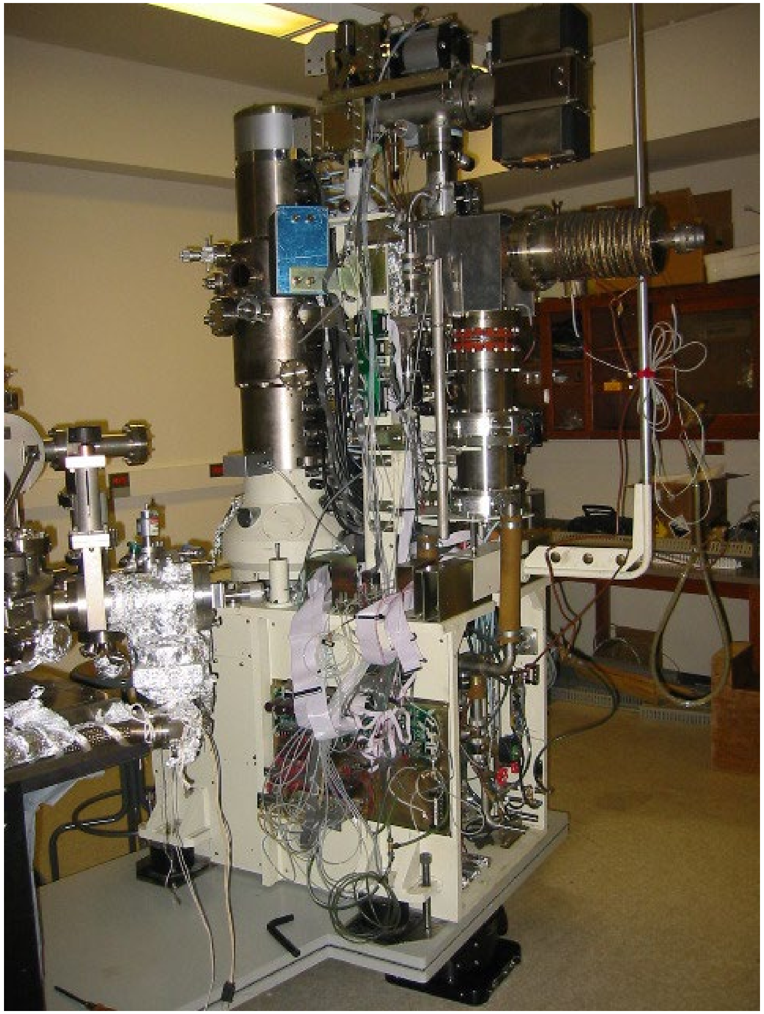
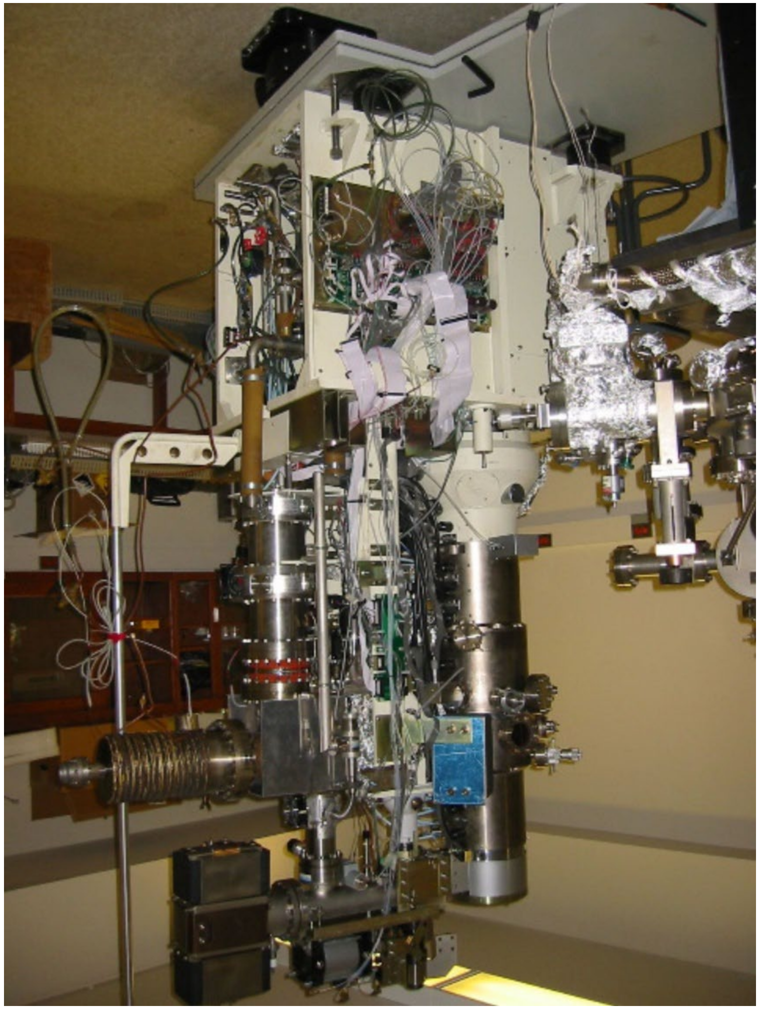
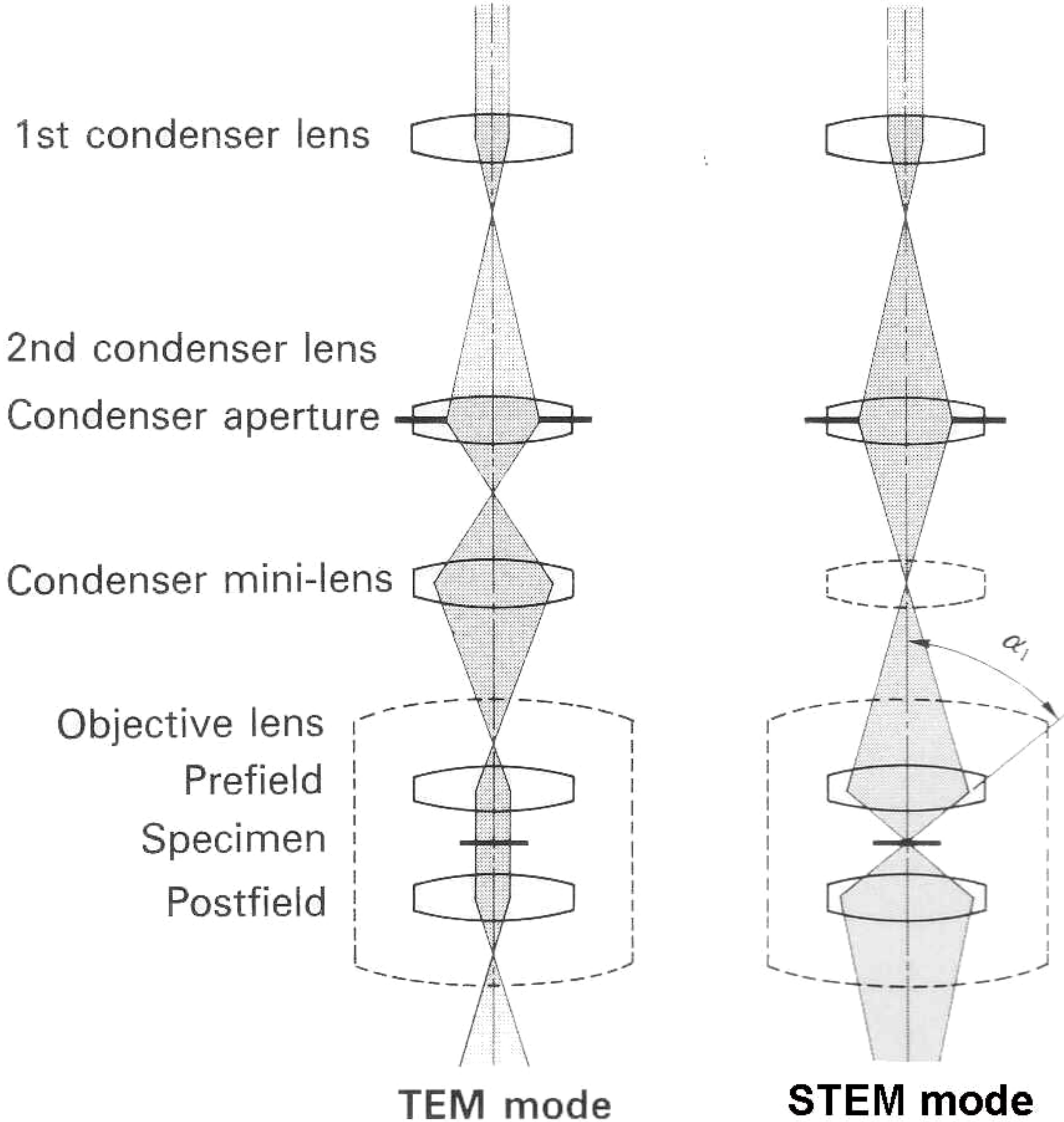
STEM

Conventional TEM



STEM

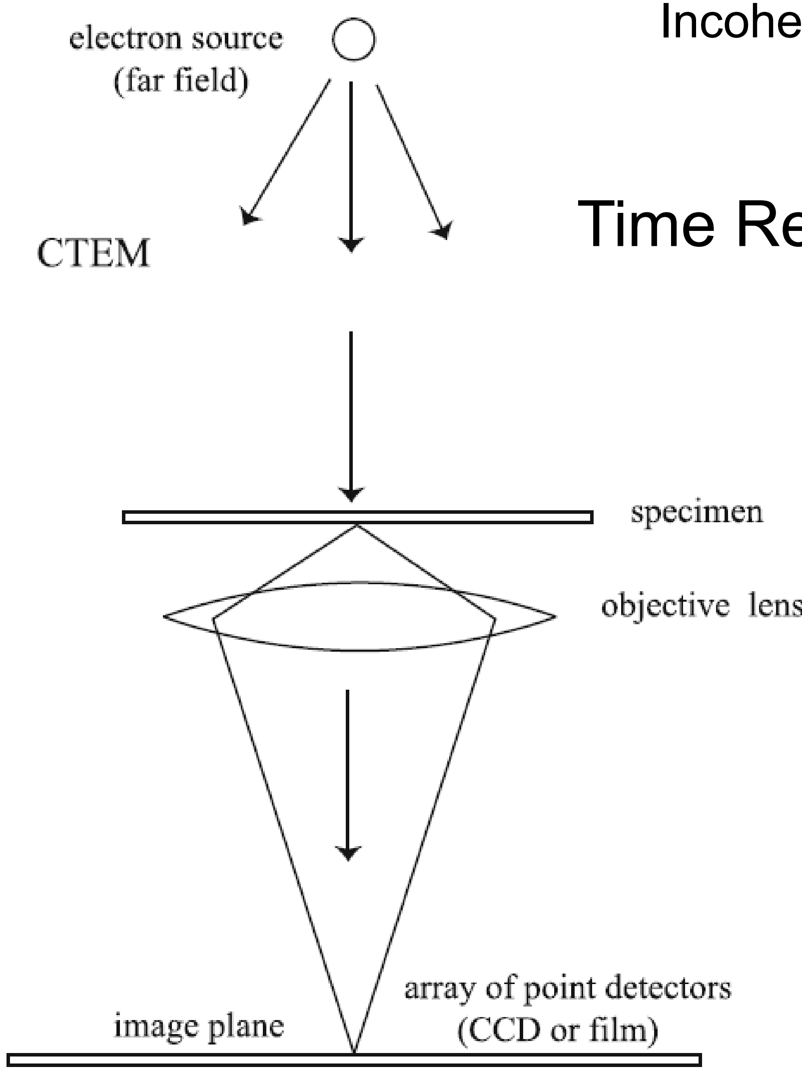



1st condenser lens
2nd condenser lens
Condenser aperture
Condenser mini-lens
Objective lens
Prefield
Specimen
Postfield
TEM mode
STEM mode

Reciprocity

Incoherent

Time Reversal



Coherent Image

Coherent Source

Comparison

TEM

- Condensor Aperture
- Objective aperture after sample
- No analogue
- Selected Area Aperture

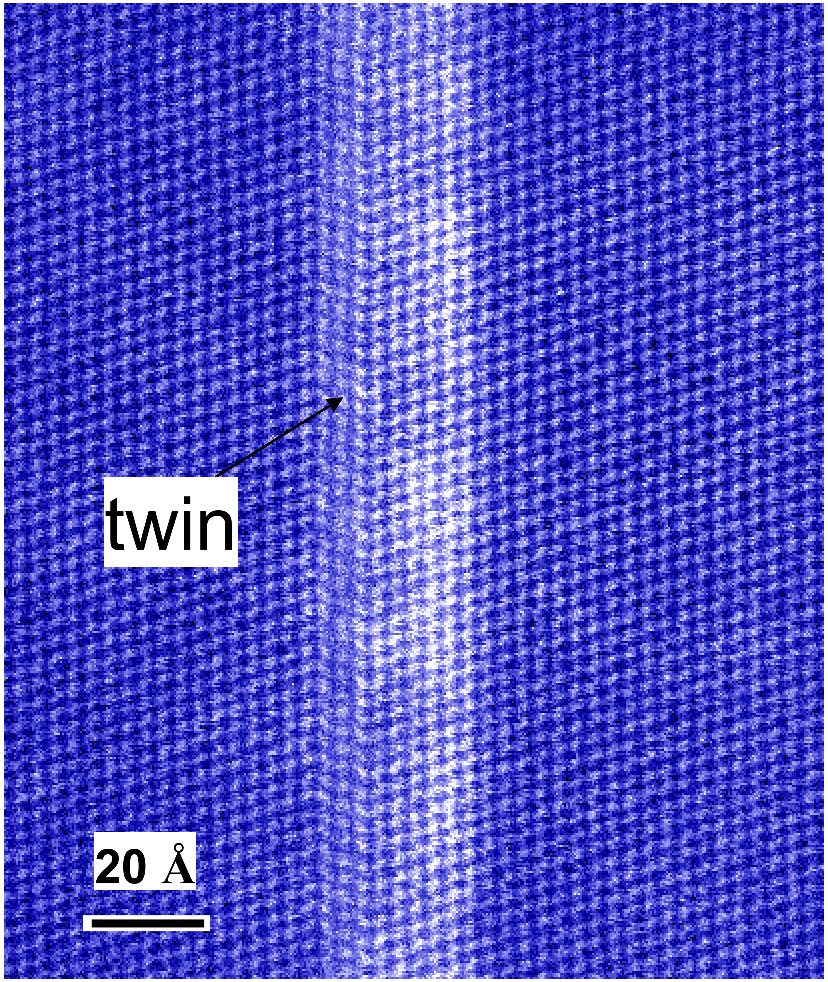
STEM

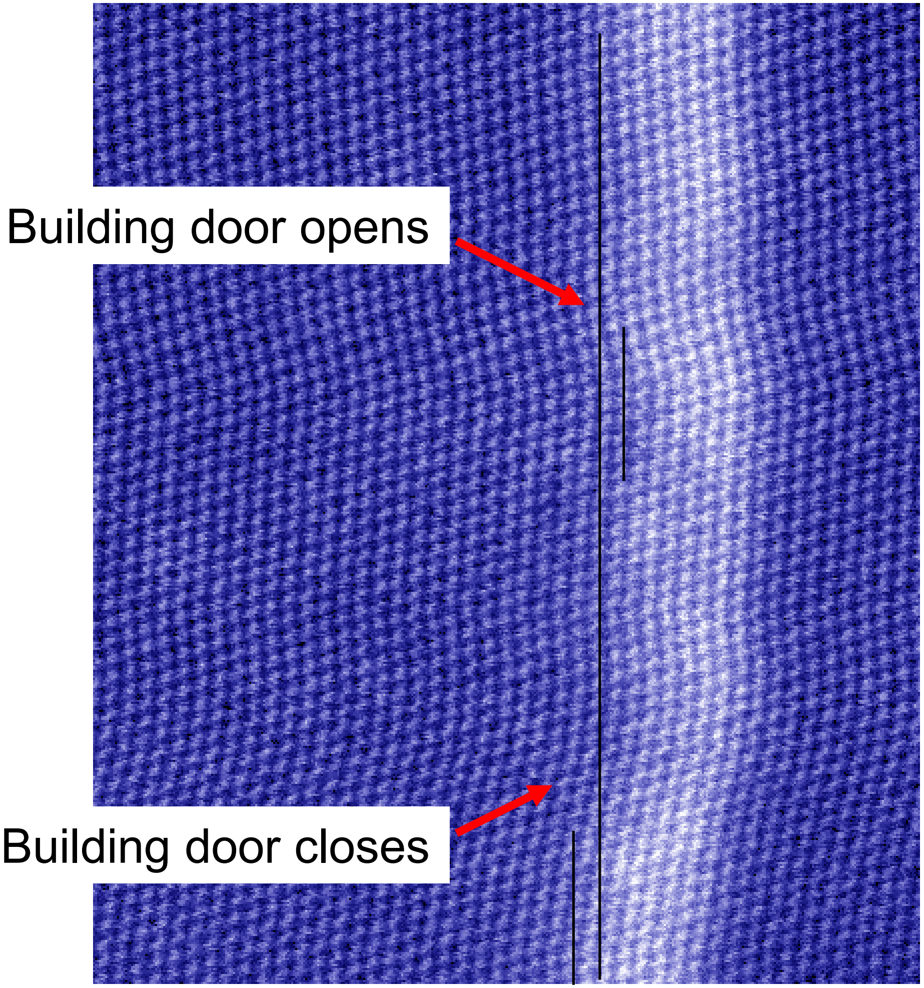
- Detector
- Objective aperture before sample
- Virtual objective aperture (condensor)
- No analogue

STEM v TEM

- STEM = TEM, but with the optics reversed (if you ignore inelastic scattering)
- BF, DF, HREM can be done in a STEM almost the same as a TEM
- However:
  - The imaging is serial as against parallel, so in general has more noise and lower signals
  - Scan coils and electronics involved, which leads to noise
  - Some types of imaging can be done easier in a STEM than a TEM (they have all been done in both)
  - STEM needs a small source (demagnified further)

Environmental Sensitivity

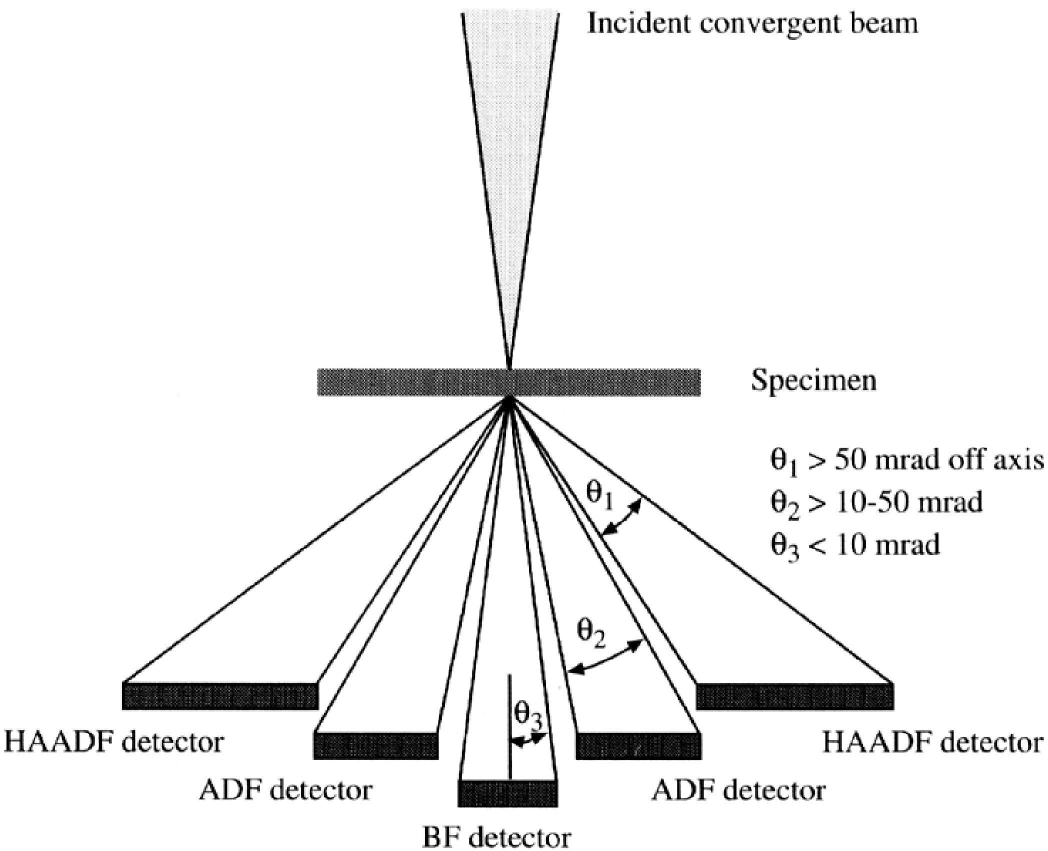


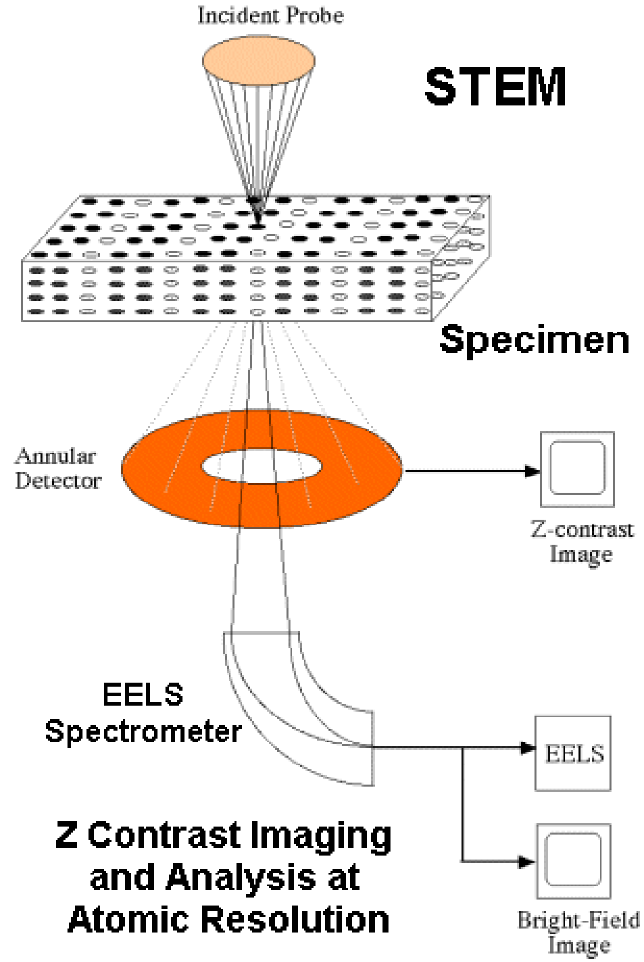


1 Pa change in air pressure → 1 Å stage deflection

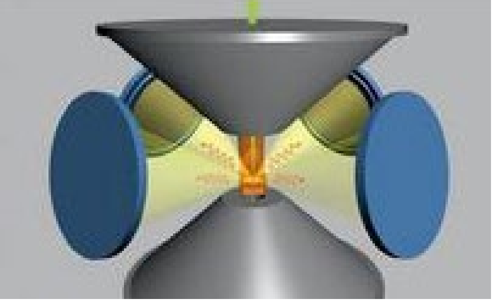
D. A. Muller JEM **50**, 219 (2001)

More detectors are available!



