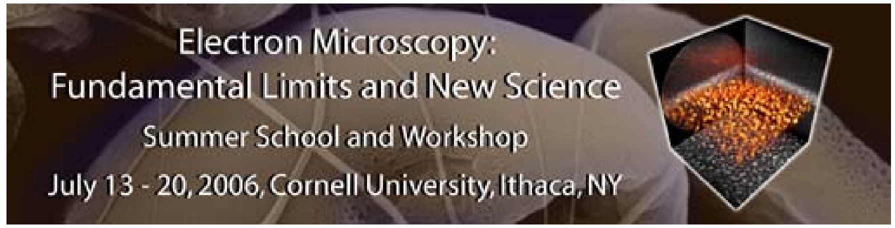
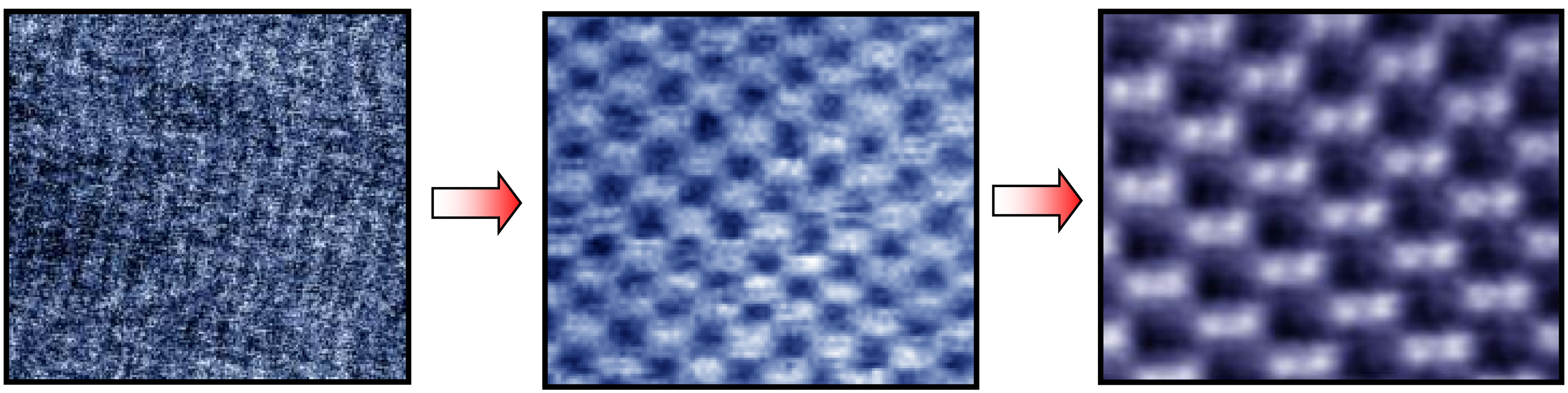
Practical STEM: More than Z Contrast



David Muller

Applied Physics, Cornell University

- Thickness dependence of images
- Common mistakes
- Alignment Tricks



3 weeks 6 months 1 year

D. A. Muller, J. Grazul, *Journal of Electron Microscopy*, **50** (2001) 219.

Acknowledgements

Matt Weyland, Zhiheng Yu,

Peter Ercius, Lena Fitting, Earl Kirkland

Applied Physics, Cornell University

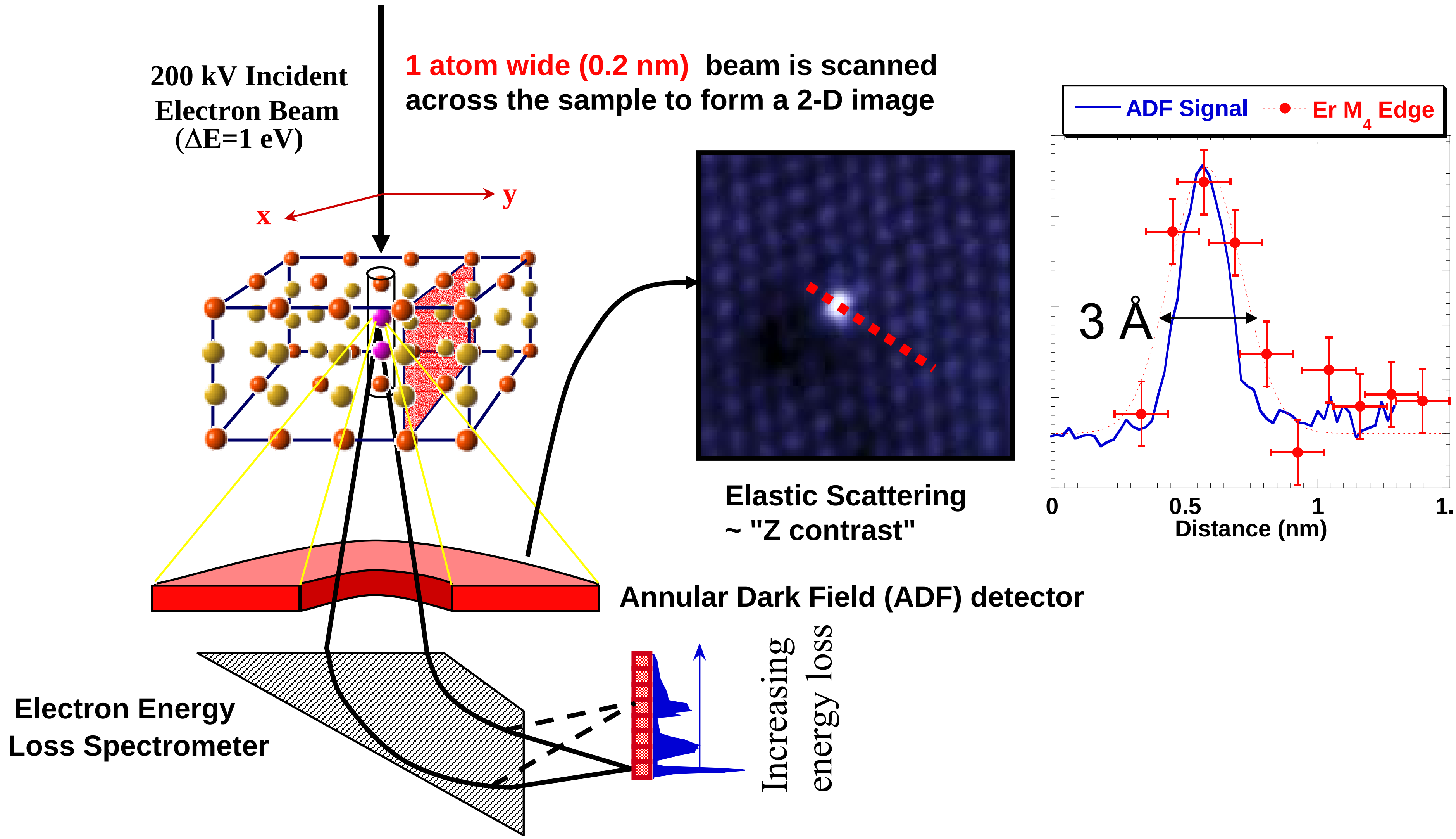
Paul Voyles

University of Wisconsin, Madison

John Grazul, Mick Thomas

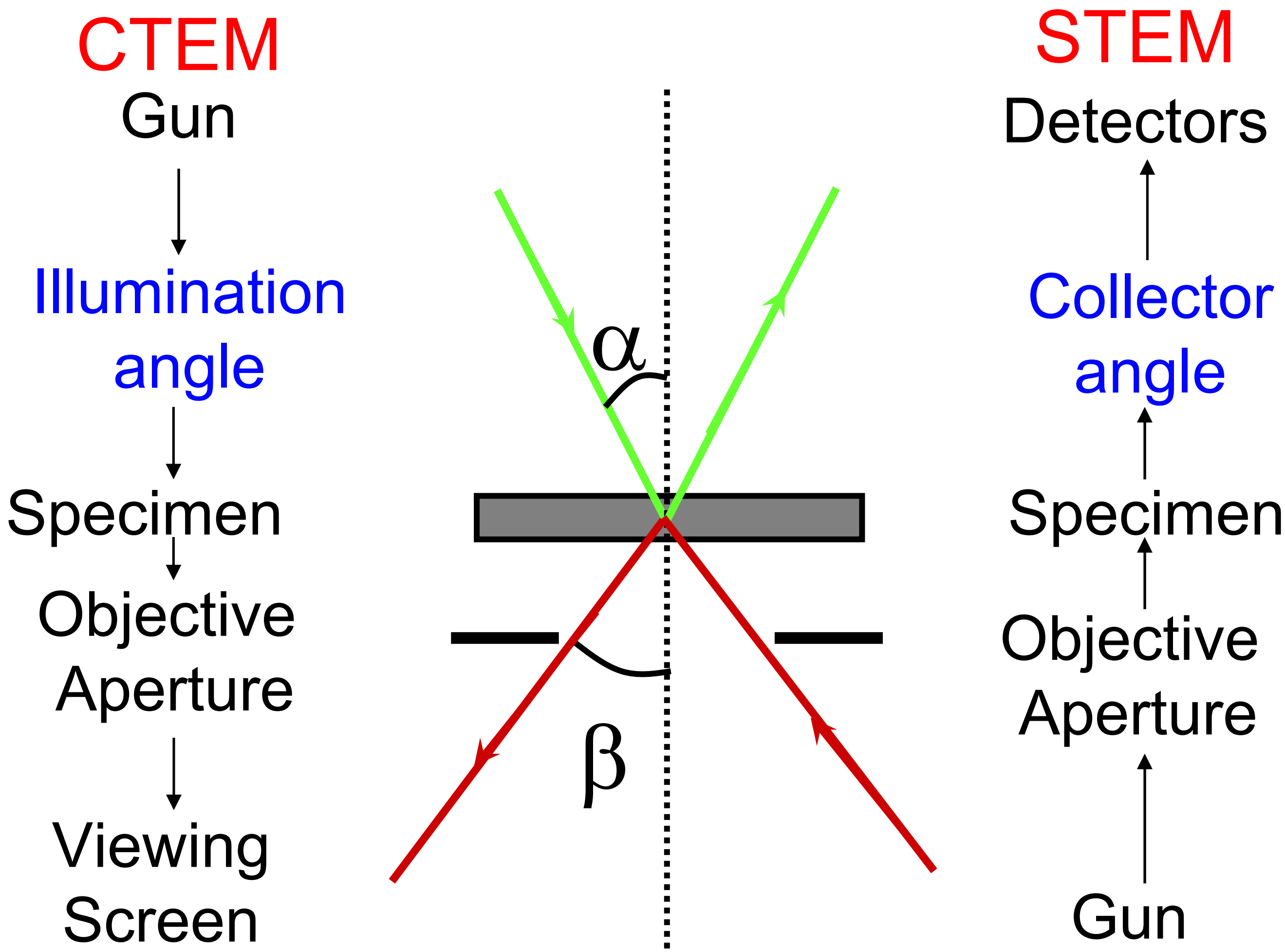
Cornell Center for Materials Research

Scanning Transmission Electron Microscopy



Single atom Sensitivity: P. Voyles, D. Muller, J. Grazul, P. Citrin, H. Gossmann, *Nature* **416** 826 (2002)
U. Kaiser, D. Muller, J. Grazul, M. Kawasaki, *Nature Materials*, **1** 102 (2002)

Reciprocity *(or STEM vs. CTEM)*



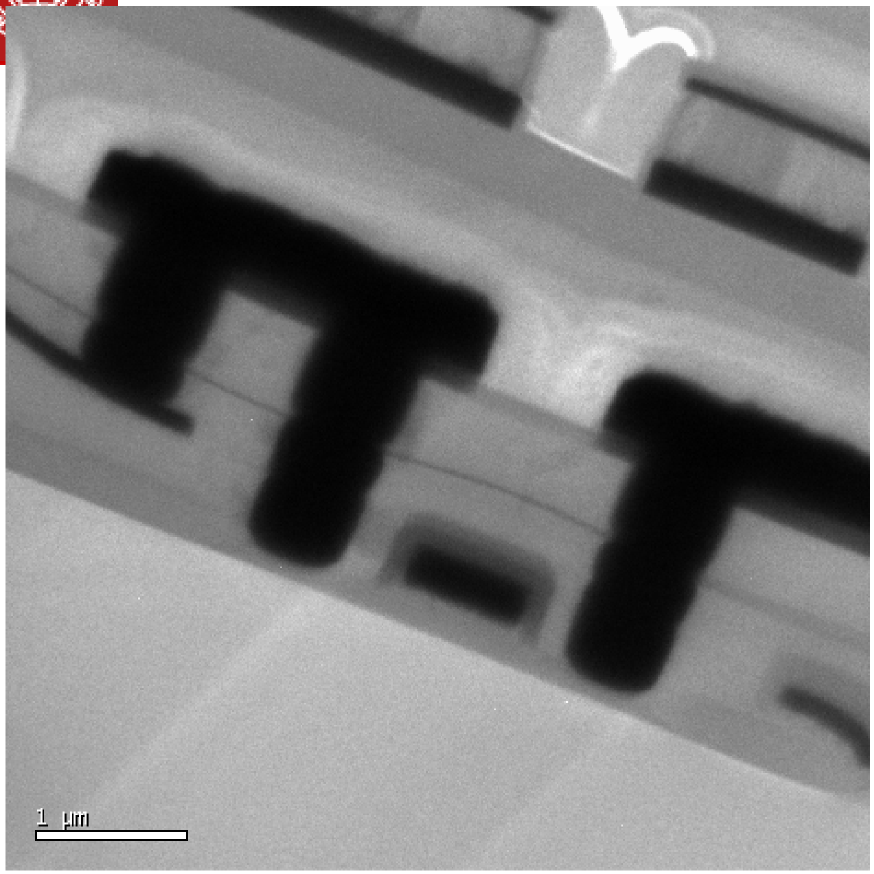
Reciprocity (for zero-loss images):

A hollow-cone image in CTEM ⟺ *an annular-dark field image in STEM.*

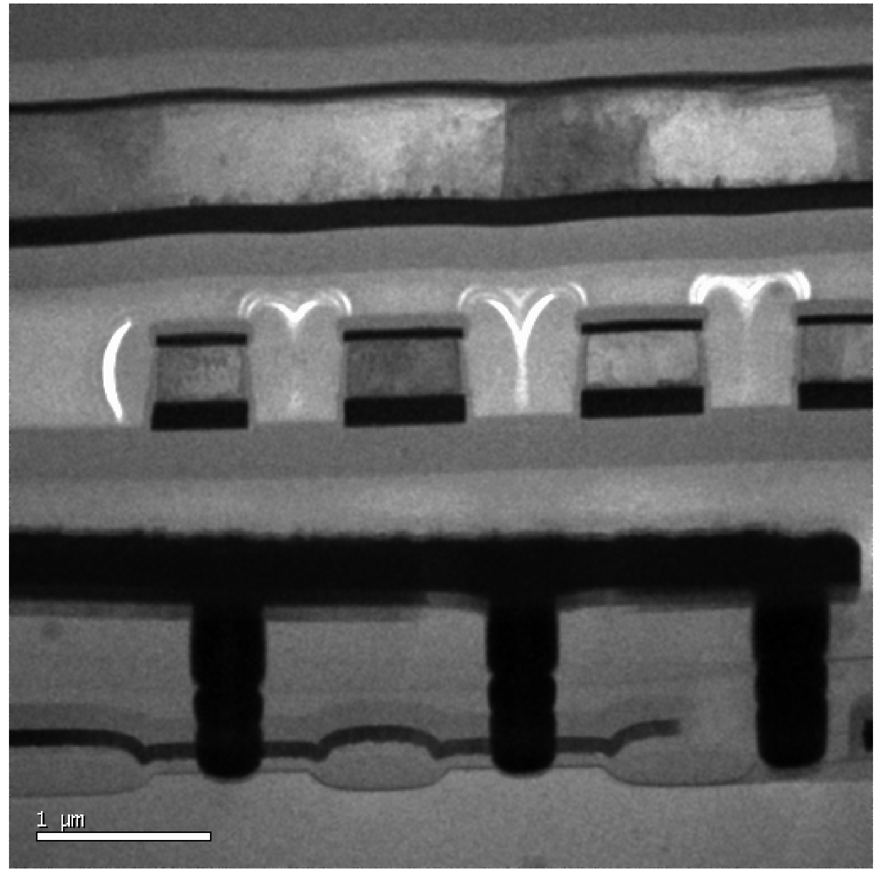
However: In STEM, energy losses in the sample do not contribute to chromatic aberrations (Strong advantage for STEM in thick specimens)

Imaging Thick Samples at 200 kV

BF-TEM



BF-STEM (~2mr θ_c)

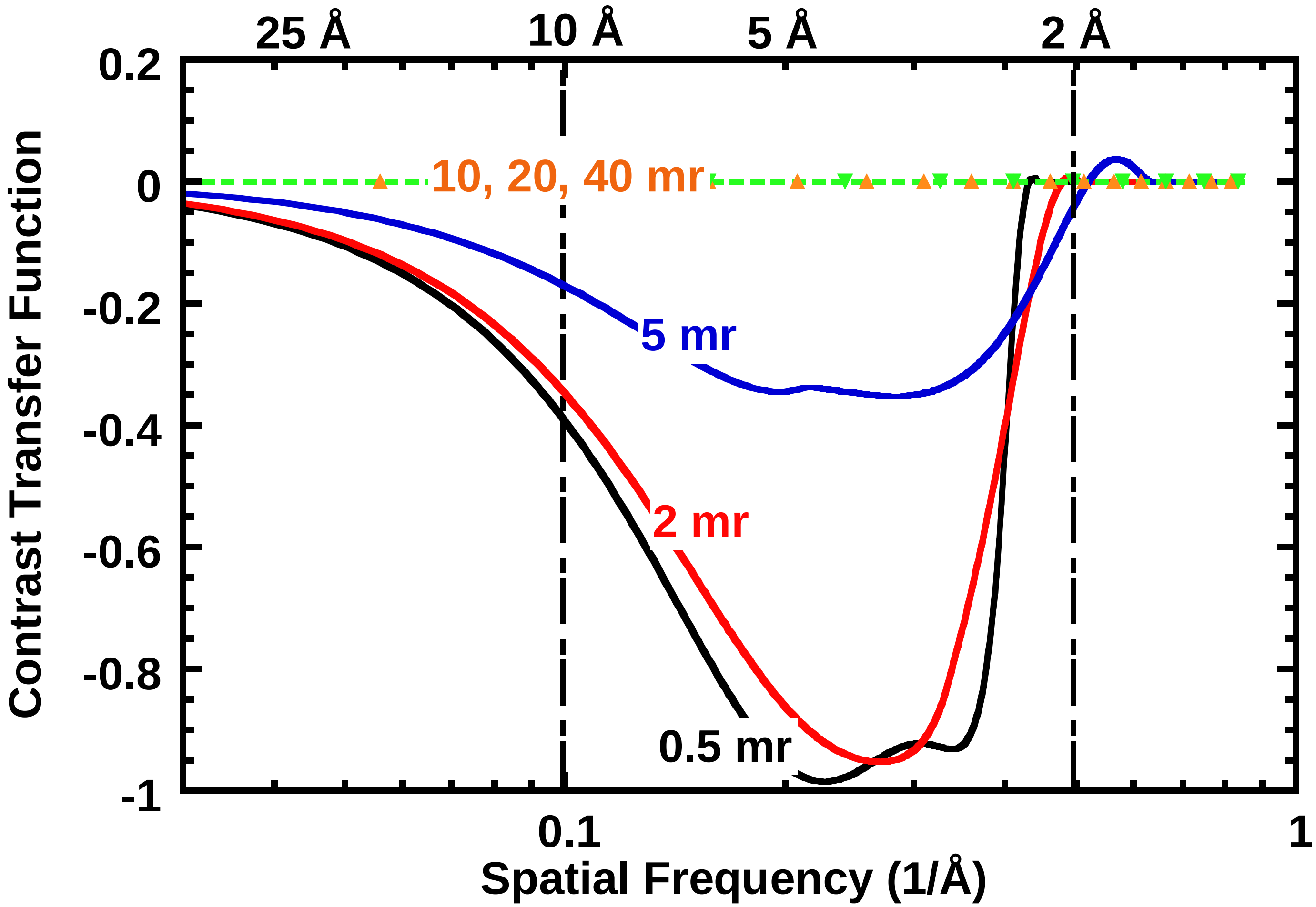


- Less blurring, more contrast in thick samples with STEM
- No Signal in W plugs, diffraction in poly

  → unsuitable for tomography

Phase Contrast for Different Illumination Angles

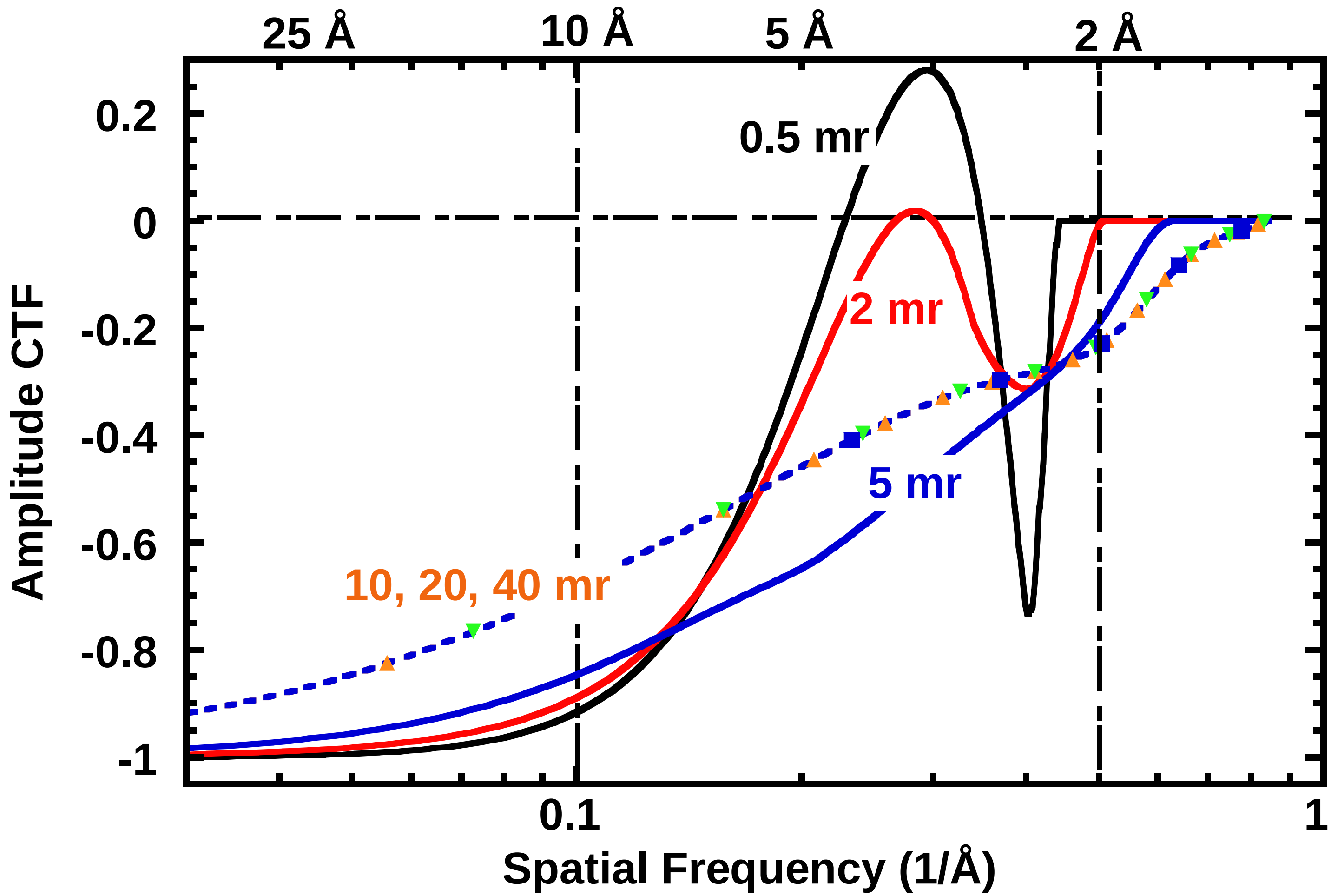
(10.5 mr Objective, E_0=200kV, Cs=1 mm, Scherzer Defocus)



- *For distances larger than 1 nm, there is little phase contrast to start with.*
- *When the illumination angles exceeds the objective aperture,* all phase contrast is suppressed!

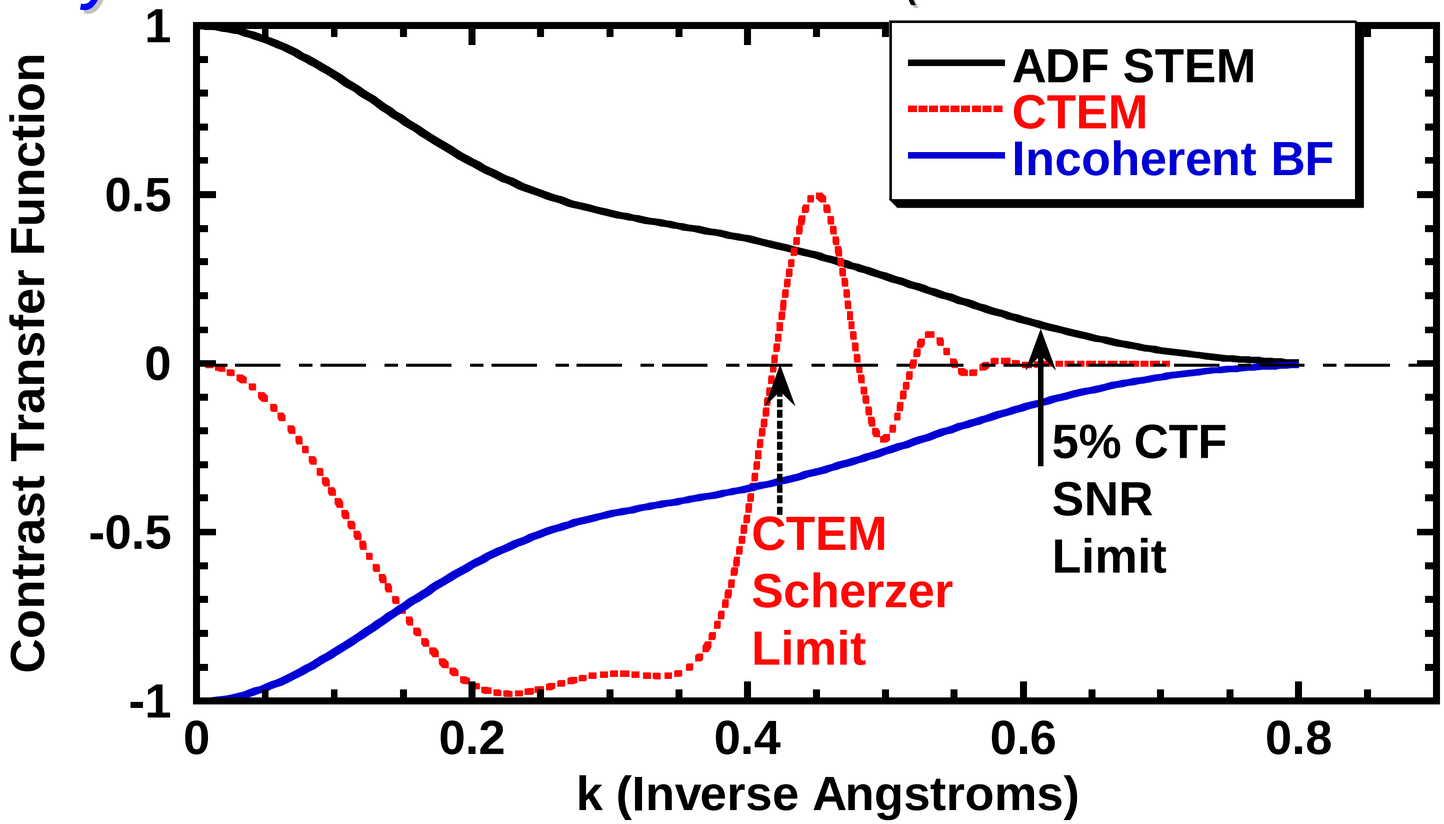
Amplitude Contrast for Different Illumination Angles

(10.5 mr Objective, E_0=200kV, Cs=1 mm, Scherzer Defocus)



- *For distances larger than 1 nm, there is little phase contrast to start with.*
- *When the illumination angles exceeds the objective aperture,*
  all contrast reversals are removed and the resolution is increased!

