O 80: Scanning Probe Microscopy: Light Matter Interaction at Atomic Scales II

Time: Thursday 10:30–12:30 Location: MA 041

O 80.1 Thu 10:30 MA 041

All-optical subcycle microscopy with atomic resolution — •Valentin Bergbauer, Thomas Siday, Johannes Hayes, Felix Schiegl, Fabian Sandner, Peter Menden, Martin Zizlsperger, Svenja Nerreter, Sonja Lingl, Jascha Repp, Jan Wilhelm, Markus A. Huber, Yaroslav A. Gerasimenko, and Rupert Huber — Department of Physics and Regensburg Center for Ultrafast Nanoscopy (RUN), University of Regensburg, 93040 Regensburg, Germany

Near-field microscopy has prompted a revolution in nano-videography of quantum systems on subcycle timescales and with a spatial resolution down to ∼10 nm. Yet, the nanoscale geometry of the tip apex has prevented access to atomic resolution. We present a fundamentally new approach which brings all-optical microscopy to the atomic scale while retaining subcycle temporal resolution for the first time, by exploiting extreme nonlinearities within tip-confined evanescent light fields. We demonstrate the capabilities of this Near-field Optical Tunnelling Emission (NOTE) microscope by imaging nanometre-sized packing defects on the surface of gold, alongside tracing the subcycle quantum flow of electrons between the scanning tip and a semiconducting van der Waals trilayer in real-time. NOTE microscopy is not only compatible with insulating samples, where no rectified currents can flow but also enables us to combine all-optical subcycle spectroscopy with atomic resolution. In doing so, NOTE provides direct access to atomic scale quantum light-matter interaction and dynamics on their natural length and timescales.

O 80.2 Thu 10:45 MA 041

Excitation mechanism for STM-induced luminescence — •Song Jiang[1], Tomáš Neuman[2], Rémi Bretel[2], Alex Boeglin[1], Fabrice Scheurer[1], Eric Le Moal[2], and Guillaume Schull[1] — [1]Université de Strasbourg, CNRS, IPCMS, UMR 7504, F-67000 Strasbourg, France — [2]Institut des Sciences Moléculaires d'Orsay (ISMO), UMR 8214, CNRS, Université Paris-Saclay, 91405 Orsay Cedex, France

Fluorescence of neutral and charged molecules has been reported with subnanometer resolution in scanning tunneling microscope induced-luminescence (STML) experiments over the last years. Despite these significant developments, the detailed mechanisms of STML of individual molecules remained to be fully understood. Here, we proposed a "universal" model relying on successive carrier tunneling events that is based on a systematic experimental STML study of quinacridone (QA) molecules adsorbed on four-monolayer (4 ML) NaCl/Ag(111). Based on a comparison between the spatial, bias voltage, and tunneling current dependences of the charged and neutral emission, and on conductance data, a many-body description of the system is set. It reveals that four different charged states of QA (QA^-, QA^0, QA^+, and QA^{2+}), involving three different spin multiplicities, can be populated within a single voltage sweep. This excitation mechanism is backed up by data obtained for QA deposited on NaCl/Au(111) where the ground state of the molecule is a doubly positively charged singlet state (S^{2+}). This model bears a universal character that can be applied to STML experiments dealing with charged and neutral molecules.

O 80.3 Thu 11:00 MA 041

Structure and optical characterisation of Dibenzoterrylene on Au(111) and C_{60} — •Andreas Reutter[1,2], Yannis Hilgers[1,2], Franziska Hirt[2,3], Stefan Kück[2,3], Markus Etzkorn[1,2], and Uta Schlickum[1,2] — [1]Institut für Angewandte Physik, TU Braunschweig — [2]Laboratory for Emerging Nanometrology, TU Braunschweig — [3]Physikalisch-Technische Bundesanstalt, Braunschweig

