Coherent Images of the Nanocrystalline World

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Colloquium, June 2003

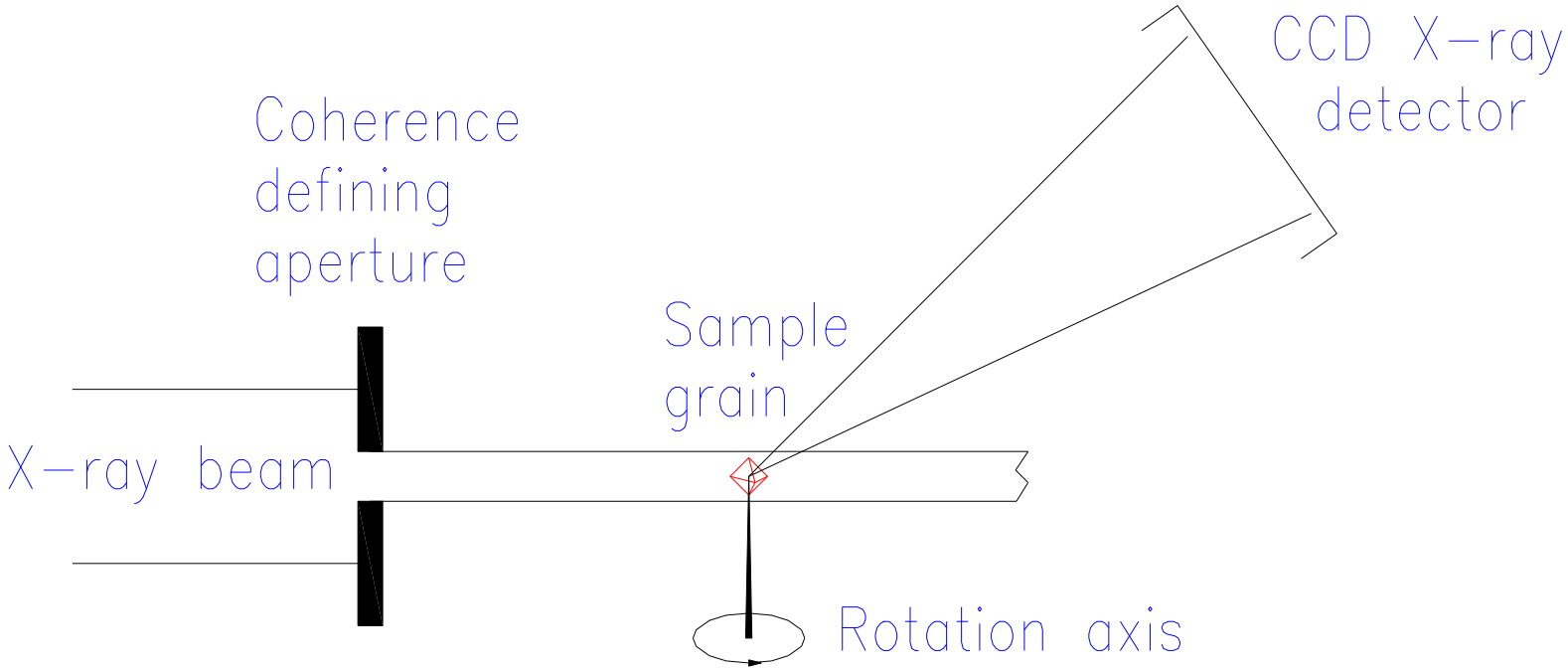
Outline

- Coherence in Diffraction
- The Phase Problem
- Coherent Diffraction from Surfaces
- Nanocrystal Shapes
- How small can we go?
- Future Applications of CXD

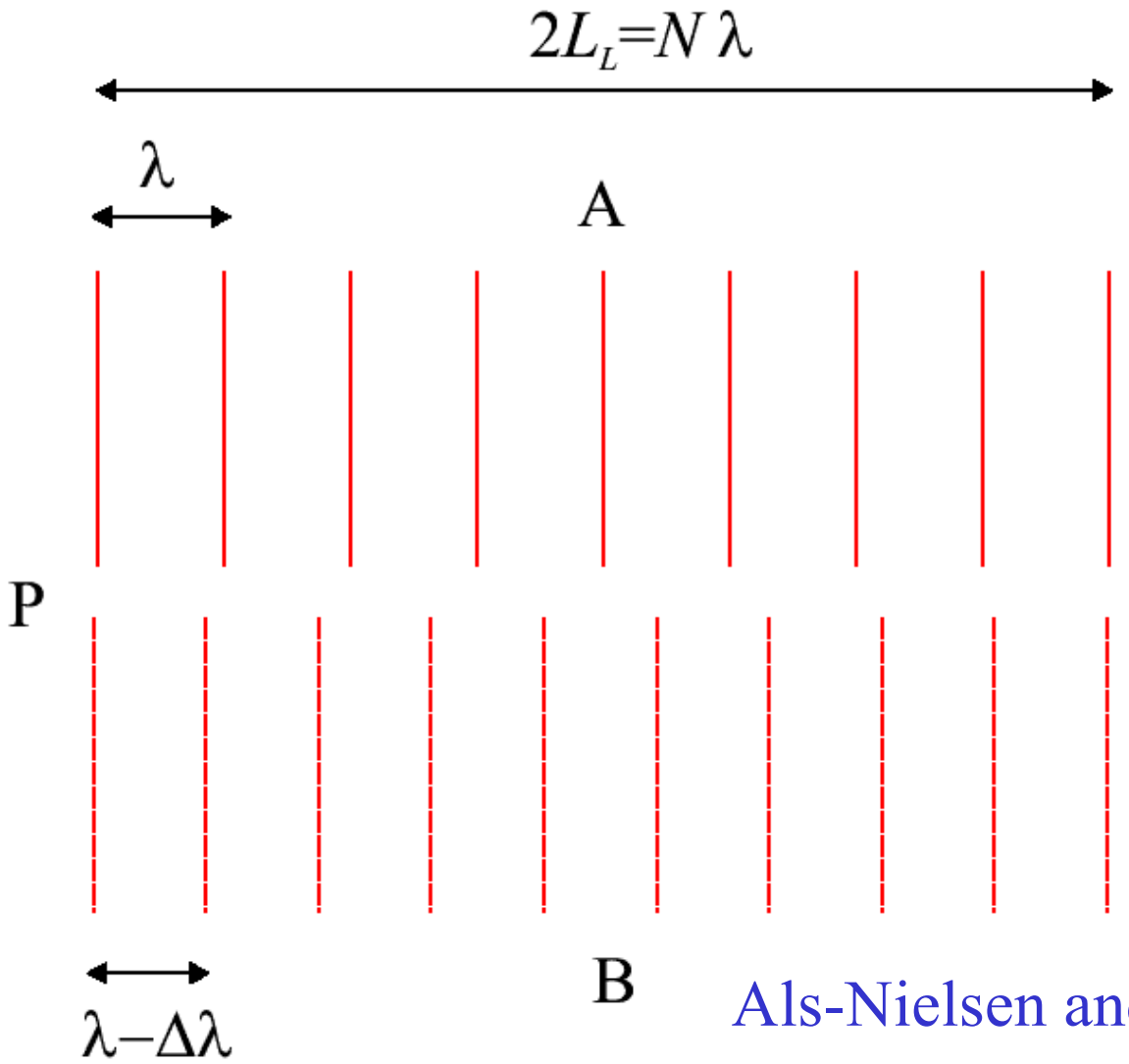
Goals of Coherent Diffraction

- Thermodynamic fluctuations
  - No ensemble average in CXD
- Probe of structure on nm scale
  - 1D, 2D and 3D
  - non-periodic object gives continuous F(**q**)
- Oversampling (in reciprocal space) permits solution of the phase problem

Lensless X-ray Microscope



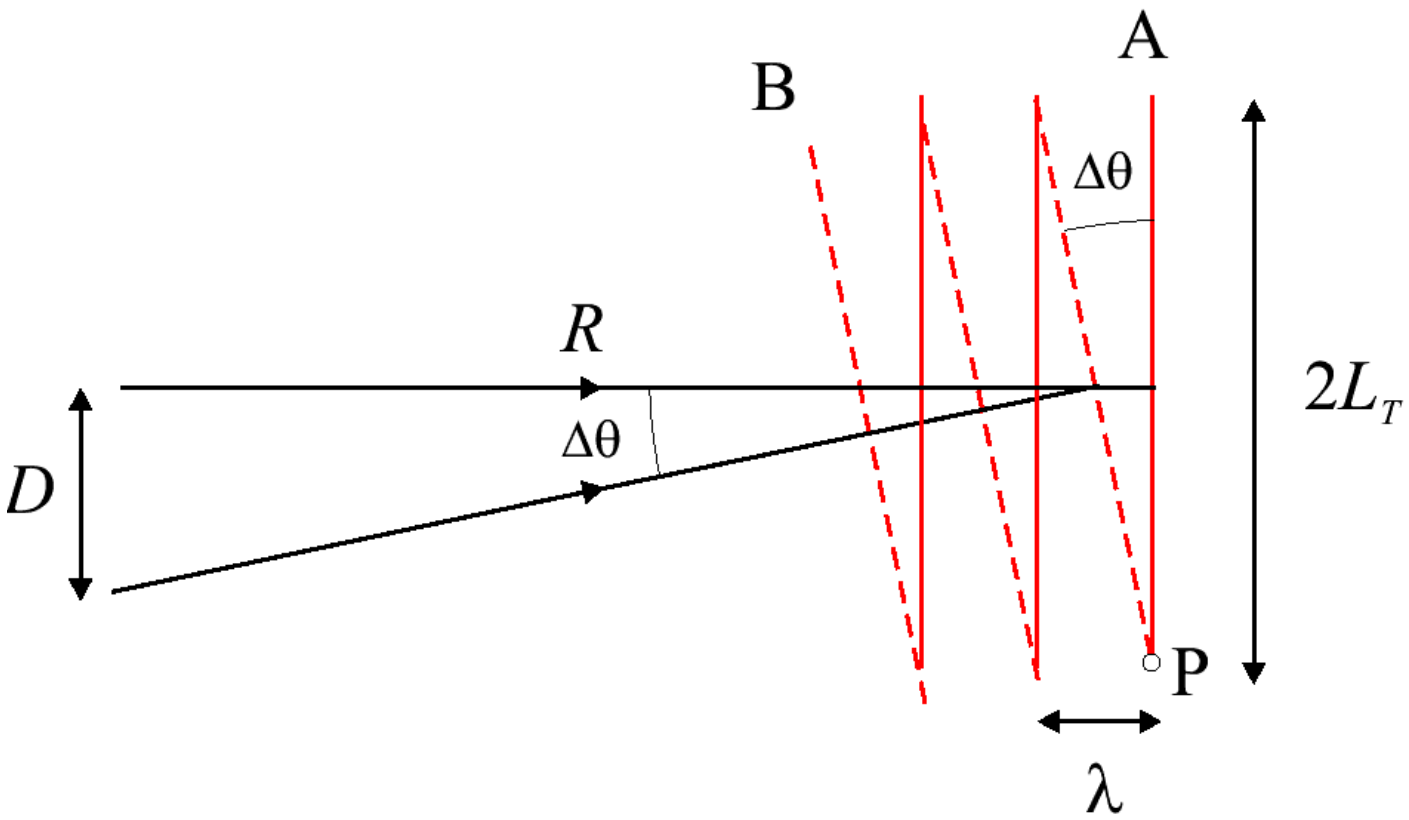
Longitudinal Coherence



$$L_L = \frac{1}{2}\frac{\lambda^2}{\Delta\lambda}$$

Als-Nielsen and McMorrow (2001)

Lateral (Transverse) Coherence



$$L_T = \frac{\lambda}{2}\frac{R}{D}$$

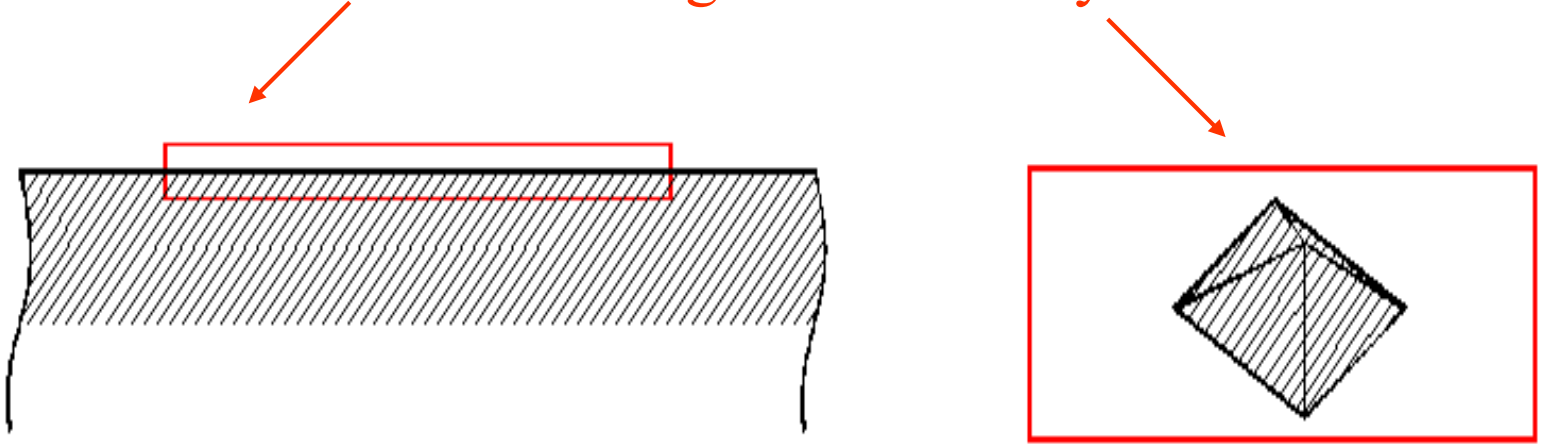
Als-Nielsen and McMorrow (2001)

Coherence at the APS

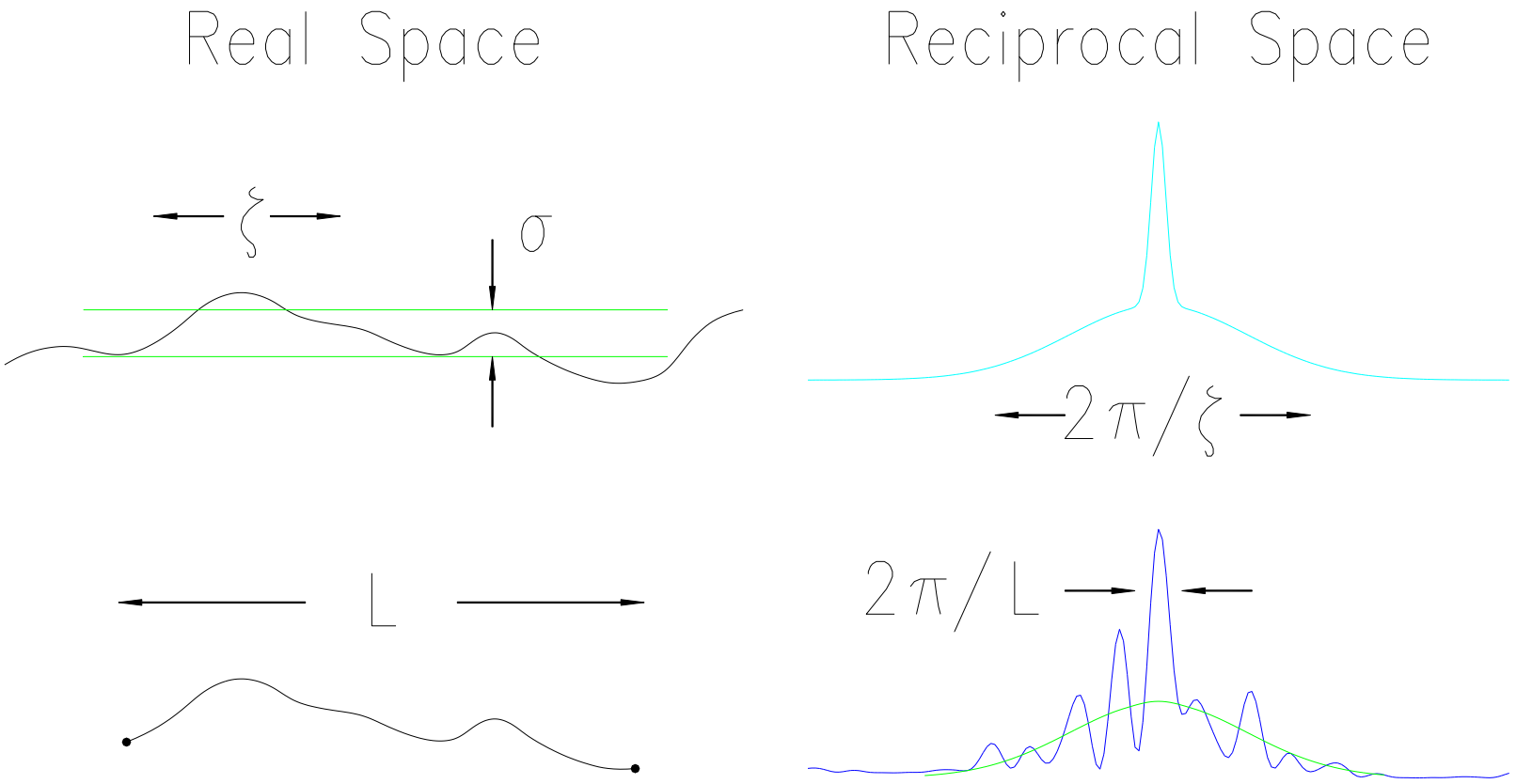
Typical 3rd Generation (undulator) Synchrotron Source

