Interpretation of Complex Images in Coherent X-ray Diffraction

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NSLS-2 Seminar
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Outline

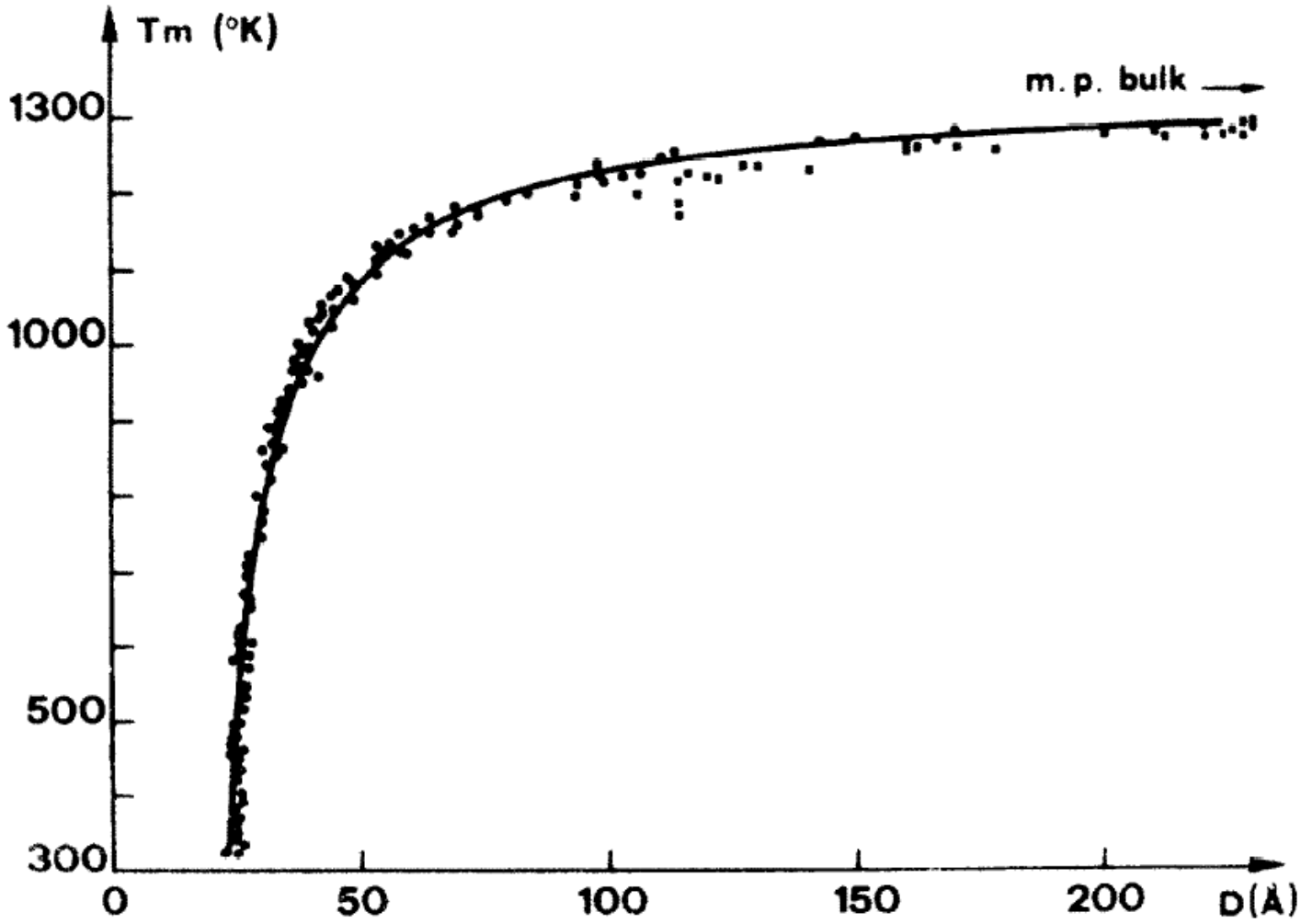
- Coherent X-ray Diffraction
- How to Solve the Phase Problem
- Nanocrystal Shapes
- Extension to Phase Objects
- New Beamline Concepts
- Opportunities with Electrons and FELs

Underlying Issues

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

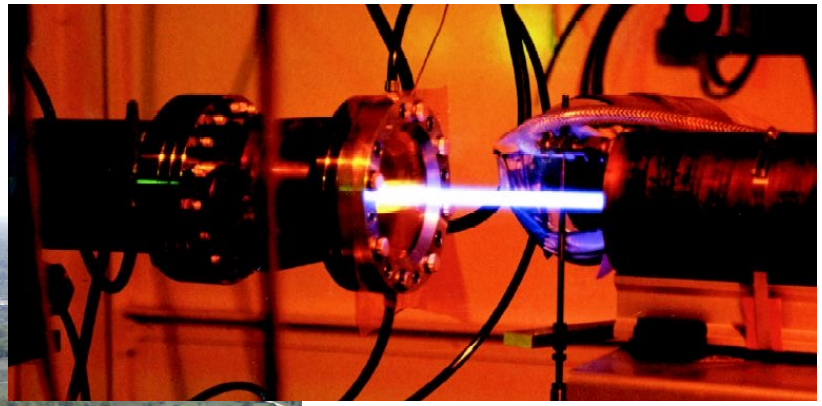
Size-dependent Melting of Au Particles

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



Synchrotron Radiation

Urbana



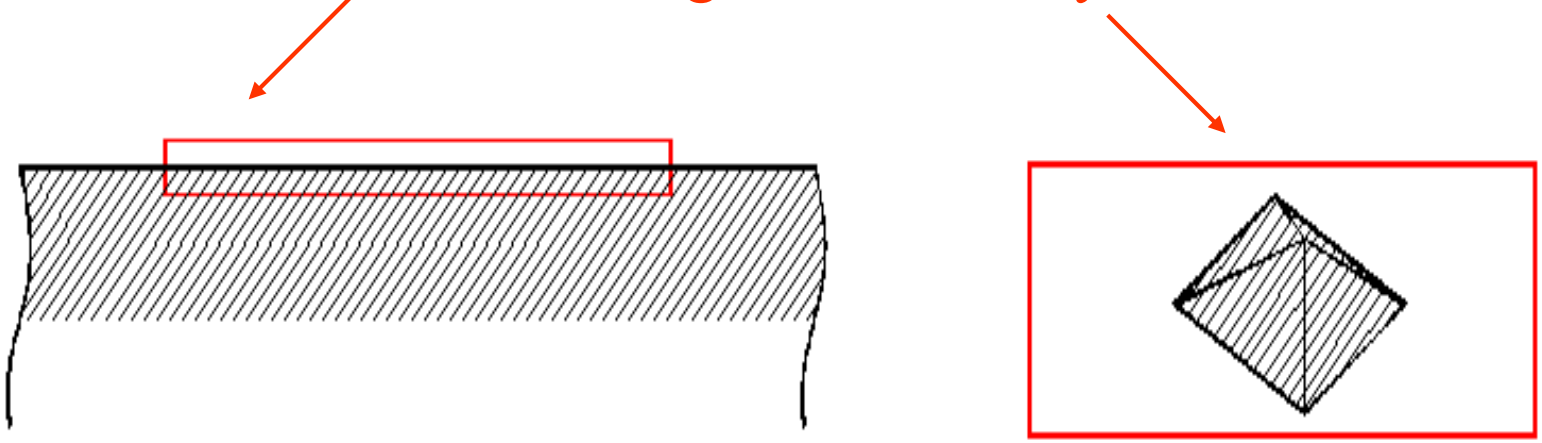
34-ID-C

Coherence at APS, ESRF or DLS

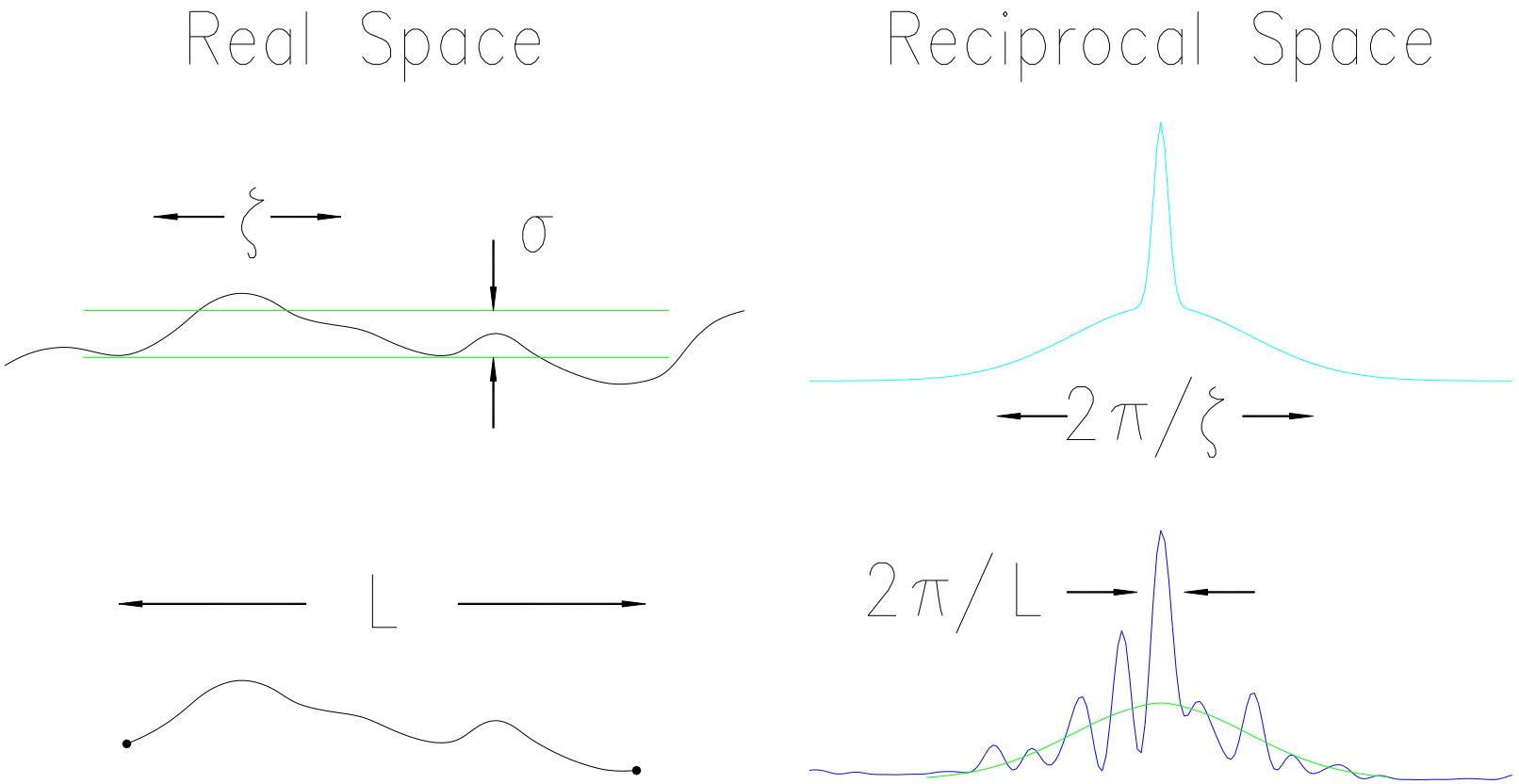
Typical of 3rd Generation (undulator) Synchrotron Source

Coherence of	ξ_{VER}	ξ_{HORIZ}	ξ_{LONG}	Flux
Raw Undulator	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}

