SCHLOSS RINGBERG

May 20 to 23, 2015

Symposium on Dynamics at Surfaces

Wednesday May 20th	
16:00-18:00	**Arrival and registration**
18:00-19:30	**Dinner**
19:30-20:00	Prof. Alec Wodtke (Göttingen): *Welcoming Remarks and Overview*.
20:00-21:00	Prof. Daniel J. Auerbach: *Desorption measurements as a probe of dynamics at surfaces*
21:00-	**Poster Session I**
Thursday May 21st	
8:00-9:00	**Breakfast**
Session Chair:	Prof. Dirk Schwarzer
9:00-10:00	Prof. Geert-Jan Kroes: *Quantum and classical dynamics of reactive scattering of* H_2 *from metal surfaces*
10:00-10:30	**Coffee Break**
10:30-11:00	Dr. Alexander (Sascha) Kandratsenka: *Independent-electron surface hopping theory: necessity of the correct PES to reproduce the experimental data*
11:00-11:30	Dr. Vanessa Bukas: *"Hot" adatoms hopping: phononic dissipation & equilibration dynamics from first-principles*
11:30-12:00	Dr. Dan Harding: *Surface scattering experiments using spatial imaging detection*
12:00-13:30	**Lunch Break**
Session Chair:	Dr. Tim Schäfer
13:30-14:00	Yvonne Dorenkamp and Hongyan Jiang: *Inelasticity in hydrogen atoms scattering from Au(111): The role of electron-hole-pair excitation*
14:00-14:30	Bastian Krüger: *Surface scattering of highly vibrationally excited NO from Au(111): Time-of-flight experiments and final rotational state distributions*
14:30-15:00	Svenja Janke: *An accurate full-dimensional potential energy surface for H at Au(111): The importance of nonadiabatic electronic excitation in energy transfer and adsorption*
15:00-18:00	**Free time**
18:00-20:00	**Dinner**
20:00-21:00	Prof. John Tully: *Energy transfer at surfaces*
21:00-	**Poster Session II**

Friday May 22nd	
8:00-9:00	**Breakfast**
Session Chair:	Prof. Jörg Schroeder
9:00-10:00	Prof. Daniel Farias: *Helium diffraction and low-energy phonon dynamics of graphene on metals*
10:00-10:30	**Coffee Break**
10:30-11:00	Jan Geweke: *State-to-state scattering of HCl molecules from Au(111) surfaces: Investigating absolute vibrational excitation probabilities (VEPs)*
11:00-11:30	Dr. Simon Rittmeyer *Quo vadis electronic fiction? Assessing vibrational lifetimes of diatomics beyond the independent atom approximation*
11:30-12:00	Jannis Neugebohren *Imaging of molecular beam scattering at Beamer II*
12:00-13:30	**Lunch Break**
Session Chair:	Dr. Oliver Bünermann
13:30-14:00	Roman Wagner: *Production of Stark decelerated CO molecules in the ground electronic state*
14:00-14:30	Sven Kaufmann: *Associative Desorption of Hydrogen Isotopes from Copper surfaces*
14:30-15:00	Dr. Sam Meek: *Toward precision infrared spectroscopy on molecules*
15:00-18:00	**Free time**
18:00-20:00	**Dinner**
20:00-21:00	Prof. Eckart Hasselbrink: *What have thin metal film heterostructures so far told us about non-adiabaticity in surface chemical reactions*
21:00-	**Poster Session III**
Saturday May 23rd	
8:00-9:00	**Breakfast**
Session Chair:	Prof. Theo Kitsopoulos
9:00-10:00	Prof. Gert von Helden: *IR spectroscopy on peptides and proteins after ion mobility selection and in liquid helium droplets*
10:00-10:30	**Coffee Break**
10:30-11:00	Dr. Kumar Sumit: *Direct measurement of vibrational lifetime of CO on Au(111)*
11:00-11:30	Prof. John Moore-Furneaux: *A system for precise spectroscopy with examples from* $^{130}TE_2$

11:30-12:00	Dr. Tim Schäfer: *Vibrational relaxation of NO in collisions with different surfaces: “All that glitters is not gold” (“Es ist nicht alles Gold, was glänzt”)*
12:00-12:15	Prof. Alec Wodtke: *Closing Remarks*
12:15-13:30	**Lunch Break**
14:00	**Departure**

SPEAKER'S

ABSTRACTS

Desorption Measurements as a Probe of Dynamics at Surfaces

Daniel Auerbach
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Scattering experiments with control of the angle, kinetic energy, internal state, and orientation of incident molecules provides a wealth of information on dynamics of molecule-surface interactions. Often, however, it is difficult to prepare incident beams over the range of kinetic and internal energies which are of significance to the process under study. Measurements of angle, energy, and state resolved desorption complement the use of adsorption measurements. If we study associative desorption of Hydrogen, for example, we automatically get information on the full range of angles, kinetic energies, vibrational, and rotational states that play a significant role in the process of dissociative adsorption. In this talk, I will review and give examples of the use of desorption to probe chemical dynamics at surfaces. The talk will be mainly historical with some details of recent work on the interpretation of desorption data.

Quantum and classical dynamics of reactive scattering of H_2 from metal surfaces.

Geert-Jan Kroes
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H_2-metal surface scattering systems serve as benchmark systems for the accurate determination of the interaction of molecules with transition-metal surfaces. I will discuss applications of dynamics methods to this topic, looking at the following questions: (i) To what extent is the scattering of H_2 from metal surfaces affected by electronically non-adiabatic effects (electron-hole pair excitation (ehps)) and by phonon effects, (ii) what is the effect of pre-coverage of the metal on the dissociation dynamics, (iii) what is the effect of defects like steps, (iv) are there similarities between dissociative chemisorption and diffraction at (hyper)thermal incidence energies (up to 0.85 eV) on the one hand, and dissociative scattering and diffraction at fast grazing incidence on the other hand, and (v) to what extent can we now describe molecular beam scattering experiments and associative desorption experiments with chemical accuracy, using a semi-empirical implementation of the specific reaction parameter approach to density functional theory (SRP-DFT).

