Fritz-Haber-Institut der Max-Planck-Gesellschaft Berlin

17th Meeting of the Fachbeirat

Berlin, 12th - 14th February 2014

Reports

Fritz-Haber-Institut der
Max-Planck-Gesellschaft
Berlin

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Reports

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Director: R. Schlögl

Department of Chemical Physics (CP)

Director: H.-J. Freund

Department of Molecular Physics (MP)

Former Director: G. Meijer
Acting Director: M. Scheffler

Department of Physical Chemistry (PC)

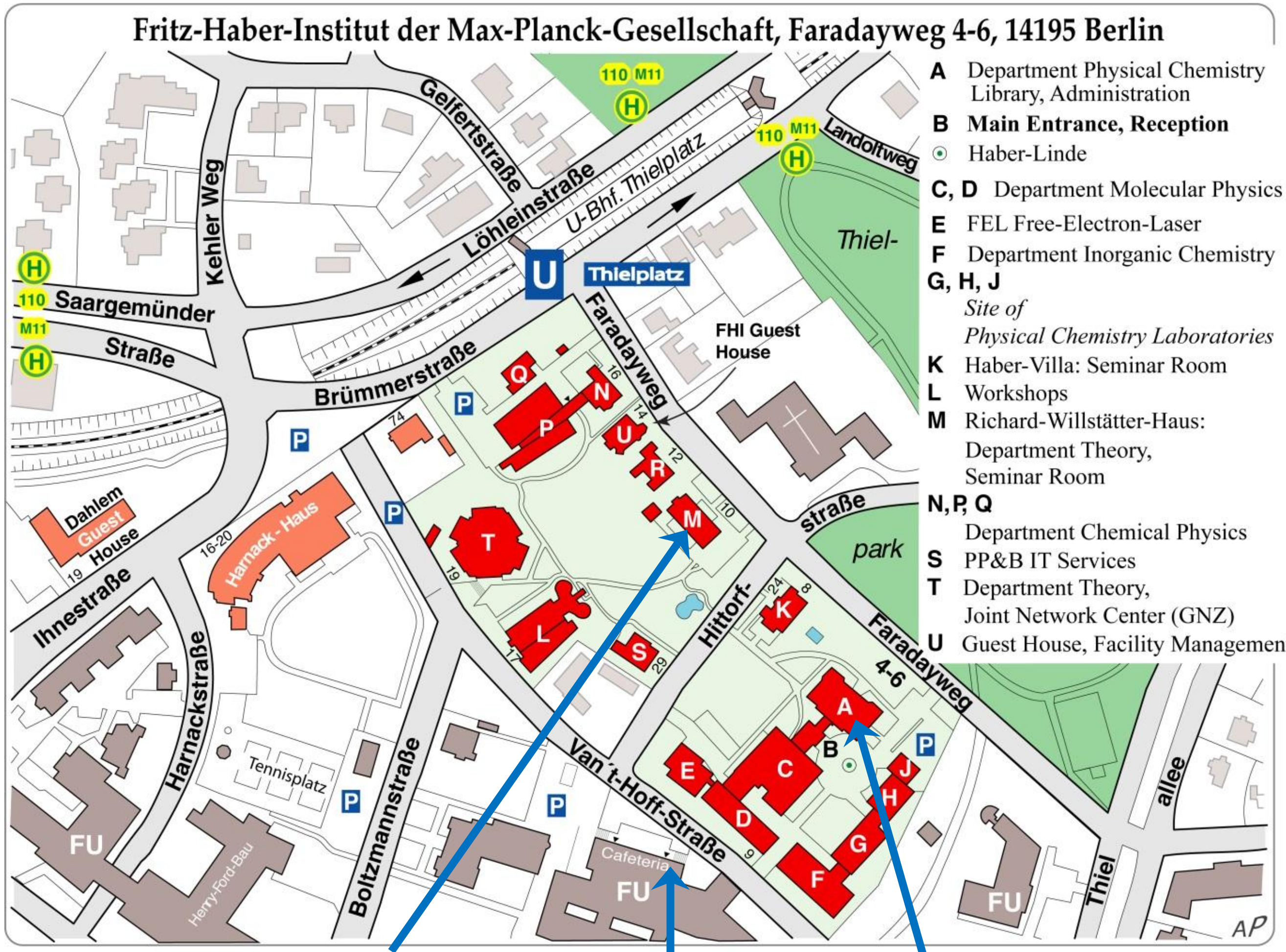
Director: M. Wolf

Head: R. Ernstorfer

Theory Department (TH)

Director: M. Scheffler

Local Map


Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin

A	Department Physical Chemistry Library, Administration
B	**Main Entrance, Reception**
⊙	Haber-Linde
C, D	Department Molecular Physics
E	FEL Free-Electron-Laser
F	Department Inorganic Chemistry
G, H, J	*Site of* *Physical Chemistry Laboratories*
K	Haber-Villa: Seminar Room
L	Workshops
M	Richard-Willstätter-Haus: Department Theory, Seminar Room
N, P, Q	Department Chemical Physics
S	PP&B IT Services
T	Department Theory, Joint Network Center (GNZ)
U	Guest House, Facility Management

All Friday: Meetings and office of the Fachbeirat at the *Seminaris Hotel and Conference Center*

Wednesday from 18:00
and all of Thursday:
Office of the Fachbeirat
(FHI, Faradayweg 10)

Thursday 9:00-13:00:
Lectures by members of the five departments
(FU, Van´t-Hoff-Str. 8)

Lunch buffet on
Thursday
(FHI library)

Research Departments of the Fritz Haber Institute

Inorganic Chemistry Robert Schlögl	**Chemical Physics** Hajo Freund	**Molecular Physics** Matthias Scheffler	**Physical Chemistry** Martin Wolf	**Theory** Matthias Scheffler
Group leaders				
Malte Behrens	Markus Heyde	Knut Asmis[2]	Kramer Campen	Carsten Baldauf
Maik Eichelbaum	Helmut Kuhlenbeck	Bretislav Friedrich	Markus Eiswirth	Luca Ghiringhelli
Axel Knop-Gericke	Thomas Schmidt	Gert von Helden	Leonhard Grill[5]	Sergey Levchenko
Julian Tornow	Shamil Shaikhutdinov	Gabriele Santambrogio[3]	Karsten Horn	Patrick Rinke
Annette Trunschke		Nicolas Vanhaecke[4]	Tobias Kampfrath	
Marc Willinger			Takashi Kumagai	
			Alexej Melnikov	
			Alexander Mikhailov	
			Julia Stähler	
Emmy Noether Group Raimund Horn[1]	ERC Group Martin Sterrer		Max Planck Research Group Ralph Ernstorfer	Angel Rubio[6]
	ERC Group Swetlana Schauermann			ERC Group Alexandre Tkatchenko

1) Since June 2013 at Technical University Hamburg-Harburg, part time at FHI
2) From February 2014 at Leipzig University, part time at FHI
3) From January 2014 at Institute for Optics of the CNR in Florence, part time at FHI
4) From February 2014 at European Patent Office in The Hague, part time at FHI
5) Since August 2013 at University of Graz, part time at FHI
6) Main affiliation: University of the Basque Country in San Sebastián

Administration, Facilities, and Service Groups

Administration
Gert von Helden/
Jörn Kändler

Free Electron
Laser
W. Schöllkopf

Joint Network
Center (GNZ)
G. Schnapka

IT Services
(PP&B)
H. Junkes

Library

Electronics
Workshop
G. Heyne

Mechanical
Workshop
P. Bischoff

Electron
Microscopy

Crystal Lab

As per December 1, 2013

Recent Developments in the Fritz Haber Institute

Executive Director: Matthias Scheffler

I. General Remarks

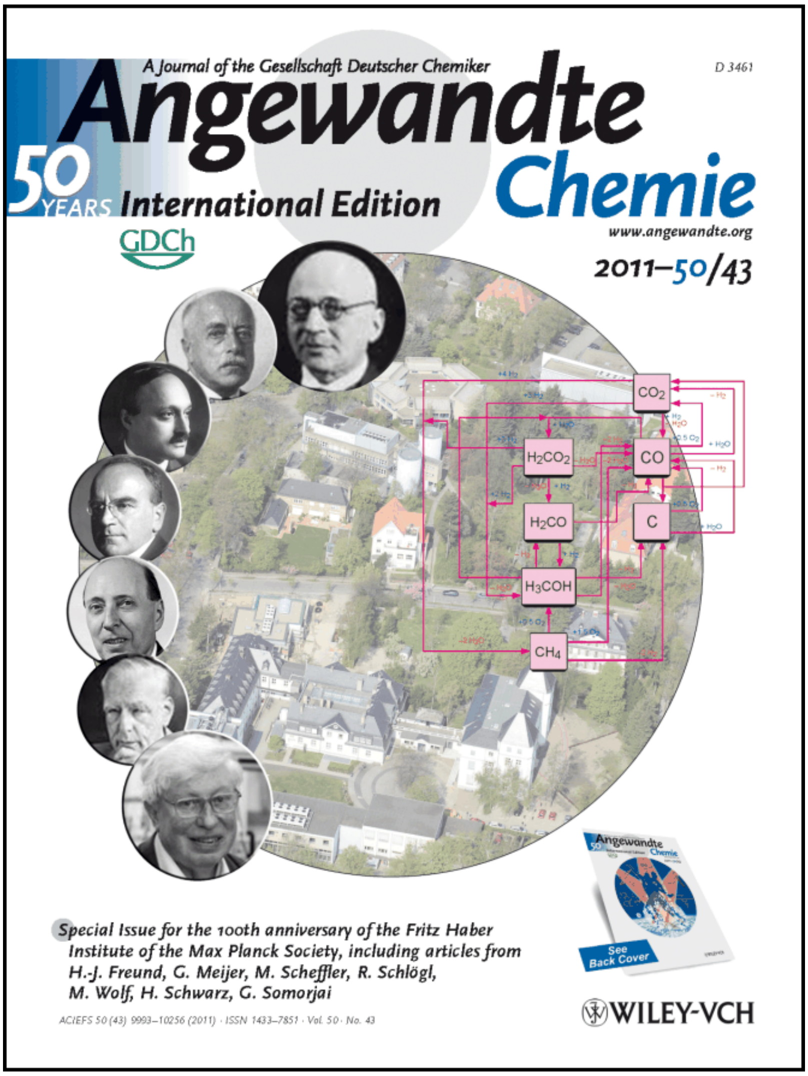
The upcoming visit of the *Fachbeirat* in February 2014 is special in several ways.

- It will be a so-called extended medium-term evaluation, and the President of the Max Planck Society has appointed two additional colleagues as *external rapporteurs*, Ben L. Feringa (Groningen) and Daan Frenkel (Cambridge). They will participate in order to compare the evaluations of the five Max Planck Institutes (MPIs) that constitute what is known in the MPS as *Research Field V*: MPI for Chemical Energy Conversion, MPI für Kohlenforschung ("Kohlen" refers to carbon), MPI of Colloids and Interfaces, MPI for Polymer Research, and the FHI. In this extended evaluation not just the last two, but the last six years, should be covered. As this would have significantly increased the necessary material for the Fachbeirat, we are putting a clear focus on the last two years (from fall 2011 to fall 2013). Only a few aspects from the earlier period will be mentioned.
- Unexpectedly, one of our director colleagues, Gerard Meijer, has left the Institute, with the consequence that severe financial cuts have been imposed. We have conducted an extensive search for a new director and started to re-structure and phase-out of most of the groups in the *Department of Molecular Physics.* More details are given below and in the report on this department, p. 185 ff.
- The scientific concept for the Free Electron Laser (FEL), a project originally initiated by Gerard Meijer, also needed to be re-considered. Since November 2013 the FEL is successfully operating in its basic mode for vibrational spectroscopy of molecules and clusters, in some ways comparable to FELIX in the Netherlands. A special section of this booklet (p. 27 ff) describes the details.

This report by the *executive director* concentrates on general and organizational aspects of the institute. The directors of the five departments present departmental issues and recent research results in their reports, beginning on p. 55.

The general research direction of the FHI concerns *fundamental aspects of the physical chemistry of surfaces, interfaces, clusters, and nanostructures.* This includes *the characterization* *of structure, composition, and dynamics, and the analysis and control of (elementary) processes*. All these attempts concern the interface between physics and chemistry. Basic research of heterogeneous catalysis is playing a noticeable role, but other topics of materials science (functional surfaces and interfaces) and biophysics are being studied as well.

Two highlights of the last six years were the *FIESTAE* symposium (Frontiers in InterfacE Science: Theory And Experiment; http://th.fhi-berlin.mpg.de/th/meetings/fiestae/), on the occasion of the 60th birthdays of Hajo Freund and Matthias Scheffler (June 28 - July 1, 2011), and the *Centennial Celebration* (100th anniversary of the founding of the Kaiser Wilhelm Institute for Physical Chemistry and Electrochemistry/Fritz Haber Institute of the Max Planck Society; October 25 – 28, 2011; http://www.fhi-berlin.mpg.de/centenary/). Both events were spectacular due to the perfect organization by Gerard Meijer as well as to the outstanding scientific programs and the discussions they engendered. The festivities for the anniversary included a scientific symposium on "Current topics in Interface and Molecular Science – Theory and Experiment", a symposium in honour of the 75th birthday of Gerhard Ertl, and a Festcolloquium including speeches by the eminent historian Fritz Stern, godson of Fritz Haber, and by Gerhard Ertl, followed by an "Open House" and a social program in the institute.


A Journal of the Gesellschaft Deutscher Chemiker

D 3461

Angewandte Chemie

50 YEARS International Edition

GDCh

www.angewandte.org

2011–50/43

Special Issue for the 100th anniversary of the Fritz Haber Institute of the Max Planck Society, including articles from H.-J. Freund, G. Meijer, M. Scheffler, R. Schlögl, M. Wolf, H. Schwarz, G. Somorjai

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See Back Cover

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Figure 1: *Cover page of the Angewandte Chemie Special Issue for the 100th anniversary of the FHI. Shown are the faces of seven Nobel laureates (F. Haber, M. von Laue, J. Franck, H. Wieland, E. Wigner, E. Ruska, and G. Ertl), an areal view at the FHI, and a reaction network typical for studies performed in the institute (example: C1 chemistry).*

The Journal *Angewandte Chemie* published in 2011 a special issue, which contains a summary of the history of the institute as well as an impressive collection of scientific papers. The five FHI directors also contributed a joint paper. Figure 1 shows the cover page of the "Angewandte".

Figure 2: *Gerhard Ertl Lecture December 10, 2013. The award winner, John T. Yates Jr. (center) together with Gerhard Ertl (left), and Matthias Scheffler (right).*

Other highlights of the last years were the *Gerhard Ertl Lectures*, which are awarded by the three Berlin Universities, the Cluster of Excellence *UniCat*, BASF, and the FHI since 2008 to esteemed colleagues from the fields of surface science and heterogeneous catalysis. The 2013 award recipient was John T. Yates Jr., shown in Fig. 2. Previous winners were: Gabor Somorjai (2008), Jens Nørskov (2009), Sir John Meurig Thomas (2010), Roald Hoffmann (2011), Charles T. Campbell (2012).

In late February 2012 Gerard Meijer informed the other directors that he had been offered the post of "voorzitter college van bestuur" (President) of the Radboud University Nijmegen, the Netherlands, and that he was seriously considering accepting. For us this came as a shock, and we tried everything possible to keep Gerard in Berlin. On February 27 we arranged a meeting with the MPS President, Peter Gruss, who also tried to persuade him to stay. For several days, Gerard went through a hard struggle to reach a final decision, but on March 7, 2012 he informed the Board of Directors that he would leave. He signed the contract with Radboud University on March 20. By September 1 he had left, and the *executive director* assumed the responsibility for the *Molecular Physics Department* and for the *FEL*. The latter is now operated as a "FHI facility".

With regard to a new director, the *Outlook Committee* (*Perspektivenkommission*) of the CPT Section of the MPS advised us that we should not restrict the search to the previous research fields of the *Molecular Physics Department*, and that we should consider primarily the long-term future of the institute. Clearly, this was also the opinion of the remaining FHI directors, and the discussions and search were performed without constraints, apart from the requirement of scientific excellence. Moreover, the research field should be in, or linked to, the wider area of the research field of the FHI as sketched above (p. 1, 2). After many meetings and further discussions, also with colleagues from the *Fachbeirat*, the Board of Directors identified a promising young candidate who is currently being evaluated by an appointment committee of the Max Planck Society. We trust that we can report more at the meeting of the *Fachbeirat* in February 2014.

In the spirit of this decision, many discussions took place with the entire *Molecular Physics Department* and with the scientists individually, in order to inform them as early as possible that the next director would probably come from a different research field. In fact, we pointed out that it would be better for many scientists to leave an albeit active and successful environment to take up an equivalent or better position elsewhere, rather than to stay in what in the MPS is called a "Restgruppe". As a result, by February 2014 the department will be essentially "empty". From the seven group leaders that existed under Gerard Meijer's directorship, only two will remain. These are:

- Bretislav Friedrich, who is very successful in the area of "interaction of (cold) molecules with fields" as well as in the history of science. He holds an honorary professor position at the TU Berlin and is also involved in research projects at other Berlin universities.
- Gert von Helden, who is a most successful scientist in the field of "infrared excitation of gas-phase molecules and clusters". He is very well linked with other research groups at the FHI and in Berlin, and is one of the members of the *Max Planck-EPFL Center for Molecular Nanoscience and Technology*. Since July 2012 Gert von Helden is also the *Administrative General Manager* (Administrativer Geschäftsführer) helping the Executive Director with many duties.

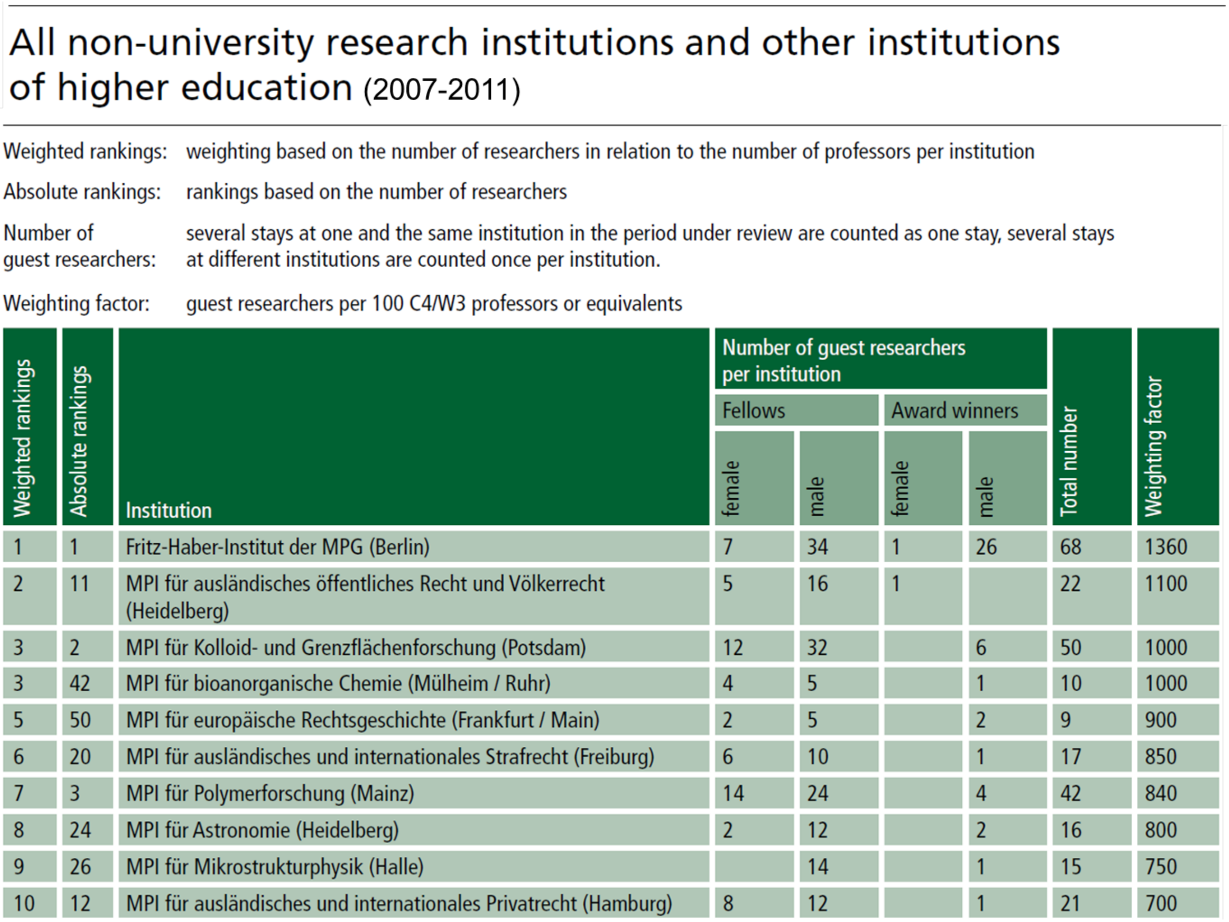
Although the department is being phased out over a rather short period of time, we have taken care that the young postdocs and students are penalized as little as possible. Thus, although most group leaders will have left by February 1, 2014, they will stay in close contact with the institute for several more months to ensure that their younger colleagues can successfully finish their projects and/or their PhD.

With respect to the other four departments we have to report that Martin Wolf (appointed as director in 2008) is still largely carrying out his experimental studies in temporary laboratory space that is rented in a nearby building in the Fabeckstraße. Finally, in early 2013, all financial and technical difficulties concerning new laboratory space at the FHI were solved, and the demolition of the building that previously had hosted part of the machine shop, the large lecture hall, the library, and the administration began in fall 2013. The new building, with its high-tech labspace for the *Department of Physical Chemistry*, should be ready in 2016.

The *Department of Inorganic Chemistry* also went through noticeable organisational and scientific changes due to the foundation of the new *Max Planck Institute for Chemical Energy Conversion (CEC)* in Mülheim. Robert Schlögl is the founding director and driving force behind the MPI-CEC, and this topic is therefore addressed in his report (p. 55 ff).

During the last six years the institute has nominated two new external scientific members. These have been duly appointed:

- Angel Rubio (University of the Basque Country, San Sebastian, Spain) who is closely linked to the *Theory Department* where he heads a research group on *Theoretical Spectroscopy*. He spends about two months per year in Berlin. Incidentally, we note that the *MPI for the Structure and Dynamics of Matter* in Hamburg has recently proposed him as a future director at their institute. He is an external scientific member of the FHI since November 2011.



All non-university research institutions and other institutions of higher education (2007-2011)

Weighted rankings: weighting based on the number of researchers in relation to the number of professors per institution

Absolute rankings: rankings based on the number of researchers

Number of guest researchers: several stays at one and the same institution in the period under review are counted as one stay, several stays at different institutions are counted once per institution.

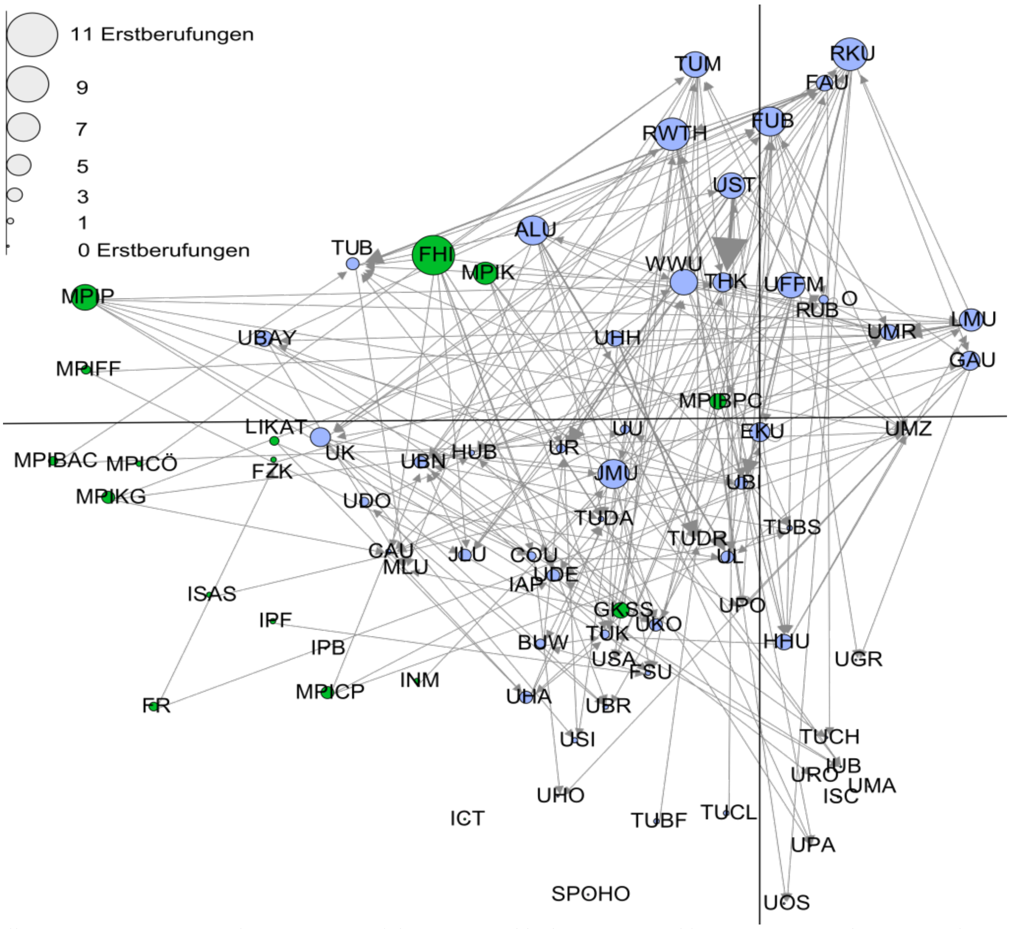
Weighting factor: guest researchers per 100 C4/W3 professors or equivalents

Weighted rankings	Absolute rankings	Institution	Number of guest researchers per institution				Total number	Weighting factor
			Fellows		Award winners			
			female	male	female	male		
1	1	Fritz-Haber-Institut der MPG (Berlin)	7	34	1	26	68	1360
2	11	MPI für ausländisches öffentliches Recht und Völkerrecht (Heidelberg)	5	16	1		22	1100
3	2	MPI für Kolloid- und Grenzflächenforschung (Potsdam)	12	32		6	50	1000
3	42	MPI für bioanorganische Chemie (Mülheim / Ruhr)	4	5		1	10	1000
5	50	MPI für europäische Rechtsgeschichte (Frankfurt / Main)	2	5		2	9	900
6	20	MPI für ausländisches und internationales Strafrecht (Freiburg)	6	10		1	17	850
7	3	MPI für Polymerforschung (Mainz)	14	24		4	42	840
8	24	MPI für Astronomie (Heidelberg)	2	12		2	16	800
9	26	MPI für Mikrostrukturphysik (Halle)		14		1	15	750
10	12	MPI für ausländisches und internationales Privatrecht (Hamburg)	8	12		1	21	700

Table 1: *The top 10 institutes in Germany chosen by AvH fellows and AvH award winners. The full list of the "Humboldt Rankings" can be found at* www.humboldt-foundation.de/web/humboldt-rankings-2012.html#s2

- Gerard Meijer (Radboud University Nijmegen, the Netherlands) who for ten years as a full scientific member made outstanding contributions to the research and scientific life of the FHI. He is an external scientific member since August 2012.

We also mention briefly that one of the best known institutions in Germany funding scientific excellence, the Alexander von Humboldt (AvH) Foundation, regularly publishes an analysis of where foreign scientists choose to go with their AvH fellowships or awards. They call it "voting with their feet". Table 1 shows the top 10 entries of the last AvH survey which covers the period from 2007 to 2011.



ALU U. Freiburg, BUW U. Wuppertal, CAU U. zu Kiel, COU U. Oldenburg, EKU Tübingen, FAU U. Erlangen-Nürnberg, FHI Fritz-Haber-Inst. der MPG, FR Forschungszentrum Rossendorf, FSU U. Jena, FUB F.U. Berlin, FZK Forschungszentrum Karlsruhe, GAU U. Göttingen, GKSS Inst. für Polymerforschung, HHU U. Düsseldorf, HUB HU. Berlin, IAP Fraunhofer-Inst. für angewandte Polymerforschung, ICT Fraunhofer-Inst. Chemische Technologie, INM Leibniz-Inst. für Neue Materialien, IPB Leibniz-Inst. für Pflanzenbiochemie, IPF Leibniz-Inst. für Polymerforschung Dresden, ISAS Inst. for Analytical Sciences, ISC Fraunhofer-Inst. für Silicatforschung, IUB International University Bremen, JLU Justus-Liebig-U. Gießen, JMU U. Würzburg, LIKAT Leibniz-Inst. für Katalyse e. V., LMU U. München, MLU U. Halle-Wittenberg, MPIBAC MPI für bioanorganische Chemie, MPIBPC MPI für biophysikalische Chemie, MPICÖ MPI für chemische Ökologie, MPICP MPI für chemische Physik fester Stoffe, MPIFF MPI für Festkörperforschung, MPIK MPI für Kohlenforschung, MPIKG MPI für Kolloid- und Grenzflächenforschung, MPIP MPI für Polymerforschung, RKU U. Heidelberg, RUBO U. Bochum, RWTH TH Aachen, SPOHO Sporthochschule Köln, THK U. Karlsruhe, TUB TU Berlin, TUBF TU Bergakademie Freiberg, TUBS TU Braunschweig, TUCH TU Chemnitz, TUCL TU Clausthal, TUDA TU Darmstadt, TUDR TU Dresden, TUK TU Kaiserslautern, TUM TU München, UBAY U. Bayreuth, UBI U. Bielefeld, UBN U. Bonn, UBR U. Bremen, UDE U. Duisburg-Essen, UDO U. Dortmund, UFFM U. Frankfurt, UGR U. Greifswald, UHA U. Hannover, UHH U. Hamburg, UHO U. Hohenheim, UK U. Köln, UKO U. Konstanz, UL U. Leipzig, UMA U. Magdeburg, UMR U. Marburg, UMZ U. Mainz, UOS U. Osnabrück, UPA U. Paderborn, UPO U. Potsdam, UR U. Regensburg, URO U. Rostock, USA U. des Saarlandes, USI U. Siegen, UST U. Stuttgart, UU U. Ulm, WWU U. Münster

Figure 3: *Number of "calls" to junior scientists in different academic institutions of the German chemistry community. The larger the circle, the more "calls" have been received at the institution concerned (see legend; "Erstberufung" denotes the first academic "call" for a researcher). The graph was published in: Baier, C., and R. Münch. "Institutioneller Wettbewerb und Karrierechancen von Nachwuchswissenschaftlern in der Chemie." KZfSS Kölner Zeitschrift für Soziologie und Sozialpsychologie* ***65****.1: 129-155 (2013).*

Recently we were informed about an analysis of the education of young academics in Germany. The data acquisition was conducted by the *German Council of Science and Humanities* (Wissenschaftsrat). Figure 3 illustrates for the institutions of the German chemistry community how many of their junior researchers received an academic “call”. Apparently, the FHI has produced the largest number of professors in the field. We believe that this tells us something about the quality of our young scientists and about the education and training provided by the FHI. While Fig. 2 covers the period of 2001-2005, the trend remains unbroken as can be seen in Table 2, which lists the 34 former members of the FHI who have been appointed to positions at a professorship-level in the last five years (2008-2013).

Person (Dep.)	**University**	**since**	**Person (Dep.)**	**University**	**since**
Hiroko Ariga	Hokkaido University, JP	2009	Andreas Osterwalde	École Polytechnique Fédérale de Lausanne, CH	2009
Knut R. Asmis	Universität Leipzig, DE	2014	Xinguo Ren	University of Science and Technology of China, CN	2013
Volker Blum	Duke University, Durham, US	2013	Zefeng Ren	Peking University, CN	2012
Ignacio Franco	University of Rochester, US	2013	Harm Hinrich Rotermund	Dalhousie University, Canada, CA	2008
Leonhard Grill	Universität Graz, AT	2013	Karsten Reuter	Technische Universität München, DE	2009
Bothina Hamad	University of Jordan, Amman, JO	2012	Thomas Risse	Freie Universität Berlin, DE	2010
Ville Havu	Aalto University School of Science, FI	2013	Melanie Schnell	Center for Free-Electron Laser Science, DE	2010
Christian Hess	Technische Universität Darmstadt, DE	2008	Aloysius Soon	College of Engineering Yonsei University, KR	2010
Steven Hoekstra	Rijksuniversiteit Groningen, NL	2009	Dario Stacchiola	Michigan Technological University, US	2009
Raimund Horn	Technische Universität Hamburg Harburg, DE	2013	Dangsheng Su	Chinese Academy of Sciences, CN	2011
Friederike Jentoft	University of Oklahoma, US	2008	Sebastiaan Y.T. v. d. Meerakker	Radboud University Nijmegen, NL	2011
Hong Jiang	Peking University, CN	2009	Zhihui Qin	WIT, Chinese Academy of Sciences, CN	2009
Jochen Küpper	Universität Hamburg, DE	2010	Simon Wall	Institute for Photonic Science, ES	2012
Suljo Linic	University of Michigan, US	2010	Kazuo Watanabe	Tokyo University of Science, JP	2010
Jonathan Lyon	Clayton State University, USA	2009	Hua Wu	Fudan University, CN	2011
Angelos Michaelides	University College London, UK	2009	Bum Suk Zhao	ULSAN National Institute of Science and Technology, KR	2011
Niklas Nilius	Carl von Ossietzky Universität Oldenburg, DE	2012	Qiang Zhang	Tsinghua University, CN	2011

Table 2 *List of 34 former members of the FHI who were appointed to academic posts (comparable to W-level positions in the German system) in the last five years (2008-2013).*

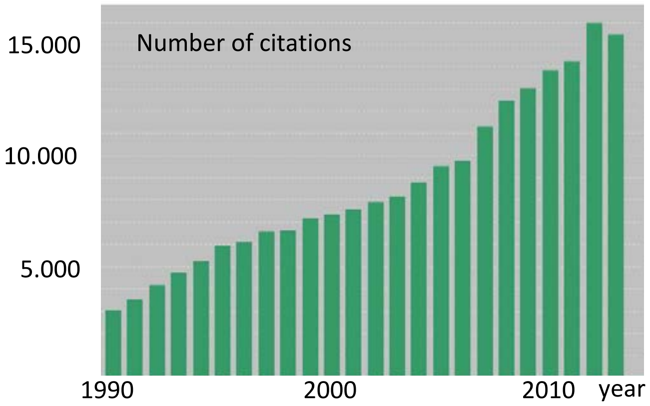


Figure 4: *Number of citations of papers with the address of the FHI according to ISI Web of Science, December 10, 2013.*

Scientific productivity can be judged using several indicators. The number of publications may be the least useful. The annual number of citations for papers with an FHI affiliation is generally considered a better indicator. It reached 16,000 in 2012 (see Fig. 4).

Another measure is the number of invited talks, since people are invited because of their novel results and the insight they offer. The total number of invited talks of members of the FHI in the last five years was 303 in 2009, 363 in 2010, 318 in 2011, 343 in 2012, and 312 during the first 10 months of 2013. Thus, in average almost every day of the year, there is one invited talk by a member of the FHI somewhere on this planet!

The awards, important grants and other distinctions they have earned should also be mentioned. Again, only the period between fall 2011 and October 2013 is taken into account:

- Malte Behrens: Jochen-Block-Preis 2013, Deutsche Gesellschaft für Katalyse
- Leonhard Grill: Feynman Prize 2011, Foresight Institute
- Raimund Horn: Hanns-Hofmann Preis 2011
- Leonid Lichtenstein: Otto Hahn Medal 2012
- Dietrich Menzel:
  - Elettra Fellow 2011, Sincrotrone Trieste
  - Gaede Langmuir Award 2012, AVS
- Laurenz Rettig: Carl Ramsauer Award 2013, Berlin Physical Society
- Mariana Rossi Carvalho: Otto Hahn Medal and Otto Hahn Award 2012
- Swetlana Schauermann:
  - Dozentenstipendium des Fonds der Chemischen Industrie, Fellowship of the Association of Chemical Industry (2012-2017)
  - ERC Starting Grant (1.6 Mio Euro) "Enantioselective Reactions on Model Chirally Modified Surfaces" (2013)
  - Minerva Research Group in the scope of "Minerva Program" of the MPS (2013)
- Georg Simon: Otto Hahn Medal 2011

Other awards and distinctions from the last two years that we could mention include,

- Hans-Joachim Freund:
  - Fellow and Member of the European Academy of Sciences (2012 and 2013)
  - Blaise Pascal Medal in Material Science of the European Academy of Sciences (2012)
  - Award for Service 2012 of the EuCheMS
  - Corresponding Member of the North Rhine-Westphalian Academy of Sciences, Humanities and the Arts (since 2012)
  - Corresponding Member of the Chemical Sciences Section and Honorary Member of the Hungarian Academy of Sciences (2013)
  - Doctor Honoris Causa of Aix-Marseille Université (2013)
- Robert Schlögl: honorary professor at the Faculty of Chemistry, University of Duisburg-Essen (2013)

The FHI continues to be involved in many activities within the Berlin-Brandenburg region, across Germany, and throughout the world, of which only a few are mentioned here.

The Cluster of Excellence (CoE) *UniCat* and four Collaborative Research Centers (CRCs, Sonderforschungsbereiche) and one Research Training Group (Graduiertenkolleg, GRK) are funded by the German Research Foundation (Deutsche Forschungsgemeinschaft), and connect the Institute with Berlin universities:

- CRC 658: Elementary Processes in Molecular Switches at Surfaces
- CRC 910: Control of Self-Organizing Non-linear Systems: Theoretical Methods and Concepts of Application
- CRC 951: Hybrid Inorganic/Organic Systems for Optoelectronics
- CRC 1109: Understanding of Metal Oxide/Water Systems at the Molecular Scale: Structural Evolution, Interfaces, Dissolution
- GRK 1558: Non-equilibrium Collective Dynamics in Condensed Matter and Biological Systems

Two more CRCs with FHI participation are currently in preparation.

In fall 2012 the CoE *UniCat* obtained a building at the TU Berlin for its new headquarters. It was named the “Gerhard Ertl Center”. All future major joint projects and the corresponding interdisciplinary training of young scientists in the fields of catalysis and chemical energy research in the Berlin area will be centred there. It will serve as a think tank, a host institution for outstanding guest scientists, a location for press conferences and strategy meetings and the home of the graduate school BIG-NSE (Berlin Graduate School of Natural Sciences and Engineering). In 2011 *UniCat* and the chemical company BASF SE founded the new joint laboratory BasCat at the Technische Universität Berlin (TU Berlin), also with noticeable input from the FHI. The current research program of BasCat focuses on oxidation catalysis (see also http://www.bascat.tu-berlin.de/bascat/).

The cooperation with the University of California at Santa Barbara (UCSB) has been very fruitful; a *MPS-UCSB program* supports joint activities and exchange of students and postdocs from the entire Max Planck Society with research in materials science at UCSB. Furthermore, the FHI is a partner in the *Max Planck-EPFL Center for Molecular Nanoscience and Technology* (http://www.fkf.mpg.de/214704/) in Lausanne/Switzerland and the *Max Planck-UBC Center for Quantum Materials* (http://www.mpg-ubc.mpg.de/) in Vancouver/Canada.

Directors and coworkers of the FHI frequently give lectures at the Berlin universities and they also organize many international workshops and schools. The latter are listed in the departmental reports of the directors.

The International Max Planck Research School (IMPRS) “Complex Surfaces in Material Sciences”, launched in 2003, creates an interdisciplinary platform for PhD education that combines cutting edge research with a thorough training of the students. It was evaluated in May 2013 and prolonged for another funding period until September 2020. The new spokesperson is Martin Wolf, and Tobias Kampfrath acts as the coordinator. A detailed description is given in a separate report (p. 15 ff).

The budget of the institute was stable until early 2012. However, the word “stable” in this context means that there had been essentially no compensation for inflation for more than 10 years. The *MPS funding* (Fig. 5, upper blue line) is at about 19 Mio € with a dip at 17 Mio € in 2005, 2006 and 2007, when we were searching for a successor of Gerhard Ertl. When Gerard Meijer announced his intention of leaving in March 2012, the budget originally allocated for his department and for the free electron laser was reduced sharply and his remaining start-up funds were withdrawn, although they had actually been earmarked for the FEL. This occurred at the same time as a budget crisis in the MPS, so that since September 2012 things have been somewhat difficult. Figure 5 shows the development of the budget over the last ten years. Note that everything is included, i.e. running costs, such as electricity, and personnel for gardening, plumbing, the workshops, etc. Only building costs are not taken into account.

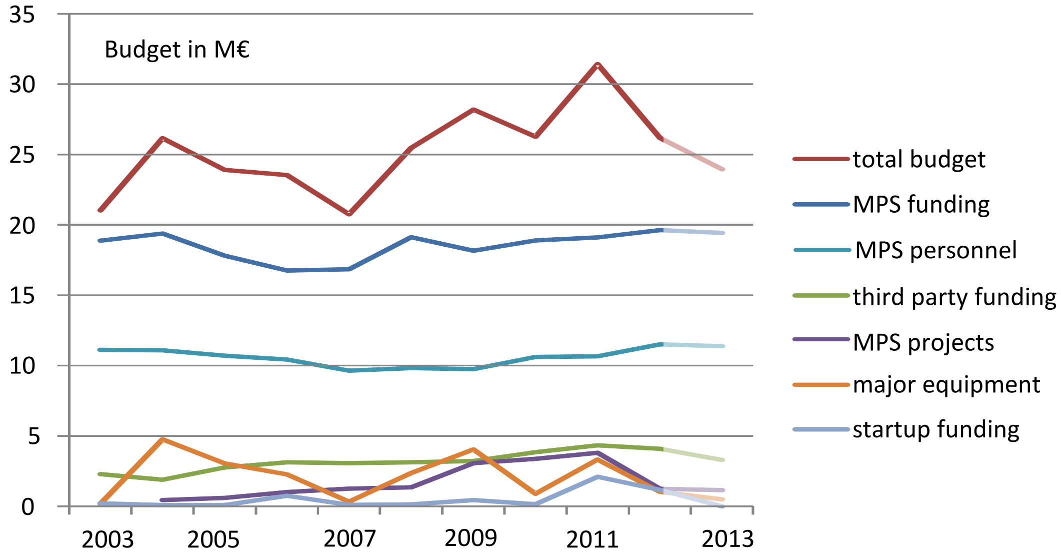


Figure 5: *Budget (in Mio €) of the institute over the period 2003-2013. For the year 2013 the target figures are used.*

As mentioned above, additional flexibility in research comes from contracts from external funding sources and from peer-reviewed, internal MPS project schemes. Among the external funding sources, the DFG, the EU including three ERC grants (Meijer, Sterrer, Tkatchenko), network-funded activities with the federal ministry of research (BMBF), and the Alexander von Humboldt Foundation are the largest contributors.

Our service groups form a decisive part of the institute's infrastructure, since they provide important support for the construction of complex equipment not available on the market. Here we would like to mention the *Electronics Laboratory*, headed by Georg Heyne, in which high-tech equipment is developed from the design stage to the delivery of the complete instrument, together with operations and safety instructions. In a similar fashion, the *Mechanical Workshop*, headed by Petrik Bischoff (appointed successor to Horst Schwäricke in April 2013), provides many fine, in many cases completely unique instruments for scientific research. Their activities are described on p. 49 and p. 50, respectively. Both service groups are highly appreciated by the scientists; they also continue to train apprentices.

The *Joint Network Center* (Gemeinsames Netzwerkzentrum, GNZ) of the Berlin-Brandenburg Max Planck institutions is a regional IT competence center and focusses on networking, back-up, virtualization and security services. It is headed by Gerd Schnapka and its activities are briefly outlined on p. 45. Based on the data network infrastructure provided by GNZ, the PP&B group (headed by Heinz Junkes) deals with the installation and maintenance of desktop systems, a compute cluster, facility management system, and telecommunication systems (more information can be found on p. 47).

It is appropriate perhaps to finish with the first lines from a recent paper published by Sir John Meurig Thomas (*"Intellectual Freedom in Academic Scientific Research under Threat",* Angew. Chem. Int. Ed. **52**, 5654 (2013))

> *"Research at the institute is primarily curiosity driven, which is reflected in the five sections comprising this Review" (on the oxidation of carbon monoxide). So wrote H.-J. Freund, G. Meijer, M. Scheffler, R. Schlögl, and M. Wolf in a Review in the special issue of Angewandte Chemie to mark the centenary of the Fritz Haber Institute (FHI) of the Max Planck Society (Angew. Chem. Int. Ed. 2011, 50, 10064). These words were music to my ears. The philosophy that animates research at the FHI also prevailed in almost all universities of the United Kingdom in former times.*

The Board of Directors will continue to act in this spirit.

We are very pleased to report that the whole institute, including the non-academic staff, create an extremely pleasant, friendly atmosphere that helps our scientists to work hard and be creative. Many thanks go to all our coworkers for their invaluable efforts. We trust that the Fachbeirat will also sense some of this spirit during its visit.

As Executive Director I would like to thank Gert von Helden, Administrative General Manager (since July 2012) for his invaluable help and support during the last 18 months. Moreover, on behalf of the Board of Directors and the whole Institute, I would also like to thank the members of the Fachbeirat, for the support and guidance they have given us over the years and to wish them an exciting and fruitful meeting.

Berlin-Dahlem, December 20, 2013

Matthias Scheffler

Matthias Scheffler

International Max Planck Research School
‘Complex Surfaces in Material Science’

Spokesperson

Hans-Joachim Freund until 06/2013 Martin Wolf from 07/2013

Coordinator

Niklas Nilius until 12/2012 Tobias Kampfrath from 01/2013

Members

Mario Dähne, TU (since 03/13)
Claudia Draxl, HU (since 03/13)
André Fielicke, TU (since 03/13)
Katharina Franke, FU (since 03/13)
Hans-Joachim Freund, FHI
Paul Fumagalli, FU (until 02/13)
Klaus Hermann, FHI (until 09/12)
Karsten Horn, FHI (until 06/13)
Norbert Koch, HU (since 03/13)
Gerard Meijer, FHI (until 09/12)
Beate Paulus, FU (since 03/13)
Klaus Rademann, HU
Thomas Risse, FU
Peter Saalfrank, UP (since 03/13)
Joachim Sauer, HU
Matthias Scheffler, FHI
Robert Schlögl, FHI
Reinhard Schomäcker, TU
Helmut Schwarz, TU (until 09/12)
Martin Weinelt, FU (since 03/13)
Helmut Winter, HU (until 09/12)
Martin Wolf, FHI

FU: Freie Universität Berlin; HU: Humboldt-Universität zu Berlin;
TU: Technische Universität Berlin; FHI: Fritz-Haber-Institut; UP: Universität Potsdam

PhD-Student Members

Alexandr Alekhin, *Russia*
Matthias Baldofski, *Germany*
Stefan Böttcher, *Germany*
Bo-Gaun Chen, *Taiwan*
Jan-Christoph Deinert, *Germany*
Petr Dementyev, *Russia (since 03/13)*
Emre Emmez, *Turkey (since 02/12)*
Felix Feiten, *Germany (since 11/12)*
Robert L. Fenger, *Germany (until 06/13)*
Vinzenz Fleischer, *Germany (since 11/12)*
Francesca Genuzio, *Italy*
Philipp Giese, *Austria (until 05/12)*
Vivekanand Gobre, *India*
Mathis Gruber, *Austria (until 02/12)*
Lukas Hammerschmidt, *Germ. (since 03/13)*
Maria Heenemann, *Germany (since 03/13)*
Arpan Kundu, *India (since 11/12)*
Xiaoke Li, *China (since 11/13)*
Leonid Lichtenstein, *Germany (until 11/12)*
Jens Lienemann, *Germany*
Bo-Hong Liu, *Taiwan*
Axel Luchterhand, *Germany (until 02/12)*
Lukas Marsoner, *Italy (since 10/13)*
Christopher Nicholson, *Scotland (since 08/12)*
Andrzej Niedziela, *Poland*
Leandro Pascua, *Spain*
GiovanniMaria Piccini, *Italy*
Nina Richter, *Germany (since 02/12)*
Norina A. Richter, *Germany (until 12/13)*
Alessandro Sala, *Italy (until 06/13)*
Nora Salas Illanes, *Chile (since 06/13)*
Philip Schambach, *Germany (until 10/13)*
Viktor Scherf, *Kazakhstan (until 05/12)*
Xin Song, *China (since 02/12)*
Christian Stiehler, *Germany (since 01/12)*
Stefanie Stuckenholz, *Germany*
Hui-Feng Wang, *China (until 10/12)*
Martin Weser, *Germany (until 04/13)*
Xin Yu, *China (until 09/13)*
Jonas Wirth, *Germany (since 03/13)*

General Remarks

The International Max Planck Research School (IMPRS) 'Complex Surfaces in Materials Sciences' (http://www.imprs-cs.mpg.de), launched in 2002, is a structured PhD program that tries to attract outstanding students from all over the world to the Fritz-Haber-Institut (FHI). It creates an interdisciplinary platform for PhD education that combines cutting-edge research with a thorough training of the students in the broad field of surface and interface science. The school is settled not only at the five departments of the FHI, but also involves various groups at the Freie Universität, the Humboldt-Universität, and the Technische Universität (all Berlin-based), and recently also at the Universität Potsdam (see Figure 1). Owing to its interdisciplinary character, the IMPRS is able to offer PhD projects in rather diverse fields, including heterogeneous catalysis, surface science, thin-film magnetism, physics of ultrafast processes, and computational chemistry. This variety in the scientific program in conjunction with close and regular interactions between the school members is the main strength of the IMPRS and an important prerequisite for its success.

The IMPRS has now nearly completed its second funding period which started with the first positive evaluation of the school in 2006 and will finish in October 2014. As detailed below, the IMPRS has successfully applied for a third funding period which will be accompanied by several gentle readjustments of the school structure.

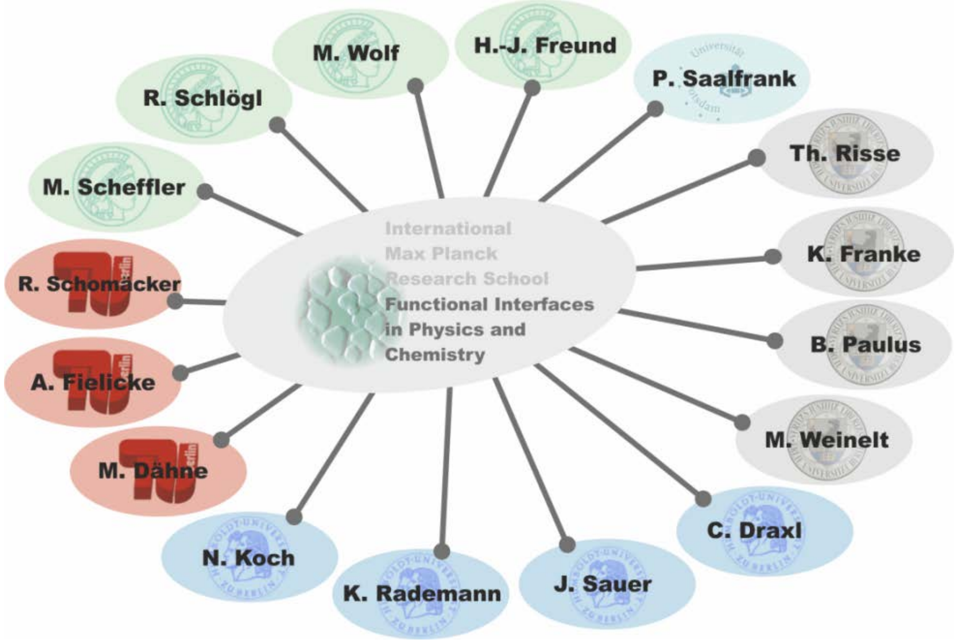


Figure 1: Scientific members of the IMPRS 'Complex Surfaces in Materials Science' as of October 2013 (green: FHI, grey: Freie Universität Berlin, blue: Humboldt-Universität zu Berlin, red: Technische Universität Berlin, light-blue: Universität Potsdam). The figure already shows the new name of the School ('Functional Interfaces in Physics and Chemistry') after renewal in a third period (2014-2020).

The initially approved budget of the IMPRS was supposed to evolve from 350 T€ in 2007 to 256 T€ in the year of 2014. However, unexpected budget cuts on the order of 20% (announced by the General Administration of the Max Planck Society in 2012) have unfortunately altered these direct allocations (see Figure 2). On the other hand, an additional 200 T€ per year have entered our budget in the form of stipends and salaries paid directly by the FHI and the partner universities. The sum of these two contributions provided the financial frame for our activities and allowed us to simultaneously award up to ten fellowships to our school members. The remaining budget was used to pay our secretary and to finance all school activities including guest speakers, seminars and workshops for the students.

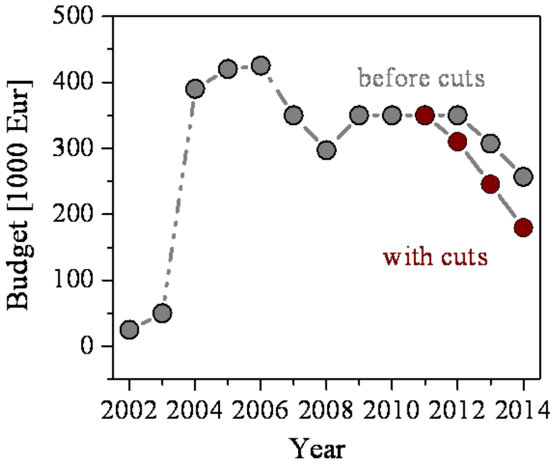


Figure 2: *Development of the IMPRS budget since its inauguration. The first funding period (2003-2008) started with an initialization time of two years (2002 and 2003). The budget of the second period started declining in 2012. The budget cut announced by the General Administration of the Max Planck Society is marked in red.*

We currently look at a total of 73 IMPRS students, counting the present (28) students as well as those (45) who graduated from 2002 to 2013. About 55% of the students came from twelve different foreign countries, for example Italy, Russia, China, Taiwan, and India. The total published output amounts to an average of 4-6 papers per student. On a statistical average, 3.7 years were required to obtain a doctoral degree, close to the target value of 3.5 years. Most of the students finished their PhD either with mark ‘very good’ (‘magna cum laude’: 76%) or ‘with distinction’ (‘summa cum laude’: 16%), and no student failed to graduate at the end of their membership in the school. Particularly positive events were the awards of distinguished prizes to five of our students, who either received the ‘Otto Hahn Medal’ of the Max Planck Society for an outstanding thesis, the ‘Klaus Tschira Award’ or the ‘Wilhelm-Ostwald-Nachwuchspreis’.

Two general trends may be identified when analyzing the career perspectives of our students after obtaining a PhD. The first group almost instantaneously found positions in the R&D or management units of big German enterprises, in particular in technologically oriented companies (Siemens, Bosch, Volkswagen, SAP), in the chemical industry (BASF and Bayer) and in the financial and administration sector (Deutsche Bank, McKinsey, Allianz Group). This first route was primarily chosen by students who were fluent in the German language. The other portion of our members continued a scientific career, typically with a postdoctoral position in a recognized research institute or a university outside Germany.

Research Achievements

The research conducted in the framework of the IMPRS mirrors the development in surface science over the last decades. The activities progressed from studies of highly idealized single crystals towards complex surfaces and interfaces, comprising nanocrystalline and amorphous materials, layered systems, materials with a well-defined dopant level or magnetic order. All those systems were analyzed with respect to their structural, electronic, magnetic and optical properties. Adsorption and reactivity studies were performed in order to provide insight into chemical processes taking place on complex surfaces. These studies aimed at gaining a better understanding of elementary steps taking place in heterogeneous catalysis and bridge the experiments performed in a laboratory environment to industrial applications. Other experimental achievements were obtained in the fields of ultrafast electron dynamics and surface magnetism, in the characterization of gas-phase molecules and clusters, and in the synthesis of complex materials with yet unrivalled catalytic properties. In addition to experimental work, there is a broad range of theoretical activities being accomplished within the school, and the close connection between theory and experiment is one particular aim of the IMPRS.

Student Training

A comprehensive education of our students, both in scientific aspects and secondary skills is one of the central goals of the IMPRS ‘Complex Surfaces in Materials Sciences’. In the second funding period, we have dedicated major efforts to achieve this goal, as detailed in the following. On the basis of a structured curriculum that is mandatory for our students, all academic school members and many external experts were engaged in this training program. The curriculum is based on three fundaments, (i) two week-long block-courses per year, (ii) monthly discussion meetings with the coordinator, and (iii) a number of soft-skill seminars organized by external experts.

The *block courses* try to impart the necessary background knowledge in the methodology, concepts, and theoretical foundations of surface science and chemistry. They are divided into two parts. The fundamental courses (held in spring) provide an introduction into the basics of physics and chemistry of surfaces. Typical topics are ‘Structures of crystalline solids’, ‘Electronic properties of surfaces', and 'Experimental methods of surface science'. To give an example of a fundamental course, we have added the schedule of the 2011 spring course as Table 1 to this report. The advanced courses (held in fall) address more specialized topics associated with surfaces and interfaces, such as 'Group Theory' and 'Ultrafast Chemistry'. Each block course contains an average of 16 lectures of 90 minutes duration and is given by senior scientists of the FHI, professors from the Berlin universities, and external specialists in the field. As our school recruits students with different backgrounds (chemistry, physics, material science), the block courses bridge language barriers between the different fields and to provide a common experimental and theoretical background for all students.

Time	Day 1	Day 2	Day 3	Day 4
9:00-12:00	*R. Schlögl* Experimental methods in catalysis	*T. Risse* Introduction into phonons and vibrations	*H.-J. Freund* Adsorption on surfaces	*J. Sauer* Electronic properties of surfaces
14.00-17:30	*K. Hermann* Introduction into surface crystallography	*S. Levchenko* Computational approaches to surface properties	*K. Horn* Physics of semiconductor interfaces	*H. Winter* Surface analysis via atom and ion probes

***Table 1:** Program of the lecture series held in March 2011.*

The *discussion meetings* (held every two months) include all students and the coordinator and provide an informal platform to review recent research developments in the school, welcome new members and organize coming activities. In addition, three students per meeting present their scientific results in a 20-minutes talk followed by 15 minutes of discussion. These regular talks in front of the school stimulate the students to improve their presentation skills, but also provide critical feedback on their scientific work. In addition, they enable the students' supervisors to gain insight into the status of a given PhD project and to rectify the goals of the project. In general, the discussion meetings are important to establish and maintain the scientific coherence of the school, as all ongoing research activities are introduced and analyzed. As the main advantage of these meetings, we want to emphasize their open, casual, and informal atmosphere that fosters an active participation (especially of young students) and provides room for scientific exchange.

Finally, the *soft-skill seminars* aim at improving the soft and secondary skills of the school members, such as their presentation and writing abilities. For this purpose, we have organized a variety of courses and workshops in order to prepare the young scientists for a possible academic or non-academic career. In the period from 2007 to 2012, the soft-skill seminars shown in Table 2 have been organized by the School.

Date	Topic	Lecturer
22.11.2011 25.11.2011	How to optimize applications in order to increase chance of employment	Wolfgang Täger
11.07., 08.08.2012	Scientific writing	Heidi Allene Henrickson
19.-22.02.2013	Introduction to LabView programming	Marcel Krenz

Table 2: *Examples of soft-skill seminars in the IMPRS.*

Special highlights of our activities are the external workshops, the last of which was carried out at the Teikyo-University Schmöckwitz in February 2012. The school members had the unique possibility to select and invite twenty outstanding researchers from all over the world. A special conference concept that was developed in particular for this occasion provided a maximum of communication and interaction between the invited speakers and the school members. Due to this open atmosphere, the meeting was an unrivalled experience for our students, giving them insight into the international scientific landscape and the chance to make contacts for their future professional career.

Third Funding Period (10/2014-09/2020)

Encouraged by the success of the IMPRS 'Complex Surfaces in Materials Science' in the past decade, we recently applied for a prolongation of the School by another funding period (10/2014-09/2020). Following our application, the School was very positively evaluated in May 2013 by a committee of international, high-ranking reviewers. In November 2013, funding over the third period was approved by the President of the Max Planck Society. The actual amount of financial support will be communicated by the end of 2013.

In the new funding period, we will continue the proven and successful core of the School, that is, its focus on surface and interface research (with a special emphasis on heterogeneous catalysis) as well as the main structure of the curriculum. However, in order to account for the latest developments in interface science, we will include new topics such as interface-specific and ultrafast spectroscopies, charge-transfer across interfaces of organic photovoltaic materials as well as the theoretical description of these structures and phenomena. To emphasize the application-related aspects of this research, the School's name will be changed from 'Complex Surfaces in Materials Science' to 'Functional Interfaces in Physics and Chemistry' in the new funding period. To integrate these topics in the IMPRS, we have gained nine high-ranking experts in these fields as new faculty members, many of which were recently appointed at the partner universities and excellently augment the IMPRS research profile. In particular, we are happy to announce that we have gained the University of Potsdam as a new partner university.

The changed member structure implies natural modifications and a broadening of the curriculum. Students will now also enjoy courses on the basics and latest developments in surface/interface science, including spin physics, ultrafast processes, and organic/inorganic interfaces which are, for example, of central importance in organic light-emitting diodes. It is very important to mention that these scientifically diverse phenomena are based on very similar fundamental principles (such as the theory of electronic structure and interface states, perturbation theory, or the concept of charge transfer). We will also strengthen the practical component of the specialized courses such that they enable more hands-on experience of various experimental and theoretical methods. Finally, we will bundle and extend our efforts to implement a quick-start help which enables new, especially foreign IMPRS students a start that is as smooth as possible.

List of lecture courses organized by the IMPRS

IMPRS-Block Course: ‘Experimental and Theoretical Methods in Surface Science’,
Teikyo University, Berlin-Schmöckwitz, February 27 - March 01, 2012.

IMPRS-Block Course: ‘Complex Surfaces in Materials Science’,
Fritz-Haber-Institut der MPG, Campus Tour, October 04 - 10, 2012.

Lecture Series: ‘Modern Methods in Heterogeneous Catalysis Research’,
Fritz-Haber-Institut der MPG, October 2012 - February 2013.

IMPRS-Block Course: ‘Aspects of Physics and Chemistry of Surfaces’,
Fritz-Haber-Institut der MPG, March 21 - 26, 2013.

IMPRS-Block Course: ‘Dynamic Processes at Interfaces and Surfaces’,
Fritz-Haber-Institut der MPG, September 27 - October 02, 2013.

Lecture Series: ‘Modern Methods in Heterogeneous Catalysis Research’,
Fritz-Haber-Institut der MPG, October 2013 - February 2014.

PhD-Projects within the IMPRS 2012 - 2013

Alekhin, Alexandr, Russia, Supervisors: Prof. Wolf/Dr. Melnikov (FHI),
“Hot carrier-driven magnetization dynamics”.

Baldofski, Matthias, Germany, Supervisors: Prof. Sauer/Prof. Freund (HU/FHI),
“Micro-kinetic modeling of the oxidative coupling of methane”.

Böttcher, Stefan, Germany, Supervisors: Prof. Risse/Prof. Horn (FU/FHI),
“Functionalization of Graphene”.

Chen, Bo-Gaun, Taiwan, Supervisors: Prof. Rademann/Dr. Christen (HU),
“Interactions of clusters with modified silicon surfaces”.

Deinert, Jan-Christoph, Germany, Supervisors: Prof. Wolf/Dr. Stähler (FHI),
“The electronic structure and ultrafast dynamics of the heterogeneous electron transfer at molecule-zinc oxide interfaces”.

Dementyev, Petr, Russia, Supervisors: Prof. Freund/Dr. Schauermann (FHI),
“Microcalorimetric measurements on oxide supported model catalysis”.

Emmez, Emre, Turkey, Supervisors: Prof. Rademann/Prof. Freund (HU/FHI),
“The Reactivity Studies of Aluminosilicate (Zeolite) Thin Films grown on Ru (0001) Substrate”.

Feiten, Felix E., Germany, Supervisors: Prof. Freund/Prof. Risse (FHI/FU),
“Surface termination of V_2O_3(0001) on Au (111)”.

Fenger, Robert, Germany, Supervisors: Prof. Rademann/Prof. Freund (HU/FHI),
“Catalytic investigations of supported noble metal nanoparticles with spectroscopic methods”.

Fleischer, Vinzenz, Germany, Supervisors: Prof. Schomäcker/Prof. Schlögl (TU/FHI),
“Resolution of the surface and gas phase contributions to the mechanism of the oxidative coupling of methane (OCM) at Na_2WO_4-Mn-SiO_2-catalyst”.

Genuzio, Francesca, Italy, Supervisors: Prof. Freund/Prof. Winter (FHI/HU),
“Spectroscopic characterization of metal nano-particles on thin hematite films”.

Giese, Philipp, Austria, Supervisors: Prof. Wolf/Prof. Freund (FHI),
“Oxidative coupling of methane”.

Gobre, Vivekanand, India, Supervisors: Prof. Scheffler/Dr. Tkatchenko (FHI),
“Van der Waals interactions for adsorption on ionic solids”.

Gruber, Mathis, Austria, Supervisors: Prof. Hermann/Prof. Freund (FHI),
“Interaction and reaction of hydrocarbons on oxides: DFT model investigations of electronic structure and spectroscopic properties”.

Hammerschmidt, Lukas, Germany, Supervisor: Prof. Paulus (FHI),
“Theoretical investigation of nano-structured thermoelectric materials”.

Heenemann, Maria, Germany, Supervisors: Prof. Schlögl/Dr. Eichelbaum (FHI),
“The influence of charge transport in high performance oxidation catalysts on activity and selectivity: Contactless measurements of Hall mobilities and charge carrier concentrations under reaction conditions”.

Kundu, Arpan, India, Supervisors: Prof. Sauer/Dr. Christen (HU),
“Accurate energy and free energy calculation of small molecule and surface”.

Li, Xiaoke, China, Supervisors: Prof. Sauer/Dr. Paier (HU),
“Density functional calculations of interactions between molecules and oxide surfaces”.

Lichtenstein, Leonid, Germany, Supervisors: Prof. Freund/Prof. Fumagalli (FHI/FU),
“Noncontact dynamic force microscopy on oxide thin layers and oxide bulk systems”.

Lienemann, Jens, Germany, Supervisors: Prof. Winter/Dr. Busch (HU),
“Untersuchung der Eigenschaften von Festkörperoberflächen mittels streifender Ionenstreuung”.

Liu, Bo-Hong, Taiwan, Supervisors: Prof. Freund/Prof. Risse (FHI/FU),
“Structure and reactivity of zinc oxide based model catalysis”.

Marsoner Steinkasserer, Lukas E., Italy, Supervisor: Prof. Paulus (FU)
"Theoretical first-principles investigation of graphene nano-structures on metallic surfaces".

Nicholson, Christopher, Scotland, Supervisors: Prof. Wolf/Prof. Weinelt (FHI/FU),
"Ultrafast Dynamics of Correlated Materials".

Niedziela, Andrzej, Poland, Supervisors: Prof. Sauer/Dr. Sierka (HU),
"Efficient optimization methods for large scale systems as used in heterogenous catalysis".

Pascua Arcusin, Leandro, Argentina, Supervisors: Prof. Freund/Prof. Pascual (FHI/FU),
"Construction of an optical near-field microscope to investigate optical centers in oxides".

Piccini, GiovanniMaria, Italy, Supervisor: Prof. Sauer (HU),
"Entropy barriers for surface reactions in Zeolites".

Richter, Nina, Germany, Supervisors: Prof. Risse/Prof. Freund (FU/FHI),
"Defect chemistry of thin metal oxide films".

Richter, Norina Anna, Germany, Supervisor: Prof. Scheffler (FHI),
"Reaction dynamics at lithium doped MgO (001)".

Sala, Alessandro, Italy, Supervisors: Prof. Freund/Prof. Christmann (FHI/FU),
"Investigation of catalytically active nano-cluster with high-resolution spectro-microscopy".

Sala Illanes, Nora, Chile, Supervisors: Prof. Draxl (HU),
"Electronic Structure of organic semiconductors by means of self-consistent *GW* approaches".

Schambach, Philip, Germany, Supervisors: Prof. Wolf/Prof. Freund (FHI),
"Tip enhanced Raman spectroscopy".

Scherf, Viktor, Kazakhstan, Supervisors: Prof. Christmann/Prof. Freund (FU/FHI),
"Adsorption of Ag on the Re(10-10) surface".

Song, Xin, China, Supervisors: Prof. Weinelt/Prof. Freund (FU/FHI),
"Surface phases of (Ti, V) mixed oxide on TiO2 (110)".

Stiehler, Christian, Germany, Supervisors: Prof. Freund/Prof. Weinelt (FHI/FU),
"Investigation of the influence of adsorption processes on the electronic properties of ultra-small, oxide-supported metal particles with discrete electronic structure via low temperature scanning tunneling microscopy".

Stuckenholz, Stefanie, Germany, Supervisors: Prof. Freund/Prof. Risse (FHI/FU),
"From a thin film to a bulk oxide system – MgO on Mo(100)".

Wang, Hui-Feng, China, Supervisors: Prof. Freund/Prof. Christmann (FHI/FU),
"STM investigations of solid/liquid interfaces".

Weser, Martin, Germany, Supervisors: Prof. Horn/Prof. Bovensiepen (FHI/FU),
"Electronic and magnetic properties of graphene based systems".

Yu, Xin, China, Supervisors: Prof. Freund/Prof. Schomäcker (FHI/TU),
"Structure-reactivity relationships of metal and metal oxide aggregates on ordered silica films".

FHI Facility Free Electron Laser

Head: Wieland Schöllkopf

Introduction

In 2008 the Max Planck Society decided to go ahead with the FHI free-electron laser (FEL) project. The aim was to set up an FEL that is capable of providing pulsed laser radiation at any wavelength between several micron in the infrared and several hundred micron in the THz regime. The purpose of the FEL is to serve the experimental groups of the FHI as a source of intense and continuously tunable laser pulses for IR and THz spectroscopy studies. Located in-house, the FEL avoids the need to move complex and expensive experimental set-ups to one of the existing IR FEL user facilities.

Following the 2008 decision, the FEL team, which was assembled by Gerard Meijer, has been working on the project. It consists of Wieland Schöllkopf (scientist in charge), Sandy Gewinner (laser engineer), and Gert von Helden (scientist) who were later joined by Wolfgang Erlebach (designer/engineer) and Andreas Liedke (electrician), both from the MP-Department, as well as by Heinz Junkes (computer expert) from the FHI group "PP&B". In addition, Weiqing Zhang, who is now heading the deep-UV FEL project in Dalian, China, was joining the FHI FEL team from 1/2010 to 9/2012 as a postdoctoral fellow supported by the Alexander von Humboldt Foundation. The FEL team dealt with the following tasks, which were successfully completed:

1. Planning of the new FEL building (construction from 3/2010 to 5/2011) including the technical facility infrastructure needed for an FEL
2. Radiation safety including applications for the legally required permits from the Berlin state authorities to build the radiation-safe facility and to run the electron accelerator ("*Errichtungsgenehmigung*" and "*Probe-Betriebsgenehmigung*") as well as the mandatory training as radiation safety officers ("*Strahlenschutzbeauftragte*")
3. Design, procurement, and installation of the electron accelerator (including electron beamline), the mid-infrared undulator, and the FEL cavity
4. Design and in-house fabrication of the IR beamlines transferring the FEL radiation to the experimental stations; design and implementation of the IR diagnostics

5. Design and implementation of the computer control system and its integration with the FHI experimental and facility systems
6. Commissioning of the mid-IR FEL system, system operation and maintenance as well as user operation

Parts of the tasks (2) – (5) were dealt with in fruitful collaborations with colleagues from FELBE (Helmholtz-Zentrum Dresden Rossendorf) and from BESSY II (Helmholtz-Zentrum Berlin). Both groups provided expertise regarding topics such as electron-accelerator design, undulators, radiation safety, etc., which is not readily available at the FHI. In addition, the FEL project has gotten excellent support from the FHI service groups including the electronics shop, machine shop, computer groups (GNZ, PP&B), facility management, and administration.

Design of the FHI FEL; Electron Accelerator

Early on it became clear that two different undulator lines will be needed to cover the full wavelength range of interest from about 4 to 500 μm. Thus, the system outline, shown in Fig. 1, includes two FELs; a mid-infrared (MIR) branch for wavelengths up to about 50 μm and a far-infrared (FIR) branch for wavelengths from about 40 to 500 μm. A normal-conducting linear accelerator provides electrons of up to 50 MeV energy with a beam transport system that feeds either of the FEL branches or the electron diagnostics beamline.

Normal-conducting RF accelerators (in contrast to superconducting machines) are powered by intense RF (~GHz) pulses, several microseconds long, with a relatively low (Hz – kHz) repetition rate. As a result, bunch trains of electrons are accelerated. The repetition rate of the bunches (*micro bunches*) within a bunch train (*macro bunch*) is determined by the RF frequency (1 GHz in our system), while the length and repetition rate of the macro bunches is determined by the RF pulse length and repetition rate, respectively (~10 μs and typically 10 Hz in our system). As the time structure of the FEL radiation essentially reflects the one of the electrons, the FHI FEL generates IR radiation in macro-pulses, several microseconds long and each consisting of thousands of micro-pulses. This peculiar time structure has shown to be very advantageous for experiments where lots of photons are needed on a time scale of tens of microseconds, e.g. for multi-photon spectroscopy in the gas-phase. For other studies, such as e.g. pump-probe experiments in the condensed phase, a pulse structure with a reduced micro-pulse repetition rate is more advantageous. As described below, this additional mode of operation will also be available at the FHI FEL.

The accelerator and electron beamline system of the FHI FEL was designed, built, and installed by the US company Advanced Energy Systems, Inc. together with Dave Dowell (formerly SLAC/LCLS), Lloyd Young (formerly LANL), Kevin Jordan (JLAB), and Ralph Lange (BESSY II) working as consultants to this company. The accelerator system consists of a thermionic gridded electron gun, a sub-harmonic buncher cavity and two standing-wave copper linacs. The first of the two S-band (3 GHz) normal-conducting electron linacs accelerates the electron bunches to a nominal energy of 20 MeV, while the second one accelerates or decelerates the electrons to deliver any final energy between 15 and 50 MeV. Each linac is powered by a separate high-power klystron providing microwave pulses of up to 20 MW peak power and 18 μs pulse length. A chicane between the linacs allows for adjustment of the electron bunch length as required. The device was designed to meet challenging specifications, including low longitudinal (< 50 keV-psec) and transverse emittance (< 20 π mm-mrad) at more than 200 pC bunch charge with a micro-bunch repetition rate of 1 GHz and a maximum macro-bunch length of at least 8 μs. The maximum macro-

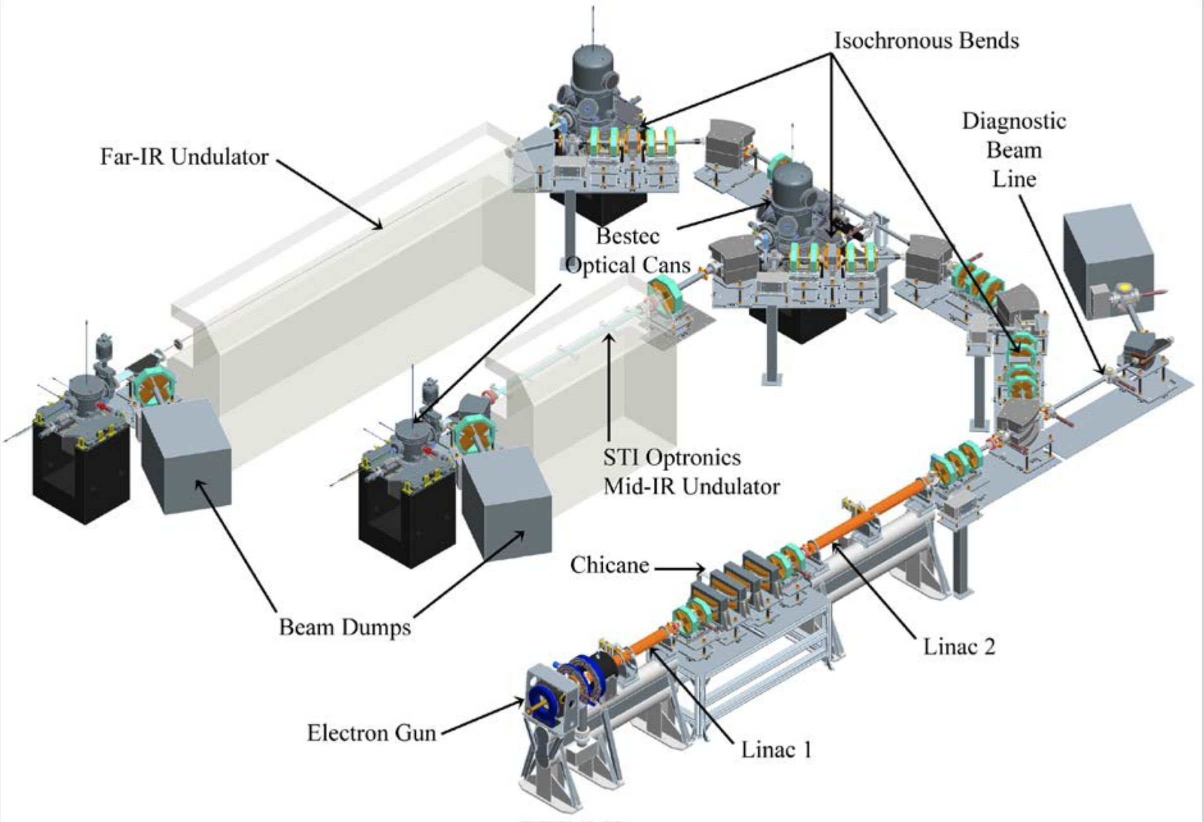


Figure 1: *Overview of the FHI FEL installation showing the electron accelerator system and the MIR FEL (operational) and the FIR FEL (to be installed in the future).*

bunch repetition rate is 20 Hz. Given these performance specifications the accelerator system is a state-of-the-art normal-conducting RF accelerator. In Table 1 we summarize the top-level electron beam performance.

The final design has optimized the specifications of the linac that are most relevant for the IR and THz FEL performance. For instance, the target bunch charge of the micro-pulses has been increased to 300 pC. In addition, the target length of the macro-bunches has been increased to 15 µs, a feature that will be especially beneficial for multi-photon experiments carried out at the FHI. Exploring if or how close the accelerator system can reach the target parameters will be among the tasks for 2014.

As of fall 2013, commissioning of the accelerator and electron-beam transport system is nearing completion. The manufacturer has demonstrated almost all of the specifications listed in Table 1. What remains to be done to finalize the accelerator system is, essentially, the implementation and commissioning of the reduced repetition-rate mode (not listed in Tab. 1).

Parameter	Unit	Specification	Target
Electron energy	MeV	20 – 50	15 - 50
Energy spread	keV	50	< 50
Energy drift per hour	%	0.1	< 0.1
Bunch charge	pC	200	> 200
Micro-bunch length	ps	1 - 5	1 - 10
Micro-bunch rep. rate	GHz	1	1
Micro-bunch jitter	ps	0.5	0.1
Macro-bunch length	µs	1 - 8	1 - 15
Macro-bunch rep. rate	Hz	10	20
Normalized *rms* transverse emittance	π mm mrad	20	20

Table 1: *Summary of electron beam parameters of the S-band linear accelerator.*

The 1 GHz mode corresponds to an electron bunch separation of 1 ns in time and, hence, 30 cm in space. As the MIR FEL cavity is 5.4 m long, there are 36 equally spaced IR pulses circulating the cavity in the 1 GHz mode. Thus, having just 1 or 2 or 3, … IR pulses circulating the cavity can be achieved by reducing the electron-bunch repetition rate to 1 or 2 or 3, … times (1/36) GHz, which is equal to 27.6, 55.6, 83.3 … MHz. Implementation of the reduced rep. rate mode is scheduled for spring 2014.

Mid-Infrared FEL; Undulator and IR cavity

Both, the MIR FEL and the FIR FEL, consist of an undulator placed within an IR cavity as summarized in Table 2. The MIR FEL includes a 2-m-long planar wedged-pole hybrid undulator manufactured by STI Optronics, Inc. with a period length of 40 mm. At a minimum gap of nominally 16.5 mm, a maximum root-mean-square undulator parameter K_{rms} of more than 1.6 is reached. This, in combination with the minimum electron energy of 15 MeV corresponds to a theoretical maximum wavelength of more than 50 µm for the MIR system. The MIR undulator, as installed in the vault of the FHI FEL building, can be seen in the center of Fig. 2.

Undulator	MIR	FIR
Type	Planar hybrid	Planar hybrid or PPM
Material	NdFeB	NdFeB or SmCo
Period (mm)	40	110
No. of periods	50	40
Length (m)	2.0	4.4
K_{rms}	0.5 – 1.6	1.0 – 3.0
IR-cavity	**MIR**	**FIR**
Length (m)	5.4	7.2
Waveguide	none	1-D 10 mm high

Table 2: *Summary of essential parameters of the MIR (commissioned) and FIR (prospected) FEL setups.*

Commissioning of the MIR undulator and oscillator cavity (built by Bestec GmbH) was completed in 2011 already. First lasing of the MIR FEL was achieved at a wavelength of 16 µm in 2012. First user experiments with mid IR FEL radiation have been made in 2013, as will be described below.

The FIR FEL has not yet been installed, but its design has been outlined. The right column of Table 2 gives the essential design parameters for the FIR undulator and cavity. In addition, the electron branch-off beamline has been included in the current system, thereby allowing for implementation of the FIR system at a later point of time.

Facility infrastructure and user beamline commissioning

The IR pulses, extracted from the MIR FEL, enter the IR beamline through a CVD diamond window under Brewster angle. The window separates the ultra-high vacuum of less than 1×10^{-8} mbar in the undulator and cavity-mirror chambers from the high vacuum (10^{-5} mbar) in the IR beamline. The IR beamline consists of 10 cm inner-diameter stainless steel pipes interconnected at right angle by vacuum chambers housing 90°-deflection broadband IR mirrors. The mirrors are either flat or toroidal (focusing) and are made out of copper with a gold coating. The nominal mirror reflectivity is 99.2% over the entire IR range. A total of 6

Figure 2: *MIR FEL with undulator (center) and IR-cavity mirror chambers (left and right) as of 2013.*

such mirrors steer the IR beam from the FEL cavity in the vault to the IR diagnostic station located in the neighboring building (Building D) over a total length of 18 m (see Fig. 3).

The diagnostic station comprises different commercial IR detectors including a liquid-nitrogen cooled MCT (HgCdTe) detector and a large area (5 cm diameter) pyro detector for IR power and pulse energy measurements. Additional equipment in the diagnostic station includes a Czerny-Turner grating spectrometer as well as a 5 stage IR beam attenuator. The diagnostic signals are online available to the users. It is planned to set up additional equipment for characterizing and manipulating the IR beam in the future including a photo-acoustic cell and a polarization rotator.

Another IR beamline system (user beamlines) connecting the diagnostic station with the first six experimental stations has been installed and commissioned. It transfers the IR beam to either one of the user experiments located in the basement and the ground floor of Building D as indicated in Fig. 3.

The EPICS-based computer control system of the FEL has been integrated with the facility control system and with the users' experimental control systems, thereby allowing the users to control the FEL remotely and run wavelength scans. As of fall 2013 five experiments from the area of gas-phase spectroscopy of bio-molecules and clusters have started experiments using the MIR FEL radiation. A sixth experiment from the field of solid-state physics (spin-phonon coupling in a ferrimagnet, as described below) is being set up by a group from the Physical Chemistry Department;

1. André Fielicke (TU Berlin, Lab D0.13, 1st floor Building D): *Chemistry of transition metal clusters*

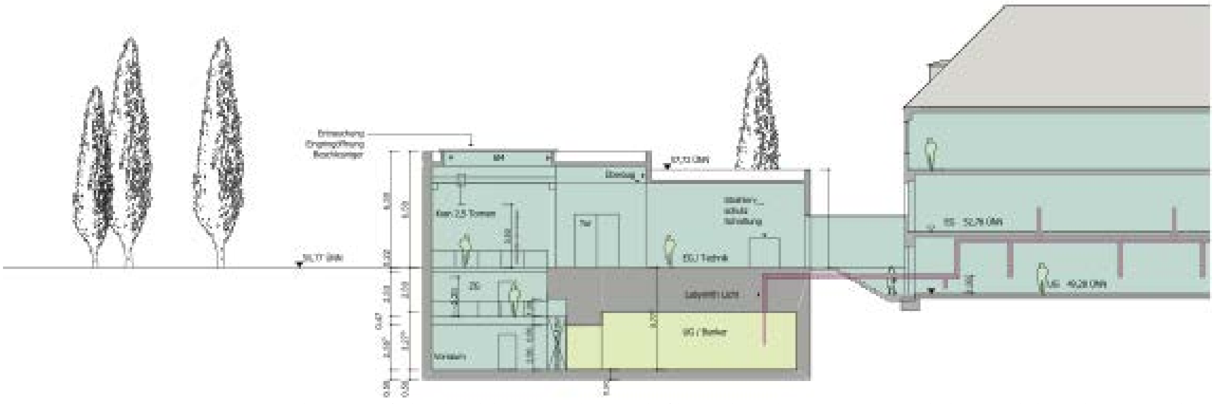


Figure 3: *Cross sectional view of the buildings E (FEL, left) and D (spectroscopy labs, right) indicating the IR transport beamline system connecting the FEL vault with the first six user stations located in the basement and ground floor of Building D.*

2. Knut Asmis I (Dept. MP, Lab DK.05, basement Building D): *Vibrational spectroscopy of gas phase clusters: catalysis, astrochemistry and new materials*
3. Knut Asmis II (Dept. MP, Lab DK.05, basement Building D): *Vibrational spectroscopy of gas phase clusters: atmospheric chemistry and ion solvation*
4. Gert von Helden I (Dept. MP, Lab DK.05, basement Building D): *Bio-molecules embedded in superfluid helium nano droplets*
5. Gert von Helden II (Dept. MP, Lab D0.11, 1st floor Building D): *IR spectroscopy combined with ion mobility spectrometry*
6. Alexander Paarmann (Dept. PC, Lab DK.08, basement Building D): *Spin-phonon coupling in a ferrimagnet*

Based on the modular beamline design it is straightforward to add more user-beamline branches in the future.

First spectroscopy experiments

Figure 4 shows a spectrum of the laser line centered at a wavelength of 5 μm obtained for an electron energy of 38 MeV. The line width (full width at half maximum) in this measurement is about 33 nm corresponding to a relative line width of less than 0.7%, which is as expected. The line width can be varied to some extent by adjusting the FEL cavity length. The stability of the 5.4-m-long IR cavity has turned out to be excellent. Even when the feedback system for cavity-length stabilization (which uses the signal from a HeNe laser interferometer) is not running, we do not observe significant length drifts. We attribute this to the careful design of the mirror chambers and to the good temperature stability in the FEL vault.

As of fall 2013 lasing of the MIR FEL was demonstrated in the wavelength range from 4 to 24 μm. Characterization of the spectral, temporal, and spatial properties of the FEL radiation in more detail as well as lasing at longer wavelength is work in progress. So far, a maximum IR pulse energy of 130 mJ was observed for a wavelength of 9 μm. It is expected that further tuning of the many parameters of the accelerator system will allow for an even better performance of the MIR FEL.

The FHI FEL started user operation in fall 2013. The first experiments using the FEL radiation were made in collaboration with the group of André Fielicke (FHI Dept. MP and TU

Berlin) investigating transition-metal clusters in the gas phase. Figure 5 shows vibrational spectra of carbon monoxide attached to small cationic ruthenium and cobalt-manganese clusters, measured with the FHI FEL in the wavelength range from 1800 to 2200 cm^{-1}. For cobalt-manganese clusters, for instance, one finds a red shift of the CO vibrational absorption

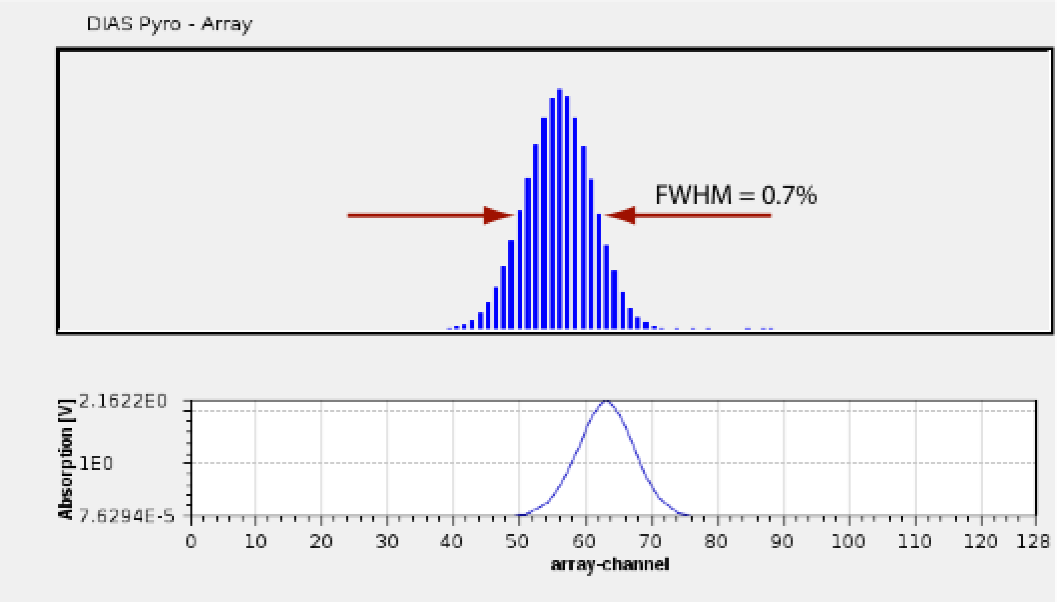


Figure 4: *Spectrum of the FEL laser line centered at 5 μm measured with a grating spectrometer. The x-axis represents the wavelength with its full scale corresponding to 400 nm.*

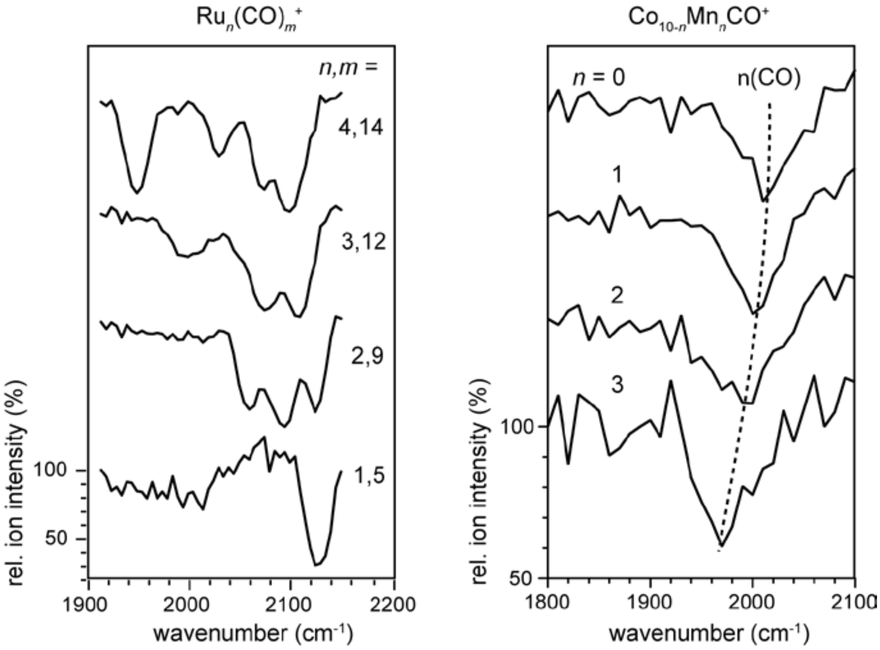


Figure 5: *In the first FHI FEL user experiments the IR radiation was used to measure vibrational spectra of cationic transition-metal carbonyl clusters. The left picture shows spectra of carbon monoxide attached to small ruthenium clusters showing intense absorption peaks. The right hand side shows spectra of a single CO attached to decamer cobalt-manganese clusters of various Mn to Co number ratios. These results were obtained in collaboration with the Fielicke group (FHI Dept. MP and TU Berlin).*

line with increasing manganese fraction. These studies allow to probe the effects of alloying on the *d*-band center that is controlling the strength of the *p*-backbonding to the CO. For more details on transition-metal cluster spectroscopy see the report from the Molecular Physics Department in this booklet.

Taking advantage of the widely tunable radiation from the FHI FEL, another experiment with gas-phase clusters was made in collaboration with the group of Knut Asmis (FHI Dept. MP). In these experiments small negatively charged aluminum oxide clusters were prepared in an ion trap, cooled to temperatures on the order of 20 K, and tagged with a few deuterium molecules per cluster. Mass-selective vibrational spectra, as those shown in Fig. 6, were measured by way of infrared photodissociation of the messenger-tagged complexes in order to probe (close to) the linear absorption regime. More details on vibrational spectroscopy of metal-oxide clusters in the gas phase are given in the report from the Molecular Physics Dept. within this booklet.

In an experiment performed in collaboration with the group of Gert von Helden (FHI Dept. MP), the FEL has been used to study the structure of biomolecules in the gas phase. In a very

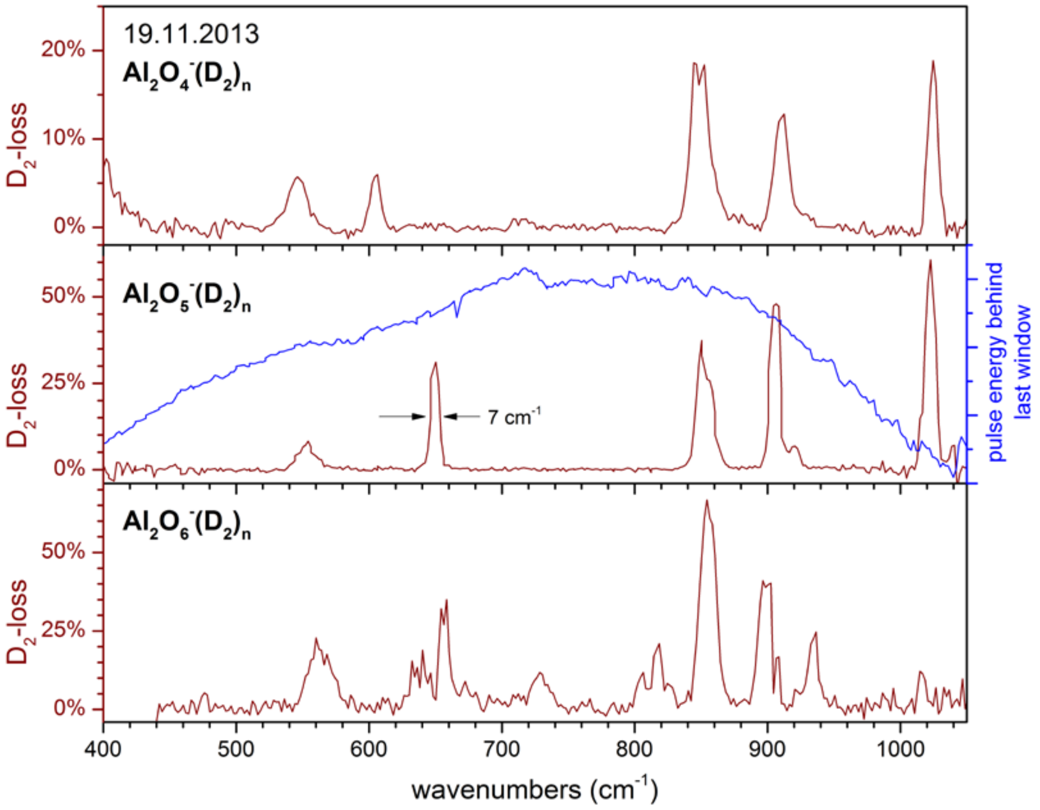


Figure 6: *Gas-phase vibrational spectra (400 to 1050 cm*$^{-1}$*) of mass-selected, cryogenically-cooled aluminum-oxide cluster anions* $Al_2O_n^-$ *with n=4-6 (Asmis group, FHI Dept. MP).*

first attempt, the protein Ubuiquitin (76 amino acids) in the charge state 7+ and complexed with a crown-ether molecule is investigated (see Fig. 7). A key feature of the experiment is that prior to the interaction with the FEL, the molecules are not only mass/charge selected but also conformer selected using gas-phase ion mobility methods. Because of this, it is very likely that the structure investigated resembles closely the so called “A-State” of ubiquitin (see picture in Fig. 7). The spectrum is characterized by an intense amide-I band near 1650 cm^{-1} and an intense amide-II band near 1500 cm^{-1}. Both bands appear surprisingly narrow.

FEL experiments currently being set up

In addition to the gas-phase studies, the FHI FEL will also be used in solid-state and surface-science experiments that are being prepared by groups from the experimental departments of the FHI. One such example is vibrational spectroscopy of metal clusters deposited on flat ordered substrates, so-called model catalysts (Fig. 8). Such deposited metal aggregates cannot be studied directly with IR absorption spectroscopy and therefore action spectroscopy is employed. The clusters are tagged with weakly bound rare-gas atoms. When the FEL radiation is in resonance with a vibrational excitation of the cluster, the rare gas atoms will desorb due to the rapid heating of the clusters. Thus, rare-gas pressure maxima as measured with a quadrupole mass spectrometer will be used to observe vibrational spectra of the

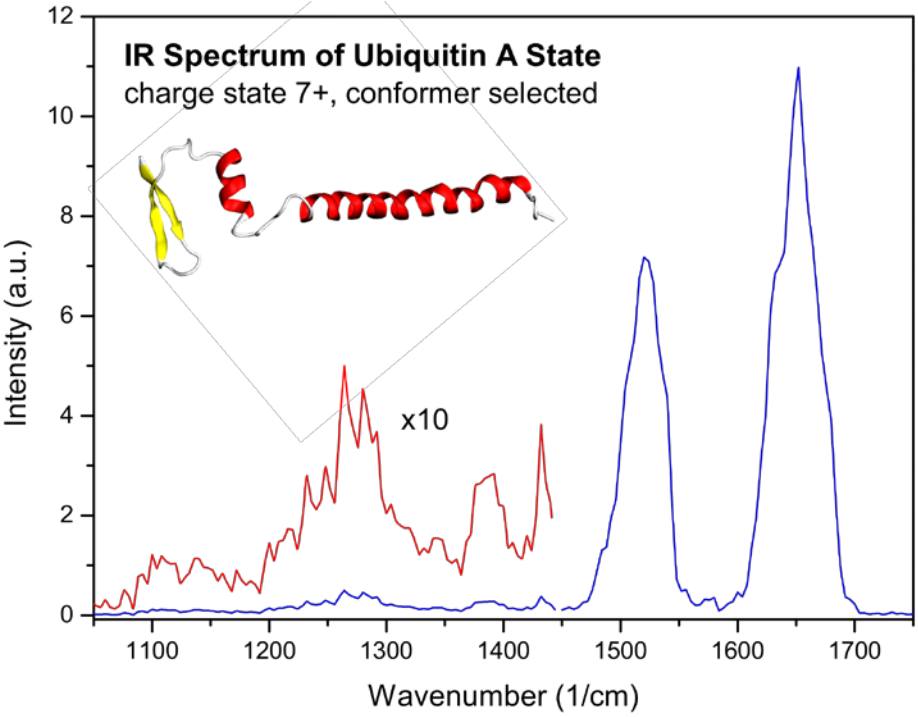


Figure 7: *Gas-phase IR spectrum of the protein ubiquitin (76 amino acids) in the charge state 7+, measured with the FHI FEL. Prior to the interaction with the FEL radiation, the molecules are conformer selected using ion mobility methods as well as mass/charge selected (von Helden group, FHI Dept. MP).*

deposited clusters.

Another example are the experiments currently being set up by the PC Department (group of A. Paarmann) which will investigate spin-phonon and phonon-phonon couplings in solids using mode-selective phonon excitation with the FEL radiation. A first experiment complements studies performed in the group of T. Kampfrath who investigate ultrafast demagnetization of the ferrimagnet Yttrium Iron garnet (YIG) after resonant excitation with laser-based broadband THz pulses. In contrast, the FEL experiment makes use of the narrow line width of the FEL radiation not accessible from laser-based sources, and will specifically address the question of mode selectivity in the demagnetization process (see Posters PC 14 and 17). The experiment will investigate the nonlinearities in the phonon absorption spectra of YIG by FEL-based reflectance spectroscopy, as well as the spectral dependence of the

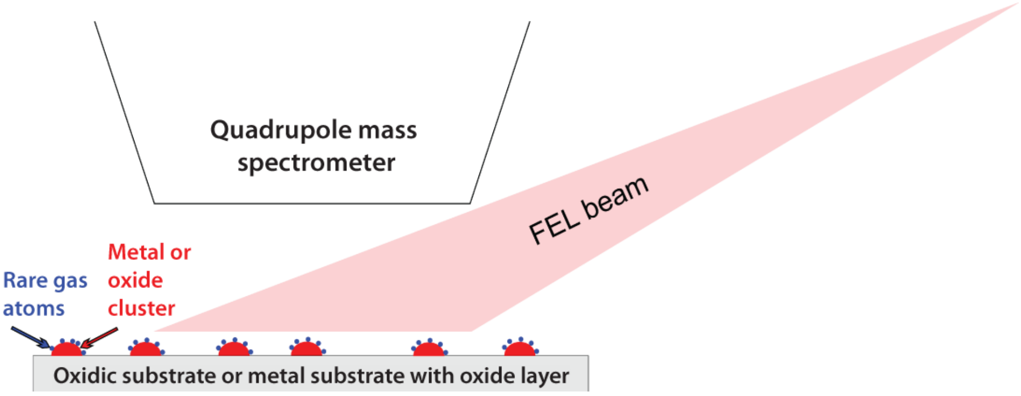


Figure 8: *Proposal for vibrational spectroscopy of metal clusters deposited on a metal oxide surface (Helmut Kuhlenbeck, FHI Dept. CP).*

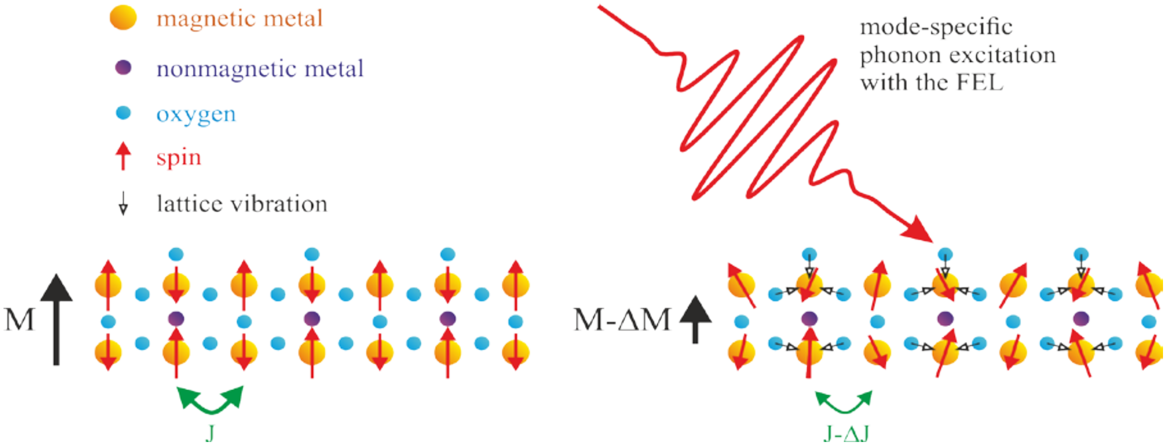


Figure 9: *Schematic of the experiment on spin-phonon coupling in a ferrimagnet. The FEL radiation is used to resonantly excite specific phonon modes in the crystal to investigate their resonant coupling to the spin system (e.g. modification of the exchange couplings J) by monitoring the FEL induced demagnetization of the sample (Alexander Paarmann, FHI Dept. PC).*

demagnetization due to the resonant FEL excitation using Faraday rotation of a visible probe laser beam.

Furthermore, it is noteworthy that external groups (including several from universities in Berlin, but also groups from other institutes within Germany and from abroad) have shown strong interest in using the FHI FEL. The reason is that the FHI FEL is one of just a handful of IR free-electron lasers worldwide and, compared to the existing FELs working with normal-conducting electron accelerators, it is designed to surpass the performance of the existing ones. As the FHI FEL is not organized as a user facility offering beam-time based on a formal application-selection procedure, external groups will, possibly, start collaborations with FHI scientists to use the FHI FEL in the future.

Summary

A new mid-infrared (4 – 50 μm) free-electron laser has been commissioned at the Fritz-Haber-Institut. It is operated with 15 – 50 MeV electrons from a normal-conducting S-band linac equipped with a gridded thermionic gun and a chicane for controlled bunch compression. First lasing was achieved in 2012. Peak macro-pulse energies in excess of 100 mJ have been observed, and first user beamlines have been commissioned. First user experiments (spectroscopy of transition-metal carbonyl clusters, aluminum oxide clusters as well as biomolecules in the gas-phase) have been made, with more to come before the end of 2013. In addition, a second FEL branch covering the FIR/THz regime from 40 to 500 μm has been designed leaving the option for a system upgrade in the future.

Publications of the FHI FEL Group

2011 (late publications)

Todd, A.M.M., H. Bluem, V. Christina, M.D. Cole, J. Ditta, D. Dowell, K. Jordan, R. Lange, J.H. Park, J. Rathke, T. Schultheiss, L.M. Young, W. Schöllkopf, S. Gewinner, W. Erlebach, H. Junkes, A. Liedke, W. Zhang, G. von Helden, G. Meijer and S.C. Gottschalk: Commissioning Status of the Fritz Haber Institute THz FEL. In: Proceedings of 2nd International Particle Accelerator Conference, IPAC'11, San Sebastián, Spain. THPC106 (2011). ISBN 978-92-9083-366-6.

2012

Schöllkopf, W., S. Gewinner, W. Erlebach, H. Junkes, A. Liedke, G. von Helden, W. Zhang, G. Meijer, H. Bluem, V. Christina, M.D. Cole, J. Ditta, D. Dowell, K. Jordan, R. Lange, J.H. Park, J. Rathke, T. Schultheiss, A.M.M. Todd, L.M. Young, U. Lehnert, P. Michel, W. Seidel, R. Wünsch and S.C. Gottschalk: Status of the Fritz Haber Institute THz FEL. In: Proceedings of 33th International Free Electron Laser Conference, Shanghai, China. TUPB30 (2012). ISBN 978-3-95450-117-5.

Todd, A.M.M., H. Bluem, J. Ditta, D. Dowell, K. Jordan, R. Lange, H. Loos, J.H. Park, J. Rathke, L.M. Young, W. Erlebach, S. Gewinner, G. von Helden, H. Junkes, A. Liedke, G. Meijer, W. Schöllkopf, W. Zhang, U. Lehnert, P. Michel, W. Seidel, R. Wuensch and S.C. Gottschalk: Commissioning of the Fritz Haber Institute Mid-IR FEL. In: Proceedings of 3rd International Particle Accelerator Conference, IPAC'12, New Orleans, Louisiana, USA. TUPPP087 (2012). ISBN 978-3-95450-115-1.

2013

Bluem, H.P., D. Dowell, H. Loos, J. Park, A.M.M. Todd, L.M. Young, S. Gewinner and W. Schöllkopf: Accelerator Beamline Performance for the IR FEL at the Fritz-Haber-Institut, Berlin. In: Proceedings of 34th International Free Electron Laser Conference, Nara, Japan. WEOC04 (2013). ISBN 978-3-95450-123-6.

Gottschalk, S.C., T.E. DeHart, R.N. Kelly, M.A. Offenbacker, A.S. Valla, H. Bluem, D. Dowell, J. Rathke, A.M.M. Todd, S. Gewinner, H. Junkes, G. Meijer, W. Schöllkopf, W. Zhang and U. Lehnert: Design and Performance of the Wedged Pole Hybrid Undulator for the Fritz-Haber-Institut IR FEL. In: Proceedings of 34th International Free Electron Laser Conference, Nara, Japan. THPD13 (2013). ISBN 978-3-95450-123-6.

Junkes, H., W. Erlebach, S. Gewinner, U. Hoppe, A. Liedke, G. Meijer, W. Schöllkopf, G. von Helden, M. Wesemann, A.M.M. Todd, L.M. Young, H. Bluem, D. Dowell, R. Lange and S.B. Webb: Integration the FHI-FEL into the FHI Research Environment - Design and Implementation Aspects. In: Proceedings of 35th International Free Electron Laser Conference, New York, USA. WEPSO30 (2013). ISBN 978-3-95450-126-7.

Junkes, H., W. Schöllkopf, M. Wesemann and R. Lange: Design and Implementation Aspects of the Control System at the FHI FEL. In: Proceedings of 14th International Conference on Accelerator & Large Experimental Physics Control Systems, San Francisco, CA, USA. In press.

Schöllkopf, W., S. Gewinner, W. Erlebach, G. Heyne, H. Junkes, A. Liedke, V. Platschkowski, G. von Helden, W. Zhang, G. Meijer, H. Bluem, M. Davidsaver, D. Dowell, K. Jordan, R. Lange, H. Loos, J. Park, J. Rathke, A.M.M. Todd, L.M. Young, U. Lehnert, P. Michel, W. Seidel, R. Wünsch and S.C. Gottschalk: First Lasing of the IR FEL at the Fritz-Haber-Institut Berlin. In: Proceedings of 34th International Free Electron Laser Conference, Nara, Japan. MOOB0 (2013). ISBN 978-3-95450-123-6.

Schöllkopf, W., S. Gewinner, W. Erlebach, G. Heyne, H. Junkes, A. Liedke, G. Meijer, V. Platschkowski, G. von Helden, H. Bluem, M. Davidsaver, D. Dowell, K. Jordan, R. Lange, H. Loos, J. Rathke, A.M.M. Todd, L.M. Young, U. Lehnert, P. Michel, W. Seidel, R. Wünsch and S.C. Gottschalk: The IR and THz Free-Electron Laser at the Fritz-Haber-Institut. In: Proceedings of 35th International Free Electron Laser Conference, New York, USA. WEPSO62 (2013). ISBN 978-3-95450-126-7.

Joint Network Center (GNZ) (Head: Gerd Schnapka)

The *Joint Network Center* (Gemeinsames Netzwerkzentrum, GNZ) of the Berlin-Brandenburg Max Planck institutions is a regional IT competence center. This facility focusses on networking, backup, virtualization and security services. At present, the center supports 13 Max Planck institutions in the Berlin-Brandenburg area including all 8 Max Planck Institutes.

The GNZ deals with wide area (DFN X-WiN), metropolitain area (BRAIN and to Potsdam) and local area (LAN and WLAN) networks. Special emphasis is placed on network security and central manageability. Apart from its network activities the GNZ offers IT services such as backup, server virtualization and virtual private network (VPN). In addition, the GNZ provides consulting services and coordinates IT projects from planning to purchase.

In 2012, the GNZ has installed a uniform new network infrastructure at all participating institutions [Figure 1]. Beyond this, a redundant 10GBit backbone was created across the entire campus of the FHI. Because of that, a better performance and higher reliability was achieved.

