Advanced Ion Microscopy and Ion Beam Nano-engineering Focus Topic
Room B231-232 - Session HI+AS+CA-WeA

Advanced Ion Microscopy and Surface Analysis Applications

Moderators: Richard Livengood, Intel Corporation, USA, Armin Gölzhäuser, Bielefeld University, Germany

2:20pm **HI+AS+CA-WeA1 Analytical Capabilities on FIB Instruments using SIMS: Applications, Current Developments and Prospects**, ***Tom Wirtz,*** Luxembourg Institute of Science and Technology, Luxembourg*; J.-N. Audinot,* Luxembourg Institute of Science and Technology, Luxembourg, Luxembourg*; J. Lovric, O. De Castro,* Luxembourg Institute of Science and Technology, Luxembourg **INVITED**

Secondary Ion Mass Spectrometry (SIMS) is an extremely powerful technique for analyzing surfaces, owing in particular to its ability to detect all elements from H to U and to differentiate between isotopes, its excellent sensitivity and its high dynamic range. SIMS analyses can be performed in different analysis modes: acquisition of mass spectra, depth profiling, 2D and 3D imaging. Adding SIMS capability to FIB instruments offers a number of interesting possibilities, including highly sensitive analytics, in-situ process control during patterning and milling, highest resolution SIMS imaging (~10 nm), and direct correlation of SIMS data with data obtained with other analytical or imaging techniques on the same instrument, such as high resolution SE images or EDS spectra [1,2].

Past attempts of performing SIMS on FIB instruments were rather unsuccessful due to unattractive detection limits, which were due to (i) low ionization yields of sputtered particles, (ii) extraction optics with limited collection efficiency of secondary ions and (iii) mass spectrometers having low duty cycles and/or low transmission. In order to overcome these limitations, we have investigated the use of different primary ion species and of reactive gas flooding during FIB-SIMS and we have developed compact high-performance magnetic sector mass spectrometers operating in the DC mode with dedicated high-efficiency extraction optics. We installed such SIMS systems on different FIB based instruments, including the Helium Ion Microscope [3-5], a FIB-SEM DualBeam instrument and the npSCOPE instrument, which is an integrated Gas Field Ion Source enabled instrument combining SE, SIMS and STIM imaging with capabilities to analyse the sample under cryo-conditions.

Here, we will review the performance of the different instruments with a focus on new developments such as cryo-capabilities and new detectors allowing parallel detection of all masses, present a number of examples from various fields of applications (nanoparticles, battery materials, photovoltaics, micro-electronics, tissue and sub-cellular imaging in biology, geology,…) and give an outlook on new trends and prospects.

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[2] F. Vollnhals, J.-N. Audinot, T. Wirtz, M. Mercier-Bonin, I. Fourquaux, B. Schroeppel, U. Kraushaar, V. Lev-Ram, M. H. Ellisman, S. Eswara, Anal. Chem. 89 (2017) 10702

[3] D. Dowsett, T. Wirtz, Anal. Chem. 89 (2017) 8957

[4] T. Wirtz, D. Dowsett, P. Philipp, Helium Ion Microscopy, ed. by G. Hlawacek, A. Gölzhäuser, Springer, 2017

[5] T. Wirtz, O. De Castro, J.-N. Audinot, P. Philipp, Ann. Rev. Anal. Chem. 12 (2019)

3:00pm **HI+AS+CA-WeA3 Correlated Materials Characterization via Multimodal Chemical Imaging using HIM-SIMS**, *A. Belianinov,* Oak Ridge National Laboratory*; S. Kim,* Pusan National University, South Korea*; A. Trofimov,* ***Olga S. Ovchinnikova,*** Oak Ridge National Laboratory

Multimodal chemical imaging simultaneously offers high resolution chemical and physical information with nanoscale, and in select cases atomic, resolution. By coupling modalities that collect physical and chemical information, we can address a new set of scientific problems in biological systems, battery and fuel cell research, catalysis, pharmaceuticals, photovoltaics, medicine and many others. The combined multimodal platforms enable local correlation of material properties with chemical makeup, making fundamental questions in how chemistry and structure drive functionality approachable. The goal of multimodal imaging is to transcend the existing analytical capabilities for nanometer scale spatially resolved material characterization at interfaces through a unique merger of advanced microscopy, mass spectrometry and optical spectroscopy.Combining helium ion microscopy (HIM) and secondary ion mass spectrometry (SIMS) onto one platform has been demonstrated as a method for high resolution spot sampling and imaging of substrates. To advance this approach and to expand its capabilities I will present our results of multimodal chemical imaging using this technique on test substrates and show application of this approach for the multimodal analysis of perovskite (HOIPs) materials. I will discuss the performance metrics of the multimodal imaging system on conductive and non-conductive materials and discuss our results on understanding the chemical nature of ferroelastics twin domains in methylammonium lead triiodide (MAPbI3) perovskite using HIM-SIMS.