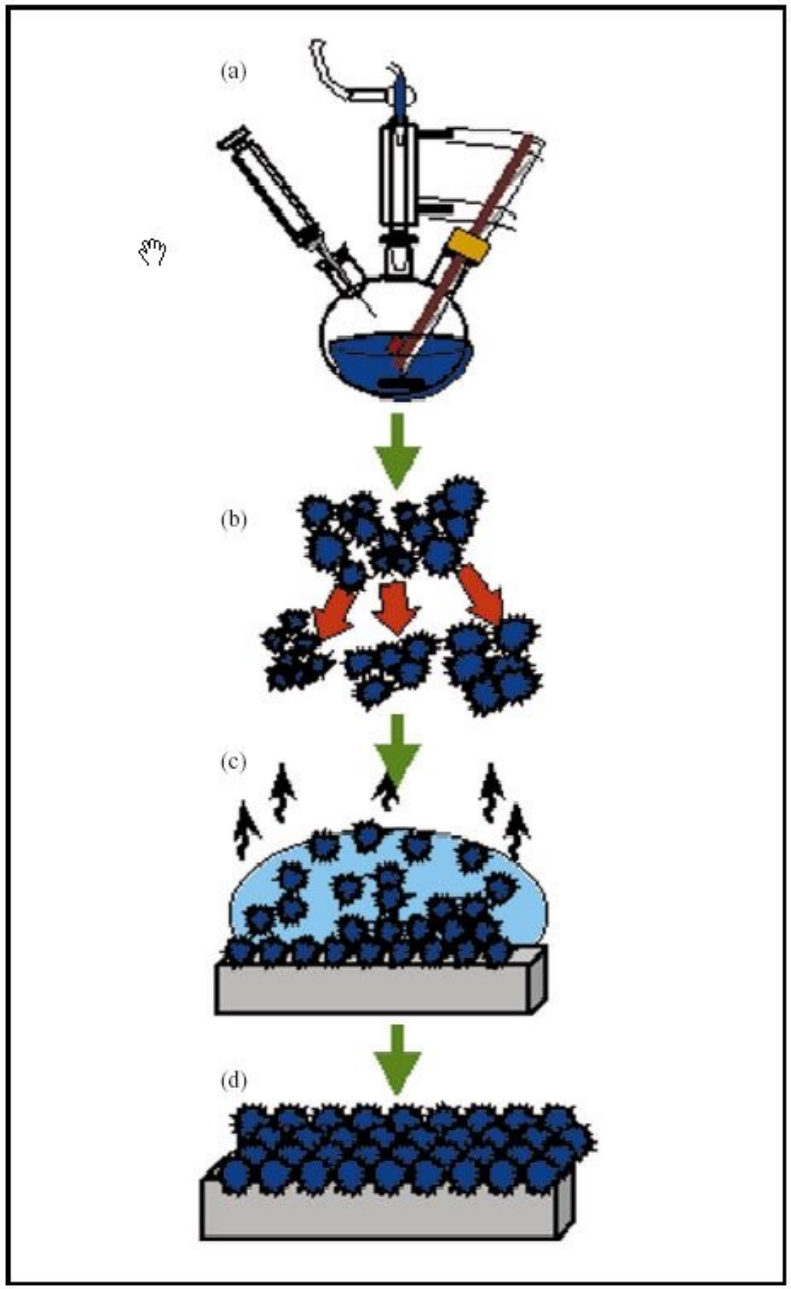
Coherent region defined by slits



Diffuse Scattering acquires Structure using CXD

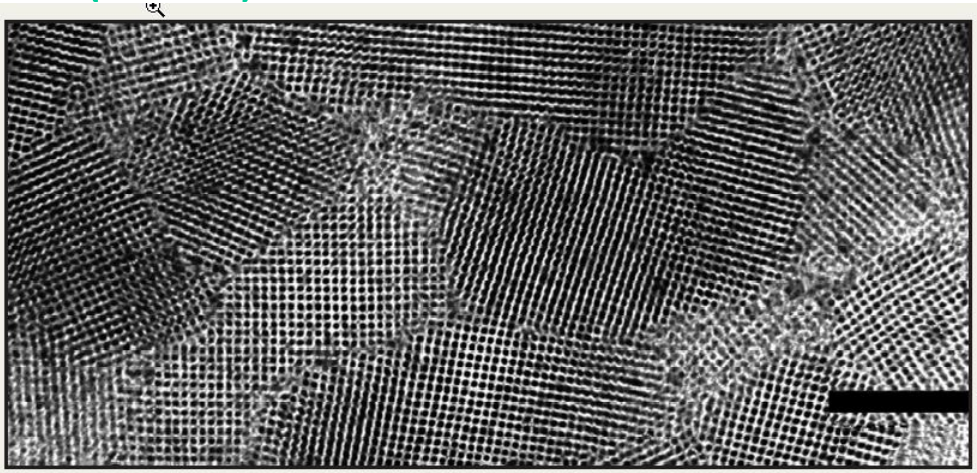


Chemical Synthesis of Nanocrystals



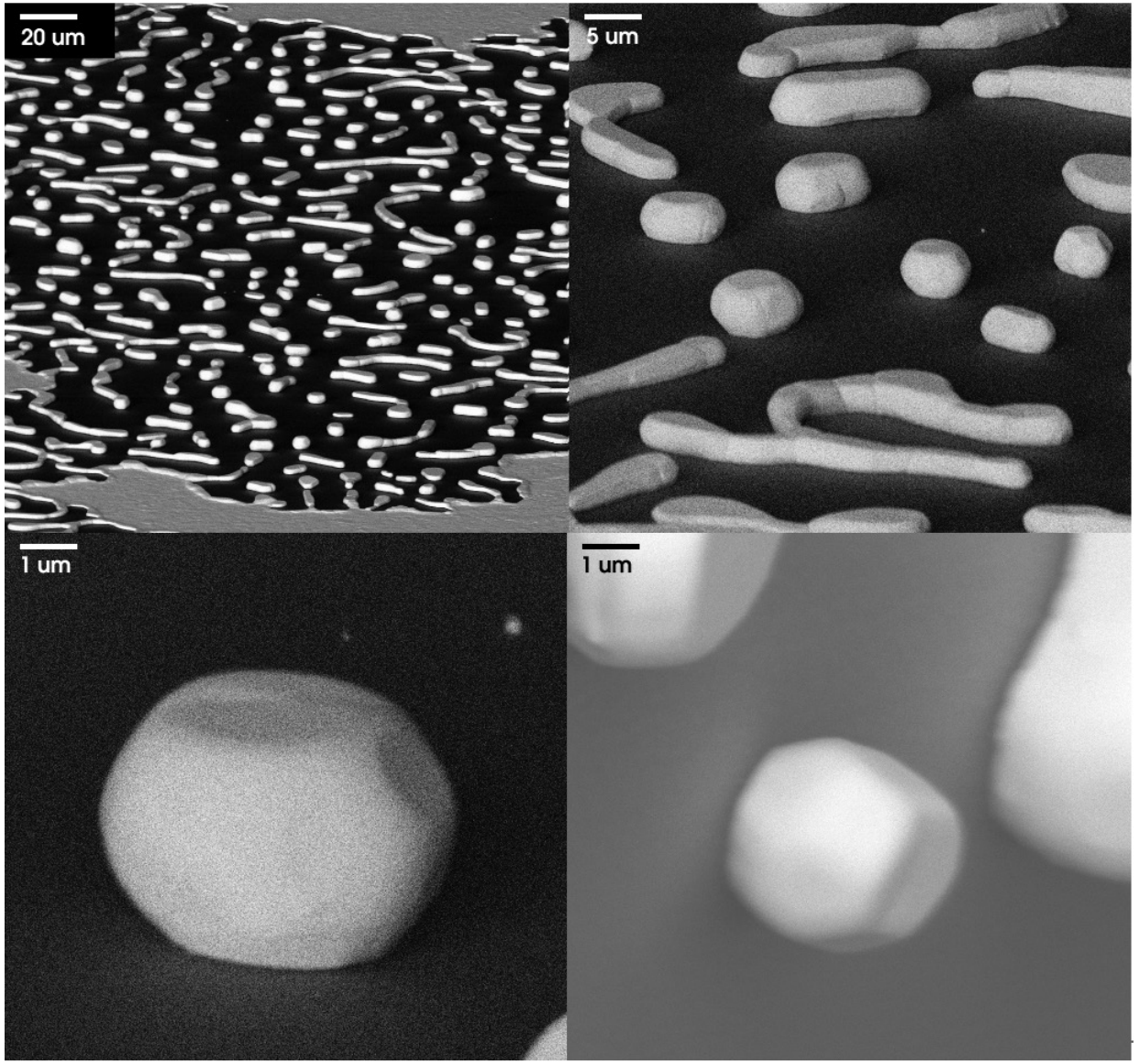
- Reactants introduced rapidly
- High temperature solvent
- Surfactant/organic capping agent
- Square superlattice (200nm scale)

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

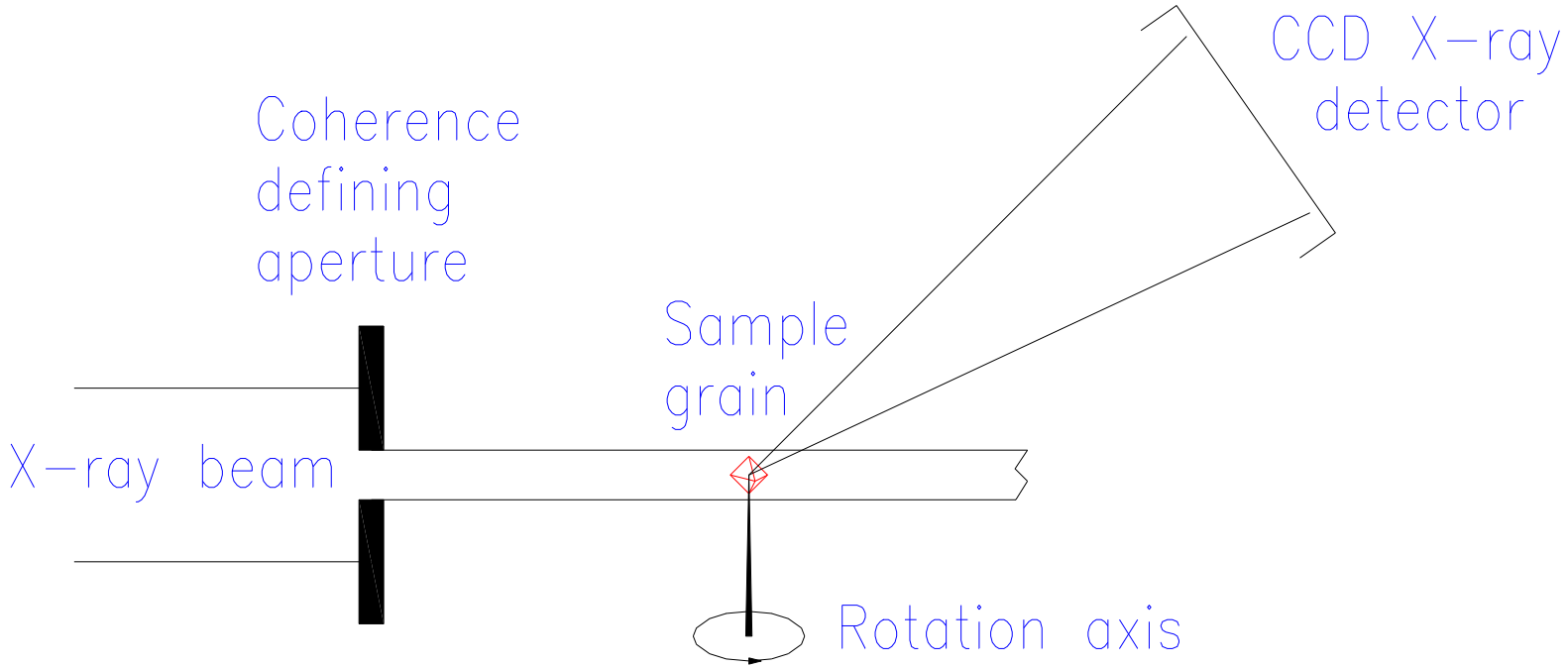


SEMS

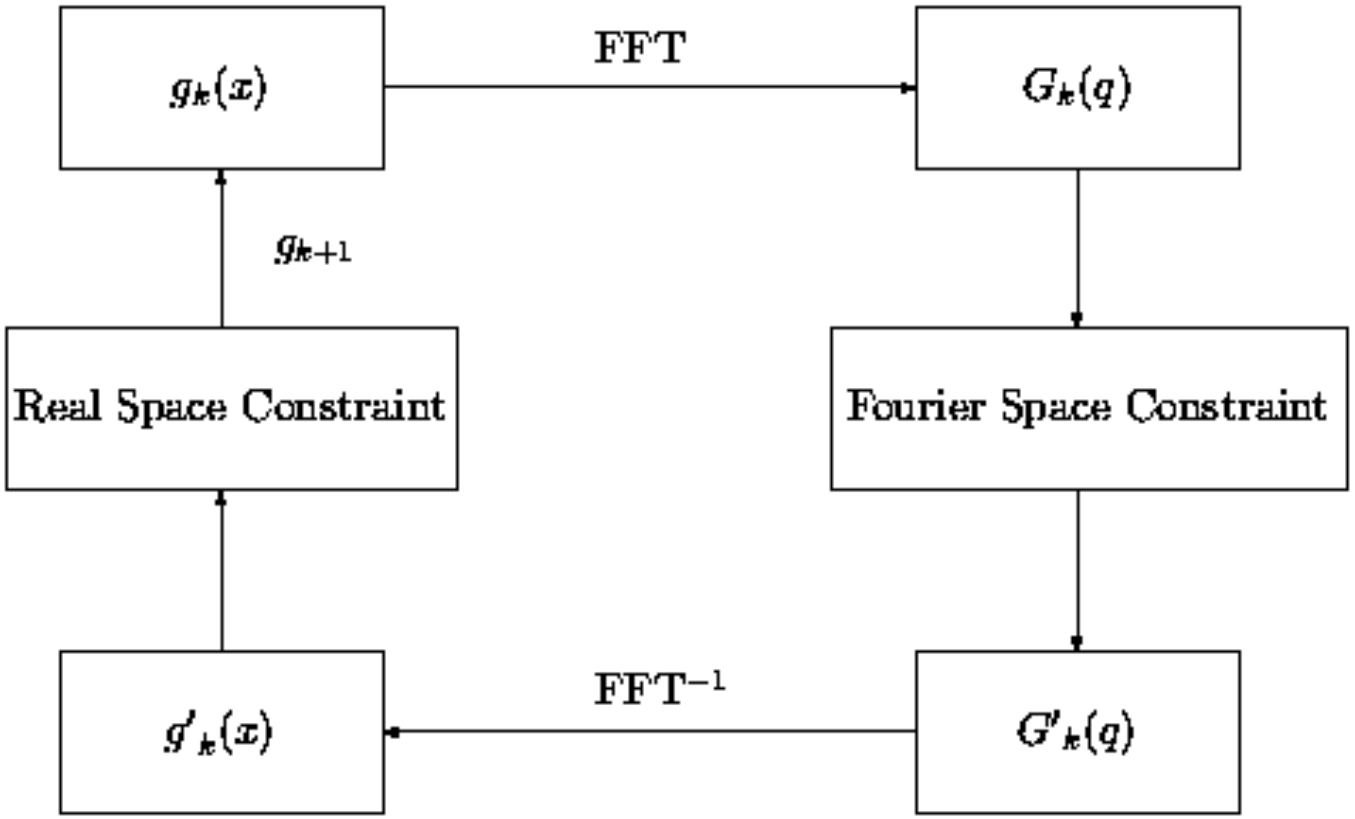
- Au blanket film
- Quartz substrate
- Annealed at 950°C for 70 hrs.



