O 36: Poster: Ultrafast Electron Dynamics at Surfaces and Interfaces

Time: Tuesday 18:00–20:00 Location: Poster C

O 36.1 Tue 18:00 Poster C

Differences in the electron dynamics of inequivalent (001) surfaces of *Td*-WTe_2: top and bottom or topological trivial and topological non-trivial? — •Klaas Opitz[1], Hauke Beyer[1], Eduard Moos[1], Kai Rossnagel[1,2,3], and Michael Bauer[1,2] — [1]Institute of Experimental and Applied Physics, Kiel University, 24098 Kiel, Germany — [2]Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, 24118 Kiel, Germany — [3]Deutsches Elektronen-Synchrotron DESY, Ruprecht Haensel Lab, 22607 Hamburg, Germany

ARPES spectra of [100]-oriented *Td*-WTe_2 show clear differences, which in the past have been associated either with different topological phases [1] or alternatively interpreted as a signature for the small but finite structural differences of the (001) and (00$\bar{1}$) surface resulting from the broken inversion symmetry of the material [2]. In this contribution we will present time-resolved ARPES data of *Td*-WTe_2 that clearly hint to distinct differences also in the electron dynamics for these two surfaces. We will discuss our results under consideration of the different interpretations of the ARPES data.

[1] Y. Wu *et al.*, Phys. Rev. B. **94**, 121113(R) (2016).
[2] Y. Wan *et al.*, Phys. Rev. B. **105**, 085421 (2022).

O 36.2 Tue 18:00 Poster C

Second harmonic spectroscopy of Fe-porphyrin/Cu(001) interfaces — •Newsha Vesalimahmoud, Mahendra Kabbinahithlu, Ping Zhou, Uwe Bovensiepen, and Andrea Eschenlohr — University of Duisburg-Essen, Faculty of Physics and CENIDE, Lotharstr. 1, 47057 Duisburg, Germany

Hybrid systems of metallo-porphyrin/metal interfaces, which are known as spinterfaces, are promising candidates to control the spin-dependent transport in spintronic devices [1]. This study focuses on the iron porphyrin molecules (FeOEP)/Cu(001) interface using interface-sensitive time-resolved second harmonic generation (SHG) spectroscopy. We analyze the polarization and wavelength dependent SHG for different molecular adsorbate thicknesses on Cu(001) using a fundamental beam in the range of 500 - 700 nm. Polarization dependent measurements show a higher p-P SHG yield than s-P SHG, as the intrinsic absolute value of $|\chi^{(2)}_{zzz}|$ is much larger than $|\chi^{(2)}_{zxx}|$. Moreover, SHG spectra show an increasing intensity up to a maximum at 2.3 eV for Cu(001) due to one ω resonance. After adsorption of 2 monolayers (ML) and 5ML FeOEP/Cu(001), the spectral dependence remains similar to the Cu(001) surface except for an enhancement at 2.25 eV for 5ML FeOEP/Cu(001). We attribute this enhancement to the energetic position of the lowest unoccupied molecular orbital (LUMO) at the FeOEP/Cu(001) interface. Moreover, we discuss the electron dynamics analyzed through pump-probe SHG at on- and off-resonant photon energies.

[1] Wende, H., et al., Nat. Mater, **6**, 516 (2007).

O 36.3 Tue 18:00 Poster C

Transient Absorption Spectroscopy of NiO — •Mahendra Kabbinahithlu[1], Tobias Lojewski[1], Sergey Kovalenko[2], Nico Rothenbach[1], Katharina Ollefs[1], Heiko Wende[1], Uwe Bovensiepen[1], Julia Stähler[2], and Andrea Eschenlohr[1] — [1]Universität Duisburg-Essen, Fakultät für Physik und Center for Nanointegration (CENIDE), Lotharstraße 1, 47057 Duisburg, Germany — [2]Humboldt-Universität zu Berlin, Institut für Chemie, Brook-Taylor-Straße 2, 12489 Berlin, Germany