3:20pm **HI+AS+CA-WeA4 Compositional Characterization of Biogenic Nanoparticles using the ORION NanoFab with SIMS**, ***Christelle Guillermier***, *F. Khanom,* Carl Zeiss PCS, Inc.*; D. Medina,* Northeastern University*; J.-N. Audinot,* Luxembourg Institute of Science and Technology, Luxembourg

Over the past several years, the use of both nanoparticles and nanostructured surfaces have emerged as an alternative's solution to antibiotic resistant bacteria as they effectively decrease bacterial survival without being highly toxic to mammalian cells. These nanoparticles whose sizes span 10 nm to several hundred nm are composed of a variety of materials such as pure metals, metal oxides, and metalloids. Their chemical characterization however remains a challenge due to their small sizes. Scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX) are the conventional analytical techniques of choice to determine the nanoparticles' morphology, size, and elemental composition. However, although sensitive enough to detect trace elements, SEM and EDX cannot provide elemental information for the smallest features on a bulk sample, or for the lightest elements.

The ORION NanoFab is an ion microscope that allows for high resolution secondary electron (SE) imaging with a He^+ focused ion beam that can be focused to a 0.5 nm probe size. The same instrument offers a Ne^+ ion beam with a focused probe size of 2 nm. Recently, this same platform has been configured with a custom-designed magnetic sector secondary ion mass spectrometer (SIMS). It allows for the detection of all periodic table elements including H and Li which EDS cannot easily detect. Importantly, SIMS with neon provides elemental imaging with spatial resolution smaller than 20 nm. The combination of high resolution He^+ imaging (0.5 nm) with Ne^+ SIMS elemental mapping yields a direct correlative technique particularly attractive for exploring nanoparticles and nanofeatures in general.

NanoFab-SIMS has already yielded information-rich images in diverse fields of applications. We will here illustrate its potential for the characterization of biogenic nanoparticles made by bacteria and plants.

4:20pm **HI+AS+CA-WeA7 Effects of Ion Irradiation on Two-Dimensional Targets: What is Different from Bulk Materials**, ***Arkady V. Krasheninnikov,*** Helmholtz-Zentrum Dresden-Rossendorf, Germany **INVITED**

Ion irradiation has successfully been used for introducing impurities and creating defects in two-dimensional (2D) materials in a controllable manner. Moreover, focused ion beams, especially when combined with in-situ or post-irradiation chemical treatments, can be employed for patterning and even cutting 2D systems with a high spatial resolution. The optimization of this process requires the complete microscopic understanding of the interaction of energetic ions with the low-dimensional targets.

In my presentation, I will dwell upon the multi-scale atomistic computer simulations of the impacts of ions onto free-standing (e.g., suspended on a TEM grid) and supported (deposited on various substrates) 2D materials, including graphene and transition metal dichalcogenides (TMDs), such as MoS2 and WS2. I will emphasize the differences between defect production under ion irradiation in 2D materials and bulk solids. The theoretical results will be augmented by the experimental data obtained by the coworkers. I will further present the results of multi-scale simulations of ion irradiation of free-standing [1] and supported [2] graphene and 2D TMDs, and demonstrate that depending on ion mass and energy, the defect production can be dominated by direct ion impacts, back scattered ions or atoms sputtered from the substrate [2]. Finally, I will touch upon the interaction of highly-charged [3] and swift heavy ions [4] with 2D systems and overview recent progress in modelling this using non-adiabatic approaches including time-dependent density functional theory and Ehrenfest dynamics [5].

1. M. Ghorbani-Asl, S. Kretschmer, D.E. Spearot, and A. V. Krasheninnikov, 2D Materials 4 (2017) 025078.

2. S. Kretschmer, M. Maslov, S. Ghaderzadeh, M. Ghorbani-Asl, G. Hlawacek, and A. V. Krasheninnikov, ACS Applied Materials & Interfaces 10 (2018) 30827.