Independent-Electron Surface Hopping Theory: necessity of the correct PES to reproduce the experimental data

Alexander Kandratsenka
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We study in detail a recently developed first-principles approach, which employs an Independent Electron Surface Hopping (IESH) algorithm to model the nonadiabatic dynamics on a Newns-Anderson Hamiltonian derived from density functional theory. This approach has been successful when compared to previously reported NO/Au scattering data. The new experiments show that vibrational relaxation probabilities increase with incidence energy of translation. The theoretical simulations incorrectly predict high relaxation probabilities at low incidence translational energy. We show how the adiabatic interaction potential influences the description of electronically nonadiabatic vibrational energy transfer in molecule-surface collisions.

“Hot” adatoms hopping: Phononic dissipation & equilibration dynamics from first-principles

Vanessa Jane Bukas and Karsten Reuter
Technische Universität München, Garching, Göttingen

Exothermic surface chemical reactions may easily release several electron volts of energy. This challenges first-principles based dynamical simulations to account for adequate energy dissipation channels. Focusing on phononic dissipation, a novel embedding scheme for metallic substrates allows for energy to be dissipated out of a quantum-mechanically described reaction zone and into a computationally undemanding, yet reliably described, extended bath1. In the application to oxygen dissociation over Pd(100) this predicts “hot” dissociation fragments traveling ballistically over several lattice constants as a consequence of non-immediate energy transfer to the metal surface. This raises fundamental questions when considering the short-range transient mobility measured experimentally for the similarly exothermic O2/Pd(111) reaction2. Here, we elucidate phononic dissipation within the (111) as compared to the (100) surface and explain the small net displacement by revealing randomized trajectories of hyperthermal hops between neighboring hollow sites due to scattering by the strongly corrugated chemisorption potential. We therefore invalidate the simplistic physical picture implying a minimal number of hops for small adatom separations and anticipate further paradigm shifts required to accommodate such “hot chemistry”, for example, in our current understanding of heterogeneous catalysis.

1 J. Meyer and K. Reuter, Angew. Chem. Int. Ed., 53, 4721 (2014).
2 M.K. Rose et al., Surf. Sci., 561, 69 (2004)

Surface Scattering Experiments using Spatial Imaging Detection

Dan J. Harding, J. Neugebohren, D. J. Auerbach, T. N. Kitsopoulos and
A. M. Wodtke
Georg-August University, Göttingen and Max Planck Institute for Biophysical Chemistry, Göttingen

We describe the recent progress made on Beamer 2, in particular, molecular beam - surface scattering experiments using spatial imaging. The imaging set up, combined with laser ionization, allows us to directly measure the speed and angular distributions of scattered molecules. The scattering of NO from Au(111) was used to test the speed calibration and resolution of the set up, which compare well with state-of-the-art IR-tagging time-of-flight measurements. We present results for the scattering of N_2 from Au(111), not possible with IR-tagging experiments, allowing us to probe the (anti-)correlation between final translational and rotational energy of the molecules.

Inelasticity in Hydrogen atom scattering from Au(111): The role of electron-hole-pair Excitation

Yvonne Jeannette Dorenkamp, Hongyan Jiang, Alec Wodtke, and Oliver Bünermann
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Tammannstr. 6, 37077 Göttingen, Germany

Atomic and molecular beam surface scattering experiments are of fundamental importance to investigate the dynamics of energy conversion at surfaces. The presented experimental setup focuses on the atomic energy transfer to solids by scattering Hydrogen atoms from a single crystalline surface.

A monochromatic Hydrogen atom beam with tunable kinetic energy is formed by laser photolysis of Hydrogen Iodide and hits a Au(111)-surface at defined incident angles. Using Rydberg Atom Tagging the final kinetic energy and scattering angle of the scattered Hydrogen atoms is measured.

Hydrogen atom scattering from Au(111) is investigated at different experimental conditions (e.g. incidence kinetic energy, incidence and scattering angles) to obtain a detailed picture of the scattering process. By comparison to theoretical calculations the role of electronically non-adiabatic processes and penetrating collision is discussed.

Surface scattering of highly vibrationally excited NO from Au(111): Time-of-flight experiments and final rotational state distributions

Bastian C. Krüger[1], Hannes C. Gottschalk, Niklas Henning, Nils Bartels[1], Alec M. Wodtke[1,2], Tim Schäfer[1]

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Nitric oxide (NO) can lose a huge amount of vibrational energy when scattered from the (111) surface of a gold single crystal.[1] The most probable scattering channel for an initially highly vibrationally excited molecule (vibrational quantum number $v = 16$) involves the transfer of nearly 2 eV of energy from the molecule to the surface.[2] Due to the large energy mismatch between the vibrational spacing in NO and phonon modes in gold it can be definitely ruled out that this process proceeds purely mechanically. Rather, it is thought to proceed non-adiabatically involving the transient formation of the nitric oxide anion and resulting in the excitation of electrons.

In order to quantify the exact amount of energy transferred to the surface, the molecules final translational and rotational energy has to be known in addition to the final vibrational energy. Time-of-flight experiments on initially highly vibrationally excited NO ($v = 11$) similar to previous experiments on NO ($v = 3$)[3] allow us to reveal the coupling between these molecular degrees of freedom.

We find that the molecules are scattered directly from the surface regardless of the vibrational energy loss. Only a small amount of the vibrational energy lost during the collision is converted into final translational energy of the vibrationally relaxed molecule. The more vibrational relaxation occurs, the higher is the final translational energy. Furthermore, stronger rotational excitation of the scattered molecule is correlated with a lower final translational energy. This means that the incidence translational energy is partly converted to final rotational energy. Based on these findings, we can determine for all possible scattering events how much energy is transferred to the surface. Furthermore, the results allow us to report exact final vibrational and translational state distributions in terms of flux.

