O 23: Ultrafast Electron Dynamics at Surfaces and Interfaces III

Time: Tuesday 10:30–13:15 Location: MA 041

O 23.1 Tue 10:30 MA 041

Emergence of Floquet band structure in Dirac Hamiltonians by short pulse irradiation — •Yuriko Baba[1], Alejandro Sebastián Gómez[2], Vanessa Junk[3], Francisco Domínguez-Adame[4], Rafael A. Molina[2], and Klaus Richter[3] — [1]Universidad Autónoma de Madrid, Madrid, Spain — [2]Instituto de Estructura de la Materia, IEM-CSIC, Madrid, Spain — [3]Fakultät für Physik, Universität Regensburg, Germany — [4]GISC, Universidad Complutense de Madrid, Madrid, Spain

Floquet theory is a well-established approach to describe time-dependent quantum systems driven by a periodic external field. In the presence of such a driving, the spectrum of the system is described by replicas of the original dispersion, shifted by integers of the driving frequency [1]. Using the Floquet formalism, this work studies the emergence of the Floquet structure in the energy spectrum of Dirac Hamiltonians subject to short pulses in order to understand the limits of this formalism as well as to interpret recent results of subcycle lightwave-ARPES [2].

[1] V. Junk, P. Reck, C. Gorini, K. Richter, Physical Review B, 101, 134302 (2020). [2] S. Ito, et al, Nature, 616, 696 (2023).

O 23.2 Tue 10:45 MA 041

The challenge of realizing Floquet effects in epitaxial graphene — •Leonard Weigl[1], Niklas Hofmann[1], Johannes Gradl[1], Peter Richter[2], Thomas Seyller[2], and Isabella Gierz[1] — [1]University of Regensburg — [2]Technische Universität Chemnitz

Floquet engineering where the band structure of a solid is modified by periodic driving with strong laser pulses is a promising method for tailoring the electronic properties of solids on ultrafast time scales. One particularly tempting proposal is the opening of a topologically non-trivial gap in the Dirac cone of graphene with circularly polarized light [1]. The size of the dynamical gap is predicted to scale linearly with the inverse driving frequency [1]. On the other hand, charge carriers inside the Dirac cone need to be able to follow the driving field coherently over many cycles, which requires driving frequencies that are large compared to the inverse scattering time of the carriers [2]. Broers et al. [3] recently predicted a sweet spot for the driving frequency around 30 THz. We implemented a setup for time- and angle-resolved photoemission spectroscopy that combines a strong-field variable-wavelength multi-THz pump source with extreme ultraviolet probe pulses for putting these predictions to the test. In this talk we present our progress and discuss remaining challenges towards the experimental observation of Floquet effects in epitaxial graphene.

[1] T. Oka and H. Aoki, Phys. Rev. B 79, 081406(R) (2009) [2] Dunlap et al., Phys. Rev. B 34, 3625 (1986) [3] L. Broers and L. Mathey, Phys. Rev. Research. 4, 013057 (2022)

O 23.3 Tue 11:00 MA 041

Subcycle time-resolved THz-ARPES of Bi_2Te_3 in two-dimensional momentum space — •Tim Bergmeier, Suguru Ito, Jens Güdde, and Ulrich Höfer — Fachbereich Physik, Philipps-Universität Marburg

Angle-resolved photoemission spectroscopy (ARPES) combined with Terahertz (THz) excitation and subcycle temporal resolution offers unique capabilities for investigating the ultrafast electron dynamics of Dirac currents in the surface band of topological insulators [1]. At field strengths that are strong enough for high-harmonic generation [2], it has recently been shown in Bi_2Te_3 that this technique can even track the buildup and dephasing of Floquet-Bloch states along one particular direction parallel to the surface [3]. A full two-dimensional (2D) momentum mapping of optically excited Dirac currents, however, has been so far demonstrated only without subcycle time resolution [4].

Here, we present first results on subcycle-resolved THz-ARPES on Bi_2Te_3 in the full 2D momentum space of the surface with a recently developed experimental setup. It combines the generation of few-cycle THz-pulses in the frequency range of 12-90 THz, which allow for field strengths of several MV/cm on the sample surface, with an ultrashort 400-nm two-photon probe of less than 15 fs duration at a repetition rate of 200 kHz.