3. R. A. Wilhelm, E. Gruber, J. Schwestka, R. Kozubek, T.I. Madeira, J.P. Marques, J. Kobus, A. V. Krasheninnikov, M. Schleberger, and F. Aumayr, Phys. Rev. Lett. 119 (2017) 103401.

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5. A. Ojanperä, A. V. Krasheninnikov, and M. Puska, Phys. Rev. B 89 (2014) 035120.

5:00pm **HI+AS+CA-WeA9 Effects of He Ion Irradiation on Gold Nanoclusters: a Molecular Dynamics Study**, ***Sadegh Ghaderzadeh****, M. Ghorbani-Asl, S. Kretschmer, G. Hlawacek,* Helmholtz-Zentrum Dresden Rossendorf, Germany*; A.V. Krasheninnikov,* Helmholtz-Zentrum Dresden-Rossendorf, Germany

The interpretation of helium ion microscopy (HIM) images of crystalline metal clusters requires microscopic understanding of the effects of He ion irradiation on the system, including energy deposition and associated heating, as well as channeling patterns. While channeling in bulk metals has been studied at length, there is no quantitative data for small clusters. We carry out molecular dynamics simulations to investigate the behavior of gold nano-particles with diameters of 5-15 nmunder 30 keV He ion irradiation. We show that impacts of the ions can give rise to substantial heating of the clusters through deposition of energy into electronic degrees of freedom, but it does not affect channeling, as clusters cool down between consecutive impact of the ions under typical imaging conditions. At the same time, high temperatures and small cluster sizes should give rise to fast annealing of defects so that the system remains crystalline. Our results show that ion-channeling occurs not only in the principal low-index, but also in the intermediate directions. The strengths of different channels are specified, and their correlations with sputtering-yield and damage production is discussed, along with size-dependence of these properties. The effects of planar defects, such as stacking faults on channeling were also investigated.

Finally, we discuss the implications of our results for the analysis of HIM images of metal clusters.

5:20pm **HI+AS+CA-WeA10 Low Damage Imaging of Polymers with the Helium Ion Microscope**, ***Doug Wei,*** Carl Zeiss, RMS, Inc.*; J.A. Notte,* Carl Zeiss PCS, Inc.*; A. Stratulat,* Carl Zeiss Microscopy, Ltd., UK

Polymers present a combination of challenges for high magnification imaging with the conventional SEM or FIB. Because they are electrically insulating, polymers are susceptible to charge accumulation and can produce imaging artifacts. Or worse, the implanted charge and surface charge can generate fields large enough to induce catastrophic dielectric breakdown. The interaction of the primary beam with the chemical bonds can cause radiolysis, cross-linking, and chain scissions which alter their morphology and other properties. Ion beams of relatively heavy species (Ga and Xe) can cause appreciable sputtering especially at high magnifications. In some cases, the sputtering can be preferential for light atoms, causing disproportionate hydrogen loss. Further difficulties include heating effects, since the typical polymers are good thermal insulators. Compounding matters, they are often temperature sensitive and can be damaged at even modest temperature rises ~ 50 deg C.

However, some of the newly available light ion beams (H, He, Li) offer unique advantages that help to circumvent the problems traditionally encountered when imaging polymers. First, the charging effects are greatly diminished compared to the SEM. In part, this is because the incident ion is likely neutralized as it enters the sample, and remains in a mostly neutral state as it penetrates deeply. This leaves only a net surface charge, which is overall positive and made more so by the ejection of secondary electrons from the surface. This is easily resolved using a collection of low energy electrons provided by a flood gun. The light ion beams also have relatively low sputtering rates compared to the heavier ions. Their interactions are primarily with the electrons of the sample. So while they can affect bonding, they are much less likely to cause sputtering. The light ions will generally be implanted deeply, often hundreds of nanometers under the surface, and helium in particular is known to diffuse out over time. Thermal effects are also much reduced with the light ion beams compared to heavier ions or the SEM. The ion's initial kinetic energy is converted to random thermal energy over a relatively large volume. And much of the transferred energy goes to the electrons in the sample, and their relatively long mean free path helps to dissipate this energy into a larger volume.

Numerous imaging examples will be provided from a variety of polymers using the helium beam from the Zeiss ORION NanoFab. These serve as representative examples of the unique sample interaction of light ions and the advantages they offer for imaging polymers.

