O 42 Oberflächenreaktionen II

Zeit: Dienstag 10:45–13:00 Raum: TU EB407

O 42.1 Di 10:45 TU EB407

Chemically induced electronic excitations during scattering and Eley Rideal reaction of hydrogen on gold surfaces — •Beate Mildner[1], Yanka Yeliazova[1], Eckart Hasselbrink[1], and Detlef Diesing[2] — [1]Institute of Physical and Theoretical Chemistry, University of Duisburg-Essen, Campus Essen — [2]Institut für Schichten und Grenzflächen 3, Forschungszentrum Jülich

It is well known that highly exothermic reactions on metal surfaces can cause electron emission, if an electron is ejected with an energy exceeding the metal workfunction. Electrons with low excitation energy can be detected using thin film Schottky or tunnel diodes with barrier energies $E_B < \Phi_{metal}$. At first, we studied the scattering of H-atoms at room temperature. On the gold surface of a $Ta/Ta_2O_5/Au$ tunnel junction we observed a H-atom induced tunnel current at 0 V bias voltage. Exposure to H_2 does not cause any detectable currents. When the sample temperature is lowered to 170 K (≈ desorption temperature of atomic hydrogen) the hydrogen induced tunnel current increases by a factor of 6. Moreover, the shape of the transient current caused by the adsoption of hydrogen from a pulsed atom beam changes from a simple square form to a shape with an initial overshoot followed by a slow decay. We assign the overshoot and the increase of the tunnel current to an Eley Rideal reaction ($H_{gas}+H_{ad} \rightarrow H_2$). Applying a bias voltage to the tunnel junction, we compared the energetic structure of the electronic excitation during ER reaction with the one from a scattering process. The bias dependence reveals that electrons with increased energy are produced during the ER-reaction .

O 42.2 Di 11:00 TU EB407

Reversible isomerization of an individual photochrome molecule by tunneling electrons — •Jörg Henzl[1], Karina Morgenstern[2], Heiko Gawronski[1], Michael Mehlhorn[1], and Karl-Heinz Rieder[1] — [1]Freie Universität Berlin, Institut für Experimentalphysik, Arnimallee 14, 14195 Berlin — [2]Universität Hannover, Institut für Festkörperphysik, Appelstr. 2, 30167 Hannover

Azobenzene molecules are well known for the light-stimulated cis-trans isomerization. We have investigated Disperse Orange 3 on the Au(111) surface at 5K under UHV conditions with a scanning tunneling microscope. Disperse Orange 3 ($C_{12}H_{10}N_4O_2$) is an azobenzene derivate ($C_6H_4N{=}NC_6H_4$) with a NO_2 and a NH_2 endgroup. At a deposition temperature of about 230K the molecules form dimers, which can be separated into monomers. We induce isomerization of individual molecules between the cis- and trans-state for several times both within dimers and monomers by tunneling electrons. Detailed insights in the switching process will be presented in this talk.

O 42.3 Di 11:15 TU EB407

The adsorption of gold on gold surfaces – where does the binding energy go ? — •Michael Kayser[1], Eckart Hasselbrink[1], and Detlef Diesing[2] — [1]Institute of Physical and Theoretical Chemistry, University of Duisburg-Essen, Campus Essen — [2]Institut für Schichten und Grenzflächen 3, Forschungszentrum Jülich

The chemistry and physics of the deposition of a metal atoms on a metal surface is not only interesting for the understanding of fundamental processes like charge transfer, chemical bonding and energy dissipation but also for altering or even tailoring properties and behaviour of surfaces [1] for their industrial application. The deposition of a metal atom on a metal surface leads to the release of the heat of sublimation which is completely transferred to the substrate. The question to be answered is to which extend the binding energy is transferred to the electronic system and to the phononic system of the gold. Electronic excitations within their short lifetime of several tens of fs [2] can be detected using a tunnel junction realized by a metal-insulator-metal system. We present measurements which prove that the binding energy is at least partly transferred to the electronic system of the gold substrate. The adsorption induced electronic excitation is measured as a tunnel current at 0 V bias voltage across the tunnelling barrier. The observed tunnel current is directly proportional to the gold atom flux. Additional processes as photo induced tunnelling play a minor role in the experiment.