[1] J. Reimann *et al.*, Nature **562**, 396 (2018).
[2] C. P. Schmid *et al.*, Nature **493**, 385 (2021).
[3] S. Ito *et al.*, Nature **616**, 696 (2023).
[4] J. Reimann *et al.*, Sci. Rep. **13**, 5796 (2023).

O 23.4 Tue 11:15 MA 041

Nonperturbative Floquet-Landau-Zener Mechanism in Multiphoton Photoemission on Metal Surface — •Yun Yen[1,2], Marcel Reutzel[3], Andi Li[4], Hrvoje Petek[4], and Michael Schüler[1,5] — [1]Laboratory for Materials Simulations, Paul Scherrer Institute, Villigen PSI, Switzerland — [2]Ecole Polytechnique Fédérale de Lausanne (EPFL), Lausanne, Switzerland — [3]Physikalisches Institut, Georg-August-Universitat Gottingen, Gottingen, Germany — [4]Department of Physics and Astronomy and Pittsburgh Quantum Institute, University of Pittsburgh, Pittsburgh, Pennsylvania, USA — [5]Department of Physics, University of Fribourg, Fribourg, Switzerland

In the Floquet engineering picture, time periodic optical fields perturbatively replicate states shifted by photon energy quanta, and cause field-dependent Autler-Townes splitting. As the field intensifies, light matter interaction shows more non-perturbative nature. Here we reveal the onset of non-perturbative responses in multiphoton photoemission (mPP) process for a driven two-level system of Cu(111) surface states. With strong enough driving, Floquet side bands form avoided crossing gaps, and thus lead to Landau-Zener (LZ) non-adiabatic tunneling within subcycle time scale. We further simulate the population dynamics with instantaneous Floquet state (IFS) formalism, and successfully reproduce experimental mPP features. Interpretation of the mPP process by Floquet-LZ theory elaborates the importance of non-adiabatic dynamics in strong field regime.

O 23.5 Tue 11:30 MA 041

Quantification of local electric fields in ultrafast photoemission experiments — •Marco Merboldt[1], Michael Schüler[2], David Schmitt[1], Jan Philipp Bange[1], Wiebke Bennecke[1], David Ramcke[1], Karun Gadge[3], Salvatore R. Manmana[3], Sabine Steil[1], G. S. Matthijs Jansen[1], Daniel Steil[1], Michael Sentef[4], Marcel Reutzel[1], and Stefan Mathias[1] — [1]Georg-August-Universität Göttingen, I. Physikalisches Institut, Germany — [2]Department of Physics, University of Fribourg, Fribourg, Switzerland — [3]Georg-August-Universität Göttingen, Institut für Theoretische Physik, Germany — [4]Institute for Theoretical Physics, University of Bremen, Bremen, Germany

The optical control of material properties using femtosecond laser pulses is one of the driving goals in ultrafast condensed matter physics. Optical parameters to control, e.g., far-from-equilibrium phases or hidden states, are the optical wavelength, the pulse duration and sequence, the polarization, and the electric field strength. For the latter, it is important to realize that the dielectric properties of the sample screen the incident light fields. Therefore, it is desirable to be able to determine the local field strength directly from the data of the time-resolved measurement.

Here, we show how to quantify the local electric field strength in trARPES experiments. We achieve this by analyzing the polarization- and momentum-resolved photoemission intensity originating from band replica generated via the laser-assisted photoelectric effect (LAPE).

O 23.6 Tue 11:45 MA 041

Momentum transfer in the pondermotive potential of VIS/NIR laser pulses detected by time-resolved ARPES — •Christian Strüber, Xinwei Zheng, and Martin Weinelt — Freie Universität Berlin, Berlin, Germany

Pump-probe photoemission measurements of ultrafast phenomena at reflective surfaces, e.g. demagnetization process in 3d or rare earth metals detected by tr-ARPES, are affected by the interaction of electrons with the ponderomotive potential of the light field. Oscillatory energy shifts have been detected when the probe laser pulse arrives before the pump pulse [1].

