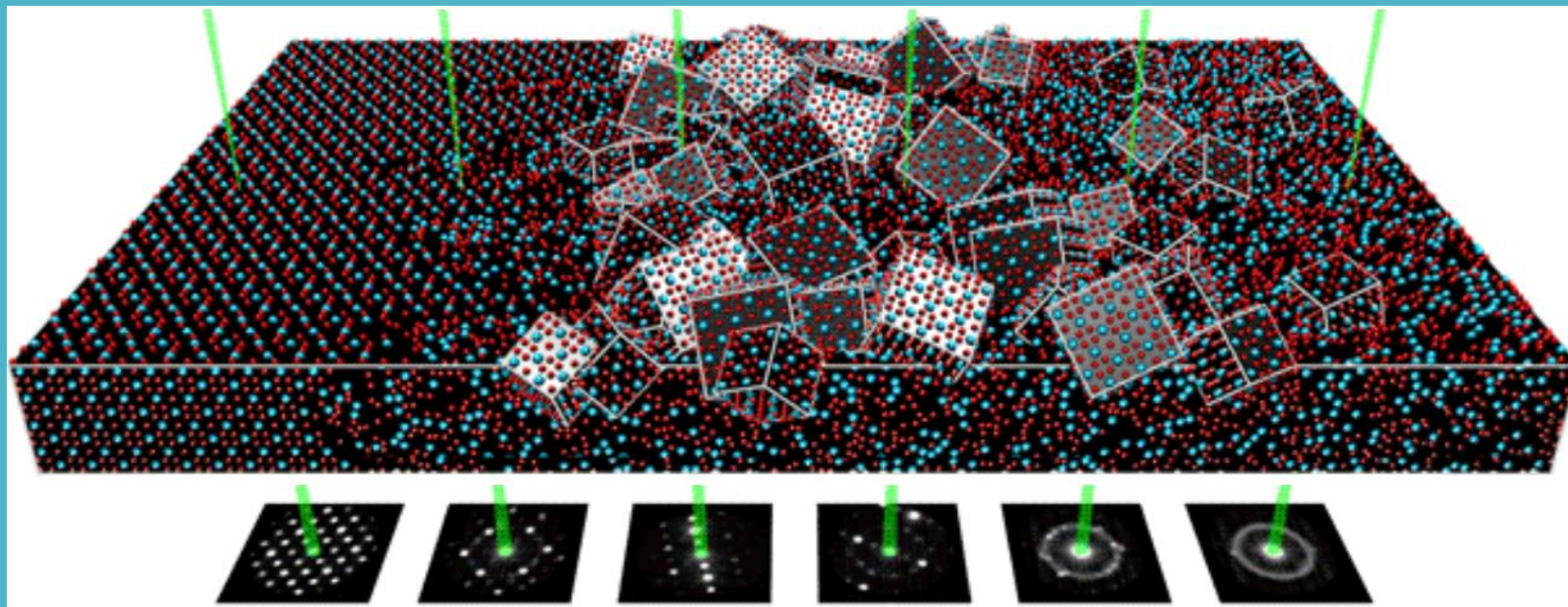
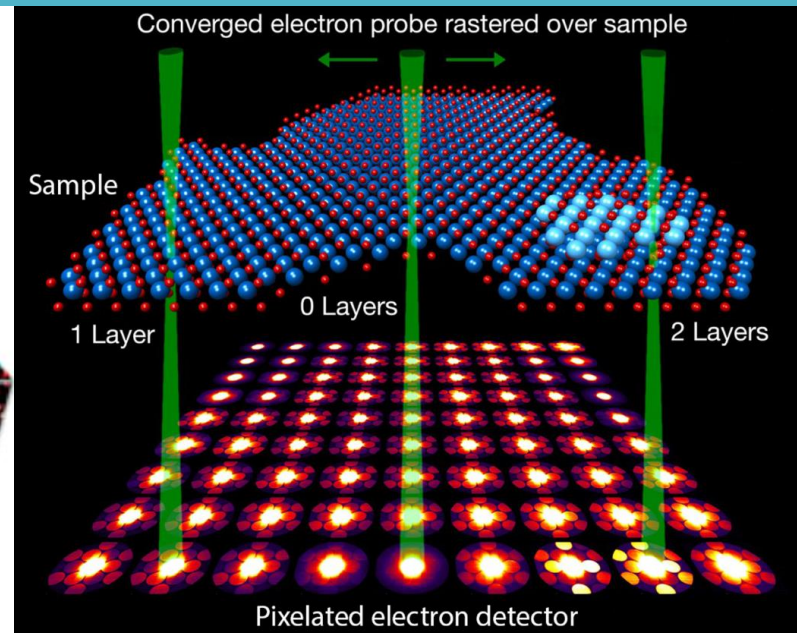


Multidimensional Transmission Electron Microscopy



[Microscopy and Microanalysis (2021), 27, 712–743]



[Microscopy and Microanalysis (2019), 25, 563–582]

25.5.2023

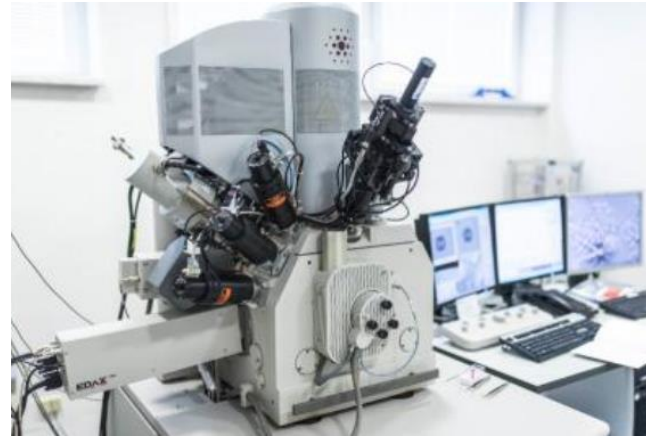
Marek Vronka

Laboratory of electron microscopy (LEM)

RNDr. Mariana Klementová, Ph.D.
Head of Laboratory LEM



- Mgr. Jarmila Balogová
- Ing. Jan Duchoň
- Martin Dušák
- Ing. Miloslav Klinger
- RNDr. Tomáš Kmječ, Ph.D.
- RNDr. Jaromír Kopeček, Ph.D.
- RNDr. Jaroslav Kupčík
- Ing. Jan Maňák
- Ing. Petr Svora, Ph.D.
- Ing. Marek Vronka, Ph.D.



FEI Quanta 3D FEG



Tescan FERA 3



FEI Tecnai TF20 X-Twin



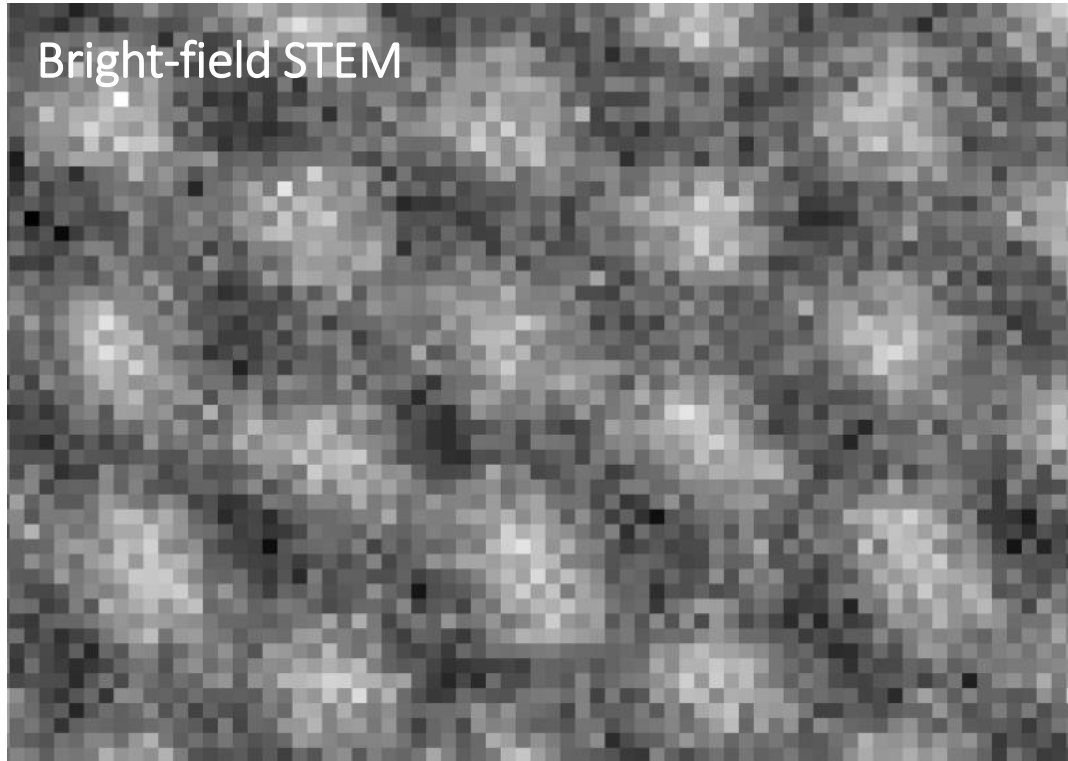
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SCIENCE
LUNCH

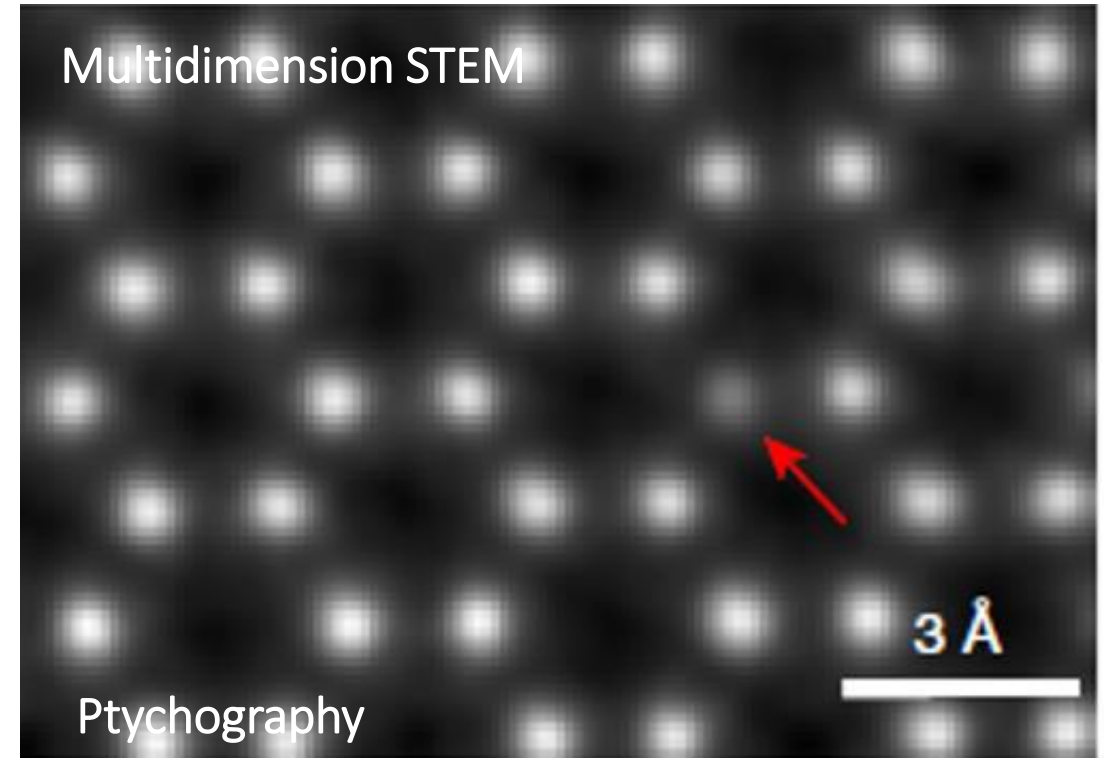
Introduction 1

Resolution 200 pm



monolayer MoS₂ at 80 kV

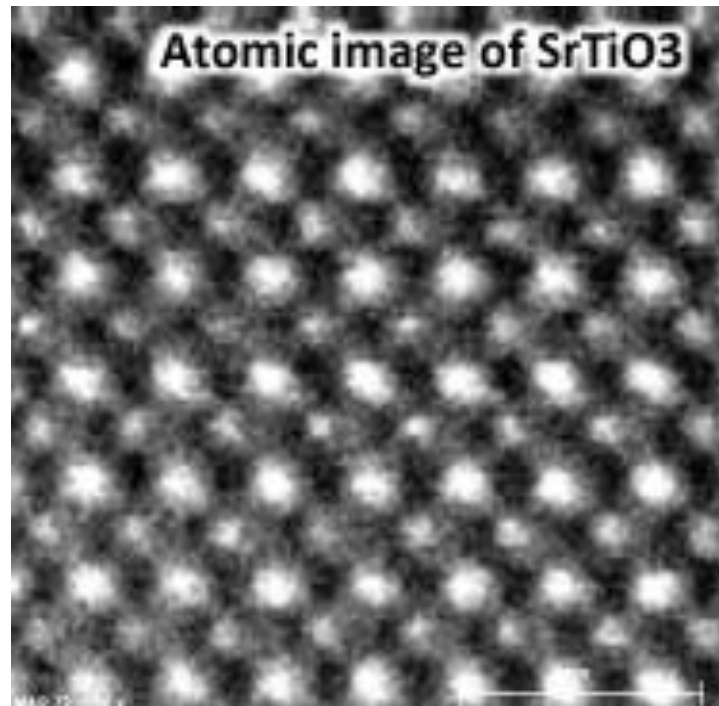
Resolution down to 20 pm



[Nature (2018), 559, 343-349]

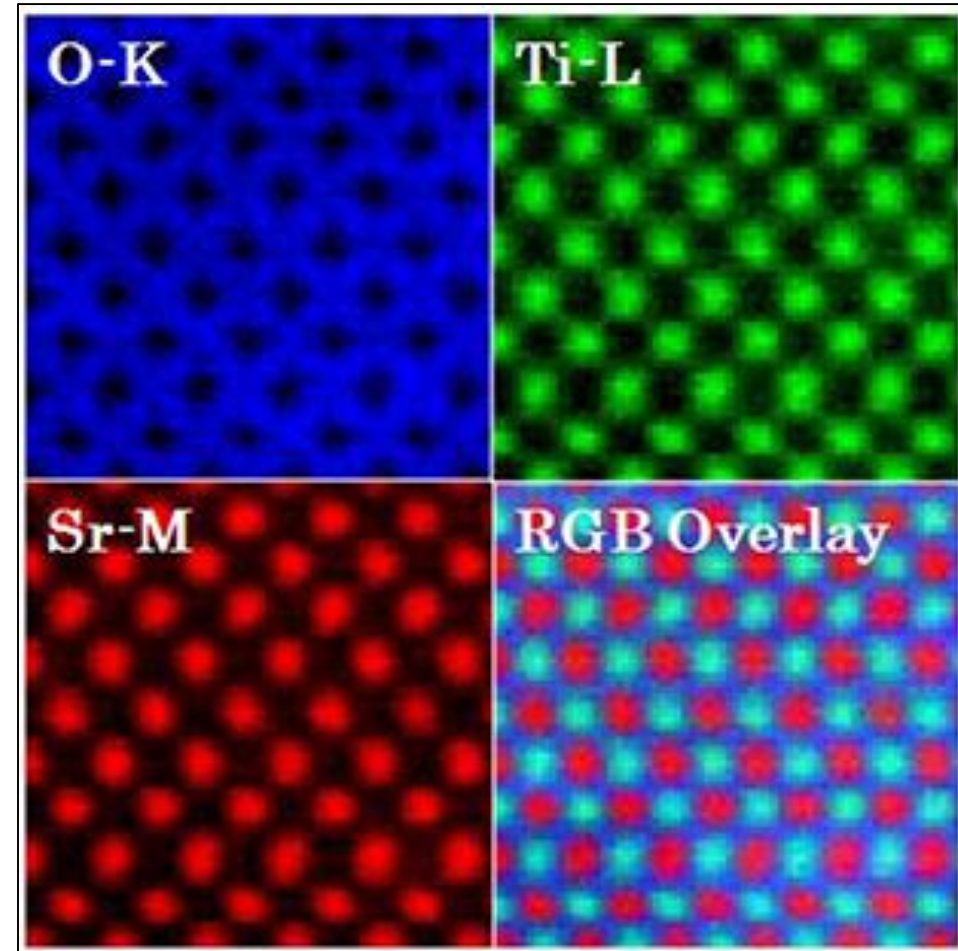
Introduction 2

ADF STEM



Atomic Resolution Elemental Mapping on SrTiO₃ crystal

Multidimension STEM – element map



[<https://www.jeol.com/words/emterms/20121023.030059.php#gsc.tab=0>]



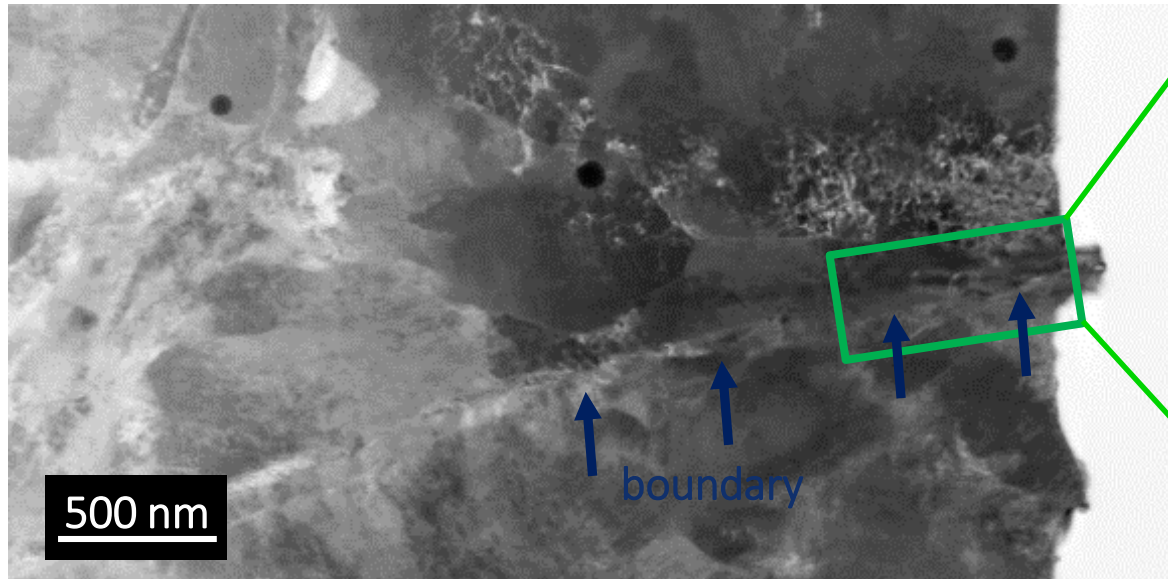
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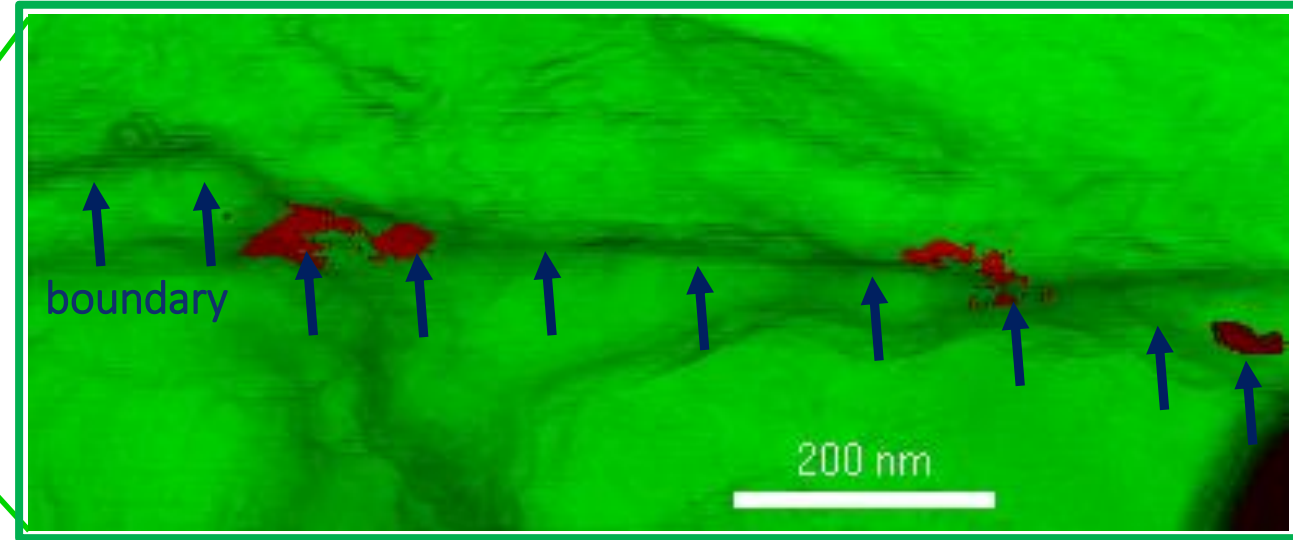
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Introduction 3

HAADF STEM



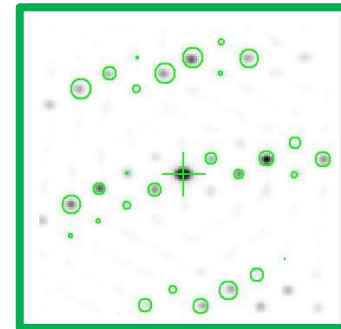
Multidimension STEM – phase map



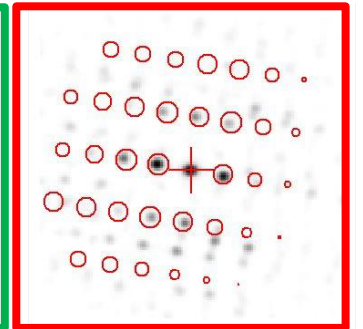
High strength maraging stainless steels with Ti addition

- traces of austenite affecting crack initiation and propagation

Martensite



Austenite



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Academy of Sciences

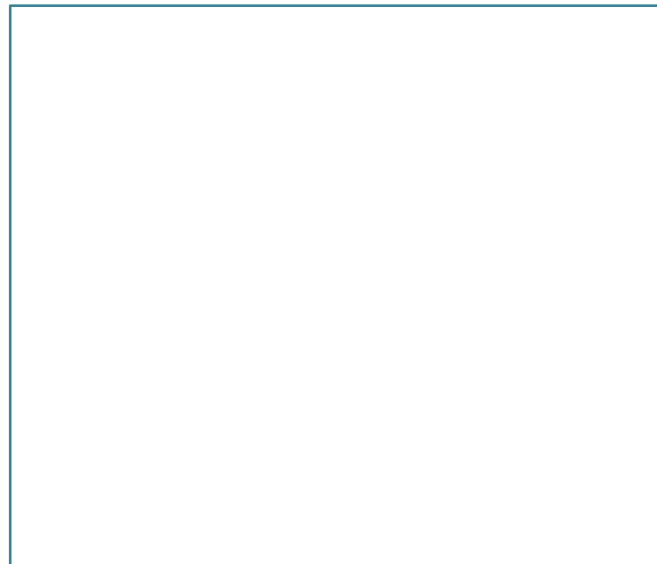
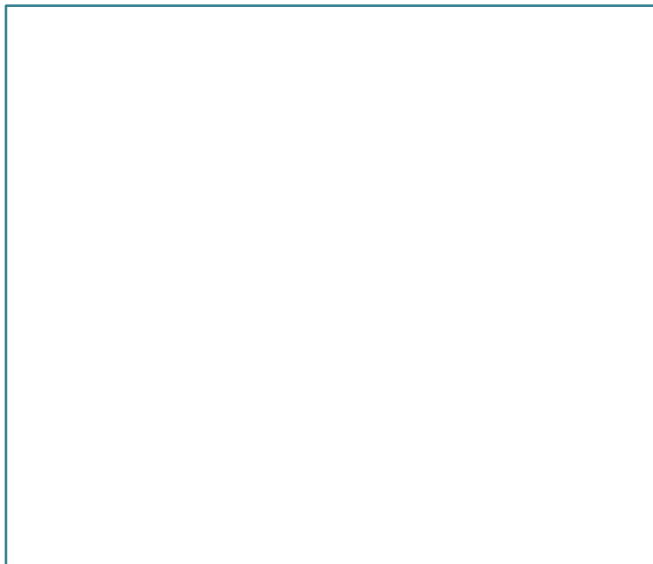
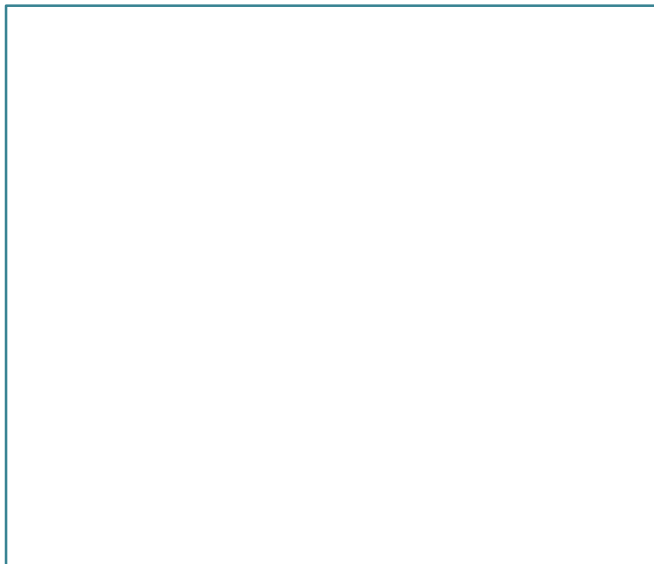
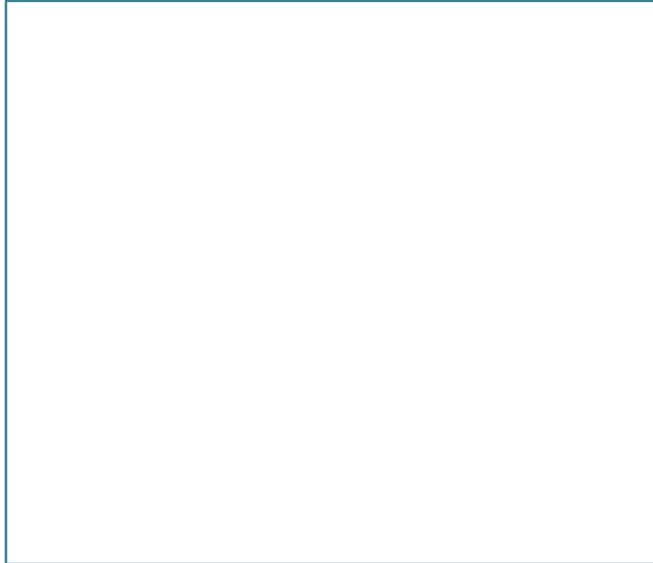
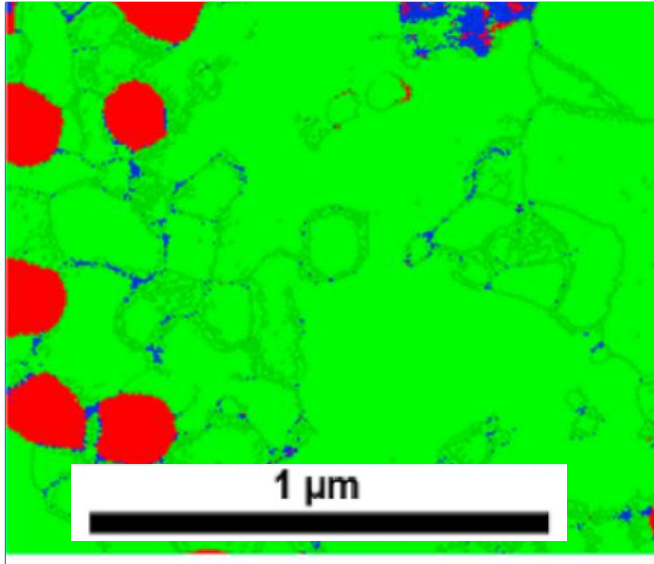
SCIENCE
LUNCH

4D-STEM

Overview and outline

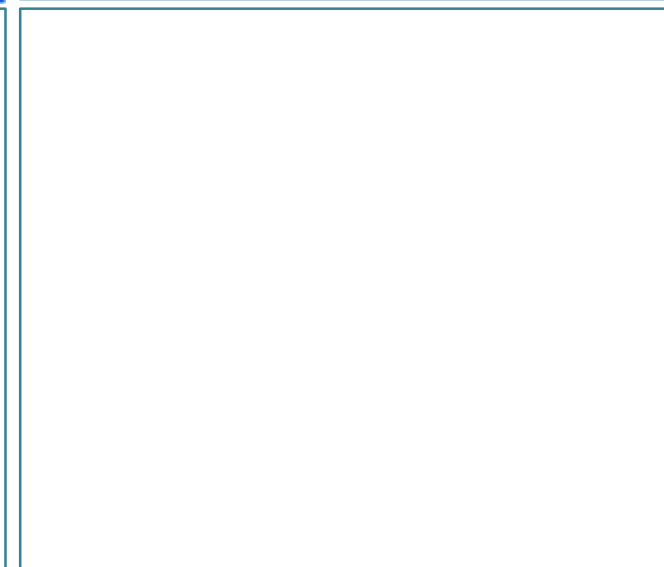
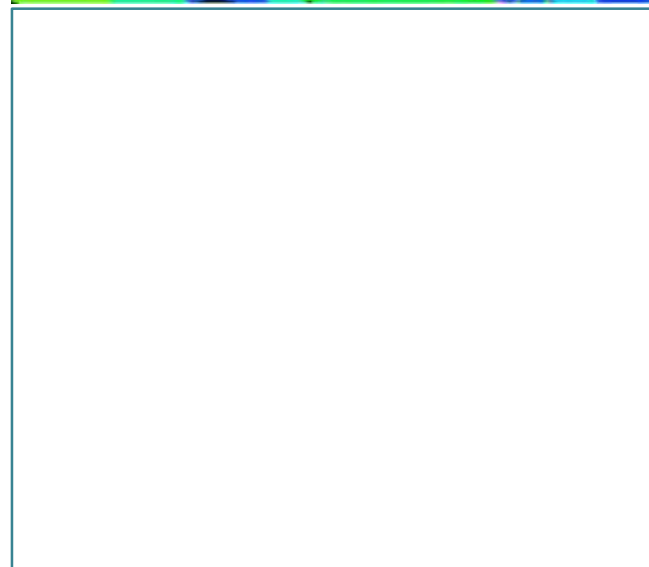
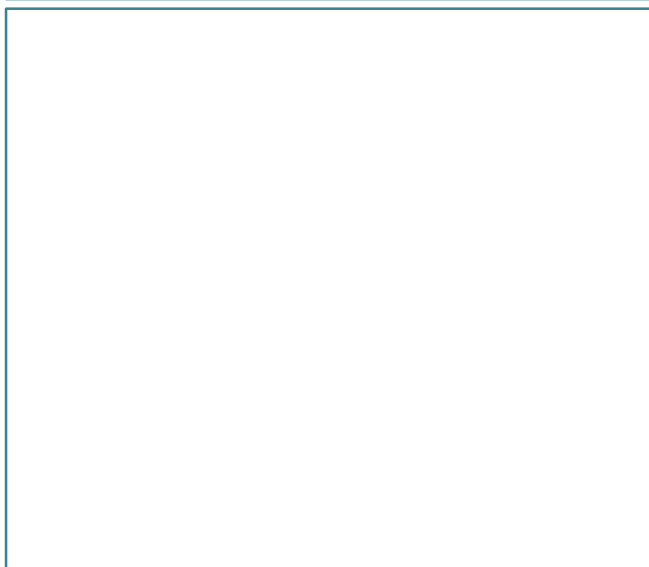
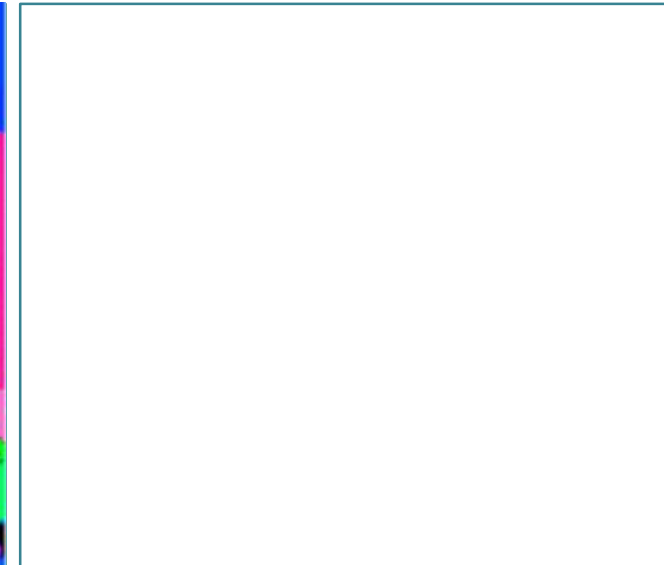
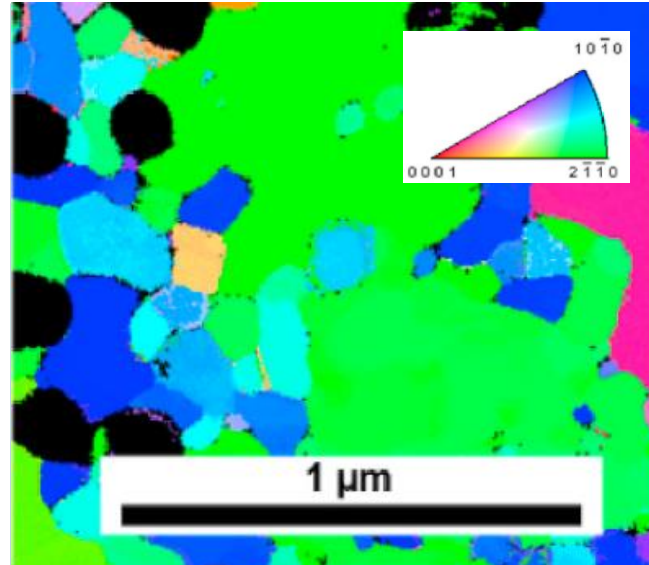
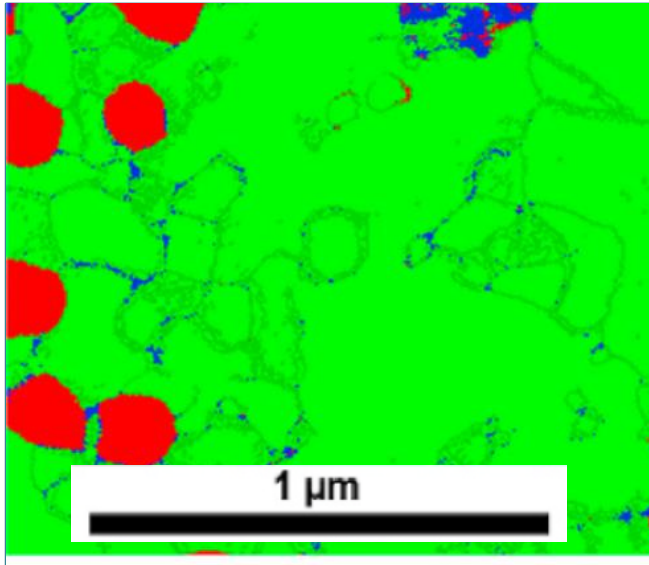
4D-STEM

