Thursday Morning, October 24, 2019

Advanced Ion Microscopy and Ion Beam Nano-engineering Focus Topic
Room B231-232 - Session HI+NS-ThM

Novel Beam Induced Material Engineering and Nano-Patterning

Moderators: Olga S. Ovchinnikova, Oak Ridge National Laboratory, Shinichi Ogawa, National Institute of Advanced Industrial Science and Technology (AIST)

8:00am **HI+NS-ThM1 Tuning out-of-plane Piezoelectricity in 2D Materials using Ion Beams***, **Yunseok Kim,*** Sungkyunkwan University, Republic of Korea **INVITED**

Two-dimensional (2D) transition metal dichalcogenides (TMDs) have been extensively studied owing to their ultra-thin nature as well as superior material properties. In particular, after the experimental observation of intrinsic in-plane piezoelectricity in the 2D MoS$_2$, fundamental studies on the piezoelectricity as well as piezoelectric device applications of the 2D TMDs have attracted significant interest. However, their applications are strongly limited due to the fact that crystallographically only in-plane piezoelectricity exists in the 2D TMDs. In this presentation, I will summarize our recent effect on the realization of tunable out-of-plane piezoelectricity in the 2D TMDs using He ion beams. Among various 2D TMDs, we have chosen MoTe$_2$ because it is very sensitive to the external stimuli such as strain. We first examined the realization of the out-of-plane piezoelectriciy by local asymmtery breaking based on the surface corrugation to check its feasibility. Then, He ion irradiation as a function of dose were performed onto the MoTe$_2$ surface. It was found that the out-of-plane piezoelectricity was indeed induced by He ion beams and, further, the magnitude of the induced out-of-plane piezoelectricity was depedent on the dose level. The proposed strategy for modulation of tunable out-of-plane piezoelectricity can be easily applied to a broader class of 2D TMD materials that have not been used for applications with out-of-plane piezoelectricity. Accordingly, it can stimulate the expansion of practical energy device applications with 2D TMDs.

8:40am **HI+NS-ThM3 Defect Engineering of Ferroelectric Thin Films – Leveraging Ion Beams for Improved Function***, **Lane Martin,*** University of California at Berkeley **INVITED**

Modern approaches to epitaxial thin-film growth have enabled unprecedented control of ferroelectric materials including the realization of enhanced polarization and ordering temperatures, production of ordered-domain structures, and improved properties. Today we are looking beyond simple lattice mismatch control for new ways to manipulate and control ferroic response and to produce unexpected or emergent effects. In this talk, we will investigate a number of observations of such emergent or unexpected properties in epitaxial thin films made possible via innovative synthesis and processing methodologies. In particular, we will explore recent examples of how synthesis, defects, and epitaxial constraint can be combined to produce exotic effects in ferroic systems. Primary focus will be given to the *ex situ* production of defects with ion bombardment to control defect-induced electronic states that can drive dramatic changes in leakage currents and impact ferroelectric response in materials like BaTiO$_3$, PbTiO$_3$, BiFeO$_3$, and others. For example, we will explore how high-energy-ion beams (>3 MeV beams of helium ions) can induce nonequilibrium densities of intrinsic point and defect clusters that have unintended positive effects – including reducing leakage in films by as much as 3-4 orders of magnitude, tuning coercive fields for switching, and much more. At the same time, leveraging focused-helium-ion bombardment, it is possible to create nanoscale patterns of defect-engineered material where emergent function, such as multi-state switching, is accomplished. Finally, we will explore how ion-bombardment procedures can also provide a knob to tune local energy competition in materials like relaxor ferroelectrics to gain new insight into material physics. All told, we will highlight specifics about the routes to produce defect-engineered ferroelectric thin films, will explore approaches to characterize and study the nature of defects that are produced – including application of techniques like deep-level transient spectroscopy, and will examine the implication of such defect structures for dielectric and ferroelectric properties – including studies of defect-based effects on switching processes and kinetics. We will end with an exploration of what further growth of defect-engineering approaches might enable in the way of novel function and applications in these materials.

