Use of Coherent X-ray Diffraction to Image Surface Structure

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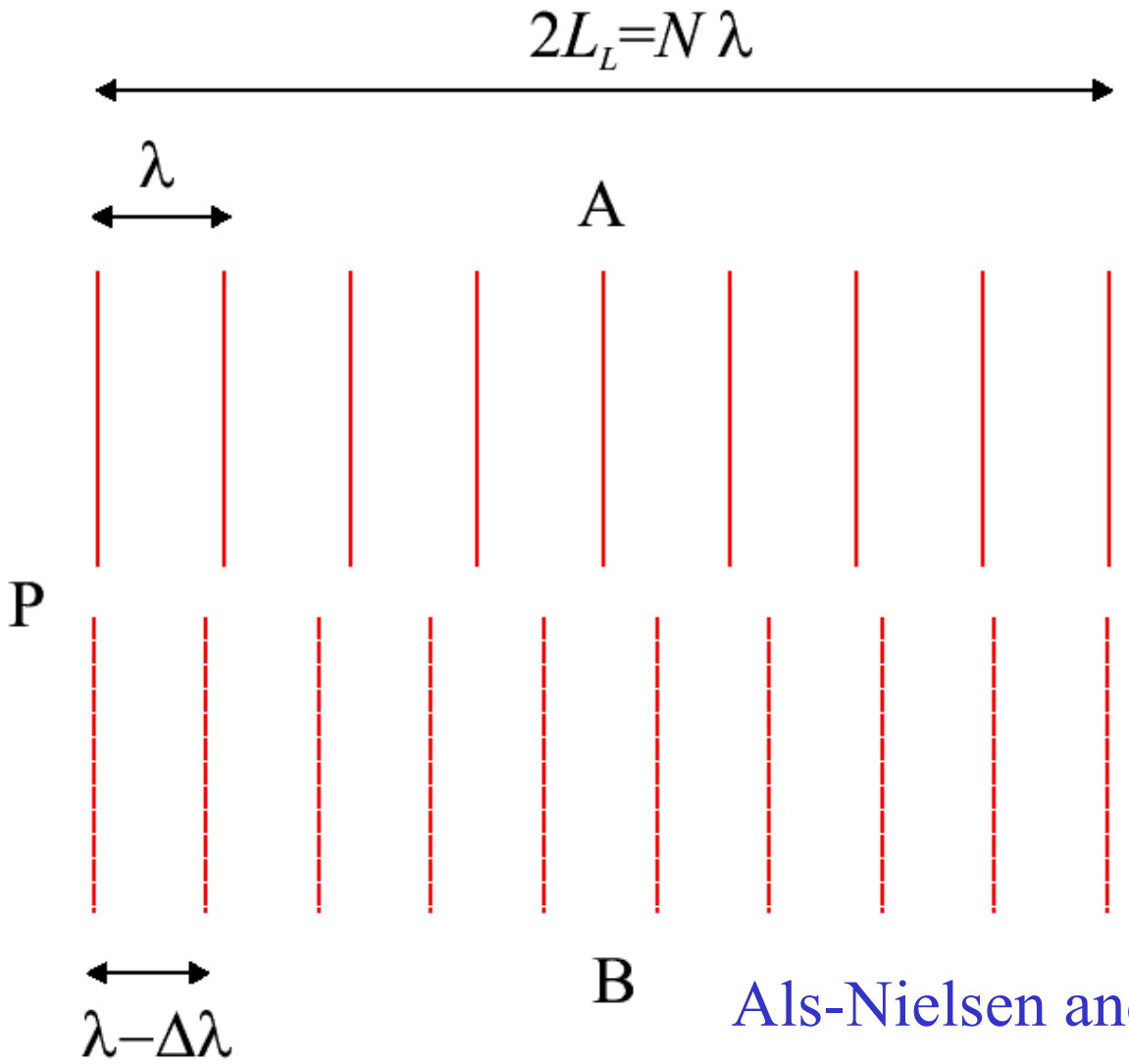
Outline

- Coherence in Diffraction
- The Phase Problem
- Coherent Diffraction from Surfaces
- Nanocrystal Facets
- Applications

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

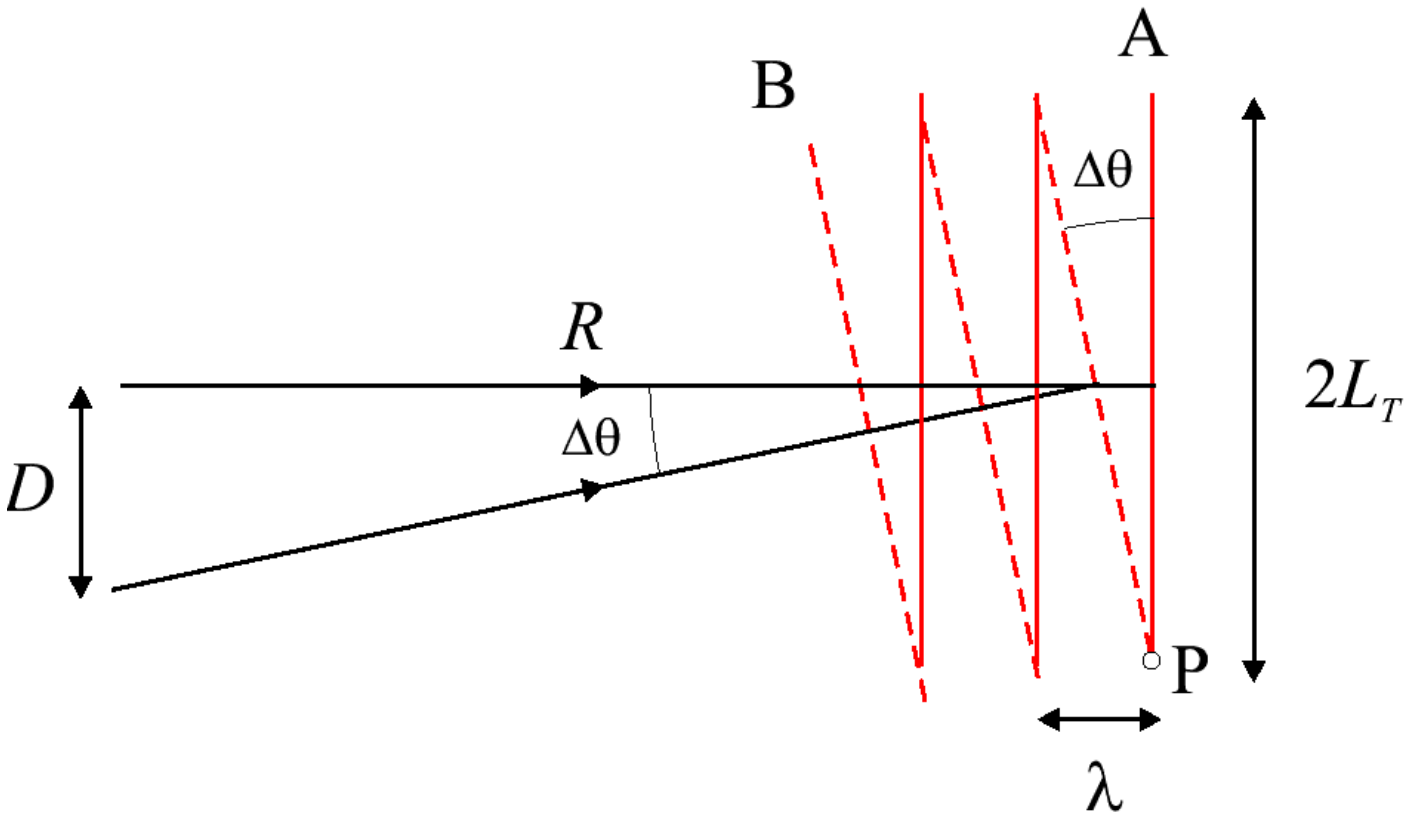
Longitudinal Coherence



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

Als-Nielsen and McMorrow (2001)

Lateral (Transverse) Coherence



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

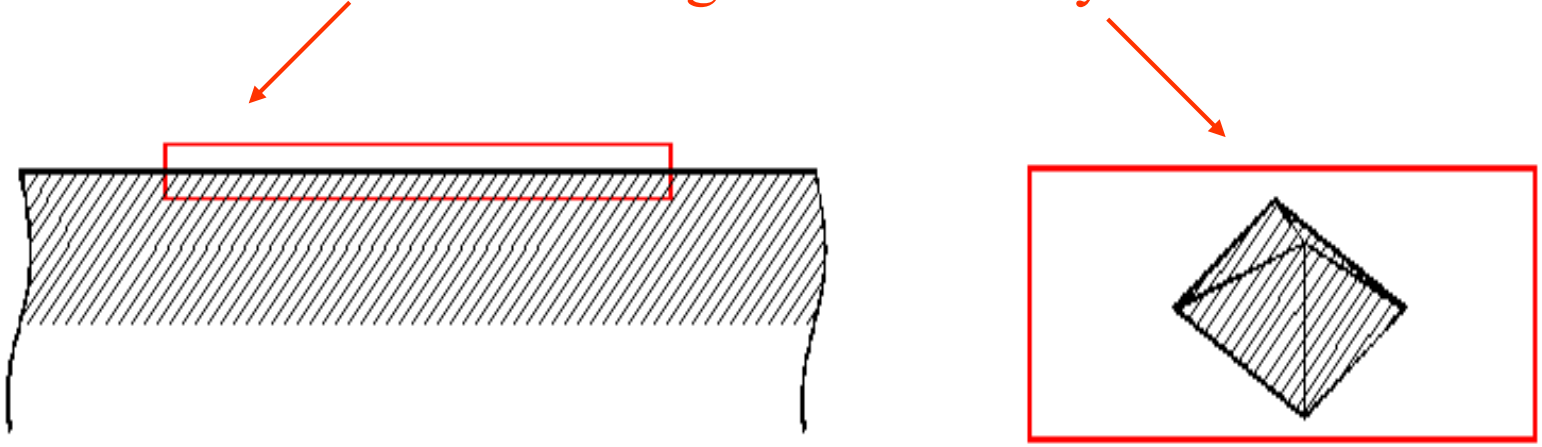
Als-Nielsen and McMorrow (2001)

