O 12: Scanning probe microscopy: light matter interaction at atomic scales

Time: Monday 15:00–17:45 Location: HSZ/0403

O 12.1 Mon 15:00 HSZ/0403

Upconversion electroluminescence in VOPc via adsorption-induced interaction of unpaired spin — •Lukas Gerhard[1], Vibhuti Rai[1], Christof Holzer[1], Carsten Rockstuhl[2], and Wulf Wulfhekel[1] — [1]Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology (KIT), D-76344 Eggenstein-Leopoldshafen, Germany — [2]Institute of Theoretical Solid State Physics, Karlsruhe Institute of Technology (KIT), D-76131 Karlsruhe, Germany

Molecules with unpaired spins (radicals) promise favourable spin statistics in their electroluminescence. Here, we combine scanning tunneling microscopy induced luminescence and density functional theory to study single vanadyl phthalocyanine molecules, which are stable neutral radicals. Two distinct adsorption geometries of the molecule on NaCl/Au(111) lead to a difference in the interaction of the unpaired electron with the substrate, which in turn allows us to investigate its effects on the light emission process [1]. Remarkably, we observe that up-conversion electroluminescence is gated by the adsorption geometry of the molecule, an effect we attribute to a reordering of excited states and enhanced excited state transition probabilities. The profound influence of the unpaired electron via state reordering opens new possibilities for tuning not only molecular electroluminescence but also many other spin dependent phenomena.

[1] V. N. Rai, C. Holzer, C. Rockstuhl, W. Wulfhekel, and L. Gerhard, arXiv:2508.11501.

O 12.2 Mon 15:15 HSZ/0403

Machine Learning for Atomic-Scale Molecular Discovery from Tip-Enhanced Raman Spectroscopy — Harshit Sethi[1], Markus Juntilla[1], •Orlando J Silveira[1], and Adam S Foster[1,2] — [1]Aalto University, Espoo, Finland — [2]Kanazawa University, Kanazawa, Japan

Tip-enhanced Raman spectroscopy (TERS) combines the nanoscale spatial resolution of scanning probe microscopy with the chemical specificity of Raman spectroscopy, offering a powerful tool for resolving both molecular structure and composition. However, extracting precise atomic-scale information from complex TERS hyperspectral data remains a significant challenge. In this work, we address this gap by developing an automated machine learning framework to decode simulated TERS images for molecular structure discovery. Our approach uses convolutional neural networks (CNNs) to analyze hyperspectral datasets generated by modeling the plasmonic enhancement at a probe tip and the vibrational properties of planar molecules via density functional theory. To handle varying numbers of vibrational modes, we introduce a spectral binning procedure that standardizes the input for network training. This model provides a foundational proof of concept for the autonomous identification of molecular structures from TERS data, advancing the development of automated platforms for atomic-scale materials characterization and discovery.

O 12.3 Mon 15:30 HSZ/0403

THz-induced bilayer stacking dynamics in 1T-TaS$_2$ — Shaoxiang Sheng[1,2], Yang Yang[3], Mohamad Abdo[1], Li Chen[4], Peize Lin[5], •Kurt Lichtenberg[1], Xinguo Ren[3,5], Sheng Meng[3,5], Susanne Baumann[1], and Sebastian Loth[1,6] — [1]University of Stuttgart, Inst. for Functional Matter and Quantum Technologies — [2]Tsientang Inst. for Advanced Study — [3]IOP, Chinese Academy of Sciences — [4]Songshan Lake Materials Laboratory — [5]Inst. of Light Resources and Environmental Science, Henan Academy of Sciences — [6]Center for Integrated Quantum Science and Technology (IQST), University of Stuttgart

The electronic properties of layered materials change dramatically with variations in the interlayer coupling and the stacking order [1]. We employ terahertz pump-probe spectroscopy in conjunction with scanning tunneling microscopy (STM) to investigate the role of stacking order dynamics and of domain structures in layered 1T-TaS2 at the atomic scale. TaS2 exhibits diverse charge-density wave (CDW) phases and an insulating ground state at low temperatures [2]. We show that terahertz pulses can induce insulator-to-metal transitions (IMTs) and reveal hidden electronic phases in 1T-TaS2 by manipulating the bilayer stacking at femtosecond time and atomic length scales. This tunability promises unprecedented ultrafast control over the electronic phases also in other layered materials, suggesting applications in reconfigurable quantum devices and neuromorphic computing.

