O 57: Ultrafast Electron Dynamics I

Time: Wednesday 10:30–13:00 Location: H2

O 57.1 Wed 10:30 H2

Imaging the subcycle dynamics of topological surface currents in two-dimensional momentum space — •Tim Bergmeier[1], Suguru Ito[1], Jens Güdde[1], and Ulrich Höfer[1,2] — [1]Fachbereich Physik, Philipps-Universität Marburg, Germany — [2]Fachbereich Physik, Universität Regensburg, Germany

Angle-resolved photoemission spectroscopy (ARPES), combined with THz-electric fields and subcycle temporal resolution, offers unique capabilites to explore light-matter interaction on timescales faster than the oscillation of a lightwave. With this approach, it is possible to directly observe the dynamics of lateral Dirac currents along the surface of topological insulators, as well as the ultrafast buildup and dephasing of Floquet-Bloch states. However, so far the momentum resolution of such experiments was restricted to only 1-dimensional cuts within the surface momentum space.

Here, we present first results on the subcycle dynamics of surface currents resolving the full Dirac cone of Bi_2Te_3. In our recently established setup in Marburg, a 200 kHz laser system allows for MV/cm field strengths by parametric amplification of frequency-tunable THz-pulses in the range of 20-40 THz, together with optimal focussing at large angles of incidence inside of the UHV chamber by astigmatism compensation. Combined with ultrashort 400nm two-photon probe pulses (<15 fs) and a Scienta DA30 photoelectron analyzer, we are able to measure the whole 2D-momentum space with subcycle resolution.

O 57.2 Wed 10:45 H2

Measuring the non-equilibrium electronic structure of phonon-driven 2D materials — •Niklas Hofmann and Isabella Gierz — University of Regensburg

Quasi-periodic driving of solids with tailored light fields has emerged as a promising pathway for non-equilibrium materials design. To bring this approach to the next level, tailored driving schemes targeting specific degrees of freedom need to be combined with ultrafast probes of the atomic and electronic structure. We combine pump pulses tunable all the way from the Terahertz to the visible spectral range with a time- and angle-resolved photoemission (trARPES) probe to gain access to the transient electronic structure of driven materials. We recently implemented a narrow-band, strong-field Terahertz source [1] that allows for the selective excitation of phonon modes in materials with strong spin-orbit coupling and extreme ultraviolet probe pulses with tunable pulse duration to trace band structure dynamics on sub-cycle as well as cycle-averaged time scales. In this talk we present proof-of-principle experiments on graphene and show the transient electronic structure of WS_2 driven at resonance to the E_{1u} phonon mode.

[1] Optics Letters 42, 129 (2017)

Invited Talk O 57.3 Wed 11:00 H2

Floquet engineering in black phosphorus — •Changhua Bao[1,2], Shaohua Zhou[2], Benshu Fan[2], Michael Schüler[3], Teng Xiao[2], Hui Zhou[4], Zhiyuan Sun[2], Peizhe Tang[5], Sheng Meng[4], Wenhui Duan[2], and Shuyun Zhou[2] — [1]Department of Physics, University of Regensburg, Regensburg, Germany — [2]Department of Physics, Tsinghua University, Beijing, China — [3]Laboratory for Materials Simulations, Paul Scherrer Institute, Switzerland — [4]Institute of Physics, Chinese Academy of Sciences, Beijing, China — [5]School of Materials Science and Engineering, Beihang University, Beijing, China

The time-periodic light field has emerged as a control knob for manipulating quantum states in solid-state materials, dubbed as Floquet engineering. In this talk, I will present our progress on the experimental realization of Floquet band engineering in a model semiconductor, black phosphorus. Driven by strong mid-infrared light fields, transient gap opening and band renormalization are directly resolved with exotic pseudospin selectivity. In addition to band-structure engineering, manipulation of symmetry properties through Floquet engineering is also explored, including ultrafast glide-mirror symmetry breaking and parity symmetry manipulation. This series of works provides a comprehensive understanding of Floquet engineering in semiconductors and important guidance for extending Floquet engineering into more materials.