Coherence at the APS

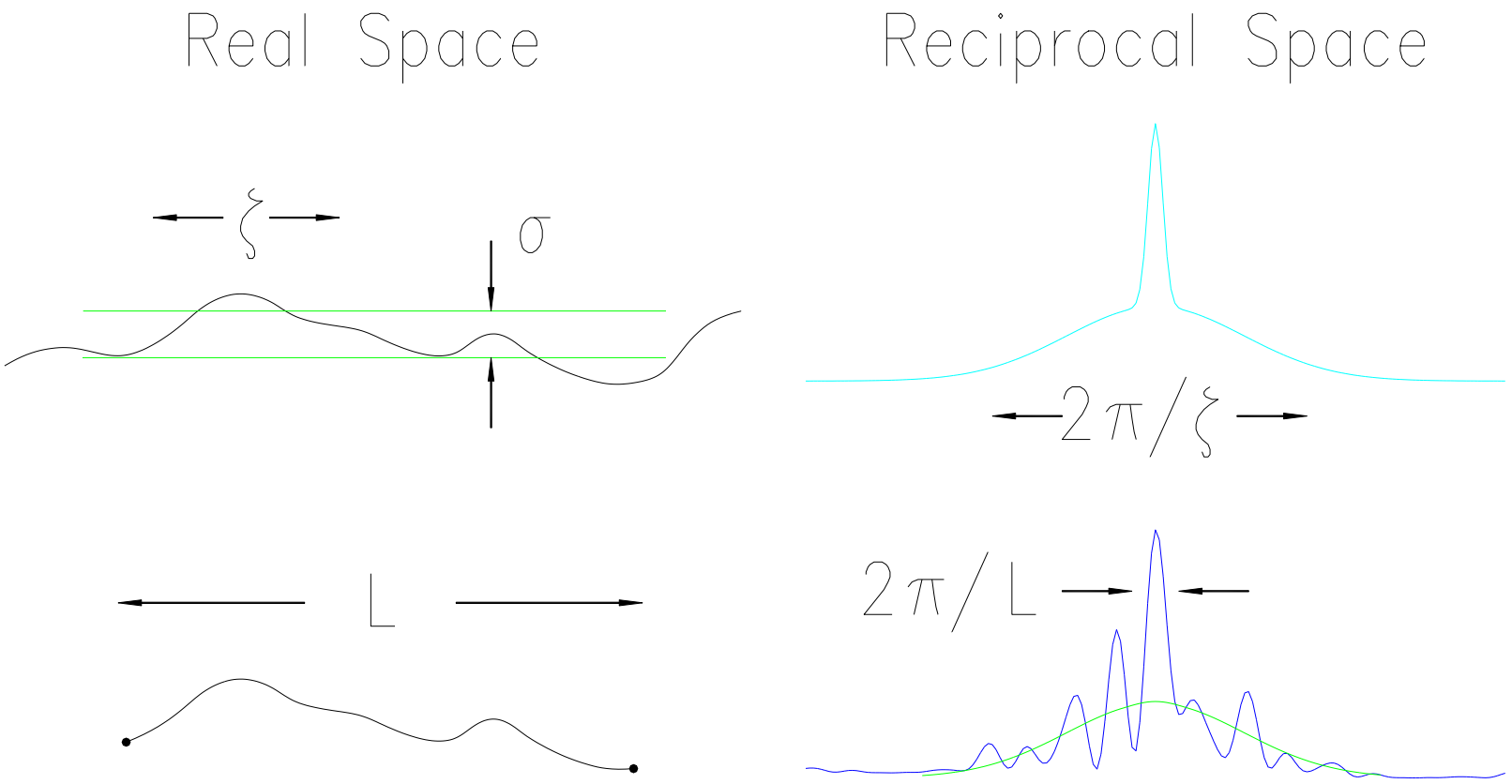
Typical 3rd Generation (undulator) Synchrotron Source

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

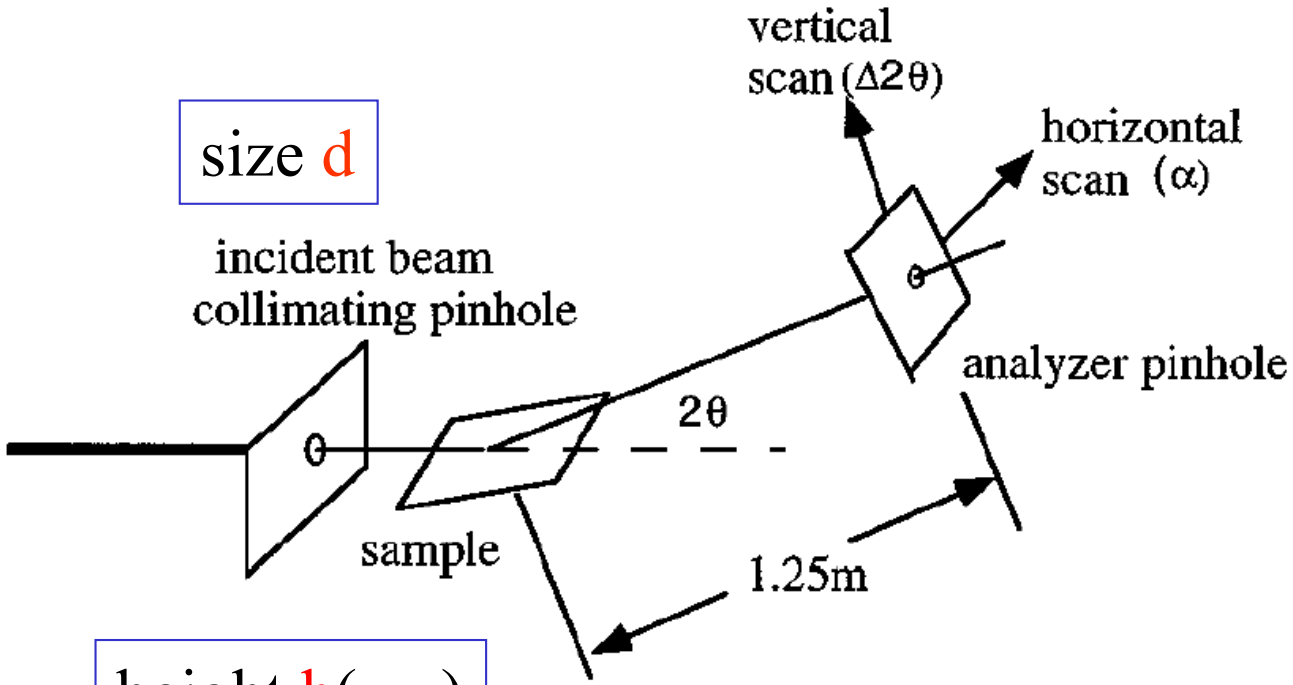


Diffuse Scattering acquires Structure using CXD



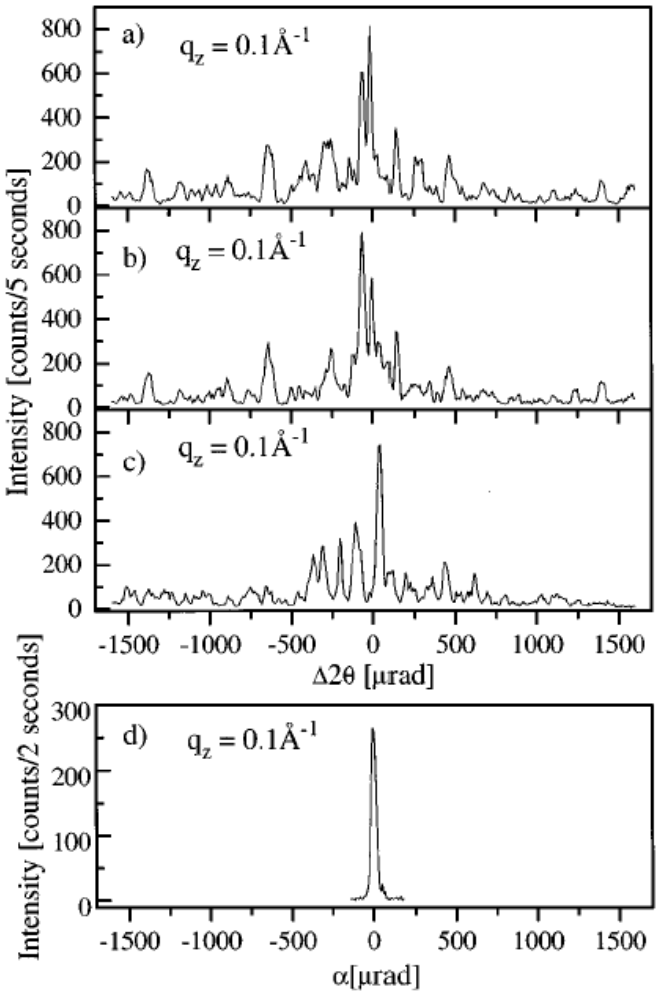
Surface Coherent X-ray Diffraction

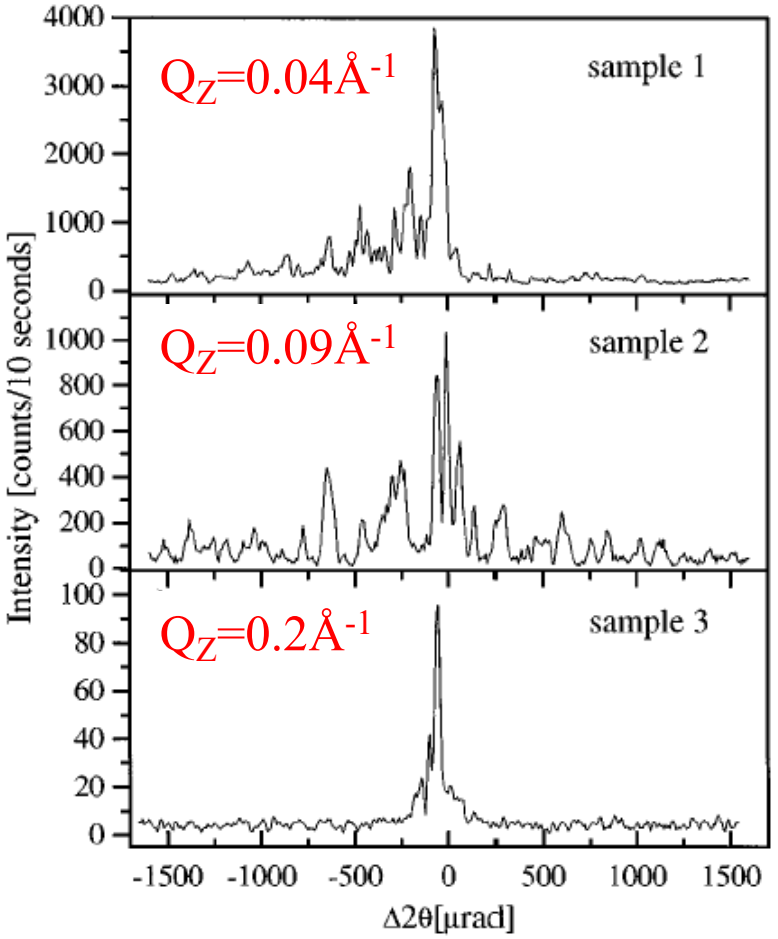
$$A(q_x, q_y) = \int_{-d/2}^{d/2} dx dy e^{iq_z h(x,y)} e^{iq_x x} e^{iq_y y}.$$



