

O 75: Ultrafast Electron and Spin Dynamics

Time: Wednesday 15:00–18:45

Location: TRE Phy

O 75.1 Wed 15:00 TRE Phy

Ultrafast electronic band gap control in an excitonic insulator — ●SELENE MOR¹, MARC HERZOG², DENIS GOLEZ³, PHILIPP WERNER³, MARTIN ECKSTEIN⁴, CLAUDE MONNEY⁵, and JULIA STÄHLER¹ — ¹Fritz-Haber-Institut, Berlin, Germany — ²Potsdam Univ., Potsdam, Germany — ³Fribourg Univ., Fribourg, Switzerland — ⁴MPG for Structural Dynamics, Hamburg, Germany — ⁵Zurich Univ., Zurich, Switzerland

Ultrafast control of material properties is of both fundamental and technological interest. Here, we investigated the sub-picosecond dynamics of the electronic structure of Ta₂NiSe₅ by means of time- and angle-resolved photoelectron spectroscopy (trARPES). The system was proposed to support an excitonic insulator (EI) phase below $T_c \approx 328$ K. Such an EI phase is expected to occur in small gap semiconductors with strong electron-hole interaction as excitons can form spontaneously and condense into a ground state with a larger gap. trARPES below T_c reveals a strong excitation-density-dependent valence band depopulation, until absorption saturates at a critical fluence $F_C = 0.2$ mJ cm⁻². Below F_C the band gap shrinks transiently due to photoenhanced screening of Coulomb interaction, while it widens above F_C for and relaxes after ≈ 1.5 ps. Hartree-Fock calculations reveal that the band gap widening can be explained by photoenhancement of the density of the exciton condensate, an effect that persists until the system undergoes interband relaxation. These results prove the possibility to manipulate condensates of excitons with light and to gain ultrafast control of the size of semiconductor band gaps.

O 75.2 Wed 15:15 TRE Phy

Ultrafast photoinduced valence transition in the heavy-fermion compound YbInCu₄ — ●LARS-PHILIP OLOFF¹, LUKAS WENTHAUS², KERSTIN HANFF¹, GIUSEPPE MERCURIO³, MICHAEL BAUER¹, WILFRIED WURTH^{2,3}, and KAI ROSSNAGEL¹ — ¹Institut für Experimentelle und Angewandte Physik, Universität Kiel, 24098 Kiel, Germany — ²DESY Photon Science, 22607 Hamburg, Germany — ³Department of Physics and Center for Free-Electron Laser Science CFEL, University of Hamburg, 22761 Hamburg, Germany

Upon cooling to a temperature below 42 K, the heavy-fermion compound YbInCu₄ undergoes an isostructural transition from a semimetallic, paramagnetic phase with an Yb valence of +3 to a metallic, non-magnetic phase with an Yb valence of +2.85. Here, we combine HHG and FEL-based time-resolved photoemission spectroscopy techniques to gain novel insights into the valence transition in the (sub-)surface region. Through the combination of time-resolved ARPES and time-resolved XPS, we are able to directly track different characteristic spectroscopic signatures of the photoinduced valence change in the (transient) valence and core electronic structure on the femtosecond to picosecond timescale. Our results constitute the first *direct* experimental observation of an ultrafast photoinduced valence change in a heavy-fermion compound.

O 75.3 Wed 15:30 TRE Phy

Ultrafast dynamics in Fe-pnictides analyzed by femtosecond time-resolved x-ray spectroscopy — ●ANDREA ESCHENLOHR¹, MOHAMMADMAHDI AFSHARI¹, KLAUS SOKOLOWSKI-TINTEN¹, NIKO PONTIUS², HIDENORI HIRAMATSU³, HIDEO HOSONO³, JÖRG FINK⁴, and UWE BOVENSIEPEN¹ — ¹Univ. Duisburg-Essen — ²Helmholtz Zentrum Berlin — ³Tokyo Institute of Technology — ⁴IFW Dresden

