Aberration-corrected imaging and spectroscopy for multidisciplinary materials characterization

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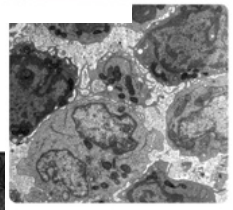
Electron Microscopy Unit

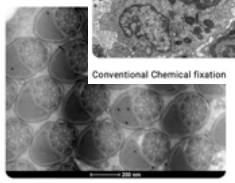
Core facility for Life Science and Materials Science Microscopy

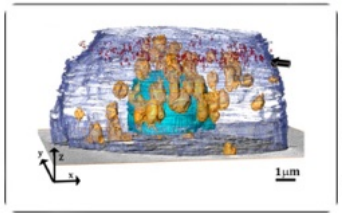
WEIZMANN INSTITUTE OF SCIENCE

Advanced light microscopy | Micro CT | Biological FIB-SEM | Biological SEM

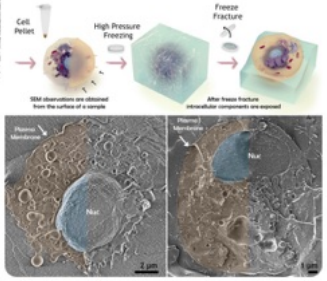
Material science FIB-SEM | Material science SEM | Material science TEM | Biological TEM



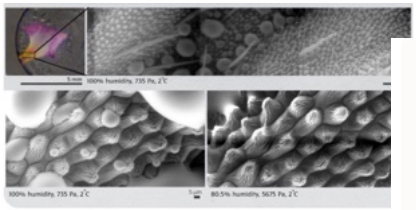




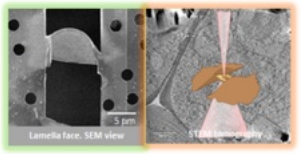
Correlative LM and EM (CLEM)



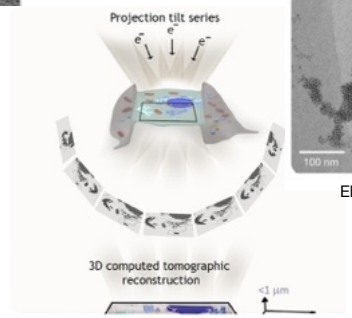
Cryo SEM



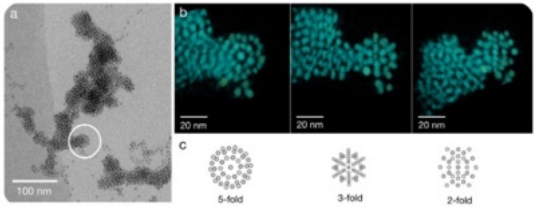
Wet mode SEM



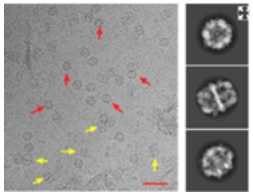
Cryo lamellae preparation for Cryo TEM/STEM



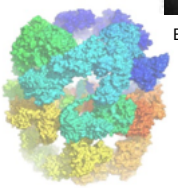
Electron tomography

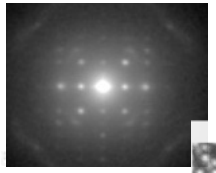


Electron tomography

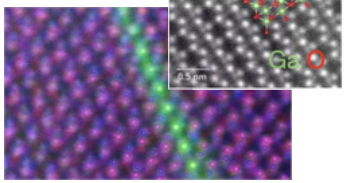


SPA





Electron Diffraction



Atomic resolution analytical TEM

Staff

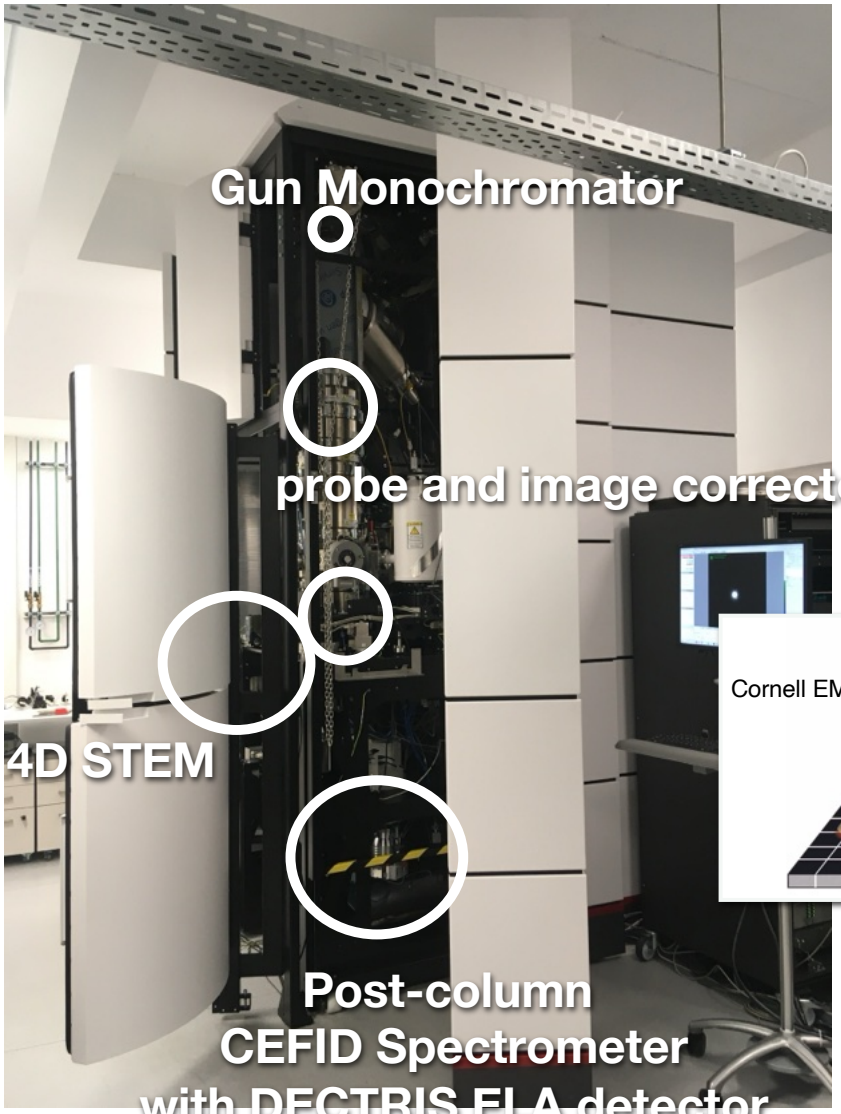
PhD level	13
Technician	2
Admin	1++

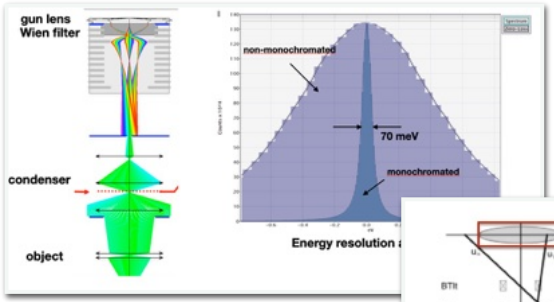
Users - PIs/Groups

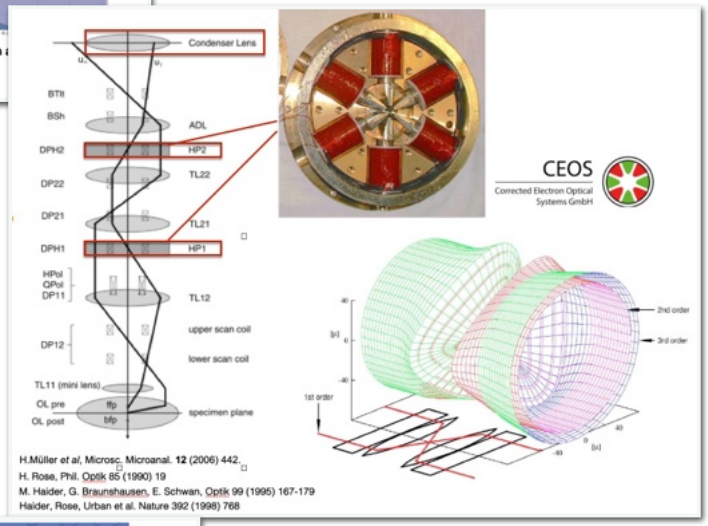
2022	2023	2024
84	82	83

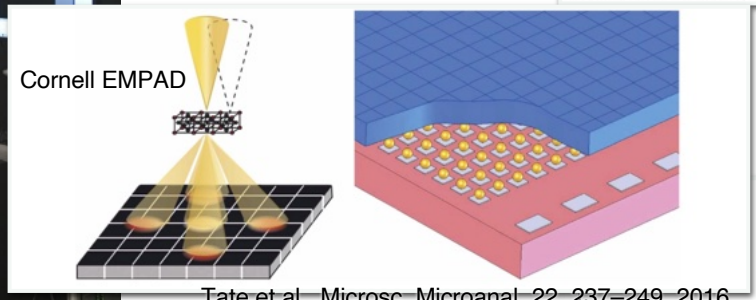
Instruments in Service

2022	2023	2024
47	54	44

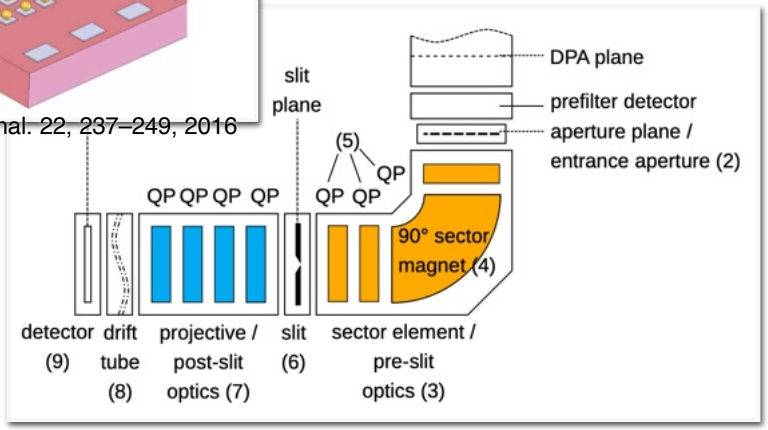




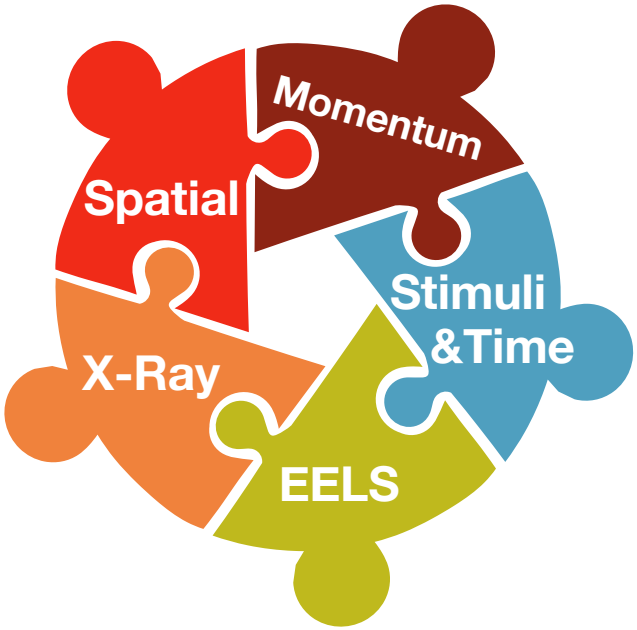


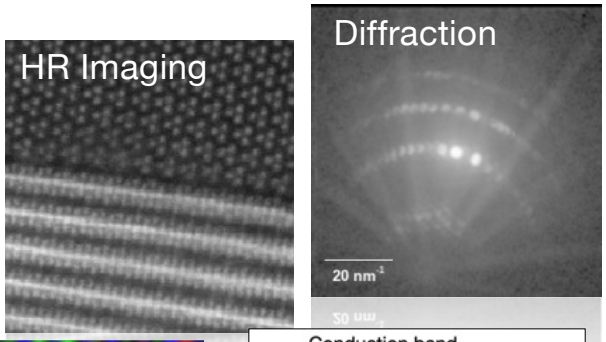


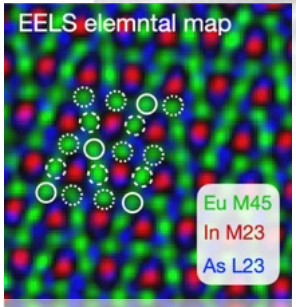
Tate et al.. Microsc. Microanal. 22, 237–249, 2016