Why Increased Resolution? *(coherent vs. incoherent imaging)*



- E_0=200kV
- C_s=1.0 mm
- Scherzer aperture
- and defocus

- Coherent imaging PSF is the probe wavefunction,
- Incoherent imaging PSF is the square of wavefunction

Eg: $\varphi(r) \approx \exp\left(-r^2/\sigma^2\right)$

$$|\varphi(r)|^2 \approx \exp\left(-2r^2/\sigma^2\right) \Rightarrow \sigma' = \sigma/\sqrt{2}$$

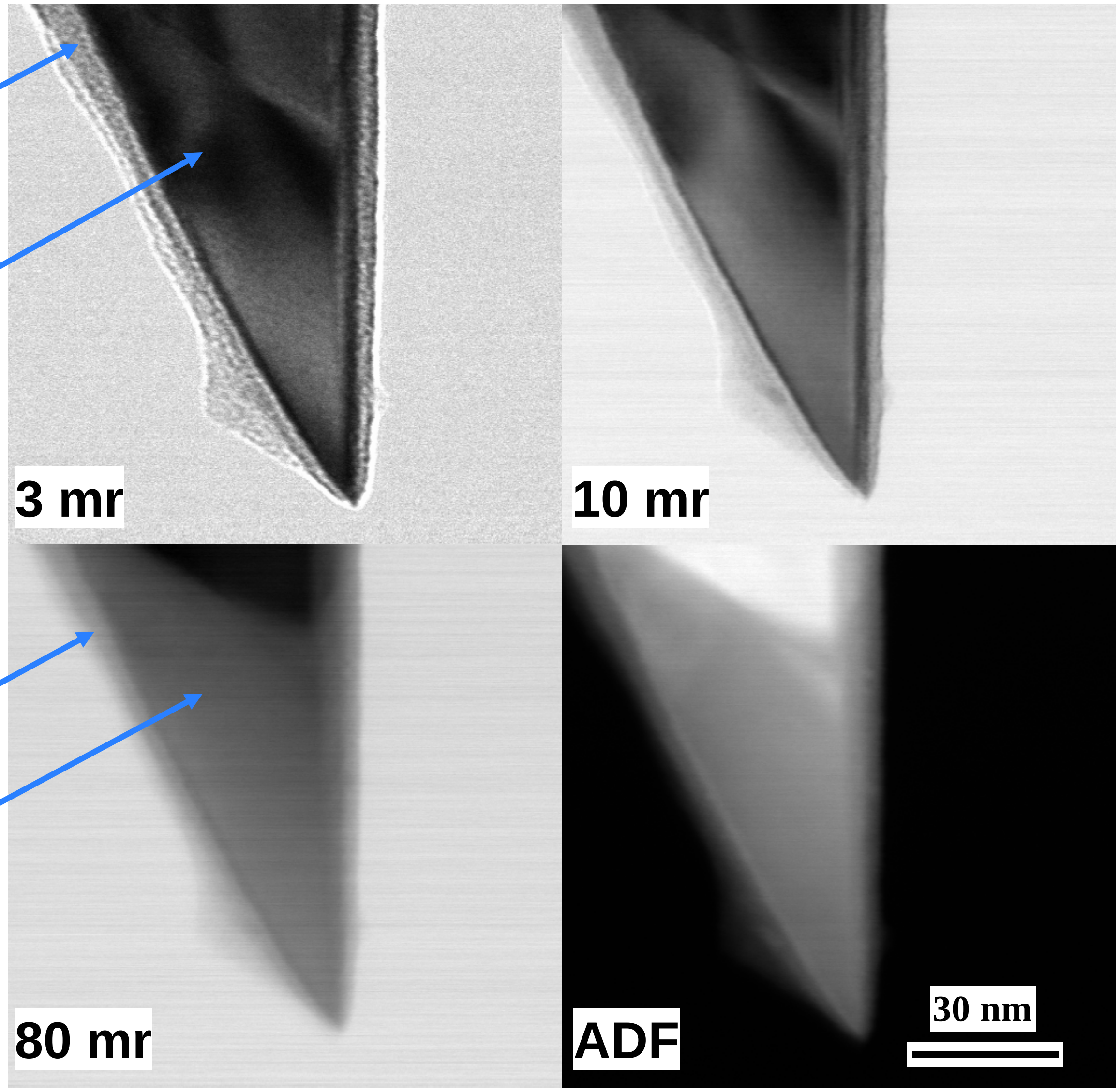
Increasing the Collector Angle (θ_c)

$\theta_c << \theta_{obj}$

- Phase contrast
- Diffraction contrast

$\theta_c \approx \theta_{obj}$

- No Phase contrast
- Diffraction contrast

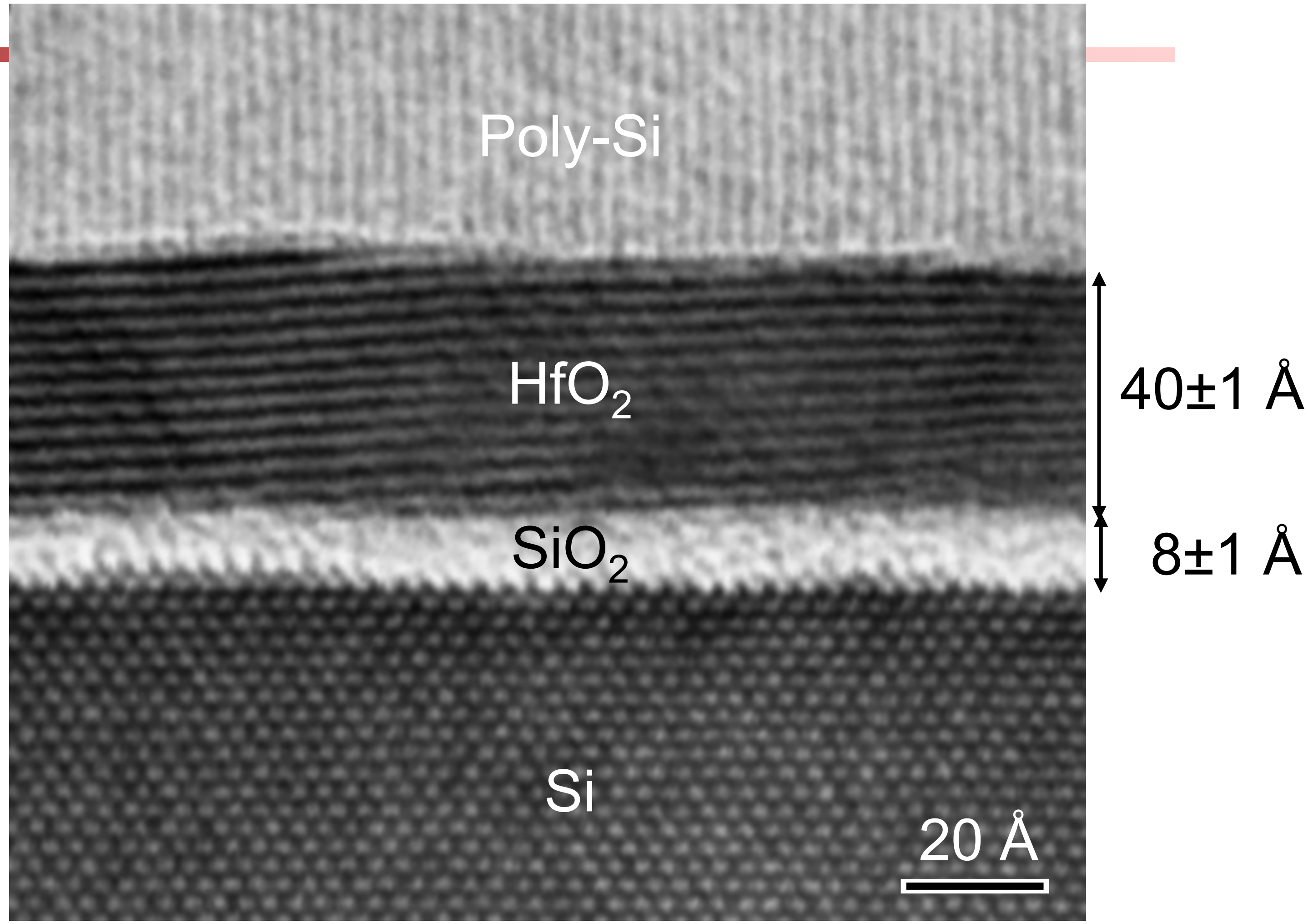


$\theta_c >> \theta_{obj}$

- No Phase contrast
- No Diffraction contrast

The incoherent BF image is the complement of the ADF image

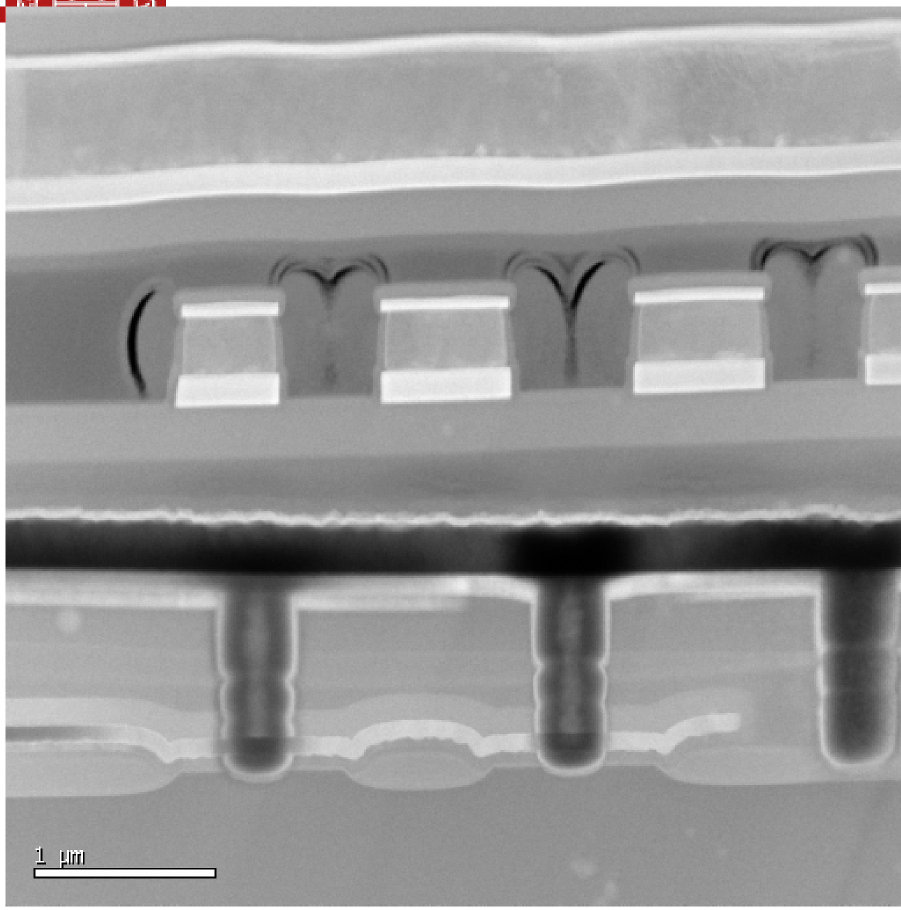
WiSi40 - 40Å HfO_2 on O_3 Underlayer + 850C/Spike/NO (6/12/2)



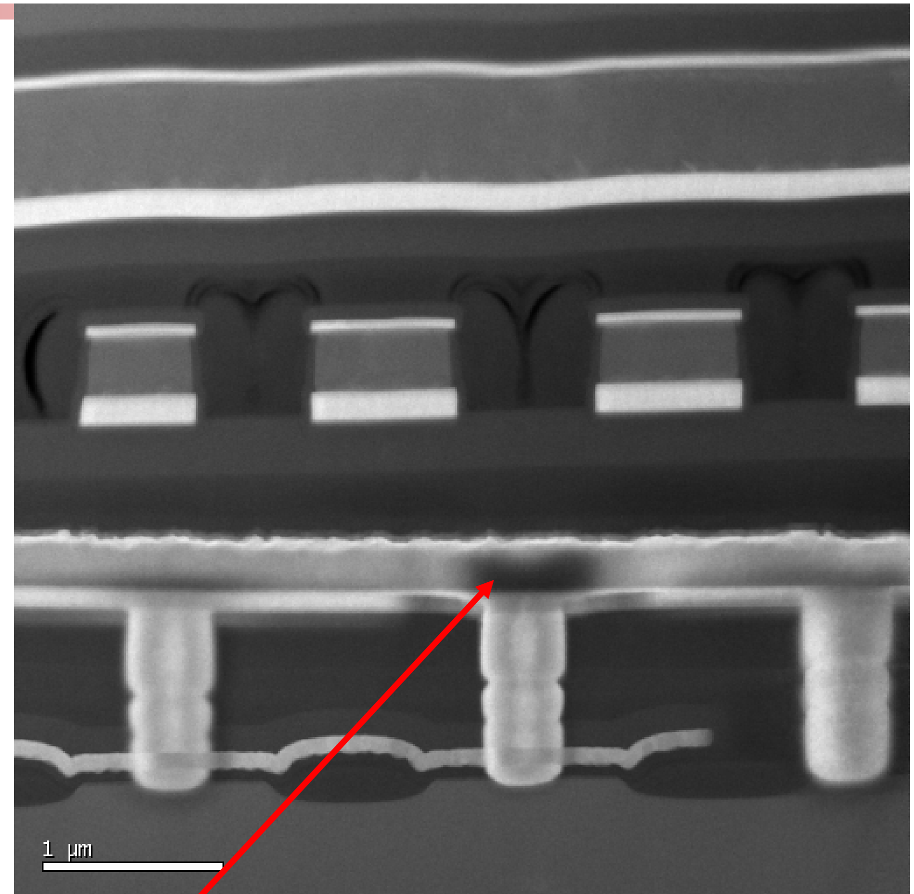
Bright-field STEM with small collector- like conventional TEM

Imaging Thick Samples at 200kV

ADF-STEM (θ_c>45 mr)

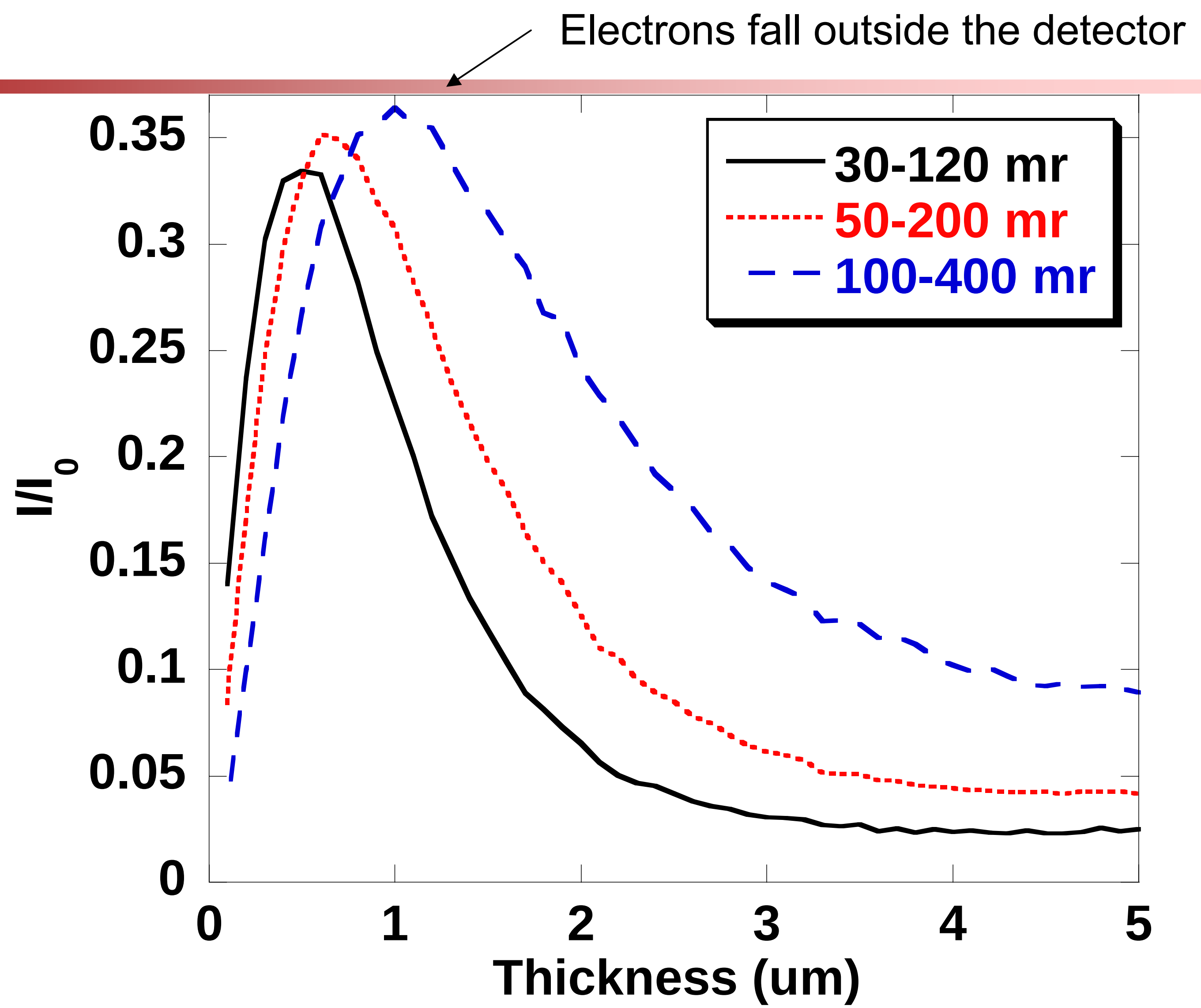


ADF-STEM (θ_c>75 mr)



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

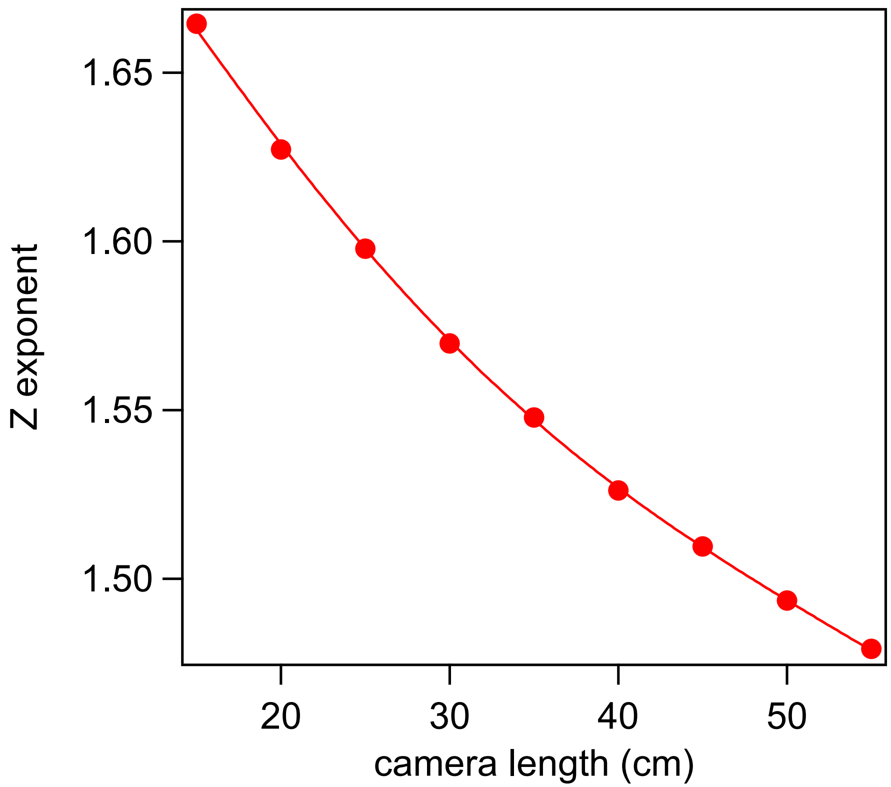
Annular Dark Field STEM at 200 kV

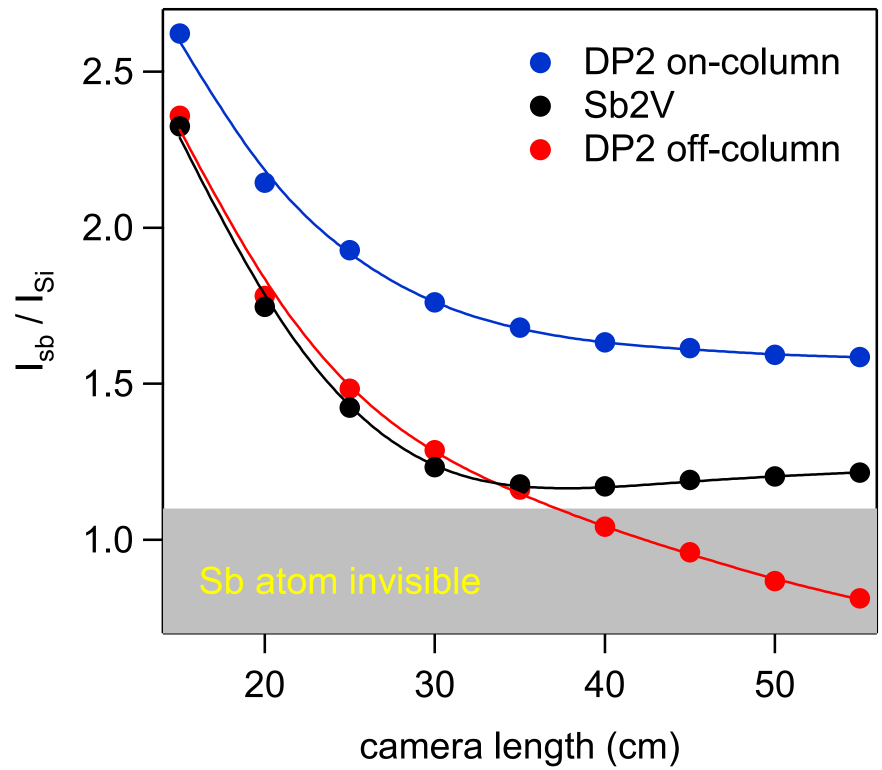


•Interpretable (monotonic, single valued) signal in Silicon to ~1 um depth

•There is a geometric limit to the detector angles (~200-400 mr)

Effect of Camera Length





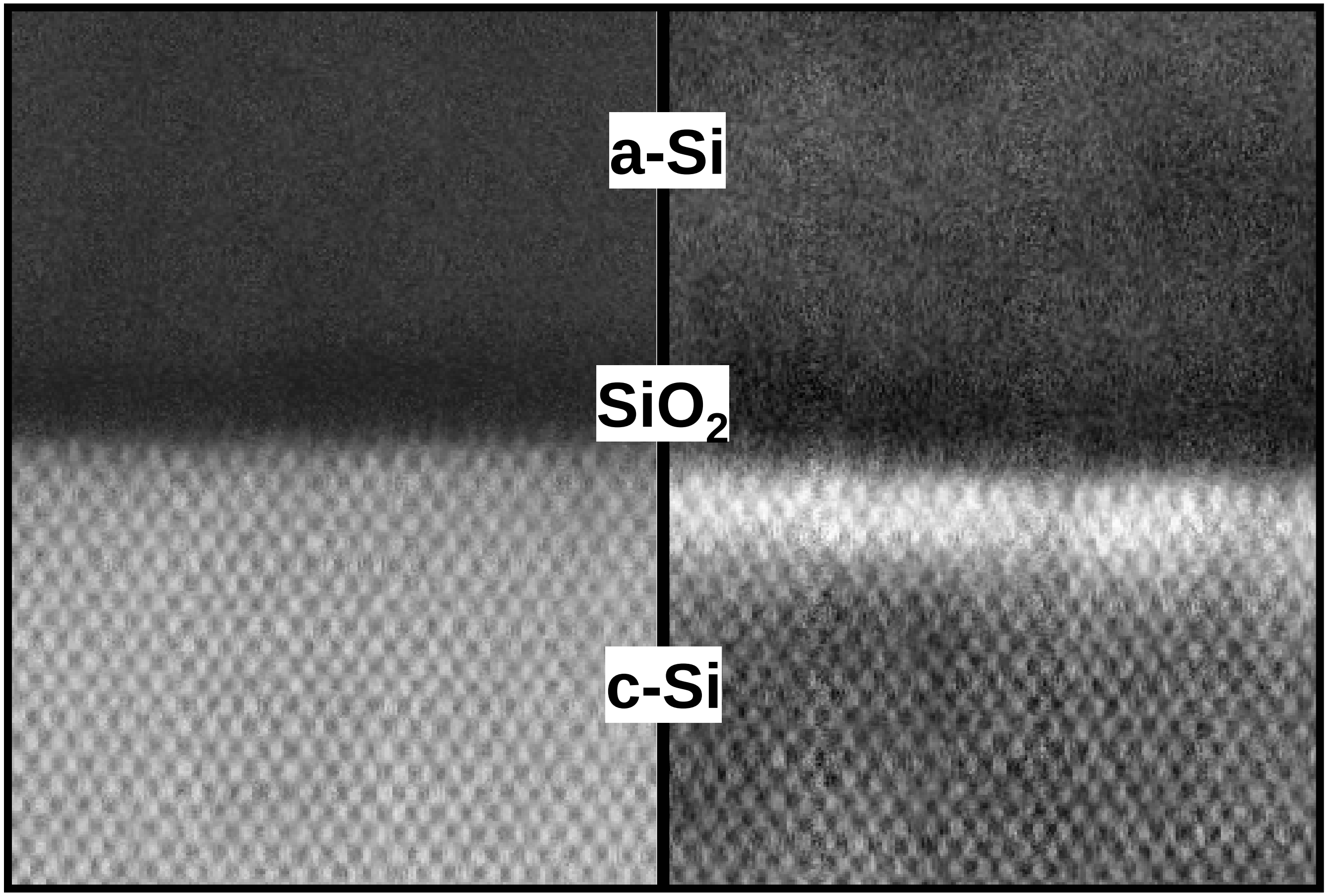
Channeling enhances on-column Sb intensity.

Strain Contrast at Si/SiO_2 Interfaces

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

ADF Inner angle:

50 mrad 25 mrad



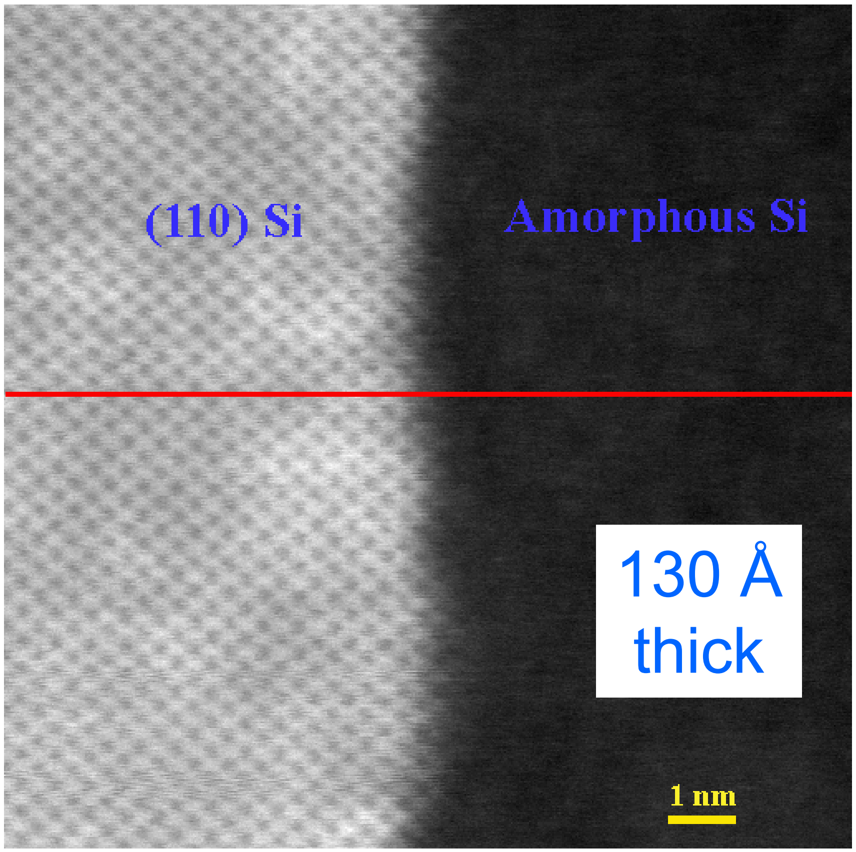
Strain Fields cause dechanneling (and scattering to small angles)

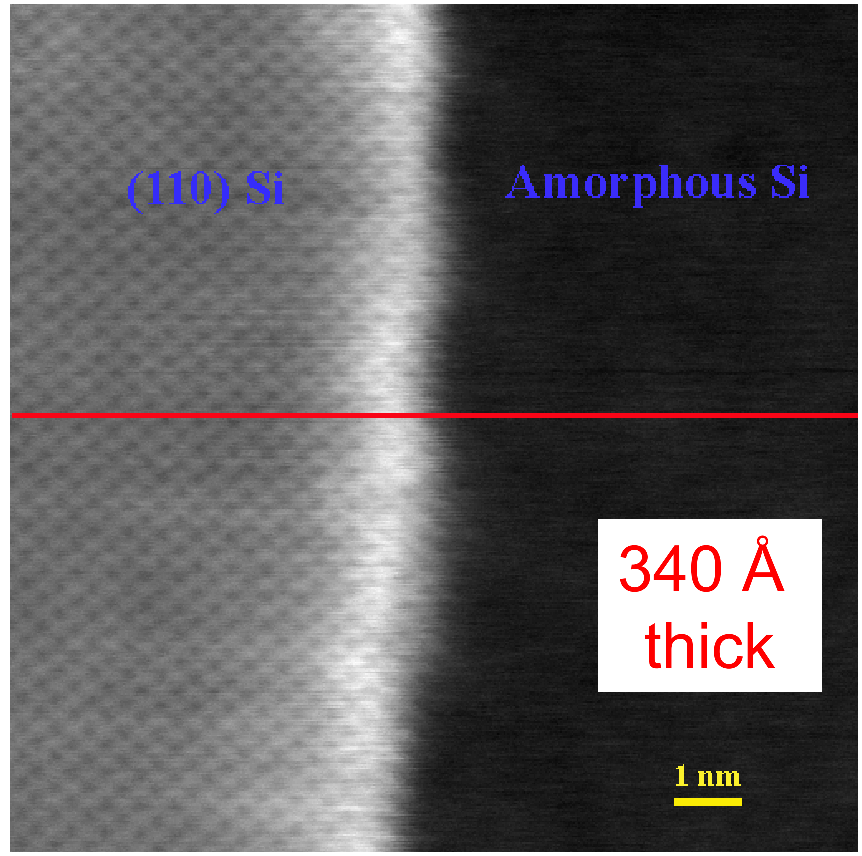
Z. Yu, D. A. Muller, and J. Silcox, *J. Appl. Phys.* **95**, 3362 (2004).

Strain Contrast vs. Sample Thickness

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

100 kV, 45 mrad ADF inner angle





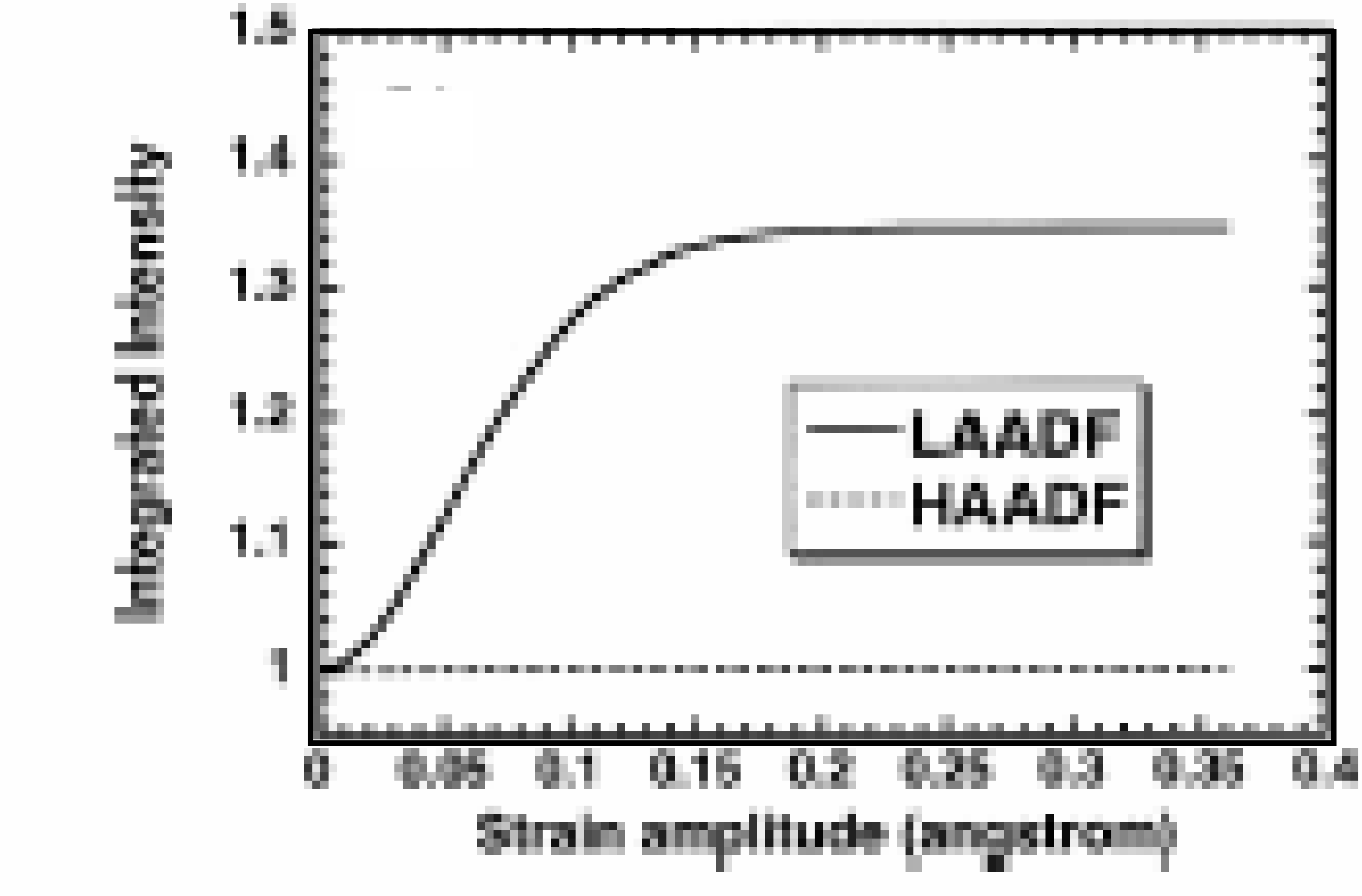
Strain Contrast effects at the interface:

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).