Overview and outline



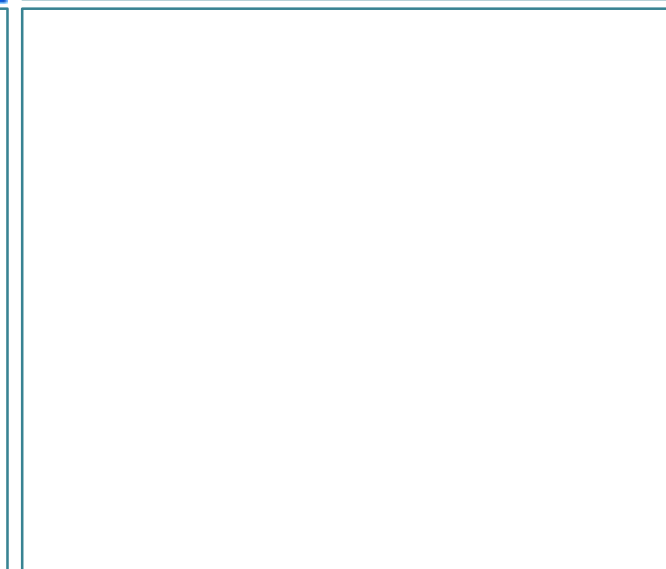
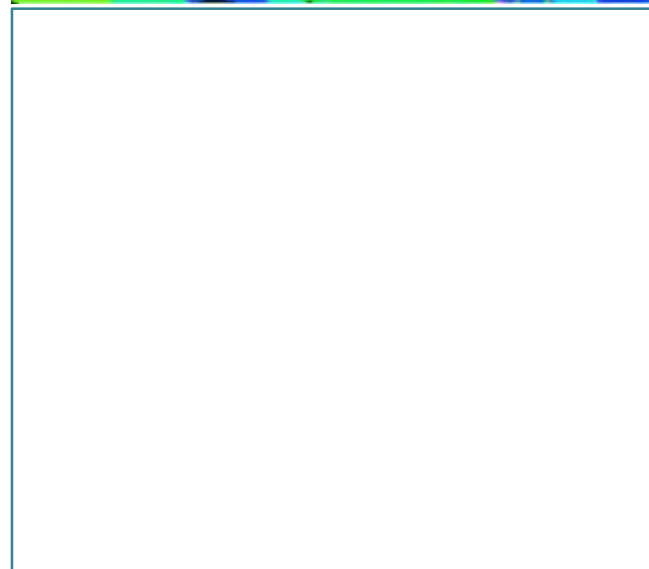
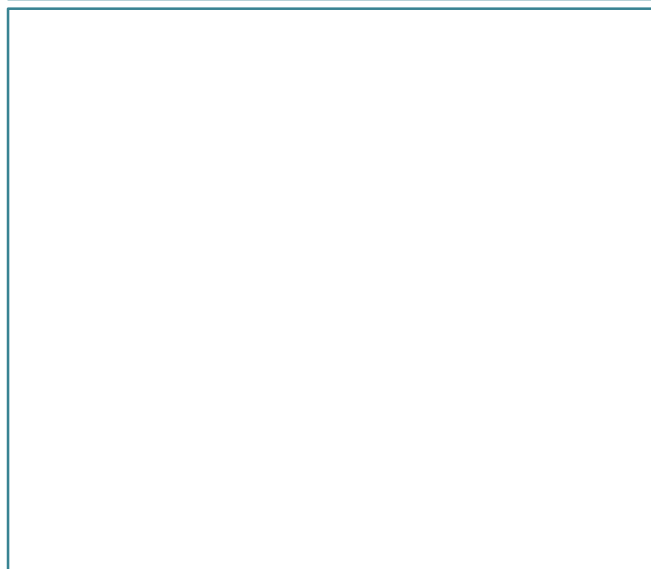
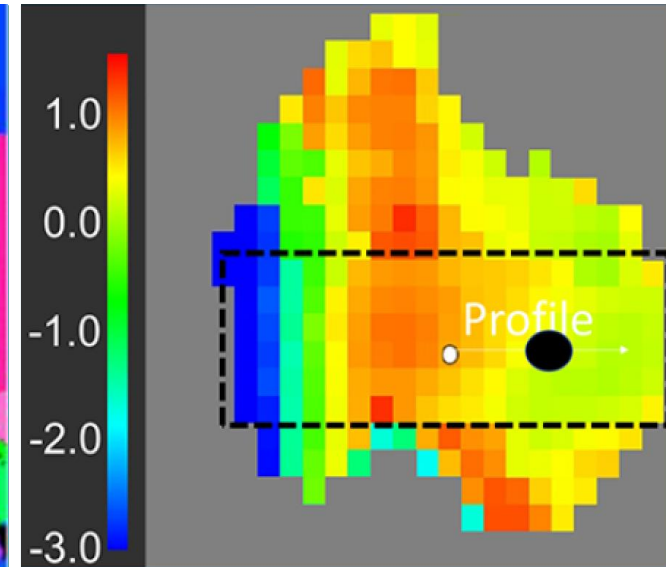
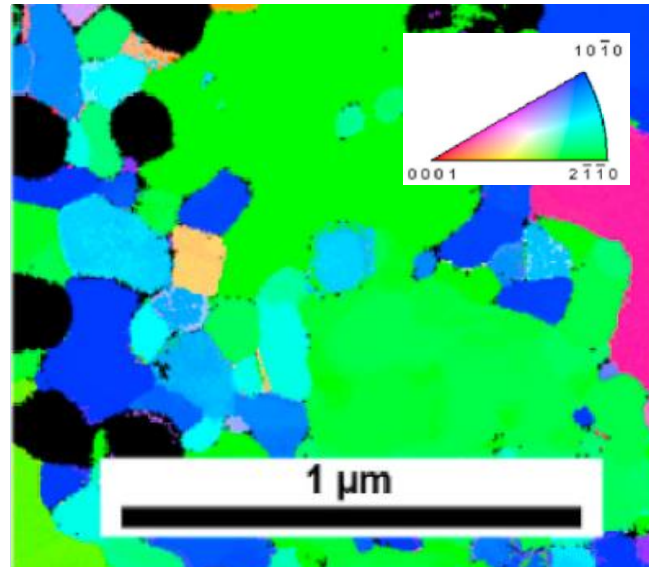
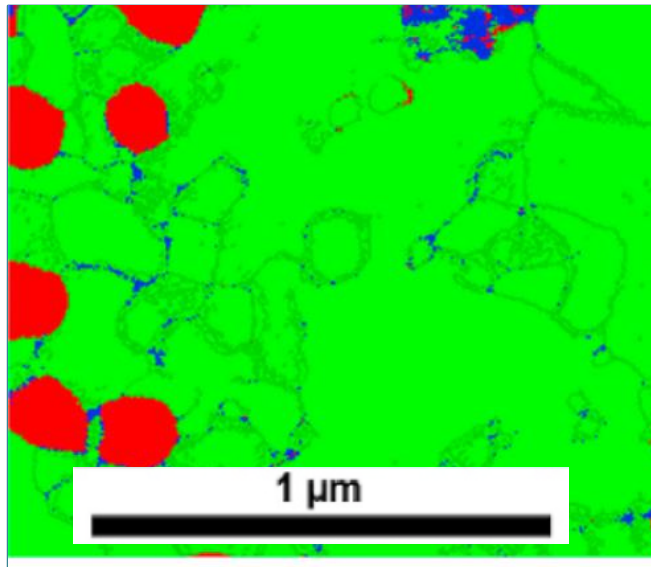
4D-STEM

Overview and outline



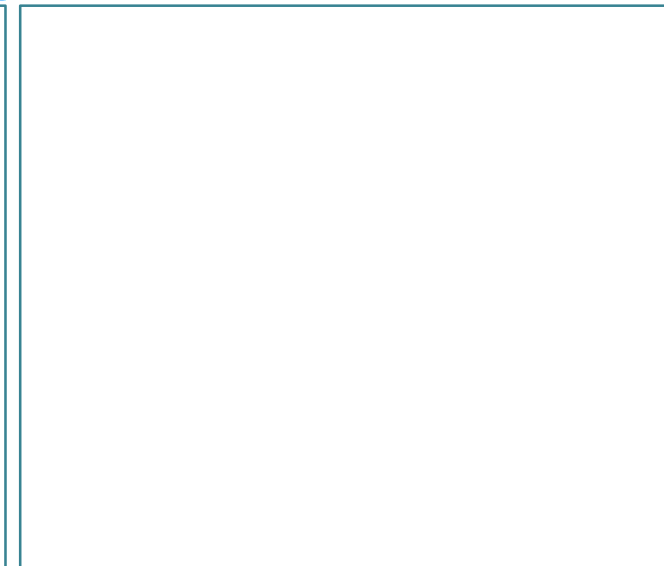
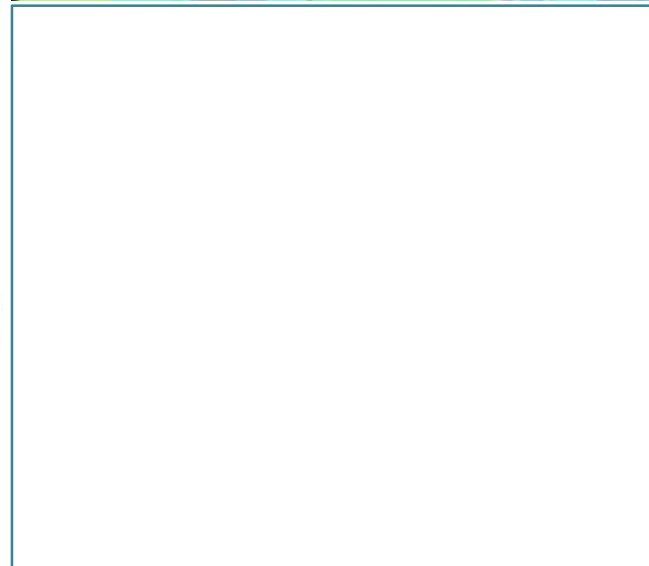
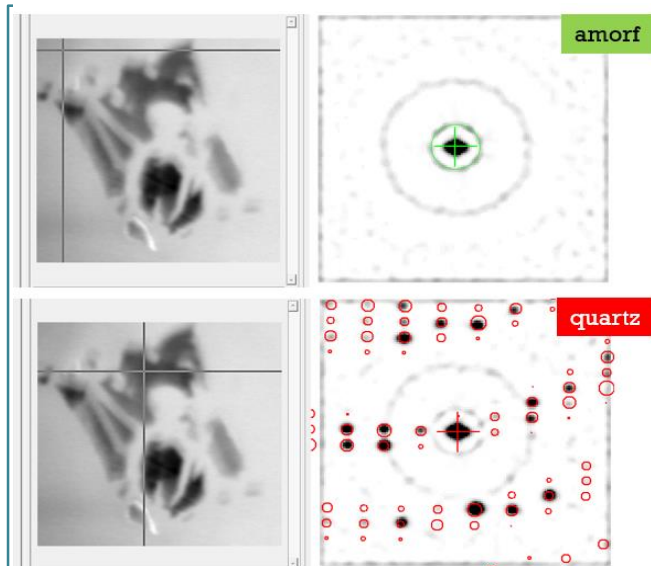
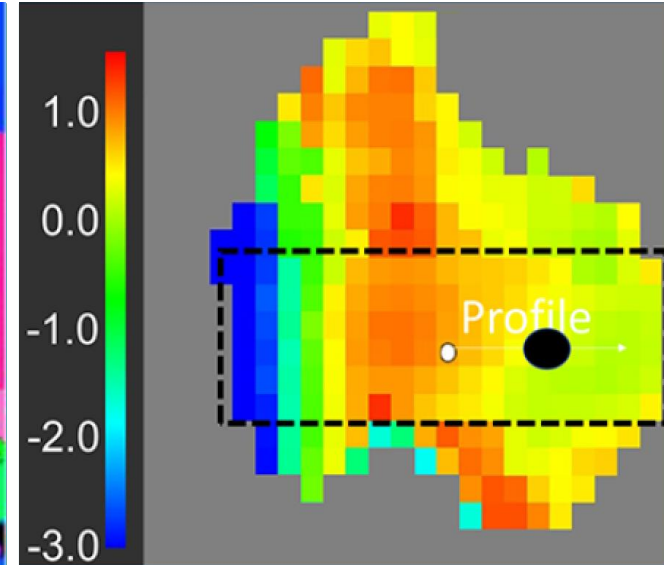
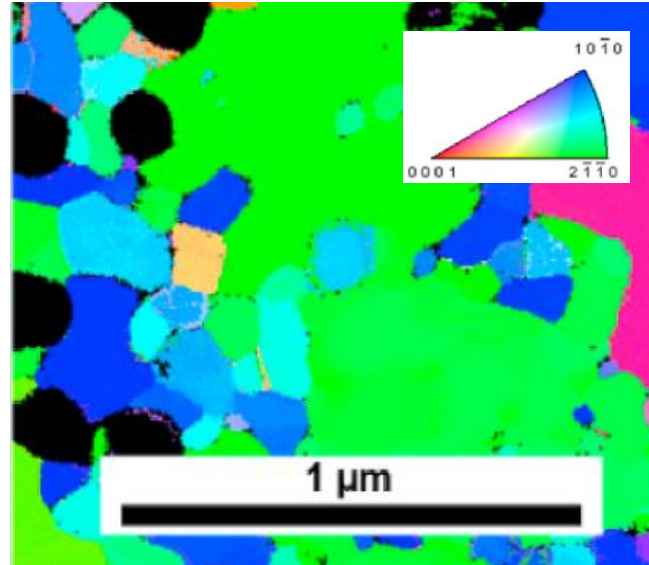
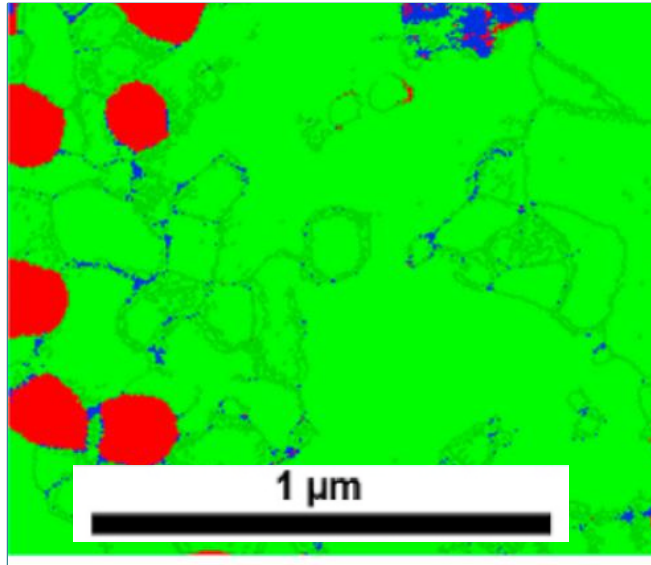
4D-STEM

Overview and outline



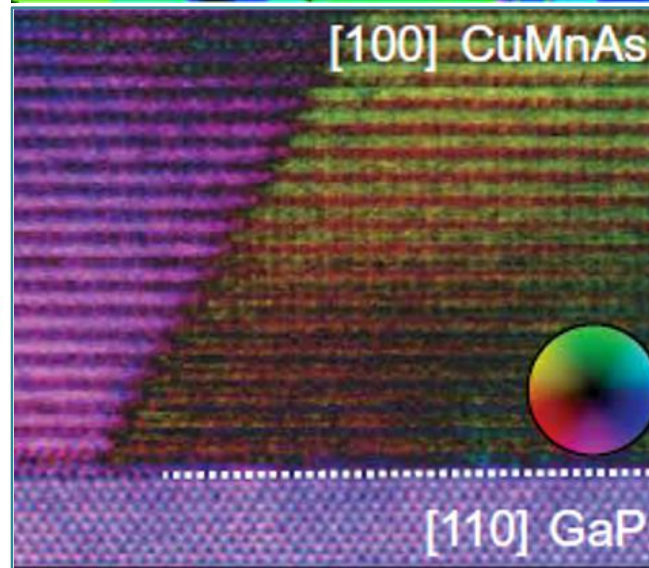
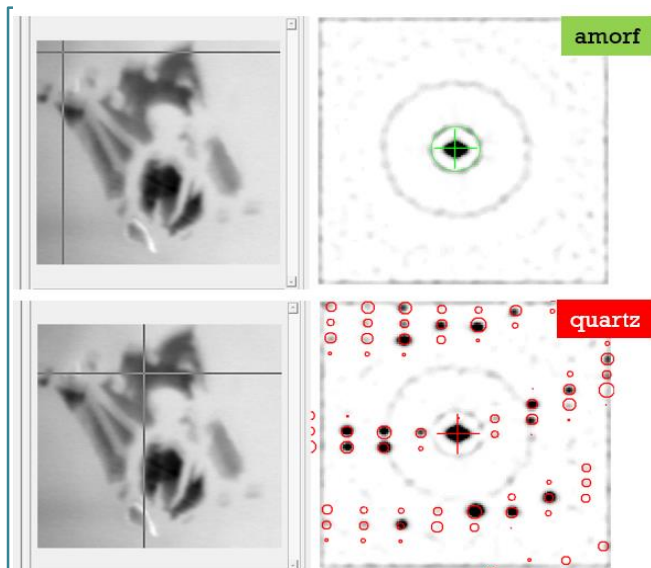
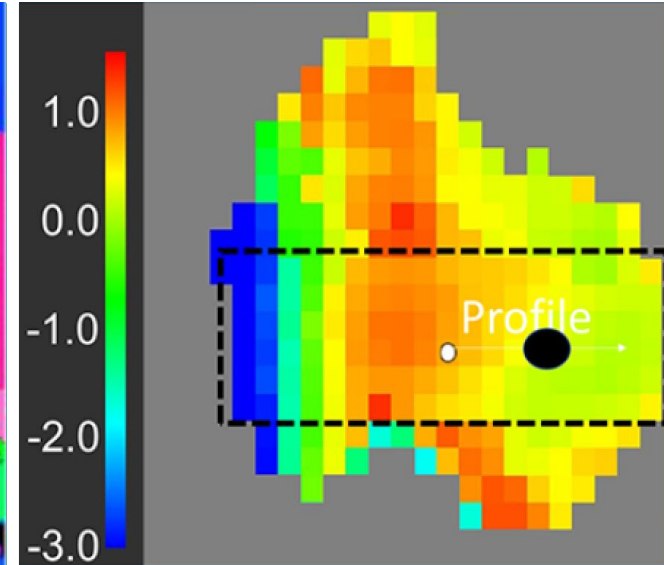
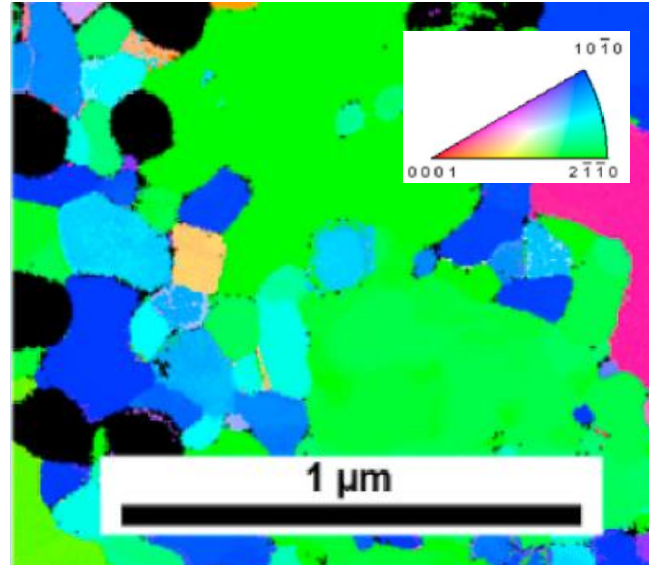
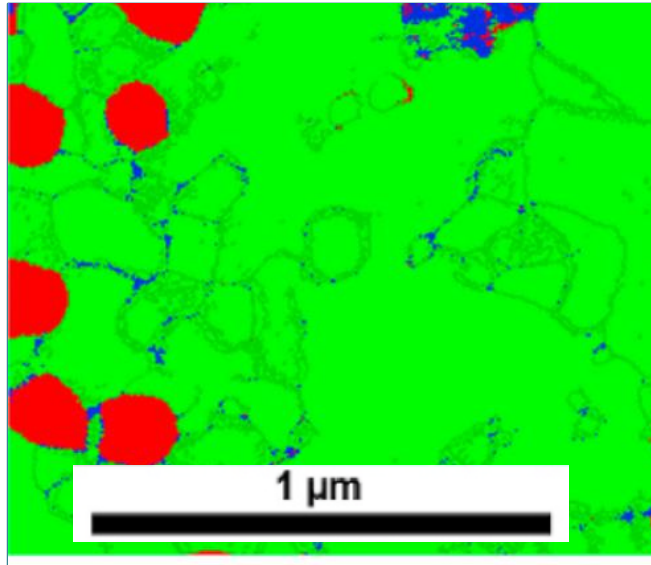
4D-STEM

Overview and outline



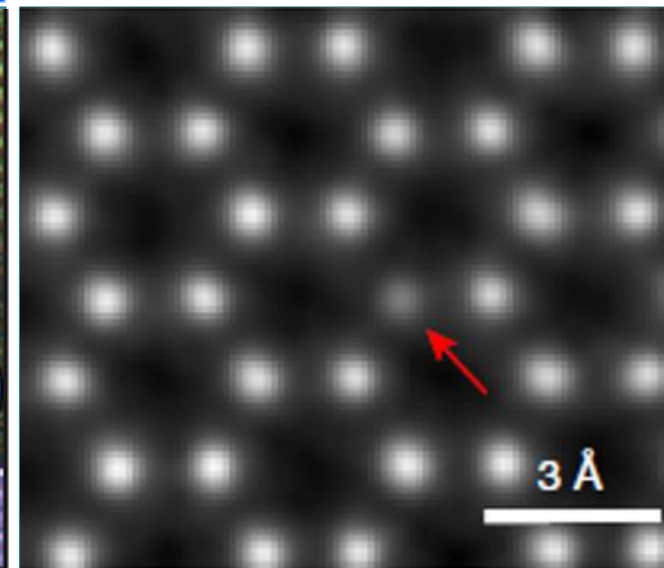
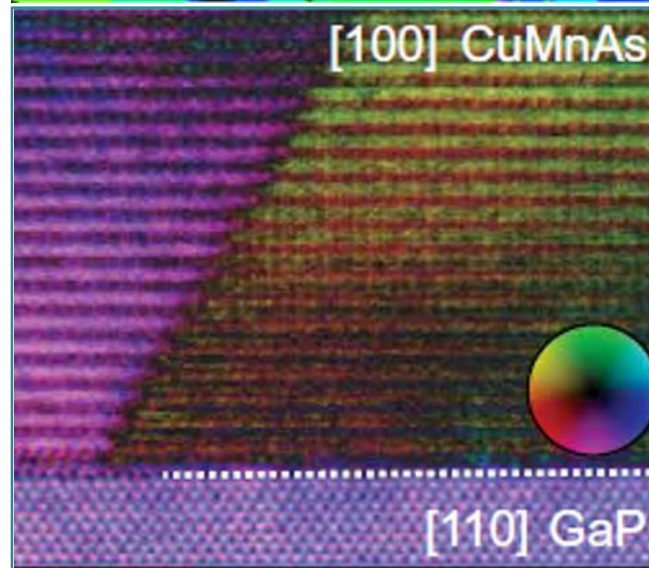
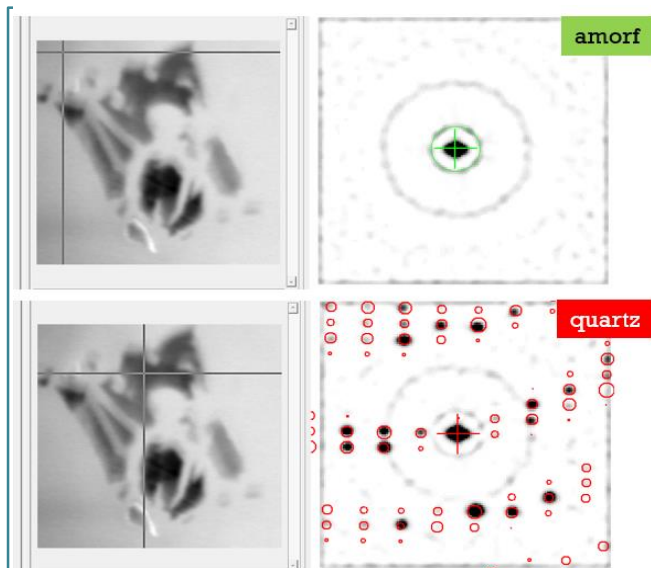
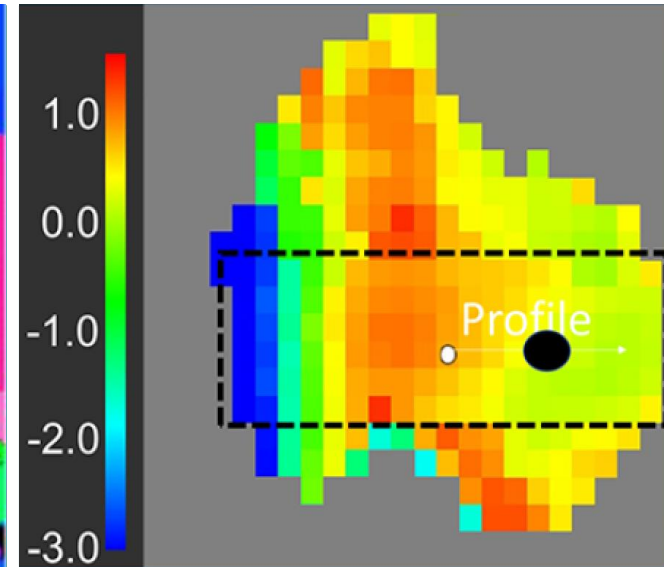
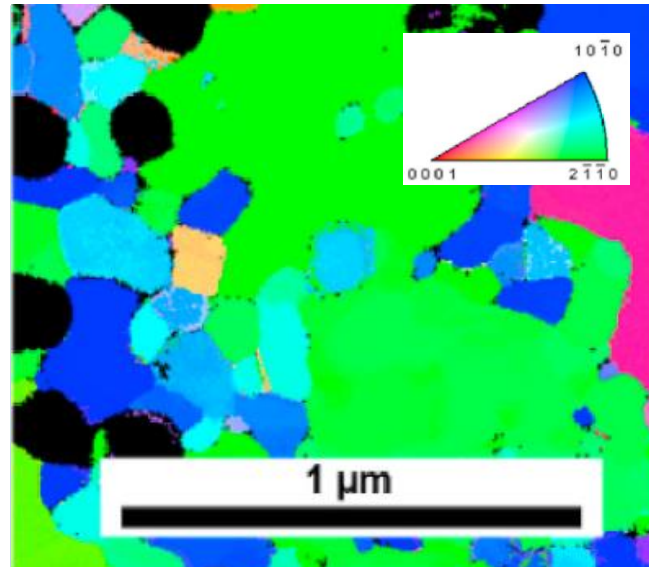
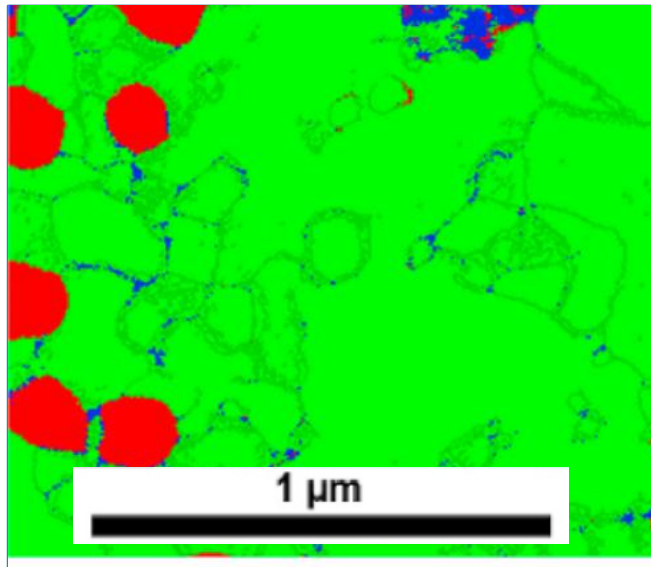
4D-STEM

Overview and outline



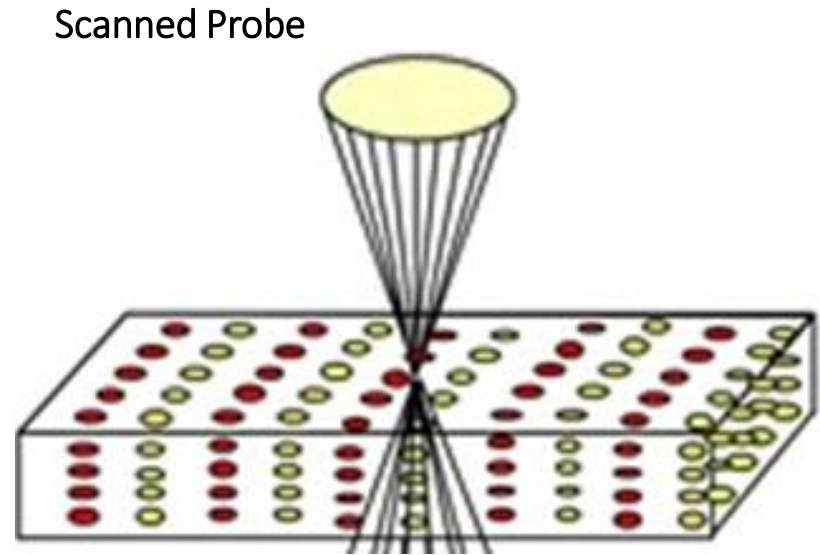
4D-STEM

Overview and outline



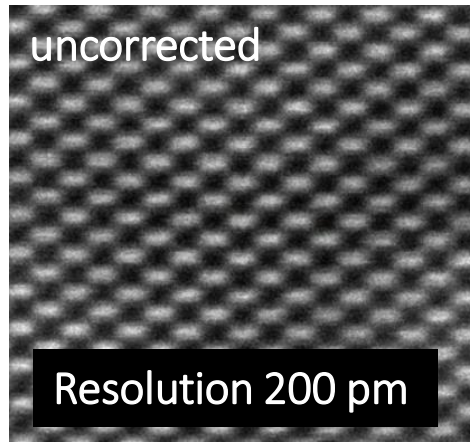
Introduction to STEM

- STEM probe focused on the sample surface, where it propagates through and scatters

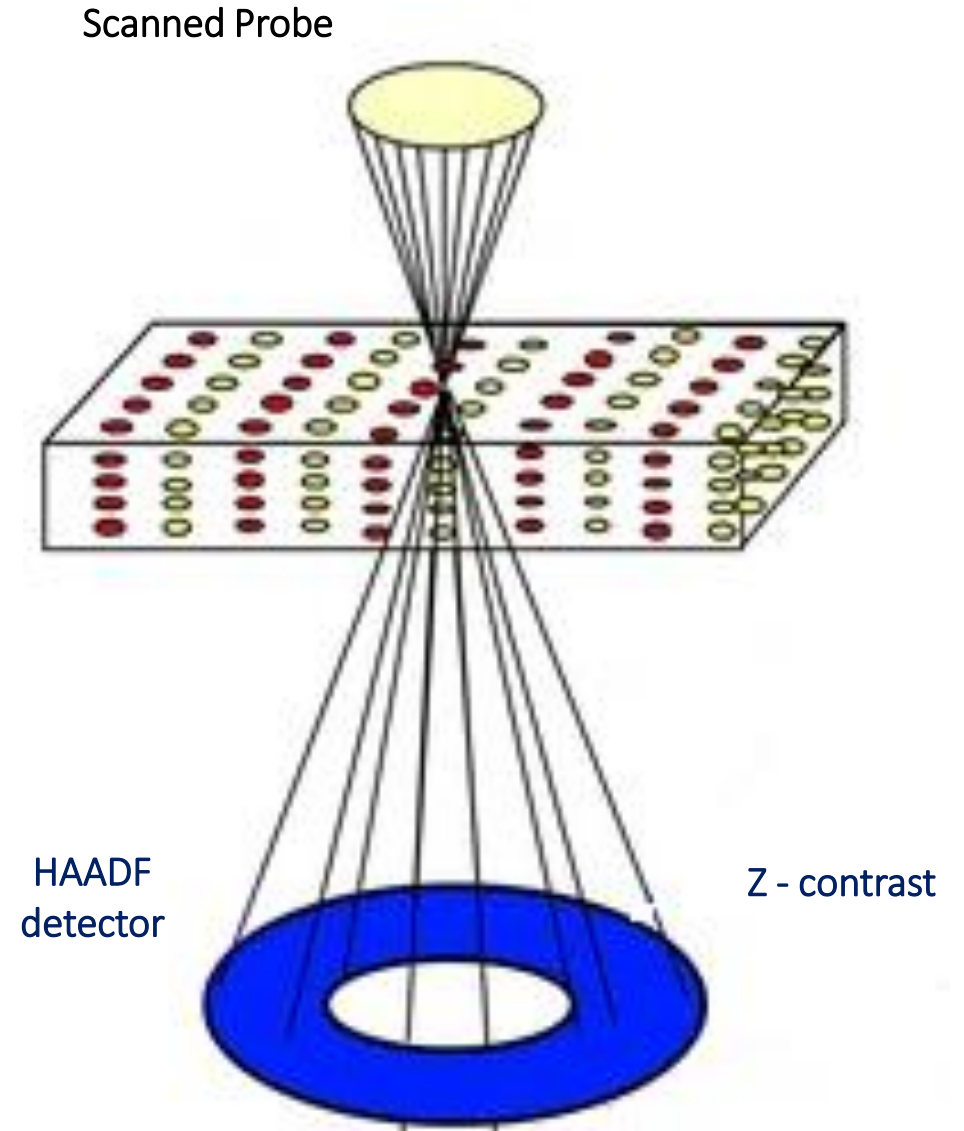


Introduction to STEM

- STEM probe focused on the sample surface, where it propagates through and scatters
- In conventional STEM up to 1D information from a 2D grid of probe positions