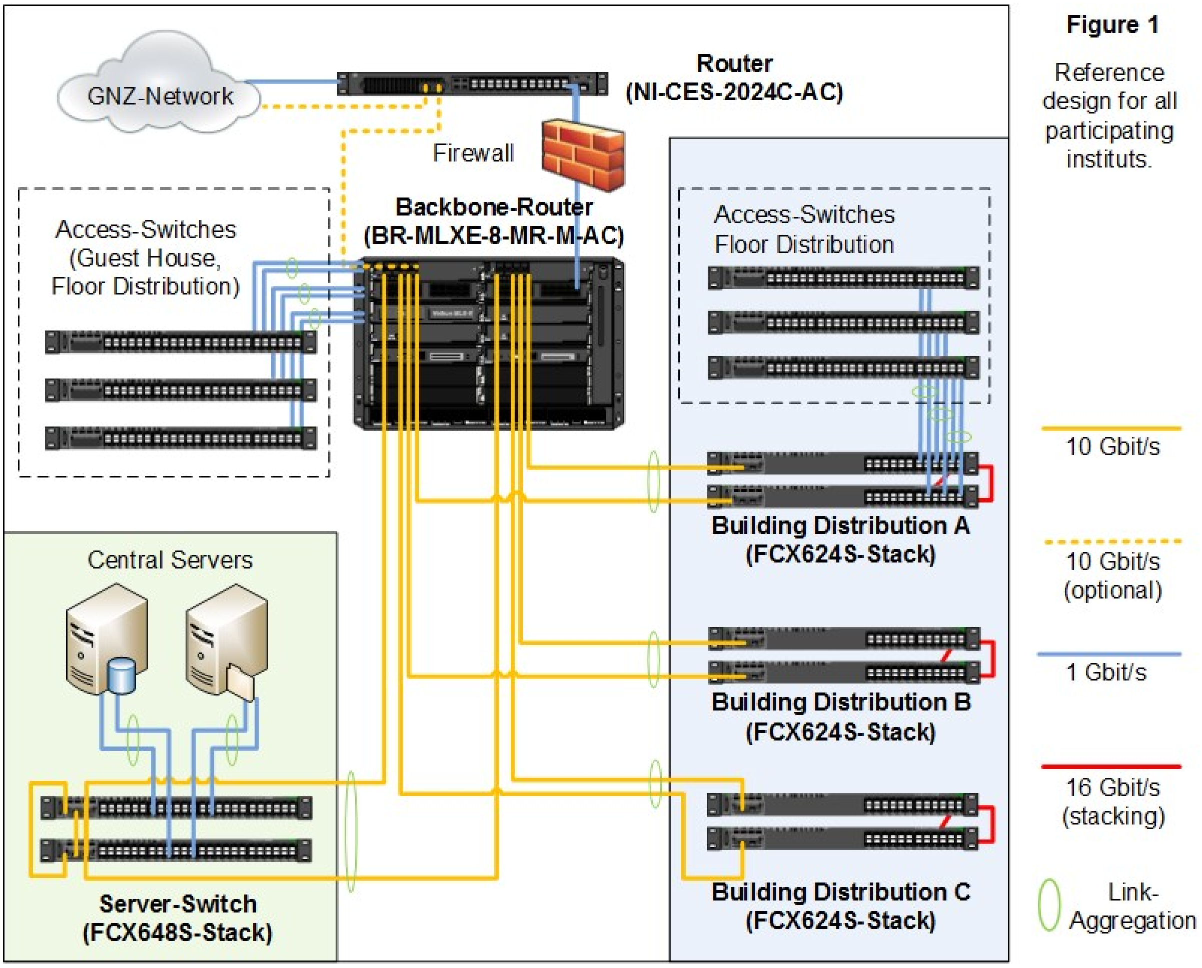


Figure 1

Reference design for all participating instituts.

The WLAN (Wi-Fi) was renewed by the GNZ in 2013 in all participating institutions. In the FHI an entirely new system was installed. This benefits in addition to our scientists also guests in the Eduroam and guest Wi-Fi network. A new intrusion prevention system (IPS) was conceived by the group in 2013 between the main connection of the GNZ and the DFN. This system was taken in operation in the second half of 2013. Malware, botnets etc. are blocked and the institute-firewalls remain free of Distributed Denial of Service (DDoS) attacks due to the IPS. The group had a leading role in the MPG IPv6 project. The IPv6 address allocation from RIPE NCC (Réseaux IP Européens Network Coordination Centre) were distributed to all Max Planck Institutes. Training for IT staff was also planned.

At present, the data area of the backup contains about 5 Petabyte on approximately 6000 LTO-Tapes. For increased security, a secondary copy of all archived data is saved in a tape library of the Gesellschaft für wissenschaftliche Datenverarbeitung Göttingen (GWDG). In 2012 the group had extend the existing backup strategy to incremental-forever incl. deduplication. After that the plattform strategy was changed to Intel/Linux. Mobile clients (Notebooks, Tablets and Smartphones) are becoming increasingly important. A internal data protection for mobile clients are also available since 2013. The whole communication incl. the in-house data storage are highly encrypted.

PP&B (Personal Computers, Realtime & Image Processing) (Head: Heinz Junkes)

Based on the data network infrastructure, provided by the *Joint Network Center* (GNZ), the PP&B group is responsible for the installation and maintenance of the desktop computer systems (including laptops), server systems, telephone system and the electronic building control system. Several server systems perform services for mail, web, printing, number crunching and databases for different archives (elogbook, channel archiver, document archives).

The core of it all is a high available storage system (Netapp metro cluster, 70 TB), interconnected with redundant 10 Gbit switches. Most server systems are running as so called virtual systems on well equipped hardware devices that are also connected via redundant paths to this 10Gbit Network. The technology used allows us to create snapshot copies while applications are running. Each user at the institute can recover files and directories that were deleted, corrupted or altered on the network drives without PP&B intervention.

The technical consulting service supports scientists of the institute in the configuration, modification, acquisition, and adaption of computers equipment to the experiments (in close cooperation with the service group of electronics). Since the last meeting, the group has been deeply involved in the EPICS (Experimental Physics and Industrial Control System) based control and archiving system of the new free electron. The group developed key components such as cavity stabilization, beam diagnostic, gun control and a machine protection system (see Fig. 1).

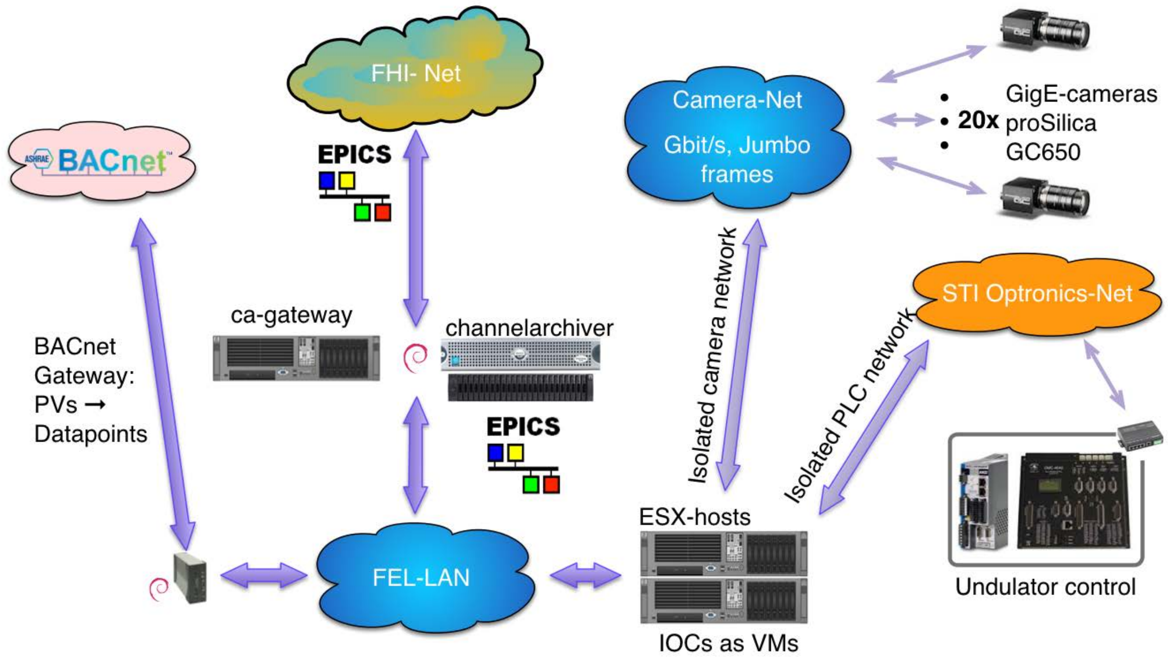


Figure 1

The FEL facility should be easy to use by the scientists at the institute and is seamlessly integrated into the existing research environment. The graphical operator interface covers radiation safety monitoring as well as controlling the complete set of building automation and utility devices, regardless of their particular function. A user interface (subset of the operator interface) allows user-provided experimental-control software (LabVIEW, Matlab, …) to connect with an Gateway providing secured access. A channel-archiver continuously records selected process variable data and provides a web server offering archive and near real-time data [1, 2].

For the next semester, a block course at the IMPRS "Complex Surfaces in Materials Science" with the title "Data Acquisition – How to" is provided. It will cover theoretical foundations of signal analysis. In this course, the Students will be able to get hands-on experience with real systems.

The current compute cluster (xfhix) was put into operation in early 2009 and is to be replaced next year (2014). An application for a system consisting of commodity hardware connected to a 10 Gbit switching factory and centralized storage will be submitted to the BAR (Beratender Ausschuss für Rechnersysteme).

[1] Junkes, H., W. Erlebach, S. Gewinner, U. Hoppe, A. Liedke, G. Meijer, W. Schöllkopf, G.v. Helden, M. Wesemann, A.M.M. Todd, L.M. Young, H. Bluem, D. Dowell, R. Lange and S.B. Webb: Integration the FHI-FEL into the FHI Research Environment - Design and Implementation Aspects. In: Proceedings of 35th International Free Electron Laser Conference, Manhattan, USA. ISBN 978-3-95450-126-7

[2] *Junkes, H., W. Schöllkopf, M. Wesemann and R. Lange:* Design and Implementation Aspects of the Control System at the FHI FEL. In: Proceedings of 14th International Conference on Accelerator & Large Experimental Physics Control Systems, San Francisco, CA, USA. In press.

Mechanical Workshop (Head: Petrik Bischoff)

The mechanical workshop consists of three groups: Precision Engineering (15 technicians, 2 trainees), Mechanical Technology (2 technicians) and Wood Technology (1 technician).

In general, the groups deal with:

1. The construction and development of scientific instruments and apparatus.
2. Fine equipment and fixtures as well as repairs.
3. The creation of CAD drawings.
4. The processing of different types of material through conventional manufacturing processes as well as soldering, welding, CNC milling / turning, electrical discharge machining, grinding and polishing.

In April 2013 Petrik Bischoff became the head of the workshop. He and his team are building on the previous high quality of the workshop and adapt to the increasing demands set by new materials, technology, and experimental requirements. Those ever more demanding requirements set by science of increased dimensional accuracy and of new materials require that the equipment in the workshops as well as the working practice must be continuously modernized.

A first step was the purchase of a 5-axis CNC milling machine (DMU 60evo) in May. The manufacturing of complex, intricate parts and assemblies is now easier to implement. Another important specialization of the precision engineering department is wire and die-sinking electrical discharge machining (EDM). The eroding team will receive a special CNC machine on the 18th of December 2013 that will be able to drill holes of diameter 0.3 mm to a depth of 15 mm. In addition, in the first quarter of 2014, an extension module (axis of rotation) will be mounted on the wire EDM machine. With the help of the rotation axis, special and unusual shapes are feasible.

To reduce or eliminate arcing of high voltage parts in high and ultrahigh vacuum, a process for surface refinement (vibratory grinding) has been adapted and specialized over the last few years. Our expertise in this area is recognized both within the FHI and beyond.

Regular meetings of the technicians in the precision engineering group with the designers situated in the various departments of the FHI result in strong cooperation, which allow for new ideas to be discussed and which help to optimize the workflow in the mechanical workshop.

Electronics Workshop (Head: Georg Heyne)

The service group for electronics is a central facility to support the scientific groups on different levels. The primary task is the development and construction of complex measuring- and control equipment, required for critical experiments, which are not available on the commercial market. In the last 2 years the E-Lab developed about 195 different devices.

These include for example very fast high voltage amplifiers for the Project Cold Molecules on a Chip (ERC), an ultra-fast high voltage switch (50kV) for the e-gun of the FEL and a complete control system for a Spatial Profile Macroreactor. Furthermore, components for computer-controlled data acquisition, for example very compact microprocessor units with AD- and DA converters with WLAN interfaces were developed.

The technical consulting service supports the scientists in the configuration and modification, the acquisition and adaptation of electronic-equipment for their experiments. A large stock of component and rental equipment allows for fast and inexpensive pilot tests. With new developments and optimized workflow, programming of logic array (FPGA) microcontroller using for example LabView as well as layout and construction of printed circuit boards is now a highly efficient process, which brings more effectiveness and transparency to the electronics workshop.

The cooperation with electronics development engineers of other MPG institutes is aided by regular meetings. Further, to keep the knowledge up to date, our staff members are send on a regular basis to training courses.

Library (Head: Uta Siebeky)

The library collects special literature covering the research fields of the institute. The range of electronic services of the FHI library includes eBook collections (i.e. RSC eBook Collection 1968-2009), electronic journals (about 30,000 currently subscribed), various databases as well as catalogues. The library holds about 15,500 monographs displayed in a new designed reading room. It is a common room for the whole institute.

The Fritz Haber Institute is practicing open access to scientific results as stated in the *Berlin Declaration on Open Access to the Knowledge in the Sciences and Humanities*. This declaration is the most significant result of the first Berlin Conference in October 2003. Berlin Conferences are annual congresses organized by the Max Planck Society and other research organizations and universities. This year's Berlin Conference was held on the occasion of tenth anniversary of the Berlin Declaration. (More information at http://openaccess.mpg.de/)

In the ten years since, the signatories of the Berlin Declaration and many other organizations have fostered a remarkable growth of Open Access, by supporting repositories, Open Access publishing, and campaigns to raise awareness of Open Access in the scientific, publishing, and political communities. But still only about 10% of peer-reviewed articles are published in Open Access journals. The Max Planck Society believes that the most important goal for the coming years is to ensure that 90% of the scholarly research literature is published with an Open Access model. The Max Planck Society calls on the signatories of the Berlin Declaration and on other organizations to make the research findings of its scientists available for the benefit of humanity, free of charge whenever possible.

The Fritz Haber Institute is a pioneer within the Max Planck Society in the field of Open Access publishing. The board of directors of the Fritz Haber Institute recommends that researcher deposit a copy of each of their published journal articles on MPG PuRe (pubman.mpdl.mpg.de) according to legal conditions and submit their work to Open Access journals. The library collects the copyright transfer agreements.

We are successful with the implementation of the *Berlin Declaration*. The library of the Fritz Haber Institute, together with the scientists, has developed and established an elaborated Open Access workflow and the FHI achieves now a level of 30% Open Access publications. The publications are collected in the repository of the Max Planck Society, MPG PuRe. Our perspective for the next decade is to increase our Open Access publications.

Department of Inorganic Chemistry Director: Robert Schlögl

Staff scientists:

Malte Behrens
Frank Girgsdies
Axel Knop-Gericke
Detre Teschner
Olaf Timpe (FHI Safety Officer)
Julian Tornow
Annette Trunschke
Marc Willinger
Sabine Wrabetz

Elmar Zeitler (Emeritus), Manfred Baerns, Klaus-Peter Dinse (Guest Emeriti)

Scientists, staying for at least six months, paid by FHI:

Elias Frei
Klaus Hermann
Tulio Rocha *(until 9/2013)*
Christoph Sprung
Dangsheng Su *(part-time)*
Andrey Tarasov
Elena Willinger-Kudrenko *(maternity leave)*
Gregor Wowsnick

Scientists, staying for at least six months, paid from external funds:

Rosa Arrigo
Idalia Bilecka
Raoul Blume
Sebastien Cap
Bambar Davaasuren *(until 11/2013)*
Maik Eichelbaum
Benjamin Frank *(until 12/2013)*
Matthias Friedrich
Nicole Giliard *(until 12/2013)*
Michael Haevecker
Xing Huang
Benjamin Johnson
Travis Jones
Stefanie Kühl *(until 12/13)*
Thomas Lunkenbein
Johannes Noack
Manfred Schuster

Alexander von Humboldt grants:
Mark Greiner
Juan Velasco Velez

Graduate students: 23 (10 from external funds)

Technicians: 7 (+ 4 for Service Group Electron Microscopy)

Trainees: 4 (including 2 master students)

Emmy-Noether Group:
Raimund Horn *since 6/2013 Full Professor at Technical University Hamburg-Harburg*

Service Group Electron Microscopy: 4 staff members

Library Service Group: Uta Siebeky, Katrin Quetting, Evelyn Prohn

Recent developments in the Department of Inorganic Chemistry

Director: Robert Schlögl

1. General

The department continued its research as outlined in the last report. The dominating process was the foundation of the new MPI CEC in Mülheim. Planning and operating this Institute represented substantial challenges that were met as will be described in the respective report of the MPI CEC. The coming into existence of the MPI CEC created significant organisational and scientific consequences for the AC department at the FHI.

The mission of the MPI CEC is to understand and develop the chemical processes that we require in order to convert chemical energy carriers. Catalysis is the underlying science and technology. The foundation concept of the Institute assumes that catalysis has a common science base independent of molecular or interfacial catalytic systems. Further, a combination of theory and experiment can deliver the knowledge for designing and optimizing chemical energy conversion reactions. The experimental program focuses on synthesis of both molecular and solid catalysts and their functional ("in-situ") analysis to arrive at a description of the operation suitable for theoretical analysis for its mechanism and its optimization potential. A generic target of the work is to replace the still essential use of noble metals in energy-related chemical conversion reactions.

This mission is pursued for the foreseeable future considering the basic processes of energy storage from renewable primary electricity and its integration into the energy system. The reactions of water splitting and of hydrogenation of N_2 and CO_2 were chosen as key reaction systems. It is obvious that such an institutional activity is of relevance for the development at the FHI. It was thus clarified that this plan is not in conflict with the evolution of the FHI as a whole. This was also discussed with the President of the MPG and with the Perspective Commission of the CPT section. The AC department forms a bridge between the two institutes FHI and MPI CEC at least for the service period of R. Schlögl as founding director. The activities of the AC department were thus re-structured to effectively transfer solid-state chemical knowledge to MPI CEC and to benefit from the knowledge on molecular systems available at MPI CEC.

The activities on carbon chemistry were transferred to the MPI CEC. There the synthesis of novel carbon forms was initiated as a platform activity to generate electrodes and electro-catalysts with and without addition of metal components. The CARBOCAT and GRAFOL projects were left at the FHI as these activities are not related to the core focus of the MPI CEC.

The electrochemistry activity as described in the outlook of the last report was initially set up at the AC department with branches of liquid phase reactions associated with water splitting and of solid state electrochemistry with the aim to understand modes of operation of Li ion battery systems. The liquid phase activity was transferred as a key activity of the new MPI in order to avoid duplication. Remaining are solid state synthetic efforts to generate OER electrodes based upon Mn oxides at the FHI whereby the testing of the systems will be done in MPI CEC in exchange for AC-FHI supporting synthesis efforts for noble metal oxide reference systems. The battery activity will stay at FHI (*J. Tornow*) and is now concentrating upon the Si-C interface as high-performance anode. The activity is embedded in a cooperation project with VW from where multiple insights into the conventional slurry process of electrode manufacturing provide the relevant research questions that we address with a model synthetic effort (CVD synthesis) as well as with a new design concept (Si on hierarchical carbon synthesized at MPI CEC).

The electron spectroscopy project interrogating liquid-solid interfaces at reaction conditions as announced in the outlook of the last report has been brought to life in the EMIL cooperation with the HZB Berlin. We have completed the design and building (*A. Knop-Gericke*) of a NAP-XPS end station that is capable of operating at up to 7 keV electron energy and at pressures compatible with liquid water surfaces. This end station is now used (at lower energies) at the ISSIS beamline. The twin undulator EMIL beamline is under construction and will become operational in 2016. The MPG holds 50% of the beam time. The unforeseen success of NAP-XPS as method has generated a huge demand of user beam time that we try to fulfil with our team at ISSIS. This service now overloads the capacity of the AC-FHI team. We plan to extend the activity in “chemical electron spectroscopy” into a joint laboratory MPG-HZB. This will involve the teams of MPI CEC and of AC-FHI, several groups of the HZB (*Bär, Lips, van de Krool*) and university users (*Jägermann, Darmstadt*) and aims at a unification of the NAP-XPS and related spectroscopic methods applied in chemistry into a common experimental platform. We plan to set up a cluster of two bending magnet beamlines and one undulator beamline in a common chemical research environment for in-situ spectroscopy at the gas-solid and the liquid-solid interfaces. AC-FHI will transfer its two end-stations to this cluster, *Jägermann* will provide his liquid-solid analysis end-station and HZB will provide the beamlines and a group of beamline scientists. AC-FHI will provide expertise and training and will thus make sure that a centre of competence for in-situ analysis of reacting interfaces develops in this joint lab. To ensure continuity beyond the closure of the department AC (in 2021) and to support collaboration with the theory department at MPI-CEC (*F. Neese*) and the there existing activity in near edge X-ray spectroscopy (*S. De Beer*) the whole group "electron spectroscopy" (*A. Knop-Gericke*) will be transferred into the MPI CEC when RS finishes service as founding director at MPI CEC.

As announced in the outlook of the last report the department entered into a process of renewing its microscopy hardware. As recommended in the last report of the Beirat the department analyzed the evolution in the field of electron microscopy techniques. A conference on the topic "EM-CAT 2013" was organized bringing together worldwide expertise in the field of chemical applications of electron microscopy. Two key developments are relevant for the work of the department in "chemical electron microscopy". One concerns the high-resolution abilities of various STEM techniques with probe aberration correctors and cold field-emission electron sources. The other concerns the advent of sample holders with windows in MEMS technique allowing with additional developments to perform in-situ TEM with environmental control at atomic resolution without compromising the performance of the microscope. An analytical aberration corrected TEM with 300 keV acceleration voltage is essential for this application.

Another technological trend is the fully automated electron microscope. We engage in a co-operation with the manufacturer to develop user profiles and sample holder systems allowing synthetic chemists to examine their samples with material science TEM techniques without having to be trained in electron microscopy from their desks. In addition the respective hardware will allow using TEM in normal laboratory rooms without special building requirement bringing down drastically the cost of ownership of TEM. We plan to install test systems at FHI and at MPI CEC and to perform the necessary development work in close cooperation with the manufacturer sending a co-worker in residence to FHI allowing access to the technology of the microscope.

This development allowed us to order a new double-aberration corrected TEM to be installed in the AC department without building activities. We obtained an external grant of 2.3 mio € in support. We also were able to buy the basic technologies for the environmental sample holder development and begin with the implementation and testing. We need to exchange our existing TITAN instrument for effective in-situ work with an upgraded version including a dynamical piezo stage, new software and effective EDX capabilities. This instrument shall be exchanged for our existing TITAN and the development versions of the automated TEM instrumentation. A package offer was negotiated. We have submitted a grant application to the MPG.

The high-end instrumentation works with better results in a dedicated building without interfering electrical fields that limit the performance in Berlin. It is thus planned to transfer the electron microscopes also to MPI CEC where a dedicated stand-alone building for electron microscopes will be erected. There the instruments will be used in collaboration with MPI KOFO who recently bought an ultrahigh performing analytical instrument complementing the performances of our instruments. In this way a MPG centre for electron microscopy focussing on chemical application and spectroscopy (EELS) will form uniting instrumentation and competence at the MPG chemistry campus in Mülheim. We expect to move the instruments in 2017. The material science microscopy supporting the AC department may stay in Berlin, in particular when other departments need access to this infrastructure.

2. Structure of the Department

The structure of the department followed the proven concept. *R. Horn* took a W3 position at TUHH following the termination of the Emmy Noether group. The research on chemical engineering aspects of high temperature catalytic reactions was discontinued. The equipment was given to *R. Horn* and a collaboration agreement was signed to perform work on profile reactor measurements of propane oxidation.

A new group was established around an activity developed in the group “Reactivity” (*A. Trunschke*). The method of microwave perturbation analysis allowed in-situ analysis of electrical resistivity on the chemical potential of the environment. It was found to be a sensitive probe[1] of the operational state of semiconducting oxidation catalysts. The new

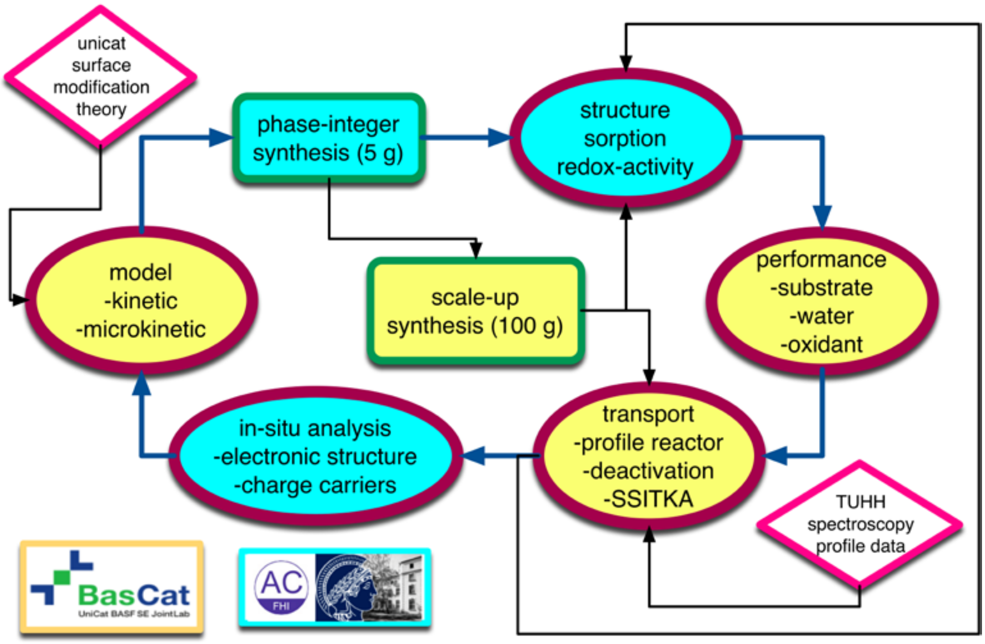


Figure 1: *The role of the AC department in the BASF-TUB Joint lab BASCAT. The COE UniCat contributes as well as R. Horn formerly from the AC department now at TUHH.*

group leader (*M. Eichelbaum*) will now extend the method to measure the Hall effect of the reacting sample in order to quantify the participation of charge carriers in catalytic redox reactions. The general target of this group is to quantitatively establish the role of semiconducting properties of oxide catalysts in regulating the charge carrier dynamics (equivalent to redox equivalents) in selective oxidation reactions. In this way several qualitative ideas about selectivity in oxidation catalysis will be traced back to physical observables following historic concepts emphasizing the role of semiconductors in catalysis. The group will also be active in using EPR for several projects. As the department does not have substantial EPR instrumentation we explore collaborations with MPI CEC and the FU Berlin where substantial EPR facilities exist.

The group “Reactivity” (*A. Trunschke*) is deeply involved in the new joint laboratory BASF-TU Berlin “BASCAT” established in 2012. Its mission is to find the mode of operation of selective oxidation catalysis beyond the current set of phenomenological “pillars” with predictive potential. This activity is scheduled for a ten-year lifetime and includes a dedicated building on the TUB campus. The project represents the contribution of the AC department to the COE “Unifying concepts in catalysts (UniCat)”. The joint lab was largely made possible through the involvement of the AC department as it is indicated by the essential role the department plays in the operational concept of BASCAT given in Figure 1. The collaboration allows the AC department to participate in extensive catalytic testing and kinetic data analysis that will be supported by a dedicated W3 appointment at the TUB.

The AC department contributes with its extensive synthetic competence using hydrothermal methods and with several in-situ analytical tools. Amongst them for us novel tools such as resonance RAMAN spectroscopy and low-energy PES are being implemented. The department received a substantial grant from BASF to set up the necessary infrastructure at the FHI. This partnership strengthened the focus on selective oxidation and the department can expect to accelerate substantially its activity in finding the generic mode of operation of selective oxidation with oxide systems. Details will be reported in section 3.

In the group "Electronic Structure" (*A. Knop-Gericke*) *T. Rocha* one of the competence carriers on NAP-XPS received a position LSLS in Brazil. *M. Greiner,* a post doc on an AvH research grant and *A. Klyushin* who is finishing his doctoral thesis in a collaborative project with the Boreskov Institute of Catalysis (*V. Bukhtiarov*) fill the gap.

The AC department attracted *Prof. K. Hermann*, a former member of the theory department who retired. He is supporting the interpretation of X-ray spectroscopic results in several activities and advises us on theoretical aspects of our work.

The structure of department with its two branches at FHI and MPI CEC is shown in the table. The activities of the CEC team will be reported in the CEC report. As multiple interactions exist between the two parts the full structure is shown here. In the following research highlights the results of the FHI team will be described.

Nr	Name	Leader	Competence 1	Competence 2	Competence 3	Core project
	AC FHI					
1	C1 Chemistry	Behrens	Low pressure synthesis	Diffraction	Thermal analysis	Methanol synthesis
2	Reactivity	Trunschke	High pressure synthesis	IR, RAMAN	Optical spectroscopy	Alkane oxidation
3	Solid state electro-chemistry	Tornow	CVD synthesis	Battery testing		Si as Li electrode
4	Electronic structure	Knop-Gericke	NAP-XPS	XAS	Adsorption	Coin metal oxidation
5	Charge carrier analysis	Eichelbaum	Microwave perturbation analysis	Hall effect	EPR	Charge carriers in oxide catalysts
6	Electron microscopy	Willinger	TEM, SEM	ESEM, ETEM	EELS	Method development
	AC CEC					
7	Nanocarbons	Reiche	Solvothermal synthesis	Mass spectrometry		Synthetic carbon
8	Water electrochemistry	Ranjan	Electro-chemistry	In-situ RAMAN		OER electrodes
9	Hybrid materials	Bulller	CVD synthesis	Nanoparticle synthesis		Electrocatalysts
10	Energy Catalysis	Bukhtiyarova	High pressure reactors	Catalytic probe reactions		Ammonia synthesis

The table reveals the portfolio of competencies in the department. The role of chemical synthesis under controlled conditions was strengthened through the groups at MPI CEC and the analytical capabilities were maintained mainly in the FHI team. The groups at MPI CEC are substantially smaller than in the FHI teams as the group had not even adequate office space until October 2013. The department in Berlin is reaching its limits in chemical laboratory work that would greatly benefit from the addition of technical support staff for which we have no positions available.

3. Graduate Students

Graduate students contribute substantially to the operation of the department. A suite of measures are thus in practice to support education and supervision of the students in addition to the regulations of the respective universities from where they are given their degrees.

Members of the department actively take part in the teaching activities of graduate students in relevant schools. We lecture in courses of the FHI-centred IMPRS “Complex Surfaces in Materials Sciences” and in the graduate school of the COE “BIG-NSE”. In addition the department organizes (*A. Trunschke*) a lecture series “Modern Methods in Catalysis” jointly with the three Berlin universities. Members of the FHI and external lecturers cover broadly basics and advanced aspects of heterogeneous catalysis in a 3-year curriculum. The program and details about crediting can be found on the website: http://www.fhi-berlin.mpg.de/acnew/department/pages/teaching.html. For the AC-FHI students the participation is compulsory and is pre-requisite for thesis submission. Additional requirements are the presentation of the thesis work in at least two conferences and the submission of ideally 3 publications.

The dissertation projects are defined in an initial period of 3 months in which the students get acquainted with the respective literature. They write a project proposal that is agreed with the group leader and is approved by the director. This text has the quality of the introductory chapter of the thesis and serves as definition of the work to be accomplished in the thesis period. The format of the thesis is a cumulative presentation of manuscripts together with an introduction and a final summarizing section of the results.

The group leaders supervise the students who work in teams within the groups and within the department. The project structure of the work at the department requires collaboration between the students. Project meetings held every 6 months and workshops of the students with the director ("Doktorandentage") every 6 months ensure progress monitoring and give opportunities to adjust guidelines of the work.

4. Cumulative progress of the past six years

In 2007 the department formulated a working hypothesis of how to close the material gap between model and performance heterogeneous catalysts. On the basis of the then concluded extensive studies of the iron oxide catalyst in ethylbenzene dehydrogenation[2] and of zirconium oxides in alkane isomerization[3] it was concluded that performance catalysts come into existence by a reaction of the as-synthesized pre-catalyst with components of the reactants. In this way an active phase different from the initial form of the catalyst should contain the active sites. This active phase was assumed to be unstable against decomposition in several stable end phases. The respective transformations causing eventual catalyst deactivation are made possible by the reaction energy transported through the catalyst.

In the past 6 years this working hypothesis was broadly substantiated and generalized for several classes of heterogeneous catalysts and several reactions. We consider performance catalysts now as dynamical systems. An active phase different in structure and composition from the equilibrium phase of the pre-catalyst is terminated by supra-molecular structures that fluctuate around a mean structure and form in this way active sites with short life times. After its conversion into the active complex by adsorption of reactants and desorption of products the deactivated site falls back into the matrix where it is regenerated into the active phase from where it may be converted into an active site. The local chemical potential that may differ from the overall chemical potential by transport phenomena controls the dynamics and kinetics of these transformations. Enabling for this is the real structure of the precursor that is defined by deliberate nano-structuring and often by unintentional defect structures referred to as "chemical memory" or "black magic" during synthesis of the pre-catalyst. Figure 2 reports the complexity of the mode of operation of a performance catalyst with some of the critical feedback loops.

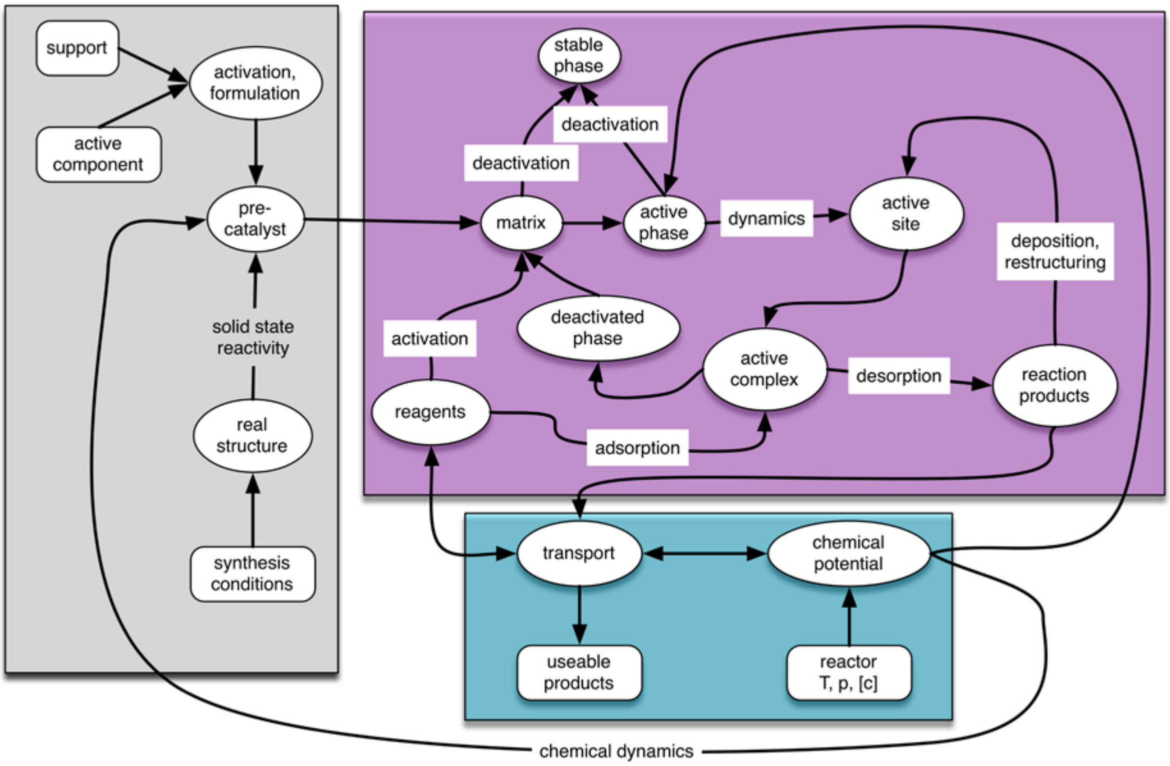


Figure 2: *The mode of operation of a performance catalyst indicated by a network of generic material states and processes. The coloured areas indicate the discipline of synthesis (grey), the area of chemical engineering (blue) and the field of atomic scale operation (red).*

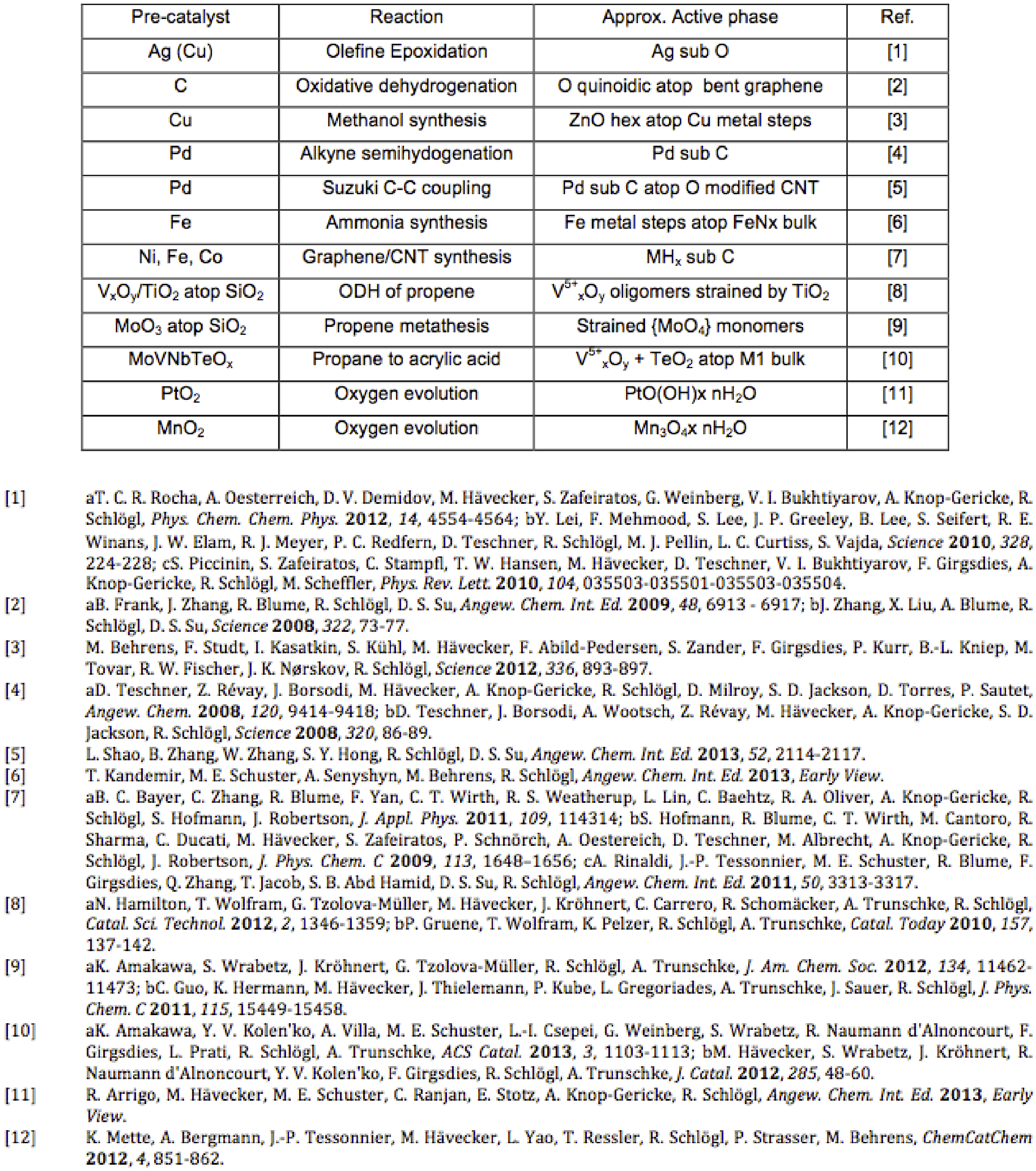
Their existence prevents the separation of the complexity into the areas indicated in the Figure. They may be named according to traditional branches of catalysis science as “catalyst synthesis” and “chemical engineering operation” and “atomic scale operation”. It is obvious from the Figure that the traditional separation of catalyst studies will barely describe the operating state and thus creates the “material gap”. We have developed in the AC department a suitable toolbox of in-situ techniques[4] that allow through combination of their individual results to derive the picture of operating performance catalysts without having to refer to model approaches. This has been done for a variety of different systems indicated in the following table. In all these cases the identified active phase was not the pre-catalyst. It occurs that the toolbox and the approach carry by now some generic character.

5. Selected results from the past two years

The following selection intends to highlight the progress in understanding of catalytic systems and their rational synthesis. A more complete account of the activities of the last two years may be found on our website under the group entries. Detailed reports on selected results are presented in the poster session.

5.1 MeOH synthesis (M. Behrens)

Based on a series of differently prepared Cu/ZnO/(Al_2O_3) catalysts that in part has been described in the previous report, a model[5] of the active site for methanol synthesis has been developed. The site ensemble exhibits two components: Steps at the Cu surface represent sites of increased activity and the presence of Zn further accelerates the conversion of O-bound intermediates. The steps were found to be stabilized by underlying planar defects in the Cu



Pre-catalyst	Reaction	Approx. Active phase	Ref.
Ag (Cu)	Olefine Epoxidation	Ag sub O	[1]
C	Oxidative dehydrogenation	O quinoidic atop bent graphene	[2]
Cu	Methanol synthesis	ZnO hex atop Cu metal steps	[3]
Pd	Alkyne semihydogenation	Pd sub C	[4]
Pd	Suzuki C-C coupling	Pd sub C atop O modified CNT	[5]
Fe	Ammonia synthesis	Fe metal steps atop FeNx bulk	[6]
Ni, Fe, Co	Graphene/CNT synthesis	MH_x sub C	[7]
V_xO_y/TiO_2 atop SiO_2	ODH of propene	$V^{5+}{}_xO_y$ oligomers strained by TiO_2	[8]
MoO_3 atop SiO_2	Propene metathesis	Strained $\{MoO_4\}$ monomers	[9]
$MoVNbTeO_x$	Propane to acrylic acid	$V^{5+}{}_xO_y$ + TeO_2 atop M1 bulk	[10]
PtO_2	Oxygen evolution	PtO(OH)x nH_2O	[11]
MnO_2	Oxygen evolution	Mn_3O_4x nH_2O	[12]

[1] aT. C. R. Rocha, A. Oesterreich, D. V. Demidov, M. Hävecker, S. Zafeiratos, G. Weinberg, V. I. Bukhtiyarov, A. Knop-Gericke, R. Schlögl, *Phys. Chem. Chem. Phys.* **2012**, *14*, 4554-4564; bY. Lei, F. Mehmood, S. Lee, J. P. Greeley, B. Lee, S. Seifert, R. E. Winans, J. W. Elam, R. J. Meyer, P. C. Redfern, D. Teschner, R. Schlögl, M. J. Pellin, L. C. Curtiss, S. Vajda, *Science* **2010**, *328*, 224-228; cS. Piccinin, S. Zafeiratos, C. Stampfl, T. W. Hansen, M. Hävecker, D. Teschner, V. I. Bukhtiyarov, F. Girgsdies, A. Knop-Gericke, R. Schlögl, M. Scheffler, *Phys. Rev. Lett.* **2010**, *104*, 035503-035501-035503-035504.

[2] aB. Frank, J. Zhang, R. Blume, R. Schlögl, D. S. Su, *Angew. Chem. Int. Ed.* **2009**, *48*, 6913 - 6917; bJ. Zhang, X. Liu, A. Blume, R. Schlögl, D. S. Su, *Science* **2008**, *322*, 73-77.

[3] M. Behrens, F. Studt, I. Kasatkin, S. Kühl, M. Hävecker, F. Abild-Pedersen, S. Zander, F. Girgsdies, P. Kurr, B.-L. Kniep, M. Tovar, R. W. Fischer, J. K. Nørskov, R. Schlögl, *Science* **2012**, *336*, 893-897.

[4] aD. Teschner, Z. Révay, J. Borsodi, M. Hävecker, A. Knop-Gericke, R. Schlögl, D. Milroy, S. D. Jackson, D. Torres, P. Sautet, *Angew. Chem.* **2008**, *120*, 9414-9418; bD. Teschner, J. Borsodi, A. Wootsch, Z. Révay, M. Hävecker, A. Knop-Gericke, S. D. Jackson, R. Schlögl, *Science* **2008**, *320*, 86-89.

[5] L. Shao, B. Zhang, W. Zhang, S. Y. Hong, R. Schlögl, D. S. Su, *Angew. Chem. Int. Ed.* **2013**, *52*, 2114-2117.

[6] T. Kandemir, M. E. Schuster, A. Senyshyn, M. Behrens, R. Schlögl, *Angew. Chem. Int. Ed.* **2013**, *Early View*.

[7] aB. C. Bayer, C. Zhang, R. Blume, F. Yan, C. T. Wirth, R. S. Weatherup, L. Lin, C. Baehtz, R. A. Oliver, A. Knop-Gericke, R. Schlögl, S. Hofmann, J. Robertson, *J. Appl. Phys.* **2011**, *109*, 114314; bS. Hofmann, R. Blume, C. T. Wirth, M. Cantoro, R. Sharma, C. Ducati, M. Hävecker, S. Zafeiratos, P. Schnörch, A. Oestereich, D. Teschner, M. Albrecht, A. Knop-Gericke, R. Schlögl, J. Robertson, *J. Phys. Chem. C* **2009**, *113*, 1648–1656; cA. Rinaldi, J.-P. Tessonnier, M. E. Schuster, R. Blume, F. Girgsdies, Q. Zhang, T. Jacob, S. B. Abd Hamid, D. S. Su, R. Schlögl, *Angew. Chem. Int. Ed.* **2011**, *50*, 3313-3317.

[8] aN. Hamilton, T. Wolfram, G. Tzolova-Müller, M. Hävecker, J. Kröhnert, C. Carrero, R. Schomäcker, A. Trunschke, R. Schlögl, *Catal. Sci. Technol.* **2012**, *2*, 1346-1359; bP. Gruene, T. Wolfram, K. Pelzer, R. Schlögl, A. Trunschke, *Catal. Today* **2010**, *157*, 137-142.

[9] aK. Amakawa, S. Wrabetz, J. Kröhnert, G. Tzolova-Müller, R. Schlögl, A. Trunschke, *J. Am. Chem. Soc.* **2012**, *134*, 11462-11473; bC. Guo, K. Hermann, M. Hävecker, J. Thielemann, P. Kube, L. Gregoriades, A. Trunschke, J. Sauer, R. Schlögl, *J. Phys. Chem. C* **2011**, *115*, 15449-15458.

[10] aK. Amakawa, Y. V. Kolen'ko, A. Villa, M. E. Schuster, L.-I. Csepei, G. Weinberg, S. Wrabetz, R. Naumann d'Alnoncourt, F. Girgsdies, L. Prati, R. Schlögl, A. Trunschke, *ACS Catal.* **2013**, *3*, 1103-1113; bM. Hävecker, S. Wrabetz, J. Kröhnert, R. Naumann d'Alnoncourt, Y. V. Kolen'ko, F. Girgsdies, R. Schlögl, A. Trunschke, *J. Catal.* **2012**, *285*, 48-60.

[11] R. Arrigo, M. Hävecker, M. E. Schuster, C. Ranjan, E. Stotz, A. Knop-Gericke, R. Schlögl, *Angew. Chem. Int. Ed.* **2013**, *Early View*.

[12] K. Mette, A. Bergmann, J.-P. Tessonnier, M. Hävecker, L. Yao, T. Ressler, R. Schlögl, P. Strasser, M. Behrens, *ChemCatChem* **2012**, *4*, 851-862.

nanoparticles, while the presence of Zn was observed to be due to strong metal support interaction (SMSI) probed by high resolution TEM (HRTEM) and near ambient pressure XPS (NAP-XPS). In situ-neutron diffraction under realistic methanol synthesis conditions[6] and a line profile analysis procedure sensitive to the presence of stacking faults revealed the stability of these defects under working conditions of the catalyst supporting their role as carrier of the highly active step sites.

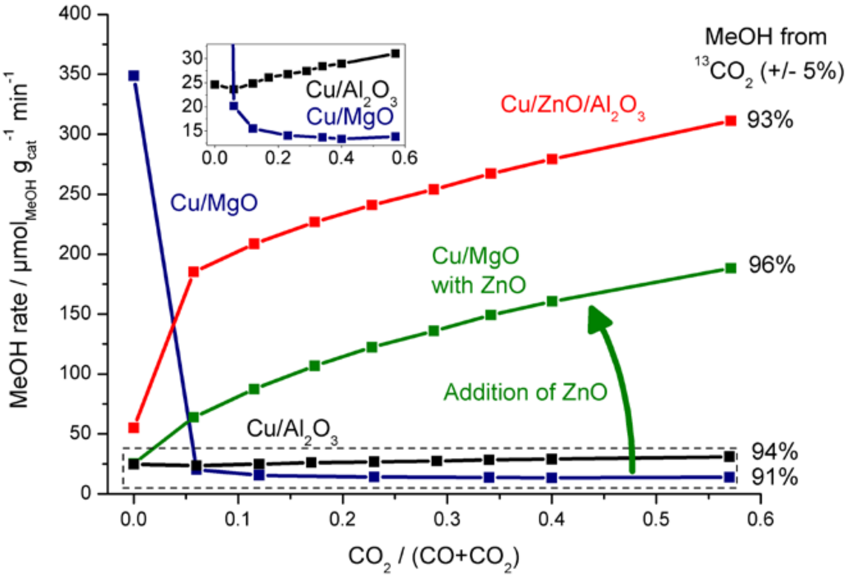


Figure 3: Methanol synthesis performance of different Cu-based catalyst in varying syngas compositions. Cu/ZnO/Al$_2$O$_3$ corresponds to a state-of-the-art industrial catalyst, Cu/MgO and Cu/Al$_2$O$_3$ are high- and low-surface area ZnO-free models and Cu/MgO/ZnO was prepared from Cu/MgO by impregnation with 5 wt.% ZnO. The percentage figures at the right hand side report the results of the ^{13}CO$_2$-labelling study using the CO$_2$-richest syngas composition.

The critical role of the ZnO component has been characterized using a Zn,Mg-substitution experiment[7] in the industrially applied catalysts synthesis procedure. Utilization of Mg instead of Zn enabled an analogous meso- and nanostructuring of the Cu/MO catalysts (M = Mg, Zn) due to the equivalence and similar ionic radii of Mg^{2+} and Zn^{2+} ions. The catalytic properties of the two high-surface area Cu/MO catalysts, however, revealed a completely different and inverse behaviour: Cu/MgO was highly active in a CO/H_2 feed and showed poor performance in CO_2-containing feeds, while Cu/ZnO was a much better catalysts in CO_2/(CO)/H_2 compared to a CO/H_2 feed (Figure 3). NAP-XPS showed that in case of the Cu/ZnO catalyst, a thin over-layer of ZnO_x species was formed at the surface of the catalyst, confirming SMSI between Cu and the reducible ZnO, which was absent for the irreducible MgO. New HRTEM evidence suggests that the Zn supply to the Cu metal surface is aided by the formation of a layered ZnO phase. The addition of a small amount of ZnO to the Cu/MgO catalysts completely changed its catalytic properties making it a reasonably good methanol synthesis catalyst from CO_2-contaning feeds and quenching its activity in CO hydrogenation at the same time. This result confirms that only small amounts of ZnO_x at the Cu surface, likely just enough to form the ZnO_x over-layer, are needed for the synergistic effect.

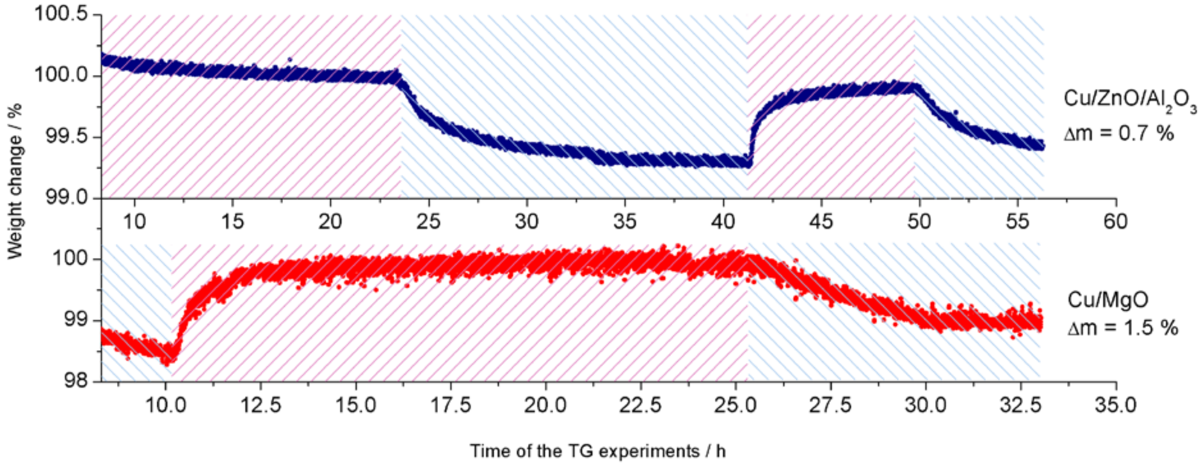


Figure 4: *In situ thermogravimetric analysis of the* $Cu/ZnO/Al_2O_3$ *(top) and the Cu/MgO catalysts (bottom) under working conditions (250 °C, 30 bar) in changing syngas feeds (magenta:* $CO_2/CO/H_2$*, blue:* CO/H_2*).*

The comparison of Cu/MgO and Cu/ZnO in Figure 3 suggests that the ZnO_x over-layer plays an important role for the hydrogenation of CO_2, while CO hydrogenation is negatively affected. Experiments with labelled $^{13}CO_2/CO/H_2$ syngas revealed that for all catalysts the major carbon source for methanol is CO_2 independent if the reaction is performance at a high rate on Cu/ZnO or at a low rate on Cu/MgO. This result shows that the high CO hydrogenation activity of Cu/MgO was completely poisoned by the presence of CO_2 in the feed as well as by the addition of ZnO to the catalyst. *In situ* TGA measurements under working conditions (30 bar, 250 °C) showed that both catalysts, Cu/ZnO and Cu/MgO, gain weight in the $CO_2/CO/H_2$ feed and reversibly lose weight in CO/H_2 (Figure 4). The weight change is related to the formation of a heavier formate-containing adsorbate layer in the presence of CO_2.

Based on the results, a consistent picture of industrial methanol synthesis slowly emerges:

1. CO_2 is the precursor for methanol.
2. Zn-decorated Cu steps are required to convert CO_2 (or formate).
3. These steps are generated and stabilized by planar defects in the bulk.
4. Zn is supplied to the defective Cu surface by SMSI.

5. This effect is probably favoured by the formation of a layered phase of graphitic ZnO and promoted by the introduction of defects in ZnO through “promotion”.
6. Cu catalysts can efficiently convert CO to methanol, but only if it is Zn-free and in the absence of CO_2, a situation that only occurs during low conversion model studies.

5.2 Complex metal oxides in propane oxidation (A. Trunschke)

The activation of small alkane molecules originating from natural gas or renewable resources on metal oxide surfaces regains increasing scientific interest since direct utilization of saturated hydrocarbons will offer alternative pathways to higher value chemicals in a post-petroleum era. The focus of the project lies on oxidation catalysis of small alkane molecules including methane, ethane, propane and butane. Oxidative dehydrogenation[8] of alkanes to the corresponding olefines or selective oxidation to functionalized monomers suffers from selectivity due to complex reaction networks[9]. The project addresses, therefore, the limits of selectivity in oxidation catalysis. Are the same active sites or catalyst components responsible for alkane activation, oxygen insertion and total combustion or will it be possible to avoid undesired consecutive reactions by intelligent catalyst design? What are the critical reaction parameters that determine the selectivity? We address these questions following a comprehensive approach that involves controlled synthesis of well-defined catalysts[10], detailed kinetic analysis of reaction networks, and the investigation of the molecular nature of the surface[11] considering also collective properties of the solid state, like crystal and electronic structure of the catalyst under reaction conditions. From the materials point of view, we concentrate on vanadium and molybdenum oxides, which are synthesized as crystalline bulk catalysts or supported monolayers[12] on meso-structured silica. Molybdenum and vanadium carbides[13] and nano-structured carbons are included for the purpose of reference. In the oxidative coupling of methane, doped and non-doped alkaline earth oxides have been investigated as model catalysts. Phase-pure M1 can be achieved by hydrothermal synthesis. We have monitored the formation of the prototypical mixed MoVTeNb oxide under hydrothermal conditions by in situ Raman spectroscopy[10a]. The spectroscopic information enabled the design of a new, accelerated, and reproducible hydrothermal route towards phase-pure M1 using a cascade synthesis approach. Modular cross-linking of structural building blocks by stepwise addition of V and Te under pressure directs the synthesis to the desired product, thereby avoiding the co-formation of unwanted phases. The isolated precipitate can be crystallized to phase-pure M1. The new M1 catalyst is characterized by high selectivity to acrylic acid in selective oxidation of propane, resulting in an increase in space–time yield by a factor of five. Current synthetic activities are aimed at controlled chemical modification of the M1 structure by restricting the composition to Mo and V only using the same methodology.

The analysis of catalytic data of acrylic acid synthesis from propane measured over a period of more than ten months under varying feed composition, temperature and contact times reveals that phase-pure M1 is absolutely stable[9] over such a long term. This is a very important finding with respect to the usability of the acquired kinetic data. The achieved phase purity is crucial for this high stability as demonstrated by constant lattice parameters observed during in-situ XRD experiments.

Analysis of the three-dimensional experimental parameter field measured in fixed bed reactors revealed that the complexity of the reaction network in propane oxidation over

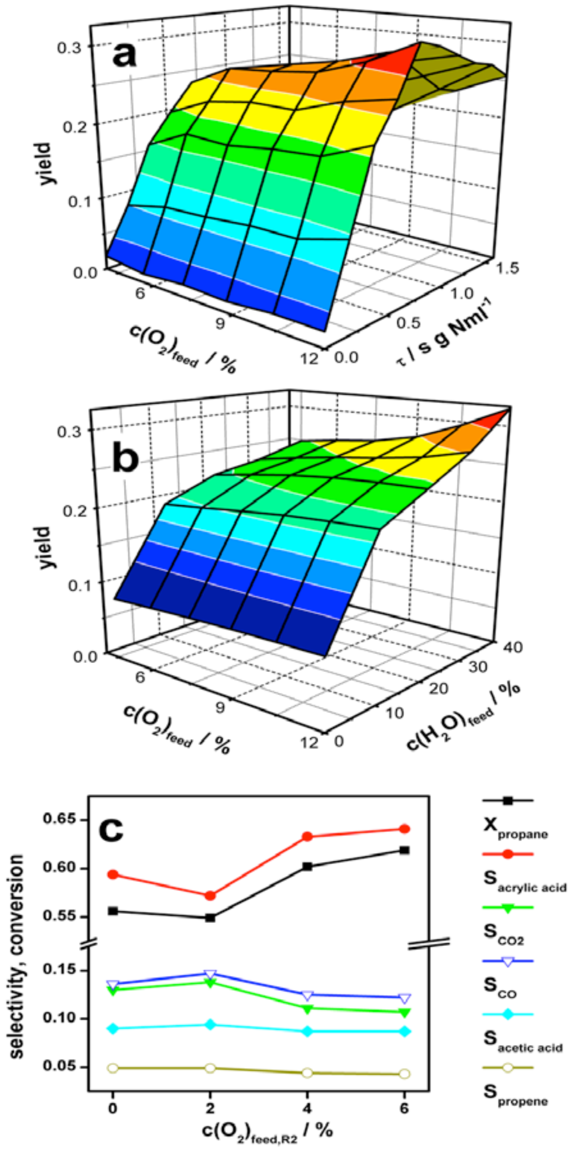


Figure 5: *(a) Influence of contact time and oxygen concentration of the feed on the yield of acrylic acid in oxidation of propane over M1 MoVTeNb oxide using feed gas mixtures that contain 3 % propane and 40 % steam. (b) Influence of steam and oxygen concentration of the feed on the yield of acrylic acid measured at 653 K and a contact time of 0.72 s g Nml-1. (c) Results of addition of* O_2 *between two reactors in series operated at a temperature of 673 K and a contact time of 0.4 s g* Nml^{-1} *per reactor in an initial feed containing 3 % propane, 6 % oxygen and 40 % steam balanced with nitrogen (amount of* O_2 *added between the two reactors in vol% is indicated on the x-axis).*

MoVTeNb oxide is reduced compared to less-defined catalysts due to phase purity and homogeneity. The oxidative dehydrogenation of propane to propene followed by allylic oxidation of propene comprises the main route to acrylic acid. Water and oxygen both fulfil besides functions as reactants critical roles in establishing and stabilizing the reactive termination structure. The oxygen partial pressure was identified as an important process parameter that controls the activity in propane oxidation over phase-pure M1 without negative implications on the selectivity (Fig. 5). High O_2 concentration in the feed keeps the catalyst in a high oxidation state, which provides an increased number of active sites for propane activation. Auxiliary steam increases activity and selectivity of M1 by changing the chemical nature of the active sites and by facilitating acrylic acid desorption.

The M1 surface composition and the vanadium oxidation state change dynamically and reversibly when the feed composition is varied, but only in the outermost surface layer. This characteristic behaviour is observable[14] by (NAP-XPS). One function of the thin active layer is to act as “oxygen buffer”, since excess of oxygen increases activity but does not deteriorate the selectivity showing that the chemical potential of oxygen does not directly control deep oxidation. The „oxygen buffer“ function can be realized by a structural flexibility allowing for interchange between bridging and terminal oxygen groups according to the gas phase chemical potential. Such a site may be envisaged as a molecular structure similar to a poly vanadate. The dynamic electronic structure of the orthorhombic M1 phase was also investigated by noncontact microwave conductivity technique. By measuring the binding energies of the core and valence levels and the secondary electron cut-off energy under different reaction conditions the alkane-dependent modulation of the work function, electron affinity, and band bending were determined. Whereas the alkane-dependent change of the electron affinity is explained by the formation or modification of the dipolar surface layer, the binding energy shifts and hence band bending are caused by Fermi energy pinning to the surface state energy of the V^{4+}/V^{5+} redox couple in the active layer. The complementary microwave conductivity studies indicate furthermore that the electron density in the conduction band is modulated by the gas phase as well. If the charge carrier transport scheme is known in detail and the energetic positions of the interfaces can be determined, then the reaction can be cast in a semiconductor band scheme allowing a quantitative energetic description more adequate than assigning formal oxidation states to isolated atoms. However, much speaks for a covalently bonded molecular feature as active site. In this picture the specific electronic structure of the support will become an important factor in explaining the reason why not every support of a polymeric V structure can act as selective oxidation catalyst.

5.3 Battery electrochemistry (J. Tornow)

The material system for our research on the battery anode is silicon due to its enormous theoretical charge storage capacity of 3580 mAh/g. We follow two different preparative approaches, the first one bases on the classical battery manufacturing, where a paste of nanometer sized silicon powder, carbon powder and binder is tape casted onto a metallic current collector. The carbon material increases the electrodes conductivity and also provides porosity where the silicon can expand into upon charging, since silicon undergoes volume expansions up to about 300% with lithiation. The disadvantage of this preparation method is its complexity by having several different components mixed in the electrode, which all might influence the interface chemistry. Furthermore, the particles of the silicon powder are always covered with an ill-defined native oxide layer. To circumvent this oxidation and also to be able to study the influence of the electrode components separately, we use a second preparation route based on a thin silicon film made by chemical vapour deposition (CVD) allowing also to generate artificial "SEI" (solid electrolyte interphase) systems of C, SiC, or SiN. We deposit the CVD-silicon either onto planar carbon or metal films to fundamentally study the reaction of pure silicon, but we also deposit the silicon into porous carbon substrates to increase the capacity and verify the potential of the CVD-preparation for realistic battery electrodes. The analysis of such systems is challenging as neither air nor nitrogen must be in contact with system to allow probing the Li species disposition. The department developed all necessary infrastructure for handling polycrystalline and planar model systems in SEM, TEM, XPS and FT-IT (ATR). Figure 6 stands as representative result revealing the complexity of the analytical challenge.

A microscopical analysis of paste based silicon anodes by SEM reveals the formation of an SEI overlayer after cycling the battery, Elemental analysis by energy filtered transmission electron microscopy (EFTEM) reveals that this SEI covers both, the silicon and the carbon particles. It is remarkable that the SEI besides carbon also contains silicon, even where it surrounds only the carbon particles.

We observe a linear relation of the charge storage capacity to the film thickness of CVD-prepared model silicon electrodes. We conclude that charges are stored in the silicon bulk and

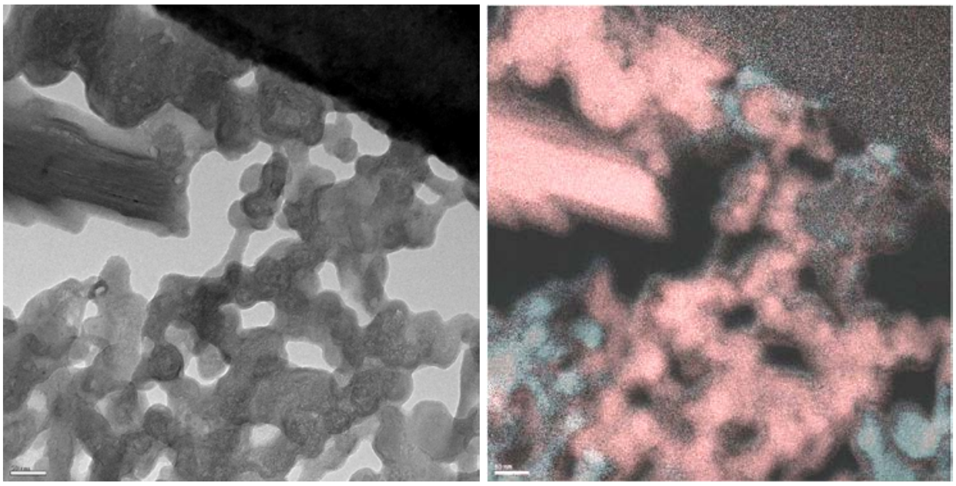


Figure 6: *TEM and EFTEM of a cycled battery anode consisting of silicon nanopowder, carbon and binder. The EFTEM shows a mixture of carbon (pink) and silicon (blue). The bright areas are nanoparticles of either silicon or carbon, surrounded with a silicon and carbon containing overlayer.*

not in the SEI. Combining the findings of the silicon dissolution in the SEI and the location of the charge storage inside the silicon, we conclude that the dissolution of silicon is a degradation mechanism.

On the cathode side[15] we currently focus on electrode materials that are state-of-the-art in battery technology. Here we aim to understand where the charge is stored inside the cathode material and explicitly if the lithium retains also part of the charge. We therefore compared the olivine $LiFePO_4$ and the layered $LiCoO_2$ by analyzing the electron energy loss spectra (EELS) of their components including that of Li. Figure 7a shows for $LiCoO_2$ a sharp Li-K-edge for the pristine and the discharged (lithiated) electrode at 61 eV, while the sharp edge vanishes in the charged state. The presence of a sharp Li-K-edge is an indication for a pure ionic state of lithium. Therefore we conclude that a lithium ion is intercalated into $LiCoO_2$ and that there is no charge transfer from the host phase towards the lithium. This reflects in fact the classical understanding of a lithium ion battery. However we could not determine this behaviour for $LiFePO_4$. Figure 7b shows the Li-K-edge of $LiFePO_4$ in different states of charge. There is one relatively sharp edge at 57 eV observable, but this is related to the Fe-M-edge, while the Li-K-edge is a broad shoulder from 60-70 eV. This indicates that lithium is not purely ionically bond, but unexpectedly part of the charge is transferred towards the lithium.

We conclude that the literature concepts on the mode of operation of Li"ion" batteries discriminating between intercalation and conversion systems is oversimplified and that predictions about theoretical capacities and electrode stabilities must be taken with great reservation.

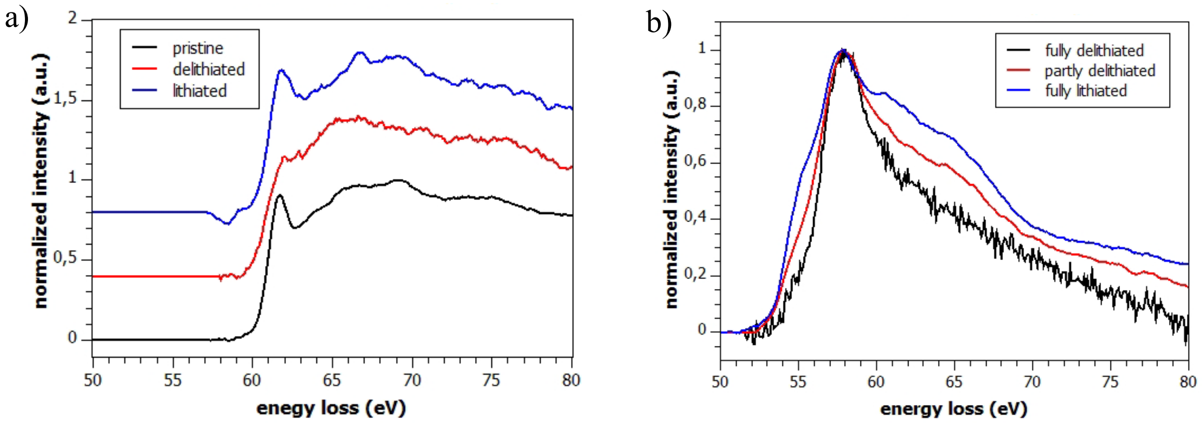


***Figure 7:** Lithium K-edge and metal M-edge EELS of a) $LiCoO_2$ and b) $LiFePO_4$ at different states of charge.*

5.4 Electrochemical stability of carbon materials

One fundamental problem in fuel cells and in water electrolysis systems is the inacceptable stability of carbon-supported electrodes. Attempts to compensate this by exceedingly high metal loadings are only mildly successful and are partly responsible for the prohibitive prize of such devices. In the absence of well-developed methodologies for synthesis and characterization of the carbon support[16] the literature concentrates on the metal part of the challenge and largely ignores the support. In combination with the activities at MPI CEC we focus on this important aspect motivated by our ability to synthesize carbon structures from molecular precursors. The electrochemical analysis of the corrosion behaviour was established at AC-FHI by a combination of electrochemical analysis with TEM, RAMAN and thermo-analytical methods.

We assume that the decomposition of carbon can be kinetically inhibited from its thermodynamic value of 0.2 V (RHE) when suitable structural modifications are applied. Chronoamperometric measurements on multiwall carbon nanotubes (MWCNT) are shown in Figure 8a. We identify four kinetically distinct regions. A large initial current abruptly falls because of combustion of the defective outer surface present in all carbon materials stored in air (region A). Subsequently, the current increases (region B), which we do not observe for low-surface area carbons. In this episode pitting corrosion enhances the reactive surface area as seen by TEM. The resolution between the two phases is hampered by transport phenomena for highly reactive samples. Having consumed the outer regions of the sample the corrosion occurs now on the bulk carbon exhibiting much less defects and thus reactive sites. This leads to a decrease of the current density (region C). In addition the formation of stable surface oxides begins passivating the carbon as a specialty of ambient temperature corrosion.

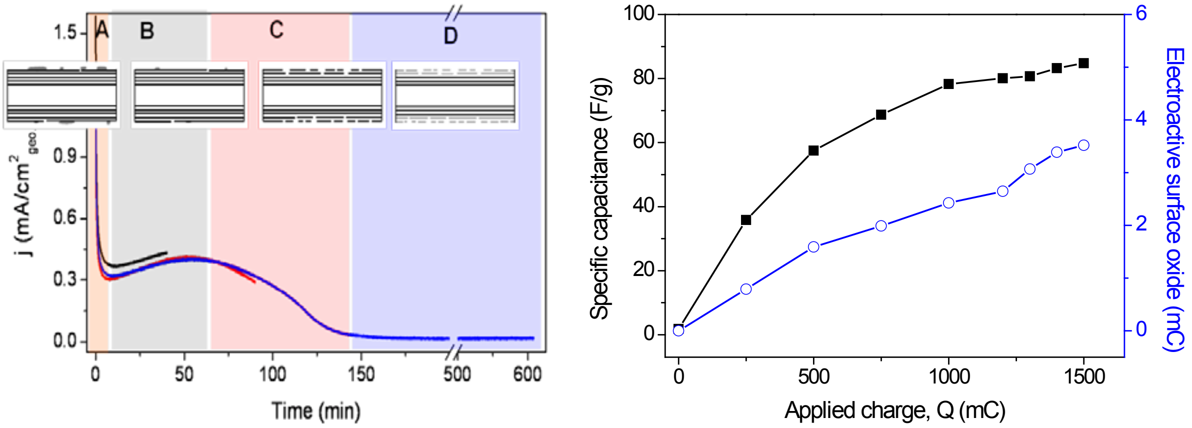


Figure 8: ***(a)*** *Chronoamperommogram on MWCNTs at 1.8 V vs. RHE in 0.5 M H_2SO_4 with a schematic diagram for the electrochemical degradation of MWCNTs in the inset.* ***(b)*** *capacitance and active surface oxide determined from cyclic voltammetry.*

In region D, the MWCNTs are covered with a passivating surface oxide, allowing for only small constant current densities. That the surface oxide is stable and prevents further corrosion of the MWCNTs is also indicated by the saturation of electrochemical capacitance (Fig. 8b) with passing higher amounts of charge through the electrode. Furthermore the oxygen concentration of the MWCNTs also saturates and the shape of the impedance spectra is constant with higher amount of passed charge. These finding have important consequences for the anchoring of active metal species that must not use, as conventionally done, the defective outer surface of the supporting carbon. We are now exploiting this by sub-surface anchoring procedure.

5.5 Electron spectroscopy

The group operated the beamline ISSIS at BESSY successfully[17] for in-house activities and for many external collaborations. The method receives substantial international attention[18] leading to a massive overbooking of our instrumental capacities. Measures to consolidate this situation are described in the general section. The activities mentioned in this report concern only projects originating in the AC department. In addition, the group designed the novel end station for EMIL allowing for high-energy NAP-XPS and built the instrument in collaboration between the FHI facilities and the SPECS company in Berlin. Attractive features are a new analyzer front lens with much improved acceptance angle and a modular sample environment allowing exchanging the sample environment for several applications such as gas phase catalysis, electrochemistry, low-temperature reactions and UHV model studies in such a way that operation can be resumed in less that a day after chamber exchange. Furthermore the group is heavily engaged in the scientific and technical development of the EMIL project joint between the HZB Berlin and the two MPI FHI and MPI-CEC. In addition a laboratory instrument is operated and currently upgraded to study electrodes from nearby performed electrochemical experiments with a transfer excluding air contact.

Silver in ethylene epoxidation

We study this reaction extensively as it serves as prototypical case for understanding the interplay between atomic oxygen species and chemical selectivity. The reason is the absence of oxide phases of the catalyst under working conditions despite the high chemical potential of oxygen. The uniqueness of Ag for this reaction is still an open question in the literature. To shed new light onto this traditional question, we use near ambient pressure X-ray photoelectron spectroscopy (NAP-XPS) to correlate the modifications at the Ag catalyst surface during reactions conditions with the catalytic performance[19] measured at the same time. We have also recently developed an innovative in situ cell in collaboration with the group of *L. Duda* from the Uppsala University to measure Resonant Inelastic X-ray Scattering (RIXS) on working Ag catalysts at ambient pressure, which allows the evaluation of possible role of bulk species at elevated pressures.

Based on our extensive analysis of O1s spectra and previous reaction models, we used the dynamics of a non-promoted catalyst to develop surface spectroscopic descriptors that captured trends in the catalytic performance. Changes in selectivity were correlated to modifications in the distribution of different forms of oxygen on the catalyst surface, namely electrophilic (Oelec) and nucleophilic species (Onucl). We further used this correlation to investigate to promotion effect of Cl on the epoxidation reaction. Figure 9 shows that the presence of Cl on the Ag surface, changes the distribution of oxygen species, which in turn renders a more selective catalyst. Hence, instead of the typical phenomenological correlation of catalytic performance with added amounts of promoters, our new results allowed a causal relationship involving the distribution of electrophilic and nucleophilic oxygen species, which are proposed active sites for the reaction.

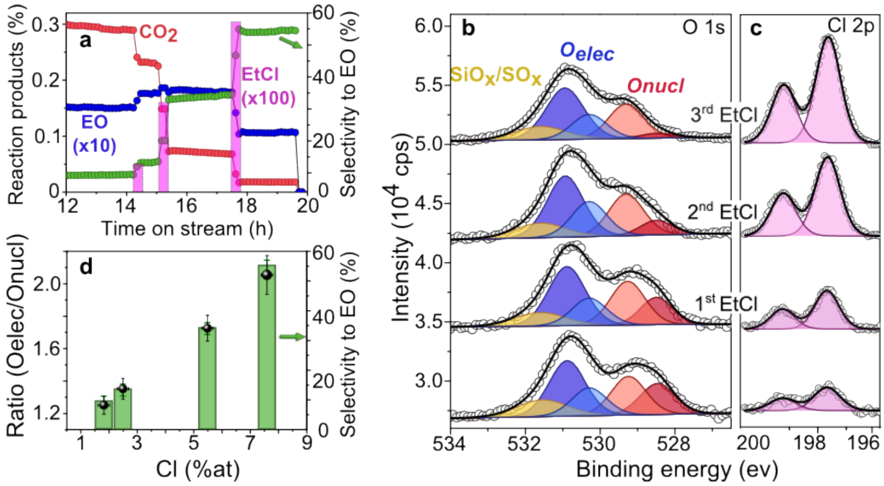


Figure 9: *In situ Cl promotion of Ag catalyst under ethylene epoxidation* ***(a)*** *Mole fraction of reaction products* CO_2 *(red) and EO (blue) together with selectivity to EO (green) as function of time. EtCl pulses are represented by the purple bars.* ***(b)*** *O1s XPS spectrum before (bottom) and after each Cl promotion step (*1^{st}*,* 2^{nd}*,* 3^{rd} *EtCl pulses).* ***(c)*** *Corresponding Cl 2p spectra before and after each promotion step.* ***(d)*** *Ratio of electrophilic to nucleophilic oxygen species Olelec/Onucl (black dots) and selectivity (green bars) as function of the Cl atomic concentration.*

Complementary to the X-ray studies, we have performed DFT calculations in collaboration with Simone Piccinin from SISSA/Trieste to improve the assignments and interpretation of the spectroscopic fingerprints and correlations. Core-level energy shifts, electronic structure and surface energies were calculated for a several Ag-O structures and compared with reference measurements on different single crystal surfaces.

Active oxygen species on metal and metal-alloy surfaces

We are investigating the partial oxidation behaviour of several metallic and bimetallic systems, including Au, Cu, Re, AgCu, and AgRe. With these systems, we can generate a full spectrum of oxygen surface species. By examining the influence of various oxygen species on catalytic performance, and comparing with the well-known silver system, we gain insight into the fundamental factors that influence catalytic performance.

Gold as an oxidation catalyst

Gold is very inactive as an extended bulk material, but is known to exhibit activity when used in the form of nanoparticles. It is very difficult for any kind of oxygen species to exist on the surface of gold. We investigated ways of inducing the formation of active oxygen species on gold using highly oxidizing atmospheres containing O_3. We induced transient oxidation of god surfaces including significant persistent restructuring. Pure gold either in the form of nanstructures or a s massive surface in the absence of an oxide support is unable to sustain catalytic oxidation of CO as heterogeneous process. This clearly proves that it is not gold alone acting as catalyst but a synergy between nano-gold and some support is essential to induce chemical reactivity.

Copper in partial oxidation of ethylene

Copper is easily oxidized, and forms thick layers of mixed oxides under partial-oxidation conditions. We have found that CuO is catalytically active for partial oxidation, while Cu_2O is active for total oxidation. Ethylene partial oxidation occurs through reaction of chemisorbed ethylene with surface oxygen species from the oxide. These species are replenished in a consecutive reaction involving activation of gas phase oxygen through reduced oxide sites after desorption of ethylene oxide. The successful catalyst consists of a sub-stoichiometric oxide—i.e. a mixture of Cu^{2+} and Cu^{1+} species. The catalytic selectivity can be tuned by tuning the oxide stoichiometry; the absolute values for ethylene epoxide were low (ca. 2.5%) under the low-pressure conditions of the NAP XPS experiment.

Rhenium in partial oxidation of ethylene

Rhenium is metal that, when alloyed with silver, provides selectivity enhancements for ethylene epoxidation. However, rhenium is different from copper in that, under epoxidation conditions, bulk rhenium oxides are not stable. Rather, under these conditions, a unique surface oxide ('ReO') is formed. This oxide does not exist as a stable bulk phase, but is found only on the rhenium metal surface. It is believed that the redox couple of the surface oxide with the silver bulk is involved in the epoxidation process by enhancing the presence of electrophilic oxygen.

Graphene Growth on Transition Metal Surfaces

Within the frame of the Grafol project the electrical, mechanical, surface and thermal properties of graphene were thoroughly investigated during and after growth by chemical vapour deposition (CVD) of different hydrocarbons and by carbon diffusion processes on various transition metal (TM) surfaces. Complementary analysis techniques such as NAP-XPS, X-ray Absorption Near Edge Structure (XANES), scanning tunneling microscopy (STM) and (environmental) scanning electron microscopy ((E)SEM) reveal that the major key to large scale graphene growth of sufficient quality is a suitable solubility of carbon sub-surface of the working catalyst[20]. This quality, which is dependent on the respective TM and in strong relation with the catalyst temperature, controls the number of carbon atoms available for graphene growth on the surface. For high (Fe) and low solubility (Cu) systems this leads to virtually immediate growth of graphene via a single reaction path. However, for intermediate solubilities, e.g. on Ni(111), several, partially superimposing reaction paths were observed. While graphene of rather low quality can be grown dominantly via a surface carbide at low temperatures (< 450°C) a higher quality can be achieved above $T = 450°C$ for which more carbon atoms can diffuse into the Ni bulk. Low temperature growth on Ni(111) leads to epitaxial growth with a strong interaction between Ni and graphene which is not even affected by subsequent exposure to air, whereas at high temperatures rotated, quasi free standing graphene is observed. A similar situation is found for Cu on which during CVD strongly bound graphene is formed. Quasi-free standing graphene results from exposure to air via intercalation of oxygen.

Noble metal free HCl oxidation over ceria-based catalysts

The heterogeneously catalyzed gas-phase oxidation of HCl to Cl_2 (Deacon reaction) is an energy-efficient and sustainable route to recycle chlorine from HCl-containing industrial waste streams. CeO_2 catalysts were identified[21] as an attractive alternative to RuO_2 because of their middle-high reactivity, good stability as well as their lower cost. X-ray diffraction and electron microscopy of CeO_2 samples exposed to reaction feeds with different O_2:HCl ratios provide evidence that CeO_2 does not suffer from bulk chlorination in O_2-rich feeds (O_2:HCl ≥ 0.75), while it does form chlorinated phases in stoichiometric or sub-stoichiometric feeds (O_2:HCl ≤ 0.25). The quantitative analysis of the chlorine uptake by X-ray photoelectron spectroscopy[22] indicates that chlorination under O_2-rich conditions is confined to the surface and possibly one subsurface layer of CeO_2 particles.

Micro-calorimetric experiments with CO_2 adsorption suggest that the basic character of CeO_2 has been eliminated upon reaction in HCl oxidation, indicating that most of the basic lattice O sites are exchanged by chlorine and that the OH groups formed are rather acidic. Thus, HCl adsorption is retarded by the loss of basic (O^{2-}) sites required for H abstraction during dissociative HCl adsorption. Furthermore, FTIR and TPD adsorption experiments using NH_3 and CO as probe molecules reveal that the density and the strength of surface acidic functions increased significantly upon reaction. EPR experiments on fresh and post-reaction samples using O_2 as probe molecule strongly suggest that oxygen activation is inhibited by the high degree of surface chlorination. DFT simulations reveal that Cl activation from O vacancy positions to surface Ce atoms is the most energy-demanding step, though Cl-O competition may render re-oxidation as the rate-determining step. The coverage of the most abundant surface intermediates, OH and Cl, was monitored under various reaction conditions by in situ FTIR and by in situ Prompt Gamma Activation Analysis. The results indicate that only a small fraction of surface sites is actively involved in the reaction, and most of the surface species probed are spectators. The role of trivalent (La, Sm, Gd, and Y) and tetravalent (Hf, Zr, and Ti) dopants in the catalytic, structural, and electronic properties of ceria was also investigated.

5.6 Electron microscopy

The group concentrated on methodical improvements of the TEM studies conducted in the context of many projects in the department. The image simulation activities were substantially intensified requiring better control of the image acquisition conditions as well as the sample preparation efforts. The improvements achieved can be seen in the respective project reports. Furthermore the group engaged heavily in the identification and experimental testing of novel developments in electron microscopy techniques. It was discovered that our TITAN instrument had a substantial malfunction of the corrector system resulting in the necessity to exchange the corrector unit. In addition the group set out to implement methods for observing catalysis-induced structural modifications. We chose the way to subject samples to practical operation conditions and study them by “identical location” TEM after processing steps and multiple transfer between the TEM and a dedicated reactor system. Alternative approaches using MEMS-based micro-reactors on a TEM specimen holder are currently developed for several applications. A completely new sample holder capable of studying a battery in-situ under potential and temperature control was developed and implemented.

A second line of development concerned the mesoscale observation of catalysts under realistic operation conditions that were often chosen such to exactly represent the condition at the ISSIS experiment at BESSY in order to compare morphology and spectroscopy of the same system under the same conditions. In order to improve the experimental set-up for in-situ experiments in the ESEM, the vacuum system was upgraded to an oil-free pumping system. Furthermore, the instrument was equipped with a mass spectrometer for the analysis of gas species in the chamber and detection of reaction atmosphere and products. For high stability and precise controllable heating experiments we designed and realized a laser heating stage which allows heating up to 1000°C. In contrast to the commercial heating stage, only the sample and a small crucible consisting of inert material are heated. Laser heating enables ultra-stable direct heating with low-drift and without undesired side-reactions on hot walls of a conventional heating system.

Carbon/Graphene growth was enabled through metal catalyzed chemical vapour deposition using ethylene, methane or benzene as hydrocarbon source (Figure 10). In the case of growth on copper substrates, the whole CVD cycle, including substrate annealing, CVD growth and subsequent cooling was studied in detail. The experiments reveal substantial surface dynamics during annealing as well as the segregation of contaminations from the bulk to the surface (mainly SiO_2). It was found that graphene growth in low pressure CVD at temperatures close to 1000°C proceeds on a surface pre-melted, highly mobile Cu surface. Growth happens after an incubation time, during which the Cu surface is enriched with carbon atoms and clusters (growth species) until super saturation is reached and nucleation starts. During growth, growth species are consumed and the growth speed decreases. With increasing Cu coverage, the formation of growth species by catalytic decomposition of the hydrocarbon feed is reduced and limits the growth speed. Finally, growth stops once a balance is reached between growth species formation, recombination and desorption from the surface and hydrogen induced graphene etching. After growth, when the sample is cooled down, a particular reconstruction of copper under the influence of covering graphene can be observed. The observed reconstruction is related to the chemical inertness of graphene and the action of small amounts of oxygen. In-situ investigations in the ESEM revealed the effect of temperature, grain orientation, copper surface mobility and sublimation on the nucleation behaviour, growth dynamics and the shape of the formed graphene.

Studies on nickel substrates show characteristic differences due to the high solubility of carbon in nickel. Carbon dissolution and segregation control the carbon formation. A reversible hydrogen-induced surface segregation of bulk-dissolved carbon was observed by ESEM. The findings are in perfect agreement with the results of in-situ XPS studies obtained by the AG “Electron Spectroscopy” at BESSY. Since both techniques operate at the same pressures conditions and the ESEM experiments can be performed over a large range of spatial resolutions, the two complementary instruments work in perfect synergy, for a combination of visual information about surface dynamics with spectroscopic data.

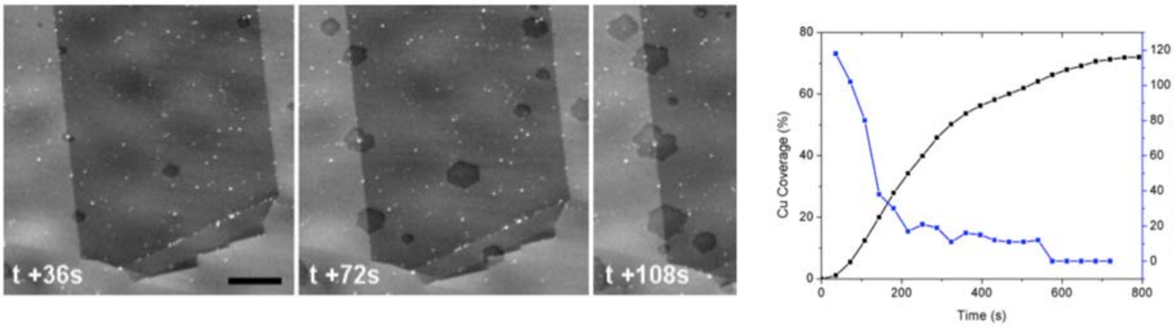


Figure 10: *Initial stages of graphene growth on a Cu foil at 1000°C. Images were recorded with a scanning speed of one frame per 36s. The scale bars measure 5µm. Quantitative analysis allows deriving details about nucleation and growth kinetics by direct observation.*

A high pressure TEM Grid (HITEM) micro reactor was realized in a flow-through geometry. Using this reactor we investigated the methanol synthesis over Cu/ ZnO/Al_2O_3 catalysts at 30 bars and 230 °C. Due to the high sensitivity of the PTR-MS detector, the amount of catalyst on the TEM grid can be varied and optimized for best imaging conditions and sample handling. Although the amount of catalyst is minuscule (ca. 1 microgram), the evolution of CO_2 and Methanol was quantified.

Our investigations were focused on the formation of a thin ZnO_x layer on the surface of Cu particles during reduction of the oxide precursor. Recent research on model systems showed that the strong interaction between the ZnO layer and the support metal causes the formation of a metastable depolarized graphitic ZnO polymorph. However, these findings were obtained on model systems that are far away from industrial relevant catalytic systems in terms of composition, structure, and activity. We now identified this metastable graphitic ZnO overlayer on highly active industrial relevant Cu/ZnO/Al2O3 catalysts for the methanol synthesis. Bright field TEM (Figure 11A) reveals a slightly distorted but generally layered structure that is covering the Cu particles. Electron energy loss spectra recorded from exposed Cu particles (Figure 11B) show the presence of reduced Cu species as well as the presence of Zn. Verification for the existence of a ZnO overlayer was obtained from energy filtered TEM (EFTEM) elemental maps using the O-K and Cu-L edges (Figure 11 C). Unambiguously, the EFTEM maps reveal the presence of oxygen in the shell covering the copper particles. Since copper was detected in the reduced state by EELS and the presence of Zn was confirmed, it can safely be concluded that the overlayer consists of ZnO in its metastable graphitic polymorph.

5.7 Theory support group

In December 2011 the Theory Support team, headed by *K. Hermann* with up to three postdoctoral researchers started to work in the Inorganic Chemistry Department. The activities of this team have focused mainly on applications of density-functional theory (DFT), on theoretical electron and photon spectroscopy (near-edge X-ray, NEXAFS and infrared, IR), and on bulk as well as surface crystallography. The results of collaborations with all groups of the department have proven to be valuable asset. Here we mention support for the Reactivity group (*A. Trunschke*) in the interpretation of structure-reactivity relationships of small supported transition metal (MoO_x, TiO_x) oxide particles[12a, 23] of catalytic interest. Further, we have contributed to theoretical studies on the ODH of hydrocarbons at spin-active carbon catalysts[24]. With the Electronic Structure group (*A.*

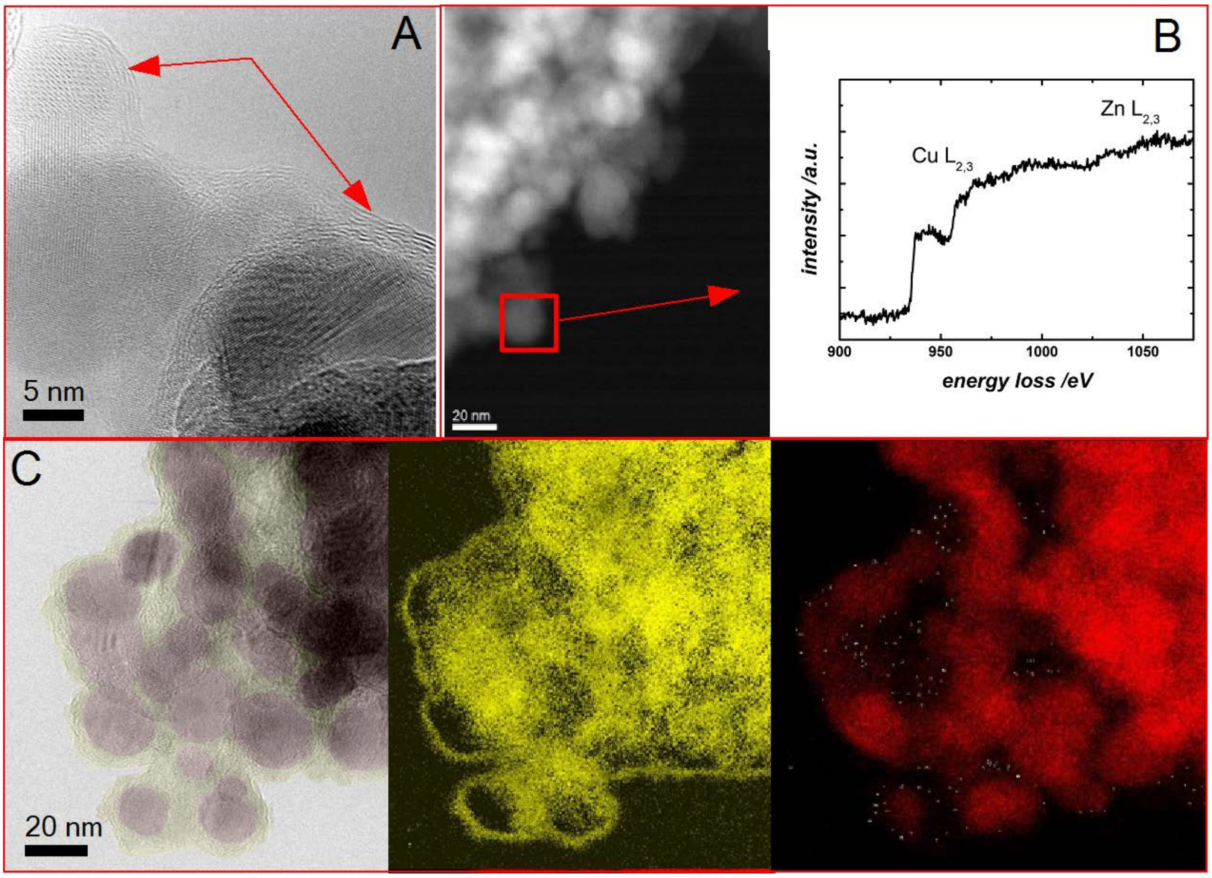


Figure 11: ***(A)*** *Brightfield TEM image showing the surface decoration of Cu particles with a layered structure.* ***(B)*** *representative STEM image of reduced Cu/ZnO/Al2O3 catalysts. The EELS spectrum was recorded from the indicated region. EFTEM maps created using the O-K and Cu-L edges are shown in* ***(C)****.*

Knop-Gericke) we worked on the interpretation of core excitation spectra (XAS) of small molecules adsorbed at metal surfaces. The experimental results were measured by FHI and evaluated in periodic DFT calculations by *Simone Piccinin* et al. (SISSA/Trieste). With the Electron Microscopy group (*M. Willinger*) we discussed crystallographic shapes of small metal particles (Pd) formed on carbon substrates as well as of graphene-induced faceting of copper surfaces. In addition, the Theory Support team studied various subjects not immediately connected with experimental work in the department but also in collaboration with external groups. This includes work on dynamics of molecular machines (nanorotors) with *M.A. Van Hove* et al., Hong Kong Baptist University and City University Hong Kong.

5.8 Collaboration with the CP department

The AC department engaged in several collaborative projects with the CP department providing essential physico-chemical insights that the AC department was able to translate to performance catalysts. This approach is working in a generic mode and was thus carried forward in two distinct project requiring extensive collaboration and efforts on both sides.

Among the various inorganic materials that have been evaluated as heterogeneous catalysts for the oxidative coupling of methane[25] (OCM), alkaline earth oxides doped with alkali elements or transition metal ions have received particular attention. In a recent study, Nilius, Freund, and co-workers et al. provide evidence that strongly bound O^{2-} species, precursors of dissociatively adsorbed oxygen, are formed on ordered CaO films doped with Mo2+ ions. The results indicate that molecular activation on doped oxides does not require any surface structural defects. Accordingly, it is suggested that the activation of methane on smooth surfaces of transition- metal-doped wide-band-gap oxides may involve such activated oxygen species. We put the concept to test (Fig. 12) with powder catalysts[26] working at high

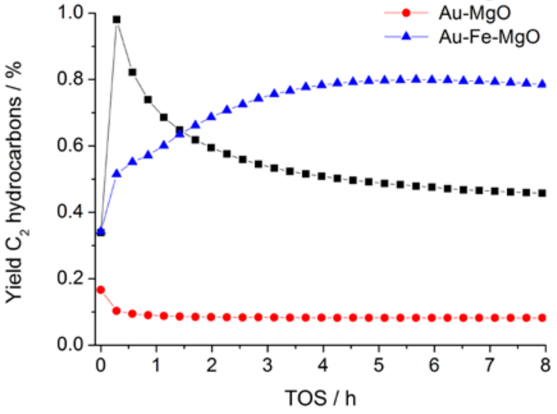


Figure 12: *Yield of ethane and ethene in the oxidative coupling of methane over MgO and doped MgO as a function of time on stream (TOS) at T=1023 K; W/F= 0.0167 g·s·ml-1;* $CH_4/O_2/N_2=3/1/1$.

temperature. Doped MgO was prepared by introduction of Fe and Au in ppm quantities. Gold seems to block the active sites on the surface of MgO since Au-MgO shows only negligible activity.

This is ascribed to the propensity of gold to adsorb at step edges of the MgO surface and thus cover the active sites related to edges as shown by electron microscopy. Fe-MgO is more active and selective resulting in higher yield. Further increase in methane conversion and selectivity is achieved by co-doping with Fe and Au. Two types of gold were evidenced based on the catalytic results: In single doped Au-MgO only the step-decorating poisoning effect was found. Through the presence of subsurface iron species in the co-doped system the edge-decorating effect was massively overruled by the beneficial effect of creating structurally stable novel active sites at terraces of the MgO without introducing vacancies that may destabilize the system at longer time on stream. The second gold species apparently arises from a significant gold–support interaction stemming from the strain in the MgO due to iron doping. Investigation of the bulk structure by HAADF STEM and TEM reveals the presence of local strain. The possible effects of isotropic unstrained gold particles are negligible due to their very low abundance as a consequence of the synthesis strategy. Electronic doping of MgO terraces is also achieved through iron dissolution only, but with significant lower effectiveness. It is tempting to conclude that the co-doping creates highly active sites for oxygen activation to give a peroxy species, whereas iron doping ends up with sites creating oxo species that are less active in creating methyl radicals for OCM.

We investigated the strong metal support interaction on a model system consisting of Pt nanoparticles on the surface of a thin Fe_3O_4 film, which was grown on a Pt (111) substrate (Figure 13). Annealing at 850K in vacuum results in the encapsulation of the Pt particles by a monolayer of FeO. A combination of high-resolution microscopic techniques was used to study the nature of the interfaces between Pt and the oxide film and to observe the surface termination of the Pt particles in cross-section. It was found that the interfaces between the iron oxide film and the Pt support on the one side and the Pt particles on the other, is characterized by a direct Pt-Fe contact. While a Kagome layer of Fe_3O_4 is exposed to the Pt (111) of the deposited particles, the interface to the Pt substrate corresponds to a (111) layer of iron in FeO (Figure 13A). The nature of the interfaces was confirmed by quantitative simulation of the STEM image contrast (see insets in Figure 5A). Additional confirmation is provided by the observed lattice mismatch, causing periodic miss-alignments between the Pt and Fe atomic columns (indicated by arrows in Figure 5A). In agreement with STM, the

HRTEM investigation revealed the presence of an atomic layer with different lattice spacing on the surface of the Pt particles (Figure 13B). The final proof for the encapsulation of the particles with a film of iron oxide is shown in Figure 14. Firstly, contrast variations in annular bright field (phase-contrast) and HAADF (Z- and thickness contrast) images of the Fe_2O_3 film indicate a loss of structural order and material in the vicinity of the supported particles. Secondly, EELS line-scans recorded across the film and supported particles show intensity due to oxygen and iron on the Pt particles. Although signal is collected from only a few atoms, the oxygen K- and iron L edges can be detected, hence demonstrating the potential of chemical electron microscopy.

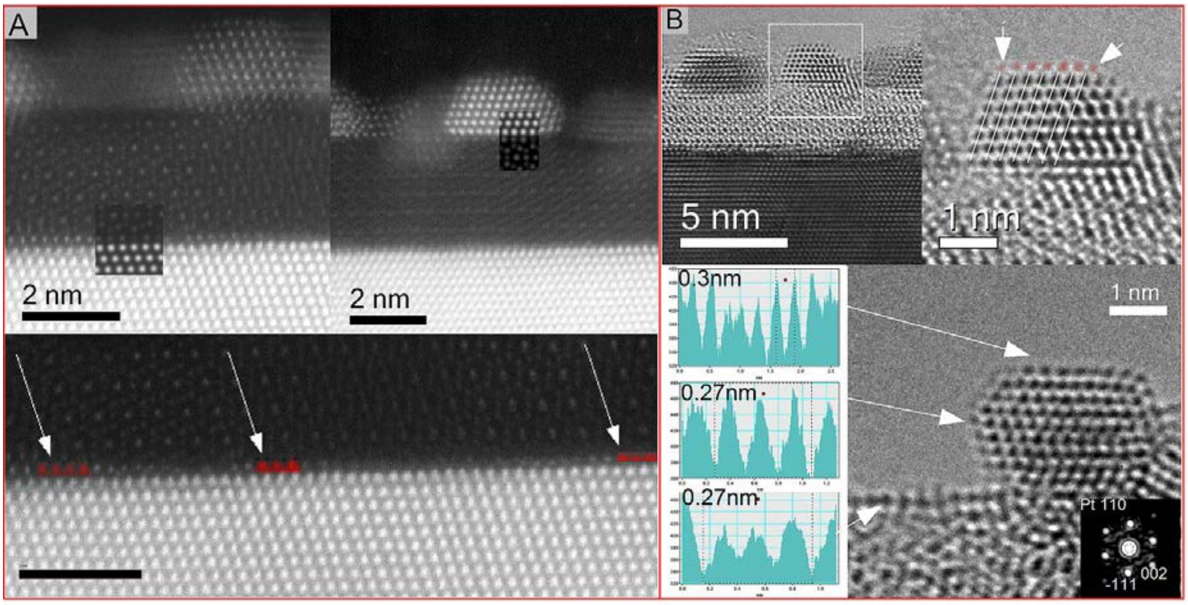


Figure 13: *(A) HAADF images of the interfaces with simulations as insets. Arrows and colored Fe atoms indicate areas where the iron columns are in line with the Pt columns.* ***(B)*** *HRTEM images reveal that the surface atomic layer shows larger lattice spacings than expected for Pt.*

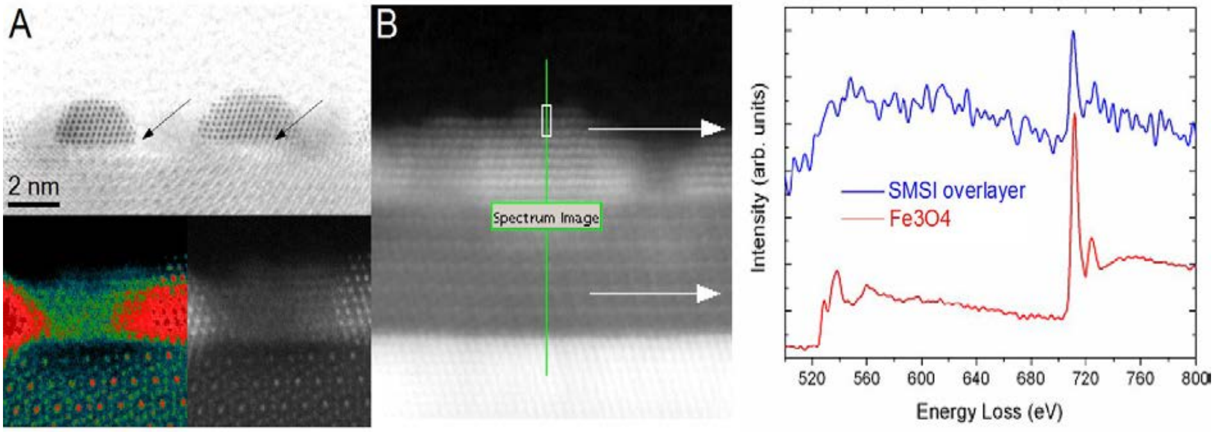


Figure 14: *(A) Annular bright field and HAADF STEM images reveal variations in the contras of the Fe_3O_4 layer in the vicinity of Pt particles, indicating a loss of iron due to formation of an overgrowth layer. (B) EELS line-scan demonstrates that the surface of the Pt particles contains iron and oxygen atoms.*

6. Future developments

The department AC is expected to consolidate the modifications induced by the foundation of the MPI CEC in the coming years. We will concentrate on our core projects, as there are C1 chemistry, selective oxidation and metal-oxygen interactions in coin group metals. The application focus on energy–related reactions will be maintained and with this also the project on functional aspects of Li"ion" batteries. Common to all projects is the guidance through the general concept of dynamical interfacial processes as briefly described in section 4 of this report. We expect to find a way to translate this concept into a synthesis strategy of novel systems in which the surface dynamics is controlled by the bulk synthesis in a rational way and not the consequence of unintentional effects occurring when the catalyst is brought in contact with chemical potential of the reactants.

We will be busy completing our large-scale instrument development projects in electron microscopy and in chemical applications of electron spectroscopy. In the microscopy project several methodical developments concerning chemical applications of TEM for chemistry at large will be of relevance for a larger user community and will thus require extensive and careful testing with a large number of case studies. In this context and also for the best use of the MEMS devices as sample holders it would be of enormous help if we could acquire a FIB instrument for target preparation in combination with a low-energy Ar ion-milling device. This would allow target preparations from catalysts making them more amenable for high resolution analytical and imaging work and thus greatly enhance the applicability of electron microscopy for the samples of relevance at the AC department.

The collaborative activities will concentrate on the cooperation with the MPI-CEC and on intra-FHI activities. Within the MPG the re-erected collaborative network MAXNET ENERGY will strengthen the material science and electrochemical aspects of our work. The work in BASCAT, a project with BASF on electro-catalysts and a project with VW (Li-Si electrodes) will be our principal industry collaborations. With HZB Berlin we intend to operate the Joint Laboratory "Chemical applications of electron spectroscopy".

References

[1] M. Eichelbaum, M. Hävecker, C. Heine, A. Karpov, C.-K. Dobner, F. Rosowski, A. Trunschke, R. Schlögl, *Angew. Chem. Int. Ed.* **2012**, *51*, 6246-6250.
[2] O. Shekhah, W. Ranke, A. Schüle, G. Kolios, R. Schlögl, *Angew. Chem. Int. Ed.* **2003**, *42*, 5760-5763.

[3] B. S. Klose, F. C. Jentoft, R. Schlögl, I. R. Subbotina, V. B. Kazansky, *Langmuir* **2006**, *21*, 10564-10572.

[4] aM. Salmeron, R. Schlögl, *Surface Science Reports* **2008**, *63*, 169-199; bR. Schlögl, *Advances in Catalysis* **2008**, *52*, 273-338; cA. Knop-Gericke, E. Kleimenov, M. Hävecker, R. Blume, D. Teschner, S. Zafeiratos, R. Schlögl, V. I. Bukhtiyarov, V. V. Kaichev, I. P. Prosvirin, A. I. Nizovskii, H. Bluhm, A. Barinov, P. Dudin, M. Kiskinova, *Advances in Catalysis* **2009**, *52*, 213-272; dS. Wrabetz, X. Yang, G. Tzolova-Müller, R. Schlögl, F. C. Jentoft, *J. Catal.* **2010**, *269*, 351-358; eV. B. Kazansky, I. R. Subbotina, F. C. Jentoft, R. Schlögl, *J. Phys. Chem. B* **2006**, *110*, 17468-17477.

[5] M. Behrens, F. Studt, I. Kasatkin, S. Kühl, M. Hävecker, F. Abild-Pedersen, S. Zander, F. Girgsdies, P. Kurr, B.-L. Kniep, M. Tovar, R. W. Fischer, J. K. Nørskov, R. Schlögl, *Science* **2012**, *336*, 893-897.

[6] T. Kandemir, F. Girgsdies, T. C. Hansen, K.-D. Liss, I. Kasatkin, E. L. Kunkes, G. Wowsnick, N. Jacobsen, R. Schlögl, M. Behrens, *Angew. Chem. Int. Ed.* **2013**, *52*, 5166-5170.

[7] M. Behrens, S. Zander, P. Kurr, N. Jacobsen, J. Senker, G. Koch, T. Ressler, R. W. Fischer, R. Schlögl, *J. Am. Chem. Soc.* **2013**, *135*, 6061-6068.

[8] aM. Eichelbaum, R. Glaum, M. Hävecker, K. Wittich, C. Heine, H. Schwarz, C.-K. Dobner, C. Welker-Niewoudt, A. Trunschke, R. Schlögl, *ChemCatChem* **2013**, *5*, 2318-2329; bK. Amakawa, Y. V. Kolen'ko, A. Villa, M. E. Schuster, L.-I. Csepei, G. Weinberg, S. Wrabetz, R. Naumann d'Alnoncourt, F. Girgsdies, L. Prati, R. Schlögl, A. Trunschke, *ACS Catal.* **2013**, *3*, 1103-1113.

[9] R. Naumann d'Alnoncourt, Y. V. Kolen'ko, R. Schlögl, A. Trunschke, *Combinatorial Chemistry & High Throughput Screening* **2012**, *15*, 161-169.

[10] aM. Sanchez Sanchez, F. Girgsdies, M. Jastak, P. Kube, R. Schlögl, A. Trunschke, *Angew. Chem. Int. Ed.* **2012**, *51*, 7194-7197; bY. V. Kolen'ko, K. Amakawa, R. Naumann d'Alnoncourt, F. Girgsdies, G. Weinberg, R. Schlögl, A. Trunschke, *ChemCatChem* **2012**, *4*, 495-503.

[11] W. Zhang, A. Trunschke, R. Schlögl, D. S. Su, *Angew. Chem. Int. Ed.* **2010**, *49*, 6084-6089.

[12] aC. Guo, K. Hermann, M. Hävecker, A. Trunschke, R. Schlögl, *J. Phys. Chem.* **2012**, *116*, 22449-22457; bB. Beck, M. Harth, N. G. Hamilton, C. Carrero, J. J. Uhlrich, A. Trunschke, S. Shaikhutdinov, H. Schubert, H.-J. Freund, R. Schlögl, J. Sauer, R. Schomäcker, *J. Catal.* **2012**, *296*, 120-131.

[13] aB. Frank, K. Friedel, F. Girgsdies, X. Huang, R. Schlögl, A. Trunschke, *ChemCatChem* **2013**, *5*, 2296-2305; bT. Cotter, B. Frank, W. Zhang, R. Schlögl, A. Trunschke, *Chem. Mater.* **2013**, *25*, 3124-3136.