[1]: S.-H. Lee, PRL 122, 106404 (2019) [2]: V Petkov, J. Phys.: Cond. Matt. 34 345401 (2022)

O 12.4 Mon 15:45 HSZ/0403

Coherent coupling between molecular dipoles probed with STML — •Quentin Stein[1], Jakob Kuhlke[3], Song Jiang[1], Tomas Neuman[2], Michelangelo Romeo[1], Fabrice Scheurer[1], Katharina Kaiser[3], and Guillaume Schull[1] — [1]Université de Strasbourg, CNRS, IPCMS, UMR7504, F-67000 Strasbourg, France — [2]Institute of Physics, Czech Academy of Sciences Cukrovarnick'a 10, Prague, 16200, Czech Republic — [3]Georg-August-Universität Göttingen, Physikalishes Institut, 37077 Göttingen, Germany

The clustering of chromophores allows their excited states to hybridize, enabling the formation of coherently coupled dipoles spanning multiple molecules. This is of key importance for optical processes occurring in both artificial and natural molecular assemblies.

Probing these coherently coupled dipoles with atomic-scale precision cannot be achieved through conventional optical techniques because of the diffraction limit set by Abbe's law. This difficulty has recently been overcome by scanning tunneling microscopy induced luminescence (STML), which relies on the extreme confinement of the electromagnetic field in the plasmonic picocavity formed at the apex of a metallic STM-tip. The radiative decay rate of a molecular exciton placed in such a cavity is greatly increased, allowing for the detection of states that are typically optically dark. It also provides nanometer-resolved maps of molecular transition dipole moments, revealing the spatial character of the excitonic states with atomic precision. In this work, we used this approach to investigate the fluorescence of coherently coupled dipoles within simple molecular assemblies.

O 12.5 Mon 16:00 HSZ/0403

Continuous-wave all-optical microscopy at the atomic scale — •Valentin Bergbauer[1], Felix Schiegl[1], Svenja Nerreter[1], Valentin Giessibl[1], Fabian Sandner[1], Franz J. Giessibl[1], Yaroslav A. Gerasimenko[1], Tom Siday[2], Markus A. Huber[1], and Rupert Huber[1] — [1]Department of Physics and Regensburg Center for Ultrafast Nanoscopy (RUN), University of Regensburg, 93040 Regensburg, Germany — [2]School of Physics and Astronomy, University of Birmingham, Birmingham B15 2TT, U.K.

Exploring matter at the most fundamental level demands optical microscopy with ever-improving spatial resolution. By confining light to the apex of a sharp metallic tip, scanning near-field optical microscopy (SNOM) circumvents the diffraction limit, though its spatial resolution is still limited by the mesoscopic tip shape, inhibiting atomic resolution. Here, we observe atomically confined optical signals with Ångström-scale lateral modulations using continuous-wave mid-infrared radiation. These signals, characterized by a picometric vertical decay and a distinct optical phase shift, are consistent with emission from lightwave-driven tunnelling currents, a mechanism recently discovered using intense THz pulses. Observing near-field optical tunnelling emission (NOTE) under continuous-wave illumination is surprising, as strong-field effects such as coherent electron driving typically require ultrafast or free-electron lasers. Demonstrating the accessibility of this tunneling-mediated contrast mechanism with standard optical setups paves the way towards routine optical imaging with atomic-scale resolution.

