O 37: Poster: Ultrafast Electron Dynamics at Surface and Interfaces II

Time: Tuesday 18:00–20:00 Location: P2/EG

O 37.1 Tue 18:00 P2/EG

Probing alternative pathways for electron transfer across a monomolecular film — Saunak Das[1], Zhiyong Zhao[1], Takanori Fukushima[2], Andreas Terfort[3], and •Michael Zharnikov[1] — [1]Angewandte Physikalische Chemie, Universität Heidelberg, 69120 Heidelberg, Germany — [2]Laboratory for Chemistry and Life Science, Institute of Innovative Research, Tokyo Institute of Technology, Yokohama 226-8503, Japan — [3]Institut für Anorganische und Analytische Chemie, Johann Wolfgang-Goethe-Universität Frankfurt, 60438 Frankfurt am Main, Germany

Charge transport (CT) across a two-dimensional molecular assembly can involve alternative pathways. Using specifically designed monomolecular films we probe (i) competition of intramolecular and intermolecular pathways in a molecular assembly and (ii) competition of different intramolecular pathways within a single molecule. For this purpose, we apply so-called core-hole-clock approach in the framework of resonant Auger electron spectroscopy, allowing the measurement of the characteristic CT time from the terminal tail group of the assembled molecules to the substrate. We show that the intramolecular CT is generally preferable and so-called matrix effects play a negligible role for CT, strongly favoring the through-bond CT model. In the case of availability of several alternative pathways within an individual molecule, a pathway with the highest conductance becomes highly dominant, while other pathways contribute minorly to the entire CT.

O 37.2 Tue 18:00 P2/EG

Pump-probe second harmonic spectroscopy of molecule/metal interfaces — Jinghao Chen[1], Rui Shi[2], Ping Zhou[1], Uwe Bovensiepen[1], Wolfgang Hübner[2], Georg Lefkidis[2], and •Andrea Eschenlohr[1] — [1]Faculty of Physics and CENIDE, Uni Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany — [2]Department of Physics, TU Kaiserslautern, Box 3049, 67653 Kaiserslautern, Germany

Achieving a microscopic understanding of charge transfer dynamics and the relaxation of optically excited electrons and holes at molecule/metal interfaces requires an interface-sensitive analysis on the respective femtosecond timescales. Second harmonic spectroscopy (SHS) is such an interface-sensitive probe in centrosymmetric materials. We perform pump-probe SHS in the visible wavelength range (1.9-2.5 eV) with <20 fs pulse duration. A prototypical molecule/metal interface is prepared by adsorption of iron octaethylporphyrin (FeOEP) molecules on Cu(001) and analyzed *in situ* in ultrahigh vacuum. By comparison with first principles theory, we identify a molecule-induced resonance at about 2.2 eV fundamental photon energy in the SHS of one monolayer of FeOEP/Cu(001). At this resonance, we observe a markedly slower relaxation time of the pump-induced changes in SHS compared to the bare Cu(001) surface, which indicates an increased lifetime of the electronic molecular state.

We thank H. Wende, J. Güdde and E. Riedle for valuable experimental advice, and the German Research Foundation for funding via SPP 1840 QUTIF and Sfb 1242.

O 37.3 Tue 18:00 P2/EG

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

The nonlinear optical technique of second harmonic generation (SHG) is a surface- and interface-sensitive tool in centrosymmetric materials, which is used to study electron dynamics at metal surfaces. We characterize the polarization and wavelength dependent SHG on a Cu(001) surface using a fundamental beam in the wavelength range of 500 - 700 nm. Polarization dependent measurements show the p-P SHG yield is almost a factor of 1.5 larger than s-P SHG, because more non-zero susceptibility tensor elements are involved in the former and the intrinsic absolute value of $|\chi^{(2)}_{zzz}|$ is much larger than the only relevant component $|\chi^{(2)}_{zxx}|$ in the latter. We also measure the spectral dependence of the SHG intensity, which shows an increase up to a maximum at 2.33 eV. Since the 3d band peak of copper lies about 2.3 eV below Fermi level, a fundamental beam at this photon energy can resonantly excite 3d electrons to the Fermi level, which largely increases the overall probability of SHG processes. Moreover, we discuss electron dynamics analyzed through pump-probe SHG at on and off-resonant photon energies.

