O 22: Ultrafast Surface Dynamics

Time: Tuesday 10:30–13:00 Location: MA 042

Invited Talk O 22.1 Tue 10:30 MA 042

Electron dynamics at molecule-semiconductor interfaces — •Katrin R. Siefermann — Leibniz-Institute of Surface Modification (IOM), Leipzig, Germany

Charge transfer processes across hybrid interfaces, such as formed by the connection of molecules to semiconductors, play an increasingly important role in a variety of emerging technologies. Detailed understanding of interfacial charge transfer in these systems, however, remains a major challenge for experiments and theory.

In my talk I will present a new approach to monitor photo-induced electron transfer from a molecule to a semiconductor material with sub-picosecond temporal resolution and from the perspective of well-defined atomic sites [1]. Combining femtosecond time-resolved X-ray photoelectron spectroscopy with constrained density functional theory, we are able to identify the nature of an intermediate electronic state that precedes free charge carrier generation in a film of dye-sensitized ZnO nanocrystals after photoexcitation of the dye with visible light. The findings demonstrate a new capability to monitor charge transfer in complex hybrid materials. This presentation will further include our latest results of electron dynamics at interfaces.

[1] K. R. Siefermann, C. D. Pemmaraju, S. Neppl, A. Shavorskiy, et al., J. Phys. Chem. Lett. 5, 2753-2759 (2014).

O 22.2 Tue 11:00 MA 042

Time-resolved hard-x-ray photoelectron spectroscopy at the x-ray free-electron laser SACLA — •Lars-Philip Oloff[1,2], Masaki Oura[2], Ashish Chainani[2], and Kai Rossnagel[1,2] — [1]Institut für Experimentelle und Angewandte Physik, Universität Kiel, 24118 Kiel, Germany — [2]RIKEN, SPring-8 Center, Sayo-cho, Hyogo, 679-5148, Japan

Time-resolved hard x-ray photoelectron spectroscopy (trHAXPES) is established as a novel ultrafast spectroscopy technique using the x-ray free-electron laser SACLA (Japan) [1,2]. The technique extends time-resolved photoemission into the hard x-ray regime and, as a core-level spectroscopy, combines atomic-site and chemical-state specificity with femtosecond time-resolution and bulk sensitivity. It can thus open novel opportunities for probing ultrafast electron dynamics in strongly excited materials, at buried interfaces, and in electronic devices under *in operando* conditions.

The viability of trHAXPES using 8 keV x-ray free-electron laser radiation is demonstrated by a systematic investigation of probe and pump pulse-induced vacuum space-charge effects. The measured time and excitation energy dependencies of spectral shifts and broadenings are compared to the results of N-body numerical simulations and simple analytic (mean-field) models.

[1] M. Oura *et al.*, J. Synchrotron Rad. **21**, 183 (2014)
[2] L.-P. Oloff *et al.*, New J. Phys., in press (2014)

O 22.3 Tue 11:15 MA 042

Narrowband high harmonic pulses for trARPES using frequency-upconverted Ti:Sapphire lasers — •S. Emmerich[1], S. Eich[1], A. Stange[3], A.V. Carr[2], J. Urbancic[1], T. Popmintchev[2], M. Wiesenmayer[1], K. Jansen[3], A. Ruffing[1], S. Jakobs[1], T. Rohwer[3], S. Hellmann[3], C. Chen[2], P. Matyba[2], L. Kipp[3], K. Rossnagel[3], M. Bauer[3], M.M. Murnane[2], H.C. Kapteyn[2], S. Mathias[1], and M. Aeschlimann[1] — [1]University of Kaiserslautern and Research Center OPTIMAS, 67663 Kaiserslautern, Germany — [2]JILA, University of Colorado and NIST, Boulder, CO 80309-0440, USA — [3]Institute of Experimental and Applied Physics, University of Kiel, D-24098 Kiel, Germany

trARPES using femtosecond XUV high harmonics has recently emerged as a powerful tool for investigating ultrafast quasiparticle dynamics in correlated-electron materials. However, the full potential of this approach has not yet been achieved because, to date, high harmonics generated by 800 nm wavelength Ti:Sapphire lasers required a trade-off between photon flux, energy and time resolution. Photoemission spectroscopy requires a quasi-monochromatic output, but dispersive optical elements that select a single harmonic can significantly reduce the photon flux and time resolution. Here we show that shorter wavelength driven high harmonic extreme-ultraviolet trARPES is superior to using 800 nm laser drivers since it eliminates the need for any spectral selection, thereby increasing photon flux and energy resolution while preserving excellent time resolution [Eich et al., J. Electron Spectrosc. 195 (2014) 231-236].

