FEMMS 2024
Frontiers of Electron Microscopy and Materials Science

Frontiers of Electron Microscopy and Materials Science FEMMS 2024

29 September
4 October
2024

Catania - Sicily, Italy

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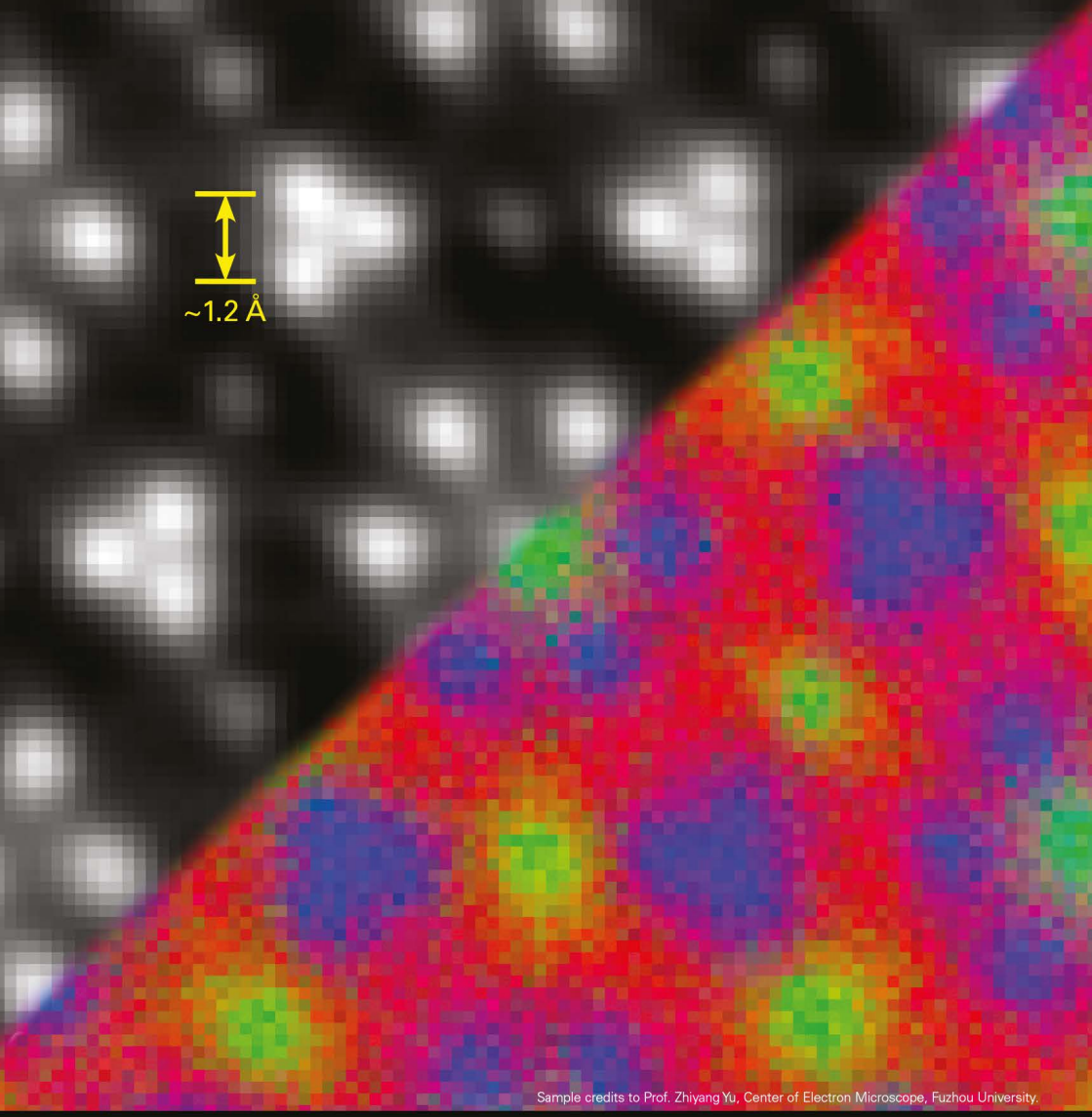
SCIENTIFIC PROGRAM AND ABSTRACT



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WELCOME

We are delighted to host the 18th Frontiers of Electron Microscopy in Materials Science (FEMMS) meeting in Catania this year! Since the last FEMMS meeting in Asheville, USA, in 2019, this journey to FEMMS 2024 has been challenging, with the global COVID-19 pandemic forcing us to postpone FEMMS multiple times. After a five-year hiatus, we are excited to welcome you to this week-long conference and hope you find it both enjoyable and enriching.

FEMMS 2024 is being held in the picturesque town of Aci Castello, Sicily. Known for its medieval Norman castle perched on volcanic rock overlooking the Ionian Sea, Aci Castello offers breathtaking views, rich history, and a tranquil atmosphere, an ideal setting for FEMMS' return. This marks the first time the conference is being held in Italy, and we have carefully chosen this unique location for FEMMS 2024.

Our program focuses on the latest advancements in transmission electron microscopy, featuring invited presentations from leading experts in experimental and theoretical science. FEMMS 2024 will include eight sessions highlighting emerging techniques in electron microscopy, with topics covering advances in spectrometers, detectors, in-situ experiments, data analysis, machine learning, and modeling.

FEMMS has always attracted a distinguished group of electron microscopists from across the globe, and FEMMS 2024 will be no exception. We believe that the combination of an inspiring program and this beautiful location will spark stimulating discussions and will generate new ideas to drive the future of electron microscopy and its applications in materials science.

We look forward to an exciting and productive conference over the next five days!

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FEMMS 2024
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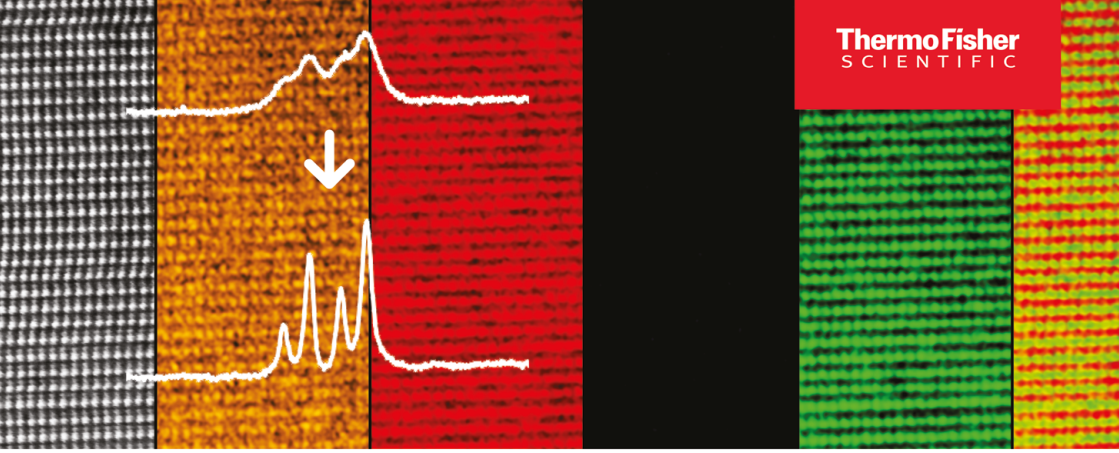

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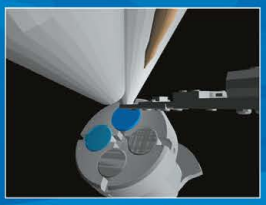

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FEMMS Program Overview

Starting Time	Sunday, September 29th	Monday, September 30th	Tuesday, October 1st	Wednesday, October 2nd	Thursday, October 3rd	Friday, October 4th
9.00		**Session I** New Instrumentation and Methods	**Session III** Pushing the Limits of Quantitative Analysis of Materials	**Session V** Time-resolved TEM	**Session VI** Theory of Microscopy and Modeling of Materials	**Session VIII** Frontiers in High Resolution Electron Spectroscopy
10.00						
11.00						
12.00					Lunch	
12.30			Lunch	Visit to Taormina (packed lunch)		
13.00		Lunch	Time for discussion		Time for discussion	Lunch Talk by Protochips
13.30		Time for discussion	Advisory Committee Meeting			
14.00					**Session VII** Artificially Intelligent Imaging and Spectroscopy: Powerful, Trustworthy, and Sustainable?	
15.00	Registration	**Session II** 4-D STEM and New Detectors	**Session IV** In-Situ, cryo-and low-dose analysis		Plenary talk: Frances M. Ross	Departure
16.00						
17.00						
18.00	Welcome reception				Visit to Catania	
19.00	Poster Session preview (prosecco & food)	Dinner	Dinner	Dinner "Cantine Firriato"	Gala Dinner	
20.00		Poster Session (wine & food)	Poster Session (wine & food)			
21.00						

Distinguished Lecture

Frances M. Ross

Frances M. Ross is a distinguished scientist and professor at the Massachusetts Institute of Technology (MIT), renowned for her groundbreaking research in nanotechnology and electron microscopy. She received her B.A. in Physics and Ph.D. in Materials Science from Cambridge University, UK, where she became captivated by electron microscopy. She continued this interest during her postdoc at AT&T Bell Laboratories, as a Staff Scientist at the National Center for Electron Microscopy, Lawrence Berkeley National Laboratory, and later as a Research Staff Member at the IBM T. J. Watson Research Center.

Prof. Ross joined MIT in 2018, where she holds the Ellen Swallow Richards Chair. Her research focuses on pushing the boundaries of electron microscopy techniques, including in-situ electron microscopy techniques to help understand self-assembly, electrochemical and other liquid phase processes. Her pioneering work in in-situ electron microscopy has revolutionized the field, allowing for the real-time observation of dynamic processes, such as crystal growth, nanoparticle assembly, and surface reactions. By leveraging advanced microscopy tools and innovative experimental methods, Ross and her team have uncovered fundamental insights into the behavior of nanomaterials, paving the way for advancements in diverse areas including materials science, physics, and nanotechnology. Prof. Ross is a Fellow of the American Association for the Advancement of Science (AAAS), the American Physical Society (APS), the Microscopy Society of America (MSA) and the Royal Microscopical Society (RMS). Prof. Ross is the author of more than 200 scientific original papers in this field, and has given uncountable invited talks at international and domestic conferences. In 2018 she was awarded the International Federation of Societies for Microscopy Hatsujiro Hashimoto Medal.

PROGRAM Sunday, September 29th

15:00-17:00	**Registration**
18.00-19:00	**Welcome reception**
19:00-21:00	**Poster Session preview** (prosecco & food)

PROGRAM Monday, September 30th

	Session I: New Instrumentation and Methods Session Chairs: Giuseppe Nicotra, Robert Klie and Paolo Longo	
09:00-09:30	**OPENING DISCUSSION**	
09:30-10:00	The Advantages of Sparse Sampling and Inpainting for High Resolution, In-situ and Ultrafast Electron Microscopy	Nigel Browning
10:00-10:30	Magnetic-field-free Atomic Resolution STEM for Magnetic Materials	Naoya Shibata
10:30-11:00	**COFFEE BREAK**	
11:00-11:30	A Fresh Look at Polymer Microstructure using 4D-STEM	Paul Midgley
11:30-12:00	The TOMO Project – Integrating a Fully Functional Atom Probe in an Aberration-Corrected TEM	Joachim Mayer
12:00-12:30	EELS at Extreme Energy Losses (XEELS™); an opportunity to obtain X-ray absorption spectroscopy-like information on Iliad microscope	Sorin Lazar Thermo Fisher Vendor Talk
12:30-13:00	Quantum Materials Properties Explored Through New Eyes: Advancing the Detection Capabilities of STEM-EELS	Juan Carlos Idrobo
13:00-13:30	**LUNCH**	
13:30-15:00	**TIME FOR DISCUSSION**	

	Session II: 4-D STEM and New Detectors Session Chairs: Jo Verbeek and Knut Muller-Caspary	
15:00-15:30	Electron Beam shaping and computational imaging: Present and perspectives	Vincenzo Grillo
15:30-16:00	Time-resolved nanothermometry using photon-electron pump-probe spectroscopy	Florian Castioni
16:00-16:30	Observation of atoms in motion by fast electrons	Christoph Koch
16:30-17:00	**COFFEE BREAK**	
17:00-17:30	Making Every Electron Count; Ptychography under low dose conditions	Angus Kirkland
17:30-18:00	EELS of Beam Sensitive Samples Using In-Situ Spectrum Imaging	Ray Twesten Gatan Vendor Talk
18:00-18:30	Event driven electron microscopy: What and why?	Daen Jannis
18:30-19:00	The Data Revolution in Electron Diffraction for Materials Characterization at Atomic and Nanoscopic Scales	Jian-Min Zuo
19:00-20:00	**DINNER**	
20:00-21:00	**POSTER SESSION** (wine & food)	

	Session III: Pushing the Limits of Quantitative Analysis of Materials Session Chairs: Marta Rossell and Ryo Ishikawa	
09:00-09:30	Crystal Nucleation and Growth in High-Entropy Alloys Revealed by Atomic Electron Tomography	Jianwei Miao
09:30-10:00	New Ways for Probing Real-Space Topological Polar Textures and Their Phase Transitions	Yu-Tsun Shao
10:00-10:30	Unveiling Structural and Electronic Order in 2D Materials with Cryogenic STEM and EELS	Miaofang Chi
10:30-10:45	COFFEE BREAK	
10:45-11:15	Probing magnetic properties at the nanoscale: A novel setup for in-situ Hall measurements in a TEM	Darius Pohl
11:15-11:45	Topological Polar Structures in Freestanding Ferroelectric Membranes	Gabriel Sanchez Santolino
11:45-12:00	Democratizing Advanced Methods of Precession-Assisted Electron Diffraction for Characterization of Advanced Materials and Thin Films	Daniel Nemecek Tescan Vendor Talk
12:00-12:30	Atomic scale mechanisms of grain rotation in nanocrystalline materials by In-Situ 4D TEM	Xiaoqing Pan
12:30-13:00	LUNCH	
13:00-15:00	TIME FOR DISCUSSION ADVISORY COMMITTEE MEETING	

	Session IV: In-Situ, cryo-and low-dose analysis Session Chairs: Shelly Conroy and Jungwon Park	
15:00-15:30	Visualizing catalytic processes at the atomic-scale	Stig Helveg
15:30-16:00	Understanding $LiNi_xCo_YMn_zO_2$ batteries by advanced transmission electron microscopy	Paulo Ferreira
16:00-16:30	**COFFEE BREAK**	
16:30-17:00	Electron Videography and Machine Learning of Soft Matter	Qian Chen
17:00-17:15	Advanced MEMS-Based In Situ Systems: Introducing Our Cutting-Edge Cooling-Heating-Biasing Solutions and Environmental Systems for Cross-Platform Research	Luca Carnevale DENSsolutions Vendor Talk
17:15-17:45	Combining in-situ synchrotron and electron microscopy techniques to study NiFe catalysts for CO_2 methanation	Manfred Schuster
19:00-20:00	**DINNER**	
20:00-21:00	**POSTER SESSION** (wine & food)	

	Session V: Time-resolved TEM Session Chairs: Thomas LaGrange and Mitra Taheri	
09:00-09:30	Mapping nanoscale near fields with attosecond electron microscopy	Murat Sivis
09:30-10:00	Probing Phonons and Magnons using Ultrafast Electron Microscopy	Yimei Zhu
10:00-10:30	Attosecond Electron Microscopy	Peter Baum
10:30-11:00	COFFEE BREAK	
11:00-11:30	Ultrafast coherent manipulation of free electrons via quantum interaction with shaped optical fields for advanced imaging approaches	Giovanni Maria Vanacore
11:30-12:00	JEOL Recent Trials for Scanning Transmission Electron Microscopy	Yu Jimbo JEOL - Vendor Talk
12:00-12:30	Microsecond Time-Resolved Cryo-EM	Ulrich Lorenz
12:30-13:00	LUNCH	

	Session VI: Theory of Microscopy and Modeling of Materials Session Chairs: Jani Kotakoski and Rohan Mishra	
09:00-09:30	Ferroic materials - designed and probed using electrons	Rohan Mishra
09:30-10:00	From modelling to generation: EELS and Materials Development	Teruyasu Mizoguchi
10:00-10:30	**COFFEE BREAK**	
10:30-11:00	Evolution of the Atomic Structure of 2D Materials under Electron Beam in a TEM: Insights from First-Principles Calculations	Arkady Krasheninnikov
11:00-11:30	A Comparative Analysis of Iterative and Non-Iterative Electron Ptychography	Colum O'Leary
11:30-12:00	Simulations of phonon and magnon EELS/EEGS including dynamical effects and multiple inelastic excitations	Jan Rusz
12:00-12:30	**LUNCH**	
12:30-14.00	**TIME FOR DISCUSSION**	

	Session VII: Artificially Intelligent Imaging and Spectroscopy: Powerful, Trustworthy, and Sustainable? Session Chairs: Lewys Jones and Steven R. Spurgeon	
14:00-14:30	Using TEM Data to Understand Bias in Machine Learning Algorithms	Katherine Sytwu
14:30-15:00	Fused Multi-Modal Electron Tomography for 3D Chemistry at Lower Dose and Higher Resolution	Robert Hovden
15:00-15:30	Reward-Driven Decision Making in Automated Microscopy	Sergei Kalinin
15:30-16:00	Machine Learning Enabled Advances in Lorentz Microscopy	Charudatta Phatak
16:00-17:00	**PLENARY TALK** Materials dynamics in situ, from nanoscale phase changes to motion within individual atomic columns	Frances M. Ross

	Session VIII: Frontiers in High Resolution Electron Spectroscopy Session Chairs: Demie Kepaptsoglou	
09:00-09:30	Single-Atom Spectroscopy in the meV to keV Energy-Loss Regime	Wu Zhou
09:30-10:00	Advanced STEM-EELS for materials discovery: challenges and opportunities	Berit Goodge
10:00-10:30	Exploring the dynamics of semiconductors with an ultrafast transmission electron microscope	Sophie Meuret
10:30-11.00	COFFEE BREAK	
11:00-11:30	Probing Nanoscale Light-Matter Interactions Using Fast Electrons	Andrew Yankovich
11:30-12:00	Correlating emission, absorption, and composition in perovskite nanocrystals	Sean Collins
12:00-12:30	Atomic-Level Visualizations of Frequency-Dependent Phonon Eigenvectors and Symmetry-Induced Vibrational Anisotropies	Xingxu Yan
12:30-13:00	LUNCH TALK BY PROTOCHIPS	

FEMMS 2024
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ABSTRACT SESSION I

...

New Instrumentation and Methods

PROGRAM Monday, September 30th

The Advantages of Sparse Sampling and Inpainting for High Resolution, In-situ and Ultrafast Electron Microscopy

Nigel D. Browning

- School of Engineering, University of Liverpool, Liverpool, L69 3GH, UK.
- SenseAI Innovations Ltd, Liverpool, L69 3GH, UK.
- Sivananthan Laboratories, 590 Territorial Drive, Bolingbrook, IL 60440, USA.

For many imaging and microanalysis experiments in high-resolution/in-situ/operando scanning transmission electron microscopy (STEM), the resolution and precision of the final result is primarily determined by the tolerance of the sample to the applied electron beam dose.

If the dose is not controlled, the stability of structures and the kinetics of dynamic observations can be dramatically changed by the beam, leading to a different structure and/or chemistry than would be expected from an ex-situ experiment under similar reaction conditions.
Recent results at the University of Liverpool (UoL) have shown that the optimal solution for dose control in any form of scanning/transmission electron microscopy is to form the image from discrete locations of a small electron beam separated by as far as possible in space and time. Instead of forming the image with an extended beam (as with TEM) or from a regular raster pattern (as in conventional STEM) this condition is satisfied ideally by moving the STEM probe over the area of the image using large jumps between the acquisition pixels, i.e. sparse sampling of the image.
This form of STEM imaging presents numerous challenges to the stability of the microscope, but these stability issues can be routinely overcome with minor changes to the hardware (an external scan controller) and by using either a form of random walk scanning, a calibrated random scan or a mixture of conventional scanning and rapid beam blanking. The larger than standard jumps between pixel acquisition locations in this sparse sampling methodology creates problems with image interpretation, as the gaps between locations of acquisition are missing information.
Fortunately, we can use Inpainting to retrieve the missing information and form a full image. Here I will discuss the methodology of Inpainting, with particular reference to the speed/efficiency of the reconstruction method and the potential for real-time video rate imaging across all forms of scanning images.

In addition, the use of simulations to provide a starting point for image interpretation and the use of deep learning approaches to allow the microscope to adapt its own imaging conditions will be demonstrated. Finally, the integration of these methods into the hardware design for both a new JEOL 300kV AI-STEM currently being installed at the UoL and the recently funded Relativistic Ultrafast Electron Diffraction and Imaging (RUEDI) UK national facility will be discussed.

Magnetic-field-free Atomic Resolution STEM for Magnetic Materials

Naoya Shibata

- Institute of Engineering Innovation, The University of Tokyo, Tokyo 113-8656 Japan.
- Nanostructures Research Laboratory, Japan Fine Ceramic Center, Nagoya 456-8587, Japan.
- Quantum-Phase Electronics Center (QPEC), The University of Tokyo, Tokyo 113-8656, Japan.

In scanning transmission electron microscopy (STEM), atomic-resolution observation and analysis of magnetic materials is normally very difficult since high magnetic fields are always exerted on samples inside the magnetic objective lens. In recent years, we have succeeded in developing a new magnetic objective lens system that realizes a magnetic field free environment at the specimen position [1].
Using this new objective lens system combined with the DELTA corrector, atomic-resolution imaging of magnetic materials is realized. This electron microscope (Magnetic-field-free Atomic Resolution STEM: MARS) is now used for research and development of many magnetic materials and devices [2,3]. By combining the MARS with differential phase contrast (DPC) imaging, it is shown that real-space visualization of atomic-scale magnetic fields of an antiferromagnet is possible [4].

In addition, recently developed tilt-scan averaging system for DPC imaging is installed in MARS [5] and enables local electromagnetic field imaging at heterointerfaces and grain boundaries by minimizing diffraction contrast [6,7]. In this talk, some recent application of MARS will be reported.

References

[1] N. Shibata et al., Nature Comm. 10, 2380 (2019).
[2] T. Seki et al., Nature Comm., 14, 7806 (2023).
[3] Y. Kohno et al., Microsc., in press.
[4] Y. Kohno et al., Nature 602, 234-239 (2022).
[5] Y. Kohno et al., Microsc., 71, 111-116 (2022).
[6] S. Toyama et al., Nature Nanotech., 18, 521-528 (2023).
[7] S. Toyama et al., submitted.
[8] The author thanks all the collaborators of this research, especially Y. Kohno, T. Seki, S. Toyama, Y.O Murakami, M. Takamoto, K. Tabata, T. Matsumoto, S.D. Findlay and Y. Ikuhara for their contribution to the works shown in this presentation. The author acknowledges support from JST ERATO Grant Number JPMJER2202, Japan.

A Fresh Look at Polymer Microstructure using 4D-STEM

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2 Department of Materials Science and NanoEngineering, Rice University, 6500 Main Street, Houston, TX 77005, USA.

Of the three 'traditional' classes of materials (metals, ceramics, polymers) [1], polymers have, until recently, perhaps been the least-studied by transmission electron microscopy despite their global commercial importance and concerns regarding the impact of polymers on the environment. Broadly, polymers can be split into two applications: the first structural, the second functional. In both cases the physico-chemical properties of the polymer are determined both by the 'ideal' polymeric packing and also by the deviations away from that ideal, through the introduction of defects, and the distribution of those defects to form what may be termed the polymer microstructure.

Over the past few decades the majority of polymer TEM (investigating crystalline and semi-crystalline polymers) has been based on imaging polymer microstructure using relatively large 'bespoke' crystals [2] or through using ingenious contrast-enhancing protocols [3]. The advent of fast, sensitive cameras and easy microscope control has opened up the use of 4D-STEM techniques to study (highly beam-sensitive) polymers in their 'native state' at much higher spatial resolution and thus reveal in more detail any underlying changes in polymer crystallinity [4,5].

In this paper we illustrate the use of a variant of 4D-STEM, scanning electron diffraction (SED), to study a number of polymers, with both structural and functional applications. SED is chosen as it provides high quality diffraction data, balancing the desire for near-parallel beams (for sharp spots) with probe size (spatial resolution). Two examples will be discussed in detail. The first, on polyethylene (PE), reveals how SED can elucidate the complex nanoscale twinning and twisting of PE lamellae [4] and reveal the subtle conformational disorder in regions between lamellae. The second, from an organic mixed ionic-electronic conductor (OMIEC) polymer [6], reveals the topology of polymer disclinations (rotational defects) and their interactions, and illustrates how the elastic constants (splay and bend) of the polymer may be quantified [7] through careful disclination analysis.

References

[1] Wyatt O. and Dew-Hughes D., 'Metals, Ceramics and Polymers' Cambr. Univ. Press, (1974)
[2] Allan P. and Bevis M., Proc. Soc. Lond. A, Vol. 341, (1974), 75-90
[3] Hudson S.D. and Thomas E.L. Phys Rev Lett 62 (1989) 1993
[4] Kang S.-J. et al. (2016). EMC 2016 Proc. doi: 10.1002/9783527808465.EMC2016.6121
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[6] Keene, S.T. et al, Nat. Mater. 22, (2023) 1121–1127.
[7] Zhang S., Terentjev E.M., and Donald A.M. Macromolecules, 37 (2004), 390-396.