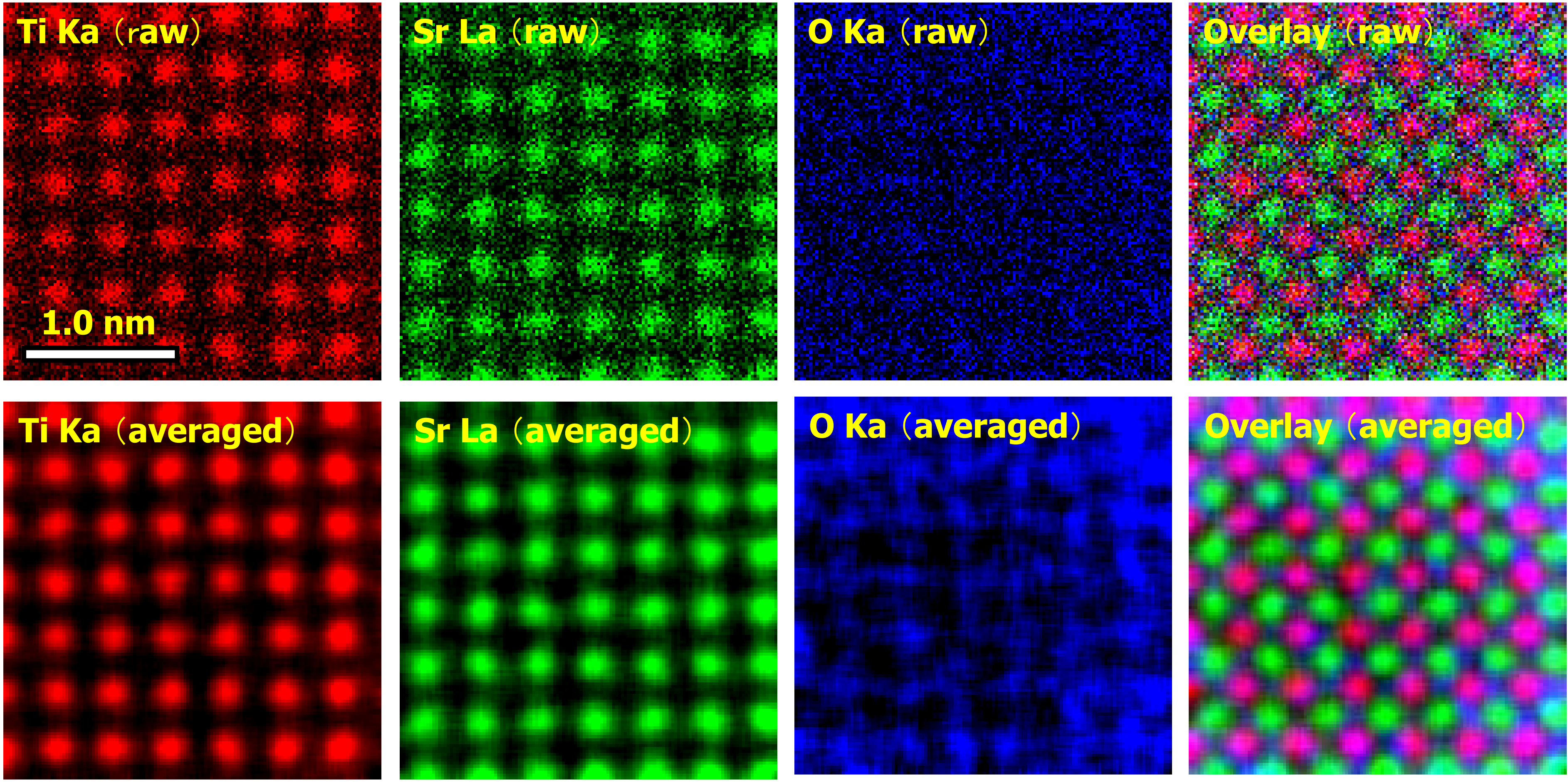


Spectroscopy

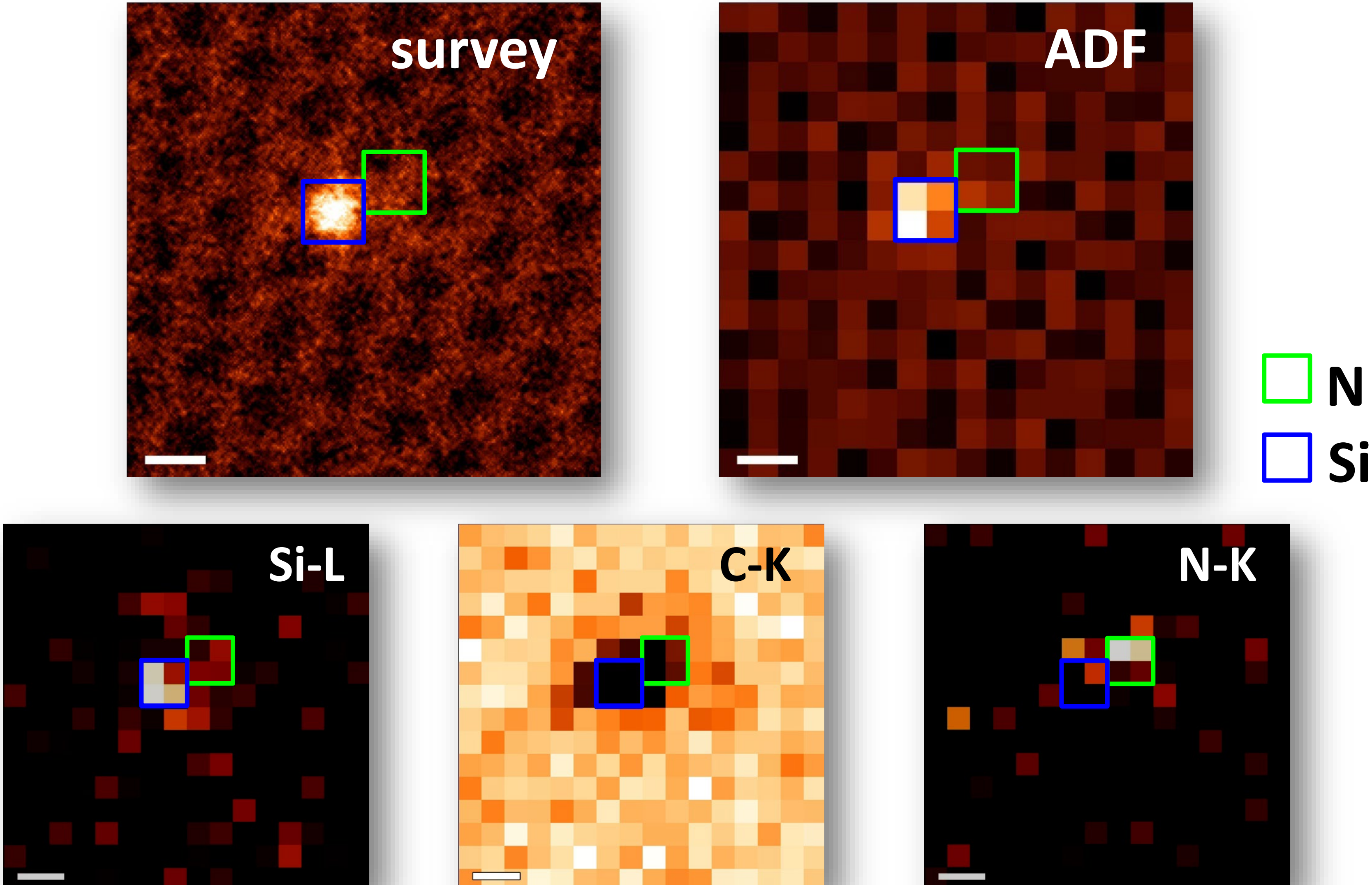


Atomic Resolution EDS

128x128, **175f**, T4, 0.2ms
RT: **9m33s**, DT: 3.71%
Count rate: 1119.57cps



Atom by atom spectroscopy

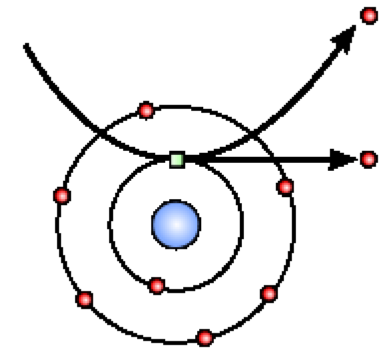


Zhou, W., Lee, J., Nanda, J., Pantelides, S. T., Pennycook, S. J., & Idrobo, J.-C. (2012). Atomically localized plasmon enhancement in monolayer graphene. *Nature nanotechnology*, *7*(3), 161–165.

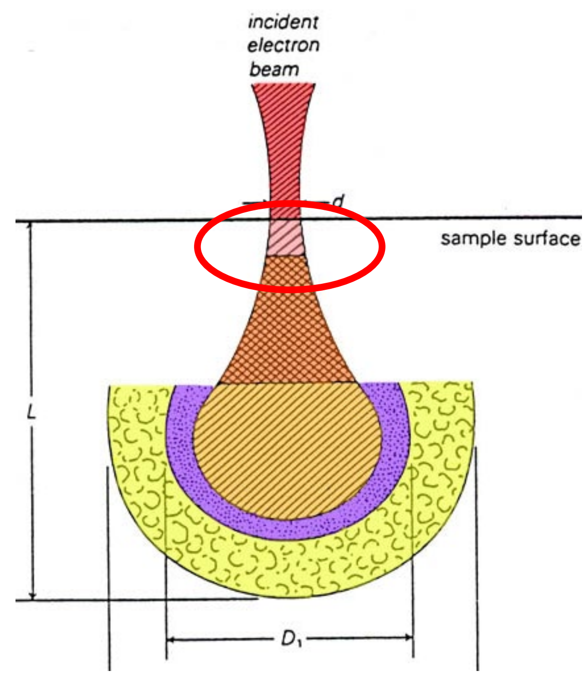
Materials Science and Engineering and Scientific User Facilities Divisions

Secondary Electrons

Secondary electrons (SE)

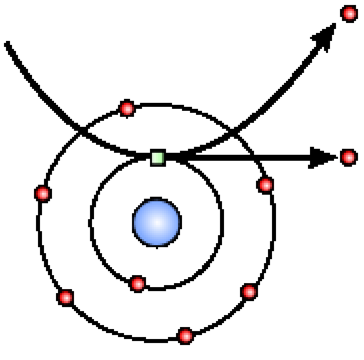


- Generated from the collision between the incoming electrons and the loosely bonded outer electrons
- Low energy electrons (~10-50 eV)
- Only SE generated close to surface escape (topographic information is obtained)
- Number of SE is greater than the number of incoming electrons
- We differentiate between SE1 and SE2



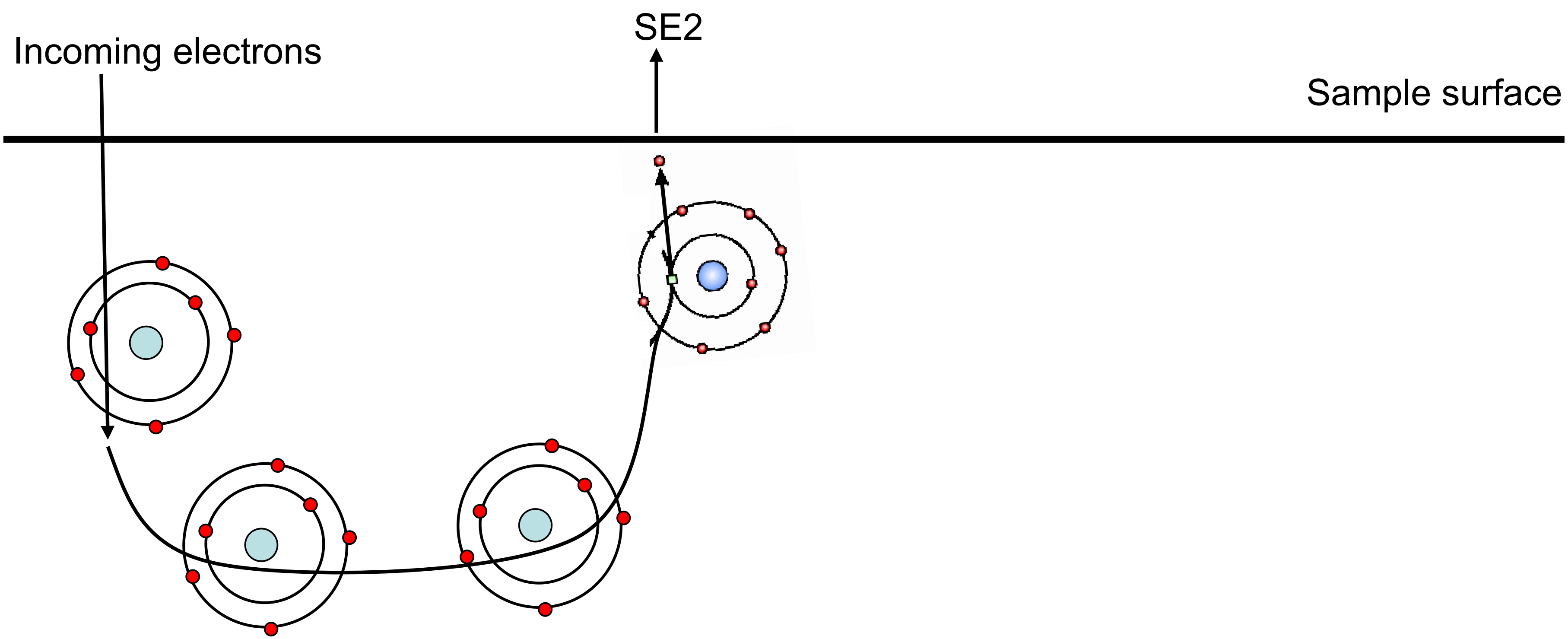
SE1

- The secondary electrons that are generated by the incoming electron beam as they enter the surface
- High resolution signal with a resolution which is only limited by the electron beam diameter

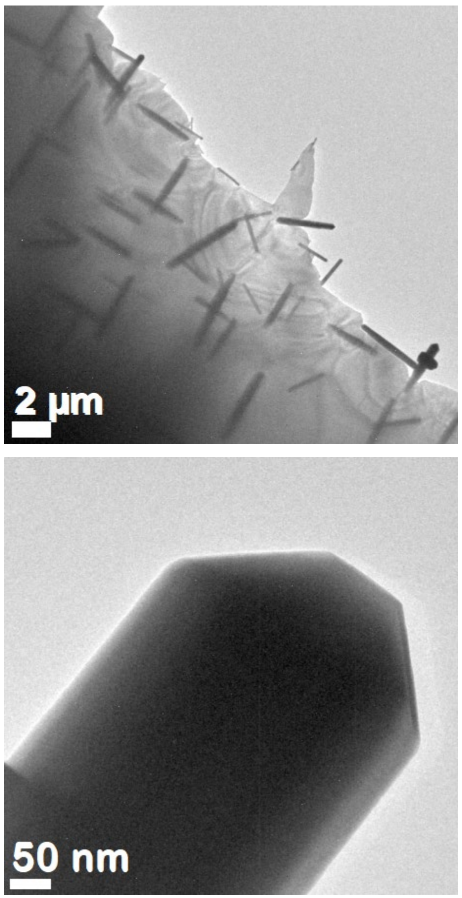


SE2

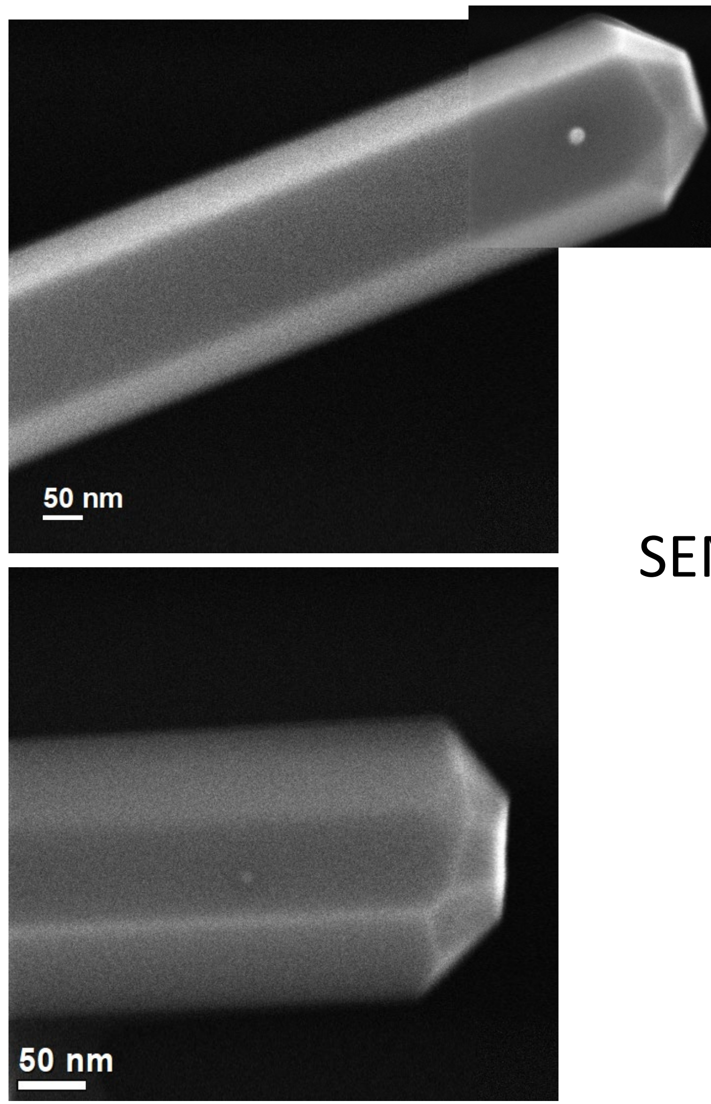
- The secondary electrons that are generated by the backscattered electrons that have returned to the surface after several inelastic scattering events
- SE2 come from a surface area that is bigger than the spot from the incoming electrons → resolution is poorer than for SE1 exclusively