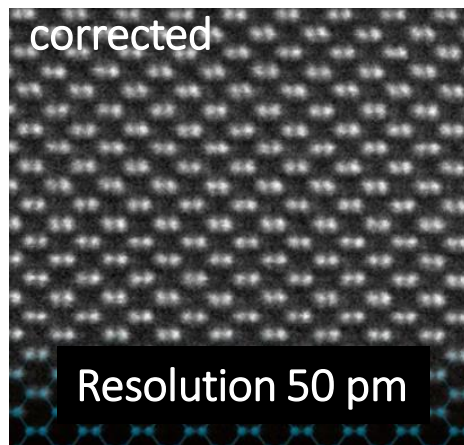
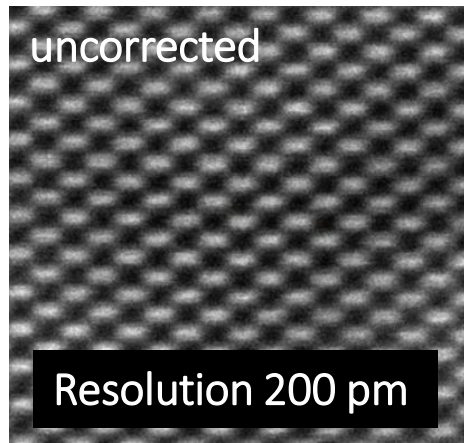
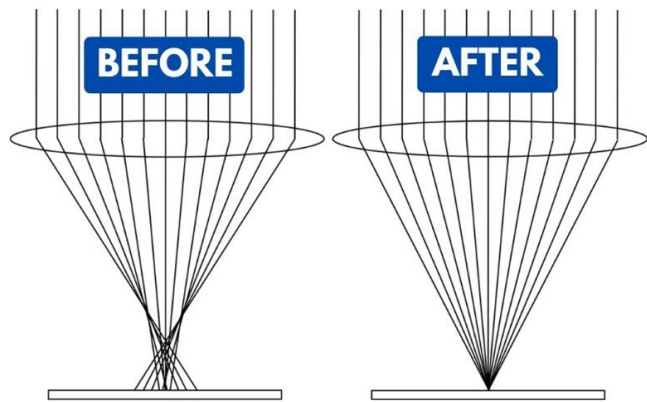
Si crystal



Introduction to STEM

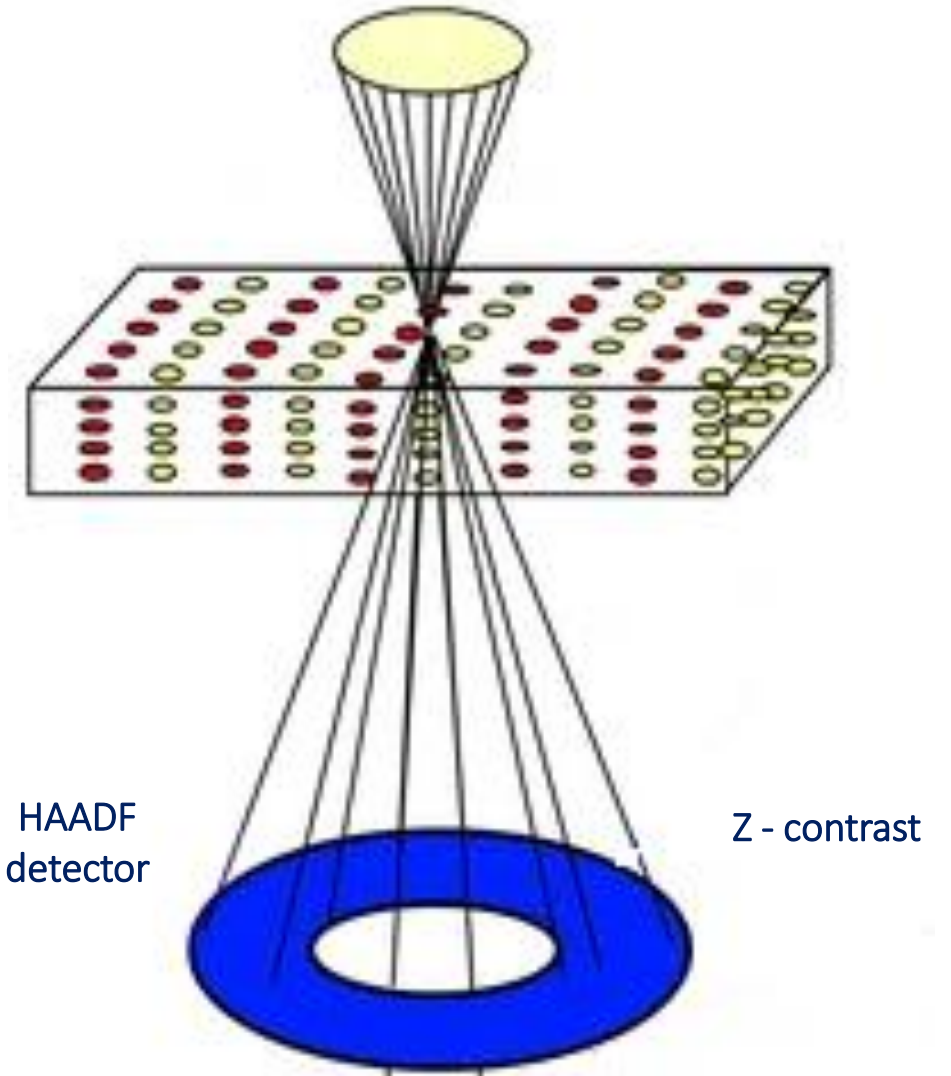
- STEM probe focused on the sample surface, where it propagates through and scatters
- In conventional STEM up to 1D information from a 2D grid of probe positions
- Probe size is critical for resolution

Cs-CORRECTED S/TEM



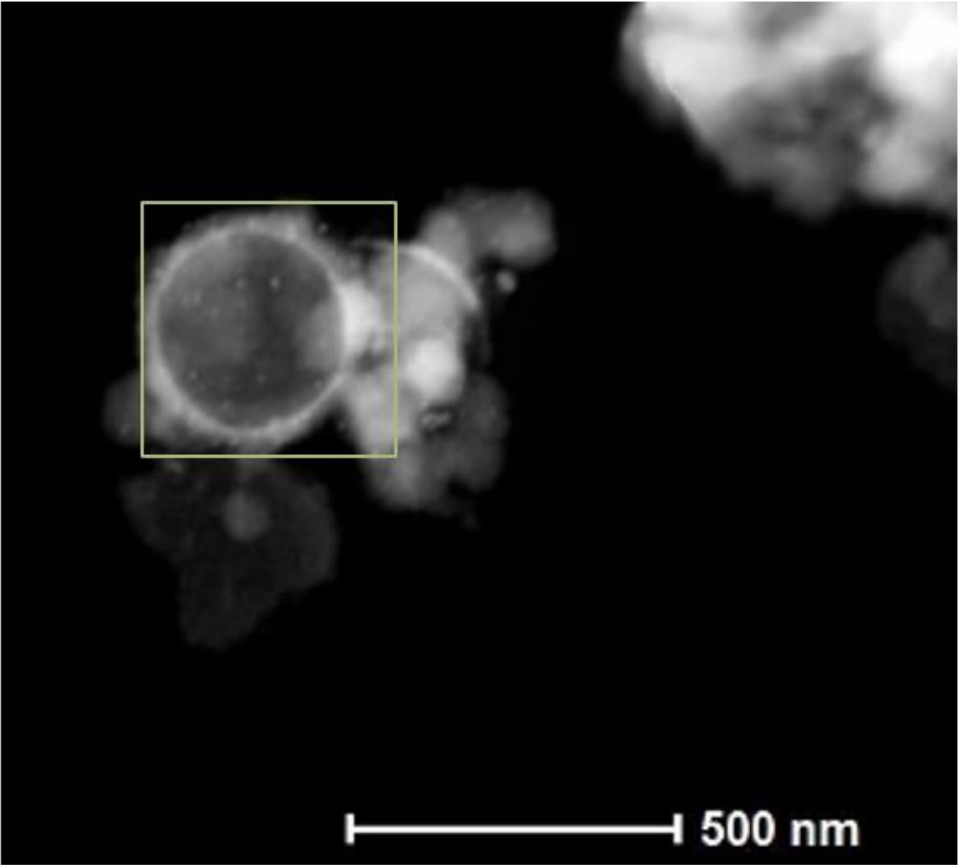
[Ultramicroscopy 189 (2018) 46–53]

Scanned Probe

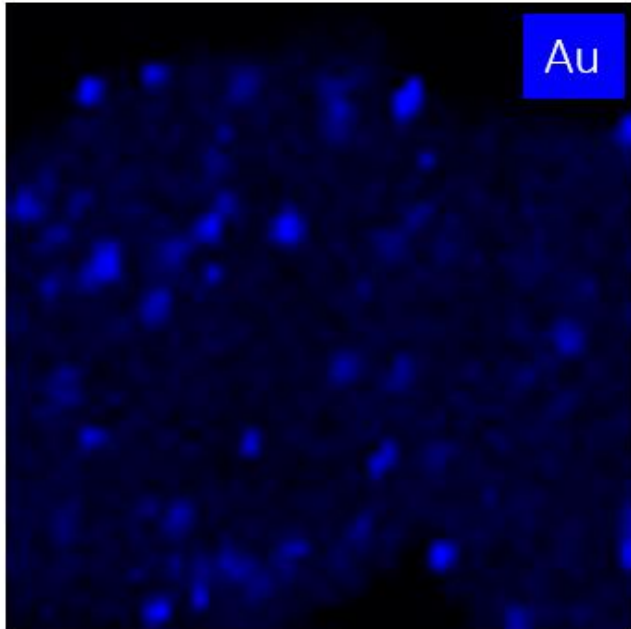
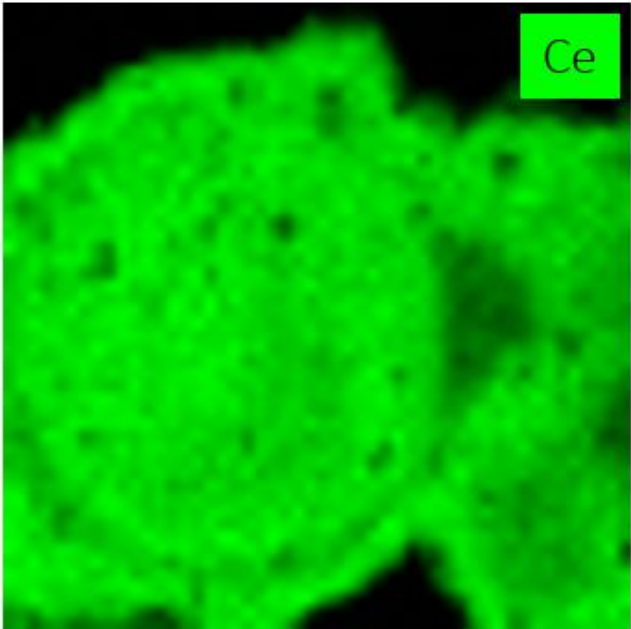
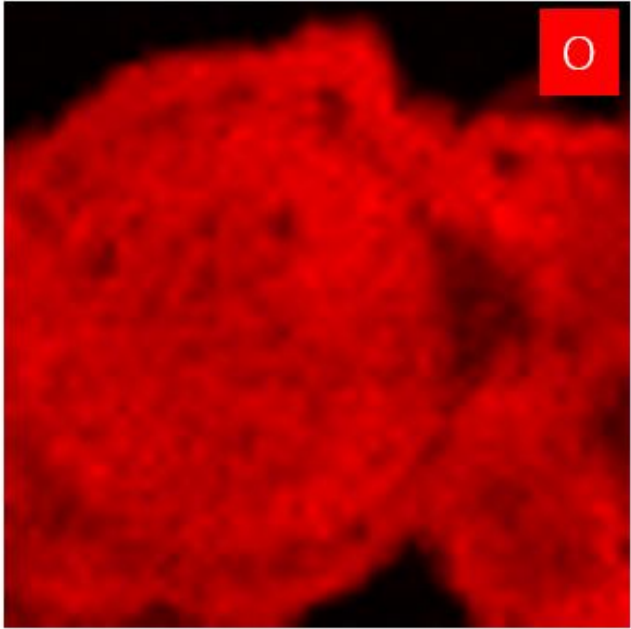
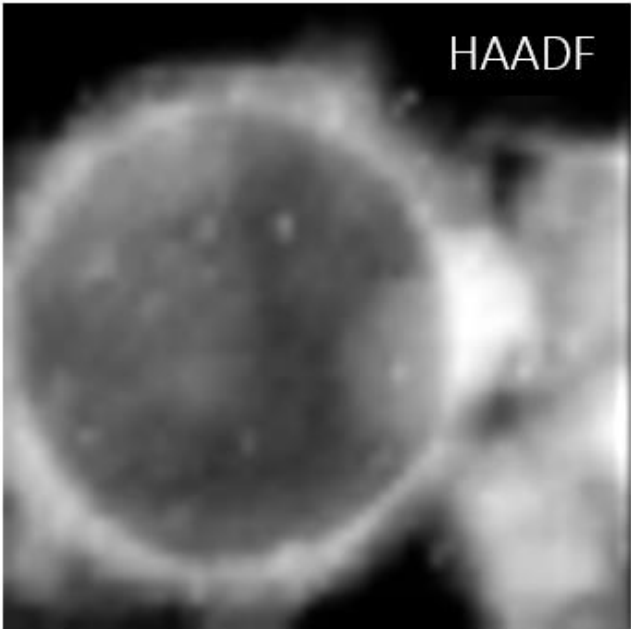


Si crystal, dumbbells separated at 130 pm in silicon [110]

A path to multidimension STEM

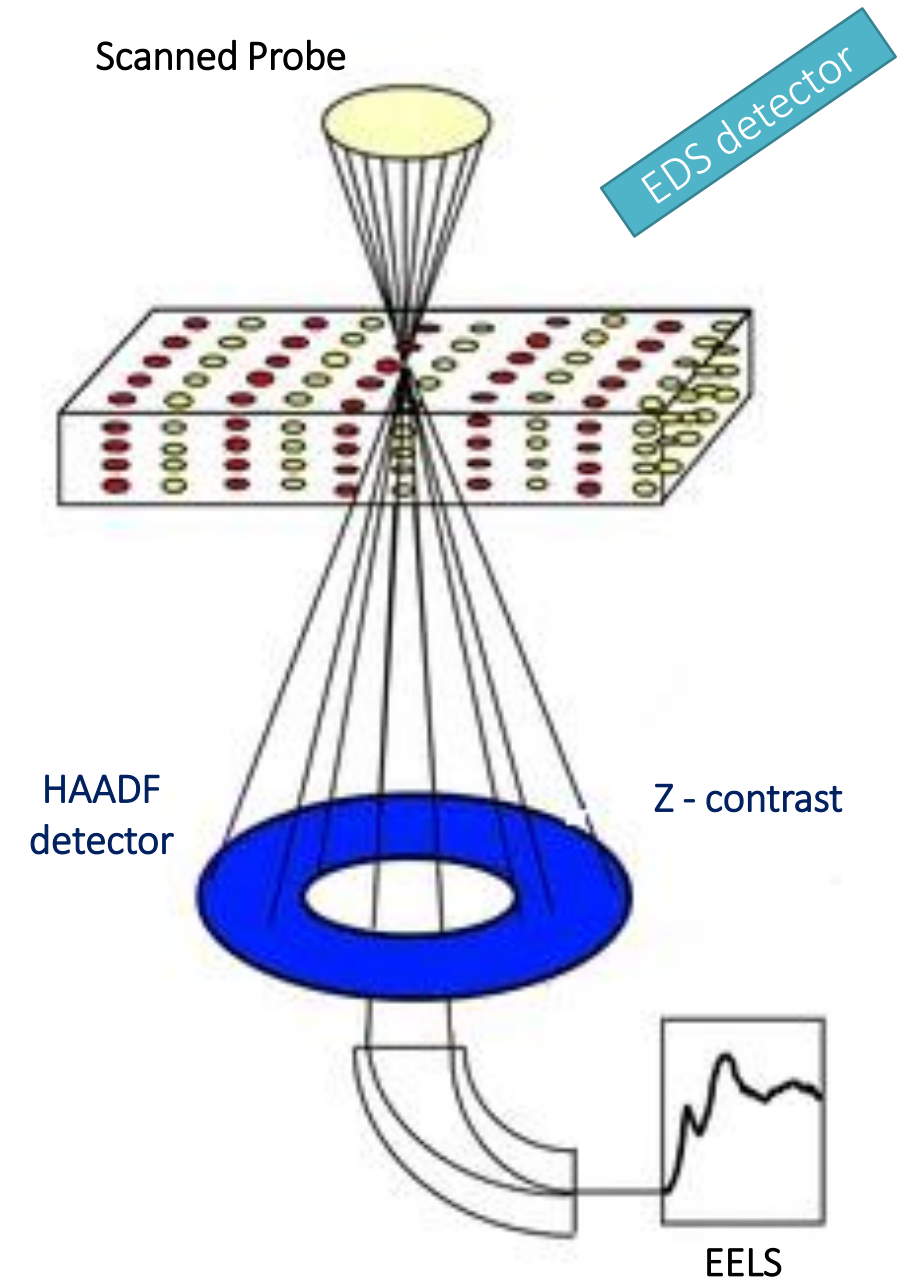


Au nanoparticles on CeO₂ spheres



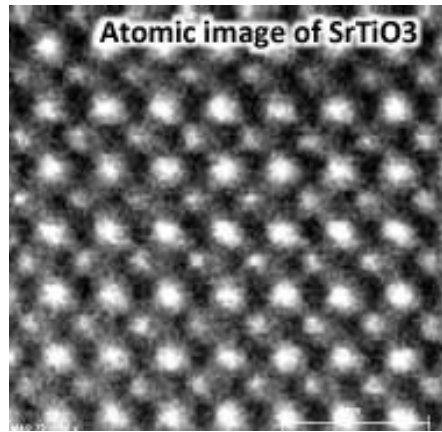
A path to multidimension STEM

- STEM probe focused on the sample surface, where it propagates through and scatters
- In conventional STEM up to 1D information from a 2D grid of probe positions
- Probe size is critical for resolution
- After the STEM probe scatters from the sample, spectroscopic signals that can be simultaneously measured

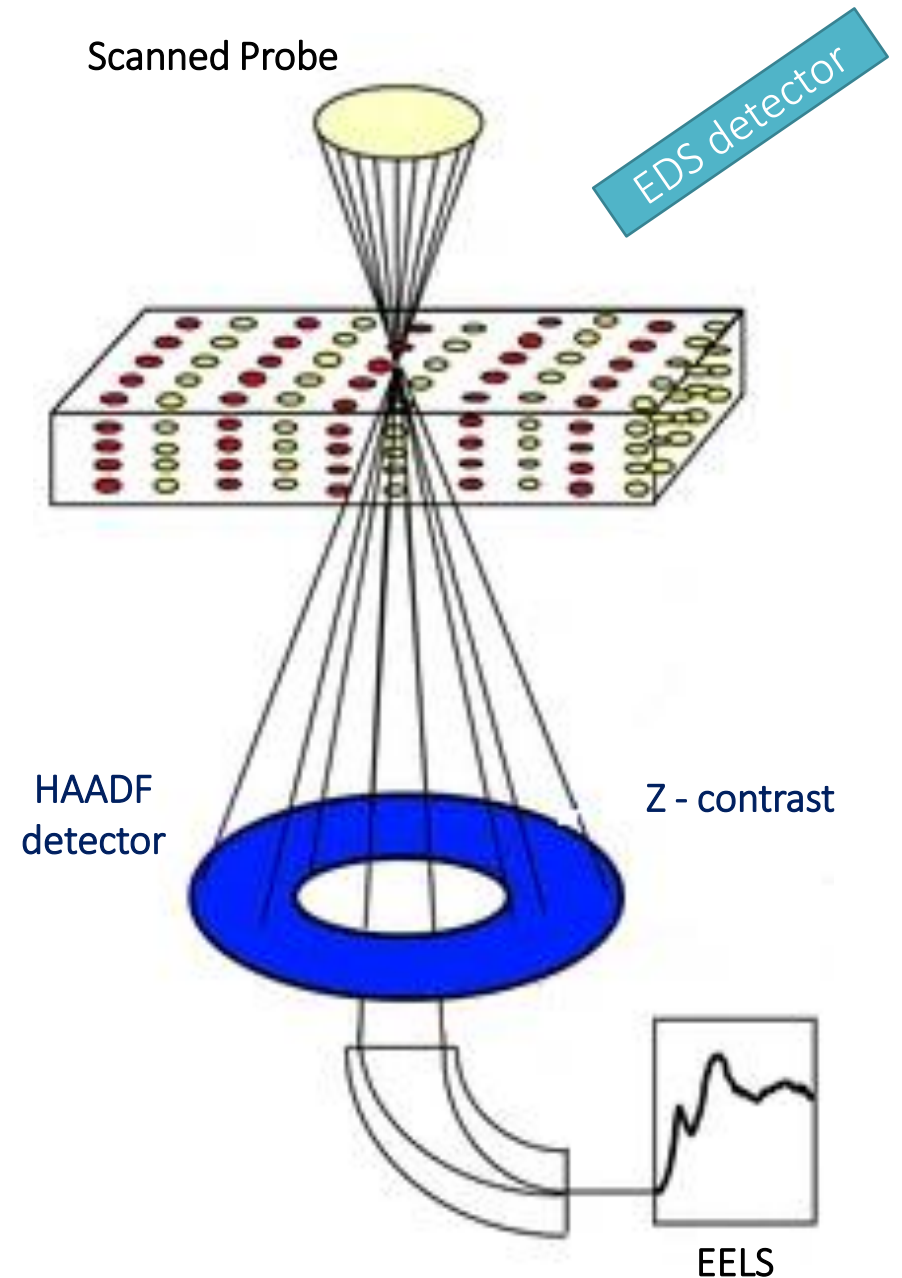
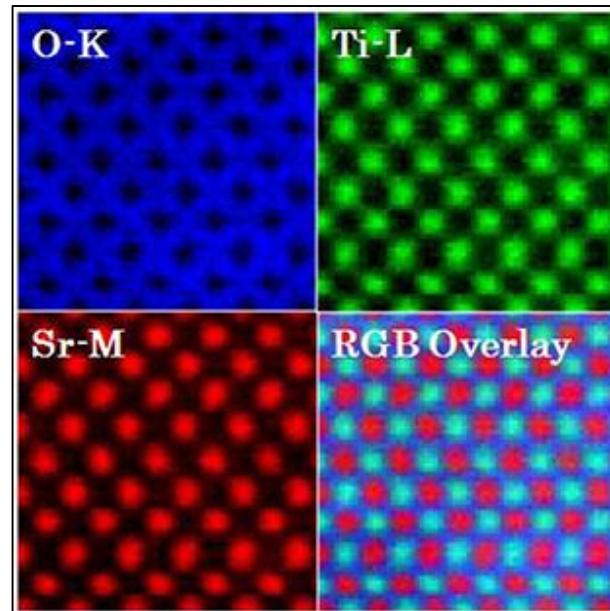


A path to multidimension STEM

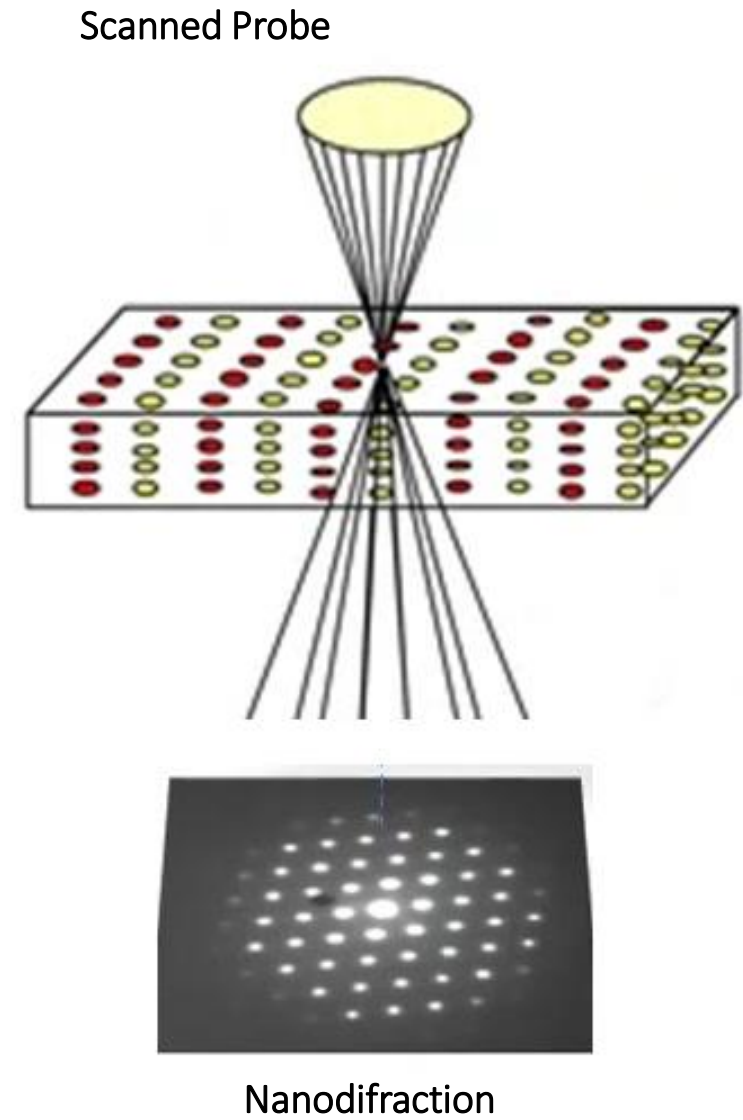
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Atomic Resolution Elemental Mapping on SrTiO₃ crystal

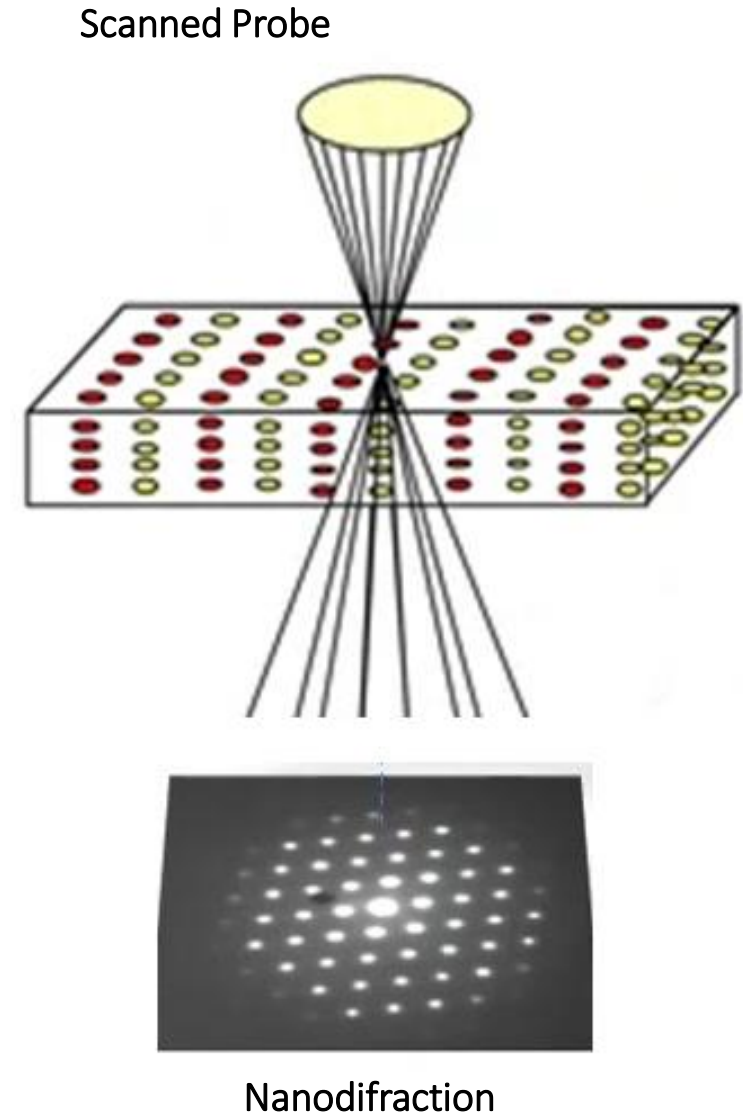


A path to 4D STEM



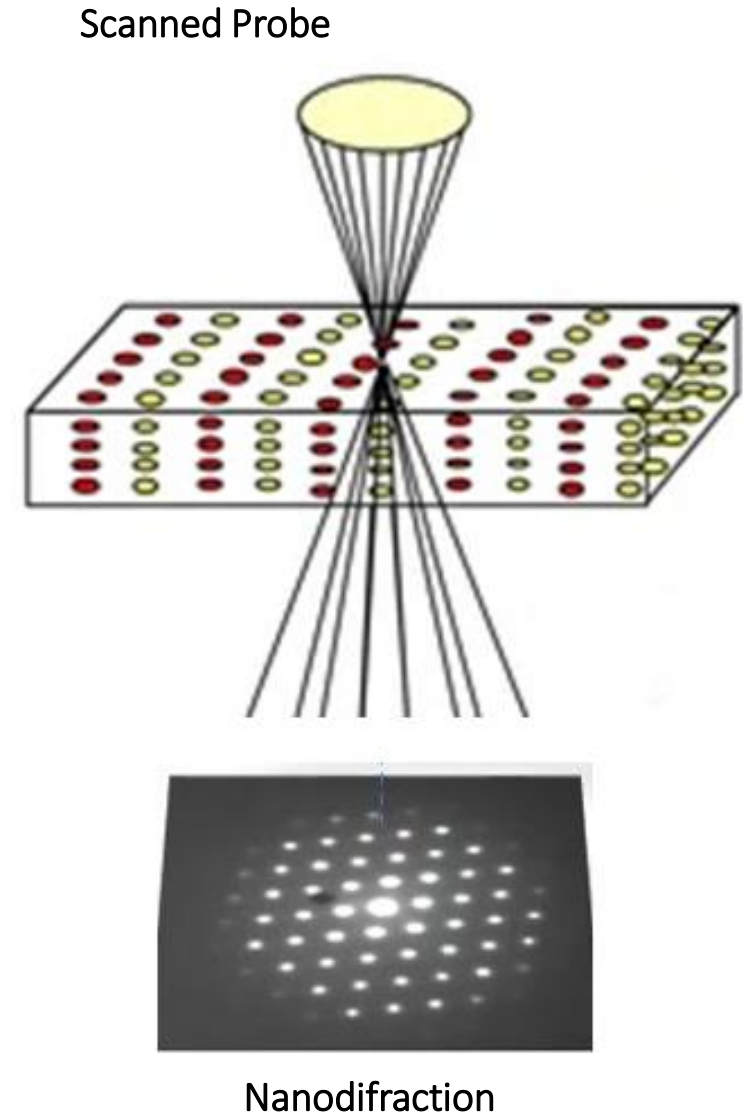
A path to 4D STEM

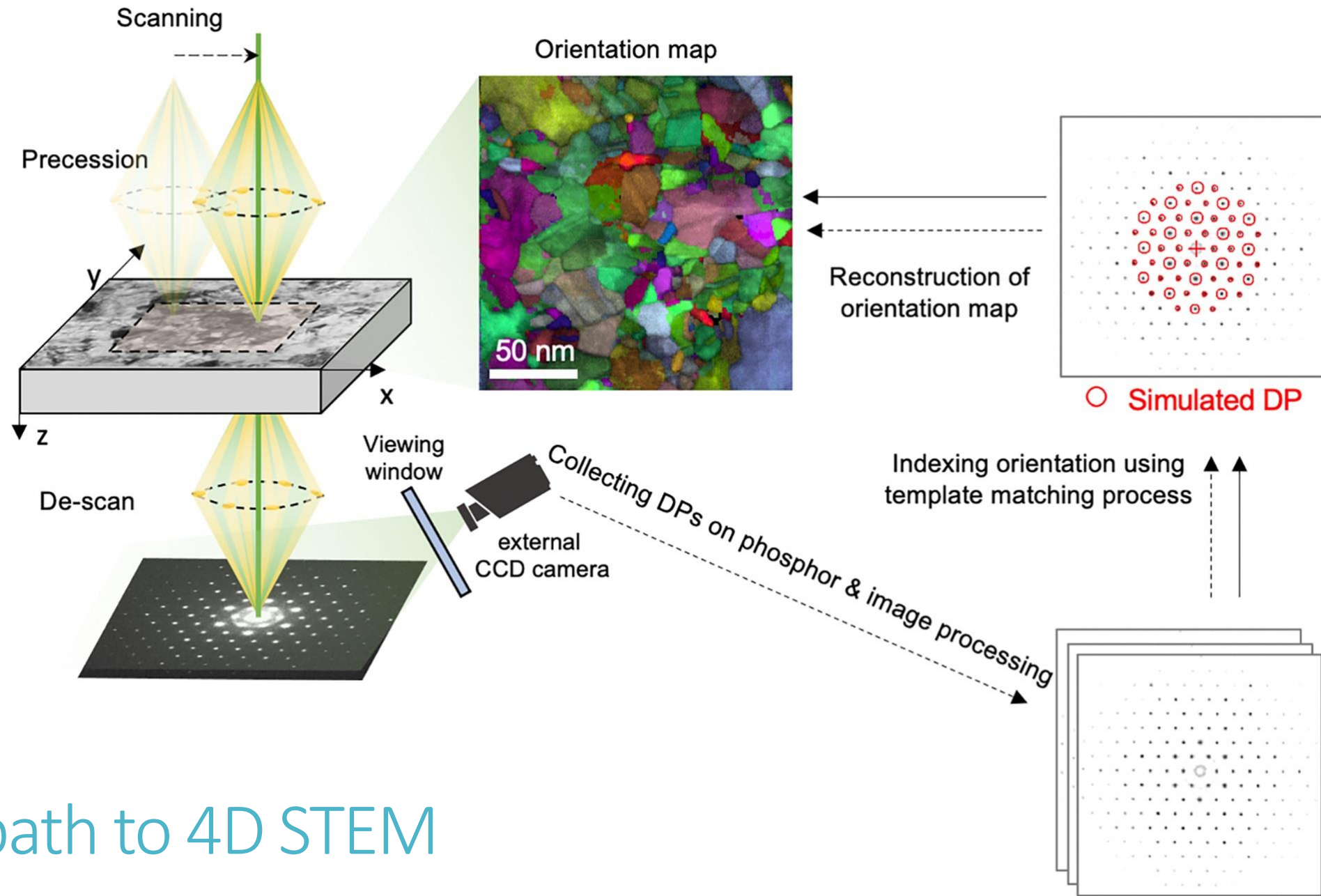
- The name “4D-STEM” refers to recording 2D images of a converged electron probe, over a 2D grid of probe positions
- STEM detectors - single value per STEM probe position
- TEM cameras – limited readout speeds and dynamic range



