TEM
Overview and Challenges

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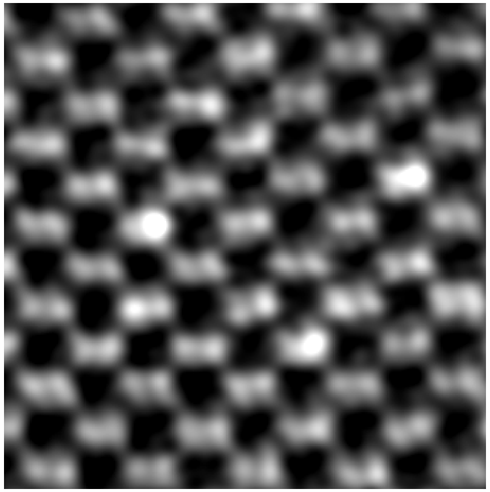
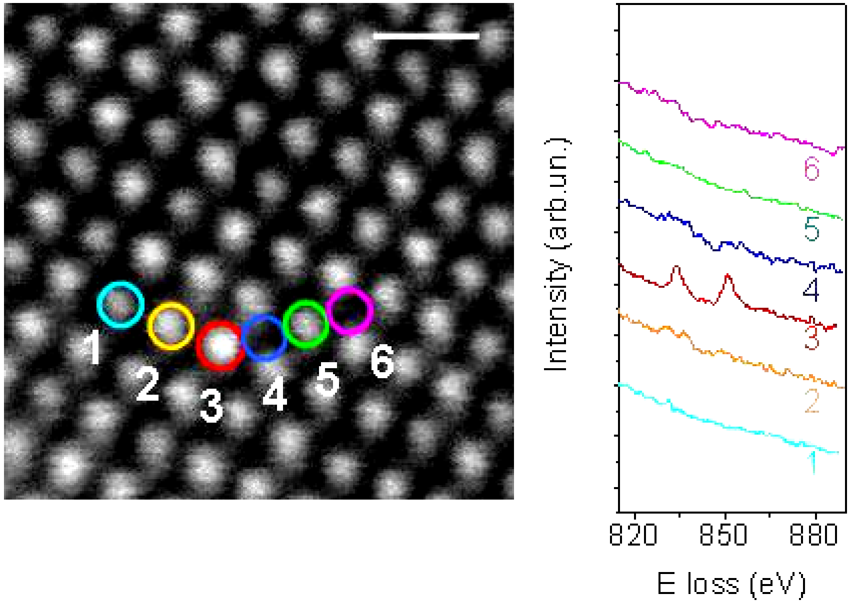
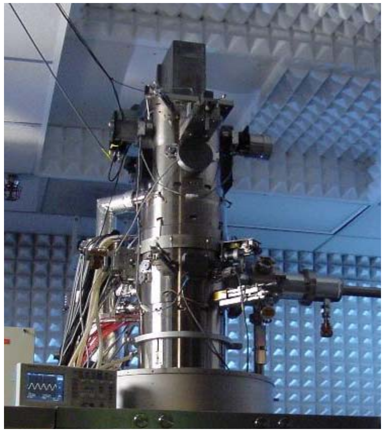


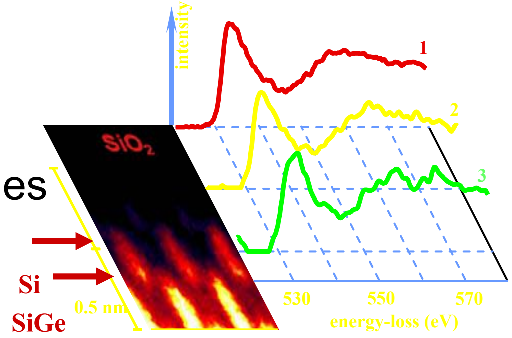


Research funded by DOE BES Division of Materials Sciences

Collaborators:



Electron Microscopy:	ORNL:	M. F. Chisholm M. Varela A. R. Lupini Y. Peng A. Borisevich
		O. L. Krivanek P. D. Nellist N. Dellby
Semiconductor Interfaces:	ORNL/NCSU:	G. Duscher S. Lopatin
Materials Theory:	Vanderbilt/ORNL:	S. T. Pantelides R. Buczko
Imaging Theory:	University of Melbourne:	L. J. Allen S. Findlay M. Oxley



Challenges for electronic materials

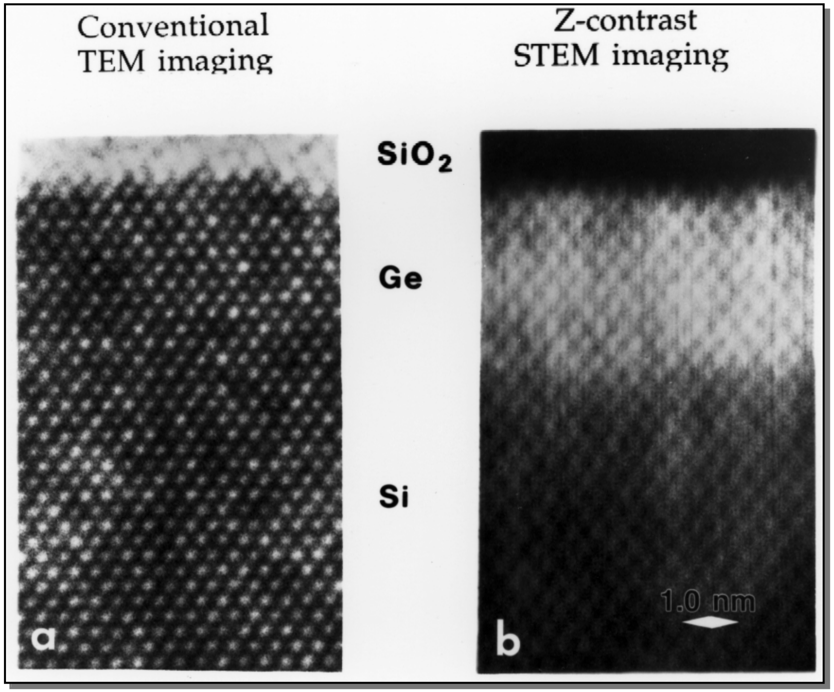
- Gate Dielectrics
  - Thickness: geometric vs. electronic
  - Stoichiometry and crystallinity
- Single atom detection
  - Image
  - Spectroscopy: localized states
- 3D tomography?

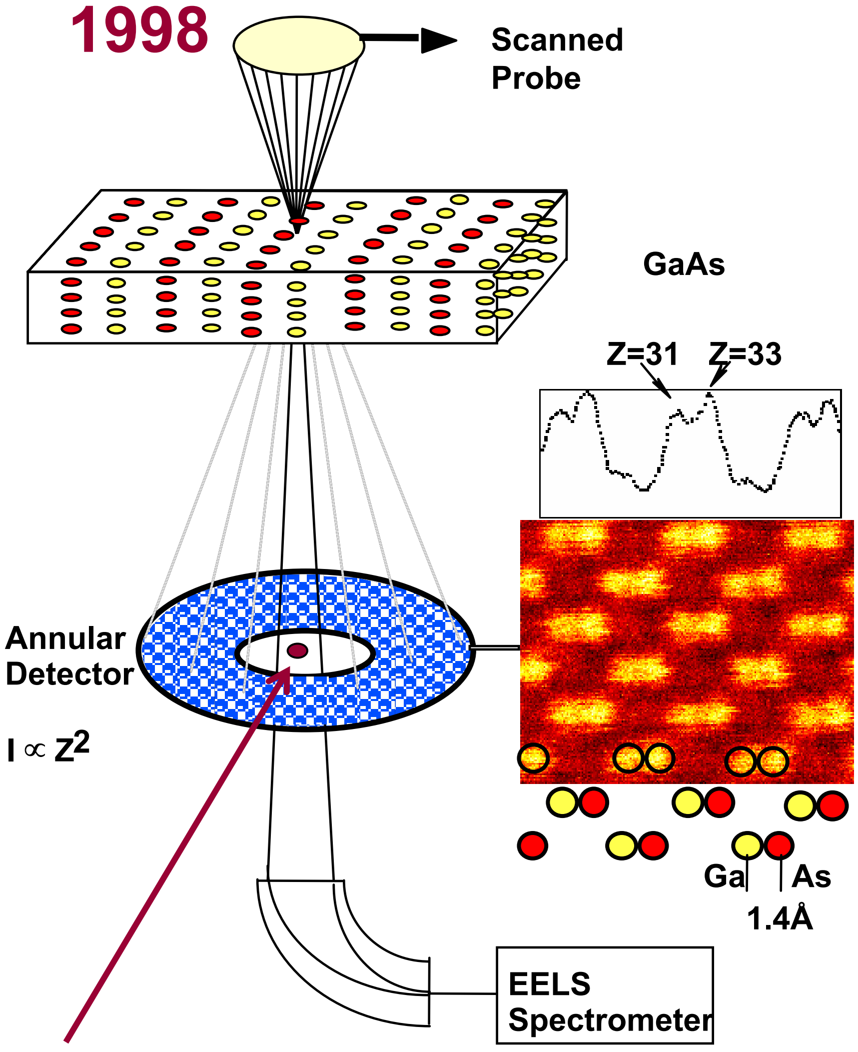
STEM ⇒ simultaneous imaging and electronic properties

Aberration correction ⇒ single atom sensitivity

TEM or STEM?

1988



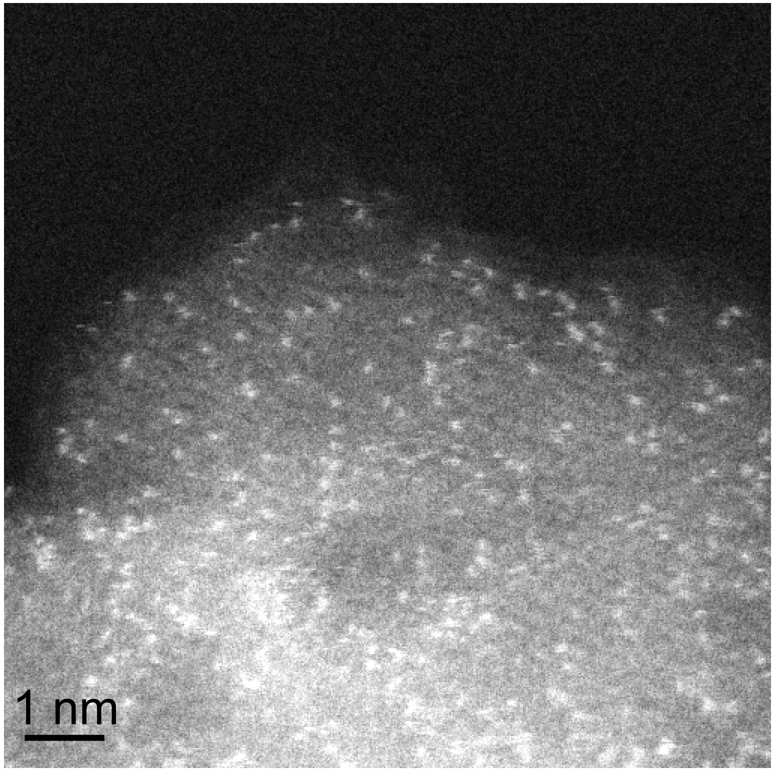


Phase contrast detector

- **STEM invented by Crewe in 1960's**
- **Incoherent imaging with electrons**
- **Atomic resolution spectroscopy**
- **Now standard on commercial TEMs**

STEM or TEM?

γ - Al_2O_3 + 3% La



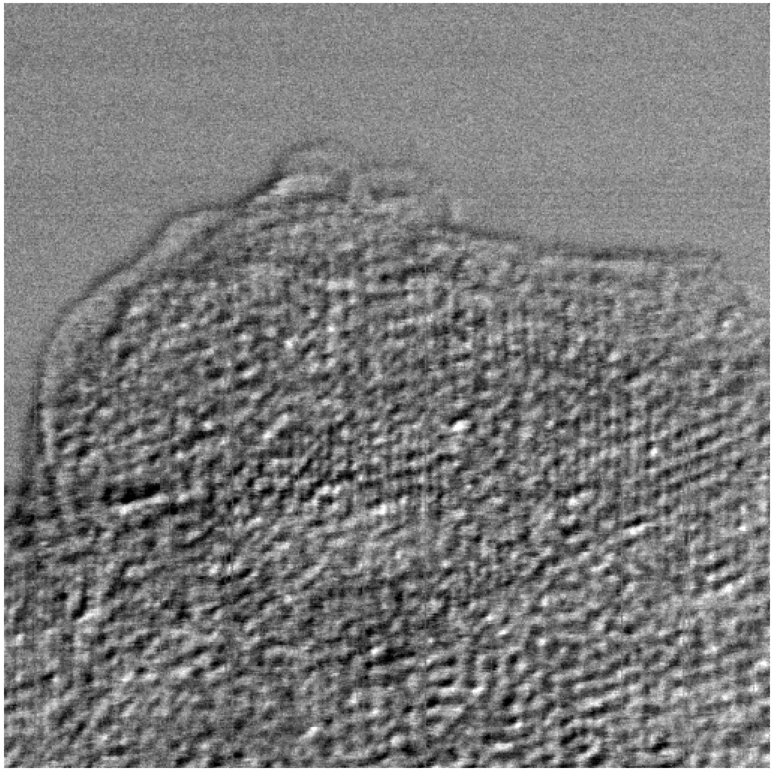
HAADF (Z-contrast)

Resolution = probe size

= 0.61λ/α

Z-contrast

Local



BFSTEM (conventional TEM)

Resolution = λ/α

Phase contrast

Non-local

Probe Size is Limited by Spherical Aberration

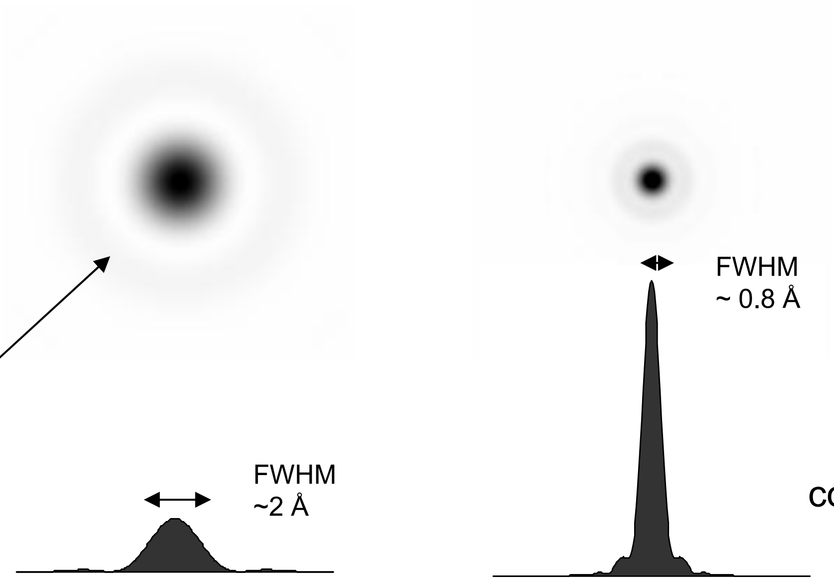
VG Microscope’s HB501UX, 100 kV

Aberration limited

C_s= 1.3 mm

C_c=1.3 mm

Significant current is lost in probe “tails”



No spherical aberration

C_5= 10 mm

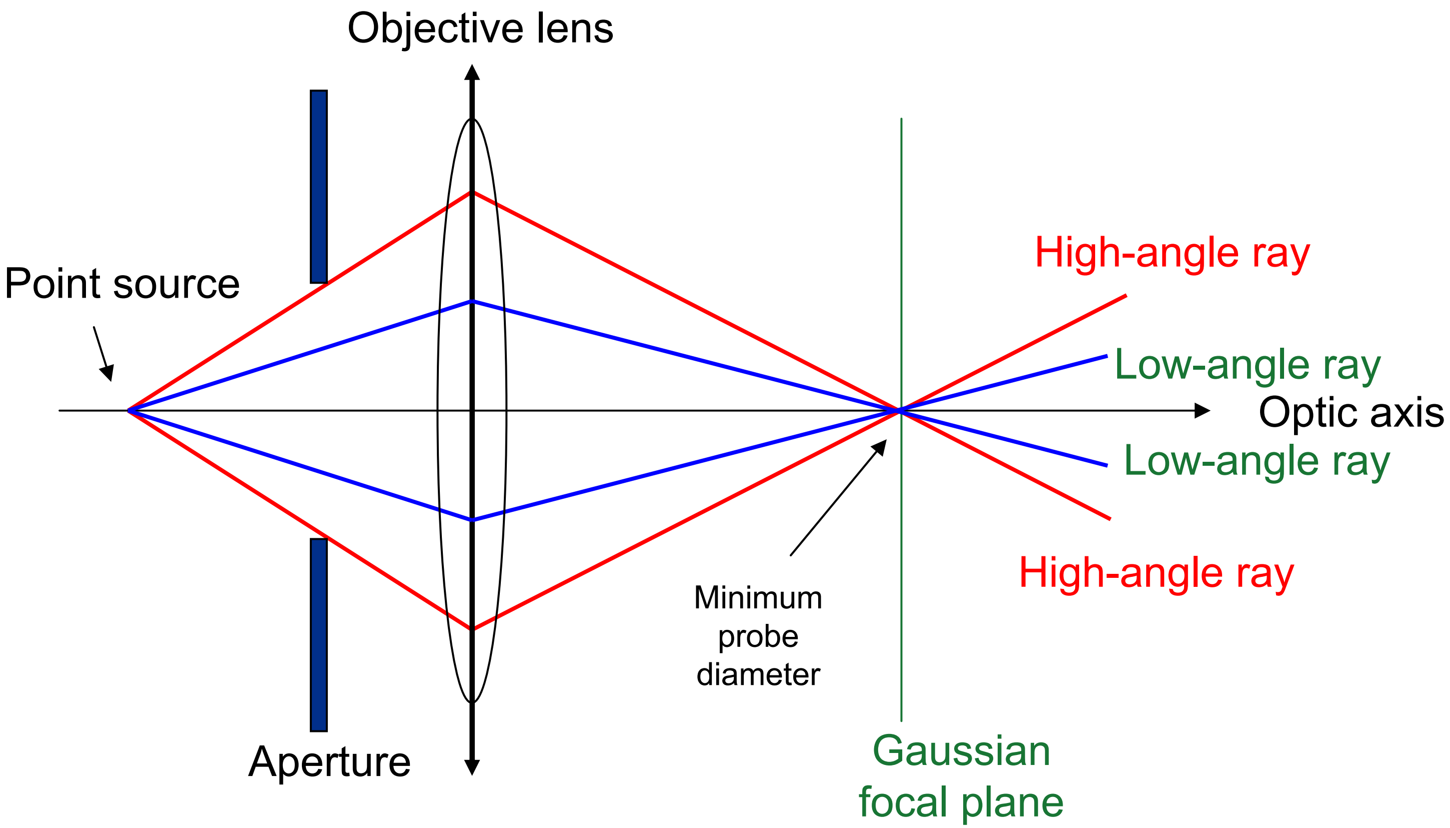
C_c=1.7 mm

Current density is concentrated into central maximum

(Normalized to same total current)

