

Interpretation of Complex Image Density obtained by Coherent X-ray Diffraction

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CMMP Theory Seminar

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Outline

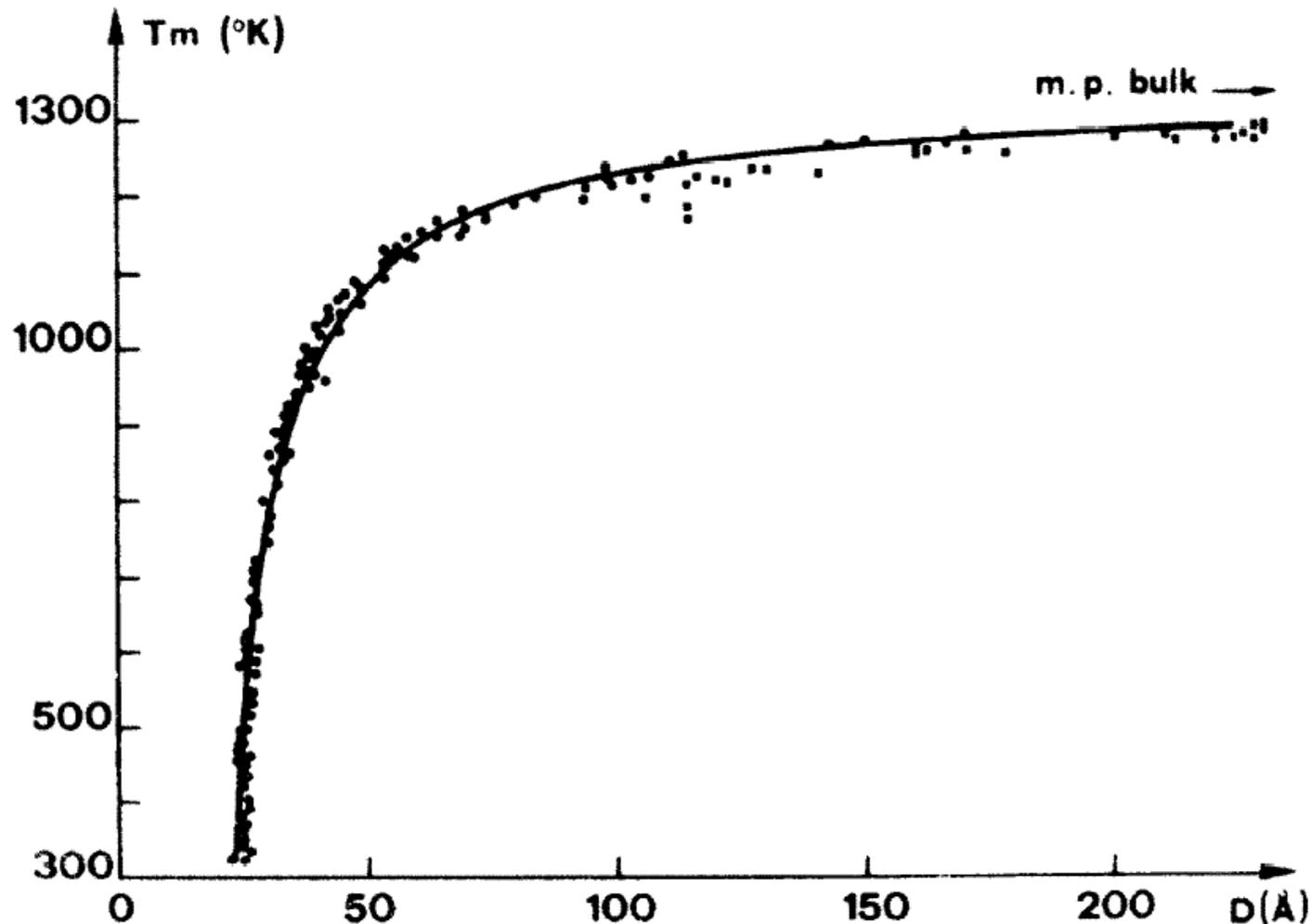
- Coherent X-ray Diffraction
- How to Solve the Phase Problem
- Nanocrystal Shapes
- Extension to Phase Objects
- Opportunities with Electrons and FELs

Theory Topics

- Uniqueness of phasing
- Phase Vortices
- Convergence of phasing algorithms
- “Noisy” Fourier Transforms
- Strain distribution within nanomaterials

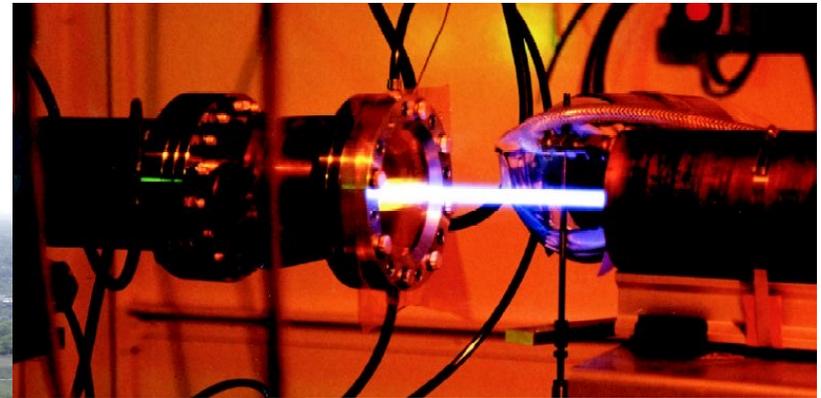
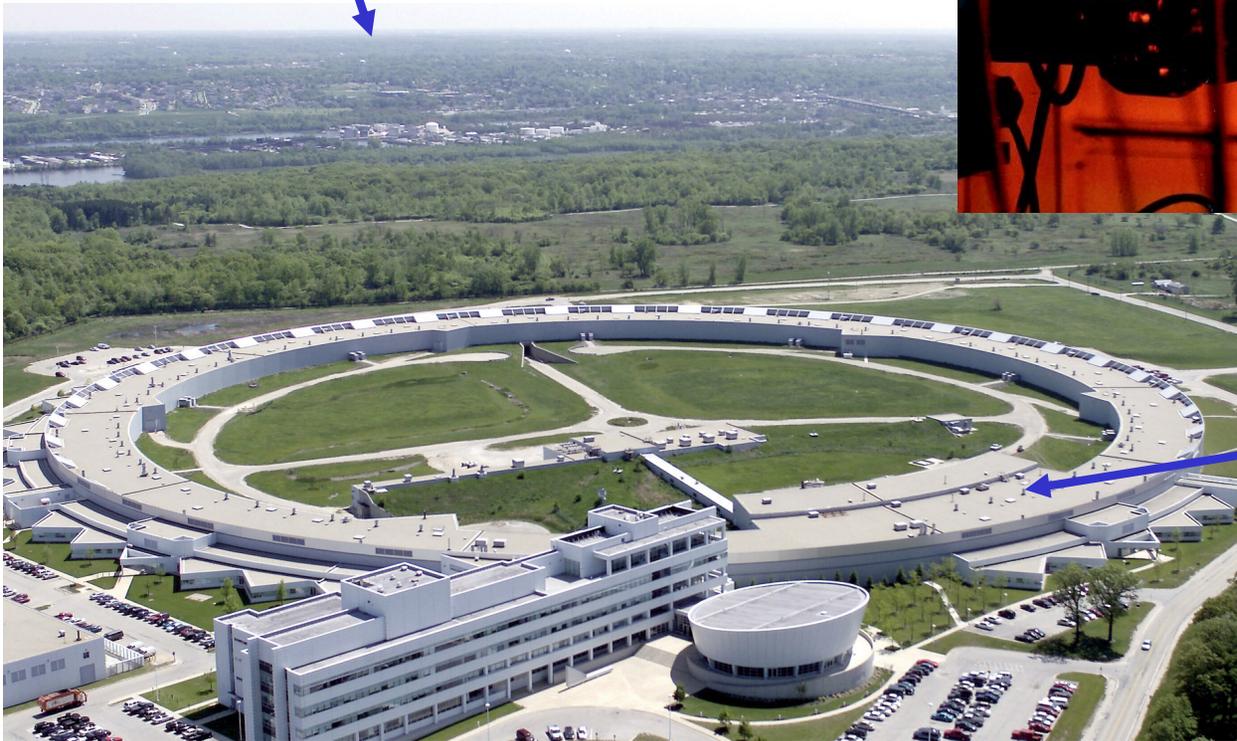
Size-dependent Melting of Au Particles

P. Buffat and J-P. Borel, Phys. Rev. A 2287-97 (1975)



Synchrotron Radiation

Urbana



34-ID-C

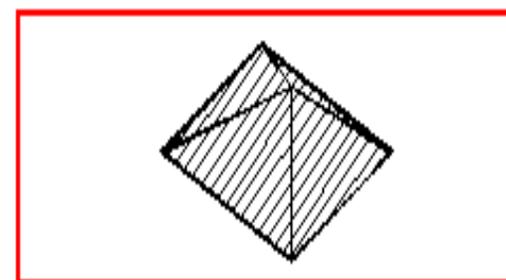
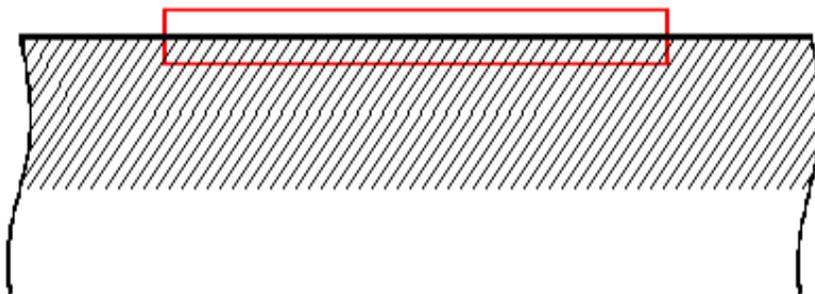


Coherence at APS, ESRF or DLS

Typical of 3rd Generation (undulator) Synchrotron Source

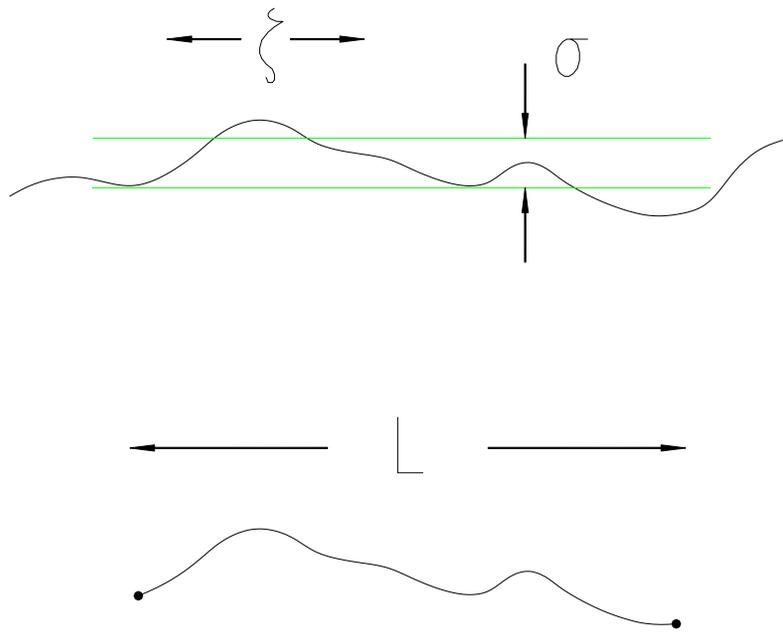
Coherence of	ξ_{VER}	ξ_{HORIZ}	ξ_{LONG}	Flux
Raw Undulator	35 μm	9 μm	0.004 μm	2×10^{12}
Si(111) Monochromator	35 μm	9 μm	1 μm	1×10^{10}
C(111) Monochromator	35 μm	9 μm	3 μm	3×10^9

Coherent region defined by slits

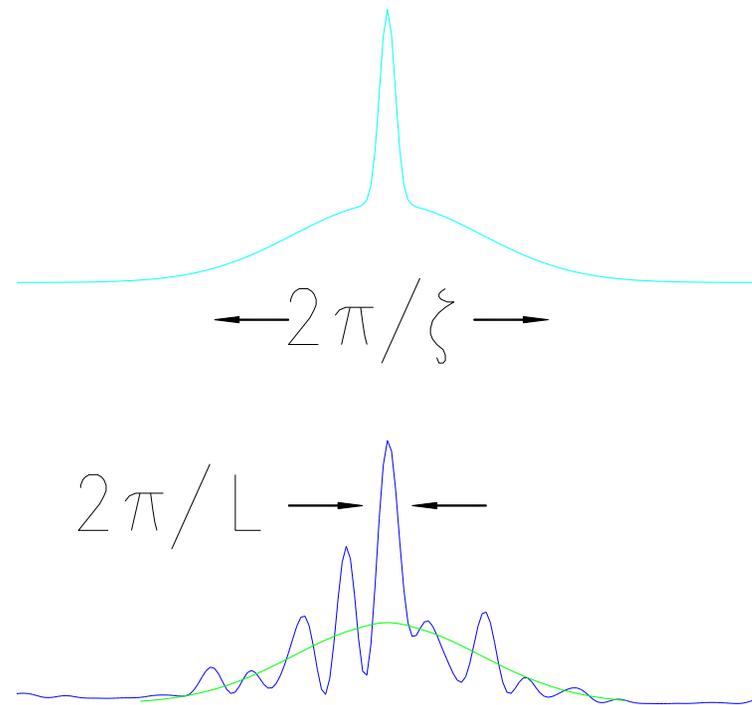


Diffuse Scattering acquires Structure using CXD

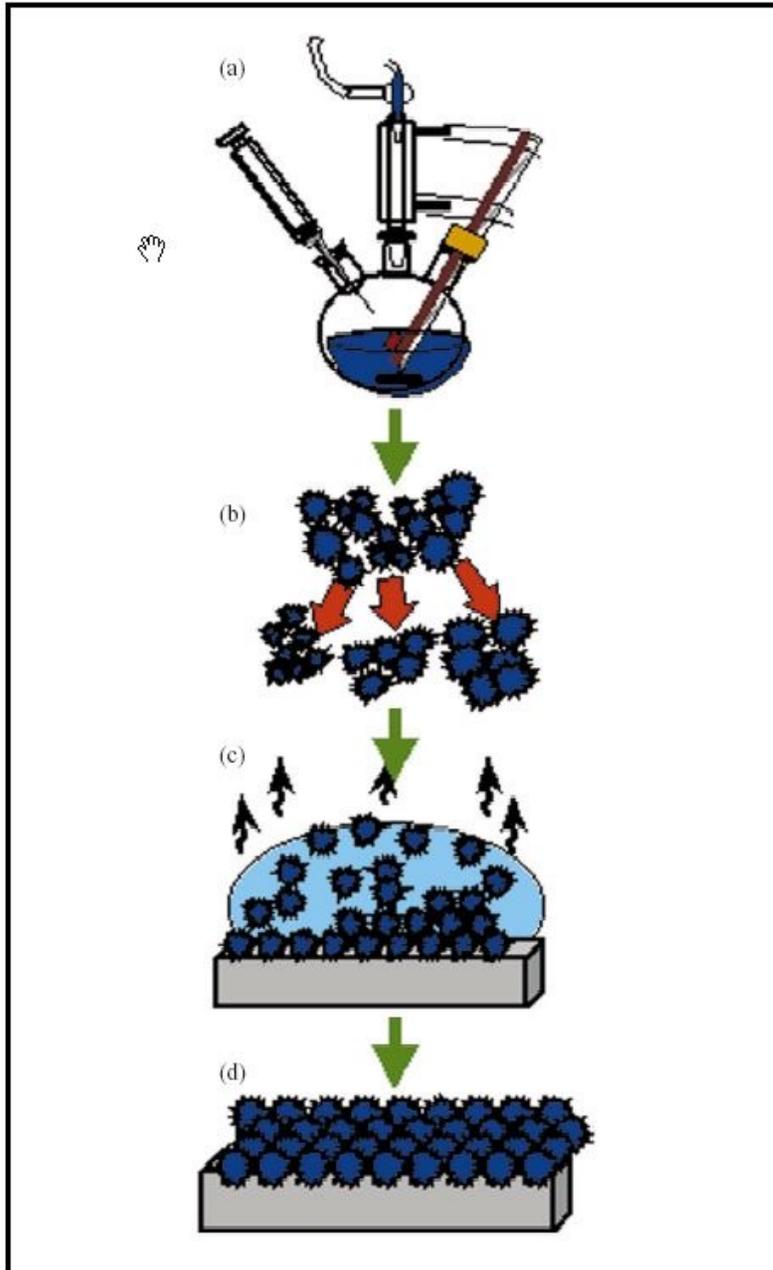
Real Space



Reciprocal Space

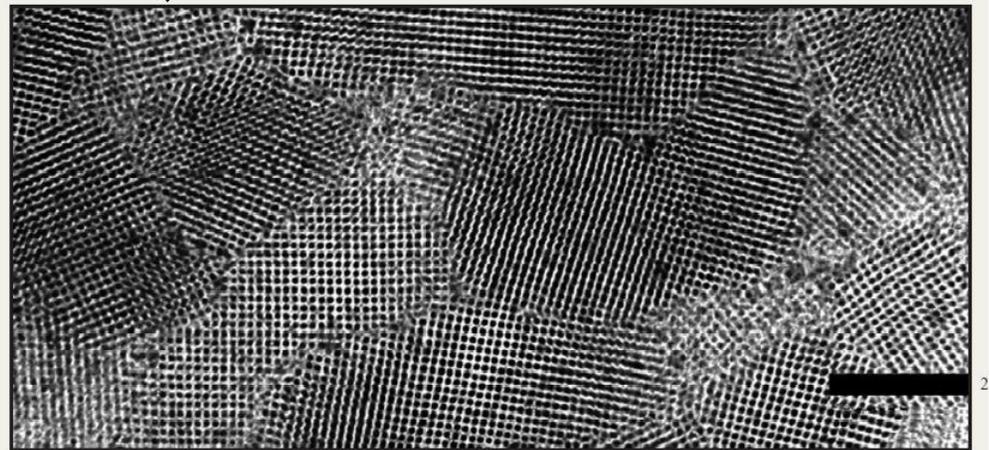


Chemical Synthesis of Nanocrystals



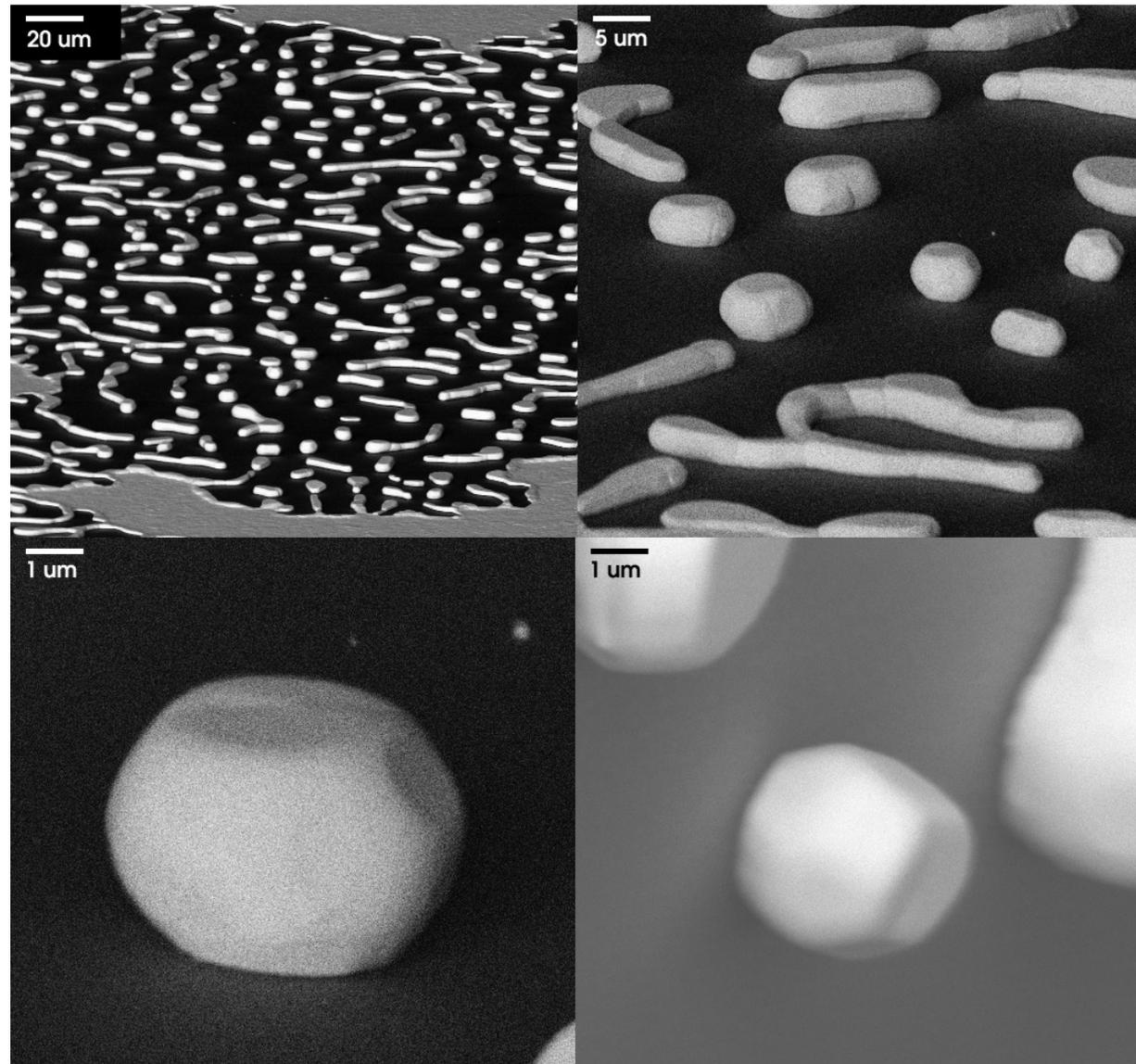
- Reactants introduced rapidly
- High temperature solvent
- Surfactant/organic capping agent
- Square superlattice (200nm scale)