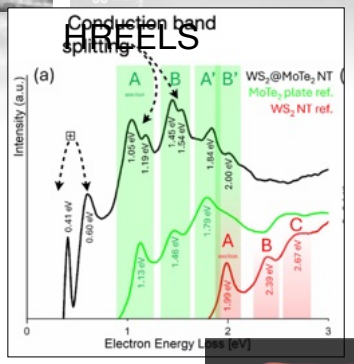


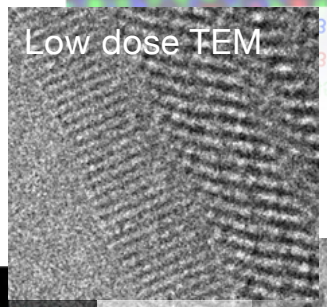
Multimodal characterisation

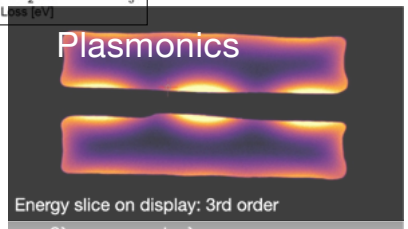


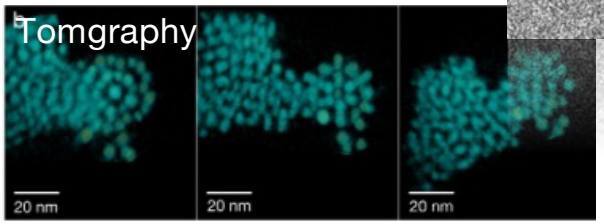


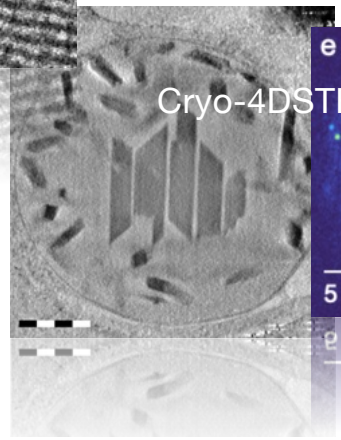


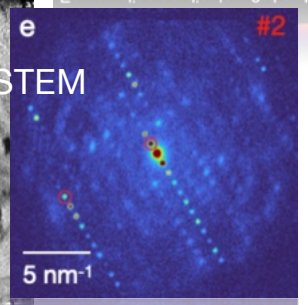




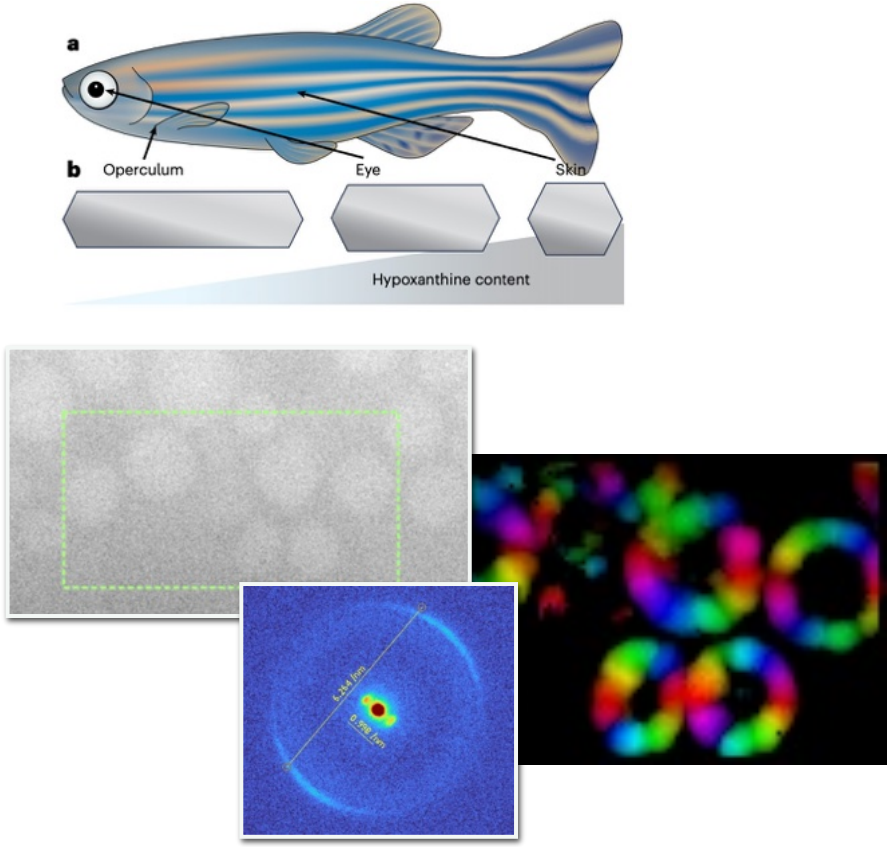




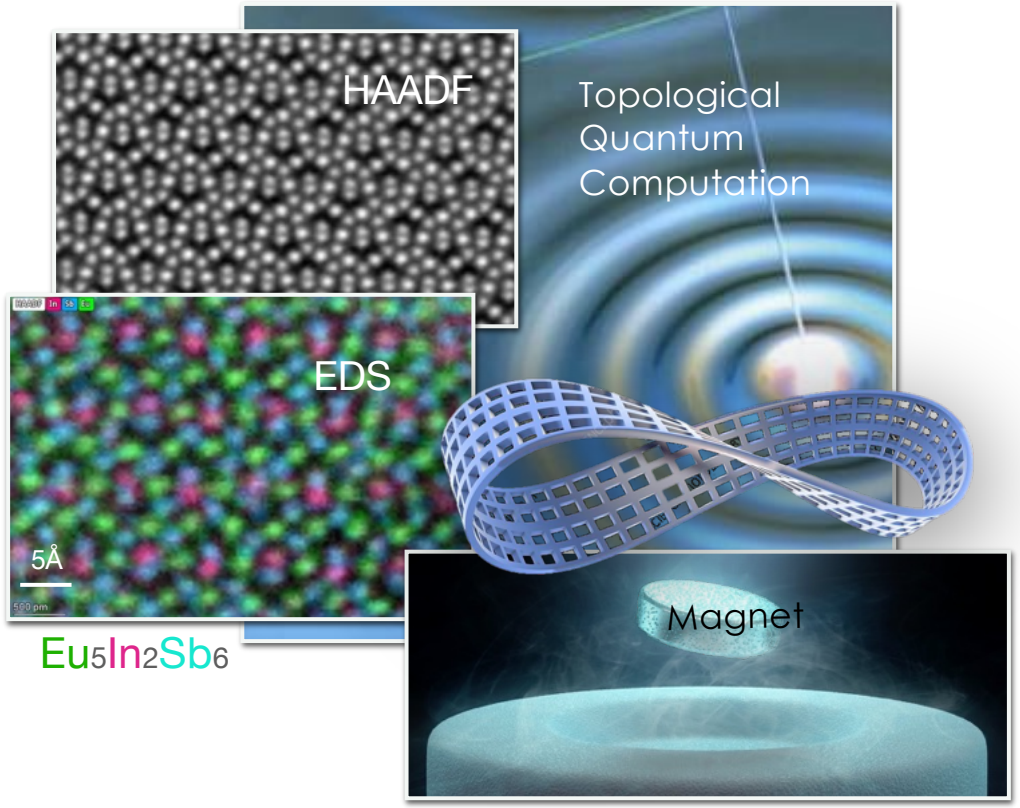




Cross-discipline Characterisation



Genetic regulation of **guanine crystal formation** in pigment cells in zebrafish.



New low-dimensional **Axion Insulators** for quantum phenomena and fault-tolerant quantum devices

Practical Considerations for High-resolution Microscopy

Sample limitations

Dose limitation, preparation limitations, stability and contamination

Achieving the objective

Which information do you seek and can you get it from your sample?

The readiness factor (be ready when the sample is)

Sample limitations

Radiation damage

charging, heating, radiolysis (e-/e- interaction), knock-on damage (e-/nucleus) → breaking of chemical bonds, structural damage on atomic scale, longer-range order disruption of structure, mass loss of chemical elements

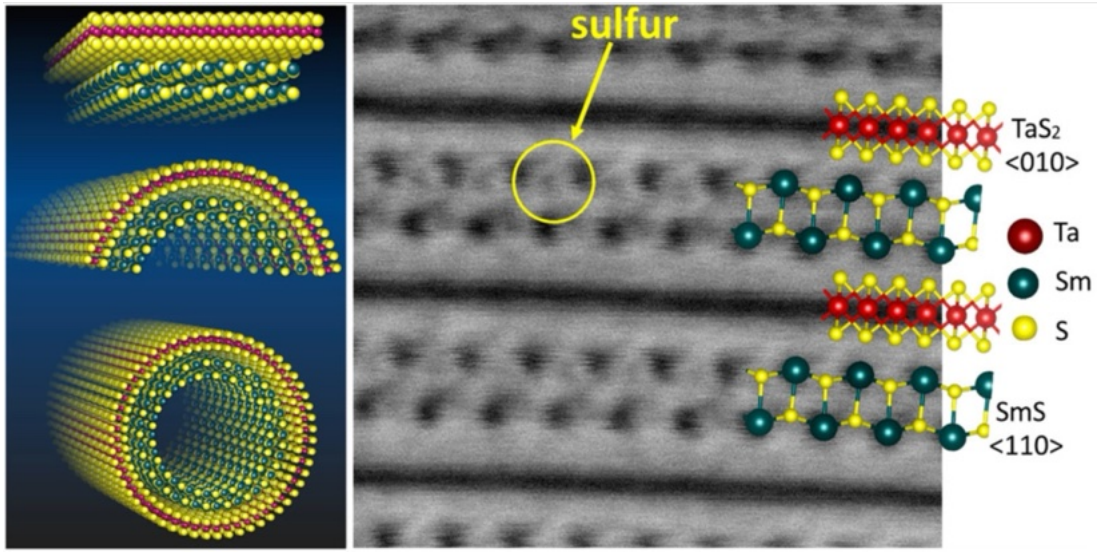
K. Nordlund, Journal of Nuclear Materials 512 (2018) 450479
R. Egerton, *Micron* 119 (2019); Microscopy Today 29 (2021) 56.
C. Russo, MRS Bulletin 44 (2019) 935

Preservation of the material context

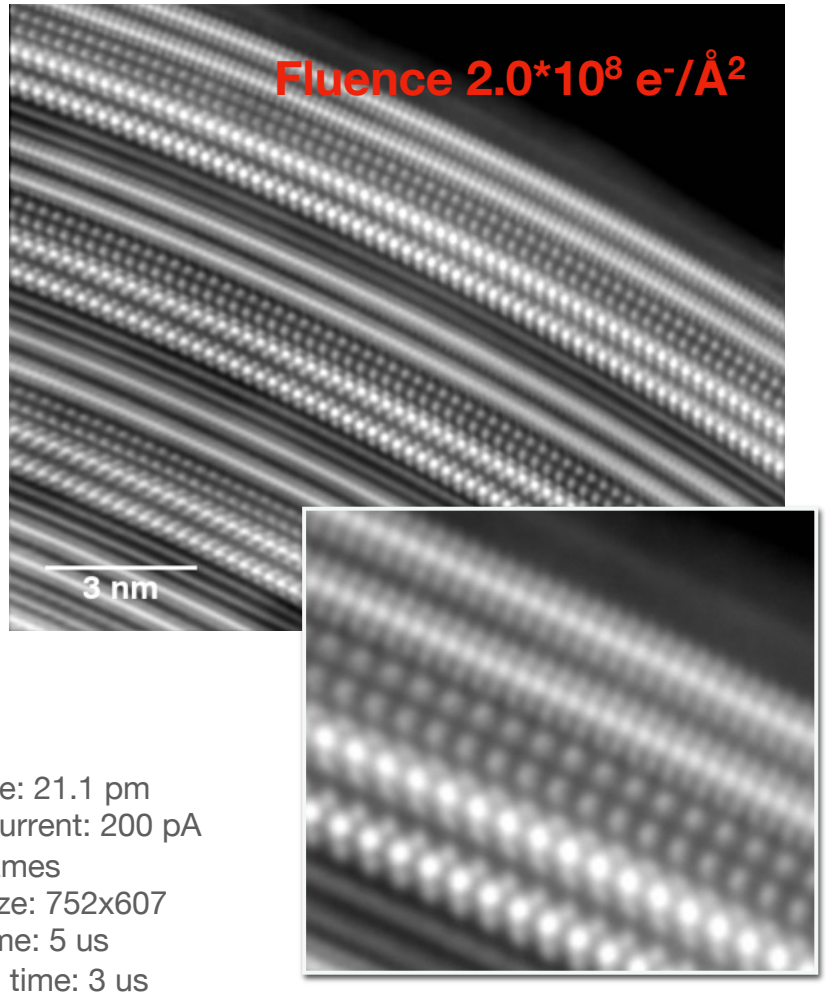
preparation of thin transparent samples (cutting, FIB-milling, drying)
transfer through ambient → structural and morphological artefacts, alteration of chemistry, phases, order

Dose-limited resolution: Extreme case of a resistant material

Misfit layer compound nanotube



cumulation up to 1127 frames
=> spatial resolution for localisation of Se, La, Ta



Pixel size: 21.1 pm
Beam Current: 200 pA
1127 frames
Array Size: 752x607
Dwell time: 5 us
Shaping time: 3 us

Total acquisition time: 1 h 10 min

Sreedhara MB, Chemistry of Materials **34** 2022

HRTEM imaging of beam sensitive materials …

Organics, polymers, defects in 2D materials, surfaces

Characteristic dose D_c, equivalent fluence ($D_{ec} = D_c/e$) and damage cross section ($\sigma_d = 1/D_{ec}$) determined by several techniques (Reimer and Kohl, 2007; [a] Isaacson, 1977; [b] Hobbs, 1975a; [c] Egerton et al., 1987; [d] Pan and Crozier, 1993; [e] Li and Egerton, 2004; [f] Egerton, 2012; [g] Hooley et al., 2017; [h] Hollenbeck and Buchanan, 1990; [s] Stark et al., 1996: 7 Å spots). Unless otherwise stated, the data are for 100 keV electrons and a specimen at room temperature. The last column shows the factor by which the measured characteristic dose increases when the specimen temperature is reduced to 100 K.

Method	Material	dose D_c (C/cm^2)	$D_{ec} = D_c/e$ (e/Å^2)	σ_D (Mb) = 100/D_{ec}	$D_c(100)$ $D_c(300)$
Fading of spots in electron diffraction patterns	Bacteriorhodopsin[s]	0.0008	0.5	200	9
	Amino acid (glycine)[a]	0.0025	1.6	64	
	Polyethylene	0.01	6.0	16	
	Coronene[e]	0.11	70	1.5	
	Phthalocyanine (Pc)	0.2	120	0.83	
	Cu-phthalocyanine	2.5	1600	0.06	
	Chlorinated Cu-Pc	20	12000	0.008	
	ZSM-5 zeolite[d]	0.5	300	0.33	
	Calcite (200 kV)[g]	62	39,000	0.0026	

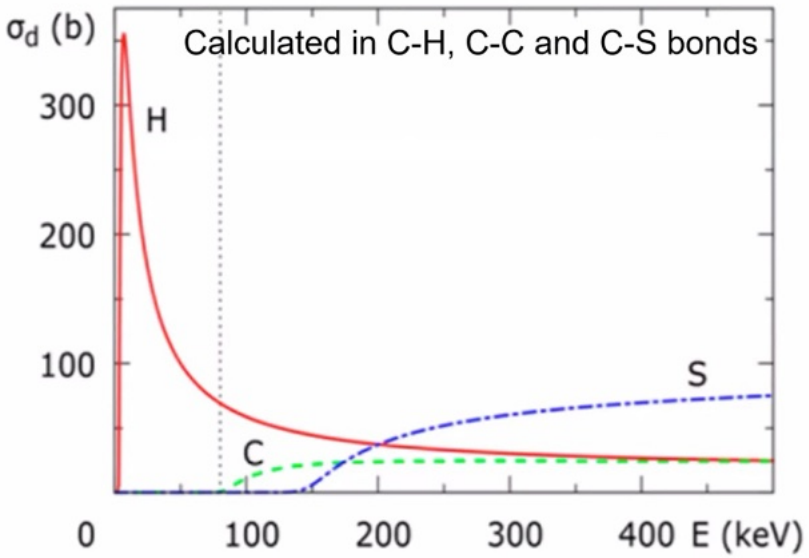
Ray Egerton, Radiation damage to organic and inorganic specimens in the TEM, Micron 119 (2019) 72–87

For comparison: TMDC or graphene - typically 10^4-10^5 e/A^2

Organics: Low Voltage doesn't solve the knock-on problem

Ballistic displacement cross section for H in C-H, C in C-C, and S in C-S bond

$$\sigma_d = 4\pi\left(\frac{Ze^2}{4\pi\varepsilon_0 2m_0c^2}\right)^2 \frac{1-\beta^2}{\beta^4}\left\{\frac{T_m}{E_d} - 1 - \beta^2\ln\left(\frac{T_m}{E_d}\right) + \pi\frac{Ze^2}{\hbar c}\beta\left[2\left(\frac{T_m}{E_d}\right)^{1/2} - \ln\left(\frac{T_m}{E_d}\right) - 2\right]\right\}$$



$$T_{max} = \frac{2ME(E+2mc^2)}{(M+m)^2c^2 + 2ME}$$

Due to low atom mass

T_{max} (^{1}H)= 188 eV @ 80kV

E_d (H in C-H) = 6.1 eV

Atomic charge Z

Atomic mass M

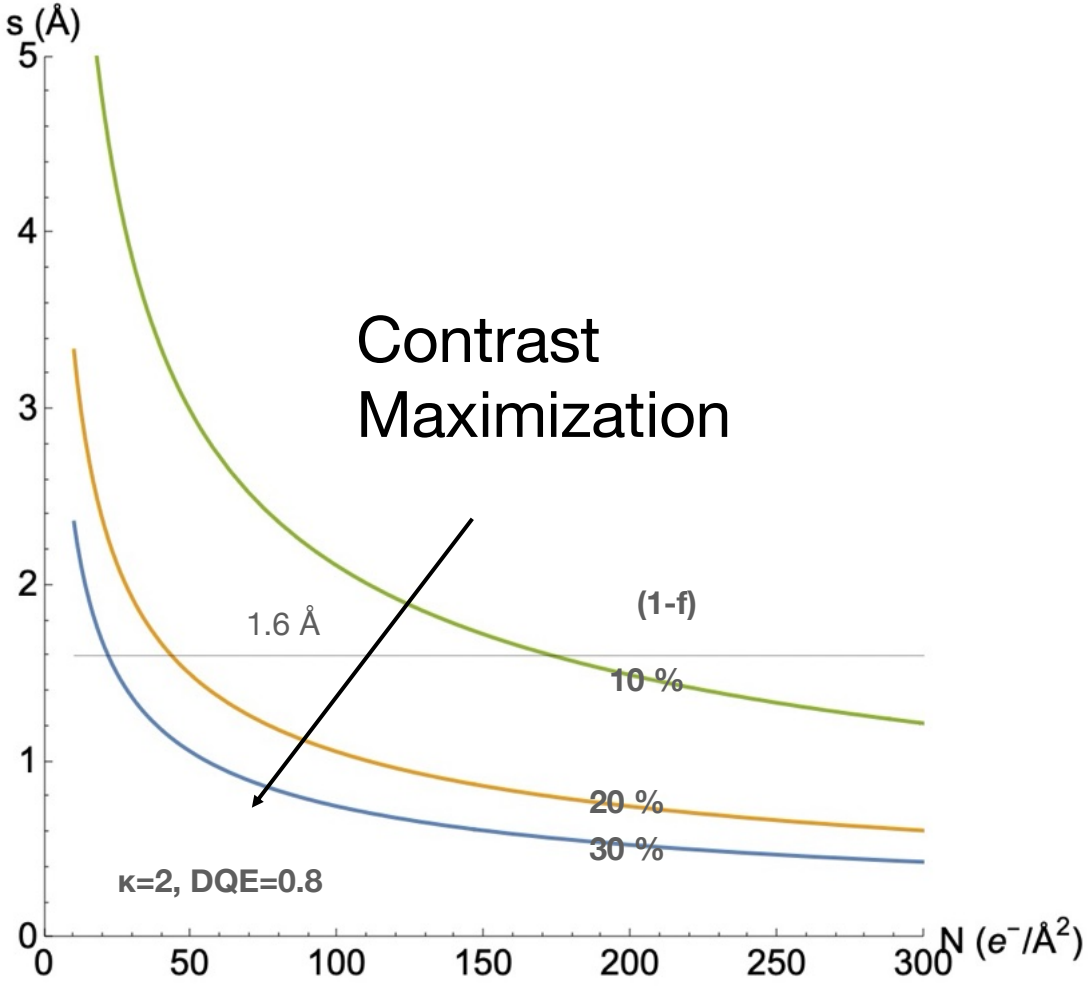
Electron velocity $\beta = v_e/c$

Electron energy E

McKinley, Feshbach, Phys. Rev. 74 (1948) 1759

Slide data courtesy of U. Kaiser, Ulm University

Dose-limited resolution - Shot Noise Case



Back-of-the-envelope pixel signal/noise calculation following the **Rose criterion**:

$$s = \frac{\kappa}{(1-f)\sqrt{N}\sqrt{\mathrm{DQE}}}$$

s: resolution defining element

κ: detection threshold for the identification of an image feature in an image element of size s^2 (threshold SNR for detection)