In nickel oxide (NiO), a correlated transition-metal oxide, the strong electron-electron repulsion splits the d-bands into an occupied lower Hubbard band (LHB) and an unoccupied upper Hubbard band (UHB). The presence of additional oxygen p-bands from adjacent sites, located between the Hubbard bands makes NiO a charge-transfer insulator. We present excited state dynamics in NiO thin films via time-resolved optical spectroscopy. NiO is pumped above the band gap with 3.98 eV photons and the transient absorption spectrum is probed using a time-delayed supercontinuum. The time-resolved absorption spectrum shows within the first 500 fs pump-induced changes at 2.2 eV, 3.2 eV and 4.3 eV. The negative change represents the reduced transitions from the bleached ground state and the positive changes represent the increased transitions from the photo-excited state. Around 2 ps, relaxation from the photo-excited state leads to emergence of a fluence dependent positive feature. We discuss this feature as transitions from polarons and shallow defect states lying below the UHB edge.

O 36.4 Tue 18:00 Poster C

Laser-induced ultrafast electron dynamics based on a two-band model — •Stephanie Roden, Tobias Held, Sebastian T. Weber, and Bärbel Rethfeld — Department of Physics and Research Center OPTIMAS, RPTU, Kaiserslautern-Landau

After an ultrashort laser excitation, the energy of an optical laser pulse is absorbed by the electrons of a solid. The thermalization of the excited electron system and the relaxation with the phonons to a joint temperature can be calculated in a kinetic manner by coupled Boltzmann collision integrals.

Previous implementations are based on an effective one-band model for the electron system [1]. This approximation does not allow a distinction between intra- and interband relaxation within the electron system. However, a separate consideration of these relaxation processes can strongly influence calculated optical parameters [2] as well as the electron-phonon coupling [3]. In this work, we extend the existing one-band model to an energy-resolved two-band model for a thin gold film that distinguishes between free sp- and more localized d-electrons. We show selected results of the intertwined relaxation dynamics following an ultra-short laser pulse.

[1] B. Y. Mueller and B. Rethfeld, PRB 87, 035139 (2013)
[2] P. D. Ndione, S. T. Weber, D. O. Gericke and B. Rethfeld, Sci. Rep. 12, 4693 (2022)
[3] T. Held, S. T. Weber, and B. Rethfeld, arXiv:2308.01067 (2023)

O 36.5 Tue 18:00 Poster C

Coherent and incoherent ultrafast lattice dynamics in WTe_2 — •Hanqian Lu[1,2], Victoria Taylor[2], Hyein Jung[1,2], Jannik Malter[2], Ralph Ernstorfer[1,2], and William Windsor[1,2] — [1]Technische Universität Berlin — [2]Fritz Haber Institute of the Max Planck Society

WTe_2 is a layered transition metal dichalogenide, which has attracted significant attention in recent years, in particular due to its potential as a topological material and a polar metal. Here we study its response to ultrafast photoexcitation using femtosecond electron diffraction (FED). We observe both coherent and incoherent photoinduced lattice dynamics, in particular the long-lived 0.23 THz shear mode. We quantify these responses along different crystal axes, and present a microscopic model of atomic vibrations of all atoms in the unit cell.

O 36.6 Tue 18:00 Poster C

Time-resolved photoelectron spectroscopy of charge separation at the PTCDA/TiOPc heterointerface on Ag(111) — •Marcel Theilen, Alexa Adamkiewicz, Alexander Lerch, Robert Wallauer, and Ulrich Höfer — Philipps-University Marburg, Germany

A key element for charge separation in organic donor/acceptor heterostructures is the formation of charge transfer (CT) states at the interface between the donor and acceptor molecules. A promising model system for studying such excitonic states is the well-defined heterosystem of PTCDA/TiOPc/Ag(111), which we examine by means of time-resolved two-photon photoemission (2PPE). Based on previous 2PPE measurements, the charge carrier dynamics upon selective excitation of excitons in the molecular layers are dominated by transfer processes into the interface state (IS) between molecule and metal. Increasing the molecule-metal distance results in a more effective decoupling to the IS and a possible formation of CT excitons at the donor/acceptor interface. However, the previous 2PPE measurements couldn't access the population of the excited molecular states, which is possible by means of time-resolved photoemission orbital tomography (tr-POT), as it provides a direct insight into the momentum-space dynamics of the excited molecular states. Here we demonstrate our first measurements of the charge carrier dynamics it the 2 ML PTCDA/2 ML TiOPc heterosystem using tr-POT. We follow the momentum-space dynamics of the excited molecular states and see, according to the 2PPE measurements, a strong coupling to the IS between molecule and metal.