Contrast from Random Strain Fields

(treated as a static Debye-Waller Factor)



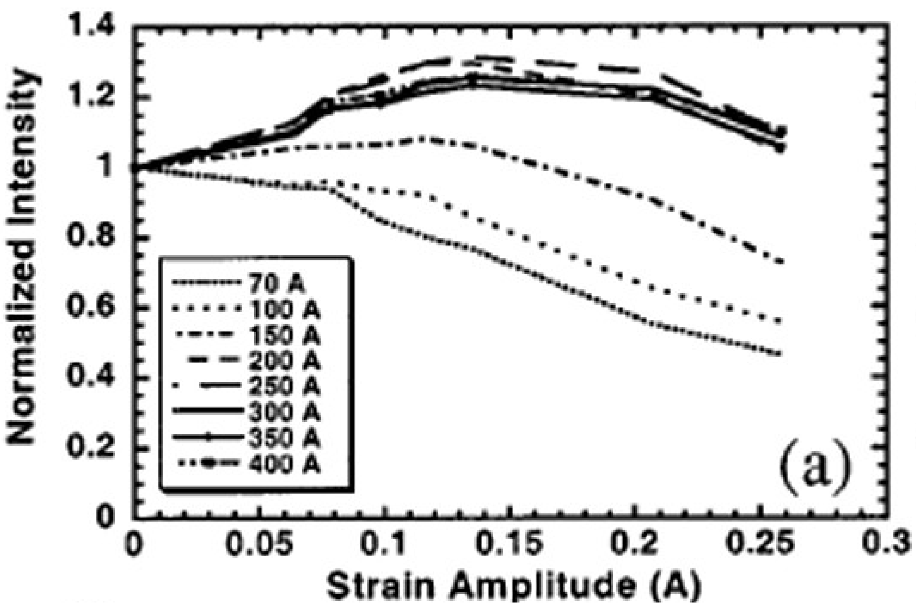
No channeling ⇒ No thickness dependence

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

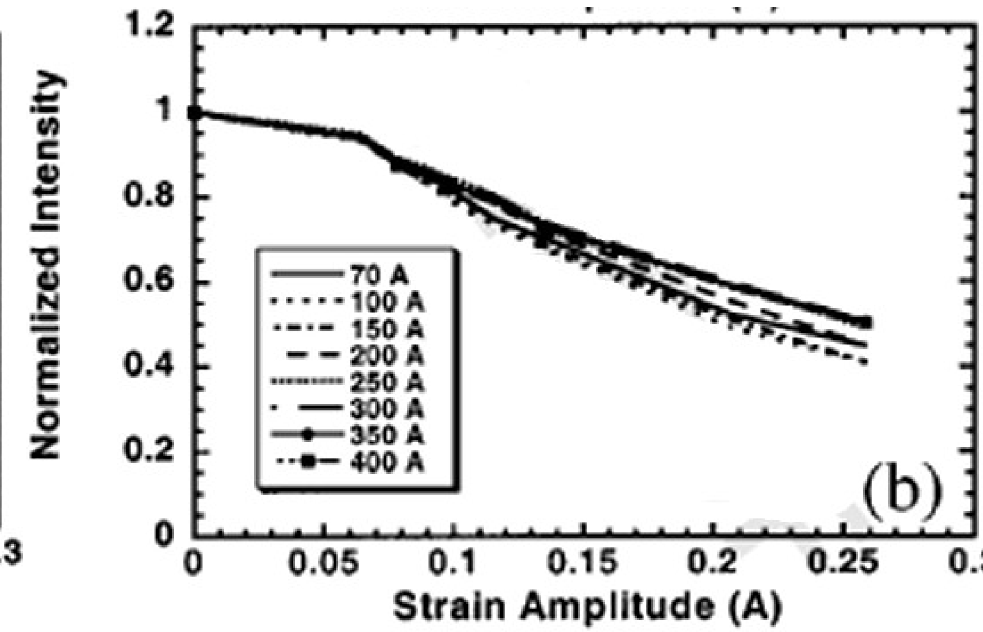
Contrast from Random Strain Fields

(using frozen phonons in multislice)

LAADF



HAADF

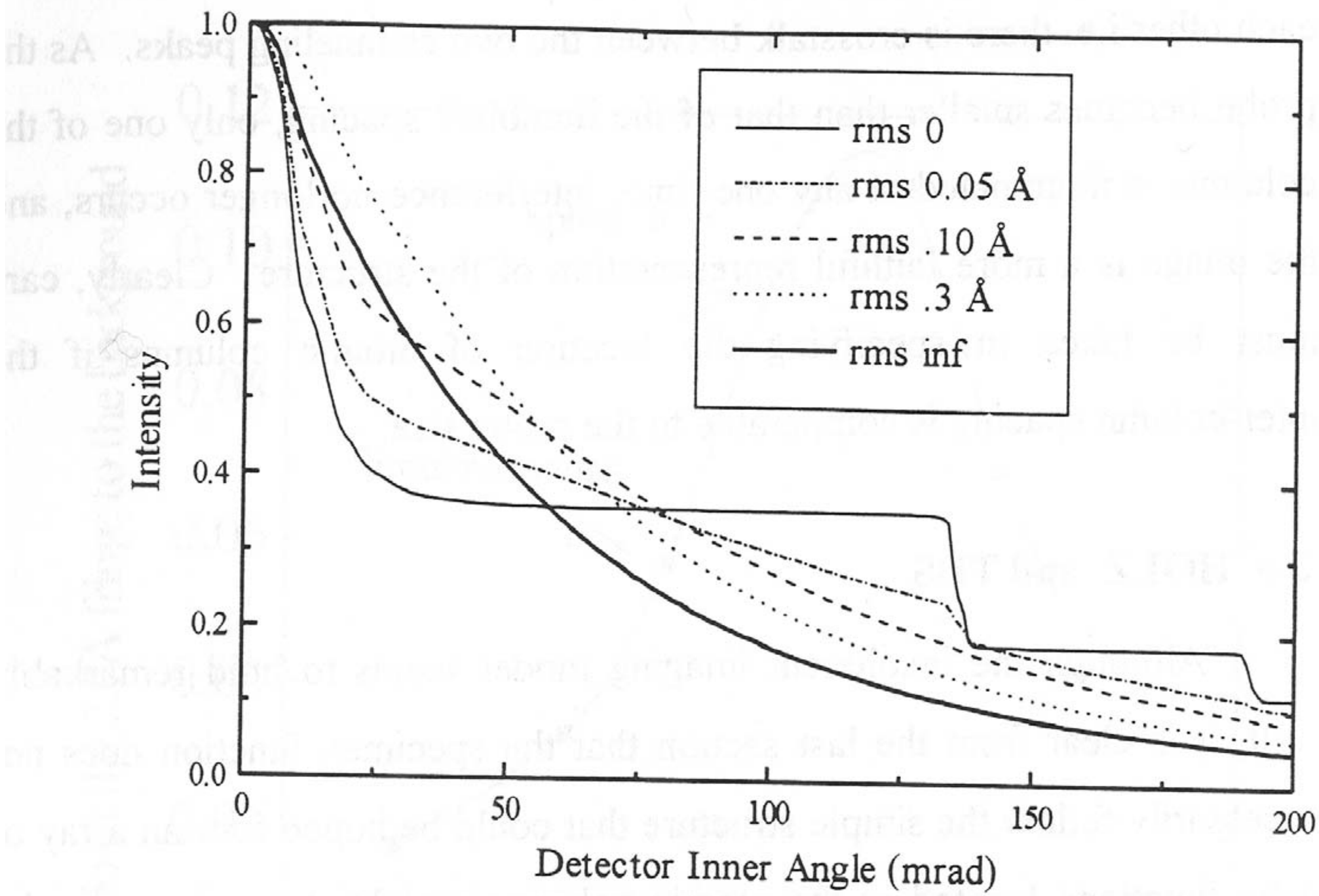


Channeling → Thickness dependence

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

Contrast from Random Strain Fields

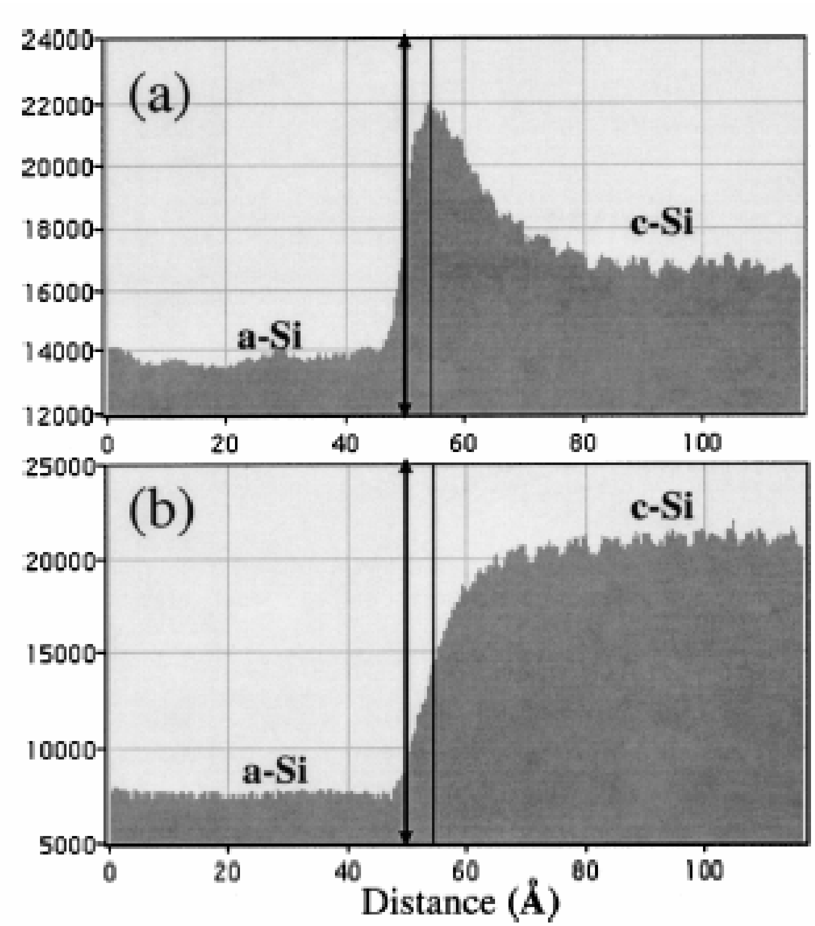
(using frozen phonons in multislice)



Channeling → Thickness dependence

Contrast from Random Strain Fields

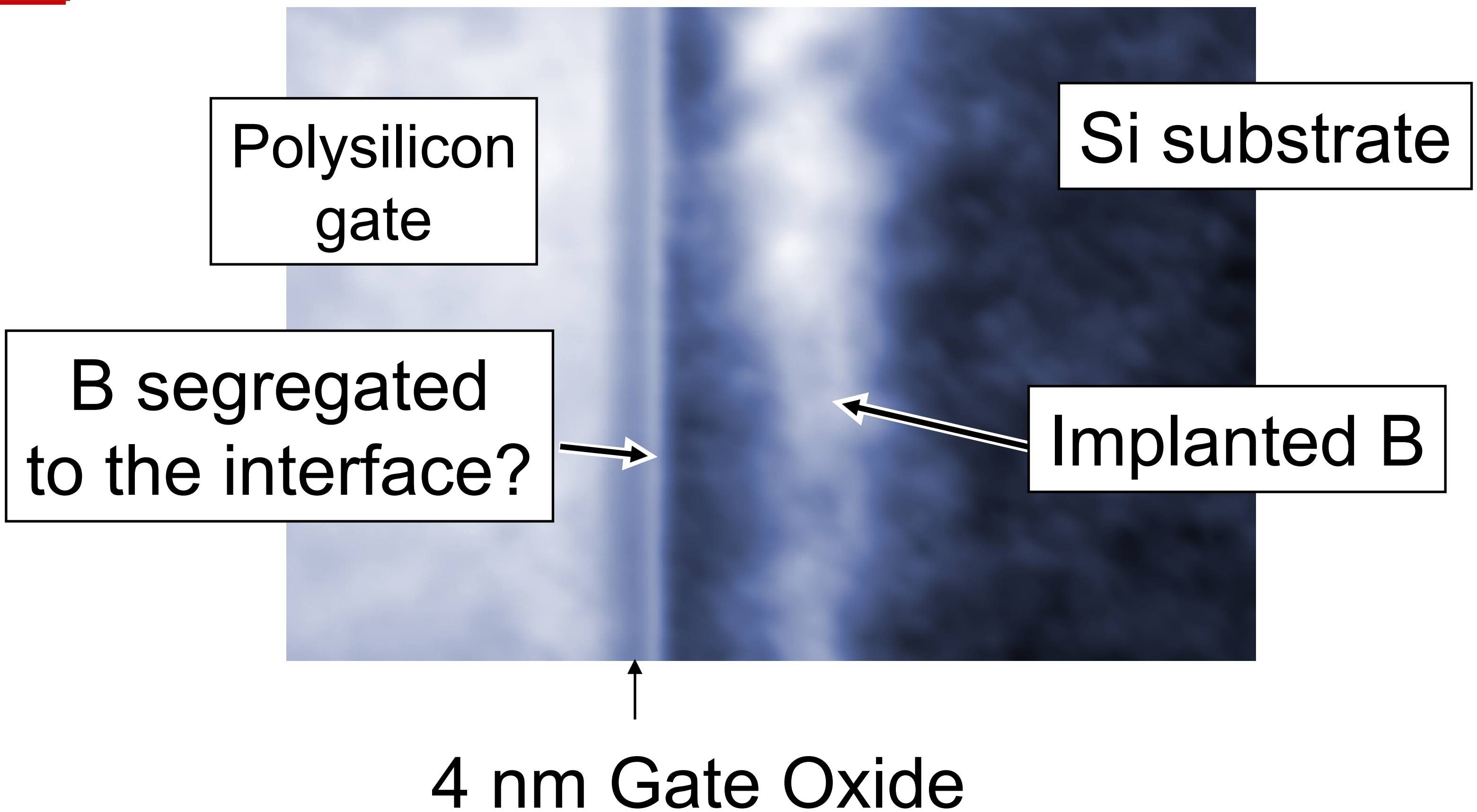
(treated as a static Debye-Waller Factor)



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

Imaging Light Atoms

Dechanneling contrast from the Strain Field around impurities



Single atom contrast is expected at 77K (Hillyard and Silcox)

Imaging Vacancies?

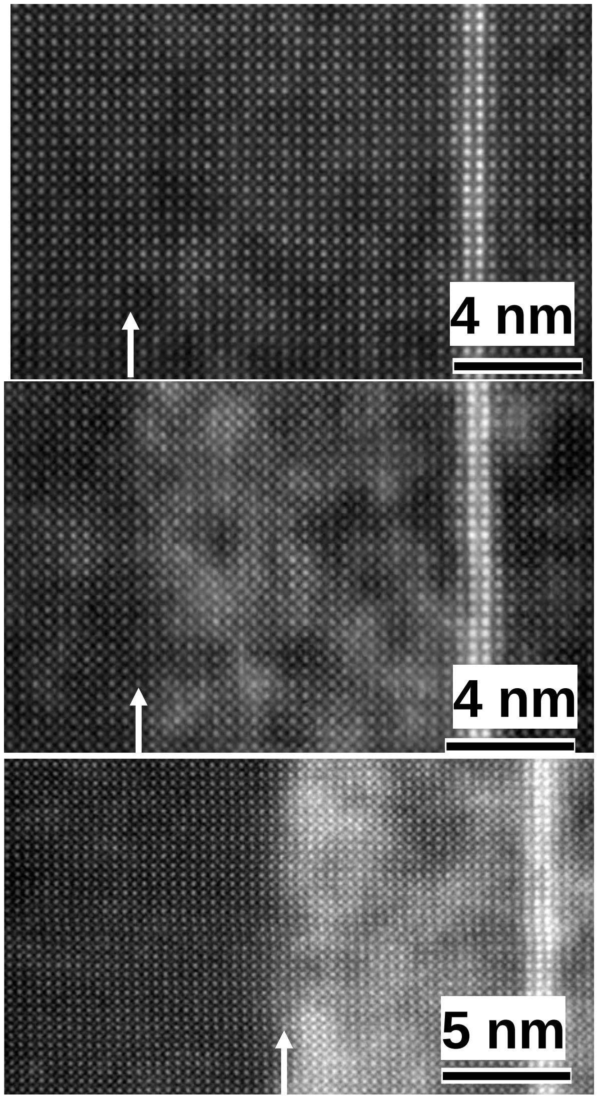
(grow 25 layers of $SrTiO_{3-\delta}$ on $SrTiO_3$)

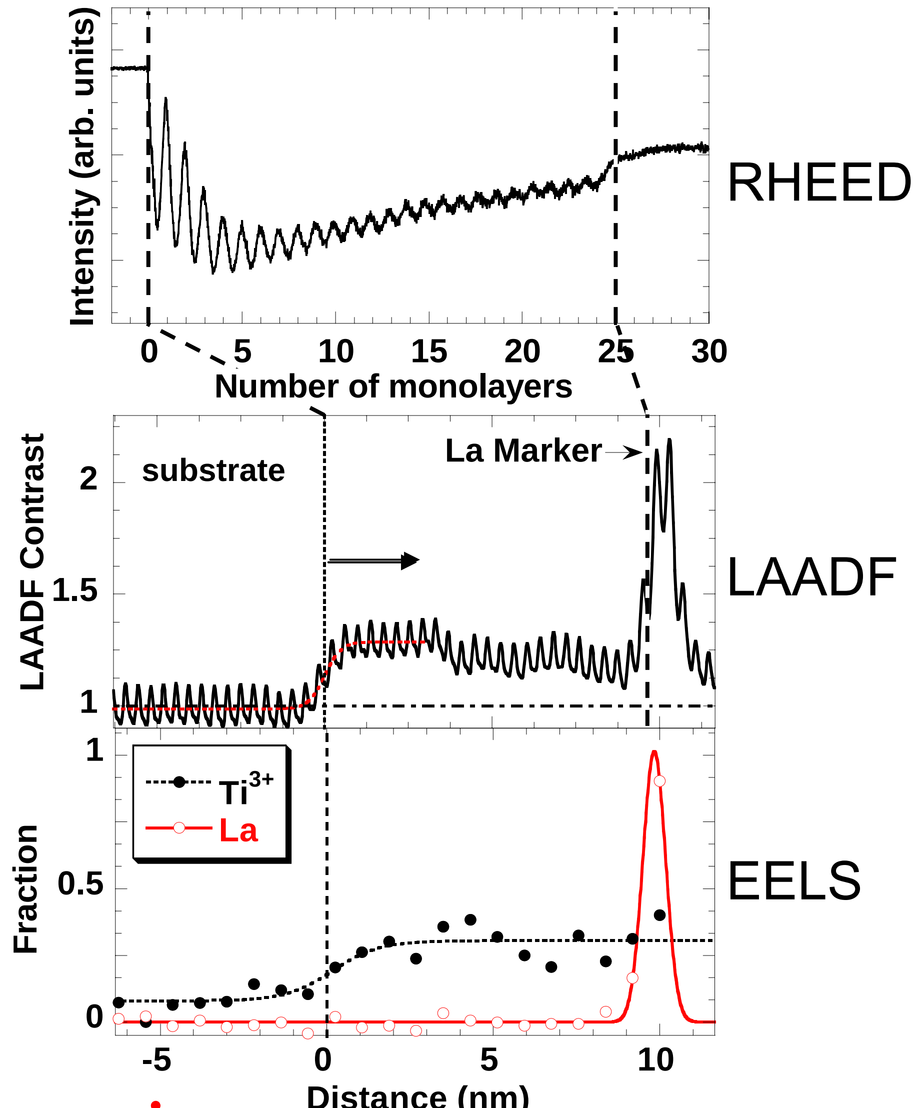
D. A. Muller et al *Nature* **430**, 657-661 (2004).

HAADF
"Z" map

LAADF
"Strain" map
Thin x/s

LAADF
"Strain" map
Thick x/s





Detection sensitivity: 1-4 Oxygen vacancies

Ronchigrams

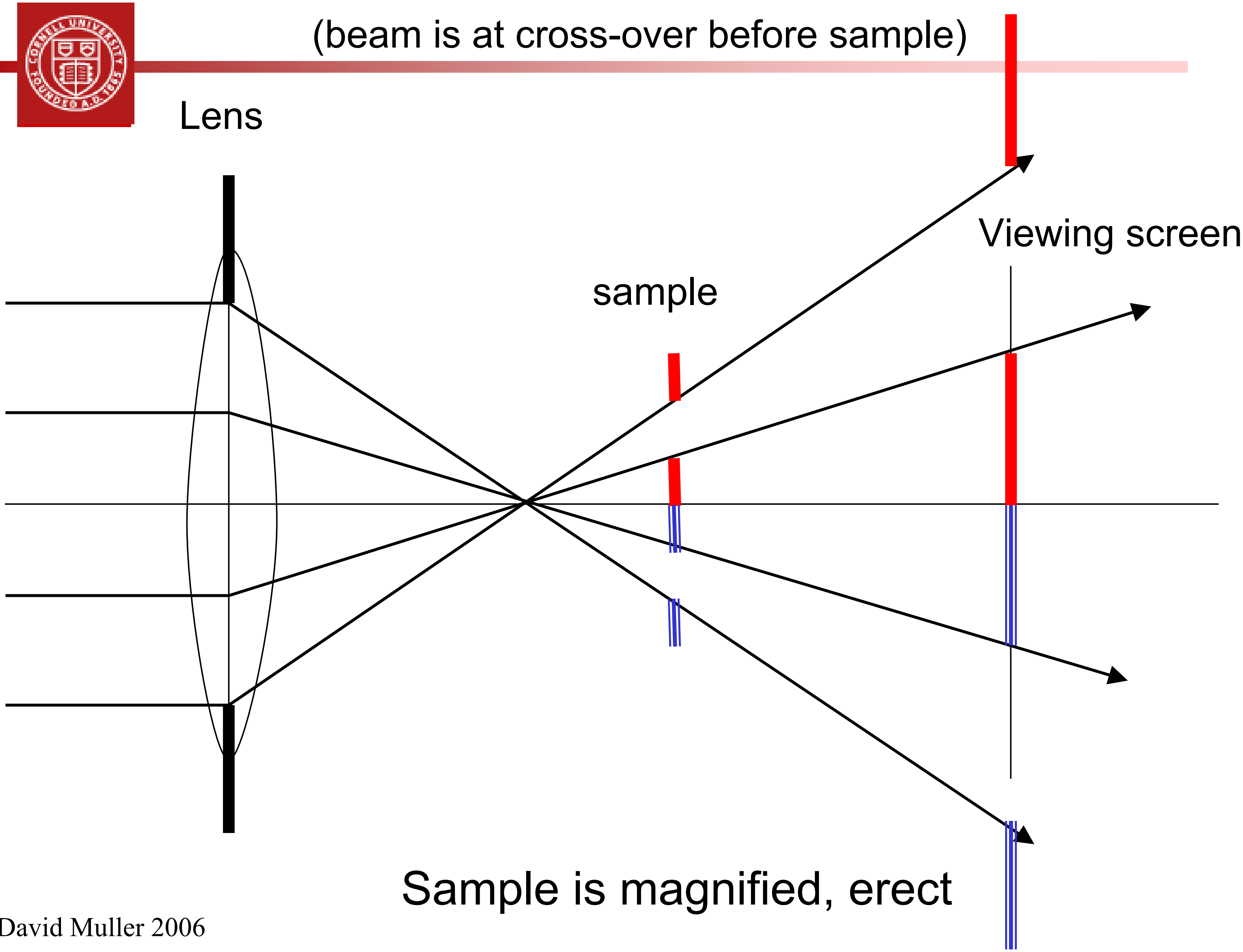
- Most accurate manual method of alignment
- Easy to find the optic axis
- Easy to correct serious astigmatism
- Easy to bring the sample into focus
- Works best on an amorphous layer
- Start with the largest aperture

J. M. Cowley, *Ultramicroscopy* **4,** 413-418.(1979)

E. M. James, N. D. Browning, Ultramicroscopy, 78 (1999) 125-139

Ronchigrams – no C_s

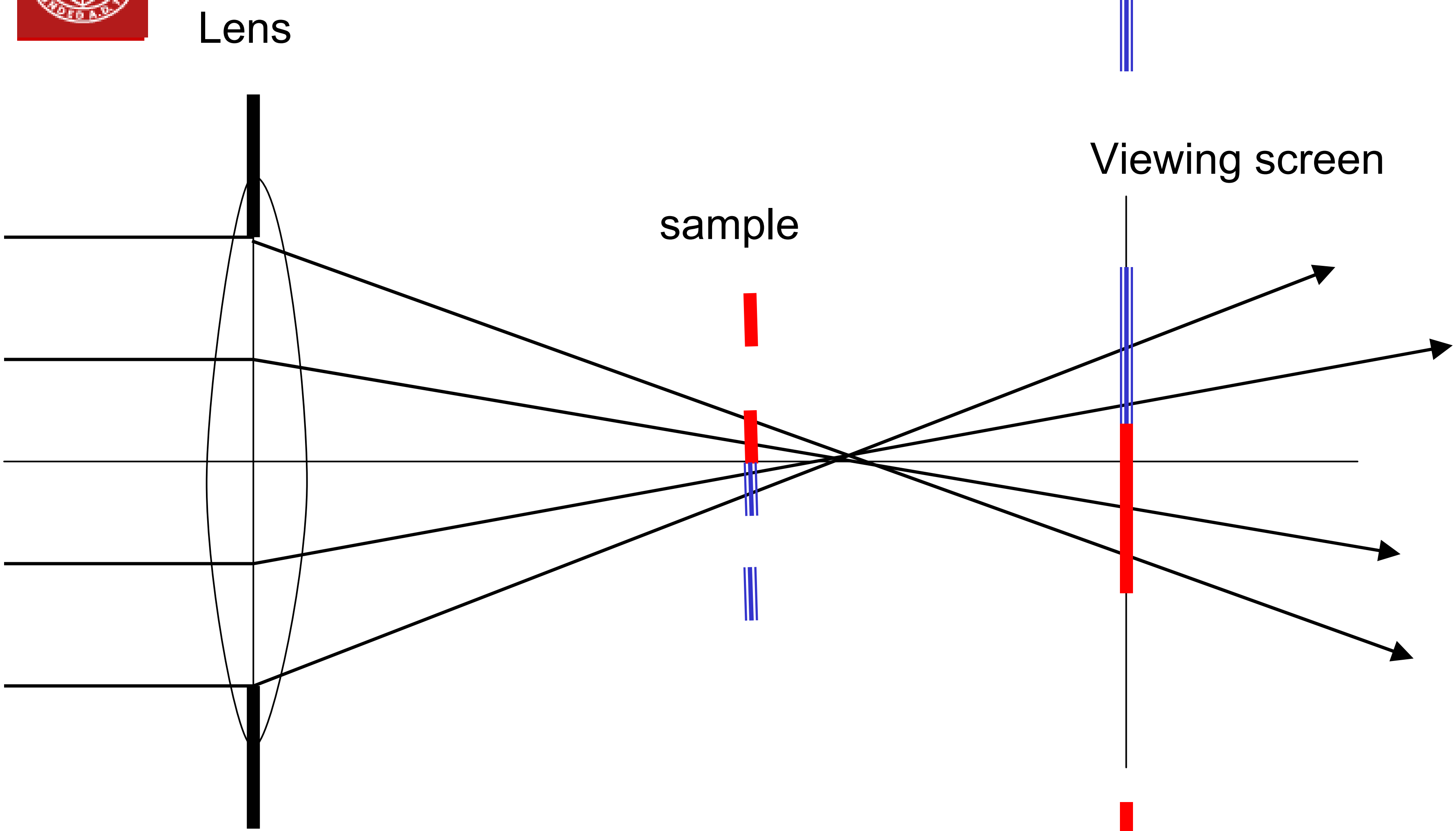
(beam is at cross-over before sample)



Sample is magnified, erect

Ronchigrams – no C_s

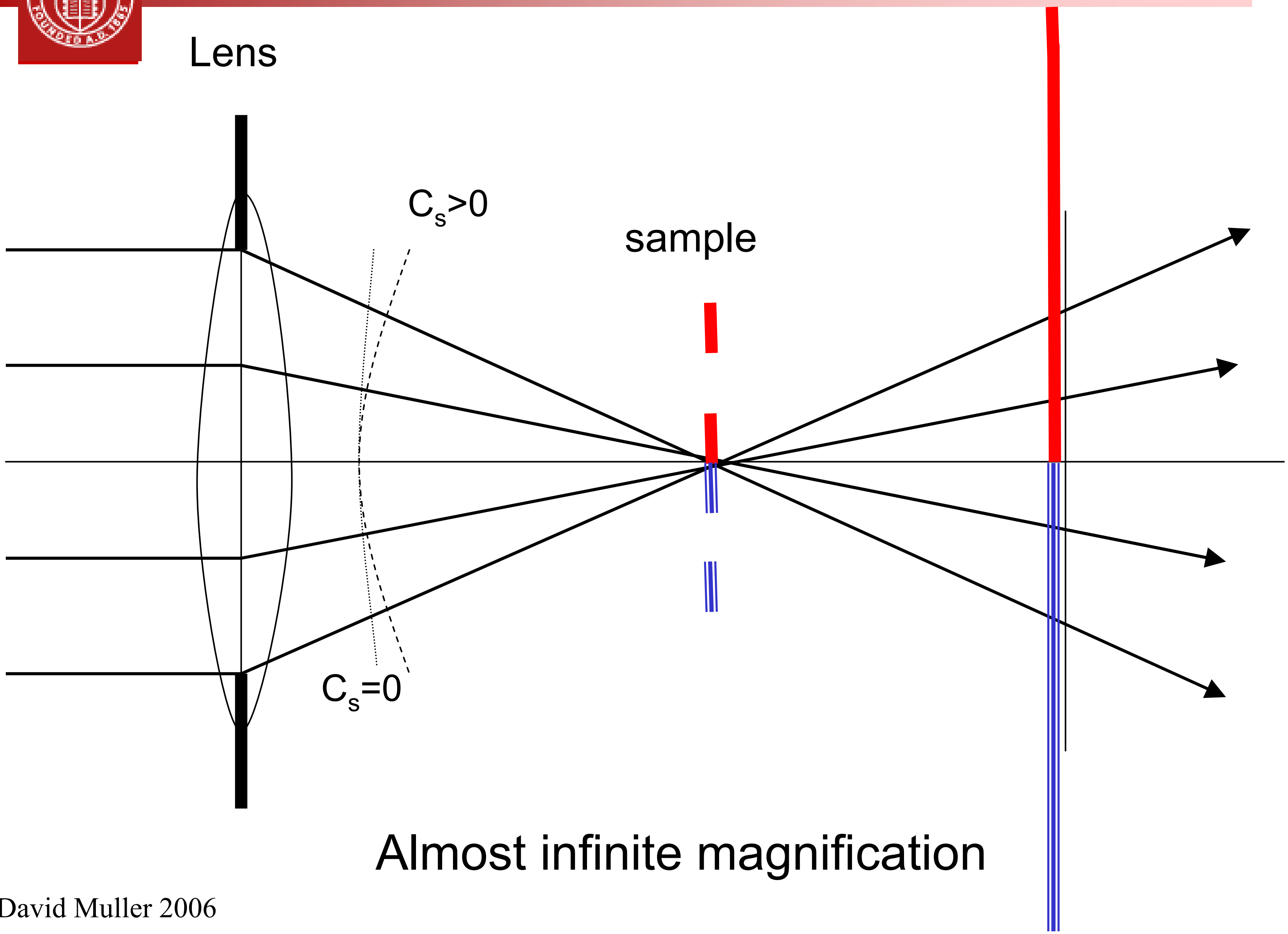
(beam is at cross-over after sample)



Sample is magnified, inverted

Ronchigrams – no C_s

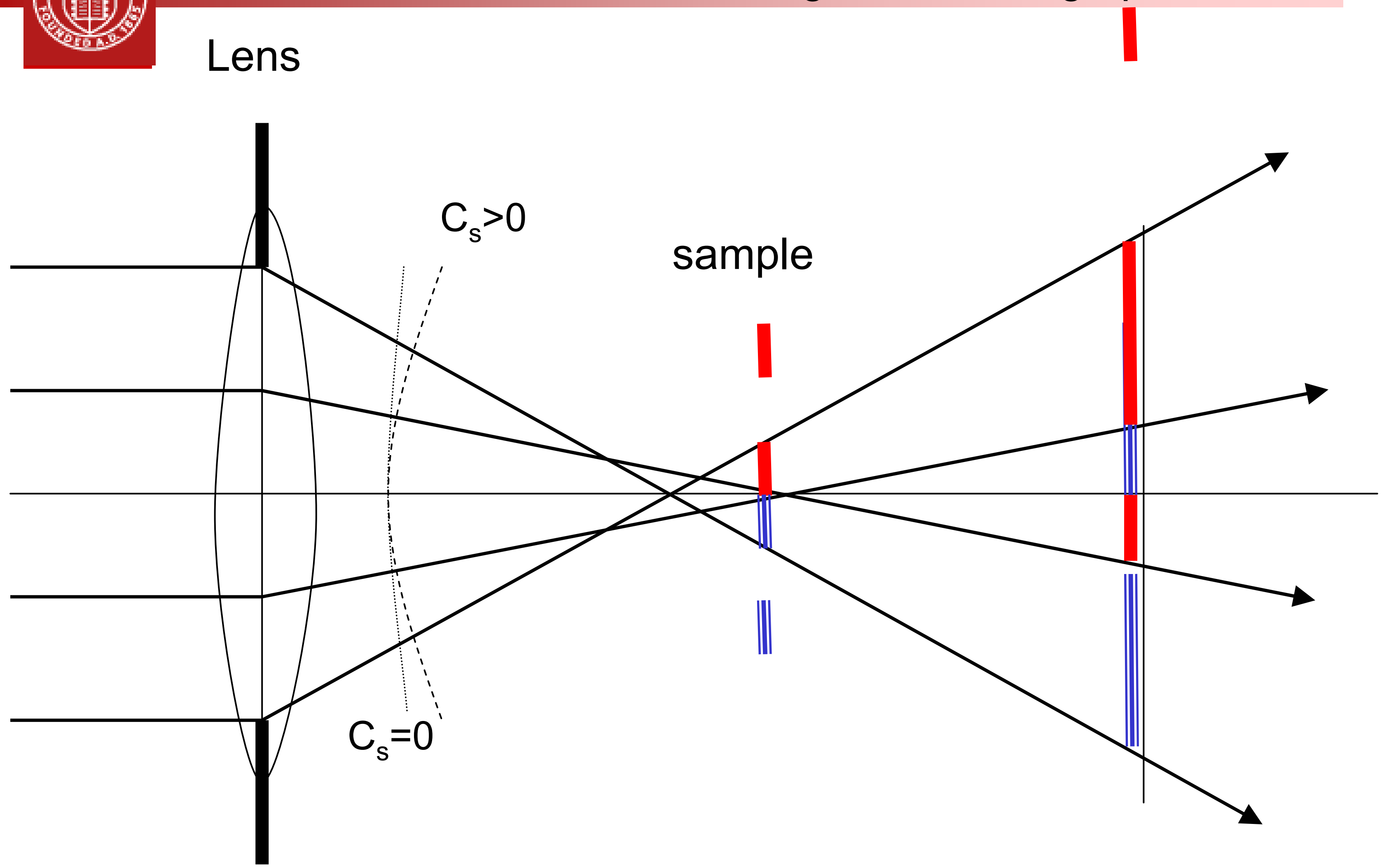
(beam is at almost cross-over on the sample)



Almost infinite magnification

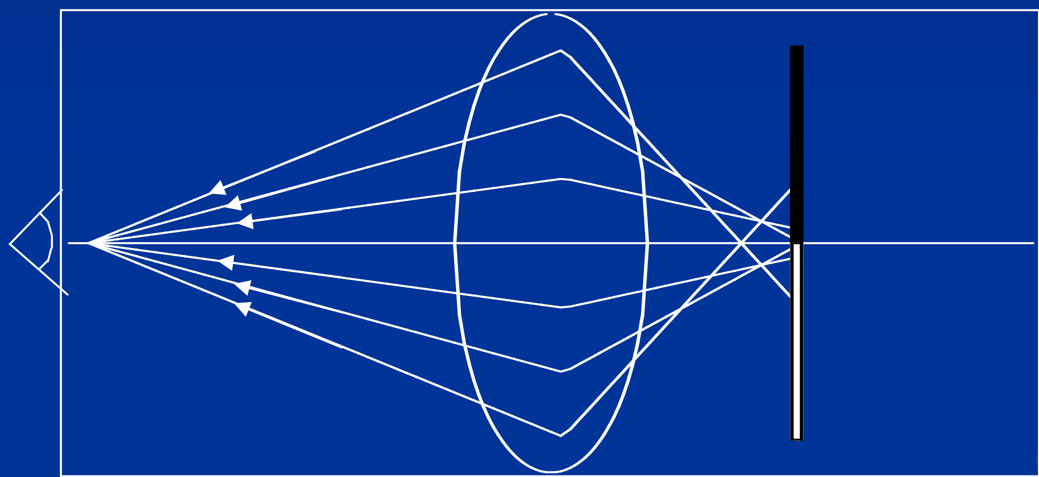
Ronchigrams with Spherical Aberration, C_s

For Cs>0, rays far from the axis are bent too strongly and come to a crossover before the gaussian image plane.