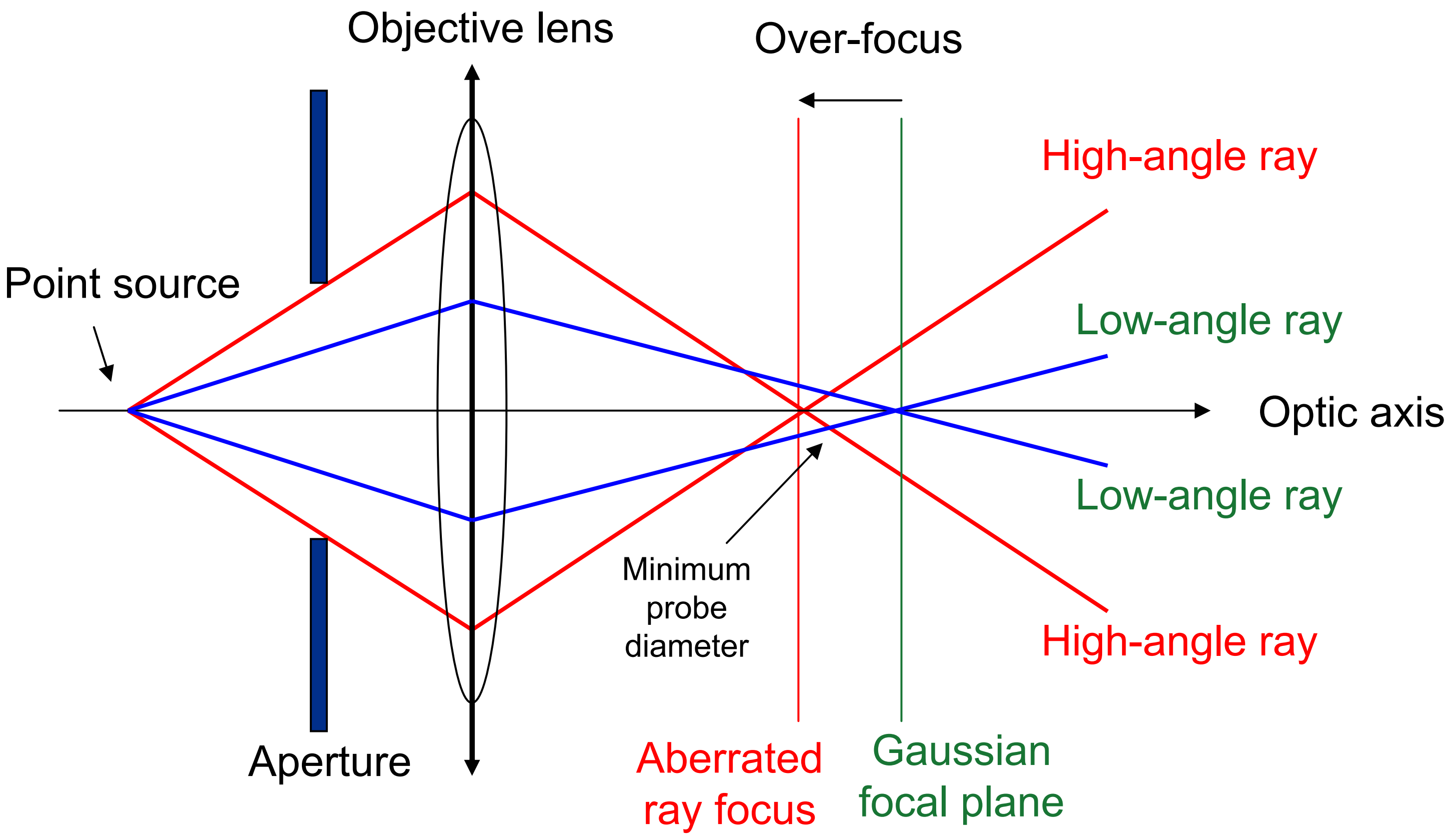
Aberration correction ⇒ smaller brighter probe
Critical for single atom sensitivity

Resolution with a perfect Lens



Ideal lens achieves diffraction limit

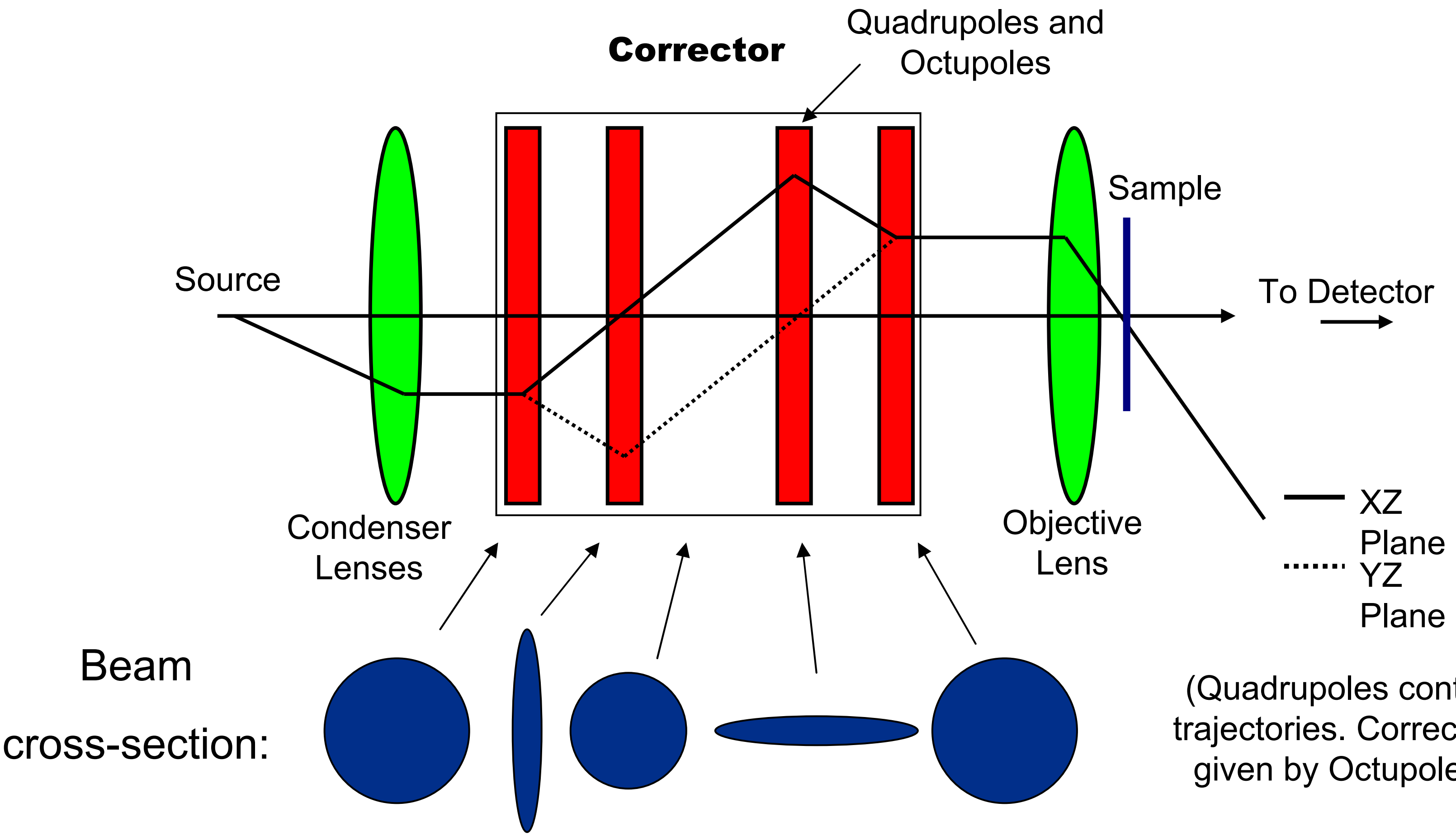
Resolution with a Real Lens



A real lens has spherical aberration

resolution ~ 50 times worse than the diffraction limit

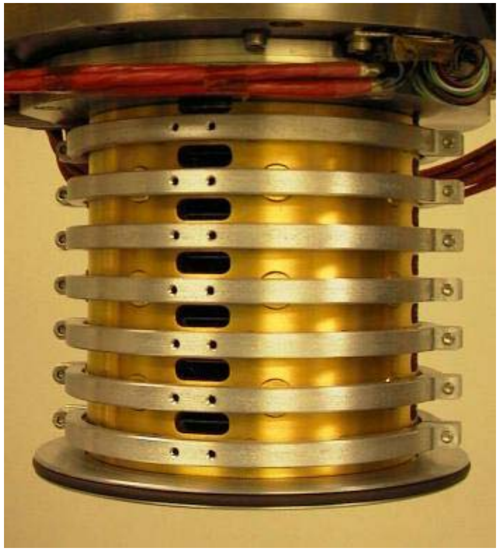
Correction of Spherical Aberration



(Quadrupoles control trajectories. Correction given by Octupoles)

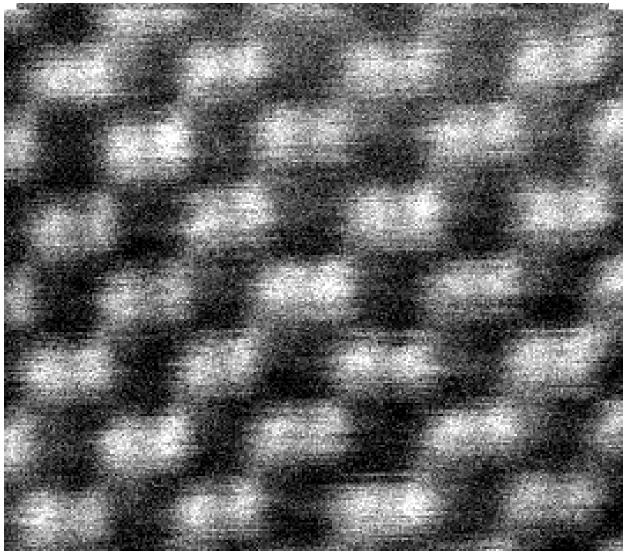
Before Correction

After Correction

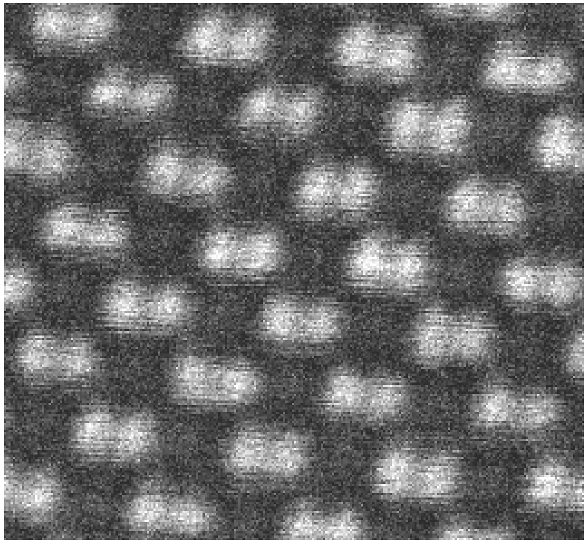


Uncorrected 100kV
STEM: 2.2 Å

Corrected 100 kV
STEM: 1.3 Å

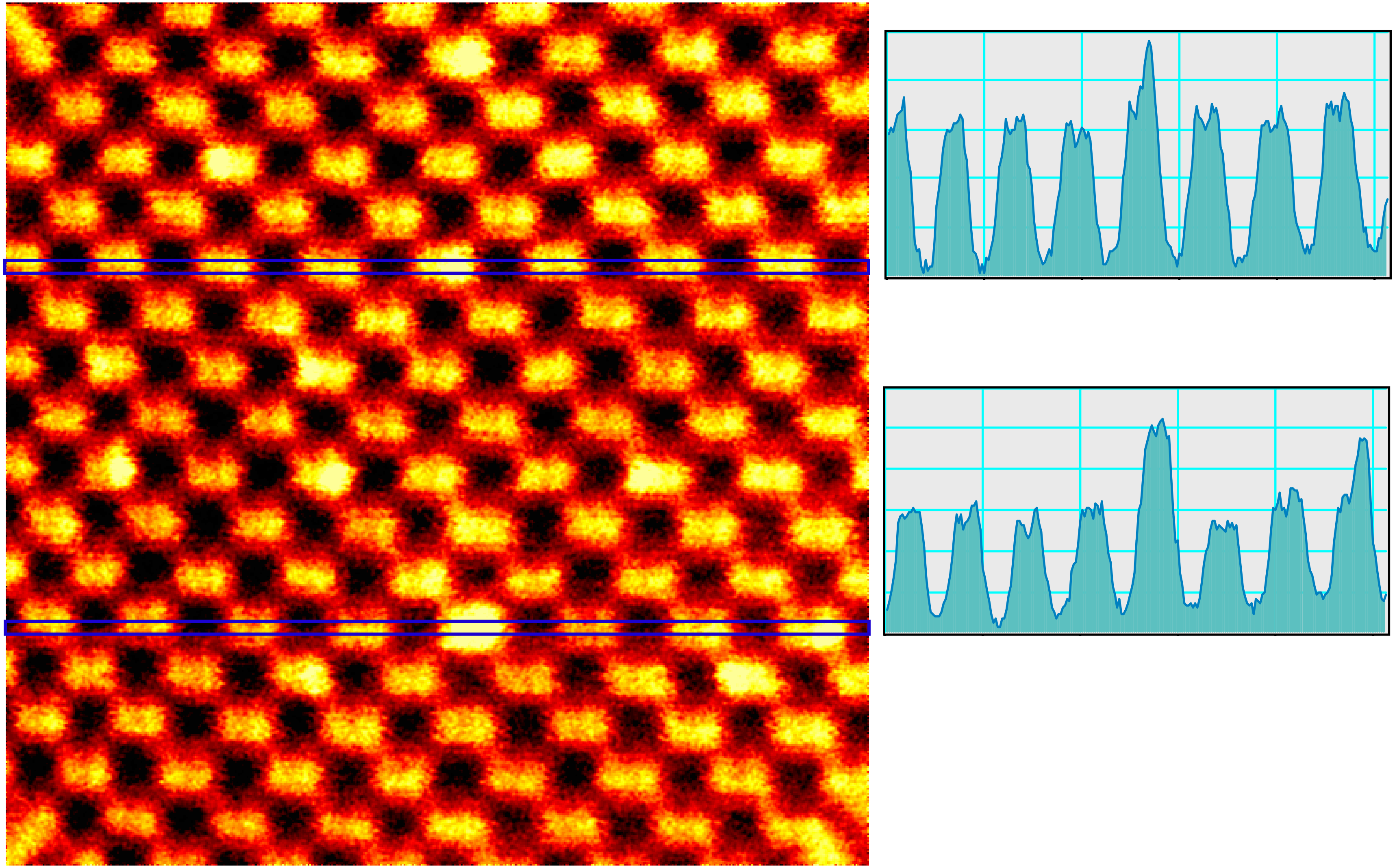


Uncorrected 300kV
STEM: 1.3 Å

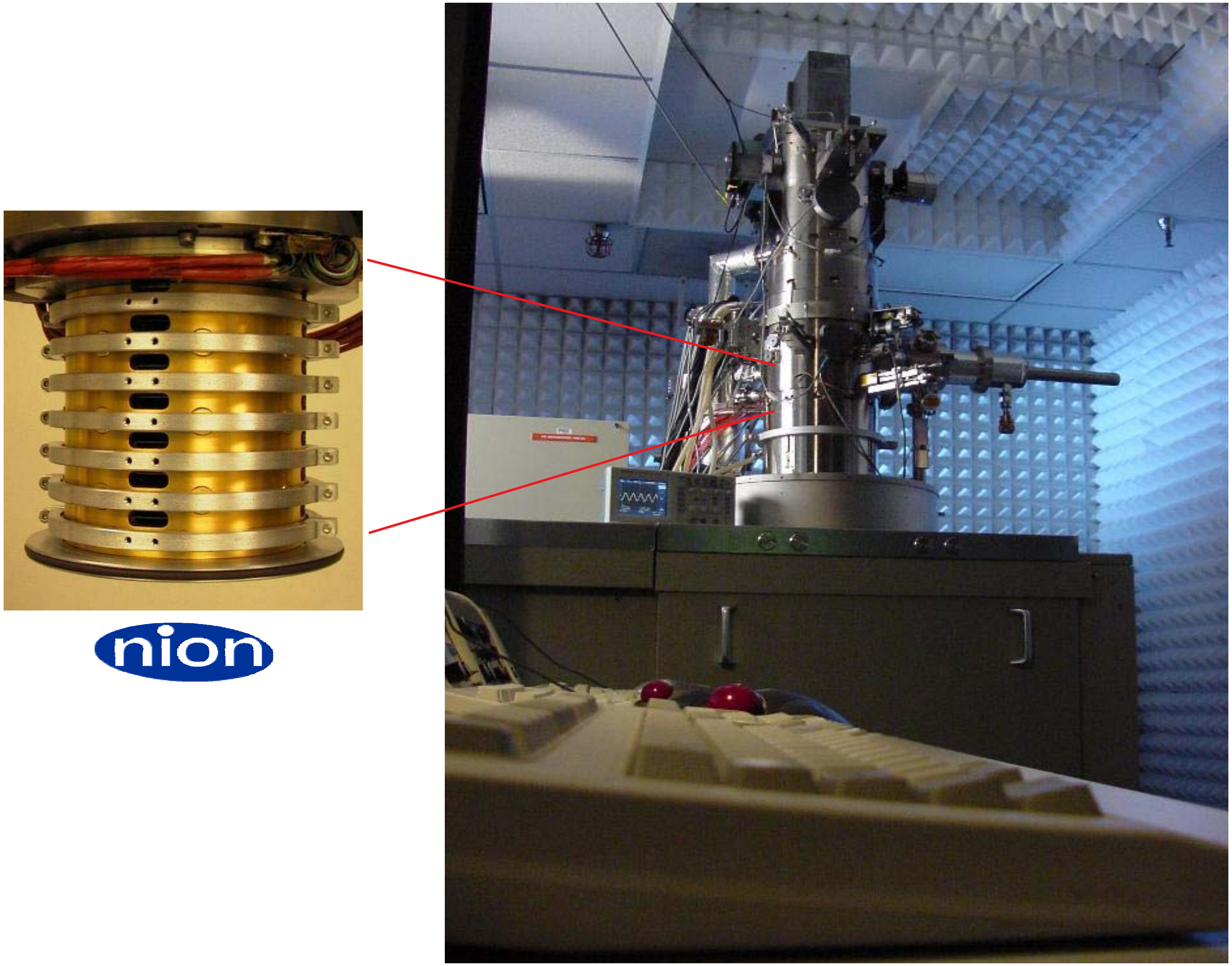


100 kV microscope now rivals 300 kV performance

Imaging of Single Bi Atoms in Si(110)



