# Inversion of Coherent Diffraction Images of Nanocrystals


- Ian Robinson
- Ivan Vartanyants
- Franz Pfeiffer
- Mark Pfeifer
- Garth Williams
- Curtis Benson

Department of Physics
University of Illinois


SRI Coherence Workshop,
Berkeley, August 2003

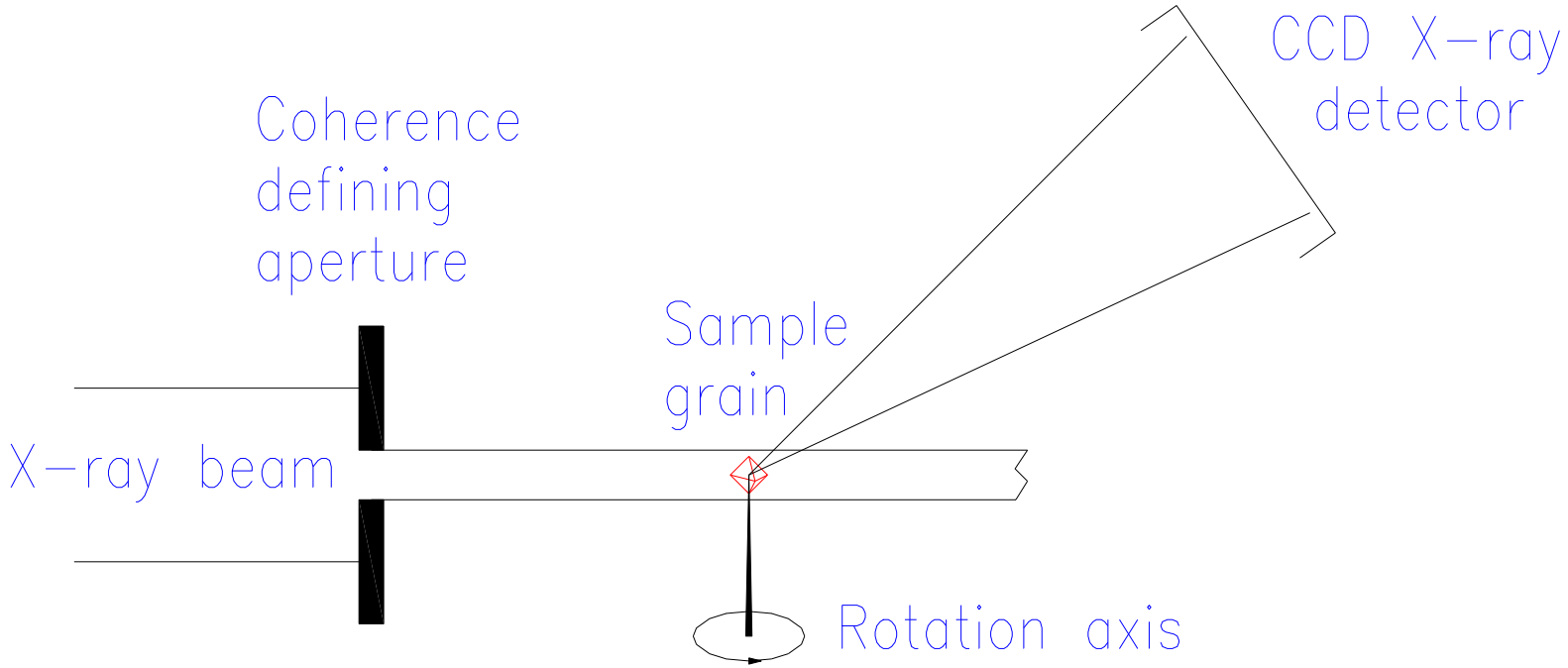
# Outline

- Coherence in Diffraction
- Oversampling and the Phase Problem
- Nanocrystal Shapes
- How small can we go?
- Future Applications of CXD

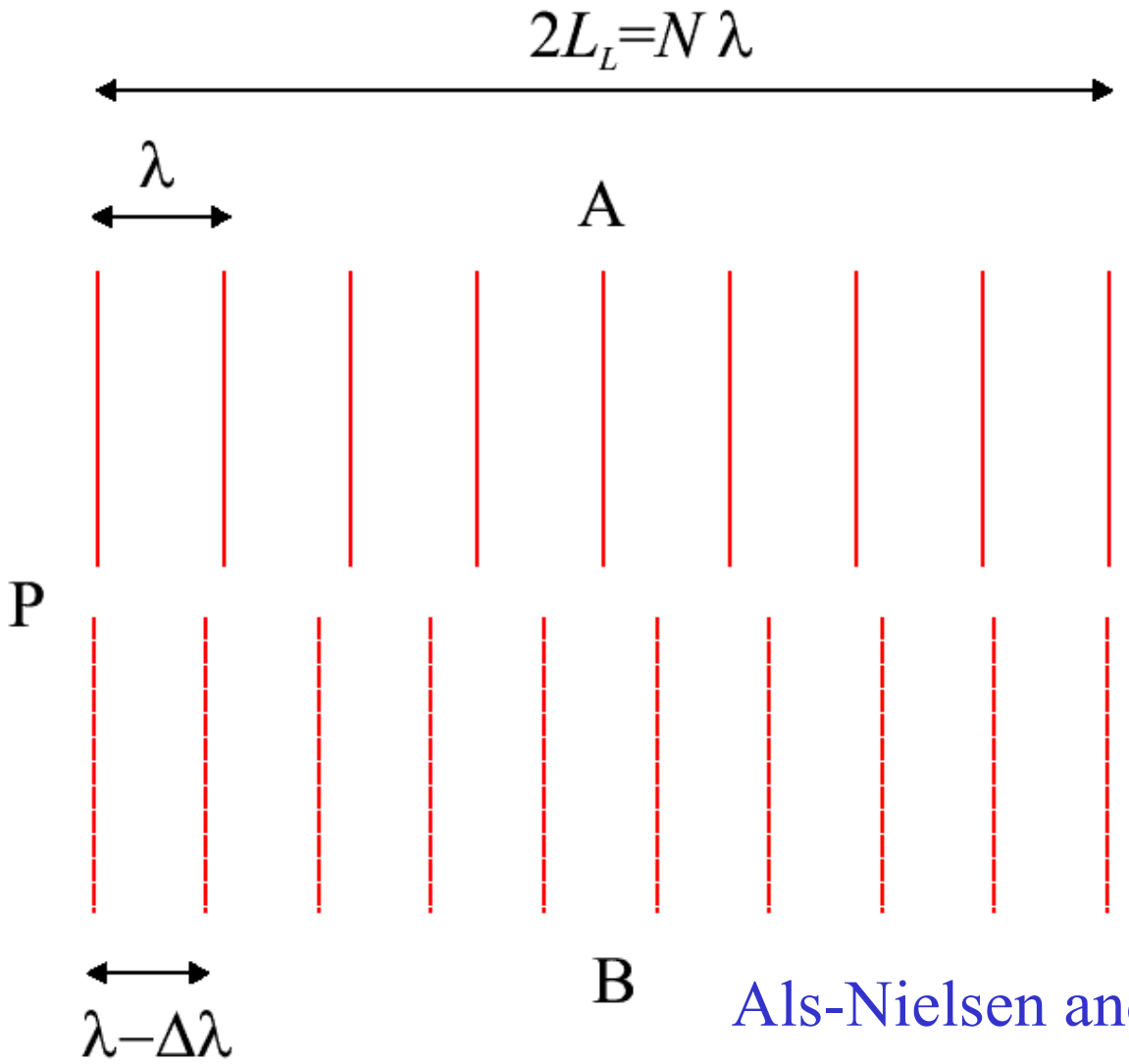
# Uses of Coherence in Diffraction

- 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



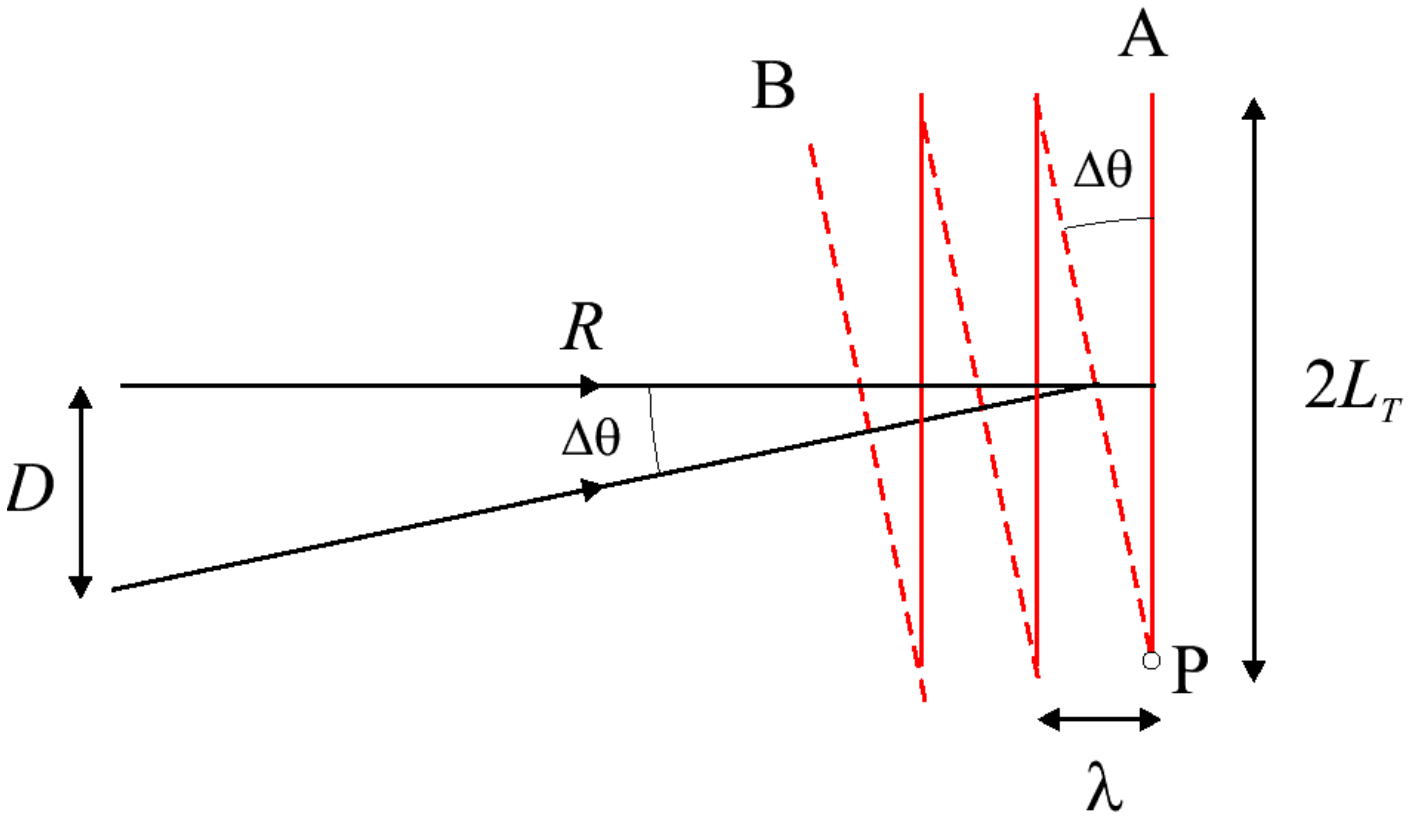
# Longitudinal Coherence




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

Als-Nielsen and McMorrow (2001)

# Lateral (Transverse) Coherence




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

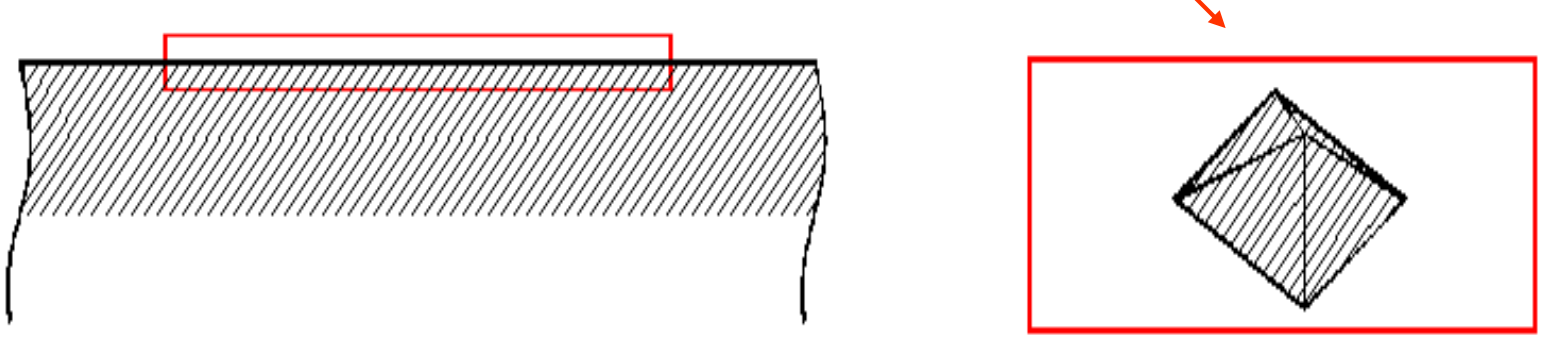
Als-Nielsen and McMorrow (2001)

