



FRITZ-HABER-INSTITUT  
MAX-PLANCK-GESELLSCHAFT

# Poster Abstracts and Bibliographic Information

2017–2019

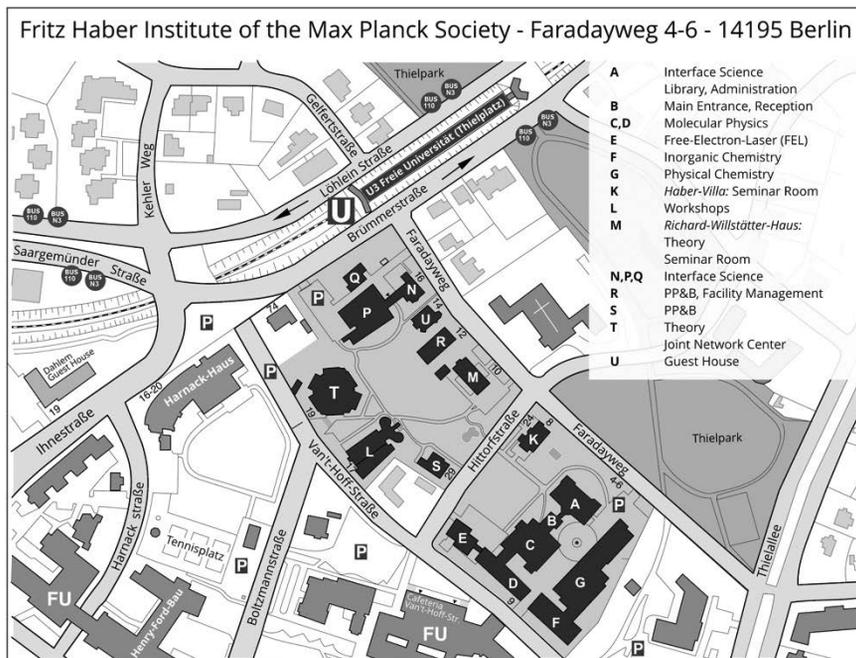


**Poster Abstracts and  
Bibliographic Information**  
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Fritz-Haber-Institut der Max-Planck-Gesellschaft

Berlin, September 2019  
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## Map



The department poster sessions will take place on Tuesday (Nov. 26, 2019) between 2:30 and 5:30 pm in the following buildings:

Department	Building
Inorganic Chemistry	F
Chemical Physics	P
Interface Science	P
Molecular Physics	A
Physical Chemistry	G
Theory	T

## Table of Contents

Map of the FHI	2
Table of Contents	3
<b>Abstracts</b>	
Department of Inorganic Chemistry	4
Department of Interface Science	30
Department of Molecular Physics	51
Department of Physical Chemistry	72
Theory Department	96
Department of Chemical Physics	112
<b>Scientific Output</b>	
Department of Inorganic Chemistry	
Publications	120
Invited Talks	136
Department of Interface Science	
Publications	144
Invited Talks	148
Department of Molecular Physics	
Publications	152
Invited Talks	160
Department of Physical Chemistry	
Publications	166
Invited Talks	175
Theory Department	
Publications	188
Invited Talks	196
Department of Chemical Physics	
Publications	209
Invited Talks	214
FHI-FEL Publications	219
PhD projects of the IMPRS	222

# Department of Inorganic Chemistry

## Poster List

- AC 1.0 Catalysis on Oxides**
- AC 1.1 Perovskites in Oxidation Reactions**  
Giulia Bellini, Gregor Koch, Michael Hävecker, Frank Girgdsies, Thomas Lunkenbein, Walid Hetaba, Pierre Kube, Frank Rosowski, Annette Trunschke, and Robert Schlögl
- AC 1.2 Supported Liquid Phase Catalysts for Oxidative Dehydrogenation of Propane**  
Ezgi Erdem, Pierre Kube, Andrey Tarasov, Frank Rosowski, Annette Trunschke, and Robert Schlögl
- AC 1.3 Activation of Molecular Oxygen on Metal Oxides**  
Yuanqing Wang, Gregory Huff, Anna Wernbacher, Frank Rosowski, Annette Trunschke, and Robert Schlögl
- AC 1.4 Clean Data in Oxidation Catalysis**  
Peter Kraus, Pierre Kube, Spencer Carey, Frank Girgdsies, Sabine Wrabetz, Andrey Tarasov, Olaf Timpe, Detre Teschner, Michael Hävecker, Liudmyla Masliuk, Michael Geske, Raoul Naumann d'Alnoncourt, Frank Rosowski, Luca M. Ghiringhelli, Matthias Scheffler, Annette Trunschke, and Robert Schlögl
- AC 1.5 Activation of Oxygen - Key to the Oxidative Coupling of Methane (OCM) Reaction**  
Lukas Thum, Maik Rudolph, Reinhard Schomäcker, Wiebke Riedel, Thomas Risse, Yuanqing Wang, Andrey Tarasov, Annette Trunschke, and Robert Schlögl
- AC 2.0 Metals**
- AC 2.1 X-ray spectroscopy investigation of the cathodic electroreduction of CO<sub>2</sub> into valuable chemical feedstocks onto Copper based catalysts**  
Juan J. Velasco-Vélez, Qingjun Zhu, Cheng-Hao Chuang, Travis Jones, Dunfeng Gao, Emilia Carbonio, Beatriz Roldan Cuenya, Robert Schlögl, and Axel Knop-Gericke
- AC 2.2 In operando XPS and XAS on oxygen evolution reaction catalysts**  
Lorenz Falling, Rik V. Mom, Travis E. Jones, Juan J. Velasco-Vélez, Detre Teschner, Axel Knop-Gericke, and Robert Schlögl
- AC 2.3 Synthetic Manganese Oxides at Water-Oxidation Catalysis Conditions: Phase Changes and Surface vs. Bulk Properties**  
Katarzyna Skorupska, Travis Jones, Frederic Sulzmann, Jens Melder, Frank Girgdsies, Christian Rohner, Philipp Kurz, Axel Knop-Gericke, and Robert Schlögl
- AC 2.4 In-situ Synchrotron Radiation based Studies on MnOx Water-Oxidation Catalysts: Electronic Structure and Reaction Mechanism**  
Katarzyna Skorupska, Travis Jones, Rik Mom, Detre Teschner, Lifei Xi, Jens Melder, Kathrin Aziz-Lange, Philipp Kurz, Cheng-Hao Chuang, Axel Knop-Gericke, and Robert Schlögl

- AC 2.5 **SO<sub>x</sub> species on silver in alkene epoxidation**  
Emilia A. Carbonio, Alexander Yu. Klyushin, Frederic Sulzmann, Elias Frei,  
Michael Hävecker, Simone Piccinin, Axel Knop-Gericke,  
Travis E. Jones, and Robert Schlögl
- AC 3.0 **Untangling Structural Complexity by Chemical Electron Microscopy**  
AC 3.1 **Implementation and Development of Methodological Strategies for TEM  
Characterisation of Catalysts**  
Gerardo Algara-Siller, Christoph Pratsch, Christian Rohner, Walid Hetaba,  
Thomas Lunkenbein, and Robert Schlögl
- AC 3.2 **Exploring Structural Complexity of Heterogeneous Catalysts by Quasi in  
situ (S)TEM**  
Liudmyla Masliuk, Kassioqe Dembele, Jan Folke, Holger Ruland,  
Annette Trunschke, Thomas Lunkenbein, and Robert Schlögl
- AC 3.3 **ESEM -A Versatile Tool for the Study of Heterogeneous Catalysts**  
Luis Sandoval-Diaz, Stéphane Poitel, Milivoj Plodinec, Ezgi Erdem,  
Juan J. Velasco-Vélez, Rick Mom, Lorenz Falling, Eugen Stotz, Annette Trunschke,  
Axel Knop-Gericke, Thomas Lunkenbein, and Robert Schlögl
- AC 3.4 **Operando TEM for the Study of Heterogeneous Catalysts under Relevant  
Reaction Conditions**  
Milivoj Plodinec, Hannah C. Nerl, Luis Sandoval-Diaz, Liudmyla Masliuk,  
Thomas Lunkenbein, and Robert Schlögl
- AC 4.1 **Reaction Calorimetry: Exploring a Solid Catalyst In-Situ**  
Andrey Tarasov, Elias Frei, Sabine Wrabetz, Travis Jones, Ezgi Erdem,  
Annette Trunschke, and Robert Schlögl
- AC 4.2 **Mechanisms of Deactivation of Supported Pt Catalysts and their  
Regeneration in Propane Dehydrogenation**  
Siamak Nakhaie, Hannah C. Nerl, Andrey Tarasov, Jutta Kröhnert, Frank Girgsdies,  
Annette Trunschke, Katarzyna Skorupska, and Robert Schlögl
- AC 5.1 **Development of a Multimodal Experiment for the Investigation of the  
Heterogeneous Chemistry of Liquid/Vapor Interfaces**  
Holger Oertel, Robert Schlögl, and Hendrik Bluhm
- AC 5.2 **Compression of a Stearic Acid Surfactant Layer on Water Investigated by  
Ambient Pressure XPS**  
Harmen Hoek, Timm Gerber, Clemens Richter, Kevin R. Wilson, Robert Schlögl,  
and Hendrik Bluhm
- AC 6.0 **Department of Heterogeneous Reactions**  
Mark Greiner, Saskia Heumann, Anna K. Mechler,  
Holger Ruland, and Robert Schlögl
- AC 7.1 **EMIL: Bridging the “Pressure Gap” in Spectromicroscopy**  
Juan J. Velasco-Vélez, Christoph Pratsch, Michael Hävecker, Eugen Stotz,  
Mihaela Gorgoi, Stefan Hendel, Franz Schäfers, Andreas Gaupp,  
Thomas Lunkenbein, Axel Knop-Gericke, and Robert Schlögl
- AC 7.2 **Research in Collaboration with BasCat**  
Annette Trunschke, Frank Rosowski, and Robert Schlögl

## Catalysis on Oxides

In heterogeneous catalysis over metal oxides the delicate interplay between electronic bulk structure and surface termination is used to control the activation of molecules at the catalyst surface and to govern selectivity in complex reaction networks.

Our current research is focused on the investigation of metal oxides as catalysts in selective oxidation of short-chain alkanes. The work is performed in collaboration with the BasCat - UniCat BASF JointLab at the Technical University Berlin.

High selectivity to value-added products is achieved over the semiconductors vanadyl pyrophosphate  $(VO)_2P_2O_7$  or  $MoVTeNbO_x$  of M1 structure in *n*-butane or propane oxidation, respectively, even at high alkane conversion (see also Poster AC 3.2). Here, the abundance of highly reactive electrophilic surface oxygen species is controlled by a self-limiting charge carrier transport situation that is created by surface modification under the influence of the feed and the corresponding band bending.

Due to their structural and chemical versatility, pure or partially substituted  $AA'A''BB'B''(O,X)_3$  perovskites have been selected as model system to investigate the universality of such a concept in oxidation catalysis. The functions of various oxygen species present on the surface under operation conditions were analyzed by NAP-XPS and NEXAFS (Poster AC 1.1).

In addition, we apply the concept of supported liquid-phase (SLP) catalysts to investigate the impact of phase transitions in thin vanadium oxide surface layers on catalyst performance in propane oxidation (see also Poster AC 3.3). To achieve melting, the addition of fluxing agents is necessary. In the present work, the influence of the fluxing agent on the catalytic properties was investigated with the aim to clarify whether the additive or the phase transition is primarily responsible for changes in catalyst performance (Poster AC 1.2).

Electronic properties of polycrystalline model catalysts were investigated by conductivity measurements and photoelectron spectroscopy. Complementary, Raman spectroscopy in combination with oxygen exchange studies was applied to analyze the interaction of  $O_2$  with metal oxides (Poster AC 1.3).

The dynamic and manifold response of catalysts to process conditions complicates the identification of descriptors in oxidation catalysis. In this respect machine learning may contribute to disclose hidden relations. This requires the definition of standards and clear procedures, which will enable quality control and the collection of reliable and balanced experimental datasets. For this purpose, a handbook of selective oxidation was developed (Poster AC 1.4).

The importance of oxygen activation in directing the selectivity towards the desired products was also shown in the oxidative coupling of methane over binary alkaline earth oxides. Moreover, the impact of additives on the electronic and catalytic properties was investigated (Poster AC 1.5).

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## Perovskites in Oxidation Reactions

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The multifaceted compositional and structural chemistry of perovskites turns this mineral class into an attractive experimental ground to explore links between solid-state chemistry and catalysis. The concept has been pursued for long time<sup>1</sup>. Today, advanced experimental and computational methods are available that facilitate descriptor-based identification of potential catalysts based on a broad variety of materials also for more complex reactions and challenging operating conditions<sup>2</sup>.

It has been shown in previous studies that the total combustion catalyst manganese oxide can be converted into a more selective oxidation catalyst by embedding MnO<sub>2</sub> species into a tungsten oxide matrix<sup>3</sup>. The concept was developed further in the present work by investigating perovskites that contain various redox active elements in oxidation reactions.

The Pechini route was applied to synthesize single B-site perovskites ABO<sub>3</sub> (A = La, Pr, Nd, Sm; B = V, Cr, Mn, Fe, Co, Ni) and double B-site perovskites AB<sub>(1-x)</sub>B'<sub>x</sub>O<sub>3</sub> (A = La, Pr; (B, B') = (Mn, Cu), x=0.1-0.5). All materials were phase-pure oxides presenting a porous structure defined by a coral-like morphology and specific surface areas that vary between 1.8 and 29 m<sup>2</sup>·g<sup>-1</sup>. The catalysts were tested in the oxidation of CO and propane. Substitution of La by Pr, Nd, and Sm in ABO<sub>3</sub> led to an increased distortion of the structure, reflected in an enhanced catalytic performance if Fe occupies the B-site. The Co-based samples presented the highest catalytic activity. Due to increased selectivity, AMnO<sub>3</sub> (A = La, Sm) catalysts were analyzed in-depth. Abundance of Mn on the surface and the presence of steam in the feed determine the selectivity to propene. The function of various oxygen species present on the surface under operation conditions was analyzed by operando XPS and NEXAFS<sup>4</sup>. Further increase in propene selectivity was observed when Mn was partially substituted by Cu in the double B site-based catalysts, underlying how the synergistic effect of two elements in the B-site exerts a beneficial influence under oxidative catalytic conditions.

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## Supported Liquid Phase Catalysts for Oxidative Dehydrogenation of Propane

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Annette Trunschke, and Robert Schlögl

Recently, it has been shown that melting phenomena influence the catalytic performance in oxidative dehydrogenation of propane (ODP) significantly when silica-supported vanadium oxide is modified by potassium<sup>1</sup>. Activity drops and selectivity increases markedly when the supported V-K oxide layer and/or segregated crystalline potassium vanadate phases are melting. The present study deals with the question whether liquefaction has a similar effect when other alkali oxides A<sub>2</sub>O (A = Cs, Rb) are used as fluxing agents, *i.e.*, whether the phenomena described above arise from an impact of potassium<sup>2,3</sup>, or phase transition. For comparison, SiO<sub>2</sub>-supported vanadium oxide catalysts that contain K, Li, or Na oxide, respectively, were also prepared.

Aqueous suspensions of alkali carbonates and V<sub>2</sub>O<sub>5</sub> with SiO<sub>2</sub> (Aerosil 300) were spray-dried and the resulting powders were treated in synthetic air at 620°C for 16 hours. The catalyst that contains 37 mol% K exhibited the formation of only one crystalline phase (K<sub>3</sub>V<sub>5</sub>O<sub>14</sub>) in the solid state at room temperature. A sharp melting signal was observed by differential scanning calorimetry (DSC) at 410°C, which was attributed to melting of K<sub>3</sub>V<sub>5</sub>O<sub>14</sub><sup>3</sup>. CsV<sub>2</sub>O<sub>5</sub> was the only detectable crystalline phase in all Cs-containing catalysts at low temperature. The phase transformation occurred in a broad temperature range between 430°C and 446°C. Rb addition led to formation of the mixtures of Rb<sub>2</sub>V<sub>4</sub>O<sub>11</sub> and Rb<sub>3</sub>V<sub>5</sub>O<sub>14</sub> phases with melting points below 400°C.

The activity in the ODP reaction dropped when the melting point of the alkali vanadate phases was reached. Operando DSC revealed a clear correlation between melting and decrease in activity. In case of the K- and Rb-containing catalysts, selectivity rapidly increased after melting. The effects were less pronounced for the Cs-containing catalysts, perhaps due to the broad temperature range of melting. At higher temperatures, the selectivity of the all catalysts approached similar values.

Decrease in specific surface area or loss of oxygen can be excluded as reasons for the drop in activity. Structural reasons for the performance changes will be discussed based on spectroscopic investigations of the materials under operation and comparison with Li- and Na- catalysts, which shows no melting during catalysis.

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## Activation of Molecular Oxygen on Metal Oxides

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Control over occurrence and distribution of various activated oxygen species on the surface of oxide catalysts is the key issue in terms of achieving high selectivity to specific selective oxidation products and the abatement of total combustion to undesired CO<sub>2</sub> in alkane oxidation.

Relations between the electronic properties of V-based (mixed) metal oxides and their catalytic performance in the oxidation of ethane, propane, and *n*-butane were studied by operando conductivity measurements and near-ambient-pressure X-ray photoelectron spectroscopy (NAP-XPS). V<sub>2</sub>O<sub>5</sub>, vanadyl pyrophosphate (VO)<sub>2</sub>P<sub>2</sub>O<sub>7</sub> (VPP), and MoV(TeNb) oxides with orthorhombic M1 structure were included in the study.

Experimentally observed correlations between performance and charge carrier dynamics give hints to universal design concepts of high performance catalysts for selective oxidation. Catalysts with high selectivity at almost complete alkane conversion (VPP and MoVTeNb M1 oxide) reveal the property of a "buffer" for charge carriers in its bulk that can be transported to the active sites when the delivery of redox equivalents between adsorbed species is too slow. Likewise, the surface layer containing the active sites is robust against chemical changes induced by the lack of redox equivalents leading to chemical oxidation or reduction of the sites<sup>1,2</sup>.

Surface molecular aspects of oxygen activation have been studied at different temperatures and gas-phase compositions by Raman spectroscopy combined with DFT calculations. Phonon calculations were performed using Quantum Espresso 5.4.0 to refine the band assignment of the V<sub>2</sub>O<sub>5</sub> Raman spectrum with the aim to allow a clear identification of the type of oxygen that undergoes an exchange reaction. Labelling experiments with <sup>18</sup>O<sub>2</sub> show that the type of oxygen in V<sub>2</sub>O<sub>5</sub>, which is exchanged, strongly depends on gas phase composition and temperature. High temperatures are required under oxidizing conditions and vanadyl oxygen is the only species that is exchanged at 580°C, whereas in presence of propane and oxygen all types of oxygen (vanadyl and bridging) are exchanged. Oxygen vacancies generated by propane in the surface region facilitate the incorporation of gaseous oxygen into the lattice of V<sub>2</sub>O<sub>5</sub>. Under normal reaction conditions of alkane oxidation (400 – 420 °C), mainly vanadyl oxygen O1 is involved in oxygen exchange. The experiments demonstrate that spectroscopic analysis of the working catalyst is needed for generating insight that is causally related to the catalytic phenomena.

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## Clean Data in Oxidation Catalysis

Peter Kraus, Pierre Kube, Spencer Carey, Frank Girgsdies, Sabine Wrabetz, Andrey Tarasov, Olaf Timpe, Detre Teschner<sup>a</sup>, Michael Hävecker<sup>a</sup>, Liudmyla Masliuk, Michael Geske<sup>b</sup>, Raoul Naumann d'Alnoncourt<sup>b</sup>, Frank Rosowski<sup>c</sup>, Luca M. Ghiringhelli<sup>d</sup>, Matthias Scheffler<sup>d</sup>, Annette Trunschke, and Robert Schlögl

Heterogeneous catalysis in general, and oxidation catalysis in particular, is a complex and highly dimensional research field. While a large amount of data has been produced over the last 40 years<sup>1</sup>, the resulting datasets can usually not be compared directly, as the applied conditions are incomparable and frequently incomplete. Transparent and open handling of data is becoming a topic of crucial importance across scientific disciplines. Here, we propose a “Clean Data Handbook”, which aims to define a minimum standard for catalytic testing and characterization of catalysts in selective oxidation, enabling comparison of materials under reference conditions.

Catalytic testing and characterizations are performed on an initial training set of 10 materials, which are synthesized in a large batch size of 20 g each. The analyzed reactions include CO, ethane, propane and *n*-butane oxidation, using a prescribed schedule of steps, including defined pre-treatment conditions, temperature and contact time variation, feed composition as well as co-feed of intermediates and steam. The catalysts are characterized by standard techniques including XRD, surface area determination, a temperature programmed oxidation and reduction study, elemental analysis, and a near surface analysis by XPS. Selected characterization techniques are applied to the „spent“ materials following catalytic testing.

Further methods can be included as part of the clean dataset, but do not form the required minimum standard. In the current iteration, such methods are: oxygen and alkane adsorption calorimetry, contactless operando conductivity measurements, and near-ambient pressure operando XPS.

The work is performed in collaboration with the Theory Department with the aim to use the toolbox of artificial intelligence for establishing a bridge between theory and experiment by searching for hidden relations and causally determined connections in experimental data with the help of machine learning and data science. Theory could then predict the combination of optimal conditions and materials for a given process.

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## Activation of Oxygen - Key to the Oxidative Coupling of Methane (OCM) Reaction

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Yuanqing Wang<sup>c</sup>, Andrey Tarasov, Annette Trunschke, and Robert Schlögl

The major challenge in OCM is the direction of the reaction towards the desired higher hydrocarbons ethane and ethane ( $C_{2+}$ ) while suppressing the thermo-dynamically favored deep oxidation towards carbon monoxide and carbon dioxide. The yield of 30%  $C_{2+}$  apparently represents an insurmountable barrier that has yet to be surpassed<sup>1</sup>. Non-reducible basic oxides such as alkaline earth oxides as well as rare earth oxides have been found as promising catalyst materials. Even though the bond dissociation energy of methane is large with 423 kJ/mol, controlling the activation of oxygen is much more important to direct the selectivity towards the desired products<sup>2</sup>. In case of basic non-reducible oxides, e.g., CaO, the capability of the material to activate oxygen appears to be directly linked to the OCM reactivity, and it appears that the dissociation of  $O_2$  is most likely the bottleneck of the reaction<sup>2</sup>. The strong basicity, which is needed for the reaction, is also a major weakness of those materials, forming stable hydroxides and carbonates. The latter are incapable of activating oxygen even at high temperatures<sup>2</sup>. To improve the  $O_2$  activation, electron-rich transition metal centers can be inserted into the alkaline earth host lattice, which provide electrons via tunneling to adsorbed oxygen lowering its dissociation barrier<sup>3</sup>. First-row transition metal-doped calcium oxides were synthesized, characterized and tested in the OCM reaction. In a screening Zn, Ni and Mn were found to have a beneficial effect for the selectivity and activity in the OCM reaction, whereas Co and Cr dopants were found to be disadvantageous. The impact of the dopant is most effective in low doping concentrations (< 0.1 at%), showing a minimum in the apparent activation energy. At higher concentrations, clustering appears to reduce the efficiency. An increased line broadening in EPR signals due to dipole-dipole interactions is observed for Mn-doped CaO as well as a diminishment of the surface derived photoluminescence for the higher doped samples, indicating the generation of sites for non-radiative relaxation and, therefore, a significantly changed electronic surface structure.

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## Metals

The electrocatalytic route of energy conversion is becoming an important technology to convert CO<sub>2</sub> into valuable chemical feedstocks using renewable sources of energy. However, finding an efficient and stable electrocatalyst for the reduction of CO<sub>2</sub> in aqueous solution is challenging. The electrochemically prepared copper-redox electro-cathodes show higher activity for the production of hydrocarbons at lower oxidation state. In-situ experiments yielded unambiguous information of the catalyst redox processes governing the CO<sub>2</sub> reduction reaction (CO<sub>2</sub>RR), as well as the nature of the active sites. In addition, the active/inactive and stable/unstable oxidation states depending on the applied potential and electrolyte were revealed. (AC 2.1)

The insufficient green production of hydrogen is a bottleneck in the development of a sustainable hydrogen network. Electrolytic water splitting in acidic electrolytes addresses this problem by making use of the intermittent nature of renewable energies to produce hydrogen at high pressures. The limiting process is the anodic oxygen evolution reaction (OER), which is plagued by high overpotentials and electrode degradation. We have studied the potential-driven restructuring of Ru, Ir, Pt and Au oxides in 0.1 M H<sub>2</sub>SO<sub>4</sub> during the oxygen evolution reaction using NAP-XPS, XAS and DFT. Our results show the distribution of cation oxidation state at the surface of the catalyst and the oxidation and deprotonation kinetics with relevant time resolution. (AC 2.2)

The CaMn<sub>4</sub>O<sub>x</sub> cluster of the enzyme Photosystem II, has spurred great interest in MnO<sub>x</sub>-catalyzed electrochemical oxidation of H<sub>2</sub>O for renewable fuel synthesis. Different manganese oxides were synthesized and tested in the OER. It was found that the contribution of surface versus bulk plays a crucial role in their performance, therefore different techniques, such as XPS, NEXAFS, XRD, IR, SEM and TEM, were applied to characterized samples. Due to the unstable character of Mn<sup>2+</sup>, higher oxidation states of manganese were already observed at the surface of all MnO<sub>x</sub> powders. There is no evidence supporting involvement of manganese species with oxidation states higher than IV in the catalytic mechanism. (AC 2.3/4)

The selective oxidation reaction of ethylene over Ag is very attractive due to the economic importance of ethylene oxide (EO) as a versatile chemical intermediate. The nature of the oxygen species that can produce EO on Ag catalysts have been recently identified as originating from adsorbed SO<sub>4</sub>. NAP-XPS experiments demonstrated that the EO selectivity tracks the coverage of SO<sub>4</sub>. In-situ Raman characterization of Ag shows the relevance of these findings for the industrial process. In contrast to ethylene epoxidation, SO<sub>4</sub> is titrated under propylene oxidation conditions and SO<sub>3</sub> is seen as a titration product. The co-existence of SO<sub>4</sub> and atomic O on the Ag surface appears to dictate the (high) EO and (low) PO selectivity. (AC 2.5)

## X-ray spectroscopy investigation of the cathodic electroreduction of CO<sub>2</sub> into valuable chemical feedstocks onto Copper based catalysts

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Among technologies to minimize CO<sub>2</sub> gas emission, the electrocatalytic route of energy conversion becomes a key issue because the electricity produced by renewable sources of energy, like solar and wind, enables the conversion of CO<sub>2</sub> into valuable chemical feedstocks. During the last decades several materials, which are able to electrochemically reduce CO<sub>2</sub> to hydrocarbons in aqueous solution, were identified, but their efficiency and stability were not suitable for applications. In this direction, copper is unique due to its ability to electro-reduce CO<sub>2</sub> to hydrocarbons and alcohols in aqueous electrolytes, as probed by Hori et al<sup>1</sup>. Nevertheless, the selective electroreduction of CO<sub>2</sub> into fuels is challenging due to the multiple complex proton-coupled electron transfer steps that occur<sup>2</sup>. This complex network makes the cathodic CO<sub>2</sub> reduction reaction (CO<sub>2</sub>RR) run with relative low current density and high overpotential and in addition electrode deactivation may occur over time. Moreover, the lack of information on the electronic structure of Cu during both the fabrication process and under the catalytic reaction makes it difficult to design more efficient and stable electrocatalysts. By tracking the electronic structure of the Cu catalysts, using in situ X-ray spectroscopies, we were able to tune and precisely set the initial Cu redox state, such as Cu<sup>0</sup>, Cu<sup>+</sup> and Cu<sup>2+</sup>, by controlled applied potential protocols<sup>3</sup>. Also, we traced the variations and modifications in the electronic structure (oxidation state) of the Cu catalysts during applied potential scans or steps and, in particular, under catalytic CO<sub>2</sub>RR conditions. These experiments yielded unambiguous information of the catalyst redox processes governing the CO<sub>2</sub>RR, as well as the nature of the active sites. In addition, we revealed the active/inactive and stable/unstable oxidation states depending on the applied potential and the electrolyte. Here, we report on the preparation of catalysts and on the in situ monitoring of their electronic structure during preparation and electrocatalytic reaction for the following systems:

1. Electrodeposited Cu with different oxidation state
2. PVD copper with different thicknesses
3. oxide-derived copper (OD-Cu)
4. Electrodeposited alloys: CuNi and CuZn

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## In operando XPS and XAS on oxygen evolution reaction catalysts

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Knowledge of the oxidation state of electrocatalysts under operating conditions is essential for proper understanding of their function. Methods to obtain such knowledge are, however, limited. Here, we present a new approach for interfacing a near-ambient pressure XPS end station with a flexible electrochemical cell, and its application to oxygen evolution reaction (OER) catalysts.

Catalyst nanoparticles deposited on a proton exchange membrane and covered with graphene. The proton exchange membrane, which is in contact with electrolyte on the back, supplies a steady flow of electrolyte to the electrodes. Meanwhile, the X-ray and electron-transparent graphene layer greatly reduces the evaporation of water into the NAP-XPS chamber. We show that this can lead to the formation of a thin layer of liquid electrolyte between graphene and the polymer membrane. Thus, electrocatalysts can be probed under operating conditions using surface-sensitive soft X-ray XPS and XAS.<sup>1,2</sup>

With this methodology, we have studied the potential-driven restructuring of Ru, Ir, Pt and Au oxides in 0.1 M H<sub>2</sub>SO<sub>4</sub> during the oxygen evolution reaction. Using 4f XP spectra or 3d M-edges X-Ray absorption spectra, we reveal the distribution of cation oxidation states at the surface of the catalyst particles. Meanwhile, the combination of O K-edge spectra and DFT simulations identifies the deprotonation behavior as a function of potential and coordination number. Due to the high detection efficiency of the employed method, the oxidation and deprotonation kinetics were followed with a time resolution on the order of seconds.

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## Synthetic Manganese Oxides at Water-Oxidation Catalysis Conditions: Phase Changes and Surface vs. Bulk Properties

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Synthetic powders of the manganese oxide (MnOx) phases birnessite, bixbyite, hausmannite and manganosite were screen-printed onto FTO-coated glass and sintered in air at 450°C.<sup>1</sup> The prepared pristine oxide layers were then conditioned at WOC potentials in a neutral phosphate buffer. Pronounced differences between surface and bulk sensitive methods were detected for MnO<sub>x</sub> as powders, as pristine electrodes and after extended operation as WOC electrocatalysts. For surface sensitive analyses, synchrotron radiation based Near Edge X-ray Absorption Fine Structure (NEXAFS) at the Mn L- and O K-edges and X-ray photoelectron spectroscopy (XPS) were used. The bulk was characterized with X-ray diffraction (XRD) and infrared spectroscopy (IR) and morphology with electron microscopy (SEM, TEM). For K-birnessite, a structural amorphization from the synthesized powder via the pristine electrode to the post-operando sample was observed. This is accompanied by an increase in the hydroxide groups after WOC, while the average Mn oxidation of +4 was hardly changed. No significant differences between the bulk and the surface were found, which could be attributed to the known porous and flexible structure of birnessite materials. Due to the easily oxidized Mn<sup>2+</sup> cations present in manganosite (MnO), some Mn<sup>3+</sup>/Mn<sup>4+</sup> ions are already present at the surface of the powder when exposed to air. From the sintering step during preparation, the MnO-coated electrodes exhibit surface oxidation to hausmannite (Mn<sup>2+</sup>/Mn<sup>3+</sup>) and phase changes of the bulk to a mixture of MnO (Mn<sup>2+</sup>), Mn<sub>3</sub>O<sub>4</sub> (Mn<sup>2+</sup>, Mn<sup>3+</sup>) and Mn<sub>2</sub>O<sub>3</sub> (Mn<sup>3+</sup>). At WOC conditions, a further oxidation to a Mn(IV)-rich birnessite was observed for the surface of the catalyst material. In the cases where birnessites are the WOC catalysts (either from the synthesis or formed from MnO during operation), both the electrode surface and the bulk of the porous material seem to contribute to the electrochemical process. Additionally, the flexibility of their structures might explain the better performance of these two electrodes. Much worse WOC rates are found for hausmannite, where the bulk material stays unchanged and only the surface is not fully oxidized to birnessite. However, as Mn<sub>3</sub>O<sub>4</sub> is known for poor carrier transport, the electrical conductivity across the 0.1-1 μm large oxide grains to the catalytic surface layer may inhibit the performance of these electrodes. Finally, the rigid structure of bixbyite (Mn<sub>2</sub>O<sub>3</sub>) seems to be largely unaffected by both the preparation and the WOC operation steps and the only observed change is a 3 nm thick layer of hydroxides at the surface of such electrode found after electrochemical testing. Nevertheless, here the good conductivity of the bulk material might explain why bixbyite is still a much better WOC electrocatalyst than hausmannite.

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## In-situ Synchrotron Radiation based Studies on MnO<sub>x</sub> Water-Oxidation Catalysts: Electronic Structure and Reaction Mechanism

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The presented studies are based on the results obtained by in-situ synchrotron radiation spectroscopy using soft X-rays (Mn L- and O K- absorption edges) for simultaneous electrochemical and spectroscopic experiments on manganese oxide water-oxidation catalysts. In these spectra, the dipole selection rule allows us to observe transitions involving empty 3d states of Mn (2p→3d) as well as empty O 2p orbitals (1s→2p), thus making it possible to monitor changes in the oxidation state of Mn and the Mn 3d/O 2p hybridization as a function of applied bias. Furthermore, the different information depths of the surface sensitive total electron yield (~10 nm) in contrast to the bulk sensitive fluorescence yield (FY, ~30 μm) or the transmission mode (information from the entire catalyst volume) also made it possible to study these potential-induced changes for different parts of the MnO<sub>x</sub> catalyst layer. To carry out the advanced surface sensitive spectroelectrochemical measurements an electrochemical cell containing a Nafion film was used to separate the pressures of 1 bar in the electrolyte from the mbar pressures on the side of the X-ray spectrometer. The price to pay in this surface sensitive detection setup was that the MnO<sub>x</sub> catalyst had to be placed on the low-pressure side of the Nafion membrane, in order to keep liquid water within the catalyst an overlayer of graphene was applied.<sup>1</sup> For the fluorescence and transmission approaches, a detection through a silicon nitride membrane covered with 20 nm of gold could be realized and this allowed a true measurement of the catalyst immersed in the liquid electrolyte. A series of previous ex-situ experiments on different synthetic MnO<sub>x</sub> helped to identify amorphous, layered Mn(III/IV) oxides (so-called birnessites) as suitable samples for the above described in-situ studies. For all samples, the surface (TEY) as well as the bulk sensitive (FY, transmission) Mn L-edge spectroscopy indicated an increase of the Mn<sup>4+</sup> concentration at water-oxidation conditions (confirmed by the detection of the O<sub>2</sub> product). At the same time an increase in the covalent character of the Mn-O bonds led to increases of the O K-edge white line.<sup>2</sup> These phenomena were observed both for neutral pH 7 (0.1M phosphate buffer) and alkaline (0.1M KOH) solutions.

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## SO<sub>x</sub> species on silver in alkene epoxidation

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The nature of the oxygen species that can produce ethylene oxide (EO) on Ag catalysts have recently been identified as originating from adsorbed SO<sub>4</sub> (SO<sub>4,ads</sub>). On a surface free of oxygen SO<sub>4</sub> forms an unreactive SO<sub>4</sub>(7 × √3)rect surface reconstruction. The Ag/SO<sub>4</sub> reconstruction is partially lifted when oxygen induced surface reconstructions are formed. This results in the formation of SO<sub>4,ads</sub>. DFT calculations and TPR experiments show SO<sub>4,ads</sub> can produce EO. NAP-XPS experiments demonstrated that the EO selectivity tracks the coverage of SO<sub>4,ads</sub>.<sup>1</sup>

To prove the relevance of these findings for the industrial process in-situ Raman characterization of Ag powder was done at 1 bar. In-situ Raman shows the presence of a peak at 996 cm<sup>-1</sup> under ethylene epoxidation conditions. This peak at 996 cm<sup>-1</sup> is very close to Ag<sub>2</sub>SO<sub>4</sub>, however it does not match the literature value of 962 cm<sup>-1</sup>.<sup>2</sup> A possible candidate for this Raman peak is then the SO<sub>4</sub>(7 × √3)rect phase, which has a computed S-O stretch mode of 985 cm<sup>-1</sup>.

In direct propylene oxidation Ag favors total combustion.<sup>3</sup> DFT calculations show SO<sub>4,ads</sub> can produce PO. Thus, we use NAP-XPS to study the Ag surface under propylene epoxidation conditions. We find that SO<sub>4,ads</sub> is not present under steady state propylene oxidation conditions. SO<sub>4,ads</sub> can, however, be formed on the Ag surface by introducing an SO<sub>2</sub> pulse into the reaction feed. This results in an increase in selectivity to PO. As for ethylene epoxidation, it appears that SO<sub>4,ads</sub> is active for propylene epoxidation. However, while under ethylene epoxidation SO<sub>4,ads</sub> is stable on the Ag surface, SO<sub>4,ads</sub> is titrated under propylene oxidation conditions and SO<sub>3,ads</sub> is seen as a titration product, resulting in a low steady state SO<sub>4,ads</sub> coverage. This is due to a lower atomic oxygen coverage under propylene epoxidation conditions compared to ethylene epoxidation conditions. Low coverage of adsorbed atomic O not only prevents the formation of O/Ag reconstructions, needed to lift the Ag/SO<sub>4</sub> reconstruction, it also precludes the re-oxidation of SO<sub>3,ads</sub> to SO<sub>4,ads</sub>. We conclude that the atomic O coverage on Ag has a critical role in mediating the coverage of the active species SO<sub>4,ads</sub> in alkene epoxidation under steady state conditions. The co-existence of SO<sub>4</sub> and atomic O on the Ag surface appears to dictate the (high) EO and (low) PO selectivity.

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## Untangling Structural Complexity by Chemical Electron Microscopy

High performance heterogeneous catalysts are metastable compounds that adapt dynamically to the chemical potential. In addition, their performance is strongly influenced by structures and compositions beyond translational symmetry. These deviations induce additional complexity into the structure of such functional materials, which are difficult to assess. To date only little is known on how these local structures participate in the catalytic reaction. The electron microscopy group aims to unravel this structural complexity and their roles in the catalytic reaction. Our applied strategy of analysis can deliver new understanding of working catalysts and can disclose chemical potential induced dynamics. The investigative strategy tackles the understanding of electron-matter interaction, the atomic-scale characterization of local structures and the disclosure of reaction induced dynamics using different *operando* approaches.

Due to the metastable nature of catalysts, beam damage, such as knock-damage, radiolysis or thermodynamic ageing, may easily occur during the electron microscopic investigation. To understand the limitation of electron microscopy for beam sensitive materials we performed systematic studies to assess the pristine structure of zeolites. The cation ordering in zeolites afforded the development of numerical post-acquisition workflows (AC 3.1).

We further investigated open orthorhombic  $(\text{Mo},\text{V})\text{O}_x$  and  $(\text{Mo},\text{V},\text{Te},\text{Nb})\text{O}_x$  by controlled electron beam imaging. These catalytically active oxides exhibit a rich structural and compositional diversity on the nanoscale. To elucidate the extension of their participation in the catalytic reaction we developed quasi *in situ* setups to decouple catalysis and analysis. Changes of identical particles can be tracked at different stages of the reaction with atomic resolution (AC 3.2).

Although these quasi *in situ* TEM studies reduce the influence of the electron beam to a minimum their largest drawback is the lack of real time information. We have implemented routines to conduct *operando* measurements inside the chamber of an environmental scanning electron microscope (ESEM, AC 3.3) and inside the column of a transmission electron microscope (TEM, AC 3.4). Recent developments for the ESEM include the possibility to investigate contamination-free oxophilic metallic surfaces and individual nanoparticles of powder catalysts under relevant reaction conditions. Using this setup, we were able to experimentally prove that the phase transition from a metastable NiO to metallic Ni can initiate a catalytic reaction (AC 3.3). In addition, our homebuilt gas feeding and analysis setup for *operando* TEM measurements allows to disentangle chemical potential induced morphological and structural changes. As we will show on AC 3.4 morphological and structural changes can be correlated to different catalytic regimes. Morphological transformations were found to be thermodynamically controlled and correspond to surface faceting. Structural dynamics occur in the bulk and are the key to maintaining a high catalytic activity.

Local and global scale analysis of catalysts under static and *operando* conditions with different techniques of electron microscopy showed that structural complexity is a general phenomenon in heterogeneous catalysis. This complexity can be increased during reaction. Using this multi-modal approach, we are able to convert visual observables into understanding.

## Implementation and Development of Methodological Strategies for TEM Characterisation of Catalysts

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Functional materials that are applied as heterogeneous catalysts vary greatly in composition, structure and morphology. These inhomogeneities on the local scale influence their physical and chemical properties and can be followed by electron microscopy. In order to describe the relevant atomic and molecular structure of such materials, each transmission electron microscopy (TEM) characterisation experiment has to be adjusted and individually optimized in order to avoid denaturing effects and to be able to image catalysts in their pristine states. This information of the pristine sample is a first step towards establishing new structure-properties relationships.

Here, we will present methodological strategies to elucidate the pristine structure of heterogeneous catalysts. Initially, beam damage studies are applied to understand electron-matter interactions and limitations given by the electron irradiation. In addition, the strategy includes new routes of sample preparation in order to achieve homogeneous and pre-oriented TEM samples. This procedure enhances reproducibility and enables quantification. Quantification of electron irradiation induced effects can be obtained by combining imaging, diffraction and spectroscopy and allows to extract threshold values at which it is possible to investigate the sample before its irreversible destruction or structural modification. Such strategies tackle the dependency of the electron dose, time and beam energy on the damage. Although in some cases the beam damage studies suggest to acquire images with a low signal-to-noise ratio, these images can still contain sufficient information for the application of post-acquisition signal recovery workflows. These workflows use algorithms that work by location, identification and classification of repeated substructures. Using this technique, we can enhance the significance of the real data beyond the limits that would be feasible with simple filtering and denoising approaches. It has, thus, been possible to retrieve atomic local and molecular structural information for highly beam sensitive samples ranging from LTA type zeolites to complex and open-structured metal oxides.

As the characterization of any catalyst by TEM is complementary to its chemical analysis, we have further established the ChemiTEM project that aims to generate standard procedures and workflows optimized for non-expert TEM users in material science and chemistry. The standardized workflows and guiding tools for sample preparation and investigation were implemented into a tablet app, helping the users with the decision process during TEM measurements. Experienced users as well as non-expert TEM users have already benefited from the implementation of this project. The ChemiTEM project will deliver a knowledge base of standard procedures and workflows necessary for TEM investigations and can make electron microscopy a standard tool in chemical laboratories.

The wealth of information added by the optimized and developed TEM methodologies and strategies has been of the utmost benefit towards a more complete understanding of a catalyst.

## Exploring Structural Complexity of Heterogeneous Catalysts by Quasi *in situ* (S)TEM

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The performance of a heterogeneous catalyst is strongly influenced by local alterations in its structure. This also includes surface modifications and bulk defects. Such local variations beyond translational symmetry of the morphology and composition at the atomic level can be revealed by (scanning) transmission electron microscopy ((S)TEM). In addition, to track the structural response of a catalytic system to the applied conditions different *in situ* TEM approaches have been developed.<sup>1</sup> However, to date these *in situ* approaches have to be a compromise between image resolution, beam influence, and applicable pressure range.

To overcome these limitations, we have developed quasi *in situ* TEM grid reactors dedicated for atmospheric pressure<sup>2</sup> and elevated pressure up to 30 bar that combine relevant, homogeneous and well-controlled conditions for all catalyst particles with high resolution identical location imaging. We demonstrate the performance of the high-pressure setup at 10 bar using the reduction of a commercially non-stoichiometric iron oxide (wustite) as an example, which is used as a precursor for ammonia synthesis. Using this setup, we are able to get insights into the phase transformation that correspond to the wustite disproportionation into magnetite and iron during its activation process with atomic resolution. We are further able to identify and compare structural changes in the orthorhombic phases of (Mo,V)O<sub>x</sub> and (Mo,V,Te,Nb)O<sub>x</sub> catalysts during catalytic reactions. These kinds of oxides exhibit a significant structural<sup>3</sup> and compositional<sup>4</sup> variability even in their pristine state, which include the presence of structural defects<sup>3</sup> and differences in the occupation and local geometry of certain crystallographic sites. Atomic scale identical location imaging revealed changes that are strongly dependent on the applied conditions including relevant conditions for the activation of the catalysts and the partial oxidation of ethane and propane.

The identified correlations between structure and activity of the catalysts can provide important insights towards prospective knowledge-based tailoring of catalysts with enhanced performance.

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## ESEM -A Versatile Tool for the Study of Heterogeneous Catalysts

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Heterogeneous catalysts are metastable materials that dynamically adapt to the chemical potential during the catalytic reaction.<sup>1</sup> Minute changes in synthesis and testing protocols influence strongly the catalytic performance. Thus, in order to obtain fundamental insights into their working principles, such functional materials have to be investigated under relevant conditions. For instance, important surface dynamics that can occur during a catalytic reaction can be assessed by environmental scanning electron microscopy (ESEM). In order to judge on the relevance of the study the function of the material has to be detected simultaneously.<sup>2</sup>

Here, we will present new developments that have rendered our existing ESEM device into a versatile tool to investigate different processes related to heterogeneous catalysis. Our modifications include the ability to study heterogeneous catalysts under clean and reproducible conditions. As we will show the modified setup allows for the study of surface dynamics that occur during dry reforming of methane (DRM) over oxophilic Ni catalyst. In the low active state the surface is oxidized, while it is metallic in the high active state. The correlation of performance with the visual information revealed that the catalytic reaction is initiated by the phase transition from the oxide to the metal. In addition, we have implemented analytical routines to study not only metal foams or foils, but also non-conductive powder catalysts. ESEM investigations that were conducted on alkaline vanadates supported on silica revealed a phase transition in the activation step for the catalytic dehydrogenation of propane. This observation is in line with differential scanning calorimetric (DSC) data. The developments also include strategies to not only visualize processes that are related to thermal catalysis, but also to electrochemistry. To proof the concept, we studied a Ni-based solid oxide fuel cell for which we observed an increase of the open circuit voltage with the reaction temperature in the oxidation of methane. Furthermore, a dedicated setup was developed to study the liquid-phase electrodeposition of Cu on an electrode film during cyclic voltammetry using a 3-electrode array. The observed surface features can be directly correlated to the applied electrochemical conditions.

With the outlined multi-tool approach, we are now able to link a variety of reaction conditions, with surface states of a variety of different catalysts and catalytic performance inside the chamber of an ESEM. This setup will thus contribute to a more detailed understanding of the working principles of heterogeneous catalysis.

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## ***Operando* TEM for the Study of Heterogeneous Catalysts under Relevant Reaction Conditions**

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Heterogeneous catalysts are metastable materials that adapt to the chemical potential of the environment.<sup>1</sup> The *in situ* generated structures fluctuate locally and are in many cases only stable during operation. This highlights the importance of developing *operando* techniques for spatially resolved electron microscopy. More robust MEMS nanoreactors give rise to new opportunities for *operando* transmission electron microscopy (TEM) studies under relevant working conditions with added detection of conversion.

Here, we show a home-built gas-feeding and analysis setup that is combined with a mass spectrometer for *operando* experiments inside a TEM using commercially available gas-flow TEM holders.<sup>2</sup> Our setup can provide new insights to generate fundamental understanding of how heterogeneous catalysts interact in different gas environments. To test the capabilities of our *operando* TEM setup, we used CO oxidation over Pt nanoparticles as a model reaction. Using this technique we were able to correlate structural and morphological changes of the Pt catalyst nanoparticles with activity.<sup>3</sup> Catalytic cycling inside the TEM and simultaneous recording of bright-field images, selected area electron diffraction patterns, and catalytic conversion data suggested that chemical dynamics have different origins as well as effects on activity. It can lead to morphological transformations and/or structural dynamics. Morphological transformations can be best described as the formation of equilibrated surface facets which are detrimental to the catalytic activity. These transformations are induced by gradients of the chemical potential. Structural dynamics are a result of frustrated phase transitions<sup>4</sup> and occur in the bulk when the chemical potential is constant. They are initiated by reactant diffusion through the Pt nanoparticles. This dynamic process is thought to be beneficial for the catalytic performance. The results were confirmed by findings from global environmental scanning electron microscopy and quasi *in situ* TEM.<sup>5</sup> In addition, we used our *operando* TEM setup to study the process of deactivation in propane dehydrogenation using Pt nanoparticles.

Prospectively our multi-modal *operando* approach will be used to combine the local information gained from *operando* TEM with a more macroscopic information to deliver new insights into the fundamental understanding of how heterogeneous catalysts work under relevant reaction conditions.

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## Reaction Calorimetry: Exploring a Solid Catalyst In-Situ

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Recent discoveries in heterogeneous catalysis in the academic and industrial field have largely been accelerated by the advances in analytical tools and wide application of complementary in-situ methods. To monitor the catalyst's properties on the fly under conditions of the catalytic process always is a non-trivial task. However, it represents a direct approach for mechanistic studies of the heterogeneous solid-gas system which provides experimental evidence on the active state of the working catalyst. Here we report on application of in-situ calorimetry in two different process reactions of high industrial relevance. The first process is ethylene epoxidation on a high-performance silver-based catalyst. This study targets the quantitative assessment and thermochemical properties of reactive oxygen on supported silver catalysts (Ag/ $\alpha$ -Al<sub>2</sub>O<sub>3</sub>). The determination of the binding strength of surface- and subsurface-oxygen is significant for understanding the relevance of the gas-solid oxygen exchange in ethylene epoxidation and its effect on the selectivity towards ethylene epoxide (EO).<sup>1,2</sup> As shown by in-situ calorimetry the reaction of oxygen with the catalyst's surface in its active state is very favorable  $260 \pm 30 \text{ kJ/mol(O}_2\text{)}$ . This effect is explained by the defect-rich silver structure and presence of subsurface oxygen as suggested by theoretical calculations.<sup>3</sup> Energy of interaction decreases with a lower total oxygen content in/on the catalyst. For the oxygen deficient, oxygen poor and pure silver surfaces the values of  $\sim 100$ , 25 and  $< 10 \text{ kJ/mol(O}_2\text{)}$ , respectively, are measured. Our results unambiguously demonstrate that silver catalysts in its active state act as oxygen reservoir. Isotope exchange experiments showed that prestored oxygen <sup>16</sup>O diffuses out of the subsurface region to the surface and incorporates explicitly into the CO<sub>2</sub> (C<sup>16</sup>O<sup>18</sup>O trace). EO in contrary contains only <sup>18</sup>O isotope. This experiment clearly demonstrates that the oxygen travels from the gas phase into the subsurface layer of the catalyst and diffuses back to the surface during the course of the reaction. The second process is the oxidative propane dehydrogenation on the supported liquid phase catalysts which have melting point in the temperature range of reaction. Alkali based vanadates on Aerosil were investigated under reaction conditions in temperature range 300-500°C. It was shown that the melting of the supported phase correlates with drop of activity and increase of selectivity. This phenomenon was shown to have a general character for K, Cs and Rb vanadates.

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## Mechanisms of Deactivation of Supported Pt Catalysts and their Regeneration in Propane Dehydrogenation

Siamak Nakhaie, Hannah C. Nerl, Andrey Tarasov, Jutta Kröhnert, Frank Girgsdies, Annette Trunschke, Katarzyna Skorupska, and Robert Schlögl

Propylene and hydrogen are the two important products of non-oxidative propane dehydrogenation (PDH). Pt-based catalysts offer remarkable propane conversion and propene yields, and therefore have already found the way to industrial PDH1. However, coking and morphological/structural changes that the catalyst may undergo are thought to lead to rapid deactivation and frequent need for costly regeneration. Initially, samples of coked catalyst with increasing time on stream (TOS) were analyzed using Raman spectroscopy, temperature programmed oxidation (TPO) as well as transmission electron microscopy (TEM). The Raman spectra showed contributions mainly from an ordered  $sp^2$ -C phase which increased in amount with TOS, some molecular and H-containing species and very limited amounts of amorphous C. This was confirmed by TPO, where the  $CO_2$  emission peak was consistently shifted to higher temperatures (up to 16 °C) with TOS. TEM also revealed changes in appearance of the coke with TOS. In early stages, loose strands of C wrapped up in tangled balls were found surrounding small Pt particles. With longer TOS, the C became more ordered with several layers of C forming and found in a number of locations including around the support as well as tightly wrapped around Pt particles. The Pt catalyst appeared to have an indirect effect on the coke formation with Pt particles of several nm in diameter supporting the structured coke formation, while smaller particles of < 2 nm did not appear to cause coke buildup. Furthermore, increased faceting of the Pt particles appeared to lead to increased coke formation. In an attempt to systematically optimize a regeneration recipe, the effect of temperature in regeneration of the catalyst in air was investigated using FTIR and findings correlated with TEM imaging. It was found that oxidative regeneration at 600 °C resulted in significant loss of active Pt surface. Despite the harsh regeneration treatment, TEM imaging revealed the presence of some residues of structured coke. In contrast, regenerations below 500 °C, was found to have only slight adverse effects on the catalyst itself. Raman, TPO and TEM showed that the regenerated and subsequently tested catalyst samples contained more highly structured C than coked samples prior to regeneration. Carbon nanotubes were found to have formed around Pt particles in several locations in the regenerated, tested samples. Furthermore, TEM imaging revealed increased faceting of the Pt. Overall, blockage of Pt sites through the coke build-up with TOS and with each regeneration cycle was found to be the major cause of deactivation. Coke build-up is thought to be the result of an interplay between dissolution-precipitation, overgrowth of C deposits followed by condensation to form a hard graphite-like coke where factors such as particle size and faceting play crucial roles in the type and amount of coke build-up.

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## Development of a Multimodal Experiment for the Investigation of the Heterogeneous Chemistry of Liquid/Vapor Interfaces

Holger Oertel, Robert Schlögl, and Hendrik Bluhm

The properties of liquid/vapor interfaces play a major role in natural processes. For example, they strongly influence the abundance and reactivity of trace gas molecules that govern heterogeneous processes in atmospheric and environmental chemistry. In particular aqueous aerosols catalyze heterogeneous reactions in the troposphere and can act as both sinks (e.g.,  $\text{HNO}_3$ ,  $\text{HCl}$ ,  $\text{N}_2\text{O}_5$ ) and sources (e.g., halogen radicals) for atmospheric trace gases.<sup>1</sup> To date, little is known about the concentration of a wide range of solution phase species at the liquid/vapor interface, which can significantly differ from that in the bulk.

Even less is known about the fundamental pathways in heterogeneous reactions of gas phase species at liquid/vapor interfaces and the role of surfactants in these reactions. While there is a wealth of reactor studies that monitor changes in the gas phase composition to conclude upon reactions at the liquid/vapor interface, these methods do not provide direct information about the reaction products and other properties of the liquid/vapor interface. Over the years a number of surface sensitive optical spectroscopies, such as sum frequency generation (SFG) and reflection-absorption infrared spectroscopy (RAIRS), as well as X-ray based methods, such as grazing incidence small angle X-ray scattering (GISAXS) have been developed to investigate liquid/vapor interfaces under realistic conditions. Ambient pressure X-ray photoelectron spectroscopy (APXPS) has been used in the past to determine the concentration of ions near aqueous solution/vapor interfaces in the presence of the equilibrium vapor pressure. It offers the opportunity to investigate these interfaces also under relevant pressures for all important environmental trace gases, such as  $\text{CO}_2$ ,  $\text{NO}_x$  and  $\text{O}_3$ , including the reaction of these gases with surfactants at the interface.

We are currently planning and designing a new experimental setup for a more comprehensive investigation of liquid/vapor interfaces. This instrument will combine spectroscopy, scattering and microscopy for a more complete understanding of reactions at liquid/vapor interfaces. The instrument will combine APXPS, RAIRS, GISAXS and Brewster Angle Microscopy, which deliver complementary information on the composition, chemical nature and structure of the interfacial region. Since diffusion rates are high in liquids it is also important to monitor the bulk of the solution which will be accomplished by X-ray fluorescence measurements. Liquid/vapor interfaces will be prepared using a Langmuir trough setup which allows dynamic control over the surfactants density at the interface. The instrument will use a laboratory X-ray source so that systematic, long term studies of the correct methodology of preparing liquid/vapor interfaces with high control over their chemical purity can be performed. This requires also to design a vacuum-compatible Langmuir trough and force sensor for the surface pressure measurements. Experiments on a prototype instrument, developed for operation at the Advanced Light Source, are described in a different poster.

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## Compression of a Stearic Acid Surfactant Layer on Water Investigated by Ambient Pressure XPS

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Under realistic environmental conditions the interface between liquids and vapor are most likely partly or totally covered by hydrophobic or amphiphilic molecules, which originate either from the bulk of the solution or have adsorbed to the interface from the surrounding gas atmosphere. These mostly carbonaceous surfactants can potentially have a strong influence on the interface chemistry by, e.g., drastically changing the gas transport from the bulk to the atmosphere and vice versa. Recent experiments on the influence of long-chained alcohol surfactant layers on the evaporation coefficient of neat water have shown that an increase in the carbon chain length by four carbons can decrease the evaporation coefficient by several orders of magnitude.<sup>1</sup> Similar effects are also expected in the opposite gas diffusion direction, i.e., for the migration of vapor-borne molecules through a surfactant layer towards the liquid interface.