300 kV STEM

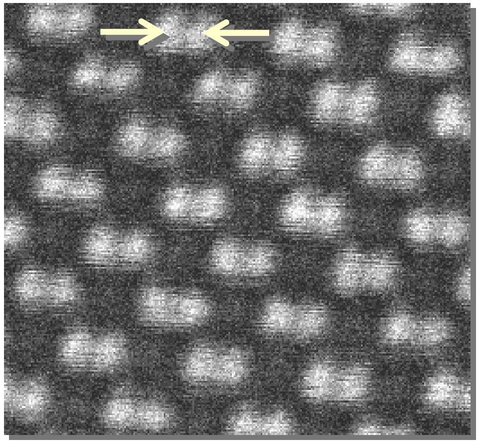


nion

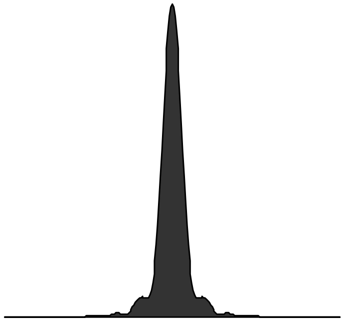
300 kV STEM

Before correction:
1.3 Å theoretical

1.3 Å achieved in Si ⟨110⟩

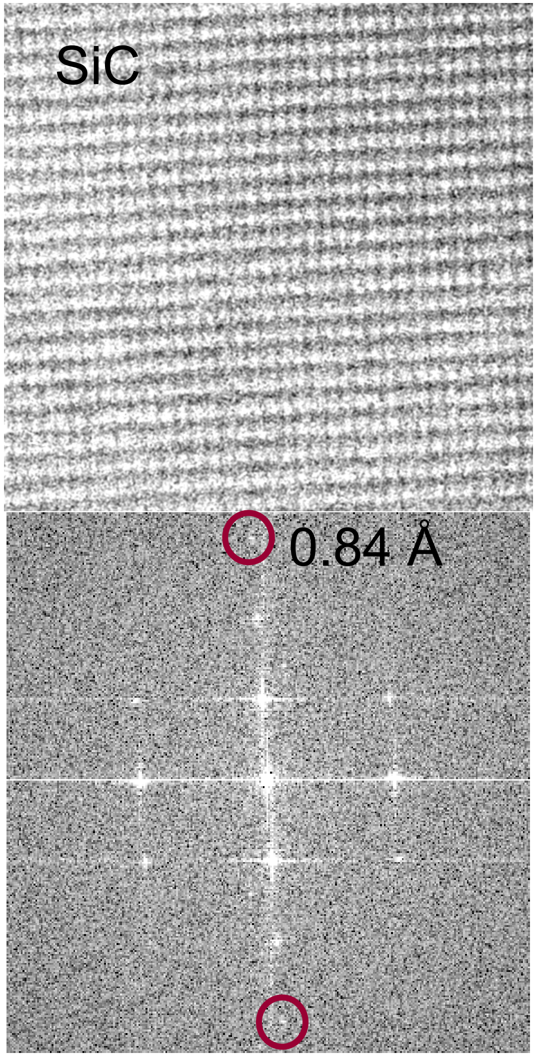


After correction:
0.5 Å theoretical



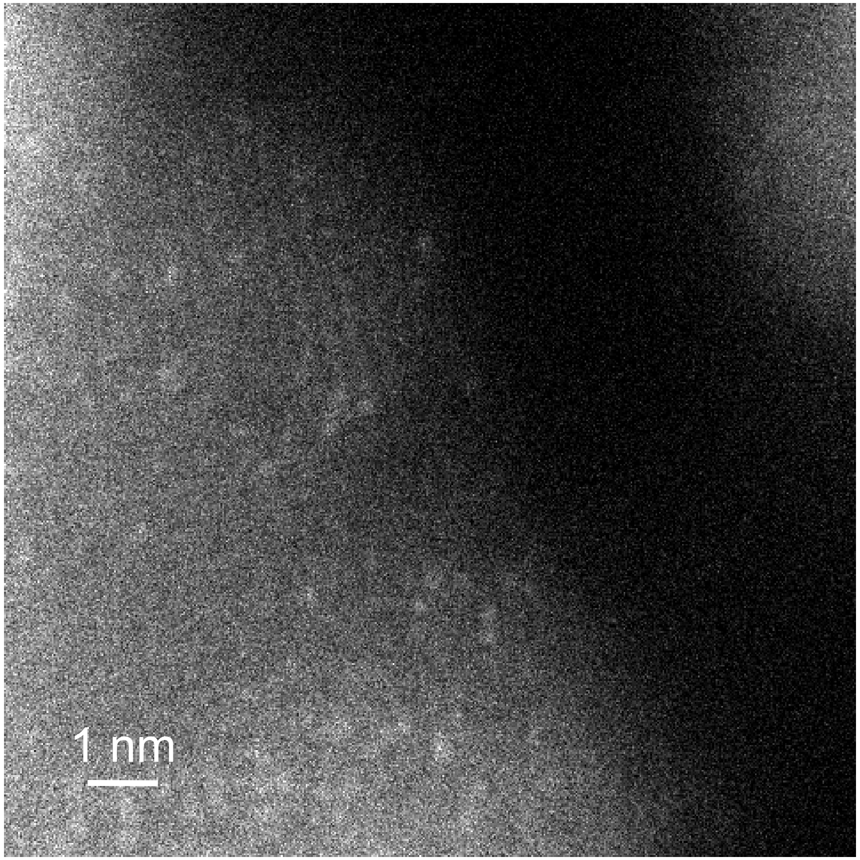
Squeeze the same current into a smaller, brighter probe

Image limited to 0.84 Å by instabilities

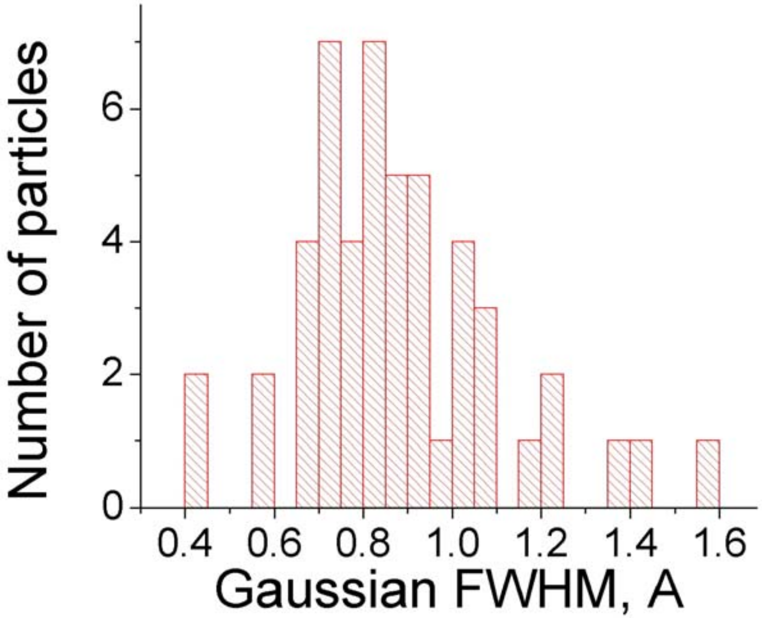


La-stabilized γ-alumina

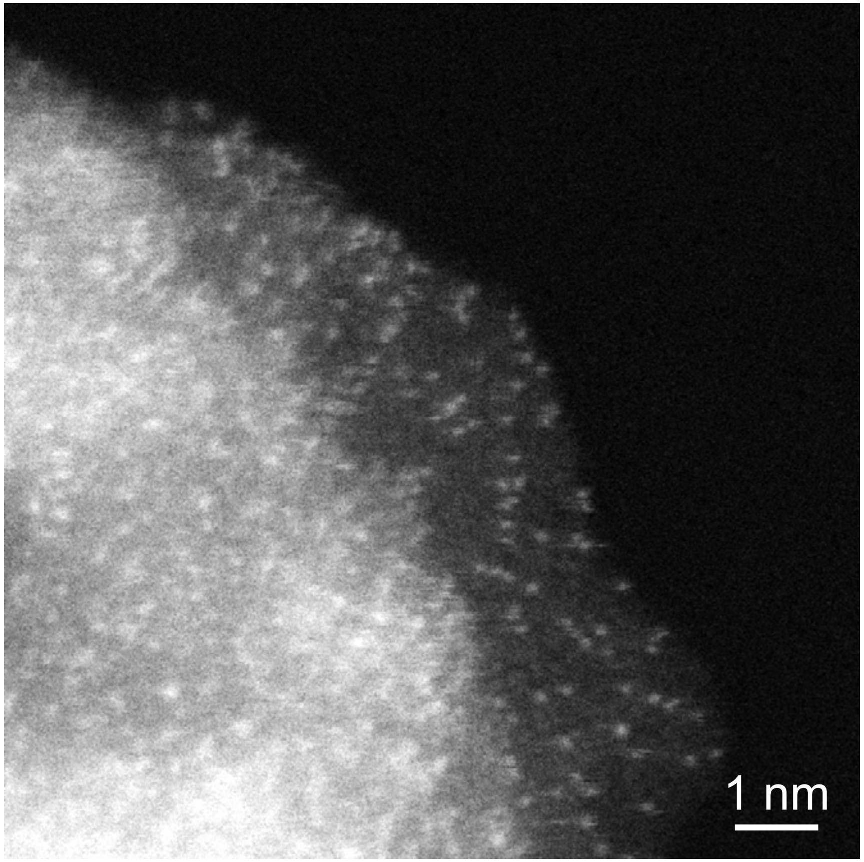
Before correction



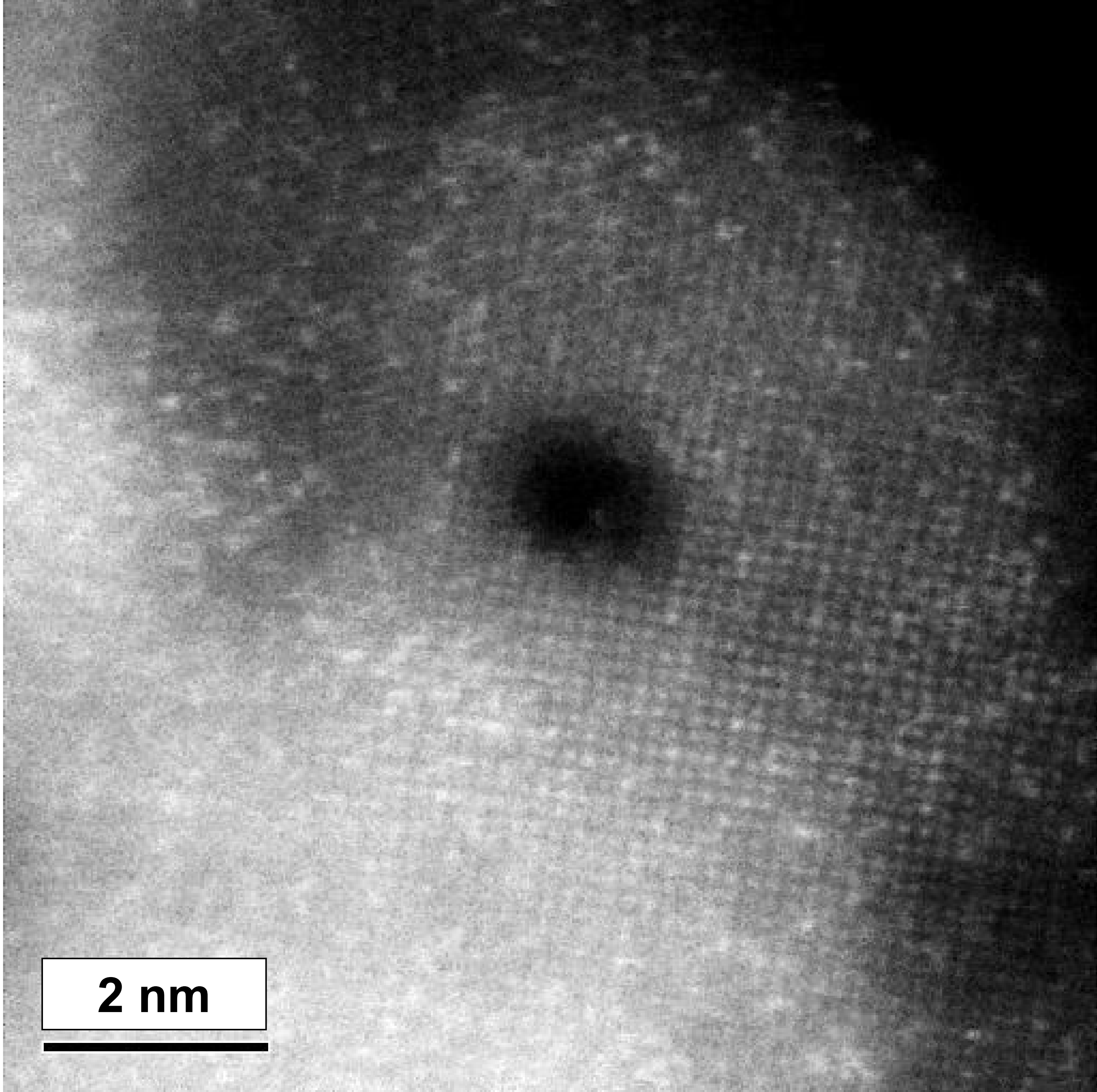
La-stabilized γ-alumina



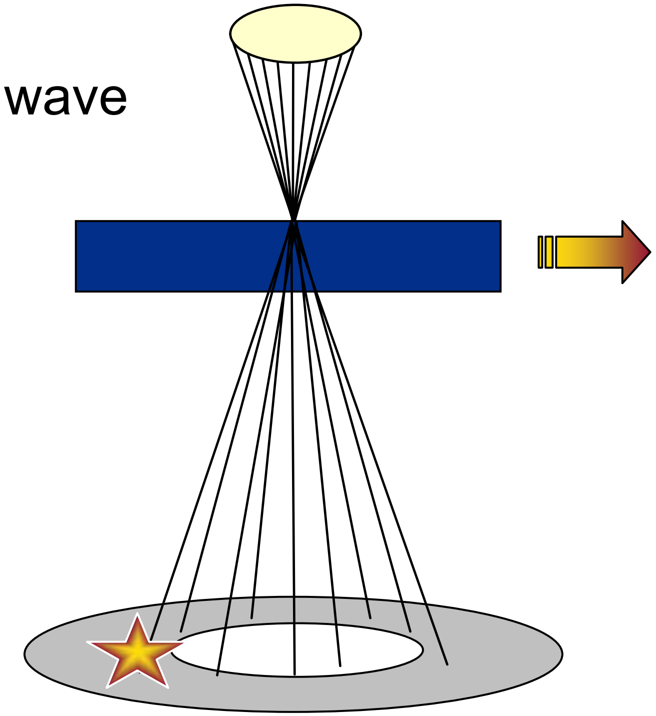
After correction



La atoms located on Al sites



Quantum aspects of STEM – Schrödinger’s cat microscope :



Electron prepared as converging spherical wave

Dynamical scattering through specimen

Propagation to detector

1m

10^8 m/s

10 ns

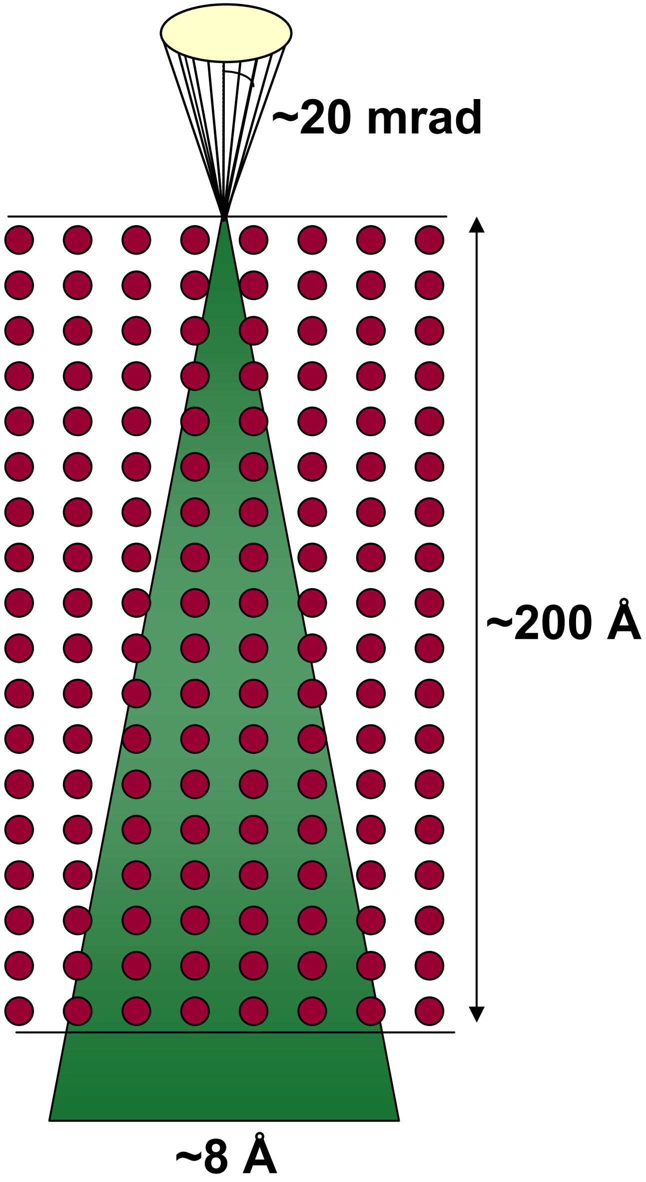
Collapse of wave function

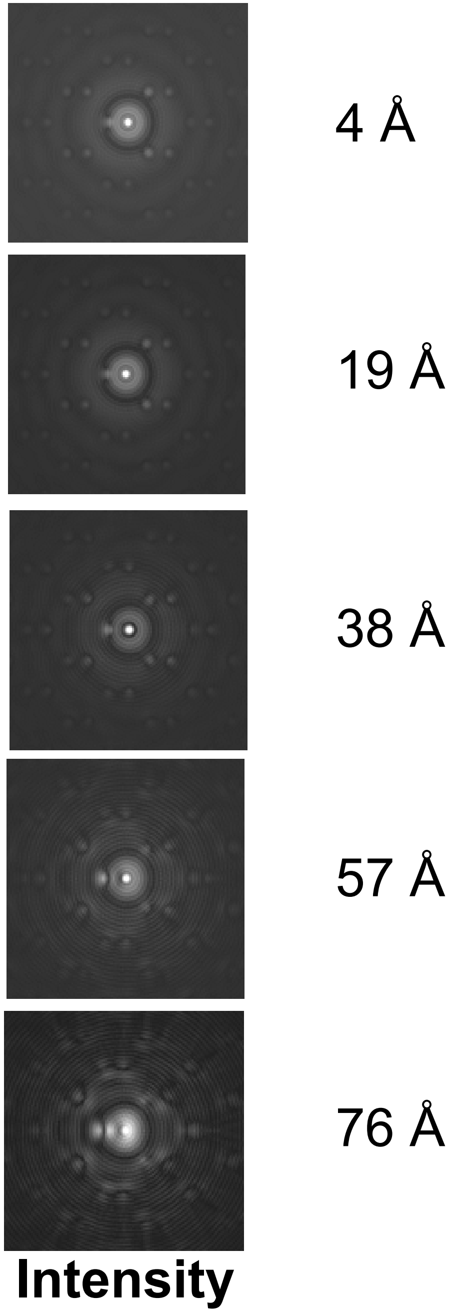
Sample recoil

Detector must be included in the observation

Observable and non-observable in the STEM

How do we get atomic resolution imaging and spectroscopy?



