

Nanometer-scale Science and Technology Division Room A222 - Session NS+2D+QS-ThM

Direct Atomic Fabrication by Electron and Particle Beams & Flash Session

Moderators: Canhui Wang, National Institute of Standards and Technology (NIST), Xiaolong Liu, Northwestern University

8:00am **NS+2D+QS-ThM-1 Multiprobe Scanning Tunneling Microscopy and Spectroscopy: Atomic-level Understanding of Quantum Transport in Functional Systems**, *Marek Kolmer¹, W Ko, A Li*, Oak Ridge National Laboratory

Techniques based on multiprobe scanning tunneling microscopy (MP-STM) allow determination of charge and spin transport in variety of systems supported on surfaces of solid materials. In classical 2- and 4-probe methods STM tips are navigated by scanning electron microscope or high-resolution optical microscope typically in micrometer scales down to hundreds of nanometers. These MP-STM methods are currently regarded as universal tools for in-situ characterization of mesoscopic transport phenomena [1,2].

Such a mesoscopic experimental paradigm has recently been changed by downscaling of 2-probe STM experiments towards the atomic level [3,4]. In this case current source and drain probes are positioned in atomically defined locations with respect to the characterized nanosystems. Our experiments rely on fully STM-based tip positioning protocol with probe-to-probe separation distances reaching tens of nm [3,4]. Such probe-to-probe lateral positioning precision is combined with about pm vertical sensitivity in probe-to-system contacts. These two factors enable realization of two-probe scanning tunneling spectroscopy (2P-STs) experiments, where transport properties can be characterized by macroscopic probes kept in atomically defined tunneling conditions [4].

Here, we will apply 2P-STs methodology to probe quantum transport properties in functional systems: graphene nanoribbons (GNRs) epitaxially grown on the sidewalls of silicon carbide (SiC) mesa structures. These GNRs display ballistic transport channels with exceptionally long mean free paths and spin-polarized transport properties as proven by mesoscopic multiprobe transport experiments [5-7]. Interestingly, the nature of these ballistic channels remains an open question. We will show that 2P-STs experiments give new insight into quantum origin of the transport behaviors.

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- [4] Kolmer M. et al., *Nat. Commun.*, 10, 1573 (2019)
- [5] Baringhaus J., *Nature*, 506, 349-354 (2014)
- [6] Aprojan J. et al., *Nat. Commun.*, 9, 4426 (2018)
- [7] Miettinen A.L. et al., submitted, arXiv:1903.05185 (2019)

This work was conducted at the Center for Nanophase Materials Sciences (CNMS), which is a DOE Office of Science User Facility.

8:20am **NS+2D+QS-ThM-2 Light and Heavy Ions from New Non-classical Liquid Metal Ion Sources for Advanced Nanofabrication**, *Paul Mazarov*, RAITH GmbH, Germany; *T Richter, L Bruchhaus, R Jede*, Raith GmbH; *Y Yu, J Sanabia*, Raith America; *L Bischoff*, Helmholtz Zentrum Dresden-Rossendorf, Germany; *J Gierak*, CNRS—Université Paris-Sud, France
INVITED

Nanofabrication requirements for FIB technologies are specifically demanding in terms of patterning resolution, stability and the support of new processing techniques. Moreover the type of ion defines the nature of the interaction mechanism with the sample and thus has significant consequences on the resulting nanostructures [1]. Therefore, we have extended the technology towards the stable delivery of multiple ion species selectable into a nanometer scale focused ion beam by employing a liquid metal alloy ion source (LMAIS) [2]. This provides single and multiple charged species of different masses, resulting in significantly different interaction mechanisms. Nearly half of the elements of the periodic table are made available in the FIB technology as a result of continuous research in this area [3]. This range of ion species with different mass or charge can

be beneficial for various nanofabrication applications. Recent developments could make these sources to an alternative technology feasible for nanopatterning challenges. In this contribution the operation principle, the preparation and testing process as well as prospective domains for modern FIB applications will be presented. As example we will introduce a GaBiLi LMAIS [4]. It enables high resolution imaging with light Li ions and sample modification with Ga or heavy polyatomic Bi clusters, all coming from one ion source. For sub-10 nm focused ion beam nanofabrication and microscopy, the GaBiLi-FIB or the AuSiGe-FIB could benefit of providing additional ion species in a mass separated FIB without changing the ion source.