[1] M. Ø. Pedersen, S. Helveg, A. Ruban - Surf. Sci. 426 (1999) 395 - 409
[2] S. Meyer et al., Phys. Rev. Lett. 93, 137601 (2004)

O 42.4 Di 11:30 TU EB407

Kinetic electron excitation in atomic collision cascades — •Stefan Meyer[1], Detlef Diesing[2], and Andreas Wucher[1] — [1]Institut für experimentelle Physik, Universität Duisburg- Essen, 45117 Essen — [2]Institut für Schichten und Grenzflächen 3, Forschungszentrum Jülich, 52425 Jülich

In atomic collision cascades, initiated by the impact of a fast particle onto a solid surface, a part of the deposited energy is transferred into the electronic system and leads to the creation of hot electrons. Recently we have shown [1] that Metal- Insulator- Metal (MIM) tunnel junctions are an appropriate tool to investigate low energy electron excitation. Electrons which are excited by particle bombardment in the top electrode of the junction travel elastically within their mean free path to the insulator interface and tunnel to the base electrode where they are detected as a tunnel current. By applying a dc- voltage across the junction, it is possible to compare the energy distribution of excited electrons caused by collision reactions with different kinetic energies. Comparative collision experiments with neutral atoms will distinguish which part of the energy transferred into the substrate is due to potential energy effects.

[1] S. Meyer, D. Diesing, A. Wucher, Physical Review Letters 93 (2004) 137601

O 42.5 Di 11:45 TU EB407

Detection of low energy electronic excitations($E < E_{vacuum}$) during the interaction of metastable rare gas atoms with metal surfaces. — •Domokos Kovacs[1], Johannes Berndt[1], Jörg Winter[1], and Detlef Diesing[2] — [1]Experimentalphysik 2, Ruhr-Universität Bochum — [2]Institut für Schichten und Grenzflächen 3, Forschungszentrum Jülich

The interaction of metastable rare-gas atoms with a metal surface gives rise to deexcitation accompanied by electron emission. While the electron emission into the vacuum is well investigated, the low energy electronic excitations cannot be detected by classical spectroscopic methods. For their detection, a thin film tunnel junction was used. It consists of a silver film (15 nm) exposed to the metastable atom beam and of an aluminum film (30 nm), separated by a very thin (2-3 nm) aluminum-oxide layer. In such a multilayer system, low energy electronic excitations can be detected as a pulsed tunnel current in the base aluminum electrode by chopping the metastable atom beam. A bias induced reversible polarity change in the metastable induced tunnel current was observed. It depends strongly on the intrinsic properties of the metastable atoms (excitation energy), but does not depend on the intensity of the metastable atom beam. In order to explain the experimental data a theoretical model considering tunnel processes between a ground state electron gas and an excited free electron gas is proposed.

O 42.6 Di 12:00 TU EB407

Energy partitioning in the femtosecond-laser induced $D_{ads} + D_{ads} \rightarrow D_2$ desorption reaction from Ru(001) — •S. Wagner[1], M. Rutkowski[2], H. Zacharias[2], C. Frischkorn[1], and M. Wolf[1] — [1]Freie Universtät, Berlin — [2]Westfälische Wilhems-Universität, Münster

The energy transfer to different degrees of freedom during the recombinative desorption of D_2 from a deuterium covered Ru(001) surface induced by femtosecond-laser excitation has been investigated with (1+1') resonance enhanced multi-photon photoionization (REMPI) and time-of-flight (TOF) measurements. Rovibrational population distributions of the reaction product are detected via various $B\ ^1\Sigma_u^+ \leftarrow X\ ^1\Sigma_g^+$ Lyman bands using tunable vacuum ultraviolet (VUV) laser radiation in the excitation step. Rotational quantum states in the vibrational ground state and the first excited state are measured yielding rotational temperatures of $E_{\rm rot}/k_{\rm B} = 800\,{\rm K}$ and 1500 K, respectively, at an absorbed laser fluence F of 85 J/m^2. Also, a vibrational temperature of the desorbing molecules is extracted which amounts to $E_{\rm vib}/k_{\rm B} = 1200\,{\rm K}$. In addition, comprehensive TOF results ($E_{\rm trans}/k_{\rm B} = 2500\,{\rm K}$ at $F = 85$ J/m^2) enable complete energy balancing and underline the non-thermal and anisotropic energy partitioning during reaction. The effects of multi-dimensional friction between substrate and adsorbate layer and peculiarities of the potential energy landscape governing the D_2 recombination are discussed.