Langmuir troughs are an excellent method to prepare liquid/vapor interfaces with adjustable surfactant coverage and chemistry. Changes in the packing and the structure of the surfactant layer are traditionally monitored through measurements of the surface pressure as a function of the mean molecular area (MMA). The relatively straightforward nature of a Langmuir trough setup lends itself for a combination with other surface sensitive methods, such as infrared spectroscopy and grazing-incidence small-angle X-ray scattering. Ambient pressure X-ray photoelectron spectroscopy (APXPS) allows to directly observe changes in the surfactant chemistry as well as all the other constituents of the solution with high interface sensitivity. In a proof-of-principle experiment we have combined a Langmuir trough setup with an ambient pressure XPS experiment to monitor the density and orientation of a stearic acid ( $C_{17}H_{34}COOH$ ) surfactant layer on neat water. From the measured C 1s and O 1s peak areas one can quantify the coverage of water by stearic acid as a function of compression, and in addition also determine changes in the orientation of the stearic acid molecules at the surface. For low coverages of stearic acid on water, i.e. at low compression, the O 1s signal from water is expected to be less attenuated by stearic acid and there are less stearic acid molecules per unit area at the water surface, resulting in a low C 1s/O 1s peak area ratio. This ratio will increase as the stearic acid layer is compressed. Changes in the orientation of the stearic acid molecule with respect to the water surface can be detected through changes in the C 1s signal of the  $CH_x$  chain ( $I_{CH_x}$ ) with respect to that originating from the acid group ( $I_{COOH}$ ), where it is expected that this ratio will increase with increasing upward tilt of the hydrophobic  $CH_x$  chains. Our measurements do indeed show these trends and correlate well with off-line compression curves from surface pressure measurements. These measurements also show that even with careful preparation the surface of neat water is always at least partially covered by a carbon contamination layer, which demonstrates that better methods for the controlled preparation of static liquid/vapor interfaces need to be developed.

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## Department of Heterogeneous Reactions

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The energy challenge can be seen as the major challenge for today's society and future generations. Chemistry plays a central role in the energy challenge, since most energy conversion systems work on (bio)chemical energy carriers and require, for their use, suitable process and material solutions. The enormous scale of their application demands optimization beyond the incremental improvement of empirical discoveries. For this reason, we work on the development of knowledge-based systematic approaches in order to arrive at scalable and sustainable solutions.

Analysis of the processes that are essential to convert the current energy systems into sustainable systems indicates that the conversion of electricity into chemical energy is a critical process in the network of chemical energy conversion reactions. Both electrolysis and heterogeneous (photo)chemical reactions are of relevance here. The difficult elementary steps are in the oxygen evolution reaction.

In a concerted effort the department develops a concept of carbon-based functional materials that operate in oxygen evolution either alone or doped with functional transition metal oxides. In parallel, we study with advanced in-situ spectroscopic tools the reaction on performing systems to learn about design requirements for systems operating with materials as used in the biological analogue. The resulting material solutions and synthesis tools will be transferred to catalytic processes binding primary hydrogen onto carrier molecules such as CO<sub>2</sub> and N<sub>2</sub> to arrive at practically useful solar fuels.

The work of the department is strictly knowledge-oriented to generate generic insight and solutions for synthesis and analysis of chemical energy conversion systems. Theory and molecular model studies with the other departments critically deepen our insight. The department engages into method development for operating advanced spectroscopic methods on heterogeneous and on homogeneous systems, as well as advanced data analytics methods to gain deep insights from large quantities of data. Projects are performed in close collaboration with the Fritz-Haber-Institute of the Max Planck Society in Berlin.

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## EMIL: Bridging the “Pressure Gap” in Spectromicroscopy

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Determine the electronic structure of an electrified interface under reaction conditions is key in the development of next generation of energy materials used for example in batteries and/or electrocatalysis. This requires simultaneously capturing of the electronic structure, morphology and chemical composition that an electrode of interest undergoes in contact with an electrolyte and in the presence of an applied electrical bias<sup>1</sup>. However, the lack of surface sensitive techniques able to monitor a catalyst's electronic structure under reaction conditions in aqueous environments hinders the detailed understanding of such processes. Recently, the advent of reaction cells based membranes that are electron transparent at low kinetic energy has been stimulated significant interest<sup>2</sup>. Access to tunable X-ray sources with suitable photon energy range and photon flux is indispensable to facilitate these in situ studies of gas/solid and liquid/solid interfaces, in particular for electrochemical experiments. EMIL, the Energy Material In-situ Laboratory Berlin at BESSY II, a joint project of the Helmholtz-Zentrum Berlin (HZB) and the Max Planck Society represented by the FHI and the MPI CEC, seized this need. This novel facility includes a new laboratory building hosting beamlines providing an unusually broad photon energy range from the soft to the tender/hard X-ray regime, i.e. 80eV-8000eV, in one spot<sup>3,4</sup>. In addition, this energy range is not so common enabling XANES spectroscopy at useful absorption edges not accessible in most of the beamlines around the world. The soft X-ray branch is under commissioning now, whereas the implementation of the tender X-ray branch is close to its completion.

The EMIL facility will be expanded significantly with the implementation of complementary microscopic in situ capabilities. For that purpose an in situ STXM (scanning transmission X-ray microscope) is under construction. This spectromicroscopy setup combines in one instrument the possibility to acquire the electronic structure of the element of interest with a lateral resolution of a few nanometers, providing a powerful link between high resolution morphology and electronic structure, under reaction conditions i.e. in presence of liquids or gases at controlled temperature and potential. STXM is a bulk sensitive technique that provides information of the whole catalyst, however it is expected that the surface sensitivity of this technique will be enhanced by combining nanofabrication with new detection modes enhancing the surface sensitivity of this method and thus yielding valuable information of the functional interface with noticeable lateral resolution. High spatial resolution is essential for any investigation that studies a catalyst on a local scale under a dynamic feed.

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## Research in Collaboration with BasCat

Annette Trunschke, Frank Rosowski<sup>a</sup>, and Robert Schlögl

Heterogeneous catalysis is crucial for meeting the challenges of our future. The development of sustainable processes based on alternative resources in the chemical industry requires multidisciplinary approaches to surpass the current limits defined by established industrial benchmark catalysts. The Department of Inorganic Chemistry collaborates with the BasCat laboratory at the Technische Universität Berlin, founded in 2011 by the Cluster of Excellence “Unifying Concepts in Catalysis” (UniCat) and the chemical company BASF SE, to develop new design concepts for catalysts and to accomplish a knowledge-based prediction of catalyst performance through scaling from atomistic to industrial level.

The current research program focuses on the activation of C-H, C-C, C-O and N-H bonds as well as molecular oxygen on the surface of high-performance model catalysts. The level of complexity of the well-defined materials needs to assure the functionality of industrial relevant systems.

The materials basis comprises nano-structured metal oxides as well as supported metal nano-particles. Research topics include the investigation of the selective oxidation of short-chain alkanes<sup>1</sup>, synthesis gas reactions<sup>2</sup>, and the direct amination of benzene to aniline as a challenging model reaction to investigate metal-support interactions, the formation of core-shell structures, and alloys.

The sophisticated tasks require the development of new methods for catalyst synthesis and characterization, in-situ spectroscopy, modelling, and data analysis. Data that describe the functional properties as well as the corresponding data and metadata concerning catalyst characterization of fresh and spent materials analysed ex-situ and under operation will be collected and stored in appropriate databases. An important methodical aspect of the work is the definition of the concept of clean data generation in heterogeneous catalysis research. In this respect a “Clean Data Handbook” will be proposed (see Poster AC 1.4).

The research within the collaboration projects with BasCat requires access to all analytical tools established in the Department of Inorganic Chemistry, in particular, kinetic measurements, electron microscopy, X-ray diffraction, thermal analysis, and synchrotron-based methods including the know-how of the experts in these fields.

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## Department of Interface Science

### Poster List

- ISC 1 **Highly-Selective CO<sub>2</sub> Electroreduction under Potentiodynamic Reaction Conditions: Facet-Dependent Electrochemistry and Structural Response of Shape-Selected Cu Nanocubes**  
Rosa M. Arán-Ais, Arno Bergmann, Janis Timoshenko, Fabian Scholten, Clara Rettenmaier, Antonia Herzog, Beatriz Roldan Cuenya
- ISC 2 **In Situ AFM on Copper Surfaces Relevant to CO<sub>2</sub> Electroreduction**  
Georg H. Simon, Christopher S. Kley, Beatriz Roldan Cuenya
- ISC 3 **Spectromicroscopic Investigation of Plasma Activated Copper for Carbon Dioxide Electroreduction Reaction**  
Liviu C. Tănase, Mauricio J. Prieto, Lucas de Souza Caldas, Philipp Grosse, Fabian Scholten, Thomas Schmidt, Beatriz Roldan Cuenya
- ISC 4 **Ultra-High Vacuum-Based Design and Characterization of Model Single-Crystal Copper Electrodes for CO<sub>2</sub> Electroreduction**  
Juan Jesus Navarro, Rosa M. Arán-Ais, Sebastian Kunze, Jens Hartmann, Markus Heyde, Beatriz Roldan Cuenya
- ISC 5 **Enhanced CO<sub>2</sub> Electroreduction towards Multicarbon Products via Shape- and Size-Controlled Nanostructured Copper Catalysts**  
Philipp Grosse, Fabian Scholten, Ilya Sinev, Miguel Bernal Lopez, Beatriz Roldan Cuenya
- ISC 6 **Direct Observations of Copper Nanocube Growth during Electrochemical Cycling**  
See Wee Chee, Rosa M. Arán-Ais, Ruben Rizo, Philipp Grosse, Beatriz Roldan Cuenya
- ISC 7 **Crystalline NP-Decorated Cu<sub>2</sub>O Nanocubes for CO<sub>2</sub> Electroreduction: Catalyst Design and Operando Structure-Composition-Reactivity Insight**  
Rosa M. Arán-Ais, Antonia Herzog, Clara Rettenmaier, Yong-Wook. Choi, Ilya Sinev, Janis Timoshenko, Beatriz Roldan Cuenya
- ISC 8 **Size and Composition Effects in the Electroreduction of CO<sub>2</sub> on Mono- and Bimetallic Nanoparticles**  
Hyo Sang Jeon, Miguel Bernal Lopez, Ilya Sinev, Arno Bergmann, Janis Timoshenko, Beatriz Roldan Cuenya
- ISC 9 **Electrochemical Reduction and Structure Evolution in Copper-Zinc Nanocatalysts: Deciphering In Situ EXAFS Data Using Neural-Networks**  
Janis Timoshenko, Hyo Sang Jeon, Beatriz Roldan Cuenya
- ISC 10 **Operando NRXS and XAFS Investigation of Segregation Phenomena in Fe-Cu and Fe-Ag Nanoparticles during CO<sub>2</sub> Electroreduction**  
Sebastian Kunze, Miguel Bernal Lopez, Philipp Grosse, Ilya Sinev, Ioannis Zegkinoglou, Nuria Jimenez Divins, Janis Timoshenko, Beatriz Roldan Cuenya
- ISC 11 **Pt-Sn-Co Nanocubes as Highly Active Catalysts for Ethanol Oxidation Reaction**  
Rubén Rizo, Arno Bergmann, Fabian Scholten, Yen-Ting Chen, Beatriz Roldan Cuenya

- ISC 12     **Size Effects and Active State Formation of Model Oxide Nanoparticle Catalysts: Co-Based Oxides during Oxygen Evolution Reaction**  
Felix Haase, Arno Bergmann, Beatriz Roldan Cuenya
- ISC 13     **Encapsulation of Copper Oxide Structures through a Silica Thin Film**  
Juan Jesus Navarro, Markus Heyde, Beatriz Roldan Cuenya
- ISC 14     **Surface Action Spectroscopy of Thin Film Oxide Surfaces**  
Yun Liu, Helmut Kuhlbeck, Beatriz Roldan Cuenya
- ISC 15     **Thin Film Catalyst Models**  
Earl M. Davis, Helmut Kuhlbeck, Beatriz Roldan Cuenya
- ISC 16     **Thermal Properties and Particle-Support Interactions in Platinum Nanoparticles Revealed by Advanced Analysis of In Situ XAS Data**  
Janis Timoshenko, Mahdi Ahmadi, Beatriz Roldan Cuenya
- ISC 17     **Particle Shape Effects in Cu-based Catalysts of CO<sub>2</sub> Hydrogenation to Methanol**  
David Kordus, Simon Widrinna, Nuria Jimenez Divins, Beatriz Roldan Cuenya
- ISC 18     **Towards Rational Design of Fe-based Catalysts for CO<sub>2</sub> Hydrogenation to Olefins**  
Mauricio Lopez Luna, Osman Karslioglu, Jian Qiang Zhong, Shamil Shaikhutdinov, Beatriz Roldan Cuenya
- ISC 19     **Ambient Pressure XPS Studies of CuZn Nanoparticles in CO<sub>2</sub> Hydrogenation**  
David Kordus, Osman Karslioglu, Mauricio Lopez Luna, JianQiang Zhong, Shamil Shaikhutdinov, Beatriz Roldan Cuenya

## Highly-Selective CO<sub>2</sub> Electroreduction under Potentiodynamic Reaction Conditions: Facet-Dependent Electrochemistry and Structural Response of Shape-Selected Cu Nanocubes

Rosa M. Arán-Ais, Arno Bergmann, Janis Timoshenko, Fabian Scholten,  
Clara Rettenmaier, Antonia Herzog, Beatriz Roldan Cuenya

The electrochemical conversion of CO<sub>2</sub> to chemicals and fuels powered by the electricity derived from renewable energy sources is a promising strategy towards sustainable energy. Highly active and selective electrocatalysts for multicarbon products are urgently needed to improve the energy efficiency of the CO<sub>2</sub> reduction reaction (CO<sub>2</sub>RR). Therefore, a multi-pathway strategy is applied considering the design of advanced catalysts with tailored activity/selectivity-determining properties and electrochemical routines leading to an *in situ* transformation towards highly selective catalysts and/or to optimized reaction conditions for C<sub>2+</sub> formation.<sup>1,2</sup>

In this work, we present strategies to gain simultaneous control over the surface structure and composition of Cu single crystal electrodes achieved by using a pulsed potential technique. Quasi *in situ* X-ray photoelectron spectroscopy (XPS) helped us to identify the oxidation state of surface species generated by the anodic pulses, while the surface structure of the electrodes was monitored by cyclic voltammetry (CV). Our results point out that the concurrency of (100) sites and Cu(I) surface species are the best combination towards the C<sub>2+</sub> products pathway.

Following these findings, we prepared shape-selected Cu<sub>2</sub>O nanocubes exposing (100) facets and investigated their catalytic activity under potentiodynamic (pulsed) CO<sub>2</sub>RR conditions and compared it to the corresponding results under potentiostatic conditions. To identify the dynamic response of the nanocubes, in particular, metal oxidation state, local atomic structure as well as crystallographic structure, we applied various *in situ* (synchrotron-based) methods.

With *in situ* high-energy X-ray diffraction (HE-XRD), we can follow the fast reduction process of the Cu<sub>2</sub>O nanocubes to metallic Cu under CO<sub>2</sub>RR conditions and the subsequent slow changes in the coherence length reflecting the crystallite/domain size in the nanocubes. Interestingly, we identified that the Cu lattice spacing and thus the Cu-Cu distances follow the potential pulses. Complementary *in situ* X-ray absorption spectroscopy shows a reversible partial formation of oxidized Cu atoms under potentiodynamic conditions, which is in agreement with the findings from quasi *in situ* XPS for single crystals.

Thus, our study gives a comprehensive structural insight into C<sub>2+</sub>-forming state of Cu based catalysts and will thus shed light into the selectivity-determining catalyst properties for CO<sub>2</sub>RR.

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## In Situ AFM on Copper Surfaces Relevant to CO<sub>2</sub> Electroreduction

Georg H. Simon, Christopher S. Kley<sup>a</sup>, Beatriz Roldan Cuenya

Observing surfaces in fluids while reactions proceed is a logical extension of ultrahigh vacuum (UHV) based surface science. Structural and morphological changes of catalysts under reaction conditions are thermodynamically and kinetically among the most relevant deviations from the equilibrium states typically observed. However, *operando* methods analysing properties of the surface during reaction while monitoring reaction products are still far from being fully established. Currently known approaches are extended and new ones are under development.

In this context, we are utilizing *in situ* atomic force microscopy (AFM), i.e. electrochemical AFM<sup>1</sup> experiments, for the study of copper crystal surfaces under CO<sub>2</sub> electroreduction (CO<sub>2</sub>RR) conditions. Surfaces under study are single and polycrystalline copper surfaces after UHV preparation or electropolishing, which are suitable to model morphological aspects relevant to CO<sub>2</sub>RR.<sup>2</sup>

On UHV prepared Cu(hkl) single crystal facets, large morphological changes are observed upon contact with the electrolyte. Nevertheless, atomically stepped surfaces can be established under reducing conditions.<sup>3</sup> From this defined starting point, we follow morphological changes in potentiostatic and potentiodynamic image series. Preliminary results show drastic and at least partially reversible morphological changes over a wide potential range, which proceed at largely different rates during anodic and cathodic scans. With the combination of electrochemical electrode characterization and local real space morphology information, this work sheds light on the general behaviour of polarized electrode surfaces, as well as on interface dynamics associated with trains of electrochemical potential pulses used to enhance C<sub>n</sub> product selectivity in copper CO<sub>2</sub>RR. Product analysis for correlation with surface reactivity is under development.<sup>4</sup>

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## Spectromicroscopic Investigation of Plasma Activated Copper for Carbon Dioxide Electroreduction Reaction

Liviu C. Tănase, Mauricio J. Prieto, Lucas de Souza Caldas, Philipp Grosse, Fabian Scholten, Thomas Schmidt, Beatriz Roldan Cuenya

For CO<sub>2</sub> electroreduction, copper is unique due to its ability to produce hydrocarbons and alcohols with a significant yield. Interestingly, an oxidized copper surface as a starting electrode enhances the activity and selectivity towards C<sub>2</sub> and C<sub>3</sub> products such as ethylene, ethanol and propanol.<sup>1</sup> It is still unclear, whether this is due to morphological changes or specific oxide sites. In order to gain a better understanding we utilize the SMART spectromicroscope<sup>2</sup> at BESSY-II, combining surface sensitive tools such as XPS, NEXAFS, LEED, XPEEM and LEEM, to investigate both, *in situ* oxygen plasma-activated and *in situ* electro-chemically treated copper samples.

First, we studied the effect of the oxygen plasma treatment over time on two crystalline orientations of copper: Cu(100) and Cu(111). We found that on the more active (100) surface, first solely Cu<sub>2</sub>O is formed and afterwards CuO on top. However, on the (111) surface both oxides structures are formed from the beginning on, but dominated by CuO at the end. Furthermore, LEED revealed the atomic surface structure during the plasma treatment. Whereas on Cu(100) the order of the topmost two to three surface layers disappeared immediately, the Cu(111) exhibited a weak order even after 30 min of plasma treatment. In both cases the surface was rough with a distance between the objects of about 30 nm as seen in LEEM and chemically homogenous as proven in XPEEM. The thermal treatment of these plasma-oxidized Cu single crystals was observed in real time. The oxide film dewetted and a few hundred nanometer large Cu<sub>2</sub>O objects were formed, randomly distributed on the crystalline surface. These islands can be used to support catalytic reactions that need more than one type of active sites [Cu, Cu(I), Cu(II)] on the same substrate, or they can even be used as a support to deposit other metallic species in order to study path-driven coupled reactions.

On the other hand, Cu crystals were also used as electrodes for *in situ* electrochemistry. We applied pulsed voltages (+0.6 V and -1.0 V vs. RHE) and interrupted the process at certain states (OCP, after positive and negative pulse), and investigated the chemical composition, structure and lateral inhomogeneities. After 15 min of pulsed treatment, interrupted at the negative potential, the surface shows the presence of Cu<sub>2</sub>O [Cu(I)] homogeneously distributed, coexisting with some metallic Cu particles a few hundreds of nanometer in size. XPEEM exhibits that the Cu<sub>2</sub>O area is covered by a C species, whereas the Cu particles contain K but are free of carbon.

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## Ultra-High Vacuum-Based Design and Characterization of Model Single-Crystal Copper Electrodes for CO<sub>2</sub> Electroreduction

Juan Jesus Navarro, Rosa M. Arán-Ais, Sebastian Kunze, Jens Hartmann, Markus Heyde, Beatriz Roldan Cuenya

Electricity generated by renewable energy sources such as solar cells or wind turbines could be stored in the form of chemicals and fuels through the electroreduction of CO<sub>2</sub> (CO<sub>2</sub>RR) into hydrocarbons or alcohols. Copper is the electrocatalyst with the highest selectivity into hydrocarbons, in detriment of the competing hydrogen evolution reaction (HER).<sup>1</sup> However, high overpotentials, among other issues, are important obstacles for its use in real applications. The characterization and modification of well-defined model systems by using single-crystal copper electrodes is a promising way to gain insights into such problems. In addition, the implementation of electrode preparation using ultra high vacuum (UHV) facilities in combination with traditional surface science characterization has been proposed as a solution to overcome uncontrolled pretreatment of electrode materials under changing ambient conditions.<sup>2</sup> This approach is now employed to evaluate copper electrodes pretreated with different methods.

Both surface reconstruction and oxidation state of copper play an important role in CO<sub>2</sub>RR. It has been demonstrated that pretreatment of the copper electrode with an oxygen plasma increases the activity to multicarbon hydrocarbons and alcohols due to the generation of subsurface oxygen, which stabilizes Cu<sup>+</sup> species.<sup>3</sup> Following this result, a special chamber equipped with an oxygen plasma source was designed and assembled into the employed UHV system.

Another important issue during electrochemical reaction is the stability of the electrocatalyst. Two-dimensional (2D) materials have been proven to be effective for stabilizing surface reconstructions.<sup>4</sup> In this context, 2D silica is of interest for the modification of the electrocatalyst and likewise also provides chemical selectivity.<sup>5</sup> Taking these ideas into account, a growth study of silica thin films on copper single-crystals has been initiated. The structural stability of these samples has also been tested towards ambient conditions and different solvent solutions. Another goal of this in-progress project is to perform a UHV oxygen plasma treatment of copper single crystals followed by the growth of a 2D silica layer, from which an enhanced performance during CO<sub>2</sub>RR might be expected.

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## Enhanced CO<sub>2</sub> Electroreduction towards Multicarbon Products via Shape- and Size-Controlled Nanostructured Copper Catalysts

Philipp Grosse, Fabian Scholten, Ilya Sinev, Miguel Bernal Lopez, Beatriz Roldan Cuenya

A feasible strategy to improve the electrocatalytic conversion of CO<sub>2</sub> to hydrocarbons is to tune the catalyst morphology as well as chemical state. Utilizing *in situ* and *operando* spectroscopic and microscopic methods, we investigated size- and shape-controlled ligand-free Cu nanocubes<sup>1</sup> as well as high surface area Cu dendrites<sup>2</sup>. From this, we gained insight into the correlation between structure, chemical state, and reactivity during CO<sub>2</sub> electroreduction (CO<sub>2</sub>RR).

X-ray absorption fine-structure spectroscopy (XAFS) and *quasi in situ* X-ray photoelectron spectroscopy (XPS) were used to investigate and define the chemical composition under reaction conditions for both types of catalysts. In addition electrochemical atomic force microscopy (EC-AFM) was applied to monitor dynamic changes in our Cu cube-based catalyst under potential control.

Under reaction conditions, the roughening of the nanocube surface, disappearance of the Cu(100) facets, formation of pores, loss of Cu and reduction of Cu<sub>x</sub>O species was observed. This led to a suppression of the selectivity for multicarbon products (i.e. C<sub>2</sub>H<sub>4</sub> and ethanol) versus CH<sub>4</sub>. Similar behaviour was observed for our Cu-den catalyst, as we saw the Cu<sub>x</sub>O species vanish, accompanied by an increase in the catalysts roughness while preserving the high surface area dendritic structure.

A comparison of our Cu-cubes and Cu-dendrites on different supports (i.e. Cu and Ag/Pt-foils) revealed the importance of the underlying support for the selectivity towards multicarbon products, as we found Cu(I) to persist under CO<sub>2</sub>RR for our Cu-cube catalyst. This led to a higher C<sub>2</sub>/C<sub>1</sub> selectivity whilst for our Cu dendrite catalysts the carbon monoxide originating from the Ag support was further reduced towards multicarbon products resulting in a better overall selectivity.

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## Direct Observations of Copper Nanocube Growth during Electrochemical Cycling

See Wee Chee, Rosa M. Arán-Ais, Ruben Rizo,  
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The electrocatalytic reduction of CO<sub>2</sub> to useful chemicals is increasingly viewed as a viable strategy towards sustainable fuel generation.<sup>1,2</sup> However, the selective reduction of CO<sub>2</sub> to multi-carbon products is still challenging and requires the discovery of electrocatalysts with better performance. To enable the rational design of these electrocatalysts, we need understand how their structural parameters influences their activity and selectivity for the reduction reaction.<sup>3</sup> Recently, our department showed that cubic Cu nanoparticle (NP) catalysts exhibit highly dynamic morphological and chemical changes during the CO<sub>2</sub> reduction reaction and these changes also affect the catalyst selectivity.<sup>4</sup>

To fine-tune the properties of these catalysts, it is important that we also understand how the synthesis parameters control the NP structure and surface morphology. Hence, high spatial resolution insights into the formation of these nanostructures are needed. Liquid cell transmission electron microscopy is an emerging technique that can provide detailed information about nanostructures in a liquid.<sup>5</sup> Furthermore, by incorporating thin film electrodes into these so-called “liquid cells”, we can probe the electrochemical response of specimens encapsulated within an electrolyte. So far, this technique has been used to study the electrodeposition of metals,<sup>6</sup> lithiation processes<sup>7,8</sup> and metallic corrosion.<sup>9</sup> We aim to use this technique to capture the dynamical evolution of these cubic Cu NPs from their initial creation from the precursor Cu solution to their behaviors under reduction reaction conditions.

Here, we will show that we can understand from our experiments why the described synthesis protocol<sup>4</sup> generates cubic NPs. Although randomly shaped NPs were deposited on the electrode during the deposition stage of the voltammetry cycle, the cubic NPs were the most stable against dissolution as the applied voltage increases. Hence, repeated cycling leads to a shape selection towards cubic nanoparticles. Further opportunities to extend this approach towards studying the same NPs for the CO<sub>2</sub> reduction reaction will be discussed.

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## Crystalline NP-Decorated Cu<sub>2</sub>O Nanocubes for CO<sub>2</sub> Electroreduction: Catalyst Design and Operando Structure-Composition-Reactivity Insight

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Direct electrochemical conversion of CO<sub>2</sub> into multicarbon fuels and chemicals by using the power of intermittent sources such as wind or solar offers the potential of closing the anthropogenic carbon cycle and store renewable energy. Among the metals studied for the CO<sub>2</sub> reduction reaction (CO<sub>2</sub>RR), Cu has been found to be the most efficient catalyst to produce hydrocarbons due to its binding energy of CO, neither too strong nor too weak for its further reduction into several products such as methane, ethylene and ethanol. Catalytic performance of Cu can be further enhanced by preparing nanostructured catalysts. The observed improvement has been usually linked to structural (facets, defects and low-coordinated sites) and compositional (oxidation state) effects. Furthermore, besides the activity and selectivity, the structural and compositional stability of the nanomaterials during the reaction play a crucial role in the resulting electrocatalytic performance.<sup>1,2</sup>

Recent development of nanostructured electrocatalysts has shown that by using cubic-shaped Cu oxide nanoparticles, which are mainly composed by (100) facets, the selectivity for ethylene is increased while suppressing methane production.<sup>3,4</sup> In addition, bimetallic Cu-M (M = Pd, Ag, Au) catalysts have shown promising activity and selectivity for the production of hydrocarbons,<sup>5-7</sup> although the shape effect has not been explored in depth for bimetallic systems.

In this work, we combine both, shape and bimetallic effects, by successfully preparing well-defined Cu<sub>2</sub>O cubes decorated with metal (M) nanoparticles through a facile wet-chemical, ligand-free synthesis. By means of *ex situ*, *in situ* and *operando* characterization methods, we have gained insight into the morphology and chemical state of the active catalyst during CO<sub>2</sub>RR. This allows a direct correlation of the catalytic performance with the changes in structure, chemical state and surface composition of the catalysts, thus gaining a mechanistic understanding of how the catalysts adapt their atomic and nanoscale properties to the reactive environments.

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## Size and Composition Effects in the Electroreduction of CO<sub>2</sub> on Mono- and Bimetallic Nanoparticles

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Electrochemical CO<sub>2</sub> reduction reaction (CO<sub>2</sub>RR) is a promising process to produce useful chemicals and fuels, but suffers from low energy efficiency and poor product selectivity displayed by metal electrodes. Nanoparticle (NP) catalysts have demonstrated greatly enhanced reactivity and selectivity as compared to their bulk counterparts. In addition, NPs with well-controlled size, shape, and composition are useful to understand catalytic performance trends and design efficient catalysts.

In this work, using the inverse micelle encapsulation synthesis method, Zn, CuZn, and CuCo NPs have been prepared and investigated as catalysts for CO<sub>2</sub>RR. In particular, *in situ* and *operando* spectroscopic methods have been used to gain insight into the correlation between the structure, chemical state, and reactivity of NPs during CO<sub>2</sub>RR. For Zn NPs, a drastic increase in the activity was observed with decreasing NP size, along with an increase in Faradaic selectivity towards H<sub>2</sub>.<sup>1</sup> This selectivity trend was associated with the enhanced content of low-coordinated sites on small NPs.<sup>2,3</sup> In addition, *operando* X-ray absorption fine-structure spectroscopy (XAFS) measurements revealed the presence of residual cationic Zn species in the catalysts during CO<sub>2</sub>RR, which was expected to correlate with the CO selectivity.<sup>4</sup> For CuZn NPs, we found that the alloying of Cu atoms with Zn takes place under reaction conditions and plays a determining role in the product selectivity. Time-dependent XAFS analysis showed that the local structure and chemical environment around the Cu atoms continuously evolves over time due to the presence of cationic Zn species. Such Cu-Zn interaction was responsible for the changes in the selectivity with increasing reaction time. For CuCo NPs, we found that the selectivity of CO<sub>2</sub> reduction could be assigned to reaction-induced segregation of Cu atoms to the NP surface.<sup>5</sup> Density functional theory (DFT) calculations from collaborating groups were used to provide new insights into the surface phenomena and the interaction of Cu facets under expansion and compression with key reaction intermediates. Consequently, various parameters proven from the given NPs systems (*i.e.*, under-coordinated sites, cationic metal species, alloy formation, and surface segregation) can determine the performance of CO<sub>2</sub>RR catalysis. Our findings will be useful for the rational design of more efficient and effective CO<sub>2</sub> reduction catalysts.

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## Electrochemical Reduction and Structure Evolution in Copper-Zinc Nanocatalysts: Deciphering In Situ EXAFS Data Using Neural-Networks

Janis Timoshenko, Hyo Sang Jeon, Beatriz Roldan Cuenya

Extended X-ray absorption fine structure (EXAFS) spectroscopy is a premier method for studies of catalysts (and other functional materials) due to its element-specificity and unique sensitivity to the oxidation state of absorbing atoms and details of the local environment around the absorbers. Moreover, EXAFS analysis is well suited for *in situ* and *operando* investigations, providing the possibility to track the changes in the structure of a catalyst under working conditions. Unfortunately, the intrinsic heterogeneity and enhanced disorder, a characteristic for catalytic materials experiencing structural transformations under harsh reaction conditions hinder the application of conventional data analysis approaches. In particular, bimetallic catalysts under reaction conditions often exist as mixtures of species in different oxidation states with different composition and structure.<sup>1</sup> Identification of these components from experimental data is a challenging task that needs, however, to be addressed to elucidate the main structural motifs responsible for the observed catalytic properties.

Here we approach this problem, by complementing experimental data interpretation with theoretical simulations of catalyst structure and machine learning methods. We demonstrate that an artificial neural network (NN)<sup>2,3</sup> trained on theoretical EXAFS data obtained from molecular dynamics simulations<sup>4</sup> can be successfully used to establish the relationship between EXAFS features and structural motifs in metals as well as oxides and related materials. The trained NN can then be employed to decipher experimental *in situ* EXAFS spectra. Moreover, the ability of the NN to interpret EXAFS data not only for the well-defined initial (e.g., oxidized) and final (e.g., metallic) states of the catalyst, but also for materials undergoing structural transformations,<sup>5</sup> makes this approach especially attractive for *in situ* investigations of catalytic processes. Here, we apply this approach to the interpretation of time-dependent EXAFS data in Cu-Zn nanoparticles employed as a catalyst for electrochemical CO<sub>2</sub> conversion to higher-value chemicals and fuels. Using NN-EXAFS method, we follow *in situ* reduction of copper and zinc species and observe gradual formation of brass alloy under reaction conditions. The observed time-dependent changes in catalyst structure can be linked to the time-dependent selectivity of the catalyst to formation of different reaction products.

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## Operando NRIXS and XAFS Investigation of Segregation Phenomena in Fe-Cu and Fe-Ag Nanoparticles during CO<sub>2</sub> Electroreduction

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Nuclear resonance inelastic x-ray scattering (NRIXS) is a synchrotron technique that probes nuclear vibrational levels, and by extension, enables the extraction of the (partial) phonon density of states specifically for certain element isotopes, such as <sup>57</sup>Fe. This offers information on the structure and composition of materials containing Mössbauer isotopes. NRIXS, and (extended) x-ray absorption fine-structure spectroscopy (E)XAFS were used here to investigate structural and chemical transformations in nanoparticles taking place under electrochemical reaction conditions.

We show here an *operando* application of the former techniques for the investigation of the structure and composition of iron-based (Fe-Cu, Fe-Ag) nanoparticles for the electrochemical reduction of CO<sub>2</sub> (CO<sub>2</sub>RR). We complement this with *operando* EXAFS and extract correlations with their activity and selectivity. The combination of the techniques reveals the evolution of the nanoparticles from an oxidized as-prepared state to a reduced, crystalline state CO<sub>2</sub>RR.

Silver segregation and the formation of a Ag shell in the case of Fe-Ag and Fe-Cu particles is inferred. Similar faradaic selectivity was found for Fe-Ag particles when compared to pure Ag particles of analogous size at -1.1 V versus RHE (reversible hydrogen electrode) in 0.1 M KHCO<sub>3</sub>.

## Pt-Sn-Co Nanocubes as Highly Active Catalysts for Ethanol Oxidation Reaction

Rubén Rizo, Arno Bergmann, Fabian Scholten, Yen-Ting Chen, Beatriz Roldan Cuenya

Electrochemical energy conversion and storage play an essential role in addressing the global energy challenge. In particular, direct ethanol fuel cells (DEFC) represent an attractive alternative to resources based on fossil fuel combustion for portable power generation. Ethanol is an eco-friendly and renewable fuel and its complete oxidation to CO<sub>2</sub> and H<sub>2</sub>O would lead to high energy densities. In addition, the easy handling, storage and transportation of a liquid fuel avoids some of the problems associated with gaseous fuel (e.g. H<sub>2</sub>) systems.<sup>1,2</sup>

Although Pt has been proven to be the most efficient metal catalyst among all the pure metals for the ethanol oxidation reaction (EOR),<sup>3</sup> its high cost and scarcity limits its use in DEFC. To increase the cost/activity ratio of the catalysts, the amount of Pt in the catalyst has to be decreased. Additionally, applying shaped nanoparticles (NP) which expose only the catalytically most active crystal facets will help in this respect.

In the ISC department, we rationally designed Pt-Sn-Co core-shell cubic NPs with outstanding stability and activity toward EOR by controlling the exposed facets and the composition. Valuable mechanistic insights were obtained by combining different *in situ* spectro-electrochemical techniques, such as differential electrochemical mass spectrometry (DEMS) and X-ray photoelectron spectroscopy.

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## Size Effects and Active State Formation of Model Oxide Nanoparticle Catalysts: Co-Based Oxides during Oxygen Evolution Reaction

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Earth-abundant transition-metal oxides are promising anode materials for a cost-efficient water oxidation in alkaline media. Cobalt oxides have been shown to exhibit high catalytic activity for the oxygen evolution reaction (OER) at neutral and alkaline pH. Recently, structurally-different  $\text{CoO}_x(\text{OH})_y$  thin film catalysts were shown to transform until similar structural motifs are observed under reaction conditions, independently of the initial metal oxidation state and metal-oxygen coordination. The high catalytic activity was assigned to the presence of reducible  $\text{Co}^{3+}\text{-O}$  sites<sup>1</sup>. Furthermore, crystalline  $\text{Co}_3\text{O}_4$  spinel-type materials were found to reversibly change their near-surface metal coordination during OER forming mainly di- $\mu$ -oxo-bridged  $\text{Co}^{3+/4+}$  ions.<sup>2,3</sup> In addition, so-called X-ray amorphous catalysts were found to excel in their catalytic activity<sup>4</sup>.

In this work, we applied size-selected nanoparticle model catalysts and investigated trends in catalytic activity and redox electrochemistry. The inverse micelle route is adopted to create oxide nanoparticles with desired size<sup>5</sup> and special emphasis is placed on nanoparticles less than 5 nm in size covering the transition between crystalline and X-ray amorphous nanoparticles. The catalytic activity is rigorously normalized to the catalysts loading determined by ICP-MS, the nominal physical surface area and the electrochemical surface area are determined by various electrochemical methods. Furthermore, we apply *in situ* XAFS and quasi *in situ* XPS for selected nanoparticle sizes to track changes in the oxidation state and local coordination especially under OER conditions, as well as irreversible changes in surface chemistry and catalyst loading. The measurements will be referenced to bulk crystalline  $\text{Co}_3\text{O}_4$  and  $\text{CoFe}_2\text{O}_4$  materials.

We will show the correlations between catalytic activity and the electrochemical surface area proofing. Furthermore, we will show how the degree of metal oxidation under OER conditions vary with the nanoparticle size based on the *in situ* XAFS results. These results allow us to differentiate the degree of surface to bulk oxidation of the NPs during catalysis. This study will give important insights into the catalysis and active state formation of size-selected nanoparticles and represents the basis for future *in situ* investigations.

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## Encapsulation of Copper Oxide Structures through a Silica Thin Film

Juan Jesus Navarro, Markus Heyde, Beatriz Roldan Cuenya

The catalytic properties of copper-based materials are extremely important in a variety of reactions, including methanol synthesis, CO oxidation or CO<sub>2</sub> reduction, among others. These processes are highly sensitive to the oxidation state of copper,<sup>1</sup> which can dynamically change during the reaction, triggering surface reconstructions, determining product selectivity or leading to deactivation. Therefore, the stabilization of the oxidation state of copper is highly desirable. A promising strategy to preserve the properties of a certain catalyst is employing a two-dimensional material as an encapsulating system.<sup>2</sup> In this work, the growth of a silica film on a pre-oxidized Cu(111) surface under ultra-high vacuum conditions is explored.

Well defined copper oxide structures can be grown when exposing a Cu(111) surface to oxygen pressures in the 10<sup>-7</sup> – 10<sup>-5</sup> mbar range and annealing to 473-623 K.<sup>3</sup> The study of these structures has been revisited and, in particular, their electronic properties have been investigated employing scanning tunneling spectroscopy (STS). A band gap of ca. 2.3 eV has been found, while an estimation of the work function was accomplished by looking into their field emission resonances (FER).

It was possible to grow a thin silica film on top of the oxidized Cu(111) surface. The obtained silica film exhibits a hexagonal crystalline structure, according to scanning tunneling microscopy (STM) and low energy electron diffraction (LEED) measurements. However, a vitreous phase was not found, in contrast with other works.<sup>4</sup> Although silica films have been widely studied in the past,<sup>5</sup> its properties as an encapsulating layer are promising to be exploited further. STM images suggest the preservation of the underlying copper oxide structure up to temperatures as high as 1000 K.

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## Surface Action Spectroscopy of Thin Film Oxide Surfaces

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Surface action spectroscopy (SAS) is a vibrational spectroscopy method developed at the Fritz Haber Institute in recent years. It is a derivative of the action spectroscopy method used for vibrational spectroscopy of gas phase aggregates.<sup>1</sup> The working principle is that weakly adsorbed inert gas atoms/molecules are desorbed by surface vibrations excited with infrared light (in the present case coming from the infrared free electron laser of the Fritz Haber Institute).<sup>2-3</sup> A plot of the desorption rate vs. the infrared energy is a vibrational spectrum. Taking spectra of microscopic surface vibrations of dielectric solids is rather straight forward with SAS. Such spectra are fingerprints of the surface structure and can be used for its determination with the help of theoretical model calculations as has recently been shown for  $\text{Fe}_3\text{O}_4(111)/\text{Pt}(111)$ . Presently, copper clusters synthesized via a inverse micelle encapsulation method are being studied. Ordered copper oxide layers on  $\text{Au}(111)$  and copper oxide prepared by oxygen plasma treatment are investigated as reference samples. Crystalline cobalt ferrite layers will be another system to be studied. First results will be presented on the poster.

Another activity is the improvement of cooling, so that temperatures low enough for helium adsorption can be reached, which requires temperatures below 3 K. Helium is very weakly bound, which would extend the usable energy range of the method to lower energy and probably also increase the sensitivity. At present  $\sim 1.5$  K are reached at the sample holder but only about 5 K at the sample. Improvement of the heat conductivity between the sample and the sample holder and an improved radiation shield may solve this problem. At present, molecular hydrogen messengers are used which are more sensitive than neon messengers while having a similar binding energy. We attribute this to the larger number of vibrational coupling channels available for energy transfer into the bond of the messenger to the surface.

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## Thin Film Catalyst Models

Earl M. Davis, Helmut Kuhlbeck, Beatriz Roldan Cuenya

A working catalyst may be very different from the as-prepared one, since the catalyst's morphology and electronic structure depends on the type of reaction, the reaction conditions, and the initial catalyst structure. As a consequence, the catalytic properties of the catalyst also depend on these parameters, since they are intimately related to the catalyst's properties. These aspects are central topics of the group's activities.

This group is part of a German collaborative research program focusing on oxidative catalytic reactions at gas/solid and liquid/solid interfaces using cobalt ferrite based catalysts. The work program involves the preparation of thin crystalline cobalt ferrite films with different terminations and defined stoichiometry variations, and the investigation of the modification of these films under thermal and electrocatalytic reaction conditions. OER (oxygen evolution reaction), as well as alkane, alkene and alcohol oxidation reactions will be studied. Thus, this project combines detailed UHV preparation and characterization studies, partially on an atomic level, with reaction studies under more realistic conditions. The thin film group deals with the UHV part of this project - thin film preparation and characterization. We have already prepared the first ordered  $\text{CoFe}_2\text{O}_4(100)$  films on  $\text{Pt}(100)$ , using a recipe similar to that used for the preparation of  $\text{Fe}_3\text{O}_4(100)$  layers on  $\text{Pt}(100)$ . XPS, STM, and LEED have been applied for characterization.  $\text{CoFe}_2\text{O}_4(111)$  layers on  $\text{Pt}(111)$  will be prepared in the next step using a recipe adapted from that for the preparation of  $\text{Fe}_3\text{O}_4(111)$  layers on  $\text{Pt}(111)$ . The performance of these films under more realistic reaction conditions, at higher pressure and in liquids, will be studied in cooperation with other groups in the department.

## Thermal Properties and Particle-Support Interactions in Platinum Nanoparticles Revealed by Advanced Analysis of In Situ XAS Data

Janis Timoshenko, Mahdi Ahmadi, Beatriz Roldan Cuenya

Understanding the unique properties (e.g., thermal properties and catalytic activity) of small (< 3 nm) metal nanoparticles (NPs) remains a challenging task, both for experimental characterization as well as from theoretical perspective. The intrinsic heterogeneity of these materials, *in situ* transformations of their structure and oxidation state, NP interactions with the support, ligands and adsorbates, surface-induced disorder and strong anharmonicity of atomic thermal motion often are the key characteristics of NPs responsible for their properties and functionality. Probing these effects experimentally, however, is a challenging task, and they are also challenging to account for in theoretical modeling.

Here we address this problem by (i) focusing on a well-defined model system – single-crystal supported size-selected platinum NPs measured in ultra-high vacuum (UHV) – and (ii) combining the experimental characterization (*in situ* X-ray absorption fine structure (XAFS) spectroscopy) of NPs structure with theoretical approaches: reverse Monte Carlo simulations (RMC),<sup>1</sup> classical molecular dynamics (MD)<sup>1</sup> and machine learning (ML) methods.<sup>2,3</sup> In particular, the ML approaches allow us to establish the relation between XAFS spectral features and NP structure descriptors from theoretical simulations, and apply it to the interpretation of experimental *in situ* XAFS data. Using this combined approach, as well as the complementary STM technique, we reveal differences in the interactions between Pt NPs and SiO<sub>2</sub> and TiO<sub>2</sub> supports, and probe the support-induced anisotropy in the bond length distribution within the NPs by analyzing polarization-dependent XAFS data.<sup>4</sup> We also follow the temperature-induced evolution of the structure and dynamics in Pt NPs on SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>, and use ML-XAFS method to extract the accurate asymmetric, non-Gaussian shapes of bond-length distributions in these materials,<sup>5</sup> and to address the long-standing controversy regarding the observed anomalous thermal properties of small Pt NPs: suppressed (or even negative) thermal expansion and enhanced Debye temperature.<sup>6</sup>

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## Particle Shape Effects in Cu-based Catalysts of CO<sub>2</sub> Hydrogenation to Methanol

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The selective CO<sub>2</sub> hydrogenation to methanol is a promising process for establishing a methanol economy and recycling CO<sub>2</sub>. The industrial process for this reaction currently uses Cu/ZnO/Al<sub>2</sub>O<sub>3</sub> catalysts in a gas mixture consisting of H<sub>2</sub>, CO and CO<sub>2</sub>. Although this process is known for many years, the reaction mechanism and the active phase involved in the reaction remain under debate. In this study, we focused on elucidating structure (shape) sensitivity of the Cu based catalysts via synthesis of the Cu cube particles primarily exposing (001) facets.

Copper(I)-oxide cubes can be synthesized using NaOH, ascorbic acid and CuCl<sub>2</sub> or CuSO<sub>4</sub> as described in the refs.<sup>1,2</sup> The recipe was adapted here to tune the size of cube-shaped Cu nanoparticles. Furthermore, it was modified to have Cu cubes encapsulated by Zn species. The cubes (bare or decorated with Zn) were then supported onto ZnO or ZrO<sub>2</sub> supports. The catalytic activity was tested in a fixed-bed flow reactor in the CO<sub>2</sub> and H<sub>2</sub> (ratio 1:3) mixture, and the products were detected with online-GC. Structure and chemical state of the catalysts before and after reaction were characterized by STEM, XPS and XRD.

The catalysts showed reasonable activity for the CO<sub>2</sub> hydrogenation while suppressing the reverse-WGS reaction as compared to “conventional” catalysts having rather ill-defined Cu particles. While the Cu cubes supported on ZnO showed higher activity than those supported on ZrO<sub>2</sub>, the formation of a Zn shell on ZrO<sub>2</sub>-supported Cu cubes further increased the selectivity and improved activity.

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## Towards Rational Design of Fe-based Catalysts for CO<sub>2</sub> Hydrogenation to Olefins

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Chemical conversion of CO<sub>2</sub> via hydrogenation into value-added products such as hydrocarbons, olefins and alcohols is one of the most promising technologies to reduce CO<sub>2</sub> emissions to the atmosphere.<sup>1,2</sup> Among commonly used catalysts based on highly dispersed 3d metals (Cu, Co, Ni, and Fe), the Fe-based catalysts have received considerable attention as they showed high selectivity towards C<sub>2+</sub> hydrocarbons. Since Fe and Co catalysts are active in the Fischer-Tropsch (FT) and reverse water-gas-shift (r-WGS) reactions, the process, often referred to as modified FT (or CO<sub>2</sub>-FT), may also proceed via sequential r-RWGS (to form CO from CO<sub>2</sub>) and FT reactions. It has been proposed that the active phase is an iron carbide layer formed during the reaction.<sup>3</sup> The selectivity towards higher hydrocarbons may be improved by adding another metal (e.g., Cu, Co) or alkali promoters like K. However, full understanding of the reaction pathways, in particular the role of a support material and promoters as well as particle size effects on the catalytic performance are still missing. To get more insight into these issues, we employ model studies aimed at finding structure-reactivity relationships for the Fe based catalysts in CO<sub>2</sub> hydrogenation.

Metal (Fe, Co) and bimetallic (Fe-Co) particles were synthesized by inverse micelle encapsulation and deposited by dip coating onto SiO<sub>2</sub>/Si and Al<sub>2</sub>O<sub>3</sub> (sapphire) substrates that allowed to study model catalysts with narrow particle size distribution and controlled inter-particle distance, measured by AFM. Chemical state and catalysts transformations were monitored *in situ* using NAP XPS in the 1 mbar range of pressures. In addition, XPS studies were performed on samples treated in a high-pressure reactor under the same reaction conditions (pressure and temperature) as used for the preparation of powder catalysts from the same precursor solutions. Further characterization was performed by IR spectroscopy using adsorption of CO as a probe molecule.

The results revealed a considerable resistance of the “as prepared” iron oxide nanoparticles towards reduction in H<sub>2</sub> atmosphere. The particles do not form carbide phase under reaction conditions used (CO<sub>2</sub>/H<sub>2</sub> = 1/3, 10 bar, 350°C), thus suggesting that the reduction step may be crucial for the principal reaction mechanism.

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## Ambient Pressure XPS Studies of CuZn Nanoparticles in CO<sub>2</sub> Hydrogenation

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Cu/ZnO catalysts are commercially used for methanol synthesis from CO<sub>2</sub> and hydrogen and thus are widely explored in the catalytic community. It appears that the intimate interaction of Cu and Zn (ZnO) phases constitutes the key factor for superior catalytic performance of the Cu/ZnO catalysts. In this study, we carried out near ambient pressure (NAP) X-ray photoelectron spectroscopy (XPS) measurements (using both conventional X-ray sources and synchrotron light) of the Cu-Zn Nanoparticles (NPs) to monitor the chemical composition and electronic state of the model catalysts during calcination/reduction treatments as well as in CO<sub>2</sub> hydrogenation reaction conditions in the mbar pressure range. Note that X-ray beam damage effects was minimized in such experiments by regularly changing sample spot position.

The measured Cu/Zn ratio in “as deposited” polymer-free 6 nm large Cu<sub>70</sub>Zn<sub>30</sub> NPs (after *ex situ* O<sub>2</sub>-plasma and *in situ* oxidation at 400°C in ~ 1 mbar O<sub>2</sub>) is close to that expected from the elemental composition of the synthesized particles. Nevertheless, the surface is slightly enriched with Cu oxide species, as extracted from a depth profile analysis using variable photon energy. Reduction in ~1 mbar H<sub>2</sub> at 350°C, however, leads to strong Zn segregation to the surface. Furthermore, the Cu/Zn surface ratio in reduced NPs does not change (at least, considerably) in the CO<sub>2</sub> + H<sub>2</sub> (1:3) reaction mixture at 250 - 350°C at these pressures. Comparative *ex situ* XPS measurements (high pressure reactor cell interfaced to the UHV system) on the samples treated in O<sub>2</sub> and H<sub>2</sub> at 1 bar revealed the same trend, albeit the Zn segregation was not as strong as observed at low pressures in the NAP XPS experiments. Complementary infrared spectroscopy studies of the NPs via adsorption of CO and CO<sub>2</sub> as probe molecules are under way which will shed more light on the surface structure of the prepared NP<sub>s</sub> as well as surface species formed under reaction conditions.

## Department of Molecular Physics

### Poster List

- MP 1 **Photo-electron spectroscopy and photo-electron circular dichroism of cold chiral anions**  
Jenny Triptow, Gerard Meijer, and André Fielicke
- MP 2 **Spectroscopy of neutral boron clusters using tunable VUV generated by 4-wave mixing**  
Sascha Schaller, Alan Günther, Gerard Meijer, and André Fielicke
- MP 3 **Direct Evidence for Neighboring and Remote Participation Revealed by Cryogenic Vibrational Spectroscopy**  
Kim Greis, Eike Mucha, Mateusz Marianski, Sooyeon Moon, Gerard Meijer, Gert von Helden, Kerry Gilmore, Peter H. Seeberger, and Kevin Pagel
- MP 4 **Gas-Phase Analysis of Fluorine-Specific Interactions**  
Rayoon Chang, Waldemar Hoffmann, Christian Manz, Gert von Helden, and Kevin Pagel
- MP5 **Probing the Conformational Landscape and Thermochemistry of Dinucleotides via Helium Nanodroplet Infrared Action Spectroscopy**  
Daniel A. Thomas, Rayoon Chang, Eike Mucha, Gerard Meijer, and Gert von Helden
- MP 6 **Characterization of the Intriguing Products of Fluoride-Carbon Dioxide Chemistry by Helium Nanodroplet Infrared Action Spectroscopy**  
Daniel A. Thomas, Eike Mucha, Maike Lettow, Mariana Rossi, Gerard Meijer, and Gert von Helden
- MP 7 **Towards enantiomer-specific spectroscopy and control of cold, chiral molecules**  
Johannes Bischoff, A. O. Hernandez-Castillo, Ju Hyeon Lee, Marco De Pas, Gerard Meijer, and Sandra Eibenberger-Arias
- MP 8 **Electronic and rotational spectroscopy of cold molecules**  
A. O. Hernandez-Castillo, Johannes Bischoff, Jennifer Langenhan, Henrik Haak, Gerard Meijer, and Sandra Eibenberger-Arias
- MP 9 **Dynamics of polar polarizable rotors acted upon by unipolar electromagnetic pulses**  
Mallikarjun Karra, Marjan Mirahmadi, Burkhard Schmidt, and Bretislav Friedrich
- MP 10 **Microsolvation and rotation of 'large' molecular impurities inside helium nanodroplets upon electronic excitation**  
Mallikarjun Karra, Alexander A. Auer, Mikhail Lemeshko, Giacomo Bighin, Igor Cherepanov, Giovanni Bistoni, Johannes Fischer, Stefan Fuchs, Eva-Maria Lottner, Alkwin Slenczka, and Bretislav Friedrich
- MP 11 **Molecules as a dark matter detector**  
Jesús Pérez-Ríos, Hariskrishnan Ramani, Oren Slone and Rouven Essig
- MP 12 **Accurate potential energy curves of AlF molecules: from quantum chemistry calculations to experimentally derived potentials**  
Jesús Pérez-Ríos, Gerrit Groenenboom, and Gerard Meijer

- MP 13 **The FHI FEL Facility: Status and 2-Color Upgrade Design**  
W. Schöllkopf, S. Gewinner, M. De Pas, H. Junkes, G. von Helden, and G. Meijer,  
W.B. Colson, D. Dowell, S.C. Gottschalk, J.W. Rathke, T.J. Schultheiss,  
A.M.M. Todd, and L.M. Young
- MP 14 **Matter-Wave Diffraction from a Periodic Array of Half Planes**  
Ju Hyeon Lee, Lee Yeong Kim, Bum Suk Zhao, and Wieland Schöllkopf
- MP 15 **Spectroscopic characterization of aluminium monofluoride with  
relevance to laser cooling and trapping**  
Stefan Truppe, Silvio Marx, Sebastian Kray, Maximilian Doppelbauer,  
Simon Hofsäss, H. Christian Schewe, Jesús Pérez-Ríos, Nicole Walter,  
Boris G. Sartakov, and Gerard Meijer
- MP 16 **A cryogenic buffer gas molecular beam of AlF molecules and optical  
cycling on a  $A^1\Pi \leftarrow X^1\Sigma^+$  transition**  
Stefan Truppe, Simon Hofsäss, Maximilian Doppelbauer, Sebastian Kray,  
Jesús Pérez-Ríos, and Gerard Meijer
- MP 17 **Aqueous-phase TiO<sub>2</sub> Nanoparticles: Nature of Water Adsorption and  
Structure of the Electric Double Layer**  
Hebatallah Ali, Robert Seidel, Jie Xiao, Bernd Winter, and Gerard Meijer
- MP 18 **A Unique Setup for Detection of Photoelectron Circular Dichroism from  
Aqueous-Phase Chiral Molecules**  
Sebastian Malerz, Florian Trinter, Christian Schewe, Henrik Haak, André Knie,  
Iain Wilkinson, Uwe Hergenbahn, Laurent Nahon, Ivan Powis,  
Bernd Winter, and Gerard Meijer
- MP 19 **Cryogenic Liquid Jet Photoemission from Alkali-Metal – Liquid Ammonia:  
From Electrolyte to Metallic Solutions**  
Christian Schewe, Hebatallah Ali, Stephan Thürmer, Bernd Winter, Gerard Meijer,  
Stephen Bradforth, and Pavel Jungwirth

## Photo-electron spectroscopy and photo-electron circular dichroism of cold chiral anions

Jenny Triptow, Gerard Meijer, and André Fielicke

Photoelectron circular dichroism (PECD) has been observed in the photoionization of chiral molecules by circularly polarized light. It becomes apparent by an intensity difference of electrons emitted in forward and backward direction with regard to the propagation direction of the light. Its sign changes when the polarization changes from left to right-handed or when changing the enantiomer. While by now intensively studied for the photoionization of chiral neutral molecules this effect has not yet been demonstrated in photodetachment of anions.