References

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- [3] J. Gierak, P. Mazarov, L. Bruchhaus, R. Jede, L. Bischoff, *Review of electrohydrodynamical ion sources and their applications to focused ion beam technology*, *JVSTB* 36, 06J101 (2018).
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9:00am **NS+2D+QS-ThM-4 Visualizing the Interplay between Spatial and Magnetic Confinement in Graphene Quantum Dots**, *Joseph Stroschio*, National Institute of Standards and Technology (NIST)
INVITED

At the heart of the wave nature of quantum mechanics is the quantization of energy due to quantum confinement, taking place when the particle's de Broglie wavelength becomes comparable to the system's length scale. In a quantum dot (QD), electrons are confined in all lateral dimensions using geometric constraints or a combination of electric and magnetic fields. Being a tunable quantum workbench, QDs have found a ubiquity of applications. Behaving as artificial atoms they have found extensive use as qubits in quantum information technologies, and tools for emulating basic models of condensed-matter physics. QDs offer an ideal platform for studying the interplay between quantum confinement, caused by spatial constraints or by large magnetic fields via cyclotron motion, and interaction effects. Recently, the ability to apply local nanometer scale gate potentials in graphene heterostructures has enabled the creation of QDs for Dirac quasiparticles. Graphene QDs are formed inside circular p-n junctions, where one has detailed control of electron orbits by means of local gate potentials and magnetic fields. We study the interplay between spatial and magnetic confinement using scanning tunneling spectroscopy measurements of the energy spectrum of graphene QDs as a function of energy, spatial position, and magnetic field. In zero field, the Dirac quasiparticles are confined by Klein scattering at large incident angle at the p-n junction boundary. The confined carriers give rise to an intricate eigenstate spectrum, effectively creating a multi-electron artificial atom. Applying a weak magnetic field results in a sudden and giant increase in energy for certain angular momentum states of the QD, creating a discontinuity in the energy spectrum as a function of magnetic field. This behavior results from a π -Berry phase associated with the topological properties of Dirac fermions in graphene, which can be turned on and off with magnetic field. With increased applied magnetic field, the QD states are observed to condense into Landau levels, providing a direct visualization of the transition from spatial to magnetic confinement in these artificial graphene atoms. With further increase in magnetic fields, an intricate interplay between Coulomb charging of compressible Landau levels separated by incompressible rings emerges, which we map as a function of energy, spatial position, and magnetic field utilizing the exceptional capabilities of scanning tunneling spectroscopy.

9:40am **NS+2D+QS-ThM-6 Using Controlled Manipulation of Molecules to Trace Potential Energy Surfaces of Adsorbed Molecules**, *O Dagdeviren, C Zhou*, Yale University; *M Todorovic*, Aalto University, Finland; *Eric Altman, U Schwarz*, Yale University

The development of scanning probe microscopy techniques has enabled the manipulation of single molecules. More recently it has been demonstrated that the forces and energy barriers encountered along the manipulation path can be quantified using non-contact atomic force microscopy (AFM). To explore the practicality of using this novel approach to experimentally measure the energy barriers an adsorbed molecule encounters as it moves across a surface decorated by other molecules

¹ NSTD Early Career Award Finalist

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including potential reaction partners, we have been studying benzene molecules on Cu (100) as a model system. We first choose a specific manipulation path and then move the tip repeatedly along this path as the tip-sample distance is reduced while recording the AFM cantilever oscillation amplitude and phase. To preserve the accuracy of the recovered tip-sample interaction potentials and forces, we use oscillation amplitudes significantly larger than the decay length of the tip-sample interaction potential are used. Operating the microscopy in the tuned-oscillator mode and analyzing the resulting cantilever oscillation amplitude and phase as functions of the spatial coordinates allows recovery of the potential energy of the interaction between the tip and the sample, the force on the tip normal to the surface, and the lateral force acting on the tip along the manipulation path, all as functions of tip vertical and lateral position with 0.01 Å resolution. In over 50 distinct manipulation events, the molecules were either pushed, pulled, jumped to the tip, or did not move depending on the chemical environment surrounding the molecule and the chemical identity of the tip. For further insight, we have compared the experimentally measured energy landscapes and manipulation outcomes with computational results obtained using a Bayesian Optimization Structure Search protocol.