C. B. Murray, IBM J. Res. & Dev. **45**
47 (2001)

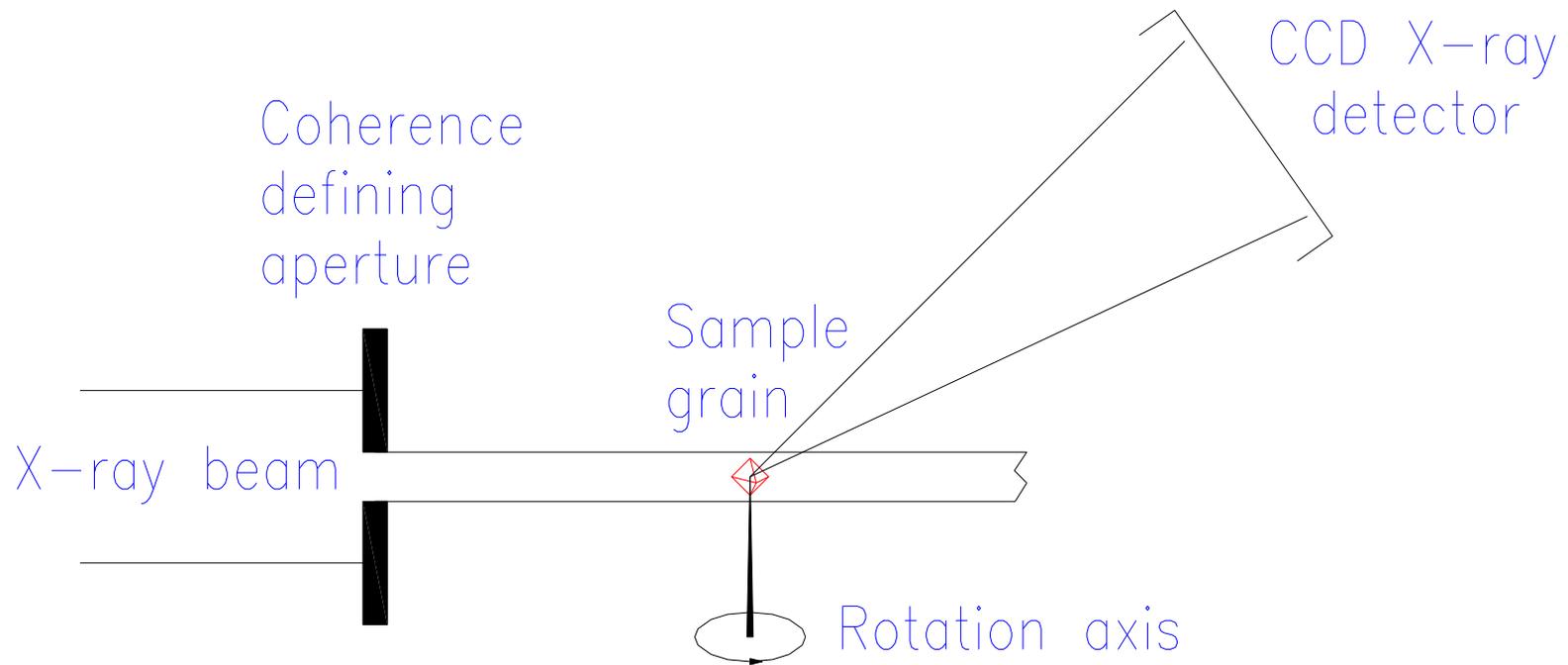


SEMS

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



Lensless X-ray Microscope



Real-space Constraints in Crystallography

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

- ‘Positivity’ and ‘Atomicity’ constraints (Sayre)
- Finite **support**, molecular envelope
- Solvent flattening/Molecular replacement
- Non-crystallographic symmetry
- Non-uniqueness is ‘pathologically rare’ ($d > 1$)
- Uses memory to avoid stagnation (Fienup HIO)

Methode HIO de Fienup

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

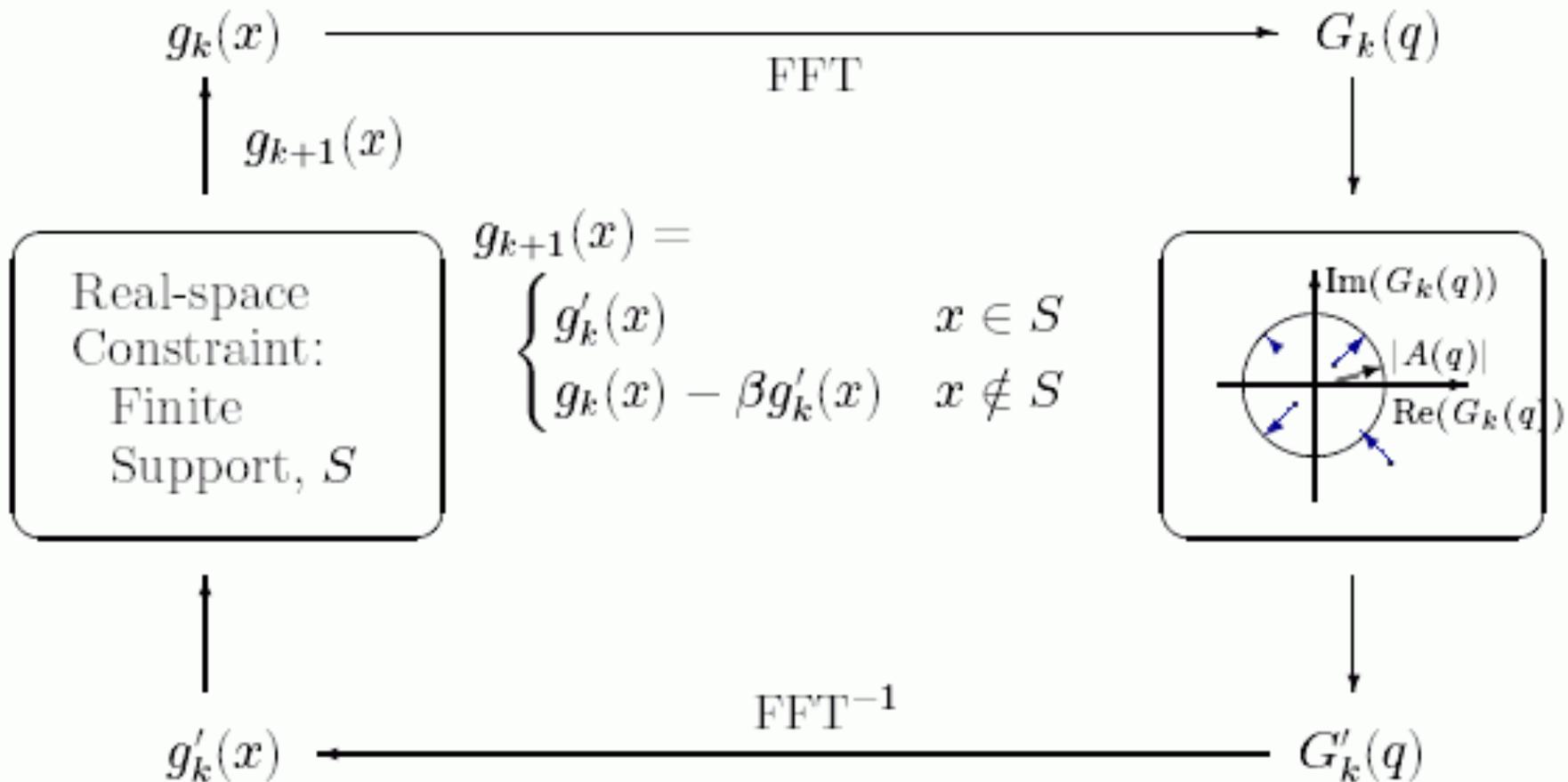
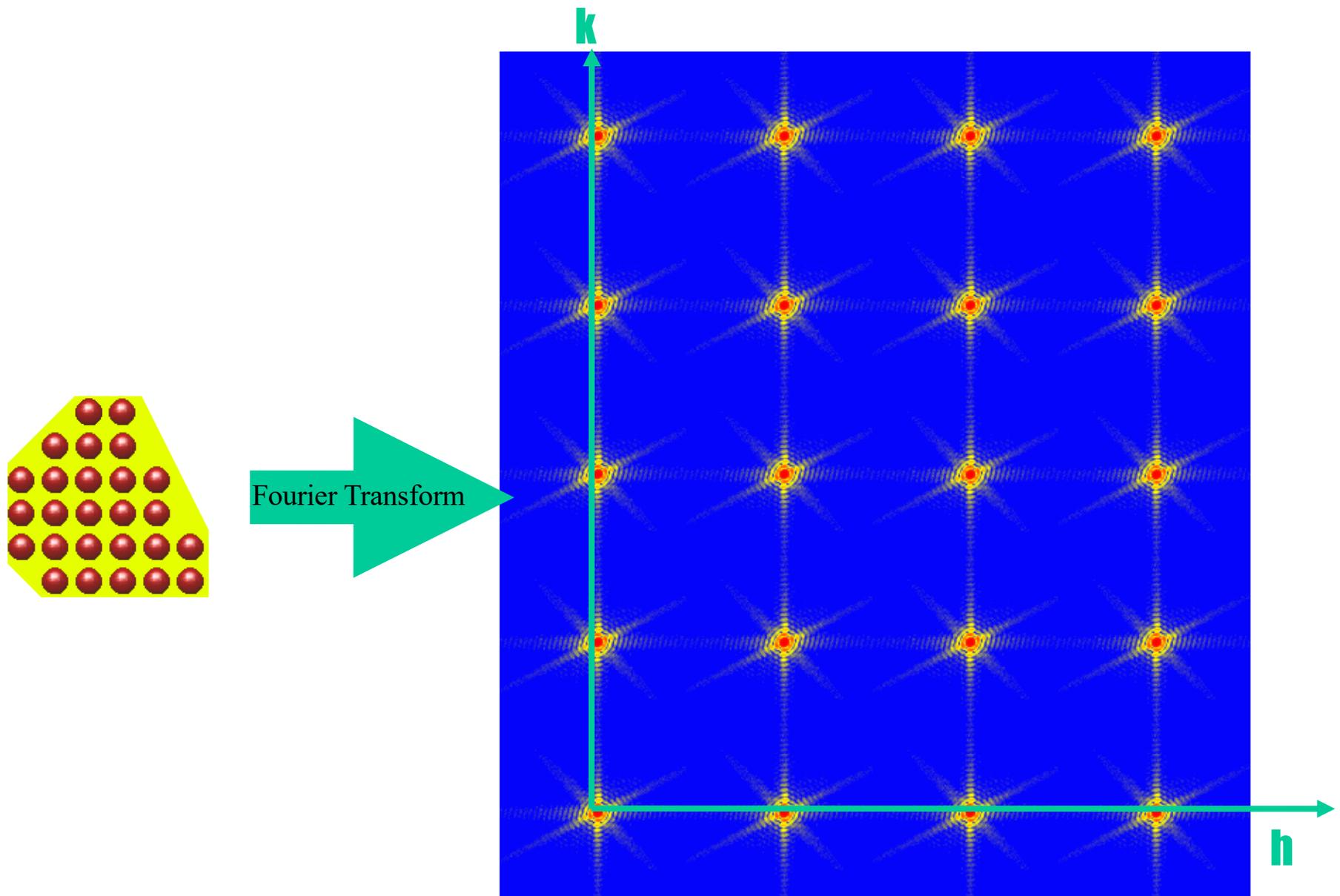
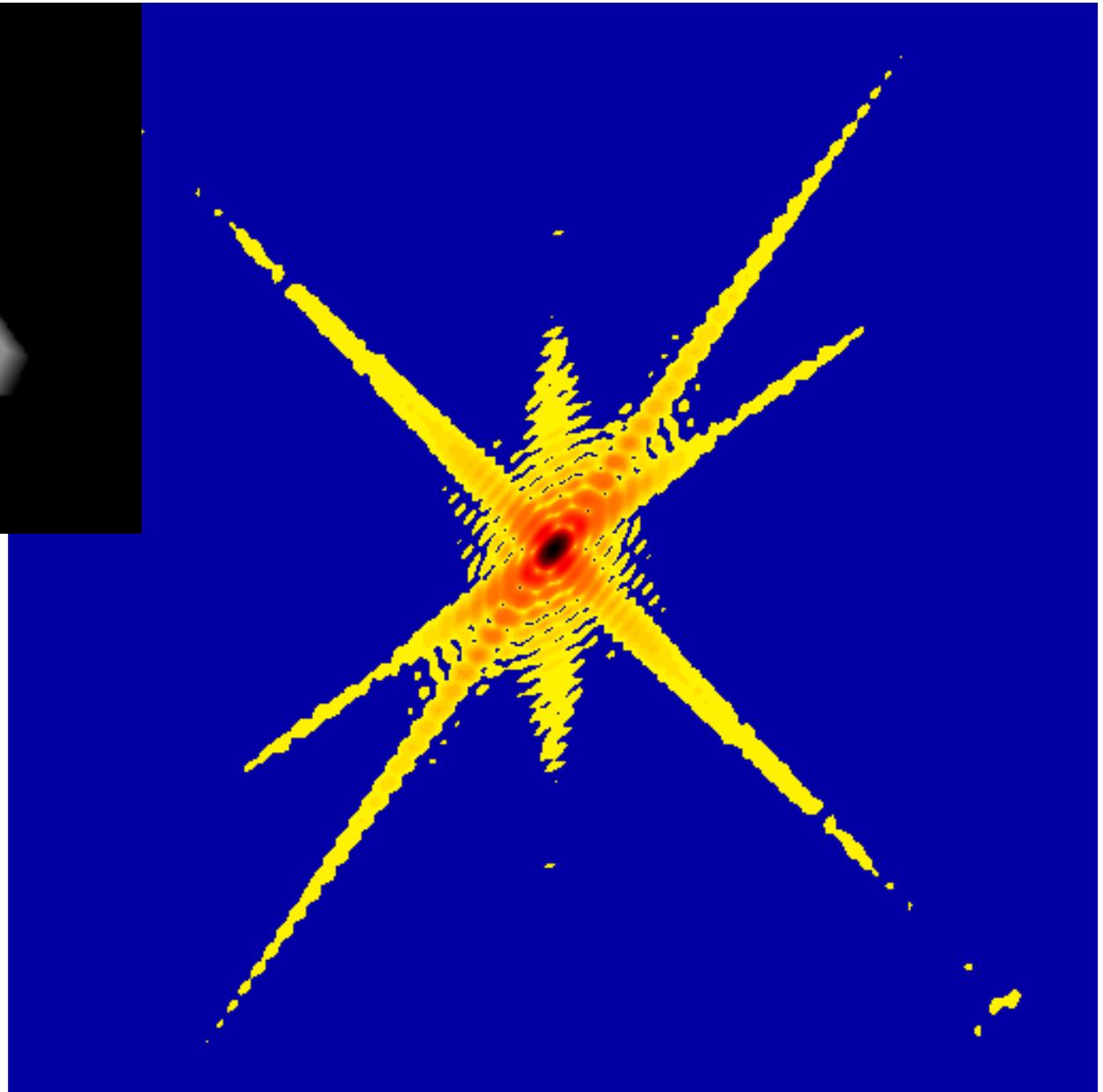
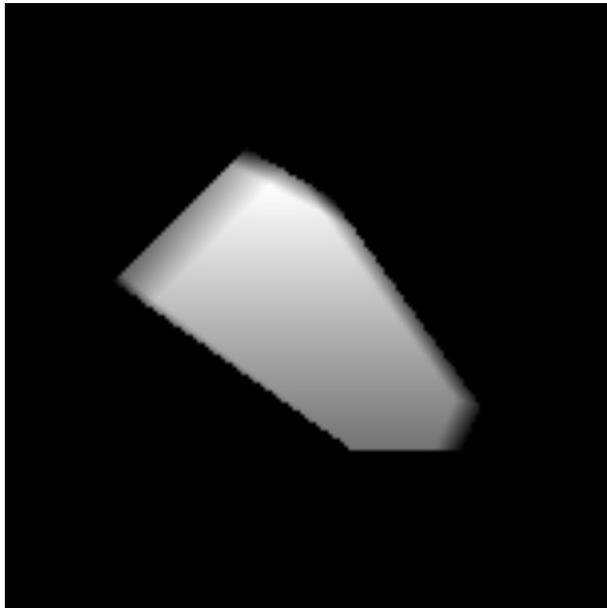


Figure 2.3: Fienup HIO

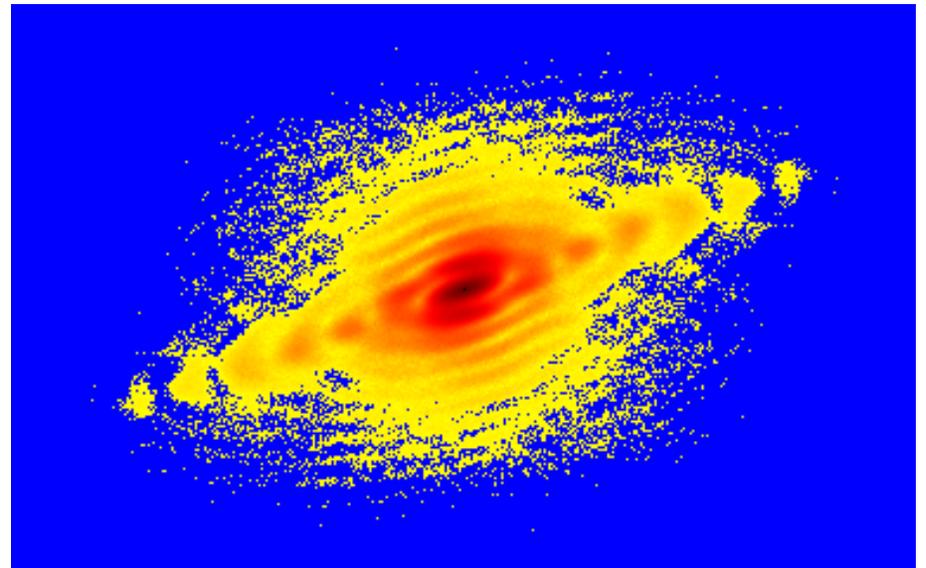
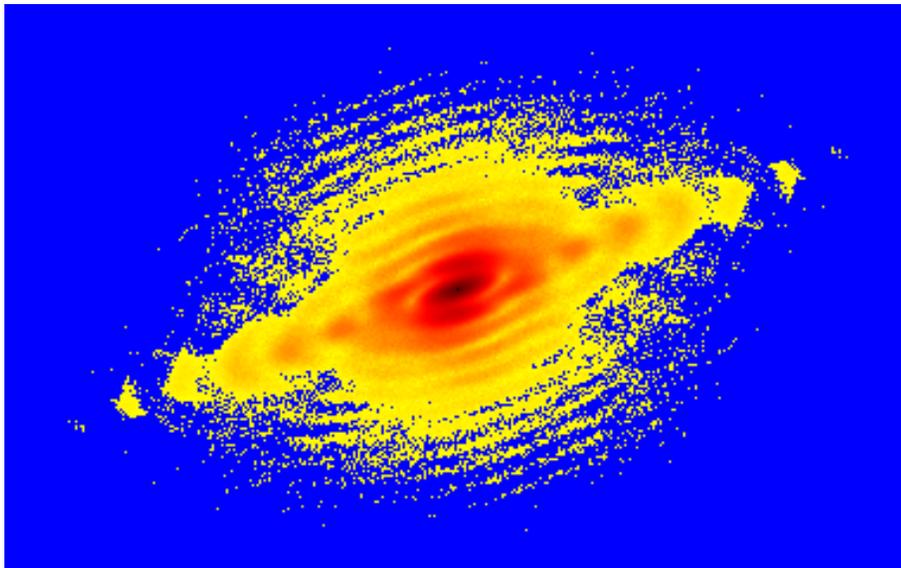
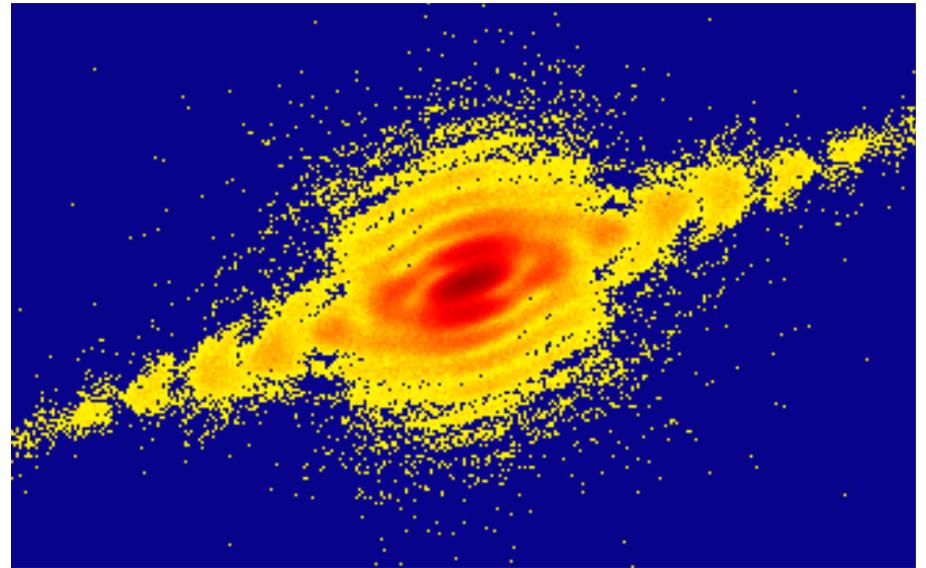
Coherent Diffraction from Crystals