[14] M. Hävecker, S. Wrabetz, J. Kröhnert, R. Naumann d'Alnoncourt, Y. V. Kolen'ko, F. Girgsdies, R. Schlögl, A. Trunschke, *J. Catal.* **2012**, *285*, 48-60.

[15] J. Popovic, R. Demir-Cakan, J. Tornow, M. Morcrette, D. S. Su, R. Schlögl, M. Antonietti, M.-M. Titirici, *Small* **2011**, *7*, 1127-1135.

[16] aD. S. Su, J. Zhang, A. Thomas, X. Wang, J. Paraknowitsch, R. Schlögl, *ChemSusChem* **2010**, *3*, 169-180; bD. S. Su, R. Schlögl, *ChemSusChem* **2010**, *3*, 136-168.

[17] R. Arrigo, M. Hävecker, M. E. Schuster, C. Ranjan, E. Stotz, A. Knop-Gericke, R. Schlögl, *Angew. Chem. Int. Ed.* **2013**, *Early View*.

[18] aV. Papaefthimiou, M. Shishkin, D. K. Niakolas, M. Athanasiou, Y. T. Law, R. Arrigo, D. Teschner, M. Hävecker, A. Knop-Gericke, R. Schlögl, T. Ziegler, S. G. Neophytides, S. Zafeiratos, *Adv. Energy Mater.* **2013**, *3*, 762-769; bB. Bayer, C.

Castellarin-Cudia, R. Blume, S. A. Steiner III, C. Ducati, D. Chu, A. Goldoni, A. Knop-Gericke, R. Schlögl, C. Cepek, J. Robertson, S. Hofmann, *RSC Adv.* **2013**, *3*, 4086-4092; cC. Rameshan, S. Stadlmayr, S. Penner, H. Lorenz, N. Memmel, M. Hävecker, R. Blume, D. Teschner, T. Rocha, D. Zemlyanov, A. Knop-Gericke, R. Schlögl, B. Klötzer, *Angew. Chem. Int. Ed.* **2012**, *51*, 3002-3006; dV. Papaefthimiou, T. Dintzer, V. Dupuis, A. Tamion, F. Tournus, A. Hillion, D. Teschner, M. Hävecker, A. Knop-Gericke, R. Schlögl, S. Zafeiratos, *ACS Nano* **2011**, *5*, 2182 - 2190

[19] T. C. R. Rocha, A. Oesterreich, D. V. Demidov, M. Hävecker, S. Zafeiratos, G. Weinberg, V. I. Bukhtiyarov, A. Knop-Gericke, R. Schlögl, *Phys. Chem. Chem. Phys.* **2012**, *14*, 4554-4564.

[20] aR. Weatherup, B. C. Bayer, R. Blume, C. Baehtz, P. R. Kidambi, M. Fouquet, C. T. Wirth, R. Schlögl, S. Hofmann, *ChemPhysChem* **2012**, *13*, 2544-2549; bR. Weatherup, B. C. Bayer, R. Blume, C. Ducati, C. Bähtz, R. Schlögl, S. Hofmann, *Nano Letters* **2011**, *11*, 4154-4160.

[21] D. Teschner, R. Farra, L. Yao, R. Schlögl, H. Soerijanto, R. Schomaeker, T. Schmidt, L. Szentmiklósi, A. Amrute, C. Mondelli, J. Pérez-Ramirez, G. Novell-Leruth, N. López, *J. Catal.* **2012**, *285*, 273-284.

[22] D. Teschner, G. Novell-Leruth, R. Farra, A. Knop-Gericke, R. Schlögl, L. Szentimiklósi, M. G. Hevia, H. Soerijanto, R. Schomäcker, J. Pérez-Ramírez, N. López, *Nature Chemistry* **2012**, *4*, 739-745.

[23] C. Guo, K. Hermann, M. Hävecker, J. Thielemann, P. Kube, L. Gregoriades, A. Trunschke, J. Sauer, R. Schlögl, *J. Phys. Chem. C* **2011**, *115*, 15449-15458.

[24] O. Khavryuchenko, B. Frank, A. Trunschke, K. Hermann, R. Schlögl, *J. Phys. Chem. C* **2013**, *117*, 6225-6234.

[25] aS. Arndt, G. Laugel, S. Levchenko, R. Horn, M. Baerns, M. Scheffler, R. Schögl, R. Schomäcker, *Catalysis Reviews: Science and Engineering* **2011**, *53*, 424-514; bU. Zavyalova, G. Weinberg, W. Frandsen, F. Girgsdies, T. Risse, K.-P. Dinse, R. Schlögl, R. Horn, *ChemCatChem* **2011**, *3*, 1779-1788; cU. Zavyalova, M. Geske, R. Horn, G. Weinberg, W. Frandsen, M. Schuster, R. Schlögl, *ChemCatChem* **2011**, *3*, 949-959; dP. Myrach, N. Nilius, S. V. Levchenko, A. Gonchar, T. Risse, K.-P. Dinse, L. A. Boatner, W. Frandsen, R. Horn, H.-J. Freund, R. Schlögl, M. Scheffler, *ChemCatChem* **2010**, *2*, 854-862.

[26] P. Schwach, M. G. Willinger, A. Trunschke, R. Schlögl, *Angew. Chem. Int. Ed.* **2013**, *52*, 11381-11384.

Publications of the Department of Inorganic Chemistry

2011 (late publications)

Arndt, S., G. Laugel, S.V. Levchenko, R. Horn, M. Baerns, M. Scheffler, R. Schlögl and R. Schomäcker: A Critical Assessment of Li/MgO-Based Catalysts for the Oxidative Coupling of Methane. Catalysis Reviews: Science and Engineering **53** (4), 424-514 (2011).

Becker, M.J., W. Xia, J.-P. Tessonnier, R. Blume, L. Yao, R. Schlögl and M. Muhler: Optimizing the synthesis of cobalt-based catalysts for the selective growth of multiwalled carbon nanotubes under industrially relevant conditions. Carbon **49** (15), 5253-5264 (2011).

Bielz, T., H. Lorenz, P. Amann, B. Klötzer and S. Penner: Water-Gas Shift and Formaldehyde Reforming Activity Determined by Defect Chemistry of Polycrystalline In_2O_3. The Journal of Physical Chemistry C **115** (14), 6622-6628 (2011).

Blume, R., M. Hävecker, S. Zafeiratos, D. Teschner, A. Knop-Gericke, R. Schlögl, L. Gregoratti, A. Barinov and M. Kiskinova: Ruthenium Active Catalytic States: Oxidation States and Methanol Oxidation Reactions. In: Nanostructured Catalysts: Selective Oxidations, Vol. 19. (Eds.) C. Hess and R. Schlögl. The Royal Chemical Society, Cambridge, UK, 248-265 (2011). ISBN 978-0-85404-186-2.

Boualleg, M., S. Norsic, D. Baudouin, R. Sayah, E.A. Quadrelli, J.M. Basset, J.-P. Candy, P. Delichere, K. Pelzer, L. Veyre and C. Thieulieux: Selective and regular localization of accessible Pt nanoparticles inside the walls of an ordered silica: Application as a highly active and well-defined heterogeneous catalyst for propene and styrene hydrogenation reactions. Journal of Catalysis **284** (2), 184-193 (2011).

Chlosta, R., G. Tzolova-Müller, R. Schlögl and C. Hess: Nature of dispersed vanadium oxide: influence of the silica support structure and synthesis methods. Catalysis Science & Technology **1** (7), 1175-1181 (2011).

Csepei, L.-I. and C. Bolla: The effect of salicylic acid on the Briggs-Rauscher oscillating reaction. Studia Universitatis Babes-Bolyai Chemia **56** (1), 285-300 (2011).

Demidov, D.V., I.P. Prosvirin, A.M. Sorokin, T. Rocha, A. Knop-Gericke and V.I. Bukhtiyarov: Preparation of Ag/HOPG model catalysts with a variable particle size and an in situ xps study of their catalytic properties in ethylene oxidation. Kinetics and Catalysis **52** (6), 855-861 (2011).

Farra, R., K. Thiel, A. Winter, T. Klamroth, A. Pöppl, A. Kelling, U. Schilde, A. Taubert and P. Strauch: Tetrahalidocuprates(II)-structure and EPR spectroscopy. Part 1: Tetrabromidocuprates(II). New Journal of Chemistry **35** (12), 2793-2803 (2011).

Favry, E., D. Wang, D. Fatauzzi, J. Anton, D.S. Su, T. Jacob and N. Alonso-Vante: Synthesis, Electrochemical Characterization and Molecular Dynamics Studies of Surface Segregation of Platinum Nano-Alloy Electrocatalysts. Physical Chemistry Chemical Physics **13** (20), 9201-9208 (2011).

Fiorito, S., A. Mastrofrancesco, G. Cardinali, E. Rosato, F. Salsano, D.S. Su, A. Serafino and M. Picardo: Effects of carbonaceous nanoparticles from low-emission and older diesel engines on human skin cells. Carbon **49** (15), 5038-5048 (2011).

Frank, B., R. Blume, A. Rinaldi, A. Trunschke and R. Schlögl: Katalyse der Sauerstoffinsertion durch sp^2-Kohlenstoff. Angewandte Chemie **123** (43), 10408-10413 (2011).

Frank, B., R. Blume, A. Rinaldi, A. Trunschke and R. Schlögl: Nanocarbons as Catalyst for Selective Oxidation of Acrolein to Acrylic Acid. In: Proceedings of the DGMK-Conference Catalysis – Innovative Applications in Petrochemistry and Refining, Dresden, Germany. 211-216 (2011). ISBN 978-3-941721-17-3.

Frank, B., R. Blume, A. Rinaldi, A. Trunschke and R. Schlögl: Oxygen Insertion Catalysis by sp^2 Carbon. Angewandte Chemie International Edition **50** (43), 10226-10230 (2011).

Frank, B., S. Wrabetz, O.V. Khavryuchenko, R. Blume, A. Trunschke and R. Schlögl: Calorimetric Study of Propane and Propylene Adsorption on the Active Surface of Multiwalled Carbon Nanotube Catalysts. ChemPhysChem **12** (15), 2709-2713 (2011).

Freund, H.-J., G. Meijer, M. Scheffler, R. Schlögl and M. Wolf: CO oxidation as a prototypical reaction for heterogeneous processes. Angewandte Chemie International Edition **50** (43), 10064-10094 (2011).

Freund, H.-J., G. Meijer, M. Scheffler, R. Schlögl and M. Wolf: Die CO-Oxidation als Modellreaktion für heterogene Prozesse. Angewandte Chemie **123** (43), 10242-10275 (2011).

Friedrich, B., D. Hoffmann and J. James: One hundred years of the Fritz Haber Institute. Angewandte Chemie **123** (43), 10198-10225 (2011).

Friedrich, B., D. Hoffmann and J. James: One hundred years of the Fritz Haber Institute. Angewandte Chemie International Edition **50** (43), 10022-10049 (2011).

Gao, Y., D. Ma, G. Hu, P. Zhai, X. Bao, B. Zhu, B.S. Zhang and D.S. Su: Layered-Carbon-Stabilized Iron Oxide Nanostructures as Oxidation Catalysts. Angewandte Chemie International Edition **50** (43), 10236-10240 (2011).

Gao, Y., D. Ma, G. Hu, P. Zhai, X. Bao, B. Zhu, B.S. Zhang and D.S. Su: Stabilisierung von Eisenoxid-Nanostrukturen mit mehrlagigem Kohlenstoff und Einsatz als Oxidationskatalysator. Angewandte Chemie **123** (43), 10419-10423 (2011).

Gaudet, J., K.K. Bondo, Z. Song, T. Fujitani, W. Zhang, D.S. Su and S.T. Oyama: Effect of gold oxidation state on the epoxidation and hydrogenation of propylene on Au/TS-1. Journal of Catalysis **280** (1), 40-49 (2011).

Hess, C. and R. Schlögl: Nanostructured Catalysts: Selective Oxidations. The Royal Chemical Society, Cambridge, UK (2011). ISBN 978-0-85404-186-2.

Hollmark, H.M., J.R. Vegelius, P.T. Kristiansen, L. Werme and L.C. Duda: Exposure of Oxidized Copper Surfaces to Aqueous Na_2S Solution Studied with Soft X-Ray Spectroscopy. Journal of the Electrochemical Society **158** (1), C1-C5 (2011).

James, J., T. Steinhauser, D. Hoffmann and B. Friedrich: One hundred years at the intersection of chemistry and physics: The Fritz Haber Institute of the Max Planck Society 1911-2011. De Gruyter, Berlin (2011). ISBN 978-3-11-023953-9.

Kolen'ko, Y.V., W. Zhang, R. Naumann d'Alnoncourt, F. Girgsdies, T.W. Hansen, T. Wolfram, R. Schlögl and A. Trunschke: Synthesis of MoVTeNb oxide catalysts with tunable particle dimensions. ChemCatChem **3** (10), 1597-1606 (2011).

Korup, O., S. Mavlyankariev, M. Geske, C.F. Goldsmith and R. Horn: Measurement and analysis of spatial reactor profiles in high temperature catalysis research. Chemical Engineering and Processing **50** (10), 998-1009 (2011).

Lee, J.K., H.-H. Ann, Y. Yi, K.W. Lee, S. Uhm and J. Lee: A stable Ni–B catalyst in hydrogen generation via $NaBH_4$ hydrolysis. Catalysis Communications **16** (1), 120-123 (2011).

Liu, X., A. Wang, T. Zhang, D.S. Su and C.-Y. Mou: Au–Cu alloy nanoparticles supported on silica gel as catalyst for CO oxidation: Effects of Au/Cu ratios. Catalysis today **160** (1), 103-108 (2011).

Majoulet, O., J.G. Alauzun, L. Gottardo, C. Gervais, M.E. Schuster, S. Bernard and P. Miele: Ordered mesoporous silicoboron carbonitride ceramics from boron-modified polysilazanes: Polymer synthesis, processing and properties. Microporous and Mesoporous Materials **140** (1-3), 40-50 (2011).

Miedema, P.S., M.M. van Schooneveld, R. Bogerd, T. Rocha, M. Hävecker, A. Knop-Gericke and F.M.F. de Groot: Oxygen Binding to Cobalt and Iron Phthalocyanines As Determined from in Situ X-ray Absorption Spectroscopy. The Journal of Physical Chemistry C **115** (51), 25422-25428 (2011).

Prati, L., A. Villa, C.E. Chan-Thaw, R. Arrigo, D. Wang and D.S. Su: Gold catalyzed liquid phase oxidation of alcohol: the issue of selectivity. Faraday Discussions **152**, 353-365 (2011).

Qiao, B., A. Wang, J. Lin, L. Li, D.S. Su and T. Zhang: Highly effective CuO/Fe(OH)$_x$ catalysts for selective oxidation of CO in H_2-rich stream. Applied Catalysis B **105** (1-2), 103-110 (2011).

Radziuk, D., A. Skirtach, A. Geßner, M.U. Kumke, W. Zhang, H. Möhwald and D. Shchukin: Ultrasonic Approach for Formation of Erbium Oxide Nanoparticles with Variable Geometries. Langmuir **27** (23), 14472-14480 (2011).

Ranke, W.: Styrene Synthesis: In-Situ Model Catalysis Studies on Ethylbenzene Dehydrogenation. In: Nanostructured Catalysts: Selective Oxidations, Vol. 19. (Eds.) C. Hess and R. Schlögl. The Royal Chemical Society, Cambridge, UK, 266-296 (2011). ISBN 978-0-85404-186-2.

Renn, J., R. Schlögl and H.-P. Zenner: Herausforderung Energie. Edition Open Access, Berlin (2011). ISBN 978-3-8442-0500-8.

Sahin, S., P. Mäki-Arvela, J.-P. Tessonnier, A. Villa, S. Reiche, S. Wrabetz, D.S. Su, R. Schlögl, T. Salmi and D.Y. Murzin: Palladium Catalysts Supported on N-functionalized Hollow Vapour-Grown Carbon Nanofibers: The Effect of the Basic Support and Catalyst Reduction Temperature. Applied Catalysis A **408** (1-3), 137-147 (2011).

Shao, L., W. Zhang, M. Armbrüster, D. Teschner, F. Girgsdies, B. Zhang, O. Timpe, M. Friedich, R. Schlögl and D.S. Su: Nanopartikuläre intermetallische Verbindungen auf Kohlenstoffnanoröhren: aktive und selektive Hydrierungskatalysatoren. Angewandte Chemie **123** (43), 10414-10418 (2011).

Shao, L., W. Zhang, M. Armbrüster, D. Teschner, F. Girgsdies, B. Zhang, O. Timpe, M. Friedrich, R. Schlögl and D.S. Su: Nanosizing Intermetallic Compounds onto Carbon Nanotubes: Active and Selective Hydrogenation Catalysts. Angewandte Chemie International Edition **50** (43), 10231-10235 (2011).

Sonderegger, H., C. Rameshan, H. Lorenz, F. Klauser, M. Klerks, M. Rainer, R. Bakry, C.W. Huck and G.K. Bonn: Surface-assisted laser desorption/ionization-mass spectrometry using TiO_2-coated steel targets for the analysis of small molecules. Analytical and Bioanalytical Chemistry **401** (6), 1963-1974 (2011).

Steinhauser, T., J. James, D. Hoffmann and B. Friedrich: Hundert Jahre an der Schnittstelle von Chemie und Physik: Das Fritz-Haber-Institut der Max-Planck-Gesellschaft zwischen 1911 und 2011. De Gruyter, Berlin (2011). ISBN 978-3-11-023914-0.

Su, D.S.: Anorganische Materialien mit Doppelhelixstruktur. Angewandte Chemie **123** (21), 4843-4847 (2011).

Su, D.S.: Inorganic Materials with Double-Helix Structures. Angewandte Chemie International Edition **50** (21), 4747-4750 (2011).

Su, D.S. and G. Sun: Nichtedelmetallkatalysatoren für die H_2-Brennstoffzelle. Angewandte Chemie **123** (49), 11774-11777 (2011).

Su, D.S. and G. Sun: Nonprecious-Metal Catalysts for Low-Cost Fuel Cells. Angewandte Chemie International Edition **50** (49), 11570-11573 (2011).

Tessonnier, J.-P.: Carbon Nanomaterials: Synthetic Approaches. In: Carbon Nanomaterials, (Ed.) C. Kumar. Wiley-VCH, Weinheim, 3-33 (2011). ISBN 978-3-527-32169-8.

Tessonnier, J.-P. and D.S. Su: Recent Progress on the Growth Mechanism of Carbon Nanotubes: A Review. ChemSusChem **4** (7), 824-847 (2011).

Thielemann, J., G. Weinberg and C. Hess: Controlled Synthesis and Characterization of Highly Dispersed Molybdenum Oxide Supported on Silica SBA-15. ChemCatChem **3** (11), 1814-1821 (2011).

Vogel, D., C. Spiel, Y. Suchorski, A. Urich, R. Schlögl and G. Rupprechter: Mapping the local reaction kinetics by PEEM: CO oxidation on individual (100)-type grains of Pt foil. Surface Science **605** (23-24), 1999-2005 (2011).

Weatherup, R.S., B.C. Bayer, R. Blume, C. Ducati, C. Baehtz, R. Schlögl and S. Hofmann: In Situ Characterization of Alloy Catalysts for Low-Temperature Graphene Growth. Nano Letters **11** (10), 4154-4160 (2011).

Westerström, R., M.E. Messing, S. Blomberg, A. Hellman, H. Grönbeck, J. Gustafson, N.M. Martin, O. Balmes, R. van Rijn, J.N. Andersen, K. Deppert, H. Bluhm, Z. Liu, M.E. Grass, M. Hävecker and E. Lundgren: Oxidation and reduction of Pd(100) and aerosol-deposited Pd nanoparticles. Physical Review B **83** (11), 115440 (2011).

Yi, Y., B.S. Zhang, X. Jin, L. Wang, C.T. Williams, G. Xiong, D.S. Su and C. Liang: Unsupported NiMoW sulfide catalysts for hydrodesulfurization of dibenzothiophene by thermal decomposition of thiosalts. Journal of Molecular Catalysis A **351**, 120-127 (2011).

Zavyalova, U., M. Holena, R. Schlögl and M. Baerns: Statistical Analysis of Past Catalytic Data on Oxidative Methane Coupling for New Insights into the Composition of High-Performance Catalysts. ChemCatChem **3** (12), 1935-1947 (2011).

Zavyalova, U., G. Weinberg, W. Frandsen, F. Girgsdies, T. Risse, K.P. Dinse, R. Schlögl and R. Horn: Lithium as Modifier for Morphology and Defect Structure of Porous Magnesium Oxide Materials Prepared by Gel Combustion Synthesis. ChemCatChem **3** (11), 1779-1788 (2011).

Zhang, B.S., X. Ni, W. Zhang, L. Shao, Q. Zhang, F. Girgsdies, C. Liang, R. Schlögl and D.S. Su: Structural rearrangements of Ru nanoparticles supported on carbon nanotubes under microwave irradiation. Chemical Communications **47** (38), 10716-10718 (2011).

Zhang, B.S., D. Wang, W. Zhang, D.S. Su and R. Schlögl: Structural Dynamics of Low-Symmetry Au Nanoparticles Stimulated by Electron Irradiation. Chemistry - A European Journal **17** (46), 12877-12881 (2011).

Zhang, W., B. Zhang, T. Wolfram, L. Shao, R. Schlögl and D.S. Su: Probing a Redox Behavior of TiO_2/SBA-15 Supported V_xO_y Catalyst Using an Electron Beam in a 200 kV Transmission Electron Microscope. The Journal of Physical Chemistry C **115** (42), 20550-20554 (2011).

Doctoral Thesis

Kühl, S.: Synthesis and Characterization of Cu-based Catalysts resulting from Cu,Zn,X Hydrotalcite-like Compounds. Technische Universität Berlin 2011.

Rameshan, C.: CuZn, PdGa und PdZn Modellkatalysatoren: Herstellung, in-situ Charakterisierung und selektive Katalyse in der Methanoldampfreformierung. Leopold-Franzens-Universität Innsbruck 2011.

Schuster, M.E.: Structure and Reactivity of Diesel Soot Particles from Advanced Motor Technologies. Technische Universität Berlin 2011.

Zheng, W.: Nanomaterials for Ammonia Decomposition. Technische Universität Berlin 2011.

2012

Allen, J.W., C.F. Goldsmith and W.H. Green: Automatic estimation of pressure-dependent rate coefficients. Physical Chemistry Chemical Physics **14** (3), 1131-1155 (2012).

Amakawa, K., S. Wrabetz, J. Kröhnert, G. Tzolova-Müller, R. Schlögl and A. Trunschke: In Situ Generation of Active Sites in Olefin Metathesis. Journal of the American Chemical Society **134** (28), 11462-11473 (2012).

Amrute, A.P., C. Mondelli, M. Moser, G. Novell-Leruth, N. López, D. Rosenthal, R. Farra, M.E. Schuster, D. Teschner, T. Schmidt and J. Pérez-Ramírez: Performance, structure, and mechanism of CeO_2 in HCl oxidation to Cl_2. Journal of Catalysis **286**, 287-297 (2012).

An der Brügge, D., N. Kumar, A. Pukhov and C. Rödel: Influence of Surface Waves on Plasma High-Order Harmonic Generation. Physical Review Letters **108** (12), 125002 (2012).

Armbrüster, M., M. Behrens, F. Cinquini, K. Föttinger, Y. Grin, A. Haghofer, B. Klötzer, A. Knop-Gericke, H. Lorenz, A. Ota, S. Penner, J. Prinz, C. Rameshan, Z. Révay, D. Rosenthal, G. Rupprechter, P. Sautet, R. Schlögl, L. Shao, L. Szentmiklósi, D. Teschner, D. Torres, R. Wagner, R. Widmer and G. Wowsnick: How to Control the Selectivity of Palladium-based Catalysts in Hydrogenation Reactions: The Role of Subsurface Chemistry. ChemCatChem **4** (8), 1048-1063 (2012).

Armbrüster, M., K. Kovnir, M. Friedrich, D. Teschner, G. Wowsnick, M. Hahne, P. Gille, L. Szentmiklósi, M. Feuerbacher, M. Heggen, F. Girgsdies, D. Rosenthal, R. Schlögl and Y. Grin: $Al_{13}Fe_4$ as a low-cost alternative for palladium in heterogeneous hydrogenation. Nature Materials **11** (8), 690-693 (2012).

Arrigo, R., M.E. Schuster, S. Wrabetz, F. Girgsdies, J.-P. Tessonnier, G. Centi, S. Perathoner, D.S. Su and R. Schlögl: New Insights from Microcalorimetry on the FeO_x/CNT-Based Electrocatalysts Active in the Conversion of CO_2 to Fuels. ChemSusChem **5** (3), 577-586 (2012).

Arrigo, R., S. Wrabetz, M.E. Schuster, D. Wang, A. Villa, D. Rosenthal, F. Girgsdies, G. Weinberg, L. Prati, R. Schlögl and D.S. Su: Tailoring the morphology of Pd nanoparticles on CNTs by nitrogen and oxygen functionalization. Physical Chemistry Chemical Physics **14** (30), 10523-10532 (2012).

Baerns, M.: Development of Catalytic Materials. In: Catalysis: From Principles to Application, (Eds.) M. Beller, A. Renken and R.A. van Santen. Wiley-VCH, Weinheim, 445-492 (2012). ISBN 978-3-527-32349-4.

Bauer, M., R. Schoch, L. Shao, B.S. Zhang, A. Knop-Gericke, M.G. Willinger, R. Schlögl and D. Teschner: Structure–Activity Studies on Highly Active Palladium Hydrogenation Catalysts by X-ray Absorption Spectroscopy. The Journal of Physical Chemistry C **116** (42), 22375-22385 (2012).

Bayer, B., M. Fouquet, R. Blume, C.T. Wirth, R. Weatherup, K. Ogata, A. Knop-Gericke, R. Schlögl, S. Hofmann and J. Robertson: Co-Catalytic Solid-State Reduction Applied to Carbon Nanotube Growth. The Journal of Physical Chemistry C **116** (1), 1107-1113 (2012).

Beck, B., M. Harth, N. Hamilton, C. Carrero, J. Uhlrich, A. Trunschke, S.K. Shaikhutdinov, H. Schubert, H.-J. Freund, R. Schlögl, J. Sauer and R. Schomäcker: Partial oxidation of ethanol on vanadia catalysts on supporting oxides with different redox properties compared to propane. Journal of Catalysis **296**, 120-131 (2012).

Behrens, M. and M. Armbrüster: Methanol Steam Reforming. In: Catalysis for Alternative Energy Generation, (Eds.) L. Guczi and A. Erdôhelyi. Springer, New York, 175-235 (2012). ISBN 978-1-4614-0343-2.

Behrens, M. and R. Schlögl: X-Ray Diffraction and Small Angle X-Ray Scattering. In: Characterization of Solid Materials and Heterogeneous Catalysts: From Structure to Surface Reactivity: Part Two: Macroscopic Techniques, Vol. 2. (Eds.) J.C. Védrine and M. Che. Wiley-VCH, Weinheim, 611-653 (2012). ISBN 978-3-527-32687-7.

Behrens, M., F. Studt, I. Kasatkin, S. Kühl, M. Hävecker, F. Abild-Pedersen, S. Zander, F. Girgsdies, P. Kurr, B.-L. Kniep, M. Tovar, R.-W. Fischer, J.K. Nørskov and R. Schlögl: The Active Site of Methanol Synthesis over $Cu/ZnO/Al_2O_3$ Industrial Catalysts. Science **336** (6083), 893-897 (2012).

Bielz, T., S. Soisuwan, F. Girgsdies, B. Klötzer and S. Penner: Reduction of Different GeO_2 Polymorphs. The Journal of Physical Chemistry C **116** (18), 9961-9968 (2012).

Checa, A., E.M. Harper and M.G. Willinger: Aragonitic dendritic prismatic shell microstructure in Thracia (Bivalvia, Anomalodesmata). Invertebrate biology **131** (1), 19-29 (2012).

Chen, C., Q. Zhang, J.-Q. Huang, W. Zhang, X.-C. Zhao, C.-H. Huang, F. Wei, Y.-G. Yang, M.-Z. Wang and D.S. Su: Chemically derived graphene-metal oxide hybrids as electrodes for electrochemical energy storage: pre-graphenization or post-graphenization? Journal of Materials Chemistry **22** (28), 13947-13955 (2012).

Chen, C., Q. Zhang, X.-C. Zhao, B.S. Zhang, Q.-Q. Kong, M.-G. Yang, Q.-H. Yang, M.-Z. Wang, Y.-G. Yang, R. Schlögl and D.S. Su: Hierarchically aminated graphene honeycombs for electrochemical capacitive energy storage. Journal of Materials Chemistry **22** (28), 14076-14084 (2012).

Chen, R., V. Trieu, A.R. Zeradjanin, H. Natter, D. Teschner, J. Kintrup, A. Bulan, W. Schuhmann and R. Hempelmann: Microstructural impact of anodic coatings on the electrochemical chlorine evolution reaction. Physical Chemistry Chemical Physics **14** (20), 7392-7399 (2012).

Chen, X., B.S. Zhang, C. Li, Z. Shao, D.S. Su, C.T. Williams and C. Liang: Structural and electrochemical properties of nanostructured nickel silicides by reduction and silicification of high-surface-area nickel oxide. Materials Research Bulletin **47** (3), 867-877 (2012).

Chinthaginjala, J.K., A. Villa, D.S. Su, B.L. Mojet and L. Lefferts: Nitrite reduction over Pd supported CNFs: Metal particle size effect on selectivity. Catalysis today **183** (1), 119-123 (2012).

Conrad, F., C. Massue, S. Kühl, E.L. Kunkes, F. Girgsdies, I. Kasatkin, B.S. Zhang, M. Friedrich, Y. Luo, M. Armbrüster, G.R. Patzke and M. Behrens: Microwave-hydrothermal synthesis and characterization of nanostructured copper substituted ZnM_2O_4 (M = Al, Ga) spinels as precursors for thermally stable Cu catalysts. Nanoscale **4** (6), 2018-2028 (2012).

Delgado, J.J., X. Chen, B. Frank, D.S. Su and R. Schlögl: Activation Processes of Highly Ordered Carbon Nanofibers in the Oxidative Dehydrogenation of Ethylbenzene. Catalysis today **186** (1), 93-98 (2012).

Diller, K., F. Klappenberger, M. Marschall, K. Hermann, A. Nefedov, C. Wöll and J.V. Barth: Self-metalation of 2H-tetraphenylporphyrin on Cu(111): An x-ray spectroscopy study. The Journal of Chemical Physics **136** (1), 014705 (2012).

Dinse, A., C. Carrero, A. Ozarowski, R. Schomäcker, R. Schlögl and K.-P. Dinse: Characterization and Quantification of Reduced Sites on Supported Vanadium Oxide Catalysts by Using High-Frequency Electron Paramagnetic Resonance. ChemCatChem **4** (5), 641-652 (2012).

Dlubak, B., M.-B. Martin, R.S. Weatherup, H. Yang, C. Deranlot, R. Blume, R. Schlögl, A. Fert, A. Anane, S. Hofmann, P. Seneor and J. Robertson: Graphene-Passivated Nickel as an Oxidation-Resistant Electrode for Spintronics. ACS Nano **6** (12), 10930-10934 (2012).

Eichelbaum, M., M. Hävecker, C. Heine, A. Karpov, C.-K. Dobner, F. Rosowski, A. Trunschke and R. Schlögl: The Intimate Relationship between Bulk Electronic Conductivity and Selectivity in the Catalytic Oxidation of n-Butane. Angewandte Chemie International Edition **51** (25), 6246-6250 (2012).

Eichelbaum, M., M. Hävecker, C. Heine, A. Karpov, C.-K. Dobner, F. Rosowski, A. Trunschke and R. Schlögl: Über den Zusammenhang zwischen elektronischer Volumenleitfähigkeit und Selektivität in der katalytischen Oxidation von n-Butan. Angewandte Chemie **124** (25), 6350-6354 (2012).

Eichelbaum, M., R. Stößer, A. Karpov, C.-K. Dobner, F. Rosowski, A. Trunschke and R. Schlögl: The microwave cavity perturbation technique for contact-free and in situ electrical conductivity measurements in catalysis and materials science. Physical Chemistry Chemical Physics **14** (3), 1302-1312 (2012).

Fellinger, T.-P., D.S. Su, M. Engenhorst, D. Gautam, R. Schlögl and M. Antonietti: Thermolytic synthesis of graphitic boron carbon nitride from an ionic liquid precursor: Mechanism, structure analysis and electronic properties. Journal of Materials Chemistry **22** (45), 23996-24005 (2012).

Fernandes, C.M., M.G. Willinger, M.T. Vieira and A.M.R. Senos: Interface Exploring of Tungsten Carbide-Stainless Steel Composites through HRTEM. Microscopy and Microanalysis **18** (S5), 109-110 (2012).

Fouquet, M., B.C. Bayer, S. Esconjauregui, R. Blume, J.H. Warner, S. Hofmann, R. Schlögl, C. Thomsen and J. Robertson: Highly chiral-selective growth of single-walled carbon nanotubes with a simple monometallic Co catalyst. Physical Review B **85** (23), 235411 (2012).

Friedrich, M., D. Teschner, A. Knop-Gericke and M. Armbrüster: Influence of bulk composition of the intermetallic compound ZnPd on surface composition and methanol steam reforming properties. Journal of Catalysis **285**, 41-47 (2012).

Friedrich, M., D. Teschner, A. Knop-Gericke and M. Armbrüster: Surface and Subsurface Dynamics of the Intermetallic Compound ZnNi in Methanol Steam Reforming. The Journal of Physical Chemistry C **116** (28), 14930-14935 (2012).

Girgsdies, F. and M. Behrens: On the structural relations of malachite. I. The rosasite and ludwigite structure families. Acta Crystallographica B **68** (2), 107-117 (2012).

Girgsdies, F. and M. Behrens: On the structural relations of malachite. II. The brochantite MDO polytypes. Acta Crystallographica B **68** (6), 571-577 (2012).

Girgsdies, F., R. Schlögl and A. Trunschke: In-situ X-ray diffraction study of phase crystallization from an amorphous MoVTeNb oxide catalyst precursor. Catalysis Communications **18** (10), 60-62 (2012).

Glaum, R., C. Welker-Nieuwoudt, C.-K. Dobner, M. Eichelbaum, F. Gruchow, C. Heine, A. Karpov, R. Kniep, F. Rosowski, R. Schlögl, S.A. Schunk, S. Titlbach and A. Trunschke: Resource-Efficient Alkane Selective Oxidation on New Crystalline Solids: Searching for Novel Catalyst Materials. Chemie-Ingenieur-Technik **84** (10), 1766-1779 (2012).

Goldsmith, C.F.: Estimating the Thermochemistry of Adsorbates Based Upon Gas-Phase Properties. Topics in Catalysis **55** (5-6), 366-375 (2012).

Goldsmith, C.F., W.H. Green and S.J. Klippenstein: Role of O_2 + QOOH in Low-Temperature Ignition of Propane. 1. Temperature and Pressure Dependent Rate Coefficients. The Journal of Physical Chemistry A **116** (13), 3325-3346 (2012).

Goldsmith, C.F., G.R. Magoon and W.H. Green: Database of Small Molecule Thermochemistry for Combustion. The Journal of Physical Chemistry A **116** (36), 9033-9057 (2012).

Guo, C., K. Hermann, M. Hävecker, A. Trunschke and R. Schlögl: Silica-Supported Titania Species: Structural Analysis from Quantum Theory and X-ray Spectroscopy. The Journal of Physical Chemistry C **116** (42), 22449-22457 (2012).

Guo, C., L. Sun, K. Hermann, C.F. Hermanns, M. Bernien and W. Kuch: X-ray absorption from large molecules at metal surfaces: Theoretical and experimental results for Co-OEP on Ni(100). The Journal of Chemical Physics **137** (19), 194703 (2012).

Haghöfer, A., K. Föttinger, F. Girgsdies, D. Teschner, A. Knop-Gericke, R. Schlögl and G. Rupprechter: In Situ Study of the Formation and Stability of Supported Pd_2Ga Methanol Steam Reforming Catalysts. Journal of Catalysis **286**, 13-21 (2012).

Halevi, B., E.J. Peterson, A. Roy, A. DeLariva, E. Jeroro, F. Gao, Y. Wang, J.M. Vohs, B. Kiefer, E.L. Kunkes, M. Hävecker, M. Behrens, R. Schlögl and A.K. Datye: Catalytic reactivity of face centered cubic $PdZn_{\alpha}$ for the steam reforming of methanol. Journal of Catalysis **291**, 44-54 (2012).

Hamilton, N., T. Wolfram, G. Tzolova-Müller, M. Hävecker, J. Kröhnert, C. Carrero, R. Schomäcker, A. Trunschke and R. Schlögl: Topology of silica supported vanadium–titanium oxide catalysts for oxidative dehydrogenation of propane. Catalysis Science & Technology **2** (7), 1346-1359 (2012).

Hao, Q.-Q., Z.-W. Liu, B.S. Zhang, G.-W. Wang, C. Ma, W. Frandsen, J. Li, Z.-T. Liu, Z. Hao and D.S. Su: Porous Montmorillonite Heterostructures Directed by a Single Alkyl Ammonium Template for Controlling the Product Distribution of Fischer-Tropsch Synthesis over Cobalt. Chemistry of Materials **24** (6), 972-974 (2012).

Hävecker, M., S. Wrabetz, J. Kröhnert, L.-I. Csepei, R. Naumann d'Alnoncourt, Y.V. Kolen'ko, F. Girgsdies, R. Schlögl and A. Trunschke: Surface chemistry of phase-pure M1 MoVTeNb oxide during operation in selective oxidation of propane to acrylic acid. Journal of Catalysis **285**, 48-60 (2012).

Hermann, K.: Periodic overlayers and moiré patterns: Theoretical studies of geometric properties. Journal of Physics: Condensed Matter **24** (31), 314210 (2012).

Hill, L.J., M.M. Bull, Y. Sung, A.G. Simmonds, P.T. Dirlam, N.E. Richey, S.E. DeRosa, I.-B. Shim, D. Guin, P.J. Costanzo, N. Pinna, M.G. Willinger, W. Vogel, K. Char and J. Pyun: Directing the Deposition of Ferromagnetic Cobalt onto Pt-tipped CdSe@CdS Nanorods: Synthetic and Mechanistic Insights. ACS Nano **6** (10), 8632-8645 (2012).

Holeňa, M., N. Steinfeldt, M. Baerns and D. Ŝtefka: Computing the correlation between catalyst composition and its performance in the catalysed process. Computers and Chemical Engineering **43** (10), 55-67 (2012).

Huang, C.-H., Q. Zhang, T.-C. Chou, C. Chen, D.S. Su and R.-A. Doong: Three-Dimensional Hierarchically Ordered Porous Carbons with Partially Graphitic Nanostructures for Electrochemical Capacitive Energy Storage. ChemSusChem **5** (3), 563-571 (2012).

Huang, J.-Q., M.-Q. Zhao, Q. Zhang, J.-Q. Nie, L. Yao, D.S. Su and F. Wei: Efficient synthesis of aligned nitrogen-doped carbon nanotubes in a fluidized-bed reactor. Catalysis today **186** (1), 83-92 (2012).

Huang, X., L. Shao, G.-W. She, M. Wang and X.-M. Meng: Catalyst-free synthesis of single crystalline ZnO nanonails with ultra-thin caps. CrystEngComm **14** (24), 8330-8334 (2012).

Huang, X., M. Wang, M.G. Willinger, L. Shao, D.S. Su and X.-M. Meng: Assembly of Three-Dimensional Hetero-Epitaxial ZnO/ZnS Core/Shell Nanorod and Single Crystalline Hollow ZnS Nanotube Arrays. ACS Nano **6** (8), 7333-7339 (2012).

Jeon, T.-Y., N. Pinna, S.J. Yoo, D. Ahn, S.H. Choi, M.G. Willinger, Y.-H. Cho, K.-S. Lee, H.-Y. Park, S.-H. Yu and Y.-E. Sung: Selective deposition of Pt onto supported metal clusters for fuel cell electrocatalysts. Nanoscale **4** (20), 6461-6469 (2012).

Johnson, B., J. Klaer, C.-H. Fischer and I. Lauermann: Depth profiling of a CdS buffer layer on $CuInS_2$ measured with X-ray photoelectron spectroscopy during removal by HCl etching. Thin solid films **520** (7), 2829-2832 (2012).

Kandemir, T., F. Girgsdies, I. Kasatkin, E.L. Kunkes, K.-D. Liss, V.K. Peterson, R. Schlögl and M. Behrens: Heterogeneous Catalysis under pressure - In-situ neutron diffraction under industrial conditions. Journal of Physics: Conference Series **340** (1), 012053 (2012).

Kandemir, T., D. Wallacher, T. Hansen, K.-P. Liss, R. Naumann d'Alnoncourt, R. Schlögl and M. Behrens: In situ neutron diffraction under high pressure - Providing an insight into working catalysts. Nuclear Instruments and Methods in Physics Research A **673** (1), 51-55 (2012).

Kolen'ko, Y.V., K. Amakawa, R. Naumann d'Alnoncourt, F. Girgsdies, G. Weinberg, R. Schlögl and A. Trunschke: Cover Picture: Unusual Phase Evolution in MoVTeNb Oxide Catalysts Prepared by a Novel Acrylamide-Gelation Route. ChemCatChem **4** (4), 417-417 (2012).

Kolen'ko, Y.V., K. Amakawa, R. Naumann d'Alnoncourt, F. Girgsdies, G. Weinberg, R. Schlögl and A. Trunschke: Unusual Phase Evolution in MoVTeNb Oxide Catalysts Prepared by a Novel Acrylamide-Gelation Route. ChemCatChem **4** (4), 495-503 (2012).

Korup, O., R. Schlögl and R. Horn: Carbon formation in catalytic partial oxidation of methane on platinum: Model studies on a polycrystalline Pt foil. Catalysis today **181** (1), 177-183 (2012).

Kühl, S., M. Friedrich, M. Armbrüster and M. Behrens: Cu,Zn,Al layered double hydroxides as precursors for copper catalysts in methanol steam reforming – pH-controlled synthesis by microemulsion technique. Journal of Materials Chemistry **22** (19), 9632-9638 (2012).

Langfeld, K., B. Frank, V.A. Strempel, C. Berger-Karin, G. Weinberg, E.V. Kondratenko and R. Schomäcker: Comparison of oxidizing agents for the oxidative coupling of methane over state-of-the-art catalysts. Applied Catalysis A **417-418**, 145-152 (2012).

Leary, R., Z. Saghi, M. Armbrüster, R. Schlögl, J.M. Thomas and P. Midgley: Quantitative HAADF-STEM tomography of unsupported intermetallic Ga-Pd catalysts. Journal of Physics: Conference Series **371** (1), 012024 (2012).

Leary, R., Z. Saghi, M. Armbrüster, G. Wowsnick, R. Schlögl, J.M. Thomas and P. Midgley: Quantitative High-Angle Annular Dark-Field Scanning Transmission Electron Microscope (HAAF-STEM)Tomography and High Resolution Electron Microscopy of Unsupported Intermetallic $GaPd_2$ Catalysts. The Journal of Physical Chemistry C **116** (24), 13343-13352 (2012).

Lee, J., H. Ju, Y. Yi, J. Lee, S. Uhm, J.K. Lee and H.J. Lee: High-Density Nanoporous Structures for Enhanced Electrocatalysis. The Journal of Physical Chemistry C **116** (4), 2915-2918 (2012).

Li, L., W. Wei and M. Behrens: Synthesis and characterization of α-, β-, and γ-Ga_2O_3 prepared from aqueous solutions by controlled precipitation. Solid State Sciences **14** (7), 971-981 (2012).

Li, L., B.S. Zhang, E.L. Kunkes, K. Föttinger, M. Armbrüster, D.S. Su, W. Wei, R. Schlögl and M. Behrens: Ga-Pd/Ga_2O_3 Catalysts: The Role of Gallia Polymorphs, Intermetallic Compounds, and Pretreatment Conditions on Selectivity and Stability in Different Reactions. ChemCatChem **4** (11), 1764-1775 (2012).

Li, X.-H., J.-S. Chen, X. Wang, M.E. Schuster, R. Schlögl and M. Antonietti: A Green Chemistry of Graphene: Photochemical Reduction towards Monolayer Graphene Sheets and the Role of Water Adlayers. ChemSusChem **5** (4), 642-646 (2012).

Liu, J., L. Jiang, Q. Tang, B.S. Zhang, D.S. Su, S. Wang and G. Sun: Coupling Effect Between Cobalt Oxides And Carbon For Oxygen Reduction Reaction. ChemSusChem **5** (12), 2315-2318 (2012).

Loo, A.Y.Y., Y.P. Lay, M.G. Kutty, O. Timpe, M. Behrens and S.B. Abd Hamid: Spectrophotometric Determination of Mercury with Iodide and Rhodamine B. Sains Malaysiana **41** (2), 213-218 (2012).

Lublow, M., B. Bouabadi and S. Kubala: Au–Pt core–shell nanoemitters on silicon for photoelectrochemical solar energy conversion. Solar Energy Materials and Solar Cells **107**, 56-62 (2012).

Lunkenbein, T., D. Rosenthal, T. Otremba, F. Girgsdies, Z. Li, H. Sai, C. Bojer, G. Auffermann, U. Wiesner and J. Breu: Access to Ordered Porous Molybdenum Oxycarbide/Carbon Nanocomposites. Angewandte Chemie International Edition **51** (51), 12892-12892 (2012).

Lunkenbein, T., D. Rosenthal, T. Otremba, F. Girgsdies, Z. Li, H. Sai, C. Bojer, G. Auffermann, U. Wiesner and J. Breu: Über die Zugänglichkeit zu geordneten porösen Molybdänoxycarbid/Kohlenstoff-Nanokompositen. Angewandte Chemie **124** (51), 13066-13070 (2012).

Mansor, N.B.A., J.-P. Tessonnier, A. Rinaldi, S. Reiche and M.G. Kutty: Chemically Modified Multi-walled Carbon Nanotubes (MWCNTs) with Anchored Acidic Groups. Sains Malaysiana **41** (5), 603-609 (2012).

Marichy, C., J.-P. Tessonnier, M.C. Ferro, K.-H. Lee, R. Schlögl, N. Pinna and M.G. Willinger: Labeling and monitoring the distribution of anchoring sites on functionalized CNTs by atomic layer deposition. Journal of Materials Chemistry **22** (15), 7323-7330 (2012).

Mette, K., A. Bergmann, J.-P. Tessonnier, M. Hävecker, L. Yao, T. Ressler, R. Schlögl, P. Strasser and M. Behrens: Nanostructured Manganese Oxide Supported on Carbon Nanotubes for Electrocatalytic Water Splitting. ChemCatChem **4** (6), 851-862 (2012).

Mota, N., M.C. Alvarez-Galván, R.M. Navarro, S.M. Al-Zahrani, A. Goguet, H.B. Daly, W. Zhang, A. Trunschke, R. Schlögl and J.L.G. Fierro: Insights on the role of Ru substitution in the properties of $LaCoO_3$-based oxides as catalysts precursors for the oxidative reforming of diesel fuel. Applied Catalysis B **113-114**, 271-280 (2012).

Mou, X., Y. Li, B.S. Zhang, L. Yao, X. Wei, D.S. Su and W. Shen: Crystal-Phase- and Morphology-Controlled Synthesis of Fe_2O_3 Nanomaterials. European Journal of Inorganic Chemistry **2012** (16), 2684-2690 (2012).

Mou, X., B.S. Zhang, Y. Li, L. Yao, X. Wei, D.S. Su and W. Shen: Rod-Shaped Fe_2O_3 as an Efficient Catalyst for the Selective Reduction of Nitrogen Oxide by Ammonia. Angewandte Chemie International Edition **51** (12), 2989-2993 (2012).

Mou, X., B.S. Zhang, Y. Li, L. Yao, X. Wei, D.S. Su and W. Shen: Stäbchenförmiges Fe_2O_3 als effektiver Katalysator für die selektive katalytische Reduktion von NO mit NH_3. Angewandte Chemie **124** (12), 3044-3048 (2012).

Moya, S.F., R.L. Martins, A. Ota, E.L. Kunkes, M. Behrens and M. Schmal: Nanostructured supported palladium catalysts—Non-oxidative methane coupling. Applied Catalysis A **411-412**, 105-113 (2012).

Müller, J.-O., B. Frank, R.E. Jentoft, R. Schlögl and D.S. Su: The oxidation of soot particulate in the presence of NO_2. Catalysis today **191** (1), 106-111 (2012).

Naumann d'Alnoncourt, R., Y.V. Kolen'ko, R. Schlögl and A. Trunschke: A new way of probing reaction networks: analyzing multidimensional parameter space. Combinatorial Chemistry & High Throughput Screening **15** (2), 161-169 (2012).

Ni, X., B.S. Zhang, C. Li, M. Pang, D.S. Su, C.T. Williams and C. Liang: Microwave-assisted green synthesis of uniform Ru nanoparticles supported on non-functional carbon nanotubes for cinnamaldehyde hydrogenation. Catalysis Communications **24**, 65-69 (2012).

Ota, A., E.L. Kunkes, I. Kasatkin, E. Groppo, D. Ferri, B. Poceiro, R.M. Navarro Yerga and M. Behrens: Comparative study of hydrotalcite-derived supported Pd_2Ga and PdZn intermetallic nanoparticles as methanol synthesis and methanol steam reforming catalysts. Journal of Catalysis **293**, 27-38 (2012).

Ovsitser, O., R. Schomäker, E.V. Kondratenko, T. Wolfram and A. Trunschke: Highly selective and stable propane dehydrogenation to propene over dispersed VO_x-species under oxygen-free and oxygen-lean conditions. Catalysis today **192** (1), 16-19 (2012).

Papaefthimiou, V., T. Dintzer, M. Lebedeva, D. Teschner, M. Hävecker, A. Knop-Gericke, R. Schlögl, V. Pierron-Bonnes, E.R. Savinova and S. Zafeiratos: Probing Metal-Support Interaction in Reactive Environments: An in Situ Study of PtCo Bimetallic Nanoparticles Supported on TiO_2. The Journal of Physical Chemistry C **116** (27), 14342-14349 (2012).

Parapat, R.Y., V. Parwoto, M. Schwarze, B.S. Zhang, D.S. Su and R. Schomäcker: A new method to synthesize very active and stable supported metal Pt catalysts: Thermo-destabilization of microemulsions. Journal of Materials Chemistry **22** (23), 11605-11614 (2012).

Pucci, A., M.G. Willinger, F. Liu, X. Zeng, V. Rebuttini, G. Clavel, B. Xue, G. Ungar and N. Pinna: One-Step Synthesis and Self-Assembly of Metal Oxide Nanoparticles into 3D Superlattices. ACS Nano **6** (5), 4382-4391 (2012).

Qian, X.F., B.S. Li, Y.Y. Hu, G.X. Niu, D.Y.H. Zhang, R.C. Che, Y. Tang, D.S. Su, A.M. Asiri and D. Zhao: Exploring Meso-/Microporous Composite Molecular Sieves with Core–Shell Structures. Chemistry - A European Journal **18** (3), 931-939 (2012).

Rameshan, C., H. Lorenz, L. Mayr, S. Penner, D. Zemlyanov, R. Arrigo, M. Hävecker, R. Blume, A. Knop-Gericke, R. Schlögl and B. Klötzer: CO_2-selective methanol steam reforming on In-doped Pd studied by in situ X-ray photoelectron spectroscopy. Journal of Catalysis **295**, 186-194 (2012).

Rameshan, C., W. Stadlmayr, S. Penner, H. Lorenz, L. Mayr, M. Hävecker, R. Blume, T. Rocha, D. Teschner, A. Knop-Gericke, R. Schlögl, D. Zemlyanov, N. Memmel and B. Klötzer: In situ XPS study of methanol reforming on PdGa near-surface intermetallic phases. Journal of Catalysis **290**, 126-137 (2012).

Rameshan, C., W. Stadlmayr, S. Penner, H. Lorenz, N. Memmel, M. Hävecker, R. Blume, D. Teschner, T. Rocha, D. Zemlyanov, A. Knop-Gericke, R. Schlögl and B. Klötzer: Hydrogen Production by Methanol Steam Reforming on Copper Boosted by Zinc-Assisted Water Activation. Angewandte Chemie International Edition **51** (12), 3002-3006 (2012).

Rameshan, C., W. Stadlmayr, S. Penner, H. Lorenz, N. Memmel, M. Hävecker, R. Blume, D. Teschner, T. Rocha, D. Zemlyanov, A. Knop-Gericke, R. Schlögl and B. Klötzer: Steigerung der Wasserstoffproduktion in der Methanol-Dampfreformierung auf Kupfer durch Zink-unterstützte Wasseraktivierung. Angewandte Chemie **124** (12), 3057-3061 (2012).

Rocha, T., A. Knop-Gericke and R. Schlögl: Comment on “Strongly-Bound Oxygen Species on Silver Surfaces: A Molybdenum Oxide Contamination?” The Journal of Physical Chemistry C **116** (20), 11408-11409 (2012).

Rocha, T., A. Oestereich, D.V. Demidov, M. Hävecker, S. Zafeiratos, G. Weinberg, V.I. Bukhtiyarov, A. Knop-Gericke and R. Schlögl: The silver–oxygen system in catalysis: New insights by near ambient pressure X-ray photoelectron spectroscopy. Physical Chemistry Chemical Physics **14** (13), 4554-4564 (2012).

Rocha, T., H. Winnischofer and D. Zanchet: Structural Aspects of Anisotropic Metal Nanoparticle Growth: Experiment and Theory. In: Complex-Shaped Metal Nanoparticles: Bottom-Up Syntheses and Applications, (Eds.) T.K. Sau and A.L. Rogach. Wiley-VCH, Weinheim, 215-238 (2012). ISBN 978-3-527-33077-5.

Rosenthal, D., R. Widmer, R. Wagner, P. Gille, M. Armbrüster, Y. Grin, R. Schlögl and O. Gröning: Surface Investigation of Intermetallic $PdGa(\bar{1}\bar{1}\bar{1})$. Langmuir **28** (17), 6848-6856 (2012).

Sanchez Sanchez, M., F. Girgsdies, M. Jastak, P. Kube, R. Schlögl and A. Trunschke: Aiding the Self-Assembly of Supramolecular Polyoxometalates under Hydrothermal Conditions To Give Precursors of Complex Functional Oxides. Angewandte Chemie International Edition **51** (29), 7194-7197 (2012).

Sanchez Sanchez, M., F. Girgsdies, M. Jastak, P. Kube, R. Schlögl and A. Trunschke: Aiding the Self-Assembly of Supramolecular Polyoxometalates under Hydrothermal Conditions To Give Precursors of Complex Functional Oxides. Angewandte Chemie **124** (29), 7306-7309 (2012).

Schäfer, S., S.A. Wyrzgol, R. Caterino, A. Jentys, S.J. Schoell, M. Hävecker, A. Knop-Gericke, J.A. Lercher, I.D. Sharp and M. Stutzmann: Platinum Nanoparticles on Gallium Nitride Surfaces: Effect of Semiconductor Doping on Nanoparticle Reactivity. Journal of the American Chemical Society **134** (30), 12528-12535 (2012).

Schlögl, R.: Die Energiewende: Eine Herausforderung für die Chemie. Nachrichten aus der Chemie **60** (6), 621-621 (2012).

Schüth, F., R. Palkovits, R. Schlögl and D.S. Su: Ammonia as a possible element in an energy infrastructure: Catalysts for ammonia decomposition. Energy & Environmental Science **5** (4), 6278-6289 (2012).

Schwarz, H., R. Schlögl and R. Horn: Radical detection in harsh environments by means of laser-induced fluorescence using a single bidirectional optical fiber. Applied Physics B **109** (1), 19-26 (2012).

Shao, L., B.S. Zhang, W. Zhang, D. Teschner, F. Girgsdies, R. Schlögl and D.S. Su: Improved Selectivity by Stabilizing and Exposing Active Phases on Supported Pd Nanoparticles in Acetylene-Selective Hydrogenation. Chemistry - A European Journal **18** (47), 14962-14966 (2012).

Simo, A., J. Polte, N. Pfänder, U. Vainio, F. Emmerling and K. Rademann: Formation Mechanism of Silver Nanoparticles Stabilized in Glassy Matrices. Journal of the American Chemical Society **134** (45), 18824-18833 (2012).

Som, T., A. Simo, R. Fenger, G.V. Troppenz, R. Bansen, N. Pfänder, F. Emmerling, J. Rappich, T. Boeck and K. Rademann: Bismuth Hexagons: Facile Mass Synthesis, Stability and Applications. ChemPhysChem **13** (8), 2162-2169 (2012).

Su, D.S.: Editorial: Chemistry of Energy Conversion and Storage. ChemSusChem **5** (3), 443-445 (2012).

Su, D.S., S. Perathoner and G. Centi: Preface: Catalysis on nano-carbon materials: Going where to? Catalysis today **186** (1), 1-6 (2012).

Teschner, D., R. Farra, L. Yao, R. Schlögl, H. Soerijanto, R. Schomäcker, T. Schmidt, L. Szentmiklósi, A. Amrute, C. Mondelli, J. Perez-Ramirez, G. Novell-Leruth and N. Lopez: An integrated approach to Deacon chemistry on RuO_2-based catalysts. Journal of Catalysis **285**, 273-284 (2012).

Teschner, D., G. Novell-Leruth, R. Farra, A. Knop-Gericke, R. Schlögl, L. Szentimiklósi, M.G. Hevia, H. Soerijanto, R. Schomäcker, J. Pérez-Ramírez and N. López: In situ surface coverage analysis of RuO_2-catalysed HCl oxidation reveals the entropic origin of compensation in heterogeneous catalysis. Nature Chemistry **4** (9), 739-745 (2012).

Teschner, D., A. Wootsch and Z. Páal: Preferential CO oxidation in hydrogen (PROX) on unsupported PtSn catalyst. Applied Catalysis A **411-412**, 31-34 (2012).

Toghan, A., R. Arrigo, A. Knop-Gericke and R. Imbihl: Ambient pressure X-ray photoelectron spectroscopy during electrochemical promotion of ethylene oxidation over a bimetallic Pt–Ag/YSZ catalyst. Journal of Catalysis **296**, 99-109 (2012).

Vogel, D., C. Spiel, Y. Suchorski, A. Trinchero, R. Schlögl, H. Grönbeck and G. Rupprechter: Local Catalytic Ignition during CO Oxidation on Low-Index Pt and Pd Surfaces: A Combined PEEM, MS, and DFT Study. Angewandte Chemie International Edition **51** (40), 10041-10044 (2012).

Vogel, D., C. Spiel, Y. Suchorski, A. Trinchero, R. Schlögl, H. Grönbeck and G. Rupprechter: Lokale katalytische Zündung der CO-Oxidation auf individuellen niedrig-indizierten Pt- und Pd-Oberflächen: kombinierte PEEM-, MS- und DFT-Untersuchungen. Angewandte Chemie **124** (40), 10185-10189 (2012).

Wang, L., J.J. Delgado, B. Frank, Z. Zhang, Z. Shan, D.S. Su and F.-S. Xiao: Resin-Derived Hierarchical Porous Carbon Spheres with High Catalytic Performance in the Oxidative Dehydrogenation of Ethylbenzene. ChemSusChem **5** (4), 687-693 (2012).

Wang, L., W. Zhang, D.S. Su, X. Meng and F.-S. Xiao: Supported Au nanoparticles as efficient catalysts for aerobic homocoupling of phenylboronic acid. Chemical Communications **48** (44), 5476-5478 (2012).

Wang, L., W. Zhang, S. Zeng, D.S. Su, X. Meng and F. Xiao: Mg-Al Mixed Oxides Supported Bimetallic Au-Pd Nanoparticles with Superior Catalytic Properties in Aerobic Oxidation of Benzyl Alcohol and Glycerol. Chinese Journal of Chemistry **30** (9), 2189-2197 (2012).

Weatherup, R.S., B.C. Bayer, R. Blume, C. Baehtz, P.R. Kidambi, M. Fouquet, C.T. Wirth, R. Schlögl and S. Hofmann: On the Mechanisms of Ni-Catalysed Graphene Chemical Vapour Deposition. ChemPhysChem **13** (10), 2544-2549 (2012).

Wohlgemuth, S.-A., R.J. White, M.G. Willinger, M.-M. Titirici and M. Antonietti: A one-pot hydrothermal synthesis of sulfur and nitrogen doped carbon aerogels with enhanced electrocatalytic activity in the oxygen reduction reaction. Green Chemistry **14** (5), 1515-1523 (2012).

Wulfers, M.J., G. Tzolova-Müller, J.I. Villegas, D.Y. Murzin and F.C. Jentoft: Evolution of carbonaceous deposits on H-mordenite and Pt-doped H-mordenite during n-butane conversion. Journal of Catalysis **296**, 132-142 (2012).

Xu, Z., H. Zhang, H. Zhong, Q. Lu, J. Wang and D.S. Su: Effect of particle size on the activity and durability of the Pt/C electrocatalyst for proton exchange membrane fuel cells. Applied Catalysis B **111-112**, 264-270 (2012).

Yang, F., M. Lublow, S. Orthmann, C. Merschjann, T. Tyborski, M. Rusu, S. Kubala, A. Thomas, R. Arrigo, M. Hävecker and T. Schedel-Niedrig: Metal-Free Photocatalytic Graphitic Carbon Nitride on p-Type Chalcopyrite as a Composite Photocathode for Light-Induced Hydrogen Evolution. ChemSusChem **5** (7), 1227-1232 (2012).

Yoong, Y.L.A., P.L. Yap, M.G. Kutty, O. Timpe, M. Behrens, S.B. Abd Hamid and R. Schlögl: Sorption profile of Hg(II) onto Mixed Phase of Copper Sulphide and Copper Sulphate. Advanced Materials Research **356-360**, 537-546 (2012).

Zafeiratos, S., S. Piccinin and D. Teschner: Alloys in catalysis: Phase separation and surface segregation phenomena in response to the reactive environment. Catalysis Science & Technology **2** (9), 1787-1801 (2012).

Zander, S., B. Seidlhofer and M. Behrens: In situ EDXRD study of the chemistry of aging of co-precipitated mixed Cu,Zn hydroxycarbonates – consequences for the preparation of Cu/ZnO catalysts. Dalton Transactions **41** (43), 13413-13422 (2012).

Zeitler, E.: Chicago exploration days. Ultramicroscopy **123**, 13-21 (2012).

Zhang, B.S., W. Zhang and D.S. Su: Analysis of Particle Size Distribution of Supported Catalyst by HAADF-STEM. Microscopy and Analysis **26** (2), 15-21 (2012).

Zhang, C., F. Yan, B.C. Bayer, R. Blume, M.H. Van der Veen, R. Xie, G. Zhong, B. Chen, A. Knop-Gericke, R. Schlögl, B.D. Capraro, S. Hofmann and J. Robertson: Complementary metal-oxide-semiconductor-compatible and self-aligned catalyst formation for carbon nanotube synthesis and interconnect fabrication. Journal of Applied Physics **111** (6), 064310 (2012).

Zhang, G.-R., D.S. Zhao, Y.-Y. Feng, B.S. Zhang, D.S. Su, G. Liu and B.-Q. Xu: Catalytic Pt-on-Au Nanostructures: Why Pt Becomes More Active on Smaller Au Particles. ACS Nano **6** (3), 2226-2236 (2012).

Zhang, J., R. Wang, E. Liu, X. Gao, Z. Sun, F.-S. Xiao, F. Girgsdies and D.S. Su: Spherical Structures Composed of Multiwalled Carbon Nanotubes: Formation Mechanism and Catalytic Performance. Angewandte Chemie International Edition **51** (30), 7581-7585 (2012).

Zhang, J., R. Wang, E. Liu, X. Gao, Z. Sun, F.-S. Xiao, F. Girgsdies and D.S. Su: Sphärische Partikel aus mehrwandigen Kohlenstoff-Nanoröhren: Bbildungsmechanismus und katalytische Leistung. Angewandte Chemie **124** (30), 7699-7704 (2012)

Zhang, M., J. Guan, B.S. Zhang, D.S. Su, C.T. Williams and C. Liang: Chemical Vapor Deposition of $Pd(C_3H_5)(C_5H_5)$ to Synthesize Pd@MOF-5 Catalysts for Suzuki Coupling Reaction. Catalysis Letters **142** (3), 313-318 (2012).

Zhao, X.-C., Q. Zhang, C. Chen, B.S. Zhang, S. Reiche, A. Wang, T. Zhang, R. Schlögl and D.S. Su: Aromatic sulfide, sulfoxide, and sulfone mediated mesoporous carbon monolith for use in supercapacitor. Nano Energy **1** (4), 624-630 (2012).

Zhao, X.-C., Q. Zhang, B.S. Zhang, C. Chen, A. Wang, T. Zhang and D.S. Su: Dual-heteroatom-modified ordered mesoporous carbon: Hydrothermal functionalization, structure, and its electrochemical performance. Journal of Materials Chemistry **22** (11), 4963-4969 (2012).

Zhu, H., Y. Xu, B.-L. Gu and W. Duan: Robust linear dependence of thermal conductance on radial strain in carbon nanotubes. New Journal of Physics **14** (1), 013053 (2012).

Doctoral Theses

Korup, O.: Methane Oxidation on Platinum Catalysts Investigated by Spatially Resolved Methods. Technische Universität Berlin 2012.

Li, H.: Determination of Oxygen Functionality on Highly Oriented Pyrolytic Graphite (HOPG). Freie Universität Berlin 2012.

Ota, A.: Synthesis, Characterization and Application of intermetallic Pd-X (Ga, Zn) Nanoparticles derived from ternary Hydrotalcite-like precursors. Technische Universität Berelin 2012.

Reiche, S.: Insights into heterogeneous catalysts for the HMF synthesis from biomass. Technische Universität Berlin 2012.

Vogel, D.: Local reaction kinetics of CO oxidation on heterogeneous Platinum, Palladium and Palladium oxide surfaces. Technische Universität Wien 2012.

Zander, S.: Preparation and characterization of Cu/ZnO catalysts for methanol synthesis. Technische Universität Berlin 2012.

Bachelor Thesis

Junk, T.: Charakterisierung von siliziumbasierten Anoden für Lithium-Ionen-Batterien. Freie Universität Berlin 2012.

2013

Abate, S., M. Freni, R. Arrigo, M.E. Schuster, S. Perathoner and G. Centi: On the Nature of Selective Palladium-Based Nanoparticles on Nitrogen-Doped Carbon Nanotubes for the Direct Synthesis of H_2O_2. ChemCatChem **5** (7), 1899-1905 (2013).

Amakawa, K., Y.V. Kolen'ko, A. Villa, M.E. Schuster, L.-I. Csepei, G. Weinberg, S. Wrabetz, R. Naumann d'Alnoncourt, F. Girgsdies, L. Prati, R. Schlögl and A. Trunschke: Multifunctionality of Crystalline MoV(TeNb) M1 Oxide Catalysts in Selective Oxidation of Propane and Benzyl Alcohol. ACS Catalysis **3**, 1103-1113 (2013).

Amakawa, K., L. Sun, C. Guo, M. Hävecker, P. Kube, I.E. Wachs, S. Lwin, A.I. Frenkel, A. Patlolla, K. Hermann, R. Schlögl and A. Trunschke: How Strain Affects the Reactivity of Surface Metal Oxide Catalysts. Angewandte Chemie, in press.

Amakawa, K., L. Sun, C. Guo, M. Hävecker, P. Kube, I.E. Wachs, S. Lwin, A.I. Frenkel, A. Patlolla, K. Hermann, R. Schlögl and A. Trunschke: How Strain Affects the Reactivity of Surface Metal Oxide Catalysts. Angewandte Chemie International Edition, in press.

Arkhangel'skii, I.V., A.V. Dunaev, I.V. Makarenko, N.A. Tikhonov, S.S. Belyaev and A. Tarasov: Non-Isothermal Kinetic Methods. Edition Open Access, Berlin (2013). ISBN 978-3-8442-4693-3.

Armbrüster, M., M. Behrens, B. Engendahl, M. Oezaslan, M. Rose and C. Sprung: German Catalysis, Celebrated in Weimar. ChemCatChem **5** (6), 1297-1298 (2013).

Armbrüster, M., M. Behrens, K. Föttinger, M. Friedrich, É. Gaudry, S.K. Matam and H.R. Sharma: The Intermetallic Compound ZnPd and its Role in Methanol Steam Reforming. Catalysis Reviews: Science and Engineering **55** (3), 289-367 (2013).

Arrigo, R., M.E. Schuster, S. Abate, S. Wrabetz, K. Amakawa, D. Teschner, M. Ferni, G. Centi, S. Perathoner, M. Hävecker, R. Schlögl: Dynamics of Palladium on nanocarbon in the direct synthesis of H_2O_2. ChemSusChem, in press.

Arrigo, R., M. Hävecker, M.E. Schuster, C. Ranjan, E. Stotz, A. Knop-Gericke and R. Schlögl: In Situ Study of the Gas-Phase Electrolysis of Water on Platinum by NAP-XPS. Angewandte Chemie International Edition **52** (44), 11660-11664 (2013).

Arrigo, R., M. Hävecker, M.E. Schuster, C. Ranjan, E. Stotz, A. Knop-Gericke and R. Schlögl: In-situ-Studie der Gasphasen-Wasserelektrolyse auf Platin mittels NAP-XPS. Angewandte Chemie **125** (44), 11874-11879 (2013).

Artyushkova, K., B. Kiefer, B. Halevi, A. Knop-Gericke, R. Schlögl and P. Atanassov: Density functional theory calculations of XPS binding energy shift for nitrogen-containing graphene-like structures. Chemical Communications **49** (25), 2539-2541 (2013).

Barbosa, R.L., V. Papaefthimiou, Y.T. Law, D. Teschner, M. Hävecker, A. Knop-Gericke, R. Zapf, G. Kolb, R. Schlögl and S. Zafeiratos: Methanol Steam Reforming over Indium-Promoted Pt/Al_2O_3 Catalyst: Nature of the Active Surface. The Journal of Physical Chemistry C **117** (12), 6143-6150 (2013).

Bayer, B.C., C. Castellarin-Cudia, R. Blume, S.A. Steiner, C. Ducati, D. Chu, A. Goldoni, A. Knop-Gericke, R. Schlögl, C. Cepek, J. Robertson and S. Hofmann: Tantalum-oxide catalysed chemical vapour deposition of single- and multi-walled carbon nanotubes. RSC Advances **3** (12), 4086-4092 (2013).

Behrens, M. and A.K. Datye: Catalysis for the conversion of biomass and its derivates. Edition Open Access, Berlin (2013). ISBN 978-3-8442-4282-9.

Behrens, M., G. Lolli, N. Muratova, I. Kasatkin, M. Hävecker, R. Naumann d'Alnoncourt, O. Storcheva, K. Köhler, M. Muhler and R. Schlögl: The effect of Al-doping on ZnO nanoparticles applied as catalyst support. Physical Chemistry Chemical Physics **15** (5), 1374-1381 (2013).

Behrens, M. and R. Schlögl: How to Prepare a Good Cu/ZnO Catalyst or the Role of Solid State Chemistry for the Synthesis of Nanostructured Catalysts. Zeitschrift für anorganische und allgemeine Chemie, in press.

Behrens, M., S. Zander, P. Kurr, N. Jacobsen, J. Senker, G. Koch, T. Ressler, R.W. Fischer and R. Schlögl: Performance Improvement of Nano-Catalysts by Promoter-Induced Defects in the Support Material: Methanol Synthesis over Cu/ZnO:Al. Journal of the American Chemical Society **135** (16), 6061-6068 (2013).

Checa, A.G., J.T. Bonarski, M.G. Willinger, M. Faryna, K. Berent, B. Kania, A. González-Segura, C.M. Pina, J. Pospiech and A. Morawiec: Crystallographic orientation inhomogeneity and crystal splitting in biogenic calcite. Journal of the Royal Society Interface **10** (86), 20130425 (2013).

Cheng, L., C. Yin, F. Mehmood, B. Liu, J. Greeley, S. Lee, B. Lee, S. Seifert, R. Winans, D. Teschner, R. Schlögl, S. Vajda and L. Curtiss: Reaction Mechanism for Direct Propylene Epoxidation by Alumina-Supported Silver Aggregates: The Role of the Particle / Support Interface. ACS Catalysis, in press.

Cotter, T.P., B. Frank, W. Zhang, R. Schlögl and A. Trunschke: The impact of V Doping on the Carbothermal synthesis of Mo Carbides. Chemistry of Materials **25** (15), 3124-3136 (2013).

Deutsch, C., H. Detz, T. Zederbauer, A.M. Andrews, P. Klang, T. Kubis, G. Klimeck, M.E. Schuster, W. Schrenk, G. Strasser and K. Unterrainer: Probing scattering mechanisms with symmetric quantum cascade lasers. Optics Express **21** (6), 7209-7213 (2013).

Dinse, A., T. Wolfram, C. Carrero, R. Schlögl, R. Schomäcker and K.-P. Dinse: Exploring the structure of paramagnetic centers in SBA-15 supported vanadia catalysts with pulsed one- and two-dimensional Electron Paramagnetic Resonance (EPR) and Electron Nuclear Double Resonance (ENDOR). The Journal of Physical Chemistry C **117** (33), 16921-16932 (2013).

Doll, T., J.J. Velasco-Velez , D. Rosenthal, J. Avila and V. Fuenzalida: Direct observation of the electroadsorptive effect on ultrathin films for microsensor and catalytic-surface control. ChemPhysChem **14** (11), 2505-2510 (2013).

Eichelbaum, M., R. Glaum, M. Hävecker, K. Wittich, C. Heine, H. Schwarz, C.-K. Dobner, C. Welker-Nieuwoudt, A. Trunschke and R. Schlögl: Towards Physical Descriptors of Active and Selective Catalysts for the Oxidation of n-Butane to Maleic Anhydride. ChemCatChem **5** (8), 2318-2329 (2013).

Farra, R., M. Eichelbaum, R. Schlögl, L. Szentmiklósi, T. Schmidt, A.P. Amrute, C. Mondelli, J. Pérez-Ramírez and D. Teschner: Do observations on surface coverage-reactivity correlations always describe the true catalytic process? A case study on ceria. Journal of Catalysis **297**, 119-127 (2013).

Farra, R., M. García-Melchor, M. Eichelbaum, M. Hashagen, W. Frandsen, J. Allan, F. Girgsdies, L. Szentmiklósi, N. Lopez and D. Teschner: Promoted ceria: A structural, catalytic and computational study. ACS Catalysis **3** (10), 2256-2268 (2013).

Farra, R., F. Girgsdies, W. Frandsen, M. Hashagen, R. Schlögl and D. Teschner: Synthesis and catalytic performance of CeOCl in Deacon reaction. Catalysis Letters **143** (10), 1012-1017 (2013).

Farra, R., S. Wrabetz, M.E. Schuster, E. Stotz, N. Hamilton, A.P. Amrute, J. Pérez-Ramírez, N. López and D. Teschner: Understanding CeO_2 as a Deacon catalyst by probe molecule adsorption and in situ infrared characterisations. Physical Chemistry Chemical Physics **15** (10), 3454-3465 (2013).

Frank, B., K. Friedel, F. Girgsdies, X. Huang, R. Schlögl and A. Trunschke: CNT supported Mo_xC catalysts: Impact of loading and carburization parameters. ChemCatChem **5** (8), 2296-2305 (2013).

Frank, B., M. Hävecker, W. Zhang, R. Schlögl and A. Trunschke: The Impact of Carbon Dynamics on the Molybdenum Carbide Surface during Catalytic Propane Dehydrogenation. Chemistry - A European Journal, in press.

Frank, B., R. Schlögl and D.S. Su: Diesel Soot Toxification. Environmental Science and Technology **47** (7), 3026-3027 (2013).

Frank, B., M.E. Schuster, R. Schlögl and D.S. Su: Emission aktivierter Rußpartikel: die Kehrseite der Medaille moderner Dieselmotoren. Angewandte Chemie **125** (10), 2736-2741 (2013).

Frank, B., M.E. Schuster, R. Schlögl and D.S. Su: Emission of Highly Activated Soot Particulate—The Other Side of the Coin with Modern Diesel Engines. Angewandte Chemie International Edition **52** (10), 2673-2677 (2013).

Frank, B., Z. Xie and A. Trunschke: Higher Alcohol Synthesis: Product Analysis using the Concept of Effective Carbon Numbers. Chemie-Ingenieur-Technik **85** (8), 1290-1293 (2013).

Friedrich, M., S.A. Villaseca, L. Szentmiklósi, D. Teschner and M. Armbrüster: Order-Induced Selectivity Increase of $Cu_{60}Pd_{40}$ in the Semi-Hydrogenation of Acetylene. Materials **6** (7), 2958-2977 (2013).

Geske, M., O. Korup and R. Horn: Resolving kinetics and dynamics of a catalytic reaction inside a fixed bed reactor by combined kinetic and spectroscopic profiling. Catalysis Science & Technology **3** (1), 169-175 (2013).

Goldsmith, C.F., A.S. Tomlin and S.J. Klippenstein: Uncertainty propagation in the derivation of phenomenological rate coefficients from theory: A case study of n-propyl radical oxidation. Proceedings of the Combustion Institute **34** (1), 177-185 (2013).

Gruber, M. and K. Hermann: Elementary steps of the catalytic NO_x reduction with NH_3: Cluster studies on adsorbate diffusion and dehydrogenation at vanadium oxide substrate. The Journal of Chemical Physics **138** (19), 194701 (2013).

Gruber, M. and K. Hermann: Elementary steps of the catalytic NO_x reduction with NH_3: Cluster studies on reactant adsorption at vanadium oxide substrate. The Journal of Chemical Physics **138** (9), 094704 (2013).

Guczi, L., Á. Molnár and D. Teschner: Hydrogenation Reactions: Concepts and Practice. In: Comprehensive Inorganic Chemistry, Vol. 2. (Eds.) J. Reedijk and K. Poeppelmeier. Elsevier, Oxford, 421-457 (2013). ISBN 0-08-097774-X.

Heine, C., F. Girgsdies, A. Trunschke, R. Schlögl and M. Eichelbaum: The model oxidation catalyst α-V_2O_5: Insights from contactless in situ microwave permittivity and conductivity measurements. Applied Physics A **112** (2), 289-296 (2013).

Huang, X., M. Wang, L. Shao, M.G. Willinger, C.S. Lee and X.-M. Meng: Polarity-Free Epitaxial Growth of Heterostructured ZnO/ZnS Core/Shell Nanobelts. The Journal of Physical Chemistry Letters **4** (5), 740-744 (2013).

Itkis, D.M., D.A. Semenenko, E.Y. Kataev, A.I. Belova, V.S. Neudachina, A.P. Sirotina, M. Hävecker, D. Teschner, A. Knop-Gericke, P. Dudin, A. Barinov, E.A. Goodilin, Y. Shao-Horn and L.V. Yashina: Reactivity of carbon in lithium-oxygen battery positive electrodes. Nano Letters **13** (10), 4697-4701 (2013).

Jentoft, F.C., J. Kröhnert, I.R. Subbotina, and, V.B. Kazansky: Quantitative Analysis of IR Intensities of Alkanes Adsorbed on Solid Acid Catalysts. The Journal of Physical Chemistry C **117** (11), 5873-5881 (2013).

Kaichev, V.V., A.Y. Gladky, I.P. Prosvirin, A.A. Saraev, M. Hävecker, A. Knop-Gericke, R. Schlögl and V.I. Bukhtiyarov: In situ XPS study of self-sustained oscillations in catalytic oxidation of propane over nickel. Surface Science **609**, 113-118 (2013).

Kandemir, T., F. Girgsdies, T.C. Hansen, K.-D. Liss, I. Kasatkin, E.L. Kunkes, G. Wowsnick, N. Jacobsen, R. Schlögl and M. Behrens: In Situ Study of Catalytic Processes: Neutron Diffraction of a Methanol Synthesis Catalyst at Industrially Relevant Pressure. Angewandte Chemie International Edition **52** (19), 5166-5170 (2013).

Kandemir, T., F. Girgsdies, T.C. Hansen, K.-D. Liss, I. Kasatkin, E.L. Kunkes, G. Wowsnick, N. Jacobsen, R. Schlögl and M. Behrens: In-situ-Untersuchung von katalytischen Prozessen bei industriell relevanten Drücken: Neutronenbeugung an einem Methanolsynthesekatalysator. Angewandte Chemie **125** (19), 5271-5276 (2013).

Kandemir, T., I. Kasatkin, F. Girgsdies, S. Zander, S. Kühl, M. Tovar, R. Schlögl and M. Behrens: Microstructural and defect analysis of metal nanoparticles in functional catalysts by diffraction and electron microscopy: The Cu/ZnO catalyst for methanol synthesis. Topics in Catalysis, in press.

Kandemir, T., M.E. Schuster, A. Senyshyn, M. Behrens and R. Schlögl: The Haber–Bosch Process Revisited: On the Real Structure and Stability of "Ammonia Iron" under Working Condition. Angewandte Chemie International Edition, in press.

Kandemir, T., M.E. Schuster, A. Senyshyn, M. Behrens and R. Schlögl: Über die Realstruktur von "Ammoniakeisen" und ihre Stabilität während des Haber-Bosch-Verfahrens. Angewandte Chemie, in press.

Khavryuchenko, O., B. Frank, A. Trunschke, K. Hermann and R. Schlögl: Quantum-chemical investigation of hydrocarbon oxidative dehydrogenation over spin-active carbon catalyst clusters. The Journal of Physical Chemistry C **117** (12), 6225-6234 (2013).

Kidambi, P.R., B.C. Bayer, R. Blume, Z.-J. Wang, C. Baehtz, R.S. Weatherup, M.G. Willinger, R. Schlögl and S. Hofmann: Observing Graphene Grow: Catalyst-Graphene Interactions during Scalable Graphene Growth on Polycrystalline Copper. Nano Letters **13** (10), 4769-4778 (2013).

Klemm, E., L. Greiner and R. Horn: Reaktionstechnik - Von der Synthese zum Prozess. Chemie-Ingenieur-Technik **85** (4), 391 (2013).

Korup, O., C.F. Goldsmith, G. Weinberg, M. Geske, T. Kandemir, R. Schlögl and R. Horn: Catalytic partial oxidation of methane on platinum investigated by spatial reactor profiles, spatially resolved spectroscopy, and microkinetic modeling. Journal of Catalysis **297**, 1-16 (2013).

Kozlowski, J.T., M. Behrens, R. Schlögl and R.J. Davis: Influence of Precipitation Method on Acid-Base Catalyzed Reactions over Mg-Zr Mixed Oxides. ChemCatChem **5** (7), 1989-1997 (2013).

Kreikemeyer-Lorenzo, D., W. Unterberger, R. Blume, M. Hävecker, T. Rocha, A. Knop-Gericke, R. Schlögl, T.J. Lerotholi, D.A. Duncan and D.P. Woodruff: Quantitative adsorbate structure determination under catalytic reaction conditions. Physical Review B **87** (12), 125420 (2013).

Kühl, S., A. Tarasov, S. Zander, I. Kasatkin and M. Behrens: Cu-based Catalyst resulting from Cu,Zn,Al Hydrotalcite-like Compound - microstructural, thermoanalytical and in-situ XAS study. Chemistry - A European Journal, in press.

Kunkes, E.L. and M. Behrens: Methanol Chemistry. In: Chemical Energy Storage, (Ed.) R. Schlögl. De Gruyter, Berlin, 413-442 (2013). ISBN 978-3-11-026407-4.

Li, G., L. Jiang, B. Zhang, Q. Jiang, D.S. Su and G. Sun: A highly active porous Pt-PbO_x/C catalyst toward alcohol electro-oxidation in alkaline electrolyte. International Journal of Hydrogen Energy **38** (29), 12767-12773 (2013).

Lorenz, H., C. Rameshan, T. Bielz, N. Memmel, W. Stadlmayr, L. Mayr, Q. Zhao, S. Soisuwan, B. Klötzer and S. Penner: From Oxide-Supported Palladium to Intermetallic Palladium Phases: Consequences for Methanol Steam Reforming. ChemCatCham **5** (6), 1273-1285 (2013).

Lunkenbein, T.: Das 63. Nobelpreisträgertreffen in Lindau 2013. Chemie in unserer Zeit **47** (4), 209 (2013).

Maganas, D., M. Roemelt, M. Hävecker, A. Trunschke, A. Knop-Gericke, R. Schlögl and F. Neese: First principles calculations of the structure and V L-edge X-ray absorption spectra of V_2O_5 using local pair natural orbital coupled cluster theory and spin-orbit coupled configuration interaction approaches. Physical Chemistry Chemical Physics **15** (19), 7260-7276 (2013).

Marichy, C., P.A. Russo, M. Latino, J.-P. Tessonnier, M.G. Willinger, N. Donato, G. Neri and N. Pinna: Tin dioxide-carbon heterostructures applied to gas sensing: Structure-dependent properties and general sensing mechanism. The Journal of Physical Chemistry C **117** (38), 19729-19739 (2013).

Mayr, L., H. Lorenz, M. Armbrüster, S.A. Villaseca, Y. Luo, R. Cardoso, U. Burkhardt, D. Zemlyanov, M. Hävecker, R. Blume, A. Knop-Gericke, B. Klötzer and S. Penner: The catalytic properties of thin film Pd-rich $GaPd_2$ in methanol steam reforming. Journal of Catalysis, in press.

Mette, K., S. Kühl, H. Düdder, K. Kähler, A. Tarasov, M. Muhler and M. Behrens: Stable Performance of Ni-Catalysts in Dry Reforming of Methane at High Temperatures for an Efficient CO_2-Conversion into Syngas. ChemCatChem, in press.

Moser, M., C. Mondelli, T. Schmidt, F. Girgsdies, M.E. Schuster, R. Farra, L. Szentmiklósi, D. Teschner and J. Pérez-Ramírez: Supported CeO_2 catalysts in technical form for sustainable chlorine production. Applied Catalysis B **132–133**, 123-131 (2013).

Niemantsverdriet, J.W. and R. Schlögl: Heterogeneous Catalysis: Introduction. In: Comprehensive Inorganic Chemistry, Vol. 2. (Eds.) J. Reedijk and K. Poeppelmeier. Elsevier, Oxford, 1-6 (2013). ISBN 0-08-097774-X.

Oh, M.H., T. Yu, S.-H. Yu, B. Lim, K.-T. Ko, M.G. Willinger, D.-H. Seo, B.H. Kim, M.G. Cho, J.-H. Park, K. Kang, Y.-E. Sung, N. Pinna and T. Hyeon: Galvanic Replacement Reactions in Metal Oxide Nanocrystals. Science **340** (6135), 964-968 (2013).

Ota, A., E.L. Kunkes, J. Kröhnert, M. Schmal and M. Behrens: Particle size effect in methane activation over supported palladium nanoparticles. Applied Catalysis A **452**, 203-213 (2013).

Papaefthimiou, V., I. Florea, W. Baaziz, I.M. Janowska, D. Begin, W.H. Doh, R. Blume, A. Knop-Gericke, O. Ersen, C. Pham-Huu and S. Zafeiratos: Effect of the Specific Surface Sites on the Reducibility of α-Fe_2O_3/Graphene Composites by Hydrogen. The Journal of Physical Chemistry C **117** (39), 20313-20319 (2013).

Papaefthimiou, V., M. Shishkin, D.K. Niakolas, M. Athanasiou, Y.T. Law, R. Arrigo, D. Teschner, M. Hävecker, A. Knop-Gericke, R. Schlögl, T. Ziegler, S.G. Neophytides and S. Zafeiratos: On the Active Surface State of Nickel-Ceria Solid Oxide Fuel Cell Anodes During Methane Electrooxidation. Advanced Energy Materials **3** (6), 762-769 (2013).

Patera, L., C. Africh, R. Weatherup, R. Blume, S. Bhardwaj, C. Castellarin-Cudia, A. Knop-Gericke, R. Schlögl, G. Comelli, S. Hofmann and C. Cepek: In Situ Observations of the Atomistic Mechanisms of Ni Catalyzed Low Temperature Graphene Growth. ACS Nano **7** (9), 7901-7912 (2013).

Parapat, R.Y., M. Wijaya, M. Schwarze, S. Selve, M.G. Willinger and R. Schomäcker: Particle shape optimization by changing from an isotropic to an anisotropic nanostructure: preparation of highly active and stable supported Pt catalysts in microemulsions. Nanoscale **5** (2), 796-805 (2013).

Rodemerck, U., M. Holeňa, E. Wagner, Q. Smejkal, A. Barkschat and M. Baerns: Catalyst Development for CO_2 Hydrogenation to Fuels. ChemCatChem **5** (7), 1948-1955 (2013).

Russo, P.A., S. Lima, V. Rebuttini, M. Pillinger, M.G. Willinger, N. Pinna and A.A. Valente: Microwave-assisted coating of carbon nanostructures with titanium dioxide for the catalytic dehydration of D-xylose into furfural. RSC Advances **3** (8), 2595-2603 (2013).

Schlögl, R.: Chemical Energy Storage. De Gruyter, Berlin (2013). ISBN 978-3-11-026632-0.

Schlögl, R.: The Solar Refinery. In: Chemical Energy Storage, (Ed.) R. Schlögl. De Gruyter, Berlin, 1-34 (2013). ISBN 978-3-11-026407-4.

Schwach, P., M.G. Willinger, A. Trunschke and R. Schlögl: Methane Coupling over Magnesium Oxide: How Doping Can Work. Angewandte Chemie International Edition **52** (43), 11381-11384 (2013).

Schwach, P., M.G. Willinger, A. Trunschke and R. Schlögl: Methane Coupling over Magnesium Oxide: How Doping Can Work. Angewandte Chemie **125** (43), 11591-11594 (2013).

Shao, L., B.S. Zhang, W. Zhang, S.Y. Hong, R. Schlögl and D.S. Su: Einfluss der Dynamik von Palladium in der Oberflächenkatalyse von Kupplungsreaktionen. Angewandte Chemie **125** (7), 2168-2171 (2013).

Shao, L., B.S. Zhang, W. Zhang, S.Y. Hong, R. Schlögl and D.S. Su: The Role of Palladium Dynamics in the Surface Catalysis of Coupling Reactions. Angewandte Chemie International Edition **52** (7), 2114-2117 (2013).

Starr, D.E., Z. Liu, M. Hävecker, A. Knop-Gericke and H. Bluhm: Investigation of solid/vapor interfaces using ambient pressure X-ray photoelectron spectroscopy. Chemical Society Reviews **42** (13), 5833-5857 (2013).

Su, D.S.: Carbon Nanotubes and Related Carbonaceous Structures. In: Nanomaterials in catalysis, (Eds.) P. Serp and K. Philippot. Wiley-VCH, Weinheim, 331-374 (2013). ISBN 978-3-527-33124-6.

Su, D.S., S. Perathoner and G. Centi: Nanocarbons for the Development of Advanced Catalysts. Chemical Reviews **113** (8), 5782-5816 (2013).

Tarasov, A., S. Kühl, J. Schumann and M. Behrens: Thermokinetic study of the reduction process of a $CuO/ZnAl_2O_4$ catalyst. High Temperatures-High Pressures **42** (4), 377-386 (2013).

Trunschke, A.: Synthesis of Solid Catalysts. In: Chemical Energy Storage, (Ed.) R. Schlögl. De Gruyter, Berlin, 277-310 (2013). ISBN 978-3-11-026407-4.

Vogel, D., Z. Budinska, C. Spiel, R. Schlögl, Y. Suchorski and G. Rupprechter: Silicon Oxide Surface Segregation in CO Oxidation on Pd: An in situ PEEM, MS and XPS Study. Catalysis Letters **143** (3), 235-240 (2013).

Vogel, D., C. Spiel, M. Schmid, M. Stöger-Pollach, R. Schlögl, Y. Suchorski and G. Rupprechter: The Role of Defects in the Local Reaction Kinetics of CO Oxidation on Low-Index Pd Surfaces. The Journal of Physical Chemistry C **117** (23), 12054-12060 (2013).

Wang, D., A. Villa, D.S. Su, L. Prati and R. Schlögl: Carbon-Supported Gold Nanocatalysts: Shape Effect in the Selective Glycerol Oxidation. ChemCatChem **5** (9), 2717-2723 (2013).

Wang, L., H. Wang, W. Zhang, J. Zhang, J.P. Lewis, X. Meng and F.-S. Xiao: Aerobic homocoupling of phenylboronic acid on Mg–Al mixed-oxides-supported Au nanoparticles. Journal of Catalysis **298**, 186-197 (2013).

Wang, L., B. Zhang, W. Zhang, J. Zhang, X. Gao, X. Meng, D.S. Su and F.-S. Xiao: Positively charged bulk Au particles as an efficient catalyst for oxidation of styrene with molecular oxygen. Chemical Communications **49** (33), 3449-3451 (2013).

Wang, T., J. Jelic, D. Rosenthal and K. Reuter: Exploring Pretreatment–Morphology Relationships: Ab Initio Wulff Construction for RuO_2 Nanoparticles under Oxidising Condition. ChemCatChem **5** (11), 3398-3403 (2013).

Weatherup, R.S., C. Baehtz, B. Dlubak, B.C. Bayer, P.R. Kidambi, R. Blume, R. Schlögl and S. Hofmann: Introducing Carbon Diffusion Barriers for Uniform, High-Quality Graphene Growth from Solid Sources. Nano Letters **13** (10), 4624-4631 (2013).

Wei, L., Z.-Y. Zhou, S.-P. Chen, C.-D. Xu, D.S. Su, M.E. Schuster and S.-G. Sun: Electrochemically shape-controlled synthesis in deep eutectic solvents: triambic icosahedral platinum nanocrystals with high-index facets and their enhanced catalytic activity. Chemical Communications **49** (95), 11152-11154 (2013).

Wowsnick, G., D. Teschner, M. Armbrüster, I. Kasatkin, F. Girgsdies, Y. Grin, R. Schlögl and M. Behrens: Surface dynamics of the intermetallic catalyst Pd_2Ga, Part II – Reactivity and stability in liquid-phase hydrogenation of phenylacetylene. Journal of Catalysis, in press.

Wowsnick, G., D. Teschner, I. Kasatkin, F. Girgsdies, M. Armbrüster, A. Zhang, Y. Grin, R. Schlögl and M. Behrens: Surface dynamics of the intermetallic catalyst Pd_2Ga, Part I – Structural stability in UHV and different gas atmospheres. Journal of Catalysis, in press.

Xia, Z., S. Wang, Y. Li, L. Jiang, H. Sun, S. Zhu, D.S. Su and G. Sun: Vertically oriented polypyrrole nanowire arrays on Pd-plated Nafion® membrane and its application in direct methanol fuel cells. Journal of Materials Chemistry A **1** (3), 491-494 (2013).

Yashina, L.V., J. Sánchez-Barriga, M.R. Scholz, A.A. Volykhov, A.P. Sirotina, V.S. Neudachina, M.E. Tamm, A. Varykhalov, D. Marchenko, G. Springholz, G. Bauer, A. Knop-Gericke and O. Rader: Negligible Surface Reactivity of Topological Insulators Bi_2Se_3 and Bi_2Te_3 towards Oxygen and Water. ACS Nano **7** (6), 5181-5191 (2013).

Zander, S., E.L. Kunkes, M.E. Schuster, J. Schumann, G. Weinberg, D. Teschner, N. Jacobsen, R. Schlögl and M. Behrens: The Role of the Oxide Component in the Development of Copper Composite Catalysts for Methanol Synthesis. Angewandte Chemie International Edition **52** (25), 6536-6540 (2013).

Zander, S., E.L. Kunkes, M.E. Schuster, J. Schumann, G. Weinberg, D. Teschner, N. Jacobsen, R. Schlögl and M. Behrens: Die Rolle der Oxidkomponente für die Entwicklung von Kupfer-Komposit-Katalysatoren zur Synthese von Methanol. Angewandte Chemie **125** (17), 6664-6669 (2013)

Zemlyanov, D., B. Klötzer, H. Gabasch, A. Smeltz, F.H. Ribeiro, S. Zafeiratos, D. Teschner, P. Schnörch, E.M. Vass, M. Hävecker, A. Knop-Gericke and R. Schlögl: Kinetics of Palladium Oxidation in the mbar Pressure Range: Ambient Pressure XPS Study. Topics in Catalysis **56** (11), 885-895 (2013).

Zhang, B., L. Shao, W. Zhang and D.S. Su: Clothing carbon nanotubes with palladium rings: Constructing carbon-metal hybrid nanostructures under electron-beam irradiation. ChemCatChem **5** (9), 2581-2585 (2013).

Zhang, B.S., W. Zhang, L. Shao and D.S. Su: Optimum Energy-Dispersive X-Ray Spectroscopy Elemental Mapping for Advanced Catalytic Materials. ChemCatChem **5** (9), 2586-2590 (2013).

Zheng, W., T.P. Cotter, P. Kaghazchi, T. Jacob, B. Frank, K. Schlichte, W. Zhang, D.S. Su, F. Schüth and R. Schlögl: Experimental and Theoretical Investigation of Molybdenum Carbide and Nitride as Catalysts for Ammonia Decomposition. Journal of the American Chemical Society **135** (9), 3458-3464 (2013).

Doctoral Thesis

Amakawa, K.: Active site for propene metathesis in silica-supported molybdenum oxide catalysts. Technische Universität Berlin 2013.

Kandemir, T.: Solid catalysts for methanol and ammonia synthesis investigated by in-situ neutron diffraction. Technische Universität Berlin 2013.

Mavlyankariev, S.: Investigation of Gas-Phase Methane Oxidation by Reactor Profile Measurements and Microkinetic Modeling. Technische Universität Berlin 2013.

Wowsnick, G.: Surface dynamics of Pd_2Ga and its reactivity in the liquid phase hydrogenation of phenylacetylene. Technische Universität Berlin 2013.

Master's Thesis

Homburg, T.: Metal-support-interaction of nanostructured Ag/ZnO catalysts. Carl von Ossietzky Universität Oldenburg 2013.