The initial approach of producing chiral anions was to form complexes of atomic gold anions and chiral molecules (alaninol, fenchone, menthone, 3-hydroxy-tetrahydrofuran) in an existing laser evaporation cluster source. The classical anion photoelectron spectra revealed a weak interaction of the gold atom and the chiral ligand with only moderate shifts of the observed bands from the binding energies of free Au<sup>-</sup>. For these anionic complexes no PECD on a magnitude seen typically in ionization of neutrals (~10%)<sup>1</sup> has been detected. Possible reasons under consideration were the formation of intrinsically hot complexes in the laser evaporation source, stability and sensitivity issues in the electron detection by the velocity map imaging setup or that anions behave differently compared to the neutrals because of changes in the relevant interactions between the emitted electron and the final state.

For improvements of sensitivity and stability a photo-elastic modulator has been implemented for turning the light polarization from left to right-handed on a shot-by-shot basis. Together with other measures to improve ion and laser beam alignment, mechanical stability, and magnetic shielding a sensitivity for detecting PECD of better than 1 % is now reached. Further, with a combination of discharge source and entrainment by a cold supersonic Ar expansion cold molecular anions<sup>2</sup> can be formed e.g. via deprotonation of alcohols.

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## Spectroscopy of neutral boron clusters using tunable VUV generated by 4-wave mixing

Sascha Schaller, Alan Günther, Gerard Meijer, and André Fielicke

Boron clusters have been found to show interesting binding behaviour with delocalized aromatic systems due to the only three valence electrons of the boron atom. According to previous experiments in combination with calculations, anionic boron cluster<sup>1,2</sup> are able to form versatile structures like planar, ring-like and cages. For neutral B<sub>40</sub> that is suggested to be formed in these experiments by electron detachment from a minor abundant, high energy isomer of the anion, a highly stable cage structure has been predicted<sup>2</sup>. Parallels to the carbon fullerenes might be drawn, which call for a more direct investigation of the neutral clusters' structures.

For this, the neutral clusters can be size-selectively characterized by vibrational spectroscopy using the IR-VUV two color ionization scheme. The resulting infrared spectra are structural fingerprints and, by comparison with calculated IR spectra, can allow for isomer assignment. In IR-VUV two color ionization, VUV photons ionize the clusters near the ionization threshold where the ionization efficiency for cold clusters is low. However, if the clusters can be heated in a preceding resonant IR excitation step a strong increase in the ionization efficiency can be detected. By measuring the ion yield as a function of the IR frequency, cluster size-specific IR spectra can be determined. It should be noted that for a given VUV photon energy, there is access to only a limited number of cluster sizes as the ionization energy depends on cluster size. So far, with a commercial VUV laser (F<sub>2</sub> laser, 7.9 eV) very few sizes of boron clusters (B<sub>11</sub>, B<sub>16</sub>, B<sub>17</sub>) could be characterized<sup>3</sup>. To overcome the limitation in photon energy VUV photons are now generated by four-wave mixing in Xenon. In the present configuration which shall cover an energy range of 6.5 eV to 8.2 eV, two dye lasers interact in a static gas cell and the resulting 2+1 difference frequency is used for ionization. After creating 7.9 eV photons in that way the spectra reproduce the results obtained via F<sub>2</sub> laser ionization.

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## Direct Evidence for Neighboring and Remote Participation Revealed by Cryogenic Vibrational Spectroscopy

Kim Greis<sup>a</sup>, Eike Mucha<sup>a</sup>, Mateusz Marianski<sup>b</sup>, Sooyeon Moon<sup>a,c</sup>, Gerard Meijer, Gert von Helden, Kerry Gilmore<sup>c</sup>, Peter H. Seeberger<sup>a,c</sup>, and Kevin Pagel<sup>a</sup>

The chemical synthesis of glycans is traditionally very cumbersome. While the regiochemistry and branching is controlled *via* elaborate protecting group (PG) strategies, stereoselectivity of a glycosidic bond can be induced by participating PGs. It has been shown that acetyl PGs at the neighboring C2-position interact with the anomeric center to form  $\beta$ -glycosidic bonds, while it is only theorized that acetyl PGs and the remote C4- or C6-positions can induce  $\alpha$ -selectivity<sup>1</sup>. Little is known about the structure of the short-lived intermediate that is formed during these reactions – the glycosyl cation.

As a first step, we have shown that glycosyl cations can be generated and trapped in a mass spectrometer to probe their vibrational modes using cryogenic IR spectroscopy. In combination with first-principles theory, we showed that participating C2-acetyl PGs form a covalent bond with the anomeric carbon leading to dioxolenium-type structures in glucose, mannose, and galactose<sup>2</sup>.

Second, a consistent set of fully benzylated, 4-acetylated, 6-acetylated and 4,6-di-acetylated galactose was investigated in order to determine whether remote participation can occur in these building blocks. Overall an overwhelming match between the experimental and the theoretical IR spectra was achieved leading to high-resolution structures of the respective intermediates. The data clearly shows that remote participation exists in C4-acetyl, but not by C6-acetyl protected galactose. Moreover, a newly discovered type of interactions between benzyl PGs and the anomeric carbon, leading to oxonium-type structures, was discovered. The data was compared to synthetic results, which showed that there is a clear correlation between the gas-phase data and solution-phase chemistry<sup>3</sup>. As a result, the outcome of glycosylation reactions can be predicted. In the future, the presented approach will be used to investigate further protecting group combinations and use the obtained data to tune reactions in glycosynthesis.

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## Gas-Phase Analysis of Fluorine-Specific Interactions

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In the pharmaceutical industry, fluorine is considered a key element to modulate the biophysical and biochemical properties of peptides, proteins and other biomacromolecules. The new interactions emerging from H/F substitution, however, are weak and difficult to study using conventional condensed-phase methods. As a result, fluorine-specific interactions are currently poorly understood. Here we take advantage of the "clean room" environment of the gas phase to obtain fundamental insights on intra- and intermolecular fluorine-specific interactions in molecules and molecular aggregates. In particular, we follow the impact of H/F substitutions by means of size- and shape-sensitive ion mobility-mass spectrometry (IM-MS), gas phase infrared spectroscopy and *ab initio* calculations.

One of the simplest self-assembling model systems in biology are amino acids. At elevated concentrations they spontaneously aggregate into clusters, which are stabilized by an interplay of various non-covalent interactions. Recently, we used IM-MS to systematically determine the geometrical size of the individual clusters and ranked them according to their packing density. Based on this data, a novel hydrophobicity scale solely based on the interaction between individual amino acid was derived.<sup>1</sup> Application of this approach to non-natural amino acids enabled us to study the effect of side chain H/F substitution on the interaction and aggregation propensity. A relative ranking of the investigated amino acids revealed surprising differences in their intrinsic hydrophobicity.

Fluorinated amino acid variants are often implemented in peptides to modulate their proteolytic stability, folding and aggregation properties. To obtain insights on the underlying interactions, we studied the impact of fluorination on the aggregation propensity of the hexapeptide NFGAIL. The peptide NFGAIL is known to undergo a spontaneous transition from a soluble, mostly unfolded form, into insoluble and  $\beta$ -sheet-rich amyloid assemblies. IM-MS results revealed the presence of polydisperse, polymorph, and compact oligomers as well as extended intermediates during the early phase of assembly. Especially the extended forms exhibited an enhanced  $\beta$ -sheet content as shown by gas-phase IR spectroscopy.<sup>2</sup> Kinetic data suggest that fluorination of phenylalanine (F) in NFGAIL significantly accelerates fibril formation and changes the morphology of the resulting fibrils. Surprisingly, compact species with presumably unordered conformation are observed *via* IM-MS and gas-phase IR spectroscopy, which indicates a significantly changed aggregation pathway in fluorinated NFGAIL.

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## Probing the Conformational Landscape and Thermochemistry of Dinucleotides via Helium Nanodroplet Infrared Action Spectroscopy

Daniel A. Thomas, Rayoon Chang, Eike Mucha, Gerard Meijer, and Gert von Helden

The extremely low equilibrium temperature of 0.4 K afforded by the helium nanodroplet environment is ideal for the acquisition of well-resolved infrared spectra. Importantly, the rate at which entrained molecules are cooled is typically much faster than the rate of molecular isomerization, yielding a “shock-freeze” of high-temperature conformational distributions. Thus, cold-ion spectra collected at varying pre-pickup ion temperature can be utilized to investigate conformational distributions and obtain thermochemical parameters, providing a map of the conformational energy landscape. This methodology has been utilized previously to obtain thermochemical parameters (relative enthalpy and entropy) for small neutral species.<sup>1,2</sup> We extend this approach to explore the conformational thermochemistry of deprotonated dinucleotides, which have been shown previously to exhibit significant structural rearrangements with low activation barriers.<sup>3</sup> Deprotonated dinucleotide species were generated by nanoelectrospray ionization and confined in a helium-buffer-gas ion trap at temperatures between 80 and 300 K. The dinucleotide ions were then captured in helium nanodroplets, and infrared action spectra were recorded with varying ion trap temperature. The acquired infrared spectra showed a strong dependence on pre-pickup temperature, consistent with the preservation of conformer populations prior to cooling in the helium nanodroplet. Non-negative matrix factorization was utilized to deconvolute infrared spectra and thereby determine temperature-dependent conformer populations. Relative enthalpies and entropies of conformers were subsequently obtained from a Van't Hoff analysis. These initial results demonstrate the promise of this technique to elucidate competing intramolecular interactions and experimentally measure thermochemical parameters, even for larger molecules.

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## Characterization of the Intriguing Products of Fluoride-Carbon Dioxide Chemistry by Helium Nanodroplet Infrared Action Spectroscopy

Daniel A. Thomas, Eike Mucha, Maike Lettow, Mariana Rossi, Gerard Meijer, and Gert von Helden

Fluoride, as a result of its small ionic radius and high Lewis basicity, often exhibits intriguing chemistry distinct from that of larger halides. In the gas phase or in aprotic solvent, fluoride typically acts as a potent nucleophile. In the presence of protic molecules, fluoride forms strong ionic hydrogen bonds that can lead to pronounced local ordering and the disruption of solvent hydrogen bonding networks. Herein we present an investigation of the complex reaction pathways encountered in nanoelectrospray ionization of an aqueous sodium fluoride solution in the presence of gas-phase carbon dioxide, which involves the intricate interplay of fluoride nucleophilic attack, strong hydrogen bonding, and proton transfer reactions. Two particularly fascinating species produced under these conditions, fluoroformate<sup>1</sup> and a carbonic acid-fluoride complex<sup>2</sup>, were characterized by helium nanodroplet infrared action spectroscopy. The low temperature of ca. 0.4 K provided by the helium nanodroplet environment enables the acquisition of well-resolved infrared spectra that are matched to high-level *ab initio* calculations to enable a comprehensive analysis of the observed structures. Fluoroformate,  $\text{FCO}_2^-$ , has long been of interest as a simple example of reductive  $\text{CO}_2$  derivatization, but the isolated ground-state anion has not been previously characterized experimentally. The carbonic acid-fluoride complex,  $\text{F}(\text{H}_2\text{CO}_3)$ , is produced in abundance under the experimental conditions, suggesting that the complex is highly stable. The ionic hydrogen bonds between the acidic groups of the carbonic acid and fluoride yield substantial anharmonic effects for the vibrational modes corresponding to acidic proton displacement. Specifically, a strong blue-shift of the symmetric hydrogen stretching fundamental relative to predictions from the harmonic approximation is traced to significant coupling between the hydrogen stretching and bending vibrations that results in an effective weakening of the  $\text{OH}\cdots\text{F}^-$  ionic hydrogen bonds.

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## Towards enantiomer-specific spectroscopy and control of cold, chiral molecules

Johannes Bischoff, A. O. Hernandez-Castillo, Ju Hyeon Lee, Marco De Pas, Gerard Meijer, and Sandra Eibenberger-Arias

Chiral molecules are important in nature and exist in one of two mirror-image versions (enantiomers), that cannot be transformed into each other by mere rotation or translation. They are of high importance in many biological and chemical reactions, specifically also in the human body. Even though most physical properties of enantiomers are identical – which makes them intrinsically difficult to separate – their handedness often determines their functionality. There is a plethora of scientific methods to study chirality, however, chiral analysis, particularly for complex samples, is still challenging.

We present the recently developed enantiomer-specific state transfer method<sup>1</sup>, where tailored microwave fields are used to enantiomer-selectively populate or depopulate a chosen rotational state. Three mutually orthogonally polarized, resonant, phase-controlled microwave pulses are used to drive connected rotational transitions, resulting in controlled, enantiomer-specific population transfer. The technique builds on microwave three wave mixing<sup>2</sup>, a recently established method for detecting chiral molecules enantiomer-specifically using microwave spectroscopy.

We present our new experimental setup at the MP department, where we implement the enantiomer-specific state transfer method and we discuss further outlook on its applications for fundamental physics studies as well as for physical chemistry.

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## Electronic and rotational spectroscopy of cold molecules

A. O. Hernandez-Castillo, Johannes Bischoff, Jennifer Langenhan, Henrik Haak, Gerard Meijer, and Sandra Eibenberger-Arias

We present our molecular beam apparatus for rotational and electronic spectroscopy experiments, as well as a new design of a pulsed slit valve.

We designed, built and preliminarily characterized a new, compact rotational spectroscopy setup including microwave inputs with three perpendicular polarizations. This new experiment allows for experiments in the 2-25 GHz regime, making studies of a large variety of molecules possible. For initial tests of the new apparatus, the molecule OCS was used and a free induction decay signal could be successfully measured. In addition, we tested the instruments broadband capabilities by recording the rotational spectrum of benzonitrile in the frequency range from 7-18 GHz. These measurements lay an important foundation for future, enantiomer-sensitive measurements with chiral molecules<sup>1</sup>.

We have investigated various molecular species, of specific interest is 1-Indanol. This alcohol is relatively rigid and simple, and it has a chiral center and an aromatic ring, making it an attractive molecule to explore. We performed resonance-enhanced multiphoton ionization experiments using helium, neon, and argon as carrier gases. The choice of carrier gas determines the abundance of different conformers of a molecule. When using argon as a carrier gas, only the lowest-energy conformer of 1-indanol is observed in the beam. We also performed double resonance experiments to determine the lifetime of the first excited electronic state. For this, two-color, resonant, two-photon ionization was carried out by using a pulsed dye laser for the resonant excitation ( $S_0$ - $S_1$ ). Consequently, with a tunable time delay, an ArF excimer laser was used for ionization out of  $S_1$ . The lifetime of the excited electronic state of the lowest energy conformer of 1-indanol is measured to be around 34 ns. Similar measurements have been performed for two other conformers.

Controlled sources for cold gas-phase molecules are often needed and used in experimental molecular physics and physical chemistry. Pulsed supersonic jet valves are abundantly implemented, also in our experiments described above. Typically, these valves have a pinhole aperture, since these can be opened and closed easily, fast, and efficiently. However, for many spectroscopic experiments it would be desirable for the pulsed valves to exhibit a long, narrow slit as an aperture since slit valves have favourable expansion properties. The technical implementation of such slit valves has shown to be challenging, therefore most past slit valve designs have not found wide application<sup>2</sup>. In the molecular physics department, a new slit valve design based on piezoelectric stack arrays was developed, built, and implemented. We performed characterization experiments of this new design using atomic Argon as a gas and miniature capacitive microphones for detection. First results are promising for future applications in molecular spectroscopy experiments.

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## Dynamics of polar polarizable rotors acted upon by unipolar electromagnetic pulses

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We study, analytically as well as numerically, the dynamics that arises from the interaction of a polar polarizable rigid rotor with single unipolar electromagnetic pulses of varying length,  $\Delta\tau$ , with respect to the rotational period of the rotor,  $\tau_r$ . In the sudden, non-adiabatic limit,  $\Delta\tau \ll \tau_r$ , we derive analytic expressions for the rotor's wavefunctions, kinetic energies, and field-free evolution of orientation and alignment. We verify the analytic results by solving the corresponding time-dependent Schrödinger equation numerically and extend the temporal range of the interactions considered all the way to the adiabatic limit,  $\Delta\tau > \tau_r$ , where general analytic solutions beyond the field-free case are no longer available. The effects of the orienting and aligning interactions as well as of their combination<sup>1</sup> on the post-pulse populations of the rotational states are visualized as functions of the orienting and aligning kick strengths in terms of population quilts. Quantum carpets that encapsulate the evolution of the rotational wavepackets provide the space-time portraits of the resulting dynamics. The population quilts and quantum carpets reveal that purely orienting, purely aligning, or even-break combined interactions each exhibit sui generis dynamics. In the intermediate temporal regime, we find that the wavepackets as functions of the orienting and aligning kick strengths show resonances that correspond to diminished kinetic energies at particular values of the pulse duration.<sup>2</sup>

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## Microsolvation and rotation of 'large' molecular impurities inside helium nanodroplets upon electronic excitation

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High-resolution electronic spectroscopy (Stark or field-free) of large organic molecules and molecular complexes doped into superfluid helium nanodroplets poses many intriguing questions at the interface of physics and chemistry – especially with regard to solvation and rotation. In our recent work, we have noted the apparent absence of the predicted free rotation of phthalocyanine and porphine in He droplets<sup>1,2</sup> upon electronic excitation. Extending the study to clusters formed and probed in helium droplets, we also present our surprising finding of a lack of additional helium-stabilized phthalocyanine-water cluster configurations as compared to the gas-phase.<sup>3</sup> The absence of a rotational sub-structure in the electronic spectra of dopants larger than glyoxal begs for an alternative multi-pronged approach to clarifying the exact nature of the dopant-bath interaction before and after electronic excitation. To this end, we study the glyoxal-helium ab initio PESs both in the ground and the lowest excited state of the glyoxal molecule. Finally, we make use of the Angulon Theory<sup>4</sup> in the strong coupling regime to assess the dopant-bath PES anisotropies in the two electronic states that are required in order to drastically squeeze the rotational sub-structure.

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## Molecules as a dark matter detector

Jesús Pérez-Rios, Hariskrishnan Ramani<sup>a</sup>, Oren Slone<sup>b</sup> and Rouven Essig<sup>c</sup>

The analysis of the temperature of the cosmic microwave background shows that only 4.6% of the energy budget of the universe is baryonic matter. However, 23% of this energy content is in the form of dark matter (DM), as proposed by Zwicky in the early 30's<sup>1</sup>. The existence of dark matter has been corroborated from astrophysical and cosmological observations. These observations reveal that dark matter interacts through the gravitational force with regular matter, and that dark matter is self-frictionless. However, no information about the particle nature of the dark matter is obtained from these observations. Therefore, it is mandatory to propose and to perform experiments probing different mass-range for particle DM, as well as to different DM-standard model particle interactions.

We present a novel approach to dark matter detection based on molecular excitations. Our idea relies on using molecules in a well-controlled environment as a dark matter detector. In particular, we envision to have a gas of CO molecules at 50 K. Under these conditions, all the molecules are in the ground vibrational state. Thus, if a DM particle collides with a molecule, the momentum transfer to the nuclei will induce a vibrational excitation of the molecule. Afterward, the molecule will decay by emitting a certain number of photons, which is our detection signal<sup>2</sup>. The typical energy scaling for vibrational excitation in CO is around 0.25 eV, therefore with this setup, we will be sensitive to light dark matter, i.e DM particles lighter than the nucleon. This mass range is unachievable with accelerator experiments and hard to reach with noble gas-liquid detectors such as the xenon experiment<sup>3</sup>.

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## Accurate potential energy curves of AIF molecules: from quantum chemistry calculations to experimentally derived potentials

Jesús Pérez-Ríos, Gerrit Groenenboom<sup>a</sup>, and Gerard Meijer

The realization of ultracold molecules offers unique applications and novel scenarios to different disciplines of physics and chemistry<sup>1</sup>. For instance, ultracold polar molecules are excellent quantum simulators of different Hamiltonians, mimicking realistic solid-state physics systems. From the chemistry side, having molecules in well-defined quantum states (even translational degrees of freedom) opens up the possibility of tailoring chemical reactions. All these applications rely on having a precise knowledge of the electronic structure of the molecules and more precisely, about the Franck-Condon factors relevant for the cooling scheme.

We present the most precise electronic potentials up to date, and hence reliable Franck-Condon factors for AIF. This is the molecule of interest for the group of Dr. Truppe at the Fritz Haber Institute (see poster by S. Truppe for details). The relevant potential energy curves for the cooling scheme are obtained by inverting the accurate available and measured (in-house) spectroscopic information<sup>3</sup>. In this way, we can get very precise Franck-Condon factors that compare well with the experimental observations. The same potential energy curves as well as the transition dipole moments are calculated by ab initio quantum chemistry methods<sup>2</sup>. In that way, we can benchmark the theoretical calculations and reveal the uncertainty of these highly accurate quantum chemistry methods.

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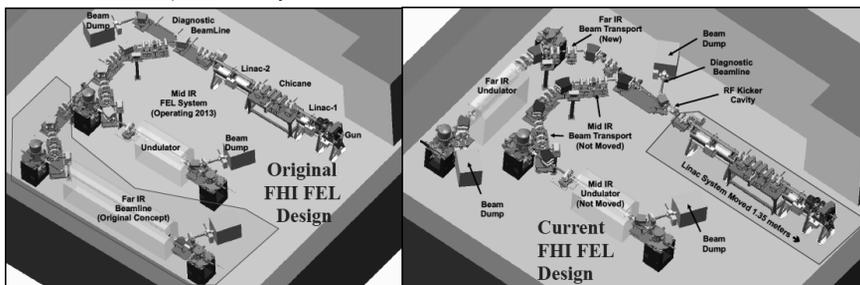
## The FHI FEL Facility: Status and 2-Color Upgrade Design

W. Schöllkopf, S. Gewinner, M. De Pas, H. Junkes, G. von Helden, and G. Meijer, W.B. Colson\*, D. Dowell\*, S.C. Gottschalk\*, J.W. Rathke\*, T.J. Schultheiss\*, A.M.M. Todd\*, and L.M. Young\*

Since coming on-line in November 2013, the FHI Free-Electron Laser (FEL) has provided intense mid-infrared (MIR) radiation, tunable from  $<3 \mu\text{m}$  to  $>50 \mu\text{m}$ , to FHI user groups. It has enabled experiments in diverse fields ranging from bio-molecular spectroscopy to studies of clusters and nanoparticles, nonlinear solid-state spectroscopy, and surface science, resulting in some 55 peer-reviewed publications so far. A significant upgrade to the original FHI FEL was proposed at the last Fachbeirat Meeting and was funded by the Max Planck Society in 2018.

The FHI FEL Upgrade essentially includes the addition of a second short Rayleigh range undulator FEL beamline (see Figure). The second FEL branch will permit lasing in the far-infrared (FIR) regime from  $<5 \mu\text{m}$  to  $>160 \mu\text{m}$ . In addition, a 500 MHz kicker cavity will be inserted downstream of the electron accelerator. It will permit simultaneous 2-color operation of both FEL beamlines by deflecting alternate 1 GHz electron bunches into each of the two undulator branches. In order to do this, the electron gun, sub-harmonic buncher, linacs and chicane will be moved back 1.35 m to allow the insertion of the kicker cavity, as indicated in Fig. 1. The kicker cavity, powered by a 65 kW solid-state amplifier, will operate in a dipole mode using the strong electric field between two vanes to deflect the electron bunches alternately left and right. It can deflect 50 MeV electrons by an angle of  $\pm 2^\circ$ . It is surrounded by two small  $2^\circ$ -deflection dipole magnets. For 2-color operation the beam is alternatively bent  $-1^\circ, -2^\circ, -1^\circ \Rightarrow -4^\circ$  by the dipole-kicker-dipole combination into the FIR line and  $-1^\circ, +2^\circ, -1^\circ \Rightarrow 0^\circ$  for the MIR beamline. Conventional single-color operation of either the MIR or the FIR FEL will still be possible when the 500 MHz field is off: with both dipoles off, every electron bunch is sent to the MIR branch; with both dipoles on deflecting  $-2^\circ, -2^\circ \Rightarrow -4^\circ$  every electron bunch will be sent to the FIR branch.

We describe the upgraded FHI FEL physics and engineering design and present the plans for 2-colour FEL operations by the end of 2020.



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## Matter-Wave Diffraction from a Periodic Array of Half Planes

Ju Hyeon Lee,\* Lee Yeong Kim,\* Bum Suk Zhao,\* and Wieland Schöllkopf

We have observed reflection and diffraction of beams of He and D<sub>2</sub> from square-wave gratings of a 400- $\mu\text{m}$  period and strip widths ranging from 10 to 200  $\mu\text{m}$  at grazing-incidence conditions. In each case we observe fully resolved matter-wave diffraction patterns including specular reflection and diffracted beams up to the second diffraction order. With decreasing strip width, the observed diffraction efficiencies exhibit a transformation from the regime of quantum reflection<sup>1</sup> from the grating strips<sup>2</sup> to the regime of edge diffraction from a half-plane array.<sup>3</sup> The latter is described by a single-parameter model<sup>4</sup> developed previously to describe phenomena as diverse as quantum billiards, scattering of radio waves in urban areas, and reflection of matter waves from microstructures. Our data provide experimental confirmation of the widespread model. Moreover, our results demonstrate that neither classical reflection nor quantum reflection are essential for achieving coherent reflection of matter waves from a structured solid, but it can result exclusively from half-plane edge diffraction.

In our experiment the quantum reflection probabilities are larger than those of half-plane array diffraction. This is due to the weak interaction of He and D<sub>2</sub> with the surface in combination with a rather sparse grating with a relatively large period of  $d = 0.4$  mm (see Figure). The latter can readily be reduced making it possible to increase the edge diffraction induced reflectivity significantly. Furthermore, half-plane edge diffraction occurs independent of the interaction potential between the atom or molecule and the half-plane surface. Therefore, it can be applied to more strongly interacting species and to weakly-bound molecules, such as the He dimer and trimer, for which classical and quantum reflection is inhibited or inefficient, respectively.

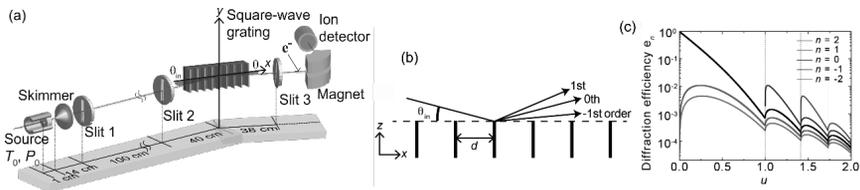


Figure: (a) Schematic of the experimental setup of grazing-incidence matter-wave diffraction. The reflection gratings are 50-mm-long micro-structured arrays of 4-mm-long parallel strips made out of 1- $\mu\text{m}$ -thick photoresist patterned on a commercial gold mirror. Four gratings of identical period  $d = 400$   $\mu\text{m}$  and different strip widths of 10, 30, 100, or 200  $\mu\text{m}$  have been used. (b) Schematic of the idealized situation of diffraction from an array of half planes (corresponding to vanishing strip width). (c) Diffraction efficiencies calculated by the single-parameter theory of half-plane array diffraction (Bogomolny-Schmit model), which we confirmed in the experiment.

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## Spectroscopic characterization of aluminium monofluoride with relevance to laser cooling and trapping

Stefan Truppe, Silvio Marx, Sebastian Kray, Maximilian Doppelbauer, Simon Hofsässs, H. Christian Schewe, Jesús Pérez-Ríos, Nicole Walter, Boris G. Sartakov, and Gerard Meijer

The aluminium monofluoride molecule (AlF) is an excellent candidate for laser cooling and magneto-optical trapping (MOT). All Q-lines of the  $A^1\Pi, v=0 \leftarrow X^1\Sigma^+, v'=0$  band near 227.5 nm are rotationally closed and can be used for laser cooling. Each molecule can scatter on average  $10^4$  photons by using one repump laser to address the  $A^1\Pi, v=0 \leftarrow X^1\Sigma^+, v'=1$  band. AlF is a closed shell molecule with a binding energy of almost 7 eV and the  $A^1\Pi, v=0$  state has a lifetime of 1.9 ns. This permits efficient production and slowing of the molecules and results in a large capture velocity of the MOT ( $> 40$  m/s), an excellent basis to trap AlF molecules with high density.

We present the precise spectroscopic characterization of all the states involved in the laser cooling scheme. We determine the rotational and hyperfine energy levels in  $X^1\Sigma^+, v=0$  and all three  $\Omega$ -manifolds in  $a^3\Pi, v=0$  with kHz and in  $A^1\Pi, v=0$  with MHz accuracy and infer precise spectroscopic constants for states. We determine the transition strengths between these states, measure their magnetic  $g$ -factors, their electric dipole moments and the lifetime of the  $A^1\Pi, v=0$  state. We also determine the vibrational branching ratio to  $X^1\Sigma^+, v'=1$  and compare the results to precise calculations using accurate molecular potentials (see poster by J. Pérez-Ríos).

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## A cryogenic buffer gas molecular beam of AlF molecules and optical cycling on a $A^1\Pi \leftarrow X^1\Sigma^+$ transition

Stefan Truppe, Simon Hofsäss, Maximilian Doppelbauer, Sebastian Kray,  
Jesús Pérez-Ríos, and Gerard Meijer

A bright and slow molecular beam is essential for further laser cooling and trapping experiments. Therefore, we develop a cryogenic buffer gas molecular beam source; a continuous flow of cryogenic helium gas is mixed with  $SF_6$  (300 K) and flows through a copper cell that is cooled to 3 K. We use a pulsed Nd:YAG laser to ablate an aluminium rod that is fixed inside the copper cell. The hot aluminium reacts with the  $SF_6$  and forms AlF. The hot AlF molecules are subsequently cooled via collisions with the cryogenic helium and extracted into a molecular beam through a small aperture in the cell. We routinely produce beams that contain  $5 \times 10^{12}$  molecules/sr/pulse, have a mean forward velocity of 160 m/s, and a rotational temperature of 4 K. This AlF beam is about 100 times brighter than similar cryogenic beams of diatomic molecules. We investigate the influence of the geometry of the copper cell on the beam properties and find the optimum conditions for the helium and  $SF_6$  flow rate. We compare the molecular flux of the helium buffer gas beam to the flux of a pulsed supersonic beam of AlF seeded in Neon. The neon beam has a speed of 800 m/s, and the total number of molecules delivered to a detector 35 cm downstream is over 100 times lower. The buffer gas molecular beam of AlF is so intense that it is possible to measure its density via absorption of a resonant laser beam, even 35 cm from the source. This molecular beam is ideal for trapping the molecules in a magneto-optical trap with a high density.

We use the buffer gas molecular beam to demonstrate optical cycling on the Q(1) line of the  $A^1\Pi, v=0 \leftarrow X^1\Sigma^+, v''=0$  band and compare it to a theoretical model that includes all 72 Zeeman sub-levels that are involved in the optical cycling scheme.

## Aqueous-phase TiO<sub>2</sub> Nanoparticles: Nature of Water Adsorption and Structure of the Electric Double Layer

Hebatallah Ali, Robert Seidel,<sup>a</sup> Jie Xiao,<sup>a</sup> Bernd Winter, and Gerard Meijer

Titanium dioxide (TiO<sub>2</sub>) is a promising material to catalyse water dissociation for the production of H<sub>2</sub> fuel as a clean and renewable energy source.<sup>1,2</sup> One of the processes limiting the conversion efficiency in water-splitting reactions<sup>1,3</sup> is the fast back-reaction of proton (H<sup>+</sup>) and hydroxide (OH<sup>-</sup>) recombination into water molecules. Our studies aim (i) at exploring the mechanism of water dissociation at the anatase – aqueous solution interface, and (ii) at characterizing the electric double layer (EDL) that forms at this interface, and which has a strong influence on ion distributions and mobilities. This information can be obtained from X-ray spectroscopy. One particular requirement is that the solid surface is exposed to bulk aqueous solution, at adjustable pH, which can be realized in nanoparticle (NP) aqueous solutions, provided that the NPs can be stabilized.

We have applied *in-situ* liquid microjet X-ray spectroscopies to characterize the EDL around 20-nm TiO<sub>2</sub> NPs dispersed and stabilized in NH<sub>4</sub>OH aqueous solution. By combining non-resonant (PES) and resonant (RPES) photoelectron spectroscopy, partial electron and partial fluorescence yield X-ray absorption spectroscopy (PEY-XAS, PFY-XAS), we find that at low concentration of NH<sub>4</sub><sup>+</sup> stabilizer ions – but still assuring that NPs do not aggregate – there remains sufficient NP surface area to interact with water. PES and PEY-XAS measurements at the N 1s and O 1s core levels, along with the measured (slightly basic) pH, identify the Stern layer to consist of < 1 monolayer NH<sub>4</sub><sup>+</sup> stabilizer ions. Moreover, the PFY measurements at the O K-edge reveal an unexpected large amount of OH<sup>-</sup> in the NP aqueous solution (despite near-neutral pH) that can only be explained by water dissociation. These OH<sup>-</sup> species are proposed to be located within a > 0.5-nm-thick diffuse layer, charge-balanced by the positive Stern layer. The PFY measurements furthermore suggest a prevailing 3-coordinated OH<sup>-</sup> hydration structure, OH<sup>-</sup>(H<sub>2</sub>O)<sub>3</sub>, within the diffuse layer which is not the case for OH<sup>-</sup> hydration in bulk-solution where the 4-coordinated structure, OH<sup>-</sup>(H<sub>2</sub>O)<sub>4</sub>, is more probable. Some of the protons from water dissociation are argued to be responsible for neutralization reactions in bulk solutions.

Analogous measurements performed in acidic solutions show very different behaviour, with water being molecularly adsorbed.<sup>4</sup>

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## A Unique Setup for Detection of Photoelectron Circular Dichroism from Aqueous-Phase Chiral Molecules

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Water molecules play an invaluable role in governing the structure, stability, dynamics, and function of biomolecules. Indeed, biological activity of proteins and nucleic acids requires the presence of water. A fascinating aspect of such activity and (bio)chemistry is chirality. We have developed a unique liquid-jet photoelectron spectroscopy (LJ-PES)<sup>1</sup> instrument, *Electronic structure from Aqueous Solutions and Interfaces (EASI)*, that adds unprecedented sensitivity to the geometric property of chirality to more generally afforded liquid-phase electronic structure information.

The effect exploited here is the chirality-selective discrimination mechanism, 'PhotoElectron Circular Dichroism' (PECD)<sup>2-4</sup> which so far has been demonstrated for gas-phase chiral molecules, and with *EASI* it is now possible to perform PECD measurements from aqueous solution. PECD is a purely electric dipole effect, and has a typical strength of a few percent, *i.e.*, at least three orders of magnitude greater than typical chiroptical effects.<sup>2-4</sup> It can be used to specifically probe chiral properties of each orbital contributing to the electronic structure without prerequisites, while at the same time retaining the information on electronic charge distributions and oxidation states known from PES.

PECD is observed upon photoionization of a chiral molecule by left and right circularly polarized light (l- or r-CPL), and manifests in a forward – backward asymmetry in the flux of the ejected electrons with respect to the direction of light propagation (*k* vector). The difference seen between l- and r-CPL, for a given enantiomer and detection angle, defines the actual PECD.<sup>2-4</sup> *EASI*, consisting of a state-of-the-art, hemispherical electron analyser is capable to detect this specific asymmetry in the photoelectron angular distributions, and thus enables to determine the so-called chiral parameter  $b_1$ .<sup>2-4</sup>

First PECD results from liquid fenchone and from aqueous-phase amino acid alanine measured near the carbon 1s core-level ionization threshold will be presented. We discuss the overall scientific goal of this project and detail the necessary technical developments required for an efficient detection of LJ-PECD. One aspect of the latter is the need for novel liquid jet designs.

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## Cryogenic Liquid Jet Photoemission from Alkali-Metal – Liquid Ammonia: From Electrolyte to Metallic Solutions

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Liquid ammonia is often used as a medium for organic chemistry due its ability to support long-lived highly reducing solvated electrons. Solvated electrons can be generated simply by adding an alkali metal to neat liquid ammonia, and unlike in water they have lifetimes on the order of months for solutions which are free from oxygen and moisture. One of the most fascinating aspects of these solutions is a gradual transition of a blue electrolyte into a bronze/gold coloured metallic solution upon increasing the alkali metal concentration. The blue solution is characteristic of solvated electrons, and the metallic state corresponds the solvated electrons being genuinely delocalized.<sup>1,2</sup> Despite tremendous experimental and theoretical efforts, a detailed microscopic understanding of metal–ammonia solutions across the entire concentration (and composition) range, from electrolyte to metallic, is still lacking.<sup>1</sup>

We have designed an experiment that allows for the first time to measure the electronic structure of neat liquid ammonia and of metal – ammonia solutions. Specifically, we have developed a refrigerated vacuum liquid microjet (at approximately -70°C) which can be used for photoelectron (PE) spectroscopy measurements. PE spectra, obtained with soft X-rays, reveal the valence and nitrogen core-level binding energies of neat liquid ammonia.<sup>3</sup> The vertical ionization energy of the highest occupied orbital (HOMO,  $3a_1$ ) is determined to be 9.1 eV. When adding low-concentration alkali metal, up to 0.2 M, a narrow peak at approximately 2 eV binding energy identifies the vertical photodetachment energy (VDE) of localized electrons (and dielectrons<sup>1</sup>) solvated in liquid ammonia. No noticeable dependence of the energy of the ammoniated electron on the chemical nature of the alkali metal, Li *versus* Na, *versus* K, is observed. Note that VDE is referenced to the vacuum energy; also, all binding energies from liquid water and aqueous solutions reported to date are with respect to the vacuum level. For alkali metal concentrations above 1 M, when the solution is golden, the PE spectra align at the Fermi edge; a structured band is observed to extend up to 4.5 eV below the Fermi energy. In addition, the  $3a_1$  (HOMO) peak is found to be broadened and shifted as compared to neat liquid ammonia which can be assigned to  $(\text{NH}_3)_n$  – metal complexes.

Our study shows that this electrolyte-to-metal transition is a gradual process rather than an abrupt first order transition, in accord with previous suggestions.<sup>1</sup> The interpretation of PE spectra of ammoniated electrons in terms of a complex structural, dynamical, and molecular orbital picture is enabled by electronic structure calculations.

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## Department of Physical Chemistry

### Poster List

- PC 1 **Multidimensional Photoemission Spectroscopy: The Concept**  
Shuo Dong, Patrick Xian, Maciej Dendzik, Samuel Beaulieu, Tommaso Pincelli, Julian Maklar, William Windsor, Alexander Neef, Marios Zacharias, Christian Carbogno, Vincent Stimper, Bernhard Schöllkopf, Stefan Bauer, Martin Wolf, Laurenz Rettig, and Ralph Ernstorfer
- PC 2 **Multidimensional Photoemission Spectroscopy: Application to Complex Materials**  
Julian Maklar, Sam Beaulieu, Tommaso Pincelli, Maciej Dendzik, Shuo Dong, William Windsor, Martin Wolf, Ralph Ernstorfer, and Laurenz Rettig
- PC 3 **Femtosecond Electron and Lattice Dynamics in Black Phosphorus**  
Hélène Seiler, Maciej Dendzik, Daniela Zahn, Shuo Dong, Patrick Hildebrandt, Samuel Beaulieu, Tommaso Pincelli, Julian Maklar, Thomas Vasileiadis, Patrick Xian, William Windsor, Christian Carbogno, Marios Zacharias, Claudia Draxl, Fabio Caruso, Martin Wolf, Laurenz Rettig, and Ralph Ernstorfer
- PC 4 **Structural and Electron Dynamics in Nanoscale Materials**  
Thomas Vasileiadis, Daniela Zahn, Faruk Krecinic, Hélène Seiler, Shuo Dong, Ivana Lapšanská, William Windsor, Yingpeng Qi, Richard Palmer, Robert Schneider, Rudolf Bratschitsch, and Ralph Ernstorfer
- PC 5 **Ultrafast Dynamics of Antiferromagnetism Studied Using Resonant X-ray Diffraction**  
Yoav William Windsor, Sang Eun Lee, Daniela Zahn, and Laurenz Rettig
- PC 6 **Probing Ultrafast Dynamics by Attosecond Diffraction Spectroscopy and Development of Nonlinear X-ray Spectroscopy**  
Lars Hoffmann and Michael Zürc
- PC 7 **Complex Non-Equilibrium Dynamics in Transition Metal Oxides and Inorganic/Organic Hybrid Systems**  
Lukas Gierster, Sessa Vempati, Angelika Demling, Sarah B. King, and Julia Stähler
- PC 8 **Dynamic Screening of Quasiparticles in WS<sub>2</sub> Monolayers en route to Ultrafast Phenomena at TMDC/Organic Hybrid Interfaces**  
Stefano Calati, Boubacar Tanda Bonkano, Samuel Palato, and Julia Stähler
- PC 9 **Ultrafast Spin- to Charge-current Conversion in Magnetic Metal Bilayers: Driving Force and Role of the Interface**  
Oliver Gückstock, Reza Rouzegar, Lukáš Nádvořník, Martin Wolf, and Tobias Kampfrath
- PC 10 **Probing the Response of Ferromagnetic Order to Terahertz Excitation**  
Julius Heitz, Alexander Chekhov, Yannic Behovits, Lukáš Nádvořník, Martin Wolf, and Tobias Kampfrath
- PC 11 **Phonon Polaritons in Polar Dielectric Heterostructures**  
Nikolai C. Passler, Christopher J. Winta, Ilya Razdolski, Martin Wolf, and Alexander Paarmann

- PC 12 **Sum-Frequency Generation Spectroscopy and Microscopy using the FHI Infrared Free-Electron Laser**  
Riko Kiessling, Yujin Tong, Christopher Winta, Sandy Gewinner, Wieland Schöllkopf, Martin Wolf, and Alexander Paarmann
- PC 13 **Towards Nano-Scale Optical Spectroscopy Using Plasmonic Scanning Tunneling Microscope Junctions**  
Shuyi Liu, Hannes Böckmann, Melanie Müller, Adnan Hammud, Martin Wolf, and Takashi Kumagai
- PC 14 **Towards Atomic-Scale Vibrational Spectroscopy Using Tip-Enhanced Raman Spectroscopy**  
Shuyi Liu, Melanie Müller, Adnan Hammud, Martin Wolf, and Takashi Kumagai
- PC 15 **Implementation and Characterization of a Photoexcited THz-gated Scanning Tunneling Microscope**  
Melanie Müller, Natalia Martín-Sabanés, Sarojini Mahajan, and Martin Wolf
- PC 16 **High Resolution Scanning Gate Microscopy**  
Alexander Kölker, Takashi Kumagai, Martin Wolf, and Matthias Koch
- PC 17 **Quantifying the Interfacial Contribution in Vibrationally Resonant Sum Frequency Generation Spectroscopy**  
Martin Thämer, Tobias Garling, Martin Wolf, and R. Kramer Campen
- PC 18 **Characterizing Potential Dependent Structure at the Electrode/Electrolyte Interface**  
Gregor Zwaschka, Xin Gong, Martin Wolf, R. Kramer Campen, and Yujin Tong
- PC 19 **Exploring the Mechanism of the Hydrogen Evolution Reaction on Pt and Au with Femtosecond Potential Perturbation Experiments**  
Gregor Zwaschka, François Lapointe, Martin Wolf, Yujin Tong, and R. Kramer Campen
- PC 20 **Characterizing the Oxide/Water Interface from the Oxide's Perspective**  
Yujin Tong, Yanhua Yue, Giacomo Melani, Peter Saalfrank, and R. Kramer Campen
- PC 21 **Nonlinear THz-Raman Spectroscopy Reveals Structural Effects and Energy Transfer Pathways of Ionic Solutions**  
Vasileios Balos, Martin Wolf, and Mohsen Sajadi
- PC 22 **Distinct Terahertz Magnetic- and Electric-Field Response of Molecules**  
Vasileios Balos, Genaro Bierhance, Martin Wolf, and Mohsen Sajadi

## Multidimensional Photoemission Spectroscopy: The Concept

Shuo Dong, Patrick Xian, Maciej Dendzik, Samuel Beaulieu, Tommaso Pincelli, Julian Maklar, William Windsor, Alexander Neef, Marios Zacharias, Christian Carbogno, Vincent Stimper, Bernhard Schöllkopf, Stefan Bauer, Martin Wolf, Laurenz Rettig, and Ralph Ernstorfer

The electronic structure of solids represents a multidimensional problem, where basic (opto-)electronic properties, complex quantum states or transient non-equilibrium properties are reflected in specific features of the multidimensional quasi-particle self-energy. We are pioneering an approach which embraces multidimensionality at each level of the experimental process making use of state-of-the-art instrumentation and novel data processing pipelines.

We have developed a time- and angle-resolved photoemission spectroscopy (trARPES) facility featuring a high-repetition rate XUV laser source and a time-of-flight momentum microscope spectrometer. Every detected photoelectron is considered a multidimensional object defined by intrinsic (energy, parallel momentum) and extrinsic parameters (pump-probe time, laser photon energies, laser polarisations, sample temperature, etc.). Depending on the material properties of interest, the single-event data is projected to volumetric data in a subspace of lower dimensions. We discuss the approach based on a few examples of use for one material, the layered semiconductor  $\text{WSe}_2$ . The ultrafast dynamics of excitons is investigated in the default trARPES dimensions of energy, parallel momentum ( $k_x, k_y$ ) and time. In contrast, we obtain the linear dichroism in the photoelectron angular distribution (LDAD) as well the anisotropy of the constant-energy maps in the vicinity of  $K$  and  $K'$  valleys from angular scans of the sample-laser geometry. As previously demonstrated for graphene, these quantities are related to the wavefunction symmetry, its chirality as well as its Berry phase. The use of different probe photon energies, as third example, provides additional access to core levels. Relating the core level line shapes to valence band dynamics reveals the sensitivity of core levels to semiconductor Mott transitions.

The high data rate of the experiment results in a data challenge. Therefore, we develop new approaches like unsupervised learning algorithms for extracting experimental band structure from multidimensional data sets. This leads to effective reduction in data volume and makes photoemission data accessible to a broader community, for instance for benchmarking electronic structure calculations. We share this development with the community by developing an open-source multidimensional photoemission data analytics toolbox (<https://github.com/mpes-kit>).

## Multidimensional Photoemission Spectroscopy: Application to Complex Materials

Julian Maklar, Sam Beaulieu, Tommaso Pincelli, Maciej Dendzik, Shuo Dong, William Windsor, Martin Wolf, Ralph Ernstorfer, and Laurenz Rettig

Complex materials are characterized by their fascinating properties originating from the strong interactions among their microscopic subsystems, typically characterized by broken symmetries or non-trivial topologies. Often, such states have characteristic fingerprints in the electronic structure of the materials, and studying its dynamics during photoinduced phase transitions (PIPTs) can provide very important information on the nature of the ordered state and the underlying reaction mechanisms and pathways. By employing our recently developed methodology of ultrafast multidimensional photoelectron spectroscopy (MPES), we can study changes in the topology of the electronic structure or follow the transition from an insulating ground state into a metallic excited state. By comparison of the experimental data to electronic band structure calculations, their validity for predicting excited state band dispersion and potential deviations, e.g. due to many-body interactions, can be assessed.

A fascinating topologically nontrivial system is the type-II Weyl semimetal  $T_d$ - $\text{MoTe}_2$ , which shows a strong sensitivity of the Fermi surface topology to electronic correlations [1]. Time-resolved Fermi-surface mapping enabled by MPES allows us to observe a topological phase transition identified through the emergence of additional pockets on the Fermi surface after strong infrared photoexcitation. This ultrafast Lifshitz transition demonstrates the strong enhancement of electronic screening due to photoexcited carriers, which suppresses electronic correlations.

Charge density waves (CDWs) are a coupled instability of lattice and electronic system where large energy gaps open up in the electronic energy dispersion. Using the momentum microscope, we track the transient electronic fingerprints of the CDW transition in the model system  $\text{TbTe}_3$  [2] in a wide energy and momentum range throughout the BZ, including the unoccupied band structure. A detailed investigation of the fluence dependent dynamics of the spectral weight in the gapped regions yields access to the dynamics of the electronic order parameter of the CDW, which we describe within a time-dependent Ginzburg-Landau model. We also exploit the spatial resolution of the instrument by investigating the dynamics of a CDW step-edge defect, where CDW order is locally suppressed.

Finally, in the proposed Mott-insulating 2D ad-atom system  $\text{Sn-Si}(111)$  we study the dynamics of the upper Hubbard band after photoexcitation. The ultrafast relaxation of carriers injected into the upper Hubbard band, occurring within the time resolution of the experiment, as well as its fluence dependence shows a close resemblance to other Mott insulators [3], providing strong evidence for the Mott-insulating nature of the ground state in the system.

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## Femtosecond Electron and Lattice Dynamics in Black Phosphorus

Hélène Seiler, Maciej Dendzik, Daniela Zahn, Shuo Dong, Patrick Hildebrandt, Samuel Beaulieu, Tommaso Pincelli, Julian Maklar, Thomas Vasileiadis, Patrick Xian, William Windsor, Christian Carbogno, Marios Zacharias, Claudia Draxl<sup>a</sup>, Fabio Caruso<sup>a</sup>, Martin Wolf, Laurenz Rettig, and Ralph Ernstorfer

Black phosphorus (BP) has recently emerged as a promising material for novel optoelectronic devices due to its highly anisotropic thermal, electronic and optical properties [1]. Macroscopic quantities such as thermal and electrical conductivities, which dictate device performance, relate to microscopic interactions between a solid's constituents. Therefore, a fundamental understanding of both the electronic and atomic structures as well as their interactions upon photo-excitation is essential. We present a femtosecond time-resolved study of black phosphorus which combines the complementary perspectives of the electronic and atomic sub-systems. To probe the transient electronic structure over the entire Brillouin zone (BZ), we employ XUV time- and angle-resolved photoemission spectroscopy (trARPES) at 500 kHz repetition rate featuring a momentum microscope [2]. The lattice dynamics is probed with momentum resolution by femtosecond electron diffuse scattering (FEDS). The combination of the two complementary techniques yields a microscopic picture of the evolution of the non-equilibrium states in black phosphorus, governed by electron-electron, electron-phonon and phonon-phonon couplings.

The electronic band structure measured with trARPES is found to be in good agreement with density functional theory calculations. When the black phosphorus flake is impulsively driven out-of-equilibrium with 800 nm laser pulses, a hot-carrier distribution around the BZ center is observed, which scatters to two other valleys in the conduction band within ca. 15 fs. We further observe a strong linear dichroism in the optical absorption of BP. Interestingly, the inter-valley scattering dynamics are also found to depend on the pump polarization direction.

On the lattice side, the measurements demonstrate an anisotropic femtosecond response along the zigzag and armchair directions. Momentum-resolved analysis of the diffraction signals enables to map phonon populations with femtosecond resolution. At early times, phonons build up along the zigzag direction, consistent with the trARPES results. At later times, phonons also build up along the armchair direction. The data shows that the lattice's response following photo-excitation is profoundly non-thermal, offering a direct proof that hot electrons couple preferably to specific phonons in black phosphorus and highlighting the important role of phonon-phonon interactions in the material. Electron-phonon couplings and phonon-phonon couplings are calculated over the whole BZ and used to build a non-thermal lattice model to rationalize the experimental results.

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## Structural and Electron Dynamics in Nanoscale Materials

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We employ femtosecond electron pulses and single-electron wave packets as probes for ultrafast structural and electron dynamics in nanoscale materials. Time-resolved electron diffraction is used to measure the ultrafast energy flow between electronic excitation and atomic vibrations within a material as well as across heterostructure interfaces. We employ this approach to size-selected Au nanocrystals, which were soft-landed on different thin-film substrates like graphene [1,2]. A combined analysis of the time-dependent diffraction peak intensities, positions and shapes reveals rich information on the nanocrystal's non-equilibrium properties, including their structural stability. In addition, we exploit the sensitivity of diffraction to the crystal orientation in order to study ultrafast rotational motions, so-called librations, of nanocrystals on flexural substrates. Applied to 2D materials, we study the structural response of monolayer WSe<sub>2</sub> to optical excitation of A, B and C excitons. In all cases, we observe a rapid transfer of electronic excitation to vibrations, which supports the notion of rapid relaxation of the optically prepared bright excitons to finite-momentum dark excitons [3].

Complementary to time-resolved momentum-space techniques, we aim to observe ultrafast charge dynamics in real space with femtosecond electron point-projection microscopy. This pump-probe technique uses low-energy (<200 eV) electrons as an imaging probe and is highly sensitive to the electric field distributions in the vicinity of nanostructures. Time-resolved experiments can therefore reveal ultrafast transient changes of electric field distributions near nanoscale objects, which allow for the reconstruction of the underlying motion of charges. The electric field sensitivity and the spatial resolution of this technique can be further enhanced, down to the level of a single elementary charge, by taking advantage of the phase information of the imaging electron wave packet [4]. Improvements in the mechanical stability of the microscope, and optimization of the photoemission process from nanotips in terms of source coherence, have recently allowed us to record first images with clear signatures of electron interference fringes in the laser-triggered imaging mode. This is a prerequisite for, and a first possible indication of, the realization of femtosecond electron holography with a combined spatio-temporal resolution below 30 nm / 30 fs.

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## Ultrafast Dynamics of Antiferromagnetism Studied Using Resonant X-ray Diffraction

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Antiferromagnetic spintronics are a promising route towards more efficient and stable devices, because antiferromagnets (AF) are less susceptible to external fields and foster a broad range of magnetic interactions with the potential for higher speeds and energy efficient manipulation. The prospect of employing antiferromagnets in devices opens new functionality pathways through properties that aren't available with ferromagnets. These include driving transitions between different spin arrangements, manipulating the ordering wave vector and more. In particular, light-induced control of the AF state is a promising route that has been recently demonstrated, and the use of femtosecond light pulses opens the path towards ultrafast AF dynamics.

Here we present ultrafast pump-probe experiments conducted at the FemtospeX slicing facility at BESSY II, a unique source of ultrashort pulses (100fs) of soft x rays (i.e. photon energies of 500-1500 eV). We employ time-resolved resonant x-ray diffraction (RXD), which is a direct probe of the long-range antiferromagnetic ordering. Using its polarization dependence, it provides detailed information about the microscopic antiferromagnetic spin arrangement (e.g.  $\uparrow\downarrow\uparrow\downarrow$ , spin spirals, helices etc.). The resonant enhancement at the L2/3 or M4/5 edges provides direct sensitivity to 3d and 4f ("magnetic") states, alongside with element sensitivity.

We used this technique to study the ultrafast magnetization dynamics in a family of prototypical Lanthanide ("RE") intermetallics  $RERh_2Si_2$ . For the case of  $GdRh_2Si_2$ , which is characterized by its weak in-plane anisotropy [1], we observe a coherent and deterministic rotation of the entire AF spin arrangement about a crystal axis via optical excitation. This effect, which occurs on a time scale of a few 10s of ps, is due to a transient change of the magnetocrystalline anisotropy. Thereby, we demonstrate a route towards optical control of an AF spin structure.

Extending this study, we directly compare 4f magnetization dynamics of different Lanthanide ("RE") ions, by systematically studying the optically-induced demagnetization in various members of  $RERh_2Si_2$ . This series of nearly-identical materials is an ideal test bed, which can be considered as a system of AF-ordered RE ions embedded in a  $Rh_2Si_2$  cage. We observe a strong variation in demagnetization time scales, from 100s of fs to 100s of ps. A systematic comparison of the RE ions reveals that their angular momentum transfer rates scale with the de Gennes factor  $(g-1)^2J(J+1)$ , indicating that the magnitude of the indirect RKKY exchange interaction is dominant in determining the demagnetization rates.

Lastly, a major promise of antiferromagnetic dynamics is inter-sublattice exchange of angular momentum, which would allow overcoming speed limits associated with angular momentum dissipation to the lattice, as is inherent to ferromagnets. To address this, we study AF systems based on 3d magnetic ions, which are mediated by *direct* exchange. We observe much faster demagnetization dynamics than with 4f magnetic ions, approaching the experimental resolutions ( $\sim 110$ fs).

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## Probing Ultrafast Dynamics by Attosecond Diffraction Spectroscopy and Development of Nonlinear X-ray Spectroscopy

Lars Hoffmann and Michael Zürich

Attosecond diffraction spectroscopy is a multimodal approach to probe the long-range spin order and the electronic and lattice dynamics in magnetic thin films leveraging the material-selectivity of X-rays and extremely short pulse duration achievable with attosecond tabletop sources. In our experiments, we generate isolated attosecond pulses in a broad spectral range covering several absorption edges present in the multilayer thin films and interrogate the spin order by resonant magnetic diffraction, while the electronic-structural response is simultaneously measured with atomic specificity by transient absorption spectroscopy [1-3]. Using this scheme enables us following the ultrafast optical excitation of the carriers out of equilibrium and the subsequent demagnetization. In a first experiment, we probe the structural response and the magnetic order in Co-Pt magnetic multilayers following ultrafast photoexcitation observing that the onset of demagnetization is time-delayed by approximately 4 fs indicating the interfacial spin transport in the photoexcited system.

