O 33: Poster Tuesday: Ultrafast Processes 2

Time: Tuesday 11:00–13:00 Location: P3

O 33.1 Tue 11:00 P3

Anisotropic carrier dynamics in single crystalline graphite — •Hauke Beyer, Petra Hein, Kai Rossnagel, and Michael Bauer — Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, Germany

Time- and angle-resolved photoelectron spectroscopy at 35 fs time-resolution is employed to study the carrier dynamics in the Dirac cone of graphite upon excitation with linearly polarized light. Due to the pseudo spin degree of freedom, the nascent photogenerated carrier distribution exhibits a strong anisotropy in momentum space. We observe the formation of a quasi-thermalized distribution on ultrafast timescales (~ 10 fs) through e-e interactions exhibiting a clear azimuthal anisotropy similar to findings for graphene [1]. For the azimuthal thermalization our data reveal a characteristic time scale of 40 ± 10 fs. The residual non thermal part of the electron distribution shows a time-dependent shift in energy by ~ 150 meV, which we assign to the formation of a steplike distribution due to e-ph interactions. The results are in good qualitative agreement with calculations based on a model introduced in Ref. [2].

[1] S. Aeschlimann *et al.*, Phys. Rev. B **96**, 020301(R) (2017).
[2] E. Malic *et al.*, Phys. Rev. B **84**, 205406 (2011).

O 33.2 Tue 11:00 P3

High-harmonic generation from the surface state of Bi_2Se_3 with THz driving fields — •Tim Bergmeier, Suguru Ito, Jens Güdde, and Ulrich Höfer — Fachbereich Physik, Philipps-Universität Marburg, Germany

The acceleration of charge carriers through the Dirac-point in the topologically protected surface state (TSS) of topological insulators (TIs) by strong electric fields with frequencies in the THz range gives rise to a nontrivial type of unusually efficient high-harmonic generation (HHG) as demonstrated for Bi_2Te_3 [1]. The long scattering times in the TSS resulting from spin-momentum locking are imprinted in the observation that the high-harmonic orders can be continuously shifted in frequency by varying the carrier-envelope phase of the driving field. This makes it possible to investigate the unusual transport properties in the TSS by this all optical method even at buried interfaces.

Here, we present first results for Bi_2Se_3 by using a newly developed setup that enables THz-HHG with a stable carrier-envelope phase at a repetition rate of 200 kHz reaching field strengths of up to 10 MV/cm in the frequency range of 12-90 THz. We show how the contribution of electrons in the TSS to the HHG spectra can be determined by measurements with varying field strengths and THz-frequencies above and below the bulk band gap, while analyzing the polarization of the high harmonic signal for different directions of the excitation in momentum space.

[1] C. P. Schmid *et al.*, Nature 593, 385 (2021).

O 33.3 Tue 11:00 P3

Towards time-resolved photoemission orbital tomography on van-der-Waals heterostructures — •Wiebke Bennecke, David Schmitt, Jan Phillip Bange, Mattis Langendorf, Katharina D. Feeser, Daniel Steil, Sabine Steil, Marcel Reutzel, G. S. Matthijs Jansen, and Stefan Mathias — I. Physikalisches Institut, Georg-August-Universität Göttingen

Heterostructures of two-dimensional van-der-Waals materials with molecular thin films provide an exceptional platform to tailor electronic energy level alignments that govern the optoelectronic response of such materials. In addition to the energy-level alignment, a detailed knowledge of the electronic wavefunctions in these systems would help to fundamentally understand optical excitations, exciton generation, charge-transfer and relaxation processes. Here, time-resolved orbital tomography is a promising method that potentially provides such information. On this poster, we show first results of femtosecond orbital tomography of PTCDA monolayers adsorbed on bulk as well as monolayer WSe_2.

O 33.4 Tue 11:00 P3

Transient optical properties in non-equilibrium laser excited noble metals — •Marius Wenk[1], Pascal D. Ndione[1], Sebastian T. Weber[1], Dirk O. Gericke[2], and Baerbel Rethfeld[1] — [1]Department of Physics and OPTIMAS Research Center, Technische Universität Kaiserslautern — [2]CFSA, Department of Physics, University of Warwick

Ultrashort laser pulses can induce strong modifications of material properties of solids such as creating highly transient optical parameters. After excitation with lasers of high power, the conduction electrons thermalize quickly to a hot Fermi distribution. Yet, the band occupation numbers can still be far from equilibrium.

We study excitation of noble metals such as copper and gold with visible photons. We use a two-temperature model and construct electron density-resolved rate equations to simulate the non-equilibrium band occupation [1]. Applying the results of the simulation, particularly the occupation and temperature data, we compute the time-resolved dielectric function based on the Drude-Lorentz formalism. In order to compare the results with experimental data, we calculate the transient optical properties such as probe reflectivity and transmissivity. Our predictions are compared with time-resolved measurements of optically excited metals, thus providing insights to electron dynamics on femto- and picosecond timescales.

