

## O 43: Scanning probe microscopy: light matter interaction at atomic scales – Poster

Time: Tuesday 14:00–16:00

Location: P2

O 43.1 Tue 14:00 P2

**Modelling the near-field enhancement for tip-enhanced photoluminescence on non-conductive layers** — ●JINHUI GUO and LAERTE PATERA — Department of Physical Chemistry, University of Innsbruck, 6020 Innsbruck, Austria

While scanning tunnelling microscopy (STM)-based tip-enhanced photoluminescence (TEPL) has been demonstrated for molecules adsorbed on 2-3 monolayers (ML) of NaCl/Ag(111) [1], nonradiative quenching due to charge tunnelling through the insulating layer is expected to affect the exciton lifetime. Employing sufficiently thick insulating layers ( $> 14$  ML) strongly suppresses electron tunnelling [2], potentially preserving the photoexcited state. However, such thick insulating layers are incompatible with STM operation, making atomic force microscopy (AFM) an ideal choice. To elucidate the field enhancement in AFM-based TEPL experiments, we performed three-dimensional finite element method (3D-FEM) simulations [3] for various AFM Cu tip geometries and Cu substrates with and without NaCl layers up to 20 ML. Our results indicate that below the tip, a pronounced electric field enhancement in the plane parallel to the sample surface ( $E_{\parallel}$ ) persists even on a 20 ML-thick NaCl layer. This effect arises mainly from the reduced screening of  $E_{\parallel}$  within the NaCl layers compared to the metallic Cu substrate. These findings support the exploitation of non-conductive layers for TEPL experiments.

[1] J. Doležal *et al.*, *Nano Lett.* 2024, 24, 1629-1634. [2] W. Steurer *et al.*, *Nat. Commun.* 2015, 6, 8353. [3] COMSOL Multiphysics® v. 6.3. www.comsol.com. COMSOL AB, Stockholm, Sweden.

O 43.2 Tue 14:00 P2

**Sub-nanoscale photochemistry without plasmons: Debromination of dibromoterphenyl on Au(111)** — HAO JIANG<sup>1,2</sup>, YOUNGWOOK PARK<sup>1</sup>, ADNAN HAMMUD<sup>1</sup>, MARTIN WOLF<sup>1</sup>, QIANG SUN<sup>2</sup>, and ●AKITOSHI SHOTARI<sup>1</sup> — <sup>1</sup>Fritz Haber Institute of the Max Planck Society, Berlin, Germany — <sup>2</sup>Materials Genome Institute, Shanghai University, Shanghai, China

Local control of light-induced reactions is a crucial process for developing molecule-based optodevices and designing photocatalysis. Nanometer- and angstrom-scale photoreaction controls have been demonstrated using scanning tunneling microscopy (STM) combined with laser irradiation [1,2]; however, so far they rely on localized surface plasmon resonance (LSPR) using plasmonic metal tips. Here we report a novel light-assisted reaction that can be induced locally without LSPR. Using laser-coupled low-temperature STM, we observed 4,4'-dibromo-p-terphenyl (DBTP) on Au(111), which is a representative system undergoing Ullmann coupling by heat or far-field irradiation [3,4]. By tuning both the bias voltage at the STM junction and the wavelength of the incident visible laser, the debromination of DBTP is induced at the sub-nanometer scale using non-plasmonic tips. This research opens up the prospect of performing the single-molecule photochemistry for various non-plasmonic systems.

[1] E. Kazuma *et al.*, *Prog. Surf. Sci.* 93, 163 (2018). [2] Y. Park *et al.*, *Nat. Commun.* 15, 6709 (2024). [3] H. Jiang *et al.*, *ACS Nano* 18, 1118 (2024). [4] H. Jiang *et al.*, *Nano Lett.* 25, 9597 (2025).