Coherence of	ξ_{VER}	ξ_{HORIZ}	ξ_{LONG}	Flux
Raw Undulator	35μm	9μm	0.004μm	2×10^{12}
Si(111) Monochromator	35μm	9μm	1μm	1×10^{10}
C(111) Monochromator	35μm	9μm	3μm	3×10^{9}

Coherent region defined by slits

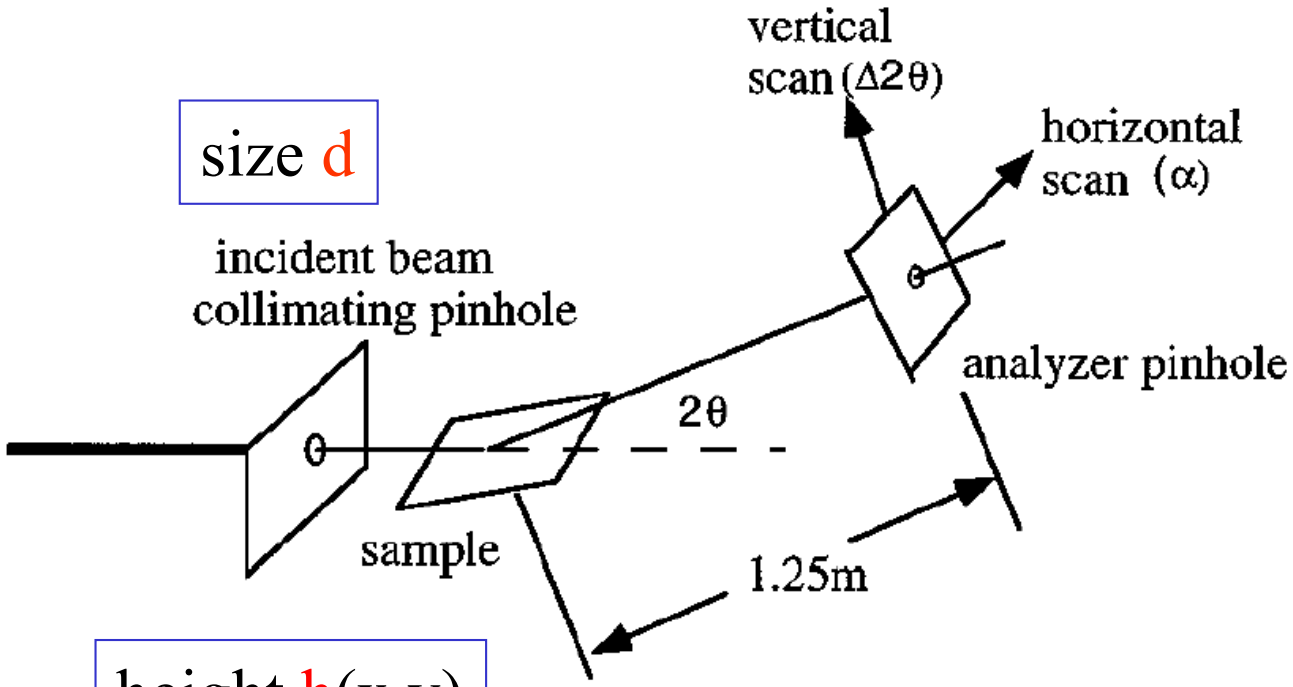


Diffuse Scattering acquires Structure using CXD



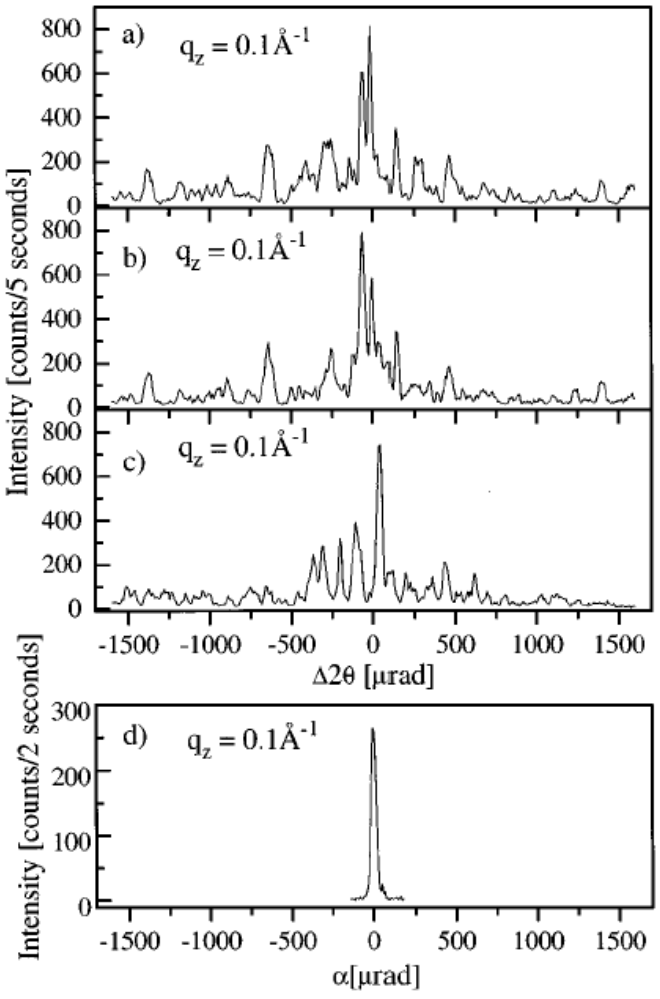
Surface Coherent X-ray Diffraction

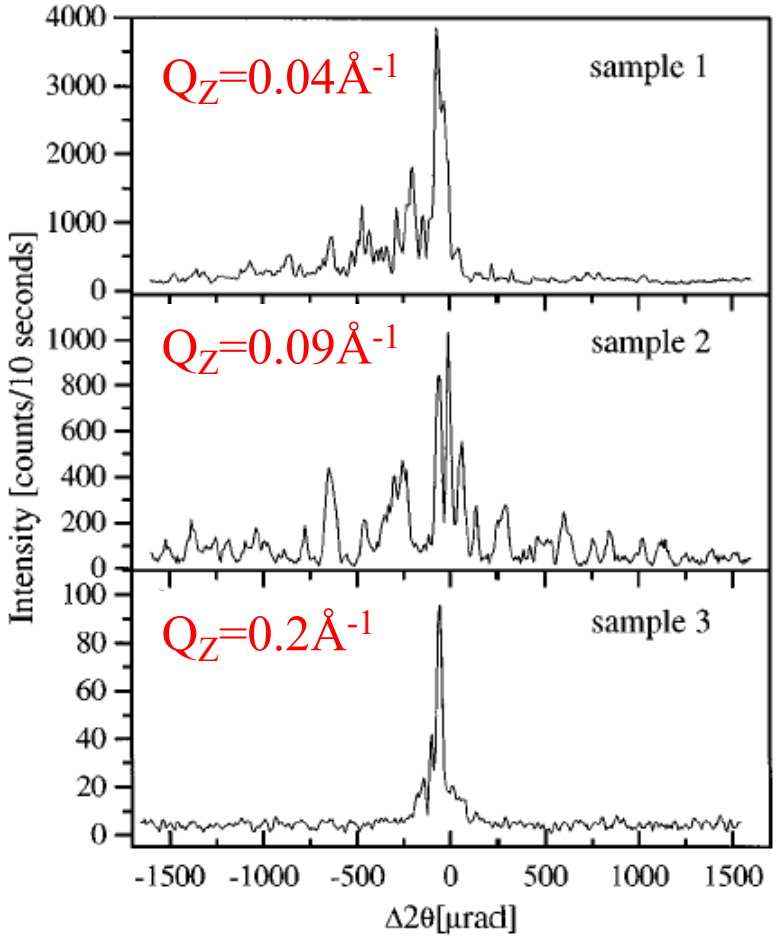
$$A(q_x, q_y) = \int_{-d/2}^{d/2} dx dy e^{iq_z h(x,y)} e^{iq_x x} e^{iq_y y}.$$



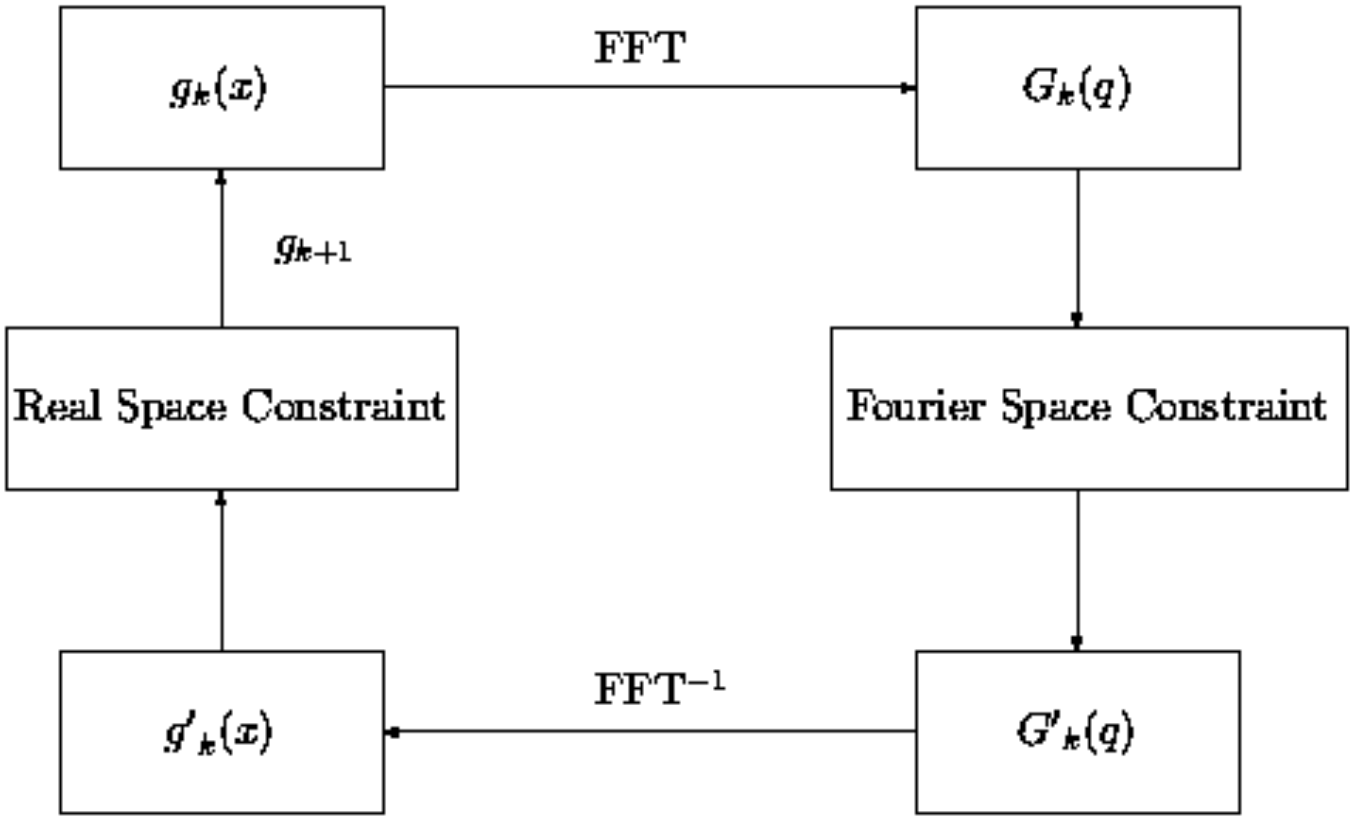
Si Samples of Different Roughness

J. L. Libbert *et al* Phys. Rev. B **56** 6454 (1997)





Generic "Error Reduction" method



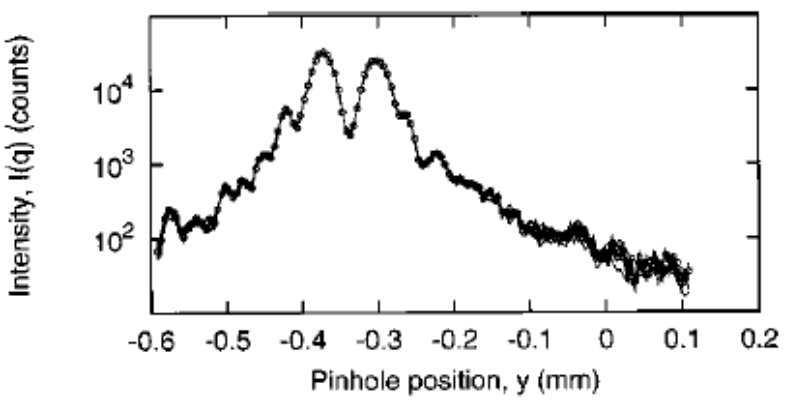
J. R. Fienup Appl. Opt. 21 2758 (1982)
R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)

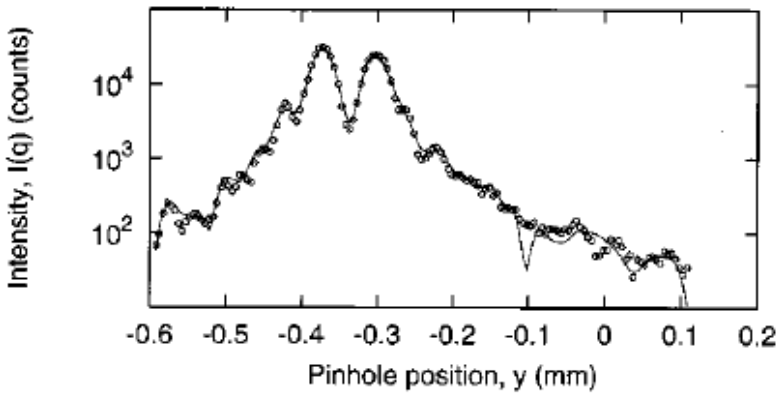
Real-space Constraints in Crystallograhy

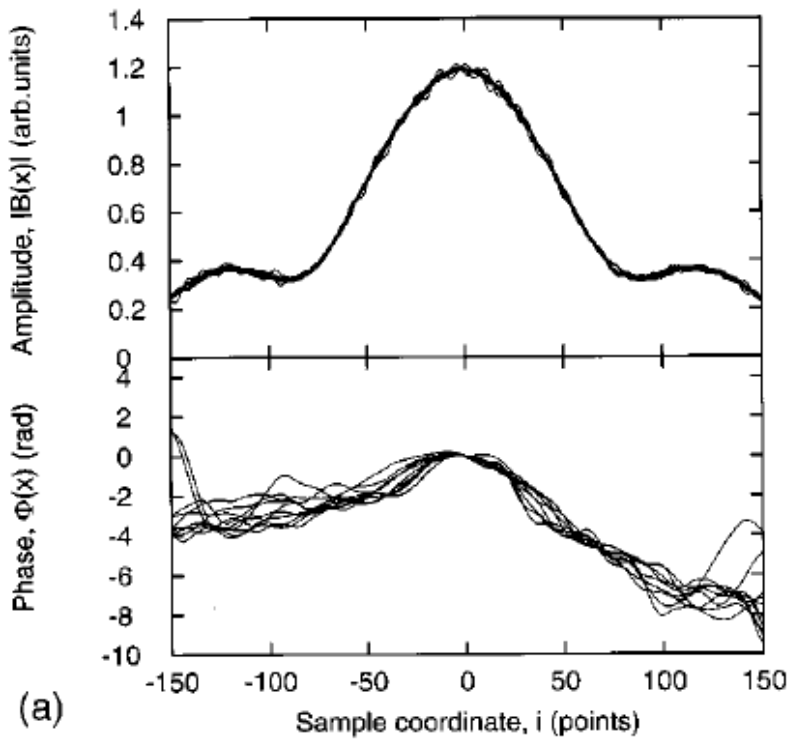
R. P. Millane, J. Opt. Soc Am. A **13** 725 (1996)