Symmetrized Data and two best fits

Chisq=0.0005



Homometric structures

$$F(Q) = F_1(Q)F_2(Q)$$

$$I(Q) = |F_1(Q)|^2|F_2(Q)|^2$$

$$F_j(Q) = \int \rho_j(x)e^{iqx} dx$$

$$F_j^*(Q) = \int \rho_j(-x)e^{iqx} dx$$

$$I(Q) = \left| \int \underline{\rho_1(x) \circ \rho_2(x)} e^{iqx} dx \right|^2$$

$$I(Q) = \left| \int \underline{\rho_1(-x) \circ \rho_2(x)} e^{iqx} dx \right|^2$$

Identical diffraction
from two structures
whenever the structure
factor is factorizable

Demonstration of Uniqueness

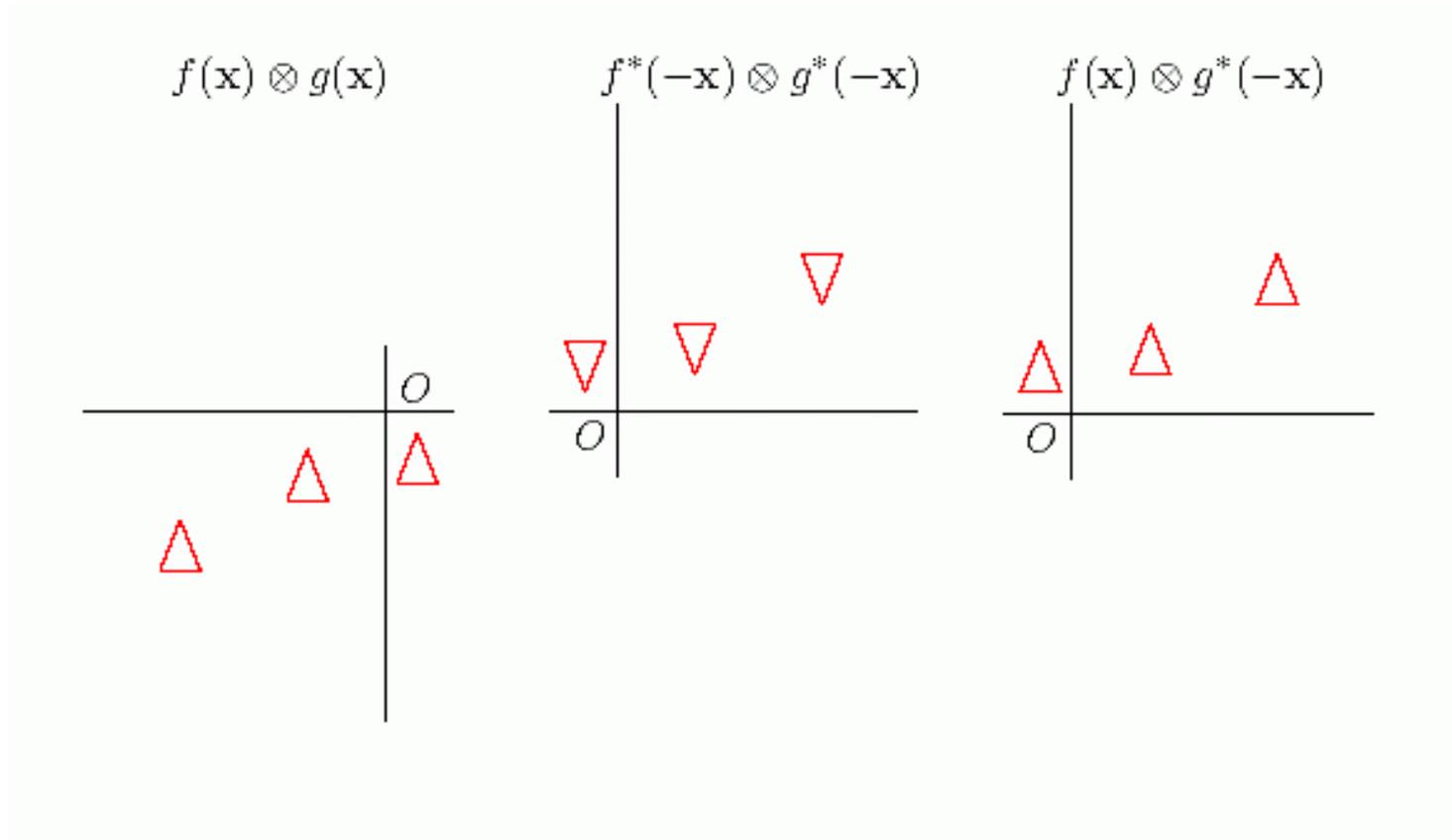
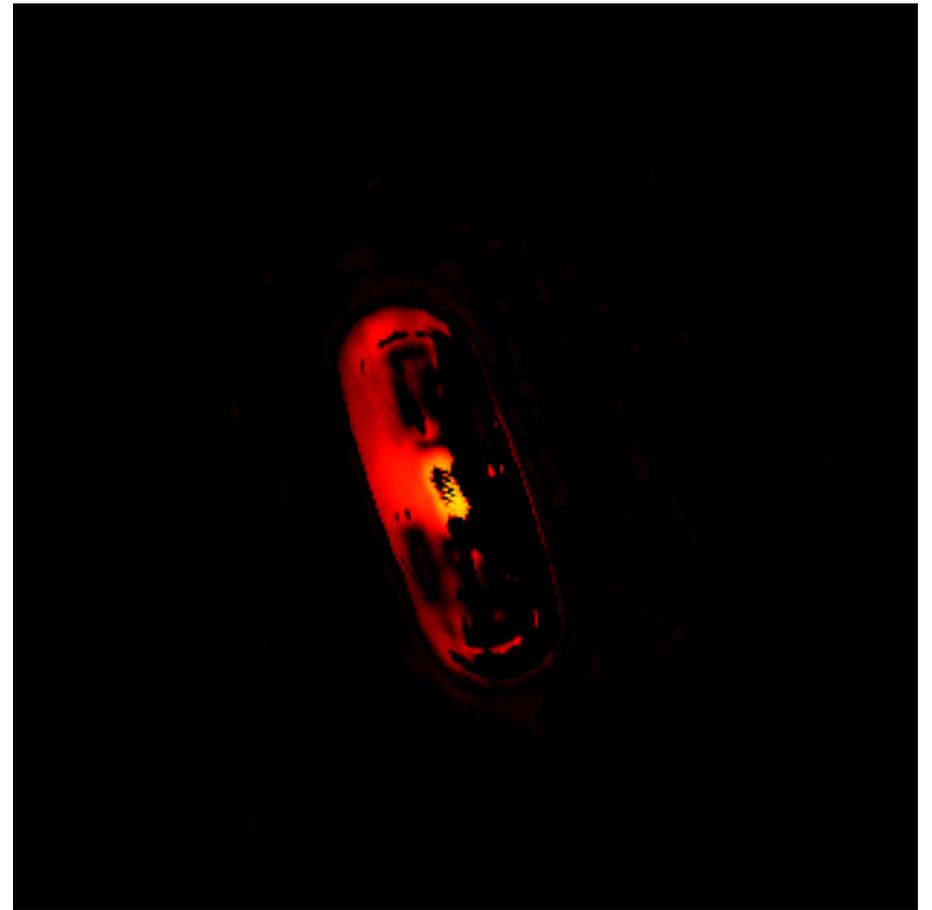
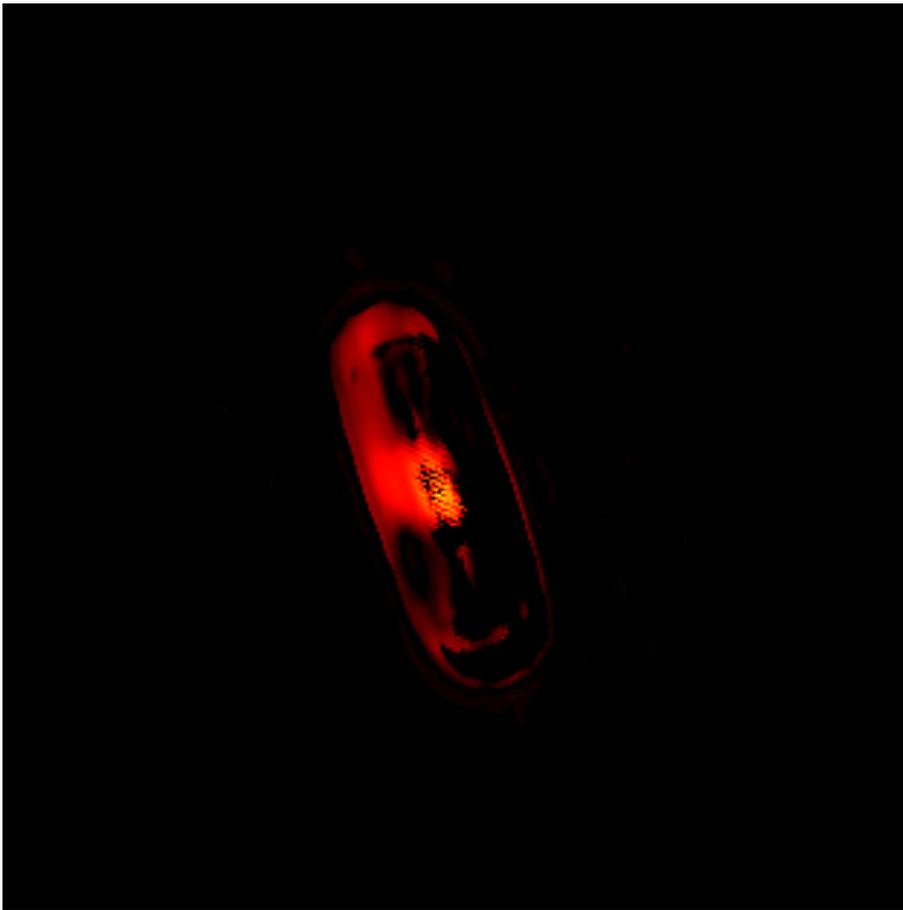


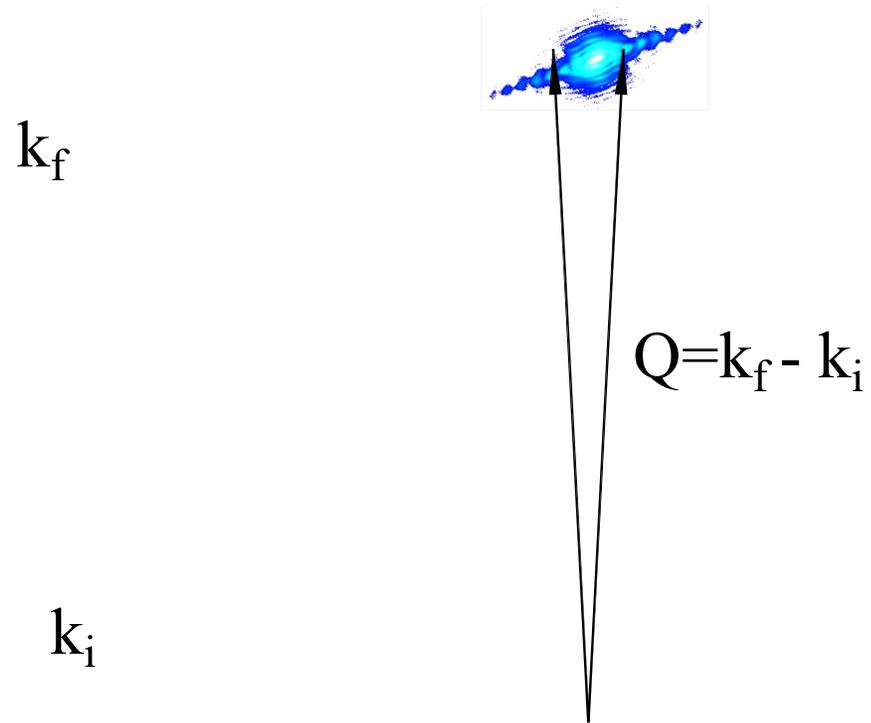
Figure 1.1: All three examples have the same Fourier magnitude, demonstrating that ambiguous solutions are possible in 2D. $f(\mathbf{x})$ is the shape of the triangle. $g(\mathbf{x}) = \delta(\mathbf{x} - \mathbf{x}_1) + \delta(\mathbf{x} - \mathbf{x}_2) + \delta(\mathbf{x} - \mathbf{x}_3)$, where the vectors \mathbf{x}_i are from the origin to the center of each triangle. The leftmost two are the “correct” solution and its twin—a trivial ambiguity—while the third demonstrates that if $|F(\mathbf{k})||G(\mathbf{k})|$ can be factorized an ambiguous solution may exist.

2D Reconstructions

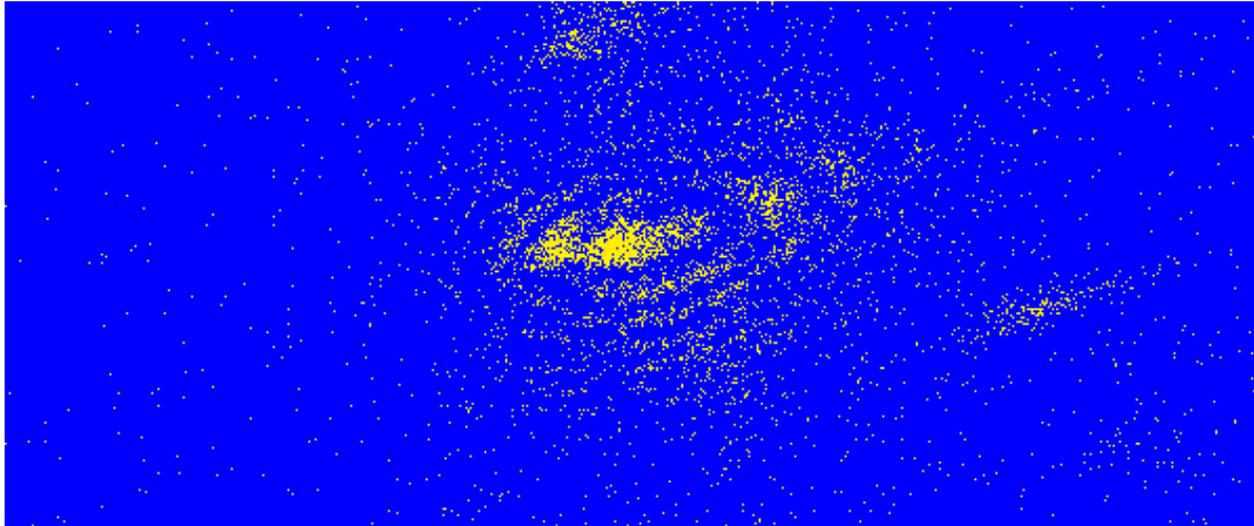
chisquare = 0.0005

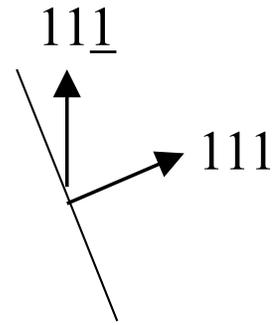
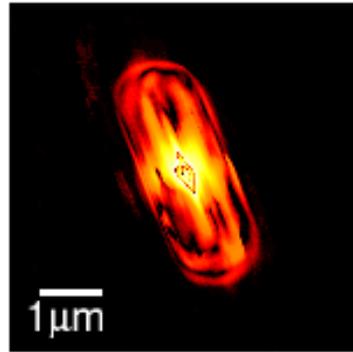
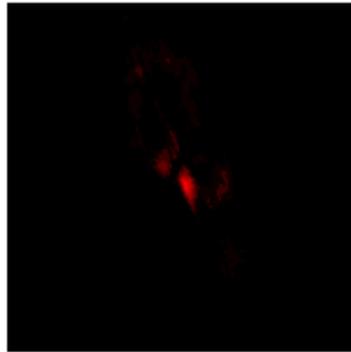


3D Diffraction Method

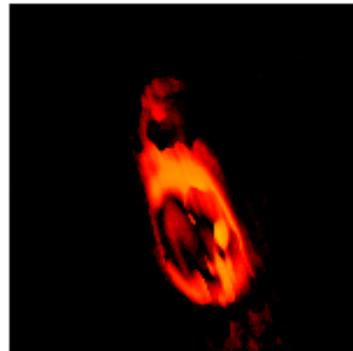
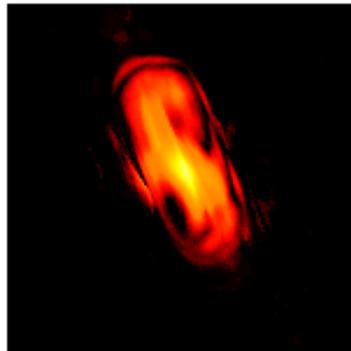
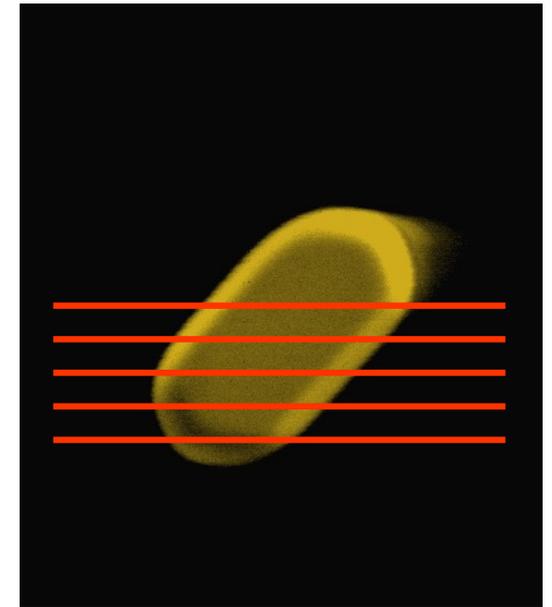
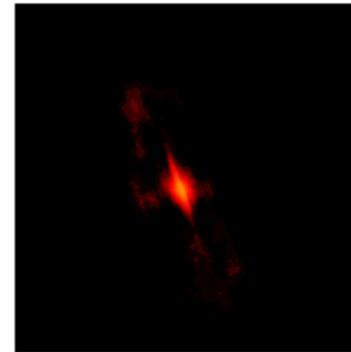
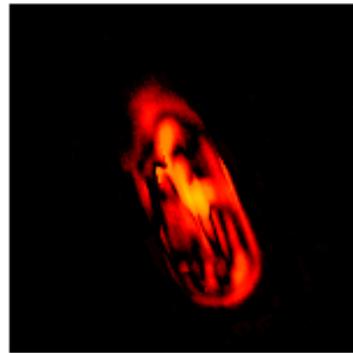
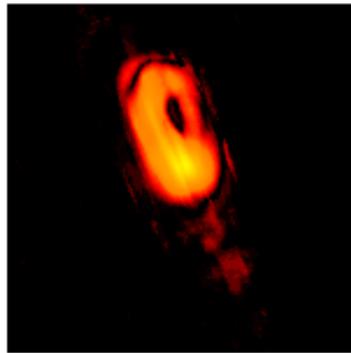
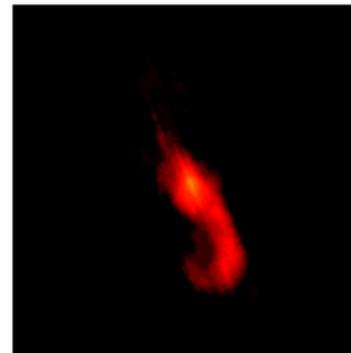
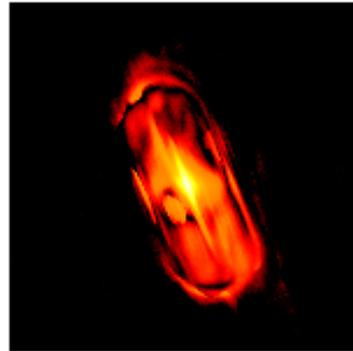
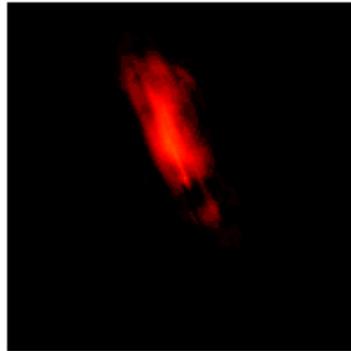


