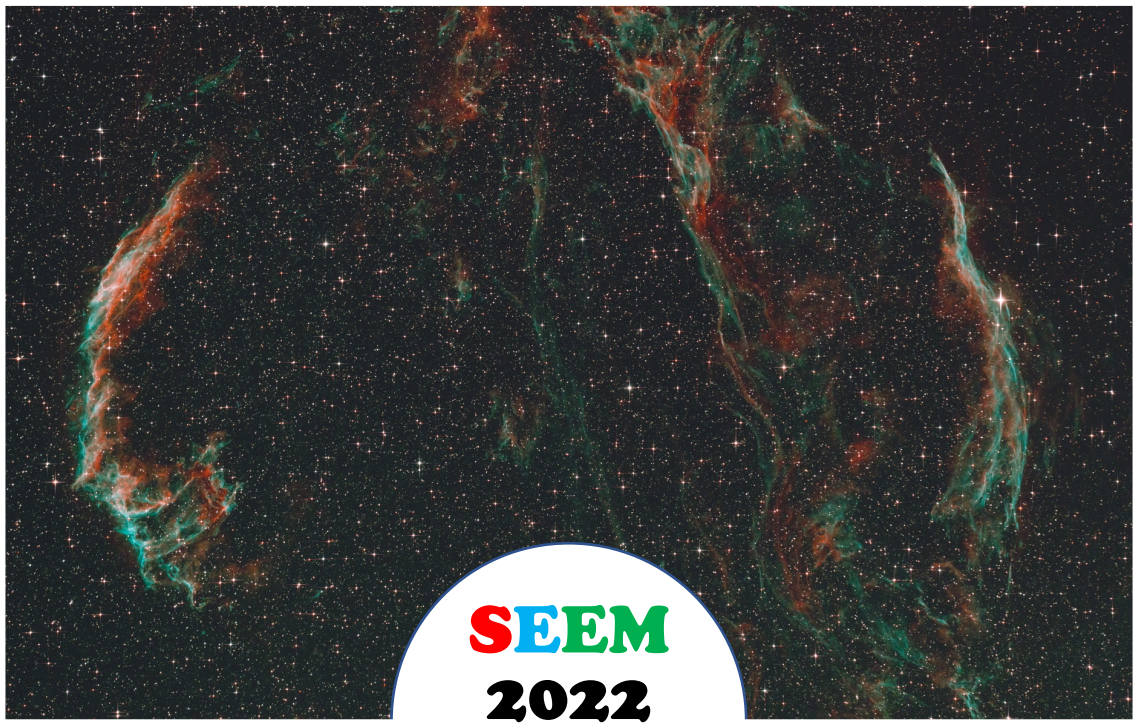
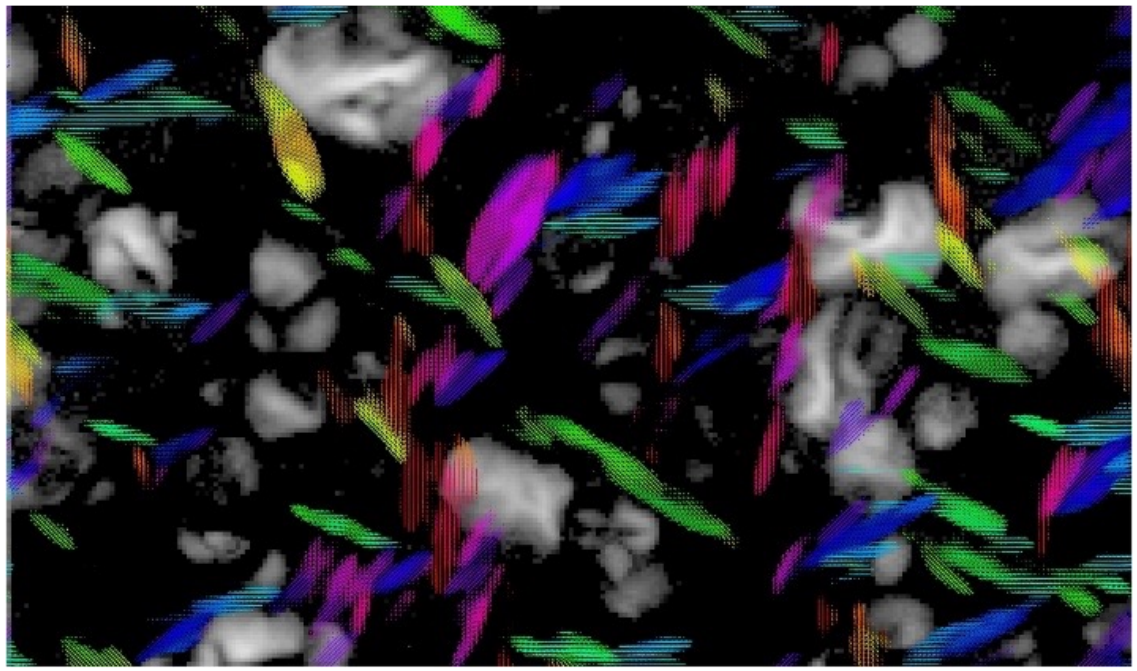