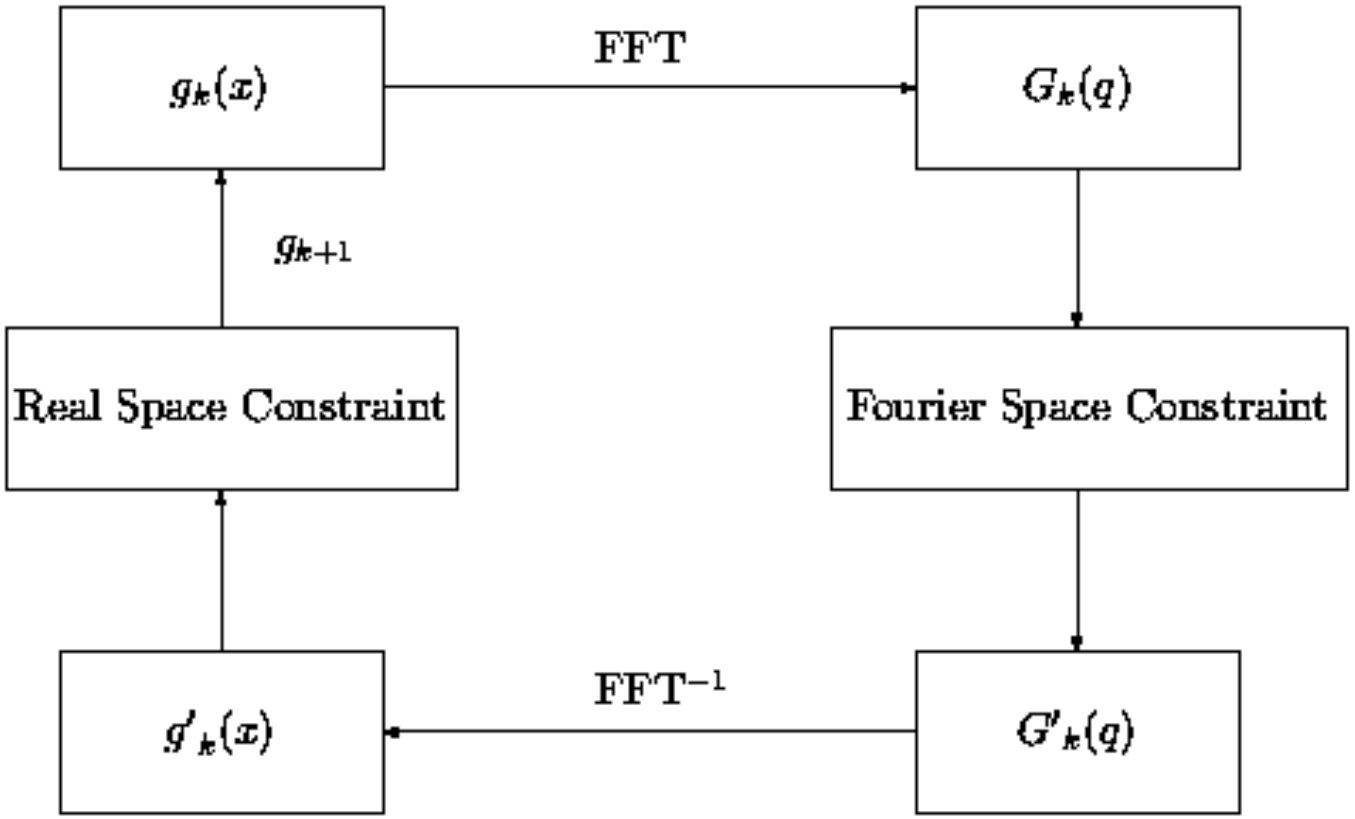
Si Samples of Different Roughness

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





Generic "Error Reduction" method



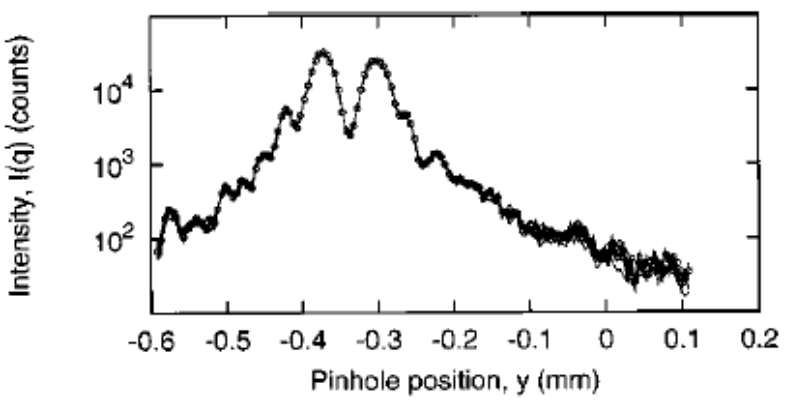
J. R. Fienup Appl. Opt. 21 2758 (1982)
R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)

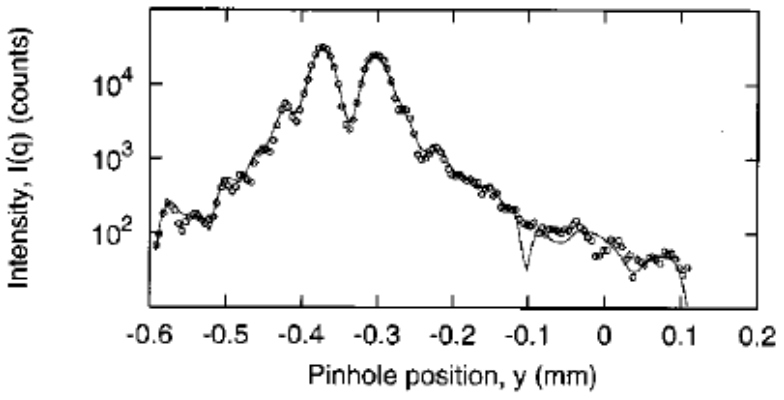
"ER" Methods in Crystallograhy

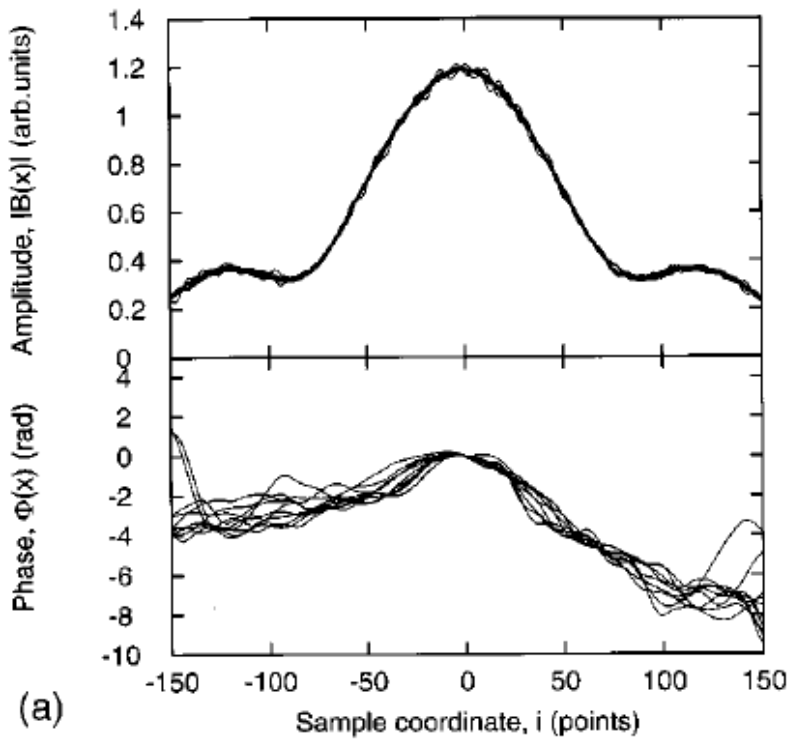
R. P. Millane, J. Opt. Soc Am. A **13** 725 (1996)

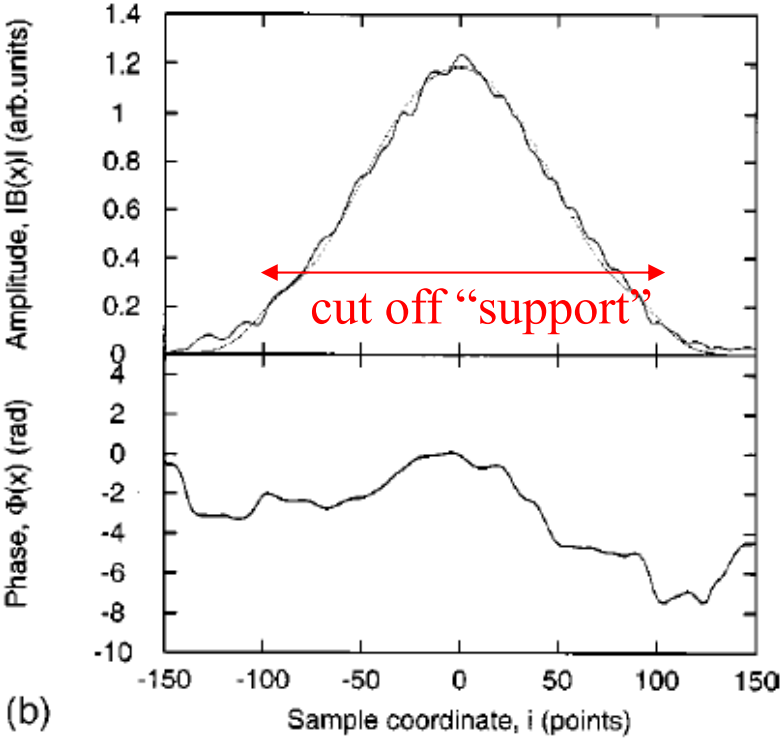
- 'Positivity' and 'Atomicity' constraints
- Finite support, molecular envelope
- Solvent flattening
- Molecular replacement
- Non-crystallographic symmetry
- Basis of 'direct methods' (Sayre, Bricogne)
- Non-uniqueness is 'pathologically rare' (d>1)