The TOMO Project – Integrating a Fully Functional Atom Probe in an Aberration-Corrected TEM

Joachim Mayer[1 2], Juri Barthel[1], Ashok Vayyala[1], Rafal Dunin-Borkowski[1], Maarten Bischoff[3], Hugo van Leeuwen[3], Stephan Kujawa[3], Joe Bunton[4], Dan Lenz[4], Thomas F. Kelly[5]

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3 Thermo Fisher Scientific, Eindhoven, The Netherlands
4 CAMECA Instruments, Inc., Madison, WI, United States
5 Steam Instruments, Inc., Madison, WI, United States.

In the present contribution, we want to introduce the TOMO microscope, which will be based on a combination of two well-known but fundamentally different materials analysis techniques in one device: a state-of-the-art atom probe will be integrated into a high-performance TEM. This plan represents a revolutionary step towards atomic-precision analysis and has not previously been realized. The first important advantage results from the almost complete complementarity of the techniques, which we will be able to apply to the same object simultaneously [1-4]. In a correlative approach, the segregation of a specific element to a defect or any other microstructural feature in a given matrix phase can most sensitively be analysed with the atom probe technique. However, the atom probe cannot deliver atomically resolved information on the atomistic structure and the local bonding situation, which in a complimentary way can be given by HRTEM/STEM and EELS.
The TOMO instrument, which will be installed at the Ernst Ruska-Centre in 2025, will benefit from the almost complete complementarity of the techniques, which can be applied to the same object simultaneously. The expected synergistic gain obtained by combining the technologies results from the fact that it will be possible to observe the shape of the tip apex during an experiment and thus correct atom trajectories for greater spatial precision. For the first time, the TOMO instrument will permit the determination of the types and locations of millions of atoms in a volume of several hundred thousand cubic nm, down to each individual lattice position, in one measurement. The three-dimensional distributions of functional elements can be studied to almost the single-atom level, while linking it to atomic structures with picometre precision and electronic structures with sub-eV resolution. [5]

References

[1] Michael K. Miller, Thomas F. Kelly, Krishna Rajan, and Simon P. Ringer, Materials Today, April 2012, vol. 15, no. 4, pp. 158-165.
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[3] Thomas F. Kelly, Microscopy and Microanalysis, vol. 23 (2017) pp. 34-45. doi:10.1017/S1431927617000125
[4] Thomas F. Kelly, Brian P. Gorman, and Simon P. Ringer, "Atomic-Scale Analytical Tomography: Concepts and Implications," (Cambridge University Press 2022).
[5] The authors acknowledge funding from the German Ministry of Education and Research (BMBF) in the framework of the National Research Infrastructure Roadmap.

EELS at Extreme Energy Losses (XEELSTM); an opportunity to obtain X-ray absorption spectroscopy-like information on Iliad microscope

Sorin Lazar[1], Maria Meledina[1], Claudia S. Schnohr[2], Thomas Höche[3], Peter Tiemeijer[1], Paolo Longo[1], Bert Freitag[1]

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3 Fraunhofer-Institute for Microstructure of Materials and Systems IMWS, Walter-Hülse-Straße 1, 06120 Halle (Saale), Germany.

EELS in the electron microscope and XAS in the synchrotron are techniques for material characterization which both are based on ionizing core electrons. Both techniques have similar energy resolution but where the spatial resolution of EELS can go down to atomic scales, the spatial resolution of XAS is limited to micrometer scale. XAS is widely used technique for analysis of the extended fine structure of ionization edges, mainly thanks to the large range ~5keV ... ~40keV of ionization energies that can be probed [1]. In contrast the performance of EELS above about 2keV energy loss is often insufficient with respect to signal strength and energy resolution to perform ELNES or EXELFS [2]. The primary cause is that the energy of the energy-lost electrons does not match anymore the operating energy of the electron microscope thus leading to large chromatic effects. With the introduction of Iliad EELS spectrometer and its integration in the optic of the column, we developed lens settings which have minimized chromatic effects, as well as automated calibration and correction routines to correct the remaining chromatic defocus using the optics of the spectrometer. These improvements offer a greatly increased range of ionization energies up to 30 keV. Moreover, the EELS data quality is of enough quality to allow obtaining similar information as from XAS.

We exemplify the improvements by recording EELS near-edge fine structure (ELNES) of Zr L-edges at 2.3 keV from metallic glass precipitates, extended fine structure EELS (EXELFS) of Cu K-edge at ~9 keV, and EELS of Mo K-edge at 20 keV and Sb K-edge at ~30 keV (***Figure** 1*). We benchmark our data against near-edge and extended fine structure X-ray absorption (XANES and EXAFS) data and we quantitatively analyze the Cu Kedge EXELFS, demonstrating the capability to determine element-specific bond lengths and to distinguish different oxidation states such as metallic Cu, Cu2O, or CuO on a sub-micrometer scale.

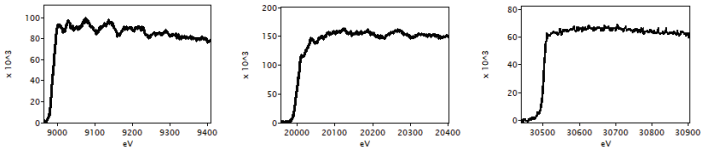

Figure 1. From left to right: Cu K-edge, Mo K-edge and Sb K-edge

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Quantum Materials Properties Explored Through New Eyes: Advancing the Detection Capabilities of STEM-EELS

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Scanning transmission electron microscopy (STEM), when combined with electron energy-loss spectroscopy (EELS), has the potential to detect properties associated with quantum materials with unprecedented spatial resolution. These properties include the emergence of magnetic ordering, valley polarization, phonon chirality, and topological characteristics such as Hall effects. In this study, we will show that achieving such measurements requires a configuration that ensures that electron momentum transfer in EELS mimics the role of polarization in light and X-rays.

Here, we will present two examples. [1] The first example demonstrates the detection of ferromagnetic ordering in lanthanum strontium manganite (LSMO) at room temperature. [2] The second example illustrates that orbital angular momentum, through the orbital Hall effect (OHE), can be detected and characterized at the nanometer scale. Subsequently, I will delve into the the detection of chiral phonons at the nanoscale, and the prerequisites for achieving two-dimensiona isotopic mapping with sub-nanometer accuracy. Throughout the talk, I will showcase experimental findings on these subjects and explore how their detection is ushering in a new era in materials characterization. [3]

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[1] The EELS part of this research was supported by the Center for Nanophase Materials Sciences, which is a Department of Energy Office of Science User Facility. This research was conducted, in part, using instrumentation within ORNL's Materials Characterization Core provided by UTBattelle,
LLC under Contract No. DE-AC05-00OR22725 with the U.S. Department of Energy.
This work was also partly funded under the Laboratory Directed Research and Development Program at Pacific Northwest National Laboratory, a multiprogram national laboratory operated by Battelle for the U.S. Department of Energy.
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FEMMS 2024
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ABSTRACT SESSION II

...

4-D STEM and New Detectors

PROGRAM Monday, September 30th

Electron Beam shaping and computational imaging: Present and perspectives

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Electron beam shaping is based on the idea that we can control the electron probes beyond standard electron optics elements by producing complex amplitude and phase landscapes for the electron beam.
Conversely 4D-STEM is the measurement of the diffraction plane for different probe positions.

The beam shaping, the interaction and the comprehensive detection like in 4D-STEM produce a triad of a quantum experiment: state preparation, interaction and analysis.
For techniques of computational imaging we find a “duality” relation between beam shaping and 4DSTEM. In facts in Computational Ghost Imaging (CGI) that uses multiple probes and a single detection and ptychography that uses a single probe and multiple position detection.

We are now approaching experimentally high resolution CGI and potentially testing the ability to reconstruct the probe. We are therefore following with some year delay the progresses of ptychography that has already reached maturity. However we believe there are more intermediate possibilities with different advantages in mixed schemes between CGI and ptychography.
In elastic scattering the symmetry between the CGI and ptychography as computational imaging schemes is related to the time reverse invariance of elastic scattering, which incidentally is not complete due to the dissipative character of the unavoidable diffuse scattering.

Conversely structuring the probe and the detection could be an essential approach for a quantum description of inelastic scattering.
I will show here how to extend methods of computational imaging in the inelastic regime in what can be called more properly Quantum State Tomography of the inelastically scattered electrons.

In this context I will discuss an early experimental example in the angular momentum subspace and a few theoretical proposals in a more general context.

Time-resolved nanothermometry using photon-electron pump-probe spectroscopy

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Understanding the thermal transport in nanostructures is a crucial and complex phenomenon for many technologies, especially as devices shrink in size towards the nanometer scale. In this case, studying out-of-equilibrium behavior requires access to timescales from ps to µs. Existing methods [1] that provide both these temporal and spatial resolutions remain limited. However, recent advances in event-based direct detectors in electron microscopy [2] open new possibilities.

In this study, we use distinct EELS signatures to measure the temperature of 2D semiconductors, Si3N4, and Al films after focused (~1 µm) and pulsed (~20 ns) laser excitation in the visible range ***(Fig. 1a)***. By synchronizing the laser pulse with a Timepix3 detector, we introduce a novel method for measuring temperature with nanometer ***(Fig. 1b)*** and nanosecond ***(Fig. 1c)*** resolutions.

A simple 2D diffusion model allows us to model the observed temperature dynamics. Since various spectroscopic excitations from IR to far UV are used, our approach provides a universal tool for probing nanosecond thermal dynamics in nanostructures. The temporal resolution here is limited by the technology of the Timepix3 detector and will be improved in the future. Furthermore, this method can be extended to diffraction-based techniques like 4D-STEM to detect subtle dynamical changes in crystalline lattices under different stimuli.

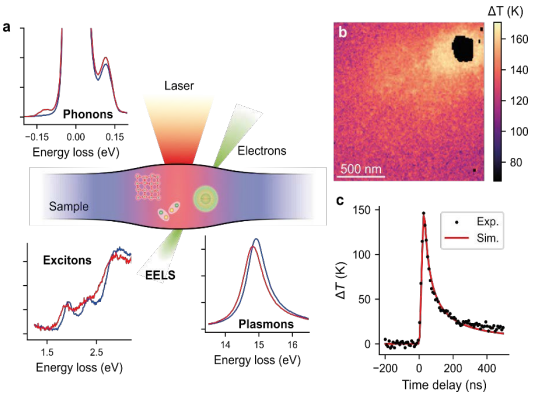

Figure 1. Description of the photon-electron pump-probe experiment.

(a) A pulsed laser in the visible range heats the sample, inducing changes in energy absorption probed by EELS across different energy ranges.
(b) Differential temperature mapping of an aluminum film following the laser exposition.
(c) Temporal profile of experimental temperature evolution after the laser pulse, compared with a heat diffusion simulation in an infinite 2D film.

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Observation of atoms in motion by fast electrons

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More than 20 years ago it was shown that proper modelling of phonons being thermally excited in a crystal yields highly quantitative electron diffraction simulations of bulk crystals [1]. More recently, it was demonstrated that quantitative fitting of 3D atomistic models to 4D-STEM data sets requires the thermal motion of the atoms to be included. Doing so, allowed the atom positions to be determined with a precision in the single pm regime [2]. We have recently shown that, even without assuming any atomistic model, the effect of thermal motion can be incorporated into iterative ptychographic reconstruction schemes, allowing the contribution of all atoms in the sample to be included, i.e. also the amorphous surface layers or contamination [3].

In a parallel development the instrumental improvement of simultaneous spatial and energy-resolution in the (S)TEM has reached a level that makes it possible to study spatial variations in the excitation of atomic vibrations [4]. Both types of information about the thermal motion of atoms are complimentary to one another, and their accessibility within the same instrument makes it possible to combine them. This talk will present recent experimental results of atomically resolved observation of variations in the phonon spectrum at different grain boundaries in silicon [5]. It will also be experimentally demonstrated that multi-object 3D ptychography, which accounts for thermal diffuse und multiple scattering, has superior resolution and reveals spatially-resolved statistics of the motion of atoms. The correlation of both measurements on the same atomic column may yield valuable information about details of the interaction between atoms on a very local level, i.e. including individual defects which cannot be studied directly using bulk diffraction or bulk spectroscopy methods.

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Making Every Electron Count; Ptychography under low dose conditions

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Acquisition of a ptychographic dataset typically requires the collection of a series of far field diffraction patterns as a function of probe position at the specimen plane. This can then be used to recover the complex specimen object function using either iterative or non-iterative algorithms. Importantly, ptychography is an inherently dose efficient technique, enabling effective the reconstruction of the exit wavefunction of radiation sensitive objects.
For applications in the life sciences cryo-electron ptychography holds much promise particularly when used with a defocused probe to scan across a specimen with highly overlapped probe positions.

This can be applied in a variant of conventional single particle analysis to provide 3D structures taking advantage of the known resolution variation of the effective ptychographic transfer function with convergence angle to provide wide spatial frequency
bandwidth transfer.
This geometry also allows datasets from wide fields of view to be collected that are suitable for studies of biologically relevant structures in a low concentration cellular context.
Ultimately the resolution of reconstructions of radiation sensitive samples is limited by radiation damage which inherently scales with electron fluence and in general most Ptychographic datasets have extremely low signal to noise.

Methods to overcome this will be discussed including sparse scan geometries optimised based on diffusion equations and the use of neural networks for processing of the raw input data. For the latter accurate centring of the bright field disk, data denoising and deconvolution of the detector MTF provides a typical 3-4 X enhancement of the SNR.

Finally, regularised and Fourier Ptychography as alternative data acquisition and processing strategies will be discussed.

EELS of Beam Sensitive Samples Using In-Situ Spectrum Imaging

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Acquiring time-series (or in-situ) EELS data allows for fractionating the electron dose over many data acquisitions, which are then parsed and summed to obtain the desired accumulated dose. Often, the critical dose threshold for a given material is unknown. While acquiring data from dosesensitive specimens such as biological cells, polymers, and ceramics, the ability to save time series
allows the removal of compromised data and only keep those acquired from the sample in a pristine state. The readout noise of traditional EELS detectors hinders this approach, but the new class of counting direct detection cameras are ideal for the method. The high-speed of these cameras even allows the extension to time series (or in-situ) spectrum images for combined spatial, spectra and temporal analysis.
Here, we demonstrate how in-situ spectrum imaging can be utilized to moderate the total dose to a sample over time and how it can be used to monitor changes to a sample by observing changes in the ADF images and ELNES shapes. We will demonstrate how in-situ spectrum imaging can moderate sample damage, obtain a high spatial resolution, and detect weak spectral signals in dosesensitive samples. Calcium carbonate (CaCO3) nanoparticles are used as a model system as the total dose thresholds for reduction and mass loss are well characterized [1]. Through in-situ spectrum imaging, we were able monitor the positions of the eg and t2g peaks of the Ca L2,3 edge to study how radiolysis drives the phase change from CaCO3 to CaO (Figure 1). Polymer-based materials are extremely dose-sensitive and undergo phase changes at low total doses of 100-1000 e-/nm2. We show that using the K3 counted camera with in-situ spectrum imaging doubled the resolution of ELNES phase mapping in a poly(styrene-acrylonitrile) polycarbonate polymer blend compared with a previous study that used a CCD-style camera to acquire EELS spectrum images [2]. Additional applications of this dose fractionated spectrum imaging will be presented.

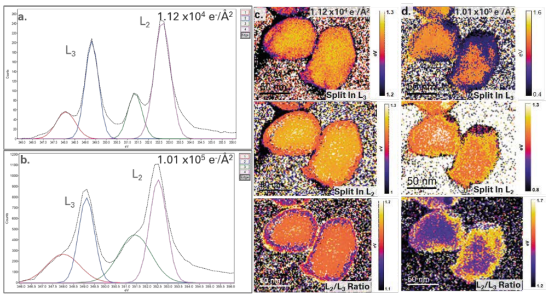

*Figure. 1. **(a)*** Extracted Ca L2,3 ELNES (Edge) from pristine CaCO3 ***(b)*** and after a total dose of 1.01e5 e-/Å2. Gaussian functions were fitted to each peak (1-4) using NLLS fitting. The split in the L3 and the L2 edges, as well as the L2/L3 edge intensity ratio, are calculated in ***(c,d)*** using the fitted parameters from Gaussians shown in ***(a)***.

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Event driven electron microscopy: What and why?

Daen Jannis, Jo Verbeeck

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- NANOlab Center of Excellence, University of Antwerp, Antwerp, Belgium.

* Currently at Thermo Fisher Scientific.

The electron microscope commonly uses a frame-based method to detect electrons where the signal generated from the interaction between the electron and the detector material is accumulated over a specific period of time. This implies that the ultimate temporal resolution is defined by the speed of the detector when no stroboscopic methods are used. The ultimate speeds of these detectors are currently 8 µs which is not fast enough to keep up with the conventional STEM dwell times which are around 1-2 µs [1].

Additionally, these fast frame-based detectors compromise in the number of pixels to accommodate for their increase in speed. In recent years, another method of detecting electrons has been developed which collects continuously individual events and stores for each event, the pixel position and the time of arrival where the time resolution is on the order of nanoseconds. This event driven measurement comes at the cost of having to use a low beam current (order off 1 pA) on the detector, although as shown in this presentation these type of detectors still have their applications. The first application which is being explored is performing low dose 4D STEM experiments at microseconds dwell time which is essential for dose sensitive materials[2].

Another use case for the event driven detectors is by correlating the outgoing electrons with the emitted x-rays. This coincidence technique enables detection of low concentrations and provides a unbiased estimate on the background signal in electron energy loss spectroscopy[3,4].

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[5] This project has received funding from the FWO project G042920N
'Coincident event detection for advanced spectroscopy in transmission electron microscopy'.

The Data Revolution in Electron Diffraction for Materials Characterization at Atomic and Nanoscopic Scales

Jian-Min Zuo[1 2]*, Robert Busch[1 2], Kaijun Yin[1 2], Hsu-Chih Ni[1 2], Haoyang Ni[1 2]

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The recent developments in electron optics and fast detectors have revolutionized the way materials are characterized at atomic and nm scale. Rather than relying primarily on electron images collected inside transmission electron microscopes (TEMs), increasingly, these instruments are designed more like diffractometers for the collection of massive diffraction datasets for 4D-STEM, taking advantage of the large electron elastic scattering cross-section and the electron focusing properties, down to hundreds of picometers. By data-mining diffraction datasets, the materials microstructure can be characterized in a quantitative way, and yet at unprecedented resolution and details. This form of data-driven electron microscopy is fundamentally different from traditional imaging approaches that were based on lenses and apertures. A major advantage is being able to form direct images of crystal structural properties and thus solves the perennial problem of electron image interpretation [1].

Innovations in diffraction techniques and analyses are critical to realize the full potential of data-driven electron microscopy. This talk will report on new developments in electron diffraction analysis from disordered materials to ordered crystals. It will be shown that the cepstral transformation of diffraction patterns is a powerful method for electron diffuse scattering analysis, and the use of cepstral signals for scanning transmission electron microscopy imaging allows the detection of chemical short-range ordering and deformation microstructure in advanced alloys [2,3]. For radiation sensitive materials, compressive sensing with sparse sampling potentially allows the imaging of molecular crystals [4]. Large angle rocking beam electron diffraction can be collected from crystals with large unit cells using direct electron detectors for structure and bonding analysis. These capabilities are in addition to strain and orientation mapping and ptychography, which makes 4D-STEM a powerful and yet versatile tool for materials characterization.

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ABSTRACT SESSION III

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Pushing the Limits of Quantitative Analysis of Materials

PROGRAM Tuesday, October 1st

Crystal Nucleation and Growth in High-Entropy Alloys Revealed by Atomic Electron Tomography

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High-entropy alloys (HEAs) have emerged as a pinnacle of materials design, spanning both structural and functional fields. For example, they offer an exceptional balance between strength and ductility in metallurgy, while exhibiting near-continuum adsorbate binding energies in catalysis. However, despite significant advancements in synthesizing HEAs, characterizing their structural and chemical order, and pursuing their broad applications, fundamental insights into the crystal nucleation and growth of HEAs at the atomic scale—essential for understanding their synthesis-structure-property relationships—remain elusive.

Here, we advance atomic electron tomography to determine the three-dimensional atomic structure and chemical composition of 8,094 HEA nuclei, captured at different stages of nucleation. We observe that early-stage nuclei exhibit a structural order that gradually decreases from the cores to the boundaries and is correlated with local chemical order. As the nuclei grow, their cores become more crystalline, and the structural order extends further along the radial direction. In the late stage of nucleation, most nuclei coalesce without misorientation, while a small fraction forms twin boundaries.

Since these results differ from classical nucleation theory and the two-step nucleation model, we develop an equation called gradient nucleation pathways to better explain our experimental observations. These findings provide a fundamental understanding of crystal nucleation and growth in HEAs. Moreover, we expect that gradient nucleation pathways can be broadly applied to elucidate a wide range of nucleation processes.

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New Ways for Probing Real-Space Topological Polar Textures and Their Phase Transitions

Yu-Tsun Shao[1], Ting-Ran Liu[1], Koushik Jagadish[1], Sergei Prokhorenko[2], Yousra Nahas[2], Lucas Caretta[3], Sujit Das[4], Zijian Hong[5], Ruijuan Xu[6], Fernando Gómez-Ortiz[7], Pablo García-Fernández[7], Long-Qing Chen[8], Harold Y. Hwang[9], Javier Junquera[7], Lane W. Martin[10], Laurent Bellaiche[2], Darrell G. Schlom[11], Ramamoorthy Ramesh[10], David A. Muller [12]

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Real-space topological dipolar textures, such as polar vortices, skyrmions, and merons, emerge in ferroelectric heterostructures due to the interplay of elastic, electrostatic, and gradient energies. These dipolar textures often exhibit functionalities like emergent chirality and local negative capacitance, offering potential applications in next-generation nanodevices. Given that these textures are inherently three-dimensional (3D) and nanometer-sized, characterizing their detailed atomic structures and understanding their topological phase transitions present significant challenges especially for TEM-based analysis that rely on 2D projection images. In this study, we investigate the topological phase transitions in epitaxial perovskite oxide heterostructures, such as (PbTiO3)/(SrTiO3) and (BiFeO3)/(TbScO3) systems, using a combination of atomic resolution imaging and four-dimensional scanning transmission electron microscopy (4D-STEM). Through 4D-STEM multislice ptychography [1], we discovered a novel phase in multiferroic BiFeO3, characterized by 3D dipolar waves with incommensurate periodicities and antiferrodistortive modes [2]. Furthermore, the 4D-STEM approach enabled direct observation of transitions among polar skyrmions, merons, and anti-merons, with some transitions accompanied by changes in chirality [3]. Research supported by AFOSR Hybrid Materials MURI (FA9550-18-1-0480) and ARO ETHOS MURI (W911NF-21-2-0162), facilities support from the NSF (DMR-1429155, DMR-1719875, DMR-2039380). Researchers at the University of Arkansas also thank the Vannevar Bush Faculty Fellowship Grant No. N00014-20-1-2834 from the Department of Defense. YTS acknowledge financial support from the USC Viterbi Startup Fund.

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Unveiling Structural and Electronic Order in 2D Materials with Cryogenic STEM and EELS

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2 Mechanical Engineering and Materials Sciences, Duke University, Durham, NC.

Quantum materials exhibit unique phenomena and functionalities beyond classical physics. The use of 2D sheets and constructing hetero- and moiré structures out of them has emerged as a promising method to induce exotic quantum effects. Studying these materials using cryogenic scanning transmission electron microscopy (STEM) and electron energy loss spectroscopy (EELS), was previously limited by stage instability.

Recent improvements in stage designs by manufacturers offer new opportunities for related research. In this talk, I will present our ongoing research using atomic-scale cryogenic STEM and monochromated EELS to investigate lattice-electronic structure coupling in several presentative 2D van der Waals structures for magnetic storage and spintronic applications.

Key examples include the discovery of layer-number-dependent phase transitions in CrCl3 as cooling, the elucidation of complex local symmetry breaking in long-wavelength charge density wave EuAl4, and the mapping of local excitons of moiré structured MoTe2. These studies demonstrate the electronic and magnetic properties of 2D materials can be tuned by manipulating the layer number or creating moiré structures. they also highlight the power of combining high-resolution cryogenic STEM imaging and spectroscopy for studying quantum materials in general [1].

References

[1] This work is supported by the U.S. DOE, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division.

Probing magnetic properties at the nanoscale: A novel setup for in-situ Hall measurements in a TEM

Darius Pohl[1], Yejin Lee[2], Dominik Kriegner[3], Sebastian Beckert[3], Sebastian Schneider[1], Andy Thomas[2 3], and Bernd Rellinghaus[1]

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Non-trivial magnetic textures, such as skyrmions, are – besides others – frequently characterized by measurements of the anomalous Hall effect and imaged using Lorentz TEM (LTEM). Frequently, however, these investigations are conducted on different samples, which may impede a direct correlation of transport and imaging data.