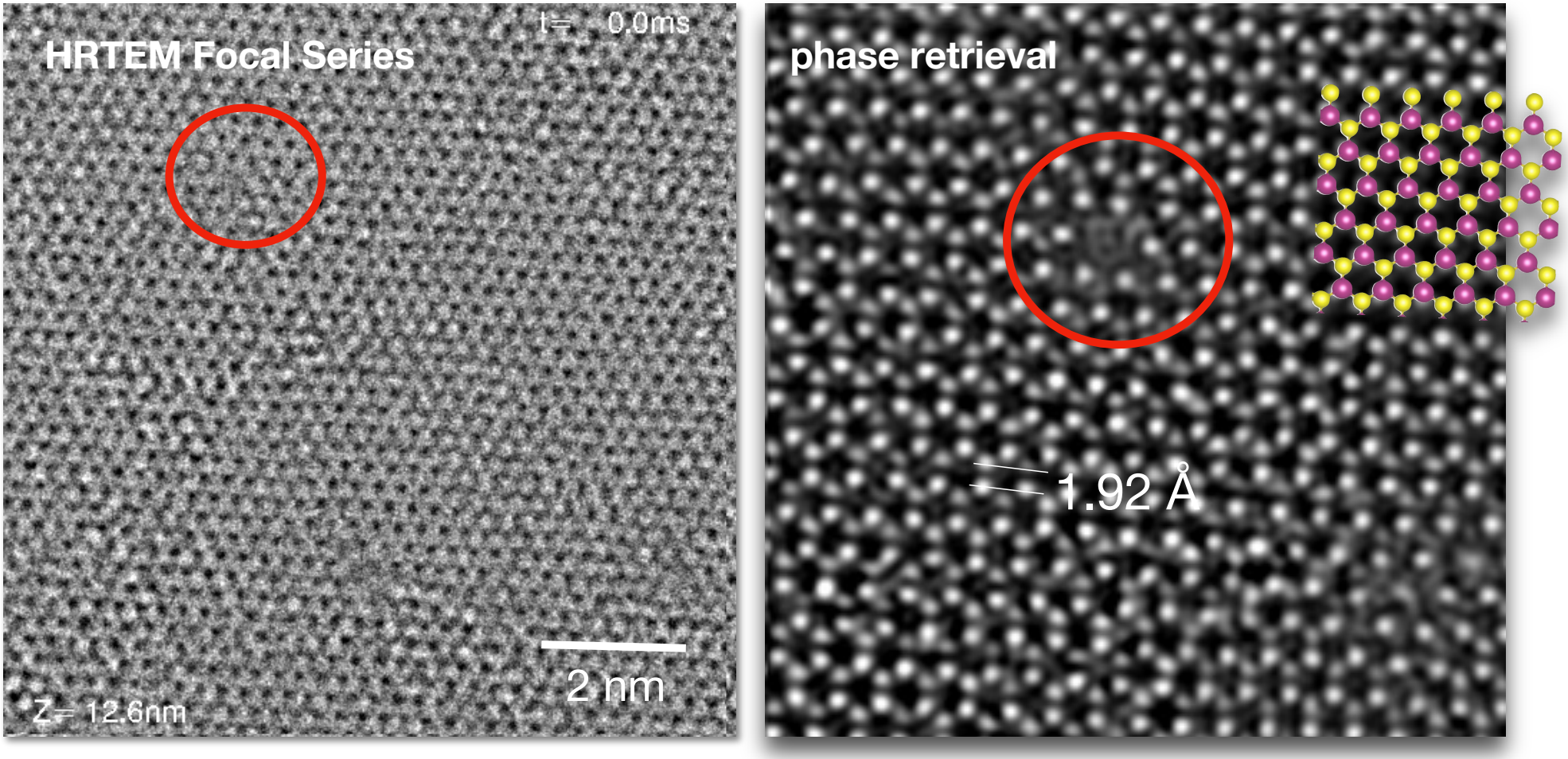
f: fraction of electrons that contribute to the background, (1-f) is the signal

N: total dose, in e^- per A^2

DQE: detective quantum efficiency

Measurement Strategies :: Maximum detection efficiency

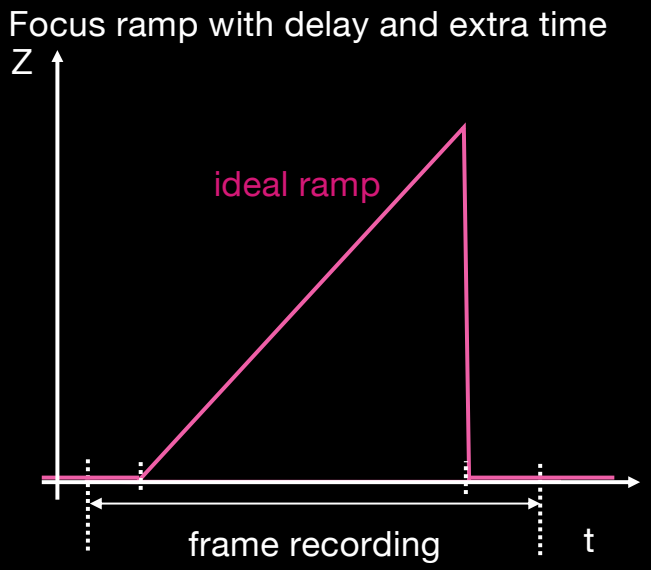
WS_2 @ 80 kV

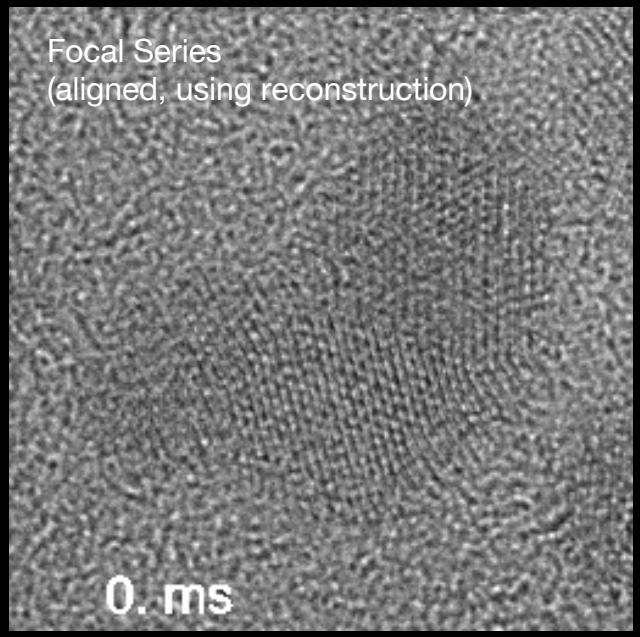


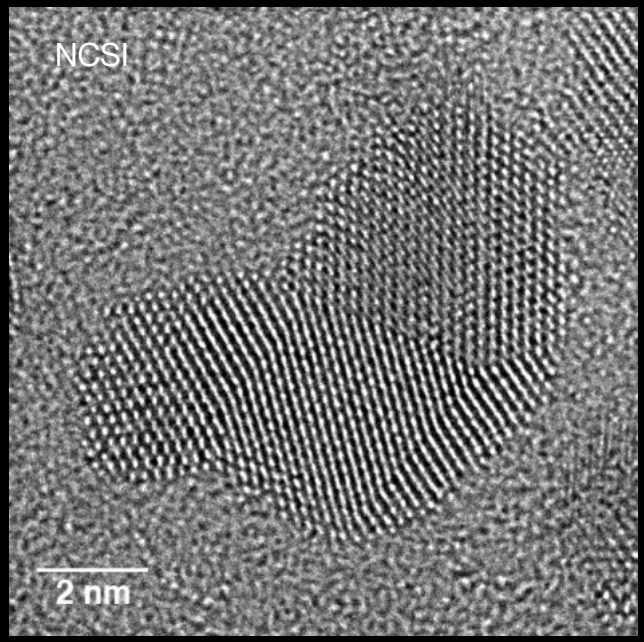
Left: Dynamic focal series in a Themis-Z, Gatan OneView,
400 images in 4 s while focus ramps from +12 nm to -12 nm .

Fluence 1000 e/A^2

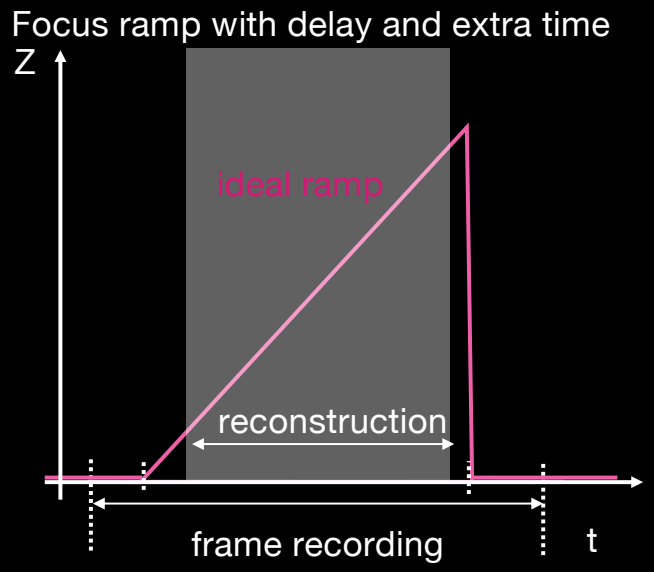
Sample: A. Cohen, A. Ismach (TAU)

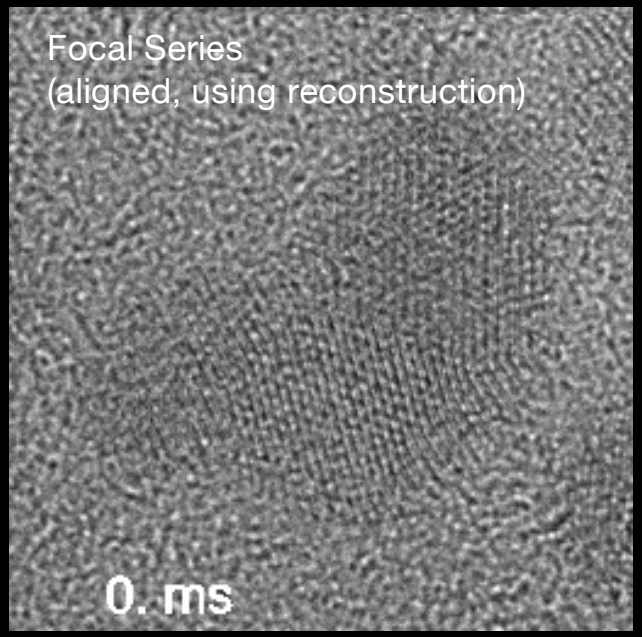


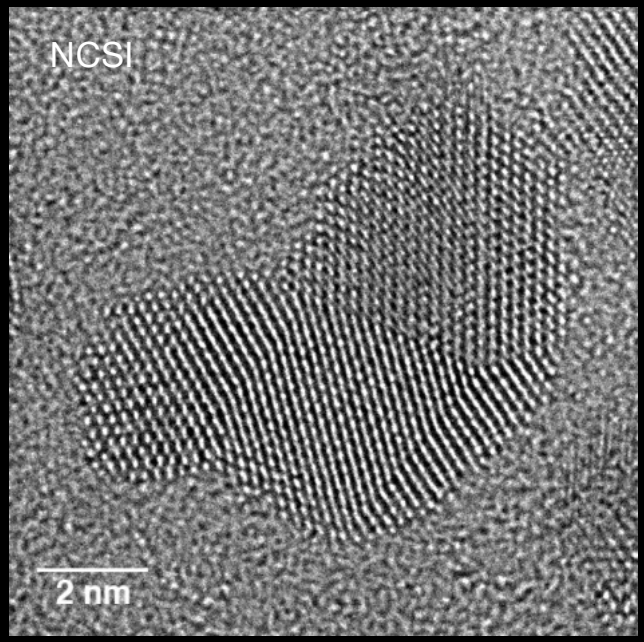


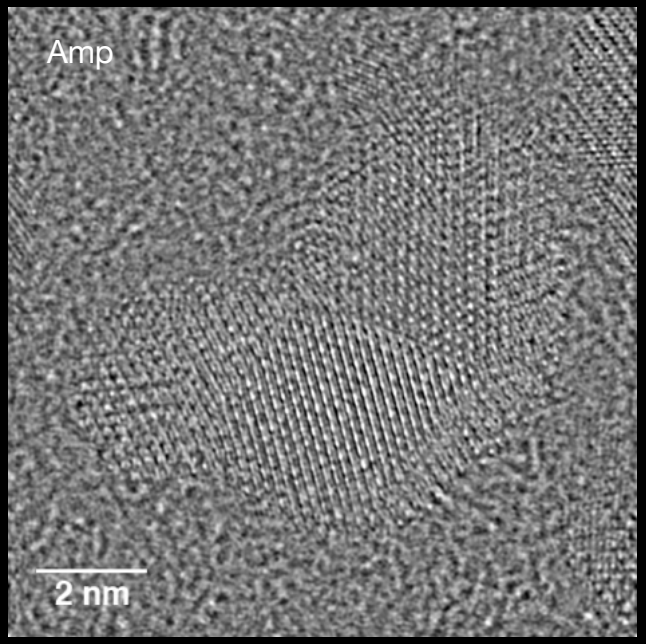


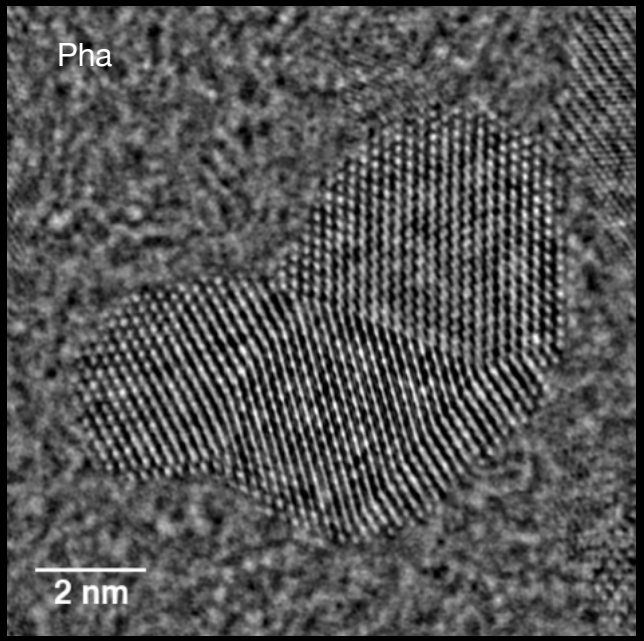
Themis-Z, AuPd, Gatan OneView, 512x512, 5ms per frame original recording, 2624 frames, 13 s total recording time
16 frame boxcar average, N=164