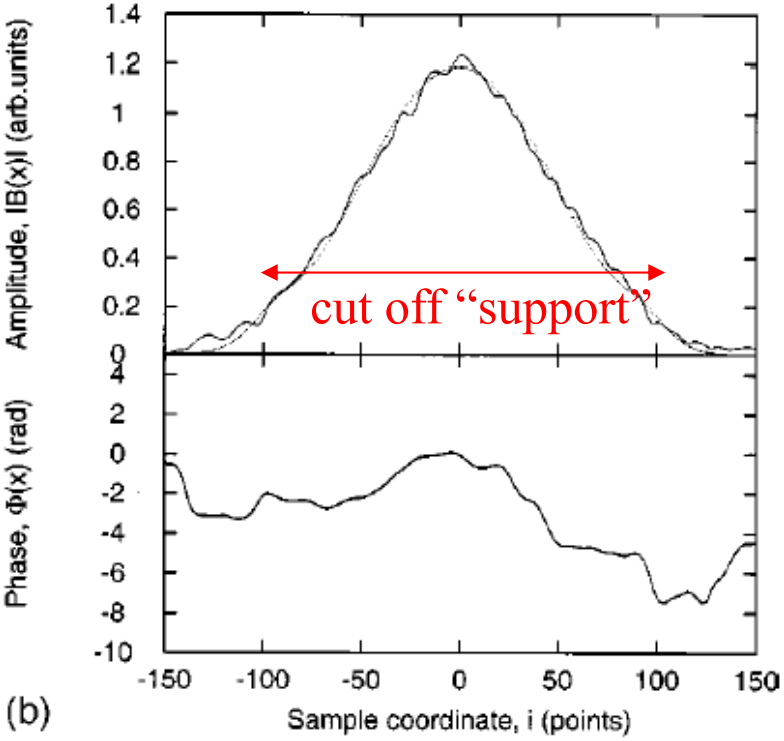
- 'Positivity' constraint (Sayre)
- Finite support, molecular envelope
- Solvent flattening
- Molecular replacement
- Non-crystallographic symmetry
- Non-uniqueness is 'pathologically rare' (d>1)

Phasing using G-S Algorithm

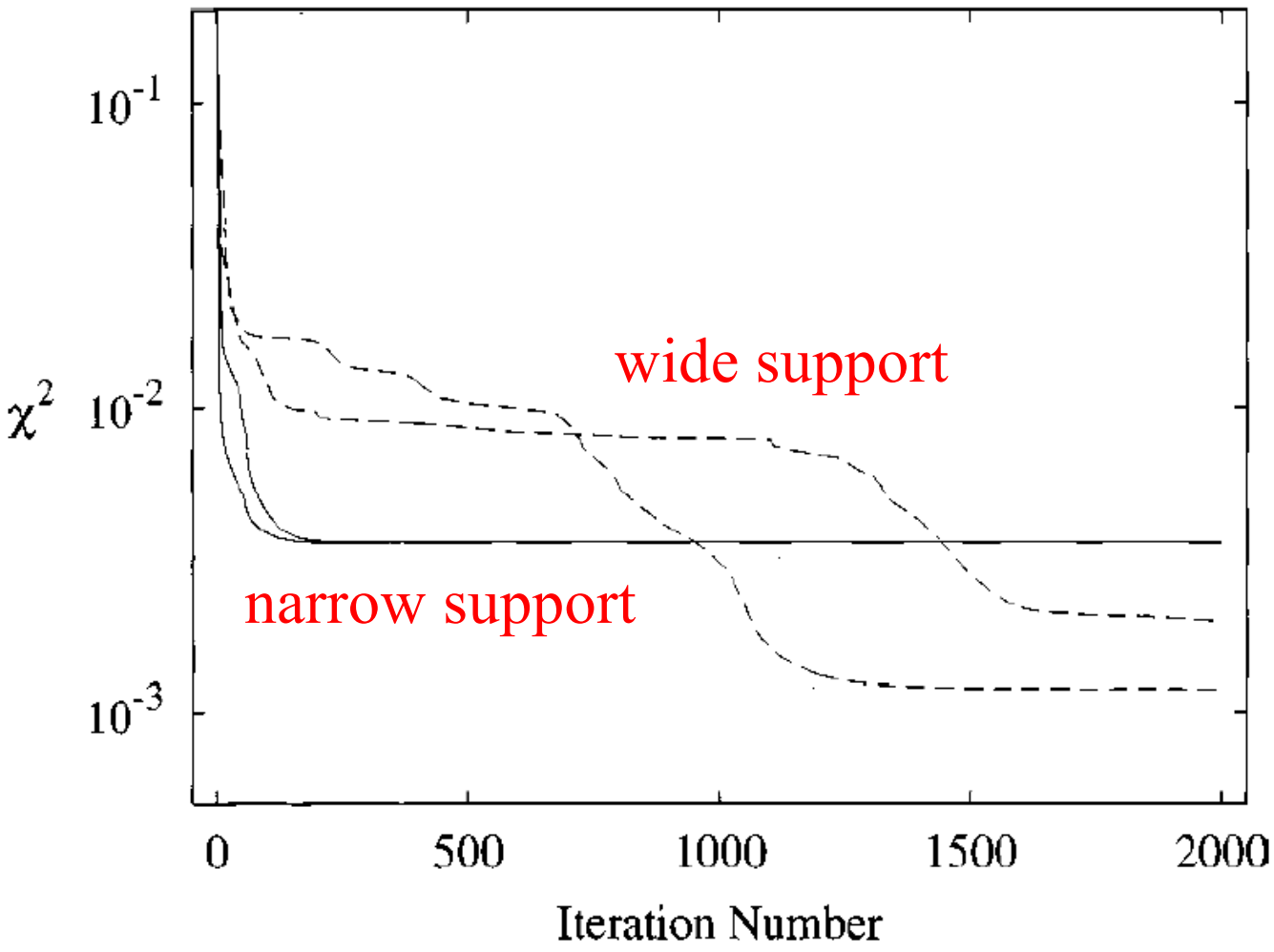








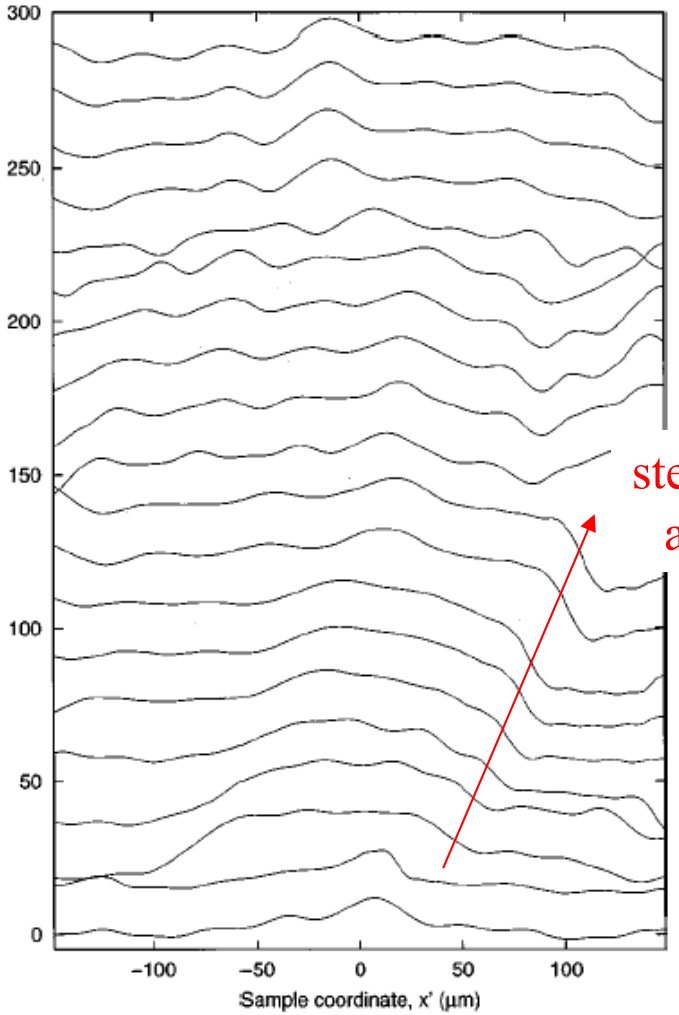
Convergence Trajectory



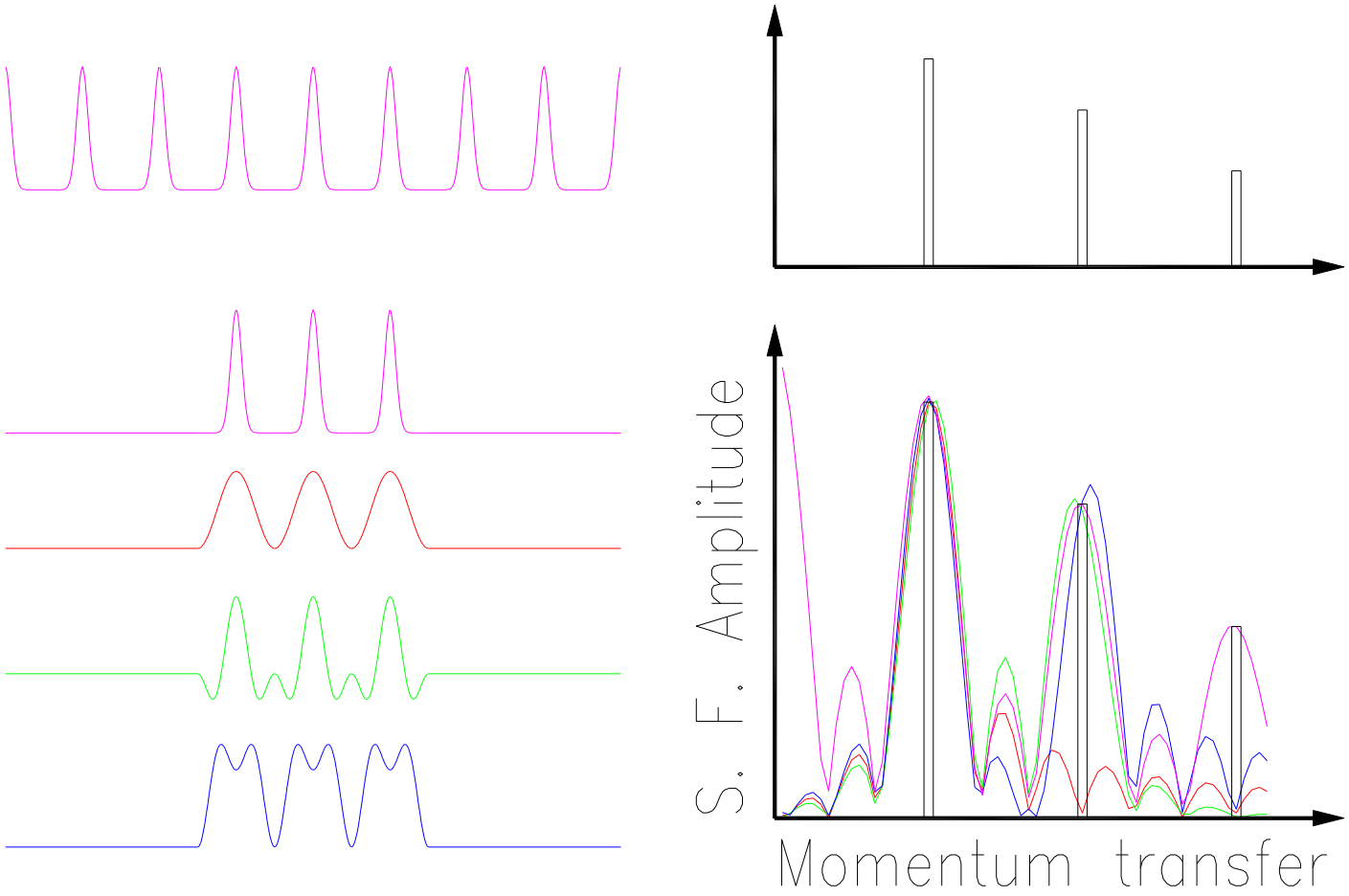
In-situ Regrowth of Oxide on Si

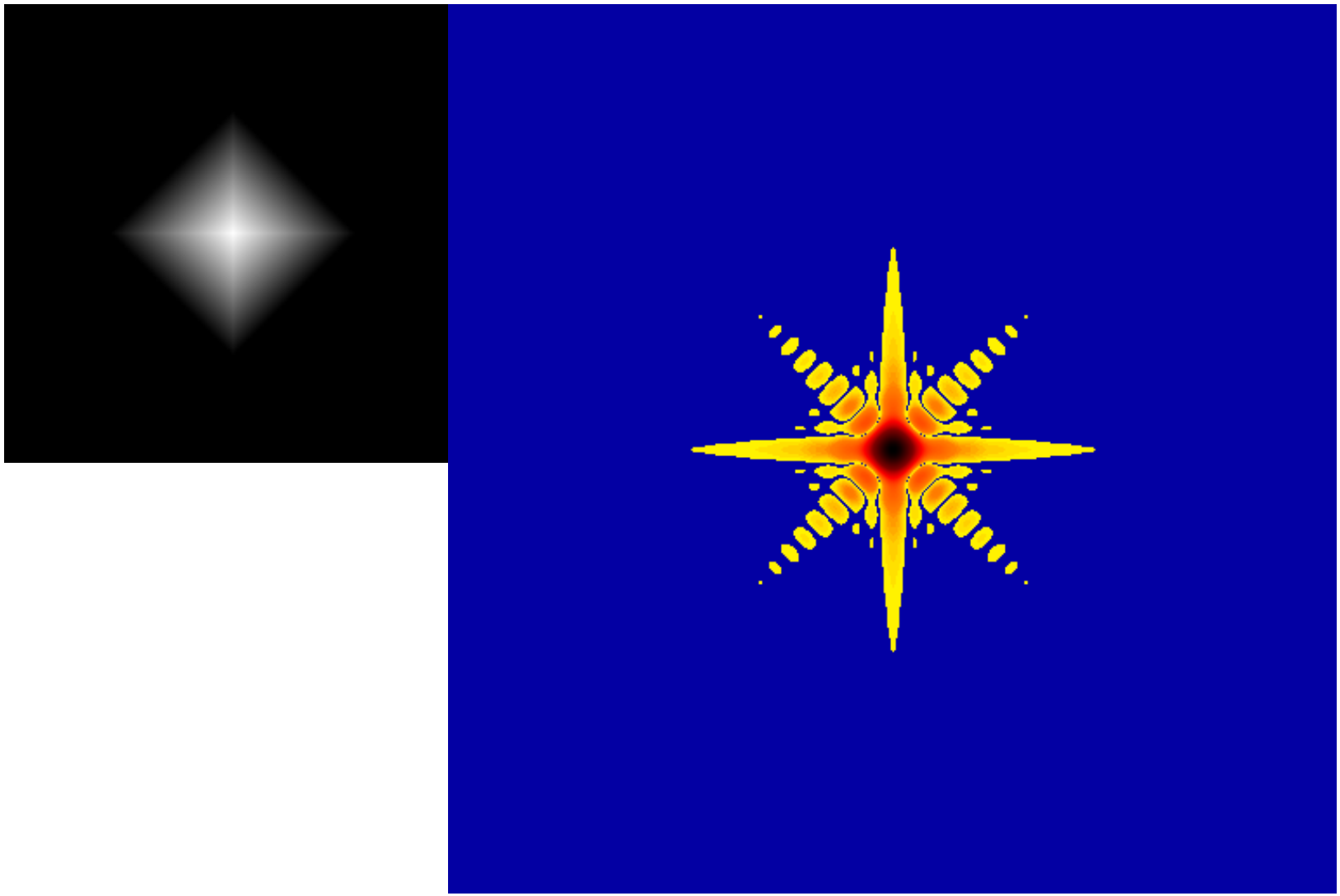
Phys. Rev. B **60** 9965 (1999)

- Wafer stripped with HF at t=0
- CXD pattern measured every 180 sec
- Each profile reconstructed independently
- Random starting phase each time