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[3] K. Golibrzuch, P. R. Shirhatti, J. Altschäffel, I. Rahinov, D. J. Auerbach, A. M. Wodtke, C. Bartels, *J. Phys. Chem. A* **2013**, *117*, 8750-8760.

An Accurate Full-Dimensional Potential Energy Surface for H at Au(111): The Importance of Nonadiabatic Electronic Excitation in Energy Transfer and Adsorption

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We have parameterized a potential energy surface for H atoms interacting with *fcc* gold based on Effective Medium Theory. It describes both the interatomic forces and electron densities in *full dimension* with the accuracy of the *ab initio* energies used in its construction. The analytic expression for the total energy contains the embedded electron density leading to a self-consistent approach to simulating nonadiabatic trajectories. We find that electron-hole pair excitation is the most important energy loss pathway for the H atom and determines the probability and mechanism for its adsorption.

Energy Transfer at Surfaces

John Tully
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Chemical processes at solid surfaces exhibit much of the complexity of condensed phase chemistry, including multiple local environments and energy scrambling via a heat bath. As a result, our understanding of the detailed pathways of surface chemical reactions has been quite murky until relatively recently. Modern experimentation is changing the situation. Gas-surface encounters are now being examined with the same exquisite detail that has been so illuminating for gas phase chemistry, with reactants prepared with controlled velocities in specific quantum states, coupled with state-selective detection of gas phase products. These experiments have revealed unexpected behavior that presents significant challenges to theory. This talk will examine some of the underlying principles that govern the exchange of energy between molecules and surfaces. Examples will include non-thermal desorption, “chemistry with a hammer”, vibrational energy pooling, and the role of nonadiabatic electronic transitions.

Helium Diffraction and Low-Energy Phonon Dynamics of Graphene on Metals

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Helium atom scattering (HAS) is a well-established technique for investigating the structural and dynamical properties of solid surfaces [1]. Because of the low energies used (10-100 meV), monoenergetic beams of neutral He atoms probe the top most surface layer of any material in a completely nondestructive manner, and can be used equally well to investigate insulating or conducting materials. HAS provides high-resolution (0.5 meV) spectral information in the acoustic phonon region (0-50 meV) [2] and is highly sensitivity to surface defects; detection of defect concentration down to 0.005 ML is routine [3]. From the phonon dispersion curves it is possible to draw conclusions about the strength of the graphene-substrate interaction, which is a difficult piece of information to get with other experimental techniques.

After briefly describing the HAS method, I will summarize recent He-diffraction and phonon measurements of graphene grown on Ru(0001), Ni(111) and Cu(111) surfaces obtained in our laboratory. In the final part, I will briefly describe the project of the Neutral Microscope (NEMI), which uses a focused beam of neutral He atoms as imaging probe [4,5]. The potential lateral resolution of NEMI is 20 nm. It would allow real–space investigations of fragile samples, without distortions due to charging effect. Both the inertness and the high reflectivity measured for epitaxial graphene suggest that it is perhaps the ideal external surface for the NEMI mirror [6].

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State-to-state scattering of HCl molecules from Au(111) surfaces: Investigating absolute vibrational excitation probabilities (VEPs)

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The development in computational chemistry has led to more advanced theoretical predictions than ever before. Nevertheless, since *ab initio* calculations are limited to very simple systems, it is of extreme importance to challenge their results by accurate measurements of com-paratively simple systems, such as small (diatomic) molecules, monocrystalline surfaces and ultra-high vacuum (UHV) conditions.

Based on previous experiments by Ran *et al.*[1] and theoretical predictions by Liu *et al.*[2] we currently examine nonreactive, inelastic as well as reactive scattering of HCl from Au(111) surfaces, with a focus on vibrational excitation and dissociative chemisorption.

We employ molecular beam surface scattering under UHV conditions. Using a narrow-linewidth, high-intensity pulsed IR laser, we can excite HCl molecules in the incoming molecular beam to vibrational states v=1,2; alternatively we can use the abundant v=0. After scattering, the molecules are detected using REMPI *via* the $E^1\Sigma^+$ state.

Absolute excitation probabilities can be derived from a comparison of the intensities in the REMPI spectra of the relevant rovibrational states after correction for experimental conditions such as laser intensity and detection efficiency and intrinsic properties like Franck-Condon factors. Preliminary results suggest an increase in excitation probability with increasing translational energy of the incident molecules (as reported before) and a higher VEP for v=1→2 excitation than for v=0→1 excitation. Furthermore, temperature dependent measurements hint at a nonadiabatic mechanism involving a coupling of electronic and vibrational states.

[1] Ran, Q.; Matsiev, D.; Auerbach, D.; Wodtke, A. *Phys. Rev. Lett.* **2007**, *98*, 237601.
[2] Liu, T.; Fu, B.; Zhang, D. H. *J. Chem. Phys.* **2013**, *139*, 184705.

Quo vadis electronic friction? Assessing vibrational lifetimes of diatomics beyond the independent atom approximation

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The quest for a both accurate and numerically efficient first-principles-based treatment of electronically non-adiabatic adsorbate dynamics on metal surfaces is still ongoing.A promising candidate in the regime of weak non-adiabaticity is the concept of electronic friction within the local density friction approximation (LDFA) [1]. The latter has recently been applied to describe the nuclear dynamics of "real" adsorption problems even directly in ab initio molecular dynamics simulations [2, 3].

The numerical efficiency of this approach stems from an intrinsic decomposition and mapping of the interacting system to independent atoms individually embedded in a free electron gas. This inherent simplicity, entirely disregarding the interaction of the adsorbate with the metal substrate, as well as intra-molecular interactions, has raised conceptual concerns about the accuracy of this theory [4]. On the other hand, it is not clear how much these approximations actually affect the description of "real" observables. In this regard, vibrational lifetimes of high-frequency adsorbate modes on metal surfaces provide a sensitive measure to gauge a non-adiabatic theory [5].

