

## O 52: Time-Resolved Spectroscopy III

Time: Wednesday 15:15–18:30

Location: MA 042

O 52.1 Wed 15:15 MA 042

**Interferometric control of spin-polarized photoemission from Cu(001)** — ●AIMO WINKELMANN, WEN-CHIN LIN, and JÜRGEN KIRSCHNER — Max-Planck-Institut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle(Saale), Germany

We demonstrate that we can tune the spin-polarization of photoelectrons emitted in a three-photon process from Cu(001) by interferometric control of the delay between two ultrashort optical pulses in a pump-probe experiment. As a function of pulse delay, the spin polarization can be changed from  $\pm 20\%$  to  $\mp 40\%$  using circularly polarized light for excitation. We differentiate between the regime of optical interference for overlapping pulses and, for longer delays, the influence of the material response. The influence of the coherent material response is detected by observing interference oscillations at twice the optical frequency. These are created by the interference of the pump pulse induced electromagnetic polarization at the surface and the subsequent probe pulse. The spin polarization of the observed electrons for delays longer than the pulse length also shows the oscillations at twice the optical frequency.

O 52.2 Wed 15:30 MA 042

**Spin-dependent inter- and intraband scattering in image-potential-state bands on 3d ferromagnets** — ●ANKE B. SCHMIDT<sup>1</sup>, MARTIN PICKEL<sup>2</sup>, MARKUS DONATH<sup>1</sup>, and MARTIN WEINELT<sup>2</sup> — <sup>1</sup>Physikalisches Institut, Universität Münster, Wilhelm-Klemm-Straße 10, 48149 Münster, Germany — <sup>2</sup>Max-Born-Institut, Max-Born-Straße 2A, 12489 Berlin and Freie Universität Berlin, Fachbereich Physik, Arnimallee 14, 14195 Berlin, Germany

We have observed ultrafast electron-magnon scattering of image-potential-state electrons on ultrathin ferromagnetic films in three different regions of energy and momentum transfer directly in the time domain. In our spin- and time-dependent photoemission experiment we employ the dispersing image-potential-state electron as observer or primary electron, thus effectively separating low-energy losses from direct decay into d-holes. For the quasielastic processes resonant interband scattering and dephasing at the band minimum as well as intraband scattering, an energy relaxation process, we found evidence to suggest that electron-magnon scattering constitutes a significant scattering channel for excited minority-spin electrons on iron even on the femtosecond timescale.

O 52.3 Wed 15:45 MA 042

**Coherent dynamics at Gd(0001) and Tb(0001) surfaces: magnon-enhanced damping of optical phonons** — ●ALEXEY MELNIKOV<sup>1</sup>, ALEXEY POVOLOTSKIY<sup>2</sup>, and UWE BOVENSIEPEN<sup>1</sup> — <sup>1</sup>Freie Universität Berlin, Institut für Experimentalphysik, Arnimallee 14, D-14195 Berlin, Germany — <sup>2</sup>St. Petersburg State University, Laser Research Institute, St. Petersburg, 198504 Russia

Coherent lattice and spin dynamics were studied at the Gd(0001) surface by time-resolved second harmonic generation [1]. To understand the elementary processes responsible for optical excitation and damping of coherent phonons (CP) and magnons (CM), we perform temperature-dependent studies in Gd and Tb. With lowering the temperature  $T$  from the Curie point  $T_C$  to 40K amplitudes of CP and CM increase by more than an order following the increase of spin ordering and showing an essentially "magnetic" nature of the excitation. The CP damping rate  $\Gamma$  increases linearly with the temperature in transition metals, which is attributed to anharmonic phonon-phonon scattering [2]. In Gd we observe a monotonous increase of  $\Gamma(T)$  at  $T < T_C$  but near  $T_C$  it reduces significantly. We explain this by phonon-magnon scattering contributing to  $\Gamma$  at  $T < T_C$ . This contribution scales with the magnetoelastic constant mediated by the spin-orbit interaction. This conclusion is confirmed by  $\Gamma(T)$  measured in Tb ( $L=3$ ), where the spin-lattice coupling is much larger than in Gd ( $L=0$ ): the phonon-magnon contribution to  $\Gamma(T)$  is found to be 6 times larger than in Gd.

[1] A. Melnikov et al., Phys. Rev. Lett. **91**, 277403 (2003).[2] M. Hase et al., Phys. Rev. B **71**, 184301 (2005).

