Opportunities for Coherent X-ray Diffraction at Diamond Light Source

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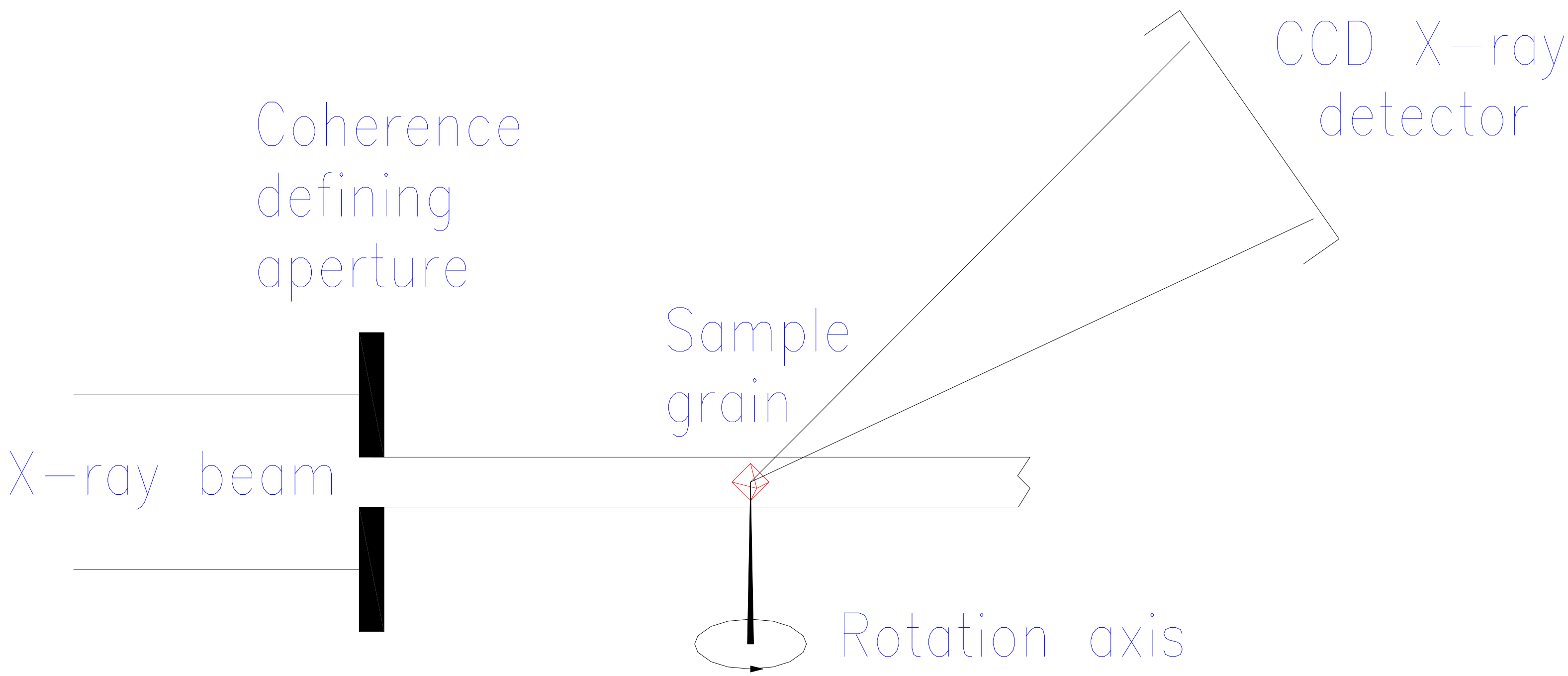
Outline

- Coherence in Diffraction
- The Phase Problem
- Nanocrystal Shapes
- Crystallization of Proteins
- Future Applications
  - Smaller Nanocrystals
  - Dislocation Structure
  - Quantum Dot Structures
  - Single-Molecule Imaging with XFEL

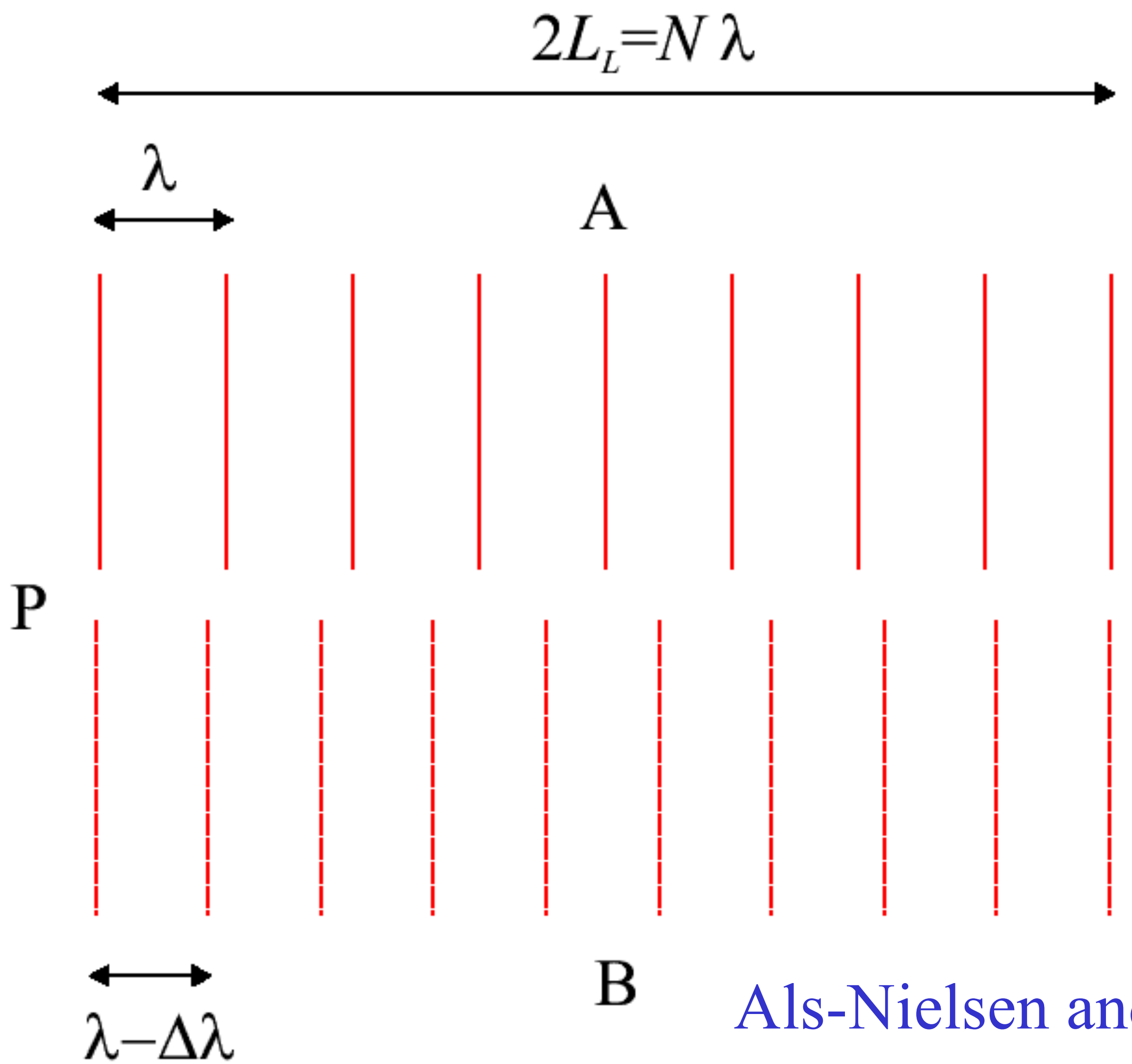
Goals of Coherent Diffraction

- 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
based on diffraction from crystal lattice



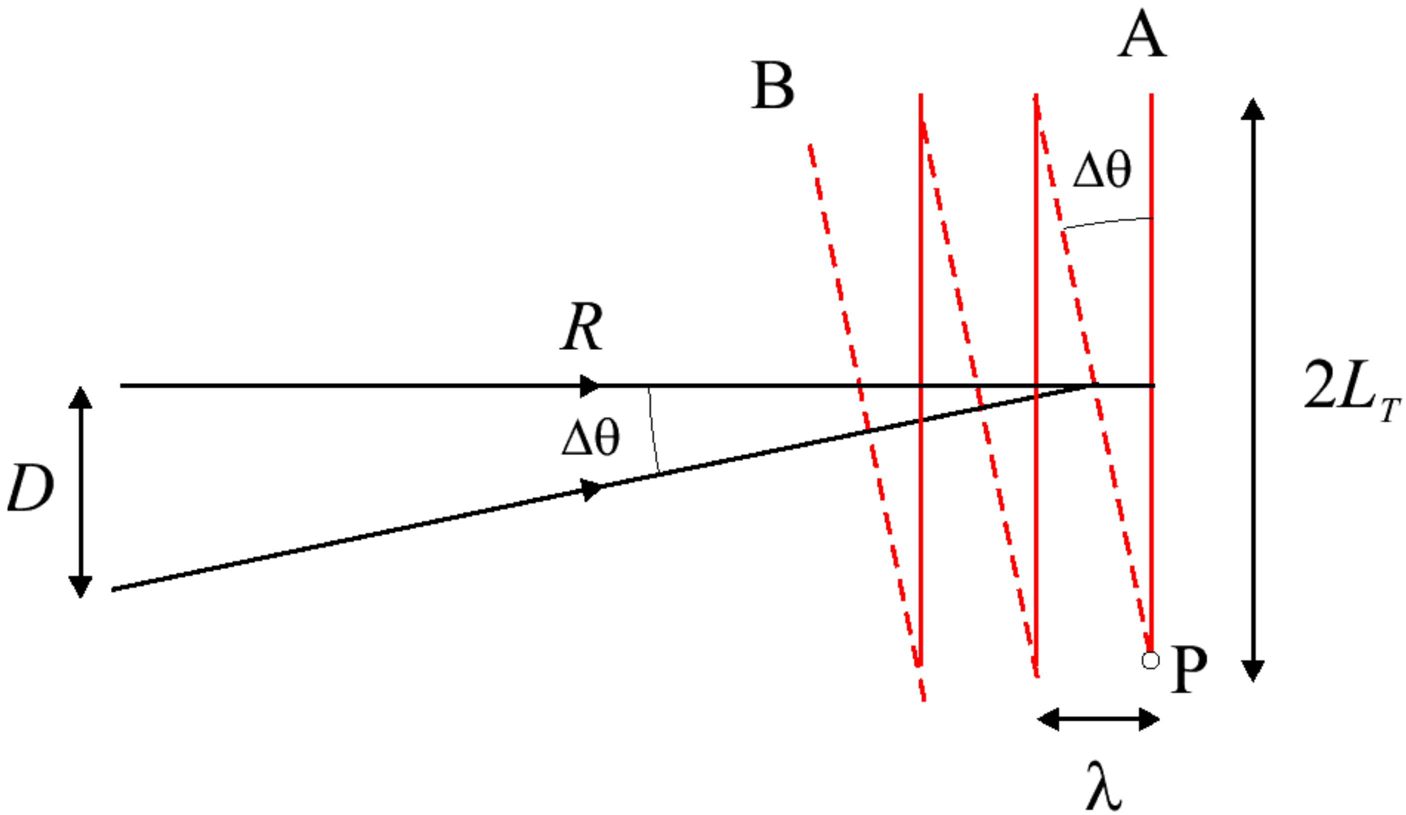
Longitudinal Coherence



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

Als-Nielsen and McMorrow (2001)

Lateral (Transverse) Coherence



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

Als-Nielsen and McMorrow (2001)

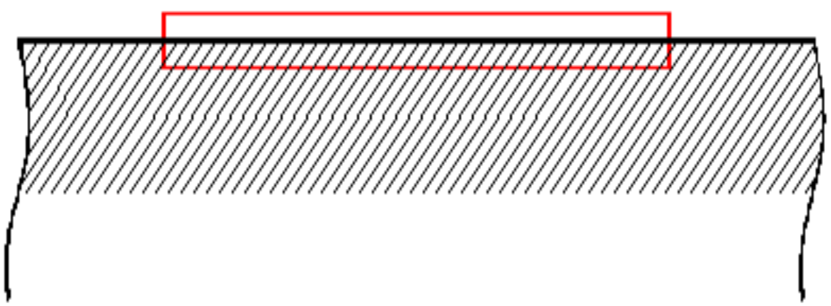
Coherence at the APS

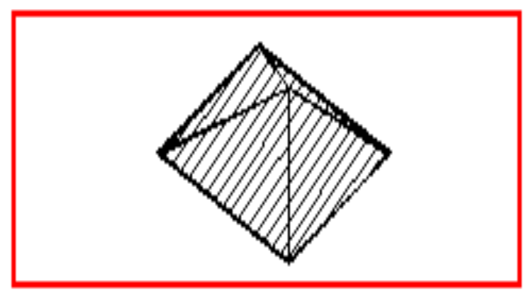
Typical 3rd Generation (undulator) Synchrotron Source

DLS coherence and flux at 8keV will be similar

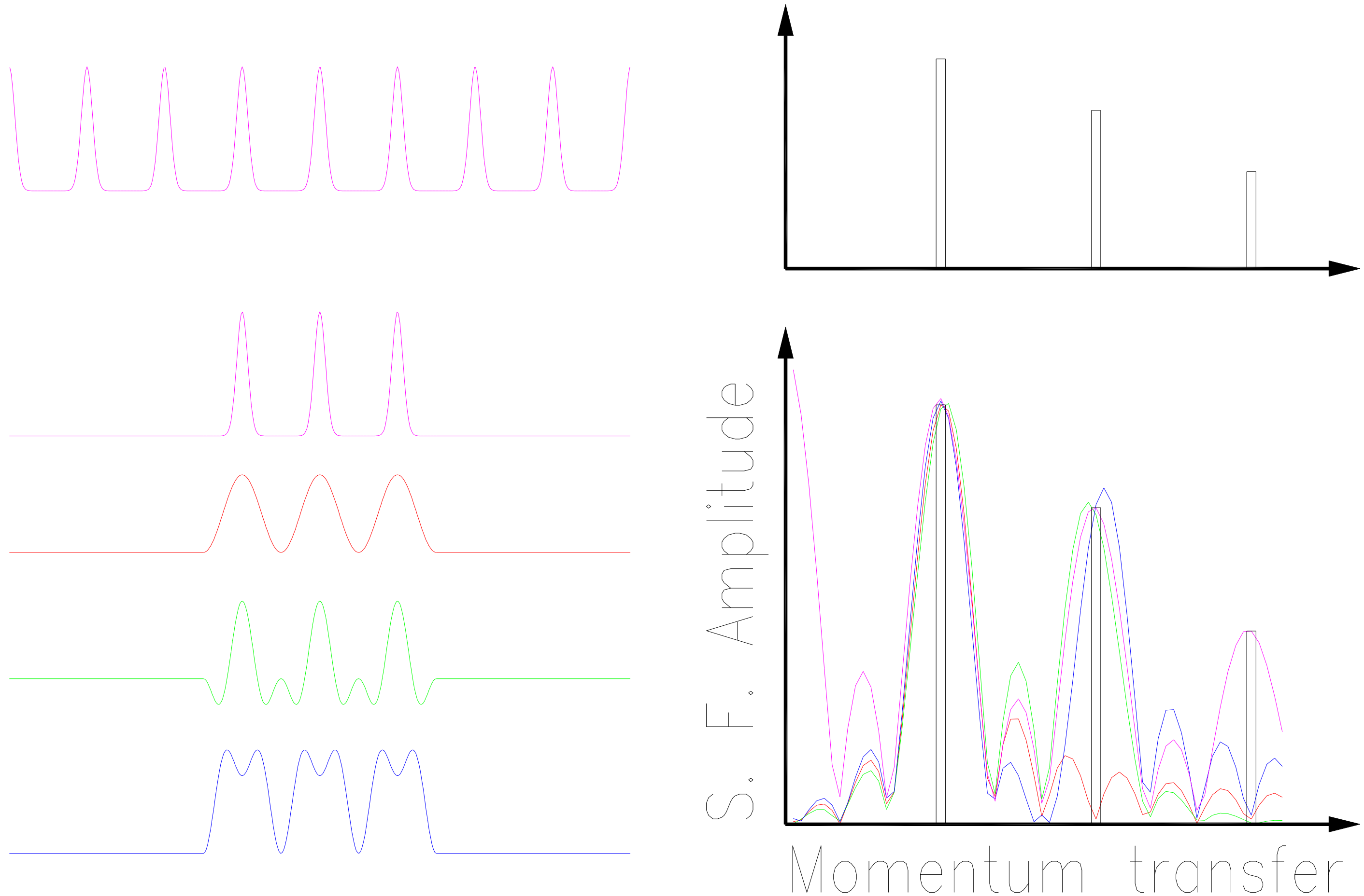
Coherence of	ξ_{VER}	ξ_{HORIZ}	ξ_{LONG}	Flux
Raw Undulator	35μm	9μm	0.004μm	2×10^{12}
Si(111) Monochromator	35μm	9μm	1μm	1×10^{10}
C(111) Monochromator	35μm	9μm	3μm	3×10^{9}