# Coherence at the APS

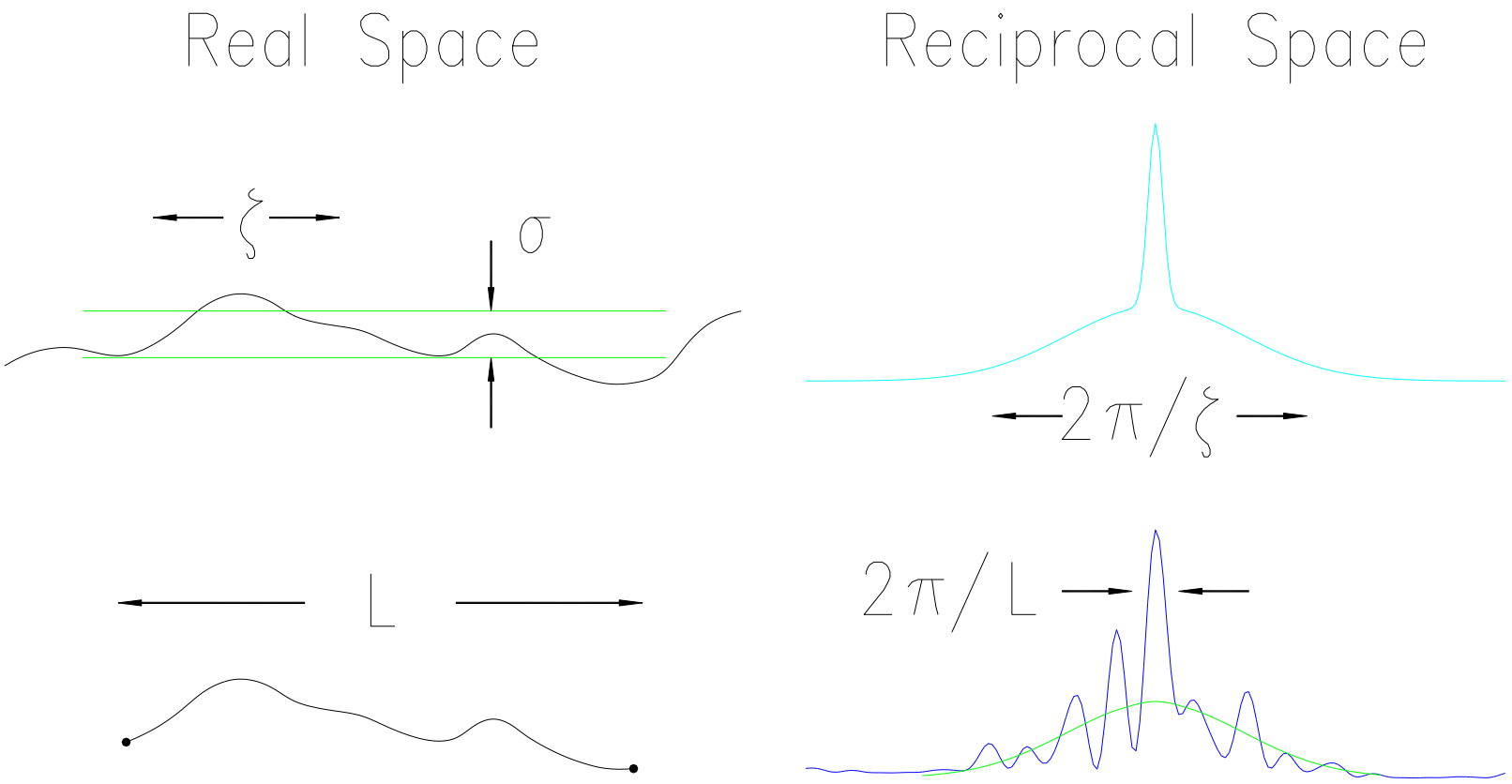
## Typical of Undulator-based Synchrotron Source

| Coherence of | $\xi_{VER}$ | $\xi_{HORIZ}$ | $\xi_{LONG}$ | Flux |
|---|---|---|---|---|
| Raw Undulator | 35μm | 9μm | 0.004μm | $2\times10^{12}$ |
| Si(111) Monochromator | 35μm | 9μm | 1μm | $1\times10^{10}$ |
| C(111) Monochromator | 35μm | 9μm | 3μm | $3\times10^{9}$ |

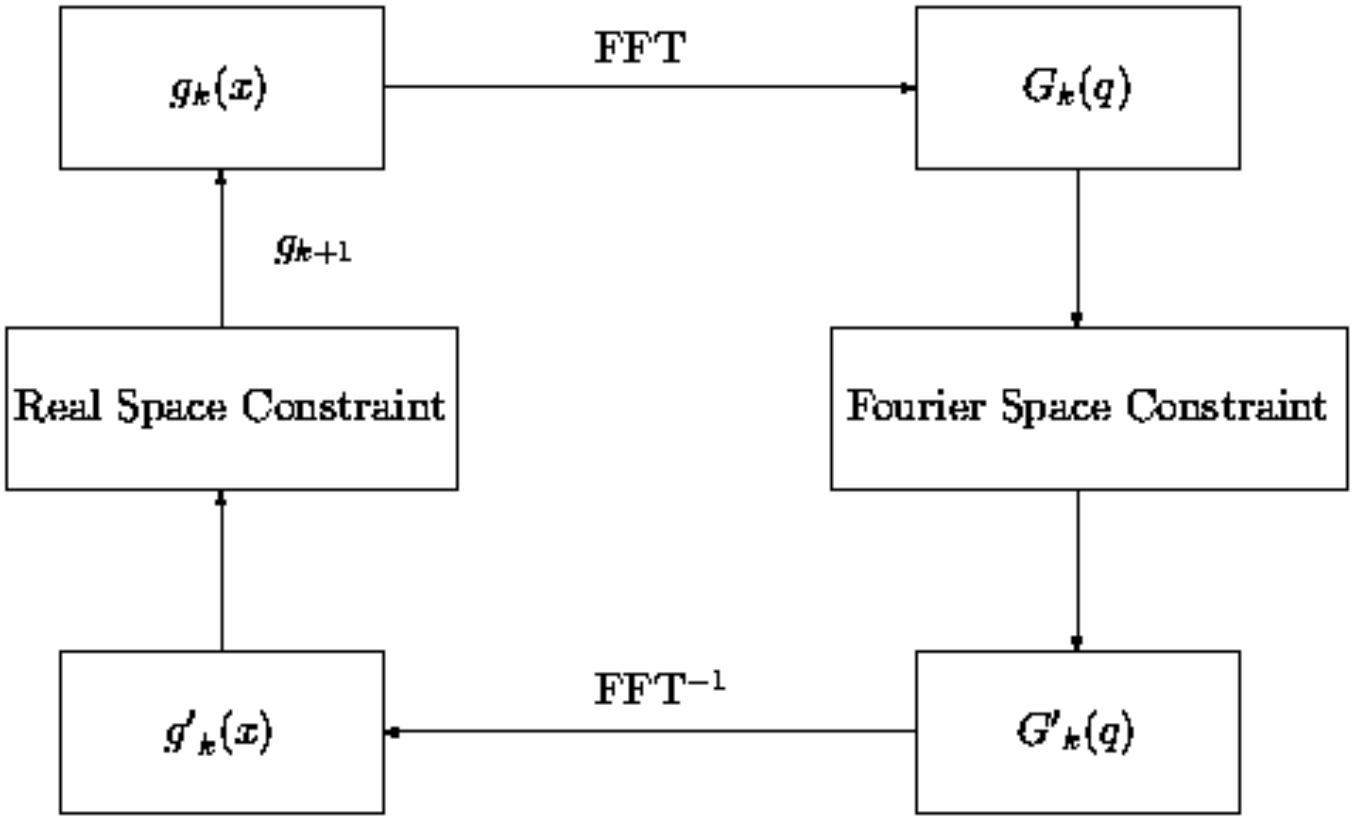
Coherent region defined by slits



# Diffuse Scattering acquires Structure using CXD



# Generic "Error Reduction" method



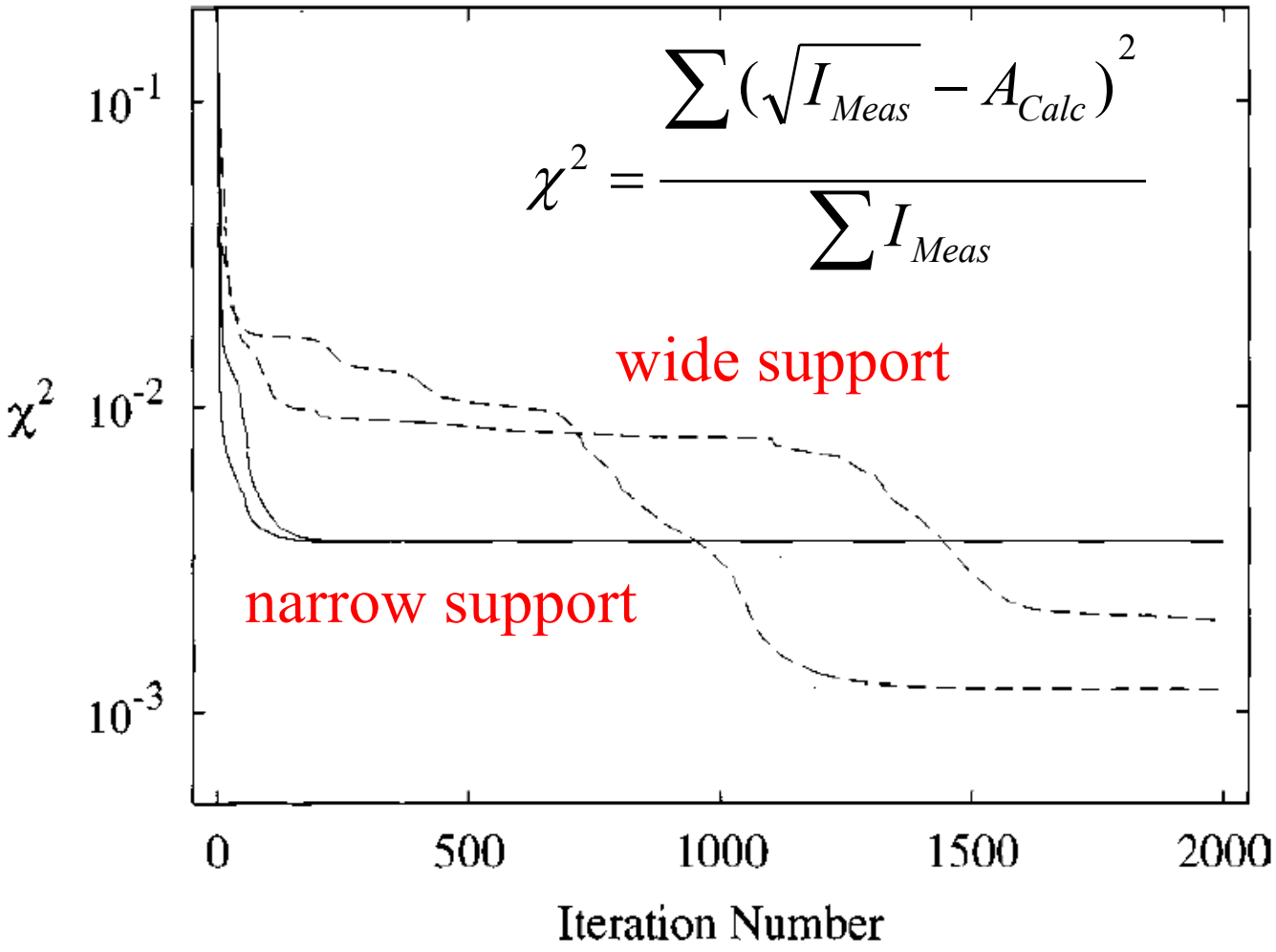

J. R. Fienup Appl. Opt. 21 2758 (1982)
R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)

# Real-space Constraints in Crystallograhy

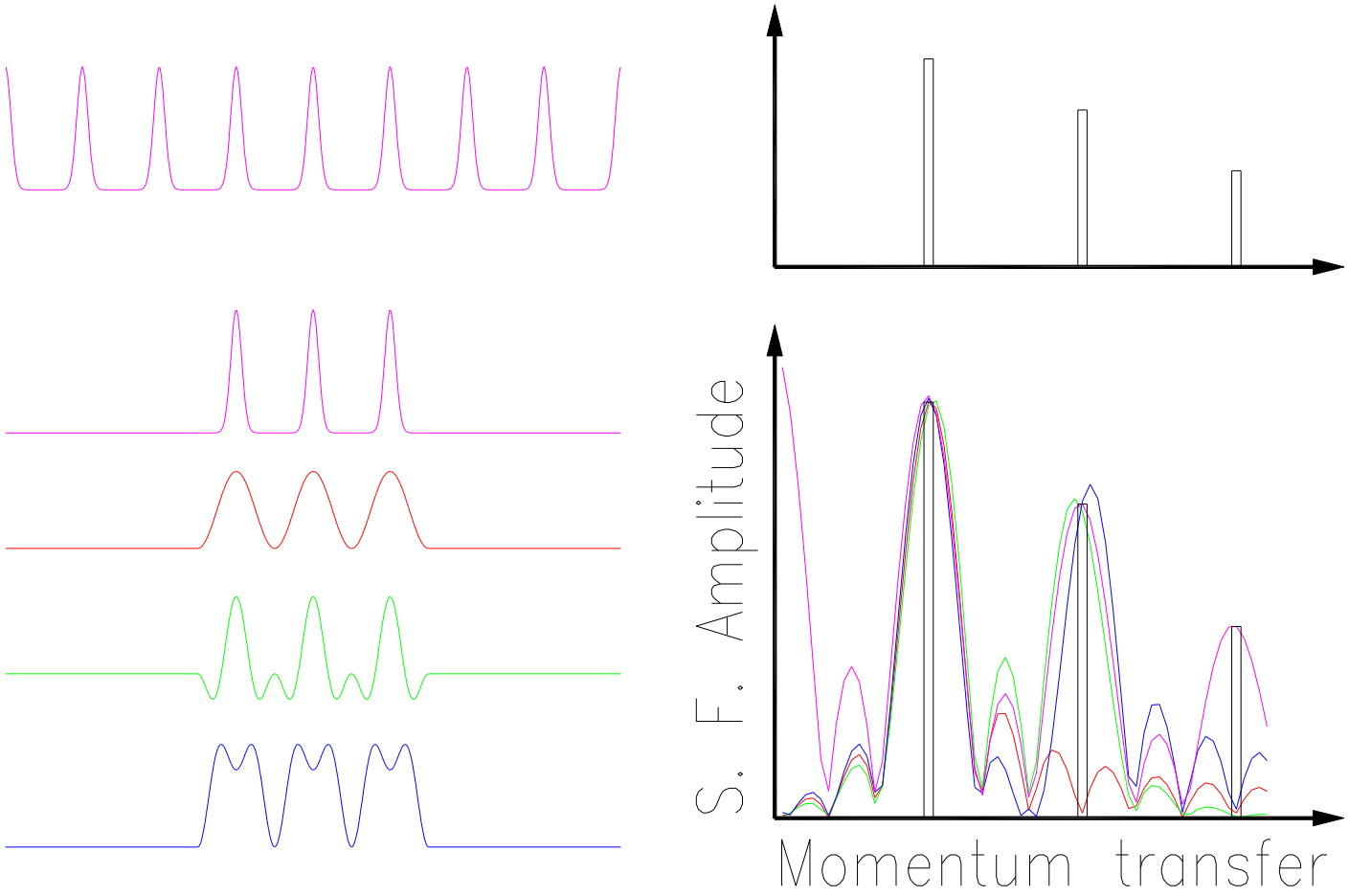
R. P. Millane, J. Opt. Soc Am. A **13** 725 (1996)

