Program EUFN Workshop 2022

Wednesday

		09:30	**Get Together & Registration**
Tutorial		10:00	FIB preparation of lamellae on chips for in-situ TEM and other techniques – J. Reuteler
		12:00	**LUNCH**
Session n°1	Talk 1	13:00	Tungsten based SQUID Nanofabrication by means of Focused Ion Beam Induced Deposition – F. Sigloch
	Talk 2	13:25	Comparing Contrast in nano-CT and FIB/SEM Tomography of an Al Cast Alloy – A Correlative Microscopy Study – C. Pauly
	Exi 1	13:50	Explore unlimited process pathways for FIB nanopatterning and ion imaging using VELION – T. Richter (Raith)
	Talk 3	14:15	3D Nanoprinting of Electrical AFM Nanoprobes – L. M. Seewald
		14:45	**COFFEE BREAK**
Session n°2	Inv 1	15:30	Manipulation and Study of Antiferromagnetic Order Enabled by Focused Ion Beam Fabrication – S. Haley
	Talk 4	16:05	Backside FIB/SEM analysis strategy to identify a new failure mode at an automotive magnetic sensor device – M. Simon-Najasek
	Exi 2	16:30	in situ Force Measurements – the FIB/SEM as a Mechanical Characterization Tool – A.J. Smith (Kleindiek)
	Talk 5	16:55	Development and SEM integration of the Nano Aperture Ion Source – M.L. Simons
	Exi 3	17:20	Extending microstructure characterization from nm to mm scale using the latest multiple ion species plasma FIB integrated with femtosecond laser. – M. Wu (Thermo-Fisher)
		17:45	**Food Truck + beers + posters**

Thursday

		08:30	**Get Together**
Session n°3	Talk 6	09:00	The patterning toolbox FIB-o-mat: Exploiting the full potential of focused ion beams for nanofabrication – K. Höflich
	Inv 2	09:25	Focused Ion Beam Induced Strain Generation in Silicon Membranes – D. Cox
	Exi 4	10:00	Two Microscopes are better than One – In-situ Correlative Analysis by combination of AFM, SEM, and FIB – H. Frerichs (Quantum Design)
		10:25	**COFFEE BREAK**
Session n°4	Talk 7	11:00	Direct-Write 3D Nanoprinting of High-Resolution Magnetic Force Microscopy Nanoprobes – M. Brugger-Hatzl

	Inv 3	11:25	Using the Helium Ion Microscope for Imaging and Modification of Nanostructures, 2D Materials, and SARS-CoV-2 infected Cells – Armin Gölzhäuser
	Exi 5	12:00	Next chapter in Nanoprototyping in the new generation of FIBSEM systems – Miloš Hrabovský (Tescan)
		12:30	**LUNCH + Discussions**
Session n°5	Talk 8	14:00	Sample preparation and analysis of LLZO ceramics for solid state batteries with Cryo FIB/SEM and aberration corrected analytical STEM – C. J. Burkhardt
	Talk 9	14:25	Helium ion microscopy and sectioning of Spider Silk – J. Fiutowski
	Inv 4	14:50	Best Practices for Xe PFIB Preparation of Materials for Transmission Electron Microscopy – S. Vitale
	Exi 6	15:25	New Applications in Energy Research Enabled by a Triple Beam, Dual Chamber FIB with Isotropic Tomographic Voxels – B. Tordoff (Zeiss)
	Talk 10	15:50	Application of FIB-TOF-SIMS for 3D high-resolution chemical characterization of Li-ion solid-state batteries – A. Priebe
		16:15	**POSTER SESSION** **COFFEE / BEER**
		18:30	**Meet at Rickmer Rickmers for Conference Dinner**

Friday

		08:30	**Get Together**
Session n°6	Talk 11	09:00	On demand spatially controlled fabrication of single photon emitters in Si – G. Hlawacek
	Inv 5	09:25	Magnetic patterning using Ne, Co, and Dy FIB – K. Lenz
	Exi 7	10:00	TOFWERK – fibTOF: The strength of SIMS capabilities on FIB-SEM microscopes – L. Pillatsch
		10:25	**COFFEE BREAK**
Session n°7	Talk 12	11:00	Positioned generation of luminescence defects in 2D materials by helium ion beams – A. Hötger
	Talk 13	11:25	Fabrication of microstructured devices for grain boundary investigations in unconventional superconductor $CeCoIn_5$ – S. Mishra
	Talk 14	11:50	Exploring Layered Conductors by 3D FIB micro-machining – C. Putzke
	Talk 15	12:15	Investigation of the Interaction of a Ga+ Focused Ion Beam with Zirconia by Electron Backscatter Diffraction – N. Brachhold
		12:40	**CLOSING**

Invited Speakers

David Cox | University of Surrey | United Kingdom
„Focused Ion Beam Induced Strain Generation in Silicon Membranes"

Armin Gölzhäuser | University of Bielefeld | Germany

Shannon Haley | University of California, Berkeley | United States
„Manipulation and Study of Antiferromagnetic Order Enabled by Focused Ion Beam Fabrication"

Kilian Lenz | Helmholtz Zetnrum Dresden Rossendorf | Germany
„Magnetic patterning using Ne, Co, and Dy FIB"

Suzy Vitale | Carnegie Institution for Science | United States
„Best Practices for Xe PFIB Preparation of Materials for Transmission Electron Microscopy"

Invited Tutorial

Joakim Reuteler | ETH Zürich | Switzerland
„Tutorial: FIB preparation of lamellae on chips for in-situ TEM and other techniques"

Posters: (presenting author, title)

Braun et al. – Focused Ion Beam Induced Nanoscale Phase Transitions in Layered Structures

Elyas et al. – The Manufacture of van der Waals Heterostructures Using He Ion Beam Patterning

Gotszalk et al. – High throughput tips manufacturing for active piezocantilevers with xenon ion beam with mass control

Guo et al. – Crystalline anisotropic curtaining effect in Bismuth

Hunter et al. – Micromachined samples for uniaxial strain studies with laser-ARPES

Kreps et al. – In-situ sample preparation of oxidizing and contaminating samples for high quality EDS and WDS quantification using FIB-SEM

Leissner et al. – Dual focused ion beam nanofabrication of V-grooves in monocrystalline gold for efficient excitation of organic single photon emitters

Nadzeyka et al. – Novel FIB nanofabrication strategies facilitated by light and heavy ions from GaBiLi Liquid Metal Alloy Ion Sources

Neupane et al. – Effect of Focused Helium-ion Beam on Surface Morphology of Polypropylene Thin-films for Power Capacitors

Smith et al. – Fault Localization in FIB/SEM – combining delayering and EBAC into a cohesive workflow

Vinograd et al. – Second order Zeeman interaction and ferroquadrupolar order in TmVO4

Weitzer et al. – Using Electron Beam Curing (EBC) for the Controlled Bending of 3D-Nanoprinted FEBID Structures

Weitzer et al. – High-Precision 3D-Nanoprinting for Sheet-like Structures via FEBID

Winiarski – Correlative Microscopy for Aviation and Aerospace

Winiarski et al. – Plasma FIB spin milling hastens battery research

Winiarski et al. – Plasma FIB-SEM-based Kintsugi Imaging for Battery Electrodes

Winkler et al. – The FIB as 3D Nanoprinter – Overview of the Activities in Graz

Oral Contributions

Tutorial: FIB preparation of lamellae on chips for in-situ TEM and other techniques

J. Reuteler[1*]

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This tutorial targets a wide range of FIB operators, from beginners to long time expert users across fields, interested in preparation of lamellae on chips (LoC). Starting with a recap of the in-situ lift-out technique for preparing a TEM lamella mounted to a half-grid (Omnigrid), the terminology used throughout the tutorial is defined. Important concepts for achieving high quality TEM lamellae for different purposes are summarized.

There is an increasing demand for thin samples mounted to a specific chip, typically MEMS. Thus the core part of this tutorial discusses several protocols for preparation of lamellae on chips. Examples will cover the preparation of samples for in-situ TEM observations, synchrotron-based x-ray techniques under local magnetic field as well as for heating experiments, and a lamella precisely mounted on an optical waveguide. Furthermore, different routes for dealing with unconventional geometries are discussed.

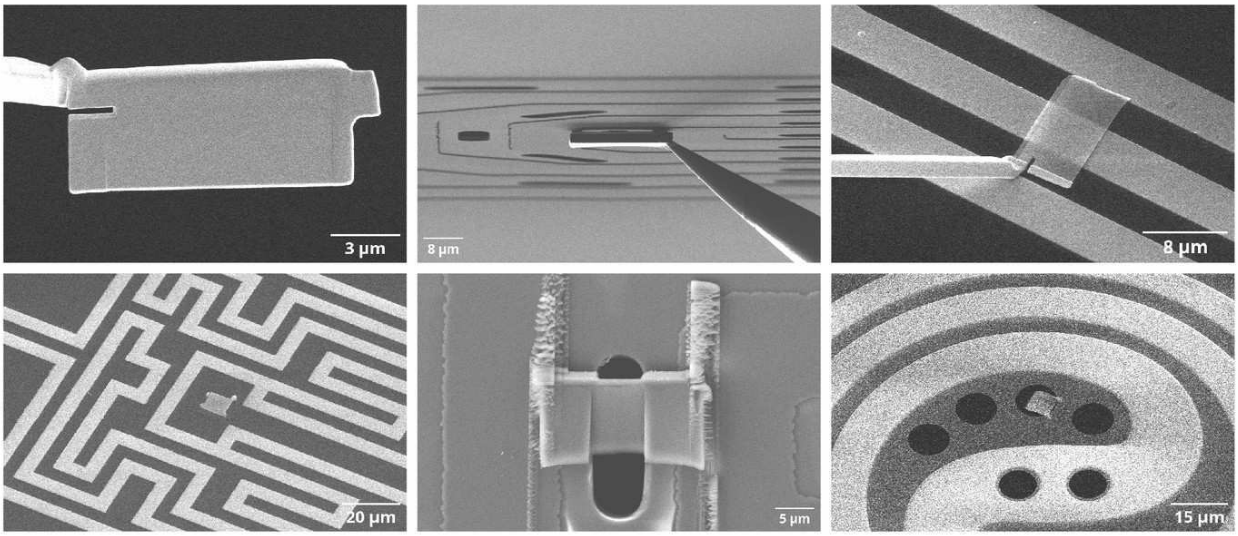

Fig. 1: Snapshots taken while preparing various lamellae on chip.

Tungsten based SQUID Nanofabrication by means of Focused Ion Beam Induced Deposition

F. Sigloch[1], S. Soraya[1,2], P. Orús[1,2] and J. M. De. Teresa[1,2]

[1] Instituto de Nanociencia y Materiales de Aragon (INMA), Universidad de Zaragoza-CSIC, 50009 Zaragoza, Spain
[2] Laboratorio de Microscopía Avanzadas (LMA), Universidad de Zaragoza, 50018 Zaragoza, Spain
[*] corresponding author email: fsigloch@unizar.es

Direct write techniques such as Focused Electron/Ion Beam Induced Deposition (FEBID/FIBID) constitute versatile, resist-free techniques for the fabrication of nanostructures, offering an alternative to conventional methods such as Optical or Electron Beam Lithography.
The deposition of the comercially available precursor gas $W(CO)_6$ with Ga^+ ions results in a film of W-C with well-established superconducting properties [1]. Planar deposits exhibit a critical temperature of T_c = 4 K – 5 K, an upper critical magnetic field of $B_{c,2}$ = 7 T – 8.5 T and a critical current density of J_c = 0.001 MA/cm^2 – 0.01 MA/cm^2 [2]. The London penetration depth is reported to be λ_L = 850 nm and the superconducting coherence length as ξ = 6 nm – 9 nm [2]. With Ga^+ FIBID and the $W(CO)_6$ precursor, superconducting nanostructures with a linewidth of 50 nm are feasible to fabricate with high precision and reproducibility.

In this work we present the fabrication of nanoscale Superconducting Quantum Interference Devices (nanoSQUIDs) of W-C by means of Ga^+ FIBID. The SQUID loop is formed by a 50 nm thick and 200 nm wide film with a rectangular inner loop of 300 x 700 nm^2. The Josephson Junctions (JJs) are formed by 300 nm long constrictions with a cross-sectional area of down to 50 x 50 nm^2. The SQUIDs obtained show a critical temperature of up to T_c = 4.3 K and a critical current of up to I_c = 8.5 μA. The normal state resistance of the JJs is R_N = 496 Ω. Upon variation of the external magnetic field we observe periodic oscillations in the critical current, I_c, and upon injection of a constant bias current $I_b \sim I_c$ the voltage dropping across the structure displays a sinusoidal dependence on the external magnetic field. The transfer coefficient is remarkably high, with up to V_Φ = 1301 μA/ϕ_0 due to the high normal-state resistance of the JJs [3]. Recent efforts towards improving the nanoSQUIDs properties will be introduced.

[1] P. Orús, F. Sigloch, S. Sangiao and J. M. De Teresa. *Superconducting Materials and Devices Grown by Focused Ion and Electron Beam Induced Deposition*, Nanomaterials 12, (2022) 1367
[2] P. Orús, R. Córdoba, J. M. De Teresa. *Nanofabrication – Nanolithography techniques and their application,* IOP Publishing (2020), ch. 5.
[3] F. Sigloch, S. Sangiao, P. Orús, J. M. De Teresa. *Large output voltage to magnetic flux change in nanoSQUIDs based on direct-write Focused Ion Beam Induced Deposition technique*, arXiv [Prepint] (2022), [arXiv:2203.05278].

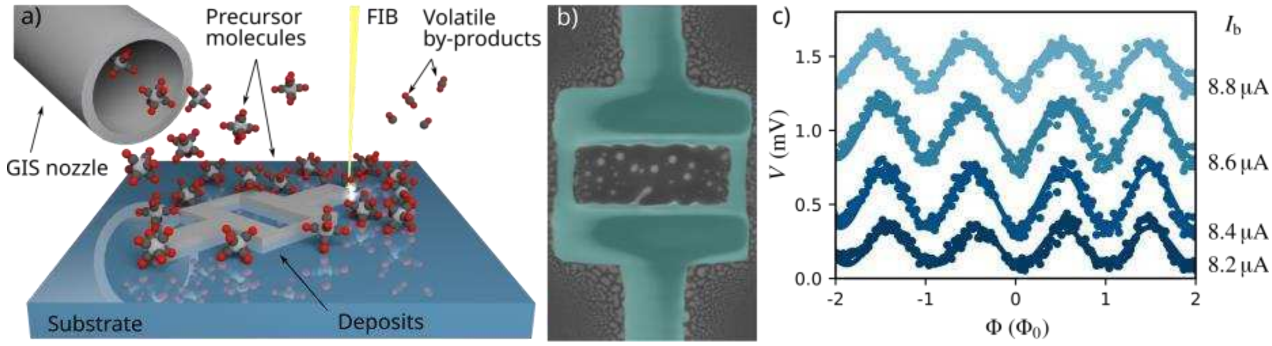

Fig. 1: a) Schematic representation of the FIB induced deposition of a nanostructure. b) A SEM image of a W-C (blue) nanoSQUID fabricated by Ga^+ FIBID. c) The corresponding periodic modulation of the voltage, V, in dependence of the magnetic flux threading the SQUID loop, ϕ. The different lines correspond to different bias currents $I_b \sim I_c$. Reproduced from [3].

Comparing Contrast in nano-CT and FIB/SEM Tomography of an Al Cast Alloy – A Correlative Microscopy Study

C. Pauly*[1], M. Engstler[1], J. Fell[2], H.-G. Herrmann[2], F. Mücklich[1]

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Aluminum alloys with silicon as the main alloying element are widely used materials for general machine and automotive parts. The microstructure of hypereutectic Al-Si-alloys (>12 wt.% Si) comprises primary Si particles and a complex Al-Si eutectic. Elements such as Cu, Mg an Ni are added to form hard intermetallic precipitates which improve the mechanical properties at elevated temperatures. Size, shape and connectivity of these complex-shape intermetallic particles play a decisive role for the properties and are thus of special interest. As classic 2D methods cannot capture connectivity, a 3D analysis of the microstructure is necessary. Due to the particle size in the µm range, high-resolution 3D imaging techniques are required.

The x-ray computed tomography (XCT) has made strong progress in the past years enabling a resolution of ~ 50 nm in lab-scale instruments. In the field of focused ion beam (FIB) serial sectioning, the introduction of the xenon plasma source has significantly increased the upper volume to an edge length of ~100-200 µm. Thus, both techniques overlap in terms of volume and resolution and correlative studies become possible.

In this work we use both XCT and FIB serial sectioning to image the microstructure of an AlSi13 cast alloy. Data was partly acquired from the same sample volume to compare the information given by the different techniques. Pros and cons of both techniques are discussed.

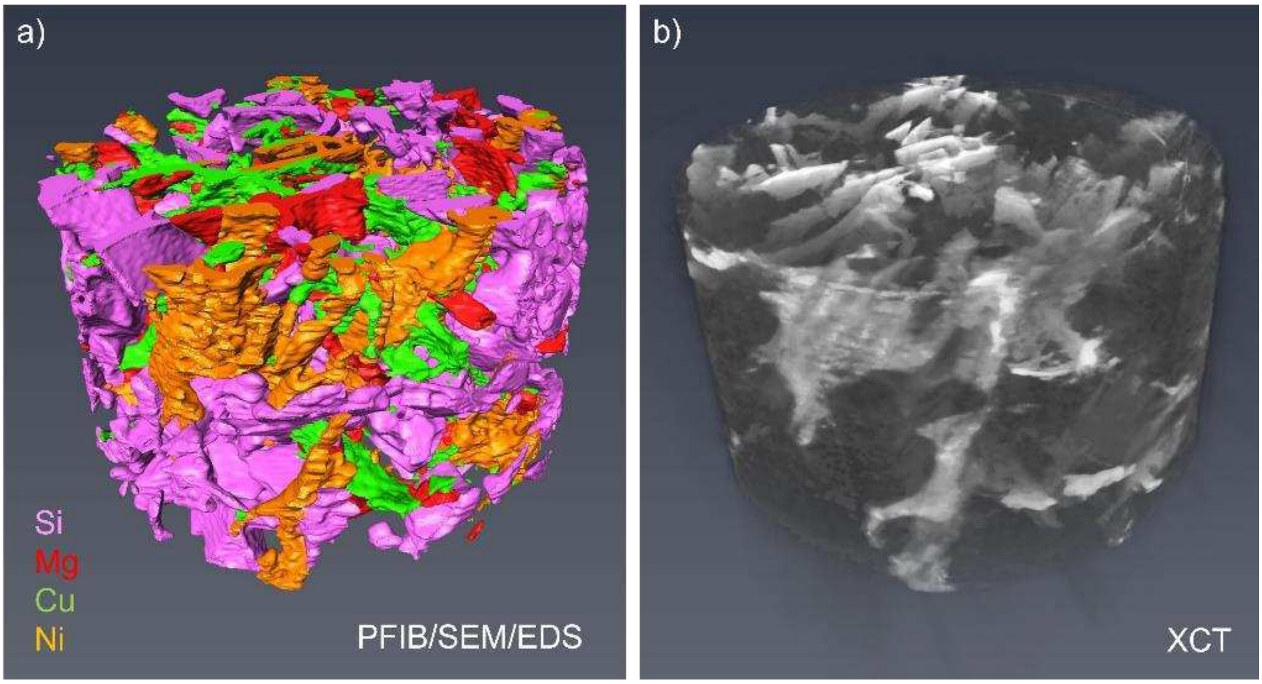

Figure 1: a) Reconstructed phases based on EDS data. Al matrix phase not shown for clarity, coloring according to principal elements. b) Volume rendering of XCT absorption contrast data. Phases containing heavy elements (Ni, Cu) appear bright.

Explore unlimited process pathways for FIB nanopatterning and ion imaging using VELION

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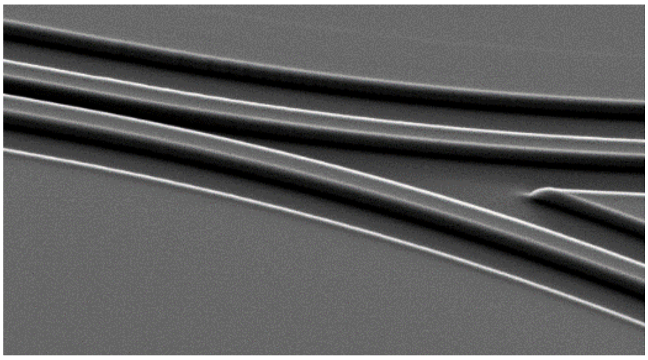
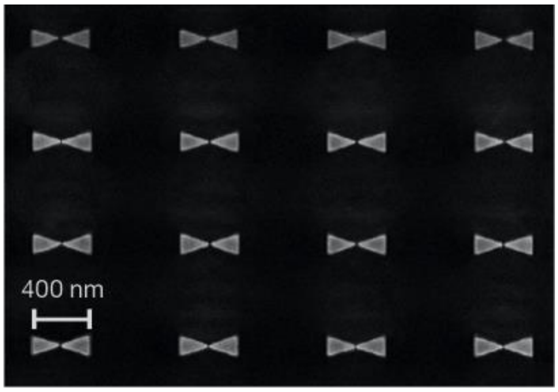
Focused Ion Beam (FIB) nano patterning has become established as a versatile and precise fabrication method of manifold features at the nanoscale. Applications in nanoscale science require high resolution fabrication techniques at high fidelity, accuracy and reproducibility over multiple write fields in an automated manner.
VELION's configurable multi-ion species FIB technology enables tailoring of various nanostructures according to application related challenges. Various ion species can be selected from universal ion sources providing fast or slow and light or heavy ions from a single source [1]. This approach paves the way for unlimited process pathways based on a FIB system merged with a true lithography platform. Unmatched large-area FIB patterning and unlimited perfect write field stitching or patterning overlay facilitates various patterning strategies according to specific applications. As VELION utilizes comprehensive automation for unattended, uninterrupted reliable nanofabrication over several days, the instrument is the ideal companion for machine driven nanofabrication.
In this contribution we present the instrument concept, an overview of various nanofabrication approaches and application such as direct patterning, hard masking, or sample functionalization.
Beyond nanopatterning, VELION FIB with its Liquid Metal Alloy Ion Sources (LMAIS) provides excellent ion beam imaging capabilities [2]. Lithium is the lightest ion for LMAIS available from periodic table and provides sub 2nm lateral image resolution. Latest results of 3D Mill & Image sample analysis will be presented utilizing the best depth milling resolution with Bismuth and superior lateral resolution with Lithium ions.

[1] 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).
[2] N. Klingner, G. Hlawacek, P. Mazarov, W. Pilz, F. Meyer, and L. Bischoff, Imaging and milling resolution of light ion beams from helium ion microscopy and FIBs driven by liquid metal alloy ion sources, Beilstein J. Nanotechnol. 11, 1742 (2020)

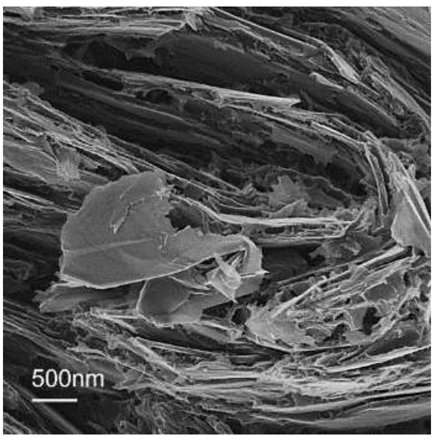
Left: General VELION setup with vertical FIB column, Laser Interferometer Stage and SEM
Right: Multi ion species FIB from Liquid Metal Alloy Ion Sources.



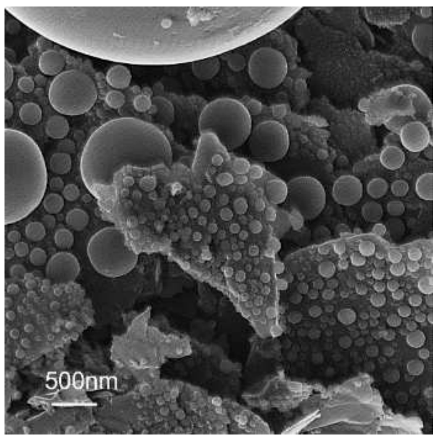
Waveguide coupler



Stepwise bowtie fabrication with Bi and Li ions



Lithium-ion image of graphite



Lithium-ion image of Sn/C

3D Nanoprinting of Electrical AFM Nanoprobes

L. M. Seewald[1], R. Winkler[1], M. Brugger-Hatzl[2], G. Kothleitner[2,3] and H. Plank[1,2,3]

[1] Christian Doppler Laboratory – DEFINE, Graz University of Technology, 8010 Graz, Austria
[2] Institute of Electron Microscopy and Nanoanalysis, Graz University of Technology, 8010 Graz, Austria
[3] Graz Center for Electron Microscopy, 8010 Graz
* corresponding author email: harald.plank@felmi-zfe.at

Along other Scanning Probe Microscopy techniques Atomic Force Microscopy (AFM) has evolved into a widely used characterization technology at the micro- and nanoscale. Highest lateral resolution is enabled using very sharp probes which are scanned across a sample. This approach allows to surpass Abbe's limit of diffraction and has thereby paved the way towards new insights and innovative applications in research and development. In addition to high resolution topo-graphical imaging, AFM enables simultaneous mapping of surface properties such as mechanical, electrical, chemical, magnetic or thermal, with nanometer resolution. Advanced operation modes, however, rely on functionalized probes. Commonly, standard Si tips are coated with relevant materials to induce a desired functionality. Disadvantages are a larger apex radius, reducing resolution capabilities, and the risk of delamination effects due to mechanical stress during AFM operation, which reduce or even eliminate the targeted sensitivity. Hence fully functional uncoated probes with very sharp apex radii would be desired. With its unrivaled flexibility in terms of structural delicacy, geometrical flexibility and minimal requirements to substrate material and morphology, Focused Electron Beam Induced Deposition (FEBID)[1] is perfectly suited for the fabrication of AFM probes. Post-growth treatments enable accurate tailoring of material's properties towards the intended application[2]. With this motivation in mind, we here present a Pt-based 3D hollow cone concept for application in electrical AFM modes (CAFM, EFM, KPFM). The presentation will cover the entire interlinked design and fabrication process to achieve mechanically stable probes with sub-10 nm apex radii as well as their chemical transfer into highly crystalline Pt structures preserving the mentioned shape aspects (Fig.1b). We then present AFM studies to compare the performance of our FEBID probes to commercially available probes both in terms of resolution (Fig.1c,d) and functionality (Fig.1e). Together with the fact, that those concepts are meanwhile patented in collaboration with our industrial partners, this contribution clearly shows the industrial relevance of 3D-FEBID in the area of Atomic Force Microscopy.

