O 68: Focus Session: Time-Resolved Momentum Microscopy

Time-resolved momentum microscopy is a new experimental technique to study electron dynamics in momentum space. In momentum microscopy, the reciprocal image plane of photoemitted electrons is mapped onto a position-sensitive detector. In addition, the kinetic energy is extracted by energy dispersive elements, such as Time-of-Flight drift tubes or hemispherical analysers. Hereby, the complete information about the electronic structure of two-dimensional materials can be obtained (kx, ky, E). At the same time the signal can be restricted to micrometer-size spots, which is important for many types of quantum materials. By combining such electron spectrometers with pump-probe laser techniques, optical excitations, photo-induced phase transitions and charge transfer processes within the entire Brillouin zone can be imaged on ultrashort time scales. Several groups, with many of them in Germany, have established this technique during the last years in lab-based setups. In addition, first setups at large scale facilities, like ELI-ALPS or FLASH, are being commissioned or in operation. With higher photon energies available at free electron lasers, also structural dynamics will be observable through time-resolved electron diffraction.

Organizer: Robert Wallauer (Universität Marburg)

Time: Thursday 15:00–18:30 Location: H3

Topical Talk O 68.1 Thu 15:00 H3

Exploring Excitonic Excitations in Momentum Space — •Keshav Dani — Okinawa Inst. of Science and Technology, Graduate University, Onna-son, Japan

Optical techniques have provided us with rich information about the exciton * a two-particle photoexcited state in semiconductors and insulators. Yet, they have left a fundamental degree of freedom of the exciton inaccessible * it*s momentum! In this talk, I will discuss the application of time-resolved photoemission techniques to access the momentum coordinate of excitons in 2D semiconductors, thereby providing us with the formation pathways of momentum-forbidden dark excitons [1], an image of the electron around the hole in the exciton, the observation of the long-predicted anomalous dispersion of the exciton-bound electron [2], the momentum distribution of the exciton-bound hole, and the confinement of the interlayer exciton in a moiré cell [3].

References (*equal authors) [1] J. Madeo*, M. K. L. Man*, et al. Science 370, 1199 (2020). [2] M. K. L. Man*, J. Madeo*, et al. Science Advances 7, eabg0192 (2021). [3] O Karni*, E. Barr*, V. Pareek*, J. D. Georgaras*, M. K. L. Man*, C. Sahoo*, et al. Nature 603, 247 (2022).

Topical Talk O 68.2 Thu 15:30 H3

Moiré interlayer and charge-transfer excitons in space and time: new experiments enabled by time-resolved momentum microscopy — •Stefan Mathias — I. Physikalisches Institut, Georg-August-Universität Göttingen, Germany

In my talk, I introduce the time-resolved momentum microscopy setup that we developed in Göttingen (Germany) [1], which includes a MHz repetition rate extreme-ultraviolet beamline, and has recently been upgraded with a spin-imaging detector. In the following, I will discuss exemplary research projects that we cover with this new instrument. In particular, I will focus on the spatio-temporal identification and dynamics of moiré interlayer excitons in twisted WSe2/MoS2 heterostructures [2], and on an orbital-resolved study [3] of charge-transfer exciton dynamics in C60 thin films [4].

[1] Keunecke et al., Rev. Sci. Instr. 91, 063905 (2020)
[2] Schmitt et al., arXiv:2112.05011(2021)
[3] Jansen et al., New Journal of Physics 22, 063012 (2020)
[4] Stadtmüller et al., Nature Communications 10, 1470 (2019)

O 68.3 Thu 16:00 H3

The complete interface molecular movie: One-stop imaging of orbital, electronic, and structural dynamics — •Markus Scholz[1], Kiana Baumgärtner[2], Marvin Reuner[3], Christian Metzger[2], Michael Heber[1], Dmytro Kutnyakhov[1], Friedrich Reinert[2], Daria Popova-Gorelova[3], Martin Beye[1], and Kai Rossnagel[4] — [1]Deutsches Elektronen-Synchrotron DESY, Notkestrasse 85, 22607 Hamburg, Germany — [2]Experimentelle Physik 7, Julius-Maximilians-Universität, Am Hubland, 97074 Würzburg, Germany — [3]I. Institute for Theoretical Physics and Centre for Free-Electron Laser Science, Universität Hamburg, Luruper Chaussee 149, 22607 Hamburg, Germany — [4]Institut für Experimentelle und Angewandte Physik, Christian-Albrechts-Universität zu Kiel, 24098 Kiel, Germany

Reactions of molecules on surfaces involve a complex interplay between electronic reorganization and the motion of atoms. The recent developments of higher-harmonics generation (HHG) and free-electron laser (FEL) based sources open up new opportunities for ultrafast spectroscopy toward simultaneously imaging subtle changes in the electronic structure and atomic positions on a femtosecond time scale and with sub-Ångström spatial resolution. Here, using a dual-electron messenger mode of momentum microscopy, we trace a photoinduced charge transfer process in energy-momentum space using valence electrons and, synchronously, at atomic sites using core electrons, in a single experimental setup for a molecule-2D material interface.