In our time- and angle-resolved photoemission spectroscopy (tr-ARPES) setup [2] using a hemispherical energy analyzer we measure the ponderomotive momentum transfer in dependence of the pump intensity and wave vector, the lateral momentum of the photoelectron and energy of the electronic state in the solid. We employ sub-50fs near infrared-pulses at 1300 nm to create the ponderomotive poten-

tial. Extreme ultraviolet pulses at 35 eV allow to detect a large range of lateral and perpendicular momenta. Additional to energy shifts of energetically narrow electronic states we observe squeezing of extended electron wavepackets leading to intensity modulations. Measurement results are simulated and fitted with high accuracy. Complete reversion of the ponderomotive momentum transfer allows for retrieval of the undisturbed initial state.

[1] U. Bovensiepen et al., *Phys. Rev. B* **79** (2009) 045415
[2] B. Frietsch et al., *Rev. Sci. Instrum.* **84** (2013) 075106

O 23.7 Tue 12:00 MA 041

Three-dimensional electron dynamics in PtBi2 — Paulina Majchrzak[1], •Charlotte Sanders[2], Yu Zhang[2], Andrii Kuibarov[3], Oleksandr Suvorov[3], Emma Springate[2], Bernd Büchner[3], Alexander N. Yaresko[4], Sergey Borisenko[3], and Philip Hofmann[1] — [1]Department of Physics & Astronomy, Interdisciplinary Nanoscience Centre, Aarhus University, 8000 Aarhus C, Denmark — [2]Central Laser Facility, STFC Rutherford Appleton Laboratory, Research Complex at Harwell, Harwell, OX11 0QX, UK — [3]Leibniz IFW Dresden, Helmholtzstr.. 20, 01069, Dresden, Germany — [4]Max-Planck-Institute for Solid State Research, D-70569, Stuttgart, Germany

Interplay between the three-dimensionally dispersing Weyl states of trigonal PtBi2 and complex many-body physics at the surface has been the subject of an intriguing recent study [1]. Key to these interesting physics is a set of Weyl points in conduction band states that are not occupied at room temperature and therefore cannot be directly probed by traditional methods of angle-resolved photoemission spectroscopy (ARPES). Using pump-probe time-resolved ARPES at the UK Artemis facility, along with the capability at Artemis for photon-energy scanning, we have been able to investigate three-dimensional dynamics in the conduction band of this interesting material, and to compare the results to theory predictions relating to the quasiparticle band dispersion and topologically non-trivial states. [1] arXiv:2305.02900 [cond-mat.supr-con]

O 23.8 Tue 12:15 MA 041

Efficient carrier multiplication in the topological insulator Bi2Se3 — •Michael Herb[1], Leonard Weigl[1], Niklas Hofmann[1], Johannes Gradl[1], Jason Khoury[2], Leslie Schoop[2], and Isabella Gierz[1] — [1]University of Regensburg — [2]Princeton University

Carrier multiplication, where the absorption of a single photon results in the formation of multiple electron-hole pairs, significantly increases the quantum yield of photodetectors and solar cells. It commonly occurs in semiconductors for incident photon energies that significantly exceed the size of the band gap. This makes the topological insulator Bi2Se3 with a bulk band gap of only ˜300meV a promising candidate for efficient carrier multiplication in the visible spectral range. Previous investigations of the non-equilibrium carrier dynamics in Bi2Se3 mainly focused on the topological surface state [1] and photovoltage effects [2]. We use time- and angle-resolved photoemission spectroscopy with visible 2eV pump pulses to investigate carrier dynamics inside the bulk bands. We find that the both the hole concentration in the valence band and the electron concentration in the conduction band keep increasing after the pump pulse is gone, providing direct evidence for the occurrence of carrier multiplication.