O 37.4 Tue 18:00 P2/EG

Formation mechanism of defect levels in rutile TiO_2(110) — •Xiang Zhang, Lukas Gierster, and Julia Stähler — Humboldt-Universität zu Berlin, Institut für Chemie

The electron dynamics at TiO_2 surfaces have been widely studied as TiO_2 is a prototypical photocatalyst. Recent studies have specifically addressed the Band Gap State (BGS) at 0.8 eV below the Fermi level and have revealed its role as a trapping center for electrons in the conduction band of rutile TiO_2(110) (with a trapping time of around 45 fs) [1]. However, the origin of the BGS itself remains debated. Using time-resolved photoelectron spectroscopy, we show here that the BGS must be - at least partially - due to long-lived (>5 μs) photoexcited charge carriers which form a photostationary state in pump-probe experiments. The potential formation mechanism of the BGS and implications of this state for previous time-resolved experiments are discussed.

Reference:
[1] Zhang et al. J. Phys. Chem. Lett. 10(52) (2019).

O 37.5 Tue 18:00 P2/EG

Investigation of low-temperature electron emission properties from sharp needle tips — •Manuel Knauft, Stefan Meier, Norbert Schönenberger, and Peter Hommelhoff — Department Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), 91058 Erlangen

Spectroscopic and microscopic techniques benefit from the small linewidth of coherent electron sources with high brightness. Reducing the operating temperature of the emitter is known to positively influence coherence of field emitted electron beams. This is especially true for superconducting needle tips as shown for niobium [1].

We present an experimental setup capable of cooling electron emitters to liquid helium temperature in ultra-high vacuum. Irradiation of samples with ultrashort laser pulses is possible through optical access to the chamber. Detection of charged particles is realized by a multichannel plate. First results for sharp needle tips of various materials are shown. In particular, we investigate the emission characteristics and compare them for field emission and laser triggered emission as a function of temperature.

[1] Nagaoka *et al.*, Nature **396**, 557 (1998)

O 37.6 Tue 18:00 P2/EG

Magnetic field effects on ultrafast driven electrons in topological insulator surface states — •Alexander Riedel[1], Vanessa Junk[1], Wolfgang Hogger[1], Cosimo Gorini[2], and Klaus Richter[1] — [1]Institut für Theoretische Physik, Universität Regensburg, Germany — [2]Universite Paris-Saclay, CEA, CNRS, SPEC, 91191, Gif-sur-Yvette, France

When strong-field light pulses of various durations and shapes interact with a solid, their electric field component acts as an a.c. bias accelerating electrons through the bandstructure and driving non perturbative transitions. These processes can lead to high-order harmonic emission of radiation which encodes characteristic properties of the underlying material.

However, in the vast majority of cases the magnetic field component of the pulse is completely neglected in theoretical descriptions. In this contribution we incorporate a magnetic field into the description by adding a Zeeman term into an effective minimal model Hamiltonian describing the surface states of the topological insulator material bismuth telluride. We present how magnetic field components influence the Berry curvature and the transitions induced by the pulse. The resulting currents and higher harmonic spectra are discussed and compared to those in the absence of magnetic field effects. All statements rely on both a semiclassical analysis and full quantum mechanical simulations.

O 37.7 Tue 18:00 P2/EG

Use cases for Picosecond Ultrasonics with X-rays (PUX) — •Alexander von Reppert[1], Maximilian Mattern[1], Steffen

Zeuschner[1,2], Jan-Etienne Pudell[1,2,3], Marc Herzog[1], and Matias Bargheer[1,2] — [1]Institut für Physik und Astronomie, Universität 14476 Potsdam, Potsdam, Germany — [2]Helmholtz-Zentrum Berlin, 12489 Berlin, Germany — [3]European XFEL, 22869 Schenefeld, Germany

Most studies of picosecond ultrasound have been and will be conducted with flexible all-optical setups, but here we discuss scenarios where ultrashort hard x-ray probe pulses excel. We focus on the extraction of the strain response from Bragg peak shifts in the symmetric diffraction condition for layered, nanoscopic structures upon excitation of metallic transducers with femtosecond laser pulses. This type of experiment can yield direct, layer-specific and quantitative information on the shape and amplitude of picosecond strain pulses and the quasi-static strain. The strain response may serve as a proxy for the local energy-density and temperature rise. The presented use-cases encompass ultrahin as well as opaque metal-heterostructures, nanostructures and negative thermal expansion materials, that each pose a challenge to established all-optical techniques.

