

## O 46: Time-resolved spectroscopies I

Time: Thursday 10:30–13:00

Location: SCH A315

O 46.1 Thu 10:30 SCH A315

**Heat transport beyond thermodynamic equilibrium** — ●MARTIN KAMMLER, ANJA HANISCH-BLICHARSKI, BORIS KRENZER, SIMONE MÖLLENBECK, PAUL SCHNEIDER, and MICHAEL HORN-VON HOEGEN — Department of Physics, Universität Duisburg-Essen, 47048 Duisburg, Germany

The heat transport of a thermal excited film into the substrate is determined by the thermal boundary resistance at the interface and well described by the Acoustic Mismatch Model (AMM) and Diffuse Mismatch Model (DMM), both well-accepted bulk models. For ultra thin films we observe a deviation for the AMM model.

After excitation of an ultra thin epitaxial Bi(111) film with fs-laser pulses the heat transport into the silicon substrate is determined by studying the cooling process with ultrafast time resolved electron diffraction in RHEED geometry using the Debye Waller effect. For Bi on Si(111) the observed exponential temperature decay time is well described by the AMM and DMM up to only 2.5 nm thin films but not for Bi/Si(001). Here, we observe a slight saturation of the cooling process with a decay constant  $\tau = 500$  ps on films thinner than 6 nm; the heat is trapped!

O 46.2 Thu 10:45 SCH A315

**Freezing Hot Electrons in Crystalline NH<sub>3</sub> and D<sub>2</sub>O on Metal Surfaces** — ●JULIA STÄHLER<sup>1,2</sup>, MICHAEL MEYER<sup>1</sup>, UWE BOVENSIEPEN<sup>1</sup>, CORNELIUS GAHL<sup>3</sup>, and MARTIN WOLF<sup>1,4</sup> — <sup>1</sup>Freie Universität Berlin, Fachbereich Physik, Arnimallee 14, 14195 Berlin — <sup>2</sup>University of Oxford, Department of Physics, Clarendon Laboratory, Parks Road, Oxford OX1 3PU, United Kingdom — <sup>3</sup>Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, Max-Born-Str. 2A, 12489 Berlin — <sup>4</sup>Fritz-Haber-Institut der MPG, Abteilung Physikalische Chemie, Faradayweg 4-6, 14195 Berlin

Electron transfer across molecule-metal interfaces is crucial in technologically highly relevant fields as, for instance, the design of nanoscale electronic devices and solar cells. Knowledge of the impact of the molecular structure and morphology on the interfacial electron dynamics is hereby essential for the development of efficient and reliable appliances. We use time-resolved two-photon photoelectron (2PPE) spectroscopy to study the electron transfer and solvation dynamics at the NH<sub>3</sub>/Cu(111) and the D<sub>2</sub>O/Ru(001) interface. We show that (i) the electron dynamics are significantly influenced by crystallization of the adlayer, (ii) the structural reorganization results in electron lifetimes on the order of minutes, and (iii) the microscopic and structural properties of the solvent determine the energetic relaxation even on macroscopic timescales. In addition, we present the first direct observation of electron trapping in pre-existing traps and demonstrate that the remarkably long lifetimes cannot be attributed to a purely morphological transition.

O 46.3 Thu 11:00 SCH A315

**Vibrational response of D<sub>2</sub>O molecules on Ru(001) upon electron injection observed with time-resolved sum-frequency generation** — ●CHRISTIAN FRISCHKORN, JURAJ BDZOCH, JAN ZACHARIAS, and MARTIN WOLF — Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, 14195 Berlin