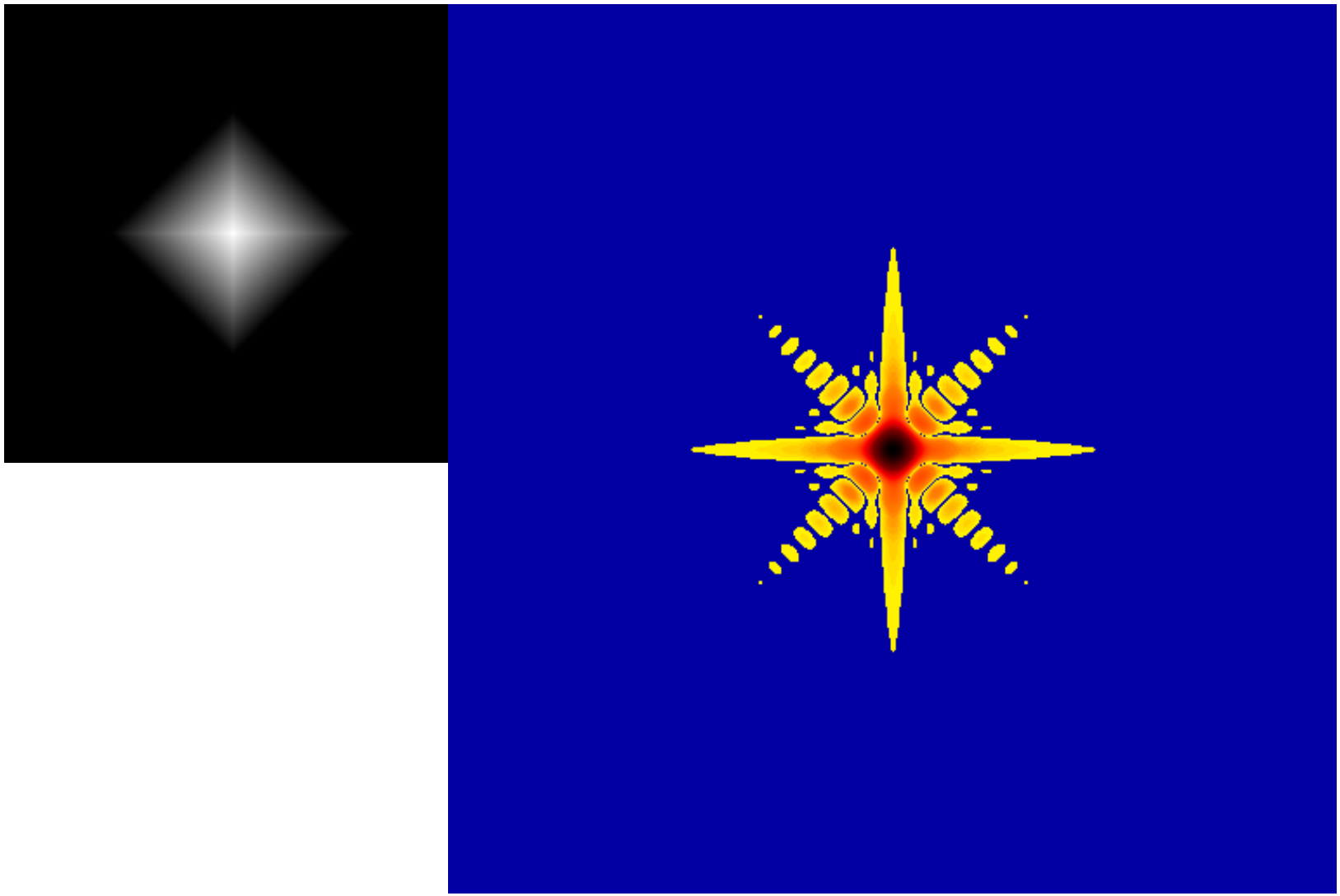
- 'Positivity', 'Atomicity' constraints (Sayre)
- Solvent flattening
- Non-crystallographic symmetry
- Molecular replacement
- Finite support = molecular envelope
- Non-uniqueness is 'pathologically rare' (d>1)