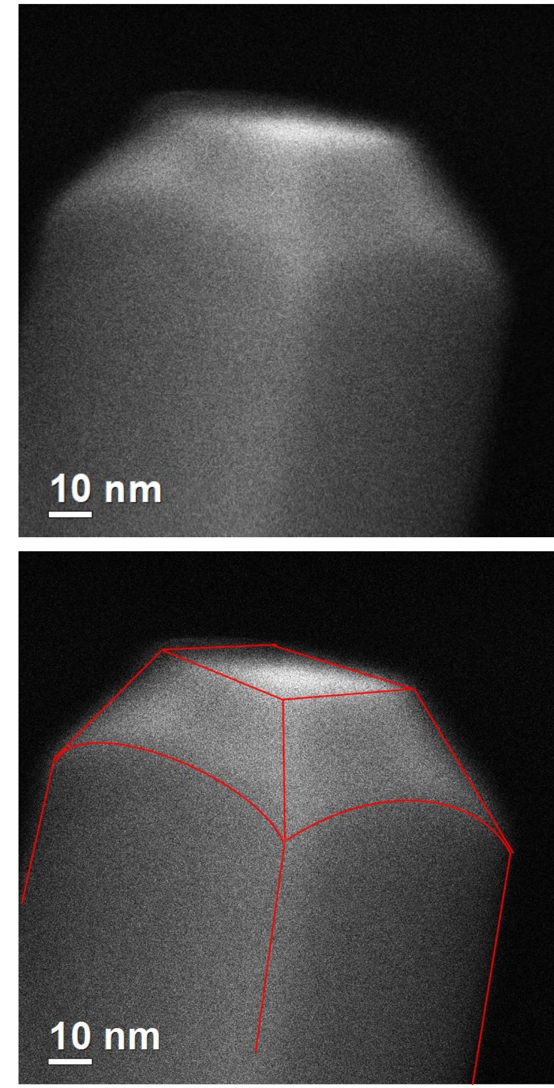
Rods in $KTaO_3$

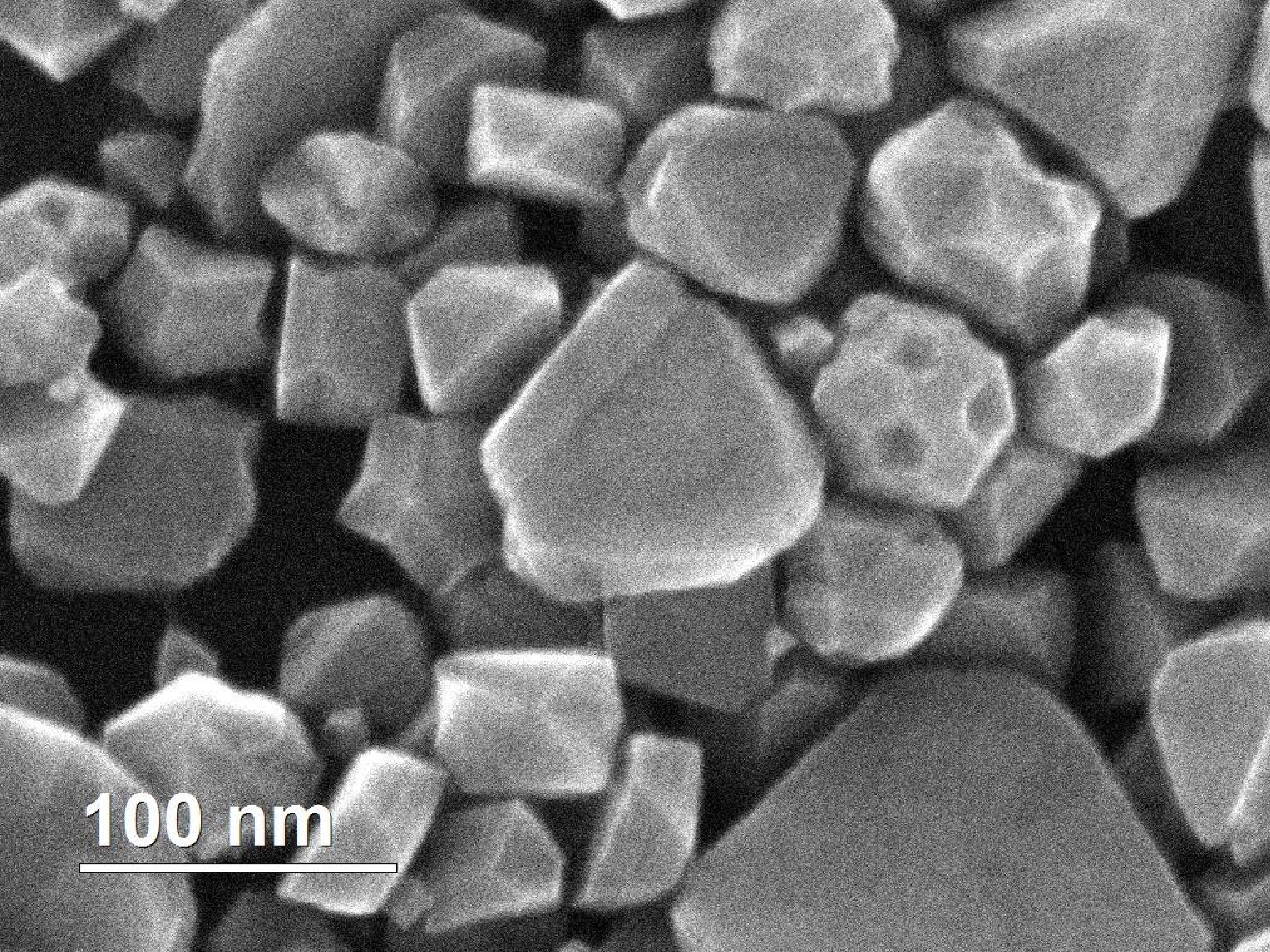


Bright Field TEM

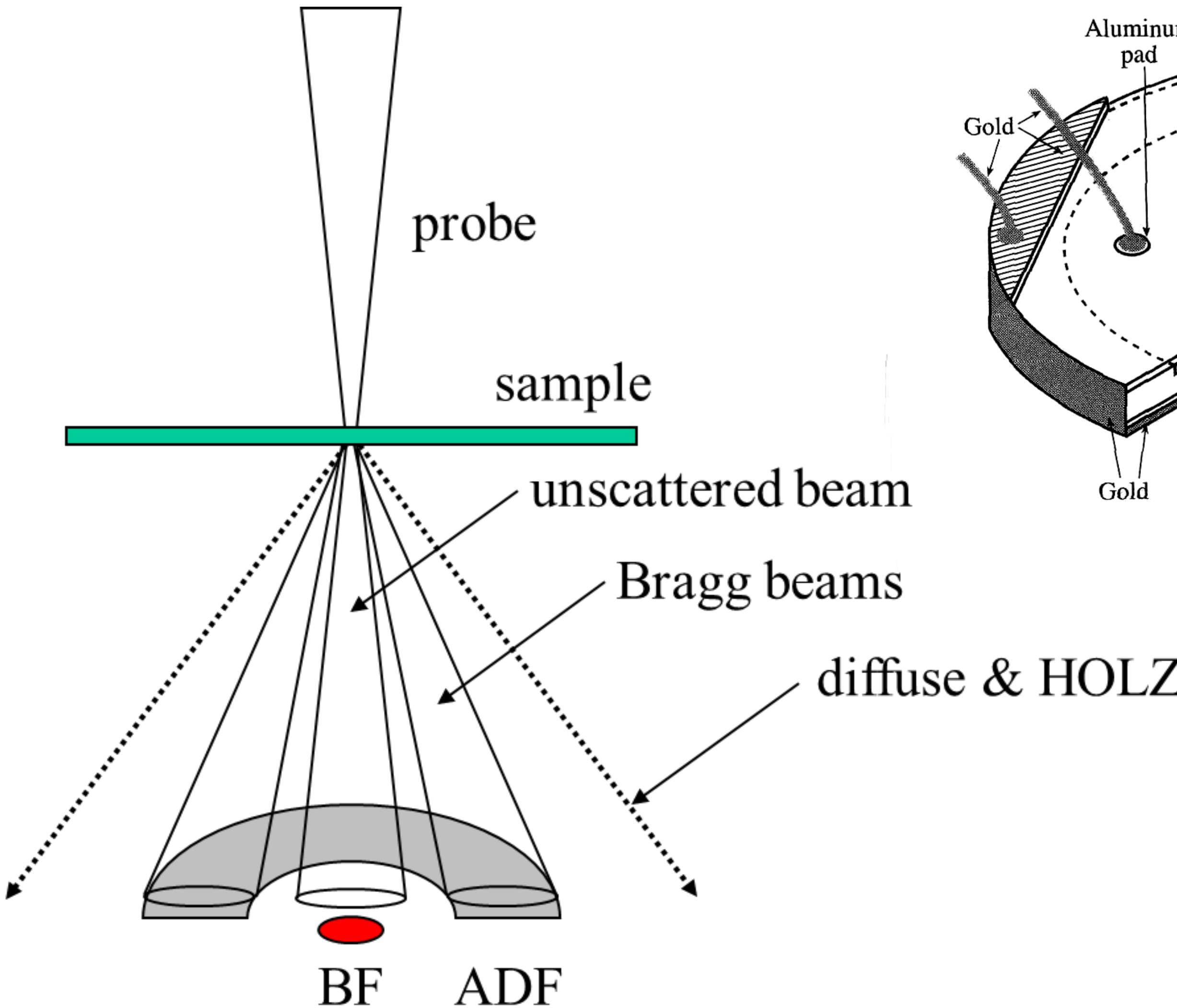


SEM

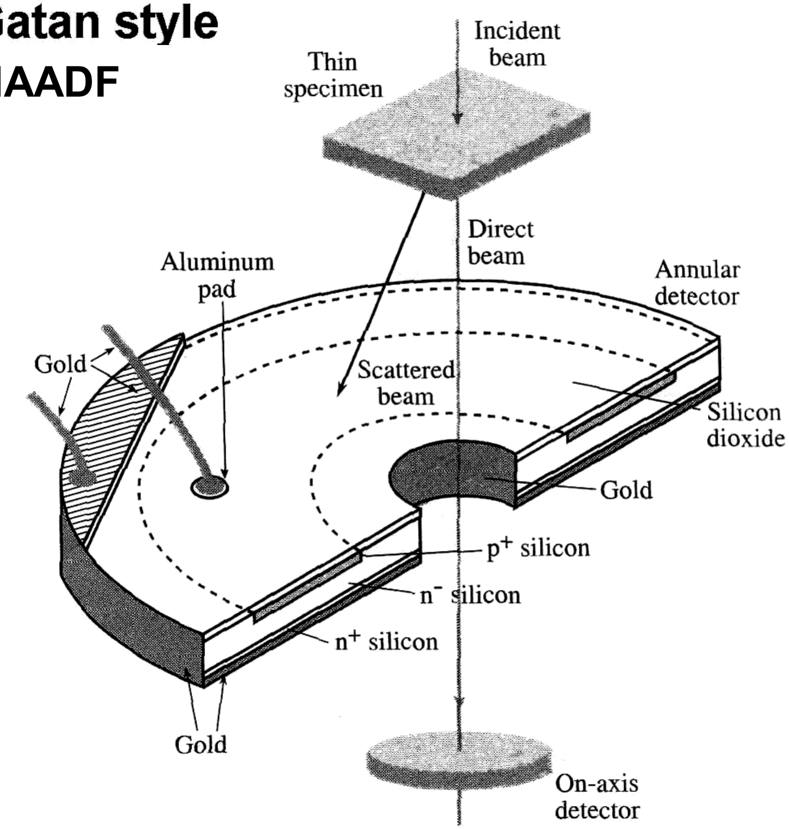



100 nm

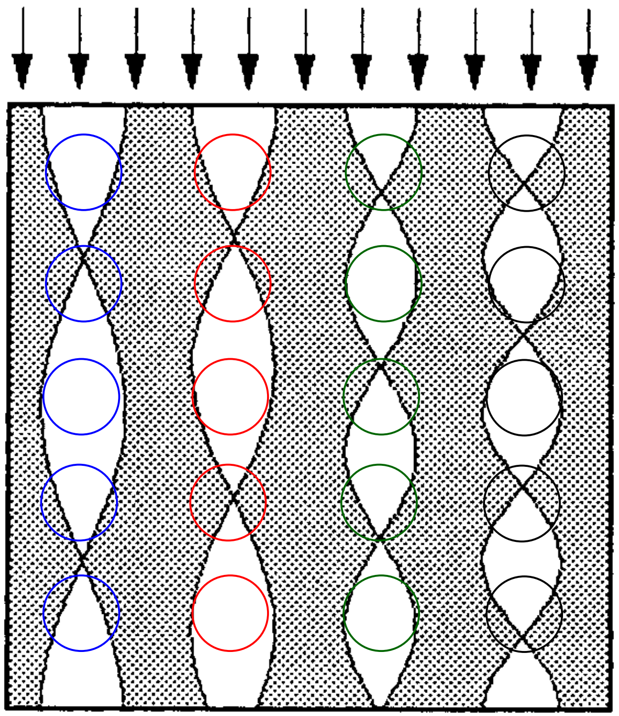
HAADF v ADF

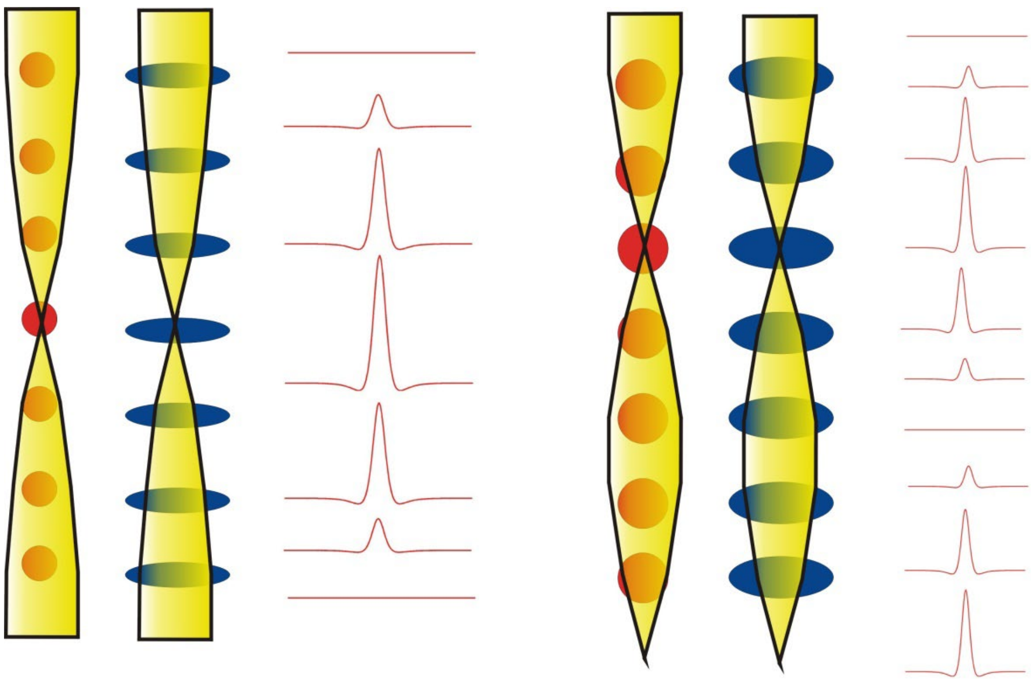


Gatan style HAADF



$$\psi(\mathbf{R},z)-1 \approx \sum_{i,j} C_i \Phi_i(\mathbf{R}-\mathbf{R}_j)\left(\exp(-i\pi\varepsilon_n z)-1\right)$$

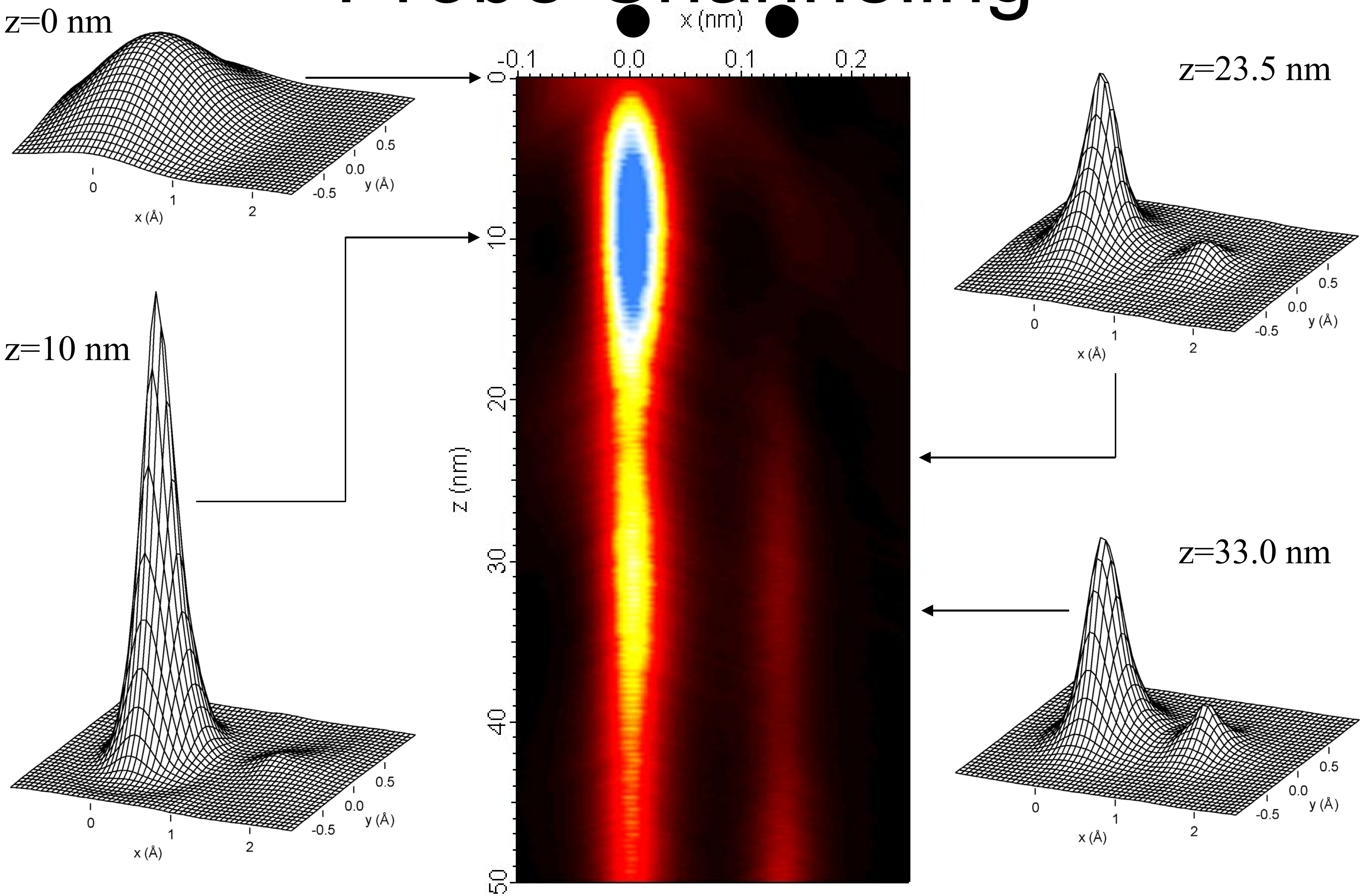




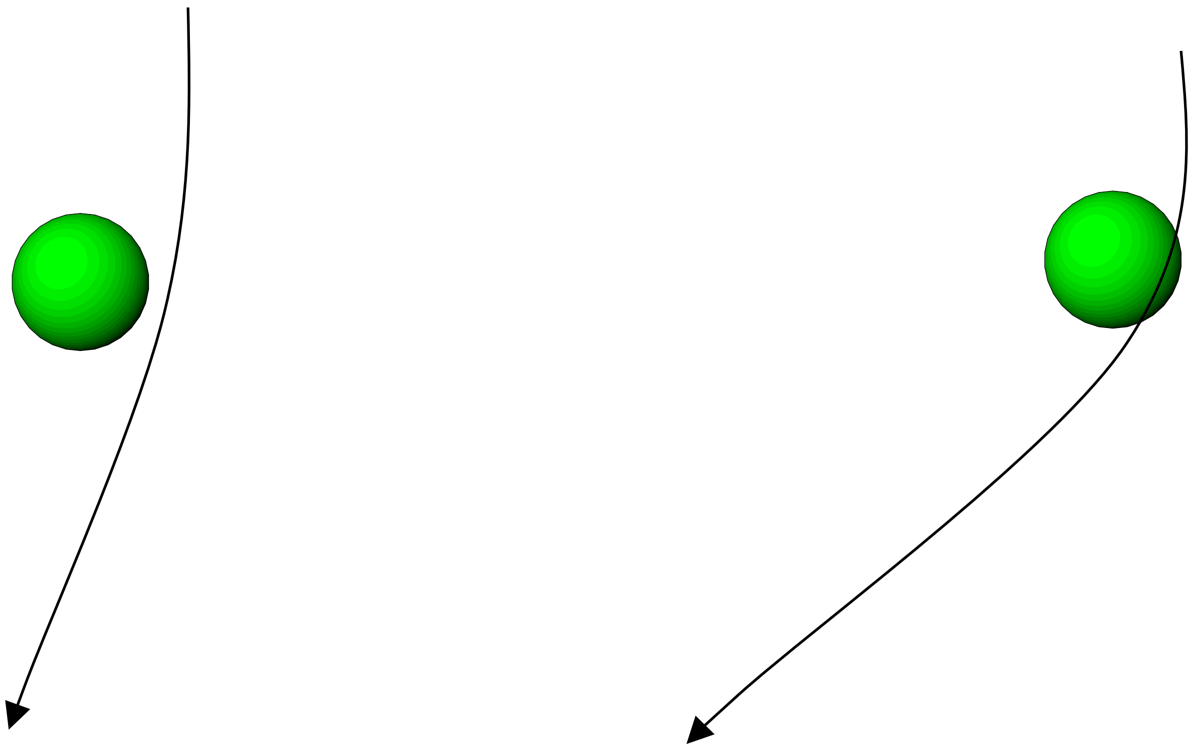
light atoms heavy atoms light atoms heavy atoms

Curtesy D. Van Dyck

Probe Channeling

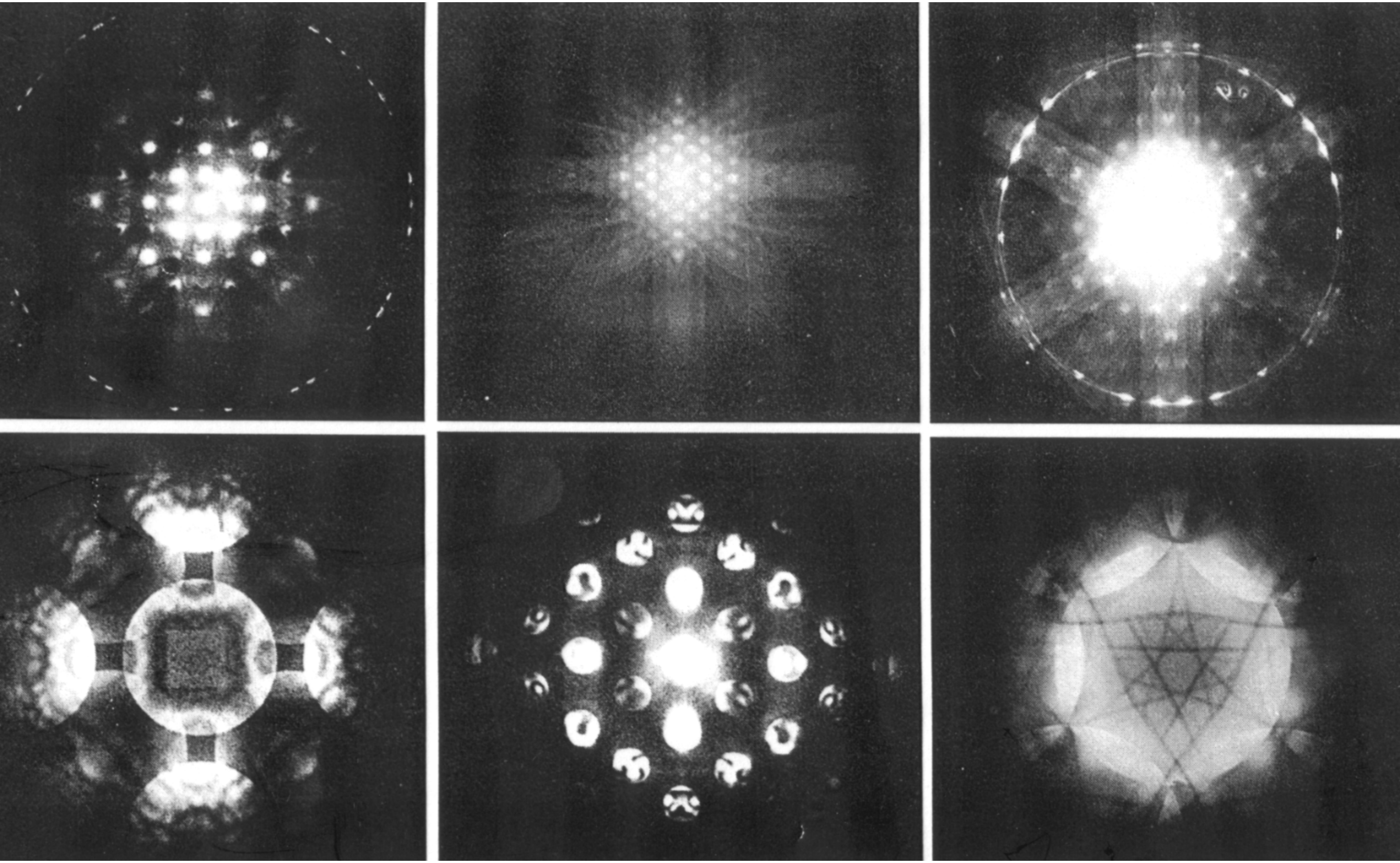


HAADF



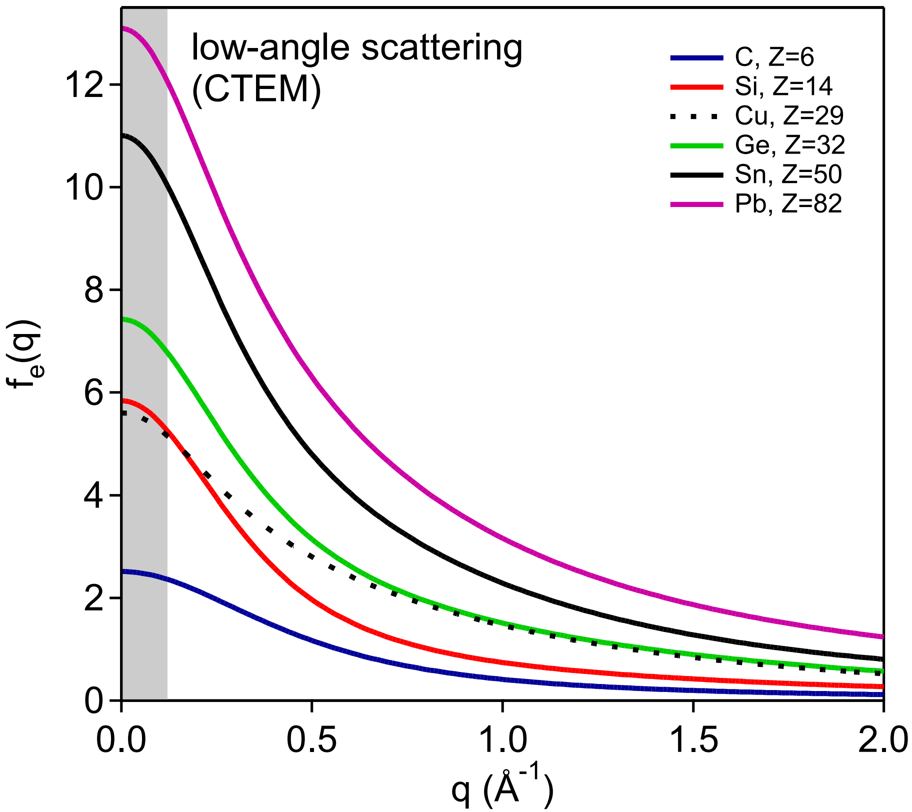
The closer the electron comes to the nucleus, the higher the probability of high-angle scattering (elastic or inelastic)

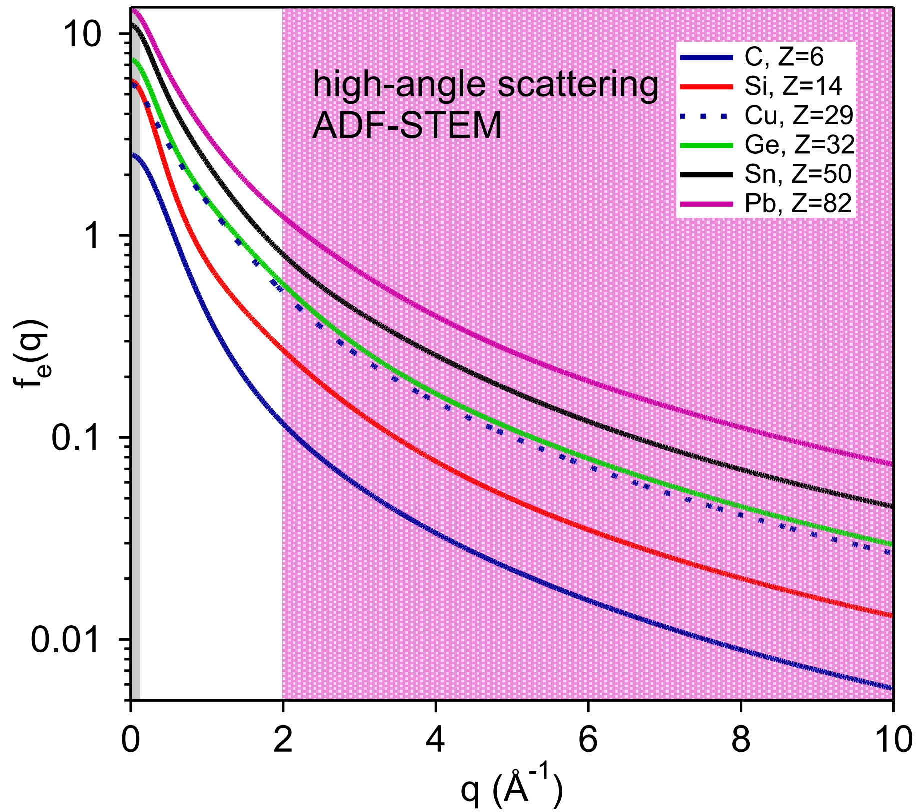
1s states dominate the high-angle scattering, incoherent when averaged over detector



Partitioning Signal in Scattering Space

Scattering from one atom is proportional to scattering factor $f_e(q)$.