While linear X-ray spectroscopies are sensitive to the bulk response of a system, the electronic properties important to applications such as catalysis are most relevant at surface and interfacial layers [4]. In the past decades nonlinear spectroscopy has become an important technique for surface analysis in the optical and near UV energy regimes. This technique exploits the enhancement in the second-order nonlinear response to a coherent photon beam at a symmetry-broken interface in an otherwise centrosymmetric media. Developing this technique towards the X-ray regime offers the unique ability to study the electronic structure of interfacial regions with core-level spectroscopy, effectively allowing for X-ray absorption spectroscopy (XAS) of the first molecular layer of a bulk sample or of a buried interface using a photon-in/photon-out detection scheme. Such measurements coupled with first-principles electronic structure calculations for spectral interpretation effectively open the door to a new field of surface analysis. Following first demonstrations of X-ray second harmonic generation, we seek to develop this technique towards X-ray sum-frequency generation and performed first free-electron laser experiments studying the nonlinear X-ray response in solid and laser-molten graphite crystals.

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## Complex Non-Equilibrium Dynamics in Transition Metal Oxides and Inorganic/Organic Hybrid Systems

Lukas Gierster, Sesha Vempati<sup>a</sup>, Angelika Demling, Sarah B. King<sup>b</sup>, and Julia Stähler

ZnO and Fe<sub>2</sub>O<sub>3</sub> are oxide semiconductors with applications that range from (photo-) catalysis to light-harvesting devices. A fundamental understanding of their non-equilibrium properties remains challenging but desirable, not least for the exploitation of novel functionalities [1]. Here we use time- and angle-resolved photoelectron spectroscopy (trARPES) to follow the ultrafast evolution of the electronic properties of the ZnO(10-10) and Fe<sub>2</sub>O<sub>3</sub>(1-102) surface upon photoexcitation. Intriguingly, we find that ZnO undergoes a semiconductor-to-metal transition (SMT) when pumped resonantly at high excitation densities. The metallic phase is characterized by a dispersive band that is crossed by the equilibrium Fermi level  $E_F$  and arises within only 20 fs. Furthermore, the photoelectron spectra show a Fermi-Dirac distribution with a delay-dependent temperature, the signature of a hot electron gas. Notably, the metallic state also decays on an ultrafast timescale ( $\sim 200$  ps). In exact temporal coincidence with the build-up and decay of the transient metal, the valence band shifts down and up again, respectively. Our observations can be explained by charge accumulation at the ZnO surface, which induces surface band bending, in close analogy to the (equilibrium) SMT induced by surface doping of ZnO and other oxides [2]. Tuning the pump photon energy below the fundamental gap proves that the depopulation of in-gap states (likely related to surface defects) is responsible for the SMT *via* an inverse surface photovoltage (SPV) effect. Complementarily, we observe similar transient band bending effects for Fe<sub>2</sub>O<sub>3</sub>, albeit weaker, and will discuss these with regard to the SPV effect. Our results on ZnO uncover a pathway to create confined surface electrons on an ultrafast timescale.

Photoinduced surface charges are also relevant at low excitation densities: Hybrid solar cells require efficient charge separation (CS) at the inorganic-organic interface. The CS efficiency of ZnO-organic interfaces is notoriously low, likely a result from the formation of hybrid charge transfer excitons (HCTX), which favor recombination at the interface [3]. So far, a direct observation of the HCTX, its formation and decay on ultrafast timescales remains elusive. Here we disentangle the CS sequence at a model ZnO-organic chromophore interface using trARPES. The system forms a type II heterojunction [4]. Upon photoexcitation of the organic across its optical gap the electrons decay from the LUMO to the ZnO CB within  $\sim 350$  fs. However, within  $\sim 100$  ps, the electrons return to the interface and form a strongly bound state  $\sim 0.5$  eV below  $E_F$  and with a  $\mu$ s lifetime. We conclude that HCTX formation occurs and is mediated by the long-range Coulomb interaction after completing the first CS step.

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## Dynamic Screening of Quasiparticles in WS<sub>2</sub> Monolayers *en route* to Ultrafast Phenomena at TMDC/Organic Hybrid Interfaces

Stefano Calati, Boubacar Tanda Bonkano, Samuel Palato, and Julia Stähler

The low dimensional nature of transition metal dichalcogenides (TMDCs) and the resulting reduced screening significantly influence their non-equilibrium optical properties, as dynamic screening by photoexcited quasiparticles governs the transient response.

In this work, we investigate the role of different photoexcited quasiparticles on the dynamic response of WS<sub>2</sub> monolayers on SiO<sub>2</sub>: excitons generated by resonant pumping and quasi-free carriers photoexcited by above-resonance pumping. Drastic changes in the reflectivity contrast upon photoexcitation are observed in both cases and the contributions to the neutral exciton and the trion are isolated. The main observation is a pump photon energy-dependent blue/red shift of the neutral exciton for resonant/above-resonance pumping, respectively. Both, photoexcited excitons and quasi-free carriers screen the Coulomb interaction, leading to a weaker/stronger renormalization of the quasi-free-particle band gap compared to the dynamic screening-induced reduction of the exciton binding energy. The relative interplay between these two effects explains the observed blue/red shift crossover. Using a simple rate equation model, it is possible to, for instance, extract exciton formation and decay times from the multiexponential dynamics of the exciton and trion resonances.

The combination of TMDCs with organic dye molecules is promising for the next generation of optoelectronic and light harvesting devices, provided the chromophores exhibit appropriate energy levels and sufficient excited state lifetimes to allow for charge or energy transfer processes. As a first step towards the investigation of hybrid interfaces, we studied terrylene molecules in solution. Time-resolved photoluminescence and broadband transient absorption experiments showed textbook-like relaxation dynamics of the excited states and a 3.2 ns lifetime. The effect of molecule condensation will be studied on passive substrates before moving towards terrylene/WS<sub>2</sub> samples.

Beyond their sensitivity to dynamic screening, TMDC monolayers are also highly sensitive to *local* effects arising from edges, defects, strain, and variations of the dielectric constant in the template and/or adsorbate. To study femtosecond dynamics on optical energy scales *and* at the nanoscale, we are developing time-resolved interferometric optical nanoscopy (TRION). This will yield unprecedented observations of the spatial dependence of electron dynamics in TMDC monolayers and TMDC/organic heterojunctions with nm spatial resolution and fs temporal resolution. The poster will show first AFM and near-field images acquired using our femtosecond laser system.

## Ultrafast Spin- to Charge-current Conversion in Magnetic Metal Bilayers: Driving Force and Role of the Interface

Oliver Gückstock, Reza Rouzegar, Lukáš Nádvořník, Martin Wolf, and Tobias Kampfrath

Transport of spin angular momentum and the conversion of the resulting spin current to a charge current are fundamental operations required for future spin-electronic devices. To be competitive and compatible with other information carriers such as light, it is required to push the bandwidth of these operations to the terahertz (THz) frequency range.

Here, we use femtosecond laser pulses (duration of  $\sim 10$  fs) to excite prototypical F|N bilayers consisting of a ferromagnetic metal F and a nonmagnetic metal N [1,2]. Following absorption of the pulse, a spin current from the F to the N layer is launched. At the same time, the spin current is converted into a transversely (e.g. in-plane) oriented charge current that gives rise to the emission of an electromagnetic pulse with frequencies extending into the THz range. This effect is interesting for building efficient THz emitters with a bandwidth covering the difficult-to-reach range from 5 to 15 THz [1]. To better understand and eventually optimize this effect, we (i) investigate the impact of the F-N interface on the spin- and charge transport and (ii) made a successful attempt to identify the driving force of the laser-triggered spin current.

Regarding (i), we study ultrafast spin-to-charge-current conversion (S2C) in prototypical F|N bilayers consisting of a ferromagnetic layer F (Py, Co or Fe) and a nonmagnetic layer N with strong (Pt) or weak (Cu, Al) spin-orbit coupling. We find a surprisingly strong interface contribution to S2C which dominates the S2C process when N is a material with weak spin-orbit coupling. By reversing the growth order of F and N layers or by dusting the interface region demonstrates that the F-N interface has a drastic impact on the polarity of the emitted THz pulse. We consistently ascribe the interfacial S2C to skew scattering of spin-polarized electrons at interface imperfections.

Regarding (ii), we compare laser-driven spin transport in F|N bilayers to another fundamental process: laser-driven ultrafast magnetization quenching, which occurs already in single magnetic layers F. By measuring both the laser-induced spin transport in the F|N bilayer and the ultrafast magnetization quenching of F with THz emission spectroscopy, we make a surprising observation: The spin current in F|N is proportional to the rate of change of the magnetization in F. This result can consistently be explained by the assumption that both quantities scale with the instantaneous difference of the chemical potential of spin-up and spin-down electrons in the F layer.

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## Probing the Response of Ferromagnetic Order to Terahertz Excitation

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To push writing of magnetic information to ultrafast time scales, it is essential to understand the response of a magnetically ordered solid to an instantaneous perturbation. A model experiment is the excitation of a ferromagnet (such as Fe) by a femtosecond laser pulse. The resulting magnetization quenching is known to proceed on a surprisingly fast time scale of 100 fs, and this observation is still a topic of extensive research. One open question is how the transfer of spin angular momentum to the crystal lattice depends on the nature of the photoexcited electron distribution, that is, thermal (Fermi-function-like) vs nonthermal.

In a first experiment, we tune the electron distribution directly after optical excitation by variation of the photon energy. Low photon energies (in particular smaller than the thermal energy of 25 meV at 300 K) are expected to induce a transient distribution that enables a much smaller phase space for electron scattering than a distribution induced with optical photon energies (~1 eV). We perform a direct comparison of the ultrafast magnetization dynamics in Fe thin films following excitation with ultrashort optical (3.1 eV) and terahertz (4 meV) pump pulses. By thorough deconvolution of the shape of the pump pulse, the intrinsic time constants of magnetization quenching and magnetization recovery are extracted for both optical and terahertz excitation.

In a second experiment, we apply THz electric-field pulses to a Fe|Pt bilayer sample to generate THz spin currents by means of the spin Hall effect. To probe the population dynamics of spin-polarized electrons in the ferromagnetic Fe layer, we measure the instantaneous magnetic linear birefringence of our sample by means of a femtosecond laser pulse. For the first time, we find clear signatures of THz-field-driven spin accumulation. Our observations indicate that the lifetime of the injected excess spins in the Fe layer is shorter than 50 fs. We correlate this finding to the THz-induced magnetization of the first experiment.

## Phonon Polaritons in Polar Dielectric Heterostructures

Nikolai C. Passler, Christopher J. Winta, Ilya Razdolski, Martin Wolf,  
and Alexander Paarmann

Midinfrared nanophotonics can be realized using the sub-wavelength spatial confinement and field enhancement of surface phonon polaritons (SPhPs) supported in polar dielectrics. The long lifetimes of these excitations offer unique advantages for infrared nanophotonics over short-lived plasmon polaritons. However, SPhPs propagating on a single polar crystal surface are barely tunable, exhibit moderate field enhancements and are intrinsically limited to p-polarization. Furthermore, SPhPs are restricted to a small frequency window bound by the crystal's transverse and longitudinal optical phonon frequencies. Presenting four different model systems, we here demonstrate that these limitations can be overcome by employing stratified polar crystal heterostructures.

For ultrathin polar dielectric films (10-100 nm), the SPhP dispersion is strongly modified, resulting in unique ultrathin film modes that naturally reside at the zero-crossing of the dielectric function (epsilon-near-zero, ENZ). This ENZ condition leads to many attractive properties such as infinite phase velocity, scatter-free propagation, and enhanced nonlinear-optical effects. Experimentally, we show the full hybridization of ENZ and surface polariton modes in an ultrathin AlN film on a SiC substrate, and demonstrate that the coupling strength can be tuned using different AlN layer thicknesses [1]. For free-space excitation employing the FHI free electron laser, the same sample yields an enhanced second harmonic response, arising from the immense optical field enhancement associated with the ENZ ultrathin-film Berreman mode [2].

In contrast, a (non-polar) dielectric ultrathin film placed on a polar crystal surface results in a compression of the substrate polariton dispersion. We here employ a thin Ge<sub>3</sub>Sb<sub>2</sub>Te<sub>6</sub> (GST) film on SiC, where the non-volatile phase transition of GST enables active tuning with outstanding figures-of-merit for both the SiC substrate SPhP and the simultaneously supported s-polarized GST-film waveguide mode. Strikingly, for thin GST films, the confinement, dispersion and field enhancement of both modes can become comparable [3].

When multiple atomically thin layers of polar semiconductors are combined in a superlattice, new vibrational modes arise due to the modified chemical bonding at interfaces. This results in a unique polaritonic response, distinct from its constituent materials, which can be altered through layer thickness variation. Simultaneously, the strong crystal anisotropy in such a superlattice leads to hyperbolic polaritons — volume-confined modes with unbound momenta. Atomic-scale heterostructures thus provide a platform for novel user-designed SPhP materials [4].

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## Sum-Frequency Generation Spectroscopy and Microscopy using the FHI Infrared Free-Electron Laser

Riko Kiessling, Yujin Tong, Christopher Winta, Sandy Gewinner, Wieland Schöllkopf, Martin Wolf, and Alexander Paarmann

Infrared-visible vibrational sum-frequency generation (SFG) spectroscopy is a valuable optical method for the selective detection of low-energy vibrational excitations at surfaces and interfaces or in non-centrosymmetric media. Besides the spectroscopic information obtained by tuning the IR frequency in resonance with the vibrational modes, symmetry selection rules allow for interface-sensitivity. Moreover, when used for microscopy, the coherent second-order nonlinear process enables higher spatial resolution compared to linear infrared microscopy techniques [1]. However, due to the nonlinear nature of the process, intense infrared and visible light sources should be employed.

Here, we utilize the FHI free-electron laser (FEL) as unique source of narrow-band, wavelength-tunable ps-pulsed coherent IR radiation. In combination with a synchronized table-top fs oscillator, we have implemented FEL-based SFG spectroscopy. By means of balanced optical cross-correlation, the FEL pulse jitter was determined to be as low as ~100 fs (rms) and a timing drift in the order of a few ps/hour [2]. SFG measurements on polar dielectric materials with broken inversion symmetry allowed for a first benchmarking of this approach.

Adding spatial resolution to SFG spectroscopy is a promising way to visualize heterogeneous interfaces with vibrational contrast. Here, we used subdiffractive nanostructures supporting localized surface phonon polaritons to control electromagnetic fields at the nanoscale, and demonstrated SFG microscopy with long-wave FEL-based IR radiation. In our proof-of-concept experiment, a scanning-probe approach of tightly focused incident beams is employed to map out the localized polaritonic field enhancements [3].

Ultimately, we will implement FEL-based distortion-corrected wide-field SFG microscopy. This approach combines the advantages of nonlinear microscopy with those of oblique imaging techniques including an all-in-focus field of view. Here, in contrast to scanning-probe, the whole image is acquired simultaneously, enabling observation of dynamic processes, avoiding imaging distortions, and severely reducing sample damage thresholds. We anticipate this novel technique will provide new insight in various fields ranging from IR nanophotonics to electrochemistry.

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## Towards Nano-Scale Optical Spectroscopy Using Plasmonic Scanning Tunneling Microscope Junctions

Shuyi Liu, Hannes Böckmann, Melanie Müller, Adnan Hammud, Martin Wolf,  
and Takashi Kumagai

Spectro-microscopy on the atomic scale using the confined near-field in plasmonic STM junctions has attracted increasing attention. We have developed a new low-temperature STM for local optical excitation and detection, to perform atomic-scale linear and nonlinear optical spectroscopy and investigate plasmon-mediated physical and chemical processes in nanocavities. This new instrument was setup in June 2017 and has now achieved superior performance. A key advancement is the implementation of a parabolic mirror in UHV with a relatively high numerical aperture which can be precisely aligned by five-axis motion using piezo stages even at 5 K.

In addition, accurate control of the localized surface plasmon resonance (LSPR) in the STM junction is of fundamental importance for atomic-scale spectro-microscopy. We have demonstrated that the LSPR can be tuned by nanoscale tip fabrication by means of focused ion beam (FIB) milling [1]. One example are Fabry-Pérot type resonators for surface plasmons which we demonstrated for tip with a single groove on their shaft. Scanning tunneling luminescence spectra of these Fabry-Pérot tips exhibit spectral modulation resulting from interference between localized and propagating surface plasmon modes. In addition, the quality factor of the plasmonic Fabry-Pérot interference can be improved by optimizing the overall tip shape. We also showed that the FIB tip can be used to study plasmon-induced single-molecule reactions [2].

We also demonstrated a simple but novel plasmon-mediated phenomenon, namely plasmon-assisted resonant electron tunneling from an Ag or Au tip into field emission resonances (FERs) of a Ag(111) surface induced by visible CW laser excitation of the STM junction [3]. As a hallmark of plasmon-assisted resonant tunneling, we observe a downshift of the bias of the first peak in the FER spectra equal to the incident photon energy. STM-induced luminescence measurement for the Ag and Au tip reveals the clear correlation between the laser-induced change in the FER spectra and the plasmonic properties of the junction. Our results clarify a novel resonant electron transfer mechanism in a plasmonic nanocavity.

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## Towards Atomic-Scale Vibrational Spectroscopy Using Tip-Enhanced Raman Spectroscopy

Shuyi Liu, Melanie Müller, Adnan Hammud, Martin Wolf, and Takashi Kumagai

Tip-enhanced Raman spectroscopy (TERS) has recently demonstrated its intriguing capability to resolve vibrational modes within a single molecule at nearly angstrom resolution [1]. This finding has raised fundamental and intriguing questions of light-matter interactions on the atomic scale.

We have developed low-temperature TERS with home-build setup and investigated ultrathin ZnO layers epitaxially grown on Ag(111). We have clarified the role of physical and chemical enhancement mechanisms in TERS and demonstrated tip-enhanced “resonance” Raman scattering (TERRS) where both enhancement mechanisms are operative [2]. In combination with scanning tunneling spectroscopy (STS), it was demonstrated that the TERRS intensity strongly varies with the local electronic structure at the ZnO/Ag(111) interface. The spatial resolution of TERRS depends on the tip-surface distance reaching nearly 1 nm in the tunneling regime, which can be rationalized by strong field confinement resulting from an atomic-scale protrusion on the tip apex. Comparison of STS and TERRS mapping clearly shows a correlation between resonantly enhanced Raman scattering and the local electronic states with near-atomic resolution. This suggests that TERRS offers a new approach for the atomic-scale optical characterization of local electronic states.

Furthermore, we also demonstrate in experiments at 5K chemical bond formation and breaking in the atomic contact regime between the STM tip and the ZnO layer. Upon formation of a chemical bond between the tip apex atom and oxygen of the outermost ZnO layer, a characteristic new vibrational mode appears in TERS. It appears that the stable atomic contact results in unexpectedly large enhancement of Raman scattering where conventional selection rules also break down. Our results may provide atomistic insight into the structure and dynamics of atomic and molecular junctions as well as light induced transport phenomena.

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## Implementation and Characterization of a Photoexcited THz-gated Scanning Tunneling Microscope

Melanie Müller, Natalia Martín-Sabanés, Sarojini Mahajan, and Martin Wolf

THz-gated Scanning Tunneling Microscopy (THz-STM) uniquely combines sub-molecular spatial with femtosecond temporal resolution, as first impressively demonstrated in 2016 [1]. Combined with optical photoexcitation THz-STM provides a powerful platform to study the local atomically-resolved dynamics of photoexcited nanostructures and surfaces, giving access to a broad variety of spatio-temporal phenomena relevant in surface science. In this regard, we developed an ultrahigh-vacuum STM/AFM system optimized for broadband excitation from the visible through the THz spectral range using two large-aperture motorized parabolic mirrors integrated on the STM platform. After installation of the instrument end of 2018, our setup is operational at room temperature since February 2019 employing a broadband spintronic emitter (STE) as source of single-cycle THz pulses to gate the tunneling junction.

Optimal operation of the THz-STM requires exact knowledge and control of the THz near-field in the STM junction. As this can deviate significantly from its corresponding far-field waveform measured by electro-optic sampling, we characterized the THz near-field directly in the time-domain via THz-induced modulation of photoemission from the tip [2] at distances down to few nanometers relevant for STM. We find pronounced spectral filtering due to frequency-dependent spatial overlap of the point-like STM tip inside the THz focus, and only little variations of the waveform between different tips. We further identify variations of the waveform's shape and polarity by variation of the STE excitation conditions, allowing for precise THz near-field control in the STM junction.

As a first step towards probing femtosecond photocarrier dynamics on the nanoscale, we investigate the influence of long-lived charge carriers on the sampled THz waveform in a metal-HOPG junction, and study effects of photoelectron propagation in the tunneling junction. Next steps include the ultrafast probing and imaging of photoexcited semiconductor surfaces and metal-semiconductor interfaces at room temperature. At last, upgrade of the system to cryogenic temperatures is currently under way to access a wider class of samples and to ensure sufficient stability of the system.

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## High Resolution Scanning Gate Microscopy

Alexander Kölker, Takashi Kumagai, Martin Wolf, and Matthias Koch

Scanning probe microscopy (SPM) is a versatile technique for the characterization of the surface. The high spatial resolution enables not only to imaging of materials with atomic resolution but also manipulation of individual atoms or molecules. However, a typical setup is limited to two electrodes: the SPM tip and the (potentially conducting) sample. Therefore, in transport experiments the current is averaged over the whole tip-sample junction, although the chemical composition of the junction has a strong influence on the measured signal [1]. We address this problem by integrating additional electrodes on the sample in close proximity to the surface. These electrodes can then be used for transport experiments or scanning gate microscopy, where the SPM tip is biased and acts as a flying nanosized gate.

We realize the electrodes by ultra-shallow ion-implantation in an otherwise at low temperature insulating silicon crystal. The ion-implanted wires bridge the gap from mm large conductive patches down to  $\mu\text{m}$  small contacts. We demonstrate ohmic behavior of these electrodes, even after high temperature surface preparation. Atomically flat surfaces are observed which makes the sample compatible to high quality SPM experiments. Adding an insulating region between two electrodes leads to a large non-conductive region which appears in the IV traces. These insulating gaps withstand high temperature sample treatment  $>1000^\circ\text{C}$ , indicating that dopant diffusion is sufficiently low.

In the future we want to extend these  $\mu\text{m}$  large electrodes with STM hydrogen resist lithography to pattern truly nano-scale contacts. These can be use to electrically contact organic compounds or two-dimensional transition-metal dichalcogenide flakes for transport experiments or excitation of molecular processes.

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## Quantifying the Interfacial Contribution in Vibrationally Resonant Sum Frequency Generation Spectroscopy

Martin Thämer, Tobias Garling, Martin Wolf, and R. Kramer Campen

Much of the chemistry that controls the world in which we live occurs at the interface between two condensed phases. Because such systems cannot be probed by electrons or atoms, their experimental characterization is challenging. Vibrationally resonant Sum Frequency Generation (VSFG) spectroscopy is an all-photon nonlinear optical technique in which the output of pulsed infrared and visible lasers are overlapped spatially and temporally at an interface and emitted sum frequency field detected. In the limit of two bulk phases that are inversion symmetric and whose optical response is dominated by dipolar contributions, VSFG is an interface-specific vibrational spectroscopy. Inspired by this limiting behaviour homodyned VSFG spectroscopy, in which the intensity of the emitted sum frequency field is measured as a function of photon energy of the incident infrared, has been increasingly applied to probe the interface of a variety of condensed phases, and the chemistry that occurs at them, over the last 30 years. It is far from clear, however, that many systems of interest behave in this idealized manner. For example, many solids are not inversion symmetric in bulk; and both liquids and solids can have a considerable thickness of the interfacial layer, often have broad, weakly-frequency dependent spectral features that overlap with narrow vibrational resonances and may have substantial quadrupolar contributions.

Quantitatively separating these contributions requires more information than present in a homodyned VSFG spectra. As a first step in this direction we have recently constructed a collinear, heterodyned, balance-detected, time domain VSFG spectrometer capable of high sensitivity measurements of the absolute phase and amplitude of the emitted sum frequency field [1]. Using this spectrometer, and leveraging the different structural symmetries of surface and bulk, we quantified the surface spectral response at the air/ $\alpha$ -quartz interface ( $\alpha$ -quartz is a prototypical non-inversion symmetric solid) [2]. To extend this approach to buried interfaces in which one or both of the bulk phases is dispersive requires quantitative compensation of the “walk-off” between the reference pulse used in the heterodyned measurement and the incident fields driving the sum frequency process at the sample surface. We have developed a compensation scheme, using a birefringent calcite crystal, that makes this process quantitative and straightforwardly reproducible. Finally, because our spectrometer is both collinear and measures in the time domain, it is possible to record sum and difference frequency (VDFG) spectra concurrently. Because material resonances are related by  $\pi$  in the two optical processes, this capability makes separation of narrow molecular resonances from a broad background straightforward as we demonstrate for an octadecanethiol SAM on Au. Additionally, because phase matching differs between SFG and DFG, combining these two measurements creates a novel optical observable of depth-dependent interfacial structure.

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## Characterizing Potential Dependent Structure at the Electrode/Electrolyte Interface

Gregor Zwaschka, Xin Gong<sup>a</sup>, Martin Wolf, R. Kramer Campen, and Yujin Tong

The most promising pathways to a sustainable global energy economy involve the electrocatalytic conversion of small organic molecules and water splitting. Building the best possible devices based on this chemistry requires understanding the *mechanism* of each of the relevant reactions. Despite decades of study, gaining experimental insight into mechanism in heterogeneous electrocatalysis has proven extremely challenging. A prerequisite to such insight is molecular-scale characterization of all aspects of the electrode/electrolyte interface: we require experimental insights that allow characterization of reactants/products, ions and water (for the case of aqueous electrolyte).

Vibrationally resonant Sum Frequency Generation (VSFG) spectroscopy is a nonlinear optical spectroscopy that is interface specific by its symmetry selection rules and, because it is a photon-in / photon-out technique, in principle applicable to interfaces between condensed phases. Here we apply VSFG spectroscopy to probe the surface of Pt and Au electrodes in contact with aqueous electrolyte as a function of potential in a thin film spectro-electrochemical cell for three different systems: (i) we describe the potential dependent VSFG response of adsorbed  $\text{SO}_4^{2-}$  on a Pt(111) working electrode and show how it reports both on adsorbed anion structure and interfacial electric field [1,2] (ii) we characterize the Pt-H spectral response of so-called "under-potential deposited" hydrogen and relate its structure to  $\text{H}_2$  formation and (iii) we probe the structure of a prototypical small organic molecule, *i.e.* dimethylaminopyridine, adsorbed on an Au working electrode and show how it changes orientation and (de)protonates with changing potential. This body of work, in conjunction with our earlier study probing the potential dependent structure of water in contact with an Au working electrode [3] demonstrate the capability to fully characterize the molecular-scale structure of all aspects of the electrolyte at metal electrode / electrolyte under potential control: reactants/products, *i.e.* adsorbed H and dimethylaminopyridine, ions, *i.e.* adsorbed  $\text{SO}_4^{2-}$ , and water.

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## Exploring the Mechanism of the Hydrogen Evolution Reaction on Pt and Au with Femtosecond Potential Perturbation Experiments

Gregor Zwaschka, François Lapointe<sup>a</sup>, Martin Wolf, Yujin Tong, and R. Kramer Campen

Our hydrocarbon combustion based global energy economy has significant adverse consequences on climate and the environment. An energy economy based on H<sub>2</sub> combustion would avoid many of these consequences but requires a sustainable means of producing H<sub>2</sub>. Perhaps the most straightforward way of doing so is water electrolysis using a renewable energy source. Building the best possible devices using this chemistry requires experimental insight into the mechanism of H<sub>2</sub> formation. Gaining such insight is challenging for, at least, two reasons: (i) Characterization of molecular-level structure at electrode/electrolyte interfaces *operando* is not possible using conventional surface science tools (ii) Even if were possible characterizing the interface under steady-state reactive conditions provides little definitive insight into mechanism: one cannot distinguish intermediates from unwanted side-products. Here we address these challenges by performing perturbation experiments: we initiate charge transfer on Au or Pt working electrodes and then track the resulting interfacial structure with femtosecond time-resolution by measuring one and two pulse induced photocurrents or voltages and probing the electrode/electrolyte interface using Vibrationally resonant Sum Frequency Generation (VSFG) spectroscopy as a function of delay.

In particular we show by measuring the photocurrent induced by femtosecond optical pulses on various Pt single crystals that while both the elementary, femtosecond, step of charge transfer and slower structural dynamics in the electrolyte are important in understanding H<sub>2</sub> production, trends in reactivity between different Pt single crystal surfaces correlate with, off-line measurements of H<sub>2</sub> activity [1]. Measurements of the Pt-H VSFG spectral response as a function of delay provide a bond-resolved picture of this fs induced charge transfer. While H adsorbs strongly on Pt at steady-state potentials at which H<sub>2</sub> is not produced, it does not on Au. Here we show, using a two-pulse photovoltage technique, that fs optical excitation leads to the production of an interfacial solvated electron at the Au electrode/aqueous electrolyte interface. Some portion of the transferred electrons are delocalized, ~ 5% populate pre-existing defect sites. Over the enduring 500 fs, delocalized electrons either relax back to Au or form localized solvated electrons whose optical response different from those in bulk and that, over nano-microsecond timescales drive H<sub>2</sub> production.

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## Characterizing the Oxide/Water Interface from the Oxide's Perspective

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Characterizing the surface chemistry of the oxide/water interface *in-situ* is important for a wide variety of geochemical systems and any device based on semiconductor (photo)-electrochemistry. Despite such clear justification, molecular-scale characterization of this interface from the perspective of the oxide, *e.g.* probing solvation or aqueous chemistry induced reconstruction or the link between oxide surface structure and reactivity, is extremely challenging. Part of the challenge lies in the absence of analytical tools that allow characterization of oxide surface structure and bonding while in contact with water: techniques that employ electrons or molecules as atoms or probes can generally not be employed. In principle surface phonon spectroscopy offers such insight however the technique typically employed, *i.e.* Electron Energy Loss Spectroscopy (EELS), is all-electron and thus cannot be employed in ambient conditions. Vibrationally Resonant Sum Frequency Generation (VSFG) spectroscopy is a laser-based nonlinear optical technique that is, by its symmetry selection rules, interface-specific for systems with two bulk phases with inversion symmetry. Because it is photon-in/photon-out it overcomes the EELS limitation.

We have previously demonstrated the application of VSFG spectroscopy to characterize the surface structure of the  $\alpha$ -Alumina (0001) surface in ambient [1]. Here we extend our prior studies and characterize the surface phonon spectra and its azimuthal dependence, of well-defined  $\alpha$ -Alumina (0001) and (11-20) single crystal surfaces in ultra-high vacuum in contact with well-defined amounts of water. In comparison with the high frequency O-H stretching modes, surface phonon vibrations are easily distinguishable from those of the adsorbed water thus allow unambiguous determination of the surface structures relevant to surface macroscopic properties. Taken together these measurements, in conjunction with normal mode calculations of well-defined single crystal surfaces, offer insight into the origin of the relative unreactivity of the (0001) surface and the evolution of surface structure (in particular its symmetry) with solvation for both surfaces.

In addition, using a meniscus sample geometry, we demonstrate the application of this approach to oxide formation: the electrooxidation of an Au working electrode in contact with acidic aqueous electrolyte. While the mechanism has been well studied previously both experimentally and theoretically, the rate limiting step has been controversial. Here, by showing the potential dependent appearance of surface Au-OH species, we demonstrate that hydroxylation controls the kinetics of Au electrooxidation.

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## Nonlinear THz-Raman Spectroscopy Reveals Structural Effects and Energy Transfer Pathways of Ionic Solutions

Vasileios Balos, Martin Wolf, and Mohsen Sajadi

Intermolecular dynamics in the terahertz (THz) frequency range are decisive to the physical, chemical and biological properties of liquid water. To capture microscopic insights on the intermolecular interactions, we use intense THz pulses to resonantly drive nonlinear effects in liquid water and resolve its real-time Raman response [1]. We show that, upon THz excitation of the rotational degrees of freedom of water molecules, the deposited THz energy is transferred to the neighboring molecules and increase their translational kinetic energy [2].

The strength of Rot-Trans coupling is then used as an intermolecular probe to investigate the ion-induced distortion of the hydrogen bonding network of water. We directly observe that in the presence of strong cations the signal amplitude increases, while strong anions reduce the signal amplitude. This experimental finding is in line with the picture that cations stick to the electron lone pair of water and polarize it even further, and thus the Rot-Trans coupling is strengthened. Highly charged anions on the other hand, accommodate themselves in the H-bonding network, thus the Rot-Trans coupling is weakened. Our results are in direct contrast to the current view that assigns the structure making ability of ions, independent of their sign, solely to their charge [3].

We use the same concept also to provide a novel experimental means for transferring energy from solvent to solutes. Here, we use the collision induced Raman response of highly polarizable ions as the energy transfer indicator. By direct coupling of the THz energy into the rotational degrees of freedom of protic and aprotic polar liquids, we resolve the Raman response of the dissolved ions. This finding can play a decisive role in the future development of coherent control of chemical reactions.

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## Distinct Terahertz Magnetic- and Electric-Field Response of Molecules

Vasileios Balos, Genaro Bierhance, Martin Wolf, and Mohsen Sajadi

Intense single-cycle terahertz (THz) pulses have great potential to reveal unprecedented insight on the low-frequency dynamics of molecular systems [1]. Our table-top THz sources can now routinely generate THz pulses with  $E \approx 3$  MV/cm electric- and  $B = E/c \approx 1$  T magnetic-field strength [2]. Such phase-locked electromagnetic pulses are short enough to drive excitations with sub-picosecond time resolution, and also long enough to create the real-time “slow-motion movie” of light-matter interaction.

Using intense THz magnetic fields, we induce ultrafast rotation of the plane of polarization of a short optical pulse traversing polarizable molecular liquids. We explain the effect as the bending of the electric polarization induced by the optical short laser pulse under the influence of a Lorentz force emanated from the THz magnetic field. The observed effect scales linearly with the THz magnetic field and quadratically with the molecular polarizability.

Using such intense THz electric fields, we demonstrate the tunability of the molecular rotational distributions in liquids, by obtaining partially aligned and anti-aligned states. The mechanism of this process is discussed in terms of an efficient mechanical THz torque to the permanent molecular dipoles. It is shown that by tuning the THz pulse frequency, phase and also the strength of the THz electric coupling to the molecular dipole moment, different rotational distributions can be achieved. THz-control of the molecular rotational distribution in liquids opens a new path to study intermolecular interactions and offers a new means for coherent control of chemical reactions.

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# Theory Department

## Poster List

- TH 1 **From NOMAD (Novel Materials Discovery) to FAIR-DI (FAIR Data Infrastructure)**  
Claudia Draxl and Matthias Scheffler *et al.*
- TH 2 **Advances in and for FHI-aims**  
Volker Blum, Sebastian Kokott, Christian Carbogno *et al.*
- TH 3 **Advanced Electronic-Structure Approaches in FHI-aims**  
Sheng Bi, Maria Dragoumi, Igor Ying Zhang, Christian Carbogno, Sergey V. Levchenko, and Matthias Scheffler
- TH 4 **Electronic Transport at Finite Temperature Beyond Perturbation Theory**  
Marios Zacharias, Zhenkun Yuan, Matthias Scheffler, and Christian Carbogno
- TH 5 **Learning Materials Properties Across the Chemical Space with SISSO, Multi-Task Learning, and Neural Networks**  
Emre Ahmetcik, Christopher J. Bartel, Christian Carbogno, Guohua Cao, Bryan R. Goldsmith, Huijun Liu, Charles B. Musgrave, Runhai Ouyang, Christopher Sutton, Zhenyu Zhang, Matthias Scheffler, and Luca M. Ghiringhelli
- TH 6 **Identifying Interpretable Descriptors for Materials Properties with Subgroup Discovery and Information Theory**  
Mario Boley, Francesc Illas, Sergey V. Levchenko, Aliaksei Mazheika, Benjamin Regler, Matthias Rupp, Christopher Sutton, Jilles Vreeken, Yang-Gang Wang, Luca M. Ghiringhelli, and Matthias Scheffler
- TH 7 **Oxidation of Elemental and Alloyed Transition-Metal Surfaces at Realistic Temperature and Pressure Conditions**  
Zhong-Kang Han, Santiago Rigamonti, Maria Troppenz, Martin Kuban, Axel Hübner, Christopher Sutton, Luca M. Ghiringhelli, Sergey V. Levchenko, Claudia Draxl, and Matthias Scheffler
- TH 8 **Stable and Metastable Crystal Structures of Transparent Semiconducting Oxides from First Principles, Experiments, and Crowd-Sourced Machine Learning**  
Christopher Sutton, Martin Albrecht, Lars Blumenthal, Jacek Golebiowski, Xiangyue Liu, Yury Lysogorskiy, Thomas Hammerschmidt, Toni Markurt, Robert Schewski, Charlotte Wouters, Takenori Yamamoto, Angelo Ziletti, Luca M. Ghiringhelli, and Matthias Scheffler
- TH 9<sup>OHG</sup> **Nuclear Quantum Effects in Response Properties of Materials and Electron-Vibrational Coupling**  
Yair Litman, Karen Fidaryan, Hagen-Henrik Kowalski, Matthias Scheffler, Christian Carbogno, and Mariana Rossi
- TH10<sup>OHG</sup> **The Role of Temperature on Interface Level Alignment of Strong Organic Electron Acceptors Adsorbed on 3D and 2D Semiconductors**  
Haiyuan Wang, Sergey V. Levchenko, Dmitrii Maksimov, Carsten Baldauf, Matthias Scheffler, and Mariana Rossi

- TH11<sup>OHG</sup> ***In Silico* Characterisation of Anharmonic Vibrational Properties of Molecular Crystals**  
Nathaniel Raimbault, Marcin Krynski, and Mariana Rossi
- TH 12 **Atomic and Electronic Structure of  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> Surfaces at Realistic Conditions: Theory and Experiment**  
Konstantin Lion, Sergey V. Levchenko, Matthias Scheffler, and Claudia Draxl
- TH 13 **High-Precision yet Efficient High-Throughput Search for Thermal Insulators**  
Florian Knoop, Thomas Purcell, Marcel Hülsberg, Maja-Olivia Lenz, David Hicks, Stefano Curtarolo, Matthias Scheffler, and Christian Carbogno

## From NOMAD (Novel Materials Discovery) to FAIR-DI (FAIR Data Infrastructure)

Claudia Draxl<sup>a</sup> and Matthias Scheffler and the big team of NOMAD and FAIR-DI colleagues<sup>1,2</sup>

This poster addresses the fourth paradigm of materials research, big-data driven materials science, and our approach<sup>3</sup> to implement it in computational materials science. We discuss the state-of-the-art as well as key issues that hamper progress. And, we sketch extensions towards experimental materials sciences.

For furthering big-data-driven computational materials science, Open Data and an all-embracing sharing, an efficient and FAIR<sup>4</sup> data infrastructure and consideration of the rich ecosystem of computer codes used in the community are of critical importance. With this intention, the NOMAD Repository was developed (open since 2014) and advanced into the NOMAD Center of Excellence (since 2015).<sup>1</sup> With its “open to all codes” concept, NOMAD is orthogonal to but synergistically interacts with other data collections in computational materials science worldwide, in particular the Materials Project, AFLOW, and OQMD.

The largely unsolved issues are the A and I of FAIR<sup>4</sup>, specifically the development of metadata and their intricate relationships, which should be reflected in ontologies. Obviously, a FAIR data infrastructure – for being accepted by the community – should work without bureaucratic hurdles or the needs for special training. For NOMAD we therefore accept the input and output files as they are produced by the employed code (this raw data goes into the NOMAD Repository). Then, NOMAD converts this data into a standardized and normalized form (the NOMAD Archive). Only the Repository and Archive together make the data FAIR. With the NOMAD Encyclopedia even the non-expert can look at the various results contained in the Archive. And the NOMAD Analytics Toolkit offers several artificial-intelligence tools for analyzing the data further and for identifying possible correlations and patterns and building “maps of materials”.<sup>1,3</sup> The Toolkit also offers tools for identifying statistically exceptional regions in the data space and for error estimates (see TH 5 and TH 6).<sup>5</sup> The poster concentrates on computational material science but also addresses the ongoing extension of the “NOMAD concept” to experimental works which brings in additional, highly complex but also scientifically challenging aspects. For example, for experimental data, we need a full characterization of the sample, the description of the apparatus, the measurement conditions and the measured quantity. This is detailed on the poster AC 1.4 by Trunschke et al. This advancement is also the concept behind the international association FAIR-DI e.V.<sup>2</sup>, which recently submitted a proposal for a large-scale consortium of the NFDI (German Research-Data Infrastructure).<sup>6</sup>

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1. NOMAD CoE (Novel Materials Discovery Center of Excellence): <https://nomad-coe.eu>.
2. FAIR-DI e.V. is an international non-profit association aimed at the development of a FAIR Data Infrastructure for Physics, Chemistry, Materials Science, and Astronomy: <https://fairdi.eu>.
3. C. Draxl and M. Scheffler. Plenary Chapter in Handbook of Materials Modeling (eds. Yip and Andreoni), Springer (2019): <https://arxiv.org/ftp/arxiv/papers/1904/1904.05859.pdf>.
4. FAIR stands for Findable, Accessible, Interoperable and Re-usable. The FAIR Data Principles: <https://www.force11.org/group/fairgroup/fairprinciples>.
5. C. Sutton *et al.*, Domains of Applicability of Machine-Learning Models for Novel Materials Discovery, to be published.
6. FAIRmat is a proposed Consortium for the German Research-Data Infrastructure (NFDI) on FAIR Data Infrastructure for Materials Science and Related Fields: <https://fairdi.eu/fairmat>.

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## Advances in and for FHI-aims

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FHI-aims (Fritz Haber Institute *ab initio* molecular simulations) is an all-electron, general-purpose electronic-structure code used for computational molecular and materials research by a global community of developers and users at FHI, in academia, and in industry. Numeric atom-centered basis sets are used as basis sets, offering numerical precision on par with the best available benchmark codes in density functional<sup>1,2</sup> and many-body<sup>3</sup> theory, at a computational cost that is comparable with plane-wave pseudopotential methods. The code is routinely applicable to thousands of atoms using semilocal and hybrid functionals and shows excellent scalability on current high-performance platforms. Further strengths include advanced electronic-structure developments for molecules and solids (poster TH 3) and seamless integrability into complex, externally managed simulation environments, including as a parallel library through python or using internet sockets (e.g., poster TH 13).

We here describe recent developments in FHI-aims and in open-source high-performance libraries on which the code relies. Developments focused on the physical description of molecules and materials include:

- Relativistic effects in FHI-aims are routinely treated by a carefully benchmarked scalar relativistic approach, with spin-orbit coupling added for band structures.<sup>4</sup> For the electronic structure, a new software layer extends the existing spin-orbit coupling infrastructure to access different topological invariants. Separately, a newly developed self-consistent quasi-four-component treatment gives access to total energies and band structures that recover the Dirac-Kohn-Sham equations essentially exactly.
- For FHI-aims, a recent in-depth extension of “+*U*” provides access to enhanced localized projectors, providing significant control over a defining aspect of “+*U*” that is often taken as a black box in the literature.<sup>5</sup>
- In the realm “beyond DFT”, we describe progress in a new periodic all-electron **GW** implementation. Furthermore, a precise **GW** contour deformation treatment of the self-energy on the real axis was developed<sup>6</sup>, particularly enabling accurate core-level spectroscopies for molecular systems. The particle-particle random-phase approximation (RPA) was recently incorporated.<sup>7</sup>
- For large and complex systems, core-level spectroscopy in FHI-aims was recently developed and analyzed in depth based on traditional DFT-based core-hole occupation constraints<sup>8</sup>, with a protocol for accurate results demonstrated for semilocal DFT.
- Existing and new many-body dispersion treatments are now incorporated in a new, open-source library with enhanced scalability.<sup>9</sup>
- Implicit solvation for finite systems<sup>10</sup> is extended to more complex solute geometries and particularly to solutes at liquid-liquid interfaces with different dielectric properties.

On a technical level, developments that extend the applicability, scalability, and usability of FHI-aims include:

- For hybrid density-functional theory, a refined load-balancing approach further reduces both CPU and memory requirements, especially for heavy-element containing systems.

- The code supports semilocal DFT simulations on GPU architectures at the heart of today's top supercomputers.
- Ongoing work focuses on symmetry exploitation in solid systems, benefitting, e.g., rapid symmetry-guided structure search algorithms used in the community.
- A reanalysis of structure relaxation for bulk solids including unit cell degrees of freedom provides significant speed-ups especially for systems with symmetries. A new fine-grained parallelization scheme additionally benefits structure optimization for large systems (thousands of atoms).

For the majority of problem sizes, up to thousands of atoms, the ELPA eigenvalue solver<sup>11,12</sup> is the bedrock of scalability in FHI-aims. We describe the latest optimizations of the ELPA library, a solver for dense generalized eigenproblems, for new HPC architectures including GPU-acceleration.<sup>13</sup> Besides confirming that “*ELPA outperforms ScaLAPACK on all architectures tested and all matrix sizes and concurrencies*”, as recently found by NERSC and Cray<sup>14</sup>, we demonstrate that these advantages are accessible in an unprecedented user-friendly fashion by the new API<sup>12</sup>, which allows to choose performance-critical parameters in an automated fashion. Finally, solvers beyond the  $O(N^3)$  limit of traditional eigensolvers for large-scale density-functional theory are accessible through the ELSI infrastructure<sup>15</sup>, paving the way for handling system sizes beyond thousands of atoms in all-electron simulations using FHI-aims. This work is supported by the projects ERC TEC1p and ZIM (BMW).

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## Advanced Electronic-Structure Approaches in FHI-aims

Sheng Bi, Maria Dragoumi, Igor Ying Zhang<sup>a</sup>, Christian Carbogno,  
Sergey V. Levchenko<sup>b</sup>, and Matthias Scheffler

Despite the unparalleled popularity of density-functional theory (DFT), the widely used semi-local density-functional approximations (DFA) for exchange and correlation (XC) suffer from self-interaction errors (also recognized as fractional-charge errors), which lead to unphysical long-range charge transfer in molecules and an underestimation of the band gaps in solids.<sup>1</sup> To address this challenge, it has been suggested to either introduce a self-interaction correction as proposed by Perdew and Zunger (PZ-SIC)<sup>2</sup> or a localized orbital scaling correction (LOSC) as proposed by Weitao Yang and coworkers<sup>3</sup> to existing semi-local DFAs. Alternatively, quantum-chemistry methods<sup>4</sup> take the correlation effects into account in the explicit many-body picture, which solves the self-interaction error and improves the assessment of correlations. This report presents significant new developments in this regard for the all-electron code FHI-aims.<sup>5</sup>

1) *Self-consistent implementation of PZ-SIC across DFAs for finite and periodic systems*: Unlike traditional PZ-SIC implementations,<sup>1,2</sup> we solve the PZ-SIC constraint as a self-consistent eigenvalue problem, which has two advantages: First, our implementation is applicable with all semilocal DFAs including the promising meta-GGA SCAN,<sup>6</sup> since no derivatives of the XC functionals are needed. Second, the PZ-SIC can be recast in terms of Wannier functions for solids, in close analogy to FHI-aims' internal representation, thus allowing us to treat molecules and periodic systems on an equal footing. The accuracy and efficiency of our PZ-SIC implementation is demonstrated and validated for LDA, GGA, and SCAN DFAs using a test set of molecules, polymers, and solids. In particular, we investigate the performance of PZ-SIC with SCAN for simple group-IV semiconductors/insulators such as diamond and for Mott insulator such as CuO and FeO. We find that PZ-SIC improves the band gaps and the band structure in comparison to SCAN.

2) *Møller-Plesset second-order perturbation theory (MP2) and beyond for single-particle excitation energies (SPE)*: We go beyond the standard MP2 method by solving the Dyson equation, which renormalizes the MP2 correlation via an infinite summation over a selective sequence of diagrams. Moreover, our implementation features an efficient treatment of the singularity of the Coulomb potential in MP2-SPE calculations, which allows us to achieve convergence already with sparse *k*-grids. We take the band gaps for a wide range of semiconductors and insulators to demonstrate that the new approach shows a competitive or even superior performance in comparison to state-of-the-art methods, such as hybrid functionals and the GW approximation.

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## Electronic Transport at Finite Temperature Beyond Perturbation Theory

Marios Zacharias, Zhenkun Yuan, Matthias Scheffler, and Christian Carbogno

In the last decade, the first-principles assessment of electron-phonon interactions has led to fundamental insights into the electronic properties of semiconductors. For example, we can now calculate a material's temperature-dependent electronic band structure and conductivity by evaluating the electronic self-energy.<sup>1</sup> However, current treatments of the electron-phonon interactions are perturbative and thus rely on **two** fundamental approximations: a *harmonic* description of the nuclear motion and a *first-order* model of the coupling elements. Clearly, these approximations break down at elevated temperatures and/or in materials with highly mobile nuclei and weak bonding.<sup>2,3</sup>

In this work, we present a novel, non-perturbative first-principles methodology that overcomes **both** these approximations. The approach accounts for *anharmonic* effects in the nuclear motion with *ab initio* molecular dynamics (aiMD) simulations, while the evolution of the electronic structure during the aiMD trajectory is used to incorporate electron-vibrational coupling *to all orders*. To this purpose, we have implemented an efficient unfolding technique for electronic band structures in the all-electron, numeric atom-centered orbitals code *FHI-aims*<sup>4</sup>. By this means, spectral functions are obtained from aiMD simulations performed in supercells, thus giving access to fully anharmonic, temperature-dependent electronic self-energies in the fundamental Brillouin zone. First, this allows us to determine thermodynamic equilibrium properties that include the renormalization induced by the nuclear motion, i.e., temperature-dependent (direct and indirect) band gaps, effective masses, and linewidths. Second, the approach can be exploited to efficiently evaluate non-equilibrium properties: We show that the electronic charge and heat flux occurring during adiabatic aiMD can be obtained from the time-dependent spectral functions, which in turn allows to compute the thermoelectric transport coefficients within the Green-Kubo formalism<sup>5</sup>. We carefully validate the developed approach by comparing to literature results<sup>6</sup> obtained for simple, harmonic group-IV and III-V semiconductors, for which perturbative methods hold. In particular, we show that quasi-harmonic long-range effects<sup>2</sup> can be efficiently incorporated by statistical sampling in sufficiently large supercells<sup>7</sup>, which reduces numerical noise and massively accelerates the convergence both in aiMD simulation time and supercell size. Furthermore, we apply our methodology to investigate the electronic structure and transport coefficients of complex anharmonic perovskites that constitute a formidable challenge for perturbative approaches<sup>3</sup>. Eventually, we discuss the physical origin of higher-order phonon-phonon and electron-phonon couplings and critically analyze the role of the exchange-correlation functional in their assessment. This work is supported by the ERC TEC1p project.

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## Learning Materials Properties Across the Chemical Space with SISO, Multi-Task Learning, and Neural Networks

Emre Ahmetcik, Christopher J. Bartel<sup>a</sup>, Christian Carbogno, Guohua Cao<sup>b</sup>, Bryan R. Goldsmith<sup>c</sup>, Huijun Liu<sup>b</sup>, Charles B. Musgrave<sup>a</sup>, Runhai Ouyang<sup>d</sup>, Christopher Sutton, Zhenyu Zhang<sup>e</sup>, Matthias Scheffler, and Luca M. Ghiringhelli

Discovering a new functional material for a specific technological application can be a slow and uncertain process. In a computational study, the typical approach is to identify a materials class as promising for a certain function and for each material in this class the ground-state structure is determined via minimization of a first-principles, reliable potential-energy surface (PES). Then, a target property that characterizes the desired function is evaluated. In this context, artificial intelligence (AI) has attracted wide attention for building surrogate models that learn the PES on the basis of a given dataset. Once the models have been trained, they are orders of magnitude faster to evaluate than the parent model and some of them have been shown to be reliable in their predictions. Therefore, they can be ideally used to speed up the determination of ground-state structures. However, two issues still exist even if routine methods to learn reliable surrogate models were available: *i*) typically such models are not transferable across the chemical space (i.e. they can be used only with the set of atomic species they have been trained on) and *ii*) also with a quick PES evaluation, the global optimization of a structure can be an overwhelming problem.

Here, we show two complementary AI approaches addressing either issue. To address *i*), we present a strategy for learning chemically transferable PESs. This is achieved by combining a linear regression on a simple basis-set expansion of the PES over atomic relative positions, where the regression coefficients depend on atomic features (e.g., electron affinity, ionization potential) in a way that is learned via a neural-network. We show that a model trained on a set of octet binaries is able to predict stable and metastable structures for compositions not present in the training set.

To address *ii*), we have developed the *sure independence screening and sparsifying operator* (SISO) to learn a target property for a class of materials directly from their chemical formulas, whereas the actual atomic structure remains implicit in the learned model. SISO works by first systematically and unbiasedly constructing a huge number of possible descriptors, shaped as analytical formulas built from a set of given candidate features. Then, the best descriptor is identified within the set of candidates via *compressed sensing*. The method is also extended to multi-task (MT) learning in order to model with one descriptor several (related) properties. SISO is applied to predict: a) whether  $ABX_3$  materials ( $X = O, F, Cl, Br, I$ ) are stable or not in the perovskite structure and we find a simple descriptor that not only outperforms the well-known Goldschmidt tolerance factor, but also provides an estimate of the accuracy of the prediction; b) whether materials in the tetradymite family are topological insulators and we find a simple descriptor depending only on atomic numbers and electronegativities, which can be extended to layered tetradymite materials without retraining; c) in the MT-SISO flavor, the phase diagram for octet binaries comprising 5 different crystal-structures. This work is supported by the ERC TEC1p project.

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## Identifying Interpretable Descriptors for Materials Properties with Subgroup Discovery and Information Theory

Mario Boley<sup>a</sup>, Francesc Illas<sup>b</sup>, Sergey V. Levchenko<sup>c</sup>, Aliaksei Mazheika<sup>d</sup>, Benjamin Regler, Matthias Rupp<sup>e</sup>, Christopher Sutton, Jilles Vreeken<sup>f</sup>, Yang-Gang Wang<sup>g</sup>, Luca M. Ghiringhelli, and Matthias Scheffler

Most of artificial-intelligence (AI) applications to materials science are dedicated to the learning of predictive global models, i.e., maps between a *descriptor* (a set of numerical parameters that univocally identify each material), and a target property of interest. Considering an AI model as a tool that mimics human reasoning, one can identify alternative tasks besides finding a global law that reproduces some observations. For instance, a human researcher may be interested in checking whether the data should rather be separated in different classes, each with its own optimal model, or whether some control variables can form a suitable descriptor explaining an observed quantity, prior to guessing any explicit functional relationship. Here, we present two AI approaches for addressing these two challenges.

The first approach is sub-group discovery (SGD), which works by identifying data subsets that *i*) optimize a functional of the observed *distribution* of a target property (e.g., the shift in average value of the target property between the full dataset and the subset) and *ii*) are described by means of conjunctions of (in)equalities statements about a set of given features (e.g., “the volume of the unit cell is larger than ...” AND “the average electronegativity of the atomic constituent is smaller than ...”). To demonstrate the versatility of this tool, we show two conceptually very different applications. First, we address the challenge of predicting whether a metal-oxide surface is or not a good CO<sub>2</sub>-conversion catalyst. Starting from *ab initio* data, we applied SGD to find whether *indicators* of CO<sub>2</sub> activation (a prerequisite to its catalytic conversion) exist and we found that a subgroup having an *elongation of a CO bond length* contains all the experimentally tested materials that are good CO<sub>2</sub> conversion catalysts. The second application is an analysis of the prediction errors of a set of machine-learning (ML) models, trained on an *ab initio* dataset of transparent semiconducting oxides (see also TH 8). The goal is to look for the *domain of applicability* (DA) of the ML models, i.e., regions of the input space that yield particularly small prediction errors. As an example, we find that for one of the considered ML models, the average error in the DA is less than half than for the whole dataset and falls below the so-called chemical accuracy. Therefore, sampling within the DA is expected to be as reliable as the parent *ab initio* model.

Our newly developed second approach uses *information theory* to score the *content of information* that a subset of candidate features has on a target property. The algorithm identifies the optimal subset size and its composition and is applied to the identification of the optimal set of features for predicting the elastic properties of inorganic crystalline compounds.

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## Oxidation of Elemental and Alloyed Transition-Metal Surfaces at Realistic Temperature and Pressure Conditions

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Oxidation of metal surfaces plays a crucial role when functional materials are in contact with an oxidizing environment. The thermodynamics of oxidation and its dependence on the metal species and surface orientation are still not fully understood. In particular, the dependence of distribution of alloy components on temperature and partial oxygen pressure and the effect of adsorbed oxygen (including subsurface oxygen) on this distribution, remain unexplored.