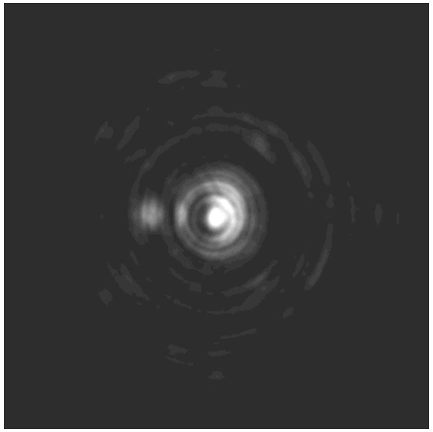


Observable and non-observable in the STEM

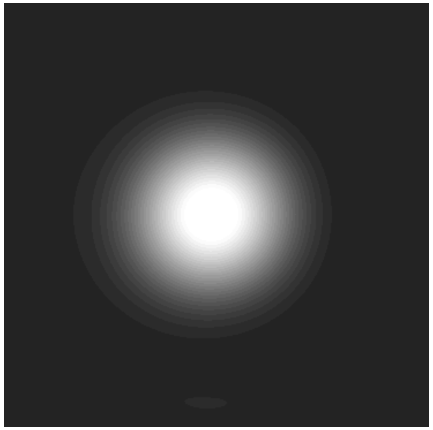
- **Electron intensity distribution inside the crystal is not an observable!**
- **We can weight the electron intensity distribution by the detector function**

- **The amount of localization in a STEM image depends on the detector used (the “observer” in QM)**

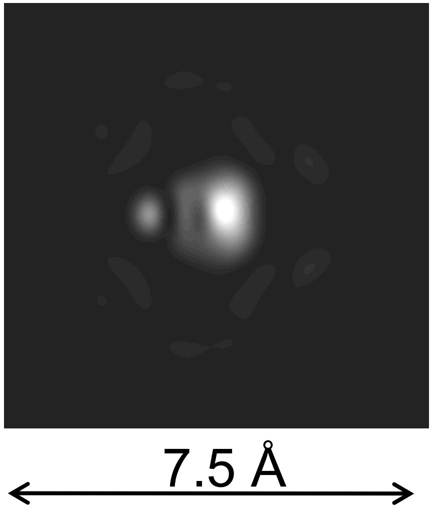
Exit wavefunction intensity



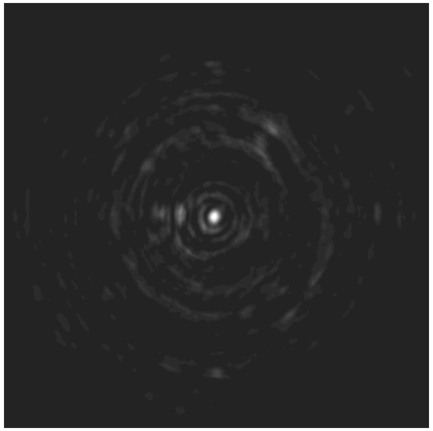
Low Angle Bright Field: Phase contrast TEM



Large Angle Bright Field: EELS



High Angle Dark Field: Z-contrast



1s Bloch States

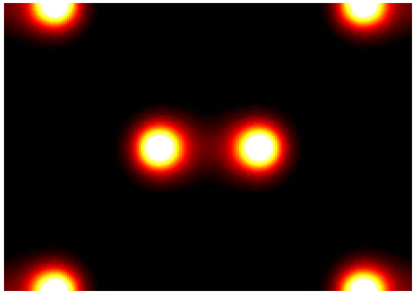
Most localized Bloch states in real space

Most extended in reciprocal space

Dominate detector integration

Most local image

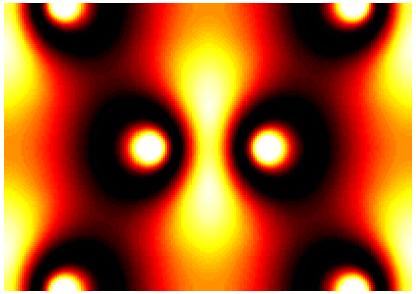
1s



2s state most highly excited

Little contribution to image

2s

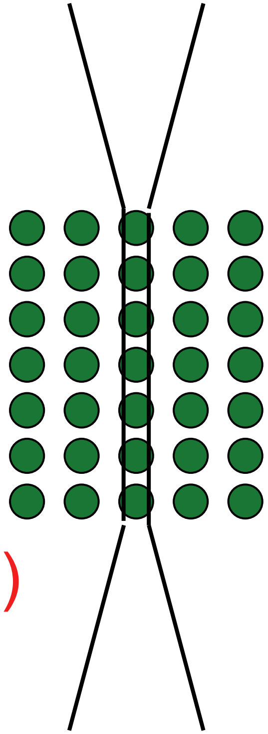


THE Z-CONTRAST IMAGE IS A DIRECT IMAGE OF 1S BLOCH STATES

Pennycook, Nellist and Rafferty, Microscopy & Microanalysis **6**, 343 (2000)

The image is a convolution

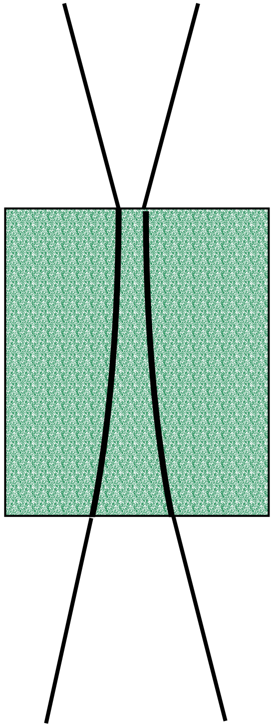
$$I(\underline{\mathbf{R}}) = O(\underline{\mathbf{R}}) * P_{eff}^{2}(\underline{\mathbf{R}})$$



Aligned crystal

$$O(\underline{\mathbf{R}}) = b^{1s^2}(\underline{\mathbf{R}})$$

$$\approx Z^2$$



Amorphous or non-aligned crystal

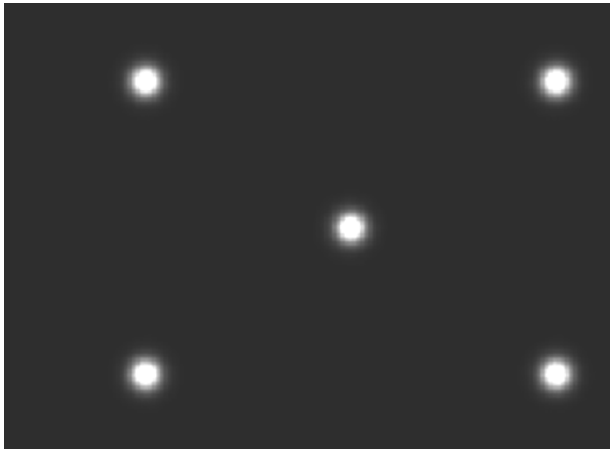
$$O(\underline{\mathbf{R}}) = n\,Z^2(\underline{\mathbf{R}})$$

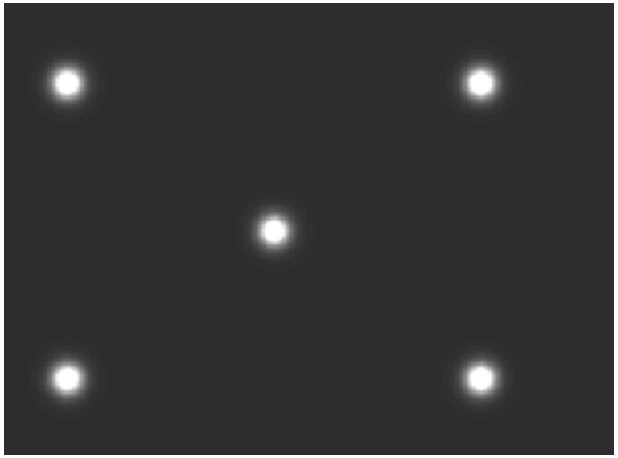
$P_{eff} = P_{incident}$ for thin specimen broadens with thickness

Does the 1s state dominance hold with larger probe angles?

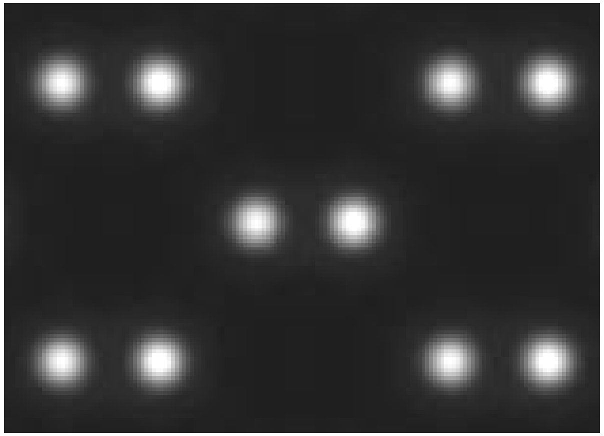
Objective aperture 23 mrad, no aberrations.

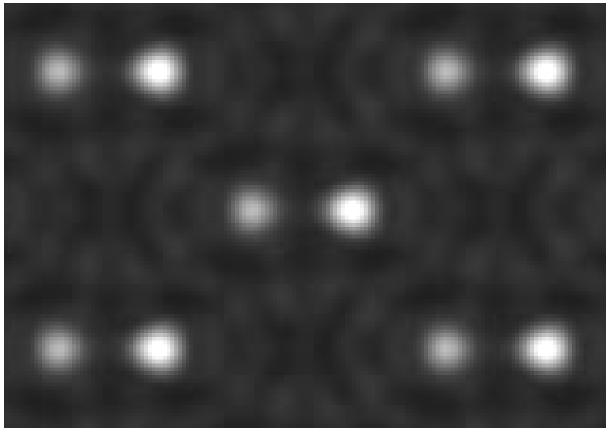
Probe size: 0.53 Å


As 1s

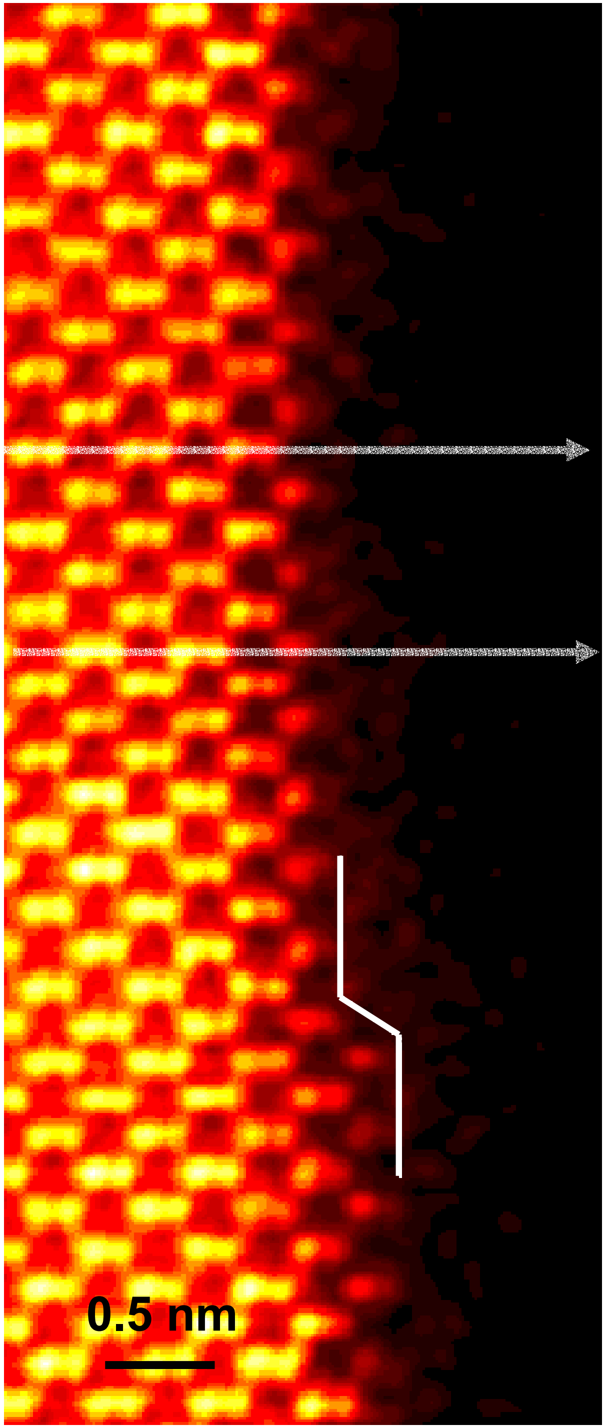

Ga 1s

YES

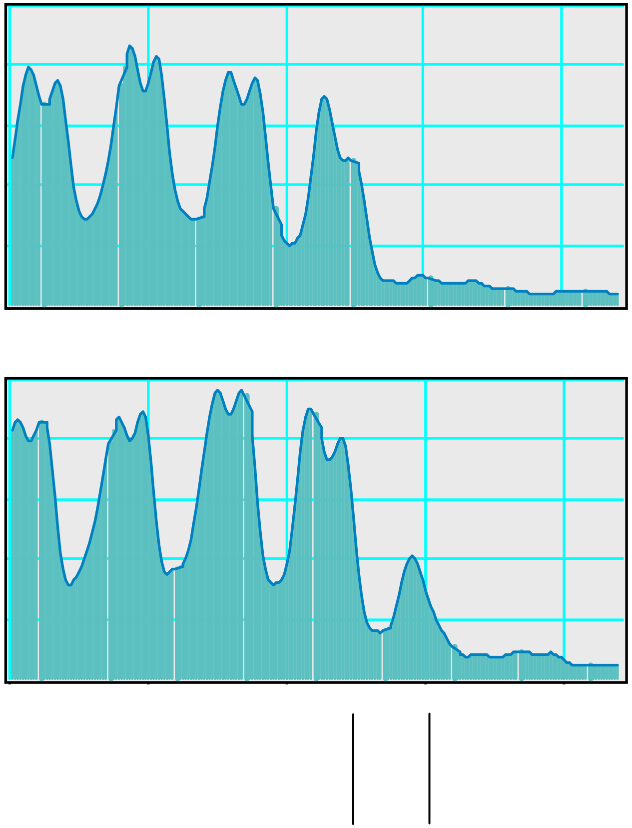

Image, t = 10 Å


Image, t = 1000 Å

Bonding at the Si/SiO$_2$ Interface



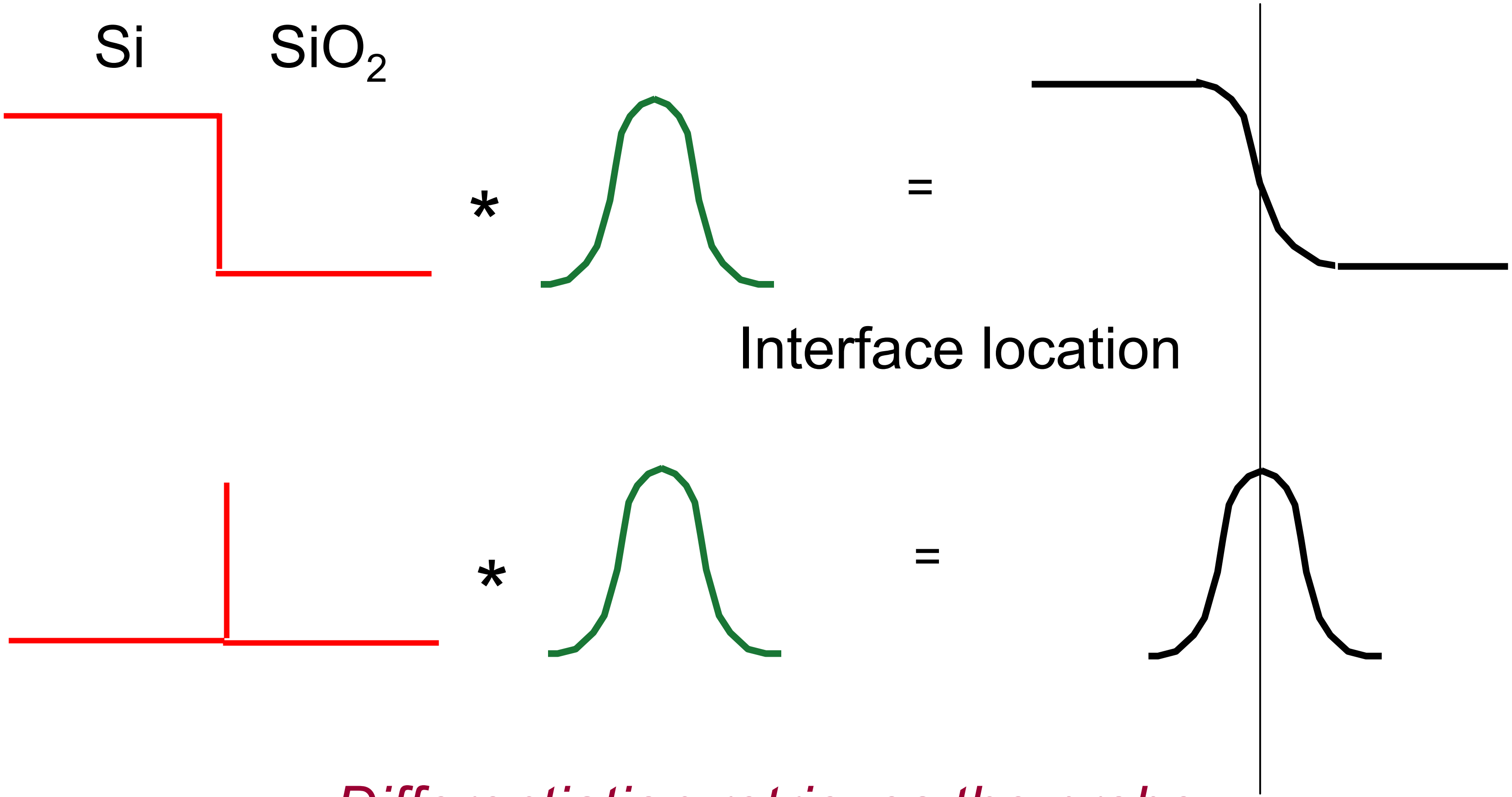
Z-contrast image with Pixon™ reconstruction



Interface width 3 - 5 Å

Imaging an *abrupt* interface:

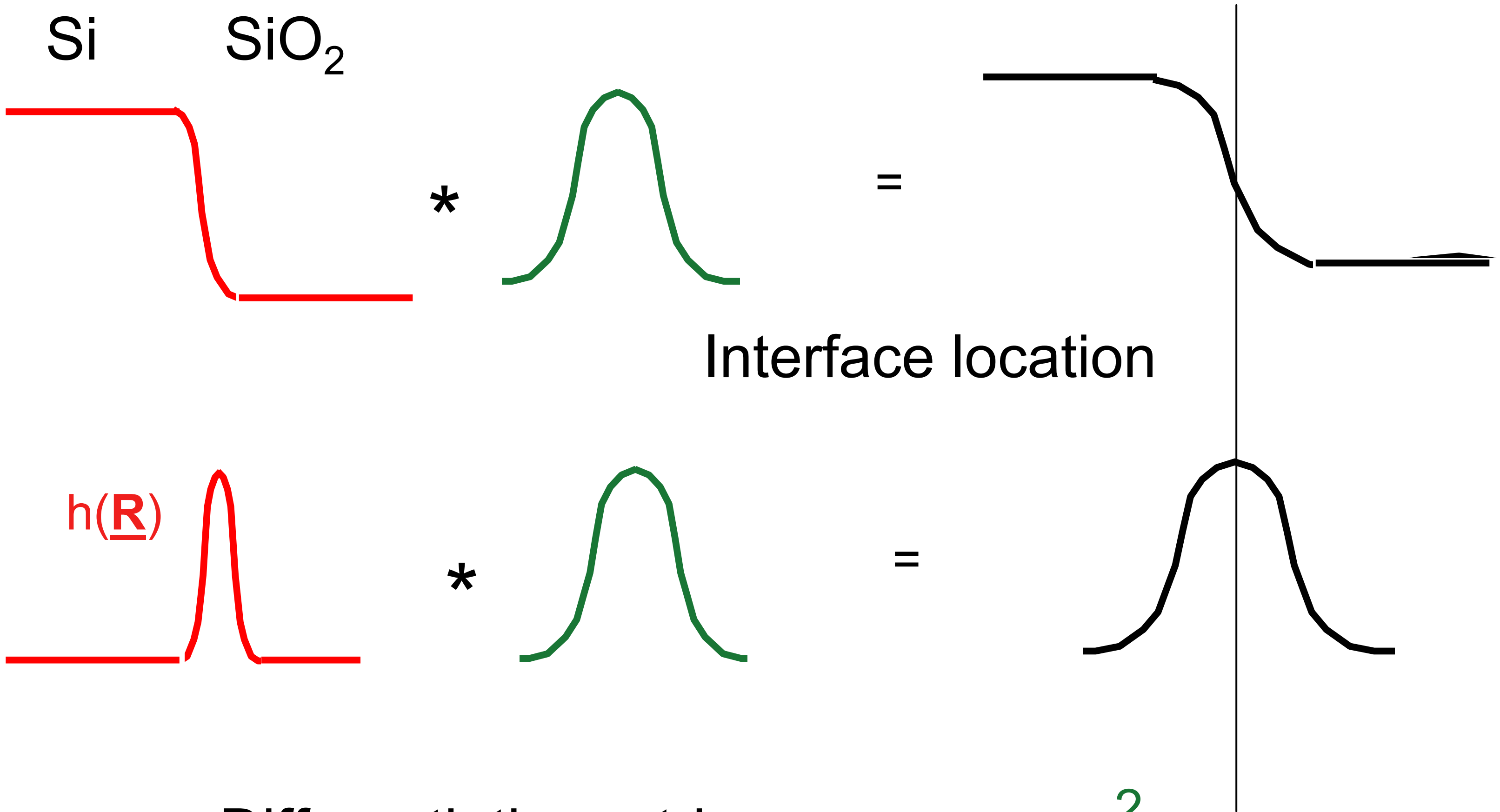
$\square\,(\underline{\mathbf{R}}) \quad * \quad P_{eff}^{2}(\underline{\mathbf{R}}) \quad = \quad I(\underline{\mathbf{R}})$