5:40pm **HI+AS+CA-WeA11 Imaging of Biological Cells with Helium-Ion Microscopy**, ***Natalie Frese****, A. Beyer, C. Kaltschmidt, B. Kaltschmidt,* Bielefeld University, Germany*; A. Thomas,* Institute for Metallic Materials Dresden, Germany*; W. Parak,* University of Hamburg, Germany*; A. Gölzhäuser,* Bielefeld University, Germany

Studies from the last decade have shown that helium-ion microscopy (HIM) is suitable for studying biological samples. In particular, cell membranes can be imaged by HIM without metallic coatings, which could lead to disturbance of the surface. In this contribution, we give two examples of biological cells imaged by HIM: (i) mouse hippocampal neurons on patterned surfaces for neuronal networks and (ii) human cells treated with colloidal nanoparticles [1, 2]. Both examples benefit from the high resolution imaging of uncoated, biological materials by HIM, as for (i) the cell adherence to patterned surfaces could be imaged and for (ii) cell morphology images indicated harmful effects of colloidal nanoparticles to cells.

[1] M. Schürmann et al., PLoS ONE 13(2), e0192647 (2018)

[2] X. Ma et al., ACS Nano 11(8), 7807-7820 (2017)

6:00pm **HI+AS+CA-WeA12 Channeling in the Helium Ion Microscope**, ***Hussein Hijazi****, C. Feldman, R. Thorpe, M. Li, T. Gustafsson,* Rutgers University*; D. Barbacci, A. Schultz,* Ionwerks

The helium ion microscope (HIM) has become a unique tool for modern materials science due to its high lateral resolution for imaging, high spatial resolution and nano-scale analysis. For crystalline materials, the incident beam may undergo ion channeling, which strongly modifies all of the basic ion-solid interactions associated with these HIM functions. Here, a 30 keV He^+ beam was used for RBS channeling in a W(111) crystal using a novel time of flight (HIM/TOF) detector developed at Rutgers University to extract critical channeling parameters. Measurements of the minimum backscattering yield (χ_{min}), surface peak (SP), and critical angle, are compared to several theoretical estimates. The results illustrate the advantage of using channeling in a backscattering mode to characterize crystalline materials with the HIM, as the backscattering intensity modifications are far greater for scattered ions than for secondary electrons. This case of "ideal" channeling with the HIM now provides a basis for analysis of more complex materials such as polycrystalline materials and textured structures, and quantifies the role of HIM induced materials modification in crystalline materials.

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

Jacob L. Swett[a], Ondrej Dyck[b], Stephen Jesse[b], Jan A. Mol[a,c]

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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)

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[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.

We acknowledge financial support of the JSPS-KAKENHI (Nos. 21101007, 26220712, 23244079, 25620001, 23740228, 26706007, 26610077, and 16K04918); JST-CREST (JPMJCR1674); and Q-LEAP. A part of this work was supported by "Nanotechnology Platform Project (Nanotechnology Open Facilities in Osaka University)" of Ministry of Education, Culture, Sports, Science and Technology, Japan [F-18-OS-0029].

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.

Thursday Afternoon, October 24, 2019

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

Emerging Ion Sources, Optics, and Applications

Moderators: Gregor Hlawacek, Helmholtz-Zentrum Dresden Rossendorf, Germany, Shida Tan, Intel Corporation

2:20pm **HI+NS-ThA1 Cold Atom Ion Sources***, **Jabez McClelland**, J.R. Gardner, W.R. McGehee,* National Institute of Standards and Technology (NIST)*; A. Schwarzkopf, B.J. Knuffman, A.V. Steele,* zeroK NanoTech Corp. **INVITED**

Ionization of laser-cooled atoms has emerged as a new approach to creating high brightness ion sources for applications such as focused ion beam (FIB) microscopy, milling, and secondary-ion mass spectrometry (SIMS). Conventional sources, such as the Ga liquid metal ion source (LMIS) or the gas field ionization source (GFIS), attain brightness by emitting from a very sharp tip. In contrast, cold atom sources attain high brightness through reducing the transverse velocity spread of the ions. With the ultracold, microkelvin-range temperatures achievable with laser cooling, the corresponding velocity spread can lead to a brightness significantly higher than typical LMIS values. Moreover, the phase-space shape of the emittance of the source – narrow in velocity, wide in space – brings new opportunities for ion optical design. For example, high currents can be obtained without the high current density present in sharp tip sources. This can result in reduced Coulomb effects, such as increased emittance and broadened energy spread (Boersch effect). Other advantages of this type of source include insensitivity to contamination, access to new ionic species, inherent isotopic purity, and fine control over emission, down to the single ion level. To date, sources have been demonstrated with Cr,[1] Li,[2] Rb,[3] and Cs[4,5] ions, realizing novel species and nanometer-scale spot sizes. In this talk I will review progress in the field and discuss recent developments in Li ion sources and applications.