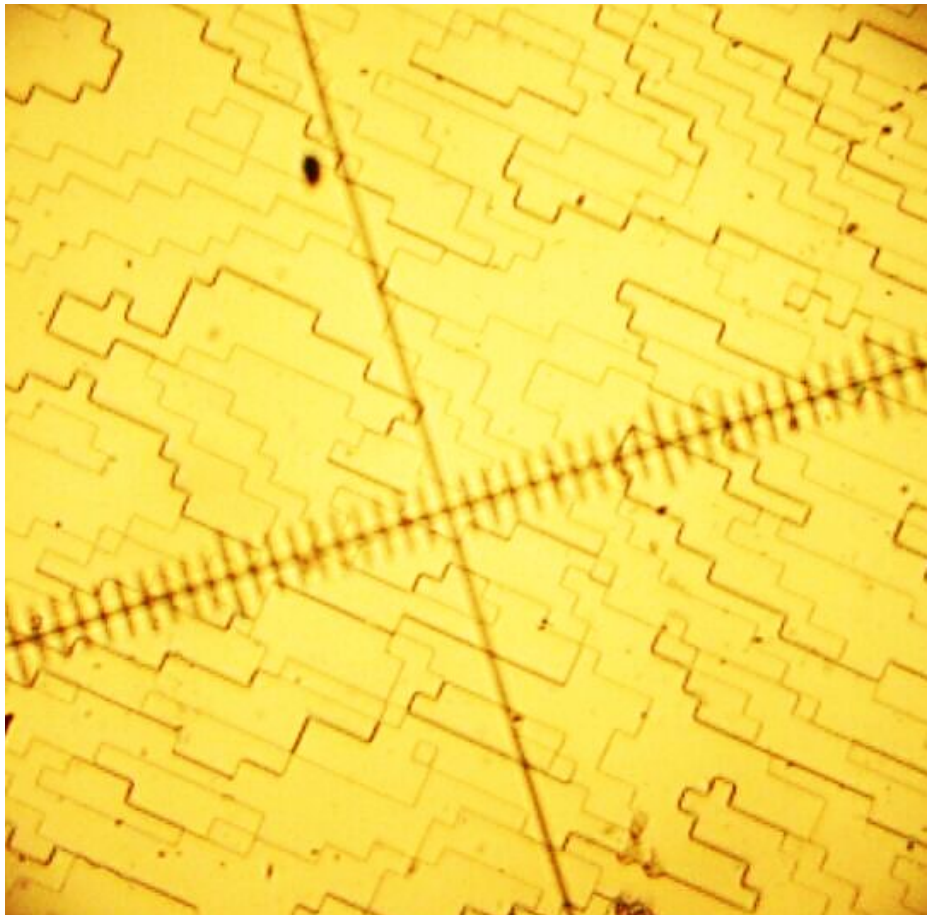


Phase Problem: Finite-size Effect

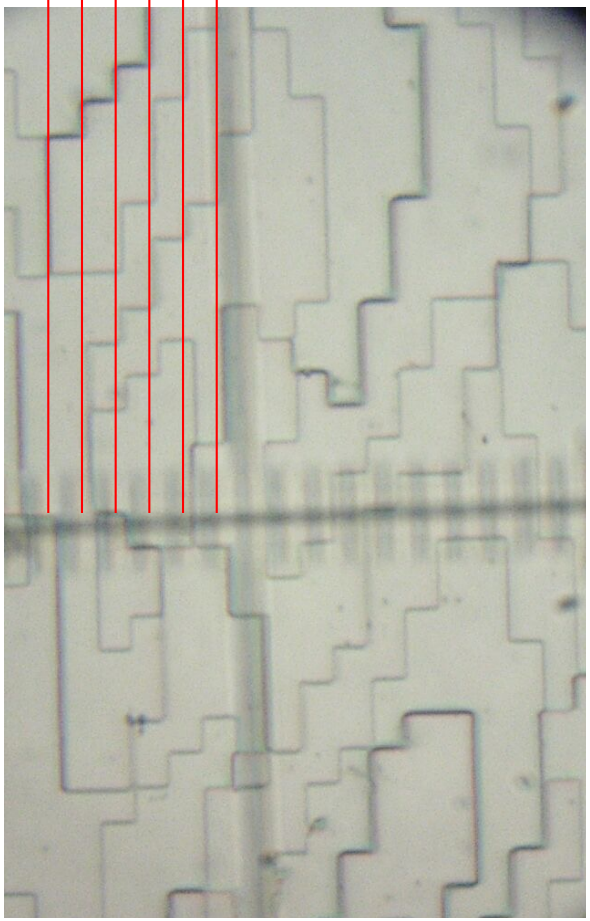


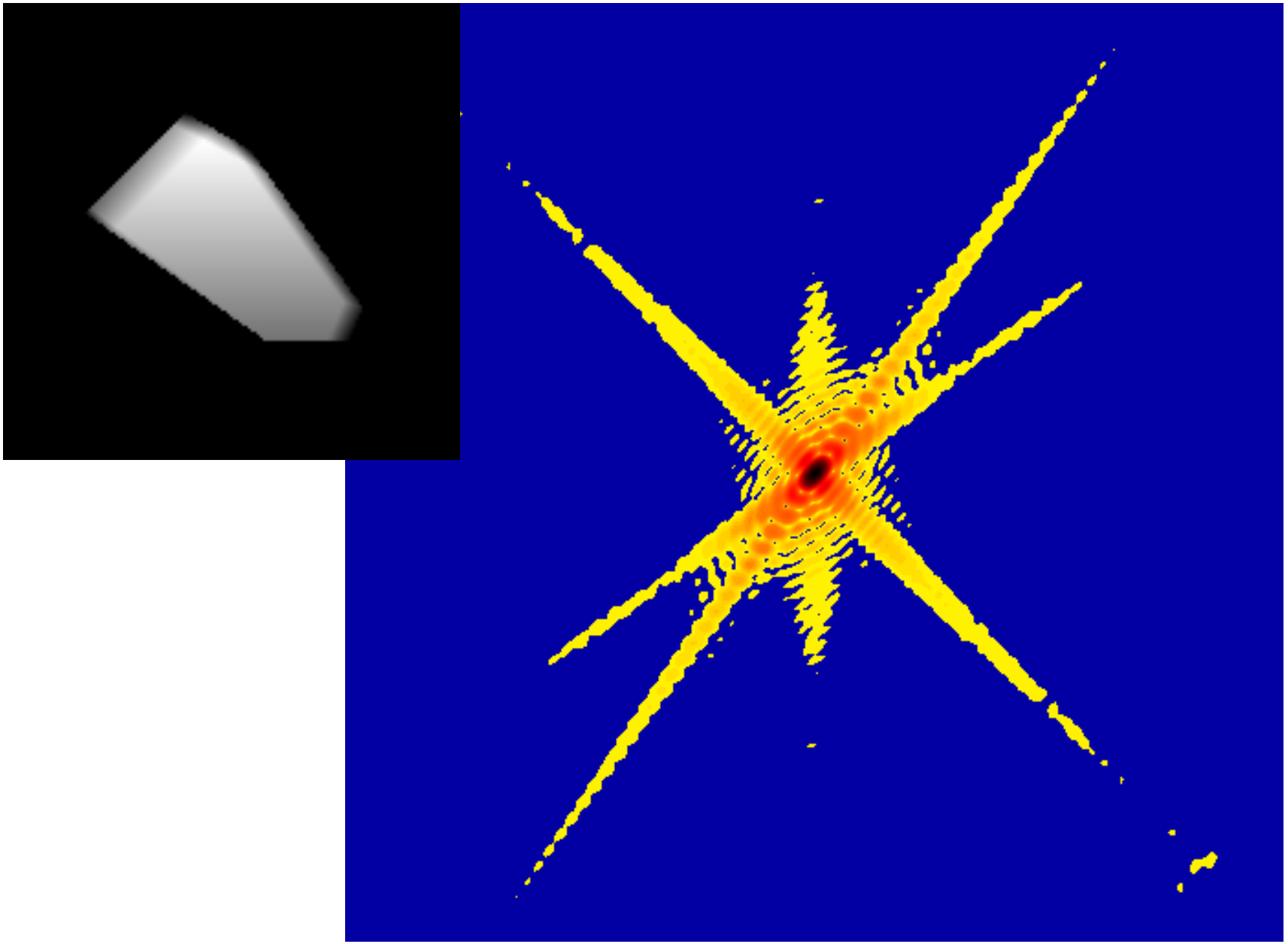


Microscope Images of Gratings



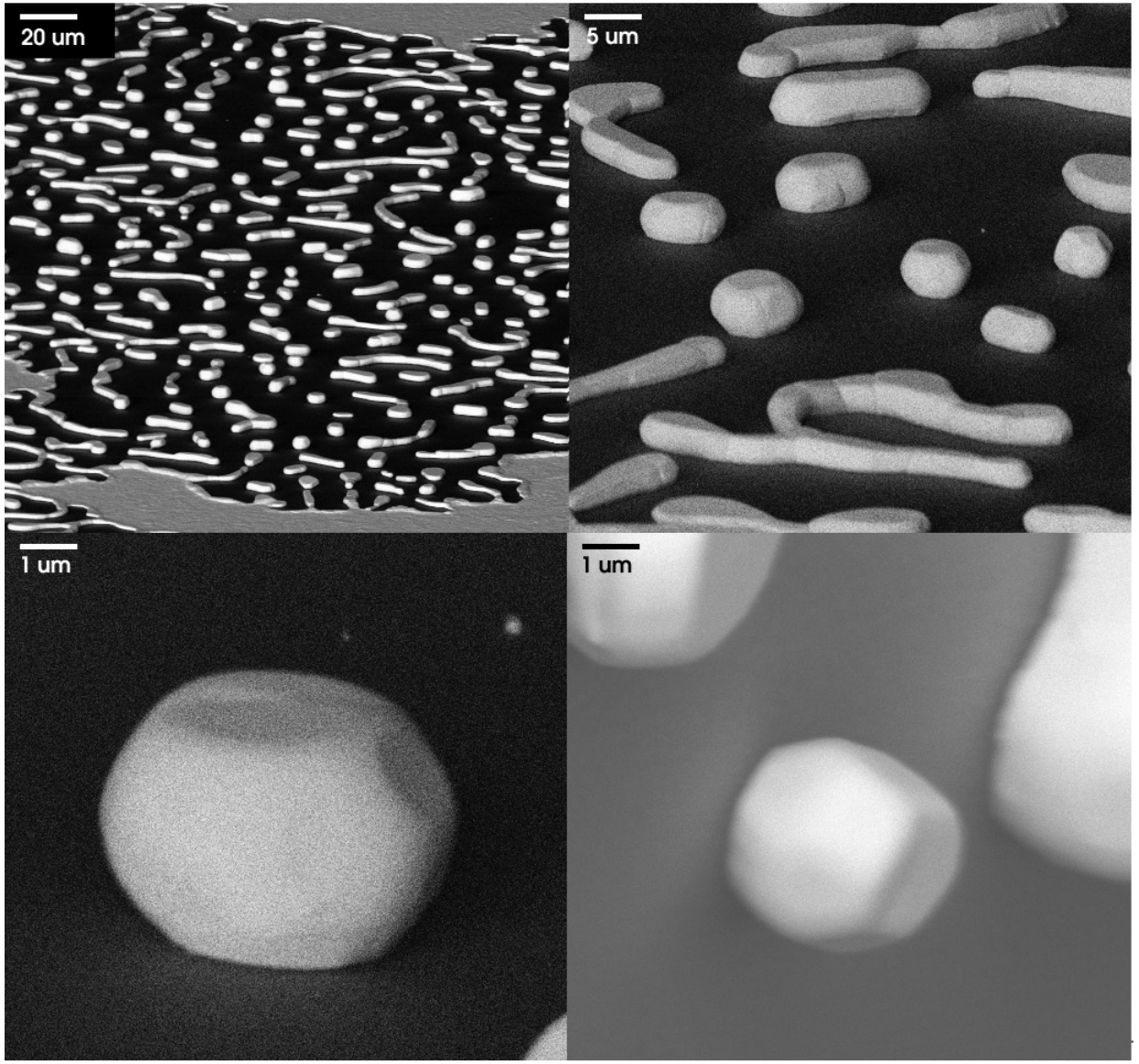
5μm grid lines



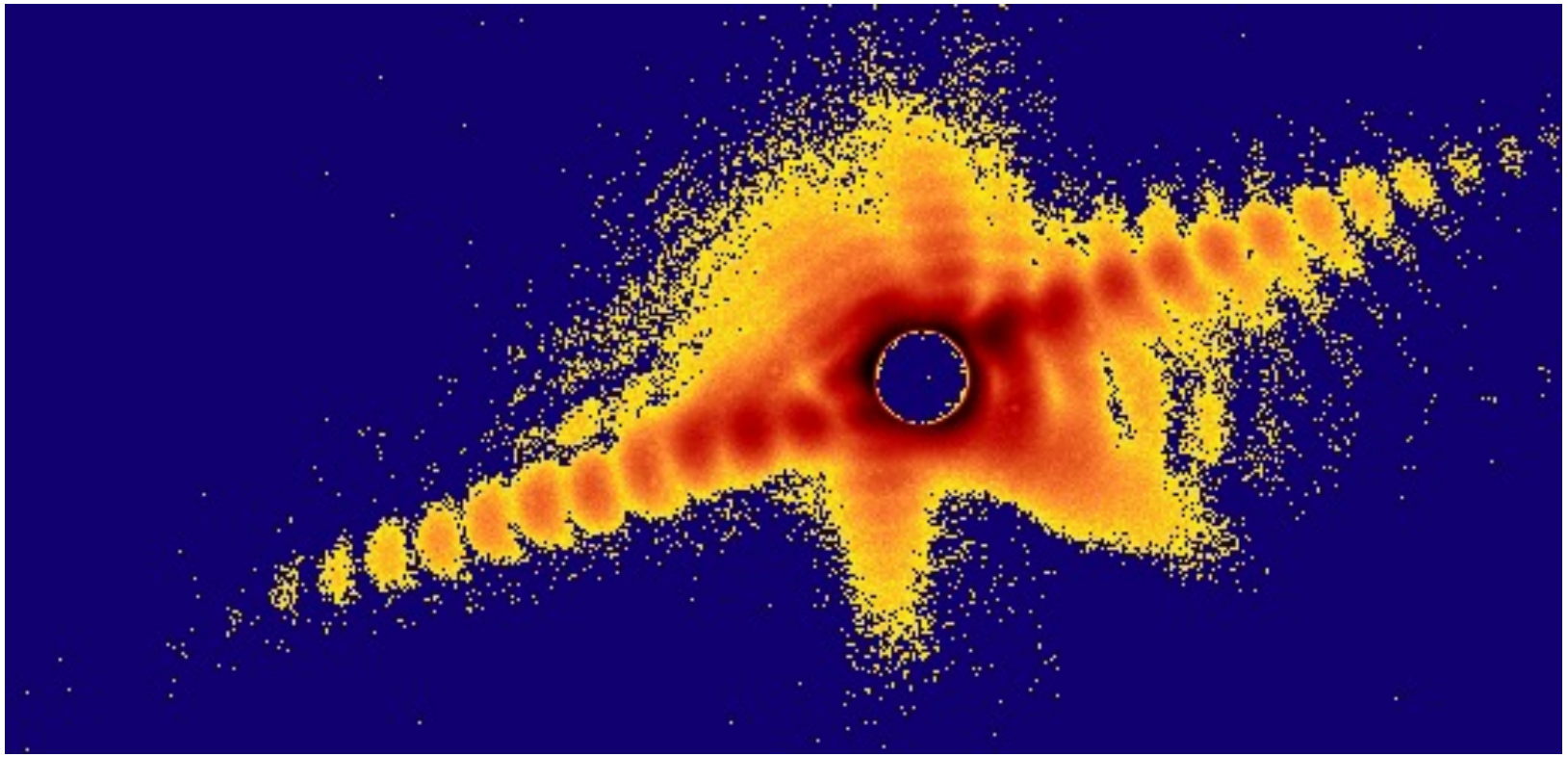


SEMS

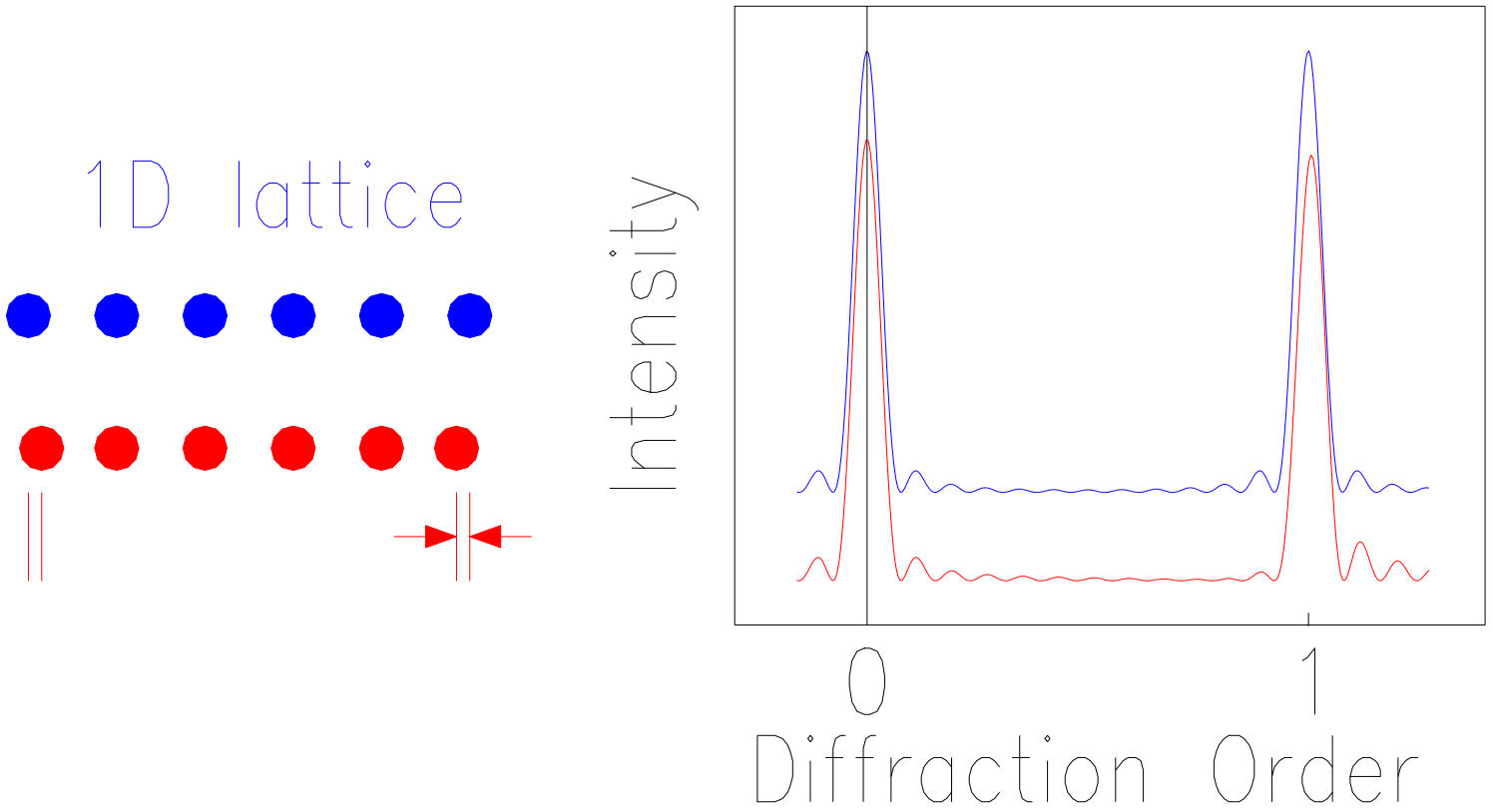
- Au blanket film
- Quartz substrate
- Annealed at 950°C for 70 hrs.



