O 8: Electron and spin dynamics

Time: Monday 10:30–13:00 Location: A 060

O 8.1 Mon 10:30 A 060

Empty topological surface state on Bi_2Se_3 — •Daniel Niesner[1], Sergey Eremeev[2], Evgueni Chulkov[3], and Thomas Fauster[1] — [1]Universität Erlangen, Lehrstuhl für Festkörperphysik, Staudtstr. 7, 91058 Erlangen, Germany — [2]Tomsk State University, pr. Lenina 36, 634050 Tomsk, Russia — [3]Donostia International Physics Center (DIPC), and CFM, Centro Mixto CSIC-UPV/EHU, Departamento de Física de Materiales, UPV/EHU, 20080 San Sebastián, Spain

Topological insulators are characterized by spin-polarized, linearly dispersing surface states that arise from a symmetry inversion of the bulk bands at band gap edges. In case of Bi_2Se_3 the Fermi surface is formed by a single Dirac-cone at the $\overline{\Gamma}$-point.

Using laser-based photoemission with 6.2 eV photon energy we find the Dirac-point 0.3 eV below the Fermi energy.

In mono- and bichromatic two-photon photoemission we find a second linearly dispersing band 1.4 eV above the Fermi level with a group velocity of $5 \cdot 10^5 \frac{\mathrm{m}}{\mathrm{s}}$. Lifetimes amount to ≈ 74 fs. Circular dichroism indicates a high degree of spin polarization.

These data are in good agreement with DFT calculations showing a topologically protected surface state 1.5 eV above Fermi level. The calculations compare favorably with the other empty electronic states found in the two-photon photoemission experiments.

O 8.2 Mon 10:45 A 060

The role of the excitation pathway in the photoinduced melting of the CDW in 1*T*-$TiSe_2$ — •Gerald Rohde, Stefan Hellmann, Timm Rohwer, Christian Sohrt, Ankatrin Stange, Kerstin Hanff, Lutz Kipp, Michael Bauer, and Kai Rossnagel — Institut für Experimentelle und Angewandte Physik der Christian-Albrechts-Universität zu Kiel

Charge-density waves (CDWs) are broken-symmetry states of low-dimensional solids that are brought about by strong electron-phonon or electron-electron interaction. In a time-resolved XUV ARPES study we recently showed that the long-range order associated with the CDW phase in the transition metal dichalcogenide 1*T*-$TiSe_2$ can be destroyed within less than 30 fs after intense illumination with a 800 nm light pulse [1]. In this contribution we present complementary data recorded at pulsed 400 nm excitation. Surprisingly we find that for the short-wavelength excitation the CDW collapse is considerably slowed down. Our time-resolved ARPES data show furthermore that the initiating absorption processes take place at well separated locations in electron momentum space for the two different wavelengths. Possible scenarios that consider the effect of the absorption pathway on the phase transition dynamics will be discussed.

[1] T. Rohwer et al., Nature **471**, 490 (2011)

O 8.3 Mon 11:00 A 060

Electronic lifetimes in a Bi quantum well with Rashba spin-splitting — •Svenja Vollmar, Andreas Ruffing, Sebastian Jakobs, Alexander Baral, Mirko Cinchetti, Stefan Mathias, Martin Aeschlimann, and Hans Christian Schneider — University of Kaiserslautern

2-photon photoemission spectroscopy and ab-initio calculations suggest that a part of the band structure of a Bi monolayer (quantum well) on a Cu(111) substrate is well described [1] by a simple Rashba Hamiltonian, which includes the effects of the spin-orbit interaction together with an asymmetric confinement potential of a quantum-well. The understanding of the consequences of the Rashba spin-split electronic bands on the electronic spin dynamics is of general importance for spin transport and diffusion lengths in such systems.

Using a combination of 1-photon and 2-photon photoemission spectroscopy, we determine the energy and momentum dependent electronic lifetimes in the Bi quantum well, and find a plateau around the intersection of the Rashba bands. To understand this characteristic energy and momentum dependence, we calculate the electronic lifetime due to carrier-carrier scattering in a simple model of a 2D electron gas including a Rashba interaction with effective Rashba parameters derived from experiment. We reproduce the observed plateau in the lifetimes using this model, and explain this behavior by a change of available scattering phase space for electrons below and above the intersection of the Rashba bands.