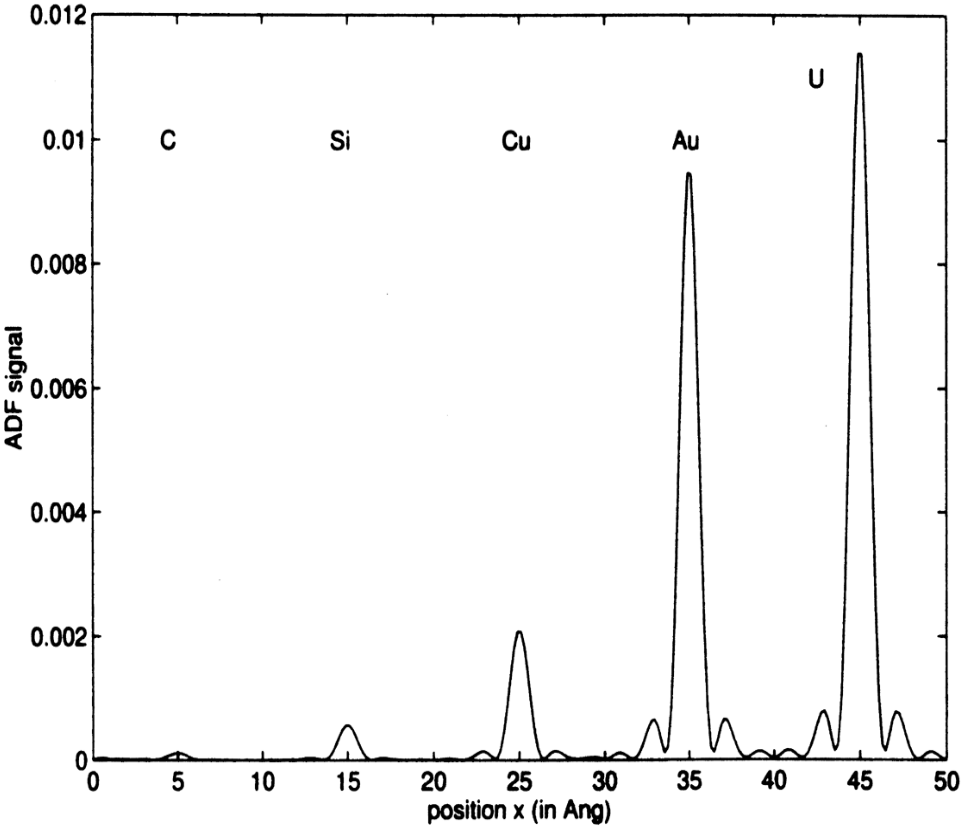


BF CTEM involves mostly low-angle scattering: Cu & Si cross.

HADF-STEM involves high-angle scattering only.

Z-contrast

- Scattering scales as ~$Z^{1.7}$ for common scattering angles
- Images are somewhat readily interpretable
- Images contain some chemical information
- Many problems with this simple picture (good enough for Govt work)



E. J. Kirkland, *Advanced Computing in Electron Microscopy* (Plenum, 1998).

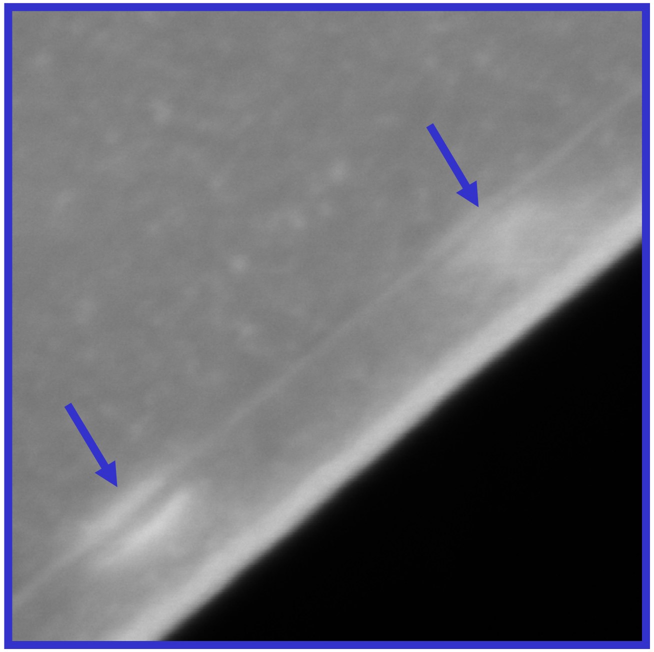
Caveat

- Consider via Fermi's Golden Rule

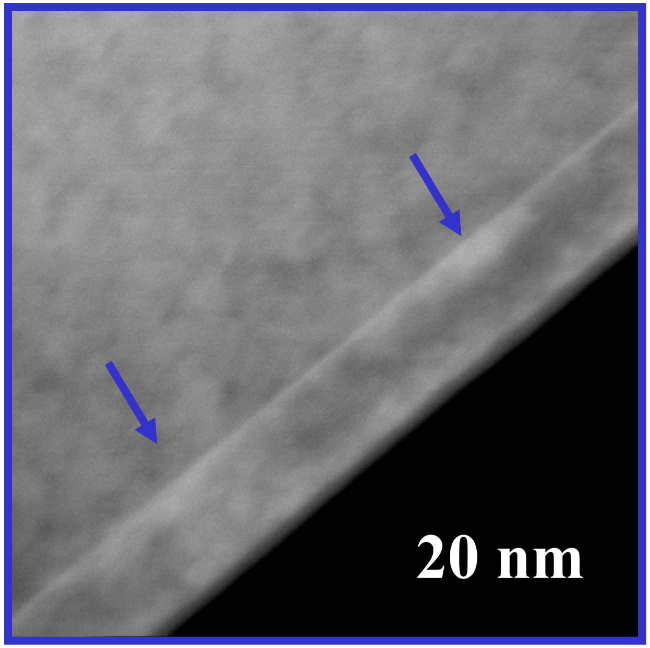
  Signal = $\rho(r)|\langle f|V|i\rangle|^2$
- "Z-contrast" only corresponds to the $|\langle f|V|i\rangle|^2$ contribution (HOLZ, phonons)
- But $\rho(r)$ comes from dynamical diffraction
- True Z-contrast occurs iff $\rho(r)$ has a simple form, which it does in special cases

Comparison between High-Angle and Low-Angle Annular Dark-Field Image

Cross–sectional sample through InGaAs quantum dots and associated quantum well grown on GaAs and capped with GaAs. Dots are arrowed.



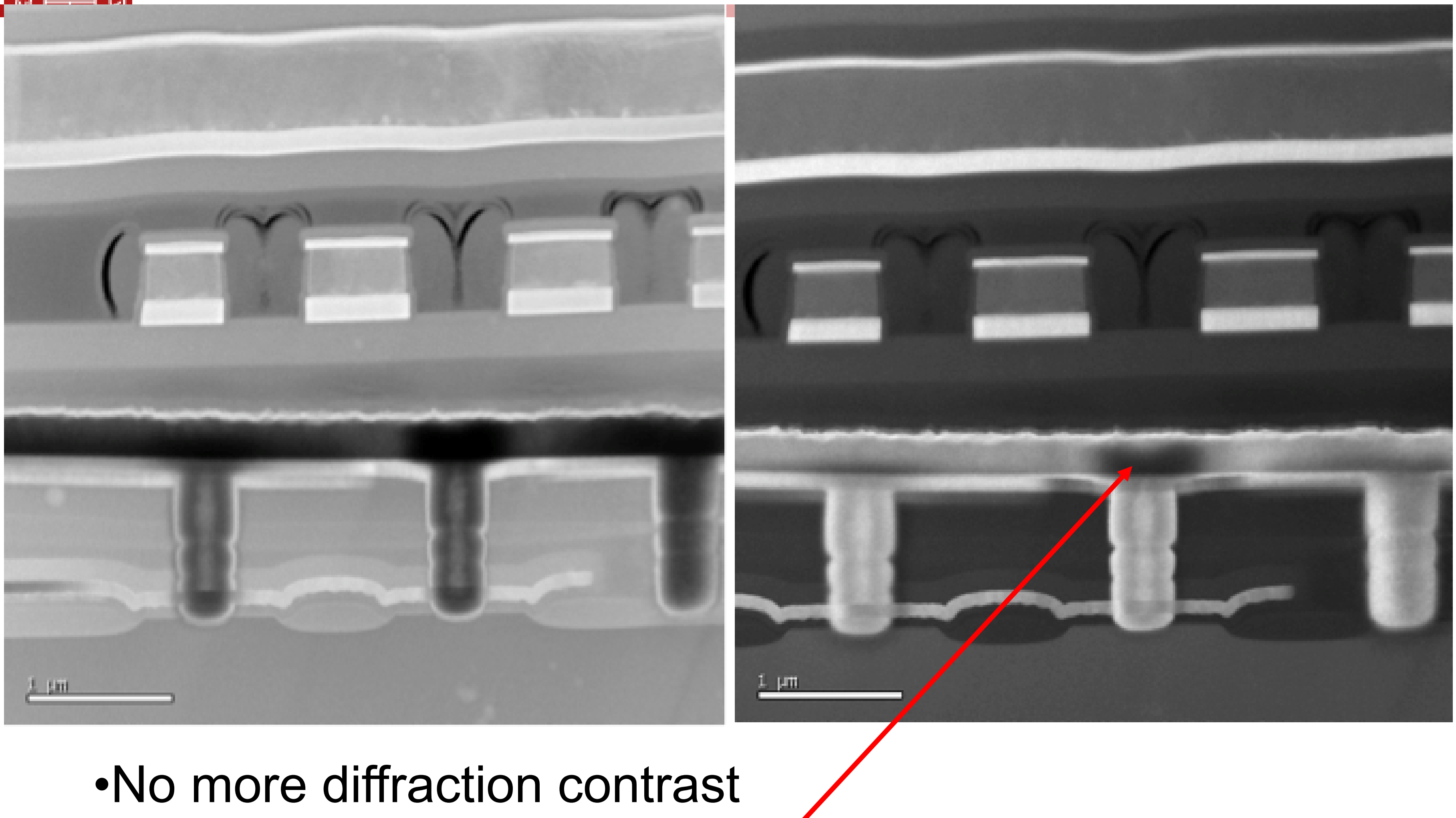
Low-angle annular dark field showing strain contast



High-angle annular dark field showing mostly Z contrast

Contrast Reversals in Thick Samples at 200kV

ADF-STEM (ϕ_c>45 mr) ADF-STEM (ϕ_c>75 mr)



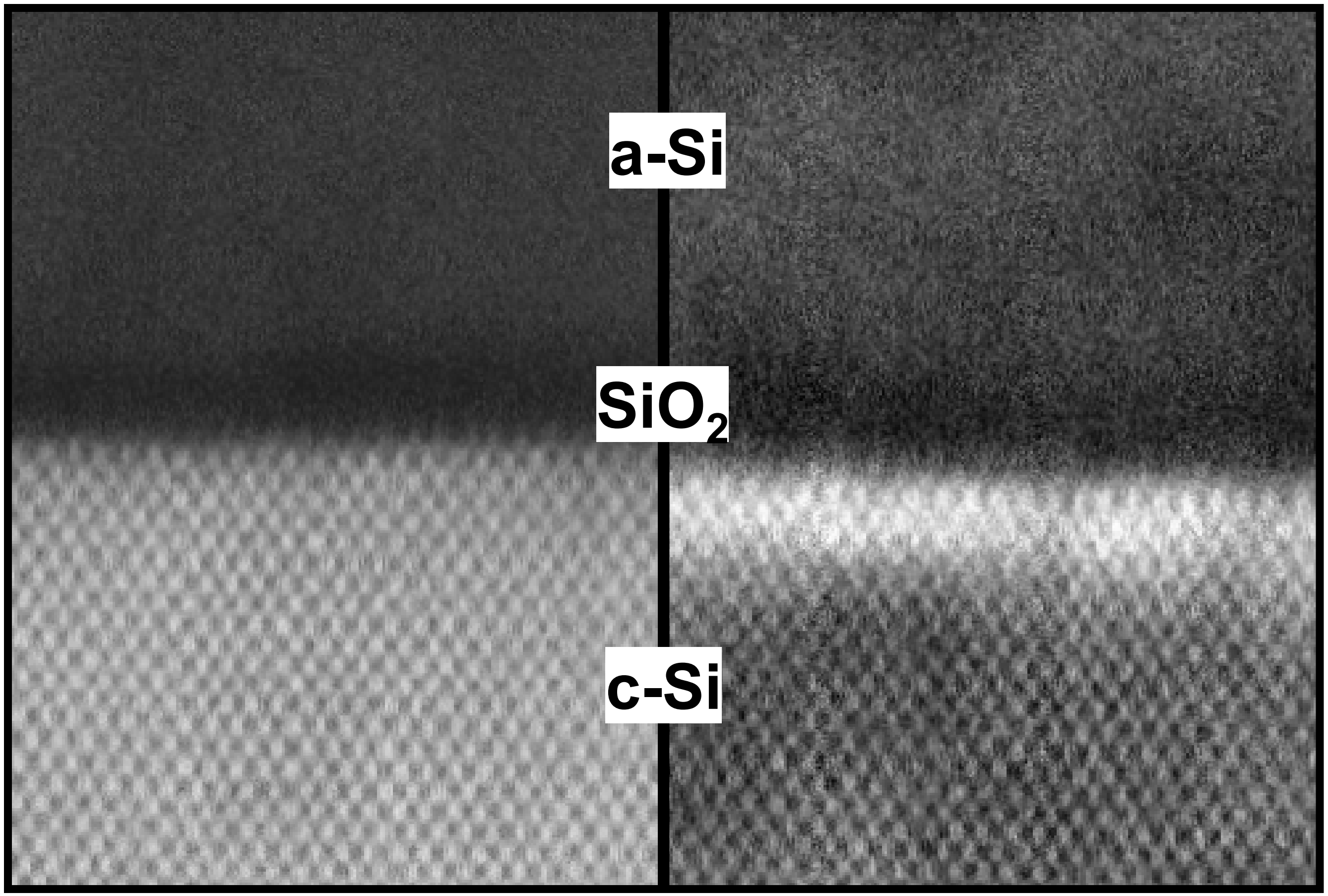
- No more diffraction contrast
- Signal in W plug not monotonic, could be mistaken for voids
- Effect reduced by increasing the collector angle

Strain Contrast at Si/SiO_2 Interfaces

(JEOL 2010F, 200 kV, C_s=1mm)

ADF Inner angle:

50 mrad 25 mrad

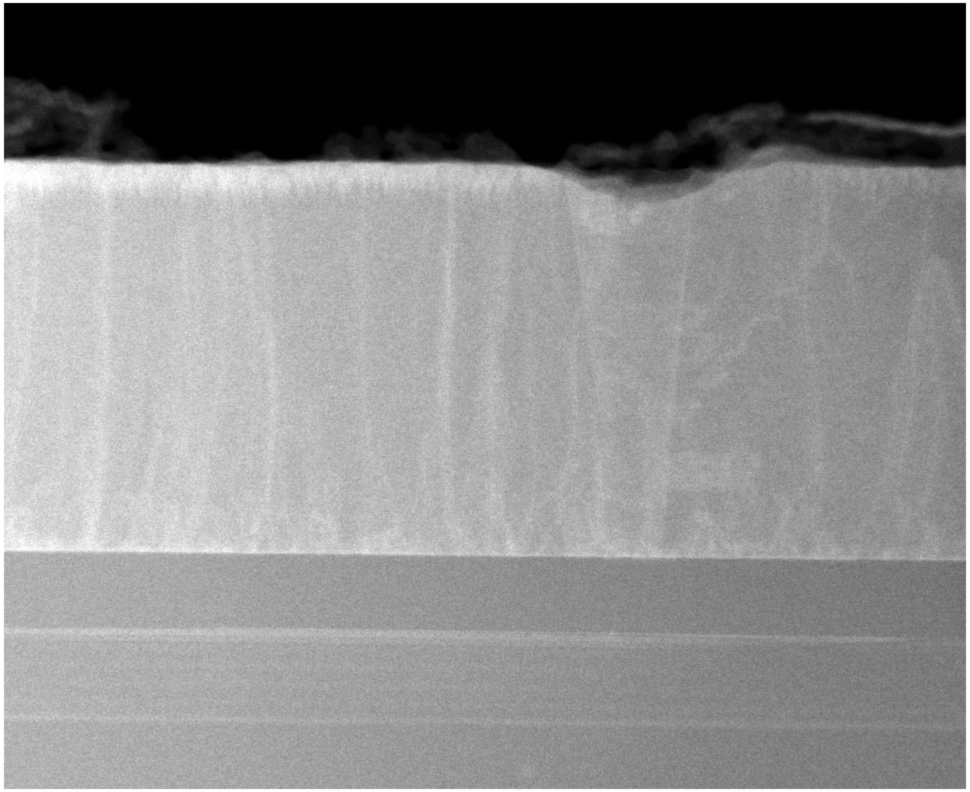


Strain Fields cause dechanneling (and scattering to small angles)

Z. Yu, D. A. Muller, and J. Silcox, *J. Appl. Phys.* **95**, 3362 (2004).

Not so High-Quality GZO

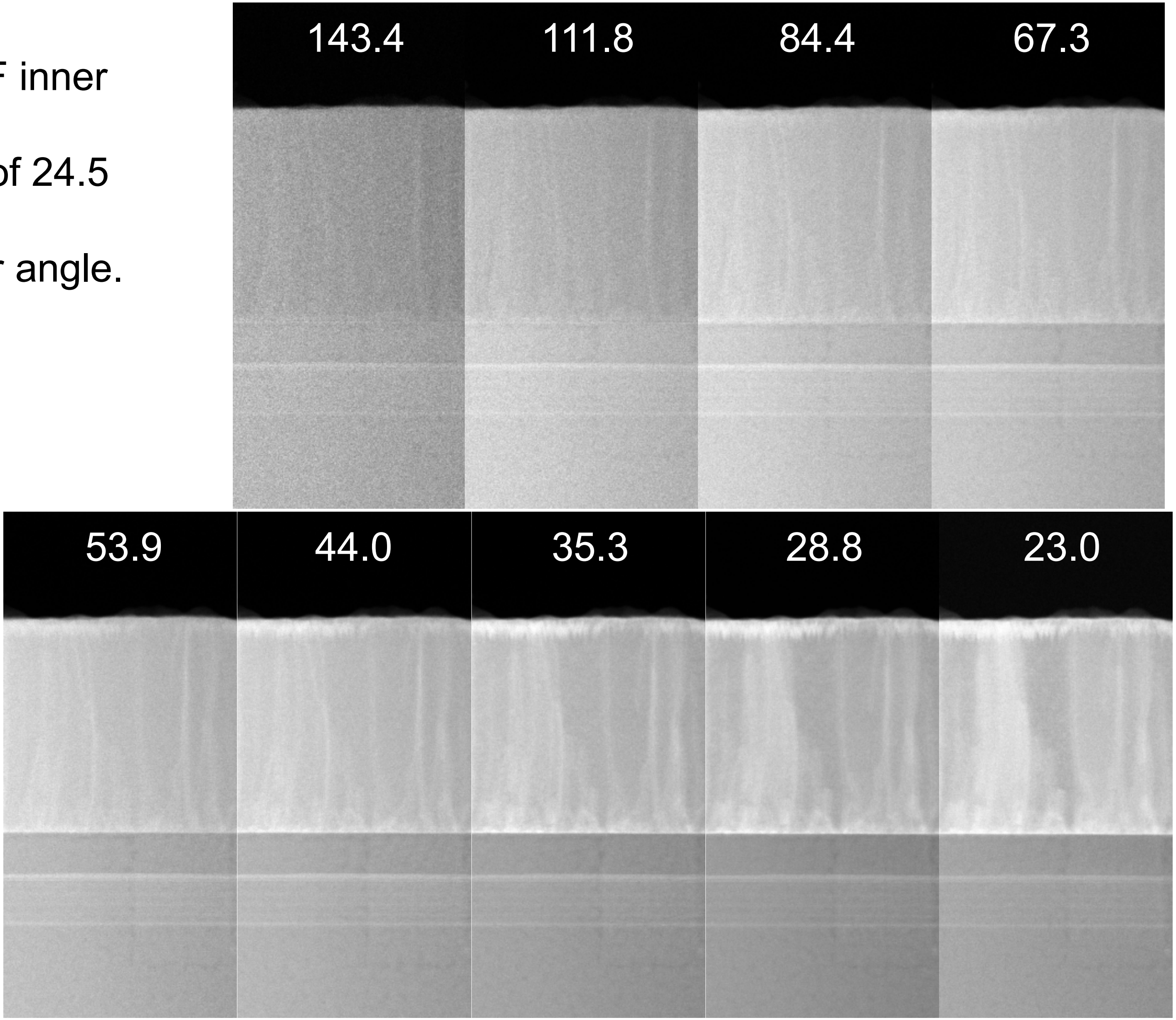
- Low-angle grain boundaries
- Inversion domain boundaries
- Interface and surface strain
- GZO brighter than GaN?