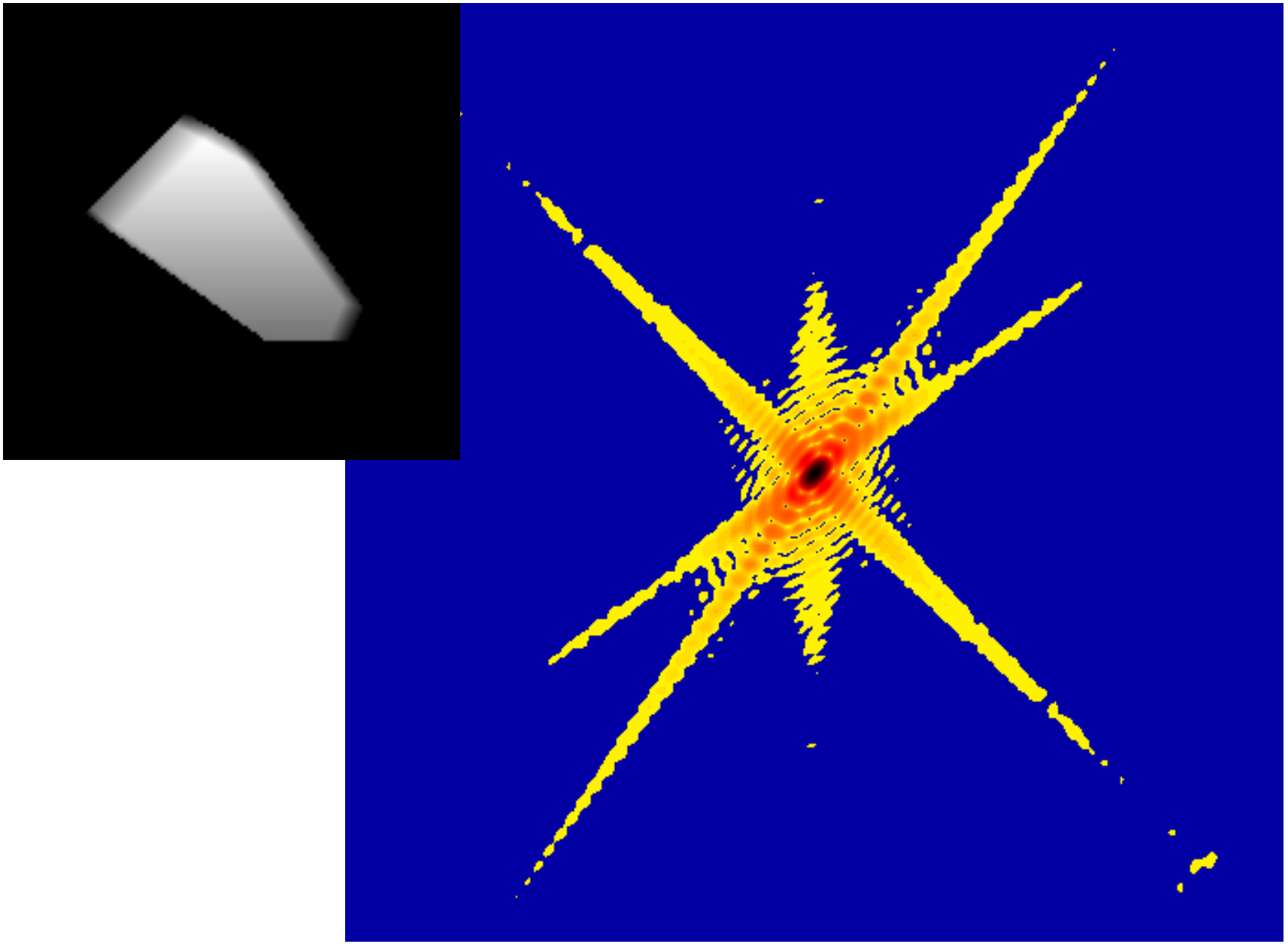
# Convergence Trajectory



# Phase Problem: Finite-size Effect

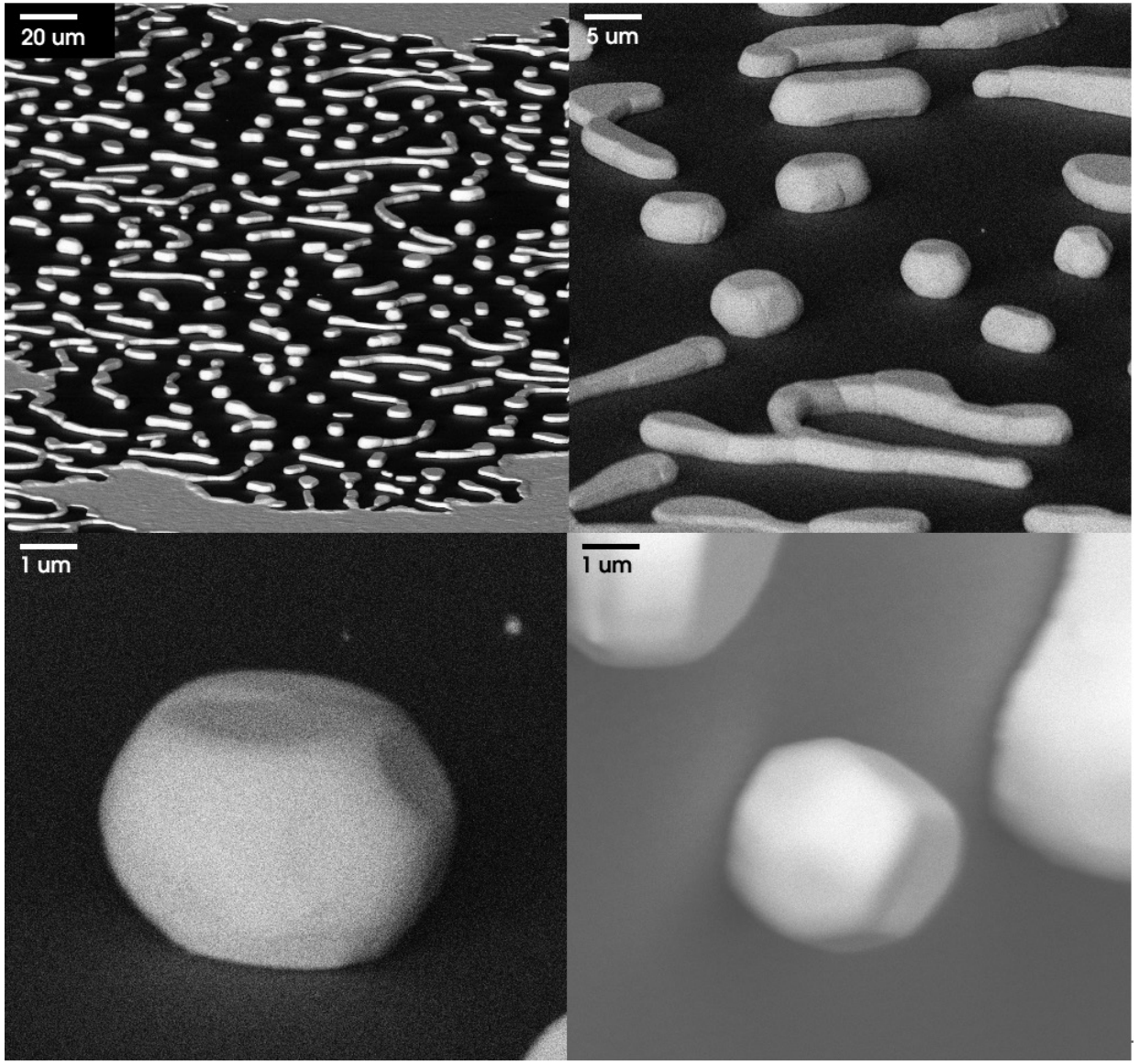




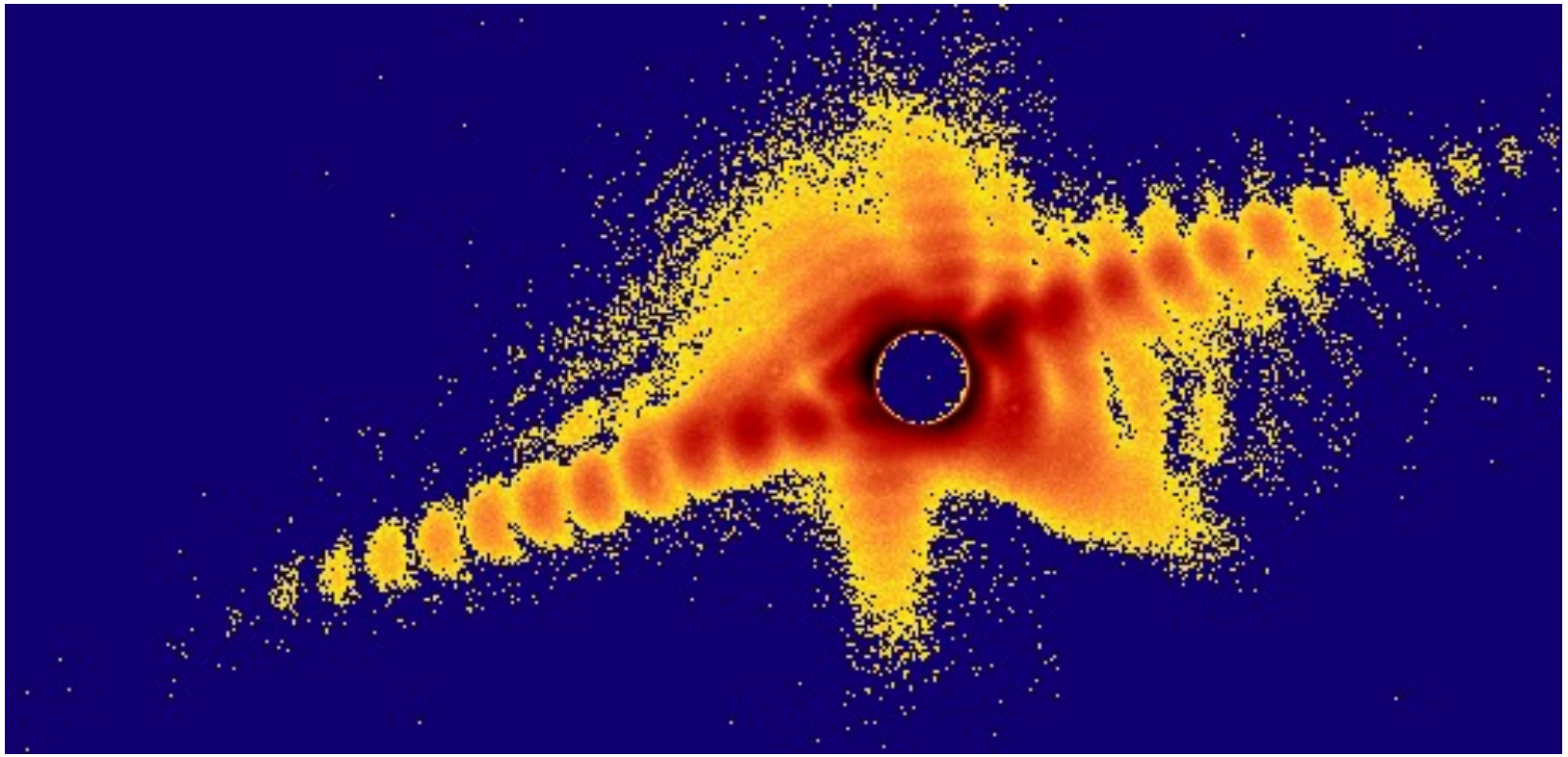


# SEMS

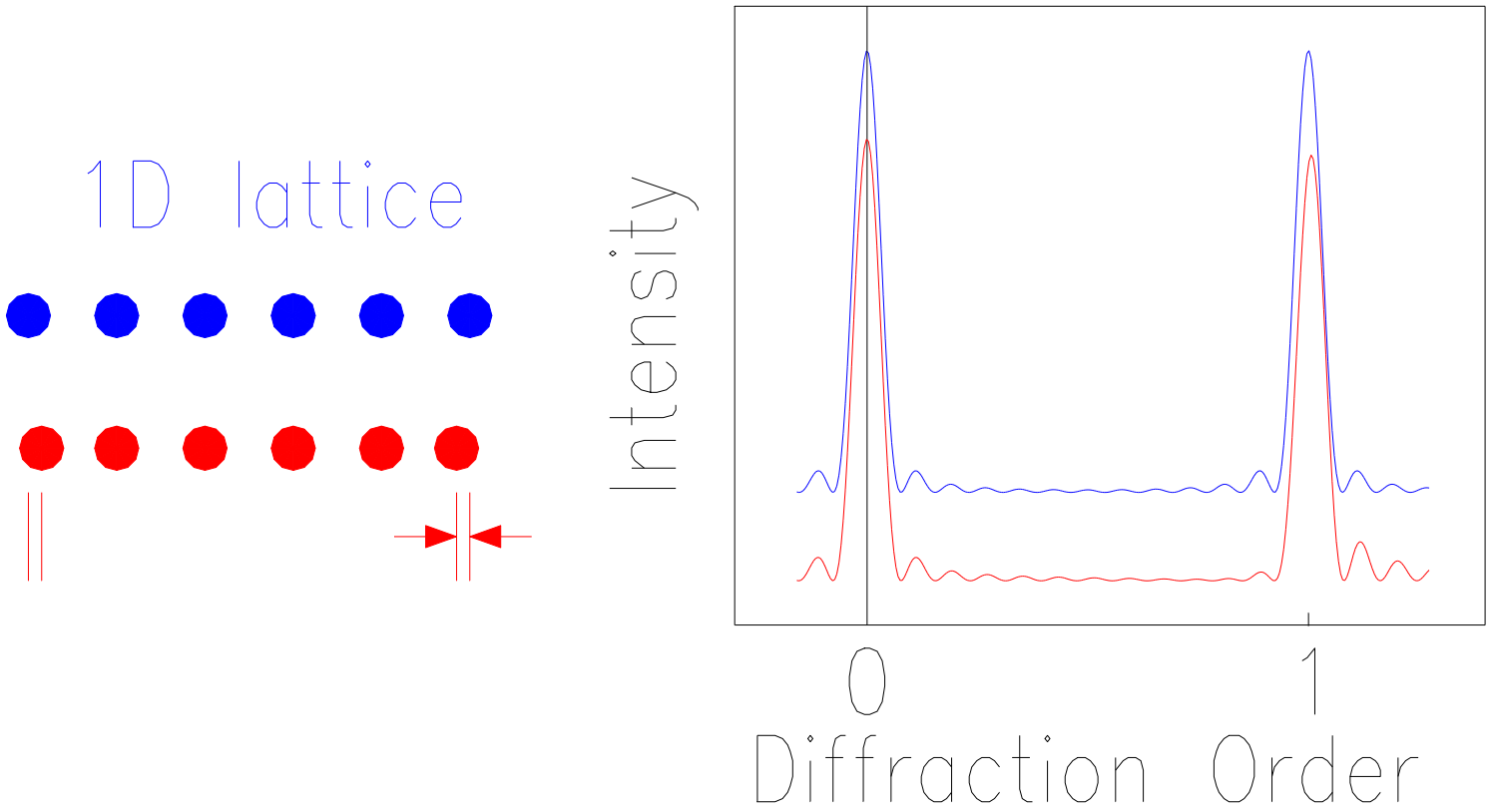
- Au blanket film
- Quartz substrate
- Annealed at 950°C for 70 hrs.



