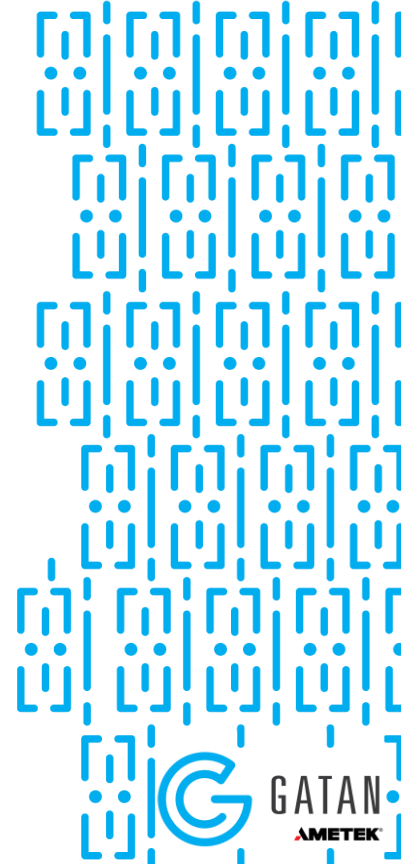


4D-STEM Unveiled: Capturing Diffraction at Every Pixel

Saleh Gorji

Application Specialist TEM - Europe



GATAN
AMETEK

Outline

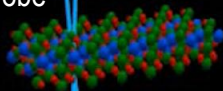
- 4D-STEM and data acquisition approaches
- Applications: Virtual apertures, strain mapping, DPC, and ptychography
- Data quality improvement:
 - Counting and energy-filtered 4D-STEM
- Multimodal 4D-STEM–EELS–EDS
- STEMx OIM orientation mapping and precession



4D-STEM and data acquisition approaches

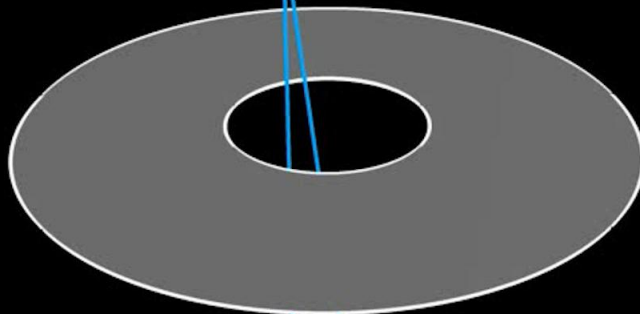
Send STEM detector signal
to the DigiScan™ system

STEM probe



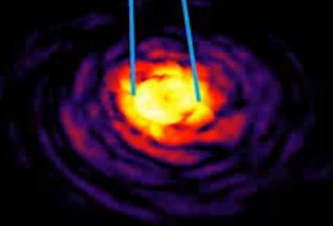
Sample

Annular STEM detector

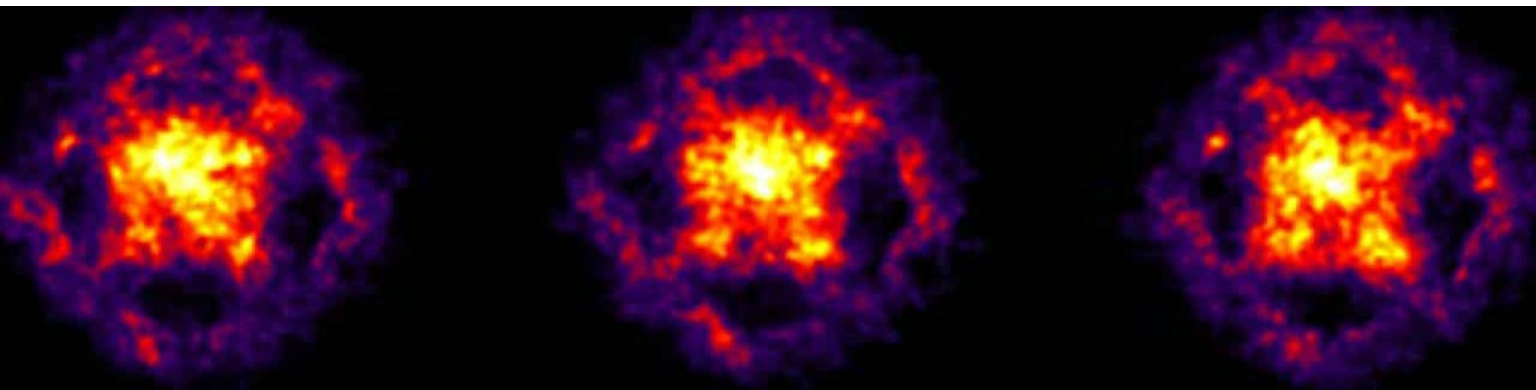


Acquire pixelated diffraction
image from the camera

Gatan camera

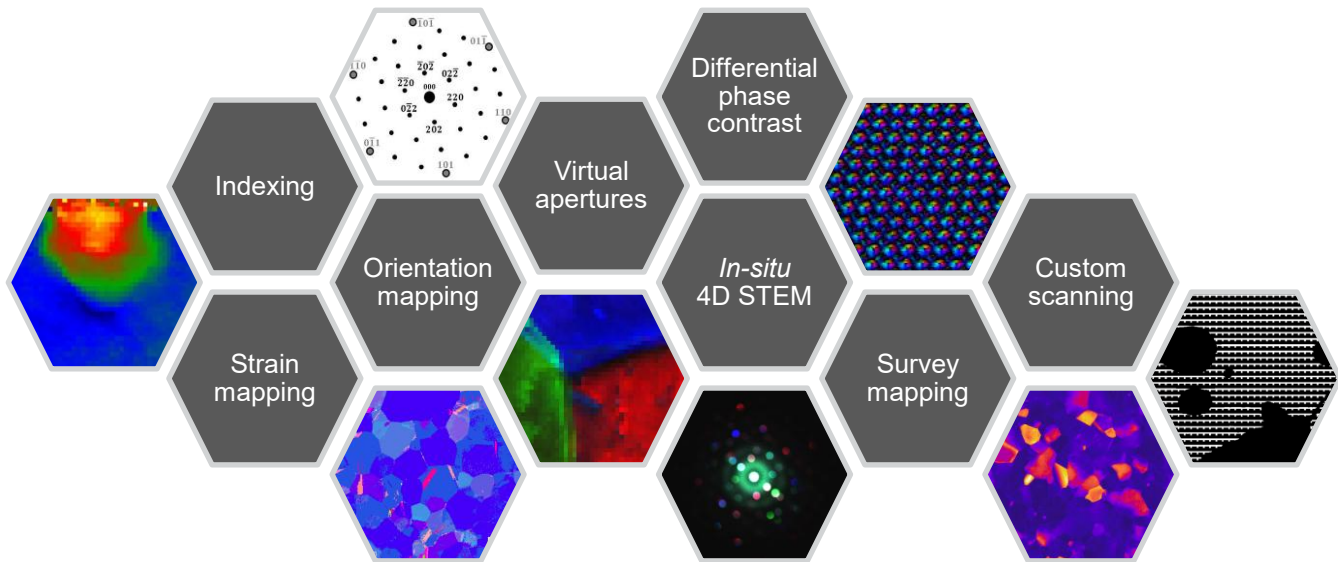


4D STEM operating principle



A 2D Diffraction Pattern for Each Real Space STEM Pixel

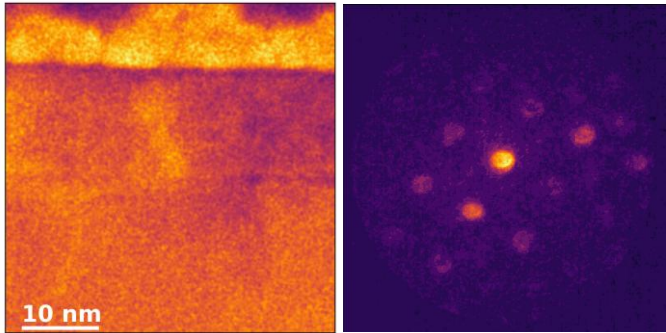
- 4D STEM dataset of a multi-layer specimen
- As you zoom into the real space STEM image you see the 2D diffraction pattern behind each real-space pixel



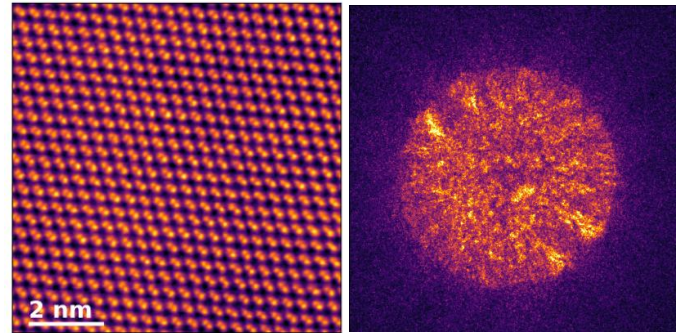
4D-STEM Can Provide a Rich Data

- Broad range of applications
 - Structural, Magnetic, and Electrical Properties

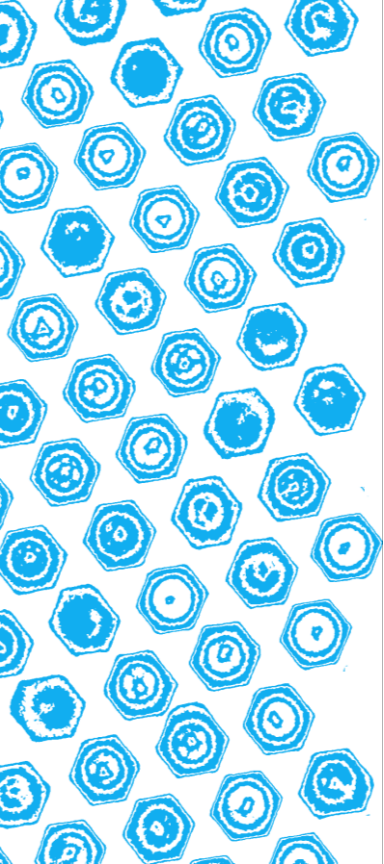
Approaches for 4D STEM experiments



- Low convergence angle, high reciprocal space resolution
- Ideal for structural analysis, strain mapping, orientation mapping