11:00am **NS+2D+QS-ThM-10 Direct Writing of Functional Heterostructures in Atomically Precise Single Graphene Nanoribbons**, **Chuanxu Ma**, Oak Ridge National Laboratory; **Z Xiao**, North Carolina State University; **J Huang**, **L Liang**, Oak Ridge National Laboratory; **W Lu**, North Carolina State University; **K Hong**, **B Sumpter**, Oak Ridge National Laboratory; **J Bernholc**, North Carolina State University; **A Li**, Oak Ridge National Laboratory

Precision control of interfacial structures and electronic properties is the key to the realization of functional heterostructures. Here, utilizing the scanning tunneling microscope (STM) both as a manipulation and characterization tool, we demonstrate the fabrication of a heterostructure in a single atomically precise graphene nanoribbon (GNR) and report its electronic properties¹. The heterostructure is made of a seven-carbon-wide armchair GNR (7-aGNR) and a lower band gap intermediate ribbon synthesized bottom-up from a molecular precursor on an Au substrate. The short GNR segments are directly written in the ribbon with an STM tip to form atomic precision intraribbon heterostructures. Based on STM studies combined with density functional theory calculations, we show that the heterostructure has a type-I band alignment, with manifestations of quantum confinement and orbital hybridization. We further investigate the negative differential resistance (NDR) devices using the GNR heterostructure based double-barrier models². Our computational results indicate that nanoscale engineering for NDR needs to consider atomic size effect in design and atomic precision in fabrication. This combined theoretical-experimental approach opens a new avenue for the design and fabrication of nanoscale devices with atomic precision.

References

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2. Xiao, Z.; Ma, C.; Huang, J.; Liang, L.; Lu, W.; Hong, K.; Sumpter, B. G.; Li, A. P.; Bernholc, J. Design of Atomically Precise Nanoscale Negative Differential Resistance Devices. *Adv. Theory Simul.* **2018**, *2*, 1800172.

11:20am **NS+2D+QS-ThM-11 Effects of Helium and Neon Processing on 2D Material Properties**, **Alex Belianinov**, Oak Ridge National Laboratory; **S Kim**, Pusan National University, South Korea; **V Iberi**, **S Jesse**, **O Ovchinnikova**, Oak Ridge National Laboratory

Recent advances in CVD-growth consistently yield high quality 2D materials for large(er) scale fabrication. Monolayers of molybdenum and tungsten diselenide and selenide, graphene, and other exotic 2D materials are becoming routine in fabrication of functional electronic and optoelectronic devices. In order to attain novel functionalities, it is critical to tune and engineer defects in 2D materials directly with nanometer precision. Advances in ion beam-based imaging and nanofabrication techniques have offered a pathway to precisely manipulate 2D materials and offer a roadmap to create junctions, amorphized areas, and introduce dopants for new types of electronic devices. Here, we demonstrate the use of a focused helium and neon ion beams in a scanning helium ion microscope (HIM) in tailoring material functionality in MoSe₂, WSe₂, CuInP₂S₆ and graphene.

The helium ion microscope can “direct-write” capabilities, capable of both imaging and nanofabrication with Helium and Neon gases, thus making it an excellent candidate for processing a wide range of 2D, and conventional
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materials. We explore milling by the helium and neon ion beams of suspended and supported samples in order to control material’s electronic and mechanical properties. We validate the results with other chemical imaging techniques such as Scanning Transmission Electron Microscopy, correlated band excitation (BE) scanning probe microscopy, and photoluminescence (PL) spectroscopy.

Acknowledgement

This work was conducted at the Center for Nanophase Materials Sciences (CNMS), which is a U.S. Department of Energy (DOE) Office of Science User Facility.

11:40am **NS+2D+QS-ThM-12 Operating Molecular Propeller in Quantum Regime with Directional Control**, **Y Zhang**, **Tolulope Ajayi**, Ohio University; **J Calupitan**, Université de Toulouse, France; **R Tumbleson**, Ohio University; **G Erbland**, **C kammerer**, CEMES-CNRS, France; **S Wang**, Ohio University; **L Curtiss**, **A Ngo**, Argonne National Laboratory; **G Rapenne**, NAIST, Japan; **S Hla**, Ohio University

Synthetic molecular machines are fascinating and have a great promise to revolutionize a large scientific and technology fields. The immense interest to this research area is evident by the 2016 Nobel Prize in Chemistry awarded for the design and synthesis of molecular machines. Unlike biological molecular machines, which typically have the sizes of a few microns, artificial machines operating at the nanometer scale are in the quantum regime. Here, we have developed a robust multi-component molecular propeller that enables unidirectional rotations on a materials surface when they are energized. Our propeller system is composed of a stator having a ratchet-shaped molecular gear designed to anchor on a gold surface and a rotator with three molecular blades. By means of scanning tunneling microscope imaging and manipulation, the rotation steps of individual molecular propellers are directly visualized, which confirms the unidirectional rotations of both left and right handed molecular propellers into clockwise and counterclockwise directions, respectively. Moreover, the mechanical manipulation of the molecular with the scanning probe tip further reveal detailed rotation mechanism, thereby opening a new research direction to investigate mechanical properties of the molecular machines with an atomic level precision.

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