Using the all-electron FHI-aims code<sup>1</sup> we present a systematic DFT study (comparing various xc approximations) for the adsorption of atomic oxygen at (111) and (110) surfaces of Ag, Au, Pt, Rh, Pd and Cu elemental metals and the Pd or Pt alloyed Cu(111) and Cu(110) surfaces. The effects of configurational entropy are evaluated by combining a cluster expansion (CE) model with Monte Carlo sampling and the Wang-Landau algorithm,<sup>2,3</sup> as implemented in our python package CELL.<sup>4</sup> The package can treat a wide variety of systems, including one-, two-, and three-dimensional materials, as well as multi-component and multi-lattice systems. Most notably, it is capable of dealing with complex materials containing several (more than 30) atoms in their parent lattice. CELL uses state-of-the-art techniques for the construction of training data sets, model selection, and finite-temperature simulations, and can be interfaced with virtually any *ab initio* code.

Our study takes into account configurational entropy for both alloy components and adsorbates simultaneously. We find that this leads to a very large number of possible compact clusters in CE, so that machine-assisted selection of the clusters is required to find a predictive model. The cluster selection is performed using compressed sensing with both  $l_0$  (penalizing the number of non-zero cluster-interactions) and  $l_1$  norms.<sup>5</sup> We show that long-range repulsive interactions between the negatively charged oxygen anions are essential to explain the stability of ordered structures of adsorbed O atoms at (110) and (111) surfaces of elemental metals. The stronger the repulsive interaction is, the higher is the temperature of order-disorder phase transitions. In general, the stability range of ordered structures at (110) surfaces is found to be larger than at (111) surfaces. The structures of adsorbed O at the Pd or Pt alloyed Cu surfaces exhibit a larger variety of patterns depending on coverage, with Pd/Pt segregating to the surface at low oxygen coverages, while at high oxygen coverages Cu segregates.

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## Stable and Metastable Crystal Structures of Transparent Semiconducting Oxides from First Principles, Experiments, and Crowd-Sourced Machine Learning

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Solid solutions of group-III sesquioxides ( $\text{Al}_x\text{Ga}_y\text{In}_z\text{O}_3$ , with  $x + y + z = 2$ ) show promise in designing new transparent *n*-type electrode or active materials for optoelectronic applications because of the possibility to tune the bandgap energies over large ranges (i.e., 3.6 to 7.5 eV) by varying the relative cation concentration. For example, alloying  $\text{Ga}_2\text{O}_3$  with  $\text{In}_2\text{O}_3$  can be useful for solar-blind photo-detector and transparent Schottky-contact applications. The computational study of these materials requires efficient sampling techniques, in order to identify the stable and metastable structures by scanning the different allowed compositions, crystal-structure prototypes, and lattice decoration. In particular, machine-learning (ML) empirical potential, fitted to accurate *ab initio* energies, can greatly speed up the exploration of the compositional and configurational space.

In this work, we present a study of the (meta)stability of the alloys  $(\text{In}_x\text{Ga}_{1-x})_2\text{O}_3$  by means of both cluster-expansion ML models trained on gradient-corrected (GGA) density - functional theory (DFT) data points in three different crystal structures and pulsed-laser deposition experiments, analysed via transmission electron microscopy. Both theory and experiments indicate that the alloy stability is determined over a broad range of temperatures by a competition among the thermodynamically stable lattices of  $\text{In}_2\text{O}_3$  (cubic phase, c) and  $\text{Ga}_2\text{O}_3$  (monoclinic phase,  $\beta$ ) and a high-temperature, high-pressure binary alloy (hexagonal phase, h). This results in a very shallow energy surface at varying composition. For the  $\beta$  and h phases, indium forms a 6-fold coordinated sub-lattice in both cases, whereas gallium forms a 4- and 5-fold sub-lattice. The presence of two lattice sites facilitates alloying even in materials that have very different ground state-structures because the preferential environment of each component is satisfied.

Furthermore, a dataset of 2 400 GGA DFT data points for the ternary sesquioxides over 6 crystal structures has been used as training set for a crowd-sourced challenge, hosted by the on-line platform Kaggle in early 2018. The contestants had to predict the formation energy and band gap for a test set of 600 data points in the same material class. About 900 teams participated in the competition. Here, we present a summary of the three top-ranked ML models, which won the competition, including an unorthodox solution (the actual absolute winner) based on a crystal-graph representation that was inspired by techniques originally developed for natural-language processing. Our analysis of the three models shows that the descriptor (the representation of the input crystal structure for the ML regression algorithm) is the key aspect determining the accuracy of the predictive model, while the actual regression algorithm plays a secondary role.

These projects demonstrate that ML empirical potentials are valid tools for accelerating the study of the compositional and configurational space of relevant materials and can help theoretical materials science to shed light on experimental observations. This work is supported by the Leibniz Center GraFOx project.

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## Nuclear Quantum Effects in Response Properties of Materials and Electron-Vibrational Coupling

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Christian Carbogno, and Mariana Rossi

The inclusion of anharmonic nuclear quantum effects (NQE) in electronic structure simulations has benefited from recent developments in the field of path integral molecular dynamics. We here address two of the main open challenges in this field, namely the calculation of nuclear quantum time-dependent response properties and electron-vibrational coupling, both in full-dimensionality and beyond the harmonic approximation. We show that our developments, which can be used in all-atom/all-electron simulations, can unravel thus-far unexplained observations regarding anharmonic hydrogen transfer dynamics and isotope-effects in electronic properties.

We have derived optimizations for techniques that approximate quantum dynamics and are able to describe vibrational spectra, heat transport, and reaction rates. These approximations are the ring-polymer instanton method<sup>1</sup> and thermostatted ring-polymer molecular dynamics<sup>2</sup> (TRPMD), which we have implemented in the i-PI code<sup>3</sup>. In particular, regarding the challenging subject of heat transfer, we extend the *ab initio* heat-flux definition proposed in Ref.<sup>4</sup> to TRPMD and centroid molecular dynamics<sup>5</sup>, in order to achieve a formulation of the Green-Kubo formalism that can be applied to situations where both nuclear quantum effects and strong anharmonicity play a role – a regime that no current theory can address and is relevant for, e.g., organic electronics. We also take advantage of an acceleration technique we have recently proposed<sup>6</sup>, tailored for weakly-bonded organic-inorganic interfaces, to study electron-vibrational coupling through the calculation of quantum thermal averages of electronic observables.

We first address porphycene, a prototype molecular switch. We can explain the controversial coexistence of Arrhenius (classical) behavior and strong isotope effect in its intramolecular hydrogen transfer from an interplay of a strong tunneling enhancement of the reaction that is, nevertheless, coupled to a low-frequency thermally activated mode<sup>7</sup>. This coupling, allied to zero-point-energy, is also responsible for an extreme broadening and red-shift of the NH-stretch band with respect to harmonic predictions, in accordance to experiments. It also causes a pronounced blue shift of this band with increasing temperature, which classical-nuclei dynamics is unable to even *qualitatively* describe<sup>8</sup>. Additionally, we explain the observed isotope-effect on the work-function changes and desorption energies of  $\text{C}_6\text{H}_{12}/\text{Rh}(111)$ <sup>9</sup>, a system of relevance for catalytic dehydrogenation processes. We show how temperature and NQE cause electronic level broadening and renormalization, which is particularly pronounced for C and H/D electronic states hybridized with the surface *d*-band. NQE also strengthen the H-Rh(111) bond leading to a work-function change that is larger for  $\text{C}_6\text{H}_{12}$  than for  $\text{C}_6\text{D}_{12}$ .

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## The Role of Temperature on Interface Level Alignment of Strong Organic Electron Acceptors Adsorbed on 3D and 2D Semiconductors

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Conventional semiconductor-based technologies are approaching their physical limits. In order to move forward, possibilities to scale down the material-size to the nanoscale are considered. For example, we investigate the combination of organic dopant molecules with traditional semiconductor devices or the use of 2D materials to replace traditional semiconductors. Specifically, we present a first-principles study of the structural and electronic properties of strong organic electron acceptors interfaced with standard (silicon) and novel (MoS<sub>2</sub>) semiconductors, emphasizing the influence of temperature effects on electronic properties, in particular energy level alignment.

We consider two cyanoquinodimethane derivatives, F4-TCNQ (F4) and F6-TCNNQ (F6), which are strong electron acceptor molecules. Regarding the structure search, we extend a genetic algorithm package that works on internal degrees of freedom<sup>1</sup> in order to include the position and orientation of rigid and *flexible* molecules with respect to the surface and to one another. Our study reveals that the bonding between the molecules and semiconductor surfaces is dominated by van der Waals interactions. The electronic level alignment is found to be strongly affected by the self-interaction error. Therefore, we perform our calculations with the range-separated hybrid HSE06 functional<sup>2</sup> including many-body van der Waals (vdW) contributions<sup>3</sup>.

At the pristine H-Si(111) surface, F4 and F6 adsorb with significantly different densely packed geometries in the first layer, despite their similar composition. In multilayered structures, F6 presents a much stronger intralayer interaction, dominated by vdW contributions, than F4. The densely packed geometries induce a large interface charge rearrangement that results in a work function increase of 1.11 and 1.76 eV for F4 and F6, respectively, in accordance with experimental results. Anharmonic nuclear fluctuations at the interface, described through *ab initio* molecular dynamics, cause a wide distribution of work function values at 300 K, well-modeled by a normal distribution with  $\sigma = 0.17$  eV, which corresponds to 6.5 times the thermal energy<sup>4</sup>.

Finally, we probe the role of temperature in the level alignment of F6 on a free-standing MoS<sub>2</sub> monolayer and graphite-supported MoS<sub>2</sub> monolayer (G-MoS<sub>2</sub>). For the latter, we consider commensurate cells containing 522 atoms. On these substrates, F6 lies flat at all coverages, forming a localized state within the band gap of MoS<sub>2</sub>. In G-MoS<sub>2</sub>, this localized state is situated only 80 meV above the Fermi energy. We thus build thermally-displaced geometries<sup>5</sup> and propose schemes to calculate the renormalization and the spread of this spatially-localized electronic energy level. With that, we can predict the possible temperature-dependent onset of charge transfer from graphite to F6-TCNNQ, creating an electric field across the MoS<sub>2</sub> film. This work is supported by the DFG CRC-951 (HIOS) project.

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## ***In Silico* Characterisation of Anharmonic Vibrational Properties of Molecular Crystals**

Nathaniel Raimbault, Marcin Krynski, and Mariana Rossi

Predictive simulations of molecular crystals present challenges ranging from the accurate description of their interatomic forces to accounting for conformational entropic contributions. These systems have been the subject of extensive studies<sup>1</sup> that most often neglect finite temperature anharmonic effects on their properties. We here present a framework that allows the calculation of anharmonic, finite-temperature, vibrational spectra and free energies from *ab initio* methods, with the help of different Gaussian process regression (GPR) schemes to dramatically reduce the computational cost.

We first focus on structure-sensitive vibrational Raman spectra, taking as examples different polymorphs of the Paracetamol and Aspirin crystals. The low-frequency region of these spectra is experimentally accessible and particularly sensitive to different polymorphic arrangements. We include full anharmonicity in this region by calculating Raman spectra with the time-correlation formalism, combining *ab initio* molecular dynamics (PBE functional with many-body dispersion corrections) and density-functional perturbation theory (DFPT), as implemented in the electronic structure code FHI-aims<sup>2</sup>. We show that lattice expansion strongly affects the collective low-frequency phonons, while van der Waals dispersion has a minute impact if lattice parameters are kept constant. We obtain better agreement with experimental lineshapes for the time-correlation formalism than the harmonic formalism, confirming the importance of anharmonic effects in the room-temperature phonons of these systems.<sup>3</sup> Therefore, in order to treat a wider range of systems encompassing larger unit cells and more flexible molecular units, we devise a machine-learning method to compute Raman intensities – the most costly part of these simulations. We build different GPR models based on atomic densities to predict the DFPT electronic polarisability and susceptibility tensors. We show that our methodology can efficiently predict the vibrational Raman spectra of crystals. In particular, with a local symmetry-adapted GPR model, we obtain sufficient accuracy when training with only a few hundred points. This model also proves to be scalable and transferrable between different polymorphic forms<sup>4</sup>.

Having established the need for anharmonic contributions to provide a quantitative prediction of phonon-related properties in these systems, we focus on the calculation of vibrational free energies at finite temperatures. We benchmark the performance of different exchange-correlation functionals in a fully anharmonic evaluation of this quantity, based on a series of thermodynamic integrations<sup>5</sup>, for hydrocarbon-based crystals. To achieve an *ab initio*-quality high-throughput framework, we propose a local GPR model to predict harmonic free energies, as well as the difference between anharmonic free energies from empirical force-fields and density-functional theory potential-energy surfaces.

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## Atomic and Electronic Structure of $\beta$ -Ga<sub>2</sub>O<sub>3</sub> Surfaces at Realistic Conditions: Theory and Experiment

Konstantin Lion<sup>a</sup>, Sergey V. Levchenko<sup>b</sup>, Matthias Scheffler, and Claudia Draxl<sup>a</sup>

The transparent conducting oxide Ga<sub>2</sub>O<sub>3</sub>, exhibiting a band gap of about 4.9 eV, is a very promising candidate for a number of applications, such as semiconducting lasers and transparent electrodes for UV optoelectronic devices and solar cells. As such, the bulk properties of its thermodynamically stable  $\beta$  phase have been extensively studied in the last two decades. The surface properties, however, playing a vital role in epitaxial growth, electrical contacts, and gas sensors, are still not well understood.

In this work, we study the stability and electronic structure of several non-polar low-index surfaces of  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> using *ab initio* atomistic thermodynamics.<sup>1</sup> We perform density-functional-theory (DFT) calculations with the hybrid exchange-correlation functional HSE,<sup>2</sup> which mixes a portion of exact exchange from Hartree-Fock theory with the exchange energy from a generalized gradient calculation. The mixing parameter is chosen as 26% exact exchange, which not only reproduces the experimental band gap but also satisfies the generalized Koopmans' theorem.<sup>3</sup>

We calculate numerous surface free energies of  $\beta$ -Ga<sub>2</sub>O<sub>3</sub>, also including vibrational contributions in the harmonic approximation for the two most stable surfaces. We find that the (100) surface is the most stable one, in agreement with previous reports,<sup>4</sup> but, quite surprisingly, it is followed closely by the ( $\bar{2}01$ ) surface. Here, the topmost surface layers are significantly flattened upon relaxation, reducing the surface energy to 40 % of the value of the unrelaxed surface. Vibrational contributions further slightly stabilize both surfaces but do not change the relative stability between them. These results explain recent experimental findings where ( $\bar{2}01$ ) facets were found to form during homoepitaxial growth on off-oriented  $\beta$ -Ga<sub>2</sub>O<sub>3</sub> (100) substrates.<sup>5</sup> For this surface the electronic structure reveals a surface state at the top of the valence band which can act as a recombination centre, and thus, potentially degrade device performance. This work is supported by the Leibniz Center GraFOx project.

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## High-Precision yet Efficient High-Throughput Search for Thermal Insulators

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David Hicks<sup>a</sup>, Stefano Curtarolo<sup>a</sup>, Matthias Scheffler, and Christian Carbogno

Identifying and designing materials with ultra-low thermal conductivities  $\kappa$  is the challenge in the development of novel, more efficient thermoelectric “waste heat recovery” devices<sup>1</sup>, which are pivotal for establishing a sustainable, green energy economy. The in-house developed *ab initio* Green-Kubo (aiGK) method<sup>2</sup> is ideally suited to guide and accelerate the search for such materials, since –in contrast to perturbative methods– it captures the strong anharmonic effects determining the properties of thermal insulators. Due to the elevated computational cost of aiGK calculations, however, a comprehensive high-throughput search of the materials space seems prohibitive, at first.

We overcome this bottleneck with a newly developed hierarchical, high-throughput framework that builds on the combination of the Atomic Simulation Environment ASE<sup>3</sup> with FireWorks<sup>4</sup>, the AFLOW Library of Crystallographic Prototypes<sup>5</sup>, Phonopy<sup>6</sup>, and FHI-aims<sup>7</sup>. In this workflow, we systematically explore the materials space using an accelerated geometry relaxation scheme that ensures the preservation of the target crystalline prototype<sup>8</sup>. The degree of anharmonicity is then quantified via a novel analytical descriptor based on the statistical comparison of the energy and forces obtained in the harmonic approximation to the fully anharmonic ones obtained via *ab initio* MD and/or statistical sampling. This allows us to disregard materials with large  $\kappa$  in the early stage of the high-throughput search and thus efficiently focus on actual thermal insulators instead. For these materials with strong anharmonicity and thus low  $\kappa$  the thermal conductivity is then computed with the aiGK method.

The performed search that covers thousands of materials reveals that strongly anharmonic compounds with low  $\kappa$  are much more common than originally thought. We shed light on the different qualitative mechanisms determining the low thermal conductivity using representative examples from multiple material classes. For instance, we discuss the role of strong ionic interactions in Cu halides (e.g. CuCl, CuI); the influence of van-der-Waals interactions in layered chalcogenides like SnSe; and the role of highly mobile species in inorganic halide perovskites such as CsPbBr<sub>3</sub> and CsPbI<sub>3</sub>. Eventually, we discuss how the developed approach can be further accelerated and guided by data-mining the millions of performed first-principles calculations with artificial intelligence and active learning techniques such as SISSO<sup>9</sup>. This work is supported by the ERC TEC1p project.

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## Department of Chemical Physics

### Poster List

- CP 1 **Influence of the Supporting Metal on the Atomic Structure of Ultrathin Germania Films**  
Adrian L. Lewandowski, Leonard Gura, Patrick Marschalik, Kristen M. Burson, Wolf-Dieter Schneider, Markus Heyde, and Hans-Joachim Freund
- CP 2 **Design of a Variable-Temperature High-Speed Scanning Tunneling Microscope**  
Zechao Yang, Patrick Marschalik, Leonard Gura, William Kirstädter, Jens Hartmann, Heinz Junkes, Gero Thielsch, Burkhard Kell, Kristen M. Burson, Markus Heyde, and Hans-Joachim Freund
- CP 3 **Surface Action Spectroscopy with Inert Messengers – Recent Results**  
Zili Wu, Yun Liu, Agata Plucienik, Matthias Naschitzki, Walter Wachsmann, Sandy Gewinner, Wieland Schöllkopf, Helmut Kühlenbeck, and Hans-Joachim Freund
- CP 4 **Iron Sulfide Layers on Au(111)**  
Earl M. Davis, Giulia Berti, Vedran Vonk, Andreas Stierle, Helmut Kühlenbeck, and Hans-Joachim Freund
- CP 5 **Effect of Confinement on Chemical Reactions: Water Formation Under a Silica Bilayer**  
Mauricio Prieto, Daniel Gottlob, Liviu Tănase, Dietrich Menzel, Thomas Schmidt, and Hans-Joachim Freund
- CP 6 **Water Adsorption on Magnetite Fe<sub>3</sub>O<sub>4</sub> surfaces**  
Eman Zaki, Francesca Mirabella, Shamil Shaikhutdinov, and Hans-Joachim Freund
- CP 7 **Interaction of Hydrogen with Ceria Surfaces**  
Kristin Werner, Shamil Shaikhutdinov, and Hans-Joachim Freund

## Influence of the Supporting Metal on the Atomic Structure of Ultrathin Germania Films

Adrian L. Lewandowski, Leonard Gura, Patrick Marschalik, Kristen M. Burson,  
Wolf-Dieter Schneider, Markus Heyde, and Hans-Joachim Freund

The discovery of the new two-dimensional (2D) thin film material silica has initiated an intensive research to clarify under which conditions crystalline or amorphous film phases are created on various metal supports. Early investigations on ultrathin silica films on Mo(112), Ru(0001), and Pt(111) have shown that metals with high oxygen adsorption energies favour the formation of crystalline monolayer films, whereas noble metals favour primarily vitreous silica bilayer films.<sup>1</sup>

Here we present an atomic scale STM study on another glass forming material, ultrathin germania films on Ru(0001), Pt(111), and Au(111). These metal supports exhibit different oxygen affinities and different lattice constants.

Specifically, on Ru(0001), stable hexagonal monolayer films are formed, characterised by strong GeO-Ru-bonds. This characteristic avoids the formation of atomically flat decoupled germania bilayer films.<sup>2</sup>

On Pt(111) monolayer and bilayer germania films are observed, where, depending on the preparation conditions, the structural changes from crystalline to mixed crystalline and amorphous to purely amorphous phases can be tuned in a controlled manner.<sup>3</sup>

On the reconstructed Au(111) surface only the amorphous germania bilayer film phase is detected. Moreover, it is observed that the soliton walls of the herringbone reconstruction, visible in the open space between germania film islands, penetrate below the germania bilayer film patches indicates a very weak film substrate interaction.

The orientation of adjacent rings, the ring-size distribution, and the ring-triplets analysis for each phase on each metal support, together with density functional theory (DFT) calculations performed by the group of Gianfranco Pacchioni, suggest that the film structure is clearly affected by the film-support interaction in the following way: the smallest film adhesion energy is found on Au(111) allowing an amorphous film growth, followed by a slightly larger adhesion energy on Pt(111) leading to controllable crystalline, amorphous, or intermediate germania film phases, and, finally, the largest calculated adhesion energy on Ru(0001) which limits crystalline germania film growth. A comparison of the two new 2D materials, ultrathin silica and germania films, highlights similarities and differences between these two glass formers.

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## Design of a Variable-Temperature High-Speed Scanning Tunneling Microscope

Zechao Yang, Patrick Marschalik, Leonard Gura, William Kirstädter, Jens Hartmann, Heinz Junkes, Gero Thielsch, Burkhard Kell, Kristen M. Burson, Markus Heyde, and Hans-Joachim Freund

For the first time the atomic structure of both the crystalline and the vitreous phase on metal supported thin silica films has been revealed by scanning probe microscopy (SPM).<sup>1,2</sup> Later, also atomic rearrangements in this film system were observed by transmission electron microscopy (TEM) under the impact of an electron beam.<sup>3</sup> Our new study aims to resolve the dynamic processes between the vitreous and the crystalline phases as a function of temperature in real space. For this purpose, we developed a high-speed scanning tunneling microscope (STM) and a sample holder with an integrated heating system.

An e-beam heater is integrated in the sample holder. This enables sample preparations up to 1500 K, and in-situ heating during LEED and STM measurements. For counter cooling the piezo components during high-temperature STM measurements, the STM body is attached to a continuous flow cryostat with a temperature range from 4 to 500 K.

The microscope body is compact, highly symmetric, and damped by hanging springs and eddy current, which results in high vibrational stability during scanning. The STM tip is mounted on a hybrid tube scanner. A commercial SPM control unit (Nanonis control system) controls a large tube piezo for slow scans of large areas. A small tube piezo, with a high resonance frequency, is attached to it. The high resonance frequency allows for high-speed scanning. A custom-made high-speed electronics Versa Module Eurocard bus (VMEbus) technology controls the small tube piezo. In order to increase the scan rate, we use a spiral scan geometry, which consists of smooth sine and cosine signals. The signals account for tilt correction and are created with an arbitrary waveform generator. During fast scanning, the feedback of the big piezo scanner is switched on, but set to a very low loop gain. Therefore, the tip scans in a quasi-constant height mode, where the logarithm of the tunneling current signal can be regarded as roughly proportional to the surface topography. A high-speed digitizer with a maximum data sampling rate of 250 MHz is utilized for the acquisition of the current. The scan control and the data acquisition are realized within the EPICS framework [4]. Within EPICS we transfer image chunks, which are then collected, joined, and displayed with fast averaging routines or with interpolation algorithms.

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## Surface Action Spectroscopy with Inert Messengers – Recent Results

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Surface action spectroscopy with inert messengers is a method to study the vibrational structure of surfaces. This includes surface vibrations, adsorbates, deposited aggregates, defects, essentially everything that can be vibrationally excited at a surface. The basic principle is that weakly bound messengers (He, Ne, molecular hydrogen) desorb when the energy of an excited surface vibration breaks the bond between the messenger and the surface. In the present case the surface vibrations are excited with light from the FEL of the FHI.

The main areas of application are probably the spectroscopy of rough surfaces and the study of microscopic vibrations at ionic surfaces. The competing methods HREELS, IRAS and HAS all have certain weaknesses in these areas.

Spectra of different iron oxide films on Au(111) and Pt(111) reveal that the surface vibrational spectrum is a sensitive fingerprint of the surface structure. The  $\text{Fe}_3\text{O}_4(111)$  surface vibrational spectrum was calculated by Joachim Paier and Xiaoke Li and the comparison with the experimental spectrum shows that surface is (as expected) tet1-terminated. This means that the method can be used for surface structure determination, provided that reference data are available, either from calculations or from reference experiments.

One relevant parameter for the performance of the method is the binding energy of the messenger to the surface. Breaking the bond means that the vibrationally excited state has to provide the required energy which is only possible for weakly bound messengers. This was confirmed by experiments with difference messengers. Another result was that molecular hydrogen performs much better than neon despite the similar binding energies. We tentatively trace this back to the larger number of vibrational states in the mol. hydrogen/surface system which has the consequence that more vibrational coupling channels are available for the energy transfer to the bond of the messenger to the substrate.

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## Iron Sulfide Layers on Au(111)

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Well-ordered iron sulfide layers with a thickness in the range of 10 nm on Au(111) were studied with respect to their structure and their chemical activity towards carbon dioxide. XRD data reveal that the films have a NiAs type 'bulk' structure with Fe vacancies. Indications for an ordering of the vacancies could not be found with XRD. The presence of vacancies also at the surface is indicated by XPS data, which additionally show that the film is sulfur-terminated. The surface exhibits a (2x2) reconstruction which was studied by IV-LEED, but a conclusive result could not yet be obtained.

The film turned out to be unreactive with respect to CO<sub>2</sub> in that only molecular adsorption, but no reaction was observed. In view of the sulfur-termination of the film it was attempted to increase the iron content by deposition of additional iron. However, the iron disappeared during the annealing step which was required to order the layer, probably by diffusion into the gold substrate. Further attempts involved the introduction of a FeO(111) layer between the sulfide and the substrate with the aim of hindering the iron diffusion into the substrate, and the reduction of the film by atomic hydrogen. The FeO(111) layer did work to some extent in that it was actually possible to increase the iron content of the layer, but gold from the substrate became visible in the XPS data after some time of preparation. The gold and the additional iron together could activate the CO<sub>2</sub> at the surface such that it decomposed into carbon and oxygen, which was not possible just by additional gold (prepared by annealing) or additional iron (prepared via reduction with atomic hydrogen).

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## Effect of Confinement On Chemical Reactions: Water Formation Under a Silica Bilayer

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The space between a physisorbed silica bilayer and its Ru(0001) support can be used as a model system to perform chemical reactions with the aim of addressing the effect of physical confinement on the reaction mechanism<sup>1</sup>. Using LEEM, LEED and local XPS, water formation from adsorbed O and H<sub>2</sub> on a Ru(0001) surface covered with (crystalline or vitreous) SiO<sub>2</sub> bilayers were investigated and compared to the same reaction on bare Ru(0001). We found that the reaction is characterized by moving reaction fronts both in the presence and in the absence of the silica film. The reason for this might be related to the requirement of site release by O adatoms for further H<sub>2</sub>-dissociative adsorption. Apparent activation energies ( $E_a^{app}$ ) of 0.59 eV and 0.27 eV are found for the front movement on bare Ru and under cover, respectively<sup>1</sup>. In the covered case, the activation energy obtained is roughly the same, regardless of the structure of the silica film (crystalline vs. vitreous). However, the Arrhenius plot present different pre-exponential factors, with the value for the vitreous layer being twice that of the crystalline phase. We suggest that the smaller activation energy but higher reaction temperature for the reaction on the SiO<sub>2</sub> BL covered Ru(0001) surface is due to a change of the rate-determining step. One important point is the involved diffusion. For instance, even though H<sub>2</sub> molecules can permeate the 6-member rings of the silica without a relatively low activation energy, the formed H<sub>2</sub>O molecules can hardly leave the confined space through rings of the same size, but easily through 7-member or larger ring sizes. These larger ring sizes can be found in the crystalline film only at the domain boundaries, whereas everywhere in the vitreous layer.

In addition to the temperature dependence of the reaction rate, we confirmed that the reaction front velocity exhibits a square-root like dependence with the hydrogen pressure  $p_{H_2}$ . Moreover, the width of the reaction front has shown to be dependent on both  $p_{H_2}$  and T. Basing on these experimental results we developed a kinetic model described by rate equations, involving the processes of adsorption, diffusion, reaction, and desorption. Within the collaboration with the theory group of Denis Usvyat (HU zu Berlin) and with Mark Schlutow (FU Berlin) we found that the original transition states for the water reaction are not significantly affected by the silica cover. Instead, the observed difference can be explained by a diffusion effect: the silica cover might hinder the H<sub>2</sub>O desorption and therefore increases the local concentration of adsorbed H<sub>2</sub>O molecules nearby the reaction front. This increased H<sub>2</sub>O concentration, in turn, can hinder the H diffusion between the O poor area, where it is dissociatively adsorbed, and the O rich area, where the H<sub>2</sub>O is formed. The rate limiting step would be now this diffusion.

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## Water Adsorption on Magnetite Fe<sub>3</sub>O<sub>4</sub> surfaces

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Well-ordered magnetite Fe<sub>3</sub>O<sub>4</sub>(111) and (001) thin films were grown on Pt(111) and (001) substrates, respectively. The films were characterized by LEED, AES, as well as by TPD and IRAS of CO as a probe molecule to prove the surface termination which is quite sensitive to the sample preparation. The Fe<sub>3</sub>O<sub>4</sub>(111) surface is terminated by a  $\frac{1}{4}$  monolayer of tetrahedrally coordinated Fe<sup>3+</sup> ions on top of a close-packed oxygen layer. The Fe<sub>3</sub>O<sub>4</sub>(001) surface terminated with an intact layer of oxygen and Fe<sub>oct</sub> atoms. However, a rearrangement of the subsurface cations distorts the structure leading to a  $(\sqrt{2}\times\sqrt{2})R45^\circ$  superstructure. We monitored adsorption of water at different temperatures as a function of coverage using infrared reflection-absorption spectroscopy, temperature programmed desorption, and single crystal adsorption calorimetry.

The results showed that water molecules readily dissociate on Fe<sub>3</sub>O<sub>4</sub>(111) surface Fe<sub>tet1</sub>-O ion pairs to form "monomers", i.e., terminal Fe-OH and surface OH groups. Further water molecules adsorb on the hydroxyl covered surface non-dissociatively and form "dimers" and larger oligomers, which ultimately assemble into an ordered (2×2) hydrogen-bonded network structure with increasing coverage prior to the formation of a solid water film.<sup>1</sup>

Adsorption on the  $(\sqrt{2}\times\sqrt{2})R45^\circ$ -reconstructed surface of Fe<sub>3</sub>O<sub>4</sub>(001) is strongly influenced by the surface reconstruction, which remains intact at all coverages. Close to the completion of the first monolayer, however, the ad-layer adopts a longer-range (2×2) superstructure. The formation of ordered water ad-layer on these two surfaces was directly monitored by temperature programmed LEED. The results showed that ordered structure on the (111) facet exists over a significantly larger range of temperatures and coverages than on the (001) surface.<sup>2</sup>

In both cases, the long-range order is evidence that water-water interactions exert a significant influence on the structure already prior to the nucleation of the second layer. We conclude that the stability differences stem from the smaller unit cell on the (111) surface, and the ability of water to more easily form stable hexagonal ice like-structures on the hexagonal substrate.

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## Interaction of Hydrogen with Ceria Surfaces

Kristin Werner, Shamil Shaikhutdinov, and Hans-Joachim Freund

Ceria ( $\text{CeO}_2$ ) has recently been found to be a promising catalyst in the selective hydrogenation of alkynes to alkenes, e.g. acetylene to ethylene. This reaction occurs primarily on highly dispersed Pd-based catalysts, but rarely on oxide surfaces. The origin of the outstanding activity and selectivity observed on  $\text{CeO}_2$  remains unclear. In our model studies, we focused on one key aspect of the hydrogenation reaction – the interaction of hydrogen with the oxide.

We made use of well-defined  $\text{CeO}_2(111)$  films about 5 nm in thickness grown on Ru(0001). The films can be prepared either in stoichiometric, fully oxidized state or in partially reduced  $\text{CeO}_{2-x}(111)$  state ( $x \sim 0.1-0.2$ ) by high temperature annealing in vacuum. The samples were characterized by low energy electron diffraction (LEED), x-ray photoelectron spectroscopy (XPS) and infrared reflection absorption spectroscopy (IRAS). In addition, we performed nuclear reaction analysis (NRA) experiments in University of Tokyo.

We found that hydrogen only dissociates on  $\text{CeO}_2(111)$  at  $\text{H}_2$  ( $\text{D}_2$ ) pressures in the mbar range and elevated temperatures. Hydrogen depth profiling with NRA indicates that H species stay on the surface of stoichiometric  $\text{CeO}_2(111)$  films, whereas H incorporates as a rather volatile species into the volume of partially reduced  $\text{CeO}_{2-x}(111)$  thin films. Further studies clearly showed that hydrogen may form hydroxyls at the surface and hydride (Ce-H) species below the surface. However, hydride formation is only observed when oxygen vacancies are present in the oxide. Their formation is accompanied by the transfer of an electron from a  $\text{Ce}^{3+}$  species to hydrogen, which results in the formation of  $\text{Ce}^{4+}$ , and thus in oxidation of ceria.

Complementary DFT calculations performed by the group of Prof. J. Sauer (HU Berlin) demonstrate that oxygen vacancies facilitate H incorporation below the surface and that they are the key to the stabilization of hydridic H species in the volume of reduced ceria.

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#### Doctoral Theses

Lamoth, M.: Investigations on supported nano-Silver Catalysts: Correlation of Particle Size, Silver – Oxygen Interaction and Reactivity. Technische Universität Berlin 2019.

Wernbacher, A.M.: Charge Transport in Vanadia Oxidation Catalysts. Technische Universität Berlin 2019.

## Invited talks by members of the Department of Inorganic Chemistry

Bluhm, Hendrik: Aqueous Interfaces Investigated Under Ambient Conditions by XPS. (OPSCAT 2019, Operando Surface Catalysis meeting, Oslo, Norway, Jan 2019).

Bluhm, Hendrik: Aqueous Interfaces Investigated Under Ambient Conditions by XPS. (Seminar Talk, Department of Molecular Physics, Fritz Haber Institute, Berlin, Germany, Feb 2019).

Bluhm, Hendrik: Electrical Potential Changes at Liquid/Solid Interfaces Measured by Ambient Pressure XPS. (257th ACS National Meeting & Exposition, Chemistry for new Frontiers, Orlando, FL, USA, Mar 2019).

Bluhm, Hendrik: Investigation of Aqueous Interfaces Using Ambient Pressure XPS. (2019 MRS Spring Meeting & Exhibit, Phoenix, AZ, USA, Apr 2019).

Bluhm, Hendrik: Heterogeneous Chemistry of Aqueous Interfaces. (Hamburg Photon Science Colloquium (HPSC), Hamburg, Germany, May 2019).

Bluhm, Hendrik: Heterogeneous Chemistry of Aqueous Interfaces. (Seminar, TU Wien, Vienna University of Technology, Vienna, Austria, Jun 2019).

Bluhm, Hendrik: Surface Chemistry of Ice Investigated by Photoemission Spectroscopy. (Seminar, Institute of Physical Chemistry, University of Innsbruck, Innsbruck, Austria, Jun 2019).

Carbonio, Emilia: Is Oxygen on Silver in Ethylene Epoxidation a Well Understood System? (Second NFFA Europe Science Workshop, Milan, Italy, Feb 2019).

Farra, Ramzi: In-Situ TEM and SEM. (Autumn School on Electron Microscopy in Materials Science, Berlin, Germany, Oct 2017).

Farra, Ramzi: Operando TEM/ESEM. (Lecture Series Winter Semester 2017 / 2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Nov 2017).

Frei, Elias: Small Scale Methanol Synthesis – from Pure CO<sub>2</sub> to Dynamic Operation. (35th Annual World Methanol Conference, Berlin, Germany, Sep 2017).

Frei, Elias: Thermal Desorption Spectroscopy on Powders. (Lecture Series Winter Semester 2017 / 2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Jan 2018).

Frei, Elias: How Surface and Structure Dynamic Influence Catalysis. (RC Collaboration Forum on Catalysis and Inorganic Materials, Ludwigshafen, Germany, Mar 2018).

Hermann, Klaus: Relative Fragment Forces and Torques in Molecular Systems. (CRF Seminar, Hong Kong Baptist University, Hong Kong, China, Sep 2017).

Hermann, Klaus: Mechanical Analysis of Rotor Molecules: Theoretical Concepts and Applications. (International Symposium on Nanomachines: Powering Molecules, Hong Kong, China, Nov 2017).

Hermann, Klaus: Fragment Motion in Motor Molecules: Basic Concepts and Application to Intra-molecular Rotations. (Seminar, Supercomputer Professional, Institute for Materials Research, Tohoku University Center for Computational Materials Science, Sendai, Japan, Nov 2018).

Hermann, Klaus: Compact Cubic Nanoparticles  $Me_N(n,m)$ : Structural Properties. (CRF Seminar, Hong Kong Baptist University, Hong Kong, China, Feb 2019).

Hermann, Klaus: Graphene and Other Overlayers at Surfaces: Interference Lattices and Moiré Patterns. (Faculty of Physics and Applied Informatics, University of Lodz, Łódź, Poland, May 2019).

Hetaba, Walid: Inelastic Scattering in the TEM. (Autumn School on Electron Microscopy in Materials Science, Berlin, Germany, Oct 2017).

Hetaba, Walid: Analytical Electron Microscopy. (Lecture Series Winter Semester 2017 / 2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Nov 2017).

Jang, Woosun: Nature-Inspired Algorithms for Surface Structure Predictions. (NANO KOREA 2019 Symposium, The 17th International Nanotech Symposium & Exhibition, Ilsan, South Korea, Jul 2019).

Jones, Travis: Simulating Spectra. (Lecture Series Winter Semester 2017 / 2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Jan 2018).

Knop-Gericke, Axel: Ambient Pressure X-Ray Photoelectron Spectroscopy: A Tool to Study Functional Surfaces Under Reaction Conditions. (Workshop on X-Ray and Electron based Analysis, Berlin, Germany, Sep 2017).

Knop-Gericke, Axel: Ambient Pressure X-Ray Photoelectron Spectroscopy: A Tool to Study the Electronic Structure of Active Iridium Oxides Anodes During the Oxygen Evolution Reaction. (Satellite Workshop, COEX - Combining electrons with X-rays for integrated in-operando experiments, Trieste, Italy, Sep 2017).

Knop-Gericke, Axel: The Electronic Structure of Iridium Oxides Anodes Used in the Oxygen Evolution Reaction. (8th International Symposium on Surface Science, ISSS-8, Tsukuba, Japan, Sep 2017).

Knop-Gericke, Axel: Functional Interfaces Studied by Ambient Pressure X-Ray Photoelectron Spectroscopy. (PETRA IV Workshop, Imaging of Surfaces, Interfaces and Nanoobjects: Local Structure, Defects and Composition, Hamburg, Germany, Oct 2017).

Knop-Gericke, Axel: New Insights in the Ethylene Epoxidation Over Ag. (FHI-Workshop on Current Research Topics at the FHI, Potsdam, Germany, May 2018).

Knop-Gericke, Axel: Ambient Pressure X-Ray Photoelectron Spectroscopy Studies of Catalytically Active Interfaces Using Electron Transparent Graphene Membranes. (AVS 65th International Symposium & Exhibition, Long Beach, CA, USA, Oct 2018).

Knop-Gericke, Axel: The Investigation of Catalytically Active Interfaces by Ambient Pressure X-ray Photoelectron Spectroscopy. (FHI-JST Joint Symposium, Current Topics and Challenges for Innovative Catalysts, Berlin, Germany, Oct 2018).

Knop-Gericke, Axel: Ambient Pressure X-Ray Photoelectron Spectroscopy of Active Solid-Gas Phase and Solid-Liquid Interfaces. (Symposium Catalysis under Realistic Conditions, Eindhoven, Netherlands, Nov 2018).

Knop-Gericke, Axel: Ambient Pressure X-Ray Photoelectron Spectroscopy of Catalytically Active Solid-Gas-Phase and Solid-Liquid Interfaces: Status and New Options at EMIL. (10th Joint BER II and BESSY II User Meeting, Berlin, Germany, Dec 2018).

Knop-Gericke, Axel: Ambient Pressure X-ray Photoelectrospectroscopy in Catalysis. (HZB Photon School 2019, Berlin, Germany, Mar 2019).

Knop-Gericke, Axel: In Situ X-Ray Spectroscopy on the Development of Iridium Nanoparticles in Confined Electrolyte. (Workshop, Helmholtz-Zentrum Berlin für Materialien und Energie, Berlin, Germany, Mar 2019).

Knop-Gericke, Axel: New Insights in the Ethylene Epoxidation Over Ag. (5th Russian-German Seminar on Catalysis, Bridging the Gap between Model and Real Catalysis, Synchrotron radiation in catalysis, Novosibirsk, Russia, Jun 2019).

Lamoth, Maximilian: Investigation on Supported Nano-Silver Catalysts: Correlation of Particle Size, Silver-Oxygen Interaction and Reactivity. (Seminar, Technische Universität Berlin, Berlin, Germany, Dec 2018).

Lamoth, Maximilian: Investigation on Supported Nano-Silver Catalysts: Correlation of Particle Size, Silver-Oxygen Interaction and Reactivity. (Special seminar, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Feb 2019).

Lunkenbein, Thomas: The Chemical Origin of Life. (Lecture Series Winter Semester 2017 / 2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Dec 2017).

Lunkenbein, Thomas: A Local View on Cu/ZnO/Al<sub>2</sub>O<sub>3</sub> Catalysts for Methanol Synthesis. (8th week of the young researcher, Chemical Energy Storage and Conversion, German-Russian Institute of Advanced Technologies (GRIAT), Kazan, Russia, Sep 2018).

Lunkenbein, Thomas: The Chemistry of Mineral Water. (Summer School 2018, Catalysis and Energy: from Synthesis to Application, Gelsenkirchen, Germany, Sep 2018).

Lunkenbein, Thomas: Real Structure Investigation of Heterogeneous Catalysts. (Seminar, Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons, ER-C, Jülich, Germany, Oct 2018).

Lunkenbein, Thomas: Heterogeneous Catalysts – The Presence of Local Structures and Their Temporal Evolution. (Lee Hsun Lecture Series, Institute of Metal Research, Chinese Academy of Sciences, Shenyang, China, Apr 2019).

Lunkenbein, Thomas: Local Structures in Heterogeneous Catalysts – From High Resolution Imaging to Operando TEM. (Seminar, Dalian Institute of Chemical Physics (DICP), Chinese Academy of Sciences, Dalian, China, Apr 2019).

Lunkenbein, Thomas: Insights Into the Structural Complexity of Heterogeneous Catalysts. (Colloquium for Physical and Theoretical Chemistry, Department of Chemistry, Technical University of Munich, Munich, Germany, May 2019).

Lunkenbein, Thomas: Operando Electron Microscopy. (FHI-Workshop on Current Research Topics at the FHI, Potsdam, Germany, May 2019).

Lunkenbein, Thomas: Structural Complexity of Solid State Catalysts. (Lecture, Lehrstuhl für Anorganische Chemie I, University of Bayreuth, Bayreuth, Germany, May 2019).

Lunkenbein, Thomas: Local Insights Into the Structural Complexity of Heterogeneous Catalysts. (GDCh-Kolloquium, GDCh - Ortsverband Chemnitz, Chemnitz, Germany, Jun 2019).

Lunkenbein, Thomas: TEM and Operando TEM Investigation of Heterogeneous Catalysts. (Lecture series, Facets of Materials Science, Faculty of Natural Sciences, Technische Universität Chemnitz, Chemnitz, Germany, Jun 2019).

Lunkenbein, Thomas: Towards Multiscale Operando Electron Microscopy for the Study of Heterogeneous Catalysts Under Working Conditions. (Germany-Japan Joint Seminar on Advanced Electron Microscopy and Its Application, Nagoya, Japan, Jun 2019).

Nerl, Hannah: Mapping Optical Properties in the Electron Microscope. (XXXIIIrd International Winterschool on Electronic Properties of Novel Materials, IWEPMN 2019, Kirchberg in Tirol, Austria, May 2019).

Schlögl, Robert: Chemical Dynamics of Selective Oxidation Catalysts. (8th World Congress on Oxidation Catalysis (WCOC 2017), Krakow, Poland, Sep 2017).

Schlögl, Robert: Dynamik macht Grenzflächen reaktiv. (GDCh-Wissenschaftsforum Chemie 2017 - Jubiläumskongress, GDCh - 150 Jahre, Berlin, Germany, Sep 2017).

Schlögl, Robert: Concepts in Heterogeneous Catalysis. (Lecture Series Winter Semester 2017 / 2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Oct 2017).

Schlögl, Robert: Die Struktur des Energieproblems. (49. Kraftwerkstechnisches Kolloquium 2017, Dresden, Germany, Oct 2017).

Schlögl, Robert: Photoelectrons From Working Catalysts. (GeCatS Infoday, Synchrotron Radiation and Neutrons for Catalysis, Materials Research and Development, Frankfurt/Main, Germany, Oct 2017).

Schlögl, Robert: Social Impact Through Collaborative Research. (Industrial Innovation Information Days 2017, Brussels, Belgium, Oct 2017).

Schlögl, Robert: Wie chemische Dynamik der Energiewende Beine macht. (GDCh-Kolloquium des Ortsverbandes Köln-Leverkusen, Köln, Germany, Oct 2017).

Schlögl, Robert: Material Dynamics in Heterogeneous Catalysis. (Seminarvortrag, BASF Catalysts, Iselin, NJ, USA, Dec 2017).

Schlögl, Robert: Methanol – A Corner Stone in Sustainable Energy Systems. (4th Methanol Technology and Policy Commercial Congress, Frankfurt/Main, Germany, Dec 2017).

Schlögl, Robert: Energiewende: Was wir wollen und was wir können. (Vortrag mit Diskussion in der Urania, Berlin, Germany, Jan 2018).

Schlögl, Robert: Keine Energiewende ohne Chemie! (Ringvorlesung, ENERGIE(W)ENDE - Denken, forschen und handeln in Jahrzehnten und Jahrhunderten, Justus-Liebig-Universität Gießen, Gießen, Germany, Jan 2018).

Schlögl, Robert: Sustainable Energy: Systems and Science. (275th Anniversary Symposium, Future of Research – Research of the Future, Friedrich-Alexander-Universität Erlangen-Nürnberg, Erlangen, Germany, Jan 2018).

Schlögl, Robert: Chemical Energy Conversion. (Vortrag anlässlich der Ehrenprofessur an der TU München, Munich, Germany, Apr 2018).

Schlögl, Robert: Erneuerbare Energien in der Mobilität: Das Potential synthetischer Kraftstoffe auf der Basis von CO<sub>2</sub>. (39. Internationales Wiener Motorensymposium, Vienna, Austria, Apr 2018).

Schlögl, Robert: Contribution of Catalysis to Future Sustainable Energy Systems. (BASF Symposium, New York, NY, USA, May 2018).

Schlögl, Robert: Fundamentals and Challenges of CCU. (International Workshop, What potential does Carbon Capture and Utilization (CCU) have?, Potsdam, Germany, May 2018).

Schlögl, Robert: Carbon Materials in Chemical Energy Conversion. (CarboCat-VIII, 8th International Symposium on Carbon for Catalysis, Porto, Portugal, Jun 2018).

Schlögl, Robert: Synthetic Fuels As Critical Element in Renewable Energy Strategies. (2nd Workshop, Perspectives on Power-to-Liquids and Power-to-Chemicals, Freiburg, Germany, Jun 2018).

Schlögl, Robert: Energy Sector Coupling in Research: An Integrated Approach. (Plenary Lecture, iSEnEC 2018, Integration of Sustainable Energy Expo & Conference, Nuremberg, Germany, Jul 2018).

Schlögl, Robert: Chemical Dynamics of Heterogeneous Catalysts. (SurfCat Summer School 2018, The Science of Sustainable Fuels and Chemicals, Gilleleje, Denmark, Aug 2018).

Schlögl, Robert: The Working State of the Oxidation State Catalyst. (256th ACS National Meeting & Exposition, Nanoscience, Nanotechnology & Beyond, Boston, MA, USA, Aug 2018).

Schlögl, Robert: Chemistry – Selected Aspects in the Energy Context. (FHI-JST Joint Symposium, Current Topics and Challenges for Innovative Catalysts, Berlin, Germany, Oct 2018).

Schlögl, Robert: The Future Role of Liquid Fuels. (50th anniversary of the ExxonMobil European Technology Center, Machelen, Belgium, Oct 2018).

Schlögl, Robert: The Role of Material Dynamics in Interfacial Catalysis. (The Royal Danish Academy of Sciences and Letters, Copenhagen, Denmark, Oct 2018).

Schlögl, Robert: Digitalization in Catalysis: Opportunities, Challenges, Next Steps. (GeCatS Infoday, Digitalization in catalysis: taking the next step, DECHEMA, Frankfurt/Main, Germany, Nov 2018).

Schlögl, Robert: Dynamics of Heterogeneous Catalysis. (Topical Joint Workshop IMPRS-SURMAT & IMPRS-RECHARGE, Max-Planck-Institut für Eisenforschung GmbH, Düsseldorf, Germany, Nov 2018).

Schlögl, Robert: Energiewende 2.0 - geht nicht ohne Chemie. (Festkolloquium, Jubiläen der Chemie in Leipzig, Leipzig, Germany, Nov 2018).

Schlögl, Robert: Katalysatorforschung-Schlüssel für innovative Energietechnologien. (Vortragsreihe Zukunft - Energie - Zukunft, Technische Universität Dresden, Dresden, Germany, Dec 2018).

Schlögl, Robert: A Systems View. (Synthetic Fuel Workshop, Royal Society, London, UK, Jan 2019).

Schlögl, Robert: In Operando XPS and Its Use in Catalysis and Electrocatalysis. (ELCoREL Workshop, Surface Electrochemistry and Spectroscopy, Prague, Czech Republic, Jan 2019).

Schlögl, Robert: Kraftstoffe aus CO<sub>2</sub>: Warum und wie? (6. Internationaler Motorenkongress 2019, Baden-Baden, Germany, Feb 2019).

Schlögl, Robert: Was tanken wir morgen? (Infotag Synthetische Kraftstoffe - Optionen für eine nachhaltige Mobilität, DECHEMA, Frankfurt/Main, Germany, Mar 2019).

Schlögl, Robert: CO<sub>2</sub> as Feedstock for Synthetic Fuels – Status and Perspectives. (17th International Conference on Carbon Dioxide Utilization - ICCDU 2019, Aachen, Germany, Jun 2019).

Schlögl, Robert: No Energy Transition Without Heterogeneous Catalysis or Material Science of Heterogeneous Catalyst. (5th Russian-German Seminar on Catalysis: Bridging the Gap Between Model and Real Catalysis, Synchrotron Radiation in Catalysis, Novosibirsk, Russia, Jun 2019).

Schlögl, Robert: Circular Economy of Carbon. (Chemistry meets Industry and Society, CIS 2019, Salerno, Italy, Aug 2019).

Schlögl, Robert: Dynamics of Catalysts for Energy Applications. (RACIRI Summer School 2019, Structure, Real-time Dynamics and Processes in Complex Systems, Svetlogorsk, Russia, Aug 2019).

Schmidt, Franz: Electron Beam Direction Dependent Light Emission. (StEM Workshop at Ringberg Castle, Kreuth, Germany, Jun 2019).

Siebeky, Uta: Praxisbericht Transformation - Publikationsprozess am FHI. (1. Fraunhofer Fachforum Bibliothek und Fachinformation, Leipzig, Germany, Oct 2017).

Siebeky, Uta: Open Access und Publikationsprozessunterstützung. (Seminar, MPI für Chemische Energiekonversion, Mülheim, Germany, Apr 2018).

Siebeky, Uta: Publikationssupport der Bibliothek des Fritz-Haber-Instituts der MPG. (Clubabend, Soroptimist International, Club Potsdam, Potsdam, Germany, Jun 2018).

Tarasov, Andrey: Operando Thermal Analysis. (Lecture Series Winter Semester 2017 / 2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Dec 2017).

Teschner, Detre: Mechanism and Kinetics on the Example of the Heterogeneous Catalytic Deacon Reaction. (5th International School-Conference on Catalysis for Young Scientists, Catalyst Design: From Molecular to Industrial Level, Moscow, Russia, May 2018).

Trunschke, Annette: Synthesis of Supported Active Phases. (Lecture Series Winter Semester 2017 / 2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Oct 2017).

Trunschke, Annette: Surface Area and Pore Size Distribution. (Lecture Series Winter Semester 2017 / 2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Nov 2017).

Trunschke, Annette: Analysis of Active Interfaces in Real Catalysts. (4th International Symposium on Chemistry for Energy Conversion and Storage, ChemEner2018, Berlin, Germany, Jan 2018).

Trunschke, Annette: Atomic-Scale Insights Into Structure and Dynamics of (Mn,Fe)O<sub>x</sub>-Promoted Rh Nanoparticles. (Faraday Discussion, Designing Nanoparticle Systems for Catalysis, London, UK, May 2018).

Trunschke, Annette: Spectroscopic Analysis of Functional Materials Under Operation. (Seminar, Institut für Optik und Atomare Physik (AG Kanngießler), Technische Universität Berlin, Berlin, Germany, Jun 2018).

Trunschke, Annette: Phase Transitions on the Surface of Alkane Oxidation Catalysts. (256th ACS National Meeting & Exposition, Nanoscience, Nanotechnology & Beyond, Boston, MA, USA, Aug 2018).

Trunschke, Annette: Selectivity in C-H Activation over Metal Oxides. (Mini Symposium, Frontiers in Catalysis and Electrocatalysis, Massachusetts Institute of Technology, Cambridge, MA, USA, Aug 2018).

Trunschke, Annette: Synthesis of Catalysts by Solvothermal Techniques. (Summer School 2018, Catalysis and Energy: from Synthesis to Application, Gelsenkirchen, Germany, Sep 2018).

Trunschke, Annette: C-H Activation in Alkanes Over Metal Oxides. (FHI-JST Joint Symposium, Current Topics and Challenges for Innovative Catalysts, Berlin, Germany, Oct 2018).

Trunschke, Annette: Concepts in Heterogeneous Catalysis. (Lecture, Berlin International Graduate School of Natural Sciences and Engineering (BIG-NSE), Technische Universität Berlin, Berlin, Germany, Oct 2018).

Trunschke, Annette: Clean Data in Oxidation Catalysis. (Irsee VIII Symposium, Kloster Irsee, Irsee, Germany, May 2019).

Trunschke, Annette: Specifics of Metadata and Design of Experiment in Heterogeneous Catalysis Research. (Shared metadata and data formats for Big-Data Driven Materials Science: A NOMAD-FAIRDI workshop, Berlin, Germany, Jul 2019).

Velasco Vélez, Juan: Semiconductors in Presence of Gases: The Relationship Between Charge Transfer and Charge Transport. (Lecture Series Winter Semester 2017 / 2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Dec 2017).

Velasco Vélez, Juan: X-Ray Spectroscopy of Chemical Energy Conversion Related Processes Under Aqueous Conditions: CO<sub>2</sub>RR on Cu and OER on IrO<sub>x</sub> Catalysts. (4th International Symposium on Chemistry for Energy Conversion and Storage, ChemEner2018, Berlin, Germany, Jan 2018).

Velasco Vélez, Juan: Graphene as Transparent Electrode for Electrochemistry and X-Ray Spectroscopy Applications in Liquid Environment. (Freigeist Workshop 2018, Carbon Nanomaterials in Aqueous Environment: From Characterization to Applications, Berlin, Germany, Feb 2018).

Velasco Vélez, Juan: Electrocatalytic Energy Conversion Related Processes Investigated by In Situ Soft X-Ray Synchrotron Radiation Spectroscopy. (Seminar, Instituto IMDEA Energía, Madrid, Spain, Apr 2018).

Velasco Vélez, Juan: Procesos Relacionados Con La Conversión Electroquímica De La Energía Investigados In Situ Mediante Espectroscopía De Rayos X. (Seminar, Department of Applied Physics, University of Granada, Granada, Spain, Apr 2018).

Velasco Vélez, Juan: Electrochemical Energy Conversion Related Processes Investigated by Operando X-Ray Spectroscopy. (Seminar, Department of Physics, Tamkang University, New Taipei, Taiwan, Jun 2018).

Velasco Vélez, Juan: 1 Bar XPS. (APXPS 2018, 5th Annual Ambient Pressure X-ray Photoelectron Spectroscopy Workshop 2018, Berlin, Germany, Dec 2018).

Velasco Vélez, Juan: Rising the Pressure Up From UHV to Bulk-Liquid in PES for Operando Electrocatalytic Investigations: The Case of OER on IrO<sub>x</sub>. (Seminar, Lawrence Berkeley National Laboratory, Berkeley, CA, USA, May 2019).

Willinger, Elena: Structure Analysis Through the Pair Distribution Function. (Autumn School on Electron Microscopy in Materials Science, Berlin, Germany, Oct 2017).

Willinger, Marc Georg: Multi-Scale Observation of Active Metal Catalysts by in Situ Scanning and Transmission Electron Microscopy. (FEMMS 2017, The 16th Frontiers of Electron Microscopy in Materials Science International Conference, Johannesburg, South Africa, Sep 2017).