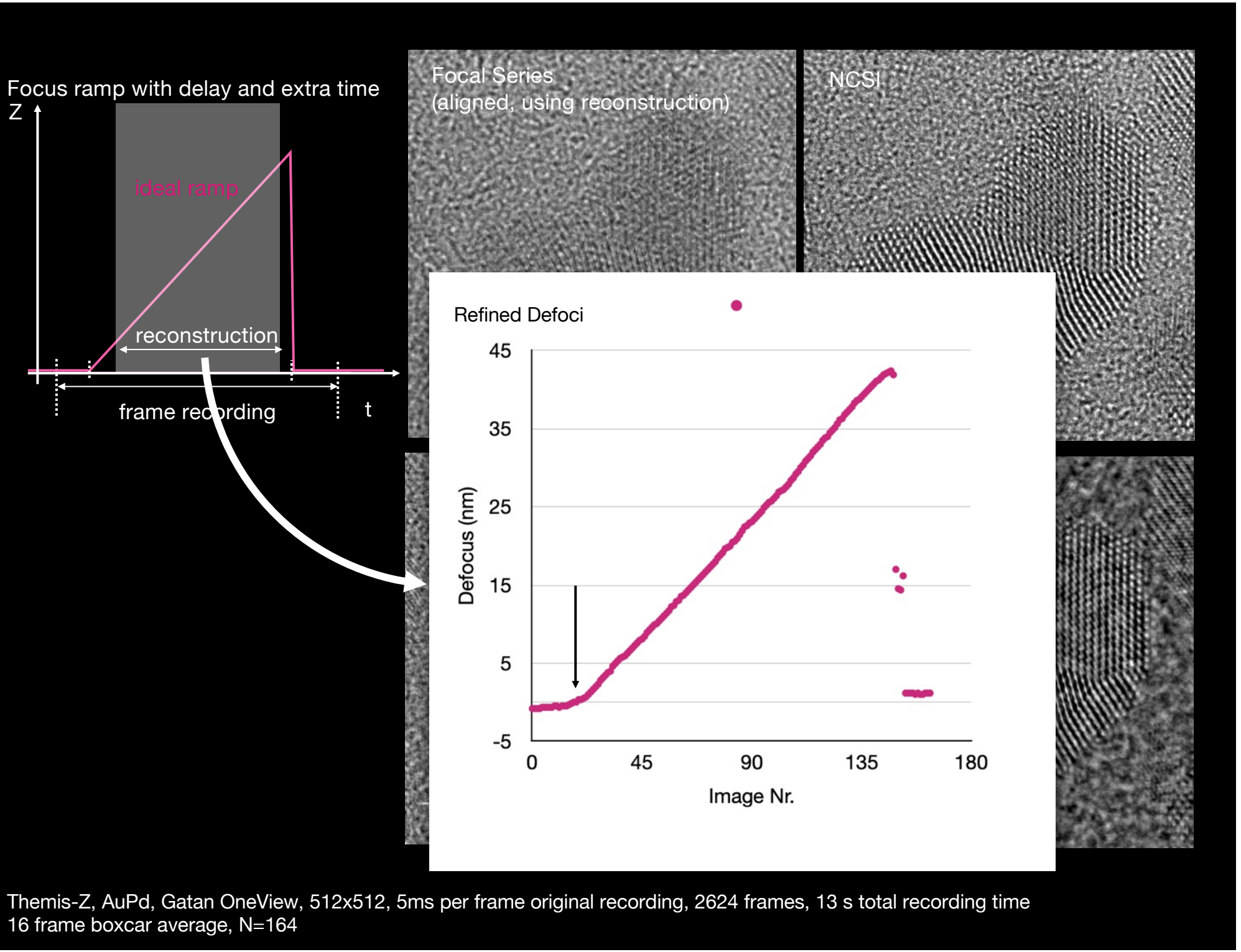






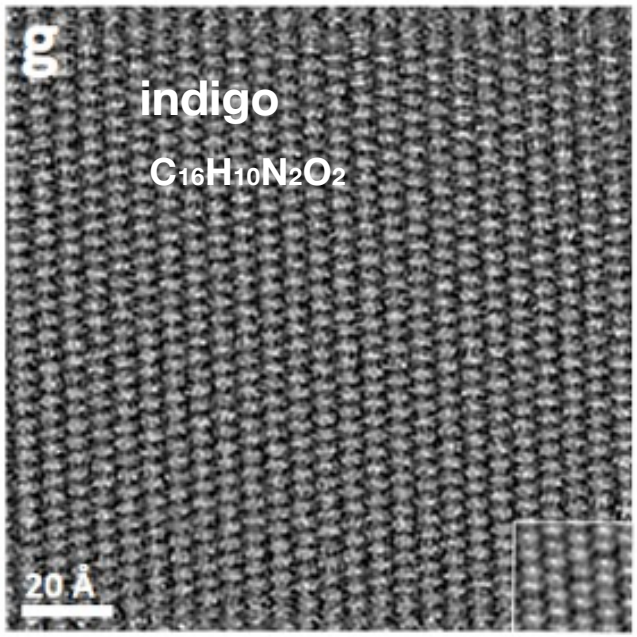


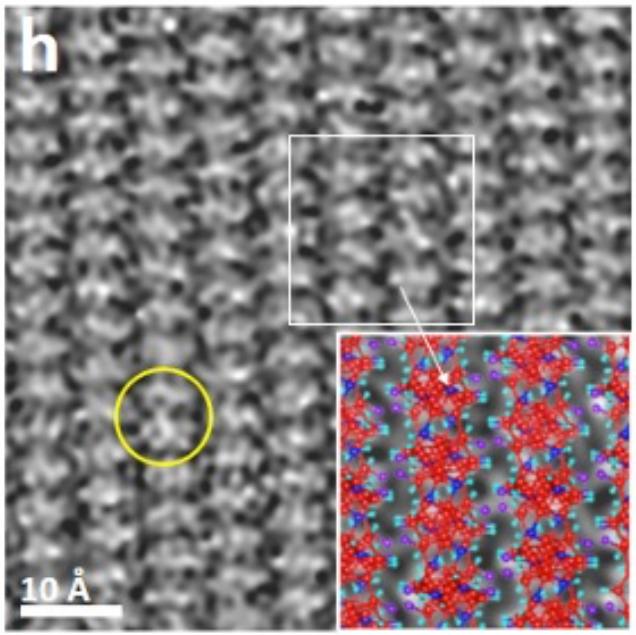
Themis-Z, AuPd, Gatan OneView, 512x512, 5ms per frame original recording, 2624 frames, 13 s total recording time
16 frame boxcar average, N=164

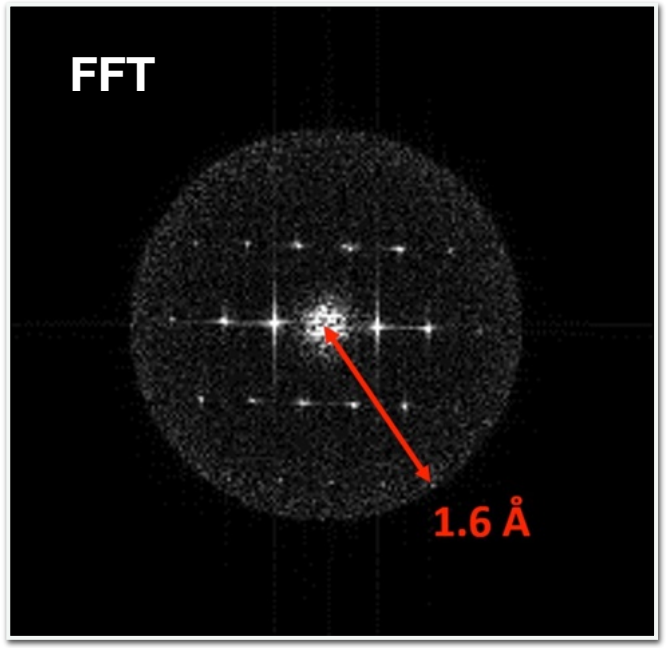


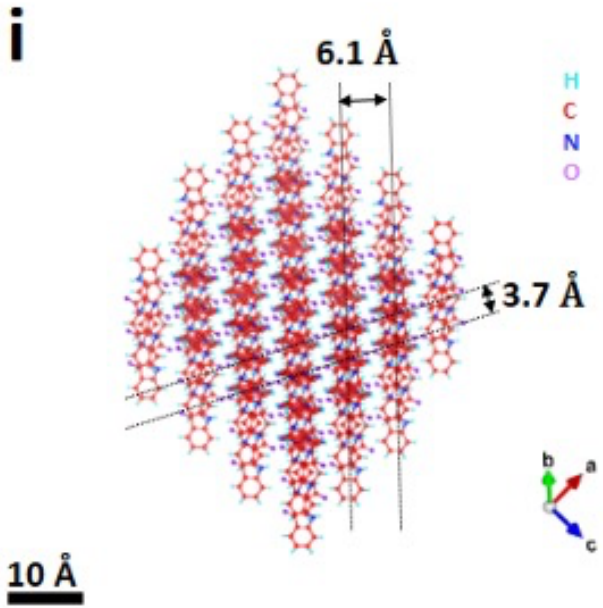
Themis-Z, AuPd, Gatan OneView, 512x512, 5ms per frame original recording, 2624 frames, 13 s total recording time
16 frame boxcar average, N=164

Maximum detection efficiency in Cryo-Microscopy









Focal Series Reconstruction

KRIOS DATA/K3

not aberration-corrected

C_S = 3.7 mm

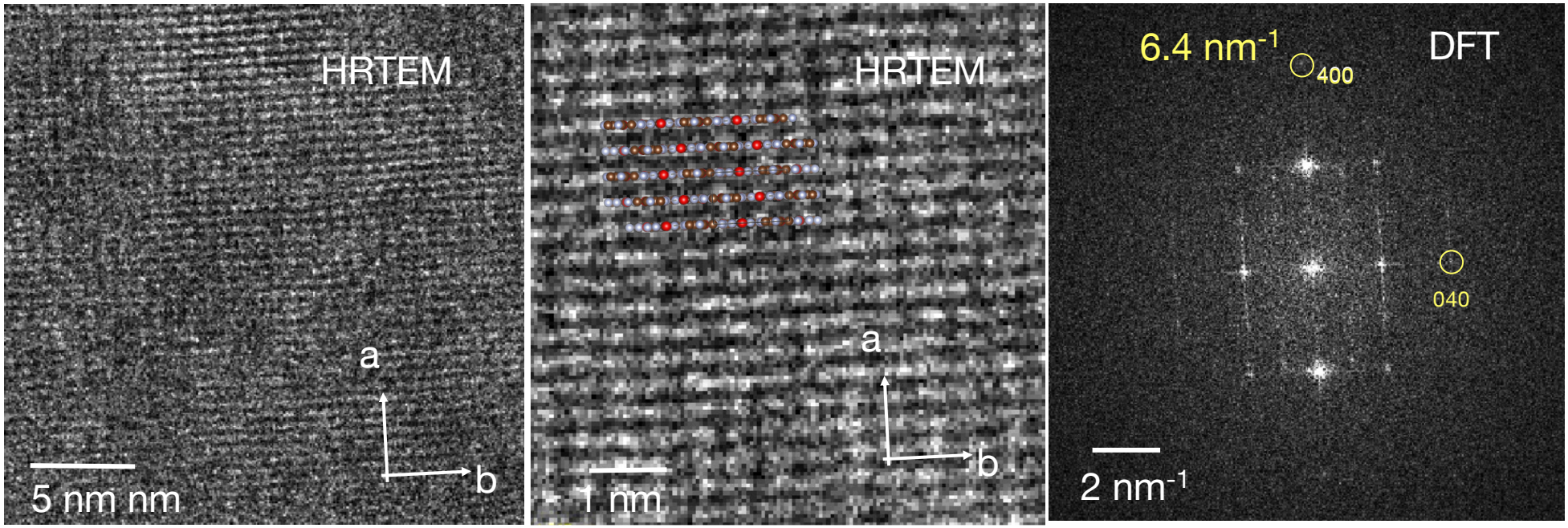
Z= -250 to -50 nm

300 images in 4 s

Total fluence: 112 $e^-/Å^2$.

Biran et al., Advanced Materials 34 (26), 2202088

The aberration-corrected case

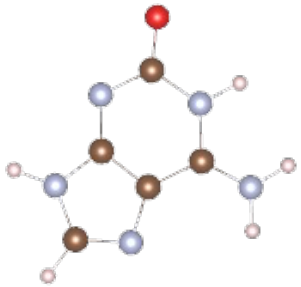


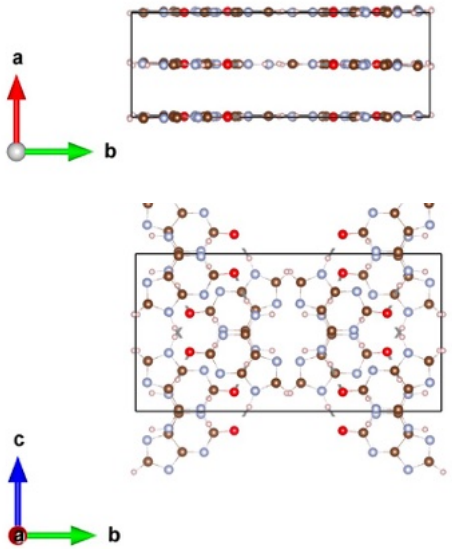
Energy-filtered **aberration-corrected** bright-field HRTEM at 200 kV, Dectris ELA

Series of 50 frames, 50 ms per frame, sum of 7 frames #34-40
Elastic channel fluence for sum of 7 frames: 21 e-/Å^2

