

SCHLOSS RINGBERG

May 18 to 21, 2016

Symposium on Dynamics at Surfaces



For the participants arriving by train at Tegernsee station, we recommend to use a taxi from the train station towards the castle [Taxi Kaufmann](#) phone **+49 (0)8022 / 5555**.

Wednesday May 18th	
16:00-18:00	Arrival and registration
18:00-19:30	Dinner
19:30-20:00	Informal Discussions
20:00-21:00	Informal Discussions
21:00-	Poster Session I
Thursday May 19th	
08:00-9:00	Breakfast
<i>Session Chair:</i> Prof. Theofanis Kitsopoulos, <i>Welcoming Remarks and Overview.</i>	
9:00-9:30	Prof. Theofanis Kitsopoulos, <i>Using Ion Imaging in Surface Science</i>
9:30-10:00	Altschäffel, Jan: <i>Ab initio molecular dynamics simulations of CO and NO scattering from Au(111)</i>
10:00-10:30	Coffee Break
10:30-11:00	Kammler, Marvin: <i>Hydrogen atom scattering from M(111) surfaces: Molecular dynamics simulations on a full-dimensional EMT Potential Energy Surface</i>
11:00-11:30	Dr. Park, Barratt: <i>Toward Vibrationally Inelastic Direct Scattering of Formaldehyde from Au(111)</i>
11:30-12:00	Hahn, Hinrich: <i>Measuring the kinetics of chemical reactions on metal surfaces in a molecular beam surface scattering experiment</i>
12:00-13:30	Lunch Break
<i>Session Chair:</i> Dr. Tim Schäfer	
13:30-14:00	Jiang, Hongyan: <i>H atom scattering from epitaxial graphene on Pt(111)</i>
14:00-14:30	Ahrens, Jennifer: <i>Ultrafast photo-induced electron transfer in self-assembled donor-acceptor coordination cages</i>
14:30-15:00	Kaufmann, Sven <i>Associative Desorption of Hydrogen from Cu(111) and Cu(211)</i>
15:00-18:00	Free time
18:00-20:00	Dinner
20:00-	Poster Session II

Friday May 20th

8:00-9:00	Breakfast
<i>Session Chair:</i> Prof. Jörg Schroeder	
9:00-10:00	Prof. Spiegelman, Fernand: <i>The Density Functional Tight Binding approach for silver and bulk systems</i>
10:00-10:30	Coffee Break
10:30-11:30	Prof. Beck, Rainer : <i>Exploring Gas/Surface Reaction Dynamics via Quantum State Resolved Molecular Beam Experiments</i>
11:30-12:00	Chen, Li: <i>Dispersed laser-induced infrared fluorescence from a monolayer CO on NaCl (100)</i>
12:00-13:30	Lunch Break
<i>Session Chair:</i> Dr. Oliver Bünermann	
13:30-14:30	Priv. Doz. Dr. Flege, Jan Ingo <i>Metal oxidation and epitaxial oxide growth at the nanoscale: an in situ perspective</i>
14:30-15:30	Prof. Zhang, DongHui <i>Quantum Dynamics Studies of Chemisorption of H₂O and CH₄ on Metal Surfaces</i>
15:00-18:00	Free time
18:00-20:00	Dinner
21:00-	Poster Session III

Saturday May 21st

8:00-9:00	Breakfast
<i>Session Chair:</i> Prof. Dirk Schwarzer	
9:00-10:00	Prof. Rothmann, Johann: <i>HgCdTe APDs for low photon number IR detection</i>
10:00-10:30	Coffee Break
10:30-11:30	Prof. Rahinov, Igor: <i>Gas-phase high-temperature synthesis of catalytic materials</i>
11:30-12:00	Prof. Alec Wodtke: <i>Closing Remarks</i>
12:00-13:30	Getting Lunch Packages
	Departure

ABSTRACTS

Ultrafast photo-induced electron transfer in self-assembled donor-acceptor coordination cages.

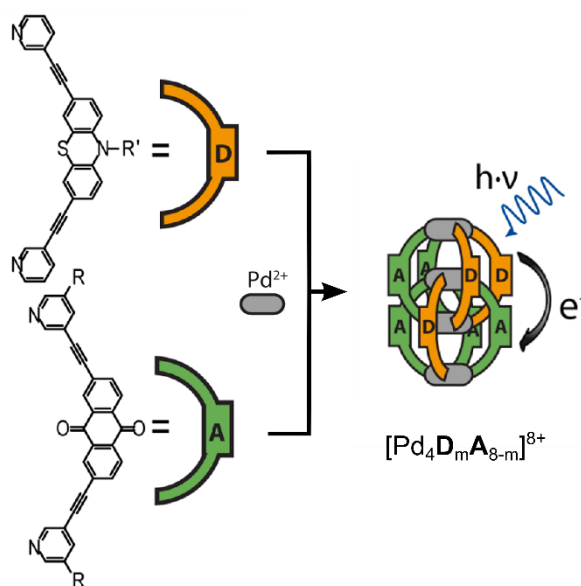
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Electron donor-acceptor complexes built from self-assembled palladium coordinated phenothiazine (**D**) and 9,10-anthraquinone (**A**) based ligands, Pd₄D_mA_{8-m}, were investigated by femtosecond spectroscopy with UV excitation and Vis/mid-IR detection to explore their potential use for photovoltaics. Coordination of the acceptor ligand to palladium in the complex Pd₄A₈ had no influence on the excited state dynamics of the anthraquinone chromophore which is determined by fast intersystem crossing to a long lived triplet state. On the other hand, excitation of the pure donor cage Pd₄D₈ leads to efficient ligand-to-metal charge transfer. For mixed complexes Pd₄D_mA_{8-m} clearly ligand-to-ligand charge transfer within 200 fs is detected regardless of whether donor or acceptor ligands are photo-excited. This is evident from the appearance of a carbonyl stretching band in the mid-IR assigned to the anthraquinone radical anion. The lifetime of the charge separated state appears to be very inhomogeneous ranging from 3.0-2000 ps. The reason for this observation could be the broad distribution of donor-acceptor cage configurations, different spin states of the charge-separated diradical ion pairs or a combination of both.