Willinger, Marc Georg: Oscillatory Behavior of Active Metal Catalysts Observed by In-Situ Electron Microscopy. (Satellite Workshop, COEX - Combining electrons with X-rays for integrated in-operando experiments, Trieste, Italy, Sep 2017).

Willinger, Marc Georg: Real-Space Imaging of Dynamical Processes in Catalysis. (IMPRS Fall Block Course 2017, New Concepts in Experimental and Theoretical Surface Science, Berlin, Germany, Sep 2017).

Willinger, Marc Georg: Chemical Electron Microscopy. (19th Meeting of the Fachbeirat of the Fritz-Haber-Institut, Berlin, Germany, Nov 2017).

Willinger, Marc Georg: In-Situ Observation of Metal Catalyzed CVD Growth of Graphene. (3rd Sub-Angstrom Low-Voltage Electron Microscopy (SALVE) Symposium, Ulm, Germany, Dec 2017).

Wrabetz, Sabine: Microcalorimetry beyond Adsorption: Basics and Applications in Heterogeneous Catalysis. (Lecture Series Winter Semester 2017 / 2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Nov 2017).

Wrabetz, Sabine: Reactants-Induced Dynamic Responses of the Surface of Heterogeneous Catalysts. (ICG 2019, 4th Edition of International Conference on Catalysis and Green Chemistry, Tokyo, Japan, May 2019).

## Publications of the Department of Interface Science

### Late 2017

Ju, W., A. Bagger, G.-P. Hao, A.S. Varela, I. Sinev, V. Bon, B. Roldan Cuenya, S. Kaskel, J. Rossmeisl and P. Strasser: Understanding activity and selectivity of metal-nitrogen-doped carbon catalysts for electrochemical reduction of CO<sub>2</sub>. *Nature Communications* **8**, 944 (2017).

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Mistry, H., Y.-W. Choi, A. Bagger, F. Scholten, C.S. Bonifacio, I. Sinev, N.J. Divins, I. Zegkinoglou, H.S. Jeon, K. Kisslinger, E.A. Stach, J.C. Yang, J. Rossmeisl and B. Roldan Cuenya: Enhanced Carbon Dioxide Electroreduction to Carbon Monoxide over Defect-Rich Plasma-Activated Silver Catalysts. *Angewandte Chemie International Edition* **56** (38), 11394–11398 (2017).

Zegkinoglou, I., A. Zendegani, I. Sinev, S. Kunze, H. Mistry, H.S. Jeon, J. Zhao, M.Y. Hu, E.E. Alp, S. Piontek, M. Smialkowski, U.-P. Apfel, F. Körmann, J. Neugebauer, T. Hickel and B. Roldan Cuenya: Operando Phonon Studies of the Protonation Mechanism in Highly Active Hydrogen Evolution Reaction Pentlandite Catalysts. *Journal of the American Chemical Society* **139** (41), 14360–14363 (2017).

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Arán-Ais, R.M., D. Gao and B. Roldan Cuenya: Structure- and Electrolyte-Sensitivity in CO<sub>2</sub> Electroreduction. *Accounts of Chemical Research* **51** (11), 2906–2917 (2018).

Bergmann, A., T. Jones, E.M. Moreno, D. Teschner, P. Chernev, M. Glieth, T. Reier, H. Dau and P. Strasser: Unified structural motifs of the catalytically active state of Co(oxyhydr)oxides during the electrochemical oxygen evolution reaction. *Nature Catalysis* **1** (9), 711–719 (2018).

Bernal, M., Al. Bagger, F. Scholten, I. Sinev, A. Bergmann, M. Ahmadi, J. Rossmeisl and B. Roldan Cuenya: CO<sub>2</sub> electroreduction on copper-cobalt nanoparticles: Size and composition effect. *Nano Energy* **53**, 27–36 (2018).

Dong, Y., O. Korup, J. Gerdtz, B. Roldan Cuenya and R. Horn: Microtomography-based CFD modeling of a fixed-bed reactor with an open-cell foam monolith and experimental verification by reactor profile measurements. *Chemical Engineering Journal* **353**, 176–188 (2018).

Dutta, A., A. Kuzume, V. Kaliginedi, M. Rahaman, I. Sinev, M. Ahmadi, B. Roldan Cuenya, S. Veszteg and P. Broekmann: Probing the chemical state of tin oxide NP catalysts during CO<sub>2</sub> electroreduction: A complementary operando approach. *Nano Energy* **53**, 828–840 (2018).

Gao, D., I.T. McCrum, S. Deo, Y.-W. Choi, F. Scholten, W. Wan, J.G. Chen, M.J. Janik and B. Roldan Cuenya: Activity and Selectivity Control in CO<sub>2</sub> Electroreduction to Multicarbon Products over CuO<sub>x</sub> Catalysts via Electrolyte Design. *ACS Catalysis* **8** (11), 10012–10020 (2018).

Grosse, P., D. Gao, F. Scholten, I. Sinev, H. Mistry and B. Roldan Cuenya: Dynamic Changes in the Structure, Chemical State and Catalytic Selectivity of Cu Nanocubes during CO<sub>2</sub> electroreduction: Size and Support Effects. *Angewandte Chemie International Edition* **57** (21), 6192–6197 (2018).

Jeon, H.S., I. Sinev, F. Scholten, N.J. Divins, I. Zegkinoglou, L. Pielsticker and B. Roldan Cuenya: Operando Evolution of the Structure and Oxidation State of Size-Controlled Zn

Nanoparticles during CO<sub>2</sub> Electroreduction. *Journal of the American Chemical Society* **140** (30), 9383–9386 (2018).

Jeon, H.S., S. Kunze, F. Scholten and B. Roldan Cuenya: Prism-Shaped Cu Nanocatalysts for Electrochemical CO<sub>2</sub> Reduction to Ethylene. *ACS Catalysis* **8** (1), 531–535 (2018).

Keune, W., S. Hong, M.Y. Hu, J. Zhao, T.S. Toellner, E.E. Alp, W. Sturhahn, T.S. Rahman and B. Roldan Cuenya: Influence of interfaces on the phonon density of states of nanoscale metallic multilayers: Phonon confinement and localization. *Physical Review B* **98** (2), 024308 (2018).

Klingan, K., T. Kottakkat, Z.P. Jovanov, S. Jiang, C. Pasquini, F. Scholten, P. Kubella, A. Bergmann, B. Roldan Cuenya, C. Rot and H. Dau: Reactivity Determinants in Electrodeposited Cu Foams for Electrochemical CO<sub>2</sub> Reduction. *ChemSusChem* **11** (19), 3449–3459 (2018).

Leonard, N., W. Ju, I. Sinev, J. Steinberg, F. Luo, A.S. Varela, B. Roldan Cuenya and P. Strasser: The chemical identity, state and structure of catalytically active centers during the electrochemical CO<sub>2</sub> reduction on porous Fe–nitrogen–carbon (Fe–N–C) materials. *Chemical Science* **22** (9), 5064–5073 (2018). [OPEN ACCESS](#)

Madauf, L., I. Zegkinoglou, H.V. Muiños, Y.-W. Choi, S. Kunze, M.-Q. Zhao, C.H. Naylor, P. Ernst, E. Pollmann, O. Ochedowski, H. Lebius, A. Benyagoub, B. Ban-d`Etat, A.T.C. Johnson, F. Djurabekova, B. Roldan Cuenya and M. Schleberger: Highly active single-layer MoS<sub>2</sub> catalyst synthesized by swift heavy ion irradiation. *Nanoscale* **10** (48), 22908–22916 (2018). [OPEN ACCESS](#)

Özer, E., I. Sinev, A.M. Mingers, J. Araujo, T. Kropp, M. Mavrikakis, K.J.J. Mayrhofer, B. Roldan Cuenya and P. Strasser: Ir–Ni Bimetallic OER Catalysts Prepared by Controlled Ni Electrodeposition on Ir<sub>poly</sub> and Ir(111). *Surfaces* **1** (1), 165–186 (2018). [OPEN ACCESS](#)

Pielsticker, L., I. Zegkinoglou, N.J. Divins, H. Mistry, Y.-T. Chen, A. Kostka, J.A. Boscoboinik and B. Roldan Cuenya: Segregation Phenomena in Size-selected Bimetallic CuNi Nanoparticle Catalysts. *The Journal of Physical Chemistry B* **122** (2), 919–926 (2018).

Piontek, S., C. Andronesco, A. Zaichenko, B. Konkona, K. Junge Puring, B. Marler, H. Antoni, I. Sinev, M. Muhler, D. Mollenhauer, B. Roldan Cuenya, W. Schuhmann and U.-P. Apfel: Influence of the Fe:Ni Ratio and Reaction Temperature on the Efficiency of (Fe<sub>x</sub>Ni<sub>1-x</sub>)<sub>9</sub>S<sub>8</sub> Electrocatalysts Applied in the Hydrogen Evolution Reaction. *ACS Catalysis* **8** (2), 987–996 (2018).

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Anke, S., G. Bendt, I. Sinev, H.R. Hajiyani, H. Antoni, I. Zegkinoglou, H. Jeon, R. Pentcheva, B. Roldan Cuenya, S. Schulz and M. Muhler: Selective 2-Propanol Oxidation over Unsupported  $\text{Co}_3\text{O}_4$  Spinel Nanoparticles: Mechanistic Insights into Aerobic Oxidation of Alcohols. *ACS Catalysis* **9** (7), 5974–5985 (2019).

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Bergmann, A. and B. Roldan Cuenya: Operando Insights into Nanoparticle Transformations during Catalysis. *ACS Catalysis*, in press. 

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Scholten, F., I. Sinev, M. Bernal and B. Roldan Cuenya: Plasma-modified dendritic Cu catalyst for  $\text{CO}_2$  electroreduction. *ACS Catalysis* **9** (6), 5496–5502 (2019). 

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Todd, A.M.M., W. Schöllkopf, W.B. Colson, S. Gewinner, S.C. Gottschalk, G. von Helden, H. Junkes, G. Meijer, M. De Pas, T.J. Schultheiss, J.W. Rathke and L.M. Young: The FHI FEL Upgrade Design. In: *Proceedings of the 10th International Particle Accelerator Conference*. JACoW, Geneva, TUPRB103 (2019). ISBN 978-3-95450-208-0. 

Velasco Vélez, J., T. Jones, D. Gao, E. Carbonio, R. Arrigo, C.-J. Hsu, Y.-C. Huang, C.L. Dong, J.-M. Chen, J.-F. Lee, P. Strasser, B. Roldan Cuenya, R. Schlögl, A. Knop-Gericke and C.-H. Chuang: The

role of the copper oxidation state in the electrocatalytic reduction of CO<sub>2</sub> into valuable hydrocarbons. *ACS Sustainable Chemistry & Engineering* **7** (1), 1485–1492 (2019).

*White, G., M. Shankar, A.N. Johnson, M.L. Rivers, G. Shen, S. Veseli, K. Shroff, M. Sekoranja, T. Cobb, T. Korhonen, D.G. Hickin, H. Junkes, M.G. Konrad, R. Lange, S.M. Hartman, K.-U. Kasemir, M. Pearson, K. Vodopivec, L.B. Dalesio, M. Davidsaver, D. Zimoch and M.R. Kraimer*: The EPICS Software Framework Moves from Controls to Physics. In: Proceedings of the 10th International Particle Accelerator Conference. JACoW, Geneva, TUZZPLM3 (2019). ISBN 978-3-95450-208-0. 

*Zegkinoglou, I., L. Pielsticker, Z.-K. Han, N.J. Divins, D. Kordus, Y.-T. Chen, C. Escudero, V. Pérez-Dieste, B. Zhu, Y. Gao and B. Roldan Cuenya*: Surface Segregation in CuNi Nanoparticle Catalysts During CO<sub>2</sub> Hydrogenation: The Role of CO in the Reactant Mixture. *The Journal of Physical Chemistry C* **123** (13), 8421–8428 (2019). 

## Invited talks by members of the Department of Interface Science

Bergmann, Arno: In situ Studies to Understand (Electro)chemical Energy Conversion–XAFS and XRD on Nanocatalysts during Alcohol Oxidation and Oxygen Evolution. (13th EERA AMPEA Joint Programme Steering Committee and AMPEA-EERA workshop: Synchrotron Radiation and Neutron Scattering for Energy Materials, Berlin, Germany, Nov 2018).

Heyde, Markus: Looking Into the Atomic Structure of Glass Films. (Physikalisches Kolloquium, Institut für Physik, Carl von Ossietzky Universität Oldenburg, Oldenburg, Germany, Apr 2019).

Heyde, Markus: Looking Into the Atomic Structure of Glasses. (Joint Meeting of DGG – USTV, 93rd Annual Meeting of the German Society of Glass Technology in Conjunction with the Annual Meeting of French Union for Science and Glass Technology, Nuremberg, Germany, May 2019).

Junkes, Heinz: PVAcess on a Real-Time OS with Archiver Appliance as Metadata Store. (EPICS Collaboration Meeting, Saint-Paul-les-Durance, France, Jun 2019).

Kuhlenbeck, Helmut: Examples of Vanadium in Model Catalysis:  $V_2O_3(0001)$ ,  $V_2O_5(001)$  and Vanadium in  $TiO_2(110)$ . (Seminar, Fachbereich Physik, Osnabrück University, Osnabrück, Germany, May 2019).

Kuhlenbeck, Helmut: Surface Action Spectroscopy With Inert Gas Messengers. (Seminar, Suzhou Institute of Nano-tech and Nano-bionics (SINANO), Chinese Academy of Sciences, Suzhou, China, Jul 2019).

Kuhlenbeck, Helmut: Surface Action Spectroscopy With Inert Gas Messengers. (Seminar, Zhengzhou University, Zhengzhou, China, Jul 2019).

Roldan Cuenya, Beatriz: Operando Investigation of Structure, Chemical State, Composition and Reactivity Correlations in Nanocatalysis. (PETRA IV Workshop, Imaging of Surfaces, Interfaces and Nanoobjects: Local Structure, Defects and Composition, Hamburg, Germany, Oct 2017).

Roldan Cuenya, Beatriz: Operando Nanocatalysis. (Workshop, Scientific opportunities with electron spectroscopy and RIXS, Helmholtz-Zentrum, Berlin, Germany, Oct 2017).

Roldan Cuenya, Beatriz: In Situ and Operando Insight into  $CO_2$  Electroreduction Nanostructured Catalysts. (LightChEC Symposium 2017, From Solar Light to Chemical Energy, University of Zurich, Zurich, Switzerland, Nov 2017).

Roldan Cuenya, Beatriz: Operando Nanocatalysis. (19th Meeting of the Fachbeirat of the Fritz-Haber-Institut, Berlin, Germany, Nov 2017).

Roldan Cuenya, Beatriz: Operando Nanocatalysis. (Physik-Kolloquium, Department of Physics, Freie Universität Berlin, Berlin, Germany, Nov 2017).

Roldan Cuenya, Beatriz: Operando Nanocatalysis: Size, Shape, Composition and Chemical State Effects. (UniCat Colloquium, Unifying Concepts in Catalysis, Berlin, Germany, Nov 2017).

Roldan Cuenya, Beatriz: Operando Nanocatalysis: Size, Shape, Composition, and Chemical State Effects. (Workshop, Molecular Approaches to Heterogeneous Catalysis and Electrocatalysts, Institute for Advanced Study, Technical University of Munich, Garching, Germany, Nov 2017).

Roldan Cuenya, Beatriz: Surface Science and X-Ray Synchrotron Methods Applied to Catalysis. (Workshop, Molecular Approaches to Heterogeneous Catalysis and Electrocatalysts, Institute for Advanced Study, Technical University of Munich, Garching, Germany, Nov 2017).

Roldan Cuenya, Beatriz: Tuning CO<sub>2</sub> Electroreduction Selectivity Through Rational Catalyst and Electrolyte Design. (WREN2017, The Workshop on Renewable Energy Sources and Nanotechnology, Belém, Brazil, Dec 2017).

Roldan Cuenya, Beatriz: Tuning CO<sub>2</sub> Electroreduction Selectivity Through Rational Catalyst and Electrolyte Design. (Keynote Talk, Nature Conference, Materials Electrochemistry: Fundamentals and Applications, Shenzhen, China, Jan 2018).

Roldan Cuenya, Beatriz: Operando Nanocatalysis. (Symposium Frontiers of Science, Conacyt - Max Planck Society, Mexico City, Mexico, Feb 2018).

Roldan Cuenya, Beatriz: Catalysis at the Nanoscale. (BundeSciencia Science Café – Women in Science – The new Status Quo of the Digital Era, Berlin, Germany, Mar 2018).

Roldan Cuenya, Beatriz: Operando Nanocatalysis: Size, Shape, Composition and Chemical State Effects. (APS March Meeting 2018, American Physical Society, Los Angeles, CA, USA, Mar 2018).

Roldan Cuenya, Beatriz: Operando Nanocatalysis. (10th Meeting, From the witches cauldrons in materials science, Center of Interface Science, CIS, Goslar, Germany, Apr 2018).

Roldan Cuenya, Beatriz: Operando Nanocatalysis. (5th International School-Conference on Catalysis for Young Scientists, Catalyst Design: From Molecular to Industrial Level, Moscow, Russia, May 2018).

Roldan Cuenya, Beatriz: Tuning CO<sub>2</sub> Electroreduction Selectivity Through Rational Catalyst and Electrolyte Design. (14th International Fischer Symposium, Kloster Seeon, Seeon, Germany, May 2018).

Roldan Cuenya, Beatriz: Operando Insight Into Thermal and Electrocatalytic Conversion of CO<sub>2</sub> to Valuable Chemicals and Fuels. (Catalysis - Accelerating Catalytic Solutions to Global Grand Challenges, Gordon Research Conference, GRC, New London, NH, USA, Jun 2018).

Roldan Cuenya, Beatriz: Operando Nanocatalysis. (Colloquium, Centro Brasileiro de Pesquisas Fisicas, CBPF, Rio de Janeiro, Brazil, Jun 2018).

Roldan Cuenya, Beatriz: Operando Nanocatalysis. (VI San Luis Conference on Surfaces, Interfaces and Catalysis, Santa Fe, Argentina, Jun 2018).

Roldan Cuenya, Beatriz: Surface Science and X-Ray Synchrotron Methods Applied to Catalysis. (Humboldt Kolleg, Fundamental Surface Science applied to Catalysis, Santa Fe, Argentina, Jun 2018).

Roldan Cuenya, Beatriz: Structure and Electrolyte Sensitivity in CO<sub>2</sub> Electroreduction. (International Symposium on Electrocatalysis, Recent Advances in Electrocatalysis and Photoelectrocatalysis, Szczyrk, Poland, Aug 2018).

Roldan Cuenya, Beatriz: Operando Insight Into Electrocatalytic and Thermal Conversion of CO<sub>2</sub> to Valuable Chemicals and Fuels. (SNI2018: German Conference for Research with Synchrotron Radiation, Neutrons and Ion Beams at Large Facilities, Munich, Germany, Sep 2018).

Roldan Cuenya, Beatriz: Size, Shape, Support and Chemical State Effects in the Conversion of CO<sub>2</sub> to Valuable Chemicals and Fuels. (10th Brazilian / German Workshop on Applied Surface Science, Bad Dürkheim, Germany, Sep 2018).

Roldan Cuenya, Beatriz: Activity and Selectivity Control in CO<sub>2</sub> Electroreduction Through Catalyst and Electrolyte Design. (AIMES 2018, Americas International Meeting on Electrochemistry and Solid State Science, Cancún, Mexico, Oct 2018).

Roldan Cuenya, Beatriz: Dynamic Nanocatalysts: Environmental Effects. (AVS 65th International Symposium & Exhibition, Long Beach, CA, USA, Oct 2018).

Roldan Cuenya, Beatriz: Dynamic Nanocatalysts: Environmental Effects. (Colloquium, Max Planck Institute for Solid State Research, Stuttgart, Germany, Nov 2018).

Roldan Cuenya, Beatriz: Operando Insight Into Electrocatalytic and Thermal Conversion of CO<sub>2</sub> to Valuable Chemicals and Fuels. (13th EERA AMPEA Joint Programme Steering Committee and AMPEA-EERA workshop: Synchrotron Radiation and Neutron Scattering for Energy Materials, Berlin, Germany, Nov 2018).

Roldan Cuenya, Beatriz: Structure and Electrolyte Sensitivity in CO<sub>2</sub> Electroreduction. (Workshop on: Solar Fuel Production Based on Nanostructured Photoelectrodes and Catalysts, Braga, Portugal, Nov 2018).

Roldan Cuenya, Beatriz: Surface Science and X-Ray Synchrotron Methods Applied to Nanocatalysis. (Basic lecture program - BIG-NSE Initial Phase WS 2018, Berlin, Germany, Nov 2018).

Roldan Cuenya, Beatriz: Dynamic Nanocatalysts: Environmental Effects. (APXPS 2018, 5th Annual Ambient Pressure X-ray Photoelectron Spectroscopy Workshop 2018, Berlin, Germany, Dec 2018).

Roldan Cuenya, Beatriz: Dynamic Nanocatalysts: Environmental Effects. (Colloquium, IFIMAC – Condensed Matter Physics Center, Universidad Autonoma de Madrid (UAM), Madrid, Spain, Dec 2018).

Roldan Cuenya, Beatriz: Structure and Electrolyte Sensitivity in CO<sub>2</sub> Electroreduction. (Chemical Engineering Seminar, Institute of Chemistry and Chemical Engineering, EPFL, Lausanne, Switzerland, Mar 2019).

Roldan Cuenya, Beatriz: Structure and Electrolyte Sensitivity in CO<sub>2</sub> Electroreduction. (Highlights in Energy Research seminar series, EPFL Valais Wallis, Sion, Switzerland, Mar 2019).

Roldan Cuenya, Beatriz: In Situ and Operando Insight Into Electrocatalytic and Thermal Conversion of CO<sub>2</sub> to Valuable Chemicals and Fuels. (2019 MRS Spring Meeting & Exhibit, Phoenix, AZ, USA, Apr 2019).

Roldan Cuenya, Beatriz: Structure and Electrolyte Sensitivity in CO<sub>2</sub> Electroreduction. (International Bunsen-Discussion-Meeting, Fundamentals and Applications of (Photo)Electrolysis for Efficient Energy Storage, Taormina, Italy, Apr 2019).

Roldan Cuenya, Beatriz: Shape-Controlled Multimetallic Nanoparticle Electrocatalysts. (An Invited Symposium on Advances and Perspectives on Modern Polymer Electrolyte Fuel Cells – In Honor of Shimshon Gottesfeld, 235th ECS Meeting, Dallas, TX, USA, May 2019).

Roldan Cuenya, Beatriz: Structure, Composition, Support, and Electrolyte Effects in CO<sub>2</sub> Electroreduction. (Symposium, Renewable Fuels via Artificial Photosynthesis or Heterocatalysis, 235th ECS Meeting, Dallas, TX, USA, May 2019).

Roldan Cuenya, Beatriz: Tuning CO<sub>2</sub> Electroreduction Selectivity by Rational Catalyst and Electrolyte Design. (25th Topical Meeting of the International Society of Electrochemistry: New electrochemical processes for energy and the environment, Toledo, Spain, May 2019).

Roldan Cuenya, Beatriz: Tuning CO<sub>2</sub> Electroreduction Selectivity by Rational Catalyst and Electrolyte Design. (2nd IMPRS-RECHARGE Symposium: Catalysis Towards Greener Chemistry, Mülheim, Germany, May 2019).

Roldan Cuenya, Beatriz: Tuning CO<sub>2</sub> Electroreduction Selectivity by Rational Catalyst and Electrolyte Design. (Seminar, Department of Chemistry, University of Copenhagen, Copenhagen, Denmark, May 2019).

Roldan Cuenya, Beatriz: Tuning the Activity and Selectivity of CO<sub>2</sub> Electroreduction Catalysts Through Nanostructuring and Controlled Oxidation. (Symposium, Polyoxometallates and Nanostructured Metal Oxides in Efficient Electrocatalysis, Energy Conversion, and Charge Storage, 235th ECS Meeting, Dallas, TX, USA, May 2019).

Roldan Cuenya, Beatriz: Operando Insight Into Electrocatalytic and Thermal Conversion of CO<sub>2</sub> to Valuable Chemicals and Fuels. (International Workshop, Materials for today's energy challenges, Padova, Italy, Jun 2019).

Roldan Cuenya, Beatriz: In Situ and Operando Insight Into Electrocatalytic and Thermal Conversion of CO<sub>2</sub> to Valuable Chemicals and Fuels. (Workshop, The Theory and Practice of Catalysis, Telluride Science Research Center, Telluride, CO, USA, Jul 2019).

Roldan Cuenya, Beatriz: Tuning CO<sub>2</sub> Electroreduction Selectivity by Rational Catalyst and Electrolyte Design. (IUPAC 47th World Chemistry Congress, Symposium: Turning Solar Energy to Fuels via Artificial Photosynthesis, Paris, France, Jul 2019).

Roldan Cuenya, Beatriz: Dynamic Nanocatalysts During CO<sub>2</sub> Hydrogenation. (ACS Fall 2019 National Meeting & Exposition, San Diego, CA, USA, Aug 2019).

## Publications of the Department of Molecular Physics

### Late 2017

*Conrad, H. and M.E. Kordesch*: High Resolution Electron Energy Loss Spectroscopy, Applications. In: Encyclopedia of Spectroscopy and Spectrometry. 3. Ed. (Eds.): J.C. Lindon, G.E. Tranter, and D.W. Koppenaal. Elsevier, Amsterdam, 47–57 (2017). ISBN 978-0-12-803224-4.

*Friedrich, B. and D. Hoffmann*: Clara Immerwahr: A life in the shadow of Fritz Haber. In: One Hundred Years of Chemical Warfare: Research, Deployment, Consequences. (Eds.): B. Friedrich, D. Hoffmann, J. Renn, F. Schmaltz, and M. Wolf. Springer, Cham, 45–67 (2017). ISBN 978-3-319-51663-9. [OPEN ACCESS](#)

*Friedrich, B. and J. James*: From Berlin-Dahlem to the Fronts of World War I: The Role of Fritz Haber and his Kaiser Wilhelm Institute in German Chemical Warfare. In: One Hundred Years of Chemical Warfare: Research, Deployment, Consequences. (Eds.): B. Friedrich, D. Hoffmann, J. Renn, F. Schmaltz, and M. Wolf. Springer, Cham, 25–44 (2017). ISBN 978-3-319-51663-9. [OPEN ACCESS](#)

*Friedrich, B.*: Not Just a Branch, but the Blossom of the Tree of Knowledge: The Rise – and Blossoming – of Physical and Theoretical Chemistry. *Chemicke Listy* **111** (10), 654–662 (2017).

*Friedrich, B., D. Hoffmann, J. Renn, F. Schmaltz and M. Wolf (Eds.)*: One Hundred Years of Chemical Warfare: Research, Deployment, Consequences. Springer, Cham (2017). ISBN 978-3-319-51663-9. [OPEN ACCESS](#)

*Friedrich, B.*: The Exacting Task of Bringing Molecules to Attention. *Scientia* (115), 26–31 (2017).

*Hans, A., C. Ozga, R. Seidel, P. Schmidt, T. Ueltzhöffer, X. Holzapfel, M.N. Pohl, P. Wenzel, P. Reiß, I. Unger, E.F. Aziz, A. Ehresmann, P. Slaviček, B. Winter and A. Knie*: Soft X-ray induced ultraviolet fluorescence emission from bulk and interface of a liquid water microjet. *Journal of Physics: Conference Series* **875** (4), 042008 (2017). [OPEN ACCESS](#)

*Hoffmann, W., G. von Helden and K. Pagel*: Ion mobility-mass spectrometry and orthogonal gas-phase techniques to study amyloid formation and inhibition. *Current Opinion in Structural Biology* **46**, 7–15 (2017).

*Hoffmann, W. and K. Pagel*: Von normalen Proteinen zu unlöslichen Ablagerungen. *Nachrichten aus der Chemie* **65** (9), 874–878 (2017).

*Mucha, E., A.I. Gonzalez Florez, M. Marianski, D. Thomas, W. Hoffmann, W.B. Struwe, H.S. Hahm, S. Gewinner, W. Schöllkopf, P.H. Seeberger, G. von Helden and K. Pagel*: Glycan Fingerprinting via Cold-Ion Infrared Spectroscopy. *Angewandte Chemie International Edition* **56** (37), 11248–11251 (2017).

*Munkerup, K., D. Romanov, T. Bohinski, A.B. Stephansen, R.J. Levis and T.I. Sølling*: Conserving Coherence and Storing Energy during Internal Conversion: Photoinduced Dynamics of cis- and trans-Azobenzene Radical Cations. *The Journal of Physical Chemistry A* **121** (45), 8642–8651 (2017).

*Perrine, K.A., K.M. Parry, A.C. Stern, M.H.C.V. Spyk, M.J. Makowski, J.A. Freitas, B. Winter, D.J. Tobias and J.C. Hemminger*: Specific cation effects at aqueous solution-vapor interfaces: Surfactant-

like behavior of  $\text{Li}^+$  revealed by experiments and simulations. Proceedings of the National Academy of Sciences of the USA **114** (51), 13363–13368 (2017).

*Schön, D., R. Golnak, M.F. Tesch, B. Winter, J. Velasco Vélez, E.F. Aziz and J. Xiao:* Bulk-Sensitive Detection of the Total Ion Yield for X-ray Absorption Spectroscopy in Liquid Cells. The Journal of Physical Chemistry Letters **8** (20), 5136–5140 (2017).

*Seidel, R., K. Kraffert, A. Kabelitz, M.N. Pohl, R. Kraehnert, F. Emmerling and B. Winter:* Detection of the electronic structure of iron-(III)-oxo oligomers forming in aqueous solutions. Physical Chemistry Chemical Physics **19** (48), 32226–32234 (2017). 

*Seo, J., S. Warnke, K. Pagel, M.T. Bowers and G. von Helden:* Infrared spectrum and structure of the homochiral serine octamer-dichloride complex. Nature Chemistry **9**, 1263–1268 (2017).

*Song, X., M.R. Fagiani, S. Debnath, M. Gao, S. Maeda, T. Taketsugu, S. Gewinner, W. Schöllkopf, K.R. Asmis and A. Lyalin:* Excess charge driven dissociative hydrogen adsorption on  $\text{Ti}_2\text{O}_4^-$ . Physical Chemistry Chemical Physics **19** (17), 23154–23161 (2017).

*Truppe, S., H. J. Williams, M. Hambach, L. Caldwell, N. J. Fitch, E. A. Hinds, B. E. Sauer and M. R. Tarbutt:* Molecules cooled below the Doppler limit. Nature Physics, **13** (12), 1173–1176 (2017).

*Vanbuel, J., E.M. Fernández, P. Ferrari, S. Gewinner, W. Schöllkopf, L.C. Balbás, A. Fielicke and E. Janssens:* Hydrogen Chemisorption on Singly Vanadium-Doped Aluminum Clusters. Chemistry – A European Journal **23** (62), 15638–15643 (2017).

*Williams, H. J., S. Truppe, M. Hambach, L. Caldwell, N. J. Fitch, E. A. Hinds, B. E. Sauer and M. R. Tarbutt:* Characteristics of a magneto-optical trap of molecules. New Journal of Physics, **19** (11), 113035 (2017). 

*Wu, Z., A. Plucienik, F.E. Feiten, M. Naschitzki, W. Wachsmann, S. Gewinner, W. Schöllkopf, V. Staemmler, H. Kühlenbeck and H.-J. Freund:* Vibrational action spectroscopy of solids: New surface-sensitive technique. Physical Review Letters **119** (13), 136101 (2017).

### Doctoral Theses

*Göth, M.:* Investigation of Protein-Ligand Complexes by Native Mass Spectrometry and Ion Mobility-Mass Spectrometry. Freie Universität Berlin 2017.

*Hofmann, J.:* Ion Mobility-Mass Spectrometry of Complex Carbohydrates. Freie Universität Berlin 2017.

### 2018

*Ali, H., R. Seidel, M.N. Pohl and B. Winter:* Molecular species forming at the  $\alpha\text{-Fe}_2\text{O}_3$  nanoparticle-aqueous solution interface. Chemical Science **9** (19), 4511–4523 (2018). 

*Asmis, K.R. and A. Fielicke:* Preface: Size-Selected Clusters as Model Systems for Catalysis. Topics in Catalysis **61** (1-2), 1–2 (2018).

*Asmis, K.R. and A. Fielicke (Eds.):* Size-Selected Clusters as Model Systems for Catalysis. Topics in Catalysis **61** (1-2), (2018).

*Burri, S., P.A. Niklaus, K. Grassow, N. Buchmann and A. Kahmen:* Effects of plant productivity and species richness on the drought response of soil respiration in temperate grasslands. PLoS One **13** (12), e0209031 (2018). 

Debnath, S., H. Knorke, W. Schöllkopf, S. Zhou, K.R. Asmis and H. Schwarz: Experimental Identification of the Active Site in the Heteronuclear Redox Couples  $[AlVO_x]^+/CO/N_2O$  ( $x=3, 4$ ) by Gas-Phase IR Spectroscopy. *Angewandte Chemie International Edition* **57** (25), 7448–7452 (2018).

Esser, T.K., H. Knorke, K.R. Asmis, W. Schöllkopf, Q. Yu, C. Qu, J.M. Bowman and M. Kaledin: Deconstructing Prominent Bands in the Terahertz Spectra of  $H_7O_3^+$  and  $H_9O_4^+$ : Intermolecular Modes in Eigen Clusters. *The Journal of Physical Chemistry Letters* **9** (4), 798–803 (2018).

Ferrari, P., J. Vanbuel, K. Hansen, P. Lievens, E. Janssens and A. Fielicke: Effect of radiative cooling on the size-dependent stability of small boron clusters. *Physical Review A* **98** (1), 012501 (2018).

Fischer, J., S. Fuchs, A. Slenczka, M. Karra and B. Friedrich: Microsolvation of porphine molecules in superfluid helium nanodroplets as revealed by optical line shape at the electronic origin. *The Journal of Chemical Physics* **149** (24), 244306 (2018). 

Fowler, N. and G. Meijer: What's the big DEAL and why is it so difficult to reach? Nick Fowler and Gerard Meijer on the future of Open Access in Germany. *Elephant in the Lab* (2018).



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Urner, L.H.: Oligoglycerol Detergents for Native Mass Spectrometry of Membrane Proteins. Freie Universität Berlin 2018.

#### Master's Theses

Chang, R.: Phenylalanine in the Gas Phase - The Impact of Side-Chain Fluorination. Freie Universität Berlin 2018.

Lettow, M.: Investigation of the Gas-Phase Structures of Fucosylated Glycans Employing Cold-Ion Infrared Spectroscopy. Freie Universität Berlin 2018.

#### Bachelor Thesis

Sinaga, L.R.: Optical Spectra of Molecules Embedded in Helium Nanodroplets. Freie Universität Berlin 2018.

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#### Master's Thesis

Langenhan, J.: Experimental Characterization of a New Piezoelectric Slit Valve and REMPI Spectroscopy of Cold, Chiral Indanol Molecules. Humboldt Universität Berlin 2019.

## Invited talks by members of the Department of Molecular Physics

Ali, Hebatallah: Molecular Species Forming at the  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> Nanoparticle Aqueous Solution Interface. (EM-IMM PhD Retreat, Helmholtz-Zentrum Berlin für Materialien und Energie (HZB), Joachimsthal, Germany, Apr 2018).

Ali, Hebatallah: Molecular Species Forming at the  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> Nanoparticle-Aqueous Solution Interface. (Final Meeting of the CRC 1109 & Edith Flanigen Award Ceremony 2018, Berlin, Germany, Oct 2018).

Ali, Hebatallah: Probing the Electronic Structure of Nanoparticle Aqueous Solution Interface. (Seminar, Department of Highly Sensitive X-ray Spectroscopy, Helmholtz-Zentrum Berlin für Materialien und Energie (HZB), Berlin, Germany, Nov 2018).

Debnath, Sreekanta: CO<sub>2</sub> Adsorption on Ti<sub>3</sub>O<sub>6</sub><sup>2-</sup>: A Novel Carbonate Binding Motif. (Molecular and Ionic Clusters, Gordon Research Seminar, GRS, Barga, Italy, Feb 2018).

Debnath, Sreekanta: CO<sub>2</sub> Adsorption on Ti<sub>3</sub>O<sub>6</sub><sup>2-</sup>: A Novel Carbonate Binding Motif. (117th General Assembly of the German Bunsen Society for Physical Chemistry, Hanover, Germany, May 2018).

Eibenberger, Sandra: Enantiomer-Specific Spectroscopy and Coherent Population Enrichment of Chiral Molecules. (Free-Electron Laser Workshop, Applications of IR Free-Electron Lasers: Latest Developments and Future Directions at Ringberg Castle, Kreuth, Germany, Jan 2018).

Eibenberger, Sandra: Gas-Phase Manipulation and Spectroscopy of Cold, Chiral Molecules. (FHI-Workshop on Current Research Topics at the FHI, Potsdam, Germany, May 2018).

Eibenberger, Sandra: Rotational Spectroscopy and Internal State Control of Cold Molecules. (Workshop, Searching for New Physics with Cold and Controlled Molecules, Mainz Institute for Theoretical Physics (MITP), Johannes Gutenberg University Mainz, Mainz, Germany, Nov 2018).

Eibenberger, Sandra: Spectroscopy and Control of Cold, Chiral Molecules in the Gas-Phase. (10th International Meeting on Atomic and Molecular Physics and Chemistry 2019, Madrid, Spain, Jun 2019).

Fielicke, André: Probing the Activation of Small Molecules by Clusters Using Infrared Multiple Photon Dissociation Spectroscopy. (Free-Electron Laser Workshop, Applications of IR Free-Electron Lasers: Latest Developments and Future Directions at Ringberg Castle, Kreuth, Germany, Jan 2018).

Fielicke, André: Activation of Carbon Dioxide by Anionic Transition Metal Clusters. (International Bunsen Discussion Meeting, Gas Phase Model Systems for Catalysis - GPMC 2018, Ulm, Germany, Jun 2018).

Fielicke, André: Probing the Structures of Gas-Phase Boron Clusters Using Size Selective IR Spectroscopy. (ISSPIC XIX, International Symposium on Small Particles and Inorganic Clusters, Hangzhou, China, Aug 2018).

Fielicke, André: Probing the Structures of Gas-Phase Boron Clusters Using Size Selective IR Spectroscopy. (XXVII International Materials Research Congress (IMRC 2018), Cancún, Mexico, Aug 2018).

Fielicke, André: IR Spectroscopy of Strongly Bound Clusters Using IR Free Electron Lasers. (Lecture, Instituto de Investigaciones en Materiales, National Autonomous University of Mexico (UNAM), Mexico City, Mexico, Nov 2018).

Fielicke, André: Shedding IR Light on Small Platinum Clusters: The Chemistry of O, CO and CO<sub>2</sub>. (Seminar, Instituto de Investigaciones en Materiales, National Autonomous University of Mexico (UNAM), Mexico City, Mexico, Nov 2018).

Fielicke, André: Cluster Catalysis: Tuning Activity and Selectivity Atom by Atom. (Clusters and Nanostructures, Gordon Research Conference, GRC, Les Diablerets, Switzerland, Jun 2019).

Fielicke, André: Probing the Structures of Gas-Phase Boron Clusters Using Size Selective IR Spectroscopy. (ICBC-II, The Second International Conference on Boron Chemistry, Taiyuan, China, Jul 2019).

Fielicke, André: Shedding IR Light on Small Platinum Clusters: The Chemistry of O, CO and CO<sub>2</sub>. (Seminar, Institute of Chemistry, Chinese Academy of Sciences, Beijing, China, Jul 2019).

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Friedrich, Bretislav: Conditional Analytic Solvability of the Quantum Pendulum Eigenproblem. (256th ACS National Meeting & Exposition, Nanoscience, Nanotechnology & Beyond, Symposium in Honor of Raphael Levine, Boston, MA, USA, Aug 2018).

Friedrich, Bretislav: "Who Was Fritz Haber?" - A Biographical Summary. (Symposium on the Occasion of Fritz Haber's 150th Birthday, Berlin, Germany, Dec 2018).

Friedrich, Bretislav: Fritz Haber at 150: The Unfolding Views of and on a German Jewish Patriot. (Vortrag und Diskussion, Fritz Haber im Fokus. Eine kritische Würdigung des umstrittenen Chemikers, Zentrum für Angewandte Kulturwissenschaft und Studium Generale (ZAK), Karlsruhe, Germany, Jan 2019).

Helden, Gert von: Biomolecules on Their Way to Solvation. (6th Science Day of the Max Planck-EPFL Center for Molecular Nanoscience & Technology, Lausanne, Switzerland, Oct 2017).

Helden, Gert von: Combination of Mass Spectrometry Techniques and the Free Electron Laser. (Lecture Series Winter Semester 2017 / 2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Feb 2018).

Helden, Gert von: Coupling Mass Spectrometry with Ion Mobility Spectrometry, Helium Droplet Isolation and Infrared Spectroscopy. (European Mass Spectrometry Conference (EMSC) 2018, Saarbrücken, Germany, Mar 2018).

Helden, Gert von: Infrared Spectroscopy of Mass-to-Charge Selected Ions in Helium Nanodroplets. (101st Canadian Chemistry Conference and Exhibition, Edmonton, AB, Canada, May 2018).

Helden, Gert von: Development of IMS-MS for Structure and Complex Mixture Analysis. (John B. Fenn Distinguished Contribution in Mass Spectrometry Award Lecture, 66th ASMS Conference on Mass Spectrometry and Allied Topics, San Diego, CA, USA, Jun 2018).

Helden, Gert von: Structural Characterization of Molybdenum Oxide Nanoclusters Using MS, IMS and IR. (FHI-JST Joint Symposium, Current Topics and Challenges for Innovative Catalysts, Berlin, Germany, Oct 2018).

Helden, Gert von: Mass Spectrometry Coupled with Ion Mobility Spectrometry, Helium Droplet Isolation and Infrared Spectroscopy. (Kolloquium, Department of Chemistry, Technische Universität Kaiserslautern, Kaiserslautern, Germany, Jan 2019).

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Karra, Mallikarjun: Microsolvation of Phthalocyanine and Phthalocyanine Based Molecules in Superfluid Helium Nanodroplets. (Tulip VII Summer School on Modern Developments in Spectroscopy, Noordwijk, Netherlands, Apr 2018).

Karra, Mallikarjun: Phthalocyanine, Porphyrin and Phthalocyanine-Based Complexes in Superfluid Helium Nanodroplets. (3rd Summer School of the SPP 1807: Control of London dispersion interactions in molecular chemistry, Hamburg, Germany, Jul 2018).

Karra, Mallikarjun: Anomalous Rotational Behaviour of “Large” Rotors Solvated in Helium Nanodroplets. (4th SPP 1807 Summer School, Control of London Dispersion Interactions in Molecular Chemistry, Paderborn, Germany, Jul 2019).

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Meijer, Gerard: Open Access of Publications, a Cost-Effective Scientific Must. (Impact of Science – August Meetup, The future of open access in Germany: What's the big DEAL?, Alexander von Humboldt Institute for Internet and Society (HIIG), Berlin, Germany, Aug 2018).

Meijer, Gerard: Status Report on the Deal Negotiations With Publishers. (WIAS - PDI Open Access Tag, Weierstraß-Institut für Angewandte Analysis und Stochastik and Paul-Drude-Institut für Festkörperelektronik, Berlin, Germany, Nov 2018).

Meijer, Gerard: Where Do We Stand After the 14th Berlin Open Access Conference? (APE 2019, Academic Publishing in Europe Nr. 14, Platforms or Pipelines? Where is the Value in Scholarly Communications?, Berlin, Germany, Jan 2019).

Meijer, Gerard: Large Scale Transformation to Open Access: The Key Role of the Max Planck Society. (Wiley Society Executive Seminar, The Future is Now: Adapt, Diversify and Thrive, London, UK, Mar 2019).

Meijer, Gerard: Large Scale Transformation to Open Access: The Deal Negotiations. (SANLiC 2019 Conference, South African National Library and Information Consortium (SANLiC), Cape Town, South Africa, Jun 2019).

Meijer, Gerard: Mainly about CO, some OA, NO CRUPS. (Symposium on Chemical Dynamics, in Honor of Prof. Dr. Alec Wodtke's 60th Birthday, Göttingen, Germany, Jul 2019).

Meijer, Gerard: Open Access Transformation durch das Projekt DEAL. (Sommersitzung des Wissenschaftsrates, Giessen, Germany, Jul 2019).

Meijer, Gerard: Science with Infrared Free Electron Lasers - Sophisticated Experiments at Mature Facilities. (HFML-FELIX User Meeting 2019, FELIX Laboratory, Radboud University, Nijmegen, Netherlands, Jul 2019).

Pagel, Kevin: Gas-Phase Structural Analysis of Complex Oligosaccharides. (GDCh-Wissenschaftsforum Chemie 2017 - Jubiläumskongress: GDCh - 150 Jahre, Berlin, Germany, Sep 2017).

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Pagel, Kevin: Gas-Phase Structural Analysis of Complex Carbohydrates. (FMP Seminar, Leibniz-Forschungsinstitut für Molekulare Pharmakologie (FMP), Berlin, Germany, Mar 2018).

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Pagel, Kevin: Ion Mobility-Mass Spectrometry in Glycan Analysis - Challenges and Limitations. (Analytica Conference 2018, Munich, Germany, Apr 2018).

Pagel, Kevin: Sugars in the Gas Phase – From Structure to Reaction Mechanisms. (Berliner Chemie Symposium, Berlin, Germany, Apr 2018).

Pagel, Kevin: Gas-Phase Structural Analysis of Glycosaminoglycans. (The FUB-HUJI Glycosaminoglycan Workshop, The Hebrew University of Jerusalem, Jerusalem, Israel, May 2018).

Pagel, Kevin: Sugars in the Gas Phase – Novel Techniques to Unravel the Glycocode. (Evening Plenary Lecture, Global Analytics Conference (GACon), BASF, Ludwigshafen, Germany, May 2018).

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Pagel, Kevin: Ion Mobility Spectrometry as Separation Technique in Glycan Analysis. (Copenhagen Symposium on Separation Sciences (CSSS 2018), Copenhagen, Denmark, Jun 2018).

Pagel, Kevin: Glycan Fingerprinting Using Cold-Ion Infrared Spectroscopy. (256th ACS National Meeting & Exposition, Nanoscience, Nanotechnology & Beyond- Division of Carbohydrate Chemistry, Boston, MA, USA, Aug 2018).

Pagel, Kevin: Sugars in the Gas Phase – From Structure to Reaction Mechanism. (Gaseous Ions: Structures, Energetics and Reactions, Gordon Research Conference, GRC, Ventura, CA, USA, Feb 2019).

Pagel, Kevin: Reaction and Fragmentation Mechanisms of Oligosaccharides – Insights From Ion Mobility Spectrometry and Gas-Phase Spectroscopy. (ANAKON 2019, Fachgruppe Analytische Chemie, GDCh, Münster, Germany, Mar 2019).

Pagel, Kevin: Sugars in the Gas Phase - Novel Techniques to Unravel the Glycocode. (6th NovAliX Conference, Biophysics in Drug Discovery 2019, Cannes, France, Mar 2019).

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Pagel, Kevin: Combining Liquid Chromatography and Ion Mobility-Mass Spectrometry for the Analysis of Glycans. (HPLC 2019, 48TH International Symposium on High-Performance Liquid Phase Separations and Related Techniques, Milan, Italy, Jun 2019).

Pagel, Kevin: Sugars in the Gas Phase – From Structure to Reaction Mechanism. (AMS 2019, Advancing Mass Spectrometry for Biophysics and Structural Biology, Amherst, MA, USA, Jul 2019).

Pérez-Ríos, Jesús: The Role of Molecules in the Quest for Dark Matter. (FHI-Workshop on Current Research Topics at the FHI, Potsdam, Germany, May 2019).

Pohl, Marvin Nicolas: Intermolecular Proton and Electron Transfer in Aqueous Solution. (Photoionization and Photodetachment, Gordon Research Conference (GRC), Galveston, TX, US, Feb 2018).

Pohl, Marvin Nicolas: Intermolecular Electronic Decay Processes in Aqueous Solutions. (EM-IMM PhD Retreat, Helmholtz-Zentrum Berlin (HZB), Joachimsthal, Germany, Apr 2018).

Schöllkopf, Wieland: The Free-Electron Laser at the Fritz-Haber-Institut. (Block Course on New Concepts in Experimental and Theoretical Surface Science, IMPRS Functional Interfaces in Physics and Chemistry, Berlin, Germany, Sep 2017).

Seo, Jongcheol: Amino Acid Clusters: The Case of the Mysterious Serine Octamer. (19th Meeting of the Fachbeirat of the Fritz-Haber-Institut, Berlin, Germany, Nov 2017).

Trinter, Florian: Science at the World Best Soft X-Ray Beam Line of the Highest Brilliance Synchrotron Radiation Facility PETRA III. (Symposium on Electronic and Structural Dynamics, Tohoku University, Sendai, Japan, Jul 2018).

Trinter, Florian: Imaging the Correlated Two-Electron Wave Function of a Hydrogen Molecule. (The International Conference on Many Particle Spectroscopy of Atoms, Molecules, Clusters and Surfaces, Budapest, Hungary, Aug 2018).

Trinter, Florian: Interatomarer Stressabbau – gefilmt! (Science Slam, Feier anlässlich der 10.000. Promotion in den Naturwissenschaften, Frankfurt, Germany, Sep 2018).

Trinter, Florian: A Molecular Movie of Interatomic Coulombic Decay in NeKr. (Atomic and Molecular Physics Seminar, University of Nevada, Reno, NV, USA, Jul 2019).

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Triptow, Jenny: PhotoElectron Circular Dichroism (PECD) of Anionic Metal Complexes. (Photoionization and Photodetachment, Gordon Research Conference (GRC), Galveston, TX, US, Feb 2018).

Truppe, Stefan: Laser Cooling Molecules to Below the Doppler Limit. (2018 Conference on Cold and Controlled Molecules and Ions (CCMI), Athens, GA, USA, Mar 2018).

Truppe, Stefan: Towards a Quantum Gas of Polar Molecules. (FHI-Workshop on Current Research Topics at the FHI, Potsdam, Germany, May 2018).

Truppe, Stefan: Laser Cooling Molecules. (Department Seminar, European Laboratory for Non-Linear Spectroscopy (LENS), University of Florence, Florence, Italy, Mar 2019).

Truppe, Stefan: Laser Cooling Molecules. (Department Seminar, Institute for Theoretical Physics, TU Wien, Vienna University of Technology, Vienna, Austria, May 2019).

Winter, Bernd: Opportunities in Liquid-Jet Photoelectron Spectroscopy. (Opportunities for Soft X-ray Science at PETRA IV, DESY Hamburg, Hamburg, Germany, Sep 2017).

Winter, Bernd: Photoelectron Spectroscopy From Aqueous Solutions. (Physikalisches Kolloquium, Universität Kassel, Kassel, Germany, Nov 2017).

Winter, Bernd: Ions at the Aqueous Solution – Vacuum Interface Probed by Liquid-Jet Photoelectron Spectroscopy. (655. WE-Heraeus-Seminar: Surfaces and Interfaces of Ionic Liquids, Bad Honnef, Germany, Dec 2017).

Winter, Bernd: Photoemission Spectroscopy From Aqueous Solutions: Electron Binding Energies and Relaxation Processes. (Seminar, Department of Chemistry (AG Suzuki), Kyoto University, Kyoto, Japan, Jan 2018).

Winter, Bernd: Intra- and Intermolecular Processes in Aqueous Solutions: Photoemission From Liquid Microjets and New Developments. (10th International Meeting on Photodynamics and Related Aspects, Cartagena, Colombia, Sep 2018).

Winter, Bernd: Electronic Structure of Aqueous Solutions: Photoemission From Liquid Microjets and New Developments. (IOP Symposium, Emerging Spectroscopies: From the Gas-Phase to Complex Systems, London, UK, Nov 2018).

Winter, Bernd: Electronic Structure of Aqueous Solutions: Photoemission From Liquid Microjets and New Developments. (Theoretical Chemistry Seminar, Department of Inorganic Chemistry, University of Chemistry and Technology, Prague (UCT) and Institute of Organic Chemistry and Biochemistry of the CAS (IOCB), Prague, Czech Republic, Nov 2018).

Winter, Bernd: Photoelectron Spectroscopy of Liquid Ammonia and Metal-Ammonia Solutions. (FHI-Workshop on Current Research Topics at the FHI, Potsdam, Germany, May 2019).

Winter, Bernd: Nanoparticle – Aqueous Solution Interface Studied by Liquid Jet Photoemission. (Telluride Workshop, Water: Grand Challenges for Molecular Science and Engineering, Telluride, CO, USA, Jun 2019).

## Publications of the Department of Physical Chemistry

### Late 2017

Baierl, S., J. Mentink, M. Hohenleutner, C. Lange, T. Do, L. Braun, A. Sell, A. Zvezdin, M. Fiebig, G. Woltersdorf, T. Kampfrath, A. Kimel, R. Mikhaylovskiy and R. Huber: Efficient nonlinear control of spins by ultrashort THz-fields. In: 2017 IEEE International Magnetics Conference (INTERMAG). IEEE, New York, NY, 8007775 (2017). ISBN 978-1-5386-1086-2.

Bothschafter, E.M., E. Abreu, L. Rettig, T. Kubacka, S. Parchenko, M. Porer, C. Dornes, Y.W. Windsor, M. Ramakrishnan, A. Alberca, S. Manz, J. Saari, S.M. Koochpayeh, M. Fiebig, T. Forrest, P. Werner, S.S. Dhesi, S.L. Johnson and U. Staub: Dynamic pathway of the photoinduced phase transition of TbMnO<sub>3</sub>. *Physical Review B* **96** (18), 184414 (2017).

Cai, K., Y. Xiao, X. Lang, Y. Tong, T. He, Z. Mao and C. Wang: Porous anode of lithium-oxygen battery based on double-gas-path structure. *International Journal of Hydrogen Energy* **42** (50), 29944–29948 (2017).

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Ertl, G.: Fritz Haber and His Institute. In: One Hundred Years of Chemical Warfare: Research, Deployment, Consequences. (Eds.): B. Friedrich, D. Hoffmann, J. Renn, F. Schmaltz, and M. Wolf. Springer, Cham, 405–408 (2017). ISBN 978-3-319-51663-9. [OPEN ACCESS](#)

Franco-Cañellas, A., Q. Wang, K. Broch, D.A. Duncan, P. Kumar Thakur, L. Liu, S. Kera, A. Gerlach, S. Duhm and F. Schreiber: Metal-organic interface functionalization via acceptor end groups: PTCDI on coinage metals. *Physical Review Materials* **1** (1), 013001 (2017).

Friedrich, B., D. Hoffmann, J. Renn, F. Schmaltz and M. Wolf (Eds.): One Hundred Years of Chemical Warfare: Research, Deployment, Consequences. Springer, Cham (2017). ISBN 978-3-319-51663-9. [OPEN ACCESS](#)

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Koch, M., M. Pagan, M. Persson, S. Gawinkowski, J. Waluk and T. Kumagai: Direct observation of double hydrogen transfer via quantum tunneling in a single porphycene molecule on a Ag(110) surface. *Journal of the American Chemical Society* **139** (36), 12681–12687 (2017). [OPEN ACCESS](#)

Koch, M., S. Hecht and L. Grill: On-Surface Polymerization: From Polyarylenes to Graphene Nanoribbons and Two-Dimensional Networks. In: From Polyphenylenes to Nanographenes and Graphene Nanoribbons. (Eds.): K. Müllen and X. Feng. (Advances in Polymer Science, Vol. 278). Springer, Berlin, 99–125 (2017). ISBN 978-3-319-64169-0.

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Razdolski, I., A. Alekhin, N. Ilin, J.P. Meyburg, V. Roddatis, D. Diesing, U. Bovensiepen and A. Melnikov: Non-equilibrium magnetic effects at interfaces for ultrafast dynamics (Conference Presentation). In: *Proceedings of SPIE*. SPIE, Bellingham, Washington, 1035718 (2017).

Seifert, T., U. Martens, S. Günther, M.A.W. Schoen, F. Radu, X.Z. Chen, I. Lucas, R. Ramos, M.H. Aguirre, P.A. Algarabel, A. Anadón, H. Körner, J. Walowski, C. Back, M.R. Ibarra, L. Morellón, E. Saitoh, M. Wolf, C. Song, K. Uchida, M. Münzenberg, I. Radu and T. Kampfrath: Terahertz Spin Currents and Inverse Spin Hall Effect in Thin-Film Heterostructures Containing Complex Magnetic Compounds. *SPIN* **7** (3), 1740010 (2017). [OPEN ACCESS](#)

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#### Doctoral Thesis

Puppin, M.: Time- and angle-resolved photoemission spectroscopy on bidimensional semiconductors with a 500 kHz extreme ultraviolet light source. Freie Universität Berlin 2017.

## 2018

Böckmann, H., S. Gawinkowski, J. Waluk, M.B. Raschke, M. Wolf and T. Kumagai: Near-Field Enhanced Photochemistry of Single Molecules in a Scanning Tunneling Microscope Junction. *Nano Letters* **18** (1), 152–157 (2018).