Close to NCSI focus, -10 nm

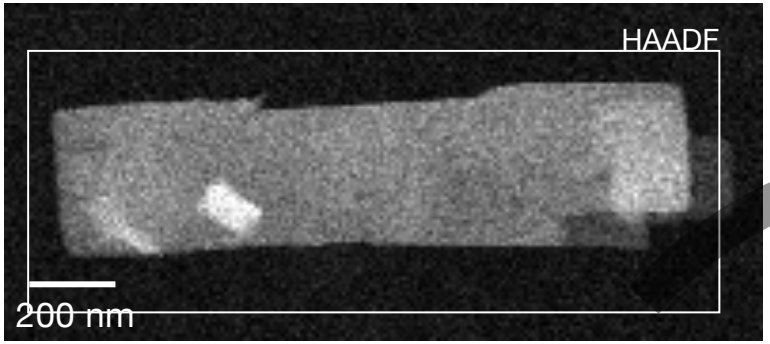
Isoguanine



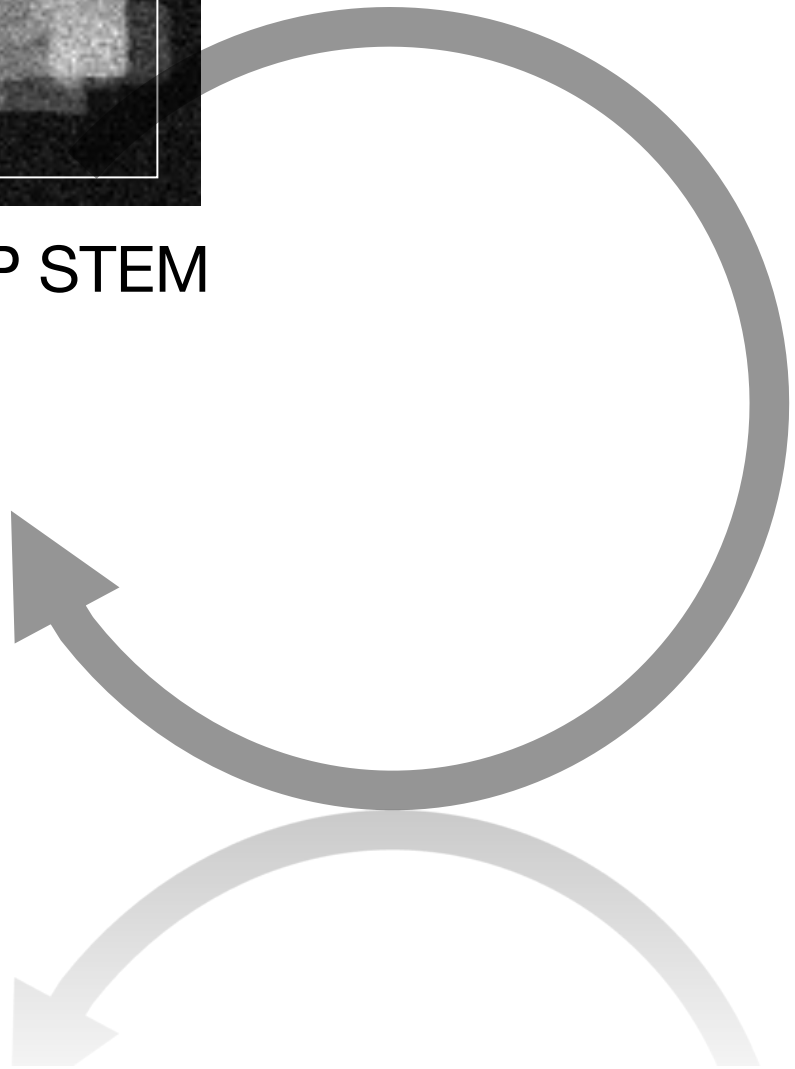


Mode Switching: Low-dose aberration-corrected TEM

When the lattice is gone before you focus …

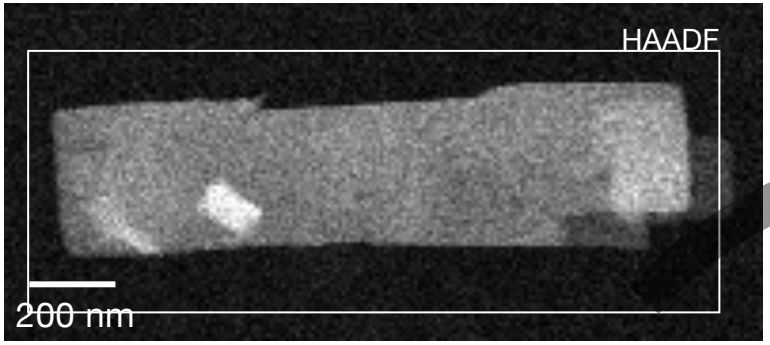


Search and focus: uP STEM

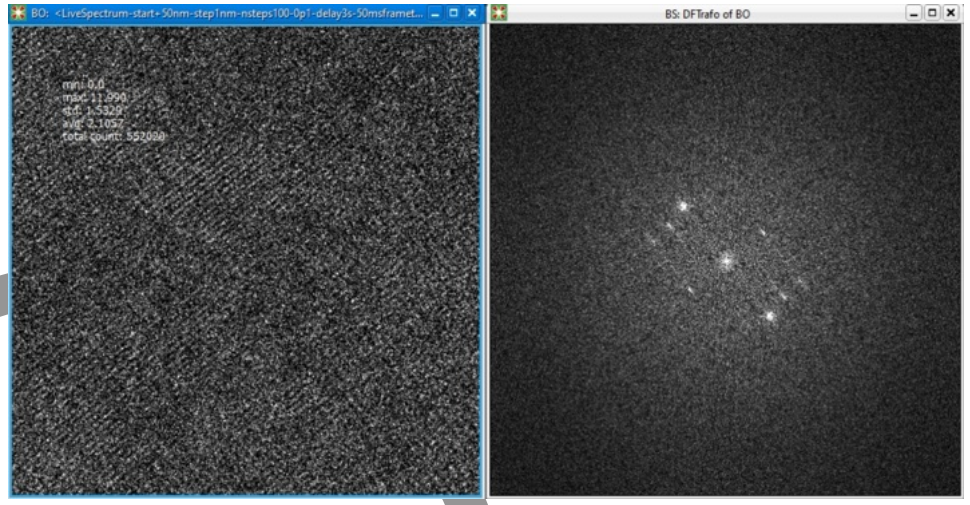


Mode Switching: Low-dose aberration-corrected TEM

When the lattice is gone before you focus …



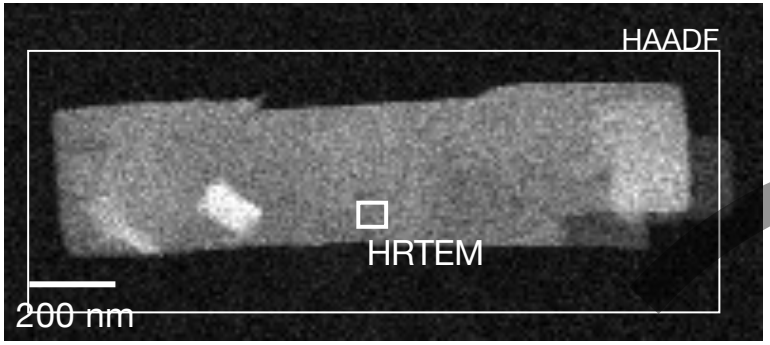
Search and focus: uP STEM



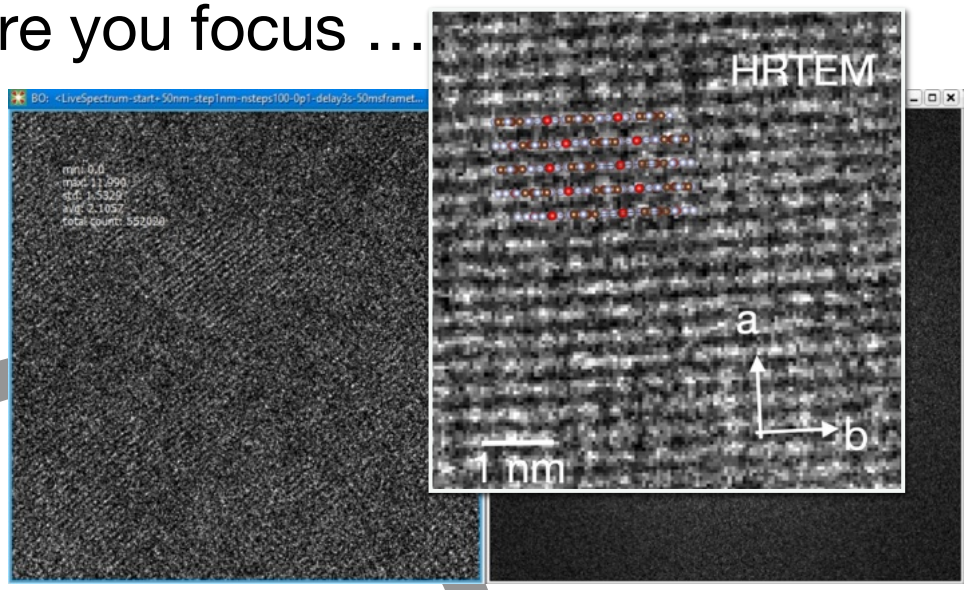
AC-HRTEM (low dose 21e/Å^2)

Mode Switching: Low-dose aberration-corrected TEM

When the lattice is gone before you focus …



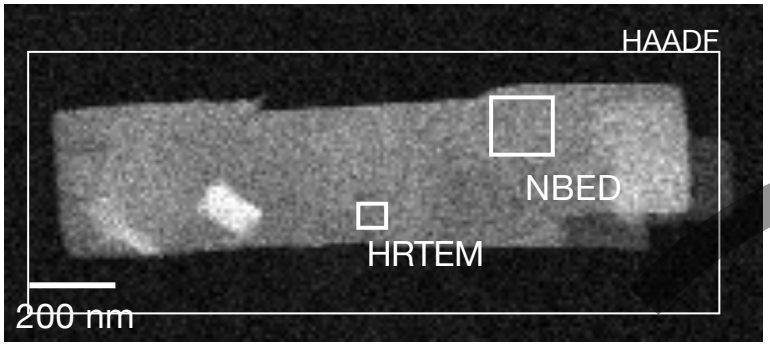
Search and focus: uP STEM



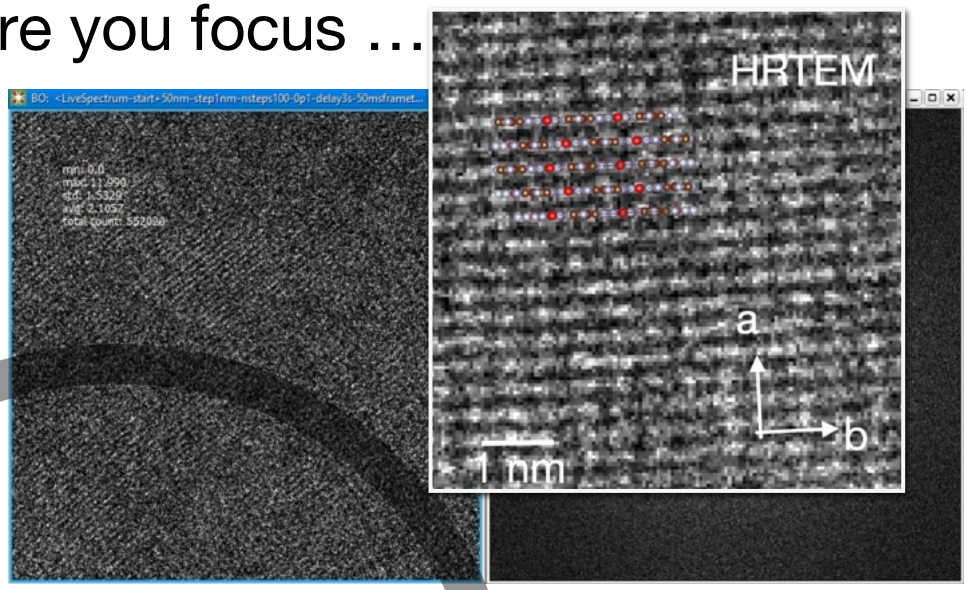
AC-HRTEM (low dose 21e/Å^2)

Mode Switching: Low-dose aberration-corrected TEM

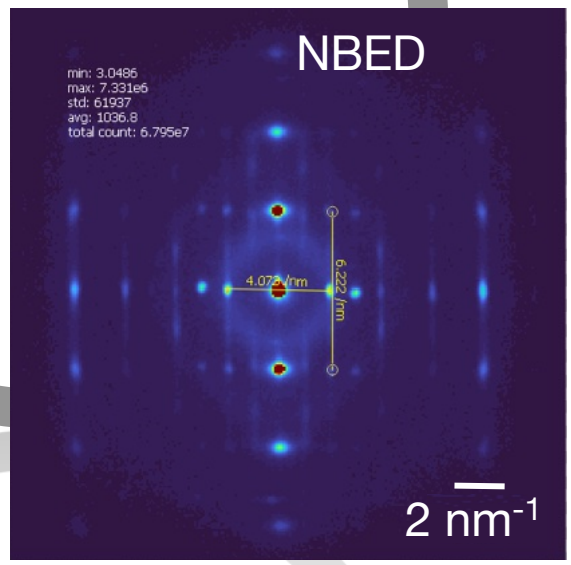
When the lattice is gone before you focus …



Search and focus: uP STEM



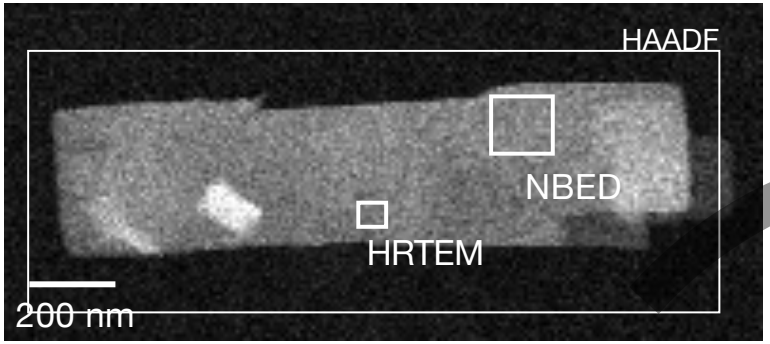
AC-HRTEM (low dose 21e/Å^2)



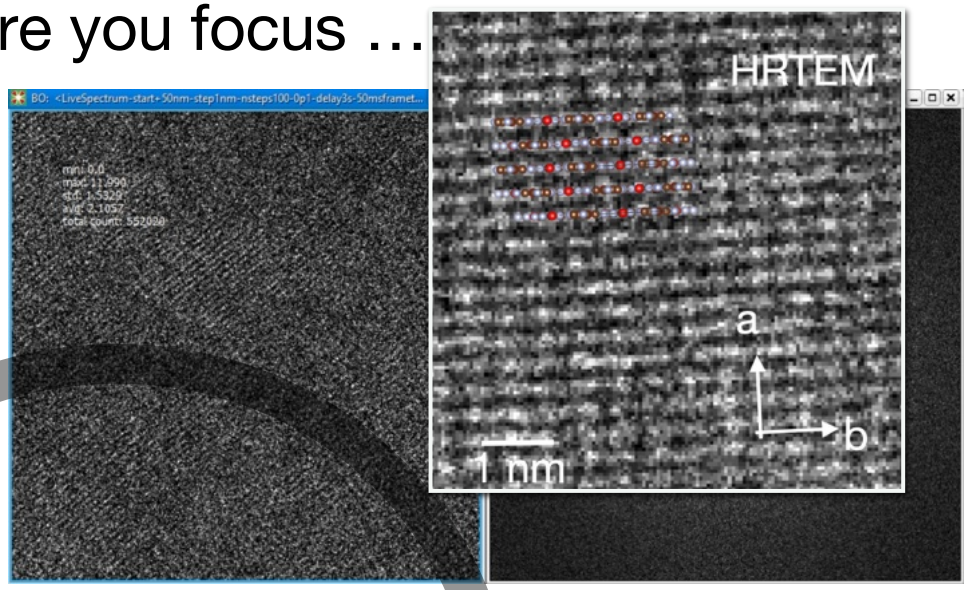
uP 4D-STEM

Mode Switching: Low-dose aberration-corrected TEM

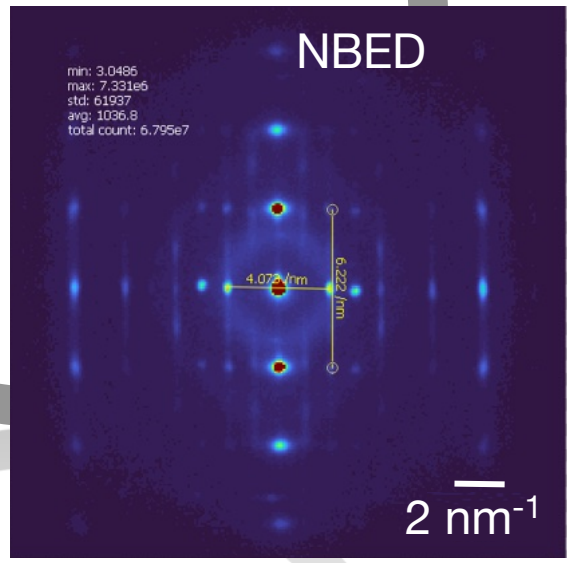
When the lattice is gone before you focus …



Search and focus: uP STEM



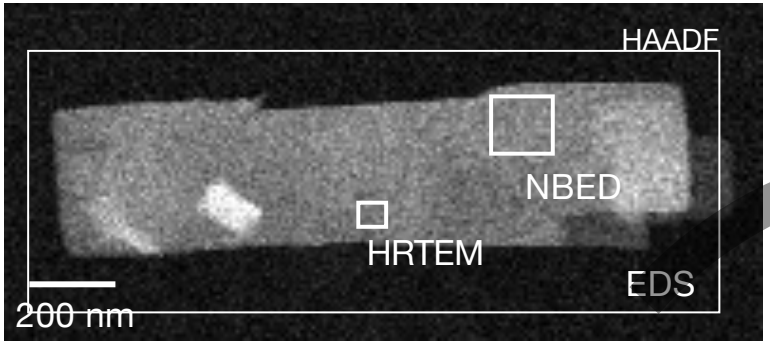
AC-HRTEM (low dose 21e/Å^2)



