O 54: Scanning Probe Microscopy: Light Matter Interaction at Atomic Scales I

Time: Wednesday 15:00–17:45 Location: MA 041

Topical Talk O 54.1 Wed 15:00 MA 041

Ultrafast scanning tunneling microscopy as a local probe of femtosecond electron and coherent phonon dynamics — •Melanie Müller — Fritz Haber Institute, Berlin, Germany

Ultrafast scanning tunneling microscopy (USTM) has emerged as a powerful tool for imaging electronic and structural excitations at surfaces with angstrom spatial and femtosecond temporal resolution. THz-lightwave-driven STM (THz-STM) and optical photon-assisted STM (ph-STM) offer two approaches [1] to probe the dynamics of charge, orbital and lattice degrees of freedom at the atomic scale. While THz-STM uses single-cycle THz pulses to drive tunneling via classical rectification, ph-STM is based on photon absorption-induced tunneling. Both allow to probe coherent phonons (CPs) and ultrafast changes in the occupation and local density of electronic states (LDOS). CPs modulate the electronic structure of solids, and thus the electronic states involved in tunneling. Here we use ph-STM to probe CPs in ultrathin ZnO/Ag(111) via their coupling to an optical resonance that enhances photon-assisted tunneling [2]. On the other hand, THz-STM allows more direct access to the LDOS by quasi-static tunneling. We study the commensurate charge density wave (CDW) phase in 1T-TaS_2, and probe the photoinduced Mott collapse and coherent CDW amplitude motion with THz-STM. The ability to probe both ultrafast local atomic and electronic structure makes USTM unique for imaging spatially inhomogeneous nonequilibrium dynamics in strongly correlated materials at the atomic scale. [1] M. Müller, Prog. Surf. Sci. 100727 (2023) [2] S. Liu et al., Sci. Adv. abq5682 (2022)

O 54.2 Wed 15:30 MA 041

THz waveform optimization for ultrafast time-domain spectroscopy in the scanning tunneling microscope — Shaoxiang Sheng[1,2], Kurt Lichtenberg[1], Felix Huber[1], Johannes Schust[1], Li Chen[3], Susanne Baumann[1], and •Sebastian Loth[1] — [1]University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany — [2]Max Planck Institute for Solid State Research, Stuttgart, Germany — [3]Program in Materials Science and Engineering, University of California San Diego, La Jolla, CA, USA

Exciting the tunnel junction of a scanning tunneling microscope (STM) with sub-cycle THz pulses enables ultrafast spectroscopy of electron, phonon and molecular motion dynamics on surfaces with atomic spatial and sub-picosecond temporal resolution. THz pulses coupled to the STM tip induce short bursts of electron tunneling that are used to sample the ultrafast dynamics. Hence, the sensitivity and time resolution achievable with this technique critically depends on the quality of the THz pulse's electric-field waveform. We use electro-optic sampling of tip-scattered THz light (s-EOS) from the STM tip and pulse-pulse correlation measurements of the THz-induced tunnel current to characterize the shape of the THz electric field in the tunnel junction. Comparison with finite-element modeling of the THz near field identifies reflections and spectral distortions that originate from THz surface plasmon propagation along the STM tip. We find an optimized tip geometry that flattens the electric field waveform in the tunnel junction and enables accurate time-domain spectroscopy.

O 54.3 Wed 15:45 MA 041

Time-domain Detection of Ultrafast Voltage Transients in an STM Tunnel Junction — Vedran Jelic[1], Stefanie Adams[1], Mohamed Hassan[1], Kaedon Cleland-Host[1], •Spencer E. Ammerman[2], and Tyler L. Cocker[1] — [1]Department of Physics and Astronomy, Michigan State University, East Lansing, MI 48824, USA — [2]EMPA, Swiss Federal Laboratories for Materials Science and Technology, 8600 Dübendorf, Switzerland

Lightwave-driven terahertz scanning tunneling microscopy (THz-STM) has emerged as a powerful tool for investigating ultrafast dynamics at atomic length scales. Illuminating the tunnel junction of an STM with a phase-stable single-cycle THz pulse has been shown to induce sub-picosecond tunneling currents coherently driven by the electric field of the THz transient, providing access to the local density of electronic states. A critical factor for interpretation of THz-STM measurements is knowledge of the ultrashort voltage transient. However, the THz transient is subject to spectral modification by variation in the nanoscale dielectric function, as shown by scanning near-field optical microscopy experiments. In order to distinguish between local samples properties and effects arising from terahertz-pulse coupling, a method for extracting the near-field transient is needed. Here, we demonstrate a THz-STM cross-correlation experiment for local time-domain sampling of the THz voltage transient. Combining self-consistent modeling and waveform shaping we introduce a generally applicable scheme for atomic-scale terahertz time-domain spectroscopy, spatially confined to an STM tunnel junction.