[1] Frank, Ahrens, Bejenke, Krick, Schwarzer, Clever, *Light-induced Charge Separation in Densely Packed Donor-Acceptor Coordination Cages*. In Preparation.

Ab initio molecular dynamics simulations of CO and NO scattering from Au(111)

Jan Altschäffel, Sascha Kandratsenka and Alec M. Wodtke
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The MD simulations of both molecules from the gold surface are performed with several GGA functionals for different conditions of surface and molecule to study the molecule translational inelasticity as well as its vibrational energy loss channels. The results are compared to both, experiment and theory, for NO in particular the Independent-Electron-Surface-Hopping (IESH) theory. Since, this theory describes the loss in translational energy not qualitatively correct but was successful in describing the vibrational excitation and multi-quantum relaxation for this system [1]-[2]. More recent, a detailed study shows that the potential energy surface (PES) for NO/Au(111) is too soft and corrugated [3]. Since, the translational energy loss is in qualitative agreement with experimental results one can conclude that the failure of the PES is attributed to the fitting procedure to obtain the PES.

Further the interaction of both molecules is investigated for different molecule orientations and adsorption sites. In case of NO charge distributions are presented which show that a description with localized basis sets are necessary to find the correct electronic ground state energy.

[1]: Neil Shenvi et al., *Science*, **326**, **5954**, p. 829–832, **2009**.

[2]: Russel Cooper et al., *Angew. Chemie Int. Edt.* **51**, **20**, p. 4954–4958, **2012**.

[3]: K. Golibrzuch et al., *J. Chem. Phys.* 140 044701, **2014**.

Exploring Gas/Surface Reaction Dynamics via Quantum State Resolved Molecular Beam Experiments

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We present recent results from our laboratory on quantum state resolved reactivity measurements for the dissociative chemisorption of methane and water and their deuterated isotopologues on Ni and Pt surfaces¹⁻⁷. Both dissociation reactions play an important role in the steam reforming process used to convert methane and water into a mixture of hydrogen and carbon monoxide by heterogeneous catalysis. Using state-selective reactant preparation by rapid adiabatic passage in a molecular beam, we prepare the surface incident methane and water molecules in specific ro-vibrational quantum states and measure the state-resolved reactivity on a single crystal surface using surface analytical techniques such as Auger electron spectroscopy, King & Wells beam reflectivity, and reflection absorption infrared spectroscopy (RAIRS). We also probe the quantum state distribution of the nonreactive scattered molecules by combining infrared laser tagging with bolometric detection. The results of our measurements provide evidence for mode- and bond-specificity as well as steric effects in chemisorption reactions and show that the dissociation of both methane and water cannot be described by statistical rate theory but require dynamical treatments including all internal vibrational and rotational degrees of freedom of the dissociating molecule. The detailed reactivity data obtained in our measurements serves as stringent test for the development of a predictive understanding of these industrially important gas/surface using first principles theory.

- [1] L. Chen, H. Ueta and R.D. Beck, *J. Phys. Chem. C*, **119**, 11199 (2015).
- [2] H.J. Chadwick and R.D. Beck, *Chem. Soc. Rev.* 2015, 10.1039/C5CS00476D
- [3] Hundt, Jiang, Van Reijzen, Guo., Beck, *Science*, **344**, 504 (2014).
- [4] P.M. Hundt, M.E. Van Reijzen, H. Ueta, and R.D. Beck, *J. Phys. Chem. Lett.*, **5**, 1963 (2014).
- [5] H. J. Chadwick, P.M. Hundt, M.E. Van Reijzen, B. Yoder and R.D. Beck, *J. Chem Phys*, **140**, 34321 (2014).
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Dispersed laser-induced infrared fluorescence from a monolayer CO on NaCl (100)

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Dispersed laser-induced infrared fluorescence from a monolayer CO on NaCl (100) surface is measured in both frequency and time domain for the first time thanks to the new emerging infrared photon detection technology-superconducting nanowire single photon detector (SNSPD)[1, 2], which has single-photon sensitivity in mid-infrared wavelength range and sub-nanosecond time response.