Electrons as excess charges in polar solvents result in molecular motion within the solvent to stabilize the new charge distribution known as electron solvation. We employ time-resolved broadband-IR-VIS SFG (sum frequency generation) spectroscopy as a powerful technique to study the changes in the vibrational spectra of the D<sub>2</sub>O water layers upon electron injection from a Ru(001) surface. Unlike in time-resolved photoemission experiments where the electron binding energy is traced and the solvent response is inferred subsequently, SFG vibrational spectroscopy *directly* monitors the molecular reorientation induced by the excess charge. If the Ru surface is excited with 266-nm (4.65 eV) photons, excess electrons injected into the adsorbate layer induce – dependent on layer morphology and layer thickness – drastic changes in the vibrational spectra of the OD stretch vibration. While in amorphous ice layers only weak changes are observed, a SFG signal increase by several orders of magnitude (enhancement factor  $\sim 10^3 - 10^4$ ) is found in crystalline D<sub>2</sub>O layers. The physical and chemical processes involved will be discussed.

O 46.4 Thu 11:15 SCH A315

**Lifetime reduction of surface states caused by impurity scattering** — ●SWANTJE HEERS, PHIVOS MAVROPOULOS, SAMIR LOUNIS, and STEFAN BLÜGEL — Institut für Festkörperforschung and Institute for Advanced Simulation, Forschungszentrum Jülich, 52425 Jülich, Germany

In order to describe the dynamics of charge transport, localization and quantum information on metal surfaces it is of crucial importance to understand the temporal evolution of quasiparticles (electrons and holes) in surface states, characterized by a lifetime  $\tau$ . This lifetime is strongly reduced by scattering at impurities, vacancies and adatoms. Using the Korringa-Kohn-Rostoker Green function method [1], the elastic scattering rate  $1/\tau$  can be calculated for adatoms as well as impurities and vacancies in and below the surface. We have performed calculations for the Cu(111) surface of finite-thickness slabs, investigating the lifetime of the Cu(111) surface state for the whole series of 3d transition metals as impurities. The thickness of the slab is varied, ranging from six to forty atomic layers. Our formalism allows us to separate the lifetime reduction induced by scattering to bulk states from scattering to surface states. We show that trends through the 3d series are qualitatively different for adatoms, compared to impurities in the surface. This work was supported by the ESF EUROCORES Programme SONS under contract N. ERAS-CT-2003-980409 and the Priority Programme SPP1153 of the DFG grant Bl 444/8-1.

[1] N. Papanikolaou, R. Zeller, P. H. Dederichs *J. Phys.: Condens. Matter* **14**, 2799 (2002).

O 46.5 Thu 11:30 SCH A315

**Mechanical Oscillations of Nanostructures** — ●RICHARD TAUBERT<sup>1</sup>, FLORIAN HUDERT<sup>2</sup>, ALBRECHT BARTELS<sup>2,3</sup>, and THOMAS DEKORSY<sup>2</sup> — <sup>1</sup>Universität Stuttgart — <sup>2</sup>Universität Konstanz — <sup>3</sup>Gigaoptics GmbH, Konstanz, Deutschland

Understanding the mechanical properties of nanostructures has been a field of large interest during the last years. We present a method to investigate the phonon dynamics in nanostructures by time-resolved optical measurements. A new technique, Asynchronous Optical Sampling (ASOPS) is used to carry out pump-probe spectroscopy. Instead of using one laser and a mechanical delay line to generate pump- and probe-beams, two separated lasers, whose repetition rates are slightly detuned by several kHz are used. This allows the measurement of mechanical oscillations with GHz line width and a signal-to-noise ratio of  $10^7$  within 100 seconds of measurement time.

Time-resolved measurements on metallic and semiconductor superlattices as well as metallic nano-triangles and nano-tetrahedrons are presented. The measurements on the superlattices clearly resolve the first-order back-folded longitudinal acoustic phonons at the zone center. Furthermore, it is possible to do precise layer thickness and superlattice-period measurements. The mechanical oscillations of metallic triangles and pyramids with different edge lengths are observed and the influence of geometry, size and substrate are investigated.