3D Diffraction Data 1 micron Au crystal



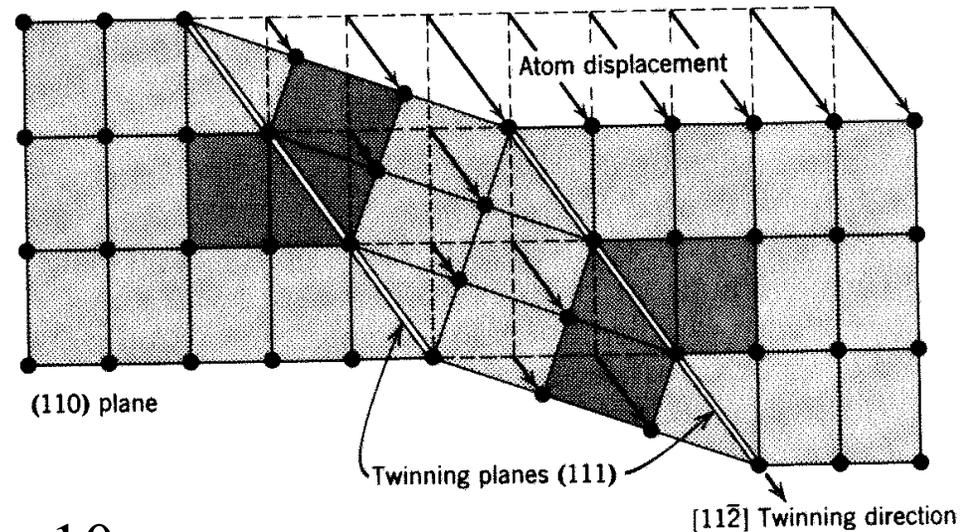
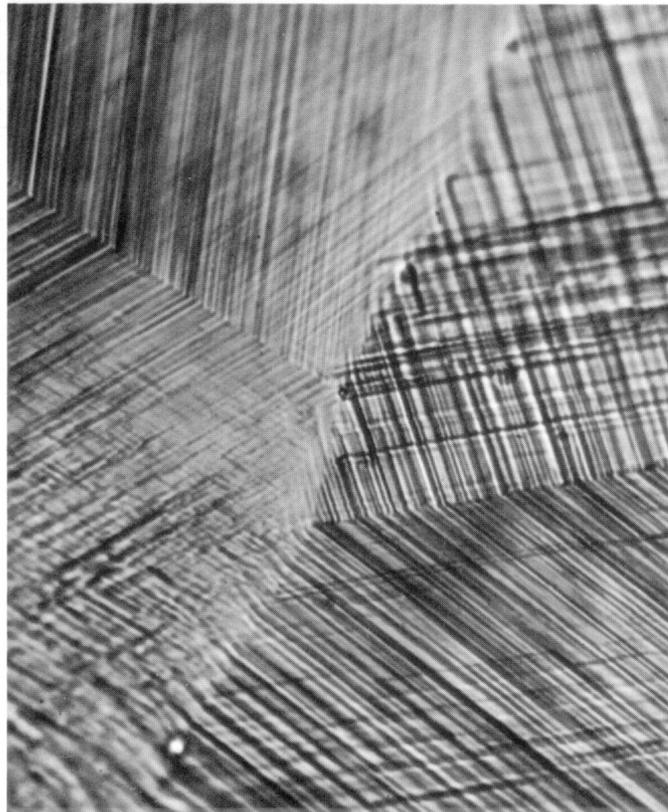


Slices through
plan view SEM:



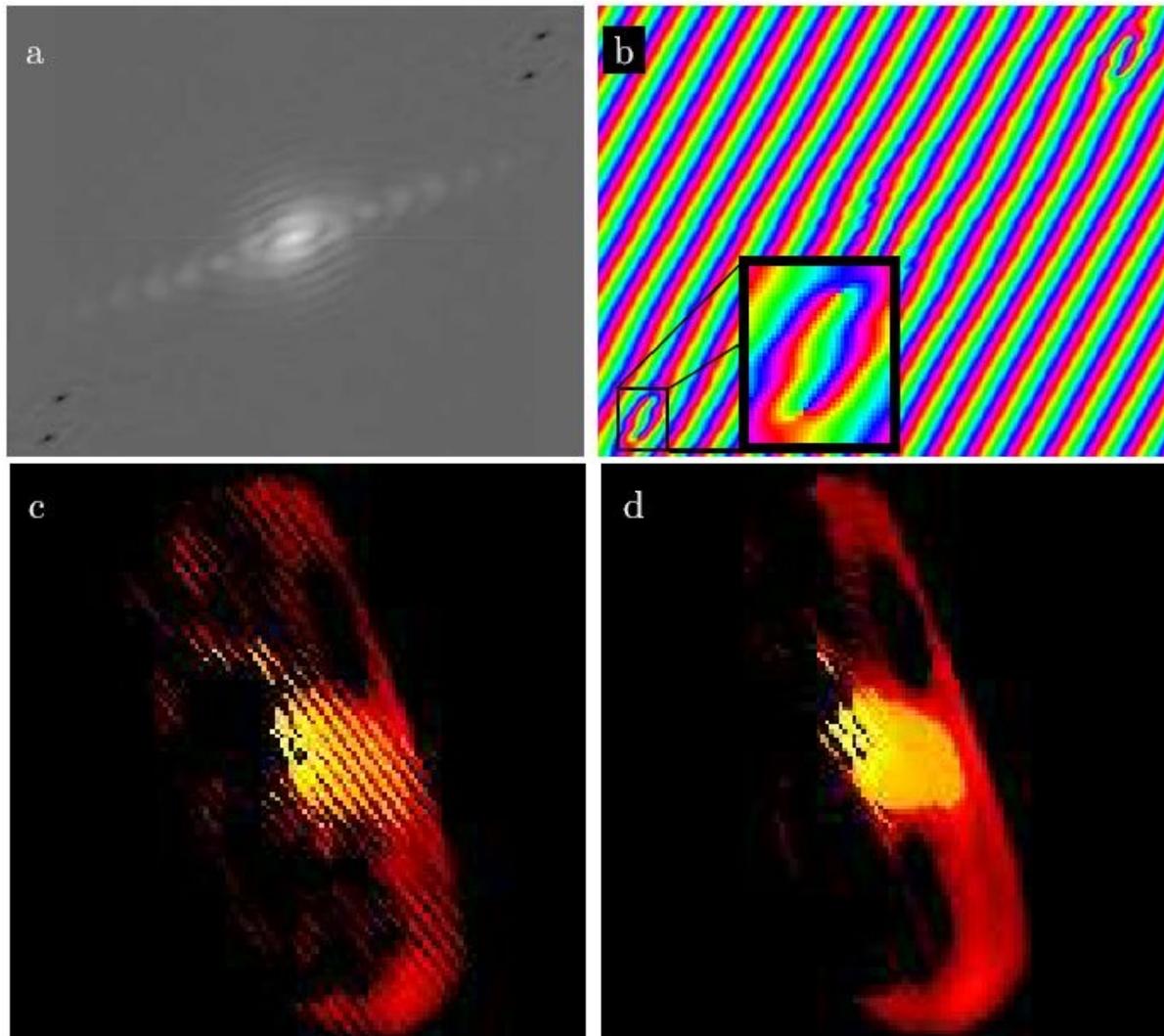
Twinning in deformed FCC metals

J. Wulff, "Structure and Property of Materials III" (1965)

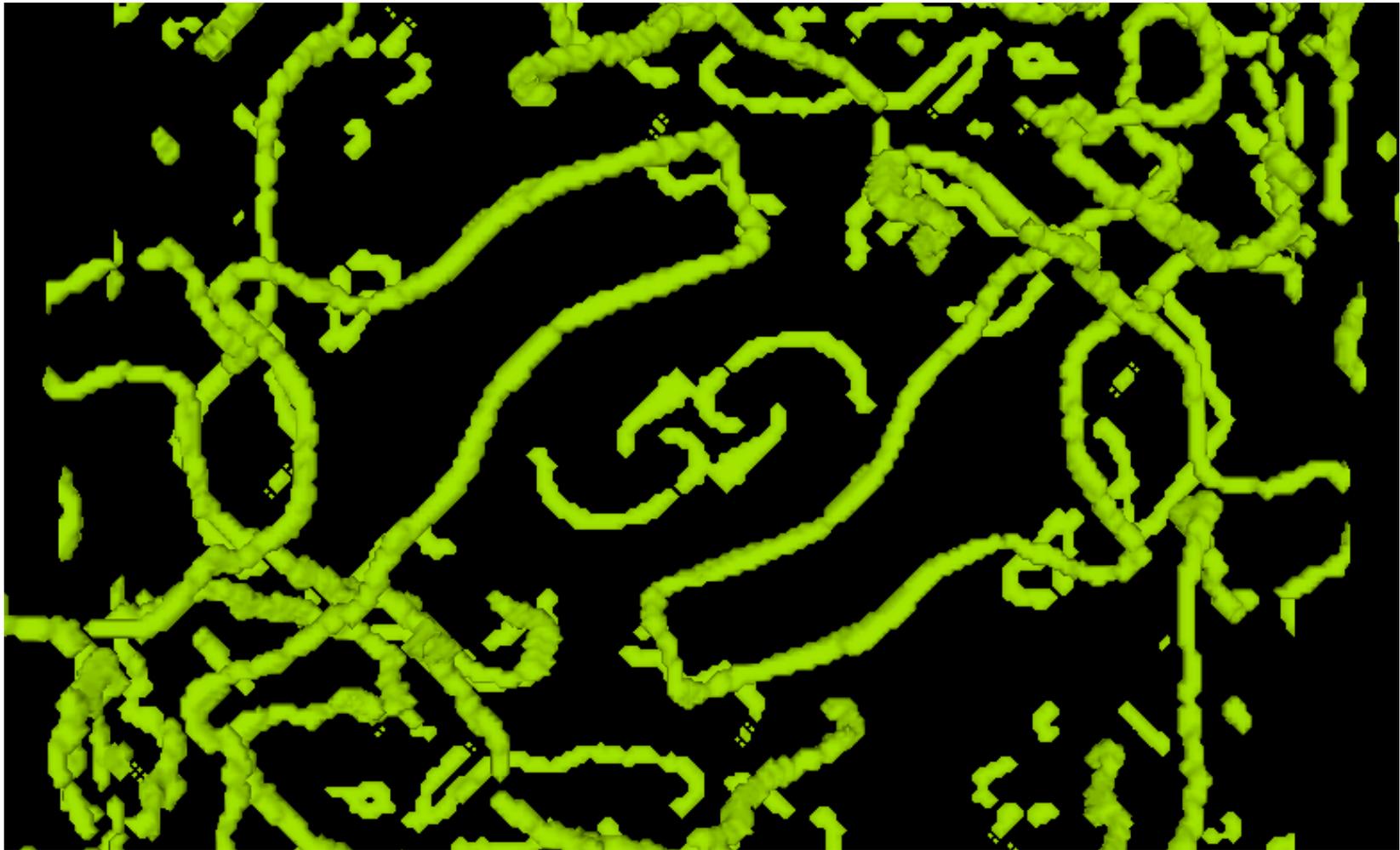


■ $\sim 10\mu\text{m}$
Cu

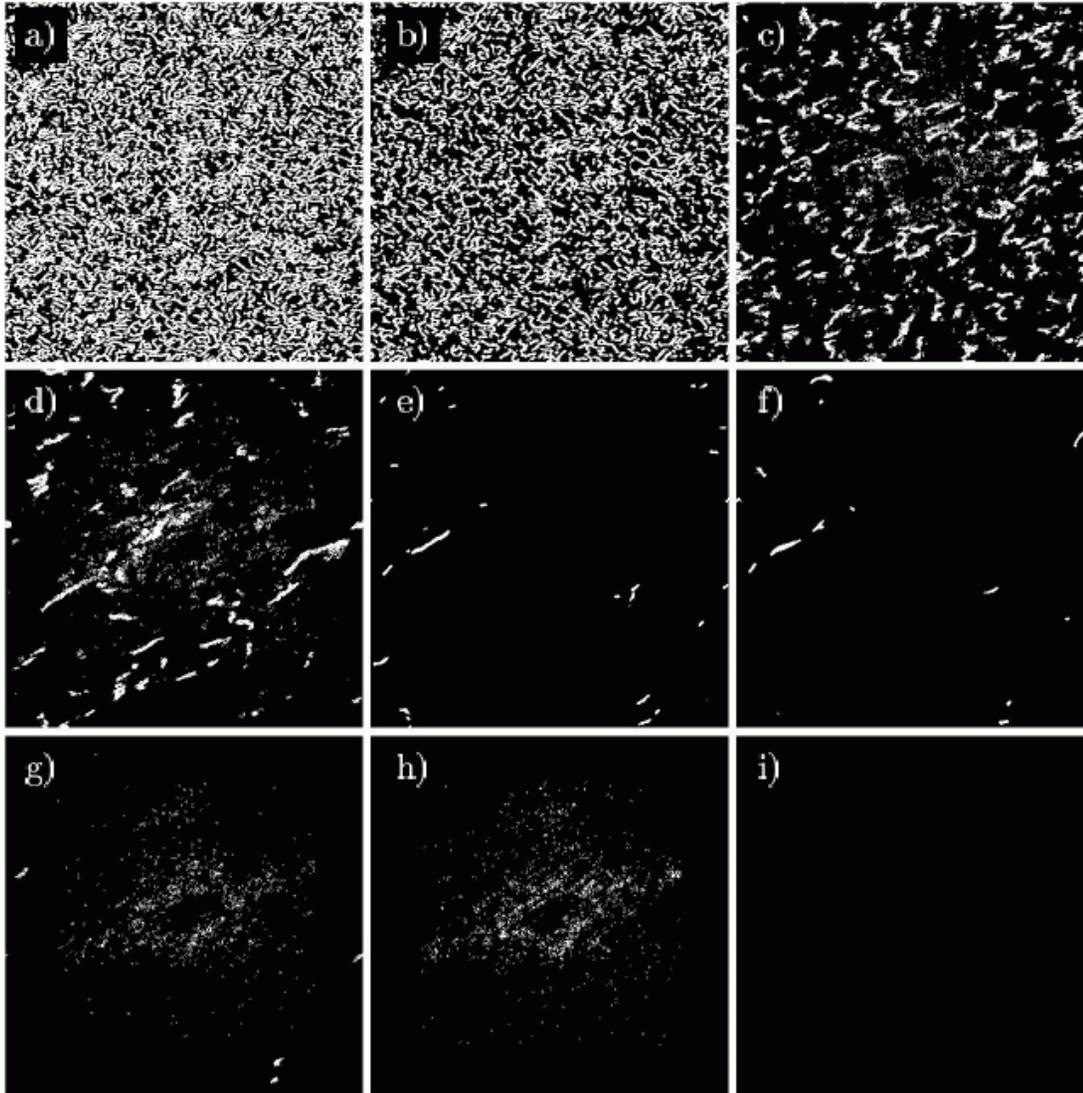
Phase Vortices and patching



'Vortices' Form Loops in 3D

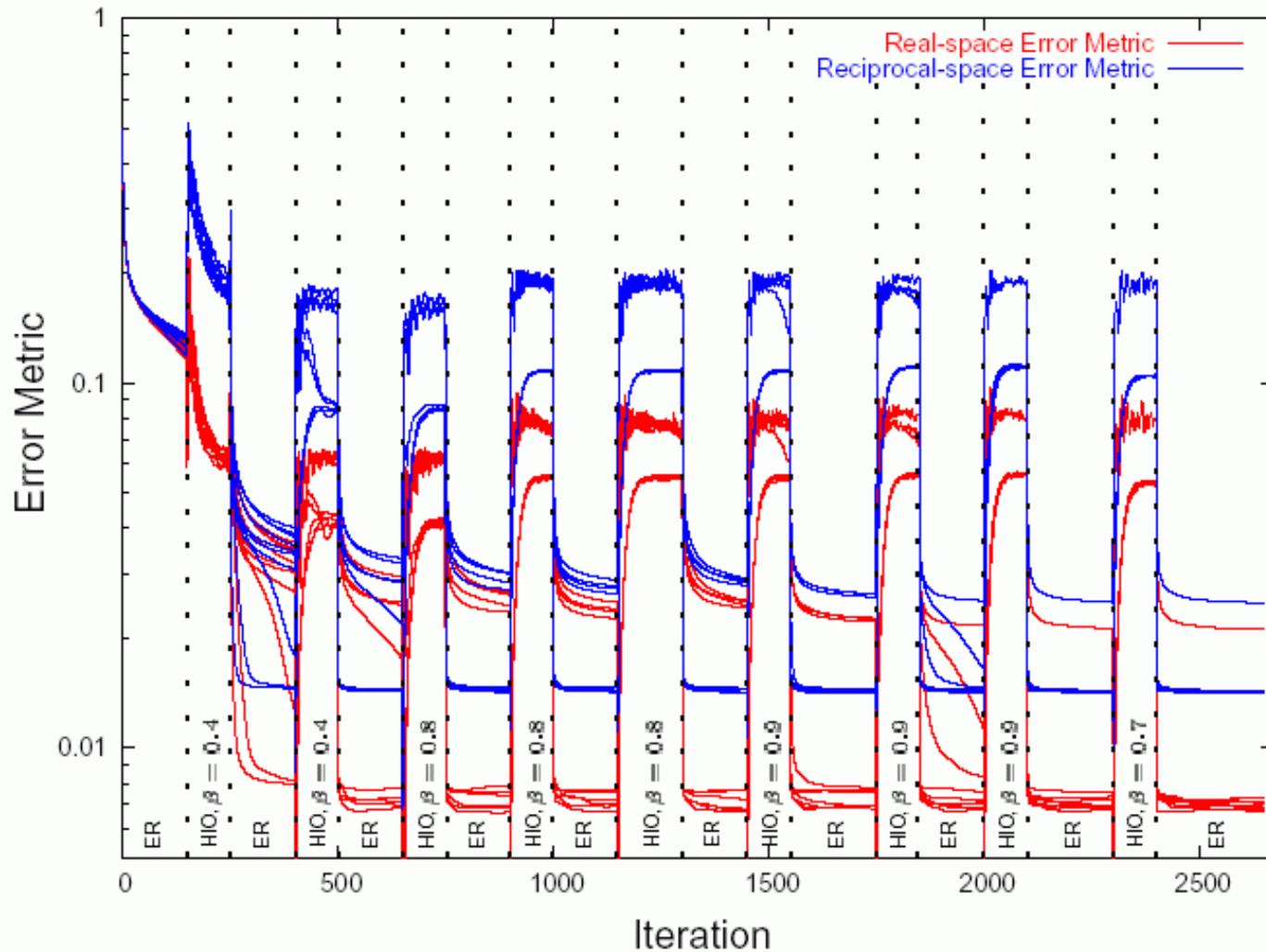


Clearing of Vortices during HIO



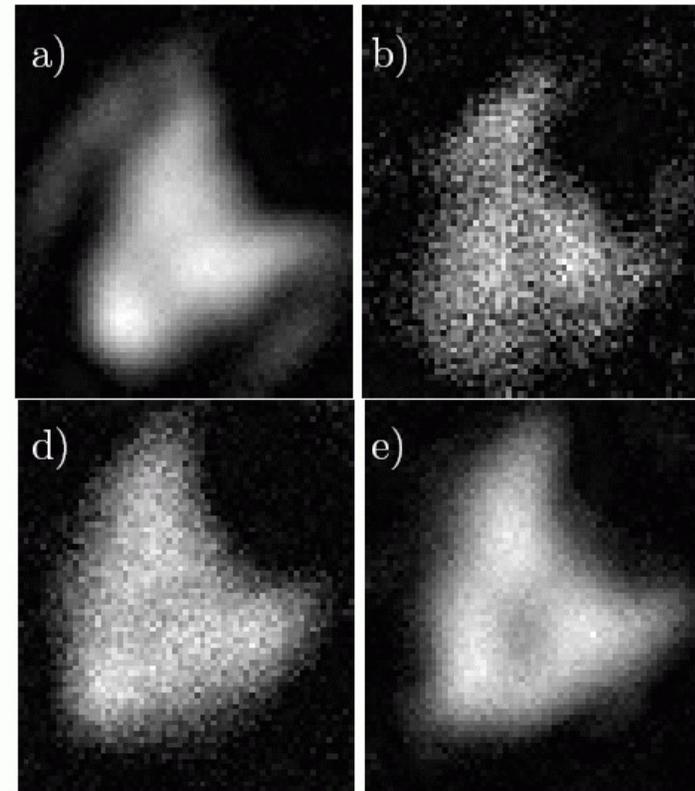
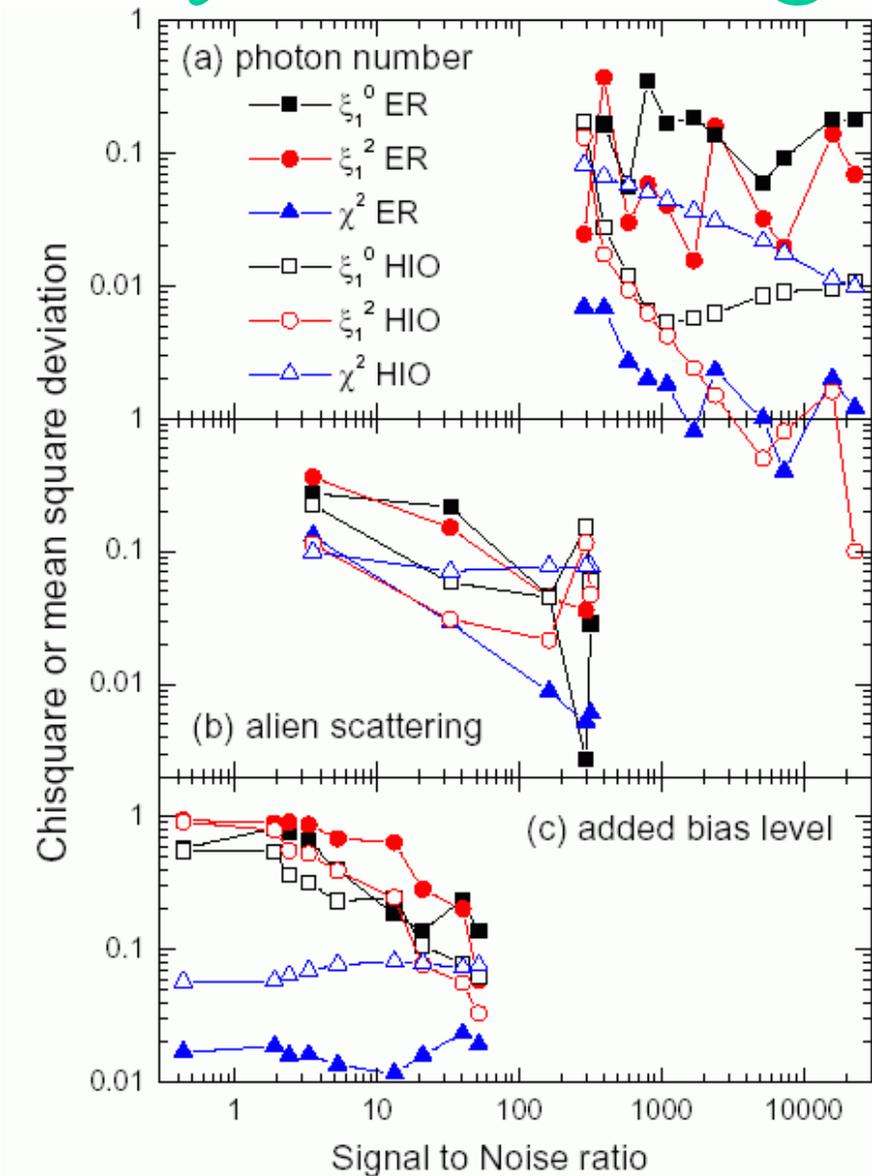
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HIO ER ER
HIO HIO ER