Following vibrational excitation of a monolayer $^{13}\text{C}^{18}\text{O}$ ($v=1$, 2054.4 cm^{-1}) on NaCl (100) at a surface temperature $T_s=7\text{ K}$ with a 10 ns laser pulse, fluorescence from $^{13}\text{C}^{18}\text{O}$ vibrational states up to $v=17$ are detected in $\Delta v=-2$ overtone emission region $3350\text{-}3390\text{ cm}^{-1}$. The dispersed fluorescence temporal profile is vibrational state dependent, the signal decays in a time range of 10 to 100 μs for detected vibrational states from $v=5$ to $v=17$. The $v=2$ to $v=4$ states signal are too weak for detection due to much faster decay. This dispersed fluorescence emission spectrum and temporal profile measurement result shows that the vibrational energy pooling process, which is an extremely efficient vibrational energy transfer process occurred in multilayer CO or bulk CO crystals[3, 4], can be also an important vibrational energy flow process for a monolayer CO at an insulator surface.

However, the observed 10 to 100 μs decay time for the dispersed fluorescence signal from the monolayer CO on NaCl (100) is about two orders of magnitude shorter than that for multilayer CO from 10 layers up to 2000 layers. This significant difference suggests a strong surface quenching effect on the laser-induced infrared fluorescence of surface adsorbed monolayer CO molecules.

The surface quenching effect observed here seems to be much stronger than that reported from a previous experiment by Ewing's group[5]. Ewing *et al* was not able to measure the dispersed but the overall fluorescence signal from a monolayer CO on NaCl(100) due to limited detection sensitivity, the measured decay time of the overall fluorescence temporal profile is 4.3 ms, two orders of magnitude longer than our measured overall fluorescence lifetime (ca. 50 μs). Hypotheses are proposed to explain the large discrepancy.

References

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Inelasticity in Hydrogen atom scattering from surfaces: A probe for energy conversion

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Developing an understanding of energy conversion at surfaces is of great fundamental and practical interest and the subject of extensive investigation over many years. Measuring the change in momentum and quantum state in collisions of atoms and molecules with surfaces are fundamental tools to evolve an understanding of how atomic and molecular degrees of freedom couple to surfaces. Despite this extensive effort, we do not have an understanding of even the simple problem of how Hydrogen atoms lose kinetic energy and become adsorbed on surfaces. Recently, we have developed an apparatus to measure energy conversion in H-atom surface collisions: A mono energetic H-atom beam is pointed towards a surface at defined incidence angles. Employing Rydberg Atom Tagging the final scattering angular and kinetic energy distributions are measured.

A first experiment showed that drastic differences are observed when Hydrogen atoms are scattered from a metal versus an insulator surface. Comparison of the experimental results to recent theoretical work indicate that electron-hole-pair excitation plays an important role in scattering processes with metals.^[1] In further studies the influence of various experimental parameters were investigated: Kinetic energy, incidence angles, isotope of the incident beam and temperature and structure of the surface.

[1] O. Bünermann et al., *Science*, 2015, 350, 1346-1349.

Metal oxidation and epitaxial oxide growth at the nanoscale: an *in situ* perspective

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Oxides are ubiquitous in our modern world and play a prominent role in a variety of every-day phenomena, e.g., as rather unwanted products of corrosion or as a crucial component in heterogeneous catalysis and microelectronics. Generally, oxide functionality is determined by the oxide's atomic structure and morphology, which are deeply intertwined with its synthesis and which result in specific chemical and electronic properties. Especially the technologically relevant transition metal and rare-earth oxides exhibit a variety of crystal structures and oxidation states, rendering the analysis of the structure-function relationship rather complex and experimentally very challenging to address. Furthermore, the materials properties are heavily influenced by the structure and composition of the oxide surface mediating the interaction of the oxide with its surrounding.

In my presentation, I will demonstrate what kind of novel insights into surface chemical redox reactions and growth processes of ultrathin oxide films and nanostructures can be gained by applying low-energy electron microscopy (LEEM). Besides offering a lateral resolution down to ten nanometers at video rates, even time-resolved probing of changes in local atomic and electronic structure is possible [1]. This experimental approach is particularly attractive in nanoheterogeneous systems exposed to reactive environments since not only the various surface components may be investigated individually, but also potential cooperative effects mediated by their coexistence and close proximity. Selected examples will include the oxidation of the Ru(0001) single crystal surface [2-4] and the subsequent growth of cerium oxide nanostructures [5-7] to illustrate the capabilities of the method.

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- [3] J. C. Goritzka, B. Herd, P. P. T. Krause, J. Falta, J. I. Flege, and H. Over, *Phys. Chem. Chem. Phys.* **17**, 13895 (2015).
- [4] J. I. Flege, J. Lachnitt, D. Mazur, P. Sutter, and J. Falta, *Phys. Chem. Chem. Phys.* **18**, 213 (2016).
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Scattering HCl from Au(111): Vibrational Excitation Probabilities and Dissociative Adsorption

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Gas phase dynamics as well as dynamics at surfaces deal with the most elementary steps of chemical reactions like energy transfer between molecular and surface degrees of freedom, electron transfer, and the making and breaking of chemical bonds. We report absolute vibrational excitation probabilities and dissociative adsorption of HCl molecules encountering a Au(111) surface.