O 37.8 Tue 18:00 P2/EG

Photocatalysis on anataseTiO2(101) in real-time — •Heshmat Noei[1,2], Michael Wagstaffe[1], Adrian Dominguez-Castro[3], Lukas Wenthaus[1], Steffen Palutke[1], Dmytro Kutnyakhov[1], Michael Heber[1], Federico Pressacco[1], Sirhei Dziarzhytski[1], Helena Gleissner[1], Verena Kristin Gupta[1], Adriel Dominguez[1], Thomas Frauenheim[3], Angel Rubio[4], and Andreas Stierle[1] — [1]Deutsches Elektronen-Synchrotron (DESY), Hamburg, Germany — [2]The Hamburg Centre for Ultrafast Imaging, Hamburg, Germany — [3]Bremen Center for Computational Material Science (BCCMS), D-28359, Bremen, Germany — [4]Max Planck Institute for the Structure and Dynamics of Matter, D-22761, Hamburg, Germany

Obtaining mechanistic insight surrounding interfacial charge transfer and the role of intermediate species during photocatalysis on metal oxide surfaces is challenging due to their ultrafast nature. By applying ultra-fast optical pump-soft X-ray probe experiments at FLASH in DESY, we have obtained first insight into the activation mechanism of CO photooxidation to CO2 on rutile and anatase TiO2 surfaces and charge transfer at the interfacxe of water and TiO2(101). By using an optical laser of 800 nm and a FEL energy of 647.8 eV, the changes in the Ti 2p, O 1s and C 1s core levels have been monitored on a picosecond timescale.

O 37.9 Tue 18:00 P2/EG

Ultrafast UV pulse generation at the SXP instrument of the European XFEL — •Ekaterina Tikhodeeva[1], Patrik Grychtol[1], Marcus Seidel[2], Christoph M. Heyl[2], Vahagn Vardanyan[1], David Doblas-Jimenez[1], and Manuel Izquierdo[1] — [1]European XFEL, Schenefeld, Germany — [2]DESY, Hamburg, Germany

At the European XFEL a new instrument, the Soft X-ray Port (SXP), is currently under commissioning. Located downstream of the SASE 3 soft X-ray undulator system, it will provide femtosecond photon pulses with variable polarization in the energy range between 260 eV and 3000 eV at MHz repetition rates. Up to 10^{12} photons per pulse will be focus at the interaction region in a micrometer spot size resulting in an intensity of more than 10^{18} W/cm^2. The energy range will enable the complete electronical, chemical and atomic characterization of solids, surfaces and interfaces using femtosecond time-resolved soft X-ray photo-electron spectroscopy. Moreover, two powerful pump lasers operating in a broad spectral range from the mid-infrared to the ultraviolet region will be available. Herriott multi-pass cells (HMPC) will be used to compress their pulses into the few femtosecond range paving the way for ultrafast pump-probe investigations at the SXP instrument combining intense and tunable soft X-rays with the versatile optical lasers. In this contribution, the development of the HMCP and the optical laser system will be presented.

O 37.10 Tue 18:00 P2/EG

Transferring photonic orbital angular momentum to metals — •Jannis Lessmeister[1], Tobias Eul[1], Eva Prinz[1], Sebastian Hedwig[1], Martin Aeschlimann[1], and Benjamin Stadtmüller[1,2] — [1]Department of Physics and Research Center OPTIMAS, TU Kaiserslautern, Germany — [2]Institute of Physics, Johannes Gutenberg University Mainz, Germany

Optical fields can carry an orbital angular momentum (OAM) in helical beams with an azimuthal phase dependence. Since the discovery of this phenomenon in 1992 [1], the interaction of photonic OAM with matter has become a steadily growing field of research. After an initial focus on atoms and molecules, in the last years, scientists also began investigating solids [2]. For example, recent studies revealed that the so-called twisted light can drive photocurrents [3] and affects ultrafast demagnetization dynamics [4].

In our research, we apply time- and angle-resolved photoelectron spectroscopy (TR-ARPES) to gain new insight into the influence of photonic OAM on the hot carrier dynamics in metals. Specifically, we investigated the influence of the added orbital angular momentum on the optical excitation efficency of photoelectrons from the spin-split band structure of Rashba surface states and the hot carrier dynamics of spin-dependent carriers in the ferromagnet Ni.