O 43.3 Tue 14:00 P2

**Photoisomerization of azobenzene derivatives on Au (111) using various photon energies and polarizations** — ●HARAPRASAD MANDAL<sup>1</sup>, PAUL SCHÖNGRUNDNER<sup>1</sup>, STEFAN HECHT<sup>2</sup>, and LEONHARD GRILL<sup>1</sup> — <sup>1</sup>Department of Chemistry, University of Graz, Austria — <sup>2</sup>Department of Chemistry, Humboldt-Universität zu Berlin, Germany

Azobenzene derivatives continue to serve as model systems for studying photoisomerization at surfaces. Among them, tetra-tert-butyl azobenzene (TBA) is particularly suitable due to its chemical stability, large steric groups, and well-defined trans and cis configurations that can easily be identified in scanning tunnelling microscopy (STM) images. In this work, we systematically studied the photon-energy- and polarization-dependent trans-to-cis isomerization of TBA, using a tunable femtosecond laser in combination with STM (working in ultra-high vacuum and at low temperatures). Illumination is done through an optical port, and STM imaging is used to identify the configuration of individual molecules before and after laser exposure. The switching efficiency was then studied for a variety of wavelengths in the range between 350 nm and 800 nm as well as different (s/p) polarization di-

rections of the incoming light. Our results provide direct insight into the parameters that steer TBA photoisomerization on Au(111) and demonstrate the capability of our laser STM platform for controlled photochemical studies of single molecules.

O 43.4 Tue 14:00 P2

**Focal phase evolution of single-cycle THz near-field pulses in a scanning tunneling microscope** — ●ANDREA ROSSETTI, VIVIEN SLEZIONA, ALKISTI VAITSIS, LUIS ENRIQUE PARRA LOPEZ, MARTIN WOLF, and MELANIE MÜLLER — <sup>1</sup>Department of Physical Chemistry, Fritz Haber Institute, 14195 Berlin, Germany

Lightwave scanning tunneling microscopy (LW-STM) employs single-cycle THz pulses as femtosecond bias, enabling imaging at sub-nanometer spatial and sub-picosecond temporal resolution. Control over the polarity and carrier-envelope phase (CEP) of such THz voltage transients is thereby crucial in determining which electronic states contribute to the ultrafast tunneling current. A simple approach for continuous CEP control of the single-cycle THz transients in LW-STM is via the Gouy phase. In this work, using a spintronic THz emitter, we use photoemission sampling from the point-like apex of the STM tip to monitor the local CEP of the tip-enhanced THz pulses along the focal axis of a focused THz beam. We find that the CEP exhibits a non-trivial evolution through the focus that strongly deviates from the expected Gouy phase shift for a monochromatic beam. Using analytical models and numerical calculations, we discuss the influence of the broad bandwidth of the THz pulses as well as their coupling to the STM tip for the local CEP in LW-STM.

O 43.5 Tue 14:00 P2

**Development of Cryogenic Photoinduced Force Microscopy: Introduction and Progression** — ●SHUXIA CHEN<sup>1</sup>, CLAUDIA FELSER<sup>1</sup>, LUKAS ENG<sup>2</sup>, and FABIAN MENGES<sup>1</sup> — <sup>1</sup>Max Planck Institute for Chemical Physics of Solids — <sup>2</sup>Technische Universität Dresden

Photo-induced Force Microscopy (PiFM) provides access to optical properties at atomic length scales through the detection of near-field optical forces. To date, PiFM experiments have been largely restricted to ambient conditions, with only limited demonstrations in extreme environments such as cryogenic temperatures or high magnetic fields. In this work, we aim to expand PiFM operation to low temperatures and large external magnetic fields. This development opens the door to exploring light matter interactions over an expanded parameter space and in exotic quantum materials.

O 43.6 Tue 14:00 P2

**Investigation of atomic defects in quasi-freestanding MoS<sub>2</sub> with THz-STM** — ●CAROLINE FIRSCHKE, JUNYOUNG SIM, VIBHUTI N. RAI, PAUL WIECHERS, FLORIAN FAABER, CHRISTIAN LOTZE, and KATHARINA J. FRANKE — Freie Universität Berlin, Germany

Transition metal dichalcogenides (TMDs) are an interesting class of materials due to their 2D-like nature and unique band structures. These materials tend to host different types of defects, heavily influencing their optoelectronic properties. THz-driven scanning tunneling microscopy is a unique technique to investigate the nature of these defects on atomic scales with ultrafast time resolution (1).

Here, we grow monolayers of MoS<sub>2</sub> on Au(111) via chemical vapor deposition, which host quasi-freestanding monolayers of MoS<sub>2</sub> with a band gap of around 2.7 eV, the so called "pits" (2). We find structural defects in these pits, which are decoupled from the metal substrate, and show sharp positive ion resonances in the band gap.