O 36.7 Tue 18:00 Poster C

Towards femtosecond momentum microscopy of field-effect gated transition metal dichalcogenides — •Bent van Winger-

den, Jan Philipp Bange, Jonas Pöhls, Wiebke Bennecke, David Schmitt, Daniel Steil, Gijsbert Simon Matthijs Jansen, Thomas Weitz, Marcel Reutzel, and Stefan Mathias — 1. Physikalisches Institut, Georg-August Universität Göttingen, Göttingen, Germany

2D-materials have received great attention due to their astounding optical and electronic properties. Several degrees of freedom (DoF) result in extensive phase diagrams and reveal manifold possibilities for applications in spin-, valley- and optoelectronics. To yield applications in technology, control over these DoF has to be established. Our goal is to study the effect of charge carrier doping by field-effect gating on the band structure and ultrafast exciton dynamics of TMDs in time-resolved momentum microscopy. Previous experimental work visualized the impact of field-effect doping on the electronic band structure for graphene and WSe_2, utilizing static μARPES endstations at synchrotrons [Nguyen *et al.* Nature 572 (2019)]. Here, we will discuss our efforts to study gated 2D-materials with our laboratory based experiment that combines a time-of-flight momentum microscope with a high-repetition rate HHG-beamline [Keunecke *et al.*, Rev. Sci. Ins. 91, 063905 (2020)]. This approach will facilitate the study of ultrafast exciton dynamics and may lead to the discovery of novel relaxation dynamics [Schmitt *et al.*, Nature 608, 499 (2022)].

O 36.8 Tue 18:00 Poster C

Coherent Electron Dynamics Probed by Interferometrically Time- and Angle-Resolved Nonlinear Plasmoemission Spectroscopy with a Birefringent Delay-Line — •Pascal Dreher, Alexander Neuhaus, 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

The early optical response of a solid to an intense light field is governed by single-particle and collective excitations within the electron system. Due to the perturbation, changes in the electron populations occur, and coherent electronic polarizations are excited. Interferometrically time-resolved two-photon photoemission (ITR-2PPE) has been highly successful at disentangling the population and polarization dynamics in various material systems. If instead multi-photon transitions are utilized (ITR-mPPE) the multitude of involved electronic states and their possible interactions complicate the extraction of lifetimes and dephasing times. We combined a birefringent delay-line with nanofocusing of surface plasmon polaritons (SPPs) on flat metal surfaces to acquire phase-locked pulses with transient electric field strengths sufficient to coherently drive highly nonlinear electronic transitions. The excited electron dynamics is observed using time- and angle-resolved photoemission spectroscopy. We discuss modelling of the dynamics with Lindblad equations and comment on the extraction of lifetimes and dephasing times in this highly nonlinear setting.

O 36.9 Tue 18:00 Poster C

100 kHz HHG at 18 eV for time-resolved ARPES — •Sunil Dahiya[1], Hermann Erk[1], Aaron Diercks[1], Stephan Jauernik[1], and Michael Bauer[1,2] — [1]Institute of Experimental and Applied Physics, Kiel University, 24098 Kiel, Germany — [2]Kiel Nano, Surface and Interface Science KiNSIS, Kiel University, 24118 Kiel, Germany

Conventional time- and angle-resolved photoemission (trARPES) employs probe pulses of approximately 6 eV, providing only limited access to momentum space. To encompass a broader momentum space, covering the entire Brillouin zone in solids, higher photon energies are required. This objective is achieved through high harmonic generation (HHG) [1], generating photon pulses with energies in the range of 10 to several hundred electron volts.

In this presentation, we introduce a 100 kHz high-harmonic generation source for trARPES. The concept, including the laser system used, is based on a recently presented setup that produces vacuum ultraviolet (VUV) pulses at a photon energy of 11 eV [2]. In our experiment, we select the next higher harmonic (5th harmonic) of the spectrum at 18 eV photon energy. The HHG source is driven by the 3rd harmonic (350 nm, 0.02 mJ pulse energy) of a Yb:KGW amplifier (1038 nm, 190 fs) and is operated with Argon or Krypton. We demonstrate the performance of the source in initial ARPES measurements of various solid targets.