[1] Winkler et al, *3D nanoprinting via focused electron beams*, J. Appl. Phys. 125, 210901 (**2019**)
[2] Geier et al., *Rapid and Highly Compact Purification*, J. Phys. Chem. C 118, 14009 (**2014**)

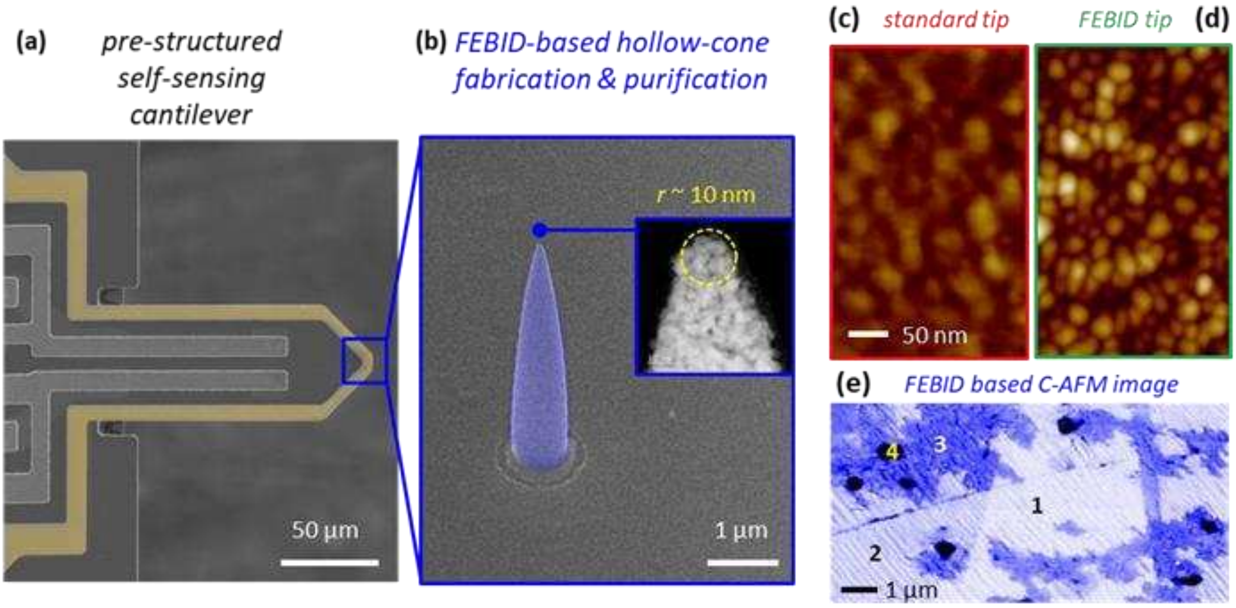


Figure 1: (a) pre-structured AFM self-sensing cantilever, which was first equipped with a PtC hollow-cone (b), further transferred into pure Pt with apex radii in the sub-10 nm regime (inset in b). (c) and (d) give a direct AFM topography comparison between commercially available C-AFM probes and the here relevant hollow-cones, respectively, which clearly shows the improved lateral resolution. (e) representative CAFM image enabling clear identification of a single-layer (1) and multi-layer graphene (2), copper oxide particles (3) and insulating Antimony particles (4) and proves CAFM applicability.

Manipulation and Study of Antiferromagnetic Order Enabled by Focused Ion Beam Fabrication

S. C. Haley[1,2], E. Maniv[1,2,3], and J. G. Analytis[1,2]*

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[2] Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA
[3] Department of Physics, Ben-Gurion University of the Negev, Beer-Sheva 84105, Israel
* corresponding author email: analytis@berkeley.edu

In ferromagnetic solids, electron spins prefer to align with each other, with macroscopically observable results and straightforward applications: souvenirs can stick to refrigerators, stable magnetic fields can translate electric currents into sound, and information can be stored in the direction of the collective magnetic moment of a material. In antiferromagnets, on the other hand, spins prefer to be anti-aligned with their neighbors. In practice, this yields much more complicated and varied patterns on a microscopic level, which are more challenging to study directly because they do not generate a net external magnetic field and are difficult to manipulate with one. We are nonetheless interested in these systems, however, both for their exotic phase diagrams and low-energy excitations, and for their use in technological applications made possible by their complex magnetic properties.

By fabricating specialized transport devices using the FIB, we have explored and tested a first-generation prototype for low-power computing components based on antiferromagnets. These devices have also found value as a novel tool for identifying unusual magnetic dynamics and textures that are challenging to study by other means.

Backside FIB/SEM analysis strategy to identify a new failure mode at an automotive magnetic sensor device

M. Simon-Najasek*[1], S. Hübner[1], M. Lejoyeux[1], A. Lindner[2], F. Altmann[1]

1 Fraunhofer Institute for Microstructure of Materials and Systems IMWS, 06120 Halle
2 TDK-Micronas GmbH, Hans-Bunte Strasse 10, 79108 Freiburg
* corresponding author email: michel.simon-najasek@imws-.fraunhofer.de

Integrated circuits (IC) used in automotive applications must be produced under high quality standards. To achieve this goal each single fail from qualification or also field application must be traced down to its physical failure root cause to increase the overall device reliability. In most cases the failure analysis (FA) strategy of IC related defects is straight forward by electrical testing, followed by defect localization and finally destructive physical analysis to identify the defect structure and derive the root cause of the electrical malfunction.

In some cases, such a strategy is not straight forward, especially if the failure site can not be exactly located or if the IC-package interaction causes the failure mode.

In this work we will demonstrate a failure root cause analysis of an automotive magnetic field sensor IC which fails by an electrical short between 2 metal layers, fig. 1. Over time a conductive path is created and at a certain resistance the devices fail with increased power consumption. This could be shown several times, but the question stays unanswered over a longer time regarding the primarily root cause. There are different hypotheses discussed, for example electrostatic discharge, package issues as well as imprints induced by probe tools.

In a standard FA procedure, the package is fully removed above the IC to get access to its functional surface structure. Then further localization and preparation steps are following. At this specific case the original root cause could not be identified from frontside, that’s why it was decided to turn the preparation starting from backside without removing the package at the frontside. The real challenge was that the failing position could not be localized in detail. Due to the electrical behavior of the sensor and the experiences collected by many fails before the position could be estimated and correlated to a several 10µm long metal network.

To get access to the ROI the Si substrate was mechanically thinned down to around 30µm thickness. After this the remaining silicon was removed by Xe-Plasma FIB trenching down to the active IC structure, fig. 2. Then the metal network was cut by Ga-FIB at several positions and investigated by passive voltage contrast to identify the shorted part, fig. 3. Finally, the remaining shorted metal lines were screened by forwarding FIB cross sectioning under SEM observation till irregularities (delamination, cracks) could be found in the IC structure. The defect origin could be identified due to the preserved mould compound on top of the IC. A large filler particle touches the IC passivation at the position of the cracks. Due to the mechanical stress during the moulding process these cracks were initiated, later metal was migrating into these cracks forming an electrically conductive path between metal 1 and 2, fig. 4. The same failure mode could be verified at several sensor devices.

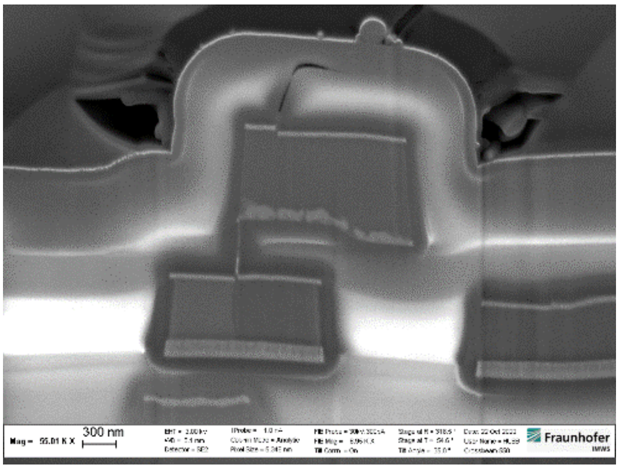

Fig. 1: SEM image of a FIB cross-section showing shorted metal lines due to a metal filled crack in-between these lines

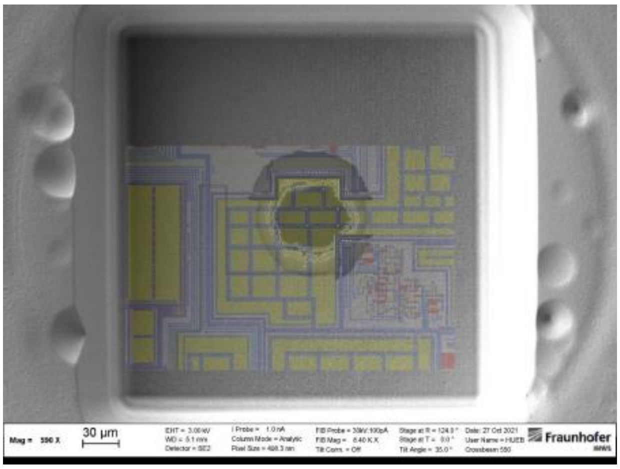

Fig. 2: P-FIB backside trenching of the Si substrate overlaid with the IC layout of the ROI, before and after detailed Ga-FIB investigation

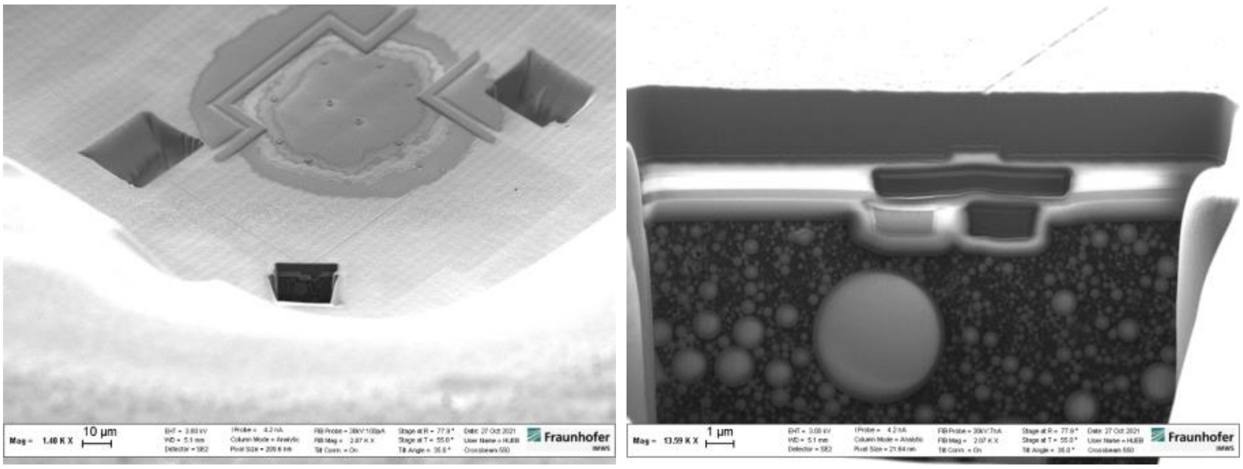

Fig.3: Passive voltage contrast investigation to isolate the defect position in the IC network

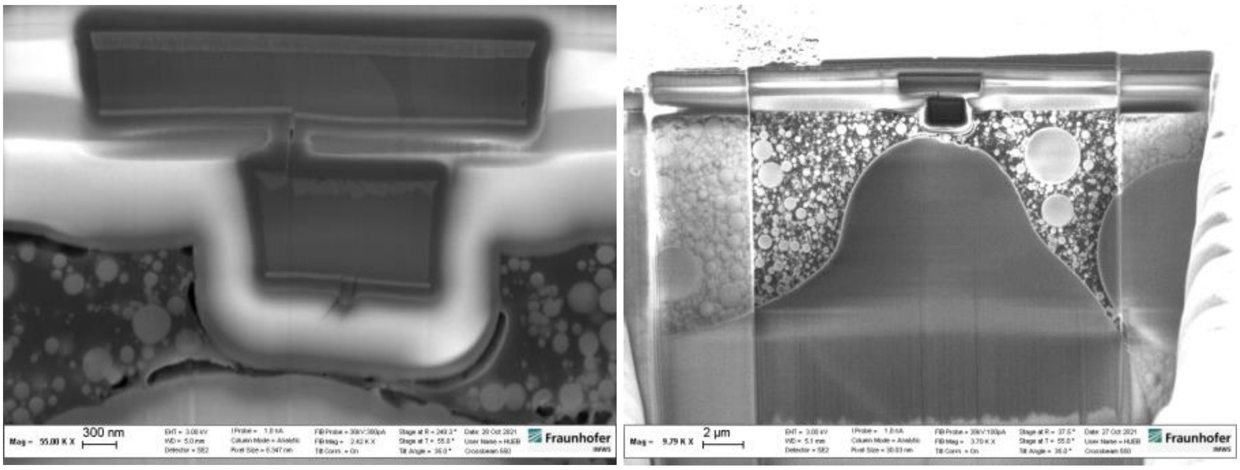

Fig.4: SEM cross-section showing a delaminated mould-passivation interface with cracked chip passivation and interlayer induced by a large filler particle

in situ Force Measurements - the FIB/SEM as a Mechanical Characterization Tool

A. J. Smith*, K. Schock, A. Rummel, S. Kleindiek

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* corresponding author email: andrew.smith@kleindiek.com

Mechanically characterizing materials inside FIB/SEMs is a common task with a myriad of use cases in various fields of research.

In this work, the authors will present several examples of how in situ force measurements can be performed using different setups inside the FIB/SEM's chamber, thus utilizing the FIB/SEMs capability to modify, customize or otherwise prepare samples for testing as well as image samples from different angles for a more comprehensive set of images for later analysis.

Examples include bending FIB cut beams and comparing EBSD results obtained prior to and post bend (Fig. 1), flat punch experiments for elucidating forging properties of novel superalloys (Fig. 2), characterizing nanowires,CNTs, and other structures (Fig. 3), etc.

The described measurements are achieved using one of three setups: 1. Smallest forces - in the range of some nN - can be measured using self-sensing, piezo-resistive AFM cantilevers. 2. Another option is to utilize moveable sample holders with precisely calibrated spring constants. In this manner the deflection observed in the FIB/SEM can be used to calculate the applied force. By choosing from spring loaded sample holders with varying spring constants, a wider range of forces can be addressed. 3. Force transducers can be used to measure large forces up to several N.

Each approach has its own distinct use cases, advantages, and disadvantages. These will be discussed, as well.

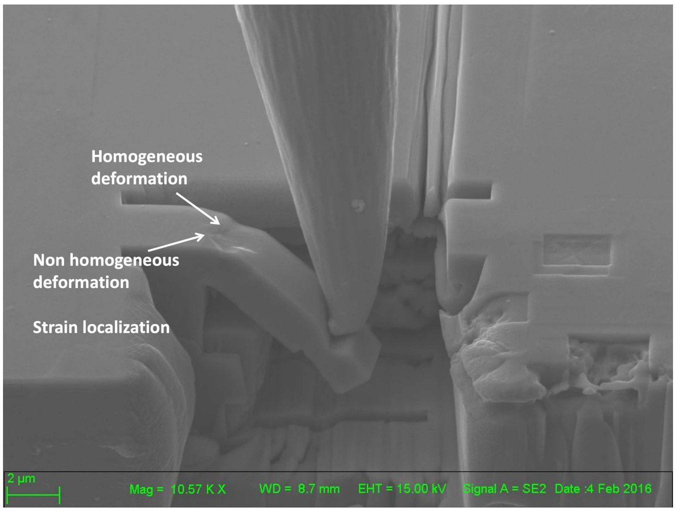

Fig. 1: FIB cut beam after bending failure (courtesy Archie, MPIE)

Fig. 2: Flat punch of a super alloy cube set in a FIB-cut base (courtesy Roesler, TU Braunschweig)

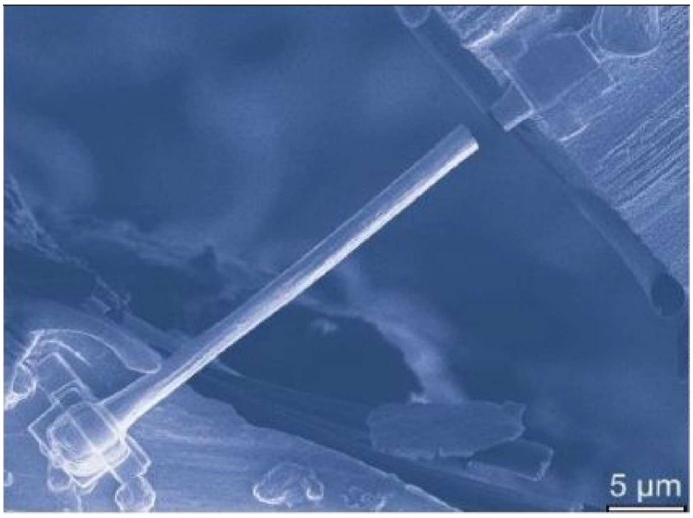

Fig. 3: Tensile test on a biological structure (courtesy Wegst, MPI Stuttgart)

Development and SEM integration of the Nano Aperture Ion Source

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[1] TU Delft
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The Nano-Aperture Ion Source (NAIS) is a new type of ion source, researched by L. van Kouwen, which should enable several ion species (Ga^+, He^+, Ne^+, Xe^+) without compromising brightness[1] $\geq 1 \cdot 10^5 A/(m^2 srV)$, or energy spread. An electron beam from a Schottky source is focused on a hole in a nano gas channel. Gas flowing through this channel is ionized due to electron impact ionization, after which it is extracted from the source by an extraction potential (*Fig.1*).[2] L. van Kouwen identified problems with chip design and experimental setup. The production yield of the NAIS chips, created by MEMS technology, was low because two membranes needed a voltage difference while maintaining a 1-micron separation. Therefore, the chip design has been altered. These changes have considerably improved production yield and the maximum pressure the chip can withstand, up to 3 bar. This alteration does negatively affect the possibility to extract the ions from the chip, since an extraction field can only be applied outside the chip.

Instead of an electric field within the chip, a structure can be used to guide the gas towards the extractor, which can be created using Electron Beam Induced Deposition (EBID) (*Fig.2*). To optimize this structure, several simulations are performed in the Rarefied Gas Regime using Python and SPARTA. Together with an electric field and ion trace simulations in GPT, a theoretical brightness can be acquired and optimized by changing the structural design. An experimental setup is being used to test chip life time and perform brightness measurements (*Fig.3*). This setup is modified by applying a new extractor with a 6D-aligner directly onto the chip and by decoupling the chip position and the electrostatic components. However, if the NAIS chip is to be used for FIB applications, a large demagnification of the source is required to reduce the ion beam spot size. Therefore, an additional setup is being developed by combining a commercially available SEM and FIB columns in one device on the same beamline (NiCole and Tomahawk columns provided by Thermo Fisher Scientific). This setup is scheduled to be ready in 2023 and should produce a usable prototype.

[1] Leon van Kouwen, Pieter Kruit; Brightness measurements of the nano-aperture ion source; Journal of Vacuum Science & Technology B (2018).

[2] Leon van Kouwen; Introduction to focused ion beams, ion sources, and the nano-aperture ion source; Advances in Imaging and Electron Physics Including Proceedings CPO-10 (2019), Start Page 181.

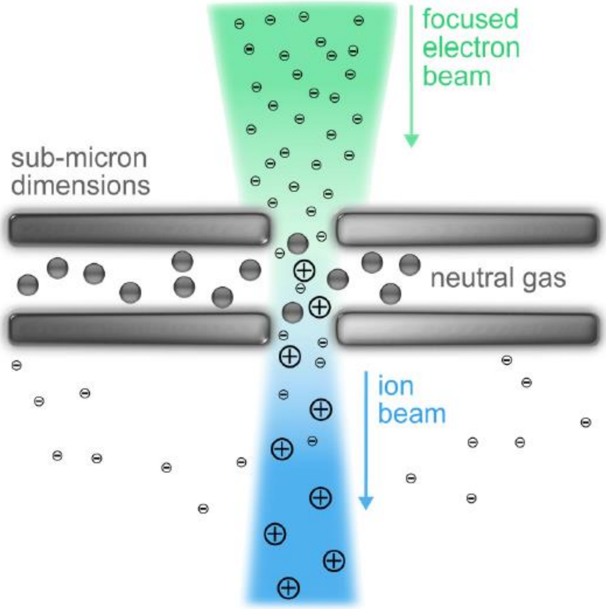

Fig.1 principle of the NAIS: gas is ionized by a focused electron beam and extracted by a strong electric field.

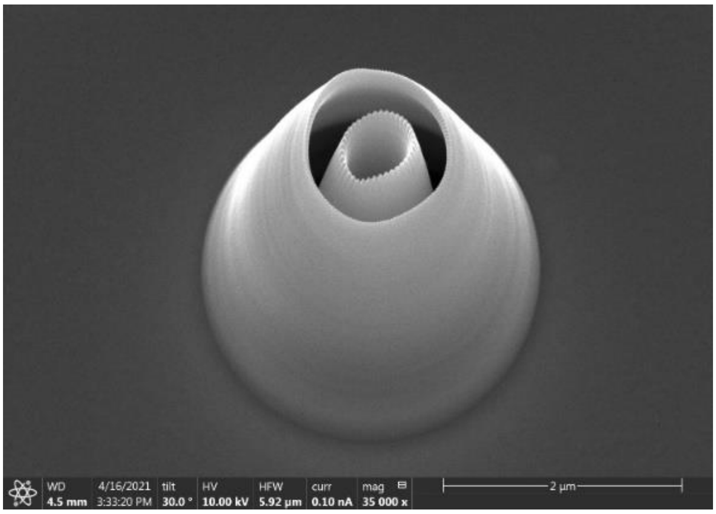

Fig.2 produced microstructure (EBID) to enhance ion beam performance.

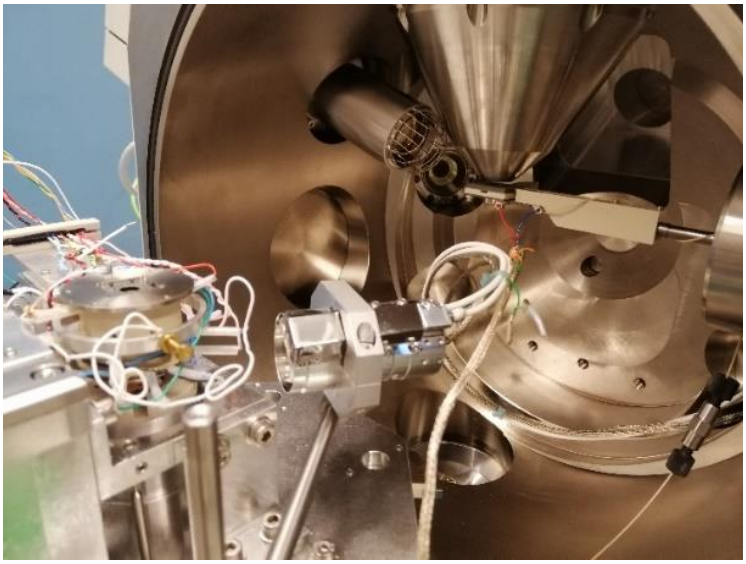

Fig.3 current experimental setup in SEM chamber.

Extending microstructure characterization from nm to mm scale using the latest multiple ion species plasma FIB integrated with femtosecond laser

Min Wu, Bartlomiej Winiarski and Remco Geurts

Thermo Fisher Scientific, Eindhoven, The Netherlands.

Conventionally 3D X-ray scanning technique such as microCT recovers large sample volume however suffers from relatively low voxel resolution, meanwhile high resolution 3D volume methods such as serial sectioning and imaging using a FIB-SEM can only recover 3D volume in the order of $\leq 40 \times 40 \times 40\ \mu m^3$. Plasma FIB-SEM expands these techniques to volume $\sim 250 \times 250 \times 250\ \mu m^3$ keeping the voxels sizes in the dozens of nm-ranges. Recently, femtosecond Laser PFIB-SEM pushed these 3D techniques further to mm-scale volumes, setting the standards for multi-modal data collection from nm to mm scales and bridge the gap between microCT and FIB-SEM, while maintaining the advantage of high resolution SEM imaging of the cut face.

In this abstract we present the latest development of multiple ion species plasma FIB integrated with femtosecond laser, the Thermo Scientific Helios 5 Laser Hydra system. We address the effects of ultra-short pulse laser ablation and discuss various application usecases using this latest versatile technology. We will also discuss the possibility of laser ablation and slicing materials with multiple ion species under cryogenic conditions using our latest 360º rotatory Aquilos cryo stage, and the newly developed inert gas transfer system CleanConnect.

We will also present the latest automation software from Thermo Fisher Scientific, AutoTEM 5 for lamellae fabrication and Auto Slice & View 5 for 3D serial sectioning and volume reconstruction.

AutoTEM 5 facilitates users to prepare the highest quality S/TEM samples and cross-sectioning with a reliable fully-robotic fashion. It is a unique solution with highly configurable workflow to enable preparation of a wide range of samples for Inverted, Top Down and Planar use cases. The complete in-situ sample preparation workflow features fully automated, unattended multi-site chunk milling, lift-out and final thinning.