Paul Voyles, UWM

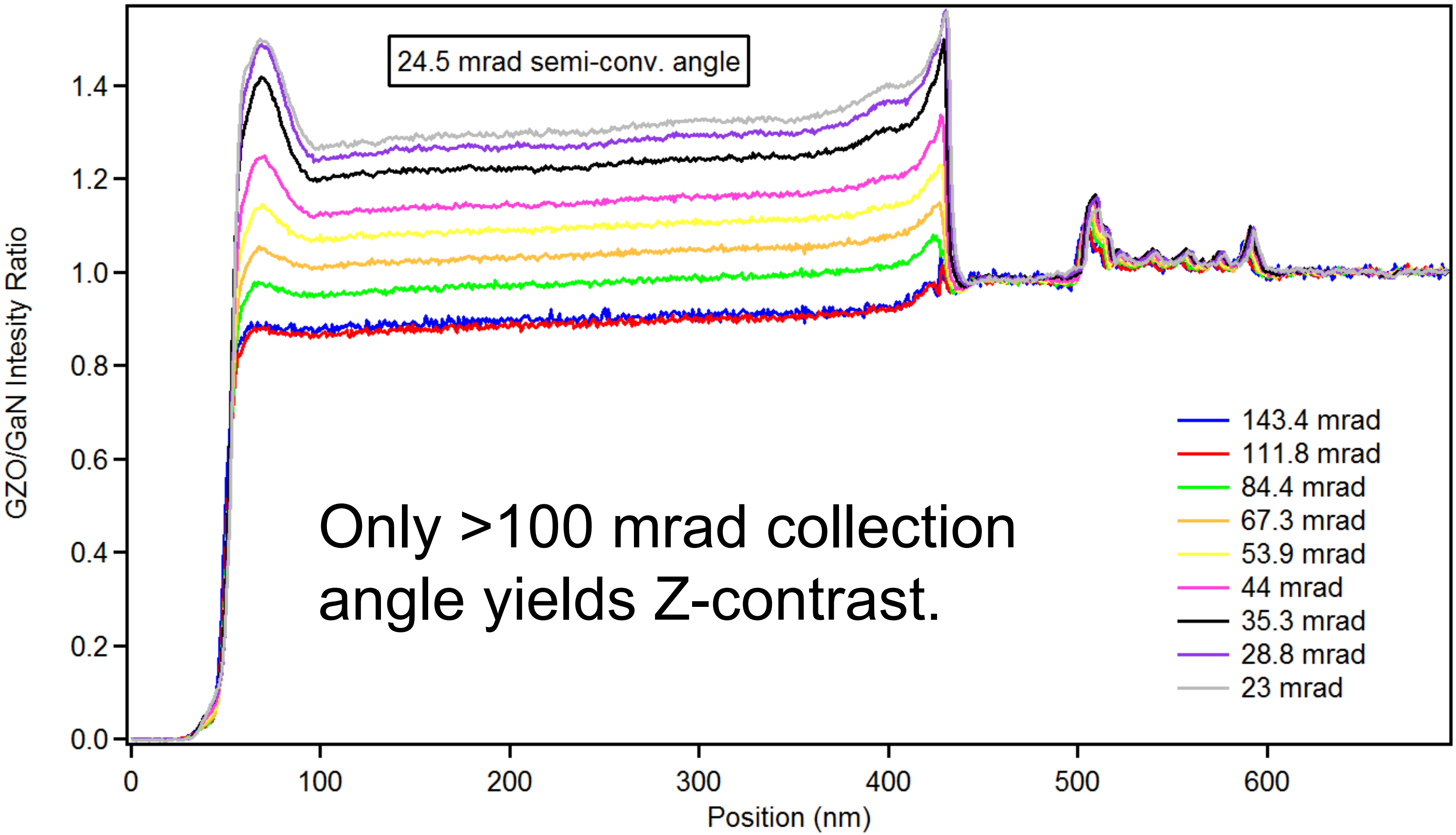
Changing Collection Angle

Change CL / HAADF inner angle at constant convergence angle of 24.5 mrad.
Outer angle 5× inner angle.



Paul Voyles, UWM

Collection Angle



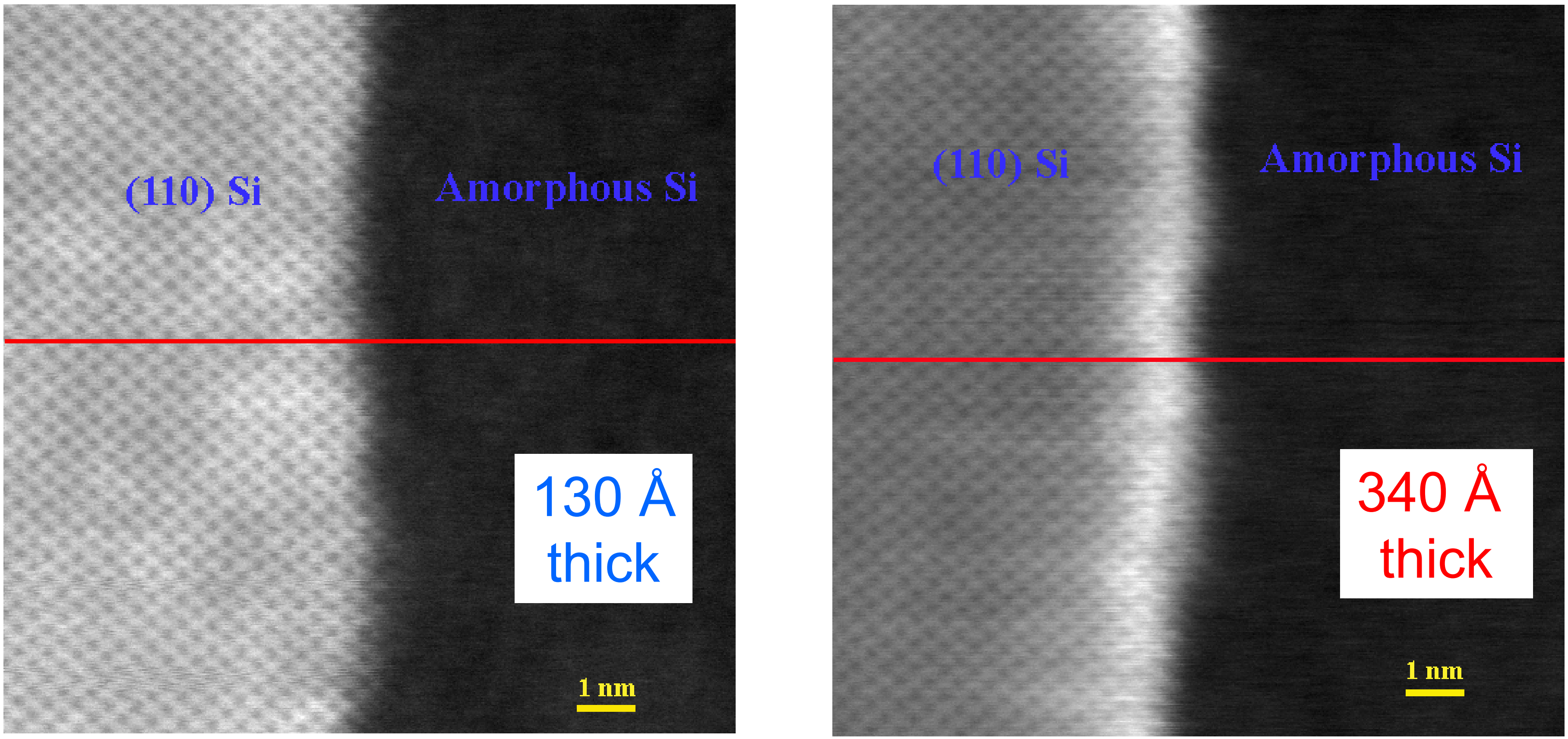
Only >100 mrad collection angle yields Z-contrast.

Paul Voyles, UWM

Strain Contrast vs. Sample Thickness

Contrast at a c-Si/-aSi is strongly depends on sample thickness

100 kV, 45 mrad ADF inner angle

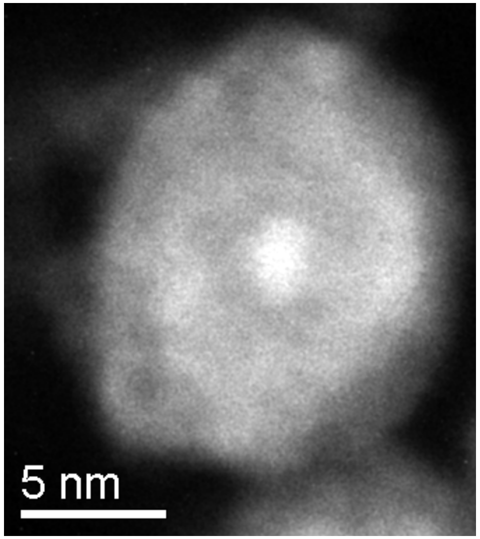


Strain Contrast effects at the interface:

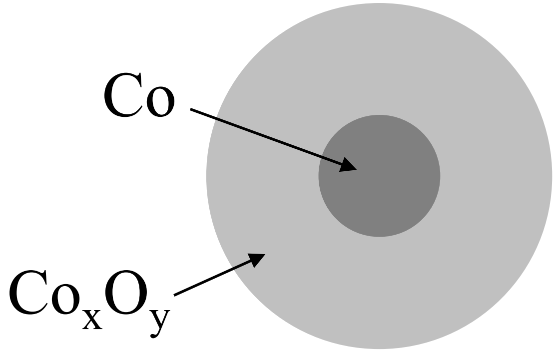
for 130 Å thick sample, ~0%; for 340 Å thick sample, 15%.

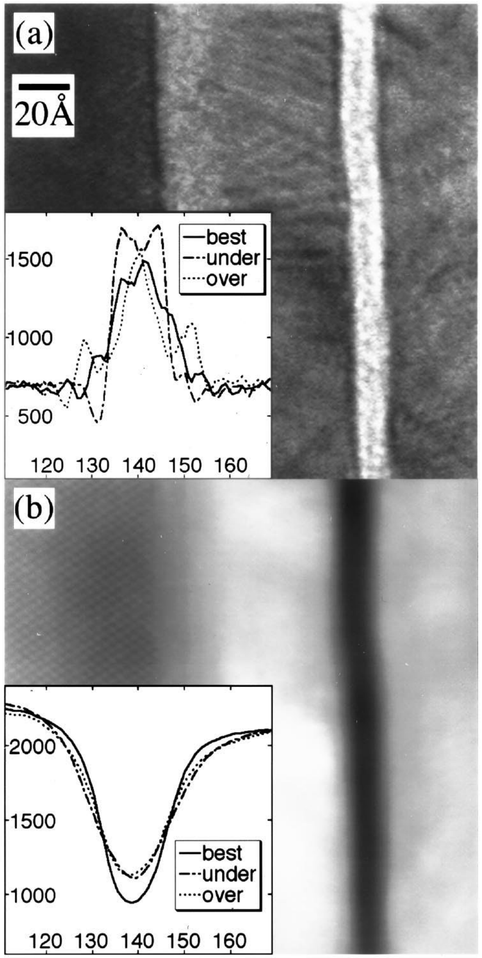
Z. Yu, D. A. Muller, and J. Silcox, *J. Appl. Phys.* **95**, 3362 (2004).

Examples: Low Resolution



oxidized Co nanoparticle





Co / AlO_x / Co tunnel junction

M. J. Plisch et al. APL **79**, 391 (2001).

Bi-Implanted Si

- A shows low-res BF
  - Can't really see implant
  - Can see damage
- B is Z-contrast image
  - Bi lights up like a Christmas Tree
  - The damage layer is not so visible.
    - No phase contrast

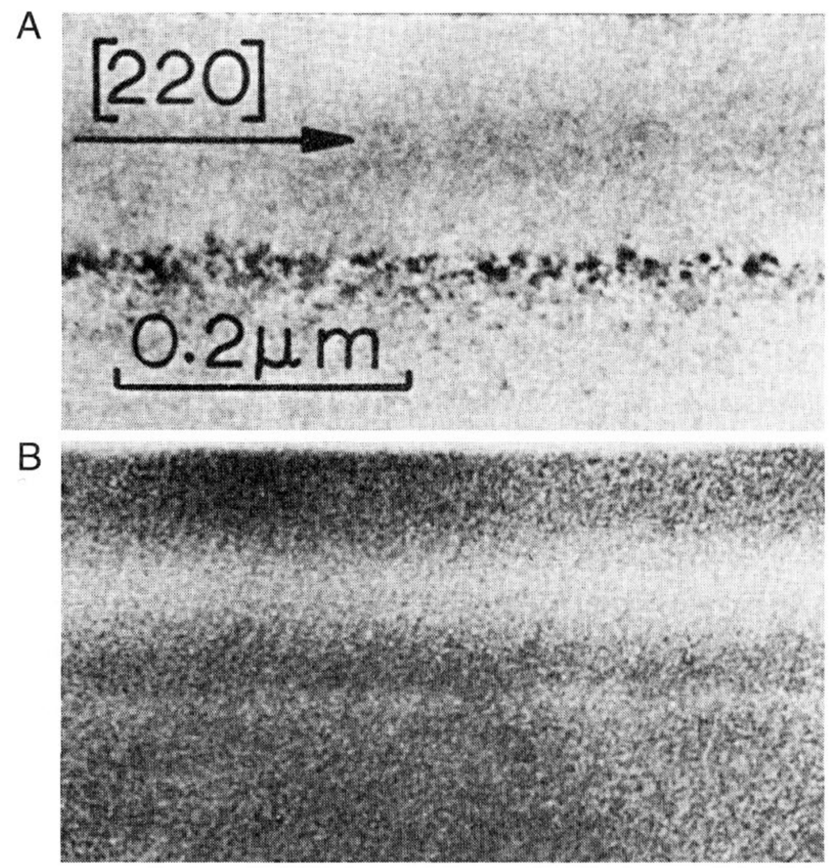
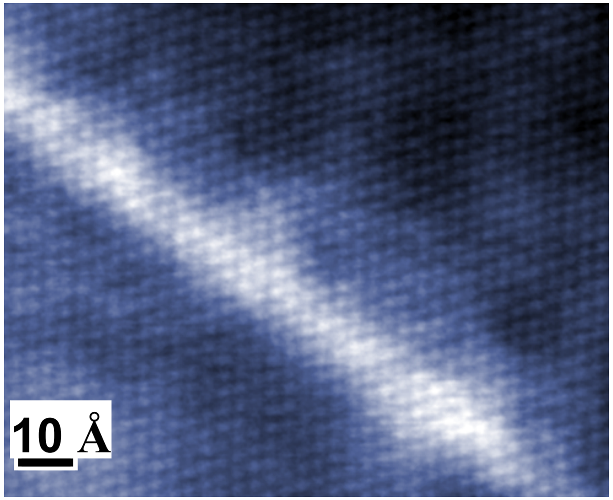


Figure 22.14. (A) Low-resolution TEM BF image showing a row of defects in Bi-implanted Si. In (B), obtained under *Z*-contrast conditions, the defects associated with the implant are invisible but the specimen is bright in the region implanted with Bi.

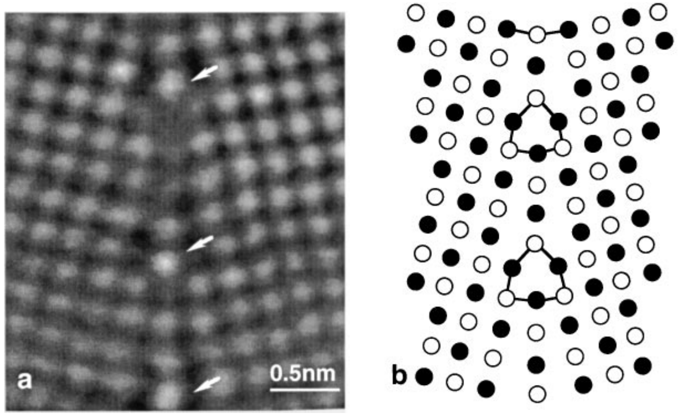
Examples borrowed from Williams & Carter

Examples: High Resolution



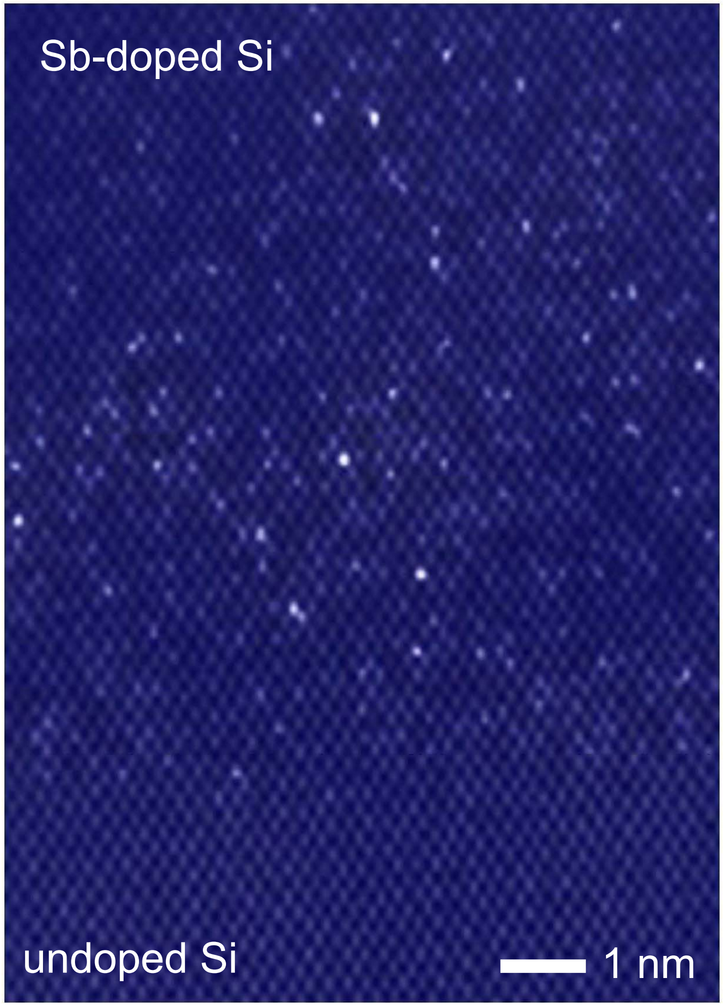
As δ-doped layer in Si

D. A. Muller PRL **83**, 3234 (1999)



Grain boundary in MgO

Y. Yan PRL **81**, 3675 (1998)

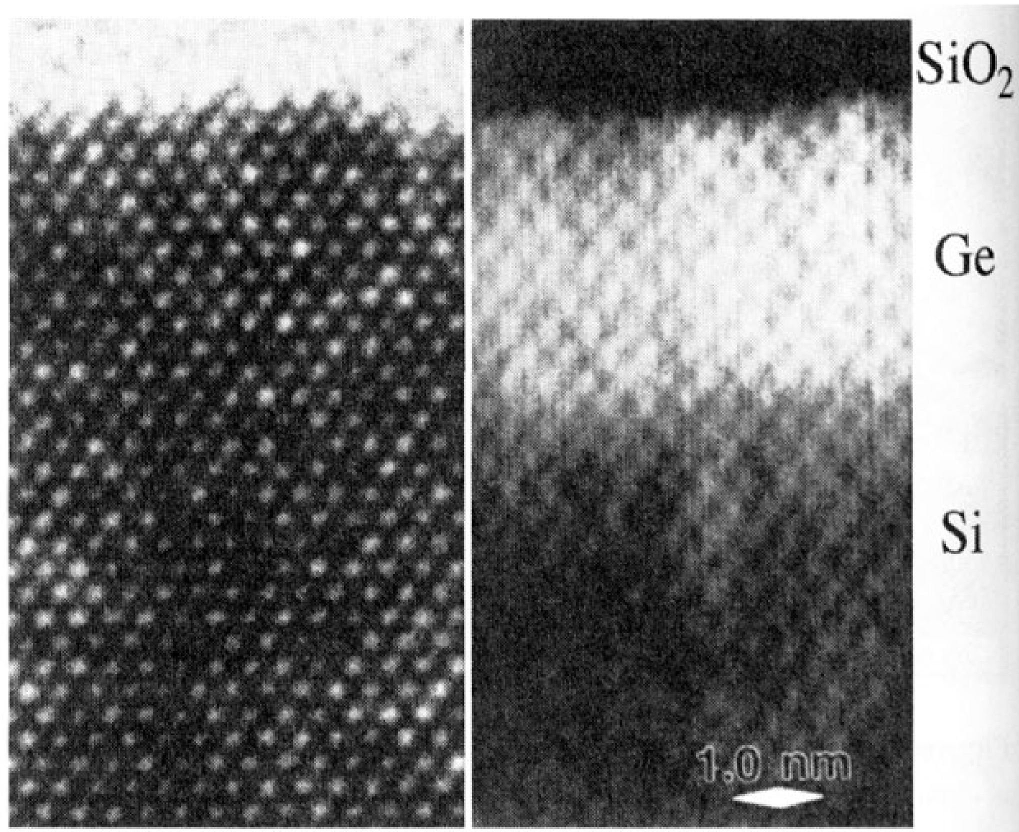


Individual Sb atoms and defect nanoclusters in Si

P. M. Voyles, Nature **416**, 826 (2002)

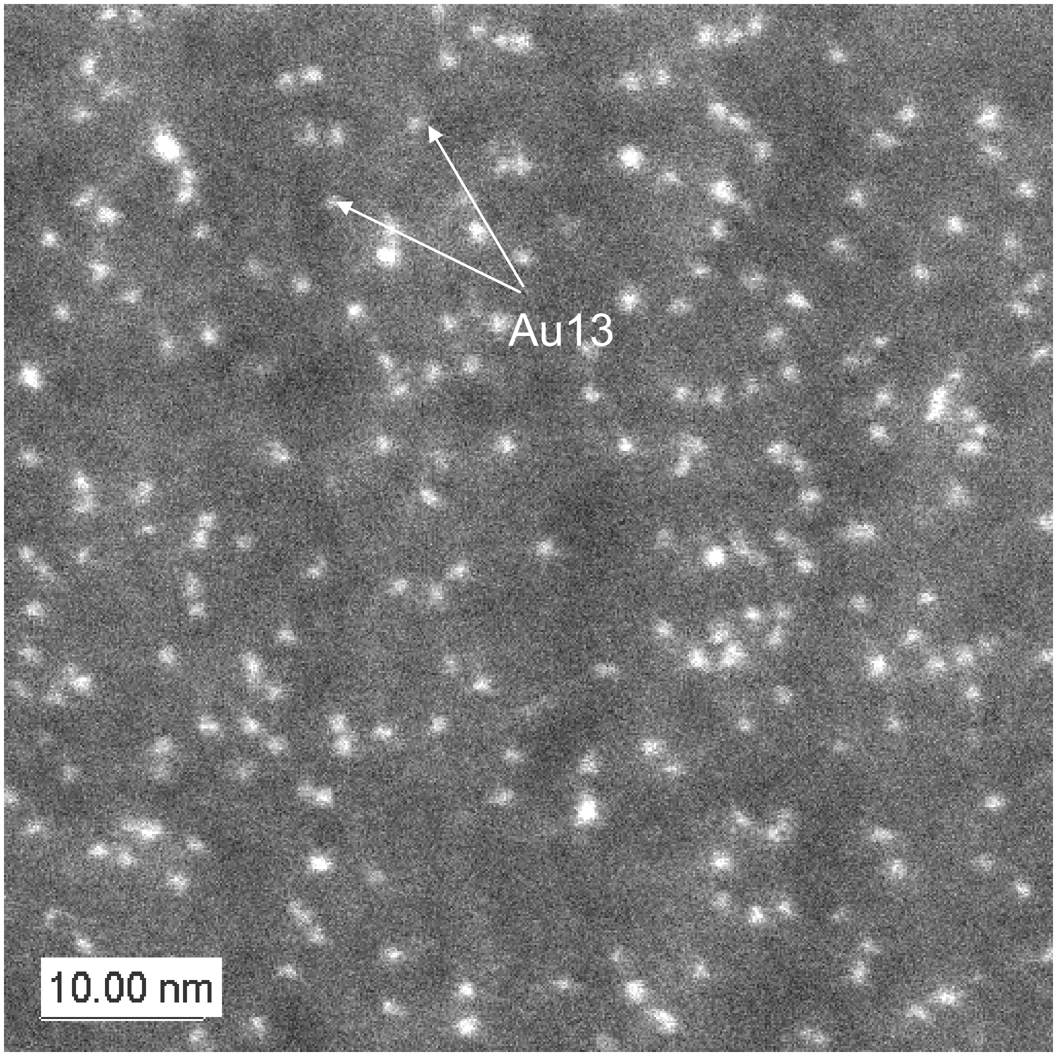
SiO_2 on Ge on Si

- Amorphous region is visible in hi-res
- Oxide is dark, Ge is light, Si in between - can see lattice