[1] T. Rohwer, et al., Nature 471, 490 (2011).
[2] C. Lee, et al, Rev. Sci. Instrum. 91, 043102 (2020).

O 36.10 Tue 18:00 Poster C

Dynamics of energy-resolved electron densities at the non-thermal stage in gold — •Markus Uehlein, Christopher Seibel, Tobias Held, Sebastian T. Weber, and Baerbel Rethfeld — Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau

During femtosecond laser excitation, the electron distribution in a metal is disturbed to a state far from equilibrium, i.e. far from a Fermi distribution. The resulting high-energy electrons (hot electrons), e.g., drive chemical reactions at surfaces and are utilized in biosensing methods. An important step for further studies is to understand how many electrons exist in a specific energy range and on what timescale. This is measurable for example with photoemission experiments.

We apply a kinetic model based on full Boltzmann collision integrals to trace dynamics of the electronic distribution [1]. Thereby, the different processes contributing to excitation and thermalization can be investigated independently. We study the temporal evolution of the electron densities in various energy ranges, pointing out the influence of primary and secondary electron generation [2]. Furthermore, we compare the microscopic description of the electron-electron scattering with a relaxation time approach and an extended temperature-based description.

[1] B.Y. Mueller, B. Rethfeld, Phys. Rev. B 87, 035139 (2013)
[2] C. Seibel, M. Uehlein *et al.*, J. Phys. Chem. C (2023) DOI: 10.1021/acs.jpcc.3c04581

O 36.11 Tue 18:00 Poster C

Nonequilibrium optical properties in laser-excited noble metals — •Pascal D. Ndione[1], Tobias Held[1], Sebastian T. Weber[1], Dirk O. Gericke[2], and Baerbel Rethfeld[1] — [1]Department of Physics and Research Center OPTIMAS, RPTU Kaiserslautern-Landau — [2]CFSA, Department of Physics, University of Warwick

We study the electron density and optical responses in gold following excitation with XUV and visible light. We develop multiband rate equations that track the density response in each active electron band [1]. The rate equations also trace the energy content of the *sp*- and *d*-electrons and couple them to the phonons. Moreover, we use a modified Drude-Lorentz approach to calculate the transient dielectric function [2]. Our results show that visible light excitation leads to an overpopulation of the *sp*-band, primarily driven by photo-excitation, while XUV irradiation results in an underpopulation of the *sp*-band, dominated by subsequent impact ionization. Comparison of our simulation for the optical response with pump-probe experiments [1, 3] shows excellent agreement, suggesting a strongly improved understanding of the relevant physical processes and their timescales.

References:
[1] P.D. Ndione et al., Sci. Rep., 12, 4693 (2022)
[2] P.D. Ndione et al., ArXiv, 2307.11874 (2023)
[3] Z. Chen et al., Nat. Commun., 12, 1638 (2021)

O 36.12 Tue 18:00 Poster C

Towards time- and angle-resolved photoemission spectroscopy of plasmon enhanced van-der-Waals heterostructures — •Mattis Langendorf, Marco Merboldt, Jan Philipp Bange, Wiebke Bennecke, Paul Werner, David Schmitt, Jonas Pöhls, Anna Seiler, Thomas R. Weitz, Marcel Reutzel, and Stefan Mathias — Georg-August-Universität Göttingen, I. Physikalisches Institut, Germany

Light matter interaction in metals is mostly determined by a collective excitation of the charge carrier density, a quasiparticle called plasmon. For the case of surface plasmon polaritons (SPPs) that are localized at the surface-vacuum interface, the SPPs can be used to excite excitons in two-dimensional transition metal dichalcogenides (TMDs) that have been exfoliated onto the metal substrate. With this contribution, we work towards the characterization of the ultrafast exciton dynamics in TMDs that have been excited by SPPs. So far, femtosecond momentum microscopy had been used to study the optically excited exciton dynamics in TMDs [1]. Here, our approach is to image the propagating SPPs with photoemission electron microscopy and then to visualize the formation of excitons in the TMD with dark-field imaging techniques [2].

