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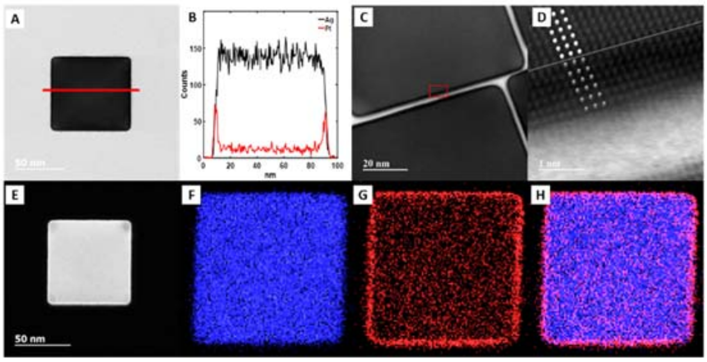

Workshop
Molecular Approaches to Heterogeneous Catalysis and Electrocatalysts

November 20-21, 2017

Organizing Committee

Suljo Linic (TUM-IAS Hans Fischer Fellow, University of Michigan, USA)
Karsten Reuter (TUM)

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European Commission

Program

MONDAY, NOVEMBER 20, 2017

Auditorium (ground floor)

1:00 – 1:15 p.m.	**Welcome Address** Ernst Rank (Director, TUM-IAS)
1:15 – 1:50 p.m.	**Technical analysis of the CO_2 emission impact and catalytic strategies for addressing the problem** Suljo Linic (TUM-IAS Hans Fischer Fellow, University of Michigan)
1:50 – 2:25 p.m.	**Catalysis for light alkanes – from methane functionalization to light olefins** Johannes A. Lercher (TUM)
2:25 – 3:00 p.m.	**Biomass conversion to fuels and chemicals** Will Medlin (University of Colorado Boulder)
3:00 – 3:30 p.m.	*Coffee Break (Foyer, ground floor)*
3:30 – 4:05 p.m.	**Introduction to electrocatalysis** Michael J. Janik (Pennsylvania State University)
4:05 – 4:40 p.m.	**Surface science and X-ray synchrotron methods applied to catalysis** Beatriz Roldan Cuenya (Fritz-Haber Institute of Max Planck Society)
4:40 – 5:15 p.m.	**Catalysis of clusters in the non-scalable size regime I** Ulrich Heiz (TUM)
5:15 – 5:50 p.m.	**Multiscale modeling of catalysis** Karsten Reuter (TUM)

TUESDAY, NOVEMBER 21, 2017

Auditorium (ground floor)

8:30 – 9:05 a.m.	***Operando* nanocatalysis: size, shape, composition, and chemical state effects** Beatriz Roldan Cuenya (Fritz-Haber Institute of Max Planck Society)
9:05 – 9:40 a.m.	**Catalysis of clusters in the non-scalable size regime II** Ulrich Heiz (TUM)
9:40 – 10:15 a.m.	**Refining first-principles photo-electrocatalysis** Karsten Reuter (TUM)
10:15 – 10:45 a.m.	*Coffee Break (Foyer, ground floor)*
10:45 – 11:20 a.m.	**Organic monolayers in heterogeneous catalysis: how “crowding” the reactants can improve catalyst specificity** Will Medlin (University of Colorado Boulder)
11:20 – 11:55 a.m.	**Lessons from enzymes - On the role of steric constraints and chemical environments for catalysis** Johannes A. Lercher (TUM)
11:55 – 12:30 p.m.	**Development of electrocatalytic materials guided by computational chemistry: fuel cells and electrolysis** Michael Janik (Pennsylvania State University)
12:30 – 13:05 p.m.	**Maximizing efficiencies of photocatalytic water splitting by engineering interfaces in multi-component photocatalysts** Suljo Linic (TUM-IAS Hans Fischer Fellow, University of Michigan)

Biographies of Speakers & Abstracts

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Biographies of Speakers & Abstracts