SEEM 2022

3rd Sino-European Early Career Researchers Virtual Workshop on Electron Microscopy in the Era of Energy Transition

Sponsor-free | Non-profit | Free registration

07 – 08 December 2022 | Online



Preface

Following the successful past two Sino-European Early Career Researchers Virtual Workshop on Emerging Techniques and Applications in Electron Microscopy (SEEM) that attracted 300+ registrations from 20+ countries and 100+ organisations in 2020 and 2021, respectively, we are pleased to announce that the 3rd of this series workshop, **SEEM 2022**, will take place again in virtual format on **07 - 08 December this year**.

In the past year, much of the globe has faced shortages and soaring prices in energy supply in the aftermath of the pandemic, the war, as well as a variety of factors. This unfortunately slows down the transition process to more sustainable energy sources. In view of this, we slightly adjust the theme of this year's workshop to **Electron Microscopy in the Era of Energy Transition** and will focus on **energy-related applications (Session I, first day) as well as cutting-edge methods (Session II, second day) using advanced electron microscopy**.

This workshop is sponsor-free, free of charge for participation, and aims to offer a bilateral forum for electron microscopists in Europe and China to discuss the latest progress. Each scientific session consists of six to seven invited talks given by early career researchers and young PIs.

The programme also features a topical forum, **Avenue towards the next phase - career advancement for early stage researchers**. A distinguished scientist and a senior research adviser will share their perspective with the participants followed by a live panel discussion.

We cordially invite colleagues to join us via a free registration at **http://shorturl.at/iGI19** (or scan the QR code below) and look forward to spending an inspiring time with you!

Sincerely,

Pei Liu (Technical University of Denmark, Denmark)
Penghan Lu (Research Centre Juelich, Germany)
Mingjian Wu (University of Erlangen-Nuremberg, Germany)

SEEM 2022
3rd Sino-European Early Career Researchers Virtual Workshop on Electron Microscopy in the Era of Energy Transition
07 - 08 December 2022

Day I	**Wednesday 07 December 2022**
Session I	**Energy-related applications**
Session chair	**Penghan Lu** (Research Centre Juelich, Germany) **Pei Liu** (Technical University of Denmark, Denmark)

Europe (CET)	China	Speaker & Title
08:30 - 08:40	15:30 - 15:40	Welcome remarks
08:40 - 09:05	15:40 - 16:05	**Xuefeng Wang** (Institute of Physics CAS, China) **Cryo-EM for battery materials and interfaces**
09:05 - 09:30	16:05 - 16:30	**Jinfei Zhou** (Chongqing University, China) **Cryo-FIB enables atomic-resolution imaging of local structures in highly sensitive bulk crystals and devices**
09:30 - 09:55	16:30 - 16:55	**Mathias Rothmann** (University of Oxford, UK) **Atomic-scale microstructure of metal halide perovskites elucidated via low-dose STEM**
09:55 - 10:15	16:55 - 17:15	Break
10:15 - 10:40	17:15 - 17:40	**Xiaoxu Zhao** (Peking University, China) **Unveiling atomic-scale Moiré features and atomic reconstructions in twisted bilayer 2D materials**
10:40 - 11:05	17:40 - 18:05	**Zhongtao Ma** (Technical University of Denmark, Denmark) **Electrochemical impedance spectroscopy combined ETEM study of solid oxide materials**
11:05 - 11:30	18:05 - 18:30	**Andreas Hutzler** (Research Centre Juelich, Germany) **Towards quantitative analysis of CO2 upcoversion via *operando* electrochemical liquid-phase TEM**
11:30 - 11:35	18:30 - 18:35	**Virtual group photo**

End of Session I (Day I to be continued)

Day I	**Wednesday 07 December 2022**
Forum	**Avenue towards the next phase - career advancement for early stage researchers**
Forum chair	**Pei Liu** (Technical University of Denmark, Denmark) **Mingjian Wu** (University of Erlangen-Nuremberg, Germany)

Europe (CET)	China	Speaker & Title
13:00 - 13:05	20:00 - 20:05	Introduction
13:05 - 13:40	20:05 - 20:40	**Dorte Juul Jensen** (Technical University of Denmark, Denmark) Distinguished scientist, Chairman of evaluation panel of many prestigious grants (including ERC Advanced Grant, 2021) **Elements of what it takes to become a successful scientist – from a personal perspective**
13:40 - 14:15	20:40 - 21:15	**Mette Christiansen** (Technical University of Denmark, Denmark) Senior research adviser on project administration and fundraising (including ERC, Marie Skłodowska-Curie, DFF, private Danish foundations) **Get a competitive CV**
14:15 - 14:40	21:15 - 21:40	Panel discussions

End of Day I

Day II	**Thursday 08 December 2022**
Session II	**Cutting-edge methods**
Session chair	**Mingjian Wu** (University of Erlangen-Nuremberg, Germany) **Penghan Lu** (Research Centre Juelich, Germany)