[1] S. Mathias et al., Phys. Rev. Lett. 104, 066802 (2010)

O 8.4 Mon 11:15 A 060

Ultrafast melting of a charge-density wave in the Peierls insulator Rb:TaS_2 — •Kerstin Hanff, Stefan Hellmann, Timm Rohwer, Michael Bauer, Lutz Kipp, and Kai Rossnagel — Institute of Experimental and Applied Physics, University of Kiel, 24098 Kiel, Germany

Ultra-short high-harmonic pulses enable us to study ultrafast dynamics of condensed matter systems on femto- and picosecond time scales. In particular, they allow us to identify the nature and strength of interactions between various degrees of freedom in complex materials, in which typically two or more of the lattice, charge, spin and orbital degrees of freedom are strongly coupled. Our recent studies focused on the investigation of electronically driven charge-density waves employing time- and angle-resolved photoelectron spectroscopy. Here, we present the ultrafast dynamics of a lattice driven CDW system. Rb adsorption on 1T–TaS_2 serves as a model system revealing a pronounced metal-to-insulator transition. Induced by a charge transfer, this intercalation process leads to a structural change into a commensurate CDW phase. Upon strong photoexcitation by an optical pump pulse the electronic gap closes and the Peierls insulator Rb:TaS_2 undergoes a transition into a transient metallic state. We are able to distinguish different time scales during the excitation and relaxation processes, which indicate the influence of the electronic and lattice systems.

O 8.5 Mon 11:30 A 060

Singlet fission and efficient electron transfer from the "dark" multi-exciton state in pentacene — Wai-Lun Chan[1], •Manuel Ligges[1,2], Askat Jailaubekov[1], Loren Kaake[1], Luis Miaja-Avila[1], and Xiaoyang Zhu[1] — [1]Department of Chemistry & Biochemistry, University of Texas, Austin, TX 78712, USA — [2]Fakultät für Physik, Universität Duisburg-Essen, 78756 Duisburg

Singlet fission refers to the creation of two or more electron-hole pairs from the absorption of a single photon in organic chromophores. This process holds great promise in the field of photovoltaics, because it may lead to power conversion efficiencies exceeding the theoretical limit for single junction solar cells, the so called Shockely-Queisser limit [1]. One of the key challenges in the implementation of singlet fission is the efficient extraction of charge carriers. Using time-resolved two-photon photoemission on the model system pentacene/C_{60}, we show that the process of singlet fission is accompanied by efficient electron transfer from a multi-electron intermediate state, previously been referred to as the " dark " multi-exciton state [2]. In addition, we show that - contrary to common belief - singlet fission in pentacene cannot be described in terms of simple rate models but involves coherent coupling between the initially excited singlet exciton and the intermediate multi-exciton state.

[1] W. Shockley & H. J. Queisser, J. Appl. Phys. **32**, 510 (1961)
[2] P. M. Zimmerman et al., Nature Chem. **2**, 648 (2010)

O 8.6 Mon 11:45 A 060

The role of intermediate states in two-photon photoemission from Si(001) — •Henning Husser and Eckhard Pehlke — Institut für Theoretische Physik und Astrophysik, Universität Kiel, Germany

Two-photon photoemission (2PPE) spectra contain detailed information about the unoccupied states [1], and time resolved 2PPE can reveal the dynamics. We have developed a non-perturbational approach to calculate photocurrents by time-dependent simulations and applied it to the calculation of normal-emission photocurrents from Si(001). Here we focus on the analysis of 2PPE Si(001) normal emission spectra from the simulations in the light of the expression known from perturbation theory. The intermediate states occurring in this expression are linked to both the initial state and to the final state by matrix elements. Intermediate states can become prominent in the spectra if they couple strongly to the initial states or the final states. Variation of the polarization of the light wave can be used to tune the matrix elements via the respective selection rules. Resonances in the emission intensity for a fixed initial state as a function of photon energy can be assigned to intermediate states. This will be used to derive information about the intermediate states in case of normal-emission 2PPE

from Si(001). In particular the role of surface resonances, as reported by Fauster et al. [2], will be investigated.

[1] W. Schattke et al., Phys. Rev. B 78, 15534 (2008).
[2] T. Fauster et al., Verhandl. DPG (VI) 46, 1/490 (2011).