These sharp non-linearities of the I-V curve provide favorable conditions for their investigation via THz pulses. We find that these defects show sharp resonances in the light-wave-driven scanning tunneling spectroscopy, which vary depending on the THz field strength, DC bias voltage and tip-sample distance. The THz response of these resonances cannot be modeled with a simple convolution of THz pulses with the static I-V of the junction, suggesting a modulation of the charge state of the defects on ultrafast timescales.

(1) T. L. Cocker *et al.* *Nature Photon* 7, 620-625 (2013)

(2) N. Krane *et al.* *Nano Letters* 2016 16 (8), 5163-5168

O 43.7 Tue 14:00 P2

**Photo-switching of azobenzene derivatives on Au(111)** —

•MATTHEW J. TIMM<sup>1</sup>, ROBERT DI VORA<sup>2</sup>, STEFAN HECHT<sup>3</sup>, BIRGITTA BERNHARDT<sup>2</sup>, and LEONHARD GRILL<sup>1</sup> — <sup>1</sup>Institute of Chemistry, University of Graz, Austria — <sup>2</sup>Institute of Experimental Physics, Graz University of Technology, Austria — <sup>3</sup>Department of Chemistry & IRIS Adlershof, Humboldt-Universität zu Berlin, Germany

The light-induced trans-cis isomerization of azobenzene and its derivatives is of great scientific and technological interest as these molecular photo-switches can be used for nano-scale sensing or information storage. Particularly, 3,3',5,5'-tetra-tert-butylazobenzene (mTBA) adsorbed upon Au(111) has seen much interest, being studied at sub-monolayer coverage (around 0.1 ML) by scanning tunneling microscopy (STM) [1,2] and near monolayer (ML) coverage (0.9 ML) by two-photon photoemission [3]. However, missing is a single-molecule level insight into whether photo-switching at low coverage differs from that at higher but still sub-monolayer coverage - does it matter if only small or large areas of the surface are covered? Here, we combine STM with a self-designed computer-vision algorithm to study photoisomerization induced by pulsed laser radiation ( $\lambda = 518$  nm) at a rather high surface coverage of 0.6 ML. This approach can track the isomerization state of thousands of individual mTBA molecules and enables us to detect subtle variations in switching yields among densely packed, similarly oriented molecules. [1] M. Alemani, et al. JACS 128 (2006) 14446. [2] M. J. Comstock, et al. Phys. Rev. Lett. 99 (2007) 038301. [3] S. Hagen, et al. Chem. Phys. Lett. 444 (2007) 85-90.

O 43.8 Tue 14:00 P2

**Sub-10-fs compression of NIR pulses for phase-resolved sampling of MIR lightwave transients** — •JUSTUS HESS, UGAITZ ELU, ANDREA ROSSETTI, MARTIN WOLF, and MELANIE MÜLLER — Department of Physical Chemistry, Fritz Haber Institute, 14195 Berlin, Germany

Lightwave scanning tunneling microscopy (LW-STM) employs single-cycle THz pulses as femtosecond bias [1], enabling imaging at sub-nanometer spatial and sub-picosecond temporal resolution. Ultrashort phase-stable mid-infrared (MIR) pulses promise to increase the time resolution in LW-STM. Phase-resolved characterization of the incident and STM tip-enhanced MIR transients requires sampling pulses with a duration of ideally less than 10 fs. For future MIR excitation in our LW-STM, we implement a MIR source and a pulse-compression scheme based on supercontinuum generation in a photonic crystal fiber (PCF) at 2 MHz in our existing LW-STM setup. Near-infrared (NIR) pulses with a duration of  $\sim 40$  fs at a center wavelength of 1030 nm are spectrally broadened via self-phase modulation in the PCF to a Fourier-limit of 6 fs. Subsequent dispersion control with a transmission-grating compressor yields few-cycle pulses at 30mW average power sufficient for MIR waveform sampling in LW-STM. [1] T. Cocker et al., Nature 539, 263\*267 (2016)