Suljo Linic

Department of Chemical Engineering, University of Michigan
Hans Fischer Fellow, Technical University of Munich

Biography

Suljo Linic obtained his PhD degree, specializing in surface and colloidal chemistry and heterogeneous catalysis, at the University of Delaware in 2003 under the supervision of Prof. Mark Barteau after receiving his BS degree in Physics with minors in Mathematics and Chemistry from West Chester University in West Chester (PA). He was a Max Planck postdoctoral fellow with Prof. Dr. Matthias Scheffler at the Fritz Haber Institute of Max Planck Society in Berlin (Germany), working on first principles studies of surface chemistry. He started his independent faculty career in 2004 at the Department of Chemical Engineering at the University of Michigan in Ann Arbor where he is currently the Class of 1983 Faculty Scholar Professor of chemical engineering.
Prof. Linic's research has been recognized through multiple awards including the 2017 Emmett Award by The North American Catalysis Society, the 2014 ACS (American Chemical Society) Catalysis Lectureship for the Advancement of Catalytic Science, awarded annually by the ACS Catalysis journal and Catalysis Science and Technology Division of ACS, the 2011 Nanoscale Science and Engineering Forum Young Investigator Award, awarded by American Institute of Chemical Engineers, the 2009 ACS Unilever Award awarded by the Colloids and Surface Science Division of ACS, the 2009 Camille Dreyfus Teacher-Scholar Award awarded by the Dreyfus Foundation, the 2008 DuPont Young Professor Award, and a 2006 NSF Career Award. Suljo Linic has presented more than 130 invited and keynote lectures and published more than 60 peer reviewed articles in leading journals in the fields of catalysis and general science. He serves as the associate editor of ACS catalysis journal.

Lecture 1

Technical analysis of the CO_2 emission impact and catalytic strategies for addressing the problem

It has become increasingly clear that human-induced CO_2 emissions are having potentially irreversible impact on the environment. In this presentation, I will share my views on the current state of the CO_2 pollution problem. I will discuss a potential pathways for the mitigation of this problem. In particular, I will focus on the potential solutions that involve catalysts. I will share my views on how achievable those solutions are, and which of those solutions gives us the highest probability to make positive impact.

Lecture 2

Maximizing efficiencies of photocatalytic water splitting by engineering interfaces in multi-component photocatalysts

Solar splitting of water to hydrogen and oxygen is a critical chemical transformation for which no commercially viable photocatalytic systems exist. Developing materials that could execute this reaction with high efficiencies would fundamentally change the environmental footprint of many segments of our economy, including chemical industry, manufacturing and energy sectors. The materials that have received the most attention for this application are hybrids that contain a semiconductor light absorber and an attached metal electrocatalyst that

performs chemical transformations. In these systems, the semiconductor serves to provide the electromotive force (voltage) that is used by the electrocatalysts to drive the reaction. The main problems in the development of these hybrid photocatalysts are the chemical instability of the desired semiconductors (with the appropriate, relatively low band gap) under the relevant water splitting conditions, and the losses associated with the presence of the semiconductor/electrocatalyst junction in these photo-catalytic system.
Recently, it has been demonstrated that these low band gap semiconductor light absorbers can in some cases be stabilized by the use of protective insulator layers by forming metal/insulator/semiconductor (MIS) photocatalysts. This strategy incorporates a stable insulator layer placed between a metal electrocatalyst and semiconductor forming a metal-insulator-semiconductor junction. So far the central focus of this area of research has been on experimentally demonstrating the enhanced stability of the semiconductors covered by these insulating layers under the reaction conditions. In this contribution, we will discuss the realistic targets for solar water splitting for these MIS photocatalyst. We will also analyze the critical problems associated with these multi-component photocatalysts. We will discuss the impact of various components on the photo-catalysts performance. We will also show how by controlling the geometries of these systems at atomistic level we can optimize their solar to hydrogen efficiencies.