Lensless X-ray Microscope



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' 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)

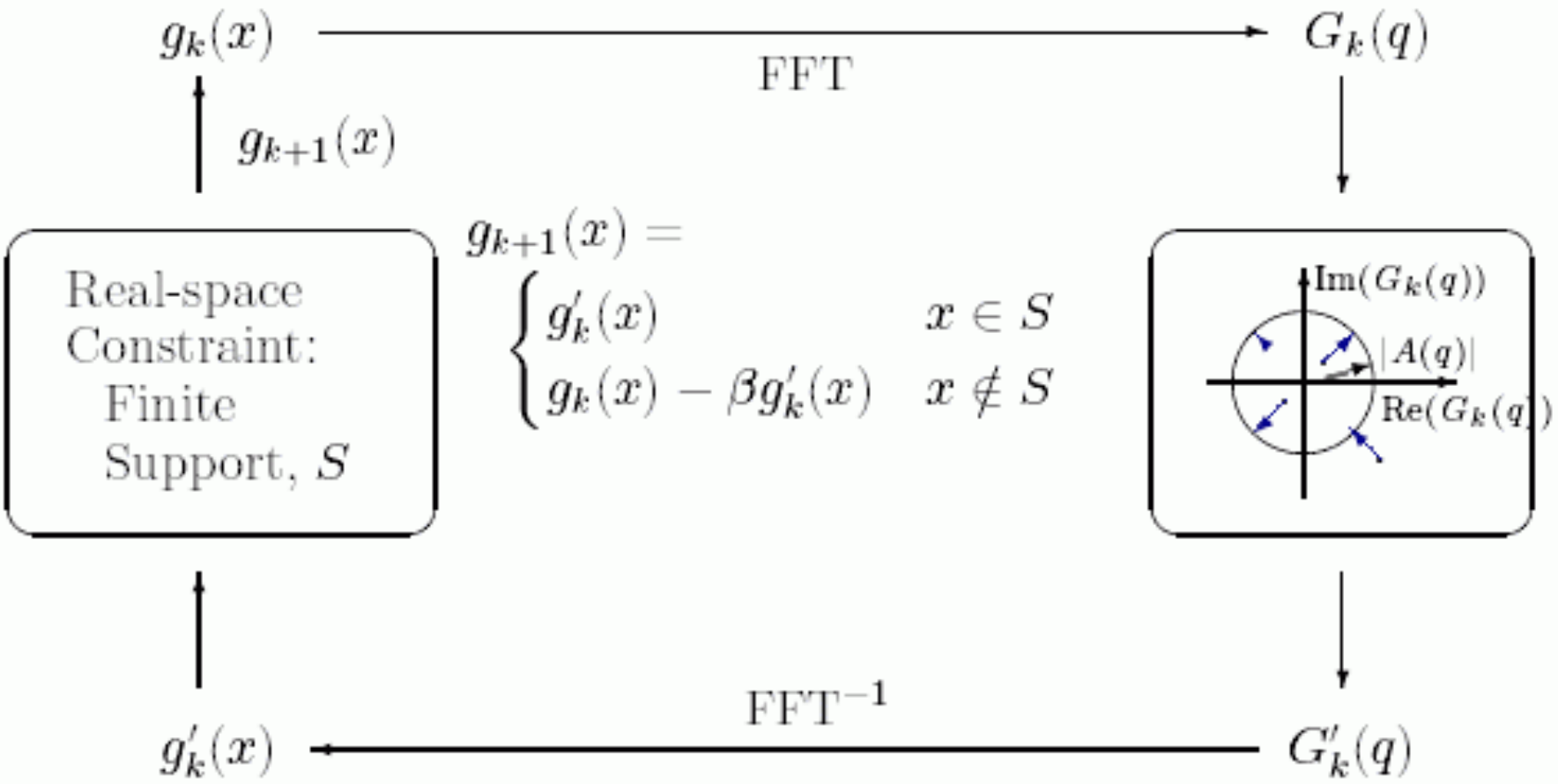
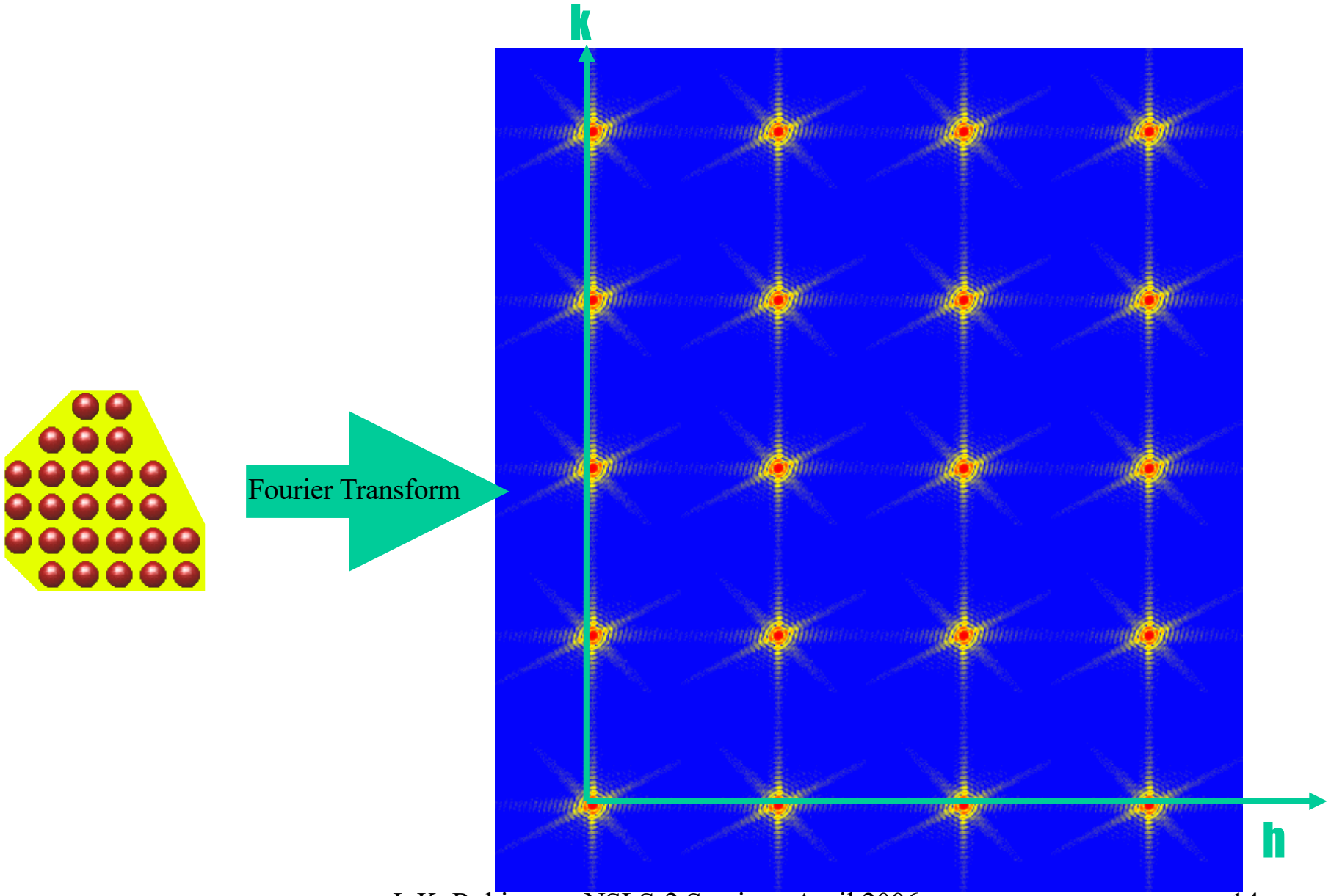
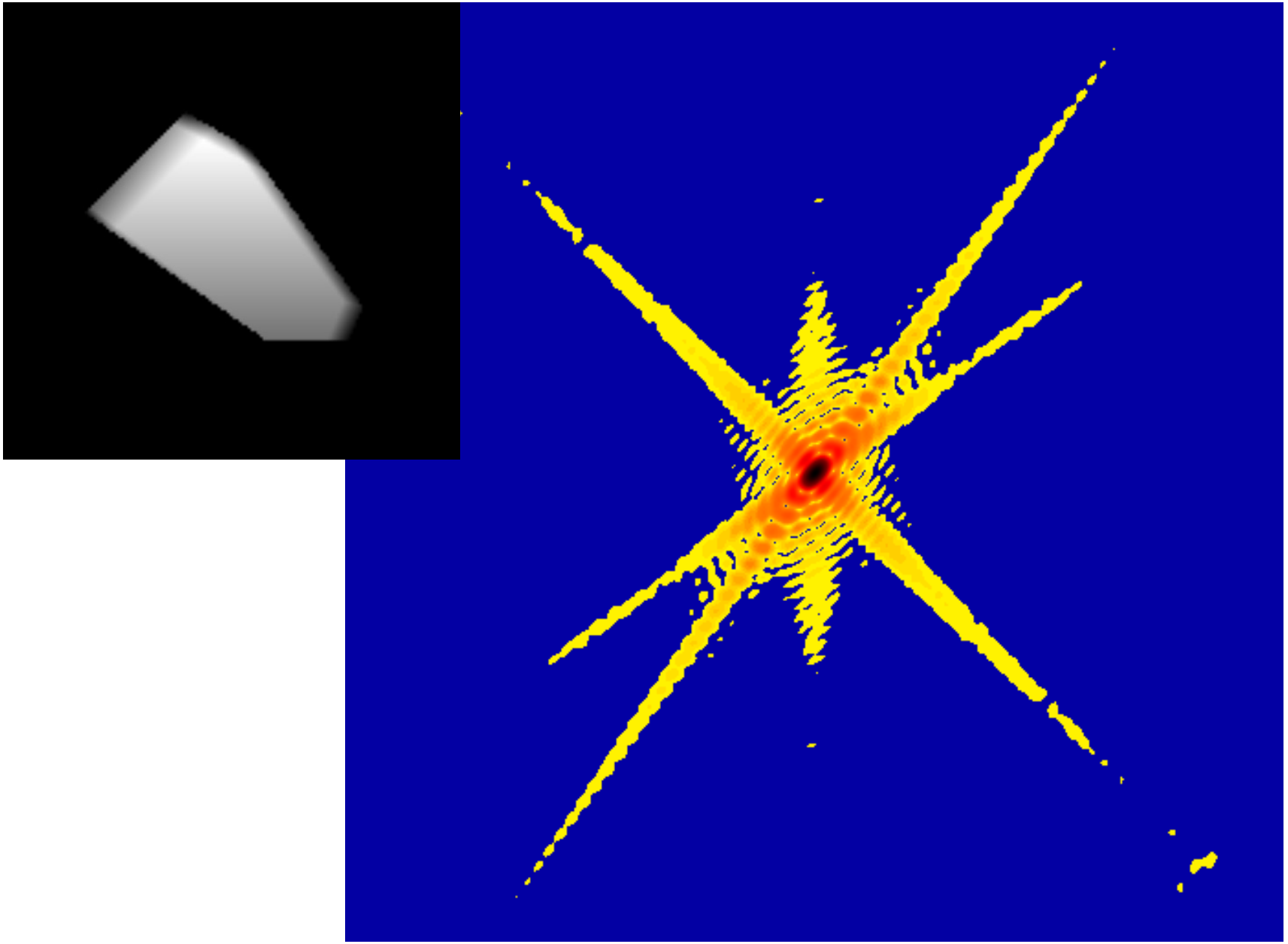