Europe (CET)	China	Speaker & Title
08:30 - 08:35	15:30 - 15:35	Introduction
08:35 - 09:00	15:35 - 16:00	**Ruochen Shi** (Peking University, China) **Measuring lattice dynamics by four-dimensional EELS**
09:00 - 09:25	16:00 - 16:25	**Yves Auad** (Paris-Saclay University, France) **Recent developments in nanosecond spectroscopies in a continuous-gun electron microscope**
09:25 - 09:50	16:25 - 16:50	**Andrea Konečná** (Brno University of Technology, Czech Republic) **Electron beam shaping using optical fields**
09:50 - 10:15	16:50 - 17:15	**Lewys Jones** (Trinity College Dublin, Ireland) **Scan smarter not harder: acquisition design in the STEM**
10:15 - 10:35	17:15 - 17:35	Break
10:35 - 11:00	17:35 - 18:00	**Haozhi Sha** (Tsinghua University, China) **Correct misorientation via adaptive propagator multislice ptychography**
11:00 - 11:25	18:00 - 18:25	**Thomas Friedrich** (University of Antwerp, Belgium) **Phase object reconstruction for 4D-STEM using deep learning**
11:25 - 11:50	18:25 - 18:50	**Jacob Madsen** (University of Vienna, Austria) **abTEM: A fast and flexible Python-based multislice simulation package for transmission electron microscopy**
11:50 - 12:00	18:50 - 19:00	Closing remarks
End of the Workshop		

Cryo-EM for battery materials and interfaces

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The emerging cryogenic electron microscopy (cryo-EM) has demonstrated its power and essential role in probing the beam-sensitive battery materials and delivering new insights. With the increasing interest in cryo-EM for battery materials and interfaces, herein we provide the strategies of obtaining fresh and native structural information with minimal artifacts, including sample preparation, transferring, imaging, and data interpretation. We show some of our recent achievements enabled by cryo-EM in terms of the Li metal, graphite, Si, and solid electrolyte interfaces. The advance of cryo-EM is expected to make contributions on probing the structures of battery materials and interface, understanding the failure mechanism, and thus facilitating to development of the higher-energy and safer batteries.

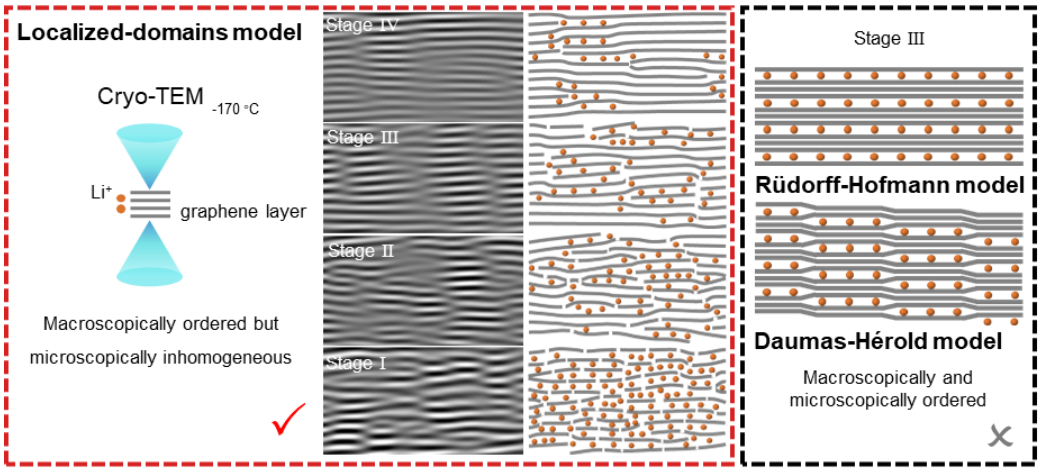

Figure 1. Proposed Localized-domains model for the staging structure of Li-intercalated graphite visualized by cryo-EM

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Cryo-FIB Enables Atomic-Resolution Imaging of Local Structures in Highly Sensitive Bulk Crystals and Devices