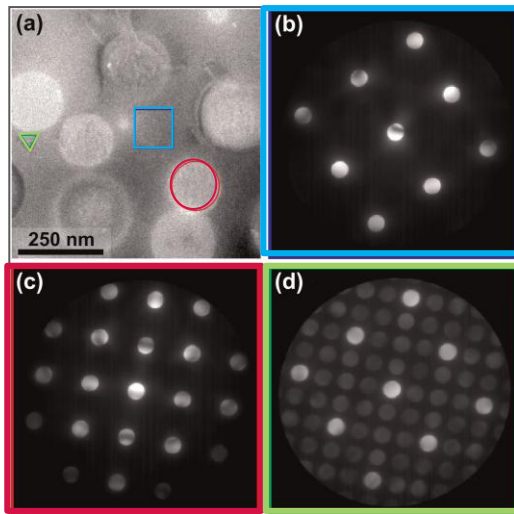


- High convergence angle, high real space resolution
- Ideal for DPC and ptychography



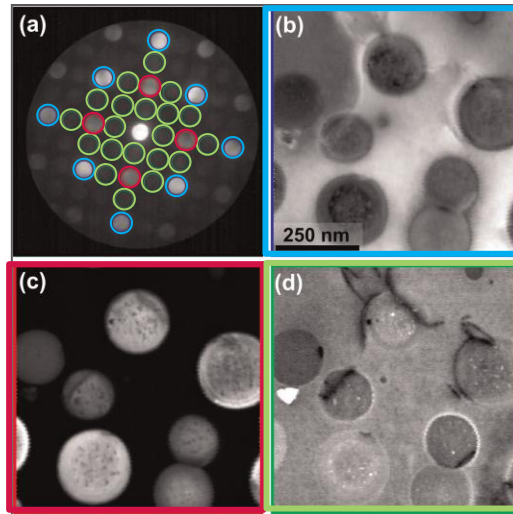
**Applications: Virtual apertures, DPC,
strain mapping, and ptychography**

Virtual SAD apertures



Matrix, Circular precipitate, Smaller precipitate

Virtual objective apertures



Reflection, Superlattice, Fe₂₃Zr₆ structure but not the other two structures

Fe-Al-Ni-Cr alloy sample; Ultramicroscopy 155, 1-10 (2015)

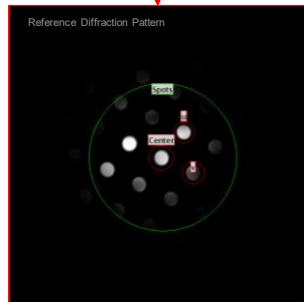
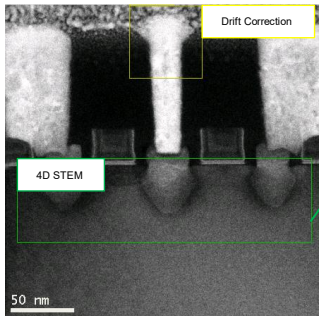
Map real and diffraction spaces with virtual apertures

- Compared to conventional STEM imaging, these images:
 - Less affected by bending and dynamical scattering
 - Provide better signal-to-noise ratio

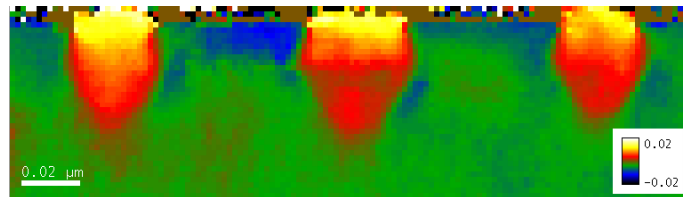
3. Strain Map Calculation

2. Strain Mapping Set-up

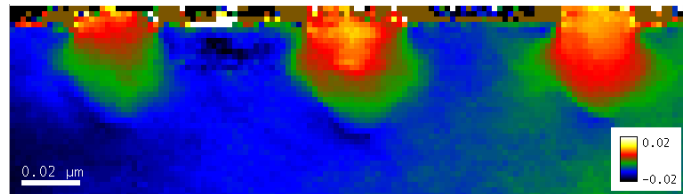
1. Data Acquisition



In-plane Strain

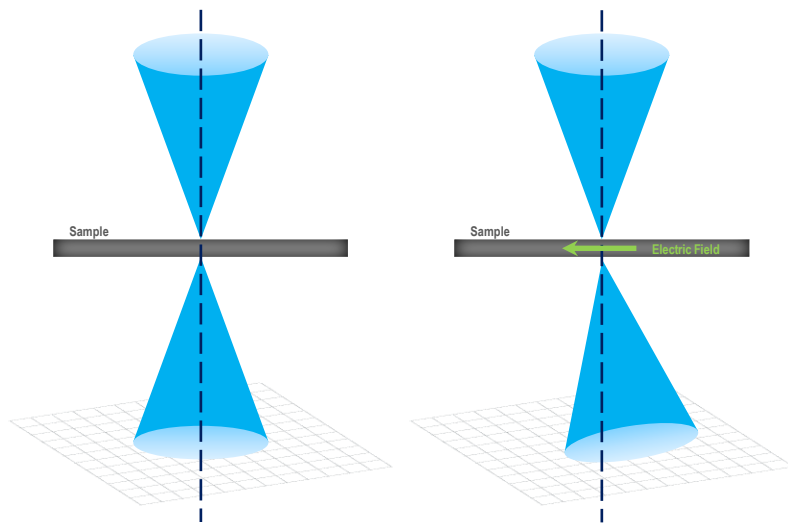


Out-of-plane Strain



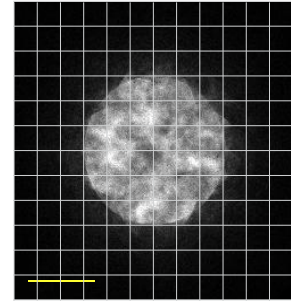
Strain Mapping of Semiconductor Devices in 3 Simple Steps

1. Acquire 4D STEM dataset
2. Set up the strain mapping parameters in DigitalMicrograph:
 - Specify the unstrained region
 - Specify the orientations of interest on the reference diffraction pattern
3. Calculate the strain maps



Beam shift measurement via:

- Center of mass
- Cross correlation
- Virtual segmented detectors



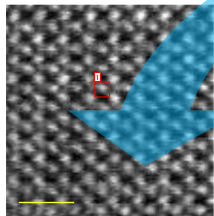
DPC: Electric & magnetic field measurements on a pixelated detector

- In a thin specimen, the beam is deflected proportional to the electric field at that probe position/pixel
- Beam displacement is measured using the center of mass, cross-correlation, or virtual segmented detectors in DigitalMicrograph
- The x and y components resulting from the beam displacement measurement can be summed to give you the beam displacement for electric field and charge density calculations

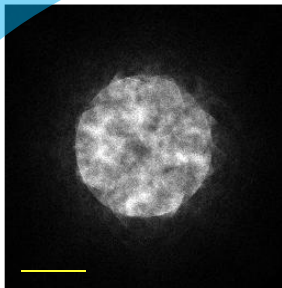
Step 1: Set up & acquire data

Step 2: Perform DPC mapping

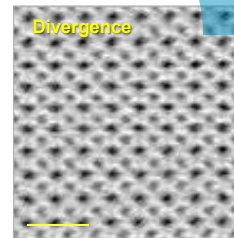
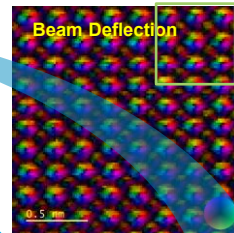
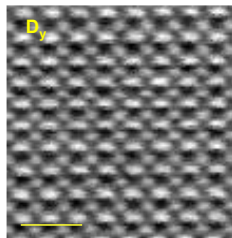
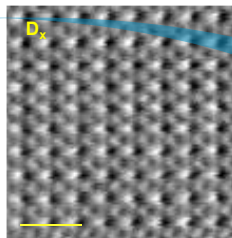
Step 3: Examine results



Acquire 4D STEM Data



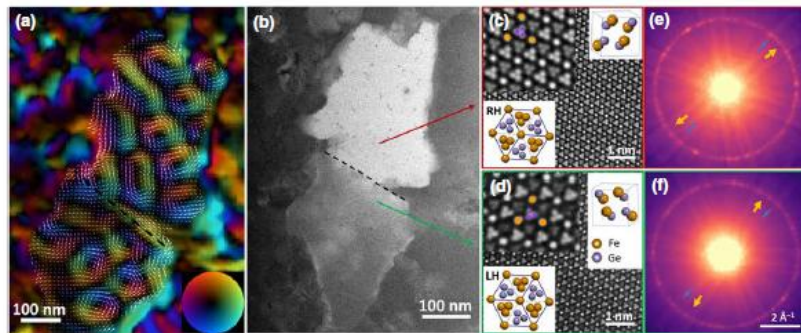
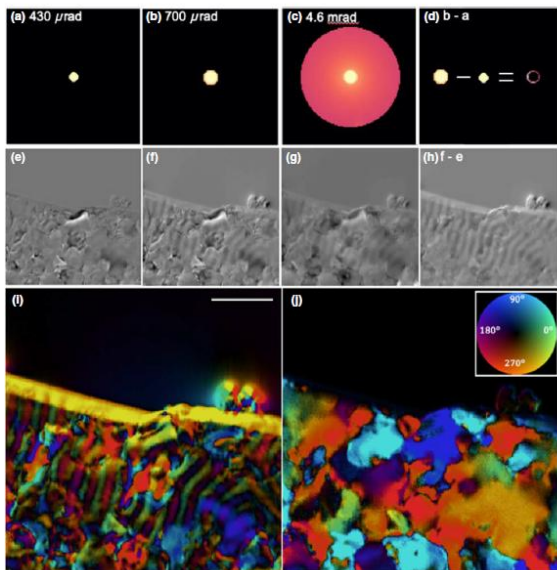
Calculate the COM at each probe position



Differential phase contrast (DPC)

- Atomic-resolution 4D-STEM data acquired from a pyrochlore oxide, with a pixel size of 0.2 Å
- Center-of-mass (COM) analysis or virtual segmens can be used to calculate and output vector maps
- Fast time to results directly at the microscope

Lorentz STEM (Low-Magnification STEM Mode)