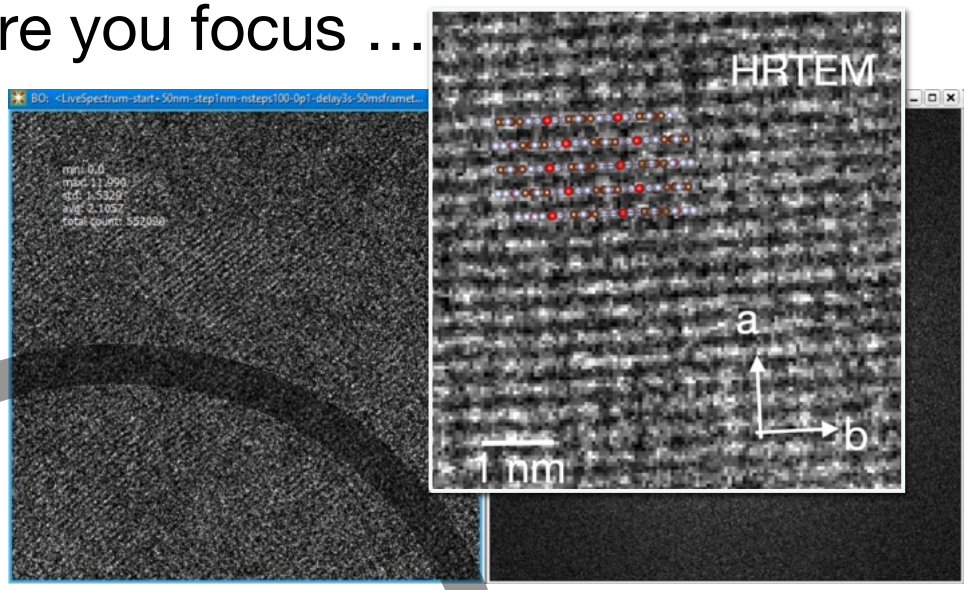
uP 4D-STEM at 4 pA

Mode Switching: Low-dose aberration-corrected TEM

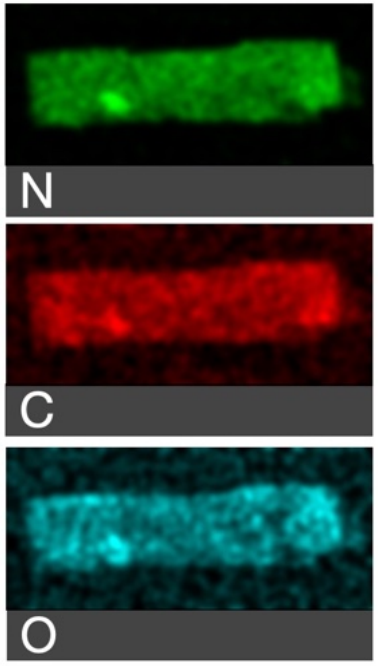
When the lattice is gone before you focus …



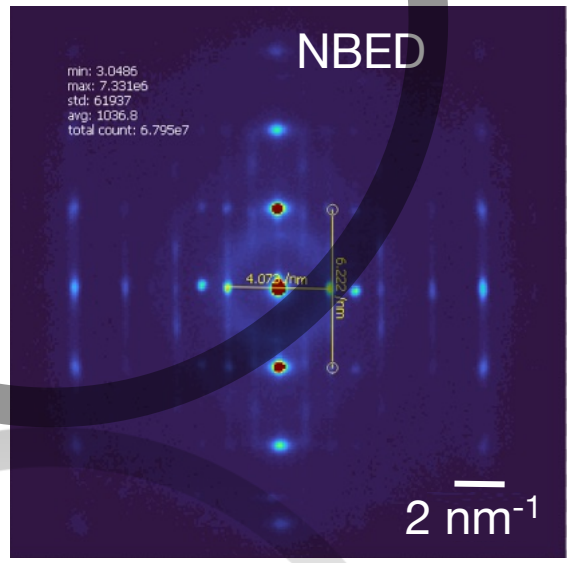
Search and focus: uP STEM



AC-HRTEM (low dose 21e/Å^2)



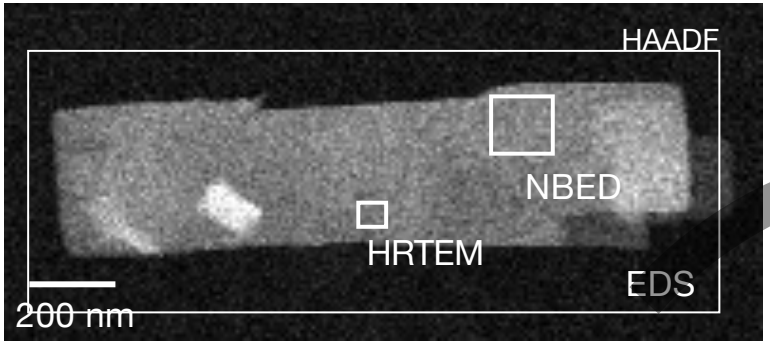
uP STEM EDS at 50 pA



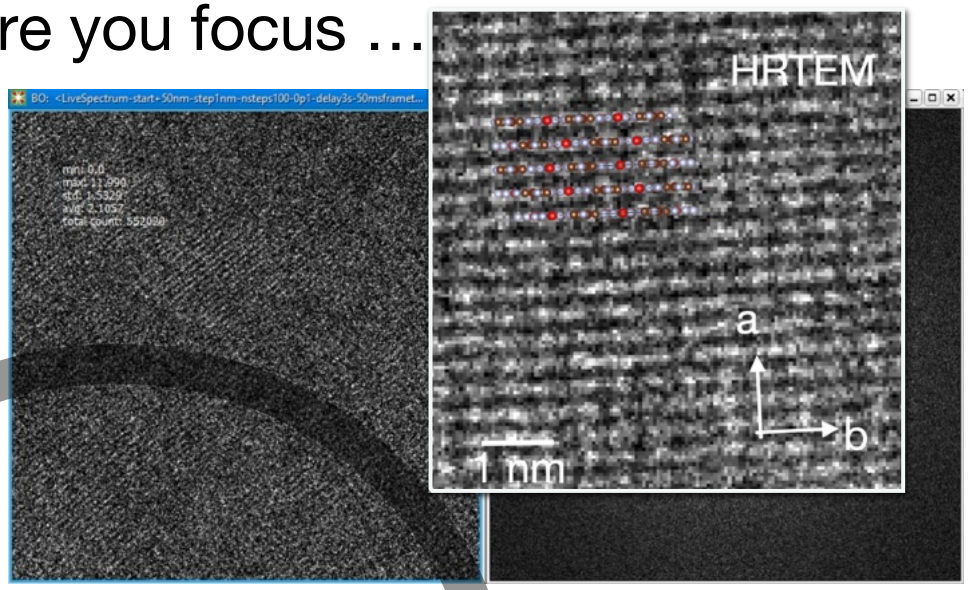
uP 4D-STEM at 4 pA

Mode Switching: Low-dose aberration-corrected TEM

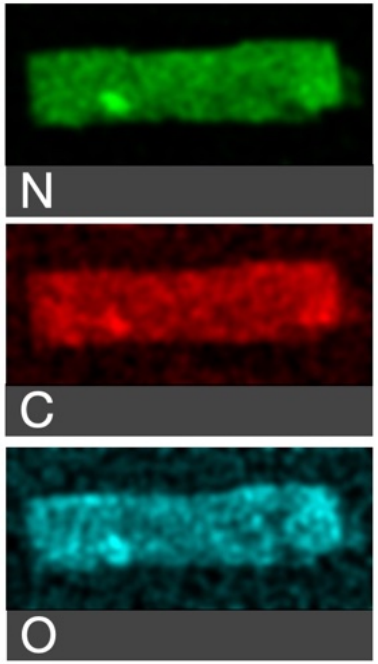
When the lattice is gone before you focus …



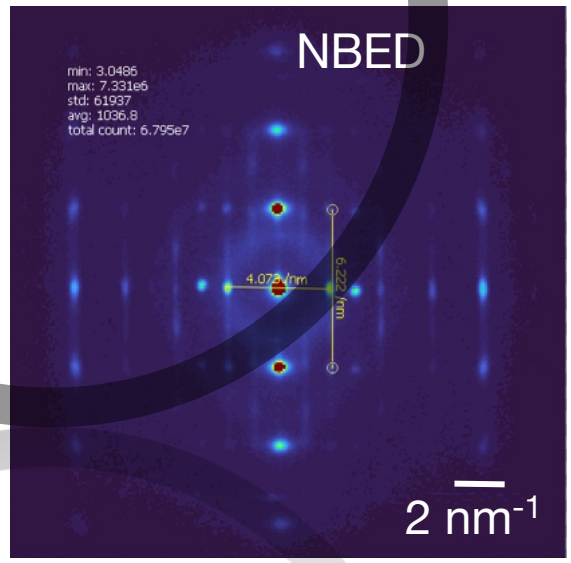
Search and focus: uP STEM



AC-HRTEM (low dose 21e/Å^2)



uP STEM EDS at 50 pA



uP 4D-STEM at 4 pA

Total recording time approx. 10 min

Achieving the objective

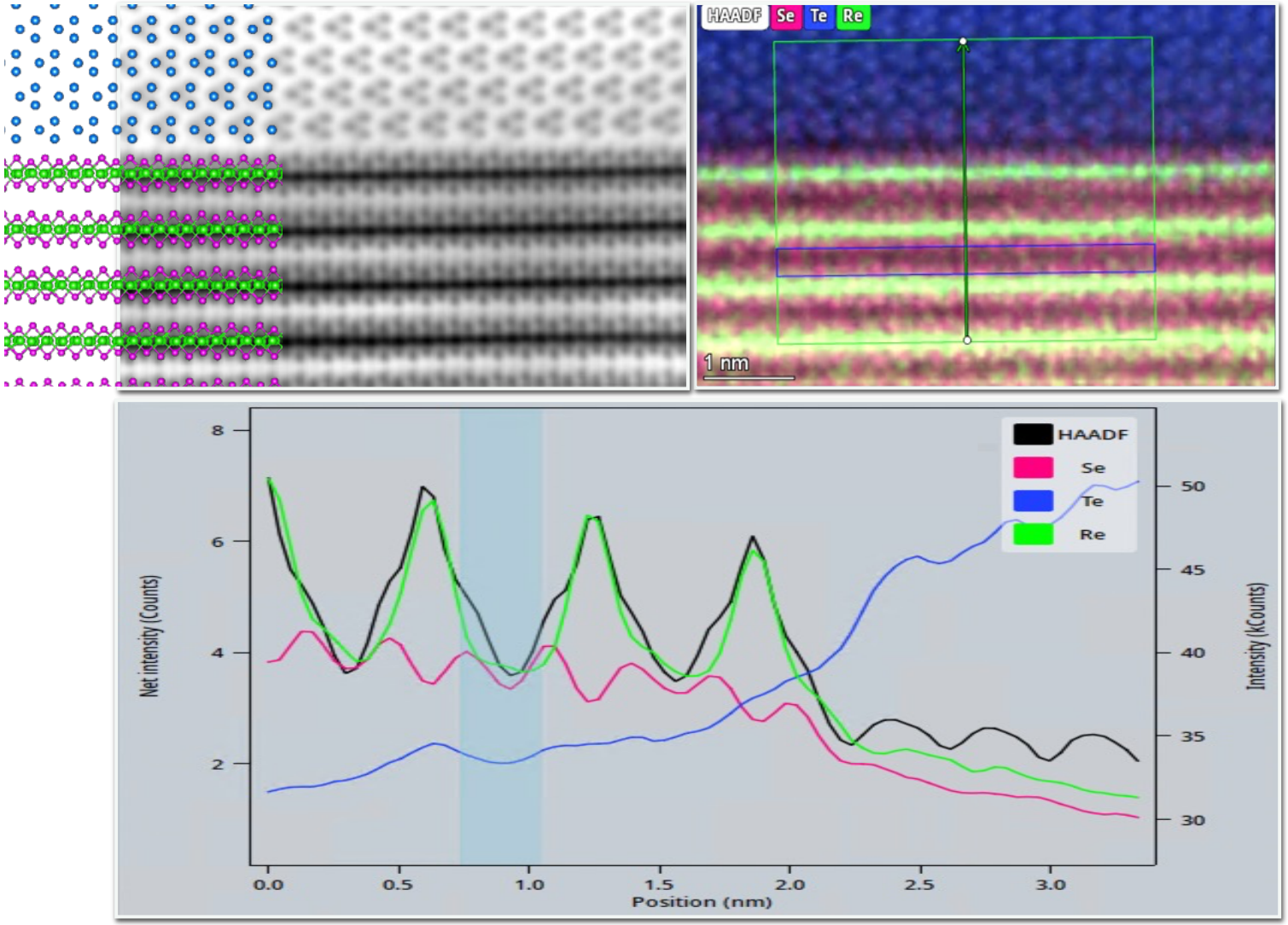
Which information do you seek and can you get it from your sample?

- optimize conditions
- learn about potential artefacts
- learn when is your result as expected

Working with simulation helps a lot!

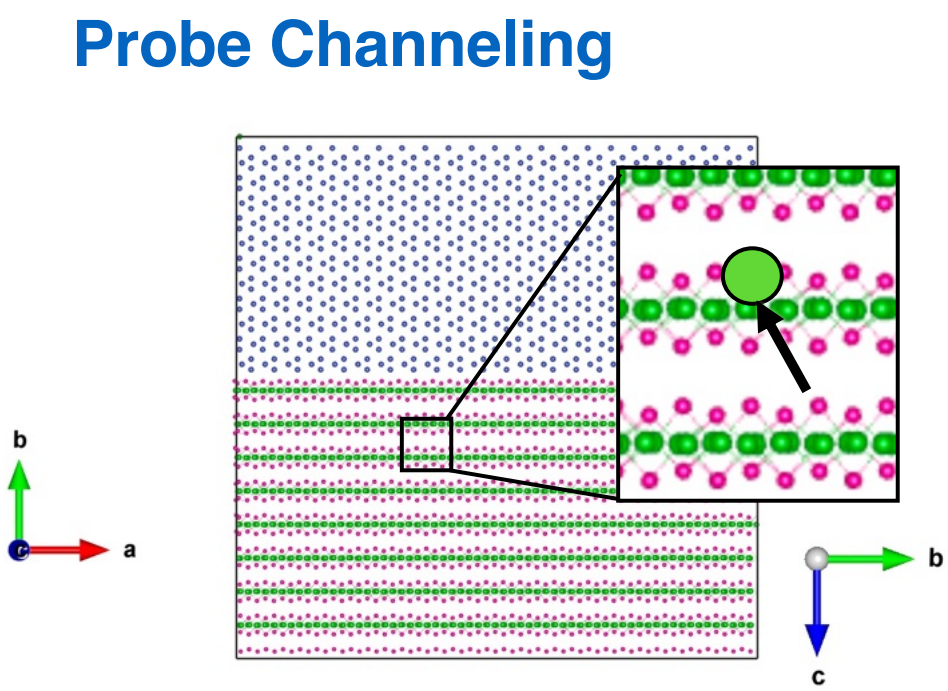
Challenge: Quantification

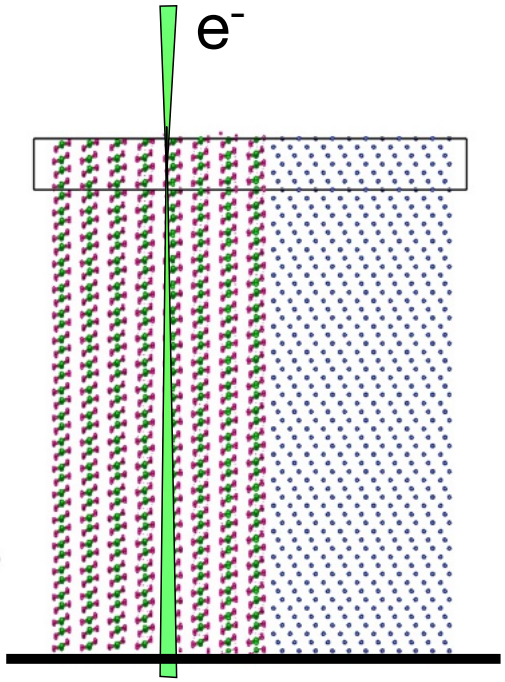
Example: EDS profiles of Te on $ReSe_2$



Modulation in the analytical signal around atomically sharp interfaces is not necessarily atomically sharp.