Figure 2.3: Fienup HIO

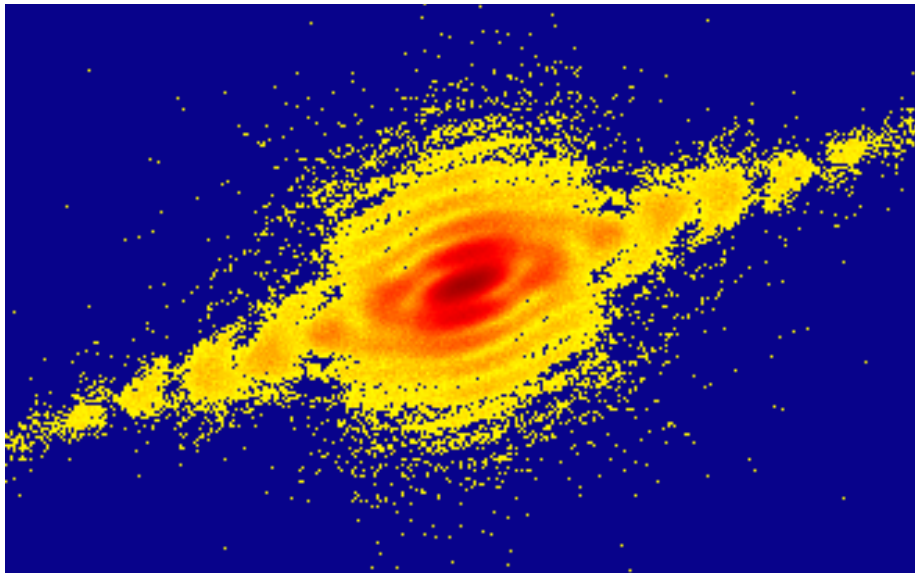
Coherent Diffraction from Crystals

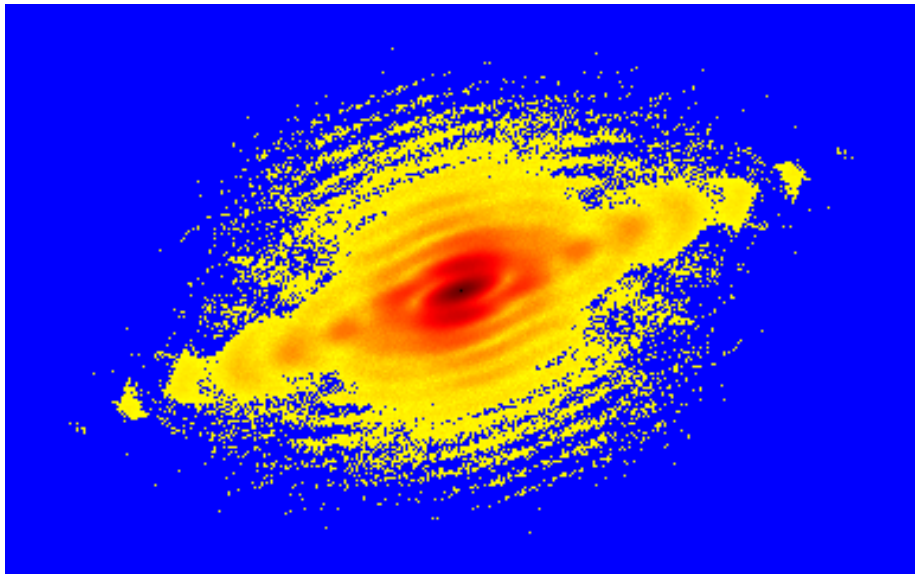


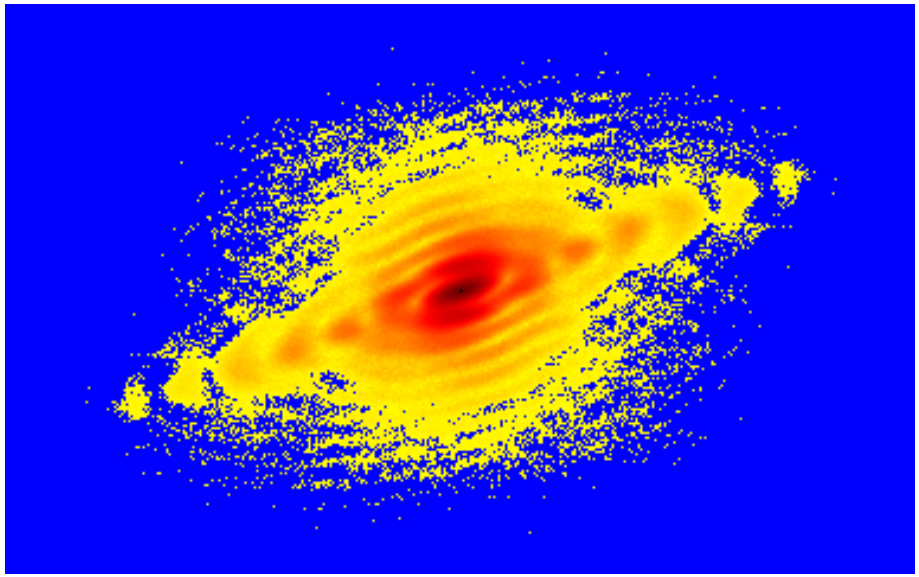


Symmetrized Data and two best fits

Chisq=0.0005







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

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

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

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

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

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

Homometric structures

Identical diffraction from two structures whenever the structure factor is factorizable

Demonstration of Uniqueness

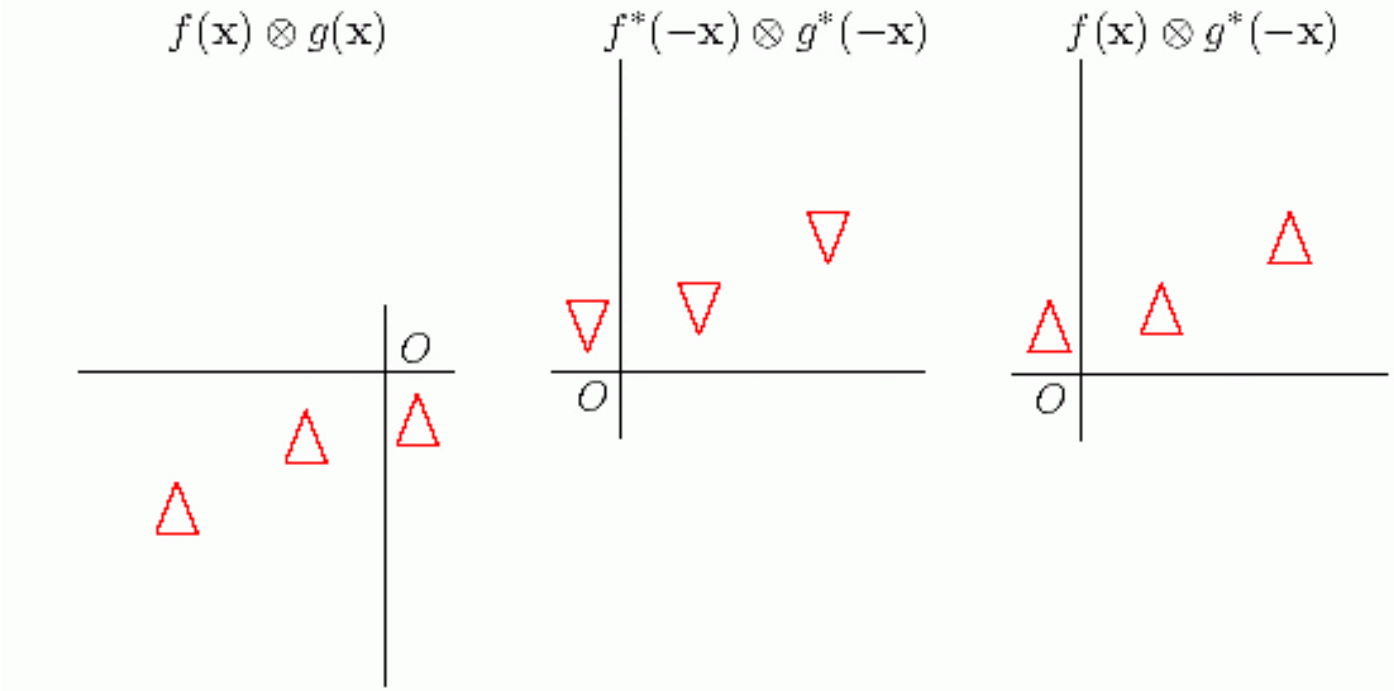
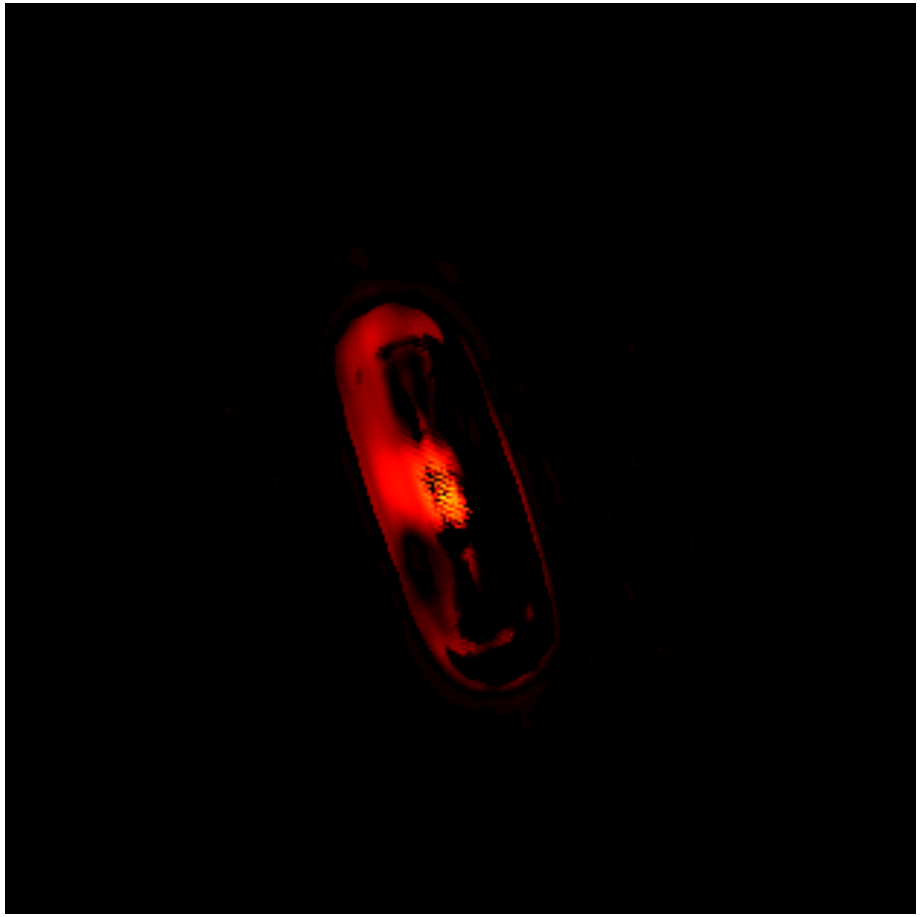
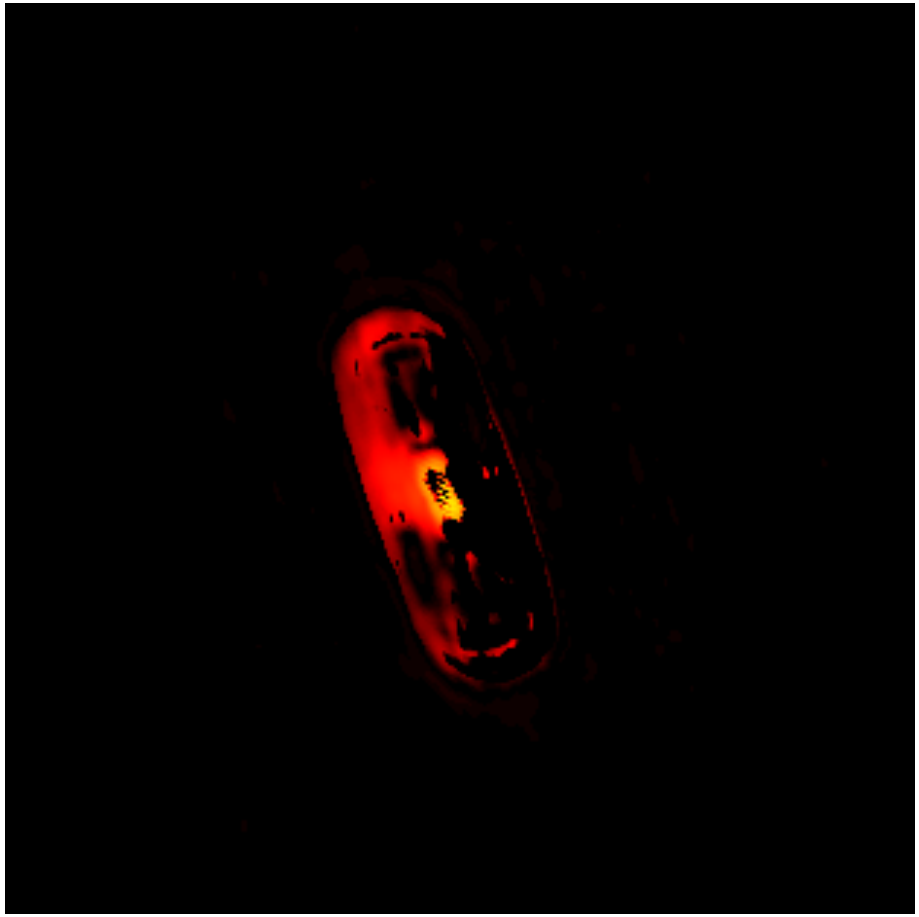


Figure 1.1: All three examples have the same Fourier magnitude, demonstrating that ambiguous solutions are possible in 2D. $f(\mathbf{x})$ is the shape of the triangle. $g(\mathbf{x}) = \delta(\mathbf{x}-\mathbf{x_1}) + \delta(\mathbf{x}-\mathbf{x_2}) + \delta(\mathbf{x}-\mathbf{x_3})$, where the vectors $\mathbf{x_i}$ 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.