Employing molecular beam techniques in ultrahigh vacuum we are able to study the scattering of gas phase molecules like HCl under well-defined, collision free conditions. A narrow-linewidth, high-intensity pulsed IR laser in combination with UV laser based resonance enhanced multiphoton ionization (REMPI) enables us to quantum state selectively prepare and detect molecules before and after scattering from the single crystal surface, thus giving insight into the flow of energy between the aforementioned degrees of freedom.

HCl exhibits vibrational excitation upon scattering which depends on incident translational energy and vibrational state as well as surface temperature. As previously reported¹, we found concurrent adiabatic (V-T coupling) and nonadiabatic (V-EHP coupling) mechanisms of excitation to occur. While an increase in the incident vibrational state from $\nu=0$ to $\nu=1$ enhanced the excitation probability by a factor of approximately 20, the excitation from $\nu=2 \rightarrow 3$ could not be observed. Presumably, the majority of these collision trajectories led to dissociation instead of further vibrational excitation.

This is in line with our findings at higher incident translational energies (> 1 eV), where HCl undergoes dissociation as proven by detection of Cl atoms using Auger electron spectroscopy (AES).² As predicted by theory^{3, 4}, we found molecules in $\nu=1$ to be more reactive than those in $\nu=0$. However, measured energetic dissociation barriers and vibrational efficacy appear to be much higher than predicted.

1. Q. Ran, D. Matsiev, D. J. Auerbach and A. M. Wodtke, *Phys. Rev. Lett.*, 2007, **98**, 237601.
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3. T. Liu, B. Fu and D. H. Zhang, *Sci. China Chem.*, 2013, **57**, 147-155.
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Measuring the kinetics of chemical reactions on metal surfaces in a molecular beam surface scattering experiment

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We use spatial ion imaging and slicing in combination with intense femtosecond laser pulses for strong field ionization to examine the kinetics of reactions at surfaces occurring after molecular beam metal surface collisions. The universal ionization method together with spatial imaging make a versatile detection scheme that enables us to measure speed and angular distributions of the mass resolved reaction products. Ions created in the detection chamber of our UHV machine are extracted by means of a pulsed electric field and accelerated towards a gated MCP/phosphor screen assembly and a CCD camera.

The reaction particularly featured in this work is the oxidation of CO on an oxygen pre-covered Pt(111) surface. While a first pulsed molecular beam of O₂ provides a certain oxygen coverage on the surface, a second, temporally delayed beam of dilute CO is shot at the surface with an incidence angle of 23°. O₂ coverage dependent CO and CO₂ desorption rates and angular distributions are measured. Two discriminable CO₂ product species that differ significantly in velocity and angular distribution are observed. Reaction rates for the two different reactions observed are derived from a kinetic analysis.

Microsecond Kinetic Measurements for Catalytic Surface Reactions via Ion Imaging.

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We describe the use of ion imaging to study molecular beam-surface scattering. Imaging detection allows the efficient measurement of the laboratory frame angular and speed distributions of products from direct inelastic scattering [1], trapping desorption, or catalytic reactions on the surface. The technique can be extended to provide direct information about the rates of desorption and reaction as a function of surface temperature. The measured rates, in turn, yield information about the barriers controlling the chemical reactivity on the surface.

We present results for the prototypical catalytic oxidation of CO by O₂ on Pt(111). We find evidence for two different reaction channels with different barriers, which produce CO₂ with different final scattering velocities. The separation of the product channels allowed by the imaging detection technique allows the kinetics of the different channels to be measured independently.

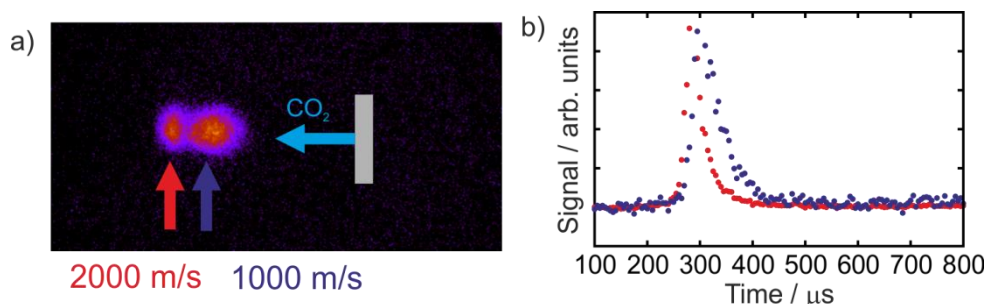


Figure 1. a) Ion image showing two different velocity components for CO₂ formed on the Pt(111) surface. b) The time dependence of the signal in the different channels measured by the delay between the molecular beam pulse and the ionization laser.

These experimental techniques can be applied to a wide range of chemical reactions occurring at surfaces, providing benchmark data for comparison with theory or for use in kinetic models. The speed and efficiency of the technique also make it suitable for reactivity screening.

[1] D.J. Harding, J. Neugeboren, D.J. Auerbach, T.N. Kitsopoulos and A.M. Wodtke, *J. Phys. Chem. A*, **119**, 12255 (2015)

H at Au(111): A full-dimensional PES and the importance of nonadiabatic effects.