Probe Channeling

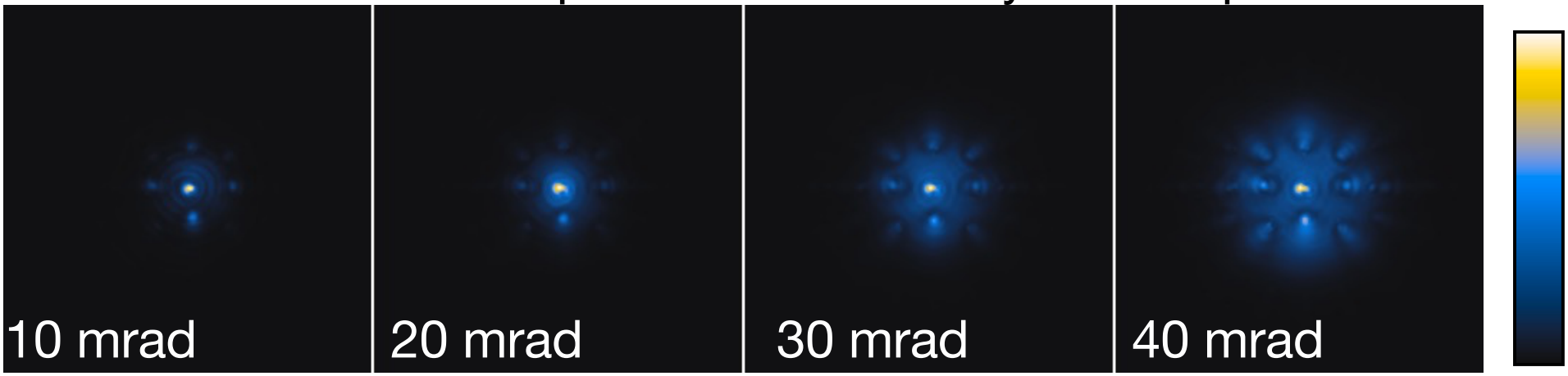




elastic channeling multislice simulation (Dr Probe)

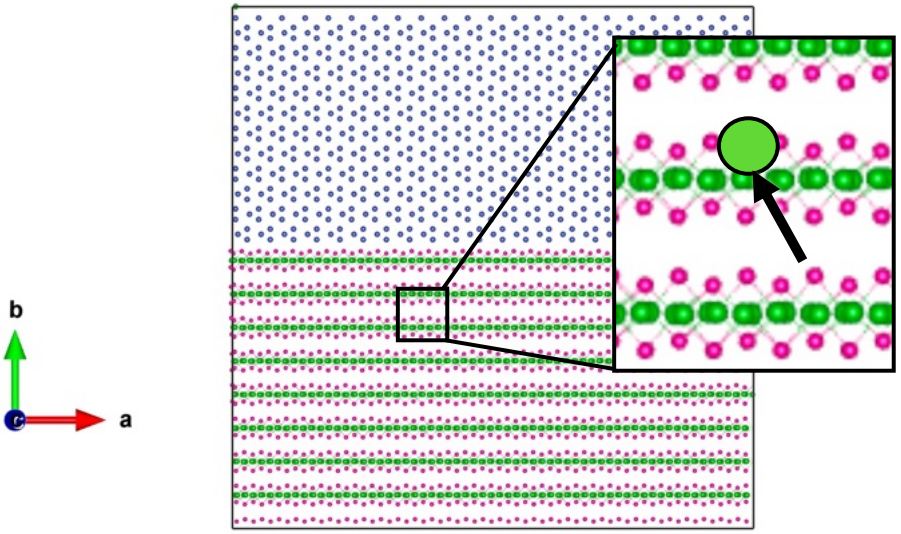
exit plane

on Re, t=10 nm exit plane wave intensity in real space

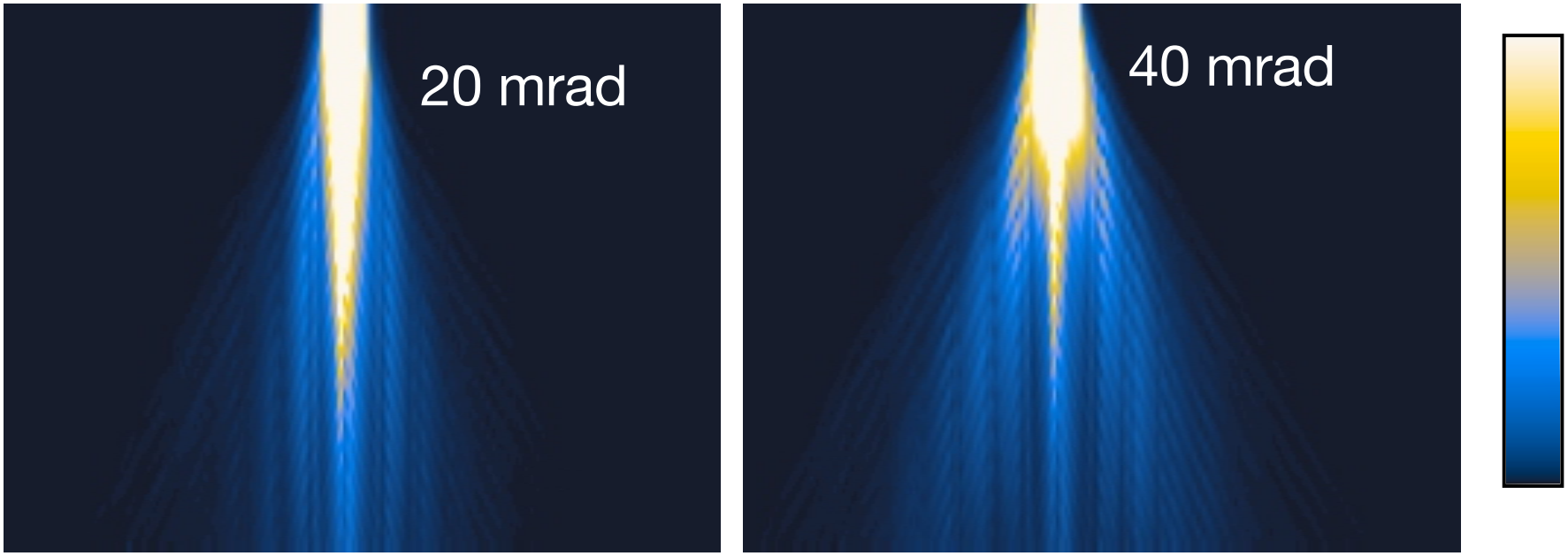


high convergence angles are not favourable

Probe Channeling



channeling wave intensity
in cross sectional plane
on Re, t=20 nm

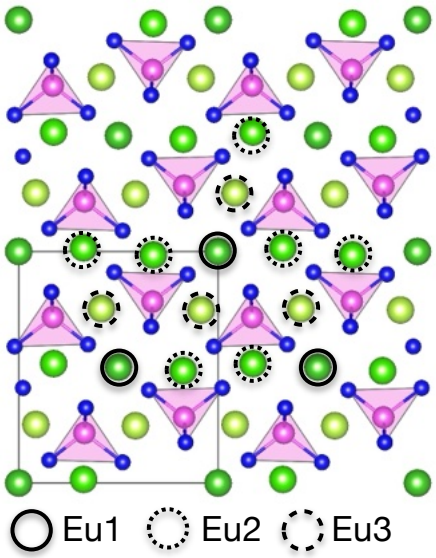


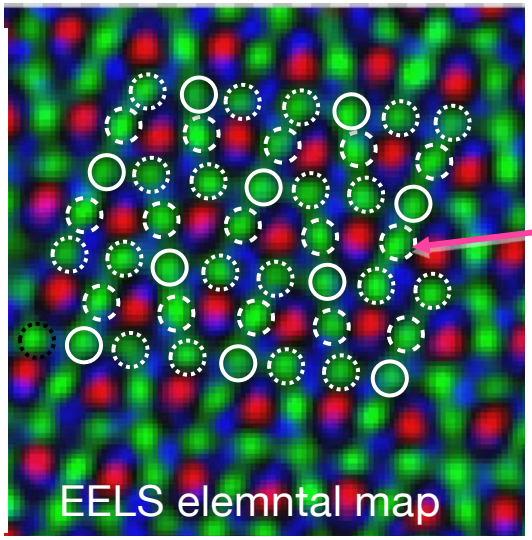
5 Å

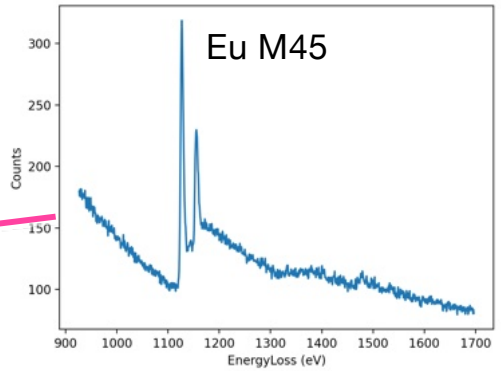
high convergence angles are not favourable unless samples are ultimately thin

Atomic-column EELS

Axion insulator $Eu_5In_2As_6$: A true Zintl-Phase



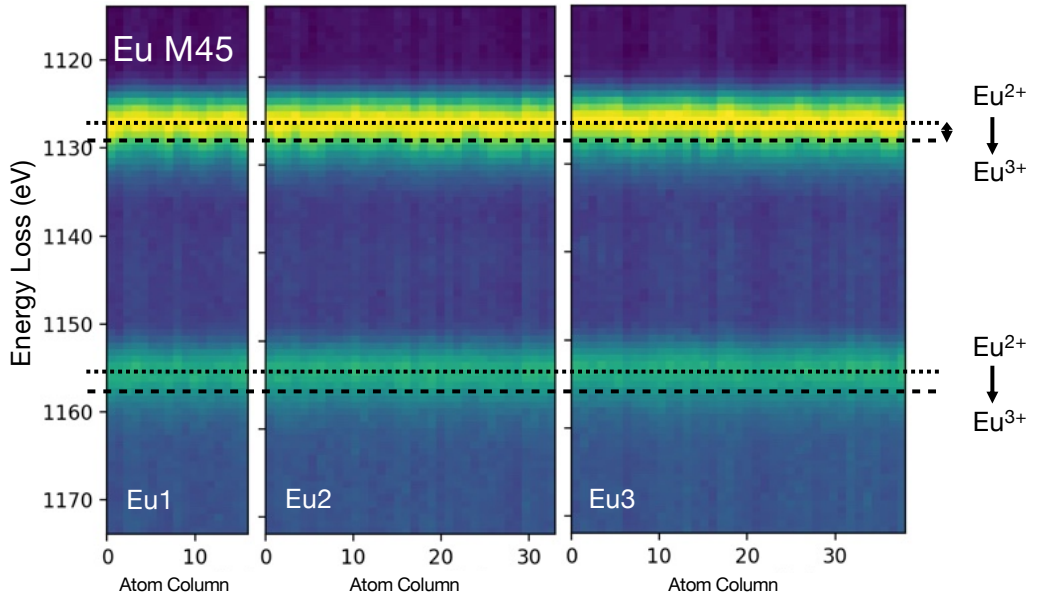




Eu M45

In M23

As L23



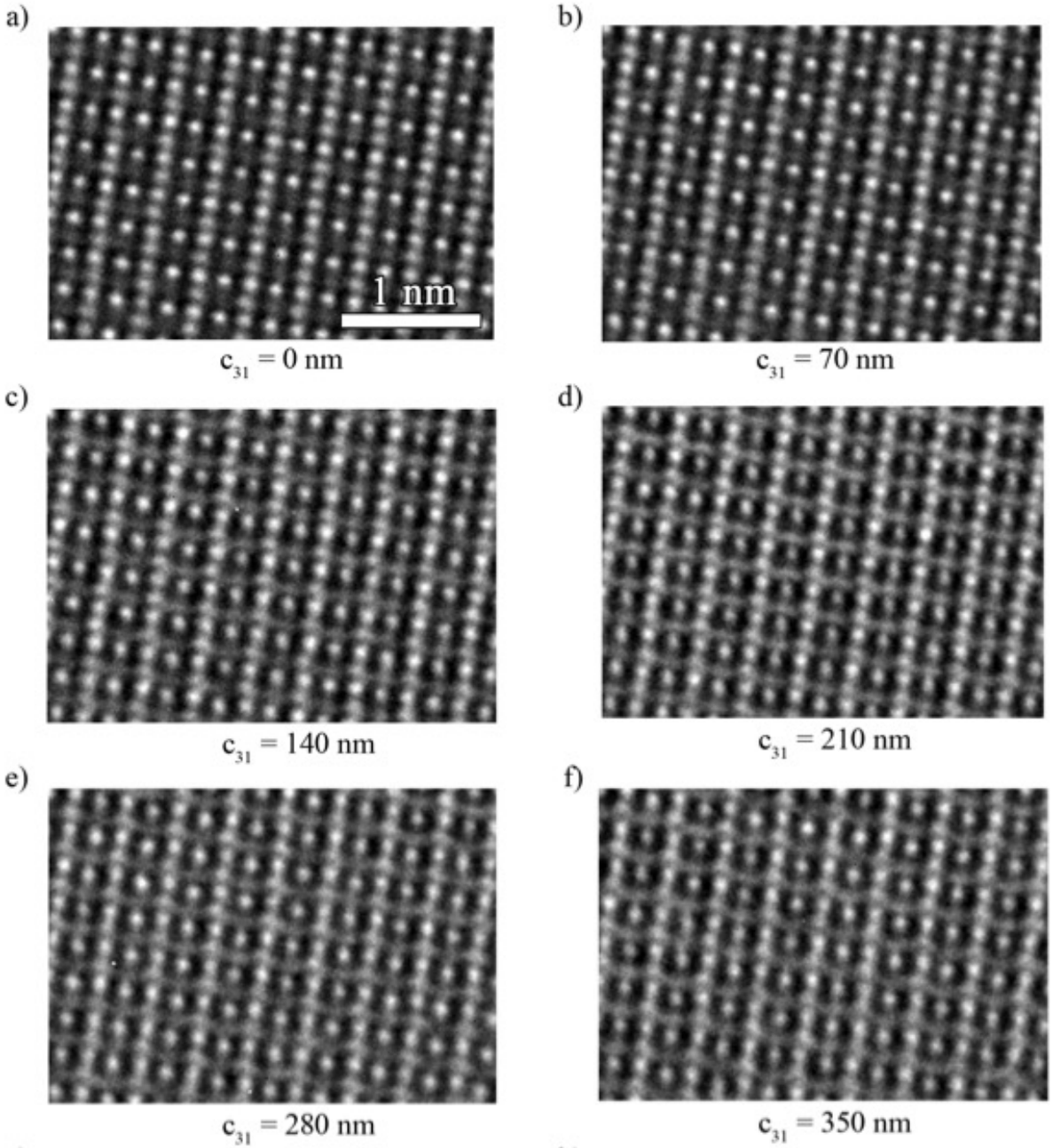
Sampling of multiple columns corresponding to the three octahedral Eu sites

=> identical electronic signature, no difference in oxidation state

Identical,

Divalent Eu Oxidation State

Effect of aberrations on images: axial coma



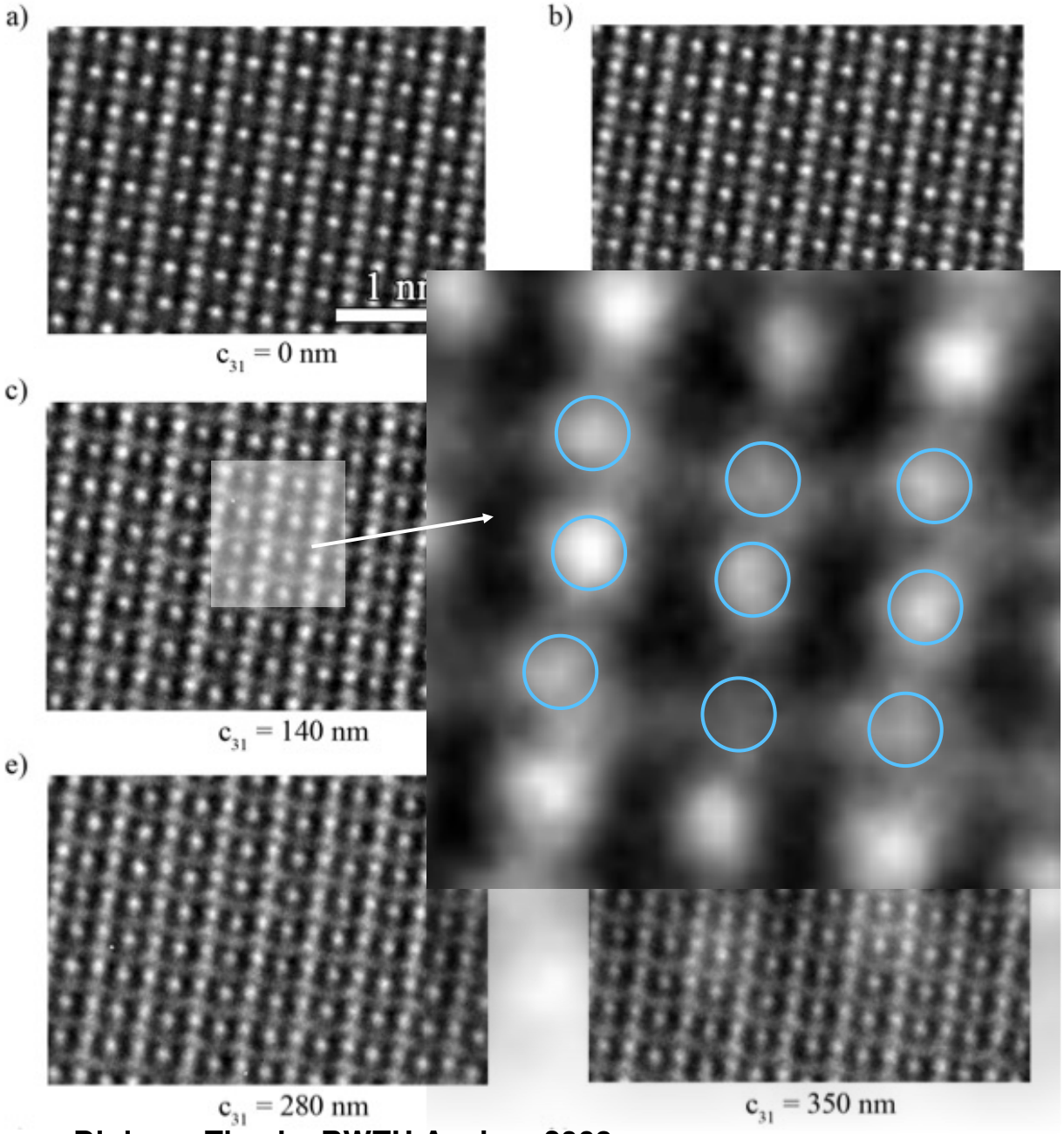
W. Hamm, Diploma Thesis, RWTH Aachen 2008

FEI Titan 300 kV

steps of ~π/2
at 80 pm

B_2 (=1/3 c_{31})

Effect of aberrations on images: axial coma



W. Hamm, Diploma Thesis, RWTH Aachen 2008

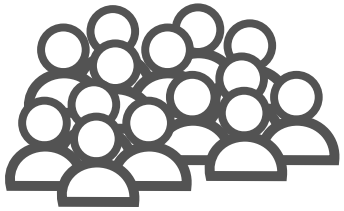
FEI Titan 300 kV

steps of ~π/2
at 80 pm

STO can look non-centrosymmetric!

