O 69: Ultrafast Electron and Spin Dynamics

Time: Wednesday 18:15–21:00 Location: Poster A

O 69.1 Wed 18:15 Poster A

Influence of linkers on electron dynamics in dye-functionalized SiC — •Nils Fabian Kleimeier[1], Deb Kumar Bhowmick[1], Linda Stegemann[1,2], Cristian Alejandro Strassert[1,2], and Helmut Zacharias[1,2] — [1]Physikalisches Institut, Universität Münster, Wilhelm-Klemm-Str. 10, 48149 Münster — [2]Center for Nanotechnology (CeNTech), Universität Münster, Heisenbergstrasse 11, 48149 Münster

Functionalized SiC surfaces have gained wide interest in the field of biosensors, transistors and OLEDs due to the bio-inertness, high stability and large, tunable band gap of SiC. The interaction between the organic dye and the SiC surface can be tuned by using different linker molecules. To investigate the effect of the linkers, benzo[ghi]perylene dye was attached to the surface using p-aminophenyltrimethoxysilane (p-APTMS), aminopropyl-triethoxy-silane (APTES) and aminoundecyltriethoxysilane (AUDTES). Fluorescence images and confocal microscopy were used to determine the distribution of the dyes on the surface and the radiative lifetimes which are in the range of 1-10 ns [1]. To determine the efficiency of electron injection into the SiC, non-radiative lifetimes of about 100 fs were measured using time-resolved two-photon photoemission (TR-2PPE).

[1] Deb Kumar Bhowmick, Steffen Linden, André Devaux, Luisa De Cola, Helmut Zacharias, Small 8, 592 (2012)

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Femtosecond time- and angle-resolved photoemission spectroscopy of 1*T*-$TaSe_2$ — •Florian Diekmann, Christian Sohrt, Ankatrin Stange, Michael Bauer, and Kai Rossnagel — Institut für Experimentelle und Angewandte Physik Universität Kiel, D-24098 Kiel

According to previous angle-resolved photoemission spectroscopy and scanning tunneling microscopy studies, the surface of 1*T*-$TaSe_2$ displays combined Peierls-Mott insulating behavior for temperatures below about 260-300 K, while bulk sensitive resistivity measurements show metallic character and no indication for a Mott transition [1]. Here, we use time- and angle-resolved extreme ultraviolet photoemission spectroscopy to directly determine the momentum-dependent electronic structure dynamics of this surface phase on the femtosecond time scale [2]. The extracted spectroscopic order parameters reveal a global two-time-scale dynamics indicating a quasi-instantaneous loss of the Mott and charge-density-wave electronic orders and a subsequent coherent suppression of the lattice distortion on a time scale related to the frequency of the charge-density-wave amplitude mode. Specifically after one half-cycle of coherent amplitude-mode vibration, a crossover state between insulator and metal with partially filled-in and partially closed Mott and Peierls gaps is reached.

[1] L. Perfetti *et al.*, Phys. Rev. Lett. **90**, 166401 (2003).
[2] C. Sohrt *et al.*, Faraday Discuss. **171**, 243 (2014).

O 69.3 Wed 18:15 Poster A

Spectroscopy and dynamics of unoccupied electronic statesof the topological insulators Sb_2Te_3 and Sb_2Te_2Se — •Johannes Reimann, Jens Güdde, Kenta Kuroda, and Ulrich Höfer — Fachbereich Physik und Zentrum für Materialwissenschaften, Philipps-Universität, D-35032 Marburg

The electronic structure and ultrafast electron dynamics of the *p*-doped topological insulator Sb_2Te_3 and its derivative Sb_2Te_2Se was investigated by means of time- and angle-resolved two-photon photoemission (2PPE). Our experiment reveal that the massless Dirac-cone like energy dispersion of topological surface states is realized above the Fermi energy in both materials. In agreement with theoretical predictions we find that the unoccupied bands of Sb_2Te_2Se are shifted to higher energies as compared to Sb_2Te_3. The observed electron dynamics in the topological surface state (TSS) of both materials is, however, very similar, which is a signature that the decay is dominated by the number of unoccupied states in the valence band. At lower temperatures at which the electron mobility in the bulk is enhanced, the decay becomes surprisingly faster, which strongly indicates that transport perpendicular to the surface into the bulk conduction band is important for the decay of the TSS.