Differentiation retrieves the probe

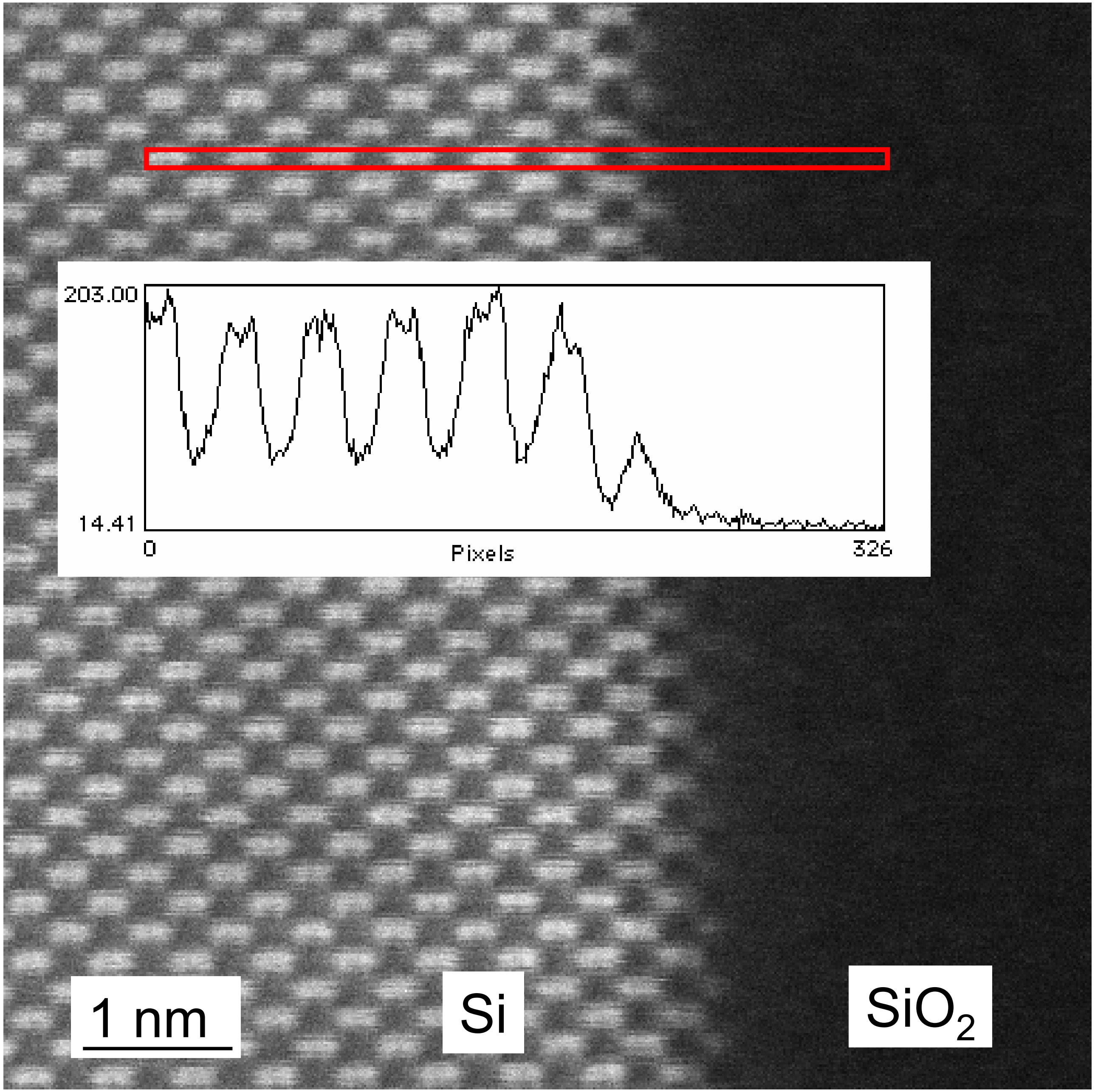
Imaging a *rough* interface:

$$\square(\underline{\mathbf{R}}) * h(\underline{\mathbf{R}}) * P_{\mathrm{eff}}^{2}(\underline{\mathbf{R}}) = I(\underline{\mathbf{R}})$$



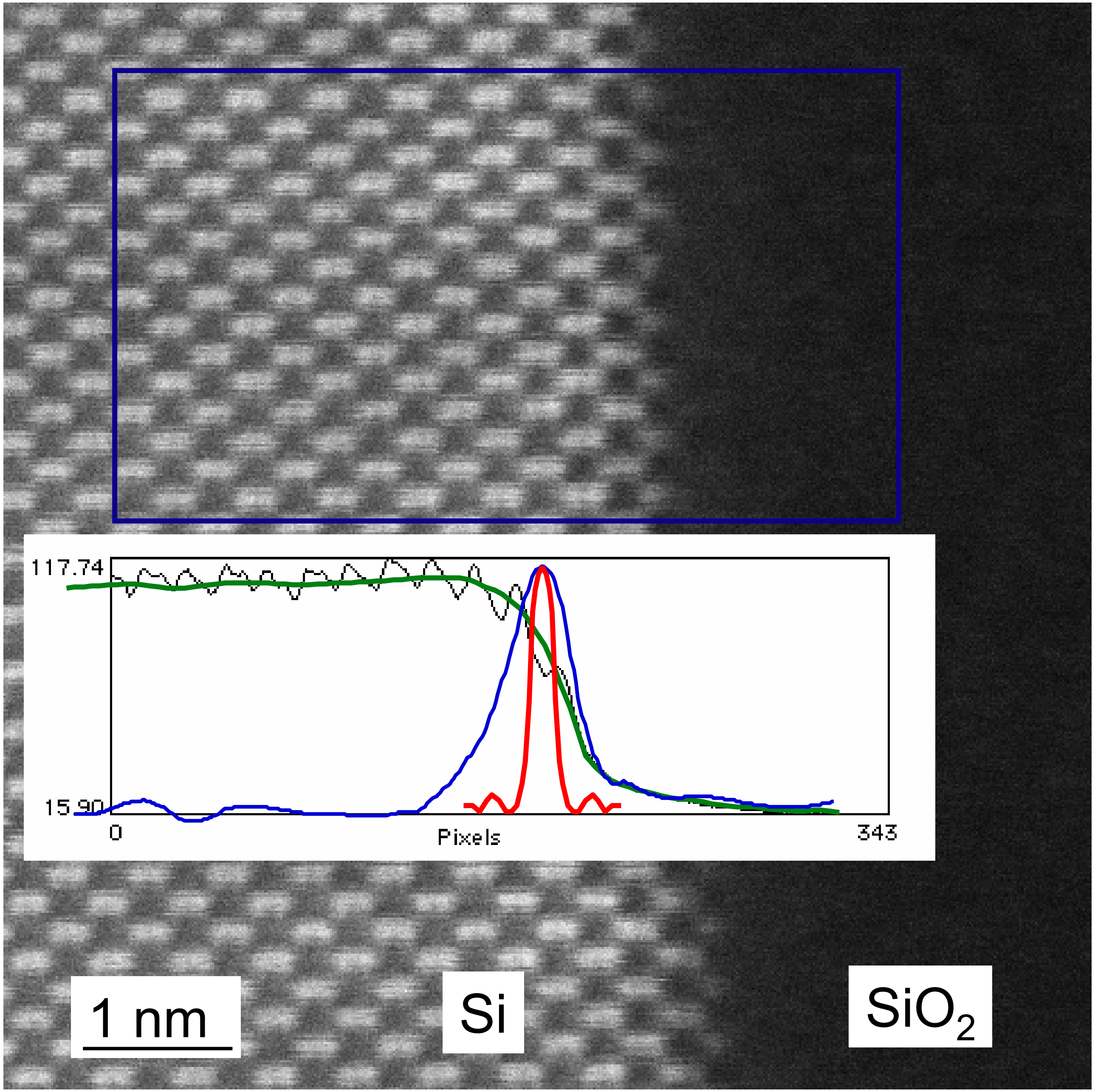
Differentiation retrieves $h(\underline{\mathbf{R}}) * P_{\mathrm{eff}}^{2}(\underline{\mathbf{R}})$

Sharp Si/SiO_2 interface



Line trace summed vertically over 3 pixels

Sharp Si/SiO$_2$ interface



Line trace summed vertically over 200 pixels

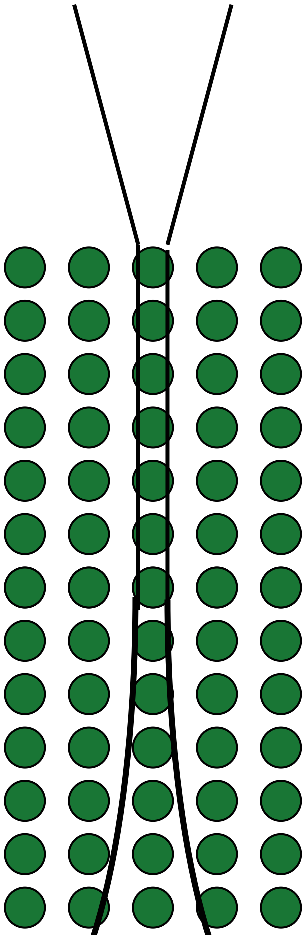
Fit

Derivative FWHM 5.1Å

Probe FWHM 1.3 Å

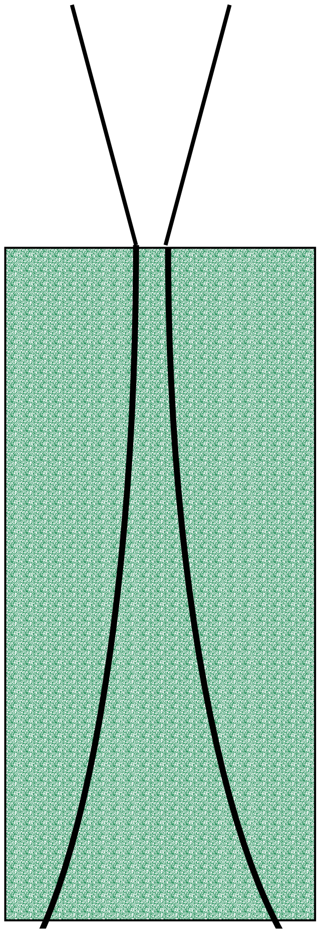
Roughness 0.49 nm

Beam broadening - thick specimen



Aligned crystal

P_{eff}^2 gains a broad background

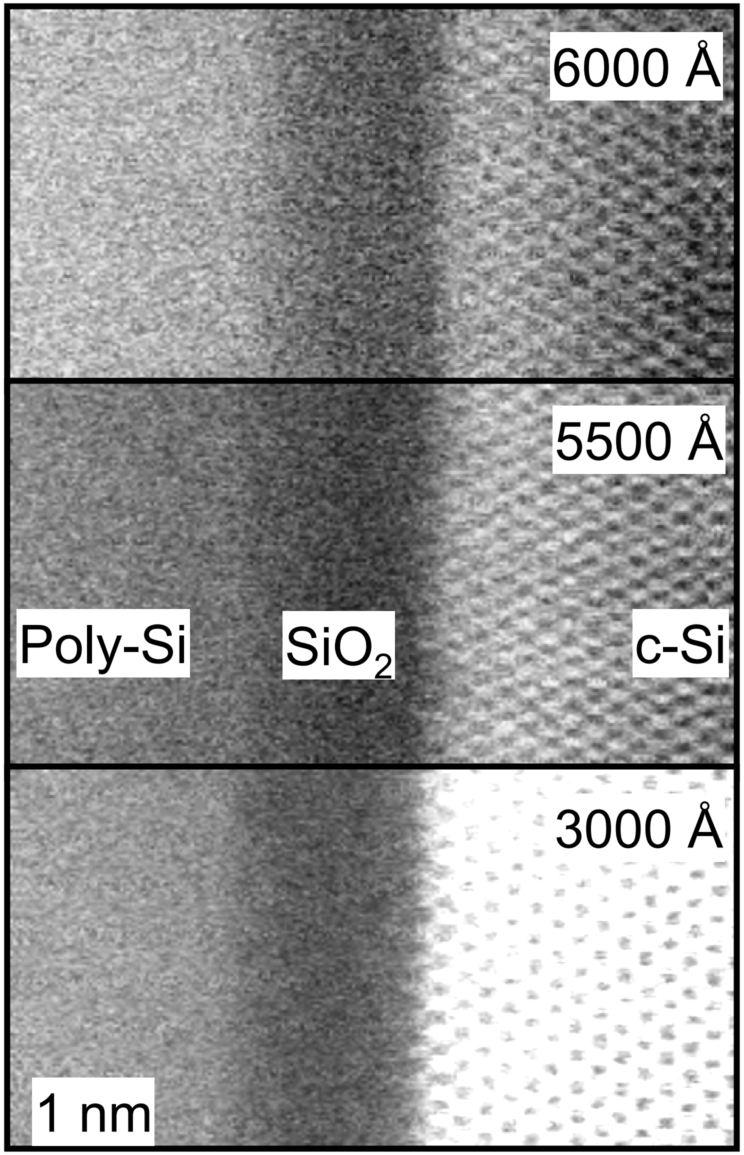


Amorphous or non-aligned crystal

P_{eff}^2 continues to increase with thickness

HA-ADF STEM

- Gate oxide width is independent of thickness
- Interface roughness appears to increase



Gate Oxide Thickness: 20 Å

Muller JEOL 2010F, 200 kV, See Diebold et al, Microscopy and Microanalysis 2003

Localization of Inelastic Scattering

- **Classical estimates based on impact parameter**

S. J. Pennycook, *Ultramicroscopy*, **26** (1988) 239

$$b^{rms} = \frac{2E_o}{X\Delta E} Log\left(\frac{4E_o}{\Delta E}\right)^{-1/2}$$

- **Quantum mechanical calculations** O′(**R**) =

$$\left(\frac{e^2}{\pi\hbar\upsilon}\right)^2 \left| \int \frac{\rho_{no}}{q^2}(\mathbf{q})e^{-i\mathbf{K}\cdot\mathbf{R}}d\mathbf{K} \right|^2$$

Based on the dipole approximation:

H. Rose, *Optik* **45**, (1976) 139

R. H. Ritchie and A. Howie, *Phil Mag A* **58**, (1988) 753

D. Muller and J. Silcox, *Ultramicroscopy* **59**, (1995) 735

Avoiding the dipole approximation:

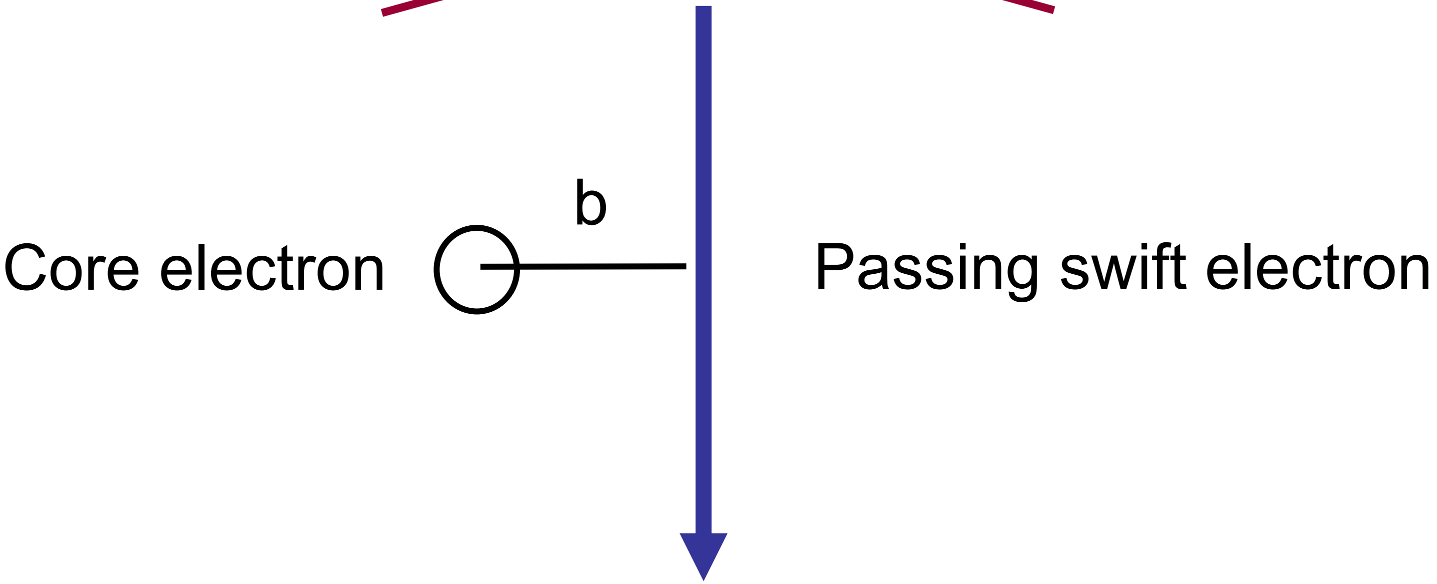
D. W. Essex, P. D. Nellist and C. T. Whelan, Ultramicroscopy, 80 (1999) 183

B. Rafferty and S. J. Pennycook, *Ultramicroscopy*, **78** (1999) 141

L. A. Allen, S. D. Findlay, M. P. Oxley and C. J. Rossouw, Ultramicroscopy (2002) submitted

Classical view - impact parameter

$$\xcancel{b = \hbar v / \Delta E}$$

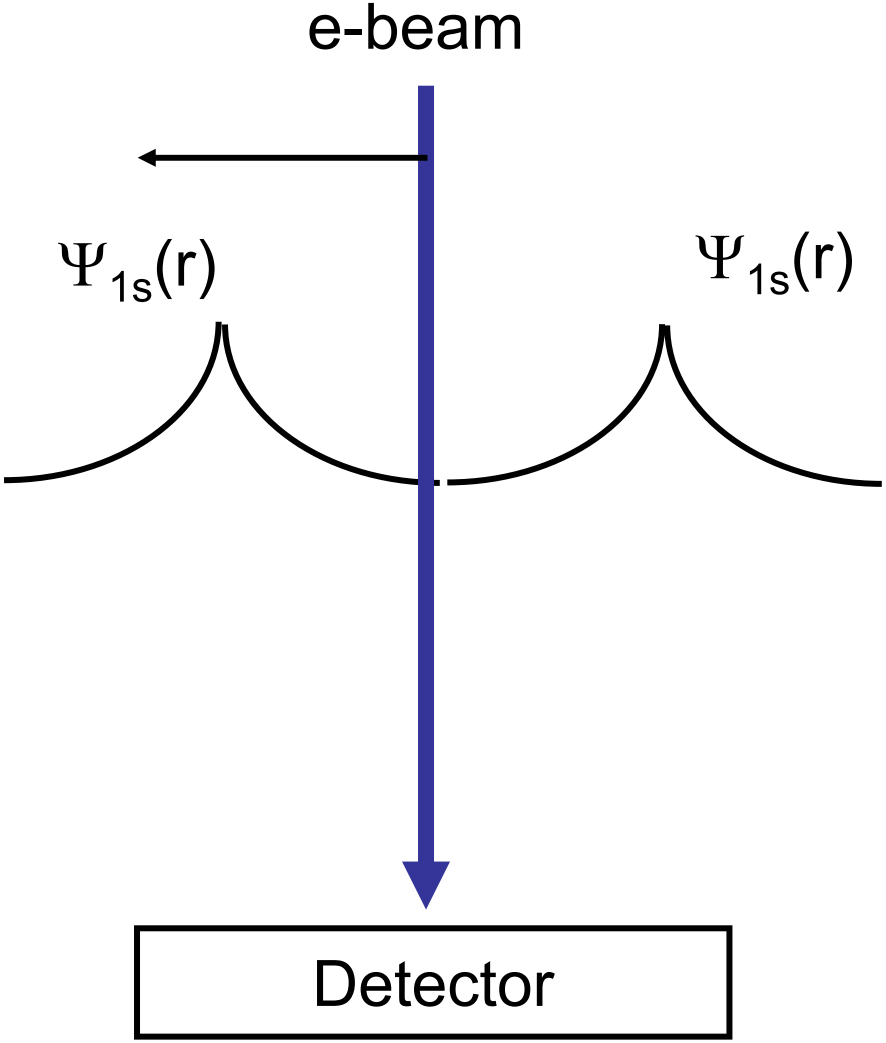


Core electron

Passing swift electron

Where is the observer?