O 43.9 Tue 14:00 P2

**Excitation of phonons by THz near fields in a quasi-suspended nanoscopic monolayer of MoS<sub>2</sub>** — •JUNYOUNG SIM, VIBHUTI RAI, CAROLINE FIRSCHKE, CHRISTIAN LOTZE, and KATHARINA J. FRANKE — Freie Universität Berlin, Department of Physics, Arnimallee 14, 14195 Berlin, Germany

Phonons play a central role in describing fundamental physical properties of materials, such as heat capacity and resistivity, and also govern many-body phenomena such as superconductivity and charge-density waves. To resolve ultrafast lattice vibrations at the atomic scale, terahertz scanning tunneling microscopy (THz-STM) has provided important groundworks, where such vibrations are influenced by the presence of atomic-scale defects [1, 2].

In this work, we investigate the phonon dynamics in a monolayer molybdenum disulfide (MoS<sub>2</sub>) grown on a Au(111) surface. We focus on nanoscopic regions, where the MoS<sub>2</sub> layer is suspended and the electronic structure is minimally hybridized with the substrate [3]. We observe an oscillatory behavior in the THz-STM pump-probe spectra on these suspended regions, which we assign to a coherent vibrational mode excited and measured by the tip-enhanced THz near field. To elucidate the excitation mechanism of the phonon modes, we vary the pump/probe field and investigate the influence of defects within the suspended area.

[1] Roelcke et al., Nature Photonics. 18, 595-602 (2024)

[2] Rai et al., arXiv:2506.08219 (2025)

[3] Krane et al., Nano Lett. 2016, 16, 8, 5163-5168 (2016)

O 43.10 Tue 14:00 P2

**THz-driven plasmonic STM luminescence** — •ALKISTI VAITSIS,

LUIS ENRIQUE PARRA LÓPEZ, VIVIEN SLEZIONA, MARTIN WOLF, and MELANIE MÜLLER — Fritz Haber Institute of the Max Planck Society, Berlin, Germany

We combine THz scanning tunneling microscopy (THz-STM) with visible light detection to investigate ultrafast STM-luminescence (STML) luminescence from atomic-scale plasmonic junctions. The luminescence is driven by single-cycle THz pulses which act as a quasi-static ultrafast bias. This enables femtosecond gating of ultrafast tunneling currents and the excitation of localized surface plasmon (LSP) modes via ultrafast inelastic tunneling. Our measurements, combined with numerical reconstruction of THz-STML spectra from static STML spectra, show that a quantum cutoff is often absent in THz-STML. In addition, THz-STML spectra frequently exhibit a distinct spectral shape that differs from that predicted from quasi-static, single-electron tunneling-induced luminescence. We discuss the role of overbias emission, hot luminescence, and femtosecond plasmon dynamics in THz-gated STML.

O 43.11 Tue 14:00 P2

**Design of a light-coupled high-magnetic-field Scanning Tunneling Microscope** — •COLIN GEUDER<sup>1</sup>, MARCO MÖNNICH<sup>1</sup>, KURT LICHTENBERG<sup>1</sup>, SUSANNE BAUMANN<sup>1</sup>, and SEBASTIAN LOTH<sup>1,2</sup> — <sup>1</sup>University of Stuttgart, Institute for Functional Matter and Quantum Technologies. — <sup>2</sup>Center for Integrated Quantum Science and Technology (IQST), University of Stuttgart.

Light-matter interaction at the nanoscale lies at the heart of many fundamental phenomena ranging from light harvesting to ultrafast lightwave electronics. However, accessing magnetic degrees of freedom through light-matter interaction at the atomic scale presents unique challenges. Scanning tunneling microscopy (STM) is the ideal tool, that can locally probe and excite these different interactions at atomic length scales. Here, we present the design of an STM that combines optical access with a large numerical aperture, magnetic fields up to 5 T, and temperatures below 2 K. We mitigate thermal loads introduced by the high-NA optical access through a combination of fiber coupling and radiation blocks at several stages of the cryostat's shields. This instrument enables studies of magnetic degrees of freedom in nanoscale light-matter interaction, such as magnon-photon coupling or exciton-magnon transport in layered materials.