We have therefore developed an advanced in-situ magneto-transport measurement setup in a transmission electron microscope, which for the first time allows for the direct correlation of the imaged magnetic textures with the Hall effect [1]. The approach enables simultaneous magnetic imaging and four-probe electrical measurements supported by high-resolution structural and chemical characterization of the same sample.

As a proof-of-principle experiment, a 20 nm thin Ni film was deposited on an electron-transparent SiN window. LTEM was then used to image the magnetic domain patterns while concurrently conducting in-situ Hall effect measurements in the microscope. These successful first measurements pave the way for previously unreported in-situ and operando Hall measurements in combination with high-resolution magnetic imaging in the TEM. Further experiments are performed on a lamella of the anti-skyrmion hosting material Mn1.4PtSn, giving insights into the magnetic signatures of anti-skyrmions.

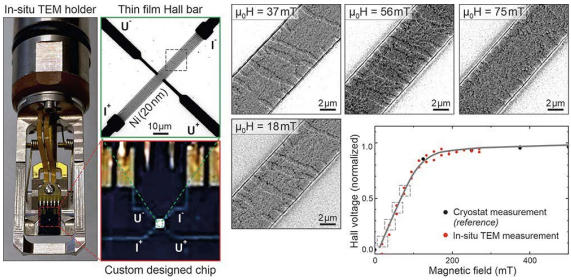

Fig. 1. Protochips Fusion Select holder with home-made measurement chip. LTEM images recorded simultaneously with Hall measurements reveal the domain growth upon magnetizing a 20 nm Ni film.

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Topological Polar Structures in Freestanding Ferroelectric Membrane

Gabriel Sánchez-Santolino[1], Víctor Rouco[1], Sergio Puebla[2], Hugo Aramberri[3], Víctor Zamora[1], Fabián A. Cuellar[1], Carmen Munuera[2 4], Federico Mompean[2 4], Mar Garcia-Hernandez[2 4], Andrés Castellanos-Gomez[2 4], Jorge Íñiguez[3 5], Carlos Leon[1 4], Jacobo Santamaria[1 4]

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Complex correlated oxides are quantum materials characterized by unshielded d-electrons in which the interaction across competing energy scales leads to diverse functionalities that can be altered by slight changes of their structure, composition, or boundary conditions [1]. In this context, recent studies on ferroelectric oxides have shown the formation of complex polar topologies, which are related to a delicate interplay between the intrinsic tendency of the material towards a uniform polarization and the electrical and mechanical constraints placed upon them [2]. However, the cube-on-cube epitaxial structure of these materials forces the use of single crystalline substrates for their growth, which restricts the possible mechanical boundary conditions and, therefore, the formation of new topological structures.

In this work, we have overcome this fact by introducing a novel approach to form a non-trivial topological polar state [3]. This is achieved by isolating epitaxially grown ferroelectric thin films from their parent substrate by a chemical exfoliation method and fabricating twisted freestanding ferroelectric bilayers, in which an unconventional strain landscape produces a vortex polarization pattern that is dependent on the twisting angle. This finding opens the path to creating novel topological textures, of just a few nm in size, that could serve as the base of innovative high-density memory devices

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Democratizing Advanced Methods of Precession-Assisted Electron Diffraction for Characterization of Advanced Materials and Thin Films

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Accessibility to advanced structural analysis of materials by electron diffraction techniques has been dramatically changed with the introduction of the new scanning transmission elecrtron diffraction microscope, the TESCAN TENSOR. The performance and usability of analytical 4D-STEM techniques boosted by beam precession is not compromised by the legacy design of conventional TEM microscopes anymore. The new STEM-dedicated design integrates state-of-the-art components such as a large direct electron detector with hybrid pixel technology, electron beam precession, electrostatic beam blanker and large dual EDS detectors. Moreover, ultra-high vacuum engineering guarantees unrestricted investigation of samples due to negligible hydrocarbon contamination from the column. Full integration of all these components facilitates their precise synchronization, true multi-modal data acquisition, and improvement in the performance and overall throughput of STEM, EDS, and 4D-STEM measurements.

Controlled from a single user interface that provides seamless user experience, acquired data are processed and analyzed on-the-fly and visualized almost in real time, making sample characterization an interactive experience instead of batch data acquisition and later post-processing. This novel approach to analytical 4D-STEM measurements has enabled simplification of sample analysis workflows that do not require TEM imaging and switching between TEM and STEM modes anymore. Advanced automation of system alignments in the background and fine guided adjustments of STEM imaging, STEM analysis and 4D-STEM nanobeam settings enable user experience comparable to SEM and FIB/SEM instruments. Users can therefore focus and spend precious instrument time on interaction with the specimen, whilst minimizing time spent on alignments of the TEM microscope. The set of collected nanoscale characterization data and results can thus be easily extended to structural information provided by the 4D-STEM and 3D-ED techniques, while data quality and accuracy of results is boosted by fully integrated beam precession.

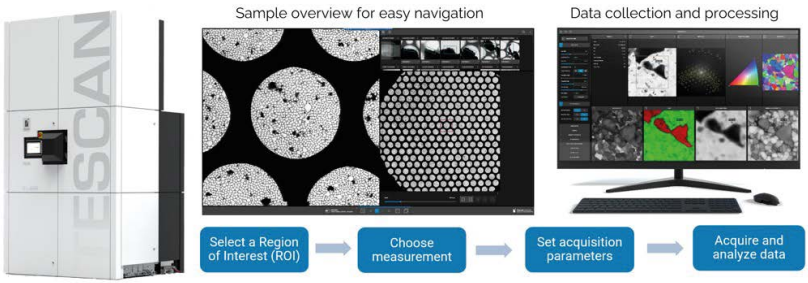

Figure 1. Streamlined sample analysis workflow using the novel multimodal analytical electron diffraction microscope, the TESCAN TENSOR.

Atomic scale mechanisms of grain rotation in nanocrystalline materials by In-Situ 4D TEM

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A polycrystalline material is an aggregate of crystallites with different lattice orientations. The crystallites, i.e., grains, can undergo nearly-rigid-body rotation. Such rotations play a central role in polycrystal dynamics (e.g., changing grain growth kinetics) and constitutes a primary deformation mechanism in nanocrystalline solids and may dominate texture evolution.

They have been widely observed in polycrystalline materials during grain growth, plastic deformation and recrystallization. Despite decades of research, the dominant mechanisms underlying grain rotation remain enigmatic, largely because of the difficulty in observing the evolution of internal interfaces. Here, we present direct evidence that grain rotation occurs through the motion of disconnections (line defects with step and dislocation character) along grain boundaries (GBs). State-of-the-art in situ four-dimensional scanning transmission electron microscopy (4D-STEM) observations reveal the statistical correlation between grain rotation and grain growth/shrinkage.

This correlation arises from shear-coupled GB migration and further the motion of disconnections, as demonstrated by in situ high-angle annular dark field STEM observations and the atomistic-simulation-aided analysis. These findings provide quantitative insights into the structural dynamics of nanocrystalline materials.

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ABSTRACT SESSION IV

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In-Situ, cryo-and low-dose analysis

PROGRAM Tuesday, October 1st

Visualizing catalytic processes at the atomic-scale

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The quest for sustainable energy technologies calls for new insight into catalysis. Catalysis of chemical reactions is commonly perceived as a complex surface phenomenon that inescapably links structural dynamics and functionality.

Uncovering the intricate relation between catalytic active surface sites and their mechanistic actions at the atomic-scale has, however, remained challenging due to the lack of microscopy tools operating at high spatial and temporal resolution under chemical meaningful conditions. Here, I will outline our efforts on advancing atomic-resolution electron microscopy as a foundation for observing the spatiotemporal behavior of catalysts in operando, and, in turn, on elucidating the role of gas-surface interactions on catalysts in their working state to bridge the so-called pressure and materials gaps in surface and catalysis sciences [1-10].

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Understanding $LiNi_xCo_yMn_zO_2$ batteries by advanced transmission electron microscopy

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$LiNi_xCo_yMn_zO_2$ (NCM) battery cathodes exhibit a specific capacity and operating voltage comparable to $LiCoO_2$, while being less toxic. They can be combined with different types of anodes and electrolytes to be used as solid state or liquid state batteries. In this regard, we have been investigating the use of Li metal cathodes and solid electrolytes, which are very sensitive to air/moisture and the electron beam. As a result, we have performed both room temperature and cryo FIB to obtain cross sections in Li metal and novel solid electrolytes. In this talk we will show the difference between the two procedures.

In addition, it has been reported that NCM is composed of a mixture of $LiMO_2$/ Li_2MnO_3 layers (where M=Ni, Co or Mn). $LiMO_2$ is associated with a trigonal (R3m) phase, whereas Li_2MnO_3 is associated with a monoclinic (C2/m) phase, which is responsible for storing and providing Li+ during Li-extraction of $LiMO_2$. Nevertheless, these layered materials exhibit a large degree of cation disorder, which disrupts the Li+ pathways and creates a continuous MO_2 layer, lowering the Li mobility and reducing the cycling performance. Yet, so far, these phenomena have been mostly studied in single-crystalline particles. However, at the industrial scale, the NCM produced is typically in the form of micrometer polycrystalline particles, where the primary particles possess a very complex microstructure, in particular an agglomeration of nanoparticles, porosity, as well as chemical and phase heterogeneity.

In this regard, this work aims to fundamentally understand the changes in chemical distribution as a function of Ni content in polycrystalline NCM cathode materials. In particular, the chemical composition and structure of the NCM polycrystalline particles with Ni contents from 0.70 up to 0.90 wt. % were investigated using FIB-SEM, aberration-corrected STEM, EDS mapping, precession electron diffraction and DPC-STEM. Slice-and-view SEM/FIB analysis revealed a geode-like morphology, independently of the composition. Yet, as the Ni content increases, the particle porosity decreases. STEM-EDS revealed variations in chemical composition across a single particle, in particular Ni content. Despite precession electron diffraction results suggesting the presence of only one crystal structure, atomic-resolution STEM images revealed that variations in the composition may be related to the presence of orientation variants within a single crystalline grain.

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Electron Videography and Machine Learning of Soft Matter

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We will present our group's recent progresses on establishing and utilizing "electron videography" to image, understand, and manipulate soft and biological systems, in space and time at a nanometer resolution. This involves systems that underpin the fundamentals of structure-function relationship for a wide range of phenomena and applications. In this talk, we will discuss in detail two systems. In the first direction, we focus on the phase behaviors of nano-sized building units as they are dispersed in solution. As a proof-of-concept, we directly image the crystallization pathways of nanosized colloids into superlattices, where the discreteness and multi-scale coupling effects complicate the free energy landscape.

Single particle tracking and simulations combined unravel a series of interesting pathways at this length scale, such as non-classical crystallization, size-dependent crystal growth habits of superlattices, and moiré patterning, enabling advanced crystal engineering. In the second direction, we shift to yet another soft matter system, to image the complicated irregular morphology of polyamide (PA) membranes to understand their morphology-property relationship. PA membranes serve as an active layer in thin-film composite (TFC) membranes used for organic solvent separation.

They exhibit rich nanomorphologies, such as crumples and interconnected networks, which are correlated with their separation performance. Here we show the combination of low-dose electron tomography and a suite of machine learning toolsets (including inpainting, segmentation, dimension reduction, and morphology classification) that allow us to digitalize the morphologies of PA membranes. We are then able to relate the morphologies to the synthetic conditions of the PA membranes to understand the morphogenesis mechanism as well as how the specific morphological parameters determine the final separation performance and mechanical properties in a quantitative manner. We see this platform can be applied to other soft materials with complicated morphologies.

Advanced MEMS-Based In Situ Systems: Introducing Our Cutting-Edge Cooling-Heating-Biasing Solutions and Environmental Systems for Cross-Platform Research

Yevheniy Pivak, Hongyu Sun, Luca Carnevale, Christian Deen-van-Rossum, Ronald Spruit, Merijn Pen, Hector Hugo Perez-Garza

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Recent advancements in in situ transmission electron microscopy (TEM) have led to the development of sophisticated systems for precise sample manipulation under various environmental conditions. MEMS technology, the core competence of our company, enables highly innovative solutions in this field. We introduce two cutting-edge platforms: the "Lightning Arctic" system and a novel environmental platform for the "Climate" and "Stream" systems, designed for in situ gas and liquid-phase TEM studies.

Lightning Arctic System: This MEMS-based system supports cooling, heating, and biasing within the TEM. It features the Nano-Chip, a MEMS-based sample carrier that delivers local stimuli. Compatible also with conventional 3 mm TEM grids, the system offers double tilt capabilities and the ability to achieve temperatures from ≤ -170°C to ≥ 800°C. The user can enable biasing experiments at any intermediate temperature. The unique modular design includes a removable liquid nitrogen dewar and an innovative holder architecture, ensuring high stability and atomic resolution while preventing bubble formation that could degrade image quality.

New Environmental TEM Platform: Enhancing the value of our "Climate" and "Stream" systems for in situ gas and liquid-phase TEM studies, respectively, this platform includes a holder with eight electrical contacts for combined thermal and electrical stimuli application alongside controlled liquid or gas flow. It supports tip rotation for seamless switching between TEM and STEM imaging modes without disrupting experimental conditions. The holder's generic removable tip design fits different TEM brands, enhancing cross-platform sample correlation. Furthermore, we demonstrate this holder´s capabilities through correlative research across multiple platforms, including TEM, SEM, and synchrotron X-ray techniques, providing researchers with unprecedented control and flexibility in studying dynamic processes across various techniques and environments.

Recent application examples will showcase how these systems, the "Lightning Arctic" and the new platform for "Climate" and "Stream" systems, are opening up new research frontiers across different application fields.

Combining in-situ synchrotron and electron microscopy techniques to study NiFe catalysts for CO_2 methanation

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To advance our understanding of catalysts and catalytic processes, direct observation is essential. Moving from traditional postmortem analyses to in-situ studies is therefore crucial. In recent years, in-situ electron microscopy has significantly enhanced our knowledge in this area, particularly for understanding dynamic structural and compositional changes during catalytic reactions [1]. The next step involves integrating in-situ transmission electron microscopy (TEM) with other in-situ techniques to bridge the length-scale gap and provide a more comprehensive view of catalytic behaviour [2].

In this study, we present findings on nickel-based catalysts, which are widely studied for CO2 methanation due to their high catalytic activity, natural abundance, and low cost [3]. However, Ni-based catalysts often deactivate [4] due to mechanisms such as carbon deposition, oxidation, sintering, and chemical poisoning. Recent research has focused on developing novel and modified Ni-based catalysts to enhance performance and durability in CO2 methanation. Additionally, studies have explored the effects of different oxide supports (e.g., CeO2, ZrO2, TiO2, MgO, and mixed oxides) on improving metal dispersion and catalytic efficiency.

We combined in-situ TEM gas-phase experiments with in-situ hard X-ray studies to investigate changes in the catalysts electronic structure during reduction and methanation. To realise this, we have constructed a specialised adapter [5] that allows us to use the same in-situ gas cell on our ARM200F TEM as well as on the spatially resolved i14 nanoprobe beamline (Diamond Light Source) enabling investigation under identical conditions. This approach allows for multi-length scale studies, linking the dynamic behaviour observed at the nanometer scale via TEM with micrometer-scale observations using nanoprobe XANES. Additionally, mass spectrometry data obtained during these experiments provide correlations between structural changes and catalytic activity.

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ABSTRACT SESSION V

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Time-resolved TEM

PROGRAM Wednesday, October 2nd

Mapping nanoscale near fields with attosecond electron microscopy

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Transmission electron microscopy (TEM) is indispensable for observing matter with extremely high spatial resolution down to the atomic scale. When combined with electron energy loss spectroscopy (EELS), it becomes possible to elucidate fingerprints of material composition and the optical properties of nanoscale structures [1]. However, EELS is limited to revealing the intrinsic properties of a sample, such as the electromagnetic near-field eigenmodes of a plasmonic nano resonator, with a spectral resolution defined by the electron microscope itself, typically in the sub-100-meV range for monochromated systems. This limitation arises because these intrinsic near-field modes are excited by the swift electrons, leading to subsequent spontaneous electron energy loss.

Recent approaches that introduce new technologies, such as pulsed-laser excitation, to conventional TEM allow for stimulated electron-light interactions which enables the study of extrinsic properties via photon-induced near-field electron microscopy (PINEM) and electron energy gain spectroscopy (EEGS). In these methods, ultrashort laser pulses excite specific polarization- and wavelength-dependent modes of a sample, and high-energy electron pulses interacting with the associated near fields undergo stimulated energy gain and loss [2]. These techniques enable the quantitative measurement of near-field strengths with nanometer spatial resolution [3] and allow for mapping the spectral response with sub-meV resolution [4] by tuning the wavelength of laser excitation. Moreover, the sensitivity of these methods to the optical phase provides access to the field evolution with sub-cycle time resolution [5,6].

In this presentation, I will provide an overview of the Göttingen UTEM project [7], where we are exploring the capabilities of ultrafast transmission electron microscopy for mapping and controlling optical near-fields [8]. In recent studies we have demonstrated mode-selective reconstruction of plasmonic near-fields [9] and introduced a free-electron homodyne detection (FREHD) scheme to measure the time evolution of these near fields with few-nanometer spatial and attosecond temporal resolution [6].

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Probing Phonons and Magnons using Ultrafast Electron Microscopy

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With recent advancement in meV EELS, there has been significant momentum in using spectroscopy methods to measure phonons and even magnons. In this presentation, I will demonstrate how ultrafast electron diffraction can be used to probe photoinduced phonon dynamics and track the evolution of optical and acoustic phonon populations in a charge-density-wave system.

Additionally, I will illustrate how a microwave-based imaging method can directly visualize magnon dynamics, including the generation, propagation, reflection, and interference of spin wave under RF-field excitation within a topological vortex and antivortex spin structure. These observations of phonon and magnon dynamics are not achievable using static EELS methods.

Attosecond Electron Microscopy

Peter Baum

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The primary step of almost any light-matter interaction is the electrodynamic response of the electrons in a material to the optical cycles of the impinging light wave on sub-wavelength and sub-cycle scales. To see such dynamics in space and time, we report here the advance of transmission electron microscopy to attosecond time resolution [1], combining the awesome spatial resolution of a transmission electron microscope with the awesome time resolution that is offered by the cycles of laser light. The idea is to use the oscillations of a laser wave to modulate the electron beam into a rapid sequence of electron pulses and then use an energy filter to resolve the electromagnetic near-fields in and around a specimen as a movie in space and time. We use an electron microscope at E0 = 183 keV and continuous-wave laser light with λ = 1064 nm. At the specimen, the electromagnetic near-fields appear frozen in time. The longitudinal fields shift the energy spectrum of the attosecond electrons towards a larger or smaller central energy, and the positive or negative energy sidebands become more or less intense, depending on position and arrival time. An imaging energy filter then selects only such electrons that have gained energy and thereby produces a movie of the optical cycles as a function of space and time. Figure 1b depicts one of our early results [1]. The specimen is a tungsten needle that is laser-excited from ~45°. The needle tip converts this excitation wave in part into a near-field surface wave that propagates along the tip and shaft. Indeed, the energy-filtered electron microscopy images reveal around the needle's surface a set of local regions with electron energy gain (white) and energy loss (black) as a function of time on attosecond dimensions. More results on chiral materials, inverse nanoantenna slits, metamaterials and chemically prepared mesocrystals show the general applicability of our novel attosecond electron microscopy to a large range of questions in contemporary research. These results establish attosecond transmission electron microscopy with field-cycle contrast as a versatile and sensitive method for visualizing the dynamics of light-matter interaction in complex materials on fundamental dimensions in space and time [2].

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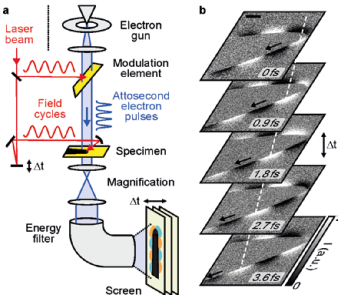

Figure 1. Attosecond electron microscopy. (a) Concept and experiment to see optical phenomena as a movie in space and time. (b) Field-cycle-contrast results at the example of a tungsten needle tip. Scale bar, 500 nm.

Ultrafast coherent manipulation of free electrons via quantum interaction with shaped optical fields for advanced imaging approaches

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The interaction between light and electrons can be exploited for generating radiation, or for controlling electron beams for dynamical investigation of materials, enabling new applications in quantum technologies and microscopy. In this contribution, I will describe an innovative method for coherent and versatile longitudinal/transverse manipulation of a free-electron wave function. Using appropriately shaped light fields in space and time, I will demonstrate how to modulate the energy, linear and orbital angular momenta, as well as spatial and temporal distributions of the electron wave function. The experiments have been performed in an ultrafast-TEM, where a pulsed electron beam was made to interact with shaped optical field generated via a spatial light modulator, and the energy-momentum exchange resulting from such interaction was directly mapped in the electron multidimensional phase space.

We will demonstrate how our approach for arbitrary longitudinal/transverse electron modulation at the sub-fs timescale is fundamental for the first time implementation of new imaging techniques, such as Ramsey-like holography and Single Pixel Imaging. Our results would pave the way for achieving unprecedented insights into non-equilibrium phenomena in advanced quantum materials, playing a decisive role in the rational design and engineering of future photonics and electronics application.

References

SMART-electron project that has received funding from the EU Horizon 2020 Programme under GA No 964591.

JEOL Recent Trials for Scanning Transmission Electron Microscopy

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Scanning Transmission Electron Microscopy (STEM) is the function hard to replace for Transmission Electron Microscope (TEM). Using STEM, local information is able to be obtained with higher resolution and therefore the applications become wider field year by year. We JEOL proceed the developments of STEM technology about optical conditions and scanning methods.

In this presentation, JEOL optical trials are introduced about magnetic field free STEM condition and nano beam diffraction STEM. In addition, shutter synchronized scan and high-speed scanning system are introduced as improved scanning methods, too.

Regarding magnetic field free STEM, JEOL has developed Magnetic field free Atomic Resolution STEM(MARS). The spatial resolution of MARS is sub-angstrom and it is possible to obtain atomic resolution magnetic information [1]. On the other hand, magnetic field free STEM observation is possible with normal TEM turning off the objective lens (OL). In this case, the condenser lens near OL, we call this lens condenser mini lens (CM), is used as focus lens. Spherical aberration corrector is able to adapt to this condition and it improves the spatial resolution. However, the focal length of CM is longer than the length from CM to the first upper cross over point, and this limits the spatial resolution of magnetic field free STEM with normal TEM. This optical condition increases the effect of chromatic aberration from other condenser lenses, because this is magnified optical system and the de-magnification is smaller.

JEOL developed higher order spherical aberration corrector, it is named DELTA corrector, in 2010 for low acceleration voltage and in 2018 for 300kV. This corrector could overcome the de-magnification issue of OL off condition. The feature of this corrector is using three hexapoles, and the aberration corrected area of Ronchigram is over 70mrad with turning on OL condition. On the other hand, it is possible to correct aberration with only two hexapoles in three.

It is possible to realize expanding the length between CM and first upper cross over point and correction of spherical aberration simultaneously with upper two hexapole spherical aberration correction. In this result, 0.5nm spatial resolution was achieved with OL off condition.

This two hexapole correction system is able to be applied to not only magnetic field free observation but nano beam diffraction. Using the lower two hexapole spherical aberration correction, it realizes convenient nano beam diffraction condition.

On the other hand, irradiation damage of electron beam is one of issues for STEM. Electron dose control is effective to avoid the sample damage. JEOL and IDES developed the synchronized scanning system with an electrostatic shutter, it is named Synchrony. Synchrony can control the dose rate at each scanning pixels and can blank the beam during the scan fly-back time with high speed shuttering rate control. Synchrony system excludes the unnecessary irradiation and change the beam dose without changing optical condition. In this talk, tempo system, which is developed by IDES and Trinity college Dublin, will be introduced.
In addition, time-resolved imaging was also dis-advantage of STEM, however it was being overcame with high speed scanning system which was developed by Tokyo university and JEOL [2]. Time-resolved imaging with fast acquisition is a crucial technology for dynamic observation with transmission electron microscope, and has been used in various studies due to the high readout speed of cameras with transmission electron microscopy. However, distinguishing the elements of atoms through phase contrast in TEM remains challenging, and information about element species through image contrast is beneficial in understanding single-atom dynamics of atom clusters on carbon film, dopants in crystals, and other related areas. On the other hand, detecting metallic atoms on a carbon film and in crystals through annular dark-field scanning transmission electron microscopy (ADF-STEM) is reasonably feasible by utilizing the high signal-to-noise (S/N) ratio of single atoms compared to the intensity of carbon atoms or crystals. Therefore, time-resolved observation with ADF-STEM is preferred for investigation the dynamics of atomic motion.

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Microsecond Time-Resolved Cryo-EM

Ulrich Lorenz

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While cryo-electron microscopy (cryo-EM) is rapidly gaining in popularity, its time resolution is frequently insufficient to directly observe proteins in action, leaving our understanding of these nanoscale machines fundamentally incomplete. My group has recently introduced a novel approach to time-resolved cryo-EM that improves its time resolution by about 3 orders of magnitude.