O 52.4 Wed 16:00 MA 042

**Observing photo-induced chemical reactions of molecules on surfaces in real-time** — ●MIHAI E. VAIDA, PETER E. HINDELANG,

ROBERT TCHITNGA, and THORSTEN M. BERNHARDT — Institut für Oberflächenchemie und Katalyse, Universität Ulm, Albert-Einstein-Allee 47, D-89069 Ulm, Germany

Time-of-flight mass spectrometry in conjunction with femtosecond resonance-enhanced multiphoton ionization is used to monitor ultrafast chemical reactions of supported molecules. A new experimental approach enables the quick and precise in situ preparation of the surface with sub-monolayer adsorbate coverage prior to photo-induced reaction. The investigation of different reaction pathways is accomplished by direct real-time monitoring of the different reaction products and intermediates as well as their kinetic energy content. First results for methyl iodide adsorbed on ultra-thin insulating magnesia films will be presented.

O 52.5 Wed 16:15 MA 042

**Reversible switching of azobenzene in direct contact with a Au(111) surface** — ●SEBASTIAN HAGEN, PETER KATE, FELIX LEYSSNER, MARTIN WOLF, and PETRA TEGEDER — Freie Universität Berlin, Fachbereich Physik

Two-photon photoemission spectroscopy is employed to analyze reversible changes in the electronic structure of the molecular switch tetra-*tert*-butyl-azobenzene (TBA) adsorbed on Au(111), which are induced by light and thermal activation. Cycles of illumination and annealing steps confirm the reversibility of the switching process, which we assign to a *trans/cis*-isomerization of TBA molecules in direct contact with the Au(111) surface. Based on wavelength dependent measurements we propose that the optically-induced conformational change (*trans*- to *cis*-isomerization) of the adsorbed molecules is not caused by direct intramolecular excitation as in the liquid phase but rather by an indirect mechanism, *viz.* a substrate mediated process. Thereby photoexcitation of holes in the Au *d*-band followed by a charge transfer to the HOMO level drives the isomerization in the photon energy range between 2.1 and 4.6 eV. In addition, for photon energies above 4.6 eV the creation of a negative ion resonance *via* attachment of excited electrons to the molecules plays a role.

O 52.6 Wed 16:30 MA 042

**Electron dynamics at the PTCDA/Ag(111) interface studied with 2PPE** — ●MANUEL MARKS<sup>1</sup>, CHRISTIAN SCHWALB<sup>1</sup>, SÖNKE SACHS<sup>2</sup>, ACHIM SCHÖLL<sup>2</sup>, EBERHARD UMBACH<sup>2,3</sup>, and ULRICH HÖFER<sup>1</sup> — <sup>1</sup>Fachbereich Physik und Zentrum für Materialwissenschaften, Philipps-Universität Marburg, D-35032 Marburg — <sup>2</sup>Universität Würzburg, Experimentelle Physik II, D-97074 Würzburg — <sup>3</sup>Forschungszentrum Karlsruhe, D-76021 Karlsruhe

We investigated epitaxially grown PTCDA (3,4,9,10-perylene-tetracarboxylic acid-dianhydride) on the Ag(111) surface as model system for a metal-organic interface by means of time- and angle-resolved two-photon photoemission (2PPE). In the presence of thin PTCDA films, an unoccupied state with an effective electron mass of  $0.39 m_e$  is observed in the projected band gap of Ag 0.6 eV above  $E_F$ . Its inelastic electronic lifetime is  $\approx 50$  fs and the state has an appreciable metallic character, significantly exceeding that of the image-potential states. We assign the new state to a mixture of the former Ag(111) Shockley surface state and the LUMO+1 of the first PTCDA monolayer (ML). In contrast to this interface state, which changes only weakly with PTCDA coverage, the binding energy of the first image-potential state shows a strong dependence. It increases by 135 meV for 1 ML, compared to clean Ag(111), but with absorption of the second ML, a subsequent drop of  $-70$  meV relative to the clean surface occurs. A similar coverage dependence can be seen in the effective electron mass, which decreases by 20% from the first to the second PTCDA layer.

O 52.7 Wed 16:45 MA 042

**Strong temperature dependence of vibrational relaxation of H/Ge(100)** — XU HAN, KRISTIAN LASS, and ●ECKART HASSELBRINK — Department of Chemistry, University of Duisburg-Essen, D-45117 Essen, Germany

The population relaxation of vibrational excitations of adsorbates is the result of various dynamical couplings. However, the underlying microscopic mechanisms at semiconductor surfaces are not yet fully understood. Germanium provides special research interests because of

its lower surface Debye temperature than silicon. On Ge(100) surface the Debye temperature is more than two times smaller than in bulk, which implies a strong anharmonicity on the surface.