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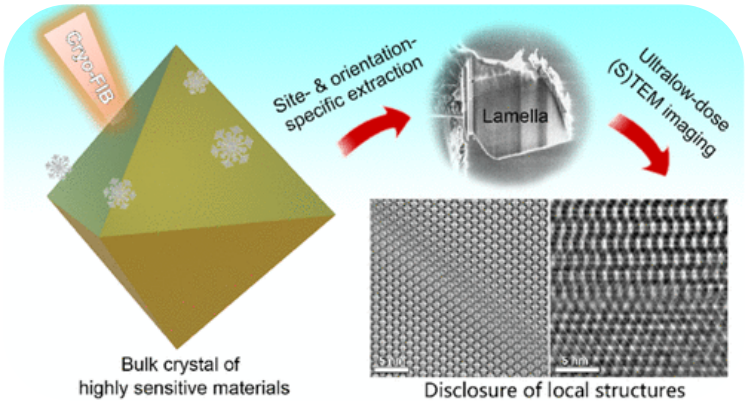
The (scanning) transmission electron microscopy ((S)TEM) specimen preparation is crucial for achieving high-resolution imaging and quantitative analysis when the sample is a bulk material. However, acquiring high-resolution TEM (HRTEM) images for electron-beam-sensitive materials, such as metal-organic frameworks (MOFs), supramolecular crystals, and organic-inorganic hybrid halide perovskites (HPs), is very challenging because their structures could easily be damaged by electron-beam irradiation. In this talk we will present our latest research progress using cryogenic focused ion beam (cryo-FIB) to prepare (S) TEM specimens of electron beam sensitive crystalline materials. We successfully extracted thin specimens from metal-organic framework (MOF) crystals and a hybrid halide perovskite single-crystal thin-film solar cell using cryo-FIB without damaging the inherent structures. The effectiveness of cryo-FIB in different application scenarios and its compatibility with various electron microscopy modes are also demonstrated. This proof-of-concept study demonstrates that cryo-FIB has a unique ability to handle highly sensitive materials, which can substantially expand the range of applications for electron microscopy.



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The atomic-scale microstructure of metal halide perovskites elucidated via low-dose (scanning) transmission electron microscopy

Mathias Uller Rothmann[1,2,7], Judy S. Kim[2,3,4], Juliane Borchert[1], Kilian B. Lohmann[1], Colum M. O'Leary[2], Alex A. Sheader[2], Laura Clark[2], Henry J. Snaith[1], Michael B. Johnston[1], Peter D. Nellist[2], Laura M. Herz[1], Wei Li[5,6], Ye Zhu[8], Weijian Chen[9], Chenquan Yang[5], Yongbo Yuan[10], Yen Yee Choo[7], Xiaoming Wen[9], Yi-Bing Cheng[5,6], Udo Bach[11,12,13,14], and Joanne Etheridge[7,15]

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In this talk, I will show how ultra-low dose electron microscopy makes it possible to elucidate fundamental crystallographic properties of hybrid perovskite thin films prepared using the same methods as used for devices, allowing us to study these properties in the native environment found in solar cell devices.

Firstly, we successfully imaged the archetypal $CH(NH_2)_2PbI_3$ ($FAPbI_3$) and $CH_3NH_3PbI_3$ ($MAPbI_3$) hybrid perovskites in their thin-film form with atomic resolution using a carefully developed protocol of low-dose HR-STEM.[2] Our images

enabled a wide range of previously undescribed phenomena to be observed, including a remarkably highly ordered atomic arrangement of sharp grain boundaries and coherent perovskite/PbI_2 interfaces, with a striking absence of long-range disorder in the crystal, as shown in Figure 1, explaining why inter-grain interfaces are not necessarily detrimental to perovskite solar cell performance.

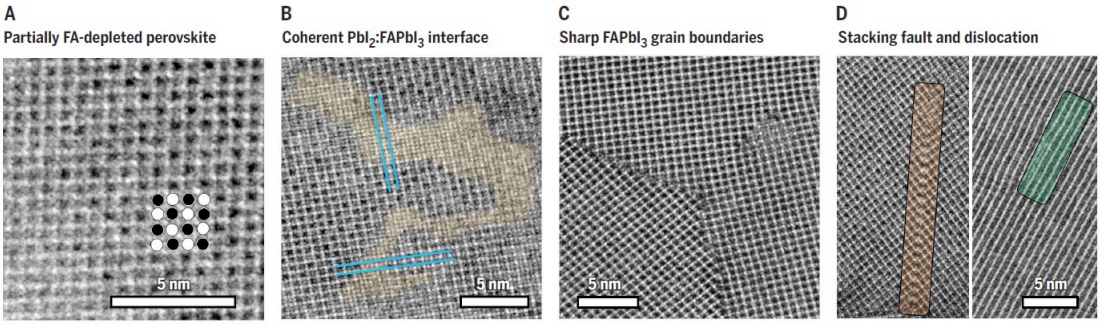

Figure 1: Crystallographic phenomena imaged by HR-STEM.

Secondly, we varied the methylammonium (MA)/formamidinium (FA) composition in $MA_{1-x}FA_xPbI_3$ (x=0-1), and compared the structure and density of intragrain planar defects with the device performance, otherwise keeping the device nominally the same. We found charge carrier lifetime, open-circuit voltage-deficit, and current-voltage hysteresis correlate empirically with the density and structure of $\{111\}_c$ planar defects (x=0.5-1) and $\{112\}_t$ twin boundaries (x=0-0.1). The best performance parameters are found when essentially no intragrain planar defects are evident (x=0.2).[3]

Our findings provide an atomic level understanding of the technologically important class of hybrid lead halide perovskites, revealing several mechanisms that underpin their remarkable performance.