Polycyclic aromatic hydrocarbons are promising candidates for molecule based single photon sources since they offer high brightness and narrow spectral emission lines. One interesting candidate is dibenzoterrylene (DBT, $C_{38}H_{20}$) which has recently been investigated in anthracene nanocrystals. However, anthracene tends to sublime at room temperature and the fabrication process has a low reproducibility. We have grown DBT on Au(111) and in C_{60} buffer layers in vacuum with organic molecular beam epitaxy. Using confocal laser scanning microscopy, we find photon emission from DBT clusters when embedded in a C_{60} Matrix. In addition, we have investigated the structure with low-temperature STM. For sub-monolayer coverage, DBT molecules self-assembly on the well-known Herringbone reconstruction on the Au(111) surface. Surprisingly, the DBT molecules prefer the fcc over the hcp areas of the herringbones in contrast to other molecules. We plan to fabricate DBT-C_{60} sandwich samples with individual DBT molecules implemented in the interlayer and to study their optical properties regarding line width and single photon characteristic of this system.

O 80.4 Thu 11:15 MA 041

Directional emission of plasmonic light by atomic-scale defects — •David Mateos[1,2], Óscar Jover[2], Miguel Varea[1,2], Koen Lauwaet[1], Daniel Granados[1], Rodolfo Miranda[1,2], Antonio Isaac Fernández Domínguez[3], Alberto Martín Jiménez[1], and Roberto Otero[1,2] — [1]IMDEA Nanoscience, Madrid, Spain — [2]Departamento de Física de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, Madrid, Spain — [3]Departamento de Física Teórica de la Materia Condensada and Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, Madrid, Spain

Manipulating the directionality of subwavelength light sources is demanded in broad applications, including sensing, photonics, optoelectronics, and energy harvesting. In this respect, nanoscale objects can control the angular distribution of scattered light below the diffraction limit. It has been demonstrated that properly engineering the size, shape, and arrangement of nanostructures in the few-nanometer scale affords control of the emission properties. Still, the effect of atomistic structures remains unexplored. In this work, we show that atomic-scale objects can also modify the directionality of plasmonic emission. The radiative density of optical states of the nanocavity formed by a sharp metallic tip at tunneling distance to a metallic surface is locally modified by a monoatomic height step. Comparison with electromagnetic calculations demonstrates that the observed changes arise from light emission tilting of the picocavity plasmons. Thus, atomic-like structures influence the light emission properties of nanoscale objects.

O 80.5 Thu 11:30 MA 041

Electroluminescence of VOPc in different adsorption configurations — Vibhuti Rai[1,2], •Lukas Gerhard[2], Christof Holzer[3], Carsten Rockstuhl[3,4], and Wulf Wulfhekel[2] — [1]Fachbereich Physik, Freie Universität Berlin, 14195 Berlin, Germany — [2]Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany — [3]Institute of Theoretical Solid State Physics, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany — [4]Institute for Nanotechnology, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany

In the last few years, scanning tunneling microscopy (STM) induced luminescence has mostly focused on planar molecules absorbed in a flat configuration. The main component of the electric field in the STM junction, however, is oriented perpendicular to the substrate. Here we show electroluminescence of Vanadyl-phthalocyanine (VOPc) deposited on NaCl on Au(111) which exhibits a strong dipole component perpendicular to the surface and different adsorption configurations [1]. As the structural change of VOPc is related to the spin structure [2], this offers a possibility to explain the observed differences in the light emission pattern in terms of charge state, lifetime and efficiency.

[1] S. Debnath et al, Angewandte Chemie 2022, 61 (25). [2] K. Kaiser et al, ACS Nano 2019, 13 (6).