# Micron-sized gold crystal: (111) Bragg reflection

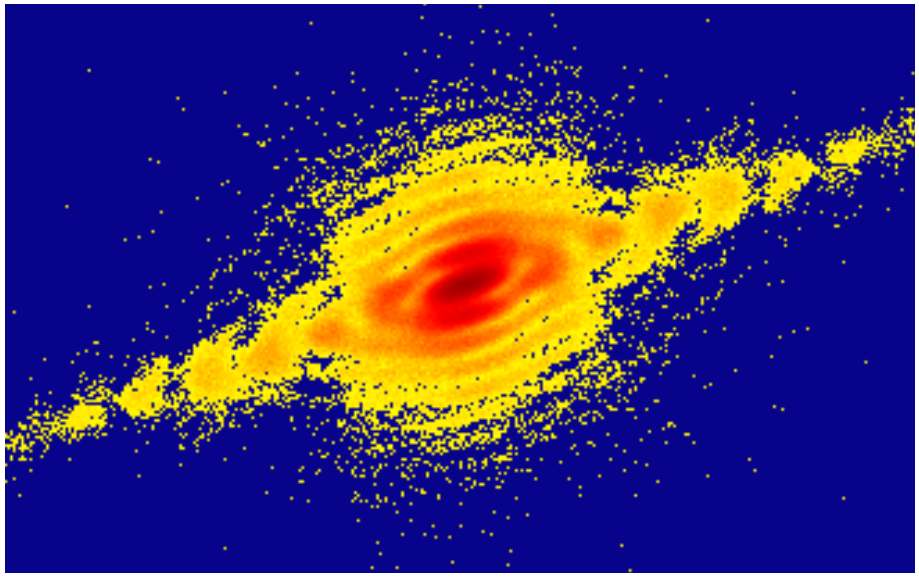


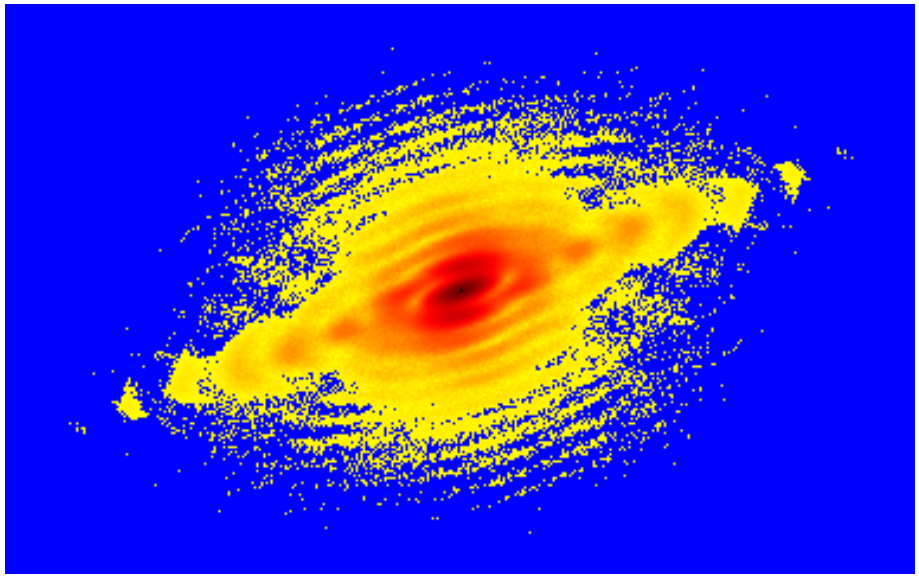
# Imaging of Lattice Strains

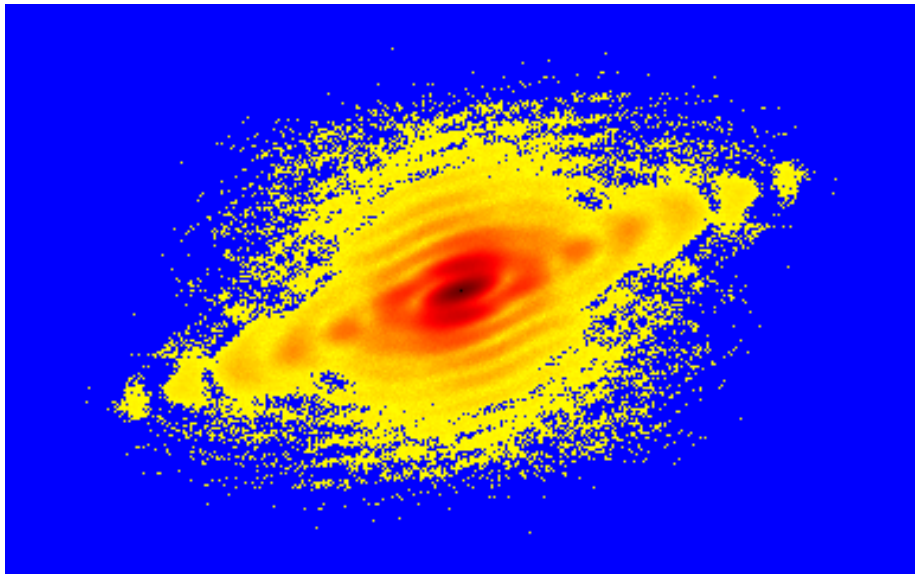


# Symmetrized Data and two best fits

Chisq=0.0005

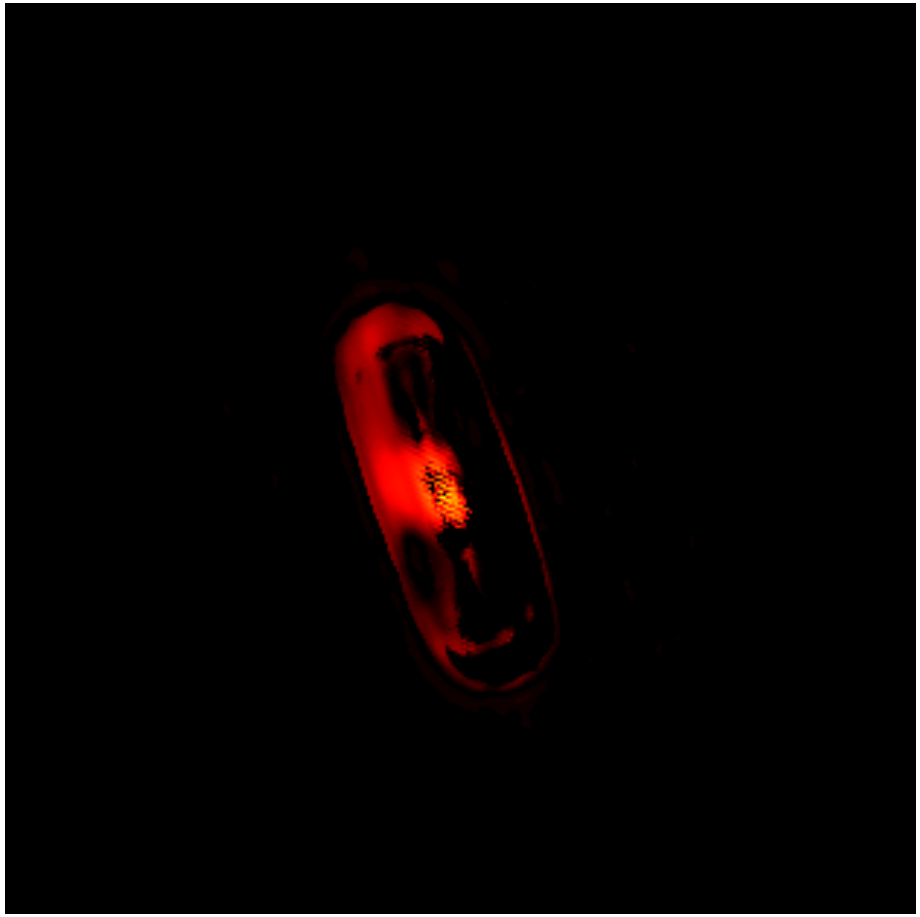


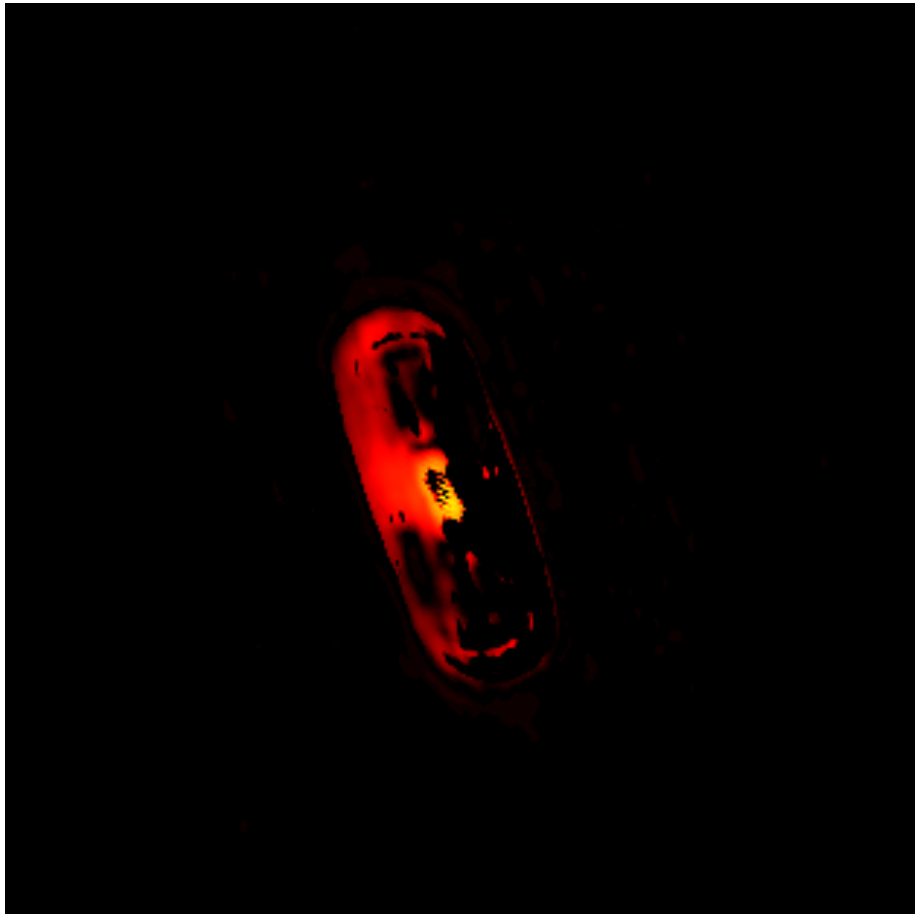




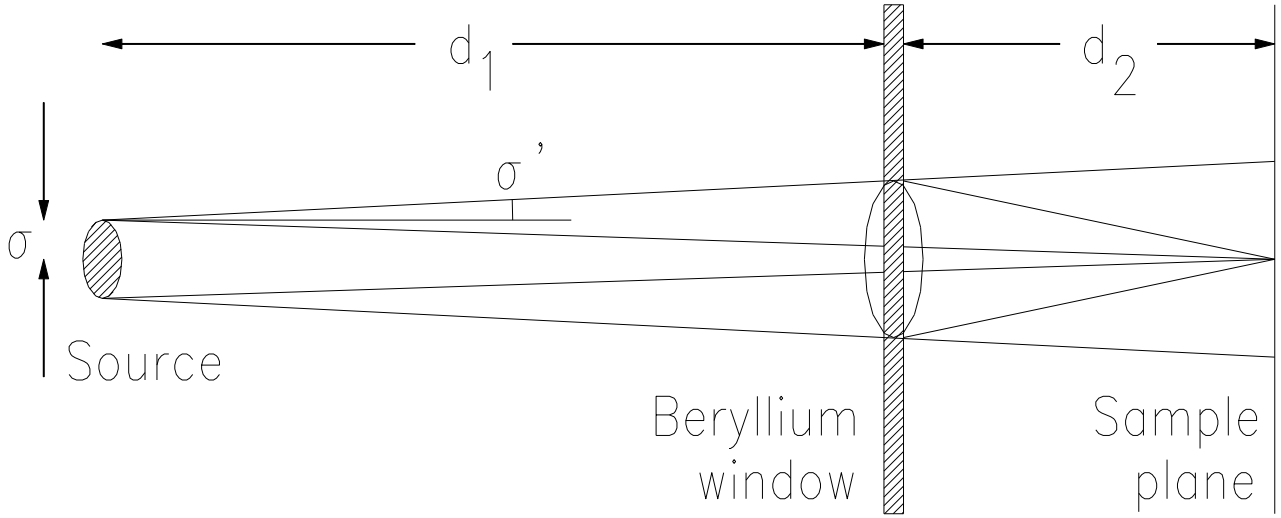
# 2D Reconstructions

## chisquare = 0.0005





# Window as a Secondary Source

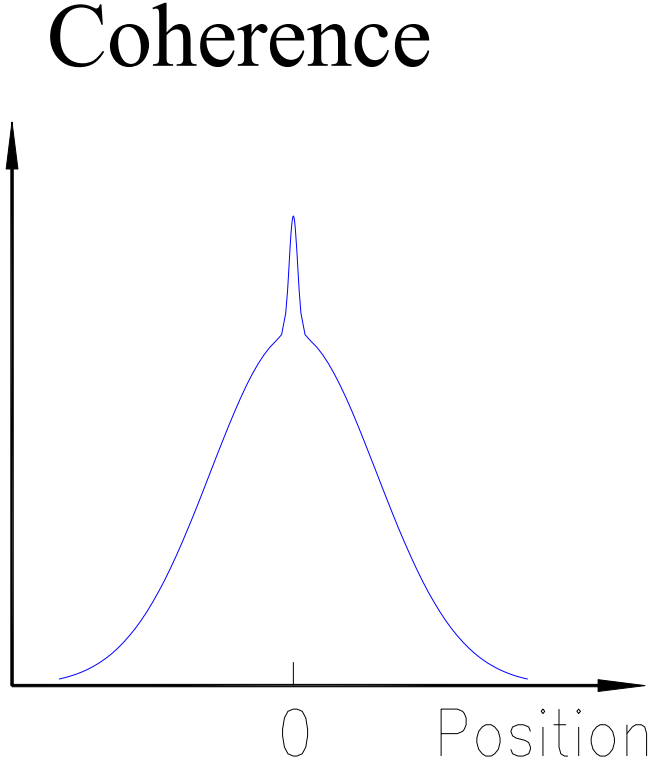



$$L_T = \frac{\lambda D}{2\sigma}$$

$$L_{T2} = \frac{\lambda D}{2\sigma} \frac{(1-\frac{d_1}{D})}{(1+\frac{\sigma' D}{\sigma}\frac{d_1}{D})}$$

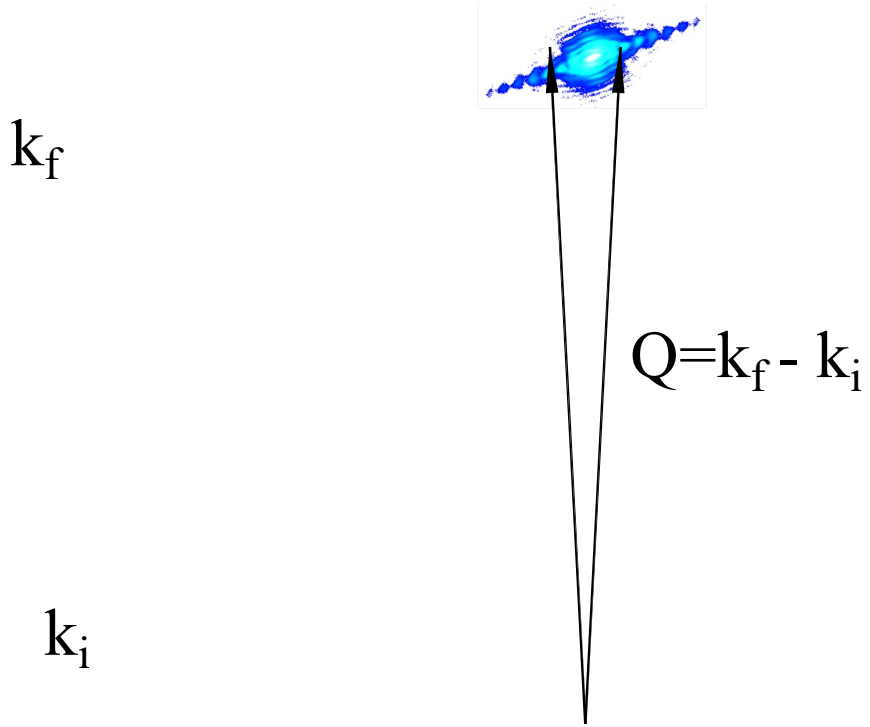
Vert ~50
Horiz ~3

# Two Components to MCF

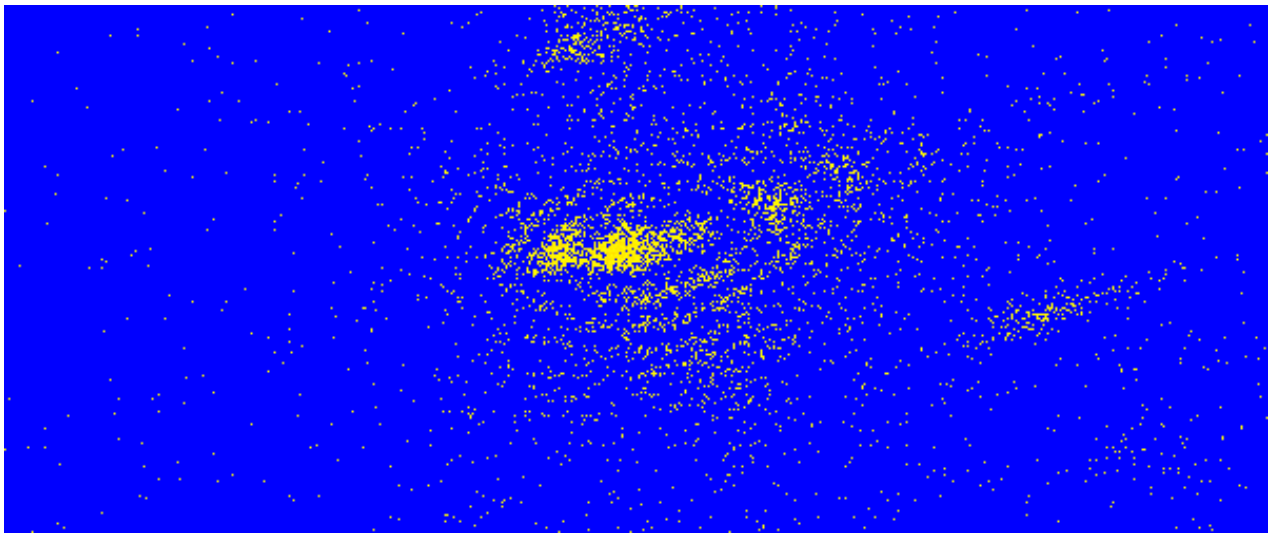



- Width of narrow component is very sensitive to location of window
- Amplitude of narrow component depends on level of diffuse scattering from window
- Amplitude increases with distance (for phase object)

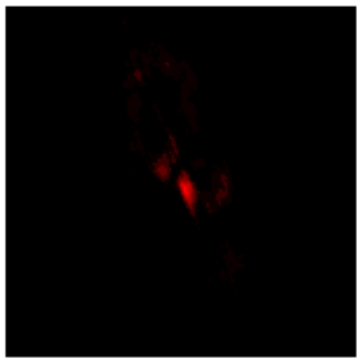
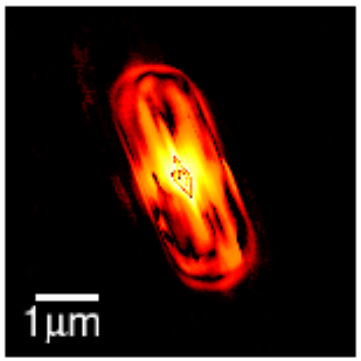
# 3D Diffraction Method

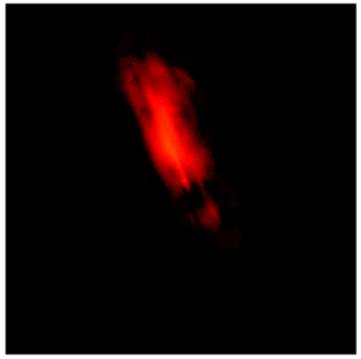
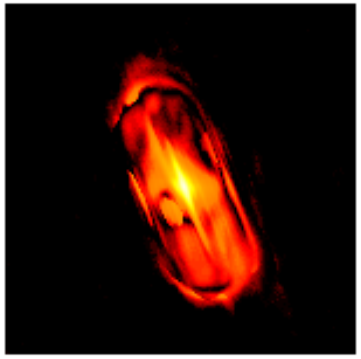
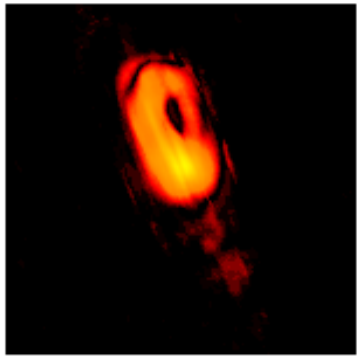
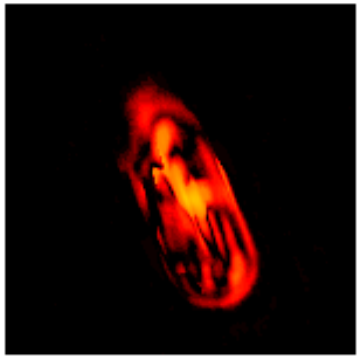
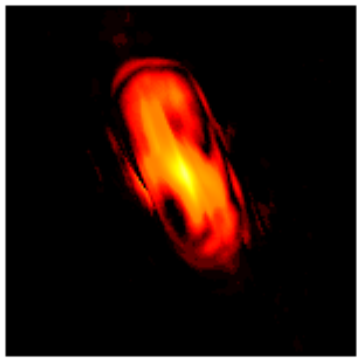
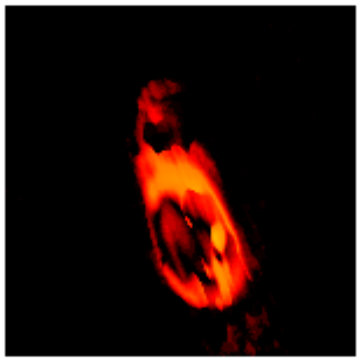
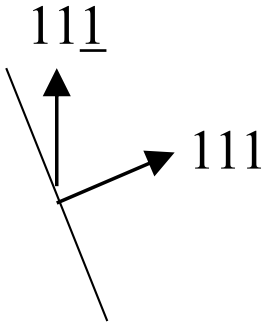


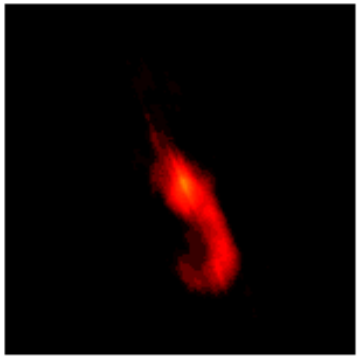
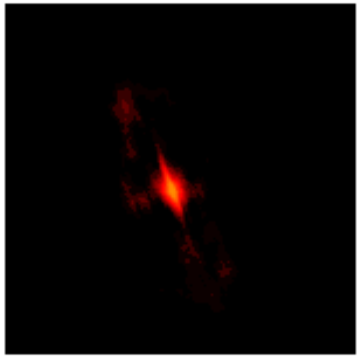
# 3D Diffraction Data
# 1 micron Au crystal