Invited Talks of the Members of the Department of Inorganic Chemistry

Rosa Arrigo

Apr 2013	Seminar zur Oberflächenforschung, Institut für Physikalische und Theoretische Chemie, University of Bonn, Bonn, Germany *In Situ Study of Water Electrolysis by Means of near Ambient Pressure X-Ray Photoemission Spectroscopy (NAP-XPS)*

Malte Behrens

Oct 2011	GÖCh-Vortrag, University of Innsbruck, Innsbruck, Austria *Vom Volumen an die Oberfläche: Festkörper- und Grenzflächenchemie von Cu/ZnO Methanolsynthese- und Methanoldampfreformierungskatalsylatoren*
Apr 2012	Euroschool on Intermetallic Compounds in Catalysis, MPI for Chemical Physics of Solids, Dresden, Germany *Synthesis of Intermetallic Catalysts*
Apr 2012	Workshop, Katalytische Materialien, University of Duisburg-Essen, Essen, Germany *Metallnanopartikel in der Katalyse*
May 2012	Festkörperchemisches und -analytisches Kolloquium, Technische Universität Berlin, Berlin, Germany *Vom Festkörper zum Klimaschutz - Materialchemie für maßgeschneiderte Kupferkatalysatoren zur Hydrierung von* CO_2 *zu Methanol*
Jun 2012	2nd Ertl Symposium on Surface and Interface Chemistry, Stuttgart, Germany *Nanostructured Catalysts for Energy Conversion*
Oct 2012	GDNÄ-Tageskongress Energiewende wohin? Berlin, Germany *Optionen für eine nachhaltige Rohstoffbasis für die Energieversorgung und für die chemische Industrie*
Dec 2012	Symposium für Anorganische Chemie, University of Ulm, Ulm, Germany *Materialchemie von nanostrukturierten Katalysatoren zur chemischen Energiespeicherung*
Mar 2013	46. Jahrestreffen Deutscher Katalytiker, Weimar, Germany *On the Active Site of Methanol Synthesis*
May 2013	AC-Kolloquium, University of Bayreuth, Bayreuth, Germany *Catalysts for* CO_2 *Conversion: Materials Chemistry of Nanostructured Cu and Ni Composites*
May 2013	HNL-Vortrag, Ruhr Universität Bochum, Bochum, Germany *Metal-Support-Interaction of Cu/ZnO: Insights into the Methanol Synthesis Catalyst from Materials Chemistry*
Jun 2013	AC-Kolloquium, University of Marburg, Marburg, Germany *Materialchemie von Feststoffkatalysatoren zur Umsetzung von* CO_2

Jul 2013	PC-Kolloquium, University of Ulm, Ulm, Germany *Development of Cu/ZnO-Based Catalysts for the Hydrogenation of* CO_2 *to Methanol: From Industrial Practice to New Materials*
Jul 2013	Seminar, University of Duisburg-Essen, Duisburg, Germany *Materialchemie von Feststoffkatalysatoren zur Umsetzung von* CO_2
Sep 2013	SUNCAT Seminar, Stanford University, Stanford, CA, USA *On the Role of the Second Metal in Cu- and Pd-Based Bi-Metallic Hydrogenation Catalysts*
Sep 2013	246th ACS National Meeting and Exposition, Indianapolis, IN, USA *Cu/ZnO Catalyst Development for* CO_2 *Hydrogenation to Methanol*

Maik Eichelbaum

Dec 2011	Schulvortrag am Gymnasium, 9.Klassenstufe, Chemie- und Physiklehrer, Jessen/Sachsen-Anhalt, Germany *Im Jahre 2011: Internationales Jahr der Chemie, 100 Jahre physikalisch-chemische Forschung am Fritz-Haber-Institut in Berlin, 100 Jahre Nobelpreis in Chemie für Marie Curie: Wie die Chemie unser Leben verändert hat, und was sie uns für die Zukunft bereit hält*
Dec 2012	Seminar, Department of Physics (AG Robert Bittl) Freie Universität Berlin, Berlin, Germany *The Intimate Relationship between Charge Transfer Properties and Catalytic Performance of Vanadium Oxide Catalysts Studied by Microwave Techniques under Reaction Conditions*
Jun 2013	23rd North American Catalysis Society Meeting, Louisville, KY, USA *The Relation between Interfacial Charge Transport and Performance of Oxidation Catalysts Studied with In Situ Microwave Techniques*
Jun 2013	7th World Congress on Oxidation Catalysis, Saint Louis, MO, USA *Microwave Conductivity Measurements in a Resonant Cavity: A Powerful In Situ Technique for Elucidating the Relevance of Interfacial Charge Transfer in Selective Oxidation Catalysis*

Klaus Hermann

Apr 2012	Seminar, Fachbereich Chemie, University of Potsdam, Potsdam, Germany *Theoretical X-Ray Spectroscopy with DFT Cluster Methods: Studies for Systems of Catalytic Relevance*
Apr 2012	Kolloquium, Faculty of Physics, University of Marburg, Marburg, Germany *Theoretical X-Ray Spectroscopy: DFT Cluster Studies for Systems of Catalytic Relevance*
May 2012	12th deMon Developers Workshop, Shanghai, China *Theoretical NEXAFS Studies Using StoBe and deMon2k: Structural Analysis of Silica-Supported Metal Oxide Catalysts*

Jul 2012	Seminar, Beijing Computational Science Research Center, BCSRC, Beijing, China *Theoretical X-Ray Spectroscopy: DFT Cluster Studies for Systems of Catalytic Relevance*
Aug 2012	Seminar, Chemical Department, Tsinghua University, Beijing, China *Theoretical NEXAFS Studies for Structural Analysis of Silica-Supported Transition Metal Oxide Catalysts*
Aug 2012	244th ACS National Meeting and Exposition, Division of Catalysis Science and Technology, Philadelphia, PA, USA *Structural Analysis of Silica-Supported Transition Metal Oxide Catalysts by X-Ray Absorption Spectroscopy: Theoretical Cluster Studies*
Aug 2012	Seminar, Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA, USA *Periodic Overlayers and Interference Lattices: Moirons at Work*
Sep 2012	Seminar, Physics Department, Pennsylvania State University, State College, PA, USA *Periodic Overlayers and Interference Lattices: Moirons at Work*
Jan 2013	8th International Conference on Computational Physics, ICCP-8, Hongkong, China *Computational Modeling of Electronic Core Excitation*
Jan 2013	8th International Conference on Computational Physics, ICCP-8, Hongkong, China *Computational Approaches to Structure, Binding, and Reaction at Solid Surfaces*
Jan 2013	Seminar, Institute of Computational and Theoretical Studies, Baptist University, Hongkong, China *Structural Analysis of Silica-Supported Transition Metal Oxide Catalysts by X-Ray Absorption: Theoretical Cluster Studies*
Jun 2013	5th International Conference on Energy Dissipation at Surfaces, SFB 616, Bad Honnef, Germany *Periodic Overlayers and Interference Lattices: Moirons at Work*
Jul 2013	International Symposium on Functional Materials and Their Theoretical Simulations, FMTS2013, Chengdu, China *Silica-Supported Transition Metal Oxide Particles in Catalysis: Theoretical X-Ray Absorption Analysis with DFT Methods*
Jul 2013	Seminar, Key Laboratory of Quantum Information, University of Science and Technology of China, Hefei, China *Theoretical X-Ray Spectroscopy: DFT Cluster Studies for Systems of Catalytic Relevance*
Jul 2013	Seminar, MPI for Chemical Energy Conversion, Mühlheim a.d. Ruhr, Germany *Theoretical X-Ray Spectroscopy: DFT Cluster Studies for Systems of Catalytic Relevance*

Raimund Horn

Jan 2012	Kolloquium, Ernst-Berl-Institut für Technische und Makromolekulare Chemie, Technische Universität Darmstadt, Darmstadt, Germany *Catalytic and Non-Catalytic Methane Oxidation at High Temperatures: Mechanistic Insight by Combination of In Situ Experiments and Kinetic Simulations*
May 2012	Jahrestreffen Reaktionstechnik, Würzburg, Germany *Einführung der Posterbeiträge des Jahrestreffens Reaktionstechnik 2012*
Nov 2012	Seminar, Fraunhofer Institute for Ceramic Technologies and Systems IKTS, Dresden, Germany *Catalytic Partial Oxidation of Methane on Ceramic Foam Catalysts*
Dec 2012	Seminar, Bayer Technology Services, Leverkusen, Germany *Catalytic and Non-Catalytic Methane Oxidation at High Temperatures: Mechanistic Insight by Combination of In Situ Experiments and Reactor Simulations*
Feb 2013	Jahrestreffen Hochtemperaturtechnik und Technische Reaktionen, Oberhausen, Germany *Heterogen katalysierte Hochtemperatur-Reaktionen*
Apr 2013	Festsymposium zum 50. Jubiläum des Instituts für Chemische Verfahrenstechnik, University of Stuttgart, Stuttgart, Germany *Heterogene Katalyse - Vom aktiven Zentrum zum Reaktor*
May 2013	Seminar, Shell Technology Center Amsterdam, Amsterdam, The Netherlands *A Novel Tool for Understanding and Optimizing Heterogeneous Catalytic Reactions under Industrial Conditions*

Timur Kandemir

May 2013	Spezialseminar, Department of Chemistry, Group of Applied Physical Chemistry, Freie Universität Berlin, Berlin, Germany *Catalysts at Work: Comparative In Situ Neutron Diffraction Studies under Industrial Synthesis Conditions*

Axel Knop-Gericke

Nov 2011	Humboldt-Kolleg, Goa, India *The Application of near Ambient Pressure X-Ray Photoelectron Spectroscopy to Study Heterogeneous Catalytic Processes*
May 2012	Seminar, Elettra: the Synchrotron Light Source, Trieste, Italy *Near Ambient Pressure X-Ray Photoelectron Spectroscopy: Recent Applications*
Jun 2012	BIT's 1st Annual World Congress of Advanced Materials, Peking, China *Characterization of Heterogeneous Catalytic Processes by near Ambient X-Ray Photoelectron Spectrscopy*
Oct 2012	Seminar, Utrecht University, Utrecht, The Netherlands *Oxygen Species on Silver Catalysts in Ethylene Epoxidation Reaction*

Nov 2012	Conference on Dynamic Behaviour of Silver Surfaces in Heterogeneous Catalytic Oxidation Reactions, Shanghai, China *Dynamic Behaviour of Silver Surfaces in Heterogeneous Catalytic Oxidation Reactions*
Feb 2013	Seminar, Helmholtz-Zentrum Berlin, Berlin, Germany *State and Outlook of near Ambient Pressure XPS Studies of Catalytic Processes at BESSY*
Sep 2013	Workshop, National Synchrotron Radiation Research Center, Hsinchun, Taiwan *Near Ambient Pressure X-Ray Photoelectron Spectroscopy in Heterogeneous Catalysis and Energy Research*
Oct 2013	15th European Conference on Applications of Surface and Interface Analysis (ECASIA13), Fort Village, Sardinia, Italy *Recent Application of near Ambient Pressure X-Ray Photoelectron Spectroscopy in Studies of Oxygen Species in Heterogeneous Catalytic Reactions*

Katrin Quetting

Nov 2011	Herbsttagung der Bibliotheken, CPT-Sektion, Berlin, Germany *Weblog@FHILibrary*
Nov 2012	Herbsttagung der Bibliotheken, CPT-Sektion, Saarbrücken, Germany *We_blog: Einfach loslegen mit dem Bibliotheksblog*

Robert Schlögl

Sep 2011	SPD-Ortsverein, Unterföhring, Germany *Die Energiewende - Grundlagen und Einsichten für eine politische Diskussion: Wissenschaft als "Bonusmaterial"*
Oct 2011	OktoberMusikFest, Bayrische Staatsoper, Munich, Germany *Die Energiewende: Wunsch und Wirklichkeit*
Oct 2011	Workshop on New Materials for Renewable Energy, Trieste, Italy *Heterogeneous Catalysis for Chemical Energy Storage: The Largest Challenge for Catalysis Ever?*
Oct 2011	Workshop on New Materials for Renewable Energy, Trieste, Italy *Material Challenges in Electro-Catalysis*
Nov 2011	Falling Walls Conference, Berlin, Germany *Energy Challenge: The Wall of Chemical Energy Storage: Novel Chemistry at the Centre Stage*
Nov 2011	The Royal Society Discussion Meeting on 'Can solar power deliver?' London, UK *The Role of Chemistry in the Renewable Energy Challenge - Is Chemistry Really Needed?*
Nov 2011	9th Nanovision, Institute of Nanotechnology, Karlsruhe, Germany *The Role of Material Chemistry in the Renewable Energy Challenge - Is Chemistry Really Needed?*

Nov 2011	Workshop on Power-to-Gas: Erdgasinfrastruktur als Energiespeicher, Bundesnetzagentur und Fraunhofer-Institut IWES, Berlin, Germany *The Role of Material Chemistry in the Renewable Energy Challenge - Is Chemistry Really Needed?*
Nov 2011	CO_2 Symposium der RWTH on Innovation in Large-Volume CO_2 recycling with Sustainable Energy Sources: Science, Politics and Business Opportunities, RWTH Aachen University, Aachen, Germany *Power to Gas: Opportunities and Challenges as for Today: Is Chemistry Really Needed?*
Nov 2011	MRS Fall Meeting, Material Research Society, Boston, MA, USA *Low-Temperature Carbon Materials for Energy Storage Processes*
Dec 2011	Workshop on Surfaces and Catalysis - The Contribution of Chemistry in Piedmont, Torino, Italy *Heterogeneous Catalysis for Chemical Energy Storage: The Largest Challenge for Catalysis Ever?*
Dec 2011	AGEF-Seminar on Electrocatalysis and Photocatalysis, Ruhr Universität Bochum, Bochum, Germany *Material Challenges in Electro-Catalysis*
Dec 2011	Vorstellungen zur Arbeit im Themennetzwerk Energie acatech- National Academy of Science and Engeneering, Berlin, Germany *Die Energiewende: Was hat der Ammoniak damit zu tun?*
Jan 2012	Evonik Industries AG, Marl, Germany *Challenges in Electro-Catalysis. Sustainable Hydrogen?*
Jan 2012	GÖCh-Vortrag, University of Duisburg-Essen, Essen, Germany *Chemical Energy Storage. Is Chemistry Really Needed?*
Jan 2012	Wissenschaftlicher Verein, Mönchengladbach, Germany *Die Energiewende: Wunsch und Wirklichkeit*
Feb 2012	2nd International Symposium on Advanced Electron Microscopy for Catalysis and Energy Storage Materials, Berlin, Germany *Expectations of Catalysis on Electron Microscopy - More than Pretty Pictures?*
Feb 2012	Institute of Polymer Chemistry, University of Stuttgart, Stuttgart, Germany *Chemical Energy Storage - After the first Rush of "Wende"*
Feb 2012	Wissenschaftliches Kolloquium anlässlich der Verabschiedung von Dr. Walter Braun, Helmholtz-Zentrum Berlin, Berlin, Germany *Synchrotronstrahlung in der Energieforschung - Geht es ohne?*
Apr 2012	Closing Symposium, Collaborative Research Centre SFB 558 on Metal-Support Interactions - How Important are they in Heterogeneous Catalysis, Electro- and Photo-Catalysis?, Bochum, Germany *Functional Analysis of the Cu/ZnO System*
Apr 2012	Physikalisches Kolloquium, Ruhr Universität Bochum, Bochum, Germany *Wie kontrollieren die Volumeneigenschaften eines Festkörpers seine Oberflächenreaktionen?*

Apr 2012	European Energy Conference - E2C, Maastricht, The Netherlands *Heterogeneous Reactions for Energy Storage - ENERCHEM: A New Pardigm in Research and Technology*
Apr 2012	Joint European C-MAC and COST CM0904 EuroSchool Intermetallic Compounds in Catalysis, Dresden, Germany *Hetrogeneous Catalysis with IMC*
Apr 2012	Deutsche Telekom-Stiftung, Bad Honnef, Germany *Energiewende - wohin?*
May 2012	4th International Congress on Operando Spectroscopy, Upton, NY, USA *From Static Model Systems to Dynamical High Performance Catalysts*
May 2012	SFB Seminar on Functional Oxide Surfaces and Interfaces (FOXSI), Vienna, Austria *Complex Metal Oxides as Heterogeneous Catalysts*
May 2012	UniCat, Technische Universität Berlin, Berlin, Germany *Solid State Chemistry and Catalysis: Structure-Function Relations?*
May 2012	3rd International Symposium on Surface Imaging / Spectroscopy at the Solid/Liquid Interface, Krakow, Poland *Structures of Interfaces Relevant in Chemical Energy Conversion*
May 2012	7th International Green Energy Conference and The 1st DNL Conference on Clean Energy, Dalian, China *The Impact of Catalysis on the Energy Challenge*
Jun 2012	Besichtigungsprogramm der Physikalischen Gesellschaft zu Berlin, Berlin, Germany *Keine Energiewende ohne Katalyse*
Jun 2012	2nd Ertl Symposium on Surface and Interface Chemistry, Stuttgart, Germany *Material Dynamics of OER Electrodes*
Jun 2012	The Conference on Carbon in Catalysis, Carbocat V, Brixen, Italy *Material Dynamics of OER Electrodes*
Aug 2012	Topsøe Catalysis Forum - Microscopy in Catalysis, Gilleleje, Denmark *Bridging the Material Gap in Heterogeneous Catalysis - The Role of Microscopy in the Concert of Synthesis and Characterization*
Sep 2012	2. Kooperationsveranstaltung des Forums ElektroMobilität e.V. und CMS Hasche Sigle, Berlin, Germany *Regenerative Energiesysteme und Speicher*
Sep 2012	GDCh-Vortrag Bayer, Leverkusen, Germany *Grenzflächenprozesse bei Elektrochemischen Oxidationsreaktionen: Dynamische Funktionsmaterialien*
Sep 2012	Herbstschule für Teilchenphysik, Kloster Maria Laach, Germany *Teller oder Tank?*
Sep 2012	BASF Research Seminar, St. Martin, Germany *Heterogeneous Catalysis - From Black Magic to Rational Design*

Sep 2012	UOP LLC, a Honeywell Company, Des Plaines, IL, USA *From Static Model Systems to Dynamical High Performance Catalysts*
Sep 2012	Symposium on The Future of Catalysis, SLAC National Accelerator Laboratory, Stanford University, Menlo Park, CA, USA *Why We Need In Situ Observation of Catalysts*
Oct 2012	VW Wolfsburg, Wolfsburg, Germany *Elektrochemie und Speicher*
Nov 2012	Shell Technology Center Amsterdam, Amsterdam, The Netherlands *The Role of Size Effects in Catalysis*
Nov 2012	International Symposium on Advanced Synchrotron Techniques for Energy and Catalysis Applications, Shanghai, China *Synchrotron Radiation in Heterogeneous Catalysis*
Nov 2012	Basic Lecture Program - Berlin International Graduate School of Natural Sciences and Engineering, Technische Universität Berlin, Berlin, Germany *Catalyst Characterization: A Challenge in Material Science*
Nov 2012	Arbeitsgemeinschaft Chemie and Energie, Karlsruhe Institute of Technology, Karlsruhe, Germany *Energiewende: Eine Herausforderung für die Chemie*
Nov 2012	6. Biomasse-Forum, Bad Hersfeld, Germany *Offene technische Herausforderungen von Power to Gas*
Nov 2012	Acatech Portfolio-Konferenz, Munich, Germany *Energiesysteme für die Zukunft*
Dec 2012	DECHEMA Infotag Kleine Moleküle als Energieträger, Frankfurt a. Main, Germany *Activation of Small Molecules - A Key Challenge for Advanced Energy Systems*
Jan 2013	Ringvorlesung WS 2012/13: Lecture Series Catalysis, Berlin, Germany *Catalysis for Energy Systems*
Jan 2013	Kolloquium der GÖCh, University of Innsbruck, Innsbruck, Austria *Katalysatoren auf die Finger geschaut - Wie wir heute heterogene Katalyse verstehen*
Jan 2013	Industrie-Club, Düsseldorf, Germany *Energiewende - wohin?*
Jan 2013	MPI-Lectures, University of Duisburg-Essen, Essen, Germany *Heterogeneous Catalysis (is Always "Nanocatalysis")*
Jan 2013	Physikalisches Kolloquium der Universität, University of Duisburg-Essen, Duisburg, Germany *Active Sites in Heterogeneous Catalysis*
Jan 2013	GDCh Vortrag, Ruhr Universität Bochum, Bochum, Germany *Carbon*
Jan 2013	Physik am Samstagvormittag, University of Bayreuth, Bayreuth, Germany *Energiewende? Wunsch und Wirklichkeit*

Feb 2013	Einweihung des NanoEnergieTechnikZentrums NETZ, Duisburg, Germany *Kleine Teilchen für große Aufgaben: Nanoparticles in Energy Science*
Feb 2013	Klassensitzung Berlin-Brandenburg Academy of Sciences and Humanities, Berlin, Germany *Energiewende? Wunsch und Wirklichkeit*
Mar 2013	6th IDECAT/ERIC-JCAT Conference (IEJCat-6) on Design Advanced Multifunctional Catalysts for Sustainable Processes, Brixen, Italy *Design and Dynamics of Heterogeneous Catalysts*
Mar 2013	Festsymposium 125 Jahre Angewandte Chemie, Berlin, Germany *Chemistry and Energy*
Mar 2013	BDEW/BMBF-Forschungssymposium, Berlin, Germany *Energiesysteme der Zukunft*
Mar 2013	Netherlands' Catalysis and Chemistry Conference NCCC-14, Noordwijkerhout, The Netherlands *In Situ Analysis of Working Catalysts: Understanding of Heterogeneous Catalysis and a Priori Models*
Apr 2013	34. Internationales Wiener Motorensymposium, Vienna, Austria *Mobilität in der Energiewende: Betrachtungen eines Chemikers zu Ausrichtungen über längere Zeiträume*
May 2013	Forschungspressekonferenz BASF, Chemistry Powers Energy - Regenerativ erzeugen, sicher speichern, effizient übertragen und nutzen, Ludwigshafen, Germany *Chemistry Enables Sustainable Energy: Keine Energiewende ohne Chemie*
May 2013	Ministry of Science,Technology and Innovation, Putrajaya, Malaysia *The Role of Chemistry in the Energy Challenge - Systemic View*
May 2013	University of Malaysia, Kuala Lumpur, Malaysia *How to Rationally Design Heterogeneous Catalysts*
May 2013	Wissenschaftliches Kolloquium, DLR Oberpfaffenhofen, Weßling, Germany *Energiewende - wohin?*
May 2013	GDCh-Kolloquium, Chemische Institute Bonn-Endenich, Bonn, Germany *Was wir über heterogene Katalysatoren heute verstehen*
Jun 2013	Institute of Metal Research Chinese Academy of Sciences, Shenyang, China *Nanocarbon: Surface Analysis Strategies*
Jul 2013	Workshop on The Future of Energy Technology, Max-Planck-Haus, Munich, Germany *Systemic Approach Towards the Energy System: The Critical Role of Chemistry*
Jul 2013	2. BHC-Symposium, Kloster Banz, Bad Staffelstein, Germany *Chemical Energy Storage: Status and Perspectives*
Jul 2013	OCM Meeting, Gerhard Ertl Center, Technische Universität Berlin, Berlin, Germany *Functional Insights into Earth-Alkali Catalyzed OCM: Status and Perspectives*

Aug 2013	Telluride Meeting Theory and Practice of Catalysis, Telluride, Collorado, USA *Dynamics in Heterogeneous Catalysis*
Aug 2013	2013 Microscopy and Microanalysis Annual Meeting, Indianapolis, IN, USA *Mesoscopic Aspects in Heterogeneous Catalysis: Dynamics at a Supramolecular Scale*
Aug 2013	2013 Microscopy and Microanalysis Annual Meeting, Indianapolis, IN, USA *Pt Nanoparticles as Electrocatalysts for Water Splitting: Atomistic Insights into Functions and Stability*
Aug 2013	SUNCAT Summer Institute 2013, SLAC National Accelerator Laboratory, Menlo Park, CA, USA *Active Sites in Heterogeneous Catalysis*
Sep 2013	GDCh-Wissenschaftsforum CHEMIE 2013, Darmstadt, Germany *Wie wir große Mengen Energie in kleinen Molekülen speichern können: Ein Statusbericht*
Sep 2013	Vollversammlung des Akademienprojektes “Energiesysteme der Zukunft”, Berlin, Germany *Energiesysteme der Zukunft*
Sep 2013	Rotary Club Berlin, Berlin, Germany *Systemic Approach Towards the Energy System: The Critical Role of Chemistry*
Sep 2013	246th ACS National Meeting and Exposition “Frontiers in Energy Conversion and Fuel Production”, Indianapolis, Indiana, USA *The Role of Chemistry in the Energy Challenge*
Sep 2013	5th International Conference on Carbon for Energy Storage/Conversion, and Environment Protection, Mülheim a.d. Ruhr, Germany *Why Carbon Materials for Energy Application?*
Sep 2013	5th ANKA/KNMF Users' Meeting 2013, Bruchsal, Germany *NAP-XPS: A Critical Tool in the Energy Challenge*
Sep 2013	Lecture, IMPRS, Berlin, Germany *Dynamics in Heterogeneous Catalysis: A Buzzword or a Fundamental Concept? A Look from the Side of the Catalyst*
Oct 2013	52. gat - Gasfachliche Aussprachetagung, Nuremberg, Germany *Ingenieure, Impulse, Innovation: Die Schnittstelle von Elektrizität und Chemie als strategische Aufgabe für nachhaltige Energiesysteme*
Oct 2013	Branchendialog, Die Photovoltaikbranche in Sachsen-Anhalt - Entwicklung, Trends und Ausblick, Bitterfeld-Wolfen, Germany *Ausbaustand und Rolle der Photovoltaik im Energiemix der Zukunft*
Oct 2013	Acatech Festveranstaltung, Konzerthaus, Berlin, Germany *Die Energiewende gestalten - Für unsere Energieversorgung in der Zukunft*

Oct 2013	SFB 1073 Symposium, Atomic Scale Control of Energy Conversion, University of Göttingen, Göttingen, Germany *The Role of Electron Microscopy in Understanding Heterogeneous Catalytic Processes*

Annette Trunschke

Mar 2012	243rd ACS National Meeting and Exposition, San Diego, CA, USA *Engineering the Synthesis of MoVTeNb Oxide Catalysts with High Stability in Oxidation of Propane*
Aug 2012	244th ACS National Meeting and Exposition, Symposium on Spectroscopy and Microscopy in Catalysis: In-situ Age, Philadelphia, PA, USA *Adding Dynamic to Electron Spectroscopy: Evolution of Active Catalyst Surfaces Studied by In Situ X-Ray Photoelectron Spectroscopy and Soft X-Ray Absorption Spectroscopy*

Marc Georg Willinger

Jan 2012	Lecture, Department of Ceramics and Material Science, University of Aveiro, Aveiro, Portugal *Morphological Dynamics of Metallic Systems Studied by Environmental SEM*
May 2012	Seminar, Institute of Electron Microscopy and Nanoanalysis, Graz University of Technology, Graz, Austria *Observation of Dynamic Processes on the Surface of Metallic Systems by Environmental SEM*
Sep 2012	European Microscopy Congress, Symposium: In Situ and Environmental EM, Manchester, UK *In Situ Observation of Graphene Growth and Dynamics of Metallic Systems by Environmental Scanning Electron Microscopy*
Nov 2012	Lecture, Institut Charles Gerhardt, Montpellier 1 University, Montpelliere, France *Inelastic Electron Scattering in the TEM: The Analytical Power of Energy Loss Spectrometry*
Jan 2013	Lecture, Department of Ceramics and Material Science, University of Aveiro, Aveiro, Portugal *Inelastic Interactions in the TEM: Advanced Imaging by Energy Filtered TEM and EELS Spectrometry*
Jan 2013	Seminar, Vienna University of Technology, Vienna, Austria *Dynamics in Metallic Systems and the Catalytic Deposition of Carbon Films Studied by In Situ Electron Microscopy*
Mar 2013	Seminar, Center for Electron Nanoscopy, Technical University, Lyngby, Denmark *In Situ Electron Microscopy Investigation of Dynamics in Metallic Systems and the Catalytic Deposition of Thin Carbon Films*

Apr 2013	Project Meeting, University of Twente, Enschede, Niederlande *In Situ Electron Microscopy Investigation of Dynamics in Metallic Systems and the Catalytic Deposition of Thin Carbon Films*
Jun 2013	Seminar, Institute of Physical Engineering, University of Technology, Brno, Czech Republic *Dynamics in Metallic Systems and the Catalytic Deposition of Carbon Films Studied by In Situ Electron Microscopy*

Sabine Wrabetz

Apr 2012	Lecture, Institut für Energietechnik, Technische Thermodynamik, Technische Universität Dresden, Dresden, Germany *Adsorption Microcalorimetry: Basics and Applications in Heterogeneous Catalysis*
Feb 2013	20. Ulm-Freiberger Kalorimetrietage, Freiberg, Germany *Adsorption Microcalorimetry in Heterogeneous Catalysis*

Department of Chemical Physics **Director: H.-J. Freund**

Staff scientists:

Dr. Markus Heyde	
Dr. Helmut Kuhlenbeck	(Habilitation)
Dr. Niklas Nilius	(Habilitation, until 03/31/2013)
Dr. Swetlana Schauermann	(Habilitation, since 09/01/2013 Minerva program)
Dr. Shamil Shaikhutdinov	
Dr. Martin Sterrer	(Habilitation)
Dr. Thomas Schmidt	

Guest scientists, staying for at least six months:

Dr. Serguei Adamovski	until 01/31/2013
Dr. Damien Cornu	until 09/30/2013
Dr. Kai Huang	until 02/28/2013
Dr. Helder Marchetto	
Prof. Niklas Nilius	until 10/31/2013
Dr. Casey O'Brien	
Dr. Yi Pan	
Dr. Hengshan Qiu	until 04/30/2013
Dr. Xiang Shao	until 03/31/2013
Prof. Dietrich Menzel	
Prof. Wolf-Dieter Schneider	
Dr. Bing Yang	until 05/31/2013

Scientists (temporary) paid from external funds:

Prof. Paul Bagus *AvH Awardee*	
Dr. Jorge Anibal Boscoboinik *AvH Fellow*	until 06/30/2013
Dr. Florencia Calaza *AvH Fellow*	
Dr. Yi Cui *AvH Fellow*	
Dr. Ahmed Ghalgaoui	until 12/31/2013
Dr. Irene Groot *AvH Fellow*	until 12/31/2012
Dr. William Kaden *AvH Fellow*	
Prof. Thomas Risse	
Dr. Fernando Stavale *AvH Fellow*	until 03/31/2013
Prof. Geoff Thornton *AvH Awardee*	

Graduate students:	17 (6 students paid from external funds) 12 IMPRS students
Trainees:	3
Diploma students:	4
Technicians:	10
PP&B Group:	Heinz Junkes (group leader) 6 Technicians, 1 Trainee, 1 Master student
Crystal Preparation Laboratory:	2 Technicians

Recent Developments in the Department of Chemical Physics

Director: H.-J. Freund

General Remarks

Since the last visit of the Fachbeirat the following changes occurred among the staff members of the department.

Dr. Niklas Nilius left the department to become Professor of Physics at University of Oldenburg. The activities will be continued within Dr. Markus Heyde´s group.

The associated group "Scanned-energy Mode Photoelectron Diffraction" headed by Prof. Phil Woodruff, was discontinued from 01.01.2012 on after the collaborative research center SFB 546 "Transition Metal Oxides" was terminated after 12 years of funding.

Prof. Wolf-Dieter Schneider started to work with the department, in particular the scanning probe microscopy groups, as a consultant, starting 01.12.2012.

Honors and Awards:

Hans-Joachim Freund

- Robert-Bunsen-Lecture 2012, German Bunsen Society for Physical Chemistry (DBG) (2012)
- Corresponding Member of the North Rhine-Westphalian Academy of Sciences, Humanities and the Arts (since 2012)
- Fellow of the European Academy of Sciences (2012)
- Dinesh O. Shah Lecture in Surface Science 2012, University of Florida, Chemical Engineering Department (2012)
- Blaise Pascal Medal in Material Science of the European Academy of Sciences (2012)
- Award for Service 2012 of the EuCheMS (2012)
- Member, European Academy of Sciences (EURASC) (since 2013)

- Corresponding Member of the Chemical Sciences Section and Honorary Member of the Hungarian Academy of Sciences (2013)
- Dr. Honoris Causa from Aix-Marseille Université (2013)
- Bernstein Lecturer, Department of Chemistry, University of Wisconsin-Madison, USA (2013-14)

Swetlana Schauermann

- 05/2012 – 05/2017 Dozentenstipendium des Fonds der Chemischen Industrie (Fellowship of the Association of Chemical Industry)
- 06/2013 ERC Starting Grant “Enantioselective Reactions on Model Chirally Modified Surfaces”
- 07/2013 Minerva Research Group in the scope of “Minerva Program” of the MPG

Dietrich Menzel

- Elettra Fellow 2011 (Sincrotrone Trieste, Dec. 2011)
- Gaede-Langmuir Award 2012 (American Vacuum Society, Oct. 2012)

Georg Simon

- Otto-Hahn Medal 2011

Leonid Lichtenstein

- Otto-Hahn Medal 2012

Members of the department have been involved in a number of other activities:

Hans-Joachim Freund

- Member, Supervisory Board, Karlsruhe Institut für Technologie (KIT) (2011-2012)
- Member, International Program Committee of the Millennium Science Initiative, Chile (since 2012)
- Member, Advisory Board of the Karl-Ziegler-Foundation of the German Chemical Society (since 2012)
- Member, Editorial Board of “Catalysis Reviews” (since 2012)
- Member, Strategy Board (Perspektivenkommission) of the CPTS (Chemical-Physical-Technical Section of the Max Planck Society) (since 2012)
- Member, International Selection Committee, Millennium Technology Prize, Technology Academy Finland (since 2013)
- Member, Scientific Advisory Committee, Indian Institute of Technology, Indore, India (since 2013)
- Member, Review Editorial Board “Frontiers in Condensed Matter Physics” (since 2013)
- Editor-in-Chief, “Catalysis Letters” and “Topics in Catalysis” (since 2013)
- Member, RIKEN Advisory Council (2012)

Dietrich Menzel

- Member, Scientific Advisory Committee of Elettra (Sincrotrone Trieste) (1998-2011)

Wolf-Dieter Schneider

- Associate Editor, Frontiers in Condensed Matter Physics (since 2013)

The following events have been organized by the department:

- Department Workshop, Döllnsee (2012)
- Faraday Discussion 162 “Fabrication, Structure and Reactivity of Anchored Nanoparticles” Berlin 2013 (with M. Bowker)
- Department Workshop, Hasenwinkel (2013)

Within the Department of Chemical Physics there are at present eight working groups:

1. Spectroscopy

   Helmut Kuhlenbeck
2. Structure and Reactivity

   Shamil Shaikhutdinov
3. Atomic Force Microscopy

   Markus Heyde
4. Scanning Probe Spectroscopy

   Niklas Nilius
5. Magnetic Resonance

   Thomas Risse (part time)
6. Catalysis/Laser Spectroscopy

   Martin Sterrer
7. Molecular Beam

   Swetlana Schauermann
8. Spectro-Microscopy (SMART)

   Thomas Schmidt

During the last two years we have continued to concentrate on structure and reactivity studies of oxide surfaces and ultrathin films, electronic structure, and adsorption on metal and oxide nanoparticles and instrumentation developments in the Department of Chemical Physics. There are several collaborations within the institute. Those include activities on nanoparticle oxide interfaces with the Department of Inorganic Chemistry, which has led to a draft of a common publication. A study on the influence of dopants in simple oxides on catalysis has led to two back-to-back papers in Angewandte Chemie, also with the Department of Inorganic Chemistry. Two collaborative projects with the Theory Department on Li doped MgO and Isophorone adsorption on Pd(111) which have led to publications. The Department has also participated in a number of projects funded from outside sources, which are listed below.

A particular project of importance has been the renewal of the "Cluster of Excellence" (CoE), "Unifying Concepts in Catalysis" (UniCat) funded by the DFG. The Department continues to represent the Institute in the Board of the CoE. Another important project for the Department has been the proposal to found another collaborative research centre (SFB 1109 "Understanding of Metal Oxide/ Water Systems at the Molecular Scale: Structural Evolution Interfaces, and Dissolution") together with the three Berlin Universities. The proposal has been favourable evaluated in 2013 and we are awaiting the final decision.

The complete list of collaborations encompasses:

- Collaboration H.-J. Freund, S. Shaikhutdinov:

  COST Action CM1104: "Reducible oxide chemistry, structure and functions"

- Collaboration H.-J. Freund, Th. Risse, N. Nilius:

  ESF COST Action D41: "Inorganic Oxides: Surfaces and Interfaces"

- Collaboration H.-J. Freund, M. Sterrer, S. Shaikhutdinov:

  SFB 1109: "Understanding of Metal Oxide / Water Systems at the Molecular Scale: Structural Evolution, Interfaces, and Dissolution"

- Collaboration H.-J. Freund and Th. Risse:

  Center of Excellence, UniCat, Teilprojekt D1: "Activation of methane"

  Center of Excellence, UniCat, Teilprojekt D2: "Chemical activation of carbon oxides"

Further collaborations include:

- Collaboration with Spain:

  S. Shaikhutdinov, N. Nilius and K.M. Neymann, University of Barcelona, Barcelona

  S. Schauermann and K.M. Neymann, University of Barcelona, Barcelona

- Collaboration with the UK:

  S. Shaikhutdinov and M. Bowker, School of Chemistry, Cardiff University, Cardiff

  T. Risse and F. Thornton, University College London, London

- Collaborations with the USA:

  H.-J. Freund, M. Sterrer, H. Kuhlenbeck and P.S. Bagus, University of North Texas, Denton

  H.-J. Freund, H. Kuhlenbeck and E. Bauer, Department of Physics, Arizona State University, Tempe

  M. Heyde and U. D. Schwarz, Yale University, New Haven

  H.-J. Freund and J.C. Hemminger, School of Physical Sciences, University of California

  H.-J. Freund, M. Heyde and S.T. Oyama, Virginia Polytechnic Institute and State University, Blacksburg

  H.-J. Freund and P.C. Stair, Northwestern University, Evanston

  H. Junkes and M. Davidsaver, Brookhaven National Laboratory, New York

  H. Junkes and S. B. Webb, Oak Ridge National Laboratory, Tennessee

  S. Schauermann and A. Gellman, Carnegie Mellon University, Pittsburgh

  S. Schauermann and B. Roldan Cuenya, University of Central Florida

  H.-J. Freund and I. Wachs, Lehigh University Bethlehem, Pennsylvania

- Collaboration with France:

  N. Nilius and J. Goniakowski, University of Aix-Marseille, Marseille

- Collaboration with Italy:

  H.-J. Freund, N. Nilius and G. Pacchioni, Università degli Studi Milano-Bicocca, Milano

  H.-J. Freund and E. Giamello, IFM, Universita degli studi di Torino, Torino

- Collaboration with China:

  H.-J. Freund and H. Gao, Chinese Academy of Sciences, Beijing

  H.-J. Freund and W. Huang, University of Science and Technology of China, Chinese Academy of Sciences, Hefei

- Collaboration with Brazil:

  H.-J. Freund and M. Schmal, Universidade Federal do Rio de Janeiro (UFRJ), Rio de Janeiro

  H.-J Freund and F. Stavale, The Brazilian Center for Physics Research, Centro Brasileiro de Pesquisas Fisicas (CBPF), Rio de Janeiro

- Collaboration with Finland:

  H.-J. Freund, N. Nilius, and H. Häkkinen, Nanoscience Center, University of Jyväskylä, Jyväskylä

- Collaboration with Hungary:

  H.-J. Freund and F. Solymosi, University of Szeged, Academy of Sciences, Szeged

- Collaboration with Japan:

  H.-J. Freund and K. Asakura, Catalysis Research Center, Hokkaido University, Sapporo

- Collaboration with South Korea:

H.-J. Freund and Jeong Park, Center for Nanomaterials and Chemical Reactions of Institute for Basic Science

- Collaboration within Germany:

  H. Junkes and R. Lange, HZB Berlin

The department has associated with it the PP&B group, and the Crystal Preparation Laboratory.

The following research highlights include results of research projects as well as on instrumental developments:

- We have for the first time atomically imaged and characterized a two-dimensional silica glass system on Ru(0001) (Angew. Chem. Int. Ed. 51, 404-407(2012)) as well as the structural transition between the ordered and the vitreous phase (Phys. Rev. Lett. 109, 106101(2012)). The results have been published partly in collaboration with Joachim Sauer´s group.

- It has been possible to extend the preparation of vitreous silica films from the Ru(0001) surface to Pt(111) (Appl. Phys. Lett. 100, 151608 (2012)). The trend in the formation of vitreous and ordered, bilayer and monolayer films, as we have prepared them over the last decade, has been established and discussed (Adv. Mater. 25, 49 (2013); Chem. Phys. Lett. 550, 1-7 (2012)).

- For the first time we have prepared and characterized a two-dimensional aluminosilicate system in an attempt to provide surface science input into zeolite chemistry in collaboration with Joachim Sauer´s group (Angew. Chem. Int. Ed. 51, 6005 (2012)).

- Within the same collaboration it was possible to prepare a model for a layered clay mineral (nontronite) on the basis of silica and ironoxide layers.

- A patterned defect structure has been observed on a silica monolayer (silicatene) film on Ru(0001), which has been predicted but never experimentally verified for graphene (Nano Lett. 13, 4422-4427 (2013)).

- In collaboration with Notker Rösch´s group (Theory) we have studied calorimetrically the heats of adsorption of CO (Phys. Chem. Chem. Phys. 13, 16800-16810 (2011)) and oxygen (Angew. Chem. Int. Ed. 52, 5175–5179 (2013)) on Fe_3O_4(111) supported Pd particles as a function of particle size. The comparison reveals for the first time the influence of low coordination sites on the particles and underlines the problems with temperature-programmed desorption studies on such systems.

- A large number of Au clusters prepared by evaporating Au onto a very thin MgO(100) film on Ag(100) have been studied by scanning tunnelling spectroscopy and the relation between energy level spacings and geometry, with regard to the question "at what size the gap closes", has been studied (Phys. Rev. B 88 (11), 115415-1-8 (2013)). These studies have been performed in collaboration with the theory group of Hannu Häkkinen, and Wolf-Dieter Schneider as a consultant.

- A summary on our activities on photochemistry and photophysics of supported metal nanoparticles has been published by our consultant Dietrich Menzel on the occasion of his Langmuir Award from the American Vacuum Society (J. Vac. Sci. Technol. A 31 (5), 050817-1-12).

- Preparation of model catalysts using wet impregnation has been studied using ambient pressure STM (Appl. Catal. A, doi:10.1016/j.apcata.2013.05.031 (2013)); Phys. Chem. Chem. Phys. 14, 11525-11533 (2012)), and directly compared with equivalent preparations using physical vapour deposition in ultrahigh vacuum.

- A detailed comparison of reactivity of five different ultrathin oxide films, completely covering the metal substrate, in CO oxidations has been reported and analysed in terms of their abilities to provide weakly bound oxygen species (ChemCatChem 5(8), 2162-2166 (2013)). For the least active ZnO(0001) films, compelling evidence was provided for the enhanced reactivity of ZnO/Pt(111) system on the oxide/metal interface (J. Catal. 301, 227 (2013)).

- We have studied CeO_2(111) surfaces with respect to the location of defects at step edges (ACS-Nano 6, 1126-1133 (2012)) in collaboration with theory from Konstantin Neyman´s group, and subsurface oxygen vacancies. The latter by correlation with Au atom decoration and in close collaboration with theory from Joachim Sauer´s group.

- Two major reviews have been written on oxide surfaces and thin films. One directly from our groups (Chem. Rev. 113, 3986-4034 (2013)) and a second one in co-authorship with Gianfranco Pacchioni (Chem. Rev. 113, 4035-4072 (2012)).

- The modified Auger-parameter concept has been applied to unravel initial and final state effects in XPS binding energy shifts for a metal atom at a buried metal oxide interface, where a straightforward interpretation would have led to wrong conclusions. This work has been carried out in collaboration with Paul Bagus and Gianfranco Pacchioni.

- The studies on the influence of dopants have been extended from the studies on the Au particle morphologies to the adsorption of O_2 on a CaO(100) surface with buried dopants (Angew. Chem. Int. Ed. 52, 11385-11387 (2013)). Our model studies have been paralleled by a study on real catalysts by the Department of Inorganic Chemistry (Angew. Chem. Int. Ed. 52, 11381-11384 (2013)) and published back to back.

- In a study on the competition between supported Au nanoparticles and adsorbed molecular oxygen for electrons from buried dopants we were able to identify a route to control nanoparticle shape by an oxygen atmosphere (Faraday Discuss. 162, 1-11(2013)).

- In order to characterize the nature of transition metal dopants, we have employed the photon-STM to excite dopant states within the bandgap (New J. Phys. 14, 033006-1-14 (2012)). In comparison with calculations from Gianfranco Pacchioni´s group assignments to the position and oxidation state of the dopant may be made (J. Am. Chem. Soc. 134, 11380-11383 (2012)).

- Progress has been made on the preparation of transition metal doped TiO_2 single crystal surfaces by homoepitaxy of TiO_2 films on TiO_2(110) single crystals which are protected by a barrier layer for diffusion (Top. Catal. 56 (15), 1389-1403 (2013)).

- Electron energy loss spectroscopy in the regime of electronic excitations has been used to unravel oxidation states on iron oxide surfaces, and their influence on deposited Au particles (Surf. Sci., submitted).

- Detailed studies have been undertaken to study the processes involved in the preparation of iron oxide surfaces on Pt(111). Also the interconversion of various Fe_3O_4, Fe_2O_3, and

FeO phases as a function of the conditions and its influence on the quality of the film have been studied (Phys. Rev. B 86, 155430 (2012)).

- The resolution of the PEEM using the SMART setup has been improved to 18 nm by including an aperture within the beam path (Ultramicroscopy 126, 23-32 (2013)). On the basis of this new concept for further improving the resolution have been developed which are now being implemented.
- High frequency EPR was shown to be able to discriminate different paramagnetic species in MgO films, which can be used to explain the unresolved asymmetric line shape observed at conventional EPR spectroscopy. Hence it shows the ability to gain additional insight onto these systems not possible with conventional EPR spectroscopy currently used.
- An instrument to combine local scanning probe measurements with temperature programmed desorption is being built.

Progress Reports

In the following, results from the various groups obtained during the last two years are summarized:

Spectroscopy Group

In the last years IV-LEED studies of MoO_3 and $V_2O_3(0001)$ on Au(111), HREELS studies of biphase Fe_2O_3 and gold clusters on biphase Fe_2O_3 on Pt(111), and TDS+STM studies of Mo+Ti and V+Ti mixed oxide layers on $TiO_2(110)$ were performed.

IV-LEED studies of $V_2O_3(0001)$ and MoO_3 on Au(111)

IV-LEED studies were performed with a modified version of the Barbieri/Van Hove Symmetrized Automated LEED package, combined with a search algorithm with some global search capability which permits to search in a somewhat larger space (as compared to Tensor LEED), and to make use of a parallel computing architecture. A LEED system with channel plate detection was used in order to minimize damage of the oxide by the electron beam.

IV-LEED studies of $V_2O_3(0001)$ on Au(111) were triggered by recent reports [Window et al, Phys. Rev. Lett. 107, 016105, 2011 and Seifert et al. Surf. Sci. 606, L41-L44, 2012] which questioned that the $V_2O_3(0001)$ surface is terminated by a layer of vanadyl groups as was assumed until then, based on vibrational spectroscopy and STM results. Based on ion scattering data the authors proposed that the oxide is terminated by the so-called O3 structure. $V_2O_3(0001)$ on different substrates has been the subject of a number of experimental and theoretical studies and therefore the surface termination is a relevant topic, especially for theoretical studies dealing with adsorption/reaction issues.

The measured IV-LEED data could be fitted with a Pendry R-factor of 0.13, assuming a vanadyl terminated structure, and with a Pendry R-factor of 0.23 for O_3 termination. Therefore, the present results indicate that the $V_2O_3(0001)$ surface is vanadyl terminated. However, while the energies of the measured structures fit well to the computed ones, the fit is somewhat worse for the intensities. The studies are going on at present in order to get a better fit for the intensities.

MoO_3 on Au(111) forms high-quality layers with a low defect density. At a thickness of more than 2 layers the oxide film assumes the structure of bulk α-$MoO_3(010)$. IV-LEED data were recorded for a monolayer thick film. Previous studies with DFT [Quek et al, Surf. Sci. 577, L71-L77, 2005] have indicated that the layer has structure similar to that of a single layer of α-$MoO_3(010)$. The IV-LEED studies confirmed this and located the Mo atoms in bridging positions on Au(111) (R-Pendry=0.05). The atomic positions are at variance with the results of the DFT study, which assumes that the Mo atoms are located in threefold hollow sites on the Au(111) surface.

HREELS studies of gold clusters on biphase $Fe_2O_3(0001)$ on Pt(111))

These studies were performed in order to characterize the catalytic activity of gold clusters on Fe_2O_3. The original intention was to study gold clusters on regular $Fe_2O_3(0001)$ but the preparation of the oxide substrate was hampered by molybdenum contaminations in the high-pressure cell. Therefore Fe_2O_3 with biphase termination was studied since this can be prepared at a lower oxygen pressure. HREELS was used to study the vibrational and the electronic excitations of the system. To cover a broader scope of properties the studies were complemented by TDS and PES investigations. Ab initio cluster calculations of excited Fe^{2+} and Fe^{3+} ion states in an oxidic surrounding were performed by Volker Staemmler (Ruhr-

Universität Bochum). For biphase Fe_2O_3 without gold it could be shown that this transforms into a slightly reduced phase by annealing at ~790 K. The reduction occurs solely below the surface; the surface structural and electronic properties are unchanged within the detection limits of the applied methods. Gold deposits on the oxygen rich surface are not active for CO oxidation while gold clusters are active, if they are deposited onto a reduced oxide layer, although the reduction occurs below the surface. This indicates that there is an interaction between the gold clusters and the oxygen vacancies below the surface. The interaction mechanism is not clear yet; one may think of some kind of charge donation to the gold clusters.

Structural studies and adsorption studies of Mo+Ti and V+Ti mixed oxide layers

Mo+Ti mixed oxide layers were prepared on $TiO_2(110)$ with the aim of investigating the role of the embedded Mo ions for chemical reactions on the surface. To this end layers with different Mo concentrations were prepared and studied with XPS and STM. Alcohol and water adsorbates were investigated in order to characterize the chemical surface activity. Ordered mixed oxide surface phases with distinct LEED patterns could not be found. Mo tends to agglomerate on the surface where it is found predominantly as Mo^{6+} at low coverages and as Mo^{4+} at high coverages. Mo^{4+} was also identified in the bulk of the mixed oxide layer.

Two types of Mo-induced features could be identified in the STM images for low Mo concentrations (in the range of 1%). At higher Mo concentrations (~50%) the surface is characterized by stripes with limited lengths in [001] direction. The concentration of bridging oxygen vacancies, which are common defects on $TiO_2(110)$, is reduced significantly even at low Mo concentrations. Methanol and ethanol TPD spectra reflect this effect by a decrease of the intensity of the features related to these surface defects. At elevated MoOx coverages, the yield of reaction products in methanol and ethanol TPD spectra are somewhat smaller than those found for clean $TiO_2(110)$ and the reactions occur at lower temperature. We assign this result (1) to a decrease of the activity related to regular $TiO_2(110)$ sites, and (2) to reactivity related to Mo induced sites with lower reaction barriers.

V+Ti mixed oxide layers were grown on $TiO_2(110)$ with a Ta+Ti mixed oxide layer between the overlayer and the substrate. The interlayer has the task to block the diffusion of vanadium into the $TiO_2(110)$ bulk. Reactions studies of alcohols were performed for layers with different vanadium concentrations and different degrees of reductions. Two reaction channels could be identified and aldehyde as well as alkane/alkene production was observed. At present STM studies are performed in order to correlate the reactivity with structural properties.

Structure and Reactivity Group

In the past two years the group has continued its work on fabrication of well-ordered thin silica films that are suited for experimental and theoretical studies of chemical reactions occurring on surfaces of silicates and related materials, such as zeolites. Another project was aimed at a deeper understanding of the structure and reactivity of metal supported ultrathin oxide films in oxidation reactions, with particular emphasis on CO oxidation at relatively low temperatures.

Preparation, growth and atomic structure of ultrathin silicate films on metals

The studies revealed a structural complexity and diversity of the silica overlayers on metals. The experimental results, complemented by DFT calculations (performed in the group of Prof. Sauer at HU Berlin), provided compelling evidence for the formation of a single-layer network of corner-sharing [SiO_4] tetrahedra, with a $SiO_{2.5}$ stoichiometric composition, on Mo(112) and Ru(0001) substrates. However, on Ru(0001) and Pt(111), it is possible to grow a bilayer SiO_2 film that is weakly bound to the metal surface. The results showed that the metal-oxygen bond strength plays the decisive role for the principal structure of the silica film. Metals with high oxygen adsorption energy favor the formation of crystalline monolayer films. Noble metal supports primarily form bilayer SiO_2 sheets. The metals with intermediate energies may form either of the structures. In addition, in contrast to monolayer films, the bilayer films exist in both crystalline and amorphous forms. These two-dimensional silica films provide unique opportunities to directly visualize the atomic structure of vitreous silica (in collaboration with the AFM group).

Besides these well-defined silica films can be used as model supports for metal and oxide clusters, the results opened new challenge for synthesis and surface science studies of structure and reactivity of zeolite-like surfaces and clay minerals. Introduction of Al into the silicate framework allowed us for the first time to fabricate ultrathin aluminosilicate films which upon hydroxylation expose surface resembling that of highly acidic zeolites. Doping a silicate film with Fe resulted in a layered (silica-iron oxide) structure that can be considered as a monolayer of Nontronite (the Fe-rich clay mineral).

CO oxidation over ultrathin transition metal oxide films supported by metals

A continuously growing body of experimental and theoretical results indicates that ultrathin oxide films, those below one nanometer in thickness when grown on transition metal, may exhibit interesting catalytic properties in their own right, which may not be observed on thicker films or respective single crystal surfaces. In particular, such films showed a higher CO oxidation rate than pure platinum group metal catalysts. In recent years, we have studied the catalytic properties of ultrathin films of transition metal oxides using the CO oxidation reaction at low temperatures and near-atmospheric pressures as a benchmark. The systems included ultrathin oxide films of Fe, Mn, Zn, and Ru, all on Pt(110) with the exception of Ru-Oxide on Ru(0001). The observed structure-reactivity relationships suggest that the oxygen binding energy in the films plays the decisive role for the CO oxidation reaction under these conditions. The more weakly bound the surface oxygen species is, the higher is the reaction rate which follows the sequence: Ru > Fe > Mn > Zn.

Closed ZnO films, i.e. films covering the surface without having access to the oxide metal interface, showed the lowest activity among the films studied. However, films only partially covering a Pt(111) support showed reaction rates by a factor of 5 – 7 higher than the closed film. Such an enhancement is rationalized in terms of the reactions occurring at the oxide/metal boundary. In addition, we have performed comparative specifically studies of CO oxidation over ZnO(0001) films supported by Pt(111), Ag(111) and Cu(111). The results permitted to formulate a structural model for elucidating the reaction mechanism on the so called “inverse model catalysts” at technologically relevant conditions.

Atomic Force Microscopy Group

The main focus has been put on the determination of surface structures. In our group, we have achieved atomic scale resolution on materials ranging from highly crystalline to amorphous. Local probe microscopy and spectroscopy can go beyond the averaging character of diffraction methods. Essential physical properties were extracted using noncontact atomic force microscopy (nc-AFM) and scanning tunneling microscopy (STM). In particular, surface structures and defects of various dimensionalities were investigated. The materials cover point defects, line defects, misfit dislocations and two-dimensional (2D) random networks, i.e. amorphous structures. One project focuses on morphological imaging and spectroscopy data taken on thin MgO films grown on Mo(001) and a new approach to directly combine these measurements with reactivity studies, while the other project is concentrated on the analysis of a bilayer silica film system as well as doping and adsorption studies on this new material class.

Combined Structure and Reactivity Studies of MgO Films Supported on Mo(001)

Distance and bias dependent nc-AFM and STM measurements were recorded on these films. The local work function shift and electronic structure of misfit induced dislocation lines in the MgO surface were measured. By using local spectroscopy techniques, which are complementary to averaging methods show the appearance of electronic properties in the real space landscape was shown. The scanning probe measurements should ideally be accompanied by reactivity studies, since detailed understanding of the structure-reactivity relationship of the same sample system is desired. Therefore, we are setting up a new temperature programmed desorption (TPD) experiment. The influence of the support and controlled doping on the properties of the thin oxide films will be investigated in our combined setup.

Silica Film Structures from Crystalline to Amorphous Ring Networks

Different thin silica films and especially the bilayer silica film have been developed in the Chemical Physics department. Here, the very first observations of amorphous silica network structures with atomic resolution were made by STM and nc-AFM and published in Angewandte Chemie together with the Structure and Reactivity group and theoretical support from Joachim Sauer´s group. Other researchers have later been able to verify these results by transmission electron microscopy (TEM). With our studies of the silica film, we have finally verified the atomic glass network, also known as random network theory, postulated by Zachariasen more than 80 years ago. The film structure resembles the original 2D drawings in all of its details. The characterization of real-space data allows for a clear assignment of atomic sites. The position of oxygen and silicon atoms can be determined by chemical sensitive imaging with STM and nc-AFM, and the ring structures, and their distribution and local neighborhood can be directly assessed. Furthermore, the structural transition from a crystalline to an amorphous domain has been investigated by STM imaging of an interface region. A smooth interface without under- or overcoordinated Si atoms was observed. The Si-Si distances appeared to remain constant in the crystalline, the vitreous, and the interfacial region. Ring statistics were computed for narrow image slices. This evaluation revealed that five- and sevenfold rings occur closer to the crystalline phase than four-, eight-, and nine-membered rings.

Impact of the Silica Network Structure on the Adsorption of Single Au and Pd Atoms

In a joint project with the groups of Martin Sterrer and Gianfranco Pacchioni we have investigated the preferred binding mechanisms for Au and Pd over bilayer silica on Ru(0001) supports using a combination of STM, nc-AFM, X-ray photoemission spectroscopy (XPS), and density functional theory (DFT). It appears that both metals prefer binding at the Ru interface after diffusing through the film, but do so with differing probabilities due to differing pore-size dependent entry barriers. While Pd may freely diffuse through pores within both crystalline and amorphous domains of the film, Au cannot enter the 6-membered rings making up the ordered phase, but, instead, binds exclusively within amorphous areas and domain boundaries, which possess larger ring structures.

Modifying the Pure Silica Film by Doping with Aluminum towards Zeolite Structures

Initial work for the preparation of zeolite films has been performed in the Structure and Reactivity group. We have reproduced these preparations to use the capabilities of our STM and nc-AFM to provide high local resolution measurements at the atomic scale. In first experiments we have compared occurring structural elements in the ring size distribution between the pure silica film and aluminosilicate films with varying doping concentrations of aluminum. Besides the investigation of different ring size distributions as well as ring combinations, we plan to use the chemical sensitive imaging contrast of our local probe method in order distinguish aluminum from silicon sites in the mixed film preparations.

In the presented cases it was shown how modern surface science techniques like nc-AFM and STM can complete and clarify the atomic models, which have been derived so far from diffraction methods. The benefits of locally resolving complex surface structures are obvious when non-periodic materials are investigated. A direct assignment from the obtained images is possible.

The employed high resolution imaging and spectroscopy significantly improves our understanding of the surface structure and chemistry of complex materials. The presented work can be referenced as a starting point for future experiments, where the tuning of certain material properties at the atomic scale becomes more and more important.

Scanning Probe Spectroscopy Group

The activities of the Scanning Probe Spectroscopy group have focused on four major topics in the past two years that are (i) preparation and analysis of semiconducting oxides with distinct optically properties (ZnO), (ii) investigation of doped oxides and their adsorption behavior, (iii) studies on ceria thin films as model system for a reducible oxide and (iv) elucidating the impact of individual molecules on the electronic structure of ultrasmall metal deposits. The experiments have been performed on two low-temperature (4K) STM setups suitable for performing conductance and inelastic tunneling spectroscopy and on a Photon STM (100 K), equipped with an optical read-out to detect photons emitted from the tip-sample junction during tunneling. For a complementary analysis of the electronic properties of the samples, we have employed X-ray photoelectron spectroscopy, using either a lab source or the synchrotron facility BESSY II. The main results of these experiments are summarized in the following:

(i) Probing the optical properties of ZnO films

The group has developed a reproducible recipe to prepare crystalline and atomically flat ZnO(0001) films of variable thickness on a Au(111) support. After a thorough morphological characterization, in particular of the predominant polarity-compensation scheme, we have focused on the photon-emission behavior of the films, as stimulated by electron injection from the STM tip into the surface. Beside the well-known band recombination peak at 373 nm wavelength, we observed three low-energy peaks that indicate the presence of defects in the ZnO lattice. Identification of the underlying defects was achieved with an extensive preparation program, in which typical ZnO point defects have been introduced in controlled quantities into the wurtzite lattice. For example, O-vacancies were generated reproducibly by thermal treatment, laser irradiation or reduction of the ZnO films with atomic hydrogen. All procedures resulted in the development of a distinct emission line at 730 nm, which was consequently assigned to an O-vacancy mediated emission process. Further experiments identified the role of Zn vacancies as well as O interstitials on the emission process. The optical response of ZnO was further modified by embedding Ag nano-particles into the oxide matrix, producing a model system for exciton-plasmon coupling as utilized in photo-catalytic and optoelectronic applications.

(ii) Tailoring physical and chemical properties of doped oxides

In continuation of earlier work, we have performed an in-depth examination of the role of Mo-donors in a crystalline CaO(001) film. We succeeded to visualize dopants in the near surface CaO layers via their characteristic charging rings that come about as the dopants reversibly change oxidation state in presence of the STM tip. The information on the spatial distribution of the dopants was complemented with XPS spectroscopy data on their preferred charge state. Whereas in the as grown CaO films, the Mo ions prefer low charge states (+2, +3), a gradual increase of the oxidation state up to 5+ and +6 is observed at higher annealing temperatures. The underlying redox-properties of the embedded Mo ions profoundly influence the binding characteristic of the doped film for adsorbates with large electron affinity. For example, gold atoms take up one extra electron from the Mo donors and modify their binding scheme to the CaO surface, giving rise to an unusual 2D growth regime of the metal. Similarly, O_2 molecules transform into negatively charged superoxo species upon adsorption, which exhibit a large propensity to dissociate into atomic oxygen. In the case of O_2 / Au co-adsorption, a competition for the Mo excess electrons governs the adsorption behavior of the doped oxide. At low O_2 partial pressure, the metal takes most of the electrons and the Au growth remains 2D, while at high pressures, oxygen is the more efficient electron acceptor and the gold remains neutral. Further experiments have explored the gradual change in the CaO properties when going to higher and higher doping levels and finally to a ternary Ca-Mo mixed oxide.

(iii) Gold adsorption on crystalline CeO2(111) films

As a model system for a reducible oxide, we have prepared $CeO_2(111)$ films with varying densities of surface and subsurface oxygen vacancies and tested the resulting adsorption properties via gold titration. In correspondence to theoretical models, the Au was found to interact with surface O defects, although the trapping ability of the defect was much smaller than expected. Interestingly, also subsurface O defects, more precisely the Ce^{3+} ions associated to them, are able to bind Au. In this case, the surface Ce^{3+} ions get re-oxidized by donating their extra electron to the Au adatom. This process is energetically favorable, because it allows for a reduction of local lattice strain, formerly introduced by the spacious Ce^{3+} ions in the film. At higher exposure, Au forms 3D aggregates on all ceria samples analyzed here, indicating that charge transfer process are not controlling the metal-oxide interaction to a large extent. Complementing experiments have been performed on the CeO_2(100) surface that is, in contrast to the (111) termination, of polar nature and features an electrostatic dipole.

(iv) Molecular adsorption on ultrasmall metal particles with quantized electronic structure

Nanosized metal deposits are the active species in heterogeneous catalysis. At reaction conditions, their intrinsic properties are heavily perturbed by molecular adsorption, which renders in situ studies of the metal-support systems advisable. In order to simulate this situation at defined conditions, we have analyzed the electronic structure of individual Au particles containing between 50-150 atoms before and after contact with isophorone as nucleophilic molecular species. The pristine particles are characterized by a series of quantum well states, as identified with STM conductance spectroscopy. The position and symmetry of the eigenstates were found to depend on size and shape of the metal islands. Especially the smaller deposits exhibit pronounced state-density gaps at the Fermi level that correlate with the eccentricity of the particle, i.e. the difference of their squared semi axes. After isophorone adsorption, a systematic downshift of the quantum-well states is revealed that is explained with a redistribution of charge from the interior of the cluster to the electron-accepting molecules at the edge. The lower charge density in the metal reduces the effect of electron-electron repulsion and thus lowers the level energies in the islands. In the limit of ultrasmall Au aggregates, the charge-transfer picture breaks down and formation of a covalent metal-molecule bond is observed. The experiments already demonstrate the strong effect of molecular adsorption on a confined quantum system, but will be reproduced with electron-donating molecules (such as NH_3) in future.

Magnetic resonance group

The remainder (note: Thomas Risse became Professor at the Freie Universität Berlin in 2010) of the magnetic resonance group still working at the Fritz-Haber-Institut focuses on the development of an ultrahigh vacuum compatible high-field electron paramagnetic resonance (EPR) experiment operating at 95 GHz. The group has integrated a commercial high filed EPR spectrometer (Bruker) into a multi-technique apparatus including STM and IR to allow for a comprehensive characterization of surfaces. In this machine the operating frequency increases by a factor of ten as compared to the X-band spectrometers operating at about 9.5 GHz used before. This offers an enhanced spectral resolution for species with very small differences in the g-matrix component. To adapt the high field spectrometer to UHV conditions it is not possible to use a monomodal resonator due to the size requirements imposed by a wavelength of about 3 mm. Instead we decided to use a Farby-Pérot resonator. Here, the single crystal metal surface serves as the planar mirror of the Fabry-Pérot resonator. The investigation of well-defined surfaces requires measurements in UHV and it turned out that the way the cavity is sealed to the ambient is of crucial practical importance. The first attempt being made used a resonator having a 100 μm thick quartz window positioned between the two mirrors as the vacuum barrier. However, it turned out that slight changes of the sample position and alignment has strong impact on the reproducibility of measurements. Therefore, a new resonator design was put in place where the cavity is sealed by a sapphire window, which is glued inside the waveguide directly onto the backside of the coupling hole of the concave mirror. Given the small space requirements and microwave incompatibilities of UHV usable glues it took quite some effort to produce He-tight concave mirrors with the necessary coupling antenna made out of 60 μm thick Au wire glued into small holes of the sapphire window in place and with very good microwave characteristics (Q ≈ 2500). With this resonator we could show that the system does not only have the required long term stability to accumulate data over more than 10 hours, but also give background stabilities such that signals in the range of 1/1000 of a monolayer can be measured.

The setup was used to investigate paramagnetic point defects on single crystalline MgO(001) films grown on Ag(100). Form the X-band studies performed earlier we know that electron bombardment of MgO(100)/Mo(100) does produce paramagnetic surface color centers and depending on the preparation condition also paramagnetic centers in the bulk. For the films grown on Ag(001) we were so far not able to find evidence for color centers on the surface of this system grown on Ag(001). However, it was possible to see paramagnetic centers in the bulk of the film. The interesting observation at this point is that the W-band experiments prove the existence of more than one species after electron bombardment of the film. An analysis of the signal revealed that there are at least three species. Two classes of signals may be discriminated: there is a single isotropic line, which makes up about 50 % of the total intensity. In addition, two anisotropic signals can be identified, which show a different temperature behavior as the isotropic one. The latter can be associated to paramagnetic centers in dislocation lines of the film while the former is consistent with the formation of a bulk color center. Interestingly, the corresponding X-band spectra of paramagnetic bulk centers show an asymmetric line shape indicating that there is more than a single species present in the film. It is now possible to understand this line shape observed at X-band on the basis of the signal components extracted from the W-band spectra, which documents the ability of high field EPR to gain additional insight into systems containing a variety of similar paramagnetic species not to be discriminated at conventional X-band spectroscopy.

Catalysis / Laser Spectroscopy Group

Over the last two years the Catalysis/Laser spectroscopy group has intensified its efforts to reveal the influence of environmental factors on the properties of single-crystalline oxide surfaces and oxide-supported metal particles used as model systems of heterogeneous catalysts. The group runs several surface science experiments equipped with analytical tools such as X-ray photoelectron spectroscopy (XPS), infrared reflection-absorption spectroscopy (IRAS), temperature-programmed desorption (TPD), and scanning tunneling microscopy (STM) that allow the electronic, vibrational, and structural properties of the samples to be determined, and is involved in the set-up of new equipment for studying oxide surfaces in contact with aqueous solutions (sum-frequency generation and electrochemical characterization techniques). Several research topics have been addressed, such as hydroxylation of oxide surfaces, the interaction of metal particles with clean and hydroxylated oxide surfaces, the science of catalyst preparation, and the extension of surface science investigations to solid-liquid interfaces. The sample systems investigated include metal-supported, thin oxide films (MgO, CaO, SiO_2, Fe_3O_4) and Au and Pd particles deposited thereon.

Electronic properties of oxide-supported metal atoms and particles

XPS binding energy (BE) shifts of supported metal particles are governed by a number of effects such as initial-state contributions including charge transfer, lattice contraction, electrostatic charging and surface core-level shifts, and size-dependent final-state screening. Disentanglement of these contributions is difficult and the inability to do so often leads to erroneous interpretations of XPS BE shifts. The Auger parameter, which is based on the determination of XPS BE´s and the kinetic energy of a properly selected Auger transition, is a useful concept that allows recorded BE shifts to be separated into initial-state and final-state contributions and has recently been used in our group to help interpreting XPS data. In order to demonstrate the applicability and usefulness of the Auger parameter we have studied Pd atoms deposited onto a bilayer-SiO_2/Ru(0001) model surface. As shown by the AFM group (Dr. Markus Heyde) and confirmed by computations (G. Pacchioni, Milano), single Pd atoms penetrate into the pores of the silica film and adsorb at the metal interface, thereby creating a sample with a large abundance of stable surface atoms on top of a dissimilar substrate, which allows the electronic properties of these under-coordinated metal species to be studied without the complicating contributions from non-surface moieties present in the bulk metal or in particles. Our own combined XPS/Auger experiments of such a sample determined the Pd 3d BE shift, which is -0.3 eV relative to Pd bulk, to be the result of the counterbalancing contributions of initial-state effects (shifting the 3d orbital energy by 1.1 eV closer to the Fermi level) and final-state screening effects (shifting the 3d orbital energy by 0.8 eV away from E_F). Typically, the -0.3 eV core-level BE shift would be interpreted as charge-transfer to Pd, which is, however, not the case according to DFT results, which indicated a partial positive charge on the interface-confined Pd atoms. Instead, the measured core-level binding energy shift is related to changes in the initial-state electronic structure of the under-coordinated Pd atoms, specifically, to a redistribution of valence-level electrons compared to Pd atoms in the bulk, leading to an upward-shift of the orbital energies, which outweighs the decreases due to extra-atomic screening and electron-density changes, in agreement with conclusions from ab-initio Hartree-Fock calculations performed by Paul Bagus.

The Auger parameter concept has in addition been applied to determine the electronic properties of metal particles on hydroxylated oxide surfaces and to correlate electronic structure changes with surface reactivity. As an example, Auger parameter analysis together with vibrational spectroscopy and TPD was used to obtain a detailed picture about the temperature dependence of the Red-Ox-reaction between Pd atoms and surface hydroxyls on a hydroxylated MgO(100) model surface. This study provided direct evidence for the interrelation between Pd oxidation by hydroxyls and the release of hydrogen as a result of the Red-Ox process.

Surface science investigations into catalyst preparation

We have previously introduced, using Pd on $Fe_3O_4(111)$ as a model system, a surface science approach to catalyst preparation that utilizes thin, single-crystalline oxide films as substrates for studying processes such as the adsorption of catalyst precursors from aqueous solutions and the formation of metal nanoparticles during the decomposition of the precursors, during typically applied wet-chemical catalyst preparation procedures. In our recent studies, we have studied the bonding of Pd precursors to the $Fe_3O_4(111)$ substrate in more detail and extended this approach also to other sample systems (Au-$Fe_3O_4(111)$ and Pd-MgO(100)). The interaction between aqueous Pd precursors (obtained from $PdCl_2$) with $Fe_3O_4(111)$ has been investigated for Pd adsorption from a strongly acidic precursor solution, where $PdCl_4^{2-}$ complexes interact with the positively charged $Fe_3O_4(111)$ surface via an electrostatic adsorption mechanism, and from a strongly alkaline precursor solution, where a direct chemical bonding between surface hydroxyls and the $Pd(OH)_4^{2-}$ precursor is favored. By controlled rinsing experiments using either water or blank solutions we were able to reveal the differences in bonding strength and solubility of the adsorbed precursors, their respeciation upon rinsing, and the effect of residual chlorine on the support surface. The latter point is particularly important and it could be shown using STM that the formation of highly dispersed Pd particles requires complete removal of chlorine by water rinsing. Along the same line, we have studied the preparation of $Fe_3O_4(111)$-supported Au particles from strongly alkaline Au^{3+} precursor solution, following a preparation procedure introduced by Masatake Haruta to obtain highly active Au catalysts. Our studies on Au-$Fe_3O_4(111)$ represent the first successful realization of the preparation of highly dispersed Au nanoparticles on a single-crystalline substrate using Haruta´s deposition-precipitation approach.

In addition to $Fe_3O_4(111)$, we used MgO(100) thin films as substrate for the preparation of supported Pd particles to investigate the influence of support dissolution during catalyst preparation. Our studies have shown that the interaction of the Pd precursor with MgO during the period of maximum MgO dissolution rate leads to a stabilization of oxidized Pd species, possibly by incorporation of Pd^{2+} in the MgO matrix during the fast MgO dissolution and re-precipitation events. These studies also revealed the strong influence of residual carbon, which was found to enhance the thermal decomposition of Pd precursors.

Construction of new equipment for in-situ investigations of oxide-liquid interfaces

As part of the ERC-funded Starting-Grant project STRUBOLI, we set up two new experiments that will in the future allow in-situ studies of oxide-liquid interfaces: A sum frequency generation spectroscopy set-up and a UHV preparation chamber combined with transfer options into liquid cells and electrochemical characterization of thin oxide films. The installation of both set-ups is almost complete and first test experiments have been performed.

Molecular Beam Group

The activities of the molecular beam group focus on atomistic-level understanding of heterogeneously catalyzed reactions including mechanisms, kinetics and thermodynamics of elementary surface processes. The main focus lies on finding detailed correlations between the structural properties of nano-sized supported catalysts and activity and selectivity of surface reactions. A variety of surface science methods - multi-molecular beam techniques, single crystal adsorption calorimetry (SCAC) and infrared reflection absorption spectroscopy (IRAS) - is employed in these studies in order to address both kinetic and thermodynamic aspects of interaction of gas phase molecules with well-defined model surfaces.

Chemoselective hydrogenation of isophorone and acrolein over Pd/Fe_3O_4/Pt(111) model catalysts

In this project detailed kinetic investigations on a complex multi-pathway surface reaction using a combination of pulsed multi-molecular beam techniques with IRAS is performed. Current research is focussed on finding structure-reactivity relationships with a particular emphasis on selectivity and on the elucidation of the reaction mechanism at the atomistic level. Currently, hydrogenation and isomerization of α,β-unsaturated ketones, such as isophorone and acrolein, over Pd nanoparticles supported on well-defined model Fe_3O_4/Pt(111) oxide film and Pd(111) we investigativ. By varying structural properties and chemical composition (e.g. by depositing surface modifiers) of model catalysts and performing detailed kinetic investigations an atomistic-level understanding of hydrocarbon conversions with hydrogen is approached, revealing the role of specific surface sites (such as e.g. low-coordinated surface sites) in competing reaction pathways.

Particularly, it was found that selectivity in partial hydrogenation of acrolein strongly depends on the nature of the catalytic surface and its chemical composition under the reaction conditions. Thus, selective hydrogenation of acrolein to an unsaturated alcohol was observed to proceed on Pd(111) while hydrogenation of the C=C bond resulting in saturated ketone was found to be favored over 6 nm-sized supported Pd nanoparticle. By employing IRAS detection of the surface reaction intermediates with a simultaneous detection of the gas phase product, it was possible to directly correlate the product formation with a particular hydrocarbon surface intermediate formed in course of the reaction. Current IRAS investigations point to a crucial role of spectator species, which are formed via initial acrolein decomposition at the early stages of reaction, in governing the selectivity in partial hydrogenation. Ongoing research is focussed on identifying the key microscopic features governing the selectivity of these processes.

Oxygen adsorption energy on Pd/Fe_3O_4/Pt(111) model catalysts by single-crystal adsorption microcalorimetry: particle size dependence

In this project, the particle size dependence of oxygen adsorption energies on well-defined Pd nanoparticles ranging from 220 to 4800 atoms per particle as compared to an extended Pd(111) single crystal surface was investigated in a direct calorimetric study. Two microscopic structural parameters have been identified to critically control the oxygen binding energies on Pd nanoparticles: the local configuration of the adsorption site and the particle size. The change of the local adsorption environment from a three-fold hollow position on extended Pd (111) single crystal to an edge site of Pd nanoparticles results in a strong increase of oxygen binding energy by about 70 $kJ \cdot mol^{-1}$. The preferential adsorption site of oxygen atoms was investigated spectroscopically by IRAS, using CO as a probe molecule for different adsorption sites, and was found to be at the edges/corners of the Pd clusters. The unexpectedly strong binding energy of oxygen at the particles edges exceeds all literature values of oxygen adsorption energies at stepped single crystal surfaces. On the other hand, if the local configuration of the adsorption site is kept constant (O adsorption at the edges/corners of Pd nanoparticles), the reduction of the cluster size leads to a pronounced decrease of oxygen binding energy from 275 kJ·mol^{-1} observed on the large clusters to 205 kJ·mol^{-1} measured for the smallest investigated nanoparticles. This latter trend coincides with the particle size dependence of CO adsorption energy on Pd particles obtained earlier. The decreasing adsorption energies observed in our studies with decreasing particle size both for CO and oxygen adsorption are indicative of a general nature of this phenomenon, which might be connected to the theoretically predicted weakening of chemisorptive interaction due to the contraction of the lattice parameter of the Pd cluster. The effects, which both of these structural parameters exhibit on the oxygen adsorption energy, result in counteracting trends – the increase of the binding strength due to adsorption at the low-coordinated surface sites, and the decrease of the adsorption energy due to reduced particle dimensions. In total, the oxygen binding energy turns out to be a convolution of these two trends resulting in a non-monotonous dependence of the oxygen adsorption energies on particle size.

Spectro-Microscopy Group (SMART)

The growth and the chemical and structural properties of thin iron oxide and silica films, as well as metal nano-particles grown on iron oxide supports have been studied *in situ* and *in real time* with the SMART microscope (SpectroMicroscope with Aberration correction for many Relevant Techniques). Parallel to these investigations the instrumental development of the new SMART-II has been pushed forward to (a) reduce the space charge effects, up to now limiting the lateral resolution in XPEEM to 18 nm and (b) to optimize the pre- and post optics and the multipole correctors in the new electrostatic Omega filter.

Iron oxide films

Applying the new recipe for the Fe_3O_4 film preparation, we produced very flat films with terraces hundreds of nanometer wide. Beside the atomic steps, appearing as significant dark lines in the LEEM (low energy electron microscopy) image, two further types of line structures could be observed. By variation of the electron energy and focus series combined with wave optics calculations, simulating the experimental contrast, the surface profiles could be estimated: (i) a smoothed step about 4 nm wide with a height, corresponding to half the unit cell size of Fe_3O_4, and (ii) Lorentzian like grooves forming a hexagonal network with a periodicity of about 70 nm. Both surface profiles can be explained by overgrown dislocation lines about 4 – 6 nm below the surface. Whereas in the first case the dislocation is caused by a missing half plane with a Burgers vector perpendicular to the surface, the dislocation in the second case is due to a missing atomic row with a Burgers vector parallel to the surface.

In the two last years the preparation parameters were successfully studied to control the α-Fe_2O_3 film growth in UHV on different metal surfaces. Compared to the growth conditions of Fe_3O_4, the oxidation temperature has to be reduced, whereas the oxygen pressure has to be enlarged. On Pt(111) surface this yields a complete film of Fe_2O_3, but always with a bi-phase termination. Moreover, we found different ways to transform reversely the two most common iron oxide structures – Fe_3O_4 and α-Fe_2O_3 – into each other. As expected from the bulk phase diagram, the annealing in oxygen transforms Fe_3O_4 into α-Fe_2O_3, whereas an intermediate phase was observed, most likely the γ-Fe_2O_3 structure. Unexpectedly, the annealing of a thin Fe_3O_4 film on Pt(111) in UHV produces also the α-Fe_2O_3 structure, i.e. oxidation, because the Fe diffusion into the Pt bulk is faster than the oxygen desorption. For the reverse process – α-Fe_2O_3 reduces to Fe_3O_4 – Fe was deposited onto the film and subsequently annealed in UHV. In case of Ag(111) as metal support for the Fe_2O_3 film, the reduction was observed if annealed for longer time; obviously, the Fe diffusion into the substrate bulk is limited and therefore the oxygen desorption is dominant.

Silica on Ru(0001)

As shown first by STM in our department, vitreous and ordered phases of the Silica bi-layer film can co-exist on the Ru(0001) surface. Taking the advantage to combine the structural contrast of LEEM with *real time* imaging, we aim for studying the formation of the vitreous phase and to clarify the existence of a structural transition between the ordered and disordered phase. Especially the influence of local coverage and substrate defects like steps and step bunches are in our focus. By LEED, the transformation from the initial (2x2) structure into a ring like, i.e. amorphous, structure was observed *in situ* and in real time during the annealing. The domains in the intermediate mixed phase are up to now too small to be imaged in dark-field. Therefore, we are optimizing the preparation condition to enlarge these domains. Various other structural domains, occurring at a SiO_2 coverage smaller than the bilayer, were found and are large enough (>50 nm) to be resolved.

SMART-2

This new aberrations-corrected LEEM-PEEM instrument with a new electrostatic Omega-filter will substitute the SMART-1 instrument, which demonstrated a lateral resolution of 18 nm in energy filtered XPEEM (X-ray stimulated Photo Emission Electron Microscopy), the world wide best value for this type of instrument. Though this value could only be achieved with aberration correction, it is limited by space charge effects due to the temporary high electron intensity caused by the pulsed time structure of the synchrotron light. However, without space charge, as shown in LEEM, the electron optics and especially the aberrations correction are sufficient for a resolution of 2.6 nm. Therefore, we are constructing a new beam splitter module with implemented apertures and slits to reduce reasonably the electron beam intensity by cutting away the part not used on the detector. The new Omega filter is optimized for both, a lateral and an energy resolution better than 2 nm and 100 meV, respectively. Nearly half of the possible second order aberrations are already compensated by the intrinsic symmetry. Half of the residual aberrations can be fully compensated by multipoles, the others not. For the latter we found the optimum design for the deceleration and the acceleration optics in combination with optimized pass energy, to reduce their effect smaller than of the unavoidable third order aberrations.

Crystal Laboratory

The Crystal laboratory provides single crystal samples for research studies in the entire Institute. The laboratory is equipped with all necessary tools for cutting a crystal rod, shaping, polishing, and final orienting the crystal surface using a Laue diffractometer. Another duty of the Crystal lab includes machinery of brittle and fragile ceramic and glass-like materials. For the last two years the laboratory prepared more than 160 crystal samples. High quality work of the Crystal-lab is also acknowledged by many other groups worldwide (Germany, China, USA, Russia, etc.) working with the crystals prepared in the Crystal-lab within several collaborative projects.

Publications of the Chemical Physics

2011 (late publications)

Benedetti, S., N. Nilius, P. Torelli, G. Renaud, H.-J. Freund and S. Valeri: Competition between polar and nonpolar growth of MgO thin films on Au(111). The Journal of Physical Chemistry C **115** (46), 23043-23049 (2011).

Duncan, D.A., W. Unterberger, D. Kreikemeyer-Lorenzo and D.P. Woodruff: Uracil on Cu(110): A quantitative structure determination by energy-scanned photoelectron diffraction. The Journal of Chemical Physics **135** (1), 014704 (2011).

Flores-Camacho, J.M., J.-H. Fischer-Wolfarth, M. Peter, C.T. Campbell, S. Schauermann and H.-J. Freund: Adsorption energetics of CO on supported Pd nanoparticles as a function of particle size by single crystal microcalorimetry. Physical Chemistry Chemical Physics **13** (38), 16800-16810 (2011).

Flores-Camacho, J.M., G. Weidlinger, L.D. Sun, K. Schmidegg, M. Hohage, D. Primetzhofer, P. Bauer and P. Zeppenfeld: Growth and optical properties of Ag clusters deposited on poly(ethylene terephthalate). Nanotechnology **22** (27), 275710 (2011).

Freund, H.-J., G. Meijer, M. Scheffler, R. Schlögl and M. Wolf: CO oxidation as a prototypical reaction for heterogeneous processes. Angewandte Chemie International Edition **50** (43), 10064-10094 (2011).

Freund, H.-J., G. Meijer, M. Scheffler, R. Schlögl and M. Wolf: Die CO-Oxidation als Modellreaktion für heterogene Prozesse. Angewandte Chemie **123** (43), 10242-10275 (2011).

Gonchar, A., T. Risse, H.-J. Freund, L. Giordano, C. Di Valentin and G. Pacchioni: Activation of oxygen on MgO:$\mathrm{O2}^{\cdot-}$ radical ion formation on thin, metal-supported MgO(001) films. Angewandte Chemie International Edition **50** (11), 2635-2638 (2011).

Gonchar, A., T. Risse, H.-J. Freund, L. Giordano, C. Di Valentin and G. Pacchioni: Aktivierung von molekularem Sauerstoff auf MgO: Bildung von $\mathrm{O2}^{\cdot-}$ auf dünnen, trägerfixierten MgO(001)-Filmen. Angewandte Chemie **123** (11), 2684-2687 (2011).

Ludwig, W., A.A. Savara, K.-H. Dostert and S. Schauermann: Olefin hydrogenation on Pd model supported catalysts: New mechanistic insights. Journal of Catalysis **284** (2), 148-156 (2011).

Nelin, C.J., P.S. Bagus, M.A. Brown, M. Sterrer and H.-J. Freund: Analysis of the broadening of X-ray photoelectron spectroscopy peaks for ionic crystals. Angewandte Chemie **123** (43), 10356-10359 (2011).

Nelin, C.J., P.S. Bagus, M.A. Brown, M. Sterrer and H.-J. Freund: Analysis of the broadening of X-ray photoelectron spectroscopy peaks for ionic crystals. Angewandte Chemie International Edition **50** (43), 10174-10177 (2011).

Pan, Y., S. Benedetti, N. Nilius and H.-J. Freund: Change of the surface electronic structure of Au(111) by a monolayer MgO(001) film. Physical Review B **84** (7), 075456 (2011).

Ringleb, F., Y. Fujimori, H.-F. Wang, H. Ariga, E. Carrasco, M. Sterrer, H.-J. Freund, L. Giordano, G. Pacchioni and J. Goniakowski: Interaction of water with FeO(111)/Pt(111): Environmental effects and influence of oxygen. The Journal of Physical Chemistry C **115** (39), 19328-19335 (2011).

Romanshyn, Y., S. Guimond, D. Göbke, J.M. Sturm, H. Kuhlenbeck, J. Döbler, M.V. Ganduglia-Pirovano, J. Sauer and H.-J. Freund: Methanol adsorption on V_2O_3(0001). Topics in Catalysis **54** (10-12), 669-684 (2011).

Shao, X., S. Prada, L. Giordano, G. Pacchioni, N. Nilius and H.-J. Freund: Einstellung der Gleichgewichtsform metallischer Nanopartikel durch Dotierung des Oxidträgers. Angewandte Chemie **123** (48), 11728-11731 (2011).

Shao, X., S. Prada, L. Giordano, G. Pacchioni, N. Nilius and H.-J. Freund: Tailoring the shape of metal Ad-particles by doping the oxide support. Angewandte Chemie International Edition **50** (48), 11525-11527 (2011).

Simon, G.H., T. König, L. Heinke, L. Lichtenstein, M. Heyde and H.-J. Freund: Atomic structure of surface defects in alumina studied by dynamic force microscopy: Strain-relief-, translation- and reflection-related boundaries, including their junctions. New Journal of Physics **13** (12), 123028 (2011).

Todd, A.M.M., H. Bluem, V. Christina, M.D. Cole, J. Ditta, D. Dowell, K. Jordan, R. Lange, J.H. Park, J. Rathke, T. Schultheiss, L.M. Young, W. Schöllkopf, S. Gewinner, W. Erlebach, H. Junkes, A. Liedke, W. Zhang, G. von Helden, G. Meijer and S.C. Gottschalk: Commissioning Status of the Fritz Haber Institute THz FEL. In: Proceedings of 2nd International Particle Accelerator Conference, IPAC'11, San Sebastián, Spain. THPC106 (2011). ISBN 978-92-9083-366-6.

Unterberger, W., T.J. Lerotholi, E.A. Kröger, M.J. Knight, D.A. Duncan, D. Kreikemeyer-Lorenzo, K.A. Hogan, D.C. Jackson, R. Włodarczyk, M. Sierka, J. Sauer and D.P. Woodruff: Local hydroxyl adsorption geometry on TiO_2(110). Physical Review B **84** (11), 115461 (2011).

Habilitation

Schauermann, S.: Interaction of gas phase molecules with nano-structured model supported catalysts. Freie Universität Berlin 2011.

Doctoral Thesis

Lewandowski, M.: Scanning tunneling microscopy study of iron oxide based model catalysts. Technische Universität Berlin 2011.

Diploma Theses

Büchner, C.: A surface science study of a catalytic model system: SiO_2 on Ru(0001). Humboldt-Universität zu Berlin 2011.

2012

Bagus, P.S., A. Wieckowski and H.-J. Freund: The contribution of lattice strain to core-level binding energy shifts in metal nanoparticles: Generality and origin of the shifts. Computational and theoretical chemistry **987**, 22-24 (2012).

Beck, B., M. Harth, N. Hamilton, C. Carrero, J. Uhlrich, A. Trunschke, S.K. Shaikhutdinov, H. Schubert, H.-J. Freund, R. Schlögl, J. Sauer and R. Schomäcker: Partial oxidation of ethanol on vanadia catalysts on supporting oxides with different redox properties compared to propane. Journal of Catalysis **296**, 120-131 (2012).

Boscoboinik, A., X. Yu, B. Yang, F.D. Fischer, R. Włodarczyk, M. Sierka, S.K. Shaikhutdinov, J. Sauer and H.-J. Freund: Modeling Zeolites with Metal-Supported Two-Dimensional Aluminosilicate Films. Angewandte Chemie International Edition **51** (24), 6005-6008 (2012).

Boscoboinik, A., X. Yu, B. Yang, F.D. Fischer, R. Włodarczyk, M. Sierka, S.K. Shaikhutdinov, J. Sauer and H.-J. Freund: Modellierung von Zeolithen durch zweidimensionale Aluminosilicatfilme auf Metallunterlagen. Angewandte Chemie **124** (24), 6107-6111 (2012).

Bowker, M., G. Hutchings, P.R. Davies, D. Edwards, R. Davies, S.K. Shaikhutdinov and H.-J. Freund: Surface structure of γ-Fe_2O_3(111). Surface Science **606** (21-22), 1594-1599 (2012).

Brown, M.A., F. Vila, M. Sterrer, S. Thürmer, B. Winter, M. Ammann, J.J. Rehr and J.A. van Bokhoven: Electronic Structures of Formic Acid (HCOOH) and Formate ($HCOO^-$) in Aqueous Solutions. The Journal of Physical Chemistry Letters **3** (13), 1754-1759 (2012).

Büchner, C., L. Lichtenstein and M. Heyde: Ein glasklares Modell. Nachrichten aus der Chemie **60** (9), 861-864 (2012).

Campbell, C.T., J.T. Yates and H.-J. Freund: D. Wayne Goodman (1945–2012). Angewandte Chemie **124** (25), 6147-6148 (2012).

Campbell, C.T., J.T. Yates and H.-J. Freund: D. Wayne Goodman (1945–2012). Angewandte Chemie International Edition **51** (25), 6043-6044 (2012).

Casu, M.B., S.-A. Savu, B.-E. Schuster, I. Biswas, C. Raisch, H. Marchetto, T. Schmidt and T. Chassé: Island shapes and aggregation steered by the geometry of the substrate lattice. Chemical Communications **48** (55), 6957-6959 (2012).

Duncan, D.A., M.K. Bradley, W. Unterberger, D. Kreikemeyer-Lorenzo, T.J. Lerotholi, J. Robinson and D.P. Woodruff: Deprotonated Glycine on Cu(111): Quantitative Structure Determination by Energy-Scanned Photoelectron Diffraction. The Journal of Physical Chemistry C **116** (18), 9885-9995 (2012).

Duncan, D.A., W. Unterberger, D.C. Jackson, M.K. Knight, E.A. Kroeger, K.A. Hogan, C.L.A. Lamont, T.J. Lerotholia and D.P. Woodruff: Quantitative local structure determination of R,R-tartaric acid on Cu(110): Monotartrate and bitartrate phases. Surface Science **606** (17-18), 1435-1442 (2012).

Duncan, D.A., W. Unterberger, D. Kreikemeyer-Lorenzo and D.P. Woodruff: Does methanol produce a stable methoxy species on Ru(0001) at low temperatures? Surface Science **606** (15-16), 1298-1302 (2012).

Fernández-Torrente, I., D. Kreikemeyer-Lorenzo, A. Stróżecka, K.J. Franke and J.I. Pascual: Gating the Charge State of Single Molecules by Local Electric Fields. Physical Review Letters **108** (3), 036801 (2012).

Freund, H.-J.: Oxide films as catalytic materials and models of real catalysts. In: Oxide Ultrathin Films: Science and Technology, (Eds.) G. Pacchioni and S. Valeri. Vol. 7. Wiley-VCH, Weinheim, 145-179 (2012). ISBN 978-3-527-33016-4.

Heyde, M., S.K. Shaikhutdinov and H.-J. Freund: Two-dimensional silica: Crystalline and vitreous. Chemical Physics Letters **550**, 1-7 (2012).

Junkes, H., R. Lange and S.B. Webb: BACnet liefert Daten der TGA am Fritz-Haber-Institut. BACnet Europe Journal **17**, 16-17 (2012).

Kim, K.H., K. Watanabe, D. Menzel and H.-J. Freund: Comparative study of thermal and photo-induced reactions of NO on particulate and flat silver surfaces. Surface science **606** (15-16), 1142-1151 (2012).

Kozlov, S.M., F. Viñes, N. Nilius, S.K. Shaikhutdinov and K.M. Neyman: Absolute Surface Step Energies: Accurate Theoretical Methods Applied to Ceria Nanoislands. The Journal of Physical Chemistry Letters **3** (15), 1956-1961 (2012).

Lewandowski, M., I. Groot, S.K. Shaikhutdinov and H.-J. Freund: Scanning tunneling microscopy evidence for the Mars-van Krevelen type mechanism of low temperature CO oxidation on an FeO(111) film on Pt(111). Catalysis Today **181** (1), 52-55 (2012).

Lichtenstein, L., C. Büchner, S. Stuckenholz, M. Heyde and H.-J. Freund: Enhanced atomic corrugation in dynamic force microscopy - The role of repulsive forces. Applied Physics Letters **100** (12), 123105 (2012).

Lichtenstein, L., C. Büchner, B. Yang, S.K. Shaikhutdinov, M. Heyde, M. Sierka, R. Wlodarczyk, J. Sauer and H.-J. Freund: Die atomare Struktur eines metallgestützten glasartigen dünnen Silikafilms. Angewandte Chemie **124** (2), 416-420 (2012).

Lichtenstein, L., C. Büchner, B. Yang, S.K. Shaikhutdinov, M. Heyde, M. Sierka, R. Wlodarczyk, J. Sauer and H.-J. Freund: The atomic structure of a metal-supported vitreous thin silica film. Angewandte Chemie International Edition **51** (2), 404-407 (2012).

Lichtenstein, L., M. Heyde and H.-J. Freund: Atomic Arrangement in Two-Dimensional Silica: From Crystalline to Vitreous Structures. The Journal of Physical Chemistry C **116** (38), 20426-20432 (2012).

Lichtenstein, L., M. Heyde and H.-J. Freund: Crystalline-Vitreous Interface in Two Dimensional Silica. Physical Review Letters **109** (10), 106101 (2012).

Lichtenstein, L., M. Heyde, S. Ulrich, N. Nilius and H.-J. Freund: Probing the properties of metal–oxide interfaces: silica films on Mo and Ru supports. Journal of Physics: Condensed Matter **24** (35), 354010 (2012).

Liu, W., A.A. Savara, X. Ren, W. Ludwig, K.-H. Dostert, S. Schauermann, A. Tkatchenko, H.-J. Freund and M. Scheffler: Toward Low-Temperature Dehydrogenation Catalysis: Isophorone Adsorbed on Pd(111). The Journal of Physical Chemistry Letters **3** (5), 582-586 (2012).

Ludwig, W., A.A. Savara, R.J. Madix, S. Schauermann and H.-J. Freund: Subsurface hydrogen diffusion into Pd nanoparticles: Role of low-coordinated surface sites and facilitation by carbon. The Journal of Physical Chemistry C **116** (5), 3539-3544 (2012).

Martynova, Y., B. Yang, X. Yu, A. Boscoboinik, S.K. Shaikhutdinov and H.-J. Freund: Low Temperature CO Oxidation on Ruthenium Oxide Thin Films at Near-Atmospheric Pressures. Catalysis Letters **142** (6), 657-663 (2012).

Menzel, D.: Electronically induced surface reactions: Evolution, concepts, and perspectives. The Journal of Chemical Physics **137** (9), 091702 (2012).

Nilius, N., S. Benedetti, Y. Pan, P. Myrach, C. Noguera, L. Giordano and J. Goniakowski: Electronic and electrostatic properties of polar oxide nanostructures: MgO(111) islands on Au(111). Physical Review B **86** (20), 205410 (2012).

Nilius, N., S.M. Kozlov, J.-F. Jerratsch, M. Baron, X. Shao, F. Viñes, S.K. Shaikhutdinov, K.M. Neyman and H.-J. Freund: Formation of one-dimensional electronic states along the step edges of CeO_2(111). ACS Nano **6** (2), 1126-1133 (2012).

Oyama, S.T., H. Zhao, H.-J. Freund, K. Asakura, R. Wlodarczyk and M. Sierka: Unprecedented selectivity to the direct desulfurization (DDS) pathway in a highly active FeNi bimetallic phosphide catalyst. Journal of Catalysis **285**, 1-5 (2012).

Pan, Y., S. Benedetti, C. Noguera, L. Giordano, J. Goniakowski and N. Nilius: Compensating Edge Polarity: A Means To Alter the Growth Orientation of MgO Nanostructures on Au(111). The Journal of Physical Chemistry C **116** (20), 11126-11132 (2012).

Pan, Y., B. Yang, C. Hulot, S. Blechert, N. Nilius and H.-J. Freund: Effect of lattice-gas atoms on the adsorption behaviour of thioether molecules. Physical Chemistry Chemical Physics **14** (31), 10987-10993 (2012).

Sala, A., H. Marchetto, Z.-H. Qin, S.K. Shaikhutdinov, T. Schmidt and H.-J. Freund: Defects and inhomogeneities in Fe_3O_4(111) thin film growth on Pt(111). Physical Review B **86** (15), 155430 (2012).

Schöllkopf, W., S. Gewinner, W. Erlebach, H. Junkes, A. Liedke, G. von Helden, W. Zhang, G. Meijer, H. Bluem, V. Christina, M.D. Cole, J. Ditta, D. Dowell, K. Jordan, R. Lange, J.H. Park, J. Rathke, T. Schultheiss, A.M.M. Todd, L.M. Young, U. Lehnert, P. Michel, W. Seidel, R. Wünsch and S.C. Gottschalk: Status of the Fritz Haber Institute THz FEL. In: Proceedings of 33rd International Free Electron Laser Conference, Shanghai, China. TUPB30 (2012). ISBN 978-3-95450-117-5.

Seifert, J., E. Meyer, H. Winter and H. Kuhlenbeck: Surface termination of an ultrathin V_2O_3-film on Au(111) studied via ion beam triangulation. Surface Science **606** (9-10), L41-L44 (2012).

Shaikhutdinov, S.K.: Surface Structures of Gold Nanoparticles. In: Gold nanoparticles for physics, chemistry and biology, (Eds.) C. Louis and O. Pluchery. Imperial College Press, London, 199-231 (2012). ISBN 978-1-84816-806-0.

Shaikhutdinov, S.K. and H.-J. Freund: Ultrathin Oxide Films on Metal Supports: Structure-Reactivity Relations. Annual Review of Physical Chemistry **63**, 619-633 (2012).

Shao, X., Y. Cui, W.-D. Schneider, N. Nilius and H.-J. Freund: Growth of Two-Dimensional Lithium Islands on CaO(001) Thin Films. The Journal of Physical Chemistry C **116** (33), 17980-17984 (2012).

Shao, X., N. Nilius and H.-J. Freund: Crossover from two- to three-dimensional gold particle shapes on CaO films of different thicknesses. Physical Review B **85** (11), 115444 (2012).

Shao, X., N. Nilius and H.-J. Freund: Li/Mo codoping of CaO films: A means to tailor the equilibrium shape of Au deposits. Journal of the American Chemical Society **134** (5), 2532-2534 (2012).

Simon, G.H., M. Heyde and H.-J. Freund: Imaging and manipulation of adatoms on an alumina surface by noncontact atomic force microscopy. Journal of Physics: Condensed Matter **24** (8), 084007 (2012).

Stavale, F., N. Nilius and H.-J. Freund: Cathodoluminescence of near-surface centres in Cr-doped MgO(001) thin films probed by scanning tunnelling microscopy. New Journal of Physics **14** (3), 033006 (2012).

Stavale, F., N. Nilius and H.-J. Freund: Probing the luminescence of single Eu_2O_3 nano-islands on MgO(001) with scanning tunneling microscopy. Applied Physics Letters **101** (1), 013109 (2012).

Stavale, F., L. Pascua, N. Nilius and H.-J. Freund: From embedded nanoislands to thin films: Topographic and optical properties of europium oxide on MgO(001) films. Physical Review B **86** (8), 085448 (2012).

Stavale, F., X. Shao, N. Nilius, H.-J. Freund, S. Prada, L. Giordano and G. Pacchioni: Donor Characteristics of Transition-Metal-Doped Oxides: Cr-Doped MgO versus Mo-Doped CaO. Journal of the American Chemical Society **134** (28), 11380-11383 (2012).

Todd, A.M.M., H. Bluem, J. Ditta, D. Dowell, K. Jordan, R. Lange, H. Loos, J.H. Park, J. Rathke, L.M. Young, W. Erlebach, S. Gewinner, G. von Helden, H. Junkes, A. Liedke, G. Meijer, W. Schöllkopf, W. Zhang, U. Lehnert, P. Michel, W. Seidel, R. Wünsch and S.C. Gottschalk: Commissioning of the Fritz Haber Institute Mid-IR FEL. In: Proceedings of 3rd International Particle Accelerator Conference, IPAC'12, New Orleans, Louisiana, USA. TUPPP087 (2012). ISBN 978-3-95450-115-1.

Wang, H., H. Ariga, R. Dowler, M. Sterrer and H.-J. Freund: Surface science approach to catalyst preparation - Pd deposition onto thin $Fe_3O_4(111)$ films from $PdCl_2$ precursor. Journal of Catalysis **286**, 1-5 (2012).

Wang, H., W. Kaden, R. Dowler, M. Sterrer and H.-J. Freund: Model oxide-supported metal catalysts – comparison of ultrahigh vacuum and solution based preparation of Pd nanoparticles on a single-crystalline oxide substrate. Physical Chemistry Chemical Physics **14** (32), 11525-11533 (2012).

Window, A.J., A. Hentz, D.C. Sheppard, G.S. Parkinson, D.P. Woodruff, W. Unterberger, T.C.Q. Noakes, P. Bailey, M.V. Ganduglia-Pirovano and J. Sauer: The structure of epitaxial V_2O_3 films and their surfaces: A medium energy ion scattering study. Surface Science **606** (21-22), 1716-1727 (2012).

Włodarczyk, R., M. Sierka, J. Sauer, D. Loeffler, J. Uhlrich, X. Yu, B. Yang, I. Groot, S.K. Shaikhutdinov and H.-J. Freund: Tuning the electronic structure of ultrathin crystalline silica films on Ru(0001). Physical Review B **85** (8), 085403 (2012).

Yang, B., W. Kaden, X. Yu, A. Boscoboinik, Y. Martynova, L. Lichtenstein, M. Heyde, M. Sterrer, R. Włodarczyk, M. Sierka, J. Sauer, S.K. Shaikhutdinov and H.-J. Freund: Thin silica films on Ru(0001): Monolayer, bilayer and three-dimensional networks of $[SiO_4]$ tetrahedra. Physical Chemistry Chemical Physics **14** (32), 11344-11351 (2012).

Yang, B., Y. Pan, X. Lin, N. Nilius, H.-J. Freund, C. Hulot, A. Giraud, S. Blechert, S. Tosoni and J. Sauer: Stabilizing Gold Adatoms by Thiophenyl Derivatives: A Possible Route toward Metal Redispersion. Journal of the American Chemical Society **134** (27), 11161-11167 (2012).

Yu, X., B. Yang, A. Boscoboinik, S.K. Shaikhutdinov and H.-J. Freund: Support effects on the atomic structure of ultrathin silica films on metals. Applied Physics Letters **100** (15), 151608 (2012).

Yudanov, l.V., A. Genest, S. Schauermann, H.-J. Freund and N. Rösch: Size Dependence of the Adsorption Energy of CO on Metal Nanoparticles: A DFT Search for the Minimum Value. Nano Letters **12** (4), 2134-2139 (2012).

Doctoral Theses

Kreikemeyer-Lorenzo, D.: Photoelectron diffraction determination of adsorbate structures. University of Warwick 2012.

Lichtenstein, L.: The structure of two-dimensional vitreous silica. Freie Universität Berlin 2012.

Pieper, B.: Festkörpergestützte Modellsysteme zur Untersuchung von Membranproteinen mittels ESR-Spektroskopie. Technische Universität Berlin 2012.

Wang, H.: Surface science approach to catalyst preparation: Palladium deposition onto iron oxide films from the liquid phase. Freie Universität Berlin 2012.

Master's Thesis

Koschnick, D.: Mobile Augmented Reality-basierte Assistenzfunktionen. Hochschule für Technik und Wirtschaft Berlin 2012.

Bachelor Thesis

Gyemant, B.: Erweiterung des Online-Fotoportals Picambio um eine Schnittstelle zu mobilen Endgeräten mit prototypischer Umsetzung einer nativen iPhone-Applikation. Beuth Hochschule für Technik Berlin 2012.

2013

Aleksandrov, H.A., F. Viñes, W. Ludwig, S. Schauermann and K.M. Neyman: Tuning the Surface Chemistry of Pd by Atomic C and H: A Microscopic Picture. Chemistry - A European Journal **19** (4), 1335-1345 (2013).

Benedetti, S., F. Stavale, S. Valeri, C. Noguera, H.-J. Freund, J. Goniakowski and N. Nilius: Steering the Growth of Metal Ad-particles via Interface Interactions Between a MgO Thin Film and a Mo Support. Advanced Functional Materials **23** (1), 75-80 (2013).

Boscoboinik, A., X. Yu, E. Emmez, B. Yang, S.K. Shaikhutdinov, F.D. Fischer, J. Sauer and H.-J. Freund: Interaction of Probe Molecules with Bridging Hydroxyls of Two-Dimensional Zeolites: A Surface Science Approach. The Journal of Physical Chemistry C **117** (26), 13547-13556 (2013).

Boscoboinik, A., X. Yu, S.K. Shaikhutdinov and H.-J. Freund: Preparation of an Ordered Ultra-thin Aluminosilicate Film Composed of Hexagonal Prisms Forming a Percolated Network. Microporous and Mesoporous Materials, in press.

Boscoboinik, A., X. Yu, B. Yang, S.K. Shaikhutdinov and H.-J. Freund: Building blocks of zeolites on an aluminosilicate ultra-thin film. Microporous and Mesoporous Materials **165**, 158-162 (2013).

Brown, M.A., A.B. Redondo, M. Sterrer, B. Winter, G. Pacchioni, Z. Abbas and J. van Bokhoven: Measure of Surface Potential at the Aqueous-Oxide Nanoparticle Interface by XPS from a Liquid Microjet. Nano Letters, in press.

Carrasco, E., A. Aumer, J.F. Gomes, Y. Fujimori and M. Sterrer: Vibrational spectroscopic observation of ice dewetting on MgO(001). Chemical Communications **49** (39), 4355-4357 (2013).

Cui, Y., K. Huang, N. Nilius and H.-J. Freund: Charge competition with oxygen molecules determines the growth of gold particles on doped CaO films. Faraday Discussions **162**, 153-163 (2013).

Cui, Y., X. Shao, M. Baldofski, J. Sauer, N. Nilius and H.-J. Freund: Adsorption, Activation, and Dissociation of Oxygen on Doped Oxides. Angewandte Chemie International Edition **52** (43), 11426-11426 (2013).

Cui, Y., X. Shao, M. Baldofski, J. Sauer, N. Nilius and H.-J. Freund: Bindung, Aktivierung und Dissoziation von Sauerstoff an dotierten Oxiden. Angewandte Chemie **125** (43), 11638-11638 (2013).

Freund, H.-J.: Modelle für heterogene Katalysatoren: Wie viel Komplexität ist notwendig? Schöningh, Paderborn (2013). ISBN 978-3-506-77728-7.

Freund, H.-J., M. Heyde, N. Nilius, S. Schauermann, S.K. Shaikhutdinov and M. Sterrer: Model studies on heterogeneous catalysts at the atomic scale: From supported metal particles to two-dimensional zeolites. Journal of Catalysis, in press.

Gonchar, A. and T. Risse: Characterisation of paramagnetic Mo impurities on MgO(100) single-crystalline films grown on Mo(100). Molecular Physics **111** (18-19), 2708-2716 (2013).

Gottschalk, S.C., T.E. DeHart, R.N. Kelly, M.A. Offenbacker, A.S. Valla, H. Bluem, D. Dowell, J. Rathke, A.M.M. Todd, S. Gewinner, H. Junkes, G. Meijer, W. Schöllkopf, W. Zhang and U. Lehnert: Design and Performance of the Wedged Pole Hybrid Undulator for the Fritz-Haber-Institut IR FEL. In: Proceedings of 34th International Free Electron Laser Conference, Nara, Japan. THPD13 (2013). ISBN 978-3-95450-123-6.

Guimond, S., D. Göbke, J.M. Sturm, Y. Romanyshyn, H. Kuhlenbeck, M. Cavalleri and H.-J. Freund: Well-Ordered Molybdenum Oxide Layers on Au(111): Preparation and Properties. The Journal of Physical Chemistry C **117** (17), 8746-8757 (2013).

Heyde, M.: Structure and Motion of a 2D Glass. Science **342** (6155), 201-202 (2013).

Heyde, M., G.H. Simon and L. Lichtenstein: Resolving oxide surfaces – From point and line defects to complex network structures. Physica Status Solidi B **250** (5), 895-921 (2013).

Junkes, H., W. Erlebach, S. Gewinner, U. Hoppe, A. Liedke, G. Meijer, W. Schöllkopf, G. von Helden, M. Wesemann, A.M.M. Todd, L.M. Young, H. Bluem, D. Dowell, R. Lange and S.B. Webb: Integration the FHI-FEL into the FHI Research Environment - Design and Implementation Aspects. In: Proceedings of 35th International Free Electron Laser Conference, Manhattan, USA. In press.

Junkes, H., W. Schöllkopf, M. Wesemann and R. Lange: Design and Implementation Aspects of the Control System at the FHI FEL. In: Proceedings of 14th International Conference on Accelerator & Large Experimental Physics Control Systems, San Francisco, CA, USA. In press.

Karslıoğlu, O., S. Xin, H. Kuhlenbeck and H.-J. Freund: Mo+TiO_2(110) Mixed Oxide Layer: Structure and Reactivity. Topics in Catalysis **56** (15-17), 1389-1403 (2013).

Kreikemeyer-Lorenzo, D., W. Unterberger, R. Blume, M. Hävecker, T. Rocha, A. Knop-Gericke, R. Schlögl, T.J. Lerotholi, D.A. Duncan and D.P. Woodruff: Quantitative adsorbate structure determination under catalytic reaction conditions. Physical Review B **87** (12), 125420 (2013).

Kreikemeyer-Lorenzo, D., W. Unterberger, D.A. Duncan, T.J. Lerotholi and D.P. Woodruff: The local structure of the azobenzene/aniline reaction intermediate on TiO_2(110). Surface Science **613**, 40-47 (2013).

Kuhlenbeck, H., S.K. Shaikhutdinov and H.-J. Freund: Well-Ordered Transition Metal Oxide Layers in Model Catalysis – A Series of Case Studies. Chemical Reviews **113** (6), 3986-4034 (2013).

Martynova, Y., B. Liu, M. McBriarty, I. Groot, M.J. Bedzyk, S.K. Shaikhutdinov and H.-J. Freund: CO oxidation over ZnO films on Pt(111) at near-atmospheric pressures. Journal of Catalysis **301**, 227-232 (2013).

Martynova, Y., S.K. Shaikhutdinov and H.-J. Freund: CO oxidation on metal-supported ultrathin oxide films: What makes them active? ChemPhysChem **5** (8), 2162-2166 (2013).

Martynova, Y., M. Soldemo, J. Weissenrieder, S. Sachert, S. Polzin, W. Widdra, S.K. Shaikhutdinov and H.-J. Freund: CO Oxidation Over Monolayer Manganese Oxide Films on Pt(111). Catalysis Letters **143** (11), 1108-1115 (2013).

Menzel, D., K.H. Kim, D. Mulugeta and K. Watanabe: Case Studies in Photochemistry on Metal Nanoparticles. Journal of Vacuum Science and Technology A **31** (5), 050817 (2013).

Nilius, N., T. Risse, S.K. Shaikhutdinov, M. Sterrer and H.-J. Freund: Model catalysts based on Au clusters and nano particles. In: Structure and Bonding, Springer, Berlin. In press.

Nilius, N., M. Sterrer, S.K. Shaikhutdinov, D. Menzel and H.-J. Freund: Model systems in catalysis for energy economy. In: Chemical Energy Storage, (Ed.) R. Schlögl. De Gruyter, Berlin, 329-352 (2013). ISBN 978-3-11-026407-4.

Pacchioni, G. and H.-J. Freund: Electron Transfer at Oxide Surfaces. The MgO Paradigm: from Defects to Ultrathin Films. Chemical Reviews **113** (6), 4035-4072 (2013).

Pan, Y., Y. Cui, N. Nilius and H.-J. Freund: Gold Adsorption on CeO_2 Thin Films Grown on Ru(0001). The Journal of Physical Chemistry C **117** (42), 21879-21885 (2013).

Pan, Y., N. Nilius, H.-J. Freund, J. Paier, C. Penschke and J. Sauer: Titration of Ce_3+ions in the CeO_2(111) Surface by Au ad-Atoms. Physical Review Letters, in press.

Peter, M., S. Adamovski, J.M. Flores Camacho and S. Schauermann: Energetics of Elementary Reaction Steps Relevant for CO Oxidation: CO and O_2 Adsorption on Model Pd Nanoparticles and Pd(111). Faraday Discussion **162**, 341-354 (2013).

Peter, M., J.M. Flores Camacho, S. Adamovski, L.K. Ono, K.-H. Dostert, C. O'Brien, B. Roldan Cuenya, S. Schauermann and H.-J. Freund: Trends in der Bindungsstärke von Oberflächenspezies auf Nanopartikeln: Wie verändert sich die Adsorptionsenergie mit der Partikelgröße? Angewandte Chemie **125** (19), 5282-5287 (2013).

Peter, M., J.M. Flores Camacho, S. Adamovski, L.K. Ono, K.-H. Dostert, C. O'Brien, B. Roldan Cuenya, S. Schauermann and H.-J. Freund: Trends in the Binding Strength of Surface Species on Nanoparticles: How Does the Adsorption Energy Scale with the Particle Size? Angewandte Chemie International Edition **52** (19), 5175-5179 (2013).

Ringleb, F., M. Sterrer and H.-J. Freund: Preparation of Pd-MgO model catalysts by deposition of Pd from aqueous precursor solutions onto Ag(0 0 1)-supported MgO(0 0 1) thin films. Applied Catalysis A, in press.

Ritter, C., M.Z. Baykara, B. Stegemann, M. Heyde, K. Rademann, J. Schroers and U.D. Schwarz: Nonuniform friction-area dependency for antimony oxide surfaces sliding on graphite. Physical Review B **88** (4), 045422 (2013).

Savara, A.A., W. Ludwig, K.-H. Dostert and S. Schauermann: Temperature dependence of the 2-butene hydrogenation over supported Pd nanoparticles and Pd(111). Journal of Molecular Catalysis A **377**, 137-142 (2013).

Savara, A.A., W. Ludwig and S. Schauermann: Kinetic Evidence for a Non-Langmuir-Hinshelwood Surface Reaction: H/D Exchange over Pd Nanoparticles and Pd(111). ChemPhysChem **14** (8), 1686-1695 (2013).

Schauermann, S., N. Nilius, S.K. Shaikhutdinov and H.-J. Freund: Nanoparticles for Heterogeneous Catalysis: New Mechanistic Insights. Accounts of Chemical Research **46** (8), 1673-1681 (2013).

Schmidt, T., A. Sala, H. Marchetto, E. Umbach and H.-J. Freund: First experimental proof for aberration correction in XPEEM: Resolution, transmission enhancement, and limitation by space charge effects. Ultramicroscopy **126**, 23-32 (2013).

Schöllkopf, W., S. Gewinner, W. Erlebach, G. Heyne, H. Junkes, A. Liedke, V. Platschkowski, G. von Helden, W. Zhang, G. Meijer, H. Bluem, M. Davidsaver, D. Dowell, K. Jordan, R. Lange, H. Loos, J. Park, J. Rathke, A.M.M. Todd, L.M. Young, U. Lehnert, P. Michel, W. Seidel, R. Wünsch and S.C. Gottschalk: First Lasing of the IR FEL at the Fritz-Haber-Institut Berlin. In: Proceedings of 34th International Free Electron Laser Conference, Nara, Japan. MOOB0 (2013). ISBN 978-3-95450-123-6.

Shaikhutdinov, S.K. and H.-J. Freund: Metal-Supported Aluminosilicate Ultrathin Films as a Versatile Tool for Studying the Surface Chemistry of Zeolites. ChemPhysChem **14** (1), 71-77 (2013).

Shaikhutdinov, S.K. and H.-J. Freund: Ultrathin Silica Films on Metals: The Long and Winding Road to Understanding the Atomic Structure. Advanced Materials **25** (1), 49-67 (2013).

Stavale, F., N. Nilius and H.-J. Freund: Lithium-molybdate nanostructures grown on the Mo(001) surface. Surface Science **609**, 78-84 (2013).

Stavale, F., L. Pascua, N. Nilius and H.-J. Freund: Morphology and Luminescence of ZnO Films Grown on a Au(111) Support. The Journal of Physical Chemistry C **117** (20), 10552-10557 (2013).

Sterrer, M. and H.-J. Freund: Towards realistic surface science models of heterogeneous catalysts - Influence of support hydroxylation and catalyst preparation method. Catalysis Letters **143** (5), 375-385 (2013).

Stiehler, C., Y. Pan, W.-D. Schneider, P. Koskinen, H. Häkkinen, N. Nilius and H.-J. Freund: Electron quantization in arbitrarily shaped gold islands on MgO thin films. Physical Review B **88** (11), 115415 (2013).

Stuckenholz, S., C. Büchner, G. Thielsch, M. Heyde and H.-J. Freund: A portable quartz micro balance for physical vapor deposition techniques. Review of Scientific Instruments **84** (8), 085118 (2013).

Yang, B., J.A. Boscoboinik, X. Yu, S.K. Shaikhutdinov and H.-J. Freund: Patterned Defect Structures Predicted for Graphene Are Observed on Single-Layer Silica Films. Nano Letters **13** (9), 4422-4427 (2013).

Yang, B., E. Emmez, W. Kaden, X. Yu, J.A. Boscoboinik, M. Sterrer, S.K. Shaikhutdinov and H.-J. Freund: Hydroxylation of Metal-Supported Sheet-Like Silica Films. The Journal of Physical Chemistry C **117** (16), 8336-8344 (2013).

Habilitation

Sterrer, M.: Katalysatorpräparationen - Ein Modellansatz. Technische Universität Berlin 2013.

Doctoral Thesis

Dowler, R.: Preparation of Catalysts on Model Supports Using Wet Chemical Methods and the Construction of a Device for the in-situ Measurement of Sum Frequency Generation Spectroscopy at the Solid-Liquid Interface. Technische Universität Berlin 2013.

Karslioglu, O.: Structural and chemical characterizations of Mo-Ti mixed oxide layers. Humboldt-Universität zu Berlin 2013.

Martynova, Y.: CO oxidation on metal supported ultrathin oxide films. Technische Universität Berlin 2013.

Ringleb, F.: Preparation and Application of Surface Science Model Catalysts in Realistic Conditions. Freie Universität Berlin 2013.

Sala, A.: Characterization of iron oxide thin films as a support for catalytically active nanoparticles. Freie Universität Berlin 2013.

Yu, X.: Ultra-thin silicate films on metal single crystals. Technische Universität Berlin 2013.

Master's Thesis

Ronneburg, H.: Combination of Reactivity Experiments with High-Resolution Scanning Probe Microscopy Studies. Freie Universität Berlin 2013.

Bachelor Thesis:

Abdou, J.: Modellstudien zur Katalysatorpräparation mit einkristallinen Oxidsubstraten. Technische Universität Berlin, 2013.