Quantum mechanical view

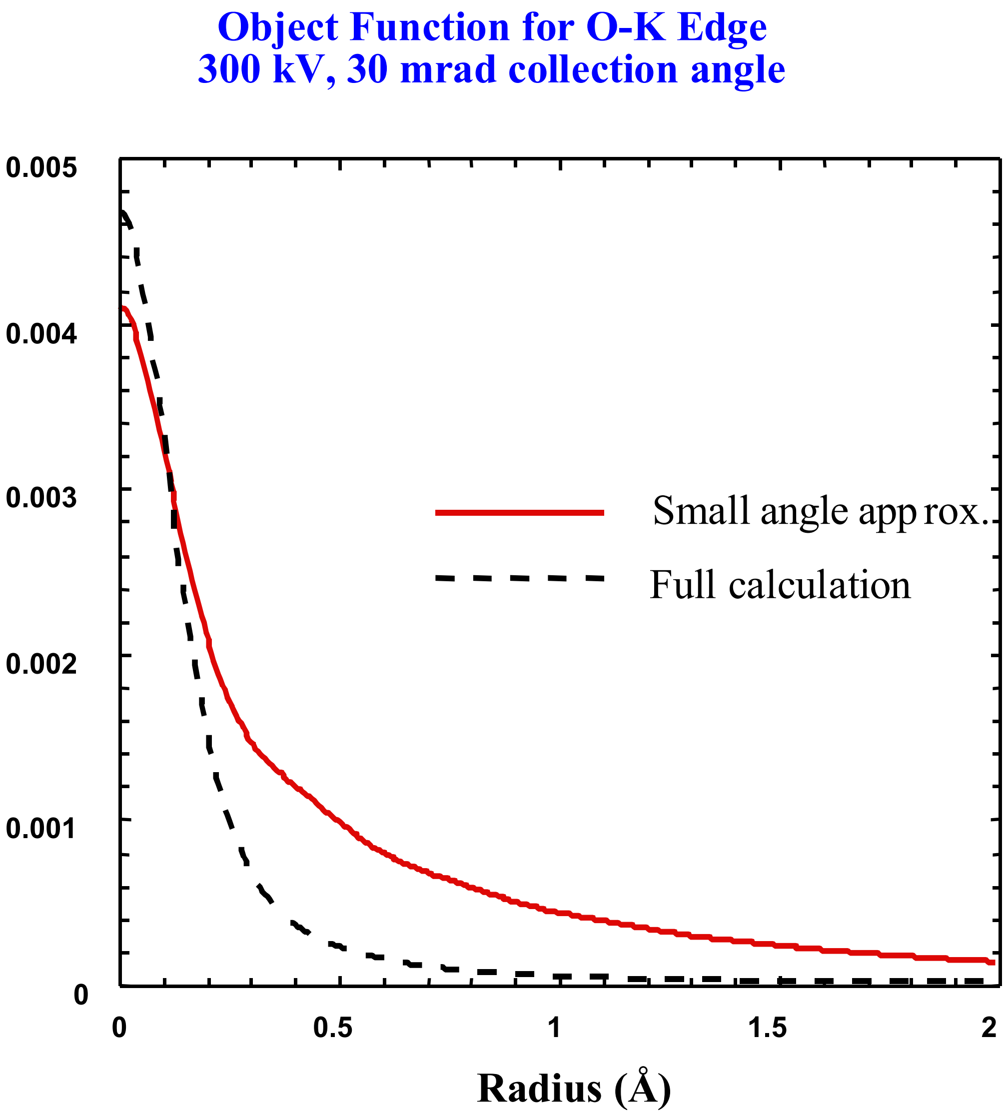


Angular range of the detector is critical

Dipole Approximation

$e^{iK.R} \sim 1 + iK.R$

Fails at large K and large R



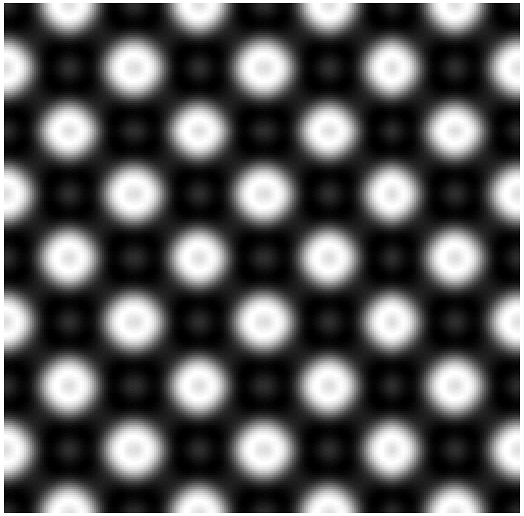
$SrTiO_3$ Ti *L*-shell EELS

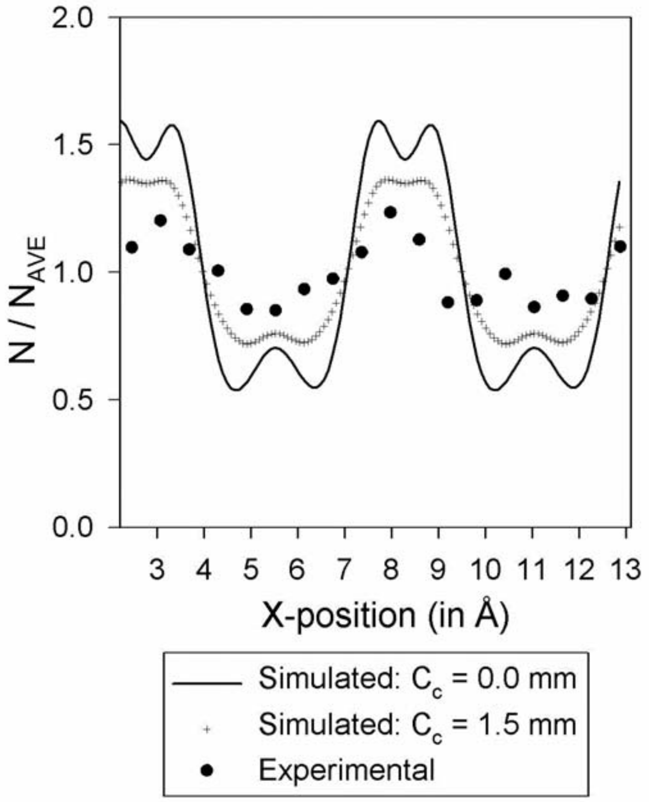
Probe: C_S = – 0.05 mm, Δf = –62 Å, C_5 = 63 mm