9:20am **HI+NS-ThM5 Exploring Proximity Effects and Large Depth of Field in Helium Ion Beam Lithography: Large-area Dense Patterns and Tilted Surface Exposure***, **Ranveig Flatabø,*** Univeristy of Bergen, Norway*; A. Agarwal,* Massachusetts Institute of Technology*; R. Hobbs,* Trinity College Dublin*; M. M. Greve,* Univeristy of Bergen*; B. Holst,* Univeristy of Bergen, Norway*; K.K. Berggren,* Massachusetts Institute of Technology

Helium ion beam lithography (HIL) is an emerging nanofabrication technique. It benefits from a reduced interaction volume compared to that of an electron beam of similar energy, and hence reduced long-range scattering (proximity effect), higher resist sensitivity and potentially higher resolution. Furthermore, the small angular spread of the helium ion beam gives rise to a large depth of field. This should enable patterning on tilted and curved surfaces without the need of any additional adjustments, such as laser-auto focus. So far, most work on HIL has been focused on exploiting the reduced proximity effect to reach single-digit nanometer resolution, and has thus been concentrated on single-pixel exposures over small areas. Here we explore two new areas of application. Firstly, we investigate the proximity effect in large-area exposures and demonstrate HIL's capabilities in fabricating precise high-density gratings on large planar surfaces (100 µm × 100 µm, with pitch down to 35nm) using an area dose for exposure. Secondly, we exploit the large depth of field by making the first HIL patterns on tilted surfaces (sample stage tilted 45°). We demonstrate a depth of field greater than 100 µm for an estimated resolution of 20 nm.

9:40am **HI+NS-ThM6 Fabrication of Plasmonic Nanostructures by Helium-Ion Milling***, **André Beyer**, M. Westphal,* Bielefeld University, Germany*; S. Stephan,* Oldenburg University, Germany*; D. Emmrich, H. Vieker,* Bielefeld University, Germany*; K. Chen,* Jinan University, Guangzhou, China*; G. Razinskas, H. Gross, B. Hecht,* Würzburg University, Germany*; M. Silies,* Oldenburg University, Germany*; A. Gölzhäuser,* Bielefeld University, Germany

Plasmonic nanostructures are essential for controlling and directing light on the nanoscale. While fabrication techniques like standard electron beam lithography (EBL) methods or focused ion beam (FIB) milling with Ga$^+$ ions are approaching their limit in the 10-nm-regime, ion beam milling with He$^+$ ions is capable of milling features below 6 nm [1,2]. In this contribution, we give two specific examples of helium-ion milled plasmonic nanostructures: (i) gold bowtie antennas milled from 100 nm thick polycrystalline gold films on mica substrates and (ii) nanoslit cavities in chemically-synthesized 40 nm thick single-crystalline gold flakes [2]. Both examples benefit from a combined approach using a Ga$^+$ FIB for milling large features and employing the fine resolution of the helium ion microscope (HIM) for milling small features. We will discuss different patterning strategies to optimize the writing speed and minimize substrate swelling. In addition, our approach to quantify the sizes of milled gaps will be shown. It is based on low dose imaging in combination with substantial line-profile averaging which we applied to few-hundred-nanometer-long homogeneous helium-ion milled lines.

[1] H. Kollmann et al., Nano Letters 14, 4778 (2014).

[2] K. Chen et al., Nanoscale 10, 17148 (2018).

11:00am **HI+NS-ThM10 Towards Atomically Precise Carbon Quantum Electronic Devices***, J.L. Swett,* University of Oxford, UK*; O. Dyck, S. Jesse,* Oak Ridge National Laboratory*; **Jan Mol,*** Queen Mary University of London, UK **INVITED**

Towards Atomically Precise Carbon Quantum Electronic Devices

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Graphene exhibits many unique properties that can be further enhanced through nanostructuring and atomic manipulation. Such nanostructured devices have potential applications as molecular junctions [1], spin qubits [2], heat engines [3], and sensors [4], providing substantial motivation for their realization. Electron and ion beams provide unique and complementary tools for realizing some of these structures due to their ability to modify the graphene with atomic and nanoscale precision, respectively. Modification may take the form of direct-write patterning [5], defect production [6], dopant introduction [7], and dopant manipulation [8]. Although much progress has been realized in these areas, transport measurements of top-down fabricated atomically precise carbon nanostructures have yet to be realized. Here we present lessons learned and key findings for this emerging direction of research leveraged from years of fabrication and transport measurements of single molecules via non-covalent bonding to graphene nanoelectrodes [9]. We will present a broad overview of the challenges and progress in understanding and controlling the transport through atomic-scale devices and discuss how these lessons inform and translate to current experiments on introducing dopants and manipulating atoms on the atomic scale with electron and ion beams in graphene and other 2D materials. Finally, practical strategies for realization of these devices will be discussed, including contamination control, fabrication strategies, and transport measurements.

References:

[1] J.K. Sowa et al., J. Chem. Phys. 149, 154112 (2018)

[2] Trauzettel, Björn, et al., Nature Physics 3.3, 192 (2007).

