O 8: Ultrafast Electron Dynamics at Surface and Interfaces I

Time: Monday 10:30–12:45 Location: TRE Phy

O 8.1 Mon 10:30 TRE Phy

Coulomb-correlated few-electron states from a laser-driven Schottky emitter — •Rudolf Haindl[1,2], Armin Feist[1,2], Till Domröse[1,2], Marcel Möller[1,2], Sergey V. Yalunin[1,2], and Claus Ropers[1,2] — [1]Max Planck Institute for Multidisciplinary Sciences, Göttingen — [2]4th Physical Institute, University of Göttingen

Observing correlation phenomena for free electrons requires a highly confined emission source, readily available by femtosecond photoemission from nanotips [1,2], and can in principle be studied using the supreme beam control provided by electron microscopes [3]. However, ensemble-averaged detection usually prevents studying multi-particle correlations in free-electron beams.

In this work, we use event-based electron microscopy to characterize laser-triggered few-electron pulses generated at a Schottky field emitter [4]. We find surprisingly strong energy correlations of about 2 eV in the two- and three-electron states hinting at a correlated emission process. Furthermore, state-sorted beam caustics show characteristic transverse pair distributions, as well as an increase and a longitudinal shift of the source.

Inducing strong few-electron Coulomb correlations facilitates non-Poissonian electron pulse statistics, promising applications in free-electron quantum optics.

[1] P. Hommelhoff, et al., Phys. Rev. Lett. 96, 077401 (2006).
[2] C. Ropers, et al., Phys. Rev. Lett. 98, 043907 (2007).
[3] A. Feist, et al., Ultramicroscopy 176, 63-73 (2017).
[4] R. Haindl, et al., arXiv:2209.12300 (2022).

O 8.2 Mon 10:45 TRE Phy

Impact of layer thickness on the anisotropic carrier dynamics of a laser-excited $Fe_n/(MgO)_m(001)$ heterostructure from real-time TDDFT — •Elaheh Shomali, Markus Ernst Gruner, and Rossitza Pentcheva — Department of Physics and Center for Nanointegration, CENIDE, University of Duisburg-Essen, Germany

The carrier dynamics of $Fe_n/(MgO)_m(001)$ metal/insulator heterostructures has been investigated from first-principles as a function of the Fe and MgO layer thickness (n=1,3,5 and m=3,5,7), excitation energy and polarization direction. The imaginary part of the dielectric tensor calculated within the random phase approximation (RPA) shows for all structures a similar metallic-like behavior in the in-plane component, $\epsilon_{xx}(\omega)$, whereas for $\epsilon_{zz}(\omega)$ the initially low response below the MgO band gap for $Fe_1/(MgO)_3(001)$ increases substantially with the number of Fe layers. Electronic excitations calculated in the framework of RT-TDDFT confirm the concerted excitation mechanism previously proposed for $Fe_1/(MgO)_3(001)$ also for the larger systems, which involves two simultaneous excitations via interface states [1]: one from occupied states of the metal to the conduction band of the insulator and simultaneously, another from the valence band of MgO into Fe states above the Fermi level. This mechanism allows for an effective bidirectional relocation of excitations between the metallic and insulating subsystems even for photon energies below the MgO gap. Funding by DFG via SFB 1242, project C02 is gratefully acknowledged. [1] E. Shomali, M. E. Gruner, and R. Pentcheva, Phys. Rev. B **105**, 245103 (2022).

O 8.3 Mon 11:00 TRE Phy

Relaxation and transport dynamics of laser-excited electrons — •Markus Heckschen[1], Yasin Beyazit[1], Florian Kühne[1], Elaheh Shomali[1], Markus Gruner[1], Rossitza Pentcheva[1], Ping Zhou[1], Detlef Diesing[2], Axel Lorke[1], Uwe Bovensiepen[1], and Björn Sothmann[1] — [1]Faculty of Physics and Center for Nanointegration (CENIDE), University of Duisburg-Essen, Lotharstraße 1, 47057 Duisburg, Germany — [2]Faculty of Chemistry, University of Duisburg-Essen, Universitätsstraße 5, 45141 Essen, Germany

Scattering of electronic excitations after laser excitation is of key importance to understand transport and relaxation effects. A recent back-pump front-probe two-photon-photoemission experiment in Fe/Au samples measured the electron distribution as a function time and energy [1]. The time at which the maxima of the electron distribution occur as a function of Au-layer thickness provides information on velocity, transport regime and relaxation processes. Here, we present a trajectory-based simulation approach to describe the energy relaxation and the transport through the sample in the ballistic regime. Our results agree qualitatively with the experiment and provide new insight into ultrafast electron dynamics.