O 46.6 Thu 11:45 SCH A315

**Fano line profiles in two-photon photoemission of image-potential states on Si(100)** — ●CHRISTIAN EICKHOFF<sup>1,2</sup>, JENS KOPPRASCH<sup>1,2</sup>, CORNELIUS GAHL<sup>1</sup>, and MARTIN WEINELT<sup>1,2</sup> — <sup>1</sup>Max-Born-Institut, Max-Born-Straße 2a, 12489 Berlin, Germany — <sup>2</sup>Freie Universität Berlin, Fachbereich Physik, Arnimallee 14, 14195 Berlin, Germany

We have studied the surface states on Si(100) by time-, energy- and angle-resolved two-photon photoemission spectroscopy. Tuning the photon energy of the pump pulse between 4.5 and 5.4 eV allows us to populate unoccupied states up to the Si(100) vacuum level. Besides the occupied dangling-bond state  $D_{up}$ , we have identified the Rydberg-like series of image-potential states. Lifetimes and binding-energies of  $n=1, 2$  and  $3$  will be presented. Significant variation of both the 2PPE peak positions and intensities were found when tuning the photon energy across the  $D_{up}$  to  $n=1$  and  $D_{up}$  to  $n=2$  transitions. Below resonance we observe the  $D_{up}$  initial state with the kinetic energy following the pump pulse photon energy. Above resonance the  $D_{up}$  intensity is significantly reduced and the peak position reflects the respective image-potential-state resonances. Around the

resonance the lowering in energy of the expected peak position and the intensity variations can be explained by a Fano-like resonance, caused by interference between transitions from  $D_{up}$  to the discrete image-potential-state resonances and the bulk continuum in combination with the finite bandwidth of the laser pulses.

O 46.7 Thu 12:00 SCH A315

**Two-photon photoemission spectroscopy of image potential states on Ag(001): Momentum-dependent lifetimes and quantum beats** — ●MARIO KIEL, KLAUS DUNCKER, and WOLF WIDRA — Martin-Luther Universität Halle-Wittenberg, Halle, Germany

Using a novel all-fiber based oscillator/amplifier system operating at a repetition rate of 1.5 MHz which pumps two noncollinear optical parametric amplifiers with tunable pulses (<30 fs) in the range of 500-950 nm, the  $n=1, 2$  and  $3$  image potential states on Ag(001) have been investigated by time and momentum-resolved two-color two-photon photoemission spectroscopy (2PPE). Lifetimes of 55, 165 and 380 fs at  $k_{||}=0$  have been determined for these states, respectively, similar to previous work [1]. The lifetime of the  $n=1$  state reveals a quadratic momentum-dependence of the decay rate. This leads to a shortening to 26 fs at  $k_{||}=0.4 \text{ \AA}^{-1}$  which can be described by a linear 34 meV/eV decay rate increase with excess kinetic energy which is comparable to results on Cu(001) [2]. It clarifies the controversy regarding the existence of additional decay channels on Ag(001) vs. Cu(001) derived from intrinsic linewidth measurements [3].

Quantum beat spectroscopy is used to determine the energy difference between the  $n=3$  and  $4$  states to 35 meV based on the 2PPE signal oscillation with a periodicity of 118 fs.

[1] I. L. Shumay, *et al.*; PRB **58** (1998), 13974

[2] W. Berthold, *et al.*; PRL **88** (2002), 056805

[3] G. Ferrini, *et al.*; PRB **67** (2003), 235407

O 46.8 Thu 12:15 SCH A315

**Chirped-pulse two-photon photoemission from Cu(111): experiment and theory** — ●ALEXANDER FISCHER<sup>2</sup>, FELIX STEEB<sup>2</sup>, STEFAN MATHIAS<sup>2</sup>, MARTIN WIESENMEYER<sup>1</sup>, MARTIN AESCHLIMANN<sup>2</sup>, and MICHAEL BAUER<sup>1</sup> — <sup>1</sup>IEAP, Sektion Physik, Christian-Albrechts-Universität zu Kiel, 24908 Kiel — <sup>2</sup>Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, 67663