Coherent region defined by slits

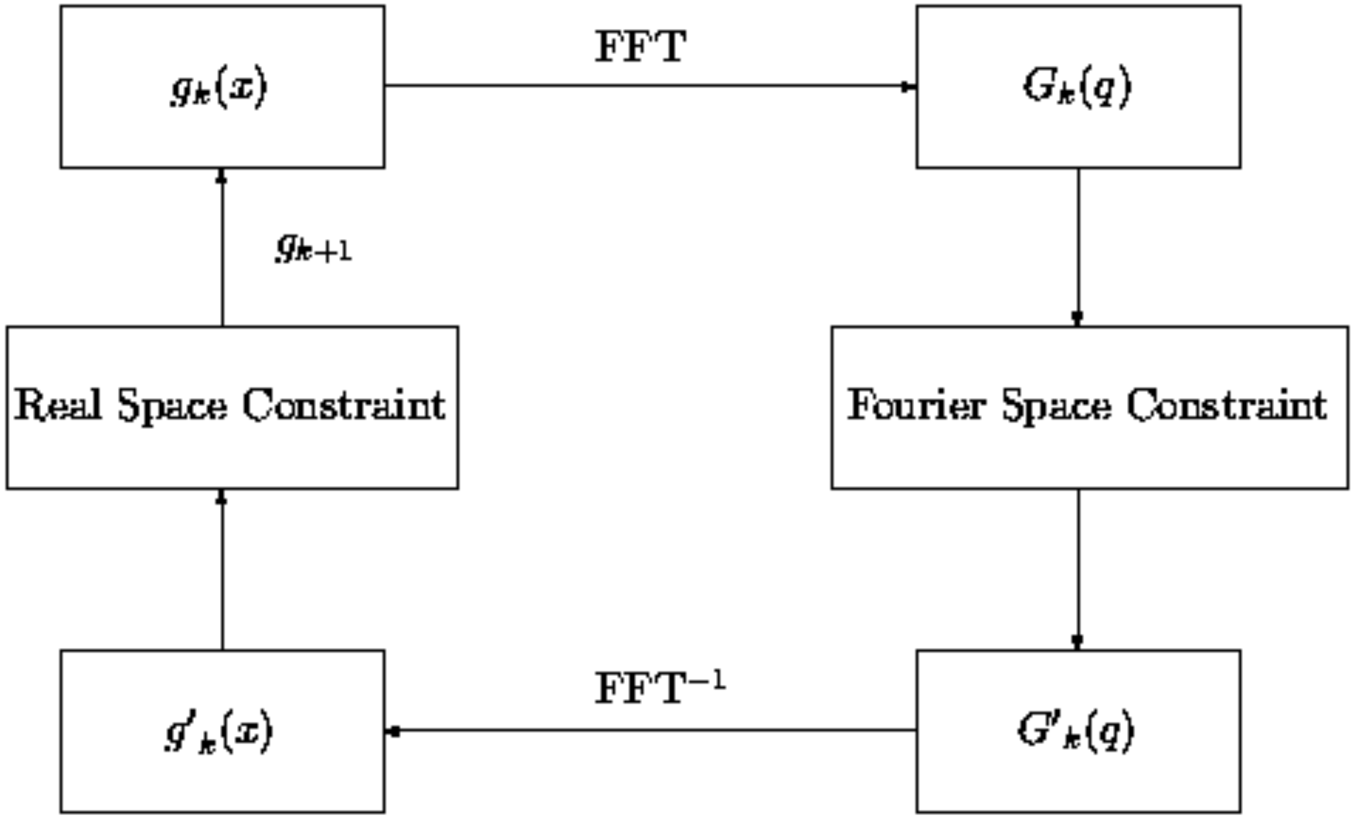




Phase Problem: Finite-size Effect



Generic "Error Reduction" method



J. R. Fienup Appl. Opt. 21 2758 (1982)

R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)

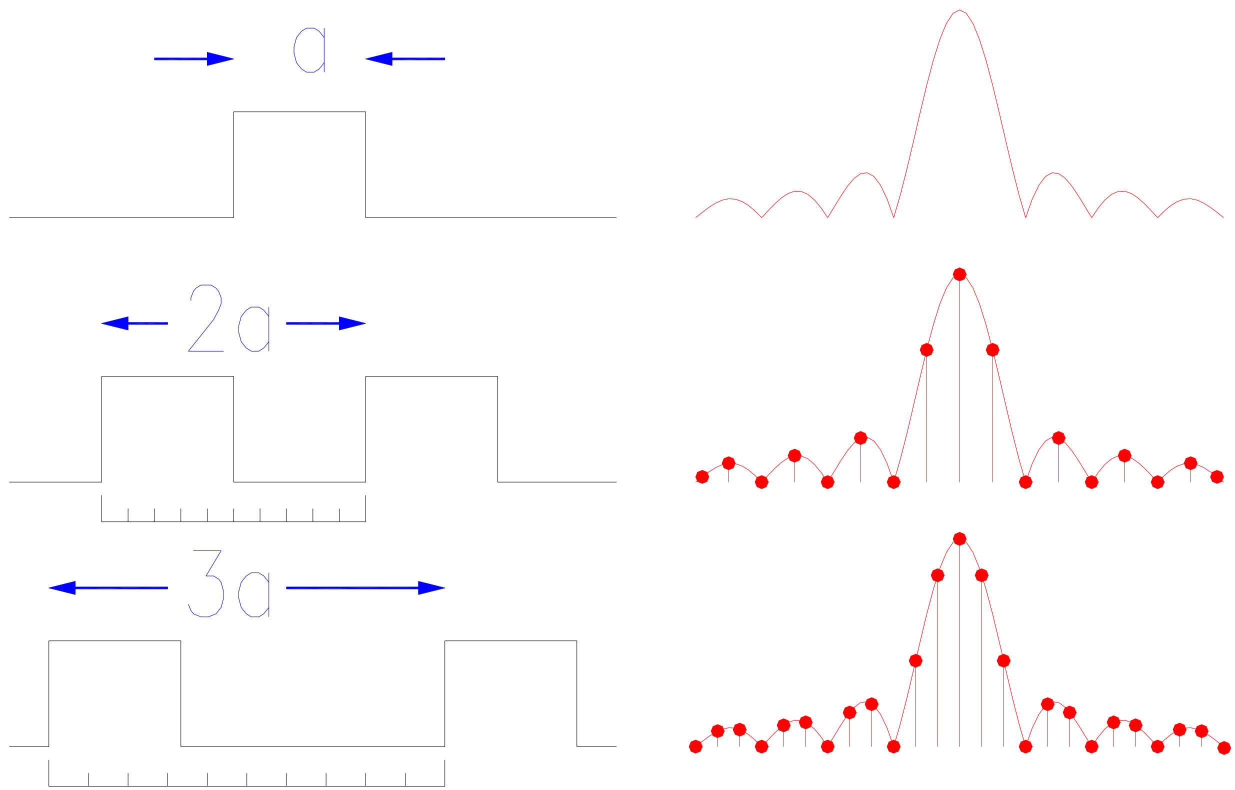
“ER” Methods in Crystallograhy

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

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

Oversampling for Small Crystals

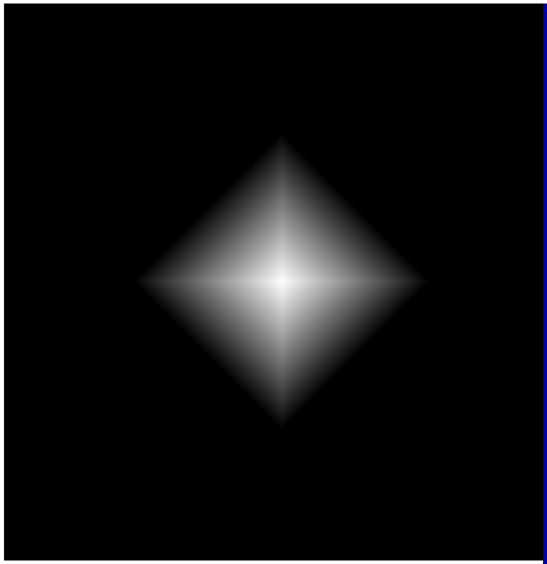
DFT connects N points with N reciprocal points

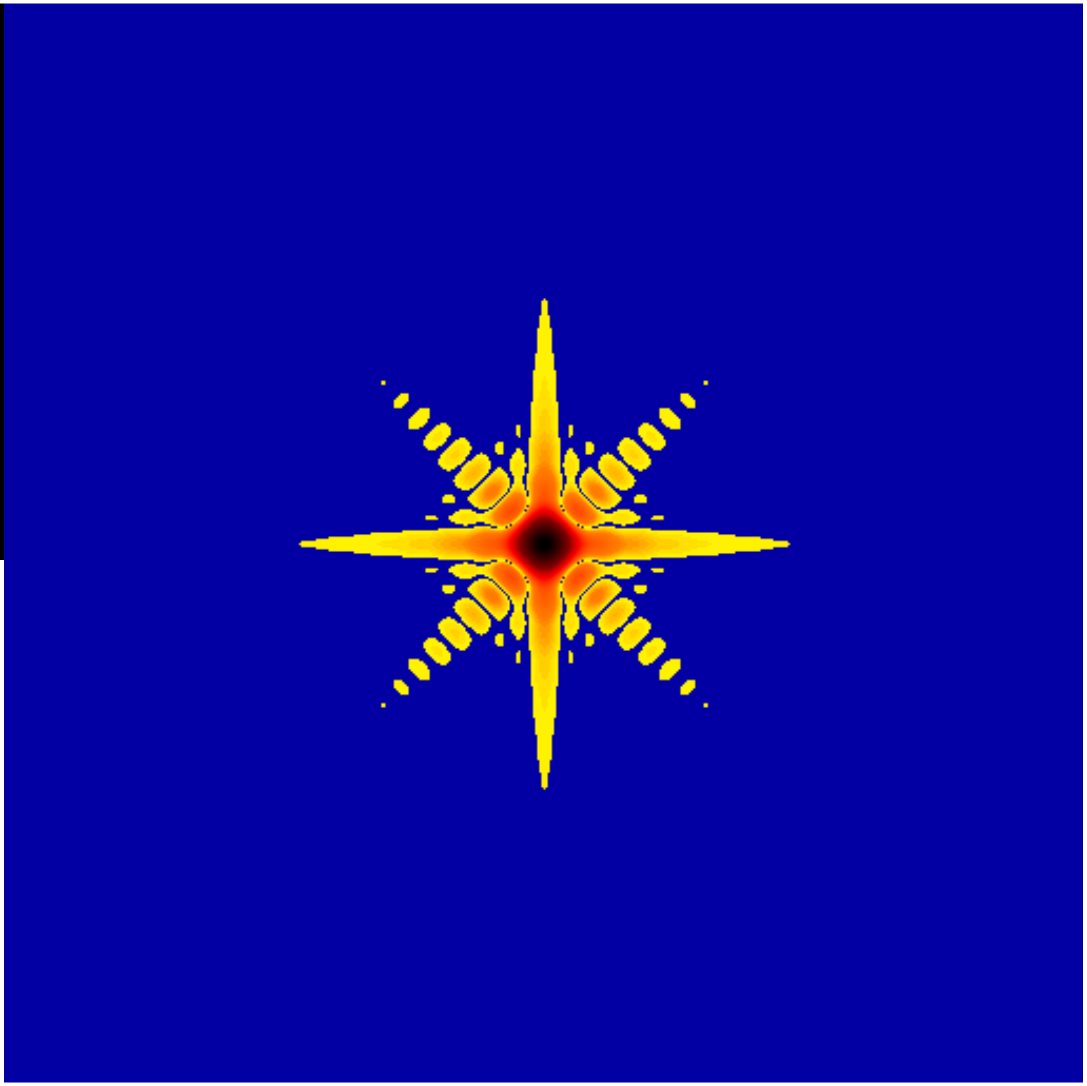


N points in 2a:
N/2 unknowns

N points in 3a:
N/3 unknowns

Always N/2 measurements





Diffraction as a Surface Integral

Die äußere Form der Kristalle in ihrem Einfluß auf die Interferenzerscheinungen an Raumgittern

Von M. v. Laue

Annalen der Physik [5] 26 55 (1936)

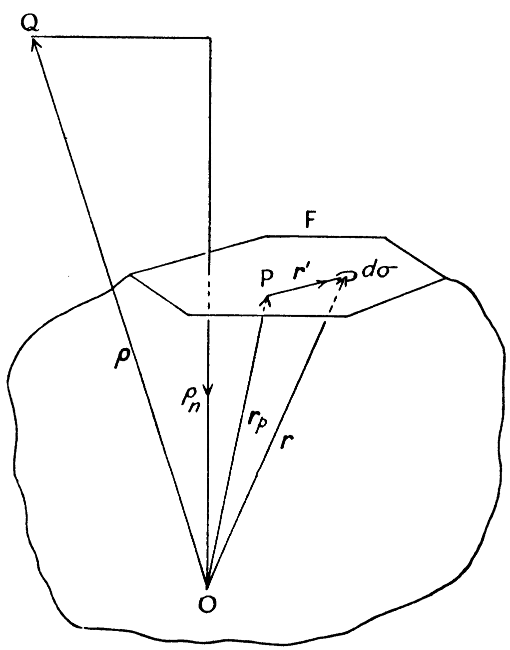
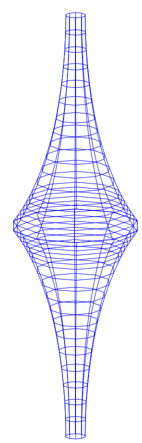
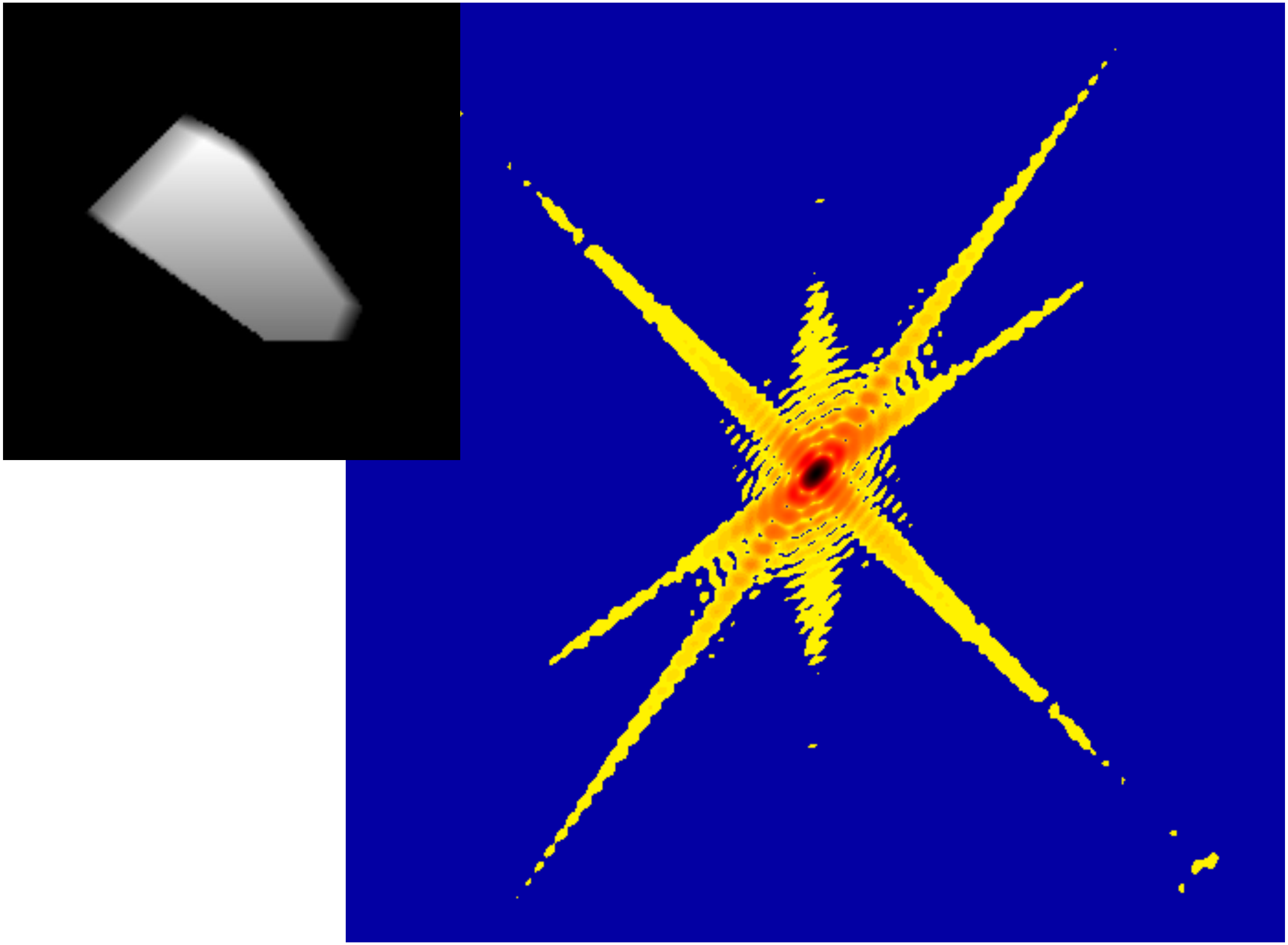


