MA 26: Ultrafast Electron Dynamics II (joint session O/MA)

Time: Tuesday 10:30–13:30 Location: WIL B321

Invited Talk MA 26.1 Tue 10:30 WIL B321

Attosecond coherent manipulation of electrons in tunneling microscopy — •Manish Garg[1] and Klaus Kern[1,2] — [1]Max Planck Institute for Solid State Research, Heisenbergstraße 1, 70569 Stuttgart, Germany — [2]Institut de Physique, Ecole Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland

We demonstrate coherent manipulation of electrons in a tunnel junction of a scanning tunneling microscope, by tuning the carrier-envelope-phase (CEP) of two-cycle long (< 6 fs) optical pulses. We explore two different tunneling regimes at the tunnel junction, photon and field-driven tunneling and demonstrate transition from one to the other regime. Spatially localized and atomically strong electric fields of strength ~ 1V/Å substantially modulate the tunneling barrier on attosecond timescales, hence allowing taming of flow of electrons to either side of the tunnel junction. Capability to tune CEP with precision of less than 0.1π enables manipulation of electron tunneling at timescales of ~ 200 as. The strong atomic confinement of tunneling current induced by laser pulses enables optical-field driven tunneling microscopy. Real-time tracing of decay dynamics of oscillations of quasiparticles (localized-surface plasmon) in a gold nanorod is studied with a nanoscale probe in tunneling contact; enabling concurrently angstrom-scale and sub-fs resolution. We expect our results to enable inducing, tracking, and controlling electronic current at atomic scales and pave the way to petahertz coherent nanoelectronics and microscopy.

MA 26.2 Tue 11:00 WIL B321

Atomically resolved femtosecond pump probe measurements on Ta2NiSe5 — •Lukas Arnhold[1], Gregory McMurtrie[1], Shaoxiang Sheng[1], Mohamad Abdo[1,2], and Sebastian Loth[1,2] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Max Planck Institute for Solid State Research, Stuttgart, Germany

The investigation of new and interesting phases in solids is one of the major goals of condensed matter physics. Ta2NiSe5, a putative excitonic insulator, is a material in which the exciton binding energy can exceed the band gap energy, leading to spontaneous condensation of excitons [1] following Bose-Einstein-statistics.

In such a material, it would be particularly interesting to locally break individual excitons and observe the re-condensation into the ground state.

To this end, we combine scanning tunneling microscopy (STM) with ultrafast THz light pulses. This boosts the STM's time resolution to the femtosecond range [2] making it possible to observe the fast electronic dynamics of correlated ground states in real space.

In static measurements, as we decrease the tunnel resistance, we observe a closing of the excitonic band gap. This closing coincides with an unusual increase of THz current in pump probe measurements. We study the dynamic response with atomic spatial resolution in particular around atomic defects in the surface layer.

[1] Lu, Y. et al., Nat Commun 8, 14408 (2017) [2] Cocker, T. et al., Nature Photon 7, 620*625 (2013)

MA 26.3 Tue 11:15 WIL B321

Ultrafast nano-imaging of the order parameter in a structural phase transition — •Thomas Danz, Till Domröse, and Claus Ropers — 4th Physical Institute – Solids and Nanostructures, University of Göttingen, Germany

Over the past decades, ultrafast optical techniques have considerably shaped our understanding of homogeneous materials, while transmission electron microscopy has greatly contributed to elucidating atomic structures and compositions on the sub-nanometer scale. Combining these concepts, ultrafast transmission electron microscopy allows for resolving femtosecond dynamics in heterogeneous materials using imaging, diffraction, and spectroscopy [1].

Here, we employ the Göttingen Ultrafast Transmission Electron Microscope (UTEM) [2] to demonstrate the ultrafast real-space mapping of the order parameter for a charge-density wave phase transition in the correlated material 1T-TaS_2. Specifically, we track the evolution of domain patterns on femtosecond to picosecond time and nanometer length scales, extracting characteristic observables not accessible by ultrafast electron or x-ray diffraction.

Additionally, we show that prominent features in the spatio-temporal domain evolution can be modeled in a time-dependent Ginzburg-Landau approach, allowing us to distinguish different regimes of the observed dynamics.

[1] A. H. Zewail, Science **328**, 187 (2010).
[2] A. Feist, Th. Danz *et al.*, Ultramicroscopy **176**, 63 (2017).