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Unveiling Atomic-Scale Moiré Features and Atomic Reconstructions in Twisted Bilayer 2D Materials

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Twisting the angle between van der Waals stacked 2D layers has recently sparked great interest as a new strategy to tune the physical properties of the materials. The twist angle and associated strain profiles govern the electrical and optical properties of the twisted 2D materials, but their detailed atomic structures remain elusive. Herein, using combined atomic-resolution electron microscopy and density functional theory (DFT) calculations, we identified five unique types of moiré features in commensurately twisted $\sqrt{7}a \times \sqrt{7}a$ transition metal dichalcogenide (TMD) bilayers. These stacking variants are distinguishable only when the moiré wavelength is short. Periodic lattice strain is observed in various commensurately twisted TMD bilayers. Assisted by Zernike polynomial as a hierarchical active-learning framework, a hexagon-shaped strain soliton network has been atomically unveiled in nearly commensurate twisted TMD bilayers. Unlike stacking-polytype-dependent properties in untwisted structures, the stacking variants have the same electronic structures that suggest twisted bilayer systems are invariant against interlayer gliding [1-2].

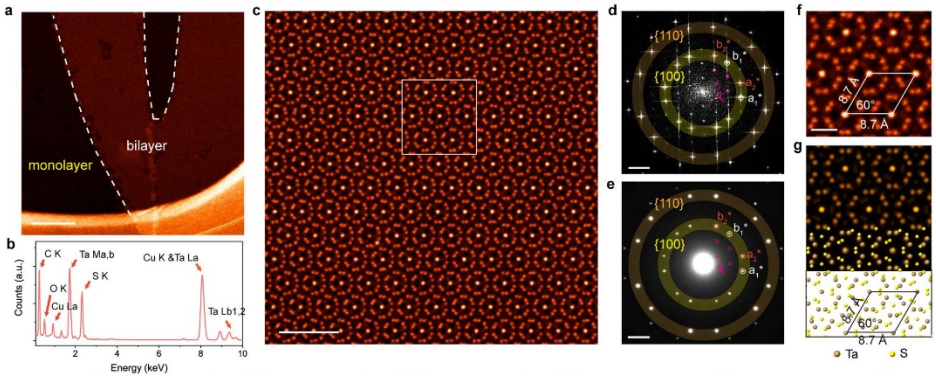

Figure 1. Commensurate superlattices in twisted TMD bilayers showing Σ = 7 superlattices.

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Electrochemical Impedance Spectroscopy combined ETEM study of solid oxide materials

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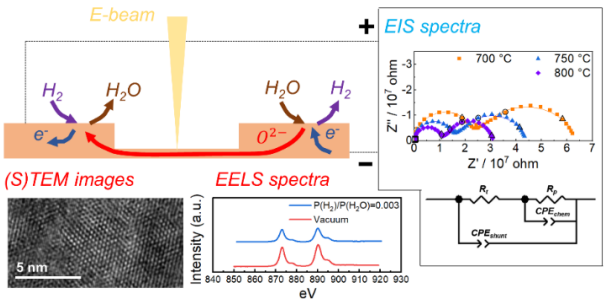
Developing related fuel production and storage technologies for sustainable energy becomes increasingly important. Solid oxide electrolysis/fuel cell has recently become a research hotspot due to their super high energy conversion efficiency[1]. However, its degradation mechanisms at high working temperatures are still unclear, advanced characterization methods are necessary, especially *operando* techniques.

Electrochemical impedance spectroscopy (EIS) is a classic technique in electrochemistry, and one of its advantages allows *operando* characterization on a running system without causing damage to the research object[2]. By fitting the EIS data with equivalent electric model circuit (ECM) of the electrochemical system, one can get the contributions of different processes in the reaction.

Environmental TEM can inlet different gases into the column[3], with a DENSsolutions lighting holder, heating and biasing can be also applied to the sample. Combining simultaneous EIS signal, one can investigate the structural/elemental evolutions and their effect on the electrochemical performance in real environments of running a cell.



In this work, we investigated the feasibility of performing EIS in ETEM. Mixed ionic and electronic conductor $Ce_{0.9}Gd_{0.1}O_{1.95-\delta}$ (CGO) was chosen as a model material[4]. Our preliminary results are consistent with the documented data in terms of ionic conductivity, electronic conductivity, activity of surface-gas exchange reactions, and chemical capacities, which verified the reliability of the EIS-TEM test.