FIG. 200

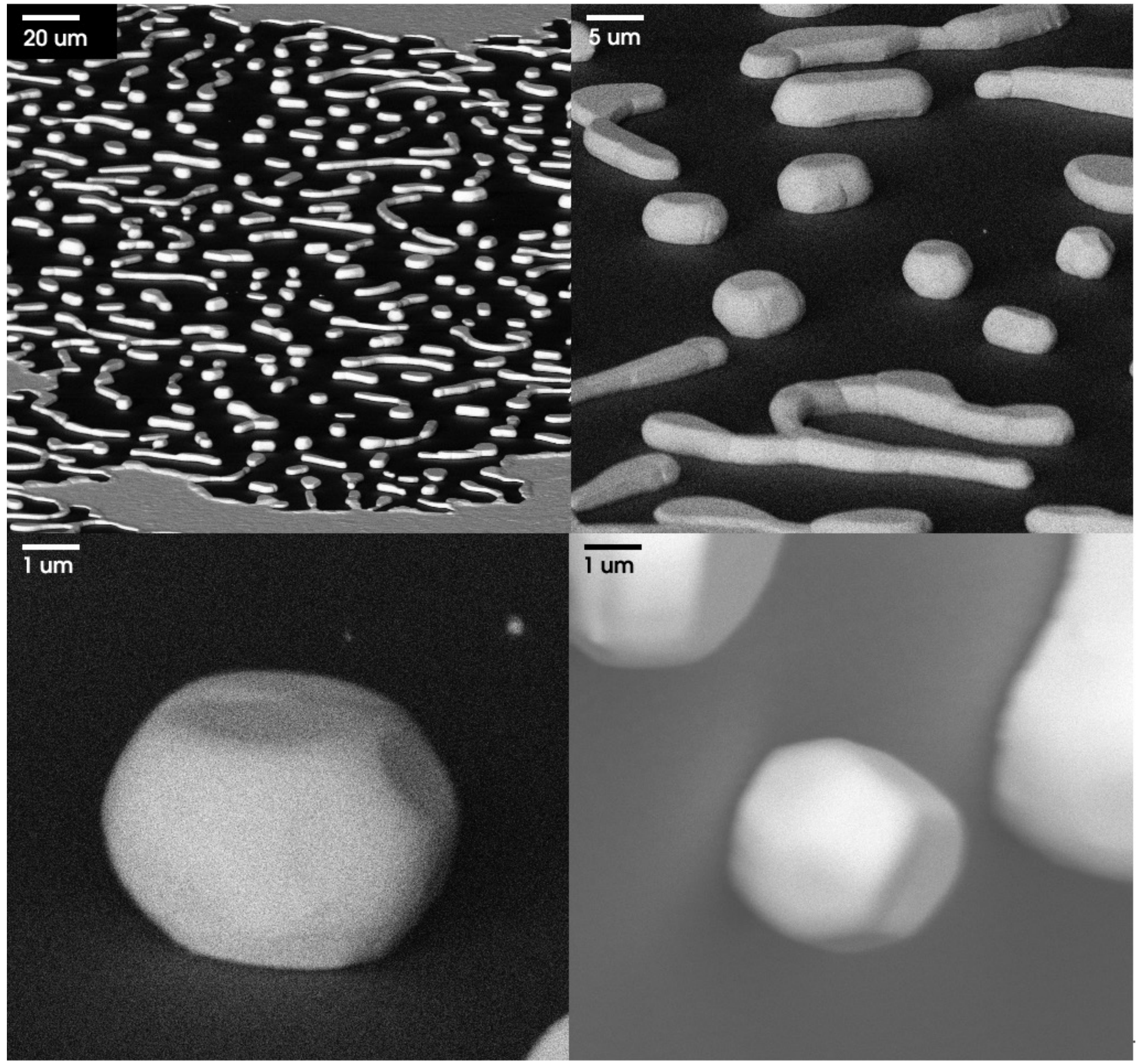


"Stacheln"

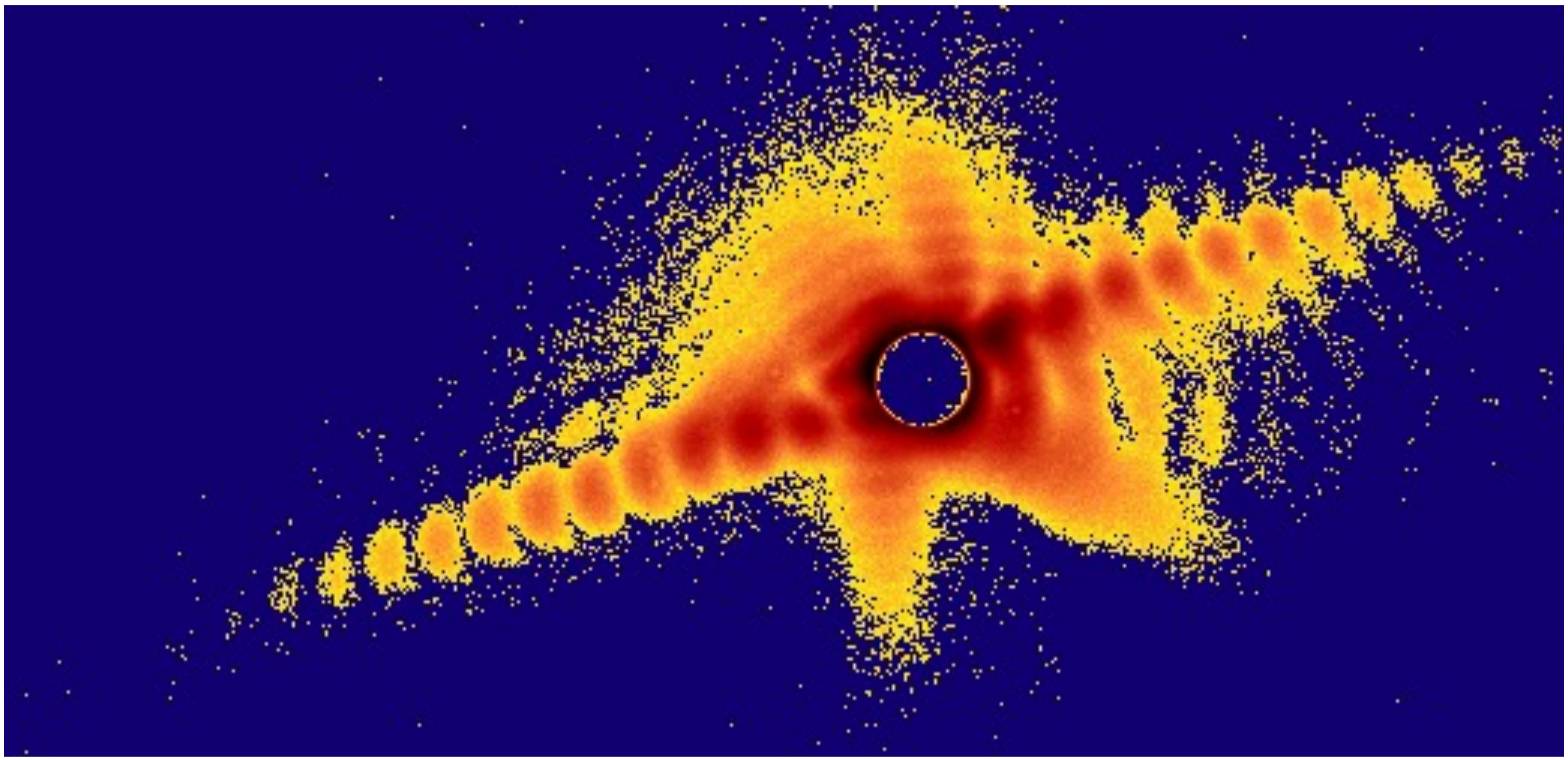


SEMS

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

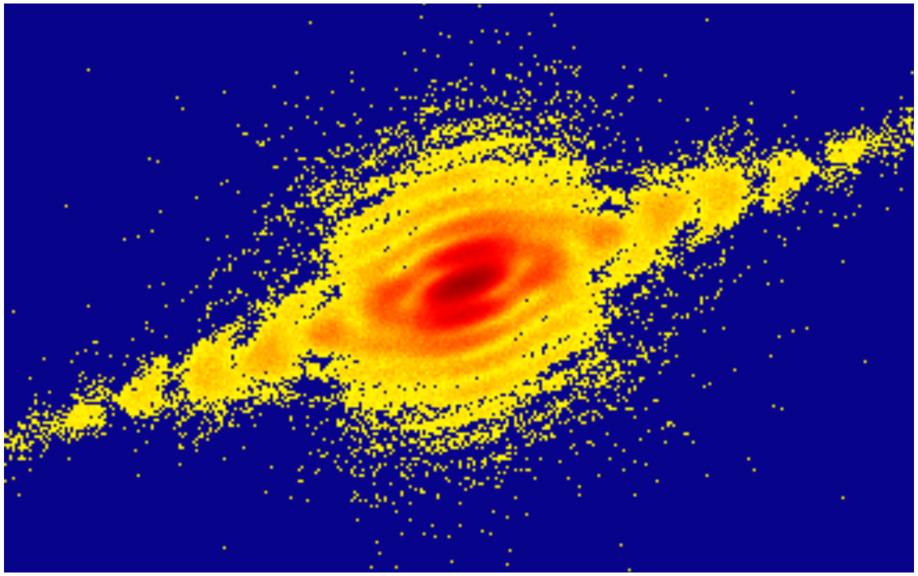


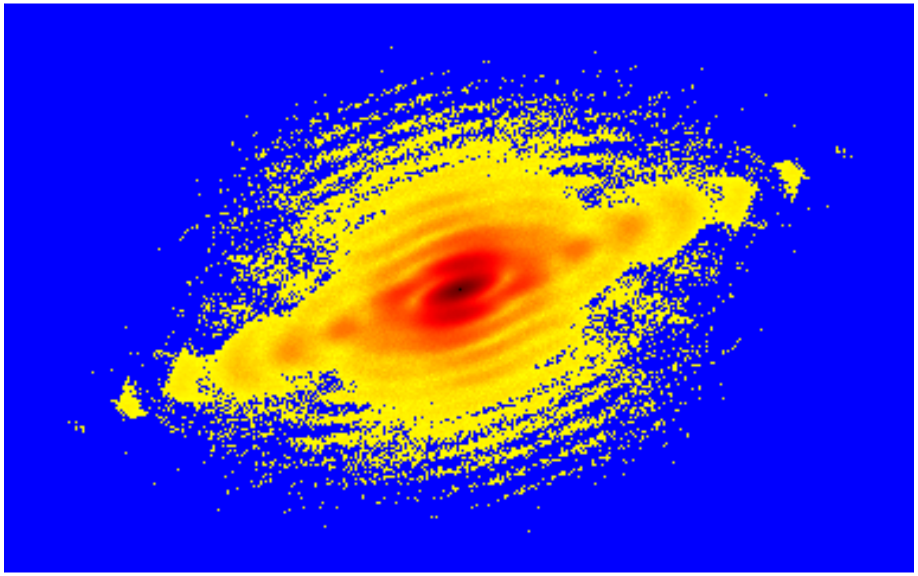
Micron-sized gold crystal: (111) Bragg reflection