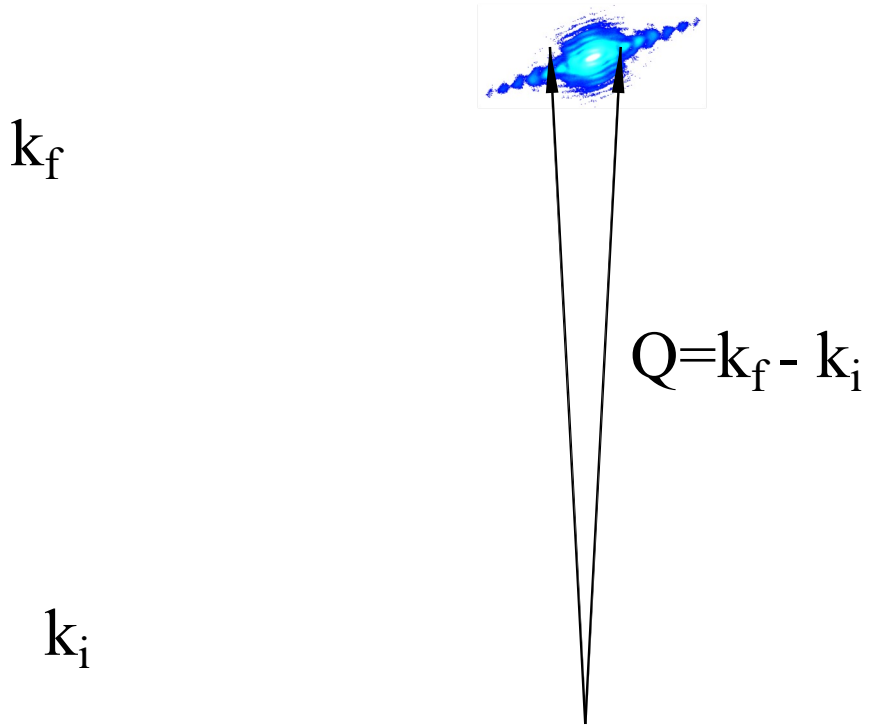
2D Reconstructions

chisquare = 0.0005

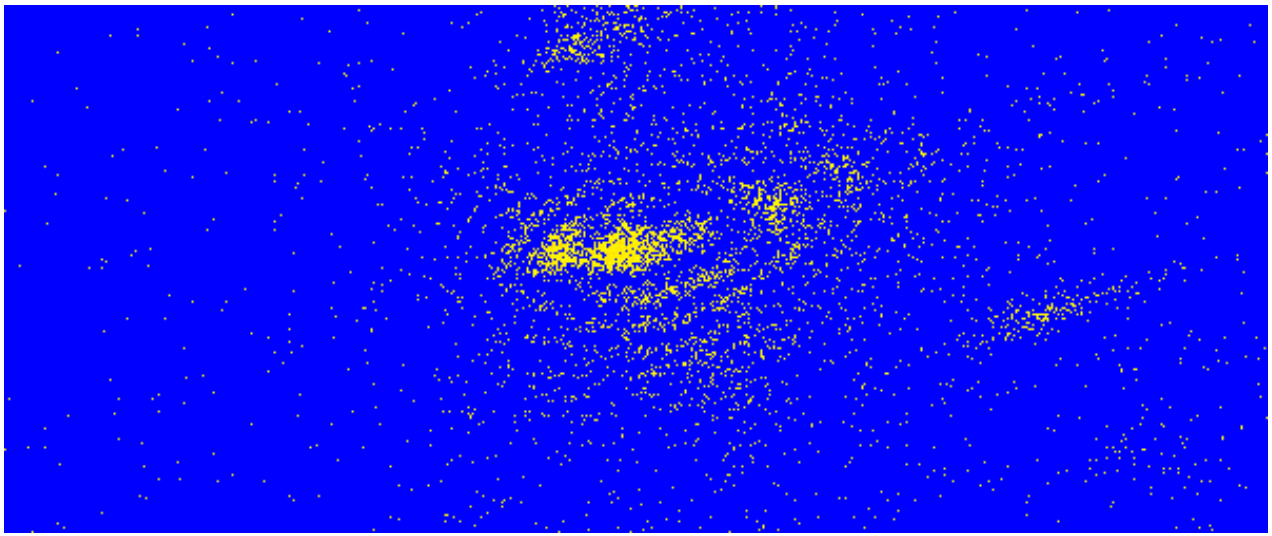


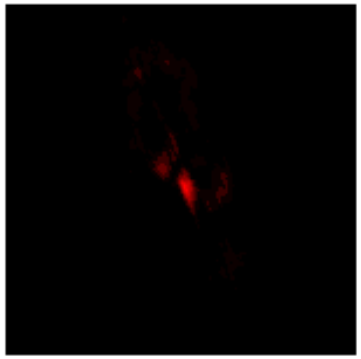
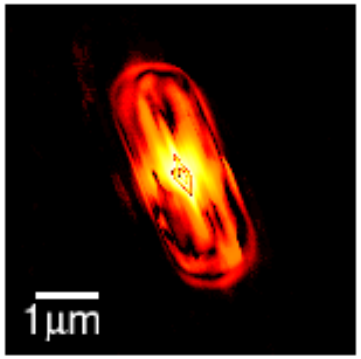


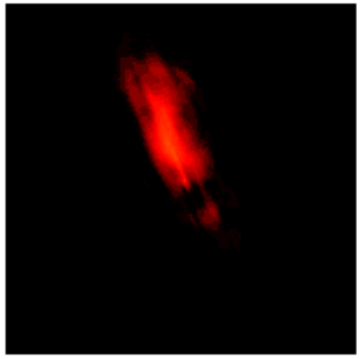
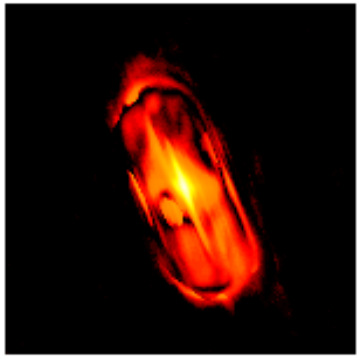
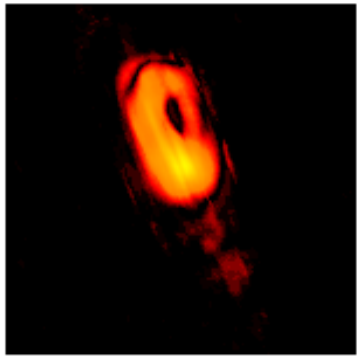
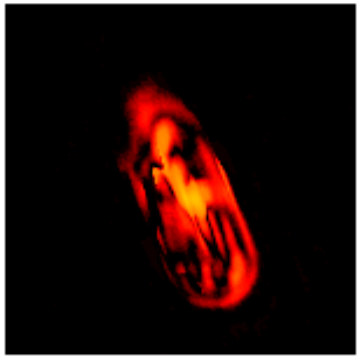
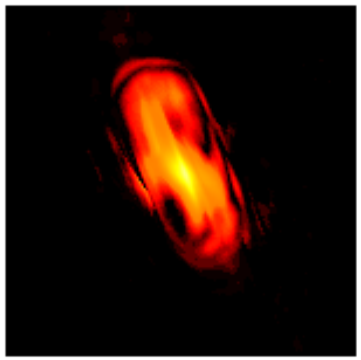
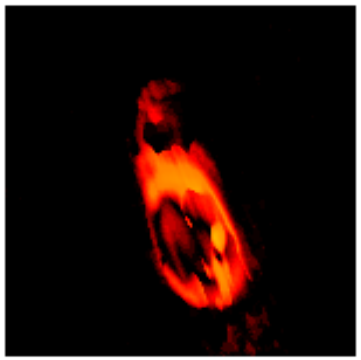
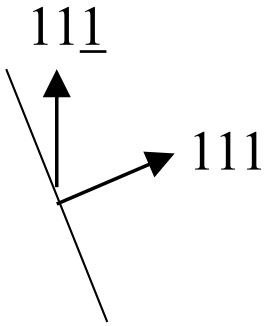
3D Diffraction Method

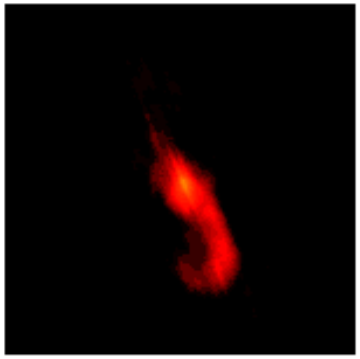
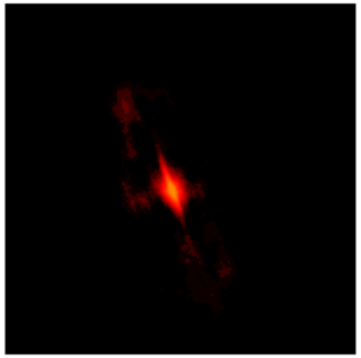


3D Diffraction Data
1 micron Au crystal

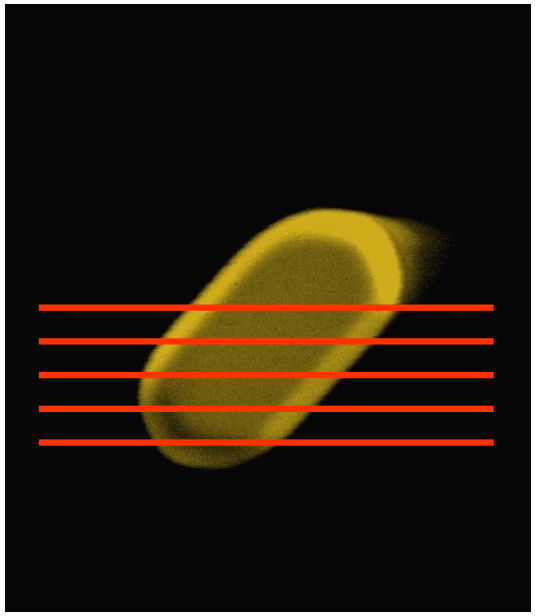


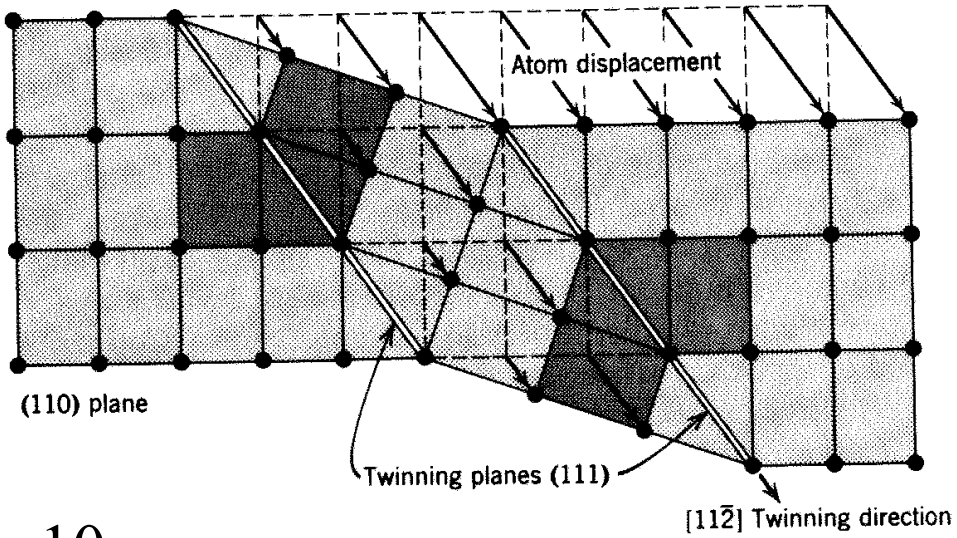



Slices through plan view SEM:



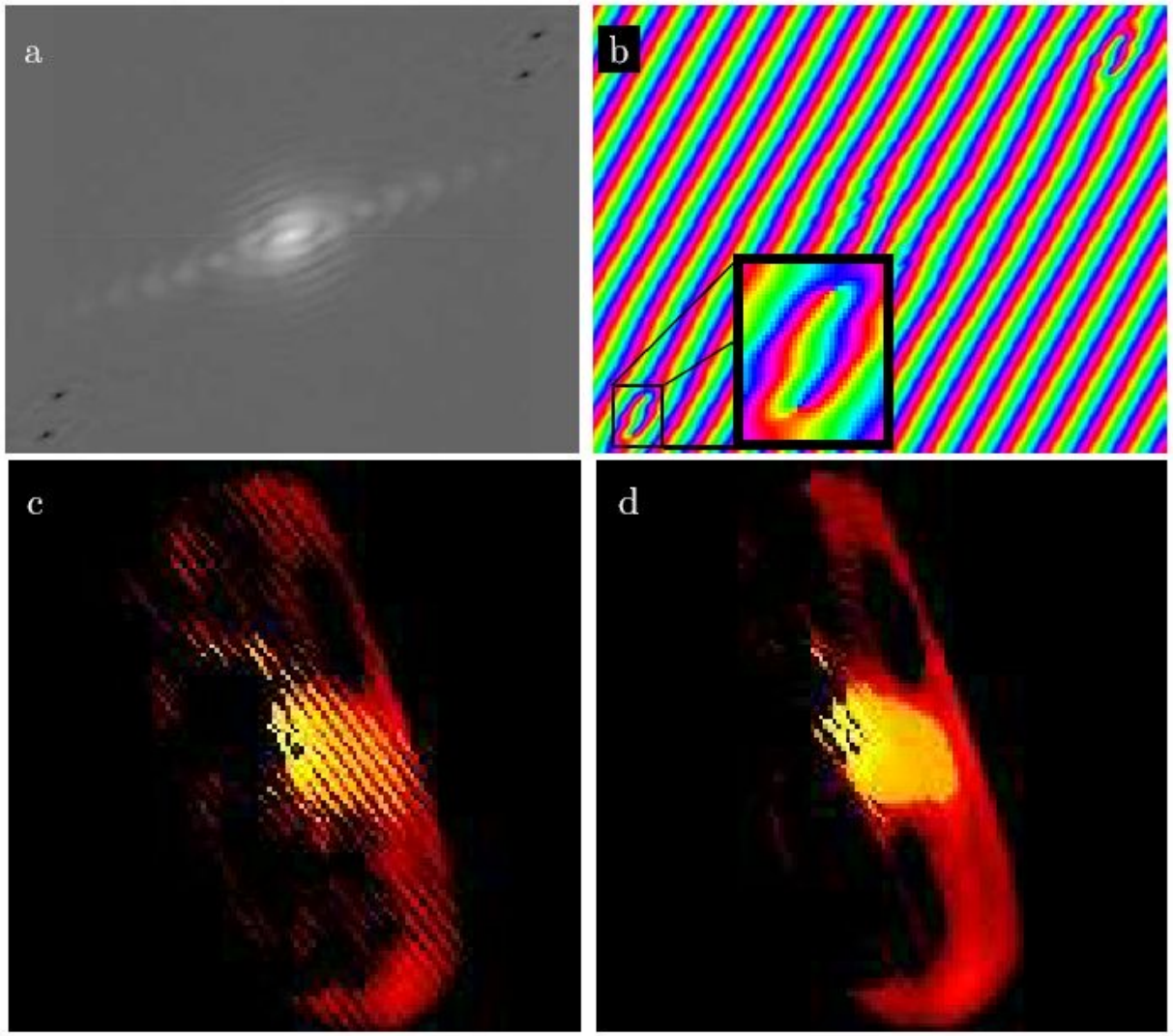
Twinning in deformed FCC metals

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

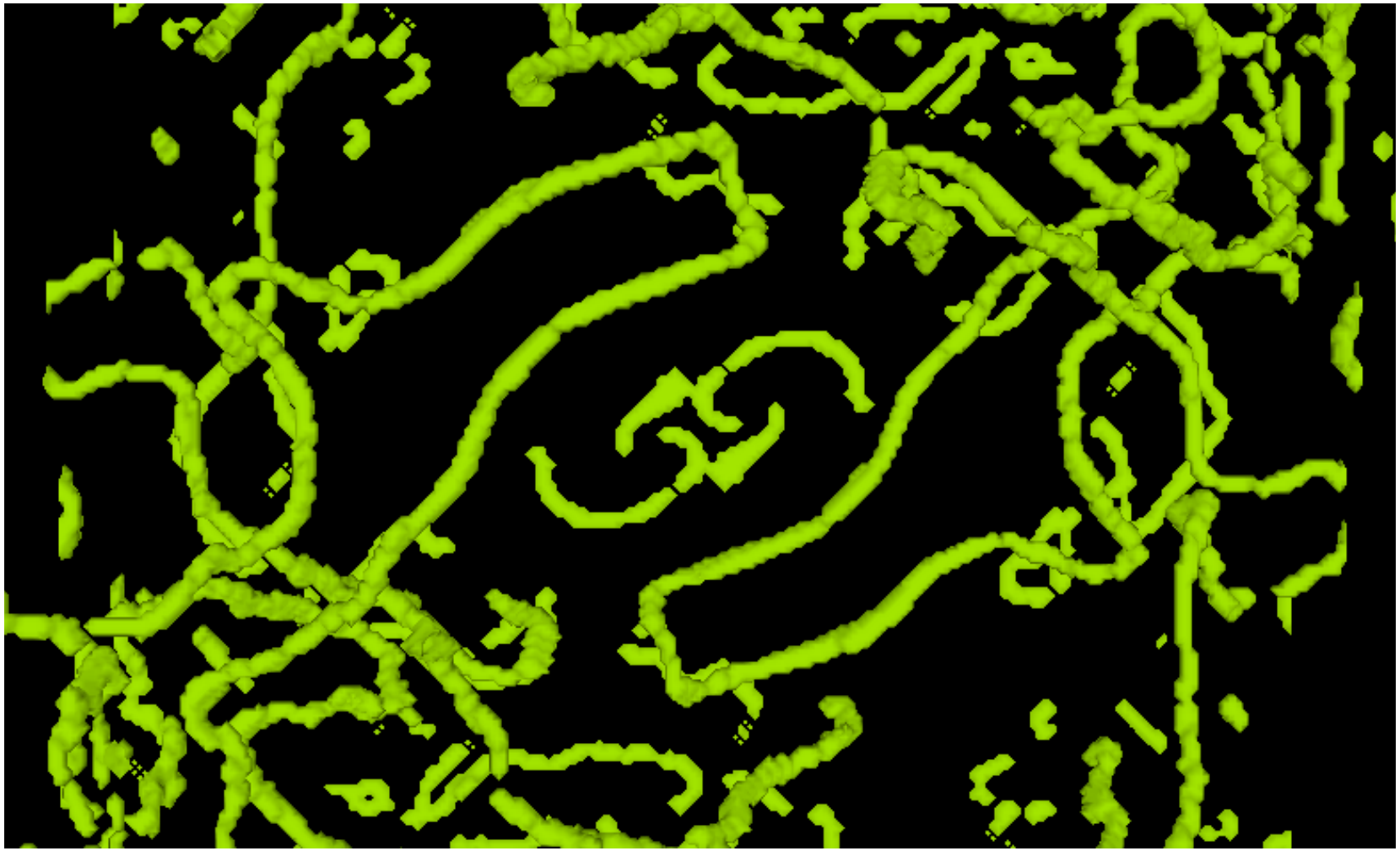


~10μm
Cu

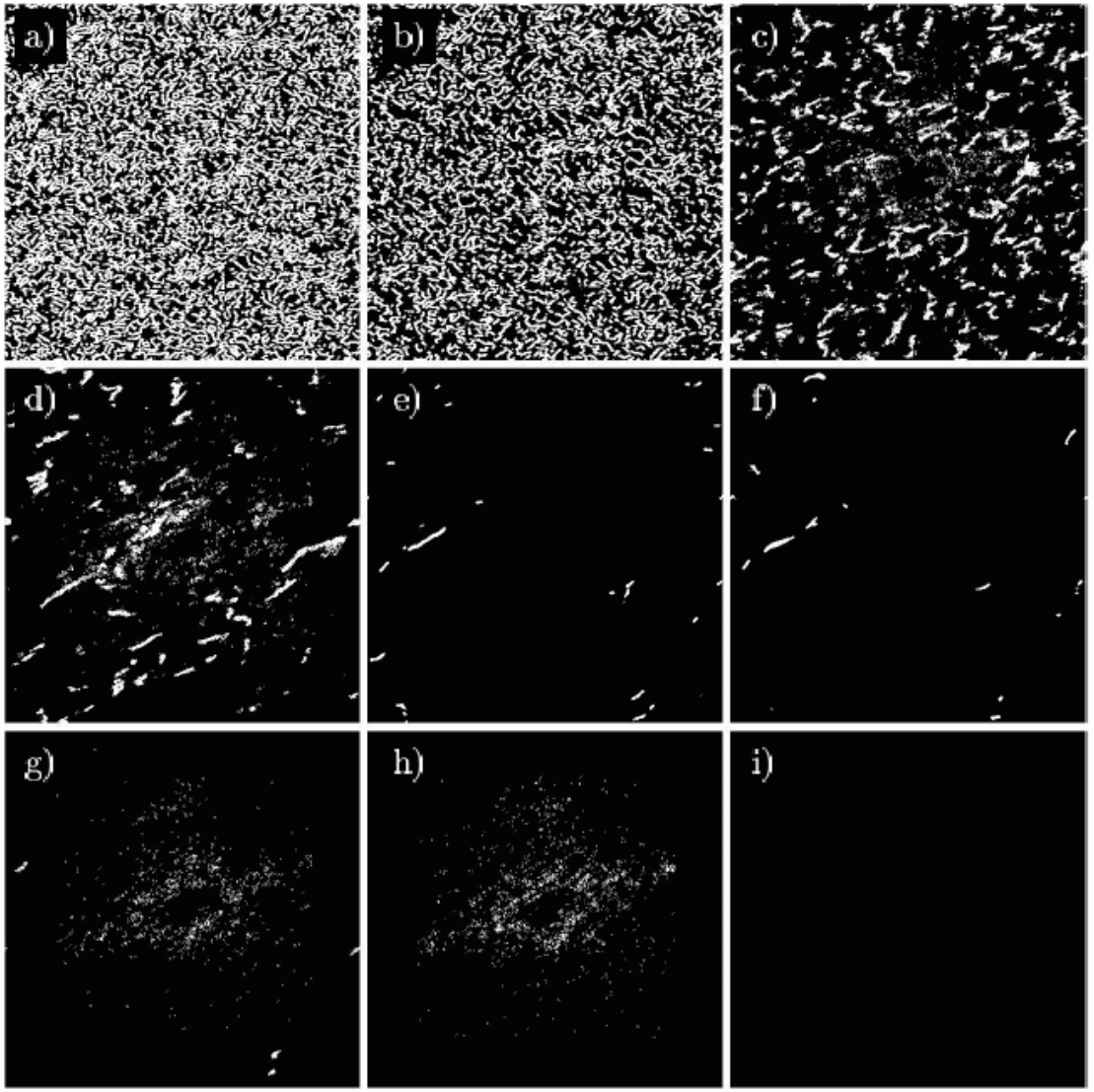
Phase Vortices and patching



‘Vortices’ Form Loops in 3D

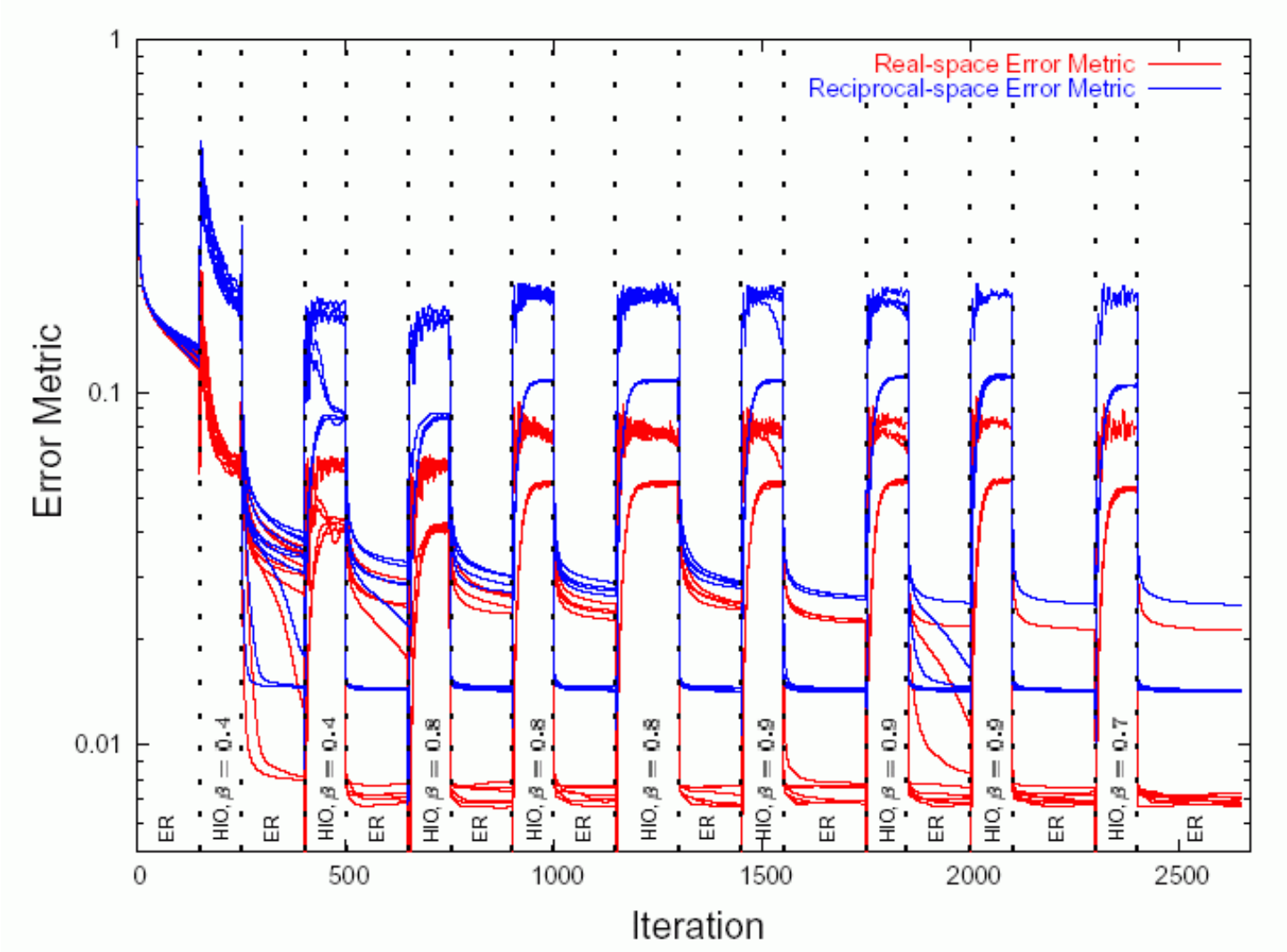