MA 26.4 Tue 11:30 WIL B321

Visualisation of coherent phonons in Bi_2Se_3 by time-resolved photoelectron diffraction — •Klara Volckaert[1], Davide Curcio[1], Dmytro Kutnyakhov[2], Steinn Agustsson[3], Kevin Bühlmann[4], Federico Pressacco[2], Michael Heber[2], Siarhei Dziarzhytski[2], Harald Redin[2], Yves Acremann[4], Jure Demsar[3], Wilfried Wurth[2], Charlotte E. Sanders[5], and Philip Hofmann[1] — [1]Aarhus University, Aarhus, Denmark — [2]DESY, Hamburg, Germany — [3]Johannes Gutenberg-University, Mainz, Germany — [4]ETH Zürich, Zürich, Switzerland — [5]Rutherford Appleton Laboratories, Harwell, United Kingdom

We have developed X-ray photoelectron diffraction (XPD) as a pump-probe technique allowing for the visualisation of structural changes on femtosecond timescales. We use this new technique to observe the structural dynamics of Bi_2Se_3 when excited by a 800 nm optical pump pulse. Terahertz oscillations of the fine structure within the resulting XPD pattern were observed, which could originate from the excited A_1g coherent phonons. These experiments were carried out at the FLASH free electron laser using a time-of-flight momentum microscope, which allows for simultaneous mapping of the emission angles in addition to kinetic energy of the electrons during a photoemission experiment (see D. Kutnyakhov et al. arXiv:1906.12155 (2019)).

MA 26.5 Tue 11:45 WIL B321

Radio frequency controlled electron pulses for time-resolved LEED — •Dennis Epp, Marcel Möller, Gero Storeck, and Claus Ropers — IV. Physical Institute, University of Göttingen, Friedrich-Hund-Platz 1, 37077 Göttingen

Solid state surface systems display complex structural and electronic phases, with properties that may drastically differ from the bulk [1]. The coupling between electronic, lattice and spin degrees of freedom can be studied by ultrafast techniques. The recently developed method of Ultrafast Low-Energy Electron Diffraction (ULEED) is suitable for studying such structural dynamics on surfaces [2,3,4]. In this stroboscopic method, miniaturised laser-driven photoelectron sources generate ultrashort low-energy electron pulses to probe pump-induced changes to the surface structure [2,3], with a temporal resolution down to 1 ps. This contribution will focus on the characterization and control of low-energy electron beams by radio-frequency fields. First measurements of the beam properties resolved by a streaking field and further strategies for pulse compression will be discussed. [1] J. M. Kosterlitz. & D. J. Thouless, J. Phys. C 6, 1181*1203 (1973). [2] G. Storeck et al., Structural Dynamics 4, 044024 (2017). [3] S. Vogelgesang, et al., Nature Physics 14,184-190 (2018). [4] G. Horstmann et al., in preparation.

MA 26.6 Tue 12:00 WIL B321

VUV user station for femtosecond transmission, reflectivity and ellipsometry experiments — •Shirly Espinoza — ELI Beamlines. Institute of Physics. Czech Academy of Science. Czech Republic

Here, we present a versatile experimental platform, located in ELI Beamlines facility in the Czech Republic, dedicated to ultrafast pump-probe VUV absorption, transmission and ellipsometry with time resolution about 100 fs [1]. The whole system is based on a 30 mJ, 15 fs, 1 kHz in-house-developed laser with central wavelength 830 nm [2]. Its pulses are subsequently transformed into the desired pump and probe beams.

The platform is enclosed in an ultrahigh vacuum chamber equipped with reflective polarizing optics. This setup is equipped with a cryostat for measurements at temperatures from 20 K to 350 K. The upcoming upgrade with switchable Helmholtz coils will enable the experiments in magnetic field up to 1.5 T at 1 kHz. We present the experimental details of this cutting edge platform and discuss the possibilities that external scientists have to carry on their measurements on it.

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[1] S. Espinoza et al., Appl. Surf. Sci. 421, 378-382 (2017)
[2] F. Batysta et al., Opt. Express 24, 17843-17848 (2016)

Invited Talk MA 26.7 Tue 12:15 WIL B321

Ultrafast dynamics of charge transfer and Frenkel excitons in molecular thin films — •Benjamin Stadtmüller — University of Kaiserslautern and Research Center OPTIMAS, Erwin-Schroedinger-Str. 46, 67663 Kaiserslautern, Germany

Molecular complexes are highly flexible materials with intriguing opportunities for future photovoltaic and spintronic applications. The crucial device-relevant processes in molecular materials are the excited state dynamics and the carrier transport. Despite their common origin - the molecular band structure - their interplay is far from being understood. In this work, we focus on the link between the excited state dynamics of excitons in fullerene thin films and the resulting ensemble dynamics of the transport states of the entire film using time- and momentum resolved photoemission with fs-XUV radiation. Upon the optical excitation of excitons in C_{60} films, we reveal a transient modification of the energy level alignment of the molecular valence states, which can be identified as the signature of charge transfer (CT) excitons in molecular materials [1]. Taking advantage of this observation, we are able to disentangle the dynamics of CT and Frenkel excitons in (endohedral) fullerene films. We find different decay dynamics depending on the sample temperature or the charge doping concentration of the molecular film. Finally, we will provide a first view onto the momentum-space signature of CT and Frenkel excitons as a first step towards imaging the orbital character of both types of excitons in molecular films. [1] Nat. Commun. 10, 1470 (2019)