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I have constructed a full-dimensional potential energy surface (PES) for a H atom interacting with a Au(111) surface by fitting the analytic form of the Effective Medium Theory [1] to density functional theory (DFT) energies. The fit used energies of the H-Au system with the Au atoms at their relaxed lattice positions as well as configurations with the Au atoms displaced from these positions. The procedure provides an accurate treatment for displacements of Au atoms and compares well to DFT energies for scattering of H atoms obtained from *ab initio* molecular dynamics (AIMD); in adiabatic molecular dynamics simulation, the PES is capable of reproducing the energy loss behavior of AIMD [2,3]. The effective medium theory also provides the background electron density which makes it possible to treat the excitation of energetically low lying electron hole pairs self-consistently in the frame of local density friction approximation. I have tested various incidence conditions for H scattering from Au(111) and find that in all cases, the energy loss by electron hole pair excitation is by far the most efficient pathway and dominates the scattering behavior and influences the adsorption mechanism significantly.

Comparison to experiment confirms that energy loss to electron hole pairs is the dominant energy dissipation pathway [4]. Molecular dynamics simulations including nonadiabatic effects for multiple incidence conditions are in very good agreement with experiment and predictions for scattering behavior at lower temperatures show a resolution of peaks occasioned by different scattering mechanisms.

[1] K. W. Jacobsen, *Surface Science*, 366, 394-402, **1996**.

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[3] G.J. Kroes, *JCP*, 141, 054705, **2014**. M. Pavanello, *JPC Letters*, 4, 3735-3740, **2013**.

[4] O. Bünermann, *Science*, 350, 1346-1349, **2015**.

H atom scattering from epitaxial graphene on Pt(111)

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The interaction of hydrogen atoms with graphene has gained considerable attention in recent years. Adsorption of hydrogen atoms on graphene is playing an important role in reversible hydrogen storage, graphene based nanoscale electronics, nuclear fusion and interstellar chemistry.

The interaction of hydrogen atoms with graphene is studied in a newly built atom-surface scattering machine. A nearly mono-energetic hydrogen atom beam is formed by laser photolysis. The hydrogen atoms are scattered from a well-defined sample held in UHV. The scattered hydrogen atoms are detected using Rydberg-atom neutral time-of-flight. This technique allows us to record angle resolved, high resolution energy loss spectra of scattered hydrogen atoms.

Scattering of hydrogen atoms from epitaxial graphene on Pt(111) shows a very different behavior than scattering from clean Pt(111). For low incidence kinetic energy, graphene acts like an atomic mirror. We observe narrow scattering angle and kinetic energy distribution and small translational energy loss. For high incidence kinetic energy, broad distribution and large translational energy loss is observed. The process obeys normal energy scaling. We speculate that this behavior is connected to the barrier of hydrogen atom adsorption on graphene.

Hydrogen atom scattering from M(111) surfaces: Molecular dynamics simulations on a full-dimensional EMT Potential Energy Surface

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Potential Energy Surfaces (PES) have long been a useful means to simulate scattering experiments much faster than using “on-the-fly” *ab initio* molecular dynamics (AIMD). This work extends the successful theoretical considerations concerning molecular dynamics (MD) studies done in this group [1,2] for H at Au(111) to the case of some other transition metal surfaces. The H atom is placed in a grid-like fashion at several hundred different positions in and above the surface and the energy of these configurations is evaluated by the PBE functional. Additionally, about a dozen AIMD trajectories are run to also sample non-equilibrium configuration space of the system. Next, the energies of one AIMD trajectory as well as of the entire grid are fitted to an analytical function resulting from Effective Medium Theory [3]. It contains seven physically meaningful parameters for each atomic species which can be used to derive structural properties, as well as to run classical MD simulations. The quality of the fit is defined by the RMS error regarding AIMD trajectories not included in the fit and by the reproduction of the metal elastic properties. This work extends the understanding of scattering processes of a hydrogen atom from various transition metals on the atomic level by employing a highly accurate PES. The dependence of H atom trapping probability and energy losses on the metal is analyzed and related to intrinsic metal properties.

References

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- [2] O. Bünermann *et al.*, Science, **350**, 1346, 2015
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Associative Desorption of Hydrogen from Cu(111) and Cu(211)

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The interaction of hydrogen molecules with metal surfaces is one of the most fundamental reactions in surface chemistry. Dissociative adsorption is often the first step in heterogeneous catalysis and tightly related to its reverse process, the associative desorption. In this experiment we studied the product energy distribution of nascent hydrogen molecules desorbing from different copper surfaces after recombination of the atoms by measuring their flight time through a field-free drift region. Using the principle of detailed balance allows us to determine the translational threshold of dissociative adsorption, E_0 , for each ro-vibrational state. Our results show that E_0 only weakly depends on the rotational state, whereas vibrational excitation results in a strong reduction of the threshold. We present data for H₂, HD and D₂ desorbing from Cu(111) and Cu(211) surfaces, respectively. We compare our results with existing data^[1,2] and with results from high-level *ab initio* quantum chemical calculations^[3,4].