In order to clarify this picture, we thus target this observable applying the LDFA for several well-studied systems and discuss it by comparing the results to other theoretical approaches as well as experimental data. Moreover, in an attempt to take both molecular effects and surface mobility into account, we introduce a simple and computationally very efficient strategy to extend the LDFA beyond the hitherto inherent independent-atom and frozen-surface approximation. Lastly, we critically compare the electronic friction-based implications to results from a recently extended perturbative approach rooted in time-dependent density functional theory (see Ref. [6] and references therein).

References

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Imaging of molecular beam scattering at Beamer II

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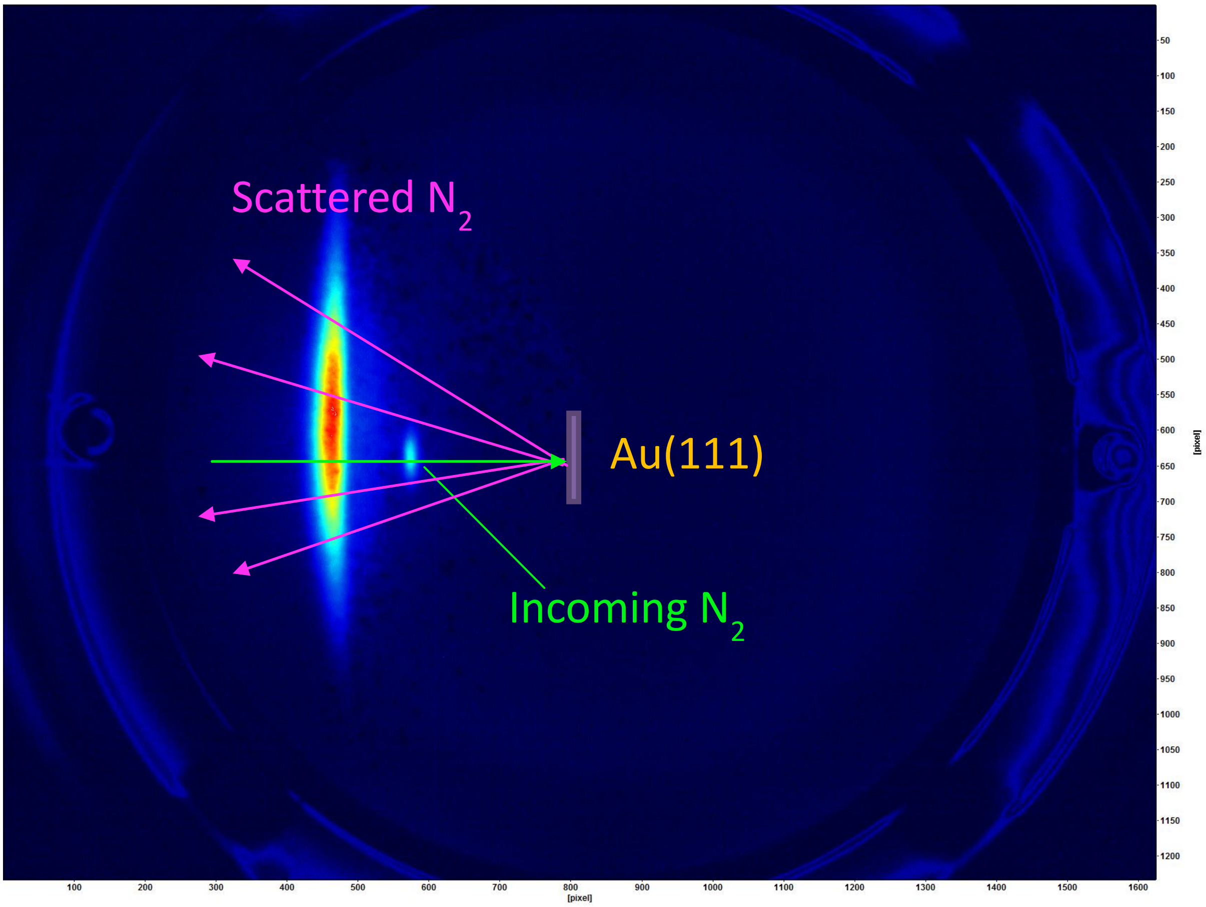
In the past months we have successfully applied spatial imaging to a surface scattering experiment. The functionality of the experiment has been proved by comparing spatial imaging data of NO scattering from Au(111) to IR-tagging time-of-flight experiments done at Beamer I. Furthermore, scattering data of N_2 from Au(111) that was previously unavailable has been taken at Beamer II. Additionally, this poster will cover the latest experimental changes and an outlook.



Production of Stark Decelerated CO Molecules in the Ground Electronic State

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Since Stark deceleration of molecular beams was accomplished in 1999 for the first time[1], it has been applied in many different experiments providing deeper insights into fields such as high resolution spectroscopy, trapping polar molecules in free space and crossed beam scattering experiments.[2] In order to perform surface scattering experiments using velocity controlled molecular beams, a new apparatus combining a Stark decelerator with a beam-surface scattering setup was built in collaboration with the group of Prof. G. Meijer.[3] By quickly switching strong electric fields, the decelerator is capable of tuning the velocity of a beam consisting of metastable CO in the $a^3\Pi_1$ state. Compared to ground state CO, metastable CO possesses a sufficiently high dipole moment for Stark manipulation. So far, surface collision experiments were performed using velocity controlled molecular beams of metastable CO.[4-6] In the next step, we plan to scatter Stark decelerated beams of CO in the ground electronic state. Thus, the CO molecules need to be transferred back to the $X^1\Sigma^+$ state after having exited the decelerator. I will report on our attempts to apply optical pumping schemes for production of Stark decelerated, ground state CO molecules. We examined the implementation of two established pumping schemes in our experimental setup. In both approaches, the transfer of the metastable species to the ground state proceeds *via* perturbed levels in the $A^1\Pi$ state. Blokland *et al.* reported on laser induced fluorescence to the vibrational and rotational ground state of $X^1\Sigma^+$ exploiting the $A^1\Pi(v = 1) / d^3\Delta(v = 5)$ level.[7] Bartels *et al.* reported on stimulated emission pumping from the laser-prepared $A^1\Pi(v = 8) / e^3\Sigma^-(v = 12)$ level leading to highly vibrationally excited ground state CO, a pumping scheme known as *pump-pump-perturb-dump*.[8]