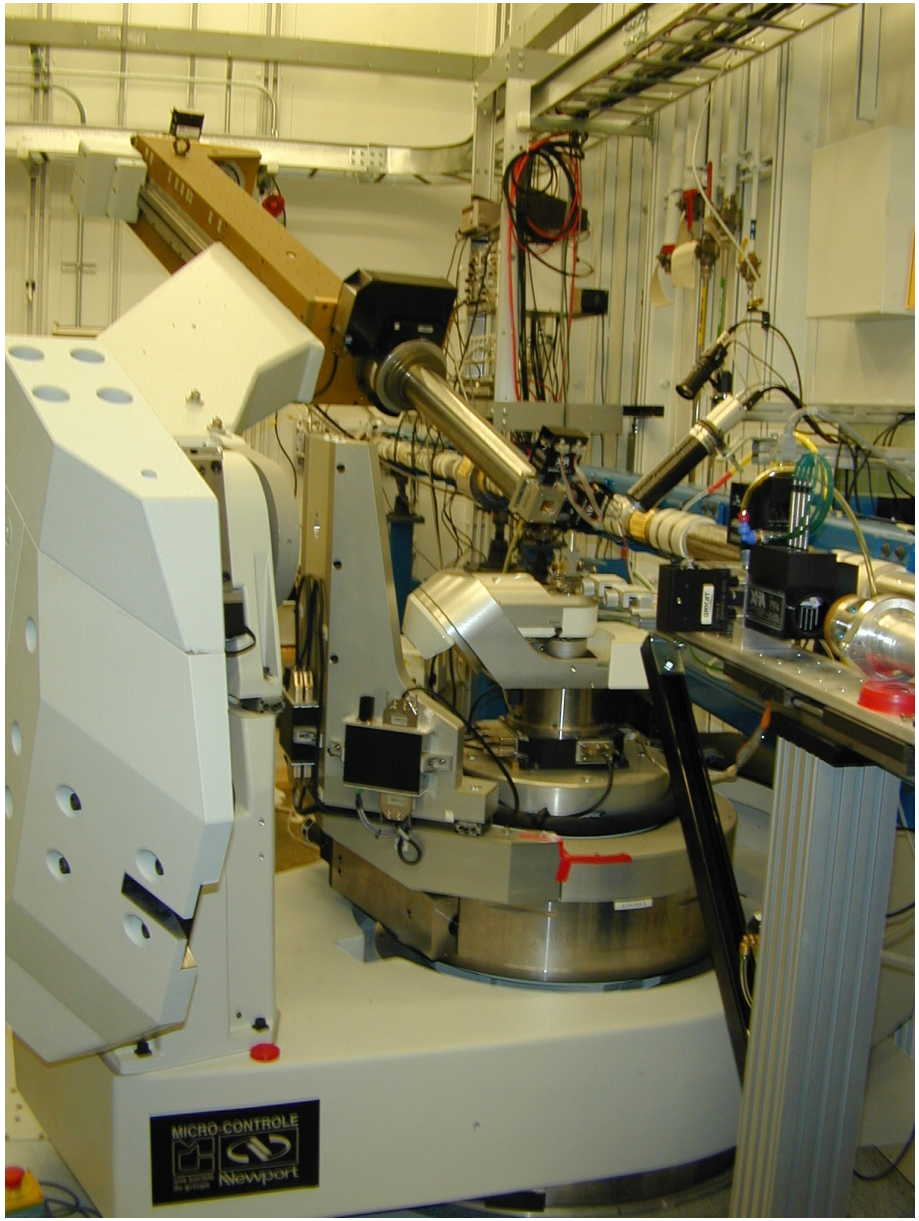
Clearing of Vortices during HIO

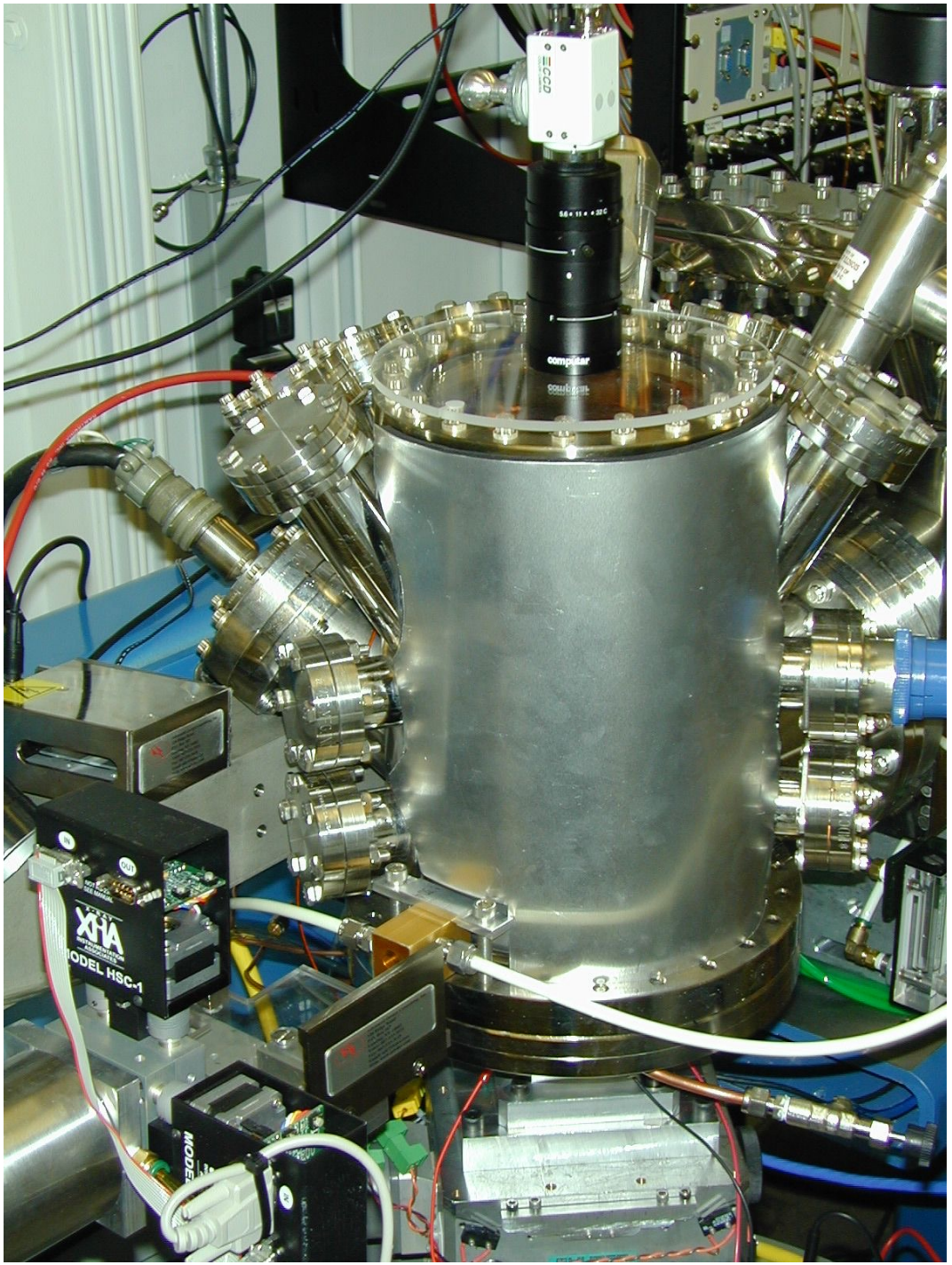


ER ER HIO
HIO ER ER
HIO HIO ER

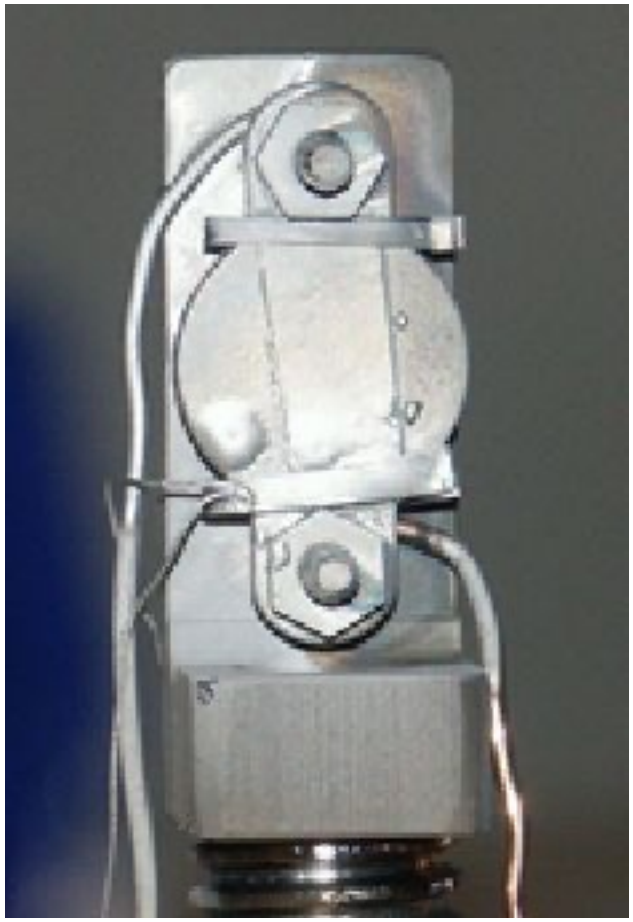
Progress of Phase Retrieval

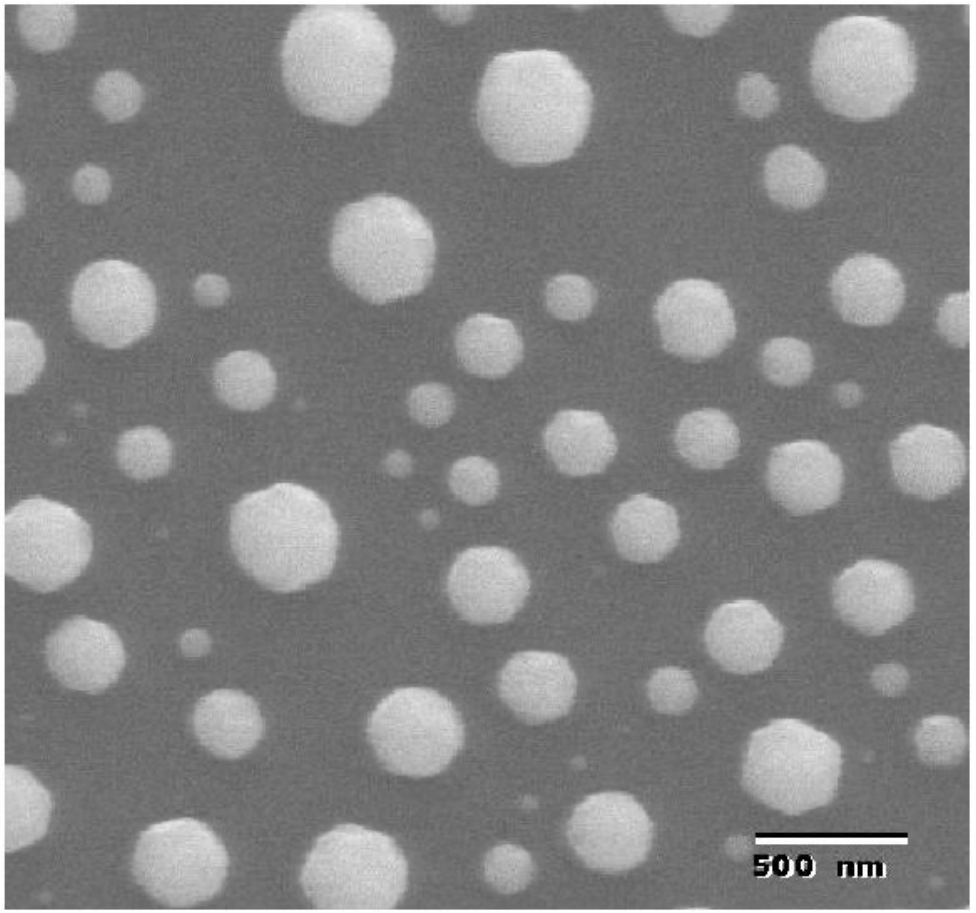



MICRO-CONTROLE
Newport