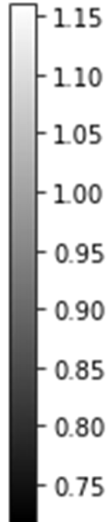
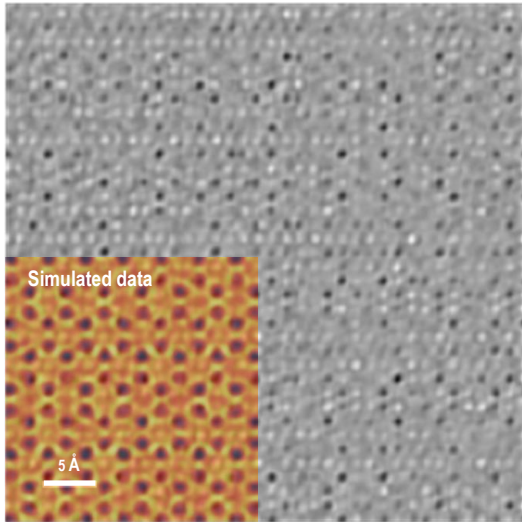
PHYS. REV. APPLIED 17, 034066 (2022)

<http://dx.doi.org/10.1103/PhysRevApplied.17.034066>

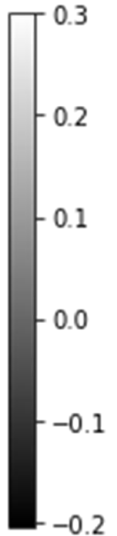
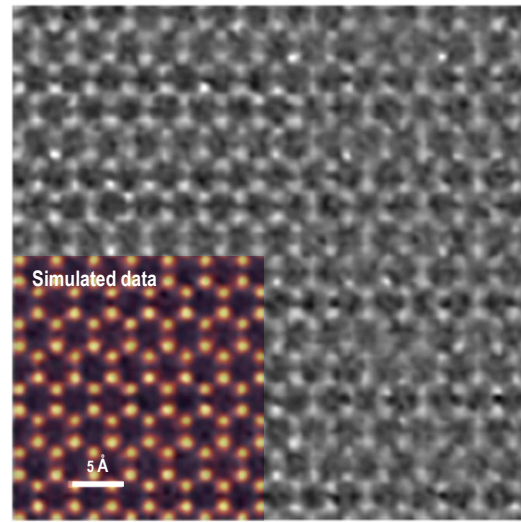
- Helicity of skyrmions coupled with crystal chirality in SiGe
- The top and bottom grains have different chirality in the SiGe crystal structure
- This is correlated with the helicity of the magnetic field in the skyrmion.

- SiGe Polycrystalline film
- Used virtual aperture to decouple contrast changes due to diffraction and the changing magnetic field.

Amplitude



Phase



Courtesy of Roberto dos Reis, Northwestern University

Incorporate Electron Ptychography in GMS with Python

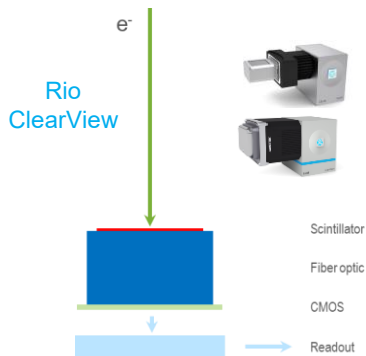
- Electron ptychography for thin specimens has shown imaging resolution beyond the diffraction limit of the lenses
- For 2D materials such low kV (<80) is required in many cases to prevent electron beam damage
- Stela enables high-resolution phase imaging of 2D materials such as MoS₂ at low kV



Data quality improvement: Counting and energy-filtered 4D-STEM

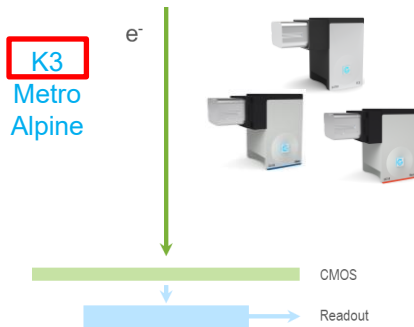
Detection technology: Indirect vs. direct detection counting

Fiber optic-coupled CMOS



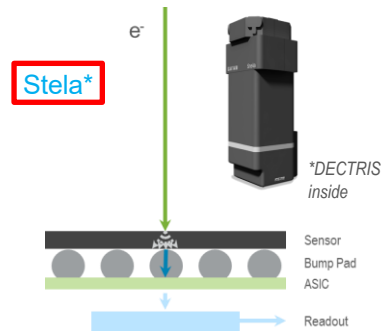
- High quality
- High speed
- More pixels

Monolithic



- High SNR/DQE
- High speed
- More pixels

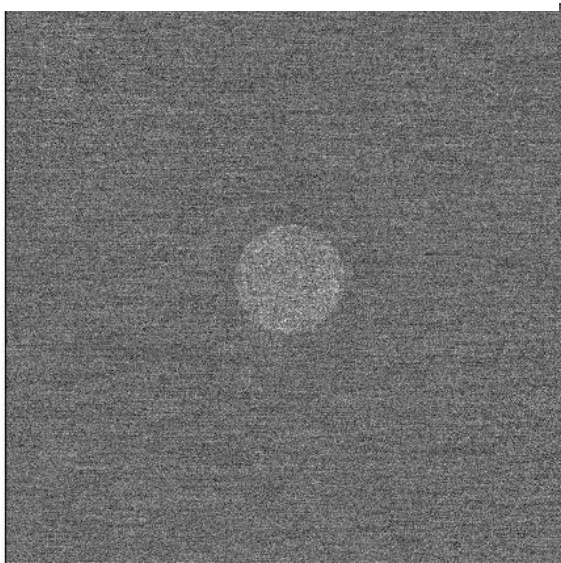
Hybrid-pixel



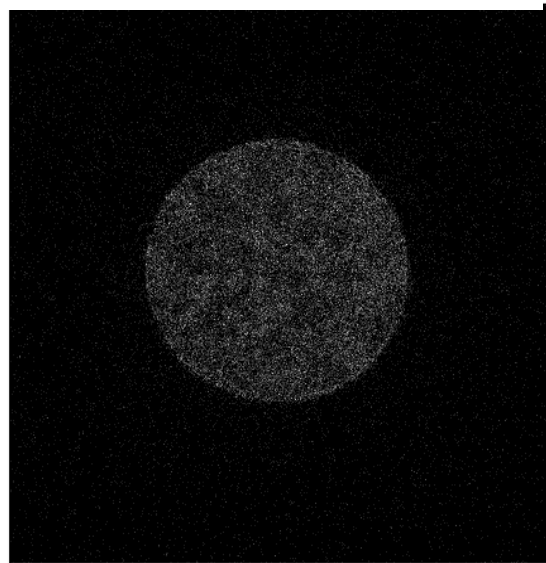
- High SNR/DQE
- High speed
- High dynamic range

Only limited by the **Shot Noise**

Single diffraction with OneView



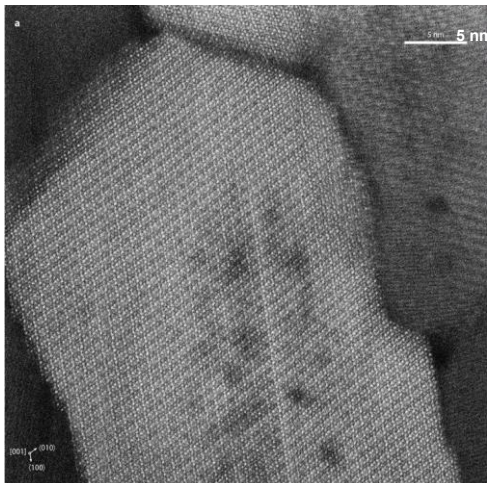
Single diffraction with K3 IS



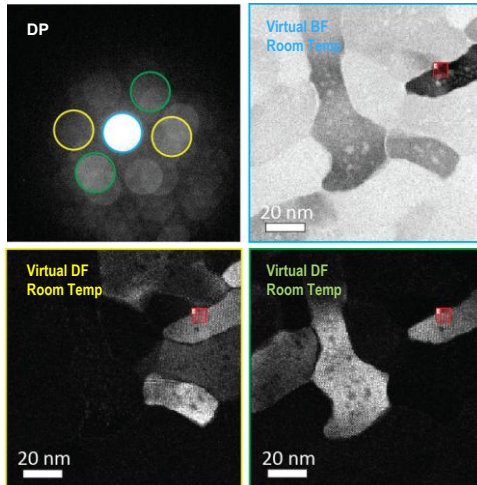
Courtesy of Roberto dos Reis, Northwestern University

Fully integrated direct detection counted 4D STEM

- Higher DQE allows collecting high-quality 4D STEM datasets at lower dose rates
- MOF and COF 4D STEM experiments were not possible with the OneView[®] camera (dose limited) and can be done on the K3 IS camera



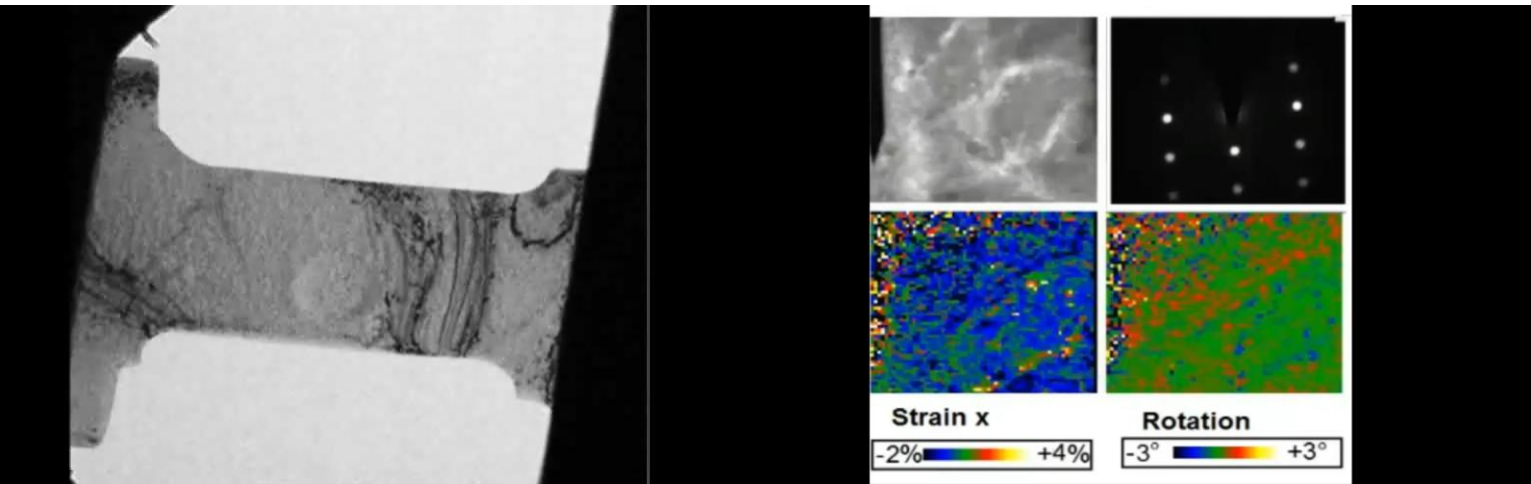
Aberration-corrected cryo-STEM-ADF lattice image
K. A. DeRocher, P. J. M. Smeets et al. *Nature* (2020)