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Towards quantitative analysis of CO_2 upcoversion via *operando* electrochemical liquid-phase TEM

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Converting carbon dioxide to fuels is one way to cope with both, global climate, and energy crisis. A promising approach for carbon-utilization is electrochemical reduction of CO_2 to CO as a precursor for other compounds[1]. However, the catalysts still suffer from low stability and structural changes. This is related to an incomplete understanding of the catalyst behavior under operational conditions. The exact mechanisms underlying this transformation remain object of debate, impeding an effective prevention of catalyst degradation. Observation of these processes are enabled by state-of-the-art liquid phase TEM (LP-TEM) techniques by performing electrochemical experiments in liquid media *in situ*. However, inelastic electron-matter interactions can strongly affect the probed system. This can yield membrane charging that may affect quantitative specimen analysis[2], or trigger secondary redox processes, even adjacent to the irradiated volume[2,3]. Moreover, the possibility of beam-induced heating needs to be accounted for[4]. Yet, most relevant to LP-TEM is radiolysis of the liquid itself, which does not only influence the redox-chemistry under observation[5], but also disables the practical applicability of pH[6]. However, utilizing holistic radiation chemistry simulations allows for quantitative assessment of the electron beam influence on the chemistry under observation[3]. Consequently, beam-induced artifacts can not only be understood, but also harnessed to trigger desired phenomena. In combination, this allows for drawing quantitative conclusions of LP-TEM studies of *operando* studies of catalytic CO_2-upconversion that are directly applicable to flask-chemistry.

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Measuring Lattice Dynamics by Four-dimensional EELS

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Phonons – quasiparticles describing lattice vibrations – play an essential role in various properties of solid-state systems, such as the thermal conductivity, electron mobility, structural stability, and conventional superconductivity. At the interface between two crystals, new phonon modes distinct from two bulk materials can emerge due to the breakdown of translation symmetry. Phonon modes localized at interfaces were predicted to exist decades ago, but they have eluded direct experimental observation to date because they are confined to only within several atom layers near the interface. To probe them, nanometer spatial resolution, millielectronvolt energy resolution and ultrahigh detection sensitivity are simultaneously required; to further measure their dispersion relation and related properties, momentum resolution is also needed. Traditional spectroscopic techniques cannot meet these stringent requirements.

To solve this problem, we developed an electron energy loss spectroscopy technique (termed 4D-EELS) based on a scanning transmission electron microscope. This technique offers high spatial, energy and momentum resolutions with high detection sensitivity, enabling nanoscale phonon dispersion measurements [1]. It can achieve a good balance between spatial and momentum resolutions, only ~15% away from the diffraction limit. Using a cubic boron nitride / diamond heterointerface as an example [2], we first measured the local phonon density of states at the atomic scale, and successfully observed phonon modes localized at the interface and phonon modes isolated from the interface. Then we measured the dispersion relation of interface modes with carefully balanced spatial and momentum resolutions. The experimental observations are in good agreement with first-principle calculations. The observed interface modes are expected to substantially affect thermal conductance across the interface and carrier mobility of the two-dimensional electron gas at the interface. The ability to measure local phonon dispersion should also be useful in studying topological interface phonon modes and even in understanding interface superconductivity.

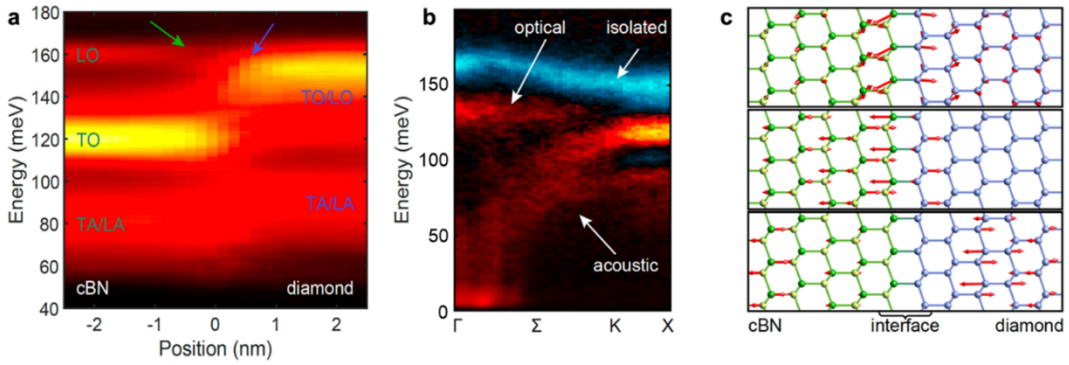

Figure 1. (a) Local phonon spectrum across the interface. (b) Dispersion relation of interface phonon modes. (c) Phonon eigenvectors of representative modes with enhanced and reduced vibration amplitude at the interface.

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Recent developments in nanosecond spectroscopies in a continuous-gun electron microscope

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Recently, the scanning transmission electron microscope (STEM) has been profoundly transformed by the advent of a new generation of electron monochromators, down to 2-5 meV spectral resolution, opening the way for new applications using electron energy loss spectroscopy (EELS). More specifically, low-loss EELS can be performed with unprecedented spectral resolution down to the infrared range [1], an energy window almost unexplored so far in an electron microscope, thus paving the way for new studies in nano-optics. Additionally, other spectroscopic techniques, combining photons and electrons, can be used in the electron microscope alongside EELS, i.e., cathodoluminescence, electron energy-gain spectroscopy, photon-induced near-field electron microscopy, etc., ultimately providing a rich and complex spectroscopic information of the sample. Further advances in STEM instrumentation are underway, and, in particular, time-resolved experiments can be critical for accessing new physical information, especially when combined with the many other techniques and advances aforementioned. Usually, time resolution is either limited by current camera technologies (in the ms range) or requires extremely fast blankers or the heavy instrumental development of a pulsed electron gun.