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Associative Desorption of Hydrogen Isotopes from Copper surfaces

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Here, we quantitatively investigated the post-permeation associative desorption reaction of hydrogen and its isotopes from copper surfaces, using the (2+1)-REMPI technique allowed for quantum state resolved measurement of the kinetic energy distribution of the desorbing flux. In accordance with the principle of detailed balance the sticking probability functions have been determined for the investigated systems in a quantum state resolved manner, which provides an ideal benchmark system for theory in surface dynamics.

Toward Precision Infrared Spectroscopy on Molecules

Sam Meek
Max-Planck-Institut für biophysikalische Chemie, Göttingen, Germany

In my talk, I will report on the progress of the precision infrared spectroscopy experiment. We have been developing a 1064-nm pumped CW OPO with idler tunable between 4.17 and 2.5 µm (2400 - 4000 cm-1) with a few watts of output power. To stabilize the OPO to sub-kHz linewidth, we have also been developing an optical frequency comb system that will make use of a narrow-linewidth 1064/532 nm CW reference laser. The reference laser will be stabilized to an iodine transition with high precision, improving both its short and long timescale performance. I will present our progress on constructing the frequency comb and the iodine reference setup. Finally, I will show first measurements of a diode laser system which will be used to detect OH radicals using LIF at 308 nm.

What Have Thin Metal Film Heterostructures So Far Told Us About Non-Adiabaticity in Surface Chemical Reactions

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The excitation of electron-hole-pairs in a substrate in the course of a chemical reaction catalysed by a metal surface is direct evidence for the release of chemical energy due to non-adiabatic processes. Besides this fundamental importance, the direct conversion of chemical energy into electrical may be considered as a source of electrical power provided that the charges can be separated. It is an attractive concept whose potential is yet largely unexplored. Devices built with thin metal films as top layers in metal-insulator-metal or metal-oxide-semiconductor structures may serve for this purpose. We have fabricated devices consisting of a 10 – 20 nm thin metal layer (Au, Ag, Pt) deposited on top of oxidized Ta, Al or Si substrates. A typically 2-3 nm thin oxide layer serves as barrier. However, electrons excited by chemical reactions at the vacuum interface of the top layer may tunnel through this barrier, which can be detected as a macroscopic current. Such a current has been observed under UHV conditions for the reaction of hydrogen on and with Au or Ag surfaces and been characterized regarding the energy distribution of the underlying electronic excitations.

Bulb experiments studying for e.g. the water formation reaction (2 H_2 + O_2 → 2 H_2O) on Pt at mbar pressures are much more complicated to interpret. The currents that are observed may be non-adiabatic, thermionic or thermoelectric in nature. There have been published a large number of reports suggesting a non-adiabatic "chemicurrent" under such reaction conditions. I will report studies carried out in my laboratory attempting to disentangle the various aspects.

IR spectroscopy on peptides and proteins after ion mobility selection and in liquid helium droplets

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IR spectroscopy has become a frequently used tool to characterize gas-phase peptides and proteins. In many experiments, ions are m/z selected, irradiated by intense and tunable IR light and fragmentation is monitored as a function of IR wavelength. The presence of different conformers can, however, complicate the interpretation, as the resulting spectra represent the sum of the spectra of the individual components. We constructed a setup, in which ion mobility methods are used to obtain m/z selected ions of defined shape on which are then further investigated by IR spectroscopy. First results on peptide aggregates are presented and for some of those, the IR spectra show a transition from helical or random coil to beta sheet structures.

In a different experiment, peptide or protein ions are captures in liquid helium droplets prior to IR spectroscopic investigation. The conditions inside a helium droplet are isothermal at 0.38 K and the interaction between the helium matrix and the molecules are weak so that only small perturbations on the molecule are expected. IR spectra for m/z selected peptides with up to 10 aminoacids and proteins containing more than 100 aminoacids have been measured. The spectra of the smaller species show resolved bands of individual oscillators, which can be used for structure assignment. For the larger species, band envelopes are obtained and for the case of highly charged proteins, a transition form helical to extended structures is observed.

Direct measurement of vibrational lifetime of CO on Au(111)

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Surface science plays a significant role in the discipline covering from physics, chemistry to biology wherever the understanding of surface and interfaces is necessary. One of the major interests in surface science is photo-driven reactions/processes. Vibrational lifetime of adsorbate on substrate surface microscopically defines these surface reactions by influencing the intermediate steps.

Vibrational lifetime of an adsorbate significantly depends upon property of substrate surface. When CO adsorbed on semiconductor or insulator, high energy vibrational mode interacts with low-frequency phonons (lattice waves) and vibrational lifetime is in nanosecond timescale.[1] Vibrational relaxation of excited (v=1) state of chemisorbed CO on metals (Cu or Pt) takes place in the order of picosecond due to electron-mediated non-adiabatic vibrational energy transfer to the metal.[2,3] In contrast, vibrational lifetime of excited (v=1) state of physisorbed CO on NaCl is in millisecond order (~4.3 ms) due to inefficient coupling of CO vibrations with surface phonons.[4] Thus, vibrational lifetime of CO on Au(111) will depend on the surface property. Gold, being the noblest metal[5], prefers a very weak adsorption of CO. In contrast, a few groups have reported some evidences of chemisorption of CO on Au at elevated pressure.[6,7] But there is no report of direct measurement of vibrational lifetime of CO on Au in the literature.