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Scattering of Formaldehyde from Au (111)

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Experimental techniques for diatomic molecule-surface scattering allow controlling the scattering process in a very precise manner. For NO / Au (111) surface scattering e. g. incidence conditions like rotational and vibrational state, translational energy, and orientation can be adjusted.¹ Furthermore, the product's distribution among rotational and vibrational states as well as its translational energy can be revealed. Up to now, it remains unclear if the concepts found for diatomic molecules can also be applied to polyatomic molecules. In addition, polyatomic molecule-surface scattering is interesting since it potentially shows much richer scattering dynamics such as mode-specificity.²

As one of the first steps towards full control of polyatomic molecule-surface scattering we examine the scattering of ground state formaldehyde from Au (111). A special heatable nozzle design allows us to produce rotationally cold beams (5-10 K) of formaldehyde seeded in different carrier gasses. The short pulse length of the molecular beam pulse (20-40 μ s) enables us to derive the final speed of scattered molecules in a TOF experiment. Here, we detect formaldehyde employing 2+1 REMPI via the $^1A_2(3p_x)$ Rydberg state.³ For a more state specific detection we developed a 1+1' REMPI scheme via the \tilde{A} state of formaldehyde which allows us to characterize the rotational state distribution of the scattered molecules. From both experiments we can derive that at incidence energies higher than 0.3 eV formaldehyde is mainly directly scattered from Au (111).

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Vibrational lifetime of small diatomic molecules physisorbed on noble metals: A new experimental setup

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I will present the construction of a new experimental setup to determine the vibrational lifetime of diatomic molecules adsorbed on noble metals using fs pump-probe IR spectroscopy. Two separate two-stage optical parametric amplifiers (OPA) based on BBO crystal followed by difference frequency mixing in AgGaS₂ crystal deliver ultrafast pulses in MID-IR range (~5 μm). The production of these ultrafast IR pulses will be discussed.

One of the early planned experiment is the direct measurement of vibrational lifetime of excited CO (v=1) on Au(111) as a function of coverage and laser fluence. TPD study of CO on Au(111) reveals that the sample surface temperature of ~35K is a requirement. A challenge to perform surface spectroscopy at low temperature using closed cycle He cryostat is the mechanical vibration of sample surface. To overcome, an entirely new design of experimental setup has been successfully implemented using Homebuilt LabVIEW program. The UHV chamber has been well-equipped for ion sputtering, annealing, Auger electron spectroscopy (AES), temperature programmed desorption (TPD) spectroscopy and other spectroscopic techniques.

Energy Transfer in Collisions of highly vibrationally excited NO with Vanadium Dioxide Surfaces

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Scattering of highly vibrationally excited NO from both insulating and conducting surfaces has increased our understanding of energy exchange channels between molecules and surfaces. It turns out that non-adiabatic effects play an important role during the interaction of the molecule with metals due to coupling of vibrational degrees of freedom to electron hole pairs (EHP).^[1,2] In this study, we present work on the interaction of highly vibrationally excited NO molecules with vanadium dioxide (VO₂). VO₂ has a Mott transition at 60°C, at which it changes its properties from insulating to conducting.^[3] This allows for switching the band gap of the surface on and off by only slightly adjusting its temperature. In the future, we hope to observe corresponding temperature effects on the vibrational energy transfer in NO/VO₂ surface collisions.

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Scattering of highly vibrationally excited NO from Ge(111)

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Scattering small molecules from well-defined surfaces is an exceptional way to investigate energy transfer processes at the gas-solid interface. Experiments on highly vibrationally excited molecules are especially interesting, because nonadiabatic effects might be enhanced due to the stretched geometry of the molecule. In this work we have scattered highly vibrationally excited NO ($v''=11$) from a semiconducting Ge(111) surface. Similar to earlier experiments on Au(111) and LiF(001), final vibrational, rotational and translational energies have been measured. In contrast to metal surfaces, scattering from germanium shows very inefficient vibrational relaxation. We explain this difference by the bandgap of the semiconducting germanium crystal and so a less efficient coupling to electron hole pairs.

Design and Construction of an UHV chamber to perform surface chemistry experiments for the practical course in advanced physical chemistry

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The study of characteristics and processes of molecules at surfaces is of great practical importance in surface science. Several experimental methods for the investigation of solid surfaces have been developed during the past decades. In order to introduce undergraduate students to some of the techniques used in surface science, we have designed and built a new UHV chamber. This can be used for thermal programmed desorption measurements (TPD), Auger electron spectroscopy (AES), and LEED. It contains a sample holder for 4 samples, which can be cooled down to cryogenic temperature by a He compressor and heated up to 1000 °C by resistive heating. In this poster the layout of the UHV chamber will be shown as well as some proof-of-principal TPD measurements of Xe on Au(100) and Xe on Au(111).