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### Doctoral Theses

*Loutchko, D.*: A Theoretical Study of the Tryptophan Synthase Enzyme Reaction Network. Humboldt Universität Berlin 2018. [OPEN ACCESS](#)

*Nicholson, C.*: Electronic Structure and Dynamics of Quasi-One Dimensional Materials. Freie Universität Berlin 2018.

*Seifert, T.*: Spintronics with Terahertz Radiation: Probing and driving spins at highest frequencies. Freie Universität Berlin 2018.

### Master's Theses

*Bierhance, G.*: Broadband Terahertz Spectroscopy of Aqueous Solutions. Humboldt Universität Berlin 2018.

*Gückstock, O.*: Characterizing Spintronic Materials by Terahertz Spectroscopy. Technische Universität Berlin 2018.

Kohlmann, M.: Grating Coupling To Surface Phonon Polaritons. Technische Universität Berlin 2018.

Quadt, F.: Development and Automation of a SFG-FROG System with Labview. Freie Universität Berlin 2018.

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Alekhin, A., I. Razdolski, M. Berritta, D. Bürstel, V. Temnov, D. Diesing, U. Bovensiepen, G. Woltersdorf, P.M. Oppeneer and A. Melnikov: Magneto-optical properties of Au upon the injection of hot spin-polarized electrons across Fe/Au(0 0 1) interfaces. *Journal of Physics: Condensed Matter* **31** (12), 124002 (2019).

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Rettig, L., A. Caviezel, S.O. Mariager, G. Ingold, C. Dornes, S.-W. Huang, J.A. Johnson, M. Radovic, T. Huber, T. Kubacka, A. Ferrer, H.T. Lemke, M. Chollet, D. Zhu, J.M. Glowia, M. Sikorski, A. Robert, M. Nakamura, M. Kawasaki, Y. Tokura, S.L. Johnson, P. Beaud and U. Staub: Disentangling transient charge order from structural dynamics contributions during coherent atomic

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*Thämer, M., T. Garling, R.K. Campen and M. Wolf*: Quantitative determination of the nonlinear bulk and surface response from alpha-quartz using phase sensitive SFG spectroscopy. *The Journal of Chemical Physics* **151** (6), 064707 (2019).

*Tuitje, F., T. Helk, M. Zürch and C. Spielmann*: Extreme ultraviolet lensless imaging of biological specimen. In: *Proceedings of SPIE*. SPIE, Bellingham, Washington, 1089029 (2019).

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*Voloshina, E., Q. Guo, B. Paulus, S. Böttcher, H. Vita, K. Horn, C. Zhao, Y. Cui and Y. Dedkov*: Electronic Structure and Magnetic Properties of Graphene/Ni<sub>3</sub>Mn/Ni(111) Trilayer. *The Journal of Physical Chemistry C* **123** (8), 4994–5002 (2019).

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*Xian, R.P., L. Rettig and R. Ernstorfer*: Symmetry-guided nonrigid registration: The case for distortion correction in multidimensional photoemission spectroscopy. *Ultramicroscopy* **202**, 133–139 (2019).

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*Zürch, M., F. Tuitje, T. Helk, J. Gautier, F. Tissandier, J.-P. Goddet, E. Oliva, A. Guggenmos, U. Kleineberg, S. Sebban and C. Spielmann*: Ptychographic Characterization of an Intense High-Harmonic-seeded Femtosecond Soft X-ray Laser. In: *Proceedings Conference on Lasers and Electro-Optics (CLEO)*. OSA, Washington, D.C., SF31.3 (2019). ISBN 978-1-943580-57-6.

*Zwaschka, G., Y. Tong, M. Wolf and R.K. Campen*: Probing the Hydrogen Evolution Reaction and Charge Transfer on Platinum Electrodes on Femtosecond Timescales. *ChemElectroChem* **6** (10), 2675–2682 (2019).

### Doctoral Theses

*Böckmann, H.*: Far- and Near-Field Photochemistry of Single Molecules on a Metal Surface. Freie Universität Berlin 2019. 

*Mor, S.*: Fundamental interactions governing the (non-)equilibrium electronic structure in low dimensions. Freie Universität Berlin 2019.

*Vasileiadis, T.*: Ultrafast Energy Flow and Structural Changes in Nanoscale Heterostructures. Freie Universität Berlin 2019.

## Invited talks by members of the Department of Physical Chemistry

Beaulieu, Samuel: Attosecond-Resolved Photoionization of Chiral Molecules. (Multiphoton Processes, Gordon Research Conference, GRC, Smithfield, RI, USA, Jun 2018).

Beaulieu, Samuel: Using Attosecond and Strong-Field Physics Toolbox to Study Photoionization of Chiral Molecules. (CRC - Seminar, CRC 1319, Extreme light for sensing and driving molecular chirality (ELCH), Kassel, Germany, Oct 2018).

Beaulieu, Samuel: Photoelectron Circular Dichroism: From Single Photon to Strong Fields and From Static to Attosecond Dynamics. (Seminar, Department of Molecular Physics, Fritz Haber Institute, Berlin, Germany, Mar 2019).

Campen, R. Kramer: Experimentally Characterizing Interfacial Anion Polarizability:  $\text{ClO}_4^-$  at the air/water interface (International Workshop on Nonlinear Optics at Interfaces, Dalian, China, July 2017).

Campen, R. Kramer: Towards Experimentally Probing the Hydrogen Evolution Reaction on Pt and Au with Femtosecond Time Resolution (Dynamics at Surfaces, Gordon Research Conference, Newport, RI, USA, July 2017).

Campen, R. Kramer: Probing Surface Potentials and their Relationship to Electron Transfer: Hopefully at High Time and Spatial Resolution (International Workshop, Surface Potential Facts, Findings and Fantasies II, École Polytechnique Fédérale de Lausanne, Lausanne, Switzerland, Sep 2017).

Campen, R. Kramer: Watching the Hydrogen Evolution Reaction on Au and Pt with Femtosecond Time Resolution: Towards Experimentally Characterizing Mechanism (Klausurtagung of the Cluster of Excellence RESOLV, Velen, Germany, March 2018)

Campen, R. Kramer: The Chemical Physics of (Solid)/Liquid Interfaces in and out of Equilibrium: Probing Electron Transfer in Real Time (Seminar, Faculty of Physics, Universität Duisburg-Essen, Duisburg, Germany, May 2018).

Campen, R. Kramer: Experimentally Characterizing Anion Polarizability at the Air/Water Interface (Telluride Workshop, Nonlinear Optics at Interfaces, Telluride, CO, USA, June 2018).

Campen, R. Kramer: Towards a Molecular View of Charge Transfer at Aqueous Electrochemical Interfaces: The Hydrogen Evolution Reaction on Pt (Minisymposium on Infrared Spectroscopy, University of Amsterdam, Amsterdam, The Netherlands, Sep 2018).

Campen, R. Kramer: Towards a Femtosecond Resolved View of the Hydrogen Evolution Reaction on Au and Pt. (FHI-JST Joint Symposium, Current Topics and Challenges for Innovative Catalysts, Berlin, Germany, Oct 2018).

Campen, R. Kramer: Searching for the Active Site in Electrocatalysis: Imaging Oxidation and Oxygen Evolution on Gold Operando. (IAP Seminar, Institute of Applied Physics, TU Wien, Vienna, Austria, Mar 2019).

Campen, R. Kramer: A Femtosecond Resolved View of Charge Transfer Between Organic Adsorbates and Solids in UHV and at Electrified Solid/Liquid Interfaces (Workshop of the Collaborative Research Center 1242: Nonequilibrium Dynamics of Condensed Matter in the Time Domain, Schleiden, Germany, Mar 2019)

Campen, R. Kramer: Towards an Operando, Femtosecond Resolved, View of Water Electrolysis on Au and Pt (FHI-Workshop on Current Research Topics at the FHI, Potsdam, Germany, May 2019)

Chekhov, Alexander L.: Ultrafast Magnetoplasmonics. (Symposium, Spins, Waves and Interactions, University of Greifswald, Greifswald, Germany, Aug 2018).

Ernstorfer, Ralph: Electron-Phonon Coupling. (Block Course on New Concepts in Experimental and Theoretical Surface Science, IMPRS Functional Interfaces in Physics and Chemistry, Berlin, Germany, Sep 2017).

Ernstorfer, Ralph: Ultrafast View on Electron and Phonon Dynamics in Momentum Space. (Keynote Lecture, Lasers for Science Facility / Artemis User Meeting, Oxford, UK, Sep 2017).

Ernstorfer, Ralph: Accessing Microscopic Coupling in Solids with Momentum-Resolving Ultrafast Techniques. (Institutskolloquium, Technische Universität Ilmenau, Ilmenau, Germany, Oct 2017).

Ernstorfer, Ralph: Ultrafast View on Electron and Phonon Dynamics in Momentum Space. (Seminar, Institut für Theoretische Physik (AG Nichtlineare Optik & Quantenelektronik), Technische Universität Berlin, Berlin, Germany, Oct 2017).

Ernstorfer, Ralph: Accessing Microscopic Coupling in Solids with Momentum-Resolving Ultrafast Techniques. (FELIX laboratory user meeting 2017, Radboud University, Nijmegen, The Netherlands, Nov 2017).

Ernstorfer, Ralph: Momentum-Resolved Excited State Dynamics: From Electron-Phonon Coupling to Phase Transitions. (19th Meeting of the Fachbeirat of the Fritz-Haber-Institut, Berlin, Germany, Nov 2017).

Ernstorfer, Ralph: Accessing Microscopic Coupling in Solids with Momentum-Resolving Ultrafast Techniques. (ICUSD 2017, 4th International Conference on Ultrafast Structural Dynamics, Trieste, Italy, Dec 2017).

Ernstorfer, Ralph: Momentum-Resolved Excited State Dynamics: From Electron-Phonon Coupling to Phase Transitions. (LACUS Day, Lausanne Centre for Ultrafast Science, Lausanne, Switzerland, Feb 2018).

Ernstorfer, Ralph: Beyond the Molecular Movie: Out-Of-Equilibrium Electronic and Atomic Structure Investigated with trARPES and Diffraction. (Seminar, Department of Physics, University of Colorado at Boulder, Boulder, CO, USA, Jun 2018).

Ernstorfer, Ralph: Beyond the Molecular Movie: Ultrafast Dynamics of Bands, Bonds and Excitons Investigated with trARPES. (Workshop, Quantum Frontiers in Molecular Science, Telluride Science Research Center, Telluride, CO, USA, Jun 2018).

Ernstorfer, Ralph: Excited State Mapping of Transition Metal Dichalcogenides. (Workshop on excitations in solids, HoW exciting! 2018, Berlin, Germany, Jul 2018).

Ernstorfer, Ralph: Momentum-Resolved View of Ultrafast Dynamics of Electrons, Excitons and Phonons in Layered Semiconductors. (ICPS 2018, 34th International Conference on the Physics of Semiconductors, Montpellier, France, Jul 2018).

Ernstorfer, Ralph: Accessing Electron- and Exciton-Phonon Coupling in Solids with Momentum-Resolving Ultrafast Techniques. (PNGFVII, Progresses in NonEquilibrium Green's Functions VII, Frascati, Italy, Aug 2018).

Ernstorfer, Ralph: Momentum-Resolved View of Ultrafast Dynamics of Electrons, Excitons and Phonons in Layered Semiconductors. (ECOSS34, 34th European Conference on Surface Science, Aarhus, Denmark, Aug 2018).

Ernstorfer, Ralph: Electron, Exciton and Phonon Dynamics in Layered Semiconductors. (Summer School on Ultrafast Dynamics in Solids, Göttingen, Germany, Sep 2018).

Ernstorfer, Ralph: Momentum-Resolved View of Ultrafast Dynamics of Electrons, Excitons and Phonons in Crystals. (UFDIM 2018 - HICONO, Ultrafast Dynamic Imaging of Matter, Agios Nikolaos, Greece, Sep 2018).

Ernstorfer, Ralph: Momentum-Resolved Dynamics of Electrons and Phonons: From Electron-Phonon Coupling to Light-Induced Phase Transitions. (Seminar, Physics Department, Aarhus University, Aarhus, Denmark, Jan 2019).

Ernstorfer, Ralph: Momentum-Resolved Dynamics of Electrons and Phonons: From Electron-Phonon Coupling to Light-Induced Phase Transitions. (Seminar, Institute for Functional Matter and Quantum Technologies, University of Stuttgart, Stuttgart, Germany, May 2019).

Ernstorfer, Ralph: Multidimensional Photoemission Spectroscopy of Solids in Non-equilibrium States. (Workshop to explore the Science Potential of high power CW VUV FELs, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany, May 2019).

Ernstorfer, Ralph: Multidimensional Photoemission Spectroscopy of Optically Prepared Non-equilibrium States of  $WSe_2$ . (CORPES 2019, International Workshop on Strong Correlations and Angle-Resolved Photoemission Spectroscopy, Oxford, UK, Jul 2019).

Gierster, Lukas: Transient Metallization of a ZnO Surface Upon Photoexcitation. (Group Seminar, Institut für Optik und Atomare Physik (AG Ulrike Woggon), Technische Universität Berlin, Berlin, Germany, Apr 2019).

Kampfrath, Tobias: Ultrafast Magnetism with Terahertz Radiation. (Bad Honnef Physics School on Magnetism: From Fundamentals to Spin based Nanotechnology, Bad Honnef, Germany, Sep 2017).

Kampfrath, Tobias: Ultrafast Spintronics with Terahertz Radiation. (International Symposium, FMS 2017, Frontiers in Materials Science, Greifswald, Germany, Sep 2017).

Kampfrath, Tobias: Probing and Controlling Spin Dynamics by Terahertz Waves. (Tutorial: Terahertz Magnetism, 62nd Annual Conference on Magnetism and Magnetic Materials, MMM2017, Pittsburgh, PA, USA, Nov 2017).

Kampfrath, Tobias: Probing and Controlling Spin Dynamics by THz Waves. (Seminar, Institute of Physics and Astronomy (Host: M. Bargheer), University of Potsdam, Potsdam, Germany, Nov 2017).

Kampfrath, Tobias: Ultrafast Spintronics with Terahertz Radiation. (Focus Session, THz Spintronics with Ferrimagnets and Dirac/Weyl Materials, PIERS 2017 Progress In Electromagnetics Research Symposium, Singapore, Singapore, Nov 2017).

Kampfrath, Tobias: Ultrafast Torque on Spins and Molecules: New Insights into Magnets and Liquids. (Seminar, Department of Chemistry (Host: X.Y. Zhu), Columbia University, New York, NY, USA, Nov 2017).

Kampfrath, Tobias: The Ultimate Speed of the Spin-Dependent and Spin Seebeck Effect: Insights and Applications. (43rd Reimei/GP-Spin/ICC-IMR International Workshop, New Excitations in Spintronics, Sendai, Japan, Jan 2018).

Kampfrath, Tobias: Ultrafast Spintronics with Terahertz Radiation. (Condensed Matter Seminar, Clarendon Laboratory (Host: M. Johnston), University of Oxford, Oxford, UK, Feb 2018).

Kampfrath, Tobias: Probing and Controlling Spin Dynamics by Terahertz Waves. (EOS Topical Meeting on Terahertz Science & Technology (TST 2018), Berlin, Germany, May 2018).

Kampfrath, Tobias: Probing and Controlling Spin Dynamics with Terahertz Radiation. (7th Workshop Core-to-Core Project Tohoku-York-Kaiserslautern, New concept spintronic devices, Kaiserslautern, Germany, May 2018).

Kampfrath, Tobias: Probing and Controlling Ultrafast Spin Dynamics by Terahertz Waves. (IEEE International Conference on Microwave Magnetics 2018, ICMM 2018, Exeter, UK, Jun 2018).

Kampfrath, Tobias: Ultrafast Terahertz Spectroscopy: Probing and Controlling Fundamental Motions of Electrons, Spins and Ions. (Kolloquium, Institut für Physikalische Chemie (Host: H. Lange), Universität Hamburg, Hamburg, Germany, Jun 2018).

Kampfrath, Tobias: Ultrafast Terahertz Spectroscopy: Probing and Controlling Fundamental Motions of Electrons, Spins and Ions. (Physikalisches Kolloquium, Institut für Physik, Universität Augsburg, Augsburg, Germany, Jul 2018).

Kampfrath, Tobias: How to Measure Ultrafast Spin and Charge Currents. (The European School on Magnetism, ESM-2018, Kraków, Poland, Sep 2018).

Kampfrath, Tobias: Probing and Controlling Spin Dynamics with Terahertz Radiation. (9th JEMS Conference 2018, Joint European Magnetic Symposia, Mainz, Germany, Sep 2018).

Kampfrath, Tobias: Probing and Controlling Spin Dynamics with Ultrashort Terahertz Pulses. (The European School on Magnetism, ESM-2018, Kraków, Poland, Sep 2018).

Kampfrath, Tobias: Spintronics: From Fundamental Concepts to Terahertz Speed. (Summer School on Ultrafast Dynamics in Solids, Göttingen, Germany, Sep 2018).

Kampfrath, Tobias: Probing and Controlling Ultrafast Spin Dynamics With Terahertz Radiation. (Workshop, Collective Spin Dynamics in Nanostructures, Kavli Institute for Theoretical Sciences, Beijing, China, Oct 2018).

Kampfrath, Tobias: Ultrafast Terahertz Spectroscopy: Probing and Controlling Fundamental Motions of Electrons, Spins and Ions. (Physikalisches Kolloquium der Fakultät für Physik (Host: A. Lorke), Universität Duisburg-Essen, Duisburg, Germany, Dec 2018).

Kampfrath, Tobias: Ultrafast Terahertz Spectroscopy: Probing and Controlling Fundamental Motions of Electrons and Molecules in Condensed Matter. (Colloquium SFB 1078, Protonation Dynamics in Protein Function, Berlin, Germany, Feb 2019).

Kampfrath, Tobias: Ultrafast Spintronics With Terahertz Radiation. (Kolloquium, Max-Born-Institut, Berlin, Germany, May 2019).

Kampfrath, Tobias: Spintronics With Terahertz Radiation: Insights and Applications for Spin Dynamics and Terahertz Photonics. (CLEO® / Europe-EQEC, 2019 Conference on Lasers & Electro-Optics / Europe and European Quantum Electronics Conference, Munich, Germany, Jun 2019).

Kampfrath, Tobias: Ultrafast Spintronics with Terahertz Radiation. (Paris Ultrafast 2019, International Conference on Ultrafast and Nonlinear Dynamics of Quantum Materials, Paris, France, Jun 2019).

Kampfrath, Tobias: Terahertz Spintronics. (Spin Dynamics in Nanostructures, Gordon Research Conference, GRC, Les Diablerets, Switzerland, Jul 2019).

Kampfrath, Tobias: Ultrafast Spintronics with Terahertz Radiation. (International Workshop, Theoretical Developments and Experimental Progresses in Quantum Matter: Dynamics of Quantum Magnetism, Shanghai, China, Aug 2019).

King, Sarah B.: Tracing the Dynamics of Interfacial Electronic Excited States from Femtoseconds to Seconds. (Chemistry Special Seminar, Department of Chemistry, The University of Chicago, Chicago, IL, USA, Dec 2017).

King, Sarah B.: Tracing the Dynamics of Interfacial Electronic Excited States from Femtoseconds to Seconds. (Seminar, Department of Chemistry and Biochemistry, University of Oregon, Eugene, OR, USA, Jan 2018).

King, Sarah B.: Tracing the Dynamics of Interfacial Electronic Excited States from Femtoseconds to Seconds. (Special Chemistry Seminar, Division of Chemistry and Chemical Engineering, California Institute of Technology (Caltech), Pasadena, CA, USA, Jan 2018).

King, Sarah B.: Tracing Electron Transfer and Localization Dynamics at a Model Electrolyte/Metal Interface From Femtoseconds to Seconds. (DIET16, 16th international workshop on Dynamics, Interactions and Electronic Transitions at Surfaces, Garmisch-Partenkirchen, Germany, Oct 2018).

Koch, Matthias: Single Shot Spin Readout With a 3D Crystalline Transistor. (Seminar, International Center for Materials Nanoarchitectonics (MANA), Tsukuba, Japan, Nov 2017).

Koch, Matthias: 3D Device Fabrication With Scanning Tunneling Hydrogen Resist Lithography. (Seminar, Department of Physical Chemistry, University of Graz, Graz, Austria, Mar 2018).

Koch, Matthias: Demonstration of Spin Read-Out in Atomic Donors in an All-Epitaxial 3D Transistor. (Seminar, Ferdinand-Braun-Institut, Leibniz-Institut für Höchstfrequenztechnik, Berlin, Germany, Jan 2019).

Koch, Matthias: Demonstration of Spin Read-Out in Ph Quantum Dots in an All-Epitaxial 3D Transistor. (Seminar, University College London, London, UK, Aug 2019).

Kumagai, Takashi: Near-Field Induced Chemistry Studied by Scanning Tunneling Microscopy. (5th Ito International Research Conference, RIKEN Centennial Anniversary & Surface and Interface Spectroscopy 2017, Forefront of Molecular Dynamics at Surfaces and Interfaces: From a single molecule to catalytic reaction, Tokyo, Japan, Nov 2017).

Kumagai, Takashi: Local Geometric and Electronic Structure of a Single Naphthalocyanine Molecule on Ultrathin ZnO Layers. (Colloquium of the CRC 951 - Hybrid Inorganic/Organic Systems for Opto-Electronics (HIOS), Berlin, Germany, Jan 2018).

Kumagai, Takashi: Single-Molecule Chemistry on Surfaces Studied by Scanning Probe Microscopy. (Lecture Series Winter Semester 2017/2018, Modern Methods in Heterogeneous Catalysis Research, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Jan 2018).

Kumagai, Takashi: Physical Chemistry with Single Molecules Studied by Scanning Probe Microscopy. (Seminar, Kyoto University, Kyoto, Japan, May 2018).

Kumagai, Takashi: Physical Chemistry with Single Molecules Studied by Scanning Probe Microscopy. (Seminar Theoretical Chemistry, University of Potsdam, Potsdam, Germany, Jun 2018).

Kumagai, Takashi: Real-Space Study of Hydrogen Transfer Reactions. (Seminar, Max Planck Institute for Polymer Research, Mainz, Germany, Jun 2018).

Kumagai, Takashi: Physical Chemistry with Single Molecules - Hydrogen Transfer Reactions in Real Space. (Section Meeting, Chemistry, Physics and Technology Section, Max Planck Society, Berlin, Germany, Oct 2018).

Kumagai, Takashi: Tip-Enhanced Raman Spectroscopy of Ultrathin ZnO Layers. (FHI-JST Joint Symposium, Current Topics and Challenges for Innovative Catalysts, Berlin, Germany, Oct 2018).

Kumagai, Takashi: Nanoscale Surface Chemistry Studied by SPM – Single-Molecule Tautomerization and Near-Field Chemistry/Physics. (Seminar, Nanoscale Science Department, Max Planck Institute for Solid State Research, Stuttgart, Germany, Nov 2018).

Kumagai, Takashi: Physical Chemistry with Single Molecules - Hydrogen Transfer Reactions in Real Space. (Seminar, Polish Academy of Sciences, Warsaw, Poland, Nov 2018).

Kumagai, Takashi: Nanoscale Surface Chemistry Studied by SPM – Single-Molecule Tautomerization and Near-Field Chemistry/Physics. (RIKEN Seminar, Saitama, Japan, Dec 2018).

Kumagai, Takashi: Nanoscale Surface Chemistry Studied by SPM – Single-Molecule Tautomerization and Near-Field Chemistry/Physics. (Seminar, Department of Chemistry, Tokyo Institute of Technology, Tokyo, Japan, Dec 2018).

Kumagai, Takashi: Nanoscale Surface Chemistry Studied by SPM – Single-Molecule Tautomerization and Near-Field Chemistry/Physics. (Seminar, Osaka University, Osaka, Japan, Jan 2019).

Kumagai, Takashi: Nanoscale Surface Chemistry Studied by SPM – Single-Molecule Tautomerization and Near-Field Chemistry/Physics. (Seminar, University of Tsukuba, Tsukuba, Japan, Jan 2019).

Kumagai, Takashi: Physical Chemistry with Single Molecules. (Seminar, Tokyo University of Science, Tokyo, Japan, Jan 2019).

Kumagai, Takashi: Physical Chemistry with Single Molecules-Hydrogen-Transfer Dynamics Studied by Scanning Probe Microscopy. (Seminar, Nagoya University, Nagoya, Japan, Jan 2019).

Kumagai, Takashi: Plasmon-Induced Photophysics and Photochemistry in Nanocavity. (9th International Workshop on Surface Physics, Trzebnica, Poland, Jun 2019).

Kumagai, Takashi: Near-Field Induced Photophysics and Photochemistry Studied by Low-Temperature STM. (Seminar, Institute for Functional Matter and Quantum Technologies, University of Stuttgart, Stuttgart, Germany, Jul 2019).

Mährlein, Sebastian: The THz Sum-Frequency Counterparts of Stimulated Raman Scattering. (SPIE Optics + Photonics 2019, San Diego, CA, USA, Aug 2019).

Mor, Selene: Ultrafast Electronic and Structural Dynamic in an Excitonic Insulator. (Seminar, Department of Physics, Politecnico di Milano, Milano, Italy, Apr 2018).

Mor, Selene: Ultrafast Electronic and Structural Dynamic in an Excitonic Insulator. (Seminar, Department of Physics, University of Fribourg, Fribourg, Switzerland, Jun 2018).

Mor, Selene: Ultrafast Electronic and Structural Dynamics in an Excitonic Insulator. (LEES 2018, International Conference on Low-Energy Electrodynamics in Solids, Portonovo, Italy, Jun 2018).

Mor, Selene: Ultrafast Electronic Band Gap Control and Inhibition of the Photoinduced Structural Phase Transition in the Excitonic Insulator. (LACUS Seminar, Lausanne Centre for Ultrafast Science, EPFL, Lausanne, Switzerland, Jun 2018).

Mor, Selene: Ultrafast Dynamics in Low Dimensional Materials. (Interview, Università Cattolica del Sacro Cuore, Brescia, Italy, Oct 2018).

Nadvornik, Lukas: Fast Optical Control of Spin in Systems Possessing High-Performance Spin Transport Properties. (19th Conference of Czech and Slovak Physicists, Presov, Slovakia, Sep 2017).

Nicholson, Christopher: Ultrafast Electronic Structure Dynamics During the Photo-Induced Phase Transition in In/Si(111). (Seminar, Department for Physics (AG Horn-von Hoegen), Universität Duisburg-Essen, Duisburg, Germany, Sep 2017).

Nicholson, Christopher: Transient Electronic Structure Dynamics Probed by trARPES at 500 kHz. (Seminar, Department of Chemistry (X.Y. Zhu Group), Columbia University, New York, NY, USA, Nov 2017).

Nicholson, Christopher: Ultrafast Dynamics of Bands and Bonds During a Photo-Induced Phase Transition. (Ultrafast Phenomena in Cooperative Systems, Gordon Research Conference, GRC, Galveston, TX, USA, Feb 2018).

Nicholson, Christopher: Ultrafast Dynamics of Bands and Bonds During a Photo-Induced Phase Transition. (SIMES Seminar, Stanford Institute for Materials and Energy Sciences, Stanford University, Stanford, CA, USA, Apr 2018).

Nicholson, Christopher: A Chemical and Physical View of Ultrafast Phase Transitions in Solids. (Seminar, Institut de physique de Rennes, Université de Rennes 1, Rennes, France, Nov 2018).

Paarmann, Alexander: Nonlinear Phonon Polariton Spectroscopy. (Seminar, Experimental Condensed Matter Physics, RWTH Aachen University, Aachen, Germany, Sep 2017).

Paarmann, Alexander: Nonlinear Phonon Polariton Spectroscopy. (Seminar, The Institute of Photonic Sciences, Castelldefels, Spain, Sep 2017).

Paarmann, Alexander: Nonlinear Phonon Polariton Spectroscopy. (Seminar, Institute of Physics, Surface Science Group, Martin-Luther-Universität Halle-Wittenberg, Halle, Germany, Dec 2017).

Paarmann, Alexander: Nonlinear Phonon Spectroscopy Using Infrared Free-Electron Lasers. (Free-Electron Laser Workshop, Applications of IR Free-Electron Lasers: Latest Developments and Future Directions at Ringberg Castle, Kreuth, Germany, Jan 2018).

Paarmann, Alexander: Nonlinear Surface Phonon Polariton Spectroscopy. (DPG-Frühjahrstagung und EPS-CMD27, Ultrafast Electron and spin dynamics at interfaces II, Berlin, Germany, Mar 2018).

Paarmann, Alexander: Nonlinear Phonon Polariton Spectroscopy Using Infrared Free-Electron Lasers. (Seminar, Department Molecular Spectroscopy, Max Planck Institute for Polymer Research, Mainz, Germany, Oct 2018).

Paarmann, Alexander: Nonlinear Phonon Polariton Spectroscopy Using Infrared Free-Electron Lasers. (Seminar, Interface and Surface Physics, University of Greifswald, Greifswald, Germany, Apr 2019).

Paarmann, Alexander: Nonlinear Phonon Polariton Spectroscopy Using Infrared Free-Electron Lasers. (Seminar, Abteilung Terahertz und Laserspektroskopie, Deutsches Zentrum für Luft- und Raumfahrt, Berlin, Germany, Jun 2019).

Paßler, Nikolai: Linear and Nonlinear Optical Response of Surface Phonon Polaritons in Polar Dielectric Heterostructures. (Group Seminar, Department of Physics (AG Paul Fumagalli), Freie Universität Berlin, Berlin, Germany, Feb 2018).

Rettig, Laurenz: Fundamental Couplings in Strongly Correlated Materials Investigated by Complementary Time-Domain Techniques. (NGSCES 2017, 8th International Conference, New Generation in Strongly Correlated Electrons Systems, Barcelona, Spain, Sep 2017).

Rettig, Laurenz: Strongly Correlated Materials Investigated by Complementary Time-Domain Techniques. (Department Workshop at Lake Griebnitzsee, Department of Physical Chemistry, FHI, Potsdam, Germany, May 2018).

Rettig, Laurenz: Fundamental Couplings in Strongly Correlated Materials Investigated by Complementary Time-Domain Techniques. (Colloquium of the CRC 1242, Non-Equilibrium Dynamics of Condensed Matter in the Time Domain, Duisburg, Germany, Jun 2018).

Rettig, Laurenz: Strongly Correlated Materials Investigated by Complementary Time-Domain Techniques. (Workshop, Ultrafast Dynamics at the Nanoscale, Fritz Haber Institute, Berlin, Germany, Jun 2018).

Rettig, Laurenz: Strongly Correlated Materials Investigated by Complementary Time-Domain Techniques. (CFEL Seminar, Desy, Hamburg, Germany, Oct 2018).

Rettig, Laurenz: Transient Potential Energy Surfaces: A Key to Photoinduced Phase Transitions. (Workshop, Nonequilibrium Quantum Dynamics and Relaxation Phenomena in Many Body Systems, Krvavec, Slovenia, Dec 2018).

Rettig, Laurenz: Strongly Correlated Materials Investigated by Complementary Time-Domain Techniques. (Lectures, CRC/Transregio 173, Spin+X: Spin in its collective environment, Mainz, Germany, Jun 2019).

Rettig, Laurenz: Ultrafast Dynamics in Ferromagnetic and Antiferromagnetic Rare-Earth Compounds. (TRR227 Retreat, CRC/Transregio 227, Ultrafast Spin Dynamics, Halle, Germany, Jun 2019).

Rettig, Laurenz: Beyond the Molecular Movie: Tracking the Transient Potential Energy Surface of Photo-Induced Phase Transitions Through the Ultrafast Electronic Structure. (International Conference on Ultrafast Spectroscopy of Correlated Quantum Materials (USCQM - 2019), Liyang, China, Jul 2019).

Sajadi, Mohsen: Intermolecular Mode Coupling in Liquids: Insights from the THz Kerr Effect. (Seminar, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany, Oct 2017).

Sajadi, Mohsen: Intermolecular Interactions in Liquids: Insights from Nonlinear THz Spectroscopy. (Ortsverbandsvortrag, Gesellschaft Deutscher Chemiker, Paderborn, Germany, Jun 2018).

Sajadi, Mohsen: Energy Transfer Within Hydrogen Bonding Network of Water Following Resonant THz Excitation. (Seminar, Institut de Physique et Chimie des Matériaux de Strasbourg, Strasbourg, France, Oct 2018).

Sajadi, Mohsen: Nonlinear THz Spectroscopy of Liquids. (Seminar, Moscow Institute of Physics and Technology, Moscow, Russia, Apr 2019).

Stähler, Julia: Ultrafast Electronic Band Gap Control and Self-Protection from a Photoinduced Phase Transition in an Excitonic Insulator. (ICEL 2017 - International Conference on Extreme Light, Szeged, Hungary, Nov 2017).

Stähler, Julia: Ultrafast Electron Solvation Dynamics and Chemistry in Polar Solvents. (Photoionization and Photodetachment, Gordon Research Conference, GRC, Galveston, TX, USA, Feb 2018).

Stähler, Julia: Ultrafast Electronic Band Gap Control and Self-Protection from a Structural Phase Transition in an Excitonic Insulator. (DPG-Frühjahrstagung und EPS-CMD27, Focus Session: Structural Dynamics in Nanoscale Materials, Probed by Ultrafast Electron Pulses II, Berlin, Germany, Mar 2018).

Stähler, Julia: Es werde Licht - Und zwar schnell! (Schule MIT Wissenschaft Hamburg, Konferenz für MINT-Lehrkräfte, Hamburg, Germany, Jun 2018).

Stähler, Julia: Ultrafast Electron Solvation Dynamics and Chemistry at the DMSO/Cu(111) Interface. (IMAMPC2018, 9th International Meeting on Atomic and Molecular Physics and Chemistry, Berlin, Germany, Jun 2018).

Stähler, Julia: Challenging Non-Equilibrium Phenomena in Condensed Matter and at Interfaces. (Selection Symposium of the Lise Meitner Excellence Program 2018, Berlin, Germany, Jul 2018).

Stähler, Julia: Ultrafast Electron Solvation Dynamics at the Origin of an Electrochemical Reaction Chain. (Workshop on excitations in solids, HoW exciting! 2018, Berlin, Germany, Jul 2018).

Stähler, Julia: Ultrafast Electronic Band Gap Control and Inhibition of a Photoinduced Phase Transition in an Excitonic Insulator. (2018 E-MRS Fall Meeting and Exhibit, Warsaw, Poland, Sep 2018).

Stähler, Julia: Ultrafast Quasiparticle Dynamics in a Transition Metal Dichalcogenide and at Hybrid Organic/Inorganic Interfaces. (International conference on Non-equilibrium dynamics of Condensed Matter in the Time Domain, CRC 1242, Kerkrade, The Netherlands, Sep 2018).

Stähler, Julia: Ultrafast Quasiparticle Dynamics in a Transition Metal Dichalcogenide and at Hybrid Organic/Inorganic Interfaces. (Nanoscience Colloquium DESY, Hamburg, Germany, Jan 2019).

Stähler, Julia: Ultrafast Quasiparticle Dynamics in ZnO and at Its Interface With an Organic Chromophore. (LACUS Day, Lausanne Centre for Ultrafast Science, EPFL, Lausanne, Switzerland, Feb 2019).

Stähler, Julia: Understanding Charge Carrier & Exciton Dynamics in ZnO and at Its Surfaces From Femto- to Microseconds. (Department Colloquium, Department of Chemistry & Chemical Biology, Cornell University, Ithaca, NY, USA, Apr 2019).

Stähler, Julia: Ultrafast Quasiparticle Dynamics in ZnO and at Its Interface With an Organic Chromophore. (Colloquium of the CRC 951, Hybrid Inorganic/Organic Systems for Opto-Electronics (HIOS), Berlin, Germany, May 2019).

Stähler, Julia: Band Gap Engineering on Ultrafast Timescales. (Paris Ultrafast 2019, International Conference on Ultrafast and Nonlinear Dynamics of Quantum Materials, Paris, France, Jun 2019).

Stähler, Julia: Band Gap Engineering on Ultrafast Timescales. (Department Seminar, Institut für Festkörper- und Materialphysik, Technische Universität Dresden, Dresden, Germany, Jul 2019).

Stähler, Julia: Inverse Surface Photovoltage-Driven Semiconductor-to-Metal Transition in ZnO Featuring Ultrafast Recovery. (Symposium on the occasion of Andrea Cavalleri's 50th Birthday, University of Oxford, Oxford, UK, Jul 2019).

Stähler, Julia: Taming ZnO: Following the Many Pathways of Optical Excitations in a Recalcitrant Material From Femto- to Microseconds. (20. Tagung Festkörperanalytik, FKA 20, Vienna, Austria, Jul 2019).

Tong, Yujin: Employing Vibrational Sum-Frequency Spectroscopy to Study Adsorbate Structure at Electrochemical Interfaces. (19th National Conference on Electrochemistry in China (NCEC), Shanghai, China, Dec 2017).

Tong, Yujin: Highlights of Nonlinear Spectro-Electrochemistry Group. (Seminar, Department of Materials Science and Engineering (Yang Shao-Horn), Massachusetts Institute of Technology, Cambridge, MA, USA, Aug 2018).

Tong, Yujin: Mechanistic Studies of OER on Gold Electrode by Interfacial Specific Vibrational Spectroscopy. (256th ACS National Meeting & Exposition, Nanoscience, Nanotechnology & Beyond, Symposium on Sustainable Energy Conversion via Innovative Electrocatalysis & Photocatalysis, Boston, MA, USA, Aug 2018).

Tong, Yujin: Mechanistic Studies of OER on Gold Electrode by Interfacial Specific Vibrational Spectroscopy. (NMSSA 2018, National Meeting of Surface Science and Application, Suzhou, China, Oct 2018).

Tong, Yujin: Probing Fundamental Electrochemical Processes With Vibrational Sum Frequency (SFG) Spectroscopy. (Seminar, Department of Chemistry (AG Igor Ying Zhang), Fudan University, Shanghai, China, Oct 2018).

Tong, Yujin: Spectroscopy at Electrochemical Interfaces. (Block Course on Fundamentals and concepts to study solids, interfaces and molecules, IMPRS Functional Interfaces in Physics and Chemistry, Berlin, Germany, Oct 2018).

Tong, Yujin: Probing Fundamental Processes in Electrochemical Catalytic Reactions With Nonlinear Spectroscopy. (International 5th Ertl Symposium on Catalytic & Adsorption Reactions in chemical Processes, Gwangju, South Korea, Nov 2018).

Tong, Yujin: Vibrational Sum Frequency Spectroscopy as a Powerful Tool for Study Fundamental Processes in Electrochemical Catalytic Reactions. (31st CGCA Annual Conference, Chinese-German Chemical Association, Leipzig, Germany, May 2019).

Tong, Yujin: Probing Anions at Electrochemical Interfaces With Vibrational Sum Frequency Spectroscopy. (International Workshop, Nonlinear Optics at Interfaces 2019, Shanghai, China, Jun 2019).

Tong, Yujin: Probing Fundamental Processes in Electrochemical Catalytic Reactions With Nonlinear Spectroscopy. (Seminar, Department of Physical Chemistry, (AG Zhanbing Yang), University of Science and Technology of China, Beijing, China, Jun 2019).

Tong, Yujin: Advanced Analysis Technology in Surface Physical Chemistry. (2019 HIT International Summer School, Harbin Institute of Technology, Harbin, China, Jul 2019).

Tong, Yujin: Probing Fundamental Processes in Electrochemical Catalytic Reactions With Nonlinear Spectroscopy. (Seminar, Physical Education Department (AG Wenxin Wang), Harbin Engineering University, Harbin, China, Jul 2019).

Winta, Christopher: Atomic-Scale Heterostructures Studied with Mid-IR Second-Harmonic Phonon Spectroscopy. (Free-Electron Laser Workshop, Applications of IR Free-Electron Lasers: Latest Developments and Future Directions at Ringberg Castle, Kreuth, Germany, Jan 2018).

Winta, Christopher: Low-Temperature Second-Harmonic Phonon Spectroscopy of  $\alpha$ -Quartz. (Group Seminar, Department of Physics (AG Wolfgang Kuch), Freie Universität Berlin, Berlin, Germany, May 2018).

Wolf, Martin: Ultrafast Dynamics of Quasi 2D Materials Probed by Time-Resolved ARPES. (Success-2017, School on UV and X-ray spectroscopies of correlated electron systems, Les Houches, France, Sep 2017).

Wolf, Martin: Ultrafast Electronic and Structural Dynamic at Interfaces. (Colloquium, Ertl Center for Electrochemistry and Catalysis, GIST – Gwangju Institute of Science and Technology, Gwangju, South Korea, Sep 2017).

Wolf, Martin: Ultrafast Electronic and Structural Dynamic in Solids Probed by Time-Resolved ARPES. (Colloquium, IBS Center for Multidimensional Carbon Materials, Suwon, South Korea, Sep 2017).

Wolf, Martin: Ultrafast Excited State Dynamics and Photoinduced Phase Transitions Probed by Time-Resolved ARPES. (International Workshop Non-Linear Effects and Short-Time Dynamics in Novel Superconductors and Correlated Spin-Orbit Coupled Systems, ICTP Asian Network on Condensed Matter and Complex Systems, Daejeon, South Korea, Sep 2017).

Wolf, Martin: Ultrafast Excited State Dynamics and Photoinduced Phase Transitions at Interfaces. (5th Ito International Research Conference, RIKEN Centennial Anniversary & Surface and Interface Spectroscopy 2017, Forefront of Molecular Dynamics at Surfaces and Interfaces: From a single molecule to catalytic reaction, Tokyo, Japan, Nov 2017).

Wolf, Martin: Introductory Talk: Molecular Materials: Excitons, Polarons, Phonon Glasses & More. (Ultrafast Phenomena in Cooperative Systems, Gordon Research Conference, GRC, Galveston, TX, USA, Feb 2018).

Wolf, Martin: Nonlinear Surface Phonon Polariton Spectroscopy Using an Infrared Free-Electron Laser. (DPG-Frühjahrstagung und EPS-CMD27, Berlin, Germany, Mar 2018).

Wolf, Martin: Beyond the Molecular Movie: Ultrafast Dynamics of Bands and Bonds During a Photo-Induced Phase Transition. (30st CGCA Annual Conference, Chinese-German Chemical Association, Berlin, Germany, Apr 2018).

Wolf, Martin: Excited State Dynamics and Spectroscopy of In Nanowires on Vicinal Silicon. (Mini-Symposium on Atomic Wires on Vicinal Si Surfaces, Duisburg, Germany, Apr 2018).

Wolf, Martin: Recent Developments in Excited State Photoemission Spectroscopy of Low-Dimensional Systems. (CRC 1242 Workshop on Light Field Induced Dynamics in Low-dimensional Systems, Duisburg, Germany, Apr 2018).

Wolf, Martin: Ultrafast Excited State Dynamics and Photo-Induced Phase Transitions Probed by Time-Resolved Photoemission. (Physical Chemistry Colloquium, Ludwig-Maximilians-Universität München, Munich, Germany, Apr 2018).

Wolf, Martin: Ultrafast Phase Transitions and Excited State Dynamics at Surfaces. (SPIE Photonics Europe 2018, Strasbourg, France, Apr 2018).

Wolf, Martin: Electronic Structure and Dynamics of In Nanowires on Si(111). (Discussion Workshop, Ultrafast Dynamics at the Nanoscale, Berlin, Germany, Jun 2018).

Wolf, Martin: Ultrafast Excited State Dynamics of Low Dimensional Solid State Materials in Momentum and Energy Space. (Workshop, Molecular Videography, Telluride Science Research Center, Telluride, CO, USA, Sep 2018).

Wolf, Martin: Ultrafast Phase Transitions and Near-Field Induced Processes at Surfaces. (STROBE Science Center Seminar, University of Colorado at Boulder, Boulder, CO, USA, Sep 2018).

Wolf, Martin: Solid State Physics Viewed From an Ultrafast Perspective. (Solid State Physics Symposium, Technische Universität Kaiserslautern, Kaiserslautern, Germany, Oct 2018).

Wolf, Martin: Ultrafast Excited State Dynamics Probed by 500 kHz Time-Resolved ARPES. (Seminar, ZX Shen Group, Stanford University, Stanford, CA, USA, Oct 2018).

Wolf, Martin: Ultrafast Electronic and Structural Dynamics at Surfaces. (International Symposium on Ultrafast Science: From the infrared to the X-rays, ISUS 2018, Lausanne, Switzerland, Nov 2018).

Wolf, Martin: Ultrafast Electronic and Structural Dynamics at Surfaces. (Laserlab-Europe Workshop, ICFO – The Institute of Photonic Sciences, Barcelona, Spain, Nov 2018).

Wolf, Martin: Beyond the Molecular Movie: The Ultrafast Electronic Structure View of Surface Dynamics. (Plenary Lecture, DPG-Frühjahrstagung 2019, Regensburg, Germany, Apr 2019).

Wolf, Martin: Ultrafast Excited State Dynamics of Solids Probed by Time-Resolved ARPES. (Second International Workshop on Ultrafast Dynamics and Metastability and Fifth International Workshop on Ultrafast Bandgap Photonics, Washington, DC, USA, Apr 2019).

Wolf, Martin: Ultrafast Excited State Dynamics Probed by trARPES & Plasmon Mediated Dynamics in STM Junctions. (11-th International Symposium on Ultrafast Surface Dynamics, USD11, Qiandao Lake, China, Jun 2019).

Wolf, Martin: Beyond the Molecular Movie: The Ultrafast Electronic Structure View on Surface Dynamics. (14th Femtochemistry Conference, FEMTO 14, Shanghai, China, Jul 2019).

Wolf, Martin: Ultrafast Excited State Dynamics and Photoinduced Phase Transitions of Solids Probed by Time Resolved Photoemission. (Symposia, Chemistry in Real Space & Time, ACS National Meeting and Exposition, San Diego, CA, USA, Aug 2019).

Xian, R. Patrick: Multidimensional Photoelectron Spectroscopy. (BiGmax Workshop 2018 on Big-Data-Driven Materials Science, Kloster Irsee, Irsee, Germany, Apr 2018).

Xian, R. Patrick: Challenges and Opportunities in Multidimensional Photoemission Spectroscopy (MPES) Data Analysis. (MaxNet data format workshop, Potsdam, Germany, Aug 2018).

Xian, R. Patrick: Challenges and Opportunities in Multidimensional Photoemission Spectroscopy (MPES) Data Analysis. (Seminar, Empirical Inference Department, Max Planck Institute for Intelligent Systems, Tübingen, Germany, Aug 2018).

Zürch, Michael: Single Shot XUV Nanoimaging Using an Intense Femtosecond Soft X-Ray Laser. (Institutskolloquium, Leibniz Institute of Surface Engineering, Leipzig, Germany, Jan 2019).

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Wagner, C., M.F.B. Green, M. Maiworm, P. Leinen, T. Esat, N. Ferri, N. Friedrich, R. Findeisen, A. Tkatchenko, R. Temirov and F.S. Tautz: Quantitative imaging of electric surface potentials with single-atom sensitivity. *Nature Materials* **18** (8), 853–859 (2019).

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Zhang, I.Y., G. Zwaschka, Z. Wang, M. Wolf, R.K. Campen and Y. Tong: Resolving the Chemical Identity of H<sub>2</sub>SO<sub>4</sub> Derived Anions on Pt(111) Electrodes: They're Sulfate. *Physical Chemistry Chemical Physics*, in press. 

Zhao, L., N. Lin, Z. Han, X. Li, H. Wang and J. Cui: Regulation of the Crystal Structure Leading to the Bandgap Widening and Phonon Scattering Increasing in Cu<sub>3</sub>SnS<sub>4</sub>-Cu<sub>3</sub>SbSe<sub>3</sub> Chalcogenides. *Advanced Electronic Materials*, in press.

## Invited talks by members of the Theory Department

Baldauf, Carsten: Biomolecules in Thin Air. (Seminar, Heidelberg Institute for Theoretical Studies, Heidelberg, Germany, Sep 2017).

Baldauf, Carsten: Prediction of Biomolecular Structure and Dynamics. (Seminar, Fachbereich V, Beuth Hochschule für Technik Berlin, Berlin, Germany, Oct 2017).

Baldauf, Carsten: Biomolecules in Thin Air. (Seminar, Cluster of Excellence RESOLV, Fakultät für Chemie und Biochemie, Ruhr-Universität Bochum, Bochum, Germany, Jan 2018).

Baldauf, Carsten: Biomolecules in Thin Air. (Seminar, Institute of Organic Chemistry and Biochemistry of the CAS, Czech Academy of Sciences, Prague, Czech Republic, Jan 2018).

Baldauf, Carsten: Biomolecules in Thin Air. (Seminar, Centre For Molecular Simulation, The University of Calgary, Calgary, AB, Canada, May 2018).

Baldauf, Carsten: Cation-Peptide Interactions from First Principles. (101st Canadian Chemistry Conference and Exhibition, Symposium: Assessing Complex Free Energy Surfaces from Molecular Simulations from Electronic Structure to Mesoscopic Processes, Edmonton, AB, Canada, May 2018).

Baldauf, Carsten: Biomolecular Structure Search. (Seminar, State Key Laboratory of Superhard Materials, College of Physics, Jilin University, Changchun, China, Jul 2018).

Baldauf, Carsten: Enhanced Sampling Methods. (Hands-On DFT and Beyond: Frontiers of Advanced Electronic Structure and Molecular Dynamics Methods, Beijing, China, Jul 2018).

Baldauf, Carsten: Biomolecules in Thin Air. (Institutskolloquium, Leibniz Institute of Surface Engineering (IOM), Leipzig, Germany, Dec 2018).

Baldauf, Carsten: Biomolecules – From the Gas Phase to the Interface. (Seminar, Max Planck Institute for Solid State Research, Stuttgart, Germany, May 2019).

Baldauf, Carsten: Biomolecules in Isolation and FAIR Research Data Infrastructure. (Seminar, Institute of Nanotechnology, Karlsruhe Institute of Technology (KIT), Karlsruhe, Germany, May 2019).

Baldauf, Carsten: Bioorganic Molecules – From the Gas Phase to the Interface. (Seminar, Fakultät für Chemie und Mineralogie, Universität Leipzig, Leipzig, Germany, Jun 2019).

Boley, Mario: Subgroup Discovery. (NOMAD Summer - A hands-on course on tools for novel materials discovery, Berlin, Germany, Sep 2017).

Boley, Mario: Subgroup Discovery in Materials Science. (NOMAD Summer - A hands-on course on tools for novel-materials discovery, CECAM, Lausanne, Switzerland, Sep 2018).

Carbogno, Christian: First-Principles High-Throughput Design and Discovery of Novel Materials, and Its Application to Thermoelectrics. (6th Science Day of the Max Planck-EPFL Center for Molecular Nanoscience & Technology, Lausanne, Switzerland, Oct 2017).

Carbogno, Christian: Strongly Anharmonic Nuclear Dynamics in Solids: Accurate Computations and Rapid Estimates. (CECAM Workshop, Anharmonicity and thermal properties of solids, Paris, France, Jan 2018).

Carbogno, Christian: All-Electron, Real-Space Perturbation Theory: Theory, Implementation, and Application within DFT. (ETSF Workshop, Electron-phonon interaction and its effects in

condensed matter and reduced dimensionality systems, Louvain-la-Neuve, Belgium, May 2018).

Carbogno, Christian: Ab Initio Materials Simulations: A Challenge for Eigensolvers. (PMAA18, 10th International Workshop on Parallel Matrix Algorithms and Applications, Zurich, Switzerland, Jun 2018).

Carbogno, Christian: Electron-Phonon Coupling and Heat Transport. (Hands-On DFT and Beyond: Frontiers of Advanced Electronic Structure and Molecular Dynamics Methods, Beijing, China, Jul 2018).

Carbogno, Christian: Numerical Quality Control for DFT-Based Materials Databases. (JMC 2018, Journées de la Matière Condensée 2018, Grenoble, France, Aug 2018).

Carbogno, Christian: Recent Advancements in the ELPA Library: Best Practices in Real Applications. (MoSSI Workshop / ELSI Conference: Solving or Circumventing Eigenvalue Problems in Electronic Structure Theory, Richmond, VA, USA, Aug 2018).

Carbogno, Christian: High-Throughput Calculations, Error Estimates in Density Functional Theory. (NOMAD Summer - A hands-on course on tools for novel-materials discovery, CECAM, Lausanne, Switzerland, Sep 2018).

Carbogno, Christian: Solving the Veracity Challenge in Electronic-Structure Theory Data. (Final M36 Nomad Review Meeting, European Commission, Luxembourg, Luxembourg, Dec 2018).

Carbogno, Christian: Quantum Materials: Obtaining Insights by Combining High-Throughput First-Principles and Machine-Learning Approaches. (Quantum Materials Workshop, Oak Ridge National Lab, Oak Ridge, TN, USA, Feb 2019).

Carbogno, Christian: Accurate Thermal Conductivities of Complex, Strongly-Anharmonic Solids From First Principles. (2019 MRS Spring Meeting & Exhibit, Phoenix, AZ, USA, Apr 2019).

Carbogno, Christian: Quantum Materials: Obtaining Insights by Combining High-Throughput First-Principles and Machine-Learning Approaches. (IMPRS Workshop: Frontiers of Quantum Materials, Stuttgart, Germany, Jul 2019).

Carbogno, Christian: Electronic Transport in Solids From First Principles. (Hands-On DFT and Beyond: High-throughput screening and big-data analytics, towards exascale computational materials science, Barcelona, Spain, Aug 2019).

Carbogno, Christian: From Harmonic Vibrations to Strongly Anharmonic Heat Transport: Ab Initio Nuclear Dynamics in Solids. (Hands-On DFT and Beyond: High-throughput screening and big-data analytics, towards exascale computational materials science, Barcelona, Spain, Aug 2019).

Draxl, Claudia: Graphene off the Dirac Point. (International Workshop on Multifunctional Dirac Materials, Stockholm, Sweden, Nov 2017).

Draxl, Claudia: Level Alignment at Organic/Inorganic Hybrid Interfaces. (SSNS'18, Symposium on Surface and Nano Science 2018, Furano, Japan, Jan 2018).

Draxl, Claudia: A Significant Raw Material of the 21st Century. (DPG-Frühjahrstagung und EPS-CMD27, Berlin, Germany, Mar 2018).

Draxl, Claudia: From Sharing to Data-Driven Research: The NOMAD Laboratory. (Colloquium, University of Luxembourg, Luxembourg, Luxembourg, Apr 2018).

Draxl, Claudia: Exciting Core Levels. (Colloquium, Abderrahmane Mira University, Béjaïa, Algeria, May 2018).

Draxl, Claudia: HPC Meets Big Data. (Digital Future 2018, Berlin, Germany, May 2018).

Draxl, Claudia: NOMAD: The FAIR Concept for Big-Data Driven Materials Science. (International Workshop, Machine Learning for Quantum Many-body Physics, Max Planck Institute for the Physics of Complex Systems, Dresden, Germany, Jun 2018).

Draxl, Claudia: Probing the LDA-1/2 Method for Solids and Molecules, and as a Starting Point for the GW Approach. (Electronic Structure (ES) '18 and Penn Conference in Theoretical Chemistry (PCTC) '18, Philadelphia, PA, USA, Jun 2018).

Draxl, Claudia: Theory of X-Ray Matter Interactions. (UXSS 2018, Ultrafast X-ray Summer Seminar 2018, Menlo Park, CA, USA, Jun 2018).

Draxl, Claudia: An Introduction to the NOMAD Project. (Workshop on excitations in solids, HoW exciting! 2018, Berlin, Germany, Jul 2018).

Draxl, Claudia: Density-Functional Theory. (Workshop on excitations in solids, HoW exciting! 2018, Berlin, Germany, Jul 2018).

Draxl, Claudia: Raman Spectroscopy. (Workshop on excitations in solids, HoW exciting! 2018, Berlin, Germany, Jul 2018).

Draxl, Claudia: SAM Structures of  $\pi$ -Conjugated Organic Molecules: Impact of Conformation, Packing and Substrate on Their Opto-Electronic Excitations. (Gordon Research Conference, GRC, Electronic Processes in Organic Materials, Barga, Italy, Jul 2018).

Draxl, Claudia: Theoretical Spectroscopy. (CAMD Summer School 2018, Electronic Structure Theory and Materials Design, Elsinore, Denmark, Aug 2018).

Draxl, Claudia: Boosting Materials Science Through Big Data and HPC. (Science at Extreme Scales: Where Big Data Meets Large-Scale Computing Tutorials, Institute for Pure and Applied Mathematics (IPAM), Los Angeles, CA, USA, Sep 2018).

Draxl, Claudia: Making Scientific Data Accessible: The NOMAD Encyclopedia. (NOMAD Summer - A hands-on course on tools for novel-materials discovery, Lausanne, Switzerland, Sep 2018).

Draxl, Claudia: NOMAD: The FAIR Concept for Big-Data-Driven Materials Science. (MRS Webinar, Data-Centric Science for Materials Innovation, Sep 2018).

Draxl, Claudia: How FAIR Are Data Repositories in Materials Science? (Workshop II: HPC and Data Science for Scientific Discovery, Institute for Pure and Applied Mathematics (IPAM), Los Angeles, CA, USA, Oct 2018).

