O 79: Poster: Ultrafast Electron and Spin Dynamics at Interfaces

Time: Wednesday 18:15–20:30 Location: Poster A

O 79.1 Wed 18:15 Poster A

Charge transfer dynamics in monomolecular films: azulene and polyacene backbones, pyridine as a resonantly addressable group — Tobias Wächter[1], Andreas Terfort[2], Mikhail V. Barybin[3], and •Michael Zharnikov[1] — [1]Applied Physical Chemistry, Heidelberg University, 69120 Heidelberg, Germany — [2]Institut für Anorganische und Analytische Chemie, Universität Frankfurt, 60438 Frankfurt, Germany — [3]Department of Chemistry, University of Kansas, Lawrence, Kansas 66045-7582, USA

We review recent progress in the application of core hole clock approach in the framework of resonant Auger electron spectroscopy to the monomolecular assembles to study electron transfer (ET) dynamics in these systems serving as prototypes of molecular electronics devices. The ET pathway to the substrate was unambiguously defined by resonant excitation of the specific tail group (nitrile) attached to the molecular backbone. Characteristic ET times within the femtosecond domain were determined, along with the attenuation factors for the ET dynamics, analogous to the case of the static transport. Three different systems were studied. The first system was 2-mercapto-6-cyanoazulene monolayer which is of importance in view of a possible correlation between the ET dynamics and molecular dipole moment. The second systems were monolayers with the acene backbone which are of importance in view of the existing controversy regarding the attenuation factor for this specific type of molecular films. The third system was a series of pyridine-substituted monolayers where, instead of nitrile, the pyridine moiety was used as the starting point for ET to the substrate.

O 79.2 Wed 18:15 Poster A

Momentum dependent hot electron lifetimes in metals — •Tobias Eul[1], Florian Haag[1,3], Philip Thielen[1,3], Martin Feidt[1], Mirko Cinchetti[2], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,3] — [1]Fachbereich Physik and Research Center OPTIMAS, Technische Universität Kaiserslautern, Germany — [2]Experimentelle Physik VI, Technische Universität Dortmund, Germany — [3]Graduate School MAINZ, Germany

Understanding the correlation between the electronic band structure and the corresponding hot carrier dynamics is of utmost importance to improve the performance of next-generation electronic and spintronic devices. In this context, we will combine momentum microscopy with two-photon photoemission to directly image the momentum dependent electron dynamics of 2D materials and metal-organic systems.

In a first step, we have chosen to study the electron dynamics of noble metals with the combination of our photoemission electron microscope with a time-of-flight detector. The experimental data obtained by time-resolved two-photon momentum microscope can be used to directly translate the transient photoemission yield into intermediate state lifetimes.

For Ag single crystals, we observe a clear momentum dependence of the electron lifetimes for all intermediate state energies. These findings are discussed in the light of static one and two photon momentum microscopy experiments which allow us to characterize the electronic states involved in the optical excitation process.

O 79.3 Wed 18:15 Poster A

Time-domain identification of electronic correlations in 1T-TaS_2 — Manuel Ligges[1], Isabella Avigo[1], Denis Golež[2], Hugo Strand[2], •Yasin Beyazit[1], Kerstin Hanff[3], Florian Diekmann[3], Ljupka Stojchevska[1], Matthias Kalläne[3], Ping Zhou[1], Kai Rossnagel[3], Martin Eckstein[4], Phillip Werner[2], and Uwe Bovensiepen[1] — [1]Faculty of Physics, Universität Duisburg-Essen — [2]Department of Physics, University of Fribourg, 1700 Fribourg, Switzerland — [3]Institute of Experimental and Applied Physics, Universität Kiel, 24098 Kiel — [4]Max Planck Research Department for Structural Dynamics, University of Hamburg-CFEL, 22761 Hamburg

We used time-resolved photoemission spectroscopy to isolate and access fundamental Mott-physics in the (quasi) 2D transition-metal dichalcogenide 1T-TaS_2. Doublon-holon recombination is found to occur on time scales as short as only a few electronic hopping cycles $\hbar/J$. Despite strong electron-phonon coupling, the dynamics can be reproduced within a single-band Hubbard model in the absence of additional bosonic coupling channels. We furthermore find that hole-doping governs the relaxation dynamics on ultrashort time scales.

O 79.4 Wed 18:15 Poster A

Time-resolved momentum microscopy using a high-repetition rate HHG lightsource — •David Schmitt, Marius Keunecke, Christina Nolte, Daniel Steil, Sabine Steil, and Stefan Mathias — I. Physikalisches Institut Göttingen, 37077 Göttingen, Friedrich-Hund-Platz 1

Recent developments in laser technology and electron spectrometers allow for the build-up of a new generation of time-resolved photoelectron emission experiments. With regard to lightsource development, coherent extreme-ultraviolet femtosecond pulses can today routinely be produced via high-harmonic generation at repetition rates up to 1 MHz, which is essential in time-resolved photoemission spectroscopy/microscopy to avoid space charge effects. In addition, new and highly-efficient electron spectrometers are available that map the full parallel momentum space of the electronic structure (ARPES), or can be used as photoemission electron microscopes (PEEM) with several tenths of nanometers spatial resolution. On our poster, we will present first measurements with such a combined time-resolved photoemission setup. We will discuss detection efficiency, space charge effects and energy-, time-, momentum- and spatial resolution that can be achieved.

