O 60: Time-Resolved Spectroscopies

Time: Thursday 15:45–18:30 Location: H42

O 60.1 Thu 15:45 H42

2PPE measurements of PTCDA on Ag(111) — •Christian Schwalb[1], Sönke Sachs[2], Achim Schöll[2], Eberhard Umbach[2], and Ulrich Höfer[1] — [1]Fachbereich Physik und Zentrum für Materialwissenschaften, Philipps-Universität Marburg, D-35032 Marburg, Germany — [2]Universität Würzburg, Experimentelle Physik II, D-97074 Würzburg, Germany

Electron transfer at a metal-molecule interface plays an important role in many chemical disciplines, ranging from molecular electronics to surface photochemistry. We present measurements using time-resolved two-photon photoemission (2PPE) to probe the energetics and dynamics of electronically excited states in epitaxial 3,4,9,10-perylenetetracarboxylic acid-dianhydride (PTCDA) thin films on a Ag(111) surface for a thicknessrange from one to ten monolayers. These measurements allow the identification and determination of the energetic positions for the lowest unoccupied molecular orbital (LUMO), and LUMO + 1. Angle-resolved 2PPE-measurements show a weak dispersion for the LUMO of ˜150 meV. Our experiments indicate that the excitation process for the LUMO is mainly done by electrons from the Ag(111) substrate. Time-resolved measurements probing the dynamics of the system show an increasing of lifetime for the LUMO for increasing layer thickness from 40 to 80 fs.

O 60.2 Thu 16:00 H42

Photoemission Electron Microscopy Study of Anthracene Thin Film Growth on Si(111) — •Niemma Buckanie and Frank Meyer zu Heringdorf — Universität Duisburg-Essen, Lotharstraße 1, 47057 Duisburg

The organic molecule anthracene consists of three linearly fused aromatic rings and has been successfully used in Organic Light Emitting Devices (OLED).The efficiency of the OLEDs depends strongly on the crystalline structure of the anthracene films. The frequency (color) of the emitted light is determined by an Exciton in the molecule.

We used linear and non-linear Photoemission Electron Microscopy (PEEM)to study the growth and the electronic properties of anthracene thin films on Si(111). Anthracene forms fractal-shaped islands with standing up molecules on top of a wetting layer that consists of flat-lying molecules. The illumination of the films with fs-laser pulses (E = 3.1 eV) enables us to determine the azimuthal alignment of the molecules in the textured films by using two photon-photoemission (2PPE). In 2PPE, a first photon excites the singlet exciton S1. The absorption of a second photon leads to the photoemission. The S1 state can only be excited with an electric field along the b-axis of the anthracene unit cell.

O 60.3 Thu 16:15 H42

Femtosecond electron dynamics in atomic wires: Si(557)-Au — •Tilman K. Rügheimer[1,2], Franz Himpsel[2], and Thomas Fauster[1] — [1]Lehrstuhl für Festkörperphysik, Universität Erlangen-Nürnberg, Staudtstr. 7, 91058 Erlangen — [2]Department of Physics, University of Wisconsin-Madison, 1150 University Ave, Madison WI 53706, USA

Atomic wires of noble metals such as gold on silicon surfaces serve as a model system for the investigation of one-dimensional electron systems. Recent experiments on Si(557)-Au have proven the existence of a spin-split surface state band below E_F [1] and have provided first information on the unoccupied part of the electronic band structure [2,3]. The dynamics of electrons has not been investigated so far.

We have carried out time-resolved two-photon photoemission experiments using femtosecond laser pulses. The lifetime of the previously reported image-potential resonance [2] is found to be smaller than 10 fs. At energies in the bulk band gap, the $I(\Delta t)$ curves are strongly asymmetric and cannot be explained by a single state and one decay channel. An interpretation using electron scattering between surface states in the gap is used to fit the data and reveals several lifetimes on the femtosecond and the picosecond scale.

[1] I. Barke et al., Phys. Rev. Lett. 97, 226405 (2006)
[2] J. A. Lipton-Duffin et al., Phys. Rev. B 73, 245418 (2006)
[3] T. K. Rügheimer et al., submitted

O 60.4 Thu 16:30 H42

Photoemission at the Si(001)-Ga surface using femtosecond lasers — Daniel Kampa, •Andrea Melzer, Jinxiong Wang, and Thomas Fauster — Lehrstuhl für Festkörperphysik, Universität Erlangen-Nürnberg, Staudtstr. 7, D-91058 Erlangen

The Si(001) (2×2)-Ga surface was used to investigate time-dependent Ga(3d) core-level shifts by pumping electrons from the valence to the conduction band. The pump-probe experiments were done by exciting the carriers with 1.59 eV laser pulses and probing the Ga(3d) core level with higher harmonics. These were generated by focussing laser pulses with 1.4 mJ energy, 30 fs pulse length and 779 nm wavelength from a multipass amplifier at a repetition rate of 1 kHz into argon. For the 25th harmonic used, the time resolution of the experiment was ∼ 400 fs after the grating monochromator. The band bending of about 110 meV of the p-doped Si(001) (2×2)-Ga surface is completely lifted by illumination of the surface with the 1.59 eV laser pulses. The Ga(3d) core level shows a slow time-dependent shift attributed to the build-up (∼ 1 ns) and decay (∼ 100 ns) of the photovoltage. The Ga(3d) core-level shift in the subpicosecond range is determined to be < 12 meV at the used pump pulse intensity of 10 mJ/cm^2.