Auto Slice & View 5 is the latest generation of Thermo Fisher Scientific's automation software for automated serial sectioning and imaging through a user-defined volume of a specimen. Auto Slice & View 5 provides all necessary features for high precision slicing and imaging through reliable automation, easy setup, monitoring and on-the-fly correction of project parameters. Some of the notable new features include editable job templates to simplify and speed up job setting, rocking beam polishing, and fast and reliable auto functions (auto focus, auto stigmation and auto lens alignment). In addition, a newly developed Spin mill function allows the software to automate sequential milling of large (up to 1 mm) areas. The sample surface is oriented almost parallel to the ion beam and the stage is periodically rotated to a series of pre-defined positions until a full rotation of 360º is fulfilled. Multiple areas for image acquisition could also be configured per every Spin mill site.

The patterning toolbox FIB-o-mat: Exploiting the full potential of focused ion beams for nanofabrication

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Future breakthroughs in nanotechnology will rely on the ability to fabricate materials and devices by design, i.e. to tailor both material properties and device geometries according to a sophisticated blueprint. Direct writing using focused beams of ions or electrons is a powerful technique not only for rapid-prototyping of novel device components but also for mask-less processing of delicate nanostructures and local modification of materials. To achieve an optimal result in the patterning process, a full control over the beam path including its rasterization is necessary. Here, we present the open-source Python toolbox FIB-o-mat [1] for automated pattern creation and optimization (cf. Fig. 1).

Patterning with an ion beam is a digital process in which the beam spot dwells for a defined time at a certain location and is then shifted by a defined distance (‘pitch’) to dwell again. This beam path can be created from a high-level geometry that is rasterized following a pre-defined routine. Therefore, FIB-o-mat provides various geometries including their Boolean combinations together with all possible rectangular and annular rasterization schemes. For complex geometries however, rasterization may lead to artifacts in narrow regions or for large curvatures. Here, FIB-o-mat offers a low-level approach where the beam path is generated by curve off-setting for any given input geometry to achieve the best patterning fidelity.

The functionalities of the Python-based toolbox FIB-o-mat are showcased for He ion beam processing of three different material systems. The magnetic properties of Co-based multilayers were locally modified using the high-level beam path generation combined with automation via stage control [2]. Plasmonic tetramer antennas were cut from single-crystalline gold on glass, demonstrating the ultimate patterning resolution of the focused He ion beam with gap sizes down to 3 nm (cf. Fig. 2). Apart from the low-level beam path generation, a local dose optimization had to be carried out. Finally, suspended single layer graphene was patterned into simple trampoline resonators but also complex phononic crystal structures [3] using the low-level beam path generation and automation via stage control.

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[2] L.M. Kern, et al.; *Deterministic Generation and Guided Motion of Magnetic Skyrmions by Focused He^{+}-Ion Irradiation*, Nano Lett. 22 (**2022**), 4028.

[3] J. N. Kirchhof, et al.; *Tunable Graphene Phononic Crystal*; Nano Lett. 21 (**2021**), 2174.

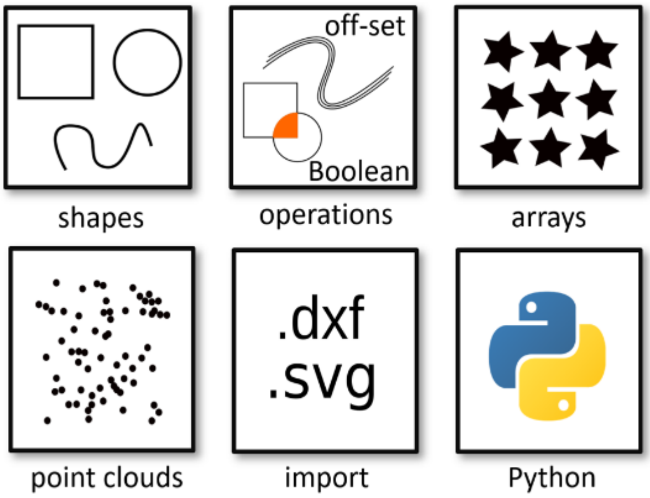

Fig. 1: FIB-o-mat tools

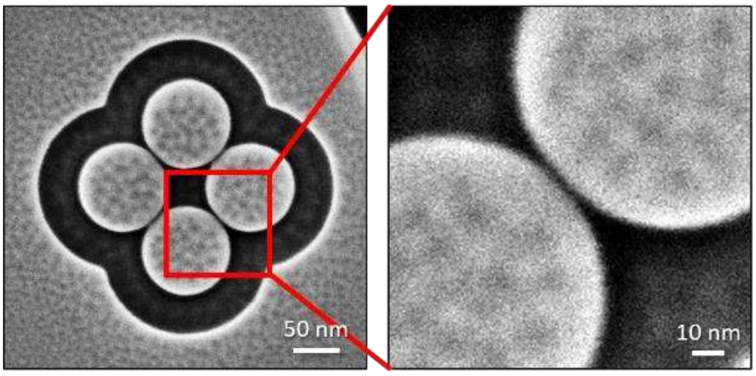

Fig. 2: Plasmonic tetramer antenna with ultimate gap resolution achieved by He ion beam patterning

Focused Ion Beam Induced Strain Generation in Silicon Membranes

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In this presentation we show an incredibly simple and controllable way of generating strain in suspended Si membranes using a Focused Ion Beam. The strain we observe approaches record levels observed in this material while retaining high crystallinity[1].

What this study also shows however is the power of serendipity. Without doubt the work contained here would not have occurred were it not for very substantial luck. The FIB induced strained region does not occur in the ion irradiated regions but in the non-irradiated regions adjacent to it. It was only possible to observe this effect due to presence of existing compressive stress, leading to buckling, in our 35 nm thick membrane substrates. This combined with an experiment that required us to irradiated them with a 30 kV ion beam led to the phenomena reported here.

FIB Irradiation of samples with energetic ion beams can create many effects such as doping, sputtering, chemical modification and damage creation. These effects depend greatly on ion energy, dose and species. The creation of damage and/or amorphisation due to the implantation of energetic ions is well understood and none more so than in the silicon system due its prominence in the semiconductor industry. For a normal incidence Ga ion at 30 kV the average ion range can be calculated to be approximately 25 nm deep, or a little less for an equivalent energy Xe ion. In both cases full amorphisation of the Si to a depth of the ion range occurs at less than 4 e14 ions/cm^2. For a focused ion beam to achieve doses of this intensity is trivial taking only a few seconds even at modest beam currents.

If 20 kV Ga ions or 30 kV Xe ions are implanted into a 35 nm thick Si membrane the ion range and hence crystal amorphisation corresponds to very close to the midpoint of the membrane thickness. This creates a bi-layer system that induces bending of the implanted membrane regions straining the adjacent unimplanted regions. Using simple FIB patterning strategies and exploiting crystal anisotropy we have shown it is possible to create biaxial strain in excess of 3% and uniaxial in excess of 8%. If the same technique were applied to germanium a transition to a direct band-gap semiconductor could be achieved at equivalent strains.

[1] Stress-strain engineering of single-crystalline silicon membranes by ion implantation: Towards direct-gap group-IV semiconductors

M.G. Masteghin, V. Tong, E.B. Schneider, C.C. Underwood, T. Peach, B. N. Murdin, R.P. Webb, S.K. Clowes and D.C. Cox. Physical Review Materials, Vol.5, 124603. 2021

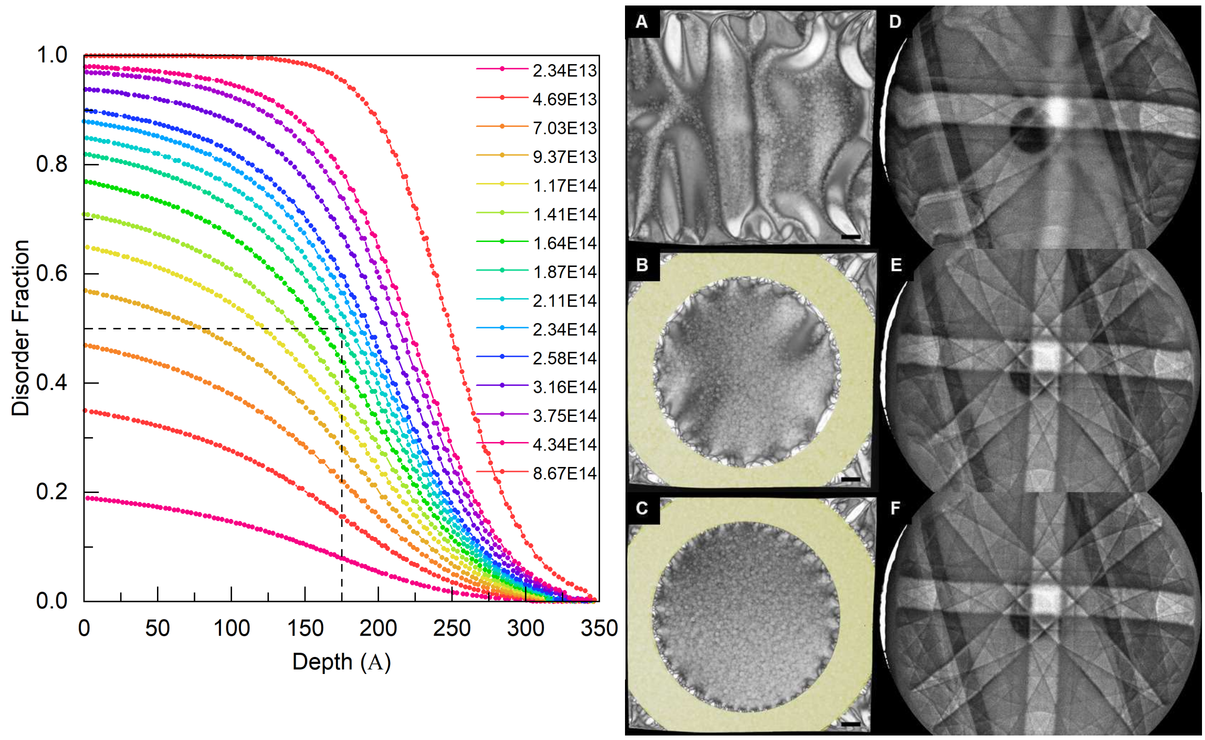

Fig. 1: (Left hand panel) Disorder fraction vs depth for 30 kV Xe ions into Si for the dose range 2.4 e13 to 8.7 e14 ions/cm^2 showing how the irradiated side of the membrane becomes amorphous but the lower side remains undamaged. (Right hand panel) The Si membrane prior to implantation (A), after partial exposure of the annular pattern, false coloured in yellow (B) and the same sample at a higher dose where the membrane buckles have been removed (C). The diffraction patterns demonstrate how the central region of the membrane is pulled flat but retains its crystallinity. The inner annular diameter is 80 μm.

Two Microscopes are better than One – *In-situ* Correlative Analysis by combination of AFM, SEM, and FIB

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[1] Quantum Design Microscopy GmbH, 64293 Darmstadt, Germany
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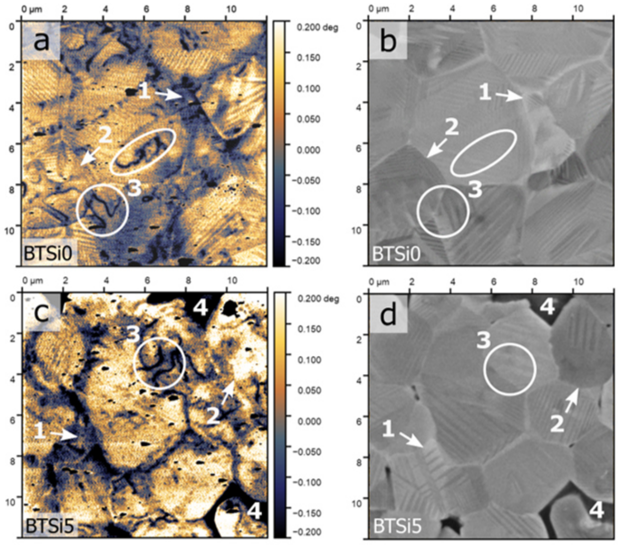
Combining different analytical methods in one instrument is of great importance for the simultaneous acquisition of complementary information. Especially the *in-situ* combination of scanning electron microscopy (SEM) and atomic force microscopy (AFM) enables completely new insights into the micro and nano-world. In this work, we present the unique *in-situ* combination of scanning electron / ion microscopy (SEM/FIB) and atomic force microscopy (AFM) for nanoscale characterization [1-2].

A particularly important aspect for advanced AFM measurements are the cantilevers itself. Especially the cantilever tips have to provide special properties to measure not only the topography, but also magnetic and electrical characteristics of materials. In this context, the FEBID process is a very promising approach for the design and fabrication of sophisticated tips with unique properties. We will present some case studies to highlight the advantages of interactive correlative *in-situ* nanoscale characterization for different materials and nanostructures, using FEBID-constructed tips. We show results for the *in-situ* electrical characterization by conductive AFM for 2D materials as well as electrostatic force microscopy (EFM) measurements of piezoceramic films that enables the precise analysis of grain boundary potential barriers in semiconducting $BaTiO_3$-based ceramics [3]. The grain boundaries were located via BSE-SEM and measured afterwards using the in-situ EFM method. The barriers were shown to be significantly thinner and more pronounced as the amount of SiO_2 was increased from 0 to 5 mol% (see Fig. 1). These results can be directly correlated with electron backscatter diffraction (EBSD) measurements in order to link the AFM and SEM data to the crystallographic microstructure.

In addition, we will present results for the *in-situ* characterization of magnetic nanostructures by combination of SEM and high-vacuum magnetic force microscopy (MFM). For the *in-situ* MFM measurements, special high aspect ratio magnetic cantilever probes fabricated by electron beam induced deposition (FEBID) were used, which surpass conventional cantilevers in terms of lateral and magnetic resolution. The SEM enables to identify the grain boundaries on multilayer thin-film samples or stainless steel in order to measure the magnetic properties directly via MFM with nanometer resolution (see Fig. 2).



	C–V [μm]	EFM [μm]
BTSi0	0.89 ± 0.04	0.92 ± 0.13
BTSi2	0.47 ± 0.02	0.50 ± 0.08
BTSi5	0.53 ± 0.03	0.52 ± 0.08

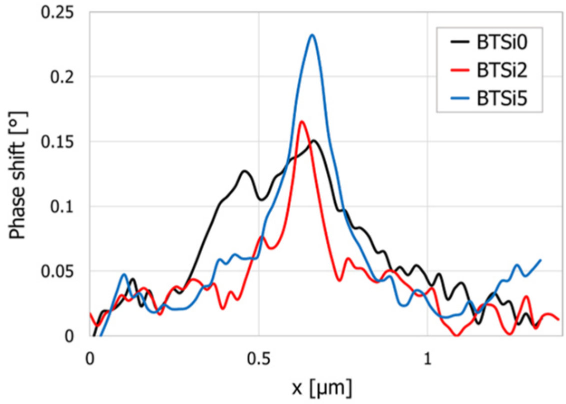


Fig 1: (Left) EFM signal of (a) BTSI0 and (c) BTSi5 with corresponding BSE-SEM images (b) and (d) of the exact same sample area. The EFM images were recorded with an external voltage of -3 V. (Right) Grain boundary potential barriers of $BaTiO_3$ based ceramics with varying SiO_2 content. The Images were extracted from [3].

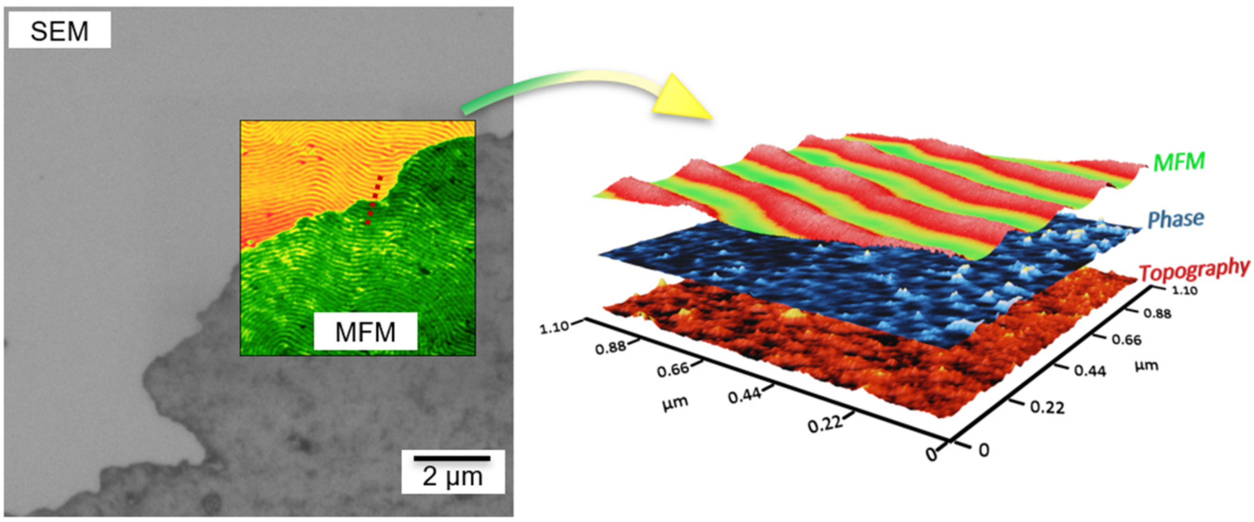


Fig 2: (Left) SEM image of a multilayer magnetic sample with correlative MFM image at a grain boundary. (Right) AFM data overlay of topography, phase, and MFM signals.

[1] D. Yablon, et al.; *Cross-platform integration of AFM with SEM: Offering the best of both worlds*; Microscopy and Analysis 31 (2) (**2017**), 14.

[2] S.H. Andany, et al.; *An atomic force microscope integrated with a helium ion microscope for correlative nanoscale characterization*; Beilstein Journal of Nanotechnology 11 (**2020**), 1272.

[3] J.M. Prohinig, J. Hütner, K. Reichmann, S. Bigl; *Complementary evaluation of potential barriers in semiconducting barium titanate by electrostatic force microscopy and capacitance-voltage measurements*; Scripta Materialia 214 (**2022**), 114646.

Direct-Write 3D Nanoprinting of High-Resolution Magnetic Force Microscopy Nanoprobes

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Magnetic devices play an important role in modern electronic, sensing or data storage applications. To exploit their full potential, high-resolution Magnetic Force Microscopy (MFM) is established as standard characterization technology as part of the research and development loop. Due to the ongoing trend towards smaller and smaller active feature sizes, the demands on high-resolution MFM tips are also increasing. Based on that motivation, we here aim on the fabrication of MFM nanoprobes with functional apex radii in the sub-10 nm regime. Traditional products mostly base on additional magnetic coatings, which increases the apex radii and therefore limits the lateral resolution during Atomic Force Microscopy (AFM) based MFM measurements. Another disadvantage of a magnetic coating is local delamination, which can occur due to the mechanical stress during scanning and lead to a change (or even complete loss) in magnetic sensitivity. Therefore, it was the goal to fabricate fully magnetic nanoscale tips, that do not require additional coating. Focused Electron Beam Induced Deposition (FEBID) was used for additive, direct-write 3D-nanoprinting of such magnetic tips on pre-finished self-sensing AFM cantilevers.[1],[2] For that, a novel $HCo_3Fe(CO)_{12}$ precursor was used, which is one of the few precursors, providing metal contents above 90 at.% after initial FEBID fabrication.[3] To explore the possibilities, we comprehensively studied the parameter space and their implications on morphology, structure and chemistry in detail by using SEM, EDX, and TEM and STEM EELS (Figure 1.a). Next, the tip geometry was further optimized by an advanced, dynamic pattering sequence to fulfil the high demands for AFM operation.[4] Additionally, the fabricated tips were subjected to different post-processing procedures such as post-irradiation with electrons, thermal treatments and purification protocols to explore and identify the most promising fabrication window. The basic performance of such MFM tips is then demonstrated with special focus on lateral resolution, magnetic phase shift and signal-to-noise ratio. Fully optimized FEBID-MFM tips were then tested on various magnetic samples (magnetic multilayer system (Figure 1.b-e), hard disc drives, magnetic recording tapes) and benchmarked to commercially available MFM tips (Figure 1.b-c). Finally, the wear resistance of such MFM nanoprobes was evaluated during a continuous operation scan over a period of 3.7 hours, which revealed the high durability of the presented concept (Figure 1.d-e). By that, we demonstrate the successful 3D-nanoprinting of MFM tips on self-sensing cantilevers, which fulfils the high requirements when aiming on industrially relevant MFM tips using FEBID-based 3D nanoprinting.

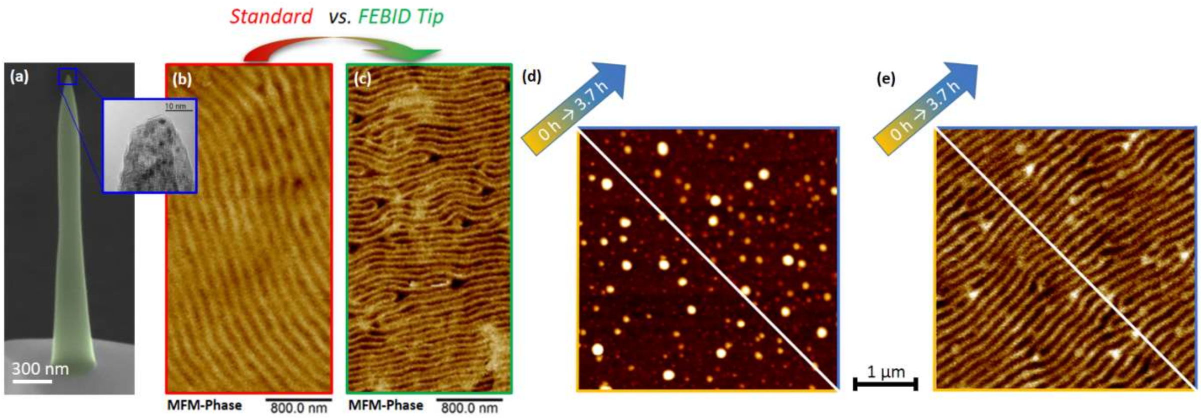

Figure 1: (a) SEM image of a specifically tailored, Co_3Fe 3D nanoprobe for Magnetic Force Microscopy (MFM). The inset shows a TEM image of the tip region, revealing a fully crystalline tip radius around 10 nm. (b) and (c) show a direct comparison of magnetic MFM maps, taken with a commercial and a FEBID-based MFM tip, respectively, which reveal the superior performance of the latter. (d) and (e) demonstrate the wear resistance of FEBID tips via topography and magnetic phase maps, respectively. The lower left parts (yellow) show the starting situation, while the upper right parts (blue) give the results after continuous AFM / MFM operation of 3.7 hours, revealing practically identical results, which underlines the durability of FEBID based MFM tips.

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Using the Helium Ion Microscope for Imaging and Modification of Nanostructures, 2D Materials, and SARS-CoV-2 infected Cells

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The Helium Ion Microscope (HIM) utilizes a focused beam of helium ions to image and modify materials with high spatial resolution, large depth of field, and chemical sensitivity [1]. HIM images show stronger chemical and topographical contrasts than images from the related scanning electron microscope, and the HIM is capable to resolve sub-nanometer features. Due to its charge compensation capability, the HIM can image insulating biological samples without additional conductive coatings. My presentation will contain examples of HIM imaging of nanomaterials, like 1 nm thick carbon nanomembranes (CNMs), 2D materials, and biological cells [2]. In an exploratory HIM study of SARS-CoV-2 infected Vero E6 cells, interactions between cells and virus particles, as well as among virus particles, could be imaged [3]. The HIM pictures show the three-dimensional appearance of SARS-CoV-2 and the surface of Vero E6 cells at a multiplicity of infection of approximately 1 with great morphological detail. The absence of a conductive coating allows a distinction between virus particles bound to the cell membrane and virus particles lying on top of the membrane. When applying higher ion currents, the HIM can be also used for the modification of materials. The capability of the HIM for nanolithography will be shown by milling 2D materials, where nanopores with diameters down to 1.3 nm were fabricated [4].

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Sample preparation and analysis of LLZO ceramics for solid state batteries with Cryo FIB/SEM and aberration corrected analytical STEM

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[1] NMI Natural and Medical Sciences Institute
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Solid state batteries are promising devices for high capacity energy storage in future electric vehicles. For stable, efficient and long life operation of these batteries stable interfaces of different layers are crucial. To optimize the interface between lithium and the solid electrolyte various interlayer materials are under development. Here we report our cryo-FIB/SEM, HRSTEM and EELS analysis of aluminum doped LLZTO (Lithium Lanthanum Tantalum Zircon Oxide) solid-state electrolyte coated with tin interlayer.

To analyze the morphology and chemical composition of coated LLZTO, FIB (focused ion beam) cross sections were prepared (Zeiss crossbeam 550 scanning electron microscope) and analyzed with EDS (Oxford X-Max 150). In a second step, a several µm thick lamella was prepared by FIB lift out technique [1]. To reduce beam induced damage the sample was finally polished inside the FIB/SEM under cryogenic conditions (-160° C, Quorum PP3010). After finishing a TEM transparent lamella, the sample was warmed up to room temperature and transferred via an argon glove box on a TEM vacuum holder. High resolution analysis of the lamella was performed in a probe corrected JEOL ARM 200F electron microscope equipped with a JEOL Dual EDS system. The Li K line is not accessible in conventional EDS, therefor we used EELS (Electron Energy Loss Spectroscopy, Gatan Quantum ER Spectrometer) to measure the distribution of Lithium.

Results of STEM imaging and EELS mapping are shown in figure 1. STEM imaging with atomic resolution of the aluminum precipitate is shown in figure 2.