O 79.5 Wed 18:15 Poster A

Time-resolved photoemission tomography of organic thin films — •Sebastian Hedwig[1], Sebastian Emmerich[1,2], Johannes Seidel[1], Dominik Jungkenn[1], Florian Haag[1], Christina Schott[1], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,2] — [1]Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, Erwin-Schrödinger-Straße 46, 67663 Kaiserslautern, Germany — [2]Graduate School Materials Science in Mainz, Erwin-Schrödinger-Straße 46, 67663 Kaiserslautern, Germany

Photoemission tomography is a powerful method to correlate the momentum resolved photoemission yield of organic thin films to the shape of the molecular orbitals in real space [1]. Using static light sources, this technique has been successfully used to identify the emitting molecular orbitals of metal-organic interfaces or to determine the density of states of different molecules in heteromolecular structures [2]. Here, we extend this approach to the ultrafast timescale. By combining momentum microscopy with fs-XUV light sources, we are able to directly image the time-resolved evolution of occupied molecular orbitals and to access the momentum space pattern of unoccupied molecular orbitals. Here, we present our first results of the optically induced transient behavior of the momentum space signature of molecular orbitals of a C_{60} thin film. These results will be analyzed in the photoemission tomography approach to gain insight into transient changes of the molecular orbital during and after the optical excitation.

[1] Science 326 702 (2009), [2] Nat. Commun. 5, 3685 (2014)

O 79.6 Wed 18:15 Poster A

High-repetition rate extreme ultraviolet HHG light source for femtosecond surface science experiments — •Christina Nolte[1], Amelie Schulte[1], Marco Merboldt[1], Germaine Arend[1], Steffen Hädrich[2], Tino Eidam[2], Jens Limpert[2], Sabine Steil[1], Daniel Steil[1], and Stefan Mathias[1] — [1]1st Physical Institute, University of Göttingen — [2]Active Fiber Systems GmbH, Jena

Table-top coherent ultrashort extreme ultraviolet light sources from high-harmonic generation have been shown to enable a wealth of novel experiments in the field of ultrafast surface science [1,2]. However, the full potential of this approach has not yet been achieved because, to date, high harmonics generated by low-repetition rate Ti:sapphire lasers required a trade-off between photon flux, repetition rate, energy and time resolution [3]. Here, we present a HHG light source driven by a nonlinearly compressed 0.5 MHz fiber laser enabling $> 10^{10}$ photons/s in single harmonics between 22-73 eV. In addition, our setup enables the direct generation of harmonics with bandwidths of the order of about 50 meV, so that the light source is ideally suited for next generation femtosecond spin- and angle-resolved photoemission experiments.

[1] Rohwer et al., Nature 471, 490 (2011)

[2] Mathias et al., JESRP 189, 164 (2013)
[3] Eich et al., JESRP 195, 231 (2014)

O 79.7 Wed 18:15 Poster A

Development of a tabletop femtosecond broadband soft X-ray source — •Oscar Naranjo-Montoya, Manuel Bridger, Alexander Tarasevitch, and Uwe Bovensiepen — Universität Duisburg-Essen, Fakultät für Physik, 47057 Duisburg, Germany

Pump-probe experiments with optical excitation and absorption spectroscopy in the soft X-ray range on a femtosecond timescale are useful to analyze electronic and magnetic properties of complex material systems and their temporal evolution. These experiments have become available at Free Electron Laser and Synchrotron sources. However, due to the limited accessibility of these large scale facilities, systematic studies remain arduous to perform. Therefore, it will be useful to develop compact laboratory based sources. A promising approach is to use high harmonic generation in noble gases driven by mid-infrared femtosecond pulses. It has already been shown that with such radiation it is possible to generate photon energies that reach the keV regime [1]. We present the development of a high power two stage Optical Parametric Chirped-Pulse Amplifier as a pump for a tabletop soft X-ray source dedicated to absorption spectroscopy. 1 ps pump pulses at 515 nm amplify seed pulses at 800 nm by a factor of 20000. Their wavelength is converted to 1500 nm with a bandwidth of 40 nm and energy of 10 mJ. The 1500 nm pulses are the seed to produce λ=3200 nm with 100 fs duration. The achieved spatial, spectral and temporal pulse characteristics and an estimate of the soft X-ray flux will be presented. This work is funded by the DFG through SFB 1242, TP A05. [1] T. Popmintchev et al., Science **336**, 1287-1291 (2012).