O 69.4 Wed 18:15 Poster A

Towards ultrafast X-ray condensed matter physics with MHz repetition rate HHG sources — •R. Klas[1,2], S. Hädrich[2,3], J. Rothhardt[2,3], S. Eich[1], M. Barkowski[1], J. Urbancic[1], M. Aeschlimann[1], J. Limpert[2,3,4], and S. Mathias[1] — [1]TU Kaiserslautern and Research Center OPTIMAS, Kaiserslautern — [2]Friedrich Schiller University Jena, Abbe Center of Photonics, Institute of Applied Physics, Jena — [3]Helmholtz-Institute Jena, Jena — [4]Fraunhofer Institute for Applied Optics and Precision Engineering, Jena

In recent years, an increasing number of ultrafast material science experiments are based on the use of table-top high-harmonic generation (HHG) lightsources [1,2]. Despite the huge success of these first experiments, a major drawback has been the limited repetition rate of these lightsources, in particular with respect to all types of photoemission experiments. Here, we combine newly developed high-repetition rate HHG sources [3,4] with state-of-the-art element-specific magneto-optical Kerr experiments. The chances and prospects for future ultrafast materials science experiments with these lightsources will be discussed.

[1] S. Mathias et al., Springer, chapter in "Ultrafast Nonlinear Optics", Editors R. Thomson, C. Leburn, D. Reid (2013)
[2] S. Mathias et al., JESRP 189, 164 (2014)
[3] J. Rothhardt et al., Opt. Lett. 39, 5224 (2014)
[4] S. Hädrich et al., Nat Phot 8, 779 (2014)

O 69.5 Wed 18:15 Poster A

Coherent and incoherent electronic excitations in the Mott insulator 1*T*-TaS_2 — •Manuel Ligges[1], Isabella Avigo[1], Simon Freutel[1], Matthias Kalläne[2], Ping Zhou[1], Lutz Kipp[2], Kai Rossnagel[2], and Uwe Bovensiepen[1] — [1]Fakultät für Physik und Zentrum für Nanointegration, Universität Duisburg-Essen — [2]Institut für Experimentelle und Angewandte Physik, Universität Kiel

Using femtosecond time- and angle-resolved photoemission spectroscopy we study the partial quench of charge order in the prototypical Mott insulator 1*T*-TaS_2 after optical excitation. An unoccupied electronic state is observed in the correlated (spectrally gapped) commensurate and nearly-commensurate charge density wave (CDW) phases of the material that is found to be abscent in the (almost metallic) incommensurate CDW phase and, thus, appears to be a direct measure of short range correlations. From its energetic position (E-E_F=170 meV), this state might be identified as the upper Hubbard band. The population of this coherent feature follows the temporal laser pulse profile and differs significantly from the incoherent electronic population dynamics in the same energetic window and $\boldsymbol{k}$-region ($\bar{\Gamma}$) that shows clear life time effects ($\tau \approx 150$ fs). We conclude that this state directly reflects the presence of charge order in the system which promptly collapses in the presence of the laser field, leaving the system behind in a state similar to the high-temperature metallic phase.

O 69.6 Wed 18:15 Poster A

2D RABBITT spectroscopy for the investigation of attosecond dynamics — •Martin Piecuch, Martin Aeschlimann, and Stefan Mathias — University of Kaiserslautern and Research Center OPTIMAS, TU Kaiserslautern, 67663 Kaiserslautern, Germany

Reconstruction of attosecond beating by interference of two-photon transitions (RABBITT) is a powerful and well-established method for attosecond pulse characterization [Paul et al., Science 292,1689 (2001)]. Additionally, the RABBITT technique has been very useful to measure and understand basic dynamics in the high-harmonic generation process itself in the past. Here, we present theoretical calculations of an extension of the RABBITT scheme by combining it with modern techniques derived from 2D spectroscopy: excitation with multiple laser pulses, and phase cycling to select specific coherence pathways. We show that these methods open up new and powerful routes to study attosecond dynamics in atoms, molecules, and materials in the future.

O 69.7 Wed 18:15 Poster A

Electronic Structure and Excitation Dynamics of the CuPc/PTCDA/Ag(111) Heterointerface — •Jonas Zimmermann, Andreas Namgalies, Nico Armbrust, and Ulrich Höfer — Fachbereich Physik und Zentrum für Materialwissenschaften, Philipps-Universität, D-35032 Marburg