Draxl, Claudia: Theoretical Spectroscopy at Hybrid Interfaces. (ASOMEA - IX, 9th Workshop on Advanced Spectroscopy of Organic Materials for Electronic Applications, Schluchsee, Germany, Oct 2018).

Draxl, Claudia: The NOMAD Encyclopedia and Visualization Tools. (Exciting Shanghai School and Hands-on workshop on density functional theory, First-principles simulations in materials science, Shanghai, China, Nov 2018).

Draxl, Claudia: Theory of Optical and Core-Level Spectroscopy. (Exciting Shanghai School and Hands-on workshop on density functional theory, First-principles simulations in materials science, Shanghai, China, Nov 2018).

Draxl, Claudia: The Forth Paradigm of Materials Science. (Colloquium, Vienna University of Technology, Vienna, Austria, Dec 2018).

Draxl, Claudia: Theoretical Spectroscopy at Interfaces. (Materials - IMX Seminar Series, École polytechnique fédérale de Lausanne (EPFL), Lausanne, Switzerland, Dec 2018).

Draxl, Claudia: Shaping the Forth Paradigm of Materials Science. (Physics Colloquium, University of Innsbruck, Innsbruck, Austria, Jan 2019).

Draxl, Claudia: From Electronic-Structure Theory to Data-Driven Science. (Physics Colloquium, University of Warsaw, Warsaw, Poland, Feb 2019).

Draxl, Claudia: Theoretical Spectroscopy at Hybrid Interfaces – Focus on Solar-Cell Materials. (International Conference on Interfaces in Organic and Hybrid Thin-Film Optoelectronics (INFORM), Valencia, Spain, Mar 2019).

Draxl, Claudia: Shaping the Fourth Paradigm of Materials Science. (Colloquium, Leibniz Institute of Polymer Research Dresden, Dresden, Germany, Apr 2019).

Draxl, Claudia: Making Scientific Data Accessible: The NOMAD Laboratory. (PQ-DFT: workshop on Precision Quantification in DFT, Louvain-la-Neuve, Belgium, May 2019).

Draxl, Claudia: Towards the 4th Paradigm of Materials Science. (Bunsentagung 2019 - 118th General Assembly of the German Bunsen Society for Physical Chemistry, Jena, Germany, May 2019).

Ghiringhelli, Luca M.: Introduction to the NOMAD Analytics Toolkit. (NOMAD Summer - A Hands-on course on tools for novel materials discovery, Berlin, Germany, Sep 2017).

Ghiringhelli, Luca M.: Searching for Materials Descriptors in Huge Feature Spaces With Compressed Sensing. (CECAM Workshop, Big-Data driven Materials Science, Lausanne, Switzerland, Sep 2017).

Ghiringhelli, Luca M.: The NOMAD European Centre of Excellence: Your Gateway to Materials-Science Data. (E-CAM Scoping Workshop: From the Atom to the Material, Cambridge, UK, Sep 2017).

Ghiringhelli, Luca M.: A Meta-Machine-Learning Method for Identifying Effective Descriptors of Materials Properties. (Workshop I: Optimization and Optimal Control for Complex Energy and Property Landscapes, IPAM, Los Angeles, CA, USA, Oct 2017).

Ghiringhelli, Luca M.: Big-Data Driven Materials Science - Critical Role of the Descriptor. (19th Meeting of the Fachbeirat of the Fritz-Haber-Institut, Berlin, Germany, Nov 2017).

Ghiringhelli, Luca M.: Big-Data-Driven Materials Science: Critical Role of the Descriptor. (Colloquium, Institute of Nanotechnology, Karlsruhe Institute of Technology (KIT), Eggenstein-Leopoldshafen, Germany, Jan 2018).

Ghiringhelli, Luca M.: SISSO: A Compressed-Sensing Method for Systematically Identifying Efficient Physical Models of Materials Properties. (Total Energy and Force Methods 2018 Workshop, Cambridge, UK, Jan 2018).

Ghiringhelli, Luca M.: Data-Driven Materials Science: Critical Role of the Descriptor. (Data Science Workshop, Scuola Internazionale Superiore di Studi Avanzati, SISSA, Trieste, Italy, Feb 2018).

Ghiringhelli, Luca M.: Novel Materials Discovery: Big-Data-Analytics Methods and Infrastructure for Building Maps of Materials. (DPG-Frühjahrstagung und EPS-CMD27, Symposium Information Driven Materials Research, Berlin, Germany, Mar 2018).

Ghiringhelli, Luca M.: Novel Materials Discovery: Big-Data-Analytics Methods and Infrastructure for Building Maps of Materials. (Materials Today Webinar, Harnessing the Potential of Open Data in Materials Science, Mar 2018).

Ghiringhelli, Luca M.: Discovering Interpretable Descriptors Through Compressed Sensing. (BiGmax Workshop 2018 on Big-Data-Driven Materials Science, Kloster Irsee, Irsee, Germany, Apr 2018).

Ghiringhelli, Luca M.: Data-Driven Materials Science: Critical Role of the Descriptor. (International Workshop, Machine Learning for Quantum Many-body Physics, Max Planck Institute for the Physics of Complex Systems, Dresden, Germany, Jun 2018).

Ghiringhelli, Luca M.: Data-Driven Materials Science: Methods and Infrastructure for Building Maps of Materials. (Symposium on Materials Genome Towards Exascale, Spetses, Greece, Jun 2018).

Ghiringhelli, Luca M.: Exploiting the New Paradigm: Crowd Sourcing Data-Driven Materials Science. (CECAM Workshop, Machine Learning at Interfaces, EPFL, Lausanne, Switzerland, Jun 2018).

Ghiringhelli, Luca M.: Novel Materials Discovery: Big-Data-Analytics Methods and Infrastructure for Building Maps of Materials. (2018 eSENCE Multiscale Modelling Meeting, Uppsala, Sweden, Jun 2018).

Ghiringhelli, Luca M.: Big-Data Analytics. (Hands-On DFT and Beyond: Frontiers of Advanced Electronic Structure and Molecular Dynamics Methods, Beijing, China, Jul 2018).

Ghiringhelli, Luca M.: Data-Driven Materials' Properties Maps: Methods and Infrastructure. (FHI-aims Developers' and Users' Meeting, Electronic Structure Theory with Numeric Atom-Centered Basis Functions, Munich, Germany, Jul 2018).

Ghiringhelli, Luca M.: Metal and Metal-Oxide Clusters at Realistic Conditions: Beyond the Static, Monostructure Description. (PASC18 Conference, Minisymposia 44 – Emerging Trends in Statistical Mechanics Applications to Nanostructured Materials, Basel, Switzerland, Jul 2018).

Ghiringhelli, Luca M.: Data-Driven Materials Science: Building Maps of Materials Properties. (2018 E-MRS Fall Meeting and Exhibit, Warsaw, Poland, Sep 2018).

Ghiringhelli, Luca M.: Data-Driven Materials Science: Statistical Methods and Infrastructure for Learning Maps of Materials' Properties. (Workshop, Molecular Simulation and Engineering, MolSimEng 2018, Milan, Italy, Sep 2018).

Ghiringhelli, Luca M.: Data-Driven Materials' Properties Maps: Methods and Infrastructure. (Leverhulme Research Centre for Functional Materials Design Inaugural Symposium, Liverpool, UK, Sep 2018).

Ghiringhelli, Luca M.: The NOMAD Analytics Toolkit: Interactive Big-Data Driven Materials Science over the Web. (NOMAD Summer - A hands-on course on tools for novel-materials discovery, Lausanne, Switzerland, Sep 2018).

Ghiringhelli, Luca M.: Data-Driven Maps of Materials' Properties: Methods and Infrastructure. (BIOVIA Materials Studio Community Days, Session: Data-Driven Materials Modelling, Brussels, Belgium, Nov 2018).

Ghiringhelli, Luca M.: HPC for Computationally and Data-Intensive Problems. (SuperMUC-NG Next-Gen Science Symposium, Munich, Germany, Nov 2018).

Ghiringhelli, Luca M.: Data-Driven Cartography of Materials Properties. (Workshop on Crystal Structure Prediction: Exploring the Mendeleev Table as a Palette to Design New Materials, International Centre for Theoretical Physics (ICTP), Trieste, Italy, Jan 2019).

Ghiringhelli, Luca M.: Data-Driven Materials Science: The Critical Role of the Descriptor. (SBP Group Seminar, Physics and Chemistry of Biological Systems - SISSA, Trieste, Italy, Jan 2019).

Ghiringhelli, Luca M.: Topics in Data-Driven Materials Science: Crowd Sourcing and Maps of Material Properties. (PRESTO International Symposium on Materials Informatics, Tokyo, Japan, Feb 2019).

Ghiringhelli, Luca M.: Compressed Sensing for Data-Driven Materials Science. (BIGmax Workshop 2019 on Big-data-driven Materials Science, Dresden, Germany, Apr 2019).

Ghiringhelli, Luca M.: Data-Driven Materials Science: The Critical Role of the Descriptor. (Seminar, University of Warwick, Coventry, UK, May 2019).

Ghiringhelli, Luca M.: Exploring the Materials Space via Regularized and Symbolic Regression. (ML4MS 2019, Young Researcher's Workshop on Machine Learning for Materials Science 2019, Aalto University, Helsinki, Finland, May 2019).

Ghiringhelli, Luca M.: Metadata Towards FAIR Data Sharing for Data-Driven Materials Science: Achievements and Open Challenges. (FHI-Workshop on Current Research Topics at the FHI, Potsdam, Germany, May 2019).

Ghiringhelli, Luca M.: Charting Materials-Properties Spaces. (E-MRS Spring Meeting 2019, SYMPOSIUM AA: Computations for materials discovery, design and the role of data, Nice, France, Jun 2019).

Ghiringhelli, Luca M.: Charting Spaces of Materials Properties. (CPMD Meeting 2019 - Pushing the Boundaries of Molecular Dynamics, Lausanne, Switzerland, Jul 2019).

Ghiringhelli, Luca M.: Metadata Towards FAIR Data Sharing for Data-Driven Materials Science: Achievements and Open Challenges. (Shared metadata and data formats for Big-Data Driven Materials Science: A NOMAD-FAIRDI workshop, Berlin, Germany, Jul 2019).

Goldsmith, Bryan: Finding Patterns, Correlations, and Descriptors in Materials Data Using Subgroup Discovery and Compressed Sensing. (255th ACS National Meeting & Exposition, Nexus of Food, Energy & Water, New Orleans, LA, USA, Mar 2018).

Hermann, Jan: Modeling Van Der Waals Interactions in Materials with Many-Body Dispersion. (FHI-aims Developers' and Users' Meeting, Electronic Structure Theory with Numeric Atom-Centered Basis Functions, Munich, Germany, Jul 2018).

Langer, Marcel Florin: Representing Molecules and Materials for Accurate Interpolation of Quantum-Mechanical Calculations. (DPG Spring Meeting, Regensburg, Germany, Mar 2019).

Levchenko, Sergey V.: Big-Data Analytics for High-Throughput Computational Materials Design: Methods and Applications. (3rd International Conference on Electronic Structure Theory for Accelerated Development of Materials: A New Tool in Materials Science, Moscow, Russia, Oct 2017).

Levchenko, Sergey V.: Catalytic Carbon Dioxide Activation on Metal Oxide Surfaces. (Energy Colloquium, Skoltech, Skolkovo Institute of Science and Technology, Moscow, Russia, Feb 2018).

Levchenko, Sergey V.: Formation and Properties of Small Polarons from First Principles. (FHI-Workshop on Current Research Topics at the FHI, Potsdam, Germany, May 2018).

Levchenko, Sergey V.: Ab Initio Thermodynamics. (Hands-On DFT and Beyond: Frontiers of Advanced Electronic Structure and Molecular Dynamics Methods, Beijing, China, Aug 2018).

Levchenko, Sergey V.: Periodic System. (Hands-On DFT and Beyond: Frontiers of Advanced Electronic Structure and Molecular Dynamics Methods, Beijing, China, Aug 2018).

Levchenko, Sergey V.: Bridging the Pressure and Materials Gaps: First-Principles Modelling of Complex Materials. (FHI-JST Joint Symposium, Current Topics and Challenges for Innovative Catalysts, Berlin, Germany, Oct 2018).

Levchenko, Sergey V.: Ab Initio Data Analytics Approaches to Catalyst Design. (Electronic Materials and Applications 2019 (EMA 2019), Orlando, FL, USA, Jan 2019).

Levchenko, Sergey V.: Ab Initio Thermodynamics. (Hands-On DFT and Beyond: High-throughput screening and big-data analytics, towards exascale computational materials science, Barcelona, Spain, Aug 2019).

Levchenko, Sergey V.: Global Effects of Doping on Surface and Interface Properties: An Ab Initio Study. (INESS-2019, The 7th International Conference on Nanomaterials and Advanced Energy Storage Systems, Almaty, Kazakhstan, Aug 2019).

Levchenko, Sergey V.: Periodic Systems: Concepts and Numeric Atom-Centered Orbitals. (Hands-On DFT and Beyond: High-throughput screening and big-data analytics, towards exascale computational materials science, Barcelona, Spain, Aug 2019).

Mazheika, Aliaksei: Carbon Dioxide Activation at Metal-Oxide Surfaces: A Compressed-Sensing Analysis. (DPG-Frühjahrstagung und EPS-CMD27, Gerhard Ertl Young Investigator Award, Berlin, Germany, Mar 2018).

Mazheika, Aliaksei: Machine Learning of CO<sub>2</sub> Activation on Metal-Oxide Semiconductor Surfaces. (Seminar, University of Barcelona, Barcelona, Spain, Jul 2018).

Mazheika, Aliaksei: CO<sub>2</sub> Activation on Semiconductor Oxide Surfaces: Machine Learning Study. (Colloquium, Technische Universität Berlin, Berlin, Germany, Aug 2018).

Mazheika, Aliaksei: Subgroup Discovery Meets Compressed Sensing: Application to CO<sub>2</sub> Activation Problem. (Theory Department Workshop 2018 at Ringberg Castle, Kreuth, Germany, Sep 2018).

Mazheika, Aliaksei: Coming to Data Analytics in Surface Science: CO<sub>2</sub> Activation on Semiconductor Oxides. (BasCat - UniCat BASF Joint Lab, Technische Universität Berlin, Berlin, Germany, Nov 2018).

Ouyang, Runhai: SISSO: A Compressed-Sensing Method for Systematically Identifying Efficient Physical Models of Materials Properties. (NOMAD Summer - A hands-on course on tools for novel-materials discovery, CECAM, Lausanne, Switzerland, Sep 2018).

Raimbault, Nathaniel: Anharmonic Raman Spectra of Molecular Crystals from DFPT. (FHI-aims Developers' and Users' Meeting, Electronic Structure Theory with Numeric Atom-Centered Basis Functions, Munich, Germany, Jul 2018).

Rossi, Mariana: Ab-Initio Simulations of Hydrogen-Bonded Systems. (IMPRS Fall Block Course 2017, New Concepts in Experimental and Theoretical Surface Science, Berlin, Germany, Sep 2017).

Rossi, Mariana: Anharmonic and Quantum Effects in (Bio)organic Systems With First-Principles Accuracy. (Total Energy and Force Methods 2018 Workshop, Selwyn College, Cambridge, MA, USA, Jan 2018).

Rossi, Mariana: First Principles Approaches to the Structure and Dynamics of Bio-Organic Interfaces. (FHI Theory Symposium, Fritz Haber Institute, Berlin, Germany, Apr 2018).

Rossi, Mariana: Towards an Accurate, High-Throughput Framework for the Prediction of Anharmonic Free Energies in Molecular Crystals. (BiGmax Workshop 2018 on Big-Data-Driven Materials Science, Kloster Irsee, Irsee, Germany, Apr 2018).

Rossi, Mariana: First Principles Approaches to the Structure and Dynamics of Hybrid Interfaces. (Seminar, Department of Physics and Atmospheric Science, Dalhousie University, Halifax, NS, Canada, May 2018).

Rossi, Mariana: Ab Initio Molecular Dynamics in Weakly Bonded Systems. (FHI-aims Developers' and Users' Meeting, Electronic Structure Theory with Numeric Atom-Centered Basis Functions, Munich, Germany, Jul 2018).

Rossi, Mariana: First Principles Approaches to the Structure and Dynamics of Hybrid Interfaces. (Seminar, Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany, Jul 2018).

Rossi, Mariana: Path Integral Based Approximations to Quantum Dynamics for High Dimensional Systems. (High Dimensional Quantum Dynamics (HDQD) Workshop, Lille, France, Aug 2018).

Rossi, Mariana: Quantum Electrons and Nuclei in Atomistic Simulations of Organic (and Inorganic) Systems. (Seminar, Theoretische Chemie, Georg-August-Universität Göttingen, Göttingen, Germany, Aug 2018).

Rossi, Mariana: Addressing the Structure and Dynamics of Weakly Bonded, Flexible Systems. (COMDI 2018, International Workshop on Computational Design and Discovery of Novel Materials, Lausanne, Switzerland, Sep 2018).

Rossi, Mariana: Addressing the Structure and Dynamics of Organic Molecules at Inorganic Interfaces. (FHI-JST Joint Symposium, Current Topics and Challenges for Innovative Catalysts, Berlin, Germany, Oct 2018).

Rossi, Mariana: Critical Interplay of Atomic Structure and Level Alignment in Organic/Inorganic Interfaces. (ASOMEA - IX, 9th Workshop on Advanced Spectroscopy of Organic Materials for Electronic Applications, Schluchsee, Germany, Oct 2018).

Rossi, Mariana: Addressing the Nuclear and Electronic Structure of Weakly Bonded Interfaces. (CMS 2019, 6th Computational Molecular Science Meeting, Warwick, UK, Mar 2019).

Rossi, Mariana: Nuclear Quantum Effects in Weakly Bonded Systems. (Seminar, Department of Chemistry, University of Warwick, Coventry, UK, Mar 2019).

Rossi, Mariana: Addressing the Structure and Dynamics of Weakly-Bonded Interfaces. (Symposium, Frontiers of Electronic-Structure Theory: Focus on the Interface Challenge, DPG-Frühjahrstagung 2019, Regensburg, Germany, Apr 2019).

Rossi, Mariana: Structure and Dynamics of Weakly Bonded Systems. (TYC Discussion Meeting: Quantum Chemistry Techniques, Thomas Young Centre (TYC), London, UK, Apr 2019).

Rossi, Mariana: Addressing the Nuclear and Electronic Structure of Weakly Bonded Interfaces. (IFT/ICTP-SAIFR Colloquium Series - 2019, Universidade Estadual Paulista, UNESP, São Paulo, Brazil, May 2019).

Rossi, Mariana: Addressing the Nuclear and Electronic Structure of Weakly Bonded Interfaces. (Materials Science Colloquium, SFB 986, Tailor-Made Multi-Scale Materials Systems, Hamburg, Germany, May 2019).

Rossi, Mariana: Nuclear Quantum Effects in Weakly Bonded Systems. (Recent developments in quantum dynamics, an E-CAM state-of-the-art workshop, Lyon, France, Jun 2019).

Rossi, Mariana: Anharmonic Nuclear Quantum Effects and Their Interplay With the Electronic Structure of Weakly Bonded Systems. (ISTCP 2019, 10th Triennial Congress of the International Society for Theoretical Chemical Physics, Tromsø, Norway, Jul 2019).

Rossi, Mariana: Structure and Dynamics of Molecular Crystals. (CPMD Meeting 2019 - Pushing the Boundaries of Molecular Dynamics, Lausanne, Switzerland, Jul 2019).

Rupp, Matthias: Interpolation of Electronic Structure Calculations via Machine Learning. (International Workshop on Atomic Physics 2017, Max Planck Institute for the Physics of Complex Systems, Dresden, Germany, Dec 2017).

Rupp, Matthias: Machine Learning for Interpolation of Electronic Structure Calculations. (Seminar, Department of Thin Films and Nanostructures, Institute of Physics of the Czech Academy of Sciences, Prague, Czech Republic, Dec 2017).

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Scheffler, Matthias: When More Data Do Not Provide a Better Description. (Workshop II: HPC and Data Science for Scientific Discovery, Institute for Pure and Applied Mathematics (IPAM), Los Angeles, CA, USA, Oct 2018).

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Qiu, H., H. Kuhlenbeck, E. Bauer and H.-J. Freund: Gold-Decorated Biphasic  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>(0001): Activation by CO-Induced Surface Reduction. *The Journal of Physical Chemistry C* **123** (13), 8221–8227 (2019).

Richter, N.F., F.E. Feiten, J. Pal, A. Plucienik, E. Emmez, S.K. Shaikhutdinov, H. Kuhlenbeck, T. Risse, H.-J. Freund, I. Goikoetxea, R. Włodarczyk and J. Sauer: Characterization of Phonon Vibrations of Silica Bilayer Films. *The Journal of Physical Chemistry C* **123** (12), 7110–7117 (2019).  
Oxidation of Reduced Ceria

Ronneburg, H., M. Trapp, R. Cubitt, L. Silvi, S. Cap, M. Ballauff and S. Risse: Surface structure inhibited lithiation of crystalline silicon probed with operando neutron reflectivity. *Energy Storage Materials* **18**, 182–189 (2019).

Schmidt, T., H. Marchetto, U. Groh, R.H. Fink and E. Umbach: Complex Monolayer Growth Dynamics of a Highly Symmetric Molecule: NTCDA on Ag(111). *The Journal of Physical Chemistry C* **123** (13), 8244–8255 (2019).  
Oxidation of Reduced Ceria

Sorek, E., J. Ankri, G. Arbiv, R. Mol, I. Popov, H.-J. Freund, S.K. Shaikhutdinov and M. Asscher: Acetylene Reactivity on Pd-Cu Nanoparticles Supported on Thin Silica Films: The Role of the Underlying Substrate. *The Journal of Physical Chemistry C* **123** (28), 17425–17431 (2019).

Sterr, M., N. Nilius, S.K. Shaikhutdinov, M. Heyde, T. Schmidt and H.-J. Freund: Interaction of water with oxide thin film model systems. *Journal of Materials Research* **34** (3), 360–378 (2019).

Weng, X., Y. Cui, S.K. Shaikhutdinov and H.-J. Freund: CO<sub>2</sub> Adsorption on CaO(001): Temperature-Programmed Desorption and Infrared Study. *The Journal of Physical Chemistry C* **123** (3), 1880–1887 (2019).

Zaki, E., Z. Jakub, F. Mirabella, G.S. Parkinson, S.K. Shaikhutdinov and H.-J. Freund: Water Ordering on the Magnetite Fe<sub>3</sub>O<sub>4</sub> Surfaces. *The Journal of Physical Chemistry Letters* **10** (10), 2487–2492 (2019). 

## Invited talks by members of the Department of Chemical Physics

Burson, Kristen: Amorphous Materials: From Two-Dimensional Glass to Bubble Rafts. (Seminar, Mount Holyoke College, South Hadley, MA, USA, Apr 2018).

Freund, Hans-Joachim: Models for Heterogeneous Catalysts: Complex Materials at the Atomic Level. (Plenary Talk, XVI Brazil MRS Meeting, Brazilian Materials Research Society (SBPMat), Gramado, Brazil, Sep 2017).

Freund, Hans-Joachim: Thin Oxide Films as Templates in Catalysis. (Workshop Surface Science Rio (WS2Rio), Rio de Janeiro, Brazil, Nov 2017).

Freund, Hans-Joachim: The Surface Science of Catalysis and More, Using Ultrathin Oxide Films as Templates: A Perspective. (ALC '17, 11th International Symposium on Atomic Level Characterizations for New Materials and Devices '17, Kauai, HI, USA, Dec 2017).

Freund, Hans-Joachim: Can Single Crystal Model Catalyst Tell Us About Real Powder Catalysts? (Workshop on Surface Science and Catalysis, Hefei, China, Jan 2018).

Freund, Hans-Joachim: Models for Heterogeneous Catalysts: Complex Materials at the Atomic Level. (International Conference on Catalysis and Surface Chemistry, Kraków, Poland, Mar 2018).

Freund, Hans-Joachim: Model Systems for Heterogeneous Catalysts at the Atomic Level. (Seminar, Institute of Physics, Czech Academy of Sciences, Prague, Czech Republic, Apr 2018).

Freund, Hans-Joachim: Engineering Surfaces at the Atomic Level. (43rd David M. Mason Lecture Series, Stanford, CA, USA, May 2018).

Freund, Hans-Joachim: Model Systems in Heterogeneous Catalysis: Towards the Design and Understanding of Structure and Electronic Properties. (Faraday Discussion, Designing Nanoparticle Systems for Catalysis, London, UK, May 2018).

Freund, Hans-Joachim: Adsorption on Oxide Surfaces and Supported Metal Particles. (Summer School, VI San Luis School and Conference on Surfaces, Interfaces and Catalysis, Santa Fe, Argentina, Jun 2018).

Freund, Hans-Joachim: Development of Instrumentation for, and Novel Concepts in Model Catalysis. (VI San Luis School and Conference on Surfaces, Interfaces and Catalysis, Santa Fe, Argentina, Jun 2018).

Freund, Hans-Joachim: Models for Heterogeneous Catalysts: Complex Materials at the Atomic Level. (NaWi Graz Physics Colloquium, Institute of Physics, University of Graz, Graz, Austria, Jun 2018).

Freund, Hans-Joachim: The Structure of Oxide Surfaces and Its Interaction with Water. (ICTAC 17, International Conference on Theoretical Aspects of Catalysis, Los Angeles, CA, USA, Jun 2018).

Freund, Hans-Joachim: Can Single Crystal Model Catalyst Based on Thin Oxide Films Tell Us About Real Powder Catalyst. (IUVSTA-86-ASEVA-28 Workshop, Nanoscale Oxides Systems in Physics and Chemistry, Avila, Spain, Jul 2018).

Freund, Hans-Joachim: Model Systems for Heterogeneous Catalysts at the Atomic Level. (2018 International Symposium on Advancement and Prospect of Catalysis Science & Technology, Sydney, NSW, Australia, Jul 2018).

Freund, Hans-Joachim: Surface Chemical Reactions on Top and Below Oxide Films: Chemistry in Open and Confined Space. (International Symposium on Relations between Homogeneous and Heterogeneous Catalysis, ISHHC18, Sydney, NSW, Australia, Jul 2018).

Freund, Hans-Joachim: Thin Films as Model Materials in Catalysis: A Perspective at the Atomic Level. (256th ACS National Meeting & Exposition, Nanoscience, Nanotechnology & Beyond, New Vistas in Heterogeneous Catalysis: Symposium in honor of Robert Grasselli, Boston, MA, USA, Aug 2018).

Freund, Hans-Joachim: Instrumentation Development and Novel Concepts in Model Catalysis. (10th Brazilian / German Workshop on Applied Surface Science, Bad Dürkheim, Germany, Sep 2018).

Freund, Hans-Joachim: Model Catalysts at the Atomic Level Studied by Advanced Experimental Techniques in Surface Science. (Lecture, Computational Spectroscopy: Bridging Theory and Experiment, Lake Como School of Advanced Studies, Como, Italy, Sep 2018).

Freund, Hans-Joachim: Model Systems in Heterogeneous Catalysts at the Atomic Level. (FHI-JST Joint Symposium, Current Topics and Challenges for Innovative Catalysts, Berlin, Germany, Oct 2018).

Freund, Hans-Joachim: Models for Heterogeneous Catalysts: Complex Materials at the Atomic Level. (ACSIN-14 & ICSPM26, 14th International Conference on Atomically Controlled Surfaces, Interfaces and Nanostructures & 26th International Colloquium on Scanning Probe Microscopy, Sendai, Japan, Oct 2018).

Freund, Hans-Joachim: Model Catalyst Design: A Materials Science Perspective at the Atomic Level. (Frontiers of Science Symposium FAPESP - Max Planck, São Paulo, Brazil, Nov 2018).

Freund, Hans-Joachim: Model Systems in Heterogeneous Catalysis: Surface Chemistry of Complex Systems at the Atomic Level. (10th Singapore International Chemistry Conference, SICC-10, Singapore, Singapore, Dec 2018).

Freund, Hans-Joachim: Model Catalysts Design: A Material Science Perspective at the Atomic Level. (Cardiff Catalysis Institute Conference, Cardiff, UK, Jan 2019).

Freund, Hans-Joachim: 40 Years of Surface Science – An Overview. (Symposium, 40 Years of Surface Science, Erlangen, Germany, Feb 2019).

Freund, Hans-Joachim: Models for Heterogeneous Catalysts: Complex Materials at the Atomic Level. (ACS Award in Surface Chemistry: Symposium in Honor of Hajo Freund, 257th ACS National Meeting & Exposition, Chemistry for New Frontiers, Orlando, FL, USA, Mar 2019).

Freund, Hans-Joachim: Models for Strong Metal Support Interaction (SMSI): From Structure (Geometric and Electronic) to Reactivity. (Ipatieff Prize: Symposium in Honor of Ivo Hermans, 257th ACS National Meeting & Exposition, Chemistry for New Frontiers, Orlando, FL, USA, Mar 2019).

Freund, Hans-Joachim: Oxide Surfaces and Its Interaction With Water. (Symposium Catalytic Chemistry over Metal Oxides, 257th ACS National Meeting & Exposition, Chemistry for New Frontiers, Orlando, FL, USA, Mar 2019).

Freund, Hans-Joachim: Can Single Crystal Model Catalyst Based on Thin Oxide Films Tell Us About Real Powder Catalysts? (CAS Distinguished International Professorship, Suzhou, China, May 2019).

Freund, Hans-Joachim: Can Single Crystal Model Catalysts Tell Us Something About Powder Catalysts? (CAS Distinguished International Professorship, Dalian, China, May 2019).

Freund, Hans-Joachim: Can Single Crystal Model Catalysts Tell Us Something About Powder Catalysts? (CAS Distinguished International Professorship, Beijing, China, May 2019).

Freund, Hans-Joachim: Model Systems for Heterogeneous Catalysts at the Atomic Level. (CAS Distinguished International Professorship, Hefei, China, May 2019).

Freund, Hans-Joachim: Model Systems for Heterogeneous Catalysts at the Atomic Level. (Seminar on Chemical Bond Activation, Carl von Ossietzky Universität Oldenburg, Oldenburg, Germany, May 2019).

Freund, Hans-Joachim: Models for Heterogeneous Catalysts: Complex Materials at the Atomic Level. (Irsee VIII Symposium, Kloster Irsee, Irsee, Germany, May 2019).

Freund, Hans-Joachim: Surface Action Spectroscopy With Inert Gas Messenger Atoms. (CAS Distinguished International Professorship, National Synchrotron Radiation Laboratory (NSRL), Hefei, China, May 2019).

Freund, Hans-Joachim: Model Systems for Heterogeneous Catalysts in Confined Space at the Atomic Scale. (NAM26, 2019 North American Catalysis Society Meeting, Chicago, IL, USA, Jun 2019).

Freund, Hans-Joachim: Model Systems for Heterogeneous Catalysts at the Atomic Level. (Colloquium, Technische Universität Clausthal, Clausthal-Zellerfeld, Germany, Jul 2019).

Freund, Hans-Joachim: Model Systems for Heterogeneous Catalysts at the Atomic Level. (IVC-21, 21th International Vacuum Congress, Malmö, Sweden, Jul 2019).

Freund, Hans-Joachim: Can Single Crystal Model Catalyst Based on Thin Oxide Films Tell Us About Real Powder Catalysts? (Procter & Gamble, Cincinnati, OH, USA, Aug 2019).

Freund, Hans-Joachim: From Crystals to Glasses at the Atomic Level in Real Space. (258th ACS National Meeting & Exposition, Symposium in Honor of Francisco Zaera & Yuriy Román-Leshkov, San Diego, CA, USA, Aug 2019).

Freund, Hans-Joachim: Interaction of H<sub>2</sub> and CO<sub>2</sub> With Ceria Surfaces: Adsorption and Absorption. (258th ACS National Meeting & Exposition, Advances in Catalysis with Ceria & Other Reducible Oxides, San Diego, CA, USA, Aug 2019).

Freund, Hans-Joachim: Reactions in Confined Space: Can Surface Science Contribute? (258th ACS National Meeting & Exposition, Symposium in Honor of Charles T. Campbell, San Diego, CA, USA, Aug 2019).

Heyde, Markus: Looking into the Structure of Glass by Designing a New 2D Material. (Dutch SPM Day 2017, Dutch Scanning Probe Microscopy Day, Leiden University, Leiden, Netherlands, Nov 2017).

Heyde, Markus: Looking Into the Structure of Glass Films. (FHI-JST Joint Symposium, Current Topics and Challenges for Innovative Catalysts, Berlin, Germany, Oct 2018).

Heyde, Markus: Looking Into the Atomic Structure of Glass Films. (257th ACS National Meeting & Exposition, Chemistry for New Frontiers, Orlando, FL, USA, Mar 2019).

Junkes, Heinz: Data Acquisition With EPICS, on the Example of Cryvisil. (CP-Workshop of the Fritz Haber Institute, Templin, Germany, Sep 2017).

Junkes, Heinz: Archiving EPICS 7 PVs. (EPICS Collaboration Meeting at ICALEPCS 2017, Barcelona, Spain, Oct 2017).

Junkes, Heinz: Blockchain im Gesundheitswesen. (34. DV-Treffen der Max-Planck-Institute, Göttingen, Germany, Oct 2017).

Junkes, Heinz: IT am Fritz-Haber-Institut. (FDM Workshop 2018, MPG Forschungsdatenmanagement Workshop, Munich, Germany, Apr 2018).

Junkes, Heinz: A Bee-Balance. (EPICS Collaboration Meeting, Lemont, IL, USA, Jun 2018).

Junkes, Heinz: RTEMSS and EPICS7. (EPICS Collaboration Meeting, Lemont, IL, USA, Jun 2018).

Junkes, Heinz: IT at the FHI "Enabling Science." (CP-Workshop at Ringberg Castle, Department of Chemical Physics, FHI, Kreuth, Germany, Sep 2018).

Junkes, Heinz: EPICS with RTEM 5. (EPICS Collaboration Meeting, Melbourne, VIC, Australia, Nov 2018).

Junkes, Heinz: Native BACnet - Vom Anspruch zur Wirklichkeit. (Zukunftstage, Smart Buildings@Internet of Things, Berlin, Germany, Nov 2018).

Kuhlenbeck, Helmut: Surface Action Spectroscopy with Rare Gas Messenger Atoms. (19th Meeting of the Fachbeirat of the Fritz-Haber-Institut, Berlin, Germany, Nov 2017).

Kuhlenbeck, Helmut: Surface Action Spectroscopy with Rare Gas Messenger Atoms. (Free-Electron Laser Workshop, Applications of IR Free-Electron Lasers: Latest Developments and Future Directions at Ringberg Castle, Kreuth, Germany, Jan 2018).

Kuhlenbeck, Helmut: Surface Action Spectroscopy – A New Tool for the Spectroscopy of Surface Vibrations. (ACS Award in Surface Chemistry: Symposium in Honor of Hajo Freund, 257th ACS National Meeting & Exposition, Chemistry for New Frontiers, Orlando, FL, USA, Mar 2019).

Pan, Qiushi: Mechanistic Insight into Heterogeneous Catalytic Reactions on Supported Catalysts. (Special seminar, Department of Inorganic Chemistry, Fritz Haber Institute, Berlin, Germany, Jan 2018).

Prieto, Mauricio: Real Time Observation of Chemical Reactions in Confined Space. (FHI-Workshop on Current Research Topics at the FHI, Potsdam, Germany, May 2018).

Schmidt, Thomas: Ultrathin Silica Films Studied by LEEM and XPEEM. (SpectroNanoscopia 2017 workshop, Seoul, South Korea, Sep 2017).

Schneider, Wolf-Dieter: STM-Induced Light Emission from Metals, Insulators, Supported Molecules, and Semiconductors: How Local Is «Local»? (CP-Workshop of the Fritz Haber Institute, Templin, Germany, Sep 2017).

Schneider, Wolf-Dieter: A Small World. (JSPS 141 Committee Award Lectures, ALC '17 11th International Symposium on Atomic Level Characterizations for New Materials and Devices '17, Kauai, HI, USA, Dec 2017).

Schneider, Wolf-Dieter: The Atomic Structure of a Crystalline Metal-Supported Ultrathin Germanium Film. (SSNS'18, Symposium on Surface and Nano Science 2018, Furano, Japan, Jan 2018).

Schneider, Wolf-Dieter: From Adatoms to Superlattices: Sensing the Spin of Individual Ce-Atoms. (3S'18, 31. Symposium on Surface Science 2018, St. Christoph am Arlberg, Austria, Feb 2018).

Schneider, Wolf-Dieter: From Adatoms to Superlattices: Sensing the Spin of Individual Ce-Atoms. (From Solid State to BioPhysics IX: From Basic to Life Sciences, Cavtat, Croatia, Jun 2018).

Schneider, Wolf-Dieter: From Adatoms to Superlattices: Sensing the Spin of Individual Ce-Atoms. (ICN+T 2018, International Conference on Nanoscience + Technology, Brno, Czech Republic, Jul 2018).

Schneider, Wolf-Dieter: A Small World: From Single Magnetic Adatoms to Two-Dimensional Kondo Lattices. (CP-Workshop at Ringberg Castle, Department of Chemical Physics, FHI, Kreuth, Germany, Sep 2018).

Schneider, Wolf-Dieter: From Adatoms to Two-Dimensional Kondo Lattices: Sensing the Spin of Individual Ce-Atoms. (10th Brazilian / German Workshop on Applied Surface Science, Bad Dürkheim, Germany, Sep 2018).

Schneider, Wolf-Dieter: Spin Excitation Spectroscopy Reveals the Magnetic Moment of a Single Ce Adatom. (SSNS'19, Symposium on Surface and Nano Science 2019, Shizuoka, Japan, Jan 2019).

Schneider, Wolf-Dieter: How C.P. Lutz Opened the Window to Nanoscale Surface Physics. (Laudatio, Verleihung der Ehrendoktorwürde an Christopher Paul Lutz, IBM Almaden Research Center, Fakultät für Physik der Universität Regensburg, Regensburg, Germany, Apr 2019).

Schneider, Wolf-Dieter: Chiral Supermolecular Self-Assembly: Scanning Probe Microscopy and Modeling. The Decisive Contributions of Alessandro De Vita. (International Symposium, Alessandro De Vita: A Celebration of his Life and Work, London, UK, May 2019).

Schneider, Wolf-Dieter: Spin Excitation Spectroscopy Reveals the Magnetic Moment of a Single Ce Adatom. (Superstripes 2019, Quantum physics in Complex Matter: Superconductivity, Magnetism and Ferroelectricity, Ischia, Italy, Jun 2019).

Schneider, Wolf-Dieter: Sensing the Spin of a Spectroscopically Dark Ce Adatom With a Scanning Tunneling Microscope. (IBS Conference on Surface Atomic Wires, Pohang, South Korea, Aug 2019).

Shaikhutdinov, Shamil K.: Ultrathin Oxide Films on Metal Substrates: Structure and Properties. (Seminar, University of Padova, Padua, Italy, Oct 2017).

Shaikhutdinov, Shamil K.: Catalysis Driven by 2D-Oxide Films. (IUVSTA-86-ASEVA-28 Workshop, Nanoscale Oxides Systems in Physics and Chemistry, Avila, Spain, Jul 2018).

## Publications based on research using the FHI-FEL

### Late 2017

*Mucha, E., A.I. Gonzalez Florez, M. Marianski, D. Thomas, W. Hoffmann, W.B. Struwe, H.S. Hahm, S. Gewinner, W. Schöllkopf, P.H. Seeberger, G. von Helden and K. Pagel:* Glycan Fingerprinting via Cold-Ion Infrared Spectroscopy. *Angewandte Chemie International Edition* 56 (37), 11248–11251 (2017).

*Seo, J., S. Warnke, K. Pagel, M.T. Bowers and G. von Helden:* Infrared spectrum and structure of the homochiral serine octamer-dichloride complex. *Nature Chemistry* 9, 1263–1268 (2017).

*Song, X., M.R. Fagiani, S. Debnath, M. Gao, S. Maeda, T. Taketsugu, S. Gewinner, W. Schöllkopf, K.R. Asmis and A. Lyalin:* Excess charge driven dissociative hydrogen adsorption on  $\text{Ti}_2\text{O}_4^-$ . *Physical Chemistry Chemical Physics* 19 (17), 23154–23161 (2017).

*Vanbuel, J., E.M. Fernández, P. Ferrari, S. Gewinner, W. Schöllkopf, L.C. Balbás, A. Fielicke and E. Janssens:* Hydrogen Chemisorption on Singly Vanadium-Doped Aluminum Clusters. *Chemistry – A European Journal* 23 (62), 15638–15643 (2017).

*Wu, Z., A. Plucienik, F.E. Feiten, M. Naschitzki, W. Wachsmann, S. Gewinner, W. Schöllkopf, V. Staemmler, H. Kühlenbeck and H.-J. Freund:* Vibrational action spectroscopy of solids: New surface-sensitive technique. *Physical Review Letters* 119 (13), 136101 (2017).

### 2018

*Debnath, S., H. Knorke, W. Schöllkopf, S. Zhou, K.R. Asmis and H. Schwarz:* Experimental Identification of the Active Site in the Heteronuclear Redox Couples  $[\text{AlVO}_x]^+/\text{CO}/\text{N}_2\text{O}$  ( $x=3, 4$ ) by Gas-Phase IR Spectroscopy. *Angewandte Chemie International Edition* 57 (25), 7448–7452 (2018).

*Esser, T.K., H. Knorke, K.R. Asmis, W. Schöllkopf, Q. Yu, C. Qu, J.M. Bowman and M. Kaledin:* Deconstructing Prominent Bands in the Terahertz Spectra of  $\text{H}_7\text{O}_3^+$  and  $\text{H}_9\text{O}_4^+$ : Intermolecular Modes in Eigen Clusters. *The Journal of Physical Chemistry Letters* 9 (4), 798–803 (2018).

*Green, A.E., J. Justen, W. Schöllkopf, A.S. Gentleman, A. Fielicke and S.R. Mackenzie:* IR Signature of Size-Selective  $\text{CO}_2$  Activation on Small Platinum Cluster Anions,  $\text{Pt}_n^-$  ( $n=4-7$ ). *Angewandte Chemie International Edition* 57 (45), 14822–14826 (2018).

*Hoffmann, W., K. Folmert, J. Moschner, X. Huang, H. von Berlepsch, B. Koks, M.T. Bowers, G. von Helden and K. Pagel:* NFGAIL Amyloid Oligomers: The Onset of Beta-Sheet Formation and the Mechanism for Fibril Formation. *Journal of the American Chemical Society* 140 (1), 244–249 (2018).

*Jia, M., J. Vanbuel, P. Ferrari, E.M. Fernández, S. Gewinner, W. Schöllkopf, M.T. Nguyen, A. Fielicke and E. Janssens:* Size Dependent  $\text{H}_2$  Adsorption on  $\text{Al}_n\text{Rh}^+$  ( $n = 1-12$ ) Clusters. *The Journal of Physical Chemistry C* 122 (22), 18247–18255 (2018).

*Kießling, R., W.B. Colson, S. Gewinner, W. Schöllkopf, M. Wolf and A. Paarmann:* Femtosecond single-shot timing and direct observation of subpulse formation in an infrared free-electron laser. *Physical Review Accelerators and Beams* 21 (08), 080702 (2018).

*Kießling, R., S. Gewinner, A. Paarmann, W. Schöllkopf and M. Wolf:* Synchronized Mid-Infrared Pulses at the Fritz Haber Institute IR-FEL. In: *Proceedings of the 38th International Free-*

Electron Laser Conference. (Eds.): K. Bishofberger, B. Carlsten, and V.R.W. Schaa. JACoW, Geneva, 188–191 (2018). ISBN 978-3-95450-179-3.

*Mucha, E., M. Lettow, M. Marianski, D. Thomas, W.B. Struwe, D.J. Harvey, G. Meijer, P.H. Seeberger, G. von Helden and K. Pagel: Fucose Migration in Intact Protonated Glycan Ions: A Universal Phenomenon in Mass Spectrometry. Angewandte Chemie International Edition 57 (25), 7440–7443 (2018).*

*Mucha, E., M. Marianski, F.-F. Xu, D. Thomas, G. Meijer, G. von Helden, P.H. Seeberger and K. Pagel: Unravelling the structure of glycosyl cations via cold-ion infrared spectroscopy. Nature Communications 9, 4174 (2018).*

*Paßler, N., I. Rzdolski, S. Gewinner, W. Schöllkopf, S. De Liberato, C. Gubbin, J. Caldwell, M. Wolf and A. Paarmann: Nonlinear Response and Strong Coupling of Surface Phonon Polaritons. In: Quantum Nano-Photonics. (Eds.): B. Di Bartolo, L. Silvestri, M. Cesaria, and J. Collins. (NATO Science for Peace and Security Series B: Physics and Biophysics). Springer, Dordrecht, 425–426 (2018). ISBN 978-94-024-1543-8.*

*Scutelnic, V., M.A.S. Perez, M. Marianski, S. Warnke, A. Gregor, U. Rothlisberger, M.T. Bowers, C. Baldauf, G. von Helden, T.R. Rizzo and J. Seo: The Structure of the Protonated Serine Octamer. Journal of the American Chemical Society 140 (24), 7554–7560 (2018).*

*Seo, J., W. Hoffmann, S. Malerz, S. Warnke, M.T. Bowers, K. Pagel and G. von Helden: Side-chain effects on the structures of protonated amino acid dimers: A gas-phase infrared spectroscopy study. International Journal of Mass Spectrometry 429, 115–120 (2018).*

*Thomas, D., M. Marianski, E. Mucha, G. Meijer, M.A. Johnson and G. von Helden: Ground-State Structure of the Proton-Bound Formate Dimer by Cold-Ion Infrared Action Spectroscopy. Angewandte Chemie International Edition 57 (33), 10615–10619 (2018).*

*Thomas, D., E. Mucha, S. Gewinner, W. Schöllkopf, G. Meijer and G. von Helden: Vibrational Spectroscopy of Fluoroformate, FCO<sub>2</sub><sup>-</sup>, Trapped in Helium Nanodroplets. The Journal of Physical Chemistry Letters 9 (9), 2305–2310 (2018).*

*Vanbuel, J., M. Jia, P. Ferrari, S. Gewinner, W. Schöllkopf, M.T. Nguyen, A. Fielicke and E. Janssens: Competitive Molecular and Dissociative Hydrogen Chemisorption on Size Selected Doubly Rhodium Doped Aluminum Clusters. Topics in Catalysis 61 (1), 62–70 (2018).*

*Weichman, M.L., S. Debnath, J.T. Kelly, S. Gewinner, W. Schöllkopf, D.M. Neumark and K.R. Asmis: Dissociative Water Adsorption on Gas-Phase Titanium Dioxide Cluster Anions Probed with Infrared Photodissociation Spectroscopy. Topics in Catalysis 61 (1-2), 92–105 (2018).*

*Winta, C., S. Gewinner, W. Schöllkopf, M. Wolf and A. Paarmann: Second-harmonic phonon spectroscopy of  $\alpha$ -quartz. Physical Review B 97 (9), 094108 (2018).*

*Wu, Z., A. Plucienik, Y. Liu, M. Naschitzki, W. Wachsmann, S. Gewinner, W. Schöllkopf, H. Kühlenbeck and H.-J. Freund: Surface action spectroscopy with rare gas messenger atoms. Review of Scientific Instruments 89 (8), 083107 (2018).*

## 2019

*Debnath, S., X. Song, M.R. Fagiani, M.L. Weichman, M. Gao, S. Maeda, T. Taketsugu, W. Schöllkopf, A. Lyalin, D.M. Neumark and K.R. Asmis: CO<sub>2</sub> Adsorption on Ti<sub>3</sub>O<sub>6</sub><sup>-</sup>: A Novel Carbonate Binding Motif. The Journal of Physical Chemistry C 123 (13), 8439–8446 (2019).*

Lettow, M., E. Mucha, C. Manz, D. Thomas, M. Marianski, G. Meijer, G. von Helden and K. Pagel: The role of the mobile proton in fucose migration. *Analytical and Bioanalytical Chemistry* 411 (19), 4637–4645 (2019).

Marianski, M., J. Seo, E. Mucha, D. Thomas, S. Jung, R. Schlögl, G. Meijer, A. Trunschke and G. von Helden: Structural Characterization of Molybdenum Oxide Nanoclusters Using Ion Mobility Spectrometry-Mass Spectrometry and Infrared Action Spectroscopy. *The Journal of Physical Chemistry C* 123 (13), 7845–7853 (2019).

Mucha, E., A. Stuckmann, M. Marianski, W.B. Struwe, G. Meijer and K. Pagel: In-depth structural analysis of glycans in the gas phase. *Chemical Science* 10 (5), 1272–1284 (2019).

Thomas, D., E. Mucha, M. Lettow, G. Meijer, M. Rossi and G. von Helden: Characterization of a trans-trans Carbonic Acid-Fluoride Complex by Infrared Action Spectroscopy in Helium Nanodroplets. *Journal of the American Chemical Society* 141 (14), 5815–5823 (2019).

Todd, A.M.M., W. Schöllkopf, W.B. Colson, S. Gewinner, S.C. Gottschalk, G. von Helden, H. Junkes, G. Meijer, M. De Pas, T.J. Schultheiss, J.W. Rathke and L.M. Young: The FHI FEL Upgrade Design. In: *Proceedings of the 10th International Particle Accelerator Conference*. JACoW, Geneva, TUPRB103 (2019). ISBN 978-3-95450-208-0.

Vanbuel, J., E.M. Fernández, M. Jia, P. Ferrari, W. Schöllkopf, L.C. Balbás, M.T. Nguyen, A. Fielicke and E. Janssens: Hydrogen Chemisorption on Doubly Vanadium Doped Aluminum Clusters. *Zeitschrift für Physikalische Chemie* 233 (6), 799–812 (2019).

## PhD Projects of the International Max Planck Research School: "Functional Interfaces in Physics and Chemistry"

### Ongoing

Attia, Smadar (FHI): Ligand-Directed Heterogeneous Catalysis on Model Surfaces.

Baldofski, Matthias (HU): TBD

Bethke, Kevin (HU): A Versatile Seebeck Potential Measurement Device for Thermoelectric Thin Films, Powders and Pastes.

Bobowski, Kamil Nick (FU): Investigation of Electronic Structure Using Time-resolved Photoemission Spectroscopy.

Böhnke, Jan (FU): Spin-resolved Photoemission for the Investigation of Ultrafast Magnetization Dynamics.

Dragoumi, Maria (FHI): Accurate Single-Particle Energies and Band Gaps of Solids from Second-Order Møller-Plesset Perturbation Theory and Beyond.

Farinacci, Laetitia (FU): The Magnetic and Electronic Properties of Fe-Porphyrin Molecules on Pb(111) Investigated by Scanning Tunneling Microscopy.

Feldt, Christoph (FU): Single Crystalline Model Surfaces to Study the Catalytic Properties of Nanoporous Gold.

Fidanyan, Karen (FHI): Theoretical Investigations of the Quantum Nature of the Nuclei in Functionalized Interface Materials.

Fischer, Eric Wolfgang (Uni Potsdam): Reduced and Exact System-Bath Quantum Dynamics of IR-driven Adsorbate Vibrations.

Fuhrich, Alexander (FHI): Ultra-thin Germania and Germania-Silica Films Growth, Structure and Reactivity.

Gura, Leonard (FHI): Highspeed Scanning Tunneling Microscopy on Thin Oxide Film Systems.

Haase, Felix (FHI): Surface Reactivity of 3d Transition Metal Based Model Catalysts for the Oxidation of Small Molecules.

Heitz, Julius (FHI): Magnetism at Terahertz Frequencies.

Herzog, Antonia (FHI): Design and Operando Investigation of Nanostructured Bimetallic CuAg Catalysts for CO<sub>2</sub> Electroreduction.

Karra, Mallikarjun (FHI): Microsolvation and Rotation of 'Large' Molecular Impurities Inside Helium Nanodroplets Upon Electronic Excitation.

Ketterl, Anna Sophia (FU): Electron Dynamics in the Spin-Split Surface States of Semiconductors with Strong Spin-Orbit Interaction.

Kordus, David (FHI): Investigation on Structure and Chemical State of Catalysts for CO<sub>2</sub> Hydrogenation.

Krane, Nils (FU): Properties and Utilization of Molybdenum Disulfide as Decoupling Layer on Au(111).

Küllmey, Tim (FU): Theoretical Investigations of Chemically Modified Kesterites under Strain.

Lawrenz, Dominic (FU): Ultrafast Magnetization Dynamics of Ferromagnetic Layered Systems.

Lewandowski, Adrián Leandro (FHI): The Atomic Structure of Ultrathin Germania Films.

Ma, Jie (HU): Electronic Structure of Transition Metal Dichalcogenides and Molecular Semiconductors.

Martyanov, Wjatscheslav (TU): Atomic Structure and Local Electronic Properties of III-V Semiconductor Surfaces and Nanostructures.

Melani, Giacomo (Uni Potsdam): From Structural Fluctuations to Vibrational Spectroscopy of Adsorbates on Surfaces: A Theoretical Study of H<sub>2</sub>O on  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>(0001).

Mucha, Eike (FHI): Vibrational Spectroscopy of Glycans in Helium Nanodroplets.

Paßler, Nikolai (FHI): Nonlinear Interactions of Surface Phonon Polaritons in Polar Dielectric Heterostructures.

Plucienik, Agata (FHI): The Interaction of Hydrogen with CeO<sub>2</sub> (111)/Ru (0001) and Surface Action Spectroscopy: Setup and First Experiments.

Rettenmaier, Clara (FHI): In-situ and Operando Nanocatalysis: Size, Shape and Chemical State Effects.

Richter (Hoffmann), Nina (FU): Characterization of Defects on Metal Supported Single Crystalline Oxide Films.

Rouzegar, Reza (FHI): Terahertz Spectroscopy of Ultrafast Spin Dynamics.

Scholz, Robert (Uni Potsdam): Laser- and Electron Driven Adsorbate Dynamics at Metal Surfaces: Electron Friction Approaches and Beyond.

Speckhard, Daniel (HU/FHI): TBD

Stemmler, Christian (FU): Analysis of Electron Correlation Effects in Strongly Correlated Systems.

Yubero, Valdivielso (FHI): High Resolution Anion Photoelectron Spectroscopy via VMI.

Vorwerk, Christian (HU): Theoretical Spectroscopy of Group-III Oxides.

Wolf, Elisabeth Hannah (FHI): Influence of the Cu/ZnO Interface on Electronic and Catalytic Properties.

Zaki, Eman (FHI): Surface-Sensitive Adsorption of H<sub>2</sub>O and Carbon Dioxide on Magnetite: Fe<sub>3</sub>O<sub>4</sub>(111) Versus Fe<sub>3</sub>O<sub>4</sub>(001).

Zwaschka, Gregor (FHI): Illuminating Water Splitting: Monitoring and Manipulating Water Electrolysis by Modern Laser Techniques.

### **Graduations 2017 - 2019**

Song, Xin (FHI): Vanadium Doped TiO<sub>2</sub>(110) Thin Layers: Preparation, Structure, and Reactivity – Methanol Partial Oxidation.

Kettemann, Frieder (HU): Electrostatically Stabilized Noble Metal Nanoparticles: Syntheses and Catalytic Properties.

Marsoner Steinkasserer, Lukas (FU): Electron Correlation Effects in Layered Structures – A Theoretical First-principles Study.

Heenemann, Maria (FHI): Charge Transfer in Catalysis Studied by In-situ Microwave Cavity Perturbation Techniques.

Nicholson, Christopher (FHI): Electronic Structure and Dynamics of Quasi-one-dimensional Materials.

Kundu, Arpan (HU): Ab Initio Prediction of Isotherms for Pure and Mixed Gas Adsorption in Metal-organic Frameworks.

Alessio, Maristella (HU): Hybrid QM: QM Methods for Extended Periodic Systems.

Mirabella, Francesca (FHI): Water and CO<sub>2</sub> Adsorption on Magnetite Surfaces: A Combined Molecular Beam and IRAS Study.

Appelfeller, Stephan (TU): Atomic Structure and Electronic Properties of Tb Silicide Nanowires.

Davis, Earl (FHI): Iron Oxide and Sulfide Thin Films: Growth Methods and Characterization.

Li, Xiaoke (HU): The Fe<sub>3</sub>O<sub>4</sub>(111) Surface-structure and Reactivity Studied by Density Functional Theory.

Schultz, Thorsten (HU): Energy Level Alignment Mechanisms at Inorganic-organic Semiconductor Interfaces Investigated with Photoelectron Spectroscopy.

Rolf, Daniela (FU): Fundamental Properties of Atoms and Molecules on Surfaces – A Combined STM and Shot-Noise Study.

Salas Illanes, Nora (HU): Electronic Structure of Selected Materials by Means of the QSGW Method within the LAPW+LO Framework.

Ralaiarisoa, Maryline (HU): Electronic Properties of Hybrid Organic-inorganic Perovskite Films: Effects of Composition and Environment.

Werner, Kristin (FHI): On the Role of Oxygen Vacancies in the Surface Chemistry of CeO<sub>2</sub> (Ceria).

The image features a solid yellow background. In the lower half, there are several thin, orange, abstract lines that create a sense of movement and depth, resembling a stylized landscape or architectural structure. These lines vary in length and orientation, some running parallel to each other and others crossing at different angles.

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