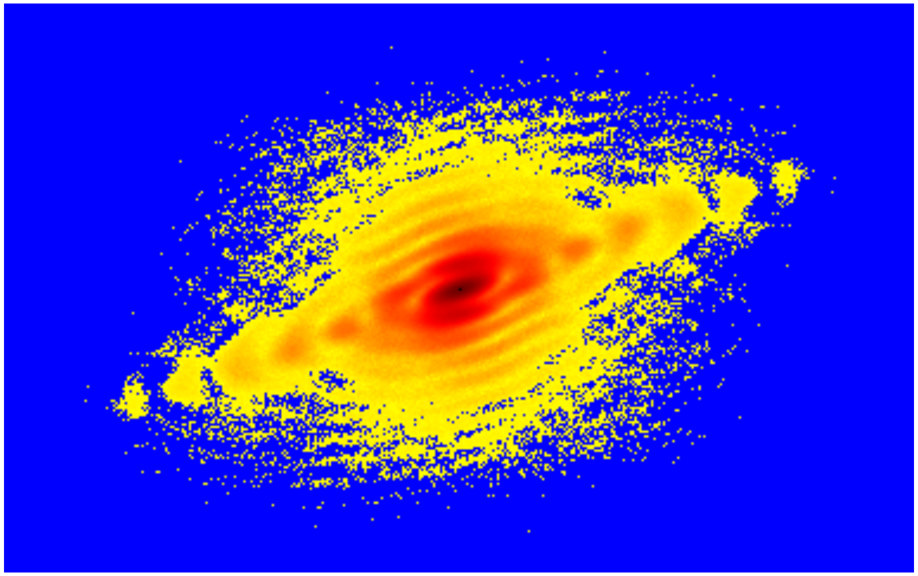


Symmetrized Data and two best fits

Chisq=0.0005

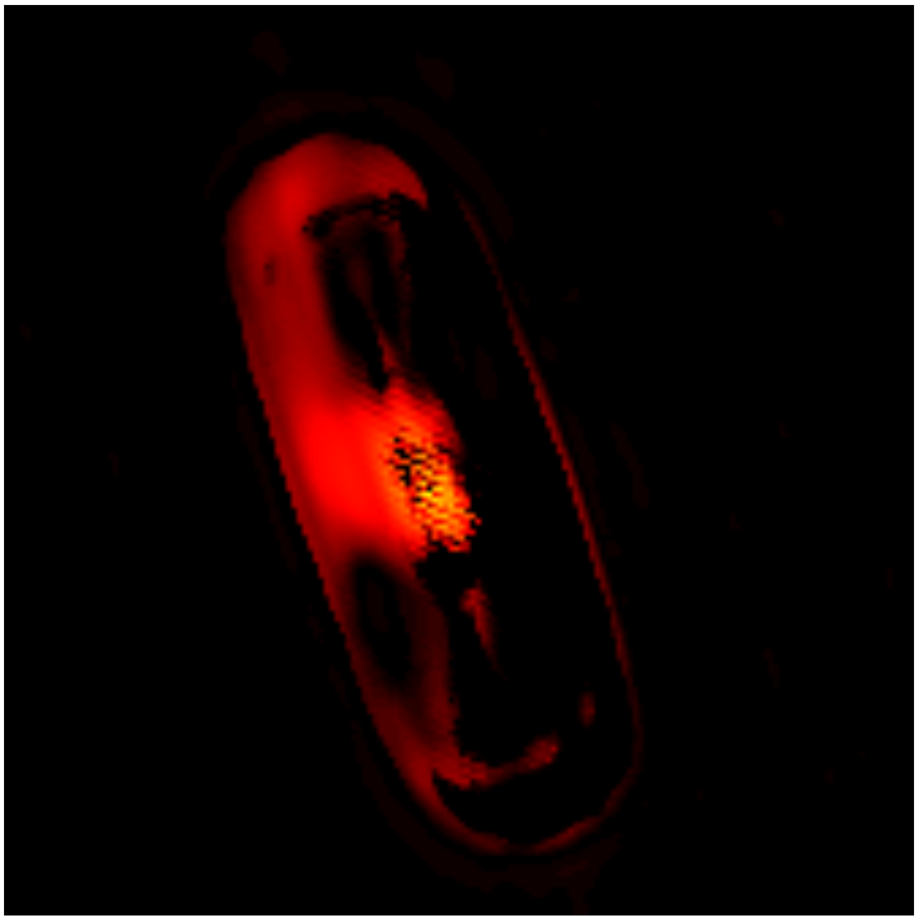


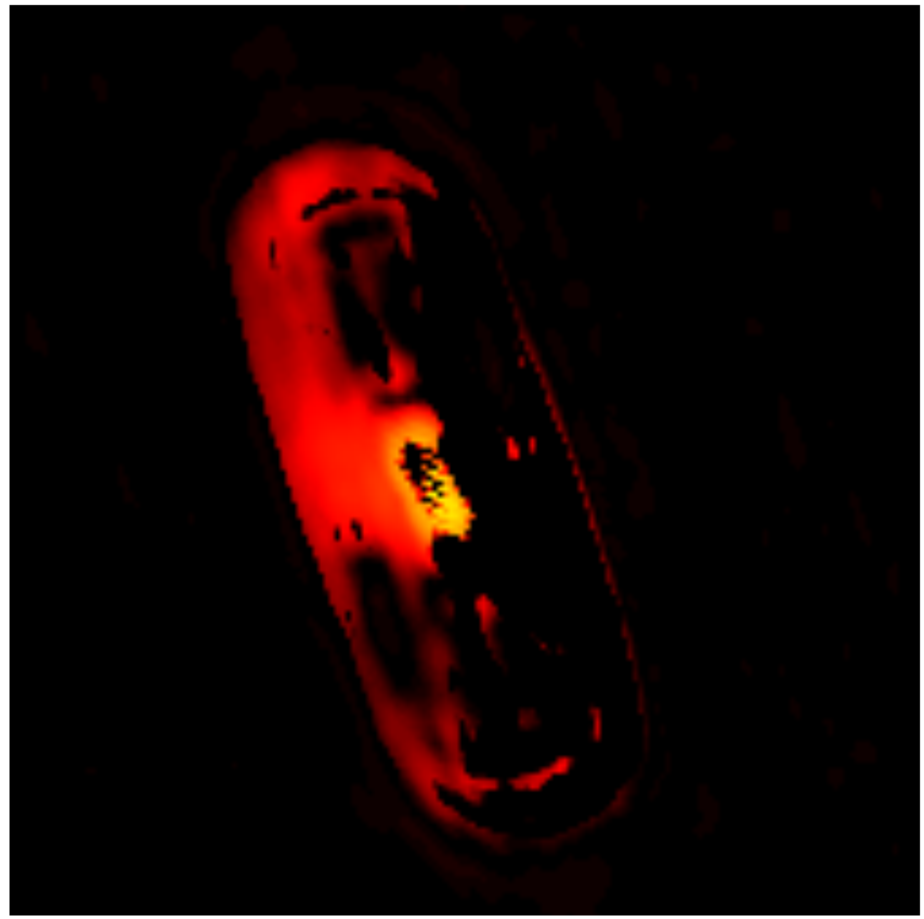




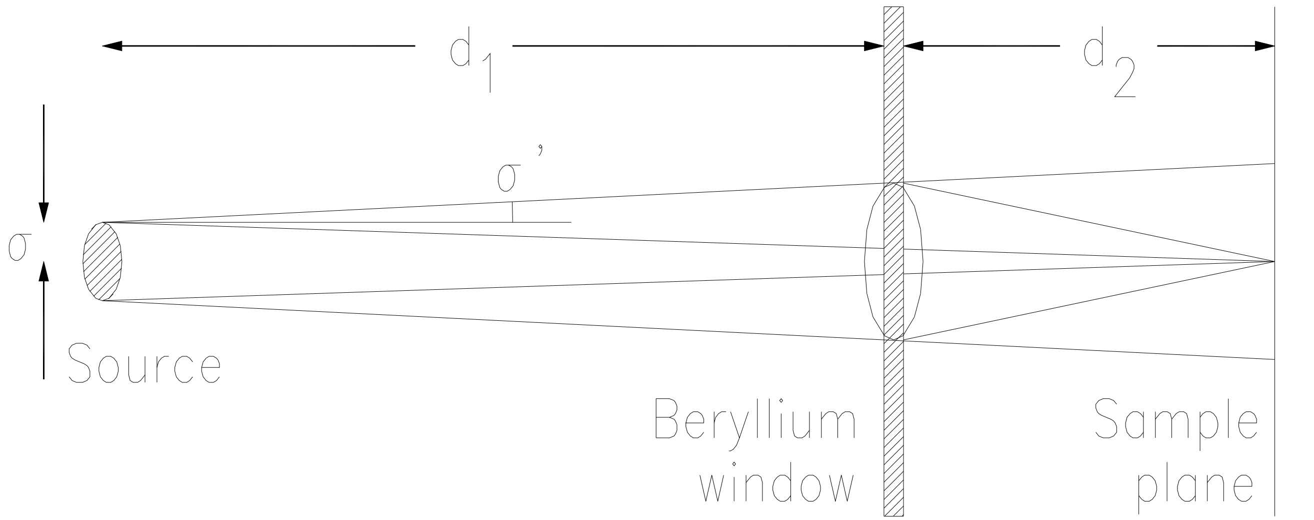
2D Reconstructions

chisquare = 0.0005





Window as a Secondary Source

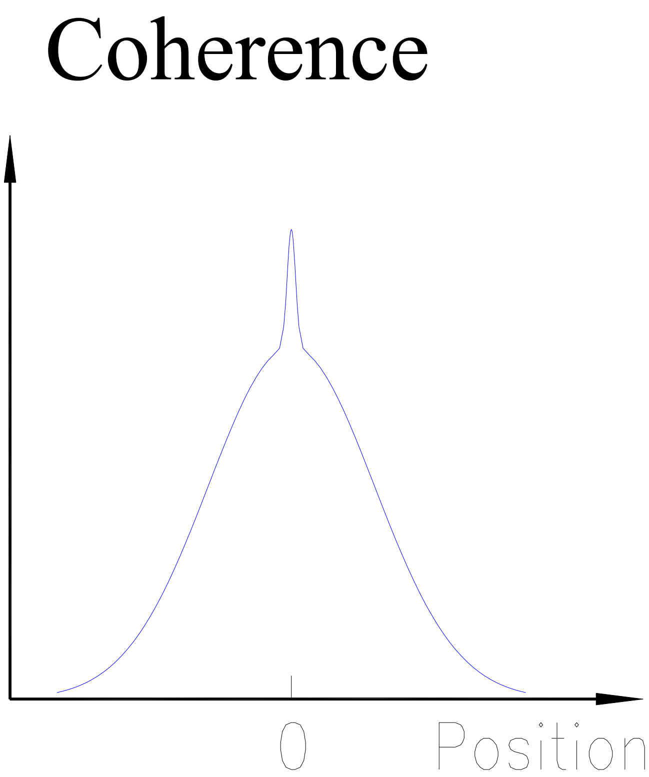


$$L_T = \frac{\lambda D}{2\sigma}$$

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

Vert ~50
Horiz ~3

Two Components to MCF



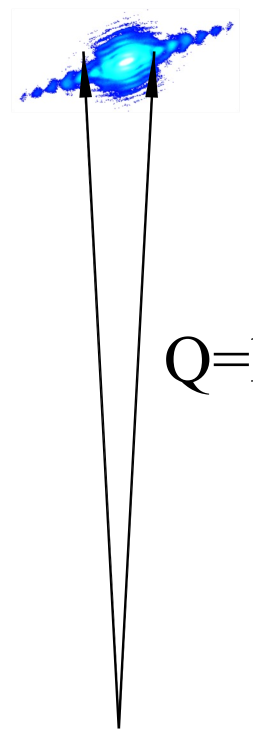
•Width of narrow component is very sensitive to location of window

•Amplitude of narrow component depends on level of diffuse scattering from window

•Amplitude increases with distance (for phase object)

3D Diffraction Method

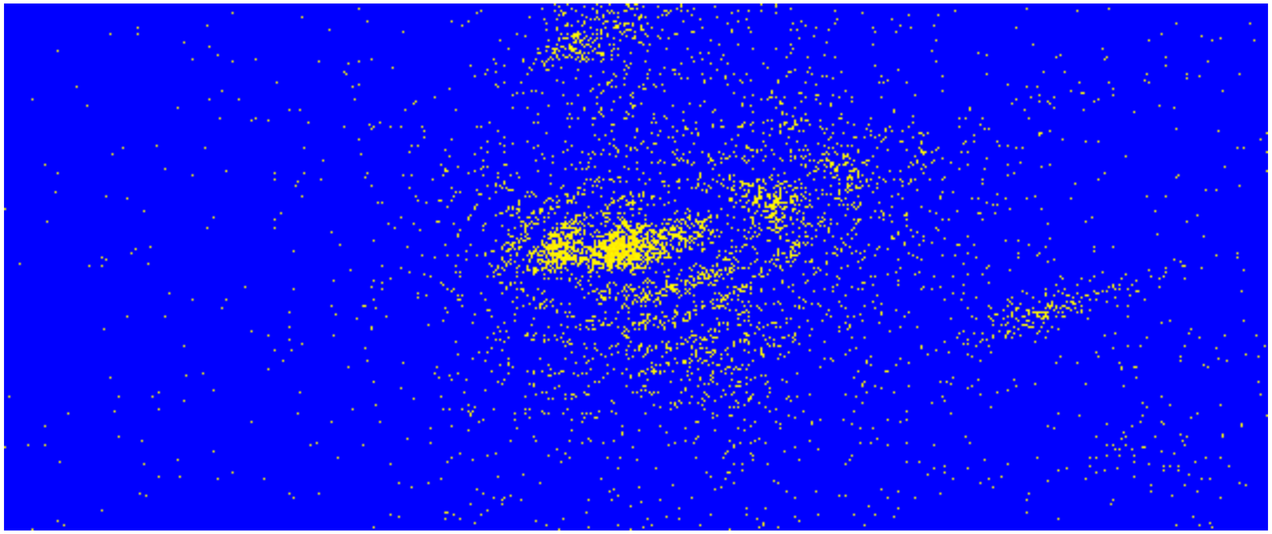
k_f



$Q = k_f - k_i$

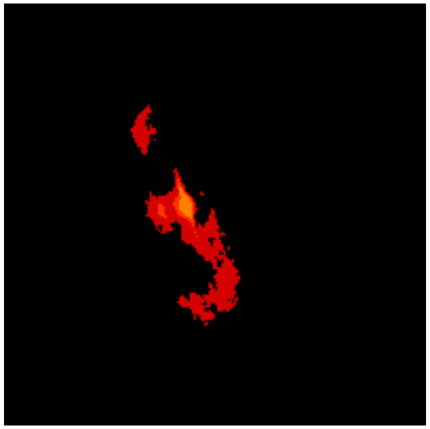
k_i

3D Diffraction Data
1 micron Au crystal

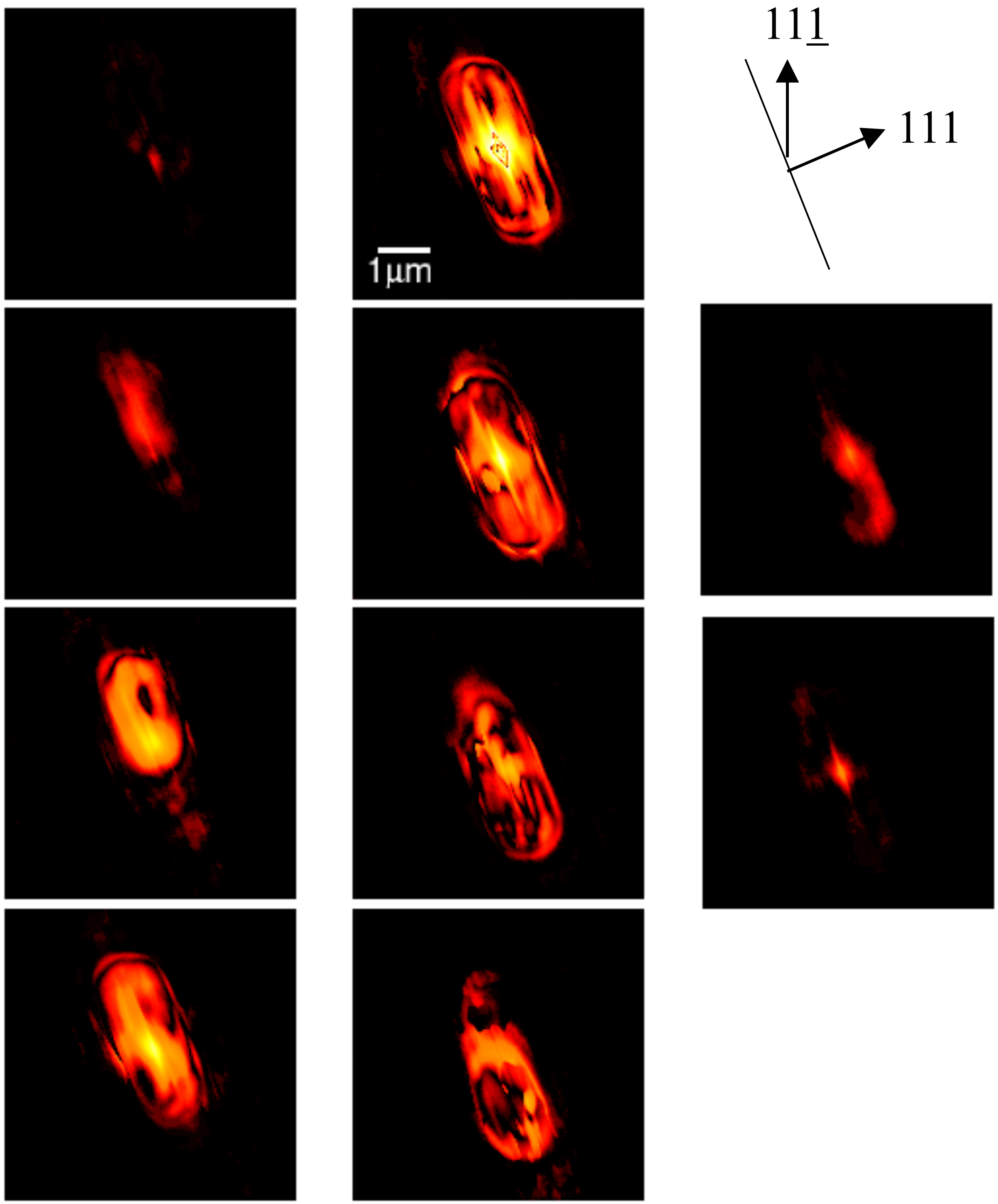


* Center is Symmetric *

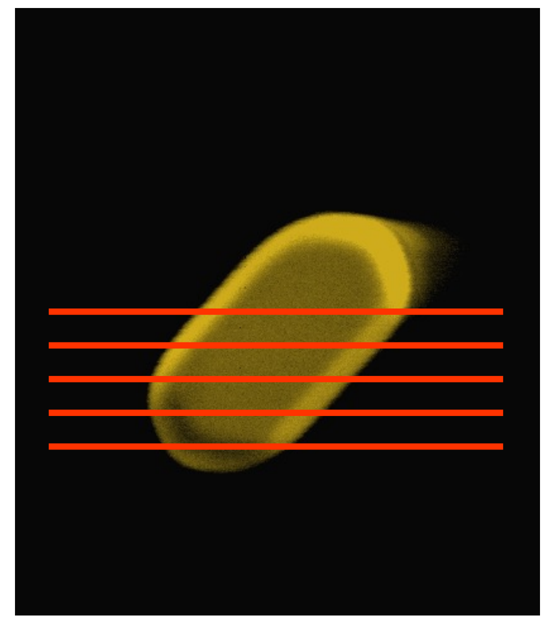
30 frames of 3D reconstruction 1 micron gold crystal