O 68.4 Thu 16:15 H3

Electron dynamics after a spin- and valley-polarized electronic excitation in WS_2 — •Lasse Münster[1], Sarah Zajusch[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

Atomically thin layers of TMDCs offer an ideal playground to study ultrafast electron dynamics. After optical excitation electrons scatter throughout the Brillouin zone to form a variety of excitonic states. We have shown that this formation process can be imaged by time-resolved momentum microscopy with tunable pump and high harmonic probe [1].

We recently incorporated a new pumping scheme, that allows us to excite the sample under an incident angle close to 0° with circularly polarized light. This results in an excitation, which is located purely within the K valley for one helicity and in the K' valley for the other helicity and provides access to all possible scattering processes. In the case of an excitation at K, electron scattering to K' and Σ is mediated by strong electron-phonon coupling with time constants of a few tens of femtoseconds. In addtion, we observe the formation of spin-forbidden excitons in the K valley and electron scattering towards Σ'. Both of these processes involve a spin-flip and are significantly slower (50 - 100 fs).

[1] R. Wallauer et al., Nano Lett. 21, 5867 (2021)

Topical Talk O 68.5 Thu 16:30 H3

Momentum and energy dissipation of hot electrons in metals and metal-molecular heterostructures — •Benjamin Stadtmueller — Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, 67663 Kaiserslautern, Germany — Institute of Physics, JGU Mainz, 55128 Mainz, Germany

The dynamics of optically excited carriers in condensed matter play a crucial role in many fundamental and device-relevant processes in materials. Here, I discuss the potential of time-resolved two-photon momentum microscopy (tr-2PMM) [1] to access the k-space signatures and the corresponding quasi-particle lifetimes of excited carriers in metals [2,3] and metal-molecular heterostructures. For metallic systems, I will demonstrate that tr-2PMM allows us to uncover anisotropies in the orientations of optical transition dipoles for highly free electron like noble metal surfaces [2] and to follow the ultrafast

intra- and interband scattering processes of optically excited carriers in metallic quantum well systems [3] in real-time. In this context, I will also introduce two approaches to alter the electron dynamics of metal surfaces by the adsorption of molecular complexes or by periodically modulating the surface potential using porous molecular networks on surfaces. Finally, I will show our efforts to exploit the spatial resolution in tr-2PMM to disentangle the k-space signatures of photo- and plasmon-induced hot carriers at surfaces [4]. [1] F. Haag et al., Rev. Sci. Instrum. 90, 103104 (2019); [2] T. Eul et al. Nat. Commun. (2022); [3] F. Haag et al. Phys. Rev. B 104, 104308 (2021); [4] M. Hartelt et al. ACS Nano 15, 19559 (2021)

O 68.6 Thu 17:00 H3

Coherent response of the electronic system driven by non-interfering laser pulses — •Tobias Eul[1], Eva Prinz[1], Michael Hartelt[1], Benjamin Frisch[1], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,2] — [1]Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, Germany

The strength of light-matter interaction in condensed matter is fundamentally linked to the orientation and oscillation strength of the materials' optical transition dipoles. Structurally anisotropic materials, e.g. elongated molecules, exhibit optical transition dipoles with fixed orientations that govern the angular-dependent light-matter interaction. Contrary, free electron like metals should exhibit isotropic light-matter interaction with the light fields dictating the orientation of the optical transition dipoles. Here, we demonstrate that an anisotropic direction of the optical transition dipoles even exists in highly free electron like noble metal surfaces. Our time- and phase-resolved photoemission experiment reveals coherent interference effects on the (110)-oriented silver surface after optical excitation with two non-interfering cross-polarized pulses. We explain this coherent material response within the density matrix formalism by an intrinsic coupling of the non-interfering light fields mediated by optical transition dipoles with fixed orientations in silver.

O 68.7 Thu 17:15 H3

Time-resolved photoemission orbital tomography of CuPc on Cu(001)-2O — •Alexa Adamkiewicz[1], Miriam Raths[2], Monja Stettner[2], Francois C. Bocquet[2], Christian Kumpf[2], Robert Wallauer[1], F. Stefan Tautz[2], and Ulrich Höfer[1] — [1]Fachbereich Physik, Philipps Universität Marburg, Germany — [2]Peter Grünberg Institute (PGI-3), Jülich Research Centre, Germany

Charge transfer across molecular interfaces is reflected in the population of electronic orbitals. For ordered organic layers, time-resolved photoemission orbital tomography (tr-POT) is capable of spectroscopically identifying the involved orbitals and deducing their population from the measured angle-resolved photoemission intensity with high temporal resolution. In a first example, we found that for PTCDA/Cu(001)-2O, two distinct excitation pathways could be observed with visible light [1]. The parallel component of the electric field induces a direct HOMO-LUMO transition, the perpendicular component transfers a substrate electron into the molecular LUMO. While in this case, a distinct excitonic signature was not observed, changes in the momentum pattern can in general serve as a measure of detecting excitonic processes. Here, we show such time-dependent change of the pattern for CuPc/Cu(001)-2O. We demonstrate how the temporal evolution of the LUMO momentum distribution can be systematically disentangled from contributions of the projected HOMO. Moreover, we observe LUMO excitation of selected molecular orientation at normal incidence by aligning the pump polarization along the molecular axis. [1] R. Wallauer et al., Science 371, 1056 (2021).