[1] Beyazit et al., PRL **125**, 076803(2020)

O 8.4 Mon 11:15 TRE Phy

Capturing non-thermal electrons: To what extend can a temperature-based model compete with a kinetic description? — •Markus Uehlein, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, RPTU, Kaiserslautern, Germany

The development and usage of laser systems in research allows to study the dynamics of laser-excited electrons in metals on sub-picosecond timescales. Kinetic models can trace non-equilibrium distributions in great detail for instance by using full Boltzmann collision integrals [1]. Due to the excitation of non-thermal electrons, this dynamics cannot be described with purely temperature-based models. However, there are a number of extensions of the well known two-temperature model to trace the non-thermal electrons in a numerically simplified way.
We aim to clarify, whether an extended TTM (first published in Refs. [2, 3], improved in Ref. [4]) can describe this electronic non-equilibrium dynamics by comparing it to the kinetic description. In particular, we compare the spectral particle dynamics to a time-resolved two-photon photoemission measurement [5].

[1] B. Y. Mueller and B. Rethfeld; Phys. Rev. B **87**, 035139 (2013)
[2] E. Carpene; Phys. Rev. B **74**, 024301 (2006)
[3] G. D. Tsibidis; Appl. Phys. A **124**, 311 (2018)
[4] M. Uehlein, S. T. Weber and B. Rethfeld; Nanomaterials **12**, 1655 (2022)
[5] Y. Beyazit *et al.*; Phys. Rev. Lett. **125**, 076803 (2020)

O 8.5 Mon 11:30 TRE Phy

Radio-frequency controlled electron pulses for time-resolved LEED — •Dennis Epp[1], Benjamin Schröder[1], Marcel Möller[1], and Claus Ropers[1,2] — [1]Max-Planck-Institut für Multidisziplinäre Naturwissenschaften — [2]Georg-August-Universität Göttingen

In this contribution, we demonstrate temporal compression of low-energy electron pulses using rf-fields. Specifically, we combine a millimetre-sized electron gun [1] with a rf-cavity driven by a sinusoidal wave. A phase-locked loop synthesizer controls the timing between the incident electron pulses and the sinusoidal signal. By electron energy filtering, we measured an initial energy spread of ΔE*1.5 eV for the uncompressed pulses with mean kinetic energies in the range of 50 eV to 100 eV [2]. In perspective, ULEED with sub-ps resolution will allow for the observation of the impact of electron-phonon coupling and coherent vibrational motion on the efficiencies of surface phase transitions and the resulting changes in materials functionality. [1] Vogelgesang, et al., Nature Physics 14,184-190 (2018). [2] Epp et al., in preparation

O 8.6 Mon 11:45 TRE Phy

High-harmonic generation in topological insulator surface states using wave-packet approaches — •Vanessa Junk[1], Wolfgang Hogger[1], Alexander Riedel[1], Cosimo Gorini[2], and Klaus Richter[1] — [1]Institute for theoretical physics, University of Regensburg, Germany — [2]Université Paris-Saclay, CEA, CNRS, SPEC, 91191, Gif-sur-Yvette, France

The interaction of matter with strong-field light gives rise to highly non-linear electron dynamics within the material. The electric field of the light accelerates electrons through the band structure and drives non-perturbative transitions, leading to strongly anharmonic electron velocities and the emission of high-order harmonics.

Here, we want to study these electron dynamics for the surface states of topological insulators by solving the time-dependent Schrödinger equation for wave packets. That the separation of bulk and surface high harmonics is experimentally possible has been demonstrated recently [1]. Furthermore, an alternating polarization rotation between odd and even order harmonics was found and attributed to the hexagonal warping present in Bi_2Te_3 surface states. Using effective Dirac models and taking into account effects of the Fermi sea, we can reproduce these experimental features with our wave-packet approach.

[1] C. Schmid, L. Weigl, P. Grössing, V. Junk, C. Gorini, S. Schlauderer, S. Ito, M. Meierhofer, N. Hofmann, D. Afanasiev, J. Crewse, K.