O 80.6 Thu 11:45 MA 041

Electron and phonon driven mobility of the ammonia clusters adsorbed on a copper surface — •Prashant Srivastava[1], Daniel Miller[2], and Karina Morgenstern[1] — [1]Chair of physical chemistry I, Ruhr University of Bochum, Germany — [2]Department of Chemistry, Hofstra University, United States2Department of Chemistry, Hofstra University, United States

Understanding molecular kinetics induced by interfacial energy transfer at metal-molecule interfaces is important in molecular electronics and battery research. We explore the ultrashort laser pulse-induced mobility of ammonia clusters adsorbed on a copper surface at a single-molecule level by a combination of low-temperature scanning tunneling microscopy with an adjoined femtosecond laser and density functional theory. The diffusion of single molecules and desorption of molecules from clusters above an area of 1.35 nm2 is observed at laser fluences below 0.390 mJ/cm2. At higher laser fluences, single molecules desorb. In this talk, we present fluence-dependent changes in the ammonia coverage to understand the excitation mechanism. We discuss how the ammonia molecules and clusters are excited by electrons and phonons in a fluence range from 0.117 mJ/cm2 to 0.625 mJ/cm2. The study contributes to a control of molecular kinetics via interfacial energy transfer.

O 80.7 Thu 12:00 MA 041

TDDFT study of the orthosilicic acid molecule under electrostatic field and laser irradiation — •Tommaso Morresi — ECT* - FBK, Trento, Italy

The Tomographic Atom Probe (TAP) is a rapidly developing technique in the field of condensed matter analysis. Recently, a combination of TAP with intense THz pulses to control ion emission has been proposed [1]. This mechanism enables high spatial resolution and chemical sensitivity. In this configuration, the TAP analysis could be extended to bio-samples as an alternative to 4D microscopy. This investigation is the ultimate goal of the EIC Pathfinder project MIMOSA. Within this collaborative framework with a strong experimental endeavour, the theoretical understanding and computational modelling of the atom evaporation mechanism is of paramount importance. In this work, we study the interaction of THz-lasers with the orthosilicic acid molecule, which is the main ingredient of the matrix embedding the bio-material in the experimental setup. By using time-dependent density functional theory for electrons in combination with molecular dynamics for ions, we analyse the dynamics of laser-assisted field evaporation of OH ions from the molecule. While the scheme is similar to the one proposed in Ref. [2], the novelty of this work is mainly represented by the frequency of the laser in TDDFT simulations. We show the critical fields allowing the evaporation of OH ions in the THz regime and also the effect of additional external electrostatic fields on top of the time-dependent ones. [1] - A. Vella et al., Sci. Adv. 7, 7 (2021) [2] - E.P. Silaeva et al., PRB 92, 155401 (2015)

O 80.8 Thu 12:15 MA 041

Selectively Addressing Plasmonic Modes and Excitonic States in a Nanocavity Hosting a Quantum Emitter — •Alberto Martin-Jimenez[1], Oscar Jover[2], Koen Lauwaet[1], Daniel Granados[1], Rodolfo Miranda[1,2], and Roberto Otero[1,2] — [1]IMDEA Nanoscience, Madrid, Spain — [2]Dep. De Física de la Materia Condensada & IFIMAC, Universidad Autónoma de Madrid, Madrid, Spain

Controlling the interaction between the excitonic states of a quantum emitter and the plasmonic modes of a nanocavity is key for the development of quantum information processing devices. In this contribution we demonstrate that the tunnel electroluminescence of electrically insulated C60 nanocrystals enclosed in the plasmonic nanocavity at the junction of a scanning tunneling microscope can be switched from a broad emission spectrum, revealing the plasmonic modes of the cavity, to a narrow band emission, displaying only the excitonic states of the C60 molecules by changing the bias voltage applied to the junction. Interestingly, excitonic emission dominates the spectra in the high-voltage region in which the simultaneously acquired inelastic rate is low, demonstrating that the excitons cannot be created by an inelastic tunnel process. These results point toward new possible mechanisms for tunnel electroluminescence of quantum emitters and offer new avenues to develop electrically tunable nanoscale light sources.