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3:00pm **HI+NS-ThA3 Silicon Lithiation by Direct-writing with a Focused Li^+-ion Beam***, W.R. McGehee, **Evgheni Strelcov**, V. Oleshko, C. Soles, N.B. Zhitenev, J.J. McClelland,* National Institute of Standards and Technology (NIST)

Improving the performance of Li-ion batteries requires understanding and controlling nanoscale ion transport at the level of interfaces, grain boundaries and defects. While in the last decades a range of electron and scanning probe microscopy techniques have been developed for probing local transport, no reliable method exists for quantitative and controllable nanoscale lithiation. Moreover, wet-cell electrochemical lithiation is significantly complicated by electrolyte decomposition, formation of solid-electrolyte interfacial (SEI) layer and parasitic reactions running in parallel to lithium insertion.

Building on our previous work,[1] here we introduce a new method of direct-write quantitative lithiation of battery-relevant materials in vacuo, in the absence of SEI or liquid electrolyte. To benchmark the technique, we use a focused, several keV Li^+-ion beam to inject lithium into 35-nm thick crystalline Si membranes with a sub-micron lateral precision. The lithiated regions, undergoing morphological, structural, chemical and functional transformations, were characterized with a combination of electron and scanning probe microscopy techniques. We observed saturation of interstitial lithium in the silicon membrane at ≈ 10 % dopant number density and spill-over of excess lithium onto the membrane's surface. The implanted Li^+ remains electrochemically active, and the spill-over effect can possibly be avoided by cooling the sample. The presented method is especially useful for probing non-equilibrium and low-concentration phases of lithiated materials that form because of incomplete lithium extraction or during initial states of pristine anode lithiation. Focused ion beam lithiation will enable controlled studies and improved understanding of Li^+ ion interaction with local defect structures and interfaces in electrode and solid-electrolyte materials.

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W.R.M. and E.S. contributed equally.

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3:20pm **HI+NS-ThA4 A New FIB for Deterministic Single Ion Implantation***, **Nathan Cassidy**,* UK National Ion Beam Centre, University of Surrey, UK*; D. Cox,* Advanced Technology Institute, University of Surrey, UK*; R. Webb,* UK National Ion Beam Centre, University of Surrey, UK*; B. Murdin,* Advanced Technology Institute, University of Surrey, UK*; B. Blenkinsopp, I. Brown,* Ionoptika Ltd., UK*; R. Curry,* The Photon Science Institute, University of Manchester, UK

Single isolated dopant atoms implanted into solid state devices have been shown to be a viable architecture for quantum technologies. Ion implantation provides many advantages as a manufacturing method for such devices, such as speed and scalability, however controlling the number of implanted ions with single-ion precision poses a significant challenge. In this paper we will present a new instrument designed for the deterministic implantation of single ions with high precision.

The SIMPLE (Single Ion Multi-species Positioning at Low Energy) tool, is a new focused ion beam tool in operation designed for the manufacture of quantum technologies. The tool has a 25kV LMIG set up for femtoAmp sample currents, with ultra-fast beam blanking, neutral blocking and a highly efficient secondary electron detection system. Deterministic ion implantation is achieved through extraction of single ions through fast beam blanking with low currents, ion implant detection through collection of secondary electron (SE) signal from the target and high spatial precision in ion placement.

To date we have demonstrated > 85% probability of implanting a single Bi+ ion into silicon without error, with a 20nm beam determining dopant placement precision. This surface secondary electron detection efficiency has been validated through simultaneous measurements of a transmitted electron signal, achieved by implanting through thin lamellae. The ion placement precision has been determined through imaging of ion induced damage on highly oriented pyrolytic graphite (HOPG) surfaces. Much work has taken place maximizing the detection efficiency for secondary electrons and investigating the factors which affect the SE yield.