In the current work, we will directly measure the vibrational lifetime of CO on Au(111). The measurement will be performed with the help of femtosecond pump-probe IR spectroscopy. We will record transient IR absorption spectra of vibrationally excited CO (v=1) on Au(111) as a function of adsorbate coverage area i.e., surrounding of adsorbate, surface temperature and laser fluence. The study leads to a requirement of very low surface temperature as TPD study shows that CO monolayer on Au(111) is stable below 50 K. Therefore, we are building an experimental setup with entirely new design, where we are hoping to attain a very low surface temperature (~10 K).

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A System for Precise Spectroscopy with Examples from $^{130}Te_2$

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In the group of Neil Shafer-Ray within the Homer L. Dodge Department of Physics and Astronomy at the University of Oklahoma, we wished to develop a system for making precise molecular spectroscopy measurements that could be used for setting limits of the electron’s electric dipole moment utilizing PbF. Such a goal sets severe limits on the requirements for the system. 1. It must be able to precisely ($1:10^{10}$) and accurately ($1:10^{8}$) scan approximately 1 GHz in a designated region near 442 nm (22,600 cm^{-1}, 678.3 THz). 2. It must be lockable for days without interruption with a precision of 1 MHz. 3. It must be absolutely re-lockable to within 10 MHz and this lock must be confirmable. 4. A second scanning mode accurate to better than 100 MHz with a much quicker and wider scanning range is desirable. 5. Minimal resources ($\leq$ \$ 15 k) were available. I will describe the resulting system, based on a Toptica TA-SHG Pro laser system and a stabilized ($1:10^{9}$ to $1:10^{10}$) HeNe laser available in the lab, which exceeds all the requirements. This system was validated using saturated absorption spectroscopy of $^{130}Te_2$. We were able to obtain results to previously unreported precision and accuracy. Much of this work is the subject of James Cocker’s dissertation.

Vibrational relaxation of NO in collisions with different surfaces: "All that glitters is not gold" ("Es ist nicht alles Gold, was glänzt")

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In recent years we extensively studied the vibrational relaxation of NO scattered from Au(111), where loss or gain of vibrational energy in collisions of an NO molecule with the single crystal gold surface proceeds via electron transfer. [1, 2] With the advent of new optical pumping and orientation methods, we were able to control all molecular degrees of freedom important to this electron transfer mediated process, providing the most detailed look yet into the inner workings of an electron transfer reaction and showing how to control its outcome.[3, 4]

We now extend these studies and focus on the interaction of highly vibrationally excited NO molecules with different surfaces like semiconductors (germanium) and other metals (silver). Analyzing our preliminary results we can already clearly state: "All that glitters is not gold".

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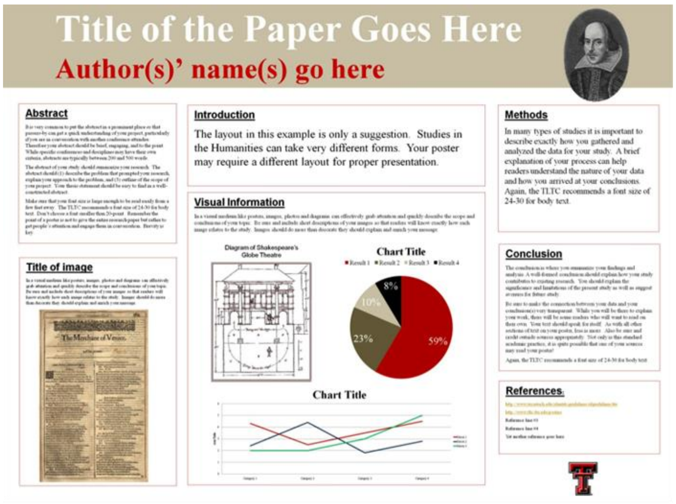

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Abstract

Introduction

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Methods

In many types of studies it is important to describe exactly how you gathered and analyzed the data for your study. A brief explanation of your process can help readers understand the nature of your data and how you arrived at your conclusions. Again, the TLTC recommends a font size of 24-30 for body text.

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Visual Information

Conclusion

References

POSTER

ABSTRACTS

Ab initio Molecular Dynamics Simulations of the NO Scattering from Au(111)

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The AIMD simulations of the NO molecule from the Au(111) surface are performed with several GGA functionals to study the NO molecule vibrational energy loss channels as well as its translational energy inelasticity. The influence of the initial NO vibrational state on the scattering events is also investigated. It is shown that the NO incidence energy losses agree qualitatively with the experimental results.

Further the NO/Au(111) interaction for different adsorption sites and orientations of the NO molecule is shown. Also the influence of different GGA functionals and supercells on the interaction is presented.

Rotational rainbow scattering with oriented molecules: “Rotational cooling upon vibrational relaxation” and the role of electron transfer

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In everyday language, the term “rainbow” refers to an optical and meteorological phenomenon caused by “reflection, refraction and dispersion” of light in water droplets and has been studied by fascinated scientists as early as 350 B.C.E. [1].

I want to convince the participants of the symposium of that the beauty of rainbows can also be found in surface dynamics. Here, so–called *rotational rainbows* [2] (non–thermal maxima in the rotational state distributions) are observed when diatomic molecules scatter from surfaces, e.g. when highly vibrationally excited NO scatters from Au(111) [3-5]. The rotational structure (which features pronounced rotational rainbows) strongly depends on the incidence orientation (N–atom first versus O–atom first collisions), the incidence translational energy and the probed incidence and final vibrational states. The results are explained with classical molecular dynamics simulations and provide deep insight into the dynamics of the collision system. In particular, a puzzling effect known as “rotational cooling upon vibrational relaxation” [7] is interpreted as resulting from a filtering effect of the rotational state distributions by vibrational relaxation.