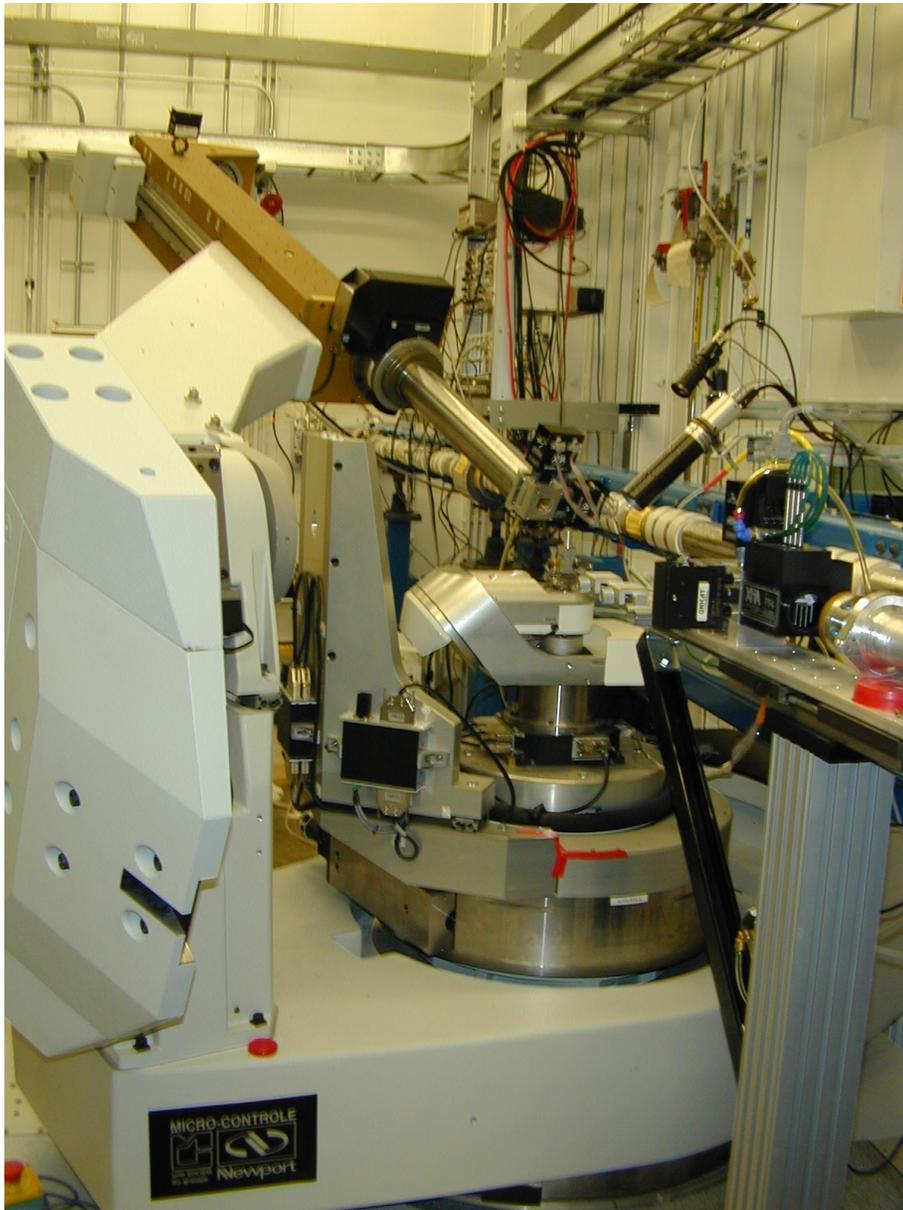
Progress of Phase Retrieval

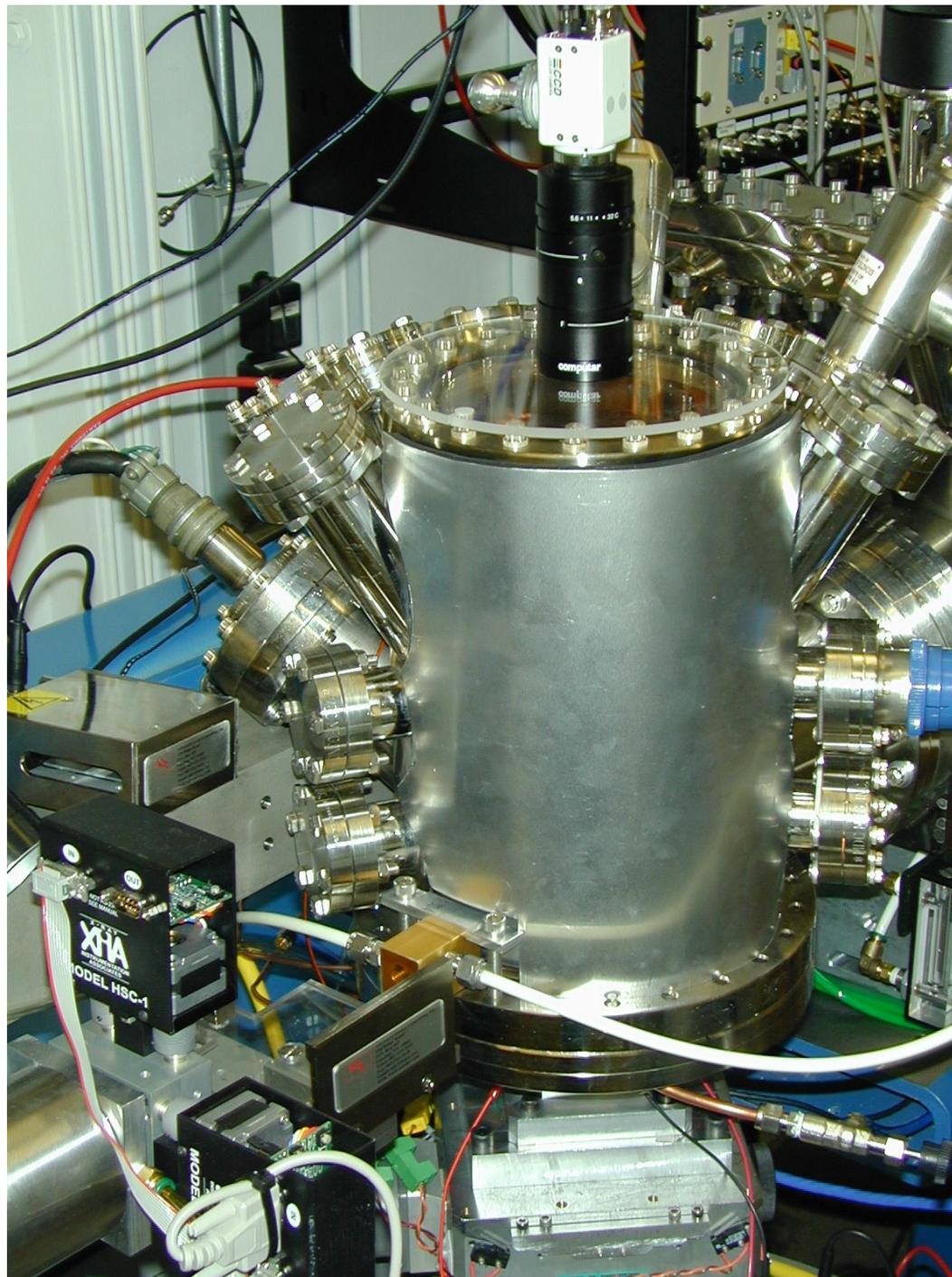


Study of Convergence with Noise

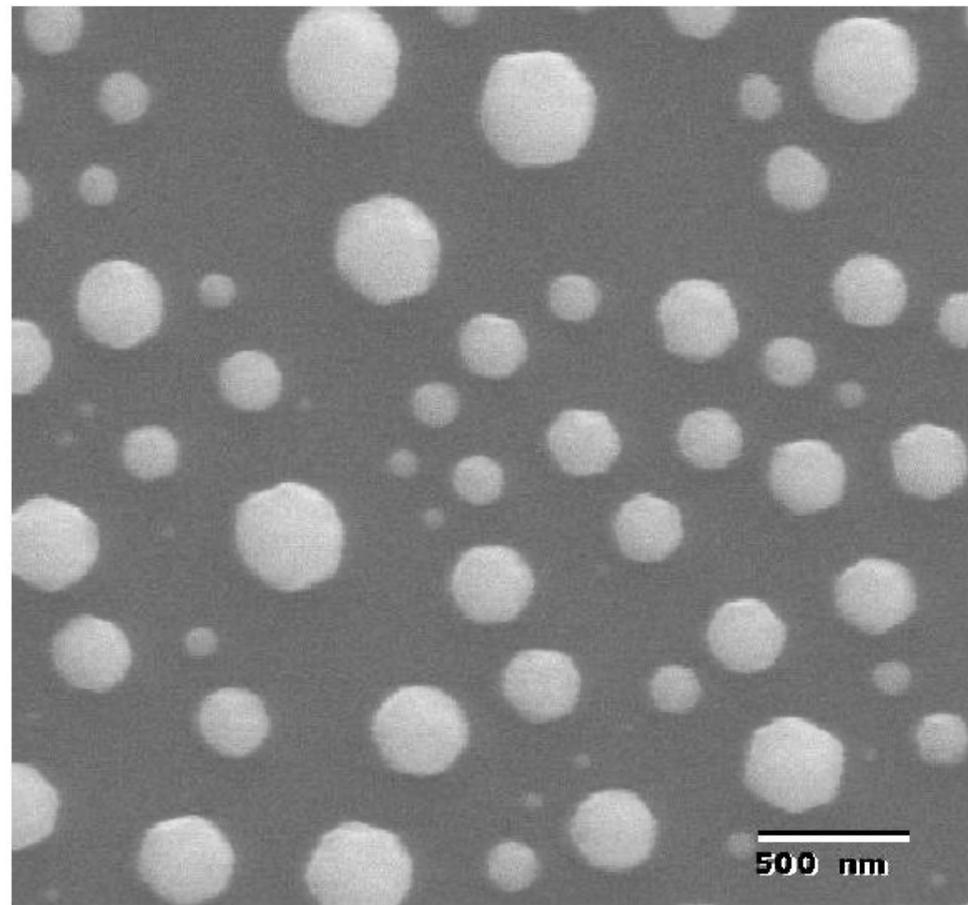
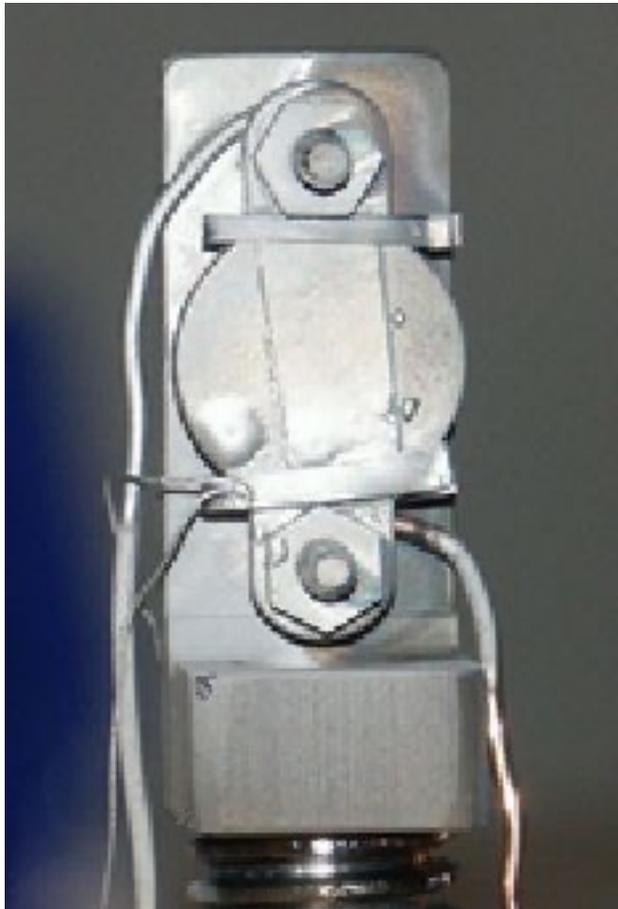
Garth Williams



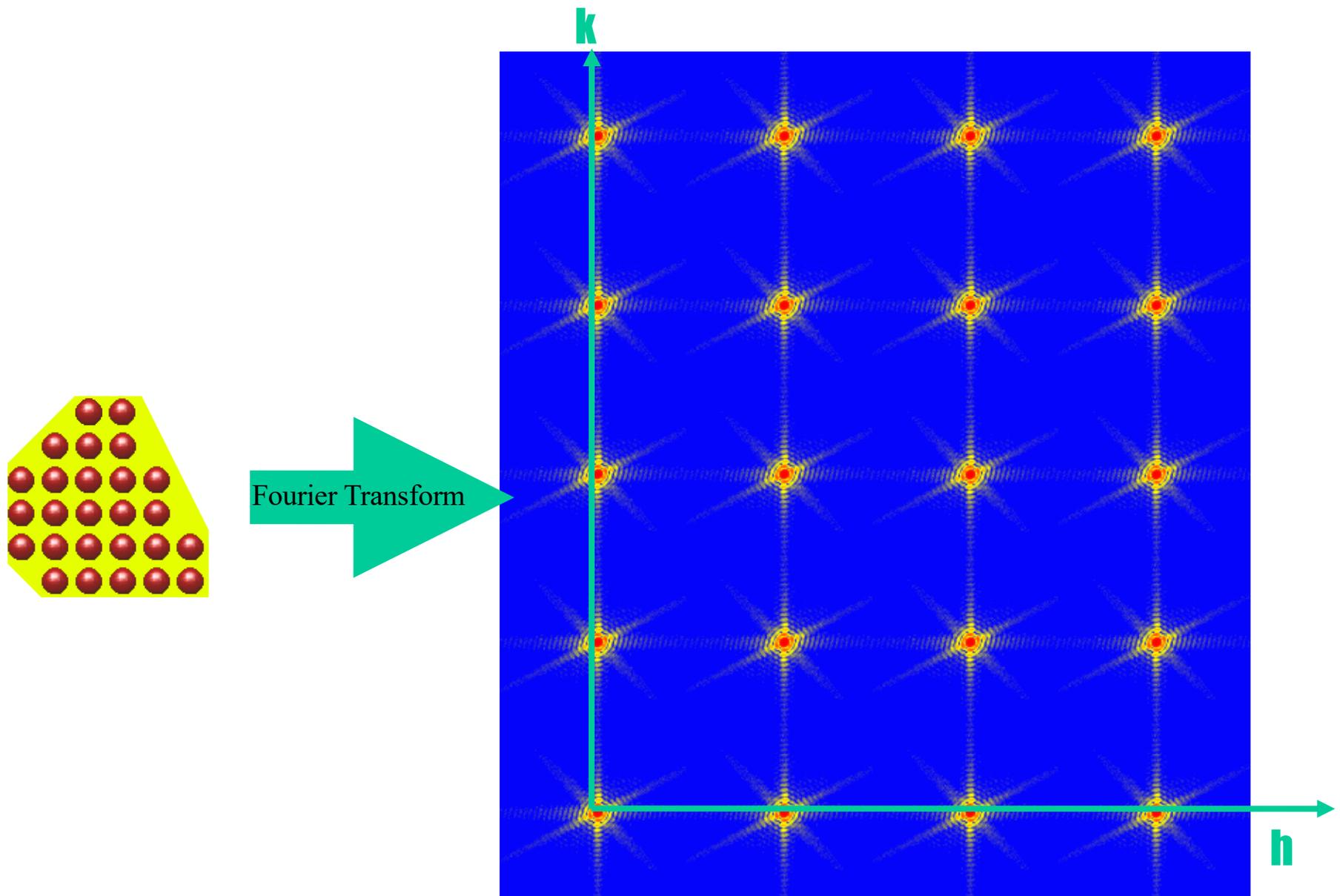




In situ growth of Pb crystals



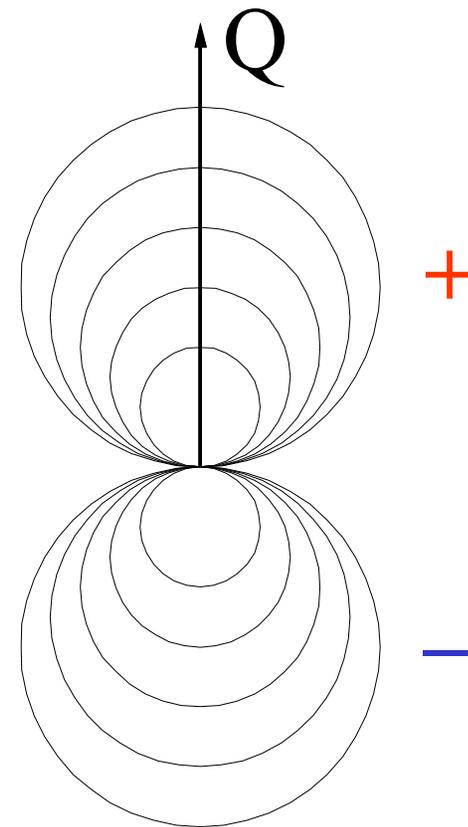
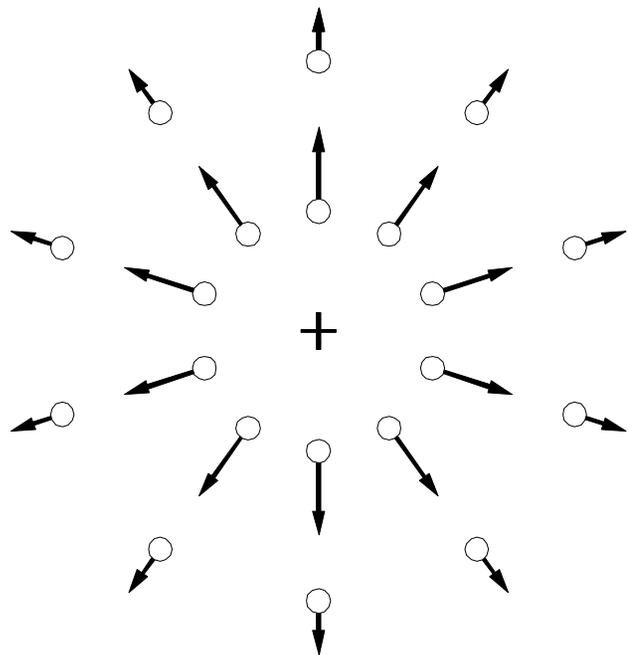
Coherent Diffraction from Crystals



Diffraction by Strain of Point Defect

$$A \sim \sum e^{i\mathbf{Q}\cdot(\mathbf{R}_j+\mathbf{u}_j)}$$
$$\approx \sum e^{i\mathbf{Q}\cdot\mathbf{R}_j} (1+i\mathbf{Q}\cdot\mathbf{u}_j)$$