Almost infinite magnification,
Only at small angles

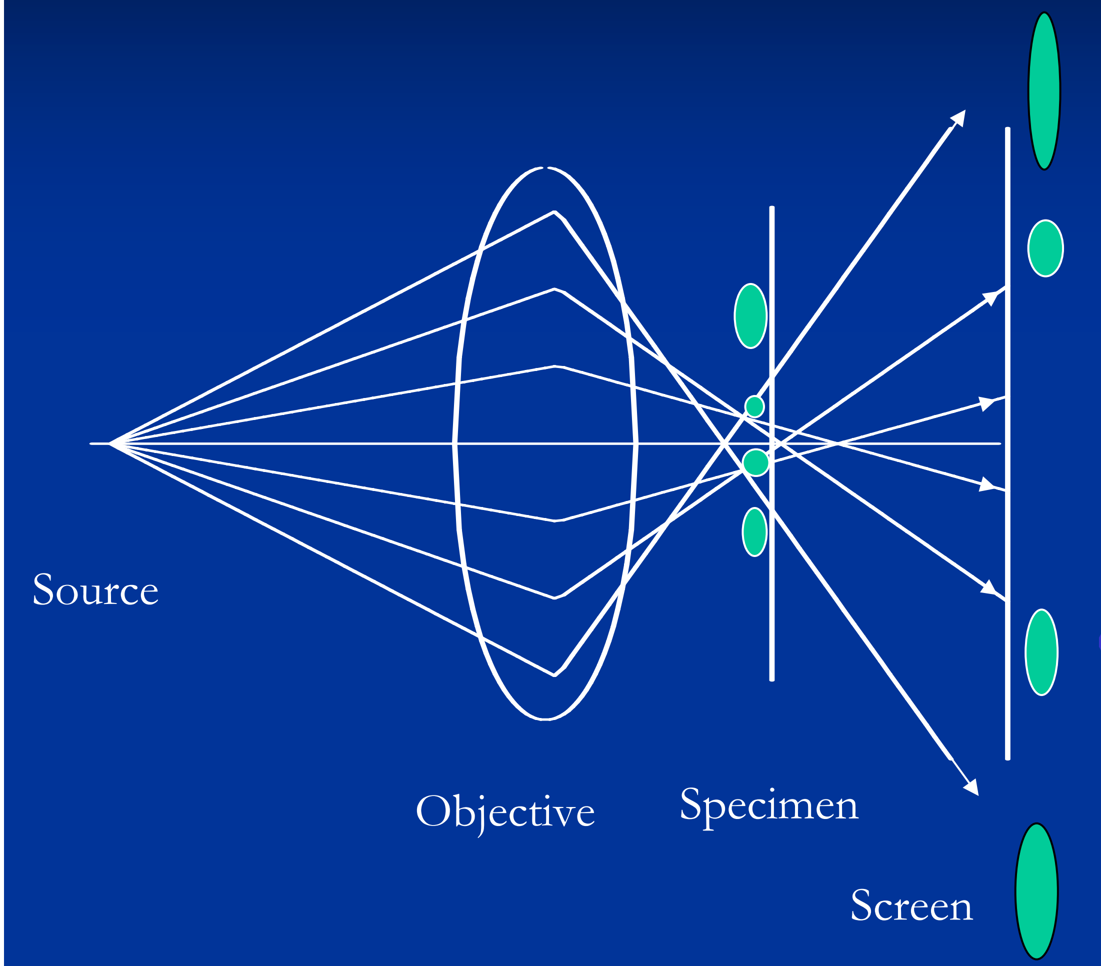
Spherical Aberration

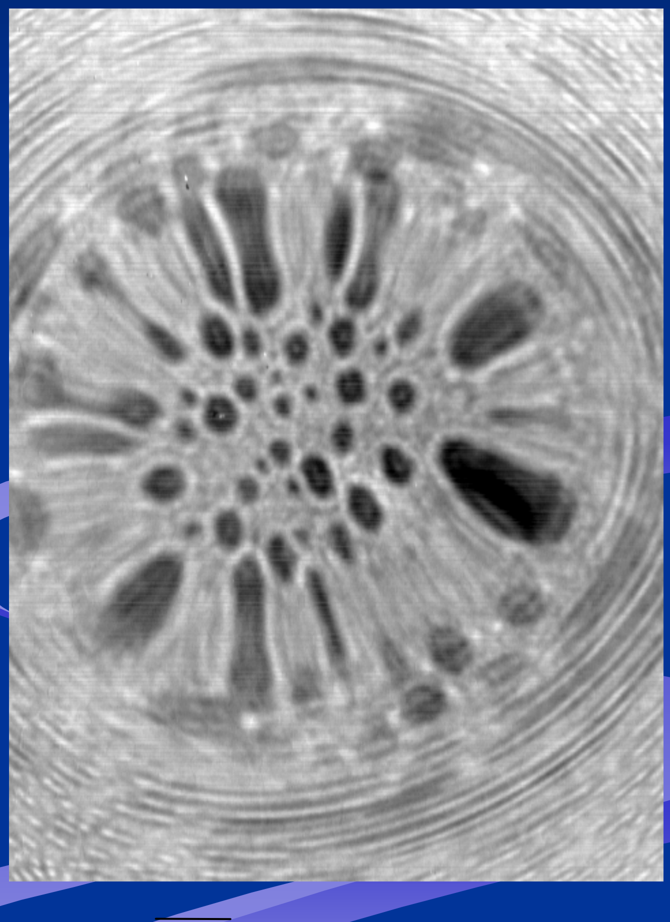


Apparent deflection at the object is proportional to the cube of the distance off-axis within the imaging lens. Deflection towards axis is always too strong.

Phil Batson, IBM

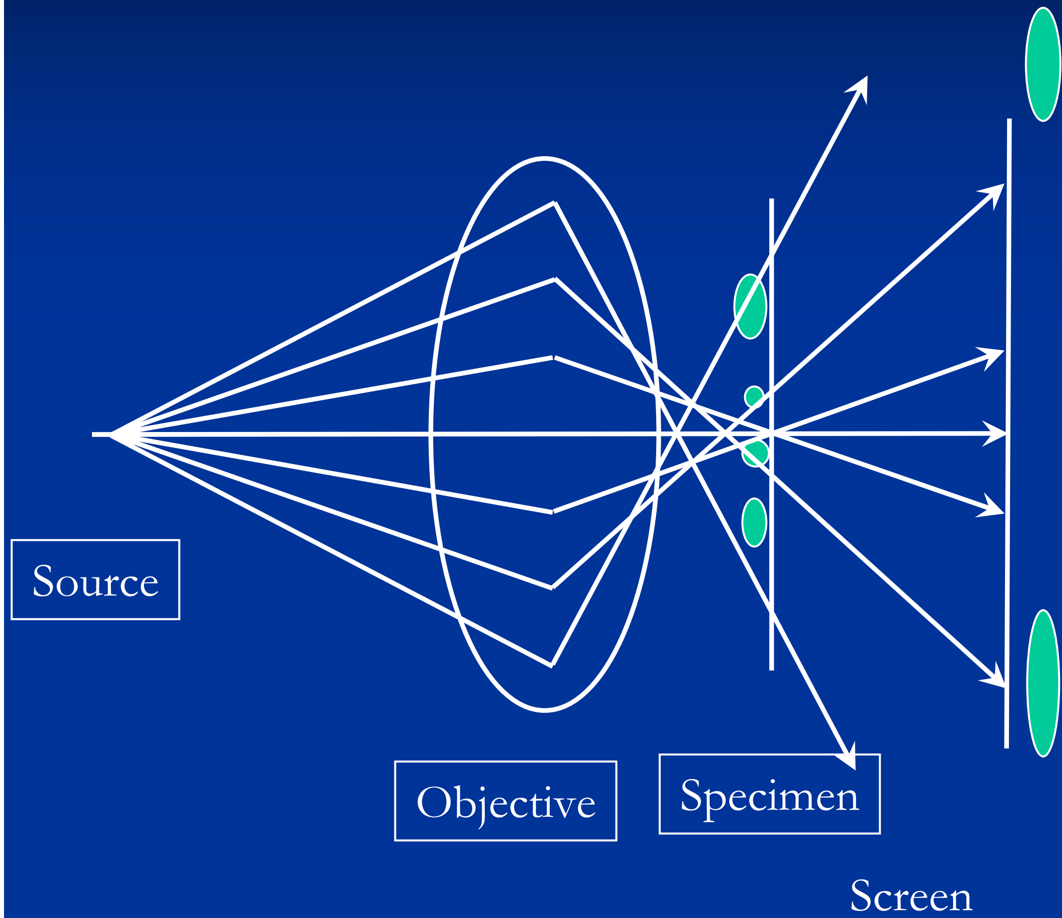
Shadow Map: "Ronchigram"

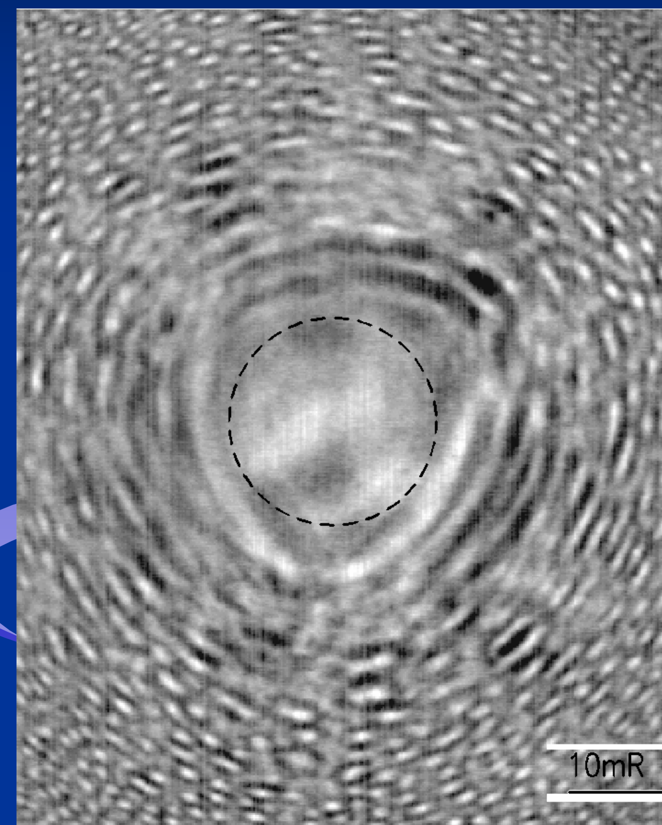




Ronchi, V. (1964) Forty years of history of a grating interferometer. Applied Optics 3, 437 – 450. Phil Batson, IBM

Shadow Map: "Ronchigram"

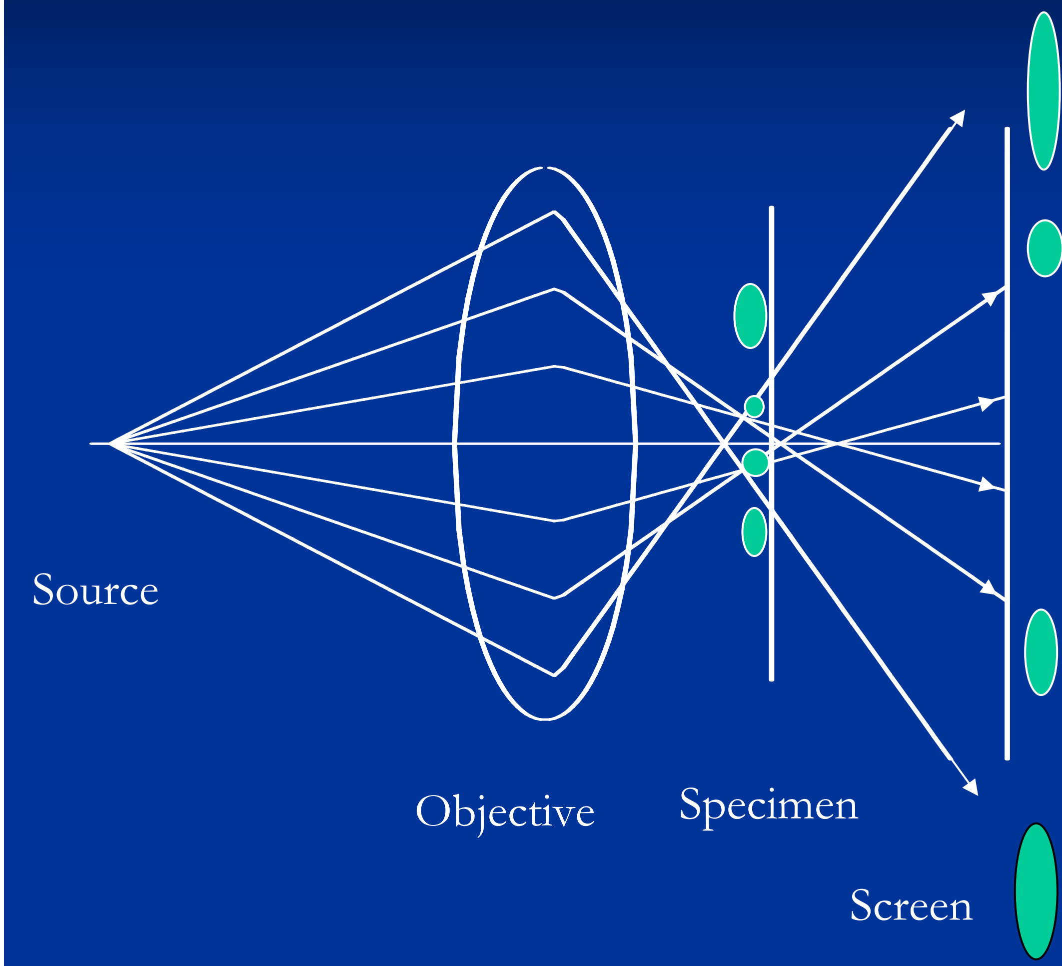


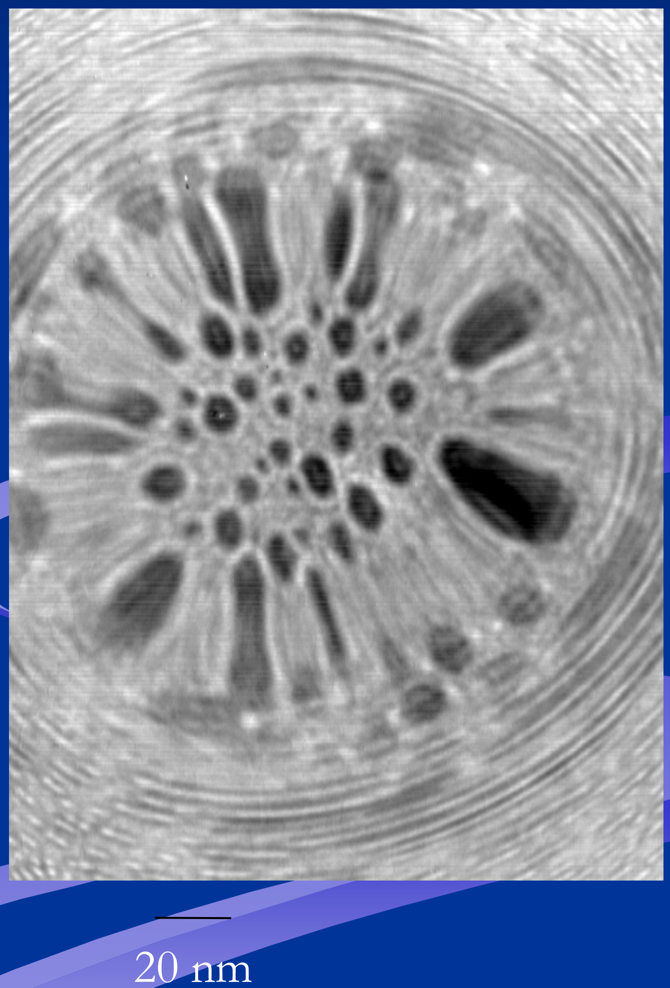


20 nm

Ronchi, V. (1964) Forty years of history of a grating interferometer. Applied Optics 3, 437 – 450. Phil Batson, IBM

Shadow Map: "Ronchigram"





Ronchi, V. (1964) Forty years of history of a grating interferometer. Applied Optics 3, 437 – 450. Phil Batson, IBM

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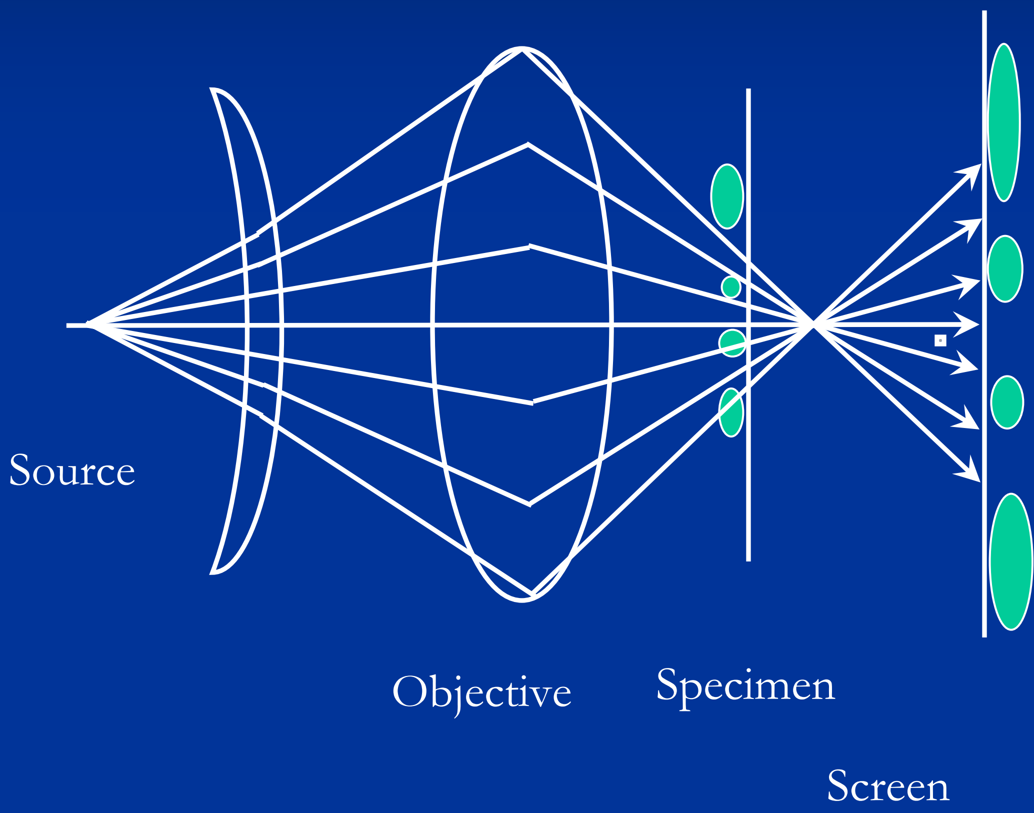
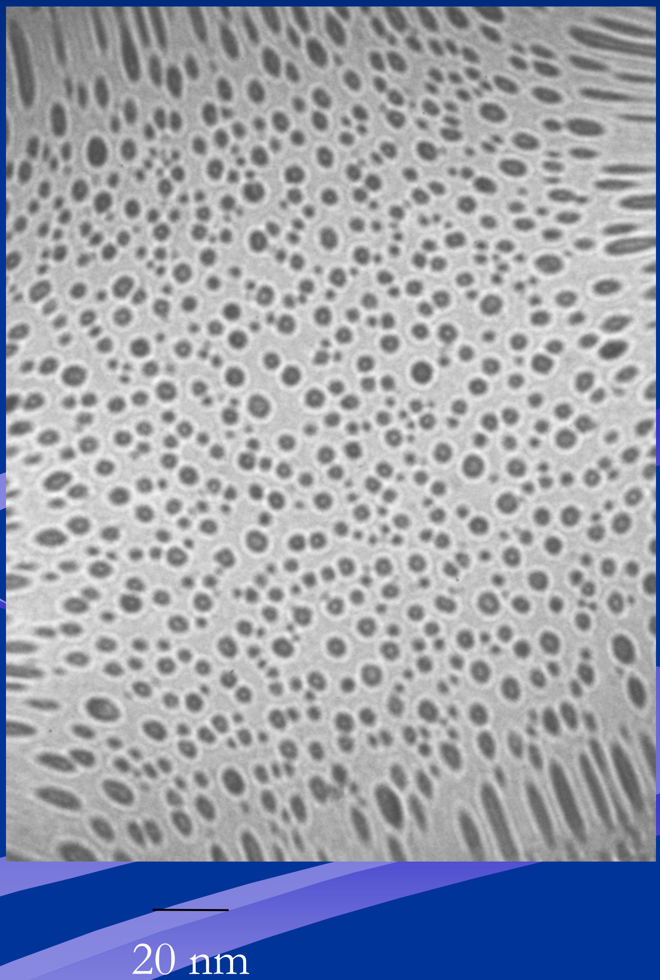
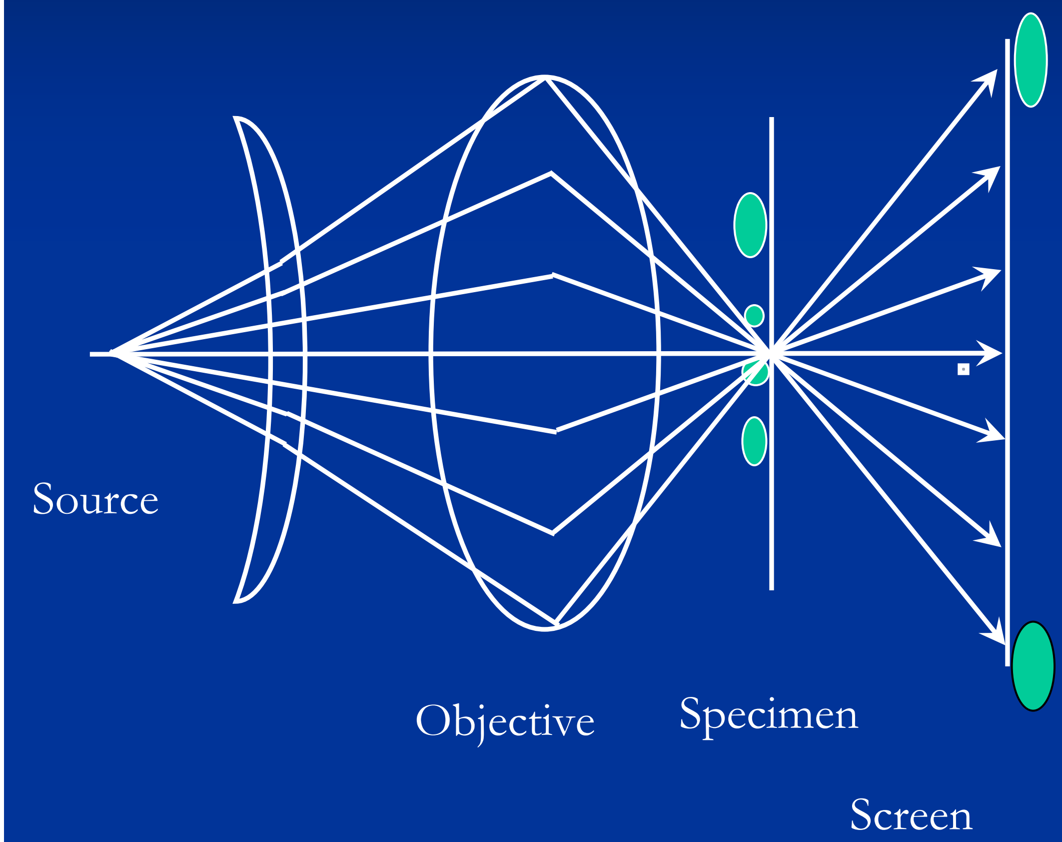
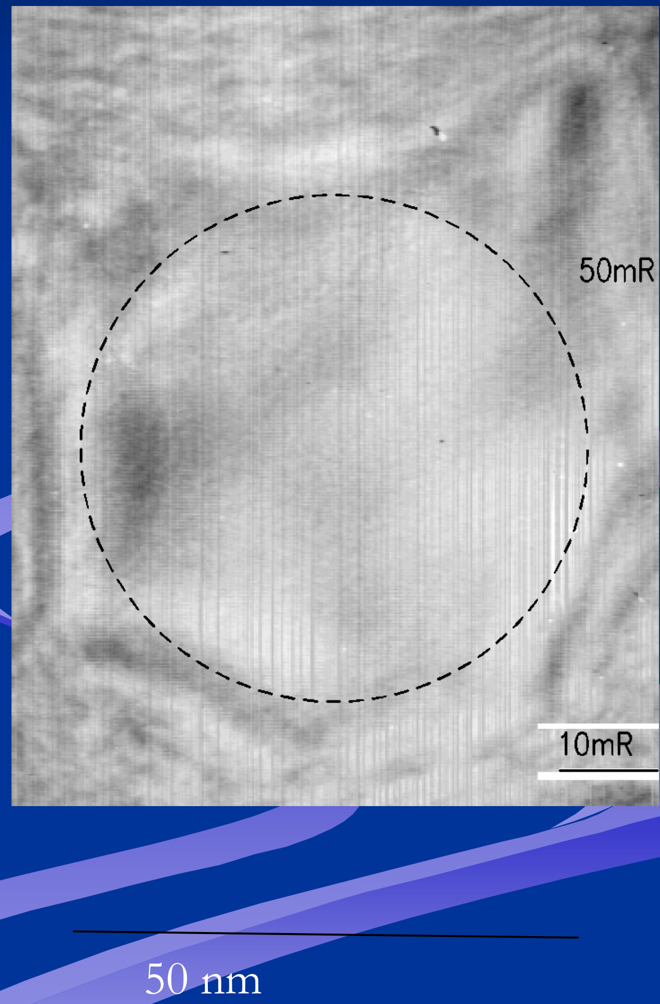




Ronchi, V. (1964) Forty years of history of a grating interferometer. Applied Optics 3, 437 – 450. Phil Batson, IBM

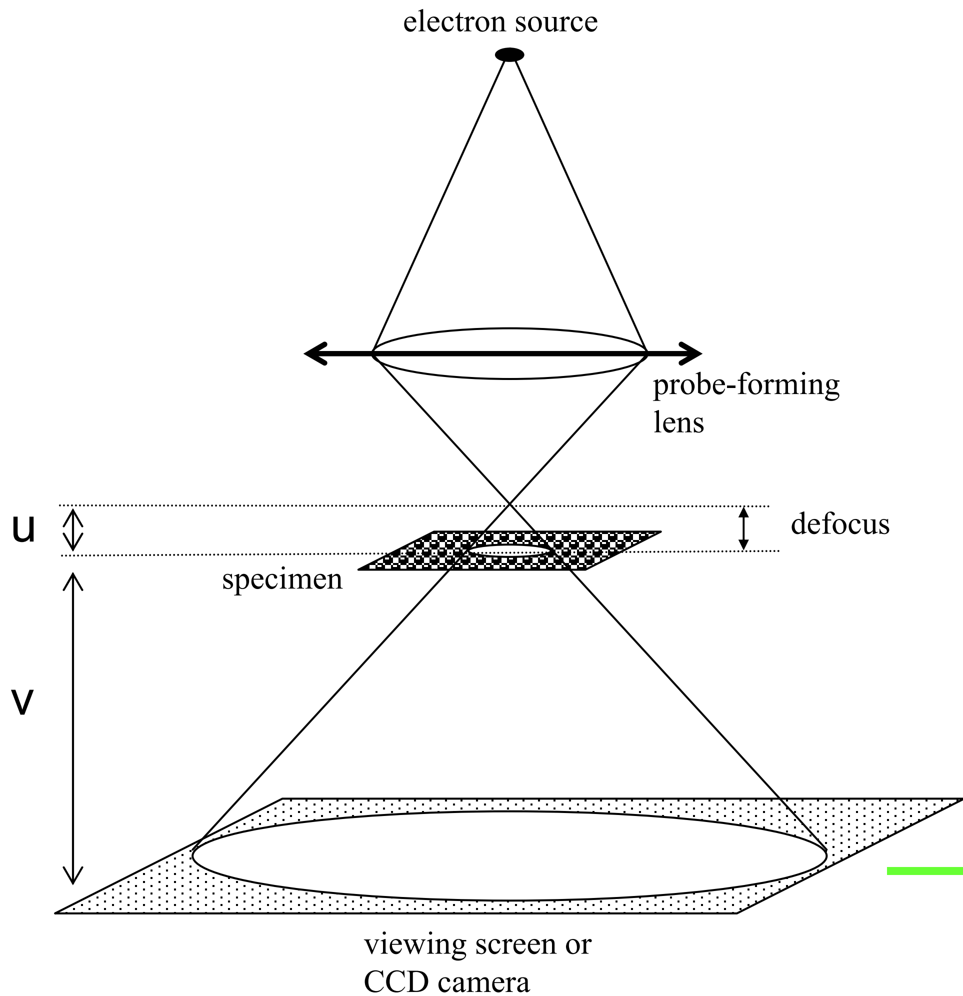
Shadow Map: "Ronchigram"

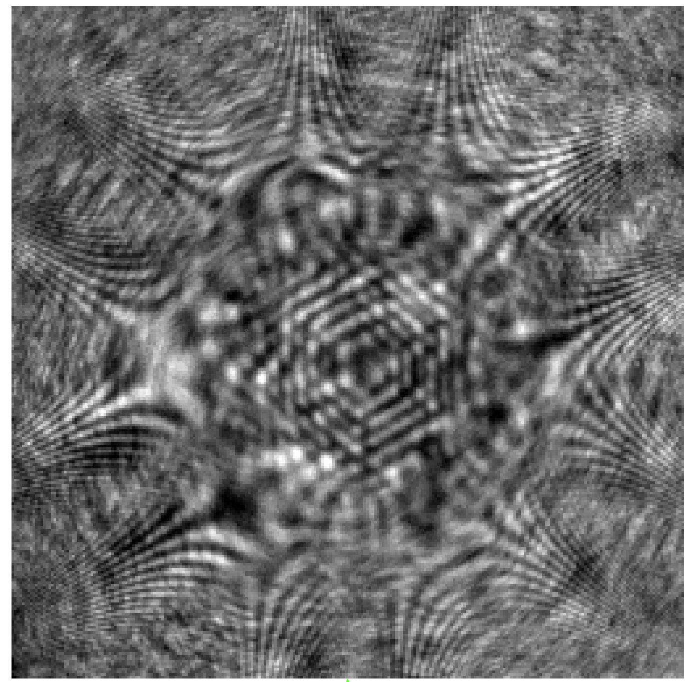




Ronchi, V. (1964) Forty years of history of a grating interferometer. Applied Optics 3, 437 – 450. Phil Batson, IBM