O 54.4 Wed 16:00 MA 041

Efficient and continuous THz carrier-envelope phase control for ultrafast lightwave-driven STM — •Jonas Allerbeck[1], Joel Kuttruff[2], Laric Bobzien[1], Lysander Huberich[1], Maxim Tsarev[2], and Bruno Schuler[1] — [1]nanotech@surfaces, Empa, Swiss Federal Laboratories for Material Science and Technology, Überlandstrasse 129, 8600 Dübendorf Switzerland — [2]University of Konstanz, Universitätsstrasse 10, 78464 Konstanz, Germany

THz lightwave-driven scanning tunneling microscopy (THz-STM) facilitates the study of atomically precise structures with sub-cycle picosecond temporal resolution, leveraging the fundamental understanding of quantum systems and carrier dynamics at the nanoscale. Here, I present a versatile THz-STM toolbox, operating at up to 41 MHz repetition rate and using an efficient method to continuously control the carrier-envelope phase of single-cycle THz pulses, established by frustrated internal reflection in a right-angle polymer prism. The setup achieves peak THz voltages at the tunneling junction ranging from 1 V at 41 MHz to few-10 V at 1 MHz, where multi-MHz repetition rates enable state-selective tunneling to localized orbital states of atomic quantum defects below 0.01 electrons per THz pulse. Photoemission sampling and lightwave-driven THz cross correlation benchmark the near-field THz amplitude. We study atomic point defects in few-monolayer transition metal dichalcogenide epitaxial crystals, featuring few-ps charge state lifetime. The precise control of the transient THz waveform at the tunneling junction paves the way towards exploring local carrier dynamics with atomic resolution.

O 54.5 Wed 16:15 MA 041

Towards STM-based atomic-scale scanning near-field optical microscopy — •Fabian Schulz[1], Jun Nishida[2], Adnan Hammud[3], Shuyi Liu[4], Takashi Kumagai[2], Martin Wolf[3], Akitoshi Shiotari[3], and Melanie Müller[3] — [1]CIC nanoGUNE, San Sebastian, Spain — [2]Institute for Molecular Science, Okazaki, Japan — [3]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [4]Huazhong University of Science and Technology, Wuhan, China

Scattering-type scanning near-field optical microscopy (sSNOM) enables measuring the optical properties of surfaces with a lateral reslolution beyond the diffraction limit. Conventional sSNOM is based on cantilever atomic force microscopy with the resolution limited to typically 10 - 20 nm. Here, using plasmonic nanocavities in a low-temperature scanning tunneling microscope (LT-STM), we demonstrate the potential of STM-based sSNOM in the visible regime to reach a resolution of at least ∼1 nm. As a prerequisite for future plasmonic sSNOM using LT-STM, we investigate the interplay between the measured near-field signal and the gap plasmon formed inside the STM junction.

Topical Talk O 54.6 Wed 16:30 MA 041

Ultrafast scanning tunnelling spectroscopy of a phonon-driven atomic vacancy in a monolayer crystal — Carmen Roelcke, Lukas Kastner, Maximilian Graml, Andreas Biereder, Jan Wilhelm, Jascha Repp, Rupert Huber, and •Yaroslav Gerasimenko — Department of Physics and Regensburg Center for Ultrafast Nanoscopy (RUN), University of Regensburg, 93040 Regensburg, Germany

Directly observing in actual microscopic videography how the motion of a specific atom affects the electronic structure and the functionality of solids has been a long-held dream of modern science. Here, we break this ground with tomographic lightwave-driven scanning tunnelling spectroscopy by taking snapshots of the electronic spectrum of a phonon-driven atomic defect faster than the vibration period. We directly resolve in space, time, and energy how atomic motion transiently modulates the bound state of a selenium vacancy, the archetypal

single-photon emitter, in a WSe_2 monolayer. By combining atomically-confined excitation of a drum-like phonon with ultrafast tunnelling spectroscopy reaching atomic spatial and 300 fs temporal resolution, we reveal transient energy shifts of the lowest bound defect state by up to 40 meV, depending on the amplitude and phase of the phonon. This combination marks a disruptive development towards understanding and control of quantum matter by accessing key mechanisms at their intrinsic length, time and energy scales.