The electronic properties of Fe-pnictide high temperature superconductors are strongly influenced by the Fe-As distance, which is linked to the superconducting critical temperature [1] and can be modified not only through doping and external pressure, but also transiently by the coherent A_{1g} phonon mode [2]. Element- and orbital-specific x-ray absorption spectroscopy (XAS) provides information on the local electronic structure. By employing femtosecond (fs) x-ray pulses from the BESSY II Femtoslicing source, we perform fs time-resolved XAS of BaFe₂(AsP)₂. We observe pump-induced changes at the Fe L₃ edge that are described by the interplay of a transient decrease in absorption, shift in binding energy and peak broadening. This complex behavior will be discussed in the context of electronic and phononic excitations in the Fe-pnictides, in particular photodoping and related changes of the transient chemical potential [3].

We acknowledge funding by the DFG through SPP 1458 and Sfb

1242, and the BMBF through FEMTOSPEX.

[1] Mizuguchi et al., Supercond. Sci. Technol. **23**, 054013 (2010). [2] Rettig et al., PRL **114**, 067402 (2015); Gerber et al., Nat. Commun. **6**, 7377 (2015). [3] Yang et al., PRL **112**, 207001 (2014).

O 75.4 Wed 15:45 TRE Phy

Polarization control of ultrafast photocurrents in the Dirac cone of the topological insulator Sb₂Te₃ — JOHANNES REIMANN¹, KENTA KURODA^{1,2}, ●JENS GÜDDE¹, and ULRICH HÖFER¹ — ¹Fachbereich Physik und Zentrum für Materialwissenschaften, Philipps-Universität, 35032 Marburg — ²Institute for Solid State Physics (ISSP), University of Tokyo, 277-8581 Chiba, Japan

We present energy-momentum mapping of the photocurrent in the Dirac cone of the topological insulator Sb₂Te₃ by means of time- and angle-resolved two-photon photoemission (2PPE) after optical excitation with ultrashort linear and circular polarized mid-infrared laser pulses. Recently, we have demonstrated that linear polarized mid-infrared pulses permit the generation of photocurrents with ps-lifetime in the initially unoccupied part of the Dirac cone of Sb₂Te₃ by a direct optical excitation [1]. Here, we show that the magnitude and direction of this current depend on the azimuthal orientation of the sample and reflect the threefold symmetry of the surface. For orientations where no photocurrent is generated by linear polarized light, we are instead able to generate a photocurrent by circular polarized light and fully control its direction and magnitude by varying the light helicity.

[1] K. Kuroda *et al.*, Phys. Rev. Lett. **116**, 076801 (2016).

O 75.5 Wed 16:00 TRE Phy

On the origin of photocurrents in the topological insulator Bi₂Se₃ — ●SOPHIA KETTERL¹, SEBASTIAN OTTO², MARTIN BASTIAN¹, CORNELIUS GAHL¹, BEATRICE ANDRES¹, JAN MINAR³, HUBERT EBERT³, THOMAS FAUSTER², JÜRGEN BRAUN³, and MARTIN WEINELT¹ — ¹Freie Universität Berlin, Arnimallee 14, 14195 Berlin — ²Friedrich-Alexander-Universität Erlangen-Nürnberg, Staudtstr. 7, 91058 Erlangen — ³Ludwig-Maximilians-Universität München, Butenandtstr. 5-13, 81377 München

Topological insulators (TIs) host metallic topological surface states (TSS) with helical spin structure. This makes them promising materials for the generation of spin-polarized currents. The TI Bi₂Se₃ is intrinsically n-doped and thus the TSS at the Γ point with a Dirac cone (DC) dispersion is occupied. Two-photon photoemission (2PPE) experiments have shown that Bi₂Se₃ exhibits a second, unoccupied DC in the band gap between the second and third conduction band [1].