O 22.4 Tue 11:30 MA 042

Thermalization dynamics in graphite - Phonon-phonon interaction vs. supercollision — •Ankatrin Stange, Christian Sohrt, Lexian Yang, Gerald Rohde, Petra Hein, Lars Oloff, Kerstin Hanff, Kai Rossnagel, and Michael Bauer — Institute of Experimental and Applied Physics, University of Kiel, Germany

Recent studies reported on a defect-assisted (supercollision) process yielding an enhanced coupling between optically excited electrons and acoustic phonons in graphene [1,2] and accelerating electron-lattice thermalization down to timescales in the few picosecond regime. In this work we address the question to what extent also the relaxation dynamics in the parent compound graphite is governed by supercollision. Time- and angleresolved photoelectron spectroscopy is used to monitor the transient carrier population in the π-bands at the H point at absorption of intense near-infrared laser pulses. In correspondence to other works the relaxation due to interaction with the lattice proceeds on two characteristic timescales: A fast (≈250 fs) cool down of the electron gas arising from the interaction of the electronic system with strongly coupled optical phonons (SCOPs) [3] is followed by a picosecond dynamics that can consistently be reproduced within a three-temperature model only if the coupling between SCOPs and acoustic phonons is taken into account [3]. In contrast, the supercollision model clearly fails in describing the recorded electron temperature transients.

[1] J. C. W. Song *et al.*, Phys. Rev. Lett. **109**, 106602 (2012)
[2] J. C. Johannsen *et al.*, Phys. Rev. Lett. **111**, 027403 (2013)
[3] T. Kampfrath *et al.*, Phys. Rev. Lett. **95**, 187403 (2005)

O 22.5 Tue 11:45 MA 042

Second Harmonic Spectroscopy in the Reststrahlen Band of SiC Using an Infrared Free Electron Laser — •Alexander Paarmann, Alexey Melnikov, Sandy Gewinner, Wieland Schoellkopf, and Martin Wolf — Fritz Haber Institute of the Max Planck Society, Berlin

We experimentally study the efficiency of optical second harmonic generation in the highly reflective Reststrahlen spectral region of the wide-band gap semiconductor 6H-SiC, employing tunable picosecond mid-infrared laser pulses generated from the FHI free electron laser. The Reststrahlen region, located between longitudinal and transversal optical phonon resonances, provides a unique window to study nonlinear effects in the phononic response of a crystal. We observe sharp resonances of the second harmonic signal at both transverse and longitudinal optical phonon fundamental frequencies, the latter being unexpected since no resonance in the optical nonlinearity is predicted for longitudinal modes. We discuss the underlying mechanism of local field enhancement, which is tightly linked to the largely dispersing linear optical properties in the Reststrahlen band. The negative real part of the dielectric function results in rapidly attenuated evanescent waves in this region, making these experiments sensitive to a thin near-surface layer. The resulting large amplitude atomic motions induced by intense mid-infrared laser excitation opens the door to future studies of nonlinear phononic response at semiconductor surfaces and interfaces.

O 22.6 Tue 12:00 MA 042

Experimental determination of the potential energy curve of diethyl ether on Si(001) — Marcel Reutzel[1], Marcus Lipponer[1], •Michael Dürr[1,2], and Ulrich Höfer[1] — [1]Philipps University, 35037 Marburg — [2]Justus Liebig University, 34392 Giessen

The adsorption dynamics of organic molecules on semiconductor surfaces is in most cases controlled by a precursor or intermediate state. For molecules containing a heteroatom such as nitrogen or oxygen, this intermediate state involves lone pair electrons of the heteroatom and, in the case of Si(001), the empty dangling bond of the lower silicon atom. Most recently, we have shown that in the case of ether molecules one can isolate this datively bonded intermediate at low temperatures; at elevated temperatures cleavage of the O-C bond of the otherwise inert ether group was observed [1].

Here we show that, using optical second harmonic generation, we

can follow in situ the conversion of diethyl ether from the intermediate to the final adsorption configuration. Measuring the kinetics as a function of surface temperature allows us to determine the associated energy barrier. Complementary information is obtained from sticking probabilities measured by means of molecular beam experiments. The dependence of the initial sticking probability on surface temperature reveals the energy difference between the conversion and desorption barrier out of the datively bonded intermediate state. Combination of the two results give full information on the potential energy curve for the adsorpton of diethyl ether on Si(001).