IR pump-SFG probe measurements of the vibrational population relaxation on H ( $2 \times 1$ )/Ge(100) surfaces revealed a strong temperature dependence of the vibrational lifetime.  $T_1$  was measured to be 0.5 ns for the Ge-H symmetric stretch at RT. The relaxation of vibrational energy follows a single exponential decay. The order of measured decay constants decreases from one nanosecond to 100 picoseconds, with increasing the substrate temperature from 273 K to 400 K. The model fitting of the temperature dependence suggests that relaxation of the Ge-H stretch mode initiates simultaneous excitation of three Ge-H bending quanta and four bulk phonons above RT. Our DFT calculations suggest that the vibration-phonon coupling, via anharmonicity, dominates the vibrational decay processes. The strong temperature dependence is facilitated by the quick increase of the density of states associated with low-frequency phonons.

O 52.8 Wed 17:00 MA 042

**Time-resolved investigation of laser-induced diffusion of CO on terraces of vicinal Pt(111)** — ●JENS GÜDDE, MARCO LAWRENZ, and ULRICH HÖFER — Fachbereich Physik und Zentrum für Materialwissenschaften, Philipps-Universität Marburg, D-35032 Marburg

We report on a time-domain study of diffusion of CO on a vicinal Pt(111) surface at low substrate temperature induced electronically via substrate electrons excited by femtosecond laser pulses. Diffusion is observed by monitoring the occupation of step sites on vicinal substrates by nonlinear optical second harmonic generation (SHG). Two-pulse correlation experiments were done to study the dynamics of energy flow from the initially excited substrate electrons to the adsorbate degrees of freedom. In contrast to O/Pt(111) [1], CO has a substantially higher binding energy at the step sites than at the terrace sites. For this reason the steps can be employed as traps for adsorbed CO at low temperatures and laser-induced diffusion can be observed by SHG not only from the steps to the terraces but also from terrace to step sites. For terrace diffusion of CO with a barrier of only 0.2 eV our correlation measurements at substrate temperatures of 40 and 60 K indicate a transition from an electronically driven process at low temperatures to a phonon driven process at higher substrate temperatures.

[1] K. Stépán *et al.*, Phys. Rev. Lett. **94**, 236103 (2005).

O 52.9 Wed 17:15 MA 042

**Image-potential states and resonances on the clean and Ar-gon covered (111)-surfaces of Copper and Silver** — ●ANDREAS DAMM, KAI SCHUBERT, JENS GÜDDE, and ULRICH HÖFER — Fachbereich Physik und Zentrum für Materialwissenschaften, Philipps-Universität, D-35032 Marburg, Germany

We report on a comparative 2PPE study of the influence of Ar adlayers on the properties of image-potential states of Ag(111) and Cu(111). On both surfaces only the  $n = 1$  state is non resonant with bulk states. As a result of Ar adsorption a drop of the workfunction and a systematic lowering of the binding energies of the image-potential electrons is observed. On Cu(111) however the first image state is bound close to the conduction band minimum. Thus this state becomes a resonance for Ar coverages  $\Theta \geq 2$  ML, whereas it remains in the band gap of the bulk states on Ag(111) for coverages up to  $\Theta = 4$  ML. This leads to a strikingly different dependence of the first image state's lifetime on layer thickness for the two surfaces. On Ag(111) the lifetime of the first image state depends exponentially on the number of adsorbed Ar layers, and rises from 32 fs on the clean surface to about 6 ps for Ar coverages of 4 ML. On Cu(111) the initial lifetime increase with layer thickness is essentially the same, but at an Ar coverage of  $\Theta = 2$  ML a kink occurs in correlation with that state becoming resonant with the conduction band of bulk Copper. For higher coverages the increase of lifetime shows again an exponential behavior, but surprisingly with a reduced thickness dependence compared to the non resonant case.

O 52.10 Wed 17:30 MA 042

**Towards time resolved core level photoelectron spectroscopy with femtosecond X-ray free-electron lasers** — ●ANNETTE PIETZSCH, ALEXANDER FÖHLISCH, MARTIN BEYE, MARTIN DEPPE, FRANZ HENNIES, MITSURU NAGASONO, and WILFRIED WURTH — Institut für Experimentalphysik, Universität Hamburg, Luruper Chaussee 149, D-22761 Hamburg

We have conducted an investigation of space-charge induced peak shift and peak broadening in W 4f core-level photoemission from a W(110)

single crystal surface using femtosecond X-ray pulses from the Free-Electron Laser at Hamburg (FLASH). We have established experimentally and through theoretical modelling what should be generally applicable conditions for photoelectron spectroscopy on solids and surfaces using femtosecond X-ray pulses from free-electron laser facilities. As a next step, we have studied within this working range time-resolved core-level photoelectron spectroscopy on a surface combining a femtosecond optical laser synchronized to the femtosecond X-ray pulses. Here, side bands to the W 4f core level lines give evidence of the cross-correlation between the femtosecond x-ray and optical pulses needed for future studies of femtosecond time-resolved core-level photoelectron spectroscopy on solids and surfaces. This work is supported in the framework of the BMBF Forschungsschwerpunkt 301 FLASH: Matter in the light of ultrashort and extremely intense x-ray pulses.