Johannes A. Lercher

Department of Chemistry, Technical University of Munich
Institute for Integrated Catalysis, Pacific Northwest National Laboratory

Biography

Johannes A. Lercher, studied Chemistry at TU Wien, receiving his PhD in 1981 at the same institution. After a year at Yale University, he joined TU Wien as Lecturer, later Associate Professor. 1993 he was appointed Professor in the Department of Chemical Technology at the University Twente, the Netherlands and in 1998 in the Department of Chemistry of TU München, Germany. Since 2011 he is also Director of the Institute for Integrated Catalysis at the Pacific Northwest National Laboratory, USA. Being member of the Austrian Academy of Sciences, Member of the Academia Europaea, the European Academy of Sciences, and the US National Academy of Engineering, he is currently President of the European Federation of Catalysis Societies and Editor-in-Chief of the Journal of Catalysis. Recent awards include the ENI Award (hydrocarbons), the David Trimm and Noel Cant Lectureship of the Australian Catalysis Society, the R.B. Anderson Award of the Canadian Catalysis Society, the Francois Gault Lectureship Award of the European Federation of Catalysis Societies, and the Robert Burwell Lectureship in Catalysis of the North American Catalysis Society. Research interests are related to catalysis in nanostructured catalysts, focusing on understanding the influence of the steric and chemical environment to tailor catalytic sites.

Lecture 1

Catalysis for light alkanes – From methane functionalization to light olefins

The easy availability of light alkanes opened new opportunities to synthesize light olefins and oxygenates, challenged only by the high dispersion of the carbon sources, requiring dedicated processes. Three different principal catalysis pathways will be discussed, the conversion of methane to methanol, the oxidative dehydrogenation of ethane and the dehydrogenation of propane to propene, eventually followed by aromatization. For the three approaches it will be shown, how the atomistic understanding of the catalyst properties including an atomistic description of the active site by combining electron microscopy and X-ray absorption spectroscopy and the molecular pathways by identifying elementary steps via NMR spectroscopy and isotope labelling allows to drastically improve catalysts and catalytic pathways. Combining rigorous kinetics with spectroscopy allowed for all three cases to describe rates linked to specific active sites and maximize their concentration. The potential and limitations of this fundamental approaches to improve and re-invent practical catalysts will be discussed.

Lecture 2

Lessons from enzymes - On the role of steric constraints and chemical environments for catalysis

Elementary organic transformations are catalyzed by enzymes at large unmatched rate and selectivity. The local constraints at active sites of enzymes and the local chemical environment are two key properties to achieve such extraordinary catalytic vectors. We use the sites in crystalline nanoporous materials to mimic such well-defined reaction space. Molecules experience constraints that can be subtly adjusted via direct synthesis, as well as via the addition of cations, oxidic clusters or organic fragments. The role of water and other solvents in such environments changes drastically the reaction pathways compared with the catalytic pathways in a homogeneous environment of chemistry in solution. The lecture will compare reactions such as elimination reactions of alcohols, alkylation of aromatic molecules and the role of acid sites for hydrogenation and hydrogenolysis. Experimental methods to define the state of the reacting molecules combined with detailed kinetic analysis and theory will be used to explain the principal contributions of the interactions and the confinement to determine reaction rates. It will be discussed how reaction rates and pathways can be tailored using the space available for a transition state and the chemical constituents around the active site.

Will Medlin

Department of Chemical and Biological Engineering, University of Colorado Boulder

Biography

Will Medlin received his BS and PhD degrees in chemical engineering from Clemson University and the University of Delaware, respectively. After conducting postdoctoral research at Sandia National Laboratories in Livermore, California, he joined the faculty of the University of Colorado, where he now serves as the Denver Business Challenge Endowed Professor in the Department of Chemical & Biological Engineering. His research focuses on the design of solid catalysts for energy and environmental applications. His work has particularly emphasized catalyst surface modification using organic self-assembled monolayers or inorganic thin films to enhance control over catalyst surface and near-surface properties. He has received several research and teaching awards, such as the NSF CAREER award and the AIChE Himmelblau Award. He has been a visiting professor at ETH-Zurich and is currently a guest professor at the Chalmers University of Technology.