Invited Talks of the Members of the Department of Chemical Physics

Hans-Joachim Freund

Jan 2012	8th International Workshop on Oxide Surfaces (IWOX-VIII), Baqueira-Beret, Spain *Thin Oxide Films: The Expected and the Unexpected*
Jan 2012	Lecture, Toyota Technology Institute, Nagoya, Japan *Model Catalysts: From Supports to Supported Nanoparticles*
Feb 2012	3rd GRI Symposium on Cluster Science Functionality of Clusters (GRIS III), Nagoya, Japan *Electronics, Thermodynamics, and Kinetics of Supported Clusters*
Feb 2012	Lecture, Wacker Chemie AG, Burghausen, Germany *Ultradünne Oxidschichten: Die "Physik" der Katalyse*
Feb 2012	International Workshop on Advanced Materials (IWAM), Ras Al Khaimah, United Arab Emirates *Ultrathin Oxide Films Grown on Metals: New Thoughts on Catalytic Materials*
Mar 2012	UCI Seminar, Irvine, CA, USA *Thin Oxide Films and the "Physics" of Catalysis: The Expected and the Unexpected*
Mar 2012	243rd ACS National Spring Meeting, San Diego, CA, USA *Ultra-Thin Film Oxides: Materials with Tailored Properties*
May 2012	Robert-Bunsen-Vorlesung, Zurich, Switzerland *Models in Catalysis: The Expected and the Unexpected*
Jun 2012	Physikalisch-Chemisches Kolloquium, Ludwig-Maximilians-Universität München, Munich, Germany *Model Catalyst Design: A Perspective at the Atomic Level*
Jul 2012	15th International Conference on Catalysis, ICC 15, Munich, Germany *Model in Heterogeneous Catalysis: What Has Been Learned?*
Jul 2012	16th International Conference on Solid Films and Surfaces, ICSFS 16, Genoa, Italy *Model Catalyst Studies Based on Thin Oxide Films*
Jul 2012	International Workshop on Nanomaterials and Nanodevices, Beijing, China *Ultrathin Oxide Films: The "Physics" of Catalysis*
Jul 2012	International Workshop on Nanomaterials and Nanodevices, Xining, China *Nanoparticles as Models for Heterogeneous Catalysts: An Atomic view*
Jul 2012	Klassensitzung, North Rhine-Westphalian Academy of Sciences, Humanities and the Arts, Düsseldorf, Germany *Modelle für heterogene Katalysatoren: Wie viel Komplexität ist notwendig?*
Jul 2012	Lecture, Faculty of Chemistry and Biochemistry, Ruhr Universität Bochum, Bochum, Germany *Thin Films as Model Materials in Catalysis: A Perspective at the Atomic Level*

Aug 2012	Symposium on Progress in Electronic and Vibrational Spectroscopy of Catalytic Materials and Catalytic Reactions, 244th ACS National Meeting and Exposition, Philadelphia, PA, USA *Three-Dimensional Silicolites and Zeolite: A New Field for Surface Science*
Aug 2012	Symposium on Spectroscopy and Microscopy in Catalysis: In Situ Age, 244th ACS National Meeting and Exposition, Philadelphia, PA, USA *Ultra-Thin Film Oxides: Grown on Metals: Materials with Tailored Properties*
Sep 2012	International Conference on Theory and Applications of Computational Chemistry, TACC-2012, Pavia, Italy *Models in Catalysis: Synergy Between Experiment and Theory*
Sep 2012	The 6th International Conference on Gold Science Technology and its Application, Gold 2012, Tokyo, Japan *The Surface Science of Supported Au Nanoparticles and its Relation to Catalysis*
Oct 2012	Lecture, Dinesh O. Shah Annual Lectureship in Surface Science, Chemical Engineering Department, University of Florida, Gainesville, FL, USA *Ultrathin Oxide Films: The Expected and the Unexpected*
Oct 2012	Lecture, Blaise Pascal Medal Award Ceremony, European Academy of Sciences, Liege, Belgium *Ultrathin Oxide Films: The Expected and the Unexpected*
Jan 2013	Seminar, Particles and Catalysis Research Group, School of Chemical Sciences and Engineering, University of New South Wales, Sydney, Australia *Oxide Supported Nanoclusters*
Feb 2013	Lecture, Monash Chemical Society Lecture, Monash University, Melbourne, Australia *Model Catalyst Design: A Material Science Perspective at the Atomic Level*
Feb 2013	Seminar, Department of Chemistry, Otago University, Dunedin, New Zealand *Photochemistry and Photodesorption from Oxide Supported Metal Nanoparticles*
Feb 2013	AMN-6 Conference, Auckland, New Zeeland *From Oxide Film Supported Nanoparticles to Two-Dimensional Zeolite Films*
Feb 2013	Seminar, Temple University, Philadelphia, PA, USA *Model Studies on Heterogeneous Catalysts at the Atomic Scale: From Supported Metal Particles to Two-Dimensional Zeolites*
Mar 2013	Joint Meeting of UniCat and Northwestern University, Berlin, Germany *Model Catalysts for Oxidation Reactions: Are Dopants in Simple Oxides Relevant?*
Mar 2013	528th Wilhelm-und-Else-Heraeus-Seminar, Bad Honnef, Germany *Fundamental Studies Using Surface Science Techniques*

Apr 2013	245th ACS National Meeting, Symposium in Honor of Prof. Alex Bell, New Orleans, LA, USA *Thin Films as Model Materials in Catalysis: A Perspective at the Atomic Level*
Apr 2013	245th ACS National Meeting, Symposium on Arthur W. Adamson Award, New Orleans, LA, USA *Ultra-Thin Oxide Films as Designable Model Catalysts*
May 2013	Cardiff Chemistry Conference, Cardiff, UK *Model Studies on Heterogeneous Catalysts at the Atomic Scale: From Supported Metal Particles to Two-Dimensional Zeolites*
Jun 2013	10 Years CeNTech, CeNTech Day, Münster, Germany *From Oxide Film Supported Nanoparticles to Two-Dimensional Zeolite Films*
Jun 2013	7th World Congress on Oxidation Catalysis, Saint Louis, MO, USA *Oxygen Activation on Oxide Surfaces*
Jul 2013	NIMS Conference, Tsukuba City, Japan *Model Catalysts Design: A Material Science Perspective at the Atomic Level*
Jul 2013	International Workshop on Nanomaterials and Nanodevices, Beijing, China *Model Studies on Heterogeneous Catalysts at the Atomic Scale: From Supported Metal Particles to Two-Dimensional Zeolites*
Jul 2013	Hayashi Conference: Next Decades of Surface Science, Hayama, Japan *From Oxide Film Supported Nanoparticles to Two-Dimensional Zeolite Films*
Jul 2013	Meeting on Catalysis, Telluride, CO, USA *From Oxide Film Supported Nanoparticles to Two-Dimensional Zeolite Films*
Aug 2013	The 16th International Symposium on Relations between Homogeneous and Heterogeneous Catalysis, Sapporo, Japan *Model Studies on Heterogeneous Catalysts at the Atomic Scale*
Sep 2013	8th German-Brazilian Workshop, Bamberg, Germany *From Pure Metal Supported Oxide Films to Two-Dimensional Zeolite Films*

Markus Heyde

Nov 2011	493rd Wilhelm-und-Else-Heraeus-Seminar on Latest Developments in Scanning Probe Techniques Focused on Nanotechnology, Bad Honnef, Germany *AFM - From Nanostructuring Under Ambient Conditions to the Ultimate Atomic Resolution and Beyond*
Mar 2012	3rd International Workshop on Advanced Atomic Force Microscopy Techniques (AAFMT 2012), Karlsruhe, Germany *The Structure of 2D Vitreous Silica*

Apr 2012	Seminar, Institute of Physics, Academy of Sciences, Prague, Czech Republic *Resolving Amorphous Solids - The Atomic Structure of a 2D Glass*
Oct 2012	AVS 59th International Symposium and Exhibition - Nanoscale Science and Technology Division, Tampa, FL, USA *Resolving Amorphous Solids - The Atomic Structure of Glass*
Nov 2012	Seminar, Department of Mechanical Engineering, Yale University, New Haven, CT, USA *The Structure of 2D Crystalline and Vitreous Silica*
Mar 2013	DPG-Frühjahrstagung, Post Deadline Session of the Surface Science Division, Ratisbon, Germany *The Structure of Glass*
Jun 2013	Condensed Matter Physics Seminar, University of Oxford, Oxford, UK *The Structure of Vitreous Silica*
Sep 2013	246th ACS National Meeting and Exposition, Indianapolis, IN, USA *Resolving Two-Dimensional Complex Oxide Film Structures*
Sep 2013	Workshop on Forces and Photons in Molecular Tunnelling Junctions, Berlin, Germany *The Impact of Atomic Force Microscopy and Spectroscopy to Illuminate Complex Oxide Surfaces*

Dietrich Menzel

Mar 2012	3S'12 (International Symposium on Surface Science), St. Christoph, Austria *Photochemistry on Nanoparticles. An Overview of a Typical System*
Jul 2012	DIET-13 Conference, Stratford-on-Avon, UK *Photochemistry at Metal Nanoparticles*
Jul 2012	ICNT-2012 (International Conference NanoScience and Technology), Paris, France *Photochemistry on Nanoparticles: A Detailed Investigation of an Exemplary System*
Sep 2012	Workshop of the Chemical Physics Department of the FHI, Döllnsee, Germany *Ultrafast Charge Transfer at Graphene Surfaces*
Oct 2012	AVS International Symposium, American Vacuum Society, Tampa, FL, USA *Surface Photochemistry on Compact Metal Crystals and on Metallic Nanoparticles*
Jan 2013	EWEG 2013 (European Workshop on Epitaxial Graphene), Aussois, France *Ultrafast Charge Transfer at Graphene Surfaces*
Sep 2013	IVC/ICSS-19 (International Conference Solid Surface), Paris, France *Ultrafast Charge Transfer at Graphene Surfaces with Widely Varying Coupling to Their Substrates*

Sep 2013	8th German-Brazilian Workshop, Bamberg, Germany *Ultrafast Charge Transfer at Graphene Monolayers with Strongly Varying Coupling Strength*
Oct 2013	AVS International Symposium, American Vacuum Society, Long Beach, NY, USA *Ultrafast Charge Transfer at Graphene Monolayers on Ni(111) and Other Substrates*

Niklas Nilius

Sep 2011	28th European Conference on Surface Science (ECOSS-28), Wroclaw, Poland *Tailoring the Properties of Oxide Materials via Doping*
Sep 2011	15th International Workshop on Surface Physics, Ladek Zdroj, Poland *Optical Spectroscopy at the Nanometer Scale Using a Scanning Tunnelling Microscope*
Nov 2011	Seminar, Department of Physics, Nano-Optics Group, University of Oldenburg, Oldenburg, Germany *Das STM: Eine spektroskopische Werkzeugkiste*
Dec 2011	Seminar, Department of Chemistry, Catalysis Research Center, Technische Universität München, Munich, Germany *Doped Oxide Materials: Their Electronic, Optical and Adsorption Properties*
Jan 2012	Seminar, Department of Physics (AG Jose Ignacio Pascual), Freie Universität Berlin, Berlin, Germany *Doping of Oxide Materials: An STM study*
Mar 2012	Statusseminar SFB 855 on Magnetoelectric Composites, Sankelmark, Germany *Tailoring the Optical Properties of Oxide Materials via Doping*
May 2012	SFB 616 Seminar, University of Duisburg-Essen, Duisburg, Germany *Doped Oxide Materials: Their Electronic, Optical and Adsorption Properties*
Jul 2012	International Conference on Nanoscience and Technology, Paris, France *Tailoring the Properties of Oxide Materials via Doping*
Jul 2012	Seminar, Institute for Nanoscience, Pierre and Marie Curie University, Paris, France *Exploring Polarity at the Nanoscale: MgO(111) Islands on Gold Supports*
Sep 2012	CRC-FHI Joint Meeting on Complex Surfaces in Material Science, Berlin, Germany *Tailoring the Shape of Metal Particles on Oxide Supports*
Oct 2012	Technologies and Measurements on Atomic Scale, Surface Science School, Khosta, Russia *STM Studies on Model Catalysts*

Dec 2012	German-Korean Symposium on Nano-Optics and Nano-Technology, Delmenhorst, Germany *Optical Properties of Doped Oxide Materials*
Feb 2013	Physikalisches Kolloquium, University of Oldenburg, Oldenburg, Germany *Functionalized Oxide Surfaces in the STM: From Polarity Tuning to Doping*
Apr 2013	162. Faraday Discussion on Fabrication, Structure and Reactivity of Anchored Nanoparticles, Berlin, Germany *Competition with Oxygen Molecules Determines the Growth of Gold Particles on Doped CaO Films*

Swetlana Schauermann

Mar 2012	Nanoscience Workshop, National University, Yokohama, Japan *Hydrocarbon Conversions on Oxide Supported Model Catalysts*
Mar 2012	Surface Science Workshop, University of Tokyo, Tokyo, Japan *Interaction of Gas Phase Molecules with Nanostructured Model Supported Catalysts: Thermodynamics and Kinetics*
Mar 2012	45. Jahrestreffen Deutscher Katalytiker, Weimar, Germany *Interaction of Gas Phase Molecules with Nanostructured Model Supported Catalysts: Thermodynamics and Kinetics*
May 2012	German-Russian Conference on Fundamentals and Applications of Nanoscience, Berlin, Germany *Olefin Conversions on Model Supported Catalysts: New Mechanistic Insights*
Sep 2012	14th International Conference on Vibrations at Surfaces, Kobe, Japan *Surface Vibrational Spectroscopy of Unsaturated Hydrocarbons on Model Supported Catalysts*
Oct 2012	Seminar, Vienna University of Technology, Vienna, Austria *Model Studies on Nanostructured Supported Catalysts*
Dec 2012	Lorentz-Center Workshop on Elementary Reactive Processes at Surfaces, Leiden, The Netherlands *Role of Subsurface Hydrogen Diffusion in Olefin Hydrogenation*
Feb 2013	Seminar, Technische Universität Berlin, Berlin, Germany *Heterogene Katalyse auf Nanostrukturierten Modellkatalysatoren: Mechanismen, Kinetik und Thermodynamik*
Mar 2013	DPG-Frühjahrstagung, Sektion Kondensierte Materie, Ratisbon, Germany *Interaction of Gas Phase Molecules with Nanostructured Model Supported Catalysts: Thermodynamics and Kinetics*
Mar 2013	Winterschool on Chemical Reaction Dynamics at Surfaces, Max-Planck-EPFL-Center for Molecular Nanoscience and Technology, Ringberg Castle, Germany *Kinetics of Surface Reactions by Molecular Beam Techniques*

Apr 2013	245th ACS National Meeting and Exposition, New Orleans, LA, USA *Interaction of Gas Phase Molecules with Nanostructured Model Supported Catalysts: Thermodynamics and Kinetics*
Apr 2013	162. Faraday Discussion on Fabrication, Structure and Reactivity of Anchored Nanoparticles, Berlin, Germany *Energetics of Elementary Reaction Steps Relevant for CO Oxidation: CO and O_2 Adsorption on Model Pd Nanoparticles and Pd(111)*
Apr 2013	Gordon Research Conference on Chemical Reactions at Surfaces, Les Diablerets, Switzerland *Hydrocarbon Conversions on Supported Model Catalysts*
Jun 2013	Seminar, ETH Zurich, Zurich, Switzerland *Elementary Steps in Surface Reactions: Mechanisms, Kinetics and Thermodynamics*
Jul 2013	Seminar, Heidelberg University, Heidelberg, Germany *Elementarschritte von Oberflächenreaktionen: Mechanismen, Kinetik und Thermodynamik*
Sep 2013	Workshop on VUV FELs in Molecular, Cluster and Surface Science: A Future Frontier for Energy Research, Dalian, China *Chemical Reactions on Nanostructured Model Catalysts*

Wolf-Dieter Schneider

Sep 2011	Workshop of the Chemical Physics Department of the FHI, Hasenwinkel, Germany *Growth and Characterization of Fullerene Nanocrystals on NaCl/Au(111)*
Jan 2012	Symposium on Surface and Nano Science 2012 (SSNS'12), Shizukuishi, Japan *Growth and Characterization of Fullerene Nanocrystals on NaCl/Au(111)*
May 2012	CEITEC, Brno University of Technology, Brno, Czech Republic *Luminescence Experiments with the Scanning Tunneling Microscope*
Jun 2012	CECAM Workshop on Control and Enhancement of Superconductivity in Conventional and High Tc Nanostructures, Lausanne, Switzerland *Dynamical Coulomb Blockade, Quantum Oscillations, and Superconductivity in Nanosized Electrical Contacts*
Jun 2012	International Conference NanoSEA, Cagliari, Italy *Dynamical Coulomb Blockade, Quantum Oscillations, and Superconductivity in Nanosized Electrical Contacts*
Jul 2012	CECAM Workshop on Theoretical Challenges in the Electronic Structure of Clusters and Nanoparticles, Lausanne, Switzerland *Dynamical Coulomb Blockade, Quantum Oscillations, and Superconductivity in Nanosized Electrical Contacts*

Jul 2012	Superstripes 2012, Ettore Majorana Foundation and Centre for Scientific Culture, Erice-Sicily, Italy *Dynamical Coulomb Blockade Observed in Nanosized Electrical Contacts*
Sep 2012	Workshop of the Chemical Physics Department of the FHI, Döllnsee, Germany *Dynamical Coulomb Blockade, Quantum Oscillations, and Superconductivity in Nanosized Electrical Contacts*
Oct 2012	2nd International School on Surface Science, Technologies and Measurements on the Atomic Scale, Khosta, Russia *Spectroscopic Manifestations of Low-Dimensional Physics: A Local View*
Oct 2012	International Workshop on Controlling the Electronic Structure of Semiconductor Nanoparticles by Doping and Hybrid Formation, Technische Universität Berlin, Berlin, Germany *Dynamical Coulomb Blockade Observed in Nanosized Electrical Contacts*
Jan 2013	Symposium on Surface and Nano Science 2013 (SSNS'13), Zao, Japan *Transport Properties and Electronic Structure of Individual Metallic Nanocontacts on Dielectric Supports: A Local View*
Jan 2013	Discussion Workshop to Access the Present Stand and Future Challenges in Surface Science, Schladming, Austria *Spectroscopic Manifestations of Low-Dimensional Physics: A Local View*
Apr 2013	45th ACS National Meeting, Symposium in Honor of Ulrike Diebold: Arthur W. Adamson Award for Distinguished Services in the Advancement of Surface Chemistry, New Orleans, LA, USA *Transport Properties and Electronic Structure of Individual Metallic Nanocontacts on Dielectric Supports: A Local View*
Jun 2013	Stripes 2013, Quantum in Complex Matter, Ischia, Italy *Transport Properties and Electronic Structure of Individual Metallic Nanocontacts on Dielectric Supports: A Local View*
Jul 2013	International Workshop on Nanomaterials and Nanodevices, Beijing, China *Transport Properties and Electronic Structure of Individual Metallic Nanocontacts on Dielectric Supports: A Local View*
Sep 2013	Workshop of the Chemical Physics Department of the FHI, Hasenwinkel, Germany *Electron Confinement: Past, Presence, and Future*

Shamil Shaikhutdinov

Oct 2011	1st Russian Congress on Catalysis, Moscow, Russia *Monolayer Oxide Catalysts: New Perspectives from Surface Science*
Mar 2012	Seminar, Martin Luther University Halle-Wittenberg, Halle, Germany *Thin Oxide Films: Structure and Reactivity*
Oct 2012	CECAM Workshop on Future Challenges in CO_2 Reduction, Bremen, Germany *Model Studies in Catalysis by Oxides*

Sep 2012	CRC-FHI Joint Meeting on Complex Surfaces in Material Science, Berlin, Germany *Reactivity of Oxide Ultrathin Films in CO Oxidation*
Jul 2013	Irsee VI Symposium, Kloster Irsee, Germany *Model Studies of Ceria-Based Oxidation Catalysts*

Martin Sterrer

Oct 2011	Annual Meeting, American Institute of Chemical Engineers, Minneapolis, MN, USA *Model Studies of Supported Catalyst Preparation: Pd Deposition on Iron Oxide Films from the Liquid Phase*
Nov 2011	54th Annual Meeting, American Vacuum Society, Nashville, TN, USA *Charge-Mediated Chemisorption on Supported Clusters*
Jan 2012	Seminar, University of Graz, Graz, Austria *Surface Science Approach to Supported Metal Catalyst Preparation - From Ultrahigh Vacuum to Metal Deposition from Solution*
Mar 2012	DPG-Frühjahrstagung, Berlin, Germany *Surface Science Approach to Supported Metal Catalyst Preparation - From Ultrahigh Vacuum to Metal Deposition from Solution*
Aug 2012	244th ACS National Meeting and Exposition, Philadelphia, PA, USA *Surface Science Approach to Supported Metal Catalyst Preparation - From Ultrahigh Vacuum to Metal Deposition from Solution*
Sep 2012	11th Pannonian Symposium on Catalysis, Obergurgl, Austria *Water on Oxide Surfaces*
Sep 2012	CRC-FHI Joint Meeting on Complex Surfaces in Material Science, Berlin, Germany *Model Studies of Catalyst Preparation*
Nov 2012	Seminar, Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany *Surface Science Models in Catalysis - Towards Realistic Catalyst Preparation*
Apr 2013	Department Seminar, Department of Physical Chemistry, FHI, Berlin, Germany *Water on Oxide Surfaces - Monolayer, Ice, Hydroxylation*
May 2013	Habilitationskolloquium, Technische Universität Berlin, Berlin, Germany *Optogenetics*
Jun 2013	Seminar, Institute of Physics, University of Graz, Graz, Austria *Surface Science Models of Heterogeneous Catalysts - From Ultrahigh Vacuum to Catalyst Preparation*

Department of Molecular Physics **Former Director: Gerard Meijer**

Acting Director: Matthias Scheffler

Staff scientists (as of November 2013):

Knut Asmis	(Habilitation)
Bretislav Friedrich	(Habilitation)
Gert von Helden	
Gabriele Santambrogio	
Nicolas Vanhaecke	

Guest scientists, staying for at least six months:

Mark Abel	(AvH Fellow)	Kevin Pagel	
Doo-Sik Ahn	(AvH Fellow)	Xiaowei Song	
Janneke Blokland	(AvH Fellow)	Xingan Wang	(AvH Fellow)
John Bowlan	(AvH Fellow)	DongdongZhang	(AvH Fellow)
Chris Eyles	(AvH Fellow)	Weiquing Zhang	(AvH Fellow)
Daniel Harding	(AvH Fellow)	Bum Suk Zhao	
Ling Jiang	(AvH Fellow)		

Guest scientists (temporary), paid from external funds:

Ad van der Avoird	(AvH Awardee)	University of Nijmegen, The Netherlands
Mike Bowers	(AvH Awardee)	University of California, Santa Barbara, CA, USA
Phil Bunker		National Research Council of Canada, Ottawa, Canada
Mike Duncan	(AvH Awardee)	University of Georgia, Athens, GA, USA
André Fielicke		Technische Universität Berlin
Kopin Liu	(AvH Awardee)	IAMS, Taipei, Taiwan
Boris Sartakov		Russia Academy of Science, Moscow, Russia

Graduate students:	10
Technicians:	7

Free Electron Laser Service Group:

Head: Wieland Schöllkopf

Staff members: 8

Recent Developments at the Department of Molecular Physics

Former director: Gerard Meijer (until August 31, 2012)
Acting director: Matthias Scheffler

General Remarks

In March 2012, the director of the *Molecular Physics Department*, Gerard Meijer, signed a contract with the Radboud University Nijmegen, the Netherlands, his *alma mater*, to assume the position of its "voorzitter college van bestuur" (president) and announced that he will depart from the FHI on August 31, 2012. His decision came as a shocking surprise not only to the members of his department but to the institute as a whole. In response, the Board of Directors of the FHI, now a four-member body, was able to secure a go-ahead from the MPG Outlook Committee (*Perspektivenkommission*) to begin a search for a fifth FHI director. The Outlook Committee made it clear that the search should not be biased toward the present research at the *Molecular Physics Department*. Instead, scientific excellence together with the ability to collectively lead the institute and to develop its future research directions were to be the main criteria for choosing a new director. For the interim period, Matthias Scheffler, the FHI executive director, has taken over as an acting director of the *Molecular Physics Department*.

Since its inauguration in 2003, the research of the "Meijer Department" has focused on two main research areas: (I) an area termed "molecular physics studies with infrared radiation" where IR spectroscopy and related methods have been used to explore the structure and dynamics of (bio)molecules, clusters, and cluster-adsorbate complexes in the gas phase, and (II) an area termed "cold molecules", where experimental techniques have been developed to achieve full control of both the internal and external degrees of freedom of neutral molecules and subsequently used to investigate the properties of the molecules.

At present, the decision about the future direction of the department has not been finalized. The CPT Section of the Max Planck Society has installed an *Appointment Committee*, but this had not even met before this report was written. The activities of the individual research groups in the field of "molecular physics studies with infrared radiation" have a significant overlap with the research in other departments of the FHI and at the Berlin-area universities. However, for "cold molecules" this is not the case and, therefore, the Board of Directors decided that this research direction will not be continued at the FHI. Utmost care has been taken to ensure that all those affected would obtain the support they need for the completion of their projects. In particular, the PhD students will be able to finish their theses and the postdocs will be given the opportunity to find adequate positions elsewhere. Individual discussions, conducted with the members of the department, were aimed at smoothing the transition and making the best of the situation for which they were not responsible. The two groups that are most affected are those of Nicolas Vanhaecke (Manipulation and control of molecular beams; group leader since September 1, 2011) and Gabriele Santambrogio (Micro-structured devices to manipulate molecules; group leader since September 1, 2010). Since the last meeting of the Fachbeirat, Karsten Horn has officially retired on December 31, 2011. Nonetheless, he remains actively involved in research with his research activities transferred to the *Physical Chemistry Department* of Martin Wolf.

Very recently, on November 15th, 2013, Uwe Becker passed away. Uwe Becker was a member of the *Molecular Physics Department* until his official retirement on June 30, 2012. However, being a successful and eager researcher, he remained active until the end and in 2012 alone, he (co)-authored seven publications. We will remember Uwe Becker as a colleague and researcher, who contributed significantly to the general understanding of the quantum mechanical process in ionization events.

Since September 2012, Gerard Meijer has visited the Department about every four weeks, and brought five of his PhD students to their successful graduation (two at Radboud University and three at the Freie Universität Berlin).

Despite these troubling developments, very exciting and highly successful research has been carried out at the Molecular Physics Department since Gerard Meijer’s departure. The ongoing projects are described below in two main sections.

Section I deals with “molecular physics studies with infrared radiation.” There have been four research groups active in this area. They all will make an extensive use of the FHI free electron laser, an FHI service group, which is described in a separate chapter of this report. The research focus of the *research group of Knut Asmis* is the structure and dynamics of small molecular ions and ionic clusters. During the past period, the employment status of Knut Asmis has changed. In April 2013 he has been granted a twelve-month leave of absence in order to take up the position of a visiting professor (W3-Lehrstuhlvertretung) at the Wilhelm-Ostwald-Institute of the University of Leipzig. In July 2013 he received a call for the W3-Chair in Physical Chemistry at the same institute and will assume this position as of February

1, 2014. Additionally, he will continue to supervise a research group at the FHI. Among the most important results are IR-spectra of protonated water clusters (see poster MP1). The *research group of André Fielicke* investigates strongly-bound metal clusters and their complexes with small chemisorbed molecules. As of January 1, 2013, André Fielicke moved to the Technical University Berlin. Part of his research activities, in particular those that use the FHI-FEL, remain at the FHI. A highlight is the observation of the activation of molecular oxygen on gold clusters (see poster MP3). The *research group led by Gert von Helden* investigates biological molecules in the gas phase. Since summer 2012, Gert von Helden also acts as the "administrative general manager" of the FHI for half of his time. Despite the significant workload in administration, a variety of fruitful research projects have been performed by this group. A particularly interesting result is the direct observation of the effects of microsolvation on the structure of biological molecules in the gas phase (see poster MP5). *Wieland Schöllkopf* is the *scientific leader of the FEL* service group. However, he is also heading research on atom and molecule optics and has been able to observe, for instance, quantum reflection of helium clusters from surfaces (see poster MP8).

In Section II, the activities of the three research groups active in the area of "cold molecules" are presented. The *research group of Bretislav Friedrich* has been studying interactions of molecules with and in fields, focusing on the manipulation of molecular rotation, quantum computing, and vector correlations in collisions. Poster MP9 features their work on the effects of far off-resonant light on polar paramagnetic molecules subject to combined electric and magnetic fields. In addition, Bretislav Friedrich has continued his exploits in the history of science, in particular of quantum mechanics. *Gabriele Santambrogio heads a group* that is financed by Gerard Meijer's *ERC advanced grant.* He investigates new methods for miniaturizing equipment to guide and decelerate polar molecules. Gabriele Santambrogio recently received an offer as a research group leader (Primo Ricercatore) from the Institute for Optics of the Italian Research Council (CNR) in Florence. He will assume this post in January 2014, but continue with some activities at the FHI until the end of January 2015. One important outcome of this research is the imaging of molecules on a chip (see poster MP10). *Nicolas Vanhaecke heads a group* at the FHI that exploits the Stark- or Zeeman-effect to control molecular beams in large machines. An important result of those efforts is the investigation of collisions of ortho- and para-H_2 with decelerated OH radicals (see poster MP12). Nicolas Vanhaecke recently accepted an offer from the European Patent Office, and he will assume the position of patent examiner in The Hague by February 1, 2014. He will continue some research activities at the FHI until June 2014.

Research

I) Molecular physics studies with infrared radiation

I A) *Gas Phase Vibrational Spectroscopy of Clusters (Knut Asmis)*

During the last two years, the research in our group was aimed at improving and refining the technique of mass-selective infrared photodissociation (IRPD) spectroscopy on gas phase cluster ions. As part of a joint project in the *Molecular Physics Department* aimed at the study of cluster- and biomolecular ions using the FHI-FEL (the CBI-Berlin project), a second ion-trap tandem mass-spectrometer was constructed. Both instruments make use of cryogenically cooled radio-frequency ion-traps, which we use as pulsed sources for mass-selected, thermalized and messenger-tagged cluster ions. The setups now allow measuring single- and double-resonance IRPD spectra of ions (<4000 amu) at variable temperature (6-300 K) and across a wide range of the IR spectral range (40-4000 cm^{-1}). As tunable and intense IR light sources we use the radiation from IR free electron lasers as well as table-top OPO/OPA laser systems. Both instruments are coupled to the beam line of the FHI FEL, allowing to use either this light source or a combination of the other available widely tunable and intense light sources.

The most notable break-through during the last period was the design, construction and application of a dual-acceleration 180° reflectron time-of-flight mass spectrometer, which replaced the linear TOF spectrometer in both instruments. This device allows double-resonance measurements with intermediate mass selection, a prerequisite for IR/IR ion-dip experiments. The advantage of the IR/IR approach over ion dip spectroscopy, albeit experimentally more demanding, is that these do not require the presence of a UV/VIS chromophore for isomer-specificity and are thus truly generally applicable. The experimental technique, named IR^2MS^2 spectroscopy, was introduced by Johnson and coworkers (Yale, USA) in 2008, but has been limited to the spectral range of commercially available table-top lasers (>600 cm^{-1}).

In our laboratory, the IR^2MS^2 technique has been successfully combined with radiation from the free electron laser FELIX (FOM Institute Rijnhuizen, NL) to measure isomer-specific IR^2MS^2 spectra of the Eigen-type and Zundel-type conformers of the protonated water hexamer across nearly the complete IR spectral range (260-3900 cm^{-1}). In collaboration with the Blum group (Theory Department) we then assigned the IR spectra on the basis of *ab initio* molecular dynamics simulations. We were able to gain insight into the mechanism responsible for the characteristically broad IR absorptions attributed to hydrogen-bonded O-H stretching modes. Furthermore, we observed the first hydrogen-bond stretching vibrations in protonated water clusters in the terahertz region (<400 cm^{-1}). This study was followed up by measurements on the larger protonated water clusters $H^+(H_2O)_n$ with n=7-10, which addressed the question of how the number of isomers evolves with the size of the hydration shell.

The characterization of how conjugate base anions, like nitrate and sulfate, are solvated at the molecular level, was extended by replacing water with inorganic acid molecules, like HNO_3 and H_2SO_4, as a microsolvent. Such clusters are among the most abundant anions in the atmosphere and have been predicted to play a crucial role in the formation mechanism of cloud condensation nuclei. As part of a long standing collaboration with the Neumark group (Berkeley, USA) we measured IRPD spectra of HSO_4 $(HSO_4)_n$, NO_3 $(HNO_3)_n$ as well as of mixed clusters of both acids at the FELIX facility. The key results of our studies are: *(i)* Hydrogen dinitrate contains an equally shared proton ($NO_3^- \cdots H^+ \cdots NO_3$), an arrangement that is surprisingly not disrupted by addition of a single water molecule. Only additional solvation with either more water or acid molecules induces the asymmetric $NO_3^- \cdots HNO_3$ motif. *(ii)* $HSO_4^-(H_2SO_4)_m(H_2O)_n$ clusters show a recurring triply hydrogen-bound configuration, which can be disrupted by the incorporation of water. *(iii)* Charge localization depends intimately on the size and composition of the clusters and cannot be reliably predicted from known gas phase acidities. We also studied the phenomena of "IRMPD transparent" bands by comparing the IRMPD spectra of the bare and messenger-tagged cluster anions.

Work on microhydrated nitrate and biphosphate anions as well as linear dicarboxylate dianions of varying chain length was brought to completion. The assignment of these spectra was more complex than anticipated, because it either required considering anharmonic effects (nitrate and biphosphate clusters) or global optimization schemes (dicarboxylate dianions). This was achieved through collaborations with the groups of K. Jordan (Pittsburgh, USA), Z.-F. Liu (Hong Kong, China) and A. Rubio (San Sebastian, Spain), respectively.

The focus of our work on metal oxide clusters, performed in collaboration with the Sauer group (HU Berlin), was moved away from vanadium oxides and towards microhydrated aluminum-, iron- and silicon-oxide clusters. First experiments and calculations have been performed on small, He-tagged iron oxide cations. In addition, a sputter source has been setup for the efficient production of these clusters. These studies are part of a planned DFG Collaborative Research Center (CRC) in Berlin, which passed its evaluation in summer 2013 with recommendation. The final decision is expected in Nov. 2013.

Other collaborative projects included the measurement of IR spectra of the periodated dodeca-closoborates $Br_{12}B_{12}^{2-}$, $I_{12}B_{12}^{2-}$ as well as of the CID-fragments $I_nB_{12}^-$ with n=7-11 (Warnecke, Bremen) and of hydrogen- and nitrogen-containing carbon cluster anions (L. Wöste, Berlin). As part of the first project, we provided spectroscopic evidence for the predicted cage opening (3D to 2D transition of the B_{12} arrangement) upon deiodination of $I_nB_{12}^-$ in-between n=8 and n=7. Carbon-containing cluster anions have attracted much attention, in particular since their detection in outer space. However, the IR spectra of many of these species have not been measured up-to-date. We are trying to fill the gap by providing data for C_nH^- (n=3,5-9,11), C_nN^- (n=5-11) and $C_nN_3^-$ (n=3-5). The spectra are currently being evaluated.

I B) *Spectroscopy and chemistry of metal clusters and cluster complexes (André Fielicke)*

The activities of the group focus on the investigation of the structures of clusters and complexes with small molecules in the gas phase using vibrational spectroscopy. These studies aim to obtain fundamental insights into the effects of geometric and electronic structure on reactivity, resulting in a detailed local picture of reactions at well-defined metal sites. Our main experimental tool has been the Free Electron Laser for Infrared eXperiments (FELIX) as a source for intense and tunable IR radiation covering frequencies from 40 to 2400 cm^{-1}. This spectral range gives access to the internal vibrational modes of metal clusters in the far-IR as well as the characteristic vibrational modes of cluster-bound ligands in the mid-IR.

In the last two years we have continued the investigations on the structures and chemical properties of clusters of different platinum group metals. Insights into the structures of the bare metal clusters have been obtained via far-IR spectroscopy; subsequently we have characterized intermediates relevant in reactions of these species, such as the initial step in the activation of methane on Pt being a molecular adsorbate strongly bound via agostic interaction of two C-H σ-bonds with a Pt atom. Full dehydrogenation of CH_4 leads to platinum carbides with a very strongly bound C atom surrounded by 3 Pt atoms. Compared to the bare Pt clusters these carbides, as well as the additionally studied oxides, hydrides, and carbonyls show very different structures, e.g. the oxidized clusters favoring planar geometries. Further spectroscopic studies have been performed for N_2 bound to ruthenium clusters. For ammonia synthesis the N_2 activation is the rate-limiting step and surface studies have shown that N_2 dissociation on Ru is a highly structure-sensitive process. The observed N–N stretch frequencies in the range of 2110 to 2200 cm^{-1} suggest the presence of σ-bonded chemisorbed N_2 oriented perpendicularly to the cluster surface. Interestingly, for the complexes of Ru_8 and Ru_9 larger deviations from the trends in ν(N–N) are observed similar to the CO adsorbates. This may be due to special geometric or electronic structures and indeed for these sizes unusual cubic structures had been predicted.

A second topic has been the activation of molecular oxygen by gold, which is one of the key steps in the low temperature oxidation catalysis by nano-scale gold particles. We have unambiguously shown the formation of superoxo species when binding O_2 to anionic gold clusters, surprisingly the same reaction is also observed for neutral gold clusters. In contrast to what has been known so far, even bare cationic gold clusters are found to bind and activate molecular O_2; depending on the cluster size and the number of O_2 ligands different species, i.e. superoxide, physisorbed O_2, or dioxygenyl are identified. These studies on the activation of small molecules by metal clusters are funded by the DFG (DFG grant FI 893/3) and the Cluster of Excellence “Unified Concepts in Catalysis” (Unicat).

Furthermore we have investigated the structures of clusters of the lanthanide element terbium, which was motivated by the strongly size-dependent magnetism of these clusters. Using DFT with a 4f-in-core effective core potential (ECP) we can reproduce the experimental far-IR spectra well and identify structures with open 5d/6s configurations. This may become relevant in explaining the coupling between the 4f electrons that determines the overall magnetism of these clusters.

Within our activities as part of the DFG funded research group FOR 1282 “Controlling the Electronic Structure of Semiconductor Nanoparticles by Doping and Hybrid Formation” (DFG grant FI 893/4) we study the effects of doping on the structural and electronic properties of small silicon clusters. Recent results from our collaboration with the group of Otto Dopfer (TU Berlin) are i) the structural transition in $Si_{6-n}C_n$ clusters from compact pure Si clusters to chain like C clusters by subsequent replacement of Si by C atoms and ii) the analysis of the effect of Xe binding on the structure and IR spectrum of Si_4^+. Together with the group of Ewald Janssens and Peter Lievens (KU Leuven, Belgium) we continued studies of (transition) metal doped Si clusters and have investigated the structure and magnetic properties of neutral and cationic Mn doped Si clusters. The smaller Mn doped Si clusters are isostructural to the bare Si clusters, but due to the open 3d shell of the Mn, carry a significant magnetic moment. For the larger clusters we apply *ab initio* global optimization via basin hopping to locate low energy isomers for comparison with the experimental IR spectra. In this way we identified that for Ag and Au doped Si_n clusters the exohedral structures continue until unexpectedly large sizes (at least up to n=15).

Another of the key topics of the group has been the development and application of new experimental techniques for the IR spectroscopy of strongly bound gas-phase clusters in different charge states. For instance, we have developed an IR-UV two-color excitation scheme, where resonant IR excitation of neutral clusters is probed by an enhancement of the yield for (non-resonant) near-threshold single-photon UV ionization. This technique has now been applied to several cluster systems like pure and doped silicon clusters, boron clusters, and magnesium oxide clusters. While these experiments have been successful as the ionization energies of the investigated species were close to the photon energy of the F_2 laser (7.9 eV) used for the near threshold ionization, the planned use of tunable UV sources will significantly broaden the range of possible applications. Furthermore, we have explored the possibilities of multiple photon excitation of clusters using the Free Electron Laser for IntraCavity Experiments FELICE, a recent extension of FELIX providing considerably higher IR laser fluences. Together with the FELIX in-house group of Joost Bakker we have performed proof-of-principle experiments on the far-IR multiple photon ionization of neutral metal clusters and obtained the very first far-IR spectra of anionic metal clusters using far-IR multiple photon electron detachment.

Due to the relocation of FELIX from the FOM Institute for Plasmaphysics in Nieuwegein to the Radboud University Nijmegen in 2012 we had to stop all experimental activities at this facility at the end of 2011. The cluster beam experiment formerly permanently installed at FELIX has been moved to the FHI in the beginning of 2012. It was refitted and upgraded, relocated to a new dedicated laboratory and connected to the beamline of the FHI-FEL. The laboratory includes a tunable UV laser to allow for the IR spectroscopy of neutral species via the IR-UV two-color ionization method. At the end of 2012 the group moved to the Technical University Berlin and continues there this line of research in collaboration with the FHI. We aim for a continuation of our experimental activities on gas-phase clusters using IR free electron lasers at the FHI-FEL.

First experiments focusing on CO binding to small metal clusters have already been performed. We currently investigate the activation of CO by nano-alloys of early and late 3d transition metal clusters. The CO stretch frequency is used to quantitatively probe the effects of alloying on the d-band center that is controlling the strength of the π-backbonding. In a second experiment with the new FHI-FEL we have studied saturated cationic ruthenium cluster carbonyls.

I C) *Biological molecules in the gas phase (Gert von Helden)*

The focus of our research is to investigate the structures of biological molecules and learn how those depend on internal composition as well as on the environment of the molecules. To this end, novel methods are developed that couple mass spectrometry to other techniques such as ion mobility spectrometry (IMS), helium droplet isolation, and optical spectroscopy. The interpretation of the results is often guided by modeling and we have ongoing collaborations with the FHI Theory Department.

IMS coupled to mass spectrometry (IMS-MS) is a powerful method that gives direct information on the size and shape of biological molecules in the gas phase. For a long time, experiments could only be performed with prototype machines developed in research laboratories. A few years ago, however, a vendor of mass-spectrometric equipment released the first commercial IMS-MS instrument. We have a second generation of this machine, which is used in a project largely led by Kevin Pagel for the analysis of carbohydrates and carbohydrate mixtures. Compared to other classes of biological macromolecules like DNA or proteins, the structure and analysis of complex carbohydrates is uncharted territory. Due to their structural diversity, it is often not possible to distinguish isomers using established MS techniques and a promising route is to implement IMS as an additional separation step.

A second IMS-MS machine is home built and has features that are not available at the commercial instrument. Several of its design parameters are based on an apparatus constructed in the M.T. Bowers laboratory at the University of California at Santa Barbara, with whom we have an ongoing collaboration. A particularly important parameter is the use of an ultra-soft nano-electrospray ionization source, followed by a gentle front-end of the machine. This allows us to transfer weakly bound peptide and protein complexes into the instrument while preserving structural elements that are present in the condensed phase. After ion mobility separation and mass analysis, the ions can interact with an excitation laser. Another mass spectrometer is then used to detect laser-induced fragmentation. The instrument is connected to a port of the FHI-FEL beamline and we are planning to measure IR spectra of mass to charge as well as conformer (IMS) selected ions in the near future. In addition, other laser sources are used and progress will be reported upon.

In a particular exciting experiment, IMS-MS was used to investigate the impact of side-chain backbone interactions on the structure of gas-phase protein ions by non-covalent attachment of crown ethers (CE). Our results indicate that in the absence of solvent, secondary interactions between charged lysine side chains and backbone carbonyls can significantly influence the structure of a protein. Once the charged residues are capped with CEs, certain charge states of the protein are found to undergo a significant structural compaction. It appears that interactions between protonated lysine side chains and backbone carbonyls are in direct competition with the intramolecular H-bonds that determine the protein's secondary and tertiary organization. Non-covalent attachment of CE to the charged side chains of the protein can compensate for these effects by solvating the positively charged ionic groups in a similar way to solvent molecules in the condensed phase. In a more general context, this implies that CE-side-chain microsolvation can be used as a tool to manipulate and tune protein structures in the gas phase. In addition, these experiments reveal that the structure of a protein in the gas phase is not only determined by the charge state, but also by much more subtle side chain to backbone interactions.

Another interesting class of molecules that have recently been studied in the group are polymers that contain non-natural amino acids – so-called "foldamers". The advantage here is that peptide bonds involving non-natural building blocks are less prone to proteolytic cleavage and, as such, of enormous interest for drug development. The first step toward successful foldamer design is typically the identification of polymeric backbones, which fold into a well-defined and ideally native-like structures. A common strategy to achieve that is to assemble polypeptides consisting of β-amino acids in which the peptide backbone is extended by one methylene unit. In a collaborative effort with the FHI Theory Department, we use ion mobility mass spectrometry, gas phase spectroscopy, and *ab-initio* molecular dynamics simulations and obtained compelling evidence that peptides which consist exclusively of β-amino acids can adopt a helical structure that resembles the characteristic H-bonding pattern of the α-helix.

When doing spectroscopy on larger biological molecules in the gas phase, efficient cooling of the molecules is necessary in order to obtain spectra that can be interpreted. We have pioneered a method in which mass/charge selected biomolecules are first trapped in a linear ion trap and then captured by liquid helium droplets. The liquid helium environment then provides for efficient cooling down to 0.4 K (the equilibrium temperature of the droplets) while at the same time being only a weak perturbation for the trapped ions. In the past two years, we continued to characterize and improve the experiment and are now able to incorporate small peptides as well as large proteins in helium droplets with beam intensities that are high enough for spectroscopic experiments. Those experiments were first performed in the UV. However, the machine is also connected to the FHI-FEL beam distribution system and we expect first experiments with that light source to be performed very soon.

I D) *Atom optics and quantum reflection of molecular beams (Wieland Schöllkopf)*

Based on our previous work on "*quantum reflection*" of atomic and molecular beams from micro-structured surfaces we have extended our investigations of coherent (and non-destructive) scattering of dimers and trimers of ground-state helium, He_2 and He_3, from reflection gratings. In addition, we have embarked on a project to develop nano-structured surfaces tailor-made to substantially increase the quantum-reflection probabilities. Furthermore, we have continued our collaboration with the group of R. Dörner at the University of Frankfurt, where one of our diffraction grating setups is being used to selectively investigate helium dimers and trimers by the COLTRIMS method. Theses studies have the potential to map out the vibrational wave-functions of He_2 and He_3 to a level of precision where the experimental data can serve as a benchmark for quantum chemistry calculations of the helium pair potential.

In the experiments we observe "*quantum reflection*" of atoms as well as helium clusters, if the particles approach the surface at grazing incidence of ~1 mrad or even less. In a classical picture, the long-range atom-surface van der Waals interaction accelerates a particle towards the surface, thereby increasing its kinetic energy by the potential well-depth energy. The particle continues to approach the surface until it is scattered back at the classical turning point on the repulsive branch of the potential, which effectively forms a hard-wall. For helium dimers and trimers, this hard-wall impact will inevitably lead to break-up of the fragile bonds, because the binding energies of 100 neV and 10 μeV, respectively, are orders of magnitude less than the ~10 meV surface-potential well depth. Thus, our observation of non-destructive scattering of He_2 and He_3 from a reflection grating cannot be understood within a classical description.

It is readily explained, however, by quantum reflection at the attractive van der Waals potential branch, which has a slope that appears to be steep on a length scale set by the incident particle's de Broglie wavelength. If the particle's velocity component perpendicular to the surface is sufficiently small, the corresponding de Broglie wavelength gets sufficiently large and the steep slope, effectively, appears as a potential step. According to quantum mechanics, at a potential step, an incident wave-function shows a non-vanishing reflection probability that even approaches unity in the limit of zero velocity. In our experiment a sufficiently small velocity component perpendicular to the surface is achieved by minute grazing-incidence angles. Quantum reflection of He_2 and He_3 occurs tens of nanometers above the surface, where the surface induced forces are too weak to break up the fragile bonds.

We observe quantum reflection of He_2 and He_3 using a commercial plane-ruled blazed reflection grating mounted in the conical diffraction configuration, where the grating grooves are nearly (within a small azimuth angle) parallel to the scattering plane. Varying the azimuth angle and the incidence angle allows for an adjustment of both, the effective grating period and the effective blaze angle, and thereby allows for enhanced resolution and intensity of He_2 and He_3 diffraction peaks. This has made possible the observation of emerging beam resonances, which we had previously seen for helium atoms, for He_2 as well. Given the unusual fragility and size of He_2 this is a surprising observation, because emerging beam resonances imply multiple surface scattering as well as the existence of an evanescent wave of helium dimers.

To increase the quantum-reflection probabilities, we have started a collaboration with groups from UNIST in Ulsan, South Korea, to investigate the use of nano-structured (nano-ridged) surfaces. Bum Suk Zhao at UNIST, who used to be a member of our group at the FHI from 2005 to 2011, designs different surface layouts, which the group of Lee is going to fabricate using state-of-the-art micro-structuring technology available at UNIST. Substantially increased reflection probabilities can, possibly, allow the observation of inelastic diffraction of helium trimers. In addition, 2-dimensional focusing of helium atoms by reflection from concave surfaces can be tried. Inelastic diffraction, predicted in a theory paper by Köhler and Hegerfeldt, describes an incident He trimer which, upon scattering off the grating, is excited from the vibrational ground-state to the first excited state. The excitation energy is not exchanged between the grating (phonons) and the trimer, but it is transferred from the trimers's center of mass motion (kinetic energy) to the vibrational degree of freedom. Thus, the kinetic energy is reduced by just the excitation energy (~10 μeV), thereby increasing the trimer's de Broglie wavelength. The wavelength increase, in turn, results in slightly different diffraction angles, which are well detectable given the high angular resolution of our apparatus. Once we observe an inelastic diffraction peak of He_3, it will be straightforward to extract the vibrational excitation energy from an analysis of diffraction angles. Observation of the helium trimer's elusive excited state and determination of its energy is especially interesting, because this state has been predicted by numerous theory works to be the only naturally occurring *Efimov state*.

II) Cold molecules

II A*) Interactions of molecules with fields (Bretislav Friedrich)*

Over the reporting period 2011-2013, the group pursued research on (i) vector correlations and diffraction-dominated stereodynamics; (ii) interactions of polar paramagnetic molecules with combined electric, magnetic and optical fields; and (iii) quantum computing with polar molecules.

(i) The correlation between the relative momenta of the reactants and products, also known as the differential cross section, had enabled the first insights into the dynamics of molecular collisions, leading to the distinction between, e.g., the "rebound" and the "stripping" reaction mechanisms. The ability to control the polarization of the reactants and to detect the polarization of the products for atom—diatom collisions has introduced two additional vectors into the game and thereby 5 more two-vector correlations, 4 three-vector correlations and one overarching four-vector correlation, all to characterize the collision's stereodynamics. Like the differential cross section, each of these vector correlations encapsulates a different aspect of the collision dynamics and provides a glimpse of the molecular forces involved. Unlike the differential cross sections, which have been the subject of intense analytic modelling resulting in textbook-bound form factors for different types of collisions (e.g., "direct" or "via an intermediate complex"), most of the other two-vector correlations and none of the higher ones have been modelled. It was our goal to develop a model of higher vector correlations for (at least) a class of molecular collisions and thus to help understand such collisions. In our work, we chose thermal rotationally inelastic collisions dominated by repulsion, which can be analytically modelled by invoking the sudden approximation and the Fraunhofer scattering, known from optics. The resulting Fraunhofer collision model is inherently quantum, has no fitting parameters, and furnishes state-to-state scattering amplitudes in closed form. The model of vector correlations based on the Fraunhofer model is therefore likewise fully quantum and fully analytic, even for collisions taking place in fields (electric, magnetic, radiative). Our analytic model – checked against experimental data and accurate computations

– turned out to be quantitative for key three-vector correlations involving product polarization, and made it possible to derive form factors which are characteristic for scattering dominated by diffraction. These can be readily used to identify diffraction-dominated scattering at a glance. As the only model available of all the correlations among four vectors, it also represents a touchstone for any models that are yet to be developed for other types of scattering processes.

(ii) Interactions with external electric, magnetic or optical fields provide the chief means to manipulate the rotational and translational motion of neutral gas-phase molecules. The pursuit of such means is a leading frontier of chemical/molecular physics. Among recent developments are new methods to control the orientation and/or alignment of molecules as well as methods to deflect and focus their translational motion and to achieve molecular trapping. The importance of orientation comes also to light in novel applications such as time-resolved photoelectron dynamics, separation of photodissociation products, deracemization, high-order harmonic generation and orbital imaging, quantum simulation, or quantum computing. All methods to manipulate molecular rotation and translation via external fields rely on the ability to create directional states of molecules. This is because only in directional states are the molecular body-fixed multipole moments 'available' in the laboratory frame where they can be acted upon by space-fixed fields. A high degree of orientation – whereby the molecular electric dipole moment is made available in the laboratory – can now be achieved by a versatile technique that combines a static electric field with a nonresonant optical field. The dual-field combination gives rise to an amplification effect which occurs for any polar molecule, as only an anisotropic polarizability, along with a permanent dipole moment, is required.

If the polar molecule is also paramagnetic, similar amplification effects can be obtained in combined static electric and magnetic fields. The latter effects as well as their further enhancement induced by superimposing a non-resonant optical field are the subject of our ongoing investigations. The triple field-combination not only offers a high efficiency and flexibility in amplifying molecular orientation but is also of fundamental theoretical interest per se, as monodromy and quantum chaos lurk behind the combined-field effects. Among the most prominent examples of polar paramagnetic molecules are the ubiquitous $^2\Sigma$, $^3\Sigma$, and $^2\Pi$ linear species, such as SrF, SO, and OH. Heteronuclear diatomics or larger polar molecules that contain a rare-earth atom often exhibit much higher orbital and spin electronic angular momenta (e.g., CeO is a $^3\Phi_2$ molecule in its electronic ground state) and, therefore, correspondingly larger magnetic dipole moments. The recently discovered LiHe van der Waals molecule, a polar and paramagnetic halo species, would also benefit from the study of its properties in combined fields, as this would likely reveal additional particulars about its structure and the dynamics of its formation. As we have been able to show recently, at the core of the triple-field effects is the lifting of the degeneracy of the projection quantum number M by a superimposed magnetic field and a subsequent coupling of the members of the "doubled tunneling doublets" (for states with $M \neq 0$) by a weak electrostatic field (for a single molecule) or by the electric dipole-dipole interaction (for two or more polar paramagnetic molecules).

(iii) In proposals for quantum computers using arrays of trapped ultracold polar molecules as qubits, a strong external field with appreciable gradient is imposed in order to prevent quenching of the dipole moments by rotation and to distinguish among the qubit sites. That field induces the molecular dipoles to undergo pendular oscillations, which markedly affect the qubit states and the dipole-dipole interaction. We evaluated entanglement of the pendular qubit states for two linear dipoles, characterized by pairwise concurrence, as a function of the molecular dipole moment and rotational constant, strengths of the external field and the dipole–dipole coupling, and ambient temperature. Simple approximate formulas fitted to our numerical results were derived from which the key quantities needed for gate operations on the molecular qubits can be obtained in terms of unitless reduced variables. We also presented a systematic approach to implementation of basic quantum logic gates operating on polar molecules in pendular states as qubits. Multi-target optimal control theory was used as a means of optimizing the initial-to-target transition probability via a laser field. We gave detailed calculations for the SrO molecule, a favorite candidate for proposed quantum computers. Our simulation results indicate that NOT, Hadamard and CNOT gates can be realized with high fidelity, as high as 0.985, for such pendular qubit states.

II B) *Micro-structured devices to manipulate molecules (Gabriele Santambrogio)*

Many of the advantages of magnetic atom chips could also be realized with electric molecule chips: the miniaturization of field structures enables the creation of large field gradients, i.e. large forces and tight potential wells for polar molecules. Above a chip, the positions of these potential wells, and thereby the positions of the trapped molecules, can be controlled to an extreme precision. Moreover, the trapped molecules are normally in a single quantum state and have a kinetic energy of the order of few mK. A particular advantage of using molecules instead of atoms on a chip is that they can be coupled to photons over a wider range of frequencies, due to their rotational and vibrational degrees of freedom.

However, a major obstacle that has delayed the development of the molecule chip arises from this same richness of molecules' internal degrees of freedom. The complicated level structure of molecules results in a general lack of closed two-level systems that are necessary for efficient laser cooling and detection using absorption or laser-induced fluorescence. In recent years, we have demonstrated that one can exploit the cooling provided by a supersonic expansion to load a microchip with cold molecules directly from a molecular beam. We used the molecule chip to trap, decelerate and accelerate polar molecules (Fachbeirat 2009). We studied non-adiabatic transitions undergone by trapped molecules and we showed that the internal degrees of freedom of molecules on the chip can be manipulated by coupling microwave radiation to the device (Fachbeirat 2011). In this last period we improved on (a) the manipulation of trapped molecules, extending the spectral range to the infrared and addressing vibrational transitions, and, most importantly, (b) we developed on-chip detection, adding the final fundamental component to the molecule chip.

(a) To demonstrate the vibrational pumping of molecules on the chip we use carbon monoxide molecules prepared using a pulsed laser in a single rotational level (J = 1) of the first electronically excited, metastable state ($a^3\Pi_1$, v = 0). The v = 1 ← v = 0 vibrational transitions are induced using pulsed IR radiation around 5.9 μm. Molecules pumped into selected rotational levels of the vibrationally excited v = 1 state are subsequently state-selectively detected through ionization using another pulsed laser system. Besides demonstrating that pulsed IR radiation can be coupled to molecules that are trapped less than 50 μm above the chip without damaging the chip, these experiments show that vibrational transitions can be pumped without having to switch off the traps (as is the case for rotational transitions). Moreover, we show that we can accurately model the transitions in the inhomogeneous and rotating field of our microtraps and that we can address selectively a subset of molecules from our traps by choosing the appropriate polarization of the laser beam.

(b) For on-chip detection we use resonant-enhanced multi-photon ionization, which is quantum state selective, can be saturated with a few mJ/mm^2 of laser light for most molecules, is intrinsically background-free, and is of general applicability. While in the simplest implementation of our detection scheme one would simply count the ions, we take the further step of using ion optics to create a time-resolved spatial image of the molecules.

We resolve the spatial structure of an array of microtraps on our chip and use this resolution to analyze the phase space distribution of the molecules. We subsequently show a sequence of time-resolved snapshots from a ballistic expansion, in a similar fashion as for the time-of-flight imaging of atomic ensembles on atom chips. Moreover, we use this new detection method to investigate the effect of phase space manipulation sequences applied to the trapped molecules. Among other things, we observe experimentally that the time scale on which a velocity cooling manipulation can take place is comparable with the inverse of the trap frequency.

Furthermore, the development of the chip-based ring decelerator that we announced two years ago is now coming to a conclusion. We met unexpected difficulties in the realization of both the microstructures and the amplifiers for generating the electrical potentials applied to the chip. The manufacturing of the microstructures proved more difficult than was first anticipated and the company that had promised to deliver the amplifier failed to do so; we thus hired an engineer who subsequently developed a whole new device with the required specifications.

II C) *Manipulation and control of molecular beams (Nicolas Vanhaecke)*

Over the last few years our group has focussed both on the applications of Stark decelerated molecular beams and on refining the operation of the Stark decelerator. The Stark deceleration technique has now become a mature technique which offers tunability of the beam velocity, a very narrow velocity spread, and an extremely high quantum state purity. These beams are therefore ideally suited for collision studies, as was already recognized in 2006.

The existing 2.6-meter-long molecular beam machine had been used before the Fachbeirat 2011 to investigate collisions between Stark decelerated OH radicals and various rare gases (He, Ne, Ar, Kr, Xe), as well as with D_2. Moreover, the machine has been combined with a hexapole state-selected beam of NO radicals, intersecting the OH beam at right angle. Rotationally and spin-orbit inelastic scattering cross sections were measured on an absolute scale for collision energies between 70 and 300 cm^{-1}. These cross sections show fair agreement with quantum coupled-channels calculations using a set of coupled model potential energy surfaces based on *ab initio* calculations for the long-range nonadiabatic interactions and a simplistic short-range interaction. This comparison reveals the crucial role of long-range forces in complex molecular collision processes.

Then we have focussed on a second bimolecular system, OH-H_2, with several goals. First, as the OH-NO experiment showed, it is extremely difficult to obtain experimental absolute cross sections. Therefore we decided to compare only ratios of cross sections. We did it first in a rather classical way, namely measuring relative cross sections, i.e. comparing inelastic collisions leading to different final quantum states of the OH radical. We also used molecular hydrogen in its ortho and para forms, in order to compare their collision cross sections, given a final quantum state of the OH radical. A wealth of experimental results of inelastic scattering of ortho- or para-H_2 with Stark-decelerated OH ($X^2\Pi_{3/2}$, J=3/2,f) at collision energies between 40 and 150 cm^{-1} has been obtained. Clear differences in the scattering process of ortho- and para-H_2 are seen in the relative cross sections, originating from the different multipole interactions involved. Absolute cross sections of ortho-H_2 or para-H_2 colliding with OH have been calculated and compared with our experimental data, in the frame of an international collaboration involving theory groups in the USA and in the Netherlands.

In addition, by using a special collision scheme with collimated beams, we have achieved a very high collision energy resolution (below 1cm^{-1}) in our crossed beam experiment, allowing us to observe sharp quantum thresholds and resonance effects. Theoretical cross sections, calculated as a function of the collision energy, have shown very good agreement with our experimental data.

In the spring 2012, we decided to stop the merged beam experiment, for which a bent electrostatic hexapole had been designed and built downstream from a 1.2-meter-long Stark decelerator. This decision was based partially on the restructuring of the Molecular Physics Department, and partially on the extreme difficulty of such an experiment. Instead, we devoted the existing Stark decelerator to the investigation of a large, new class of switching schemes in a Stark decelerator. These schemes offer more free parameters to be optimized than in the standard mode of operation of the Stark decelerator. Therefore, there is plenty of room for optimization of the switching scheme, depending of the required six-dimensional properties of the decelerated beam (i.e. with goals in the six-dimensional phase space). We have shown that after optimization, these schemes substantially improve the efficiency of the decelerator at both low and high velocities, which are relevant for trapping experiments and collision experiments, respectively. Both experimental and simulation results show that these new modes of operation outperform the schemes which have hitherto been in use. In addition, these new modes of operation could potentially be extended to other deceleration techniques, like the Zeeman deceleration.

An important challenge in the field of cold molecules is a general technique to manipulate the motion of polar molecules in high-field-seeking states. The alternating gradient method, although very general, has proven to be extremely demanding. An alternative uses the interaction of polar molecules with near resonant radiation. The microwave decelerator developed in our department has been used to decelerate and focus ammonia molecules in high-field-seeking states. The multistage decelerator consists of a cylindrical microwave cavity oscillating in the TE_{11n} mode, with n=12 electric field maxima along the beam axis. By switching the microwave field on and off at the appropriate times, a beam of state-selected ammonia molecules with an incident mean velocity of 25 m/s is guided while being spatially focused in the transverse direction and bunched in the forward direction. Deceleration from 20.0 to 16.9 m/s and acceleration from 20.0 to 22.7 m/s have been demonstrated. In August 2012, this experiment moved to the group led by M. Schnell in Hamburg, with the goal to extend the decelerator to several microwave cavities in order to improve the range of manipulated velocities.

Early 2012, we started a new experiment, with the goal to extend our outstanding control over decelerated molecular beams to species that cannot be manipulated with electric field gradients. Using time-dependent, inhomogeneous magnetic fields, one can get control over the motion of paramagnetic molecules in a supersonic beam and can manipulate the highly chemically relevant, broad class of free-radicals. In our laboratory, a new-concept Zeeman decelerator has been designed and is being constructed, which produces a traveling magnetic trap with controllable longitudinal velocity and transverse orientation. Atoms and molecules possessing a magnetic dipole moment, in so-called low-field-seeking states, are trapped around a node of a propagating wave, provided that the initial velocity of the wave matches a velocity populated in the supersonic beam. In addition, three-dimensional confinement is achieved by controlling the radial orientation of the trap, which can be done fully independently from its longitudinal motion.

Due to the restructuring of the *Molecular Physics Department*, the ultracold atom experiment has been shut down in the spring 2012, and the molecular synchrotron was moved to the group led by H.L. Bethlem in Amsterdam.

Publications of the Molecular Physics

2011 (late publications)

Blokland, J., J. Riedel, S. Putzke, B.G. Sartakov, G.C. Groenenboom and G. Meijer: Producing translationally cold, ground-state CO molecules. The Journal of Chemical Physics **135** (11), 114201 (2011).

Bostwick, A., J.L. McChesney, T. Ohta, K.V. Emtsev, T. Seyller, K. Horn and E. Rotenberg: The interaction of Xe and Xe + K with graphene. Journal of Electron Spectroscopy and Related Phenomena **183** (1-3), 118-124 (2011).

Burow, A.M., T. Wende, M. Sierka, R. Włodarczyk, J. Sauer, P. Claes, L. Jiang, G. Meijer, P. Lievens and K.R. Asmis: Structures and vibrational spectroscopy of partially reduced gas-phase cerium oxide clusters. Physical Chemistry Chemical Physics **13** (43), 19393-19400 (2011).

Chaudhry, A. and H. Kleinpoppen: Analysis of Excitation and Ionization of Atoms and Molecules by Electron Impact. Springer, New York (2011). ISBN 978-1-4419-6946-0.

Claes, P., E. Janssens, V.T. Ngan, P. Gruene, J.T. Lyon, D.J. Harding, A. Fielicke, M.T. Nguyen and P. Lievens: Structural Identification of Caged Vanadium Doped Silicon Clusters. Physical Review Letters **107** (17), 173401 (2011).

Crassee, I., J. Levallois, D. van der Marel, A.L. Walter, T. Seyller and A.B. Kuzmenko: Multicomponent magneto-optical conductivity of multilayer graphene on SiC. Physical Review B **84** (3), 035103 (2011).

Crassee, I., J. Levallois, A.L. Walter, M. Ostler, A. Bostwick, E. Rotenberg, T. Seyller, D. van der Marel and A.B. Kuzmenko: Giant Faraday rotation in single- and multilayer graphene. Nature Physics **7** (1), 48-51 (2011).

Dhaka, R.S., A.K. Shukla, K. Horn and S.R. Barman: Photoemission study of Al adlayers on Mn. Physical Review B **84** (24), 245404 (2011).

Freund, H.-J., G. Meijer, M. Scheffler, R. Schlögl and M. Wolf: CO oxidation as a prototypical reaction for heterogeneous processes. Angewandte Chemie International Edition **50** (43), 10064-10094 (2011).

Freund, H.-J., G. Meijer, M. Scheffler, R. Schlögl and M. Wolf: Die CO-Oxidation als Modellreaktion für heterogene Prozesse. Angewandte Chemie **123** (43), 10242-10275 (2011).

Friedrich, B., D. Hoffmann and J. James: One hundred years of the Fritz Haber Institute. Angewandte Chemie **123** (43), 10198-10225 (2011).

Friedrich, B., D. Hoffmann and J. James: One hundred years of the Fritz Haber Institute. Angewandte Chemie International Edition **50** (43), 10022-10049 (2011).

Gerling, U.I.M., E. Brandenburg, H. von Berlepsch, K. Pagel and B. Koksch: Structure Analysis of an Amyloid-Forming Model Peptide by a Systematic Glycine and Proline Scan. Biomacromolecules **12** (8), 2988-2996 (2011).

Gonzalez Florez, A.I., S.A. Meek, H. Haak, H. Conrad, G. Santambrogio and G. Meijer: An electrostatic elliptical mirror for neutral polar molecules. Physical Chemistry Chemical Physics **13** (42), 18830-18834 (2011).

Gubbels, K. and H.T.C. Stoof: Interacting preformed Cooper pairs in resonant Fermi gases. Physical Review A **84** (1), 013610 (2011).

Hermes, A.C., S.M. Hamilton, W.S. Hopkins, D.J. Harding, C. Kerpal, G. Meijer, A. Fielicke and S.R. Mackenzie: Effects of Coadsorbed Oxygen on the Infrared Driven Decomposition of N_2O on isolated Rh_5^+ Clusters. The Journal of Physical Chemistry Letters **2** (24), 3053-3057 (2011).

James, J., T. Steinhauser, D. Hoffmann and B. Friedrich: One hundred years at the intersection of chemistry and physics: The Fritz Haber Institute of the Max Planck Society 1911-2011. De Gruyter, Berlin (2011). ISBN 978-3-11-023953-9.

Jiang, L., T. Wende, P. Claes, S. Bhattacharyya, M. Sierka, G. Meijer, P. Lievens, J. Sauer and K.R. Asmis: Electron Distribution in Partially Reduced Mixed Metal Oxide Systems: Infrared Spectroscopy of $Ce_mV_nO_o^+$ Gas-Phase Clusters. The Journal of Physical Chemistry A **115** (41), 11187-11192 (2011).

Meek, S.A., M. Parsons, G. Heyne, V. Platschkowski, H. Haak, G. Meijer and A. Osterwalder: A traveling wave decelerator for neutral polar molecules. Review of Scientific Instruments **82** (9), 093108 (2011).

Nie, S., A.L. Walter, N.C. Bartelt, E. Starodub, A. Bostwick, E. Rotenberg and K.F. McCarty: Growth from Below: Graphene Bilayers on Ir(111). ACS Nano **5** (3), 2298-2306 (2011).

Nielsen, J.H., P. Simesen, C.Z. Bisgaard, H. Stapelfeldt, F. Filsinger, B. Friedrich, G. Meijer and J. Küpper: Stark-selected beam of ground-state OCS molecules characterized by revivals of impulsive alignment. Physical Chemistry Chemical Physics **13** (42), 18971-18975 (2011).

Omiste, J.J., M. Gärttner, P. Schmelcher, R. Gonzalez-Ferez, L. Holmegaard, J.H. Nielsen, H. Stapelfeldt and J. Küpper: Theoretical description of adiabatic laser alignment and mixed-field orientation: the need for a non-adiabatic model. Physical Chemistry Chemical Physics **13** (42), 18815-18824 (2011).

Putzke, S., F. Filsinger, H. Haak, J. Küpper and G. Meijer: Rotational-state-specific guiding of large molecules. Physical Chemistry Chemical Physics **13** (42), 18962-18970 (2011).

Riedel, J., S. Hoekstra, W. Jäger, J.J. Gilijamse, S.Y.T. van de Meerakker and G. Meijer: Accumulation of Stark-decelerated NH molecules in a magnetic trap. The European Physical Journal D **65** (1-2), 161-166 (2011).

Scharfenberg, L., K. Gubbels, M. Kirste, G.C. Groenenboom, A.v.d. Avoird, G. Meijer and S.Y.T. van de Meerakker: Scattering of Stark-decelerated OH radicals with rare gas atoms. The European Physical Journal D **65** (1-2), 189-198 (2011).

Schnell, M. and J. Küpper: Tailored molecular samples for precision spectroscopy experiments. Faraday Discussion **150**, 33-49 (2011).

Steinhauser, T., J. James, D. Hoffmann and B. Friedrich: Hundert Jahre an der Schnittstelle von Chemie und Physik: Das Fritz-Haber-Institut der Max-Planck-Gesellschaft zwischen 1911 und 2011. De Gruyter, Berlin (2011). ISBN 978-3-11-023914-0.

Todd, A.M.M., H. Bluem, V. Christina, M.D. Cole, J. Ditta, D. Dowell, K. Jordan, R. Lange, J.H. Park, J. Rathke, T. Schultheiss, L.M. Young, W. Schöllkopf, S. Gewinner, W. Erlebach, H. Junkes, A. Liedke, W. Zhang, G. von Helden, G. Meijer and S.C. Gottschalk: Commissioning Status of the Fritz Haber Institute THz FEL. In: Proceedings of 2nd International Particle Accelerator Conference, IPAC'11, San Sebastián, Spain. THPC106 (2011). ISBN 978-92-9083-366-6.

Toennies, J.P., H. Schmidt-Böcking, B. Friedrich and J.C.A. Lower: Otto Stern (1888–1969): The founding father of experimental atomic physics. Annalen der Physik **523** (12), 1045-1070 (2011).

Voloshina, E.N., A. Generalov, M. Weser, S. Böttcher, K. Horn and Y. Dedkov: Structural and electronic properties of the graphene/Al/Ni(111) intercalation system. New Journal of Physics **13** (11), 113028 (2011).

Walter, A.L., A. Bostwick, K.-J. Jeon, F. Speck, M. Ostler, T. Seyller, L. Moreschini, Y.J. Chang, M. Polini, R. Asgari, A.H. MacDonald, K. Horn and E. Rotenberg: Effective screening and the plasmaron bands in Graphene. Physical Review B **84** (8), 085410 (2011).

Walter, A.L., S. Nie, A. Bostwick, K.S. Kim, L. Moreschini, Y.J. Chang, D. Innocenti, K. Horn, K.F. McCarty and E. Rotenberg: Electronic structure of graphene on single-crystal copper substrates. Physical Review B **84** (19), 195443 (2011).

Wei, Q., S. Kais, B. Friedrich and D. Herschbach: Entanglement of polar symmetric top molecules as candidate qubits. The Journal of Chemical Physics **135** (15), 154102 (2011).

Yacovitch, T.I., T. Wende, L. Jiang, N. Heine, G. Meijer, D.M. Neumark and K.R. Asmis: Infrared Spectroscopy of Hydrated Bisulfate Anion Clusters: $HSO_4^-(H_2O)_{1-16}$. The Journal of Physical Chemistry Letters **2** (17), 2135-2140 (2011).

Doctoral Thesis

Bierau, F.: Trapping biomolecular ions in superfluid helium droplets. Freie Universität Berlin 2011.

2012

Abel, M., S. Marx, G. Meijer and G. Santambrogio: Vibrationally exciting molecules trapped on a microchip. Molecular Physics **110** (15-16), 1829-1837 (2012).

Asmis, K.R.: Structure Characterization of Metal Oxide Clusters by Vibrational Spectroscopy: Possibilities and Prospects. Physical Chemistry Chemical Physics **14** (26), 9270-9281 (2012).

Asmis, K.R. and D.M. Neumark: Vibrational Spectroscopy of Microhydrated Conjugate Base Anions. Accounts of Chemical Research **45** (1), 43-52 (2012).

Asmis, K.R., T. Wende, M. Brümmer, O. Gause, G. Santambrogio, E.C. Stanca-Kaposta, J. Döbler, A. Niedziela and J. Sauer: Structural variability in transition metal oxide clusters: gas phase vibrational spectroscopy of $V_3O_{6-8}^+$. Physical Chemistry Chemical Physics **14** (26), 9377-9388 (2012).

Baltenkov, A.S., U. Becker, S.T. Manson and A.Z. Msezane: Interference in the molecular photoionization and Young's double-slit experiment. Journal of Physics B **45** (3), 035202 (2012).

Becker, U.: Saudi University Policy: Meaningful Cooperation. Science **335** (6072), 1041-1041 (2012).

Braune, M., T. Lischke, A. Meißner, M. Ilchen, S. Deinert, J. Viefhaus, A. Knie and U. Becker: Evidence for anisotropic final state interactions in the two-photon ionization of rare gases. Journal of Physics: Conference Series **388** (3), 032074 (2012).

Cheng, T.C., L. Jiang, K.R. Asmis, Y. Wang, J.M. Bowman, A.M. Ricks and M.A. Duncan: Mid- and Far-IR spectra of H_5^+ and D_5^+ compared to the predictions of anharmonic theory. The Journal of Physical Chemistry Letters **3** (21), 3160-3166 (2012).

Crassee, I., M. Orlita, M. Potemski, A.L. Walter, M. Ostler, T. Seyller, I. Gaponenko, J. Chen and A.B. Kuzmenko: Intrinsic Terahertz Plasmons and Magnetoplasmons in Large Scale Monolayer Graphene. Nano Letters **12** (5), 2470-2474 (2012).

Erk, B., A. Rudenko, D. Rolles, B. Rudek, L. Foucar, S. Epp, M. Cryle, I. Schlichting, C. Bostedt, S. Schorb, J. Bozek, A. Rouzee, A. Hundertmark, T. Marchenko, M. Simon, F. Filsinger, L. Christensen, S. De, S. Trippel, S. Wada, K. Ueda, C. Schröter and J. Ullrich: Coulomb explosion imaging of small organic molecules at LCLS. Journal of Physics: Conference Series **388** (2), 022108 (2012).

Erlekam, U., G. Meijer and G. von Helden: Infrared spectroscopic and ab-initio studies of the benzene dimer. AIP Conference Proceedings **1504**, 487-490 (2012).

Filsinger, F., D.-S. Ahn, G. Meijer and G. von Helden: Photoexcitation of mass/charge selected hemin$^+$, caught in helium nanodroplets. Physical Chemistry Chemical Physics **14** (38), 13370-13377 (2012).

Foucar, L., A. Barty, N. Coppola, R. Hartmann, P. Holl, U. Hoppe, S. Kassemeyer, N. Kimmel, J. Küpper, M. Scholz, S. Techert, T.A. White, L. Strüder and J. Ullrich: CASS—CFEL-ASG software suite. Computer Physics Communications **183** (10), 2207-2213 (2012).

Friedrich, B.: Kolotoc pro studene molekuly. Vesmir **91** (7), 404-407 (2012).

Friedrich, B. and D. Hoffmann: Sto let fyzikalne-chemickeho vyzkumu v Berline-Dahlemu: Ustav Fritze Habera v letech 1911-2011. Ceskoslovensky Casopis pro Fyziku **62**, 110-124 (2012).

Friedrich, B., S. Kais and D. Mazziotti: Preface: Scaling Mount Impossible: A Festschrift for Dudley Herschbach. Molecular Physics **110** (15-16), 1537-1537 (2012).

Gorkhover, T., M. Adolph, D. Rupp, S. Schorb, S.W. Epp, B. Erk, L. Foucar, R. Hartmann, N. Kimmel, K.-U. Kühnel, D. Rolles, B. Rudek, A. Rudenko, R. Andritschke, A. Aquila, J.D. Bozek, N. Coppola, T. Erke, F. Filsinger, H. Gorke, H. Graafsma, L. Gumprecht, G. Hauser, S. Herrmann, H. Hirsemann, A. Hömke, P. Holl, C. Kaiser, F. Krasniqi, J.-H. Meyer, M. Matysek, M. Messerschmidt, D. Miessner, B. Nilsson, D. Pietschner, G. Potdevin, C. Reich, G. Schaller, C. Schmidt, F. Schopper, C.D. Schröter, J. Schulz, H. Soltau, G. Weidenspointner, I. Schlichting, L.W. Strueder, J. Ullrich, T. Möller and C. Bostedt: Nanoplasma Dynamics of Single Large Xenon Clusters Irradiated with Superintense X-Ray Pulses from the Linac Coherent Light Source Free-Electron Laser. Physical Review Letters **108** (24), 245005 (2012).

Gubbels, K., Q. Ma, M.H. Alexander, P.J. Dagdigian, D. Tanis, G.C. Groenenboom, A. van der Avoird and S.Y.T. van de Meerakker: Resonances in rotationally inelastic scattering of OH($X^2\Pi$) with helium and neon. The Journal of Chemical Physics **136** (14), 144308 (2012).

Gubbels, K., S.Y.T. van de Meerakker, G.C. Groenenboom, G. Meijer and A. van der Avoird: Scattering resonances in slow NH_3–He collisions. The Journal of Chemical Physics **136** (7), 074301 (2012).

Harding, D.J., C. Kerpal, G. Meijer and A. Fielicke: Activated methane on small cationic platinum clusters. Angewandte Chemie International Edition **51** (3), 817-819 (2012).

Harding, D.J., C. Kerpal, G. Meijer and A. Fielicke: Aktiviertes Methan auf kleinen Platinclusterkationen. Angewandte Chemie **124** (3), 842-845 (2012).

Harding, D.J., C. Kerpal, D.M. Rayner and A. Fielicke: Communication: The structures of small cationic gas-phase platinum clusters. The Journal of Chemical Physics **136** (21), 211103 (2012).

Härtelt, M., A. Fielicke, G. Meijer, K. Kwapien, M. Sierka and J. Sauer: Structure determination of neutral MgO clusters—hexagonal nanotubes and cages. Physical Chemistry Chemical Physics **14** (8), 2849-2856 (2012).

Härtelt, M., J.T. Lyon, P. Claes, J. de Haeck, P. Lievens and A. Fielicke: Gas-phase structures of neutral silicon clusters. The Journal of Chemical Physics **136** (6), 064301 (2012).

Hermes, A.C., S.M. Hamilton, G.A. Cooper, C. Kerpal, D.J. Harding, G. Meijer, A. Fielicke and S.R. Mackenzie: Infrared driven CO oxidation reactions on isolated platinum cluster oxides, $Pt_nO_m^+$. Faraday Discussions **157**, 213-225 (2012).

Herschbach, D., J. Rigden and B. Friedrich: Interleaved excerpts from interviews of Dudley Herschbach (DH) by John Rigden (JR) on May 21-22, 2003 and Bretislav Friedrich (BF) on March 5-9, 2012. Molecular Physics **110** (15-16), 1549-1590 (2012).

Janssens, E., S.M. Lang, M. Brümmer, A. Niedziela, G. Santambrogio, K.R. Asmis and J. Sauer: Kinetic Study of the Reaction of Vanadium and Vanadium-Titanium Oxide Cluster Anions with SO_2. Physical Chemistry Chemical Physics **14** (41), 14344-14353 (2012).

Kirste, M., X. Wang, G. Meijer, K. Gubbels, A. van der Avoird, G.C. Groenenboom and S.Y.T. van de Meerakker: Communication: Magnetic dipole transitions in the OH $A^2\Sigma^+ \leftarrow X^2\Pi$ system. The Journal of Chemical Physics **137** (10), 101102 (2012).

Kirste, M., X. Wang, C. Schewe, G. Meijer, K. Liu, A. van der Avoird, L.M.C. Janssen, K.B. Gubbels, G.C. Groenenboom and S.Y.T. van de Meerakker: Quantum-state resolved bimolecular collisions of velocity-controlled OH with NO radicals. Science **338** (6110), 1060-1063 (2012).

Koch, R.J., M. Weser, W. Zhao, F. Viñes, K. Gotterbarm, S.M. Kozlov, O. Höfert, M. Ostler, C. Papp, J. Gebhardt, H.-P. Steinrück, A. Görling and T. Seyller: Growth and electronic structure of nitrogen-doped graphene on Ni(111). Physical Review B **86** (7), 075401 (2012).

Korica, S., A. Reinköster, M. Braune, J. Viefhaus, D. Rolles, G. Fronzoni, D. Toffoli, M. Stener, P. Decleva, O.M. Al-Dossary, B. Langer and U. Becker: Appearance of Plasmons in Fullerenes. Journal of Physics: Conference Series **388** (2), 022087 (2012).

Küpper, J., F. Filsinger, G. Meijer and H. Stapelfeldt: Manipulating the motion of complex molecules: Deflection, focusing, and deceleration of molecular beams for quantum-state and conformer-selection. In: Methods in Physical Chemistry, Vol. 1. (Eds.) R. Schäfer and P.C. Schmidt. Wiley-VCH, Weinheim, 3-28 (2012). ISBN 978-3-527-3274-54.

Langer, B., R. Hentges, O. Kugeler, M. Braune, S. Korica, J. Viefhaus, D. Rolles, U. Hergenhahn, H. Fukuzawa, X. Liu, Y. Tamenori, M. Hoshino, H. Tanaka, C. Nicolas, C. Miron, O.M. Al-Dossary, K. Ueda and U. Becker: Coherent localization exhibited by unequal Auger Doppler components. Journal of Physics: Conference Series **388** (2), 022088 (2012).

Lemeshko, M. and B. Friedrich: Interaction between polar molecules subject to a far-off-resonant optical field: entangled dipoles up- or down-holding each other. Molecular Physics **110** (15-16), 1873-1881 (2012).

Majima, T., G. Santambrogio, C. Bartels, A. Terasaki, T. Kondow, J. Meinen and T. Leisner: Spatial distribution of ions in a linear octopole radio-frequency ion trap in the space-charge limit. Physical Review A **85** (5), 053414 (2012).

Meerakker, S.Y.T. van de, H.L. Bethlem, N. Vanhaecke and G. Meijer: Manipulation and Control of Molecular Beams. Chemical Reviews **112** (9), 4828-4878 (2012).

Merz, S., N. Vanhaecke, W. Jaeger, M. Schnell and G. Meijer: Decelerating molecules with microwave fields. Physical Review A **85** (6), 063411 (2012).

Mucke, M., M. Förstel, T. Lischke, T. Arion, A.M. Bradshaw and U. Hergenhahn: Performance of a short "magnetic bottle" electron spectrometer. Review of Scientific Instruments **83** (6), 063106 (2012).

Ngan, V.T., E. Janssens, P. Claes, J.T. Lyon, A. Fielicke, M.T. Nguyen and P. Lievens: High Magnetic Moments in Manganese-Doped Silicon Clusters. Chemistry - A European Journal **18** (49), 15788-15793 (2012).

Nielsen, J.H., H. Stapelfeldt, J. Küpper, B. Friedrich, J.J. Omiste and R. Gonzales-Ferez: Making the Best of Mixed-Field Orientation of Polar Molecules: A Recipe for Achieving Adiabatic Dynamics in an Electrostatic Field Combined with Laser Pulses. Physical Review Letters **108** (19), 193001 (2012).

Putzke, S., F. Filsinger, J. Küpper and G. Meijer: Alternating-Gradient Focusing of the Benzonitrile-Argon Van der Waals Complex. The Journal of Chemical Physics **137** (10), 104310 (2012).

Romanescu, C., D.J. Harding, A. Fielicke and L.-S. Wang: Probing the structures of neutral boron clusters using infrared/vacuum ultraviolet two color ionization: B_{11}, B_{16}, and B_{17}. The Journal of Chemical Physics **137** (1), 014317 (2012).

Rudek, B., D. Rolles, A. Rudenko, S. Epp, L. Foucar, B. Erk, R. Hartmann, N. Kimmel, P. Holl, C. Reich, L. Strüder, H. Hirsemann, K. Ueda, M. Simon, N. Berrah, C. Bostedt, J. Bozek, S. Schorb, M. Messerschmidt, M. Adolph, T. Gorkhover, D. Rupp, T. Möller, J. Schulz, L. Gumprecht, A. Aquila, F. Filsinger, K.-U. Kühnel and J. Ullrich: Multiphoton Ionization of Xenon at the LCLS Free-Electron Laser. Journal of Physics: Conference Series **388** (2), 022022 (2012).

Rudek, B., S.-K. Son, L. Foucar, S. Epp, B. Erk, R. Hartmann, M. Adolph, R. Andritschke, A. Aquila, N. Berrah, C. Bostedt, J. Bozek, N. Coppola, F. Filsinger, H. Gorke, T. Gorkhover, H. Graafsma, L. Gumprecht, A. Hartmann, G. Hauser, S. Herrmann, H. Hirsemann, P. Holl, A. Hömke, L. Journel, C. Kaiser, N. Kimmel, F. Krasniqi, K.-U. Kühnel, M. Matysek, M. Messerschmidt, D. Miesner, T. Möller, R. Moshammer, K. Nagaya, B. Nilsson, G. Potdevin, D. Pietschner, C. Reich, D. Rupp, G. Schaller, I. Schlichting, C. Schmidt, F. Schopper, S. Schorb, C.-D. Schröter, J. Schulz, M. Simon, H. Soltau, L. Strüder, K. Ueda, G. Weidenspointner, R. Santra, J. Ullrich, A. Rudenko and D. Rolles: Ultra-efficient ionization of heavy atoms by intense X-ray free-electron laser pulses. Nature Photonics **6** (12), 858-865 (2012).

Schöllkopf, W., S. Gewinner, W. Erlebach, H. Junkes, A. Liedke, G. von Helden, W. Zhang, G. Meijer, H. Bluem, V. Christina, M.D. Cole, J. Ditta, D. Dowell, K. Jordan, R. Lange, J.H. Park, J. Rathke, T. Schultheiss, A.M.M. Todd, L.M. Young, U. Lehnert, P. Michel, W. Seidel, R. Wünsch and S.C. Gottschalk: Status of the Fritz Haber Institute THz FEL. In: Proceedings of 33th International Free Electron Laser Conference, Shanghai, China. TUPB30 (2012). ISBN 978-3-95450-117-5.

Todd, A.M.M., H. Bluem, J. Ditta, D. Dowell, K. Jordan, R. Lange, H. Loos, J.H. Park, J. Rathke, L.M. Young, W. Erlebach, S. Gewinner, G. von Helden, H. Junkes, A. Liedke, G. Meijer, W. Schöllkopf, W. Zhang, U. Lehnert, P. Michel, W. Seidel, R. Wuensch and S.C. Gottschalk: Commissioning of the Fritz Haber Institute Mid-IR FEL. In: Proceedings of 3rd International Particle Accelerator Conference, IPAC'12, New Orleans, Louisiana, USA. TUPPP087 (2012). ISBN 978-3-95450-115-1.

Viefhaus, J., M. Ilchen, S. Deinert, L. Glaser, F. Scholz, P. Walter, M. Braune, A. Meißner, L. Tribedi and U. Becker: Oscillatory behavior of the valence photoionization properties of N_2 and O_2 due to coherent photoelectron emission from two sites. Journal of Physics: Conference Series **388** (2), 022086 (2012).

Wofford, J.M., E. Starodub, A.L. Walter, S. Nie, A. Bostwick, N.C. Bartelt, K. Thürmer, E. Rotenberg, K.F. McCarty and O.D. Dubon: Extraordinary epitaxial alignment of graphene islands on Au(111). New Journal of Physics **14** (5), 053008 (2012).

Woodham, A.P., G. Meijer and A. Fielicke: Aktivierung von molekularem Sauerstoff durch anionische Goldcluster. Angewandte Chemie **124** (18), 4520-4523 (2012).

Woodham, A.P., G. Meijer and A. Fielicke: Activation of molecular oxygen by anionic gold clusters. Angewandte Chemie International Edition **51** (18), 4444-4447 (2012).

Yacovitch, T.I., N. Heine, C. Brieger, T. Wende, C. Hock, D.M. Neumark and K.R. Asmis: Communication: Vibrational spectroscopy of atmospherically relevant acid cluster anions: Bisulfate versus nitrate core structures. The Journal of Chemical Physics **136** (24), 241102 (2012).

Zhao, B.S. and W. Schöllkopf: Fundamental physics: Molecules star in quantum movie. Nature Nanotechnology **7** (5), 277-278 (2012).

Doctoral Theses

Härtelt, M.: Action Spectroscopy of Strongly Bound Clusters in the Gas Phase. Technische Universität Berlin 2012.

Kirste, M.: OH-NO scattering at the quantum level. Freie Universität Berlin 2012.

Putzke, S.: Alternating-gradient focusing of large neutral molecules. Freie Universität Berlin 2012.

Scharfenberg, L.: Crossed beam scattering with a Stark-decelerated molecular beam. Technische Universität Berlin 2012.

Wende, T.: Gas phase infrared photodissociation spectroscopy of mass-selected ionic clusters: metal oxides and microhydrated anions. Freie Universität Berlin 2012.

Zieger, P. C.: A synchrotron for polar molecules. Radboud Universiteit Nijmegen 2012.

2013

Badino, M. and B. Friedrich: Putting the Quantum to Work: Otto Sackur's Pioneering Exploits in the Quantum Theory of Gases. In: Traditions and Transformations in the History of Quantum Physics, (Eds.) S. Katzir, C. Lehner and J. Renn. Edition Open Access, Berlin, 61-85 (2013). ISBN 978-3-8442-5134-0.

Badino, M. and B. Friedrich: Much Polyphony but Little Harmony: Otto Sackur's Groping for a Quantum Theory of Gases. Physics in Perspective **15** (3), 295-319 (2013).

Baldauf, C., K. Pagel, S. Warnke, G. von Helden, B. Koksch, V. Blum and M. Scheffler: How Cations Change Peptide Structure. Chemistry - A European Journal **19** (34), 11224-11234 (2013).

Bluem, H.P., D. Dowell, H. Loos, J. Park, A.M.M. Todd, L.M. Young, S. Gewinner and W. Schöllkopf: Accelerator Beamline Performance for the IR FEL at the Fritz-Haber-Institut, Berlin. In: Proceedings of 34th International Free Electron Laser Conference, Nara, Japan. WEOC04 (2013). ISBN 978-3-95450-123-6.

Bowlan, J., D.J. Harding, J. Jalink, A. Kirilyuk, G. Meijer and A. Fielicke: Communication: Structure of Magnetic Lanthanide Clusters from Far-IR Spectroscopy: Tb_n^+ (n = 5-9). The Journal of Chemical Physics **138** (3), 031102 (2013).

Chang, Y.-P., F. Filsinger, B.G. Sartakov and J. Küpper: CMIstark: Python package for the Stark-effect calculation and symmetry classification of linear, symmetric and asymmetric top wavefunctions in dc electric fields. Computer Physics Communications, in press

Claes, P., V.T. Ngan, M. Härtelt, J.T. Lyon, A. Fielicke, M.T. Nguyen, P. Lievens and E. Janssens: The structures of neutral transition metal doped silicon clusters, Si_nX (n = 6-9; X = V, Mn). The Journal of Chemical Physics **138** (19), 194301 (2013).

Dietl, N., T. Wende, K. Chen, L. Jiang, M. Schlangen, X. Zhang, K.R. Asmis and H. Schwarz: Structure and Chemistry of the Heteronuclear Oxo-Cluster $[VPO_4]^{\bullet +}$: A Model System for the Gas-phase Oxidation of Small Hydrocarbons. Journal of the American Chemical Society **135** (9), 3711-3721 (2013).

Erk, B., D. Rolles, L. Foucar, B. Rudek, S.W. Epp, M. Cryle, C. Bostedt, S. Schorb, J. Bozek, A. Rouzee, A. Hundertmark, T. Marchenko, M. Simon, F. Filsinger, L. Christensen, S. De, S. Trippel, J. Küpper, H. Stapelfeldt, S. Wada, K. Ueda, M. Swiggers, M. Messerschmidt, C.D. Schröter, R. Moshammer, I. Schlichting, J.H. Ullrich and A. Rudenko: Inner-shell multiple ionization of polyatomic molecules with an intense x-ray free-electron laser studied by coincident ion momentum imaging. Journal of Physics B **46** (16), 164031 (2013).

Erk, B., D. Rolles, L. Foucar, B. Rudek, S.W. Epp, M. Cryle, C. Bostedt, S. Schorb, J. Bozek, A. Rouzee, A. Hundertmark, T. Marchenko, M. Simon, F. Filsinger, L. Christensen, S. De, S. Trippel, J. Küpper, H. Stapelfeldt, S. Wada, K. Ueda, M. Swiggers, M. Messerschmidt, C.D. Schröter, R. Moshammer, I. Schlichting, J.H. Ullrich and A. Rudenko: Ultrafast Charge Rearrangement and Nuclear Dynamics upon Inner-Shell Multiple Ionization of Small Polyatomic Molecules. Physical Review Letters **110**, 053003 (2013).

Friedrich, B.: Autobiography of Bretislav Friedrich. Manipulating Molecules via EM Fields: A Festschrift for Bretislav Friedrich, Molecular Physics **111** (12-13), 1632-1642 (2013).

Friedrich, B.: Book Review: Michael Polanyi and His Generation. Origins of the Social Construction of Science. By Mary Jo Nye. Angewandte Chemie International Edition **52** (2), 498-500 (2013).

Friedrich, B.: Book Review: M. Freemantle, Gas! Gas! Quick, Boys! How Chemistry Changed the First World War. Angewandte Chemie International Edition **52** (45), 11695-11696 (2013)

Friedrich, B.: Buchbesprechung: Michael Polanyi and His Generation. Origins of the Social Construction of Science. Von Mary Jo Nye. Angewandte Chemie **125** (2), 518-520 (2013).

Friedrich, B.: Buchbesprechung: Gas! Gas! Quick, Boys! How Chemistry Changed the First World War. Von Michael Freemantle. Angewandte Chemie **125** (45), 11911-11912 (2013).

Friedrich, B.: Viewpoint: A Fragile Union Between Li and He Atoms. Physics **6**, 42 (2013).

Ghiringhelli, L.M., P. Gruene, J.T. Lyon, D.M. Rayner, G. Meijer, A. Fielicke and M. Scheffler: Not so loosely bound rare gas atoms: finite-temperature vibrational fingerprints of neutral gold-cluster complexes. New Journal of Physics **15** (8), 083003 (2013).

Gottschalk, S.C., T.E. DeHart, R.N. Kelly, M.A. Offenbacker, A.S. Valla, H. Bluem, D. Dowell, J. Rathke, A.M.M. Todd, S. Gewinner, H. Junkes, G. Meijer, W. Schöllkopf, W. Zhang and U. Lehnert: Design and Performance of the Wedged Pole Hybrid Undulator for the Fritz-Haber-Institut IR FEL. In: Proceedings of 34th International Free Electron Laser Conference, Nara, Japan. THPD13 (2013). ISBN 978-3-95450-123-6.

Grätz, F., D. Engelhart, R.J.V. Wagner, H. Haak, G. Meijer, A.M. Wodtke and T. Schaefer: Vibrational enhancement of electron emission in CO ($a^3\Pi$) quenching at a clean metal surface. Physical Chemistry Chemical Physics **15** (36), 14951-14955 (2013).

Harding, D., C. Kerpal, G. Meijer and A. Fielicke: Unusual bonding in platinum carbido clusters. The Journal of Physical Chemistry Letters **4** (6), 892-896 (2013).

Heine, N., M.R. Fagiani, M. Rossi, T. Wende, G. Berden, V. Blum and K.R. Asmis: Isomer-Selective Detection of Hydrogen-Bond Vibrations in the Protonated Water Hexamer. Journal of the American Chemical Society **135** (22), 8266-8273 (2013).

Junkes, H., W. Erlebach, S. Gewinner, U. Hoppe, A. Liedke, G. Meijer, W. Schöllkopf, G. von Helden, M. Wesemann, A.M.M. Todd, L.M. Young, H. Bluem, D. Dowell, R. Lange and S.B. Webb: Integration the FHI-FEL into the FHI Research Environment - Design and Implementation Aspects. In: Proceedings of 35th International Free Electron Laser Conference, Manhattan, USA. In press.

Junkes, H., W. Schöllkopf, M. Wesemann and R. Lange: Design and Implementation Aspects of the Control System at the FHI FEL. In: Proceedings of 14th International Conference on Accelerator & Large Experimental Physics Control Systems, San Francisco, CA, USA. In press.

Kerpal, C., D.J. Harding, A.C. Hermes, G. Meijer, S.R. Mackenzie and A. Fielicke: Structures of Platinum Oxide Clusters in the Gas Phase. The Journal of Physical Chemistry A **117** (6), 1233-1239 (2013).

Kerpal, C., D.J. Harding, J.T. Lyon, G. Meijer and A. Fielicke: N_2 Activation by Neutral Ruthenium Clusters. The Journal of Physical Chemistry C **117** (23), 12153-12158 (2013).

Kerpal, C., D.J. Harding, D.M. Rayner and A. Fielicke: Small Platinum Cluster Hydrides in the Gas Phase. The Journal of Physical Chemistry A **117** (34), 8230-8237 (2013).

Kim, K.S., A.L. Walter, L. Moreschini, T. Seyller, K. Horn, E. Rotenberg and A. Bostwick: Coexisting massive and massless Dirac fermions in symmetry-broken bilayer graphene. Nature Materials **12** (10), 887-892 (2013).

Kirste, M., H. Haak, G. Meijer and S.Y.T. van de Meerakker: A compact hexapole state-selector for NO radicals. Review of Scientific Instruments **84** (7), 073113 (2013).

Lapoutre, V.J.F., M. Härtelt, G. Meijer, A. Fielicke and J. Bakker: Communication: IR spectroscopy of neutral transition metal clusters through thermionic emission. The Journal of Chemical Physics **139** (12), 121101 (2013).

Merz, S., C. Brieger, N. Vanhaecke, G. Meijer and M. Schnell: Manipulating the motion of polar molecules with microwave radiation. Molecular Physics **111** (12-13), 1855-1864 (2013).

Nagel, P.M., J.S. Robinson, B.D. Harteneck, T. Pfeiffer, M.J. Abel, J.S. Prell, D.M. Neumark, R.A. Kaindl and S.R. Leone: Surface plasmon assisted electron acceleration in photoemission from gold nanopillars. Chemical Physics **414**, 106-111 (2013).

Ostojic, B., P. Jensen, P. Schwerdtfeger and P. Bunker: The Predicted Spectrum and Singlet–Triplet Interaction of the Hypermetallic Molecule SrOSr. The Journal of Physical Chemistry A **117** (39), 9370 - 9379 (2013).

Pagel, K. and D.J. Harvey: Ion Mobility–Mass Spectrometry of Complex Carbohydrates: Collision Cross Sections of Sodiated N-linked Glycans. Analytical Chemistry **85** (10), 5138-5145 (2013).

Pagel, K., E. Natan, Z. Hall, A.R. Fersht and C.V. Robinson: Intrinsically Disordered p53 and Its Complexes Populate Compact Conformations in the Gas Phase. Angewandte Chemie International Edition **52** (1), 361-365 (2013).

Pagel, K., E. Natan, Z. Hall, A.R. Fersht and C.V. Robinson: Intrinsically Disordered p53 and Its Complexes Populate Compact Conformations in the Gas Phase. Angewandte Chemie **125** (1), 379-383 (2013).

Savoca, M., A. Lagutschenkov, J. Langer, D.J. Harding, A. Fielicke and O. Dopfer: Vibrational Spectra and Structures of Neutral Si_mC_n Clusters ($m + n = 6$): Sequential Doping of Silicon Clusters with Carbon Atoms. The Journal of Physical Chemistry A **117** (6), 1158-1163 (2013).

Savoca, M., J. Langer, D.J. Harding, O. Dopfer and A. Fielicke: Incipient chemical bond formation of Xe to a cationic silicon cluster: Vibrational spectroscopy and structure of the Si_4Xe^+ complex. Chemical Physics Letters **557**, 49-52 (2013).

Schnell, M., U. Erlekam, P. Bunker, G. von Helden, J.-U. Grabow, G. Meijer and A. van der Avoird: Structure of the Benzene Dimer - Governed by Dynamics. Angewandte Chemie International Edition **52** (19), 5180-5183 (2013).

Schnell, M., U. Erlekam, P. Bunker, G. von Helden, J.-U. Grabow, G. Meijer and A. van der Avoird: Die Struktur des Benzoldimers – die Dynamik gewinnt. Angewandte Chemie **125** (19), 5288-5292 (2013).

Schnell, M., U. Erlekam, P. Bunker, G. von Helden, J.-U. Grabow, G. Meijer and A. van der Avoird: Unraveling the internal dynamics of the benzene dimer: A combined theoretical and microwave spectroscopy study. Physical Chemistry Chemical Physics **15** (25), 10207-10223 (2013).

Schöllkopf, W., S. Gewinner, W. Erlebach, G. Heyne, H. Junkes, A. Liedke, V. Platschkowski, G. von Helden, W. Zhang, G. Meijer, H. Bluem, M. Davidsaver, D. Dowell, K. Jordan, R. Lange, H. Loos, J. Park, J. Rathke, A.M.M. Todd, L.M. Young, U. Lehnert, P. Michel, W. Seidel, R. Wünsch and S.C. Gottschalk: First Lasing of the IR FEL at the Fritz-Haber-Institut Berlin. In: Proceedings of 34th International Free Electron Laser Conference, Nara, Japan. MOOB0 (2013). ISBN 978-3-95450-123-6.

Wanko, M., T. Wende, M.M. Saralegui, L. Jiang, A. Rubio and K.R. Asmis: Solvent-mediated folding in dicarboxylate dianions: Aliphatic chain length dependence and origin of the IR intensity quenching. Physical Chemistry Chemical Physics, in press.

Warnke, S., G. von Helden and K. Pagel: Communication: Protein Structure in the Gas Phase: The Influence of Side-Chain Microsolvation. Journal of the American Chemical Society **135** (4), 1177-1180 (2013).

Woodham, A.P., G. Meijer and A. Fielicke: Charge Separation Promoted Activation of Molecular Oxygen by Neutral Gold Clusters. Journal of the American Chemical Society **135** (5), 1727-1730 (2013).

Woodham, A.P. and A. Fielicke: Gold Clusters in the Gas Phase. In: Structure and Bonding, (Ed.) M. Mingos. Springer, Berlin. In press.

Yacovitch, T.I., N. Heine, C. Brieger, T. Wende, C. Hock, D.M. Neumark and K.R. Asmis: Vibrational spectroscopy of bisulfate/sulfuric acid/water clusters: Structure, stability, and infrared multiple-photon dissociation intensities. The Journal of Physical Chemistry A **117** (32), 7081-7090 (2013).

Zhang, X., C. Eyles, C.A. Taatjes, D. Ding and S. Stolte: A general scaling rule for the collision energy dependence of a rotationally inelastic differential cross-section and its application to NO(X) + He. Physical Chemistry Chemical Physics **15** (15), 5620-5635 (2013).

Zhao, B.S., W. Zhang and W. Schöllkopf: Non-destructive quantum reflection of helium dimers and trimers from a plane ruled grating. Molecular Physics **111** (12-13), 1772-1780 (2013).

Zhu, J., S. Kais, Q. Wei, D. Herschbach and B. Friedrich: Implementation of quantum logic gates using polar molecules in pendular states. The Journal of Chemical Physics **138** (2), 024104 (2013).

Zieger, P.C., C.J. Eyles, G. Meijer and H.L. Bethlem: Resonant excitation of trapped molecules in a molecular synchrotron. Physical Review A **87** (4), 043425 (2013).

Doctoral Theses

Kerpal, C.: Structure and chemistry of platinum group metal clusters. Radboud Universiteit Nijmegen 2013.

Master's Thesis

Esser, T.: The effect of partially deiodination on the structure of periodinated closo-dodecaborate probed by infrared photodissociation spectroscopy. Freie Universität Berlin 2013.