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TOWARD VIBRATIONALLY INELASTIC DIRECT SCATTERING of Formaldehyde from Au(111)

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In recent decades, molecular beam scattering experiments—primarily on diatomic molecules—have demonstrated signatures for distinct mechanisms of vibrationally inelastic energy exchange between molecules and metal surfaces. Namely, mechanical kinetic energy-vibration (T - V) coupling and electronically non-adiabatic electron hole pair-vibration (eHP- V) coupling are both important processes that exhibit qualitatively different dependences on incident kinetic energy and surface temperature. In polyatomic molecules, these mechanisms might happen simultaneously and mode selectively. In formaldehyde, the eHP- V mechanism is expected to interact *exclusively* with the CO stretch (ν_2). We have designed and tested a new implementation of Dirk Schwarzer's nozzle to generate molecular beams of formaldehyde by cracking and drying a sample of paraformaldehyde directly inside the nozzle tip. We have developed a new 1+1' REMPI scheme for rotationally-resolved detection of formaldehyde via the resonant $\tilde{A} \leftarrow \tilde{X}$ transition. We characterize this scheme by studying the spectrum of our incoming formaldehyde beam, as well as a rotationally-inelastic trapping-desorption channel of formaldehyde scattered from gold surfaces. Finally, we have observed rotationally-inelastic direct scattering of formaldehyde from Au(111). We report the kinetic energy loss and rotational energy distribution as a function of incident kinetic energy. We discuss planned experiments for investigating vibrationally-inelastic scattering channels.

Gas-phase high-temperature synthesis of catalytic materials

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The current state-of-the-art in the field of gas-surface scattering dynamics calls for expanding its unique experimental approaches to the investigation of fundamental molecular interactions at complex surfaces. Despite the demonstrated importance of simple crystalline surfaces, molecular interactions with which presently dominate the field of surface dynamics, it is widely understood that more sophisticated systems are needed to make progress on realistic problems in surface chemistry. High-temperature gas-phase synthesis of solid phase functional materials (e.g. combustion synthesis and chemical vapor deposition, CVD) serves wide range of applications on laboratory and industrial scales. One of the most widespread utilizations of gas phase synthesis is the fabrication of catalytic materials. Gas-phase synthesis provides a clean, scalable and self-purifying method for catalyst production that allows to significantly reduce the number of steps required for catalyst preparation when compared to classical wet chemistry methods such as co-precipitation, impregnation, sol-gel and hydrothermal syntheses. Moreover, with respect to purity and surface composition, particles and films generated via gas-phase synthesis are preferred compared to materials from wet synthesis with surface active additives. The combination of high temperature gas phase synthesis with mass selective molecular beam sampling merges the advantages of flame-assisted synthesis for bulk nanopowders such as process stability, wide size-tuning range (particle sizes from a few to ~ 500 nm are commonly accessible) and wide material library availability with those of mass-selective cluster beam deposition in terms of direct integration of nanoparticles on support substrates, monodispersity, substrate coverage control, spatial resolution and spatial distribution manipulation, thus providing an excellent opportunity to obtain samples for systematic studies of molecular interaction with catalytic surfaces. Our research has a two-fold character: the synthesis of catalytic materials by flame pyrolysis and CVD is an aim and also an instrument of our studies. On the one hand, laser-based and molecular beam sampling methods are developed [1,2] and utilized to reveal the detailed mechanisms underlying the synthesis process, and on the other, processes are optimized to obtained catalysts with desired functionalities. I will present several recent studies devoted to development diagnostic techniques allowing to address the challenges of probing both gas-phase and solid particulates in flame aerosol and will demonstrate on the example of iron oxide nanoparticles how combination of several diagnostics techniques was used to reveal unexpected mechanistic details of particle formation [3,4]. In another example I will present our recent achievements in CVD synthesis of vanadium dioxide (VO₂) films and highlight some of preliminary molecular beam/surface scattering experiments where energy exchange in interaction of vibrationally excited NO molecules with topologically rough VO₂ films deposited on silica substrates was examined.

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HgCdTe APDs for low photon number IR detection.

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HgCdTe APDs have opened a new horizon in photon starved Infra-Red (IR) applications due to their exceptional performance in terms of high linear gain, low excess noise and high quantum efficiency which have enabled single photon detection with high efficiencies from the uv up to the mid-IR range [1]. These properties, which enables the detection of a few number of photons with an ultra-low loss of the information contained in the photon flux, equivalent to an effective quantum efficiency of 70 to 80 %, combined with a large linear dynamic range offers an unique observation window of low photon number temporal and/or spatial information in the IR range. The performances of HgCdTe APDs, such as quantum efficiency, gain, dark noise, excess noise, response time will be discussed as a function of the spectral sensitivity of the APD that can be tuned by varying the composition of the compound semi-conductor [2-4]. A particular attention will be given to the discussion of the performance of APDs which are sensitive at longer mid-IR wavelengths, approaching the 10 μm range, and under which condition low photon number, down to a single photon, detection can be achieved at such wavelengths.

The HgCdTe APD prototype detectors have been developed for applications such as time of flight 3D imaging, astrophysics, deep space optical telecommunications, photoluminescence life time measurements, gas LIDAR and single photon detection. The presently achievable detector performance will be illustrated through a number of such application demonstrations made with prototype imaging arrays and single element detectors dedicated to extract the spatial and/or temporal information low photon number IR flux.