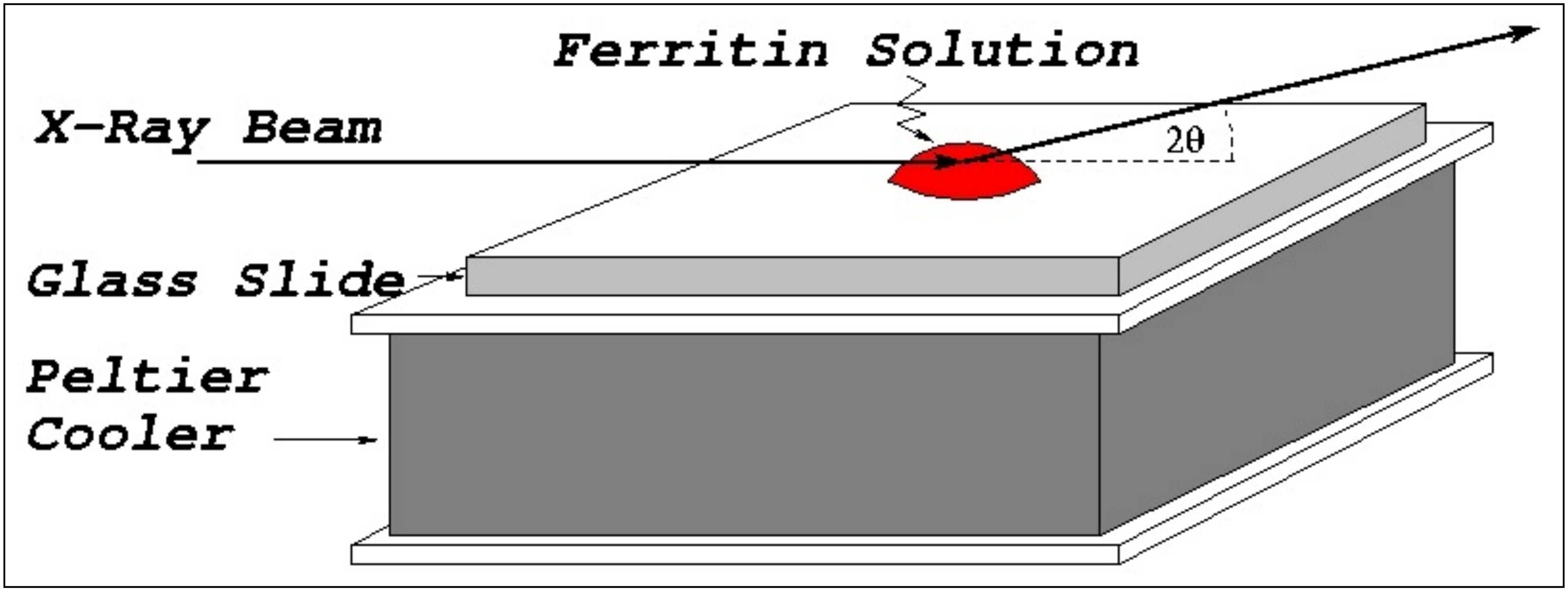
* CENTER *



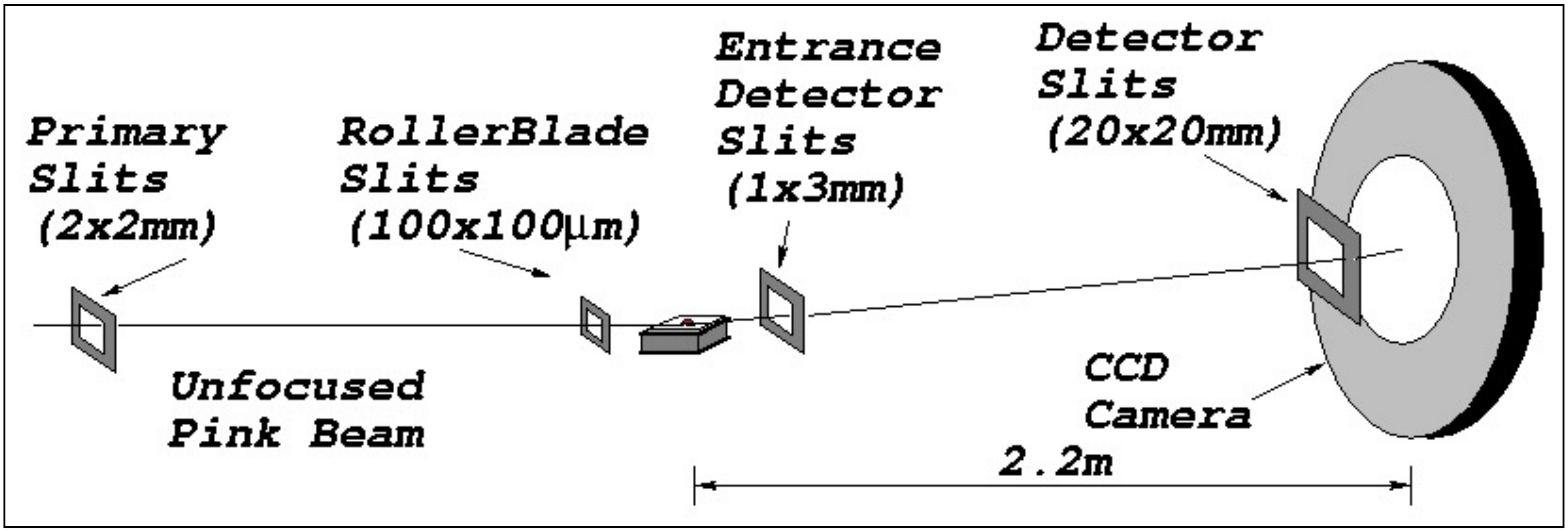
Slices through plan view SEM:



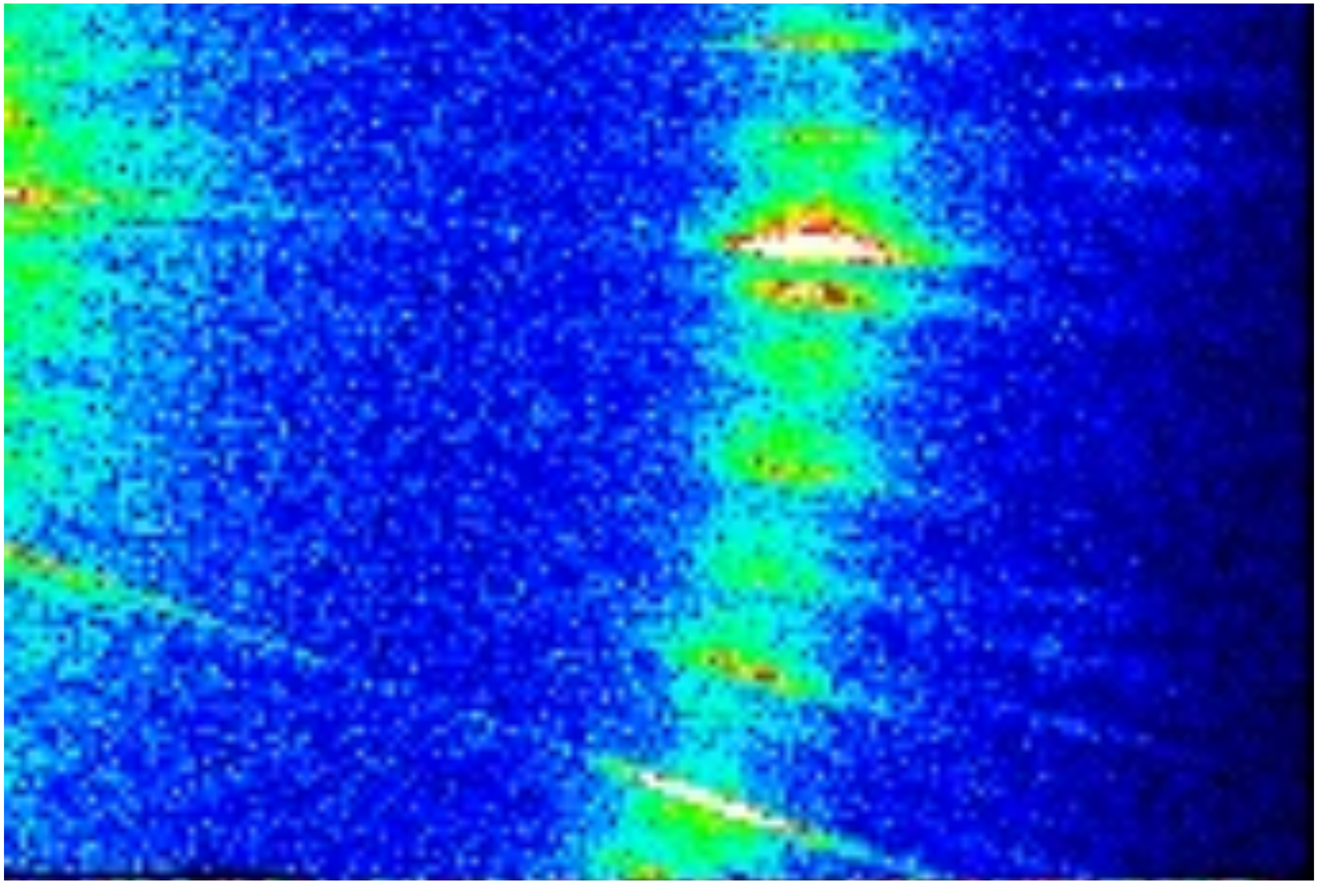
In-situ Study of Crystallization



Experiment at APS Sector 34

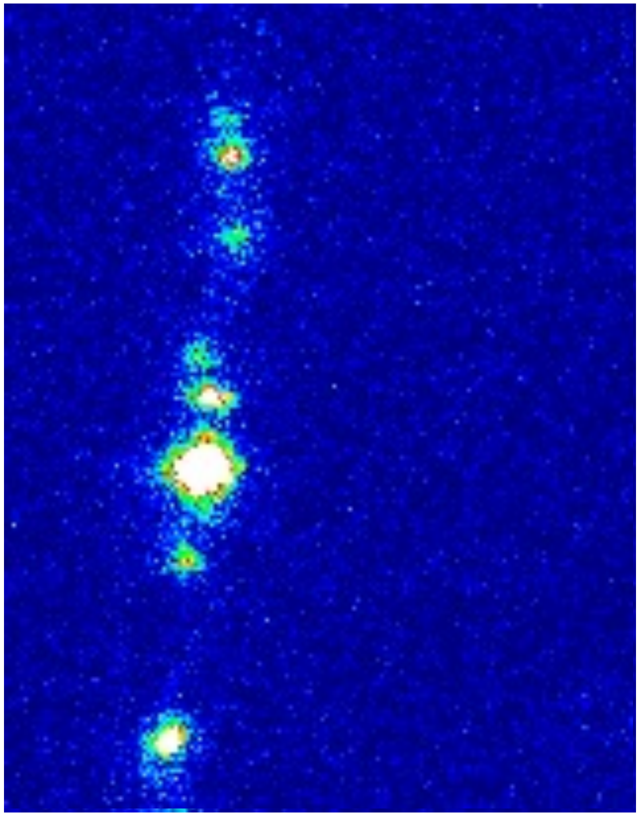


“Pink beam” sees CTRs

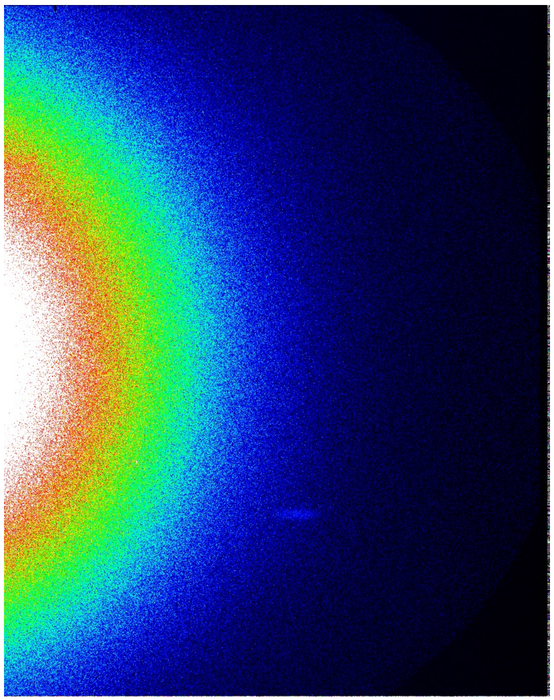


Ferritin (111) Powder Ring

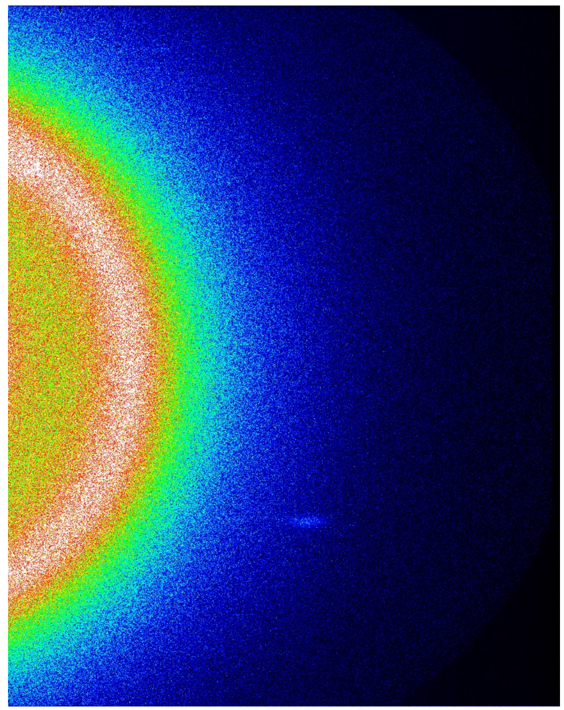
- 50 frames
- 30sec exposure
- 0.3sec playback
- 150x200 pixels of 22.5μm