Invited Talks of the Members of the Department of Molecular Physics

David Adu Smith

Feb 2013	Cold Atoms Seminar, School of Physics and Astronomy, University of Nottingham, Nottingham, UK *Prethermalization in a Non-Equilibrium Many-Body Quantum System*
Apr 2013	RTG Colloquium, Hanover, Germany *Prethermalization in a Non-Equilibrium Many-Body Quantum System*

Knut R. Asmis

Sep 2011	19th International Conference on Horizons in Hydrogen Bond Research, Göttingen, Germany *Microhydration of Conjugate Base Anions Probed by Gas Phase Vibrational Spectroscopy*
Sep 2011	10. Clustertreffen der deutschsprachigen Clusterphysik, Rothenfels, Germany *Microhydration of Conjugate Base Anions Probed by Gas Phase Vibrational Spectroscopy*
Jan 2012	GDCh-Kolloquium der University of Ulm, Ulm, Germany *Isomer-Selective Double Resonance IR-IR Spectroscopy*
Feb 2012	3rd Genesis Research Institute Symposium on Cluster Science, Nagoya, Japan *Gas Phase Vibrational Spectroscopy of Transition Metal Oxide Clusters*
May 2012	111. Bunsentagung, Leipzig, Germany *Isomer-Selective IR/IR Double Resonance Spectroscopy: Isolating the Spectral Signatures of* $H^+(H_2O)_6$ *Isomers*
Jun 2012	Kolloquium für Physikalische, Biophysikalische und Theoretische Chemie, Technische Universität München, Munich, Germany *Isomer-Selective Double Resonance IR-IR Spectroscopy*
Sep 2012	19th International Mass Spectrometry Conference, Kyoto, Japan *Isomer-Selective Double Resonance IR-IR Spectroscopy*
Sep 2012	Physical Chemistry Seminar, Tohoku University, Sendai, Japan *Recent Advances in the Vibrational Spectroscopy of Mass-Selected Cluster Ions*
Jan 2013	Seminar, Wilhelm Ostwald Institute of Physical and Theoretical Chemistry, Leipzig University, Leipzig, Germany *Isomer-selektive IR-Spektroskopie an protonierten Wasserclustern: Ein grenzübergreifender Beitrag zum Verständnis von* $H^+(aq)$

Feb 2013	24th Meeting of the Australian and New Zealand Society for Mass Spectrometry (ANZSMS24), Melbourne, Australia *Isomer-Selective Detection of Hydrogen-Bond Vibrations in the Protonated Water Hexamer*
Feb 2013	Physical Seminar, Department of Chemistry, University of Georgia, Athens, GA, USA *Recent Advances in the Vibrational Spectroscopy of Mass-Selected Ions*
Feb 2013	Gordon Research Conference on Gaseous Ions: Structures, Energetics and Reactions, Galveston, TX, USA *Recent Advances in the Gas Phase Vibrational Spectroscopy of Mass-Selected Ions*
Sep 2013	2nd COST-IOTA Workshop on Cold Molecular Ions, Arosa, Switzerland *IR^2MS^2 Population Labeling Spectroscopy: Isomer-Specific IR Signatures of Protonated Water Clusters*
Sep 2013	VUV FELs in Molecular, Cluster and Surface Science, Dalian, China *Characterizing the Structure, Reactivity and Dynamics of Gas Phase Clusters Using FEL Radiation*

Uwe Becker

May 2012	International Workshop on Correlation and Polarization Phenomena in Ionization, State University, Moscow, Russia *Coherence and Loss of Coherence in the Photoionization of Inversion Symmetric Systems*

André Fielicke

Jul 2012	CECAM Workshop on Theoretical Challenges in Electronic Structure of Clusters and Nanoparticles, Lausanne, Switzerland *Structure Determination of Free Metal Clusters via Far-Infrared Vibrational Spectroscopy*
Sep 2012	CRC-FHI Joint Meeting on Complex Surfaces in Material Science, Berlin, Germany *Vibrational Spectroscopy of Metal Cluster Complexes: Investigating the Chemistry on a Small Surface*
Oct 2012	International Workshop of the Research Unit 1282 Controlling the Electronic Structure of Semiconductor Nanoparticles by Doping and Hybrid Formation, Berlin, Germany *Structure and Dynamics of Pure and Doped Silicon Clusters*
Mar 2013	DPG-Frühjahrstagung, Hanover, Germany *Size Selective Vibrational Spectroscopy of Strongly Bound Neutral Clusters*

Jun 2013	11th European Conference on Atoms, Molecules and Photons, ECAMP 11, Aarhus, Denmark *Vibrational Spectroscopy of Metal Cluster Complexes: Investigating the Chemistry on a Small Surface*
Jul 2013	International Symposium on Atomic Cluster Collisions - ISACC 2013, Wuhan, China *Vibrational Spectroscopy of Metal Cluster Complexes: Investigating the Chemistry on a Small Surface*
Aug 2013	Seminar, Department of Chemistry, John Hopkins University, Baltimore, MD, USA *Shedding IR Light on Gas Phase Metal Clusters: Insights into Structures and Reactions*
Aug 2013	XXth International Materials Research Congress, Cancun, Mexico *Shedding IR Light on Gas Phase Metal Clusters: Insights into Structures and Reactions*
Sep 2013	7th International Conference on the Theory of Atomic and Molecular Clusters (TAMC VII), Birmingham, UK *Clusters of the Platinum Group Metals: Insights from IR Spectroscopy into Structures and Reactions*
Oct 2013	Clustertreffen der deutsprachigen Clusterphysik, Herzogenhorn, Germany *Shedding IR Light on Gas Phase Metal Clusters: Insights into Structures and Reactions*

Bretislav Friedrich

Sep 2011	Coherence and Decoherence at Ultralow Temperatures Institute for Advanced Study, Technical University Munich, Munich, Germany *Entanglement of Trapped Polar Molecules*
Oct 2011	Fundamental Science with Ultracold Molecules, ITAMP, Harvard University, Cambridge, MA, USA *Molecules in Far-Off Resonance Light*
Oct 2011	Centenary of the Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany *One Hundred Years at the Intersection of Chemistry and Physics - Book Presentation*
Dec 2012	Colloquium, ETH Zurich, Zurich, Switzerland *Shedding Nonresonant Light on Polar Molecules*
Feb 2013	Lecture, Kavli Institute for Theoretical Physics, University of California, Santa Barbara, CA, USA *Shedding Nonresonant Light on Molecules*

Nadja Heine

Jan 2012	Gordon Research Conference on Molecular and Ionic Clusters, Ventura, CA, USA *Isomer-Selective IR/IR Double Resonance Spectroscopy: Isolating the Spectral Signatures of* $H^+(H_2O)_6$ *Isomers*
Mar 2013	Symposium on Size Selected Cluster, Davos, Switzerland *Vibrational Spectroscopy of Atmospherically Relevant Cluster Anions*
May 2013	112. Bunsentagung, Karlsruhe, Germany *Vibrational Spectroscopy of Atmospherically Relevant Clusters:* $NO^-_3(HNO_3)_m(H_2O)_n$
Jul 2013	Seminar, Institute for Atmospheric and Enviromental Sciences, Goethe University Frankfurt, Frankfurt a. Main, Germany *Vibrational Spectroscopy of Atmospherically Relevant Clusters*

Gert von Helden

Oct 2011	Laboratory Astrochemistry External Retreat, Heidelberg, Germany *Spectroscopy in He Droplets*
Jan 2012	Isolated Biomolecules and Biomolecular Interactions, Les Diablerets, Switzerland *Deep-Freezing Mass/Charge Selected Biomolecular Ions in Liquid Helium Droplets*
Sep 2012	18th European Conference on the Dynamics of Molecular Systems, Oxford, UK *Proteins in the Freezer: Embedding Mass/Charge Selected Biomolecular Ions in Liquid Helium Droplets*
Jan 2013	FELIX Abschiedssymposium, Nieuwegein, The Netherlands *FELIX as a Heatgun*
Mar 2013	Symposium on Size Selected Clusters, Davos, Switzerland *Proteins in the Freezer: Embedding Mass/Charge Selected Biomolecular Ions in Liquid Helium Droplets*
Apr 2013	Seminar, École Polytechnique Fédérale de Lausanne, Lausanne, Switzerland *Biomolecules in the Gas Phase and in Liquid Helium Droplets*
May 2013	Seminar, University of Basel, Basel, Switzerland *Mass/Charge Selected Biomolecular Ions in the Gas Phase and Deep-Frozen in Liquid Helium Droplets*

Karsten Horn

Sep 2011	5. Tagung der Vereinigung der Spanischen Synchrotronstrahlungsnutzer (AUSE-5), Valencia, Spain *Photoemission Studies of Graphene*
Dec 2011	Symposium, Schwerpunktprogramm Graphen, Rothenfels, Germany *Untersuchungen der Struktur von Graphen auf Halbleitern und Metallen*

Mikhail P. Lemeshko

Sep 2011	Lecture, The Institute for Theoretical Atomic, Molecular and Optical Physics, Harvard-Smithsonian Center for Astrophysics, Cambridge, MA, USA *Interactions of Polar Molecules Dressed by Far-Off-Resonant Light: Entangled Dipoles Up- or Down-Holding Each Other*
Jun 2012	DAMOP Thesis Prize talk, Orange County, CA, USA *Molecular Interactions in and with Fields: Thermal Collisions, Ultracold Gases, Supersymmetry*

Sebastiaan Y. T. van de Meerakker

Nov 2011	International Workshop on Ultra-Cold Molecules, Stellenbosch, South Africa *Cold and Controlled Collisions Using Stark-Decelerated Molecular Beams*

Gerard Meijer

Sep 2011	Lecture Series at Summerschool on Complex Quantum Systems (CoQuS), Vienna, Austria *Manipulation of Molecules with Electric Fields, Parts I-III*
Sep 2011	Workshop on Coherence and Decoherence at Ultracold Temperatures, Munich, Germany *Collision Studies with Stark-Decelerated Molecular Beams*
Sep 2011	Opening Plenary Lecture, COMET, Oxford, UK *Collision Studies with Stark-Decelerated Molecular Beams*
Sep 2011	Instituts Kolloquium, Max-Born-Institut, Berlin, Germany *Taming Molecular Beams*
Oct 2011	Topical Meeting on Fundamental Science with Ultracold Molecules, ITAMP, Cambridge, MA, USA *Collision Studies with Stark-Decelerated Beams*
Oct 2011	ITAMP Seminars and Joint Atomic Physics Colloquium Series, Harvard University, Cambridge, MA, USA *Collision Studies with Stark-Decelerated Molecular Beams*

Jan 2012	Physikalisches Kolloquium Mainz, University of Mainz, Mainz, Germany *Taming Molecular Beams*
Feb 2012	IMM Colloquium, Radboud University Nijmegen, Nijmegen, The Netherlands *What to Do with an Infrared Free Electron Laser? Gas-Phase Experiments on Molecules and Clusters*
Apr 2012	503rd Wilhelm-und-Else-Heraeus-Seminar on Free-Electron Lasers: From Fundamentals to Applications, Bad Honnef, Germany *(F)IR-FEL Experiments on Molecules and Clusters*
Apr 2012	International Workshop on Zeeman Deceleration, ETH Zurich, Zurich, Switzerland *Manipulation of the Motion of Molecules with Microwave Fields*
Apr 2012	International Workshop on Exploiting Free Electron Lasers in Chemistry, University of Nottingham, Nottingham, UK *Vibrational Spectroscopy on Gas-Phase Molecules and Clusters*
Apr 2012	Marie Curie Symposium on Tomorrow's Physics, Radboud University Nijmegen, Nijmegen, The Netherlands *Cold Molecules; Towards a Molecular Laboratory on a Chip*
May 2012	506th Wilhelm-und-Else-Heraeus-Seminar on Infrared: Science, Technology and Applications, Bad Honnef, Germany *(F)IR-Spectroscopy with Free Electron Lasers: Studies on Molecules and Clusters*
Jul 2012	Plenary Lecture, 44th Conference of the European Group on Atomic Systems (EGAS), University of Gothenburg, Gothenburg, Sweden *Taming Molecular Beams*
Jul 2012	Gordon Research Conference on Atomic and Molecular Interactions, Stonehill College, Easton, MA, USA *Taming Molecular Beams*
Jul 2012	Gordon Research Conference on Electronic Spectroscopy and Dynamics, Bates College, Lewiston, ME, USA *On Decelerated Beams and Trapped Molecules*
Nov 2012	30th Annual Meeting of the Swiss Group for Mass Spectrometry, Beatenberg, Switzerland *Taming Beams of Neutral Molecules*
Mar 2013	Winterschool on Chemical Reaction Dynamics at Surfaces, Ringberg Castle, Germany *Molecular Beams with a Tunable Velocity*
Jun 2013	11th European Conference on Atoms, Molecules and Photons, ECAMP 11, Aarhus, Denmark *Molecules on a Chip*
Sep 2013	IOP Visitor Lecture, Queen's University, Belfast, UK *Taming Beams of Neutral Molecules*
Sep 2013	IOP Visitor Lecture, Dublin City University, Dublin, Ireland *Taming Beams of Neutral Molecules*

Sep 2013	IOP Visitor Lecture, University College Cork, Cork, Ireland *Taming Beams of Neutral Molecules*
Oct 2013	Donders Beyond Borders Lecture, Radboud University Nijmegen, Nijmegen, The Netherlands *Taming Beams of Neutral Molecules*

Gabriele Santambrogio

Dec 2011	LENS, Firenze, Italy *A Stark Decelerator on a Microchip*
Nov 2012	Seminar, MPI of Quantum Optics, Garching, Germany *Molecules on a Microchip*
Apr 2013	Gruppensemiar, Genesis Research Institute, Inc. - Toyota Technological Institute, East Tokyo Laboratories, Tokyo, Japan *A Molecules Chip*
Apr 2013	Gruppensemiar, Department of Chemistry (AG Akira Terasaki), Kyushu University, Fukuoka, Japan *A Molecules Chip*
Apr 2013	Gruppensemiar, Department Basic Sciences (AG Fumitaka Mafune), University of Tokyo, Tokyo, Japan *A Molecules Chip*
May 2013	Fotonica 2013, Milano, Italy *Molecules Chip*
Jun 2013	Seminar, Istituto Nazionale di Ricerca Metrologica, Torino, Italy *A Molecules Chip*

Wieland Schöllkopf

Mar 2012	DPG-Frühjahrstagung, Stuttgart, Germany *Quantum Reflection and Matter-Wave Optics with Helium Atoms and Molecules*
Apr 2012	Seminar, DFG-Forschergruppe FOR 1282 - Controlling the Electronic Structure of Semiconductor Nanoparticles by Doping and Hybrid Formation, Berlin, Germany *The IR FEL at the FHI - Current Status and Prospects*
Aug 2012	5th Symposium on Gas-Phase Reaction Dynamics and Laser Spectroscopy, Korea Advanced Institute for Science and Technology, Daejeon, Korea *The IR Free-Electron Laser at the FHI: A New Tool for Molecules and Cluster Spectroscopy*
Aug 2012	The 34th International Free Electron Laser Conference, Nara, Japan *First Lasing of the IR FEL at the FHI, Berlin*

Jan 2013	Kolloquium, Institute of Physics, University of Rostock, Rostock, Germany *Quantum Reflection and Matter-Wave Optics with Helium Atoms and Molecules*
Feb 2013	Lecture Series Catalysis, FHI, Berlin, Germany *Introduction to the IR Free-Electron Laser at the FHI*

Nicolas Vanhaecke

Nov 2011	IOTA-COST Workshop on molecular ions, Sandbjerg, Denmark *Manipulation and Control of Molecular Beams with Electric and Magnetic Fields*
Feb 2012	Joint seminar week, Diavolezza, Switzerland *A Traveling Wave Zeeman Decelerator*
May 2012	95th Canadian Chemistry Conference and Exhibition, Calgary, AB, Canada *Manipulation and Control of Molecular Beams with Electric and Magnetic Fields*
Nov 2012	ESF Conference in Partnership with LFUI on Cold and Ultracold Molecules, Obergurgl, Austria *Manipulation of Cold Molecular Ensembles*

Department of Physical Chemistry **Director: Martin Wolf**

Staff scientists:

Markus Eiswirth		Takashi Kumagai	*from 05/2013*
Leonhard Grill		Alexey Melnikov	
Kramer Campen		Alexander Mikhailov	
Karsten Horn	*from 01/2012*	Alexander Paarmann	*from 01/2014*
Tobias Kampfrath		Bruno Pettinger	*until 02/2013*
Patrick Kirchmann	*until 08/2012*	Julia Stähler	

Gerhard Ertl (Emeritus)

Guest scientists, staying for at least six months:

Yves Dehouc	*until 11/2011*	Kamaraju Natarajan	*until 09/2012*
Keunsu Kim	*until 06/2013*	Andrea Rubano	*until 09/2012*
Nikolaos Kouvaris	*until 01/2013*	Toru Shimada	*until 02/2012*
Arantzazu Mascaraque	*until 08/2012*	Akitoshi Shiotari	
Sajadi Hezaveh Mohsen			

Scientists (temporary), paid from external funds:

Yunpei Deng		*EU*	Melke Nascimiento		*DFG*
Holger Flechsig		*DFG*	Zefeng Ren	*until 12/2011*	*AvH Fellow*
Shigefumi Hata		*JSPS*	Alexander Saywell		*EU/DFG*
Marc Herzog		*EU/DFG*	Yujin Tong		*DFG*
Pablo Kaluza	*until 01/2013*	*DFG*	Jaeyoung Lee		*AvH Fellow*
Takashi Kumagai	*until 04/2013*	*JSPS Fellow*	Hamilton Varela		*FAPESP*
Michael Meyer		*EU/DFG*	Simon Wall	*until 08/2012*	*AvH Fellow*
Claude Monney		*AvH/SNF*	Lizandra Zimmermann		*DAAD*
Christophe Nacci		*EU*			

Graduate students:	19	(9 from external funds)
Diploma students:	12	
Trainees:	8	
Technicians:	9	

Max Planck Research Group: Structural and Electronic Surface Dynamics

Head: Ralph Ernstorfer

2 postdocs
3 graduate students
2 diploma students

Service Group Electronic Lab

Head: Georg Heyne — 15 staff members 2 apprentices

Service Group Mechanical Workshop

Head: Petrik Bischoff — 18 staff members 2 apprentices

Recent Developments in the Department of Physical Chemistry

Director: Martin Wolf

Max Planck Research Group: Structural and Electronic Surface Dynamics

Head: Ralph Ernstorfer

1. General

Since the last meeting of the Fachbeirat, the Department of Physical Chemistry has developed its experimental and laboratory infrastructure to become fully operational. Currently the research groups of the department are still distributed over several locations on the FHI campus and in rented external lab space. However, the construction of the new building for the department is now on its way and will be completed in early 2016.

Several changes have occurred among the group leaders and senior postdocs of the department:

- *Dr. Zefeng Ren* received an offer for an associate professorship at Peking University and started there in January 2012.
- *Prof. Karsten Horn* (formerly Department of Molecular Physics) joined the department in January 2012. He provides expertise in angle-resolved photoemission spectroscopy and continues his research on electronic structure of grapheme-based materials.
- *Dr. Simon Wall* received an offer for an assistant professorship at ICFO (The Institute of Photonic Science) in Barcelona and left the department in August 2012 to set up an independent research group.
- Also in August 2012 *Dr. Patrick Kirchmann* left for a staff scientist position at SLAC National Accelerator Laboratory in Stanford.
- *Dr. Leonhard Grill* accepted an offer for a full professorship at the University of Graz and started this position in August 2013. He is still affiliated with the department in order to complete a EU funded project (AtMol) until end of 2014.
- Since May 2013 *Dr. Takashi Kumagai* is heading a new research group to study elementary molecular processes at surfaces employing low-temperature atomic force and scanning tunneling microscopy (AFM/STM) as well as tip-enhanced Raman spectroscopy (TERS).
- Starting in January 2014, *Dr. Alexander Paarmann* will set up a new group to employ the new infrared free electron laser (FHI-FEL) for time-resolved optical spectroscopy of ultrafast dynamics of solids, in particular phonon dynamics. He will complement these studies using femtosecond table-top sources.

Furthermore, several postdoc associates and graduate students moved on to new positions in industry or administration.

Two large service groups of the institute, the Electronics Lab and the Mechanical Workshops (since 2013), are currently associated with the department. After the retirement of *Horst*

Schwäricke in April 2013, *Petrik Bischoff*, formerly with the Department of Molecular Physics, was appointed as the new head of the Mechanical Workshops. Currently the services of the Mechanical Workshops are expanded and the machine infrastructure is modernized, a process which will continue over the next few years. The Electronics Lab (headed by *Georg Heyne*) is well organized and continues to provide excellent service for the institute.

1.1 Research of the Department

The research of the Department of Physical Chemistry is focused on the dynamics of elementary processes at surfaces, interfaces and in solids. Our goal is to develop a microscopic understanding of the dynamics of molecular and electronic processes as well as the interactions between various (electronic, spin and lattice) degrees of freedom. The general strategy is to address these problems from several sides using complementary approaches, in particular by the development and application of various spectroscopic techniques dedicated to the specific physical questions. Research is performed by small groups with specific expertise. Although the research of each individual group is mostly curiosity driven our complementary approach creates various synergies between the different groups.

The research of the department is currently structured into two main areas, (I) ultrafast dynamics of elementary processes in solids and at interfaces, and (II) molecular processes at surfaces and is carried out by the research groups listed below.

Ultrafast Dynamics in Solids and at Interfaces

- Dynamics of Correlated Materials (*Martin Wolf*)
- Electron Dynamics at Interfaces (*Julia Stähler*)
- Electronic Structure of Surfaces and Interfaces (*Karsten Horn*)
- Terahertz Physics (*Tobias Kampfrath*)
- Time-resolved Second Harmonic Generation Spectroscopy (*Alexey Melnikov*)

Molecular Processes at Surfaces

- Nanoscale Science (*Leonhard Grill*)
- Nanoscale Surface Chemistry (*Takashi Kumagai*)
- Interfacial Molecular Spectroscopy (*Kramer Campen*)
- Complex Systems (*Alexander Mikhailov*)
- Spatiotemporal Self-organization (*Markus Eiswirth*)

Max-Planck-Research Group

- Structural and Electronic Surface Dynamics (*Ralph Ernstorfer)*

The Max-Planck-Research Group (MPRG) of *Ralph Ernstorfer* on "Structural and Electronic Surface Dynamics" is closely associated with the department and complements the research on ultrafast dynamics of solids by time-resolved electron diffraction techniques.

The first line of research aims at studying the dynamics of elementary processes on the relevant time scales of the process by ultrafast laser spectroscopy (typically with femto- or picoseconds time resolution). The department applies a broad spectrum of established as well as newly developed techniques; these are used to study the electronic excitations and low energy excitations in solids, dynamics of electron transfer processes, vibrational dynamics at interfaces, as well as optically induced phase transitions. The second line of research is the investigation of elementary molecular processes either at the single molecule level, or by employing various schemes of optical excitations including photo-induced surface reactions. Scanning probe microscopy (in part combined with optical excitation) permits imaging, manipulation and spectroscopy as well as inducing and probing chemical processes of individual molecules. Further activities address problems of molecular biophysics and electrochemistry. Here, theoretical studies of molecular machines are performed, complemented by studies of nonlinear dynamics and pattern formation in electrochemical systems.

The promotion of young scientists is an important goal and several measures are taken to help them developing their career (*e.g.* IMPRS graduate school, PhD student and department workshops, regular status discussions, nomination for awards and invited talks). Recently several prizes have been awarded to junior scientists, namely the Feynman Prize (Foresight Institute) to *Leonhard Grill*, the Carl Ramsauer Award (German Physical Society Berlin) to *Laurenz Rettig* and the Inoue Research Award for Young Scientists to *Takashi Kumagai*.

Furthermore, junior staff scientists and postdocs are guided to gain experience in grant applications and establish their scientific network. Currently, the department cooperates in several EU funded projects, four collaborative research centers (Sfb 658, Sfb 910, Sfb 951, Sfb 1109) and one research unit (FOR 1700) funded by the Deutsche Forschungsgemeinschaft (DFG). These projects are complemented by several individual DFG research grants of young scientist (see section 3 for a complete list of projects).

1.2 Selected Research Highlights

The following research highlights are a selection of results from research projects as well as instrument development in the department obtained during the last two years:

- Progress was achieved in the area of time and angle-resolved resolved photoemission spectroscopy (trARPES, (hν = 6 eV)) applied to various correlated materials (high-Tc superconductors, Fe-pnictides and charge density wave (CDW) systems). A systematic study of the tri-telluride CDW system demonstrated direct probing of the transient modulation of the CDW gap as well as vibrational coherent control of the amplitude mode using a three-pulse excitation scheme. Very recently, ultrafast melting of the spin-density-wave phase in Cr was studied with trARPES using XUV light obtained by high harmonics generation. [Phys. Rev. Lett. **108**, 097002 (2012), ibid. **107**, 097002 (2011)].
- The ultrafast evolution of the photoinduced insulator-to-metal transition in VO_2 was probed with a white light supercontinuum. Using the optical phonons of the insulating phase as a marker, an ultrafast change of the lattice potential symmetry (i.e. the restoring forces) was identified on a timescale much faster than the structural phase transition leading to a transient excited state which differs from the equilibrium metallic state of VO_2. [Nature Commun. **3**, 721 (2012), Phys. Rev. B **87**, 115126 (2013)].
- A high repetition rate (500 kHz) laser system for high harmonics generation of XUV laser pulses (hν = 20 – 40 eV) in combination with a state-of-the-art ARPES experiment is currently being developed. This system employs multiple stages of fiber lasers, a slab laser amplifier and optical parametric chirped pulse amplification (OPCPA) to generate < 20 fs pulses with more that 20W output power in the visible.
- Bilayer graphene band calculations show a feature that is very desirable but absent in the monolayer – a band gap. Transport experiments, however, have so far failed to detect a clear gap. Using angle-resolved photoemission, this contradiction is explained by showing that small twists in the relative bilayer arrangement lead to the coexistence of massive Dirac particles (expected in the bilayer) with a massless particle band that crosses the ideal bilayer gap. [Nature Materials **12**, 887 (2013)].
- It is still not clear why quasicrystals, which have perfect atomic order yet lack translational periodicity, assume their complex structures. Using bulk-sensitive hard x-ray photoemission evidence for a large pseudogap near the Fermi level is obtained, supporting a Hume-Rothery mechanism for quasicrystal formation. Quasicrystals apparently form a

metallic phase at the surface that masks, when using surface sensitive photoemission, the true bulk electronic structure. [Phys. Rev. Lett. **109**, 216403 (2012)].

- The ultrafast quasiparticle dynamics at the ZnO(10-10) single crystal surface following above-band gap excitation was probed by time-resolved two-photon photoelectron spectroscopy, exhibiting ultrafast electron cooling in the conduction band by electron-phonon scattering and followed by formation of a surface-bound exciton.
- The transport of very short bunches of spin-polarized electrons (spin-current pulses) in magnetic heterostructures has been demonstrated employing THz emission spectroscopy as a probe. Using femtosecond laser excitation, the spin transport was launched from a ferromagnetic Fe thin film into a nonmagnetic cap layer of low (Ru) or high mobility (Au), which results in spin trapping (Ru) or ballistic traversal (Au). The results are potentially useful for future spintronics circuitry operated at highest (THz) frequencies [Nature Nanotech. **8**, 256 (2013)].
- A high-precision optical setup for time-resolved linear and non-linear magneto-optical spectroscopy has been developed, operating in back pump-front probe scheme with 20 fs time resolution. Femtosecond spin current pulses were demonstrated in epitaxial Fe/Au/Fe/MgO(001) multilayers [Phys. Rev. Lett. **107,** 076601 (2011)].
- Manipulation of the spectrum or temporal shape of a light pulse has been achieved by coupling femtosecond laser pulses into waveguides. For example, by suddenly tilting the waveguide's dispersion relation the spectrum can be compressed, a process which is reversible, features high conversion efficiency and could find application as a magnifying lens for optical spectra. Furthermore, the concept for an ultrafast optical delay line could be demonstrated [Phys. Rev. Lett. **108**, 033902 (2012), *ibid.* **108**, 213901 (2012)].
- A breakthrough has been achieved using femtosecond time-resolved x-ray spectroscopy (RIXS) at the LCLS free electron laser in Stanford to probe the electronic structure of CO molecules, as their chemisorption state on Ru(0001) changes upon exciting the substrate by using a femtosecond optical laser pulse. The observed electronic structure changes are consistent with a transient weakening of the CO-metal bond without notable desorption, indicationg the existence of two distinct adsorption wells, a chemisorbed and a precursor state, separated by an entropy barrier [Science **339**, 1302 (2013); Phys. Rev. Lett. **110**, 186101 (2013)].

- Hierarchical “bottom-up” covalent binding of molecular building blocks in a well-defined pattern was demonstrated by sequentially supplying reactive sites on molecular building blocks. Copolymer networks were formed with high spatial selectivity. After dissociation of Br substituents from molecular building blocks on gold surfaces the polymerization to straight chains along the step edges was demonstrated, resulting in a pre-alignment along a given direction over the entire sample. [Nature Chemistry **4**, 215 (2012), Angew. Chem. Int. Ed. **51**, 5096 (2012)].
- The electrical current through a single molecule could be measured at different voltages over a large range. In this way, the conductance properties of an individual polymer could be correlated with its electronic states for the first time. Comparison with calculation reveals that the conductance depends on the precise atomic structure and the bending of the molecule in the STM junction. [Nature Nanotech. **7**, 713 (2012)].
- The intramolecular H-atom transfer reaction (tautomerization) within a single porphycene molecule on Cu(110) was controlled directly by low-temperature STM. The potential landscape of this process can be precisely tuned by putting single Cu adatoms nearby or by changing the orientation of neighboring molecules. Furthermore, the mechanism of thermally and vibrationally-induced tautomerization was deduced from isotope effects and the bias voltage and tunneling current dependence [Phys. Rev. Lett. **111**, 246101 (2013), Nature Chemistry (in press 2013)].
- A new experimental setup for sum frequency generation (SFG) spectroscopy has been developed allowing studies of liquid solid interfaces as well as probing low-frequency vibrations (e.g. surface phonons) down to ~700 cm^{-1}. The dissociative adsorption of water (D_2O) on α-Al_2O_3(0001) has been studied both in UHV and under ambient conditions by (1) characterizing the fragments via the OD stretch vibration and (2) the accompanying surface reconstruction using the Al-O surface phonon SFG spectral response.
- By combined experimental and computational studies of the non-hydrogen bonded (free) OH groups at the air/water interface, it was shown that these are structurally and dynamically heterogeneous on sub-picosecond timescales, and that 2/3 of their vibrational relaxation proceeds via intramolecular energy transfer and 1/3 via reorientation [J Phys. Chem. B, **116**, 9467 (2012), ibid. **117**, 11753 (2013), Proc. Nat. Acad. Sci (in press 2013)].

- Coarse-grained elastic-network numerical investigations of two macromolecules, playing a fundamental role in the cells, have been performed. For the molecular motor myosin, its strain-sensor behavior, previously found in single-molecule experiments, was explained and communication between important functional domains of the protein could be elucidated [Biophys. J. **102**, 542-551 (2012)].
- Many protein machines operate as active inclusions in lipid bilayers forming biological membranes. Fast and efficient methods have been developed for numerical simulations of lipid bilayers in membranes with active protein inclusions indicating the hydrodynamic effects should play a principal role in interactions between active membrane inclusions [J. Chem. Phys. **137**, 055101 (2012) and J. Chem. Phys. **138**, 195101 (2013)].

2. Progress Report

2.1 Ultrafast Dynamics in Solids and at Interfaces

Elementary processes in solids and at interfaces such as transfer of charge and spin, energy dissipation, or electron-phonon coupling are the underlying microscopic basis of much more complex phenomena, ranging from surface reactions to phase transitions in solids. The study of the non-equilibrium, ultrafast dynamics of such fundamental processes provides mechanistic insights into the interplay and energy exchange between electron, spin and lattice degrees of freedom. It is the strategy of the department to elucidate the dynamics of elementary processes from various sides using complementary approaches and techniques. These approaches are implemented by several groups in the department, which perform real-time studies of solids and interfaces on ultrafast time-scales, complemented by studies of the electronic structure.

2.1.1 Transient Electronic Structure of Correlated Materials

Correlated electron materials exhibit exotic electronic and magnetic properties, characterized by broken-symmetry ground states such as metal-to-insulator instabilities, unconventional superconductivity, and various cooperative ordering phenomena. One of the major challenges in this field is to understand the ground and excited state properties on a microscopic level and to disentangle the competing interactions and correlations of charge, spin, orbital and lattice degrees of freedom, which act on multiple length, energy and time scales.

Ultrafast laser spectroscopy provides a tool to access elementary scattering and relaxation processes by optically exciting the electronic system and subsequently probing the evolution of the transient electronic structure by an appropriate spectroscopic technique. For example, in a material undergoing an insulator-to-metal transition, optical excitation can induce a transient melting of the band gap whereby the timescale of the gap closing is characteristic for a mechanism driven by purely electronic correlations (Mott transition) or by ion motions (Peierls instability). The technique of time- and angle-resolved photoemission spectroscopy (trARPES) extends the benefits of momentum-resolved photoelectron spectroscopy into the time domain and provides direct access to the transient evolution of the electronic structure after optical excitation. Furthermore, the collective dynamics of lattice or spin excitations can be studied through their influence on the quasiparticle band structure.

As prototypical charge density wave (CDW) system the group of *Martin Wolf* has investigated the material class of rare-earth tri-tellurides, RTe_3 (RTe_3, R = Te, Ho, Dy) using trARPES with a 6 eV fs laser probe (in collaboration with U. Bovensiepen, Duisburg and

Z.-X. Shen, Stanford). Using a position sensitive detector for two-dimensional imaging of the photoelectron momentum, the transient changes of the Fermi surface and the opening and closing of the CDW gap could be mapped directly on a femtosecond time scale (see Fig. 1). By employing a novel three pulse "pump-reexcite-probe" photoemission scheme, the dynamics of the upper and lower CDW band edges could be resolved in great detail, indicating that the CDW gap modulation (amplitude mode) originates from a complex lattice motion whereby at least two coupled phonon modes are involved. A more detailed analysis of the band collapse reveals a transient reduction of the curvature of the Fermi surface, which is attributed to a transient change of the Te5p orbital overlap in the excited state. This leads to reduced coupling between neighbouring Te chains and thus to a modification in the dimensionality of the 2D band structure. Furthermore, vibrational coherent control has been demonstrated for the amplitude mode using double pulse excitation.

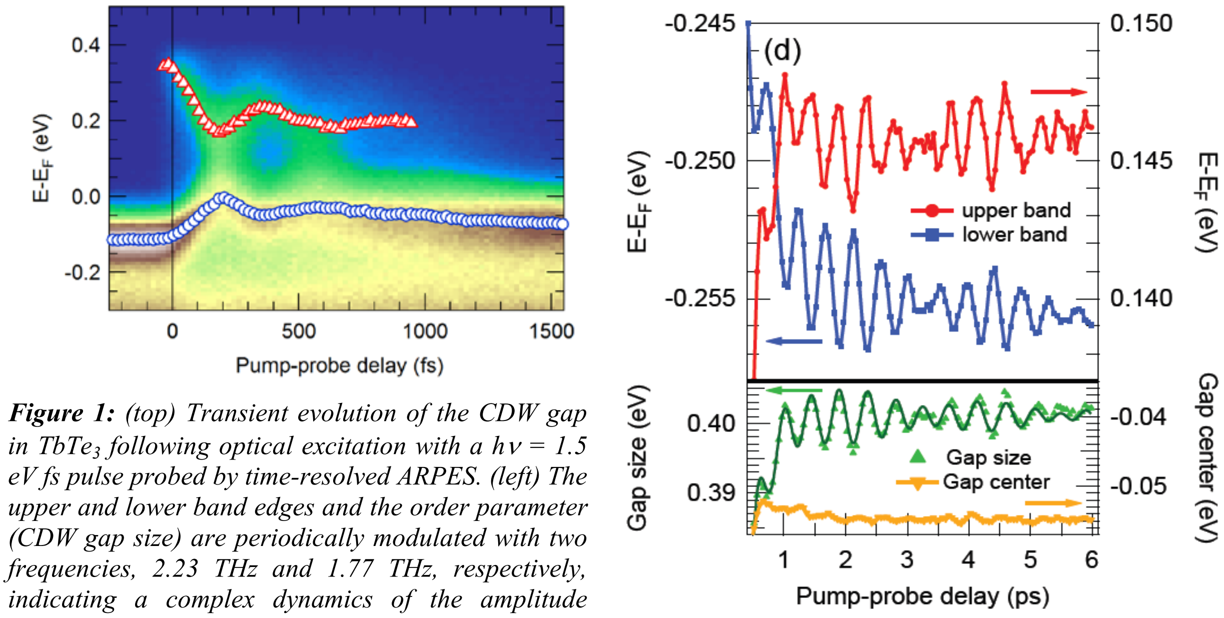


Figure 1: *(top) Transient evolution of the CDW gap in TbTe$_3$ following optical excitation with a hν = 1.5 eV fs pulse probed by time-resolved ARPES. (left) The upper and lower band edges and the order parameter (CDW gap size) are periodically modulated with two frequencies, 2.23 THz and 1.77 THz, respectively, indicating a complex dynamics of the amplitude mode.*

Further studies using trARPES with hν = 6 eV have probed the temperature-dependent relaxation times of photoexcited electrons and holes in antiferromagnetic Fe-pnictide, $EuFe_2As_2$, and have attributed their dynamics with the spin density wave gap and the single-particle band at the zone center, respectively [1]. The recovery of magnetic order after ultrafast excitation occurs four times slower compared to electron-phonon equilibration due to a smaller phase space for spin-dependent relaxation.

To overcome the limited accessible *k*-space due to the low kinetic energy of photoelectron using 6 eV laser ARPES, considerable efforts are currently undertaken to set up an advanced trARPES experiment based on a high repetition laser source (100 kHz – 1 MHz) for high harmonics generation (HHG) of XUV laser pulses (hν = 20 – 40 eV) in combination with a state-of-the-art ARPES ultrahigh vacuum system. In addition to the possibility of accessing several Brillouin zones, the HHG-based trARPES will provide much enhanced counting statistics. This experiment employs novel fibre and OPCPA laser technology and is developed in close collaboration with the MPRG group of *Ralph Ernstorfer* (see report MPRG).

On the way to develop HHG-based trARPES experiments, the group has recently performed two studies using the access to exisiting HHG setups, which operate at 1-10 kHz based on established TiSapphire laser technology. In collaboration with M. Weinelt, Freie Universität Berlin, the prototypical spin density wave (SDW) material chromium has been studied. An ultrafast melting of the SDW phase was revealed from the response of the related backfolded band, which can be compared to a transient change in electronic temperature. Very recently, the charge density wave (CDW) material $TiSe_2$ was studied with HHG-based trARPES in a beamtime at the ARTEMIS light source at Rutherford Appleton Lab (UK). The mechanism responsible for the CDW phase transition is still heavily debated. For $TiSe_2$ temperature dependent measurements were conducted as well as long-wavelength excitation to investigate the peculiar physics of this system.

In the group of *Julia Stähler,* the ultrafast electron and lattice dynamics of vanadium dioxide (VO_2) across the insulator-to-metal transition were investigated by means of transient optical spectroscopy using a white light supercontinuum, and by time-resolved two-photon photoelectron spectroscopy. VO_2 undergoes a phase transition from a monoclinic, insulating phase at low temperatures and a rutile, metallic phase above $T_{\text{crit}} = 340$ K. This transition can also be induced by photoexcitation, enabling its investigation on ultrafast time scales. Using Raman-active optical phonons of the monoclinic phase as a sensor, it could be shown that the *symmetry* of the restoring forces is lost within few femtoseconds, even before ionic motion occurs. Re-excitation of the non-equilibrated system, i.e. a pump-probe experiment of the excited state, furthermore unveils that the VO_2 has not reached the metallic phase yet at these early times after excitation. The optical response resembles that of the thermodynamically stable phase only after picoseconds [2].

[1] L. Rettig *et al.,* Phys. Rev. Lett. **108**, 097002 (2012).

[2] S. Wall, D. Wegkamp, L. Foglia, K. Appavoo, J. Nag, R. F. Haglund, J. Stähler, M. Wolf, *Nature Commun.* **3** (2012) 721; S. Wall *et al., Phys. Rev. B* **87**, 115126 (2013).

2.1.2 Ultrafast Carrier and Exciton Dynamics in Inorganic/Organic Hybrid Systems

The combination of inorganic semiconductors with organic molecules to hybrid systems (HIOS) promises superior functionality of the interface compared to a bare linear combination of the single material properties. Applications such as organic LEDs or solar cells would not only benefit from the high charge carrier mobility and stability of the inorganic compound in combination with the tunable optical properties of the organic molecules, but could also make use of interfacial hybrid states that facilitate, for example, charge or energy transfer between the constituents.

A promising candidate for such applications is zinc oxide (ZnO) due to its wide band gap (3.4 eV), n-type conductivity and abundance. Despite several decades of research, a full understanding of the surface properties of ZnO remains elusive. The group of *Julia Stähler* has investigated the electronic structure and ultrafast carrier dynamics of ZnO, its (10-10) surface, and interfaces with organic molecular layers with femtosecond (fs) time-resolved two-photon photoemission (2PPE) and optical spectroscopy. The non-polar ZnO(10-10) surface exhibits a downward surface band bending when terminated with atomic hydrogen, leading to the formation of a charge accumulation layer below the Fermi level E_F with a density on the order of 10^{-13} cm^{-2}, as shown in Fig. 2a. This goes along with a work function decrease of up to $\Delta\Phi = -0.6$ eV due to charge donation from the hydrogen. Significantly stronger reduction of Φ can be achieved using the dipolar molecule pyridine with negative electron affinity (Fig. 2b). Here, due to the lack of Fermi level pinning, a huge work function decrease of $\Delta\Phi = -2.9$ eV could be demonstrated in excellent agreement with *ab-initio* calculations by the group of *Patrick Rinke*, Theory Department. [1]

As illustrated in Fig. 2a, above band gap excitation of ZnO leads to the creation of excited electrons in the conduction band (CB). The subsequent ultrafast relaxation dynamics (see processes 1 & 2 in Fig. 2a) are monitored in time-resolved 2PPE by a second, time-delayed fs laser pulse ($h\nu_{probe}$). Fig.2c displays in false colors how the excited electron population relaxes on fs timescales by first electron-phonon scattering towards lower energies, and finally ends up in an excitonic surface state (SX) *below* E_F. The binding energy of more than 200 meV with respect to the bulk CB minimum is responsible for its long lifetime exceeding several 100 ps. Remarkably, such SX is not observed for $SrTiO_3$ (STO), another n-type transparent conducting oxide that exhibits also a 2D electron gas at its surface. Here, however, the charge density is at least one order of magnitude larger than for ZnO, which screens the electron-hole Coulomb attraction.

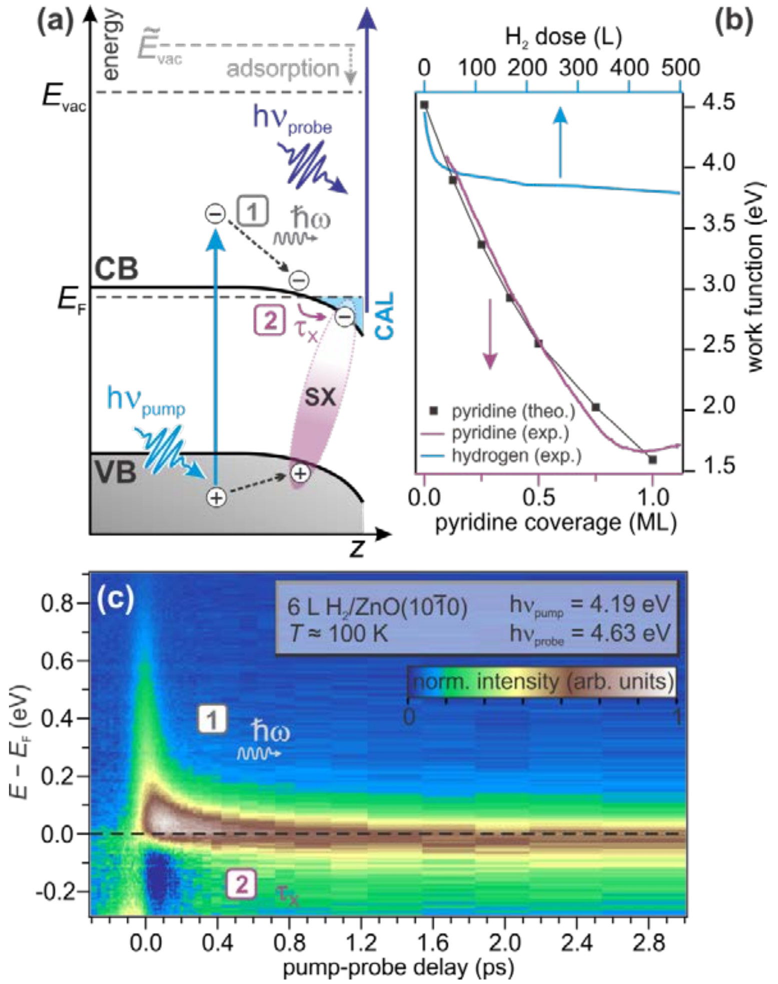


Figure 2: *(a) Two-photon-photo-emission (2PPE) and scattering processes at the ZnO(10-10) surface. (b) Work function change due to adsorption of hydrogen (blue line) and pyridine (purple line, markers DFT calculation) (c) Ultrafast electron dynamics at the ZnO surface as probed by 2PPE spectroscopy. False colors represent additional electrons excited by the pump pulse. Hot carriers in the CB relax on fs timescales by optical phonon emission. After a few 100 fs, additional electrons are observed below E_F, indicating that a surface exciton has formed.*

The combination of ZnO with organic and optically active molecules is a crucial step towards application. The group has characterized the interface of the organic molecule SP6 (*2,7-bis(biphenyl-4-yl-)2',7'-ditertbutyl-9,9'-spirobifluorene*) with several oxide surfaces by means of non-resonant Raman spectroscopy down to the monolayer level. [2] First optical transient transmission experiments probing the LUMO → LUMO+1 resonance of the molecules point at efficient charge separation at the interface to ZnO. These are complemented by 2PPE studies of the electronic structure of the SP6/ZnO(10-10) interface.

[1] O. T. Hofmann, J.-C. Deinert, Y. Xu, P. Rinke, J. Stähler, M. Wolf, M. Scheffler, *J. Chem. Phys.* **139**, 174701 (2013).

[2] J. Stähler, O. T. Hofmann, P. Rinke, S. Blumstengel, F. Henneberger, Y. Li, T. F. Heinz, *Chem. Phys. Lett.* **584**, 74 (2013).

2.1.3 Electronic Structure of Surfaces and Interfaces

Among the low dimensional materials, graphene has an exceptional status since its electronic structure brings together solid state physics and quantum electrodynamics. Also many real-world applications are envisaged and thus an enormous research effort is under way.

The group of *Karsten Horn* has investigated graphene oxidation, for example using NO_2 and SO_2, and fluorination by interaction with PF_3 and XeF_2. The latter is a particularly interesting process, as "half-fluorinated graphene" has been found, where a fluorine atom is attached to every second carbon atom, is an insulating phase with a large band gap. A metastable phase exists which transitions back to the ground state under emission of blue light. The graphene bilayer has received specific attention since it is supposed to exhibit a small band gap, desirable for electronic switching applications. This has, however, failed to appear in electronic transport measurements. Photoemission data from our collaboration with groups at the Advanced Light Source (Lawrence Berkeley Laboratory), and TU Chemnitz, reveal the reasons for this: they show the coexistence of massive Dirac Fermions with massless ones which bridge the gap predicted from tight binding calculations (see Fig. 3, e.g. features C1 and C2) [1]. An analysis of the data shows that small (~ 0.2°) twists between the two layers, unavoidable even in the highest quality samples, lead to a gradual variation of the relative lateral arrangement of the carbon atoms in the two layers, (A - B and A - A stacking), which induces the above

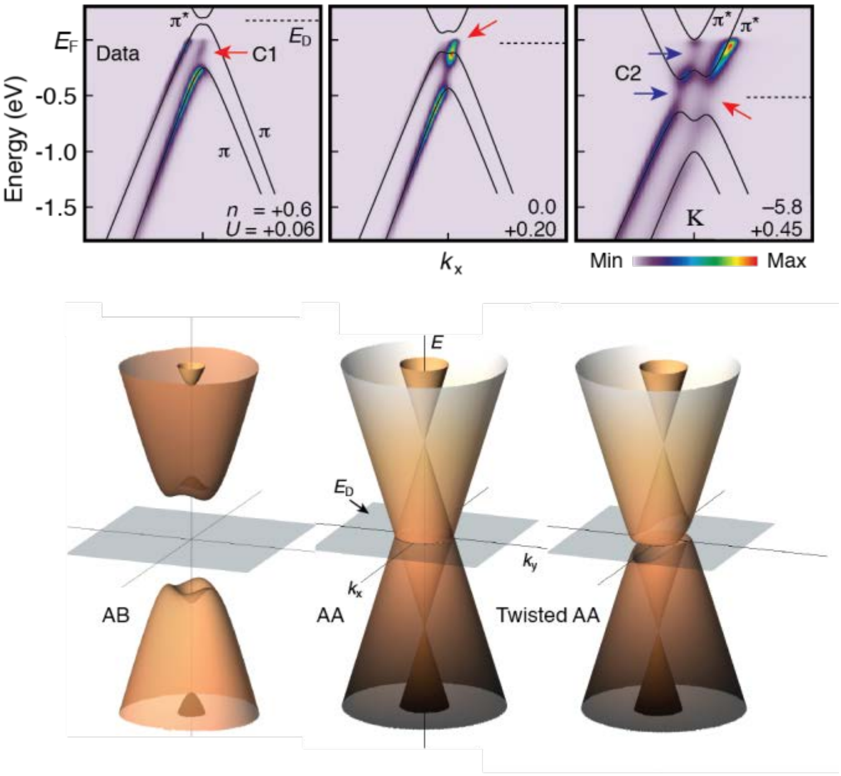


Figure 3: *Top: Experimental band dispersion of hole-doped (left), nearly-neutral (center), and electron-doped (right) bilayer graphene with overlaid lines from tight-binding band structure calculations for AB stacking, doping level n (units 10^{13} cm^{-2}) and Coulomb energy U (units eV). Bottom: Schematic band structure of bilayer graphene with AB, AA, and twisted AA stacking.*

coexistence of massive and massless states in the band structure, as shown schematically in Fig. 3 (bottom).

Furthermore, graphene's interaction with metals has been studied, for intercalation of metals (Cu, Co, Mn) in between graphene on Ir(111). Here, novel phases of these materials could be examined, since the intercalated atoms assume the (larger) lattice constant of the iridium substrate. Graphene deposited on ferromagnetic materials has been predicted to act as spin filter; hence we have studied magnetic interaction in such junctions using x-ray magnetic circular dichroism studies, for example in nickel-grapheme-cobalt "sandwich" layers, where the magnetic coupling between nickel and cobalt across the graphene layer was established. With the aim of graphene functionalization, the adsorption of ammonia and water on graphene has been studied on different substrates (Ni and Ir) showing that the strength of graphene's interaction with its substrate has a marked effect on the nature of the chemisorption interaction.

Further studies on Al-Pd-Mn and Al-Cu-Fe quasicrystals using bulk-sensitive hard x-ray photoemission (HXPES) have provided evidence for the existence of a large pseudogap near the Fermi level (characteristic for a Hume-Rothery mechanism), which is not observed in surface sensitive low energy photoemission, because the spectrum is affected by a metallic phase formed near the surface that masks the true bulk electronic structure [2].

[1] K.S. Kim *et al., Nature Materials* **12**, 887 (2013).
[2] J. Nayak *et al., Phys. Rev. Lett.* **109**, 216403 (2012).

2.1.4 Terahertz Physics: Low-energy Excitations and Control by THz pulses

The terahertz (THz) frequency range is of central relevance from a fundamental-scientific as well as from an application-related point of view. First, many elementary excitations in physical systems have transition energies on the order of 10 meV, for example excitations of quasi-free electrons in solids, crystal lattice vibrations, and excitons in semiconductors. As 1 THz corresponds to a energy of 4.1 meV, these modes can be probed resonantly and with sub-picosecond time resolution using THz electromagnetic field pulses. With recent advances in THz pulse generation providing electric-field amplitudes of ~1 MV cm^{-1} it has become possible to even drive and control such resonances on sub-picosecond time scales [1]. Second, bit rates in current information technology may soon approach the THz frequency range. Therefore, it is important to develop ultrafast techniques to manipulate electric currents or light at THz frequencies, for example with modulators and frequency shifters.

The group of *Tobias Kampfrath* makes use of ultrashort THz and optical laser pulses to investigate the interplay of low-energy excitations in complex materials and to control the properties of matter and light at the highest frequencies. Currently, the group focuses on electron spins in magnetically ordered solids, and studies new schemes to manipulate the dynamics and transport of magnetization. This goal also addresses basic questions of spin-phonon interaction and spin-orbit coupling.

Ultrafast spin transport: Future electronics will potentially not only make use of the electron charge as an information carrier, but also employ the electron spin (up/down) to encode the value of a bit. Successful implementation of such "spintronics" requires the transfer of electron spins through space as well as the manipulation of the spin state. These elementary operations should proceed at a pace exceeding that of today's computers, that is, at THz frequencies. To study and control ultrafast spin transport, a spin-polarized current pulse was launched in a Fe thin film through illumination with a femtosecond laser pulse [Fig. 4(a)].

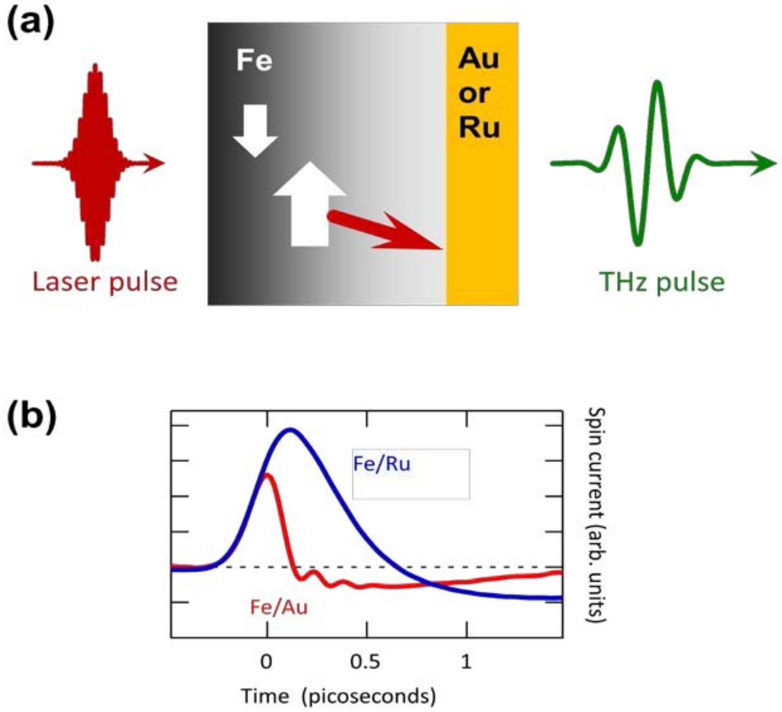


Figure 4: *(a) Schematic of generation, manipulation, and detection of THz spin currents. A femtosecond laser pulse launches a spin current from a Fe thin film (10nm) to a metallic cap layer (2nm) of either Ru or Au. While transport is slowed down in Ru (low electron mobility), it is fast in Au (high electron mobility). The spin current is converted into a charge current (inverse spin Hall effect), resulting in the emission of THz radiation whose detection allows extraction of the spin current dynamics. (b) Spin currents in Fe/Au and Fe/Ru as determined from the THz transients emitted by these heterostructures. The dynamics in Fe/Ru is much slower than in Fe/Au showing that adding layers of different mobility provide a route for manipulation of spin current bursts.*

Such a spin current arises because the laser pulse promotes spin-up electrons from d-type states with low band velocity into sp-like states with high velocity, whereas the minority spin-down electrons remain d-type and, thus, slow. To manipulate these bursts, the Fe film was contacted with another nonmagnetic film of either low (Ru) or high (Au) electron mobility. As a result, the spin transport in the Fe/Au heterostructure should proceed much faster than in

Fe/Ru. To probe the spin flow in a contactless manner, the inverse spin Hall effect is used that (through spin-orbit coupling) converts the longitudinal spin current into a transverse charge current, thereby leading to the emission of a detectable THz electromagnetic transient (see Fig. 4a). The measurements indeed verify the expected behavior: the spin current in Fe/Au exhibits a much faster dynamics than in the low-mobility Fe/Ru structure (see Fig. 4b). These findings are relevant because they show that the inverse spin Hall effect is still operative even at THz frequencies and that ultrafast spin currents can be delayed in a relatively simple manner [2].

Ultrafast spin manipulation: To manipulate the magnetization of a ferromagnetic metal film on ultrafast time scales, the sample is usually illuminated with a femtosecond laser pulse, thereby depositing energy in the electronic subsystem. The resulting reduction of magnetization on a 100-fs time scale is not yet understood, despite considerable experimental and theoretical efforts since the first experiments in 1996. To freeze out the complex electronic degrees of freedom, we recently focused on the ferrimagnetic insulator yttrium iron garnet (YIG, electronic band gap 2.8eV). Energy is brought into the system by exclusively pumping optical phonons with an intense THz pulse, thereby leaving the electronic subsystem unchanged. First results demonstrate a fast demagnetization with a 1.1 ps time constant, which is extremely surprising because spin-lattice relaxation in YIG is known to occur on much slower time scales of 1 ns and more. The results indicate that the coupling of spins with optical phonons is orders of magnitude stronger than with acoustic phonons. Currently a detailed study is under way to understand the microscopic origin of this observation.

Surface/interface sensitivity: So far, THz pulses have exclusively addressed the bulk properties of samples rather than their interfaces. The group of *Tobias Kampfrath* currently investigates the possibility to measure currents flowing at interfaces of topological insulators. Similar to the scheme of Fig. 4a, currents are launched by a circular polarized fs laser pulse in surface states of topological insulators and then measured by detecting the emitted THz electromagnetic transient (THz emission spectroscopy).

Manipulation of light on-the-fly: Besides controlling spins as information carriers, also new schemes for light manipulation have been tested. The basic idea is to couple light into a photonic structure and then vary its photonic mode structure by applying a femtosecond control pulse. The light inside the structure will then adiabatically follow these changes and finally emerge with a modified shape. To test this scheme, a photonic crystal was used and light at the telecommunication band around 200 THz coupled into it. By applying a spatially shaped femtosecond control pulse, free charge carriers were induced in selected regions of the photonic crystal, thereby (1) shifting and (2) even tilting the dispersion relation (frequency vs wave vector) of the structure. As a result, the probe pulse followed these changes adiabatically, resulting in (1) a blueshift (by up to 0.3 THz) and (2) a compression of the pulse spectrum (by up to 10%) with high efficiency (up to 80%). The frequency shifting is potentially useful for shifting signal trains in optical telecommunication to different frequency channels, whereas the spectral compression may find application as a spectral lens [3].

[1] T. Kampfrath, K. Tanaka, K.A. Nelson, *Nature Photon.* **7**, 680 (2013).

[2] T. Kampfrath *et al.*, *Nature Nanotech.* **8**, 256 (2013).

[3] D. M. Beggs, T. F. Krauss, L. Kuipers, T. Kampfrath, *Phys. Rev. Lett.* **108**, 033902 (2012);
D. M. Beggs *et al.*, *Phys. Rev. Lett.* **108**, 213901 (2012).

2.1.5 Ultrafast Spin Dynamics in Epitaxial Metallic Multilayers

Ultrafast spin dynamics induced by transport of photoexcited spin polarized carriers is of fundamental interest for magnetic applications like spintronics and data storage. To study the underlying elementary processes on femtosecond time scales, the group of *Alexey Melnikov* has developed a time domain approach that probes the spin dynamics induced by hot carriers (HC) in a back-pump-front-probe scheme. In a first experiment the transport of spin polarized HC through a Au/Fe/MgO(001) stack has been demonstrated [1]: Thereby, optical excitation of hot carriers in the Fe film is followed by superdiffusive transport to the Au surface where the carrier density and spin polarization are detected by magneto-induced second harmonic (SH) generation. Later on it has been shown that the ballistic HC fraction can be controlled by the Fe layer thickness and the duration of the ballistic spin current pulse can be as short as 30 fs. The spin dynamics in Fe was studied by a combination of SH and magneto-optical Kerr effect.

To proceed towards a new concept of metal-based elements for femtosecond spintronics, Fe/Au/Fe/MgO(001) stacks with different thickness of Fe layers were used, which allow for parallel or anti-parallel alignment of the magnetizations M_E and M_C in emitter and collector (see Fig. 5). The SH electric field consists of E_{even} and E_{odd} components which are even and odd with respect to magnetization reversal. The relative pump-induced variation $\Delta_{odd}=\Delta E_{odd}/E_{odd}$ characterize variations of the magnetization at the interfaces. Since optical phases are sensitive to interface properties, E_{odd} generated at Fe/Au and Fe/MgO interfaces can interfere constructively or destructively, which leads, respectively, to large or small magnetic contrast $\rho \approx 2\, E_{odd}/E_{even}$. In the first case Δ_{odd} is sensitive to the average (sum) of the magnetizations at both interfaces (Fig. 5b) while in the second case Δ_{odd} monitors variations of the difference between two interface magnetizations (Fig. 5a).

In Fe/Au/Fe structures with parallel alignment, minority HC which traverse the Au layer within 40 fs [1] are accumulated at the collector Fe/Au interface. This reduces the interface M_C and thus the absolute value of E_{odd}, which results in positive Δ_{odd} (Fig. 5b) due to destructive interference of interface contributions. In the case of antiparallel alignment, ballistic HC (minority HC with respect to M_E) are majority HC with respect to M_C and thus are not accumulated at the interface. The interface demagnetization increasing Δ_{odd} (Fig. 5b) occurs upon the arrival of diffusive HC (now with negative spin) on a timescale of 200 fs [1]. Finally, the HC transport leads to the demagnetization of the Fe/Au interface and occurs on the timescale of ballistic or diffusive HC transport for parallel or antiparallel configurations, respectively.

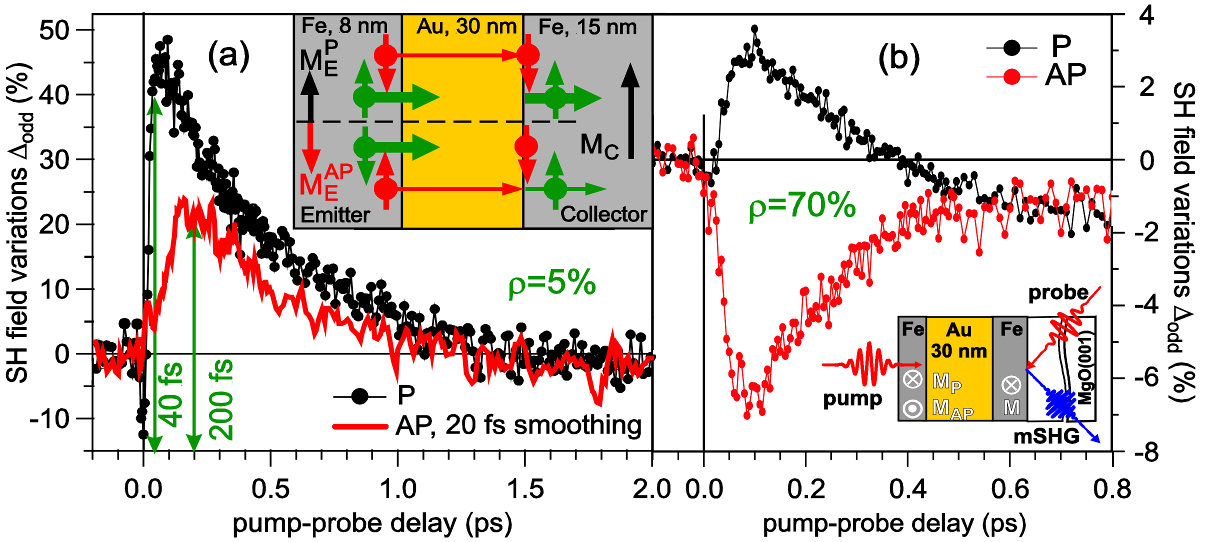


Figure 5: *Pump-induced changes of the odd second harmonic (SH) field Δ_{odd} which probes the spin dynamics in Fe/Au/Fe structures for (a) a 8 nm-thick emitter and small collector magnetic contrast ρ=5% (a) and (b) a 5 nm-thick emitter and large contrast ρ=70%. The parallel (P) and anti-parallel (AP) orientations of magnetizations in the emitter (M_E) and collector (M_C) are shown in the experimental schemes in the insets. Inset (a): The large ballistic propagation length of majority HC in Fe $\lambda^{\uparrow}_{Fe} >> \lambda^{\downarrow}_{Fe}$ leads to more effective emission of majority HC (thick green vs. thin red arrows) and to the accumulation of minority HC at the Fe/Au interface of the collector. However, in Au $\lambda^{\uparrow}_{Au} << \lambda^{\downarrow}_{Au}$ and hot carrier transport is ballistic (fast) for minority and diffusive (slow) for majority carriers (long red vs. short green arrows).*

In Fe/Au/Fe structures with large magnetic contrast (Fig. 5b), the role of ballistic HC providing small changes of interface M_C due to their small concentration is not significant and we can consider only diffusive (majority with respect to M_E) HC. The reversal of M_E permits to alternate between positive and negative variations of M_C on a femtosecond timescale, which is promising for the future development of spintronics devices.

[1] A. Melnikov *et al.*, *Phys. Rev. Lett.* **107,** 076601 (2011).

2.2 Molecular Processes at Interfaces

Understanding molecular processes at a microscopic level of single molecule reactions, interfacial charge and energy transfer and vibrational dynamics provides fundamental insight into surface reactions. Studies of molecular processes at interfaces are performed by several groups in the department which employ complementary techniques with high spatial resolution as well as chemical sensitivity using vibrational or x-ray spectroscopy. In these studies, surface reactions and molecular rearrangements are stimulated by thermal activation, excitation by light or interfacial charge transfer. These are complemented by computational studies of biomolecular machines and pattern formation in electrochemical systems.

2.2.1 Nanoscience with Functional Molecules

The research activities in the group of *Leonhard Grill* focus on the investigation and manipulation of single functional molecules on surfaces by scanning tunnelling microscopy (STM), preferentially at low temperatures of 5 K. This method makes it possible to image single molecules with very high spatial resolution, and to manipulate them by chemical forces, tunnelling electrons or the electric field in the junction.

An important class of functional molecules are molecular switches that exhibit at least two stable states with characteristic physical and/or chemical properties. The research group has studied various types of molecular switches on metal surfaces. One example follows previous work of the group on azobenzene derivatives with four *tert*-butyl side groups, but here these groups were attached at slightly different positions of the benzene rings (*para* instead of *meta* position). These molecules are found to be prochiral on a Au(111) surface, and enantiomerically pure islands are observed. In manipulation experiments chirality switching is observed, where single molecules change from one enantiomer to the other, probably by a twofold internal rotation. In another study, the group has investigated covalently connected multiple switching systems where the coupling of the different switching units is of particular interest. It could be shown that bisazobenzene molecules with two switching units can be deposited onto a Au(111) surface under clean ultrahigh vacuum conditions, and that they self-organize in large ordered islands of different arrangements depending on their chemical structure. While lateral manipulation can be achieved, no switching processes could be induced by voltage pulses over different parts of the molecules and in different environments. When changing the switching unit and studying imine derivatives on Au(111), it was found that the molecular layer gradually transforms from a nearly complete *trans*- to a nearly complete *cis*-monolayer with increasing molecular coverage [1].

Another focus in the field of functional molecules is on so-called nanomachines, which are objects with dimensions of few nanometers that perform work – a key vision in molecular nanotechnology. Functional molecules are of particular interest in this regard and the research group has studied so-called motorized nano-cars which should move on a surface upon illumination, essentially as a result of an isomerization process of the "motor" unit. After the very difficult synthesis, the intact deposition of such a complex molecule under ultrahigh vacuum conditions represents a central challenge. It could be shown that intact molecules can be sublimed in vacuum onto a Cu(111) surface and that they can be imaged there as individual entities [2]. The molecular appearance in the STM images is in good agreement with the molecular dimensions in the gas phase, and according to their chemical structure, two typical conformations are identified. However, lateral motion on the surface, in particular by activating the molecular motor, has not been achieved so far.

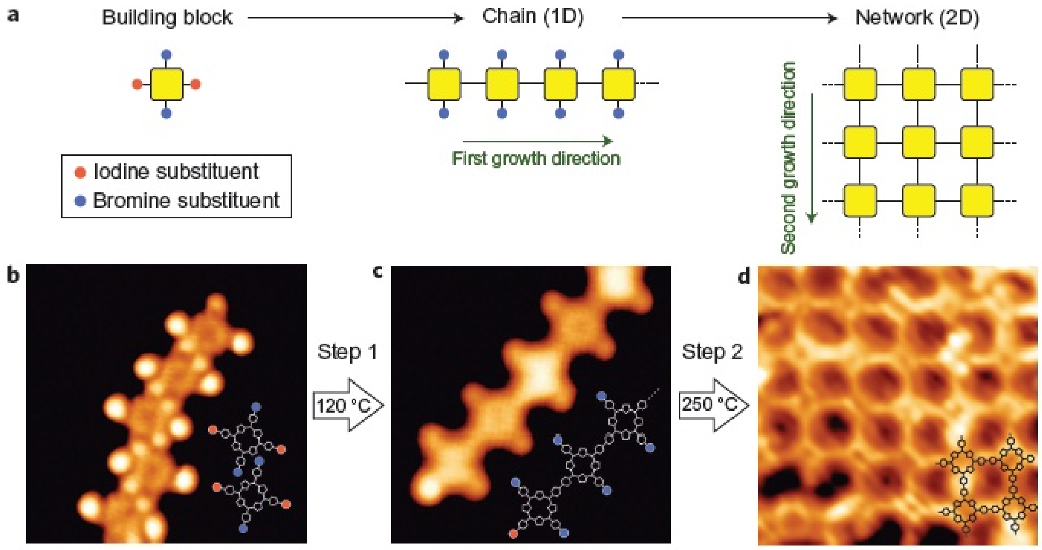


Figure 6: *Hierarchical growth following sequential thermal activation. (a) Scheme of the activation mechanism. Arrows indicate the different growth directions of the two sequential steps. (b–d) STM images (8 × 8 nm*^{2} *in (b,c) and 10 × 10 nm*^{2} *in (d)) of trans-*Br_2I_2TPP *molecules on Au(111): after deposition (at 80 K, (b)), after heating to 120°C (c) and after further heating to 250°C (d). The corresponding chemical structures are indicated.*

In addition to the fundamental understanding of physical processes and chemical reactions of single molecules on a surface, the research group is interested in the linking of molecular building blocks on surfaces by covalent bonds, i.e. on-surface polymerization that is based on previous work by the group (*Nature Nanotech. 2, 687 (2007)*). An important issue in this regard is the precise location of the molecular activation process, i.e. the dissociation of Br substituents from the molecular building blocks on the surface. By comparing flat and stepped gold surfaces, the research group could precisely identify the kink sites at the step edges as the catalytically active sites because there is a high preference for activation for that side of the molecules that points towards these sites [3]. After heating the surface, the step edges create polymers that run parallel over the surface, which is of interest for a pre-alignment in future polymerization processes. Another study has focused on the complexity of the molecular structures produced by on-surface polymerization. All approaches so far have relied on a single step process, thus resulting in very simple structures. The research group has extended this method by introducing a hierarchical growth process based on a sequential activation of the molecular building blocks as sketched in Fig.1a [4]. The molecular building blocks (*trans*-Br_2I_2TPP molecules) exhibit two types of halogen substituents (bromine and iodine) that are dissociated from the molecular core at different temperatures, due to their characteristic binding energies. This results in a programmed reactivity in which in the first step, only the iodine atoms and in the second step the bromine atoms are cleaved from the molecule. Starting from the intact molecules (Fig.1b, the two halogen species appear at different height), this leads to polymer chains after the first step (Fig.1c). It is important to note that these polymers were linked exclusively at the former iodine sites, which is visible in the bright lobe, i.e. iodine, at the terminus and darker lobes, i.e. bromine, sideways in Fig.1c. After a second heating process, the polymer chains are connected sideways in a zipping mechanism, resulting in a two-dimensional network (Fig.1d). In addition to these homomolecular polymers, also copolymers could be formed by mixing two different building blocks and the resulting structure could not be formed in a conventional single-step linking process [4].

The research group has used the expertise in on-surface polymerization for the formation of graphene nanoribbons (following a recipe by R. Fasel and co-workers) on a Au(111) surface [5]. The objective was here not the polymerization process itself, but the characterization of charge transport through the molecular wires. By pulling individual graphene nanoribbons off the surface with the STM tip, the current decay along the polymer, which is the key property for the characterization of charge transport, can be determined. In particular, this was done for various electron energies, thus for the first time correlating the conductance of individual molecules with their electronic structure from the HOMO over the gap up to the LUMO. It was found that the charge transport is most efficient if the electron energy matches either the HOMO or the LUMO level, which are both delocalized along the ribbon [6].

[1] C. Gahl *et al.*, J. Am. Chem. Soc. **135**, 4273 (2013).

[2] P.-T. Chiang *et al.*, ACS Nano **6**, 592 (2012).

[3] A. Saywell, J. Schwarz, S. Hecht, and L. Grill, Angew. Chem. Int. Ed. **51**, 5096 (2012).

[4] L. Lafferentz *et al.*, Nature Chemistry **4**, 215 (2012).

[5] M. Koch, F. Ample, C. Joachim, and L. Grill, Nature Nanotech. **7**, 713 (2012).

2.2.2 Molecular Manipulation and Spectroscopy at the Nanoscale

Molecular processes and functions are fundamental in nature and also play a key role for molecular devices in future nanotechnology. Low-temperature scanning tunneling microscopy (LT-STM) permits not only to directly observe adsorbate dynamics at the single-molecule level, but also to precisely manipulate adsorbates and to control chemical reactions of single molecules by using chemical forces between the STM tip and adsorbates, the injection of tunneling electrons as well as the electric field in the junction. The research in the group of *Takashi Kumagai* focuses on the investigation of individual molecules and molecular ensembles using LT-STM and tip-enhanced Raman scattering.

Direct observation and control of single molecule dynamics: It is known that local environments of individual molecules have a significant impact on chemical processes in condensed phases via the deformation of the potential landscape. However, such local influences have rarely examined at the level of individual molecules in experiments and the effects of nearby single atoms or molecules on chemical reactions have not been studied so far.

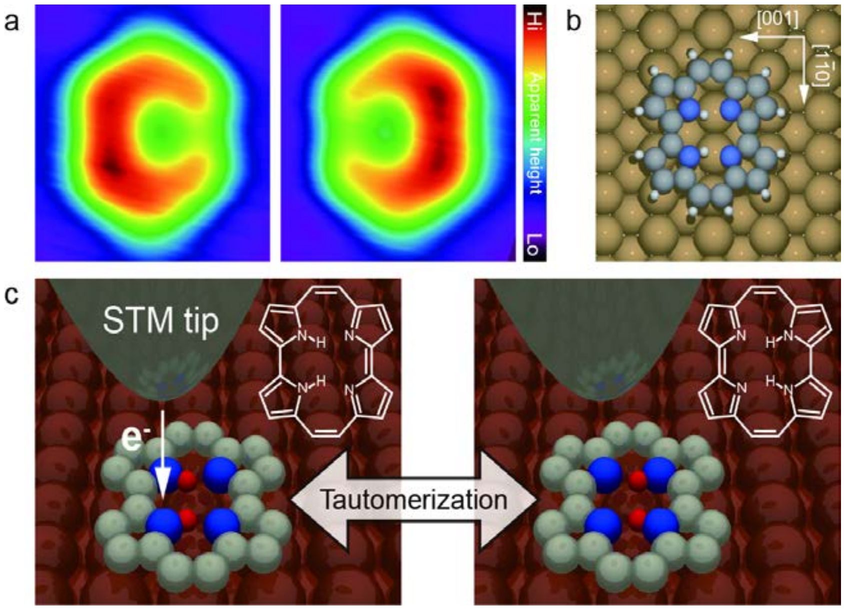


Figure 7: *Porphycene molecules on a Cu(110) surface (a) STM images of a single porphycene molecule. (b) The optimized structure determined by the density functional theory calculations. Porphycene molecule favors cis*

Takashi Kumagai and coworkers have achieved precise control of an intramolecular hydrogen transfer reaction (tautomerization) in single porphycene molecules adsorbed on Cu(110) [1]. Single porphycene molecules were imaged at 5 K and found to have a *cis* configuration in which the inner H-atoms are located on one side in the cavity (Fig.7a and b). Although the molecule is stationary at low bias voltages, the *cis*-*cis* tautomerization is induced at higher voltages and the STM image shows a flipping between the two states (Fig.7a and c). The efficiency of tautomerization depends on the lateral STM tip position with respect to a molecule, i.e., atomically precise location of the electron injection into the molecule, and shows maxima when the electron is injected over the position where the inner H-atoms exist. The tautomerization is also thermally induced at elevated temperatures and a barrier of 168±5 meV is determined from an Arrhenius plot. Furthermore, an isotope effect using deuterium-substituted porphycene in which the inner H-atoms are replaced by deuterium revealed that the STM-induced tautomerization is triggered by vibrational excitation via inelastic electron tunneling processes.

Remarkably, the probability for tautomerization can be precisely tuned by placing a single Cu adatom nearby a porphycene molecule. Cu adatoms are controlled by STM manipulation and the rate of tautomerization is significantly affected depending on the relative position between the adatom and molecule. The results demonstrate the high sensitivity of an elementary reaction to the exact position of individual atoms with respect to the molecule, surprisingly also at rather large distances much larger than a van der Waals radius [1].

The group extended this study to molecular assemblies and observed cooperative effects in the tautomerization process. Tautomerization is almost quenched in the dimer, but it becomes active in a specific molecules within larger clusters. It is revealed that even the hydrogen arrangement in the cavity of a neighboring molecule influences the tautomerization, causing positive and negative cooperativity. The results highlight the importance of local environments in the vicinity of individual molecules, and demonstrate the potential to regulate a single-molecule function. It is expected that this control over chemical reactions by subtle changes in the atomic-scale environment can be extended to other systems and will thus improve the understanding of fundamental molecular processes. It might even allow the tuning of molecular processes in functional nanostructures, which would pave the way towards information processing at the single-molecule level.

Tip-enhanced Raman spectroscopy: Tip-enhanced Raman spectroscopy (TERS) is one of the possible techniques to probe both adsorbate geometries and local vibrations, which provides fruitful insight into physical and chemical processes on surfaces. In TERS an STM tip is employed to generate a plasmonic field to enhance the Raman scattering of adsorbates as well as imaging their local structure with sub-molecular resolution. Within the last few years a TERS setup in ultra-high vacuum (UHV) has been developed to achieve a local spectroscopy at the single-molecule level [2]. Current experiments investigate graphene nano-ribbon (GNR) on a Au(111) surface, an attractive material in nano-science and technology. Local defects and alkali doping of GNR are expected to have a significant impact on its properties and the understanding of such influences is of importance for potential device applications.

[1] T. Kumagai *et al.* Phys. Rev. Lett. **111**, 246101 (2013); Nature Chemistry, (in press 2013);.

[2] B. Pettinger *et al.,* Ann. Rev. Phys. Chem. **63**, 379 (2012).

2.2.3 Real-time Observation of Photoinduced Surface Reactions

A longstanding dream has been to follow the dynamics of chemical reactions in real time and directly observe each elementary step. While for photoinduced reactions in the gas phase a remarkable level of sophistication has been reached, similar breakthroughs have not be achieved for heterogeneously catalyzed reactions at surfaces. Fundamental questions in surface reaction dynamics address how charge and energy is transferred between the adsorbates and the surface, or how the electronic structure is rearranged within the molecular unit during the elementary reaction steps. The real-time observation of chemical bond formation at surfaces requires techniques which are sensitive to the chemical state and simultaneously enable real-time probing of elementary steps with femtosecond time resolution.

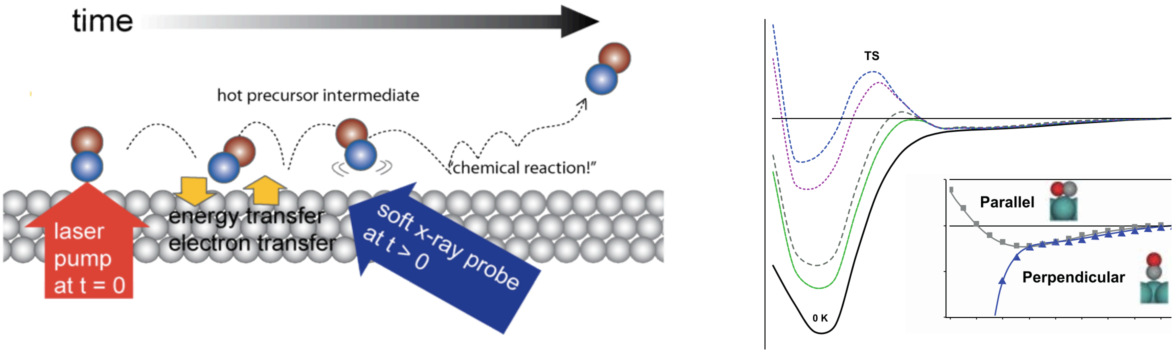


Figure 8: *(left) Schematic illustration of elementary steps in the desorption process of carbon monoxide. The reaction is stimulated by an ultrashort optical laser and probed with soft x-ray spectroscopy using femtosecond x-ray pulses. (right) Free energy diagram along the reaction pathway for CO on Ru(0001) exhibiting a chemisorption well and a shallow precursor which are separated by a temperature dependent barrier[2].*

Both x-ray absorption spectroscopy (XAS) and x-ray emission spectroscopy (XES) have the unique ability to provide an atom-specific probe of the electronic structure. With the advent of femtosecond x-ray lasers these techniques can now be transformed into time-resolved probes of transient chemical species at surfaces. For real-time probing of surface reactions, the chemical process must be initiated by a time-correlated ultrashort laser pulse, a concept which has been studied previously in the Department by the group of *Martin Wolf*: Femtosecond laser excitation of an adsorbate-covered metal surface can serve as a ultrafast “trigger” of surface reactions, whereby the non-adiabatic coupling between transiently excited metal electrons and adsorbate vibrational degrees of freedom mediates chemical processes such as associate desorption of the reactants [1].

Our collaboration with Anders Nilsson, and groups from Stanford, Stockholm, Hamburg and Berlin has performed a series of surface dynamics experiments, exploiting the unique capabilities of the Stanford x-ray free electron laser, LCLS, to probe the atom specific electronic structure changes during surface reactions induced by a strong laser pulse. Fig. 8 (left) schematically illustrates the basic principle of these experiments, exemplified for the simple case of desorption. The first experiment addressed desorption of CO from Ru(0001) induced by a 400 nm laser pulse [2]: By combining XAS and XES in the O 1s region both occupied and unoccupied states of the CO-metal bond ($5\sigma/1\pi$, d_π, $2\pi^*$ states) were probed as a function of time delay between the optical and x-ray laser pulses, demonstrating for the first time the feasibility of time-resolved resonant inelastic x-ray scattering (RIXS) in surface chemistry. Remarkably, a substantial transient weakening of the CO-Ru bond was observed, persisting for ~20 ps whereby ~30% of the *adsorbed* CO molecules resembled an electronic structure close to free CO. From DFT calculations including van-der Waals contributions of the free energy along the reaction coordinate (potential of mean force) a consistent picture could be obtained: The desorption dynamics of CO from Ru(0001) is governed by a transient energy landscape with a chemisorption well and a shallow precursor state, which are separated by a transient (entropic) barrier [2]. During desorption, a fraction of the vibrational excited CO molecules are transferred into the precursor state and are stabilized by the barrier prior to desorption (see Fig. 8 right). It should be noted that such insight into the dynamics of an excited (i.e. reacting) adlayer could not be revealed with established techniques like molecular beam scattering.

Further analysis demonstrated that by using resonant XES, selective excitation of different ensembles within the excited CO adlayer becomes possible, which exhibit different dynamical behavior and coupling to substrate electrons and phonons [3].

More recently, the technique of time-resolved RIXS has been successfully applied at LCLS to also study associative surface reactions such as ultrafast photoinduced CO_2 formation and hydrogenation of CO on ruthenium. Future planning includes the development of a dedicated surface science endstation with two XES spectrometers for molecular orientation sensitive detection and optimized sensitivity.

[1] H-J. Freund, G. Meijer, M. Scheffler, R. Schlögl and M. Wolf, Angew. Chem. **123**, 10242 (2011).

[2] M. Dell'Angela *et al.,* Science **339**, 1302 (2013).

[3] M. Beye *et al.*, Phys. Rev. Lett. **110**, 186101 (2013).

2.2.4 Interfacial Molecular Spectroscopy

The structure, reactivity and dynamics of molecules at interfaces are addressed by the group of *Kramer Campen* using the interface specific nonlinear optical technique of vibrational sum frequency (VSF) spectroscopy. Furthermore, vibrational and structural dynamics are directly probed by time-resolved infrared pump / VSF probe experiments. Over the last two years three systems have been studied: the air/water interface, CH_4 and C_2H_4 dissociation and reaction at the Ru(0001) surface in UHV, and water dissociation and reaction at the α-Al_2O_3(0001) surface both in UHV and under ambient conditions. For UHV studies a molecular beam source was developed to prepare translational and vibrational non-thermal distributions of impinging molecules, e.g. for dissociative adsorption. Under ambient conditions conventional VSF spectroscopy has been extended either by directly sampling structural dynamics, in the case of the air/water interface, or by probing low-frequency modes (e.g. the surface phonons of α-Al_2O_3) not previously observed.

Air/Water Interface: Prior VSF and simulation studies have shown that, from the H_2O molecule point of view, interfaces between liquid water and hydrophobic surfaces have a large population of water molecules with one non-hydrogen bonded (free) OH groups. Recent theoretical work has made clear that quantitative understanding of hydrophobic solvation requires an understanding of the ultrafast structural dynamics of these free OH groups [1]. We have recently applied both experiment, i.e. time resolved VSF spectroscopy (in collaboration with M. Bonn, MPI for Polymer Science), and simulation (in collaboration with A. Vila Verde, MPI of Colloids and Interfaces), to understand the dynamics of the free OH. Taken together, this work suggests: (1) The free OH rotates 3x faster than hydrogen bonded OH groups either at the air/water interface or in bulk water [2, 3]. (2) The free OH is structurally heterogeneous on picosecond timescales: free OH groups closer to the vapor have a different orientational distribution and persist longer before rotating down towards the liquid than free OH groups closer to bulk water [2-4]. (3) Relaxation of vibrationally excited free OH groups proceeds by a combination of energy transfer between the free and hydrogen bonded OH within a single water molecule (2/3 of relaxation occurs via this pathway) and structural relaxation (1/3) in which the excited free OH rotates towards the liquid and forms a hydrogen bond [5].

Methane and Ethylene at Ru(0001): Interaction of CH_4 and C_2H_4 with metal (oxide) surfaces may lead to their decomposition and the formation of higher hydrocarbons. To study this chemistry we have characterized the interaction of CH_4 and C_2H_4 with the Ru(0001) surface in UHV using temperature programmed desorption (TPD) and VSF spectroscopy as a function of sample temperature and carbon coverage. To overcome the dissociation barrier of CH_4 a molecular beam source was employed with CH_4. By probing the CH spectral response (2800-3100 cm^{-1}) during the decomposition of both CH_4 and C_2H_4 we have tracked the relative stability of all one and two carbon, CH containing species. We find that both the relative stabilities and rates of interconversion of the various hydrocarbon species present are strongly depending on surface coverage and temperature (Fig. 9 left).

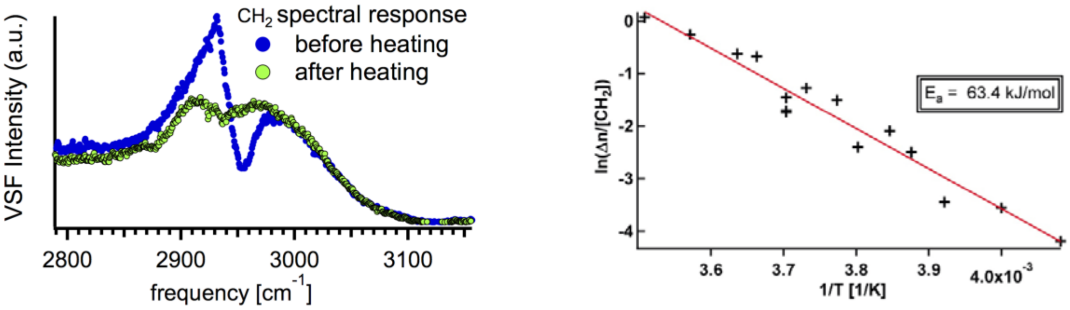


Figure 9: *Left: Measured VSF spectra showing the decrease in CH_2 spectral amplitude on heating above 350 K. Right: Change in CH_2 resonance amplitudes plotted as a function of sample heating temperature on an Arrhenius plot illustrating CH_2 to CH conversion. In the low coverage limit the E_a for CH_2 dehydrogenation is 14 kJ/mol.*

A dramatic change in reactivity was observed above 350 K. Both computation (in collaboration with S. Levchenko, Theory Department) and experiment clarify that below 350 K adsorbed hydrogen blocks energetically favorable surface sites and that above 350 K recombinative desorption of H_2 sets in. This coverage dependence of hydrocarbon reactivity is manifested by the thermal stability of CCH_2/CCH species and the thermal activation for the conversion of CH_2 to CH (Fig. 9 right).

Water Dissociation and Surface Reconstruction at α-Al_2O_3(0001): Most properties of Al_2O_3 surfaces change dramatically on exposure to even submonolayer concentrations of water. To gain insight into the mechanism(s) of such change, we here probe the elementary steps of single molecule water dissociative adsorption on α-Al_2O_3(0001) and the changes in surface structure they induce. Water adsorption was studied by preparing a well defined α-Al_2O_3(0001) surface in UHV and then dosing this surface, using the molecular beam source, with D_2O seeded in He. Next the spectral response of the frequency range corresponding to dissociated water molecules was characterized as a function of sample temperature. This data show five resonances whose relative frequencies, and intensities as a function of experimental geometry, are consistent with computation (collaboration with P. Saalfrank, University of Potsdam). Based on this agreement, these five modes are assigned to fragments resulting from three different predicted water dissociative adsorption channels. By tracking the thermal stabilities of these fragments, it was demonstrated that models of water surface reactivity must explicitly account for surface coverage effects.

While the water OD stretch is a useful probe of dissociative adsorption, it tells us little about concurrent surface reconstruction. To probe surface reconstruction both within and outside UHV we have extended VSF spectroscopy to infrared frequencies as low as 750 cm^{-1} to probe also α-Al_2O_3 surface phonons. As expected, the amplitudes/line shapes of these modes are dramatically changed by treatments known to dehydrate/rehydrate the surface. Normal mode calculation confirms our assignment and allows a full microscopic description of each mode. This work demonstrates that surface reconstruction outside of UHV can be tracked by optical probing of surface phonons.

Electron solvation at interfaces: The reactivity of excess electrons in aqueous environments is highly relevant in various disciplines ranging from atmospheric chemistry to photosynthesis. The group of *Julia Stähler* has studied excess electrons at ice-vacuum interfaces created by photoexcitation of the metal template and also by low-energy electron impact. Their relaxation and lifetime of several seconds was determined using two-photon photoelectron (2PPE) spectroscopy. In addition to pure charging, permanent changes to the surface dipole and therefore work function (up to 1 eV) were also observed, which may result from a build-up of OH^- induced by the trapped electrons. In combination with simple model calculations, these work function modifications were also used to identify the impact of work function distributions on photoelectron experiments [6].

[1] D. Chandler, *Nature*, **437**, 640 (2005).

[2] C. S. Hsieh *et al.*, *Phys. Rev. Lett.*, **107**, 116102 (2011).

[3] A. Vila Verde, P. G. Bolhuis, R. K. Campen, *J. Phys. Chem. B*, **116**, 9467 (2012).

[4] Y. Tong, A. Vila Verde, R. K. Campen, *J. Phys. Chem. B*, **117**, 11753 (2013).

[5] C. S. Hsieh, R. K. Campen, M. Okuno, E. H. G. Backus, Y. Nagata, M. Bonn, *PNAS* (in press 2013).

[6] D. Wegkamp, M. Meyer, C. Richter, M. Wolf, and J. Stähler, *Appl. Phys. Lett.* **103**, 151603 (2013).

2.2.5 Computational Dynamics of Protein Machines

The group of *Alexander Mikhailov* performs computational studies of biomolecular systems. Protein machines play a fundamental role in biological cells. Operating as molecular motors, they transport load over filaments and microtubules or generate mechanical force. They can perform operations with other molecules, such as DNA or RNA, and cut, glue or unwind them. Protein machines can act as enzymes, facilitating reaction events, or as pumps transporting ions across lipid bilayers. In all their functions, the operation of such molecular devices is based on the ability of proteins to fold into a definite conformation and to perform ordered mechanochemical motions, induced by binding and detachment of ligands or by chemical reactions with them. All machines require energy for their operation and this energy is usually provided through ATP molecules. Understanding the dynamics of protein machines is essential in biophysics of a cell; it can also open a way for engineering of synthetic molecular devices with similar properties.

The cycles of protein machines involve slow conformational motions on the scales of milliseconds or longer. Therefore, they cannot be reproduced in current full molecular dynamics simulations and coarse-grained descriptions are required. One such description consists in modeling a protein as an elastic networks (EN) formed by particles (amino acids) with effective elastic interactions between them. EN models are popular and often used in the context of normal-mode analysis. A special feature of the investigations in the group of *Alexander Mikhailov* is that complex nonlinear dynamics is considered in the EN models of various real protein machines through numerical simulations. The research is undertaken in cooperation with partners in Japan, Canada, Belgium and Taiwan.

Myosin is the molecular motor responsible for muscle contraction and for transport along actin filaments in biological cells. It has been extensively investigated in the group of T. Yanagida, Osaka University, and at RIKEN Quantitative Biology Center in Japan. Their recent single-molecule experiments have shown that this protein acts as a “strain sensor”, so that its affinity towards the actin filament is strongly modulated by the applied external forces; this behavior is of principal importance for the motor function. Numerical analysis of responses of myosin molecules to external forces could not only confirm the experimental results, but also disclose the nature of the strain-sensor behavior in this macromolecule [1].

Actin is a structural protein able to form, through polymerization, long filaments which build the skeleton of a cell and are also used for intracellular transport by myosin motors. The filaments are permanently growing at one end and dissolving at the other end through the process of “treadmilling”. While it was known that ATP is needed for filament growth, its role in the polymerization process remained unclear. The EN simulations have revealed that, when ATP binds to an actin monomer, this stabilizes its closed conformation already present as a metastable conformational state in the ATP-free molecules. Such closed conformation much better fits the growing filament end and thus the polymerization rate is greatly enhanced. The metastable conformational states of actin were previously detected in single-molecule FRET experiments by T. Yanagida with coworkers.

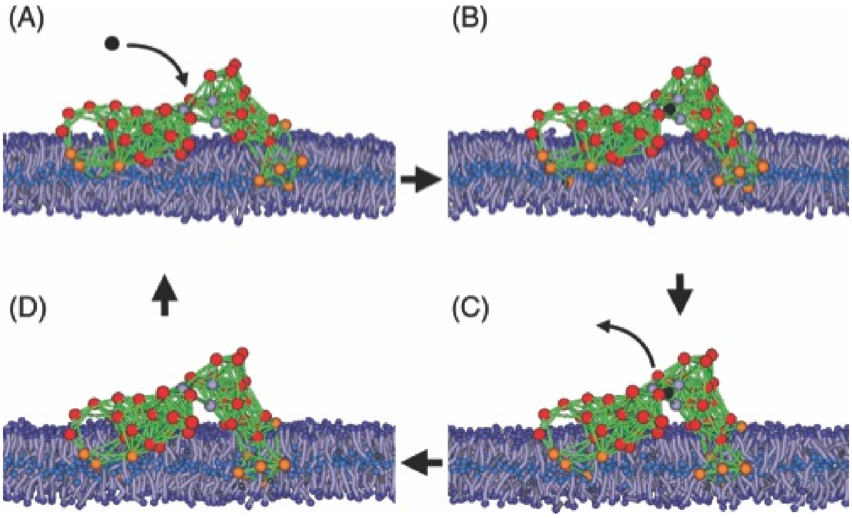


Figure 10: *The cycle of a membrane machine. (A) The ligand binds to the machine; (B) the machine conformation changes from the open state to the closed state; (C) the reaction takes place and the ligand is released; (D) the machine returns to its open state.*

Many protein machines operate as active inclusions in lipid bilayers forming *biological membranes*. In such cases, additional complications arise because of the necessity to incorporate lipids and the solvent into a model. Together with R. Kapral, Toronto University, and researchers from the National Central University in Taiwan, fast and efficient methods for such simulations, combining the EN description for proteins with a coarse-grained description for lipids and the multiparticle collision dynamics for the solvent, were developed [2,3]. Fig.10 shows a cycle of a model protein machine attached to a lipid bilayer. An important result is that strong hydrodynamic flows in the lipid bilayer, induced by operating machines, were found, indicating that hydrodynamic effects should play a principal role in interactions between active membrane inclusions. The swimming behavior of active machines in biomembranes has been further discussed [4].

Furthermore, a statistical analysis based on the NMR data for conformational ensembles in a set of 1500 different proteins was analyzed and used improvement of the accuracy of the EN method (with Y. Dehouck, ULB in Belgium). In addition to the work on biomolecular systems the group has also investigated complex dynamics in networks.

[1] M. Düttmann, Y. Togashi, T. Yanagida, A. S. Mikhailov, *Biophys. J.* **102**, 542 (2012).

[2] M.-J. Huang, R. Kapral, A. S. Mikhailov, H.-Y. Chen, *J. Chem. Phys.* **137**, 055101 (2012).

[3] M.-J. Huang, R. Kapral, A. S. Mikhailov, H.-Y. Chen, *J. Chem. Phys.* **138**, 195101 (2013).

[4] M.-J. Huang, H.-Y. Chen, A. S. Mikhailov, , *Eur. Phys. J. E* **35**, 119 (2012).

2.2.6 Electrochemical Dynamics

The co-existence of distinct timescales is an important feature in most natural and man-made systems exhibiting kinetic instabilities and oscillatory behavior. The group of *Markus Eiswirth* has concentrated on different aspects of non-linear dynamics in electrochemical systems from both experimental and theoretical points of view. For electrochemical reactions taking place at the solid/liquid interface, experimentally recorded time-series are often subjected to a long-term surface deactivation process that acts as a slowly evolving bifurcation parameter. Recently, mechanistic aspects associated to this process during the catalytic electro-oxidation of small organic molecules have been investigated. Experiments and numerical simulations were carried out in cooperation with Hamilton Varela, University of Sao Paulo at Sao Carlos, Brazil, and Jaeyoung Lee, Ertl Center for Electrochemistry and Catalysis, Gwangju, Korea).

Fig. 11 shows results for the electro-oxidation of formaldehyde on platinum in terms of the time-trace of the electrode potential, *U*, obtained under a slow galvanodynamic sweep [1]. The deliberate increase of the applied current mimics the spontaneous and slow poisoning process, also observed in many other systems. Therefore, the oscillatory patterns depicted in (c - h) and monitored at different applied currents appear spontaneously after setting a given fixed current. For a certain kind of oscillation or waveform, the mean electrode potential increases in time (Fig. 11 a). The system thus consists of two parts: the core oscillator, associated to the main dynamics or the oscillations themselves, and a slowly evolving drift.

The coupled system is characterized by the co-existence of two disparate time-scales. After each cycle, the surface would ideally return to its original state and, under those conditions, oscillations would persist as long as the reaction proceeds. Conversely, the slowly evolving parameter may arise from the fact that the surface is not completely restored to its initial conditions after one oscillatory cycle. Since the drift in acidic media is always accompanied by an increase in the mean electrode potential, it is likely that the surface is slowly getting oxidized after each cycle.

Based on the above conjectures and also on *in situ* infrared and *online* mass spectrometry, one can describe the interplay between slow and fast processes in terms of the surface coverage of adsorbates. The main aspects are summarized in the reaction scheme in Fig. 11 (bottom). Generally speaking, the electro-oxidation of organic molecules on platinum proceeds via parallel pathways: the direct pathway, with an active intermediate (adsorbed formate in this scheme) transforming to carbon dioxide relatively fast, and the indirect route, where adsorbed carbon monoxide is oxidized at comparatively high overpotentials in a Langmuir-

Hinshelwood step with adsorbed oxygenated species. Steps 1 to 6 in this scheme are associated to oscillations and thus belong to the core subsystem

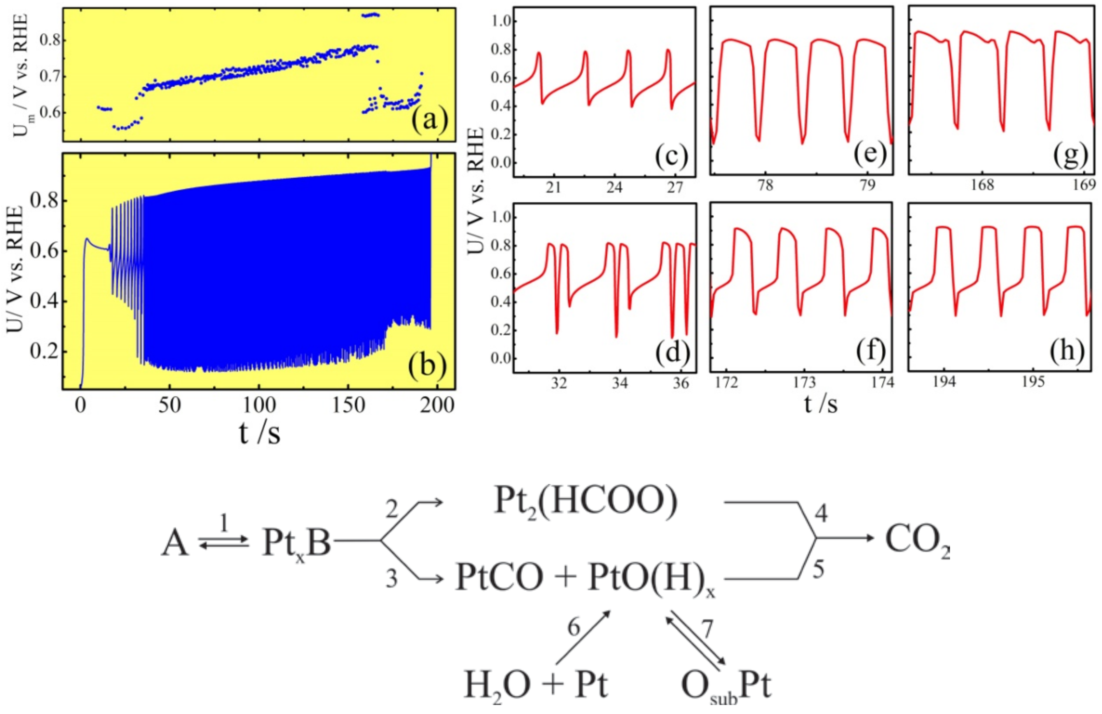


Figure 11: *(top) Time-traces of (a) the mean electrode potential, and of (b) the electrode potential, during the galvanodynamic (10.42 µA cm*$^{-2}$ *s*$^{-1}$*) electro-oxidation of formaldehyde on platinum (c-h expanded time axis). Electrolyte: 0.5 M* H_2SO_4 *aqueous solution with 0.1 mol L*$^{-1}$ *of HCHO. (Bottom): Reaction scheme of electro-oxidation of methanol and formaldehyde on platinum [1].*

The adsorbed species $PtO(H)_x$ represents a generic oxygenated adsorbate that participates in the oxidation of species such as adsorbed carbon monoxide. Further oxidation of the platinum surface leads to the so-called place-exchange process, in which surface oxygen atoms are inserted in the platinum lattice in order to allow further surface oxidation to take place. Sub-surface oxygen is in principle unavailable for Langmuir-Hinshelwood steps such as the illustrated electro-oxidation of adsorbed carbon monoxide. As a consequence, the increase in the amount of O_{sub}-Pt accompanying the increase in the electrode potential is equivalent to a decrease in the overall number of surface sites, which in turn, causes an increase in the actual current density (in contrast to the applied current, which is strictly constant). Therefore, coupled to the core oscillator, the spontaneous drift that slowly evolves in time is suggested to result from the increase of sub-surface oxygen.

After assigning the slowly evolving parameter as the coverage of a surface-blocking species, incorporated this process has been incorporated in a generic model for this family of oscillators. The resulting model consists of four ordinary differential equations, and it was investigated over a wide parameter range. Besides the bifurcation analysis, the system was studied by means of high-resolution period and Lyapunov diagrams. It was observed that the system's dynamics becomes simpler as the irreversible poisoning evolves, as evidenced by the changes in the structure of the bifurcation diagram. Nevertheless, periodic cascades are preserved in a confined region of the resistance *vs.* potential diagram.

[1] M. F. Cabral, R. Nagao, E. Sitta, M. Eiswirth, H. Varela, *Phys. Chem. Chem. Phys.* **15**, 1437 (2013).

3. Research Projects Funded from Outside Resources

Kramer Campen and Martin Wolf:

- DFG Collaborative Research Center SFB 658 “Elementary processes of molecular switches at surfaces”, project B9 ***“Molecular switching in self-assembled monolayers at liquid-solid interfaces”*** (selfassembled monolayers, liquid-solid interfaces, static and time-resolved vibrational SFG spectroscopy, from 7/2013)

Kramer Campen:

- DFG Collaborative Research Center 1109 “Understanding of Metal Oxide/Water Systems at the Molecular Scale: Structural Evolution, Interfaces and Dissolution”, Project B1, **“*Understanding water structure and reactivity at aluminum oxide surfaces using nonlinear vibrational spectroscopy and theory*”** (oxide/water interfaces, time resolved vibrational SFG spectroscopy, *ab initio* dynamics and thermodynamics, together with P. Saalfrank, Univ. Potsdam, from 4/2014)

Ralph Ernstorfer and Martin Wolf:

- DFG Reseach Unit FOR1700 (Wo 653/8-1) “Metallic nanowires on the atomic scale: Electronic and vibrational coupling in real world systems”, Project E5 ***“Time-resolved spectroscopy of photoinduced transitions and electronic excitations in quasi-1D metal wires on semiconductors”*** (electronic structure and dynamics in quasi 1D systems, time-resolved ARPES, from 11/2012)

Leonhard Grill:

- European research project (ICT FET) ***“Alternative routes towards information storage and transport at the atomic and molecular scale (ARTIST)”*** (single molecule manipulation and spectroscopy with low-temperature STM, until 7/2013)
- European research project (ICT FET) ***“Atomic scale and single molecule logic gate technologies (AtMol)”*** (formation and characterization of molecular wires with low-temperature STM, until 12/2014)
- Marie Curie Initial Training Networks (multipartner ITN) of the European Union “Actuation and characterization at the single bond limit” (ACRITAS) (Jan – July 2013, now transferred to University of Graz)
- DFG Collaborative Research Center SFB 951 “Hybrid Inorganic/Organic Systems for Opto-Electronics”, project A2 ***“Assembly and local probing of single molecules on ultrathin ZnO films on metals”*** (deposition and imaging of single molecules on utrathin

oxide films with STM, until 6/2015, project will be continued by Takashi Kumagai and Martin Wolf)

Leonhard Grill and Martin Wolf:

- Collaborative Research Center SFB 658 "Elementary Processes of Molecular Switches at Surfaces", project A5 ***"Complex switching units and molecular architectures, studied by scanning tunneling microscopy"*** (nanostructuring, single molecule spectroscopy and manipulation with low-temperature STM, until 6/2013)

Karsten Horn:

- Individual project (Ho 797/18-1) ***"Graphene-based systems for spintronics: Magnetic interactions at the graphene/3d metal interface (SpinGraph)"*** within the program "EuroGRAPHENE" of the European Science Foundation
- DFG Priority Program SPP 1459 (De 1679/3-1) "Graphene", project ***"Graphene: electronic structure, transport and functionalization"*** (growth and doping of eptaxial grapheme layers, angle-resolved photoemission spectroscopy)

Tobias Kampfrath:

- DFG individual research grant (KA 3305/2-1), ***"Femtosecond coherent control of terahertz radiation by transient nanophotonic structures"***, (ultrafast optical spectroscopy, THz photonics, from 02/2013)
- DFG priority program SPP 1666 (KA 3305/3-1), "Topological Insulators", project ***"Investigation of directional THz spin currents in topological surface states"*** (ultrafast spin dynamics in topological insulators, THz emission spectroscopy, together with C. Heiliger, Univ. Gießen and M. Münzenberg, Univ. Göttingen, from 06/2013)

Alexey Melnikov:

- DFG individual research grant (ME 3570/1-3), ***"Ballistic electron-driven magnetization dynamics induced by femtosecond laser excitation"*** (ultrafast spin dynamics and transport, epitaxial metallic multilayer films, non-linear optical SHG spectroscopy, from 10/2013)

Alexander Mikhailov:

- DFG Collaborative Research Center SFB 910 "Control of Self-Organizing Nonlinear Systems", project A6 ***"Control of self-organization in dynamical networks"*** (control of nonlinear dynamics in chemical and biological networks)

- Volkswagen Foundation, international project ***"Self-organizing networks of interacting machines"*** (design and analysis of the networks of molecular machines)
- DFG Research Training Group GRK 1558 "Nonequilibrium Collective Dynamics in Condensed Matter and Biological Systems", project ***"Active microfluidics based on floating molecular machines"*** (molecular machines at liquid interfaces)

Alexander Saywell:

- DFG individual research grant "eigene Stelle" (Sa 2625/1-1), ***"Force induced dissociation of individual molecules"*** (Single molecule spectroscopy and manipulation, low-temperature atomic force microscopy (AFM), from 1/2014)

Julia Stähler and Martin Wolf:

- Collaborative Research Center SFB 951 "Hybrid Inorganic/Organic Systems for Opto-Electronics", project B9 ***"Electronic structure and ultrafast carrier dynamics at hybrid inorganic/organic interfaces"*** (time-resolved 2PPE and non-linear optical spectroscopy, organic/inorganic semiconductor interfaces)

Julia Stähler, Tobias Kampfrath, Ralph Ernstorfer and Martin Wolf:

- European Research Project (*NMP-2011-SMALL*): ***"Time dynamics and Control in nanostructures for magnetic recording and energy applications (CRONOS)"***, project FHIB, (time-resolved 2PPE and non-linear optical spectroscopy, magneto-optical Kerr spectroscopy, THz emission, high-harmonic generation, from 6/2012)

Martin Wolf:

- German-Israel Foundation (GIF), cooperation with Prof. Micha Asscher, Hebrew University of Jerusalem, project ***"Photochemistry and electron dynamics of oriented molecules within a nano-capacitor"*** (photochemistry and charge transfer processes in thin molecular films, until 3/2013)

4. Publications of the Department of Physical Chemistry

2011 (late publications)

Cortes Rodriguez, R., L. Rettig, Y. Yoshida, H. Eisaki, M. Wolf and U. Bovensiepen: Momentum-Resolved Ultrafast Electron Dynamics in Superconducting $Bi_2Sr_2CaCu_2O_{8+\delta}$. Physical Review Letters **107** (9), 097002 (2011).

Cossaro, A., M. Puppin, D. Cvetko, G. Kladnik, A. Verdini, M. Coreno, M. de Simone, L. Floreano and A. Morgante: Tailoring SAM-on-SAM Formation. The Journal of Physical Chemistry Letters **2** (24), 3124-3129 (2011).

Flechsig, H., D. Popp and A.S. Mikhailov: In Silico Investigation of Conformational Motions in Superfamily 2 Helicase Proteins. PLoS One **6** (7), e21809 (2011).

Först, M., R.I. Tobey, S. Wall, H. Bromberger, V. Khanna, A.L. Cavalieri, Y.-D. Chuang, W.S. Lee, R. Moore, W.F. Schlotter, J.J. Turner, O. Krupin, M. Trigo, H. Zheng, J.F. Mitchell, S.S. Dhesi, J.P. Hill and A. Cavalleri: Driving magnetic order in a manganite by ultrafast lattice excitation. Physical Review B **84** (24), 241104 (2011).

Freund, H.-J., G. Meijer, M. Scheffler, R. Schlögl and M. Wolf: CO oxidation as a prototypical reaction for heterogeneous processes. Angewandte Chemie International Edition **50** (43), 10064-10094 (2011).

Freund, H.-J., G. Meijer, M. Scheffler, R. Schlögl and M. Wolf: Die CO-Oxidation als Modellreaktion für heterogene Prozesse. Angewandte Chemie **123** (43), 10242-10275 (2011).

Katayama, I., S. Koga, K.-I. Shudo, J. Takeda, T. Shimada, A. Kubo, S. Hishita, D. Fujita and M. Kitajima: Ultrafast Dynamics of Surface-Enhanced Raman Scattering Due to Au Nanostructures. Nano Letters **11** (7), 2648-2654 (2011).

Kobayashi, Y., T. Shibata, Y. Kuramoto and A.S. Mikhailov: Robust network clocks: Design of genetic oscillators as a complex combinatorial optimization problem. Physical Review E **83** (6), 060901(R) (2011).

Lee, J.K., H.-H. Ann, Y. Yi, K.W. Lee, S. Uhm and J. Lee: A stable Ni–B catalyst in hydrogen generation via $NaBH_4$ hydrolysis. Catalysis Communications **16** (1), 120-123 (2011).

Mota Lima, A.B., E.R. Gonzalez and M. Eiswirth: Kinetic Insights over a PEMFC Operating on Stationary and Oscillatory States. The Journal of Physical Chemistry A **115** (47), 13773-13782 (2011).

Mota Lima, A.B., D.H. Salmazo, H. Varela, E.R. Gonzalez and M. Eiswirth: Eletro-oxidação oscilatória de ácido fórmico sobre paládio uma variação cinética e mecanística. In: Proceedings Simposio Brasileiro de Eletroquimica e Eletroanalítica XVIII, 313-316 (2011).

Okamoto, H., K. Imura, T. Shimada and M. Kitajima: Spatial distribution of enhanced optical fields in monolayered assemblies of metal nanoparticles: Effects of interparticle coupling. Journal of Photochemistry and Photobiology A **221** (2-3), 154-159 (2011).

Rooman, M., J. Albert, Y. Dehouck and A. Haye: Detection of Perturbation Phases and Developmental Stages in Organisms from DNA Microarray Time Series Data. PLoS One **6** (12), e27948 (2011).

Scheuch, M., T. Kampfrath, M. Wolf, K. von Volkmann, C. Frischkorn and L. Perfetti: Temperature dependence of ultrafast phonon dynamics in graphite. Applied Physics Letters **99** (21), 211908 (2011).

Wegkamp, D., D. Brida, S. Bonora, G. Cerullo, J. Stähler, M. Wolf and S. Wall: Phase retrieval and compression of low-power white-light pulses. Applied Physics Letters **99** (10), 101101 (2011).

Doctoral Thesis

Flechsig, H.: Structurally Resolved Coarse-Grained Modeling of Motor Protein Dynamics. Technische Universität Berlin 2011.

Meyer, M.: Ultrafast Electron Dynamics at Alkali/Ice Structures Adsorbed on a Metal Surface. Freie Universität Berlin 2011.

2012

Arendt, P.: Nearly Unattenuated Charge Density Wave Transport in a Linear Chain Conductor Fabricated by Spark Discharge. Zeitschrift für Naturforschung A **67**, 29-38 (2012).

Beggs, D.M., T.F. Krauss, L. Kuipers and T. Kampfrath: Ultrafast Tilting of the Dispersion of a Photonic Crystal and Adiabatic Spectral Compression of Light Pulses. Physical Review Letters **108** (3), 033902 (2012).

Beggs, D.M., I.H. Rey, T. Kampfrath, N. Rotenberg, L. Kuipers and T.F. Krauss: Ultrafast Tunable Optical Delay Line Based on Indirect Photonic Transitions. Physical Review Letters **108** (21), 213901 (2012).

Bovensiepen, U. and P.S. Kirchmann: Elementary relaxation processes investigated by femtosecond photoelectron spectroscopy of two-dimensional materials. Laser & Photonics Reviews **6** (5), 589-606 (2012).

Bovensiepen, U., H. Petek and M. Wolf: Dynamics at Solid State Surfaces and Interfaces, Vol. 2: Fundamentals. Wiley-VCH, Weinheim (2012). ISBN 978-3-527-40924-2.

Chiang, P.-T., J. Mielke, J. Godoy, J.M. Guerrero, L.B. Alemany, C.J. Villagómez Ojeda, A. Saywell, L. Grill and J.M. Tour: Toward a Light-Driven Motorized Nanocar: Synthesis and Initial Imaging of Single Molecules. ACS Nano **6** (1), 592-597 (2012).

Darwish, T.A., Y. Tong, M. James, T.L. Hanley, Q. Peng and S. Ye: Characterizing the Photoinduced Switching Process of a Nitrospiropyran Self-Assembled Monolayer Using In Situ Sum Frequency Generation Spectroscopy. Langmuir **28** (39), 13852-13860 (2012).

Dedkov, Y.S., K. Horn, A. Preobrajenski and M. Fonin: Epitaxial graphene on metals. In: Graphene Nanoelectronics: Metrology, Synthesis, Properties and Applications, (Ed.) H. Raza. Springer, Heidelberg, 189-234 (2012). ISBN 978-3-642-20467-8.

Doblhofer, K., G.K. Weil: Application of the Quartz Microbalance in Electrochemistry. In: Methods in Physical Chemistry, Vol. 1. (Eds.) R. Schäfer and P.C. Schmidt. Wiley-VCH, Weinheim, 575-601 (2012). ISBN 978-3-527-3274-54.

Düttmann, M., M. Mittnenzweig, Y. Togashi, T. Yanagida and A.S. Mikhailov: Complex Intramolecular Mechanics of G-actin — An Elastic Network Study. PLoS One **7** (10), e45859 (2012).

Düttmann, M., Y. Togashi, T. Yanagida and A.S. Mikhailov: Myosin-V as a Mechanical Sensor: An Elastic Network Study. Biophysical Journal **102** (3), 542-551 (2012).

Ertl, G.: The Arduous Way to the Haber–Bosch Process. Zeitschrift für anorganische und allgemeine Chemie **638** (3-4), 487-489 (2012).

Ertl, G. and H.-J. Lewerenz: Obituary: Dieter M. Kolb (1942–2011). ChemPhysChem **13** (12), 2891-2891 (2012).

Fauster, T., H. Petek and M. Wolf: Surface States and Adsorbate-Induced Electronic Structure. In: Dynamics at Solid State Surfaces and Interfaces, Vol. 2. (Eds.) U. Bovensiepen, H. Petek and M. Wolf. Wiley-VCH, Weinheim, 115-154 (2012). ISBN 978-3-527-40924-2.

Föhlisch, A., S. Vijayalakshmi, A. Pietzsch, M. Nagasono, W. Wurth, P.S. Kirchmann, P.A. Loukakos, U. Bovensiepen, M. Wolf, M. Tchaplyguine and F. Hennies: Charge transfer dynamics in molecular solids and adsorbates driven by local and non-local excitations. Surface science **606** (11-12), 881-885 (2012).

Generalov, A.V. and Y.S. Dedkov: EELS study of the epitaxial graphene/Ni(1 1 1) and graphene/Au/Ni(1 1 1) systems. Carbon **50** (1), 183-191 (2012).

Gille, M., L. Grill and S. Hecht: Bottom-up zu molekularen Nanostrukturen. Nachrichten aus der Chemie **60** (10), 986-990 (2012).

Gonnelli, G., M. Rooman and Y. Dehouck: Structure-based mutant stability predictions on proteins of unknown structure. Journal of Biotechnology **161** (3), 287-293 (2012).

Hata, S., H. Nakao and A.S. Mikhailov: Global feedback control of Turing patterns in network-organized activator-inhibitor systems. Europhysics letters **98** (6), 64004 (2012).

Huang, M.-J., H.-Y. Chen and A.S. Mikhailov: Nano-swimmers in biological membranes and propulsion hydrodynamics in two dimensions. The European Physical Journal E **35** (11), 119 (2012).

Huang, M.-J., R. Kapral, A.S. Mikhailov and H.-Y. Chen: Coarse-grain model for lipid bilayer self-assembly and dynamics: Multiparticle collision description of the solvent. The Journal of Chemical Physics **137** (5), 055101 (2012).