Imaginary density



Good statistics, 3D diffraction data

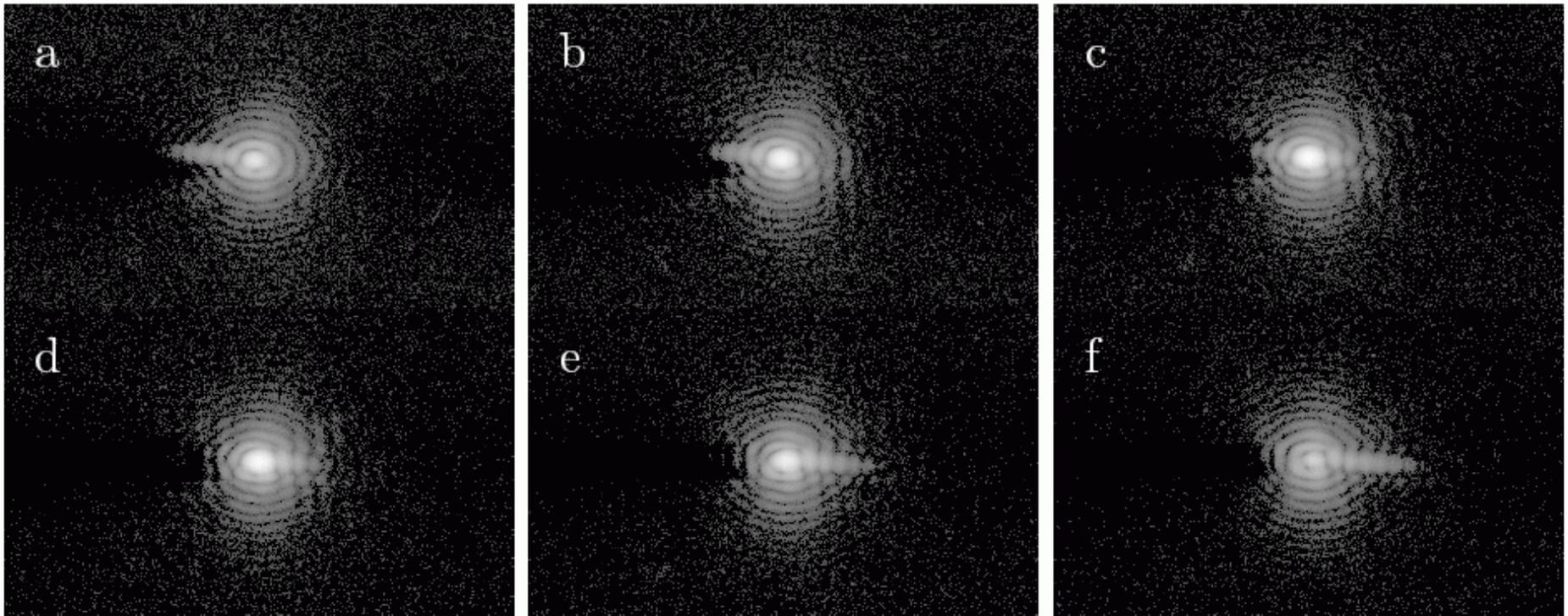
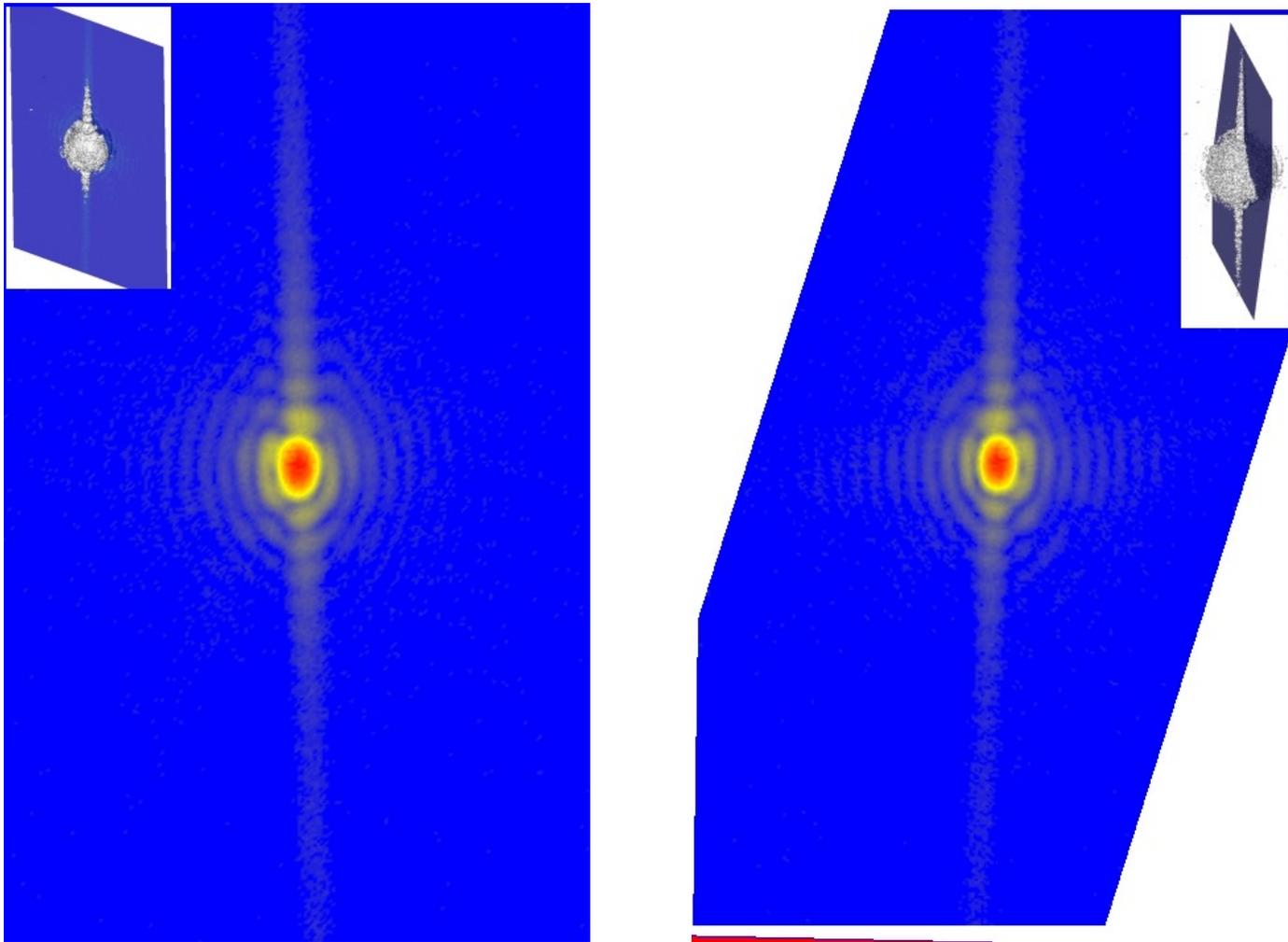
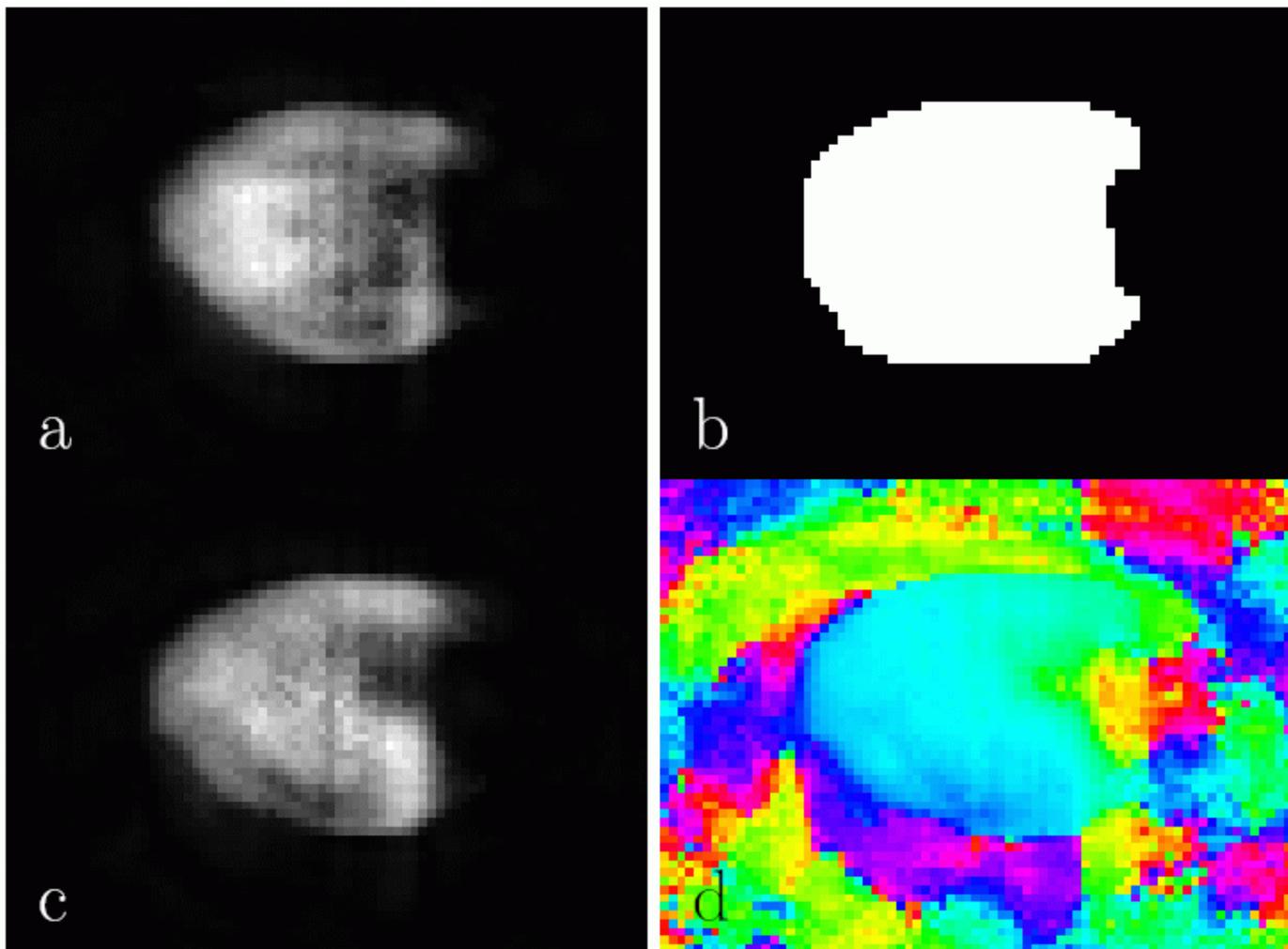


Figure 4.12: Center slices from 3D CXD pattern from Pb sample, on a log scale. Data file 296 from 10/03.