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[2] Mayo, M. and A.J. Morris, Structure Prediction of Li–Sn and Li–Sb Intermetallics for Lithium-Ion Batteries Anodes. Chemistry of Materials, 2017. 29 (14): p. 5787-5795

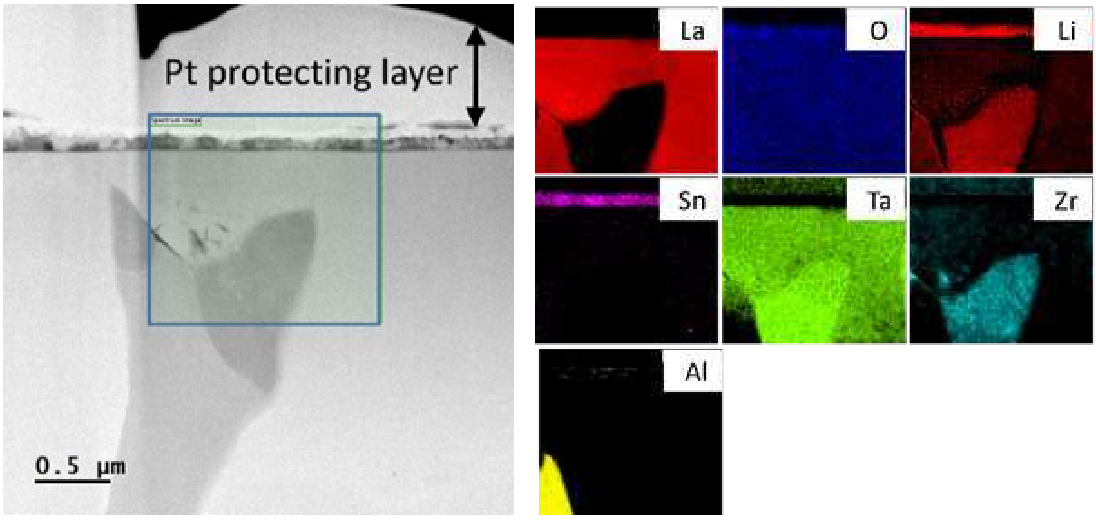

Fig. 1: STEM ADF image and EELS elemental mapping at 200keV in JEOL ARM200F.

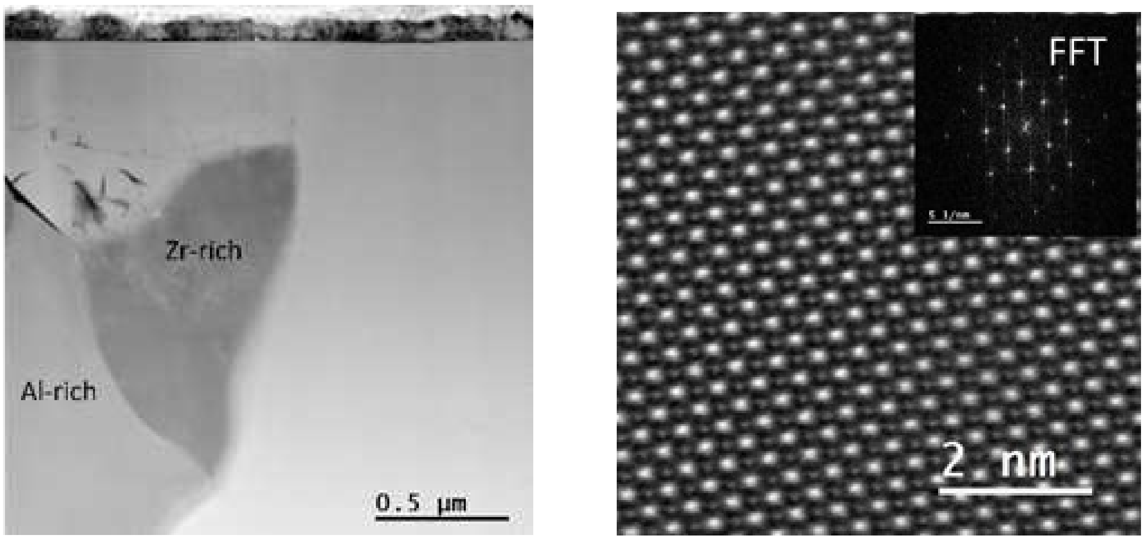

Fig.2: STEM ADF image (left) and atomic resolution image of the aluminum rich precipitate (right).

Helium ion microscopy and sectioning of Spider Silk

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Focused ion beams have recently emerged as a powerful tool for ultrastructural imaging of biological samples. In this presentation, we will show that Helium Ion Microscopy (HIM), combined with ion milling, can be used to visualize the inner structure of both Major and Minor Ampullate silk fibres of the orb-web weaving spider *Nephila madagascariensis*. The internal nanofibrils were imaged in pristine silk fibres, with little or no observed damage to the sample structure. Furthermore, a method to cut/rupture the fibres using He^+ ions combined with internal sample tension is presented.

This work showed that HIM is a valuable tool for visualizing biological samples. The inherent sputtering of the He^+ ions could be used to mill the samples without damaging the biological structures softly. This contrasted with the Ne^+ ions, which caused damage to the spider silks' internal structure.

It was also shown that combining He^+ ion milling and the inherent tension in the spider silk sample made it possible to cut the specimen and visualize the rupturing process and the internal structures in the silk. This method could be used for other fibrous structures or even with non-elastic samples if used with stretched adhesive carbon tape, which could be used to apply the rupturing force.

The HIM images of the spider silk revealed that the rupturing process was highly dynamic involving rearrangement of the material in the fibre and showing strong indications of an internal fibril structure in the silk fibres with typical dimensions of 100-200nm. We anticipate that HIM will significantly contribute to some of the most challenging imaging applications and may open new directions in future bioimaging when paired with other imaging modalities.

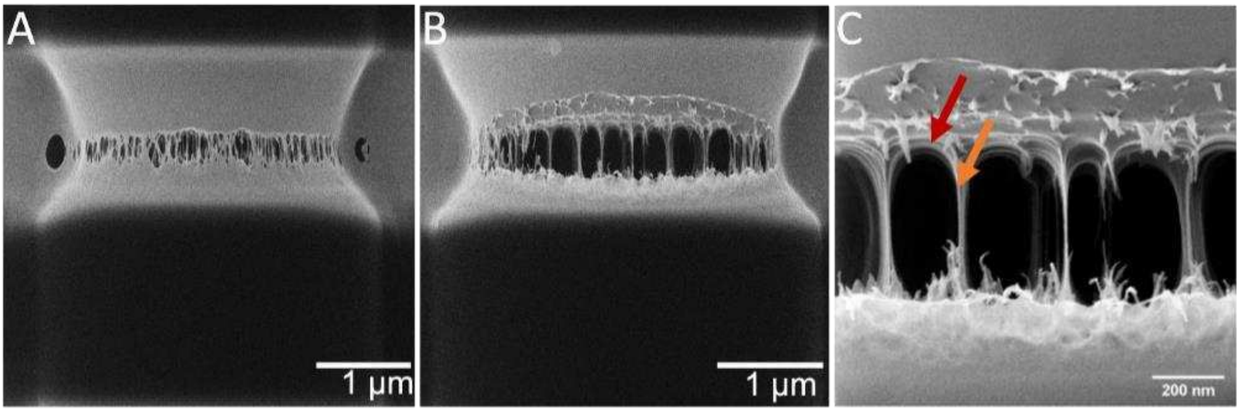

Fig. 1: HIM images of a MiS fiber that has been cut in half by He+ ion sputtering. A) HIM image of a MiS fiber before breaking. B) image of the fiber stretching. C) Further zoom in on B) showing the individual fibrils being stretched (orange arrow), causing deformation in the base of the fibril (red arrow).

Best Practices for Xe PFIB Preparation of Materials for Transmission Electron Microscopy

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Over the past decade, there has been a concerted effort to develop gallium (Ga)-free focused ion beam (FIB) instruments for materials research. The helium ion microscope (HIM), neon ion microscope (NIM), and other similar inductively coupled plasma (ICP) multi-ion source focused ion beams such as oxygen (O) and argon (Ar) have been explored, though none have shown to be effective for the preparation of samples for transmission electron microscopy (TEM). Xenon (Xe) plasma focused ion beam (PFIB), thanks to its heavier ion mass (and thus high material sputter yield) and chemical inertness, has become the go-to instrument for large length scale (>100 µm) crossing sectioning and FIB-SEM tomography. But can the Xe PFIB fabricate TEM samples appropriate for nanoscale microstructural characterization?

Xe PFIB has its own set of challenges relative to Ga FIB. Xe PFIB would not typically be the go-to tool for nanoscale milling applications like TEM sample preparation because the ICP Xe PFIB will produce a beam with a larger diameter than the liquid metal ion source (LMIS) Ga FIB in the range of currents typically used for TEM prep. A beam with a larger diameter can be more difficult to use in smaller scale (< 500 nm) applications. The Xe PFIB also has a greater range of current density across the width of the beam. The outer-most edges of the ion beam, called beam tails, are more prominent in the Xe PFIB compared to the Ga FIB whose beam tails are virtually non-existent in most modern FIB instruments. The Xe PFIB beam tails add complexity to finer scale milling applications such as thinning a TEM sample to electron transparency.

In this talk, we will demonstrate that it is possible to create Ga-quality TEM samples using Xe PFIB. This quality is defined as curtain-free and uniformly thin over any 25 μm^2 area where the relative thickness (t/λ) measured with electron energy loss spectroscopy (EELS) is less than or equal to 1. We will identify best-practices and ideal parameters settings (including deposition energy (kV), deposition height, ion beam incident milling angle, and beam placement on the sample) to show that it is possible to fabricate two types of Xe PFIB-made samples: 1) a curtain-free uniformly thin electron transparent area where $t/\lambda \approx 0.9$ over 38 μm^2, and 2) a curtain-free non-uniformly thin electron transparent area where t/λ ranges from 0.3 to 1.1 over 48 μm^2. Both sample types exhibit most, but not all, aspects of Ga-quality. Because of Xe PFIB's relatively large beam size and wide beam tails, a balance must be struck between ultimate sample thinness and the overall size of the electron transparent area.

New Applications in Energy Research Enabled by a Triple Beam, Dual Chamber FIB with Isotropic Tomographic Voxels

Ben Tordoff, Ph.D.

EUFN 2022

Energy materials research relies heavily on a deep understanding of material and device microstructure to make advancements. Moreover, because these materials exist in complex devices constructed of many materials and interfaces between them, the critical information is often buried beneath the surface of a material specimen or encapsulated in a closed-form device. Furthermore, the final devices derive their performance from the nanoscale 3D arrangement and microstructures of the constituent materials involved. Recently, a commercial focused-ion beam scanning electron microscope was developed with an integrated fs-laser mill attached to the load lock of the microscope, opening the door to new and more powerful analysis approaches in energy materials research. Applications include rapid access to deeply buried structures for high resolution imaging and analytics, large area cross-section preparation, and massive material ablation for sample preparation of structures for, e.g. FIB-SEM tomography, TEM, APT, or X-ray nanoCT. Additionally, each of these may be correlatively guided by prior imaging with techniques like 3D X-ray microscopy to enable targeted analysis and preparation. By combining the laser mill with leading technologies for 3D FIB-SEM tomography with true isotropic voxels, this platform enables comprehensive quantitative analysis of energy materials at the nanoscale. We illustrate these concepts with a number of application examples and use cases demonstrating the utility of the approach.

Application of FIB-TOF-SIMS for 3D high-resolution chemical characterization of Li-ion solid-state batteries

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The growing interest in Li-ion batteries (LIBs) is particularly driven by the vast need for electric vehicles (EVs), providing environment-friendly (i.e., renewable, clean and zero-emission) energy storage systems.[1] The development of new chemical systems, ensuring improved LIBs' capacity, power and efficiency as well as the reduction of production costs, was a great milestone. However, the safety issues as well as short lifetime are still the critical problems of the conventional LIBs. All-solid-state-batteries (ASSBs) using inorganic solid-state electrolytes (SSE) are great alternatives to commonly used LIBs based on highly flammable organic liquid electrolytes as they enable for operation in a wide range of temperatures.[2] Therefore, ASSBs are considered as one of the most promising future energy storage technologies. The characterization of ASSBs is demanding because they contain buried structures and heterogeneous interfaces. Furthermore, detection of Li and representing its 3D distribution with nanoscale resolution is attainable only to few analytical techniques. In this work, we exploit the outstanding potential of FIB-TOF-SIMS (focused ion beam time-of-flight secondary ion mass spectrometry, Figure 1) for comprehensive chemical characterization of novel Li-containing thin films, which are potential materials for the future generation batteries. This technique allows for parallel detection of all sample components with high spatial resolution (i.e., lateral resolution < 50 nm and depth resolution < 10 nm)[3–5] and high sensitivity (ppm). Our studies show that FIB-TOF-SIMS can reveal presence of 400±100 nm overlithiated grains and 100±30 nm nanoparticles with an increased $^{7}Li^{16}O^{+}$ ion content in the Li- and Ni-rich layered oxide with the composition of $Li_xNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ (LR-NMC811, x>1) as well as monitor structural changes upon air exposure (both on the surface and in the bulk).[6] Furthermore, we demonstrate that simultaneous delivery of fluorine gas during FIB-TOF-SIMS can significantly improve the quality of acquired TOF-SIMS data.[7–11] The latter is proved using a novel Au/$Li_7La_3Zr_2O_{12}$/Pt/MgO/Si multilayer. In this case, the enhanced TOF-SIMS data helped understand the operation of the system. The detection of Li within Au layer after the polarization measurements explained the previously observed formation of internal electric field.[12] Our studies prove that FIB-TOF-SIMS is a powerful technique delivering essential insights into the complex structure of novel

Li-based materials, which can help optimize the functionality of future energy storage technologies.

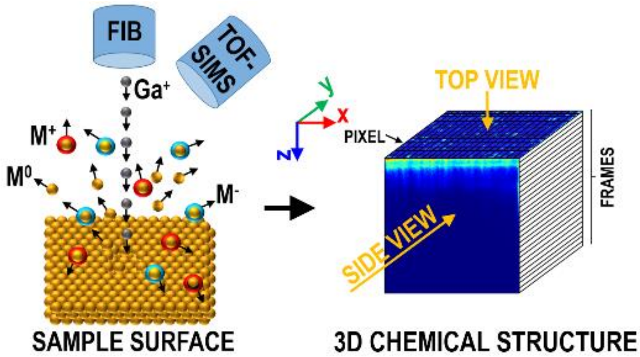

Fig. 1: FIB-TOF-SIMS is one of few analytical techniques allowing sample's 3D chemical structure to be assessed with nanoscale resolution.

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On demand spatially controlled fabrication of single photon emitters in Si

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Single photon emitters (SPE) are fundamental building blocks for future quantum technology applications. However, many approach lack the required spatial placement accuracy and Si technology compatibility required for many of the envisioned applications. Here, we present a method to fabricate at will placed single or few SPEs emitting in the telecom O-band in Silicon [1] . The successful integration of these telecom quantum emitters into photonic structures such as micro-resonators, nanopillars and photonic crystals with sub-micrometer precision paves the way toward a monolithic, all-silicon-based semiconductor-superconductor quantum circuit for which this work lays the foundations. To achieve our goal we employ home built AuSi liquid metal alloy ion sources (LMAIS) and an Orsay Physics CANION M31Z+ focused ion beam (FIB). Silicon-on-insulator substrates from different fabrication methods have been irradiated with a spot pattern. 6 to 500 Si^{2+} ions have been implanted per spot using an energy of 40 keV. For the analysis and confirmation of the fabrication of true SPEs a home build photo luminescence setup has been used. G-centers formed by the combination of two carbon atoms and a silicon atom are confirmed by measurements of zero phonon lines (ZPL) at the expected wave length of 1278 nm for the case of carbon rich SOI wafers. In the case of ultra clean SOI wafers and high ion fluxes emission from tri-interstitial Si complexes is observed. The SPE nature of these so called W-centers has also been confirmed by ZPL measurements at 1218 nm. The achieved lateral SPE placement accuracy is below 100 nm in both cases and the success rate of SPE formation is more than 50%. After a discussion of the formation statistic we also present an approach how our FIB based approach can be upscaled to wafer-scale nanofabrication of telecom SPEs compatible with complementary metal oxide semiconductor (CMOS) technology for very large scale integration (VLSI).

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Magnetic patterning using Ne, Co, and Dy FIB

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Magnetic nanostructures needed for magnonics and spintronics are usually processed by conventional lithography techniques in combination with lift-off or broad-beam ion etching. However, it has been shown [1] that the quality and shape of the structures' edges play an important role for the magnetization dynamics as structures become smaller and smaller. Furthermore, regarding optical measurement techniques, hard-to-remove resist masks that become hardened by ion etching are problematic. Direct-writing focused ion beams (FIB) do not have these issues. In addition, using non-standard ion species opens various paths for local magnetic patterning, i.e., influencing the magnetic properties locally.

I will present results for maskless magnetic patterning of ferromagnetic nanostructures using He and Ne ions as well as a few liquid metal alloy ion sources (LMAIS) for FIB systems. He/Ne FIBs are well established and commercially available. Irradiation of (paramagnetic) FeAl films by Ne ions creates local ferromagnetic nanostructures caused by disorder that are embedded in a paramagnetic matrix [2]. The precise Ne FIB also enables us to trim the edges of magnetic nanostructures enhancing their magnetic fidelity and creating certain localized magnon states at the edges of the samples. Using specifically developed LMAIS, like e.g., $Co_{36}Nd_{64}$, CoDy, or CuDy [3,4] in combination with a Wien mass filter offers further new paths for magnetic patterning. I will present results on the modification of $Ni_{80}Fe_{20}$ (permalloy) strip samples. Using the CoNd LMAIS a narrow track of Co ions was implanted. The induced magnetic changes were measured with microresonator ferromagnetic resonance (FMR) before and after the implantation. Structures as small as 30 nm can be implanted up to a concentration of 10 % near the surface. Such lateral resolution is hard to reach for other lithographic methods. Using Dy ions one can locally increase the Gilbert damping parameter of the magnetization dynamics by more than a factor of four with a lateral resolution of about 100 nm.

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fibTOF: The strength of SIMS capabilities on FIB-SEM microscopes

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The combination of a FIB-SEM microscope with a mass analyzer provides a cost-effective route to high spatial resolution chemical imaging using secondary ion mass spectrometry (FIB-SIMS) [1]. The high mass resolving power, together with the ability to collect information about all elements simultaneously, has made the use of time-of-flight mass analyzers popular for this purpose, especially in the field of materials science. Because information about all mass to charge ratios is collected, retrospective analysis of the data sets can allow for new insights into local elemental distributions and correlations without repeating the measurement. FIB-SIMS is based on the direct measurement of the sputtered ions. The yields of secondary ions are in general high for positive ions from alkali metals and, also for negative ions from halogens. This is especially true for lithium and fluorine, which can be difficult to map by other techniques. FIB-SIMS is therefore a technique well placed to support research of current and next-generation rechargeable battery materials (e.g., for lithium ion, sodium ion and fluoride ion batteries).

The correct identification of the sputtered ions is mandatory for the characterization of the material properties. Knowledge about the possible sample composition is required for the data interpretation. A high mass resolving power is essential to resolve molecules from elements at the same atomic mass unit. Molecular fragments containing the element of interest can confirm the correct elemental identification.

FIB-SIMS is thus a powerful technique that can complement other characterization methods (electrochemical, XRD, SEM imaging, or XPS). Having the specimen within the vacuum chamber of a FIB-SEM microscope means that a fresh (not oxidized) surface can be prepared, although the use of a cryostage and grazing incidence FIB beam may be necessary to get the best quality surface when using Ga+ primary (FIB) ions [3].

Recent results from rechargeable battery materials recorded by the fibTOF and methods for the correct data interpretation will be presented.

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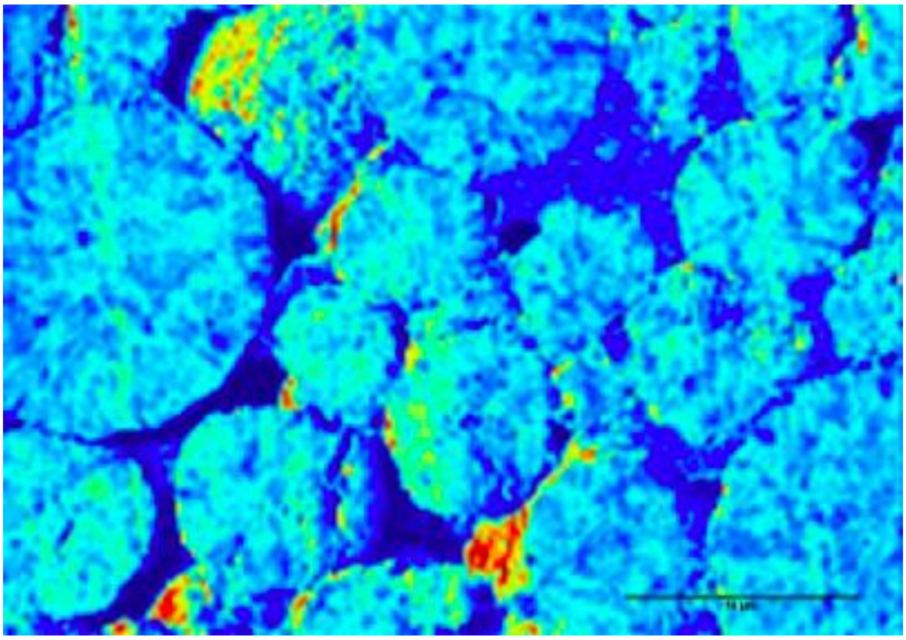

Figure 1: Lithium distribution in a $LiNi_xMn_yCo_{1-x-y}O_2$ (NMC) cathode, from [4].

Positioned generation of luminescence defects in 2D materials by helium ion beams

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Single spin defects in 2D transition-metal dichalcogenides are natural spin-photon interfaces for quantum applications. The creation of such point-defects in bulk crystals lacks either lateral or vertical precision. We overcome this disadvantage by irradiating atomically thin materials by a focused He-ion beam. Here we report on deterministic generation of optically active defects with a helium ion microscope in monolayer MoS_2. In photoluminescence (PL) measurements on the irradiated sites we measure sharp emission lines ~200 meV below the optical bandgap of MoS_2. [1] The He-ion beam dose can be decreased to create single emission lines at each irradiated spot with a yield of ~20%. These single emission lines emit single photons, which could be unambiguously proven in second-order correlation measurements. [2] In high-field magneto-photoluminescence we attribute the emission lines to single sulfur vacancies by combining experiment and ab-initio calculations. In the experiment, we reveal the lifting of spin-degeneracy of the involved defect bands even at zero magnetic field. These results highlight that defects in 2D semiconductors may be utilized for quantum technologies. [3]

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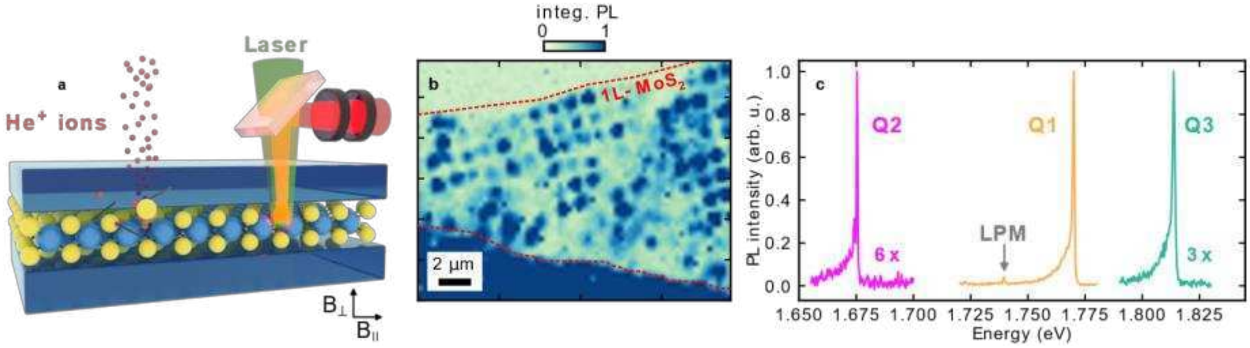

Fig. 1: a) Sketch of the He-ion irradiation and the magneto-optical measurement on the generated defect center. b) False color map of the defect luminescence showing the irradiated array pattern. c) Typical photoluminescence spectra of three different emission bands showing similar asymmetric line shapes.

Fabrication of microstructured devices for grain boundary investigations in unconventional superconductor $CeCoIn_5$

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The study of grain boundaries (GB) in superconductors has both fundamental and applied interests. In high-temperature cuprate superconductors studies of the critical currents (Jc) across GBs have provided important information on the symmetry of the superconducting order parameter and are critical for the observation of spontaneously generated half-flux magnetic quanta [1,2]. Similar to cuprate superconductors, heavy fermion superconductors (HFS) host rich physics in the form of unconventional superconducting phases with nodal quasiparticles. However, there have been relatively few phase-sensitive measurements of the superconducting order parameter thereby emphasizing the need for investigations of J_c across GBs in HFS.

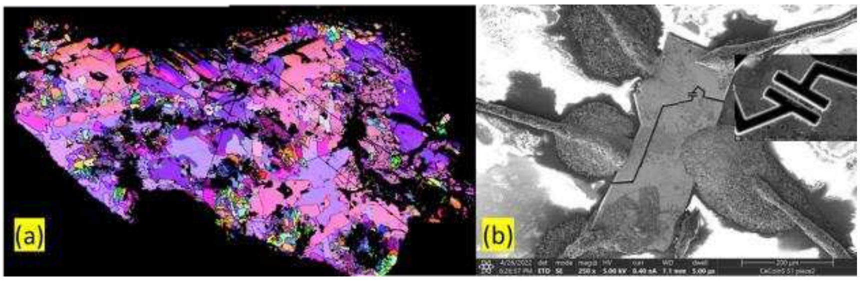

Fig. 1: (a) Shows an EBSD image of the differently oriented grains present in polycrystalline $CeCoIn_5$ sample and (b) shows the SEM image of a microstructed fabricated across a 90° GB using FIB milling.

In this talk, I will present results on GBs in polycrystalline samples of the HFS $CeCoIn_5$. Electron backscatter diffraction (EBSD) performed on well-polished samples of polycrystalline $CeCoIn_5$ reveal that majority of grains are not randomly oriented as one would expect but grow at a misorientation angle of 90° with respect to their neighboring grain. We performed J_c studies across various such GBs by fabricating microstructured devices using focused ion-beam milling. Our investigations are crucial in understanding the superconducting order parameter symmetry of $CeCoIn_5$ and its potential use in devices for quantum information science.