4D STEM data courtesy of doctors Smeets, dos Reis, and Dravid, NUANCE, Northwestern University

K3 IS counting enables 4D STEM of beam-sensitive materials

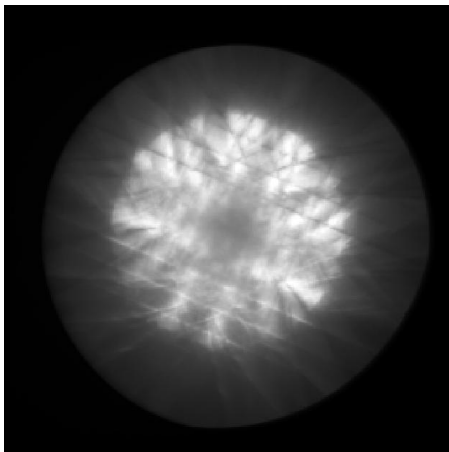
- Enamel is beam-sensitive – Only until now small cryo-HR STEM on small areas were possible
- With K3[®] IS, capture 4D STEM diffraction data at very low dose conditions
- Allows observation of a larger area (~16x) at room temperature without damaging the integrity of the specimen



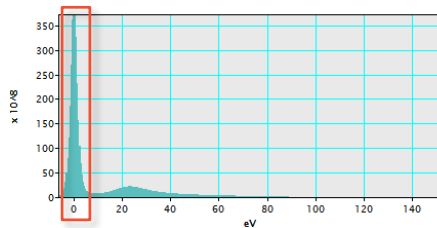
Appl. Phys. Lett. 109, 081906 (2016)

Nanoscale strain mapping during *in-situ* deformation

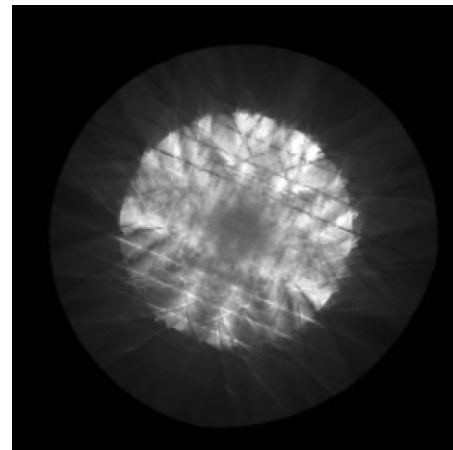
- High-speed cameras from Gatan allow the addition of the fifth dimension of time to 4D STEM experiments
- Local stress/strain during an *in-situ* deformation of a metallic sample was measured with a few nanometers resolution using the K2[®] IS camera and STEMx system



Unfiltered



10 eV slit inserted



Zero-loss filtered

Only fully integrated energy filtered 4D STEM solution

- Allow precise diffraction analysis
- Uncover hidden information
- Provide additional acquisition modalities via energy filter (EELS, EFTEM, EELS-SI)

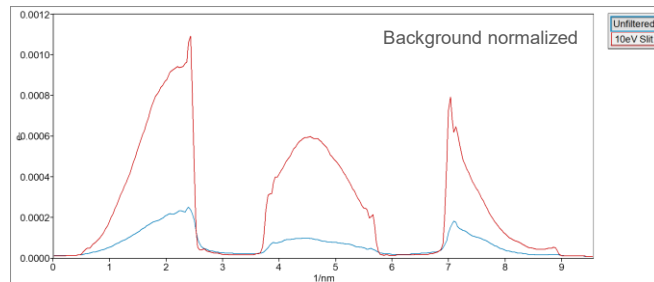
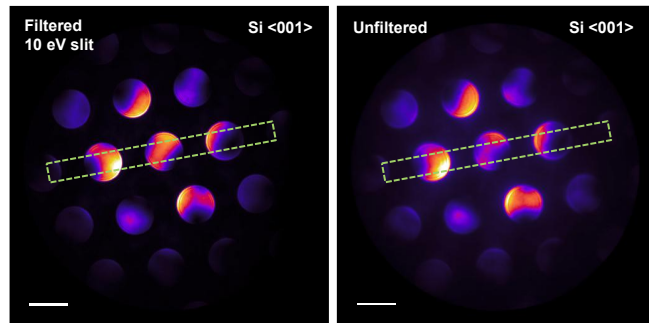
Energy-filtered strain mapping

Strain mapping in single crystal

- The strain mapping algorithm performs strain analysis by comparing CBED disc positions in a 4D STEM dataset with those in an unstrained reference pattern
- Energy-filtering improves disc sharpness and edge detection, significantly improving the accuracy of strain mapping measurements
- Single crystal silicon was used as an ideal test case to quantify the benefit of energy-filtering; a 20% improvement in precision was observed

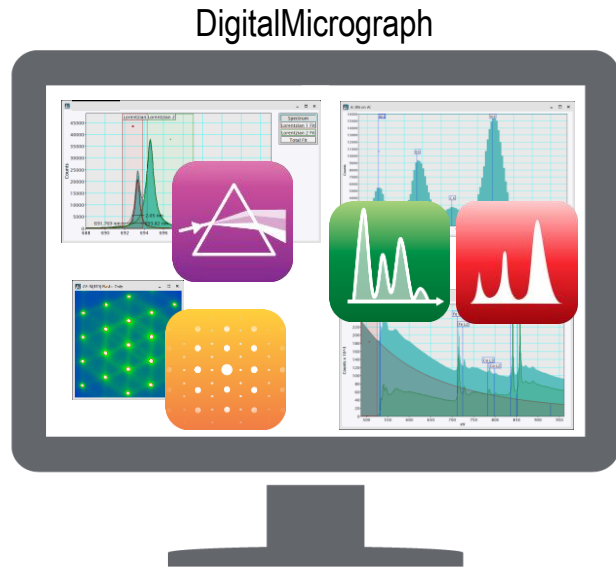
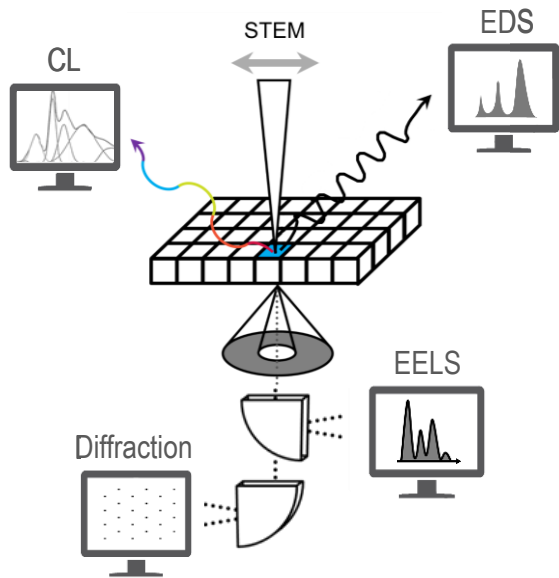
| Mean | No filter | Filtered |
|-----------------|-------------------|-------------------|
| ϵ_{uu} | 0.008 ± 0.016 | 0.021 ± 0.014 |
| ϵ_{vv} | 0.000 ± 0.016 | 0.019 ± 0.013 |
| ϵ_{uv} | 0.002 ± 0.010 | 0.000 ± 0.008 |
| Ratio n | 0.073 ± 0.630 | 0.022 ± 0.63 |

20% improvement



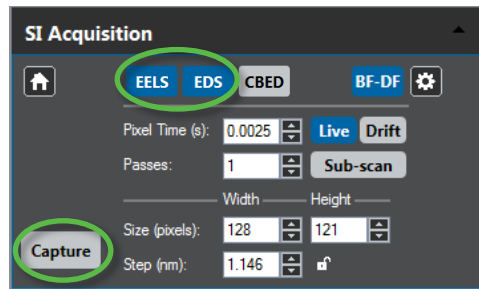
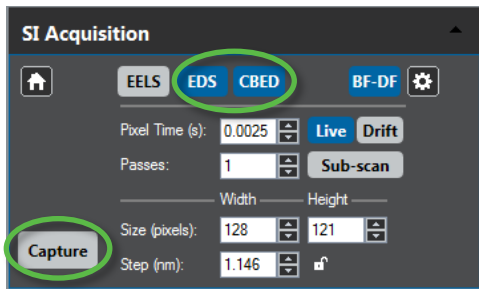


Multimodal 4D-STEM-EELS-EDS



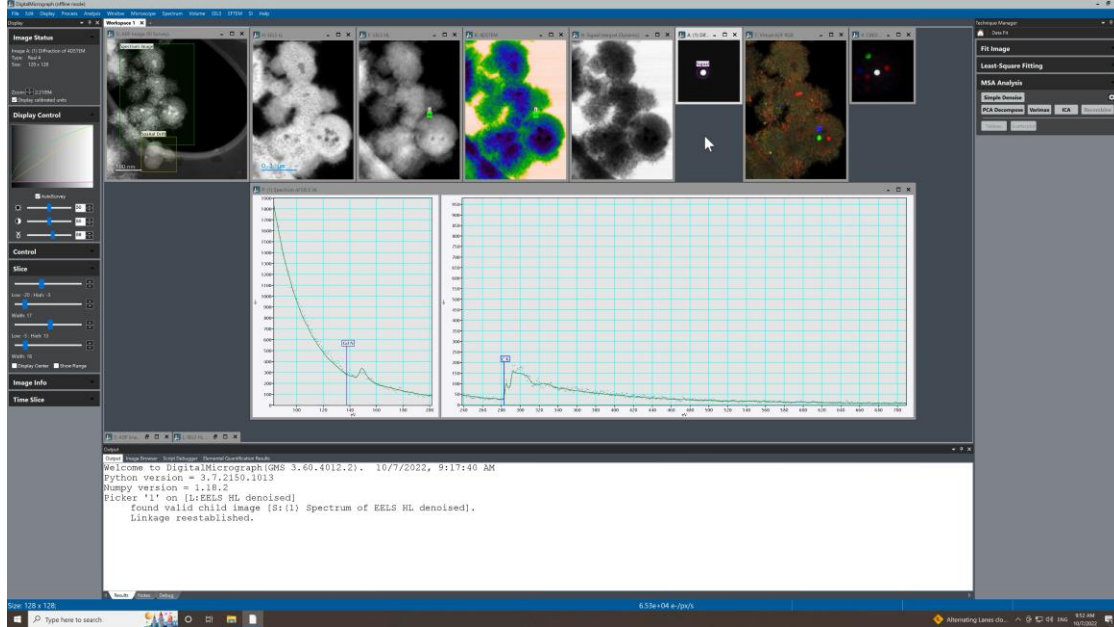
eaSI collects complementary signals from multiple detectors