Our method involves melting a cryo sample with a laser beam, which allows dynamics of the embedded particles to occur in liquid once a suitable stimulus is provided. While the dynamics occur, the heating laser is switched off at a well-defined point in time, causing the sample to rapidly recool, so that it vitrifies. The particles are thus trapped in their transient configurations, in which they can subsequently be imaged. We demonstrate that our approach affords a time resolution of 5 µs or better. Moreover, near-atomic resolution reconstructions can be obtained from revitrified samples, showing that the revitrification process leaves the protein structure intact.

Finally, I will present a microsecond time-resolved pH jump experiment, in which we observe the dynamics of the capsid of CCMV, an icosahedral plant virus. These results highlight the potential of our method to fundamentally advance our understanding of protein function through direct observation of the dynamics involved.

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ABSTRACT SESSION VI

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Theory of Microscopy and Modeling of Materials

PROGRAM Thursday, October 3rd

Ferroic materials – designed and probed using electrons

Guodong Ren[1], Gwan Yeong Jung[1], Huandong Chen[2], Boyang Zhao[2], Xin Li[3], Pravan Omprakash[1], Xiaoshan Xu[3], Jayakanth Ravichandran[2], Rohan Mishra[1]

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Advances in supercomputing capabilities and electronic structure calculations based on density-functional theory (DFT) now make it possible to design materials with new properties starting from the atomic scale — that can be used to guide synthesis experiments. Concurrent advances in scanning transmission electron microscopy (STEM) enable imaging and spectroscopy of materials with unprecedent spatial and energy resolution. Naturally then, the combination of theory and microscopy provides an unparalleled probe to unravel the atomic-scale structure-property correlations in complex materials. In this talk, I will give two examples of combining DFT and STEM to: 1) unravel the origin of ferroelectricity in nanoscale hafnia,[1] 2) and realize a relatively understudied class of multiferroic that combines ferroelectricity and chirality.[2]

The discovery of ferroelectricity in hafnia-based materials offers the potential of incorporating ferroelectrics with silicon. However, comprehending why these materials defy the common trend of reduced ferroelectric ordering at the nanoscale, and the mechanism that stabilizes the ferroelectric phase — that is absent in the phase diagram — presents significant challenges to traditional knowledge of ferroelectricity. I will show that the formation of the orthorhombic ferroelectric phase of the single-crystalline epitaxial films of 10% La-doped HfO2 (LHO) on (111)-oriented yttria stabilized zirconia (YSZ) relies on the stability of the high-pressure orthorhombic antiferroelectric phase. Our detailed structural characterization demonstrate that as-grown LHO films represent largely the o-AFE phase being thermodynamically stabilized by the compressive strain. The o-AFE phase can then be converted to the o-FE phase using mechanical or electrical poling. We find that the orthorhombic phase stability is enhanced in thinner films down to one-unit-cell thickness, a trend that is unknown in any other ferroelectric films. This is due to the vanishing depolarization field of the o-AFE phase and the isomorphic LHO/YSZ interface, supporting strain-enhanced ferroelectricity in the ultrathin films.

Ferroaxial materials have ordered rotational structural distortions that break mirror symmetry and induce chirality. When ferroaxial order is coupled with ferroelectricity arising from a broken inversion symmetry, it offers the prospect of electric-field-control of the ferroaxial distortions and opens up new tunable functionalities. I will discuss the discovery of a strain-stabilized, room-temperature chiral ferroelectric phase in single crystals of BaTiS3. Using DFT calculations, we predict the stabilization of this multiferroic phase for biaxial tensile strains exceeding 1.5% applied on the basal ab-plane of BaTiS3. We used an innovative approach using focused ion beam milling to make appropriately strained samples of BaTiS3. The ferroaxial and ferroelectric distortions, and their domains in the chiral ferroelectric were directly resolved using atomic resolution STEM. Landau-based phenomenological modeling predicts a strong coupling between the ferroelectric and the ferroaxial order making BaTiS3 an attractive test bed for achieving electric-field control of chirality-related phenomena.

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From modelling to generation: EELS and Materials Development

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Electron Energy Loss Spectroscopy (EELS) observed using scanning transmission electron microscopy has the potential to identify material properties at the sub-nanometer scale. For instance, we have achieved nanometer-scale thermal expansion measurements at the SrTiO3 interface and revealed its relationship to the interface structure [1], as well as the distribution of 4-fold/5,6-fold coordinated Al inside SiO2-Al2O3 glass [2].

To achieve such microscopic analysis, suitable simulations and modeling of materials are crucial. Recently, we developed a Python code, "interface_master," that can construct bicrystal models of interfaces consisting of any two lattices, including tilt, twist, mixed, and hetero interfaces and surfaces [3]. This code includes a function to calculate the cell of non-identical displacement (CNID), which dramatically reduces the number of candidate structures in rigid body translation states [4].

In addition to modeling, applications of machine learning to EELS have been reported. In particular, we have aimed to transcend the traditional physics of spectrum generation through machine learning. First, we constructed a spectral database comprising more than 100,000 Carbon-K edges of organic molecules [5]. Using this database, we achieved the prediction of extensive properties, such as molecular weight and internal energy, which are typically considered unrelated to ELNES features, via machine learning [6].

We have also attempted to extract valence band information, similar to that obtained from XPS, from the ELNES features [7,8]. Generative AI is also becoming a powerful tool for materials development and analysis [9,10].

In my presentation, I will discuss the modeling and application of machine learning and generative AI for EELS and materials development.

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Evolution of the Atomic Structure of 2D Materials under Electron Beam in a TEM: Insights from First-Principles Calculations

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Due to their very geometry, 2D materials, such as graphene or single sheets of transition metal dichalcogenides are the ideal systems for the atomic structure characterization using transmission electron microscopy (TEM).

Indeed, numerous TEM studies provided insights into morphology of these systems and revealed that all these materials contain defects and impurities, which frequently govern their electronic and optical properties. Moreover, defects can also appear due to the impacts of energetic electrons, and the formation of defects may give rise to phase transformations in these materials and affect their characteristics.

All of these calls upon the studies on defects in 2D materials and mechanisms of their formation under electron irradiation. In my talk, I will present the results of our recent theoretical studies on how point and line defects appear in 2D materials under electron beam irradiation [1,2] in a TEM obtained in collaboration with several experimental groups. I will also touch upon electron beam-induced transformations in the atomic structure of 2D materials [3-6].

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A Comparative Analysis of Iterative and Non-Iterative Electron Ptychography

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The development of fast pixelated detectors for scanning transmission electron microscopy (STEM) has provided a wealth of opportunities for imaging light and heavy elements with unprecedented contrast and resolution, primarily via electron ptychography. While the first experimental demonstrations of ptychography incorporated non-iterative, analytical techniques for electron microscopy data (Figure 1(a)) [1], many recent STEM ptychography routines have been based on coherent diffractive imaging techniques initially developed for X-ray illumination (Figure 1(b)) [2]. The latter class of methods use iterative numerical approaches, such as gradient descent, to converge on a faithful solution for the object (i.e., the sample) and the probe. Concurrently, non-iterative methods such as single sideband and Wigner distribution deconvolution are still frequently used in STEM, providing quantitative atomic-resolution imaging of light and heavy elements for sufficiently thin samples [3]. However, to date, there have been few efforts to discuss the relationship between non-iterative and iterative methods beyond the visual comparison of their results.

Here, we will compare and contrast iterative and non-iterative techniques for STEM ptychography, as briefly outlined in Table 1, using 2D materials as example systems [3,4]. We will discuss the key equations, experimental requirements, and computational workflows for each class of methods, and summarize the capabilities of each reconstruction technique for materials science applications. Finally, we will outline new methods based on deep learning that aim to improve and simplify the process of ptychographic reconstruction [5].

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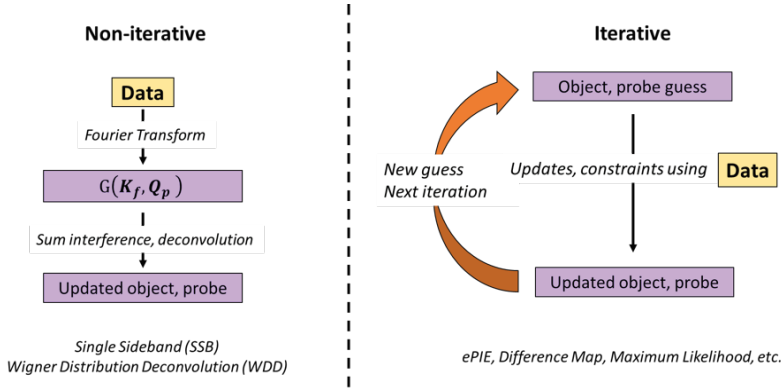

Figure 1. Simplified workflows for (a) non-iterative and (b) iterative electron ptychography.

	Non-iterative	**Iterative**
Sampling	Very restrictive in real space	Flexible, overlap-dependent
Overlap	Reciprocal space	Real and reciprocal space
Resolution	Typically ~2α, can be extended	~Maximum diffraction angle
Low-dose?	Same workflow as high-dose	Regularizations often used
Probe geometry	Focused or defocused	Focused or defocused
Thickness	Phase-object approximation	Beyond phase-object
Depth sectioning	Possible, limited to thin samples	Possible, regularization used

Table 1. Comparison of the requirements and capabilities of non-iterative and iterative electron ptychography.

Simulations of phonon and magnon EELS/EEGS including dynamical effects and multiple inelastic excitations

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Phonons and magnons play important roles in various technological applications. Phonons are essential for understanding heat flow at the nanoscale, while magnons hold promise as an information carrier (quasi-)particle for a new generation of hardware in information technologies. Instrumental advances, bringing high spatial resolution offered by scanning transmission electron microscopes (STEMs) together with high energy resolution and high dynamic range offered by direct electron detectors, offer an exciting route for studying these quasiparticles down to atomic scale [1-2].
However, the strong interaction between the electron beam and the sample, resulting in multiple elastic and inelastic scattering events, complicates the interpretation of experimental data and reliable theoretical predictions are thus of high importance.
We extend the quantum excitation of phonons method [3] to the spectroscopic domain. This is achieved by deriving a time-autocorrelation function of the so-called auxiliary wave-function and showing that it represents the desired spectral information. The exact quantum-mechanical evolution of the auxiliary wave-functions is approximated by scattering on a structure model with the atomic coordinates (or magnetic moment directions in the case of magnon EELS) evolving classically. Motion of atoms or precession of magnetic moments is simulated by molecular dynamics or atomistic spin dynamics, respectively.
This new theoretical framework treats elastic and inelastic scattering on equal footing, including energy loss and gain processes, and effects of multiple elastic and inelastic scattering events [4].
The resulting simulation method remains flexible and efficient, and in comparison with recently introduced frequency-resolved frozen phonon multislice method (FRFPMS; [5]) it includes multiple inelastic scattering, doesn't require knowledge of Debye-Waller factors, and effectively includes anisotropic vibrations (or precession of magnetic moments) and non-local absorption effects of the elastic channel. We present the method on simulations of phonon EELS/EEGS on crystalline silicon and magnon EELS/EEGS for bcc iron and compare them to available data.

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FEMMS 2024
Frontiers of Electron Microscopy and Materials Science

ABSTRACT SESSION VII

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Artificially Intelligent Imaging and Spectroscopy Powerful Trustworthy and Sustainable

PROGRAM Thursday, October 3rd

Using TEM Data to Understand Bias in Machine Learning Algorithms

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Machine learning and artificial intelligence techniques have emerged as extremely powerful data science tools that can potentially automate information extraction out of large TEM datasets with better accuracy and speed. However, current state-of-the-art data-driven methods are notorious "black boxes" which can be dangerous for scientific analysis. This "black-box" nature makes it difficult to justify results, for example distinguishing between physically plausible features or a data artifact, and it is unclear under what conditions we might expect these mistakes to happen. Alternatively, we can use our domain knowledge of our scientific datasets to identify potential sources of bias in our machine learning pipelines and aid in our interpretation of these models.

In this talk, I'll discuss how curated TEM datasets have enabled us to explore how choices in data representation and model architecture can bias the output of a machine learning model in the context of nanoparticle characterization. First, we show how data preprocessing, or the conversion of raw data into a form suitable for algorithm input, affects machine learning model performance in both nanoparticle identification and shape analysis in TEM images. By understanding and mitigating for known biases in the raw data, we are able to extract out subtle changes in nanoparticle shape over large populations and compare neural network robustness across various experimental conditions. Next, we show how neural network model architecture features, namely the receptive field, can affect model performance. By combining systematically varied neural network architectures with curated datasets, we find that a large receptive field is needed for identifying low contrast nanoparticle features. Overall, our results point towards the need for increased analysis on the data-inputs of our data-driven algorithms, and for further quantitative studies on the robustness of our trained models.

Fused Multi-Modal Electron Tomography for 3D Chemistry at Lower Dose and Higher Resolution

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For electron tomography, dose and resolution are inextricably linked—the maximum achievable 3D resolution is ultimately limited by the total allowable dose. Higher 3D resolution demands more specimen projections [1,2] and exceedingly high SNR [2,3,4]. This makes high-resolution chemical tomography from inelastic scattering intractable. The inelastic scattering signals from chemically sensitive core-electron interactions are typically one-hundred times smaller than high-angle elastic scattering signals hitting an annular dark field (ADF) detector. For any specimen, one can expect 3D chemical resolution to be five to ten times worse than ADF tomography.

Here we show fused multi-modal electron tomography is the essential to enabling 3D chemical imaging at higher theoretical resolutions and dramatically reduced dose. Fused multi-modal electron tomography notably improves both the sampling and dose constraints that limit resolution across a range of radiation sensitive materials [5]. We experimentally demonstrate fused multi-modal tomography by imaging at resolution < 1 nm in a beam sensitive material (Fe/Au embedded in Polystyrene-based ligands). The novel technique is theoretically, computationally, and experimentally validated.

We emphasize three innovations for chemical electron tomography when utilizing multi-modal data fusion: (1) By linking the physics of elastic Rutherford scattering and inelastic core-loss scattering, dose can be reduced by upwards of two orders of magnitude. This addresses the key limits to 3D chemical imaging at high-resolution or on beam-sensitive materials.

(2) Mixed inelastic and elastic signals can be sampled independently to break traditional 3D resolution limits for chemistry. (3) Chemical stoichiometry can be retrieved with higher precision and do so without knowledge of elemental inelastic cross-sections. These aspects have been discussed theoretically and demonstrated experimentally [5].

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Reward-Driven Decision Making in Automated Microscopy

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The integration of machine learning (ML) in electron microscopy is poised to revolutionize the field, the development enabled by rapid cloudification of existing experimental tools and broad adoption of edge control enabled by Python APIs. In this presentation I introduce several levels of ML applications. These start with post-acquisition data analysis to real-time data analysis to inform human decisions, to active ML agents implementing decisions on active instrument, and the more general incorporation of microscopy into experimental and theoretical workflows from sample selection to theory updates.

I will further discuss the challenges associated with the transition from real-time data analysis to ML decision-making during active microscopy experiment. We propose that this transition requires the definition of reward functions, which need to be seamlessly integrated across various domains. Should these reward functions be universally established, the entirety of experimental efforts could be conceptualized as optimization problems. Here, we identify several categories of reward functions that are discernible during the experimental process, encompassing image optimization, fundamental physical discoveries, the elucidation of correlative structure-property relationships, and the optimization of microstructures.

The operationalization of these reward functions on autonomous microscopes is demonstrated, as well as the need and strategies for human-in-the-loop intervention. This approach further allows for building the framework for incorporation of microscopy in materials discovery and optimization workflows. The very tempting opportunity this research opens is further use of the LLMs for creation of probabilistic reward functions.

Machine Learning Enabled Advances in Lorentz Microscopy

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Non-trivial, nanoscale magnetic spin textures such as skyrmions, and merons are of continued interest for both understanding their fundamental physical properties, and controlling them for potential use in next-generation spintronic applications [1]. In order to gain this understanding, it is critical to study both the microstructure and magnetic domain structure simultaneously to determine the fundamental role of inhomogeneities in microstructure as well as the effect of shape and size of nanostructures on the magnetic domain behavior. Moreover, observation of domain behavior using in situ experiments under external stimuli such as temperature, electromagneti fields and currents can provide information about the underlying energy landscape.

Lorentz transmission electron microscopy (LTEM) is ideally suited for quantitative analysis of magnetic domains at the nanometer length scale. The state of the art LTEM using aberration correctors allows for imaging down to sub-nanometer scale in field-free conditions. In order to obtain the quantitative information about the magnetization of the sample, it is essential to retrieve the phase shift of the electrons. This can be obtained using various methods such as in-line or offaxis electron holography, and more recently using 4D-STEM approaches. In this talk, we will present our recent advances in phase retrieval for LTEM using neural-network based approaches. Using the forward model for the image formation, we will demonstrate the application of automatic differentiation to solve for the phase from a through-focus series of images and show that we are able to achieve a higher phase sensitivity and spatial resolution than previous techniques [2] as well as extension of the method to single image phase retrieval [3]. Besides these approaches, we will also present our work on using machine-learning methods for fast, statistically significant analysis of magnetic spin textures such as skyrmions from in situ experiments [4]. Such analysis can give further insight into their ordering behavior and skyrmionskyrmion interactions.

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Materials dynamics *in situ*, from nanoscale phase changes to motion within individual atomic columns

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Building functional materials with atomic level precision requires a detailed understanding of the processes that control crystal growth and phase transformations at the nanoscale. In situ experiments in the transmission electron microscope offer a unique view of these phenomena and a powerful opportunity to quantify and control their outcomes.

We will describe examples of phase transformations and growth processes visualized when nanoscale materials are exposed to a reactive gas or liquid environment. Reactions can be driven with an even higher degree of spatial control by stimulating local excitation with the electron beam. We will show how beam targeting at the tens of picometer level, achieved through algorithms that mitigate the effects of drift and scan distortions, allows dynamics to be measured for individual atomic columns of a crystal.

The resulting local structural transformations may offer a pathway to tailored defect arrays with electronic properties relevant to quantum computing applications. We anticipate that continued advances in control of the sample environment and electron dose will open exciting future prospects for developing new materials with the help of in situ electron microscopy.

FEMMS 2024
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ABSTRACT SESSION VIII

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Frontiers in High Resolution Electron Spectroscopy

PROGRAM Friday, October 4th

Single-Atom Spectroscopy in the meV to keV Energy-Loss Regime

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The possibility of performing real-space imaging and spectroscopy analysis at the atomic scale via low-voltage aberration-corrected scanning transmission electron microscopy (STEM) has provided exciting new opportunities for the study of two-dimensional materials, especially for defects and interfaces. Pushing the sensitivity of STEM spectroscopy techniques down to the single atom level is expected to open a new avenue for probing the local functionalities of materials, but remains challenging. This grand challenge lies in the extremely low scattering cross-section for signals in the keV energy-loss regime, while a combination of low current for monochromated electron probe and low scattering cross-section forms the main obstacle for single-atom spectroscopy at the meV regime.

In this presentation, I will discuss our recent efforts on pushing the sensitivity of single-atom spectroscopy technique in both the meV and keV energy-loss regime using dopants in monolayer graphene as a model system. With this relatively stable single-atom model system, we can explore the ultimate sensitivity of EELS at 60 kV where the dose level is no longer the limiting factor, and the above-mentioned technical challenges could be partially overcome with state-of-the-art direct electron detectors, better electron optics and advanced signal processing algorithms. We show that the sensitivity of single-atom vibrational spectroscopy analysis can be pushed to the chemical-bonding level and this technique could be applied to explore local vibrational signatures at defects and interfaces in 2D materials [1]. Using single-atom vibrational spectroscopy, we show that it is now feasible to perform atom-by-atom isotope mapping in monolayer graphene by precise measurement of the peak frequency of the optical phonon modes. As for core-loss excitations, we show that electronic states contributed by specific unoccupied pz orbital around a four-fold coordinated Si point defect in graphene can be mapped out using atomic-resolution energy-loss near-edge fine structure (ELNES) spectroscopy [2]. In addition, local coordination measurement can be achieved with single-atom sensitivity via extended energy loss fine structure (EXELFS) analysis at the keV energy-loss regime.

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Advanced STEM-EELS for materials discovery: challenges and opportunities

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Many aspects of modern materials discovery are driven by understanding and controlling chemical interactions and electronic bonding at the atomic scale. For instance, locacompetition with competing order parameters such as lattice structure can be leveraged to engineer new properties or emergent phenomena, especially in heterostructures, thin films, or other phase-inhomogeneous geometries. The ability to locally probe elemental, chemical, and electronic states down to the atomic scale therefore plays a key role in this kind of materials design and discovery. In many cases, for example, bulk spectroscopic probes are not suitable for small sample volumes or spatially varying signals, as we demonstrated in the case of recently discovered superconducting nickelate thin films [1,2]. In other systems, exotic phases may be stable only at low temperatures or in meta-stable compounds with low damage thresholds in the high energy electron probe, both of which necessitate working under “signallimited” conditions [3, 4].

In all cases, carefully designed STEM-EELS experiments to harness recent developments in hardware and data analysis are required to uncover and thus inform new atomic-scale materials design. Here, I will present examples of our recent work leveraging a combination of high spatial- and energy-resolution STEM-EELS measurements across a variety of novel quantum materials systems, including superconductors, low-dimensional magnets, and metal-insulator systems. In parallel, I will highlight key advances in instrumentation (including detectors, electron source, and spectrometer) as well as in data processing which expand our access to new materials and phases.

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Exploring the dynamics of semiconductors with an ultrafast transmission electron microscope

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The development of time-resolved Cathodoluminescence (TR-CL) in a scanning electron microscope has enabled the measurement of the lifetime of excited states in semiconductors with a sub-wavelength spatial resolution [1]–[3]. The development of time-resolved Cathodoluminescence (TR-CL) in a scanning electron microscope enabled the measurement of the lifetime of excited states in semiconductors with a sub-wavelength spatial resolution. It was used for example to measure the influence of stacking faults on the GaN exciton [1], to probe the role of a silver layer on the dynamics of a YAG crystal[2] or to show the influence of stress on the optical properties of ZnO nanowires [3]. These results demonstrate that TR-CL is essential to study the correlation between semiconductor optical and structural properties (composition, defects, strain...). While TRCL is usually done in a scanning electron microscope, the improvement of the spatial resolution and the combination with other electron-based spectroscopies offered by transmission electron microscopes has been a step forward for TR-CL [4], [5].

Indeed, we recently succeed to do the first time-resolved cathodoluminescence experiments within an ultrafast transmission electron microscope [4], followed soon after by Ye-Jin Kim et al [5]. Our TRCL experiment are performed in a unique electron microscope, based on a cold-FEG electron gun [6]. This technology allows among other things to reach a spatial resolution of a few nanometers, essential for the study of III-N heterostructures.

In this presentation we will discuss for example the advantage and inconvenient of TRCL in a UTEM and present our results on the study of charge carrier dynamics in In0.3Ga0.7N/GaN quantum well with a resolution below 10 nm. Comparing different heterostructure we will discuss the impact of growth condition on the optical properties (spectral and carriers dynamics). We will study the QW emission dynamic both along and across the quantum well and correlate the results with strain map and high resolution HADF images.

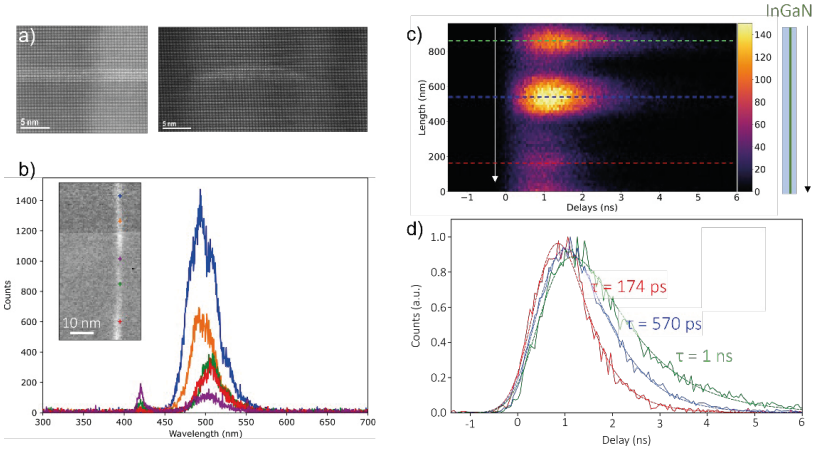

Figure 1. a) STEM-HADF image of an In0.3Ga0.7N/GaN quantum well. b) Cathodoluminescence spectra at different position of along the quantum well. EHT = 100 keV at nitrogen temperature. c) Decay map along the quantum well with different decay depending on the position of the electron beam. d) Decay trace extracted from c) with the corresponding lifetimes extracted using a mono-exponential model.