The Electron Ronchigram



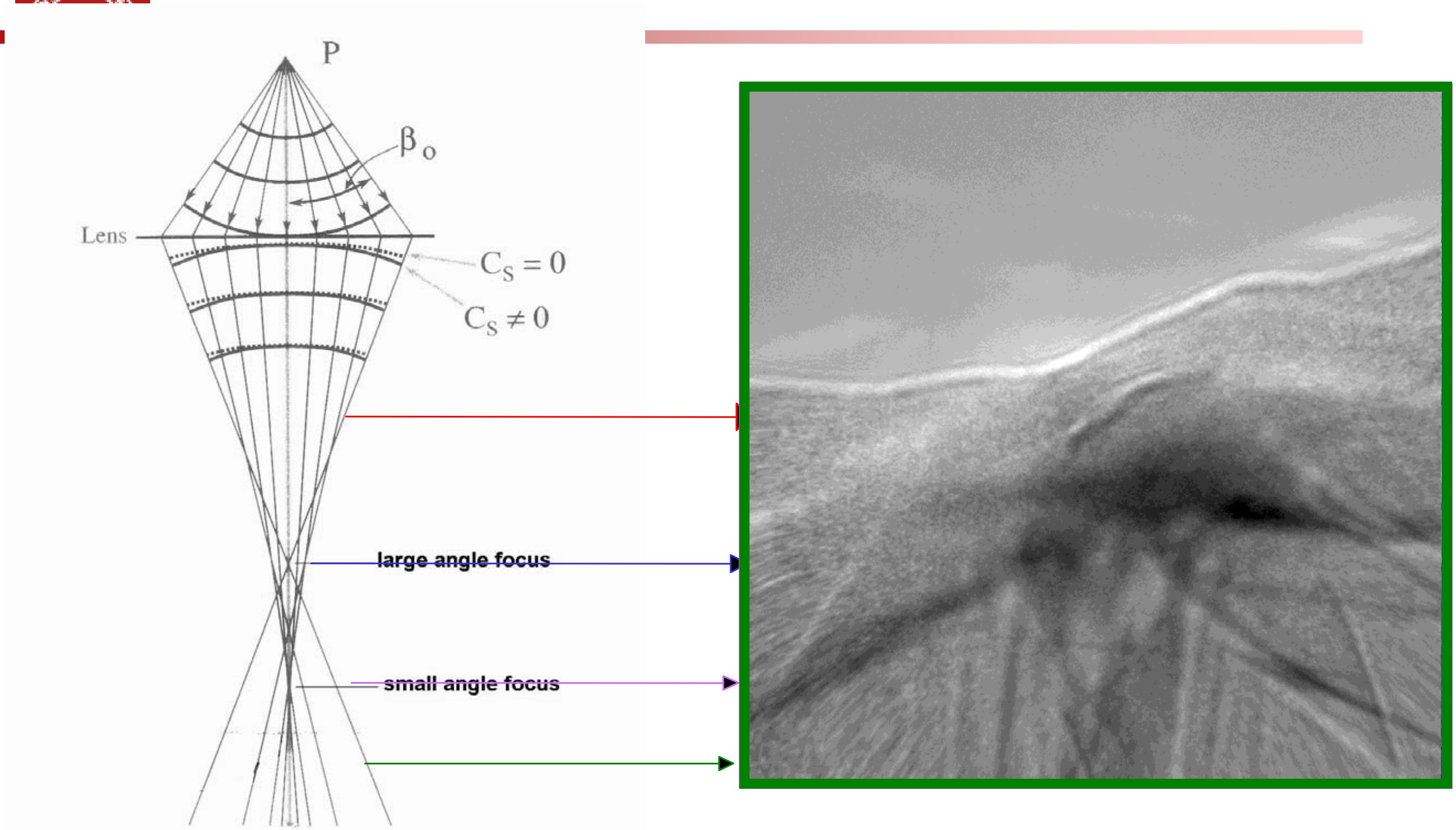


$$M = \frac{v}{u}$$

Cowley, *J. Elec Microsc Tech* **3**, 25 (1986)

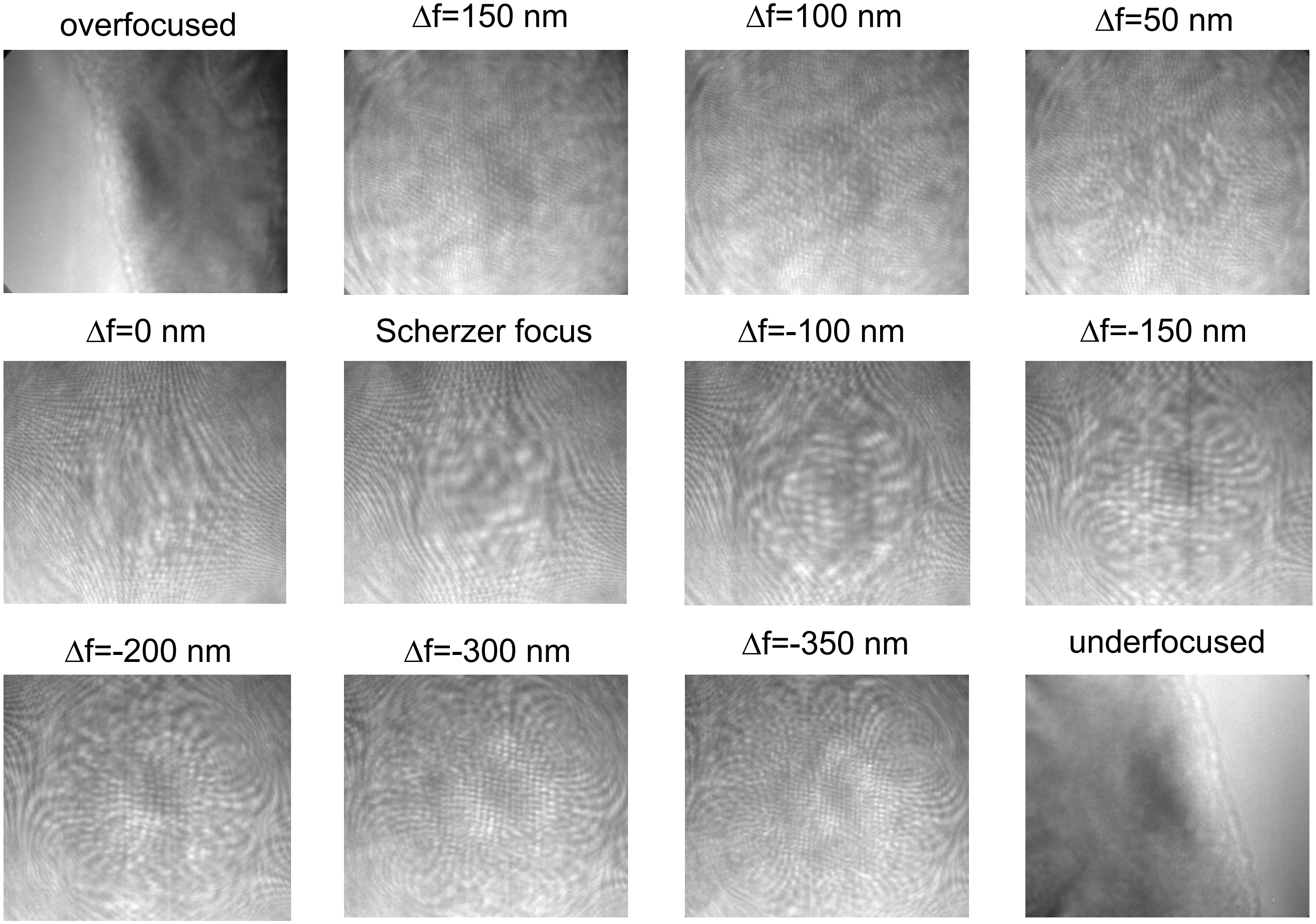
Nigel Browning, UC Davis

Effect of C_s on Ronchigram



James and Browning, *Ultramicroscopy* **78**, 125 (1999)

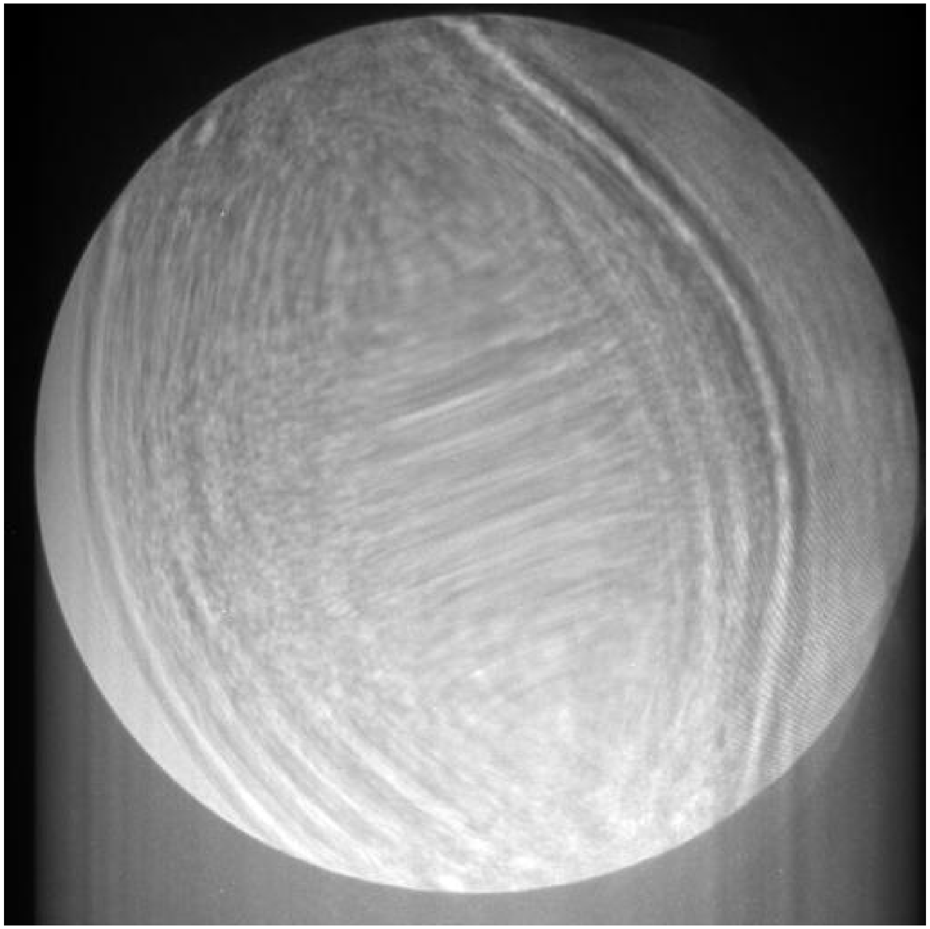
Ronchigrams from Si <110>



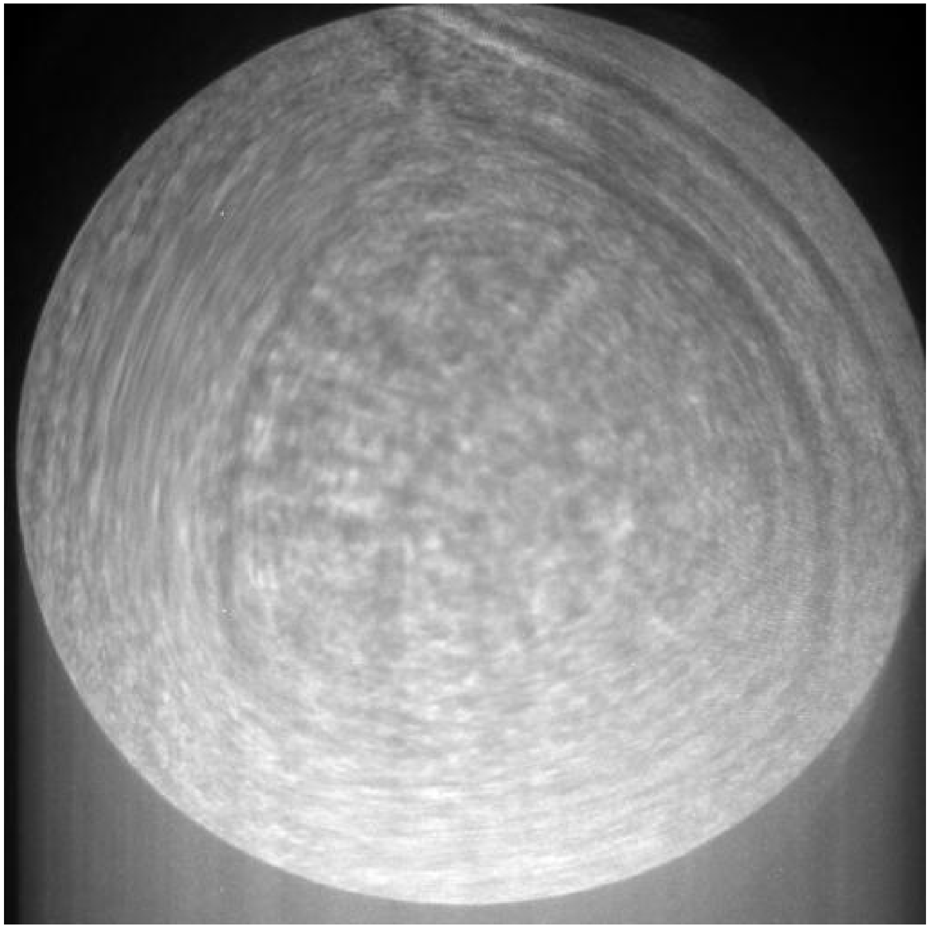
Nigel Browning, UC Davis

Correcting for Astigmatism

Two fold astigmatism

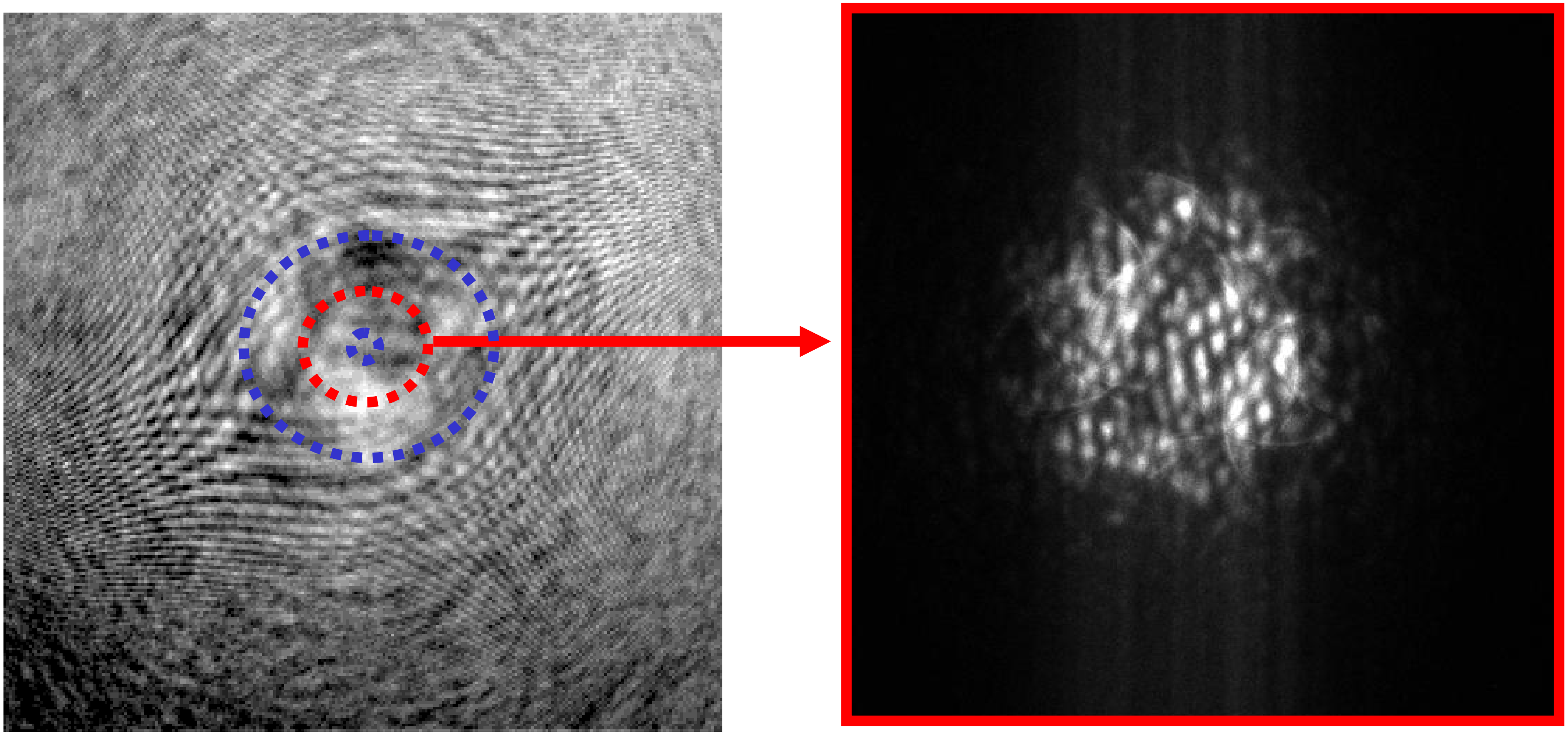


Three fold astigmatism



Nigel Browning, UC Davis

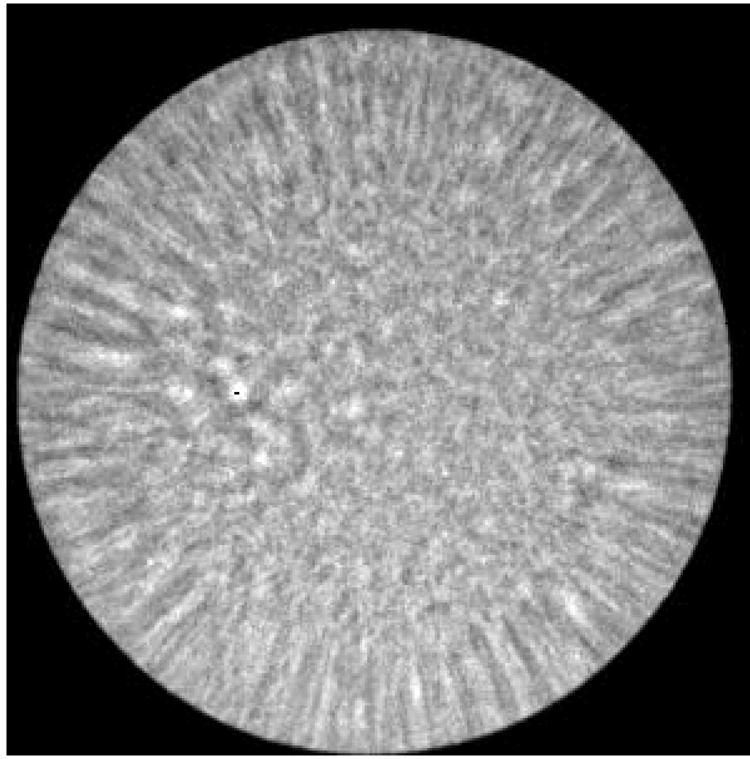
Forming the Smallest Probe



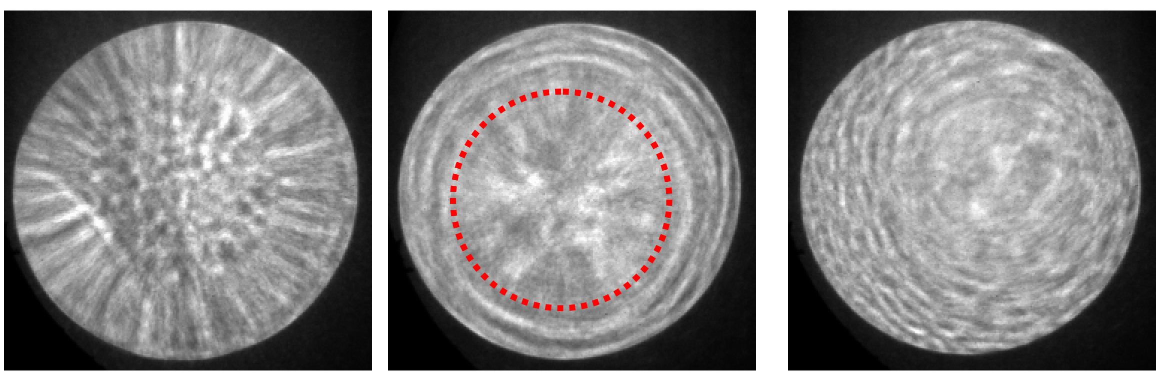
Put aperture over area of constant phase in Ronchigram to give CBED pattern

Nigel Browning, UC Davis

Ronchigram focus series on a-C

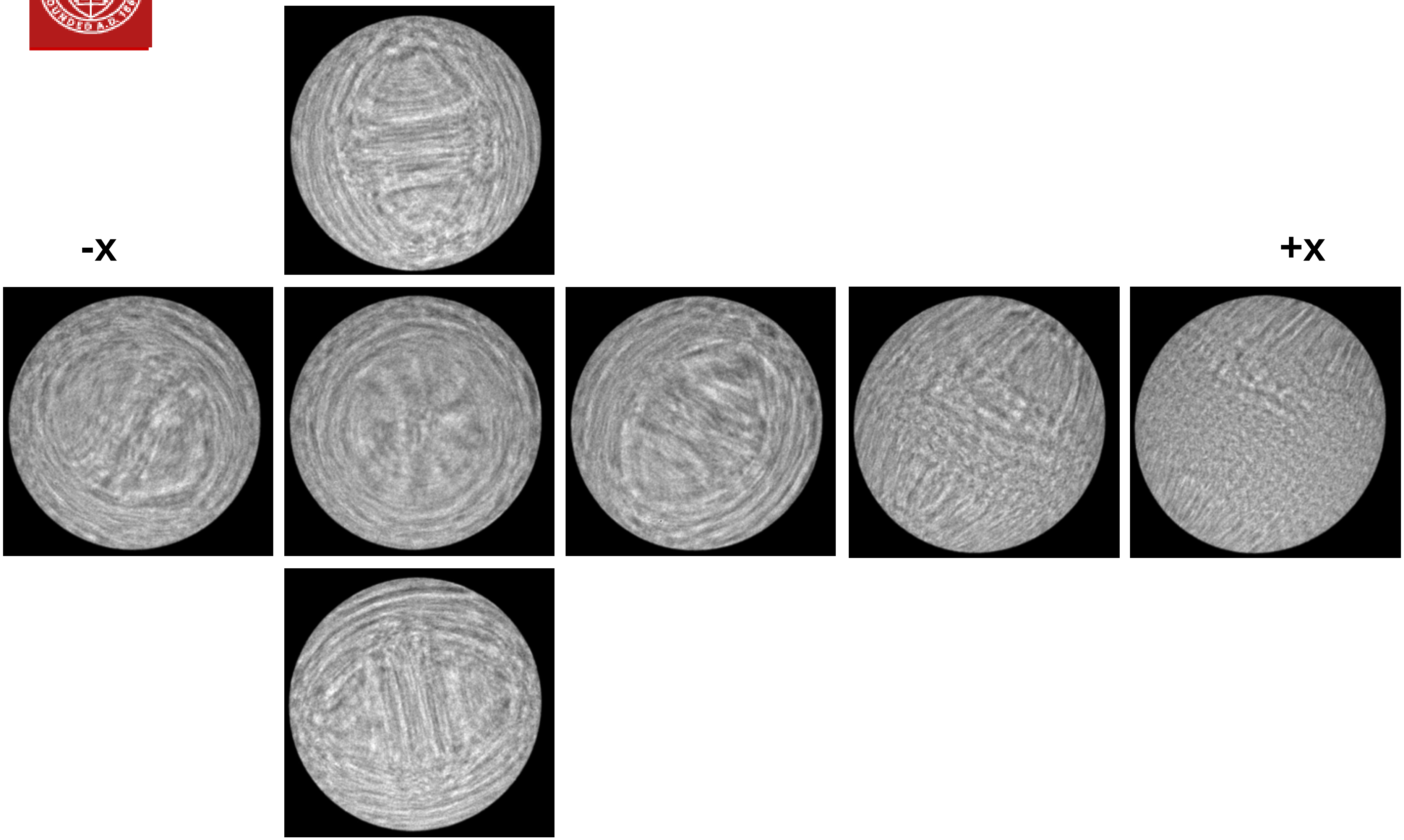


This aperture
Is too big



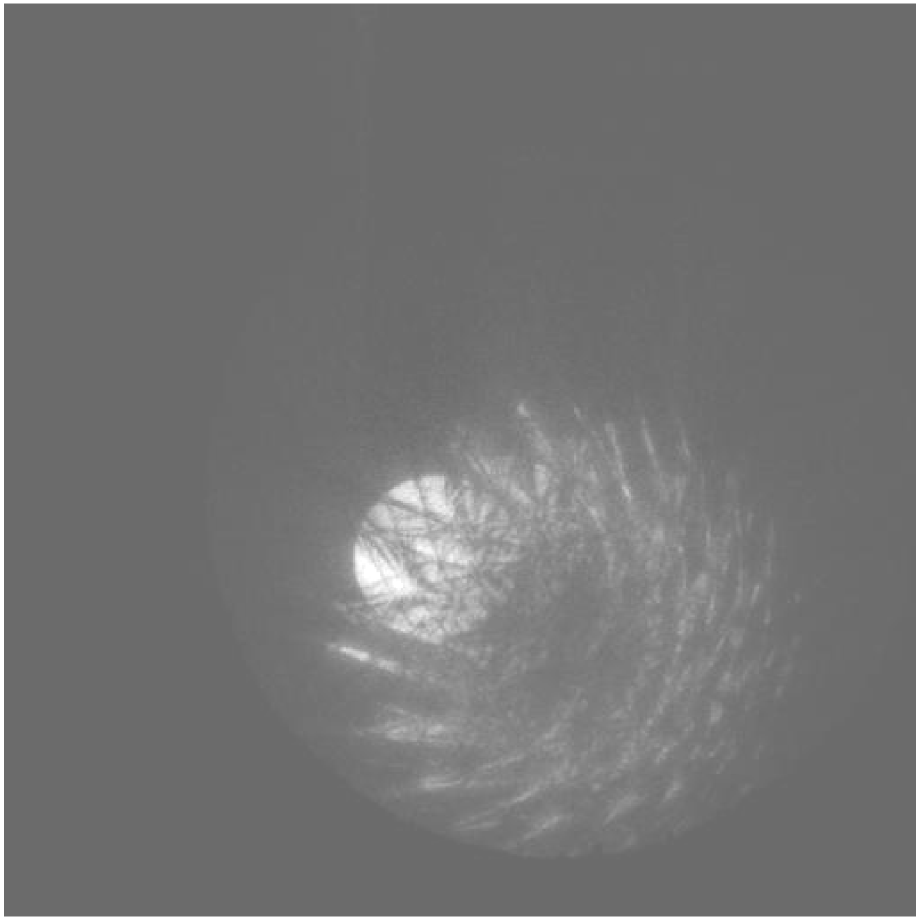
Correcting Severe Astigmatism in Ronchigrams

(don't image probe)

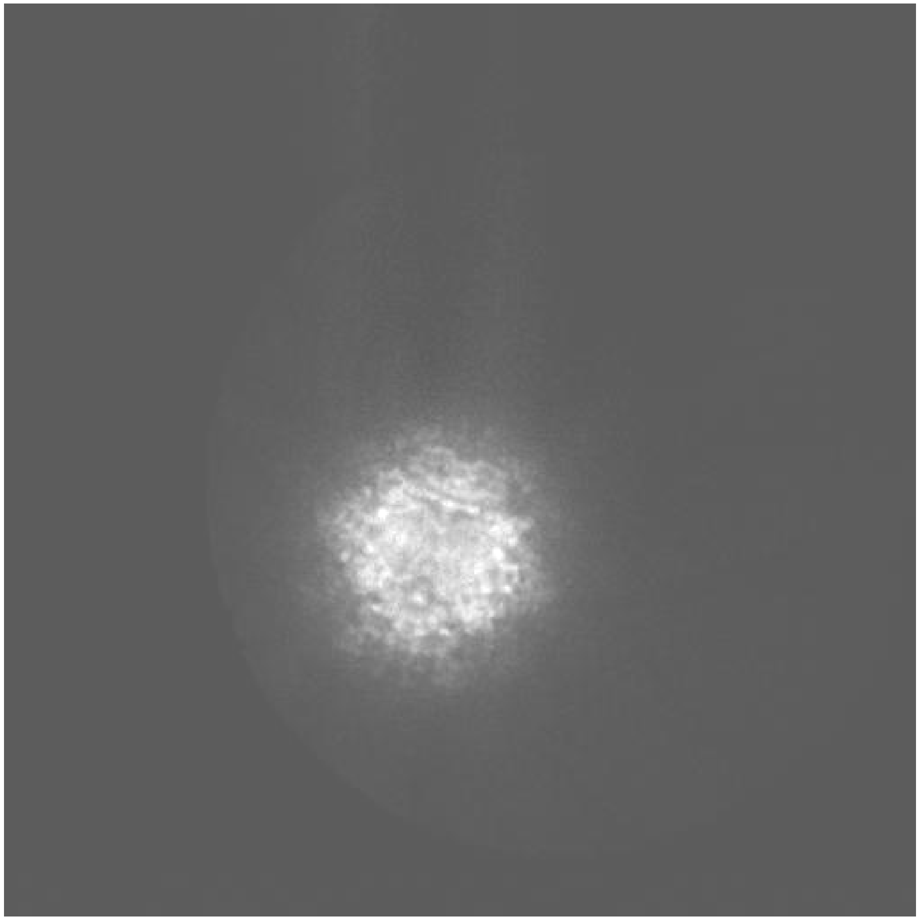


Ronchigrams on Crystals

Tilted

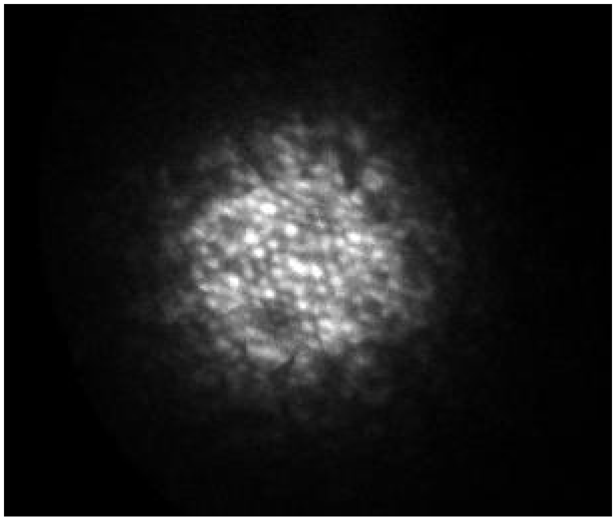


On Axis (not in focus)