[1] Y. H. Wang et al., Phys. Rev. Lett. 2012
[2] M. Neupane et al., Phys. Rev. Lett. 2015

O 23.9 Tue 12:30 MA 041

Doping-Induced Modulation of Charge Density Wave States in 1*T*-TaS_2: Ultrafast Insights — •Jesumony Jayabalan[1], Florian K. Diekmann[2], Negar Najafianpour[1], Ping Zhou[1], Walter Schnelle[3], Martin Eckstein[4], Kai Rossnagel[2], and Uwe Bovensiepen[1] — [1]Universität Duisburg-Essen, Germany — [2]Christian-Albrechts-Universität zu Kiel, Germany — [3]Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany — [4]University of Hamburg-CFEL, 22761 Hamburg, Germany

1*T*-TaS_2 displays diverse charge density wave (CDW) ordered states influenced by temperature-dependent electron-electron and electron-phonon coupling. Chemically doping 1*T*-TaS_2 provides a potential means to adjust these couplings, thus modifying the material's properties. This presentation reports the ultrafast response of doped 1*T*-TaS_2 mainly in incommensurate CDW state, aiming to understand changes in electronic and structural properties. We present results on periodic lattice distortions induced by an infra-red ultrafast pulse, measured using ultraviolet time-resolved photoemission spectroscopy. Samples were doped with W or Mo by substituting a fraction of Ta during growth. Alongside temperature-dependent static photoemission, resistivity, and low energy electron diffraction studies, we demonstrate that the amplitude of oscillation and damping time are strongly influenced by CDW formation, dependent on the level of doping. We find that the defect atoms act as scattering centers which hinders the oscillations causing an increased damping with doping. Funding by the DFG through FOR 5249 QUAST is gratefully acknowledged.

O 23.10 Tue 12:45 MA 041

Sub-picosecond photo-induced local distortion reduction in Sb_2Te chalcogenide phase-change material — •Zhipeng Huang[1], Xinxin Cheng[2], Hazem Daoud[3], Wen-Xiong Song[4], R. J. Dwayne Miller[3], Klaus Sokolowski-Tinten[1], and R. Kramer Campen[1] — [1]Faculty of Physics and Center of Nanointegration (CENIDE), University of Duisburg-Essen, Duisburg, Germany — [2]SLAC National Accelerator Laboratory — [3]Departments of Chemistry and Physics, University of Toronto — [4]Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences

Chalcogenide phase-change materials can be switched reversibly between a low resistive opaque crystalline phase and a high resistive transparent amorphous phase by heating, electrical or optical pulses with (sub-)nanosecond switching speed. Femtosecond laser excitation provides an ultrafast, energy-efficient way for precisely manipulating the optical, electronic and structural properties of these materials.

Here we present our ultrafast electron diffraction and femtosecond-resolved sum frequency generation spectroscopy studies on Sb_2Te material under femtosecond UV pulse irradiation. We observed a sub-picosecond photo-induced non-thermal structural change in the crystallized Sb_2Te sample. Through careful structural factor calculations, we quantified the structural change corresponding to Sb/Te atom movement along the [111] plane to a less-distorted local structure by coupling to the A1g coherent optical phonon mode.

O 23.11 Tue 13:00 MA 041

Ultrafast Electron-Phonon Scattering in Dirac Antiferromagnets — Marius Weber[1,2], •Kai Leckron[1], Bärbel Rethfeld[1], and Hans Christian Schneider[1] — [1]Department of Physics and Research Center OPTIMAS University of Kaiserslautern - Landau, Campus Kaiserslautern, Germany — [2]Institut für Physik, Johannes Gutenberg University Mainz, Germany

In topological antiferromagnets the corresponding band and spin structure strongly depends on the orientation of the order parameter, for instance, two Dirac-cones with nearly linear dispersion at the border of the Brillouin zone for in-plane orientation of the Néel-vector may become a gapped system in another direction. This poses a challenge for the calculations of the electronic dynamics in these bandstructures, but the problem also applies more generally to all cases in which the electronic dynamics in the whole Brillouin zone are of interest and in which the bandstructure exhibits pronounced anisotropies. For this case and effectively 2D momentum spaces, we have developed a numerical approach which we present here for electron dynamics due to electron-phonon interactions in a model antiferromagnet. After a simulated instantaneous excitation of the electronic system, which leads to anisotropic carrier distributions, we demonstrate the effects of anisotropy on the electronic distribution functions and on macroscopic quantities like magnetization, and band-resolved carrier densities. We highlight the numerical accuracy of our implementation of the electron-phonon scattering which fully accounts for the density conservation requirements necessary for a dynamic spin-resolved calculation.