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Exploring Layered Conductors by 3D FIB micro-machining.

Carsten Putzke

Some of the most enigmatic correlated states in the field of quantum matter arise in lower dimensions such as quasi-2d and quasi-1d materials. The layered crystal structure in organic and high-T_c superconductors is an example of these materials, where a quasi-2D electronic structure gives rise to several unconventional electronic instabilities. The layered crystal structure allows for exfoliation in some materials which helped to uncover electrical transport properties such as the quantum Hall effect as well as enables spectroscopic probes in single layer and few layer systems. It is the same crystallographic anisotropy that hinders the study of interlayer electrical transport and spectroscopic probes such as angle resolved photoemission spectroscopy (ARPES).

In my talk I will demonstrate the novel experimental capabilities that focused ion beam micro-structuring enables in studying layered conductors. Confining the in-plane dimension of quasi-2D high purity metals of the Delafossites to length scales smaller than the electron mean free path gives rise to a novel realization of the particle-wave duality[1,2].
Beyond these exciting physical phenomena of finite size confined pillars, novel experimental possibilities will be presented which enable previously inaccessible insight into layered materials.

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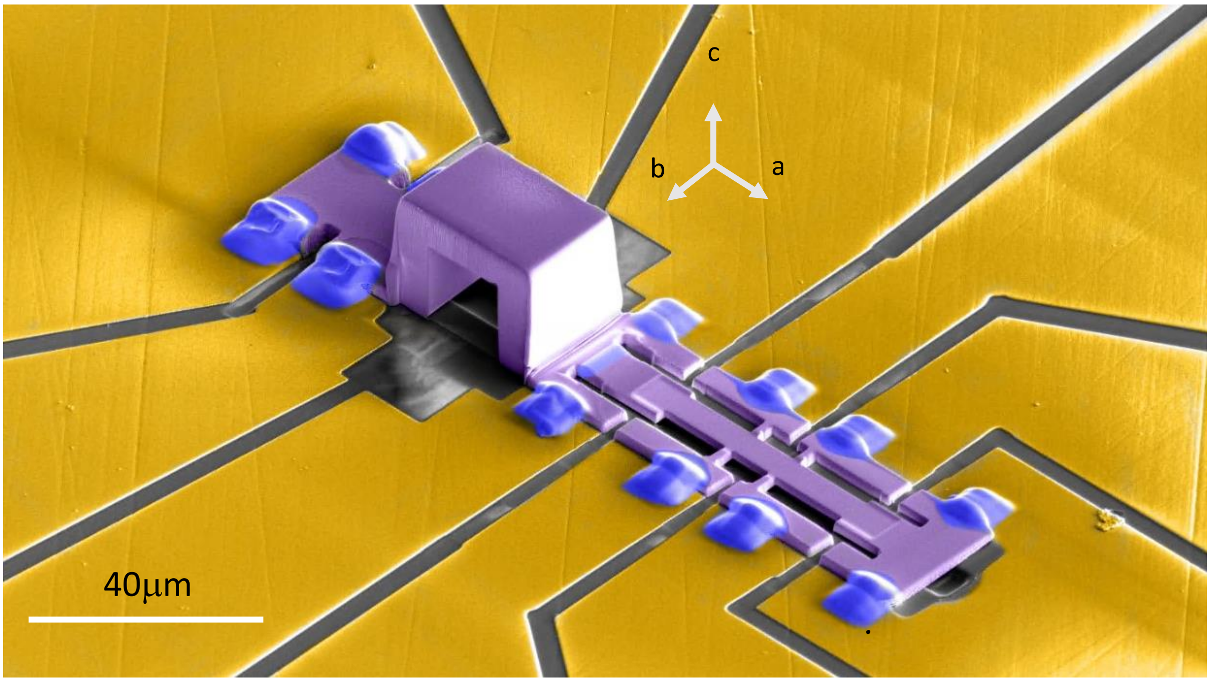

Scanning electron microscope image of a 3D PdCoO2 micro-structure enabling to study the interplay between inter- and intra-layer electrical transport.

Investigation of the Interaction of a Ga^+ Focused Ion Beam with Zirconia by Electron Backscatter Diffraction

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The interaction of a Ga^+ focused ion beam with zirconia materials (Mg-PSZ, Y-PSZ) was investigated with the aim to identify suitable parameters for the sample preparation for surface sensitive analytical methods such EBSD. The FIB parameters glancing angle (angle between ion beam and surface) and applied power (defined by acceleration voltage and ion current) were varied between two levels.

It was observed that the phase stability of monoclinic zirconia depended strongly on the FIB treatment parameters [1-2]. For a glancing angle of 5° and an applied acceleration voltage of 30 kV with an ion current of 30 nA a transformation of originally monoclinic grains to the tetragonal/cubic phase took place (Fig. 1). This was related to the implantation of Ga^+ ions and their presence as Ga_2O_3 which works as stabilizer for the high temperature phases of zirconia [3-4].

Furthermore, the statistical analysis yielded that the influence of the FIB parameters on the surface quality corresponded well to the literature. The data showed that mild parameter levels (lower glancing angle und lower power of the ion beam, such as 5° and acceleration voltage of 5 kV combined with ion current of 4.8 nA, respectively) reduced the surface damage.

[1] H. Berek, C.G. Aneziris; *Effect of focused ion beam sample preparation on the phase composition of zirconia;* Ceramics International 44 (**2018**), 176435.

[2] N. Brachhold, H. Berek, J. Fruhstorfer, C.G. Aneziris; *Focused Ion Beam Parameters for the Preparation of Oxidic Ceramic Materials;* Advanced Engineering Materials 23 (**2021**), 2001235.

[3] A. Surpi, E. Göthelid, T. Kubart, D. Martin, J. Jensen; *Localised modifications of anatase TiO_2 thin films by a Focused Ion Beam;* Nuclear Instruments and Methods in Physics Research, Section B: Beam Interactions with Materials and Atoms 268 (**2010**), 3142.

[4] T. Yamamoto, A. Kurimoto; *Ga Ion-doped ZrO_2 Catalyst Characterized By XRD, XAFS, and 2-butanol decomposition;* Analytical Sciences 36 (**2020**), 41.

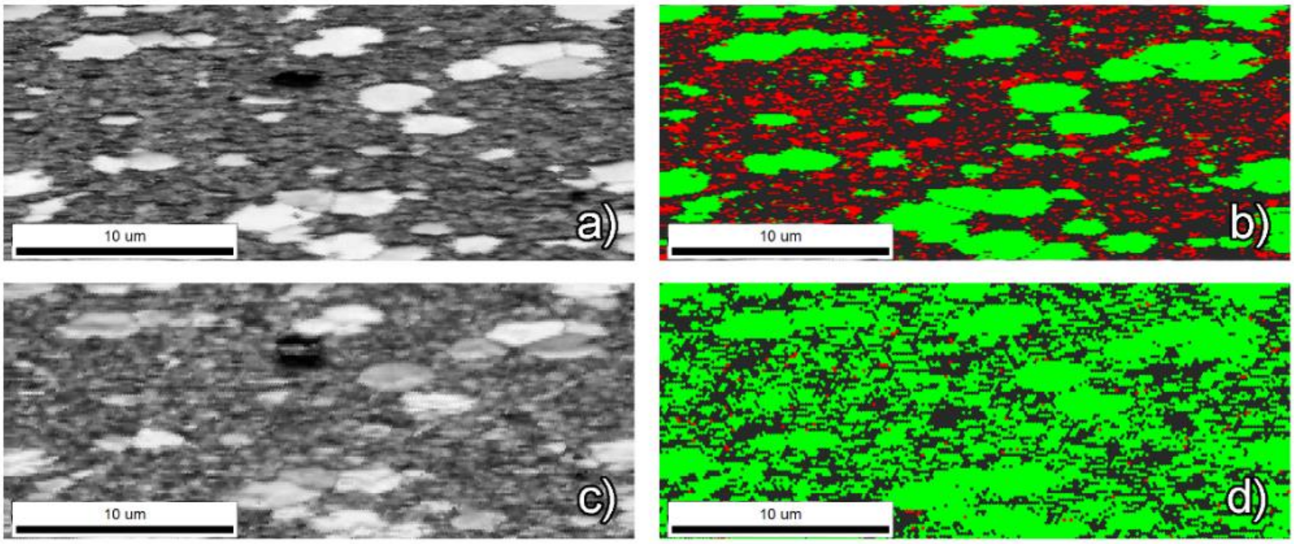

Fig. 1: Image quality (a, c) and phase distribution (b, d – red: monoclinic ZrO_2, green: cubic/tetragonal ZrO_2) of Y-PSZ, (a,b) initial state, (c,d) state after a Ga^+ FIB treatment at 30 kV and 30 nA with a glancing angle of 5°

Poster Contributions

Focused Ion Beam Induced Nanoscale Phase Transitions in Layered Structures

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Specimen preparation for Transmission Electron Microscopy (TEM) using Focused Ion Beam (FIB) is a common method [1]. While it offers many advantages over other methods, like site specific specimen preparation, it also suffers from many disadvantages. Artifacts induced by FIB range from Ion implantation to thermal effects [2,3].

In this work, we investigate thermal effects induced by FIB milling in chalcogenide-based 2D layered Sb_2Te_3 thin films covered by Cu layers. Sb_2Te_3 thin layers are epitaxially grown on p-type Si (111) substrates and polycrystalline samples grown on SiO_2 using pulsed laser deposition [4]. Dependent on beam current used during FIB lamella preparation and Sb_2Te_3 layer thickness, hole formation in the Cu layer, thickness change in the Sb_2Te_3 layer and nanoscale modifications are observed (Fig 1.). The structural changes are confirmed by in situ X-Ray Diffraction heating. The introduction of a separation layer (e.g. Pt) between the Cu and Sb_2Te_3 layers hinders thermal induced structural changes by FIB (Fig 2.). Moreover, Cr - Sb_2Te_3 and a Cu – GeTe layer systems show no modifications during preparation.

[1] T. Ishitani, H. Tsuboi, T. Yaguchi, H. Koike; Transmission electron microscope sample preparation using a focused ion beam; Microscopy (1994), 322.

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[4] H. Bryja, J.W. Gerlach, A. Prager, M. Ehrhardt, B. Rauschenbach, A. Lotnyk; Epitaxial layered Sb2Te3 thin films for memory and neuromorphic applications; 2D Materials (2021), 045027.

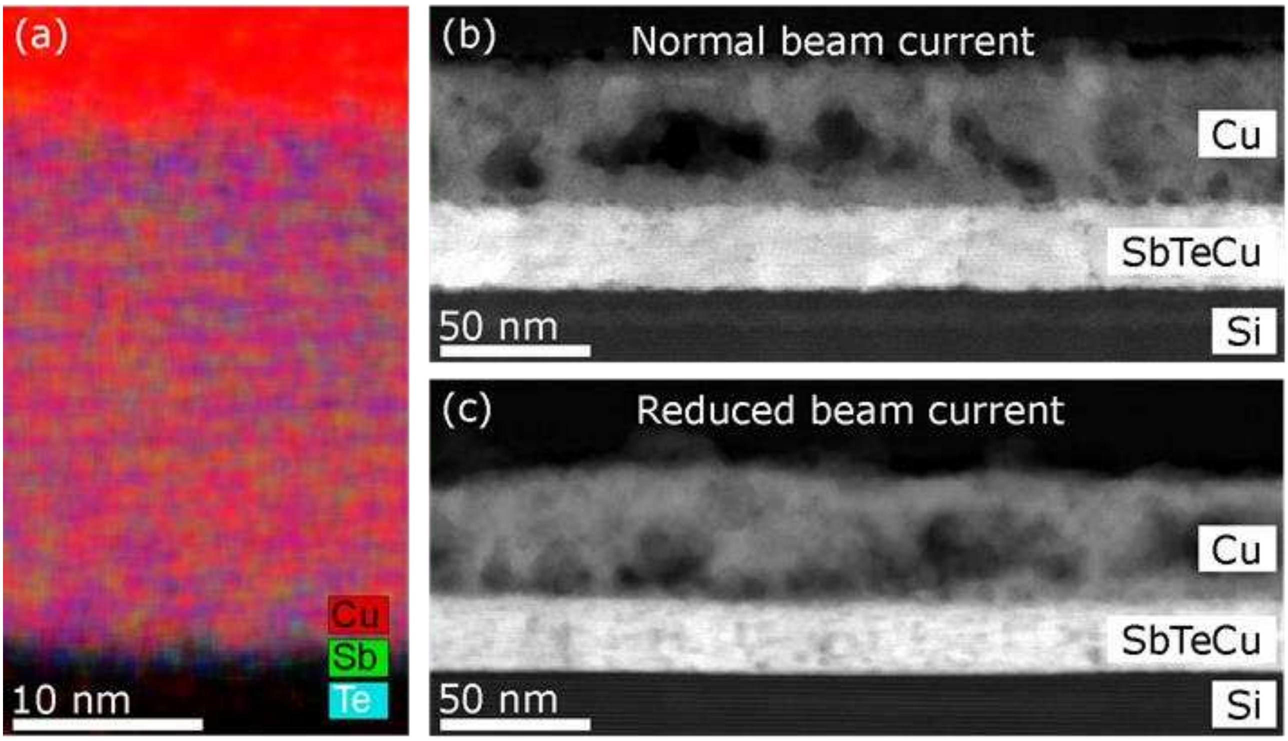

Fig. 1: (a) EDX Map of a lamella prepared with FIB, (b-c) Overview HAADF-STEM images of specimen prepared with normal and reduced FIB beam currents.

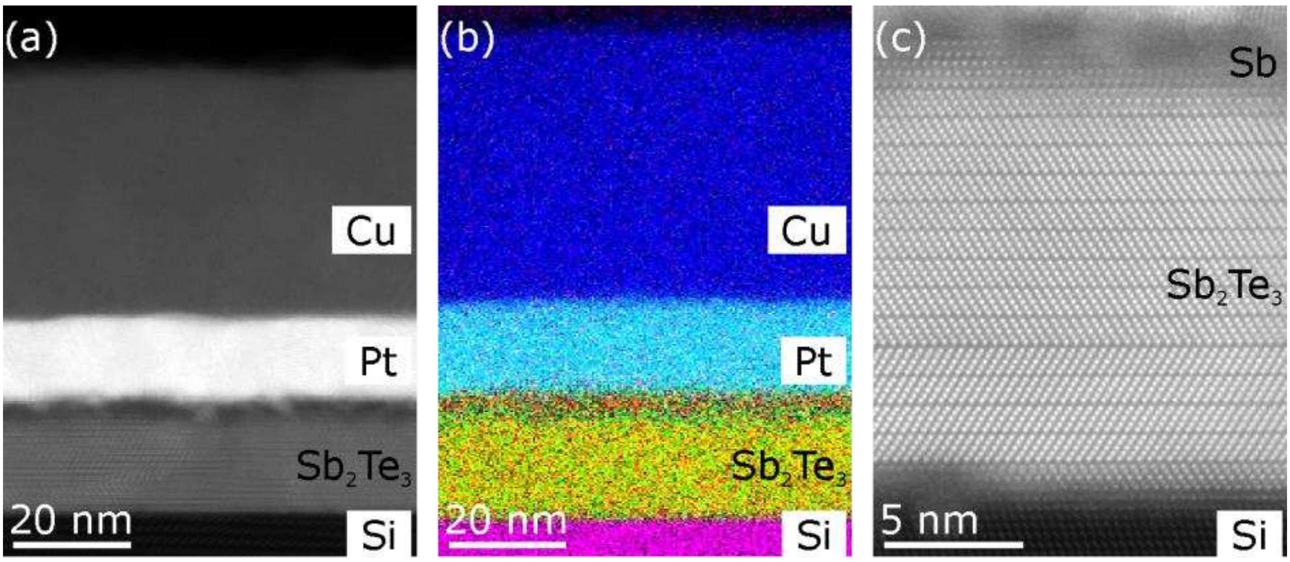

Fig. 2: Cu/Pt/Sb2Te3 layer stack. (a) Overview HAADF-STEM image. (b) Overview EDX elemental map. (c) Atomic-resolution HAADF-STEM image, showing initial Sb_2Te_3 structure and no redeposition of Cu.

The Manufacture of van der Waals Heterostructures Using He Ion Beam Patterning

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Polaritons in two-dimensional materials exhibit enhanced light-matter interactions, which makes them interesting for low-loss, highly confined light transport. A polariton is a quasiparticle that combines a photon with a dipole-carrying excitation in matter and is strongly dependent on the type and geometry of the material. The hybridization of polaritonic modes in different 2D materials may provide strong localization of plasmonic excitations with long propagation distances of phonon modes [1]. By modifying the geometry of van der Waals (vdW) heterostructures at the nanoscale, we tune hybrid polaritonic modes.

We fabricate and patterning of heterostructures based on single crystalline gold or silver flakes, graphene, and hexagonal boron nitride (hBN). For dry transfer we used polydimethylsiloxane (PDMS) and poly(propylene) carbonate (PPC) films due to their strong adhesion to 2D materials at room temperature. Therewith, single-layer to few-layer 2D materials were successfully transferred onto thin electron transparent membranes of silicon nitride.

To modify the geometry of the heterostructures at the nanoscale, a Zeiss Orion Nanofab microscope is then used for patterning by He and/or Ne ion beam milling (cf. fig. 1). As polaritonic modes are not only strongly influenced by geometry, but also by material quality, an important step in the study is therefore to investigate different currents, acceleration voltages, and ion types to determine what damaging effects they have on the crystalline lattice and the corresponding m material response. The optimization of the patterning routines is carried out with the help of FIB-o-Mat, which provides complete control over the beam path [2].

In the following step, monochromated, low-loss scanning transmission electron microscopy (STEM), electron energy-loss spectroscopy (EELS) [3] is used to map the optical properties of the fabricated heterostructures, and the results are compared to near-field optical methods.

[1] A. Woessner, M. B. Lundeberg, Y. Gao, A. Principi, P. Alonso-gonzález, M. Carrega, K. Watanabe, T. Taniguchi, G. Vignale, M. Polini, J. Hone, R. Hillenbrand, and F. H. L. Koppens; Nature Materials 14 (**2015**), 421.

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[3] O. L. Krivanek, N. Dellby, J. A. Hachtel, J. Idrobo, M. T. Hotz, N. J. Bacon, A. L. Bleloch, G. J. Corbin, M. V. Hoffman, C. E. Meyer, T. C. Lovejoy; Progress in ultrahigh energy resolution EELS; Ultramicroscopy Volume 203 (**2019**), 60.

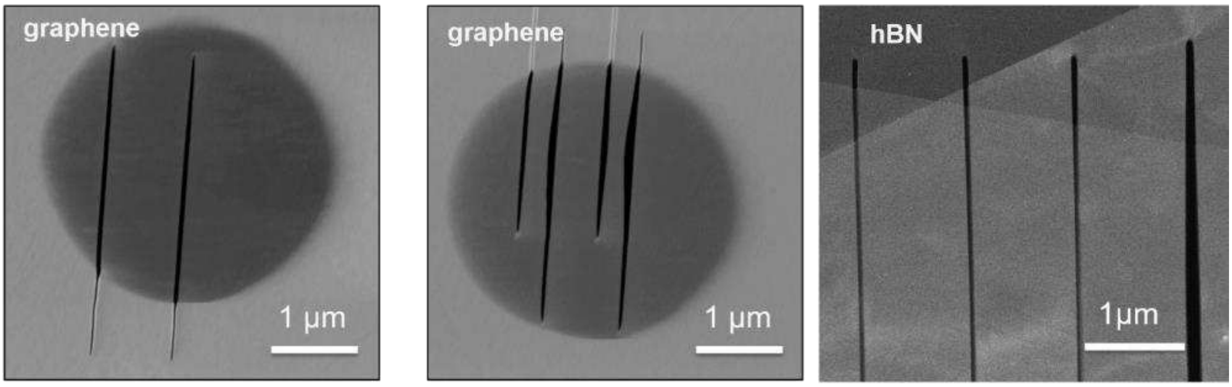

Fig. 1: SEM images of an example graphene flake and hBN flake after He ion beam patterning with different doses.

High throughput tips manufacturing for active piezocantilevers with xenon ion beam with mass control

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Microcantilevers equipped with orthogonal tips are basic tool in many scanning probe microscopy (SPM) techniques, especially in atomic force microscopy (AFM). Tips are medium for transferring interactions between surface and appropriate transducers, their durability determines life span of a tool. However, it is possible to preserve the tool by regeneration of the tip. While original tips are manufactured with use of mass production methods, regenerated tips require unitary approach. Therefore throughput of regeneration method becomes valid parameter.

For regeneration of tips focused ion beam (FIB) family of techniques can be used. It allows both for removing (milling) and building (depositing) the material. With use of nanomanipulators auxiliary materials can be applied [1]. Manufacturing the tip requires time, as required amount of material has got to be moved. To increase the throughput of process inert gas plasma beam can be utilized. Plasma beam offers greater currents than liquid metal sources or gridless ion sources. Furthermore, use of heavy element such as xenon greatly improves processing speed, as Xe ions have higher sputtering yield (2.3 atom/ion on silicon on normal incidence) than e.g gallium (2.05) and also lower penetration depth (mean 11,1 nm versus 12.2 nm for gallium) [2].

To get a hold on rapid process, external measuring circuit is applied. Active cantilever is actuated at the resonant frequency and it can be viewed as a simple harmonic oscillator (SHO). Material deposition delivers mass to the SHO, therefore reducing its resonant frequency; material milling during process raises resonance. Vibrations are observed with help of amplifying circuit on the oscilloscope, where in situ assessment of process' result is possible with measurements of mass

In this setup conductive diamond tips were delivered onto active piezoresistive cantilevers. We present used setup with results in form of sharpened tips.

[1] E. Gacka, P. Kunicki, A. Sikora, R. Bogdanowicz, M. Ficek, T. Gotszalk, I. Rangelow, K. Kwoka; *Focused ion beam-based microfabrication of boron-doped diamond single-crystal tip cantilevers for electrical and mechanical scanning probe microscopy*; Measurement (**2022**), 1

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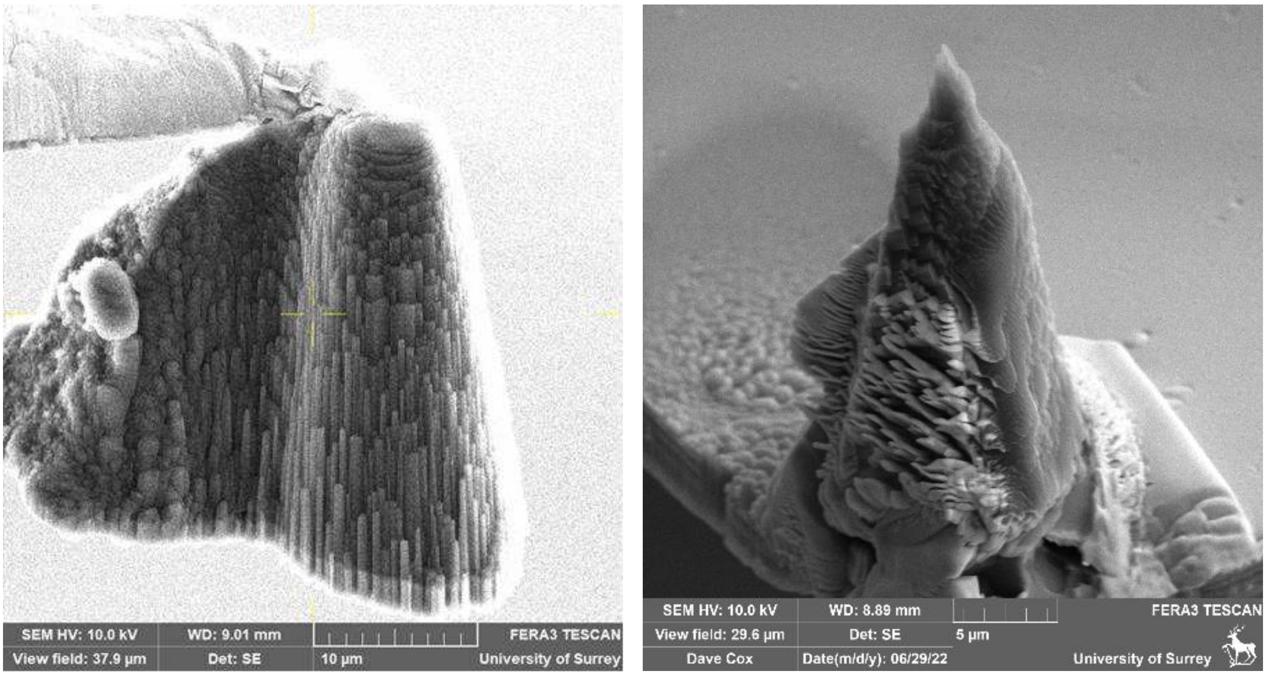

Fig. 1: Diamond tip before mounting, carried with nanomanipulator (left) then mounted and sharpened (right)

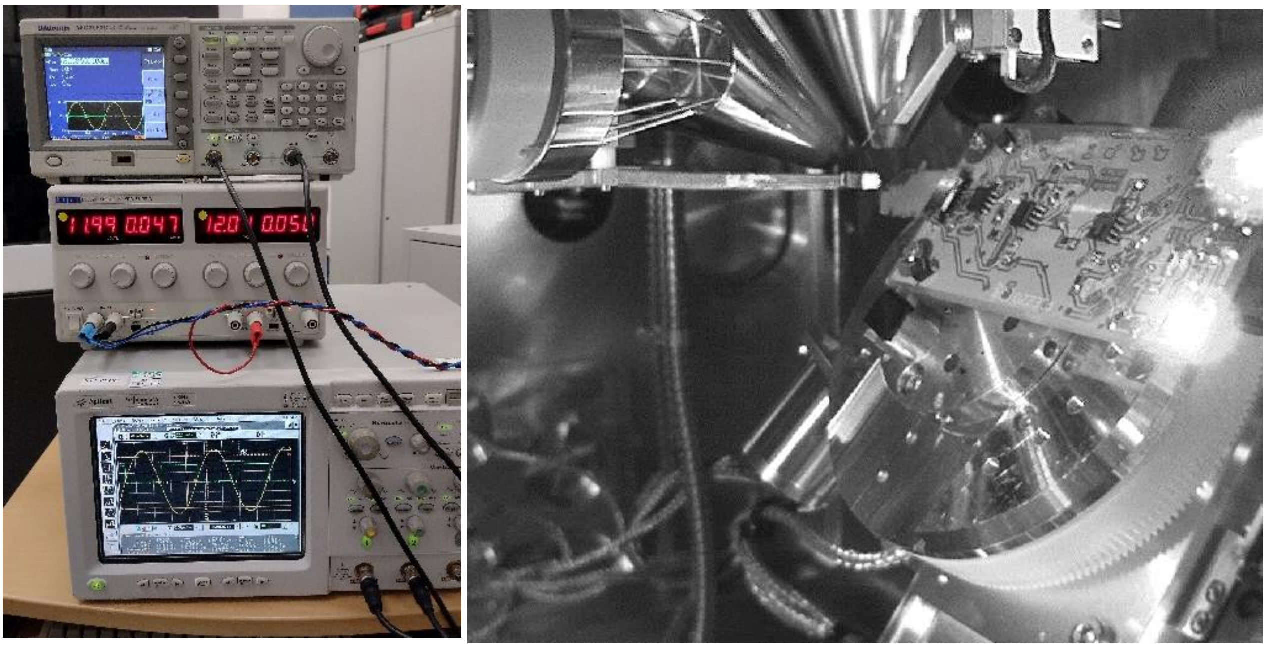

Fig. 2: Tip placement control setup consisting of: power source, function generator, oscilloscope outside chamber (left) and circuit board of amplifiers inside the chamber (right)

Crystalline anisotropic curtaining effect in Bismuth

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Elemental bismuth is a semi-metallic material with inherently strong spin-orbit coupling and high carrier mobility. These unique electronic properties makes it highly important for both fundamental researches and future applications[1]. Motivated by the recent development on higher-order topological insulator phase in Bismuth[2], it is desirably needed to explore the possibility of artificially writing topological conduction channel using focused-ion beam (FIB) technique. However, due to its low crystallization temperature, the local heating by ion beam assists the growth of Bismuth nanowires along the beam direction and therefore leads to strong curtaining effect on the polished surface, which is a major challenge for microstructure fabrication. On the other hand, we found that this curtaining effect is highly anisotropic due to the preferential growth of bismuth nanowires along different crystalline directions. By changing the ion beam direction from trigonal to bisectrix direction, the curtaining effect is strongly suppressed and therefore a clean lamella can be obtained. This sets the basis for the future development of FIB-printed topological microcircuits. Moreover, the nanowire growth along bisectrix direction sheds the light on an interesting method to develop integrated bismuth nanowire array, the pattern of which can be exactly controlled by the ion beam condition.