[1] Allen et al., Phys. Rev. A 45 (1992)
[2] Quinteiro Rosen et al., Rev. Mod. Phys 94 (2022)
[3] Ji et al., Science 368 (2020)
[4] Prinz et al., arXiv:2206.070502 (2022)

O 37.11 Tue 18:00 P2/EG

Combining THz-ARPES and THz-HHG to study electrical currents at buried interfaces of topological insulators — •Tim Bergmeier, Suguru Ito, Jens Güdde, and Ulrich Höfer — Fachbereich Physik, Philipps-UniversitätMarburg, Germany

Angle-resolved photoemission spectroscopy (ARPES) combined with terahertz (THz) excitation has shown to provide a subcycle-resolved momentum space view of lightwave-driven Dirac currents in the surface band of topological insulators [1]. At high electric field strength, these currents have shown to give rise to a unique type of efficient high-harmonic generation (HHG) that reflects the unusual transport properties of the Dirac electrons [2]. As a pure optical technique, THz-HHG is able to access also deeply buried interfaces, but lacks band structure information.

Here, we present an experimental setup which combines both techniques in order to study first the impact of thin protection layers on the current transport in a topologically protected band. The correlation of the time-resolved band structure information provided by THz-ARPES with the properties of THz-HHG will then allow us to apply the latter for thick layers as will be required for device applications. The setup includes the generation of THz-pulses with field strengths up to 10 MV/cm over a frequency range of 12-80 THz, and of ultrashort 400-nm probe pulses for two-photon photoemission with subcycle time resolution at a repetition rate of 200 kHz.

[1] J. Reimann *et al.*, Nature 562, 396 (2018).
[2] C. P. Schmid *et al.*, Nature 593, 385 (2021).

O 37.12 Tue 18:00 P2/EG

A narrow bandwidth extreme ultra-violet light source for time- and angle-resolved photoemission spectroscopy — Qinda Guo[1], Maciej Dendzik[1], Antonija Grubišić-Čabo[1], Magnus H. Berntsen[1], Cong Li[1], •Wanyu Chen[1], Bharti Matta[2], Ulrich Starke[2], Björn Hessmo[1], Jonas Weissenrieder[1], and Oscar Tjernberg[1] — [1]Department of Applied Physics, KTH Royal Institute of Technology, Hannes Alfvéns väg 12, 114 19 Stockholm, Sweden — [2]Max Planck Institute for Solid State Research, Heisenbergstraße 1, 70569 Stuttgart, Germany

Here, we present a high repetition rate, narrow bandwidth, extreme ultraviolet photon source for time- and angle-resolved photoemission spectroscopy. The narrow bandwidth pulses $\Delta E = 9$, 14, and 18 meV for photon energies $h\nu = 10.8$, 18.1, and 25.3 eV are generated through high harmonic generation using ultra-violet drive pulses with relatively long pulse lengths (461 fs). The high harmonic generation setup employs an annular drive beam in tight focusing geometry at a repetition rate of 250 kHz. Photon energy selection is achieved by a spherical focusing grating, which provides high efficiency photon flux with only a small amount of focus size increase (~30%) and temporal broadening (6.8%). A two stage optical-parametric amplifier provides $<$ 100 fs tunable pump pulses from 0.65 μm to 9 μm. Combined with a time-of-flight electron analyzer, the setup enables for high-resolution studies of ultrafast dynamics over the whole surface Brillouin zone in most quantum materials.

O 37.13 Tue 18:00 P2/EG

Time Resolved Photoelectron Spectroscopy of Thiophen based Conjugated Donor Aceptor Polymers for Organic Photovoltaic — •Tobias Reiker[1], Pradny Phanse[1], Nils Fabian

Kleimeier[1], Zitong Liu[2], Adrian Urban[1], Deqing Zhang[2], and Helmut Zacharias[1] — [1]Center for Soft Nanoscience, University of Münster, Germany — [2]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. We report results of temporally resolved photoemission studies on thiophene polymers on silicon substrates. The charge transport behaviour can be tuned by different alkyl side chains since they influence the electronic structure and aggregation. A direct assessment of the intramolecular and intermolecular dynamics may guide synthesis routes. With pF8T2, pDPP4T, pDPP4T-1 and pDPTTT we investigated the electronic dynamics of high hole-mobility organic semiconductors. Either the backbone or the side chains were modified. With the blend of pDPP4T-1 and a Fullerene we compare the excitation dynamics of singe component und blended OPV. These different molecular configurations are intended to provide insights into the change in electron configuration due to both backbone modification and intermolecular packing.