Topical Talk O 68.8 Thu 17:30 H3

Is there a perfect electron analyzer for time-resolved ARPES? — •Laurenz Rettig — Fritz-Haber-Institut der Max-Planck-Gesellschaft

Recent years have seen a huge popularity of time-of-flight based momentum microscopes (MMs), which based on advanced electrostatic optics have revolutionized angle-resolved photoemission spectroscopy (ARPES). Compared to conventional hemispherical analyzers (HAs) with angle-dispersing electron lenses, which access only a small fraction of the reciprocal space at a given configuration, MMs allow for the simultaneous detection of multiple Brillouin zones without the need to rearrange the sample geometry. However, one drawback of such instruments, in particular in time-resolved studies, arises from the large energy and momentum range covered simultaneously, which in combination with detection limitations of delay-line detectors and space charge restrictions can severely reduce the effective detection rate for selected energy-momentum regions compared to conventional HAs.

In my talk I will discuss the advantages and limitations of both types of instruments for several application scenarios in time-resolved ARPES, and present some recent highlights in charge-density wave materials, which take advantage of the combination of both types of instruments.

O 68.9 Thu 18:00 H3

Developing a fs-XUV source for time-resolved momentum microscopy on 2D materials — •Karl Schiller[1], Alan Omar[2], Lasse Sternemann[1], Matija Stupar[1], Clara Saraceno[2], and Mirko Cinchetti[1] — [1]Department of Physics, TU Dortmund University, Germany — [2]Ruhr-Universität Bochum, Germany

We introduce a newly developed setup for time-resolved momentum microscopy with femtosecond extreme ultraviolet (fs-XUV) radiation. The fs-XUV pulses with photon energy up to 30 eV are generated by high-harmonic generation in an Argon gas jet and are coupled to an energy-filtered momentum microscope (KREIOS MM, Specs GmbH). A commercial Ytterbium-based high-power laser system drives the generation with a variable high repetition rate between 100 kHz and 1 MHz (Carbide, Light Conversion). While this configuration allows for high energy resolution of 50 meV, a high temporal resolution can also be reached by coupling the laser to a tunable in-house build Herriot-type multipass cell compressor with peak powers up to 2 GW and pulse durations of $\sim$45 fs [1]. In the talk, we will present the first characterization data of this setup and motivate its capability to examine few-layer 2D materials.

[1] A. Omar, et al. *Advanced Solid State Lasers*, OSA Technical Digest (Optical Society of America, 2021), paper JM3A.55

O 68.10 Thu 18:15 H3

FEL-based time-of-flight momentum microscopy: 3 time-resolved photoemission modalities in 1 experiment — •D. Kutnyakhov[1], R.P. Xian[2], M. Dendzik[2], M. Heber[1], F. Pressacco[3], S.Y. Agustsson[4], L. Wenthaus[1], H. Meyer[3], S. Gieschen[3], K. Bühlman[5], S. Däster[5], R. Gort[5], D. Curcio[6], K. Volckaert[6], M. Bianchi[6], Ch. Sanders[6], J.A. Miwa[6], S. Ulstrup[6], A. Oelsner[7], C. Tusche[8,9], Y.-J. Chen[8,9], D. Vasilyev[4], K. Medjanik[4], G. Brenner[1], S. Dziarzhytski[1], S. Dong[2], J. Hauer[2], L. Rettig[2], J. Demsar[4], H.-J. Elmers[4], Ph. Hofmann[6], R. Ernstorfer[2], G. Schönhense[4], Y. Acremann[5], and K. Rossnagel[1,10] — [1]DESY, Hamburg — [2]FHI Berlin — [3]CFEL, Univ. Hamburg — [4]Univ. Mainz — [5]ETH Zürich — [6]Univ. Aarhus — [7]Surface Concept GmbH, Mainz — [8]FZ Jülich GmbH — [9]Univ. Duisburg-Essen — [10]CAU Kiel

Time-resolved photoemission spectroscopy with ultrashort pump and probe photon pulses is an emerging technique with wide application potential. The ultimate combination of valence-band and core-level spectroscopy with photoelectron diffraction in a single experiment for electronic, chemical, and structural dynamics analysis specifically requires tunable monochromatic soft X-ray pulses at a high repetition rate as well as highly efficient single-shot electron detectors with increased multi-hit capabilities. We have realized such a 3-in-1 ultrafast photoemission experiment at FLASH/PG2, DESY merging free-electron laser capabilities with a multi-dimensional recording scheme.