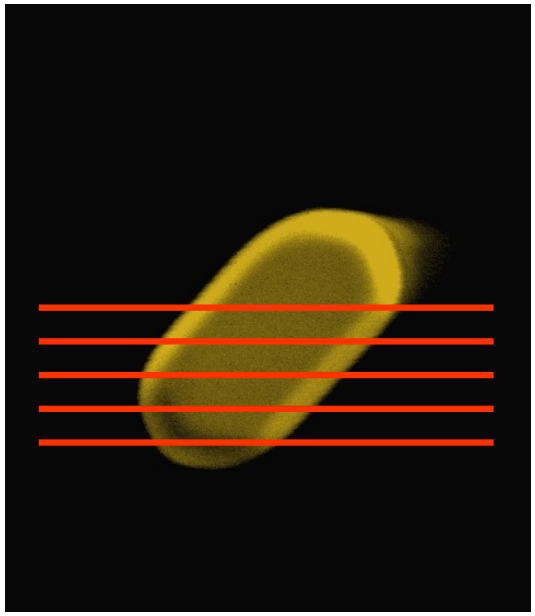
* Center is Symmetric *

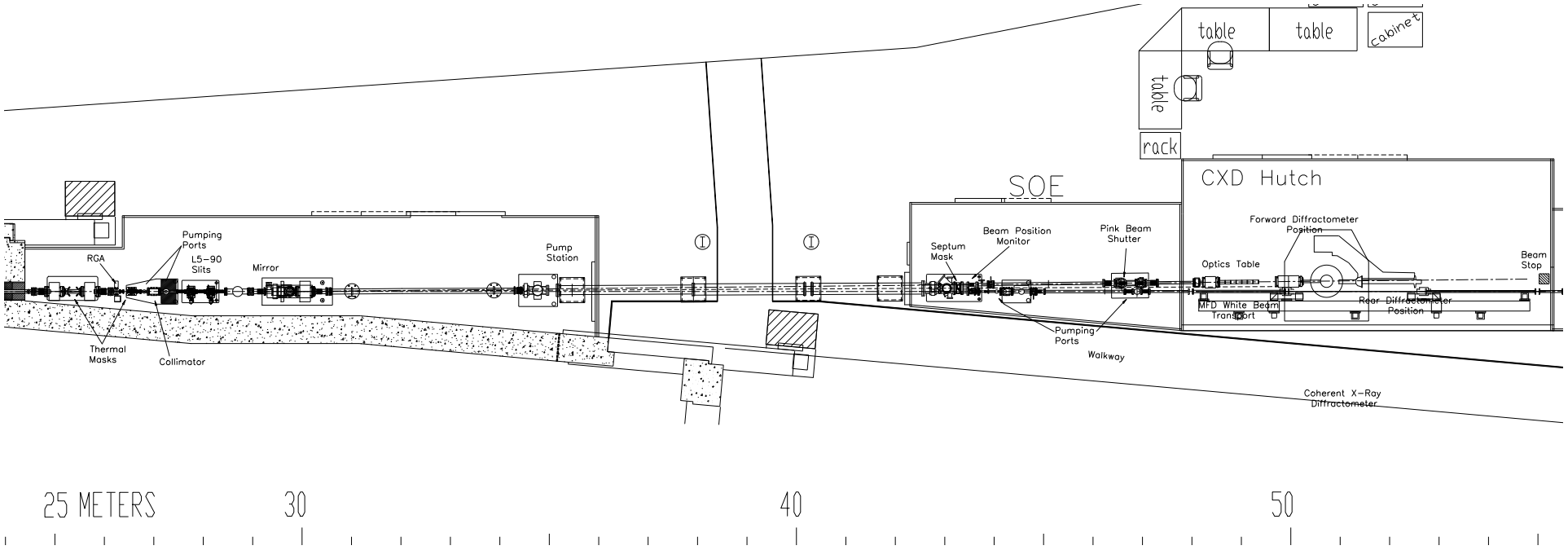








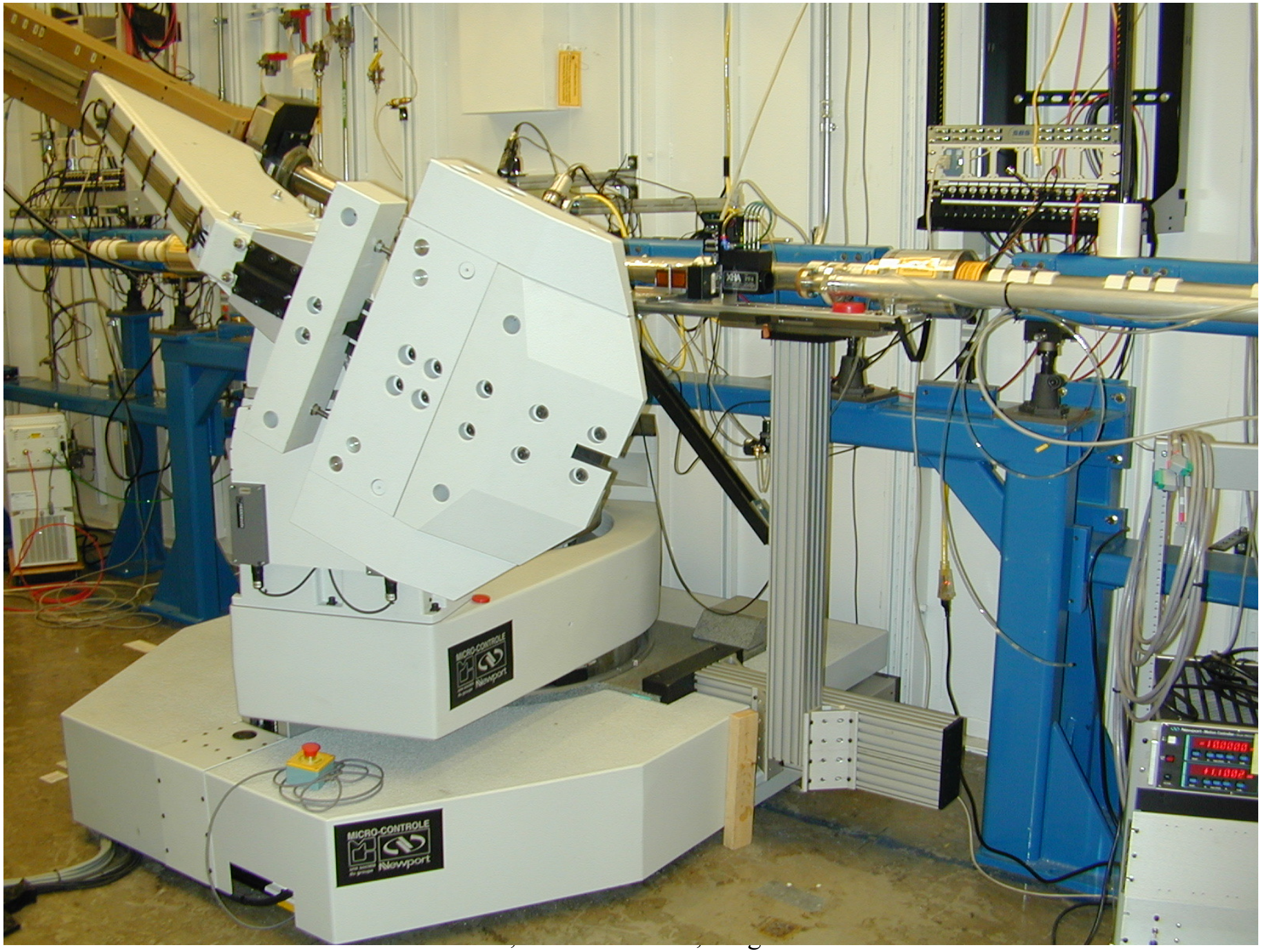



Slices through plan view SEM:

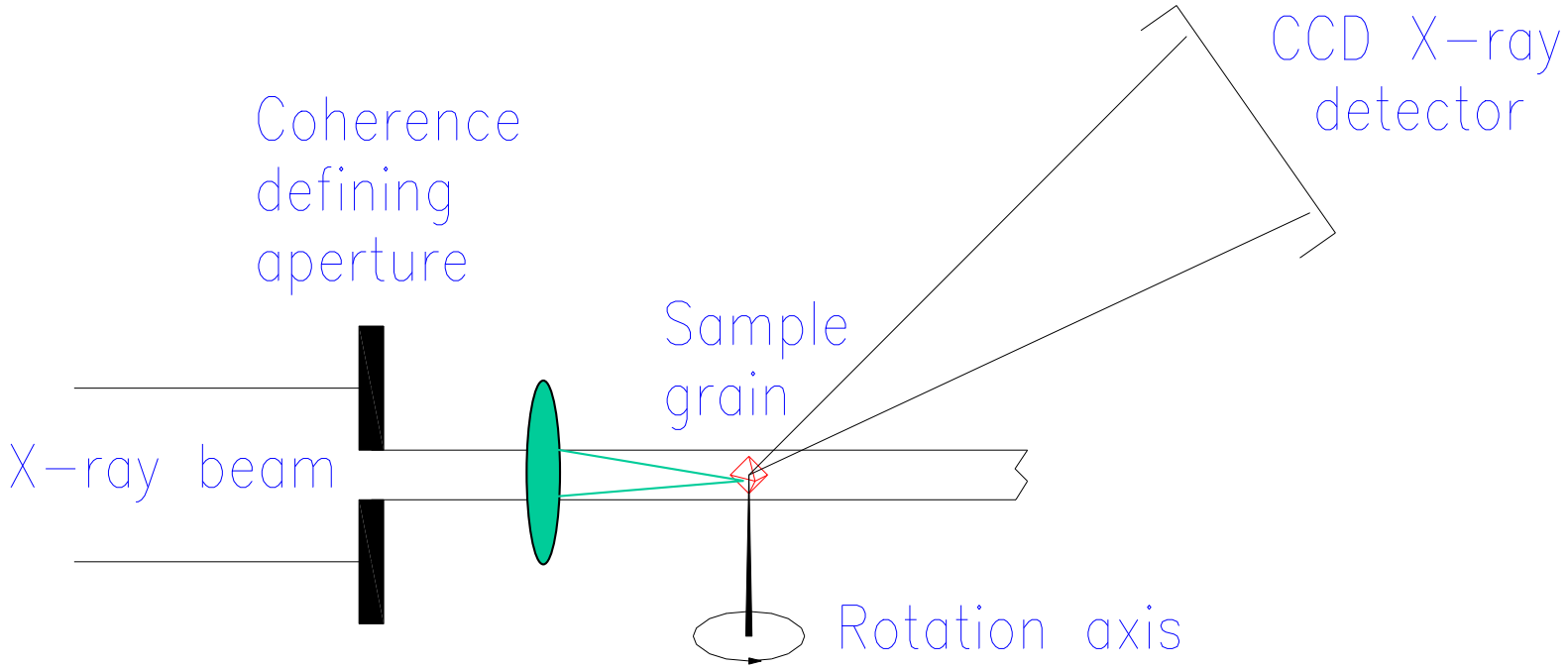


# CXD Beamline at APS Sector 34

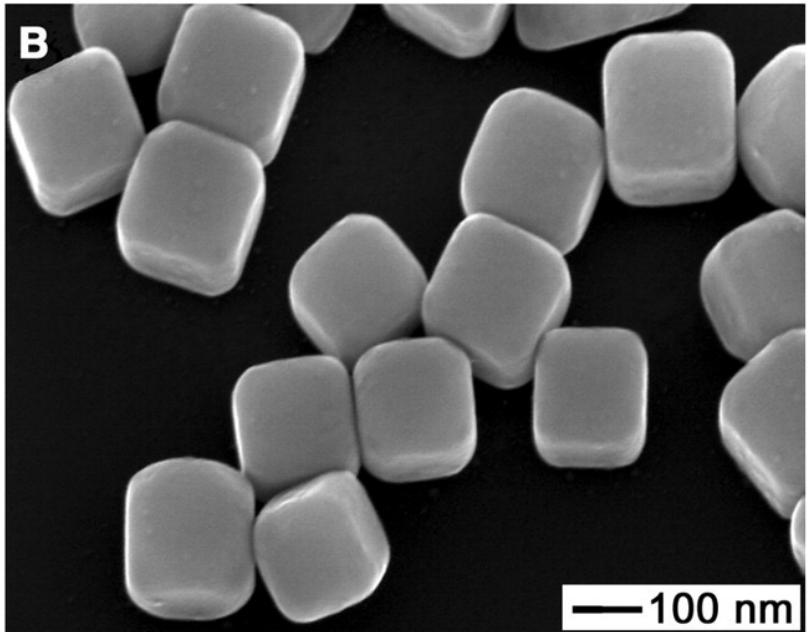


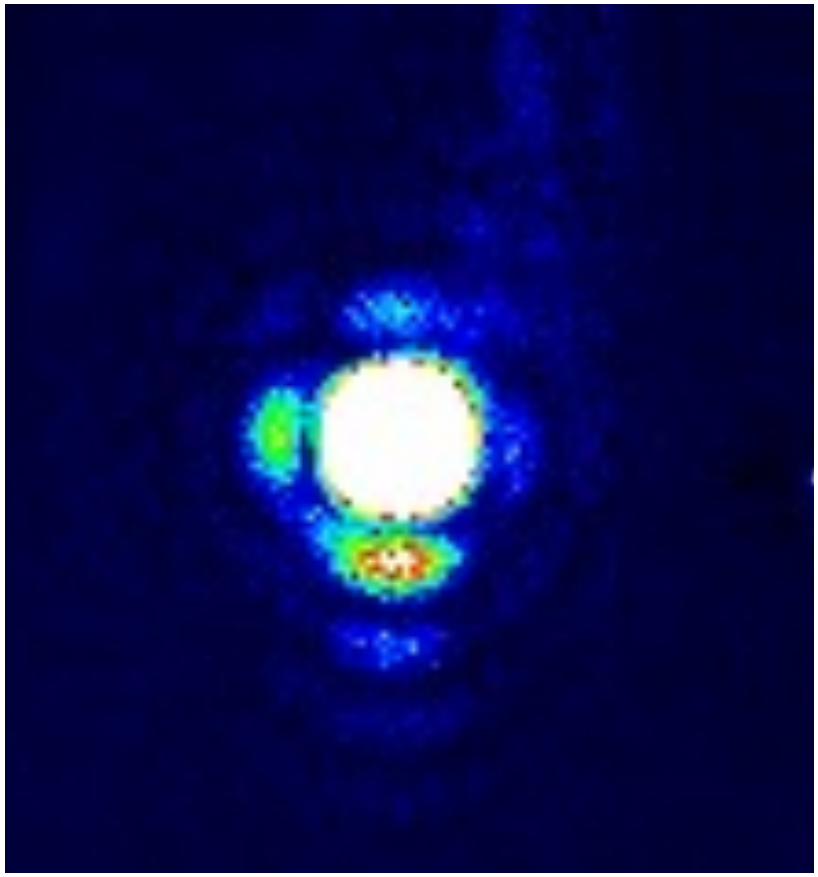

MICRO-CONTROLE
Newport
MICRO-CONTROLE
Newport

# Lensless X-ray Microscope



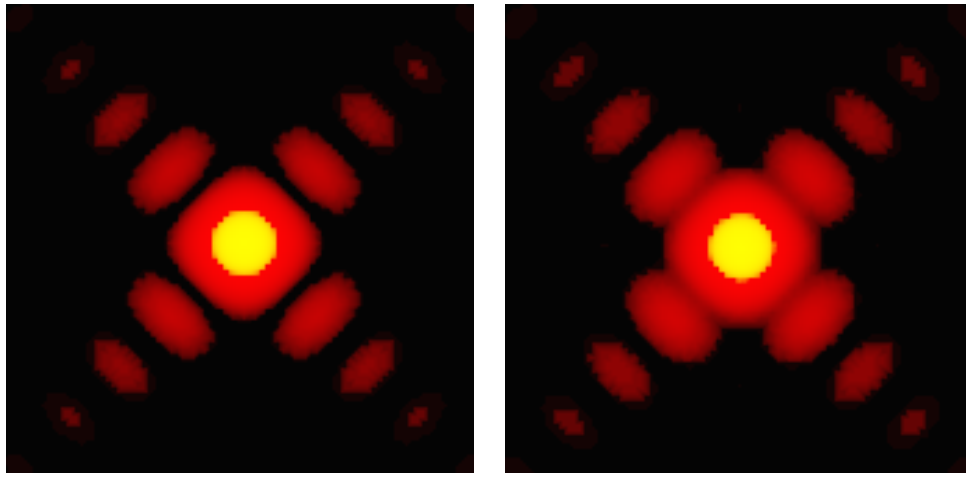
# CXD from Silver Nanocubes



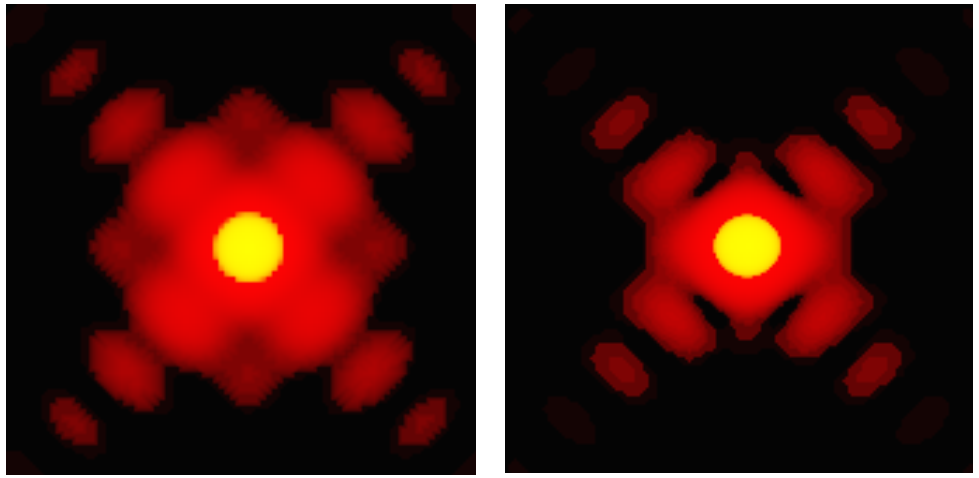
Yugang Sun and Younan Xia,
Science 298 2177 (2003)

# Spherical and cylindrical waves



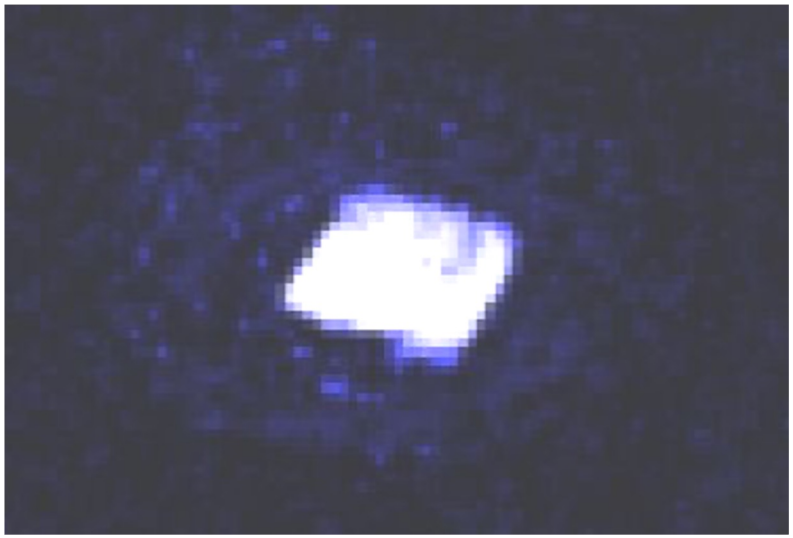
π/4

π/2

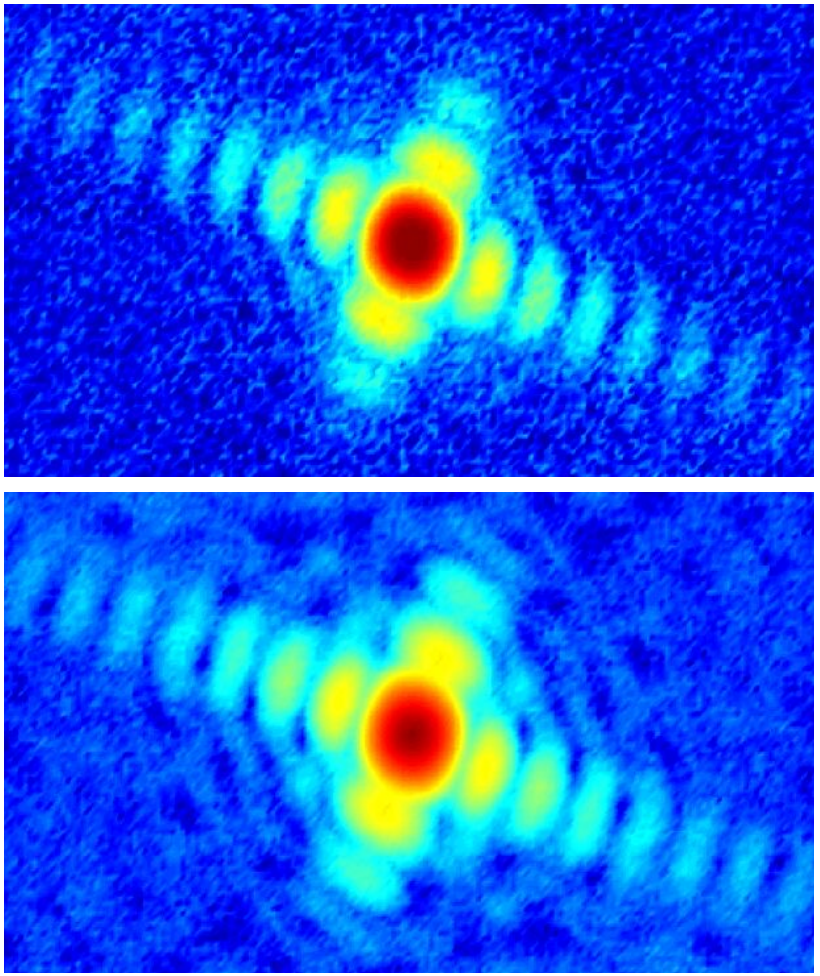


π/4 (x)