O 8.7 Mon 12:00 A 060

Orbital Symmetry Dependent Electron Transfer through Molecules Assembled on Metal Substrates — •Florian Blobner[1], Pedro Coto[2], Francesco Allegretti[1], Michel Bockstedte[2], Oscar Rubio-Pons[2], Haobin Wang[3], David Allara[4], Michael Zharnikov[5], Michael Thoss[2], and Peter Feulner[1] — [1]Physik-Department E20, Technische Universität München, 85748 Garching, Germany — [2]Institut für Theoretische Physik und Interdisziplinäres Zentrum für Molekulare Materialien, Friedrich-Alexander-Universität Erlangen-Nürnberg, 91058 Erlangen, Germany — [3]Department of Chemistry and Biochemistry, MSC 3C, New Mexico State University, Las Cruces, 88003 New Mexico, USA — [4]Departments of Chemistry and Material Science, Pennsylvania State University, 16802 Pennsylvania, USA — [5]Angewandte Physikalische Chemie, Universität Heidelberg, 69120 Heidelberg, Germany

Femtosecond charge transfer dynamics in self-assembled monolayers of cyano-terminated ethane-thiolate on gold substrates was investigated with the core hole clock method. Electrons from the nitrogen K-shell are state-selectively excited into the two symmetry-split π^* orbitals of the cyano end group with x-ray photons of well-defined polarization. The charge transfer times from these temporarily occupied orbitals to the metal substrate differ significantly. Theoretical calculations show that these two π^* orbitals extend differently onto the alkane backbone and the anchoring sulfur atom, thus causing the observed dependence of the electron transfer dynamics on the symmetry of the orbital.

O 8.8 Mon 12:15 A 060

Trapping of image-potential resonances on free-electron-like metal surface — •Ulrich Höfer[1,2], Matthias Winter[1], Eugene V. Chulkov[2], and Pedro M. Echenique[2] — [1]Fachbereich Physik, Philipps-Universität Marburg, Germany — [2]Donostia International Physics Center (DIPC), San Sebastián, Spain

Image-potential states have been studied by two-photon photoemission (2PPE) for the surface of Al(100) where the whole series is energetically degenerate with free-electron-like bulk states. In contrast to expectations, the series of resonances is not smeared out to one broad structure as a result of a strong coupling to the bulk continuum. Instead, the first resonance (n=1) is found to be suppressed and the resonances with quantum numbers n=2,3,4,5 are resolved as individual peaks in the time-resolved spectra. Both effects are suggested to be a consequence of resonance trapping [1].

Here, we combine the multiple scattering approach to describe surface states with an open quantum system formalism that properly takes into account strong coupling of resonances to a continuum. The theory is able to quantitatively describe trapping of adjacent surface resonances for simple metals.

[1] M. Winter, E. V. Chulkov, U. Höfer, PRL **107**, 236801 (2011)

O 8.9 Mon 12:30 A 060

Time-resolved 2PPE study on NiO(001) thin films — •Mario Kiel, Stephan Grosser, and Wolf Widdra — Martin-Luther-Universität Halle-Wittenberg, 06120 Halle, Germany

The electronic structure of unoccupied states within ultra-thin films of the transition metal nickel oxide has been investigated by time-resolved two-photon photoemission (2PPE) in combination with scanning tunneling spectroscopy (STS). The thin films have been prepared under UHV conditions by molecular beam epitaxy in an oxygen atmosphere on a Ag(001) substrate at room temperature.

For the NiO bilayer a series of image-potential states has been identified. From time-resolved data lifetimes of 30, 50, and 120 fs have been determined for the n=1–3 image state, respectively. The observed binding energies can be well described on the basis of the dielectric properties of NiO. For the bilayer the permittivity is already close to the bulk like. Besides the image-potential states unoccupied Ni 3*d* states can be identified for the NiO bilayer as well as for thicker films in the 2PPE spectra. These states exhibit layer-dependent energy shifts which will be compared with layer-resolved STS data in the range from 2 to 8 monlolayers NiO. Additionally, 2PPE features from occupied NiO states have been identified due to their characteristic energy shift upon photon energy variation.

O 8.10 Mon 12:45 A 060

Hot carrier relaxation in HOPG probed at the H-point by means of time-resolved XUV photoemission — •Ankatrin Stange, Christian Sohrt, Timm Rohwer, Stefan Hellmann, Gerald Rohde, Kerstin Hanff, Lutz Kipp, Kai Rossnagel, and Michael Bauer — Institut für Experimentelle und Angewandte Physik, Universität Kiel, 24118 Kiel, Germany

Time- and angle-resolved XUV photoelectron spectroscopy (tr-ARPES) is applied to study the relaxation dynamics of excited hot carriers in HOPG and graphene. The XUV probe used in the experiment allows us to record electronic structure transients at large momentum values so that in particular the boundary of the first Brillouin zone becomes accessible. In the case of HOPG this capability provides a most direct access to the momentum region relevant for optical excitation and subsequent relaxation of the hot carriers. The results are discussed with respect to other time-resolved experiments which in the past were restricted to probes in the visible/NUV regime.