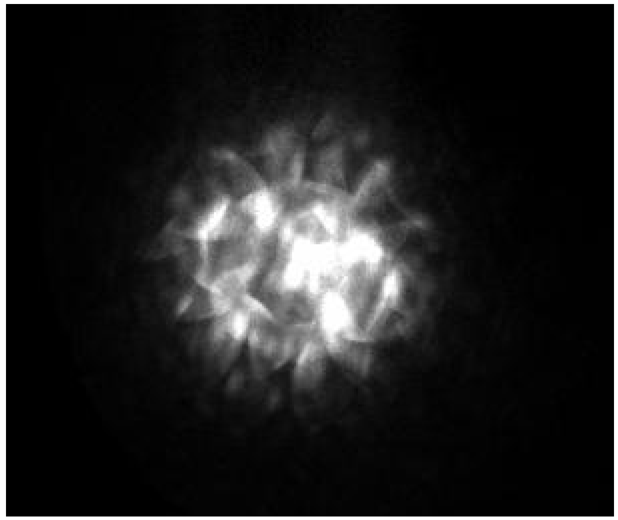


Ronchigrams on Crystals

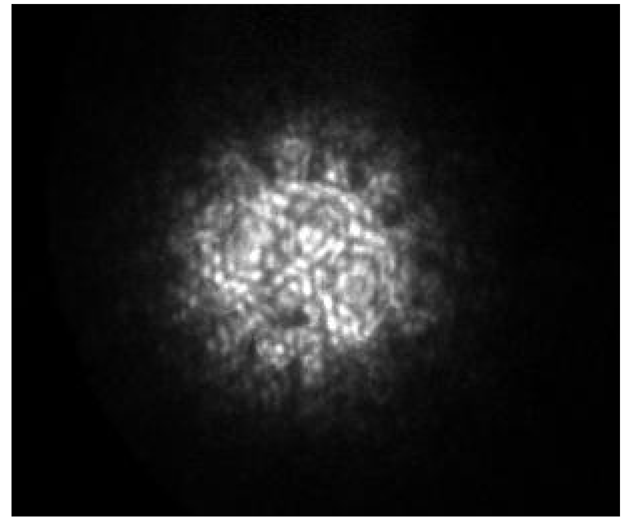
under



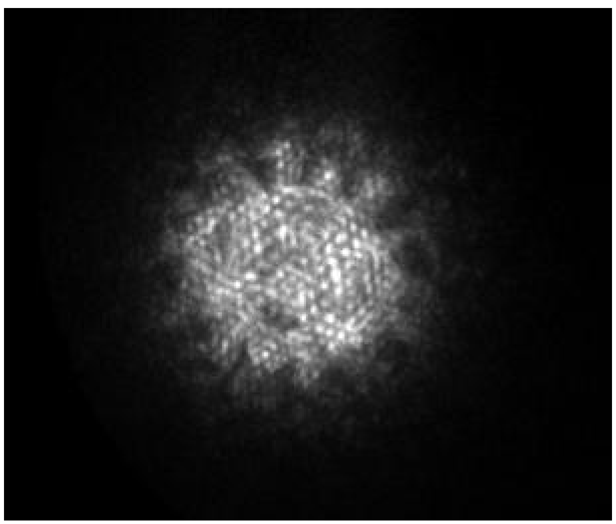
In focus



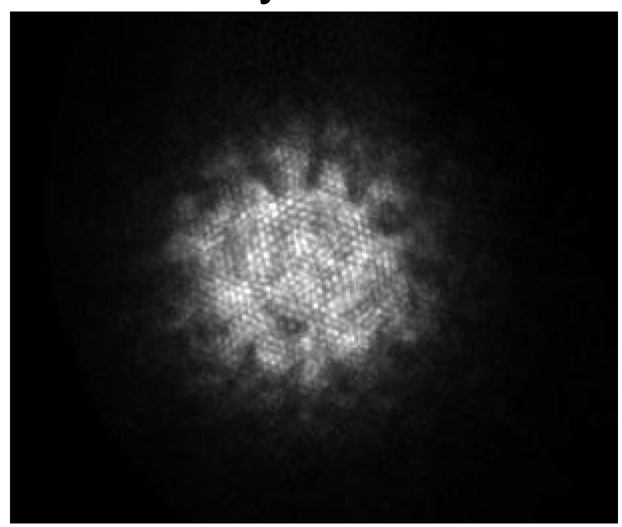
over



More over



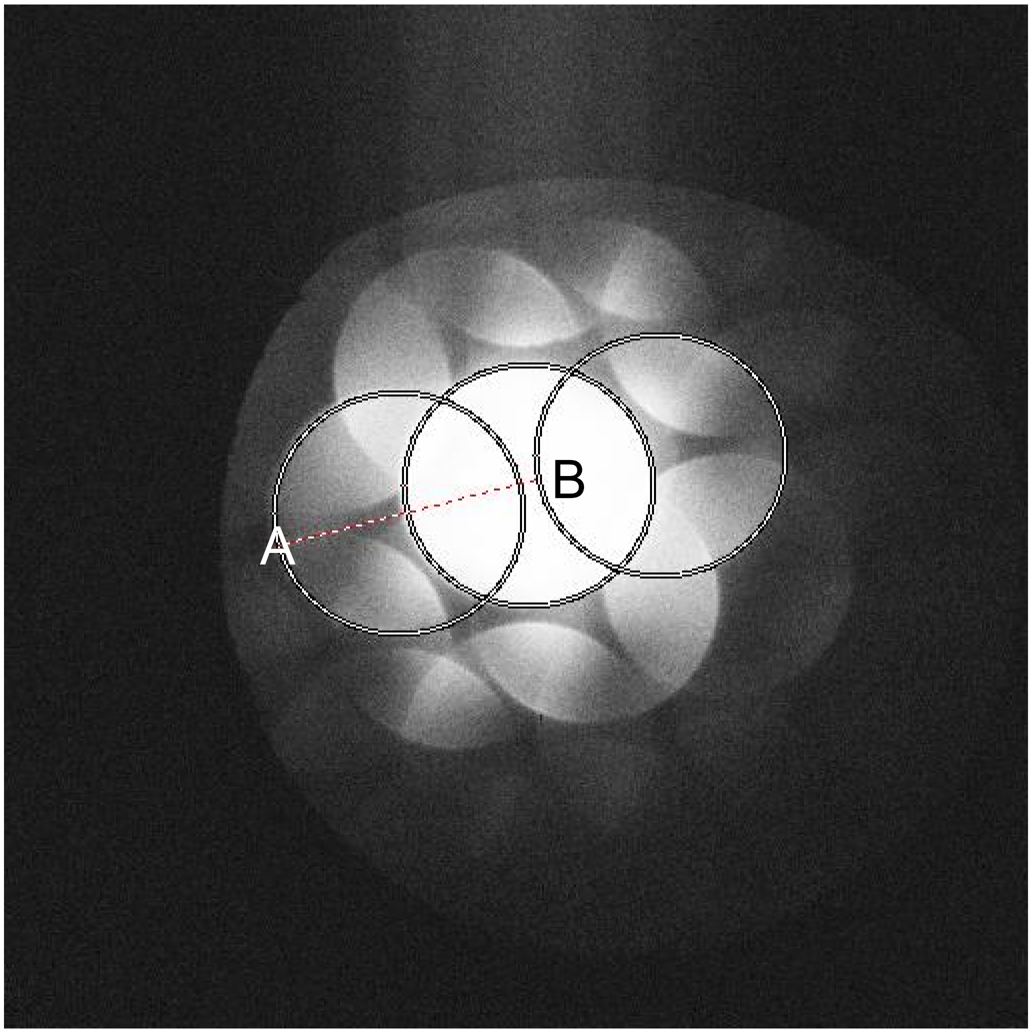
way over



Measuring the Aperture Size

(Using [110] Silicon as a reference)

Scan the beam over a small area to remove ronchigram structure



Distance AB
Is 4 x Bragg Angle

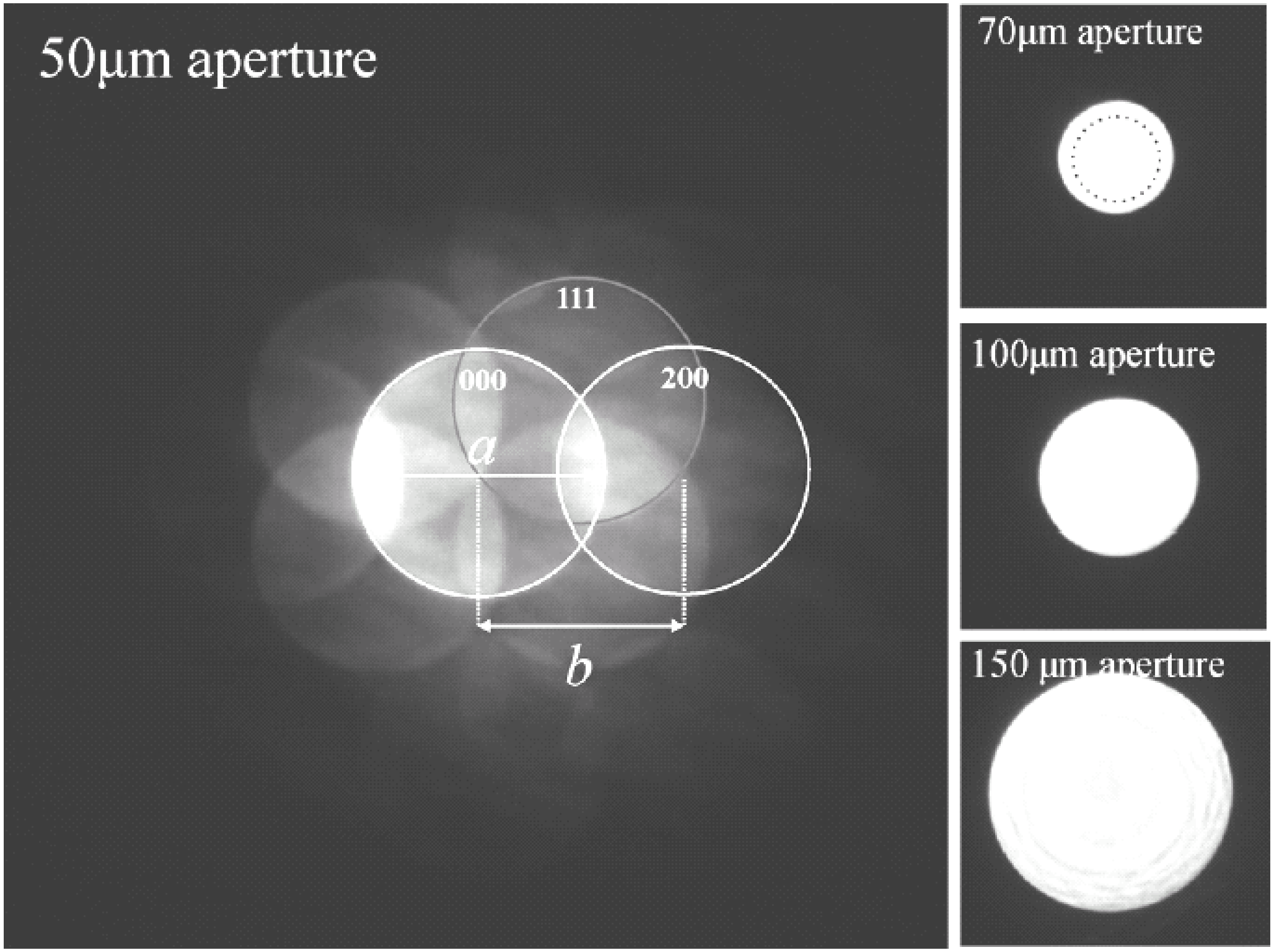


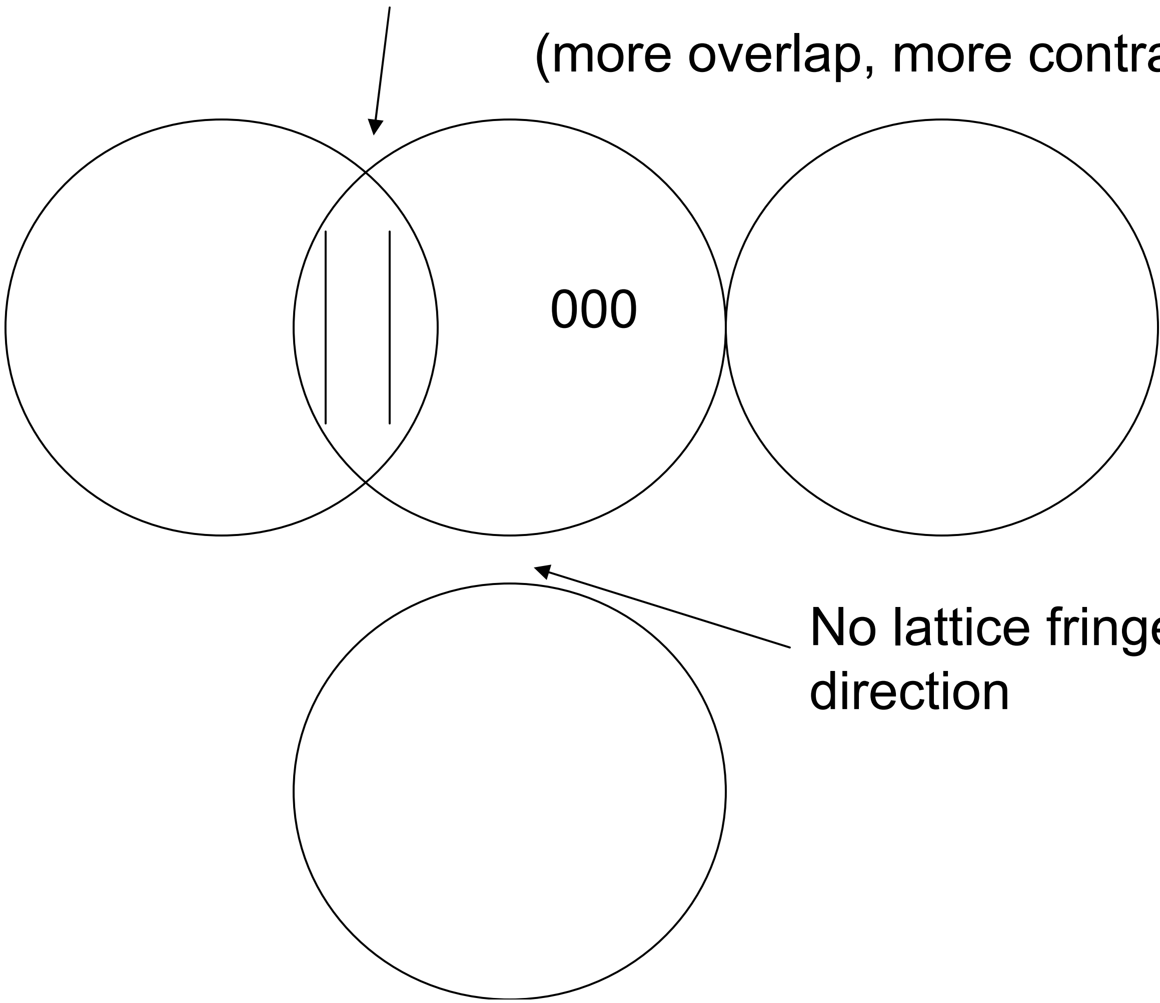
Figure 10. Measurement of STEM convergence angles in an FEI F20 SuperTWIN. The diffraction pattern is "calibrated" on the the 200 reflection of Si, oriented onto to the 110 axis. The convergence semi-angle (α) is proportional to the ratio of the disc width to the disc spacing (a/b). As b is independent of the chosen aperture the other three apertures can be calibrated by just recording the width of the zero order disk (a). The dotted line inside the 50µm aperture represents the relative scale of the 50 µm aperture.

Reality Check

(Can I see a lattice spacing)

Disks must overlap to form a lattice fringe

(more overlap, more contrast)*



No lattice fringes this direction

Balancing Spherical Aberration against the Diffraction Limit

(Less diffraction with a large aperture – must be balanced against C_s)

A more accurate wave-optical treatment, allowing less than λ/4 of phase shift across the lens gives

Minimum Spot size: $$d_{\min} = 0.43 C_s^{1/4} \lambda^{3/4}$$

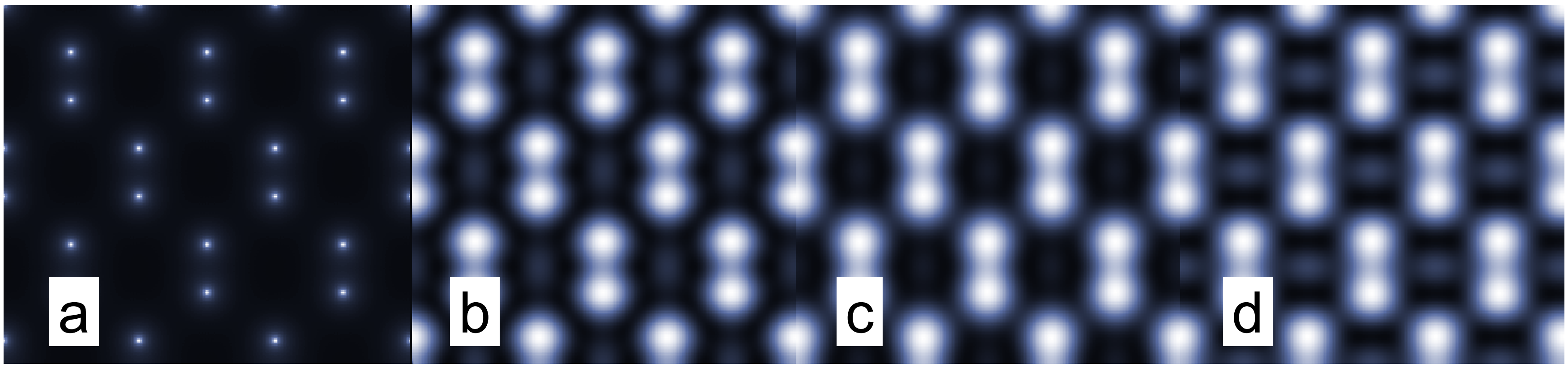
Optimal aperture: $$\alpha_{opt} = \left(\frac{4\lambda}{C_s} \right)^{1/4}$$

At 200 kV, λ=0.0257 Å, C_s = 1.0 mm, d_{min} = 1.55Å and α_{opt} = 10 mrad
C_s = 1.2 mm, d_{min} = 1.59Å and α_{opt} = 9.6 mrad

C_s = 0.5 mm, d_{min} = 1.28Å and α_{opt} = 12 mrad
C_s = 0.6 mm, d_{min} = 1.34Å and α_{opt} = 11 mrad

Multislice simulated Annular-Dark-Field Images of Silicon [110] in a 200 kV STEM

(Cs=0.5 mm, Probe forming aperture=11.9 mr, ADF inner angle=30 mr)



(a) The projected potential along [110]
(b) The ADF image for a 2.7 Å thick crystal
(c) The ADF image for a 81 Å thick crystal
(d) The ADF image for a 2.7 Å thick crystal (1.6 Å information limit)

Note: (i) The dumbbells are visible even when the (400) spot is excluded
(ii) Except the dumbbell spacing is not 1.36A, but closer to 1.6A

D. Muller, Bell Labs, 1998

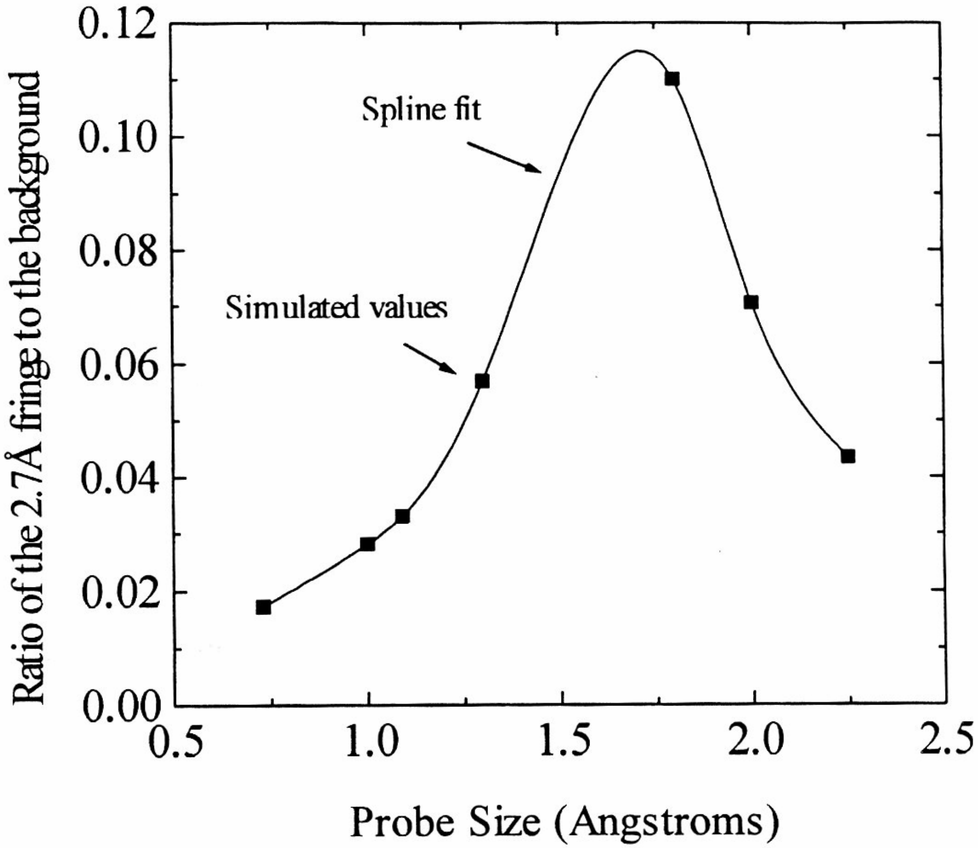
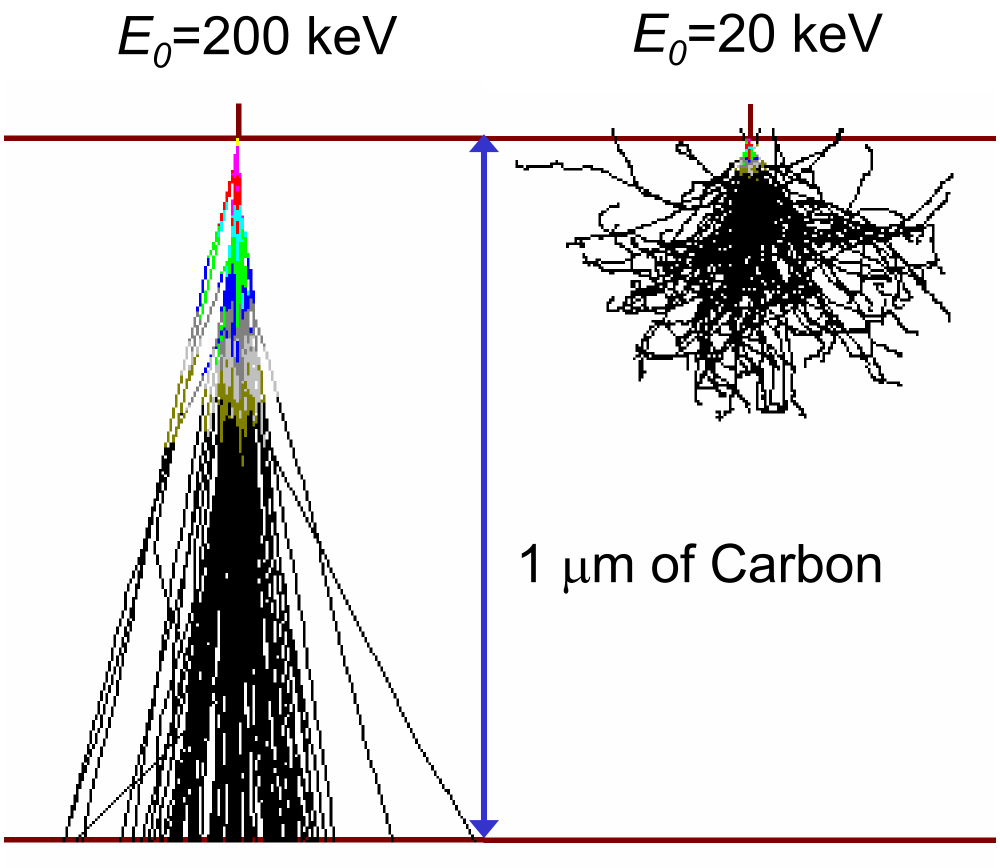


Figure 3.17: Simulated strength of 2.7 Å fringe (normalized with respect to overall linescan intensity) in 190 Å of Si (110) versus nominal probe size. For large probe sizes the fringe is beyond the resolution and is not visible, for small probes the is no 'crosstalk' between the atomic columns located 1.36 Å apart and the 'forbidden' fringe is again not visible.

S. Hillyard, PhD Thesis, Cornell University

Beam Spreading



Electron Range (in μm):

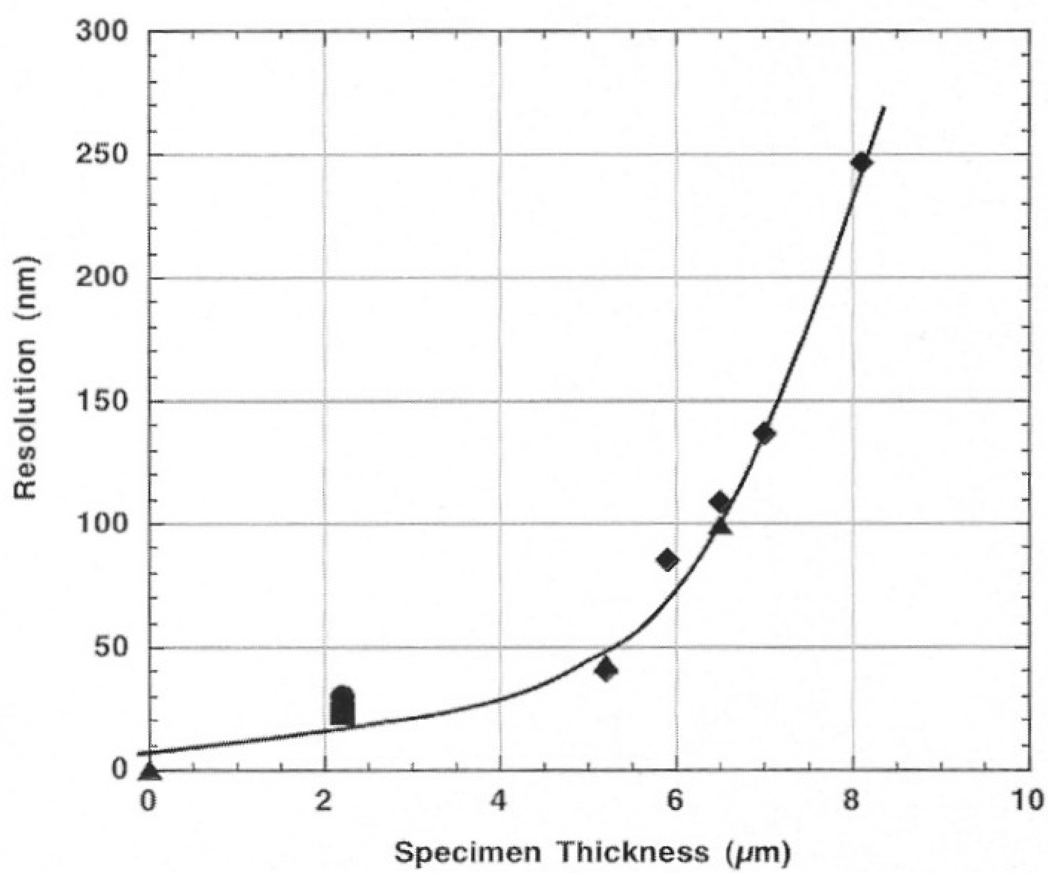
$$R \approx \frac{0.064}{\rho} E_0^{1.5}$$

(density ρ in g/cm³, E_0 in keV)

R~ 100 μm at 200 keV

Beam Spreading

At 300 kV

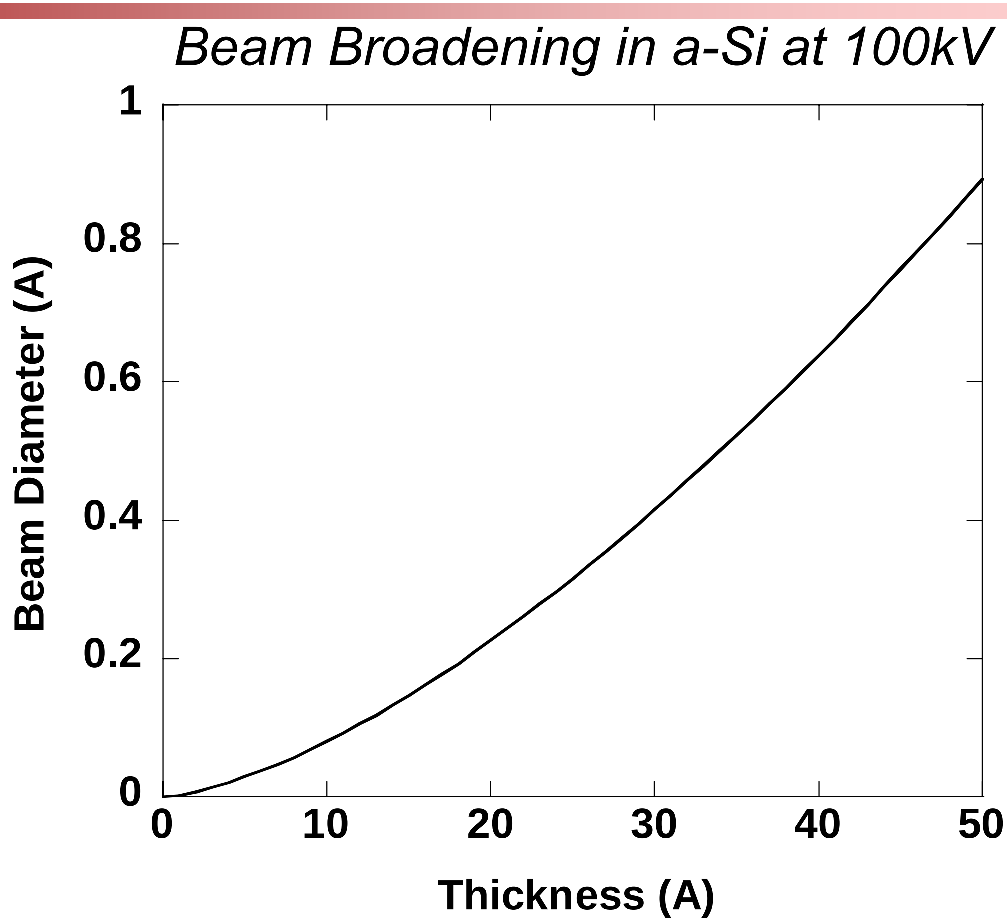


N. Zaluzec, Microscopy Today (2004)

$$\text{Beam Spreading} \propto \frac{(Zt)^{1.5}}{E_0}$$

At 100 kV
0.16 nm for 10 nm thick C
1.8 nm for 50 nm thick C

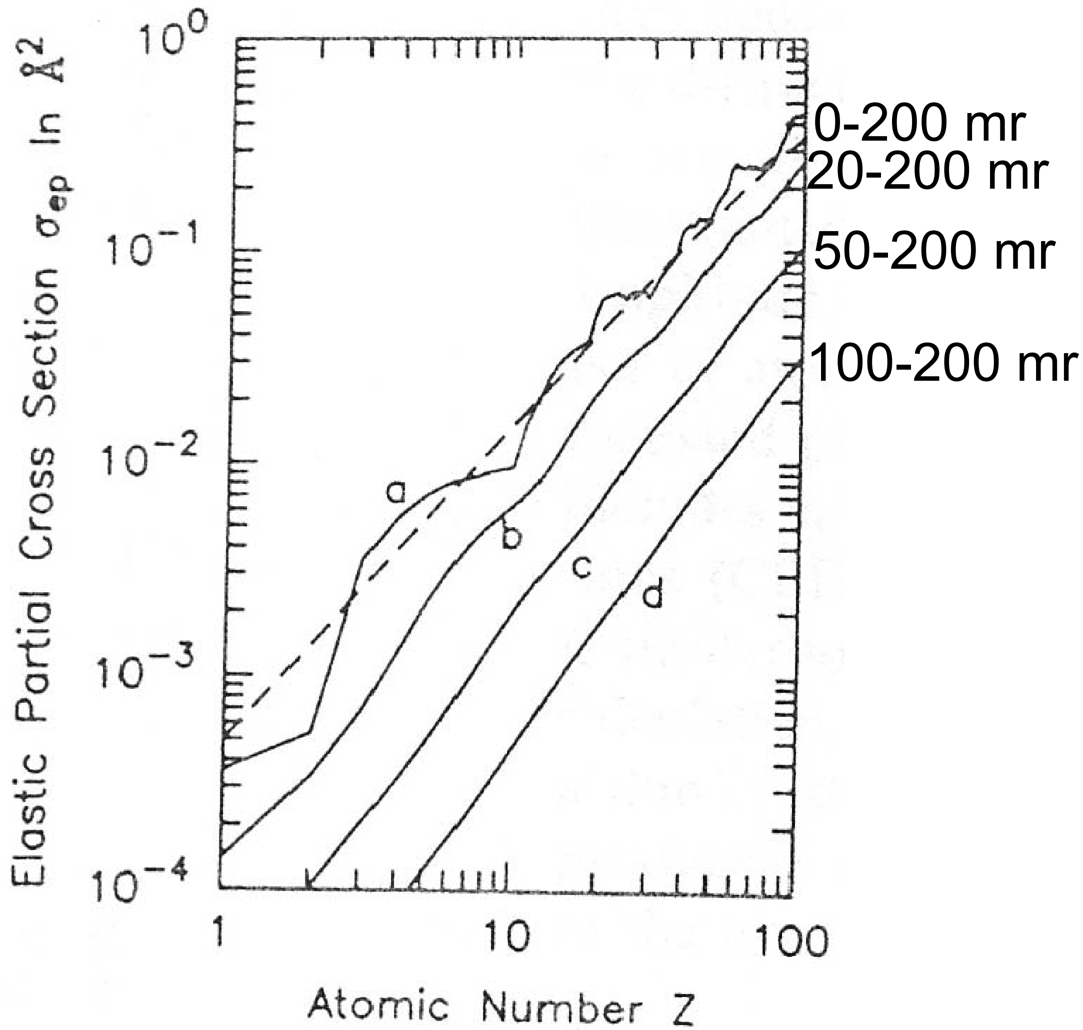
How does an amorphous layer on the entrance surface degrade resolution?