Nat. Rev. Phys. 4, 33 (2022); Nature 614, 75 (2023); PRL 131, 116401 (2023); ACS Nano 18, 32038 (2024); Nat. Commun. in press

O 57.4 Wed 11:30 H2

Observation of Floquet states in graphene — •Marco Merboldt[1], Michael Schüler[2], David Schmitt[1], Jan Philipp Bange[1], Wiebke Bennecke[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

Recent advances in the field of condensed-matter physics have unlocked the potential to realize and control emergent material phases that do not exist in thermal equilibrium. One of the most promising concepts in this regard is Floquet engineering, the coherent dressing of matter via time-periodic perturbations. However, the broad applicability of Floquet engineering to quantum materials is in question, especially with respect to (semi-)metals and graphene in particular.

Here, we resolve this long-standing debate by using electronic structure measurements to provide direct spectroscopic evidence of Floquet effects in graphene [1]. We report light-matter-dressed Dirac bands by measuring the contribution of Floquet sidebands, Volkov sidebands, and their quantum path interference to graphene's photoemission spectrum. Fully supported by experiment and theory, we demonstrate that Floquet engineering in graphene is possible.

[1] Merboldt *et al.*, arXiv:2404.12791 (2024)

O 57.5 Wed 11:45 H2

Theory of nonperturbative nonlinear transport in a Floquet-Weyl semimetal — •Juan Ignacio Aranzadi[1], Matthew Day[2,3], James McIver[2,3], and Michael Sentef[1,2] — [1]Institute for Theoretical Physics, University of Bremen/ Bremen Center for Computational Material Science, Bremen — [2]Max Planck Institute for the Structure and Dynamics of Matter, CFEL, Hamburg — [3]Department of Physics, Columbia University, New York, NY, USA

Weyl semimetals exhibit unique topological properties characterized by pairs of Weyl nodes with opposite chirality. While their transport properties have been extensively studied [1], their behavior under strong optical driving fields remains poorly understood. In this regime, the material response is governed by photon-dressed Floquet-Bloch states. Here, we investigate the photocurrent production mechanisms in Td-MoTe2, a type-II Weyl semimetal, under strong circularly polarized fields. Recent transport measurements in this material revealed a linear scaling of photocurrents at strong fields (<0.4 MV/cm) for the circular photogalvanic effect and the light-induced Hall effect [2]. We explore the microscopic theory underlying the photocurrent production in this system and disentangle its various contributing mechanisms. [1] J. Hu, S. Xu, N. Ni, Z. Mao, Annual Review of Materials Research (2019) 49:1, 207-252 [2] M. Day et. al. Nonperturbative nonlinear transport in a topological light-matter hybrid (2024)

O 57.6 Wed 12:00 H2

Hybrid Frenkel-Wannier excitons facilitate ultrafast energy transfer at a 2D-organic interface — •Wiebke Bennecke[1], Ignacio Gonzalez Oliva[2], Jan Philipp Bange[1], Paul Werner[1], David Schmitt[1], Marco Merboldt[1], Anna M. Seiler[1], Daniel Steil[1], R. Thomas Weitz[1], Peter Puschnig[3], Claudia Draxl[2], G. S. Matthijs Jansen[1], Marcel Reutzel[1], and Stefan Mathias[1] — [1]I. Physikalisches Institut, Georg-August-Universität Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen, Germany — [2]Physics Department and CSMB, Humboldt-Universität zu Berlin, 12489 Berlin, Germany — [3]Institute of Physics, NAWI Graz, University of Graz, 8010 Graz, Austria

The combination of two-dimensional transition metal dichalcogenides (TMDs) and organic semiconductors (OSCs) is a highly promising material platform for the realization of future optoelectronic devices. While the excitonic properties of their individual components have been intensively studied, much less is known about excitons at the hybrid interface. Here, we use ultrafast momentum microscopy and many-body perturbation theory to investigate the exciton landscape at the PTCDA/WSe_2 interface [1]. In particular, we find an exciton state formed via Förster resonant energy transfer, which is of hybrid nature:

Concomitant intra- and interlayer electron-hole transitions within the OSC layer and across the TMD/OSC interface, respectively, give rise to an exciton wavefunction with mixed Frenkel-Wannier character.