HAADF-STEM

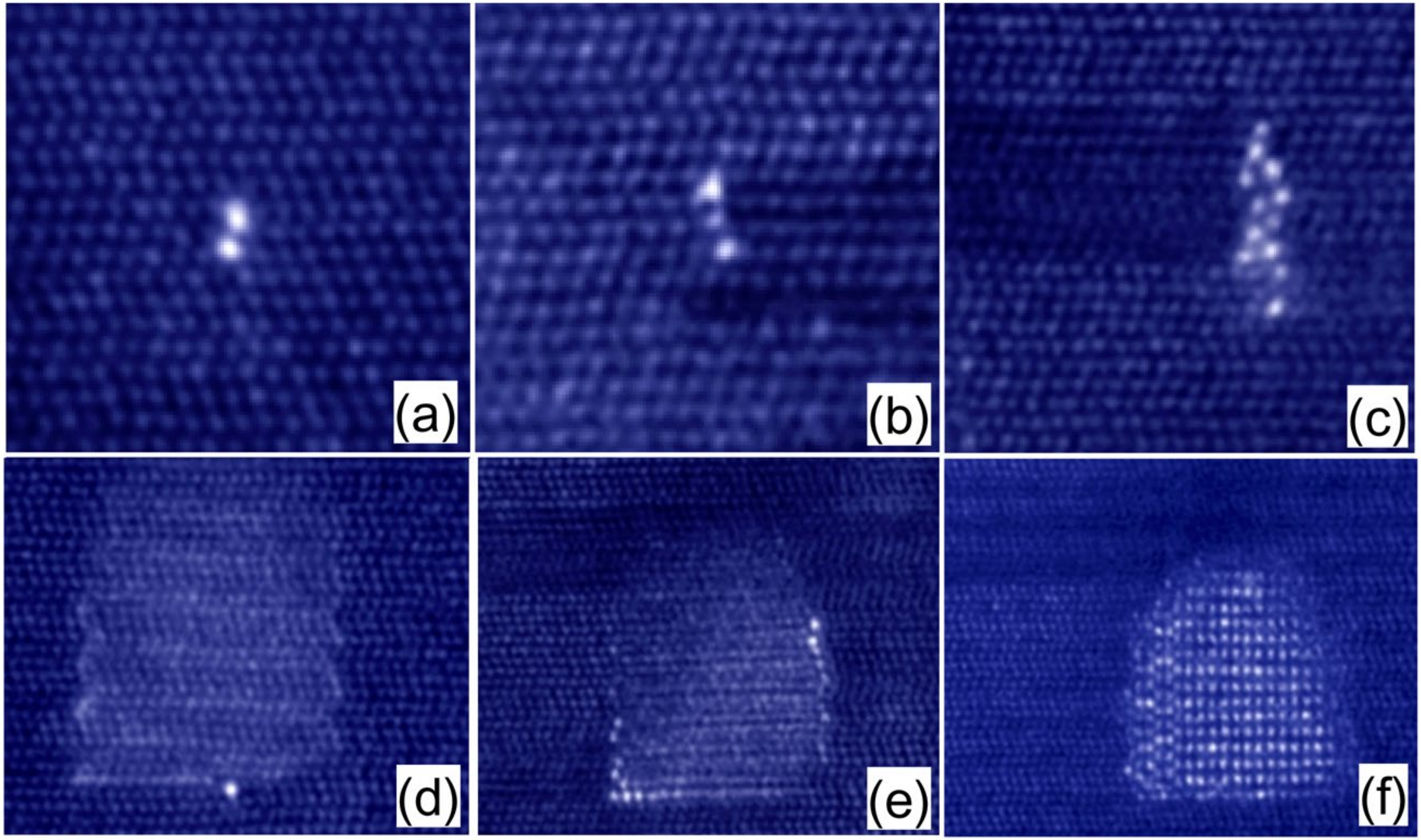
High-Angle Annular Dark-Field Scanning Transmission Electron Microscopy



Huiping Xu, Ray Twesten

A representative STEM-HAADF image (HB 501, 100kv, inner angle: 96mgrad) of sample $Au_{13}(PPh_3)_4(SC12)_4$

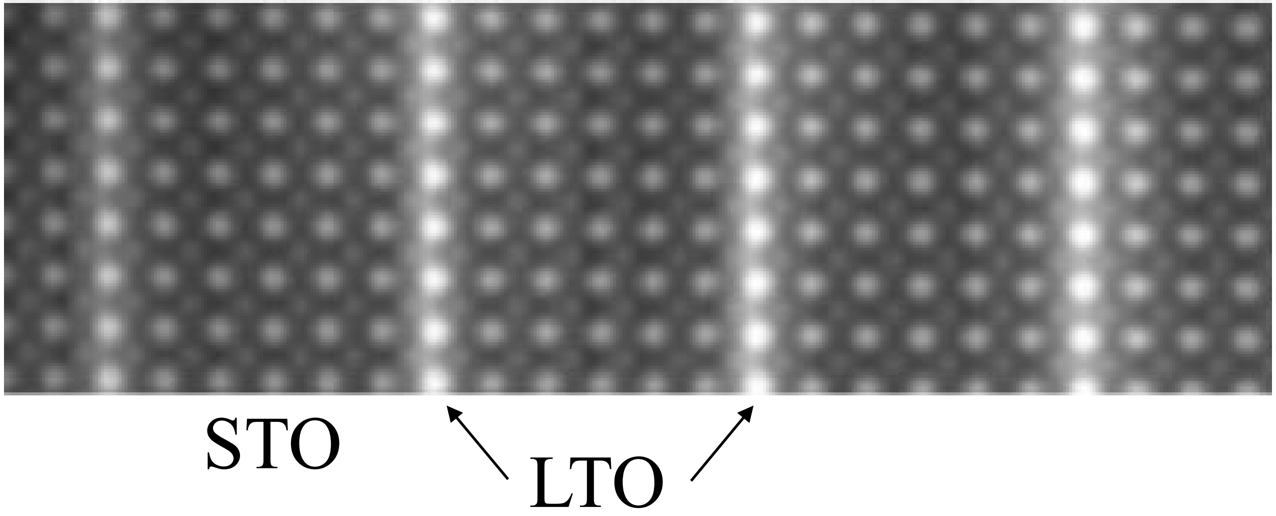
Examples



Growth of Er clusters in SiC on annealing.

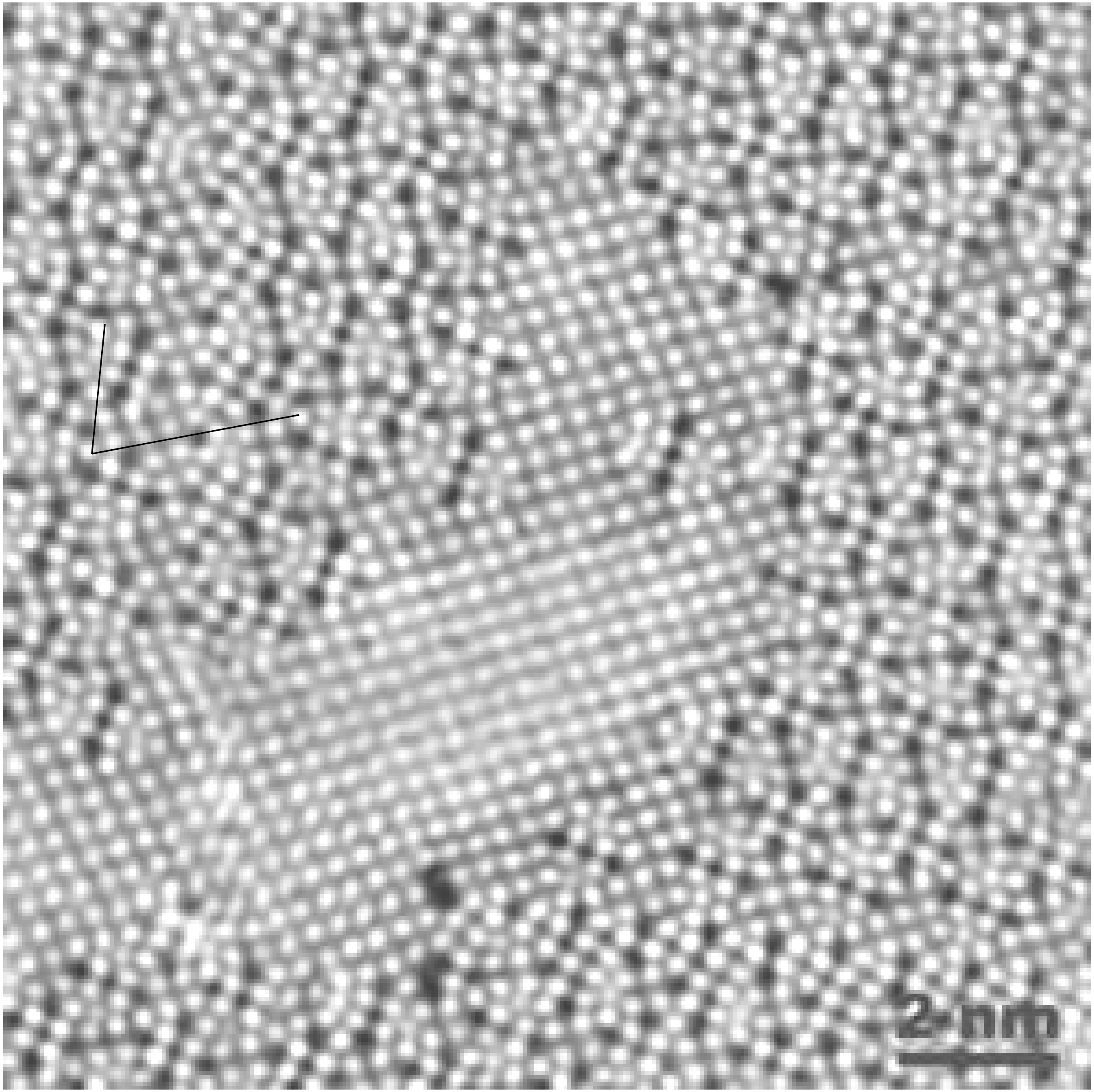
U. Kaiser, Nature Materials **1**, 102 (2002)

Examples



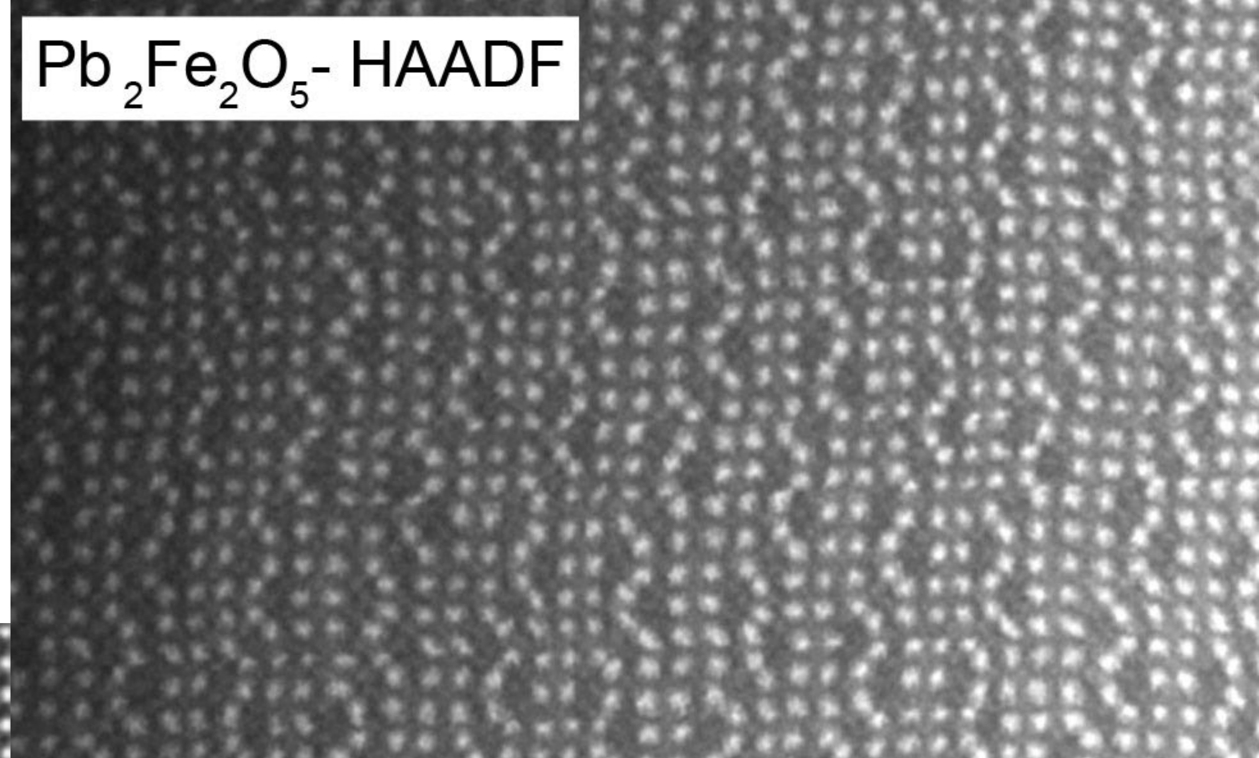
$SrTiO_3$ / $LaTiO_3$ multilayers

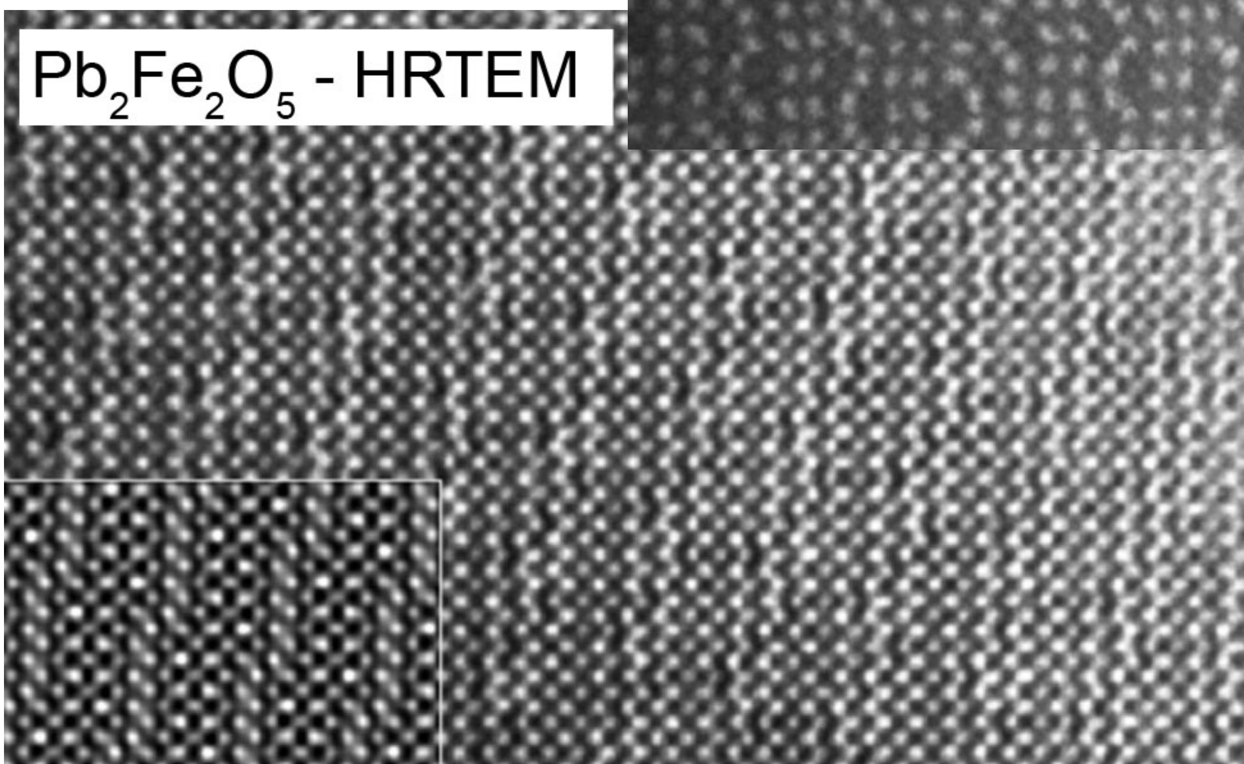
A. Ohtomo Nature **419**, 378 (2002)


2 nm

$$I = \beta Z^n$$

$Z_O = 8$
$Z_{Fe} = 26$
$Z_{Pb} = 82$

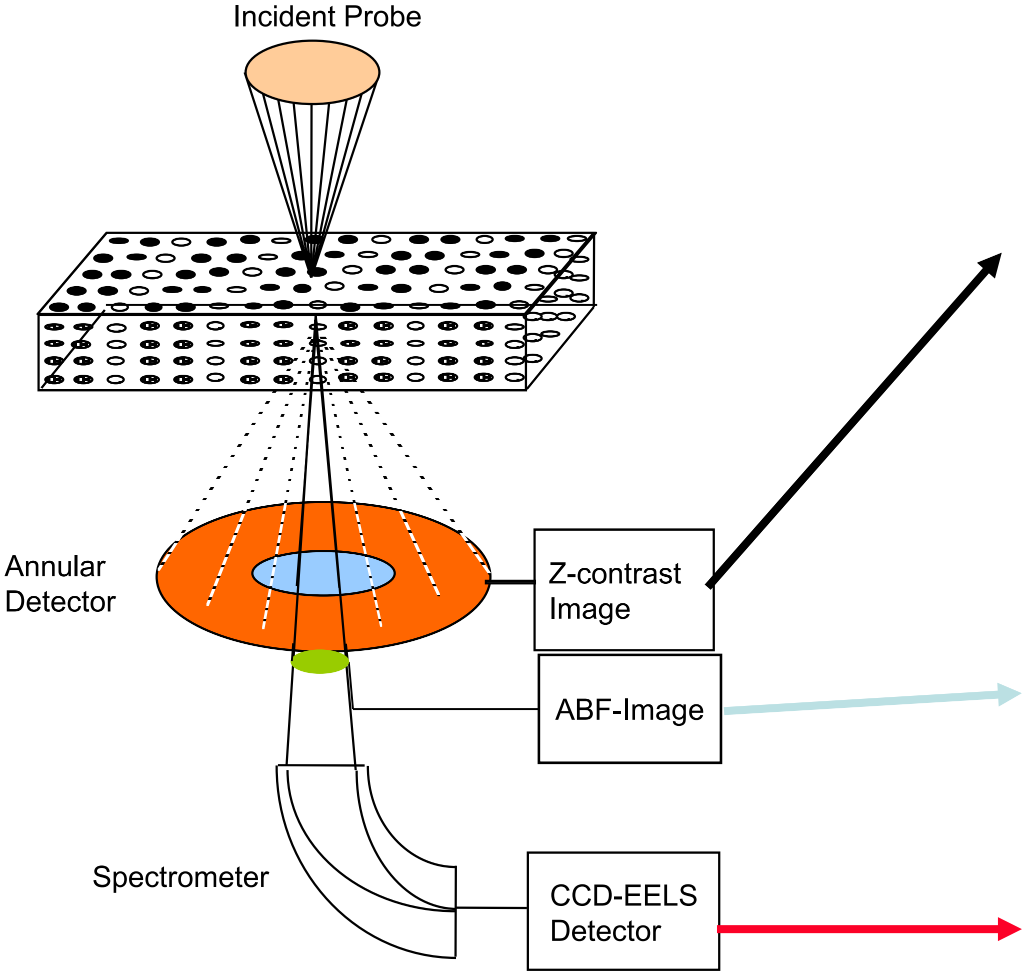


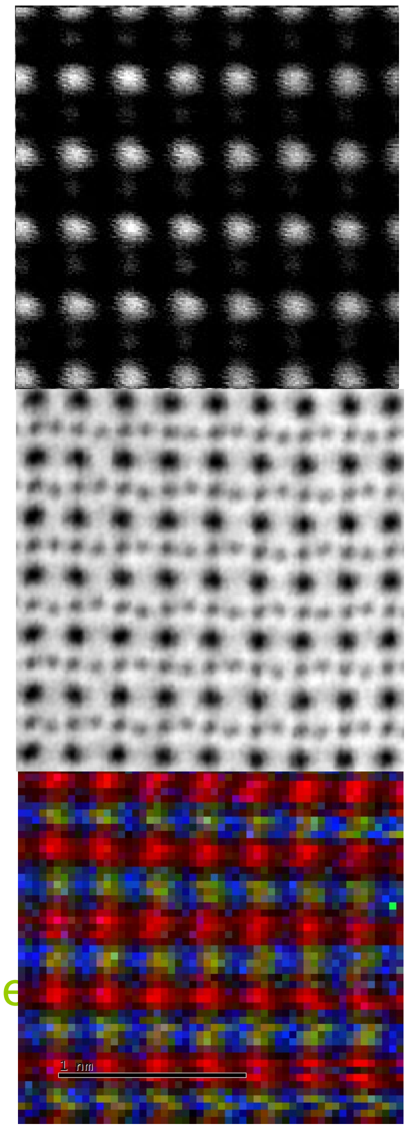


Abakumov et al.
Angewandte Chemie (2006)