Phasing using G-S Algorithm

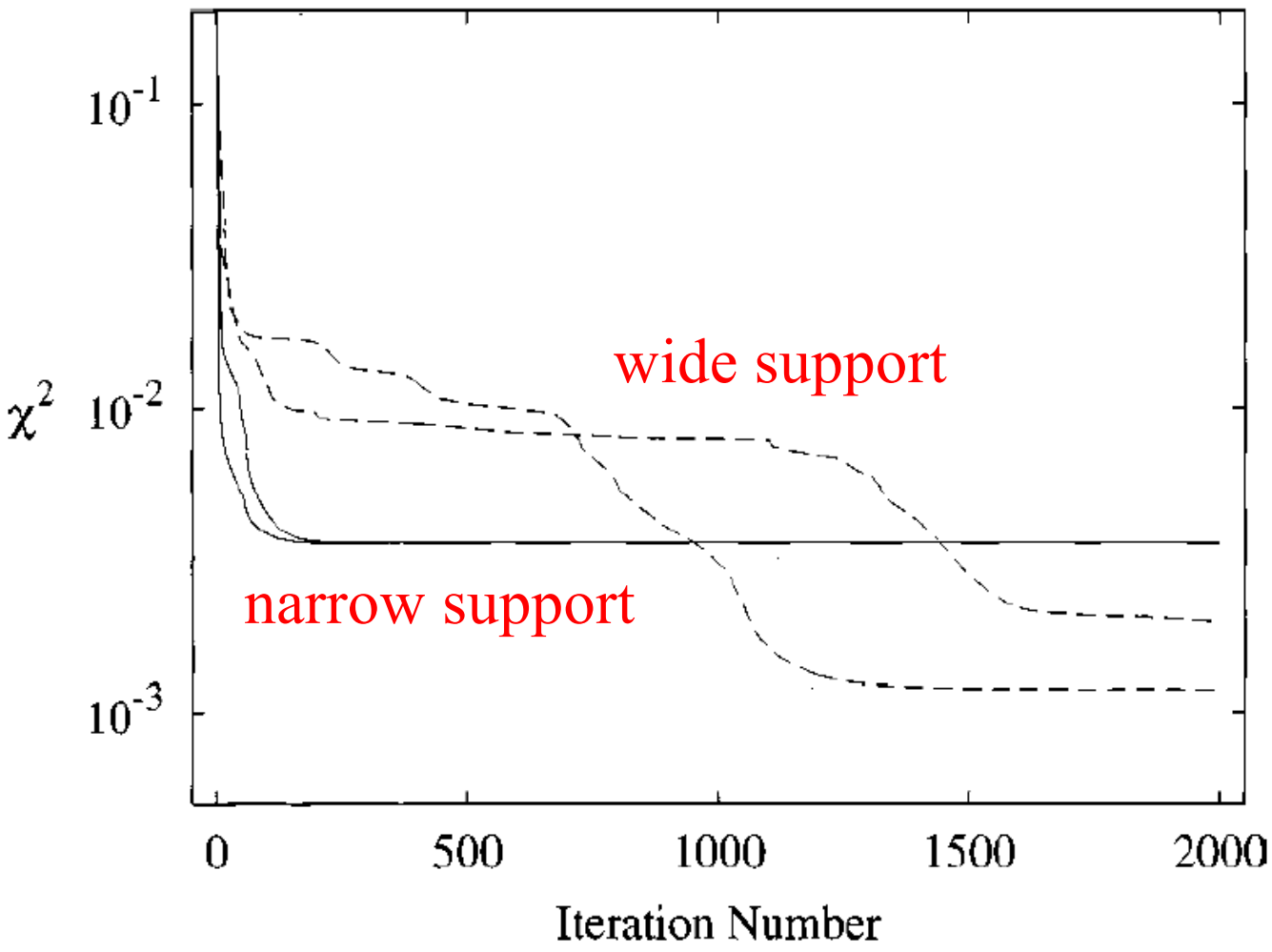








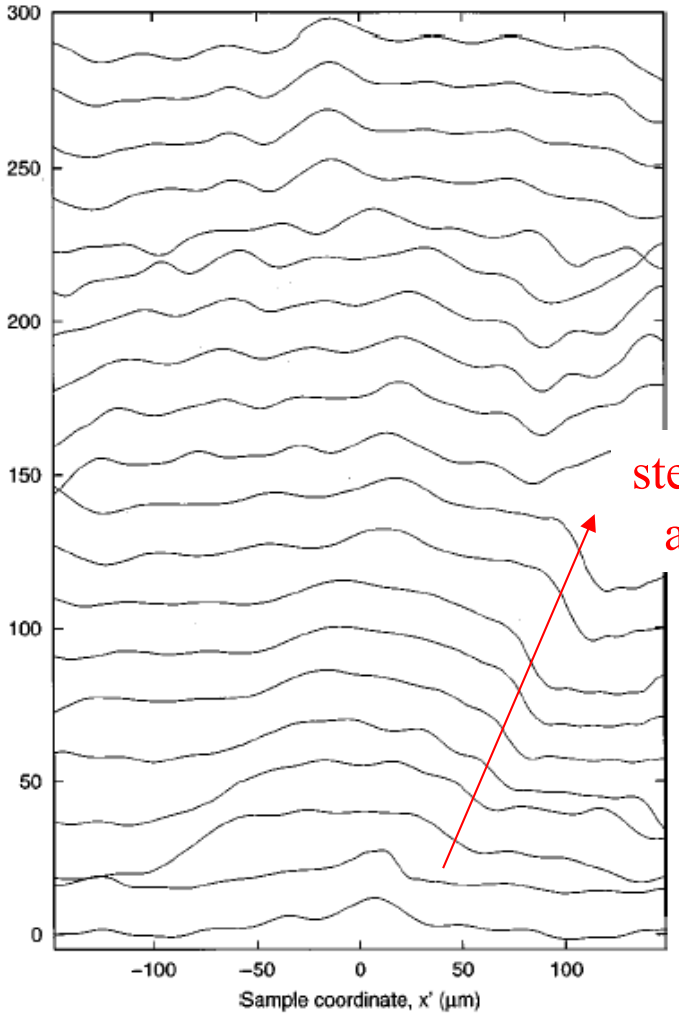
Convergence Trajectory



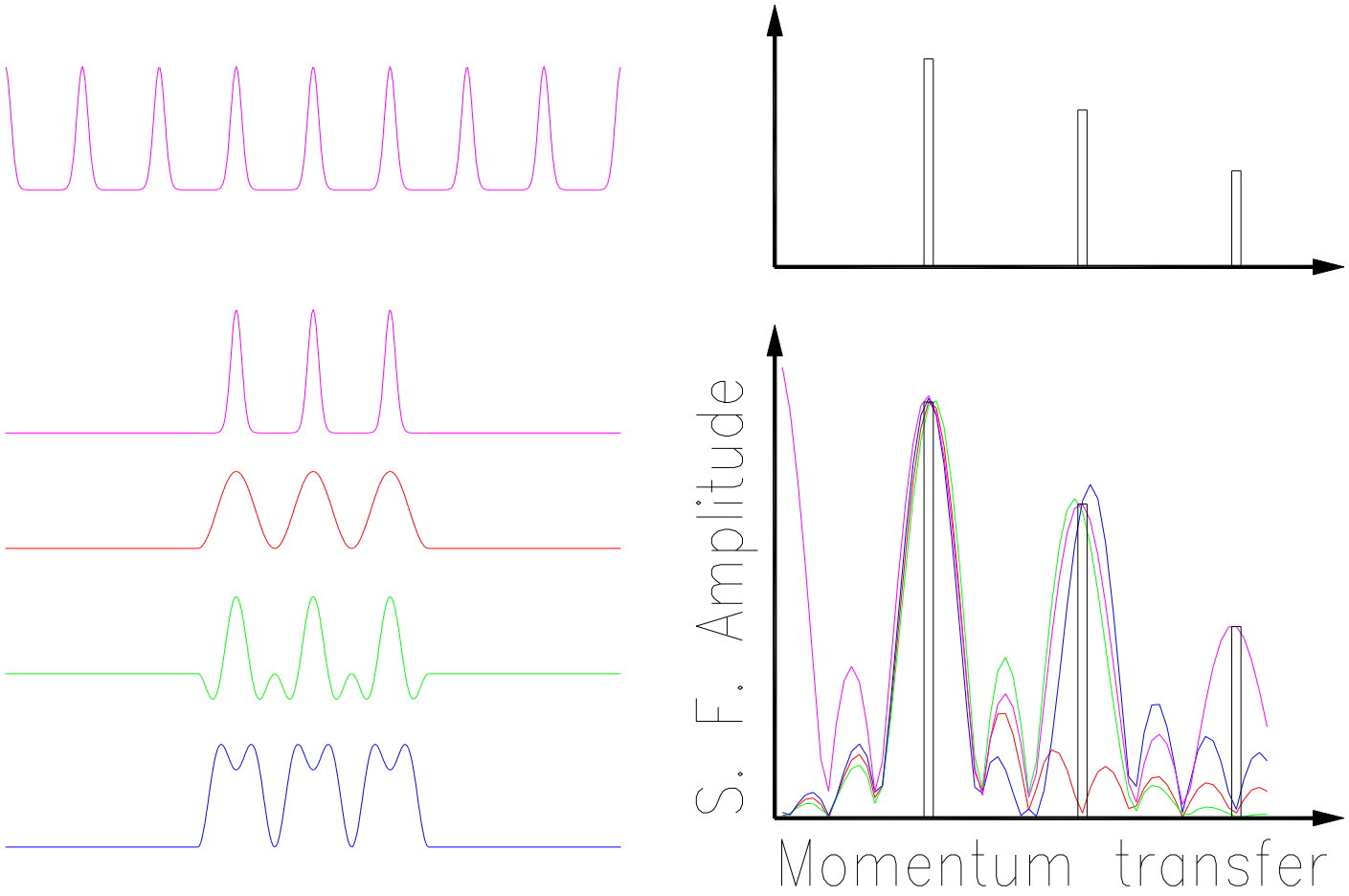
In-situ Regrowth of Oxide on Si

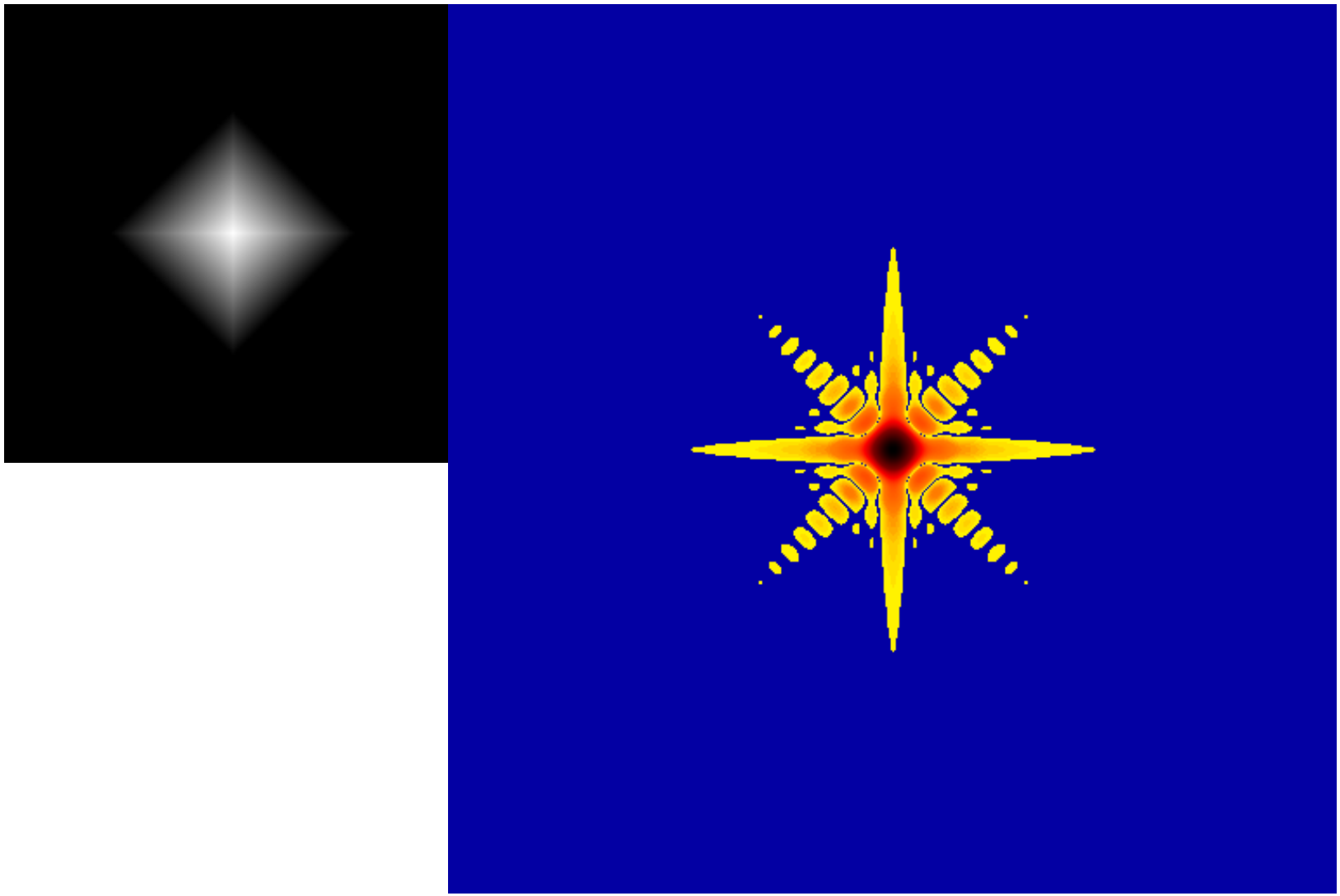
Phys. Rev. B **60** 9965 (1999)

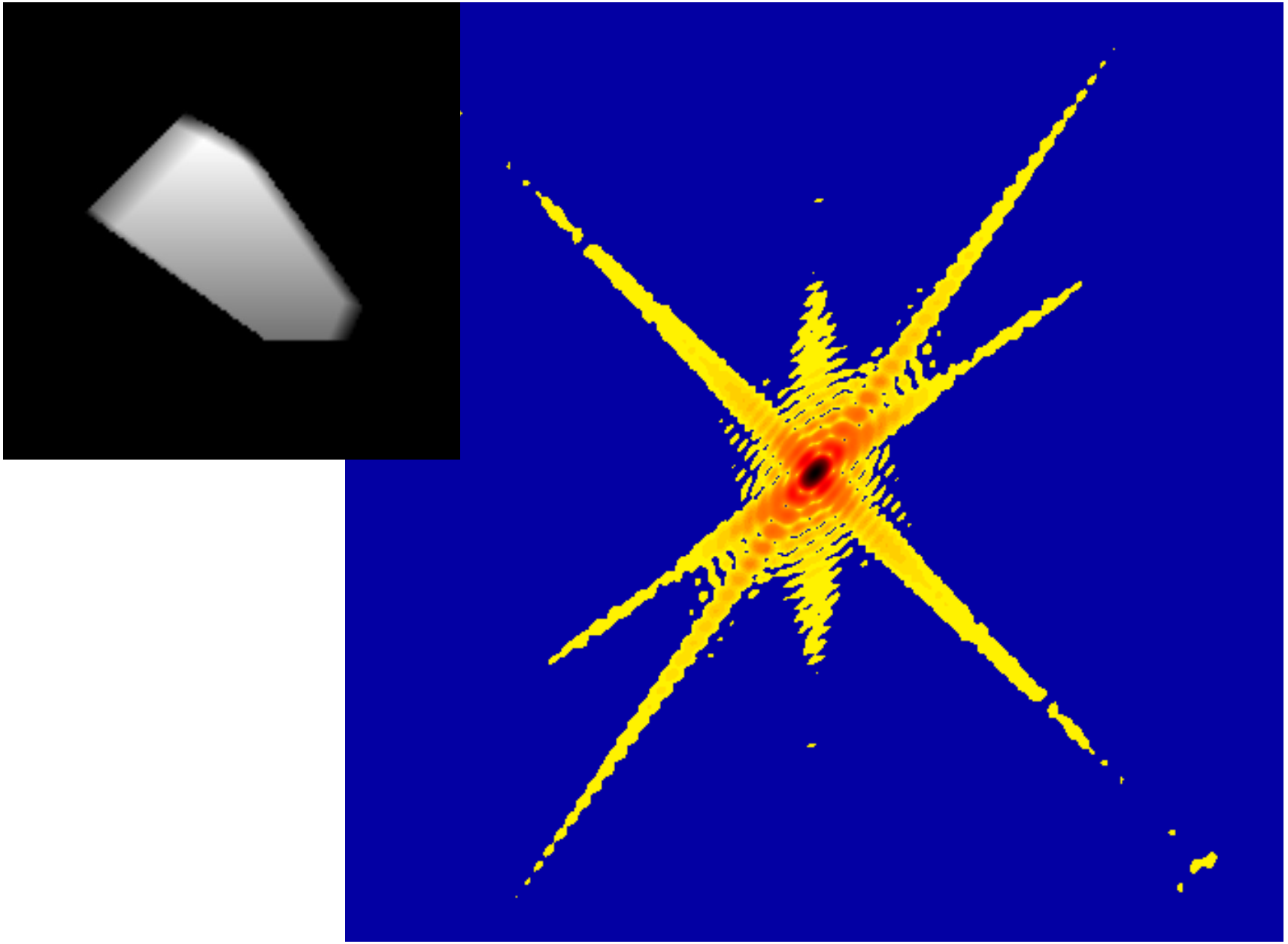
- Wafer stripped with HF at t=0
- CXD pattern measured every 180 sec
- Each profile reconstructed independently
- Random starting phase each time