O 43.12 Tue 14:00 P2

**strong-field theory model for ultrafast electron transport in laser-assisted STM** — •BOYANG MA<sup>1</sup>, MELANIE MUELLER<sup>1</sup>, and MICHAEL KRUEGER<sup>2</sup> — <sup>1</sup>Department of Physical Chemistry, Fritz Haber Institute, 14195 Berlin, Germany — <sup>2</sup>Department of Physics, Technion, Israel Institute of Technology, 32000 Haifa, Israel

In ultrafast scanning tunneling microscopy (USTM), the classification of light-driven currents is usually based on the well-known Keldysh parameter. Similar to the description of strong-field photoemission, it is often used to distinguish photon-driven (weak-field) and field-driven (strong-field) tunneling regimes. Here, we discuss the limitations of the Keldysh parameter in classifying light-induced electron transport in nanoscale gaps, and present a theoretical study of laser-induced electron transport in STM [1,2]. In contrast to strong-field effects in spatially extended systems, we show that the Keldysh parameter alone is insufficient to capture the electron transmission dynamics in a confined nanogap. Instead, it must be complemented by an additional parameter,  $\zeta$ , defined by the laser field strength, frequency, and junction width [2]. Notably, in the THz-STM,  $\zeta$  alone becomes the decisive parameter, superseding the role of the Keldysh parameter. This reflects the fact that the Keldysh parameter accounts solely for properties of the driving field and neglects geometric confinement of the tunneling junction.

[1] B. Ma and M. Krüger, Phys. Rev. Lett. 133, 236901 (2024)

[2] B. Ma and M. Krüger, Phys. Rev. A 112, 033104 (2025)

O 43.13 Tue 14:00 P2

**Addressing Atomic-scale optical Stark-shift Microscopy** — XABIER ARRIETA<sup>1,2</sup>, SOFIA CANOLA<sup>3</sup>, RUBEN ESTEBAN<sup>1,4</sup>, JAVIER AIZPURUA<sup>2,4,5</sup>, and •TOMAS NEUMAN<sup>3</sup> — <sup>1</sup>Centro de Física de Materiales (CFM-MPC), CSIC-UPV/EHU, Donostia- San Sebastián 20018, Spain — <sup>2</sup>Department of Electricity and Electronics, FCT-ZTF, UPV/EHU, Leioa 48940, Spain — <sup>3</sup>Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10, Prague, 16200, Czech Republic — <sup>4</sup>Donostia International Physics Center, Donostia-San Sebastián 20018, Spain — <sup>5</sup>IKERBASQUE, Basque Foundation for Science, Euskadi Plaza 5, Bilbao 48009, Spain

An applied external electric field can shift the optical transition energies of molecular emitters through the optical DC Stark effect. Conventional Stark-shift spectroscopy applies spatially homogeneous electric fields, allowing to probe variations in molecular dipole moment and polarizability upon photoexcitation. In contrast, molecules in natural environments experience strongly inhomogeneous internal electric fields, where the Stark effect follows different selection rules. We extend the theory of Stark-shift spectroscopy to account for such nanoscale field. Similar field localization naturally arises in light-assisted scanning tunneling microscopy, which could provide spatially resolved access to the electronic response of individual molecules under optical excitation [1,2]. We demonstrate the potential of this technique to develop into atomic-scale Stark-shift microscopy. [1] Roslowska et al., Phys. Rev. X, 12, 011012 (2022). [2] Imada et al., Science, 373(6550), 95-98 (2021).

O 43.14 Tue 14:00 P2

**Light emission from dibenzoterrylene (DBT) molecules studied with STML** — •YANNIS HILGERS, ANDREAS REUTTER, MARKUS ETZKORN, and UTA SCHLICKUM — Institute of Applied Physics, LENA - TU Braunschweig, Braunschweig, Germany

Scanning Tunneling Microscopy induced Luminescence (STML) has gathered great interest since its development in the late 1980s as it allows studying optical properties with atomic spatial resolution. With this technique, it has become possible to study quantum light sources like individual molecules exhibiting single photon emission, but obtaining high and reproducible photon yields has proven to remain challenging. With our self-built STML-setup, we achieve count rates about an order of magnitude higher than what has been reported in the past, allowing for measurements on more sensitive systems. Here, we present optical properties of dibenzoterrylene (DBT) molecules which have been reported as single photon sources when encapsulated inside a host lattice of Anthracene and Naphthalene. However, since both materials are not stable, we combine DBT molecules with a host lattice consisting of C<sub>60</sub> multilayers, which is much more robust and even stable in air at room temperature. We found that light emission of individual DBT on C<sub>60</sub> multilayers is indeed preserved. The emitted light was analyzed by means of photon maps and photon energy spectra.