[1] Asish K. Kundu et al., *Quantum size effects, multiple Dirac cones and edge states in ultrathin Bi(110) films*; arXiv: 2106.13943 **(2021)**.

[2] Frank Schindler, Zhijun Wang et al., *Higher-Order Topology in Bismuth*; Nature Physics 14 **(2018)**, 918.

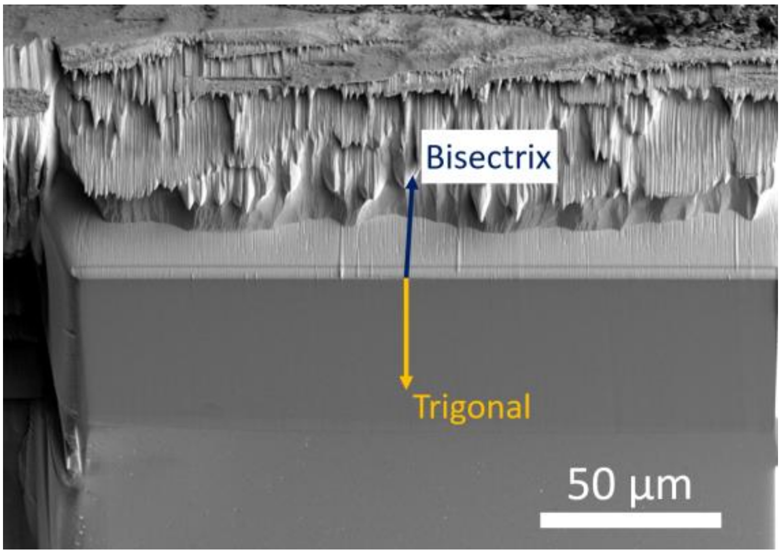

Fig. 1: Crystalline anisotropic curtaining effect in Bismuth.

Micromachined samples for uniaxial strain studies with laser-ARPES

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Uniaxial strain is an important tuning parameter in condensed matter physics, as modest pressures can induce fundamentally different characteristics in materials reversibly and reproducibly [1-3]. However, it has long been a technical challenge to systematically study the effects of uniaxial strain in experiments such as angle resolved photoemission spectroscopy (ARPES) [4-6].

We introduce a novel and generally applicable route to studying uniaxial strain by macroscopically shaping a sample with a Helios G4 PFIB such that it gains a tapered profile. This profile allows us to induce a variation in strain within the sample by applying pressure with a thermally actuated pressure cell. The induced strain gradient can then be resolved in experiments with a spatially local probe, unlocking the potential for systematic studies of the effect of uniaxial strain. We present micro-focused laser-ARPES results for Sr_2RuO_4 – a keystone material in condensed matter physics – under uniaxial strain to study the evolution of a van Hove singularity across the chemical potential.

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[2] H. Kim *et al.; Uniaxial pressure control of competing orders in a high-temperature superconductor;* Science **362** *6418* (2018), 1040.

[3] A. Steppke *et al.; Strong peak in Tc of Sr2RuO4 under uniaxial pressure;* Science **355** *6321* (2017) eaaf9398.

[4] J. A. Sobata, Y. He, Z. X. Shen; *Angle-resolved Photoemission studies of Quantum Materials;* Reviews of Modern Physics **93** (2021), 025006.

[5] S. Ricco *et al.; In situ strain tuning of the metal-insulator-transition of Ca2RuO4 in angle-resolved photoemission experiment;* Nature Communications **9** (2018), 4535.

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In-situ sample preparation of oxidizing and contaminating samples for high quality EDS and WDS quantification using FIB-SEM

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Energy dispersive and wave dispersive X-ray spectroscopy (EDS and WDS) are very important tools in materials research to obtain information about the chemical composition of a sample. Many materials like thermoelectrics show a significant loss of efficiency with elemental compositions slightly differing by a few percent. In quantitative measurements conductivity, contamination and, above all, homogeneity and the geometry of the sample have a significant influence on the quantification results with EDS or WDS systems. Especially for WDS the quality of the sample surface is very important to achieve an accuracy below 1%. Many materials are susceptible to oxidation (Fig. 1, right) or carbon contamination on their surface decreasing the accuracy and quality of WDS and EDS measurements. Furthermore, this prevents the quantification of initial oxygen or carbon contents.

The use of FIB allows the preparation of smooth cross-sections with very good smoothness and planarity (Fig.1, left). With an EDS/WDS system installed at a FIB-SEM, an in-situ, site specific preparation followed by analysis without leaving high vacuum is feasible. This enables a contamination- and oxidation-free preparation and analysis, significantly improving the quality of the measurement.

Cross-sections prepared with the FIB are usually not perpendicular to the electron beam, which is a pre-requisite for a proper quantitative EDS or WDS analysis. In this study, a new procedure is presented allowing an accurate quantification with EDS and WDS by using different tilt angles for the preparation and analysis step.

Initially, the cross-section of a thermoelectric sample is prepared at a tilt of -10° producing a flat angle between sample and Ion beam on a system including an angle of 52° between electron and ion column. By tilting the sample to +28° afterwards, the cross-section is orientated perpendicular to the electron-beam enabling a quantification with EDS and WDS (Fig: 2). The results sum up to a total weight of 100.23% indicating a reliable combined EDS/WDS measurement (Tab. 1, left). For the sample surface produced via metallographic state-of-the-art procedures, which is oxidized due to atmospheric exposure, a weight of 92.23% is measured indicating a non accurate measurement caused by the high oxygen contamination layer on the surface (Tab. 2, right). This comparison indicates that this procedure significantly improves the quality of EDS and WDS quantitative compositional results for fast oxidizing and easy contaminating samples.

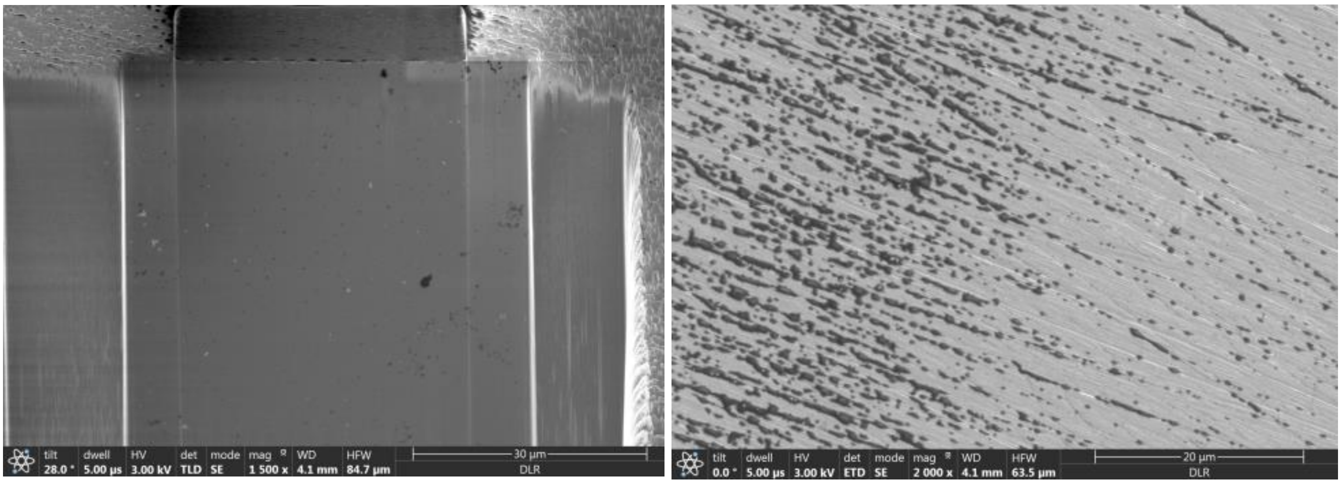

Fig. 2: Left: Surface of FIB prepared Cross-Section in a flat angle. Right: Oxidized surface of the sample

	FIB Prepared		Metallographic Preparation	
Element	Weight [%]	Atom [%]	Weight [%]	Atom [%]
Mg+	8.82	31.28	8.74	33.14
Ag	40.74	32.65	37.53	32.06
Sb	50.67	35.97	45.96	34.79
Total	100.23	100.00	92.23	100.00

Tab. 1: Left: Quantification with WDS of MgAgSb in a FIB prepared cross-section with corrected tilt. Right: Quantification with WDS of the same sample, but on its surface prepared by conventional metallography procedures. Mg was measured by WDS, AG and Sb by EDS.

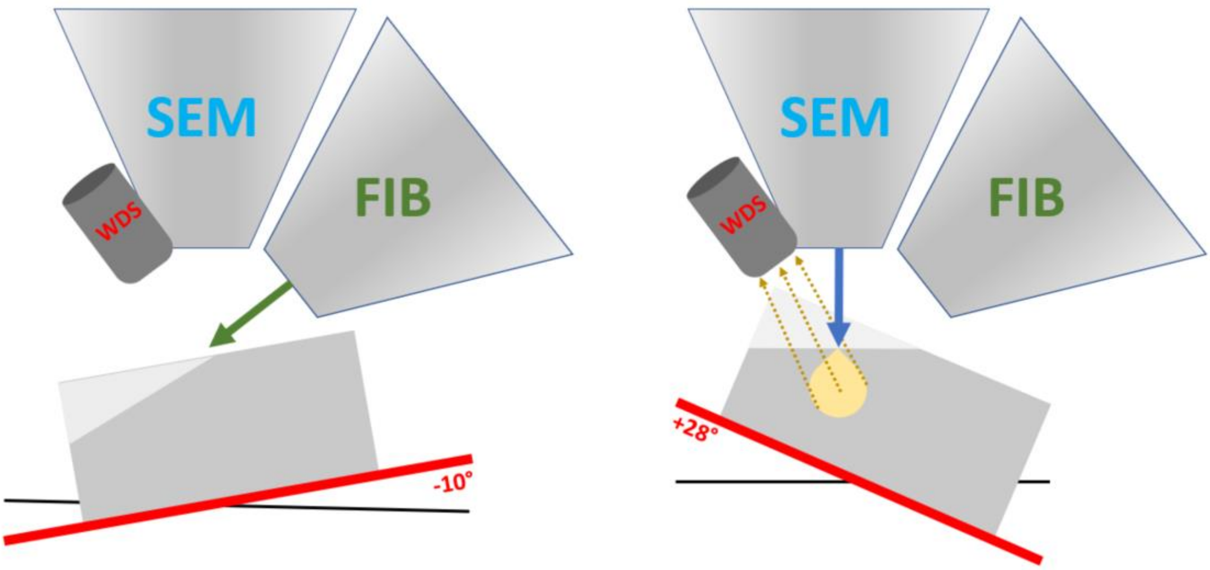

Fig. 2: Left: Cross-Section with -10° stage tilt resulting in a -28° tilt compared to the sample surface; Right: Stage tilt of +28° to achieve the geometry necessary for WDS quantification.

Dual focused ion beam nanofabrication of V-grooves in monocrystalline gold for efficient excitation of organic single photon emitters

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Single dibenzoterrylene (DBT) molecules in anthracene nanocrystals have been shown to emit single photons without blinking, bleaching or spectral diffusion. Efficient integration of these molecules into photonic circuits will pave the way toward the realization of quantum optical networks. We incorporated anthracene nanocrystals containing single DBT molecules into plasmonic V-grooves milled with a focused ion beam in monocrystalline gold flakes. The fabricated V-grooves exhibit high-quality low-loss guiding of channel plasmon polaritons with the propagation length reaching ~14 µm at a wavelength of 800 nm. For DBT molecules coupled to the V-grooves, we observe enhanced emission decay rates with up to 50% of the emission being funneled into channel plasmon polaritons [1].

In particular we utilized a two-step focused ion beam lithography process to fabricate V-grooves with defined dimensions, smooth sidewalls and inclined end-faces on monocrystalline gold flakes. The subsequent application of gallium and helium focused ion beams (GaFIB/HeFIB) provided by a commercial instrument (Zeiss Orion Nanofab) allowed us to fabricate waveguides of the required size and shape at reasonable fabrication speed.

[1] Kumar, S.; Leissner, T.; Boroviks, S.; Andersen, S. K. H.; Fiutowski, J.; Rubahn, H. G.; Mortensen, N. A.; Bozhevolnyi, S. I. Efficient Coupling of Single Organic Molecules to Channel Plasmon Polaritons Supported by V-Grooves in Monocrystalline Gold. Acs Photonics 2020, 7 (8), 2211-2218.

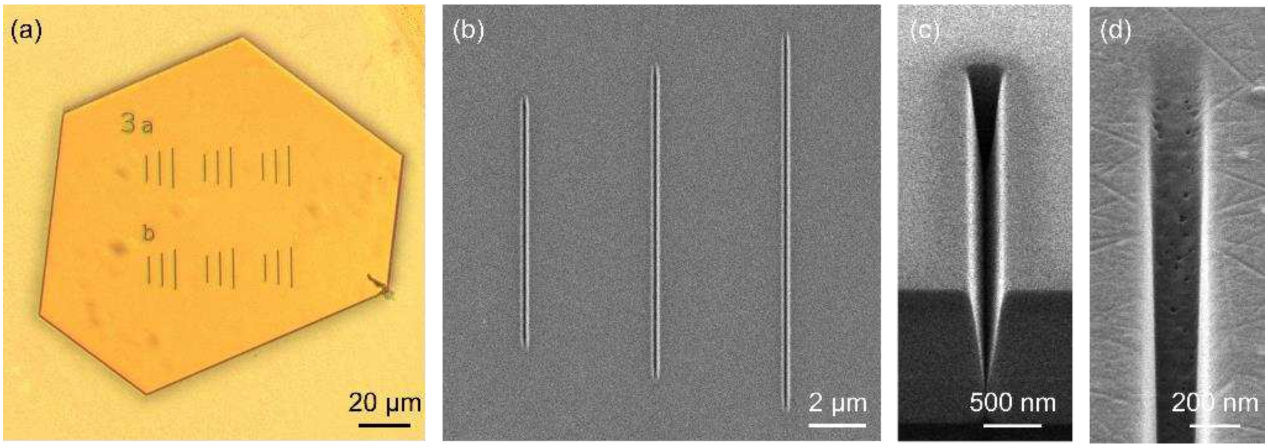

Fig. 1: (a) Microscope image of a gold flake containing V-grooves of different sizes. (b) HIM image of V-grooves. (c) HIM image of a cross-section of a V-groove together with a nano mirror. (d) HIM image of the nano-mirror at one of V-groove ends.

Novel FIB nanofabrication strategies facilitated by light and heavy ions from GaBiLi Liquid Metal Alloy Ion Sources

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Focused Ion Beam (FIB) direct nano-patterning has become established as versatile, and precise fabrication method of smallest features at the nanoscale [1]. Applications in nanoscale science are manifold and require high resolution fabrication techniques at high fidelity, accuracy, and reproducibility. As a result, high demands concerning sputter yield, beam stability and patterning resolution are made on the ion beam for direct FIB nano-patterning. Liquid Metal Alloy Ion Source (LMAIS) is an emerging FIB source technology that provides a versatile solution to deliver light or heavy and fast or slow ions from a single source for FIB nanofabrication [2]. We present unique direct nano-patterning results and a novel workflow using GaBiLi LMAIS. This workflow allows taking advantage of the benefits of the different beams. Gallium, Bismuth and Lithium ions are emitted simultaneously with subsequent ion separation in an ExB filter.
A Lithium-ion beam has the smallest beam diameter and enables highest patterning resolution as well as imaging resolution of all ions available from LMAIS[3] whereas Bismuth ion beam provides higher sputter yield at higher depth resolution [4].
To overcome challenges during patterning of sub-10nm metallic nanogaps, we present a 2-step fabrication process for bowtie nano-antennas. This approach takes advantage of large volume material milling with a Bismuth ion beam at high sputter yield to speed up the entire fabrication process and subsequent lateral fine shaping using a Lithium beam from the same ion source.
Beyond direct nanopatterning, Lithium as the lightest ion available from LMAIS provides excellent ion beam imaging capabilities. Latest results of 3D milling and ion imaging with Bismuth and Lithium ions for 3D sample tomography and reconstruction will be presented.

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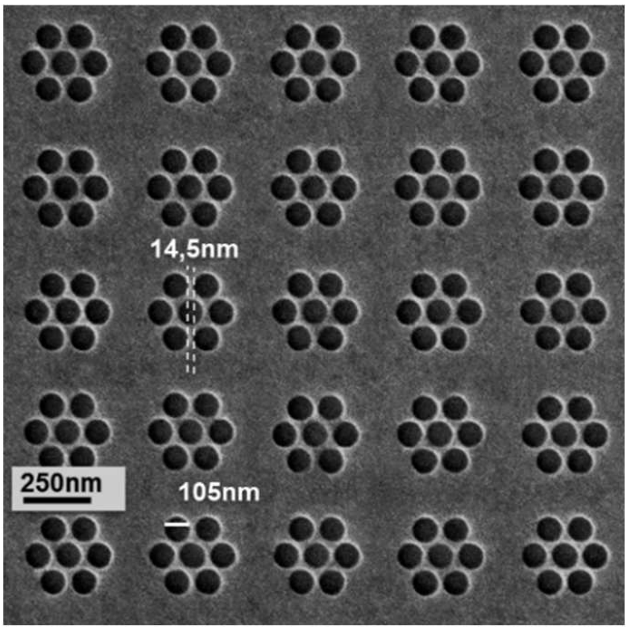

Fig. 1: *Li FIB patterning of heptamer-arranged nanohole (HNH) arrays in 50nm Au film on SiO_2 and subsequent Li ion beam imaging.*

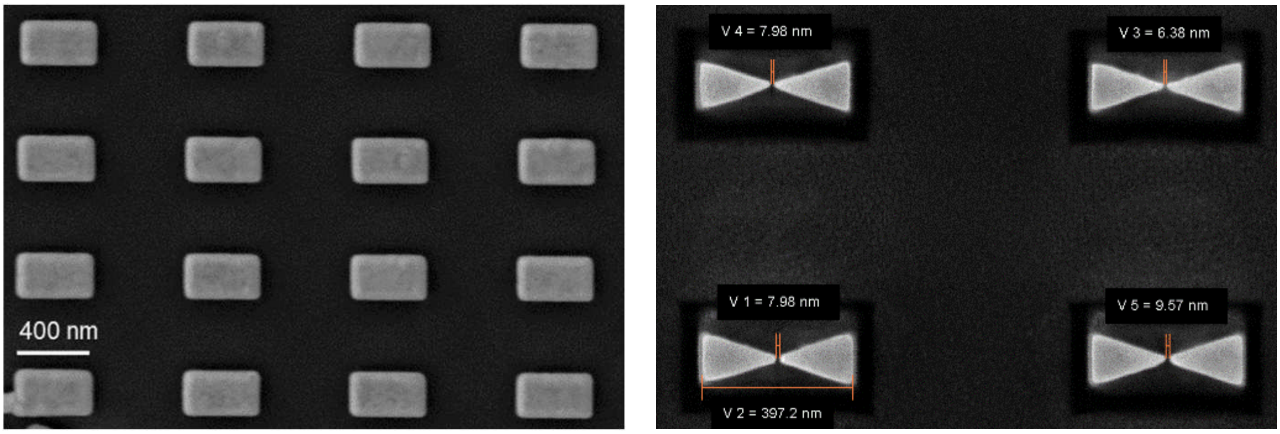

Fig. 2: *Stepwise fabrication of nano bowties in 50nm Au film on Si: a) Bi FIB for large volume milling to excavate rectangular boxes, b) Li FIB for fine shaping bowties structures at highest resolution.*

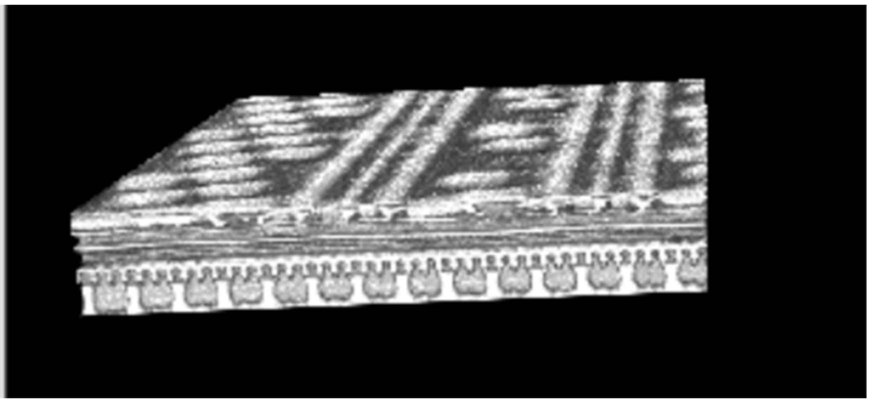

Fig. 3: *3D reconstruction (5µm x 5µm x 0.85µm) of semiconductor chip layers by Bi milling and Li imaging.*

Effect of Focused Helium-ion Beam on Surface Morphology of Polypropylene Thin-films for Power Capacitors

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Polypropylene (PP) films are one of the widely used dielectric materials for power capacitors in high-frequency and high-power applications due to their excellent dielectric strength, ease of processability, reliability, and low dissipation factor [1]. The rapid development of high-performance capacitors requires new methods of modification and advanced characterization of the PP films at the nanoscale level to enable their further miniaturization and to improve their performance. In this work, we investigated the effect of ion-beam irradiation on the surface morphology of PP films. Commercially available and deposited with spin-coating PP films were irradiated with a focused He^{+}-ion beam (He-FIB) in a Zeiss Orion NanoFab Helium Ion Microscope at landing energy of 25 keV with doses in a range of 5.4×10^{-5} nC/μm^2 to 8.07×10^{-3} nC/μm^2. Prior to irradiation, all samples were metalized with a very thin layer of a metal alloy. He-FIB was used to construct surface patterns similar to those fabricated in our previous studies [2,3] of polymer materials irradiated with FIBs. Atomic force microscopy (AFM) and optical microscopy were used to analyze the details of surface modification. The obtained results show that the irradiation of the PP films with He-FIB results in shrinkage of the polymer material and negligible surface sputtering effects. Surface ripples were observed at the borders between the irradiated and non-irradiated regions, which is attributed to the mechanical strain induced by the material modification. Among other things, our study shows that, alongside the known modification of dielectric and electric properties of PP material, the changes in the film shape in the irradiated regions should be taken into account as a geometrical factor that affects capacitance.