- Analyzing a specimen with a single STEM technique is not always sufficient to fully understand the system and explain/predict the material properties and behavior
- eaSI **combines** the broadest range of STEM-optimized EELS, EDS, CL, and 4D STEM detectors in DigitalMicrograph software



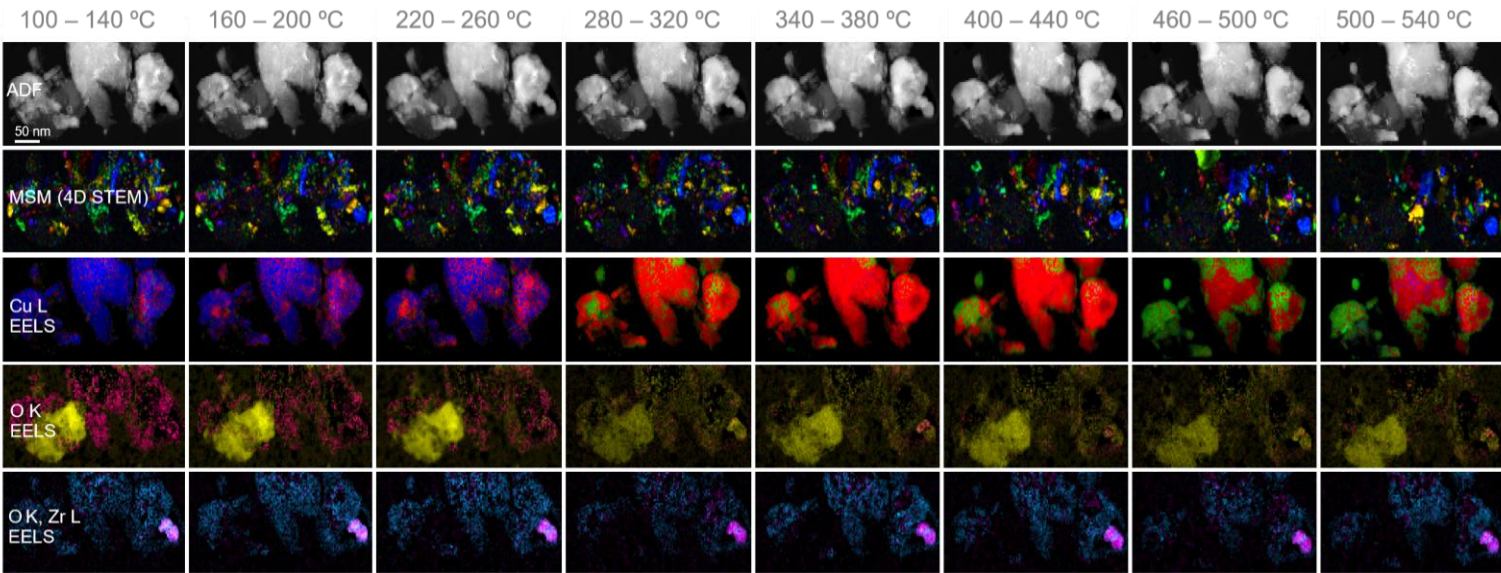
Enables and simplifies synchronization of compatible STEM detectors

- Hardware **synchronization** is required for the highest possible acquisition rates of multimodal STEM SI datasets without compromising data integrity
- Select your desired STEM signals from the STEM SI panel and start the acquisition; eaSI **synchronizes** for you!



Complements your existing techniques – 4D STEM + EELS

- One sample, one microscope session, one data format – unparalleled productivity
- 4D STEM analysis identifies distinct crystallites, and linked processing tools allow EELS data to be extracted from the exact same position

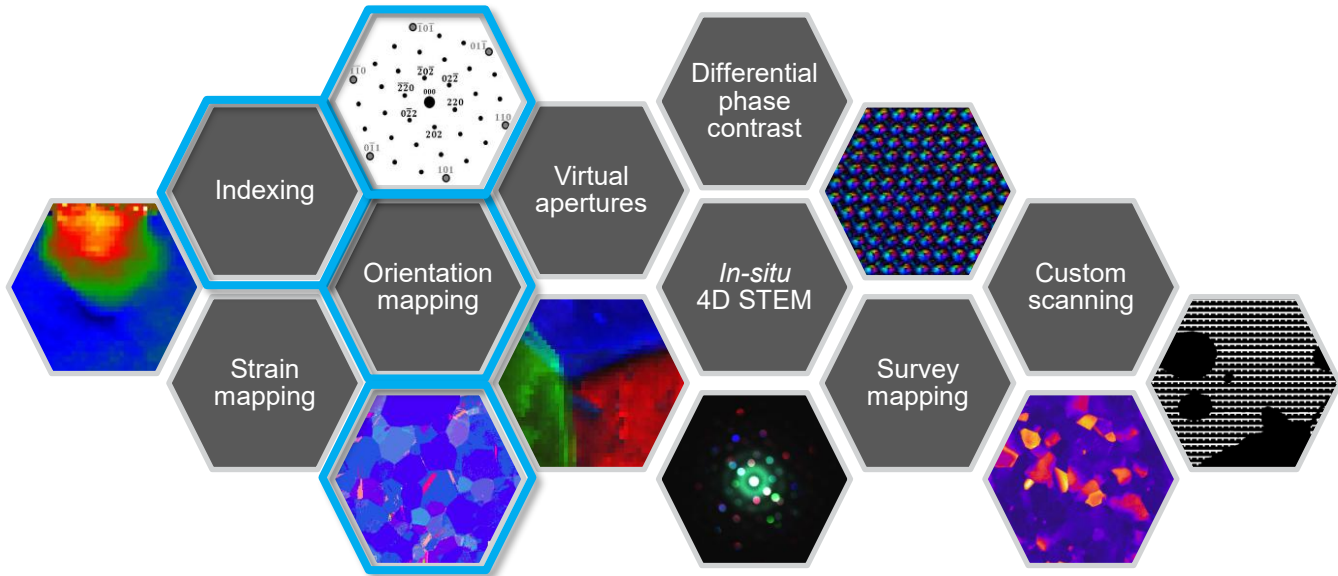


eaSI: STEM experiment combined, synced, and linked

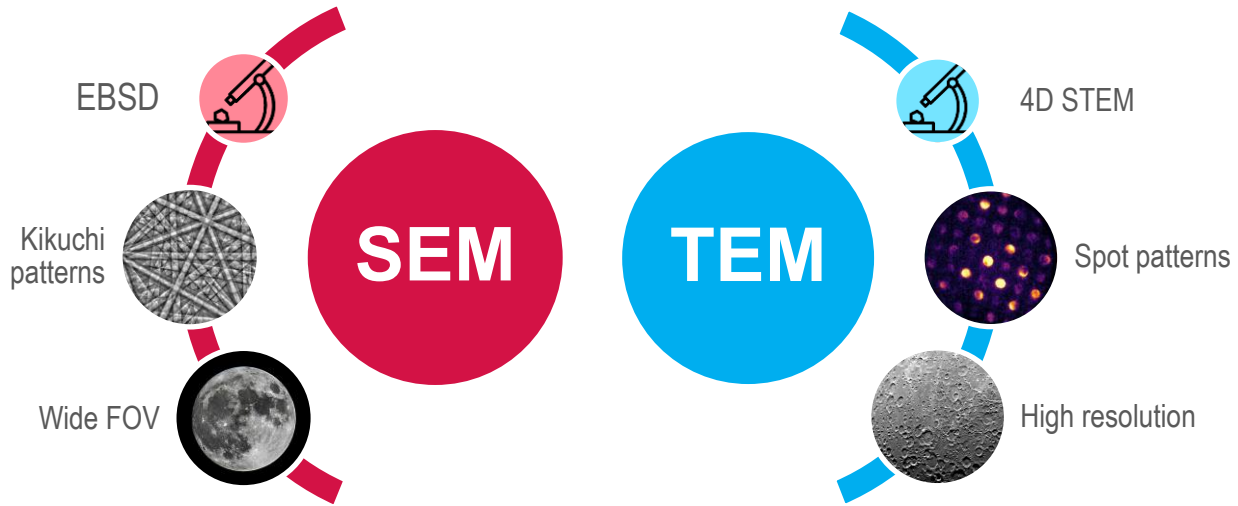
- *In-situ* EELS + 4D STEM experiment entirely collected in DigitalMicrograph software
- Confidently process and analyze acquired linked data in multiple ways, while maintaining spatial and temporal correlation



STEMx OIM orientation mapping and precession

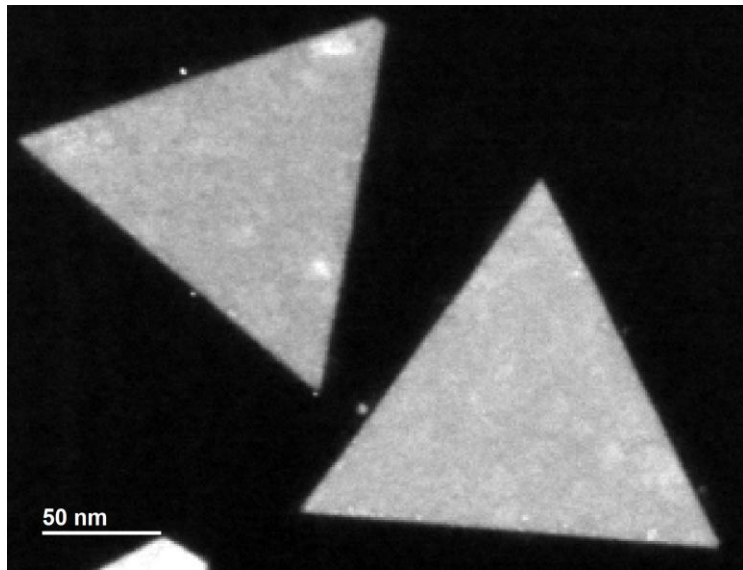


Indexing and Orientation mapping

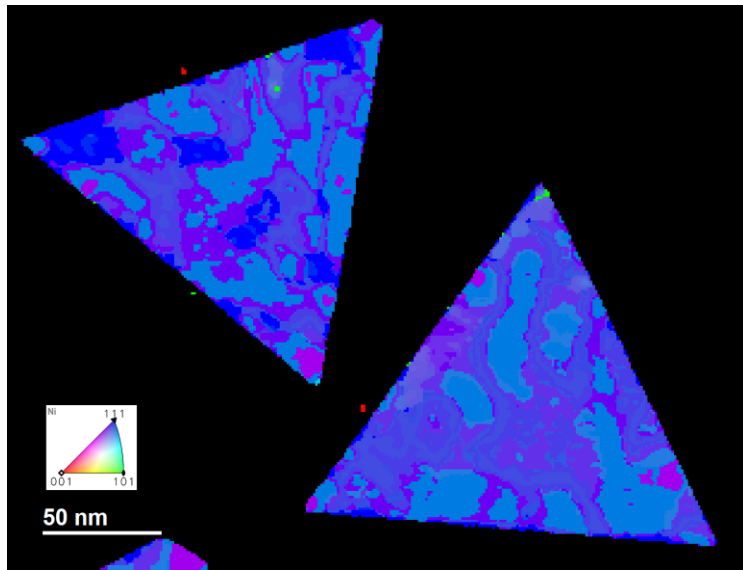


Orientation mapping in electron microscopy

- Grain orientation and phase distribution analysis is needed for understanding materials properties like strength or conductivity
- EBSD is a common approach, but nanoscale materials (2D materials, semiconductors, nanograined metals and alloys) require spatial resolutions not achievable in the SEM
- Data processing and analysis software must be straightforward to use and ideally give correlative results