A. Pietzsch *et al.*, submitted

O 52.11 Wed 17:45 MA 042

**Zeit- und ortsaufgelöste Nahfelduntersuchung an strukturierten metallischen Oberflächen** — ●ANDREAS BITZER und MARKUS WALTHER — Molecular and Optical Physics, Institute of Physics, Albert-Ludwigs-Universität Freiburg, Hermann-Herder-Strasse 3, 79104-Freiburg, Germany

Wir präsentieren eine eigens entwickelte Methode zur zeit- und ortsaufgelösten Nahfelduntersuchung an mikrostrukturierten metallischen Oberflächen. Durch gepulste Terahertz-Strahlung werden Oberflächenladungen zu elektromagnetischen Schwingungen angeregt beispielsweise an einem Metallgitter, einem Metalldraht und an einer mikrostrukturierten Metallfolie. Anhand unserem Verfahren ist es nun möglich, direkt an der Oberfläche der Probe das Nahfeld des elektrischen Feldes sowohl zeit- als auch ortsaufgelöst zu bestimmen. Mittels einer Fourieranalyse lässt sich dabei die spektrale Feldverteilung räumlich auflösen. So konnten wir an metallischen Filmen mit periodischen subwellenlängen-großen Lochgittern, welche im THz-Frequenzbereich als Bandpassfilter eingesetzt werden, charakteristische geometrische Moden beobachten. Ein weiteres Gebiet das sich mit Hilfe unserer Methode eindrucksvoll beobachten lässt ist die räumliche und zeitliche Feldentwicklung während der Feldpropagation entlang eines Wellenleiters. Am Beispiel eines Metalldrahtes werden wir den Einkopplungsvorgang eines THz Pulses in einen solchen Wellenleiter visualisieren und anschließend demonstrieren wie sich der THz-Puls entlang des Drahtes fortbewegt.

O 52.12 Wed 18:00 MA 042

**Surface States and Kramers-Kronig relations in one-dimensional Photonic Crystals** — ●MICHAEL BERGMAYER and KURT HINGLER — CD-Labor für oberflächenoptische Methoden, Institut für Halbleiter- und Festkörperphysik, Universität Linz, Austria

Surface states provide very interesting features such as large field enhancement and are very sensitive to the geometry and dielectric behaviour of the investigated structure. A thin metallic sheet allows to investigate the near field and permits a design of a system with negative refracting behaviour. A one-dimensional photonic crystal consisting of layers which have a resonant dielectric behaviour in the infrared (photon-phonon coupling) show very interesting surface states: due to the coupling of bulk and surface states around the resonant frequency a dispersion with negative group velocity occurs. In this region the damping remains small yielding a large figure of merit  $n'/n''$  which is the ratio of real and imaginary part of the dielectric function. In our work we will calculate the dispersion of such coupled surface states and unveil the mechanism that leads to this small damping values. Furthermore we investigate whether and how Kramers-Kronig relations can be applied to systems where the internal structure is on the order of the wavelength.

O 52.13 Wed 18:15 MA 042

**Enhanced photodesorption by vibrational pre-excitation: Quantum model simulations for Cs/Cu(111)** — ●DOMINIK KRÖNER — Universität Potsdam, Institut für Chemie, Theoretische Chemie, Karl-Liebknecht-Str. 24-25, D-14476 Potsdam, Germany

We present electronic and nuclear quantum model simulations for the direct, laser-induced charge transfer from a Cu(111) surface state, to an unoccupied resonance state of an adsorbed Cs-atom. An one-electron model potential for Cu(111) by Chulkov *et al.* [1] is extended by a Cs adsorbate potential to determine energies and lifetimes of electronic states of Cs/Cu(111) at low coverage. In addition, empirical nuclear potential energy surfaces of the electronic ground and the antibonding excited state along the Cs-Cu distance are constructed. For both mod-

els, electronic and nuclear, we perform open-system quantum dynamics with the goals: (i) to estimate the excitation (charge transfer) and Cs desorption probabilities [2], (ii) to optimize the charge transfer process by laser pulse shaping using optimal control theory [2], and (iii) to increase the negligible desorption yield of Cs from the Cu surface, obtained from direct UV excitation, by vibrationally pre-exciting the Cs-Cu bond using a THz-laser pulse [3].

[1] E. V. Chulkov, V. M. Silkin, P. M. Echenique, *Surf. Sci.* **437**, 330 (1999).

[2] D. Kröner, T. Klamroth, M. Nest, P. Saalfrank, *Appl. Phys. A* **88**, 535 (2007).

[3] D. Kröner, S. Klinkusch, T. Klamroth, *Surf. Sci.*, accepted (2007).