Currently the system is running with Bi source, and there are In sources available. Alongside the development of the instrument there is also research into developing a series of liquid-metal ion sources for elements with optical and quantum applications including P, Te, Se and Cd. A second SIMPLE tool has also been installed at the UK National Ion Beam Centre, which operates with a 20kV duoplasmatron arc source, capable of 50nm spot sizes. SIMPLE #2 will initially operate with nitrogen source for the fabrication of NV centres in diamond.

4:00pm **HI+NS-ThA6 Technology and Applications of a Plasma Ion Source with User-selectable Ion Species***, **Gregory Schwind**, S.M. Kellogg, J. Stiller, M. Doud, C. Rue, B. Van Leer,* Thermo Fisher Scientific **INVITED**

The focused ion beam (FIB) has become an indispensable tool for micro- and nano-machining applications. Due to its high brightness and ease of use, the gallium liquid metal ion source (LMIS) has been the source of choice over much of the nearly four decades of FIB history. At the beginning of this decade, a new generation FIB system based on the inductively coupled plasma (ICP) ion source was brought to market, offering beam current and throughput 20 times greater than LMIS-based systems. A next generation plasma source has been developed [1], offering the option to change the ion beam species by switching the feed gas supplied to the plasma source. The ability to dynamically change ion species—for example from a noble gas such as argon to an electronegative species such as oxygen—creates new design challenges for the source, the FIB optical subsystem, and the platform as a whole. Both empirical measurements and numerical simulations were used to better understand the species-specific performance of the source design. Results show that the emission properties depend on both the ion species and the plasma density, which lead to orienting the system design around specific modes of operation optimally suited to each species, FIB current and landing energy [2].

Several new and exciting application areas are enabled by the ability to switch FIB ion species dynamically. Ion-surface interactions such as sputtering, implantation, and the creation of an amorphous damage layer depend on the ion's momentum [3], which in turn depends on ion mass. Furthermore, chemical reactivity between the incoming ion and the target surface seems to play a role in the surface modification process in some instances. Several FIB application examples illustrating these interdependencies will be shown.

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4:40pm **HI+NS-ThA8 Neutral Helium Microscopy*, Bodil Holst*,** University of Bergen, Norway

Neutral helium microscopy is a new imaging technique currently under development. In a neutral helium microscope a beam of neutral helium atoms is created through supersonic expansion from a nozzle and focussed onto the surface to create a scanning instrument. The resolution is determined by the beam spotsize on the surface. The neutral helium microscope has several advantages: the very low energy of the beam (less than 0.02 eV compared to several keV for helium ion or electron microscope) , charge neutrality, and inertness of the helium atoms, a potential large depth of field, and the fact that at thermal energies the helium atoms do not penetrate into any solid material. This opens the possibility, among others, for the creation of an instrument that can measure surface topology on the nanoscale, even on surfaces with high aspect ratios. The helium microscope currently exist in two configurations: The pinhole microscope and the zone plate microscope, both are covered in this paper. We begin with a series of images which demonstrate and explores the unique contrast mechanisms of the new instrument. This is followed by a general discussion of helium microscope designs and resolution.

5:00pm **HI+NS-ThA9 GaBiLi Liquid Metal Alloy Ion Sources for Advanced Nanofabrication***, P. Mazarov,* RAITH GmbH, Germany*; T. Richter, L. Bruchhaus, W. Pilz, R. Jede,* Raith GmbH, Germany*; **Yang Yu**, R.M. Schmid, J.E. Sanabia,* Raith America, Inc.*; L. Bischoff,* Helmholtz Zentrum Dresden-Rossendorf, Germany*; G. Hlawacek,* Helmholtz-Zentrum Dresden Rossendorf, Germany

Nanofabrication requirements for FIB technologies are specifically demanding in terms of patterning resolution, stability and the support of new processing techniques. Additionally, 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 LMAIS provides single and multiple charged mono- as well as polyatomic ion species of different masses, resulting in significantly different interaction mechanisms. Nearly half of the elements of the periodic table are thus 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 an example we will introduce the 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.