Phase Problem: Finite-size Effect

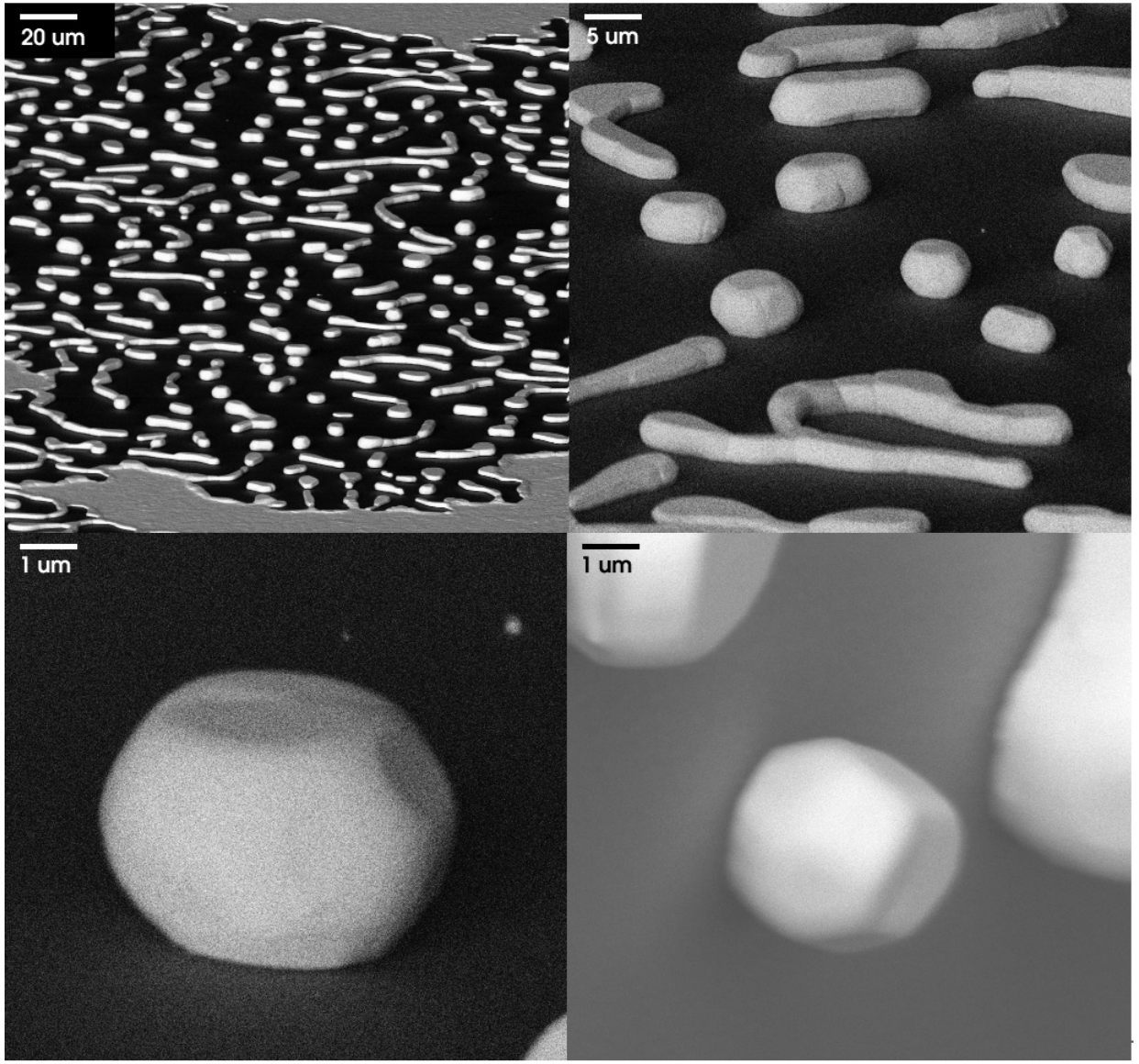




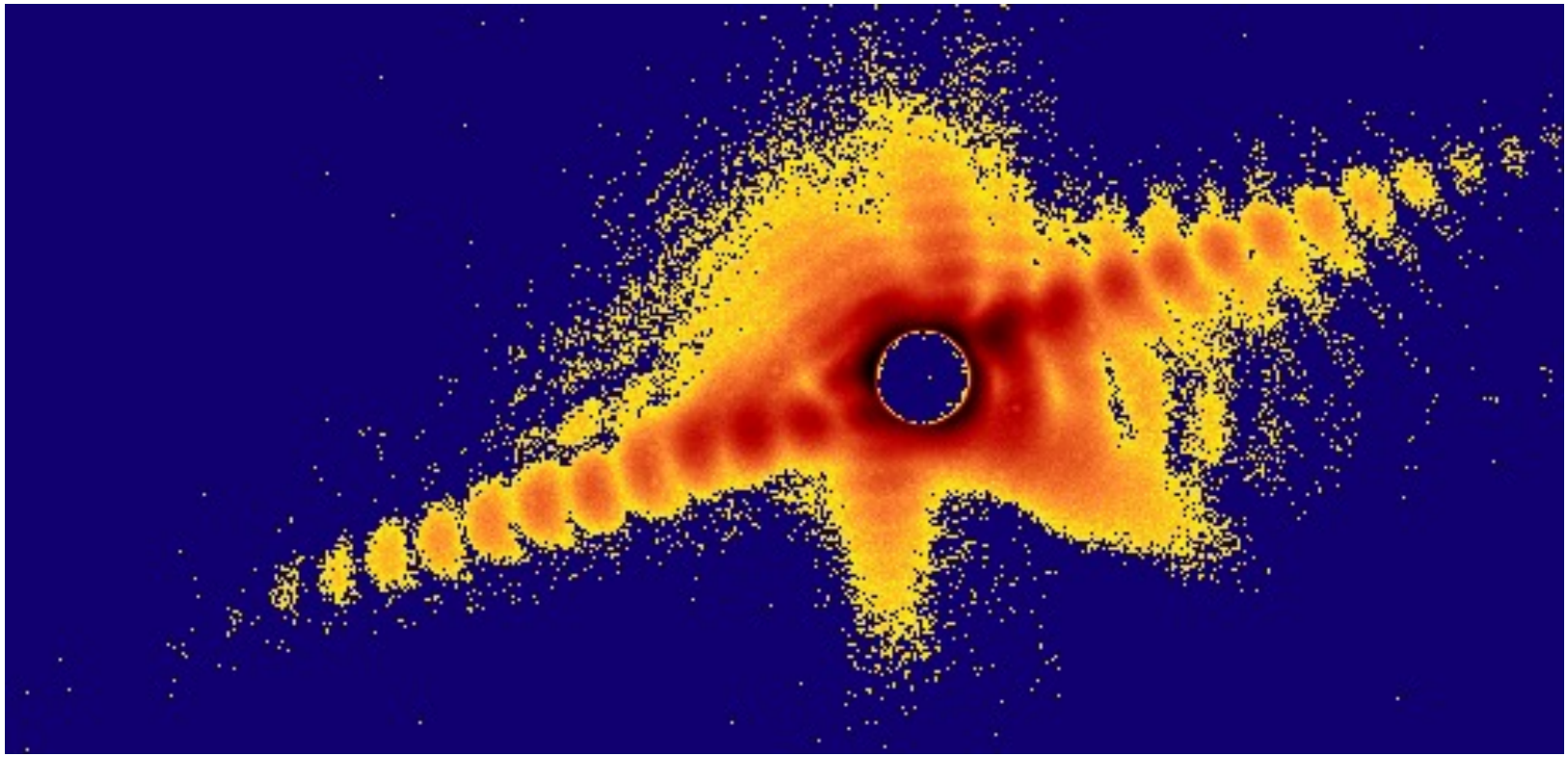


SEMS

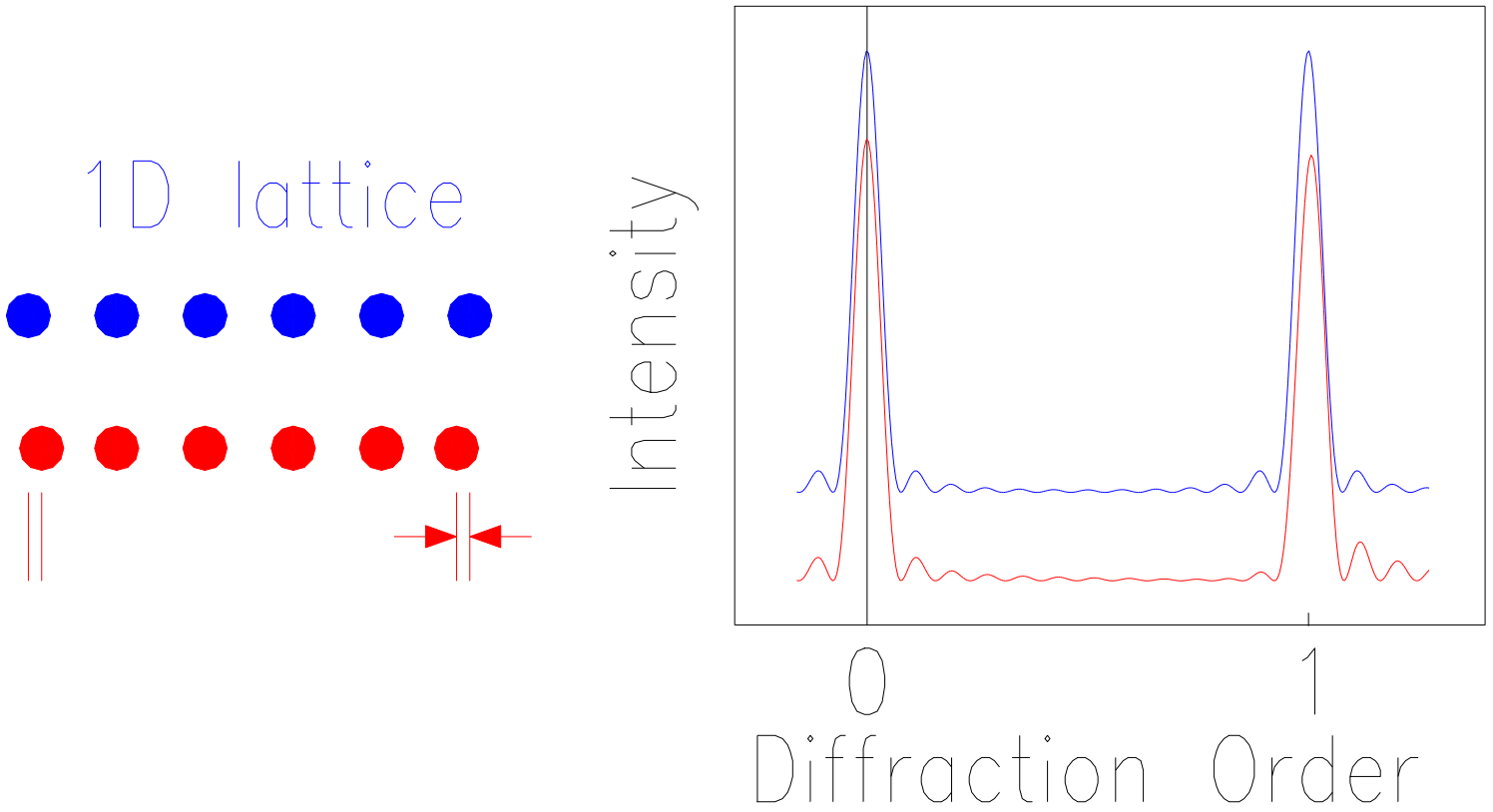
- Au blanket film
- Quartz substrate
- Annealed at 950°C for 70 hrs.