O 54.7 Wed 17:00 MA 041

Observing correlated electron dynamics in the local density of states of 1T-TaS_2 by THz-STM — •Luis Parra López[1], Alkisti Vaitsi[1], Vivien Sleziona[1], Fabian Schulz[2], Martin Wolf[1], and Melanie Müller[1] — [1]Fritz Haber Institute, Berlin, Germany — [2]CIC nanoGUNE, San Sebastian, Spain

THz scanning tunneling microscopy (THz-STM) has emerged as a promising technique for imaging surfaces with femtosecond temporal and angstrom spatial resolution. The prospect of imaging both local charge and lattice dynamics is of particular interest for the study of strongly correlated materials whose physical properties are governed by strong electron-electron and electron-phonon interactions. 1T-TaS_2 is a fascinating example, where the formation of a commensurate charge density wave (C-CDW) phase is accompanied by the opening of a Mott gap. Femtosecond optical excitation leads to ultrafast collapse of the Mott gap and launches coherent amplitude motion of the CDW. We discuss to what extent both can be probed by THz-STM and decipher the mechanisms by which transient electronic temperatures, ultrafast Mott collapse and coherent AM oscillations modulate the tunneling current. We present a model to extract ultrafast changes in the local density of states in the C-CDW phase of 1T-TaS_2. We reproduce the transient change of the rectified current measured in THz-STM after global ultrafast photoexcitation of 1T-TaS_2. Our results provide the basis to explore ultrafast insulator-to-metal transitions and coherent CDW oscillations with the angstrom spatial resolution provided by THz-STM.

O 54.8 Wed 17:15 MA 041

Combining THz Pump-Probe spectroscopy with Scanning Tunneling Luminescence — •Kurt Lichtenberg, Johannes Schust, Felix Huber, Susanne Baumann, and Sebastian Loth — University of Stuttgart, Institute for Functional Matter and Quantum Technologies, Stuttgart, Germany

Inelastic electron tunnelling in scanning tunnelling microscopes (STM) can trigger atomically-localized emission of light from metallic tunnel junctions, molecules or semiconducting surfaces. This enables studies of light-matter interaction at sub-molecular length scales. Fast gating of the tunnelling current by pulsed voltage sources gives access to the dynamics of the underlying excitation and light emission processes. Electronic pulse generators provide nanosecond time resolution [1], but much faster voltage transients are possible by exciting the STM's tunnel junction with single-cycle THz pulses, which were shown to generate electroluminescence [2]. We combine THz excitation [3] with single-photon detection in a variable-temperature STM and investigate the possibilities for ultrafast and atomically resolved measurements of electroluminescence using THz pump-probe spectroscopy.

[1] C. Grosse, et al. Appl. Phys. Lett. 103 183108 (2013)
[2] K. Kimura, et al. ACS Photonics 8 982 (2021)
[3] M. Abdo, et al. ACS Photonics 8 702 (2021)

O 54.9 Wed 17:30 MA 041

Amplitude Calibration for THz-STM of Atomic Defects in 2D Semiconductors — •Laric Bobzien, Jonas Allerbeck, Spencer Eve Ammermann, and Bruno Schuler — nanotech@surfaces, Empa -Swiss Federal Laboratories for Material Science and Technology, Überlandstrasse 129, 8600 Dübendorf Switzerland

The fundamental understanding of quantum dynamics in low-dimensional materials requires simultaneous ultrafast temporal and atomic spatial resolution, achievable with ultrafast STM. Here, single-cycle THz-pulse-driven STM (THz-STM) combines the atomic spatial resolution with picosecond temporal resolution of the THz field. Point defects in 2D semiconductors, such as vacancies or impurities, are a perfect model systems to investigate strongly correlated dynamics in low dimensions and offer a rich playground for understanding dynamics in the atomic regime. We study point defects in a prototypical family of 2D semiconductors, transition metal dichalcogenides (TMDs), with our multi-MHz repetition rates and efficient THz-STM.

I will show recent measurements of the THz rectified charge as a function of bias and THz field amplitude on 2D TMDs. This allows to map the complex energy landscape of localized states with a resolution of down to 0.01 electrons per pulse enabling precise near-field amplitude calibration. Preliminary THz-STM and THz-STS measurements of a sulfur vacancy in monolayer MoS2 demonstrate state-selective tunneling into distinct electronic orbitals of the quantum defect. These findings pave the way for exploring atomic systems at their native length and time scale.