The human factor: Daily routine to maintain reliability

Policy at a multi-user facility



Users - PIs/Groups

2022	2023	2024
84	82	83

Instruments in Service

2022	2023	2024
47	54	44

- No multiuser setups
- Maintain one basic alignment
- Maintain one default set of tunings

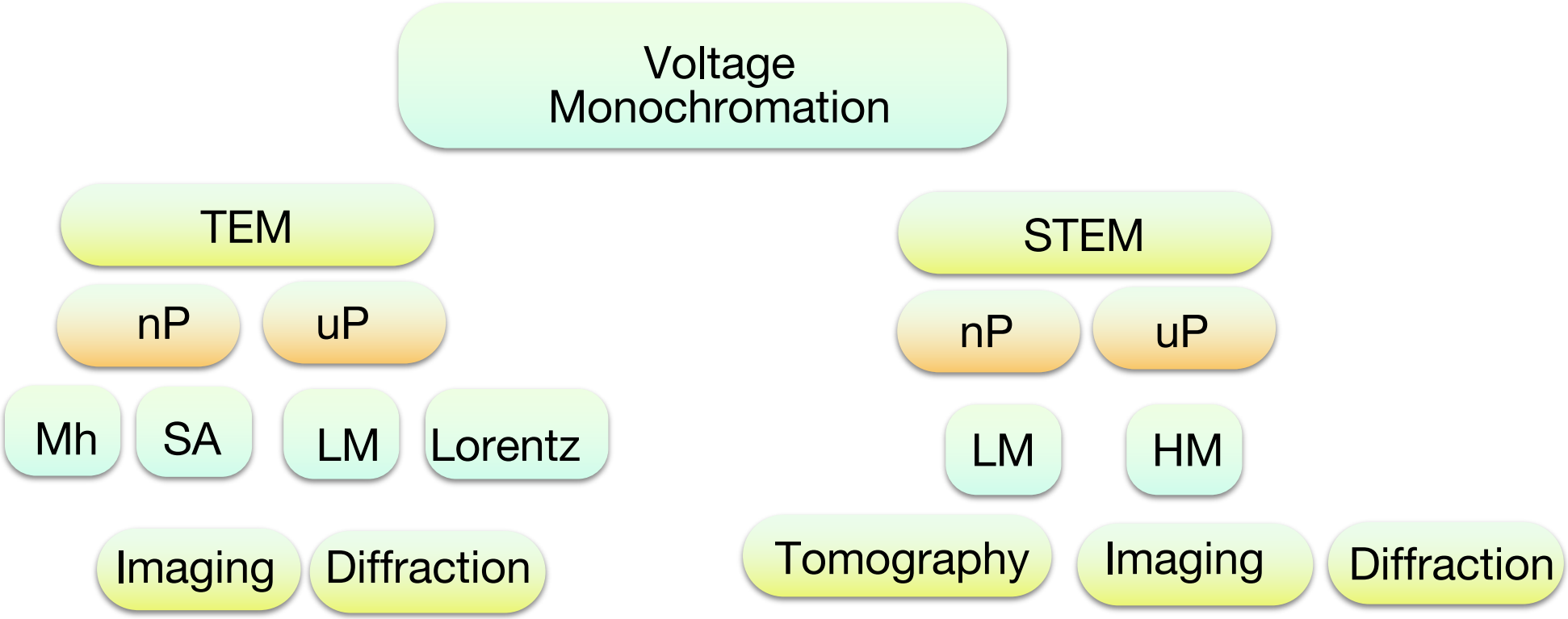
On the corrector side

- Tuning is stable
- Low maintenance
- Reproducible settings
- little issues with mode switching (on the side of the corrector)

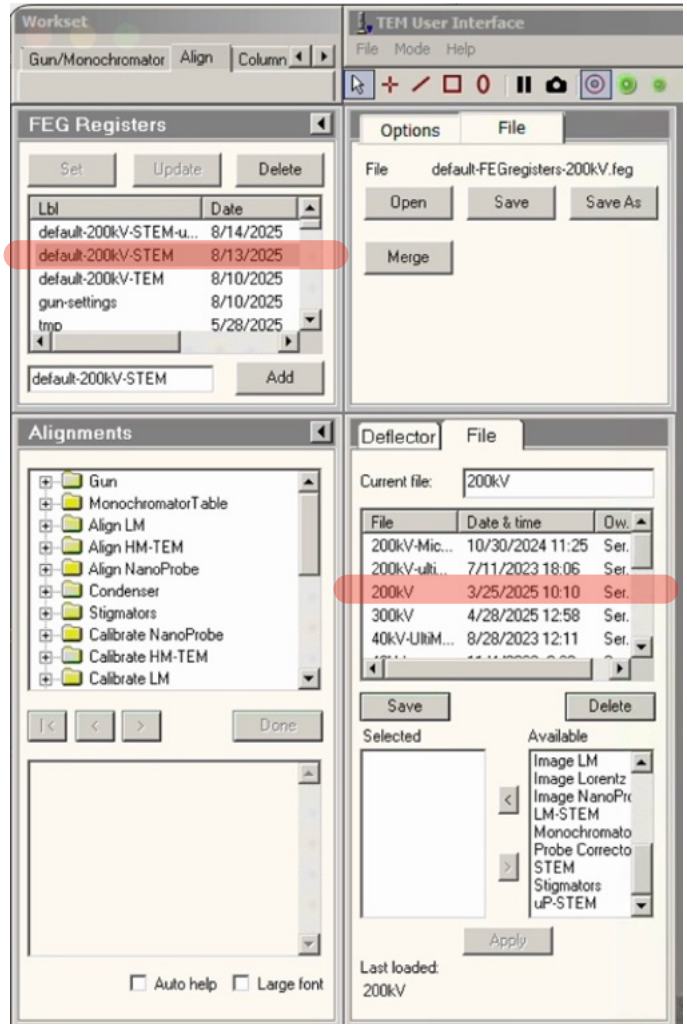
Daily routine to maintain reliability

Maintain one default set of tunings

Standard modes on a TFS TEM-STEM

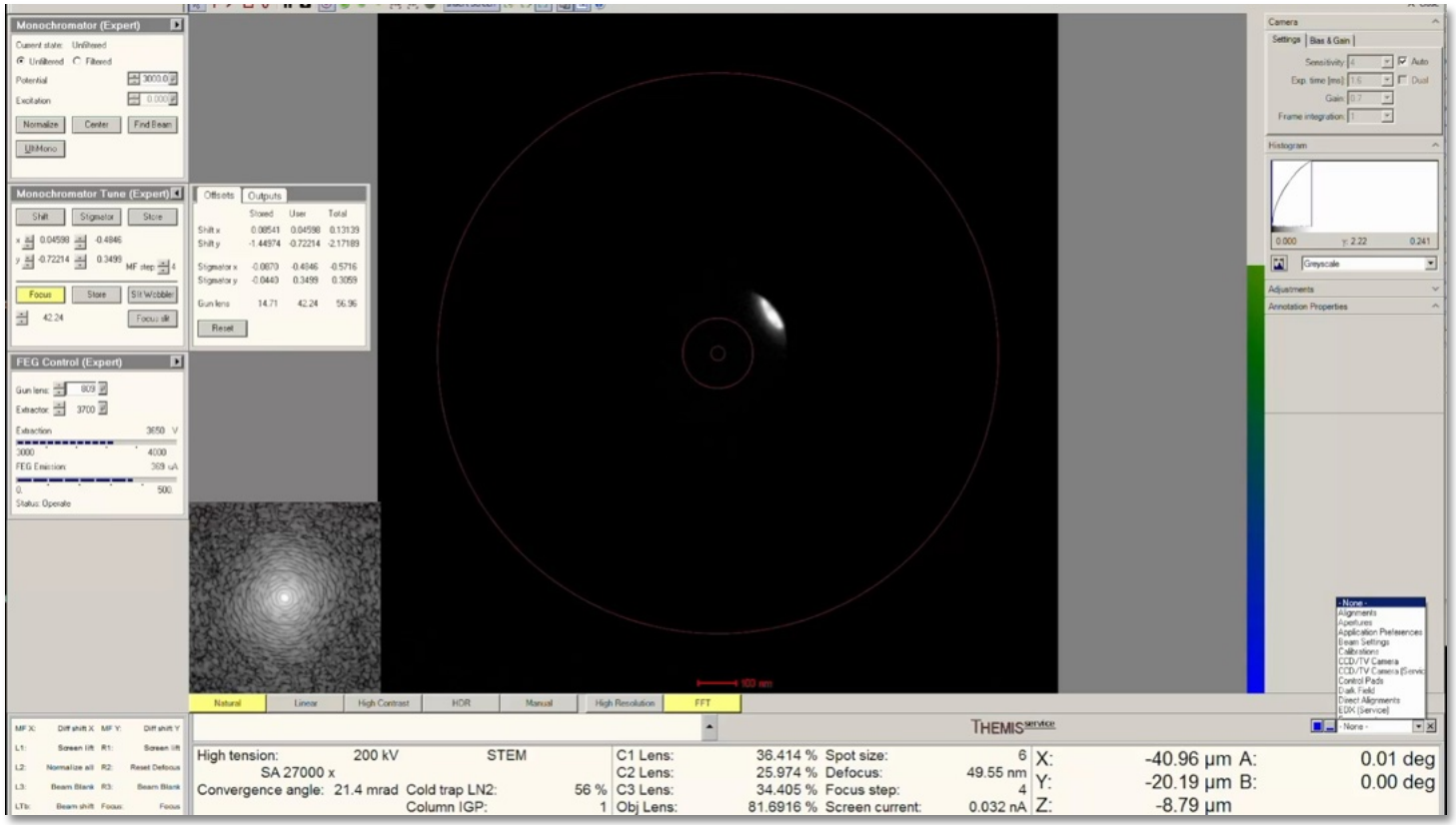


Example for the daily procedure for HRSTEM



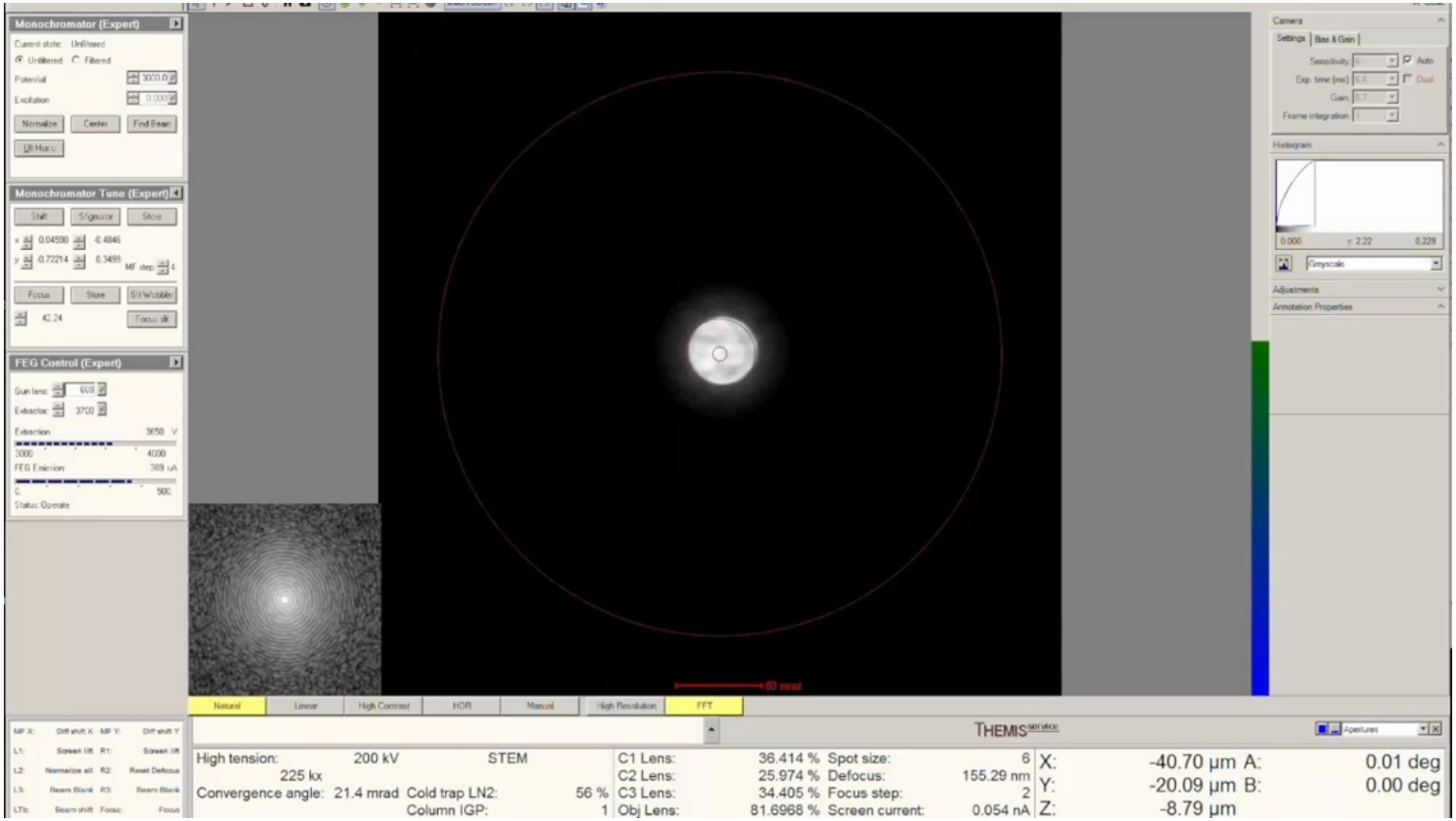
Start from default alignment data

Example for the daily procedure for HRSTEM



Loading aberration-corrector settings (probe and image-side), tune probe side (aperture & C2/C3 centering), tuning in the Ronchigram for A1 coarse, B2 via exported elements

Example for the daily procedure for HRSTEM



Tune aberration corrector (8x speed)

… ready to go ... shouldn’t take more than 20 min

Questions?