E = 100 keV   aperture = 20 mrad

ADF detector
56 – 202 mrad

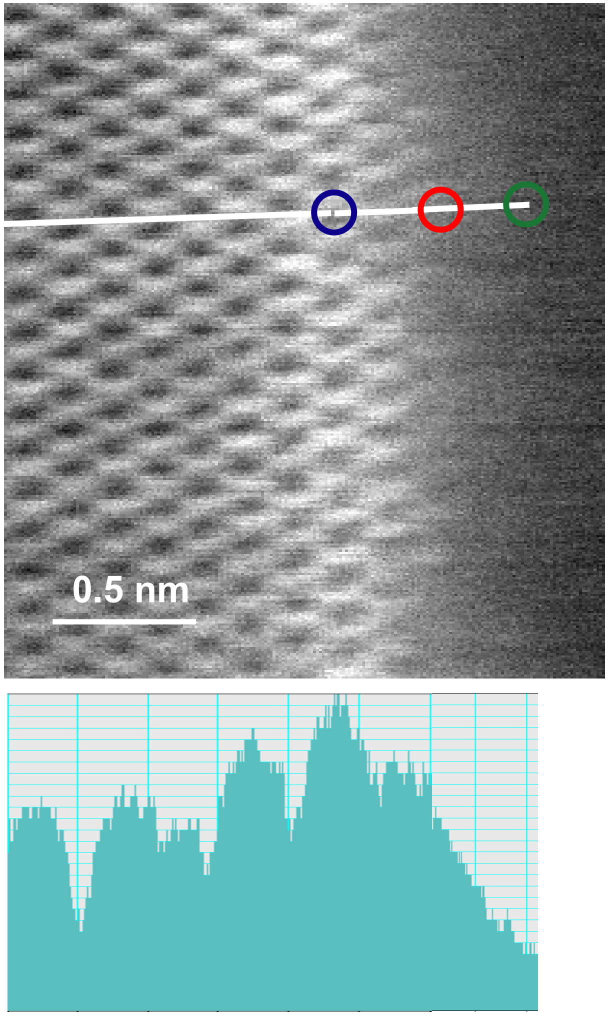
EELS detector
semiangle: 20 mrad

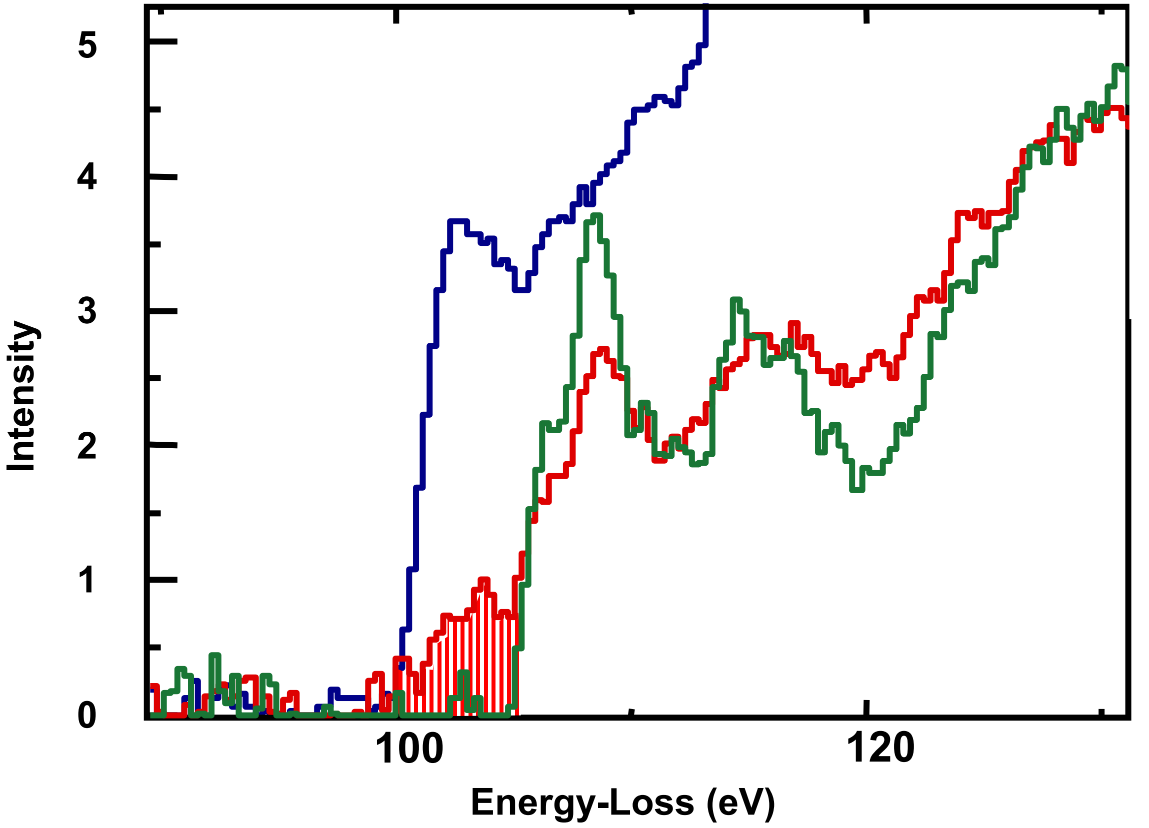




Zone-axis images   t = 200 Å

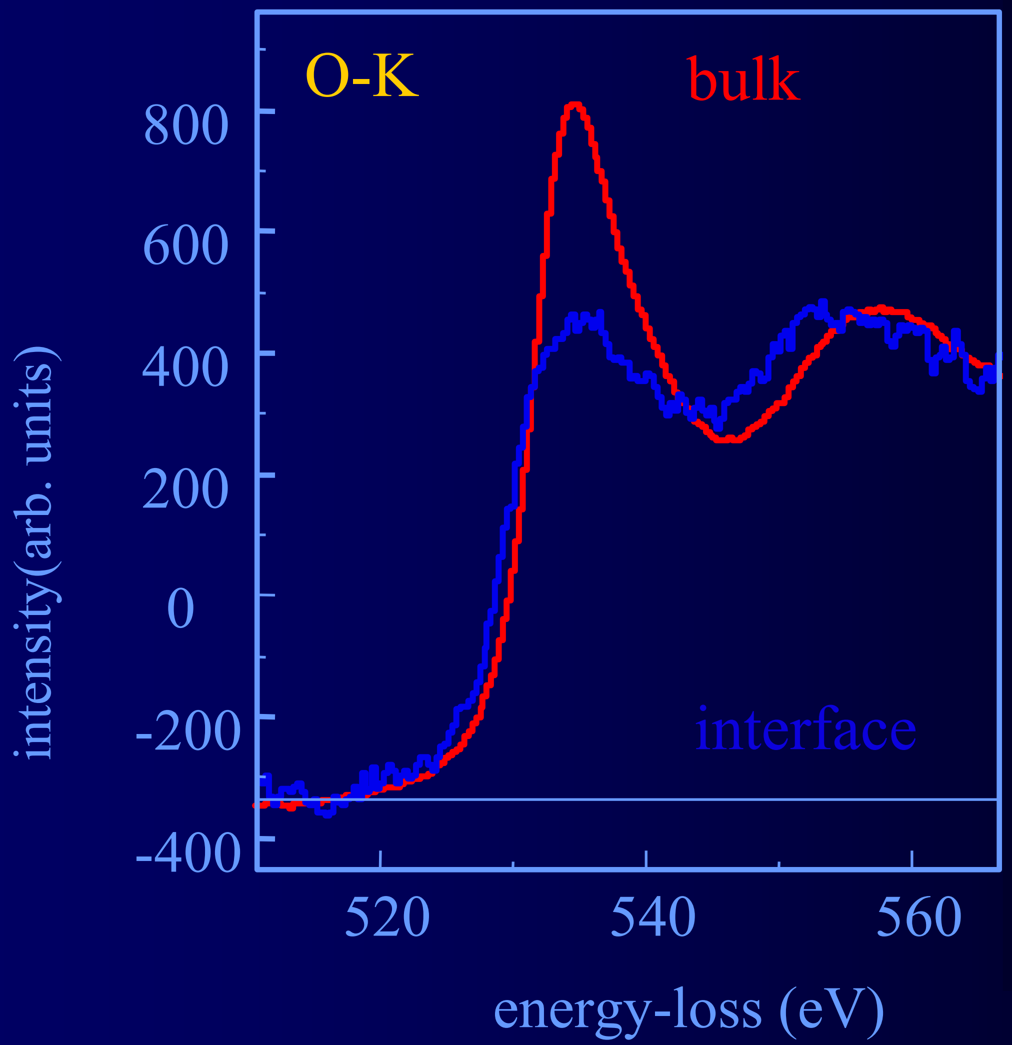
EELS Line Scan





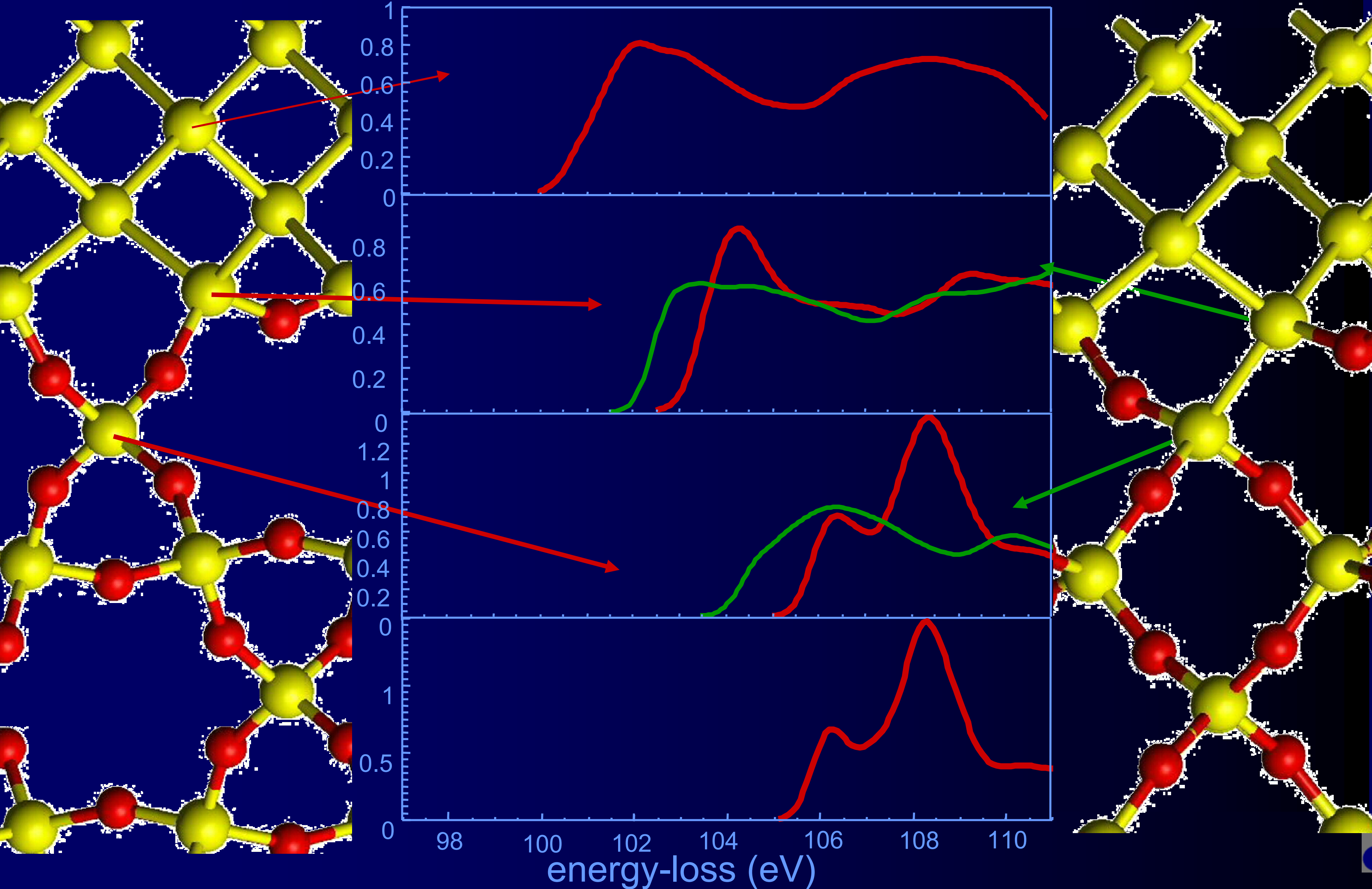
Full band gap seen 0.5 nm into oxide

O-K Ionization Edge at the Si/SiO$_2$ IF

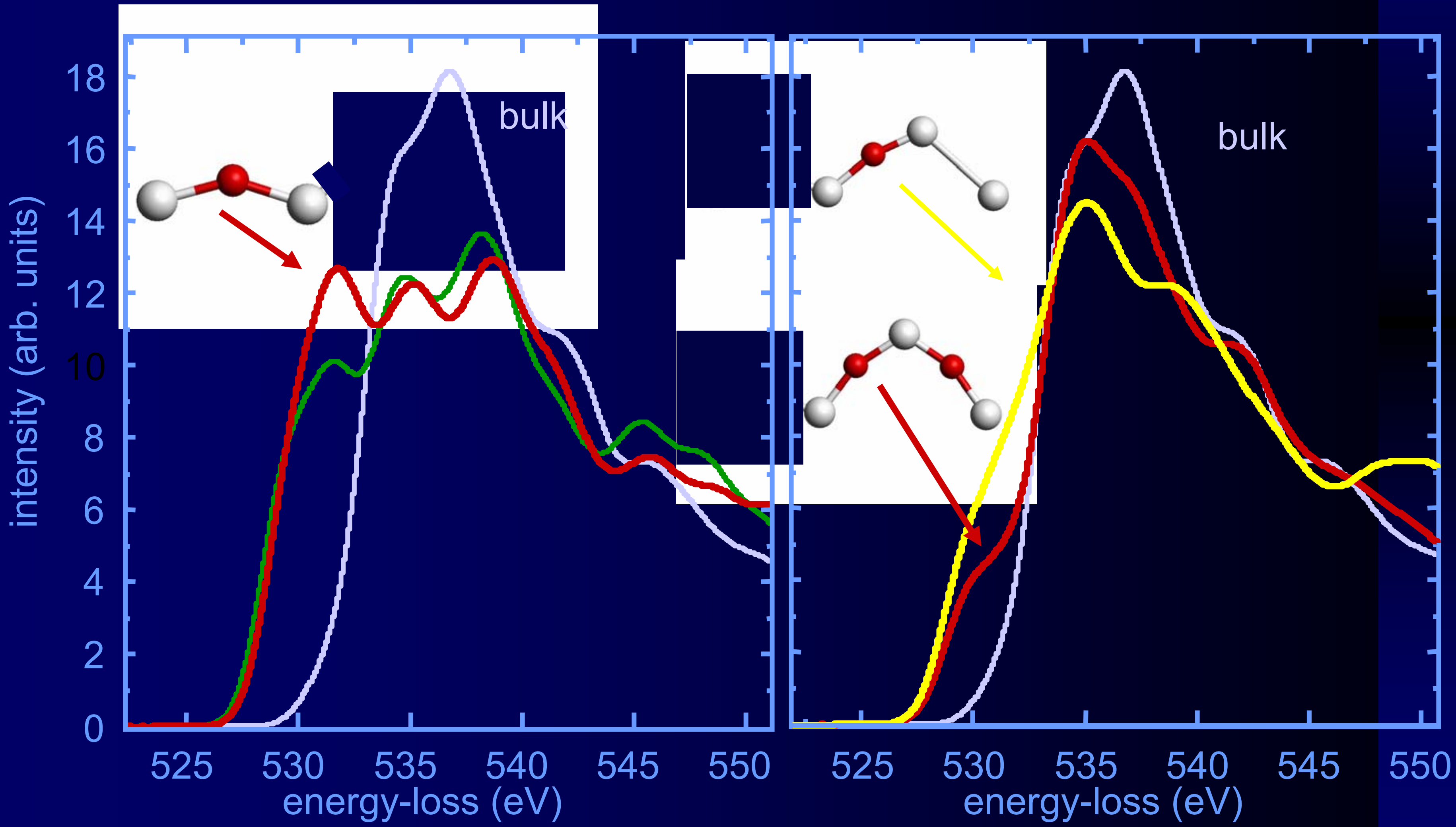


NCSU

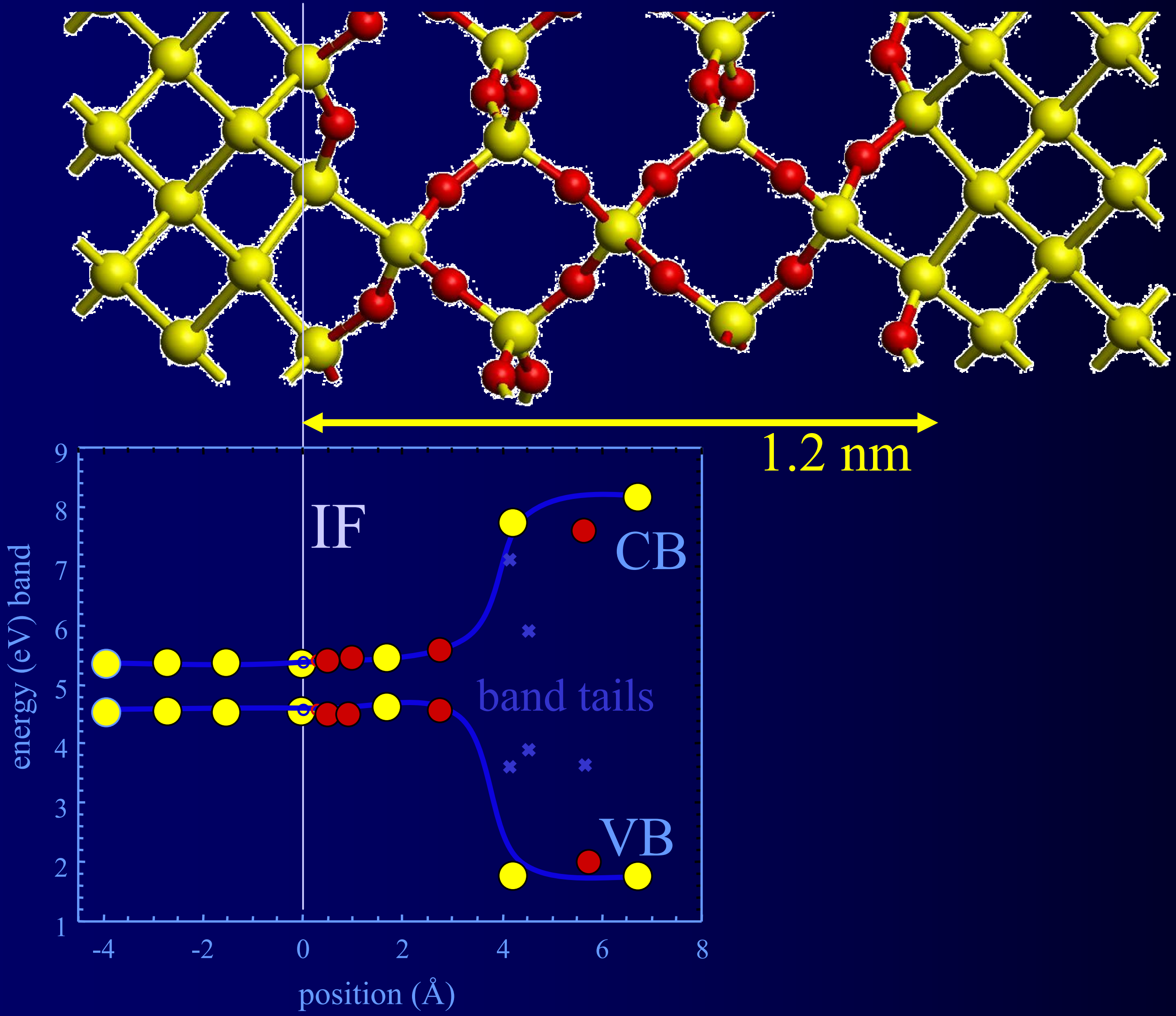
Calculated Si-$L_{2,3}$ Edges at Si-SiO_2



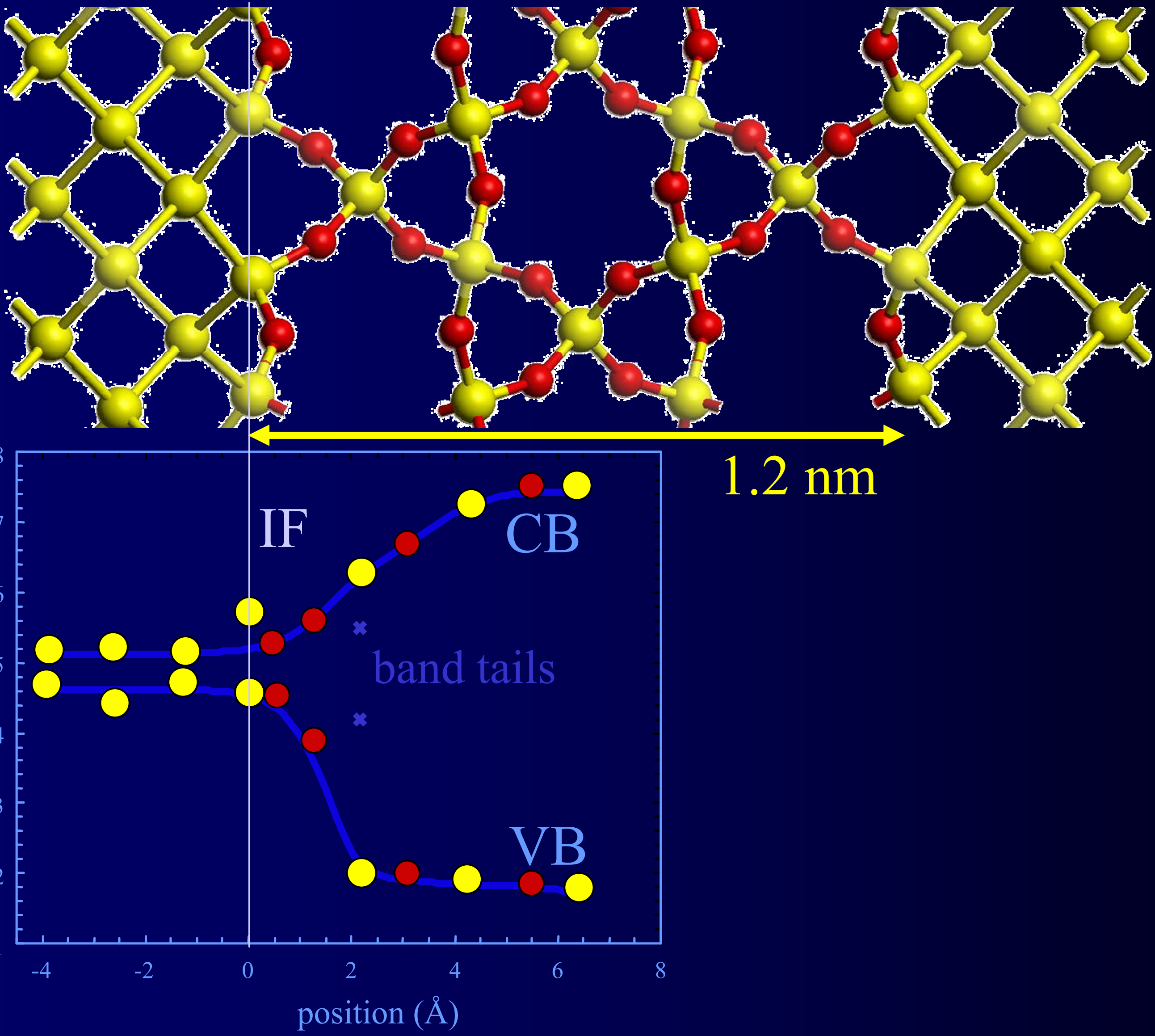
Calculated O-K Edges at the Si-SiO$_2$ IF