In the study of ultrafast processes at surfaces using femtosecond light sources the effect of the spectral phase of the excitation pulse has been considered only in a few publications, yet [1,2]. For instance, Petek *et al.* investigated the spectral changes in the 2PPE signal from the Cu(111) Shockley surface state as induced by a controlled variation of the spectral phase of the excitation light. In this case the findings were attributed to the specific ultrafast dynamics of the involved intermediate state and not to the chirp of the laser pulse. Here we report on an extension of these earlier studies, which considers next to the peak position further spectral signatures such as the peak FWHM and asymmetry. Additionally we develop a theoretical model, which enable us to reproduce and interpret our observations in a quantitatively very satisfactory manner. It reproduces almost perfectly the experimental observed changes in the surface state 2PPE signal for a non resonant excitation scheme under consideration of the complete spectral phase

of the laser pulse, especially the interaction of GVD and TOD[3].

[1] H. Petek, *et al.*, Phys. Rev. Lett. **79** 4649 (1997) [2] M. Merschdorf, *et al.*, Phys. Rev. B **70**, 1934401 (2004) [3] M. Bauer, *et al.*, The nature of a nonlinear excitation..., New J. of Phys., in print

O 46.9 Thu 12:30 SCH A315

**Observation of Surface Plasmon Propagation with Attosecond Accuracy** — ●NIEMMA MOHAMADIE BUCKANIE<sup>1</sup>, NICOLAI RASS<sup>1</sup>, and FRANK-JOACHIM MEYER ZU HERINGDORF<sup>1,2</sup> — <sup>1</sup>Universität Duisburg-Essen, Institut für Experimentelle Physik, Lotharstraße 1, 47057 Duisburg — <sup>2</sup>Center for Nano Integration Duisburg-Essen (CeNIDE), 47057 Duisburg

Surface Plasmon Polaritons (SPP) are collective oscillations of the electron gas in metallic systems that propagate along nano-particle surfaces. SPPs can be imaged by Photoemission Electron Microscopy (PEEM), using ultra short femtosecond laser pulses [1]. For the observation of the SPP wave propagation, time resolved techniques, i.e., pump-probe techniques must be applied, where delay times of attosecond accuracy between the pump and the probe pulse are required. The necessary interferometer to generate the time delay must then be stable in the attosecond time regime for the duration of the experiments, i.e., over several hours. To achieve these requirements, we designed and built an actively stabilized Mach-Zehnder-Interferometer (MZI) that has the ability to control time delays between the pump and the probe pulse in the attosecond time regime and that has a long-time stability of 15 as. With this MZI the SPP propagation in Ag islands on Si(111) is directly observed.

[1] L.I. Chelaru and F.-J. Meyer zu Heringdorf, *Surf. Sci.* **601** (2007) 4541–4545

O 46.10 Thu 12:45 SCH A315

**Correlated positron-electron emission from surfaces** — ●FRANK O. SCHUMANN, GRANT VAN RIESSEN, MICHAEL BIRKE, CARSTEN WINKLER, and JÜRGEN KIRSCHNER — Max-Planck Institut für Mikrostrukturphysik, Weinberg 2, 06120 Halle

Electrons in a solid do not move independent from each other. The Pauli principle demands that two electrons with parallel spins can not be at the same location. The Coulomb interaction makes it energetically unfavorable for electrons to be close to each other. This constitutes the concept of the exchange-correlation (xc) hole. Our electron pair emission studies demonstrate that this concept is an experimental reality. In these experiments a primary electron or photon hits a surface and emits an electron pair. In order to disentangle the different contributions to the xc-hole, one should use as primary particle a positron and study the positron-electron pair emission. In this case one does not need to consider the Pauli principle. Because of this motivation we performed the first experiment, which demonstrates that the impact of a positron causes indeed the emission of a positron-electron pair. We used the NEPOMUC facility together with a pair of hemispherical analyzers operated with a coincidence circuit. As target we selected a LiF(100) surface, which was hit with 85 eV primary positrons. We find that the emission of positron-electron pairs is time correlated.