Lecture 1

Biomass conversion to fuels and chemicals

This lecture will provide an overview of fundamental aspects of the conversion of plant-based biomass to key products, including fuels, chemicals, and materials. The goal of the presentation is to provide a broad summary of the following aspects of biomass conversion technology: (1) the prospects and challenges associated with different types of crops grown for energy; (2) various biological and thermochemical techniques for deconstruction of biomass into component sugars and other compounds; (3) approaches for upgrading of biomass-derived sugars and other intermediates to end products; and (4) potential impacts of biomass conversion on industry and society, including pressures imposed on the food supply and likely effects on climate change. The presentation will include examples of how different companies have approached development of biomass conversion technologies.

Lecture 2

Organic monolayers in heterogeneous catalysis: how “crowding” the reactants can improve catalyst specificity

One of the most important factors governing the efficiency of a reaction process is the selectivity to desired products. Achieving high selectivity is necessary in realizing efficient chemical processes that minimize energy inputs and waste, but is a major challenge for complex chemical feedstocks with multiple functional groups. Addressing this problem is important both in conventional production of chemicals and for the conversion of biomass to chemicals and fuels, and requires improved designs of the materials that catalyze these reactions.

Our group has investigated several techniques for improving selectivity during the reactions of complex molecules. One approach involves the modification of surfaces with organic self-assembled monolayers (SAMs). By changing the functional groups present within these SAMs, one can precisely control the near-surface environment to enhance specific reactant-surface interactions and thus improve catalyst performance. In this presentation, we will discuss different ways in which surface “crowding” with organic ligands can be used to improve reaction selectivity. These examples span various types of catalysts, including cases where the key reaction steps occur on metal surfaces, on metal oxide surfaces, and at interfaces between metals and metal oxides. The utility of the methods by which SAMs can be used for selectivity control will be illustrated for reaction chemistries important in biomass refining and in the production of valuable chemicals.

Michael J. Janik

Department of Chemical Engineering, Pennsylvania State University

Biography

Michael Janik is a Professor of Chemical Engineering at Pennsylvania State University. He began his appointment at Penn State in August, 2006. His research interests are in the use of computational methods to understand and design materials for alternative energy conversion systems. Current activities address a wide-range of energy technologies including fuel cells and electrolysis, heterogeneous catalysis, organic electronics, and CO_2 capture and utilization.

Research methods emphasize atomistic simulation using quantum chemical methods and kinetic modeling. Janik is affiliated with the Penn State Energy Institute, PSU-Dalian University of Technology Joint Center for Energy Research, the PSU Institutes of Energy and the Environment, and the Battery and Energy Storage Technology Center. He also holds the title of Visiting Professor at Dalian University of Technology. The Janik group currently includes 9 graduate students, 12 undergraduate students, and a post-doctoral research associate. Dr. Janik received his B. S. in Chemical Engineering from Yale University. He completed his doctoral studies at the University of Virginia under the advisement of Bob Davis and Matt Neurock. He has co-authored approximately 110 peer reviewed papers, and co-edited the book “Computational Catalysis” (with Aravind Asthagiri), published by the Royal Society of Chemistry in 2013.

Lecture 1

Introduction to electrocatalysis

In a future world with abundant renewable electricity, electrochemical technologies will become essential for generating fuels and chemicals and storing electrical energy. In this talk, I will review basic construction of an electrochemical cell, electrochemical thermodynamics, and electrochemical kinetics. This overview will then be followed by a more detailed introduction to electrocatalysis. In many “flow-through” electrochemical devices, such as fuel cells and electrolysis units, electrode catalysts, “electrocatalysts”, are needed to accelerate reaction kinetics. Electrocatalysis occurs at an interface between the electrode and an electrolyte, and involves ions and electrons as products and reactants. The electrode typically has a charge under operating conditions, motivating ions in the electrolyte to gather near the charged surface forming an electrochemical double-layer. Transport and reaction kinetics are impacted by the structure and composition of this interface. Throughout the talk, concepts will be introduced using example fuel cell and electrolysis technologies that have potential impact to future energy systems.