O 43.15 Tue 14:00 P2

**Influence of Polarons on Tunneling and Optical Spectra of Molecules Adsorbed on Dielectric Layers** — •JAN FRIESE — Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10, Prague, 16200, Czech Republic

Light-assisted scanning tunneling microscopy has enabled the simultaneous probing of transport and optical properties of individual molecules with atomic-scale precision. This technique relies on electronically decoupling the molecules from the metallic substrate by depositing them on dielectric spacer layers. However, these spacers are often formed by ionic crystals, such as NaCl. The phononic response of these ionic layers can significantly impact the observed experimental features due to polaronic effects. Their understanding requires a bottom-up theoretical description accounting for the dynamics of the dielectric layer triggered by electronic transitions in the molecule.

Here we address this challenge and use the polaron-formalism to

model the interaction of phonons in the dielectric layer with changes of the charge density in the molecule due to optical or charge transfer excitations. With that, two-time correlation functions and consequently spectral functions can be derived to account for the dynamics involved in substrate reorganization processes following a change of charge density in an adsorbed molecule. The results can be used to calculate features like the reorganization energy, and positions, widths and shapes of peaks in tunneling and optical spectra.

O 43.16 Tue 14:00 P2

**Optics of Edge-Modified Graphene Nanoribbons: A Theoretical Perspective** — •JIAN CHENG WONG<sup>1</sup>, SONG JIANG<sup>2</sup>, GUILLAUME SCHULL<sup>2</sup>, and TOMÁŠ NEUMAN<sup>1</sup> — <sup>1</sup>Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10, 16200 Prague, Czech Republic — <sup>2</sup>Université de Strasbourg, CNRS, IPCMS, UMR 7504, F-67000 Strasbourg, France

Atomic-scale engineering of graphene nanoribbons (GNRs) provides a versatile means to tune their electronic and optical properties. We present a theoretical study of edge-modified GNRs that describe the process resulting in excitonic emission under scanning tunneling microscopy-induced luminescence (STML). The modified edge structure forms localized states in GNR similar to the end states studied formerly[1]. From the electroluminescence maps, we explore how interactions between two of these localized states change the optical properties depending on the GNR structure. For this, we construct a many-body model which describes the dynamics of tip-induced charging/discharging and relaxation that eventually leads to light emission. [1] Song et al., Science, 379(6636), 1049-1054 (2023).

O 43.17 Tue 14:00 P2

**Utilizing a Tunable Laser for Tip Enhanced Photoluminescence of Single Molecules** — •MAXIMILIAN RÖDEL<sup>1</sup>, RODRIGO CEZAR DE CAMPOS FERREIRA<sup>1</sup>, JIRÍ DOLEŽAL<sup>1</sup>, and MARTIN ŠVEC<sup>1,2</sup> — <sup>1</sup>Institute of Organic Chemistry and Biochemistry (IOCB), Czech Academy of Sciences, Flemingovo náměstí 542/2. CZ16000, Praha 6, Czech Republic — <sup>2</sup>Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10/112, CZ16200, Praha 6, Czech Republic

The study of organic molecules and their optical properties provides insight into molecular interactions, guiding the design of materials for advanced opto-electronic devices. In this contribution, we present a tip-enhanced photoluminescence (TEPL) scanning tunneling microscope (STM) setup by utilizing a tunable Ti:Sapphire laser not only for resonant excitation of organic molecules like MgPc, but, moreover, for disentangling optical signals.

For this purpose, a parabolic mirror is used for coupling the laser into the nano-cavity, ensuring wavelength-independent imaging and minimizing aberrations, enabling a consistent performance across the entire visible wavelength range. By doing so, high-resolution spectra of single-molecule MgPc at temperatures of <7 K on a Ag111/NaCl surface can be detected and by changing the excitation wavelength the vibronic contribution and the Raman signal can be distinguished.

With this model system, we are able to complement our optical data set and even hope to establish photoluminescence excitation spectroscopy for future measurements.