[1] Pascal D. Ndione, Sebastian T. Weber, Dirk O. Gericke, and Baerbel Rethfeld. Scientific Reports, 12(1) 4693 (2022)

O 33.5 Tue 11:00 P3

LabVIEW based software solution for tr-ARPES experiments — •Johannes Gradl[1], Niklas Hofmann[1], Leonard Weigl[1], Yu Zhang[2], Cephise Cacho[3], Neeraj Mishra[4,5], Stiven Forti[4], Camilla Coletti[4,5], and Isabella Gierz[1] — [1]Department of Physics, University of Regensburg, 93040 Regensburg, Germany — [2]Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot OX11 0DE, United Kingdom — [3]Diamond Light Source, Harwell Campus, Didcot OX11 0DE, United Kingdom — [4]Center for Nanotechnology Innovation @NEST, Istituto Italiano di Tecnologia, 56127 Pisa, Italy — [5]Graphene Labs, Istituto Italiano di Tecnologia, 16163 Genova, Italy

Time- and angle-resolved photoemission spectroscopy (tr-ARPES) is an experimental technique used for visualizing non-equilibrium carrier dynamics as well as transient band structures of photoexcited samples as a function of energy, momentum, and time. A tr-ARPES setup ideally includes femtosecond pump pulses with tunable photon energy, femtosecond extreme ultraviolet probe pulses an ultrahigh vacuum chamber equipped with a cryo-cooled sample manipulator and a photoelectron analyzer as well as additional features for in situ sample preparation and characterization. In addition to this hardware, the execution of tr-ARPES experiments requires a user-friendly software for hardware control, data acquisition, and real time data visualization. We will present our LabVIEW based software solution and demonstrate its successful implementation with a series of data sets from a WS2/graphene van der Waals heterostructure.

O 33.6 Tue 11:00 P3

Ultrabroadband THz-STM and its application to study hot electron dynamics in metals — Natalia Martín Sabanés[1,2], Faruk Krecinic[1], •Vivien Sleziona[1], Takashi Kumagai[1,3], Fabian Schulz[1,4], Luis Enrique Parra Lopez[1], Alkisti Vaitsi[1], Martin Wolf[1], and Melanie Müller[1] — [1]Fritz-Haber Institute of the Max-Planck Society, Berlin, Germany — [2]IMDEA Nanoscience, Madrid, Spain — [3]Institute of Molecular Science, 444-8585 Okazaki, Japan — [4]CIC NanoGUNE BRTA, San Sebastian, Spain

Localized ultrafast currents across the junction of a scanning tunneling microscope (STM) can be generated by photo-assisted hot electron tunneling or *cold* lightwave-induced tunneling. In addition, but so far not considered, fs laser excitation can induce a transient thermalized electron distribution that can give rise to an ultrafast current component. Here we investigate the role of ultrafast thermionic tunneling for photoinduced hot electron tunneling from a photoexcited STM tip [1]. We access the dynamics of hot electron tunneling by phase-resolved sampling of ultrabroadband Terahertz (THz) waveforms inside the STM junction. Our results reveal the strong nonthermal character of photoinduced hot electron tunneling, and provide a new route to probe hot electron dynamics in metals using THz-STM. Furthermore, we report on the development of an ultrabroadband THz-STM with tunable optical excitation for the spatiotemporal investigation of photocarrier dynamics at metal-semiconducting interfaces. [1] N.

Martín Sabanés et al., 10.48550/arXiv.2205.08248

O 33.7 Tue 11:00 P3

Dark Exciton Formation Dynamics in TMDC Monolayers — •Sarah Zajusch[1], Lasse Münster[1], Raul Perea-Causin[1], Samuel Brem[1], Katsumi Tanimura[1], Jens Güdde[1], Yaroslav Gerasimenko[2], Rupert Huber[2], Ermin Malic[1], Ulrich Höfer[1], and Robert Wallauer[1] — [1]Fachbereich Physik, Philipps-Universität Marburg, Germany — [2]Fachbereich Physik, Universität Regensburg, Germany

Charge transfer processes in two-dimensional TMDCs are governed by the formation of excitons. The excitonic landscape comprises optically accessible bright excitons as well as momentum- and spin-forbidden dark states. Our experimental setup combines time-resolved momentum microscopy with probe energies in the XUV regime which provides direct access to k-resolved exciton dynamics within the whole Brillouin zone on an ultrafast time scale.

Although structural features are very similar for different semiconductor TMDC materials, slight variations in both electronic band ordering and excitonic binding energy can drastically modify the excited population dynamics. We present a comparison of exciton formation in the two extreme cases of WS_2 and $MoSe_2$ monolayers after valley-sensitive excitation with circular polarized light. On the one hand, in $MoSe_2$ the bright KK-exciton is the energetically most favorable state. On the contrary, in WS_2, we additionally observe the ultrafast formation of energetically lower dark KK'- and KΣ-excitons. The microscopic understanding of these processes is crucial with regard to interlayer exciton formation in TMDC heterostructures.