Increased Apparent Source size → *Loss of resolution, loss of apparent brightness*

- ***No FIB'ed samples!***
- ***Ignore for probes larger than 0.4Å***

Phase vs. ADF Contrast



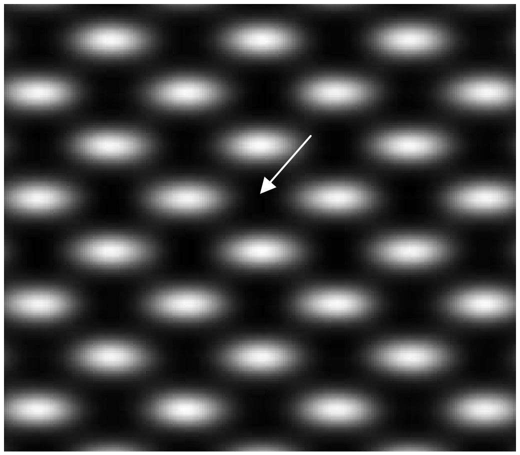
ADF Signal is much weaker than HR-TEM

E. J. Kirkland et al, Acta Cryst, 1987

Imaging a Single Antimony Atom in 4.5 nm of Silicon

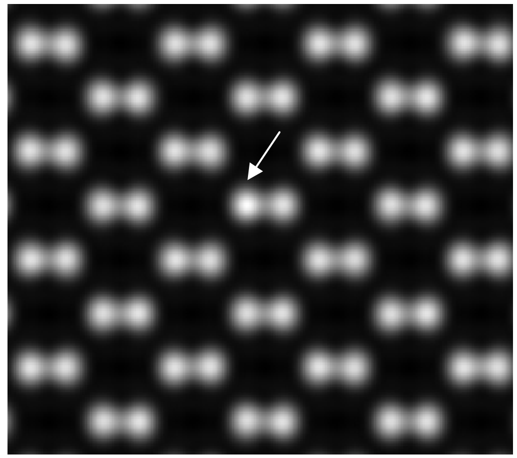
(the atom is 2.1 nm from the top surface)

HRTEM



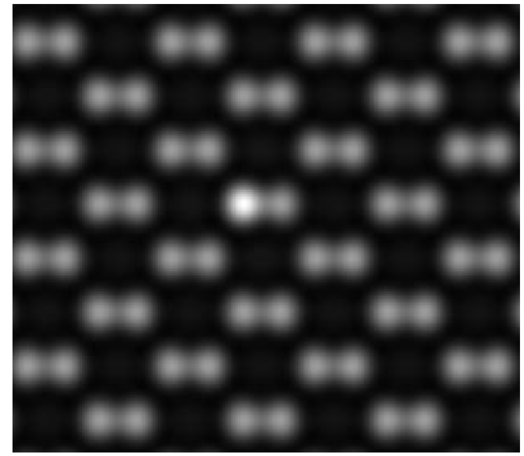
C_s=0.5 mm
(JEOL 2010F URP)

Exit Wave Reconstruction



C_s = 0 mm
0.75 Å information limit

ADF-STEM



C_s=0.5 mm
(JEOL 2010F URP)

Multislice simulations assume a 200 kV electron beam

Sb contrast: HRTEM 0%, EWR: 10% (5% at 1.2Å) , ADF-STEM:65%

Why do we get more signal in ADF? Channeling!

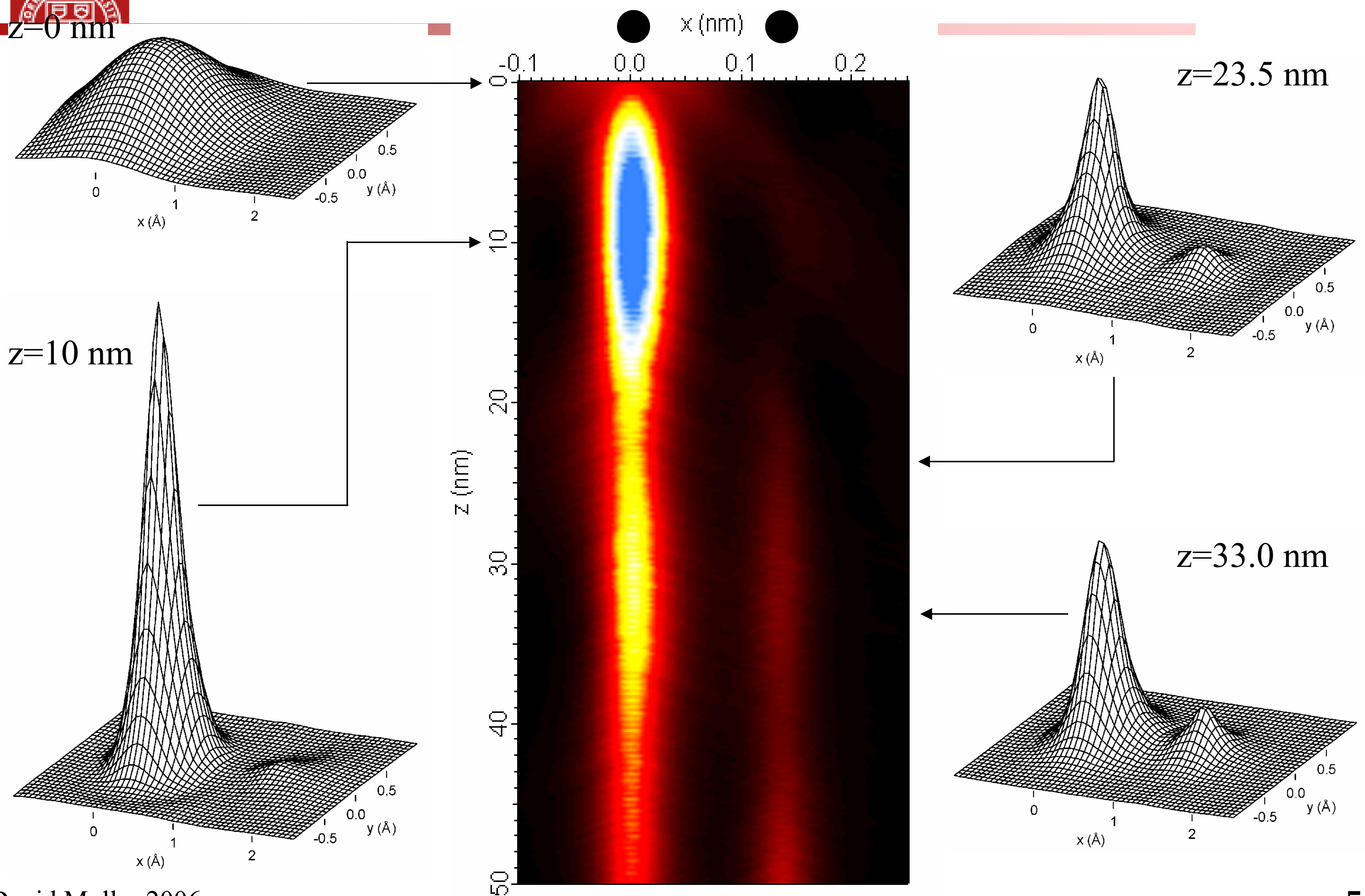
ADF in Thicker Samples

- *Simple specimen transmission function model:*

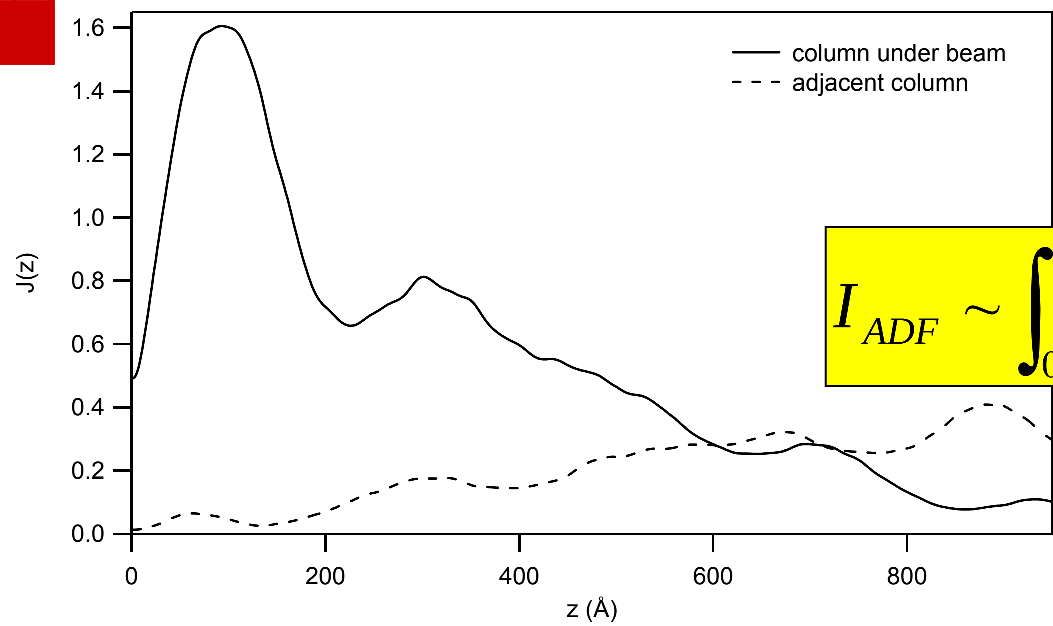
$$I_{ADF} \sim |h|^2 \otimes |t|^2$$

- Suggests that wave amplitude is important, not phase as in conventional HRTEM
- Interaction of the fast electrons with the periodic lattice including phonons is difficult
- Numerical simulations

Probe Channeling

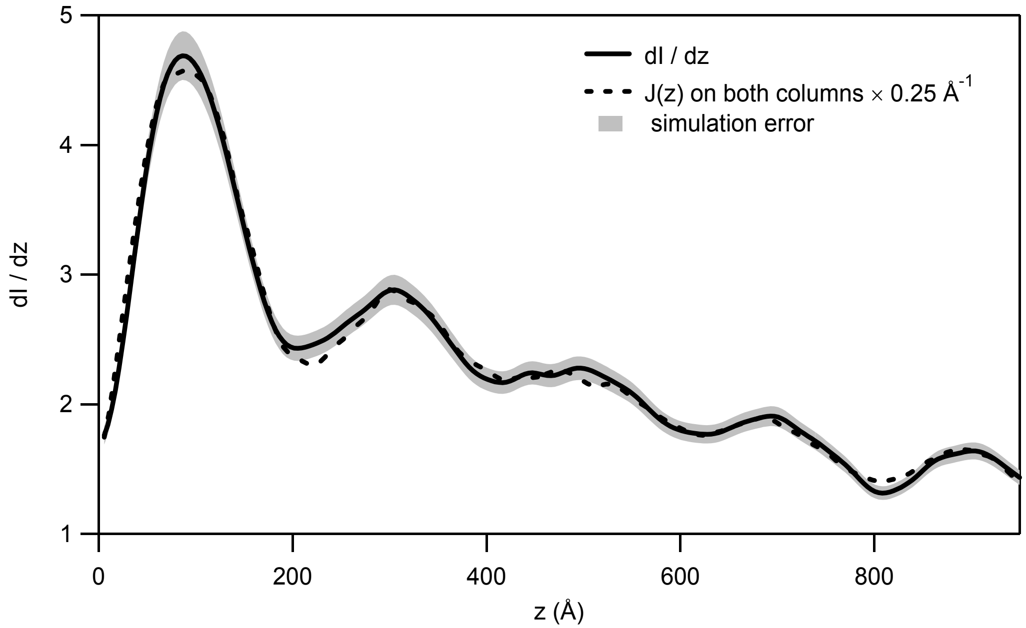


ADF Signal tracks Probe Amplitude

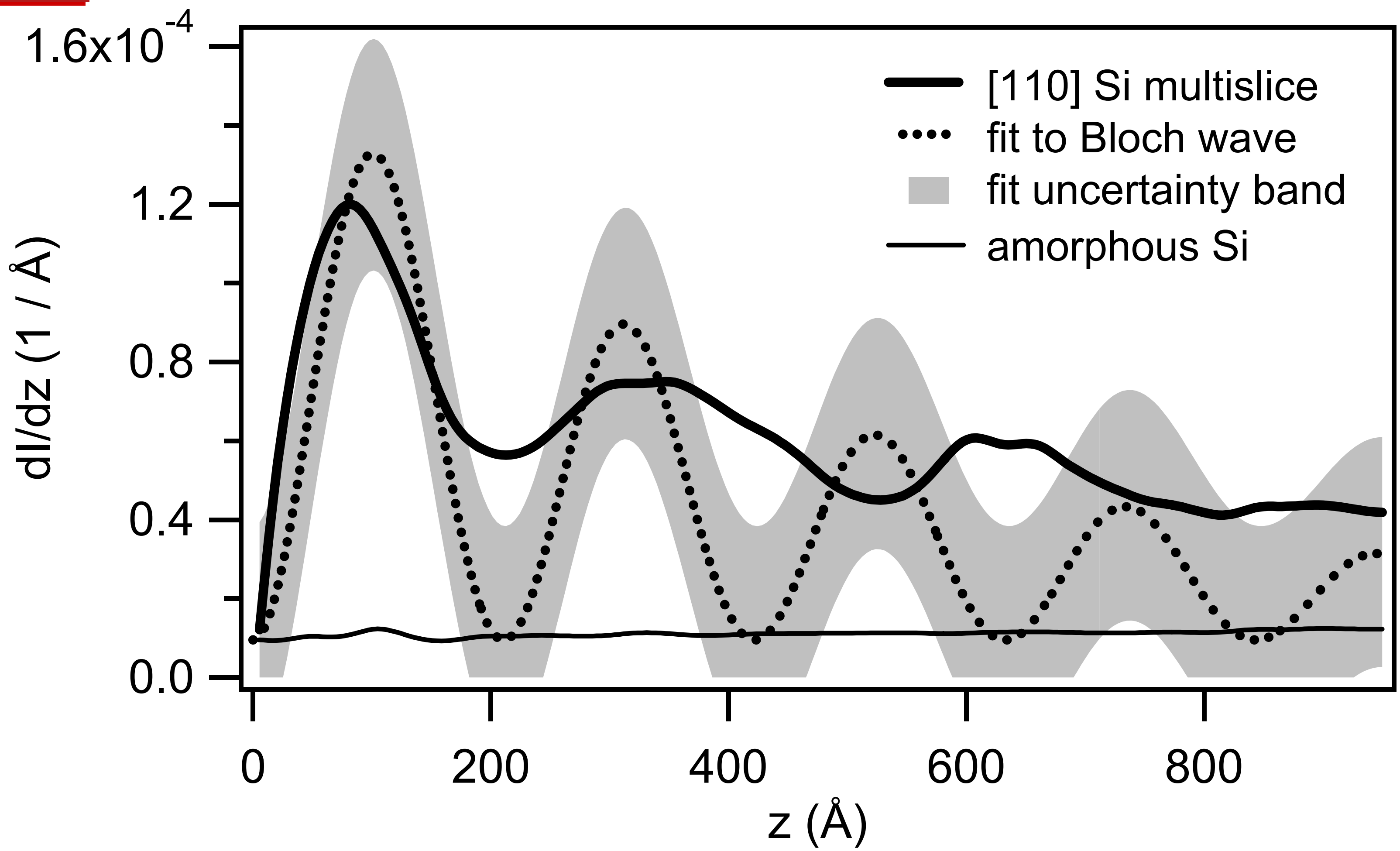


Si [110] at 300K
(200 kV, C_s=1 mm)

$$I_{ADF} \sim \int_0^{z_0} |h(\vec{r},z)|^2 \otimes_r |t(\vec{r},z)|^2 \mathrm{d}z$$

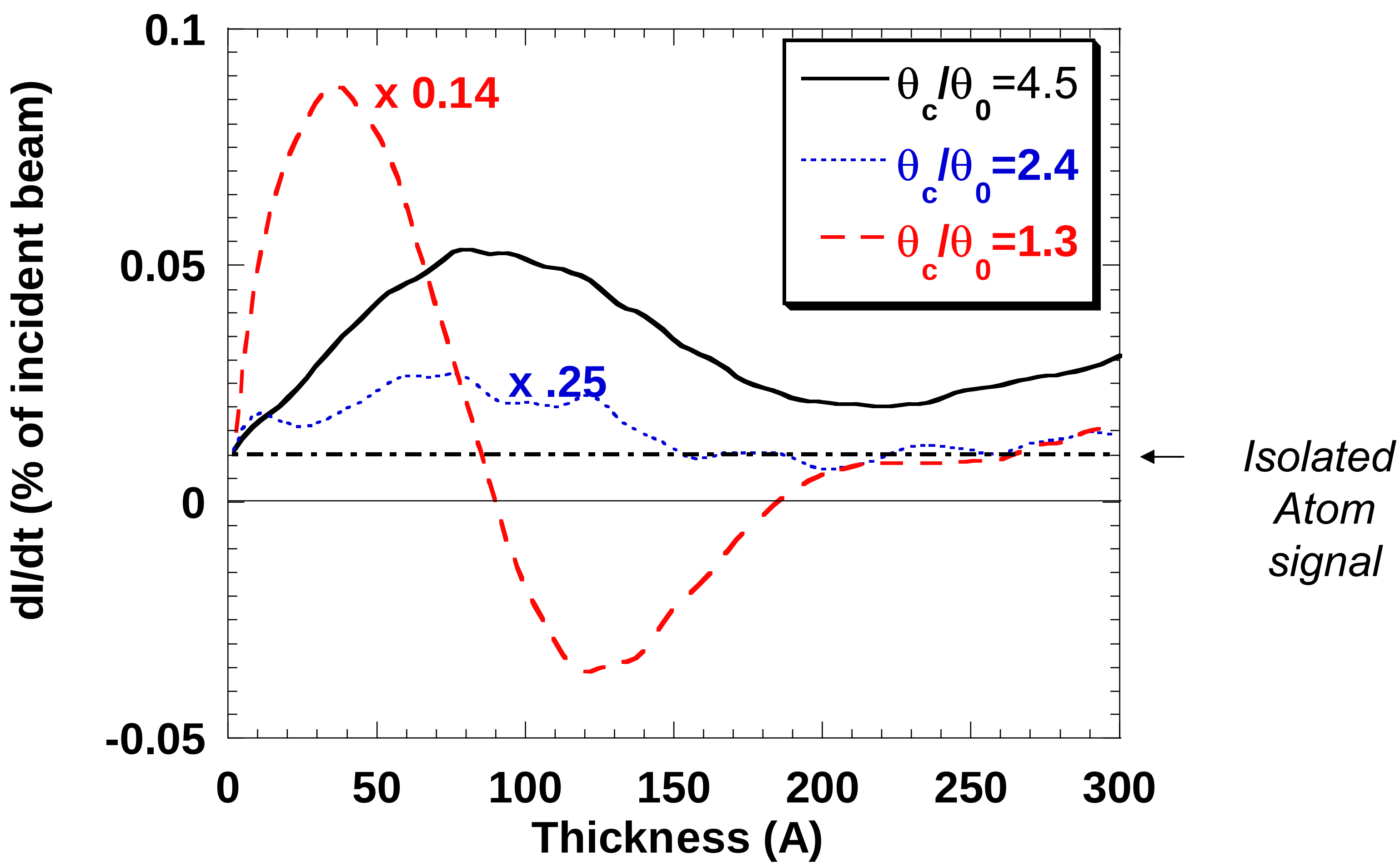


Channeling down

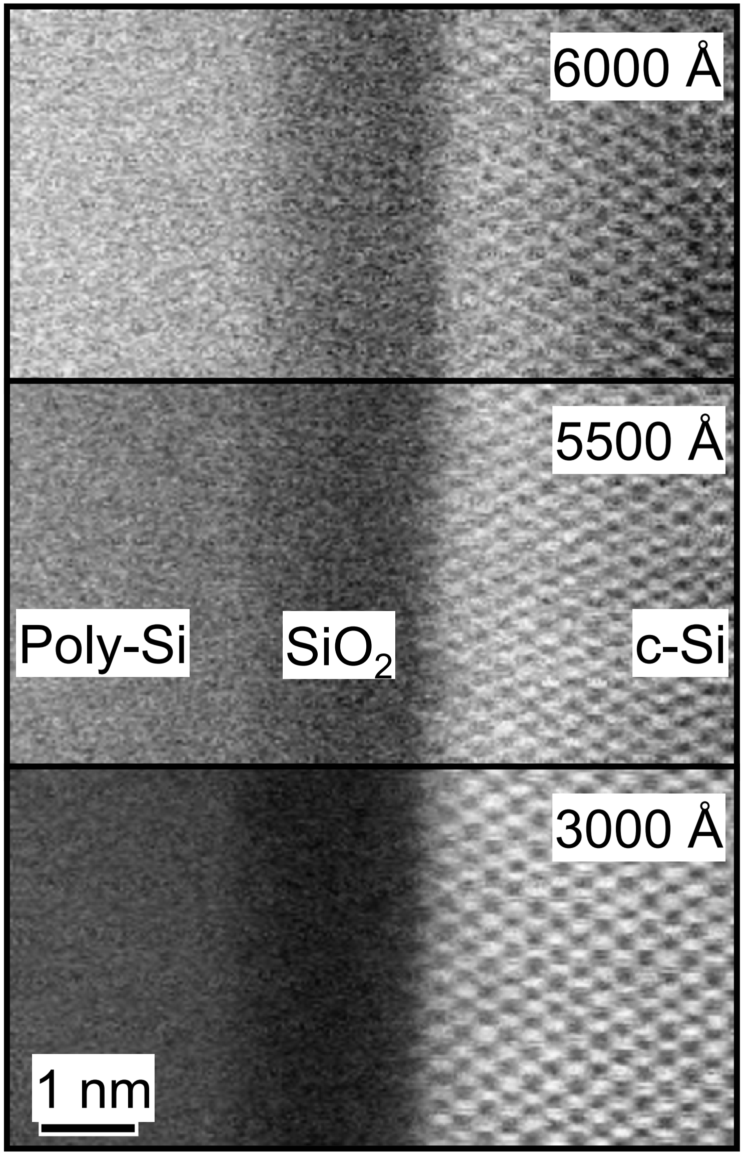


Signal vs Collection Angle

[011]Si, 300 kV, 9 mr probe angle



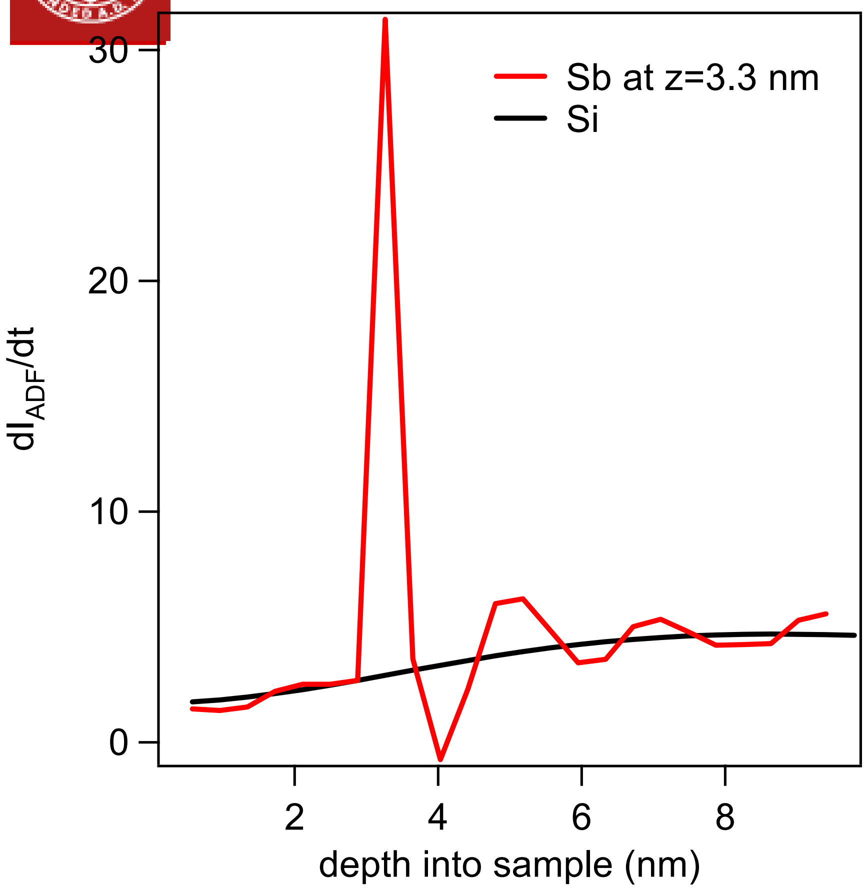
Imaging Thick Cross-Sections

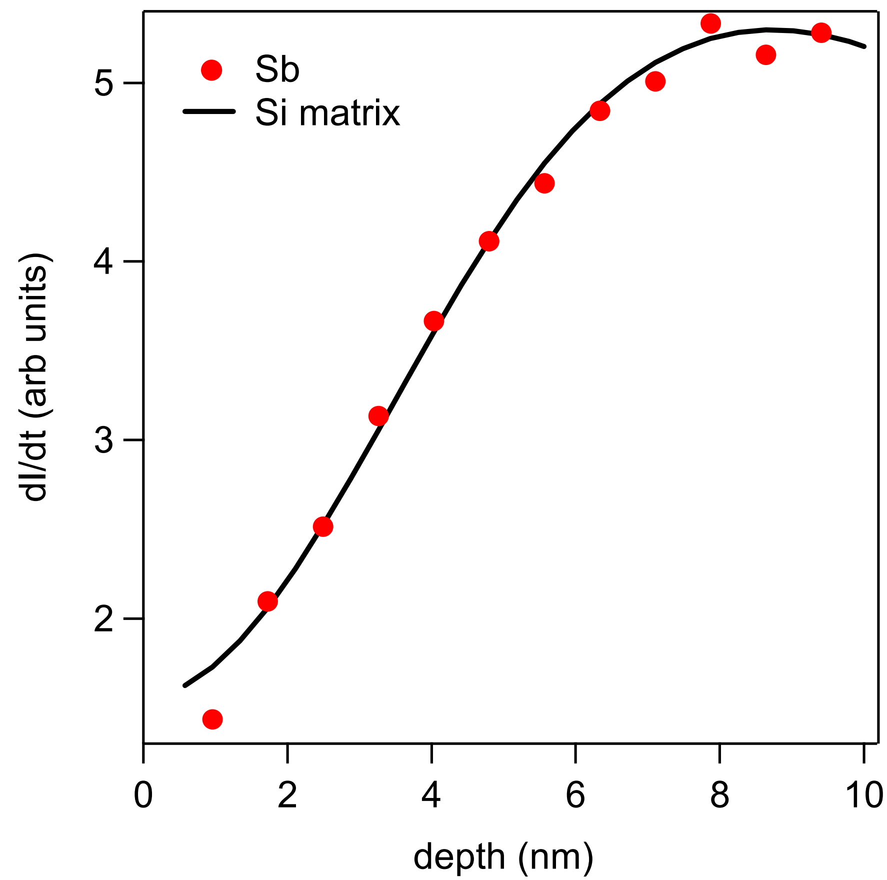


Gate Oxide Thickness: 20 Å

- ADF Images decay gracefully with increasing thickness
- Apparent Oxide Thickness is unchanged with thickness
- Apparent Interface Roughness increases from 1.6 to 2.7 Å rms
- "white band" develops (depends on thickness and ADF angles)

One Sb Atom vs. Depth

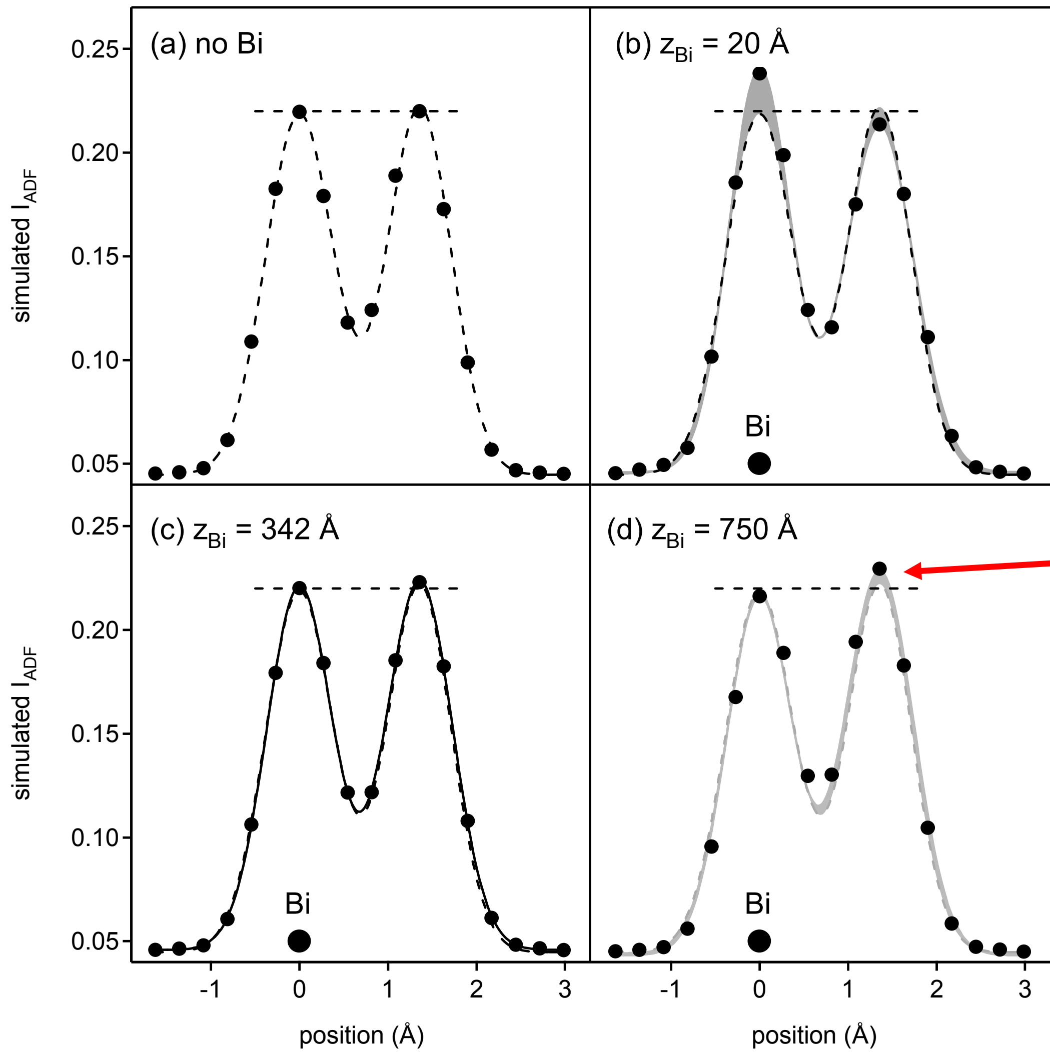




Scattering from one Sb atom ∝ Si scattering at the same depth.

Dopants as probes of Beam Spreading

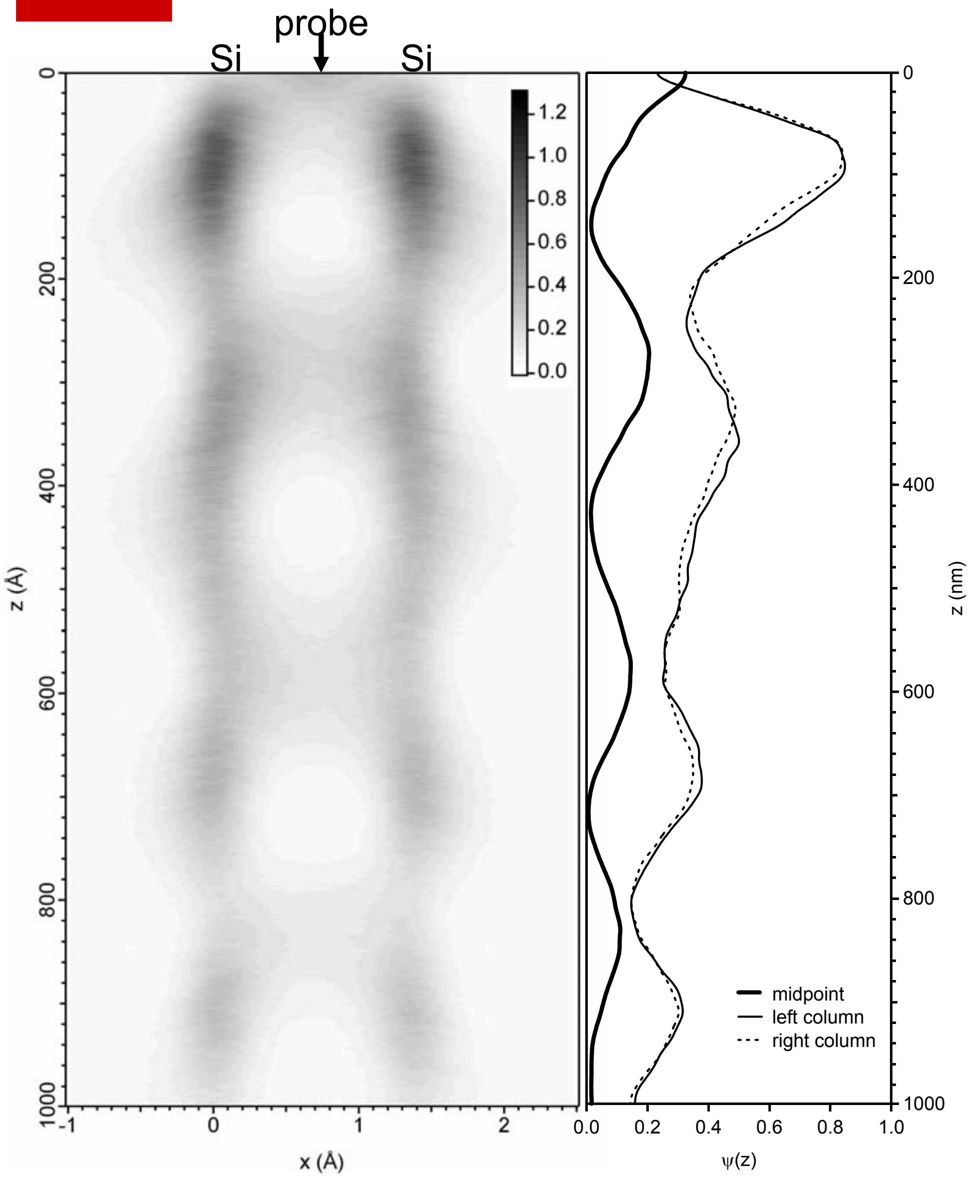
(Multislice for 75.8 nm Si in 100 kV C_s-corrected STEM)



Wrong column Appears bright!