O 79.8 Wed 18:15 Poster A

A novel setup for XUV-based time-resolved ARPES at 500 kHz repetition rate — M. Puppin, C. Nicholson, P. Xian, W. Windsor, Y. Deng, J. Feldl, C. Monney, M. Wolf, •L. Rettig, and R. Ernstorfer — Fritz-Haber-Institut der MPG, Faradayweg 4-6, 14195 Berlin

We present a novel setup for time- and angle-resolved photoemission spectroscopy (trARPES) based on ultrashort extreme ultraviolet (XUV) laser source with 0.5 MHz repetition rate. The source is driven by an optical parametric chirped pulse amplifier (OPCPA) based on a hybrid fiber-slab amplifier system. The near-infrared OPCPA output is frequency-upconverted sequentially: frequency-doubling to 3.1 eV photon energy is followed by 7th harmonic generation (21.7 eV) in a dense noble gas jet. After spectral filtering, an XUV pulse with 20 fs pulse duration, a bandwidth of $\sim$ 100 meV and a photon flux exceeding 10^{11} photons/s is available for trARPES.

We demonstrate the capabilities of the setup by mapping the excited state structure of bulk WSe_2 100 fs after optical excitation with 3.1 eV photons throughout the Brillouin zone and compare the results to DFT band structure calculations. Furthermore, in $TbTe_3$ we investigate the closing of the charge density wave energy gap and unfolding of the band structure in both occupied an unoccupied states after excitation with 1.55 eV photons. Our experimental setup allows the observation of occupied and transiently populated unoccupied states and their dynamics in the whole Brillouin zone, and the determination of direct and indirect band gaps within a single experiment.

O 79.9 Wed 18:15 Poster A

Time-resolved time-of-flight momentum microscopy at femtosecond lab sources — •Eva Sophia Walther[1], Tobias Eul[1], Florian Haag[1,2], Katerina Medjanik[3], Gerd Schönhense[3], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,2] — [1]Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, Germany — [2]Graduate School of Excellence Materials Science in Mainz, Germany — [3]Institut für Physik, Johannes Gutenberg-Universität Mainz, Germany

Time and angle-resolved photoemission spectroscopy (trARPES) is one of the most important tools to investigate ultrafast carrier dynamics in solids. The largest challenge in trARPES is the small angular acceptance of conventional hemispherical electron analysers which only allows one to image a small part of the Brillouin zone. This limitation can be overcome by momentum microscopy, as recently shown at the free-electron laser FLASH [1]. This technique uses a set of parallel-imaging optics designed for best resolution in k-space. In combination with ToF data recording it allows to collect the photoelectron distribution in the full hemisphere above the sample surface and the complete d-band complex in a single acquisition. Here, we present the first results obtained with our newly commissioned ToF-momentum microscope which was combined with fs-laser light sources. The status of the development will be demonstrated by first 2PPE momentum microscopy data obtained for noble metal surfaces and ferromagnetic thin films. Finally, we will discuss possibilities for ToF-momentum microscopy with fs-XUV light sources. [1] H.-J. Elmers et al., this conference

O 79.10 Wed 18:15 Poster A

Photoexcitation of doublons in 1*T*-TaS_2 — •Y. Beyazit[1], J. Beckord[1], I. Avigo[1], M. Ligges[1], M. Kalläne[2], P. Zhou[1], K. Rossnagel[2], and U. Bovensiepen[1] — [1]Faculty of Physics, University of Duisburg-Essen, Duisburg, Germany — [2]Institute of Experimental and Applied Physics, University of Kiel, 24098 Kiel, Germany

Layered two-dimensional materials such as transition metal dichalcogenides exhibit rich phase diagrams. 1*T*-TaS_2 is metallic at high temperatures, while below 180 K it becomes an insulator and the topmost electronic band splits due to a commensurate periodic lattice distortion into an occupied lower (LHB) and an unoccupied upper Hubbard band (UHB), separated by a 350 meV energy gap. The UHB can be populated by generating doublons with femtosecond laser pulses whose photon energy (1.55 eV) largely exceeds the electronic band gap, raising questions on the underlying excitation mechanism [2-4]. Using femtosecond time- and angle-resolved photoemission with different pump photon energies we find that upon excitation with 1.55 eV photons the UHB is observed, pumping with 0.92 eV and 1.04 eV results in a suppression of the UHB signature. We conclude that for the generation of a doublon population in the UHB a resonant dipole transition from an initial state is required, which is not the case for pumping with the lower photon energy. This confirms a dipole-like excitation scheme proposed recently [4].

[1] Sipos et al., Nature Mater. **7**, 960 (2008), [2] M. Ligges et al., arXiv:1702.05300 (2017), [3] N. ten Brinke et al., PRB **95**, 195123 (2017), [4] A. Mann et al., PRB **94**,115122 (2016)