We studied the excitation of photocurrents with circularly polarized light in the second DC with 2PPE and one-step photoemission calculations. The second DC can be populated from the first DC. We find dichroic photoemission for circularly polarized pump pulses, both at resonant and off-resonant excitation. Only for excitation out of the occupied conduction band, we observe an asymmetric electron population indicating a photocurrent. Thus our results question the topological origin of photocurrents in Bi₂Se₃ for near-IR excitation [2]. [1] D. Niesner *et al.*, Phys. Rev. B **86**, 205403 (2012). [2] C. Kastl *et al.*, Nat. Commun. **6**, 6617 (2015).

O 75.6 Wed 16:15 TRE Phy

Spin and Valley Dynamics of Free Carriers in Epitaxial Single-Layer WS₂ — ●ANTONIJA GRUBISIC-CABO¹, SOREN ULSTRUP², DEEPNARAYAN BISWAS³, JONATHAN M. RILEY³, MACIEJ DENDZIK¹, CHARLOTTE E. SANDERS¹, MARCO BIANCHI¹, CEPHISE CACHO⁴, DAN MATSELYUKH⁴, RICHARD T. CHAPMAN⁴, EMMA SPRINGATE⁴, PHIL D. C. KING³, JILL A. MIWA¹, and PHILIP HOFMANN¹ — ¹Aarhus University, DK — ²ALS Berkeley, USA — ³University of St. Andrews, UK — ⁴CLF, STFC Rutherford Appleton Laboratory, UK

The semiconducting single-layer (SL) transitional metal dichalcogenides (TMDCs) have been identified as ideal materials for accessing and manipulating spin- and valley-degrees of freedom. The current understanding of the valley properties and excited carrier dynamics relies largely on a photoluminescence and differential absorption measurements. Since excitons dominate the optical response of SL TMDCs,

studies of free carriers can only be made indirectly. Here, we use time-resolved ARPES to directly measure free carriers in epitaxial SL WS₂ grown on Ag(111). We observe that the optically generated free carrier density in a single valley can be increased by a factor of 2 using circularly polarized light. Moreover, by varying the photon energy of the excitation, we can tune the free carrier density in a given spin-split band. This allows us to selectively excite free electron-hole pairs with a given spin and within a single valley.

O 75.7 Wed 16:30 TRE Phy

Resolving initial stages of carrier thermalization in graphite using time-resolved ARPES with sub-15 fs temporal and near Fourier-limited spectral resolution — ●GERALD ROHDE¹, ANKATRIN STANGE¹, ARNE MÜLLER¹, MARCEL BEHRENDT¹, KERSTIN HANFF¹, LARS-PHILIP OLOFF¹, LEXIAN YANG², PETRA HEIN¹, KAI ROSSNAGEL¹, and MICHAEL BAUER¹ — ¹Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Germany — ²Department of Physics, Tsinghua University, Beijing, People's Republic of China

Carrier dynamics in graphitic materials have been subject of theoretical and experimental investigations in recent years. State-of-the-art experimental approaches address the time scales of excitation and relaxation of hot carrier distributions, aiming to break their evolutions down into the underlying fundamental scattering processes as ultimately governed by electron-electron and electron-phonon interaction. Using time- and angle-resolved photoelectron spectroscopy with sub-15 fs temporal and near Fourier-limited spectral resolution, we report here on the build-up of an out-of-equilibrium electron distribution in graphite within 13 fs and its further evolution during the thermalization towards a Fermi-Dirac distribution. We discuss potential processes causing this transition and compare the results to corresponding data reported for graphene [1]. Moreover, indications for a momentum redistribution of the hot carriers on ultrafast time scales are presented following a momentum-selective photoexcitation.

[1] I. Gierz *et al.*, Phys. Rev. Lett. **115**, 086803 (2015).