O 60.5 Thu 16:45 H42

Ultrafast Vibrational Dynamics of CO/Ir(111) — •Heike Arnolds, Ian Lane, and David King — Department of Chemistry, University of Cambridge, Lensfield Road, Cambridge CB2 1EW, UK

We present a set of experiments that provide a complete mapping of coherent and incoherent vibrational relaxation times for a saturated layer of CO on a metal surface, Ir(111). Using femtosecond infrared pump - sum frequency probe spectroscopy, we measure the vibrational spectrum, the free induction decay and the vibrational lifetime. We also present the first measurement of a mid-infrared photon echo from a metallic surface, some fifteen years after the analogous measurement on a semiconductor surface. We find that the spectral linewidth (∼5 cm^{-1}) in this strongly dipole-coupled system is dominated by lifetime broadening and inhomogeneity, with only a small contribution from pure homogeneous dephasing due to coupling with low frequency modes. In addition, we see a change in echo peak shape with delay time, which shows that the inhomogeneity in this system is not static, instead frequency fluctuations contribute on different time scales.

O 60.6 Thu 17:00 H42

Femtosecond laser induced diffusion of CO on vicinal Pt(111)-surfaces — •Marco Lawrenz, Krisztina Stépán, Jens Güdde, and Ulrich Höfer — Fachbereich Physik und Zentrum für Materialwissenschaften, Philipps-Universität Marburg, D-35032 Marburg, Germany

Femtosecond laser induced surface diffusion of CO from step edges onto terraces of a vicinal Pt(111) surface has been studied at low substrate temperature using optical second harmonic generation (SHG) as a sensitive probe of the step coverage. We have investigated the diffusion rate as a function of laser fluence and delay between two time-delayed excitation pulses in a two-pulse correlation scheme. Compared to our previous studies on the diffusion of atomic oxygen on vicinal Pt(111) [1], we observe a weaker nonlinear dependence of the diffusion rate on laser fluence ($\propto F^6$) in a fluence range of F = 2–4 mJ/cm^2. The two-pulse correlation, on the other hand, has a narrower width of only ≈500 fs, which unambiguously show that the excitation mechanism for CO diffusion is also mediated by the substrate electron- rather than the phonon-system. As in the case of oxygen, the narrow two-pulse correlation can be described within the generalized one-dimensional electronic friction model only, if we assume an electron-temperature-dependent friction coefficient.

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

O 60.7 Thu 17:15 H42

Spin-dependent lifetimes of hot electrons in Co/Cu(100) revisited — •Ilja Panzer[1,3], Andreas Goris[1,3], Martin Pickel[2], Anke Schmidt[1,3], Fabian Giesen[1], Markus Donath[2], and Martin Weinelt[1,3] — [1]Max-Born-Institut, Max-Born-Straße 2A, 12489 Berlin — [2]Physikalisches Institut, Universität Münster, Wilhelm-Klemm-Straße 10, 48149 Münster — [3]Freie Universität Berlin, Arnimallee 14, 14195 Berlin

There is an ongoing controversy over hot electron lifetimes in ferromagnetic metals from the experimental as well as the theoretical point

of view [1,2]. From the various experimental approaches to measure the lifetimes of excited electrons no consistent picture of the underlying processes has emerged. For ferromagnetic thin films it is widely accepted that the lifetime of excited electrons is spin dependent, but the ratio between majority and minority lifetimes is still not settled.

We have measured the spin-dependent lifetimes of hot electrons between 250 and 750 meV above E_F for 20 ML Co/Cu (100) with time-resolved two-photon photoemission. Hot electrons are created by a 1.5 eV IR pulse of 3 nJ and probed with a significantly weaker UV pulse with the UV photon energy tuned slightly below the work function thus reducing the background of direct photoemission. We find significantly different lifetimes for minority and majority electrons. The implication for ultrashort magnetic switching will be discussed.

[1] Aeschlimann et al., Phys. Rev. Lett. **79**, 5158 (1997)
[2] Zhukov et al., Phys. Rev. Lett. **93**, 096401 (2004)

O 60.8 Thu 17:30 H42

The Laser-Assisted Photoelectric Effect on Surfaces — •Aeschlimann Martin[1], Luis Miaja-Avila[2], Guido Saathoff[2], Margaret M. Murnane[2], and Henry C. Kapteyn[2] — [1]Department of Physics, University of Kaiserslautern, Kaiserslautern, Germany D-67663 — [2]Department of Physics and JILA, University of Colorado, Boulder, CO 80309-0440

While the laser-assisted photoelectric effect (LAPE) in atoms is well established for the characterization of femtosecond EUV pulses, the equivalent process in the original manifestation of the photoelectric effect -photoemission from surfaces- has not therefore been studied.