Kokh, O. Tereshchenko, J. Güdde, F. Evers, J. Wilhelm, K. Richter, U. Höfer and R. Huber, Tunable non-integer high-harmonic generation in a topological insulator, Nature **593**, 385-390 (2021)

O 8.7 Mon 12:00 TRE Phy

Subcycle surface-bulk coupling in a topological insulator — •Suguru Ito[1], Michael Schüler[2], Manuel Meierhofer[3], Josef Freudenstein[3], Dmytro Afanasiev[3], Jens Güdde[1], Michael Sentef[4], Rupert Huber[3], and Ulrich Höfer[1] — [1]Philipps-Universität Marburg, Germany — [2]Paul Scherrer Institute, Switzerland — [3]Universität Regensburg, Germany — [4]Max Planck Institute for the Structure and Dynamics of Matter, Germany

Intense lightwaves at terahertz frequencies can drive currents in electronic bands as well as non-perturbative excitations across the band gap, whose interplay leads to strong-field phenomena such as high-harmonic generation. Angle-resolved photoemission spectroscopy (ARPES) with subcycle time resolution allows us to observe such lightwave-driven processes in momentum space.

Here, we will present our investigation of subcycle coupling between the topological surface state (TSS) and the bulk conduction band (BCB) in Bi_2Te_3, where the TSS is well seperated from the bulk along the $\bar{\Gamma}$-$\bar{K}$ direction. Under exposure of a strong 25-THz lightwave, however, transient replicas of the equilibrium TSS, Floquet-Bloch states, are formed. Their hybridization with the BCB enables an efficient population transfer into the BCB. In contrast, TSS and BCB are less separated along $\bar{\Gamma}$-$\bar{M}$, where already an intraband current can induce an efficient subcycle population transfer. Both distinct mechanisms are readily seen in our experiment. The dynamical Floquet-bulk coupling is confirmed by our full quantum calculations and provides a momentum-resolved view of non-perturbative interband excitations.

O 8.8 Mon 12:15 TRE Phy

Ultrafast electron dynamics in a topological crystalline insulator — •Magnus Berntsen[1], Yuzhu Fan[1], Antonija Grubisic-Cabo[1,2], Maciej Dendzik[1], Qinda Guo[1], Cong Li[1], Yang Wang[1], Wanyu Chen[1], Dibya Phuyal[1], Jonas Weissenrieder[1], and Oscar Tjernberg[1] — [1]Department of Applied Physics, KTH Royal Institute of Technology, Sweden — [2]University of Groningen, Netherlands

Topological crystalline insulators is a class of materials that possesses helical Dirac-like electronic surface states whose existence is protected by crystal symmetries. This intricate coupling of topology and crystal structure makes it particularly interesting to study the transient electronic structure in these materials induced by an optical excitation since an accompanying symmetry-breaking lattice deformation could lead to a break-down of the topological phase on an ultrafast time scale. Here, we have studied the ultrafast electron dynamics in the archetypal topological crystalline insulator (Pb,Sn)Se by time- and angle-resolved photoemission spectroscopy. The electronic excitation occurs mainly along a preferential direction in momentum space determined by the polarization of the pump pulse and the excitation spectrum displays multiple time scales suggestive of the existence of multiple relaxation channels. Supported by time-resolved diffraction data acquired by ultrafast electron microscopy, we discuss our findings both in the light of the specific electronic structure of the surface states as well as the potential influence of lattice dynamics on the decay of the electronic excitation.

O 8.9 Mon 12:30 TRE Phy

Field-induced ultrafast modulation of Rashba coupling in ferroelectric α-GeTe(111) — Geoffroy Kremer[1,2], Julian Maklar[3], Laurent Nicolaï[4], •Chris W. Nicholson[1,3,5], Changming Yue[1], Caio Silva[3], Philipp Werner[1], J. Hugo Dil[6,7], Juraj Krempaský[6], Gunther Springholz[8], Ralph Ernstorfer[3,9], Jan Minár[4], Laurenz Rettig[3], and Claude Monney[1] — [1]Université de Fribourg, Switzerland — [2]Université de Lorraine, France — [3]Fritz Haber Institute of the Max Planck Society, Germany — [4]University of West Bohemia, Czech Republic — [5]SPECS Surface Nano Analysis GmbH, Germany — [6]Paul Scherrer Institut, Switzerland — [7]EPFL, Lausanne, Switzerland — [8]Johannes Kepler Universität, Austria — [9]Technische Universität Berlin, Germany

Rashba materials provide an ideal playground for spin-to-charge conversion in prototype devices. I will present our recent time- and angle-resolved photoemission spectroscopy and momentum microscopy results on α-GeTe(111). This is a non-centrosymmetric ferroelectric (FE) semiconductor displaying a giant FE distortion below 700 K, which hosts spin-polarized states that are promising for a new type of highly efficient non-volatile memory device based on switchable polarization. We find a transient modification of the bulk Rashba splitting corresponding to an enhancement of the FE lattice distortion. This enhanced response results from a transient surface photovoltage which induces a delayed displacive excitation of a phonon along the FE distortion direction [1].

[1] Kremer et al, *Nature Communications* 13, 6396 (2022)