Here, we have taken a different path toward nanosecond-resolved EELS. We have developed a new acquisition scheme using the event-based Timepix3 direct electron detector (providing sub 5 ns temporal resolution) as our EELS detector. In this case, hyperspectral imaging can be reconstructed alongside typical single-channel imaging systems, such as annular dark-field and bright-field detectors, removing the long-lasting limitation of the readout speed, typical of frame-based measurements [2]. In parallel, we have also developed a new type of time-correlated experiments based on temporal coincidences between inelastic electron scattering and photon emission events. This technique, called cathodoluminescence excitation spectroscopy (CLE) as a counterpart to photoluminescence excitation spectroscopy (PLE), is capable of unveiling the excitation pathways in the sample (Figure 1), circumventing one of the major limits for EELS spectroscopy, i.e., the broadband nature of the energy exchange between a fast electron and matter [3]. Additionally, it will be shown that by combining event-based hyperspectral imaging with CLE, it is possible to study these decay pathways at the nanometer scale.

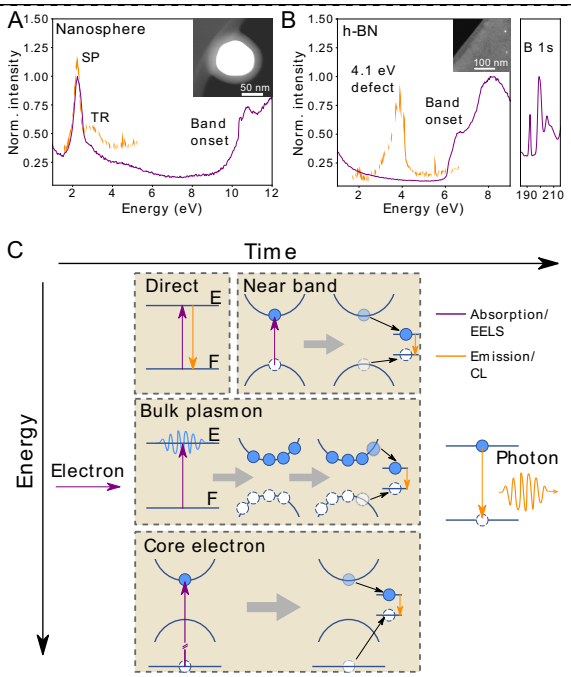

Figure 1: **Photon emission pathways upon electron scattering.** (A and B) Time-averaged CL (orange) and EELS (purple) spectra of an Au/SiO2 nanosphere and a thin h-BN flake show different absorption and emission features, described in the text. From these correlative time-averaged spectra, one cannot identify which absorption transitions lead to light emission. The small intensity emission at ≈2 eV in the h-BN CL spectrum is a replica of the 4.1-eV defect emission due to the diffraction grating. The insets show images of the nanosphere and the h-BN edge. CL and EELS spectra have been normalized and shifted vertically for clarity. (C) A relativistic inelastic electron scattering event in a solid can generate different excitations (vertical purple arrows): direct optical transition, NBE transition, bulk plasmon excitation, and core-level transitions. Excitations not involving single particles (excitons, bulk and SPs, etc.) are represented between a fundamental (F) and excited (E) state. These can relax through different pathways, leading to the excitation of a final optically bright energy level and to photon emission (vertical orange arrows).

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Electron beam shaping using optical fields

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Over the last decades, electron microscopy has become a powerful and versatile technique for nano- or atomic-scale imaging and spectroscopy. Major advancements have been made possible thanks to better spatial and temporal control over the amplitude and phase of the electron wave function, which is commonly achieved by means of complex arrangements of electric and magnetic lenses or static phase plates. We envision an alternative to traditional electron-optical elements, materialized in the concept of the optically-driven electron modulators that enable dynamical shaping of electron-beam wave functions both in space and time. This approach capitalizes recent experimental demonstrations of wave function control through optical fields [1], combined with ultrafast control over the electron-light interaction [2]. We propose two types of schemes: a photonic aberration corrector (PAC) that exploits the interaction of the electron with light scattered from a thin film; and an optical free-space electron modulator (OFEM) operating in free space. We demonstrate applications of the proposed setups in aberration correction [3] or generation of exotic electron beam shapes [3,4], which could be further exploited in novel acquisition schemes [5] or in generation of entanglement between electrons and sample excitations [6].

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Correct misorientation via adaptive propagator multislice ptychography

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Super-resolution imaging of solids is essential to explore the local symmetry breaking and derived material properties. Electron ptychography is one of the most promising schemes to realize super-resolution imaging beyond aberration correction. In order to reach both deep sub-angstrom resolution imaging and accurate measurement of atomic structures, however, it is still required for the electron beam to be nearly parallel to zone axis of crystals. Here we report an efficient and robust method to correct the specimen misorientation in electron ptychography, giving deep sub-angstrom resolution for specimens with large misorientations. The method largely reduces the experimental difficulties of electron ptychography and paves the way for widespread applications of ptychographic deep sub-angstrom resolution imaging.