Kahra, S., G. Leschhorn, M. Kowalewski, A. Schiffrin, E. Botschafter, W. Fuß, R. de Vivie-Riedle, R. Ernstorfer, F. Krausz, R. Kienberger and T. Schaetz: A molecular conveyor belt by controlled delivery of single molecules into ultrashort laser pulses. Nature Physics **8** (3), 238-242 (2012).

Kaluza, P.F. and T. Cioaca: Phase oscillator neural network as artificial central pattern generator for robots. Neurocomputing **97**, 115-124 (2012).

Kaluza, P.F. and A.S. Mikhailov: Design of robust flow processing networks with time-programmed responses. The European Physical Journal B **85** (4), 129 (2012).

Koch, M., F. Ample, C. Joachim and L. Grill: Voltage-dependent conductance of a single graphene nanoribbon. Nature Nanotechnology **7** (11), 713-717 (2012).

Kouvaris, N., H. Kori and A.S. Mikhailov: Traveling and Pinned Fronts in Bistable Reaction-Diffusion Systems on Networks. PLoS One **7** (9), e45029 (2012).

Kummer, K., D.V. Vyalikh, L. Rettig, R. Cortés, Y. Kucherenko, C. Krellner, C. Geibel, U. Bovensiepen, M. Wolf and S.L. Molodtsov: Ultrafast quasiparticle dynamics in the heavy-fermion compound $YbRh_2Si_2$. Physical Review B **86** (8), 085139 (2012).

Lafferentz, L., V. Eberhardt, C. Dri, C. Africh, G. Comelli, F. Esch, S. Hecht and L. Grill: Controlling on-surface polymerization by hierarchical and substrate-directed growth. Nature Chemistry **4** (3), 215-220 (2012).

Lee, J., H. Ju, Y. Yi, J. Lee, S. Uhm, J.K. Lee and H.J. Lee: High-Density Nanoporous Structures for Enhanced Electrocatalysis. The Journal of Physical Chemistry C **116** (4), 2915-2918 (2012).

Lublow, M., S. Kubala, J.-F. Veyan and Y.J. Chabal: Colored porous silicon as support for plasmonic nanoparticles. Journal of Applied Physcis **111** (8), 084302 (2012).

Martínez-Blanco, J., A. Mascaraque, Y.S. Dedkov and K. Horn: Ge(001) As a Template for Long-Range Assembly of π-Stacked Coronene Rows. Langmuir **28** (8), 3840-3844 (2012).

Mielke, J., S. Selvanathan, M. Peters, J. Schwarz, S. Hecht and L. Grill: Molecules with multiple switching units on a Au(111) surface: self-organization and single-molecule manipulation. Journal of Physics: Condensed Matter **24** (39), 394013 (2012).

Nacci, C., K. Kanisawa and S. Fölsch: Reversible switching of single tin phthalocyanine molecules on the InAs(111)A surface. Journal of Physics: Condensed Matter **24** (39), 394004 (2012).

Nayak, J., M. Maniraj, A. Rai, S. Singh, P. Rajput, A. Gloskovskii, J. Zegenhagen, D.L. Schlagel, T.A. Lograsso, K. Horn and S.R. Barman: Bulk Electronic Structure of Quasicrystals. Physical Review Letters **109** (21), 216403 (2012).

Paarmann, A., M. Gulde, M. Müller, S. Schäfer, S. Schweda, M. Maiti, C. Xu, T. Hohage, F. Schenk, C. Ropers and R. Ernstorfer: Coherent femtosecond low-energy single-electron pulses for time-resolved diffraction and imaging: A numerical study. Journal of Applied Physcis **112** (11), 113109 (2012).

Pettinger, B., P. Schambach, C.J. Villagomez Ojeda and N. Scott: Tip-Enhanced Raman Spectroscopy: Near-Fields Acting on a Few Molecules. Annual Review of Physical Chemistry **63**, 379-399 (2012).

Rettig, L., R. Cortes, S. Thirupathaiah, P. Gegenwart, H.S. Jeevan, M. Wolf, J. Fink and U. Bovensiepen: Ultrafast Momentum-Dependent Response of Electrons in Antiferromagnetic $EuFe_2As_2$ Driven by Optical Excitation. Physical Review Letters **108** (9), 097002 (2012).

Rettig, L., P.S. Kirchmann and U. Bovensiepen: Ultrafast dynamics of occupied quantum well states in Pb/Si(111). New Journal of Physics **14** (2), 023047 (2012).

Rubano, A., L. Braun, M. Wolf and T. Kampfrath: Mid-infrared time-domain ellipsometry: Application to Nb-doped $SrTiO_3$. Applied Physics Letters **101** (8), 081103 (2012).

Sánchez-Portal, D., J. Stähler and X.-Y. Zhu: Basic theory of heterogeneous electron transfer. In: Dynamics at Solid State Surfaces and Interfaces, Vol. 2. (Eds.) U. Bovensiepen, H. Petek and M. Wolf. Wiley-VCH, Weinheim, 155-180 (2012). ISBN 978-3-527-40924-2.

Saywell, A., J. Schwarz, S. Hecht and L. Grill: Polymerization on Stepped Surfaces: Alignment of Polymers and Identification of Catalytic Sites. Angewandte Chemie International Edition **51** (21), 5096-5100 (2012).

Saywell, A., J. Schwarz, S. Hecht and L. Grill: Polymerization on Stepped Surfaces: Alignment of Polymers and Identification of Catalytic Sites. Angewandte Chemie **124** (21), 5186-5190 (2012).

Selvanathan, S., M.V. Peters, S. Hecht and L. Grill: Island formation and manipulation of prochiral azobenzene derivatives on Au(111). Journal of Physics: Condensed Matter **24** (35), 354013 (2012).

Shimada, T., C. Frischkorn, M. Wolf and T. Kampfrath: Maximizing the amplitude of coherent phonons with shaped laser pulses. Journal of Applied Physics **112** (11), 113103 (2012).

Sicot, M., P. Leicht, A. Zusan, S. Bouvron, O. Zander, M. Weser, Y.S. Dedkov, K. Horn and M. Fonin: Size-Selected Epitaxial Nanoislands Underneath Graphene Moiré on Rh(111). ACS Nano **6** (1), 151-158 (2012).

Sobota, J.A., S. Yang, J.G. Analytis, Y.L. Chen, I.R. Fisher, P.S. Kirchmann and Z.-X. Shen: Ultrafast Optical Excitation of a Persistent Surface-State Population in the Topological Insulator Bi_2Se_3. Physical Review Letters **108** (11), 117403 (2012).

Stähler, J., C. Gahl and M. Wolf: Dynamics and Reactivity of Trapped Electrons on Supported Ice Crystallites. Accounts of Chemical Research **45** (1), 131-138 (2012).

Sultan, M., U. Atxitia, A. Melnikov, O. Chubykalo-Fesenko and U. Bovensiepen: Electron- and phonon-mediated ultrafast magnetization dynamics of Gd(0001). Physical Review B **85** (18), 184407 (2012).

Timrov, I., T. Kampfrath, J. Faure, N. Vast, C.R. Ast, C. Frischkorn, M. Wolf, P. Gava and L. Perfetti: Thermalization of photoexcited carriers in bismuth investigated by time-resolved terahertz spectroscopy and ab initio calculations. Physical Review B **85** (15), 155139 (2012).

Tobey, R.I., S. Wall, M. Först, H. Bromberger, V. Khanna, J.J. Turner, W. Schlotter, M. Trigo, O. Krupin, W.S. Lee, Y.-D. Chuang, R. Moore, A.L. Cavalieri, S.B. Wilkins, H. Zheng, J.F. Mitchell, S.S. Dhesi, A. Cavalleri and J.P. Hill: Evolution of three-dimensional correlations during the photoinduced melting of antiferromagnetic order in $La_{0.5}Sr_{1.5}MnO_4$. Physical Review B **86** (6), 064425 (2012).

Vila Verde, A., P.G. Bolhuis and K. Campen: Statics and Dynamics of Free and Hydrogen-Bonded OH Groups at the Air/Water Interface. The Journal of Physical Chemistry B **116** (31), 9467-9481 (2012).

Wall, S., M. Rini, S.S. Dhesi, R.W. Schoenlein and A. Cavalleri: Advances in Ultrafast Control and Probing of Correlated-Electron Materials. IEEE Journal of Selected Topics in Quantum Electronics **18** (1), 18-91 (2012).

Wall, S., D. Wegkamp, L. Foglia, K. Appavoo, J. Nag, R.F. Haglund, J. Stähler and M. Wolf: Ultrafast changes in lattice symmetry probed by coherent phonons. Nature Communications **3**, 721 (2012).

Yanagita, T. and A.S. Mikhailov: Design of oscillator networks with enhanced synchronization tolerance against noise. Physical Review E **85** (5), 056206 (2012).

Yang, J., C. Nacci, J. Martinez-Blanco, K. Kanisawa and S. Fölsch: Vertical manipulation of native adatoms on the InAs(111)A surface. Journal of Physics: Condensed Matter **24** (35), 354008 (2012).

Zenichowski, K., C. Nacci, S. Fölsch, J. Dokic, T. Klamroth and P. Saalfrank: STM-switching of organic molecules on semiconductor surfaces: An above threshold density matrix model for 1,5 cyclooctadiene on Si(100). Journal of Physics: Condensed Matter **24** (39), 394009 (2012).

Doctoral Thesis

Duettmann, M.: Elastic Network Models of Proteins: Uncovering the Internal Mechanics of Actin and Myosin. Technische Universität Berlin 2012.

Giese, P.: Photoinduced N_2O dissociation and CO oxidation on thin MgO films. Freie Universität Berlin 2012.

Rettig, L.: Ultrafast Dynamics of Correlated Electrons. Freie Universität Berlin 2012.

Scheuch, M.: Strong Modulation of THz Optical Properties of Layered Materials by Thermal and Ultrafast Heating. Freie Universität Berlin 2012.

Diploma Theses

Kogler, F.: Interactions of artificial Molecular Machines. Freie Universität Berlin 2012.

Plottke, H.: Präparation und Charakterisierung von Wolfram-Nanospitzen als Feldemissions-Elektronenquelle. Freie Universität Berlin 2012.

Pronobis, W.: Ultrafast Optical Spectroscopy of Lattice Vibrations in Solids. Freie Universität Berlin 2012.

Master's Thesis

Popp, D.: Elastic networks: Study of large-scale conformational motions of membrane proteins. TU Bergakademie Freiberg 2012.

2013

Avigo, I., R. Cortés Rodriguez, L. Rettig, S. Thirupathaiah, H.S. Jeevan, P. Gegenwart, T. Wolf, M. Ligges, M. Wolf, J. Fink and U. Bovensiepen: Coherent excitations and electron–phonon coupling in Ba/$EuFe_2As_2$ compounds investigated by femtosecond time- and angle-resolved photoemission spectroscopy. Journal of Physics: Condensed Matter **25** (9), 094003 (2013).

Beye, M., T. Anniyev, R. Coffee, M. Dell'Angela, A. Föhlisch, J. Gladh, T. Katayama, S. Kaya, O. Krupin, A. Møgelhøj, A. Nilsson, D. Nordlund, J.K. Nørskov, H. Öberg, H. Ogasawara, L.G.M. Pettersson, W.F. Schlotter, J.A. Sellberg, F. Sorgenfrei, J.J. Turner, M. Wolf, W. Wurth and H. Öström: Selective Ultrafast Probing of Transient Hot Chemisorbed and Precursor States of CO on Ru(0001). Physical Review Letters **110** (18), 186101 (2013).

Bothschafter, E.M., A. Paarmann, E.S. Zijlstra, N. Karpowicz, M.E. Garcia, R. Kienberger and R. Ernstorfer: Ultrafast Evolution of the Excited-State Potential Energy Surface of TiO_2 Single Crystals Induced by Carrier Cooling. Physical Review Letters **110** (6), 067402 (2013).

Cabral, M.F., R. Nagao, E. Sitta, M. Eiswirth and H. Varela: Mechanistic aspects of the linear stabilization of non-stationary electrochemical oscillations. Physical Chemistry Chemical Physics **15** (5), 1437-1442 (2013).

Chang, J.Y., G. Khalsa, L. Moreschini, A.L. Walter, A. Bostwick, K. Horn, A.H. MacDonald and E. Rotenberg: Uniaxial strain induced band splitting in semiconducting SrTiO$_3$. Physical Review B **87** (11), 115212 (2013).

Chang, J.Y., L. Moreschini, A. Bostwick, G.A. Gaines, Y.S. Kim, A.L. Walter, B. Freelon, A. Tebano, K. Horn and E. Rotenberg: Layer-by-Layer Evolution of a Two-Dimensional Electron Gas Near an Oxide Interface. Physical Review Letters **111** (12), 126401 (2013).

Dehouck, Y. and A.S. Mikhailov: Effective Harmonic Potentials: Insights into the Internal Cooperativity and Sequence-Specificity of Protein Dynamics. PLoS Computational Biology **9** (8), 1003209 (2013).

Dell'Angela, M., T. Anniyev, M. Beye, R. Coffee, A. Föhlisch, J. Gladh, T. Katayama, S. Kaya, O. Krupin, J. LaRue, A. Møgelhøj, D. Nordlund, J.K. Nørskov, H. Öberg, H. Ogasawara, H. Öström, L.G.M. Pettersson, W.F. Schlotter, J.A. Sellberg, F. Sorgenfrei, J.J. Turner, M. Wolf, W. Wurth and A. Nilsson: Real-Time Observation of Surface Bond Breaking with an X-ray Laser. Science **339** (6125), 1302-1305 (2013).

Doblhofer, K.: Electrochemical Concepts: A Practical Guide. In: Chemical Energy Storage, (Ed.) R. Schlögl. De Gruyter, Berlin, 135-150 (2013). ISBN 978-3-11-026407-4.

Errami, H., M. Eiswirth, D. Grigoriev, W.M. Seiler, T. Sturm and A. Weber: Efficient Methods to Compute Hopf Bifurcations in Chemical Reaction Networks Using Reaction Coordinates. Lecture Notes in Computer Science **8136**, 88-99 (2013).

Ertl, G.: Molecules at Surfaces: 100 Years of Physical Chemistry in Berlin-Dahlem. Angewandte Chemie International Edition **52** (1), 52-60 (2013).

Ertl, G.: Moleküle an Oberflächen: 100 Jahre Physikalische Chemie in Berlin-Dahlem. Angewandte Chemie **125** (1), 52-60 (2013).

Gahl, C., D. Brete, F. Leyssner, M. Koch, E.R. McNellis, J. Mielke, R. Carley, L. Grill, K. Reuter, P. Tegeder and M. Weinelt: Coverage- and Temperature-Controlled Isomerization of an Imine Derivative on Au(111). Journal of the American Chemical Society **135** (11), 4273-4281 (2013).

Generalov, A.V., E.N. Voloshina and Y.S. Dedkov: Structural and electronic properties of graphene-based junctions for spin-filtering: The graphene/Al/Ni(1 1 1) intercalation-like system. Applied Surface Science **267**, 8-11 (2013).

Gong, C., J. Jiang, C. Li, L. Song, Z. Zeng, Y. Zheng, J. Miao, X. Ge, Y. Deng, R. Li and Z. Xu: Observation of CEP effect via filamentation in transparent solids. Optics Express **21** (20), 24120-24128 (2013).

Grill, L.: Imaging and manipulation of single molecules by scanning tunneling microscopy. In: Engineering of Chemical Complexity, (Eds.) A.S. Mikhailov and G. Ertl. World Scientific, New Jersey, 27-49 (2013). ISBN 978-981-4390-45-3.

Hofmann, O.T., J.-C. Deinert, Y. Xu, P. Rinke, J. Stähler, M. Wolf and M. Scheffler: Large work function reduction by adsorption of a molecule with a negative electron affinity: Pyridine on ZnO(10-10). The Journal of Chemical Physics **139** (17), 174701 (2013).

Huang, M.-J., R. Kapral, A.S. Mikhailov and H.-Y. Chen: Coarse-grain simulations of active molecular machines in lipid bilayers. The Journal of Chemical Physics **138** (19), 195101 (2013).

Kaluza, P.F.: Computation with phase oscillators: An oscillatory perceptron model. Neurocomputing **116**, 127-131 (2013).

Kampfrath, T., M. Battiato, P. Maldonado, G. Eilers, J. Nötzold, S. Mährlein, V. Zbarsky, F. Freimuth, Y. Mokrousov, S. Blügel, M. Wolf, I. Radu, P.M. Oppeneer and M. Münzenberg: Terahertz spin current pulses controlled by magnetic heterostructures. Nature Nanotechnology **8** (4), 256-260 (2013).

Kampfrath, T., K. Tanaka and K.A. Nelson: Resonant and nonresonant control over matter and light by intense terahertz transients. Nature Photonics **7** (9), 680-690 (2013).

Kim, K.S., T.-H. Kim, A.L. Walter, T. Seyller, H.W. Yeom, E. Rotenberg and A. Bostwick: Visualizing Atomic-Scale Negative Differential Resistance in Bilayer Graphene. Physical Review Letters **110** (3), 036804 (2013).

Kouvaris, N. and A.S. Mikhailov: Feedback-induced stationary localized patterns in networks of diffusively coupled bistable elements. Europhysics letters **102** (1), 16003 (2013).

Kumagai, T., F. Hanke, S. Gawinkowski, J. Sharp, K. Kotsis, J. Waluk, M. Persson and L. Grill: Controlling Intramolecular Hydrogen Transfer in a Porphycene Molecule with Single Atoms or Molecules Located Nearby. Nature Chemistry, in press.

Kumagai, T., F. Hanke, S. Gawinkowski, J. Sharp, K. Kotsis, J. Waluk, M. Persson and L. Grill: Thermally and vibrationally induced tautomerization of single porphycene molecules on a Cu(110) surface. Physical Review Letters, in press.

Mikhailov, A.S. and G. Ertl: Analysis, design and control of complex chemical systems. In: Engineering of Chemical Complexity, (Eds.) A.S. Mikhailov and G. Ertl. World Scientific, New Jersey, 3-23 (2013). ISBN 978-981-4390-45-3.

Mikhailov, A.S. and G. Ertl: Engineering of Chemical Complexity. World Scientific, New Jersey (2013). ISBN 978-981-4390-45-3.

Monney, C., A. Uldry, K.J. Zhou, A. Krzton-Maziopa, E. Pomjakushina, V.N. Strocov, B. Delley and T. Schmitt: Resonant inelastic x-ray scattering at the Fe L_3 edge of the one-dimensional chalcogenide $BaFe_2Se_3$. Physical Review B **88** (16), 165103 (2013).

Mota Lima, A.B., M. Eiswirth and E.R. Gonzalez: Enhanced Efficiency of CO-Containing Hydrogen Electroxidation with Autonomous Oscillations. The Journal of Physical Chemistry C **117** (24), 12495-12501 (2013).

Müller, M., A. Paarmann, C. Xu and R. Ernstorfer: Coherent Electron Source for Ultrafast Electron Diffraction and Imaging. The European Physical Journal - Web of conferences **41**, 10007 (2013).

Öström, H., M. Krenz, I. Radu, U. Bovensiepen, M. Wolf and C. Frischkorn: Coupling of spin and vibrational degrees of freedom of adsorbates at metal surfaces probed by vibrational sum-frequency generation. Applied Physics Letters **103** (13), 132403 (2013).

Pacile, D., P. Leicht, M. Pagano, P.M. Sheverdyaeva, P. Moras, C. Carbone, K. Krausert, L. Zielke, M. Fonin, Y.S. Dedkov, F. Mittendorfer, J. Doppler, A. Garhofer and J. Redinger: Artificially lattice-mismatched graphene/metal interface: Graphene/Ni/Ir(111). Physical Review B **87** (3), 035420 (2013).

Pashkin, A., A. Sell, T. Kampfrath and R. Huber: Electric and magnetic terahertz nonlinearities resolved on the sub-cycle scale. New Journal of Physics **15** (6), 065003 (2013).

Rettig, L., R. Cortés, J.-H. Chu, I.R. Fisher, F. Schmitt, P.S. Kirchmann, R.G. Moore, Z.-X. Shen, M. Wolf and U. Bovensiepen: Time-resolved Fermi surface mapping of the charge density wave material $DyTe_3$. The European Physical Journal - Web of conferences **41**, 03025 (2013).

Rettig, L., R. Cortés, H.S. Jeevan, P. Gegenwart, T. Wolf, J. Fink and U. Bovensiepen: Electron-phonon coupling in 122 Fe pnictides analyzed by femtosecond time-resolved photoemission. New Journal of Physics **15** (8), 083023 (2013).

Roeters, S.J., C.N. Van Dick, A. Torres-Knoop, E.H.G. Backus, K. Campen, M. Bonn and S. Woutersen: Determining In Situ Protein Conformation and Orientation from the Amide-I Sum-Frequency Generation Spectrum: Theory and Experiment. The Journal of Physical Chemistry A **117** (29), 6311-6322 (2013).

Sajadi Hezaveh, M. and N.P. Ernsting: Excess Dynamic Stokes Shift of Molecular Probes in Solution. The Journal of Physical Chemistry B **117** (25), 7675-7684 (2013).

Schick, D., R. Shayduk, A. Bojahr, M. Herzog, C.K. von Schmising, P. Gaal and M. Bargheer: Ultrafast reciprocal-space mapping with a convergent beam. Journal of Applied Crystallography **46** (5), 1372-1377 (2013).

Schiffrin, A., T. Paasch-Colberg, N. Karpowicz, V. Apalkov, D. Gerster, S. Mühlbrandt, M. Korbman, J. Reichert, M. Schultze, S. Holzner, J.V. Barth, R. Kienberger, R. Ernstorfer, V.S. Yakovlev, M.I. Stockman and F. Krausz: Optical-field-induced current in dielectrics. Nature **493** (7430), 70-74 (2013).

Shin, D., B. Jeong, B.S. Mun, H. Jeon, H.-J. Shin, J. Baik and J. Lee: On the Origin of Electrocatalytic Oxygen Reduction Reaction on Electrospun Nitrogen–Carbon Species. The Journal of Physical Chemistry C **117** (22), 11619-11624 (2013).

Stähler, J., O.T. Hofmann, P. Rinke, S. Blumstengel, F. Henneberger, Y. Li and T.F. Heinz: Raman study of 2,7-bis(biphenyl-4-yl-)2',7'-ditertbutyl-9,9'-spirobifluorene adsorbed on oxide surfaces. Chemical Physics Letters **584** (1), 74-78 (2013).

Tong, Y., A.V. Verde and K. Campen: The Free OD at the Air/D_2O Interface is Structurally and Dynamically Heterogeneous. The Journal of Physical Chemistry B **117** (39), 11753-11764 (2013).

Wall, S., L. Foglia, D. Wegkamp, K. Appavoo, J. Nag, R.F. Haglund, J. Stähler and M. Wolf: Tracking the evolution of electronic and structural properties of VO_2 during the ultrafast photoinduced insulator-metal transition. Physical Review B **87** (11), 115126 (2013).

Walter, A.L., A. Bostwick, F. Speck, M. Ostler, K.S. Kim, Y.J. Chang, L. Moreschini, D. Innocenti, T. Seyller, K. Horn and E. Rotenberg: Small scale rotational disorder observed in epitaxial graphene on SiC(0001). New Journal of Physics **15** (2), 023019 (2013).

Wegkamp, D., M. Meyer, C. Richter, M. Wolf and J. Stähler: Photoinduced work function modifications and their effect on photoelectron spectroscopy. Applied Physics Letters **103** (15), 151603 (2013).

Zenker, B., H. Fehske, H. Beck, C. Monney and A.R. Bishop: Chiral charge order in 1T-$TiSe_2$: Importance of lattice degrees of freedom. Physical Review B **88** (7), 075138 (2013).

Zhou, P., C. Streubühr, A. Kalus, T. Frigge, S. Wall, A. Hanisch-Blicharski, M. Kammler, M. Ligges, U. Bovensiepen, D. von der Linde and M. Horn von Hoegen: Ultrafast time resolved reflection high energy electron diffraction with tilted pump pulse fronts. The European Physical Journal - Web of conferences **41**, 10016 (2013).

Doctoral Thesis

Koch, M.: Growth and Characterization of Single Molecular Wires on Metal Surfaces. Freie Universität Berlin 2013.

Lafferentz, L.: Covalent Molecular Architectures and Dithienylethene Switches on Metal Surfaces. Freie Universität Berlin 2013.

Mielke, J.: Investigation of Single Functional Molecules on Metal Surfaces by Scanning Tunneling Microscopy. Freie Universität Berlin 2013.

Schambach, P.: Tip-enhanced Raman spectroscopy in ultra-high vacuum. Freie Universität Berlin 2013.

Weser, M.: Electronic and magnetic properties of graphene-based systems. Freie Universität Berlin 2013.

Diploma Theses

Loutchko, D.: Tryptophas Synthase: Single Enzyme Kinetic Modeling and Numerical Simulations. Humboldt-Universität zu Berlin 2013.

Lüneburg, S.: Novel electrostatic microlens for collimation of femtosecond electron pulses photoemitted from a nano-sized tip. Humboldt-Universität zu Berlin 2013.

5. Invited Talks of the Members of the Department of Physical Chemistry

Lea Bogner

Feb 2013	Seminar, Department of Physics (Prof. Katharina Franke/ Prof. Petra Tegeder), Freie Universität Berlin, Berlin, Germany *Poly(3-Hexylthiophene) Thin Films and Dicyanovinyl-Sexithiophene on Au(111) Investigated with Two-Photon Photoemission*
Apr 2013	Seminar, Department of Physics (AG Martin Weinelt), Freie Universität Berlin, Berlin, Germany *Electronic Structure and Excited States Dynamics in Polythiophene and an Acceptor-Subtituted Oligothiophene*

Kramer Campen

Jun 2012	Workshop on Nonlinear Optics at Interfaces, Telluride Science Research Center, Telluride, CO, USA *Understanding Methane Decomposition at the Ru(0001) Interface*
Aug 2012	244th ACS National Meeting and Exposition: Materials for Health and Medicine, Philadelphia, PA, USA *Understanding Methane Decomposition at the Ru(0001) Interface*
Sep 2012	CRC-FHI Joint Meeting on Complex Surfaces in Material Science, Berlin, Germany *Methane Decomposition at the Ru(0001) Interface*
Dec 2012	Group Seminar, Theoretical Chemistry, Institute of Chemistry, University of Potsdam, Potsdam, Germany *Structure and Dynamics of the Free OH at the Air/Water Interface*
Mar 2013	Integrated Research Training Group Seminar on Elementary Processes in Molecular Switches: Collaborative Research Center 658, Technische Universität Berlin, Berlin, Germany *Interfacial Spectroscopy: A Tool for Characterization of Semiconductor Surface Structures and Surface Solvation*
Jun 2013	CECAM Workshop on Liquid/Solid Interfaces: Structure and Dynamics from Spectroscopy and Simulations, Lausanne, Switzerland *Understanding Water Dissociation and Surface Reconstruction on the α-Al_2O_3(0001) Surface*
Sep 2013	20th International Conference on Horizons in Hydrogen Bond Research, Antwerp, Belgium *Structure and Dynamics of the Non-Hydrogen Bonded (Free) OH at the Air/Water Interface*
Oct 2013	Block Course on Dynamic Processes at Interfaces and Surfaces, International Max Planck Research School Complex Surfaces in Materials Sciences, Berlin, Germany *How to Probe Interfaces with Optical Sum Frequency Spectroscopy*

Jan-Christoph Deinert

May 2013	Seminar, Institute of Solid State Physics (Prof. Mario Dähne), Technische Universität Berlin, Berlin, Germany *Electronic Structure and Dynamics at the ZnO (10-10) Surface*

Yunpei Deng

Oct 2012	Meeting on the Frontiers in Optics/Laser Science XXVIII (FiO/LS), Rochester, NY, USA *Few Cycle Infrared OPCPA System and Applications*
Dec 2013	Conference on high intensity laser and attosecond science, Tel-Aviv, Israel *High Power OPCPA system for XUV sources at 500 kHz*

Markus Eiswirth

Sep 2011	Institute of Computer Science, University of Bonn, Bonn, Germany *Stoichiometric Network Analysis*
Jan 2012	B-IT Lecture Series in Life Science Informatics, University of Bonn, Bonn, Germany *Nonautocatalytic Oscillators and Olfactory Response*
Jun 2012	2nd Ertl Symposium on Surface and Interface Chemistry, Stuttgart, Germany *Chemical Kinetics and Algebraic Geometry*
Oct 2012	GIST, School of Environmental Science and Engineering, Gwangju, Korea *Dissipative Structures*
Oct 2012	ADeKo Conference Basic Science for the 21st Century, Seoul, Korea *Frozen Dynamical Patterns*
Oct 2012	GIST, Ertl Center for Electrochemistry and Catalysis, Gwangju, Korea *Stoichiometric Network Analysis*
Nov 2012	Dagstuhl-Seminar Symbolic Methods for Chemical Reaction Networks, Schloss Dagstuhl, Germany *Stoichiometric Networks and Dynamic Instabilities*

Ralph Ernstorfer

Aug 2012	MPS 2012 International Conference on Many Particle Spectroscopy of Atoms, Clusters and Surfaces, Berlin, Germany *Attosecond Photoelectron Spectroscopy of Solids*
Sep 2012	KTH Stockholm, School of Information and Communication Technology, Stockholm, Sweden *Attosecond Photoelectron Spectroscopy of Solids*

Sep 2012	Department Seminar, Department of Chemical Physics, Lund University, Lund, Sweden *Attosecond Photoelectron Spectroscopy of Solids and Optical-Field-Induced Current in Dielectrics*
Nov 2012	Institutskolloquium, Institute of Physics, Chemnitz University of Technology, Chemnitz, Germany *Non-Equilibrium Structural Dynamics in Solids and Optical-Field-Induced Current in Dielectrics*
Mar 2013	APS March Meeting, Baltimore, MD, USA *Attosecond View of the Photoelectric Effect*
Mar 2013	Seminar, Department of Chemistry, Columbia University, New York, NY, USA *Attosecond View of the Photoelectric Effect and Nonthermal Phase Transitions in Solids*
Mar 2013	Seminar, University of Delaware, Department of Physics and Astronomy, Newark, DE, USA *Optical-Field-Induced Current in Dielectrics*
Apr 2013	Seminar, Institute of Physics, Martin Luther University Halle-Wittenberg, Halle, Germany *Attosecond View of the Photoelectric Effect and Optical-Field-Induced Current in Dielectrics*
Sep 2013	Seminar, Ultrafast X-Ray Science Laboratory, Lawrence Berkeley National Laboratory, Berkeley, CA, USA *Non-Equilibrium Structural Dynamics and Phase Transitions in Solids*
Sep 2013	246th ACS National Meeting and Exposition, Indianapolis, IN, USA *Chemical Control of Heterogeneous Electron Transfer Dynamics and Strong Electron-Lattice Coupling in* TiO_2
Sep 2013	Physics Colloquium, University of Nebraska-Lincoln, Kearney, NE, USA *Non-Equilibrium Structural Dynamics and Phase Transitions in Solids*

Gerhard Ertl

Oct 2011	Lecture, 100 Jahre Fritz-Haber-Institut, Berlin, Germany *Molecules at Surfaces*
Nov 2011	Clemens Winkler Lecture, Freiberg, Germany *Katalyse an Oberflächen: Vom Atomaren zum Komplexen*
Nov 2011	Marie Sklodowska-Curie Symposium, Warsaw, Poland *Catalysis by Surfaces: From Atoms to Complexity*
Jun 2012	2nd Ertl Symposium on Surface and Interface Chemistry, Stuttgart, Germany *Reactions at Surfaces*
Aug 2012	4th EuCheMS Congress, Prague, Czech Republic *Catalysis at Surfaces*

Oct 2012	Lecture, Opening Ertl Center, Technische Universität Berlin, Berlin, Germany *Catalysis and Surface Science*
Nov 2012	Lecture, Ponteficia Accademia delle Scienze, Rom, Italy *Complexity in Chemistry: From Disorder to Order*
Jan 2013	Lecture, Romain-Rolland-Gymnasium, Berlin, Germany *Katalyse an Oberflächen*
Jan 2013	Lecture, Technische Universität Darmstadt, Darmstadt, Germany *Moleküle an Oberflächen*
May 2013	Otto-Stern-Symposium, Hamburg, Germany *Moleküle an Oberflächen*
May 2013	Lecture, Technische Universität München, Munich, Germany *Reaktionen an Oberflächen: Vom Atomaren zum Komplexen*
Jul 2013	Nobel Laureate Meeting, Lindau, Germany *Moleküle an Oberflächen*

Leonhard Grill

Sep 2011	FUNMOLS Workshop, Barcelona, Spain *Characterization and Manipulation of Single Functional Molecules by STM: From Switches to Wires*
Sep 2011	11th European Conference of Molecular Electronics (ECME 2011), Barcelona, Spain *Model Systems for Molecular Electronics Studied by Scanning Probe Microscopy*
Oct 2011	Symposium on Transport Through Molecules, Berlin, Germany *Conductance Measurements of Single Polymers as a Continuous Function of Their Length*
Jan 2012	Seminar, Chemistry Department, University of Graz, Graz, Austria *Single Functional Molecules at the Atomic Scale: Imaging, Manipulation and Assembly*
Feb 2012	Supramolecular Chemistry Conference, Lanzarote, Spain *Bottom-Up Assembly and Manipulation of Covalent Molecular Nanostructures*
Mar 2012	Seminar, Physics and Astronomy Department, University of Nottingham, Nottingham, UK *Functional Molecules Studied by STM: From Switches to Wires*
Apr 2012	Seminar, Institute of Science and Technology, Klosterneuburg, Austria *Single Functional Molecules at the Atomic Scale: Imaging, Manipulation and Assembly*
Jun 2012	CNR Workshop on Organic Molecules on Surfaces, Parma, Italy *Covalent Linking of Functional Molecules on Surfaces: Wires and Networks*

Jul 2012	Seminar, Physics Department, University of Graz, Graz, Austria *Single-Molecules Manipulation and Molecular Assembly on Surfaces*
Sep 2012	29th European Conference on Surface Science (ECOSS-29), Edinburgh, UK *Functional Molecules on Surfaces: Assembly and STM Manipulation*
Sep 2012	38th Micro and Nano Engineering Conference (MNE2012), Toulouse, France *Assembly and Manipulation of Functional Molecules at the Atomic Scale: From Switches to Wires*
Sep 2012	CRC-FHI Joint Meeting on Complex Surfaces in Material Science, Berlin, Germany *On-Surface Chemistry by Covalent Linking of Molecules on Metals*
Nov 2012	Lorentz-Center Workshop on Dynamical Phenomena at Surfaces, Leiden, The Netherlands *Manipulating Single Molecules with Mechanical, Optical or Electrical Functions*
Dec 2012	Conference on Molecular Electronics (ElecMol12), Grenoble, France *Assembly and Manipulation of Single Functional Molecules: Switches and Wires*
Jan 2013	Foresight Technical Conference: Illuminating Atomic Precision, Palo Alto, CA, USA *Assembly and Manipulation of Molecules at the Atomic Scale: Stitching and Switching*
Mar 2013	Interdisciplinary Surface Science Conference (ISSC-19), Nottingham, UK *Manipulation of Single Functional Molecules on Surfaces: Wires and Switches*
Jun 2013	Nanopack, Workshop, Grenoble, France *Bottom-Up Assembly of Functional Molecular Nanostructures*
Jun 2013	4th European Workshop on Nanomanipulation, Krakow, Poland *Manipulating Single Molecules and Nanostructures by STM*
Jul 2013	Seminar, Swiss Federal Laboratories for Materials Science and Technology (EMPA), Dübendorf, Switzerland *Assembly and Manipulation of Molecules at the Atomic Scale: "Stitching and Switching"*
Sep 2013	Trends in Nanotechnology Conference (TNT2013), Sevilla, Spain *Assembly and Manipulation of Single Functional Molecules*
Oct 2013	Workshop on Modeling Single-Molecule Junctions: Novel Spectroscopies and Control, Berlin, Germany *Combined Imaging, Spectroscopy and Conductance Measurements of Single Molecules: Towards Molecular Wires*
Oct 2013	543th Wilhelm-und-Else-Heraeus-Seminar on Electron Transport through Atoms, Molecules and Nanowires: Advances in Theory and Experiments, Bad Honnef, Germany *STM Manipulation of Single Functional Molecules: Towards Molecular Wires*

Karsten Horn

Mar 2012	VIIIth Prague Workshop on Photoinduced Molecular Processes, Prague, Czech Republic *Electronic Structures Studies of Surfaces and Solids Using Photoelectron Spectroscopy*
May 2012	Physikalisch-Chemisches Seminar, University of Göttingen, Göttingen, Germany *Quantum Well States in Thin Metallic Films: From Textbook Physics to Spin-Resolved Photoemission*
May 2012	Statistical Physics and Low Dimensional Systems, Abbaye des Prémontrés, Pont-à-Mousson, Frankreich *Electronic Structure and Fermi Surface of Decagonal Al-Cu-Co Quasicrystals*
Sep 2012	29th European Conference on Surface Science (ECOSS-29), Edinburgh, UK *Graphene - Electronic Structure of a 2D Model Solid*
Jan 2013	European Workshop on Epitaxial Graphene, Aussois, France *Electronic Structure and Many-Body Effects in Graphene Studied by Angle-Resolved Photoemission*
Jan 2013	International Workshop on Nanoscience on Surfaces - Present Stand and Future Challenges, Schladming, Austria *Graphene - Surface Science and Solid State Physics of a 2D Model Solid*
Mar 2013	Graphene Workshop: From Fundamental Properties to Applications, Ajdovscina, Slovenia *Electronic Structure of Epitaxial Graphene on Metals and Silicon Carbide*
Apr 2013	Seminar, Department of Physics and Astrononmy, Uppsala University, Uppsala, Sweden *Graphene - What Do We Learn about Its Electronic Structure from Angle-Resolved Photoemission?*
Jun 2013	Graphene Week, Chemnitz, Germany *Graphene Based Systems for Spintronics: Magnetic Interactions - Magnetic Interactions at the Graphene/3D Metal Interface*
Oct 2013	Seminar, Paul-Scherrer-Institut, Villigen, Switzerland *Aperiodic „Quasi“-Crystals and their Electronic Structure*
Oct 2013	Seminar, Swiss Federal Laboratories for Materials Science and Technology (EMPA), Dübendorf, Switzerland *Epitaxial Graphene and its Electronic Structure*

Tobias Kampfrath

Nov 2011	User Meeting at BESSY II (Berlin Electron Synchrotron), Berlin, Germany *Intense Terahertz Pulses as Driving Force of Electronic and Spin Excitations*

Dec 2011	Center of Excellence Colloquium, University of Göttingen, Göttingen, Germany *How to Enhance Magnetic Light-Matter Interaction at Highest Frequencies*
Apr 2012	Seminar, Department of Physics, Freie Universität Berlin, Berlin, Germany *Spectroscopy and Control of Spin Dynamics Using Terahertz Pulses*
May 2012	Seminar, Department of Physics, Freie Universität Berlin, Berlin, Germany *Spectroscopy and Control of Spin Dynamics Using Terahertz Pulses*
Jun 2012	Physikalisches Kolloquium, Chemnitz University of Technology, Chemnitz, Germany *Beyond Body Scanners: How to Use Terahertz Pulses to Observe and Control Spin Dynamics in Solids*
Jan 2013	Seminar, Department of Physics, University of Duisburg-Essen, Duisburg, Germany *Beyond Body Scanners: How to Use Terahertz Pulses to Observe and Control Spin Dynamics in Solids*
Jan 2013	12th Joint MMM/Intermag Conference (MMM/Intermag 2013), Chicago, IL, USA *Engineering Ultrafast Spin Currents and Terahertz Transients by Magnetic Heterostructures*
Mar 2013	DPG-Frühjahrstagung, Ratisbon, Germany *Manipulating Terahertz Spin Currents*
Mar 2013	Workshop on Terahertz/Carbon, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany *THz Transport of Charges and Spins*
Apr 2013	Colloquium, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany *Beyond Body Scanners: How to Use Terahertz Pulses to Observe and Control Spin Dynamics in Solids*
Jun 2013	Department colloquium, MPI for Polymer Research, Mainz, Germany *Spintronics with Terahertz Radiation*
Okt 2013	UMC 2013 (Ultrafast Magnetism Conference), Strasbourg, France *Spintronics with Terahertz Radiation*

Patrick S. Kirchmann

Jan 2012	Donostia International Physics Center, San Sebastián, Spain *Femtosecond Time- and Angle-Resolved Photoemission Spectroscopy*
Apr 2012	Department Workshop, Department of Physical Chemistry, FHI, Zeuthen, Germany *The Ultrafast Dynamics of Correlated Materials - Combining Time-Resolved Photoemission and Diffraction*
Jun 2012	Faculty of Physics, University of Duisburg-Essen, Duisburg, Germany *New Light on Charge Density Waves: Electron-Phonon Coupling in the CDW Material $TbTe_3$ Seen by Femtosecond Time-Resolved ARPES and Resonant Soft X-Ray Diffraction*

Jun 2012	Chair for Ultrafast Phenomena and Photonics at the University of Konstanz, Konstanz, Germany *New Light on Charge Density Waves: Electron-Phonon Coupling in the CDW Material* $TbTe_3$ *Seen by Femtosecond Time-Resolved ARPES and Resonant Soft X-Ray Diffraction*
Jun 2012	Chair of Solid State Physics at the Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany *Ultrafast Electron Dynamics in the Topological Insulator Material* Bi_2Se_3
Aug 2012	Virtual Institute Dynamic pathways in multidimensional landscapes, Scientific Kick-Off and Summer School, Helmholtz-Centre Potsdam - GFZ German Research Centre for Geosciences, Potsdam, Germany *Ultrafast Dynamics of Amplitude and Phase Excitations in Charge- and Spin-Ordered Materials*

Harald Kirsch

Apr 2013	Gordon Research Conference on Chemical Reactions at Surfaces: Surfaces in Energy and the Environment: Selected talk from submitted poster, Les Diablerets, Switzerland CH_4 *and* C_2H_4 *Decomposition and Higher Hydrocarbon Formation on Ru(0001)*

Alexey Melnikov

Mar 2012	APS March Meeting, Boston, MA, USA *Ultrafast Transport of Laser-Excited Spin-Polarized Carriers in Metallic Multilayers*
May 2012	Seminar, Department of Physics (AG Martin Weinelt), Freie Universität Berlin, Berlin, Germany *Ultrafast Spin Dynamics Induced by Superdiffusive Transport of Laser-Excited Spin-Polarized Hot Carriers in Metallic Multilayers*
Mar 2013	DPG-Frühjahrstagung, Ratisbon, Germany *Ultrafast Spin Dynamics Induced by Laser-Generated Spin Currents in Metallic Multilayers Probed by Non-Linear Magneto-Optics*
Apr 2013	Seminar, Department of Physics (AG Martin Aeschlimann), Technische Universität Kaiserslautern, Kaiserslautern, Germany *Ultrafast Spin Dynamics Induced by Laser-Generated Spin Currents in Metallic Multilayers and Probed by Non-Linear Magneto-Optics*
Jun 2013	Seminar, Department of Physics (AG Markus Münzenberg), University of Göttingen, Göttingen, Germany *Ultrafast Non-Local Spin Dynamics Induced by Hot Carrier Transport in Metallic Multilayers and Probed by (Non-Linear) Magneto-Optics*

Sep 2013	Advanced Laser Technologies 2013, Budva, Montenegro *Time-Resolved Non-Linear Magneto-Optics as a Tool to Study Spatially Non-Uniform Non-Local Ultrafast Spin Dynamics in Metallic Multilayers*

Alexander S. Mikhailov

Nov 2011	Lecture, Department of Physics, Central National University, Johngli, Taiwan *Elastic-Network Models of Molecular Machines*
Dec 2011	Seminar, Center for Theoretical Biology, Peking University, Peking, China *Evolutionary Design of Genetic Networks*
Mar 2012	Conference on Statistical Mechanics and Nonlinear Dynamics, National S. N. Bose Center for Basic Sciences, Calcutta, India *Elastic-Network Models of Molecular Machines*
Mar 2012	Seminar, Department of Physics, National Science University, Hefei, China *Turing Patterns in Network-Organized Activator-Inhibitor Systems*
Sep 2012	Workshop on Active Dynamics on Micro Scales: Molecular Motors and Self-Propelled Particles, Leiden University, The Netherlands *Elastic-Network Models of Molecular Machines*
Oct 2012	Symposium on Complex Network Analysis: From Graph Theory to Systems Biology, Hall in Tirol, Austria *Evolutionary Design of Robust Functional Networks*
Feb 2013	Conference on Self-Organization and Emergent Dynamics in Active Matter, Yukawa Institute of Theoretical Physics, Kyoto University, Kyoto, Japan *Elastic-Network Models of Protein Machines*

Claude Monney

May 2013	Colloquium, University of Greifswald, Greifswald, Germany *Possible Exciton Condensation in* $TiSe_2$*: Experimental and Theoretical Evidences*
May 2013	V. V. Nemoshkalenko Memorial Conference and Workshop, Electronic Structure and Electron Spectroscopies, Kiev, Ukraine *Electron-Hole Fluctuations Driving the Charge Density Wave Phase Transition in* $TiSe_2$
Jul 2013	CCP9/CECAM Workshop on Electronic Excitations and Photoelectron Spectroscopy: Bridging Theory and Experiment, Oxford, UK *Charge Density Wave Phase in* $TiSe_2$*: Calculating the Spectral Function Within the Exciton Condensate Model*
Jul 2013	CORPES 13 - International Workshop on Strong Correlations and Angle-Resolved Photoemission Spectroscopy, Hamburg, Germany *Complementary Momentum-Resolved Spectroscopies on a Charge Density Wave Material: ARPES and RIXS*

Aug 2013	8th International Conference on Inelastic X-Ray Scattering (IXS 2013), Stanford, CA, USA *Determining the Short-Range Spin Correlations in Edge-Shared Chain Cuprates with RIXS*

Melanie Müller

Oct 2013	Workshop on New Trends and Faces in Ultrafast Structural Dynamics, Stellenbosch, Südafrika *Femtosecond Low-Energy Electron Diffraction and Imaging: A Compact Approach*

Bruno Pettinger

Jun 2012	Il Congresso Nazionale di Spettroscopic Raman and Effetti Ottici Non Lineari, Bologna, Italy *Recent Progress in Tip-Enhanced Raman Spectroscopy*

Andrea Rubano

Mar 2012	MAMA-Protheo Workshop, Vietri, Italy *Optical Second Harmonic Generation as a Tool for the Characterization of Buried Interfaces*
Mar 2012	Seminar, Department of Physics, University of Naples, Naples, Italy *High Frequency THz-Ellipsometry on Perovskite Oxides*
Sep 2012	XCVIIIth National Congress of the Italian Physics Society (SIF), Naples, Italy *Optical Second-Harmonic Generation: A Powerful Tool for Buried Interfaces Investigation*
Sep 2012	Seminar, Department of Physics, University of Naples, Naples, Italy *Setup of a Versatile High-Field THz Spectrometer*
Nov 2012	Seminar, Department of Materials, ETH Zurich, Zurich, Switzerland *High Frequency THz-Ellipsometry on Perovskite Oxides*

Julia Stähler

Sep 2011	16th Workshop on Electronic Excitations, European Theoretical Spectroscopy Facility (ETSF), Turin, Italy *Two Sides of the Same Coin: Ultrafast Dynamics and Interactions of Electrons and Lattices*
Nov 2011	Seminar, Nanoscale Science Department, MPI for Solid State Research, Stuttgart, Germany *Ultrafast Electron Dynamics at Organic/Inorganic Interfaces Probed by 2PPE Spectroscopy*

Jan 2012	SFB 616 Colloquium, Faculty of Physics, University of Duisburg-Essen, Duisburg, Germany *Energy Level Alignment and Ultrafast Electron Dynamics at the Pyridine/ZnO(10-10) Interface*
Feb 2012	Department Seminar, Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse (CSNSM), Orsay cedex, France *Energy Level Alignment and Ultrafast Electron Dynamics at the Pyridine/ZnO(10-10) Interface*
May 2013	8th International Symposium on Ultrafast Surface Dynamics, Estes Park, CO, USA *Ultrafast Surface Dynamics of ZnO(10-10): Photons, Electrons, and Excitons*
Oct 2013	GDNÄ Tageskongress: Karrierewege - Frauen in Naturwissenschaft, Medizin und Technik, Berlin, Germany *Was ist eine Männerdomäne?*

Lutz Waldecker

Oct 2013	Workshop on New Trends and Faces in Ultrafast Structural Dynamics, Stellenbosch, Südafrika *Ultrafast Dynamics of Phase Transitions in* $Ge_2Sb_2Te_5$

Daniel Wegkamp

Apr 2013	Seminar, Department of Physics (AG Martin Weinelt), Freie Universität Berlin, Berlin, Germany *Long-Lived Electrons Trapped in Amorphous Ice Adsorbed on Cu(111) + Hot Electrons in the 2DEG at the STO/Vacuum Interface*

Martin Wolf

Oct 2011	Conference on X-Ray Spectroscopy on Magnetic Solids (XRMS 2011), Stanford, CA, USA *Dynamics of Correlated Materials Probed by trARPES*
Nov 2011	Distinguished Lectureship, APCTP-POSTECH-MPK, Pohang, Korea *Ultrafast Photoinduced Dynamics in Correlated Materials*
Nov 2011	Physikalisches Kolloquium, University of Göttingen, Göttingen, Germany *Ultrafast Photoinduced Dynamics in Solids*
Dec 2011	Conference on Ultrafast Chemical Physics and Physical Chemistry (UCP 2011), Glasgow, UK *Transient Electronic Structure Probed by trAPRES: From Electron Solvation in Ice to Phase Transitions in Solids*

Jan 2012	RIXS/REXS Workshop, SLAC National Accelerator Laboratory, Stanford, CA, USA *Time- and Angle-Resolved Photoemission Spectroscopy of Correlated Materials*
Jan 2012	Institutskolloquium, Ilmenau University of Technology, Ilmenau, Germany *Ultraschnelle Dynamik in komplexen Festkörpersystemen*
Feb 2012	Gordon Research Conference on Photonions, Photoionization and Photo-detachment, Galveston, TX, USA *Dynamics of Photo-Excited Electrons at Ice/Metal Interfaces: Electron Transfer and Solvation*
Feb 2012	Gordon Research Conference on Ultrafast Phenomena in Cooperative Systems, Galveston, TX, USA *Introductory Talk: Exploring Complex Quantum Systems of Condensed Matter*
Mar 2012	LCLS-II New Instruments Workshop, SLAC National Accelerator Laboratory, Stanford, CA, USA *Challenges in Catalysis and Dynamics of Surface Chemistry at LCLS II*
Apr 2012	Workshop on Exploiting Free Electron Lasers in Chemistry, University of Nottingham, Nottingham, UK *Surface Femtochemistry and Low Energy Excitations in Solids: Experiments in the Lab and at LCLS*
May 2012	SFB 951 Kolloquium, Humboldt-Universität zu Berlin, Berlin, Germany *Electron Dynamics and Energy Level Alignment at ZnO(10-10) Probed with 2PPE: Effect of Hydrogen and Pyridine Adsorption*
Jun 2012	TYC Energy Materials Workshop, King's College, London, UK *Ultrafast Dynamics of Interfacial Electron Transfer*
Jun 2012	Julius Springer Forum on Applied Physics, Berlin, Germany *Transient Electronic Structure of Photoexcited Solids Probed by Time-Resolved ARPES*
Jun 2012	2nd Ertl Symposium on Surface and Interface Chemistry, Stuttgart, Germany *Dynamics of Electron Transfer and Solvation Processes at Water Ice/Metal Interfaces*
Aug 2012	Plenary Lecture, JMC 13 Conference, Montpellier, France *Transient Electronic Structure of Complex Materials Probed by Time-Resolved ARPES*
Sep 2012	Plenary Lecture, International Conference CMD-24 and European Conference on Surface Science (ECOSS-29), Edinburgh, UK *Transient Electronic Structure and Ultrafast Dynamics of Solids*
Sep 2012	IMPACT Conference, Orsay, France *Transient Electronic Structure of Complex Materials Probed by Time-Resolved ARPES*

Oct 2012	CECAM Workshop on Surface Dynamics, Zaragoza, Spain *From Surface Femtochemistry to Ultrafast Phase Transitions in CDW Systems*
Nov 2012	Lorentz-Center Workshop, Leiden, The Netherlands *Probing the Transient Electronic Structure in Surface Femtochemistry and Ultrafast Phase Transitions in Solids*
Nov 2012	Photon Science Colloquium, DESY, Hamburg, Germany *Transient Electronic Structure and Ultrafast Dynamics of Solids*
Feb 2013	International Workshop on Physics at the Borderline between 1D and 2D, Bad Honnef, Germany *Ultrafast Dynamics of CDW Systems Probed by Time-Resolved ARPES*
Feb 2013	Workshop on Short-Time Dynamics in Strongly Correlated Systems, Bochum, Germany *Transient Electronic Structure and Lattice Dynamics of Photo-Excited Solids*
Feb 2013	SFB 616 Seminar, University of Duisburg-Essen, Duisburg, Germany *Transient Electronic Structure and Ultrafast Dynamics of Solids*
Feb 2013	WUPCOM Winterschool, Winklmoosalm, Germany *Ultrafast Changes in the Structural Properties of* VO_2 *During the Photoinduced Insulator-to-Metal Transition*
Mar 2013	Winterschool on Chemical Reaction Dynamics at Surfaces, Ringberg Castle, Germany *Probing the Transient Electronic Structure in Surface Femtochemistry*
May 2013	GDCh Kolloquium, Julius Maximilian University of Würzburg, Würzburg, Germany *Interfacial Charge Transfer Dynamics and Femtochemistry of Molecular Adsorbates*
May 2013	Plenary Lecture, 112. Bunsentagung, Karlsruhe, Germany *Interfacial Charge Transfer Dynamics and Femtochemistry of Molecular Adsorbates*
May 2013	International Workshop on Controlled Atomic Dynamics at Solid Surfaces, San Sebastián, Spain *Probing Transient Electronic Structure in Femtochemistry and Charge Transfer Processes at Surface*
May 2013	Plenary Lecture, Conference on Ultrafast Surface Dynamics, USD-8, Estes Park, CO, USA *Transient Electronic Structure and Lattice Dynamics of Photo-Excited Solids*
Jun 2013	International Workshop on Energy Dissipation at Surfaces, Bad Honnef, Germany *Dynamics of Electron Transfer and Exciton Formation at Interfaces*
Jun 2013	CNR-MPG Workshop on Quantum Science and Technology, Villa Vigoni, Italy *Dynamics of Solids: Insights from Ultrafast Spectroscopy*

Sep 2013	246th ACS National Meeting and Exposition, Indianapolis, IN, USA *Dynamics of Electron Transfer and Exciton Formation at Interfaces*
Sep 2013	International Conference on Dynamic Pathways in Mulitdimensional Landscapes, Berlin, Germany *Dynamics of Electron Transfer and Exciton Formation at Interfaces*
Oct 2013	Conference on Ultrafast Dynamics of Correlated Materials, ITCP, Trieste, Italy *Ultrafast Dynamics of Complex Materials Probed by Time-Resolved Photoemission*
Oct 2013	60th International Symposium, American Vacuum Society (AVS), Long Beach, NY, USA *Dynamics of Electron Transfer and Exciton Formation at Interfaces*

Max Planck Research Group for Structural and Electronic Surface Dynamics

Head: Ralph Ernstorfer

Group members

Scientists:

Roman Bertoni (from 10.2013)
Alexander Paarmann (until 12.2013)

Doctoral students:

Melanie Müller
Michele Puppin
Lutz Waldecker

Diploma and master's students:

Sebastian Lüneburg
Henry Plottke
Niels Schröter

Research projects funded from outside resources

- European Research Project (*NMP-2011-SMALL*): *"Time dynamics and Control in nanostructures for magnetic recording and energy applications (CRONOS)"*, project within work package FHIB (*Optimal control schemes of high-harmonic generation*), together with Martin Wolf.
  Project scientist: Yunpei Deng.
- DFG Research Unit FOR1700 *"Metallic nanowires on the atomic scale: Electronic and vibrational coupling in real world systems"*, Project E5 *"Time-resolved spectroscopy of photoinduced transitions and electronic excitations in quasi-1D metal wires on semiconductors"*, together with Martin Wolf.
  Project graduate student: Christopher Nicholson.

Max Planck Research Group for Structural and Electronic Surface Dynamics

Head: Ralph Ernstorfer

General

The Max Planck Research Group *Structural and Electronic Surface Dynamics*, established in September 2010 and hosted by the Department of Physical Chemistry, performs experimental research investigating ultrafast phenomena in condensed matter. Part of the group's research, in particular in its initial phase, has focused on the development of new experimental instrumentation and methodology. The work has been carried out by two postdoctoral researchers, three graduate students and three diploma or master's students.

Dr. Alexander Paarmann left the group at the end of 2013 for a group leader position in the Department of Physical Chemistry and the graduate students entered the final stage of their respective doctoral research studies. The transition to the 2nd generation of postdoctoral and doctoral researchers is initiated.

Scientific scope

The ground state properties of complex materials as well as the reaction pathways of photo-excited states are governed by many-body phenomena and the mutual dependence of electron, spin and lattice subsystems. Ultrafast techniques provide access to these fundamental correlations as femtosecond light pulses allow for subsystem-specific excitations and time-resolved observation of the same or another subsystem's response. In the limit of weak optical excitation, this concept may be seen as an experimental perturbative approach where correlations governing ground state properties are revealed by the system's response to modest, but specific excitations. Employing intense ultrashort laser pulses, on the other hand, allows for the preparation of transient states of matter exhibiting strong non-equilibrium, in particular between electrons and lattice. The coupling of the subsystems of such states can be significantly different compared to the ground state and the induced photo-physical or photo-chemical reaction may follow a non-thermal reaction pathway not available in thermal equilibrium.

These concepts require the application of different, complementary probes revealing the temporal evolution of the different subsystems: femtosecond electron diffraction (FED) unfolds the structural dynamics, femtosecond time- and angle-resolved photoelectron spectroscopy (trARPES) reveals the evolution of the electronic structure, and femtosecond optical spectroscopy provides the optical response. We investigate correlations and photo-induced reactions by employing, and partially developing, these ultrafast techniques. As these phenomena inherently depend on the symmetry of the system, specifically on the dimensionality of the translational symmetry, we put a particular emphasis on the development of ultrafast probes highly sensitive to structural and electronic dynamics in two- and one-dimensional systems.

Research fields

Non-equilibrium structural dynamics of crystals

Transition metal oxides like TiO_2 exhibit a pronounced coupling of electronic and lattice degrees of freedom. As valence and conduction bands are formed from different atomic orbitals, namely O *2p* and Ti *3d*, respectively, interband excitation effectively causes a charge re-distribution within the unit cell modifying the potential energy surface of the lattice. We investigate the effect of electron temperature and carrier relaxation on the excited-state potential energy surface prepared by a few-fs ultraviolet laser pulse (Fig.1). The analysis of the phase of the induced coherent Ti-O stretch vibration reveals a dynamic evolution of the lattice potential synced to the extremely fast cooling of charge carriers. This correlation between electron temperature and the lattice potential is in agreement with non-equilibrium density functional theory calculations performed by the group of Martin Garcia, Universität Kassel.

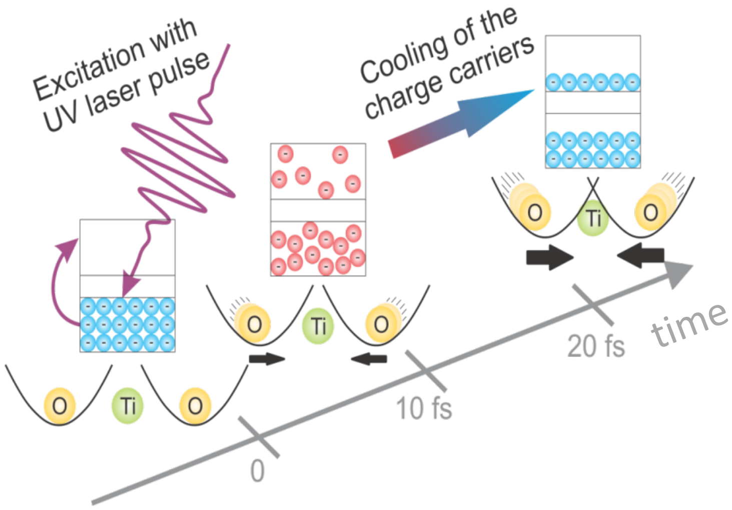


Figure 1: *Schematic representation of the effect of carrier relaxation on the lattice potential energy surface in TiO_2. E.M. Bothschafter et al., Phys. Rev. Lett.,* ***110****, 067402 (2013). Poster PC 21.*

Photo-induced phase transitions in solids

A limiting case of non-equilibrium electron-lattice interaction is the situation of electronic excitation inducing a structural phase transition. Such phase transitions may occur in a thermal fashion, i.e. after thermal equilibration between electrons and lattice, or nonthermally as a direct consequence of electronic excitation-induced changes to the lattice potential. The latter scenario occurs predominantly in covalently bound crystals as optical excitation transfers population from bonding to antibonding states. Currently, the ultrafast optical and structural response to photoexcitation of the phase change material $Ge_2Sb_2Te_5$ (GST) is investigated in collaboration with the group of Simon Wall, ICFO Barcelona. GST is commonly used as optical storage medium as it exhibits two meta-stable states with a large contrast in optical, electrical and crystalline properties. We aim to clarify the relation between the photo-induced ultrafast optical response and the structural response of the lattice by single-shot femtosecond optical spectroscopy and femtosecond electron diffraction in transmission mode employing a newly developed fs electron diffractometer providing a temporal resolution of 100 fs (Fig. 2).

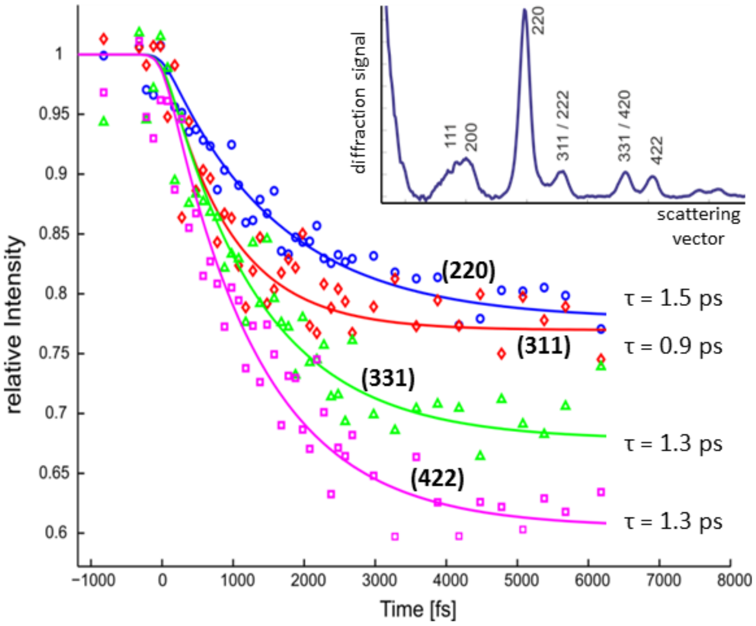


Figure 2: *Time-resolved electron diffraction data of poly-crystalline GST after reversible optical excitation with 800 nm light. The drop in diffraction intensity, the time-resolved Debye-Waller effect, reveals the rate of energy transfer from electrons to lattice through electron-phonon scattering. While the optical properties of GST exhibit a pronounced and persistent change within 100 fs after optical excitation, the lattice heats with a time constant of approximately 1 ps. This indicates that electronic effects, e.g. a transition from resonant to non-resonant bonding, dominate the ultrafast optical response. Poster PC 21.*

Femtosecond low-energy electron diffraction and imaging

As motivated above, there is strong interest in ultrafast experimental techniques that provide direct structural information, e.g. the structural evolution of a crystal or a molecule on the time scale of the fundamental vibrational modes. There has been tremendous progress in the development of femtosecond electron and x-ray diffraction and imaging techniques optimally suited for three-dimensional systems. The highest scattering cross-section, hence the highest sensitivity to structural dynamics in low-dimensional materials, however, is obtained with sub-keV electrons. We are developing a femtosecond low-energy electron diffraction and imaging apparatus based on a laser-triggered field emission electron source. The current operation modes include diffraction in transmission and point projection microscopy, see Fig.

3. Electron diffraction in reflection mode for the investigation of surface-specific structural dynamics will be added in the future.

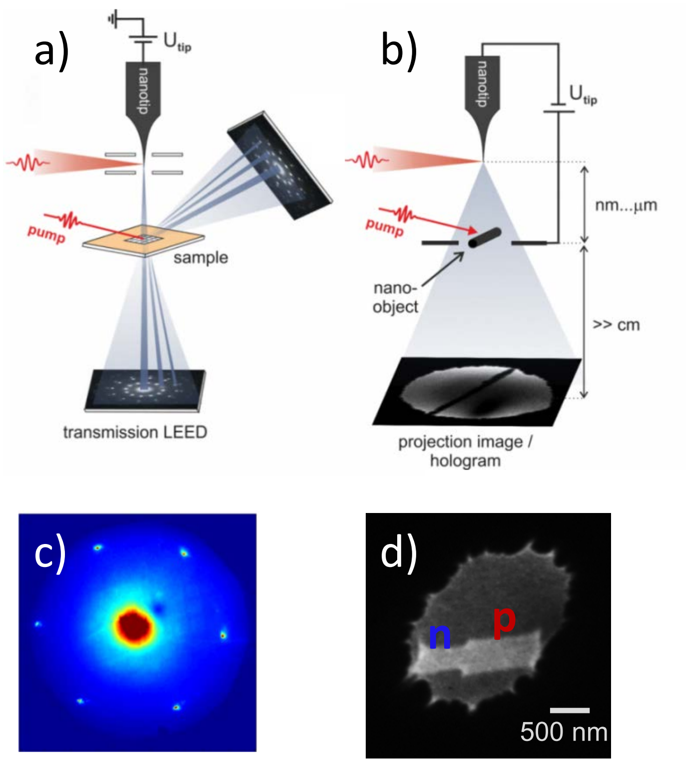


Figure 3: *Scheme of the time-resolved low-energy electron diffraction (a) and point projection microscopy (b). Coherent fs low-energy electron wave packets are generated from a laser-triggered nanotip. Employing an integrated micro-electrode for collimation, 2-D materials like free-standing graphene can be investigated in transmission mode (c). Taking advantage of the high sensitivity of low-energy electrons to electric fields, the electrostatic potential in the vicinity of a nanoobject can be investigated. Panel (d) shows the point-projection image of a spatially homogeneous InP nanowire with 30 nm diameter with integrated p-n junction. The p-n junction causes a local variation of the vacuum level visible in the projection image. In a time-resolved experiment, this approach will make it possible to visualize photo-induced currents in nanoobjects with high temporal and spatial resolution.*
Paarmann et al., J. Appl. Phys. 112, 113109 (2012); Lüneburg et al., Appl. Phys. Lett. 103, 213506 (2013). Poster PC 20.

High-repetition rate extreme ultraviolet laser for time- and angle-resolved photoelectron spectroscopy (trARPES)

Complementary to the structural probes introduced above, time-resolved ARPES provides a view on a material's electronic structure with potentially high temporal resolution such that movies of the electronic response to optical excitation may be obtained (see section 2.1.1 of the PC report and poster PC 18 for the material classes under investigation). In practice, there are two branches in the implementation of trARPES, one based on high repetition rate laser systems providing high counting statistics but limited probe photon energies, which limits the accessible range in the Brillouin zone, and another based on probing with extreme ultraviolet (XUV) light generated by high harmonic generation employing high power laser systems of limited repetition rate. In close cooperation with the research group *Dynamics of Correlated Materials* headed by Martin Wolf, we aim at bridging this technology gap by developing a high power laser system providing sufficiently short and intense laser pulses allowing for XUV generation at 500 kHz repetition rate. This approach is based on a two-stage optical parametric chirped-pulse amplifier (OPCPA) pumped with a hybrid fiber-slab laser system as front end. This laser provides spectrally tunable, both in terms of central wavelength and bandwidth, femtosecond laser pulses with average output power exceeding 20 W, see Fig. 4. As this laser system deviates from established approaches to laser-based XUV generation, a computer program for numerical simulations of high harmonic generation has been developed and employed in order to identify the optimal phase matching conditions for the given laser parameters. Next, this will be investigated experimentally.

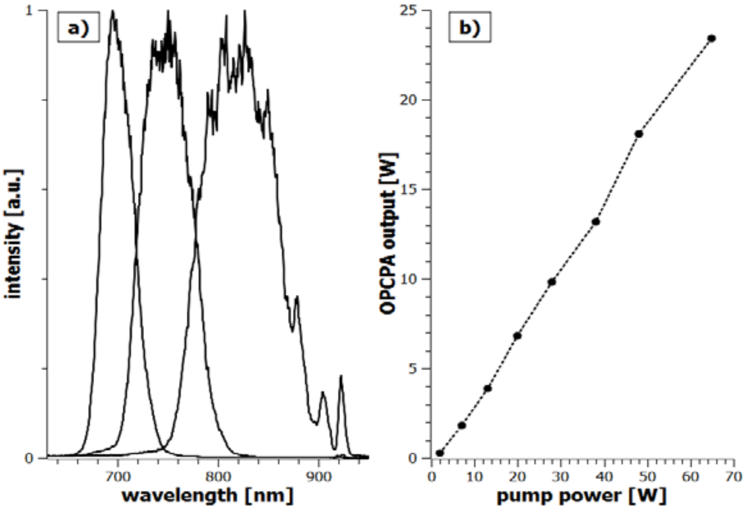


Figure 4: *Exemplary output spectra of the developed OPCPA laser system providing tunable visible and near-infrared pulses with adjustable time-bandwidth product (a). Power scaling of the second OPCPA stage output versus pump power revealing an almost constant conversion efficiency exceeding 30%.*
Poster PC 19.

Observation and control of electron motion in solids on the attosecond time scale

Propagating electrons in crystals are described as coherent superposition of Bloch states around a central wave vector. Free propagation of Bloch wave packets in crystals, however, is very limited in space and time as elastic and inelastic scattering processes destroy the wave packet's coherence. Ultrashort laser pulses with few femtosecond or attosecond duration allow for the investigation and even control of Bloch wave packet propagation in conventional crystals. For instance, attosecond photoelectron spectroscopy performed on metals covered with a variable number of adsorbate layers provides a time-resolved view on the propagation of Bloch wave packets from the substrate through the adsorbate. In an independent set of experiments, collective electron motion, which might be described as highly non-linear polarization, is induced and controlled by the shape of a laser pulse's electric field (see Fig. 5). These experiments demonstrate the feasibility of controlling macroscopic currents at optical frequencies.

These research projects had been initiated while being as scientist in the groups of R. Kienberger and F. Krausz (TU München and MPQ, Garching) and continued in collaboration since the start of the MPRG at the FHI.

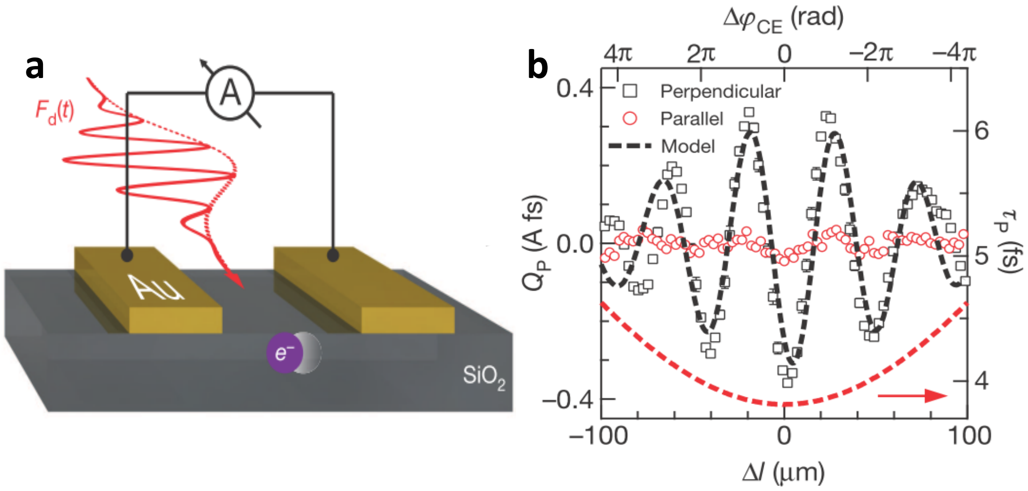


Figure 5: *Optical-field-induced current in dielectrics. A sub-two cycle laser pulse with controlled shape of its electric field is exposed to an unbiased metal-dielectric-metal nanojunction (a). Depending on the shape of the laser field, described in terms of the carrier-envelope phase* φ_{CE}*, a current is induced in the external circuit whose direction and amplitude critically depends on the symmetry of the pulse (charge flow per laser pulse:* Q_P*) (b).*

Schiffrin et al. Nature 493, 70 (2013). Paasch-Colberg et al., Nature Photonics, in print. Poster PC 22.

Publications (2012-2013)

A. Paarmann, M. Gulde, M. Müller, S. Schäfer, S. Schweda, M. Maiti, C. Xu, T. Hohage, F. Schenk, C. Ropers, and R. Ernstorfer: Coherent femtosecond low-energy single-electron pulses for time-resolved diffraction and imaging: A numerical study. J. Appl. Phys. **112**, 113109 (2012).

S. Neppl, R. Ernstorfer, E.M. Bothschafter, A.L. Cavalieri, D. Menzel, J.V. Barth, F. Krausz, R. Kienberger, and P. Feulner: Attosecond Time-Resolved Photoemission from Core and Valence States of Magnesium. Phys. Rev. Lett. **109**, 087401 (2012).

S.Kahra, G. Leschhorn, M. Kowalewski, A. Schiffrin, E. Bothschafter, W. Fuß, R. de Vivie-Riedle, R. Ernstorfer, F. Krausz, R. Kienberger,T. Schaetz: A molecular conveyor belt by controlled delivery of single molecules into ultrashort laser pulses. Nature Physics **8**, 238 (2012).

E.M. Bothschafter, A. Paarmann, E.S. Zijlstra, N. Karpowicz, M.E. Garcia, R. Kienberger, and R. Ernstorfer: Ultrafast Evolution of the Excited-State Potential Energy Surface of TiO_2 Single Crystals Induced by Carrier Cooling. Phys. Rev. Lett. **110**, 067402 (2013).

A. Schiffrin, T. Paasch-Colberg, N. Karpowicz, V. Apalkov, D. Gerster, S. Mühlbrandt, M. Korbman, J. Reichert, M. Schultze, S. Holzner, J.V. Barth, R. Kienberger, R. Ernstorfer, V.S. Yakovlev, M.I. Stockman, and F. Krausz: Optical-field-induced current in dielectrics. Nature **493**, 70 (2013).

S. Lüneburg, M. Müller, A. Paarmann, and R. Ernstorfer: Microelectrode for energy and current control of nanotip field electron emitters. Appl. Phys. Lett. **103**, 213506 (2013).

T. Paasch-Colberg, A. Schiffrin, N. Karpowicz, S. Kruchinin, Ö. Sağlam, S. Keiber, O. Razskazovskaya, S. Mühlbrandt, A. Alnaser, M. Kübel, V. Apalkov, D. Gerster, J. Reichert, T. Wittmann, J.V. Barth, M.I. Stockman, R. Ernstorfer, V.S. Yakovlev, R. Kienberger, and F. Krausz: Solid-state light-phase detector. Nature Photonics, in print.

Proceedings (2012-2013)

S. Neppl, R. Ernstorfer, A.L. Cavalieri, J.V. Barth, D. Menzel, F. Krausz, P. Feulner, and R. Kienberger: Probing ultrafast electron dynamics in condensed matter with attosecond photoemission. Proc. SPIE 8623, Ultrafast Phenomena and Nanophotonics XVII, 862312 (2013).

M. Müller, A. Paarmann, C. Xu, and R. Ernstorfer: Coherent Electron Source for Ultrafast Electron Diffraction and Imaging. EPJ Web of Conferences **41**, 10007 (2013).

E.M. Bothschafter, A. Paarmann, N. Karpowicz, E.S. Zijlstra, M.E. Garcia, F. Krausz, R. Kienberger, and R. Ernstorfer: Interband excitation and carrier relaxation as displacive driving force for coherent phonons. EPJ Web of Conferences **41**, 04021 (2013).

Diploma and master's theses

S. Lüneburg: Novel electrostatic microlens for collimation of femtosecond electron pulses photo-emitted from a nanosized tip, Humboldt-Universität zu Berlin, 2013.

N. Schröter: Numerical study of high harmonic generation as a light source for time- and angle-resolved photoelectron spectroscopy, Freie Universität Berlin, 2013.

H. Plottke: Präparation und Charakterisierung von Wolfram-Nanospitzen als Feldemissions-Elektronenquelle, Freie Universität Berlin, 2012.

Invited Talks (2012-2013)

Ralph Ernstorfer

Aug 2012	MPS 2012 International Conference on Many Particle Spectroscopy of Atoms, Clusters and Surfaces, Berlin, Germany *Attosecond Photoelectron Spectroscopy of Solids*
Sep 2012	KTH Stockholm, School of Information and Communication Technology, Stockholm, Sweden *Attosecond Photoelectron Spectroscopy of Solids*
Sep 2012	Department Seminar, Department of Chemical Physics, Lund University, Lund, Sweden *Attosecond Photoelectron Spectroscopy of Solids and Optical-Field-Induced Current in Dielectrics*
Nov 2012	Institutskolloquium, Institute of Physics, Chemnitz University of Technology, Chemnitz, Germany *Non-Equilibrium Structural Dynamics in Solids and Optical-Field-Induced Current in Dielectrics*
Mar 2013	APS March Meeting, Baltimore, MD, USA *Attosecond View of the Photoelectric Effect*
Mar 2013	Seminar, Department of Chemistry, Columbia University, New York, NY, USA *Attosecond View of the Photoelectric Effect and Nonthermal Phase Transitions in Solids*
Mar 2013	Seminar, University of Delaware, Department of Physics and Astronomy, Newark, DE, USA *Optical-Field-Induced Current in Dielectrics*
Apr 2013	Seminar, Institute of Physics, Martin Luther University Halle-Wittenberg, Halle, Germany *Attosecond View of the Photoelectric Effect and Optical-Field-Induced Current in Dielectrics*
Sep 2013	Seminar, Ultrafast X-Ray Science Laboratory, Lawrence Berkeley National Laboratory, Berkeley, CA, USA *Non-Equilibrium Structural Dynamics and Phase Transitions in Solids*
Sep 2013	246th ACS National Meeting and Exposition, Indianapolis, IN, USA *Chemical Control of Heterogeneous Electron Transfer Dynamics and Strong Electron-Lattice Coupling in* TiO_2
Sep 2013	Physics Colloquium, University of Nebraska-Lincoln, Kearney, NE, USA *Non-Equilibrium Structural Dynamics and Phase Transitions in Solids*

Melanie Müller

Oct 2013	Workshop on New Trends and Faces in Ultrafast Structural Dynamics, Stellenbosch, Südafrika *Femtosecond Low-Energy Electron Diffraction and Imaging: A Compact Approach*

Lutz Waldecker

Oct 2013	Workshop on New Trends and Faces in Ultrafast Structural Dynamics, Stellenbosch, Südafrika *Ultrafast Dynamics of Phase Transitions in* $Ge_2Sb_2Te_5$

Theory Department **Director: Matthias Scheffler**

Group leaders:

Carsten Baldauf
Volker Blum *(until Sept. 2013)*
Ignacio Franco *(until June 2013)*
Luca Ghiringhelli
Sergey Levchenko
Patrick Rinke
Alexandre Tkatchenko

Angel Rubio (Distinguished Visiting Scientist)

Guest scientists, staying for at least six months, paid by the FHI:

Heiko Appel
Carsten Baldauf
Susmita Basak
Amrita Bhattacharya
Saswata Bhattacharya
Fabio Caruso
Sucismita Chutia
Mateusz Marianski
Karsten Rasim
Xinguo Ren
Matti Ropo
Mariana Rossi Carvalho
Honghui Shang
Igor Ying Zhang

Guest scientists, staying for at least six months, paid from external funds:

Guillem Albareda P.	Marie-Curie	Christian Joas	Hist. of Science
Alberto Ambrosetti	ERC	Werner Jürgens	BMBF
Alexander Blum	History of Science	Wei Liu	ERC
Elizabeth C. Beret	AvH Fellow	Ermin Malic	TU Berlin
Wang Gao	ERC	Anthony Reilly	ERC
Katja Hansen	ERC	Yong Xu	AvH Fellow
Oliver Hofmann	Schrödinger		

Emeritus: **Alexander M. Bradshaw**

Graduate students: 19
+2 IMPRS

Diploma students: 2
+1 IMPRS

ERC Research Group: Organic Functional Materials and Molecular Interactions

Head: Alexandre Tkatchenko 6 post docs
4 graduate students
+1 IMPRS

Joint Network Center (GNZ) of the Berlin-Brandenburg Max Planck Institutions

Head: Gerd Schnapka
Staff members: 8

Recent Developments in the Theory Department

Director: Matthias Scheffler

V.1 d Importance of Many-body Effects and Self-interaction Corrections in the Adsorption Puzzle of CO on Metal Surfaces

V.1 e Importance of Space-Charge Effects for the Concentration of Defects at Metal-Oxide Surfaces

V.1 f Stability and Metastability of Clusters in a Reactive Atmosphere: Unexpected Stoichiometries of $\mathrm{Mg}_M\mathrm{O}_x$

V.1 g Critical Assessment of Li/MgO-Based Catalysts for the Oxidative Coupling of Methane

V.1 h Sub-Monolayer Water Adsorption on Alkaline Earth Metal Oxide Surfaces: A First-Principles Cascade Genetic Algorithm Study

V.1 i Unexpected Stability of O-Interstitial on Rough MgO Surfaces

V.1 j Towards Catalysis by Free Gold Clusters

V.2 Defects in Bulk Semiconductors and Insulators

V.2 a First-Principles Optical Spectra for F Centers in MgO

V.2 b Ferroelastic Switching in the Dynamics of Y- and Ti-stabilized ZrO2

V.2 c Electronic Structure and van der Waals Interactions in the Stability and Mobility of Point Defects in Semiconductors

V.3 Biophysics

V.3 a Building Blocks of Proteins

V.3 b Critical Role of Van der Waals Interactions for Polypeptide Secondary Structures

V.3 c Formation of the Protein α-Helix Motif in the Chemically HomologousClass of β-Peptides

V.3 d Water-Peptide Interaction

V.3 e Force Regulation of von Willebrand Factor (VWF) Activity

V.4 Organic Materials and Interfaces

V.4 a Reliable Modelling of Stabilities, Polymorphism, and Response Properties in Organic Materials

V.4 b Effects of Stress and Strain on the Electronic Structure of Organic Semiconductors

V.4 c Reliable Description of van der Waals Interactions for Inorganic/Organic Systems

V.4 d Charge Transfer and Charge Localization at Organic/Metal Interfaces

V.4 e Space-Charge Transfer in Hybrid Inorganic-Organic Systems

V.5 And more…

V.5 a The *f*-Electron Challenge: Lanthanides and Lanthanide Oxides in Density-Functional and Many-Body Perturbation Theory

V.5 b Van der Waals Interactions in Ionic and Semiconductor Solids

V.5 c Consistent Set of Band Parameters and Deformation Potentials for the Group-III Nitrides and Group-II Oxides

V.5 d Polymorphism in Ice at Ambient and High Pressures

V.5 e Gold Clusters at Finite Temperatures

V.5 f Sustainability and Resource Depletion

V.5 g History of Quantum Field Theoretical Methods in Many-Body Physics: The Story of RPA

I. General Remarks

The year 2013 marked the 25th anniversary of the foundation of the FHI *Theory Department*. Since the very beginning, research in the Department has been concerned with fundamental aspects of the chemical and physical properties of surfaces, interfaces, clusters, and nanostructures. Obviously, this theme is sufficiently general and covers such a wide range of topics, that there has been no need for a change, even when some years ago we added a group working in biophysics.

A significant part of our work concerns the development of new and advanced methods and their coding for high-performance computers. More specifically, validation of the electronic-structure base and linking it with methods from thermodynamics and statistical mechanics has always played a significant role in the *Theory Department*. While in the eighties, we developed the "*ab initio* atomistic thermodynamics" approach for defects in bulk semiconductors and at interfaces (together with Carl Michael Weinert and Jarek Dabrowski), in the nineties we complemented the *ab inito* molecular dynamics approach of Car and Parrinello by "*ab initio* kinetic Monte Carlo (kMC)" methods in order to reach the time scales relevant for crystal growth (together with Paolo Ruggerone, Christian Ratsch, Kristen Fichthorn, and Peter Kratzer). Then, in 2004 an "*ab initio* kMC" approach for heterogeneous catalysis was introduced (together with Karsten Reuter). Karsten continued this development by linking it to the proper description of gas-phase or fluid dynamics, in order to include mass and heat transport (he has been at the TU Munich since 2010). Recent developments in the Department have concentrated on techniques like basin hopping and a genetic algorithm for mapping out the enormous configuration space of larger molecules and clusters (Luca Ghiringhelli and Saswata Bhattacharya).

Progress in this field has been considerable in the last few years, and here, we briefly highlight some outstanding achievements. A more complete discussion of the breadth of the work of the Department is sketched in Sections IV and V.

- *Van der Waals interactions* – In 2008 Alexandre Tkatchenko joined the Department, shortly after he had finished his PhD in physical chemistry at the Universidad Autonoma Metropolitana (UAM-I), Mexico. In February 2009, the paper on "Accurate Molecular van der Waals Interactions from Ground-State Electron Density and Free-Atom Reference Data" appeared (*A. Tkatchenko and M. Scheffler, Phys. Rev. Lett.* ***102****, 073005 (2009)*), which by the end of 2013 had been cited already more than 500 times. Since then Alexandre has made several advances based on the initial idea, some are described below (Section IV). In 2011 he was awarded an ERC Starting Grant.

- *Renormalized perturbation theory, second order (rPT2)* — Another breakthrough concerns the development of advanced DFT functionals, in particular the "exact exchange plus correlation in the random-phase approximation (RPA)" approach. Identifying two contributions, which in many-body perturbation theory appear at the same level of approximation as the RPA, have yielded what we call "renormalized perturbation theory, second order (rPT2)". This approach treats electron correlation in terms of the RPA plus renormalized single excitations (rSE) plus second-order screened exchange (SOSEX): rPT2 = RPA + rSE + SOSEX. (*X. Ren, P. Rinke, G.E. Scuseria, and M. Scheffler: Renormalized Second-order Perturbation Theory for the Electron Correlation Energy: Concept, Implementation, and Benchmarks. Phys. Rev. B* ***88****, 035120 (2013)*).
- *Space-charge layer* — Work on defects at surfaces of MgO and parallel studies of the adsorption of organic molecules at ZnO surfaces showed that for charge transfer situations the build-up of a space charge layer not only gives rise to band bending, but also represents a significant energy cost. The latter had been neglected so far, largely because of technical reasons that prevented a proper theoretical description. Our developments and results have shown that the space-charge layer changes the concentration of charged defects or the amount of charge transfer at the surface significantly. It implies that carrier doping of the substrate bulk may be used for tuning defect or adsorbates concentrations and properties. (*N.A. Richter, S. Sicolo, S.V. Levchenko, J. Sauer, and M. Scheffler: Concentration of Vacancies at Metal Oxide Surfaces: Case Study of MgO (100). Phys. Rev. Lett.* ***111****, 045502* (published July 2013), and *Y. Xu, O.T. Hofmann, R. Schlesinger, S. Winkler, J. Frisch, J. Niederhausen, A. Vollmer, S. Blumstengel, F. Henneberger, N. Koch, P. Rinke, and M. Scheffler: Space Charge Transfer in Hybrid Inorganic-Organic Systems. Phys. Rev. Lett.* ***111****, 226802* (published November 2013)).
- *FHI-aims code* — The development of the FHI-aims code, which combined many of the earlier ideas and concepts developed in the Department (and even earlier), has been a great success. This applies to the daily computations in the Department, but also to the world-wide distribution of the code. FHI-aims was released in 2009 (*V. Blum, R. Gehrke, F. Hanke, P. Havu, V. Havu, X. Ren, K. Reuter, and M. Scheffler: Ab initio molecular simulations with numeric atom-centered orbitals. Comp. Phys. Comm.* ***180****, 2175 (2009)*). By the

end of 2013 the paper had been cited about 250 times. Recent developments are significant and briefly sketched out in Section IV below.

The following Section describes organizational developments and Section III personnel and related matters. Subsequently, Sections IV and V briefly outline some of the work performed in the *Theory Department*: Section IV describes conceptual, methodological, and technical developments, which enable us to study new types of problems and/or to improve the accuracy of calculations. Finally, in Section V, the main applications studied in the *Theory Department* are outlined and some recent results presented.

To keep things short, the presentation focusses on a description of progress achieved during the last two years. Nevertheless, a few studies from the last six years are also mentioned.

II. Organizational

We simply summarize the various aspects of the present situation in bullet form.

- The collaboration with the University of California Santa Barbara (UCSB, College of Engineering and College of Mathematical, Life and Physical Sciences) has been very fruitful. Since 2005 Matthias Scheffler spends about three months per year at the UCSB, although in the past two years this has been reduced to two months. The exchange of postdocs and PhD students between the institutes has turned out to be stimulating and very successful.
- The so-called “UCSB-MPS program” supports joint activities and exchange of students and postdocs between scientists working in Materials Science in the entire Max Planck Society (MPS) and the University of California Santa Barbara since 2005. For the MPS, the program is coordinated by Matthias Scheffler. The program will terminate in 2015.
- We are also part of a “Partnership for International Research and Education: Electron Chemistry and Catalysis at Interfaces” (PIRE-ecci). This activity, managed by Susannah Scott at UCSB, helps to intensify collaborations through the exchange of graduate students with chemical-physics institutes in China, in particular the Dalian Institute for Chemical Physics of the Chinese Academy of Sciences. In 2010 we added a German component to this activity by officially including the FHI, and in 2011, also the University of Göttingen.
- In 2009 the MPS decided to create “Max Planck Centers” at high-ranking universities outside Germany. The *Theory Department* is part of the “MPS-UBC Centre for Quantum Materials” which was established in October 2010 at the University of British Columbia

(UBC) in Vancouver. The Center is directed by George Sawatzky and Bernhard Keimer who chair a Scientific Board comprised of three Max-Planck directors, currently Bernhard Keimer, Hao Tjeng, and Matthias Scheffler, and three UBC professors, currently George Sawatzky, Andrea Damascelli, and Ian Affleck.

- At the École Polytechnique Fédérale de Lausanne (EPFL) the "Max Planck - EPFL Center for Molecular Nanoscience and Technology" has been initiated by Klaus Kern (MPI Stuttgart), together with Benoit Deveaud-Plédran (EPFL), Jeffrey Hubbell (EPFL), Thomas Rizzo (EPFL), Matthias Scheffler, and Alec Wodtke (Göttingen). The official start was January 1st, 2013. Several PhD students will jointly work with us and colleagues in Lausanne, receiving their PhD from EPFL.

As of December 1st, 2013 the FHI *Theory Department* is structured into 7 research groups. Nevertheless most of the work in the Department typically involves more than one of these groups. The groups and their leaders are:

- Unifying Concepts in Catalysis, headed by **Sergey Levchenko**
- *Ab Initio* Statistical Mechanics of Cluster Catalysis and Corrosion, headed by **Luca M. Ghiringhelli**
- Heat and Charge Transport, headed by **Matthias Scheffler**
- *Ab Initio* Biomolecular Simulations, headed by **Carsten Baldauf**
- Many Body Electronic Structure Theory, headed by **Patrick Rinke**
- Theoretical Spectroscopy, headed by **Angel Rubio**
- ERC group on Organic Functional Materials and Molecular Interactions, headed by **Alexandre Tkatchenko**
- In addition we host Emeritus **Alex Bradshaw** (since January 2009) who is working on photoionization in molecules and clusters as well as on questions of energy supply and resources in the context of the sustainability debate

Together with Martin Vingron (director at the MPI for Molecular Genetics) Matthias Scheffler is responsible for the *Joint Network Center* (Gemeinsames Netzwerkzentrum, GNZ) of the Berlin-Brandenburg Max Planck activities. This is a regional IT competence center, serving 8 MPIs and 5 additional institutions. It focusses on networking, backup, virtualization, and security services. It is headed by Gerd Schnapka and more information is given on p. 45.

III. Personnel and Related Matters

Since the last visit of the *Fachbeirat* in November 2011 the following noteworthy developments have taken place:

- In summer 2013 **Volker Blum** moved to his new post as Associate Professor in the *Department of Mechanical Engineering and Materials Science at Duke University*, North Carolina, USA. The department continues to collaborate with him on several projects and, together with Matthias Scheffler, he will remain coordinator of the FHI-aims developments.
- **Ignacio Franco** assumed the position Assistant Professor at the *Department of Chemistry at University of Rochester*, New York, USA in summer 2013.
- On January 1st, 2013 **Xinguo Ren** started his professorship at the *Key Laboratory of Quantum Information at the University of Science and Technology of China (USTC)* in Hefei, China. He remains one of the key developers and coordinators of the “beyond RPA” and *GW* components of the FHI-aims code.

Looking back at the last five years, 12 former members of the *Theory Department* have been appointed to lecturerships or professorships. In addition to the above, these are:

- Bothina Hamad, University of Jordan Amman, Jordan - since 2012
- Ville Havu, Aalto University School of Science, Espoo, Finland - since 2013
- Hong Jiang, Peking University, Beijing, China - since 2009
- Suljo Linic, University of Michigan, Ann Arbor, USA - since 2010
- Angelos Michaelides, University College London, London, UK - since 2009
- Rossitza Pentcheva, Universität Duisburg-Essen, Duisburg, Germany - from 2014
- Karsten Reuter, Technische Universität München, München, Germany - since 2009
- Aloysius Soon, College of Engineering Yonsei University, Seoul, South Korea - since 2010
- Hua Wu, Fudan University, Shanghai, China - since 2011

We are also proud to report that Mariana Rossi received the Otto Hahn Medal and the Otto Hahn Award in 2012. The latter awards the best PhD work concluded in the whole MPS (Sections for Humanities, for Biology and Medicine, and for Chemistry, Physics, and Technology) during the period from fall 2011 to fall 2012. About 900 PhD students finished their PhD during this period, and one of them, namely Dr. Rossi, was bestowed with the Otto Hahn Award. This gives her the opportunity to establish a small group at a Max Planck Institute of her choice when coming back to Germany after her postdoc stay abroad.

Members of the *Theory Department* take part in various national, European (EU and ESF) and international programs. The list of these activities is as follows:

- Einstein Foundation Berlin – Einstein Research Project ETERNAL: Exploring Thermoelectric Properties of Novel Materials, K.-R. Müller, C. Draxl, M. Scheffler - since April 2013.
- ERC – European Research Council, Starting Grant: Van der Waals Interactions in Complex Materials; A. Tkatchenko - since 2011.
- EU – European Commission, Marie Curie Actions, Initial Training Network (ITN): Surfaces for Molecular Recognition at the Atomic Level (SMALL), coordinator: J.N. O'Shea; M. Scheffler - since October 2010.
- DFG – German Research Foundation, Cluster of Excellence 314: Unifying Concepts in Catalysis (UniCat), coordinator: M. Driess; M. Scheffler - first funding period November 2007 - October 2012, new funding period until October 2017.
- DFG – German Research Foundation, SFB 951: Hybrid Inorganic/Organic Systems for Opto-Electronics (HIOS), spokesman: F. Henneberger; project B4: A. Knorr, P. Rinke, M. Scheffler - since July 2011.
- DFG – German Research Foundation, FOR 1543: Shear-flow regulation of Hemostasis (SHENC), spokesman: R. Schneppenheim; subgroup C1: F. Gräter, C. Baldauf - since June 2011.
- NSF – The National Science Foundation, Partnership for International Research and Education (PIRE): Electron Chemistry and Catalysis, director: S. Scott; M. Scheffler - since 2005.

- UCSB – University of California, Santa Barbara, College of Engineering and College of Mathematical, Life & Physical Sciences; M. Scheffler (distinguished visiting professor) - since 2005.
- Max Planck - EPFL Center for Molecular Nanoscience and Technology - Max Planck Society and the École Polytechnique Fédérale de Lausanne, directors: K. Kern, T. Rizzo; board members: B. Deveaud-Plédran, J. Hubbel, A. Wodtke, M. Scheffler - since 2013.
- MPS-UBC Center – Max-Planck-Gesellschaft – University of British Columbia Center for Quantum Materials, directors: B. Keimer, G.A. Sawatzky; board members: A. Damascelli, L.H. Tjeng, I. Affleck, M. Scheffler - since 2010.
- INCITE – US Department of Energy, Innovative and Novel Computational Impact on Theory and Experiment, computer allocation grant for 100 million CPU hours per year: Non-Covalent Bonding in Complex Molecular Systems with Quantum Monte Carlo, Oak Ridge/ Argonne Labs, PI: D. Alfè; M. Gillan, K. Jordan, A. Michaelides, A. Tkatchenko, M. Towler, O.A. von Lilienfeld - since 2012.
- CNMS – Center for Nanophase Materials Sciences, user project, CNMS2012-287, computer allocation grant for 450,000 CPU-hours: Finger Printing Protein Secondary Structure Motifs from First Principles: Computational Spectroscopy, Oak Ridge National Laboratory, PI: V. Blum - since 2012.
- PRACE – Partnership for Advanced Computing in Europe, PRACE-DECI-11 computer allocation grant for 14,700,000 standardized CPU-hours: Graphene Precursor Phase and Graphene-SiC(000-1) Interface by First Principles Thermodynamics and a Massively Parallel Structure Search (GraSiC-1), PI: V. Blum - since November 2013.
- Vetenskapsrådet – Swedish Research Council: “Catalysis on the atomic scale”, organizer: E. Lundgren; S. Levchenko, M. Scheffler - since 2011.

Since Professor Claudia Draxl was appointed to the Chair of Theoretical Solid State Physics at the Humboldt University Berlin in November 2011, some research of the *Theory Department* is being performed together with her and her group. In particular this concerns research on thermoelectric materials and the development of novel tools for the scientific analysis of *Big Data* that are now being generated on nearly every aspect in materials science (Christian Carbogno, Sergey Levchenko, Luca Ghiringhelli). A proposal for appointing Claudia “Max

Planck Fellow", as discussed at the last meeting of the Fachbeirat, had been put on hold by the MPS President's Office, because of a budget crises of the MPS that turned up in summer 2012.

Over the last twelve years we have strongly supported CECAM, a high-level European organization devoted to the promotion of fundamental research on advanced computational methods and their application to important problems in frontier areas of science and engineering. In particular, CECAM organizes scientific workshops and tutorials on computational molecular and materials science. It is financed by European research organizations. The Max Planck Society became a member in 2008. Since 2008 the CECAM headquarters is at the EPFL Lausanne, and "CECAM nodes" have been established in several member states. In Germany Matthias Scheffler was influential in creating a joint node, "cecam-MM1P.de", which focuses on multi-scale modeling from first principles (MM1P) with emphasis on methods and applications to materials and biophysics. The board of directors of the MM1P.de node consists of Thomas Frauenheim (University of Bremen), Eberhard K.U. Gross (MPI Halle), Matthias Scheffler, and Björn Winkler (Goethe University Frankfurt). Recently, the CECAM council endorsed the renewal of the Node agreement which will apply until 2018.

In the field of *ab initio* calculations of complex processes in materials CECAM has informal contacts to the Psi-k network. This is the umbrella organization for the electronic-structure community in Europe and beyond. Four years ago Psi-k was granted charitable status, and Matthias Scheffler is a member of the board of trustees. For example, Psi-k organizes a large Psi-k-conference every 5 years, one of the international key meetings in the field. The next one will be organized in 2015 by Angel Rubio.

Scientists of the *Theory Department* were involved in various services to the surface science and electronic-structure communities worldwide. For example, they lecture at the *Free University* and the *Technical University Berlin* as well as at the *International Max Planck Research School*. They organized or co-organized conferences, workshops, tutorials, and summer schools on topics in electronic-structure theory, multi-scale modeling, surface science, and biophysics, impacting upon the careers of numerous students of theoretical material science. This list of activities in the last two years includes:

- Conference on "SMALL is Beautiful: Theory Meets Experiment", November 9-12, 2011, Berlin, Germany; organizers: A. Tkatchenko and G. Held (University of Reading, UK)

- Symposium on “Frontiers of Electronic Structure Theory: Strong Correlations from First-Principles”, March 25-30, 2012 at DPG Spring Meeting, Berlin, Germany; organizers: S. Biermann (Centre de Physique Théorique, Palaiseau Cedex, France) and M. Scheffler
- 12th Einstein Lecture – David J. Gross - “Einstein's dream of unification - then and now”, June 29, 2012, Berlin, Germany; organizer: L. Wöste (Freie Universität Berlin, Germany) and M. Scheffler
- International Workshop on “Frontiers in Computational Material Science” July 18-21, 2012, Ringberg Castle, Germany; organizers: P. Rinke and M. Scheffler
- FHI-aims Developers' and Users' Meeting on “Density Functional Theory and Beyond with Numeric Atom-Centered Orbitals”, August 28-31, 2012, Berlin, Germany; organizers: V. Blum, M. Rossi, and M. Scheffler
- Workshop on “Towards First-Principles Description of van der Waals Interactions in Complex Materials”, October 15-19, 2012, Lausanne, Switzerland; organizers: A. Tkatchenko, A. Rubio, and M. Scheffler
- Workshop on “Vibrational coupling: most important, often ignored, and a challenge for *ab-initio* theory”, November 6-9, 2012, Lausanne, Switzerland; organizers: H. Appel, C. Carbogno, C. Draxl (Humboldt-Universität zu Berlin, Germany), and M. Scheffler
- Symposium on “Frontiers of Electronic Structure Theory: Discovery of Novel Functional Materials”, March 10-15, 2013 at DPG Spring Meeting, Regensburg, Germany; organizers: N. Marzari (École Polytechnique Fédérale de Lausanne, Switzerland), R. Drautz (Ruhr-Universität Bochum, Germany), and M. Scheffler
- International Summer School on “Basic Concepts and First-Principles Computations for Surface Science: Applications in Chemical Energy Conversion and Storage”, July 21-26, 2013, Norderney, Germany; organizers: K. Reuter (Technische Universität München, Germany) and M. Scheffler
- School on “Density-functional theory and beyond: Computational materials science for real materials”, August 6-15, 2013, Trieste, Italy; organizers: V. Blum, C. Baldauf, M. Scheffler, and R. Gebauer (International Centre for Theoretical Physics, Trieste, Italy)
- Symposium on “New Theoretical Concepts and Directions in Catalysis”, August 27-30, 2013, Santa Barbara, CA, USA; organizers: H. Metiu (University of California, Santa Barbara, USA) and M. Scheffler

- CECAM/Psi-k Research Conference on “Multi-scale Modeling from First-Principles”, September 8-13, 2013, Cap Roig, Spain; organizers: M. Mareschal (Zaragoza Scientific Center for Advanced Modeling, Zaragoza, Spain) and M. Scheffler
- IPAM (Institute of Pure and Applied Mathematics) long program on “Materials for a Sustainable Energy Future”, September 9 - December 13, 2013, IPAM/UCLA, Los Angeles, USA; organizing committee: M. Bazant (MIT, Cambridge, USA), G. Galli (University of California, Davis, USA), G. Henkelman (University of Texas, Austin, USA), K. Promislow (Michigan State University, East Lansing, USA), and M. Scheffler
- Workshop on “Fuels from Sunlight”, October 14-18, 2013, IPAM/UCLA, Los Angeles, USA; organizing committee: R. Klein (Freie Universität Berlin, Germany), J. Nørskov (Stanford University, USA), and M. Scheffler
- Workshop on “Energy Conservation and Waste Heat Recovery”, November 18-22, 2013, IPAM/UCLA, Los Angeles, USA; organizing committee: G. Galli (University of California, Davis, USA), R. James (University of Minnesota, Twin Cities, USA), J. Lukes (University of Pennsylvania, Philadelphia, USA), and M. Scheffler

One event that should be stressed was the “hands-on tutorial” in August 2013. This successful ten-day school (with 112 participants) was organized by Volker Blum, Carsten Baldauf, Matthias Scheffler, and Ralf Gebauer (ICTP). The tutorial was well received and there was very positive feedback from the participants, as can be seen in the report by the ICTP (http://www.ictp.it/about-ictp/media-centre/news/2013/9/dft_workshop.aspx).

Alex Bradshaw has served on several committees of the National Academy of Sciences (Leopoldina), National Academy of Engineering (acatech), and the Royal Society in the UK. He is currently member of an inter-academy study group on the prioritization of the various governmental and administrative measures associated with the transformation of the German energy supply system (*Energiewende*). He has also given evidence to parliamentary committees on sustainability questions and on open access.

IV. New Concepts, Methods and Techniques

A full description of the developments in the department during the last six years would exceed the space available for this summary. The 14 items below therefore give only an overview of the main highlights. We focus on the work done since summer 2011 and mention earlier work only in a few exceptional cases.

IV.1 Beyond the Random-Phase Approximation for Correlation

This work started in 2009 when we demonstrated that, and explained why, exact exchange plus correlation in the random-phase approximation (in short RPA) solves the "CO adsorption puzzle". Previous to our work it had been realized that standard exchange-correlation (XC) functionals often predict the wrong (adsorption) geometries; sometimes they even turn out to be qualitatively incorrect. The reason for this failure had not been fully understood, nor was a computationally tractable approach known that would provide a robust and reliable treatment. Our subsequent work on RPA-based methods contributed significantly to the resurgence of RPA in electronic-structure theory. A deeper analysis of RPA performance revealed that some of its shortcomings can be overcome by adding renormalized single excitations (rSE) and second-order screened exchange (SOSEX) terms. Both contributions emerge from a systematic diagrammatic expansion in many-body perturbation theory. We refer to the combined scheme of RPA+rSE+SOSEX as renormalized perturbation theory second order (rPT2), because it can be viewed as a renormalization of all second order diagrams. Systematic benchmarks show that the performance of rPT2 is well balanced across different quantities and systems e.g. atomization energies, energy barriers, and non-covalent interactions. For this reason, we are now using rPT2 as reference when assessing the accuracy of standard XC functions, such as PBE. Current and future work now addresses a) ways to go even beyond rPT2, b) to make RPA and rPT2 computationally more efficient, c) to develop new and compact basis sets that systematically approach the complete basis set limit, and d) to derive forces (Igor Zhang, Xinguo Ren, Volker Blum, Patrick Rinke).

IV.2 Self-Consistent *GW* Approach

The *GW* Green function approach has become the method of choice for the description of charged excitations in solids, and its application to molecules and nanosystems is steadily increasing. However, with few exceptions the ground-state properties of *GW* have not been explored yet. We have implemented a fully self-consistent *GW* scheme (scGW) that is based on the iterative solution of the Dyson equation into the FHI-aims code. Unlike in the more common, perturbative *GW* schemes (G_0W_0), scGW is independent of the reference state. This provides a well-defined total energy, which we calculate by means of the Galitskii-Migdal formula. To assess the scGW ground state, we calculated bond lengths, binding energies, and vibrational frequencies for a set of diatomic molecules. The accuracy of scGW is comparable to RPA, which is, however, not as good as that of rPT2. A detailed analysis of H_2 dissociation reveals that scGW and RPA are close around the equilibrium bond distance, but when it comes to bond breaking the DFT-based RPA outperforms Green-function theory. For prototypical donor/acceptor molecules, we demonstrate that scGW - due to its synergetic description of ground- and excited-state properties - is a promising method for charge-transfer systems, in which the electron density depends on the relative alignment of the molecular orbitals of the donor and the acceptor (Fabio Caruso, Patrick Rinke, Xinguo Ren, Angel Rubio).

IV.3 Full CI Quantum Monte Carlo

In quantum chemistry so called test sets have been developed, with which highest level calculations were performed for a selection of molecules. G2, HTBH38, S22, and ISO34 are well known examples of such test sets for covalent-bond interactions, reaction barrier heights, bio-oriented weak interactions, and isomerization energies, respectively. The importance of test sets is that approximations, e.g. of the XC treatment or the basis set, can be benchmarked against the almost exact results, providing a validation and error estimates of the approximate methods. Due to the lack of reliable electronic structure methods, such a test set does not exist so far for materials science.

With the mid-term goal of establishing a "test set for materials science and engineering" we are presently implementing the full configuration interaction quantum Monte Carlo (FCI-QMC) approach into the FHI-code with the aid of the post-SCF NECI code maintained by A. Alavi's group. For clusters this development has been largely completed, but for solids and surfaces significant developments are still needed. Parallel to the FCI-QMC approach we are also implementing coupled-cluster theory and double hybrid functionals (Igor Zhang).

IV.4 Consistent Combination of Hybrid Density Functionals and Quasiparticle Energy Calculations

To overcome the self-interaction error and the band-gap problem of density-functional theory (DFT), we combine hybrid density functionals with the G_0W_0 approach for quasiparticle excitations. We exploit the formally exact condition of exact DFT that the energy of the highest occupied molecular orbital (HOMO) corresponds to the ionization potential (IP) of a finite system. The α-value that controls the amount of exact exchange, and therefore the self-interaction error in the hybrid functional, is then optimized to obey this condition as closely as possible for our hybrid functional/G_0W_0 combination. This combined approach is essential for describing electron transfer (as exemplified by the TTF/TCNQ complex) and yields the vertical ionization potentials of the G2 set with a mean absolute percentage error of only ~3%. For MgO clusters, the formation energy of the optimal hybrid functional is in good agreement with coupled cluster reference calculations at the singles, doubles, and perturbative triples level (CCSD(T)). This validation then facilitated the description of oxygen vacancies at the MgO surfaces (see also **V.1 e**) (Viktor Atalla, Norina Richter, Sergey V. Levchenko, Patrick Rinke).

IV.5 Practical and Accurate Treatment of van der Waals Interactions

Van der Waals (vdW) interactions are ubiquitous in nature, playing a major role in defining the structure, stability, and function for a wide variety of molecules and materials. Our recent developments in methods for vdW interactions go beyond the pairwise Tkatchenko-Scheffler method that we introduced in 2009. We have developed the so-called many-body dispersion (MBD) method that determines the full many-body vdW energy in the dipole approximation for non-metallic systems (Alexandre Tkatchenko, Robert A. DiStasio Jr. and Roberto Car (Princeton University)). The efficiency of the method has been demonstrated by calculations on systems with more than 10,000 atoms (Alexandre Tkatchenko and Vivekanand Gobre) and the accuracy of DFT+MBD calculations has been thoroughly assessed for molecular materials (molecules, supramolecular systems, molecular crystals) and semiconductors (Alexandre Tkatchenko, Anthony M. Reilly, Noa Marom (Tulane University), Alberto Ambrosetti, Wang Gao, Guo-Xu Zhang). In all of these cases, the MBD method coupled with non-empirical PBE and PBE0 xc functionals is able to achieve the elusive “chemical accuracy” of 1 kcal/mol in the binding and cohesive energies. Further developments of the MBD method are ongoing in order to extend its applicability to highly polarizable materials (layered nanostructures) and metals. Our final goal is to systematically connect the MBD approach which depends exclusively on the electron density with RPA calculations based on Kohn-Sham DFT orbitals. Our recent demonstration that the MBD energy expression is equivalent to RPA for a model system of localized dipoles suggests that a seamless connection between MBD and RPA is eventually possible to achieve.