O 75.8 Wed 16:45 TRE Phy

Recent developments in the one-step description of time-resolved photoemission — ●JÜRGEN BRAUN¹, MICHAEL POTTHOFF², and HUBERT EBERT¹ — ¹Dept. Chemie, LMU Universität München, Germany — ²I. Institut für Theoretische Physik, Universität Hamburg, Germany

Recently a theoretical frame for the description of pump-probe photoemission has been developed. The approach is based on a general formulation using the Keldysh formalism for the lesser Green function to describe the real-time evolution of the electronic degrees of freedom in the initial state after a strong pump pulse that drives a system out of equilibrium [1]. As a first application, the theoretical description of two-photon photoemission (2PPE) for Ag(100) within the SPR-KKR-approach is briefly reviewed [2]. This formalism has now been generalized to the ferromagnetic case. First examples of fully spin-polarized 2PPE calculations on Fe(110) will be presented. Finally, a new approach is sketched that allows to account also for dynamical correlations in the non-equilibrium state caused by the pump pulse by means of a corresponding two-time dependent self-energy $\Sigma(z, z')$.

[1] J. Braun, R. Rausch, M. Potthoff, J. Minar, and H. Ebert, Pump-probe theory of angle-resolved photoemission, Phys. Rev. B **91**, 035119 (2015)

[2] J. Braun, R. Rausch, M. Potthoff, and H. Ebert, One-step theory of two-photon photoemission, Phys. Rev. B **94**, 125128 (2016)

O 75.9 Wed 17:00 TRE Phy

Interplay between metal-organic interface state and donor-acceptor interface mediated charge separation on PTCDA/TiOPc/Ag(111) — ●ALEXANDER LERCH¹, FREDERIK SCHILLER^{1,2}, and ULRICH HÖFER¹ — ¹Fachbereich Physik und Zentrum für Materialwissenschaften, Philipps-Universität, D-35032 Marburg — ²Centro de Física de Materiales, San Sebastian, Spain

Charge separation of excitons in the well ordered model system PTCDA on titanyl phthalocyanine (TiOPc) on Ag(111) was investigated by means of time-resolved two-photon photoemission (2PPE). An optical parametric oscillator provided pump pulses with photon energies ranging from 1.6 to 2.5 eV. This allowed for selective excitation of TiOPc or PTCDA. Between TiOPc and Ag(111) a shockley derived interface state (IS) approximately 0.3 eV above the Fermi energy forms. We observe a strong coupling of the first molecular layers to the IS

and determine transfer times into the IS ranging from approximately 20 fs out of the second to 160 fs out of the third and fourth molecular layers. Systematic thickness-dependent measurements quantify the distance dependence of this coupling. Whereas the IS dominates the dynamics in the second layer, the PTCDA/TiOPc acceptor-donor interface (D/A) competes for electrons from excitons in the third to fourth layer. In the fifth to sixth layer no significant IS assisted separation on a timescale below 12 ns can be detected any longer. Our results highlight the competition between IS and D/A mediated charge separation of excitons in few-layer organic systems on metal substrates.

O 75.10 Wed 17:15 TRE Phy

Ultrafast electron dynamics and small polaron formation in solid DMSO on Cu(111) — ●SARAH KING, KATHARINA BROCH, and JULIA STÄHLER — Department of Physical Chemistry, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

The ultrafast electron solvation dynamics in DMSO have been investigated using time- and angle-resolved two-photon photoemission spectroscopy of condensed DMSO films on a Cu(111) single crystal with different coverages, between 1 and 10 monolayer (ML) equivalents. At DMSO coverages higher than 1 ML, initially a delocalized electronic state is formed in DMSO at 2.85 ± 0.05 eV relative to the Fermi level, with an effective mass of approximately 1.8 times the mass of a free electron. This state simultaneously undergoes localization and energetic stabilization where the peak energy shifts to lower energies at a rate of approximately 1 eV/ps as the peak dispersion flattens. By 200 fs after formation, this state exhibits negative dispersion and tenfold slower peak shifting before decaying completely within 1 ps. We preliminarily attribute the dynamics in peak energy and localization to small polaron formation in DMSO. The decay-time of the small polaron state is concomitant with the rise-time in a DMSO/vacuum surface state at 2.30 ± 0.05 eV, which has an extremely long decay time of *seconds* and exhibits reactivity with oxygen. As DMSO is a common electrolyte, our results on non-equilibrium electronic states in DMSO are important for understanding electrochemistry.