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5:20pm **HI+NS-ThA10 Focused Ion Beams in Biology: How the Helium Ion Microscope and FIB/SEMs Help Reveal Nature's Tiniest Structures*, Annalena Wolff*,** Central Analytical Research Facility, Institute for Future Environments, Queensland University of Technology (QUT), Brisbane QLD 4000, Australia*; N. Klingner,* Helmholtz Zentrum Dresden-Rossendorf, Germany*; W. Thompson,* HeelionicsLLC*; Y. Zhou,* Queensland University of Technology (QUT), Australia*; J. Lin,* Affiliated Stomatological Hospital of Xiamen Medical College, China*; Y. Peng,* CSIRO Manufacturing, Australia*; J. Ramshaw,* St. Vincent's Hospital, University of Melbourne, Australia*; Y. Xiao,* The Australia-China Centre for Tissue Engineering and Regenerative Medicine (ACCTERM), Queensland University of Technology, Australia

Focused Ion Beam (FIB) devices such as the Helium Ion Microscope (HIM) as well as FIB/SEMs are increasingly popular within the biological sciences in recent years. High resolution imaging of uncoated non-conductive samples with the HIM helps reveal nature's tiniest structures while the FIB/SEM allows to prepare TEM lamellae, 3D reconstruct the sample or reveal sub surface structures with nanometre precision.

This presentation shows how the HIM as well as FIB/SEMs can be used in biological sciences to reveal nature's tiniest structures. The presented work then focuses on the underlying ion-solid interactions and the effect of ion beam parameters on heating induced by ion beams. The work presented here deals with gallium ion solid interactions, however the broader results are applicable to any type of FIB including the helium ion microscope (HIM) and plasma FIBs. The interactions of gallium ions in skin were simulated using Monte Carlo methods, finite element simulations and numerical modelling for different beam parameters. The program SRIM [4] was used to obtain theoretical results which permit estimation of the ion beam induced temperature increases, using the physical principles of Fourier's law of conductive heat transfer.

The technique was tested on collagen, a soft biological material which is commonly used in biomedical applications. Collagen was chosen as a suitable test sample as it loses its fibrillary structure when denaturated by heat, permitting damage to easily be recognized. Cross-sections and TEM lamellas were prepared from non-embedded collagen with conventional FIB processing parameters as well as heat reducing FIB parameters.

The results also show that heat damage can be prevented by reducing the local dose rate and area underneath the ion beam. Using lower acceleration voltages allows the operator to select higher local dose rates (ion beam currents) and minimized processing times. A TEM comparison of a microtome prepared lamella and a FIB prepared lamella (using different heat reducing parameters) shows that the fibrillar structures can be maintained, and heat damage avoided. The approach described here can be used to determine suitable parameters for other soft materials.

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Advanced Ion Microscopy and Ion Beam Nano-engineering Focus Topic
Room Union Station B - Session HI-ThP

Advanced Ion Microscopy Poster Session

HI-ThP2 Morphology Modification of Si Nanopillars under Ion Irradiation at Elevated Temperatures*, **Xiaomo Xu**, K.-H. Heinig,* Helmholtz Zentrum Dresden-Rossendorf, Germany*; W. Möller,* Helmholtz-Zentrum Dresden-Rossendorf, Germany*; H.-J. Engelmann, N. Klingner,* Helmholtz Zentrum Dresden-Rossendorf, Germany*; A. Gharbi, R. Tiron,* CEA-LETI, France*; J. von Borany,* Helmholtz Zentrum Dresden-Rossendorf, Germany*; G. Hlawacek,* Helmholtz-Zentrum Dresden Rossendorf, Germany

Ion beam irradiation of vertical nanopillar structures can be utilized to fabricate a vertical gate-all-around (GAA) single electron transistor (SET) device in a CMOS-compatible way. After irradiation of Si nanopillars (with a diameter of 35 nm and a height of 70 nm) by either 50 keV broad beam Si^+ or 25 keV focused Ne^+ beam from a helium ion microscope (HIM) at room temperature and a fluence of 2e16 ions/cm^2, strong deformation of the nanopillars has been observed which hinders further device integration. This is attributed to ion beam induced amorphization of Si allowing plastic flow due to the ion hammering effect, which, in connection with surface capillary forces, dictates the final shape. However, plastic deformation can be suppressed under irradiation at elevated temperatures (investigated up to 672 K). Then, as confirmed by bright-field transmission electron microscopy, the substrate and the nanopillars remain crystalline, and are continuously thinned radially with increasing fluence down to a diameter of 10 nm. This is attributed to enhanced forward sputtering through the sidewalls of the pillar, and found in reasonable quantitative agreement with the predictions from 3D ballistic computer simulation using the TRI3DYN program.

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