ABF

Scanning Transmission Electron Microscope (STEM)

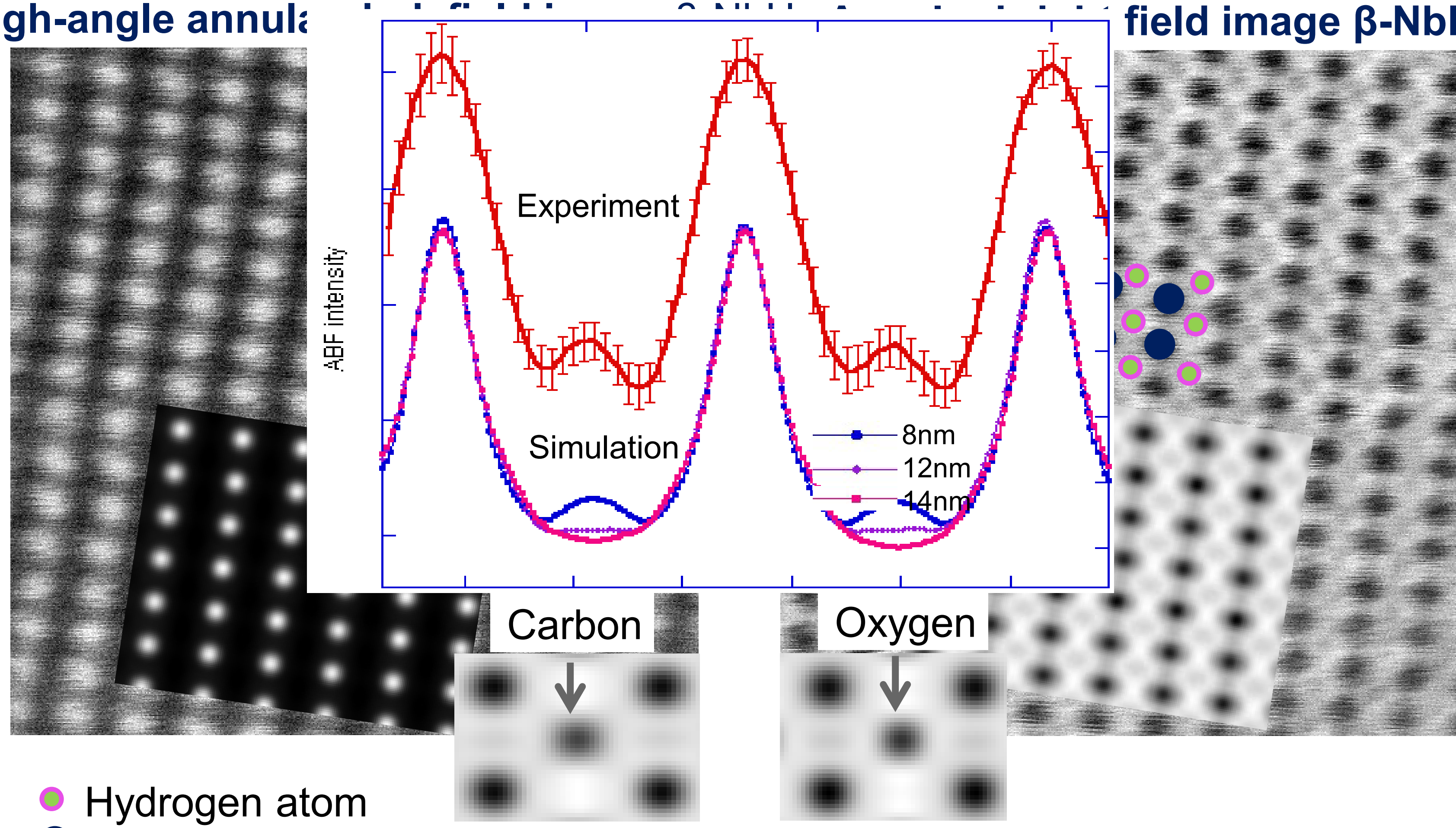




ABF detector can detect lighter ele

Direct imaging of Hydrogen in Nb:

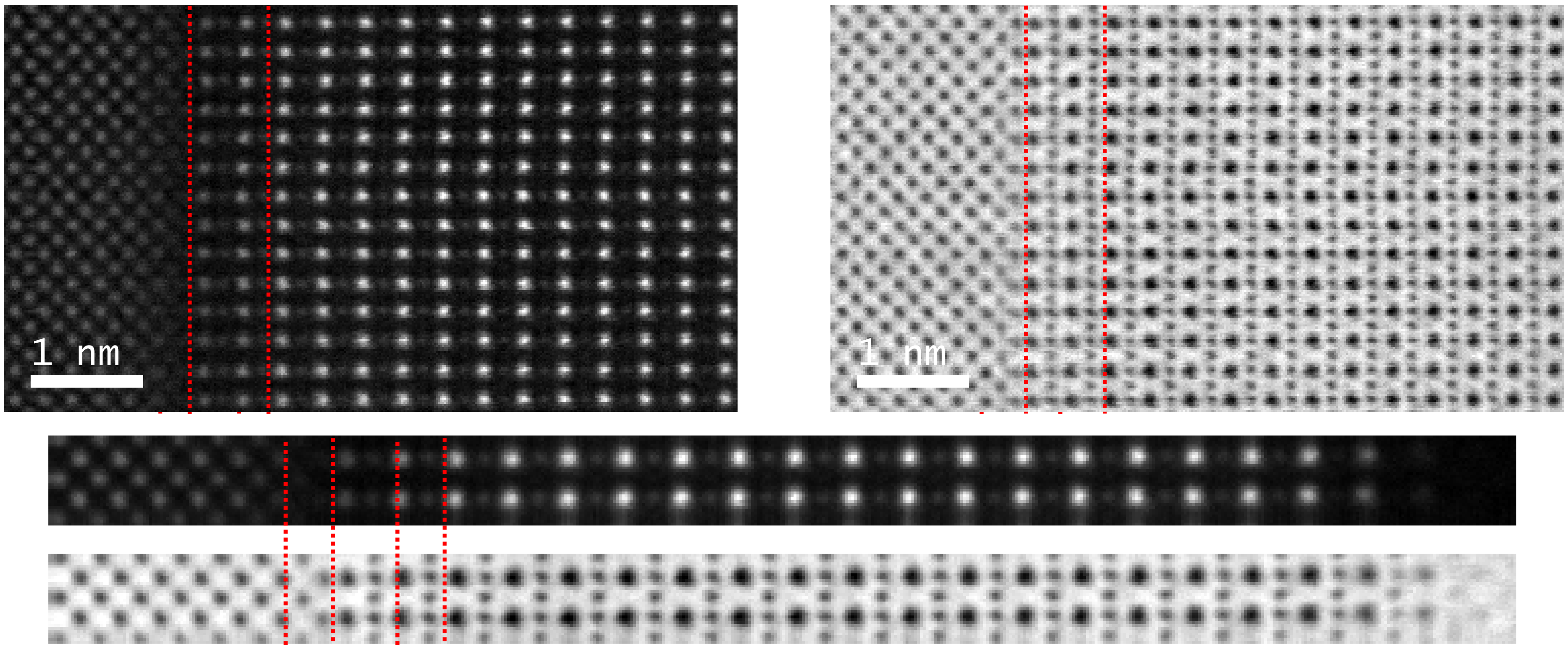
High-angle annul… field image β-NbH



Hydrogen atom
Niobium atom

YJ. Kim, R. Tao, R.F. Klie and D. Seidman, ACS nano, *7* (1), pp 732–739, (2013)

GaAs [110] substrate, $SrTiO_3$[100] buffer layer, $BaTiO_3$[100] film



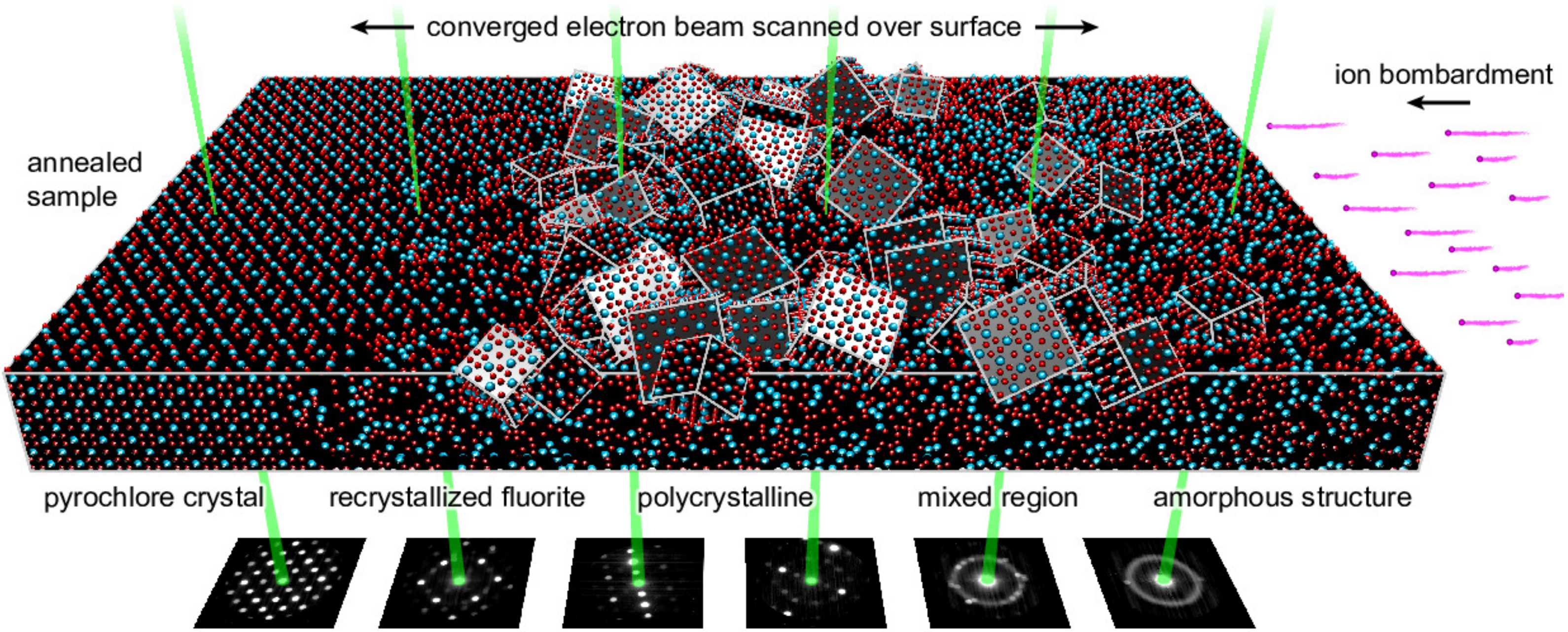
- Enlarged and averaged HAADF image shows As-SrO atomic registry
- No double layer As from c-(4x4) observed
- Visible oxygen vacancies in 1st layer SrO
- Visible displacement of O columns in STO buffer layer

4D STEM

Current Bleeding-Edge

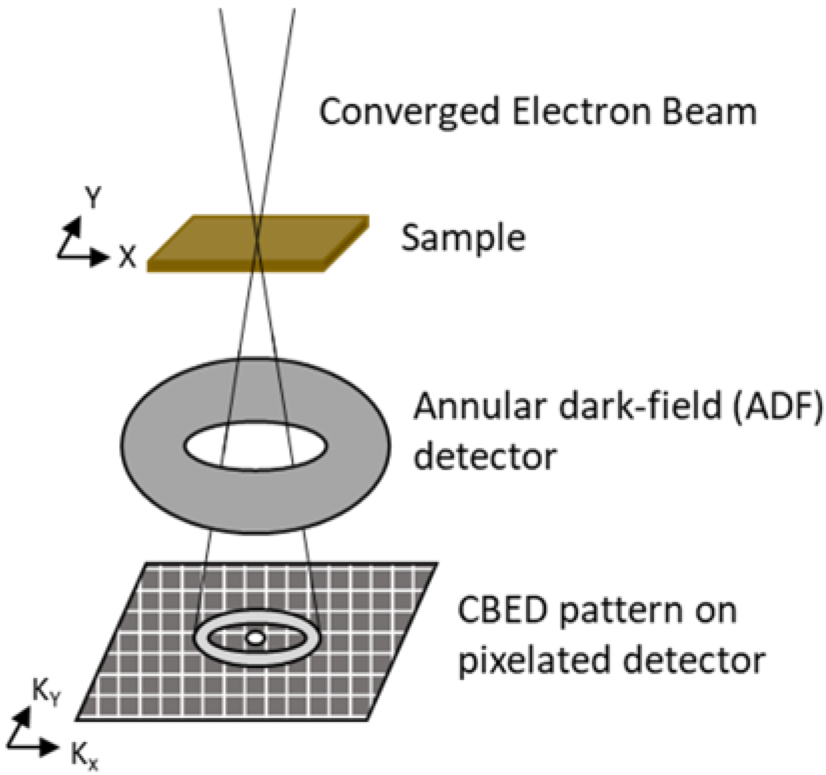
It will cut you if you get it wrong

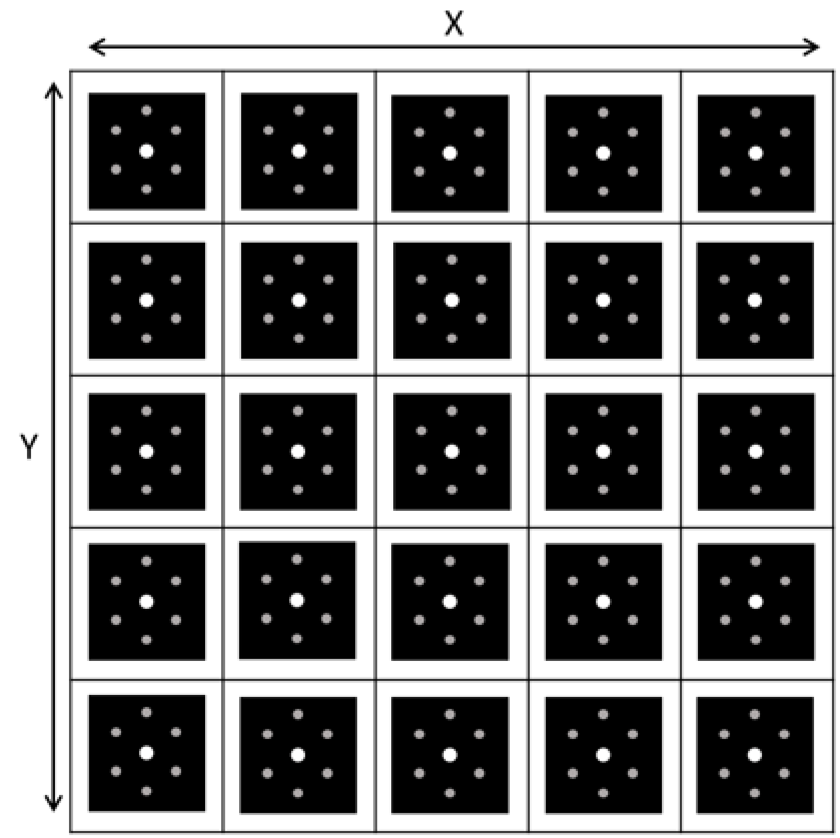
A 4D-STEM Experiment Involves Data Collection in Two Real and Two Reciprocal Space Dimensions



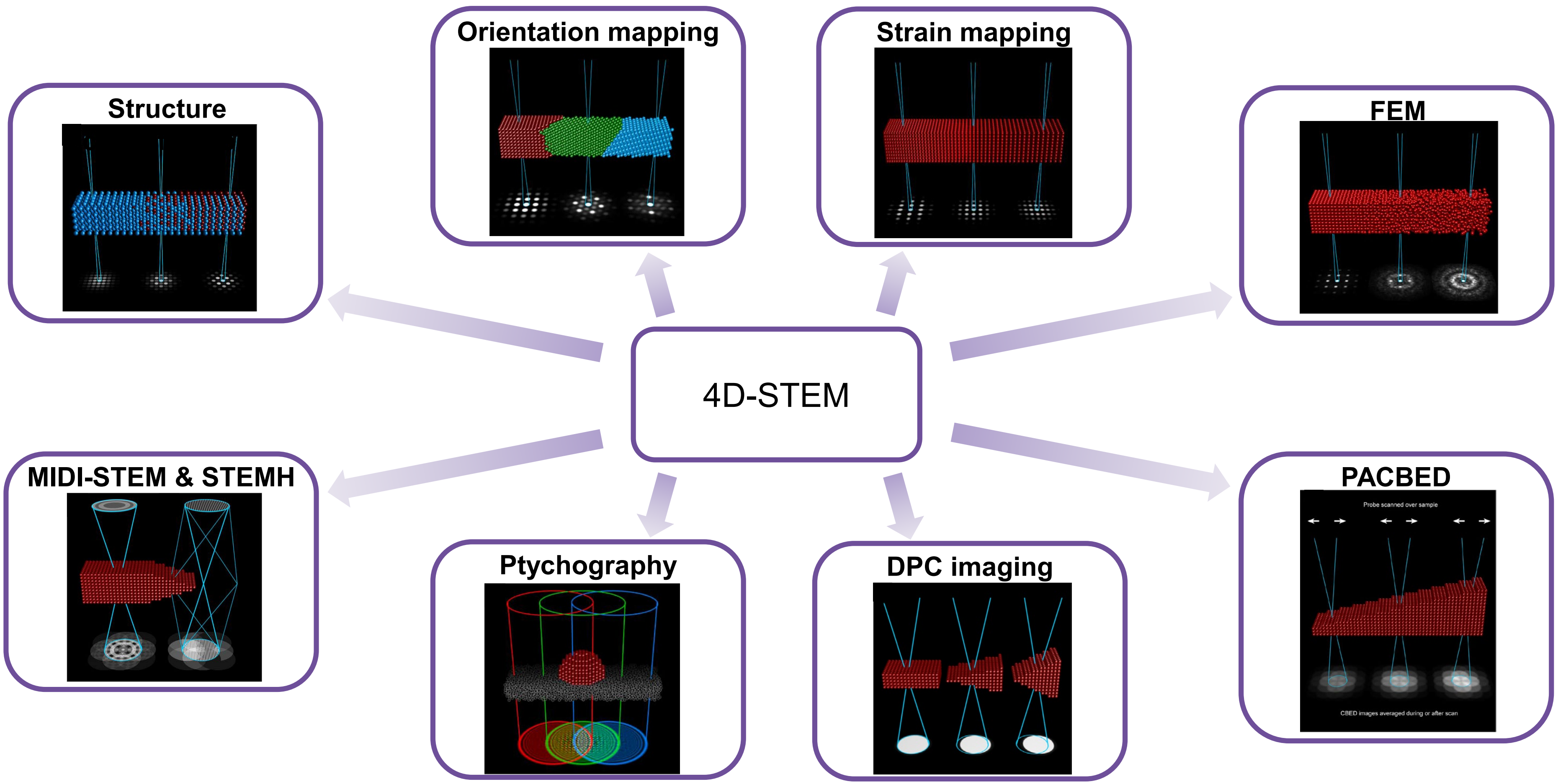
Sample: irradiated $Gd_2Ti_2O_7$
Savitzky, B.H.; Ophus, C.; *et al.*, *arXiv preprint arXiv:2003.09523* **2020.**

CBED at every point

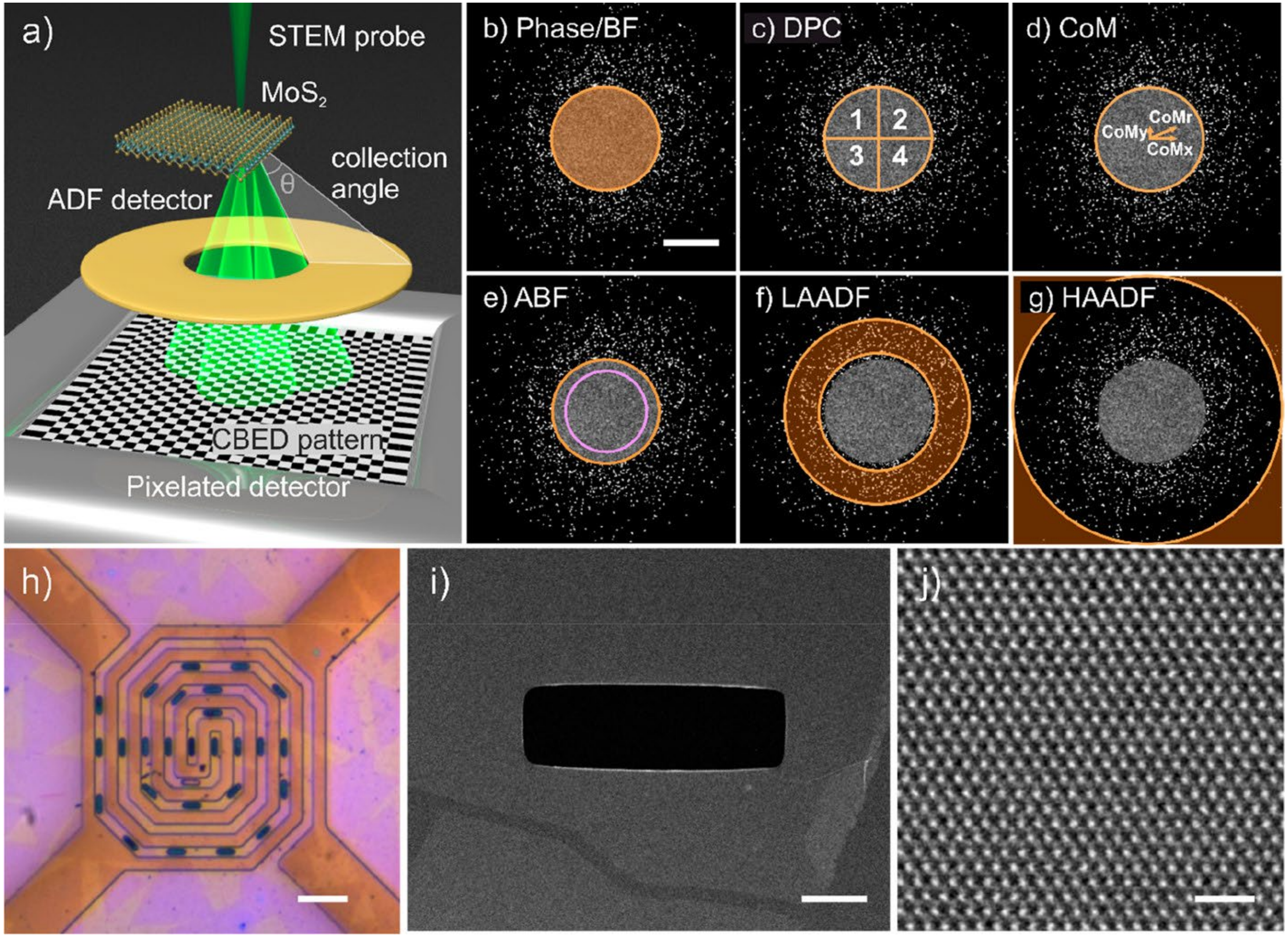




4D-STEM Can Be Applied to a Variety of Techniques

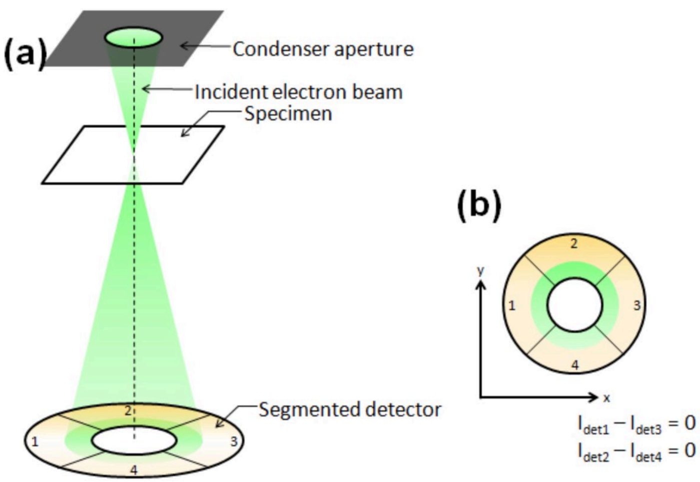


Ophus, C., *Microsc. Microanal.* **2019.**

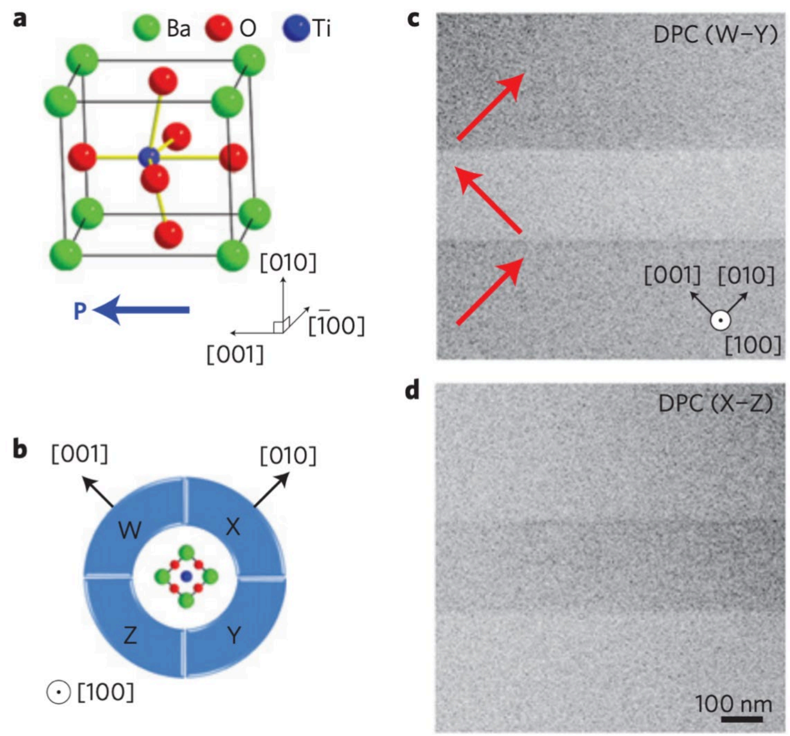


DOI: (10.1021/acs.nanolett.9b02717)

Differential Phase Contrast



DPC detector schematic



Application in measuring polarization

www.jeol.co.jp
Shibata, N.; Findlay, S. D.; Kohno, Y.; Sawada, H.; Kondo, Y.; Ikuhara, Y. *Nature Physics* **2012**, *8*, 611–615.