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Probing Nanoscale Light-Matter Interactions Using Fast Electrons

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To reach next-generation photonics applications, we need to improve our control over the interactions between light and matter at the smallest possible length scales. One route towards this is through strong light-matter interactions that occur when there is coherent energy exchange between an electromagnetic mode and a dipolar excitation within matter. When the interaction becomes strong enough, two new coupled modes called polaritons are created which have fascinating inseparable light and matter properties[1]. Polaritons have historically been studied at low temperatures using optical microcavities, but recently the field has been extended to nanoscale systems at room temperature by creating, for example, plasmon-exciton polaritons (plexcitons)[2]. Additionally, there is interest in optically dark scattering states as an alluring route towards enriching traditional nanophotonics approaches. For example, optical anapole states are created by the superposition of electric and toroidal Mie-like modes in high index dielectric materials and are characterized by vanishing scattering due to destructive interference in the far-field[3]. Optical based characterization methods have made significant advances in understanding plexciton and anapole physics, but they have significant shortcomings, including their inability to probe their characteristic nm length scale. In this work, we use experimental and theoretical monochromated STEM EELS to spatially and spectrally resolve strong coupling phenomenon. By synthesizing a hybrid system composed of Ag truncated nanopyramids coupled to the 2D TMD material WS2, we generate plexcitons, resolve their anticrossing behavior, and spatially map them to reveal unexpected nanoscale variations in their behavior[4]. Additionally, by nanopatterning WS2 nanodiscs, we confirm EELS can measure Mie-like modes, anapole states, and anapole-exciton self-hybridization. By spatially mapping these WS2 nanodisc modes and states, we find that their excitation can be selectively controlled by placing the electron beam at different positions on the nanodisc[5]. Lastly, we explore the possibility of using STEM EELS to image and characterize slab waveguide modes in TMD materials. These results demonstrate that STEM EELS is a powerful technique for expanding our knowledge of strong light-matter interactions beyond what is accessible using optical spectroscopy. Our results also provide an avenue for enhancing light-matter interactions using dark scattering states like anapoles, which can now be accessed by electron microscopy.

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Correlating emission, absorption, and composition in perovskite nanocrystals

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Lead halide perovskite (LHP) materials, a rapidly improving class of photovoltaic materials, are also prized for bright, narrow-band emission for light emitting diode applications [1], arising from strongly excitonic response at room temperature [2]. In fully inorganic LHPs, of the type CsPbX3, the emission energies are tuned by mixed anion composition [3], with mixed CsPb(Cl1-xBrx)3 suitable for highly sought after blue emitters. However, optical measurements of these mixed halide nanocrystals exhibit contributions from a heterogeneous ensemble, complicating attribution of variations in composition, absorption, and emission. This presentation will report the use of correlative cathodoluminescence (CL), electron energy loss spectroscopy (EELS) at the bandgap energy, and X-ray energy dispersive spectroscopy (EDS) for the analysis of optical properties from individual LHP nanocrystals. We selected a composite material comprising LHP nanocrystals within a metal–organic framework (MOF) glass, exhibiting bright, narrow-band photo- and cathodoluminescence (100 kV, liquid nitrogen cooled) from <50 nm CsPbX3 [3,4]. Preparation as a focused ion beam (FIB) lamella further supports low-loss EELS followed by X-ray EDS for crystal-by-crystal analysis of the emission peak, the optical onset in EELS, and the composition across >30 nanocrystals ***(Figure 1)***. Excitonic peaks were preserved in both EELS and CL in these materials, enabling relative Stokes shift analysis. Through automated curve fitting, we recovered trends in the bandgap energy, emission energy, and composition as well as in the Stokes shift and the CL emission linewidth, offering previously unobservable experimental structure–function relationships in LHP nanocrystals for advancing design of LHP-based optoelectronic technologies.

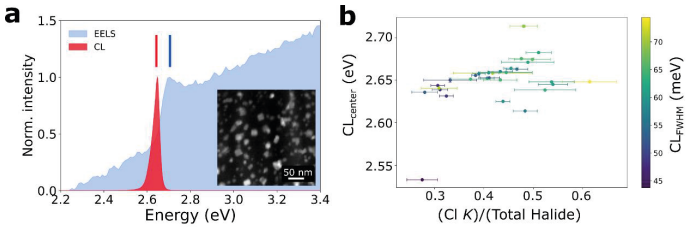

Fig. 1. a, Correlated EELS and CL from a single nanocrystal within a MOF-CsPb(Cl1-xBrx)3 crystal-glass composite. The inset shows an example annular dark field STEM image of the FIB lamella. *b*, Correlation between Cl content (halide intensity fraction) and CL emission energy.

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Atomic-Level Visualizations of Frequency-Dependent Phonon Eigenvectors and Symmetry-Induced Vibrational Anisotropies

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Phonon anisotropy gives rise to the orientation-dependent variations of dielectric, optical, and thermal properties of materials [1]. At the atomic level, the vibration of individual atoms, known as thermal ellipsoids, could also exhibit strong anisotropies due to lowered crystal symmetry and reduced local point group symmetry. Thermal ellipsoids are traditionally estimated by diffraction methods [2], which encountered critical shortcomings of lacking both spatial and energy resolutions. The missing frequency-linked information is crucial for understanding their thermal and photonic responses. The state-of-the-art monochromated electron energy-loss spectroscopy (EELS) reaches a combination of few-meV energy resolution and sub-nm spatial resolution and enables the detection of exotic vibrational states emerging at various crystalline imperfections [3, 4]. However, the functionalities such as distinguishing diverse elements, crystalline positions, or atomic vibrational anisotropies have not been fully explored. In this talk, I will first introduce the recent progress in this burgeoning field of vibrational EELS, and then present our new results on observing atomic level phonon eigenvectors and revealing atomic vibrational anisotropies. We developed a novel dark-field EELS technique with desired momentum exchanges and applied it to both centrosymmetric and non-centrosymmetric perovskite oxides. The energy-filtered vibrational signals contain distinct atomic features of different elements in different energy ranges, consistent with simulated results. By varying the momentum exchanges, we also discerned two types of oxygen atoms exhibiting contrasting vibrational anisotropies below and above 60 meV for a cubic phase SrTiO3 due to their frequency-linked thermal ellipsoids. The reduction of crystal symmetry such as tetragonality can further produce an unexpected modulation of thermal ellipsoids between apical and equatorial oxygen atoms near certain energy ranges. This method establishes a new pathway to visualize phonon eigenvectors at specific crystalline sites, thus delving into uncharted realms of dielectric, optical, and thermal property investigations with unprecedented spatial resolutions [5].

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FEMMS 2024
Frontiers of Electron Microscopy and Materials Science

POSTERS OVERVIEW

POSTER	TITLE
1	Characterization of Aberration-Corrected Lorentz TEM Applying a Magnetic Field with Objective Lens - **Yu Jimbo**
2	Enhanced Analysis of Hyperspectral Analytical Scanning Transmission Electron Microscopy Data through Blockwise Processing - **Sebastian Cozma**
3	New Method for Calibrating Electron Spectrometers with μeV precision - **Thomas LaGrange**
4	Probing the Itinerant Magnetic Behavior of FeRh using In-situ Electron Magnetic Circular Dichroism (EMCD) and Lorentz TEM - **Thomas LaGrange**
5	Enhanced time resolution with a room-temperature energy dispersive X-ray PIN photodiode detector - **Luca Serafini**
6	Quantification and Denoising of Atomic-Resolution EDX Spectrum Images Through a Multiscale Bayesian Approach - **Pau Torruella**
7	Overcoming the aberration-limit of a non-corrected Transmission Electron Microscope with computational ghost imaging - **Paolo Rosi**
8	What is the width of a grain boundary, and how to determine it? - **Klaus van Benthem**
9	Physics-based scan distortion correction in hardware - **Benjamin Bammes**
10	Atomic-Scale Characterization of Monolayer Amorphous Materials Using Advanced Low-Voltage Electron Microscopy - **Ang Li**
11	Emission characteristics and stability of the LaB_6 nanoneedle emitter - **Kanako Noguchi**
12	Edge State Enigma: An EELS Detective Story - **Frances Allen**
13	Development and Applications of a High-Energy Electron Energy-loss Spectrometry System in an Aberration-Corrected Scanning Transmission Electron Microscope - **Masashi Watanabe**
14	Streamlining Graphene Liquid Cell Preparation: VitroTEM's Naiad System - **Hans Radhoe**
15	Event-responsive Beam-modulated STEM with Multi-frame and Sparse Scanning - **Lewys Jones**
16	Technological Advances for Temperature-Dependent operando Electrochemical Studies within the Transmission Electron Microscope - **David Nackashi**
17	How to tackle the most sophisticated materials science questions with advanced TEM: towards reliable investigations of materials in their native state - **Maria Meledina**
18	*In situ* biasing DPC STEM observation of p-n junction depletion layers - **Yoshifumi Kojima**
19	Towards Atomic-Resolution Electron Energy Loss Spectroscopy (EELS) in an Uncorrected 30kVScanning Electron Microscope - **Demie Kepaptsoglou**
20	Electron beam effects in monolayer 2D materials from knock on damage and inelastic scattering effects to chemical etching - **Jani Kotakoski**

POSTER	TITLE
21	Novel scan coil design for high spatiotemporal-resolution imaging in the scanning transmission electron microscope - **Adam Phipps**
22	Bridging the gap: beam-induced structuration between crystalline interface **Chih-Ying Hsu**
23	Microstructural characterization of aging-treated aluminum alloy AA2024 with SEM, STEM, EDS, and EBSD - **Sandra Rontree**
24	Chemical and structural characterization of chalcogenide nanostructures for molecular recognition through controlled electron dose transmission electron microscopy - **Federica Florio**
25	Camera Systems and Software for Fast 4DSTEM Data Acquisition and Live Virtual Detector Calculation - **Hans Tietz**
26	In-situ characterization of the high-temperature spin-state transition in LaCoO3 via STEM imaging and EELS - **Michelle A. Smeaton**
27	Revolutionizing Nanoscale Characterization with TESCAN TENSOR's Analytical 4D-STEM - **Tomáš Morávek**
28	Unveiling rapid nanoscale dynamics in copper catalyst during HOR and CO_2 hydrogenation reactions - **Georgian Melinte**
29	Atomically Sharp Domain Walls in an Antiferromagnet - **Jan Michalička**
30	SOLARIS Cryo-EM Facility at SOLARIS National Synchrotron Radiation Centre, Kraków, Poland - **Paulina Indyka**
31	Consistency and reliability of different ptychographic deconvolution approaches - **Tizian Lorenzen**
32	Thickness-Dependent Behavior of Magnetic Domains in van der Waals Fe_3GeTe_2 during Magnetization Reversal - **Jennifer Garland**
33	Atomic scale mapping of charge and lattice coupling at polar $LaAlO_3/SrTiO_3$ oxide interfaces by vibrational EELS - **Roger Guzman**
34	In situ study of the 2D Electron Gas in p-GaN/AlGaN/GaN High Electron Mobility Transistor - **Gianfranco Sfuncia**
35	Linear dichroism in a scanning transmission electron microscope - **Jaume Gazquez**
36	Outrunning Crystallization During Flash Melting and Vitrification: Determining the Critical Heating and Cooling Rates of Pure water - **Nathan J. Mowry**
37	Seeking peak precision in atomic EELS mapping with counting mode direct electron detection - **Duncan T.L. Alexander**
38	Characterising point defects in exfoliated $PtSe_2$ - **Danielle Douglas-Henry**
39	STEM Characterization of TeO_2 Grown on CdTe Substrates: Understanding Possible Improvements to Back-Contact Passivation in CdTe Solar Cells - **Noah Kamm**
40	TEM analysis of Textured Hexagonal Silicon Crystals obtained via Nanoindentation - **Antonio M. Mio**
41	Atomic-scale chemical and spin-state analysis of one-dimensional magnetic nanochains inside carbon nanotubes - **Linxuan Li**

POSTER	TITLE
42	Machine Learning Based Characterization of Active Dopant Concentration in Si with EELS - **Jian-Min Zuo**
43	CEFID: A flexible platform for spectroscopic experiments - **Giulio Guzzinati**
44	A Machine Learning Approach for Automatic Detection and Classification of Defects in Atomic Resolution STEM Images - **Raja Abdul Wahab Ayyubi (Noah Kamm)**
45	Atomic-resolution mapping of phonon modes across crystallographic shear structures in thermoelectric (Al,Nb)-doped TiO_2 - **Shihao Wang**
46	Electron microscopy at CNR-IMM and it's efforts for experimental data trustworthy and FAIR trough NFFA-DI consortium - **Elisa Brugaletta**

1

Characterization of Aberration-Corrected Lorentz TEM Applying a Magnetic Field with Objective Lens

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Lorentz microscopy is one of the powerful observation methods without the influence of magnetic field from the objective lens (OL) of a transmission electron microscope (TEM). Normally, the magnetic field around the TEM specimen is about 2 T. This field could adversely affect the observation of magnetic material. To avoid the influence, OL needs to be turned off and the lens under the OL is used as a focus lens, where it is called an objective mini-lens (OM) in JEOL instrument. We can use the OL to apply some amount of magnetic field to samples in Lorentz microscopy [1]. Recently, the spherical aberration correction was applied to Lorentz microscopy, the resolution was improved [2]. They showed the applied magnetic field with OL is possible with spherical aberration correction. However, the optimum setting for aberration-corrected Lorentz TEM has not been understood yet. In this research, we characterized the detailed capabilities of spherical aberration-corrected Lorentz TEM while applying magnetic field by the OL.
In this time, JEM-ARM300F2 with TEM spherical aberration corrector was used. The pole-piece was wide gap pole-piece. The TEM resolutions with/without spherical aberration correction were investigated in 300 kV. The varied resolution with applying the magnetic field and the field of view were also measured at that time. The aberration measurement was done by diffractogram tableau method. The correction area was about 4 mrad and the third order spherical aberration coefficient of OM was under 10 mm.

The Young's fringe test was performed to check the TEM resolution. The resolution without spherical aberration correction was 1.83 nm without applying the magnetic field, and it was slightly improved by applying magnetic field to 1.75 nm with 0.5 T and 1.61 nm with 1T. On the other hand, they were improved with spherical aberration correction. Achieved resolution were 0.76 nm, 0.67 nm and 0.58 nm with 0 T, 0.5 T and 1 T, respectively. The effective field of view (FOV) was changed by applying magnetic field with objective lens. FOV was decreased as a function of the applied magnetic field. They were measured to be 200 µm with 0.3 T, and 20 µm with 1 T. This limitation was occurred by replicating of electron beam. The replicating was caused by the aberration of imaging system. On the other hand, the ratios of magnification were increased 6% with 0.5T and 45% with 1 T.

The principal plane of complex lens was getting closer to the sample plane as applying magnetic field. The complex lens was formed by OM and OL. This lens improved the spatial resolution, and this result showed the resolution was limited by the chromatic aberration.

We have addressed making a setting of aberration-corrected Lorentz TEM imaging mode. The TEM resolution was improved by spherical aberration correction of OM. Additionally, applying the magnetic field by the OL, the resolution was also slightly improved. The magnification and FOV were changed as a function of applied magnetic field. In the presentation, which condition could be the better for imaging a magnetic sample will be discussed. In addition, the spherical aberration correction could be applied to STEM Lorentz microscopy. These results will be introduced with the resolution and the optical constants, too.

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2

Enhanced Analysis of Hyperspectral Analytical Scanning Transmission Electron Microscopy Data through Blockwise Processing

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Processing the vast amounts of data generated by STEM spectroscopy, particularly hyperspectral EDXS data, poses significant computational challenges, including memory constraints and the risk of losing minority phases in monolithic analyses. To address these issues, here we extend our in-house, model-based non-negative matrix factorisation (NMF) method, called EspM-NMF[1] into a new tool that processes data in a blockwise manner before performing a global reconstruction.

Our method begins by splitting the hyperspectral data into spatial blocks based on the dataset's spatial structure. We then perform principal component analysis (PCA) using singular value decomposition (SVD) to identify the number of relevant components in each block, followed by an initial NMF decomposition of each block using our EspM-NMF algorithm. This step estimates the spectral components on a blockwise basis. During post-processing, we integrate a priori information about elemental composition, phase details, and chemical compounds. We then use a custom spectral clustering algorithm to identify shared features across the data blocks, reducing the number of spectral components to match the total expected for the entire dataset. Finally, we derive the corresponding spatial components by solving the least squares problem blockwise and concatenating the results.

In order to compare the results quantitatively with the regular, monolithic approach, we apply both approaches to a synthetic dataset generated using a previously-developed EDXS spectrum image simulation algorithm[2]. We find that our blockwise approach significantly improves the separation of spectral and spatial components, enhances noise resilience, and helps the identification of minority phases that monolithic processing often misses. Our method also enables efficient out-of-core processing, overcoming memory constraints and facilitating the analysis of large datasets without the need for high-performance computing clusters.

Through the integration of prior knowledge and the blockwise NMF-based decomposition, our tool not only improves the accuracy of separating spectral and spatial components, but also ensures the identification of minority phases. Spectral clustering helps in identifying shared spectral features across blocks, improving results' consistency, reliability, and interpretability. This new approach, which we plan to publish in open-source format, will enable more detailed and comprehensive analyses in STEM spectroscopy, paving the way for deeper insights into material structures and compositions.

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3

New Method for Calibrating Electron Spectrometers with μeV precision

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Recent developments of new electron sources and electron spectrometer hardware have pushed the spectroscopy resolution to the meV energy range and atomic level, orders of magnitude better than older generation equipment. However, the manufacturer hardware calibration approaches have not evolved sufficiently to be commensurate with these recent developments. Moreover, with the increased spectroscopic resolution, developing new, high-precision spectrometer calibration methods with sub-meV resolution becomes more imperative. Here, we discuss a new calibration method that uses strong scattering and coupling of the free electrons of the electron microscope with the optical field produced by photons circulating in the microring resonator chips.

The interaction of the TEM electrons with the microresonator chip photons causes the electrons to gain and lose energy in multiple orders of the resonator photon energy and produce EEL spectra exhibiting many precisely spaced sidebands spanning the pixel range of the detector. Such spectra provide a precise energy ruler for calibrating the spectrometer with a precision of few μeV- an accuracy of as much as three orders of magnitude over conventional calibration techniques. In addition, this technology can provide real-time measurements of spectrometer non-linearities and energy drift that give errors of 100s meV but can be corrected, providing unprecedented precision on μeV level. Combining these μeV precision measurements with machine learning and AI would be a paradigm shift in electron spectrometer technologies, enabling new automated spectrometer alignment and calibration methods.

4

Probing the Itinerant Magnetic Behavior of FeRh using In-situ Electron Magnetic Circular Dichroism (EMCD) and Lorentz TEM

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Research in spintronic, quantum, and other magnetic technologies aims to shrink devices down to a material's fundamental levels, i.e., to the scale of collective or individual spin states. For such developments to occur, material characterization methods must be amenable to probe magnetism on the revelant length scales spatially. In particular, a question arises in the validity of quantitative X-ray and Electron spectroscopy results when the probe size is smaller than the wavelength spin fluctuations and collective spin dynamics. For a better understanding of the influence of probe area on itinerant magnets such as FeRh, we use EMCD, an electron microscopy approach analogous to X-ray magnetic circular dichroism (XMCD) that provides nanometer spatially resolved element-specific information about the magnetic properties, e. g., spin and orbital magnetic moments.

Exploiting the first-order transition from antiferromagnetic (AFM) to the ferromagnetic (FM) phase in FeRh as a reference, we show that the ECMD measured spin-orbital magnetic moments differ by an order of magnetic between 1μm and 1nm probes. We explain these results in the context of the Self-Consistent Renormalization (SCR) theoretical framework and the validity of sum rules for itinerant systems. However, the results of this study shed light on fundamental aspects of how we interpret XMCD and EMCD spectroscopy data.

Enhanced time resolution with a room-temperature energy dispersive X-ray PIN photodiode detector

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Energy dispersive X-ray spectroscopy (EDX) has demonstrated its utility as a user-friendly analytical technique in electron microscopy for elemental analysis and mapping of specimens. Detector technology has evolved to configurations of multiple silicon drift detectors (SDDs) allowing for high count rates by having a large active area while keeping the anode capacitance low. Incident X-rays are absorbed in the bulk Si region and generate electron-hole pairs. The electrons then drift towards the anode under guidance of an electric field. The drift speed is relatively low and together with the large active area size limits the precision on the X-ray arrival time. X-rays absorbed near the anode are read out significantly faster than those absorbed farther away [1]. Currently this is the main bottleneck for EDX and energy electron loss (EELS) coincidence detection [2] where the aim is to correlate transmitted electrons and X-rays in time. This information can among others greatly enhance the sensitivity for detecting trace elements in a matrix while preserving both the EDX and EELS signal without compromise. To tackle this issue, we are working on a proof-of-concept detector consisting of a small reverse-biased Si PIN photodiode with a low-noise charge amplifier circuit. Contrary to the ongoing trend of ever-increasing size of active areas and number of detectors we ensure sufficient collection by bringing our sensitive area and specimen very close together. The sensitive area can thus remain small, keeping the photodiode's capacitance low enough to allow for high acquisition rates and low noise levels. Moreover, the small size limits the temporal broadening as initially aimed for. So far several prototypes have been made and tested in a scanning electron microscope (SEM). Over the different iterations the leakage current, signal-to-noise ratio (SNR) and form factor have been improved. With Cu X-rays the current SNR of the single X-ray pulse signals we are measuring is above 5, with pulses exhibiting sub-50ns rise times. The entire system consumes less then 750mW of power. Initial spectra show an energy resolution of around 1keV at 8,05keV (Cu K-). Currently different photodiodes are being tested and compared as well as ways of lowering the leakage current to further improve the energy resolution. In conclusion, we present the development of an in-house build room-temperature PIN photodiode X-ray detector to improve time resolution and allow for advancements in EDX and EELS coincidence experiments that so far have been hampered by the slow drift mechanism in SDD setups.

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6

Quantification and Denoising of Atomic-Resolution EDX Spectrum Images Through a Multiscale Bayesian Approach

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In this work, we propose the use of a multiscale Bayesian estimation strategy (BMS) to deal with the extreme noise and signal sparsity observed in atomic-resolution energy-dispersive X-ray (EDX) spectroscopy data. The method, originally developed for noisy LIDAR data1, relies on two factors. First, it assumes pre-known contributions to the EDX spectrum from each chemical element. In the present work, the elemental contributions to the spectra are obtained from the espm library.2 Secondly, it adopts a multiscale model that accounts for the Poisson statistics of the data and exploits spatial correlation between pixels to provide robust abundance estimates of each element in each pixel, even in the presence of high noise. The outputs of the algorithm are maps of the X-ray signal quantified in atomic percentage, which are considerably denoised, and corresponding Bayesian uncertainty maps. The method has first been tested using (non-atomic) simulated EDX datasets generated using espm, which allows comparison to a ground truth, where it demonstrates better accuracy than classical peak integration quantification, especially in low signal-to-noise ratio (SNR) situations. Next, the analysis method was also tested on experimental atomic-resolution EDX spectrum images (Figure 1) from a LaVO3 perovskite sample, as a function of integrated number of frames (or equivalently acquisition time). The Bayesian multiscale method shows better spatial resolution, SNR and quantification accuracy, especially for short acquisition times.

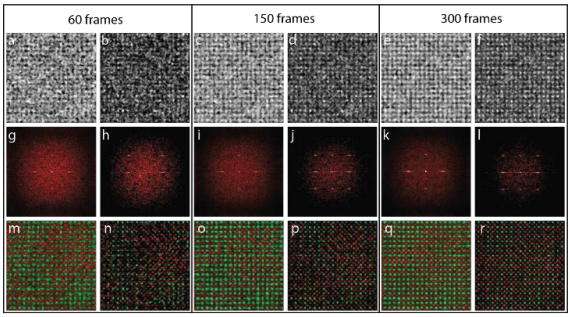

Figure 1.
BMS approach compared to processing using commercial Thermo Fisher Scientific Velox software, for an increasing number of frames from the EDX spectrum image.
Top row: quantified V maps. Middle row: Fourier transform of the map above it;
Bottom row: color mix map between the V maps (green) and La maps (red).
The columns a-g-m, c-i-o and e-k-q correspond to Velox analysis, the rest to BMS.

