O 43: Scanning Probe Microscopy: Light-Matter Interactions at the Atomic Scale II

Time: Tuesday 14:00–15:45 Location: H24

O 43.1 Tue 14:00 H24

Kinetics of nucleation and crystallization of sodium chloride based on frozen solution sample preparation apparatus — •Xinmeng Liu[1], Jiadong Guo[1], Yunzhe Jia[2], Sheng Meng[2], Enge Wang[1], and Ying Jiang[1] — [1]International Center for Quantum Materials, School of Physics, Peking University, Beijing, People's Republic of China — [2]Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing, People's Republic of China

Solution environment is ubiquitous and plays a vital role in various fields, especially in the nucleation and crystallization process. Here, we designed and built a new frozen solution sample preparation apparatus compatible with UHV environment, allowing atomic-scale SPM experiments. By utilization of this apparatus, we successfully transferred NaCl solution in glassy states onto the Au(111) substrate. The qPlus-based AFM with CO tip characterized the kinetics of ions nucleation and crystallization after annealing. We found that the ions tend to form disordered networks with water molecules at the initial stage of nucleation, and then adjust to ordered crystals. Furthermore, ions continue to crystallize on the formed island surface by way of chain growth. This is different from the previous view that NaCl nucleation follows the classical nucleation theory. In addition, we also captured the existence of a small crystal nucleus composed of several ions, which is surrounded by water molecules that help the nucleus to further grow. Our results provide new insights into solution process and will have significant effect on the mechanism of material synthesis.

O 43.2 Tue 14:15 H24

Wavefunction Reconstruction of Excitonic Edge States using Machine Learning — •Aritra Mishra, Sidhartha Nayak, and Alexander Eisfeld — Max Planck Institute for the Physics of Complex Systems

A typical problem in quantum mechanics is to reconstruct the eigenstate wave functions from measured data. In the case of molecular aggregates, the information about the excitonic eigenstates is important to understand the optical and transport properties [1]. The reconstruction of the wavefunction coefficients from the near field absorption spectra is shown for a linear and a 2D molecular arrangement [2].

Here, we consider the aggregates arranged in two sublattices in a 2D arrangement, each sublattice having a particular orientation of the molecular transition dipole moment, that shows topological edge states as described in [3]. We show the reconstruction of the excitonic wave function for $N = 50$ molecules and move to $N = 200$, in the presence of disorder in the molecular dipole orientations and noise in the spectra. We observe a better reconstruction for higher noises considering all the input spectra for the electric field polarisation of the nanotip along the x, y and z axes.

[1] X. Gao and A. Eisfeld, J. Phys. Chem. Lett. 9, 6003 (2018)
[2] F. Zheng, X. Gao and A. Eisfeld, Phys. Rev. Lett. 123, 163202 (2019)
[3] J. Yuen-Zhou, S. K. Saikin, N. Y. Yao and A. Aspuru-Guzik, Nature Materials 13, 1026 (2014)

O 43.3 Tue 14:30 H24

Probing topological Floquet states in graphene with ultrafast STM — •Nils Jacobsen[1,2], Melanie Müller[3], Michael Schüler[4,5], Martin Wolf[3], Angel Rubio[2,6], and Michael Sentef[1,2] — [1]ITP, University of Bremen, Bremen, Germany — [2]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany — [3]Fritz Haber Institute, Berlin, Germany — [4]Laboratory for Materials Simulations, Paul Scherrer Institut, Villigen, Switzerland — [5]Department of Physics, University of Fribourg, Fribourg, Switzerland — [6]Center for Computational Quantum Physics The Flatiron Institute, New York, USA

Floquet band engineering enables the control of solids via periodic laser driving. The light-induced quantum anomalous Hall effect (QAHE) in graphene with circularly polarized light [1] has been measured in ultrafast transport [2] and recently, Floquet replica bands under linearly polarized light have been reported in time-resolved photoemission spectroscopy [3]. Here, we explore the possibility of probing (topological) Floquet states in graphene with ultrafast scanning tunneling microscopy (USTM) as a complementary experimental technique [4]. Being highly sensitive to gap openings in the local density of states and the formation of edge states, USTM is a promising and versatile tool for probing light-induced topological states in quantum materials.