# Reconstruction of Ag Nanocrystal

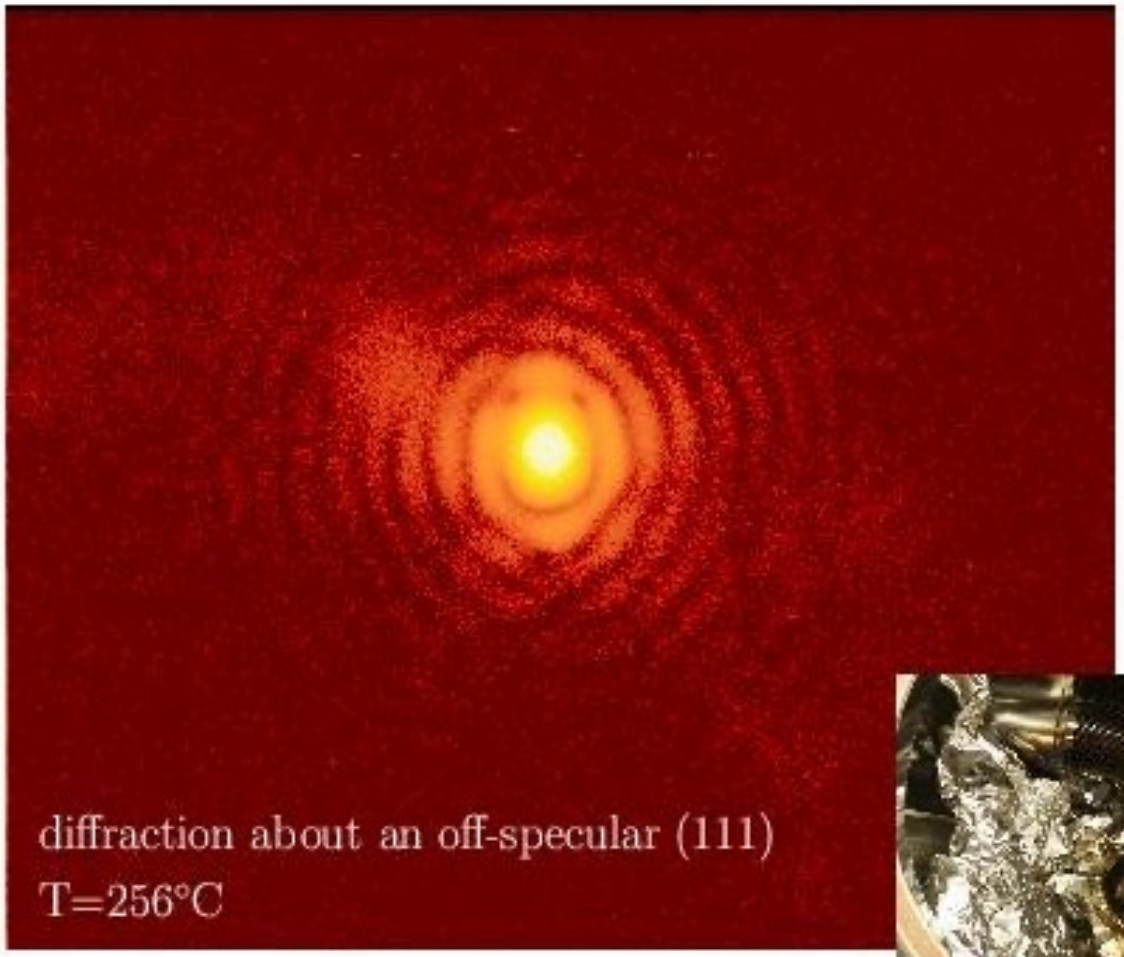


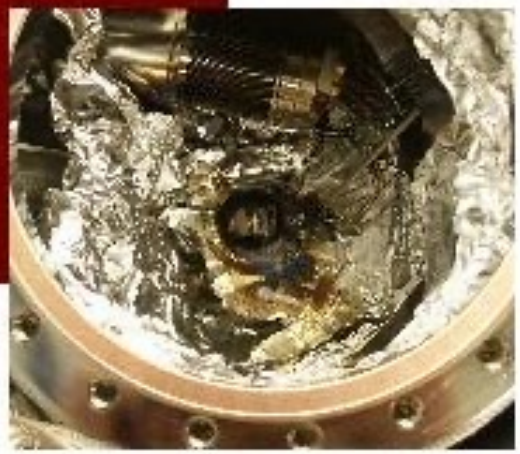
200nm



# 0.7μm Pb Nanocrystal

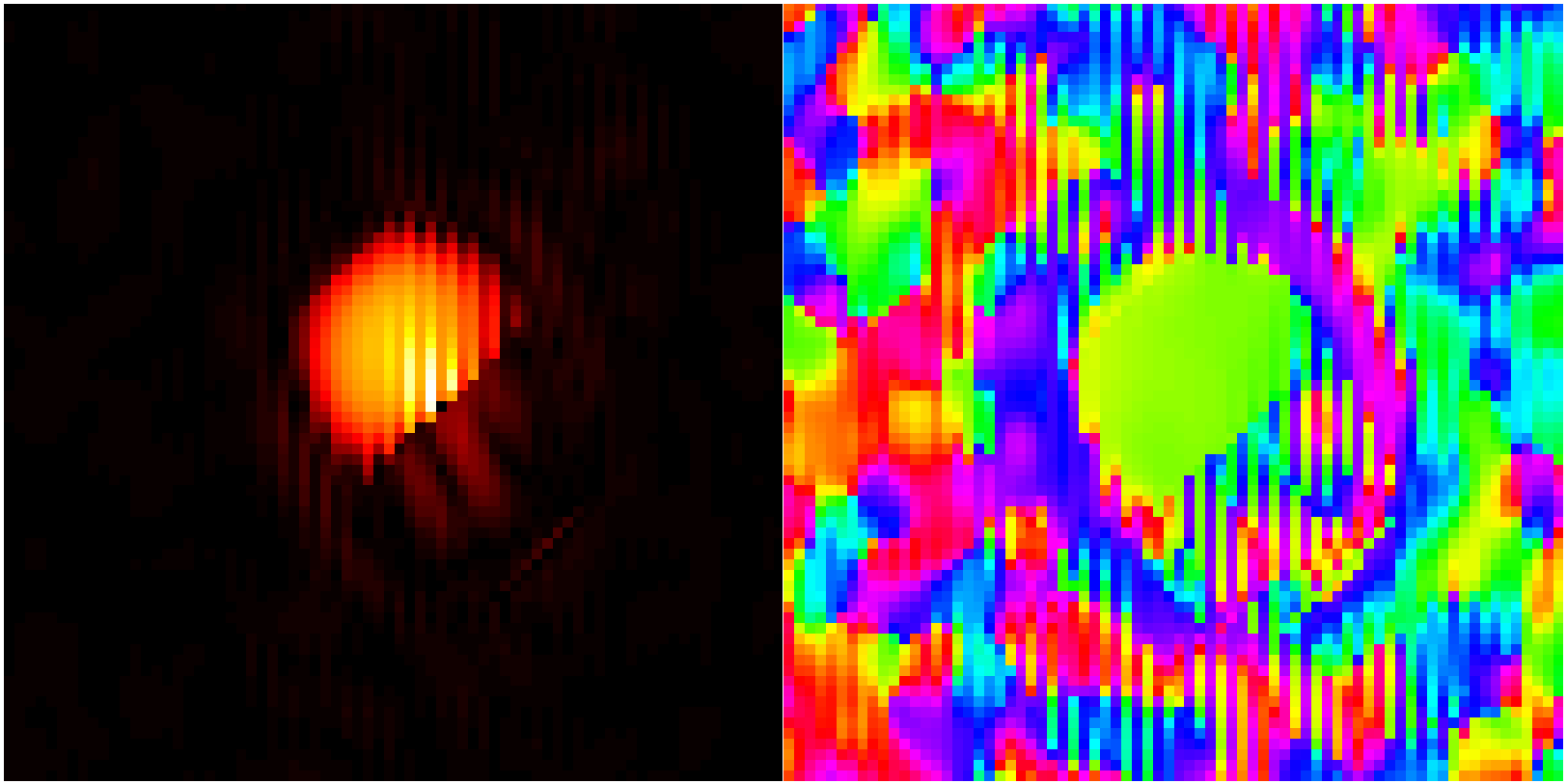
## In-situ growth at APS sector 34



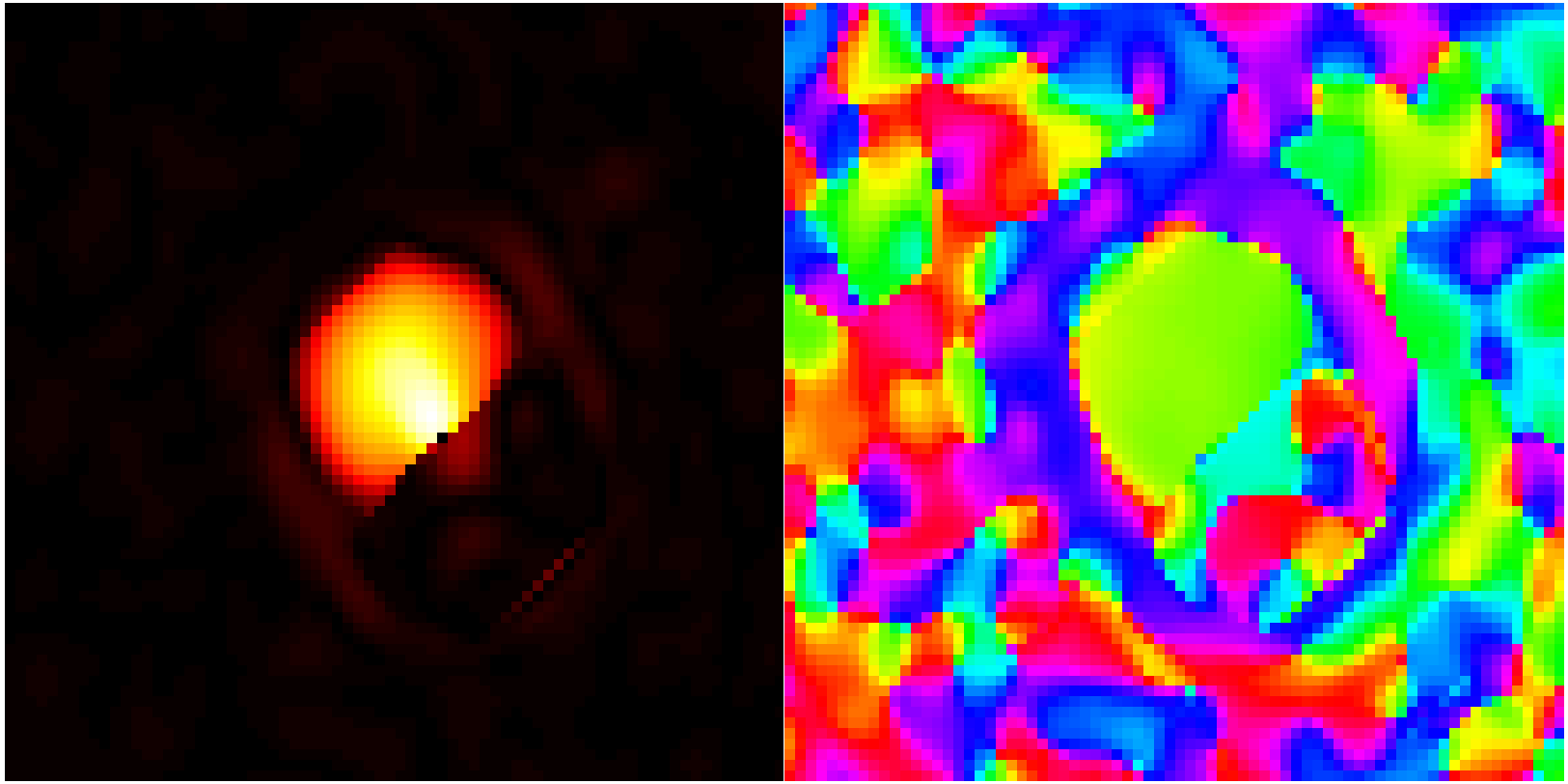



# Incomplete Reconstruction can be Striped

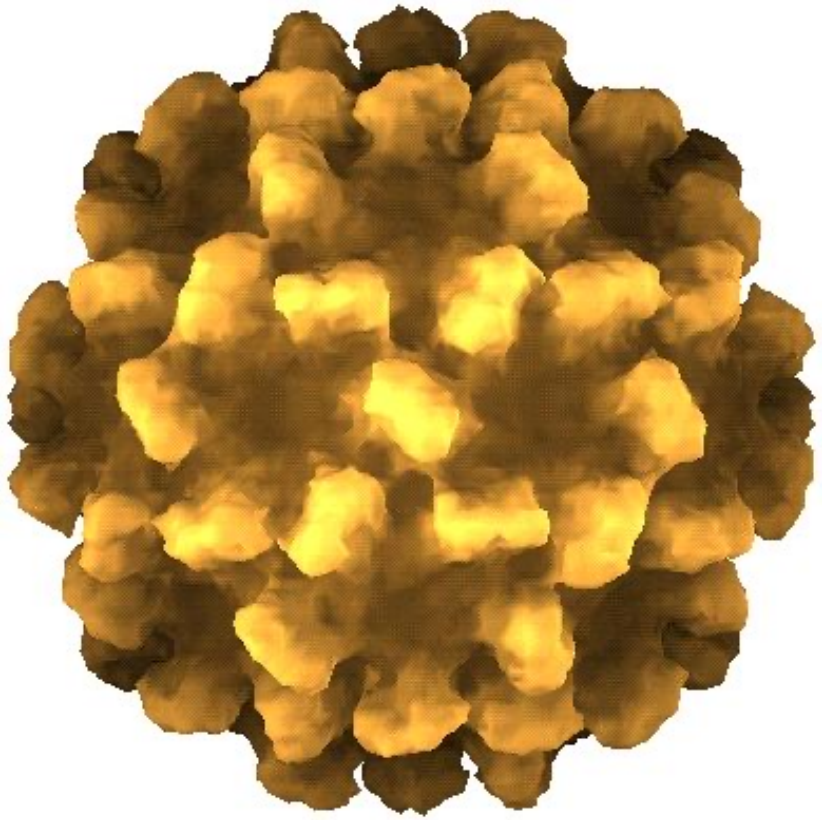
## 0.5 micron Pb crystal on $SiO_2$ substrate



# Result of “Patching” in 2D



# Tomato Bushy Stunt Virus
# 1980



# Conclusions and Outlook

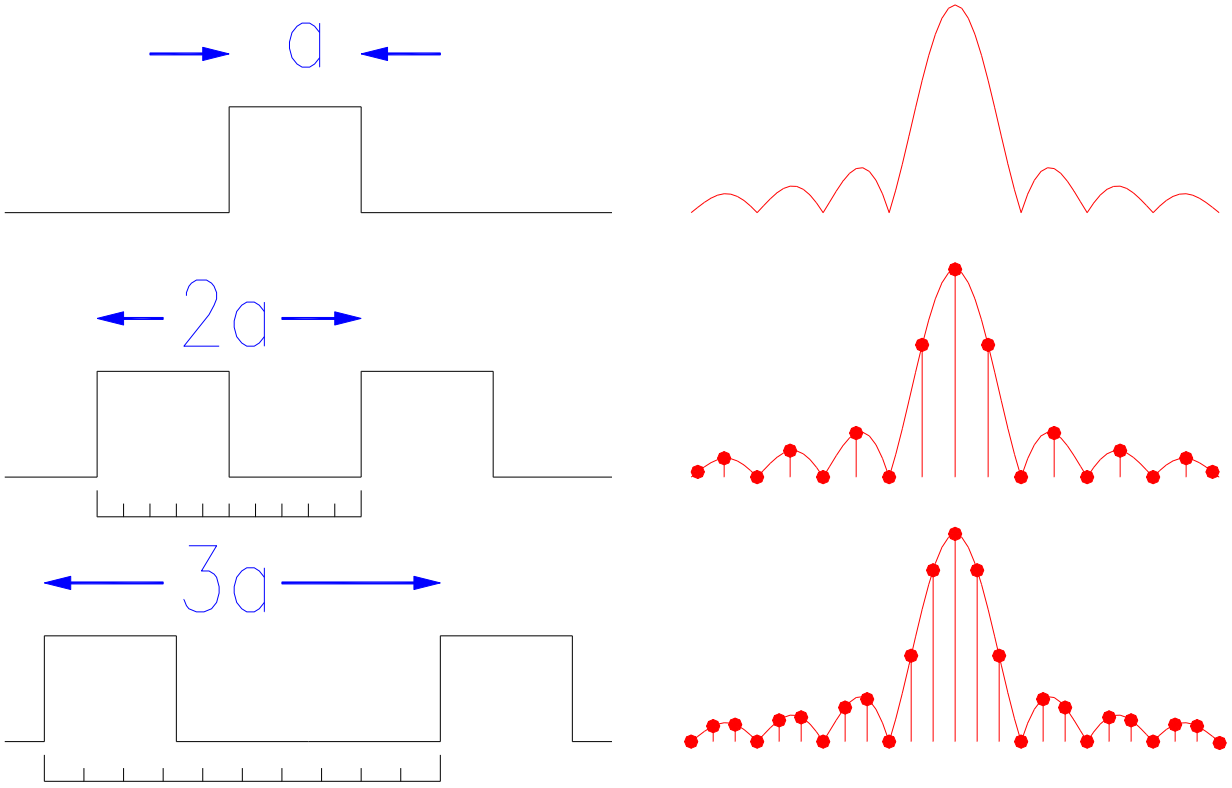
- Diffuse scattering acquires fine structure
- Inversion of CXD demonstrated
- Internal structure in Au (not Pb, Ag) nanocrystals
- Preservation of coherence upon focussing
- Small crystals possible now;
- Single molecules one day with XFELs

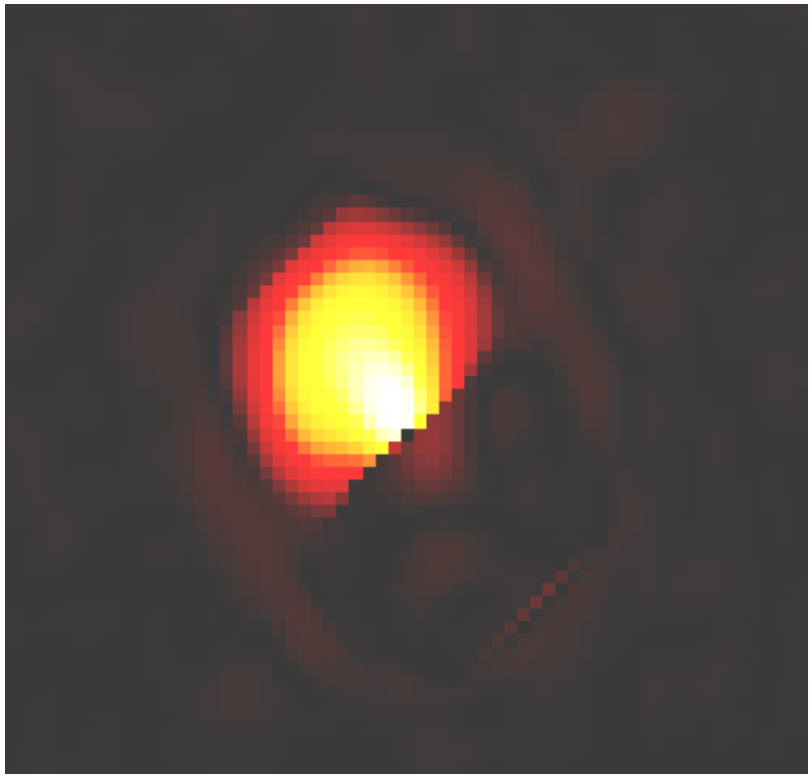
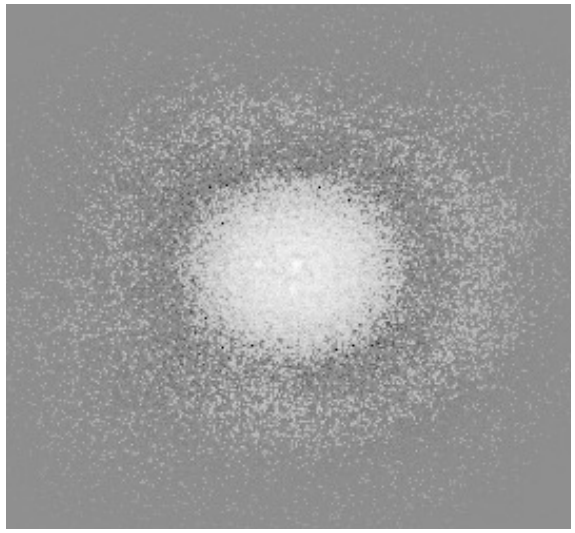
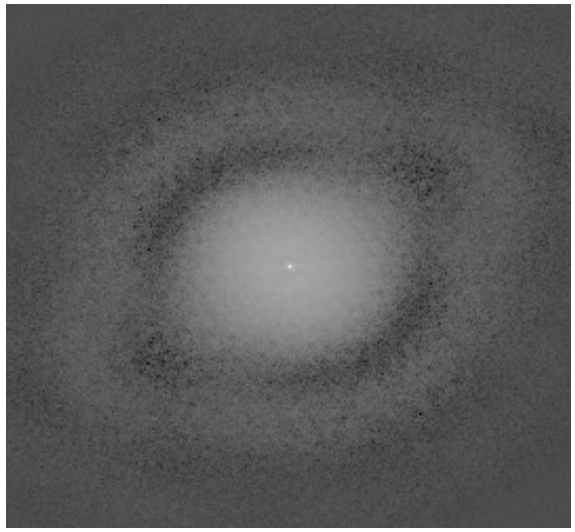
# Oversampling for Small Crystals

## DFT connects N points with N reciprocal points

N points in 2a:
N/2 unknowns

N points in 3a:
N/3 unknowns




Always N/2 measurements

# Reconstruction of Pb Nanocrystal