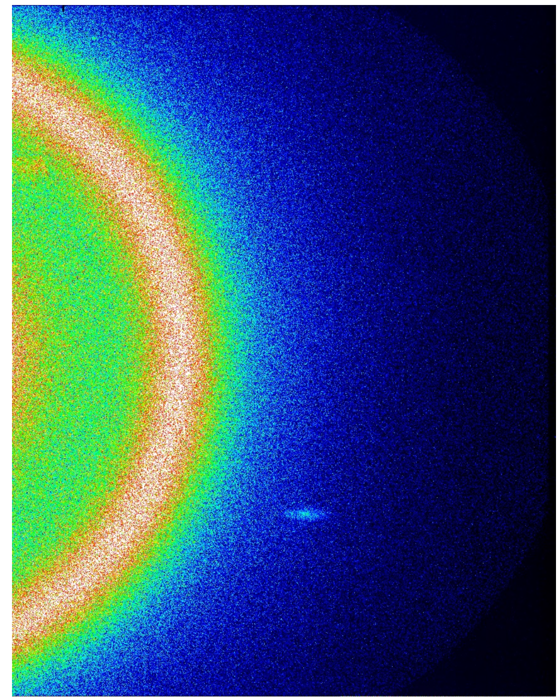
Ferritin Solution upon Freezing



T=13.3

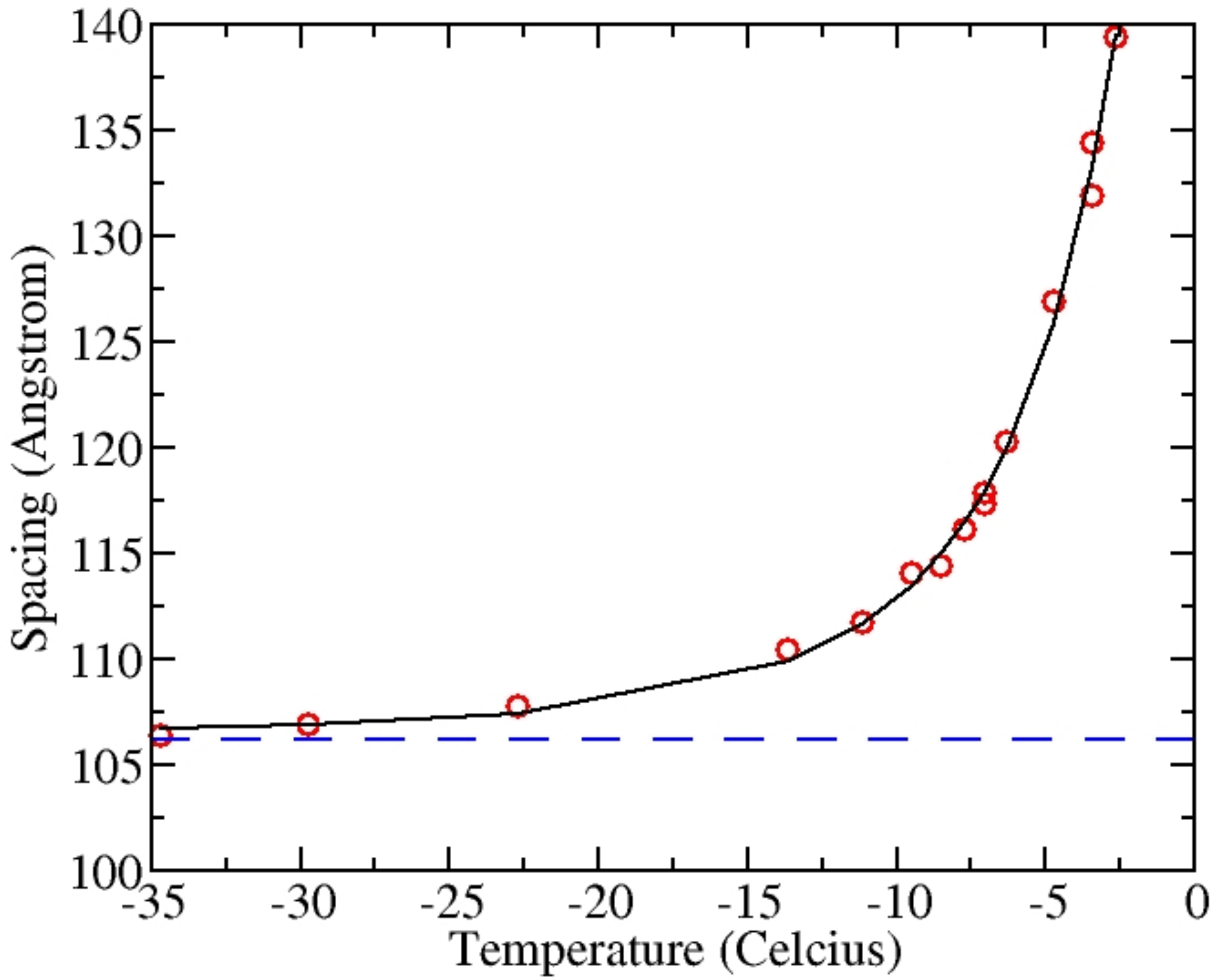


T=-4.7

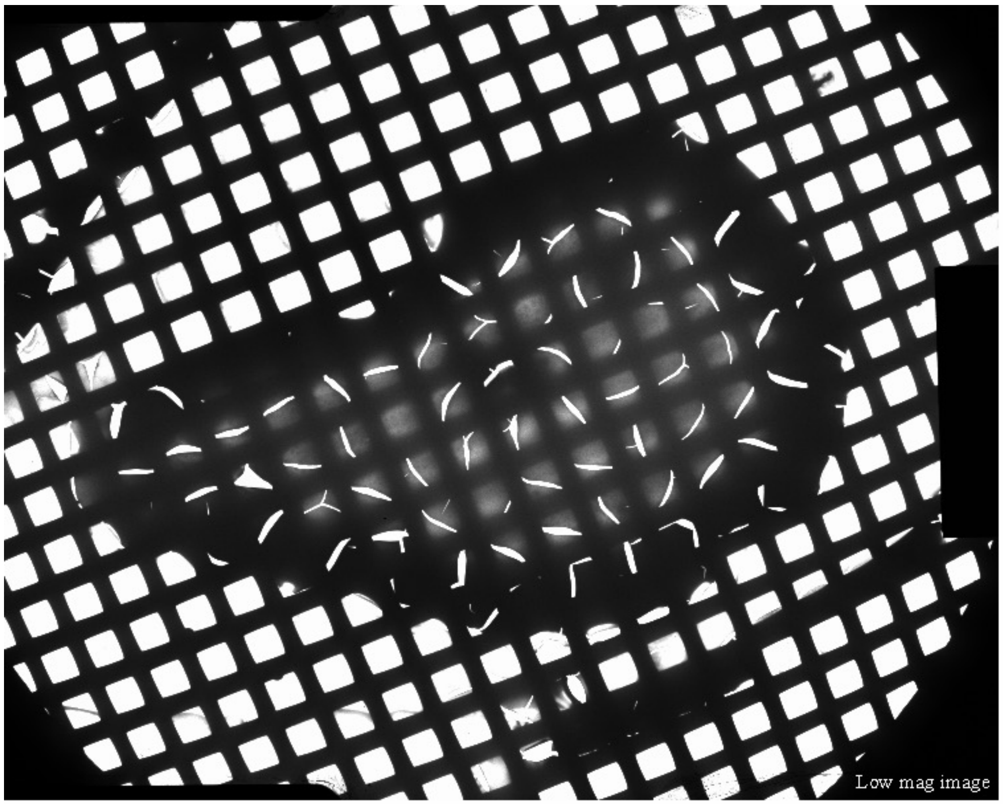


T=-34.7

Temperature Variation of Peak Position

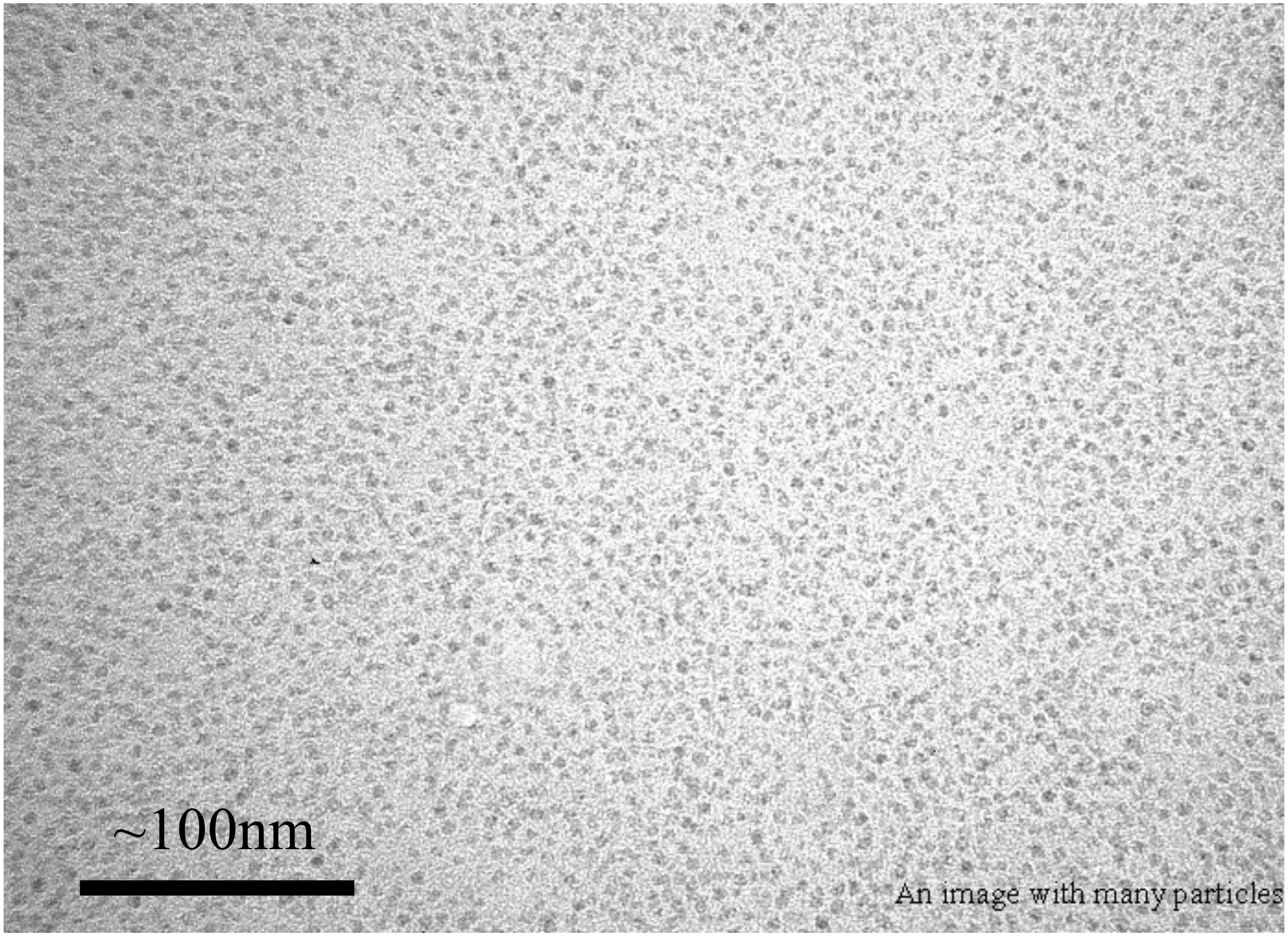


Overview of EM grid

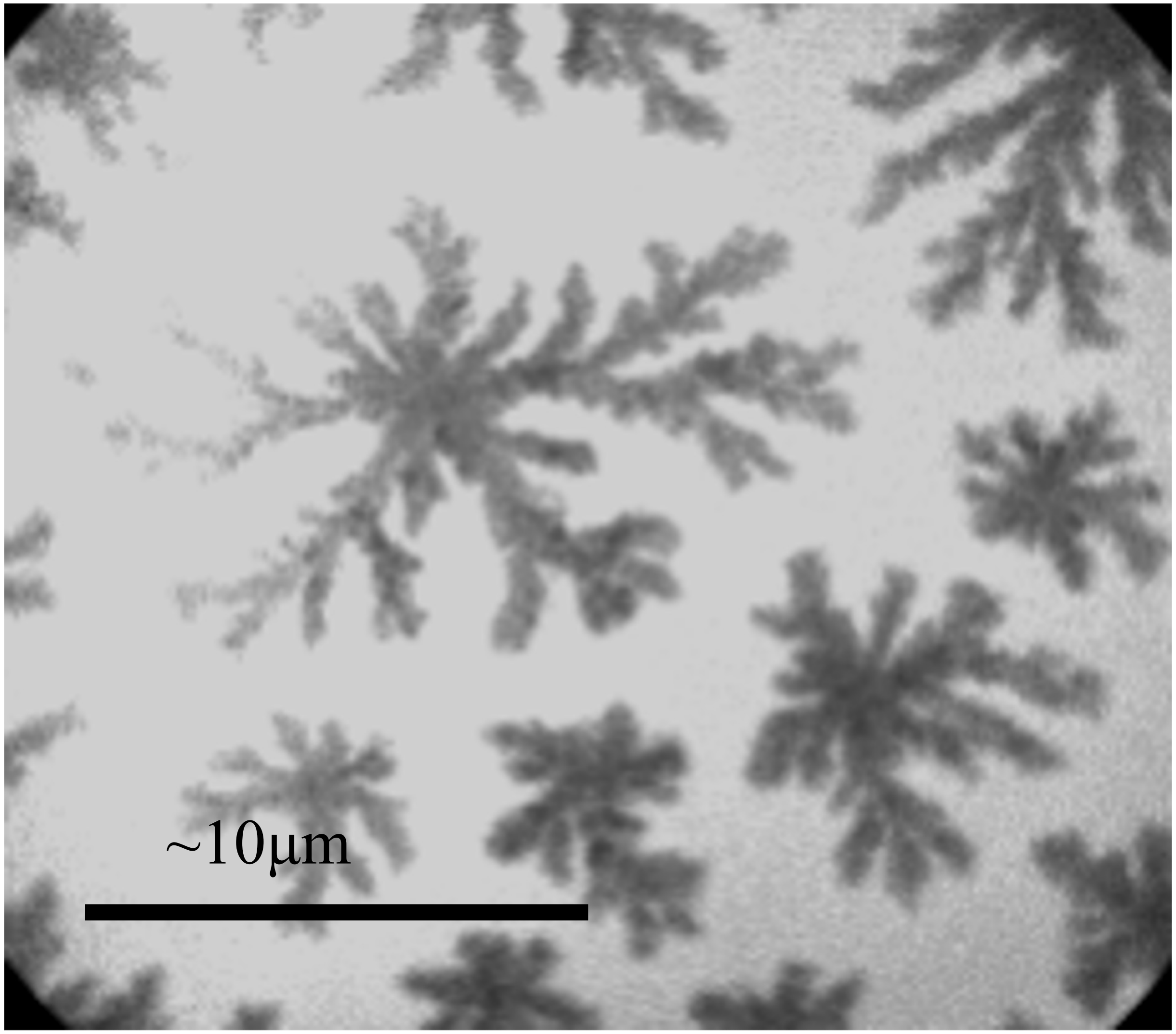


- 3mm diam grid
- a-carbon + “Formvar”
- 10mg/ml Ferritin
- 10mg/ml NaCl
- dipped grid
- dried in air
- no cryo-cooling
  - ▹three regions of different density

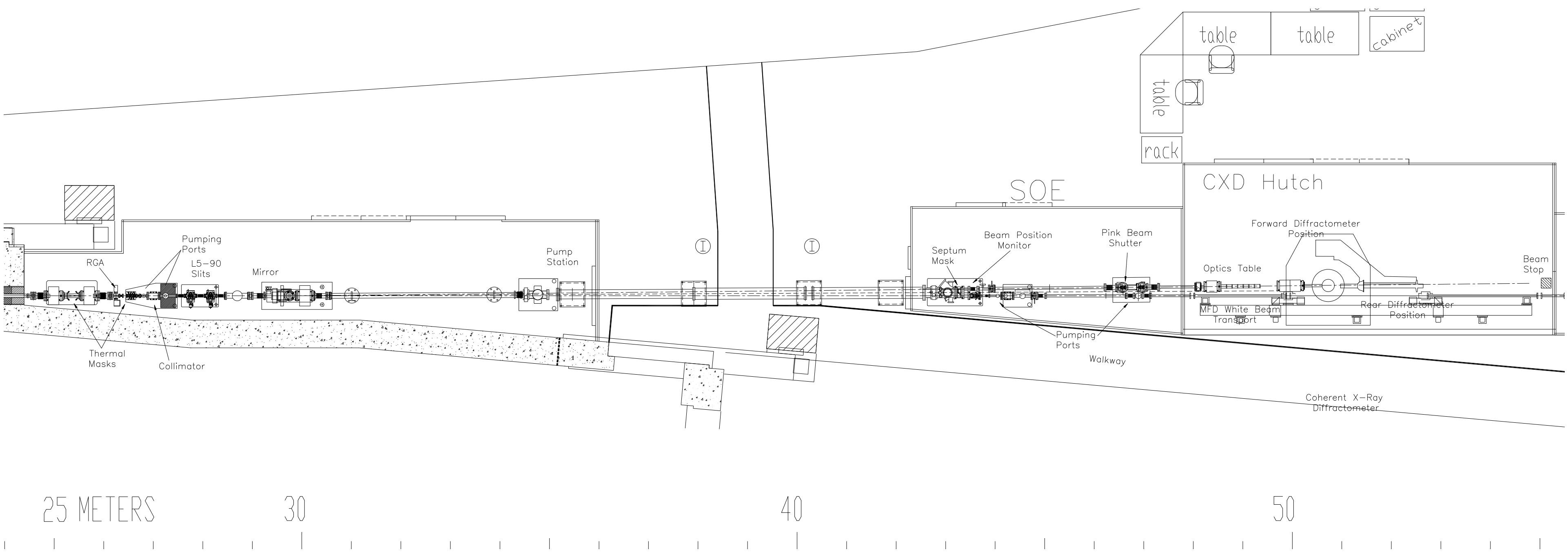
Border Region (low density)