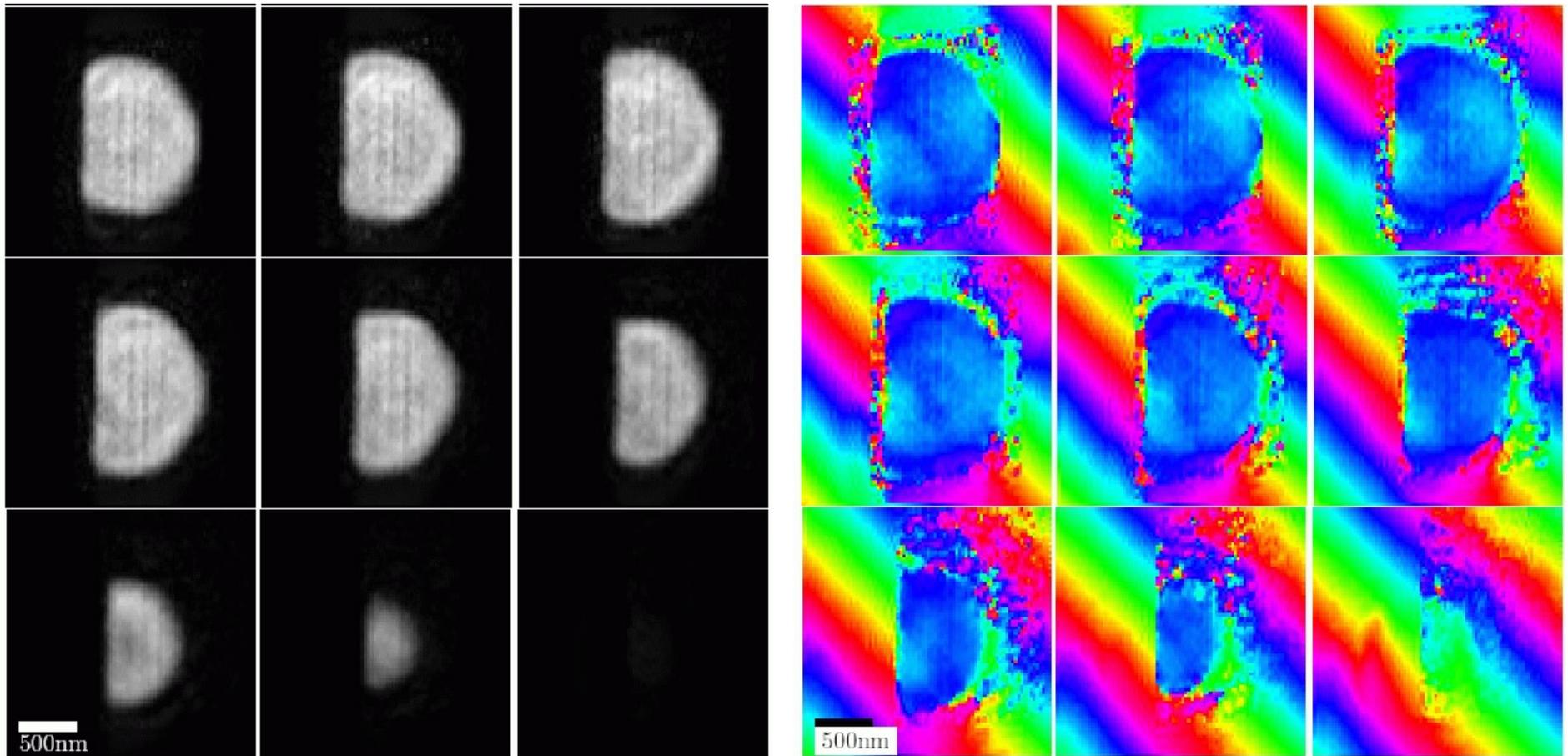
3D data along special directions

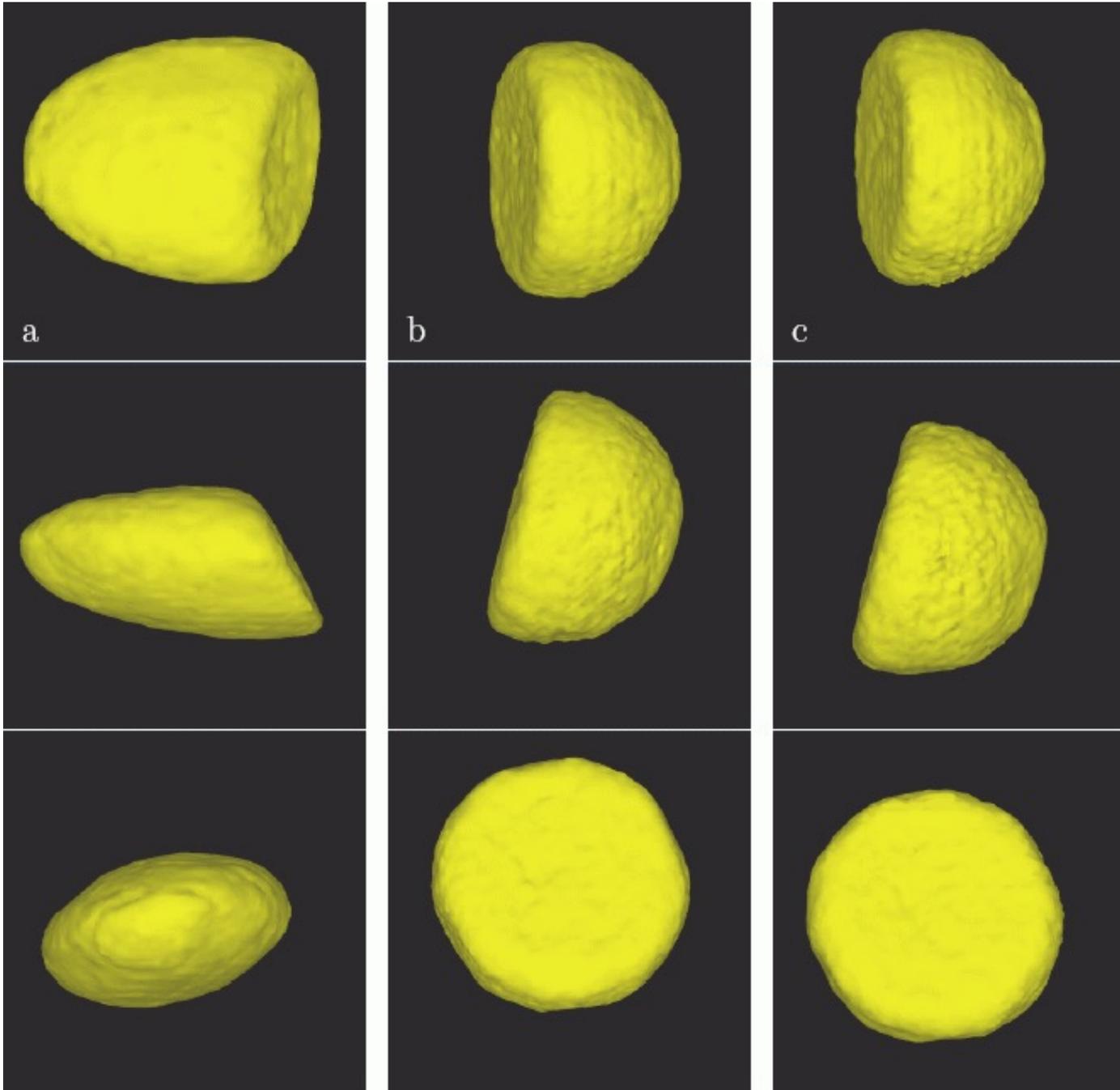


Learn shape of “tight” support

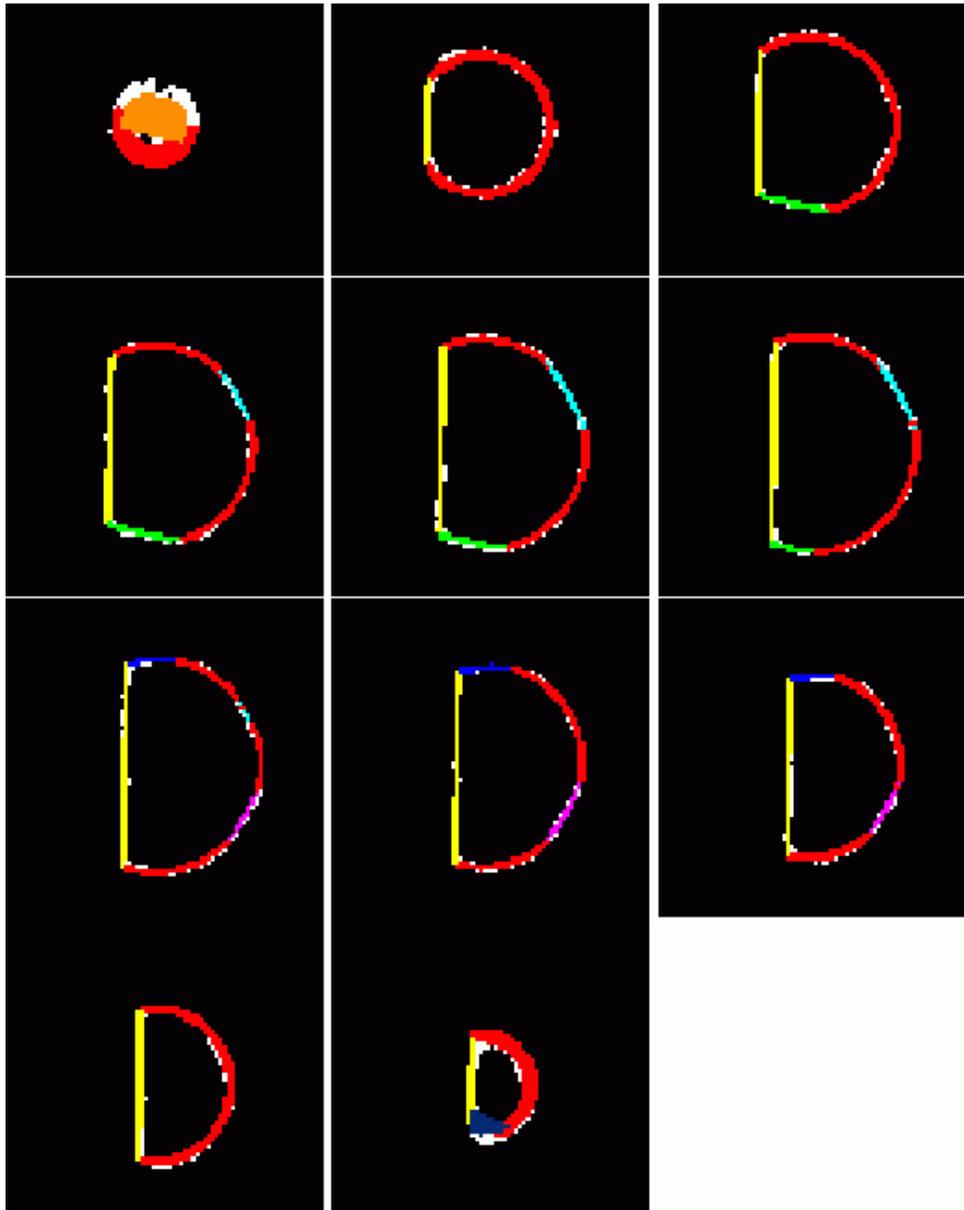


Then refine amplitude *and* phase



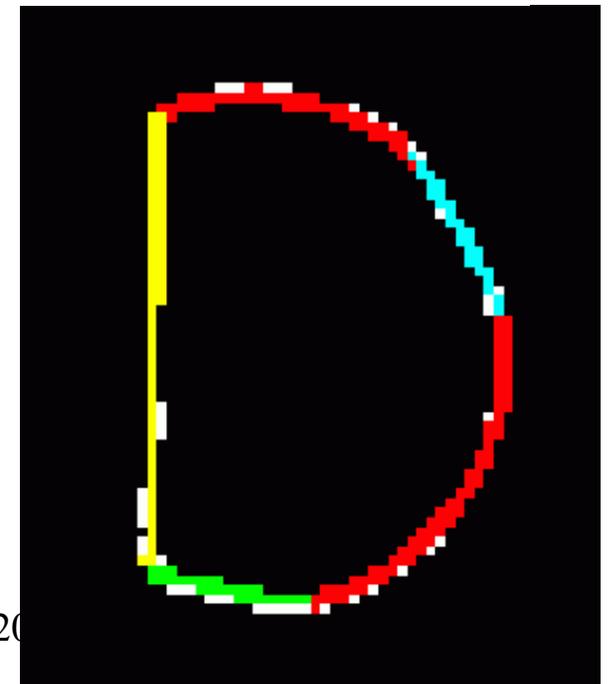


Fitting to faceted shape

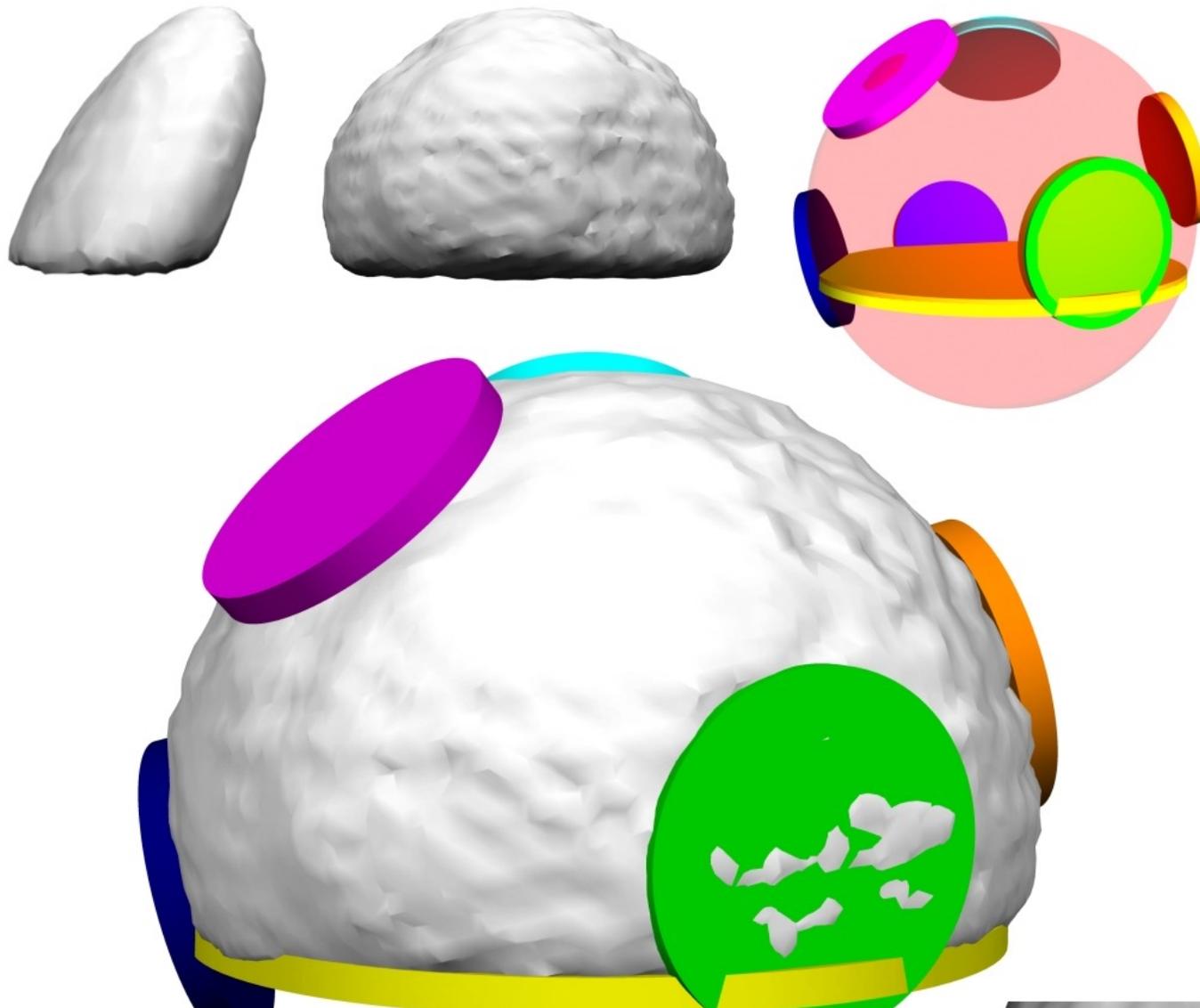


	$ R $		P0	P1	P2	P3	P4	P5	P6
	9.4	P0	0	85	149	79	134	106	71
	25.7	P1		0	123	164	83	76	102
	25.1	P2			0	72	67	74	110
	25.9	P3				0	111	106	76
	25.4	P4					0	113	68
	25.4	P5						0	176
	26.0	P6							0 0

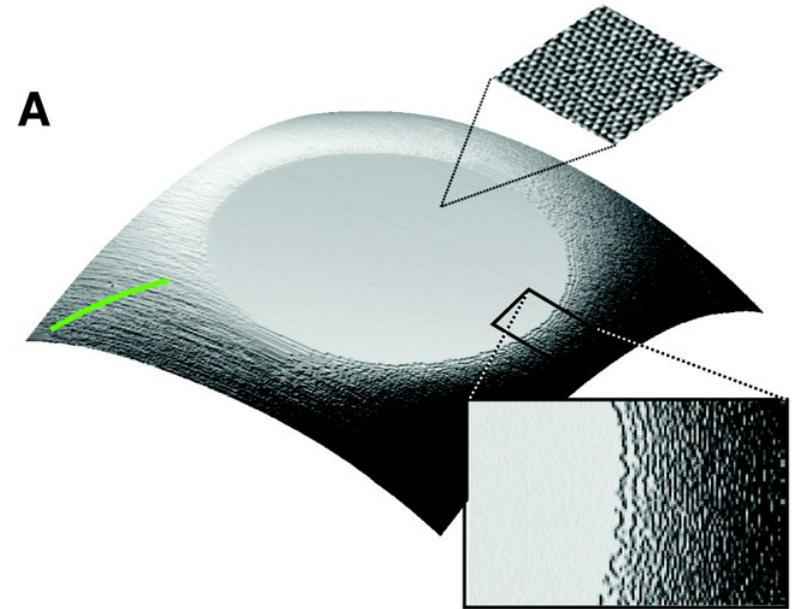
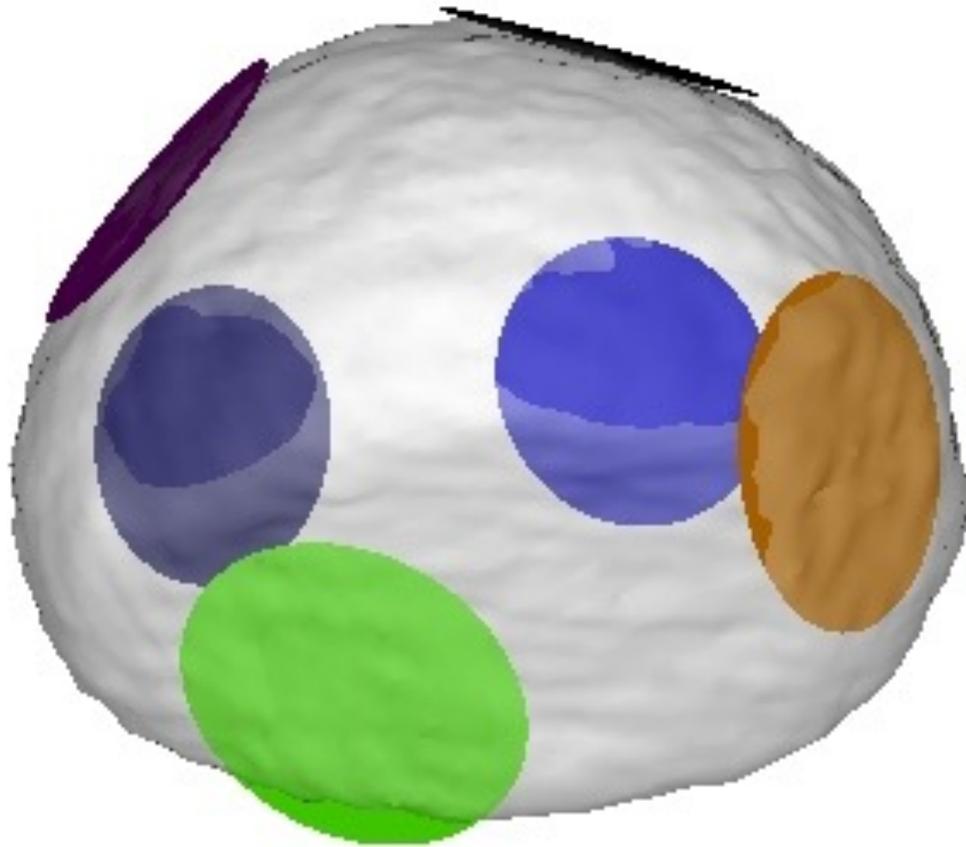
Angles between facets



Fitting of crystal shape

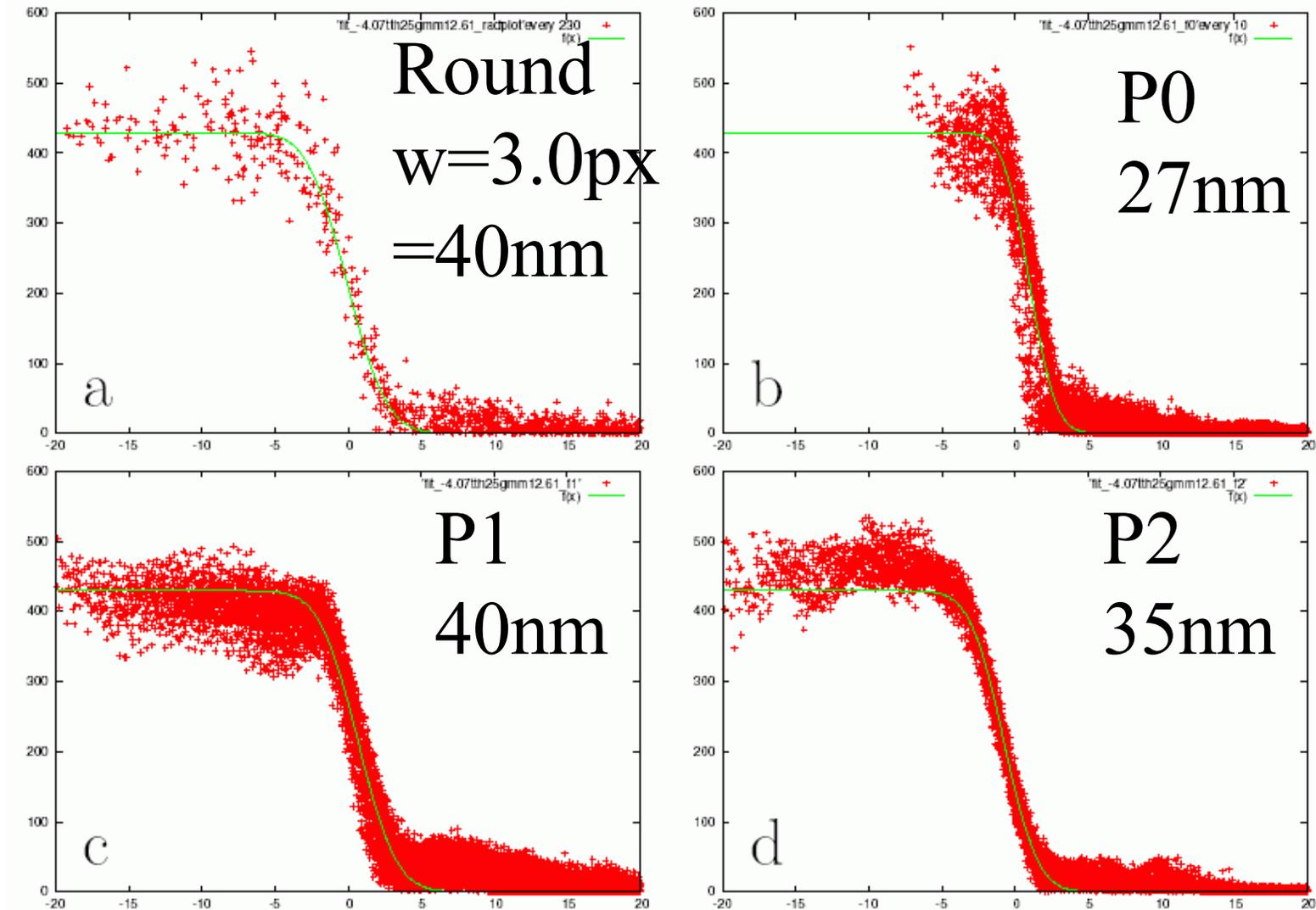


Facets of Equilibrium Crystal Shape

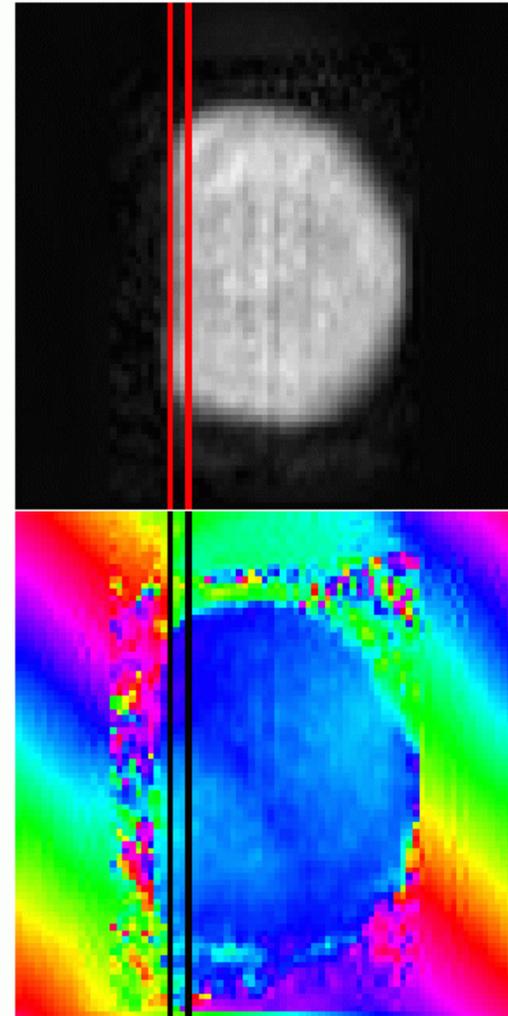
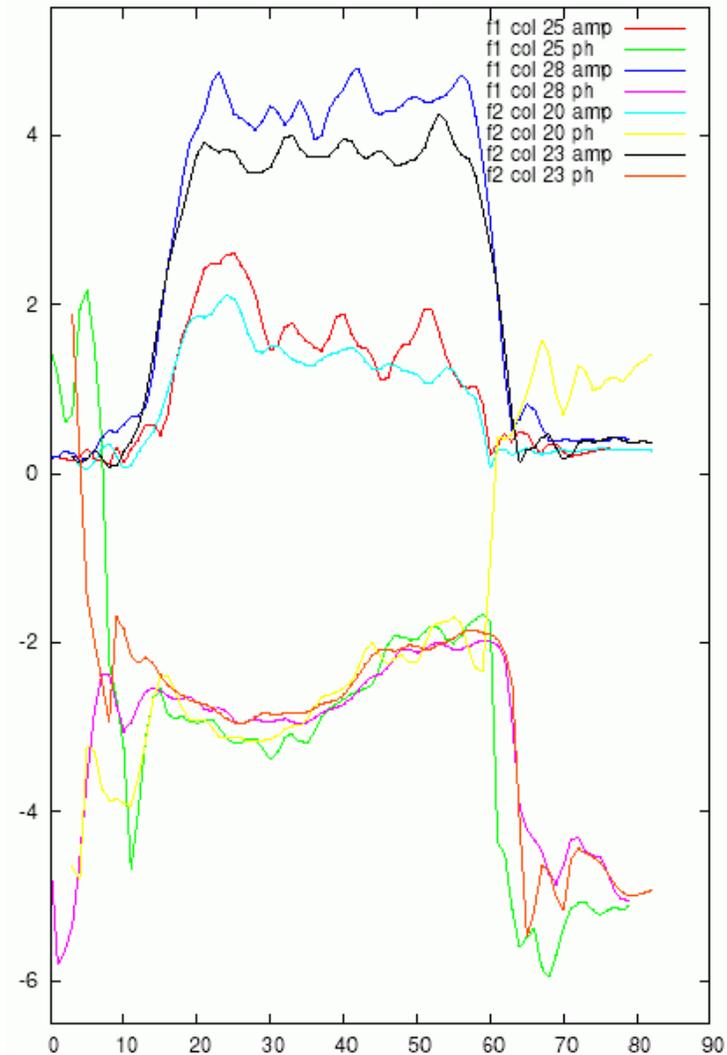


Thurmer K, Williams E, Reutt-Robey J
Science **297** 2033 (2002)

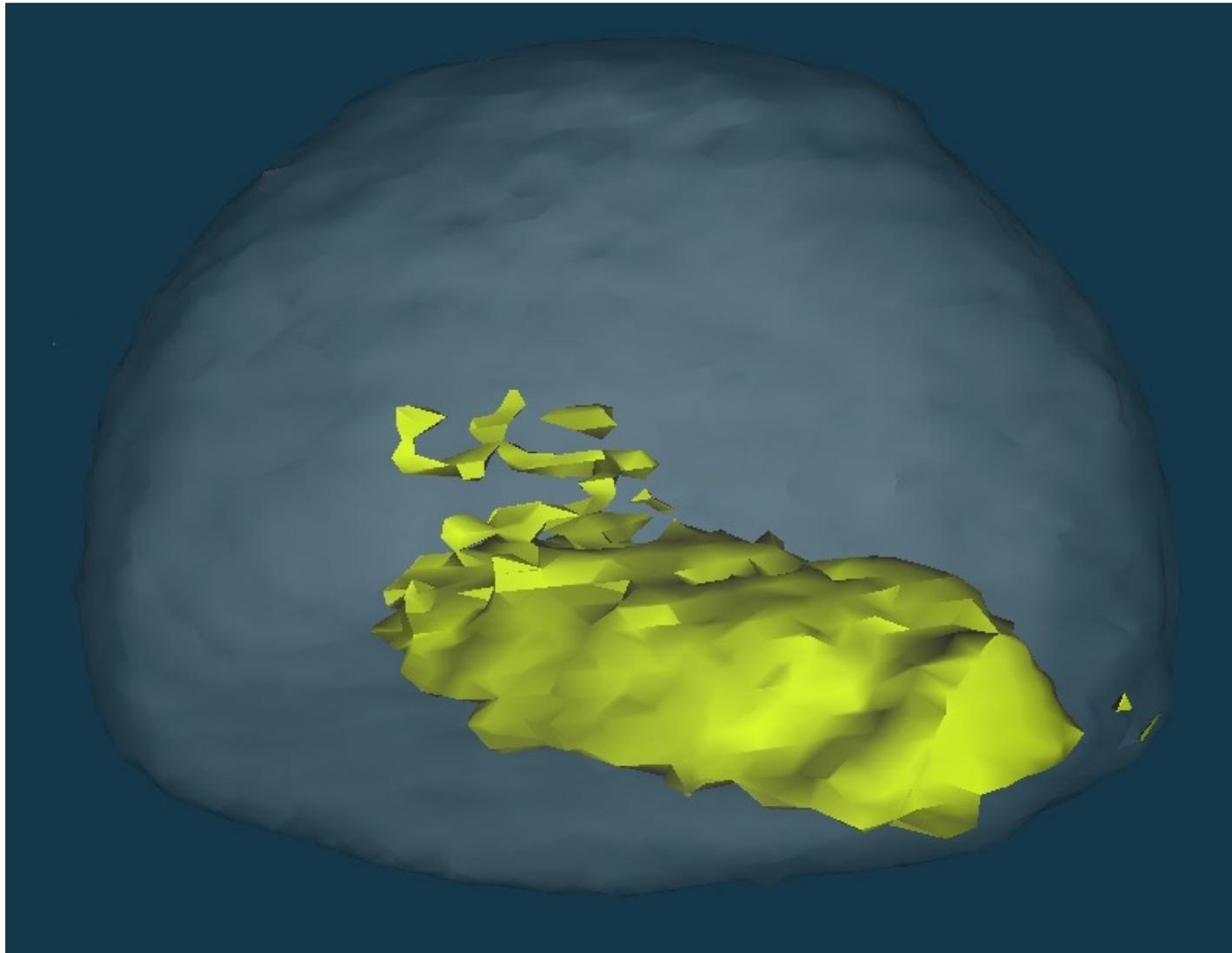
Density distribution across surface



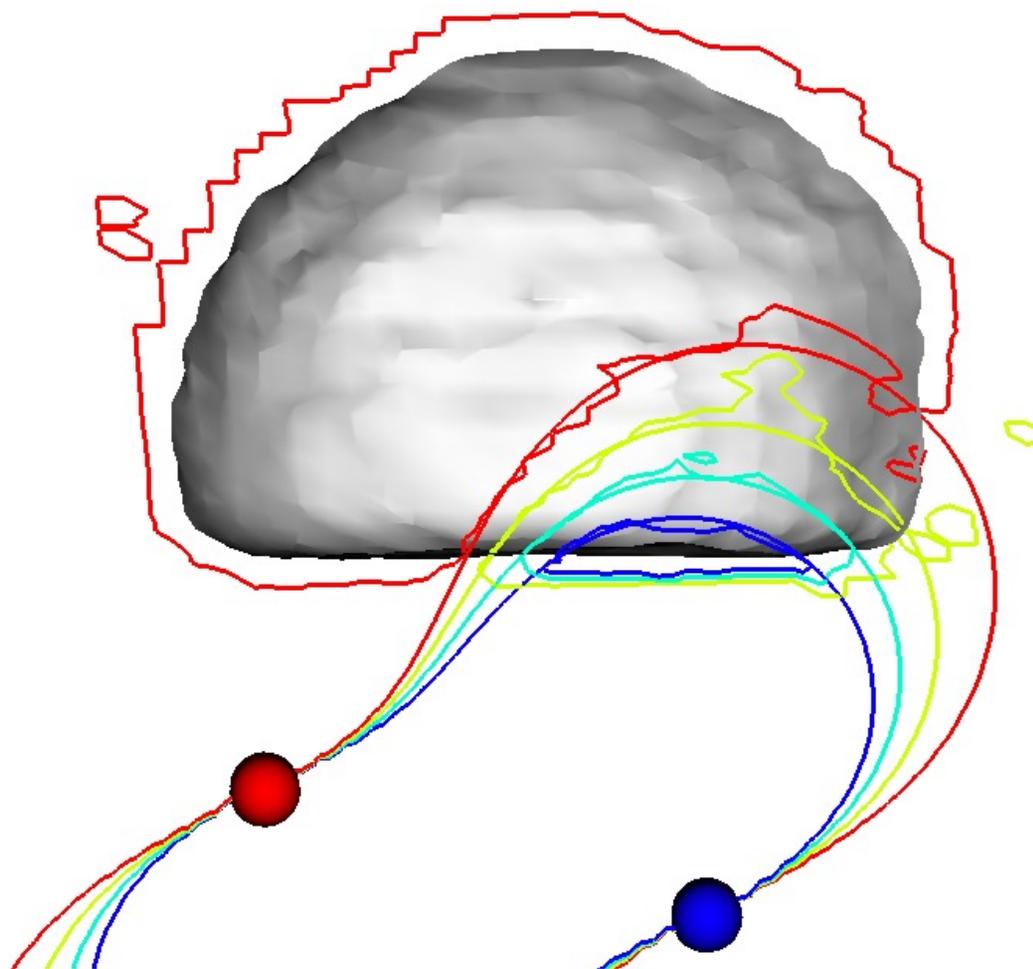
Phase structure near substrate interface