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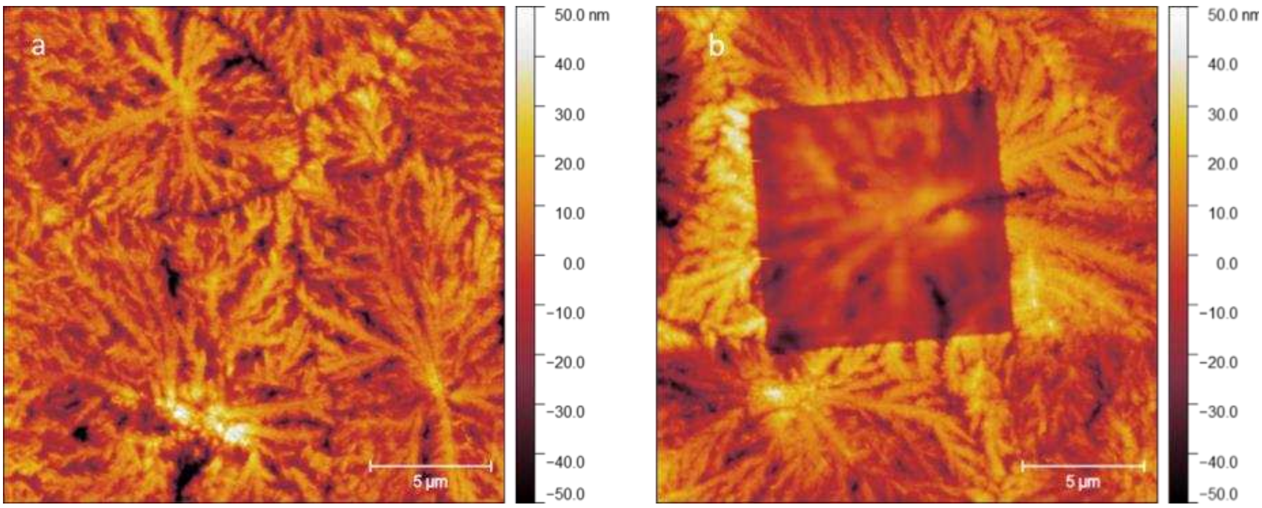

Figure 1: AFM images of the surface of spin-coated PP film, before (a) and after (b) FIB-He irradiation. The irradiated region is the dark square in b).

Fault Localization in FIB/SEM – combining delayering and EBAC into a cohesive workflow

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Fault localization and fault isolation are important, recurring tasks in many Fault Analysis laboratories. The workflow involves various techniques for deprocessing and delayering the sample including CNC milling, various polishing methods, and lately, with the advent of smaller and smaller technology nodes, PFIB delayering. The latter method yields precise control over the number of layers removed while allowing the operator to open large windows into the sample.

Once the target layer is exposed, electrical testing methods such as EBIC (Electron Beam Induced Current) or EBAC (Electron Beam Absorbed Current) as well as nanoprobing experiments for transistor characterization can be performed. Depending on the obtained results, further delayering may be required. Or, if the fault is located, a TEM slice may be prepared in order to further elucidate the fault's root cause.

Thus, it is advantageous to combine the electrical characterization measurement and the PFIB delayering into a single tool – thereby circumventing the need to move the sample back and forth between tools. This minimizes the sample's exposure to air (which minimizes contamination on the sample) and significantly speeds of the time to result. The images in Fig. 1 and Fig. 2 show the proposed setup as well as a micrograph of a typical sample with probes in place for EBAC analysis.

In this work, the proposed tool configuration for performing the combined workflow described above will be presented.

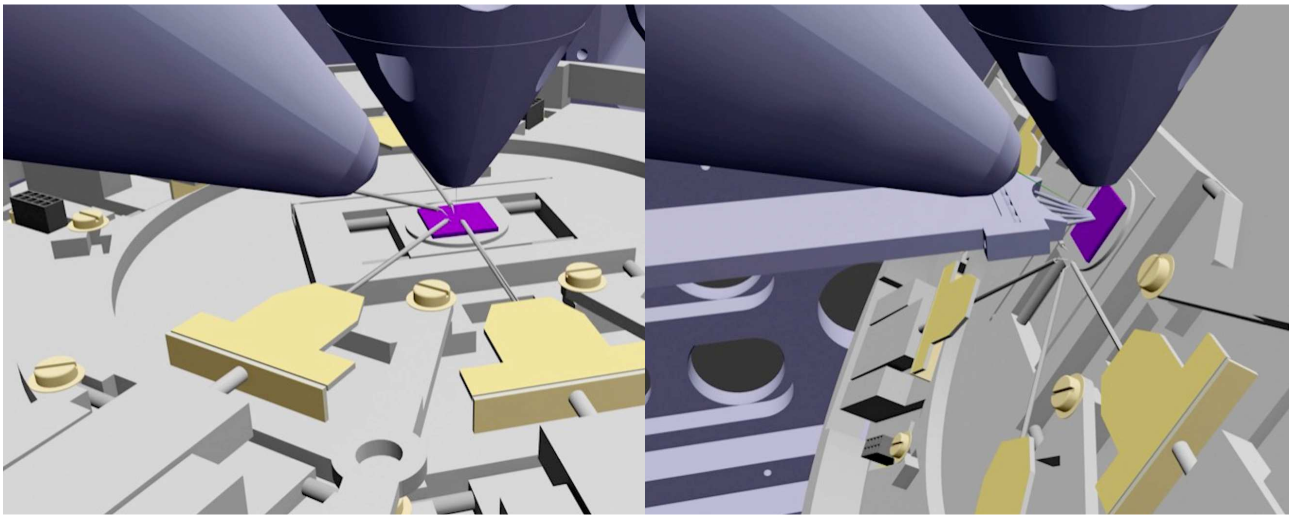

Fig. 1: EBAC configuration (left) and delayering configuration (right) with the stage tilted to FIB angle and the GIS inserted. Note that the sample was shifted using the integrated substage such that the distance between the probe tips and the FIB is maximized in order prevent contamination of the probe tips.

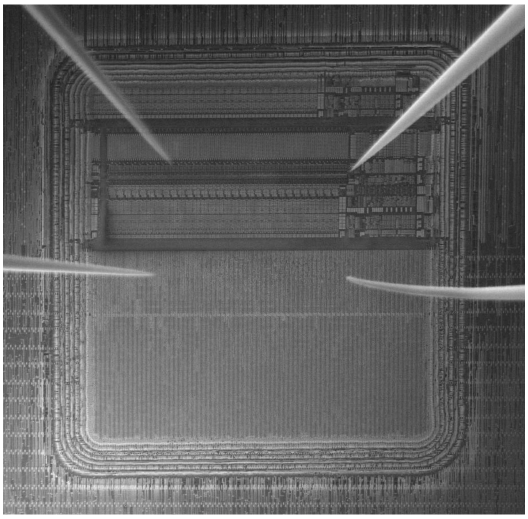

Fig. 2: Four probe tips placed in close vicinity to the sample in preparation for an EBAC analysis (courtesy AMD Singapore)

Second order Zeeman interaction and ferroquadrupolar order in $TmVO_4$

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$TmVO_4$ exhibits ferroquadrupolar order of the Tm 4f electronic orbitals at low temperatures, and is a model system for Ising nematicity. A magnetic field oriented along the *c*-axis constitutes a transverse effective field for the quadrupolar order parameter, continuously tuning the system to a quantum phase transition as the field is increased from zero. In contrast, in-plane magnetic fields couple to the order parameter only at second order, such that orienting along the primary axes of the quadrupole order results in an effective longitudinal field, whereas orienting at 45 degrees results in a second effective transverse field. Nuclear magnetic Resonance (NMR) studies of these effects are hampered by increased linewidth of the spectra due to inhomogeneous demagnetizing fields but can be minimized by cutting the sample to an ellipsoidal shape using a Xe plasma FIB.

Previous NMR measurements of $TmVO_4$ identified a scaling between the spin–lattice relaxation rate and the shear elastic stiffness constant, c_{66}, suggesting that the ^{51}V ($I = 7/2$) nuclear spins couple to the Tm orbitals through the electric field gradient (EFG), giving rise to a quadrupolar relaxation channel [1]. However, the spectra were significantly broadened by inhomogeneous demagnetization fields and the anisotropic *g*-factor of the Tm ground state doublet. In order to better discern the spectra and relaxation mechanisms at play, we reshaped a single crystal of $TmVO_4$ to an ellipsoidal shape, with a homogeneous demagnetization field (see Fig. 1). We utilized a Xe^{2+} plasma focused ion beam (FIB) by Thermo Fisher Scientific with a 30 kV, 1 µA beam to cut our sample with the long-axis along the *c*-axis of the crystal. Sample damage from the beam is only expected on the surface within a depth of 30–40 nm and energy dispersive X-ray analysis (EDX) of a test surface verifies the unchanged composition of $TmVO_4$ below. The final sample diameter is 0.4 mm and the length of 1.3 mm require a total cutting time in excess of 25 h of each side. The magnetic broadening was dramatically reduced in the FIB crystal, such that each of the seven peaks separated by the quadrupolar splitting are clearly resolved. The ability to resolve all seven peaks is important because it enables us to extract details of the magnetic and quadrupolar contributions to the spin–lattice relaxation rate that would otherwise be inaccessible, as discussed above in the section on Spin-lattice relation [2].

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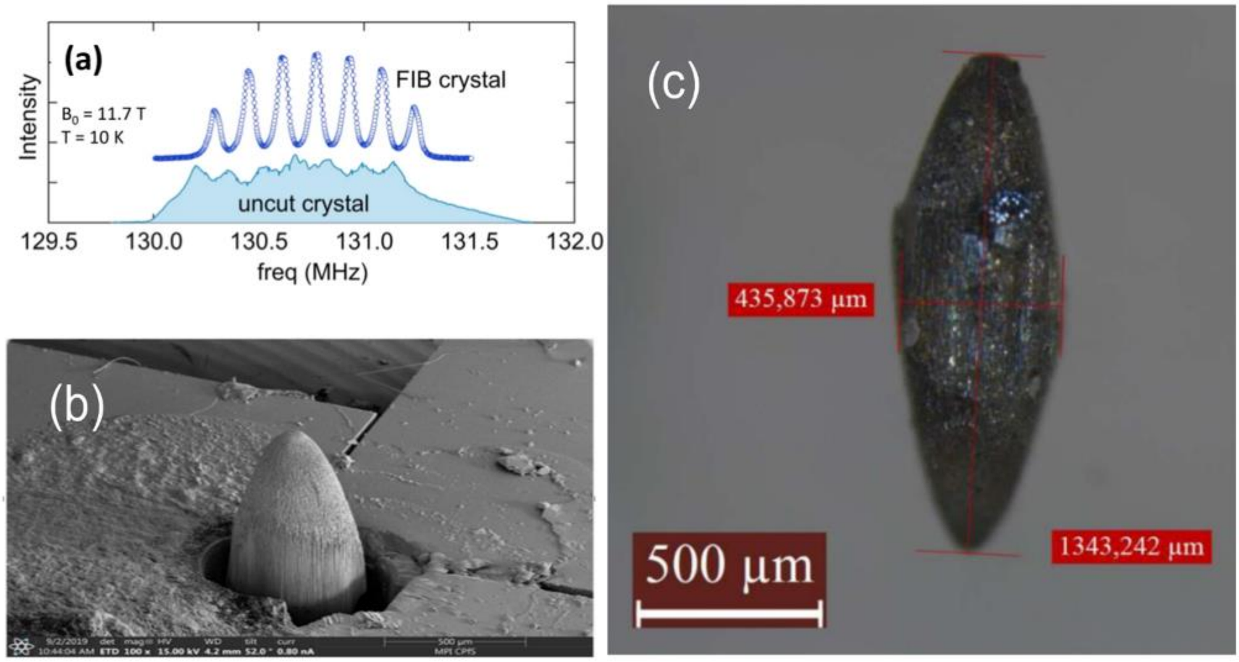

Fig. 1: (a) ^{51}V-NMR spectrum with and without ellipsoidal crystal shape. (b) Scanning electron microscopy (SEM) scan of the sample during the FIB process. (c) Crystal after FIB. Al and C are deposited on the sample surface layer during the FIB processing, but do not contribute to the NMR signal.

Using Electron Beam Curing (EBC) for the Controlled Bending of 3D-Nanoprinted FEBID Structures

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Additive manufacturing via Focused Electron Beam Induced Deposition (FEBID) is an increasingly relevant technique for depositing high-fidelity architectures on the nanoscale. While most such structures in the past were of a meshed nature [1], recent developments towards building closed (sheet-like) elements have opened up the field for a whole new range of possibilities [2]. In a next step we now explored post-growth electron beam curing (EBC) [3], where the structures are locally irradiated without precursor gas present. This process impacts the inner structure and the overall volume of exposed elements and, if only applied partially, enables controlled deformation. We therefore performed experimental series, analyzed via SEM and TEM and complemented by Monte Carlo Simulations to explore and identify ideal parameters for smooth, stable and reproducible morphological bending. Figures 1a and 1b show a vertical wall with a width of 1 µm and a height of 2 µm that was bent via electron beam irradiation within a defined area across the structure. Figure 1c shows a more complex (originally straight) screw element where two areas have been exposed to EBC, clearly illustrating the bending effect towards the incidence direction. We attribute this “forward” bending to smaller interaction volumes of the incoming electrons compared to the wall thickness, mainly influencing the front part of the elements in comparison with the back side. We evaluated the impact for a variety of parameters, such as voltage, point pitch, dwell time, overall dose and beam incidence angle to achieve controlled and reproducible results. The expansion to more complex EBC patterns leads furthermore to more sophisticated bending as will be presented as well (see Fig. 2). We thereby extended the post-growth treatment possibilities of FEBID, showing the flexibility of EBC for various applications in research and development, some of which clearly go beyond the capabilities of sole 3D FEBID (e.g. spatially tuned mechanics).

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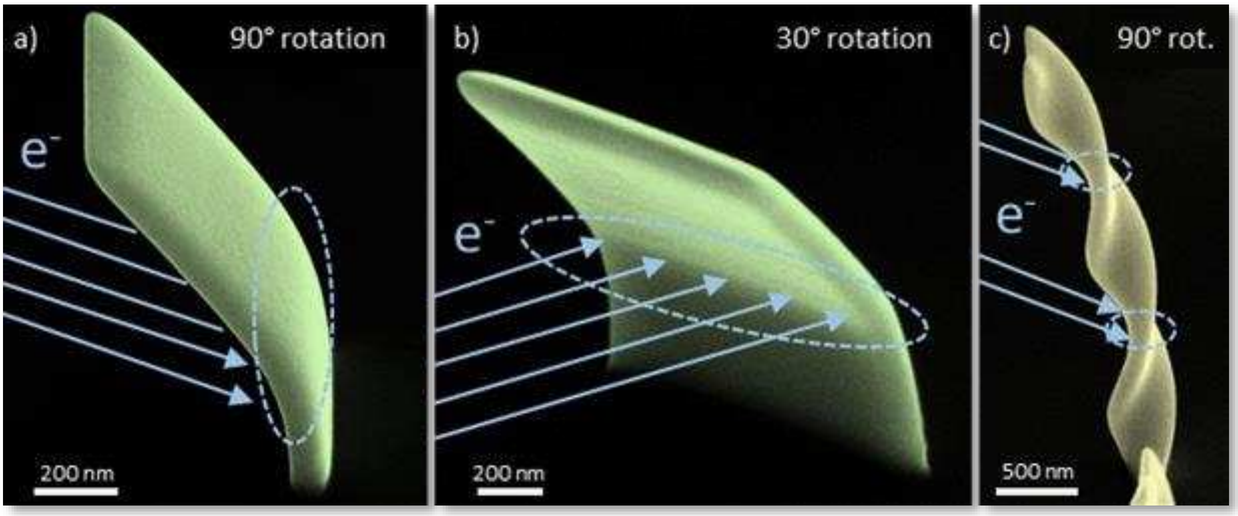

Fig. 1: Bending of sheet-like 3D FEBID elements. Bent wall from a side angle (a) and from a 30° rotated point of view (b) and twofold bent screw structure (c).

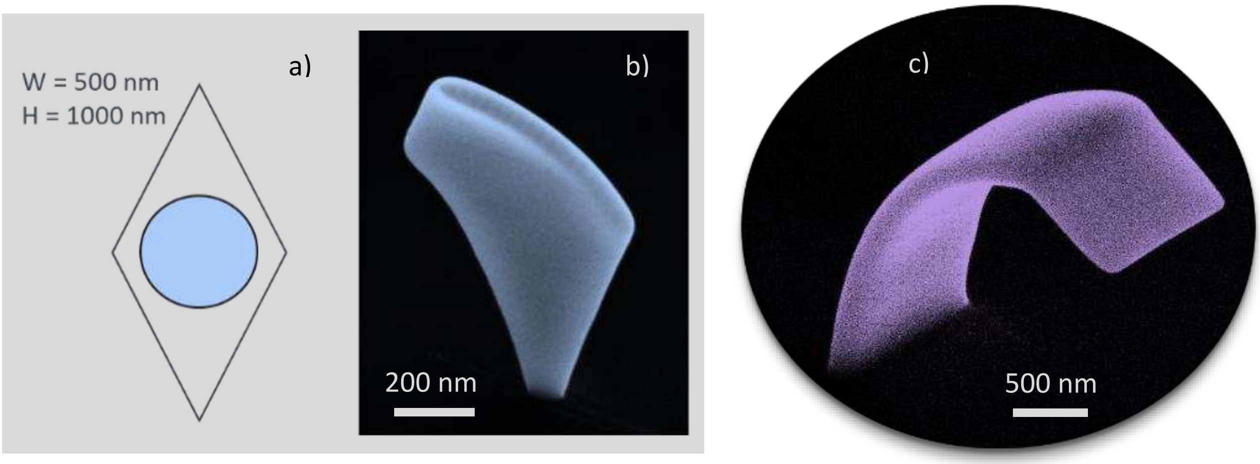

Fig. 2: More advanced electron beam curing with (a) a schematic for the bending of a diamond architecture via a circular curing pattern, (b) the corresponding SEM image of the real structure and (c) a wall element that was bent to an overhang.

High-Precision 3D-Nanoprinting for Sheet-like Structures via FEBID

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Among the few additive-manufacturing techniques capable of creating 3-dimensional objects on the nanoscale, 3D nanoprinting via Focused Electron Beam Induced Deposition (3D-FEBID) is an increasingly relevant technology for building high-fidelity nanostructures. Its capabilities of depositing feature sizes below 20 nm under optimized conditions and below 100 nm on a regular basis and its flexibility both in terms of substrate as well as precursor materials make it a unique technology with many possibilities and yet unexplored applications. While it has been used and developed further for a few years now, most fabricated structures in the past have been meshed [1], meaning a combination of differently oriented, individual nanowires, connected at specific points in 3D space according to the target application. This work leverages 3D-FEBID to the next level by expanding its capabilities from mesh-like towards closed (sheet-like) structures with a high degree of precision. The main challenge and source of most deviations from target shapes is thereby based on local beam heating and its implications on local growth rates. While well-understood in meshed structures, closed objects revealed additional dependencies on the dimensions of built objects and the XY pixel position within the structures. Furthermore, electron trajectories are more complex in closed objects, introducing additional proximity effects. To address these problems, we combined finite-difference simulations with 3D-FEBID experiments and developed a Python-based compensation tool, capable of stabilizing the growth for each XY pixel point in individual patterning planes by pre-determined parameter adjustments (Fig. 1a). The gained insight allowed further expansion, now being applicable for different element widths and -heights, as demonstrated by more advanced structures (Fig. 1b). In a last step we introduced trapezoid and inclined elements into our compensation code (Fig. 2a), which we then combined to a “construction kit” tool that is able to build compound structures (Fig. 2b-d). By that, we crucially expanded FEBID-based 3D nanoprinting by opening up design possibilities for closed and consequently mixed objects for novel applications in various fields of research and development.

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[2] A. Weitzer et al.; Expanding FEBID-Based 3D-Nanoprinting toward Closed High-Fidelity Nanoarchitectures; ACS Applied Electronic Materials, 4 (2) (**2022**), 744.

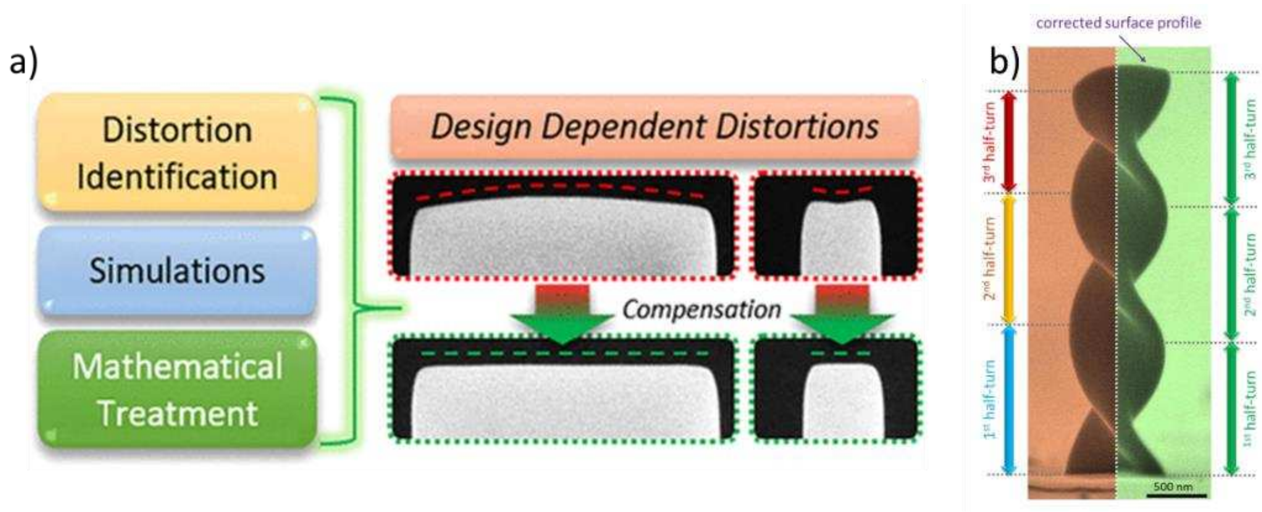

Fig. 1: Improvements due to our compensation tool on the example of (a) walls and (b) a screw [2].

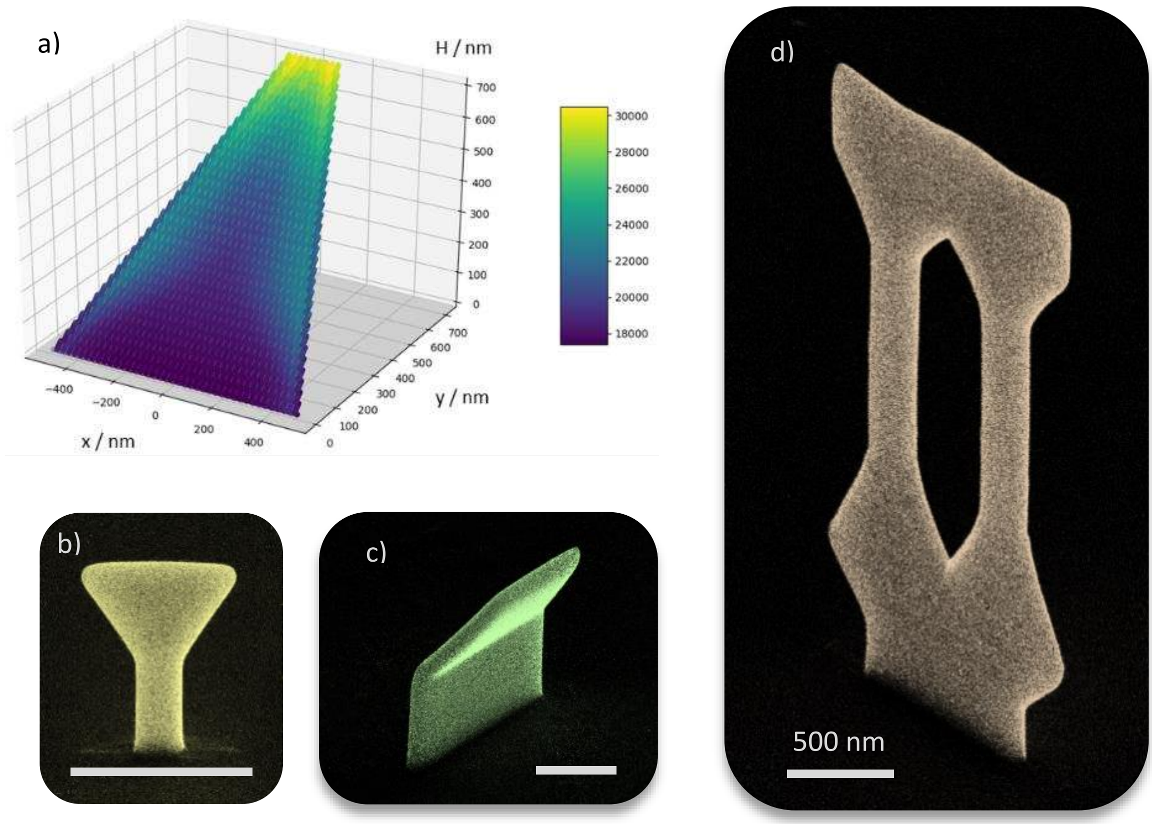

Fig. 2: Python compensation tool extensions for advanced FEBIP structures with (a) an illustration of the dwell time adjustments for a trapezoid structure and (b)-(d) SEM images of compound architectures.

Correlative Microscopy for Aviation and Aerospace

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Correlative microscopy (CM) workflows allow and aid solving a broad range of scientific and industrial problems (e.g. battery research, 3D printing, smart materials, integrated circuits, multi-physics simulation, etc.) previously unreachable by the typical experimental operando. CM workflows involve coordinated in 2D / 3D space and time (temporal 4D imaging) characterization of materials and components across a range of length scales. Various apparatus and imaging modalities contribute to the workflow, for example light, electron/ion microscopy, X-Ray computed tomography (CT), SIMS, EBSD, EDS, WDS, CL, XPS, Raman, STEM and TEM imaging and metrological techniques. These techniques use advanced cross-platform sample holders and dedicated software for automatic or guided coordinate transfer and locking solutions. CM delivers plethora of coregistrated 3D data that is often post-processed by artificial intelligence (AI) algorithms. AI-based segmentation or complex, multi-phase microstructures reduces the time-to-results from months to days or even dozens of hours. Further CM is often followed by an image-based modelling and multi-physics simulations.

As a practical example of CM for aerospace and defense industries we study a thermal barrier coating (TBC) used in the afterburner liner of turboramjet configured JT11D-20 engine (Fig. 1). The CM workflow used in this study correlates 3D data from HeliScan micro CT, femtosecond Laser Plasma FIB-SEM serial sectioning tomography across the length scales required to characterize and understand effects of long-term service on microstructural characteristics of the TBC (Fig. 2). Each system contributes structural (micro CT, Serial Sectioning) and analytical (EBSD, EDS) data directly within its characteristic length scale. The Laser Plasma DualBeam, cross-platform holder kit, Maps and Avizo software also plays a key role in integrating the workflow across the full range of length scales. Atomic scale S/TEM imaging and quantitative analysis is a logical extension of the workflow to the nano scale.