Lecture 2

Development of electrocatalytic materials guided by computational chemistry: fuel cells and electrolysis

Our group applies computational chemistry techniques to a range of catalyst and materials design challenges in energy technology. This presentation will concentrate on our work in electrocatalysis. Electrocatalysts are an essential component of fuel cells, electrolyzers, and some battery technologies. The development of composition-structure-functional relationships guides rational design of electrocatalytic materials. Quantum mechanics based computational techniques, such as density functional theory methods, are a useful tool in guiding catalyst design. Density functional theory (DFT) methods are widely used to evaluate surface catalytic reaction mechanisms and to predict the relative performance of various catalyst formulations or structures. Translation of DFT approaches to the electrocatalytic environment requires additional methodological choices due to additional complexities offered by the electrified catalyst-electrolyte interface. This talk will provide an overview of the challenges to atomistic modeling of electrochemical interfaces and describe the various DFT approaches used to model electrocatalytic systems. The use of DFT to determine electrocatalytic reaction mechanisms and guide the design of catalytic materials will be discussed using examples from our group’s research; hydrogen fuel cells, borohydride fuel cells, carbon dioxide and nitrogen reduction to fuels and chemicals.

Beatriz Roldan Cuenya

Department of Interface Science, Fritz-Haber-Institute of the Max Planck Society, Berlin

Biography

Beatriz Roldan Cuenya is currently a director of the Interface Science Department at the Fritz-Haber-Institute of the Max Planck Society in Berlin (Germany). She was previously a Chair Professor in the Department of Physics at the Ruhr-University Bochum (Germany, 2013-2017). She moved from the University of Central Florida (USA) where she was a professor of Physics from 2004 till 2013. She carried out her postdoctoral research in the Department of Chemical Engineering at the University of California Santa Barbara (2001-2003). Prof. Roldan obtained her PhD in Physics from the University of Duisburg-Essen (Germany) summa cum laude in 2001. She completed her M.S./B.S. in Physics with a minor in Materials Science at the University of Oviedo, Spain in 1998.

During her academic career Prof. Roldan received an Early CAREER Award from the US National Science Foundation (2005) and the international Peter Mark Memorial award from the American Vacuum Society (2009). In 2016 she became Fellow of the Max Planck Society in Germany.

Prof. Roldan is the author of 100 peer-reviewed publications and 3 book chapters and has given over 100 invited talks. She has also been granted 2 USA patents. She presently serves in the editorial advisory board of the Surface Science journal and in the Advisory Committee of the Office of Basic Energy Sciences of the US Department of Energy.

Prof. Roldan's research program explores the novel physical and chemical properties of nanostructured materials, with emphasis on advancing the field of nanocatalysis through *in situ* and *operando* characterization of catalysts at work. She has developed synthetic methods for preparing nanoscale catalysts with controlled size, shape, and spacing which have been shown to lead to exceptionally high catalytic activity and selectivity for chemical processes of paramount industrial, societal and environmental importance, such as the utilization of CO_2 for the production of chemicals and fuels, the reduction of poisonous nitric oxide and oxidation of carbon monoxide for applications in the automobile industry, as well as the oxidation and decomposition of alcohols for fuel cell applications.

Lecture 1

Surface science and X-ray synchrotron methods applied to catalysis

This lecture will provide an introduction to the field of Nanoparticle Catalysis and will discuss surface science and X-ray-based methods for the *in situ* and *operando* characterization of nanocatalysts.