[3] P. Gehring et al., Nano Lett. 17, 7055 (2017)

[4] P. Puczkarski et al., ACS Nano 12, 9451 (2018)

[5] Nanda, Gaurav, et al., Carbon 119, 419-425, (2017)

[6] Robertson, Alex W., et al., Nature communications 3 1144 (2012)

[7] Tripathi, Mukesh, et al., ACS nano 12.5 4641-4647 (2018)

[8] Dyck, Ondrej, et al., Small 14.38 1801771 (2018)

[9] C.S. Lau et al., Phys. Chem. Chem. Phys. 16, 20398 (2014)

11:40am **HI+NS-ThM12 Fabrication of High-Q nanofiber Bragg Cavity Using a Helium Ion Microscope***, **Hideaki Takashima,*** Kyoto university, Japan*; A. Fukuda, H. Maruya, T. Tashima,* Kyoto University, Japan*; A. Schell,* Central European Institute of Technology, Czech Republic*; S. Takeuchi,* Kyoto University, Japan

Efficient coupling between single light emitters and photons propagating in single mode fibers has been attractive attention recently for the realization of photonic quantum information devices, such as single photon sources, and quantum phase gates. Toward the realization of these devices, we have developed nanofiber Bragg cavity (NFBC), which is an optical nanofiber embedded in a microcavity in it, using a gallium focused ion beam (FIB) milling system. The NFBC has small mode volume of wavelength size, ultra-wide tunability of the resonant wavelength, and high coupling efficiency (>80%). However, experimentally achieved quality (Q) factors have been still a few hundreds. Here, we report the development of the NFBC using a helium ion microscope (ZEISS “ORION NanoFab”).

Nanofibers are fabricated by heating a single-mode fiber with a ceramic heater and stretching the end of the fiber. The diameter of the nanofibers is reduced to about 300 nm. The helium ion beam is periodically irradiated from the top side of the nanofiber to fabricate Bragg grating. The period at the center of the grating is modified for introducing a defect to be worked as a microcavity.

In order to evaluate the Q factor of the NFBC, we measure a transmission spectrum. The light of a halogen lamp is connected to the one end of the NFBC and the transmitted light is observed with a spectrometer with the resolution of 0.17 nm.

When we measure the transmission spectrum of the NFBC with the grooves of 320, a sharp resonant peak with the linewidth of 0.54 nm was observed in the center of the stop band. This agrees with the Q factor of 1260, which is more than 4 times larger than the NFBC fabricated with the Ga FIB system (Q ~ 300). Taking into account of the resolution of the spectrometer, it is expected that the real Q factor would be higher than this value.

In conclusion, we reported the fabrication of NFBC using the helium microscope. When the number of the grooves is 320, the Q factor is 1260, which is more than 4 times larger than the NFBC fabricated by the Ga FIB system.

Besides this result, we will discuss the NFBC when the number of the grooves is changed and the comparison with finite-difference time-domain (FDTD) simulation.

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12:00pm **HI+NS-ThM13 Time of Flight Secondary Ion Mass Spectrometry in the Helium Ion Microscope for Battery Materials and Other Nanoscale Problems***, N. Klingner,* Helmholtz Zentrum Dresden-Rossendorf, Germany*; **Gregor Hlawacek,*** Helmholtz-Zentrum Dresden Rossendorf, Germany*; L.J. Wheatcroft, B.J. Inkson,* University of Sheffield, UK*; R. Heller,* Helmholtz Zentrum Dresden-Rossendorf, Germany

Helium Ion Microscopy (HIM) has become a wide spread imaging and nanofabrication technology. However, existing HIM users can currently not perform elemental analysis in an easy and cost efficient way. We present results obtained using a light weight retrofitable Time of Flight Secondary Ion Mass Spectrometer (TOF-SIMS). I will briefly give an overview on new developments in our TOF-SIMS setup which allows to obtain information on the elemental composition of the sample. The lateral resolution for the presented TOF-SIMS add-on has been measured to be 8 nm. A particular advantage of the presented TOF-SIMS implementation is that it allows for charge compensation during data acquisition and thus the elemental analysis of insulators or poorly conducting materials. In addition delayed extraction can be realized which will allow a field free application of the primary beam which reduces aberrations and the setup time. While not a dedicated high mass resolution instrument it allows to answer many scientific questions by combining the high lateral resolution of the HIM with elemental information. The examples include but are not limited to battery materials and corrosion protection of steel.

[1] Nico Klingner, Rene Heller, Gregor Hlawacek, Stefan Facsko, and Johannes von Borany. Time-of-

flight secondary ion mass spectrometry in the helium ion microscope. Ultramicroscopy, 198:10–17,

2019.

[2] Nico Klingner, René Heller, Gregor Hlawacek, J. von Borany, John A. Notte, Jason Huang, and

Stefan Facsko. Nanometer scale elemental analysis in the helium ion microscope using time of flight

spectrometry. Ultramicroscopy, 162:91–97, 2016.

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