Micron-sized gold crystal: (111) Bragg reflection

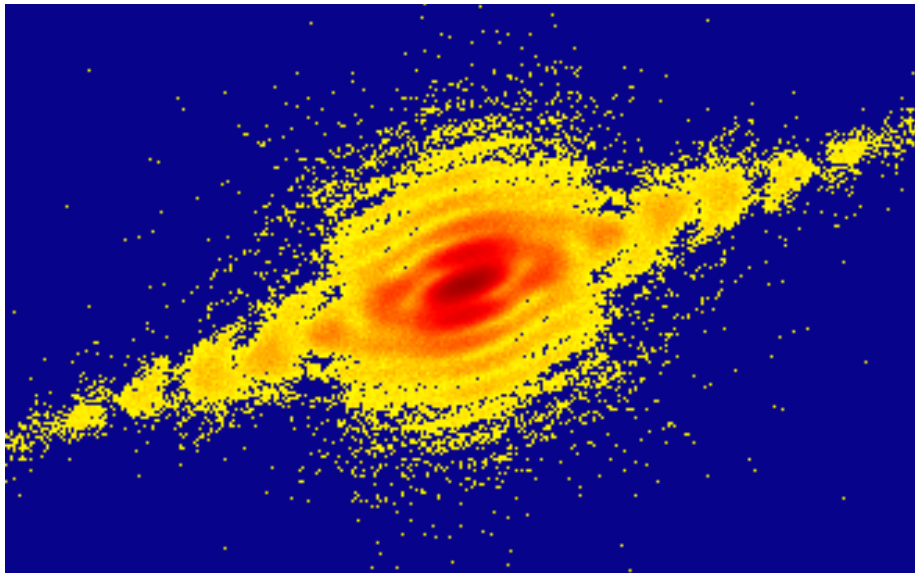


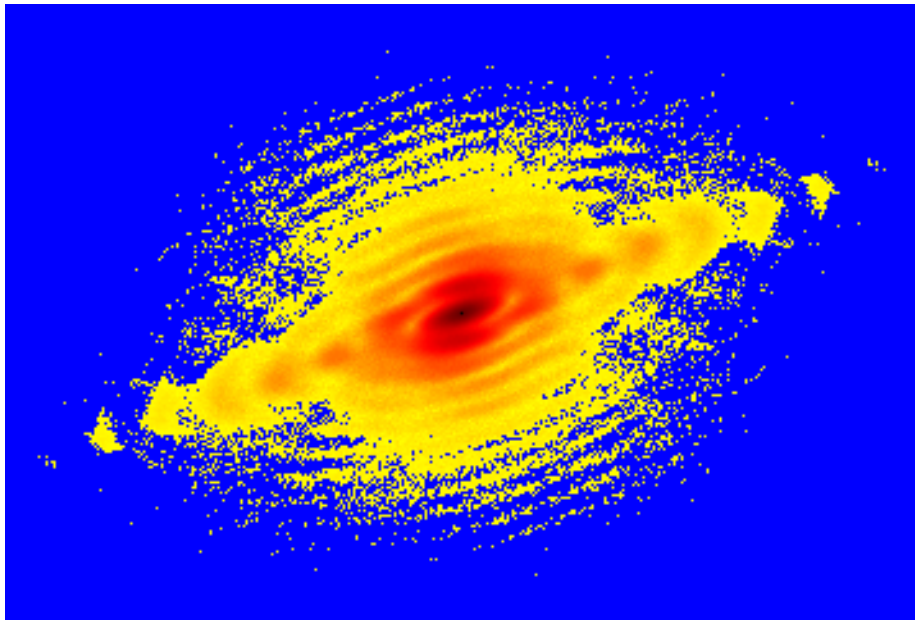
Imaging of Lattice Strains

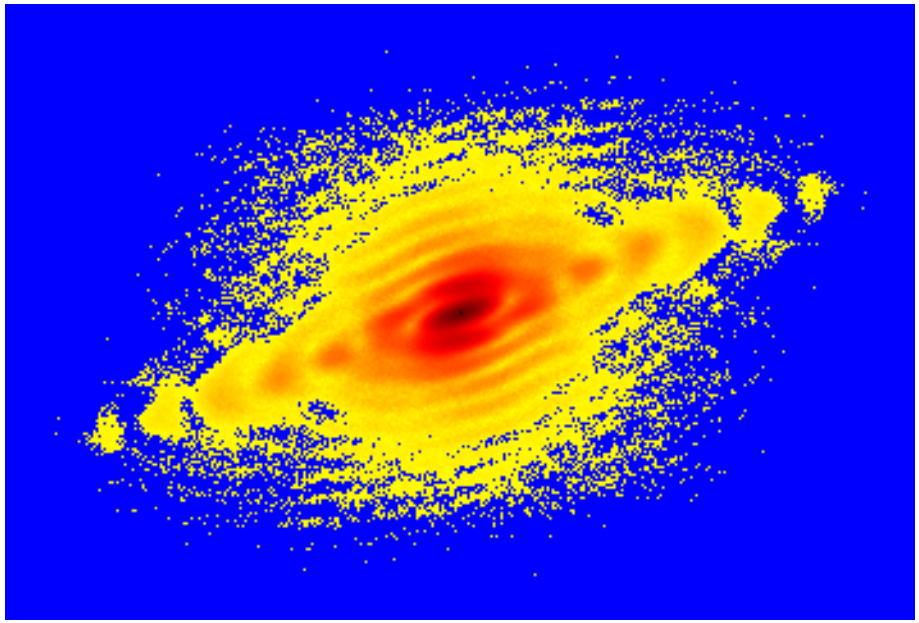


Symmetrized Data and two best fits

Chisq=0.0005

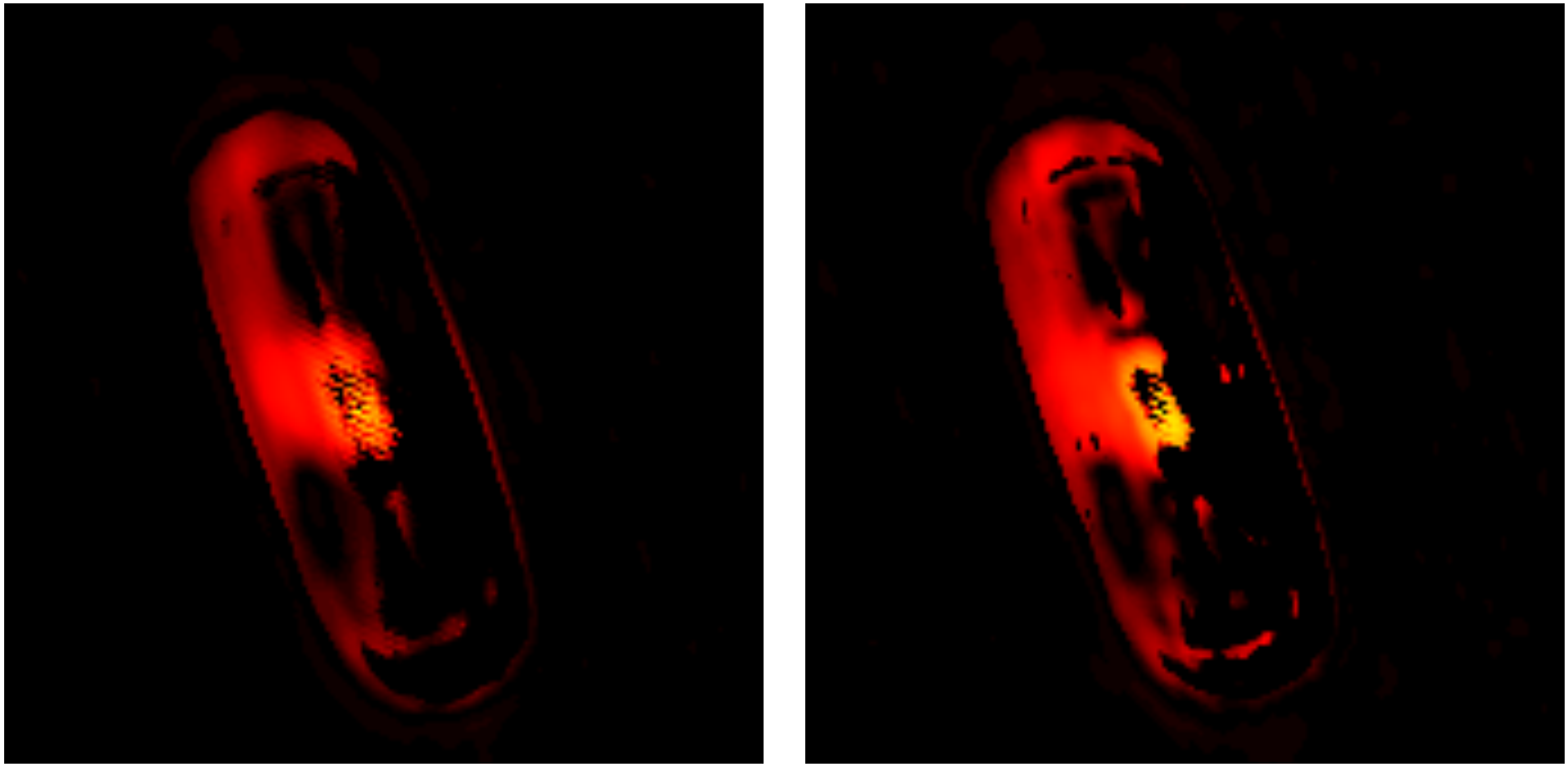




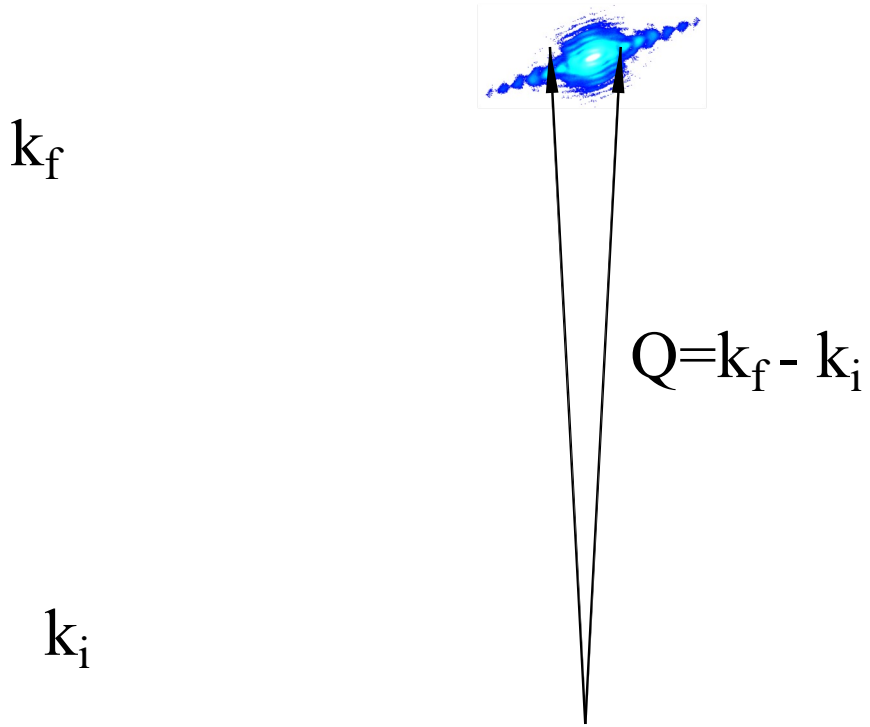