Lecture 2

Operando nanocatalysis: size, shape, composition, and chemical state effects

In order to comprehend the properties affecting the catalytic performance of metal nanoparticles (NPs) and nanostructured surfaces, their dynamic nature and response to the environment must be taken into consideration. The working state of a NP catalyst might not be the state in which the catalyst was prepared, but a structural and/or chemical isomer that adapted to the particular reaction conditions. This talk provides examples of recent advances in the preparation, *in situ* and *operando* characterization (AFM, STM, TEM, NRIXS, XAFS) of NP catalysts with well-defined sizes and shapes.

It will be highlighted that for structure-sensitive reactions, catalytic properties such as activity, selectivity and stability against sintering can be tuned through controlled synthesis. This talk will focus on model heterogeneous catalysts (Au, Ag, Zn, Cu, Cu-M with M = Zn, Co, Au, Ag) for reactions of tremendous societal and industrial relevance, namely the gas-phase hydrogenation and electrocatalytic reduction of CO_2 and the oxidation of alcohols. Emphasis will be given to elucidating the role of the NP size, shape, interparticle distance, composition, and chemical state of the catalysts in the activity and selectivity of the former reactions. The results are expected to open up new routes for the reutilization of CO_2 through its direct conversion into valuable chemicals and fuels such as methanol and ethylene.

Ulrich Heiz

Department of Chemistry and Catalysis Research Center,
Technical University of Munich

Biography

Prof. Heiz explores the physical and chemical properties of the smallest matter particles in the non-scalable size regime. In this regime, the properties of the clusters are determined by the exact number of atoms. His research findings create a better understanding of nanocatalysis, asymmetric catalysis and photocatalysis. He also works on characterizing and developing new optic materials based on size-selected clusters.

After studying chemistry at the University of Bern (1987), Prof. Heiz obtained his doctorate there in physical chemistry (1991). Thanks to grants from the Andrew Mellon Foundation and the Swiss National Science Foundation, he was able to do research at the laboratory of the Exxon Research and Engineering Company and the University of Pittsburgh. He completed his lecturer qualification in physics at the University of Lausanne in 1998. After that, he spent time in Japan and Berlin (as an Alexander von Humboldt fellow) before accepting a professorship at the University of Ulm in 2000. Since 2004, he has been full professor of physical chemistry at TUM. Prof. Heiz was a member of the Alexander von Humboldt Foundation board and in 2010, he received an ERC Advanced Grant. From 2013 to 2017, he was academic director of the Catalysis Research Center at TUM. Currently, he is also an elected member of the Advisory Board of the Max-Planck-Institute in Stuttgart.

Lecture 1

Catalysis of clusters in the non-scalable size regime I

The study of size-selected clusters on surfaces has been growing into a vital research field within cluster science and catalysis since the discovery of the astonishing size-dependent activity of small gold clusters for the oxidation of CO in the late nineties. More than one decade of research in a combined effort between theory and experiment has resulted in a detailed understanding of cluster’s structural, electronic, optical, and magnetic properties. Thus, we understand several chemical, catalytic, and photocatalytic processes on clusters on a molecular level, today. This exciting advancement was only possible by a parallel development and introduction of novel, state-of-the-art methods, both in experiment and theory. We are convinced that such a research strategy based on a deep fundamental understanding will ultimately allow us to implement knowledge-based improvements in several key technologies using materials in the nano- and subnano size range.

In this lecture, fundamental properties of clusters are discussed. We then present results on the activation of methane on free tantalum and tantalumoxide clusters and show that intrinsic cluster reactivities behave in a non-predictable way with reaction paths strongly dependent on cluster size and composition. Ab initio calculations confirm the existence of a size-dependent rate-determining step measured by kinetic and isotope exchange experiments. Furthermore, the exothermicity of the reaction strongly varies with size.