IV.6 Influence of van der Waals Interactions on Electronic Properties

The long-range vdW energy is a tiny fraction (~0.001%) of the total energy; hence it is typically assumed that it has no visible direct influence on the electronic properties of molecules and solids. To assess the reliability of this "conventional wisdom", we recently implemented a fully self-consistent version of the Tkatchenko-Scheffler DFA+vdW functional, where DFA stands for an approximate semi-local exchange-correlation density functional (Alexandre Tkatchenko, Nicola Ferri, Robert A. DiStasio Jr. and Roberto Car (Princeton University)). In agreement with previous work, we find a very small overall contribution from self-consistency in the structure and stability of vdW-bound molecular complexes. However, nonlocal vdW interactions turn out to significantly affect electronic properties of coinage metal (111) surfaces, leading to an increase of up to 0.3 eV in their workfunction, bringing it in agreement with experiments. Furthermore, vdW interactions are also found to noticeably affect workfunctions and charge transfer in hybrid organic/metal systems.

IV.7 Heat Transport by Phonons

An accurate first-principles determination of (vibrational) thermal conductivity has remained elusive: State-of-the-art techniques treat deviations from the harmonic approximations perturbatively and thus become questionable at elevated temperatures, e.g., above 70% of the melting temperature, at which the dynamics of the nuclei is significantly affected by anharmonic interactions. To overcome this limitation, we have implemented a variant of the *Green-Kubo* method that accounts for the full anharmonicity via molecular dynamics. The new and crucial step is the reformulation of the heat flux, which is required for assessing the thermal conductivity, within a *first-principles* framework. Starting from an *energy density* formalism, we have shown that the virial theorem allows the heat flux to be defined in terms of the *stress tensor*. In contrast to the energy density, this formulation is independent of the periodic boundary conditions and thereby alleviates the strong finite size artifacts that appear in the small supercells that are treatable from first principles. Whereas excellent results could be achieved with this method in the case of strong anharmonicity (low thermal conductivity), the description of good heat conductors (low anharmonicity) is still challenging due to the long time and large length scales required to achieve convergence in such cases. Currently, strategies to bridge these scales are being implemented and tested, e.g. the separation of time and length scales and the extrapolation via harmonic and quasi-harmonic potentials (Christian Carbogno, Ramamurthy Ramprasad (University of Connecticut)).

IV.8 Electronic Transport (Charge and Heat)

Many efforts have been undertaken to accurately describe the interactions (electron-phonon and electron-electron scattering) that drive electronic charge and heat transport on different time and length scales. For macroscopic transport, we have followed the lines of Sec. IV.7 and have implemented the *Greenwood-Kubo* method, in which the electronic transport coefficients are assessed from the electron-phonon scattering that arises in molecular dynamics. To bridge the necessary time and length scales, an implementation of the Boltzmann Transport formalism is used (Karsten Rasim, Christian Carbogno). On the microscopic, molecular scale, the group of Ferdinand Evers at the Karlsruhe Institute for Technology (KIT) has developed a *non-equilibrium Green-functions* module for FHI-aims that performs the calculation of transmission functions as well as charge and heat conductances (Alexei Bagrets and Ferdinand Evers (KIT)).

IV.9 Time-Dependent Density-Functional Theory for Quantum Electrodynamics - Bridging Materials Science and Quantum Optics

R.P. Feynman called Quantum Electrodynamics (QED) “the jewel of physics” for its extremely accurate predictions of quantities like the Lamb shift or the anomalous magnetic moment of the electron. It is the underlying theory for the non-relativistic Hamiltonian in Coulomb gauge that is often used as starting point for modern electronic-structure theory. Recently, we went back to the foundations of QED and, starting from the full QED-Lagrangian, have developed a time-dependent density-functional theory for QED. We have addressed how this new theory translates into lower levels of theory from the non-relativistic approximation to the Maxwell description of the photon field. Such an approach has the advantage that the quantization of the radiation field can be treated explicitly and in this way a more microscopic perspective on optical excitations which appear in photosynthesis, the vision process, solar cells, etc. can be gained. As starting point for our studies, we have constructed numerically exact solutions for QED model systems with entangled photons. Using an iterative fixed-point scheme, these exact solutions can be inverted to extract the exact Kohn-Sham potentials for our multicomponent TDDFT/QED approach. This provides an explicit route to find corrections to the classical Maxwell equations which incorporate the quantized nature of the electromagnetic field. We also have started to use our numerically exact QED solutions for the emerging field of correlated spectroscopy where, in contrast to normal spectroscopy, entangled photons are used to probe matter (Johannes Flick, Rene Jestädt, Heiko Appel, Angel Rubio).

IV.10 Electronic Decoherence in Molecules and Other Vibronically Coupled Systems

In the past two years, we have investigated electronic decoherence effects in molecules. Understanding electronic decoherence is vital to the development of approximation schemes to the full vibronic evolution of molecules and is central to our description of fundamental non-equilibrium processes such as photosynthesis and vision. Three basic contributions have been developed: 1.) We have introduced a hierarchy of measures of decoherence, termed "reduced purities", that can be used to characterize decoherence in many-electron systems. While usual measures of decoherence are based on the full many-body electronic density matrix and are thus not of general applicability, the reduced purities are based on the few-body electronic density matrix and can be used to characterize decoherence in the common case when only reduced information about the electrons is available. 2.) Using the reduced purities and related measures, we have investigated the basic electronic decoherence dynamics for the Su-Schrieffer-Heeger model of trans-polyacetylene by following the vibronic dynamics explicitly in a mixed quantum-classical Ehrenfest approximation. The simulations reveal the basic structure of the decoherence dynamics and illustrate the influence of system size and the nature of the initial state on the decoherence. 3.) Finally, in this context, we have identified a remarkable vibronic process that we call VIBRET in which the decay of an electron in the conduction band to a further inner state resonantly excites an electron in the valence band, leading to an oscillatory exchange of population between two distinct electronic states that lives for up to tens of picoseconds. Such oscillatory behavior suggests the presence of a long-lived electronic coherence. Interestingly, however, through the reduced purities VIBRET is shown to arise from a purely incoherent process (Ignacio Franco, Heiko Appel).

IV.11 Genetic Algorithm and the Cascade of Increasing Accuracy

Finding the global minimum of a given potential-energy surface (PES) is a formidable problem that cannot be solved either analytically or via grid-based methods, unless only a few degrees of freedom are involved. Only stochastic methods offer a viable alternative. The genetic algorithms (GA) form a class of stochastic global optimizers which are based on the principles of natural evolution. When applied to atomic structures and when a search for the global energy minimum is made, a GA is in practice designed to randomly select low-energy structures and combine them into a new test structure, which is then locally optimized. The accurate knowledge of the PES is one of the critical aspects of any global search. In order to avoid wasting accurate energy and force evaluations on high-energy and already-known structures, a “cascade” scheme was designed: A multi-stepped procedure in which successive steps employ higher levels of theory, with each next level using information obtained at the lower level. High-energy and already-known structures are discarded in one of the low-level steps (Luca M. Ghiringhelli, Saswata Bhattacharya).

IV.12 Simulating the Global Effects of Charge-Carrier Doping by the Virtual-Crystal Approximation

Introducing atomically dispersed extrinsic species into a material, called doping, can have diverse effects on the material's properties. In particular, it can promote or suppress formation of intrinsic defects. Theoretical description of the effects of doping have so far remained focused on local effects, namely on changes in geometric and electronic structure and formation energies of an isolated intrinsic defect in the vicinity of a dopant. However, doping usually also changes electron or hole conductivity of a material, providing new pathways for charge transfer and compensation on macroscopic length scales. The relative importance of these global effects of doping for defect formation and surface chemistry has not been assessed so far.

In order to address this problem, we developed an approach based on the virtual-crystal approximation (VCA). In this approach, instead of explicitly introducing dopants, we modify native species in the material in such a way that delocalized partially occupied states are introduced with only a slight perturbation of the valence states of the host system. This simulates the dopant-induced states responsible for changes in conductivity. In an all-electron framework, this is achieved by modifying charges of all or of a selected subgroup of atomic nuclei in the system. The advantage of this approach is that the density of the “virtual dopant” states, i.e. the dopant concentration, can be varied independently of the size of the unit cell.

This allows low dopant concentrations to be modeled using smaller unit cell sizes in periodic models (Norina Richter, Sergey V. Levchenko).

IV.13 Sensitivity Analysis in Heterogeneous Catalysis

The kinetics of heterogeneous catalysis is affected by various processes. On surfaces with steps and defects their number can reach well over a hundred. Using the (first-principles) kinetic Monte Carlo approach, we modify the rate of each process and measure the resulting change of the turnover frequency. Such a sensitivity analysis (also called "degree of rate control") identifies the key processes (typically more than one but less than ten). It is particularly important for finding (causal) descriptors for a "Big Data Analysis" of different catalyst materials (Karsten Reuter, Hakim Meskine and Sebastian Matera (TU Munich), Horia Metiu (UCSB)).

IV.14 The Atom-Centered Numeric Basis-Set Code "FHI-aims"

The FHI-aims all-electron code continues to expand as a strong base for the implementation and application of electronic structure methods developed in the department and elsewhere. Since 2011, the code has seen two major releases as well as continuous upgrades in the development branch. The number of developers that have contributed to the code in a noteworthy way now exceeds 50, most of them located in Berlin, but with several strong contributions from groups in Munich (Karsten Reuter), Helsinki (Ville Havu), Hefei (Xinguo Ren), London, and elsewhere. The key methodological choice behind the code are numeric atom-centered basis functions, allowing us to reach a computational accuracy on par with, e.g., the full-potential LAPW approach, but roughly at the same cost as plane-wave pseudopotential calculations. Several significant methodological additions to the code have been mentioned in the above sections, including: Correlation methods beyond the random-phase approximation; numeric atom-centered basis sets with valence correlation consistency for H-Ar, to obtain basis-set converged total energies for correlated methods directly; self-consistent *GW*; many-body perturbation inspired approaches to van der Waals dispersion interactions in DFT. Further important improvements include:

- Completed implementation of non-periodic and periodic hybrid functionals in an $O(N)$ scaling approach based on a localized version of resolution of the identity (RI) and enhanced auxiliary RI basis sets (Sergey V. Levchenko, Jürgen Wieferink, Arvid Ihrig, Xinguo Ren, Patrick Rinke, Matti Ropo, Volker Blum)

- Stress tensor and unit cell relaxation for local and semilocal DFT, hybrid functionals, and pairwise van der Waals contributions (Christian Carbogno, Franz Knuth, Viktor Atalla, Jürgen Wieferink, Volker Blum)

- Initial implementation of linear-response time-dependent density-functional theory for molecular optical spectra (Jan Kloppenburg, Heiko Appel, Volker Blum)

- Initial implementation of periodic *GW* and RPA methods in reciprocal-space and real-space frameworks (Xinguo Ren, Sergey V. Levchenko, Mark Glaß)

- QM/MM Embedding infrastructure for surface models as clusters by repulsive pseudopotentials and point charges (Daniel Berger and Karsten Reuter (TU Munich), Volker Blum)

- Connection to i-pi infrastructure (Michele Ceriotti and coworkers) for nuclear quantum effects modelled by path-integral Monte Carlo (Mariana Rossi)

- “aimsChain” infrastructure to compute transition states: currently, nudged elastic band and the string method (Yingyu Yao, Luca M. Ghiringhelli)

- Linear macroscopic dielectric tensor (Björn Bieniek, Christian Carbogno) and Kubo-Greenwood transport (Christian Carbogno, Karsten Rasim)

- Connection to “aitranss” infrastructure (Evers group, KIT) for molecular transport based on the Landau-Buettiker formalism (Alexei Bagrets (KIT))

- Generalized Langevin Equation colored-noise thermostat to include nuclear quantum effects (Mariana Rossi)

V. Applications – Some Highlights

This Section briefly summarizes some computational studies, results, and insights from the various research fields and topics studied in the *Theory Department* during the last six years. The selection criteria for the examples presented were their visibility or expected impact. To keep this Section to a reasonable length, emphasis is put on the last two years.

V.1 Surfaces, Adsorption, and Heterogeneous Catalysis

V.1 a Benzene Adsorbed on Transition-Metal Surfaces: Structure and Adsorption Mechanism

The adsorption of aromatic molecules on metal surfaces plays a key role in condensed matter physics, catalysis, and functional materials. We systematically studied the adsorption of benzene on transition-metal surfaces, demonstrating that our recently developed DFT+vdW$^{\text{surf}}$ method allows quantitative treatment of both weak and strong adsorption on metal surfaces, thus extending the already excellent performance found for molecules in the gas phase. Notably, we found that van der Waals forces contribute more to the binding of covalently bonded benzene than they do when benzene is physisorbed (Alexandre Tkatchenko, Wei Liu, Victor G. Ruiz, Guo-Xu Zhang, Biswajit Santra, Xinguo Ren).

V.1 b Graphene Engineering: Stability of Epitaxial Graphene and the Surface Reconstructions of 3C-SiC

SiC is an important material for growing graphene films directly on a semiconducting substrate. The underlying process is Si sublimation. Several experimental groups claim 'wafer-scale' high-quality monolayer graphene films on the Si side of SiC, and first devices are being made by groups at IBM, U Erlangen, and elsewhere. We have shown that these films in fact grow due to the establishment of specific equilibrium, or very-near equilibrium conditions, and are not just necessary kinetic intermediates during Si sublimation. This result will help focus future growth efforts on the C side (where the same growth mode is not possible) and probably also on other substrates (Volker Blum, Lydia Nemec, Florian Lazarevic, Patrick Rinke).

V.1 c Ultrathin Oxides: Bulk Oxide-Like Model Surfaces or Unique Films?

On the nanoscale, materials often reveal new and unexpected features. Ultrathin silica films on Mo(112), for example, differ significantly from the surfaces of bulk SiO_2 and should therefore be considered as new materials with interesting and novel properties of their own. The same is true for monolayer ZnO films on the (111) surfaces of Ag, Cu, Pd, and Pt. Thicker films, on the other hand, approach the structure of the bulk ZnO surface and might potentially serve as model surfaces in catalysis or for the study of hybrid inorganic/organic systems (Patrick Rinke, Christoph Freysoldt, Björn Bieniek).

V.1 d Importance of Many-body Effects and Self-interaction Corrections in the Adsorption Puzzle of CO on Metal Surfaces

Since the adsorption of CO on metals requires a good description for systems that are as diverse as metal surfaces and small molecules, it has become an important benchmark system for electronic-structure methods. We have demonstrated that RPA outperforms the common density functionals, because it combines exact exchange, which removes the self-interaction error, with a correlation energy based on the renormalized (screened) Coulomb potential that is finite for metallic systems and yields the right decay behavior outside a metal surface (Patrick Rinke and Xinguo Ren).

V.1 e Importance of Space-Charge Effects for the Concentration of Defects at Metal-Oxide Surfaces

We have calculated the concentration of oxygen vacancies at the (100) surface of doped MgO as a function of temperature, oxygen pressure, and dopant concentration, using *ab initio* atomistic thermodynamics. The global electrostatic energy of the charge separation generated by the electron transfer from the vacancies to the dopant-induced states is considered as part of the free energy of the system. We find that a space-charge layer with a thickness up to hundred nanometers forms under the surface at realistic conditions solely due to the formation of charged oxygen vacancies at the surface (Norina Richter and Sergey V. Levchchenko, Sabrina Sicolo and Joachim Sauer (HU Berlin)).

V.1 f Stability and Metastability of Clusters in a Reactive Atmosphere: Unexpected Stoichiometries of Mg_MO_x

By applying the cascade genetic algorithm (IV.11) and *ab initio* atomistic thermodynamics, we identify the stable and metastable compositions and structures of Mg_MO_x clusters at realistic temperatures and oxygen pressures. Small clusters ($M < 5$) are found to be in thermodynamic equilibrium when $x > M$. The non-stoichiometric clusters exhibit peculiar magnetic behavior, suggesting the possibility of tuning magnetic properties by changing environmental pressure and temperature conditions (Luca M. Ghiringhelli, Saswata Bhattacharya, Sergey V. Levchenko).

V.1 g Critical Assessment of Li/MgO-Based Catalysts for the Oxidative Coupling of Methane

We have used *ab initio* atomistic thermodynamics to analyze stability of different types of defects on the Li-doped MgO(100) surface. The Li substitutional defects, as well as their complexes with oxygen vacancies, are predicted to segregate from the bulk to the surface. There they form islands which are rich in Li substitutional defects and oxygen vacancies. These findings have helped to explain recent experimental observations, and prompted a critical re-assessment of the relevance of a large number of previous theoretical and experimental studies on the practical application of Li-MgO as a catalyst for the oxidative coupling of methane (Sergey V. Levchenko).

V.1 h Sub-Monolayer Water Adsorption on Alkaline Earth Metal Oxide Surfaces: A First-Principles Cascade Genetic Algorithm Study

Using *ab initio* atomistic thermodynamics and the cascade genetic algorithm (IV.11), we have studied trends in the stability of adsorbed water structures on alkaline earth metal-oxide surfaces. We find a range of H_2O chemical potentials where one-dimensional adsorbed water structures are thermodynamically stable on CaO(001). On MgO(001) and SrO(001) such structures are not found. The formation of the one-dimensional structures is explained by the balance between water-water and water-surface interactions (Xunhua Zhao, Saswata Bhattacharya, Luca M. Ghiringhelli, Sergey V. Levchenko).

V.1 i Unexpected Stability of O-Interstitial on Rough MgO Surfaces

In order to model the thermodynamic stability of rough metal oxide surfaces in a reactive atmosphere, we calculate, by combining *ab initio* atomistic thermodynamics and the cascade genetic algorithm (IV.11), the relative stability of medium-size parallelepipedal $[(\mathrm{MgO})_N]^q$ clusters (N = 18, 24, 32, 108), isolated or embedded in a point-charge matrix, at various charge states q as function of T, p_{O2}, and doping conditions (accounted for by the chemical potential of the electrons, μ_e). We find that, at realistic temperatures, pressures, and μ_e, O_2 or

O_3 interstitial states at the oxygen corners of pristine clusters are favored for *all* studied charge states (Saswata Bhattacharya, Sergey V. Levchenko, Luca M. Ghiringhelli).

V.1 j Towards Catalysis by Free Gold Clusters

Using constrained *ab initio* atomistic thermodynamics we have studied free gold clusters in contact with an atmosphere composed of O_2 and CO. The approach enables us to account for spin, temperature, and pressure effects. The results suggest that the adsorption of an incoming O_2 molecule drives the system from the singlet to the triplet spin state, and vice versa, and the oxidation reactions then occur on either the S=0 or the S=1 potential-energy surface (Elizabeth C. Beret, Merel M. van Wijk, Luca M. Ghiringhelli).

V.2 Defects in Bulk Semiconductors and Insulators

V.2 a First-Principles Optical Spectra for F Centers in MgO

The study of the oxygen vacancy in MgO has been aggravated by the fact that the positively charged and the neutral vacancy absorb at practically identical energies in the optical spectrum. We apply the Bethe-Salpeter approach on top of a newly developed scheme for defect formation energies in the G_0W_0 approach and obtain optical spectra in unprecedented agreement with experiment. Our calculations reveal that not only the absorption but also the emission spectra of different charge states peak at the same energy, which leads to a reinterpretation of the optical properties of the F center. (Patrick Rinke, André Schleife (University of Illinois at Urbana-Champaign), Emmanouil Kioupakis (University of Michigan), Anderson Janotti and Chris Van de Walle (UCSB) and Claudia Rödl and Friedhelm Bechstedt (University of Jena)).

V.2 b Ferroelastic Switching in the Dynamics of Y- and Ti-stabilized ZrO2

Using *ab initio* molecular dynamics and the *generalized solid state-nudged elastic band* method, which accounts for the unit-cell degrees of freedom, we have studied the thermodynamic properties of ZrO_2 based ceramics. By investigating extended supercells we have identified a novel phase stabilization mechanism for the high-temperature cubic phase. This particular dynamics, which can be controlled via cation doping, involves the realignment of the lattice vectors along different cartesian directions (Christian Carbogno, Carlos Levi (UCSB)).

V.2 c Electronic Structure and van der Waals Interactions in the Stability and Mobility of Point Defects in Semiconductors

Defects are abundant in real-world materials where they significantly modify the electronic, optical, and magnetic properties. Density functional theory calculations with hybrid functionals are essential to achieve a reasonable description of defect electronic levels and defect formation energies. However, these functionals significantly overestimate the experimental migration barriers. Notably, the inclusion of screened van der Waals interactions on top of hybrid functionals significantly improves the description of defect formation energies and brings migration barrier heights into close agreement with experimental values (Wang Gao, Alexandre Tkatchenko).

V.3 Biophysics

V.3 a Building Blocks of Proteins

Secondary structure (helices, sheets, turns, etc.) are the key conceptual building blocks used to understand structure formation (and ultimately, function) in peptides and proteins. Our goal has been to quantify the contributions that determine the stability of helices and turns. Regarding helices, direct comparisons to gas phase experiments enabled us to show that especially the vibrational entropy can tip the balance towards this structure, avoiding the formation of less organized, more compact “blobs”. On the other hand, we showed that specific ion-peptide interactions can in fact disrupt the neutral, organized secondary structure, using turn-forming tetrapeptides as an example. A key question that must be asked, however, is whether first-principles methods are worth the effort for solving such questions - i.e., what is the accuracy needed to match what experiment can resolve. We have addressed this question for a set of benchmark experiments of a helical peptide from the Rizzo group, showing that indeed, a hybrid functional and many-body dispersion interactions are needed to capture this 100-atom chain (Mariana Rossi, Franziska Schubert, Carsten Baldauf, Sucismita Chutia, Matti Ropo, Volker Blum, Gert von Helden (MP Department)).

V.3 b Critical Role of Van der Waals Interactions for Polypeptide Secondary Structures

Folding and unfolding processes are important for the functional capability of polypeptides and proteins. We have employed both the pairwise and many-body methods for vdW interactions to show that their accurate inclusion is essential for understanding the stability of peptides *in vacuo*. VdW interactions qualitatively change the conformational landscape of alanine polypeptides, and greatly enhance the thermal stability of helical structures, in agreement with gas-phase experiments. Many-body effects in the vdW energy increase in importance for larger and structurally more complex peptides (Mariana Rossi, Volker Blum, Alexandre Tkatchenko, Joel Ireta Moreno (Metropolitan Autonomous University, México)).

V.3 c Formation of the Protein α-Helix Motif in the Chemically HomologousClass of β-Peptides

The β-peptide backbone compensates the limitations of natural peptides for a potential use as drugs, namely by increasing lifetime, enlarging chemical space, and improving folding behavior. We have demonstrated here the potential of β-peptides to imitate the most-prominent structural motif of natural proteins, the α-helix. A first-principles conformational search served as the basis for simulations of vibrational spectra and the computation of molecular shapes (collision cross sections) which were then compared with the corresponding experimental values (Franziska Schubert, Kevin Pagel (MP Department), Volker Blum, Carsten Baldauf).

V.3 d Water-Peptide Interaction

Perhaps the most frequently emphasized question in the peptide structure area concerns the interaction with water. Here, too, a critical first step is to understand the interaction between peptides and individual water molecules quantitatively, including adsorption energies, adsorption trends, and conformational preferences. Based on a series of measured equilibrium constants from the Jarrold and Bowers groups, we showed that DFT-PBE+vdW predictions yield excellent agreement with experiments for lysine-terminated polyalanine chains, including a suprising sensitivity to peptide chain length which is traced to the vibrational contribution. This can be addressed in the harmonic approximation, but it is clear that this approach will be of limited value in future. Real (anharmonic) “nuclear quantum effects” are needed, especially for protons. We have therefore connected our approach to path integral dynamics methods, initially for small water clusters. We are continuing to work towards an incorporation of such effects in a way that is effective for larger peptide molecules and electronic structure theory (Mariana Rossi, Sucismita Chutia, Michele Ceriotti (EPFL), David Manolopoulos (University of Oxford), Volker Blum).

V.3 e Force Regulation of von Willebrand Factor (VWF) Activity

VWF is a large multimeric blood protein. Its maximal length of up to 40µm makes it sensitive to shear-flow, which acts as a trigger to regulate blood clotting mediated by cross-linking injured vessel walls and blood platelets. In this project, we perform force field simulations at the level of protein domains and have been able to elucidate a force-dependent auto-inhibition mechanism that regulates the VWF interaction with a platelet receptor and collagen. Thus, we are beginning not only to understand physiology in atomistic detail, but also to discover new drug targets to potentially intervene in threatening diseases like strokes (Camilo Aponte-Santamaría and Frauke Gräter (Heidelberg Institute for Theoretical Studies (HITS), Carsten Baldauf).

V.4 Organic Materials and Interfaces

V.4 a Reliable Modelling of Stabilities, Polymorphism, and Response Properties in Organic Materials

Organic materials are of great fundamental and applied importance. We have demonstrated that the recently developed DFT+MBD method is capable of reaching accuracies of a few kJ/mol for both crystalline and gaseous phases compared to high-level theoretical and experimental stabilities, giving a systematic and seamless method for modeling both gas-phase and condensed molecular materials. The importance of accurate treatment of dispersion goes far beyond energetic stabilities. Many response properties show even larger contributions from many-body dispersion interactions (Anthony M. Reilly, Alberto Ambrosetti, Alexandre Tkatchenko).

V.4 b Effects of Stress and Strain on the Electronic Structure of Organic Semiconductors

Stress, strain, and pressure are key determinants of the stability, mechanical and electronic properties of any material, but are particularly important in semiconductors. Being able to predict the effects of stress / strain on the electronic structure accurately, however, involves non-trivial decisions, including the electronic structure method itself, a quantitative treatment of stress in periodic systems, and the question of the impact of nuclear effects (electron-phonon interaction) on the eventual results. We have completed a full implementation of the stress tensor for numeric atom-centered basis sets in the FHI-aims code. For the prototypical organic semiconductor polyacetylene, we show that the effect of pressure on the band gap is correctly captured *if* a hybrid functional with a sufficiently high exact exchange contribution is used (HSE06 with a fraction 0.5 of exact exchange or higher). This is closely connected to the chain dimerization in the electronic ground state, i.e., the electronic gap itself can be described by a static symmetry-broken arrangement at the Born-Oppenheimer surface (Franz Knuth, Viktor Atalla, Volker Blum, Christian Carbogno).

V.4 c Reliable Description of van der Waals Interactions for Inorganic/Organic Systems

Reliable modeling of the structure, stability, and electronic properties of complex adsorption systems remains a daunting task for modern electronic structure calculations. Our recently developed DFT+vdW$^{\mathrm{surf}}$ method extends the applicability of semi-local DFT to organic/metal systems and leads to a few surprising findings: (1) The vdW energy can contribute more to the binding of covalently bonded systems than it does for that of physisorbed molecules; (2) the physically bound precursor state for aromatics on Pt(111) can be more stable than the corresponding chemisorbed state; (3) many-body vdW interactions lead to a binding energy for a fullerene molecule adsorbed on multi-layered graphene that *decreases* as a function of the number of underlying graphene layers (Wei Liu, Victor G. Ruiz, Sergey Filimonov, Javier Carrasco, Alexandre Tkatchenko).

V.4 d Charge Transfer and Charge Localization at Organic/Metal Interfaces

For organic adsorbates that strongly hybridize with a metal surface, we find that semi-local density functionals provide an adequate description of charge transfer and the associated work-function change compared to experimental data. When the hybridization between metal and adsorbate is prevented by a NaCl spacer layer, semi-local functionals predict a fractional electron transfer resulting in equally charged molecules, while hybrid functionals, are able to break the translation symmetry and produce integer charging of only a fraction of the adsorbate layer (Oliver T. Hofmann, Viktor Atalla, Patrick Rinke).

V.4 e Space-Charge Transfer in Hybrid Inorganic-Organic Systems

For the hybrid inorganic-organic system tetrafluoro-tetracyanoquinodimethane (F4TCNQ) on the ZnO(000-1) surface we have shown that the adsorption energy and the electron transfer depend strongly on the bulk doping concentration of ZnO. The associated work-function changes are large, for which the formation of space-charge layers is the main driving force. Our prediction of a large work-function change at vanishing charge transfer is in agreement with photoemission experiments on the same system (Yong Xu, Oliver T. Hofmann, Patrick Rinke, Raphael Schlesinger, Sylke Blumstengel, Fritz Henneberger and Norbert Koch (HU Berlin), Antje Vollmer (Helmholtz-Zentrum Berlin)).

V.5 And More…

V.5 a The *f*-Electron Challenge: Lanthanides and Lanthanide Oxides in Density-Functional and Many-Body Perturbation Theory

We challenge the notion that classifies the *f*-electron lanthanides as strongly correlated. We find that, unlike conventional density functionals, the advanced RPA functional predicts a pressure-induced isostructural α to γ phase transition in cerium with a transition pressure in good agreement with the experimental extrapolation to 0 K. The similarly good agreement between photoemission spectra and G_0W_0 calculations for the lanthanide sesquioxides unveils that the variation of the band gaps across the series can be explained in terms of the *f*-electron filling (Marco Casadei and Patrick Rinke, Hong Jiang (Peking University), Xinguo Ren (University of Science and Technology of China), Angel Rubio).

V.5 b Van der Waals Interactions in Ionic and Semiconductor Solids

How much do van der Waals interactions contribute to the cohesive properties of ionic and semiconductor solids? The answer to this fundamental question has remained elusive for a long time. We have developed an efficient method to determine vdW parameters for "atoms" in solids, and have assessed the relative contribution of vdW interactions to the lattice constant, cohesive energy, and bulk modulus for a wide variety of solids. While the vdW energy is clearly not the most relevant contribution in many solids, its accurate inclusion in semi-local and hybrid DFT functionals leads to significantly improved performance of these functionals when comparing to reliable experimental data for the cohesive properties of solids (Guo-Xu Zhang, Alexandre Tkatchenko).

V.5 c Consistent Set of Band Parameters and Deformation Potentials for the Group-III Nitrides and Group-II Oxides

We have derived consistent sets of band parameters and strain deformation potentials for the group-III nitrides (AlN, GaN and InN) and the group-II oxides (MgO, ZnO and CdO) in the wurtzite phase. Consistency is achieved by using the HSE hybrid density functional for the deformation potentials and the G_0W_0 approach for the band structures. It is verified by computing selected deformation potentials in G_0W_0 and vice versa. The parameters are in good agreement with available experiments and can now be used in device simulations and to interpret experiments (Patrick Rinke, Qimin Yan, Anderson Janotti and Chris Van de Walle (UCSB), Manfred Winkelnkemper, and Dieter Bimberg (TU Berlin), Abdallah Qteish (Yarmouk University, Jordan), Jörg Neugebauer (MPI Düsseldorf)).

V.5 d Polymorphism in Ice at Ambient and High Pressures

It has long been recognized that water molecules interact with each other through a combination of electrostatic dipoles, hydrogen bonding, and vdW forces. However, the way in which the interplay of such interactions leads to the rich and complex phase diagram of ice remains far from being understood. Using diffusion quantum Monte Carlo (DMC), accurate lattice energies of several ice phases have been obtained. Then, employing the DFT+vdW approach, we show that the vdW contribution to the lattice energy increases monotonically from the ambient to the high pressure phases of ice. Conversely, the strength of the hydrogen bonds between the water molecules decreases as the pressure increases. This results in the finding that in the highest pressure phase one quarter of the entire lattice energy comes from vdW forces (Biswajit Santra, Alexandre Tkatchenko, Jiří Klimeš, Dario Alfè, Ben Slater, Angelos Michaelides (UCL), Roberto Car (Princeton University)).

V.5 e Gold Clusters at Finite Temperatures

Typical theoretical studies of clusters address the T=0 K structures and then focus on the lowest-energy isomer(s). We have performed, however, unbiased sampling of the free-energy landscape of Au_N clusters ($3 \leq N \leq 20$) by means of DFT (PBE+vdW)-based replica-exchange molecular dynamics. For many sizes of pristine as well as complexed gold clusters we find a fluxional, and for some sizes, even a liquid character at room temperature. The calculations also reveal that anharmonicity as well as the presence of adsorbed noble gas atoms can have significant effects on IR spectra. The bonding between rare gas (RG) atoms and small gold clusters has been analyzed and found to be due to an overlap of the *p* orbitals of the RG with *s-d* orbitals of the gold clusters, which leads to the depletion of the electron density in the region between the gold atoms. This reduces the electron-electron repulsion between the gold atoms and effectively strengthens the Au-Au bonds (their length is shortened and the stretching vibrational frequency is increased) (Luca M. Ghiringhelli, Sergey V. Levchenko, Phillipp Gruene, Jonathan T. Lyon, André Fielicke, and Gerard Meijer (MP Department)).

V.5 f Sustainability and Resource Depletion

The transformation of the energy system in Germany (known as the *Energiewende*) involves a substantial increase in the generation of renewable energy. It is also expected to create a strong demand for many raw materials, some of which are already regarded as scarce. Examples are lithium and cobalt for batteries, rare earth elements for wind turbines, as well as cadmium, tellurium and indium for solar cells. Does this mean that regenerative energy systems are not necessarily sustainable? And what does “sustainable” actually mean when applied to mineral resources? Within the framework of the project we examine the concept of “sustainability” in general and the possible role played by resource depletion. Particular emphasis is given to raw materials required for the new energy economy and for electric vehicles. (Alex M. Bradshaw, Benjamin Reuter and Thomas Hamacher (TU Munich)).

V.5 g History of Quantum Field Theoretical Methods in Many-Body Physics: The Story of RPA

In the two decades after World War II, the quantum theory of many-body systems underwent a major transformation by extending concepts and techniques originating from quantum electrodynamics (QED) to the study of solids and nuclei. Since the late 1950s, quantum field theoretical techniques (in particular Feynman-Dyson diagrammatic perturbation theory) have become commonplace in condensed-matter theory. In order to gain a better understanding of this process of knowledge transfer, we contextualize a particular thread of this development: the story of the random-phase approximation (RPA), a perturbative technique which is of particular importance even today (Alexander S. Blum (MPIWG), Christian Joas (LMU Munich)).

Publications of the Theory Department

2011 (late publications)

Appel, H. and M. Di Ventra: Stochastic quantum molecular dynamics for finite and extended systems. Chemical Physics Letters **391** (1), 27-36 (2011).

Araghi, R.R., C. Baldauf, U.I.M. Gerling, C.D. Cadicamo and B. Koksch: A systematic study of fundamentals in alpha-helical coiled coil mimicry by alternating sequences of beta- and gamma-amino acids. Amino Acids **41** (3), 733-742 (2011).

Arndt, S., G. Laugel, S.V. Levchenko, R. Horn, M. Baerns, M. Scheffler, R. Schlögl and R. Schomäcker: A Critical Assessment of Li/MgO-Based Catalysts for the Oxidative Coupling of Methane. Catalysis Reviews: Science and Engineering **53** (4), 424-514 (2011).

Badino, M.: Mechanistic slumber vs. statistical insomnia: The early history of Boltzmann's H-theorem (1868-1877). The European Physical Journal H **36** (3), 353-378 (2011).

Beret, E.C., L.M. Ghiringhelli and M. Scheffler: Free gold clusters: Beyond the static, monostructure description. Faraday Discussions **152** (1), 153-167 (2011).

Bradshaw, A.M., T. Hamacher and U. Fischer: Is nuclear fusion a sustainable energy form? Fusion Engineering and Design **86** (9-11), 2770-2773 (2011).

Carlsson, J.M., L.M. Ghiringhelli and A. Fasolino: Theory and hierarchical calculations of the structure and energetics of [0001] tilt grain boundaries in graphene. Physical Review B **84** (16), 165423 (2011).

Cudazzo, P., M. Gatti, F. Roth, B. Mahns, M. Knupfer and A. Rubio: Plasmon dispersion in molecular solids: Picene and potassium-doped picene. Physical Review B **84** (15), 155118 (2011).

Cudazzo, P., I.V. Tokatly and A. Rubio: Dielectric screening in two-dimensional insulators: Implications for excitonic and impurity states in graphane. Physical Review B **84** (8), 085406 (2011).

Freund, H.-J., G. Meijer, M. Scheffler, R. Schlögl and M. Wolf: CO oxidation as a prototypical reaction for heterogeneous processes. Angewandte Chemie International Edition **50** (43), 10064-10094 (2011).

Freund, H.-J., G. Meijer, M. Scheffler, R. Schlögl and M. Wolf: Die CO-Oxidation als Modellreaktion für heterogene Prozesse. Angewandte Chemie **123** (43), 10242-10275 (2011).

Fuks, J.I., N. Helbig, I.V. Tokatly and A. Rubio: Nonlinear phenomena in time-dependent density-functional theory: What Rabi oscillations can teach us. Physical Review B **84** (7), 075107 (2011).

Helbig, N., J.I. Fuks, M. Casula, M.J. Verstraete, M.A.L. Marques, I.V. Tokatly and A. Rubio: Density functional theory beyond the linear regime: Validating an adiabatic local density approximation. Physical Review A **83** (3), 032503 (2011).

Helbig, N., J.I. Fuks, I.V. Tokatly, H. Appel, E.K.U. Gross and A. Rubio: Time-dependent density-functional and reduced density-matrix methods for few electrons: Exact versus adiabatic approximations. Chemical Physics **391** (1), 1-10 (2011).

Hu, X.L., J. Carrasco, J. Klimes and A. Michaelides: Trends in water monomer adsorption and dissociation on flat insulating surfaces. Physical Chemistry Chemical Physics **13** (27), 12447-12453 (2011).

Huix-Rotllant, M., A. Ipatov, A. Rubio and M.E. Casida: Assessment of dressed time-dependent density-functional theory for the low-lying valence states of 28 organic chromophores. Chemical Physics **391** (10), 120-129 (2011).

Joas, C. and S. Katzir: Analogy, extension, and novelty: Young Schrödinger on electric phenomena in solids. Studies in History and Philosophy of Science B **42** (1), 43-53 (2011).

Joas, C. and G. Waysand: Von Leitungsketten zur Paarhypothese: Die Entdeckung der Supraleitung und die wechselvolle Geschichte ihrer Erklärung. Physik Journal **10** (6), 23-30 (2011).

Lew, W., M.C. Crowe, C.T. Campbell, J. Carrasco and A. Michaelides: The Energy of Hydroxyl Coadsorbed with Water on Pt(111). The Journal of Physical Chemistry C **115** (46), 23008-23012 (2011).

Malic, E., C. Weber, M. Richter, V. Atalla, T. Klamroth, P. Saalfrank, S. Reich and A. Knorr: Microscopic Model of the Optical Absorption of Carbon Nanotubes Functionalized with Molecular Spiropyran Photoswitches. Physical Review Letters **106** (9), 097401 (2011).

Marom, N., J.E. Moussa, X. Ren, A. Tkatchenko and J.R. Chelikowsky: Electronic structure of dye-sensitized TiO_2 clusters from many-body perturbation theory. Physical Review B **84** (24), 245115 (2011).

Marom, N., X. Ren, J.E. Moussa, J.R. Chelikowsky and L. Kronik: Electronic structure of copper phthalocyanine from G_0W_0 calculations. Physical Review B **84** (19), 195143 (2011).

Marom, N., A. Tkatchenko, S. Kapishnikov, L. Kronik and L. Leiserowitz: Structure and Formation of Synthetic Hemozoin: Insights From First-Principles Calculations. Crystal Growth & Design **11** (8), 3332-3341 (2011).

Marom, N., A. Tkatchenko, M. Rossi, V. Gobre, O. Hod, M. Scheffler and L. Kronik: Dispersion Interactions with Density-Functional Theory: Benchmarking Semiempirical and Interatomic Pairwise Corrected Density Functionals. Journal of Chemical Theory and Computation **7** (12), 3944-3951 (2011).

Ropo, M., K. Kokko, E. Airiskallio, M.P.J. Punkkinen, S. Hogmark, J. Kollar, B. Johansson and L. Vitos: First-principles atomistic study of surfaces of Fe-rich Fe–Cr. Journal of Physics: Condensed Matter **23** (26), 265004 (2011).

Santra, B., J. Klimes, D. Alfè, A. Tkatchenko, B. Slater, A. Michaelides, R. Car and M. Scheffler: Hydrogen Bonds and van der Waals Forces in Ice at Ambient and High Pressures. Physical Review Letters **107** (18), 185701 (2011).

Scipioni, R., D. Donadio, L.M. Ghiringhelli and L. Delle Site: Proton Wires via One-Dimensional Water Chains Adsorbed on Metallic Steps. Journal of Chemical Theory and Computation **7** (9), 2681-2684 (2011).

Stella, L., C. Attaccalite, S. Sorella and A. Rubio: Strong electronic correlation in the hydrogen chain: A variational Monte Carlo study. Physical Review B **84** (24), 245117 (2011).

Zhang, G.-X., A. Tkatchenko, J. Paier, H. Appel and M. Scheffler: Van der Waals Interactions in Ionic and Semiconductor Solids. Physical Review Letters **107** (24), 245501 (2011).

Doctoral Thesis

Rossi, M.: Ab initio study of alanine-based polypeptide secondary-structure motifs in the gas phase. Technische Universität Berlin 2011.

2012

Andrade, X., J. Alberdi-Rodriguez, D.A. Strubbe, M.J.T. Oliveira, F. Nogueira, A. Castro, J. Muguerza, A. Arruabarrena, S.G. Louie, A. Aspuru-Guzik, A. Rubio and M.A.L. Marques: Time-dependent density-functional theory in massively parallel computer architectures: the OCTOPUS project. Journal of Physics: Condensed Matter **24** (23), 233202 (2012).

Arion, T., R. Püttner, C. Lupulescu, R. Ovsyannikov, M. Förstel, G. Öhrwall, A. Lindblad, K. Ueda, S. Svensson, A.M. Bradshaw, W. Eberhardt and U. Hergenhahn: New insight into the Auger decay process in O_2: The coincidence perspective. Journal of Electron Spectroscopy and Related Phenomena **185** (8-9), 234-243 (2012).

Baldauf, C. and H.-J. Hofmann: Ab initio MO Theory – An Important Tool in Foldamer Research: Prediction of Helices in Oligomers of omega-Amino Acids. Helvetica Chimica Acta **95** (12), 2348-2383 (2012).

Bhattacharya, S., A. Bhattacharya and G.P. Das: Anti-Kubas Type Interaction in Hydrogen Storage on a Li Decorated BHNH Sheet: A First-Principles Based Study. The Journal of Physical Chemistry C **116** (5), 3840-3844 (2012).

Bradshaw, A.M. and T. Hamacher: Nonregenerative Natural Resources in a Sustainable System of Energy Supply. ChemSusChem **5** (3), 550-562 (2012).

Carter, D.J., M. Fuchs and C. Stampfl: Vacancies in GaN bulk and nanowires: Effect of self-interaction corrections. Journal of Physics: Condensed Matter **24** (25), 255801 (2012).

Caruso, F., P. Rinke, X. Ren, M. Scheffler and A. Rubio: Unified description of ground and excited states of finite systems: The self-consistent GW approach. Physical Review B **86** (8), 081102(R) (2012).

Casadei, M., X. Ren, P. Rinke, A. Rubio and M. Scheffler: Density-Functional Theory for f-Electron Systems: The α-γ Phase Transition in Cerium. Physical Review Letters **109** (14), 146402 (2012).

Chen, J., S.A. Edwards, F. Gräter and C. Baldauf: On the Cis to Trans Isomerization of Prolyl-Peptide Bonds under Tension. The Journal of Physical Chemistry B **116** (31), 9346-9351 (2012).

Chutia, S., M. Rossi and V. Blum: Water Adsorption at Two Unsolvated Peptides with a Protonated Lysine Residue: From Self-Solvation to Solvation. The Journal of Physical Chemistry B **116** (51), 14788-14804 (2012).

Cudazzo, P., M. Gatti and A. Rubio: Excitons in molecular crystals from first-principles many-body perturbation theory: Picene versus pentacene. Physical Review B **86** (19), 195307 (2012).

Cudazzo, P., M. Gatti and A. Rubio: Plasmon dispersion in layered transition-metal dichalcogenides. Physical Review B **86** (7), 075121 (2012).

De Giovannini, U., D. Varsano, M.A.L. Marques, H. Appel, E.K.U. Gross and A. Rubio: Ab initio angle- and energy-resolved photoelectron spectroscopy with time-dependent density-functional theory. Physical Review A **85** (6), 062515 (2012).

De Oteyza, D.G., A. Sakko, A. El-Sayed, E. Goin, L. Floreano, A. Cossaro, J.M. Garcia-Lastra, A. Rubio and J.E. Ortega: Inversed linear dichroism in F K-edge NEXAFS spectra of fluorinated planar aromatic molecules. Physical Review B **86** (7), 075469 (2012).

DiStasio, R.A., O.A. von Lilienfeld and A. Tkatchenko: Collective many-body van der Waals interactions in molecular systems. Proceedings of the National Academy of Sciences of the United States of America **109** (37), 14791-14795 (2012).

Donadio, D., L.M. Ghiringhelli and L. Delle Site: Autocatalytic and Cooperatively Stabilized Dissociation of Water on a Stepped Platinum Surface. Journal of the American Chemical Society **134** (46), 19217-19222 (2012).

Elliott, P., J.I. Fuks, A. Rubio and N.T. Maitra: Universal Dynamical Steps in the Exact Time-Dependent Exchange-Correlation Potential. Physical Review Letters **109** (26), 266404 (2012).

Franco, I. and P. Brumer: Electronic coherence dynamics in trans-polyacetylene oligomers. The Journal of Chemical Physics **136** (14), 144501 (2012).

Goikoetxea, I., J. Beltrán, J. Meyer, J.I. Juaristi, M. Alducin and K. Reuter: Non-adiabatic effects during the dissociative adsorption of O_2 at Ag(111)? A first-principles divide and conquer study. New Journal of Physics **14** (1), 013050 (2012).

Gundlach, L. and F. Willig: Ultrafast Photoinduced Electron Transfer at Electrodes: The General Case of a Heterogeneous Electron-Transfer Reaction. ChemPhysChem **13** (12), 2877-2881 (2012).

Hellgren, M., D. Rohr and E.K.U. Gross: Correlation potentials for molecular bond dissociation within the self-consistent random phase approximation. The Journal of Chemical Physics **136** (3), 034106 (2012).

Iori, F., M. Gatti and A. Rubio: Role of nonlocal exchange in the electronic structure of correlated oxides. Physical Review B **85** (11), 115129 (2012).

Jiang, H., R.I. Gómez-Abal, X.-Z. Li, C. Meisenbichler, C. Ambrosch-Draxl and M. Scheffler: FHI-gap: A GW code based on the all-electron augmented plane wave methid. Comp. Phys. Com. **184**, 348-366 (2012).

Jiang, H., P. Rinke and M. Scheffler: Electronic properties of lanthanide oxides from the GW perspective. Physical Review B **86** (12), 125115 (2012).

Kim, H.-J., A. Tkatchenko, J.-H. Cho and M. Scheffler: Benzene adsorbed on Si(001): The role of electron correlation and finite temperature. Physical Review B **85** (4), 041403 (2012).

Li, X., R. Gomez Abal, H. Jiang, C. Ambrosch-Draxl and M. Scheffler: Impact of widely used approximations to the G_0W_0 method: An all-electron perspective. New Journal of Physics **14** (2), 023006 (2012).

Liu, W., J. Carrasco, B. Santra, A. Michaelides, M. Scheffler and A. Tkatchenko: Benzene adsorbed on metals: Concerted effect of covalency and van der Waals bonding. Physical Review B **86** (24), 245405 (2012).

Liu, W., A.A. Savara, X. Ren, W. Ludwig, K.-H. Dostert, S. Schauermann, A. Tkatchenko, H.-J. Freund and M. Scheffler: Toward Low-Temperature Dehydrogenation Catalysis: Isophorone Adsorbed on Pd(111). The Journal of Physical Chemistry Letters **3** (5), 582-586 (2012).

Marom, N., F. Caruso, X. Ren, O.T. Hofmann, T. Körzdörfer, J.R. Chelikowsky, A. Rubio, M. Scheffler and P. Rinke: Benchmark of GW methods for azabenzenes. Physical Review B **86** (24), 245127 (2012).

McCullagh, M., I. Franco, M.A. Ratner and G.C. Schatz: Defects in DNA: Lessons from Molecular Motor Design. The Journal of Physical Chemistry Letters **3** (6), 689-693 (2012).

Michealides, A. and M. Scheffler: An Introduction to the Theory of Crystalline Elemental Solids and their Surfaces. In: Surface and Interface Science: Concepts and Methods, Vol. 1. (Ed.) K. Wandelt. Wiley-VCH, Weinheim, 13-72 (2012). ISBN 978-3-527-41156-6.

Mucke, M., M. Förstel, T. Lischke, T. Arion, A.M. Bradshaw and U. Hergenhahn: Performance of a Short “Magnetic Bottle” Electron Spectrometer. Rev. Sci. Instrum. **83**, 063106 (2012).

Paier, J., X. Ren, P. Rinke, G.E. Scuseria, A. Grüneis, G. Kresse and M. Scheffler: Assessment of correlation energies based on the random-phase approximation. New Journal of Physics **14** (4), 043002 (2012).

Paz Borbón, L.O., G. Barcaro, A. Fortunelli and S.V. Levchenko: Au_N clusters ($N = 1$–6) supported on MgO(100) surfaces: Effect of exact exchange and dispersion interactions on adhesion energies. Physical Review B **85** (15), 155409 (2012).

Pozun, Z.D., K. Hansen, D. Sheppard, M. Rupp, K.-R. Müller and G. Henkelman: Optimizing transition states via kernel-based machine learning. The Journal of Chemical Physics **136** (17), 174101 (2012).

Ramprasad, R., H. Zhu, P. Rinke and M. Scheffler: New Perspective on Formation Energies and Energy Levels of Point Defects in Nonmetals. Physical Review Letters **108** (6), 066404 (2012).

Ren, X., P. Rinke, V. Blum, J. Wieferink, A. Tkatchenko, A.G. Sanfilippo, K. Reuter and M. Scheffler: Resolution-of-identity approach to Hartree–Fock, hybrid density functionals, RPA, MP2 and GW with numeric atom-centered orbital basis functions. New Journal of Physics **14** (5), 053020 (2012).

Ren, X., P. Rinke, C. Joas and M. Scheffler: Random-phase approximation and its applications in computational chemistry and materials science. Journal of Materials Science **47** (21), 7447-7471 (2012).

Reuter, K.: First-principles kinetic Monte Carlo simulations for heterogeneous catalysis: Concepts, status, and frontiers. In: Modeling and Simulation of Heterogeneous Catalytic Reactions: From the Molecular Process to the Technical System, (Ed.) O. Deutschmann. Wiley-VCH, Weinheim, 71-111 (2012). ISBN 978-3-527-32120-9.

Rinke, P., A. Schleife, E. Kioupakis, A. Janotti, C. Rödl, F. Bechstedt, M. Scheffler and C.G. Van de Walle: First-Principles Optical Spectra for F Centers in MgO. Physical Review Letters **108** (12), 126404 (2012).

Ruiz, V.G., W. Liu, E. Zojer, M. Scheffler and A. Tkatchenko: Density-Functional Theory with Screened van derWaals Interactions for the Modeling of Hybrid Inorganic-Organic Systems. Physical Review Letters **108** (14), 146103 (2012).

Rupp, M., A. Tkatchenko, K.-R. Müller and O.A. von Lilienfeld: Fast and Accurate Modeling of Molecular Atomization Energies with Machine Learning. Physical Review Letters **108** (5), 058301 (2012).

Rupp, M., A. Tkatchenko, K.-R. Müller and O.A. von Lilienfeld: Reply to Comment on "Fast and Accurate Modeling of Molecular Atomization Energies with Machine Learning". Physical Review Letters **109** (5), 059802 (2012).

Schatschneider, B., J.-J. Liang, S. Jezowski and A. Tkatchenko: Phase transition between cubic and monoclinic polymorphs of the tetracyanoethylene crystal: The role of temperature and kinetics. CrystEngComm (14), 4656-4663 (2012).

Skolidis, G., K. Hansen, G. Sanguinetti and M. Rupp: Multi-task learning for pK_a prediction. Journal of Computer-Aided Molecular Design **26** (7), 883-895 (2012).

Tkatchenko, A., D. Alfè and K.S. Kim: First-Principles Modeling of Non-Covalent Interactions in Supramolecular Systems: The Role of Many-Body Effects. Journal of Chemical Theory and Computation **8** (11), 4317-4322 (2012).

Tkatchenko, A., R.A. DiStasio, R. Car and M. Scheffler: Accurate and Efficient Method for Many-Body van der Waals Interactions. Physical Review Letters **108** (23), 236402 (2012).

Yan, Q., A. Janotti, M. Scheffler and C.G. Van de Walle: Role of nitrogen vacancies in the luminescence of Mg-doped GaN. Applied Physics Letters **100** (14), 142110 (2012).

Yan, Q., P. Rinke, M. Winkelnkemper, A. Qteish, D. Bimberg, M. Scheffler and C.G. Van de Walle: Strain effects and band parameters in MgO, ZnO, and CdO. Applied Physics Letters **101** (15), 152105 (2012).

Zhu, H., Y. Xu, B.-L. Gu and W. Duan: Robust linear dependence of thermal conductance on radial strain in carbon nanotubes. New Journal of Physics **14** (1), 013053 (2012).

Doctoral Theses

Gruber, M.: The selective catalytic reduction of NO by NH_3 at Brønsted and Lewis acid sites of vanadium oxide surfaces: Density functional theory studies. Freie Universität Berlin 2012.

Mangold, C.: Understanding catalytic processes and their selectivity from first principles: the oxidation of ammonia. Freie Universität Berlin 2012.

Meyer, J.: Ab initio modeling of energy dissipation during chemical reactions at transition metal surfaces. Freie Universität Berlin 2012.

Yan, Q.: Theoretical Study of Material and Device Properties of Group-III Nitrides. University of California Santa Barbara 2012.

2013

Agrawal, P., A. Tkatchenko and L. Kronik: Pair-Wise and Many-Body Dispersive Interactions Coupled to an Optimally Tuned Range-Separated Hybrid Functional. Journal of Chemical Theory and Computation **9** (8), 3473-3478 (2013).

Ambrosetti, A., F. Ancilotto and P.L. Silvestrelli: Van der Waals-Corrected Ab Initio Study of Water Ice–Graphite Interaction. The Journal of Physical Chemistry C **117** (1), 321-325 (2013).

*Ambrosetti, A., D. Alfè, R.A. DiStasio Jr. and A. Tkatchenk*o: Hard Numbers for Large Molecules: Towards Exact Energetics for Supramolecular Systems. Submitted to J. Phys. Chem. Lett. (December 08, 2013).

Ambrosetti, A., A.M. Reilly, R.A. DiStasio Jr., and A. Tkatchenko: Long-range correlation energy calculated from coupled atomic response functions. Submitted to J. Chem. Phys. (December 11, 2013).

Atalla, V., M. Yoon, F. Caruso, P. Rinke and M. Scheffler: Hybrid density functional theory meets quasiparticle calculations: A consistent electronic structure approach. Physical Review B **88** (16), 165122 (2013).

Attaccalite, C., L. Wirtz, A. Marini and A. Rubio: Efficient Gate-tunable light-emitting device made of defective boron nitride nanotubes: from ultraviolet to the visible. Scientific Reports **3**, 2698 (2013).

Baldauf, C., K. Pagel, S. Warnke, G. von Helden, B. Koksch, V. Blum and M. Scheffler: How Cations Change Peptide Structure. Chemistry - A European Journal **19** (34), 11224-11234 (2013).

Beret, E.C., M. van Wijk and L.M. Ghiringhelli: Reaction cycles and poisoning in catalysis by gold clusters: a thermodynamics approach. International Journal of Quantum Chemistry, in press.

Bhattacharya, S., S.V. Levchenko, L.M. Ghiringhelli and M. Scheffler: Stability and metastability of clusters in a reactive atmosphere: Theoretical evidence for unexpected stoichiometries of Mg_MO_x. Physical Review Letters **111** (13), 135501 (2013).

Bradshaw, A.M. and T. Hamacher: Nuclear fusion and the helium supply problem. Fusion Engineering and Design **88** (9-10), 2694-2697 (2013).

Bradshaw, A.M., B. Reuter and T. Hamacher: The potential scarcity of rare elements for the Energiewende. Green **3** (2), 93-111 (2013).

Bürker, C., N. Ferri, A. Tkatchenko, A. Gerlach, J. Niederhausen, T. Hosokai, S. Duhm, J. Zegenhagen, N. Koch and F. Schreiber: Exploring the bonding of large hydrocarbons on noble metals: Diindoperylene on Cu(111), Ag(111), and Au(111). Physical Review B **87** (16), 165443 (2013).

Caruso, F., P. Rinke, X. Ren, A. Rubio and M. Scheffler: Self-consistent GW: An all-electron implementation with localized basis functions. Physical Review B **88** (7), 075105 (2013).

Caruso, F., D. Rohr, M. Hellgren, X. Ren, P. Rinke, A. Rubio and M. Scheffler: Bond Breaking and Bond Formation: How Electron Correlation is Captured in Many-Body Perturbation Theory and Density-Functional Theory. Physical Review Letters **110** (14), 146403 (2013).

Cho, Y., S.K. Min, J. Yun, W.Y. Kim, A. Tkatchenko and K.S. Kim: Noncovalent Interactions of DNA Bases with Naphthalene and Graphene. Journal of Chemical Theory and Computation **9** (4), 2090-2096 (2013).

De Giovannini, U., G. Brunetto, A. Castro, J. Walkenhorst and A. Rubio: Simulating Pump-Probe Photoelectron and Absorption Spectroscopy on the Attosecond Timescale with Time-Dependent Density Functional Theory. ChemPhysChem **14** (7), 1363-1376 (2013).

Delle Site, L., L.M. Ghiringhelli and D.M. Ceperley: Electronic energy functionals: Levy–Lieb principle within the ground state path integral quantum Monte Carlo. International Journal of Quantum Chemistry **113** (2), 155-160 (2013).

Egger, D.A., V.G. Ruiz, W.A. Saidi, T. Bučko, A. Tkatchenko and E. Zojer: Understanding Structure and Bonding of Multilayered Metal–Organic Nanostructures. The Journal of Physical Chemistry C **117** (6), 3055-3061 (2013).

Franco, I. and H. Appel: Reduced purities as measures of decoherence in many-electron systems. The Journal of Chemical Physics **139** (9), 094109 (2013).

Franco, I., A. Rubio and P. Brumer: Long-lived oscillatory incoherent electron dynamics in molecules: Trans-polyacetylene oligomers. New Journal of Physics **15** (4), 043004 (2013).

Fuks, J.I., P. Elliott, A. Rubio and N.T. Maitra: Dynamics of Charge-Transfer Processes with Time-Dependent Density Functional Theory. The Journal of Physical Chemistry Letters **4** (5), 735-739 (2013).

Gao, W. and A. Tkatchenko: Electronic Structure and van der Waals Interactions in the Stability and Mobility of Point Defects in Semiconductors. Physical Review Letters **111** (4), 045501 (2013).

Ghiringhelli, L.M.: Application of (Kohn-Sham) Density-Functional Theory to Real Materials. In: Many-Electron Approaches in Physics, Chemistry and Mathematics: A Multidisciplinary View, (Eds.) V. Bach and L. Delle Site. Springer, in press

Ghiringhelli, L.M., P. Gruene, J.T. Lyon, D.M. Rayner, G. Meijer, A. Fielicke and M. Scheffler: Not so loosely bound rare gas atoms: finite-temperature vibrational fingerprints of neutral gold-cluster complexes. New Journal of Physics **15** (8), 083003 (2013).

Gobre, V.V. and A. Tkatchenko: Scaling laws for van der Waals interactions in nanostructured materials. Nature Communications **4**, 2341 (2013).

Guimond, S., D. Göbke, J.M. Sturm, Y. Romanyshyn, H. Kuhlenbeck, M. Cavalleri and H.-J. Freund: Well-Ordered Molybdenum Oxide Layers on Au(111): Preparation and Properties. The Journal of Physical Chemistry C **117** (17), 8746-8757 (2013).

Hamacher, T., M. Huber, J. Dorfner, K. Schaber, and A.M. Bradshaw: Nuclear Fusion and Renewable Energy Forms: Are they Compatible? Fusion Engineering Design **88**, 657-660 (2013).

Hansen, K., G. Montavon, F. Biegler, S. Fazli, M. Rupp, M. Scheffler, O.A. von Lilienfeld, A. Tkatchenko and K.-R. Müller: Assessment and Validation of Machine Learning Methods for Predicting Molecular Atomization Energies. Journal of Chemical Theory and Computation **9** (8), 3404-3419 (2013).

Heine, N., M.R. Fagiani, M. Rossi, T. Wende, G. Berden, V. Blum and K.R. Asmis: Isomer-Selective Detection of Hydrogen-Bond Vibrations in the Protonated Water Hexamer. Journal of the American Chemical Society **135** (22), 8266-8273 (2013).

Hofmann, O.T., J.-C. Deinert, Y. Xu, P. Rinke, J. Stähler, M. Wolf and M. Scheffler: Large work function reduction by adsorption of a molecule with a negative electron affinity: Pyridine on ZnO(10-10). The Journal of Chemical Physics **139** (17), 174701 (2013).

*Hofmann, O.T., N. Moll, P. Rinke, and M. Scheffler:*Interface dipoles of organic molecules on Ag(111) in hybrid density-funktional theory. Submitted to New J. Phys. (October 2, 2013).

Jiang, H., R. Gomez Abal, X. Li, C. Meisenbichler, C. Ambrosch-Draxl and M. Scheffler: FHI-gap: A GW code based on the all-electron augmented plane wave method. Computer Physics Communications **184** (2), 348-366 (2013).

Kendelewicz, T., S. Kaya, J.T. Newberg, H. Bluhm, N. Mulakaluri, W. Moritz, M. Scheffler, A. Nilsson, R. Pentcheva and G.E. Brown: X-ray Photoemission and Density Functional Theory Study of the Interaction of Water Vapor with the Fe_3O_4(001) Surface at Near-Ambient Conditions. The Journal of Physical Chemistry C **117** (6), 2719-2733 (2013).

Kioupakis, E., P. Rinke, A. Janotti, Q. Yan and C.G. Van de Walle: Energy Conversion: Solid-State Lighting. In: Computational Approaches to Energy Materials, (Eds.) A. Walsh, A.A. Sokol and C.R.A. Catlow. Wiley, Oxford, 231-260 (2013). ISBN 978-1-119-95093-6.

Liu, W., S. Filimonov, J. Carrasco and A. Tkatchenko: Molecular switches from benzene derivatives adsorbed on metal surfaces. Nature Communications **4**, 2569 (2013).

Liu, W., V.G. Ruiz, G.-X. Zhang, B. Santra, X. Ren, M. Scheffler and A. Tkatchenko: Structure and energetics of benzene adsorbed on transition-metal surfaces: Density-functional theory with van der Waals interactions including collective substrate response. New Journal of Physics **15** (5), 053046 (2013).

Marom, N., R.A. DiStasio, V. Atalla, S.V. Levchenko, A. Reilly, J.R. Chelikowsky, L. Leiserowitz and A. Tkatchenko: Many-Body Dispersion Interactions in Molecular Crystal Polymorphism. Angewandte Chemie International Edition **52** (26), 6629-6632 (2013).

Marom, N., R.A. DiStasio, V. Atalla, S.V. Levchenko, A. Reilly, J.R. Chelikowsky, L. Leiserowitz and A. Tkatchenko: Many-Body Dispersion Interactions in Molecular Crystal Polymorphism. Angewandte Chemie **125** (26), 6761-6764 (2013).

Mercurio, G., R.J. Maurer, W. Liu, S. Hagen, F. Leyssner, P. Tegeder, J. Meyer, A. Tkatchenko, S. Soubatch, K. Reuter and F.S. Tautz: Quantification of finite-temperature effects on adsorption geometries of π-conjugated molecules: Azobenzene/Ag(111). Physical Review B **88** (3), 035421 (2013).

Moll, N., Y. Xu, O.T. Hofmann and P. Rinke: Stabilization of semiconductor surfaces through bulk dopants. New Journal of Physics **15** (8), 083009 (2013).

Montavon, G., M. Rupp, V. Gobre, A. Vazquez-Mayagoitia, K. Hansen, A. Tkatchenko, K.-R. Müller and O.A. von Lilienfeld: Machine Learning of Molecular Electronic Properties in Chemical Compound Space. New Journal of Physics **15** (9), 095003 (2013).

Nemec, L., V. Blum, P. Rinke and M. Scheffler: Thermodynamic equilibrium conditions of graphene films on SiC. Physical Review Letters **111** (6), 065502 (2013).

Pitkänen, H., M. Alatalo, A. Puisto, M. ropo, K. Kokko and L. Vitos: Ab initio study of the surface properties of austenitic stainless steel alloys. Surface Science **609**, 190-194 (2013).

Reilly, A. and A. Tkatchenko: Seamless and Accurate Modeling of Organic Molecular Materials. The Journal of Physical Chemistry Letters **4** (6), 1028-1033 (2013).

Reilly, A. and A. Tkatchenko: Understanding the role of vibrations, exact exchange, and many-body van der Waals interactions in the cohesive properties of molecular crystals. The Journal of Chemical Physics **139** (2), 024705 (2013).

Ren, X., P. Rinke, G.E. Scuseria and M. Scheffler: Renormalized second-order perturbation theory for the electron correlation energy: Concept, implementation, and benchmarks. Physical Review B **88** (3), 035120 (2013).

Richter, N., S. Sicolo, S.V. Levchenko, J. Sauer and M. Scheffler: Concentration of Vacancies at Metal Oxide Surfaces: Case Study of MgO(100). Physical Review Letters **111** (4), 045502 (2013).

Rossi, M., M. Scheffler and V. Blum: Impact of Vibrational Entropy on the Stability of Unsolvated Peptide Helices with Increasing Length. The Journal of Physical Chemistry B **117** (18), 5574-5584 (2013).

Rossi, M., A. Tkatchenko, S.B. Rempe and S. Varma: The role of methyl–induced polarization in ion binding. Proceedings of the National Academy of Sciences of the United States of America **110** (32), 12978–12983 (2013).

Rozzi, C.A., S.M. Falke, N. Spallanzani, A. Rubio, E. Molinari, D. Brida, M. Maiuri, G. Cerullo, H. Schramm, J. Christoffers and C. Lienau: Quantum coherence controls the charge separation in a prototypical artificial light-harvesting system. Nature Communications **4**, 1602 (2013).

Santra, B., J. Klimeš, A. Tkatchenko, D. Alfé, B. Slater, A. Michaelides, R. Car and M. Scheffler: On the Accuracy of van der Waals Inclusive Density-Functional Theory Exchange-Correlation Functionals for Ice at Ambient and High Pressures. The Journal of Chemical Physics **139** (15), 154702 (2013).

Schatschneider, B., J.-J. Liang, A. Reilly, N. Marom, G.-X. Zhang and A. Tkatchenko: Electrodynamic response and stability of molecular crystals. Physical Review B **87** (6), 060104(R) (2013).

Schatschneider, B., S. Monaco, A. Tkatchenko and J.-J. Liang: Understanding the Structure and Electronic Properties of Molecular Crystals under Pressure: Application of Dispersion Corrected DFT to Oligoacenes. The Journal of Physical Chemistry A **117** (34), 8323-8331 (2013).

Schleife, A., P. Rinke, F. Bechstedt and C.G. Van de Walle: Enhanced Optical Absorption Due to Symmetry Breaking in $TiO_{2(1-x)}S_{2x}$ Alloys. The Journal of Physical Chemistry C **117** (8), 4189-4193 (2013).

Schlesinger, R., Y. Xu, O.T. Hofmann, S. Winkler, J. Frisch, J. Niederhausen, A. Vollmer, S. Blumstengel, F. Henneberger, P. Rinke, M. Scheffler and N. Koch: Controlling the work function of ZnO and the energy-level alignment at the interface to organic semiconductors with a molecular electron acceptor. Physical Review B **87** (15), 155311 (2013).

Schuler, B., W. Liu, A. Tkatchenko, N. Moll, G. Meyer, A. Mistry, D. Fox and L. Gross: Adsorption geometry determination of single molecules by atomic force microscopy. Physical Review Letters **111** (10), 106103 (2013).

Schumann, T., M. Dubslaff, M.H. Oliveira Jr, M. Hanke, F. Fromm,T. Seyller, L. Nemec, V. Blum, M. Scheffler, J.M. J Lopes and H. Riechert: Structural investigation of nanocrystalline graphene grown on $(6\sqrt{3}\times6\sqrt{3})$ R30°-reconstructed SiC surfaces by molecular beam epitaxy. Accepted to New J. Phys. (November 27, 2013).

Stähler, J., O.T. Hofmann, P. Rinke, S. Blumstengel, F. Henneberger, Y. Li and T.F. Heinz: Raman study of 2,7-bis(biphenyl-4-yl-)2',7'-ditertbutyl-9,9'-spirobifluorene adsorbed on oxide surfaces. Chemical Physics Letters **584** (1), 74-78 (2013).

Tkatchenko, A., A. Ambrosetti and R.A. DiStasio: Interatomic methods for the dispersion energy derived from the adiabatic connection fluctuation-dissipation theorem. The Journal of Chemical Physics **138** (7), 074106 (2013).

Wanko, M., T. Wende, M.M. Saralegui, L. Jiang, A. Rubio and K.R. Asmis: Solvent-mediated folding in dicarboxylate dianions: Aliphatic chain length dependence and origin of the IR intensity quenching. Physical Chemistry Chemical Physics, in press.

Xu Y., O.T. Hofmann, R. Schlesinger, S. Winkler, J. Frisch, J. Niederhausen, A. Vollmer, S. Blumstengel, F. Henneberger, N. Koch, P. Rinke and M. Scheffler: Space Charge Transfer in Hybrid Inorganic-Organic Systems. Phys. Rev. Lett. **111**, 226802 (2013).

Yu, J., M. Scheffler and H. Metiu: Oxidative Dehydrogenation of Methane by Isolated Vanadium Oxide Clusters Supported on Au (111) and Ag (111) Surfaces. The Journal of Physical Chemistry C **117** (36), 18475-18483 (2013).

Zhang I.Y., X. Ren, P. Rinke, V. Blum and M. Scheffler: Numeric atom-centered-orbital basis sets with valence-correlation consistency from H to Ar. Submitted to New J. Phys. (September 4, 2013).

Zheng, W., T.P. Cotter, P. Kaghazchi, T. Jacob, B. Frank, K. Schlichte, W. Zhang, D.S. Su, F. Schüth and R. Schlögl: Experimental and Theoretical Investigation of Molybdenum Carbide and Nitride as Catalysts for Ammonia Decomposition. Journal of the American Chemical Society **135** (9), 3458-3464 (2013).

Doctoral Theses

Atalla, V.: Density-functional theory and beyond for organic electronic materials. Technische Universität Berlin 2013.

Caruso, F.: Self-consistent GW approach for the unified description of ground and excited states of finite systems. Freie Universität Berlin 2013.

Casadei, M.: Density-Functional Theory for *f*-Electron Systems: The α-γ Phase Transition in Cerium. Freie Universität Berlin 2013.

Richter, N.: Charged point defects in oxides - a case study of MgO bulk and surface F centers. Technische Universität Berlin 2013.

Invited Talks of the Members of the Theory Department

Guillem Albareda Piquer

Jul 2013	International Workshop on Frontiers in Computational Material Science, Ringberg Castle, Germany *Bohmian Trajectories: A Complementary Computational Tool to Describe Nonadiabatic Molecular Dynamics*

Heiko Appel

Nov 2011	Theory Days on Stochastic and Dissipative Effects, Toulouse, France *Stochastic Unravelling of Interactions in Quantum Molecular Dynamics*
Nov 2011	Mini-Symposium: Memory Effects in Light Matter Coupling, Berlin, Germany *Markovian and Non-Markovian Stochastic Schrödinger Equations*
Jan 2012	5th International Workshop and School on Time-Dependent Density-Functional Theory: Prospects and Applications, Benasque, Spain *Electron-Ion Entanglement and Decoherence Dynamics in Trans-Polyacetylene Oligomers*
Jul 2012	International Workshop on Frontiers in Computational Material Science, Ringberg Castle, Germany *Vibrational Coupling and Real-Time Dynamics in Trans-Polyacetylene*
Nov 2012	Introductory Talk and Session Moderation at the CECAM Workshop on Vibrational Coupling: Most Important, often Ignored, and a Challenge for Ab Initio Theory, Lausanne, Switzerland *Non-Adiabatic Effects in Vibronically Coupled Systems*
Jan 2013	Seminar, Nano-Bio Spectroscopy Group, University of the Basques Country, San Sebastián, Spain *Wave Packet Revivals and Decoherence Dynamics in Vibronically Coupled Systems*
May 2013	Seminar, MPI of Microstructure Physics, Halle, Germany *Spontaneous Emission and Photon Wavepackets in Cavity QED*
Jul 2013	CECAM Workshop on Density Functional Theory: Learning from the Past, Looking to the Future, Berlin, Germany *Intra-System Derivative Discontinuities in Static DFT and Correlated Photon-Electron Wavefunctions in Cavity QED*
Aug 2013	Hands-on Workshop DFT and Beyond: Computational Materials Science for Real Materials, Trieste, Italy *Time-Dependent Density Functional Theory and Optical Properties*
Oct 2013	Advances in Time-Dependent Methods for Quantum Many-Body Systems, Trento, Italy *Wave Packet Revivals, Decoherence Dynamics and Photon Pulse Propagation in Coupled Phonon-Electron-Photon Systems*

Carsten Baldauf

Mar 2012	Seminar, Institute of Organic Chemistry and Biochemistry, Prague, Czech Republic *Paradocks - Molecular Docking with Population-Based Metaheuristics*
Mar 2013	Bio-Soft-Matter-Seminar, Department of Physics (AG Roland Netz), Freie Universität Berlin, Berlin, Germany *Formation of Protein Secondary Structure Motifs*
Oct 2013	Seminar, Department of Biological Sciences, Middle East Technical University, Ankara, Turkey *Modeling the Effect of Shear-Flow Induced Tensile Force on a Protein and How Cations Change Peptide Structure*

Björn Bieniek

Jul 2012	International Workshop on Frontiers in Computational Material Science, Ringberg Castle, Germany *Ultra-Thin ZnO Films on Metal Substrates from First Principle*

Volker Blum

Oct 2011	Theoretical Chemistry Seminar, Technische Universität Darmstadt, Darmstadt, Germany *Capturing Large Systems with First Principles: Pushing Some Limits of Electronic Structure Theory for Peptides and Surfaces*
Oct 2011	Conference on Computational Physics, Gatlinburg, TN, USA *Pushing Some Computational Limits with All-Electron Electronic Structure Theory: Eigenvalue Solvers, Surface Reconstruction, Biomolecular Structure*
Nov 2011	SMALL is Beautiful - Theory Meets Experiment, Berlin, Germany *State of the Art in Electronic Structure Codes*
Dec 2011	Theoretical Chemistry Seminar, University of Potsdam, Potsdam, Germany *Pushing Some Computational Limits with All-Electron Electronic Structure Theory*
Dec 2011	Seminar, Interdisciplinary Centre for Advanced Materials Simulations, Ruhr Universität Bochum, Bochum, Germany *Stretching the Limits of First Principles: Structure Prediction, Dynamics and Spectroscopy from Large Surfaces to BioMolecules*
Jan 2012	jDPG Theoretiker-Workshop, Burg Breuberg, Germany *Ab Initio Simulations of BioMolecules*
Jan 2012	Isolated Biomolecules and Biomolecular Interactions, Les Diablerets, Switzerland *Polyalanine-Based Secondary Structure in the Gas Phase: First-Principles Structure, Dynamics and Spectroscopy*

Jan 2012	BASF Modeller Lunch, BASF SE, GVC/S, Ludwigshafen, Germany *Pushing Some Computational Limits with All-Electron Electronic Structure Theory: FHI-Aims*
Mar 2012	Seminar, Pacific Northwest National Laboratory, Richland, WA, USA *Pushing Some Computational Limits with All-Electron Electronic Structure Theory: Surfaces and BioMolecules*
May 2012	IPAM Workshop on Computational Methods in High Energy Density Plasmas, IV: Computational Challenges in Warm Dense Matter, University of California, Los Angeles, Los Angeles, CA, USA *All Electron Ab Initio Molecular Simulations - Status, Successes, and Some Computational Challenges*
Jun 2012	I^{3}MS Seminar, Institute for Modelling and Simulation, RWTH Aachen University and Forschungszentrum Jülich, Aachen, Germany *The FHI-Aims Code: Experiences from a Large Scientific Software Project*
Jul 2012	Workshop on Efficient Localised Orbitals for Large Systems, Strong Correlations and Excitations, Cambridge, UK *Numeric Atom-Centered Basis Sets for All-Electron Ab Initio Simulations: The FHI-Aims Code, Surfaces, and Biomolecular Structure*
Jul 2012	IMS Seminar, Institute for Materials Science, University of Connecticut, Storrs, CT, USA *Predictive Theory for Real Materials and Molecules: Towards High Throughput for the Materials Genome*
Aug 2012	Seminar, Atomistic Simulation Centre, Queen's University, Belfast, UK *Pushing Some Limits with All-Electron Electronic Structure Theory: FHI-Aims, Graphene on SiC and Peptide Secondary Structure*
Sep 2012	11th European Conference on Surface Crystallography and Dynamics (ECSCD-11), Edinburgh, UK *How Far Can We Push First Principles Predictions Today? Accuracy, Large-Scale Surface Reconstructions and Graphene on SiC*
Sep 2012	29th European Conference on Surface Science (ECOSS-29), Edinburgh, UK *How Far Can We Push First Principles Predictions Today? Accuracy, Large-Scale Surface Reconstructions and Graphene on SiC*
Nov 2012	Seminar, Duke University, Durham, NC, USA *Predictive Theory and Modeling for the Materials Genome: FHI-Aims*
Jan 2013	Seminar, Department of Chemistry, Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany *All-Electron Electronic Structure Theory for Real Materials and Molecules*
Feb 2013	Seminar, Technische Universität Kaiserslautern, Kaiserslautern, Germany *Pushing Some Limits of Biomolecular Spectroscopy*
Mar 2013	Hume-Rothery Symposium, The Minerals, Metals and Materials Society, San Antonio, TX, USA *All-Electron Electronic Structure Accuracy for Real Materials and Molecules*

Apr 2013	Seminar, Competence Center of Computational Chemistry, ETH Zurich, Zurich, Switzerland *An Integrated Framework for Materials, Molecules and the Chemistry that They Control: FHI-Aims*
Jun 2013	Telluride Workshop on Thermal Transport at the Nanoscale, Telluride, CO, USA *Efficient, Accurate All-Electron Approach to Nanoscale Systems:FHI-Aims*
Jun 2013	Workshop on Stabilized Noble Metal Nanoparticles, Helsinki, Finland *First Principles for a Combinatorial Explosion: The Road to Biomolecular Structure, Spectroscopy, Dynamics*
Aug 2013	Hands-on Workshop DFT and Beyond: Computational Materials Science for Real Materials, Trieste, Italy *The Nuts and Bolts of Electronic Structure Theory*
Sep 2013	246th ACS National Meeting and Exposition, Indianapolis, IN, USA *Materials and Nanostructures, the All-Electron Way: FHI-aims*

Christian Carbogno

Aug 2012	FHI-Aims Developer's and User's Meeting on Density Functional Theory and Beyond with Numeric Atom-Centered Orbitals, Berlin, Germany *Heat Transport from First Principles in FHI-Aims*
Nov 2012	CECAM Workshop on Vibrational Coupling: Most Important, often Ignored, and a Challenge for Ab Initio Theory, Lausanne, Switzerland *Thermal Conductivity at High Temperatures from First Principles*
Dec 2012	Seminar, Theoretische Chemie, Technische Universität München, Munich, Germany *Thermal Conductivity at High Temperatures from First Principles*
Jun 2013	Telluride Workshop on Thermal Transport at the Nanoscale, Telluride, CO, USA *High Temperature Thermal Conductivity from First Principles*
Aug 2013	Hands-on Workshop DFT and Beyond: Computational Materials Science for Real Materials, Trieste, Italy *Charge and Heat Transport in Solids*

Fabio Caruso

Apr 2012	CECAM Workshop on GW Quasiparticle Calculations in Condensed Matter Physics and Nanoscience, Lausanne, Switzerland *Beyond One-Shot GW: Self-Consistency and Total Energy from GW*
Jul 2012	International Workshop on Frontiers in Computational Material Science, Ringberg Castle, Germany *Ground-State Properties from Many-Body Perturbation Theory: Is Self-Consistent GW Good Enough?*

Aug 2012	FHI-Aims Developer's and User's Meeting on Density Functional Theory and Beyond with Numeric Atom-Centered Orbitals, Berlin, Germany *Self-Consistent GW in FHI-Aims*
Dec 2012	Recent progress in Dynamical Mean-Field Theory and GW Calculations, and hands-on school on full-potential LMTO method, IPCMS, Strasbourg, France *Unified Description of Ground- and Excited-States: Fully Self-Consistent GW*
May 2013	ETSF Young Researchers Meeting, Budapest, Hungary *Introduction to the GW Approximation*

Marco Casadei

Jul 2012	International Workshop on Frontiers in Computational Material Science, Ringberg Castle, Germany *Unraveling the α-γ Phase Transition in Cerium from First Principle*

Ignacio Franco

Jan 2012	Seminar, Department of Chemistry, University of Rochester, Rochester, NY, USA *Single-Molecules Pulling: Fundamentals and Applications in Molecular Electronics and Molecular Motors*
Jan 2012	Seminar, European Theoretical Spectroscopy Facility, University of the Basque Country, San Sebastián, Spain *Single-Molecules Pulling: Fundamentals and Applications in Molecular Electronics and Molecular Motors*
Jan 2012	Spezialseminar, MPI of Quantum Optics, Garching, Germany *Optical Control of Currents Along Nanoscale Junctions*
Jun 2012	Cross Border Workshop on Laser Science 2012, McGill University, Montreal, Canada *Optical Control of Currents Along Nanoscale Junctions*
Aug 2012	Seminar, Department of Chemistry, Universidad del Valle, Cali, Colombia *Optical Control of Currents Along Nanoscale Junctions*
Aug 2012	Seminar, Department of Chemistry, Universidad Nacional de Colombia, Bogotá, Colombia *Single-Molecules Pulling: Fundamentals and Applications in Molecular Electronics and Molecular Motors*
Aug 2012	Seminar, Department of Chemistry, University of Antioquia, Medellín, Colombia *Single-Molecules Pulling: Fundamentals and Applications in Molecular Electronics and Molecular Motors*

Nov 2012	CECAM Workshop on Vibrational Coupling: Most Important, often Ignored, and a Challenge for Ab Initio Theory, Lausanne, Switzerland *Introduction to the Session on Excitations in Strong Fields and Time-Resolved Excitations*
Jan 2013	Theory Seminar, Chemistry Department, New York University, New York, NY, USA *Optical Control of Currents Along Nanoscale Junctions*
Jul 2013	Telluride Workshop on Non-Equilibrium Phenomena, Nonadiabatic Dynamics and Spectroscopy, Telluride, CO, USA *Electronic Decoherence and Coherent-Like Phenomena in Molecules: Trans-Polyacetylene*

Luca M. Ghiringhelli

Dec 2011	Workshop on the Physics and Chemistry of Energy Conversion at Interfaces, Dalian, China *Towards Catalysis by Gold Clusters: Allowed Reaction Cycles and Poisons*
Dec 2011	Workshop on Sustainable Energy, Taipei, Taiwan *Towards Catalysis by Gold Clusters: Allowed Reaction Cycles and Poisons*
Jan 2012	jDPG Theoretikerworkshop (Theorist's workshop of the Young German Physical Society), Burg Breuberg, Germany *Free-Energy Calculation for Molecules and Condensed Phases*
Mar 2012	Colloquium at Oak Ridge National Laboratory, Oak Ridge, TN, USA *Towards Catalysis by Gold Clusters: Reaction Cycles and Poisons*
Jun 2012	First PLUMED meeting, Trieste, Italy *Ab Initio Free Energy: Interface PLUMED / FHI-Aims and Application to Fluxional Metal Clusters and Beyond*
Jul 2012	International Workshop on Frontiers in Computational Material Science, Ringberg Castle, Germany *(Meta)Stability and Dynamics of Metal Clusters in Realistic Atmosphere from First Principles*
Dec 2012	Research Workshop and Graduate School on Complexity from quantum systems to emergent behaviour, Debrecen, Hungary *Levy-Lieb Constrained-Search and Classical Monte Carlo Sampling: Electron Correlations and Shannon Entropy*
Feb 2013	International Workshop on Computational Materials Design, Jodhpur, India *Ab Initio Atomistic Thermodynamics and Statistical Mechanics of Materials' Properties and Functions*
Mar 2013	Block Course on Aspects of Physics and Chemistry of Surfaces, International Max Planck Research School Complex Surfaces in Materials Sciences, Berlin, Germany *(Computational) Statistical Mechanics from First Principles*

Jul 2013	International Summer School on Basic Concepts and First-Principles Computations for Surface Science: Applications in Chemical Energy Conversion and Storage, Norderney, Germany *Treating Surfaces: Clusters vs. Supercells*
Aug 2013	Hands-on Workshop DFT and Beyond: Computational Materials Science for Real Materials, Trieste, Italy *From Ab Initio Molecular Dynamics to Statistical Mechanics*
Sep 2013	CECAM/Psi-k Research Conference:Multi-scale Modeling from First-Principles, Cap Roig, Spain *Free Metal and Metal-Oxide Clusters: Beyond the Static, Monostructure Description*
Sep 2013	Block Course on Dynamic Processes at Interfaces and Surfaces, International Max Planck Research School Complex Surfaces in Materials Sciences, Berlin, Germany *First-Principles Molecular Dynamics and Infrared Spectroscopy*
Oct 2013	IPAM Program on Materials for a Sustainable Energy Future, Los Angeles, CA, USA *Free Metal and Metal-Oxide Clusters: Beyond the Static, Monostructure Description*

Oliver T. Hofmann

Jul 2012	International Workshop on Frontiers in Computational Material Science, Ringberg Castle, Germany *Hybrid Functional Studies of Inorganic/Organic Systems*
Aug 2013	Hands-on Workshop DFT and Beyond: Computational Materials Science for Real Materials, International Centre for Theoretical Physics, Trieste, Italy *Density Functional Theory in Practise II*

Sergey V. Levchenko

Oct 2011	BIG-NSE Initial Phase WS 2011, Berlin, Germany *From Electronic Structure to Dynamics, Thermodynamics, and Reaction Kinetics: Multiscale Modelling of Materials and Interfaces*
Mar 2012	243rd ACS National Spring Meeting, San Diego, CA, USA *Electronic Structure and Thermodynamics of Li/MgO (100) and (111) Surfaces*
Oct 2012	2nd Surface Science School on Technology and Measurements at the Atomic Scale (SSS-TMAS II), Sochi, Russia *Theoretical Study of Chemical Surface Reactions*
Jul 2013	UniCat meeting on Oxidative coupling of methane, Berlin, Germany *First-Principles Description of Oxides at Realistic Conditions: Defects and Clusters*

Aug 2013	Hands-on Workshop DFT and Beyond: Computational Materials Science for Real Materials, Trieste, Italy *Beyond DFT for Periodic Systems*
Oct 2013	Seminar, Department of Mechanical Engineering and Materials Science, Duke University, Durham, NC, USA *Ab Initio Modelling of Defects at Semiconductor Surfaces: Doping as a Thermodynamic Factor*

Xinguo Ren

Nov 2011	SMALL is Beautiful: Theory Meets Experiment, Berlin, Germany *Approaches beyond DFT in Terms of Accuracy*
Jul 2012	International Workshop on Frontiers in Computational Material Science, Ringberg Castle, Germany *Renormalized Second-Order Perturbation Theory for Total and Self Energy: Concepts and Benchmarks*
Jul 2012	CECAM Workshop on Efficient Localised Orbitals for Large Systems, Strong Correlations and Excitations, University of Cambridge, Cambridge, UK *Beyond LDA and GGAs Using Numeric Atom-Centered Basis Functions*
Aug 2012	FHI-Aims Developer's and User's Meeting on Density Functional Theory and Beyond with Numeric Atom-Centered Orbitals, Berlin, Germany *Beyond RPA and GW: Renormalizated Second-Order Perturbation Theory for Ground-State and Excited-State Calculations*
Oct 2012	CECAM Workshop on Towards First-Principles Description of van der Waals Interactions in Complex Materials, Lausanne, Switzerland *Renormalized Second-Order Perturbation Theory for Weak Interactions*
Dec 2012	Recent Progress in Dynamical Mean-Field Theory and GW Calculations, and Hands-on School on Full-Potential LMTO Method, IPCMS, Strasbourg, France *Renormalized Second-Order Perturbation Theory for the Computation of Ground-State Total Energy and Self-Energy: Concept and Benchmarks*

Norina Richter

Oct 2013	IPAM Workshop on Materials for a Sustainable Energy Future, Workshop II: Fuels from Sunlight Concentration of Point Defects at Surfaces, Berlin, Germany *The Crucial Role of Temperature, Pressure, and Doping*

Patrick Rinke

Nov 2011	Recent Advancements in Density-Functional Theory: Transport and Strong Correlations, Strasbourg, France *Recent Progress in RPA and GW Calculation*

Jan 2012	Seminar, FHI, Berlin, Germany *Theoretical Spectroscopy-Success and Challenges for d- and f-Electron Systems*
Feb 2012	Towards Reality in Nanoscale Materials V, Levi, Finland *Towards a Unified Description of Ground and Excited State Properties: RPA vs GW*
Feb 2012	Seminar, Freie Universität Berlin, Berlin, Germany *Theoretical Spectroscopy - Success and Challenges for d- and f-Electron Systems*
Mar 2012	DPG-Frühjahrstagung, Dresden, Germany *Random Phase Approximation and GW for Correlated Systems*
May 2012	IPAM Workshop Computational Challenges in Warm Dense Matter, Los Angeles, CA, USA *Towards a Unified Description of Ground and Excited State Properties*
May 2012	Computational Oxides Spintronics Workshop, Cranage Hall, UK *Advanced DFT Functionals and GW for Correlated Systems*
May 2012	Seminar, York University, York, UK *Molecules on Surfaces - A Challenge for Electronic Structure Theory*
Aug 2012	Hands-on Workshop on Excitations, HoW exciting! Humboldt-Universität zu Berlin, Berlin, Germany *Towards a Unified Description of Ground and Excited State Properties: The GW approach*
Sep 2012	CRC-FHI Joint Meeting on Complex Surfaces in Material Science, Berlin, Germany *Organic Thin Films on ZnO Surfaces*
Sep 2012	HIOS Young Researcher Meeting, Wittensee, Germany *Theoretical Aspects of Hybrid Inorganic/Organic Systems*
Oct 2012	Group Seminar, Dalian Institute for Chemical Physics, Dalian, China *Towards a Unified Description of Ground and Excited State Properties: RPA vs GW*
Oct 2012	Colloquium, Dalian Institute for Chemical Physics, Dalian, China *Hybrid Inorganic/Organic Interfaces from a Semiconductor's Perspective*
Oct 2012	Seminar, Chemistry Department, Peking University, Beijing, China *Towards a Unified Description of Ground and Excited State Properties: RPA vs GW*
Oct 2012	Physics Colloquium, Tsinghua University, Beijing, China *Hybrid Inorganic/Organic Interfaces from a Semiconductor's Perspective*
Nov 2012	Physics Colloquium, University of Texas, Austin, TX, USA *Hybrid Inorganic/Organic Interfaces from a Semiconductor's Perspective*
Nov 2012	Seminar, Institute for Computational Engineering and Science, University of Texas, Austin, TX, USA

	Towards a Unified Description of Ground and Excited State Properties: RPA vs GW
Nov 2012	Seminar, IBM Research, Zurich, Switzerland *Hybrid Inorganic/Organic Interfaces from a Semiconductor's Perspective*
Jan 2013	16th International Workshop on Computational Physics and Materials Science: Total Energy and Force Methods, Trieste, Italy *Towards a Unified Description of Ground and Excited State Properties: GW vs RPA and Beyond*
Feb 2013	Seminar, Department of Materials, Imperial College, London, UK *Hybrid Inorganic/Organic Interfaces from a Semiconductor's Perspective*
Feb 2013	Seminar, Department of Materials, University of Oxford, Oxford, UK *Hybrid Inorganic/Organic Interfaces from a Semiconductor's Perspective*
Feb 2013	Seminar, Weizmann Institute, Rehovot, Israel *Hybrid Inorganic/Organic Interfaces from a Semiconductor's Perspective*
Feb 2013	Seminar, Weizmann Institute, Rehovot, Israel *Towards a Unified Description of Ground and Excited State Properties: RPA vs GW*
Apr 2013	Seminar, Center for Atomic Scale, Technical University of Denmark, Copenhagen, Denmark *Towards a Unified Description of Ground and Excited State Properties: RPA vs GW and Beyond*
May 2013	Seminar, Technische Universität München, Munich, Germany *Towards a Unified Description of Ground and Excited State Properties with View to Hybrid Organic/Inorganic Systems*
Jun 2013	Green's Function Methods: The Next Generation, Toulouse, France *Towards a Unified Description of Ground and Excited State Properties: GW vs RPA and Beyond*
Jun 2013	Stabilized Noble Metal Nanoparticles, Recent Advances and Present Challenges, Helsinki, Finland *Hybrid Systems from First Principles*
Sep 2013	Seminar, University of California, Santa Barbara, CA, USA *Towards a Unified Description of Ground and Excited States GW vs RPA and Beyond*

Mariana Rossi

Sep 2011	16th Workshop on Electronic Excitations, European Theoretical Spectroscopy Facility, Turin, Italy *Exploring the Conformations, Stability, and Dynamics of Helix-Forming Alanine-Based Polypeptides: First-Principles Predictions and Benchmarks*
Jul 2013	Seminar, Chemistry Department, Technische Universität München, Munich, Germany

	Understanding the (Thermodynamic) Stability of Isolated Polypeptides from First Principles
Aug 2012	Density Functional Theory and Beyond with Numeric Atom-Centered Orbitals, Berlin, Germany *Ab Initio Molecular Dynamics for Biomolecular Spectroscopy*
Aug 2013	Hands-on Workshop DFT and Beyond: Computational Materials Science for Real Materials, Trieste, Italy *Ab Initio Molecular Dynamics*

Angel Rubio

Oct 2011	Theorie-Kolloquium, Martin Luther University Halle-Wittenberg, Halle, Germany *Theoretical Spectroscopy: Modelling Photo-Induced Dynamical Processes in Nanostructures and Oxides*
Oct 2011	4th European School on Molecular Nanoscience (ESMOLNA2011), Peñíscola, Spain *Theoretical Concepts for the Simulation of Nanostructured-Based Devices: From Photovoltaics to Light-Emission*
Nov 2011	3rd Theory Days, Stochastic and Dissipative Effects, Toulouse, France *Non Equilibrium Dynamical Simulations of Complex Systems from TDDFT*
Dec 2011	Colloquium, Faculty of Physics, University of Duisburg-Essen, Duisburg, Germany *Modeling Optoelectronic Nanostructured Devices Made of Inorganic Nanotubes*
Feb 2012	Gordon Research Conference on Ultrafast Phenomena in Cooperative Systems, Understanding Complex Matter Far from Equilibrium and on Elementary Time Scales, Galveston, TX, USA *Ultrafast Response of Solids and Nanostructures Investigated via Many-Body Theory and Time-Dependent Density Functional Techniques*
Feb 2012	Symposium on Density Functional Theory for Chemical Physics, March Meeting of the American Physical Society, Division of Chemical Physics, Boston, MA, USA *TDDFT for Nonlinear Phenomena of Light-Matter Interactions*
May 2012	Colloquium, Materials Department of Physics, University of Oxford, Oxford, UK *Photodynamics of Nanostructures: A TDDFT Approach to the Weak and Strong Coupling Regimes*
Jun 2012	2nd TYC Workshop on Energy Materials, London, UK *TDDFT for Light-Matter Interactions in Strong Coupling Regimes*
Jun 2012	2nd International Conference on Advanced Materials Modelling (ICAMM), Institute of Materials Jean Rouxel, Université de Nantes, Nantes, France *Simulating Optoelectronic Devices at the Nanoscale: A TDDFT Perspective*

Jun 2012	Summer UPV/EHU Workshop on Concepts and Applications of Stimulus-responsive Materials, San Sebastián, Spain *Unraveling the Electronic Properties of Nanostructures and BioMolecules from Spectroscopy: A Theoretical Perspective*
Jul 2012	Colloquium, Center for Free-Electron Laser Science, Hamburg, Germany *Modelling Photo-Induced Dynamical Processes in Nanostructures and BioMolecules from First Principles: Correlation Effects and Applications*
Aug 2012	Hands-on Workshop on Excitations, HoW exciting! Humboldt-Universität zu Berlin, Berlin, Germany *Perspectives*
Sep 2012	CECAM Conference on Energy from the Sun: Computational Chemists and Physicists Take up the Challenge, Cagliari, Italy *Hybrid Organic Photovoltaics: From a Time-Dependent Density Functional Perspective*
Sep 2012	NANOWIRES12, Paul-Drude-Institute for Solid State Electronics, Berlin, Germany *Theoretical Concepts for the Simulation of Nanostructured-Based Devices: Towards Nanostructured-Based Optoelectronic Devices*
Oct 2012	Colloquium, Department of Chemistry, Technische Universität München, Munich, Germany *What Time-Dependent Density Functional Theory Can Provide to Understand and Design Nanostructured-Based Optoelectronics Materials*
Oct 2012	5th European School on Molecular Nanoscience (ESMOLNA2012), Cuenca, Spain *Electronic Properties of New Hybrids Made of BN and C*
Nov 2012	Colloquium of the Department of Physics, University of Luxembourg, Luxemburg, Luxemburg *Understanding and Designing of Energy Materials from First Principles Simulations: Optoelectronic and Hybrid-Photovoltaic Devices*
Dec 2012	Colloquium of CIC Biomagune, San Sebastián, Spain *What Theoretical Modeling Provides for Our Understanding and Predicting Properties of Nanostructures and BioMolecules*
Feb 2013	CECAM Workshop on Calculation of Optical Properties of Nanostructures from First Principles, Lausanne, Switzerland *Fundamentals of TDDFT for Nonlinear Phenomena of Light-Matter Interactions: Application to Hybrid Organic Photovoltaics and Charge Transfer Processes*
Mar 2013	International Winterschool on Electronic Properties of Novel Materials (IWEPNM2013), Kirchberg/Tirol, Austria *New Nanoscale Hybrid Structures Made of C and BN from First Principles: Optoelectronic Devices*

Mar 2013	Colloquium, San Sebastián, Spain *Supercomputación Científica en Euskadi*
Apr 2013	TD-DFT (Time-Dependent Density Functional Theory) Conference, Université de Nantes, Nantes, France *How Non Linear and Charge Transfer Processes Are Captured in Time-Dependent Density Functional Theory*
Apr 2013	Workshop on Dynamics of Matter: Advances in Theory, Center for Free-Electron Laser Science, Hamburg, Germany *Non Equilibrium Dynamical Processes in Finite and Extended Systems: A TDDFT and Many-Body Perspective*
May 2013	Electronic Structure Calculations with the GPAW Code: Users and Developers Meeting, Technical University of Denmark, Lyngby, Denmark *Time-Dependent Density Functional Theory for Non-Linear Phenomena in Solids and Nanostructures: Fundamentals and Applications*
Jul 2013	Workshop on Learning from the Past, Looking to the Future, Berlin, Germany *A TDDFT Perspective on Nonlinear Electronic Processes: Optics, Photoemission and Resonant Tunneling*
Jul 2013	Colloquium, Department of Chemistry (AG Joachim Sauer), Humboldt-Universität zu Berlin, Berlin, Germany *Static and Time-Dependent Density-Functional Schemes for Bond-Breaking and Bond Formation, Correlation Effects Including Mott Insulators*
Sep 2013	Hetero NanoCarb, Castelldefels, Barcelona, Spain *From Pure to Doped Carbon-Based Nano-Materials: Recent Advances and Applications*
Oct 2013	MPQ Colloquium, Munich, Germany *Non Equilibrium Dynamical Processes in Finite and Extended Systems from a Time-Dependent Density Functional (TDDFT) Perspective*

Matthias Scheffler

Sep 2011	CECAM Workshop on Perspectives and Challenges of Many-Particle Methods, Bremen, Germany *The Base of Multi-Scale Modeling: Assessing the Accuracy and Reliability of Electronic-Structure Theory*
Jan 2012	Symposium on Surface and Nano Science, Shizukuishi, Japan *Role of van der Waals Interactions in Adsorption and at Inorganic/Organic Interfaces*
Jan 2012	Materials Research Outreach Symposium, University of California, Santa Barbara, CA, USA *Role of van der Waals Interactions in Physics, Chemistry, and Biology*
Feb 2012	Workshop on New Theory and Methods in Quantum Research, University of British Columbia, Vancouver, Canada *Accuracy of Ab Initio Computations in Materials Science*

Feb 2012	APS March Meeting, Boston, MA, USA *Reliability and Robustness of Density-Functional Theory and GW Calculations*
Feb 2012	Colloquium on Physical Chemistry, Ludwig-Maximilians-Universität München, Munich, Germany *Role of van der Waals Interactions in Physics, Chemistry, and Biology*
Mar 2012	25th Symposium on Surface Science, St.Christoph/Arlberg, Austria *Role of van der Waals Interactions in Adsorption of Organic Molecules at Inorganic Surfaces*
Mar 2012	Plenary Talk at the DPG-Frühjahrstagung, Berlin, Germany *Role of van der Waals Interactions in Physics, Chemistry, and Biology*
May 2012	Workshop on Recent Advances in Materials Physics Symposium, North Carolina State University, Raleigh, NC, USA *Role of van der Waals Interactions in Physics, Chemistry, and Biology*
Jun 2012	Physikalisches Kolloquium, University of Duisburg-Essen, Duisburg, Germany *Role of van der Waals Interactions in Physics, Chemistry, and Biology*
Jun 2012	Plenary Talk at the Congress on Electronic Structure: Principles and Applications, ESPA 2012, Barcelona, Spain *The Random-Phase Approximation for Electron Correlation and Beyond*
Jul 2012	Keynote/Opening Talk at the Foundations of Molecular Modeling and Simulations Conference (FOMMS 2012), Mount Hood, OR, USA *Role of van der Waals Interactions in Physics, Chemistry, and Biology*
Jul 2012	Physics and GDCh Colloquium, Universität Bremen, Bremen, Germany *Role of van der Waals Interactions in Physics, Chemistry, and Biology*
Sep 2012	Workshop of the Partnership of International Research and Education, Fudan University, Shanghai, China *Role of van der Waals Interactions in Physics, Chemistry, and Biology*
Sep 2012	CECAM Workshop on Validation and Verification in Electronic-Structure Calculations: State of the Art and Perspectives, Lausanne, Switzerland *Towards the Validation Level of Electronic Structure and Total Energy Calculations of Materials*
Sep 2012	Symposium on The Future of Catalysis, Stanford University, Stanford, CA, USA *Towards the Validation Level of Electronic Structure and Total Energy Calculations of Materials*
Oct 2012	Colloquium at Oak Ridge National Laboratory, Oak Ridge, TN, USA *Role of van der Waals Interactions in Physics, Chemistry, and Biology*
Dec 2012	Plenary Talk at the Materials Gerome Initiative Workshop of the National Science Foundation, Arlington, VA, USA *Setting the Stage for a Successful MGI*
Jan 2013	Symposium on Surface on Nano Science 2013, Zao, Japan *The Materials Genome Initiative - The Basic-Science Component*

Mar 2013	3S'13 - 26th Symposium on Surface Science, Åre, Sweden *Density-Functional Theory with Many-Body, Screened van der Waals Interactions*
Apr 2013	Symposium on Theory and Simulation in Energy and Fuel Production and Utilization at the 245th ACS National Meeting and Exposition, New Orleans, LA, USA *Adsorption of Organic Molecules at Metal and Metal Oxide Surface*
Apr 2013	Symposium on Theory and Simulation in Energy and Fuel Production and Utilization at the 245th ACS National Meeting and Exposition, New Orleans, LA, USA *Understanding the Concentration of Surface Vacancies at Metal Oxides: The Example of MgO (100)*
Jun 2013	International Symposium on Recent Electronic-Structure Theories and Related Experiments, Stuttgart, Germany *Intrinsic Defects and Adsorption at Oxide Surfaces: Theoretical Challenges, Concepts, and Insights*
Jul 2013	CECAM Workshop on Density Functional Theory: Learning from the Past, Looking to the Future, Berlin, Germany *Thermodynamics and Statistical Mechanics from First Principles for Surfaces and Interfaces: Theoretical Challenges, Concepts, and Insights*
Jul 2013	Joint ICTP-NSFC School and Advanced Workshop on Modern Electronic Structure Computations, Shanghai, China *Thermodynamics and Statistical Mechanics from First Principles for Surfaces and Interfaces: Theoretical Challenges, Concepts, and Insights*
Jul 2013	Hayashi Conference: Next decades of Surface Science, Hayama, Japan *Intrinsic Defects and Adsorption at Oxide Surfaces: Theoretical Challenges, Concepts, and Insights*
Jul 2013	International Summer School on Basic Concepts and First-Principles Computations for Surface Science: Applications in Chemical Energy Conversion and Storage, Norderney, Germany *Electronic Structure Theory*
Jul 2013	International Summer School on Basic Concepts and First-Principles Computations for Surface Science: Applications in Chemical Energy Conversion and Storage, Norderney, Germany *Space Charge Transfer at Metal Oxide Surfaces*
Aug 2013	Lecture, Hands-on Workshop DFT and Beyond: Computational Materials Science for Real Materials, Trieste, Italy *Electronic Structure Overview*
Aug 2013	DOE Symposium on New Theoretical Concepts and Directions in Catalysis, University of California, Santa Barbara, CA, USA *Intrinsic Defects and Adsorption at Oxide Surfaces: Theoretical Challenges, Concepts, and Insights*

Sep 2013	Keynote Lecture, CPMD-Meeting 2013: Matter, Life, Light from Ab Initio Molecular Dynamics Simulations, Leipzig, Germany *Thermodynamics and Statistical Mechanics from First Principles for Surfaces and Interfaces: Theoretical Challenges, Concepts, and Insights*
Oct 2013	IPAM program on Materials for a Sustainable Energy Future, Los Angeles, CA, USA *Theoretical Concepts and Directions in Heterogeneous Catalysis: What Comes Next?*

Alexandre Tkatchenko

Nov 2011	Seminar, University of Vienna, Vienna, Austria *Van der Waals Interactions in Biology, Chemistry, and Physics*
Dec 2011	Seminar, University of South Florida, Tampa, FL, USA *Van der Waals Interactions in Biology, Chemistry, and Physics*
Dec 2011	Quitel Conference, Cancun, Mexico *Many-Body van der Waals Interactions in Biology, Chemistry, and Physics*
Dec 2011	SFB HIOS Kickoff Meeting, Berlin, Germany *Accurate Description of van der Waals Interactions for Inorganic/Organic Interfaces*
Feb 2012	Seminar, Pohang University of Science and Technology, Pohang, Korea *Van der Waals Interactions in Biology, Chemistry, and Physics*
Mar 2012	Third International Supercomputing Conference, Guanajuato, Mexico *Atomistic Modeling from First-Principles: Pushing the Limits of Accuracy and Speed*
Jun 2012	Seminar, Forschungszentrum Jülich, Jülich, Germany *Van der Waals Interactions in Biology, Chemistry, and Physics*
Jun 2012	Seminar, University of Pittsburgh, Pittsburgh, PA, USA *Van der Waals Interactions in Biology, Chemistry, and Physics*
Jun 2012	Telluride Meeting on Many-Body Interactions from Quantum Mechanics to Classical Potentials, Telluride, CO, USA *Accurate and Efficient First-Principles Calculation of van der Waals Energy*
Aug 2012	Seminar, Hanyang University, Seoul, Korea *Van der Waals Interactions in Biology, Chemistry, and Physics*
Aug 2012	Workshop on Density Functional Theory and Beyond with Numeric Atom-Centered Orbitals, Berlin, Germany *Van der Waals Formalisms for Molecules and Condensed Matter*
Sep 2012	European Conference on Condensed Matter (CMD-24), Edinburgh, UK *Van der Waals Interactions in Molecules, Solids, and Interfaces*
Oct 2012	Seminar, École Polytechnique Fédérale de Lausanne, Lausanne, Switzerland *Van der Waals Interactions in Biology, Chemistry, and Physics*

Oct 2012	IPAM Workshop on Material Defects, University of California, Los Angeles, Los Angeles, CA, USA *Van der Waals Interactions in Molecules and Condensed Matter, Including Systems with Defects*
Dec 2012	1st IPAM Reunion on Navigating Chemical Compound Space, Lake Arrowhead, CA, USA *Collective Many-Body van der Waals Interactions in Molecular Systems*
Jan 2013	Seminar, MPI für Eisenforschung GmbH, Düsseldorf, Germany *Van der Waals Interactions in Molecules, Solids, and Interfaces*
Mar 2013	DPG Frühjahrstagung, CPP Section, Ratisbon, Germany *Collective van der Waals Interactions in Molecules and Solids*
Mar 2013	APS March Meeting, DCOMP Section, Baltimore, MD, USA *Improving Density Functionals with Quantum Harmonic Oscillators*
Apr 2013	245th ACS National Meeting and Exposition, PHYS section, New Orleans, LA, USA *Collective van der Waals Interactions in Molecular Systems*
Apr 2013	Seminar, Chemistry, University of Nottingham, Nottingham, UK *Many-Body van der Waals Interactions with Applications in Biology, Chemistry, and Physics*
Apr 2013	Cold Atoms Seminar, School of Physics and Astronomy, University of Nottingham, Nottingham, UK *Van der Waals Interactions in Molecules, Solids, and Interfaces*
May 2013	Seminar, Thomas Young Center, London, UK *Get Real: Van der Waals Interactions in Complex Materials*
May 2013	Seminar, Queen Mary University of London, London, UK *Masterclass on van der Waals Interactions*
May 2013	Seminar, Trinity College, Dublin, Ireland *Van der Waals Interactions in Molecules, Solids, and Interfaces*
Jun 2013	Seminar, University of Potsdam, Potsdam, Germany *Van der Waals Interactions in Molecules, Solids, and Interfaces*
Jul 2013	Conference on Chemical Bonding, Kauai, HI, USA *(Un)Chemical Bonding: Surprises in Non-Covalent Interactions in Molecular Systems*
Jul 2013	International Summer School on Basic Concepts and First-Principles Computations for Surface Science: Applications in Chemical Energy Conversion and Storage, Norderney, Germany *Bonding at Surfaces and Interfaces*
Aug 2013	Hands-on Workshop DFT and Beyond: Computational Materials Science for Real Materials, Trieste, Italy *Practical Approaches to van der Waals Interactions*

Aug 2013	Meeting on Transatlantic Frontiers of Chemistry, Kloster Seeon, Germany *Collective van der Waals Interactions in Molecular Systems*
Aug 2013	VIII Congress of the International Society of Theoretical Chemical Physics, Budapest, Hungary *(Un)Chemical Bonding: Surprises in Non-Covalent Interactions in Molecular Systems*
Sep 2013	246th ACS National Meeting and Exposition, Indianapolis, IN, USA *Reliable Modeling of Bonding for Complex Interfaces with Applications in Catalysis, Molecular Electronics, and Photovoltaics*
Sep 2013	Seminar, Chemistry Department, Princeton University, Princeton, NJ, USA *Collective van der Waals Interactions in Molecules and Solids*
Sep 2013	Joint Seminar of ITAMP and Chemical Biology, Harvard University, Cambridge, MA, USA *Collective van der Waals Interactions in Molecules and Solids*
Oct 2013	CECAM Workshop on Structure-Property Relationships of Molecular Precursors to Organic Electronics, Lausanne, Switzerland *Collective Phenomena in Organic Materials Described with Coupled Quantum Harmonic Oscillators*

Yong Xu

Sep 2012	Chinese Physics Meeting, Guangzhou, China *Quantum Thermal Transport in Carbon Nanostructures*
Sep 2012	Chinese Physics Meeting, Guangzhou, China *Work Function Tuning at Hybrid Inorganic/Organic Interfaces*

Notes