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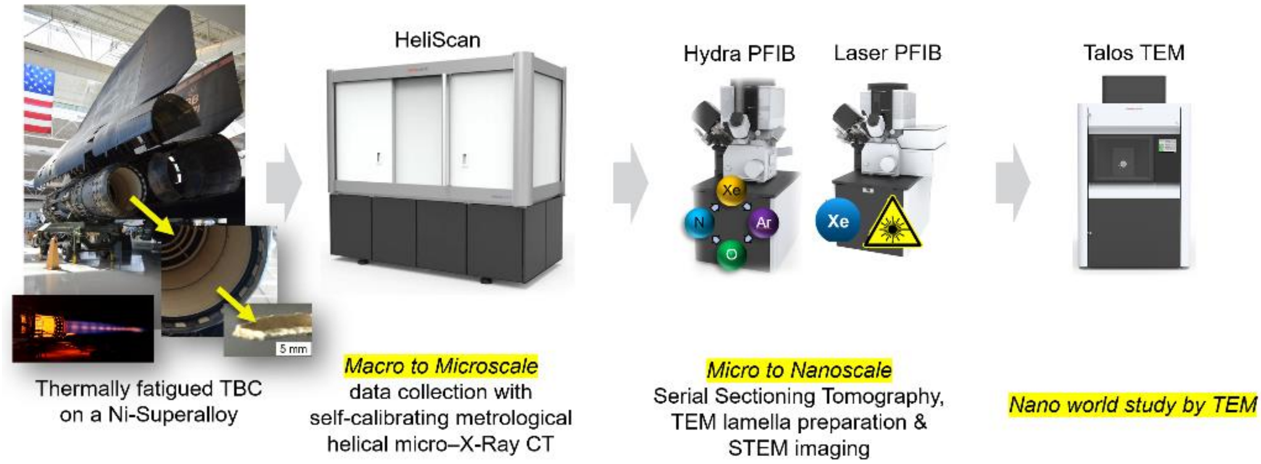
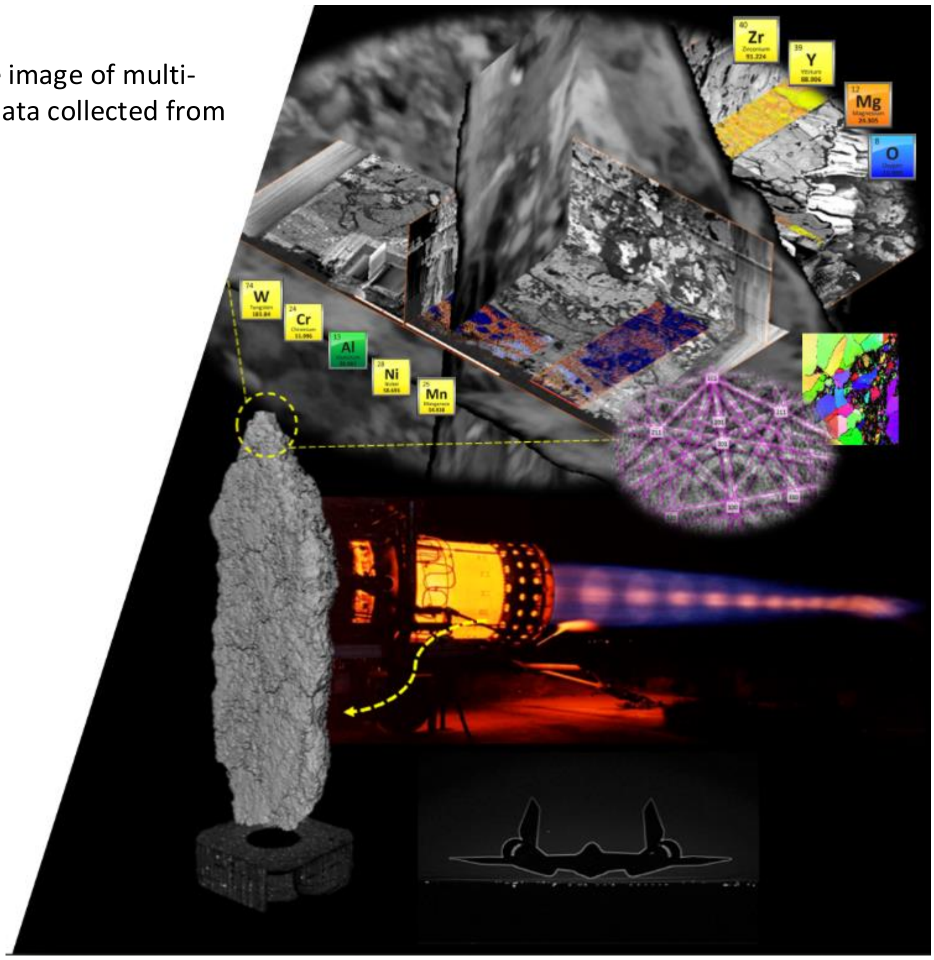


Fig. 1: Shows the experimental workflow.

Fig. 2: Shows composite image of multi-scale and multi-modal data collected from the TBC sample.



Plasma FIB spin milling hastens battery research

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The technology for energy storage systems is changing rapidly. In the past decade, the rapid growth of consumer electronics and electric vehicles market leads Li-ion batteries to attract significant attention. Currently two important industries, the energy and automobile industries, are involved in intensive research in this area where the most research takes place into: (a) solid state batteries [1], (b) lithium-ion batteries [2] and (c) sodium-ion batteries [3]. As the energy revolution gathers pace, batteries will be needed for energy storage in order to equalize the fluctuating power production of photovoltaic solar systems or wind turbines. The trend for electromobility places a lot of pressure to accelerate battery research.

Further advancement of their performance for higher energy and better safety is achieved by fundamental understanding of battery materials structures and chemistry throughout the life cycle. Various complex studies have been carries out in situ, ex situ and using 3D correlative multi-scale tomography and microscopy (CMT) [4]. Although CMT is very powerful methodology to gather 3D insight into material microstructure and chemistry [5, 6], it requires considerable amount of effort if compared to characterization of materials in two dimensions without prior screening with X-ray micro tomography (µCT). In many situations 2D information (SEM, EBSD, EDS, Raman, SIMS) from a cross-section of material give plethora of quantitative and statistical data. Plasma FIB-SEM opened fast access to 100's of microns wide and deep cross-sections allowing collecting multi-modal information with nanometer resolution [6]. Depending on the size of the cross-section, material and type of analyses needed the time-to-results (TTR) can vary from few dozens to hundreds of minutes. Considerable part of TTR is the site preparation time (SPT) for the cross-section: (a) MultiChem gas injection system deposition of a protective layer; (b) milling of the side trenches; (c) milling of the front trench to define of the cross-section and (d) polishing of the cross-section. For example, the SPT time for high resolution electron diffraction pattern analyses (HR-EBSD) over large areas (> 100's of µm2) is about < 25% of the TTR, while the SPT for high resolution SEM imaging is about > 95% of the TTR. The SPTs for other quantitative techniques sits in between the above percentage values of the TTR. In the recent years TTR was reduced by introducing new more sensitive detection systems, e.g. EDS, EBSD. Further, the femto-second Laser PFIB-SEM significantly reduced a cross-section preparation or serial sectioning time [7]. Currently, a new large area (<= 1 mm2) or large volume acquisition technique, so called Plasma FIB spin milling (PFIB-SM), was introduced by Winiarski at al. [8]. This technique collects data from superficial layers of a specimen surface and does requires minimum or no site preparation time.

This work presents for the first time the Plasma FIB spin milling method for applications in the battery research. As an example, a generic NMC cathode from a Li-ion battery cell was spin mill polished, where areas of 500 µm in diameter ware prepare within dozens of minutes. The PFIB-SM allows collecting curtain-free data from on-axis and off-axis location, thus accessing areas of about 50 mm2. This technique is well suited for rapid surface polishing and collecting multimodal information, while reaching an inert gas transfer condition is possible.

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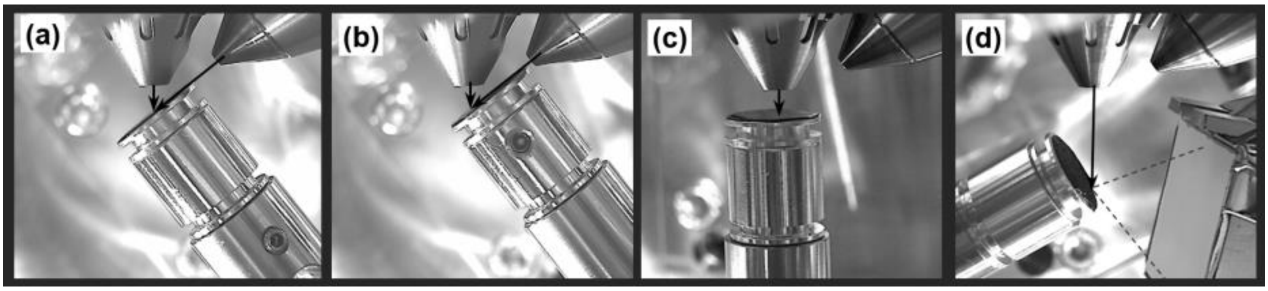

Fig. 1: Shows the experimental setup. Hydra PFIB spin milling (a) on axis and (b) off-axis; (c) SEM/EDS data collection and (d) EBSD data collection geometries.

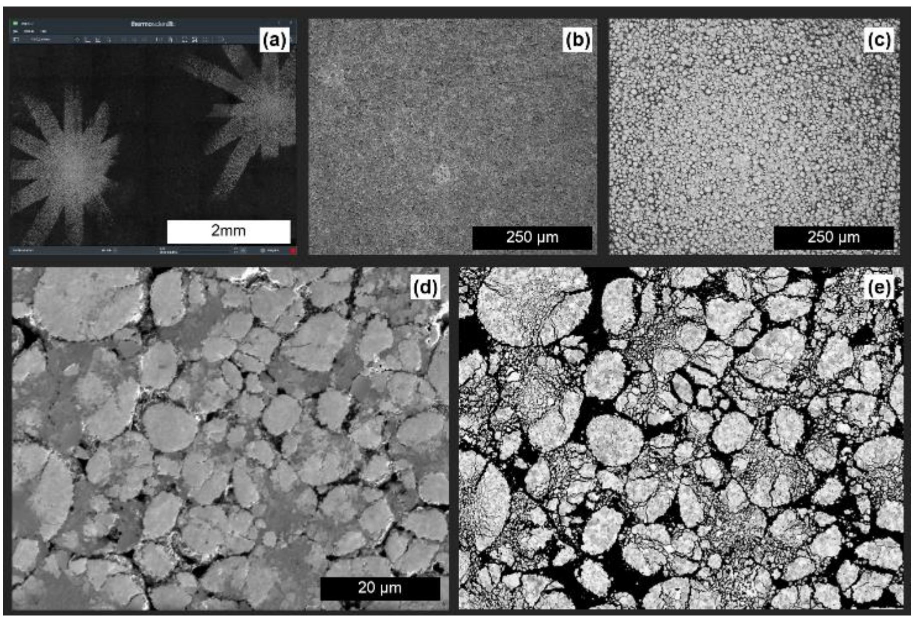

Fig. 2: Shows Xe PFIB spin milling results. (a) data is collected in a Maps project; the cathode before (b) and after spin milling (c) @ 30 kV, 60 nA, 1° glancing angle, < 60 min; (d) SEM ETD-SE 2kV, 0.8nA; (e) SEM CBS 5 kV, 1.6 nA

Plasma FIB-SEM-based Kintsugi Imaging for Battery Electrodes

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The mesostructure of porous electrodes used in lithium-ion batteries strongly influences cell performance [1]. Accurate imaging of the distribution of phases in these electrodes would allow this relationship to be better understood through simulation [2]. However, imaging the nanoscale features in these components is challenging. While scanning electron microscopy is able to achieve the required resolution, it has well established difficulties imaging porous media. This is because the flat imaging planes prepared using focused ion beam milling will intersect with the pores, which makes the images hard to interpret as the inside walls of the pores are observed. It is common to infiltrate porous media with resin prior to imaging to help resolve this issue [3], but both the nanoscale porosity and the chemical similarity of the resins to the battery materials undermine the utility of this approach for most electrodes.

In this study, a technique is demonstrated which uses *in situ* infiltration of platinum to fill the pores and thus enhance their contrast during imaging (Fig. 1). Reminiscent of the Japanese art of repairing cracked ceramics with precious metals, this technique is referred to as the *kintsugi* method. The images resulting from applying this technique to a conventional porous cathode are presented and then segmented using a multi-channel convolutional method [4] (Fig. 2). We show that while some cracks in active material particles were filled with the carbon binder phase, others remained empty, which will have implications for the rate performance of the cell. Energy dispersive X-ray spectroscopy was used to validate the distribution of phases resulting from image analysis (Fig. 2), which also suggested a graded distribution of the binder relative to the carbon additive. The equipment required to use the kintsugi method is commonly available in major research facilities and so we hope that this method will be rapidly adopted to improve the imaging of electrode materials and porous media in general.

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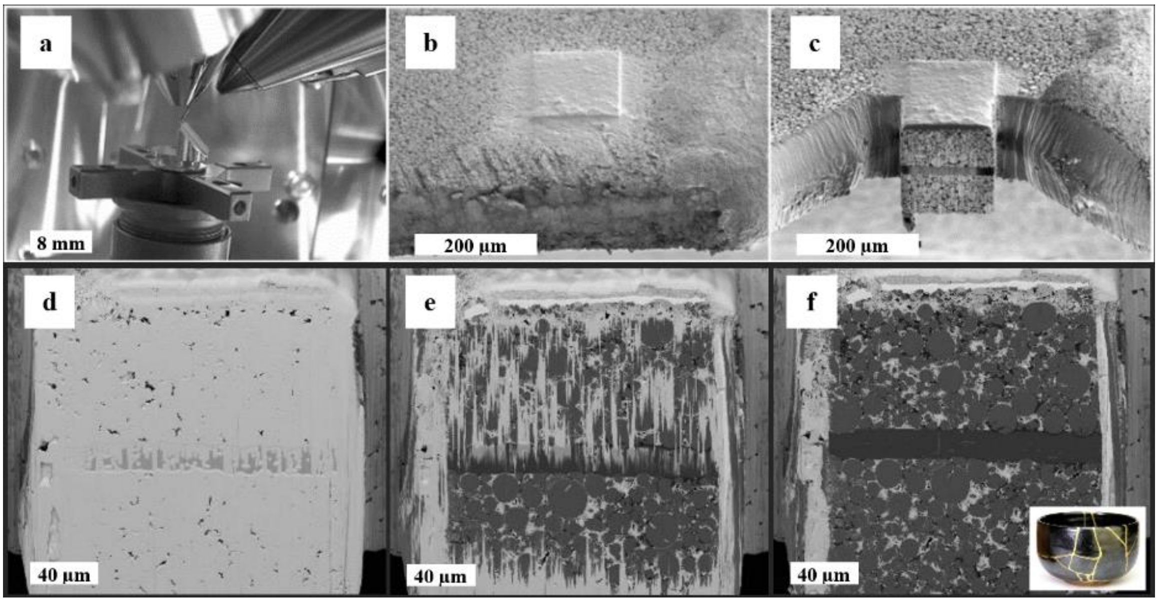

Fig. 1: The cross-section preparation workflow using Helios Hydra PFIB-SEM. (a) Image of the sample arrangement relative inside the vacuum chamber. (b) deposition of the Pt-C mix protective layer on the top surface of cathode. (c) the material block and block face preparation. (d) Pt layer deposition on the block face; (e) the automated ASV run, removing the excess of Pt layer. (f) image of the kintsugi infiltrated block face at 54 nm resolution (Inset shows an example of the pot repaired using the traditional kintsugi method). The scalebar in (a) is 8 mm; in (b, c) is 200 µm; and in (d-f) is 40 µm.

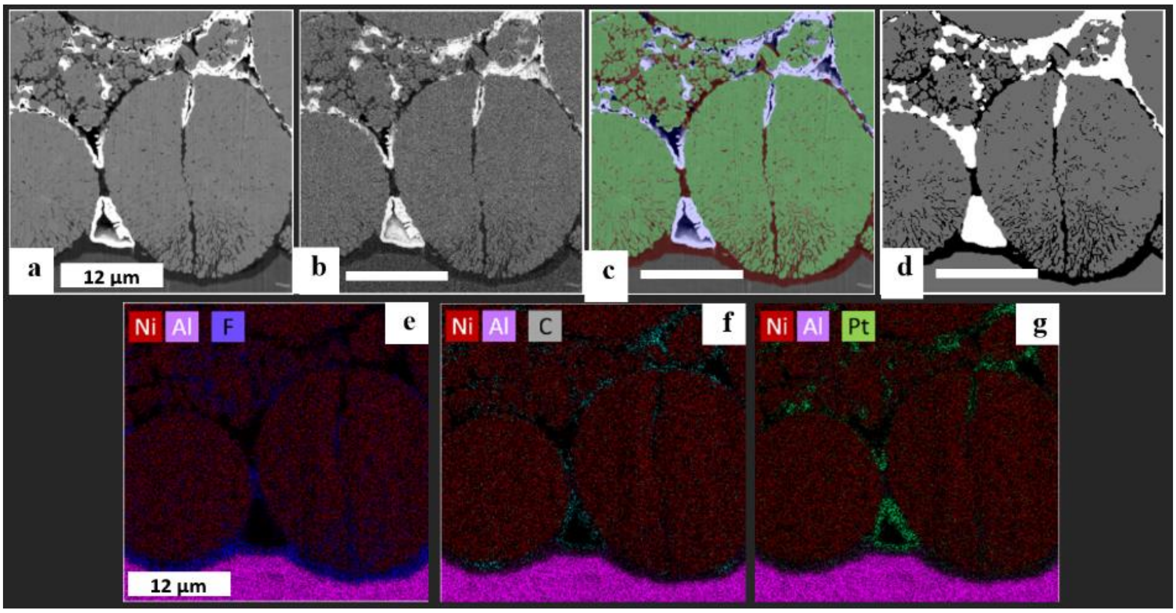

Fig. 2: Images of cathode cross-section prepared using kintsugi method. (a) SEM-BSE images using CBS detector. (b) SEM-BSE images using TLD. (c) CBS image overlaid with segmentation false colouring, where green is active material, red is CBD, and blue is pore. (d) Segmentation where white is pore, black is binder and grey is either active material or current collector. (e-g) Composite of EDS images highlighting the distributions of Al (current collector), Ni (AM particles), Pt (pores), and C (CBD), and the presence of F (polyvinylidene fluoride binder).

The FIB as 3D Nanoprinter – Overview of the Activities in Graz

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Focused particle beam microscopes are versatile tools as they allow nanofabrication in subtractive (ion beam milling, chemically assisted etching via ions or electrons) as well as in additive ways. For the latter, precursor gases are introduced by a gas injection system and locally immobilized by the focused particle beams. Those techniques, called Focused Electron / Ion Beam Induced Deposition (FEBID/FIBID), allows the deposition of nanoscale objects from different precursor materials even on extreme surface topographies and complement situations were alternative nanofabrication technologies run into their limitations (e.g. resist based lithography). Beyond planar and bulky objects, 3D printing of complex architectures becomes possible by a thoughtful control of the particle beam, leveraging both techniques into the status of a real 3D-nanoprinter [1].

In this contribution we highlight the research activities at the FELMI-ZFE in Graz (Austria), which are centered around 3D nanoprinting via FEBID. We line out actual challenges of this emerging technology on three frontiers (Fig. 1): (1) materials, (2) structure/geometries and (3) applications. For each area, we demonstrate possibilities, recent progress and remaining challenges. Furthermore, we show several applications, where such 3D-FEBID structures are used. A dedicated section presents the activities of the Christian Doppler Laboratory that focus on the direct-write fabrication of nanoprobes via 3D-FEBID with an industrial relevance. Here, we modify the tip region of Atomic Force Microscopy (AFM) cantilever with FEBID deposits [2] to enable advanced AFM modes such as conductive AFM, magnetic force microscopy or scanning thermal microscopy [3] (Fig. 2).

[1] R. Winkler et al.; *3D nanoprinting via focused electron beams.* J. Appl. Phys. (**2019**), 125, 210901

[2] H. Plank et al.; *Focused Electron Beam-Based 3D Nanoprinting for Scanning Probe Microscopy: A Review*. Micromachines (**2019**), 11, 48.

[3] J. Sattelkow et al.; *Three-Dimensional Nanothermistors for Thermal Probing*. ACS Appl. Mater. Interfaces (**2019**), 11, 22655

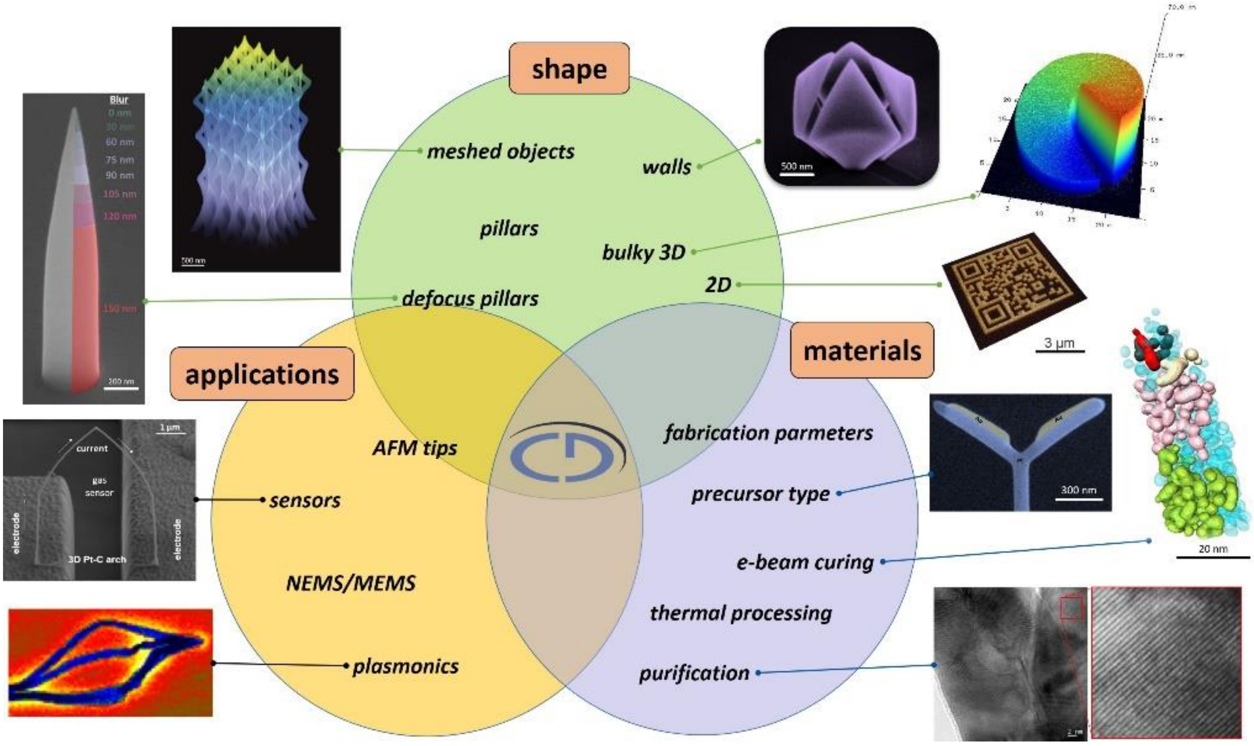

Fig. 1: Actual research areas on 3D-nanoprinting via Focused Electron Beam Induced Deposition addressed at the TU Graz, with a focus on direct-write of AFM nanoprobes.

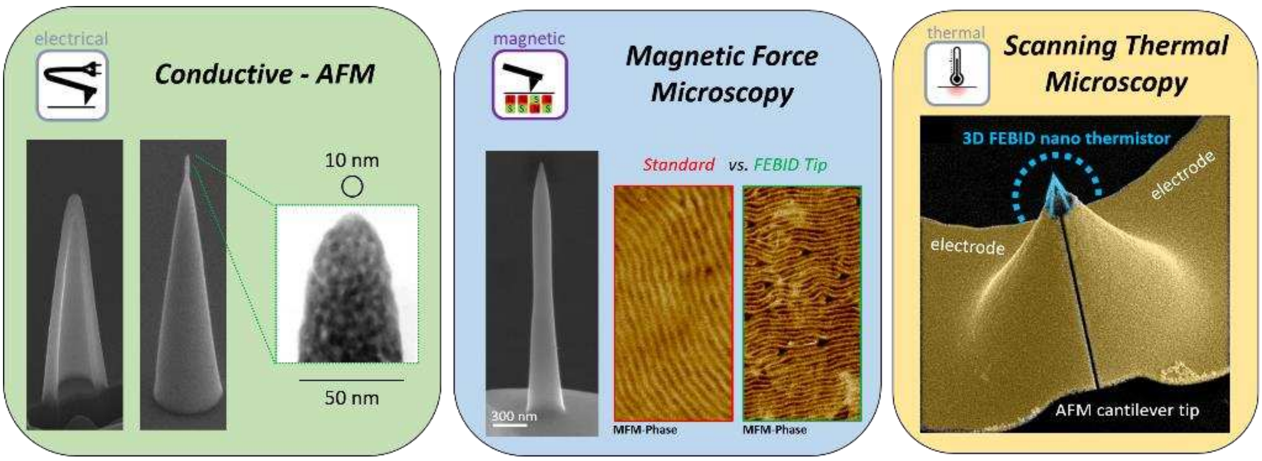

Fig. 2: Special 3D-FEBID nanoprobes for advanced AFM measurements (C-AFM, MFM, SThM), developed in the Christian Doppler Laboratory in cooperation with GETec Microscopy and Quantum Design Microscopy.