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Overcoming the aberration-limit of a non-corrected Transmission Electron Microscope with computational ghost imaging

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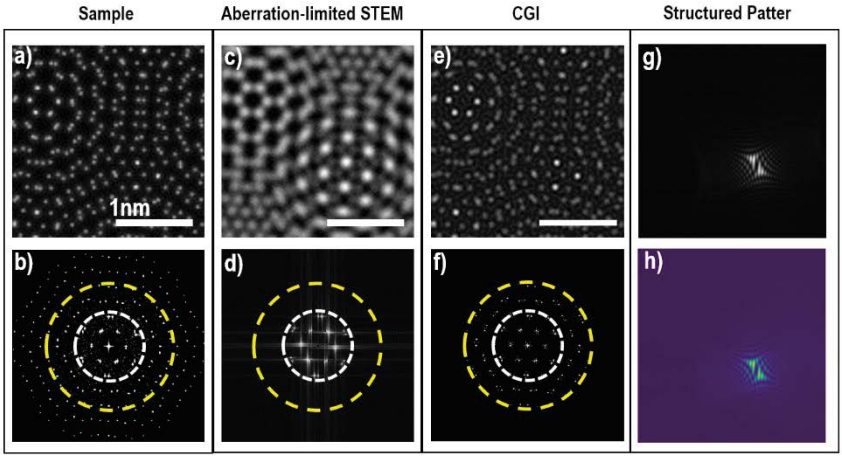
The invention of aberration correctors at the end of the 20th century has made modern Transmission Electron Microscopes (TEMs) able to image and characterize samples with the highest lateral (spatial) resolution. [1,2] In particular the ability to correct spherical aberration was the pivotal factor for high resolution imaging. [3,4] Unfortunately, aberration correctors are quite costly and are reaching performance limitations.
Here we numerically demonstrate that by using computational ghost imaging (CGI) [5] we can overcome the resolution limit imposed by aberrations in TEMs. In CGI the image of the sample (and its spatial information) is computationally recovered by illuminating the sample with a series of known structured beams and collecting the integrated transmitted intensity via a single-pixel bucket detector positioned after the sample. The electron modulator (beam shaping device) is assumed to be in the last condenser aperture, while the sample (a twisted bilayer of MoS2 – Figure1a) is conventionally positioned in the sample plane. As single pixel detector we used the annular dark field. Our CGI scheme can overcome the aberration limit imposed by the instrument (figure 1e) with a two-fold increase in spatial resolution compared to an aberration-limited STEM image (figure 1c) as confirmed by the analysis of their FTTs (figure 1f and d, respectively).
We are now working on an experimental implementation where the electron modulator has been realized using MEMS technology. Moreover, we are also working on employing machine learning to predict the structured patterns (ground truth figure 1g, predicted figure 1h) and further optimize the pattern generation. Lastly, we have started to adapt a mixed scheme where we also control the scan coil of the TEM to optimize the illumination, increase reliability and acquisition speed. This should also allow for the reconstruction of the probe.



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What is the width of a grain boundary, and how to determine it?

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Atomic-scale defect configurations determine the properties and functionalities of materials. The application of stresses such as elevated temperature, modified gas phases, or externally applied electric fields can alter interface structures and, therefore, modify microstructures and macroscopic materials properties.

Using bicrystal experiments we have previously demonstrated that electric fields directed across grain boundary planes can alter the atomic and electronic structures of (100) twist grain boundaries in SrTiO3 [1]. Electric fields directed along the interface plane alter the atomic and electronic grain boundary structures as a function of field strength and proximity to the positive and negative electrodes. EELS and XPS have revealed field-induced oxygen ion migration along the interface planes [2]. In this project electric fields were directed along a 24° tilt grain boundary in undoped SrTiO3. HAADF-STEM imaging revealed a change of the periodic structural units that constitute the grain boundary core structure as a function of location between the positive and negative electrodes during field annealing. Detailed analysis of image intensities reveals that structural units are similar at the two ends of the boundary but have different widths. ELNES and low-loss EELS experiments reveal changes of the electronic structures and local optical properties.

For the same locations HAADF-STEM micrographs, spatially resolved near-edge fine structures, and the local complex dielectric functions obtained from low-loss EELS experiments each demonstrate different interfacial widths. Preliminary 4D-STEM experiments further indicate gradual changes of charge across the grain boundary plane over yet another interval. This presentation will discuss concepts of interface expansion, i.e., width of grain boundaries determined with different STEM techniques. The relevance of this discussion is illustrated, for instance, by the classical equation for grain boundary mobility that depends on “interface width” [3]. A definition of this term remains unclear to date and hinders a quantitative and analytical description of grain growth.

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9

Physics-based scan distortion correction in hardware

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The application of scanning transmission electron microscopy (STEM) techniques to the study of dynamic specimens (in situ STEM), large specimen areas, and/or biological or other radiation-sensitive specimens, is hindered by limited acquisition speed and specimen radiation damage effects. Even with high-speed pixelated detectors for 4D STEM or the use of even faster analog detectors for 2D STEM, high sampling rates do not entirely alleviate these problems. Unfortunately, decreasing dwell time introduces significant scan distortions, requiring flyback delays that significantly reduce the time resolution of imaging and preventing the use of non-rastered scan patterns that may be beneficial for particular experiments.
The most common approach to addressing these distortions uses post-processing: analyzing acquired images, moving pixel values to their estimated correct location, and then interpolating a final distortion-corrected image. Unfortunately, image correction through post-processing may introduce artifacts that can significantly affect the interpretation of results, it does not address the potential radiation damage effects stemming from deviations in the position of the beam on the specimen, and delay introduced by post-processing hampers real-time feedback and automation. Additionally, while linear interpolation during post-processing may be straight-forward for 2D STEM, its application to more complex 4D STEM measurements can be ambiguous.

Direct Electron has developed a physics-based theoretical model of scan distortions and a patent-pending system for reducing these distortions during acquisition with their DE FreeScan scan generator by modulating the voltages delivered to the microscope's scan coils. After calibrating two or three parameters for the microscope, subsequent STEM acquisitions can be acquired with a wide variety of scan patterns, minimal delays, and minimal distortions, without applying any post-processing.

Results on waffle grids show a striking reduction in scan distortion for unconventional scan patterns, such as serpentine or rectangular spiral patterns. Notably, no flyback or other delays are required for these scan patterns and no post-processing is required. Experiments to show the impact of scan distortion correction and various scan patterns on more interesting materials (including radiation-sensitive materials) are in progress at the University of Wisconsin and will be presented at the FEMMS meeting.

We expect distortion-corrected STEM to not only increase the accuracy and speed of STEM experiments, but also enable a wide range of new scan patterns that were previously impractical due to scan distortions.

10

Atomic-Scale Characterization of Monolayer Amorphous Materials Using Advanced Low-Voltage Electron Microscopy

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Amorphous materials, which break the periodic symmetry of crystals, possess unique and variable properties that can be tailored to meet specific requirements in a wide range of applications. Due to their complex and random structures, studying the property-structure relationships in amorphous materials has been a longstanding challenge. However, recent advances in the controlled synthesis of two-dimensional (2D) amorphous materials have renewed interest in this field by providing simplified amorphous structures. [1-2]

In this study, we developed a new method for the synthesis of 2D monolayer amorphous carbon and utilized an aberration-corrected Nion HERMES-100 microscope operating at an acceleration voltage of 60 kV to investigate the structural characteristics of the synthesized samples. The relatively low acceleration voltage was chosen to minimize structural changes induced by the electron beam. In addition, for such highly disorder 2D carbon structure, low-dose conditions are proven necessary, even at 60 kV. Key structural features, such as bond lengths, bond angles, ring percentages, and pair distribution functions (PDF), were characterized from atomically resolved STEM-ADF images. Nanoscale features of structural ordering were further examined using 4D-STEM NBED technique to provide comprehensive structural insights. These structural details were then correlated with performance tests to explore the structure-property relationships at the atomic scale. Understanding the origin of the unique properties of amorphous materials and the mechanisms behind their structural formation remains a significant area of study. Atomic-scale structural characterization by low-voltage low-dose STEM is a crucial step towards achieving this understanding, potentially leading to the development of novel materials with tailored properties for advanced technological applications.

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Emission characteristics and stability of the LaB_6 nanoneedle emitter

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From materials science to biology, TEM is widely used for research. To further improve spatial and energy resolution, the development of electron sources with higher brightness and better coherence is of great importance. W(310) is the most commonly used cold cathode field emission (CFE) emitter material that can be practically applied.
In CFE emitters, the use of a material with a low work function is said to result in high brightness and narrow energy spread. The work function of W(110) is about 4.5 eV, whereas that of LaB6(100) is as low as about 2.5 eV. Therefore, LaB6(100) has been expected as a CFE material, but it has not been put to practical use because stable field emission could not be obtained due to the inability to maintain clean surface.
Recently, Tang et al. of NIMS have succeeded in stabilizing the LaB6(100) surface and fabricating a needle emitter by FIB processing [1-2]. The energy spread of this emitter ranges from 0.27 eV to 0.38 eV, which is lower than that of conventional W emitters (0.45 eV), due to its lower work function [1]. In this study, we installed the LaB6 nanoneedle emitter on the JEM-2100F and characterized its field emission characteristics at an accelerating voltage of 200 kV (Fig. 1). The stability of the probe current was also measured for 24 hours (Fig. 2). The amount of current fluctuation was approximately 3%, indicating that stable probe current could be maintained over a long time without flashing.

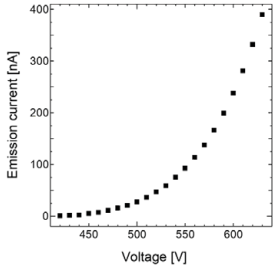

Fig 1. Filed emission I-V curve

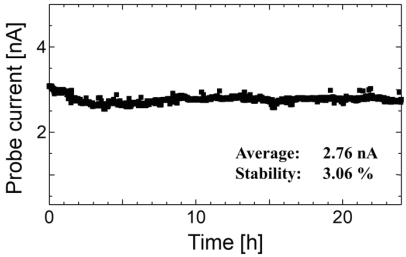

Fig 2. Stability of the probe current for 24 hours

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12

Edge State Enigma: An EELS Detective Story

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2D materials are often touted as wonder materials, with unique properties such as the exceptional electrical conductivity of monolayer graphene and the intralayer exciton landscapes of moiré superlattices [1]. Another often stated characteristic of 2D materials is their chemical stability. However, as soon as atoms are removed from the lattice to form open edges, the resulting dangling bonds render the material susceptible to chemical attack. This instability is acutely relevant for applications that specifically seek to tailor stable vacancy defect clusters, e.g. 2D nanopores for the sensing and separation of ions and small molecules. Despite substantial efforts to fabricate and characterize these structures, atomic-level understanding of the mechanisms of edge state stability and control are still lacking.

In the present study, we use electron energy loss spectroscopy (EELS) to investigate the edge states of vacancy defect clusters in monolayer hexagonal boron nitride (hBN). We target sub-nm triangular vacancy defects to test predictions of mechanosensitive ion transport [2]. Ensembles of these defects are fabricated with high precision using our recently developed de-coupled ion and electron irradiation approach. In broad-beam EELS experiments (operating in transmission electron microscopy (TEM) mode) we delve into boron K-edge fine structure to provide hints as to the edge chemistry of the defects. This dose-efficient approach leverages signal averaging over thousands of defect structures and allows one to probe the defects collectively even under hydrocarbon contamination [3]. Then we use scanning TEM (STEM)-EELS with a sub-Ångström probe to directly correlate boron K-edge chemical shifts with the atomic structure and elemental composition of individual vacancy defect edge states.

The EELS evidence collected thus far reveals tantalizing clues about pore edge stabilization mechanisms involving substitutional dopant species. We seek to harness these effects to functionalize vacancy defect structures with known species. This has implications ranging from passivated nanopores for practical applications outside of vacuum to the deterministic fabrication of dopant-vacancy color center complexes in 2D hBN for future quantum technology platforms.

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13

Development and Applications of a High-Energy Electron Energy-loss Spectrometry System in an Aberration-Corrected Scanning Transmission Electron Microscope

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Electron energy loss spectrometry (EELS) analysis can offer elemental identification, compositions, and insight on chemical bonding status. An EELS system mounted on the latest aberration-corrected scanning transmission electron microscope (STEM) has benefits over the synchrotron-based X-ray Absorption Spectrometry (XAS) approaches especially in terms of analytical spatial resolution. Due to the limitation in energy-loss ranges (~3,000 eV) in a typical EELS system, however, EELS information recorded from lower energy-loss edges is hardly compared with XAS data obtained through relatively higher energy edges. Therefore, we have developed a new EELS system acquirable for higher electron energy-loss signals (aiming to expand the acquirable energy-loss range from conventional ~3,000 eV to 13,000 eV) and installed to an existing aberration-corrected S/TEM JEOL JEM-ARM200CF together with a new scanning generator called MDP at Lehigh. The system development and installation were performed in collaboration with JEOL (USA, Germany and Japan) and CEOS (Germany).

The new EELS system is based on a CEOS Energy Filtering and Imaging Device (CEFID) [1] with two electron detectors: a CMOS-based CCD camera TVIPS TemCam-XF416 and a highly sensitive hybrid-pixel electron detector Dectris ELA. EELS signals can be acquired by either CEOS Panta Rhei software or newly developed JEOL FEMTUS software. To achieve the high energy-loss signals properly, several hardware settings were optimized, not only in the spectrometer but also in the post-specimen lenses of the microscope. After applying all the tuning, it was possible to record high energy-loss signals beyond 10 keV including the Ge K edge over 11 keV without changing the accelerating voltage.

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Streamlining Graphene Liquid Cell Preparation: VitroTEM's Naiad System

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Graphene Liquid Cells (GLCs) offer unprecedented capabilities for imaging samples in their native hydrated state, providing insights at atomic resolution [1]. However, manual assembly of GLCs presents significant challenges, limiting their widespread adoption. This poster presents VitroTEM's Naiad system, a revolutionary approach to GLC preparation. By employing a layered structure consisting of monolayer graphene sheets on standard TEM grids, the Naiad system rapidly constructs GLCs, encapsulating liquid samples for imaging [2]. Our poster showcases images of ferritin particles in their native environment, demonstrating the system's efficacy in biological materials research. Additionally, we present atomic resolution images of Au nanoparticles, highlighting its utility in nanomaterial science. The Naiad system simplifies GLC assembly, enabling researchers to focus on sample imaging rather than grappling with graphene preparation. This poster emphasizes the Naiad system's potential to accelerate discoveries in diverse fields reliant on high-resolution imaging of liquid-phase samples.

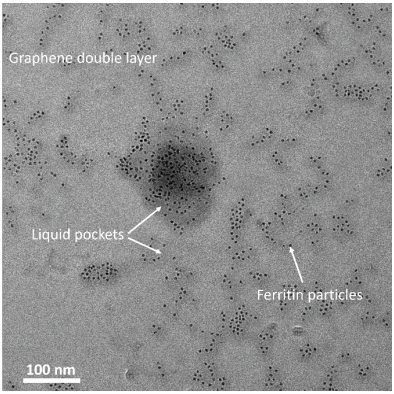

Fig 1. Ferritin particles encapsulated in GLC pockets.

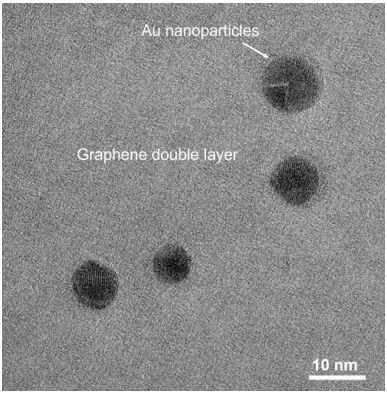

Fig 2. High resolution TEM image of Au nanoparticles encapsulated in a very thin GLC pocket.

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15

Event-responsive Beam-modulated STEM with Multi-frame and Sparse Scanning

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Recent advances allow any analog STEM detector to operate in an event-counting mode, measuring individual electron detection events in real time. This technology delivers images with SNR limited only by ideal Poisson counting statistics [1]. By combining such event-counting hardware with fast electrostatic dose-modulators (EDM) [2], a real-time 'event responsive' imaging mode can be realised [3]. We call this Trigger Event Modulated Probability Observation STEM, or TempoSTEM for short. In classical STEM, we observe fixed periods (dwell times) and record the varying numbers of transmitted/scattered events that arrive at a detector. TempoSTEM operates in a fundamentally different way, where we specify a fixed number of events and measure the varying time needed for the image signal to reach that number of electron events and the beam is blanked for the remainder of the pixel once the threshold is met, significantly reducing dose and sample damage.Information theory predicts that there is a reduced information content for every successive electron recorded per pixel, which has been confirmed experimentally [3]. This creates a balance where a lower Tempo trigger exit condition ensures the most information efficient detection per-electron, but produces low SNR images. Here we present a mode to iteratively update noisy high-variance pixels to achieve improved resolution and SNR. Multi-frame imaging is another common approach to increase the SNR of an image. Where a hardware EDM is present [2], a further option is to only revisit a subset of pixels in subsequent frames in the series. After the first noisy scan-frame of a TempoSTEM acquisition, we can find pixels that are outliers by, say, more than one standard deviation of their neighbours. On the second scan frame, we can revisit only these pixels and an additional TempoSTEM observation can be made, with the beam blanked elsewhere to minimise dose. The rescanned pixels have now received the dose equivalent of two frames, and the scattering rate estimate is updated. On a third scan for example, even fewer are rescanned. After some number of rescans the variance is converged, and the image acquisition can be considered complete. This produces a significant dose saving for equivalent SNR, up to 2x, even relative to the already low-dose TempoSTEM approach. This method pushes below one event per-pixel-per-frame, maximising the information from each electron, increasing the best achievable combination of resolution and beam damage.

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16

Technological Advances for Temperature-Dependent operando Electrochemical Studies within the TransmissionElectron Microscope

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The study of in situ or operando electrochemical systems entails observing electrochemical behavior of samples in environments as close to “real working conditions” as possible.
Scanning/transmission electron microscopy (S/TEM) enables researchers to characterize physical behavior on the sub-nanometer scale, but conventional S/TEM techniques require sample imaging and analysis in a high-vacuum environment. Closed-cell S/TEM holders allow researchers to study their electrochemical systems in liquid, including the option to flow or exchange electrolyte across the sample, by hermetically sealing the experiment from the high-vacuum environment of the TEM. This technology allows direct observation of processes at the nanoscale that influence material performance, such as the formation of the solid electrolyte interphase (SEI) layer during lithium-ion battery cycling[1], the preferential corrosion of stainless steel at sulfite[2] and silicate[3] inclusions, and the degradation of shape-controlled nanocrystals during electrocatalytic cycling[4-6].

The next generation of in situ electrochemistry systems will push closer to “real working conditions” through advanced temperature control. Many applications require temperatures beyond room temperature, such as fuels cell catalysts, batteries, and corrosion. In this poster, we will share initial results from a new in situ electrochemistry system featuring temperature contro looking at well-known redox couples (Figure 1a) and of dendrite formation (Figure 1b) as a function of the temperature the implications of this new technology to research across numerous application fields including fuel cells, ionic liquids, batteries, corrosion, and electrocatalysis.

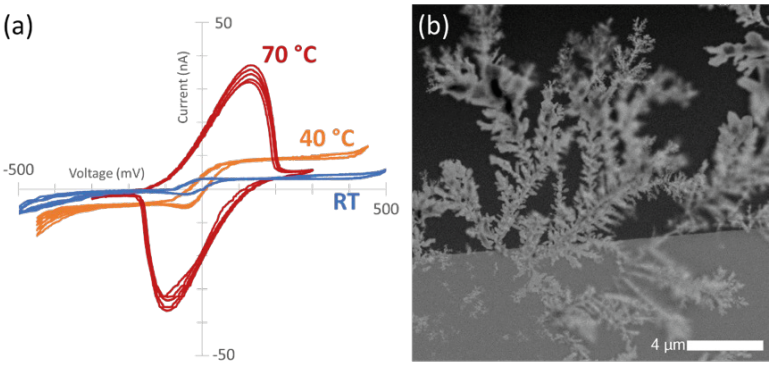

Fig 1. (a) Chemical redox cycling of 50 mM ferric and 50 mM ferrous chloride at room temperature (blue), 40 °C (orange) and 70 °C (red) within a closed-cell TEM system. (b) TEM micrograph of silver dendrites formed in real time on the platinum working electrode by chronoamperometry.

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How to tackle the most sophisticated materials science questions with advanced TEM: towards reliable investigations of materials in their native state

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The path towards a circular economy requires developing and optimizing technologies that will enable the green transition of society. Energy materials, catalysts, materials for pollutants removal and quantum materials are at the heart of these crucial sectors. Advanced microscopy is of huge importance when developing new materials. However, the afore mentioned materials are often sensitive to the electron beam or influenced by the magnetic field of the TEM, which makes them obstinate objects for the advanced TEM investigations.

The mission of Thermo Fisher Scientific is to make the world healthier, cleaner, and safer. One important way to achieve this mission is to support the rapidly developing need for atomistic investigations of electron beam and magnetic field sensitive materials. Thermo Fisher Scientific is developing dedicated workflows interlinking both hardware and software innovations.
In this contribution we will highlight and discuss the recent software and hardware innovations as well as workflows, illustrating the power of our solutions with several practical examples.

18

In situ biasing DPC STEM observation of p-n junction depletion layers

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Differential phase contrast scanning transmission electron microscopy (DPC STEM) can visualize and quantify local electric fields and charges within a specimen at high spatial resolution [1,2]. Since directly characterizing local electric fields and carriers is useful for developing better property materials and devices, DPC STEM has attracted much attention in recent years. On the other hand, in situ biasing S/TEM observation with the micro-electromechanical systems (MEMS) has been intensively developed [3]. Combining DPC STEM with the MEMS biasing techniques will be a powerful tool for directly observing local electric field dynamics at high spatial resolution. However, the optimal experimental procedures for in situ biasing DPC STEM experiments, such as sample preparation procedures for ensuring good electrical contacts, has not yet been well established. In this study, we developed a technique for in situ biasing DPC STEM observation using a MEMS-based system. We selected a p-n junction in GaAs semiconductor as a model sample, doped with 1018 and 1019 atoms/cm3 for the p-type and n-type regions, respectively. A sample was prepared with a focused ion beam (FIB) system (Helios 5 UX, Thermo Fisher Scientific, Inc.). The sample lamella containing the p-n junction was fixed to the voltage-applying circuit on the MEMS chip and was thinned to 400 nm. The in situ observation was performed using STEM with 16-segment SAAF detector (JEM-2100F, JEOL Ltd.) and an in situ MEMS biasing holder (Lightning Sample Holder DHB30, DENSsolutions). The electric field distribution across the p-n junction was observed under various external voltages. We successfully visualized the electric field changes across the p-n junction when we changed the applied voltages. We performed Poisson-Schrödinger simulations with NEXT NANO++ software [4] and compared the calculated electric field distribution with the experimental results. We evaluated several possible effects on the observed electric fields, such as FIB sample fabrication process (the effect of FIB deposited electrode, FIB damage on sample surface and so on), based on the results of both experiment and simulation. Details of this study will be discussed in the presentation.

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19

Towards Atomic-Resolution Electron Energy Loss Spectroscopy (EELS) in an Uncorrected 30kVScanning Electron Microscope

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As an era-defining technological advancement in the field of nanoscience and beyond, theeffective implementation of aberration correction has allowed electron microscopy to routinely reach deep sub-angstrom-level spatial resolution. Among many impactful consequences, these developments have seen thewidespread adoption of low-voltage instruments, which can maintain very high spatial resolutions thanks totheir aberration correctors, even down to 20kV, especially for applications in 2-dimensional materials at thesingle atom level [1]. Beyond single-atom sensitivity, low-voltage operation is highly sought-after for reasonssuch as reduced knock-on damage to samples or increased inelastic cross-sections resulting in a high signalfor spectroscopy. However, for a large number of practical materials science applications, the complexity and price of such instrumentation, especially when analytical capabilities are added, can be a drawback. Incontrast, high-throughput capabilities with lower entry barriers in terms of cost and complexity, but which maintain a relatively high-resolution, can often be preferable in order to address numerous scientific questions. One possible solution is the use of (low-voltage) scanning electron microscopes (SEMs) operated in transmission geometry – or (T)SEMs [2]. When equipped with cold field emission sources, these instruments have been shown to reach 0.2nm information transfer in bright-field STEM imaging [3], and to provide remarkable flexibility for surface and spectroscopic investigations of functional materials [4]. Here, we show how the capabilities of such a high-resolution (T)SEM can be pushed even further towards near-atomic resolution for EELS mapping. We use a Hitachi SU9000EA microscope, a low-kV (≤30kV) uncorrected (T)SEM equipped with a Hitachi electron energy-loss spectrometer developed for this instrument, which, thanks to its cold-field emitter, has a native energy resolution of ~0.3eV. In the optical configuration chosen, and at 30kV acceleration voltage, the estimated probe size was sufficient to observe 0.26nm spots in the Fourier transform of high-angle annular-dark-field STEM images of a $LaNbO_4$ A-site deficient perovskite [5] and to generate La M4,5 edge EELS map with atomic-plane resolution. The use of an edge with a high 832eV onset also highlights the applicability of EELS in this uncorrected 30kV system, even at relatively high energy losses. Other EELS applications, such as plasmonics and low primary energy core-loss (down to 3kV) will also be highlighted to further illustrate the versatility of these instruments, whose advanced capabilities as (T)SEM-EELS instruments belie their relative operational simplicity and low cost.

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20

Electron beam effects in monolayer 2D materials from knock on damage and inelastic scattering effects to chemical etching

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It is well established that electron irradiation during (scanning) transmission electron microscopy imaging can influence the structure of the studied sample due to elastic and inelastic scattering [1]. While the first of these, which dominates for conductive materials, is fairly well understood [2,3] with the help of quantitative measurements enabled by 2D materials and aberration-corrected instruments, the role of excitations arising from inelastic scattering remains under active research [4–7]. At the same time, the equally important role of the residual vacuum in the microscope is often completely neglected. However, as we have shown recently [8,9], it has an influence already at pressures typical for instruments with side-entry holders (10-7 mbar) on carbon structures without ideal sp2 bonding (hydrocarbon contamination, defects and pores in graphene) where the partial pressure of oxygen can even determine the observed termination of a graphene edge. In the case of more oxygen-sensitive materials such as MoTe2, the presence of oxygen in the column significantly accelerates the beam-induced damage even for the pristine material [10], whereas structurally similar but chemically more inert MoS2 remains unaffected. As will be shown here, controlling residual oxygen partial pressure in the microscope allows also determining the shape of pores created under electron illumination into hBN during imaging, where pores created under ultra-high vacuum show no clear edge-termination preference, whereas under a low-pressure oxygen atmosphere nitrogen-terminated triangle-shaped pores are dominant [11].