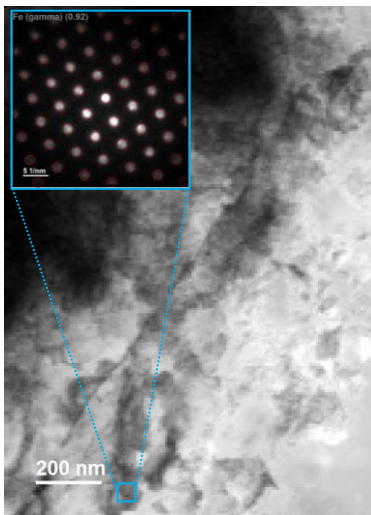
STEM ADF image



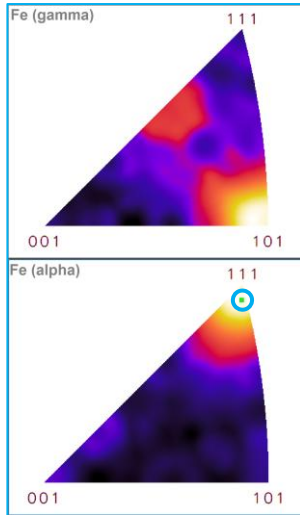
STEMx OIM orientation map

Add orientation mapping to your 4D STEM analysis toolkit

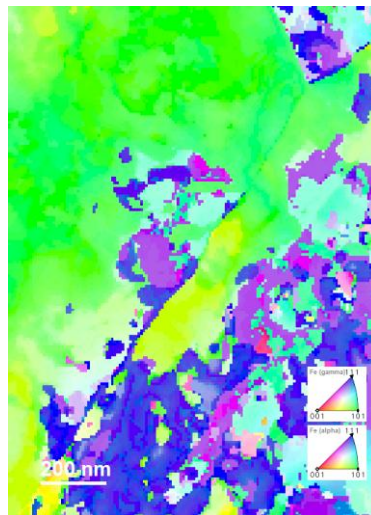
- STEM analysis enables higher-resolution data than possible via SEM
- Easily generate orientation maps in DigitalMicrograph with a template-matching algorithm requiring no scripting at all



Virtual bright field of 304 stainless



Multiphase heatmap



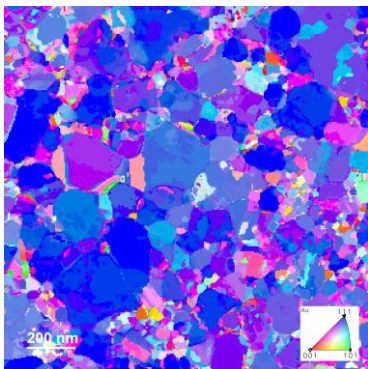
Orientation map



Fe alpha/gamma phase map

Analyze multiphase materials with STEMx OIM

- Analysis setup for multiphase orientation and phase mapping is just as easy as with single phase materials
- At each pixel position, STEMx OIM assigns the best match from all possible phases to generate the phase and orientation maps



Orientation map of Au film

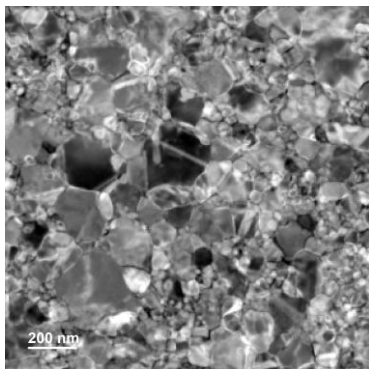
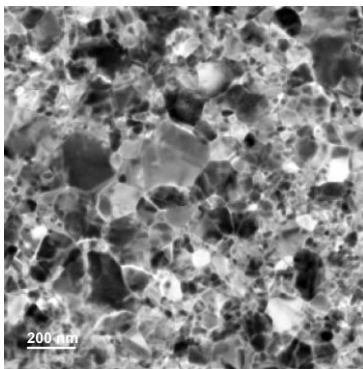
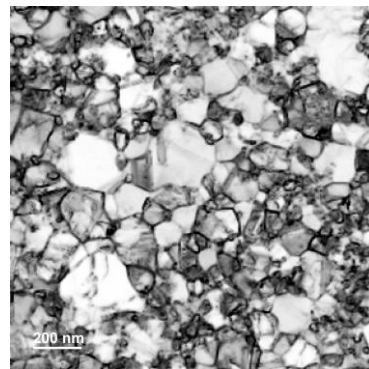


Image quality map



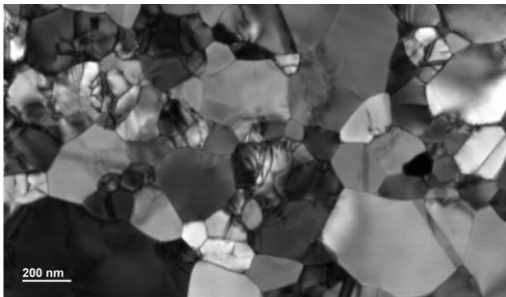
Confidence index map



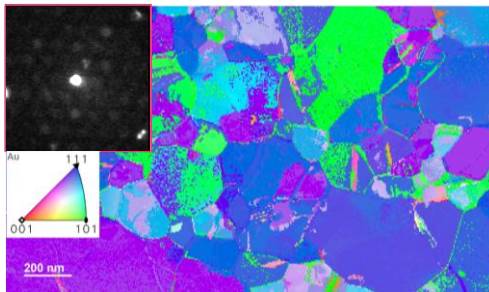
Similarity map

Assess orientation analysis results with correlative mapping options

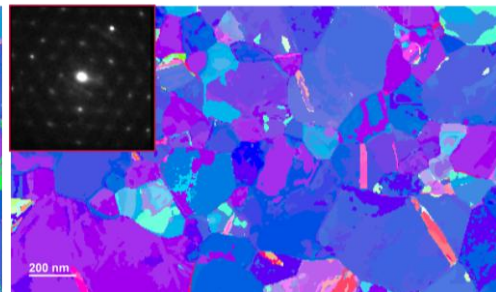
- STEMx OIM provides Image Quality and Confidence Index metrics at each pixel position for additional analysis detail and context
- Similarity maps highlight regions with (dis)similar diffraction patterns between neighboring pixels for even more insight into grain structure



Au film



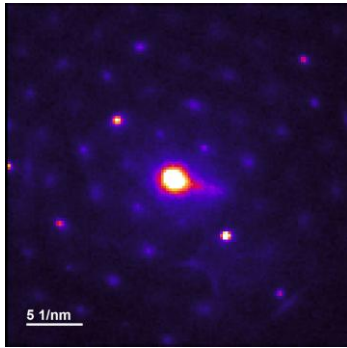
2.5 mrad – most spots have low SNR



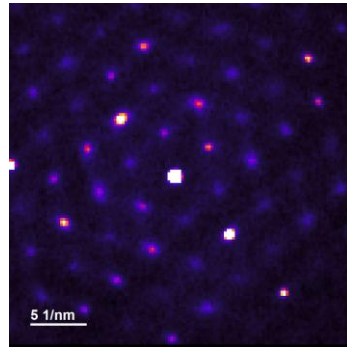
0.7 mrad – spot SNR increases and indexing improves

Smaller convergence angles improve indexing results

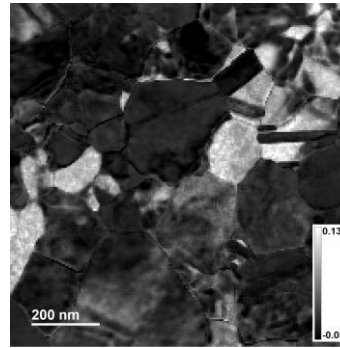
- Smaller 0.7 mrad convergence angle (JEOL STEM $\mu\mu$ DIFF mode) improves indexing significantly compared to 2.5 mrad (JEOL STEM MAG mode)
- Smaller spot size improves signal-to-noise at same dose and pixel dwell time and provides more gaussian spot profile for the template-matching algorithm