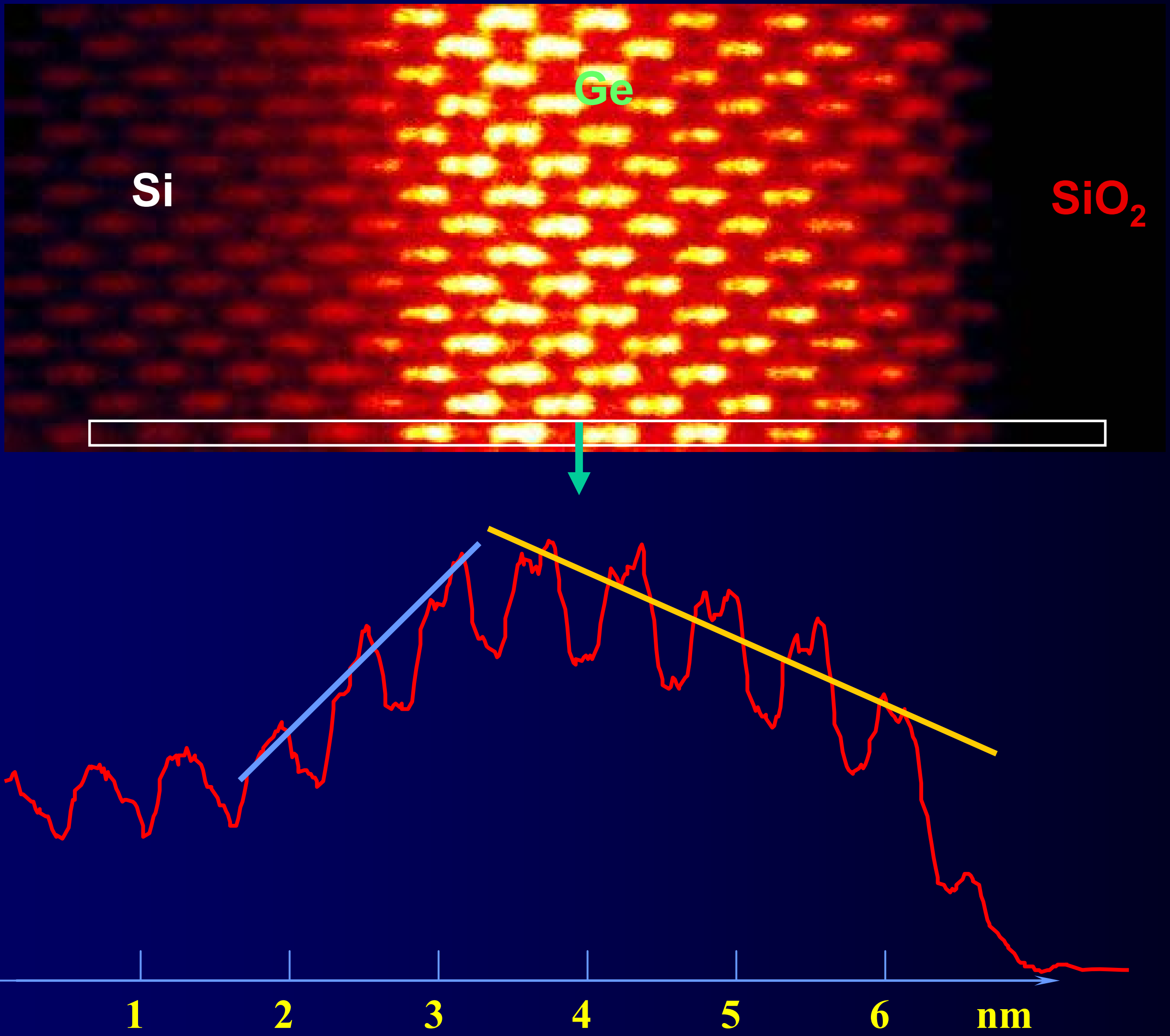
Si/SiO$_2$ Interface with Suboxide



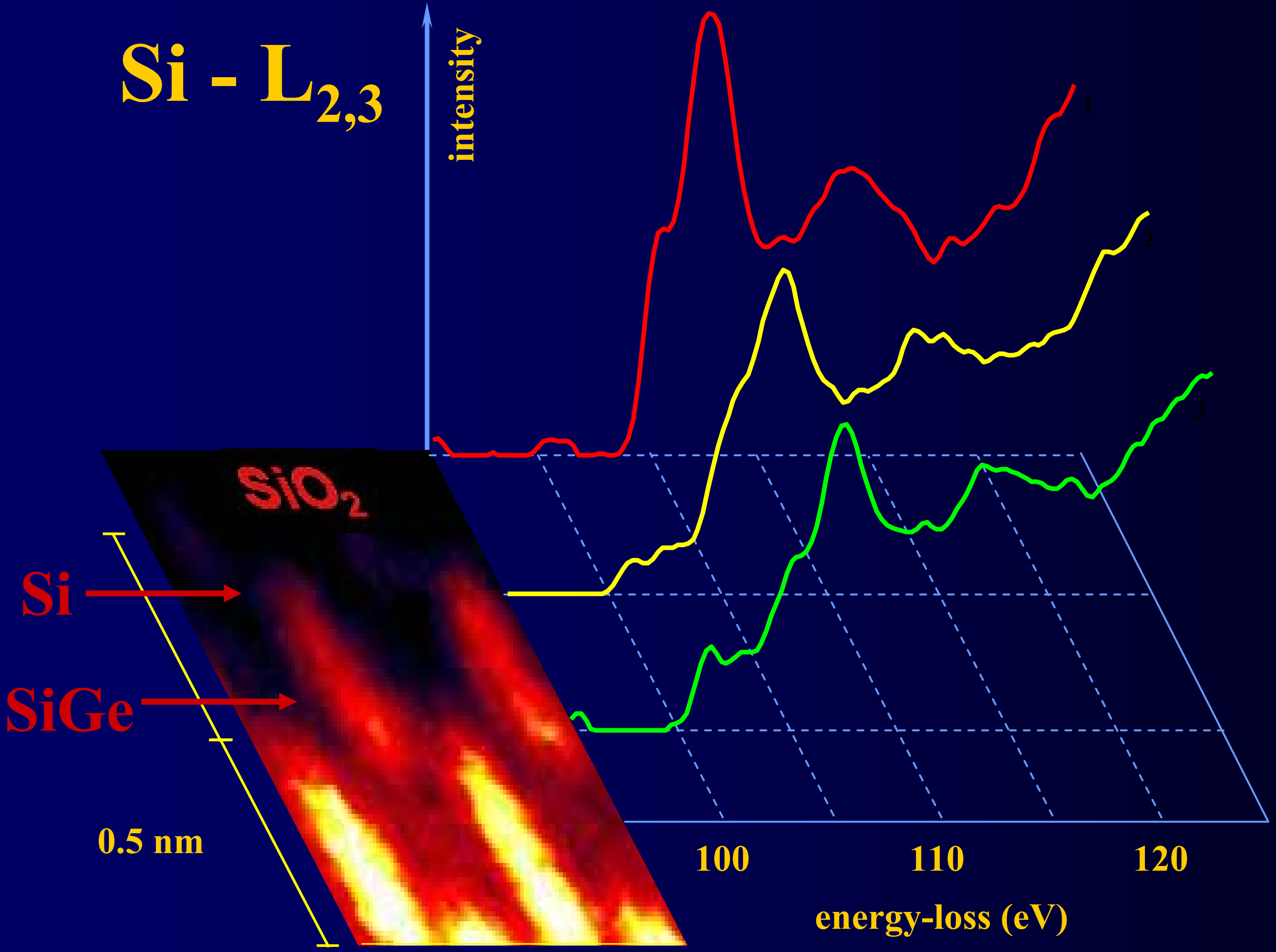
Abrupt Si/SiO$_2$ Interface



Z-Contrast at Si/Ge/SiO$_2$ Interfaces

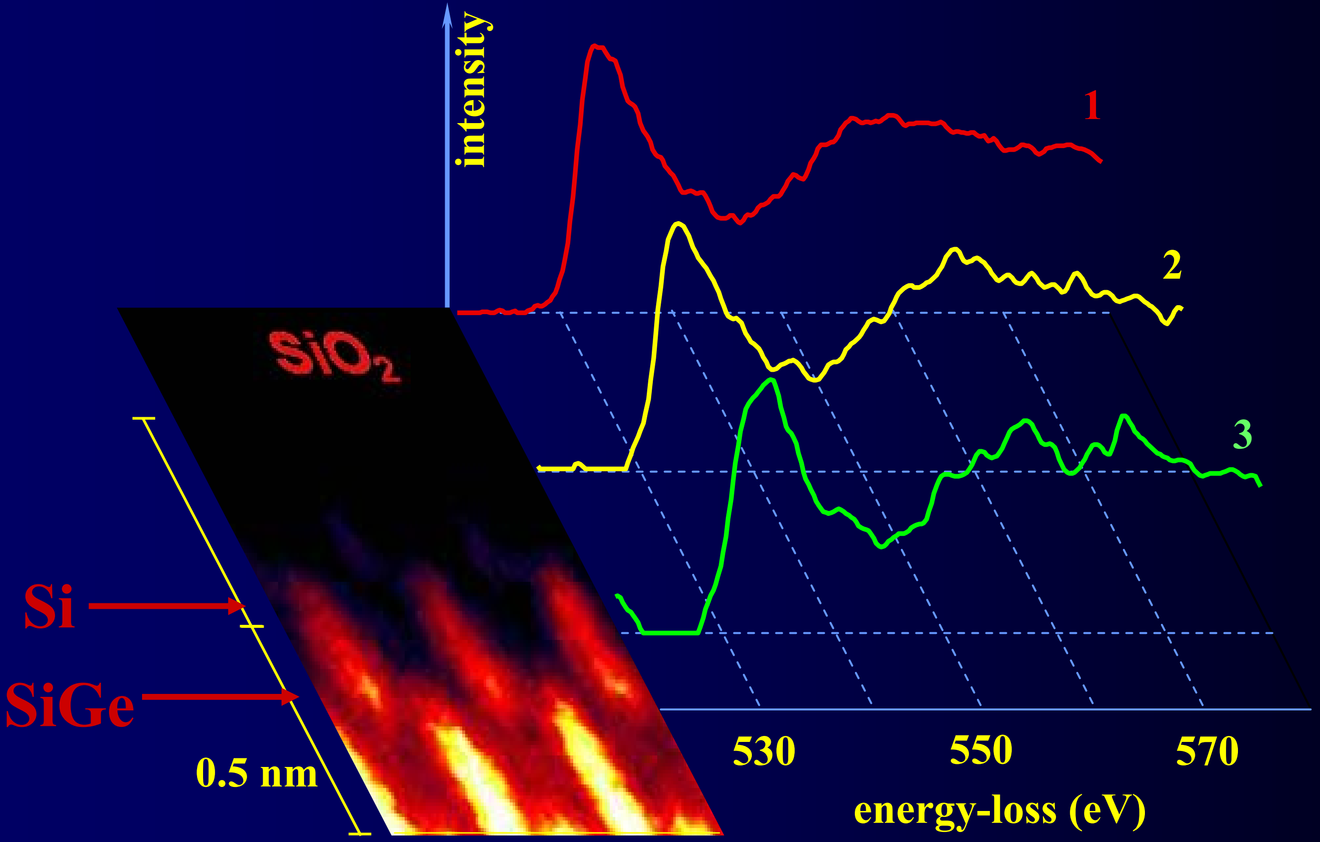


EELS at the Ge/SiO$_2$ Interfaces

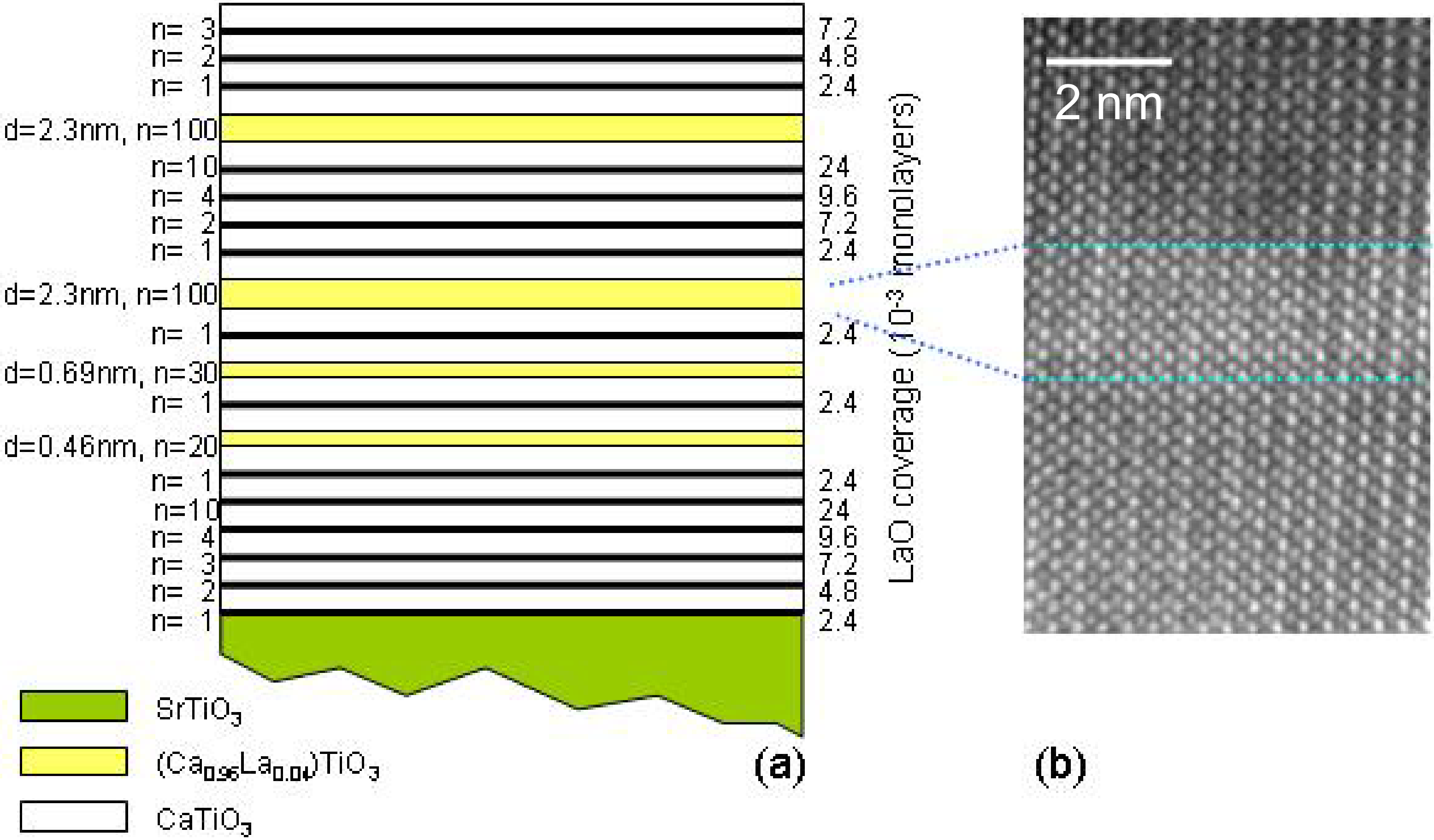


EELS at the Ge/SiO$_2$ Interface

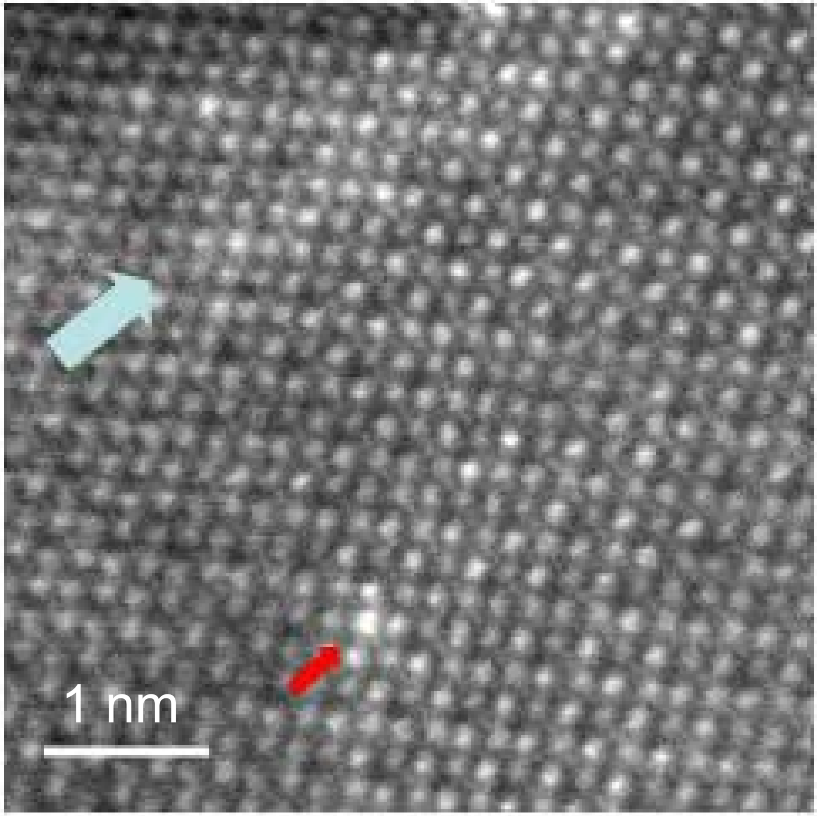
O-K

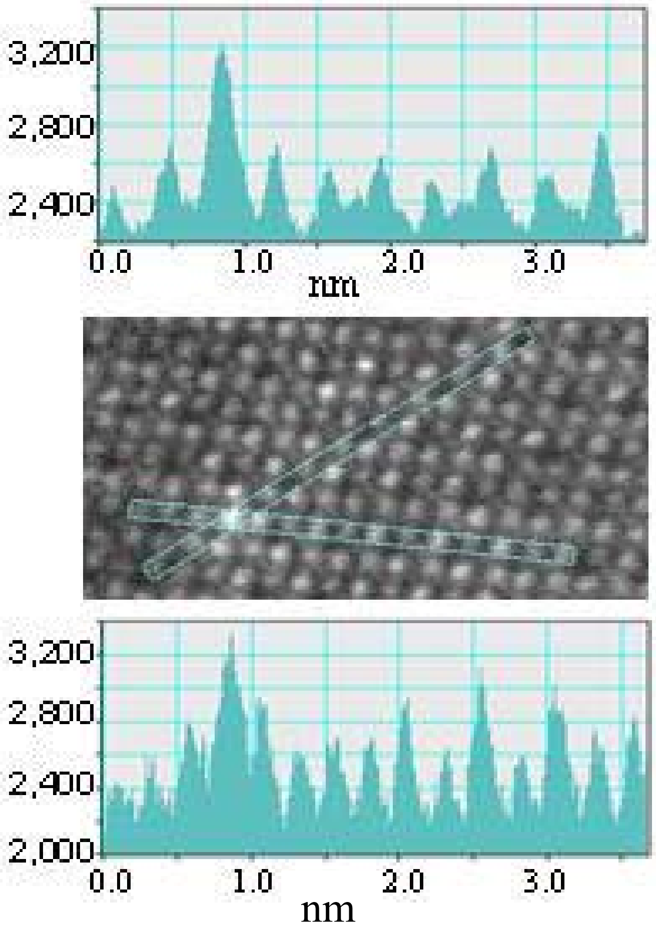


EELS: Single Atom Detection

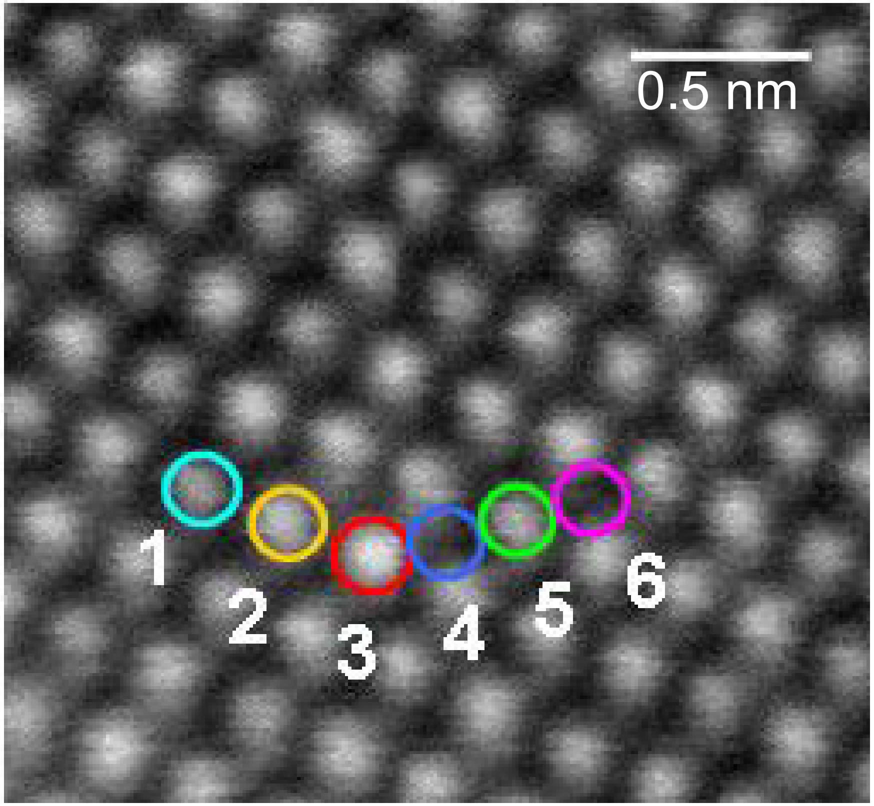


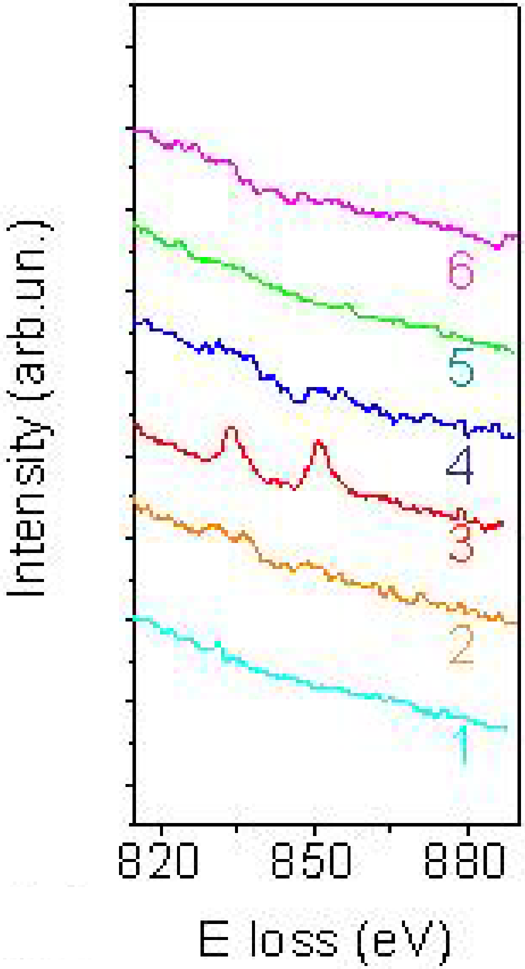
EELS: Single Atom Detection





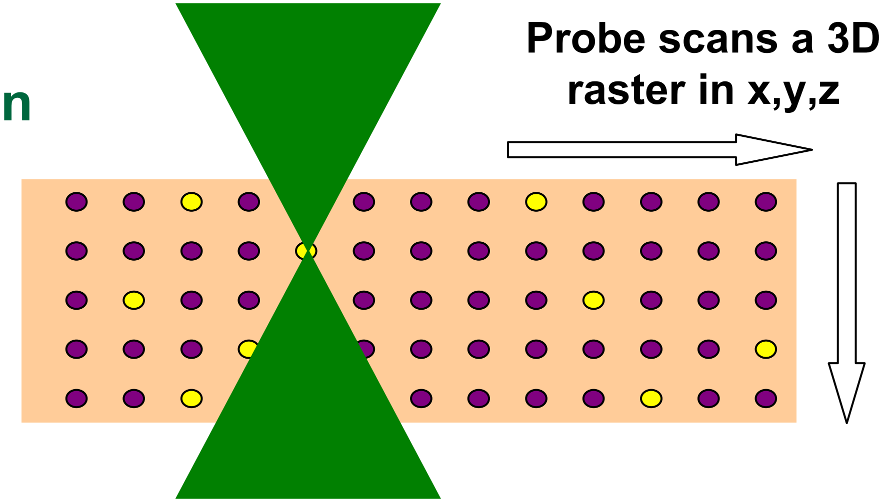
EELS: Single Atom Detection





Aberration-corrected STEM:

- **Single atom sensitivity in imaging and spectroscopy**
  - Dopants
  - Sites: interstitial or substitutional
  - Special sites: steps, dislocation cores
- **3D tomography**
  - Tilt series - limited resolution
  - Confocal STEM?



DOE Transmission Electron Aberration-corrected Microscope initiative