High Density Ring

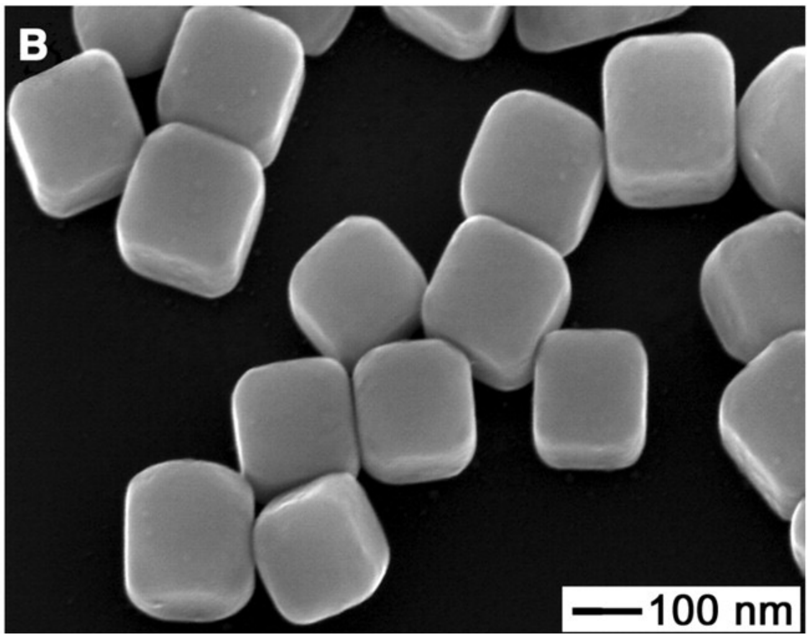


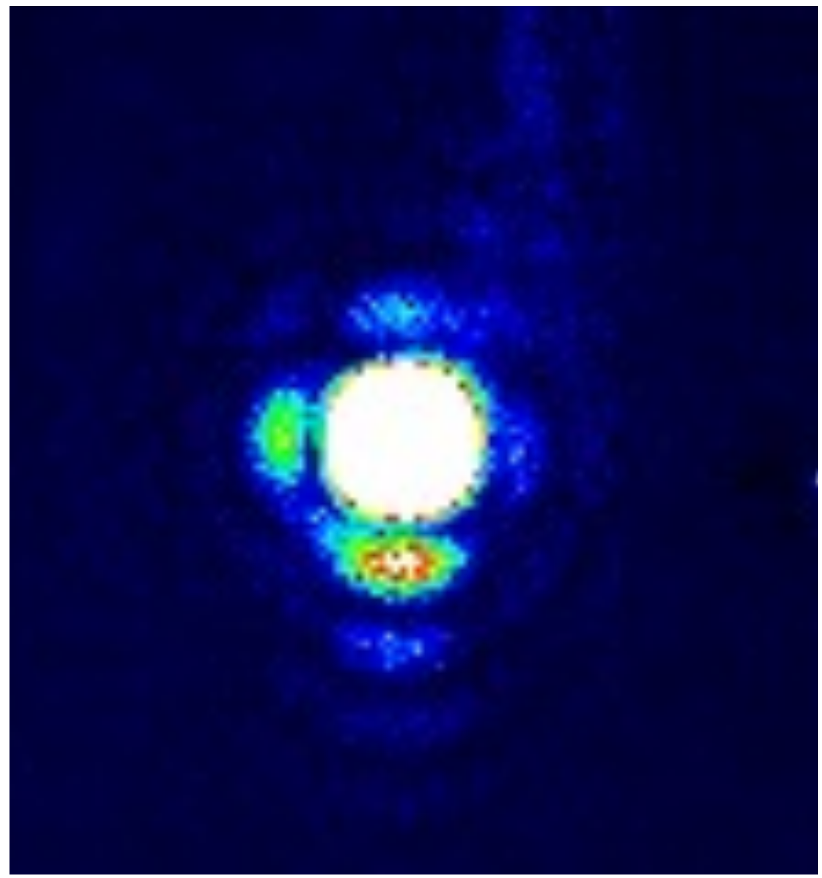
CXD Beamline at APS Sector 34




MICRO-CONTROLE
Newport
MICRO-CONTROLE
Newport

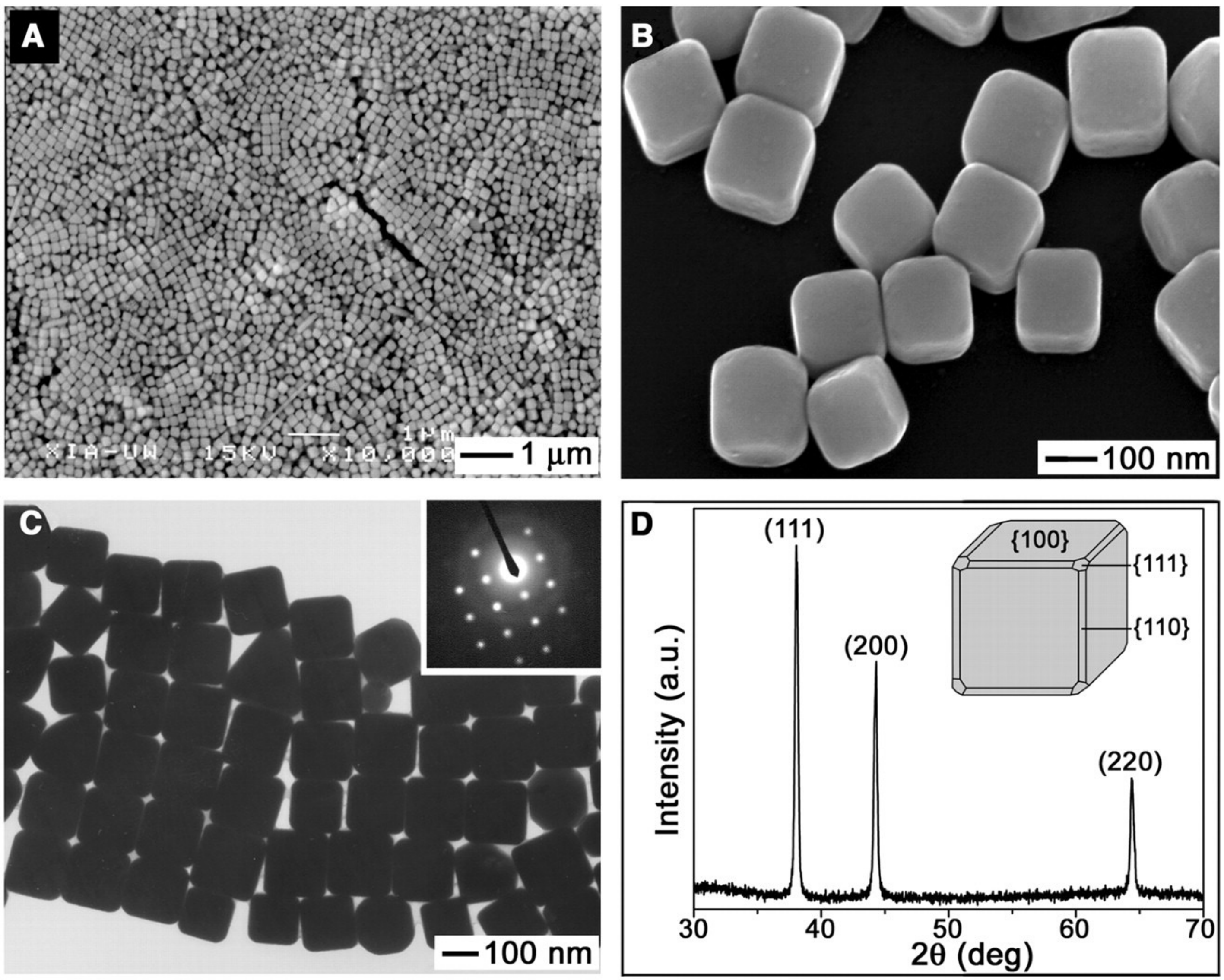
CXD from Silver Nanocubes



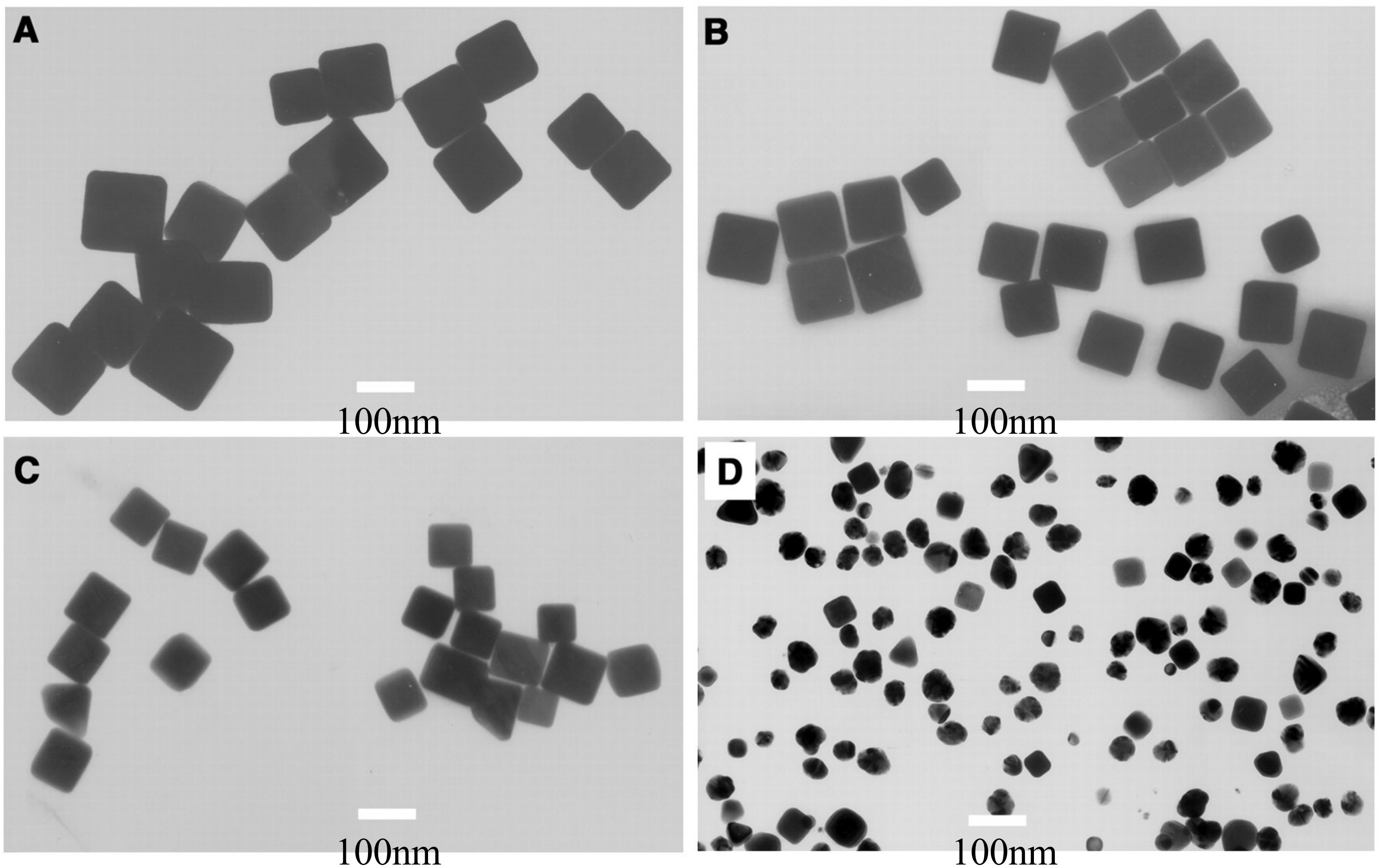


Yugang Sun and Younan Xia,
Science 298 2177 (2003)

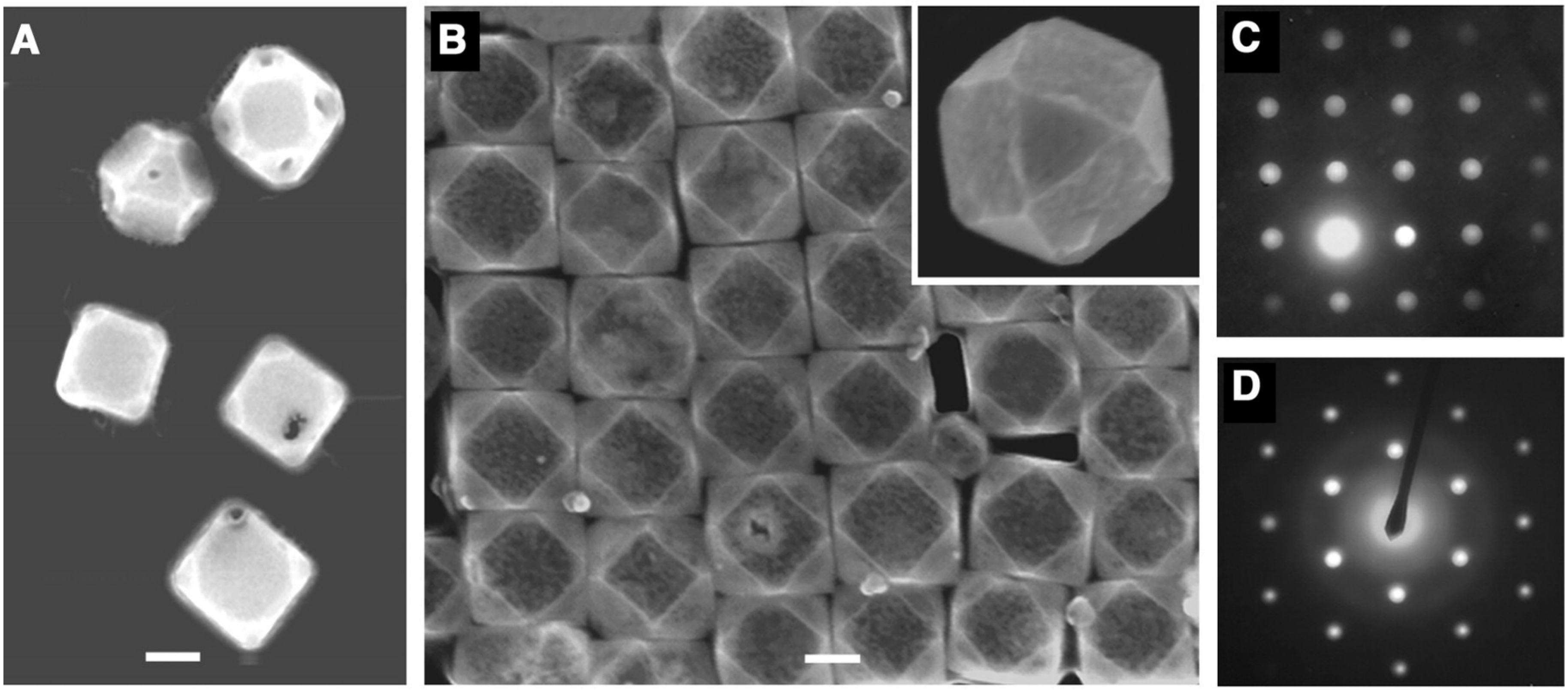
Chemical Synthesis from $AgNO_3$



Variation of growth time and concentration



Au overgrowth using $HAuCl_4$



100nm 100nm

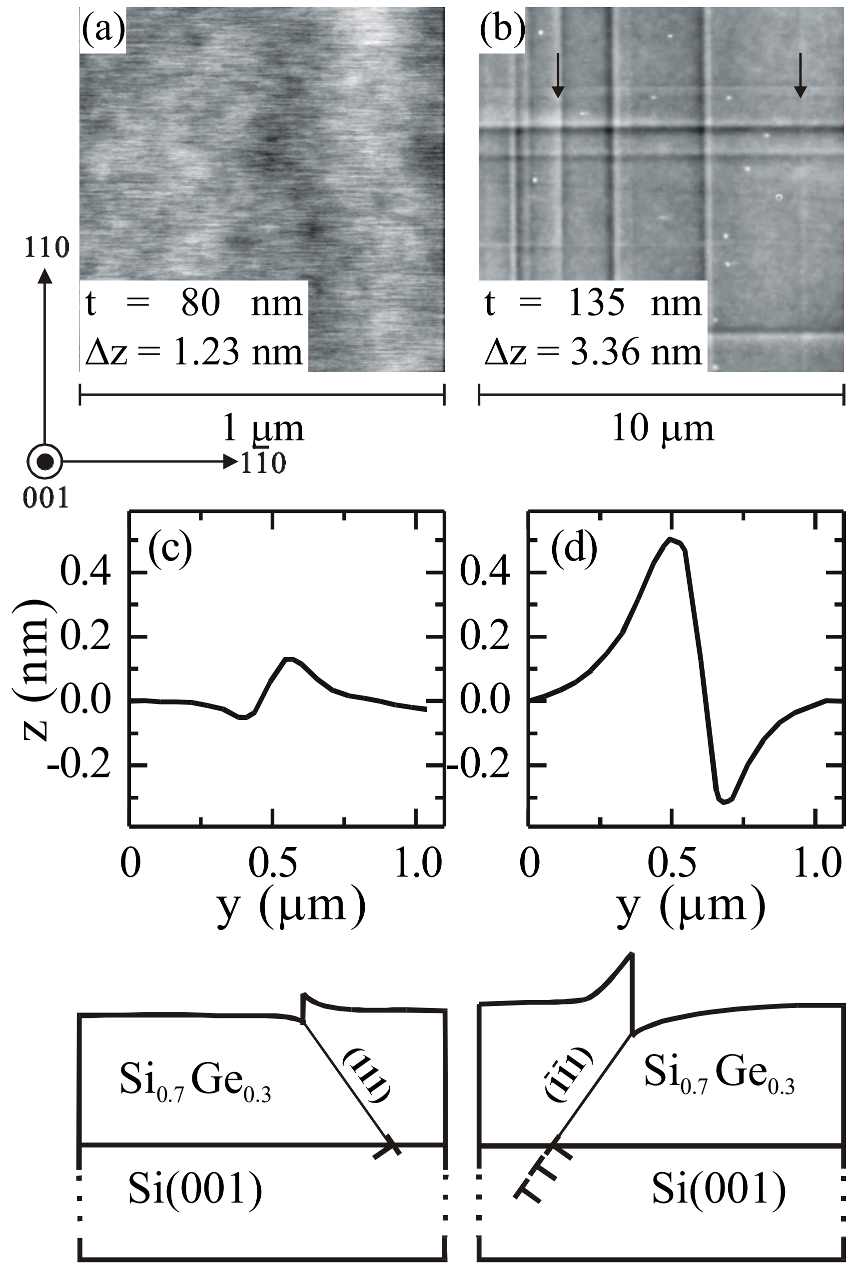
Yugang Sun and Younan Xia, Science 298 2177 (2003)