O 75.11 Wed 17:30 TRE Phy

Optically induced quasi-particle dynamics and transient polarization of a C₆₀ film — ●SEBASTIAN EMMERICH¹, BENJAMIN STADTMÜLLER¹, DOMINIK JUNGKERN¹, NORMAN HAAG¹, CHRISTINA SCHOTT¹, STEFFEN EICH¹, MARKUS ROLLINGER¹, MAHALINGAM MANIRAJ¹, MARTIN AESCHLIMANN^{1,3}, MIRKO CINCHETTI^{1,3}, and STEFAN MATHIAS² — ¹University of Kaiserslautern and Research Center OPTIMAS, Erwin-Schrödinger-Str. 46, 67663 Kaiserslautern, Germany — ²Physikalisches Institut I, Universität Göttingen, 37077 Göttingen, Germany — ³Experimentelle Physik VI, Technische Universität Dortmund, 44221 Dortmund, Germany

The performance of organic based electronic devices is determined by the energy level alignment as well as by the excitation dynamics of electrons in these materials. Although both aspects are intrinsically linked by the polarizability of organic materials, they have mainly been discussed separately so far. In this work, we present new insight into the ultrafast dynamics of thin C₆₀ films on Ag(111). The combination of time- and angle resolved photoemission with a fs-XUV light source allows us to follow the transient evolution of the unoccupied as well as of the occupied band structure after the fs-optical excitation with visible light. The unoccupied part of the C₆₀ band structure reveals the well-known quasi-particle dynamics of the exciton formation and its decay in C₆₀. Most interestingly, we observe a transient change of the linewidth of the occupied molecular orbitals upon optical excitation. This effect is attributed to the transient polarization of the molecular film due to the exciton formation in distinct C₆₀ sites.

O 75.12 Wed 17:45 TRE Phy

Vibrational relaxation and migration dynamics of excitons in sexithiophene/Au(111) — WIBKE BRONSH¹, SEBASTIAN BAUM¹, KRISTOF ZIELKE¹, MALTE L. WANSLEBEN², MARTIN WEINELT¹, and ●CORNELIUS GAHL¹ — ¹Freie Universität Berlin, Institut für Experimentalphysik, Arnimallee 14, 14195 Berlin — ²Physikalisch-Technische Bundesanstalt, Abbestr. 2-12 10587 Berlin

The primary optical excitations in organic semiconductors are excitons. The corresponding absorption bands are commonly strongly broadened due to coupling to vibrational excitations, as known from optical spectroscopy. In organic electronics, the dynamics of such excitonic states concerning their energetic evolution as well as their migration through the material are decisive for the performance of a device. We

investigated the exciton dynamics by time-resolved two-photon photoemission (2PPE) spectroscopy in the model system of sexithiophene (6T) on the Au(111) surface. In crystalline 6T films, vibrational excess energy is dissipated on a time scale of <100 fs resulting in an excitonic state which is stable in energy for >100 ps. In amorphous 6T films, however, the energetic relaxation from a broad initial distribution of excited states is strongly slowed down and reaches an excitonic state with a binding energy comparable to that in the crystalline film on a time scale >10 ps. This slow process is attributed to migration of excitons towards sites with higher binding energy. Thus the morphology of the organic layer strongly influences the excited state dynamics and energetics relevant for organic electronics.

O 75.13 Wed 18:00 TRE Phy

Femtosecond electron dynamics in the momentum space

— ●FLORIAN HAAG¹, JOHANNES SEIDEL¹, TOBIAS EUL¹, NORMAN HAAG¹, MIRKO CINCHETTI², MARTIN AESCHLIMANN¹ und BENJAMIN STADTMÜLLER¹ — ¹Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, Erwin-Schroedinger-Strasse 46, 67663 Kaiserslautern, Germany — ²Experimentelle Physik VI, Technische Universität Dortmund, 44221 Dortmund, Germany

Excited electrons play a crucial role for many fundamental chemical and physical phenomena occurring at surfaces, (hybrid) interfaces, or in bulk materials. New insight into this field can be obtained by the combination of time-resolved two-photon photoemission spectroscopy with momentum microscopy. This novel approach of time-resolved 2 photon momentum microscopy offers the possibilities to follow the electron dynamics in the accessible momentum space and hence to determine momentum resolved lifetimes of excited electrons.