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Experimental results of hydrogen recombination on Au(111) surface

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In this work, we investigated the recombinative desorption of hydrogen from Au(111) surface using a hydrogen permeation technique. We used the principle of detailed balance to extract quantitative information about dissociative adsorption dynamics from the time-of-flight (TOF) distributions of molecules desorbed from the surface. We detected highly translationally excited hydrogen molecules of different internal states. The translational excitation decreased as the vibrational excitation increased. We also investigated the dependence of the reaction probability on rotational, vibrational and translational energy of hydrogen molecules. The angular distributions of products were remarkably peaked at the surface normal, implying high activation energy of adsorption. This should be an ideal benchmark data-set for testing modern theories of surface chemistry. Specifically, we would like to know whether electronically adiabatic theory could succeed to describe these dynamical measurements. This system was chosen because of recent chemi-current measurements attributed to the recombinative desorption of hydrogen on Au.^[1, 2] The investigations should help us to understand how well the Born-Oppenheimer approximation could work in a simple chemical reaction at a metal surface.

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The Density Functional Tight Binding approach for silver and bulk systems

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The Density Functional Theory based Tight Binding (DFTB) method [1] is an approximate quantum scheme derived from Density Functional Theory (DFT). It is a computationally efficient method for electronic structure calculation to be used in large scale simulations [2], for instance global structure optimization or on-the-fly molecular dynamics targeting statistical meaning and convergence of the dynamical observables [3]. The method is implemented either within finite systems schemes or with periodic boundary conditions schemes. The scope of the contribution will be (i) to briefly introduce the principles of the DFTB approach (ii) provide a survey of its ability and accuracy to treat silver and gold systems, from molecule and clusters up to nanoparticles of a few hundreds atoms and the bulk [4], emphasizing structural, energetical and electronic properties (ionization, electron attachment) (iii) to discuss the perspectives to treat interactions of atoms or molecules with noble metal clusters and surfaces and to include excitation in the simulations and schemes in DFTB beyond the Born-Oppenheimer approximation.

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Molecular Beam Scattering from Ultrathin Metallic Films of Ag/Au(111)

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Quenching electronically and vibrationally excited carbon monoxide at metal surfaces using velocity controlled molecular beams

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When scattering highly vibrationally excited NO($v = 18$) from a low-work function surface, the molecules lose several quanta of vibrational energy during the collision.^[1] The large-amplitude vibration is converted into electronic excitation in the metal leading to electron emission from the surface.^[2] This non-adiabatic behavior of NO molecules on metal surfaces can be explained by a mechanism involving an electron transfer from the metal to the NO molecule, forming a transient anionic species which subsequently decays by auto-detachment of an electron.

Our work focuses on molecule-surface scattering events that involve electron emission. In order to compare the dynamics of CO and NO at metal surfaces, we plan to scatter highly vibrationally excited CO($v = 17$) from a low-work function surface. In addition, we investigate the de-excitation of electronically excited, metastable CO ($a^3\Pi_1$) at both clean and adsorbate covered metal surfaces, a process which also proceeds via an electron transfer process.^[3] In particular, we want to determine the importance of the velocity of incident molecules on the electron emission probability, as an inverse velocity dependence of vibrationally promoted electron emission was observed for NO($v = 18$) on metal surfaces.^[4] For these velocity dependent measurements, we use a machine that combines a Stark decelerator for accurate velocity control of molecular beams with a state-of-the-art beam-surface scattering chamber. We recently achieved to do automated velocity scans, which allow to repeat molecule-surface scattering experiments for different velocities of the CO beam at a 10 Hz rate.

I report on our progress in developing the required methods for conducting velocity dependent molecule-surface scattering experiments.

- [1] Y. Huang *et al.*, *Science* **2000**, 290, 111.
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Full-Dimensional Quantum Dynamics Studies of Chemisorption of Water on Static Metal Surfaces

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Abstract

Water dissociation on metal surfaces to form chemisorbed OH(ads) and H(ads) is an essential part in the steam reforming process used on a large scale by the chemical industry to convert methane (CH₄) to hydrogen. It is, therefore, of great importance to have an in-depth understanding of the adsorption and dissociation process. Theoretically, with only one relatively heavy atom (O atom) involved, the scattering of this three-atom molecule on a static surface is an ideal system to carry out quantum dynamics studies beyond diatomic molecules such as H₂. Recently, reduced dimensionality quantum dynamics calculations have been performed with the number of degrees of freedom included up to six, and predicted strong mode specificity, bond selectivity, and steric effects for water on Cu(111) and Ni(111). Semi-quantitative agreements were achieved between some of theoretical results and experiments. In this talk, I will present some of our recent progress made on quantum dynamics studies of chemisorption of water on static metal surfaces with full nine degrees of freedom included. Our calculations provide benchmarks for testing various approximations used to estimate chemisorption probabilities based on reduced dimensionality studies.

Short biography: Prof. Dong Hui Zhang was born in 1967 in Zhejiang, China, and received his B.S. degree in physics from Fudan University, China, in 1989. He earned a Ph.D. in chemical physics from New York University in 1994. After working as a postdoctoral research fellow in New York University and the University of Chicago, he joined the National University of Singapore in 1997. He moved to Dalian Institute of Chemical Physics, CAS, in 2006. His primary research area is in theoretical and computational studies of chemical reactions in the gas phase and gas-surface interface. He has published more than 180 peer-review papers, with 8 published in Science.

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