MA 26.8 Tue 12:45 WIL B321

Momentum resolved ultrafast organic molecular exciton dynamics — •Ralf Hemm, Martin Mitkov, Florian Haag, Sebastian Emmerich, Sebastian Hedwig, Martin Aeschlimann, and Benjamin Stadtmüller — University of Kaiserslautern (TUK) and research center OPTIMAS, Erwin-Schroedinger-Str.46, 67663 Kaiserslautern, Germany

Mapping the lowest unoccupied molecular orbitals (LUMOs) of an organic semiconducting thin film in momentum space is one of the great challenges of ultrafast surface science. Especially on the femto- to picosecond timescale, resolving the transient molecular orbital structure can help to disentangle excitonic decay mechanisms.

Here, we apply bichromatic time-resolved two-photon momentum microscopy [1] to image the excited state dynamics of excitons in molecular materials in momentum space. Our model system is a multilayer of ordered C60 on Cu(111), for which the energy dependent exciton dynamics was already reported in literature [2]. In this talk, we will therefore focus on the momentum space signatures of the excited states. We will identify the characteristic momentum space signatures of the excited states and follow their evolution above and below the critical temperature for the structural phase transition of C60. This will allow us to correlate the exciton dynamics in momentum space to the dominant energy and momentum dissipation process in molecular materials.

[1] F. Haag et al., Rev. Sci. Instr. 90, 103104 (2019)
[2] A. Rosenfeldt et al., J. Chem. Phys. 133, 23 (2010)

MA 26.9 Tue 13:00 WIL B321

Ultrafast excited state dynamics and transient band structure renormalizations in endohedral metallofullerenes — •Sebastian Hedwig, Sebastian Emmerich, Benito Arnoldi, Johannes Stöckl, Benjamin Stadtmüller, and Martin Aeschlimann — Department of Physics and Research Center OPTIMAS, University of Kaiserslautern, Erwin-Schrödinger-Straße 46, 67663 Kaiserslautern, Germany

Fullerenes have been object to intense research in the past decades, with promising potential for their implementation in optoelectronic devices. Embedding metal atoms or clusters into the fullerene by chemical synthesis can alter the cage symmetry, which strongly influences the molecular transport properties as well as the available relaxation channels of excited electronic states. Of special interest in this field is the cluster-cage electron transfer. Here, we present a time resolved photoemission study carried out on thin films of the prototypical endohedral metallofullerene $Sc_3N@C_{80}$ in a fs UV-pump XUV-probe experiment. We observe a transient broadening of all (polaronic) molecular valence states which follows the timescale of the exciton formation and decay in the molecular films, in analogy to our recent findings for the fullerene C_{60} [1]. Moreover, we show that the exciton and polaron dynamics are strongly altered upon K intercalation of the pristine film. This enables us to draw conclusions regarding cluster-cage charge transfer on ultrafast timescales. [1] B. Stadtmüller et al., Nat Commun 10, 1470 (2019)

MA 26.10 Tue 13:15 WIL B321

Temperature effects on the electron dynamics of metal-organic interface states — •Klaus Stallberg[1], Masahiro Shibuta[1,2], and Ulrich Höfer[1] — [1]Fachbereich Physik, Philipps Universität Marburg, Germany — [2]Keio Institute of Pure and Applied Sciences, Keio University, Yokohama, Japan

The presence of electronic interface states (IS) can strongly affect the electron dynamics at organic/metal interfaces. In particular, IS mediated charge transfer has been observed for model systems comprising few layers of π-conjugated organic molecules on single-crystalline metal surfaces. The formation of Shockley derived IS is well understood in the meantime and experimentally observed properties, such as the free-electron-like momentum dispersion as well as the energy onset, are well reproduced by *ab initio* calculations. In contrast, the IS electron dynamics eludes a consistent theoretical description so far, and systematic experimental studies are still missing.

Here, we systematically investigate temperature effects on the formation and the relaxation dynamics of the IS for the organic/metal systems NTCDA/Ag(111) and PTCDA/Ag(111). Using two-photon photoemission (2PPE), we observe a pronounced decrease of the IS energy for increasing temperatures, which we attribute to an extended molecule-metal binding distance due to phonons in the molecular layer. Moreover, a drastic increase of the IS lifetime with temperature is found. While it can qualitatively be explained with a reduced phase space for electron scattering with metal bulk states, this temperature effect is much stronger than expected from a simple physical model.