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21

Novel scan coil design for high spatiotemporal-resolution imaging in the scanning transmission electron microscope

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The Scanning Transmission Electron Microscope (STEM) is a highly versatile tool that is used to study a wide range of materials, from semiconductors to biological cells. It operates by scanning a beam across a specimen and then recording the beam that has been transmitted through the specimen at these points. STEM is a powerful technique for probing specimens down to the atomic scale, but the slow imaging speeds can result in excessive beam exposure, and therefore damage, and the inability to capture dynamic in-situ events. Increasing the scanning speed is therefore an attractive proposition for better control of the dose-rate on the sample as well as improved temporal resolution.

Currently, the main limitations to the scanning time in a STEM are imposed by the response time of the scanning coils, determined largely by their inductance. For this reason conventional imaging requires a flyback wait time between scan lines to reduce inductive hysteresis. Flyback time may be eliminated using novel scan paths, though at fast scan speeds the inductive effects are still problematic. Similarly, techniques such as compressed sensing can increase frame-rates but retains issues regarding the hysteresis of the scanning coils. Previous work by Ishikawa et al. shows scan coils with an inductance ~240 times less than the conventional scan coils, though with a limited scan area.

In this work we present a scan coil system to greatly improve the response time of the scanning system. Alongside the existing, main scan coils, we add additional deflectors with lower inductance to work simultaneously with the main scan coils. By using the second coils to counteract the effects of hysteresis in the main scanning coils, imaging at lower dwell and flyback times without image distortion or compromising scan area. The additional coils themselves are not perfect and further sets of coils could be used to compensate remaining hysteresis. Equally, the use of electrostatics due to the rapid response time and lack of hysteresis could be deployed in tandem. With our design, we hope to make fast scanning in STEM practicable and enable high-resolution imaging to improve dose-control and in-situ measurements.

Bridging the gap: beam-induced structuration between crystalline interfaces

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It is widely known that, as well as being a powerful analytical tool, the energetic electrons of a scanning transmission electron microscopy (STEM) probe can provide an interesting means for modifying the sample being studied, in-situ. Here, we investigate such beam effects within the gap between two crystalline, perovskite oxide lattices that are brought together by transfer and stamping [1, 2]. We study the samples using a 300 keV aberration-corrected probe, under a range of beam current, pixel dwell times, and total scan durations. In order to monitor the structural evolution, we apply a combination of Z-contrast high-angle annular dark-field (HAADF) imaging, energy dispersive X-ray spectroscopy (EDXS) and electron energy-loss spectroscopy (EELS) using a Gatan Continuum spectrometer equipped with a high sensitivity K3 direct electron camera.

The lattices are chosen to share the same basic compound, stamped manually to give a mis-tilt between them of maximum 4°. In the as-stamped condition, the gap between the lattices measures about 2 nm. Within it, an amorphous mixture of the perovskite elements and carbon is observed, which remains unaffected by the electron beam. However, when the stamped samples are subject to heat treatments, we find distinct changes in their behavior. First, an appropriate treatment reduces the gap size to ~0.9 nm. Second, while initially the gap is filled with amorphous compound, under the electron beam the atoms rearrange into a crystalline structure that progressively forms a crystalline bridge across the gap. From HAADF imaging, we observe that it adopts the same atomic structure as the perovskite compound of the upper and lower lattices, similar to what has previously been observed in structuration of ion beam amophorized perovskite oxides [3]. We monitor this structural evolution using frame-by-frame analysis of rapidly acquired scans. The nature of the crystalline bridge is further confirmed by studying near-edge structures in EELS, and by EDXS. By careful, systematic measurements we identify that this electron beam writing of the structure depends on surpassing a critical electron flux. Remarkably, at either side of a crystalline bridge, the “parent” lattices may distort locally to bring them into better structural alignment. We are studying this phenomenon with the goal of understanding better the bonding physics and chemistry behind it.

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Microstructural characterization of aging-treated aluminum alloy AA2024 with SEM, STEM, EDS, and EBSD

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The rapid evolution of the aerospace industry has driven the demand for materials that offer enhanced strength, toughness, and corrosion resistance. Among these materials, aluminum alloy AA2024 stands out due to its lightweight properties and superior mechanical performance, making it a popular choice for structural components in aircraft. The alloy's primary alloying elements, copper and magnesium, significantly contribute to its desirable properties by forming various precipitates during different heat treatment processes. This study aims to provide a comprehensive microstructural characterization of three AA2024 aluminum alloy specimens subjected to different aging treatments: T3, T6, and T81.
To achieve this, an integrated approach was employed, combining scanning electron microscopy (SEM), scanning transmission electron microscopy (STEM), energy dispersive spectroscopy (EDS), and electron backscatter diffraction (EBSD). These techniques were utilized to identify and analyze various precipitates, intermetallic particles, and dispersoids within the alloy samples. The Thermo Scientific™ Apreo™ ChemiSEM played a crucial role in this study by facilitating efficient data acquisition and processing through its integrated ChemiSEM™ Technology and streamlined EBSD workflow.

Figure 1 shows a variety of microscale intermetallic particles in the AA2024-T3 sample. Material contrast in the backscattered electron (BSE) images was used to distinguish particles with different physical characteristics. Conventional EDS analysis confirmed the presence of varying ratios of Cu, Mn, Mg, and Fe in all these particles. Automated ChemiPhase analysis was able to identify all the materials present in the analyzed area. ChemiPhase assigns each material to a specific phase and automatically calculates the associated spectrum, quantification, and area fraction.

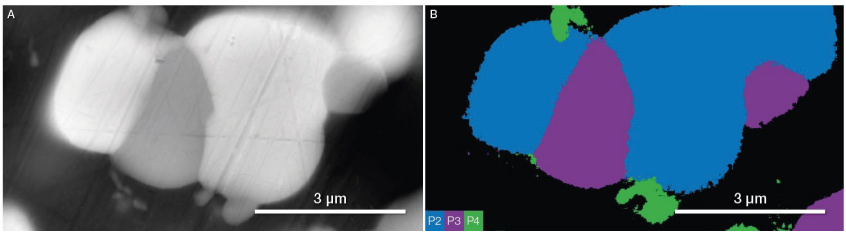

Figure 1. AA2024 sample subjected to the T3 aging treatment. Various intermetallic particles are differentiated through a combination of backscattered electron contrast imaging (A) and ChemiPhase analysis (B).
Different colors in the ChemiPhase map represent distinct phases.

24

Chemical and structural characterization of chalcogenide nanostructures for molecular recognition through controlled electron dose transmission electron microscopy

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Sensing is crucial to understand the overall condition of a given system and provide detailed and accurate information about its current state. Whether in industrial processes, environmental monitoring, healthcare or other applications, it is fundamental to detect changes and identify potential risks. Particularly, environmental molecular recognition has gained great attention for predicting and mitigating natural disasters, protecting biodiversity and safeguarding public health.1

In recent years, scientific research has focused on the development of remote sensing systems thanks to their cost and time effectiveness.2 In such a context, electrochemical sensors are excellent candidates due to their high sensitivity and selectivity, rapid response time, compact size and ease of integration.3

Transition metal dichalcogenides are ideal active materials for electrochemical sensors due to their excellent electronic properties, availability and robustness. Specifically, MoS2-based composite materials own unique properties making them even more effective for various sensing applications.4 In such a context, we synthesised MoS2 composite nanostructures by a facile one-step solvothermal method. In-depth studies of Raman spectroscopy and X-ray photoelectron spectroscopy analyses confirmed the formation of a composite material containing both metallic (1T) and semiconducting (2H) phases of MoS2. Transmission Electron Microscopy showed that our composite is composed by nanoribbon structures. This system was thus used to develop a new electrochemical sensor to monitor nitrite anions in aqueous solution as well as in real commercial water samples. Such results pave the way to the development of devices suited for real water monitoring.

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Camera Systems and Software for Fast 4DSTEM Data Acquisition and Live Virtual Detector Calculation

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The high frame rate of new detectors based on CMOS technology allows fast acquisition of large 4DSTEM datasets. We have developed a software called EMplified Scan for 4DSTEM acquisition with the TVIPS TemCam-XF416ES, the DECTRIS hybrid cameras QUADRO and the ultra-fast ARINA detector. Synchronization between image acquisition and scanning is controlled by the TVIPS Universal Scan Generator. The datasets are stored for postprocessing in HDF5 format with metadata for camera and microscope parameters.

EMplified Scan is able to calculate virtual detectors in real-time during the acquisition of 4DSTEM datasets. Therefore, several custom virtual detectors can be shown together with standard STEM detectors in a continuous mode. It supports microscope control like automatic beam blanking and detector insertion and retraction.

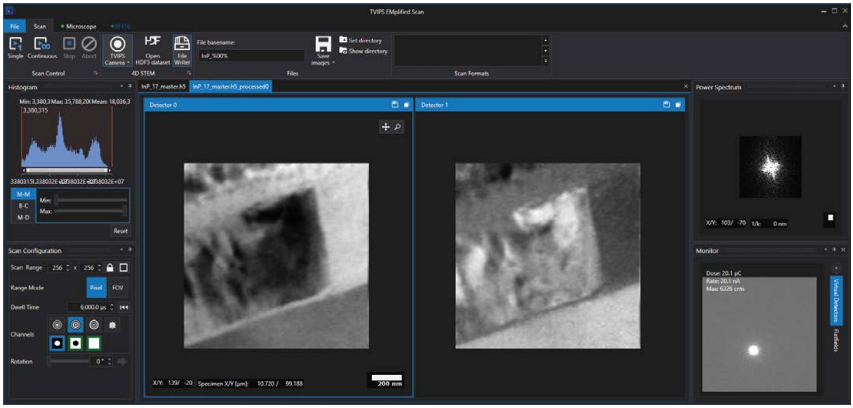

Fig 1. Screenshot of EMplified Scan showing virtual dark and bright field detector images of a cross section of sputtered InP. The monitor image (right lower corner) shows the live diffraction pattern on a TemCam-XF416.

A hybrid detector like the DECTRIS ARINA allows large scans in very short time. For example a 1024 x 1024 scan with the 2x binned pixel (96 x 96 pixel, pixel size 200x200µm) takes less than 10 sec. With 4k x 4k pixel, the TemCam-XF415ES offers a larger resolution and a higher field of view. The framerate is depending on the readout area of the sensor. A 512 x 512 pixel scan at a camera size of 256 x 256 pixel takes less than 6 min. For diffraction mapping application it is possible to scan, e.g. 100 x 100 scan points with an image field of 1024 x 1024 pixel in less than a minute.

In-situ characterization of the high-temperature spin-state transition n $LaCoO_3$ via STEM imaging and EELS

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In-situ scanning transmission electron microscopy (STEM) is a powerful platform for investigating phase transitions in materials, as it can provide time-dependent structural and spectroscopic information with unparalleled spatial resolution. This is particularly useful for the development of materials and devices for neuromorphic computing, where electronic and structural transitions are core to the device functionality. Neuromorphic, or brain-inspired, computing is based on the emulation of neurons and synapses in the brain by controllably and reversibly tuning the electrical conductivity. It has the potential to provide crucial improvements to computing speed and energy efficiency over CMOS technology at a time when computing needs are rapidly increasing. $LaCoO_3$ holds promise for neuromorphic switching devices due to its interrelated magnetic and electronic transitions [1]. Here we combine structural and spectroscopic measurements in the STEM with in-situ heating to characterize the high temperature spin transition of $LaCoO_3$.

Lanthanum cobaltite has two broad spin-state transitions between low, intermediate, and high spin states. Previous work has successfully demonstrated the measurement of the low-temperature low-to-intermediate spin-state transition through electron energy loss spectroscopy (EELS) near-edge structure analysis and cryogenic cooling [2]. The present work extends this analysis to the intermediate-to-high spin-state transition, which occurs over a temperature range (300K – 600K) relevant to microelectronics applications. We combine EELS analysis with atomic-resolution imaging to connect the spin, electronic, and structural components of the transition. We, further, discuss the challenges associated with these in-situ high temperature measurements, including oxygen loss in the high vacuum environment of the STEM, leading to formation of the brownmillerite phase ($LaCoO_{2.5}$). Challenges related to beam- and other instrument-induced effects are intrinsic to in-situ measurements, and the proper assessment and mitigation of them is crucial to achieving meaningful and repeatable results. Nonetheless, careful experimental design and analysis can produce important insights into the nanoscale behavior of phase transformation relevant to next-generation microelectronic devices.

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3. This work was supported as part of the Reconfigurable Materials Inspired by Nonlinear Neuron Dynamics (REMIND) Energy Frontier Research Center, funded by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES). Growth of the cobalt oxide film was performed at Sandia National Laboratories under contract #DE-NA-0003525. Microscopy characterization of the films was performed at the National Renewable Energy Laboratory under contract #DE-AC36-08GO28308 and at facilities of the Platform for the Accelerated Realization, Analysis, and Discovery of Interface Materials (PARADIM), which are supported by the National Science Foundation under Cooperative Agreement No. DMR-2039380.

Revolutionizing Nanoscale Characterization with TESCAN TENSOR's Analytical 4D-STEM

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Nanoscale characterization is instrumental in materials science research for enhancing the mechanical and physical properties of metals, alloys, composites, and other materials. Using the analytical 4D-STEM techniques, scientists and engineers gain detailed insights into crystallinity and structural composition at the nanoscale through diffraction patterns. This technique allows precise correlation of the structural information at specific sample locations with nanometer resolution, which is crucial for understanding and optimizing material properties such as strength, ductility, and conductivity. Through these insights, the scientists can develop advanced materials with tailored properties for various applications, from aerospace engineering to semiconductor manufacturing. The ability to obtain comprehensive images and detailed crystallographic and compositional data at nanometer resolution makes this approach indispensable for driving innovations in material science.

Although the fundamentals of 4D-STEM are well-recognized, its capabilities, precision and accuracy are substantially improved by using beam precession during data acquisition. This enhancement significantly improves the quality of diffraction patterns by minimizing (averaging out) the effects of dynamical scattering in the collected datasets and consequently increases the robustness of performed data analysis. TESCAN TENSOR leverages this technology by full integration of beam precession into the microscope hardware and software, making precession-assisted 4D-STEM measurements routine, fast and intuitive for scientists at all levels of TEM/STEM expertise. By automating nearly every function of the microscope, TESCAN TENSOR simplifies the complexity traditionally associated with TEM/STEM alignments and settings, thus broadening its accessibility. This automation, combined with real-time data analysis and processing, enables high-quality, robust 4D-STEM acquisition and analysis in contrast to the traditional approach of blind collection of diffraction data and later postprocessing after the microscope session. For experienced electron microscopists and method developers, TESCAN TENSOR additionally supports advanced methods and development through the Expert PI interface, which is flexible software environment offering full access to all microscope functions at the Python level and allowing the scientists to push the boundaries of their investigations.

The technology behind TESCAN TENSOR's advanced analytical 4D-STEM capabilities will be demonstrated using an example of Ti-Cu-Fe alloy. Rapid phase analysis enabled precise differentiation of small grains of the intermetallic Ti2Cu phase from the surrounding Ti-alpha and Ti-beta phases. Additionally, the orientation analysis revealed mutual orientations and spatial relationships of the individual grains and phases within the alloy. By leveraging these capabilities, researchers can gain a deeper understanding of the microstructural characteristics and properties of Ti-Cu-Fe alloy, enhancing their ability to tailor and optimize its performance for various applications.

28

Unveiling rapid nanoscale dynamics in copper catalyst during HOR and CO_2 hydrogenation reactions

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During their operations, catalysts undergo fast and localized structural and chemical transformations that drive them from thermodynamic stable states to non-equilibrium states.

Studying these highly dynamical transformations induced by energy absorption and/or environmental conditions, will allow a more precise structure-environment-property correlation. However, as the catalyst-environment interactions happen away from the thermodynamic equilibrium within timescales inferior to 10-3 s and involve structural changes that nucleate and propagate at the nanoscale, only a limited understanding of them exist. Aberration-corrected TEM coupled with in-situ thermal gas-phase setups have an important contribution to the field of heterogenous catalysis owing to its ability to describe morphological and structural changes, phase transitions, and to probe the reversibility of these changes. Of special interest, is its ability to reveal aspects of the non-equilibrium dynamics, as changes in the environmental conditions (e.g., temperature, gas components partial pressure) induce continuous changes in the catalyst surface shape, composition, or structure [1].

Using the hydrogen oxidation reaction (HOR) and CO_2 hydrogenation on copper nanoparticles as model systems, this study focuses on revealing in real-time the dynamic phase-transitions of copper and their nanoscale propagation. Reversible transitions during the catalytic activation, from the inactive Cu^0 to active Cu_2O, the phase coexistence and fast transformations at optimal performance, or the surface faceting evolution with the changes in the local chemical potential, will be investigated using a comprehensive in-situ gas phase TEM methodology. Using slow stimuli ramping, e.g. temperature or O_2 partial pressure, delivered by the Climate in-situ gas holder (DENS™), the aim will be to maximize the spatial resolution while maintaining a temporal resolution in the 10-3 s range. For this, the study will make use of a direct electron detection K3 Gatan™ camera and it will be focused on electron diffraction and EELS spectroscopy data.

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Atomically Sharp Domain Walls in an Antiferromagnet

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Efficient manipulation of antiferromagnetic (AF) domains and domain walls has opened up new avenues of research towards ultrafast, high-density spintronic devices [1,2]. AF domain structures are known to be sensitive to magnetoelastic effects, but the microscopic interplay of crystalline defects, strain and magnetic ordering remained largely unknown. Recently, we have explored antiferromagnetic CuMnAs thin films in which imaging by x-ray photoemission electron microscopy (XPEEM) revealed that its AF domain structure is dominated by nanoscale crystalline defects [3]. The results emphasized the crucial role of these defects in determining the AF domains and domain walls, and provided a route to optimizing device performance in term of scaling limits for the data density in the bulk of the antiferromagnet. However, even smaller magnetic objects were indirectly observed in the material, but they remained below the detection limit of the used established XPEEM methods.
Here for the first time, we achieved atomic resolution imaging of abrupt AF magnetic domain walls in CuMnAs epilayers by utilizing scanning transmission electron microscopy (STEM), differential phase contrast (DPC) and 4D-STEM techniques [4]. Identification of the magnetic domain DPC signal is based on the specific symmetry of the CuMnAs crystal, where the opposite magnetic Mn sublattices occupy crystallographically distinct noncentrosymmetric sites.

With focus on small field-of-view high-resolution imaging, we could associate the DPC-STEM signals with two types of abrupt Néel vector reversals: The first type occurs at a crystallographic antiphase boundary defect, while the second type forms in a part of the epilayer with no crystallographic perturbation detectable by STEM, as schematically shows Fig. 1.

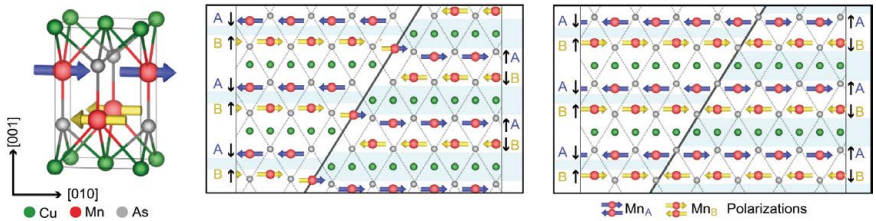

Fig 1. : CuMnAs unit cell, atomically sharp domain walls at antiphase boundary defect (left) and in unperturbed area (right) of the CuMnAs single crystal.

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SOLARIS Cryo-EM Facility at SOLARIS National Synchrotron Radiation Centre, Kraków, Poland

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SOLARIS Cryo-EM Facility at the SOLARIS National Synchrotron Radiation Centre is a modern and extensive research unit operating since 2019 in Krakow, Poland. Our priority research area revolves around the field of structural biology, where the usual goal is to understand the spatial structure of biologically relevant organic and inorganic molecules, and to understand their dynamics and interactions. We provide access to two high-end cryo-electron microscopes dedicated determining structures of biomacromolecules: Krios G3i (300 kV), and Glacios (200 kV), both ThermoFisher Scientific. In experiments, we use single particle analysis (SPA), cryo-electron tomography (cET) and microcrystal electron diffraction (MicroED) techniques allowing to analyse specimens at near-atomic resolution. **Figure 1.** demonstrates the research capabilities available at the SOLARIS Cryo-EM Facility.

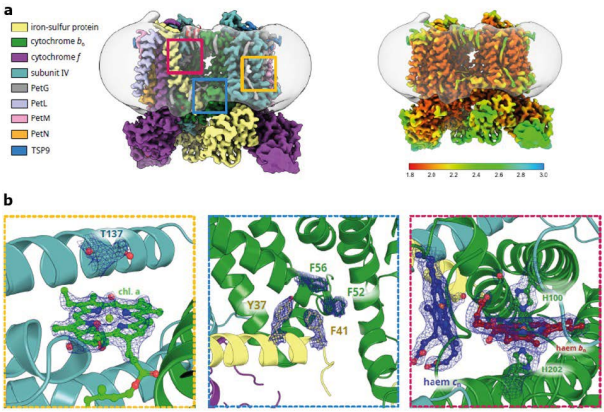

Fig 1. : An overview of the structure of cytb6f and cofactor.
a, 1.9 Å cryo-EM of DPQ-incubated cytb6f with color-coded subunits (left) and local resolution map coloured by resolution range (right);
b, close-up view of selected 1.9 Å map regions with remarkable resolution of amino-acid sidechains and cofactors. The colours of the frames correspond to regions marked by squares in **a**.

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Consistency and reliability of different ptychographic deconvolution approaches

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Knowledge of the electron probe is an important prerequisite for a successful ptychographic reconstruction as it is otherwise impossible to deconvolve the effects of specimen and illumination. While direct schemes such as the single sideband ptychography (SSB) are able to correct for lens aberrations, these have to be known beforehand [1]. Using iterative schemes, such as the extended ptychographic iterative engine (ePIE) [2], it is possible to optimize both the specimen and the probe at the same time. However, the retrieved probe is not limited to physical solutions, and it is difficult to gauge the reliability of a retrieved probe.

To investigate the consistency and quality of the retrieved probes a 4D-STEM focal series of a 2D SnS2 sheet was acquired with defoci between -20 and 20 nm. The datasets were first reconstructed using ePIE, and an iterative procedure was developed to fit probes to each result, parametrized by standard aberration coefficients. Figure 1 shows the reconstructed objects and the real space probes as well as the aberration function retrieved from ePIE, together with the respective fit results. For all investigated foci the agreement between retrieved and fitted probe is very good which can be recognized both in real space and in the phase plate. A plot of the defocus determined using the ePIE probe against the defocus set at the microscope shows the expected linear relationship (Figure 2). Additionally, the aberration coefficients for different foci are in very good agreement with each other throughout the series.

Consequently, the probes obtained by ePIE are physically reasonable electron probes and can be fully characterized using axial lens aberrations. The fitted aberration coefficients can be used as a start for other reconstruction schemes or to align the TEM. The applicability of the found aberration coefficients to other ptychographic schemes, such as SSB or gradient based methods, will be shown additionally. Probes and specimen retrieved using different reconstruction schemes will be compared and the influence of partial coherence is discussed.

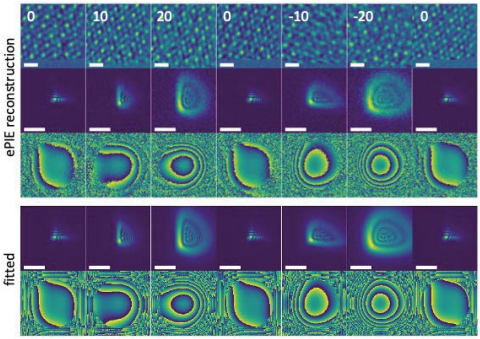

Fig 1. : Top: Phase grating (top) with foci (nm), probe (middle), aberration function (bottom) from ePIE. Bottom: Probe fit.

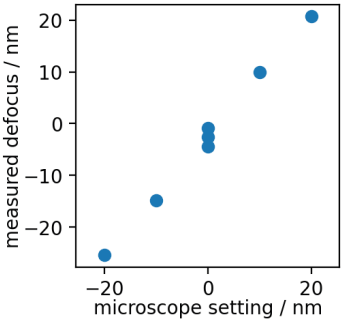

Fig 2. : Plot of the fitted defocus vs. the nominal one set at the microscope.