As most fundamental examples, we have studied the momentum resolved electronic lifetime of polycrystalline and single crystalline metal crystals. Using various combinations of light polarization for the pump and probe pulse allows us to distinguish the electron dynamics of bulk and surface bands. Our results will significantly contribute to a better understanding of momentum space dynamics of excited electrons and of inelastic und quasi-elastic scattering phenomena at interfaces and in solids.

O 75.14 Wed 18:15 TRE Phy

Spatio-Temporal Probing of Lattice Dynamics in Graphite

by **Ultrafast TEM** — ●ARMIN FEIST¹, NARA RUBIANO DA SILVA¹, WENXI LIANG², CLAUS ROPERS¹, and SASCHA SCHÄFER¹ — ¹IV. Physical Institute - Solids and Nanostructures, Göttingen, Germany — ²Huazhong University of Science and Technology, Wuhan, China

Over the past decades, ultrafast techniques have shaped a rich pic-

ture of material dynamics in spatially homogenous systems. Extending these concepts, ultrafast transmission electron microscopy (UTEM) provides access to nanoscale processes on their intrinsic femtosecond time and nanometer length scales [1,2].

Here, we apply UTEM to investigate ultrafast nanoscale lattice dynamics localized at the edge of a single-crystalline graphite thin film [3]. Time-dependent convergent beam electron diffraction (CBED) of nanoscale sample areas gives access to the complex distortions of the crystal structure. Specifically, the initial optically generated, spatially-inhomogeneous strain distribution relaxes via coherent out-of-plane breathing and in-plane shear motion coupled to a propagating compressive strain wave. We spatio-temporally map these dynamics with 30 nm spatial and sub-picosecond temporal resolution. By employing UTEM, energy transfer and dissipation in complex nanostructured systems can be tracked over a wide range of time scales, spanning from femtosecond optical excitations to coherent lattice vibrations with nanosecond lifetimes.

[1] A. H. Zewail, *Science*, **328**, 187 (2010). [2] A. Feist *et al.*, arXiv: 1611.05022. [3] A. Feist *et al.*, in preparation.

O 75.15 Wed 18:30 TRE Phy

Lattice Response of Pb/Si(111) Layers and Islands upon Femtosecond Optical Excitation

— ●TOBIAS WITTE, TIM FRIGGE, BERND HAFKE, BORIS KRENZER, and MICHAEL HORN-VON HOEGEN — University of Duisburg-Essen, Lotharstr. 1, 47048 Duisburg, Germany

Employing time resolved reflection high energy electron diffraction (tr-RHEED) the non-equilibrium dynamics of the phonon system after optical excitation of ultrathin Pb films and islands on Si(111) has been investigated. The sample is pumped by fs-IR-laserpulses, while the transient response is probed by a 30 keV electron pulse. Surface sensitivity is achieved via grazing incidence of the high energy electrons. fs-laserpulses cause impulsive excitation of the electron system of the Pb films and islands, respectively. The hot electron system thermalizes and heats the lattice system by electron-phonon coupling. The thermal motion of the (surface-) atoms is observed through the transient Debye-Waller effect. As Pb is known to exhibit strong electron-phonon coupling in the bulk the time constant for the energy transfer to the lattice system is - in the framework of the two temperature model (TTM) - expected to be in the sub-ps regime. In contrast, a rather slow time constant of $\tau = 3.2$ ps was observed at the surface. Moreover, incoherent lattice excitation of the islands occurs even slower at a timeconstant of $\tau = 4.6$ ps. These results demonstrate how low-dimensionality and quantum-size effects reduce the energy flow mediated by electron-phonon coupling.