Lecture 2

Catalysis of clusters in the non-scalable size regime II

In this lecture, strategies for optimizing cluster stability against ripening will be presented, where both, the binding of the clusters to the surface and that of the individual atoms, must be controlled. Such tuning of the interactions may be achieved through the judicious selection of surfaces with laterally modulated wettability. Furthermore, we observed that the broadening of cluster size distribution induced by Ostwald ripening is efficiently suppressed even under reaction conditions for size-selected cluster samples. As an example of the catalysis of clusters on surfaces, results on the hydrogenation of ethylene on Pt_n clusters are presented. Surprisingly we observed an onset of reactivity going from Pt_9 to Pt_{10} and Pt_{13} being the most active cluster size. The origin of the size dependent reactivity is the local charging of the reactive atoms in the cluster. We present strategies of how to tune the reactivity through the design of the local environment of the clusters.

Karsten Reuter

Department of Chemistry, Technical University of Munich

Biography

Karsten Reuter received his Doctoral Degree in Theoretical Physics from the University Erlangen-Nürnberg in 1998. After postdoctoral stays at the Fritz-Haber-Institut der Max-Planck-Gesellschaft in Berlin and the FOM-Institut for Atomic and Molecular Physics in Amsterdam, he headed an Independent Junior Research Group at the Fritz-Haber-Institut from 2005, combined with the position of a Privatdozent at the Free University Berlin. Since 2009 he holds the Chair of Theoretical Chemistry at the Technische Universität München (TUM), is an adjunct professor in the TUM Physics Department, and is affiliated to the TUM Catalysis Research Center. In 2014/15 he was a visiting professor at the Chemical Engineering Department of Stanford University. His research interests center on a quantitative multiscale modeling of materials properties and functions in energy and catalysis, in particular on linking predictive-quality quantum-mechanical electronic structure calculations with more coarse-grained statistical and continuum approaches. He has co-authored over 200 publications in this and related fields.

Lecture 1

Multiscale modeling of catalysis

First-principles electronic structure calculations have emerged as a key contributor in modern heterogeneous catalysis research. Next to the dedicated computation of thermostability, spectroscopic signals and reactivity descriptors, they are increasingly used as basis for predictive-quality multiscale modeling approaches [1]. Corresponding approaches additionally account for meso- and macroscopic aspects like statistical interplay within the catalytic cycle or heat and mass transport in the reactor. This provides unprecedented insight into the catalytic function, be that the actual reaction mechanisms or the nature of the surface of the operating catalyst. Particularly with respect to the latter, this insight points at substantial deviations from the standard picture that analyzes catalytic function merely in terms of properties of and processes at active sites as they emerge from a crystal lattice truncation of the nominal catalyst bulk material [2]. Instead, a highly dynamic picture is suggested with continuous reaction-induced complex (surface) morphological changes at (near-) ambient conditions or an evolving, possibly liquid-like phase behavior due to limited heat dissipation channels. Advancing multi-scale catalysis modeling methodology to scrutinize such a picture is the next grand challenge. In my lecture I will introduce corresponding methodologies and discuss their capabilities and current limitations.

References
[1] M.K. Sabbe, M.-F. Reyniers, K. Reuter, Catal. Sci. Technol. 2 (2012) 2010
[2] K. Reuter, Catal. Lett. 146 (2016) 541

Lecture 2

Refining first-principles photo-electrocatalysis

First-principles based computational modelling assumes an ever increasing role in understanding and partly already optimizing catalysts for photo-electrochemical reactions. The success of prevalent approaches like the computational hydrogen electrode (CHE) thereby relies largely on a numerical efficiency sufficiently high as to allow for large-scale screening of catalyst materials. Such efficiency arises out of well-chosen descriptors that ideally condense the mechanistic understanding of the ongoing catalytic reactions at the solid-liquid interface.

In recent years, there is increasing evidence that this understanding needs further scrutiny. Critical aspects in this respect are the neglect of solvation effects, the assumption of pathways that exclusively proceed via proton-coupled electron transfer steps, and the neglect of any kinetic limitations in the CHE approach. In this talk I will review our recent activities in addressing these issues with detailed first-principles based multiscale modeling approaches. Thereby, in particular embedding approaches – both on the solid and the liquid side of the interface – have the potential for a refined description without sacrificing computational efficiency.