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32

Thickness-Dependent Behavior of Magnetic Domains in van der Waals Fe_3GeTe_2 during Magnetization Reversal

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The van der Waals (vdW) ferromagnet Fe3GeTe2 (FGT) has attracted interest for studying 2D magnetism due to its relatively high Curie temperature (200 - 220 K) [1] and demonstrated ability to host Bloch- and Néel-type skyrmions [2], [3]. Characterization of emerging magnetic vdW materials is essential for understanding fundamental magnetic behavior and achieving precise control of magnetic domains for potential applications in data storage and magnetic sensing. In these materials, external stimuli such as applied magnetic field and parameters such as sample thickness affect the intrinsic energy terms governing the formation and behavior of domains and nontrivial topological spin textures. Lorentz transmission electron microscopy (LTEM) enables high-resolution observation of magnetic domains while varying key conditions such as temperature and magnetic field. In this work, we use in-situ cryo-LTEM to image the field-driven evolution of magnetic domains in an exfoliated FGT flake with thickness from 20-100 nm. First, we apply a field-cooling procedure to nucleate Néel skyrmions, then we apply field in the negative direction to study the magnetization reversal.

We observe the formation of skyrmion lattices and their growth into unique faceted domain states. In thicker regions, initial skyrmion lattices are denser with smaller radii, and stronger fields are necessary to increase domain size and reach uniform reversal. Micromagnetic simulations closely reproduce the spin-reversal effect, highlighting the interplay between anisotropy, magnetic field, and field-cooling conditions. Our systematic study demonstrates high control of skyrmion size, density, and transitions to novel phases by tuning applied field and sample thickness of FGT, as well as contributing to a better understanding of the governing energy terms during magnetization reversal. This work was supported by U.S. Department of Energy (DOE), Office of Science, Office of Basic Energy Sciences, Materials Sciences and Engineering Division. Use of the Center for Nanoscale Materials, an Office of Science user facility, was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. DE-AC02-06CH11357.

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Atomic scale mapping of charge and lattice coupling at polar $LaAlO_3/SrTiO_3$ oxide interfaces by vibrational EELS

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The polar catastrophe occurring at the $LaAlO_3/SrTiO_3$ interface leads to the formation of an interfacial superconducting two-dimensional electron gas (2DEG) [1-2]. Yet, the nature of the pairing mechanism in such dilute limit is still under debate. Recent experiments and theoretical studies [3-4] suggest a pairing mechanism mediated by ferroelectric fluctuations through transverse optical phonons involving the formation of interfacial polarons. These polarons consist of electrons coupled with strongly polarized lattice distortions at the $SrTiO_3$ surface. Here, we address these phenomena using atomically-resolved monochromated STEM-EELS in combination with first principles calculations to reveal the coexistence of inversion-symmetry breaking and electron-phonon interaction at $LaAlO_3/SrTiO_3$ superconducting interfaces. By utilizing orthogonal off-axis beam-detector geometries, we are able to observe coupling of electrons with the longitudinal and transversal optical modes of STO within the 2DEG and decompose the directionality of the atomic vibrations with respect to the interface plane. We reveal the emergence of localized vibrational modes near the interface, exhibiting strong interaction with electrons upon increasing the interfacial doping. Our findings provide new key insights into the understanding of the origin of superconductivity at quantum paraelectric oxide interfaces.

This research benefited from resources and supports from the Electron Microscopy Center at the University of Chinese Academy of Sciences, and from the Beijing Outstanding Young Scientist Program (BJJWZYJH01201914430039), the National Key R&D Program of China (2018YFA0305800) and the National Natural Science Foundation of China (52373231).

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34

In situ study of the 2D Electron Gas in p-GaN/AlGaN/GaN High Electron Mobility Transistor

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Wide band gap (WBG) semiconductors like gallium nitride (GaN) and related compunds like aluminium gallium nitride (AlxGa(1-x)N) are deeply investigated for power electronics because of their capability to sustain high current, voltage and power density. These III-V materials possess a spontaneous polarization because of their noncentrosymmetric structure. Their pseudomorphycally, strained heterostructures (eg: AlGan/GaN) shows also a piezoelectric polarization. The resulting electric field induces high charge densities at the interface, leading to the formation of a two-dimensional electron gas (2DEG) confined in the GaN side of the interface. In an AlGaN/GaN High Electron Mobility Transistor (HEMT), the GaN channel exploits the high sheet carrier concentration and mobility of the 2DEG. To achieve a normally-off behaviour a p-GaN gate is added to the device, leading to a p-GaN/AlGaN/GaN structure where the presence of the 2DEG can be modulated by applying a potential between gate and source electrodes.

In a Scanning Transmission Electron Microscope (STEM), the presence of the 2DEG near the interface of the AlGaN/GaN heterostructure is expected to induce small deviations of the electron beam, which can be studied with 4D-STEM techniques. However, STEM analysis of interface of strained heterostructures typically shows also diffraction contrast which is superimposed on the 2DEG effect. Our work aims to discriminate the 2DEG effect on the electron beam by switching the 2DEG between the on and off state, exploiting the capability of in situ TEM biasing and 4D-STEM techniques. Starting from a commercial p-GaN/AlGaN/GaN HEMT, we used a Focused Ion Beam (FIB) to extract a section of device. This lamella was transferred to a four-contacts FIB-optimized E-chip (Protochips) and ion beam induced deposition (IBID) of platinum was used to create electrical connections between device and E-chip electrodes. This allowed to verify the electrical behaviour of the device in situ which is at the basis for the following 4D-STEM analysis.

35

Linear dichroism in a scanning transmission electron microscope

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Electronic, orbital, and spin reconstructions of 3d transition-metal oxides enable the emergence of novel properties and exotic phases at the surfaces and interfaces of thin films. In ABO_3 perovskites, a metal ion in octahedral coordination creates a significant crystal field, splitting the 3d orbitals into two degenerate sets: eg states and t_2g states. Under strain or when symmetry is reduced at surfaces or interfaces, the degeneracy of these states is further lifted, leading to selective electron occupancy. Orbital polarization, which quantifies the difference in orbital occupancy, is typically measured using X-ray absorption spectroscopy (XAS) and X-ray linear dichroism (XLD). XLD, defined as the difference in absorption of light polarized parallel and perpendicular to an orientation axis, provides macroscopic insights but is limited to surface contributions, potentially overlooking changes at specific atomic sites within buried interfaces.

Here, we present a methodology to achieve Electron Linear Dichroism (ELD) with atomic resolution using electron energy-loss spectroscopy (EELS) in an aberration-corrected scanning transmission electron microscope (STEM). We applied this technique to mixed-valence manganite $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) thin films under different strain states, validating our approach against previously reported XLD results [1]. Our findings confirm a positive dichroic signal for LSMO under compressive strain and a negative signal under tensile strain. Additionally, we investigated the orbital occupancy in quantum well oxides, observing its evolution at the interface correlated with charge density.

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Outrunning Crystallization During Flash Melting and Vitrification: Determining the Critical Heating and Cooling Rates of Pure water

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Water can be vitrified if it is cooled at high rates, which makes it possible to outrun crystallization in so-called no man's land, a range of deeply supercooled temperatures where water crystallizes rapidly. Using time-resolved electron diffraction, along with different microsecond shaped laser pulses, we study the rates needed to outrun crystallization when both heating and cooling a thin-film sample of water, known as the critical heating rate and critical cooling rate. These insights play a critical role in a wide range of fields, from the cryo-preservation of biological samples to the studying of dynamic processes of proteins in cryo-electron microscopy. Using similar experimental methods, we have also shown the structure of water evolves smoothly through the rapid crystallization regime called 'No Man's Land' as water cools from room temperature to cryogenic temperatures. All these experiments help narrow down the range of possible explanations for the origin of the anomalous behavior of water and create new experimental methods to study the behavior of water as well as biological samples in water.

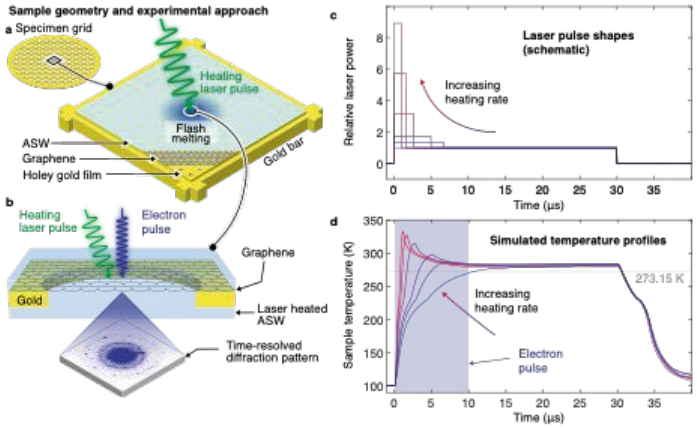

Fig 1. Constantin R. Krüger, Nathan J. Mowry, Marcel Drabbels, and Ulrich J. Lorenz *The Journal of Physical Chemistry Letters* 2024 *15* (16), 4244-4248
DOI: 10.1021/acs.jpclett.4c00315

Seeking peak precision in atomic EELS mapping with counting mode direct electron detection

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Since the advent of aberration-corrected scanning transmission electron microscopy (STEM) with electron energy-loss spectroscopy (EELS) spectrum imaging capabilities, atomic resolution STEM-EELS has become a vital technique for probing both elemental compositions and bonding states across epitaxial thin films.1 Nevertheless, data quality are often limited in terms of spatial and spectral precision. In large part, these limitations have derived from the charge-coupled devices that, until recently, have been the primary sensors used for EELS, and which have deficits of: poor detective quantum efficiency and point spread function; slow read out time and poor duty cycle; and correlated channel noise. Direct electron detection promises to address all of these deficiencies, at the same time. Here, we explore the benefits brought by direct electron detection using a large array monolithic active pixel sensor, with the goal of seeking maximum precision for both spectral peaks (EELS fine structures) and spatial peaks (atomic columns).
As test objects, we take superlattices of perovskite rare-earth nickelates, which constitute pure $SmNiO_3$ and alloyed $(Nd_{0.7}La_{0.3})NiO_3$ layers, grown on $LaAlO_3$.[2] Not only is it paramount to characterize their chemical nature for correctly interpreting their physical properties, these samples also provide interesting challenges for testing atomic-resolution STEM-EELS, such as the overlap of the rare-earth M edges with each other and with the Ni L edge. The STEM-EELS measurements are made using a Gatan Continuum ERS spectrometer equipped with a 3.4k channel K3 detector operated in counting mode, installed on a double-aberration corrected and monochromated FEI Titan Themis 60-300. Different acquisition strategies are tested: single-EELS, dual-EELS, single frame, multi-frame with on-line drift correction, multi-frame with off-line rigid and non-rigid registration. As an example of output, using a 5 integrated frame on-line drift-corrected dataset that is denoised using principal component analysis, we not only achieve atomic resolution maps across the full elemental range while preserving key EELS fine structure details, but also attain 1.2 Å resolution in the summed spectrum image signal and visualization of ± 20 pm antipolar displacements of Sm cations in the Sm map.

In conclusion, compared to legacy detectors, a latest-generation counting mode detector shows a step-change improvement for atomic resolution mapping of perovskite oxide heterostructures. While ideal acquisition strategies are still being refined, current results already demonstrate the potential for high precision mapping, both spectrally and spatially. Nevertheless, despite the nearly pure stochastic noise of the detector, initial tests show deficiencies in standard denoising approaches, stimulating a current search for more appropriate strategies.

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Characterising point defects in exfoliated $PtSe_2$

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Transition Metal Dichalcogenides (TMDs) are a class of two-dimensional (2-D) materials, comprising of a transition metal (M) and two chalcogen (X2) atoms, with the chemical formula of MX2 [1]. Platinum Diselenide (PtSe2), a group 10 TMD, has shown promise with its high room-temperature mobility, strong layer-dependent band structure and high stability in the air, thus presenting opportunities for applications as high-speed sensors and opto-electronic devices [2,3]. Processing of 2-D materials is known to have adverse effects on the material and thus needs to be well understood. Electron microscopy shines through as an essential tool in the thorough structural and chemical characterization in this regard.

In this study, the collaborative use of theoretical modeling via Density Functional Theory (DFT) and experimental characterisation through Scanning Transmission Electron Microscopy (STEM) was used to identify point defects present in PtSe2 nanoflakes. Bulk material was exfoliated through both liquid phase exfoliation (LPE) and mechanical exfoliation (ME) to achieve few layer flakes. Low-voltage aberration-corrected STEM images were recorded with a high angle annular dark-field (HAADF) detector on a Nion UltraSTEM.

Multiple point defects, including vacancies, substitutionals and various complexes, were identified in the exfoliated flakes through STEM imaging. Multislice STEM image simulations were performed to validate the cases using the abTEM code. The effect of these defects on the electronic structure of PtSe2 was then studied by first-principal calculations using DFT, while also calculating their subsequent formation energies. The structure-property correlation regarding the effect of defects on the thermoelectrical properties of PtSe2 was then investigated using the Boltzmann Transport Equation.

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STEM Characterization of TeO_2 Grown on CdTe Substrates: Understanding Possible Improvements to Back-Contact Passivation in CdTe Solar Cells

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Solar cell materials are an important part of current climate change mitigation strategies, necessitating a range of research into different suitable materials for different applications and niches. CdTe is a thin-film solar technology that has already seen some practical adoption; however, its efficiency currently lags behind that of Si, with an efficiency of 22.3% [1] compared to Si at 26.1% [2]. Currently, an area of research to improve device efficiency in CdTe is improving the electronic structure of the back-contact area. Forming an Ohmic back contact is difficult due to the high work function of CdTe; a passivation layer should help ameliorate the poor band structure and improve efficiency. TeO_2 has been shown to be a potential candidate for such a passivation layer. [3]

In this contribution, we study the nanoscale atomic and electronic structures of TeO_2 as deposited onto large-grain and polycrystalline substrates of CdTe. This is done to compare with DFT calculations and better understand the implications that it has on the band structure of the device. A thin cross-sectional lamella for use in a scanning transmission electron microscope (STEM) was prepared using a Thermo Fisher Helios 5CX focused ion beam. STEM imaging and elemental characterization was done using a JEOL JEM-ARM200CF at 200 kV, which is equipped with an Oxford XMAX100TLE energy-dispersive X-ray (EDX) spectrometer and a Gatan Quantum GIF detector for electron energy loss spectroscopy (EELS). [4-5]

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5. This material is based upon work was in part supported by the U.S. Department of Energy's Office of Energy Efficiency and Renewable Energy (EERE) under the Solar Energy Technologies Office Award Number DE-EE0008974 and funding from the Alliance for Sustainable Energy, LLC, Managing and Operating Contractor for the National Renewable Energy Laboratory for the U.S. Department of Energy.

TEM analysis of Textured Hexagonal Silicon Crystals obtained via Nanoindentation

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Hexagonal Silicon is particularly attractive for its application in light emission technologies and for the manipulation of its electronic properties with applied strain. While the fabrication of Hexagonal Silicon has been extensively demonstrated for nanowires and complex deposition systems, its synthesis by pressure, nanoindentation and annealing has been only partially explored.
In this poster, we investigate a system of nanoindented Silicon (100) subsequently annealed to induce a phase transformation to hexagonal Si. The system has been studied combining state-of-the-art characterization techniques, including electron energy loss spectroscopy (EELS) and transmission electron microscopy (TEM) that reveal a crystallographic texture of the hexagonal silicon crystals together with an energy shift in the plasmon response with respect to the surrounding cubic phase.

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41

Atomic-scale chemical and spin-state analysis of one-dimensional magnetic nanochains inside carbon nanotubes

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One-dimensional van der Waals materials—such as nanowires, nanorods, and nanotubes—provide an exceptional platform for understanding novel physical and chemical phenomena in low-dimensional systems. These materials exhibit remarkable electronics, photonics, thermal, and magnetic properties as the ultimate limit in material downscaling is reached. Nonetheless, these one-dimensional materials are susceptible to electron beam irradiation, which hinders their comprehensive characterization via electron microscopy.

To address this, carbon nanotube (CNT) encapsulation emerges as an effective technique, enhancing the stability of the encapsulated materials under electron beam exposure and facilitating their synthesis. In this study, we successfully image CNT-encapsulated one-dimensional magnetic nanochains (CrX2, where X=Cl or Br) at the atomic level. Moreover, employing a direct electron detector enabled us to perform atomic-level STEM-EELS mapping under 60 kV, facilitating the analysis of different elements and the spin-state of Cr atoms in CrX2 single chains within the CNT. Notably, both CrCl2 and CrBr2 exhibit similar spin-state, suggesting that different neighboring halogen atoms does not markedly influence the spin-state of Cr in the CrX2 compound. The experimental spin-state measurements agree well with density-functional theory (DFT) calculations, showing the power of low-voltage aberration-corrected STEM equipped with a direct electron detector in studying the properties of one-dimensional materials.

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Machine Learning Based Characterization of Active Dopant Concentration in Si with EELS

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Dopant Engineering is a key aspect of semiconductor device fabrication, dominating the electrical properties of the material. In particular, the "active" dopant concentration which can contribute to the carrier concentration can greatly influence the performance of the semiconductor device. Characterizing dopant itself has already been a challenging task due to the minute amount of dopant atoms and the low Z number for common dopant species such as B and P. It poses a bigger challenge to differentiate the active dopant concentration from the chemical dopant concentration. Traditionally, electrical measurements [1], e.g. Hall effect measurements, are used to determine the active dopant concentration. However, as the geometry of the devices becomes smaller and more complex, such as FinFET or Gate-All-Around devices, high spatial resolution techniques are required to characterize the active dopant concentration. Previously, off-axis electron holography has been used to quantify the active dopant concentration [2]. Off-axis electron holography is limited by the requirement of reference region directly adjacent to the region of interest and it is highly susceptible to thickness and alignments.

In this presentation, we propose a machine learning based approach to predict the active dopant concentration in Si from EELS data. The training dataset contains Si L edge spectra measured from 4 Si thin films with different Phosphorous doping levels. 80 nm x 200 nm Spectrum images were acquired with 2 nm step size, covering 40 nm doped film and 40 nm Si substrate, which we processed [3] and will use as the test data. A support vector machine [4] was trained to regress the active dopant concentration from the EELS spectra. Before training, the dataset was split into training and validation sets. The model was also validated by removing one of the samples from the dataset and predicting the active dopant concentration in that sample. The ability of the trained models to predict active dopant values will be demonstrated as well as current limitations, which will be discussed. The result indicates that near edge structure of Si L edge contains information about the active dopant concentration in Si and deserves more experimental and theoretical investigation.

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CEFID: A flexible platform for spectroscopic experiments

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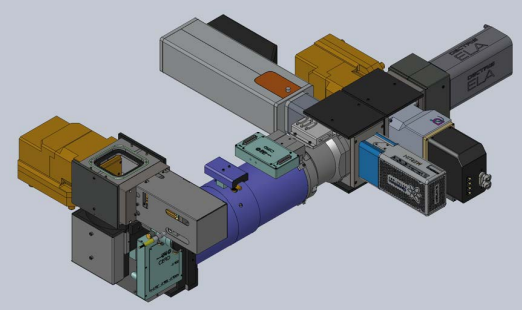
TEM-related instrumentation is developing rapidly, and a state-of-the-art experiments involve an increasingnumber of third-party components ranging from advanced in-situ specimen holders and new generation detectors to pulsed lasers and programmable phase plates. The CEOS Energy Filtering and Imaging Device (CEFID) is an energy filter and spectrometer offering state-of-the-art specifications and the flexibility to implement ambitious and unprecedented experiments.

Using a separation of function that mirrors that of a TEM itself, the filter design comprises highly optimised and stable optics up to the energy-selecting slit, and a flexible and minimalist projective stage [1]. This gives high performance and stability, while allowing to hop between modes (imaging, spectroscopic dispersions) and different detectors with little to no re-tuning. The Python/Qt-based GUI aka “Pantha Rhei” is used for the filter operation and implements interactive and automated procedures for alignments, common workflows ranging from spectroscopic imaging to 4D-STEM, and tools for on-the-fly analysis such as live DFT, EELS maps computation/quantification, Center-of-Mass, etc. The software is highly extendable and offers a scripting and plug-in API in python and a remote control interface for integration into third-party software such as LabView or MATLAB. The CEFID is compatible with the columns of the three major (S)TEM manufacturers and a wide range of detectors (see figure) and scan generators has already been integrated to use it for both, CEFID tuning and data acquisition. The CEFID/PantaRhei plattform is ideal for rapid acquisition and evaluation of spectroscopic data on daily basis [2], but also provides sufficient flexibility and compatibility for complex experiments where different tools need to work in unison, such as synchronizing acquisition with in-situ stimuli, or PINEM [3].



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44

A Machine Learning Approach for Automatic Detection and Classification of Defects in Atomic Resolution STEM Images

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Over the past few decades, electron and scanning probe microscopies have become crucial tools for investigating matter at the atomic and mesoscale levels. Advances in detectors, sensors, electron sources, and data storage have significantly increased the quantity and quality of structural and spectral data, thereby providing extensive information on material structures and functionalities. This data is usually analyzed manually to locate and measure defects, as these defects have a huge impact on the electronic, optical, and ferroic properties of the materials [1]. However, manual analysis of this vast amount of data is becoming increasingly impractical, time-consuming, and susceptible to inaccuracies. To address this issue, recent studies have explored the use of computer-vision techniques to automate defect detection in STEM and STM images [2].

We propose an unsupervised machine-learning algorithm based on an advanced neural network architecture known as a Convolutional Variational Autoencoder (CVAE) to automatically detect and highlight defects in STEM images. This architecture does not require any prior knowledge of bulk structures or defects, uses only a minimal training set, and applies to all periodic atomic structures. The architecture enables the algorithm to robustly learn and predict the bulk structure, which remains unaffected by local perturbations (defects). We find that the difference between the bulk image and its prediction is negligible, whereas the difference between an image with defects and its prediction is significant. Our study focuses on anomaly detection in atomic-resolution HAADF images of SrTiO3 and CdTe, with the potential to identify anomalies in all crystalline materials, provided that a sufficiently large area of bulk material is available for training the CVAE network.

In contrast to supervised ML algorithms, the unsupervised neural network architecture to be discussed here can also classify and cluster STEM images with a high level of confidence based on their visual similarities without any prior labeling of large datasets. [3]

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Atomic-resolution mapping of phonon modes across crystallographic shear structures in thermoelectric (Al,Nb)-doped TiO_2

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Phonons play a critical role in many physical properties of a materials including their thermal and electrical conductivities. Changes in normal phonon mode frequencies occur in the presence of defects. In thermoelectric materials (TE), such defect-induced localised modification of the vibrational response is widely used to tailor the thermal conductivity [1]. In previous work [2,3] it was shown that atomic-level defect engineering resulted in the enhancement of the TE performance of (Al,Nb)-doped TiO_2. The introduction of crystallographic shear (CS) structures leads to the reduction of lattice thermal conductivity, which is believed to occur through enhanced phonon scattering. Therefore, it becomes important to measure the spatial distribution and dispersion of the localized vibrational response across the CS structure, in order to gain insight into the heat-conduction process. Recent advances in scanning transmission electron microscopy (STEM) and electron energy-loss spectroscopy (EELS) have provided powerful and flexible tools to study phonons at high spatial resolution, down to single atom sensitivity [4]. In this work, we use STEM-EELS to probe the localised phonon response in CS structures at atomic resolution in the polar (Al,Nb)-doped TiO_2.

EELS measurements were performed on a Nion UltraSTEM100MC 'Hermes' scanning transmission electron microscope, equipped with a Nion IRIS high energy resolution EELS spectrometer with a Dectris ELA direct electron detector. The acceleration voltage was 60 kV and the probe convergence semi angle was 31.5 mrad, resulting in a 1 Å probe size. The experimental optical geometry follows the conditions in Ref [4], in which the off-axis, or dark-field EELS (DF-EELS) geometry significantly reduces the contribution to the EELS signal of electrons having undergone delocalized dipole scattering, while promoting that of localized impact phonon scattering. This approach enables in principle single-atom sensitivity of phonon scattering.

Across the CS structures, vibrational EELS measurements successfully and atomically resolved different phonon modes that are localized on specific atomic sites. These phonon modes showed a sensitivity in momentum space and a link to chemical bond. Results are discussed in context of the local structure and chemistry, determined at the same position through careful chemical mapping, while phonon map simulations using the frequency-resolved frozen phonon multislice method [5] are used to rationalize the experimental findings.

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Electron microscopy at CNR-IMM and it's efforts for experimental data trustworthy and FAIR trough NFFA-DI consortium

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NFFA-DI is a projet for realizing a Full-Spectrum Research Infrastructure for nanoscience and nanotechnology, which integrates cutting-edge instrumentation, computational resources and curation of scientific data according to the FAIR (findable, accessible, interoperable and reusable) and open science principles.

At presente, the reference repository in the field of material science is NOMAD, a free and open-source data management platform developed by FAIRmat consortium, and the common international standard data format is NeXus, built implementing basic Hierarchical Data Format (HDF5) storage elements.

Since experiments nowadays create a set of very often voluminous and diverse numerical data and metadata in many data format, NFFA-DI infrastructure will be capable of enhancing the Italian research competitiveness on the fundamental interactions of multi-atomic matter creating a global system for managing research data through the application of Fair-by-design technology to all experimental and computational resources.

CNR-IMM focused on this topic making data coming from 2 of its electron microscopes (HELIOS 5 UC and JEOL 2010) conform to NeXus format ad easy to upload on NOMAD running a python script also ables to read additional sample metadata from an internal Google Sheet.

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