We grow well-defined monolayers of copper(II)phthalocyanine (CuPc)

on top of monolayers of perylene-tetracarboxylic acid dianhydride (PTCDA) on Ag(111) as model systems for organic donor-acceptor heterointerfaces. We employ time-resolved two-photon photoemission (2PPE) to examine the dynamics of charge transfer processes at these interfaces. An optical parametric oscillator (OPO) enables us to tune the pump photon energy to the HOMO-LUMO transition of CuPc. The time-resolved 2PPE spectra reveal the subsequent decay of the excitation. In case of the PTCDA monolayer the excited electron cannot be transferred from the LUMO of CuPc to unoccupied PTCDA orbitals because the former LUMO of PTCDA is shifted below the Fermi level by chemisorption. Instead, we observe an electron transfer into the interface state of PCTDA/Ag(111). This metal-organic hybrid state arises from the Shockley surface state of clean Ag(111) and is located at 0.6 eV above the Fermi level. The electron transfer from CuPc shows up as resonantly enhanced and temporarily delayed 2PPE intensity of the interface state when the pump photon energy is tuned through the CuPc HOMO-LUMO transition at 1.8 eV.

O 69.8 Wed 18:15 Poster A

Influence of the density of states on nonequilibrium dynamics in metals — •Sebastian Weber[1], Benedikt Y. Mueller[2], and Baerbel Rethfeld[1] — [1]Technische Universität Kaiserslautern, Germany — [2]Max-Planck-Institut für Intelligente Systeme, Germany

After excitation with an ultrashort laser pulse, the electrons in a metal are in a strong nonequilibrium state. During thermalization, they interact also with the lattice, transferring energy to the phonons. On the basis of complete Boltzmann-type collision integrals, a method allowing to treat materials with an arbitrary density of states has been developed [Phys. Rev. B 87, 035139 (2013)]. This method provides insights into the response of different material classes to ultrafast laser excitation. Note that this approach also opened the way to describe ultrafast magnetization dynamics, see [Phys. Rev. Lett. 111, 167204 (2013)].

Here, we study the microscopic relaxation processes, in particular thermalization and the electron-phonon coupling strength under nonequilibrium conditions. Results for gold [Appl. Surf. Sci. 302, 24 (2014)] are compared to those obtained for other metals like copper and silver.

O 69.9 Wed 18:15 Poster A

Time-resolved photoelectron spectroscopy in a back-pump/front-probe geometry — •Gerrit Horstmann, Simon Lange, Claus Ropers, and Sascha Schäfer — IV. Physical Institute, Friedrich-Hund-Platz 1, University of Göttingen, Germany

The combination of ultrafast pump-probe techniques with angle-resolved photoelectron spectroscopy (ARPES) has enabled the detailed investigation of ultrafast electron dynamics in condensed matter. However, the influence of fundamental transport processes such as electron scattering in bulk material or at interfaces as well as propagation through functional layers is difficult to access in time-resolved photoelectron spectroscopy (tr-PES). Here, we present a flexible tr-PES setup allowing for optical sample excitation at the backside of an ultrathin film combined with time- and angle-resolved photoelectron probing at the film's front surface. In a first step, we study the photoemission from back-pumped homogeneous metal films on sapphire substrates, characterizing the delay-dependent spectral and angular distributions of photoemitted electrons. Possible future applications are discussed, e.g., for the investigation of optically-triggered conductivity changes in correlated materials.

O 69.10 Wed 18:15 Poster A

Correlation of Chirp and Delay in Attosecond Streaking Measurements from W Surfaces — •Andreas Kim[1], Johannes Barth[1], Peter Feulner[1], Konrad Hütten[1], Reinhard Kienberger[1], Ferenc Krausz[2], and Stefan Neppl[3] — [1]TU-München, Physikdepartment, Garching, Germany — [2]MPI für Quantenoptik, Garching, Germany — [3]LBNL, Berkeley, USA

Attosecond streaking, i.e., the momentum-modulation of photoelectrons emitted by ultrashort XUV pulses into the strong field of a synchronized IR laser [R. Kienberger et al., Nature 427, 817], enables chronoscopy on the attosecond timescale. It has been used to explore time delays between the release of photoelectrons from different electronic levels with unprecedented resolution, from isolated particles [e.g. M. Schultze et al., Science 328, 1658] and condensed matter [A. Cavalieri et al., Nature 449, 1029; S. Neppl et al., PRL 109, 087401]. This method also provides information on characteristic parameters of the ultrashort XUV pulses like duration, central energy and chirp, i.e., the temporal variation of the photon energy across the pulse. We measure a direct correlation between the wave-packet chirp and relative time delay between conduction band and W4f core-level photoemission, with different chirp values for conduction band and W4f electrons. We show that reliable information on the excitation pulse, the electronic transport and screening processes can only be derived from such attosecond streaking data, if detection and evaluation artifacts are identified and accounted for. Supported by the Deutsche Forschungsgemeinschaft (Munich Centre for Advanced Photonics; MAP B.1.3 & B.1.4)