Micron-sized gold crystal: (111) Bragg reflection

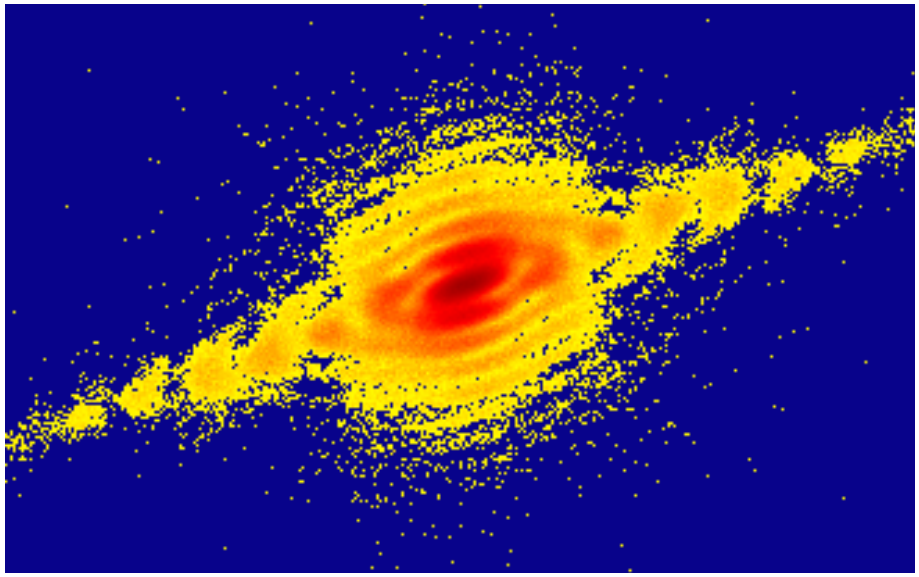


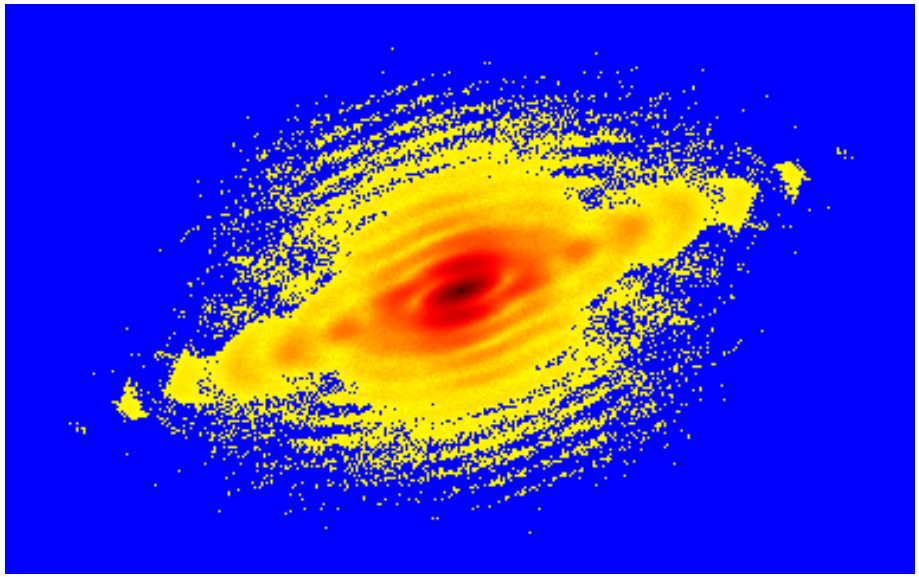
Imaging of Lattice Strains

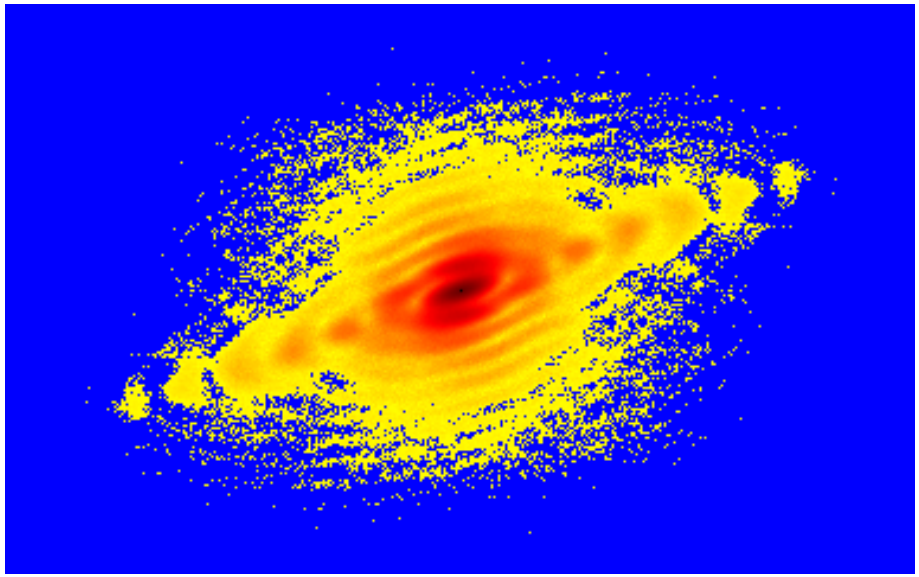


Symmetrized Data and two best fits

Chisq=0.0005

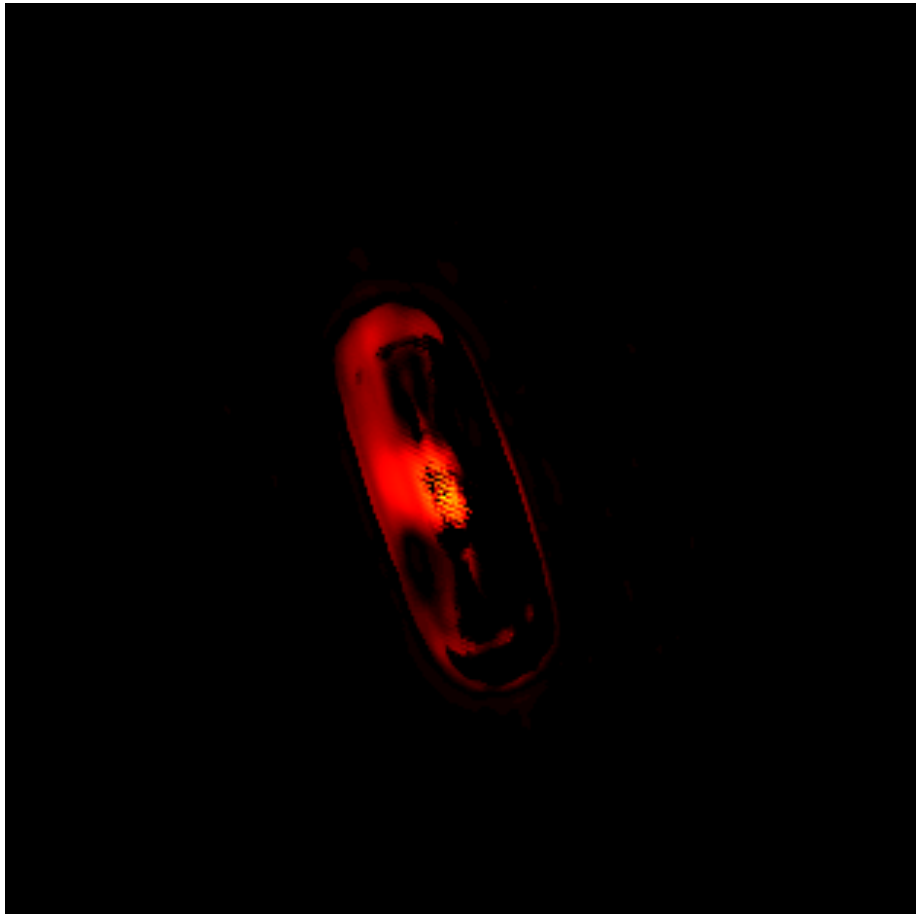


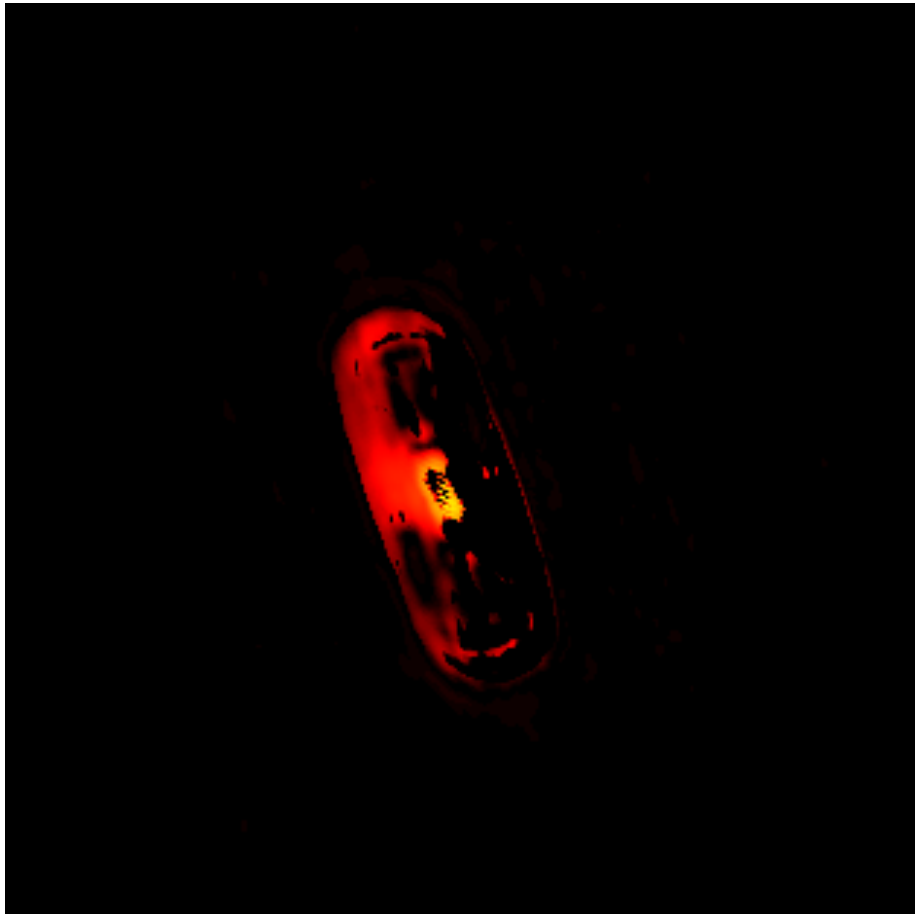




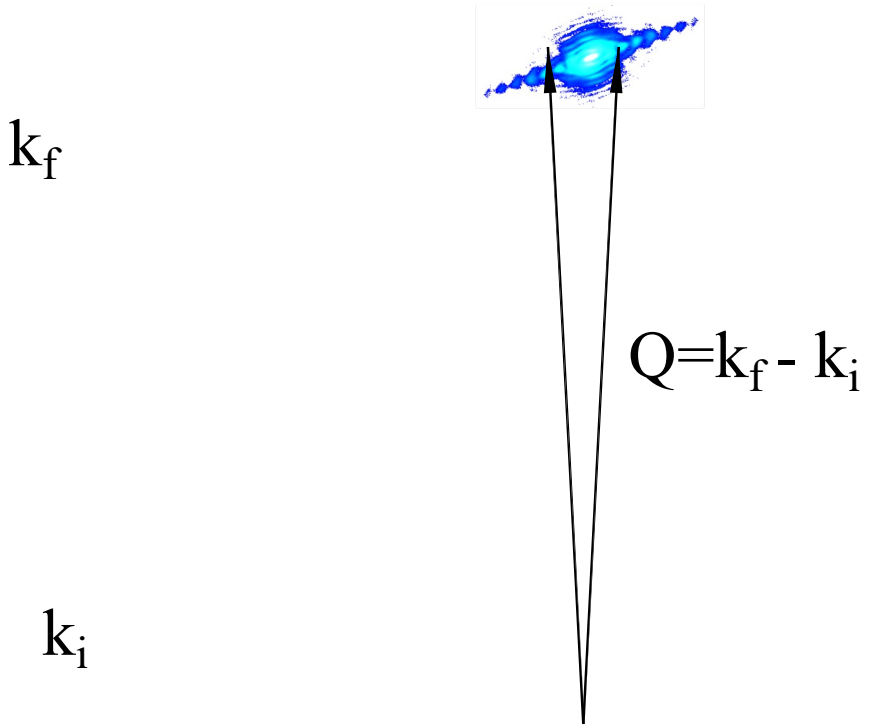
2D Reconstructions

chisquare = 0.0005

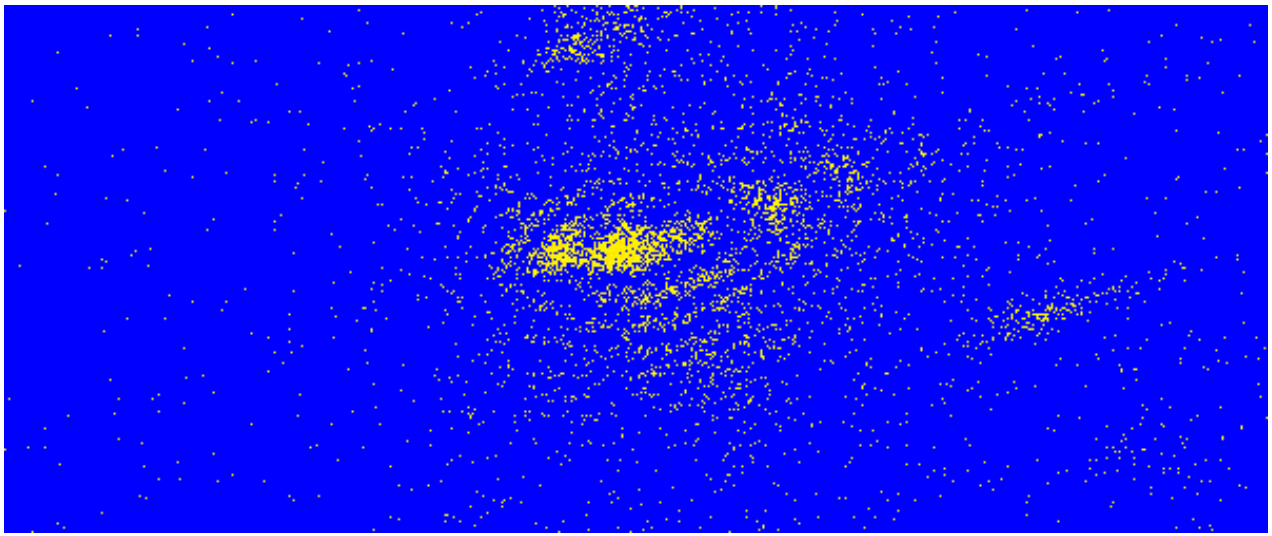




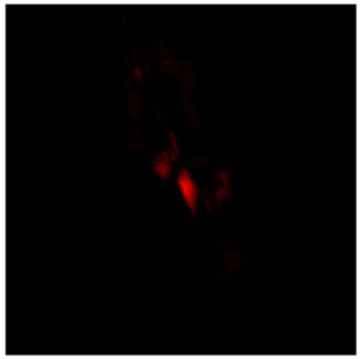
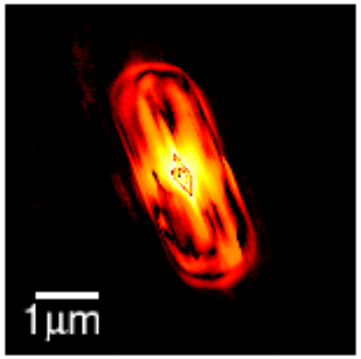
3D Diffraction Method