O 33.8 Tue 11:00 P3

Time- and Angle-Resolved Photoelectron Spectroscopy with Nano-Focused Surface Plasmon Polaritons — •Alexander Neuhaus, Pascal Dreher, David Janoschka, Michael Horn-von Hoegen, and Frank-J. Meyer zu Heringdorf — Faculty of Physics and Center for Nanointegration, Duisburg-Essen (CENIDE), University of Duisburg-Essen, 47048 Duisburg, Germany.

Non-perturbative interactions of intense light fields with the electronic band structure in a solid can result in transient electronic properties. The experimental conditions required to realize the necessary field strength can be realized in nano-optical systems, as these can be designed to provide tremendous enhancements of the local field amplitude. Ultimately, observing the non-equilibrium electron dynamics in such systems requires a combination of precise control over the local driving field, state resolution, and spatial selectivity.

Here, we explore electron emission from nano-focused femtosecond surface plasmon polariton (SPP) pulses, providing us with deep-subwavelength spatial selectivity. Time- and angle-resolved photoelectron spectroscopy with attosecond precision provides us access to the coherent and incoherent dynamics of the electron emission process. The technique is applied to the system Cs/Au(111), where we find a resonant enhancement of the electron emission by an image potential state.

O 33.9 Tue 11:00 P3

Ultra Fast Dynamics in Modified Thiophene based Conjugated Donor-Acaptor Organcic Polymers — •Tobias Reiker[1,2], Carsten Winter[1], Deb Kumar Bhowmick[1,2], Nils Fabian Kleimeier[1,2], Zitong Liu[3], Deqing Zhang[3], and Helmut Zacharias[1,2] — [1]Center for Soft Nanoscience, University of Münster, Germany — [2]Physikalisches Institut, University of Münster, Germany — [3]Institute of Chemistry, Chinese Academy of Science, Beijing, China

Thiophene-based polymers are promising candidates for solar cell, OLED or transistor applications. An internal donor - acceptor system is formed by coupling thiophene polymers with pyrrole chains. The charge transport behavior can be tuned by different alkyl side chains since they influence the electronic structure. A direct assessment of the intramolecular and intermolecular dynamics may guide synthesis routes. With pDPP4T, pDPP4T and pDPTTT we investigated the electronic dynamics of verified high hole-mobility organic semiconductors. Either the backbone or the side chains were modified. In contrast, another polymer pF8T2 with bi-thiophene in the backbone was used, but with fluorene instead of pyrrole as acceptor. These different molecular configurations are intended to provide insights into the change in electron configuration due to both backbone modification and intermolecular packing. We report results of temporally resolved photoemission studies on thiophene polymers on silicon substrates.

O 33.10 Tue 11:00 P3

Density-dependent electron-phonon coupling in multiband systems. — •Tobias Held, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Germany

If a solid is irradiated with a short-pulsed laser in the visible spectrum, the energy is almost entirely absorbed by the electrons while the lattice remains cold. The subsequent energy flow between electrons and phonons is usually described by the electron-phonon coupling parameter, which plays a central role in the Two-Temperature Model and most other temperature-based models. This coupling parameter depends on a multitude of observables. Most frequently a dependence on the electron temperature is considered.
In this work, we aim to investigate how a varying density distribution between different electron subsystems affects the coupling parameter. In gold, we distinguish between sp- and d-electrons and in magnetic nickel between majority and minority spins. Our results show that for gold, the total coupling strongly depends on the density distribution, while for nickel it is largely independent on the spin densities. In the latter case, the individual coupling contributions of the bands change significantly with density but mostly compensate each other in terms of the total coupling.

O 33.11 Tue 11:00 P3

Electron Dynamics of an intercalated Graphene Layer on Nickel — •Katharina Hilgert[1], Christina Schott[1], Eva Walther[1], KaMan Yu[1], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,2] — [1]Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, 67663 Kaiserslautern Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, 55128 Mainz, Germany

One of the great challenges in information technology is to develop novel concepts for the realization of active functional units on eversmaller length scales. The simplest approach to reduce the size of any device structure is to employ atomically thin materials as graphene with their unique electronic and optical properties. However, these are often altered on surfaces due to strong chemical interaction with the substrate material. On this poster we present our approach to restore and design the electronic properties of graphene on a Ni(111) surface by intercalation of lead atoms. The changes in the electronic band structure of graphene on Ni(111) are monitored by angle resolved photoelectron spectroscopy with extreme ultra violet radiation. On the highly reactive Ni(111) surface, the linear dispersion of the Dirac cone of the free-standing graphene sheet is completely suppressed by the strong graphene surface interaction. This changes significantly after the intercalation of Pb. The band structure of the graphene/Pb/Ni(111) system reveals again the linear dispersion resembling the behavior in free-standing graphene. This has clear consequences on the carrier dynamics near the K-point as we will discuss in this contribution.