[1] Oka et al. PRB 79, 081406(R) (2009) [2] McIver et al. Nat. Phys. 16, 38-41 (2020) [3] Merboldt et al. arXiv:2404.12791 (2024) , Choi et al. arXiv:2404:14392 (2024) [4] Müller Prog. Surf Sci. 99, 100727 (2024)

O 43.4 Tue 14:45 H24

Theoretical Study of Electronic and Optical Properties in Edge-Modified Graphene Nanoribbons — •Jian Cheng Wong[1], Song Jiang[2], Sofia Canola[1], Alex Boeglin[2], Guillaume Schull[2], and Tomáš Neuman[1] — [1]Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10, 16200 Prague, Czech Republic — [2]Université de Strasbourg, IPCMS, CNRS, UMR 7504, F-67000 Strasbourg, France

Graphene nanoribbon (GNR) exhibits electronic and optical properties tunable by its geometry. One such approach is to introduce localized electronic states by modifying the edge structure. A previous study [1] using scanning tunneling microscopy-induced luminescence (STML) revealed that the presence of localized single-particle end states in GNR contributed to the formation of localized optical excitations. Here we provide an extensive theoretical description of such excitations on the modified edge structure of GNRs, and how it interacts with the end states. To that end, we develop a many body model that incorporates ab initio electronic structure methods and elucidate the sequence of events involved that leads to the eventual light emission under STML. From this model, we compare the electroluminescence maps obtained with experimental results and show the microscopic details of the localized states probed by STML.

[1] Song et al., Science, 379(6636), 1049-1054 (2023).

O 43.5 Tue 15:00 H24

Correlations between noise and electroluminescence in graphene nanojunctions — •Sascha Korn, Michael Krieger, and Heiko B. Weber — Lehrstuhl für Angewandte Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg

Light emission in scanning tunneling microscopy is often explained by the granularity of charge and should be therefore correlated with shot noise. Also, hot electrons may create both noise and thermal luminescence [1,2]. Using planar graphene nanojunctions, we study the phenomenon of electroluminescence in the point contact regime in a simple and well controlled electromagnetic environment. A spectral analysis of such measurements perfectly follows Planck's law and unambiguously supports the thermal picture. We present experimental data that correlate electrical noise measurements and optical spectroscopy, providing an in-depth view into the microscopic processes.

[1] Ott, C., Götzinger, S. & Weber, H. B. Thermal origin of light emission in nonresonant and resonant nanojunctions. Phys. Rev. Res. 2, 042019 (2020)
[2] Korn, S., Popp, M.A. & Weber, H.B. A point-like thermal light source as a probe for sensing light-matter interaction. Sci Rep 12, 4881 (2022)

O 43.6 Tue 15:15 H24

Photon blockade in current-driven single-molecule emitters — Andrés Bejarano[1,2], •Moritz Frankerl[1], Rémi Avriller[2], Fabio Pistolesi[2], and Thomas Frederiksen[1,3] — [1]Donostia International Physics Center, Spain — [2]Univ. Bordeaux, CNRS, LOMA, France — [3]Ikerbasque, Bilbao, Spain

We consider photon emission from a single electronic level embedded in a strongly damped cavity, where photon emission is driven by electronic tunneling events. Using a Lindblad master equation approach we investigate the system dynamics, photon emission spectrum as well as the second-order coherence function $g^{(2)}(\tau)$ [1]. We demonstrate that many features observed in scanning tunneling microscopy light-emission experiments can be explained with this simple model. Specifically, restricting the applied bias to the first emission threshold, we find antibunching in the photon statistics irrespective of the coupling strength to the cavity [2]. Employing a higher bias leads to a excitation of states with a photon number greater than one and thus the system shows bunching behavior as well as an emergence of two distinct time

scales in the dynamics of $g^{(2)}(\tau)$. [1] Q. Schaeverbeke, R. Avriller, T. Frederiksen, F. Pistolesi, PRL 123, 246601 (2019) [2] P.Merino, C. Große, A. Roslawska, K. Kuhnke, K. Kern, Nat. Comm. 6, 8461 (2015)

O 43.7 Tue 15:30 H24

Attosecond charge transfer in atomic-resolution scanning tunnelling microscopy — •Katharina Glöckl[1], Simon Maier[1], Raffael Spachtholz[1], Carlos Bustamante[2], Korbinian Pürckhauer[1], Franz J. Giessibl[1], Franco Bonafé[2], Markus A. Huber[1], Angel Rubio[2], Jascha Repp[1], and Rupert Huber[1] — [1]Department of Physics & Regensburg Center for Ultrafast Nanoscopy (RUN), Universität Regensburg — [2]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg

Scanning tunnelling microscopy (STM) driven with single-cycle terahertz pulses has afforded atomic-scale slow motion videos of single molecular orbitals. Driving tunnel currents with the carrier field of near-infrared light could improve the temporal resolution from ~100 fs down to attoseconds. Yet, competing multi-photon processes and thermal effects pose severe challenges in this spectral domain.

Here, we introduce an attosecond STM concept that is largely immune against thermal artifacts. By pulse synthesis, we periodically vary the waveform of single-cycle near-infrared pulses to drive tunnelling currents while keeping the thermal load on the tip constant. In a non-degenerate pump-probe scheme, we observe clear attosecond features in the sub-cycle currents and demonstrate atomic resolution by taking snapshot images of a single Cu adatom on a silver surface. Our results pave the way to recording the fastest relevant dynamics of electrons within atoms, molecules and quantum materials in actual attosecond atomic videography.