Low dislocation density GeSi films

Thickness close to critical thickness

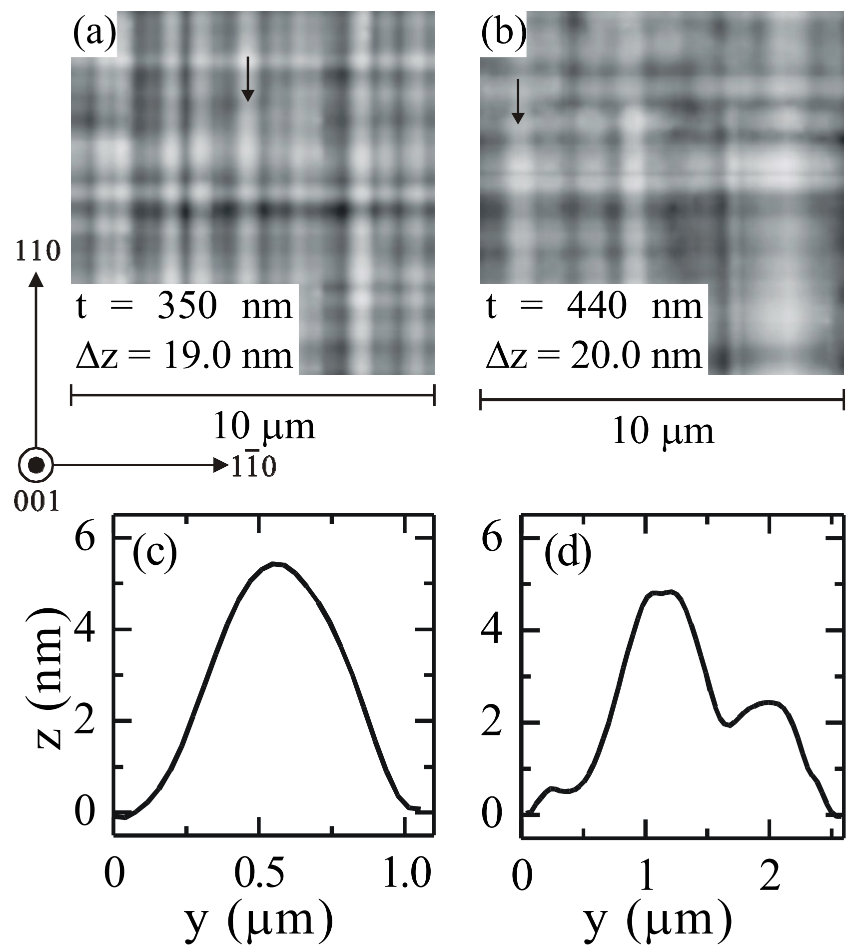
Dislocations aggregate at interface

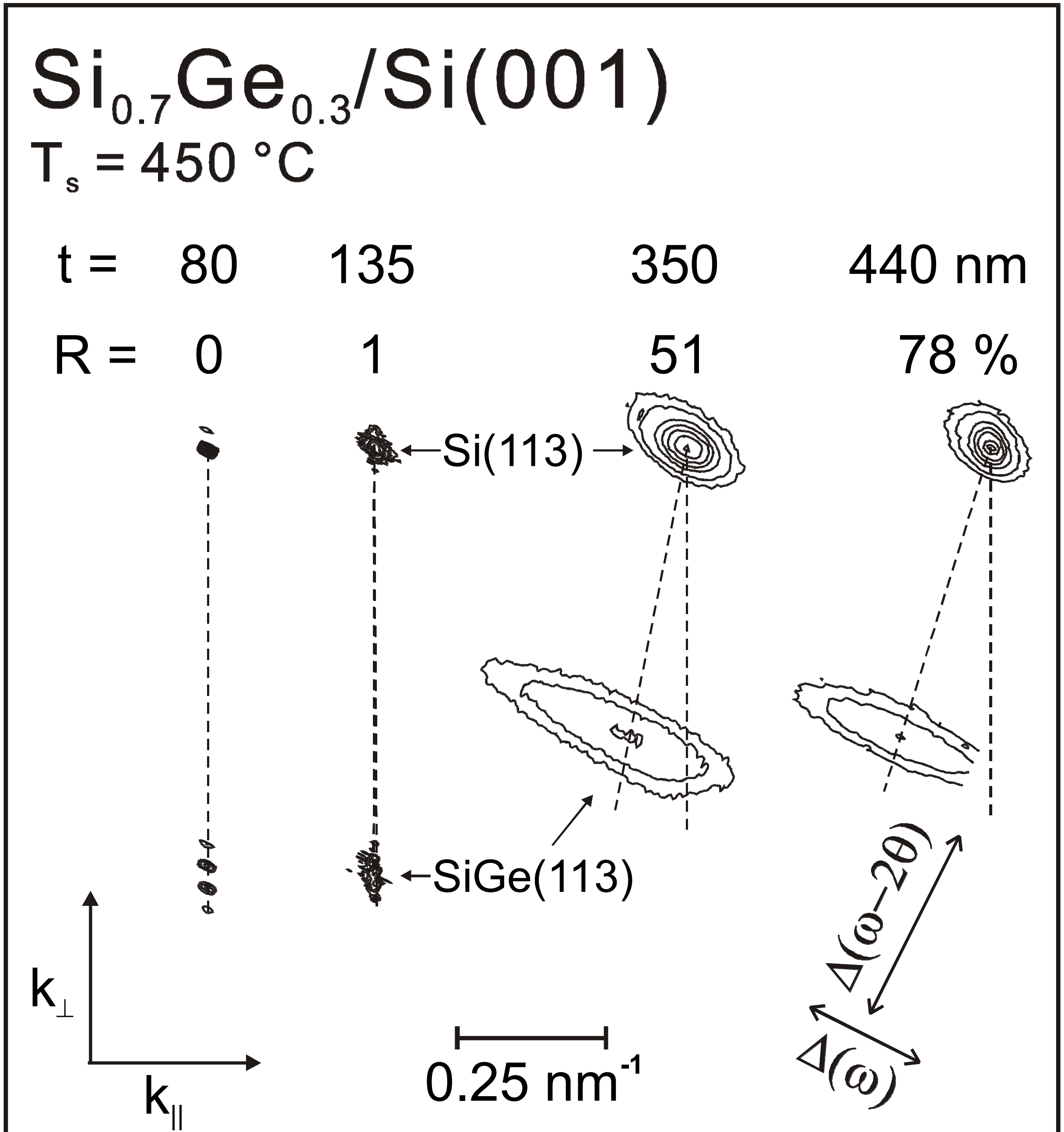
Dislocations glide to surface along {111}



AFM morphology of $Ge_{0.3}Si_{0.7}$ films

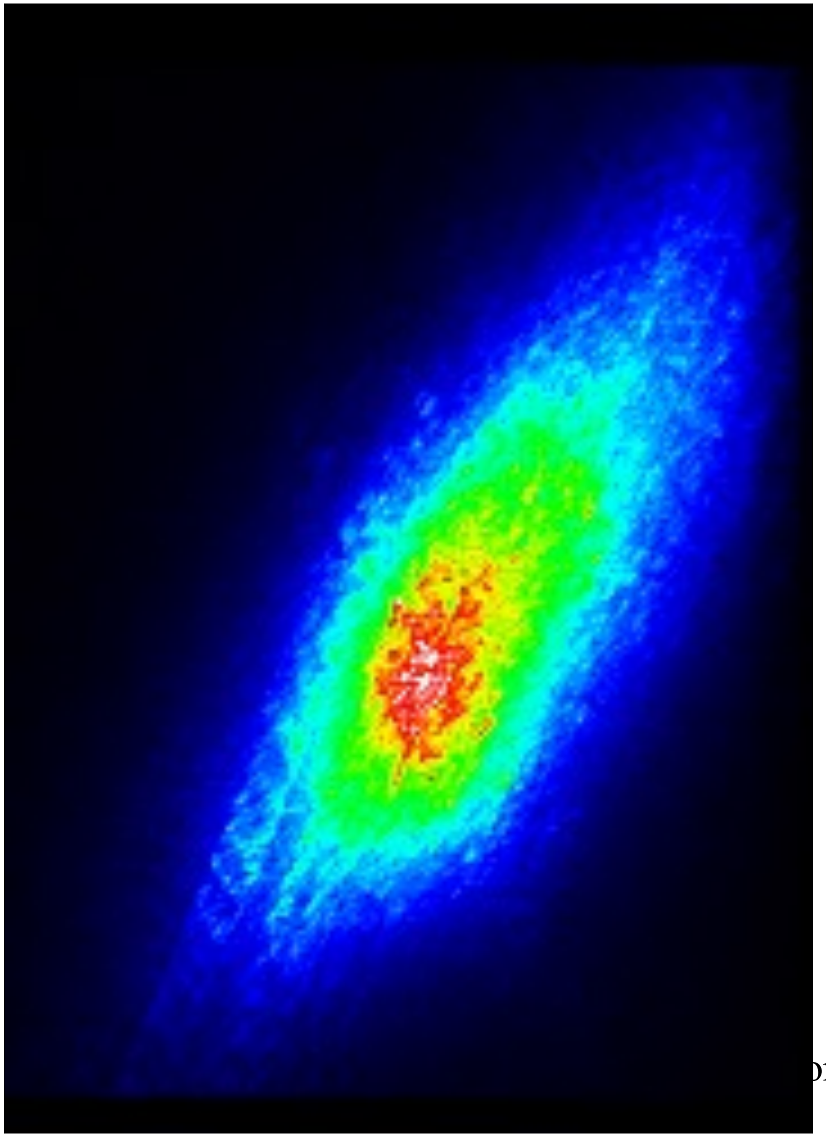
T. Spila, PhD dissertation, UIUC (2002)





Si0.7Ge0.3/Si(001)
Ts = 450 °C
t = 80 135 350 440 nm
R = 0 1 51 78 %
Si(113)
SiGe(113)
k⊥
k||
0.25 nm-1
Δ(ω–2θ)
Δ(ω)

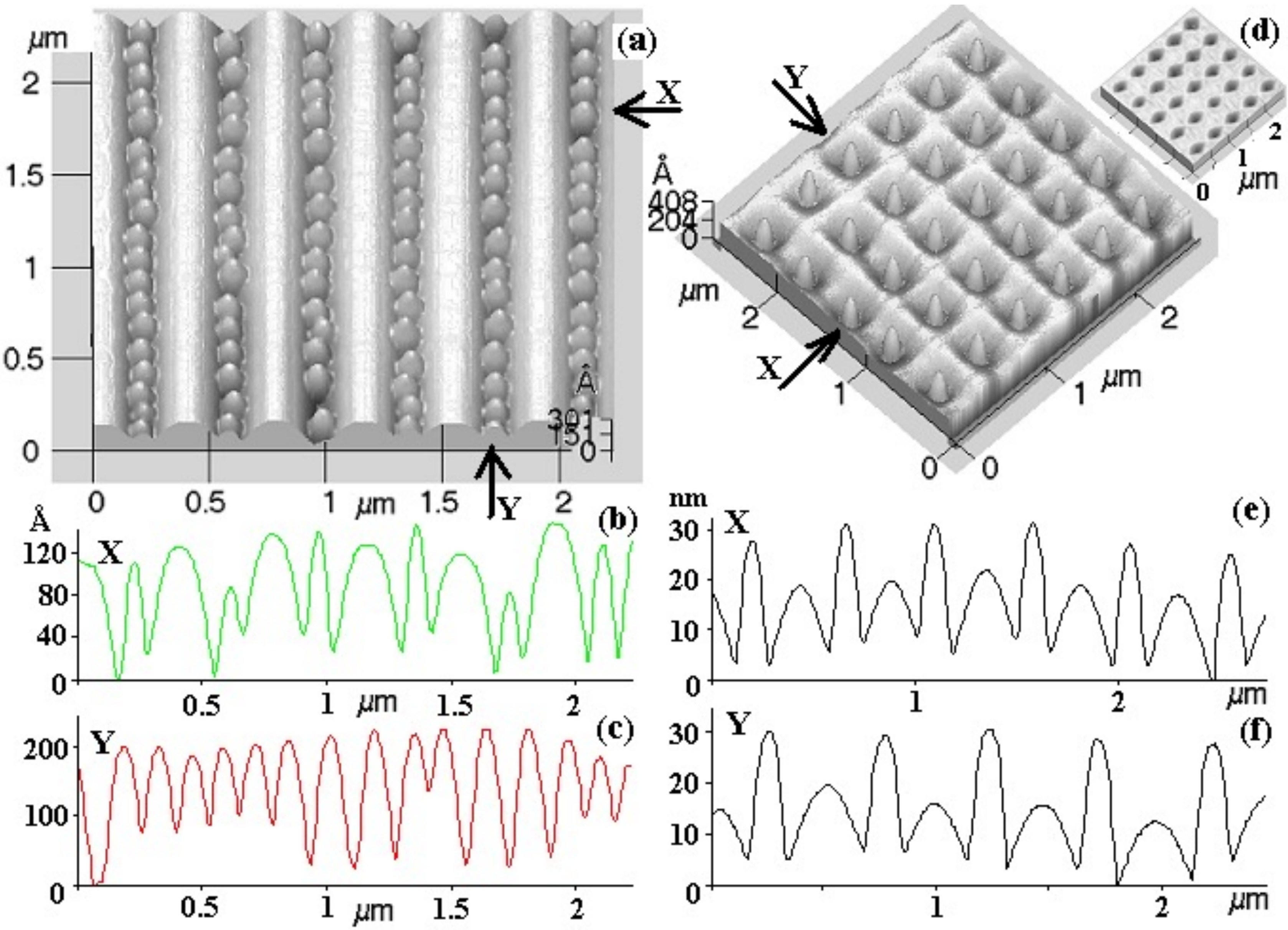
Ge_xSi_{1-x} Film Diffraction



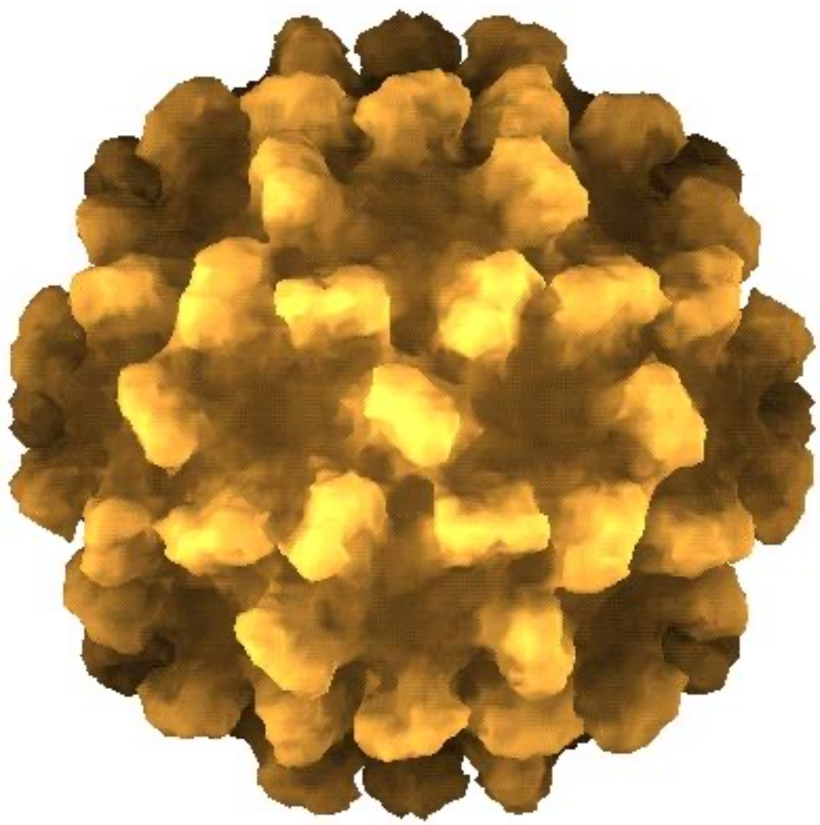
- 202 Bragg Peak
- 2° incidence angle
- 8.5 keV
- 20μm ×40μm beam onto KB mirror
- 1μm ×1μm focus
- 0.5μm sample steps
- APS 34-ID-C

1D and 2D Quantum Dot Arrays

Zhenyang Zhong, G. Bauer, Johannes Kepler Universität Linz



Tomato Bushy Stunt Virus
1980



Conclusions and Outlook

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