An open and speculative question – which I would like to discuss on the symposium – is whether the rotational state distributions might be affected by multiple electron transfer events during the collision.

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Inelastic Hydrogen Atom Scattering: Status of the RAT-Experiment and First Results

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Obtaining an atomic-level understanding of the dynamics of energy conversion at surfaces remains a complex and challenging area of modern research in physical chemistry. A general strategy to this field follows the lessons of gas-phase bimolecular chemical dynamics, where simple model systems are studied experimentally with great care while theoretical simulations are developed.

One of the simplest systems to think of is Hydrogen atom scattering from a single crystalline surface. A new apparatus to experimentally investigate this model system with extraordinary precision is presented. Laser photolysis of Hydrogen Iodide is employed to produce a monochromatic Hydrogen atom beam with well-defined initial directions. The H-atom beam strikes a single crystalline surface held on a 6-axis manipulator. The kinetic energy and angular distributions of the scattered H-atoms are measured with extraordinary resolution employing Rydberg Atom Tagging. High control of experimental parameters as well as angular and energy resolved detection give a very detailed picture of the scattering process.

Experimental results for scattering H-atoms from a metal versus an insulator surface are presented. Drastic differences are observed. The results reveal that electron-hole-pair excitation is the dominate relaxation channel in case of a metal surface. Comparison to theoretical calculations support this interpretation. Furthermore, problems of the current experimental setup are discussed and planned modifications will be presented.

Fluorescence from Multilayer CO on NaCl studied with SNSPD

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Laser induced infrared fluorescence from multilayer CO (ca. 1100 layers) on NaCl is studied in both time-domain and frequency-domain using the superconducting nanowire single photon detector (SNSPD), and compared with the result from an InSb detector.

In frequency-domain, upon the initial vibrational excitation of CO ($v=0 \rightarrow 2$), infrared emissions from $v=3$ up to $v=28$ vibrational states are observed, implying the vibrational energy pooling process. Fluorescence signal are detected in a very broad frequency range (1850 – 6000 cm^{-1}) covering the one-quanta ($\Delta v=-1$), two-quanta ($\Delta v= -2$) and three-quanta ($\Delta v= -3$) emission regions.

In time-domain, dispersed fluorescence decay signal are measured for each observed emission states. The vibrational state dependent and non-single exponential decay signal suggests a very complicated decay kinetics. The signal decays on a millisecond time scale, and the decay time constant varies from 0.5 to 10 ms depends on the vibrational state. Furthermore, thanks to the fast time response (< 1 ns) of the SNSPD detector, a fast initial rise part of the fluorescence signal on a time scale of 10 µs after the laser pulse (10 ns) excitation is revealed, suggesting the time for vibrational energy pooling up from the initial $v=2$ state to high vibration states.

The experimental results here demonstrate an application of the new emerging SNSPD detector in molecular surface science. The fast time response and single photon sensitivity in the MIR range of SNSPDs provide an unique opportunity for studying vibrational dynamics of molecules adsorbed at solid surfaces at a monolayer or even a single-molecular level.

Temperature programmed desorption of weakly bound adsorbates on Au(111)

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We have performed temperature programmed desorption (TPD) experiments to analyze the desorption kinetics of physisorbed Ar, Kr, Xe, C2H2, SF6, N2, NO and CO on Au(111). We report the desorption activation energy (Edes) and show thermal desorption spectra for each gas/surface system.

Velocity-map-imaging employing strong-field ionization - A universal approach to molecular beam studies

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Today, the resonance enhanced multi-photon ionization technique (REMPI) is a commonly used ionization method in the detection schemes of molecular beam experiments. While REMPI can provide detailed information from the detection of molecules in a well-defined quantum state, it is at the same time limited to the detection of a certain compound in one particular state only, and not all molecules have good REMPI schemes. Non-resonant multi-photon ionization in an intense laser field provides an alternative to REMPI. Although it is an established technique [1], non-resonant MPI has become a topic of major interest for molecular beam experiments recently [2]. It is not quantum state selective, however, it gives a universally applicable ionization method for the detection of atoms and molecules. In combination with an ion- or photoelectron imaging setup strong field ionization can be a powerful tool for the investigation of chemical reaction dynamics [3].

In order to test the capability of non-resonant MPI, we examined the dissociative ionization of different small molecules using our recently described photofragment imaging spectrometer [4] in combination with intense Ti:sapphire laser pulses (775 nm, 150 fs, 750 µJ). Furthermore the application of strong-field ionization in the detection scheme of molecular beam surface scattering experiments is intended, e.g. to detect molecules that are poorly ionized with REMPI or to detect different species from a catalytic reaction at once.

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The role of atomic mass in translationally inelastic atom-surface scattering

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The interaction of atoms and molecules with metal surfaces are of both practical and theoretical interest. One very important question that has not yet been answered in detail is how translational energy carried by atoms or molecules couples to the surface during atom-surface scattering. In general, atoms may lose their translational energy to the vibrations of the metal surface atoms or to electron hole pair excitations of the metal. Comparison between recent experimental and theoretical works suggests that an adiabatic picture may totally fail in describing Hydrogen atom scattering from a Au(111) surface. The influence of the experimental parameters, including atomic mass and electron spin among others, on the scattering process and how they affect the energy dissipation still needs careful investigation.

On a newly built surface scattering machine, the differences in Hydrogen and Deuterium atom-scattering from a metal surface can be studied in detail. The H- respectively D-beam is formed by photolysis of HI or DI. After the scattering process the kinetic energies and scattering angles of H or D are measured using Rydberg-atom tagging. The presented results provide valuable benchmarks for the theoretical calculations.

Potential energy surface for Hydrogen Atoms on a Palladium(111) surface

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Potential energy surfaces have long been a useful means to simulate trajectories much faster than using *ab initio* molecular dynamics (AIMD). This work opens a new branch of theoretical considerations concerning molecular dynamics (MD) studies done in this group by investigating the interaction of a hydrogen atom with a Pd(111) surface.