[1] Bennecke *et al.*, arXiv:2411.14993 (2024)

O 57.7 Wed 12:15 H2

Characterization of Excitons for bulk Black Phosphorus — •Juan Felipe Pulgarin Mosquera[1,2], Geoffroy Kremer[2,3], Claude Monney[2], and Michael Schueler[1,2] — [1]PSI Center for Scientific Computing, Theory and Data, 5232 Villigen PSI, Switzerland — [2]Department of Physics, University of Fribourg, CH-1700 Fribourg, Switzerland — [3]Institut Jean Lamour, CNRS-Université de Lorraine

Excitons (coupled electron-hole pairs) in semiconductors can form collective states that exhibit spectacular nonlinear properties and possible applications in future optoelectronic devices. We present here some theoretical methods and a workflow for determining the excitonic wave functions and the corresponding excitonic binding energies for bulk Black Phosphorus. We solve the Bethe-Salpeter equations for coherent and incoherent excitations. The theoretical/numerical results are compared to the experimental ones of angle resolved photoemission spectroscopy (ARPES), by analyzing the spectra produced after including non-equilibrium electron-phonon dynamics; solving the time-dependent Boltzmann equation. These results allow us to understand the nature and characteristics of these two-particle bound states, together with some scattering effects, being challenging due to the stronger screened potential for 3D materials, resulting in short time excitations.

O 57.8 Wed 12:30 H2

Momentum-resolved valleytronic dynamics in TMDC monolayers — •Sarah Zajusch[1], Lasse Münster[2], Marcel Theilen[1], Marleen Axt[1], Yaroslav Gerasimenko[2], Jens Güdde[1], Robert Wallauer[1], and Ulrich Höfer[1,2] — [1]Fachbereich Physik, Philipps-Universität Marburg — [2]Fachbereich Physik, Universität Regensburg

The lack of inversion symmetry allows to access the valley degree of freedom in TMDC materials. With circular polarized light, time-resolved momentum microscopy enables us to trace the formation of a rich variety of bright and dark excitons typical for these materials on an ultrafast timescale and throughout the whole Brillouin zone. We performed measurements on both $MoSe_2$ and WS_2. In WS_2, we observe a valley depolarization at room temperature within 100 fs, whereas in $MoSe_2$, this happens on a slower timescale, with a slight polarization that is still visible long after 100 fs. There are mainly two proposed leading mechanisms for this relaxation process, exciton-phonon scattering and electron-hole exchange interaction, which are differently pronounced in W- and Mo-based TMDCs due to the differences in their excitonic energy landscapes. As the ultrafast formation of dark KK'- and KΣ-excitons in WS_2 shows, mainly exciton-phonon scattering contributes to the valley relaxation here. This process is strongly temperature-dependent.

O 57.9 Wed 12:45 H2

Formation and thermalization of non-equilibrium excitonic occupations — •Paul Werner[1], Jan P. Bange[1], Wiebke Bennecke[1], Giuseppe Meneghini[2], David Schmitt[1], Marco Merboldt[1], Anna Seiler[1], AbdulAziz AlMutairi[3], Samuel Brem[2], Junde Liu[1], Daniel Steil[1], Stephan Hofmann[3], R. Thomas Weitz[1], Ermin Malic[2], Stefan Mathias[1], and Marcel Reutzel[1] — [1]I. Physikalisches Institut, Georg-August Universität Göttingen, Germany — [2]Fachbereich Physik, Philipps-Universität Marburg, Germany — [3]Department of Engineering, University of Cambridge, United Kingdom

Semiconducting transition metal dichalcogenides (TMDs) host a rich landscape of Coulomb-correlated electron-hole pairs, which makes them ideal candidates for future optoelectronic applications. After an initial optical excitation of bright excitons, it is known that subsequent scattering into optically dark excitons takes place [1]. However, these processes must involve non-thermal exciton distributions that thermalize to a quasi-equilibrium [2]. In this work we use time-resolved momentum microscopy to directly access these non-thermal exciton distributions in optically excited homobilayer MoS_2. We identify the exciton landscape and relaxation pathways, and we find clear signatures of the non-equilibrium exciton distributions that are involved in the thermalization process. We compare our data with a full microscopic model calculation that confirms our experimental findings.

[1] Bange *et al.*, 2D Materials **10** 035039 (2023)
[2] Rosati *et al.*, ACS Photonics **7**, 2756–2764 (2020)