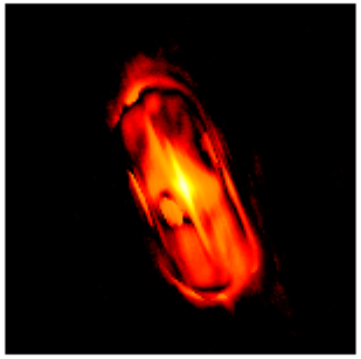
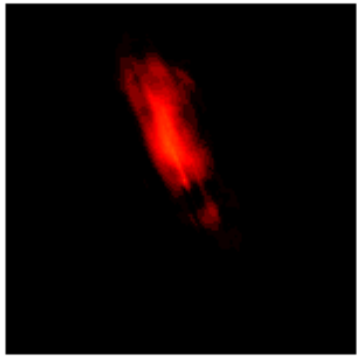
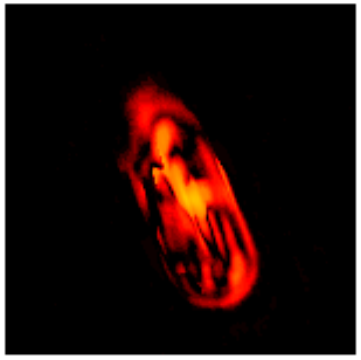
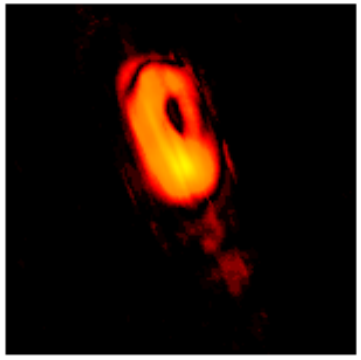
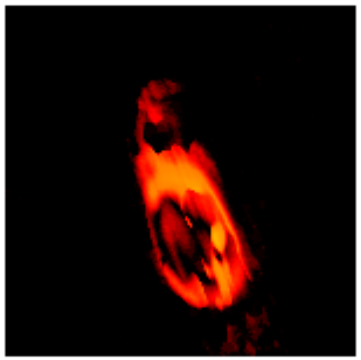
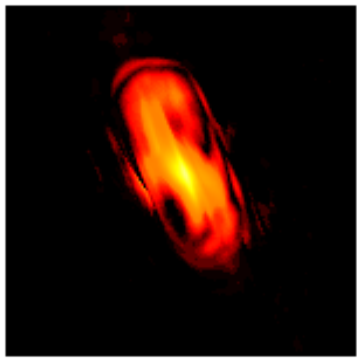
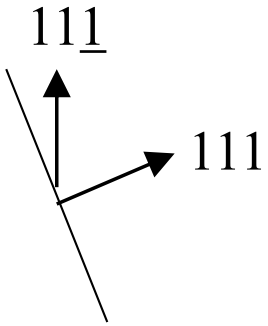


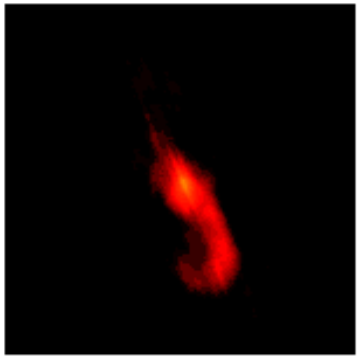
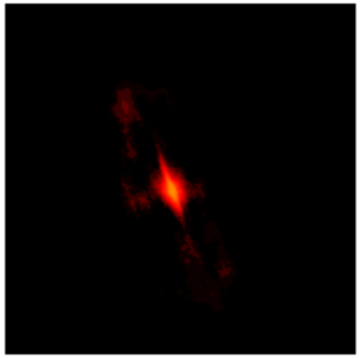
3D Diffraction Data
1 micron Au crystal



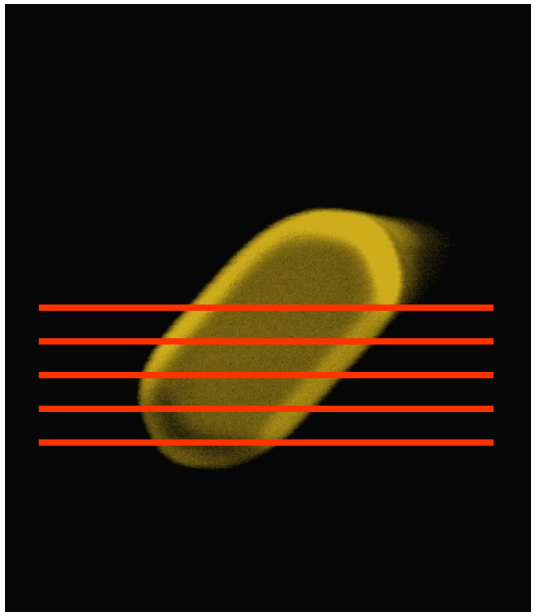
* Center is Symmetric *

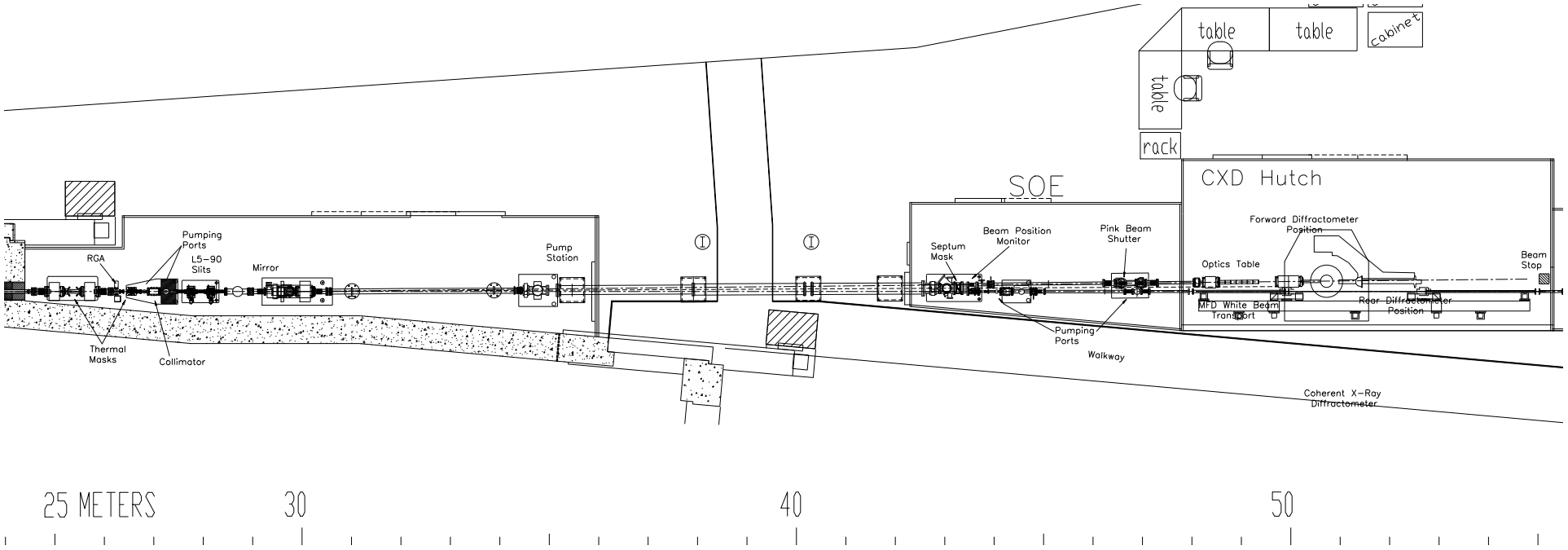








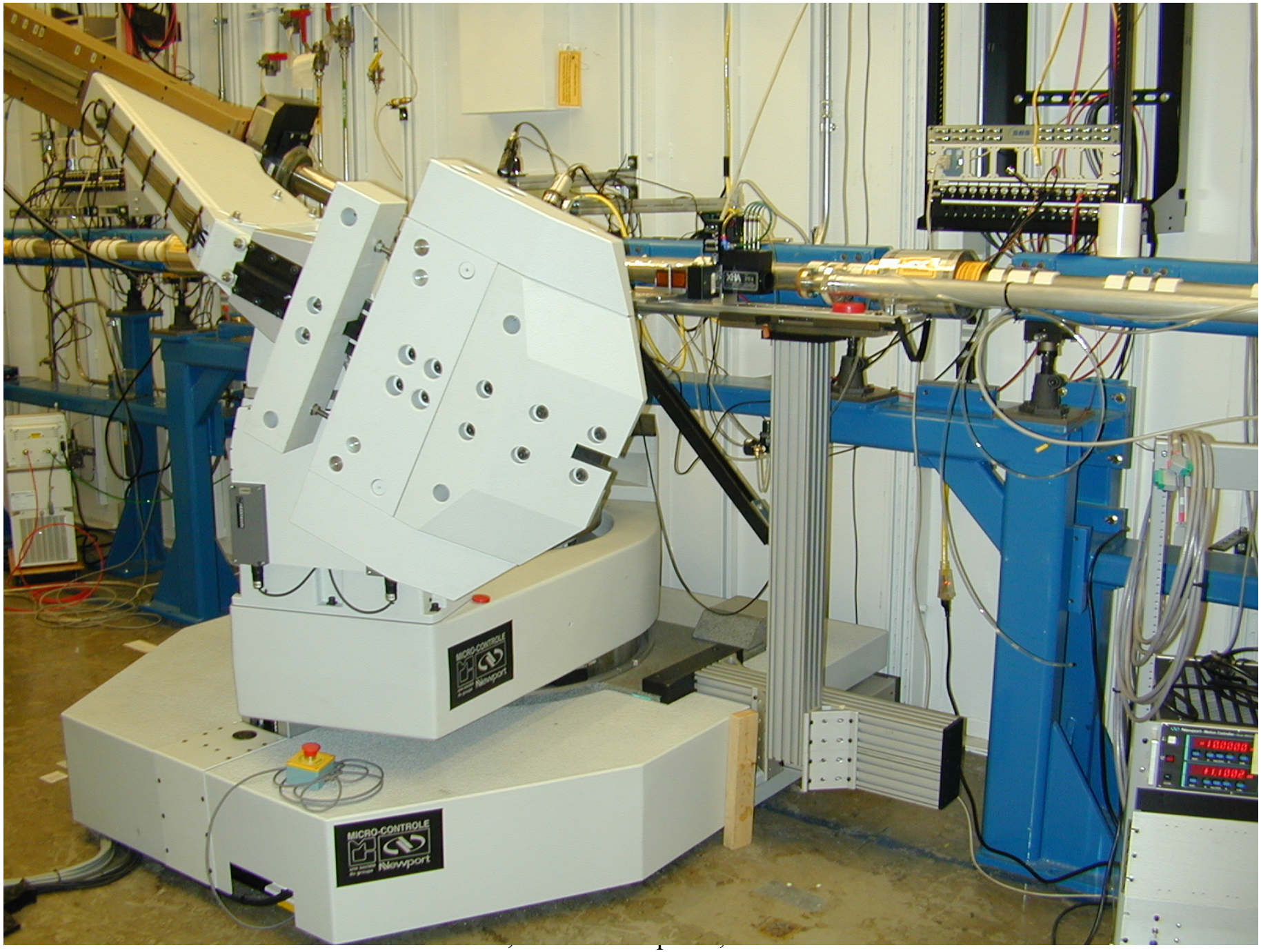



Slices through plan view SEM:

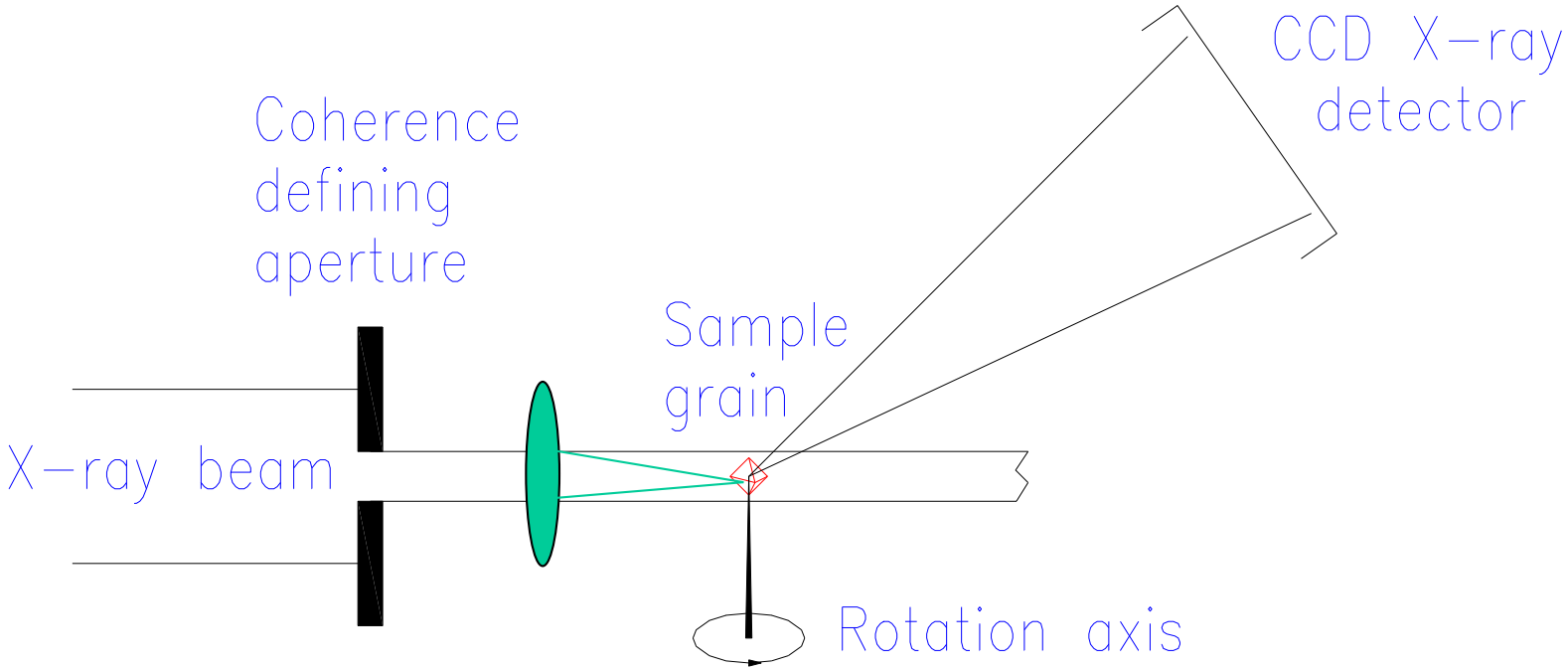


CXD Beamline at APS Sector 34

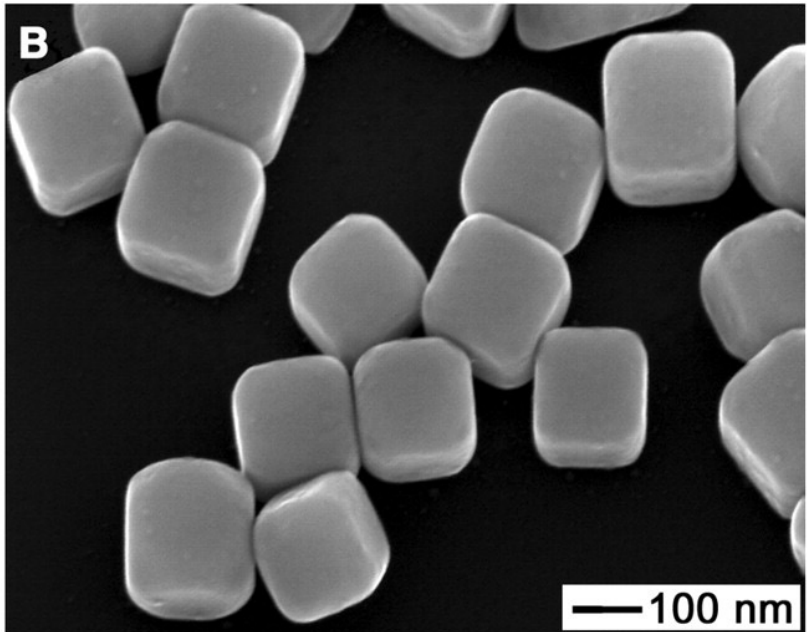


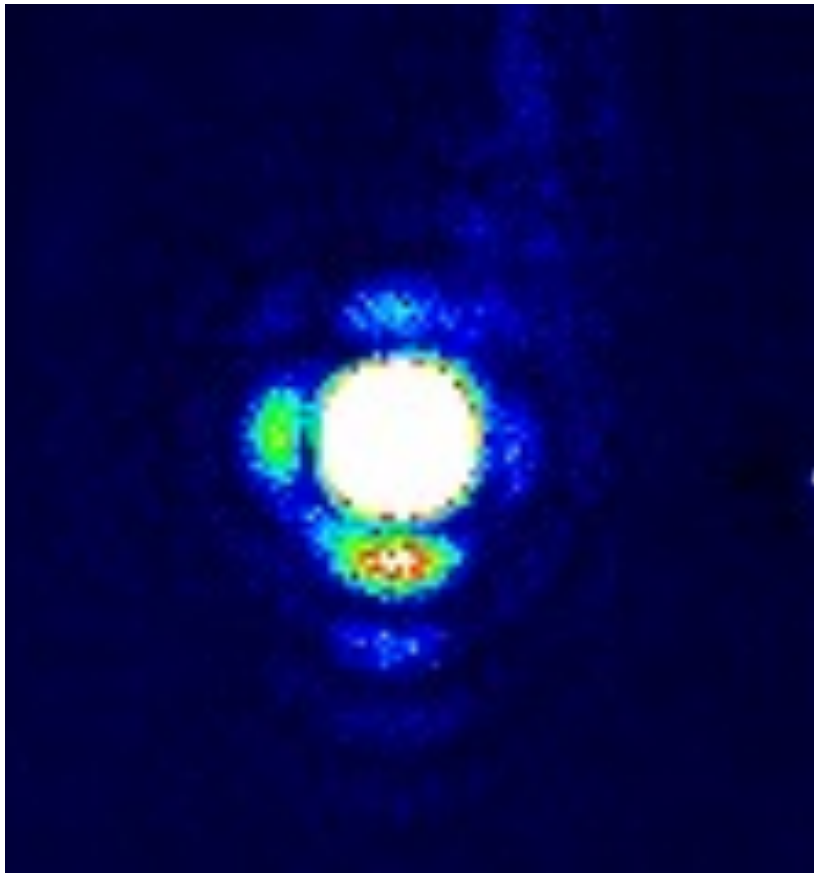

MICRO-CONTROLE
Newport
MICRO-CONTROLE
Newport

Lensless X-ray Microscope



CXD from Silver Nanocubes

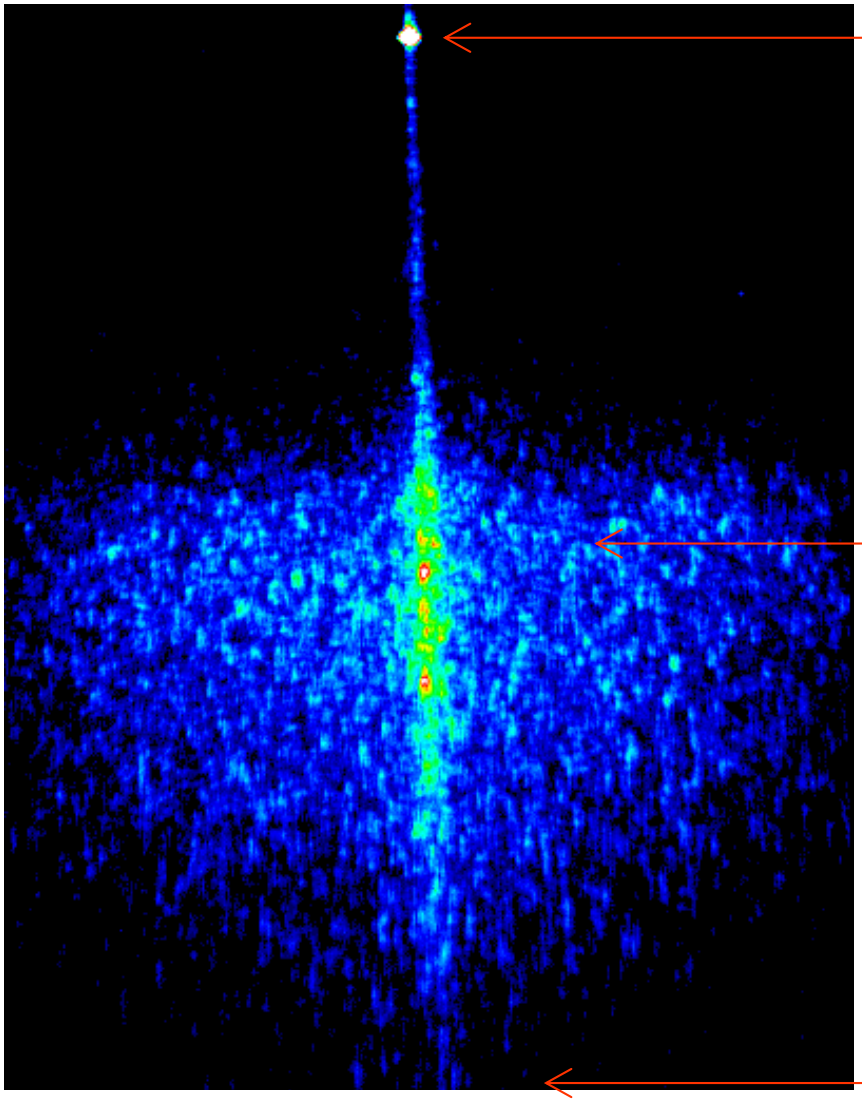




Yugang Sun and Younan Xia,
Science 298 2177 (2003)

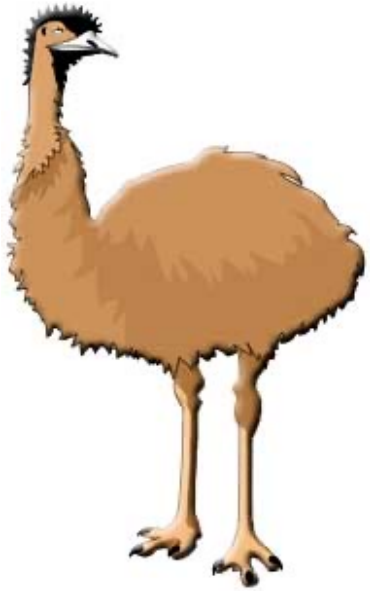
Structure in "Yoneda" Peak

Grazing-exit diffraction from a 1000A Au polycrystalline film



Specular ($\alpha_f \sim \alpha_i$)