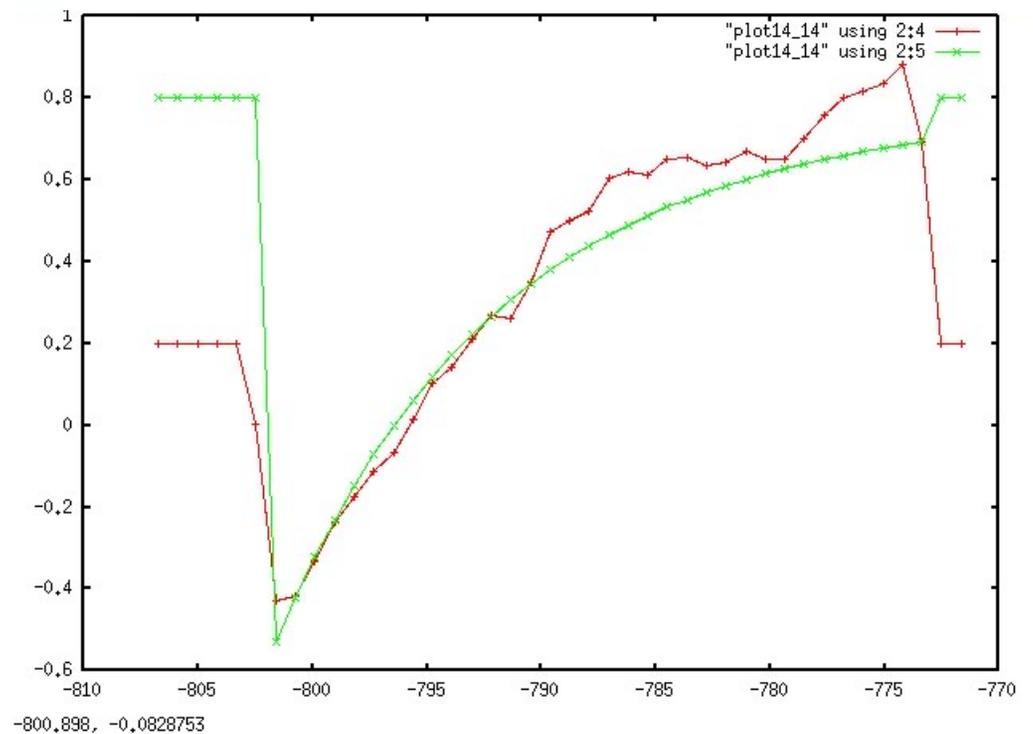
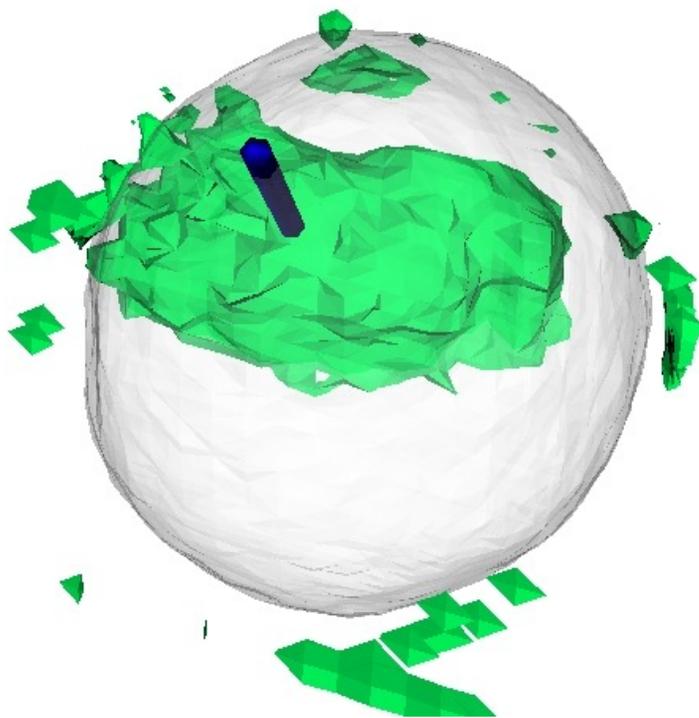
Modeling of 3D Phase Bump

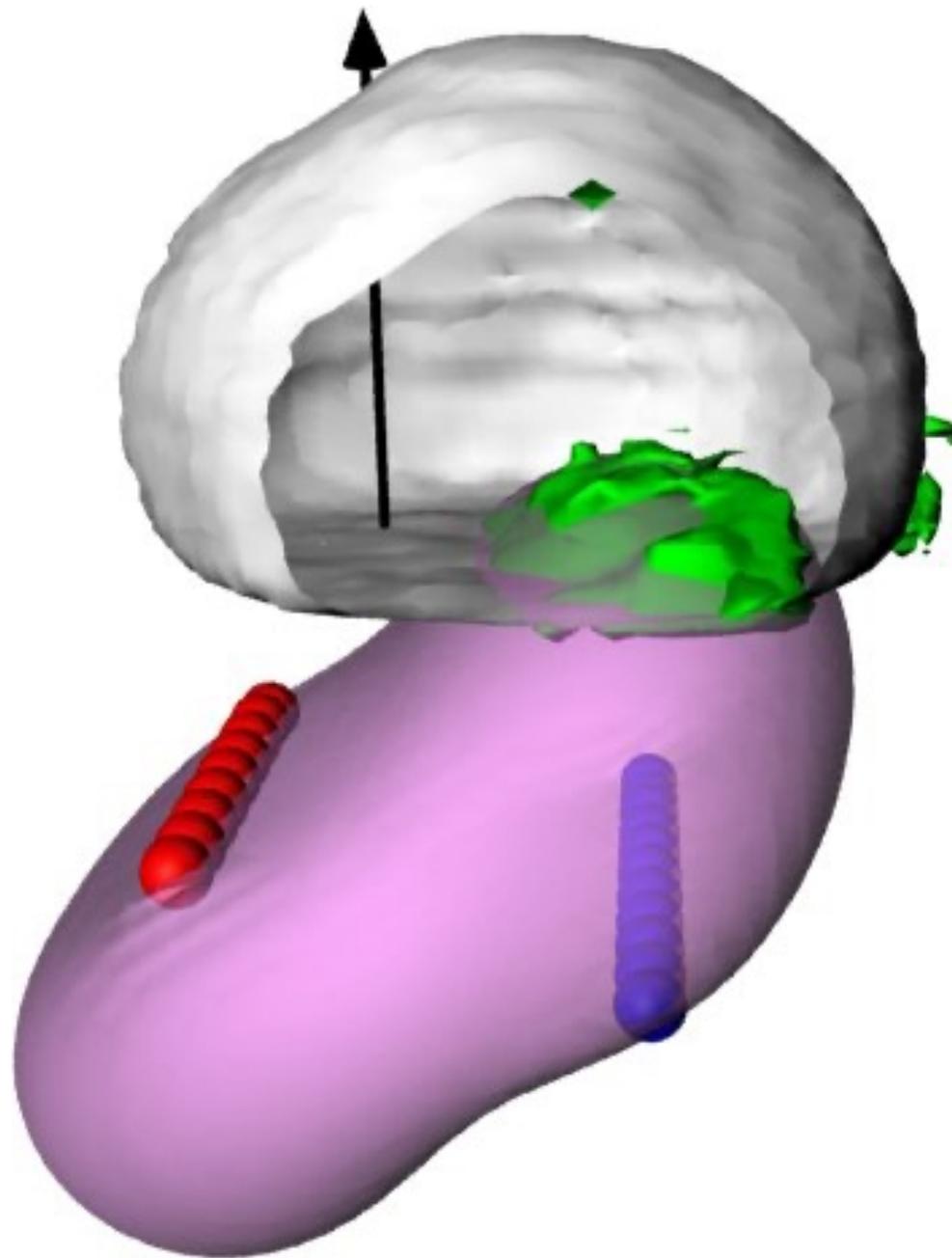


Field lines of Point Charges



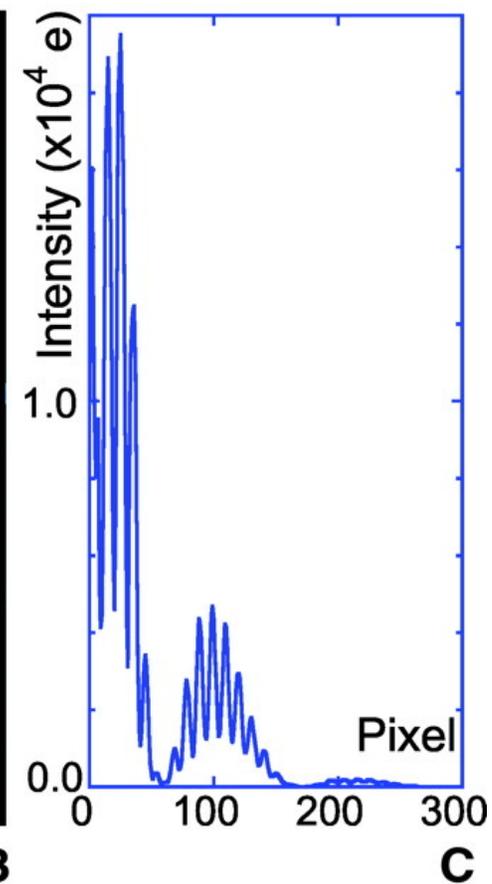
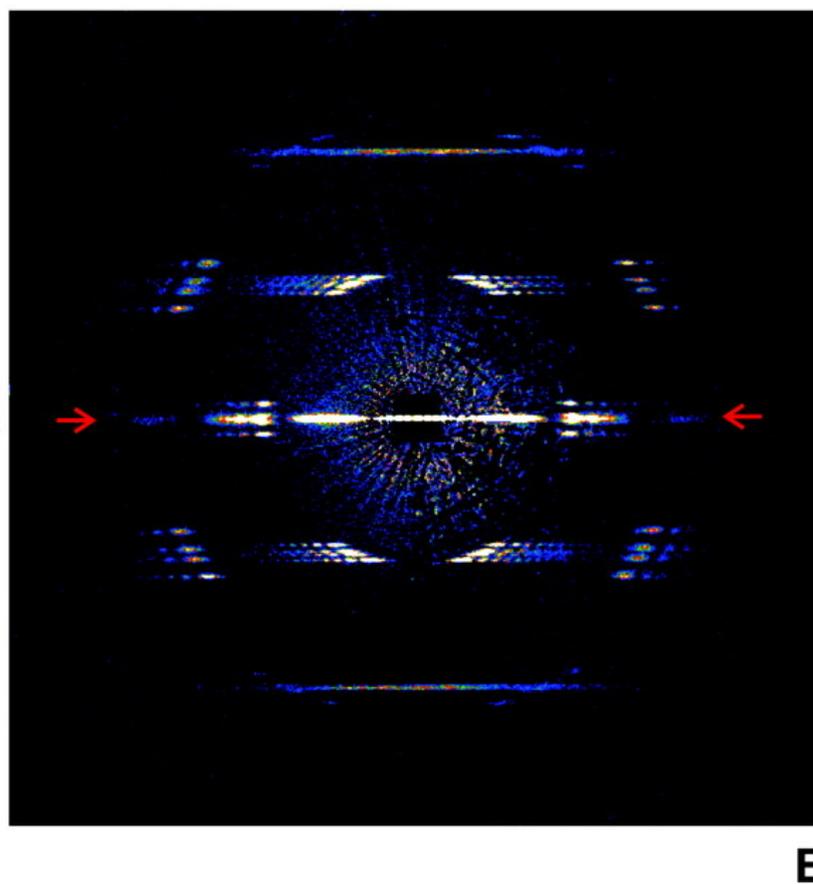
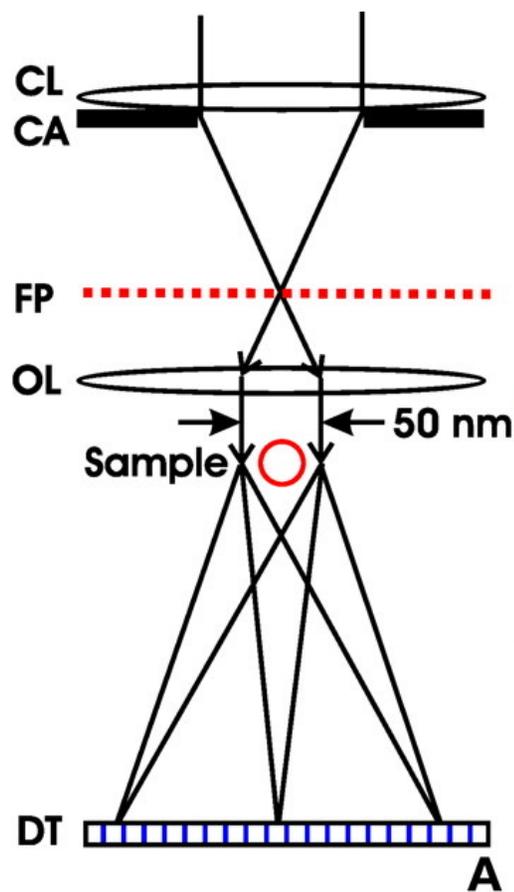
Line scan through phase





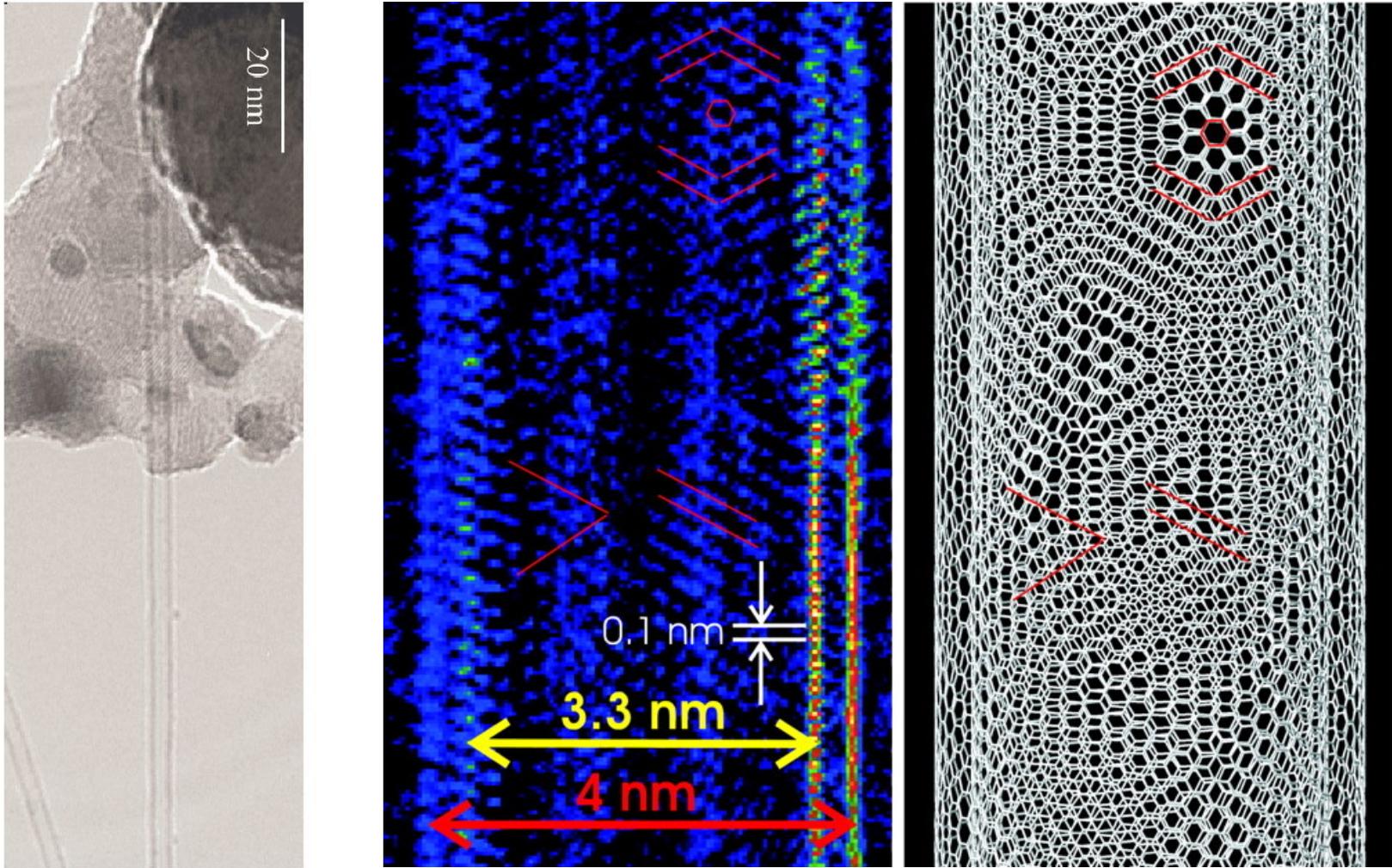
Electron Diffraction from C-nanotube

Jim Zuo et. al. Science 300 1419 (2003)

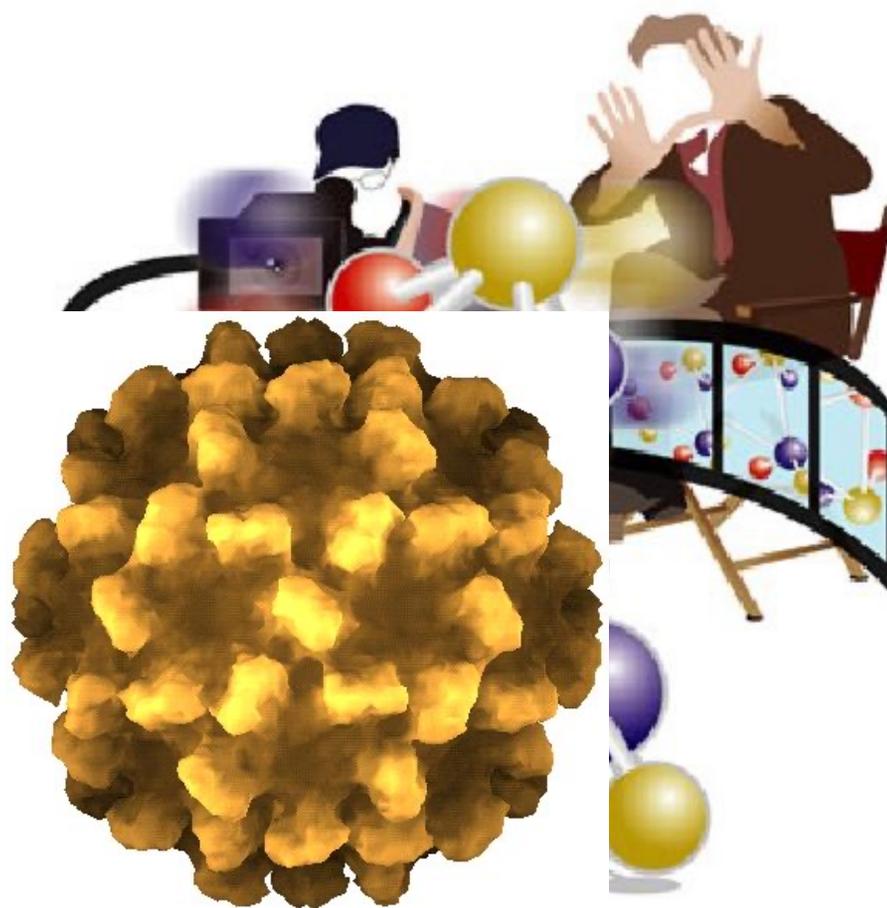


Images of DW C-nanotube

Jim Zuo et. al. Science 300 1419 (2003)



Molecular Movies using XFEL



Conclusions and Outlook

- Internal structure of Au Nanocrystals
- 3D imaging practical for nanocrystals
- Phasing by computation instead of lens
- Strain fields imaged from asymmetric patterns
- Atomic resolution with electron diffraction
- Single molecules using XFEL