O 12.6 Mon 16:15 HSZ/0403

Tuning Molecular Light Emission Through Atomic-scale Control of the Local Environment — •Thiago G. L. Brito[1], Daniel Arribas[1], Sofia Canola[2], Klaus Kuhnke[1], Tomáš Neuman[2], and Anna Rosławska[1] — [1]Max Planck Institute for Solid State Research, Heisenbergstr. 1, 70569 Stuttgart, Germany — [2]Institute of Physics, Czech Academy of Sciences, 16200 Prague, Czech Republic

Intermolecular distances, adsorption sites, and configurations define a molecule's local environment, but their combined effect on single-molecule emission across different excitation pathways remains unclear. In this study, we measure the singlet emission of zinc phthalocyanine (ZnPc) on NaCl/Ag(111) and introduce platinum phthalocyanine (PtPc) as both a controlled perturbation of the local environment and a probe of ZnPc emission via resonant energy transfer (RET). All measurements were performed using scanning tunneling microscopy induced luminescence (STML) and tip-enhanced photoluminescence (TEPL). By systematically tuning intermolecular distances, restricting molecular rotational motion, and comparing direct electrical excitation with RET-driven and photon excitations, we reveal previously inaccessible regimes of ZnPc singlet emission, including neutral exciton-libron coupling. We further disentangle the environmental factors that influence emission spectra, demonstrating how the local environment actively shapes the optical response of single molecules.

O 12.7 Mon 16:30 HSZ/0403

Dipole-dipole coupling in assembled ZnPc-molecule clusters — •Jakob Kuhlke[1], Quentin Stein[2], Song Jiang[2], Georg Traeger[1], Fabrice Scheurer[2], Tomáš Neuman[3], Katharina Kaiser[1], and Guillaume Schull[2] — [1]Georg-August University Göttingen, IV. Physical Institute, Göttingen — [2]CNRS, Institut de Physique et Chimie des Matériaux de Strasbourg, Département Surfaces et Interfaces, Strasbourg — [3]Institute of Physics, Czech Academy of Sciences Cukrovarnická 10, Prague, 16200, Czech Republic

Dipole-coupled molecules provide a promising platform to build functional structures with tailored optical properties. We use ZnPc molecules as building blocks to investigate the coupling of molecular dipoles in different molecular assemblies.

In contrast to ZnPc monomers, dimers and chains of ZnPc molecules exhibit sharper and red-shifted emission peaks, a phenomenon known as superradiance [1], [2], [3]. Here, we extend this approach to more arbitrary clusters. We use scanning tunneling microscopy-induced luminescence (STML) to study the optical properties of several cluster configurations on the nanometer scale.

[1] L. Yang, et al. Physical Review Letters 122.23 (2019)
[2] Z. Yang, et al. Nature 531.7596 : 623-627 (2016).
[3] J. Dolezal, et al. ACS nano 16.1 : 1082-1088 (2021).

O 12.8 Mon 16:45 HSZ/0403

Selectivity in tautomerization reaction pathways triggered in single molecules on ultrashort time scales — •Niklas Friedrich, Carmen Roelcke, Tzu-Chao Hung, Yaroslav Gerasimenko, Rupert Huber, and Jascha Repp — Institute of Experimental and Applied Physics and Regensburg Center for Ultrafast Nanoscopy (RUN), University of Regensburg, and Halle-Berlin-Regensburg Cluster of Excellence CCE, Germany

Lightwave-driven scanning tunnelling microscopy (LW-STM) is a powerful technique allowing to unravel dynamics of quantum systems on their intrinsic energy, spatial and temporal scales [1,2]. In LW-STM, the electric-field transient of ultrashort laser pulses focused into the junction of a tunnelling microscope acts as a bias-voltage transient and thereby controls tunnelling on ultrashort timescales.

Here, we explore a chemical reaction triggered on ultrashort timescales. We study free-base naphthalocyanine molecules (H2Nc) on 2ML NaCl on Cu(100). In DC-biased experiments, this system switches between different tautomers upon injection of high-energy tunnelling electrons [3]. We find that the same reaction is facilitated by ultrashort voltage transients in LW-STM for transients far exceeding the voltage of the lowest unoccupied molecular orbital. Furthermore, we observe different selectivities in the tautomerization reaction depending on the energy of the tunnelling electron that we tentatively assign to the orbital structure of H2Nc.

