O 3: Mini-Symposium: Ultrafast surface dynamics at the space-time limit I

Time: Monday 10:30–12:30 Location: R1

Invited Talk O 3.1 Mon 10:30 R1

Actuating and probing a single-molecule switch at femtosecond timescales — Dominik Peller, Carmen Roelcke, Lukas Z. Kastner, Thomas Buchner, Alexander Neef, Johannes Hayes, Florian Albrecht, Rupert Huber, and •Jascha Repp — Department of Physics, University of Regensburg, 93040 Regensburg, Germany

Combining scanning tunneling microscopy (STM) with lightwave electronics [1] has enabled the simultaneous femtosecond and sub-angstrom resolution in observing matter [2]. We now demonstrate the combined femtosecond and sub-angstrom access in the control of matter. Ultrafast localized electric fields in lightwave STM enable exerting atom-scale femtosecond forces to selected atoms. Utilizing these forces to excite coherent structural dynamics, we can modulate the quantum transitions of a single-molecule switch by up to 39% [3]. Further, we exploit the same single-molecule switch to quantitatively resolve the electromagnetic waveform of the tip-confined near-field transients directly inside the tunneling junction at atomic scales [4].

[1] T. L. Cocker et al., Nature Photon. 7, 620 (2013).
[2] T. L. Cocker et al., Nature 539, 263 (2016).
[3] D. Peller et al., Nature 585, 58 (2020).
[4] D. Peller et al., Nature Photonics (2020).

Invited Talk O 3.2 Mon 11:00 R1

Real space-time imaging of valence electron motion in molecules — •Manish Garg — Max Planck Institute for Solid State Research, Heisenbergstr. 1, 70569 Stuttgart, Germany

Chemical transformations in molecules are a consequence of valence electron motion and its eventual coupling to atomic motion, hence, tracking valence electron motion at the orbital level is the key to understanding and taming such transformations. Scanning tunnelling microscopy (STM) can passively and locally probe the valence electron density in molecules. Contemporary techniques in attosecond science, on the other hand, can generate and track the temporal evolution of a coherent superposition of quantum states of valence electrons by using strong laser fields, which can be probed only non-locally. In absence of the capability to trigger and probe electron dynamics at the single-orbital level, electron motion could only be inferred by reconstruction. Here, we demonstrate that the dynamics of coherent superposition of valence electron states generated by < 6 femtosecond long carrier-envelope-phase (CEP) stable laser pulses, can be locally probed with picometer spatial resolution and 300 attosecond temporal resolution simultaneously, at the single orbital-level with the help of an STM, defying the previously established fundamental space-time limit. We show that near fields of optical pulses confined to the apex of nanotip of an STM enable orbital imaging of electronic levels of molecules with pm resolution. We envisage that it will be possible to see a chemical bond formation dynamics through a transition state at the orbital level in the near future.

Invited Talk O 3.3 Mon 11:15 R1

Ultrafast structural phase transitions probed by low-energy electron diffraction — •Claus Ropers — IV. Phys. Inst., Göttingen, Germany — MPI for biophysical Chemistry

Governed by broken symmetries and an effectively reduced dimensionality, surfaces exhibit a multitude of complex phases and transitions, with prominent examples in metal-to-insulator lattice instabilities. Ultrafast measurement technology has greatly improved our understanding of the formation and non-equilibrium response of such phases, revealing transition pathways and means of optical control.

Despite these advancements, tracking the rapid structural evolution of surfaces has remained challenging. Low-energy electron diffraction (LEED) is an ideal tool to characterize the structure, symmetries and long-range order at surfaces, but it typically lacks ultrafast temporal resolution.

This talk will present the development and first applications of Ultrafast LEED, combining short-pulsed probing with ultimate surface sensitivity. I will give some methodical background on its implementation and will discuss results on optically induced structural phase transitions in charge-density wave systems [1,2].

[1] "Structural dynamics of incommensurate charge-density waves tracked by ultrafast low-energy electron diffraction" G. Storeck et al., Struct. Dyn. 7, 034304 (2020).
[2] "Coherent control of a surface structural phase transition", J.G. Horstmann et al., Nature 583, 232 (2020).

Invited Talk O 3.4 Mon 11:45 R1

Probing the ultrafast electron dynamics in the quantum spin Hall system Bismuthene with time-resolved ARPES — •Julian Maklar[1], Raul Stühler[2], Maciej Dendzik[1,3], Tommaso Pincelli[1], Shuo Dong[1], Samuel Beaulieu[1], Martin Wolf[1], Ralph Ernstorfer[1], Ralph Claessen[2], and Laurenz Rettig[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, D-14195 Berlin, Germany — [2]Physikalisches Institut and Würzburg-Dresden Cluster of Excellence ct.qmat, University of Würzburg, D-97070 Würzburg, Germany — [3]Department of Applied Physics, KTH Royal Institute of Technology, Stockholm, Sweden

Quantum spin Hall (QSH) systems are 2D topological insulators with promising device applications due to dissipationless, symmetry-protected spin currents in their edges. A promising high-temperature QSH material is Bismuthene – a 2D honeycomb lattice of bismuth atoms on a silicon carbide substrate – which features a wide bulk band gap and conductive 1D edge states located at substrate steps and domain boundaries.

Here, we investigate the electronic structure of Bismuthene upon ultrafast photoexcitation via time- and angle-resolved photoemission spectroscopy (trARPES). We map out the transiently occupied excited states in the conduction band and additionally observe faint spectral weight within the indirect band gap, which we attribute to metallic edge states. In addition, we observe a surprisingly fast recovery of excited carriers to the ground state – a further hint towards the presence of topological metallic decay channels.

Invited Talk O 3.5 Mon 12:00 R1

Atomic-resolution imaging of THz-driven dynamics on charge-ordered surfaces — •Sebastian Loth — Universität Stuttgart, Institut für Funktionelle Materie und Quantentechnologien, 70569 Stuttgart — Max-Planck-Institut für Festkörperforschung, 70569 Stuttgart

Charge order in correlated-electron materials is intimately linked to fluctuations of charge density that occur at nanometer length scales and at ultrafast speed. Such localized fluctuations are not a simple perturbation but can be the determining factor for the dynamics of phase transitions and the macroscopic response of the electron system. While these fluctuations often remain hidden to ensemble-averaged measurements, the combination of ultrafast THz spectroscopy and scanning tunneling microscopy can both locally excite and probe dynamics with atomic spatial and femtosecond temporal resolution [1]. For the quasi two-dimensional charge density wave state in niobium diselenide we find that the tip-enhanced electric field of the THz pulses excites the sample directly by driving a strong in-plane displacement current in the surface. This leads to a distortion of the charge-density wave in the vicinity of atomic pinning sites that relaxes by emitting a complex pattern of amplitude and phase excitations rather than a uniform collective response. Resolving these fluctuations in real space at the scale of individual impurities provides a new route to unraveling the electronic dynamics of disordered correlated materials.

[1] T. L. Cocker et al., Nature Photon. 7, 620 (2013). & T. L. Cocker et al., Nature 539, 263 (2016).