Best 2D Reconstructions

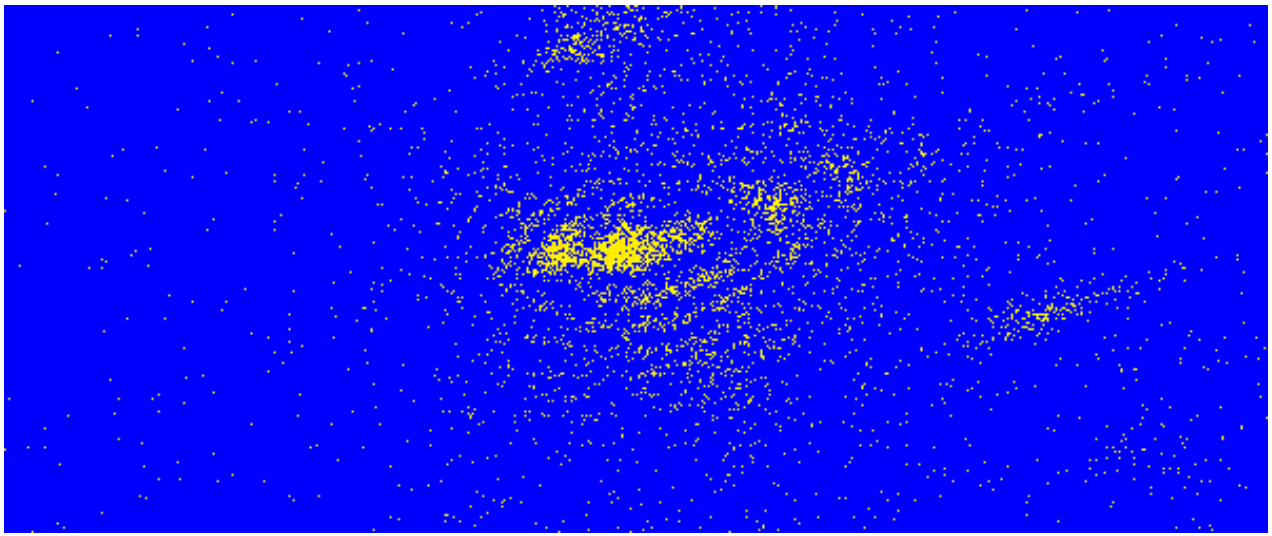
chisquare = 0.0005



3D Diffraction Method

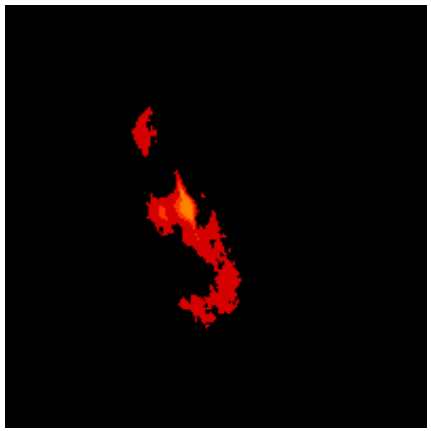


3D Diffraction Data
1 micron Au crystal

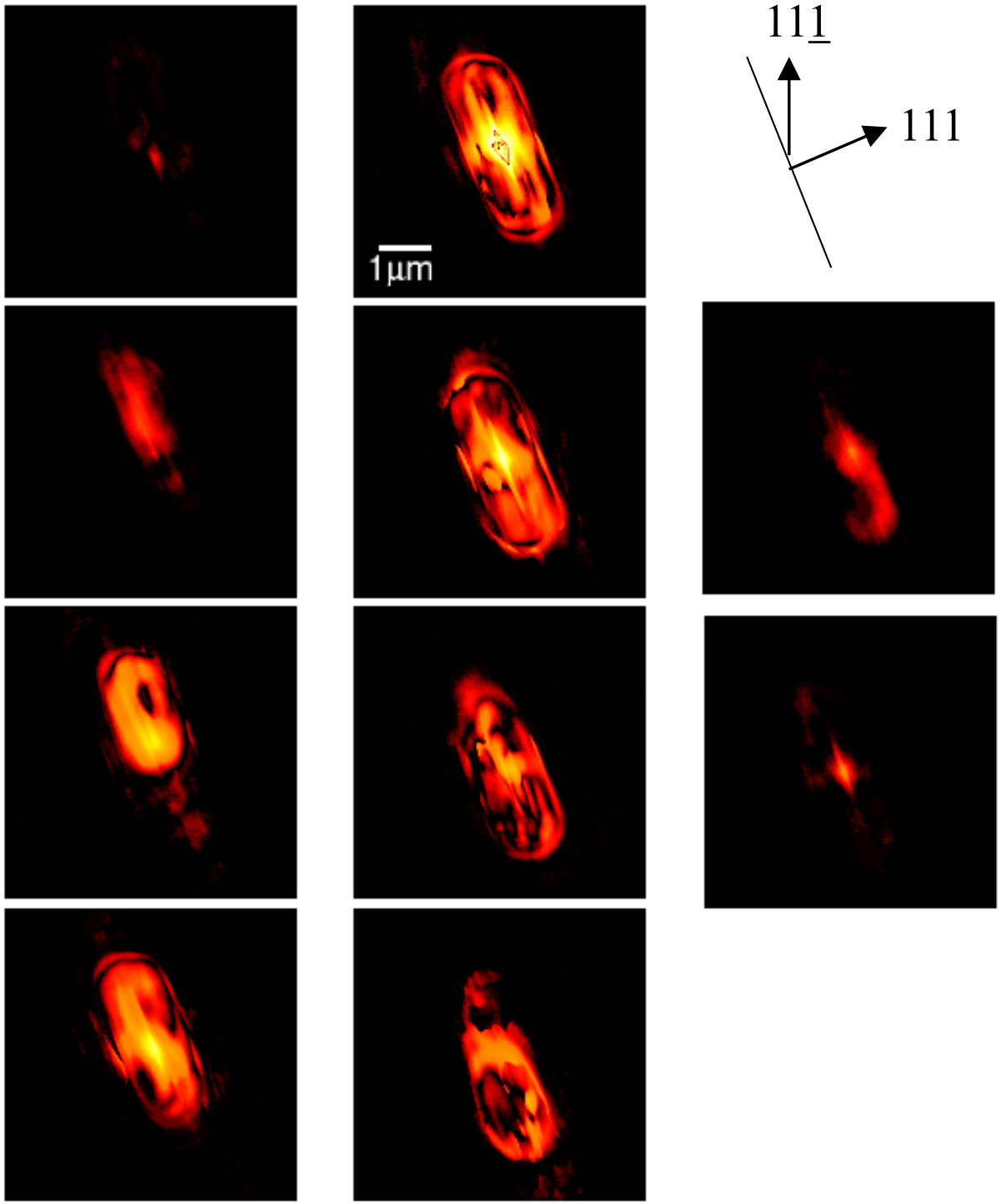


* Center is Symmetric *

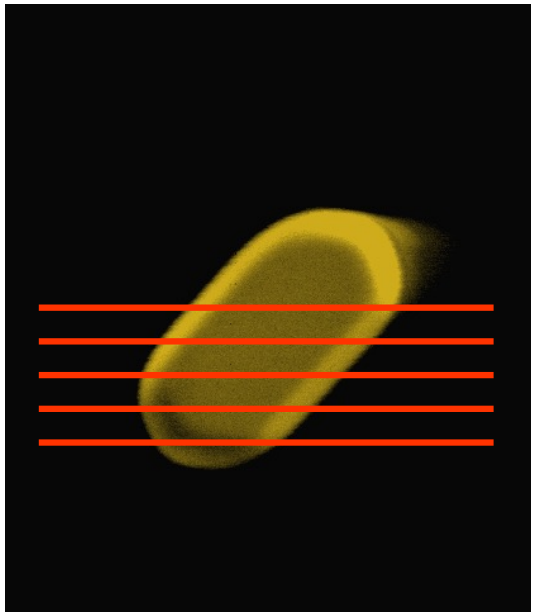
30 frames of 3D reconstruction 1 micron gold crystal