A path to 4D STEM

- The name “4D-STEM” refers to recording 2D images of a converged electron probe, over a 2D grid of probe positions
- STEM detectors - single value per STEM probe position
- TEM cameras – limited readout speeds and dynamic range
- CCD camera placed in front of fluorescent screen

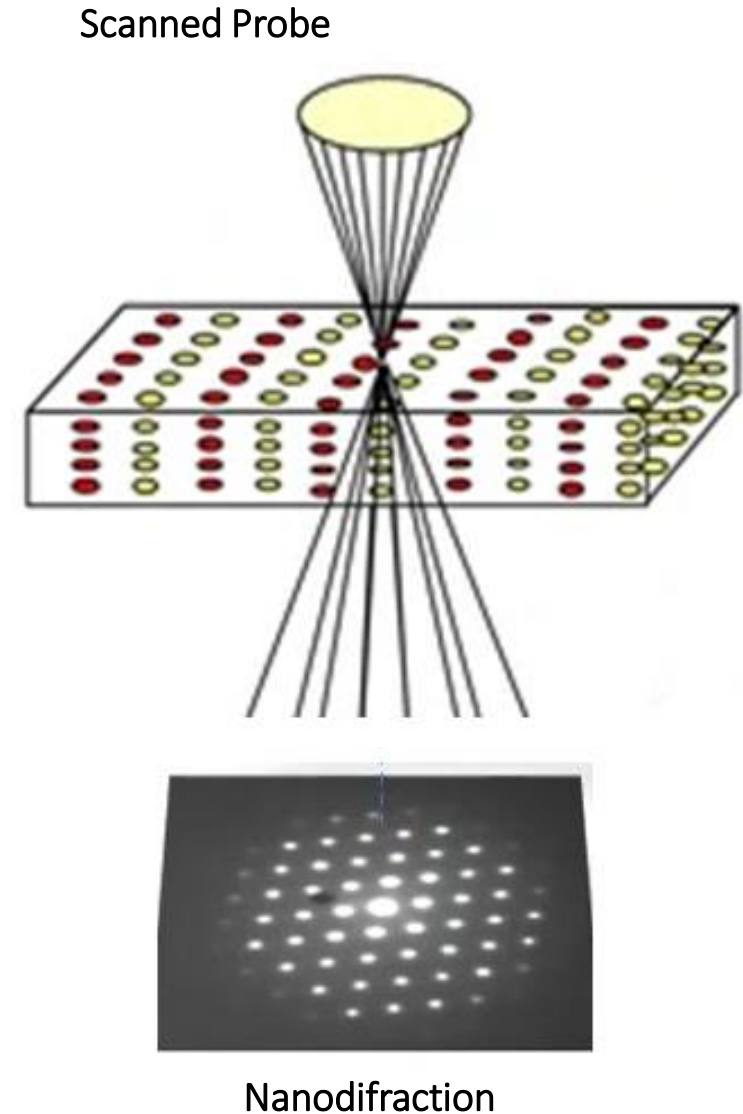




A path to 4D STEM

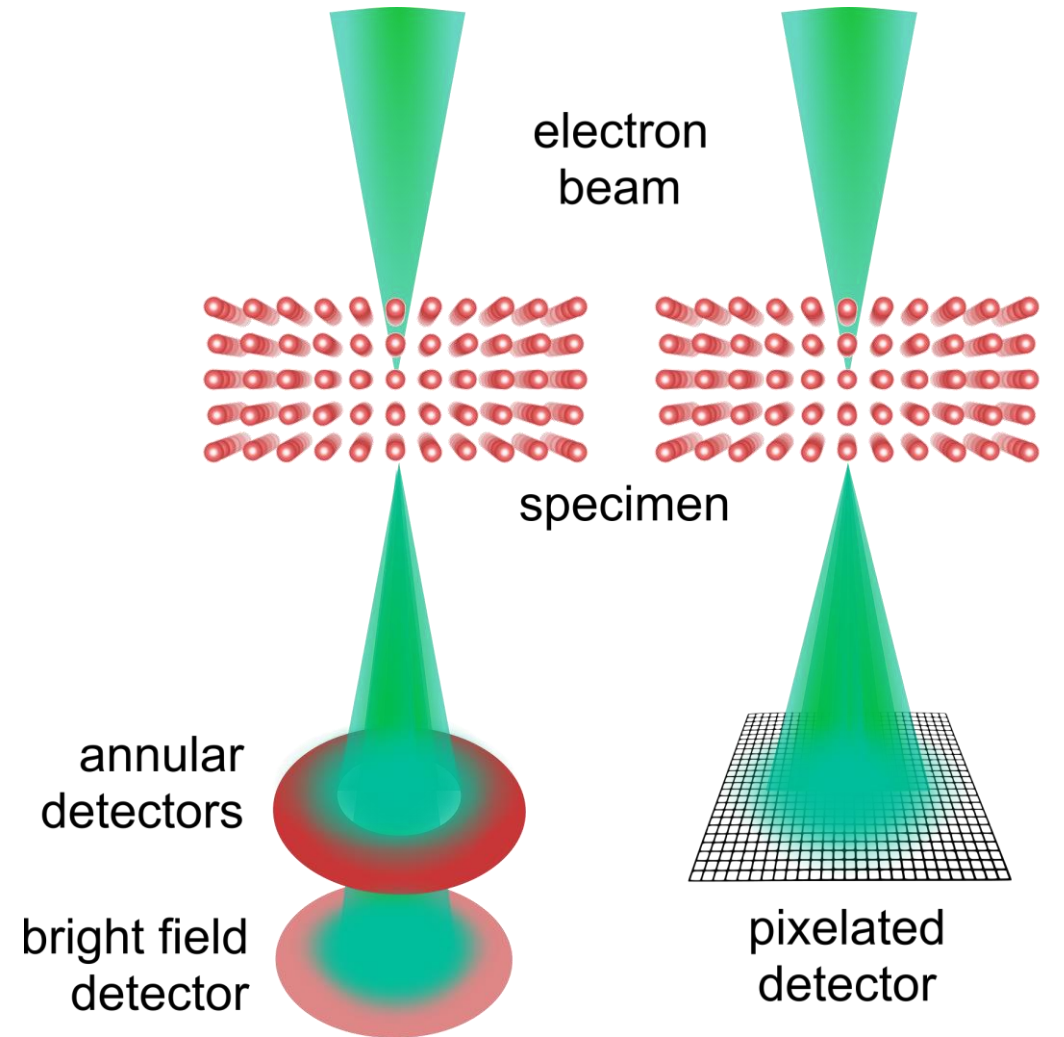
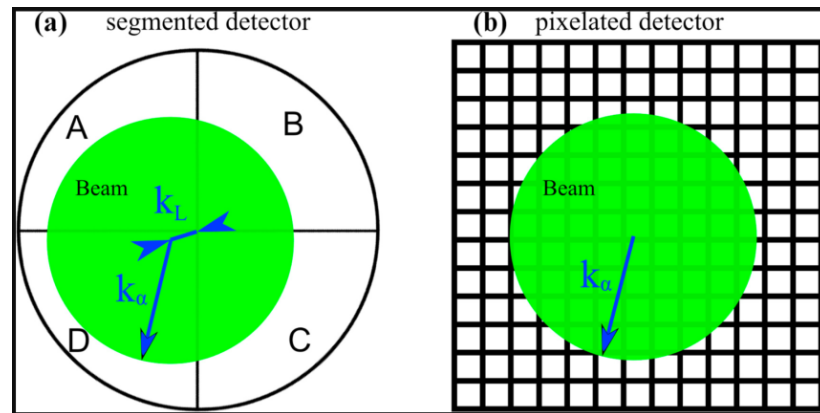
A path to 4D STEM

- The name “4D-STEM” refers to recording 2D images of a converged electron probe, over a 2D grid of probe positions
- STEM detectors - single value per STEM probe position
- TEM cameras – limited readout speeds and dynamic range



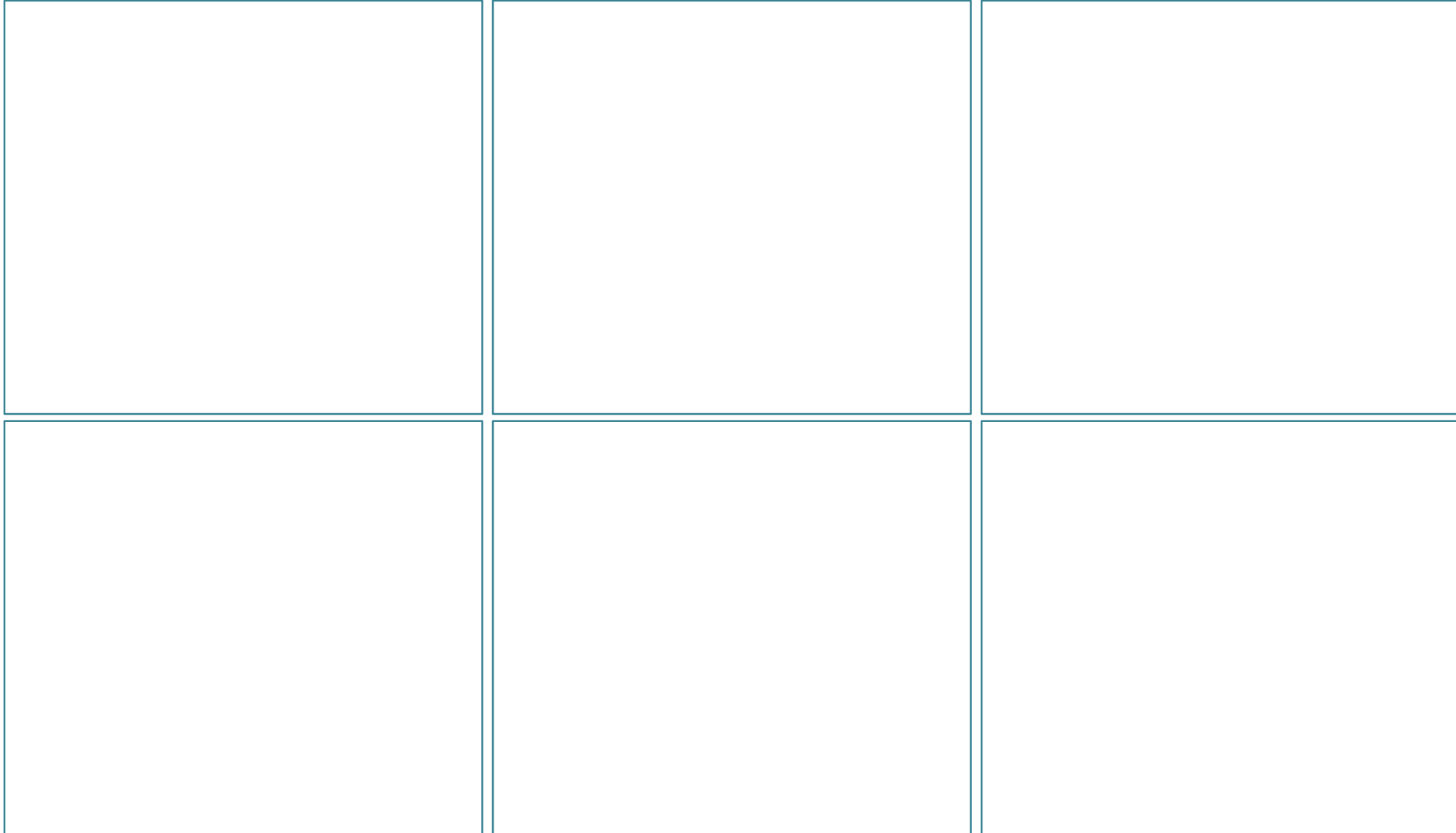
A path to 4D STEM

- The name “4D-STEM” refers to recording 2D images of a converged electron probe, over a 2D grid of probe positions
- STEM detectors - single value per STEM probe position
- TEM cameras – limited readout speeds and dynamic range
- Pixelated detectors



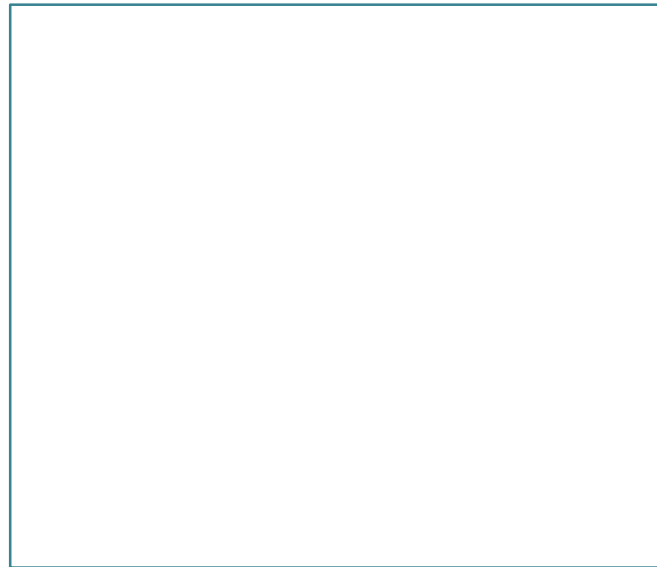
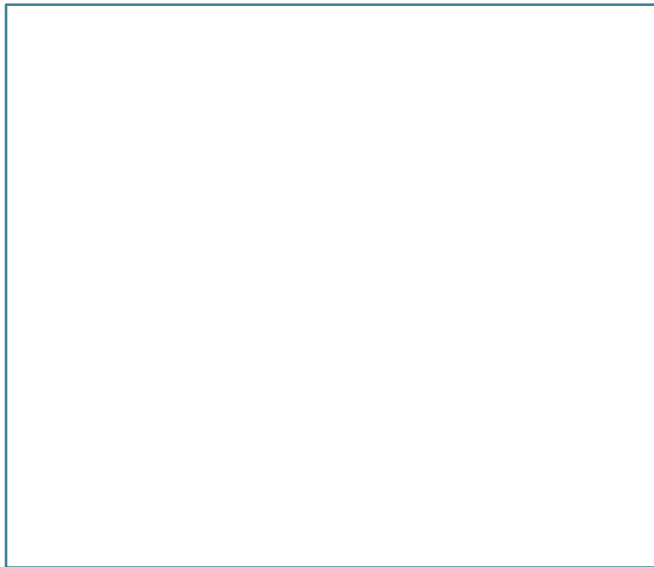
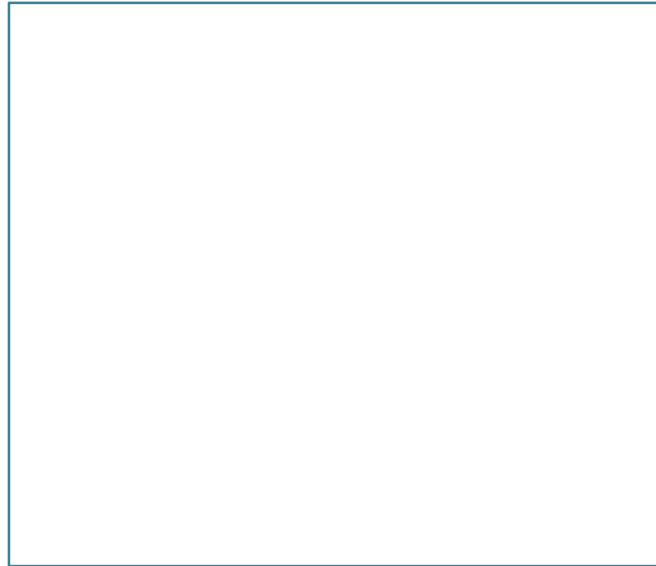
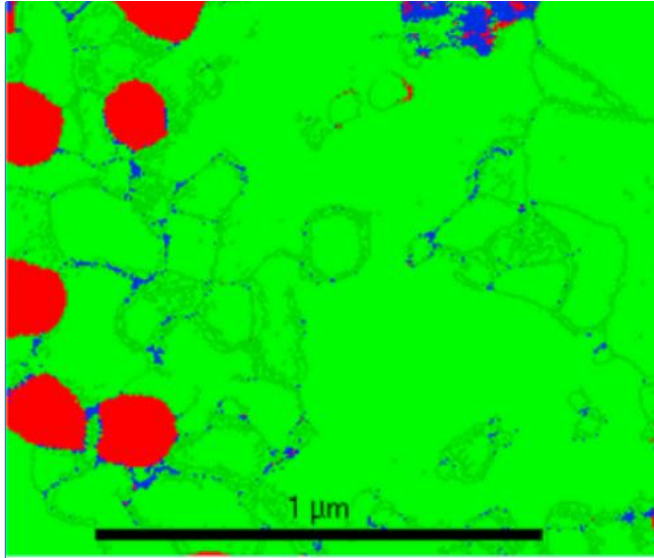
4D-STEM

- The name “4D-STEM” refers to recording 2D images of a converged electron probe, over a 2D grid of probe positions



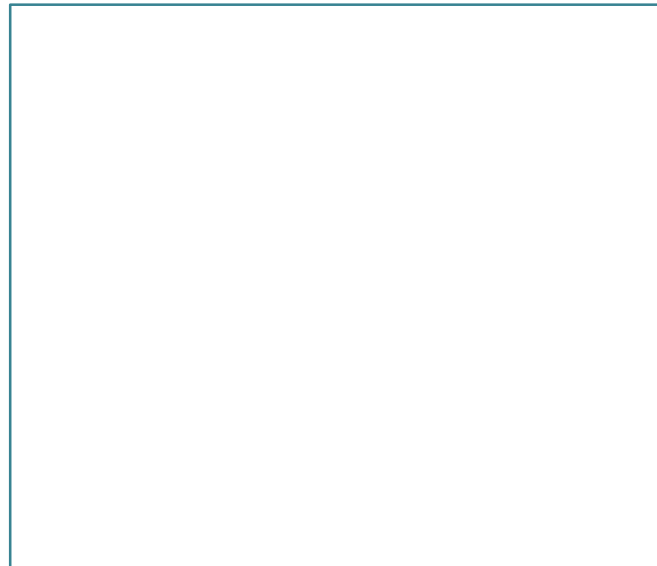
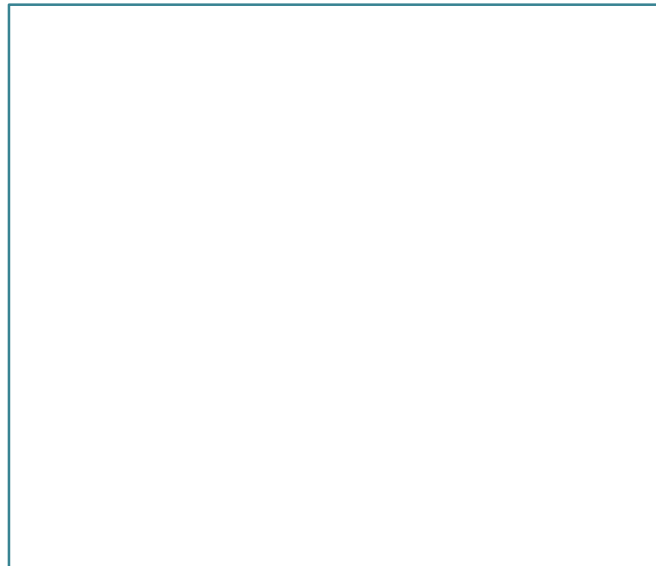
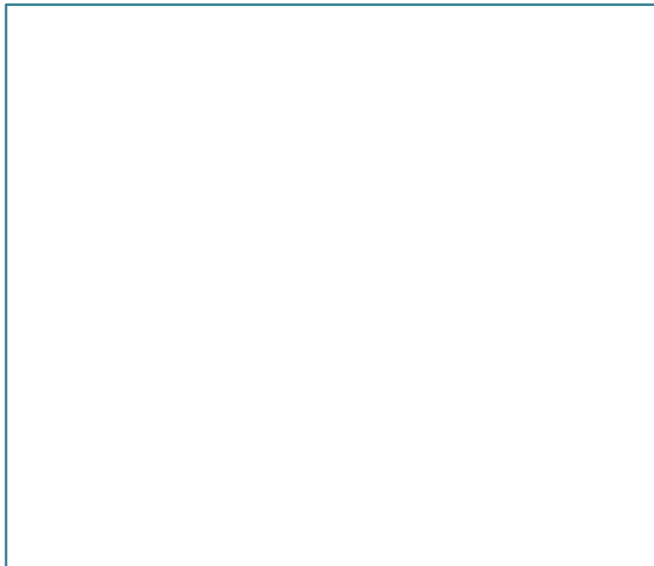
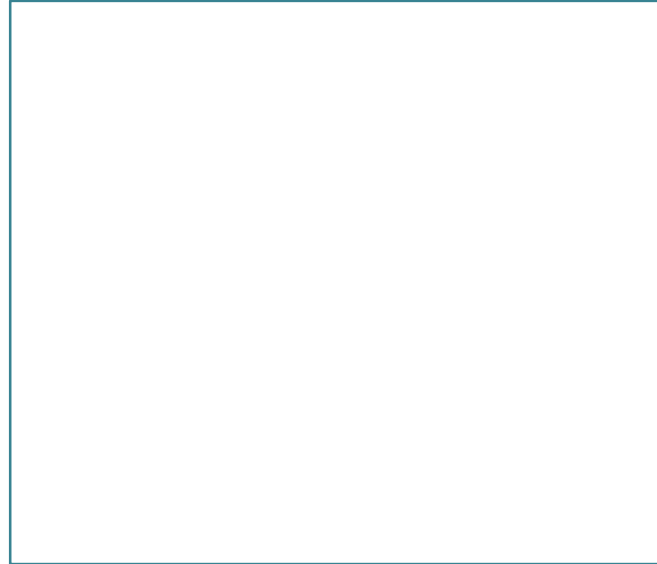
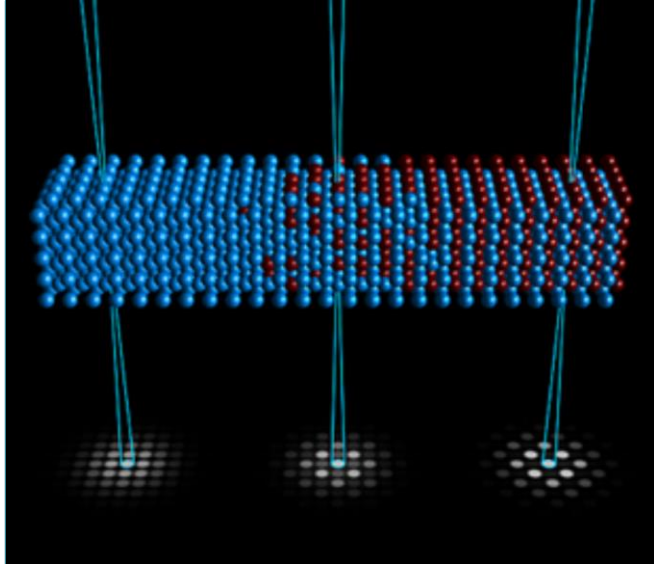
4D-STEM

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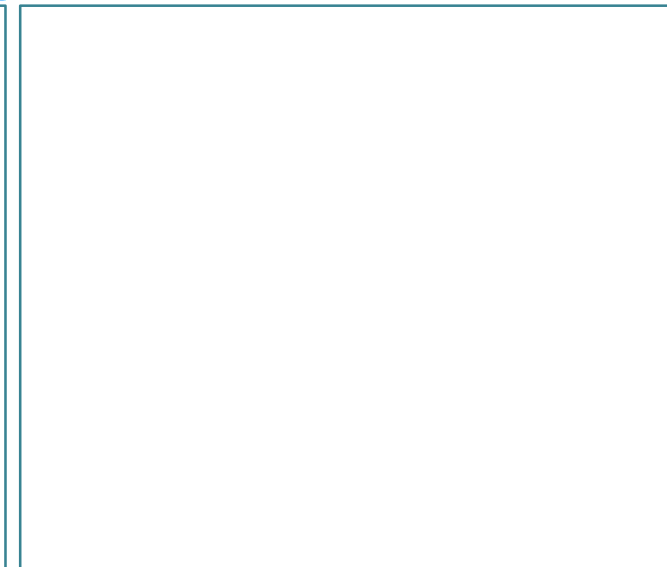
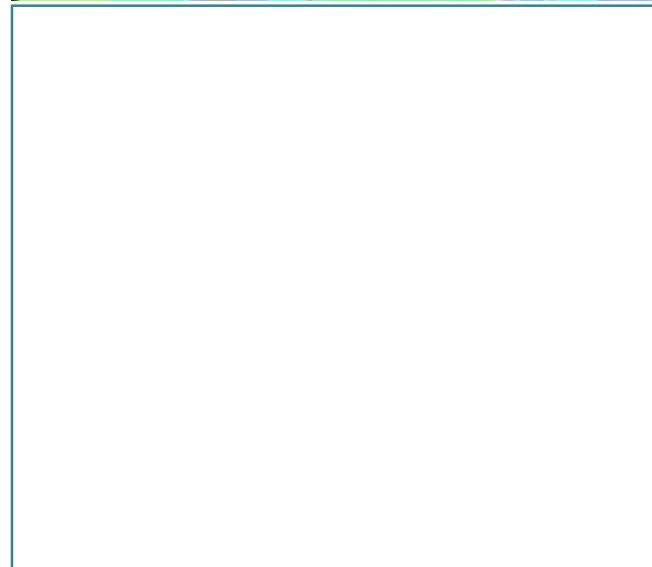
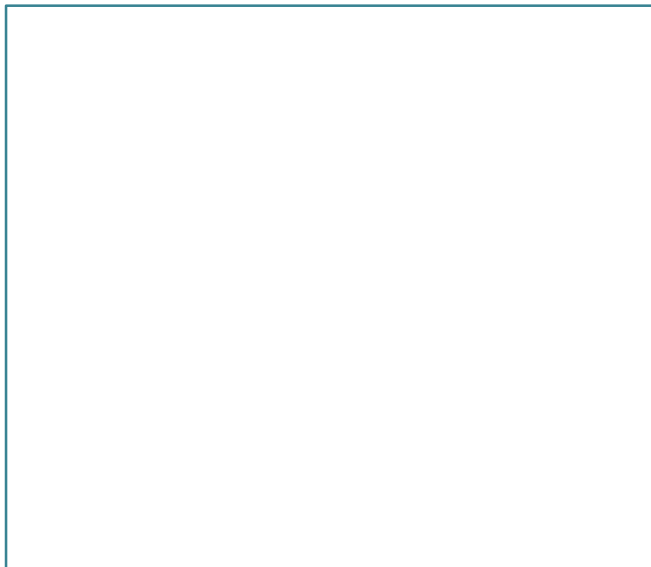
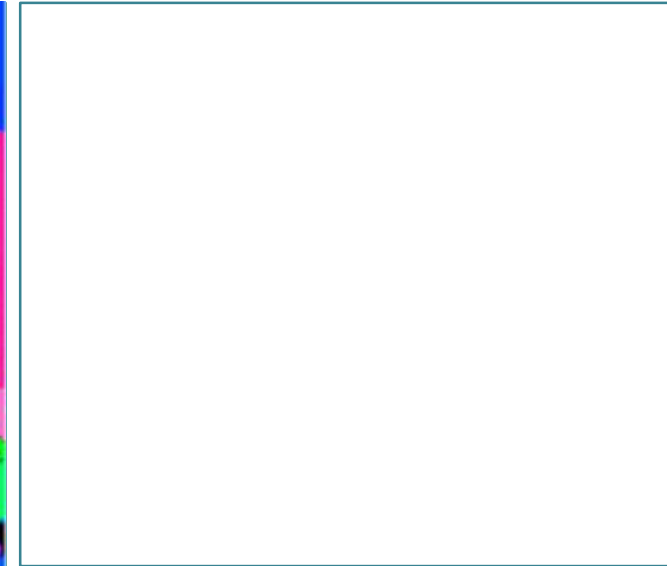
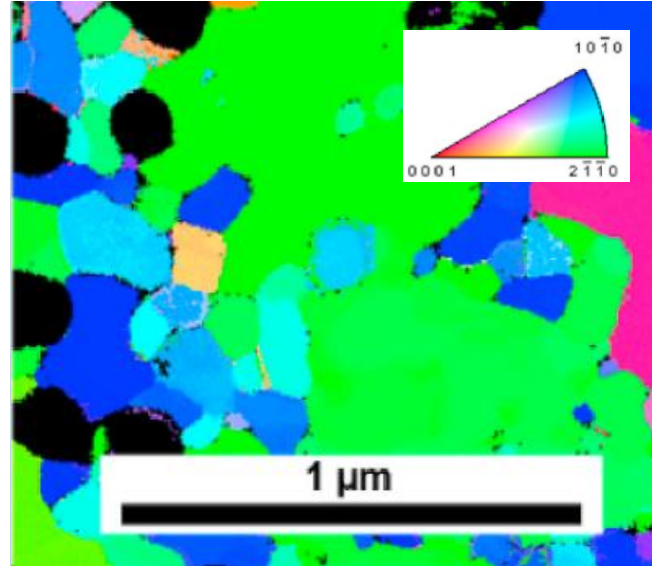
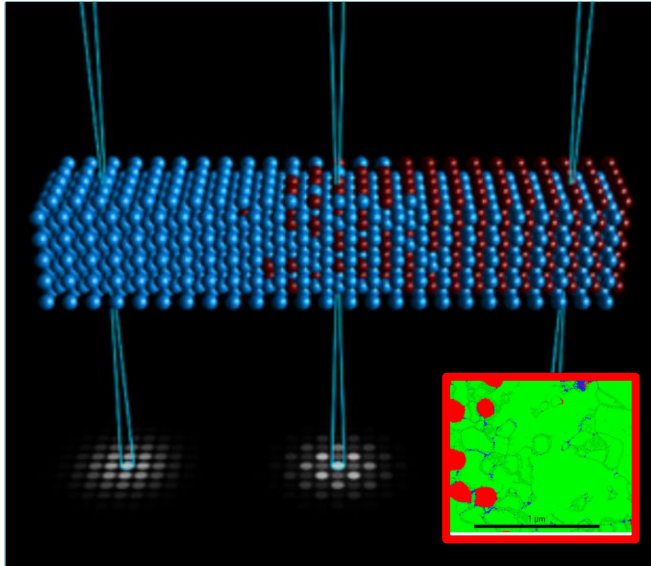
4D-STEM

- The name “4D-STEM” refers to recording 2D images of a converged electron probe, over a 2D grid of probe positions