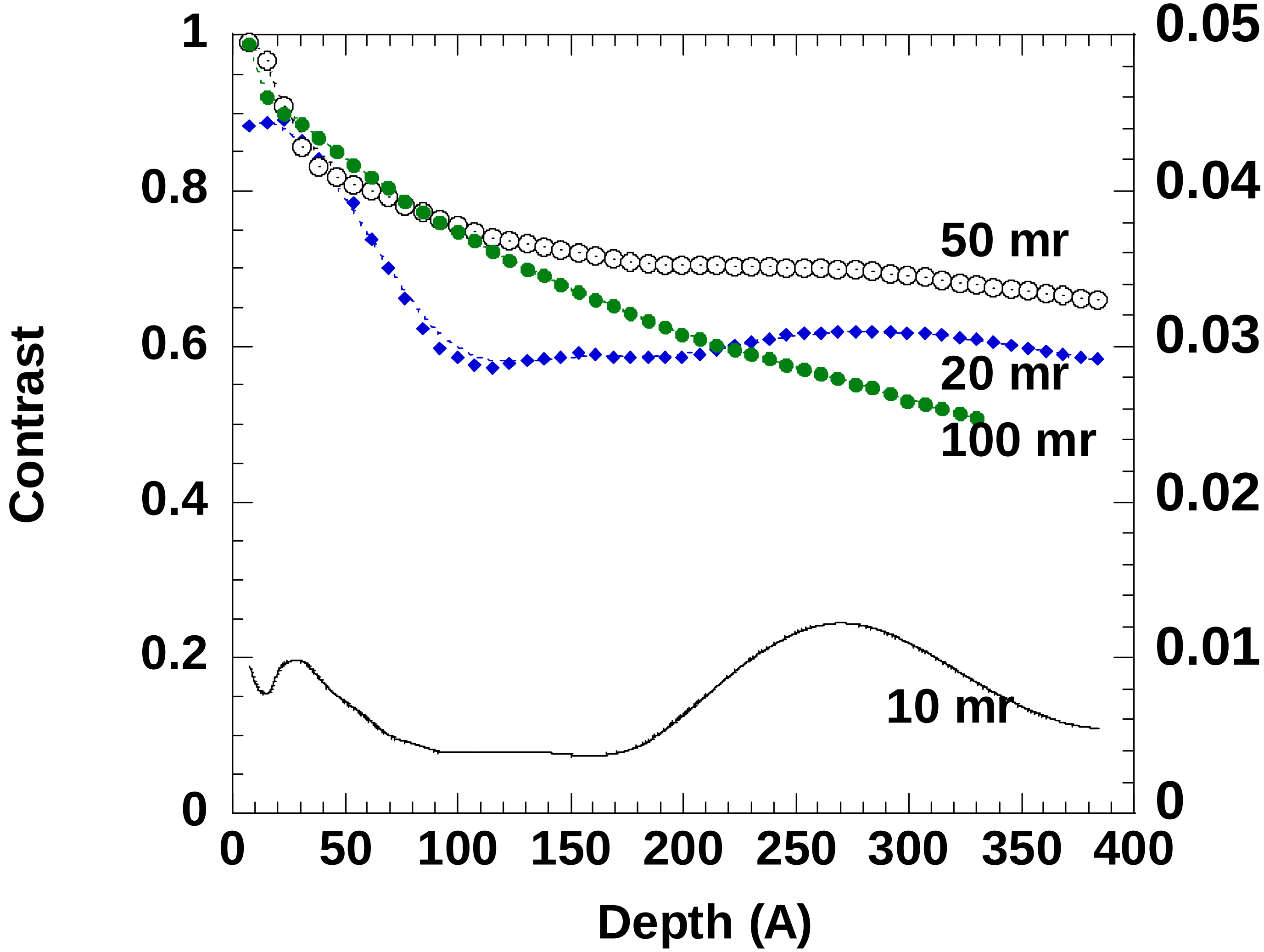
P. M. Voyles, D. A. Muller, E. J. Kirkland, *Microscopy and Microanalysis* **10,** 291-300 (2004).

Channeling Down Si [110]

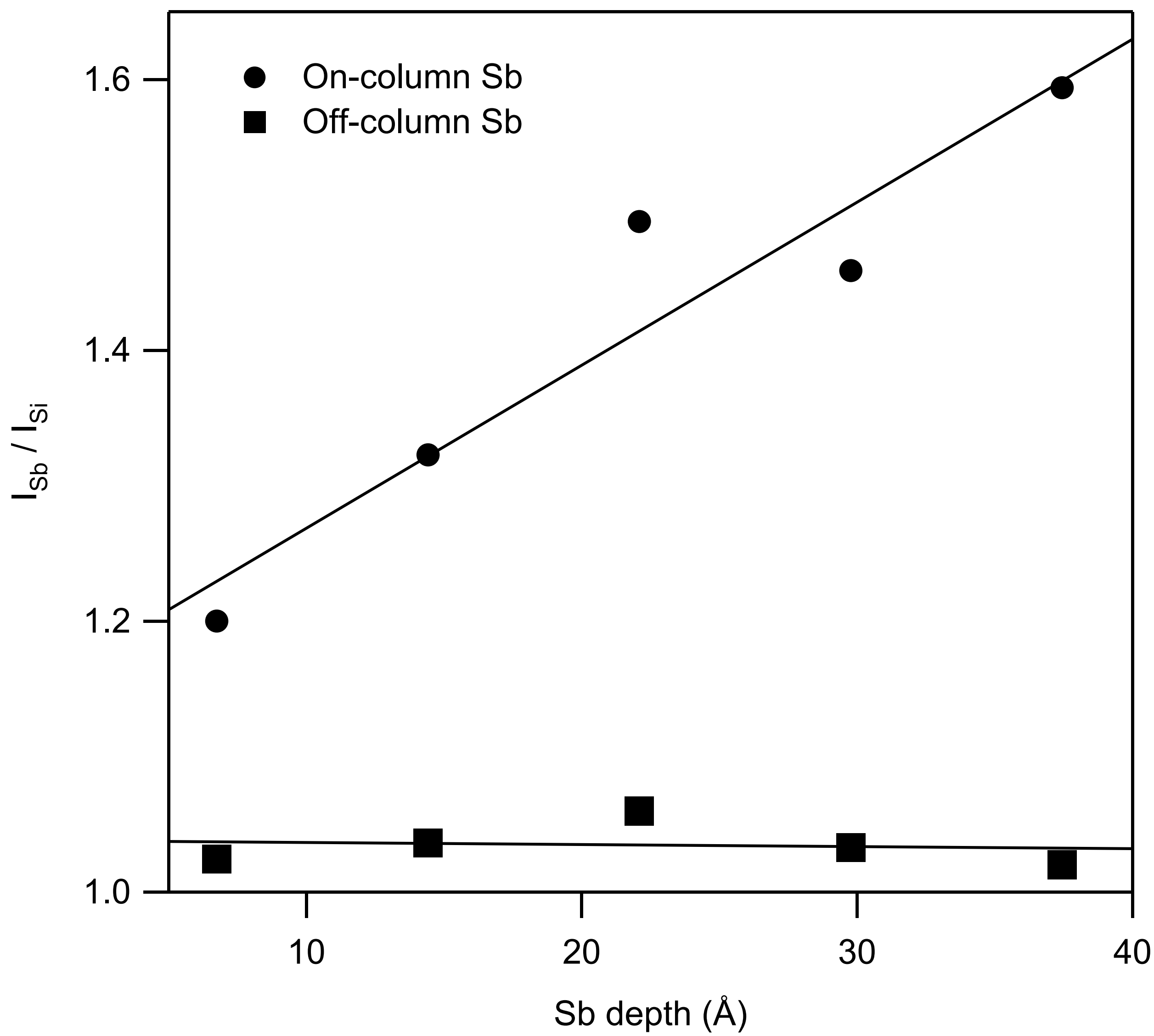


- Probe doesn't stay between atom columns -oscillates
- Almost entirely on atom columns at 100, 400Å
- When on-column, scattering is large
  - Will reduce dumbbell contrast

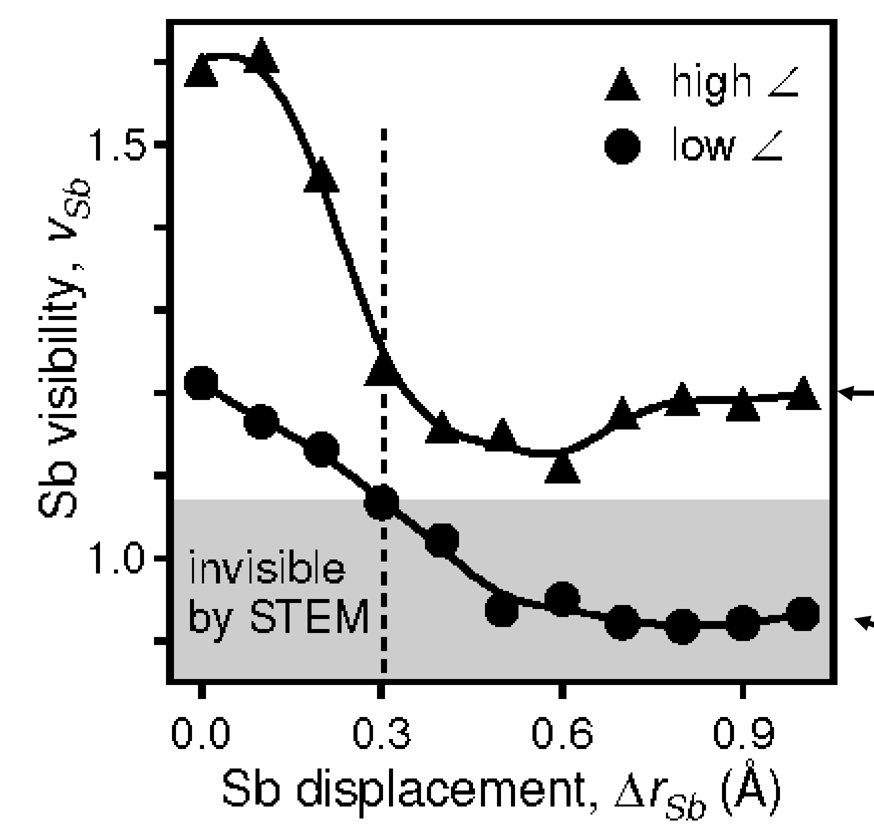
Si Dumbell Contrast vs.Probe Angle

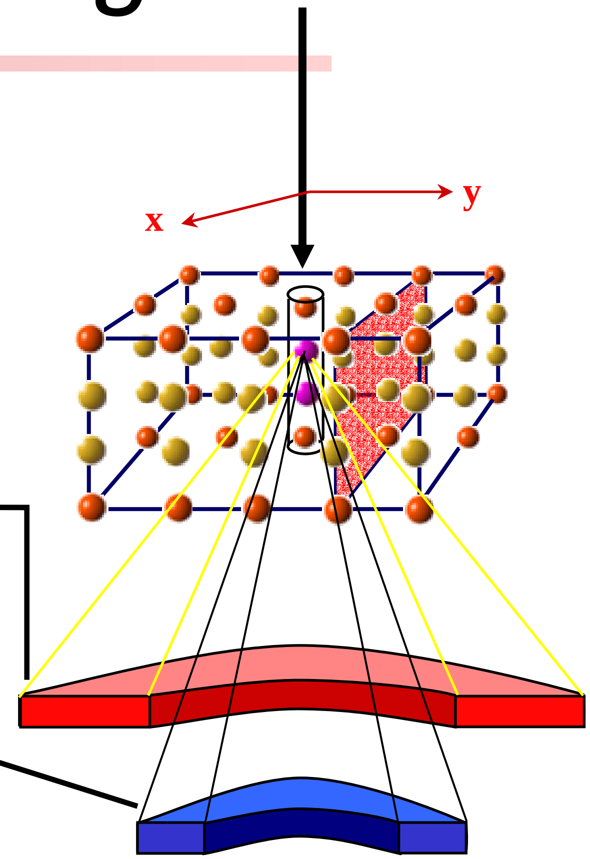


On-column vs Off



Effect of Camera Length

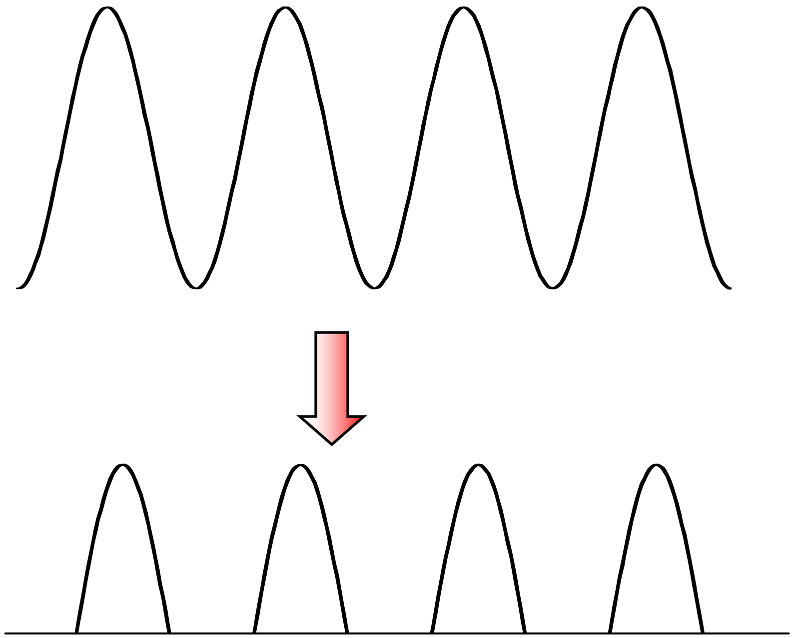




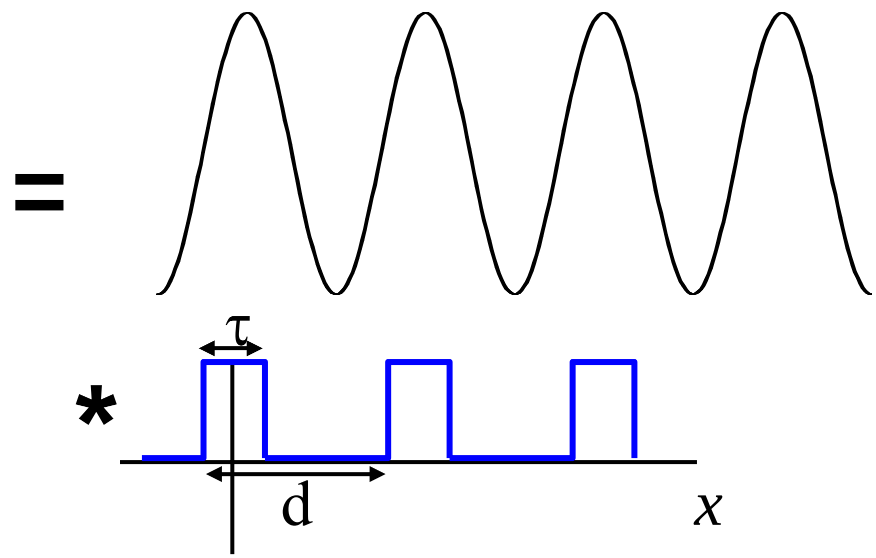
simultaneous
acquisition in
exact registration

P. M. Voyles, D. A. Muller et al, *Phys. Rev. Lett.* **91**, 125505 (2003).

Clipping an Image is Bad
(and easy to do)



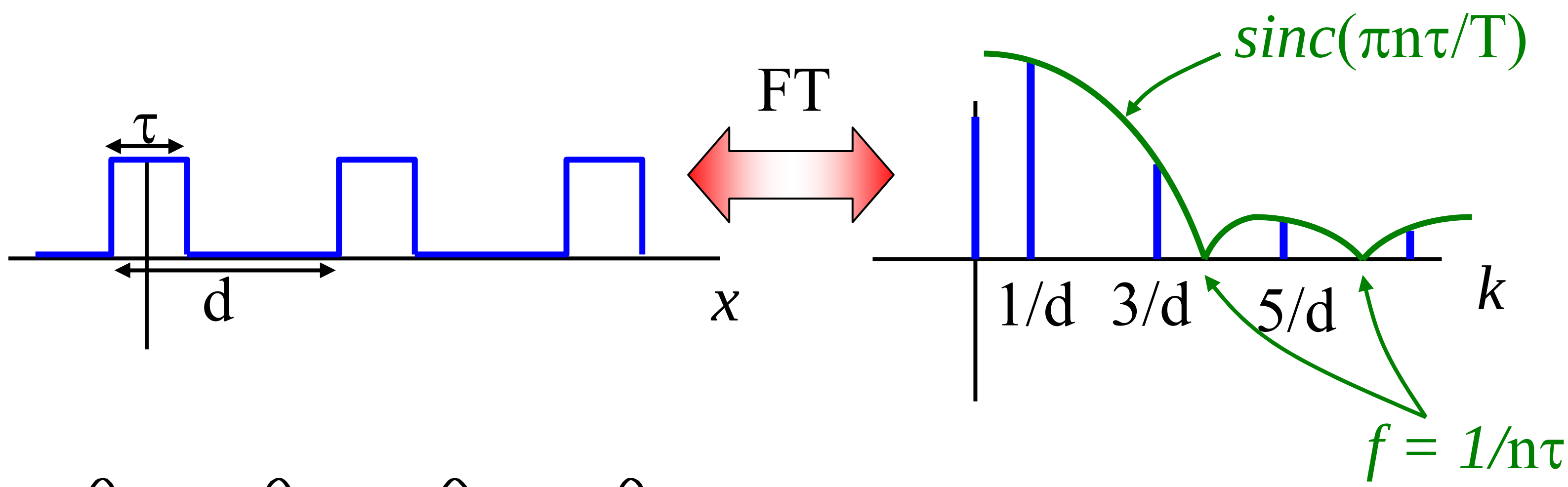
Black level set too high

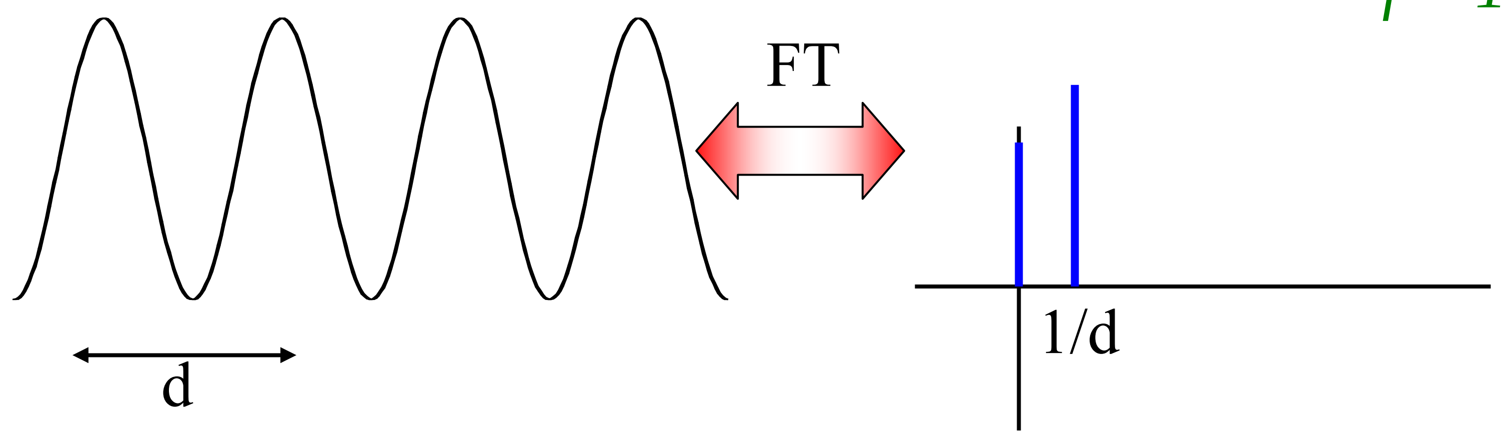


Equivalent to multiplying by square wave

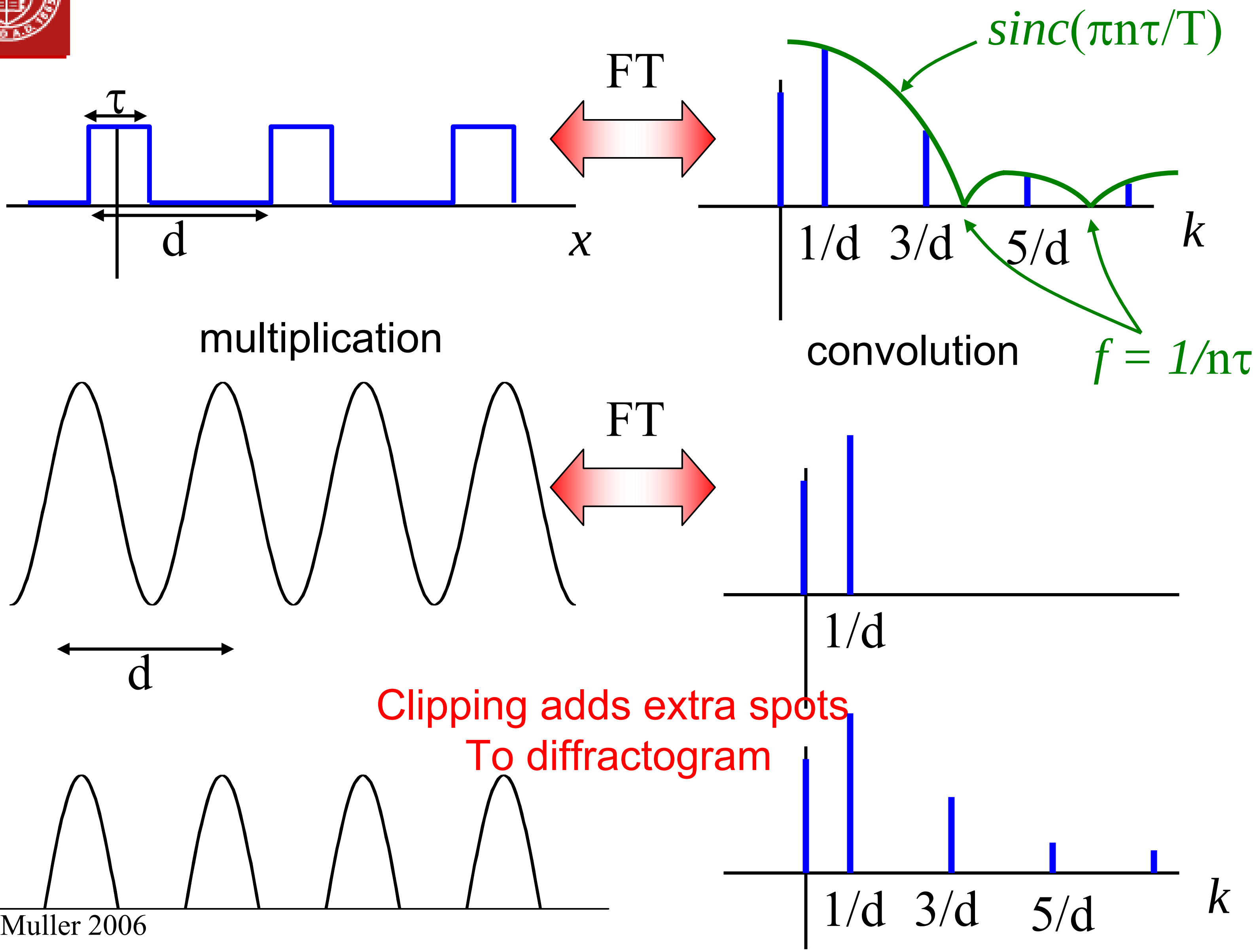
Fourier Transform of a Square Wave

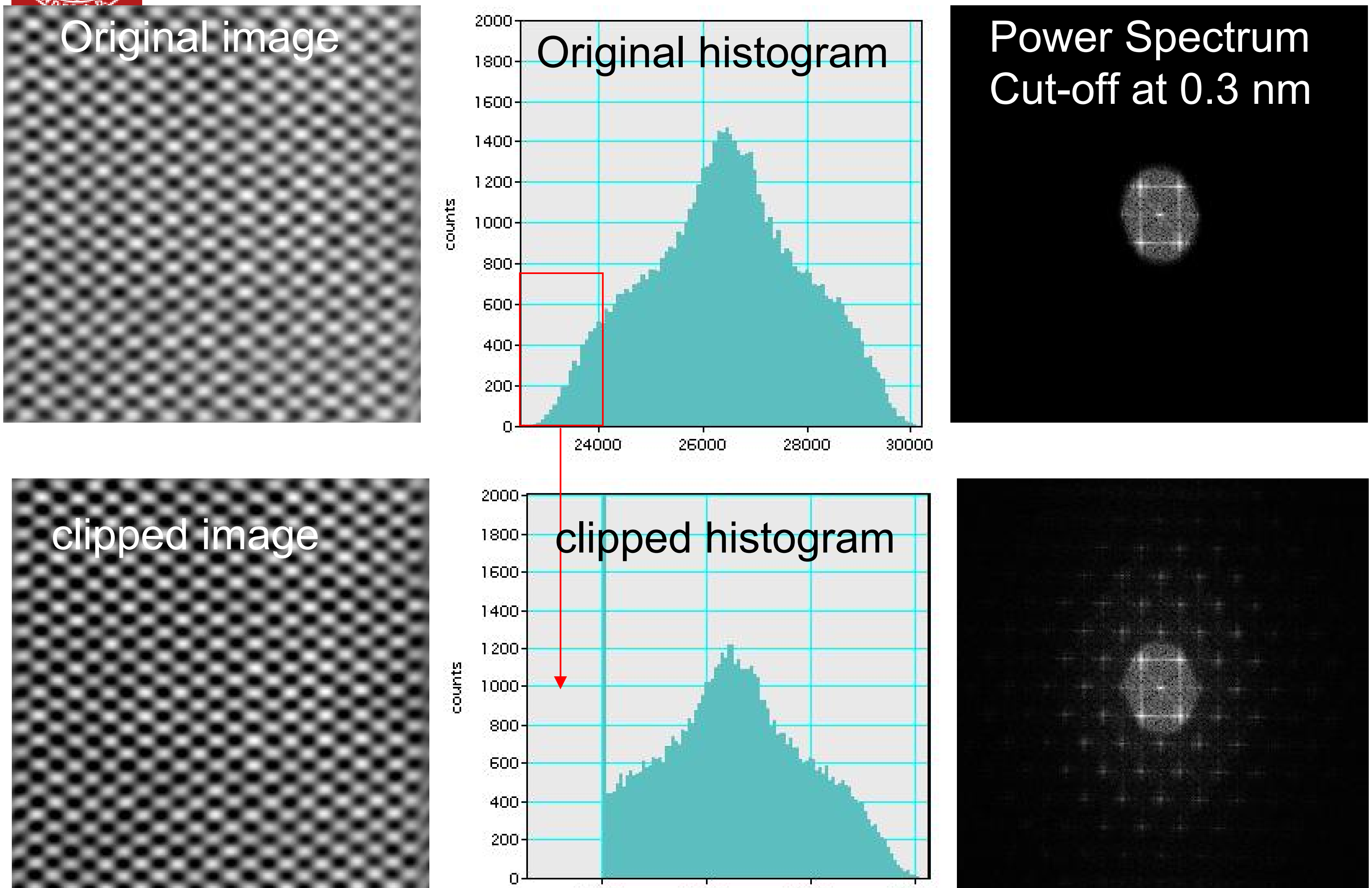
$$f(t) = \frac{A\tau}{T} + \sum_{n=1,3,5}^{\infty} \frac{2A\tau}{T} \frac{\sin(n\pi\tau/T)}{n\pi\tau/T} \cos(2\pi n f_0 t)$$





Fourier Transform of a Square Wave




Original image
Original histogram
2000
1800
1600
1400
1200
1000
800
600
400
200
0
counts
24000
26000
28000
30000
Power Spectrum
Cut-off at 0.3 nm
clipped image
clipped histogram
2000
1800
1600
1400
1200
1000
800
600
400
200
0
counts
24000
26000
28000
30000

Summary

- BF STEM – fake TEM
- LAADF STEM – strain contrast, single vacancy
- HAADF – depth dependent imaging of single dopants

- Check histograms to avoid clipping (extra spots)
- Ronchigrams – easier than imaging probe for align

Comparison of Brightness Measurements For Cold and Thermal Field Emitters

David Muller

Applied and Engineering Physics

Cornell University

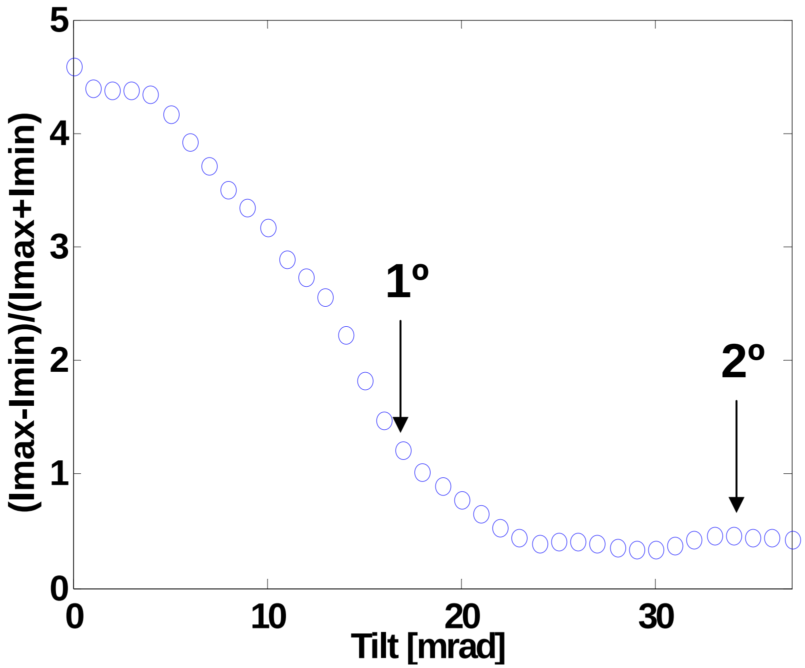
Current State of the Art:

- Few good measurements of Brightness.
- Need to measure or extract the source size (easy to overestimate)
- No reliable studies of Brightness vs. Field, Temperature or monochromation

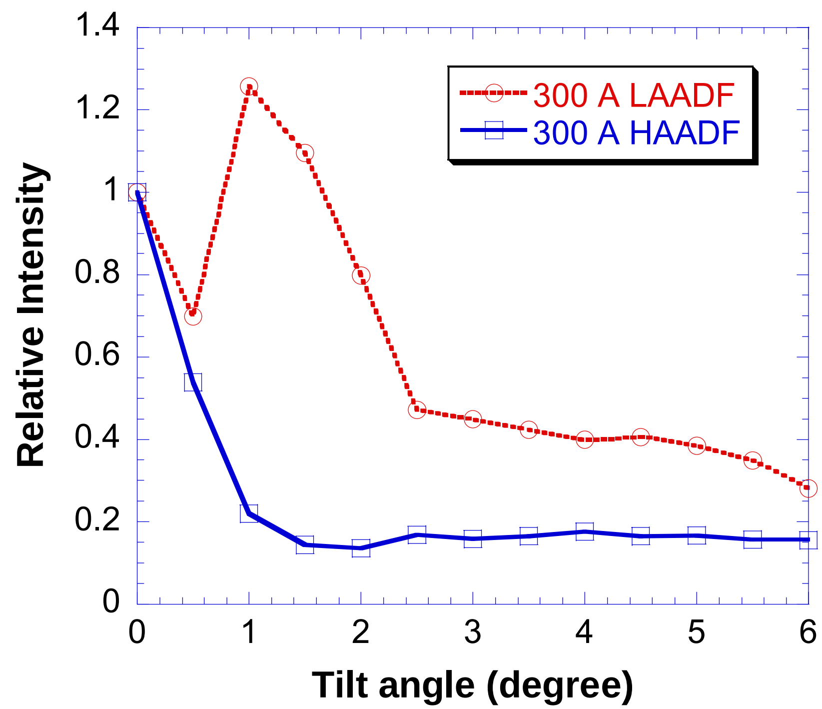
Sample Tilt in ADF-STEM

(200kV, 10 mrad)

200Å of [001] $SrTiO_3$



300Å of [011] Si



Up to~ 5 mrad of mistilt is OK before fringe contrast is reduced