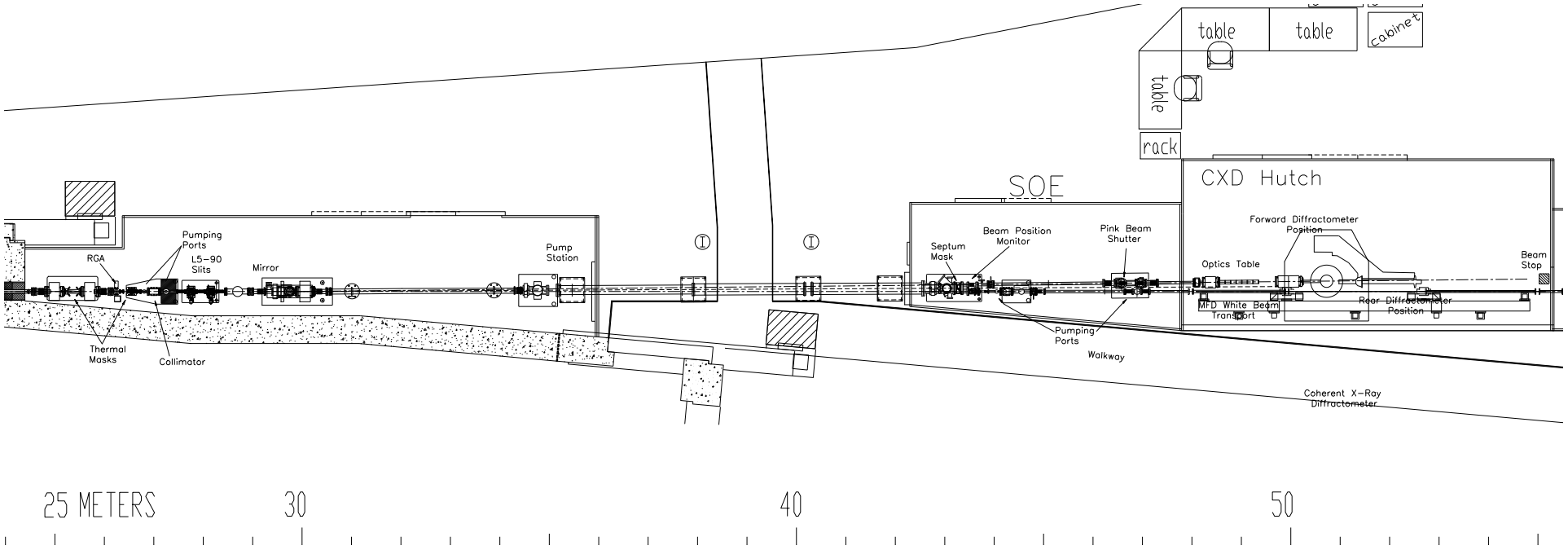
* CENTER *

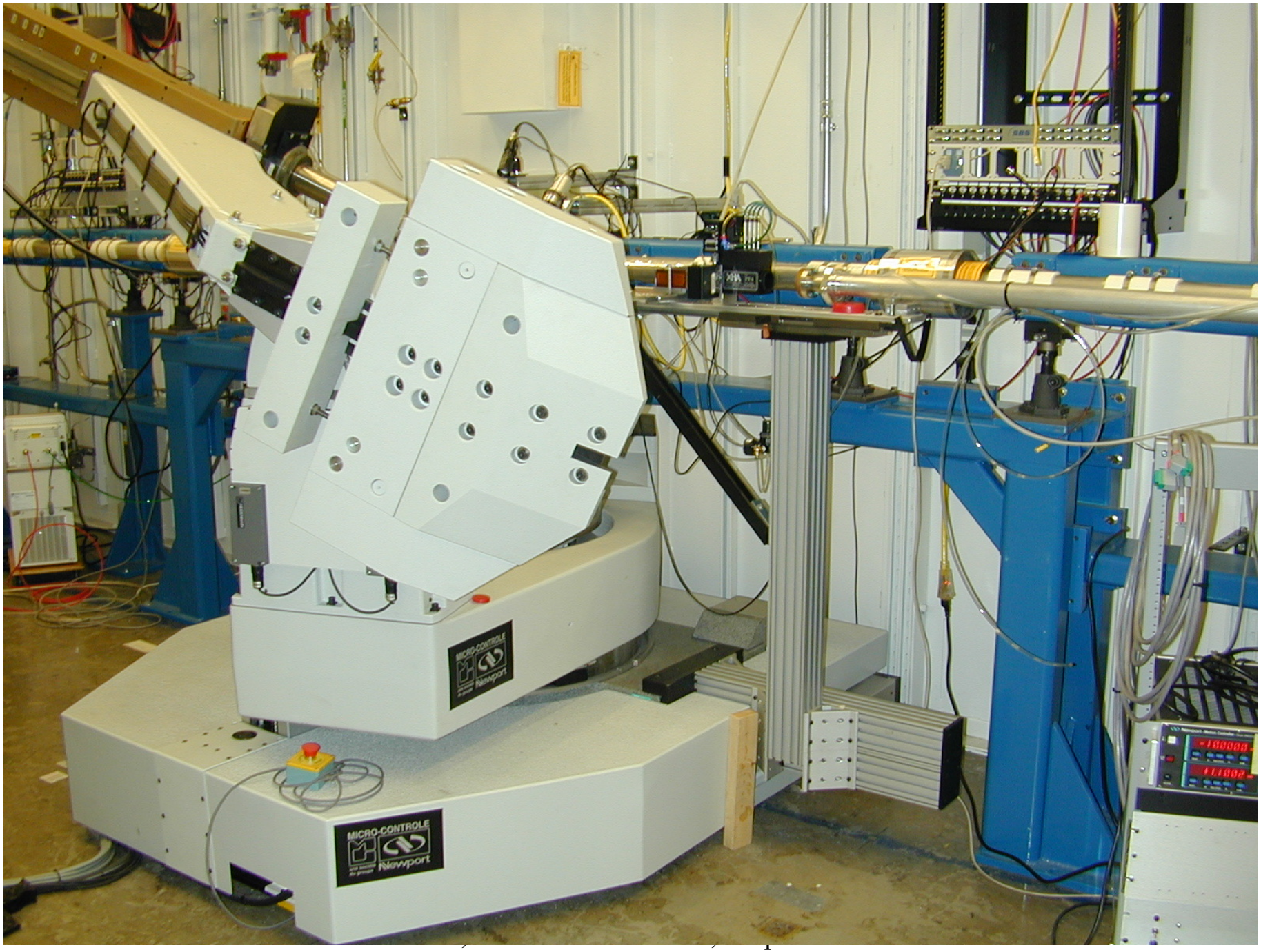


Slices through plan view SEM:



CXD Beamline at APS Sector 34

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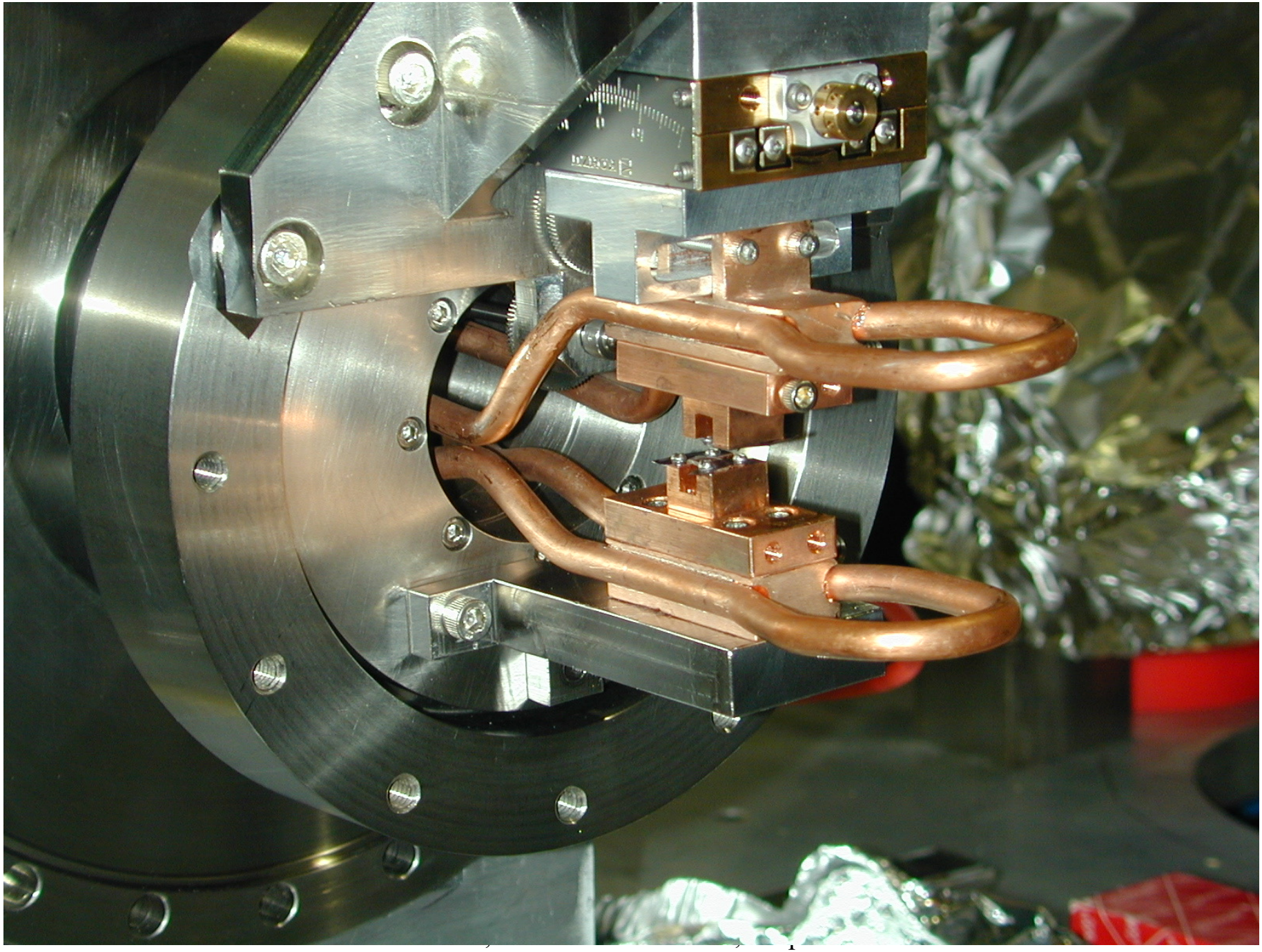

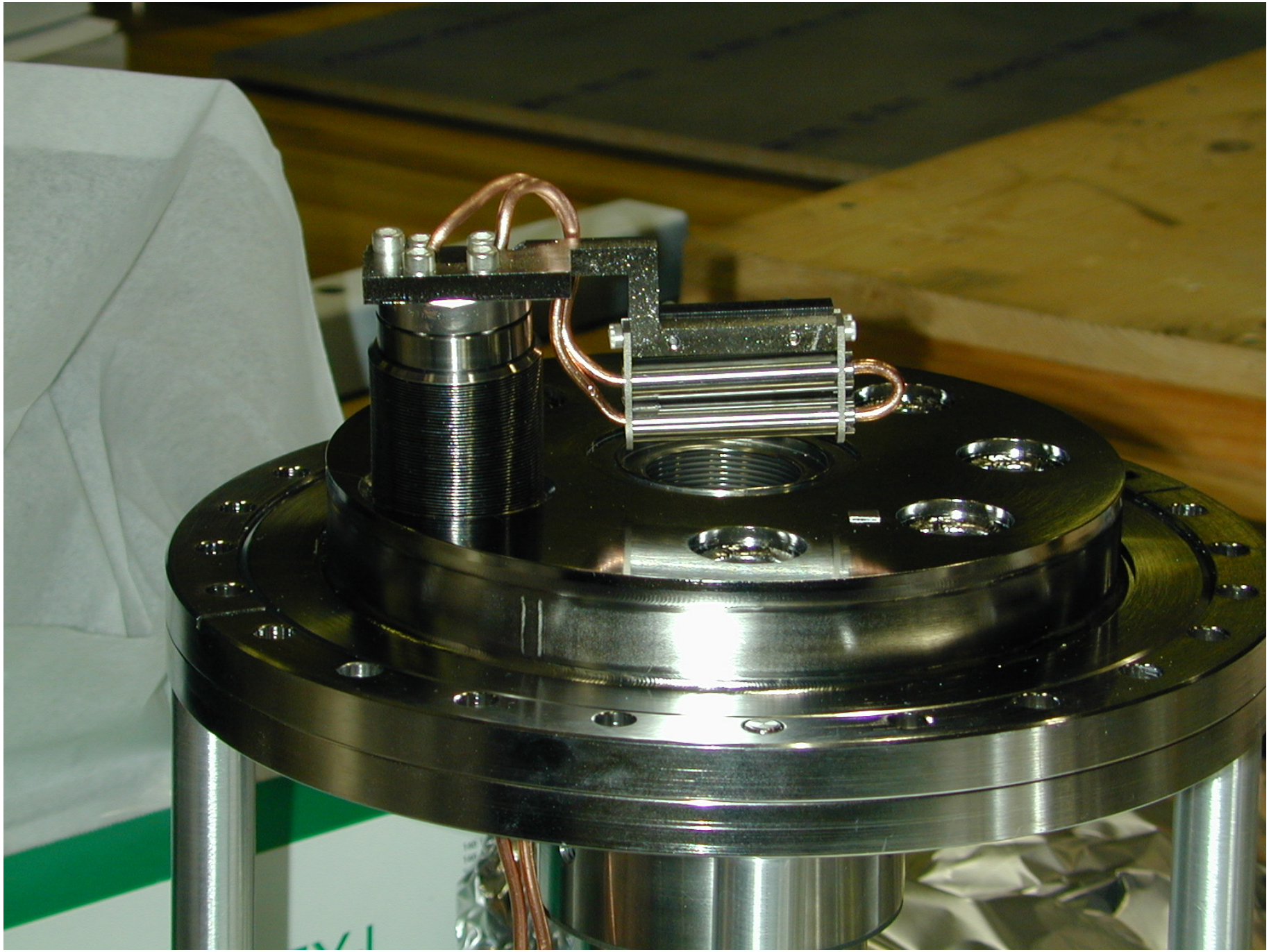

Conclusions

- "Diffuse" scattering acquires fine structure
- Surface/interface morphology
- Shapes of small particles
- CXD facility ready for experiments
- Future applications of CXD
  - Atomic-scale fluctuations
  - 3D Imaging of strain fields at the nm level