In the present work, we present the first observation of the LAPE process in two-color surface photoemission [1]. In our experiment, IR pulses of 30 fs duration and 1.5 mJ at 780 nm are produced by a Ti:sapphire laser system and split into probe and pump. The probe beam is upconverted to the EUV using phase-matched high harmonic generation in a hollow fiber. A pair of Si:Mo multilayer mirrors spectrally selects the 27th harmonic (30 nm). The pump beam is directed through an optical delay arm and spatially and temporally overlapped with the EUV beam on a Pt(111) sample. A time-of-flight detector then measures the kinetic energy of the photoemitted electrons. In the presence of the pump pulse, these electrons can either absorb or emit an IR photon leading to sidebands in the EUV photoelectron spectrum. This 'dressing' of the continuum states is visible as steps in the Fermi edge.

[1] L. Miaja-Avila et al, Phys. Rev. Lett. 97, 113604 (2006)

O 60.9 Thu 17:45 H42

Space-,Time- and Energy- resolved PEEM at deposited Clusters — •Martin Rohmer, Carsten Wiemann, Michael Bauer, and Martin Aeschlimann — Department of Physics, University of Kaiserslautern, Kaiserslautern, Germany D-67663

Photoemission electron microscopy (PEEM) is a common technique to map the lateral distribution of the photoemission yield from micro- and nanostructured samples with sub-micron spatial resolution. Energy-resolved PEEM combined with the time-resolved two-photon photoemission technique (TR 2PPE) enables us to probe at the same time the ultrafast dynamics of electron excitations at surfaces (Lifetime mapping).

In this presentation we show that this setup is capable to identify very small differences (1-3 fs) in the excited state lifetime of supported clusters as result of lateral variations in the coupling efficiency to the underlying HOPG substrate. The high sensitivity of time-resolved PEEM to these temporal inhomogenities is related to the parallel acquisition mode of this technique, which significantly reduces the resolution limiting influence of systematic error arising from temporal fluctuations in the experimental parameters.

O 60.10 Thu 18:00 H42

Laser-induced ultrafast modification of electronic structure studied by femtosecond x-ray spectroscopy — •Niko Pontius, Christian Stamm, Torsten Kachel, Marko Wietstruk, Hermann A. Dürr, and Wolfgang Eberhardt — BESSY m.b.H., Albert-Einstein-Str. 15, 12489 Berlin, Germany

Femtosecond laser-induced excitation and relaxation processes of valence electrons in metals have so far been intensely studied by time-resolved techniques exclusively probing the valence electron response. However, including core level electrons in the probing process by using ultrashort x-ray pulses we demonstrate that additional, novel information about the transient modification of the electronic structure can be obtained.

In a laser-pump and x-ray-probe experiment at the BESSY 'femtosecond slicing' source we found for Ni that upon optical excitation the L_3 absorption line undergoes a $\sim$200 meV shift to lower energies. This shift occurs within the experimental temporal resolution of 150$\pm$50 fs. Comparing the experimental data with cluster calculations suggests that the observed L_3 absorption line shift is associated with an increase of valence electron localization inducing a shift of the 2p-core level to lower binding energy. This localization effect could be caused by enhanced scattering of itinerant electrons with electron-hole excitations. Such a modification of the electronic structure as a result of the electronic system perturbation has so far been unknown.

O 60.11 Thu 18:15 H42

A soft x-ray energy-dispersive beamline for NEXAFS and other CFS/CIS studies — •D. R. Batchelor[1], Th. Schmidt[1], R. Follath[2], Ch. Jung[2], R. Fink[3], M. Knupfer[4], B. Büchner[4], and E. Umbach[1] — [1]Universität Würzburg, Experimentelle Physik II — [2]BESSY GmbH, Berlin — [3]Physikalische Chemie II, Universität Erlangen-Nürnberg — [4]IFW Dresden

Energy-dispersive near edge x-ray absorption fine structure (NEXAFS) spectroscopy is a well-known technique in the hard x-ray regime. By combining the photon energy dispersion of a plane-grating XUV monochromator with the imaging properties of a hemispherical electron energy analyzer, soft x-ray energy dispersive electron spectroscopy is possible. This multiplex technique allows not only NEXAFS without time-consuming scanning of the photon energy but also high-resolution CFS/CIS spectroscopic studies such as Auger/autoionization spectroscopy and photoelectron diffraction. We present the design for an upgrade of the BESSY UE52-PGM beamline and the results of a pilot experiment. Data from copper and its oxides together with phthalocyanines will be presented. Besides the common photoemission and Auger lines and the known participator and spectator decay channels the extensive, quasi-3-dimensional data sets clearly show that other excitations, such as "shake-up", play an important role in both, the normal and resonant Auger processes. The new technique provides data with much more accuracy and detail than hitherto achieved by simply stepping the photon energy.

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