The H-atom is placed at several hundred different positions in and above the surface and the energy of these configurations is evaluated by PBE, RPBE and PW91 DFT functionals. Next, the obtained energies corresponding to the grid-like structure of H-atom positions are fit to an analytical function resulting from Effective Medium Theory. This expression contains seven different physically meaningful parameters for each atomic species which can be used to derive structural properties. It also describes the potential energy surface used to run MD simulations by applying Newton's laws of motion.

The first attempts to fit the static energy grid already seem promising since the bulk modulus can be well reproduced and the RMS error is close to only 100 meV. Future fits are going to incorporate non-equilibrium geometries from AIMD trajectories. They raise the bar for an adequate Effective Medium fit but will also help to model trajectories at finite surface temperature.

Hydrogen-atom scattering from Platinum probed by "Rydberg Atom Tagging"

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We investigate translational inelasticity in hydrogen atom scattering from surfaces. Our goal is to understand the details of kinetic energy dissipation at surfaces. Currently, our experiments are focused on the H/Au(111) system. In future experiments, we want to extend our research to a variety of different materials. By carefully choosing the systems various parameters can be altered (e.g. mass, electronic and surface structure) and the influence of these parameters on the coupling can be studied. In comparison to Gold, Platinum is of special interest because the electronic density of states near the Fermi-energy is very different. Platinum has a significant contribution of *d*-electrons, while Gold only has contributions of *s*- and *p*-electrons. Hydrogen atoms might interact stronger with those *d*-electrons and more energy loss to the surface by means of electronically non-adiabatic coupling is expected. A similar trend was recently observed by Werdecker *et al.* for the non-adiabatic coupling of vibrational excitation of molecular nitrogen to electron-hole pair excitations of Gold and Platinum.

This poster will give an overview over the important Gold and Platinum properties as well as over the literature about scattering experiments from their surfaces. The current status of the experiment will be presented and necessary modifications to it will be discussed. Because of the high reactivity of Platinum, the planed experiments will need careful surface preparation to ensure defined and reproducible experimental conditions.

Preparation of Velocity Controlled Molecular Beams of Highly Vibrationally Excited CO

Artur Meling, Roman Wagner, Nils Bartels, Alec M. Wodtke and Tim Schäfer
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A scheme for the preparation of highly vibrationally excited CO in the $X^1\Sigma^+(v=17)$ state was implemented in a setup designed for molecular beam surface scattering experiments with a Stark decelerator. The vibrational excitation is achieved through optical pumping via two energetically low lying triplet states based on the work of Bartels et al. The implementation of this scheme in a Stark decelerator-based setup allows the complete control of the molecular beam velocity. Also a hexapole filter is placed in front of the decelerator allowing the removal of carrier gas and ground state CO from the molecular beam. This improves the quantum state purity and is an important step towards future surface scattering experiments.

Dissociation of HCl on Au(111) surface

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Quantum dynamics calculation looking at the dissociation of HCl on Au(111) surface has been reported [1]. These calculations show that the dissociation reaction is activated by both translation and vibration energy of the incident HCl molecules. We have undertaken an experimental study of this reaction and report some our important results here.

Clean Au(111) surface under UHV conditions was dosed with a pulsed, seeded molecular beam of HCl with incidence translation energies (E_i) up to 2.3 eV, prepared using a heated nozzle. Auger electron spectroscopy (AES) measurements carried out after the dosing show the presence of Cl atoms on the surface resulting from the dissociation of the incident HCl molecules.

The salient observations were:

(1) The dissociation probability depends on the E_i of the HCl molecules showing a threshold like behavior with the onset of dissociation observed at ~ 1.8 eV.
(2) Dissociation probability depends strongly on the incidence angle of the HCl molecules on the Au(111) surface indicating that the normal component of the E_i is important for crossing the activation barrier for dissociation.
(3) The Cl atom coverage measured by AES was observed to decrease with increasing surface temperature used for dosing (in the range of 130 to 370 K).

These observations will be discussed in terms of the mechanism of the dissociation reaction (role of translation vs the vibration in crossing the activation barrier). Further, the surface temperature dependence of the Cl atom coverage indicates that recombination reactions to form HCl and H_2 occur on the surface.

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Experimental results of hydrogen recombination on metal surface

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In this work, we investigated the recombinative desorption of hydrogen from metal (Cu, Ag, or Au) surfaces 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 goes down as the vibrational excitation increases. We will investigate the dependence of the reaction probability on rotational, vibrational and translational energy of hydrogen molecules. The angular distributions of products would be another intriguing issue to study. We can also study hydrogen recombination on stepped surfaces. This will be an ideal benchmark data-set for testing modern theories of surface chemistry. Specifically, we wish to know if electronically adiabatic theory can 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 will help us to understand to what extent the Born-Oppenheimer approximation can break down in a simple chemical reaction at a metal surface.

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Collision of HCl with Au(111) – Dissociation of HCl?

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In this talk, we will present recent results of the measurement of HCl molecular beams from a hot nozzle colliding with a Au(111) surface. A molecular beam of HCl (4% HCl in H_2) was lead through a resistively heated SiC tube allowing to increase the kinetic energy from initially 1.2 eV up to 2.3 eV. After dosing a surface at room temperature for several minutes with higher beam velocities, Chlorine could be detected by Auger spectroscopy. Preliminary results suggest a threshold behavior allowing dissociation of HCl on Au after achieving kinetic energies of 2.0 eV which is supported by angular dependence measurements. Dosing at lower surface temperatures leads to higher Auger signal intensities of Cl, not vanishising after heating up to room temperature and therefore excluding HCl physisorption. Possible mechanisms for the dissociation of HCl on Au(111) and subsequent desorbing processes will be discussed.

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