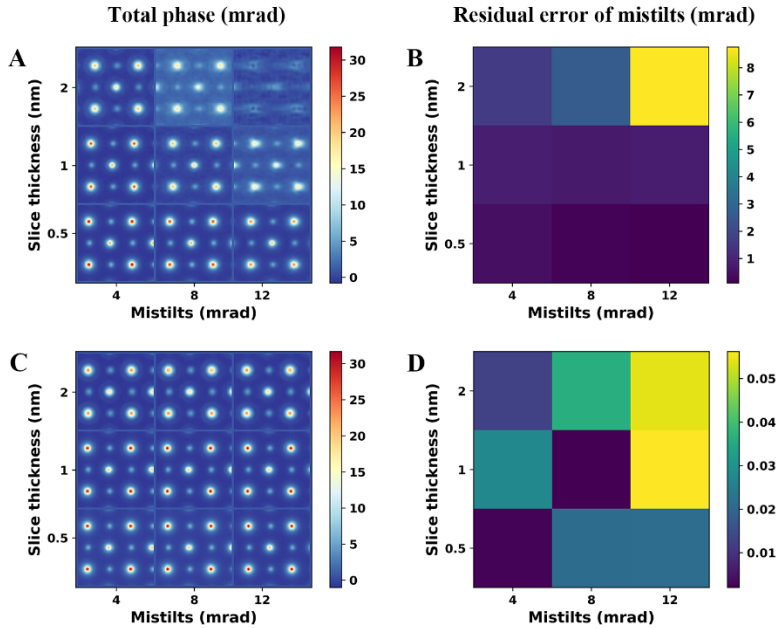

Figure 1. Figure for SEEM 2022

Reference

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Phase Object Reconstruction for 4D-STEM using Deep Learning

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In this study we explore the possibility to use deep learning for the reconstruction of phase images from 4D scanning transmission electron microscopy (4D-STEM) data. The process can be divided into two main steps. First, the complex electron wave function is recovered for a convergent beam electron diffraction pattern (CBED) using a convolutional neural network (CNN). Subsequently a corresponding patch of the phase object is recovered using the phase object approximation (POA). Repeating this for each scan position in a 4D-STEM dataset and combining the patches by complex summation yields the full phase object. Each patch is recovered from a kernel of 3x3 adjacent CBEDs only, which eliminates common, large memory requirements and enables live processing during an experiment. The machine learning pipeline, data generation and the reconstruction algorithm are presented. We demonstrate that the CNN can retrieve phase information beyond the aperture angle, enabling super-resolution imaging. The image contrast formation is evaluated showing a dependence on thickness and atomic column type. Columns containing light and heavy elements can be imaged simultaneously and are distinguishable. The combination of super-resolution, good noise robustness and intuitive image contrast characteristics makes the approach unique among live imaging methods in 4D-STEM.

*ab*TEM: A Fast and Flexible Python-based Multislice Simulation Package for Transmission Electron Microscopy

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The use of scattering simulations based on the multislice algorithm has, for decades, been an invaluable aid for transmission electron microscopy [1]. As a result, many excellent simulation codes exist; *ab*TEM is another one, however, our approach differs in key aspects [2]. The code is openly available: https://github.com/abTEM/abTEM.

*ab*TEM is a pure Python package, with effective use of open-source libraries making it highly performant. Its implementation focuses on the use of scripts and especially Jupyter notebooks [3], which inherently creates documentation and reproducibility of the simulations. A modular implementation makes *ab*TEM highly customizable.

*ab*TEM was recently reimplemented from the ground up with Dask [4]. Dask allows scaling from a laptop to thousands of nodes at high-performance computing facilities; this makes the code well-suited for diverse settings, from teaching to research.

*ab*TEM supports an expanding number of advanced features. These include accurate electrostatic potentials using density functional theory [5], a multi-GPU accelerated implementation of PRISM [6], multislice ePIE [7], and core-loss energy-filtered simulations [8]. In addition, we recently added an experimental implementation for plasmons [9], and we are working on vibrational EELS [10] and magnetism [11].

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SEEM 2022

Organized by

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Cover images: “Energy bursts from cosmic- to nano-scale”
(Top) The Veil nebula (NGC 6960, 6992): clouds of heated and ionized gases and dust found in Cygnus, which is remnant from a historical supernova explosion, between 10’000 and 20’000 years ago.
Image captured with an amature telescope from a balcony in Erlangen, DE.
(Bottom) Donor nano-crystallites in an active layer of organic solar cell. Color indicates the orientation of crystallites with their molecule lamella (so-called pi-stacking) edge-on, while gray clouds are face-on counterparts.
Image captured using 4D scanning confocal electron diffraction technique (Wu, et. al., Nat. Comm. 13 [2022] 2911).