Look at differences – yields fields & derivatives (kind-off)

Differential Phase Contrast

nature COMMUNICATIONS

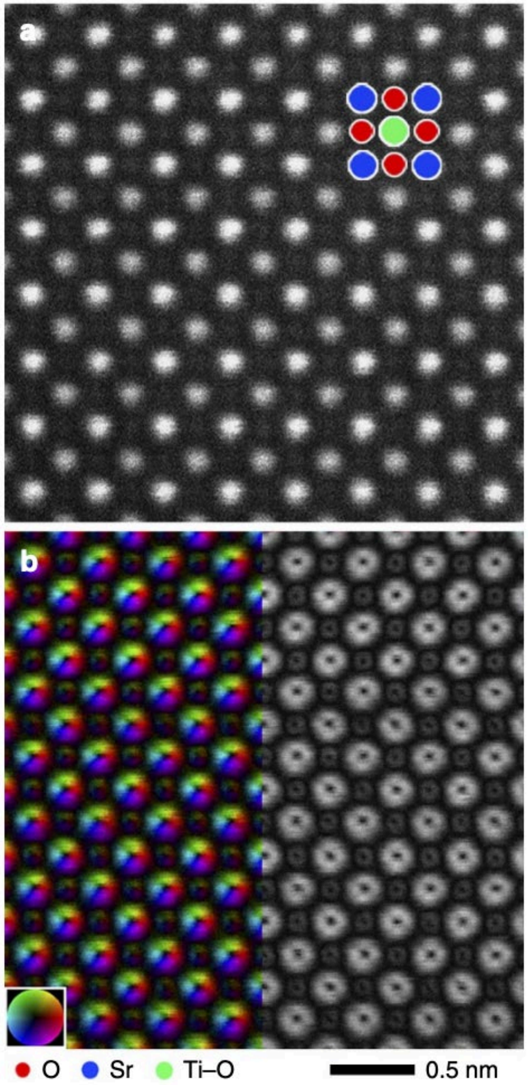
ARTICLE

Received 11 Jan 2017 | Accepted 14 Apr 2017 | Published 30 May 2017

DOI: 10.1038/ncomms15631 OPEN

Electric field imaging of single atoms

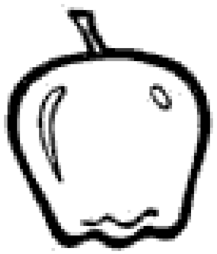
Naoya Shibata[1], Takehito Seki[1], Gabriel Sánchez-Santolino[1], Scott D. Findlay[2], Yuji Kohno[3], Takao Matsumoto[1], Ryo Ishikawa[1] & Yuichi Ikuhara[1,4]



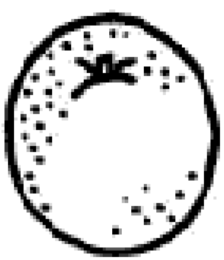
The Phase Problem

- We have an exit wave from the sample
  - $\psi(r)$ wave in real space = $a(r)\exp(-i\phi(r))$
  - $\Psi(u) = \int \exp(-2\pi i u.r)\psi(r)dr = A(u)\exp(-i\phi(u))$
- Observables
  - $I(r) = \langle|\psi(r)|^2\rangle = \langle a(r)^2\rangle$ Real Space Image
  - $I(u) = \langle|\Psi(u)|^2\rangle = \langle A(u)^2\rangle$ Diffraction Pattern
- Note: “<>” is average over incoherent aberrations and other statistical terms

Phase: Apples & Oranges



FT → $A_a \exp(-i\ \phi_a)$



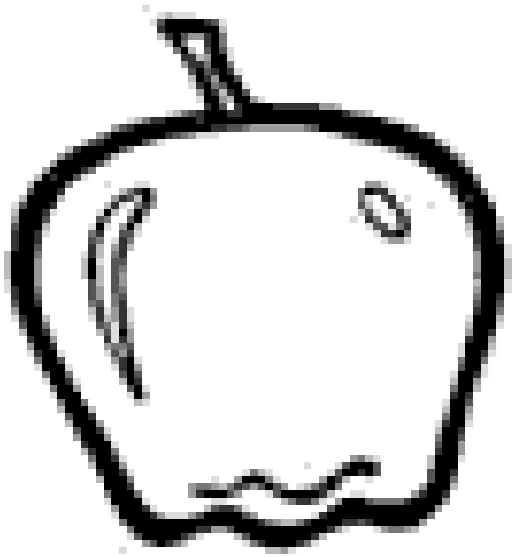
FT → $A_o \exp(-i\ \phi_o)$

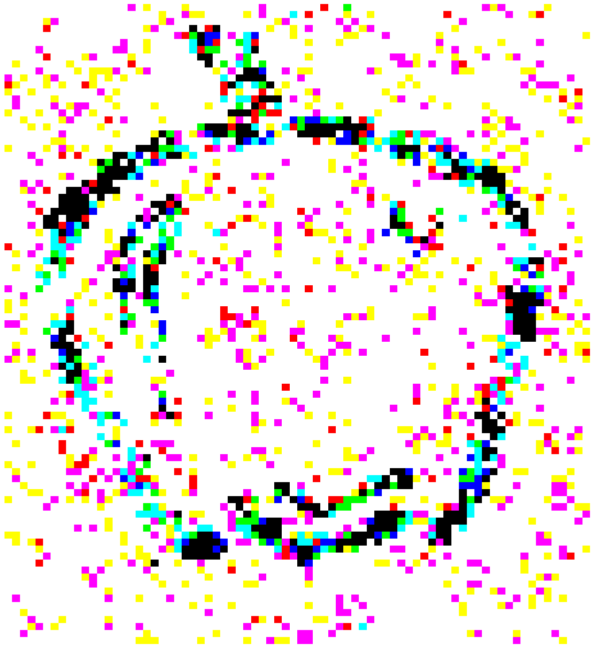
+

$A_o \exp(-i\ \phi_a)$ → IFT → { Oranle ? / Appge ? }

Phase of Apple + Amplitude of Orange = ?

Phase of Apple = Apple



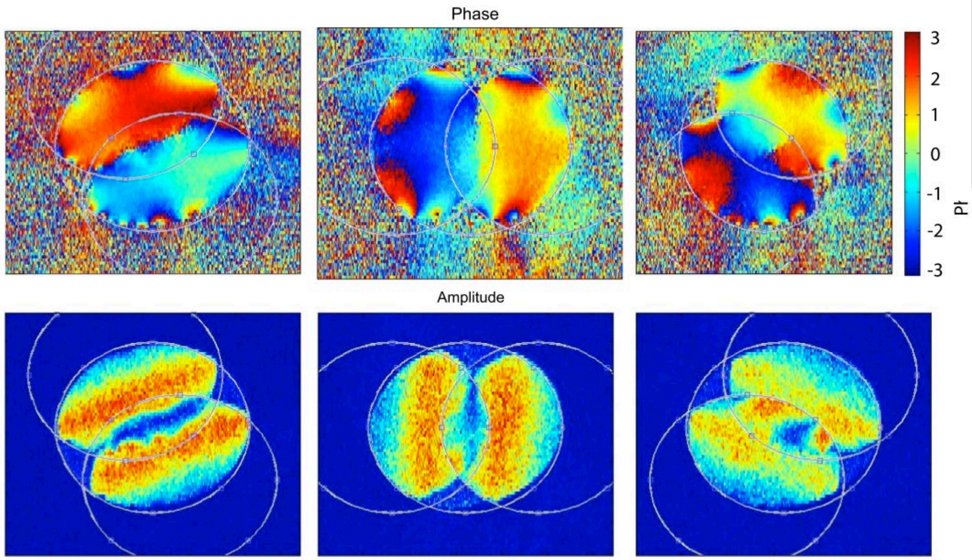


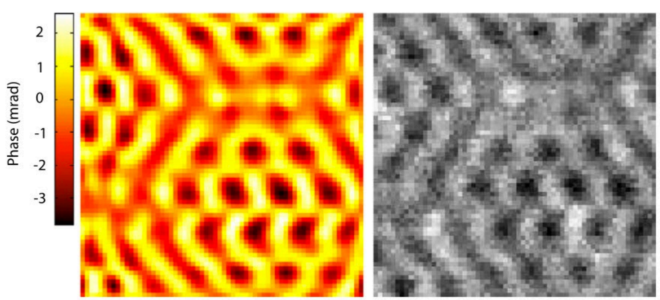
$FT^{-1}\ \{A_o\ \underline{\exp(-i\ \phi_a)}\ \}$ ⇒ Apple

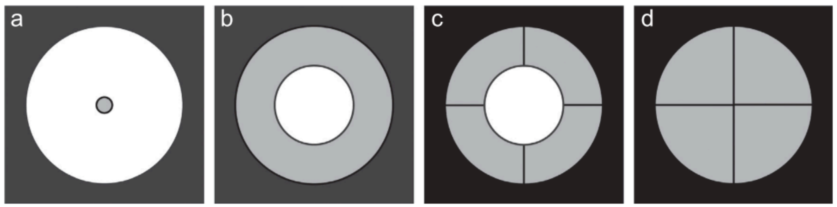
Phase is more important than amplitude

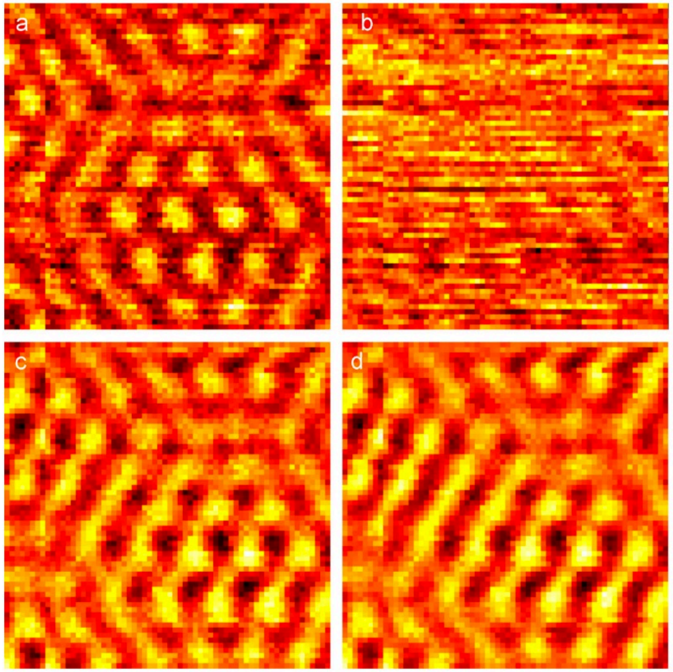
Ptychography

Analysis of 4D dataset



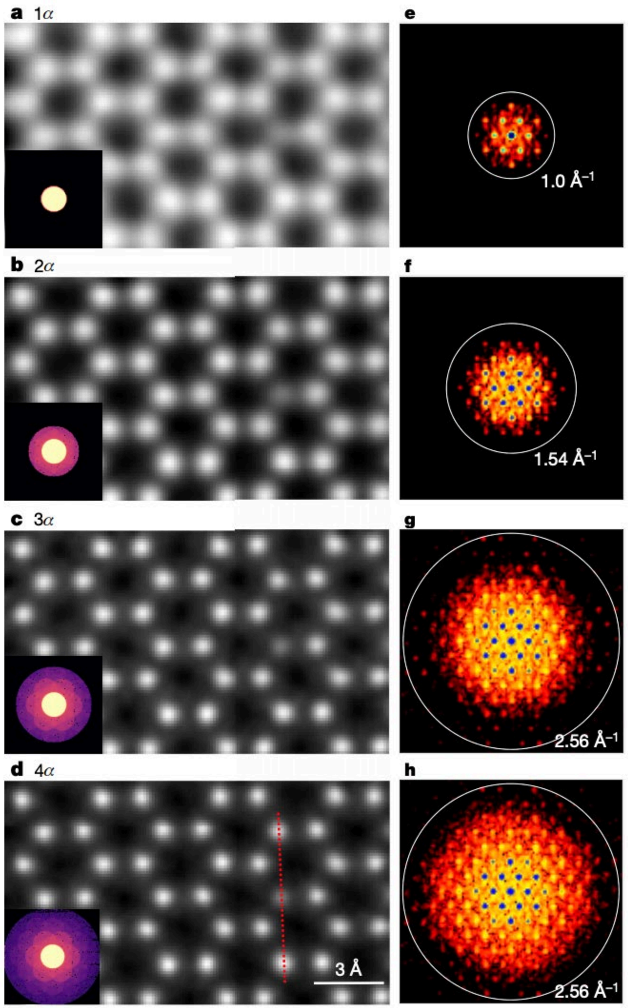


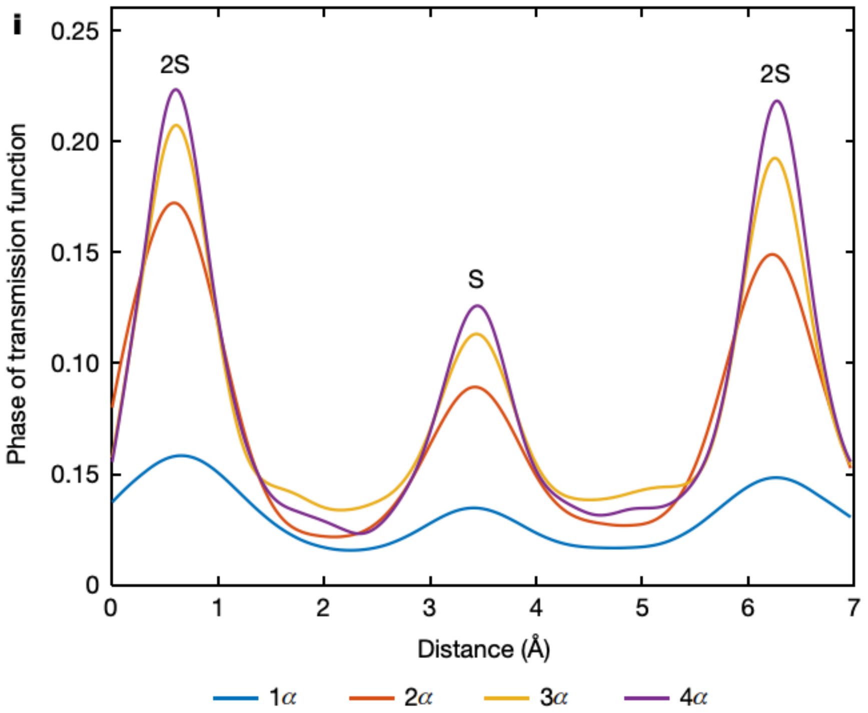




Kim, Y. S. *et al.* Localized electronic states induced by defects and possible origin of ferroelectricity in strontium titanate thin films. *Appl. Phys. Lett.* **94**, 1–4 (2009).

Ptychography

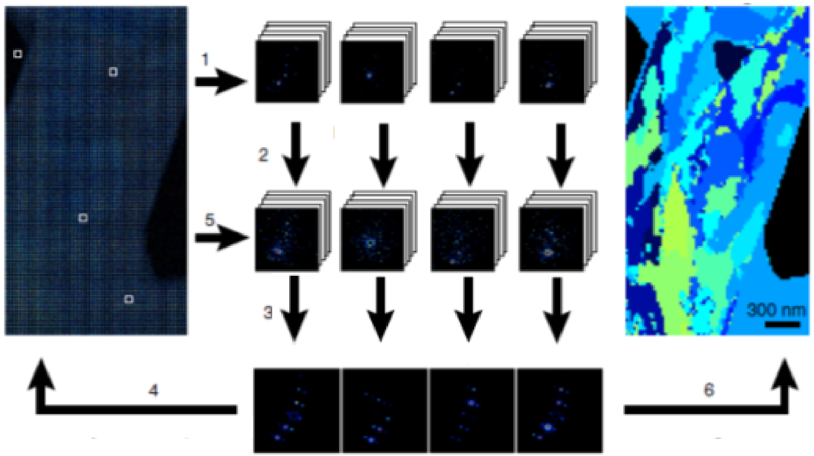




Jiang, Y., Chen, Z., Han, Y. *et al.* Electron ptychography of 2D materials to deep sub-ångström resolution. *Nature* **559,** 343–349 (2018) doi:10.1038/s41586-018-0298-5

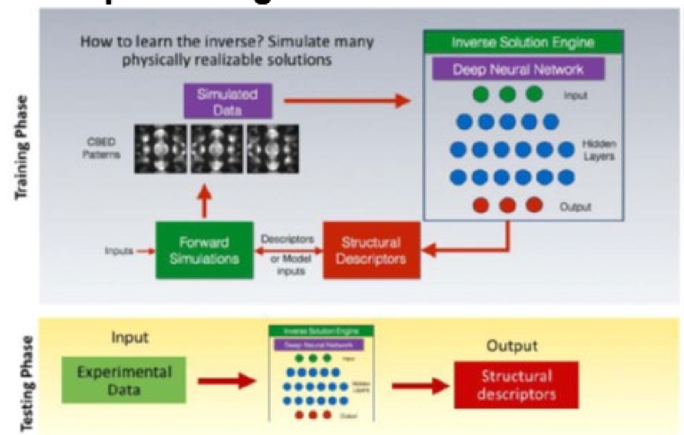
What's Next? Automation of Data Acquisition and Processing

Unsupervised data clustering



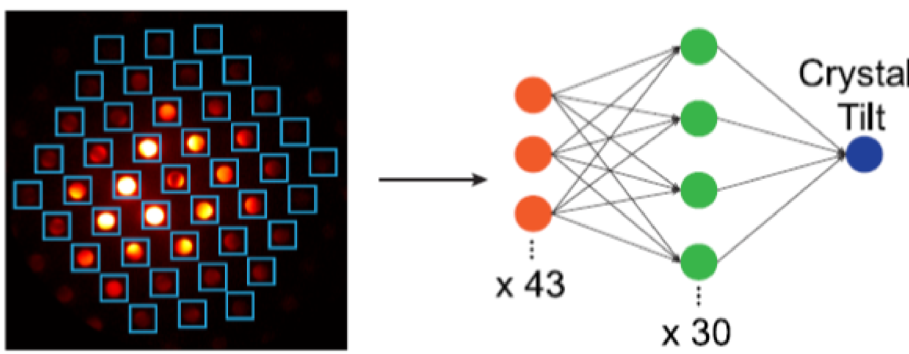
Gallagher-Jones, M.; Rodriguez, J. A.; *et al.*, *Comm. Biol.* **2019.**

Deep learning of interface structures



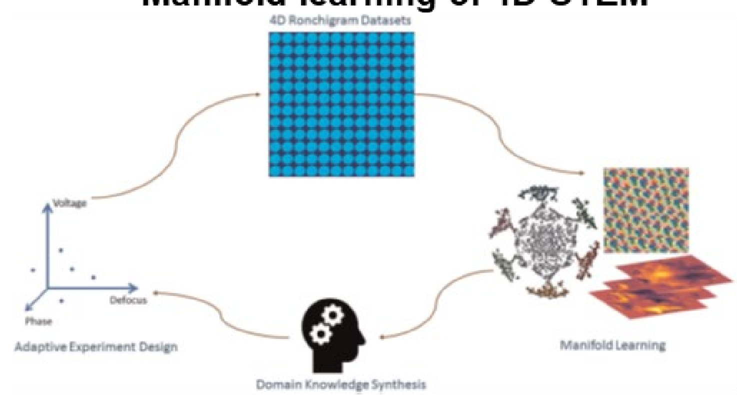
Oxley, M. P.; Kalinin, S. V.; *et al.*, *Mach. Learn.: Sci. Technol.* **2020.**

Artificial neural network



Yuan, R.; Zuo, J.-M.; *et al.*, *Ultramicroscopy* **2021.**

Manifold learning of 4D-STEM



Li, X.; Kalinin, S. V.; *et al.*, *Npj Comput. Mater.* **2019.**

Summary: STEM

- Similar signals to TEM, often easier to obtain some types
- Parallel collection of different signals, but serial detection
- Easier for EELS/EDS mapping and similar
- Annular dark-field is not as simple as often thought, serious misinterpretations exist in the current literature
- Still under development