[1] G. Mette, *et al.*, ChemPhysChem **15**, 3725 (2014).

O 22.7 Tue 12:15 MA 042

Polarization dependent photoinduced voltage in Sb_2Te_3 — •Thomas Schumann[1], Maria Mansurova[2], Fabio Boschini[3], Cinja Seick[2], Gregor Mussler[4], Tobias Kampfrath[5], and Markus Münzenberg[1] — [1]Institut für Physik, Ernst-Moritz-Arndt-Universität Greifswald,Germany — [2]I.Physikalisches Institut,Georg-August Universität Göttingen,Germany — [3]Dipartimento di Fisica,Politecnico di Milano, Italia — [4]Peter-Grünberg Instiut,Forschungszentrum Jülich,Germany — [5]Abteilung für Physikalische Chemie,Fritz-Haber-Institut Berlin(MPG),Germany

Topological insulators potentially open up a novel route in spin based electronics. The energy momentum landscape of the Dirac cone provides, via spin-momentum locking, a control of the directionality of spin currents [1]. Its femtosecond control with light pulses has been demonstrated. The used excitations are realized with femtosecond laser pulses (at 1.55 eV) with different pump-pulse polarization. We demonstrate coherent femtosecond control of the dynamics in presence of the pump-pulse and the DC detection of helicity dependent photo currents. Both are compared to possible optical transitions in ab-initio calculations.

Furthermore we thank the DFG-SPP1666.

[1] J. W. McIver et al., Nat. Nanotech. 7, 96(2012)

O 22.8 Tue 12:30 MA 042

Development of Ultrafast Low-Energy Electron Diffraction — •Max Gulde[1], Simon Schweda[1], Gero Storeck[1], Anastassia Rissanou[2], Sebastian Schramm[1], Manisankar Maiti[1], Hak Ki Yu[3,4], Alec Wodtke[3], Vagelis Harmandaris[2], Sascha Schäfer[1], and Claus Ropers[1] — [1]4th Physical Institute, University of Göttingen, Göttingen, Germany — [2]Department of Applied Mathematics, University of Crete, 71409 Heraklion, Greece — [3]Max Planck Institute for Biophysical Chemistry, 37077 Göttingen, Germany — [4]Institute for Physical Chemistry, University of Göttingen, 37077 Göttingen, Germany

Ultrafast electron diffraction experiments have offered intriguing insights into atomic-scale structural dynamics in bulk media on their fundamental time scales [1]. In contrast, comparable time-resolved studies in quasi two-dimensional systems such as surfaces and interfaces remain challenging, despite recent accomplishments [2, 3]. Here, we introduce ultrafast low-energy electron diffraction (ULEED) in transmission as a powerful new tool for ultrafast surface science [4]. In particular, we present a detailed experimental and theoretical account of the relaxation of a polymer superstructure on free-standing graphene from a highly out-of-equilibrium state. Finally, first data from a ULEED experimental setup operated in back-reflection is presented.

[1] B. J. Siwick et al., Science 302, 1382-1385 (2003).
[2] S. Schäfer et al., J. Chem. Phys. 135, 214201 (2011).
[3] A. Hanisch-Blicharski et al., Ultramicroscopy 127, 2-8 (2013).
[4] M. Gulde et al., Science 345, 200 (2014).

O 22.9 Tue 12:45 MA 042

Velocity map imaging of ultrafast electron emission from fs-laser illuminated nanoscale metal tips — Alexander Bainbridge and •William Bryan — Department of Physics, Swansea University, Singleton Park, Swansea SA2 8PP, UK

We have recently demonstrated velocity map imaging (VMI) of photoelectron emission from a metal nanotip illuminated with a strong field laser pulse [1]. We will present theoretical and experimental results demonstrating the simultaneous momentum resolution of electrons emitted from gas and solid targets. Above threshold ionization from krypton gas facilitates momentum calibration, and tunnelling from a metal nanotip exhibits a hemispherical momentum distribution defined by discrete photon absorption. Numerical simulations show VMI condition are maintained over a wide range of instrument configurations such that momentum focusing is independent of the emission location.

Forthcoming experiments will involve cryogenic sharpening of nanotips to a single atom, traditionally observed with field ion microscopy. As the nanotip approaches a single atom, the observed velocity map image will tend towards that of a gas phase atom, showing recollisional interference. This hybrid of real-space and momentum-space imaging will be decisive in the design of next-generation femtosecond electron diffraction and microscopy instruments.

[1] A R Bainbridge and W A Bryan, New J. Phys. 16 (2014) 103031