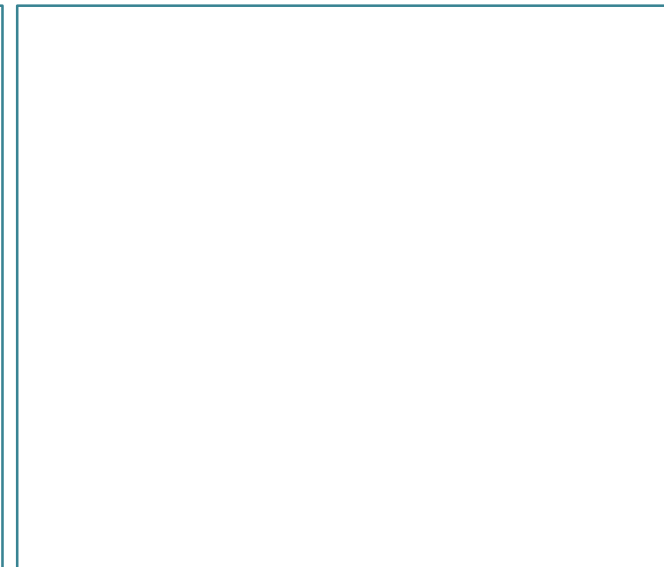
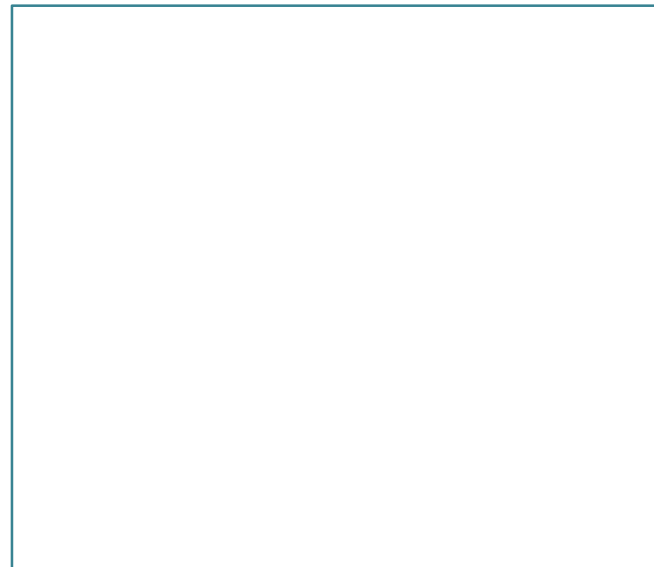
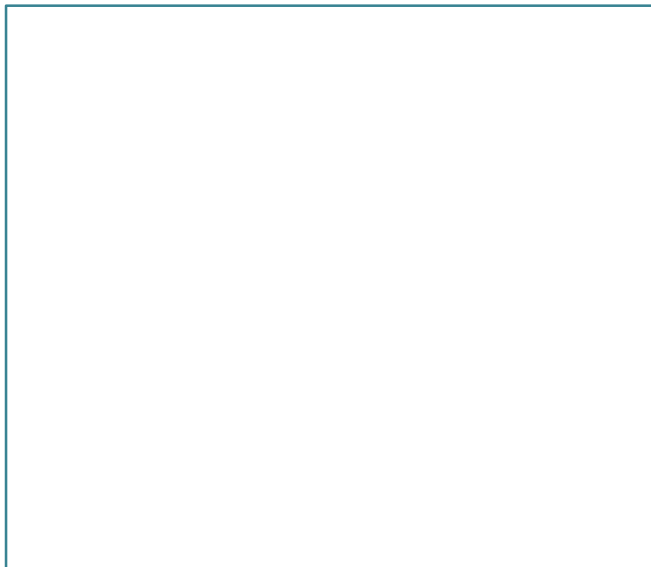
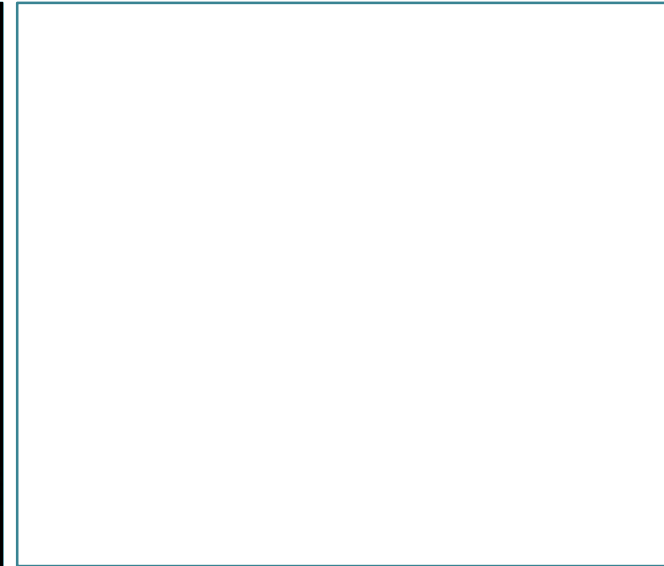
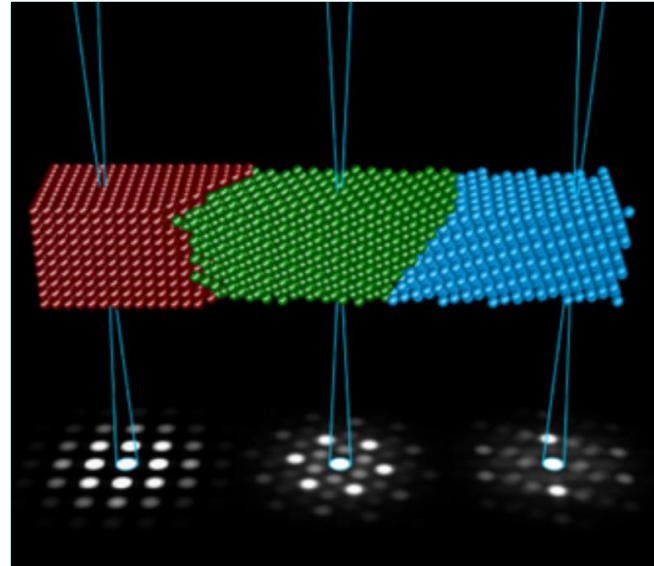
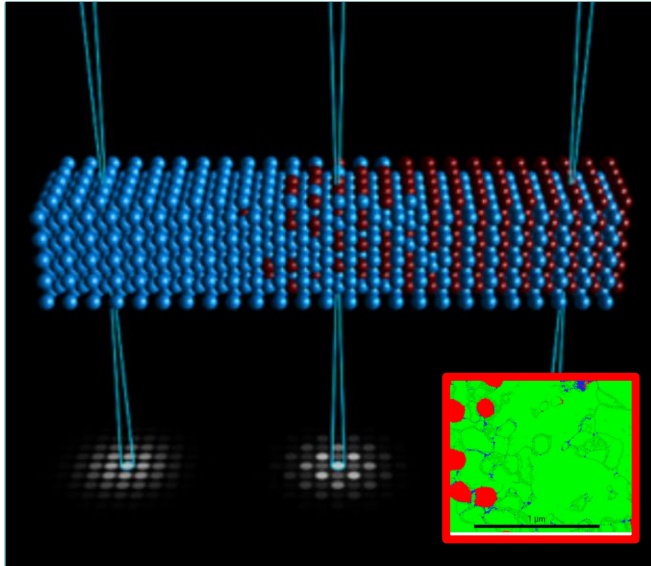
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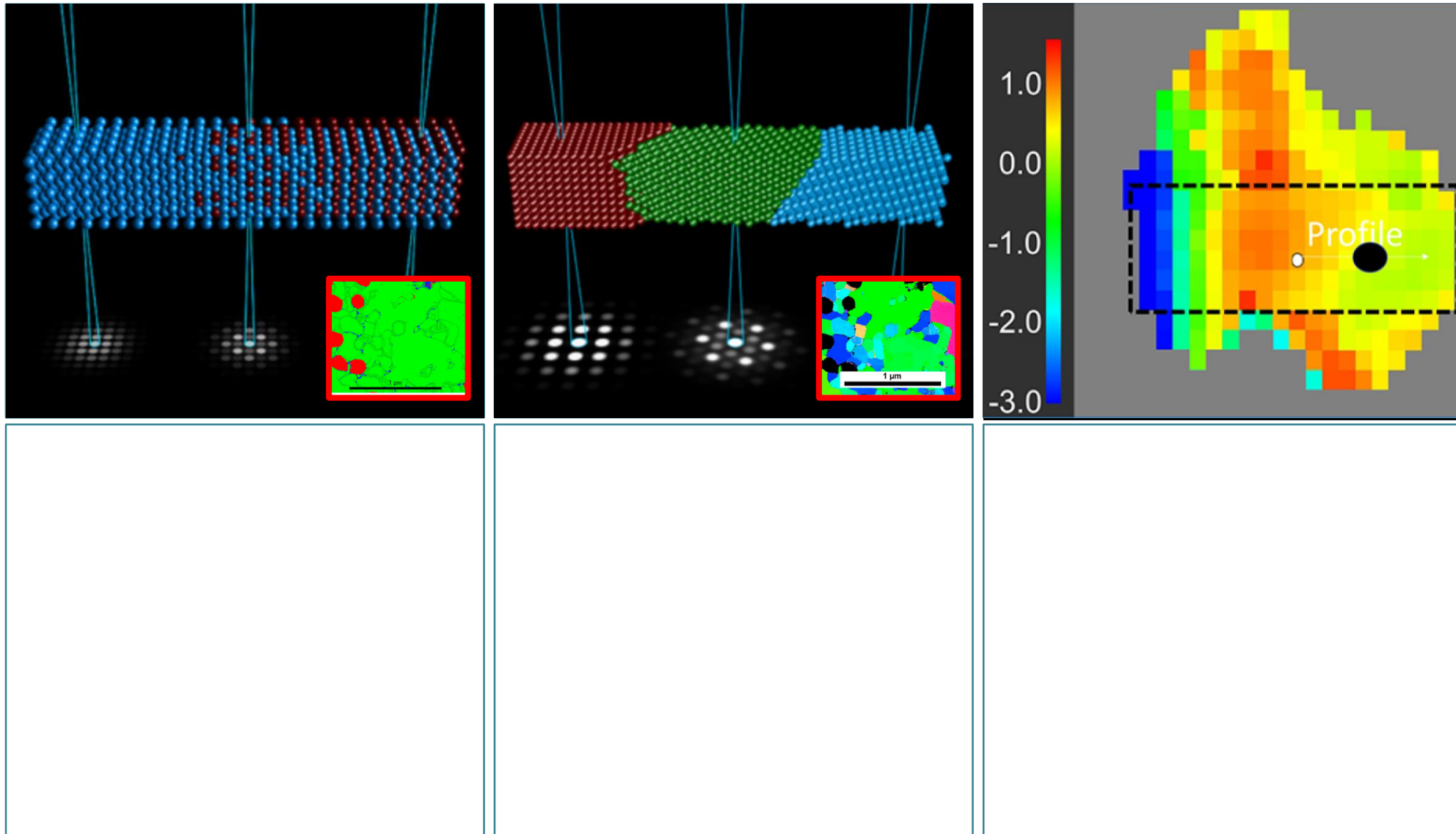
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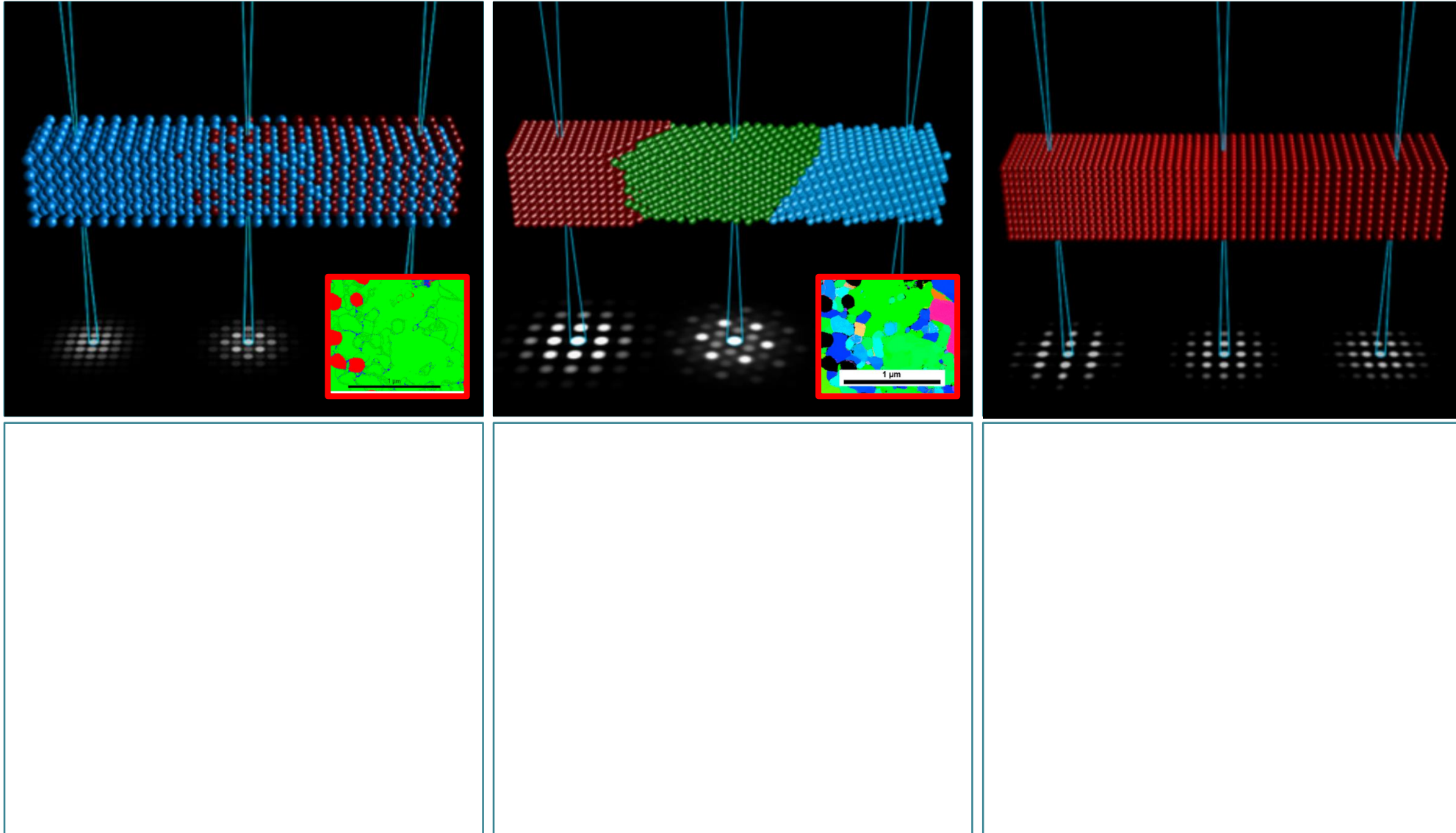
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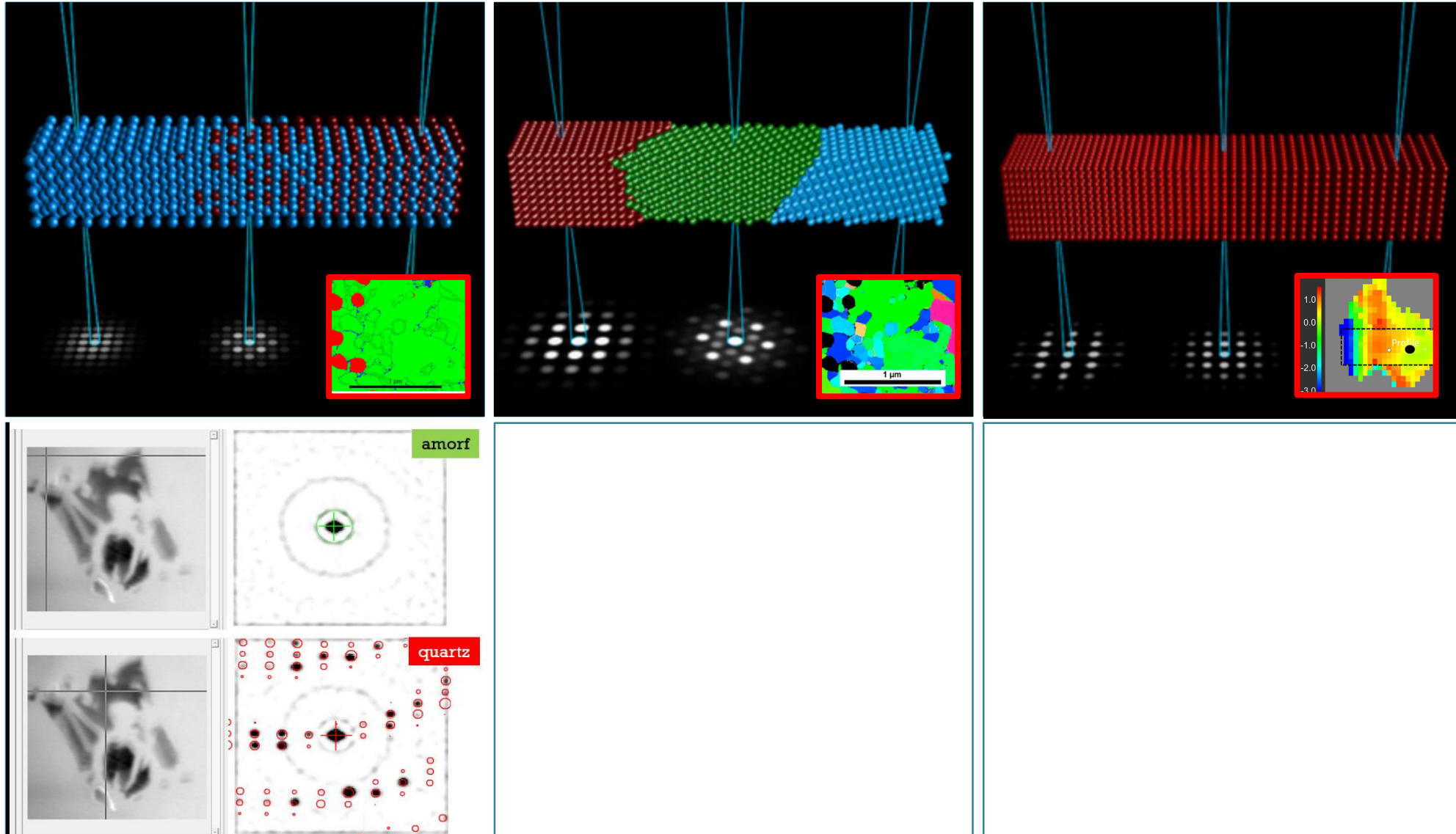
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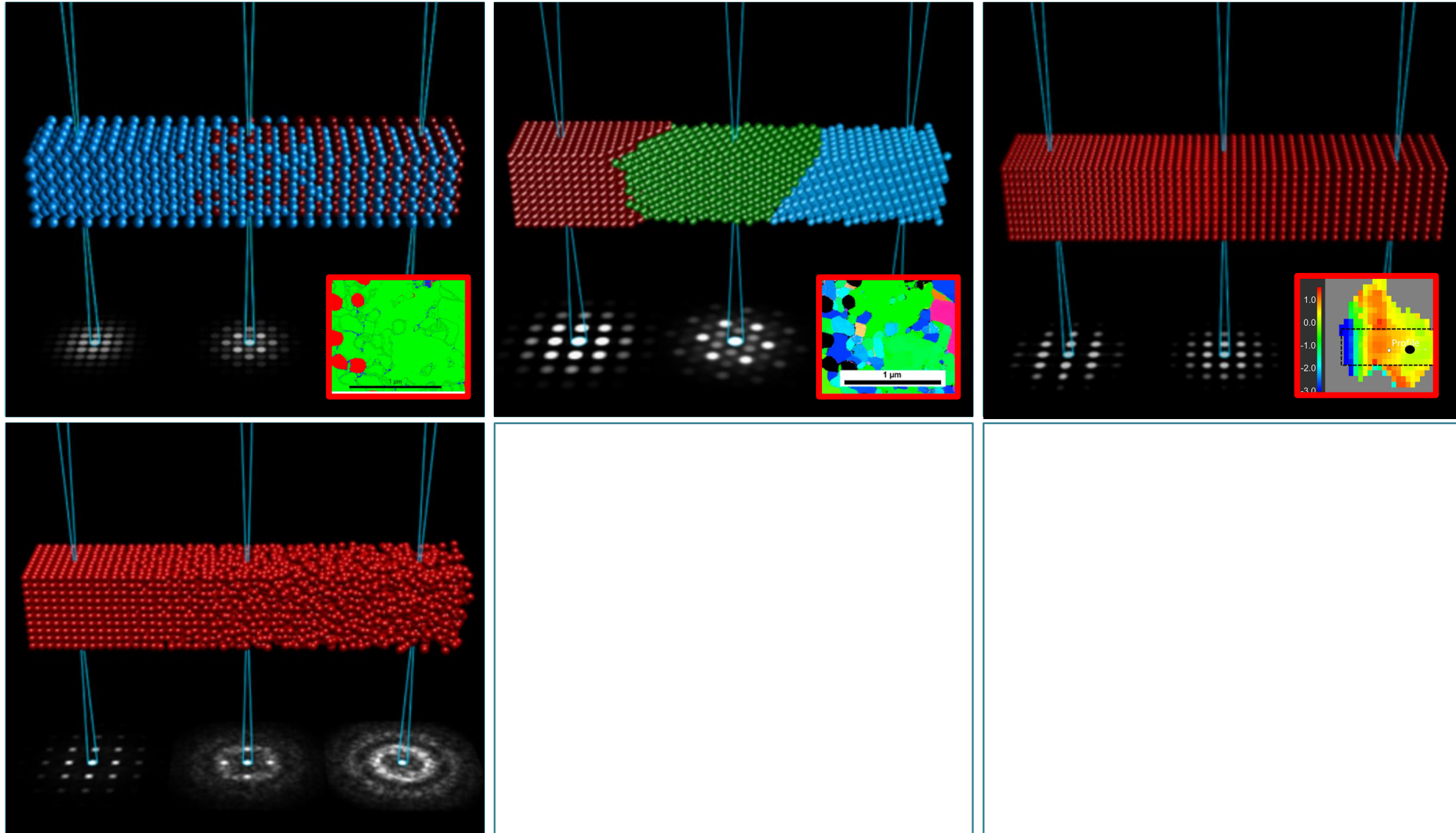
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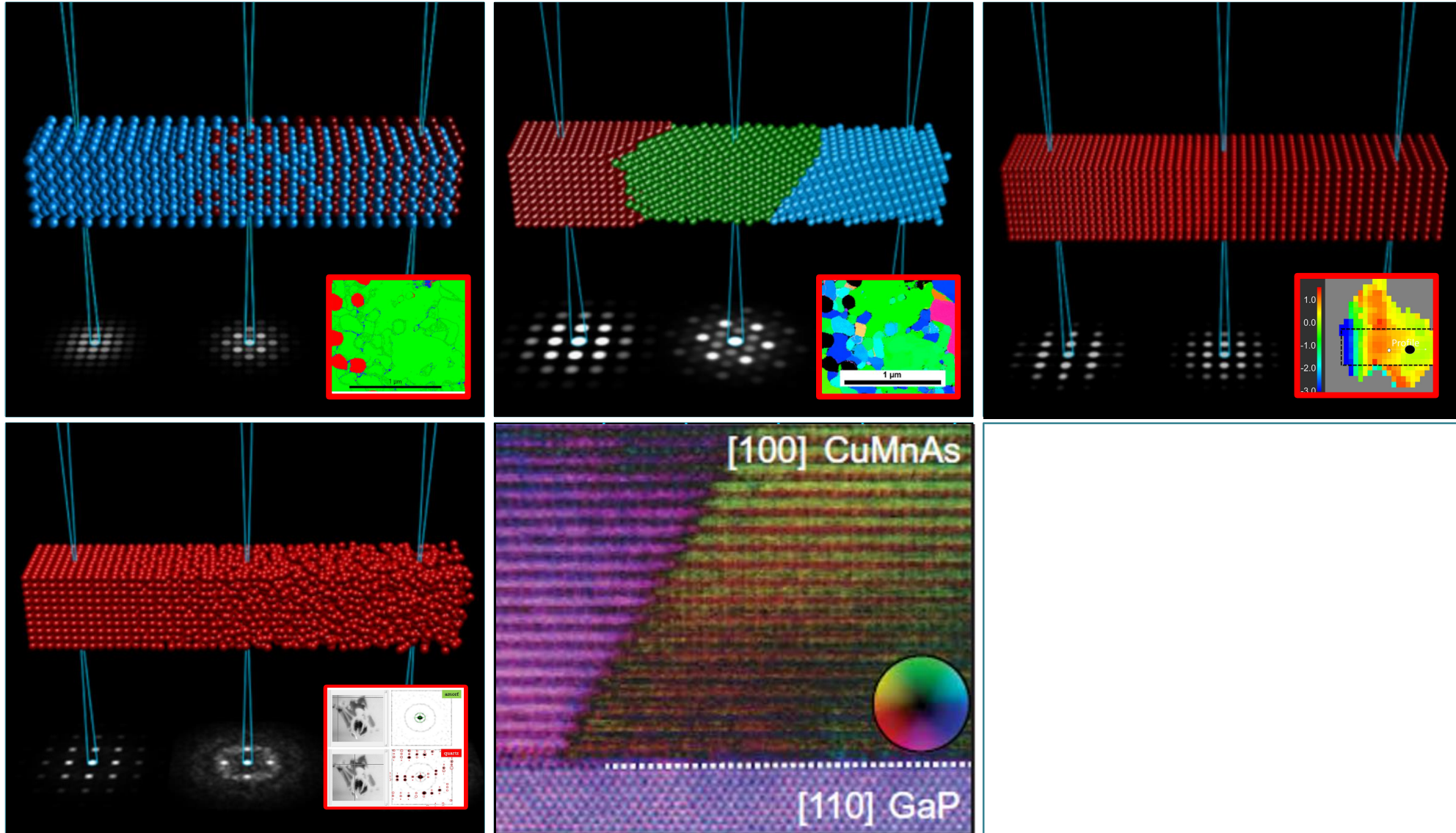
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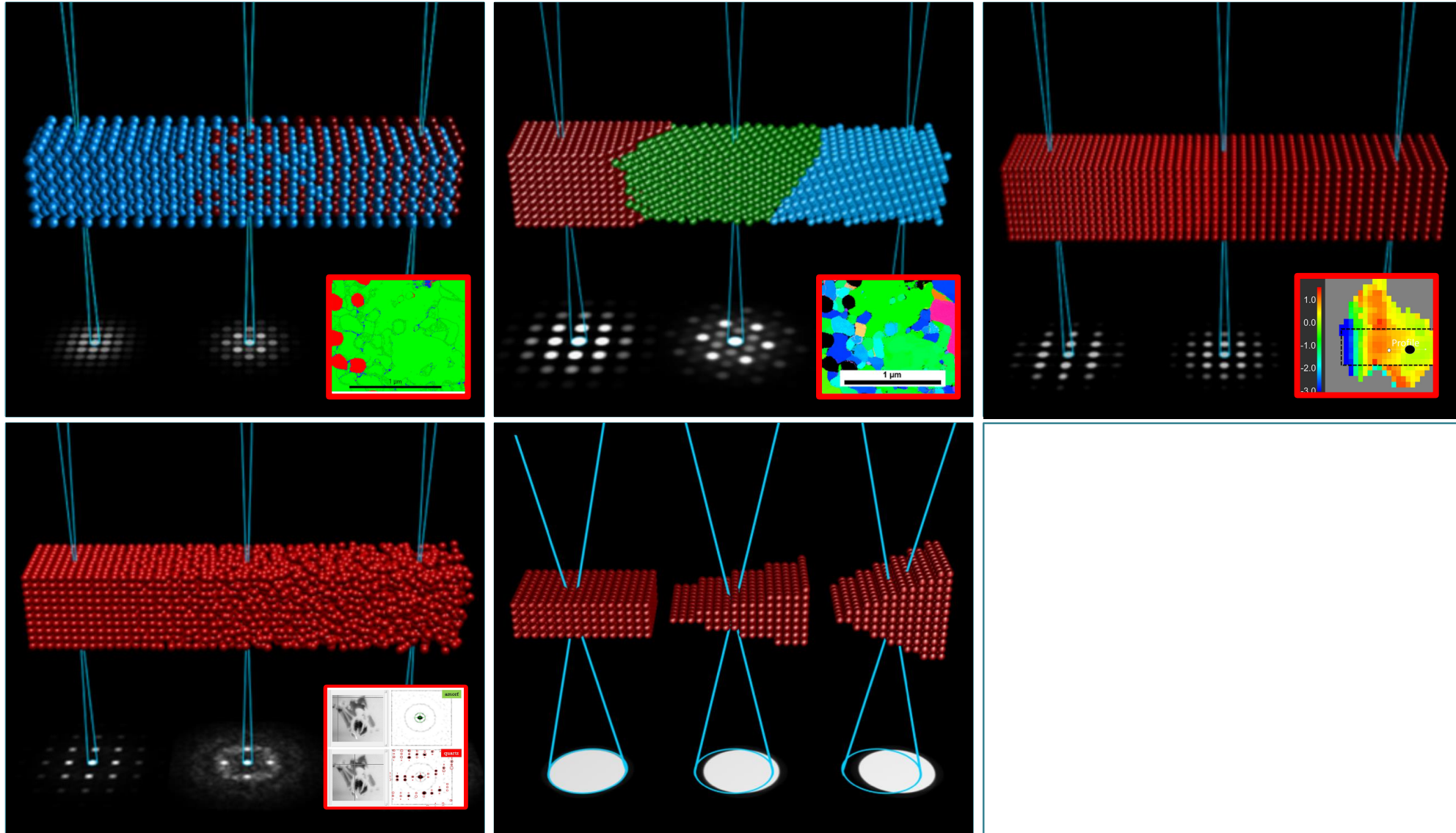
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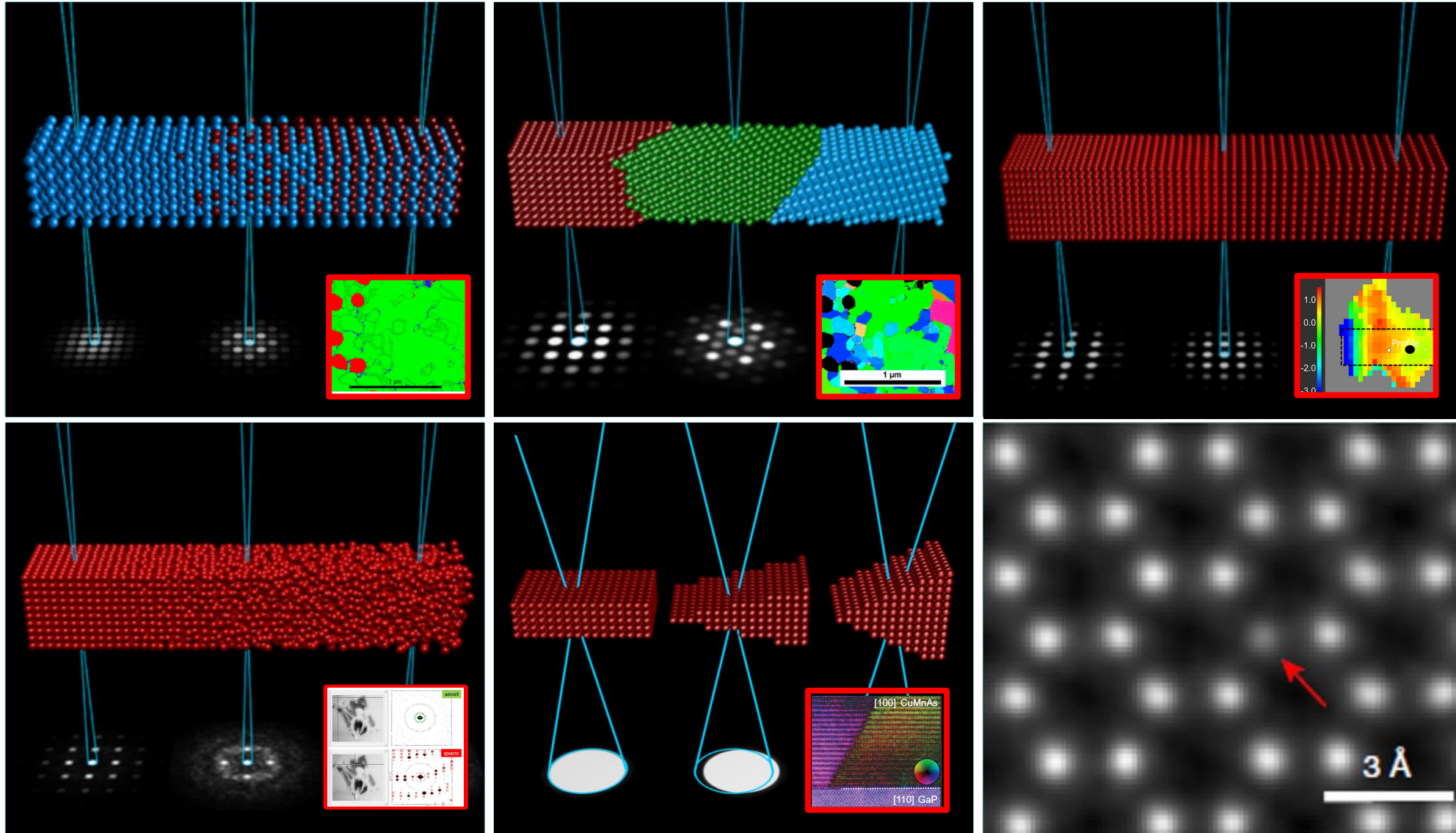
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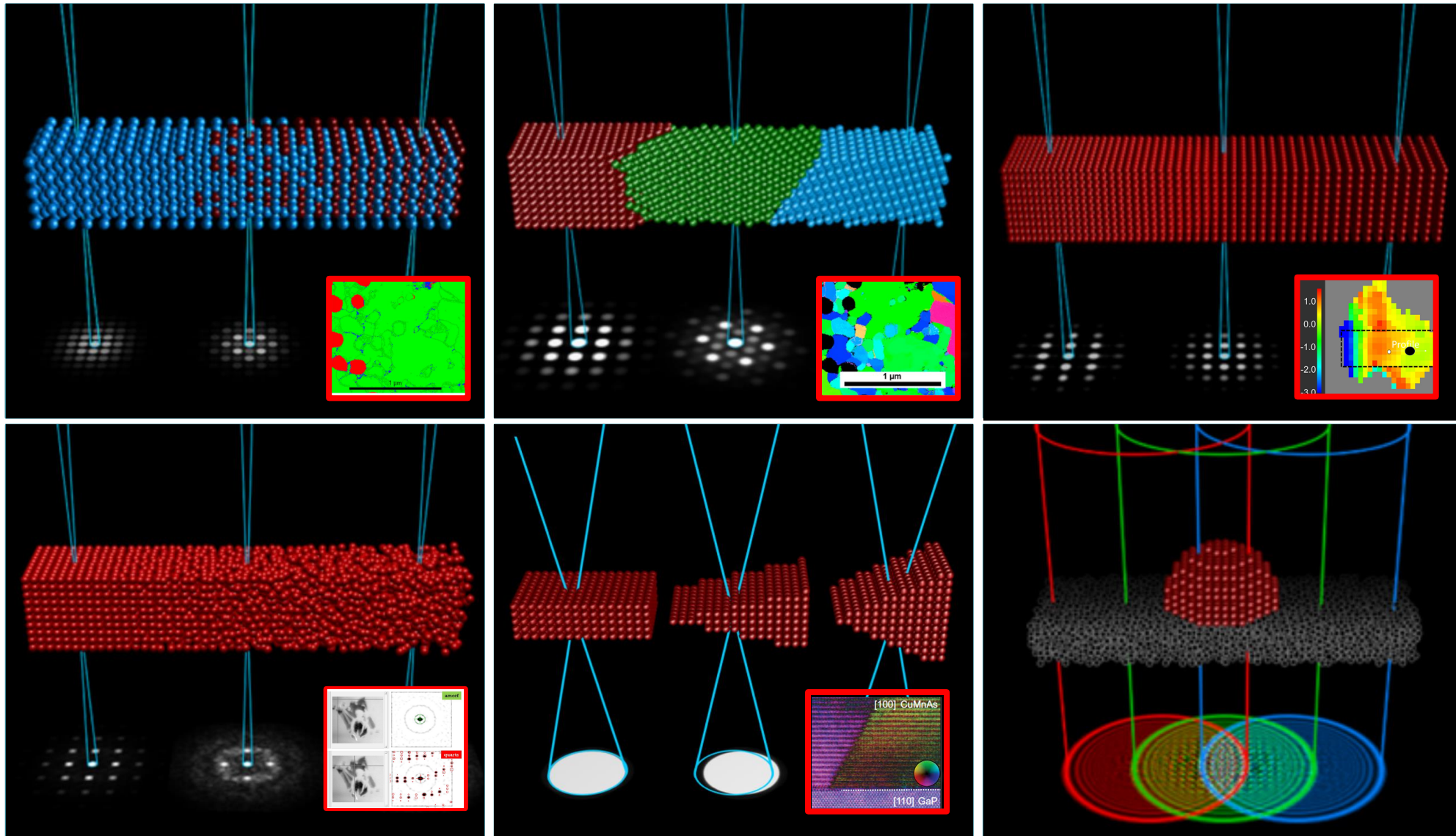
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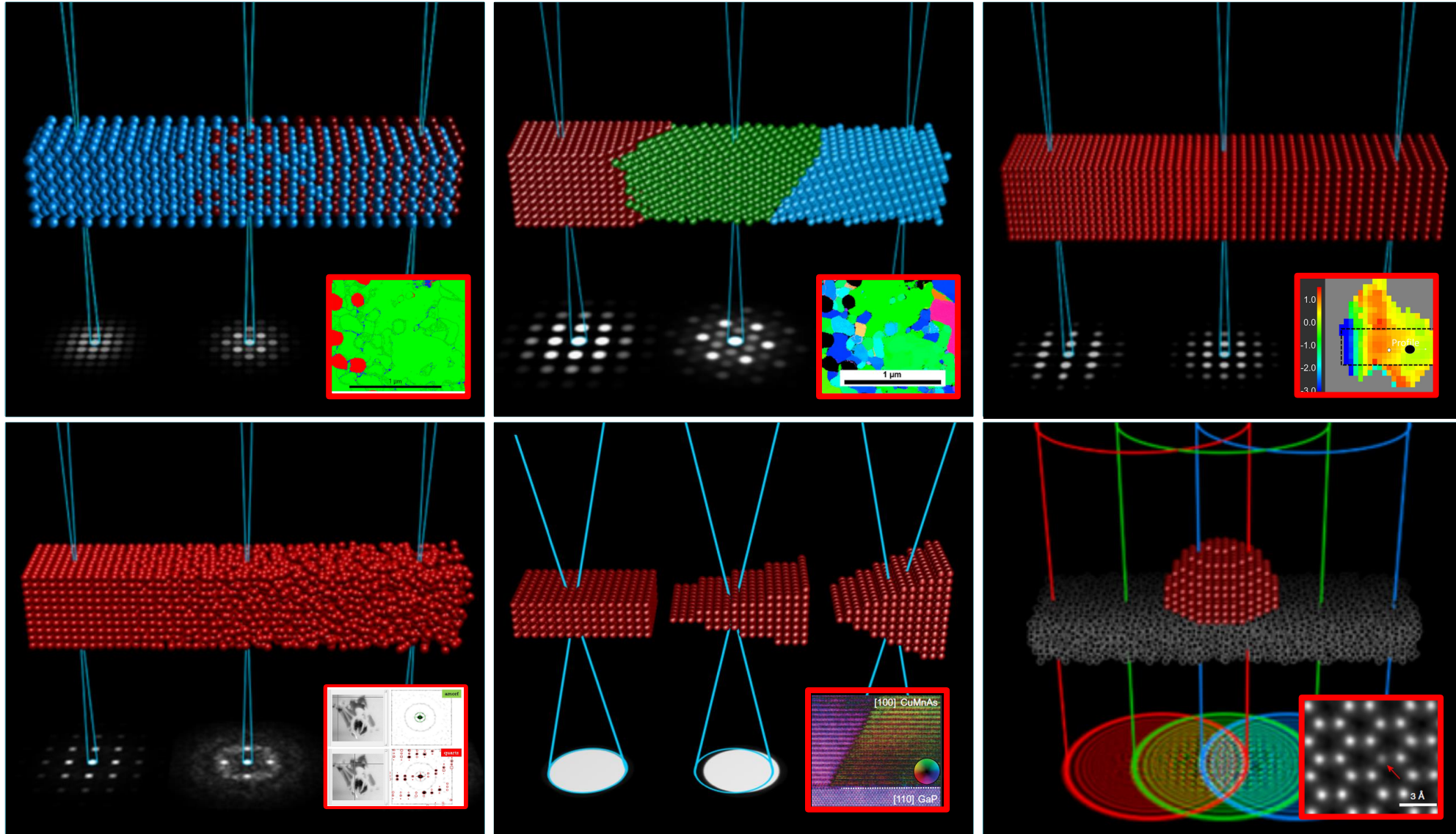
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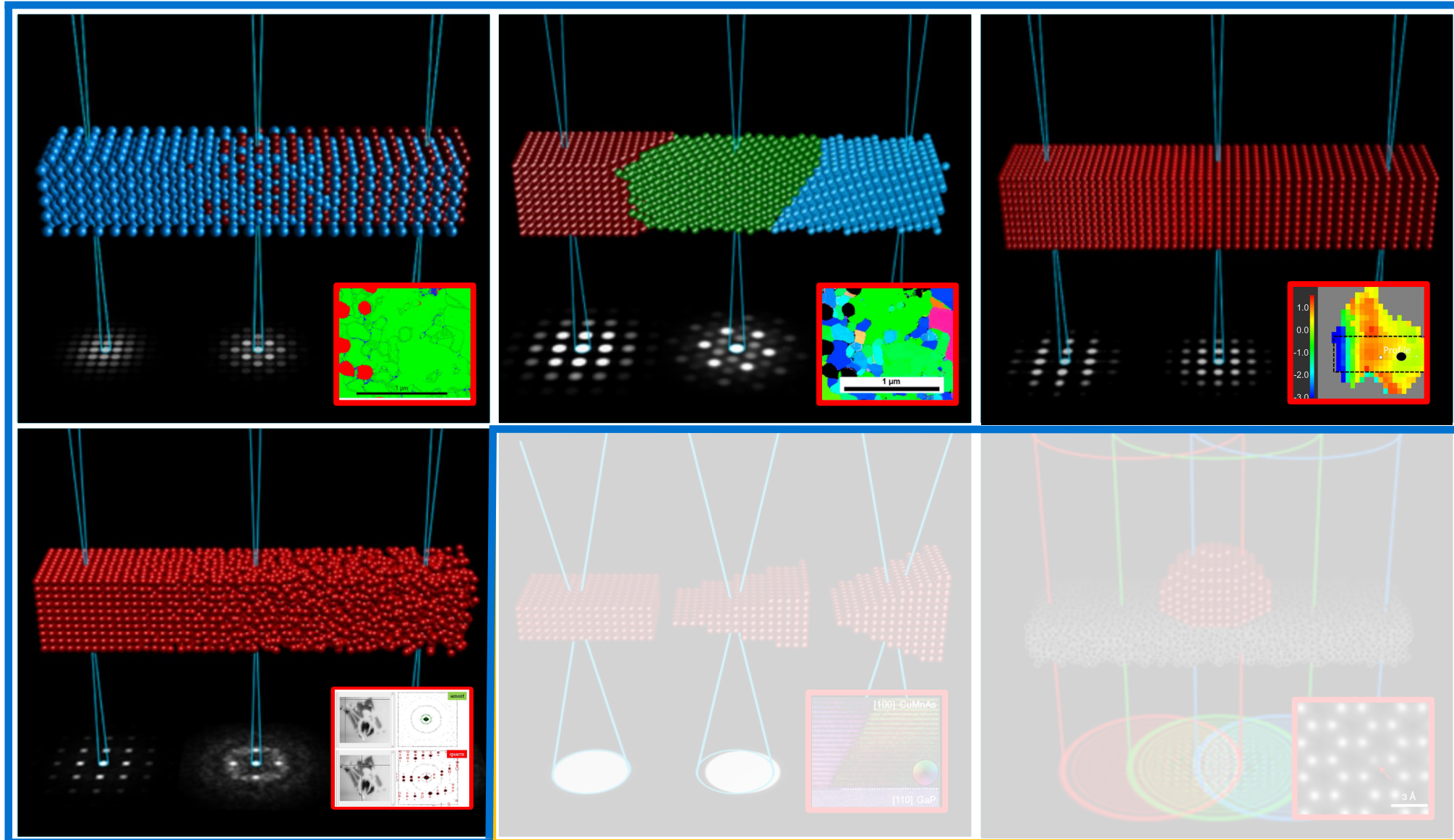
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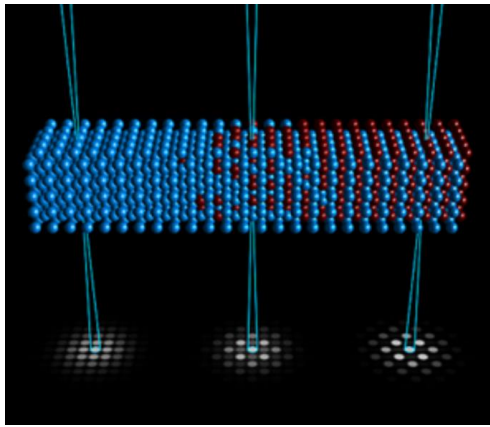
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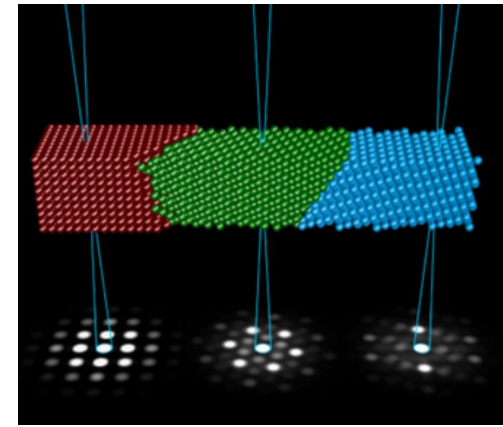