[1] Schmitt, Bange et al., Nature 608, 499-503 (2022).
[2] Schmitt et al. arXiv.2305.18908 (2023).

O 36.13 Tue 18:00 Poster C

Ultrafast Low Energy Electron Diffraction of Layered Materials — •Alp Akbiyik[1], Felix Kurtz[1], Monica Kolek Martinez de Azagra[2], Lukas Jehn[1], Hannes Böckmann[1], Dennis Epp[1], Thomas Weitz[2], and Claus Ropers[1,3] — [1]Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]1st Physical Institute, University of Göttingen, Germany — [3]4th Physical Institute, University of Göttingen, Germany

Enhanced by reduced dimensionality and the formation of heterostructures, van-der-Waals materials including few-layer graphene and transition metal dichalcogenides exhibit a large variety of tunable and correlated phenomena. Studying the non-equilibrium response of such materials, ultrafast surface-sensitive measurement techniques such as angle-resolved photoemission spectroscopy is instrumental in revealing couplings and correlations in the time domain. As a complementary method for surface-sensitive structural dynamics, we employ ultrafast low-energy electron diffraction (ULEED) [1]. In this contribution, we present recent progress in the development and application of this technique for the study of structural phase transformations and lattice thermalization. Moreover, we will discuss progress towards higher versatility of the approach in studying exfoliated samples prepared and transferred in an argon atmosphere.

[1] G. Storeck et al., Structural Dynamics 7, 034304 (2020).

O 36.14 Tue 18:00 Poster C

Photoemission electron microscopy of exciton dynamics in thinfilm TMD materials — •Lina Hansen, Kerstin Harland, Katrin Meier, Arvid Klösgen, and Jan Vogelsang — Institut für Physik, Carl von Ossietzky Universität Oldenburg, 26129 Oldenburg, Germany

Atomically thin layers of transition metal dichalcogenide (TMD) materials offer unique optical properties through their excitonic effects in the reduced dimensions. The resulting enhanced light-matter interactions are based on the arising excitons with long lifetimes being the main mechanism for light emission and recombination processes.

Using a photoemission electron microscope (PEEM) with optical excitation in the visible, near-infrared and extreme ultraviolet spectral region, thinfilm TMD materials are studied. In a first step, our investigations focus on exciton diffusion dynamics at structural interfaces. In particular, we utilize the unique tight focusing capabilities of our set-up to locally excite excitons in the laterally heterogenous TMD material and reveal the spatial behavior of the exciton decay.

O 36.15 Tue 18:00 Poster C

Coherent oscillations in the $4p_{x,y}$ valence band of 1T-$TiSe_2$ — •Jan Böhnke, Mehul Jotshi, Stephan Schmutzler, Cornelius Gahl, and Martin Weinelt — Fachbereich Physik, Freie Universität Berlin, Germany

Upon cooling below T $\sim$ 200 K, 1T-$TiSe_2$ undergoes a structural phase transition which is characterized by a charge redistribution and a periodic lattice distortion. By time-resolved ARPES it is possible to investigate the dynamics of both, electronic band structure and phonon response. Upon excitation with a fs laser pulse with 1.55 eV photon energy we observe fluence-dependent coherent oscillations in the Se $4p_{x,y}$ valence band of $TiSe_2$ which have been previously reported in a time-resolved photoemission experiment [1]. In contrast, no signature of coherent oscillations is found in the backfolded Ti 3d conduction band, suggesting that it is not a sensitive probe for the phase transition. Interestingly, we find image-potential states (n = 1, 2, 3) with lifetimes of up to 300 fs, indicating a low defect density of the surface after in-vacuum cleavage.