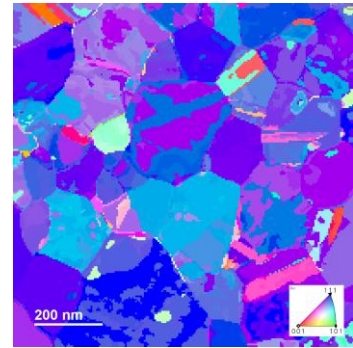
100 nm thick Au



10 eV energy filter reduces background, improves SNR



Brighter grains mark improvements in confidence index from energy filtering

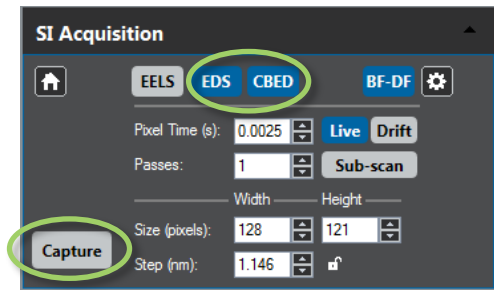


Orientation map

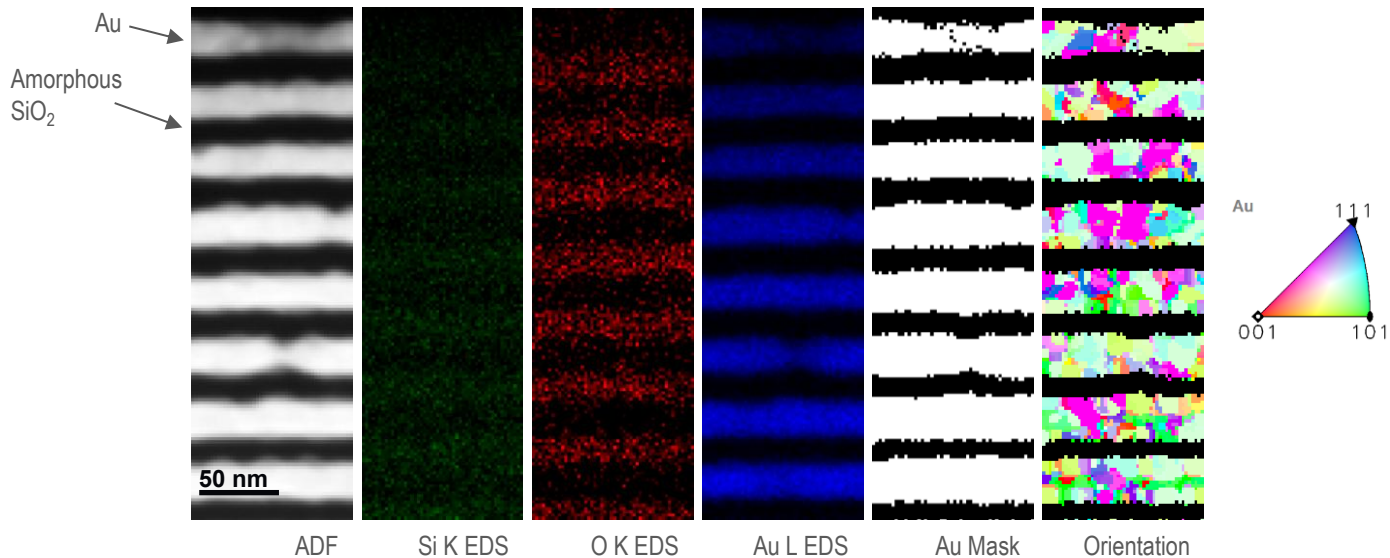
Energy filtering has positive effect on orientation map confidence

- Energy filtering reduces background signal in diffraction pattern and improves SNR of spots across pattern area
- Confidence index values in several grains of map area increase with the energy filtering
- Energy filtering should have even larger effect in thicker samples, or samples with less symmetry

eaSI: STEM experiments combined, linked, and synced

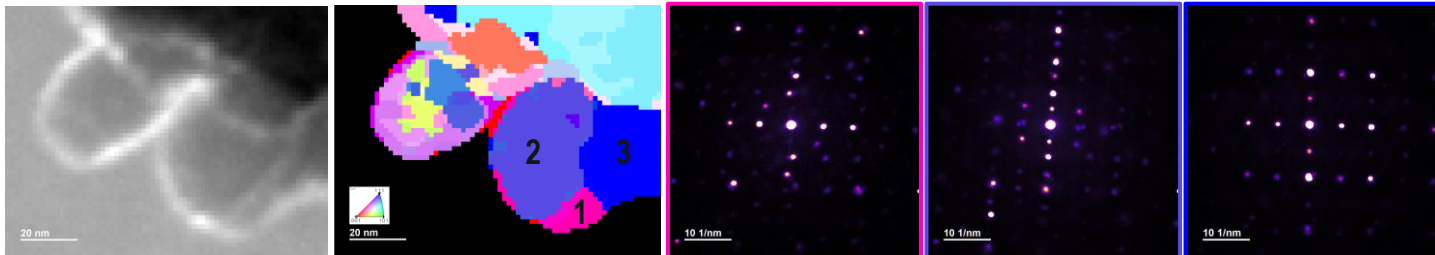


- Simply select your desired STEM signals from the STEM SI panel and start the acquisition
- eaSI enables simultaneous 4D STEM + EDS and sequential 4D STEM + EELS data acquisition
- 4D STEM orientation/phase maps along with linked EDS/EELS maps give a complete picture of structure and elemental/chemical properties of your material



Combining 4D-STEM orientation analysis with simultaneous EDS

- eaSI technology enables simultaneous EDS and 4D STEM data acquisition in DigitalMicrograph
- Linked EDS and 4D STEM data enables correlative elemental and structural analysis, and orientation maps can be masked based on elemental mapping data



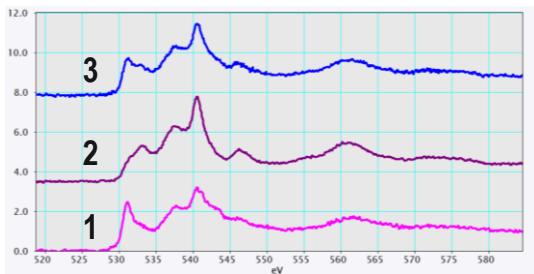
CoO virtual BF

CoO Orientation map

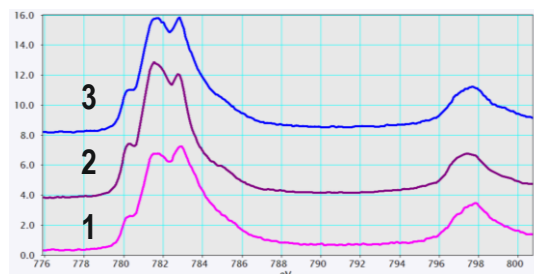
Diffraction area 1

Diffraction area 2

Diffraction area 3



O-K EELS



Co-L_{2,3} EELS

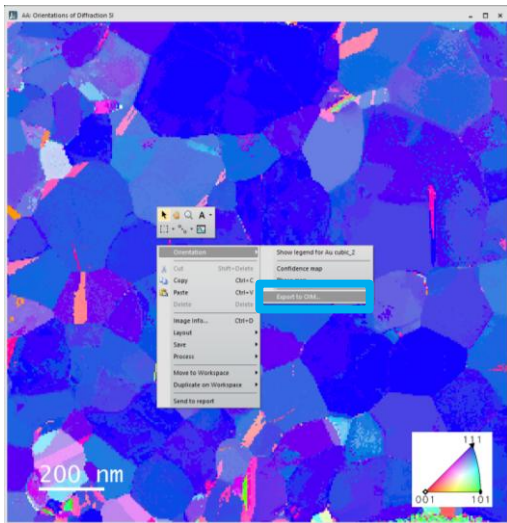
Link structure and chemistry with eaSI EELS + 4D STEM

- eaSI technology seamlessly links sequentially-acquired EELS and 4D STEM datasets to combine orientation analysis with EELS chemical analysis
- Linked data provides deeper sample analysis: orientation data reveals smaller grains with distinct EELS fine structure not expected from virtual BF image contrast

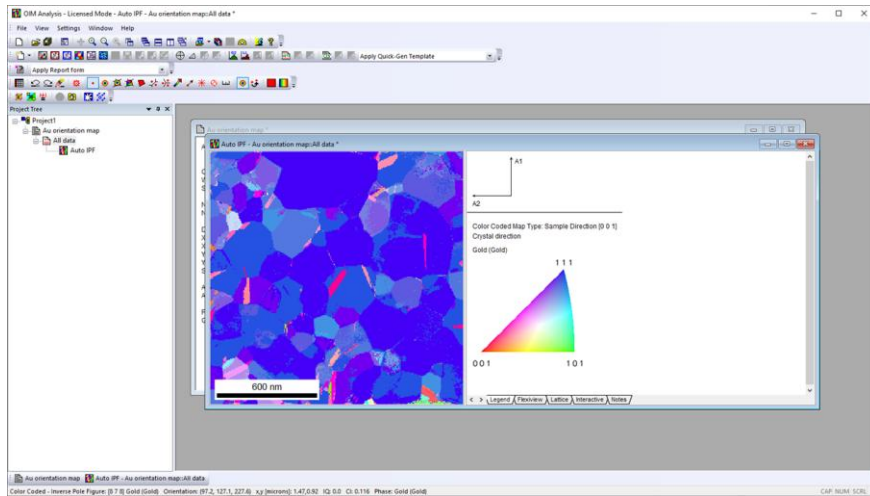


eaSI





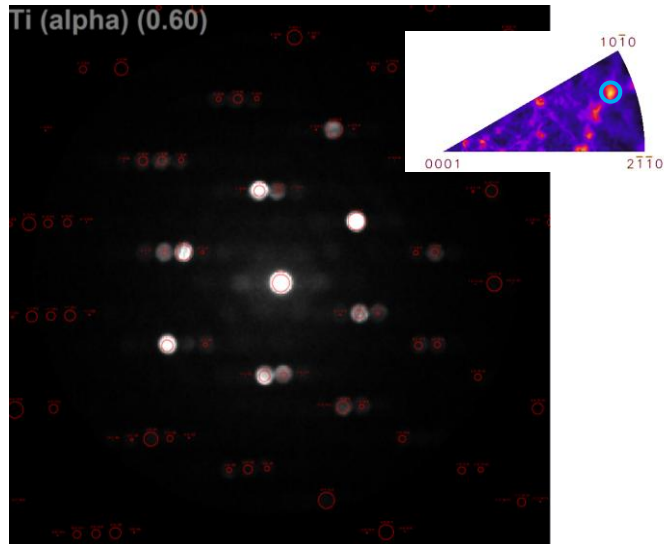
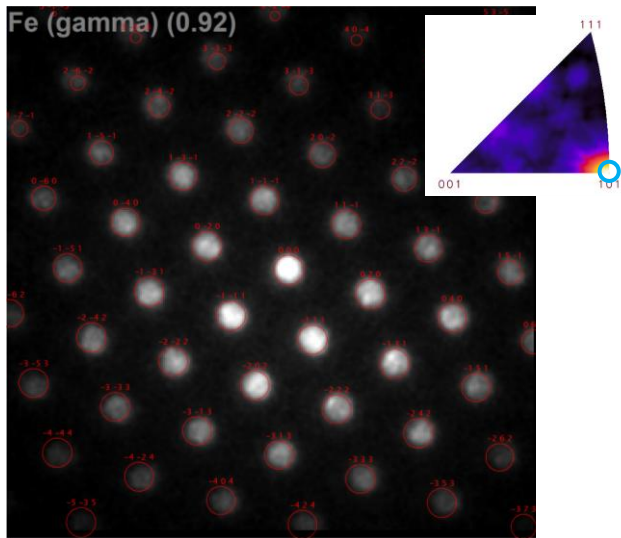
DigitalMicrograph