Possible
at FZU

AC-STEM
with
better
detector
needed

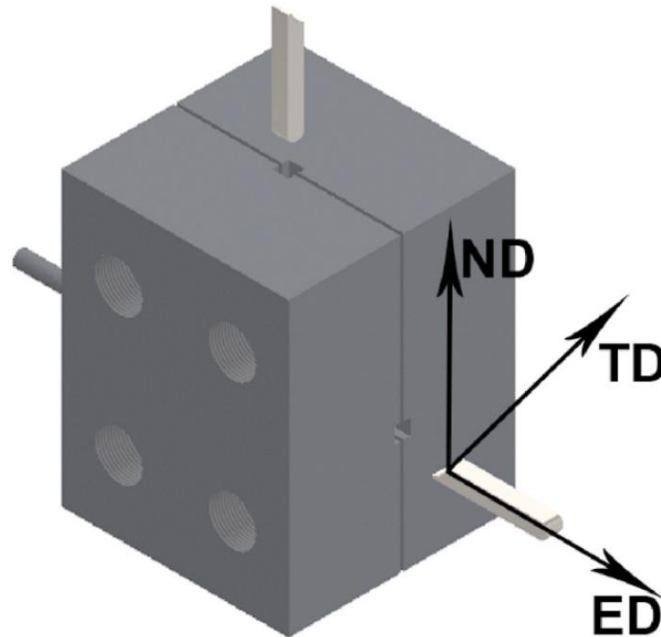


Structure and orientation mapping LEM result example

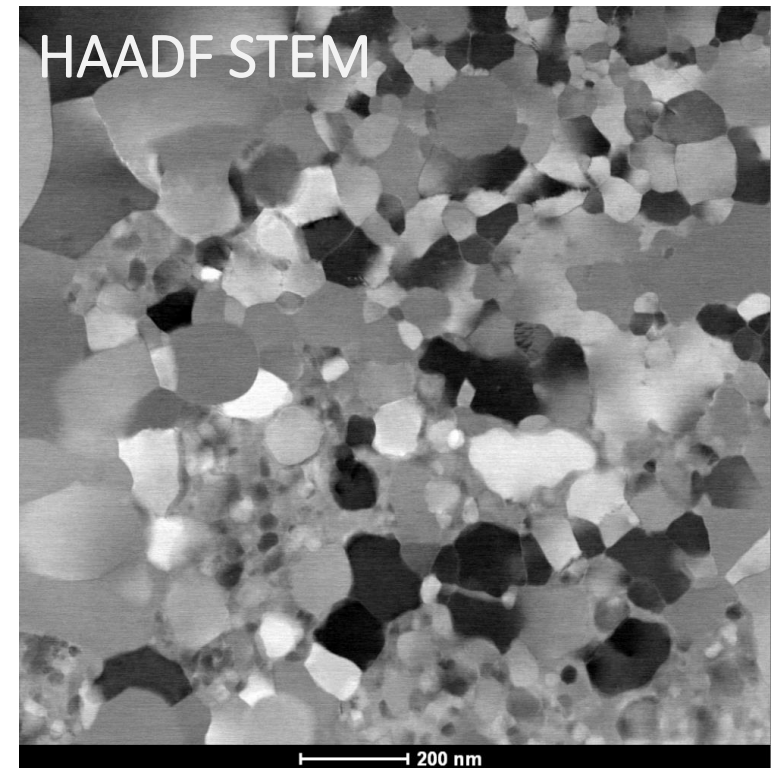


Cooperation with J. Pinc research

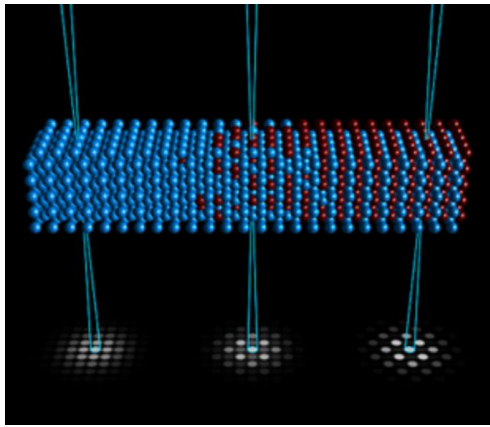
- Zn-0.8Mg-0.2Sr (wt. %) alloy processed by ECAP
- Material for biodegradable implants
- 4D STEM ideal for microstructure study



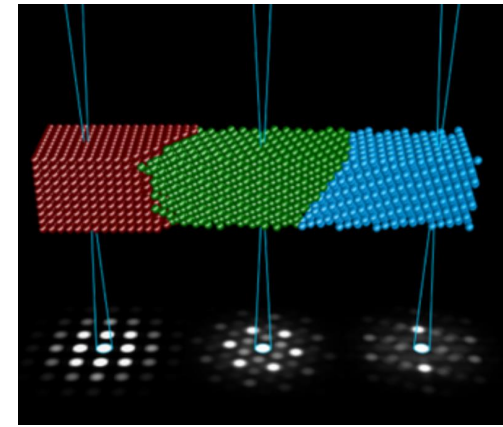
Equal channel angular pressing (ECAP)



[Materials Science & Engineering A 824 (2021) 141809]



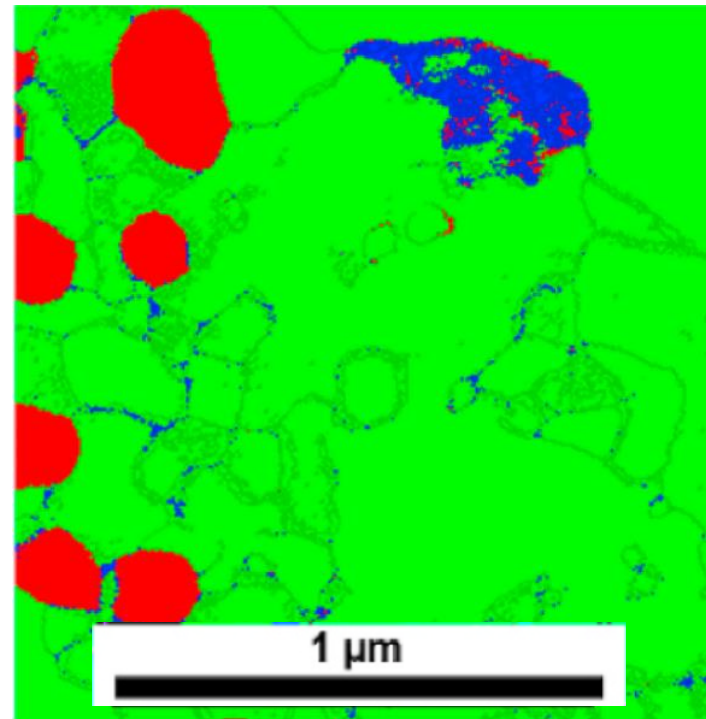
Structure and orientation mapping LEM result example



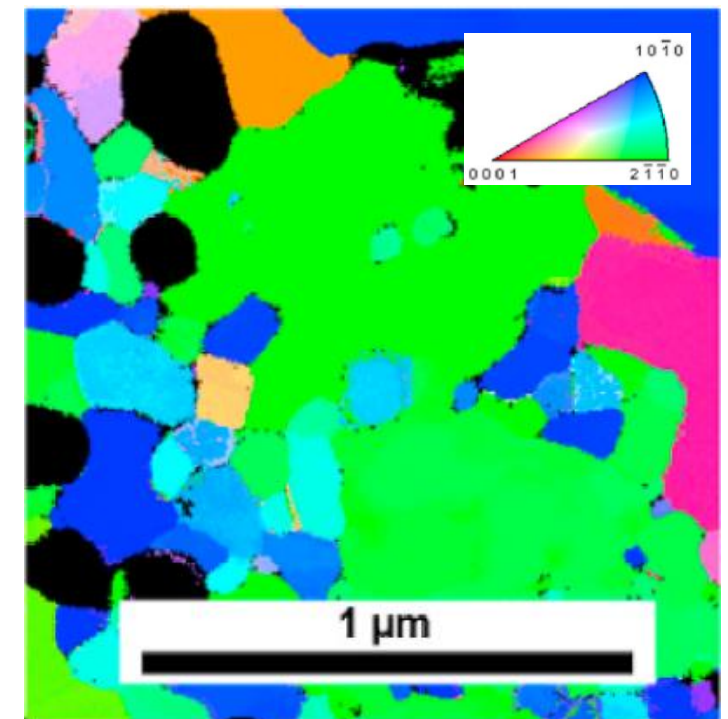
Cooperation with J. Pinc research

- Zn-0.8Mg-0.2Sr (wt. %) alloy processed by ECAP
- Material for biodegradable implants
- 4D STEM ideal for microstructure study
- Intermetallic phases Mg_2Zn_{11} (blue), $SrZn_{13}$ (red)

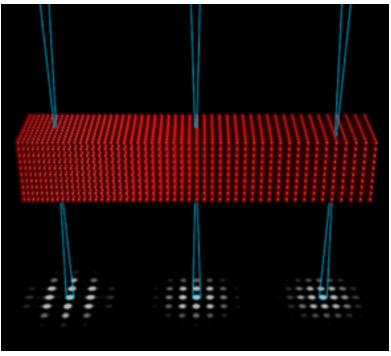
Phase map



Orientation map

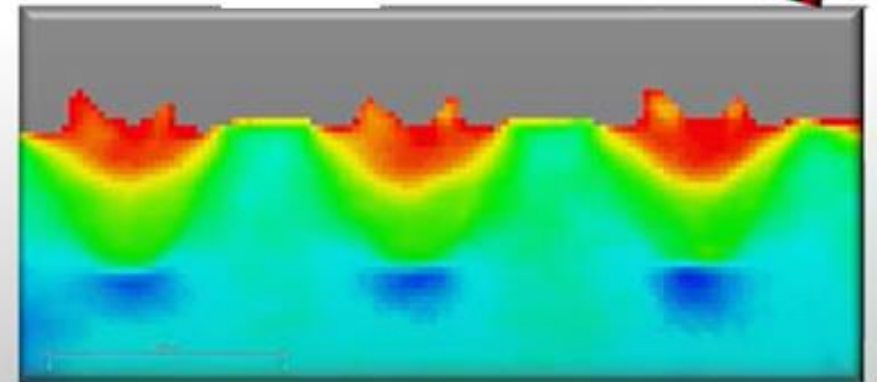
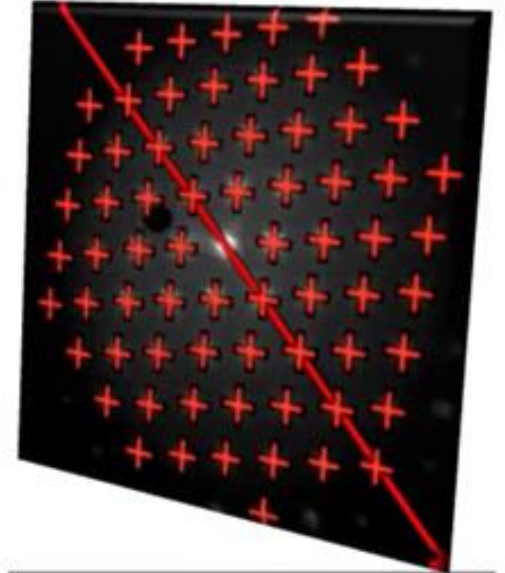


Strain mapping



- Functional materials possess a large degree of local variation in lattice parameter
- Strain measurement using NBD involves measurement of shift in individual diffraction spots
- Trade-off between resolution in real space and reciprocal space

Strain Calculation:
PED series patterns are matched against the **reference pattern**.



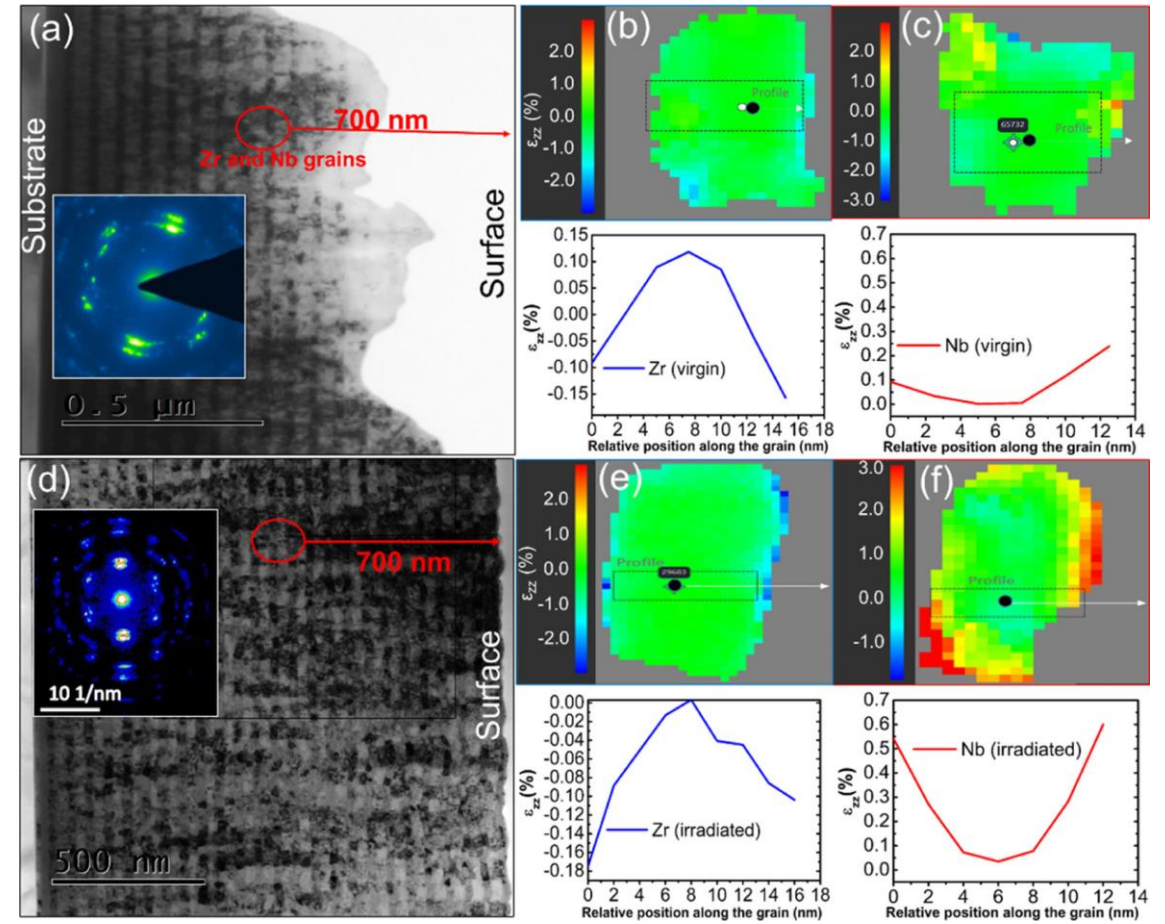
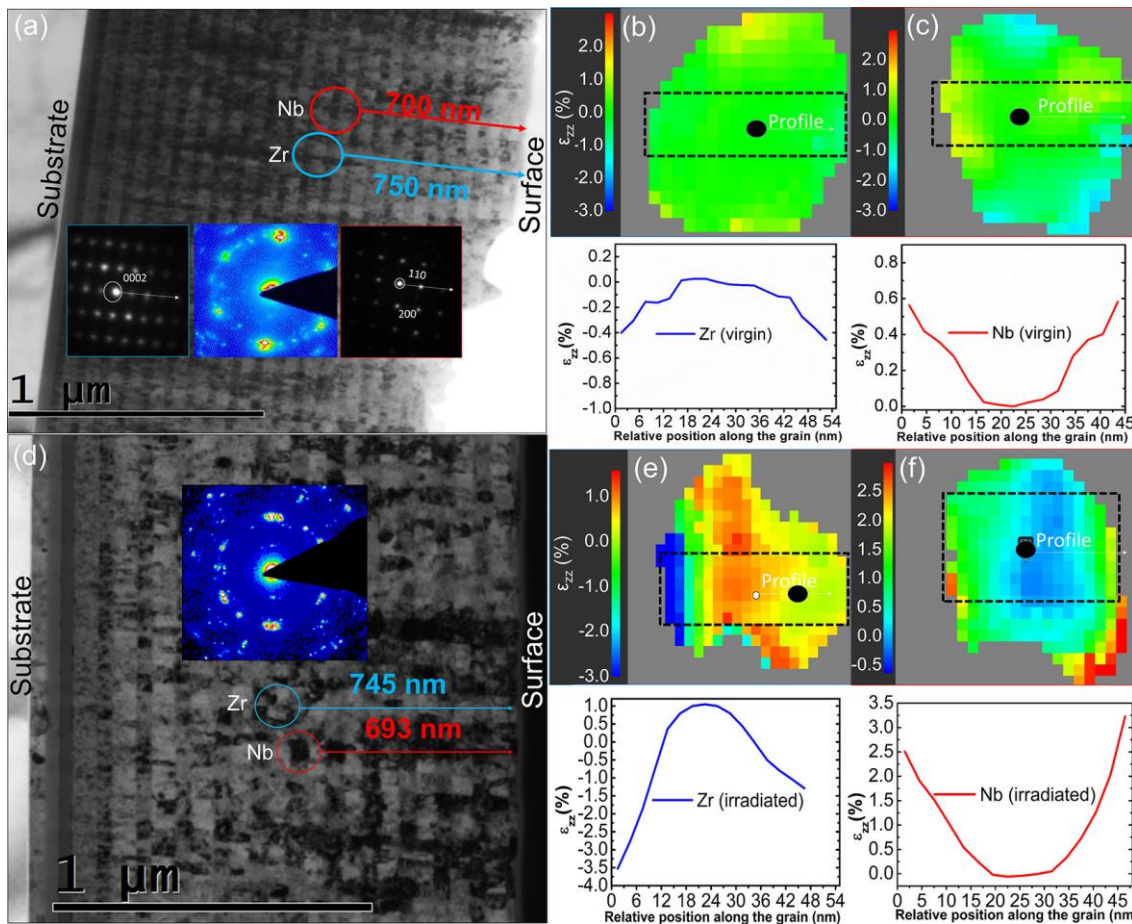
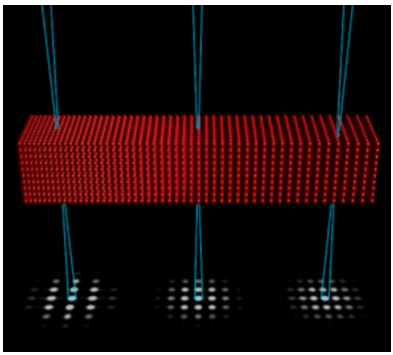
Strain Mapping

Single crystal grain area calculated

Strain mapping – LEM result example

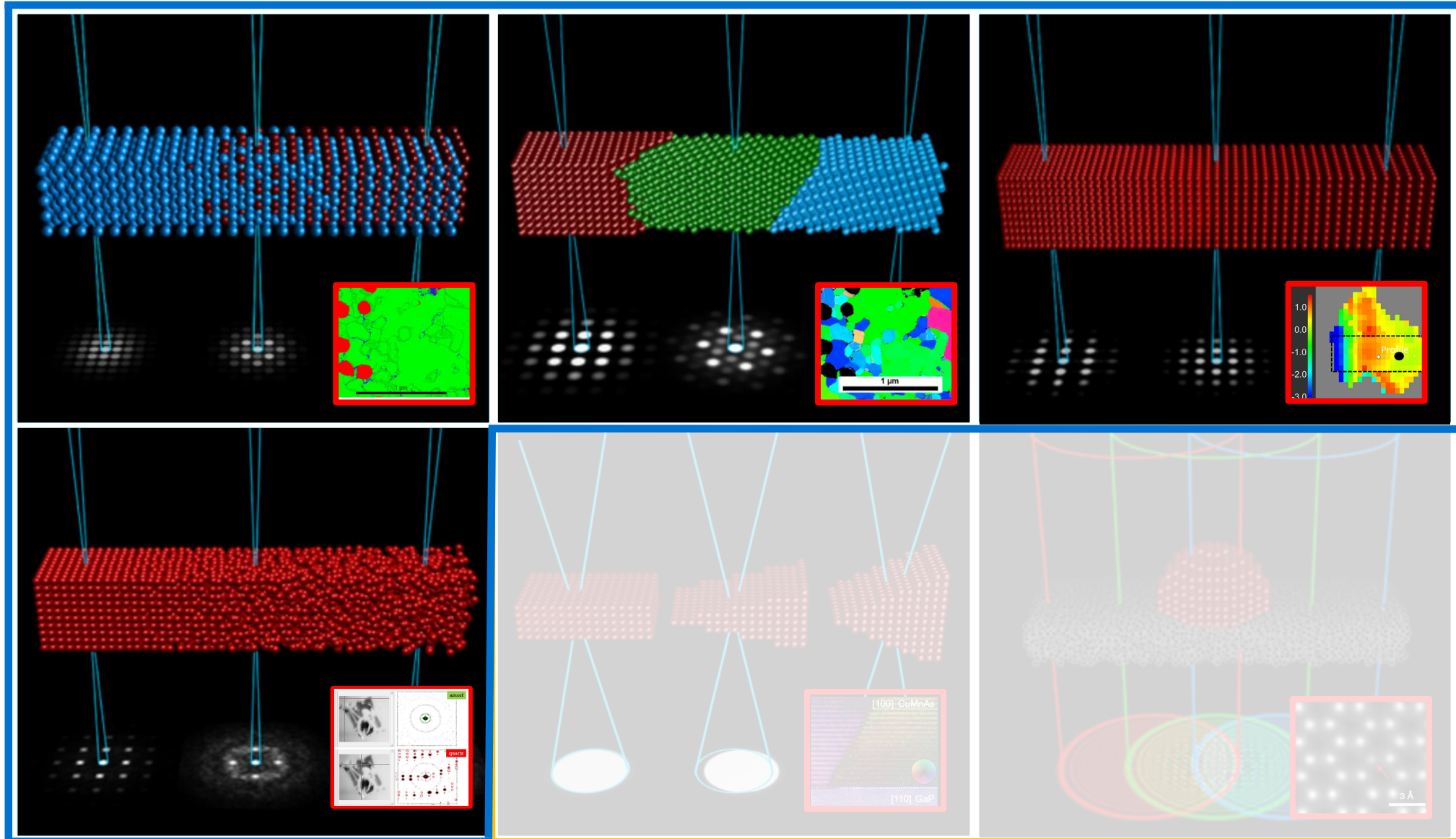
Cooperation with CTU in Prague

- Zr/Nb nanolayered alloys - promising materials for nuclear applications thanks to their resistance to radiation damage
- Zr/Nb55 with thicknesses around Zr = 24 nm and Nb = 31 nm is the most promising multilayer system with the high radiation damage resistance and minimum swelling for nuclear applications



4D-STEM

- The name “4D-STEM” refers to recording 2D images of a converged electron probe, over a 2D grid of probe positions