[1] S. Duan et al., Nature 595, 239-244 (2021)

O 36.16 Tue 18:00 Poster C

Few electron correlations from nanometric needle tips triggered by femtosecond laser pulses — •Felix Lopez Hoffmann, Jonas Heimerl, Stefan Meier, and Peter Hommelhoff — Department Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), 91058 Erlangen

Metal needle tips are a standard source of free electrons for electron microscopes. When many electrons are emitted from such tips, space charge effects due to Coulomb repulsion appear as intuitively expected [1]. Here we demonstrate that already two electrons repel each other if confined tightly in space and time. In specific, we use femtosecond laser pulses to trigger multiphoton photoemission from metallic needle tips with a few nanometer in radius. The emitted electron pairs show an energy anti-correlation with a visibility of 56%, a mean energy splitting of 3.3 eV and a correlation decay time of 82 fs [2]. Similar results have been obtained in a TEM [3]. By energy filtering the emitted beam the number distribution of electrons per laser pulse can be tuned from Poissonian to sub/super-Poissonian statistics. Sub-Poissonian emission, in our case with a second order correlation $g(2)$ as low as 0.34, allows for shot-noise reduced imaging. Using such Coulomb-induced energy splitting, also heralding of electrons becomes possible. Furthermore, we show first results on spatial correlations stemming from transverse momentum exchange.

[1] Kuwahara et al., Appl. Phys. Lett. 109(1), 013108 (2016)
[2] Meier et al., Nat. Phys. 1-8 (2023)
[3] Haindl et al., Nat. Phys. 19, 1410-1417 (2023)

O 36.17 Tue 18:00 Poster C

An XUV time- and angle-resolved photoemission spectroscopy setup for complementary experiments with high temporal and high energy resolution — •Mohamed Amine Wahada, Tommaso Pincelli, Lawson Lloyd, Alessandro de Vita, Tulio de Castro, Martin Wolf, Ralph Ernstorfer, and Laurenz Rettig — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

Time and angle resolved photoemission spectroscopy (trARPES) is a powerful technique to investigate the transient electronic band structure of materials on a femtosecond timescale. However, due to the Heisenberg's uncertainty principle, the achievable energy resolution is limited by the pulse bandwidth required for a given temporal resolution. Here, we present a new scheme to benefit separately from a high temporal resolution or a high energy resolution within one single experiment operating in the extreme ultraviolet (XUV) regime at 500 kHz. In the first case, 40 fs, 400 nm laser pulses generated by optical parametric chirped pulse amplification are used to generate the XUV. In the second case, 200 fs laser pulses at 515 nm are used. Both beams are guided to a harmonics generation chamber, where a single harmonic at ˜21.7 eV is isolated and guided into the experimental trARPES chamber. The first setup allows high ˜20 fs temporal resolution measurements while the second setup achieves an energy resolution of $<$40 meV. In conjunction with a wavelength tunable pump, this tool allows to perform pump probe ARPES experiment on the same sample granting access to complementary observables.

O 36.18 Tue 18:00 Poster C

A high repetition rate XUV source for time-resolved momentum space mapping of photoelectrons — •Yu Zhang, Adam S. Wyatt, James O. F. Thomson, Richard T. Chapman, Charlotte E. Sanders, and Emma Springate — Central Laser Facility, STFC Rutherford Appleton Laboratory, Research Complex at Harwell, Harwell, United Kingdom

With an ultrashort pulsed extreme ultraviolet (XUV) laser as the photon source, angle-resolved photoemission spectroscopy (ARPES) has extended the study of electronic structure beyond the 3D momentum space and into the temporal domain, with femtosecond resolution. Recently a new high repetition rate (100 kHz) XUV source has been developed in the Artemis facility of Central Laser Facility, UK, to provide an XUV beam source at energies from 20 to 45 eV, with a maximum flux of about 10^{10} photons/second. Together with a tunable laser source, time-resolved ARPES measurements are offered at Artemis with much higher data collection efficiency compared to the previous setup based on a 1 kHz system.

In this poster, the detailed beamline setup will be demonstrated. Besides a pulse-duration preserving XUV monochromator, a demagnification unit has been developed to focus the XUV spot size down to about 20 micrometers at the sample. This new capability allows users to perform experiments on small samples and distinct structural domains of inhomogeneous samples.

O 36.19 Tue 18:00 Poster C

Strong-Field Electron Emission in a Photoemession Electron Microscope (PEEM) — •Kerstin Harland, Lina Hansen, Katrin Meier, Arvid Klösgen, and Jan Vogelsang — Institut für Physik, Carl von Ossietzky Universität, 26129 Oldenburg, Germany

Time-resolved photoemission electron microscopy (PEEM) is well suited for investigating electric fields at nanostructures. Beyond imaging, exerting control on charge carriers with strong optical electric fields on the nanoscale is an ongoing challenge. Space charge constraints so far limit the applicable optical field strengths.