OIM Analysis

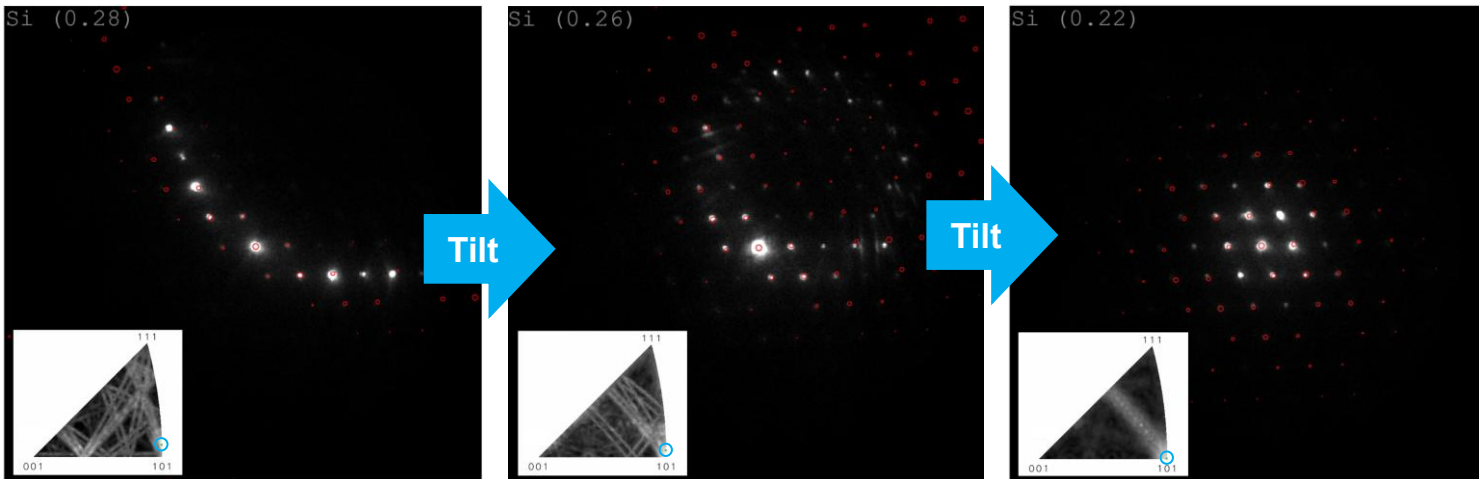
Bring 4D STEM data into EBSD analysis workflows

- Orientation map data is easily exported to *.ang format for more processing options in the EDAX OIM Analysis software
- SEM and TEM users can analyze EBSD and 4D STEM orientation data in the same way



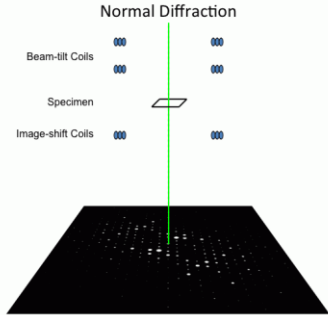
Index electron diffraction patterns from any diffraction experiment

- STEMx OIM indexing works on diffraction patterns from 4D STEM datasets as well as parallel-beam selected area electron diffraction (SAED) patterns
- Utilize the solution heatmap to visualize all viable indexing solutions and apply automated diffraction spot labeling to facilitate pattern interpretation.

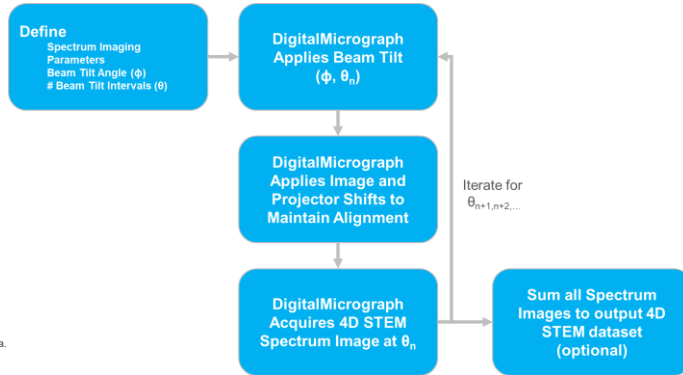


Guided experiment setup with live pattern indexing

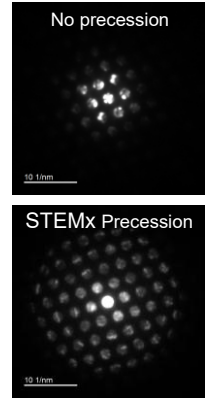
- Set up your diffraction experiment more efficiently and find your desired zone axis quicker using STEMx OIM indexing on your camera live view



Rfree1989. Geometry of electron beam in precession electron diffraction. Wikipedia. https://en.wikipedia.org/wiki/Precession_electron_diffraction. CC BY-SA 4.0.

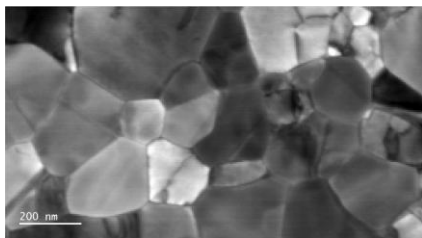
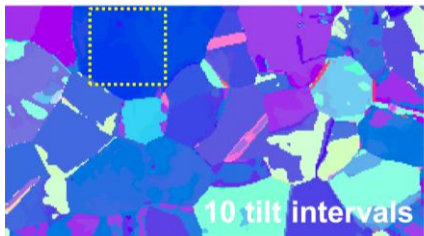
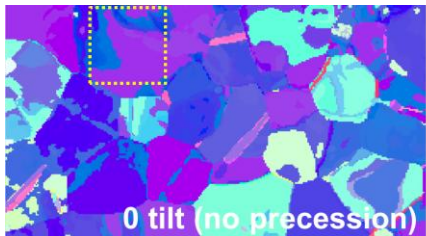


STEMx Precession *under development*



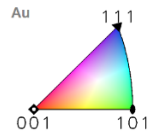
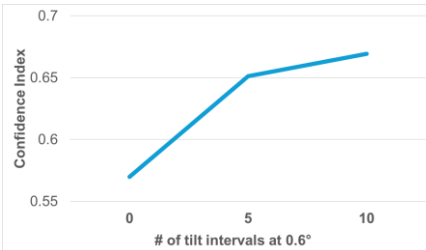
Acquire precession-assisted data with STEMx Precession

- Precession electron diffraction (PED) reduces dynamical effects and captures higher-order diffraction spots, both beneficial to 4D STEM strain and orientation & phase mapping experiments
- STEMx Precession controls STEM coils via DigitalMicrograph to precess the beam and maintain data alignment. This enables precession-assisted 4D STEM data acquisition without the need for additional hardware



Au STEM Image

Au samples courtesy of J. Kacher, GA Tech.

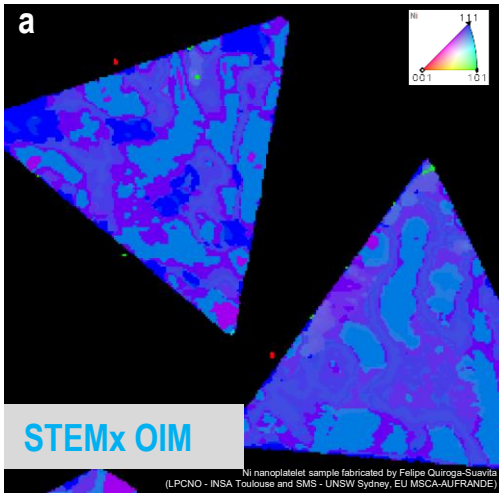


STEMx Precession results in orientation maps with higher confidence index values in highlighted grain

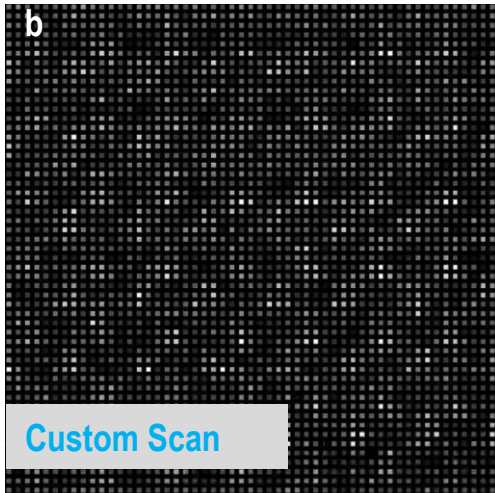
Improve indexing results with STEMx Precession orientation mapping

- STEMx Precession data improves template-matching results, yielding orientation maps with higher confidence index values
- In several grains, confidence index values increased more than 15% compared to non-precessed data, demonstrating the enhanced accuracy and reliability of STEMx Precession orientation analysis

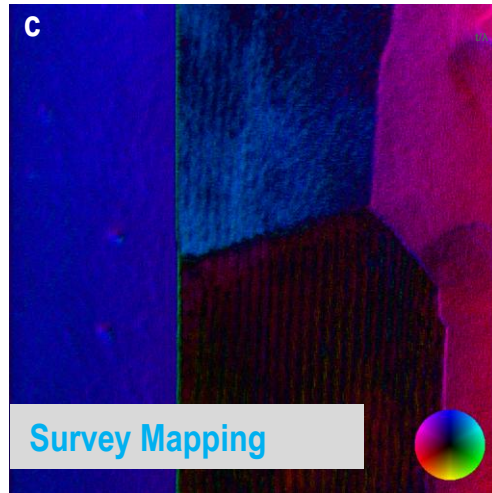
Three new Spectrum Imaging techniques in DM 3.62



4D STEM orientation mapping made eaSI

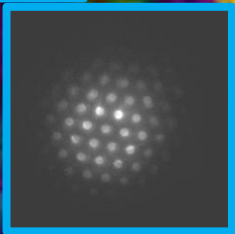


Increasing flexibility to decrease damage



Real time results, real data reduction

Thank You!



Max diffraction spot
orientation and
intensity color scale

This 128 x 128 (scan) x 512 x 512 (pattern) dataset was collected in half a minute, and processed via Python in DigitalMicrograph in 1 min

Material: boron nitride