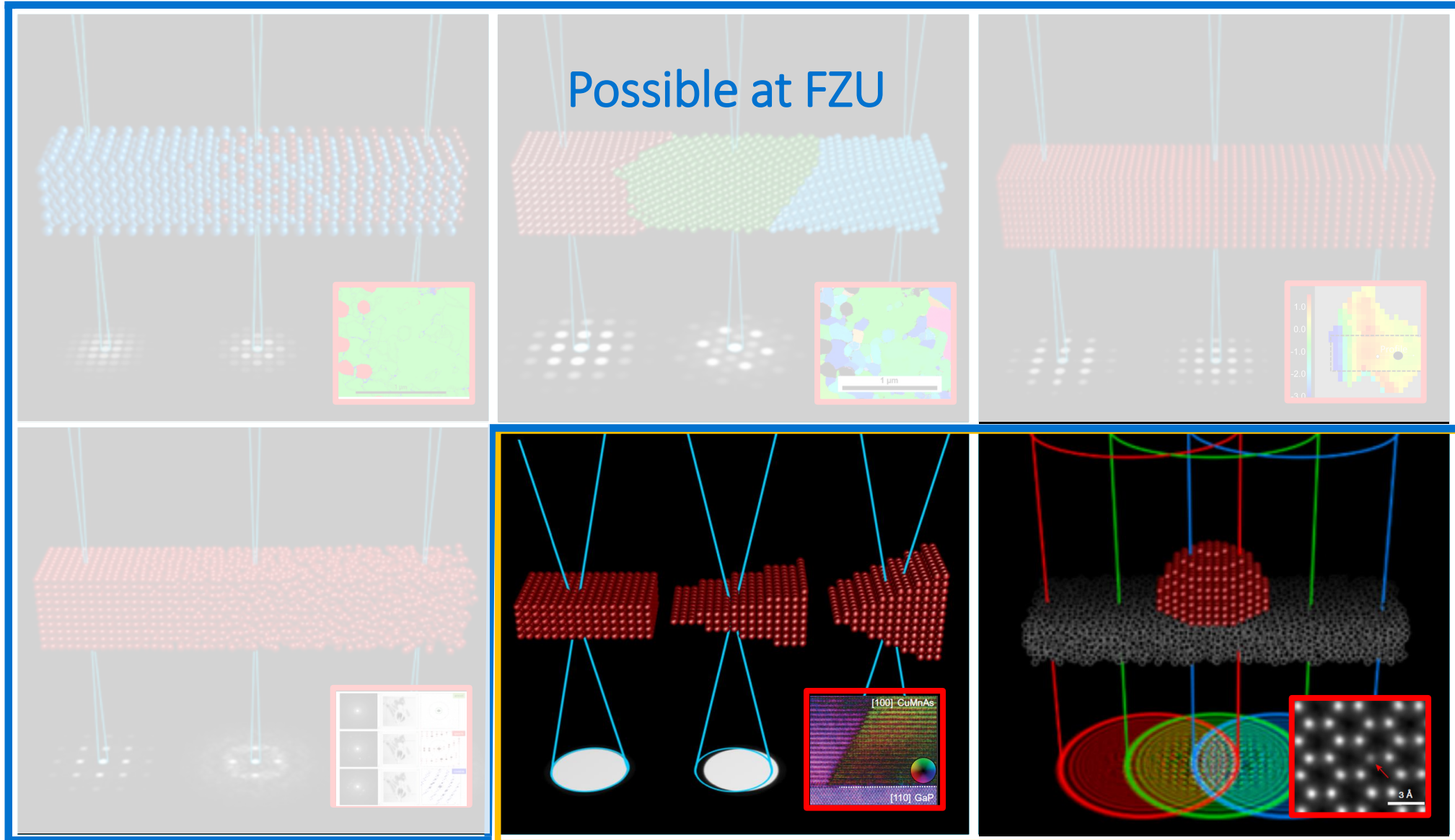


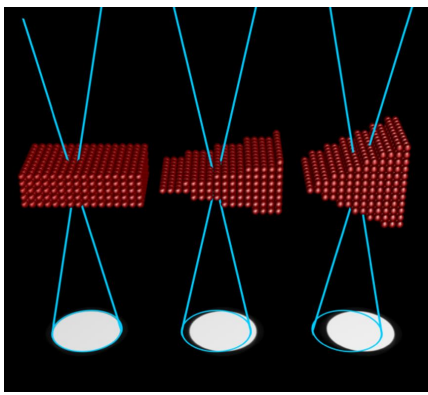
Possible
at FZU

ACTEM
with better
detector
needed

4D-STEM

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Phase Contrast Analysis

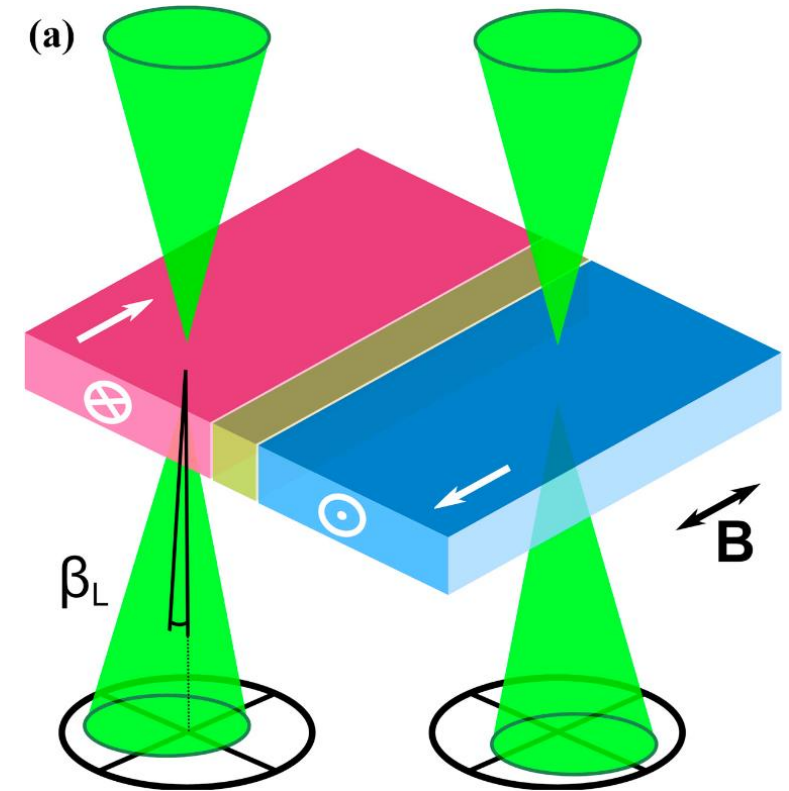
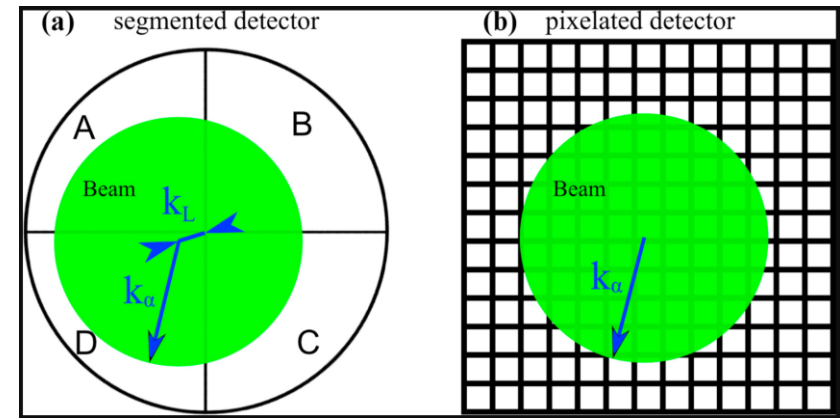
Differential Phase Contrast (DPC)

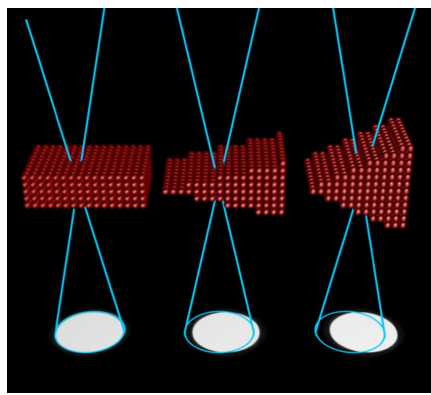
- sensitive to the magnetic field of the sample



Lorentz imaging technique in TEM

- direct measure of the two components of the Lorentz deflection
- resolution approximately equal to the electron probe size





Phase Contrast Analysis

Differential Phase Contrast (DPC)



Atomically sharp domain walls in an antiferromagnet

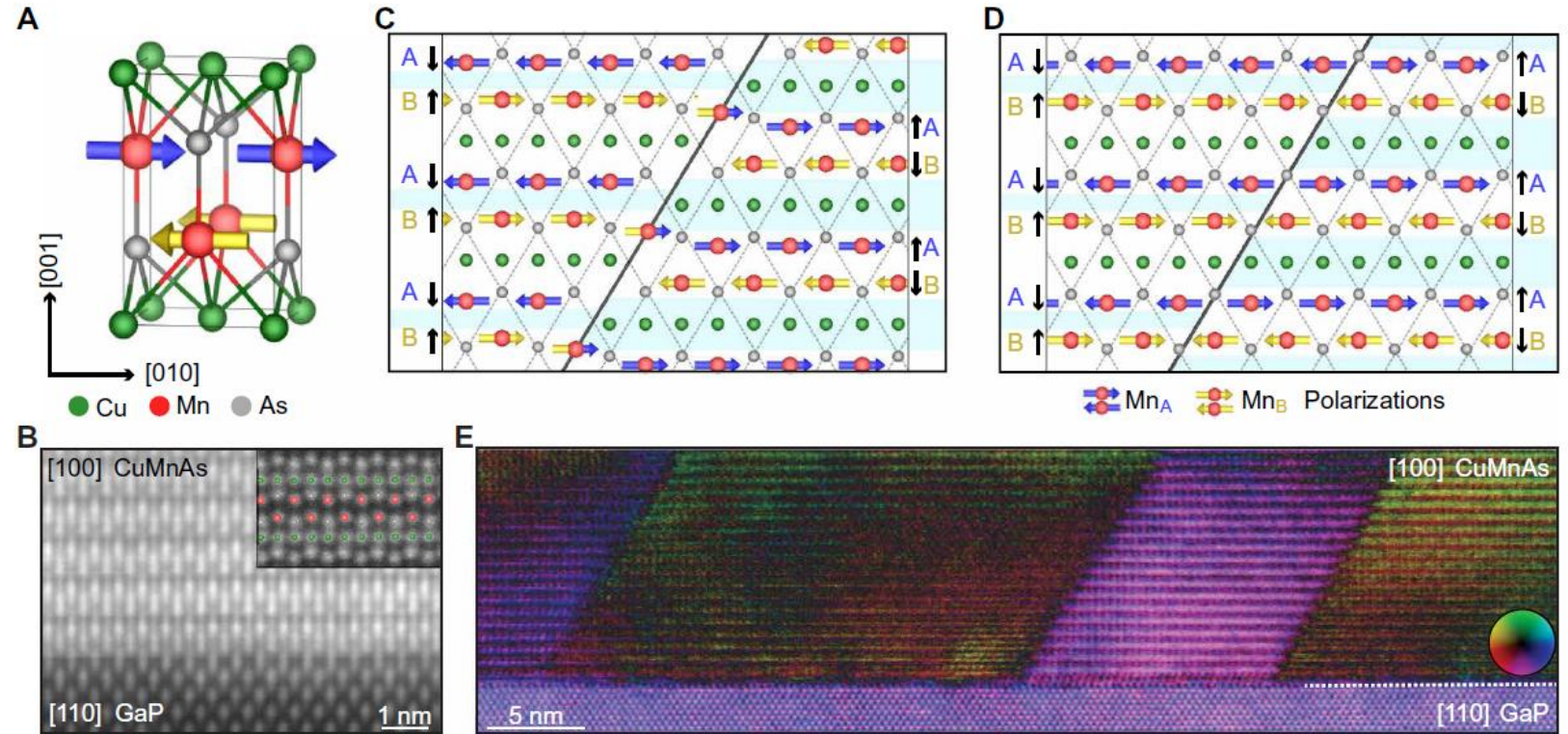
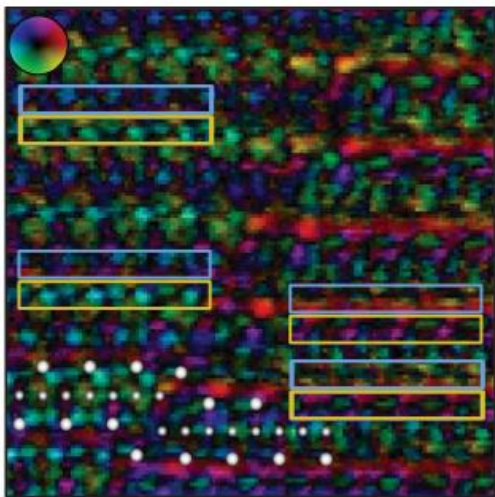
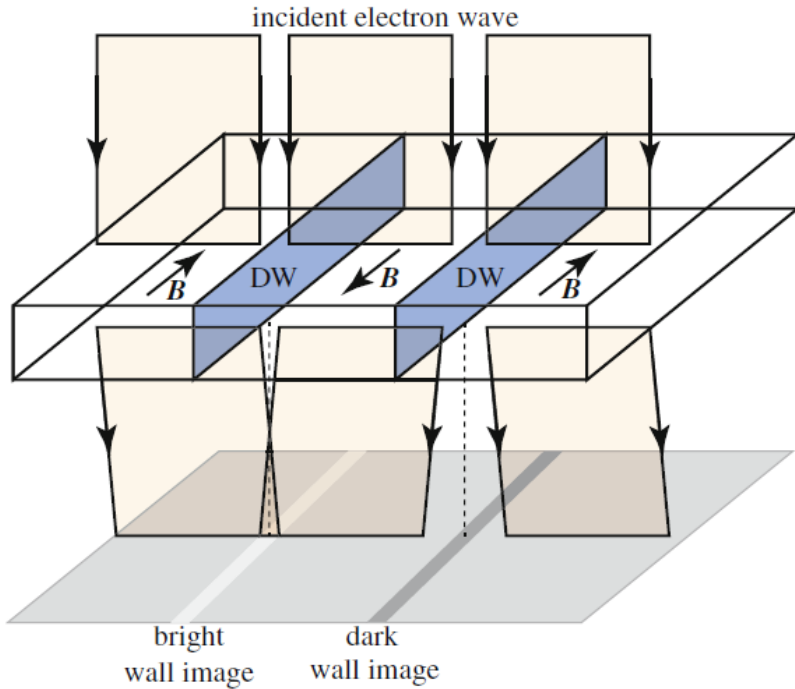


Fig. 1. Crystal structure and atomically sharp domain walls in antiferromagnetic CuMnAs. (A) Atomic model of the CuMnAs unit cell. (B) High-angle annular dark-field (HAADF)–STEM image of a [100] projection of the epitaxial CuMnAs film grown on lattice-matched GaP. (C and D) Schematics of the atomically sharp domain walls at an antiphase boundary defect and in an unperturbed area of the CuMnAs single crystal, respectively. Symbols A (blue) and B (yellow) label the upper and lower Mn sublattices from the unit cell in (A). Thin dashed lines highlight preserved As atom matrix. Black arrows represent Lorentz force direction at individual sublattices, which focuses the deflected beam into the areas with light blue overlay. (E) An overview DPC-STEM image of the atomically sharp domain walls in a CuMnAs film.

Lorentz TEM



- Fresnel (defocus) Mode
- For imaging – out-of-focus
- Magnetic domain walls - bright (convergent) and dark (divergent) lines
- Spatial resolution limited – high defocus value needed
- No information is directly available about the direction of magnetization
- Transport-of-Intensity Equation (TIE) formalism

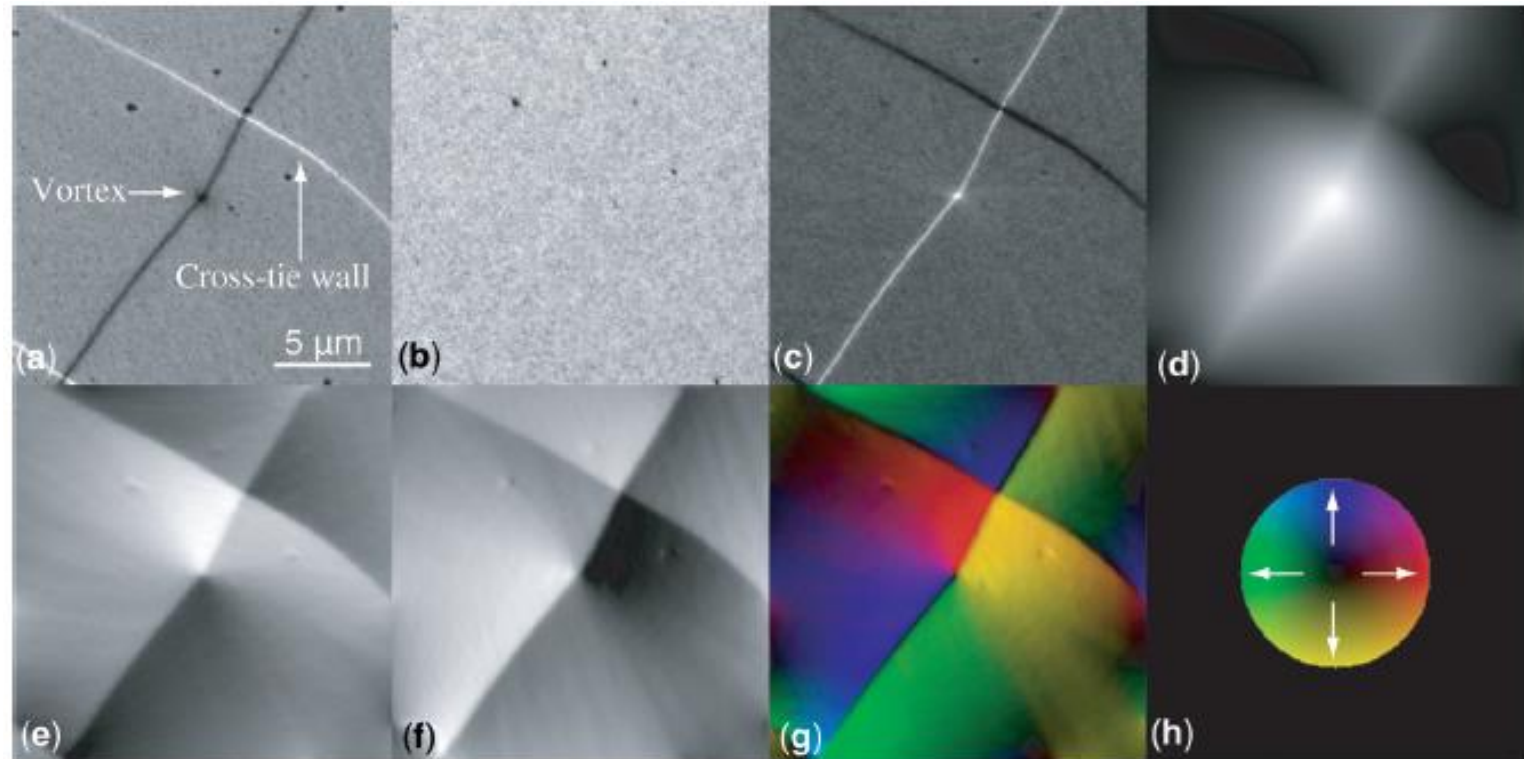
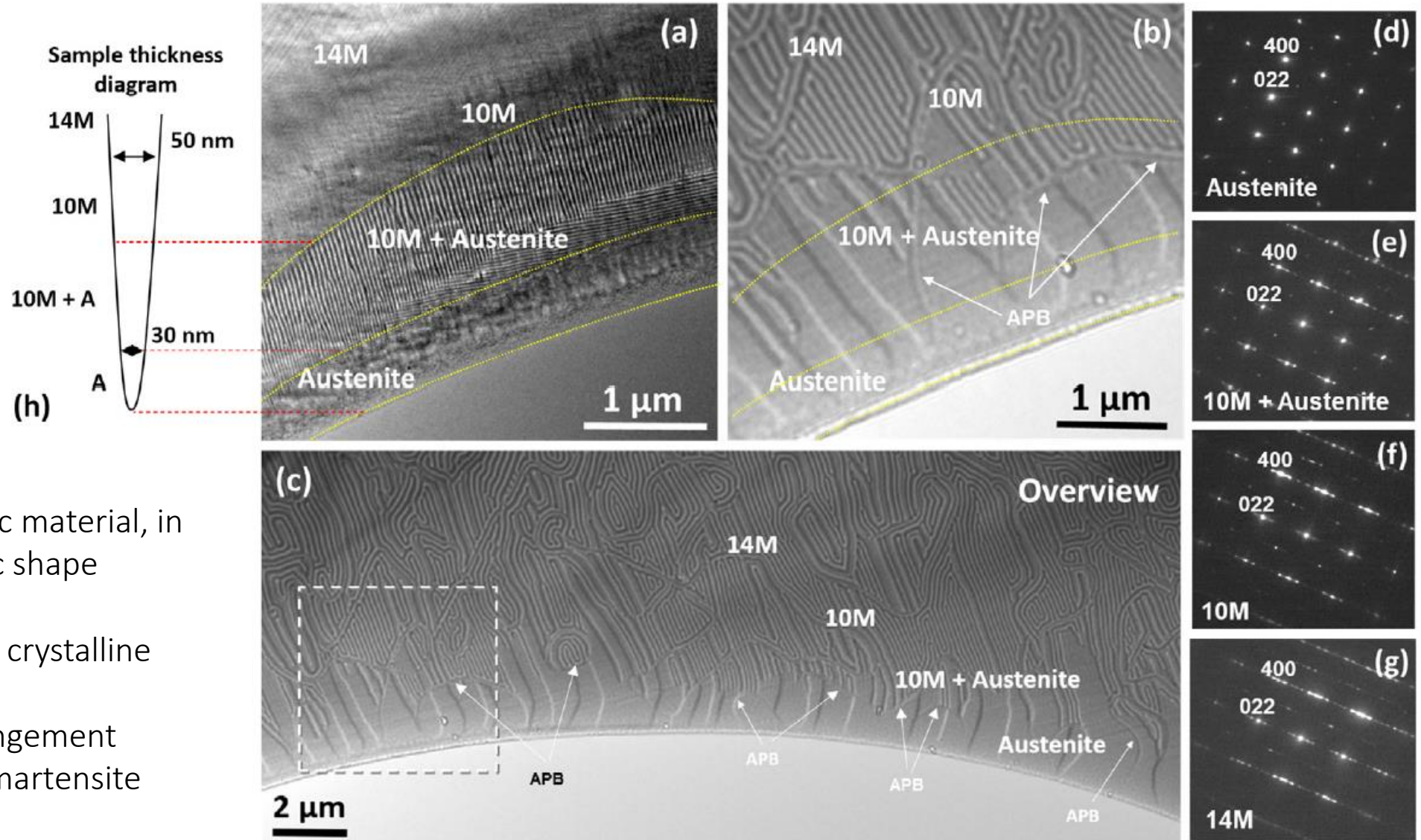


Figure: Phase reconstruction for a vortex–antivortex configuration in a Permalloy thin film (thickness 50 nm); (a)–(c) through focus series, (d) reconstructed phase, (e) and (f) gradient components of the phase, (g) and (h) color plot of the magnetization along with the color legend

LTEM in Ni-Mn-Ga Alloys - LEM result example

Cooperation
with
O. Heczko
research

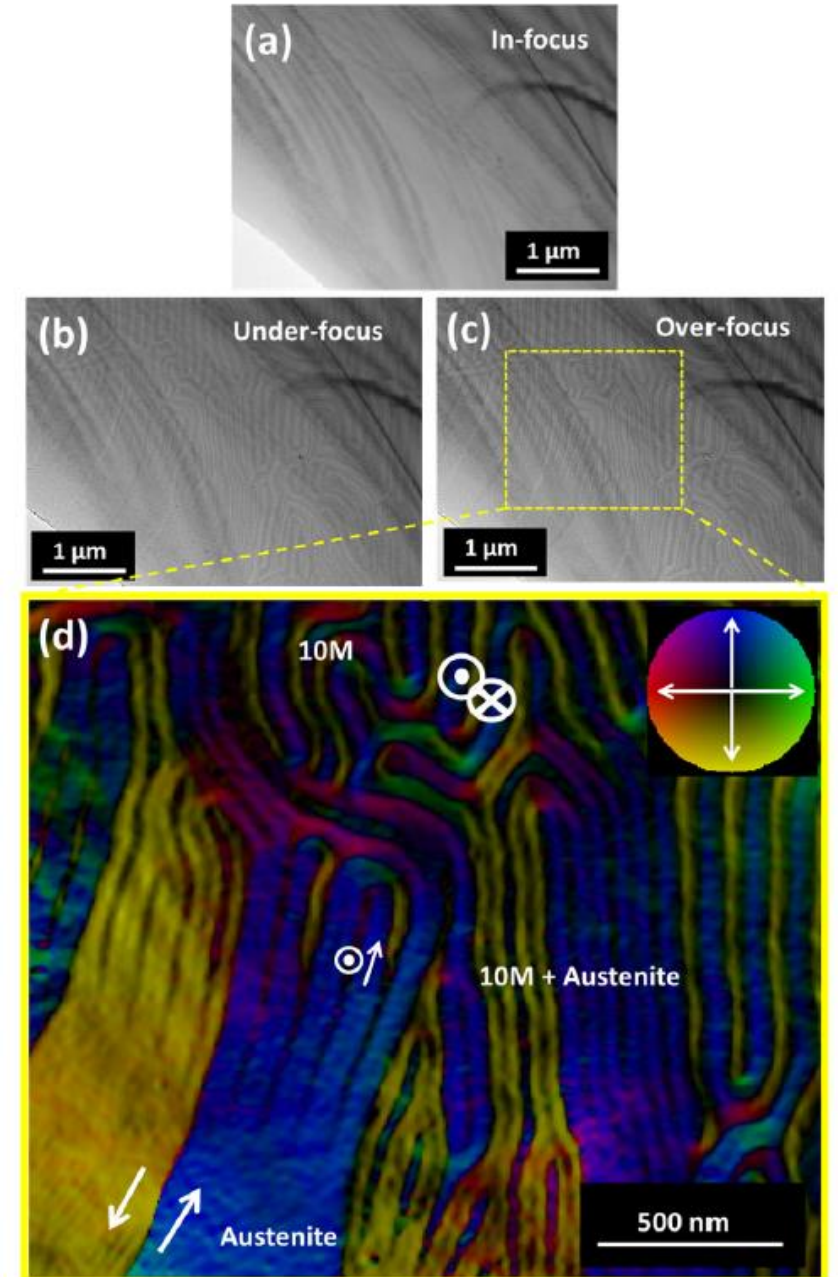
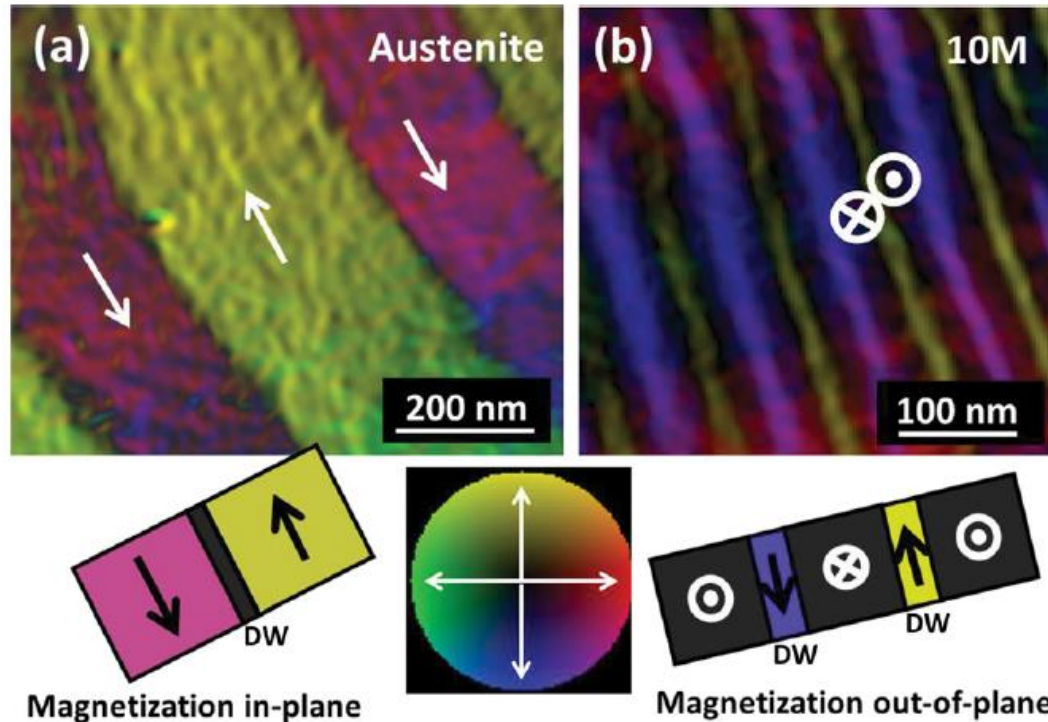


- Ni-Mn-Ga – multiferroic material, in this case ferromagnetic shape memory alloys
- $\text{Ni}_{50}\text{Mn}_{25}\text{Ga}_{20}\text{Fe}_5$ single crystalline foil
- Magnetic domain arrangement across the austenite - martensite interface

LTEM in Ni-Mn-Ga Alloys - LEM result example

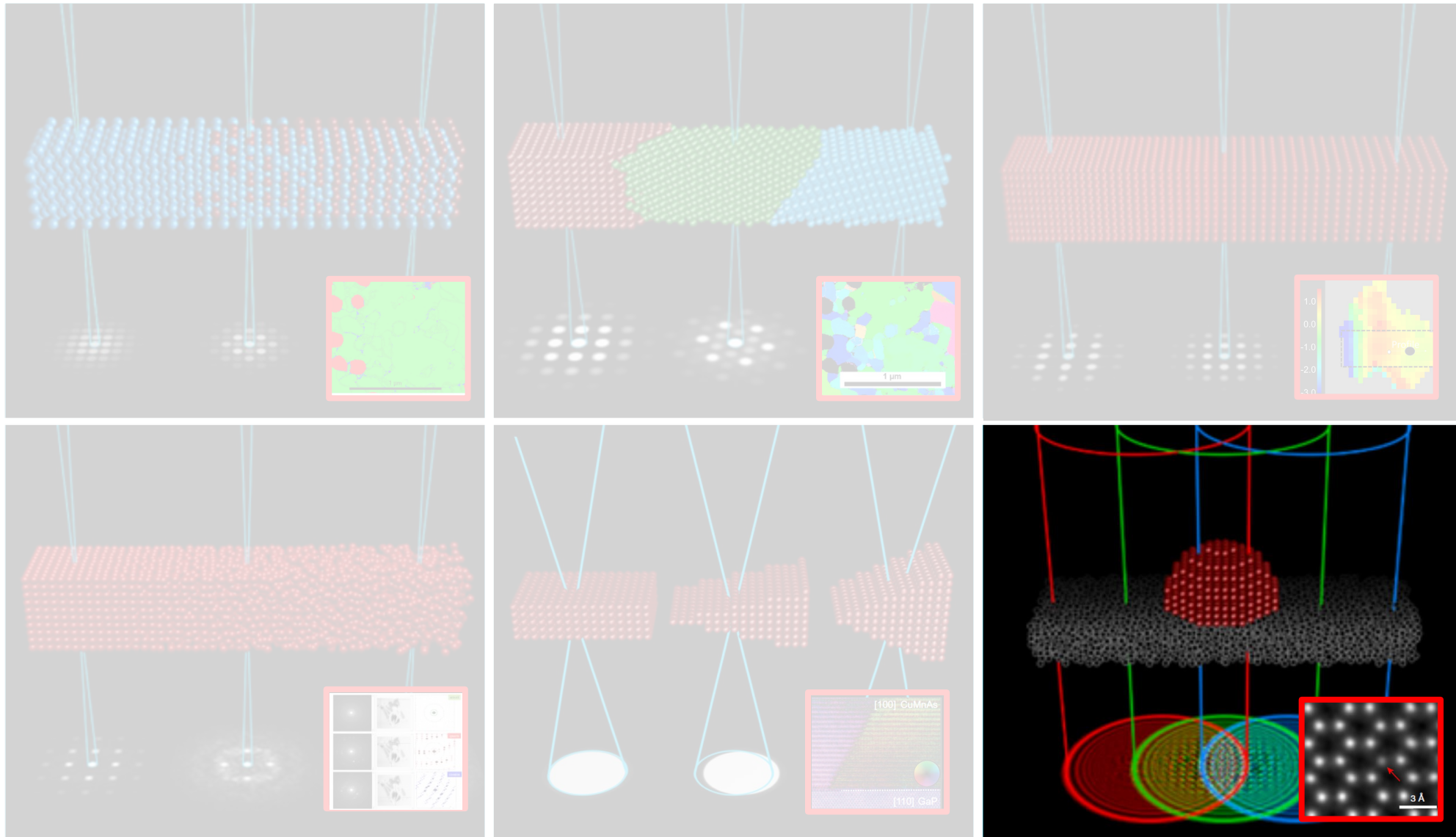
- Ni-Mn-Ga – multiferroic material, in this case ferromagnetic shape memory alloys
- $\text{Ni}_{50}\text{Mn}_{25}\text{Ga}_{20}\text{Fe}_5$ single crystalline foil
- Magnetic domain arrangement across the austenite - martensite interface
- Across the interface, the labyrinth magnetic domain structure of 10M martensite changed to the wide domain structure of austenite