To overcome this problem, we investigate electron emission from a thin, monocrystalline gold flake with our photoemission electron micro-

scope. We write an antenna in the gold flake so that surface plasmons can be excited. To reach the needed high intensity, we focus a 2000-nm wavelength laser beam with few-cycle pulses from our home-built laser system through the backside of the sample onto the gold flake antenna. Compared to conventional PEEM setups, this allows us to obtain a smaller focus size and address individual antennas without space charge problems. The antenna in the gold flake concentrates the optical energy from the laser in a small region, where electrons are emitted. We investigate the emission point and energy spectrum from these electrons with varying laser parameters.

O 36.20 Tue 18:00 Poster C

Impact of the CEP of the driving pulses on High-order Harmonic Generation — •Katrin Meier, Lina Hansen, Arvid Kloesgen, Kerstin Harland, and Jan Vogelsang — Institut für Physik, Carl von Ossietzky Universität Oldenburg, 26129 Oldenburg, Germany

The nonlinear process of High-order Harmonic Generation (HHG) is a method of generating ultrashort pulses in the extreme ultraviolet spectral range. They are of great interest to researchers because they can be used, for example, to probe ultrafast electron dynamics in various structures.

In this work, a home-built laser system is used that delivers ultrashort laser pulses centred around 2000 nm. The intense few-cycle near-infrared pulses are focused into a specially designed gas target where high harmonics of the driving laser field are generated. The carrier-envelope phase (CEP) of the driving pulses is measured with an f-to-2f interferometer. The focus of the work is on the influence of the CEP on the HHG spectrum and the corresponding temporal pulse structure.

O 36.21 Tue 18:00 Poster C

Few-cycle 2 μm light source for high-order harmonic generation at 200 kHz repetition rate — •Arvid Klösgen, Katrin Meier, Niels Cordes, Julia Altenburg, Lina Hansen, and Jan Vogelsang — Institut für Physik, Carl von Ossietzky Universität Oldenburg, 26129 Oldenburg, Germany

Electron microscopy with femtosecond time resolution requires laser sources with high repetition rates beyond 100 kHz to collect sufficient statistics without detrimental space charge effects. Investigations on the attosecond time scale additionally require high pulse energies to permit the generation of attosecond light pulses in a gas jet.

We report on a home-built high repetition rate laser system that includes an ytterbium-based 1030-nm, turn-key pump laser, white light generation, non-collinear optical parametric amplification in the visible range, difference frequency generation and additional optical parametric amplification. The system delivers broadband, few-cycle light pulses at a central wavelength of 2 μm with a pulse energy of 25 μJ and a passively stable carrier-envelope phase. Short- and long-term stability, beam quality, the temporal pulse structure and tunable parameters such as phase matching, time delay and pulse compression are subject of this study.

O 36.22 Tue 18:00 Poster C

Time-of-flight momentum microscopy for subcycle time-resolved photoemission — •Sarah Zajusch, Suguru Ito, Lasse Münster, Jens Güdde, Robert Wallauer, and Ulrich Höfer — Philipps-Universität Marburg

Time-of-flight momentum microscopy enables the simultaneous acquisition of energy and momentum distribution within the complete photoemission horizon. Thus, this technique paves the way to trace for example all exciton formations in TMD materials [1] and orbital images on ultrafast timescales [2]. In order to also investigate subcycle dynamics such as lightwave-driven currents in topological surface bands [3], we have to face additional challenges like photoelectron streaking and multi-hit detection.

Here, we present how photoelectron streaking affects time-of-flight experiments and how it can be compensated in the post processing. In addition, depending on pump frequency and field strength, it can be necessary to include a real-time streaking correction during the running experiment. We also show how multi-hit detection can be realized while maintaining good measurement performance.

[1] R. Wallauer *et al.*, Nano Lett. 21, 13, 5867-5873 (2021).
[2] R. Wallauer *et al.*, Science **371**, 1056-1059 (2021).
[3] S. Ito *et al.*, Nature **616**, 696 (2023).