[1] Nat. Photon. 18, 595-602 (2024) [2] arXiv:2507.10206 (2025) [3] Science 317, 1203-1206 (2007)

O 12.9 Mon 17:00 HSZ/0403

Atomic-scale excitonic luminescence nanoscopy of moiré superlattices in van der Waals heterostructures — •Manas Pratim Biswas[1], Fábio J. R. Costa[1], Elise Jouaiti[1], Arnaud Gloppe[1], Katharina Kaiser[1,2], Fabrice Scheurer[1], Stéphane Berciaud[1], and Guillaume Schull[1] — [1]Institut de Physique et de Chimie des Matériaux de Strasbourg (IPCMS), Université de Strasbourg (CNRS, UMR 7504), Strasbourg, France — [2]4th Physical Institute-Solids and Nanostructures, Georg-August-Universität Göttingen, Göttingen, Germany

Atomically thin two-dimensional materials are susceptible to their environment, and stacking them into van der Waals heterostructures allows precise control of their electronic and optical properties. In semiconducting transition metal dichalogenides, excitons dominate the optical response whose properties can be tuned through interfacial interactions. Twisted and/or lattice-mismatched bilayers form a moiré superlattice that periodically modulates the electronic landscape. However, diffraction-limited optical methods average over micrometer scales, concealing these nanoscale effects. To overcome this, we employ cryogenic scanning tunneling microscope-induced luminescence[1] (STML, <7K, ultra-high vacuum) to probe exciton-moiré interplay in near-aligned WSe_2/WS_2 heterobilayers[2,3]. STML enables simultaneous atomic-scale imaging and local optical readout, with the ultimate aim of demonstrating nano-optical probing of moiré-modulated systems. References: 1. Nat. Mat. 22, 482 (2023) 2. Nat. Phys. 19, 1286 (2023) 3. Nat. Mat. 20, 945 (2021)

O 12.10 Mon 17:15 HSZ/0403

Tracing vibrational anharmonicity at the submolecular level with tip-enhanced Raman spectroscopy — •Youngwook Park[1], Ikutaro Hamada[2], Martin Wolf[1], and Akitoshi Shiotari[1] — [1]Fritz-Haber Institute of the Max-Planck Society, Berlin, Germany — [2]The University of Osaka, Suita, Japan

Tip-enhanced Raman spectroscopy (TERS) can achieve atomic-scale vibrational imaging, yet overtones and combination bands remain difficult to access due to weak transition strengths. We show that point-contact TERS enables submolecular-resolution detection of intense overtones and combination bands from a single asymmetric perylene derivative on silicon. A 10-times signal increase upon tip–molecule contact allows us to resolve both mechanical and electrical anharmonicities with Å-level spatial contrast. The mapped mechanical anharmonicity reveals a site-specific vibrational energy exchange pathway, demonstrating TERS as a powerful tool for real-space tracking of intramolecular vibrational energy redistribution.

O 12.11 Mon 17:30 HSZ/0403

Orbital-resolved imaging of coherent femtosecond exciton dynamics in single molecules — •Caiyun Chen — Max Planck Institute for Solid State Research, Heisenbergstraße 1, 70569 Stuttgart

Understanding and controlling excitonic wavepackets is fundamental to uncovering mechanisms underlying energy conversion dynamics in organic molecular systems. We present orbital-resolved imaging of coherent exciton dynamics in single metal-centered naphthalocyanine (MNc) molecules*CoNc, CuNc, and ZnNc*adsorbed on NaCl/Ag(111). Using femtosecond scanning tunneling microscopy, we directly observe time-resolved photocurrent signals and spectroscopic signatures of coherent excitons, enabling selective excitation and control of excitons in isolated and coupled molecules. In single CuNc molecules, we measure a decoherence time (˜70 fs), which decreases in dimers due to intermolecular interactions. Time-resolved measurements and photocurrent imaging reveal how central metal atoms in different naphthalocyanine molecules influence decoherence dynamics of the excited electronic state. Notably, a ZnNc molecule has longer decoherence time (˜ 300 fs) compared to the CuNc and CoNc (˜ 30 fs). Unpaired electrons on Co in CoNc molecule leads to distinct photocurrent imaging contrasts with the closed shell ZnNc and CuNc molecules. The origin of faster decoherence times of the excited states in CoNc and CuNc molecules is linked to conical intersections. These findings underscore the applications of ultrafast STM to probe quantum coherence and energy transfer dynamics at the single-molecule level with atomic-scale spatial resolution.