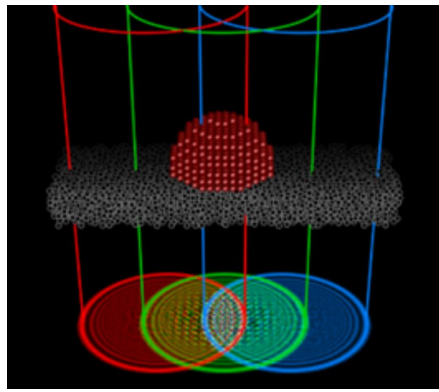
Cooperation
with
O. Heczko
research



4D-STEM

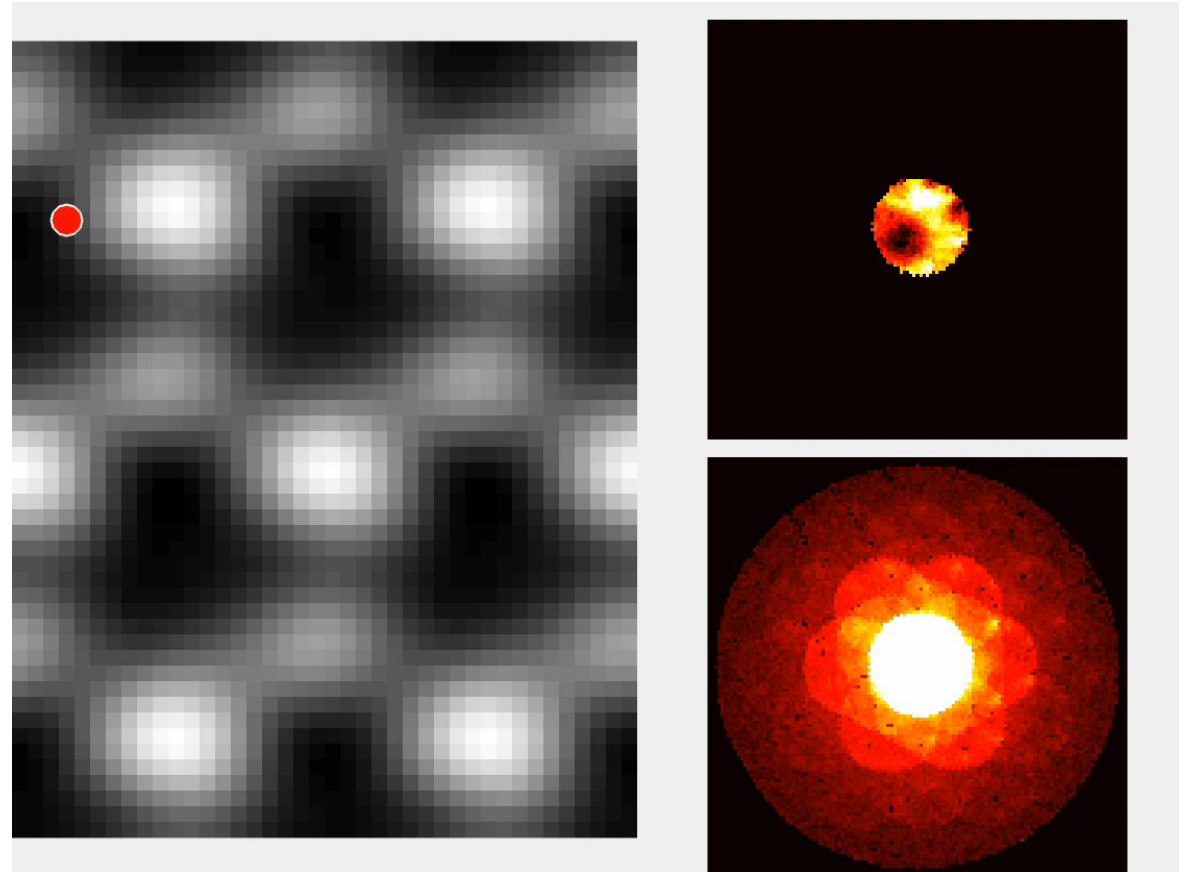
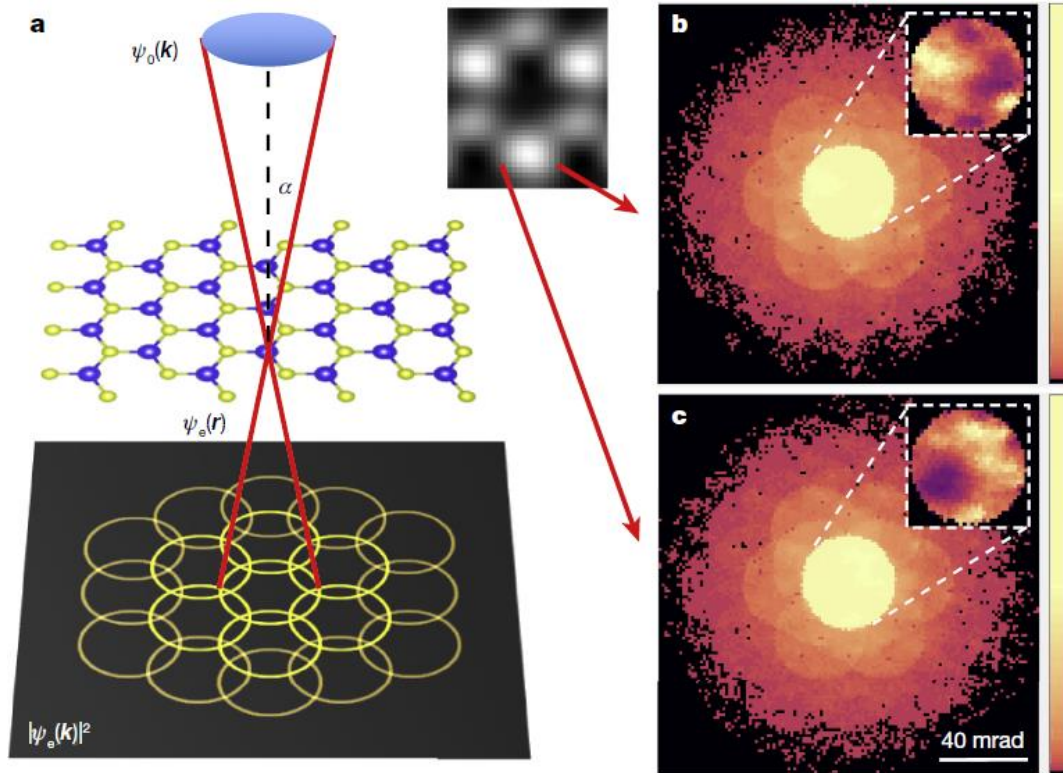
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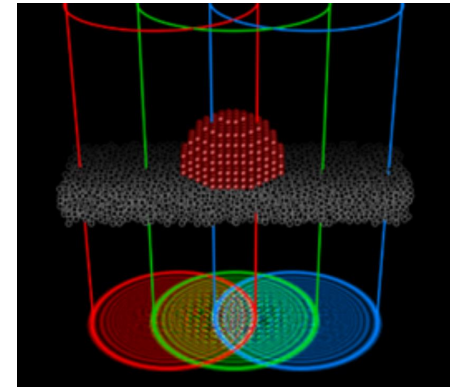
Electron Ptychography

- Computational imaging technique
- The beginnings of principle of the method in the 60s – previously in the visible-light imaging (11) and x-ray imaging
- New detectors – new possibility for ptychography in TEM
- Overlapping CBED patterns used for the construction of the complex electron probe and complex sample potential

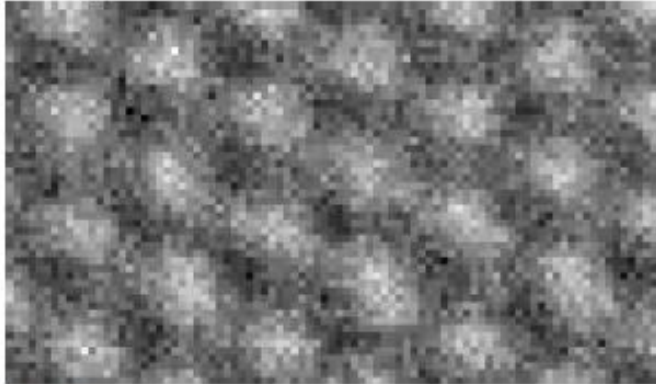


Electron Ptychography

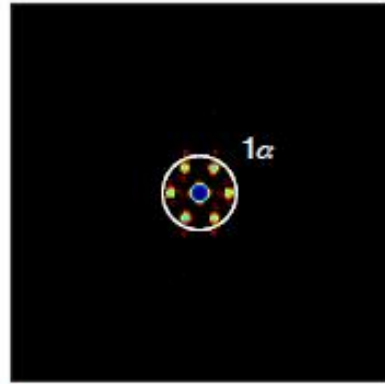
- STEM resolution is also determined by the double of the semi-convergence angle (α) - 2α
- STEM microscopes are currently near 50 pm spatial resolution
- Ptychographic reconstructions with 4D STEM data provide higher contrast than ADF or HAADF and segmented DPC imaging in STEM
- Resolution less than 20 pm



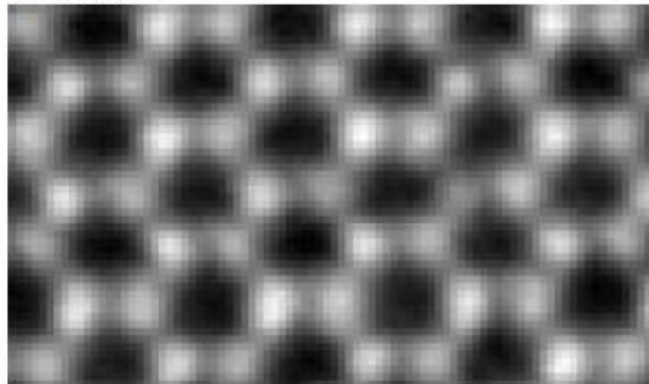
a Bright-field



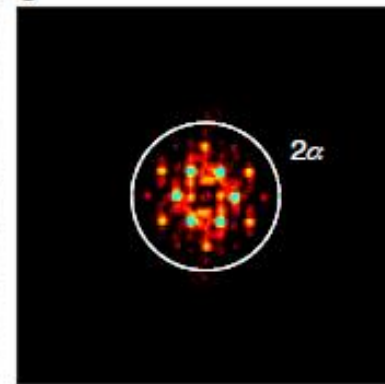
e



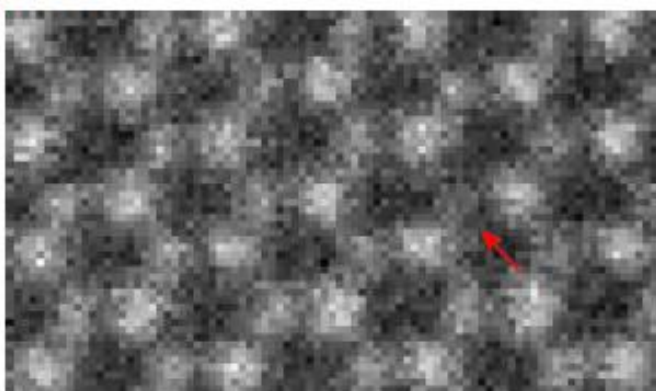
c iCoM



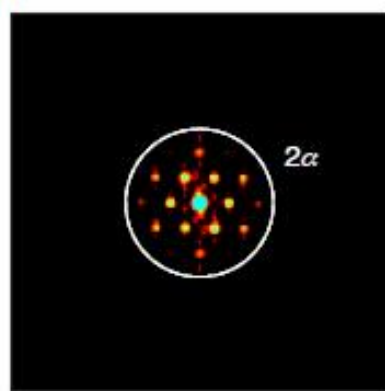
g



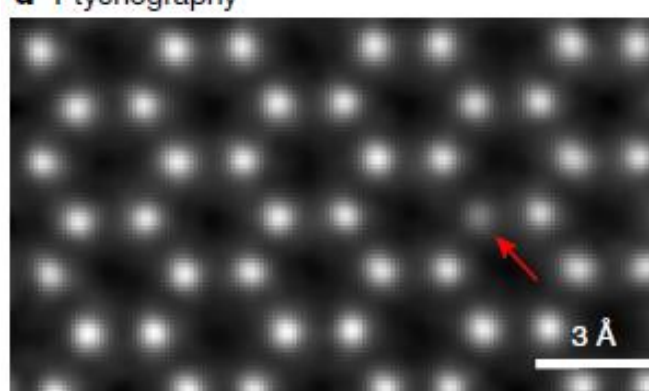
b ADF



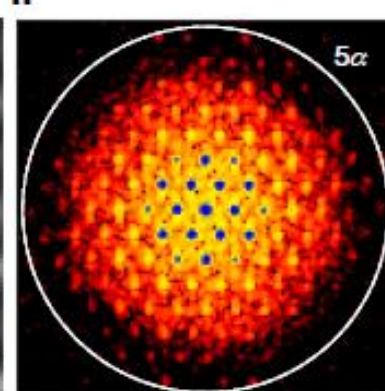
f



d Ptychography



h

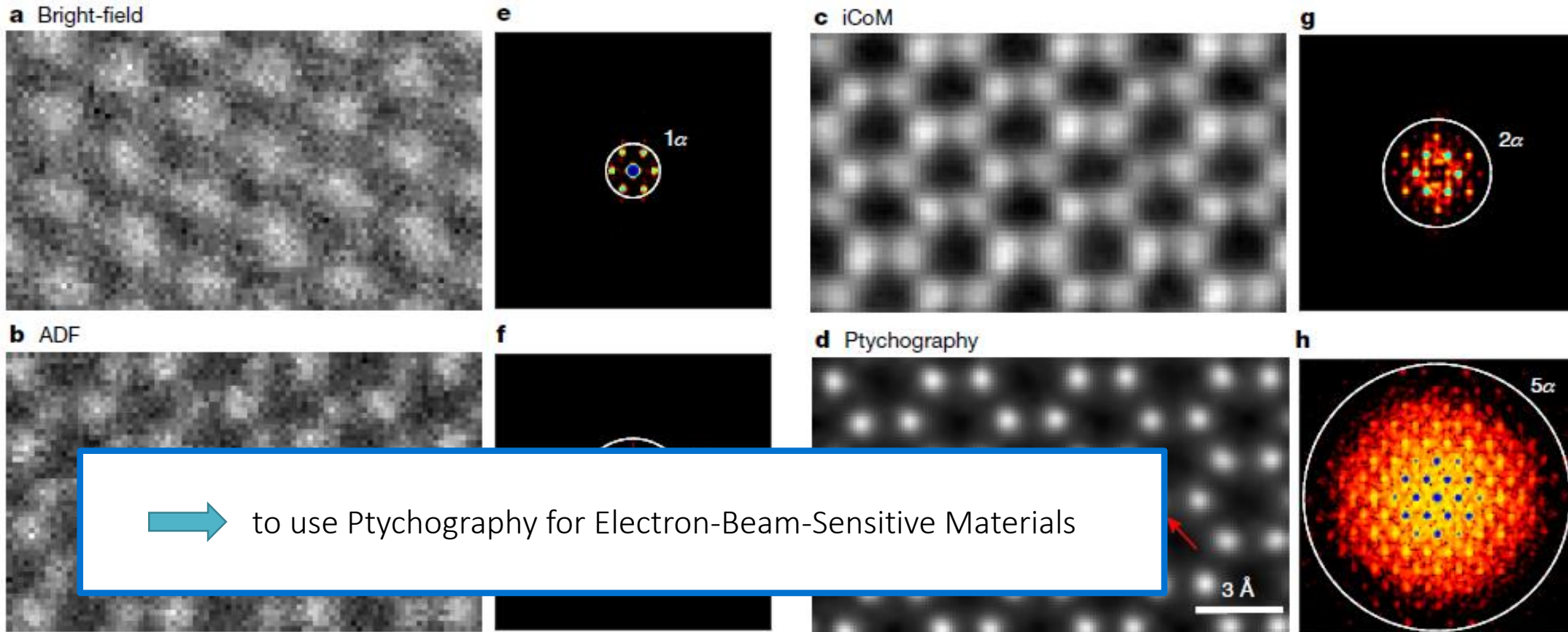
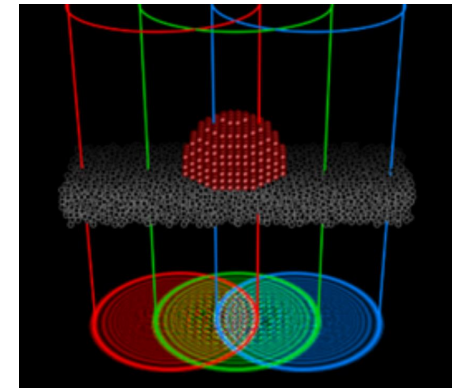


monolayer
MoS₂ at
80 kV

[Nature (2018),
559, 343-349]

Electron Ptychography

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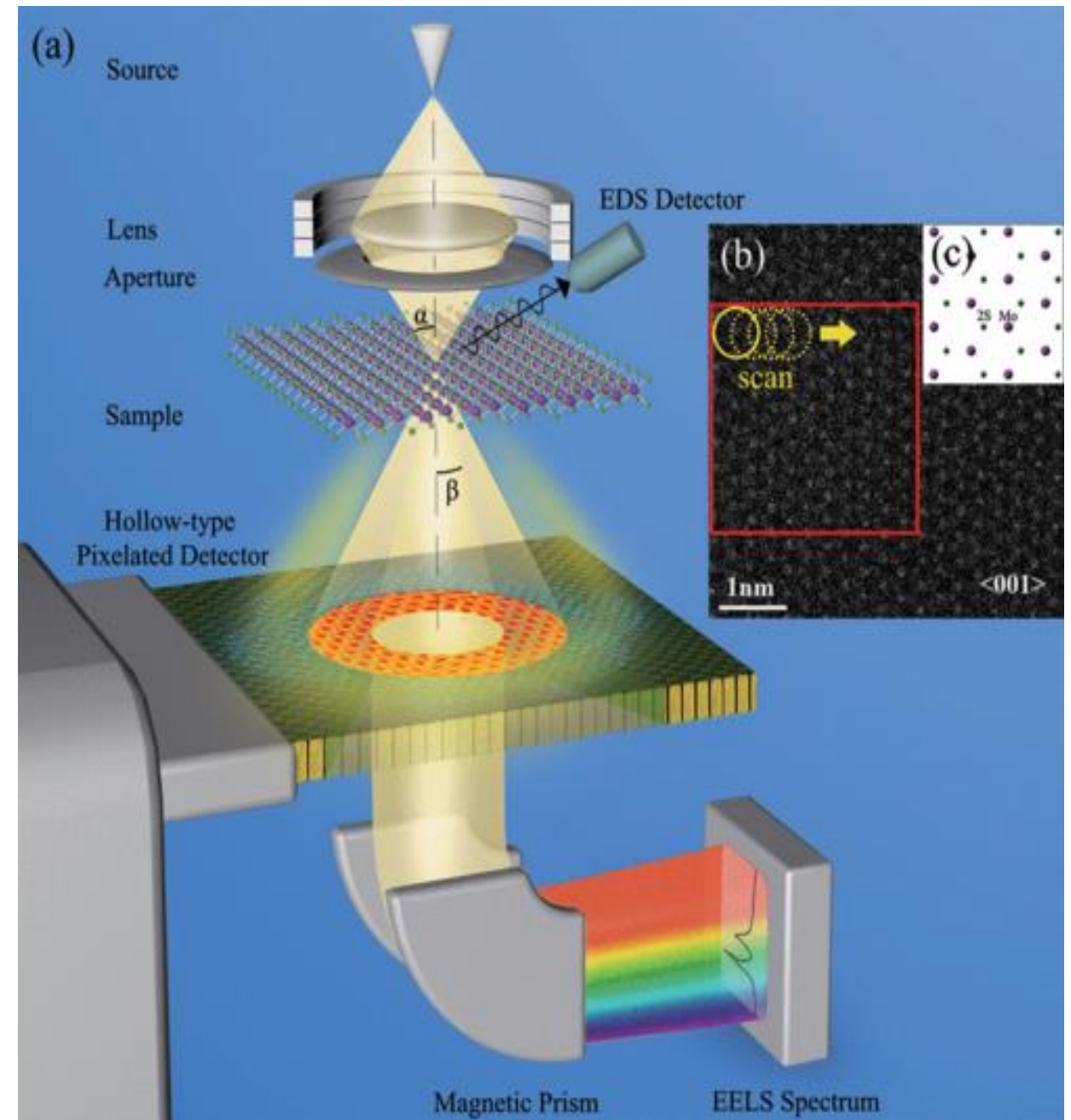
monolayer
MoS₂ at
80 kV

[Nature (2018),
559, 343-349]

Beyond Ptychography

Ptychography with Hollow Pixelated Detector (5D STEM)

- method for quantitative phase recovery and simultaneous electron energy loss spectroscopy (EELS) analysis using ptychographic reconstruction
- potential for recovering both structural and chemical information at atomic resolution

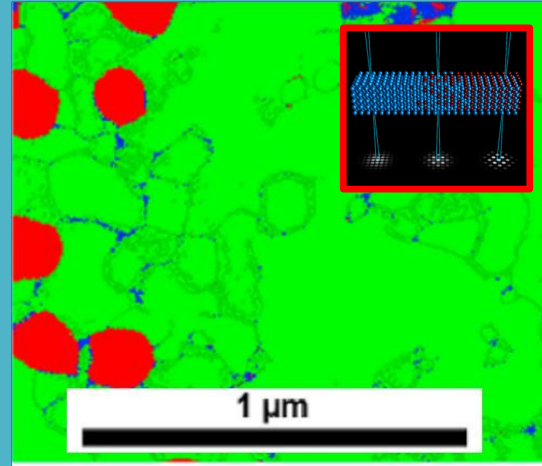


4D-STEM techniques

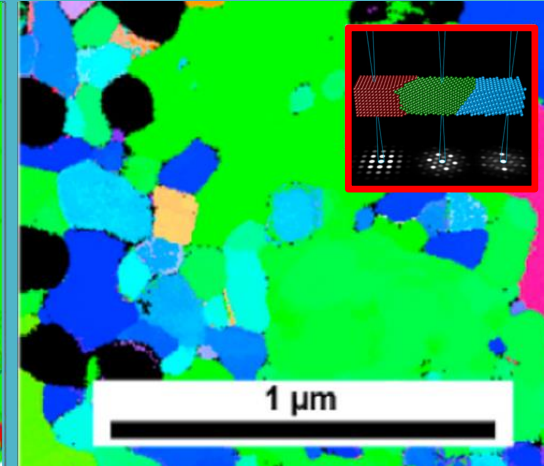
Summary of Multidimensional Transmission Electron Microscopy

- Elemental Mapping
- 4D-STEM
- Lorentz multidimensional TEM
- For atomic resolution in 4D-STEM ACTEM needed

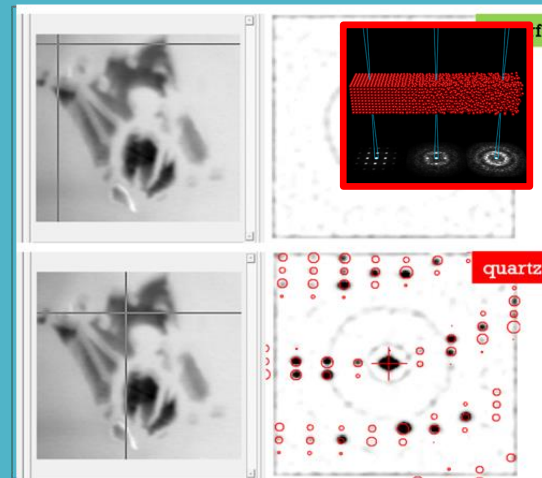
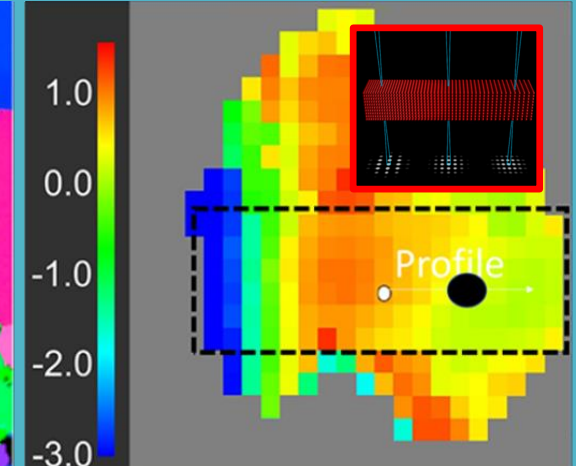
Phase map



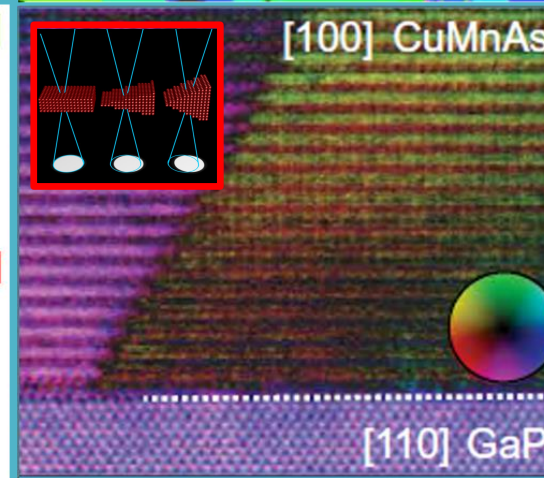
Orientation map



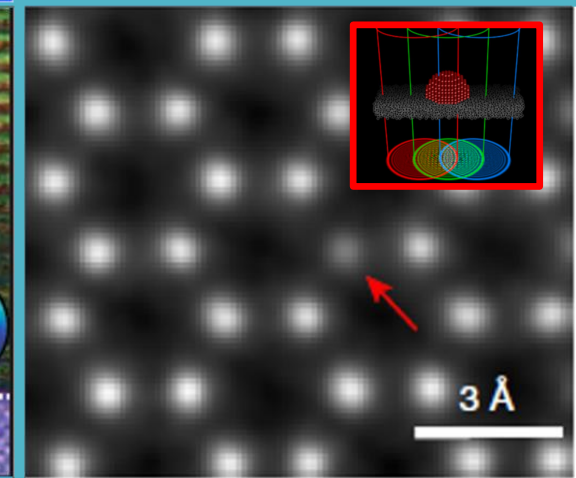
Strain map



Structural disorder



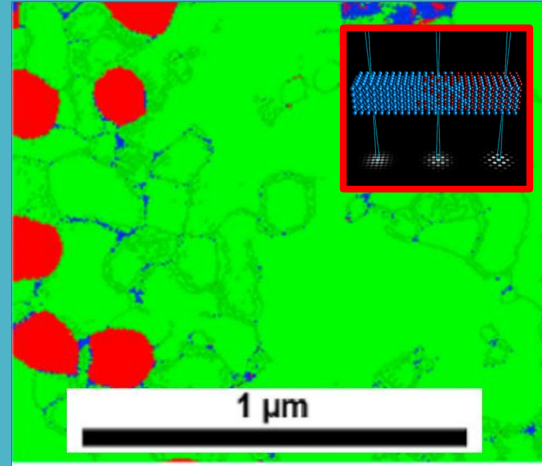
Differential
Phase Contrast



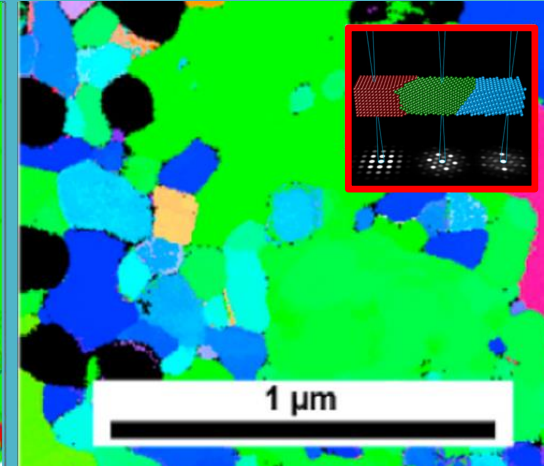
Ptychography

4D-STEM techniques

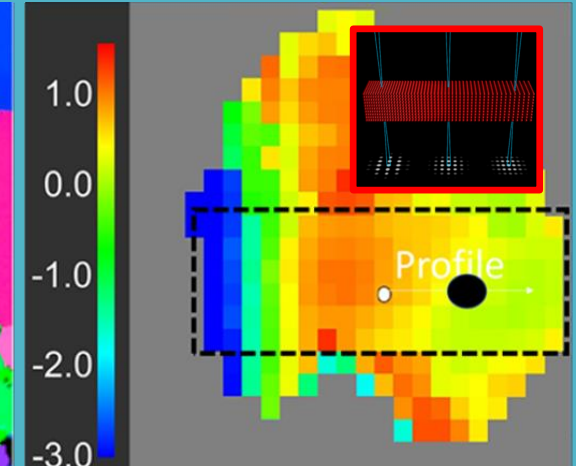
Phase map



Orientation map



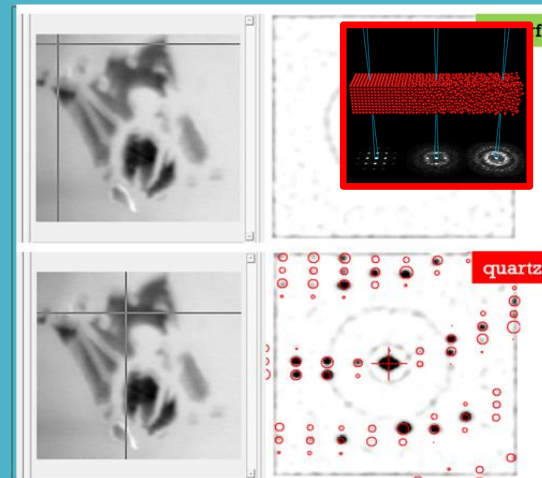
Strain map



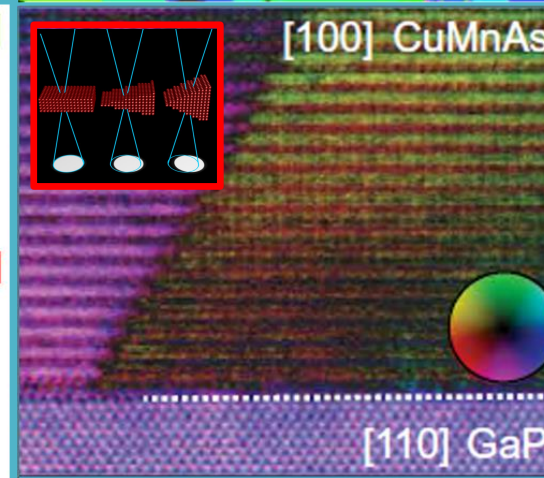
Thank you for
your attention

Marek Vronka

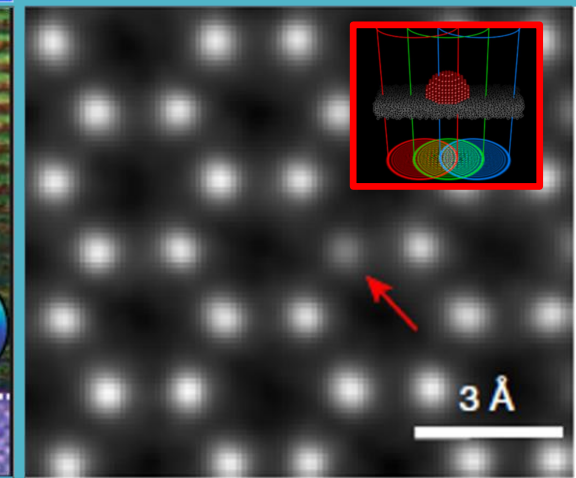
25.5.2023



Structural disorder



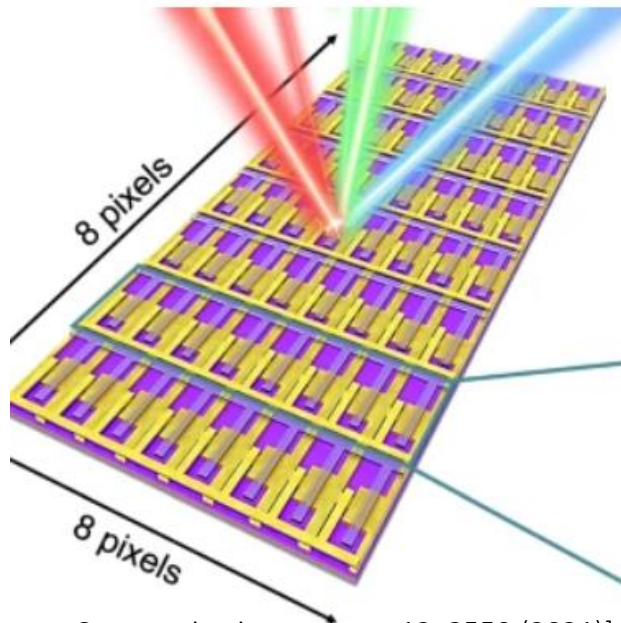
Differential
Phase Contrast



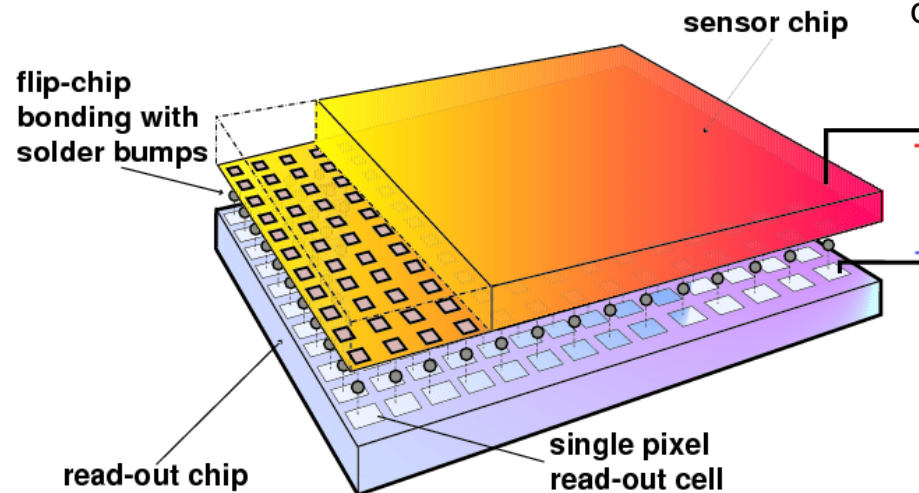
Ptychography

Pixelated detectors

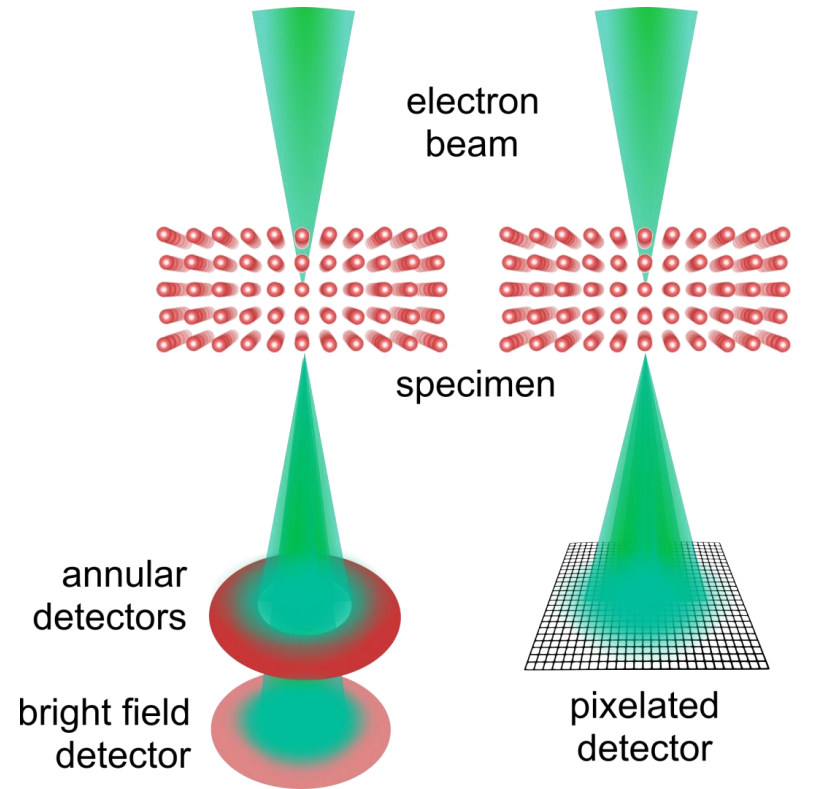
- monolithic active pixel sensors (APS) - complementary metal-oxide-semiconductor (CMOS) chip
 - sensitive doped epitaxial layer
- hybrid pixel array detector (PAD)
 - array of photodiodes is bump bonded to a specific integrated circuit



[Nature Communications volume 12, 3559 (2021)]



[Nuclear Instruments and Methods in Physics Research A 501 (2003) 239–244]



[quantumdetectors.com]

Precession Electron Diffraction

Precession electron diffraction (PED) is a specialized method to collect electron diffraction patterns in a transmission electron microscope (TEM). By rotating (precessing) a tilted incident electron beam around the central axis of the microscope, a PED pattern is formed by integration over a collection of diffraction conditions. This produces a quasikinematical diffraction pattern that is more suitable as input into direct methods algorithms to determine the crystal structure of the sample.