O 36.23 Tue 18:00 Poster C

EUV pump-probe spectroscopic imaging — •Hannah Strauch[1], Fengling Zhang[2], Bent van Wingerden[1], Daniel Steil[1], Stefan Witte[2], Thorsten Hohage[3], Stefan Mathias[1], and G. S. Matthijs Jansen[1] — [1]University of Göttingen, I. Physical Institute, Göttingen, Germany — [2]Advanced Research Center for Nanolithography, Amsterdam, The Netherlands — [3]University of Göttingen, Institute of Numerical and Applied Mathematics, Göttingen, Germany

Extreme ultraviolet (EUV) spectroscopy accesses element-specific core levels, and thereby enables an element-resolved view of electronic, structural and magnetic dynamics. Likewise, EUV diffractive imaging allows nanoscale imaging of complex structures. The combination of both strengths in femtosecond spectromicroscopy is highly promising, but typically requires long measurement runs as well as a rigorous mathematical treatment of the data.

Here, we address these challenges by combining Fourier-transform spectroscopy and Fourier-transform holography (FTH) in a table-top interferometric high-harmonic generation experiment. From the far-field diffraction pattern of two phase-locked EUV pulses, we reconstruct spectromicroscopic images using an iteratively regularized Gauss-Newton algorithm. This approach implements prior knowledge of the spectral domain and thereby reduces the sampling requirements and measurement time by an order of magnitude. We will report on progress towards time-resolved spectroscopy of light-induced dynamics in exfoliated 2D material nanostructures based on spectroscopic FTH.

O 36.24 Tue 18:00 Poster C

Characterization of THz electric field transients by time-resolved photoelectron emission spectroscopy — •Hamed Abbasi[1], Ping Zhou[1], Florian Diekmann[2], Kai Rossnagel[2], Martin Mittendorff[1], and Uwe Bovensiepen[1] — [1]University of Duisburg - Essen, Germany. — [2]Kiel University, Germany

Energy- and momentum-dependent analysis of THz electric field induced charge carrier dynamics can be achieved on the surface of materials in THz-pump photoemission-probe experiments [1]. Here we report such an experimental setup in which the THz field is generated at 250 kHz repetition rate using a photoconductive antenna [2], with which we obtained a field with 1 THz center frequency and 0.5 ps duration. We have synchronized the THz pulse with an ultrashort UV pulse for time-resolved angle-resolved photoemission spectroscopy. 45meV energy streaking of photoelectrons on the sample surface could be achieved at a cleaved surface of 1T-TaS_2 sample, which is correspond to 6 kV/cm THz electric field. The separation of field induced dynamics in the material and acceleration of the emitted photoelectron in vacuum will be discussed.

We thank the DFG for funding through CRC 1242.

[1] Reimann et al. Nature **562**, 396 (2018).
[2] Dreyhaupt et al. Appl. Phys. Lett. **86**, 121114 (2005).

O 36.25 Tue 18:00 Poster C

Singlet fission and triplet dynamics in pentacene embedded in a surface-anchored metal-organic framework — •Martin Richter[1], Zhiyun Xu[2], Philipp Ludwig[3], Pavel Kolesnichenko[1], Uwe Bunz[3], Christof Wöll[2], and Petra Tegeder[1] — [1]Physikalisch-Chemisches Institut, Universität Heidelberg — [2]Institut für Funktionelle Grenzflächen, Karlsruher Institut für Technologie — [3]Organisch-Chemisches Institut, Universität Heidelberg

It has been observed, that the rate of SF and the lifetime of the generated triplets strongly depend on the molecular arrangement.[1]

Here, a cofacial orientation of pentacene molecules is achieved by embedding them in a surface anchored metal-organic framework (SURMOF). Transient absorption spectroscopy has been used to analyze the ultrafast dynamics as well as long lived states after photoexcitation. The observed difference absorption spectra indicate, that after the initial excitation a singlet excited state generates a correlated triplet pair within a few picoseconds that retains singlet character. Subsequent dynamics show the formation of a long-lived species (40us) with triplet character. This exceeds by far the observed lifetime of triplets generated in pentacene thin films (10ns) and may enhance triplet harvesting capabilities in photovoltaic devices.[2]

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