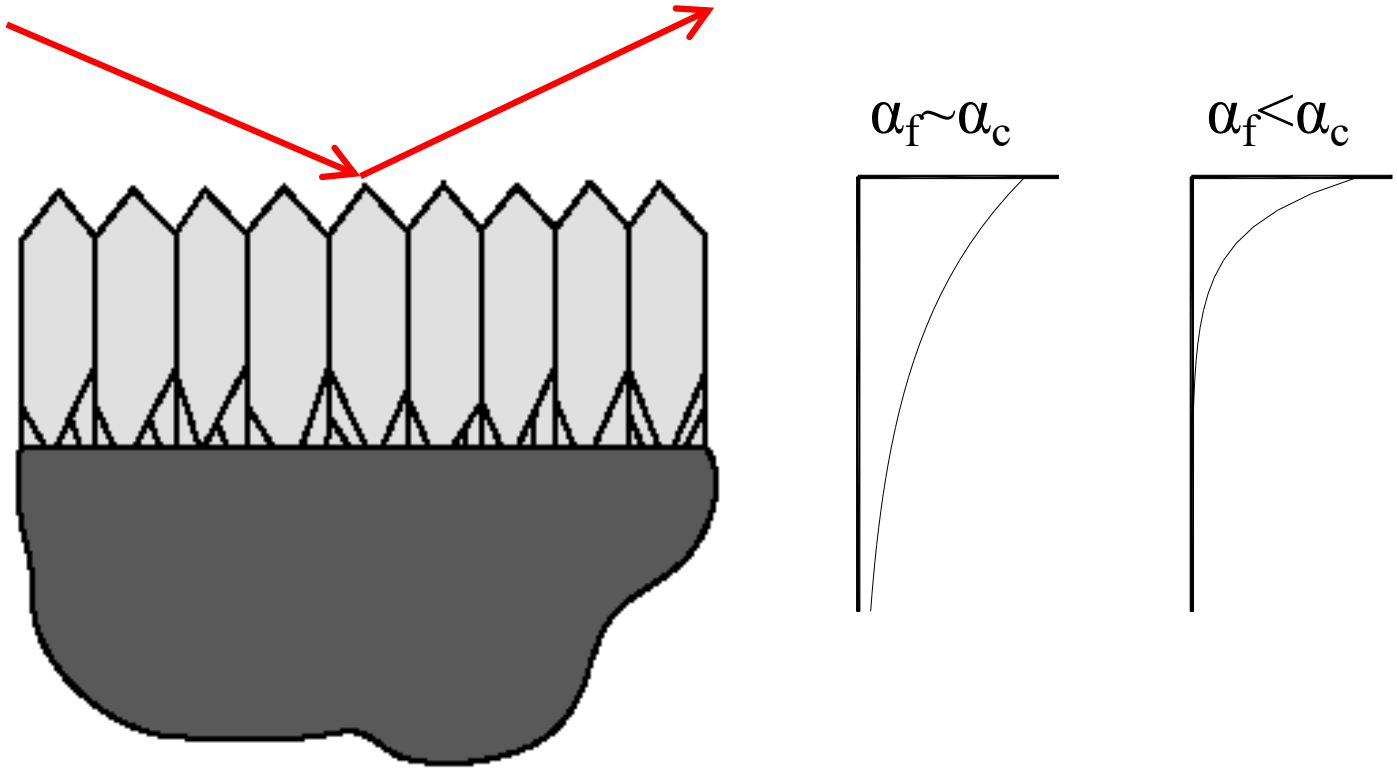
$\alpha_f \sim \alpha_c$



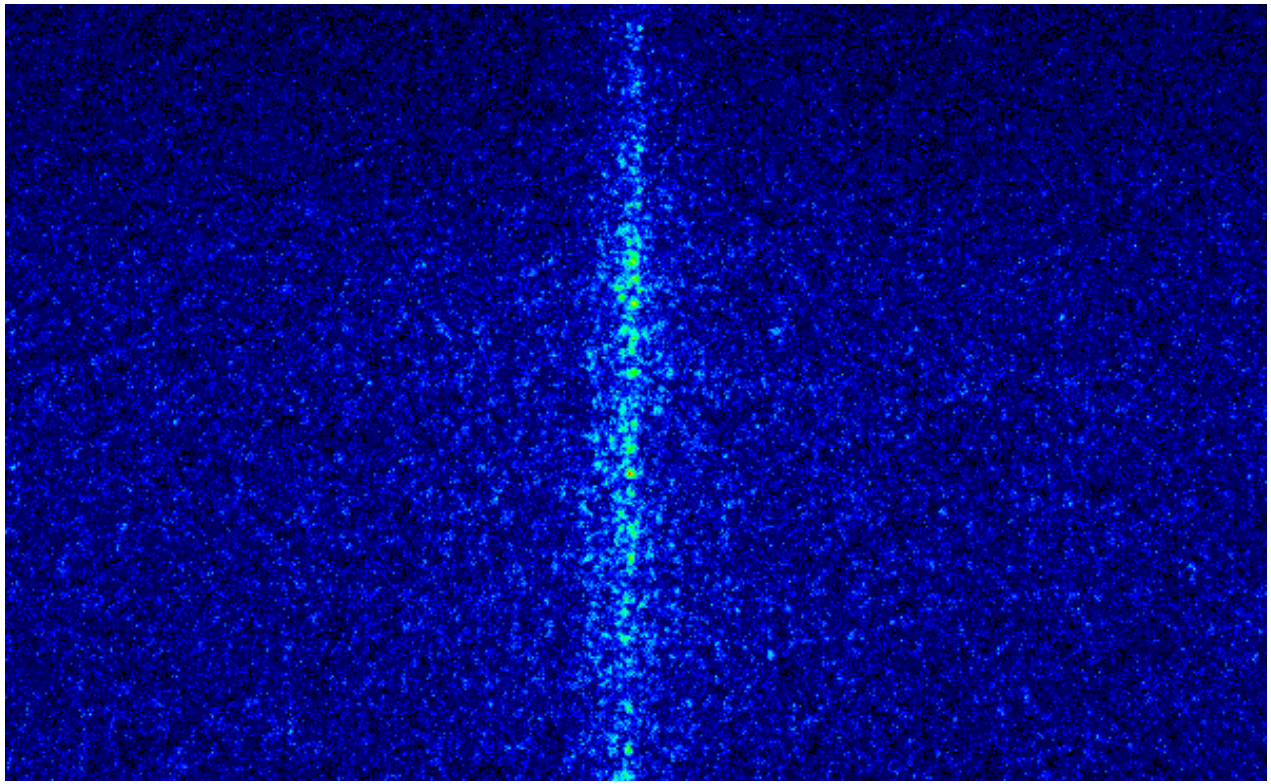
$\alpha_f \sim 0$

Competitive Grain Growth

C. V. Thompson, Ann. Rev. Mat. Sci. **30** 159 (2000)



Angle series, 0.01° steps

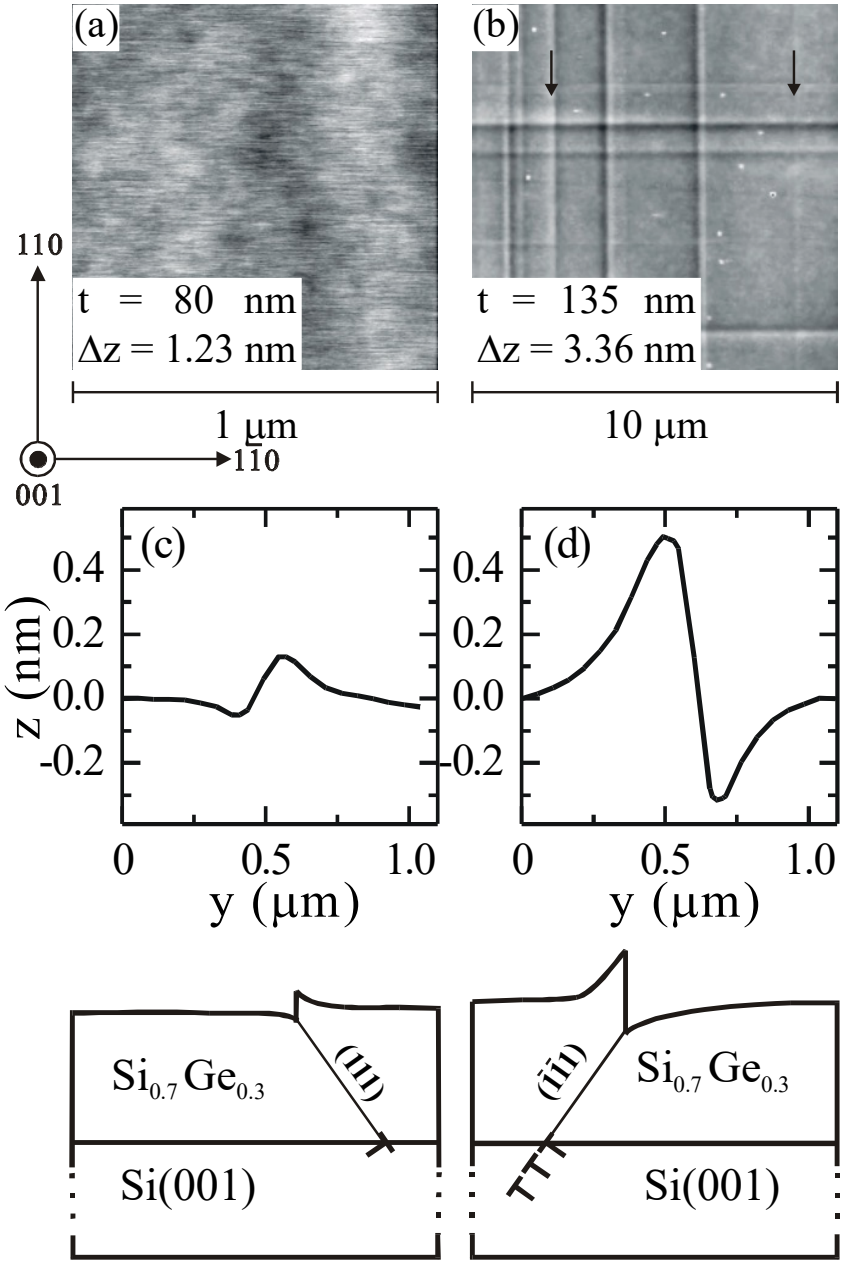


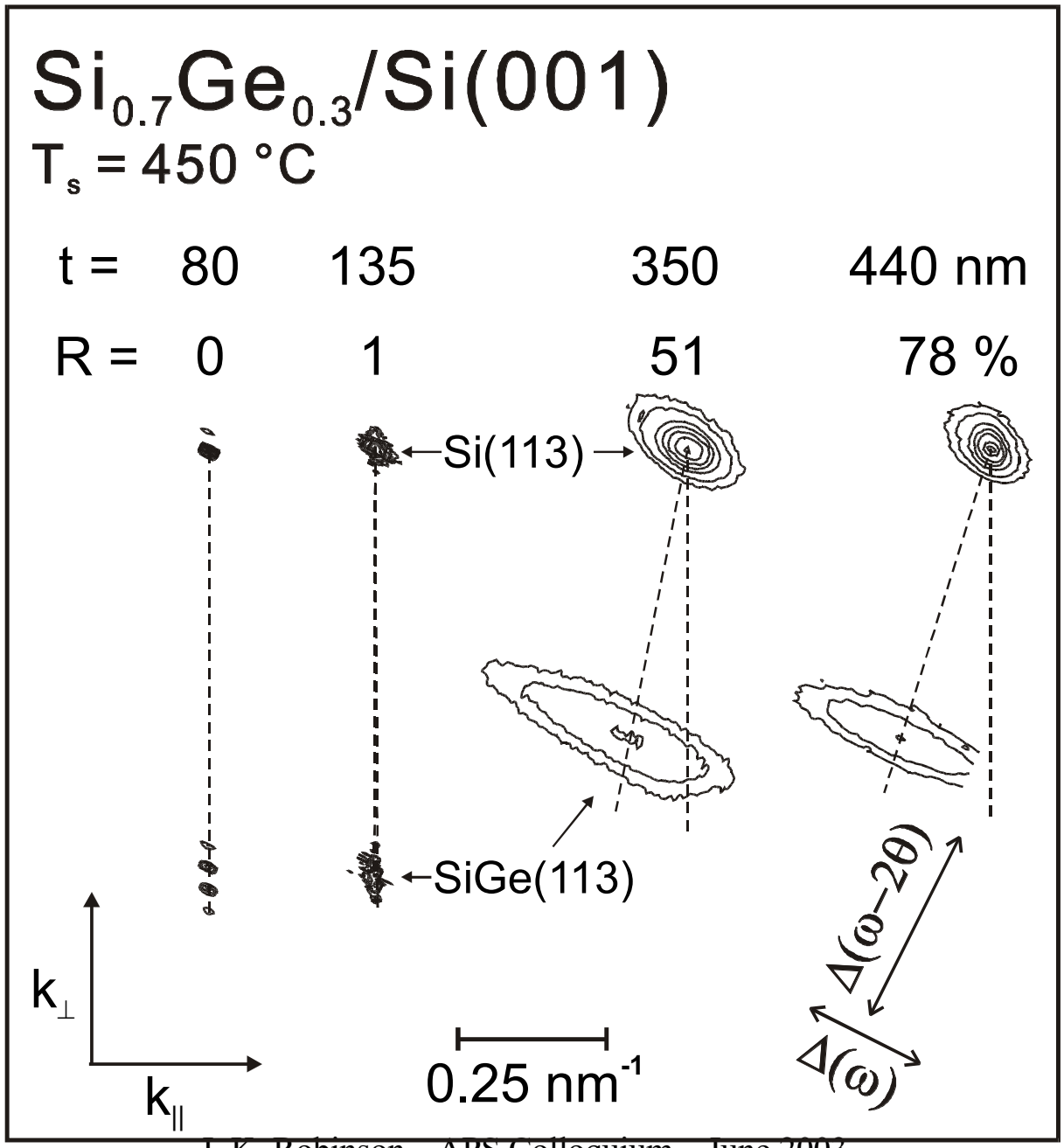
Low dislocation density GeSi films

Thickness close to critical thickness

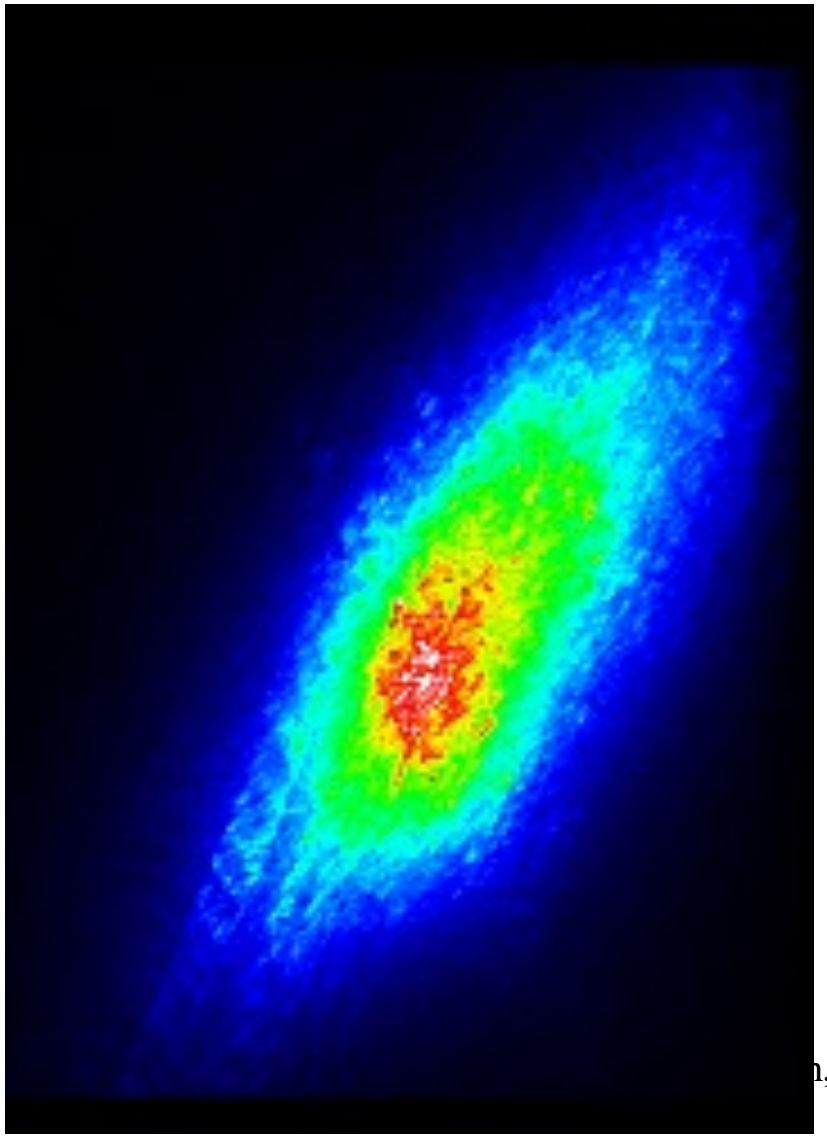
Dislocations aggregate at interface and glide to surface along {111}

T. Spila, UIUC Thesis




$Si_{0.7}Ge_{0.3}/Si(001)$
T_s = 450 °C
t = 80 135 350 440 nm
R = 0 1 51 78 %
Si(113)
SiGe(113)
$k_\perp$
$k_\parallel$
0.25 nm^{-1}
$\Delta(\omega-2\theta)$
$\Delta(\omega)$

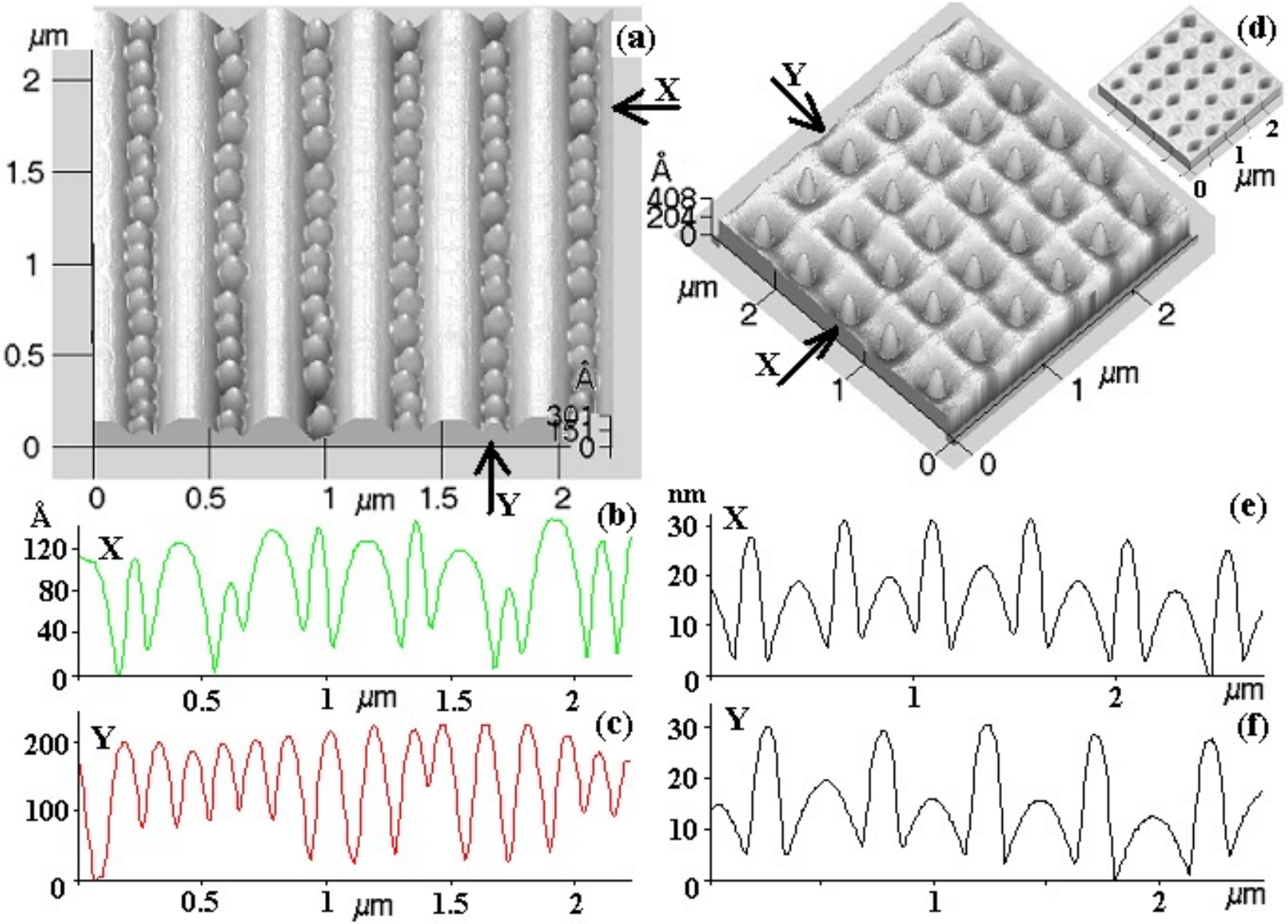
Ge_xSi_{1-x} Film Diffraction



- 202 Bragg Peak
- 2800A film
- 2° incidence angle
- 8.5 keV
- 20μm ×40μm beam onto KB mirror
- 1μm ×1μm focus
- 0.5μm sample steps
- APS 34-ID-C

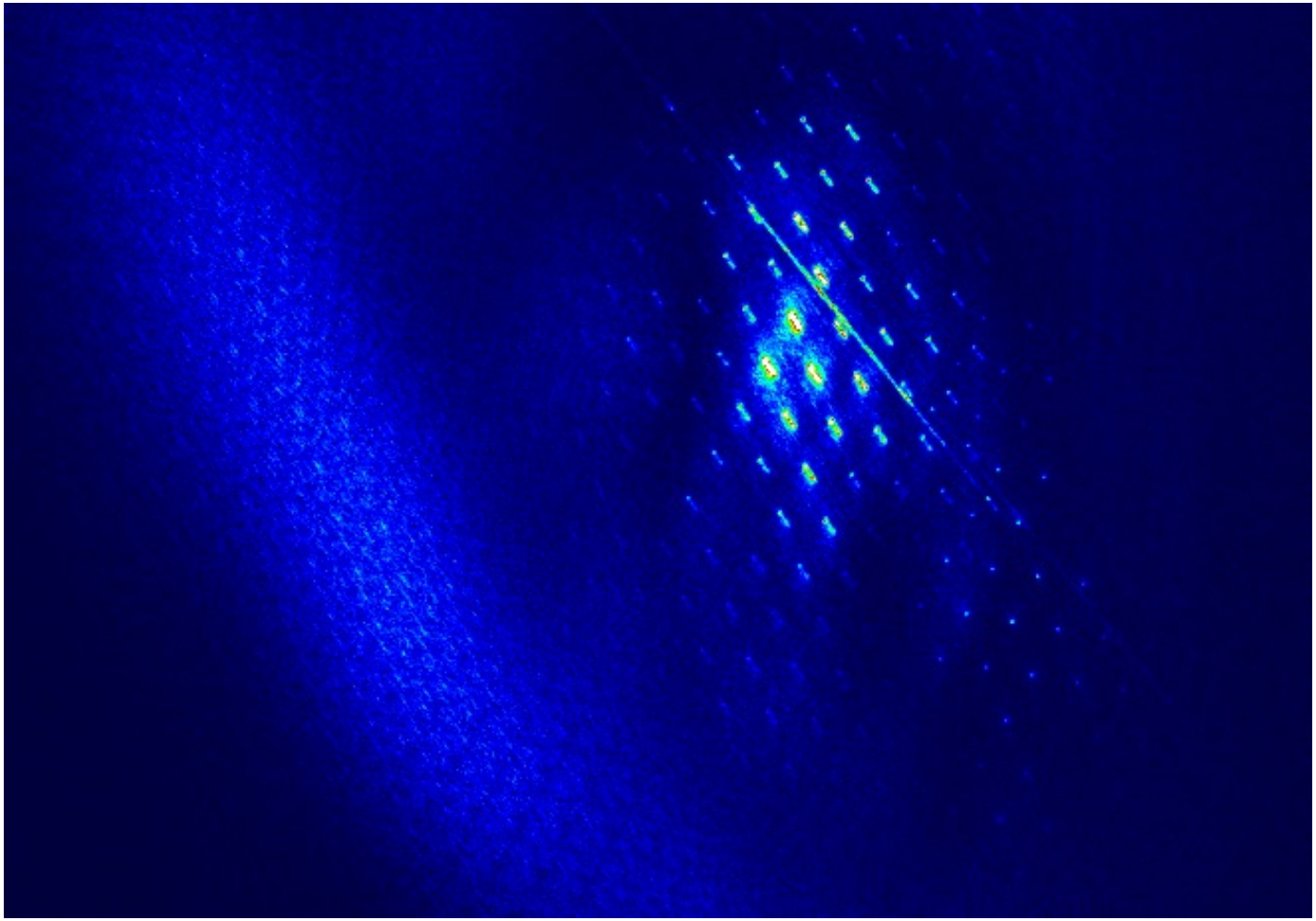
1D and 2D Quantum Dot Arrays

Zhenyang Zhong, G. Bauer, Johannes Kepler Universität Linz

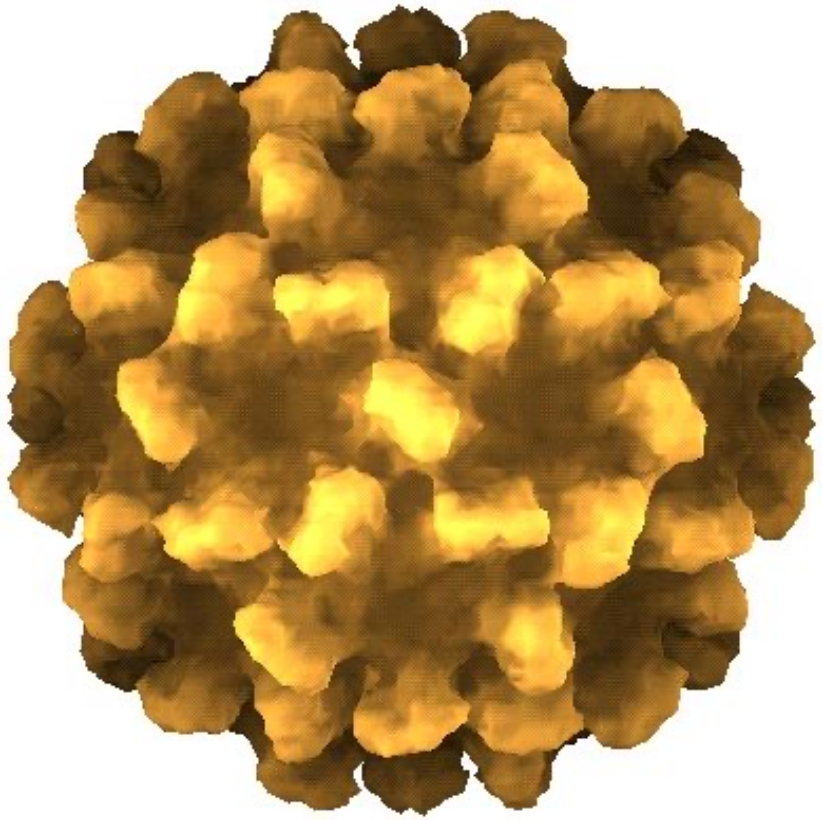


202 Diffraction from Ge QD array

with G. Bauer, Z. Zhong and T. H. Metzger



Tomato Bushy Stunt Virus
1980



Conclusions and Outlook

- Inversion of CXD demonstrated
- Internal structure of Au Nanocrystals
- Preservation of coherence upon focussing
- Dislocations, Quantum Dots all possible now
- Single molecules one day