O 42.7 Di 12:15 TU EB407

Ellipsometric Confirmation of the Phase Transition in Methanol Oxidation over Copper Surface — •Ling Zhou[1], Sebastian Günther[2], and Ronald Imbihl[1] — [1]Institut für Physikalische Chemie und Elektrochemie, Universität Hannover — [2]Department Chemie, Ludwig-Maximilians-Universität München

The kinetics of methanol oxidation over metallic and oxidized copper surface has been studied in the 10^{-2} mbar range. It was found that the different phases over the copper sample lead to different reactivity. Comparing with the metallic copper surface, the partially oxidized copper surface has higher reactivity to produce formaldehyde whereas the deep oxidized surface shows more total oxidation to CO_2. The phase transition during the reactions can be well defined by the optics and ellipsometer which is in favour of determining the active phase in methanol selective oxidation to formaldehyde.

O 42.8 Di 12:30 TU EB407

A RAIRS investigation of methanol dehydrogenation on a Rh(111)/V islands surface — •Robert Schennach, Hans Peter Koch, and Georg Krenn — Institute of Solid State Physics, Graz University of Technology, Petersgasse 16/2, 8010 Graz, Austria

Fundamental insights in the dehydrogenation reaction of methanol is of interest due to the importance of this reaction in methanol powered fuel cells. In this work the methanol dehydrogenation on a Rh(111) surface with V islands has been investigated using reflection absorption infra red spectroscopy (RAIRS). The surface consists of two to four layer high V islands with an average island diameter of about 9 nm, which cover about 25% of the Rh(111) surface. The reactivity of this surface for methanol dehydrogenation is about 6 times as high as the reactivity of the V free Rh(111) surface. But the reactivity of the Rh(111)/V islands surface decreases over time due to CO dissociation on the V islands. However, the deactivated islands are still significantly more reactive towards methanol dehydrogenation than Rh(111). The RAIRS measurements show that the reaction mechanism changes markedly going from Rh(111) to the Rh(111)/V islands surface. This is mainly due to an increased stability of methanol on the V islands. In addition a new reaction pathway via methanol C-O bond opening yielding CH_3 on the surface has been found on the Rh(111)/V islands surface. The reactivity of the deactivated islands seems to be due to a slight stabilization of methanol on this surface.

O 42.9 Di 12:45 TU EB407

Elektronen induzierte Isomerisierung: Direkte Visualisierung einer chemischer Reaktion innerhalb eines einzelnen Moleküls — •V. Simic-Milosevic[1], K. Morgenstern[2], M. Mehlhorn[1] und K.-H. Rieder[1] — [1]Freie Universität Berlin, Institut für Experimentalphysik, Arnimallee 14, 14195 Berlin — [2]Universität Hannover, Institut für Festkörperphysik, Appelstr. 2, 30167 Hannover

Der Tunnelstrom eines Tieftemperatur-Rastertunnelmikroskops wurde genutzt, um Dipolmoment und 2D- Chiralität von einzelnen Chlornitrobenzol ($C_6H_4ClNO_2$)-Molekülen auf Cu(111) bei 5 K zu verändern. Die hohe submolekulare Auflösung des RTM erlaubt die Identifizierung des Phenylrings und der verschiedenen Substituenten. Durch direkte Injektion von Tunnelelektronen in die Bindung zwischen Kohlenstoff und Chlor ist es möglich, die Position des Chlors innerhalb des Moleküls zu ändern. Als Folge ändern sich 2D-Chiralität und Dipolmoment des Chlornitrobenzols. Die Abhängigkeit der Reaktionswahrscheinlichkeit von der Anregungsenergie und dem Strom wurde untersucht. Dies erlaubt die Identifizierung des zugrunde liegenden Mechanismus. Detailierte Ergebnisse werden im Ramen des Vortrags präsentiert.