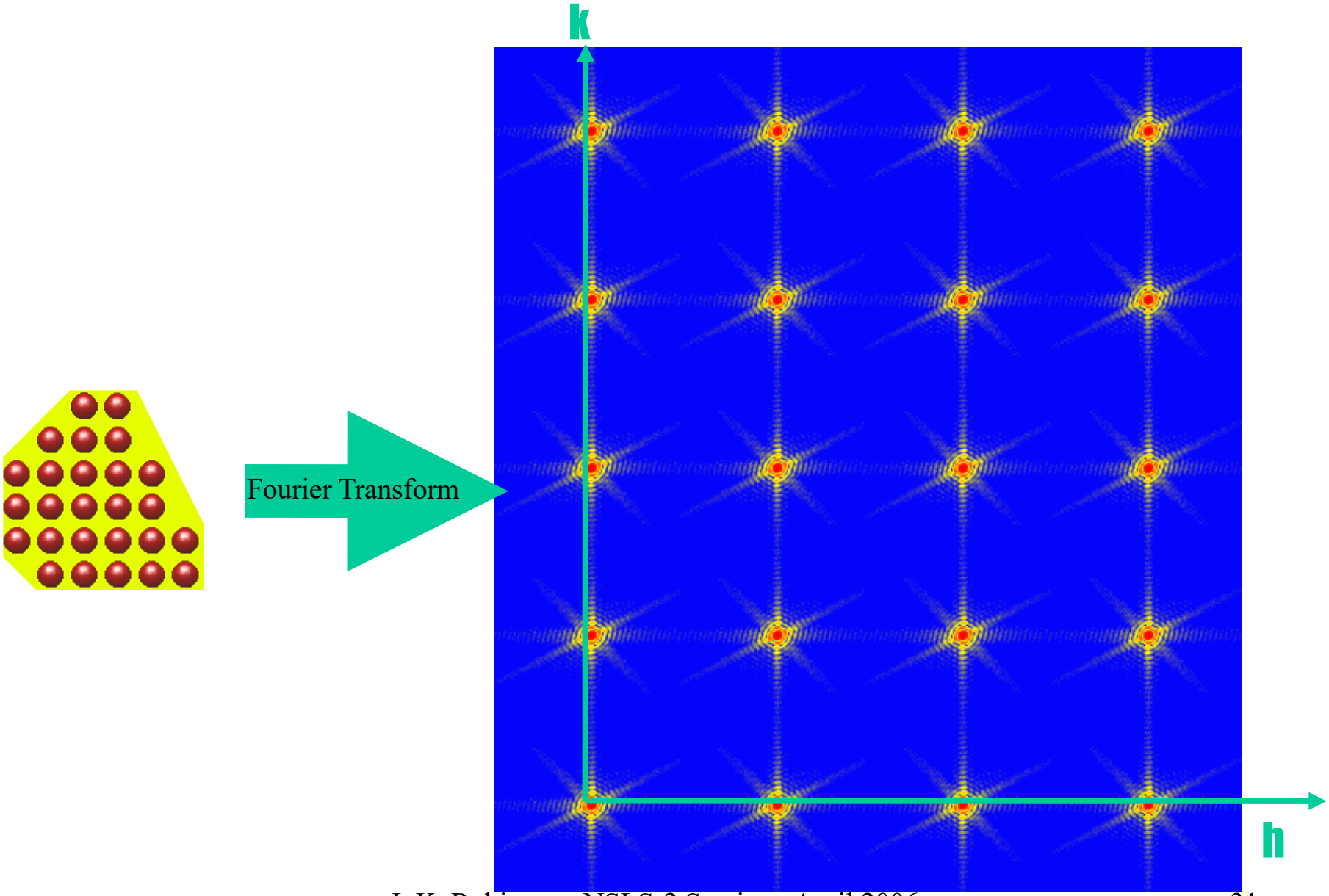

computar
MODEL HSC-1

In situ growth of Pb crystals





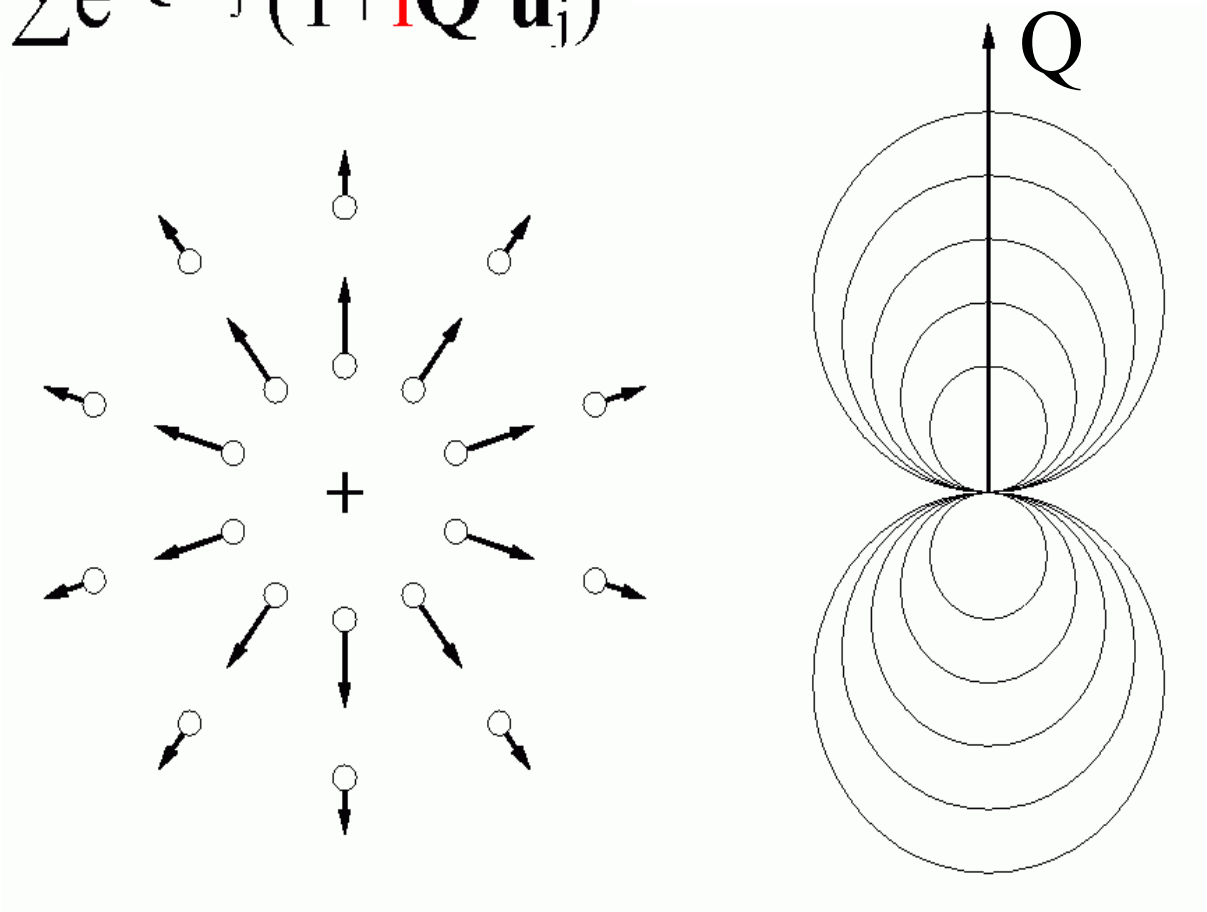
Coherent Diffraction from Crystals



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

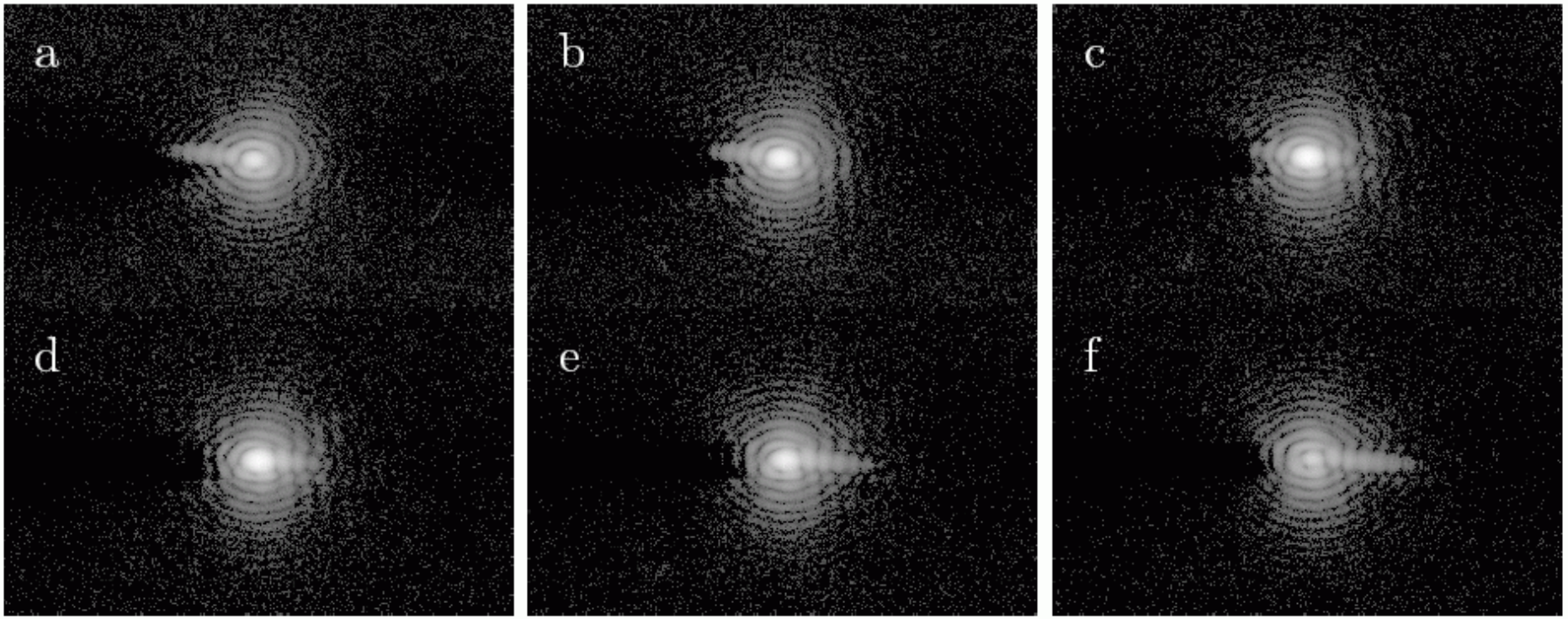
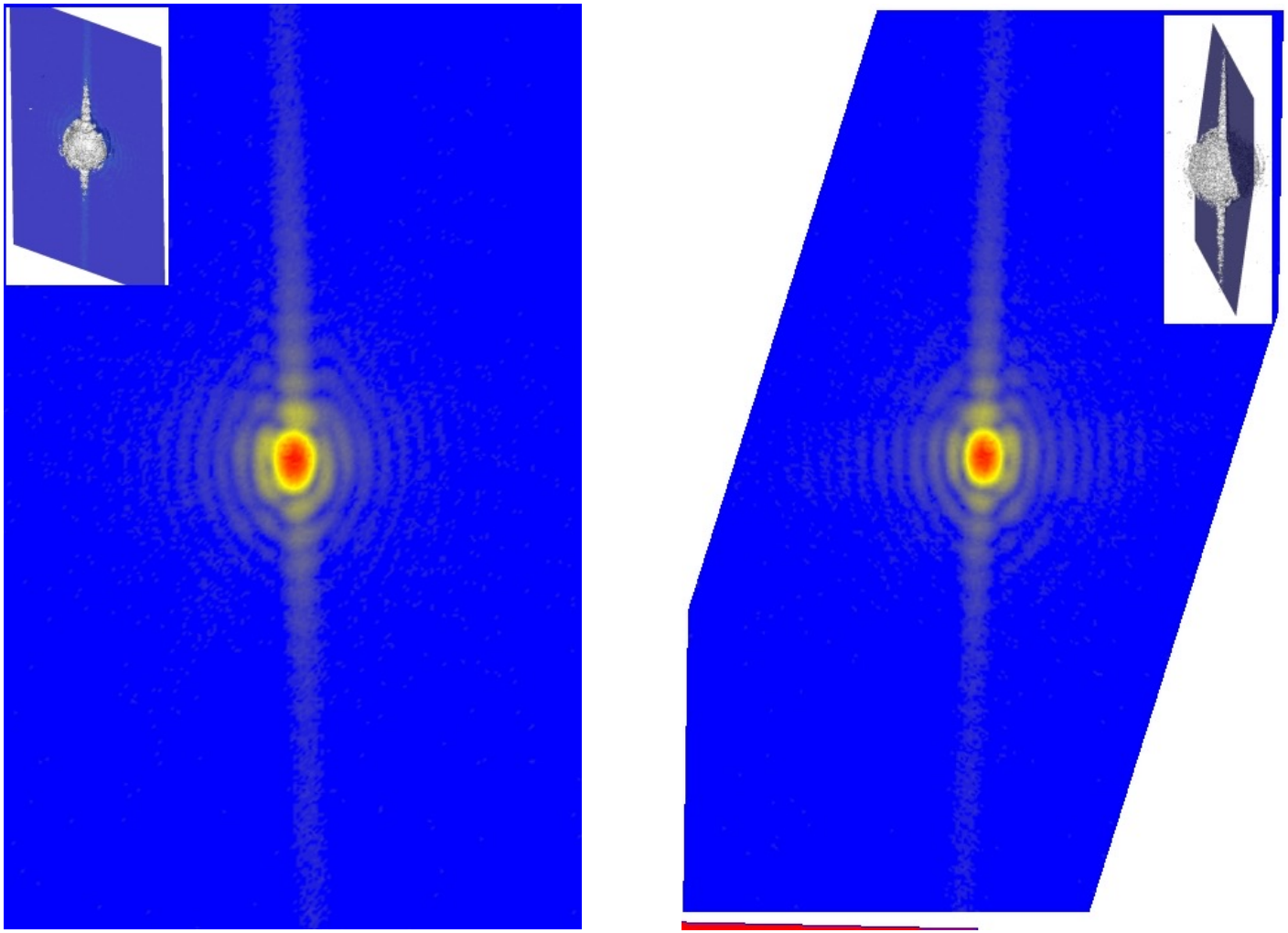
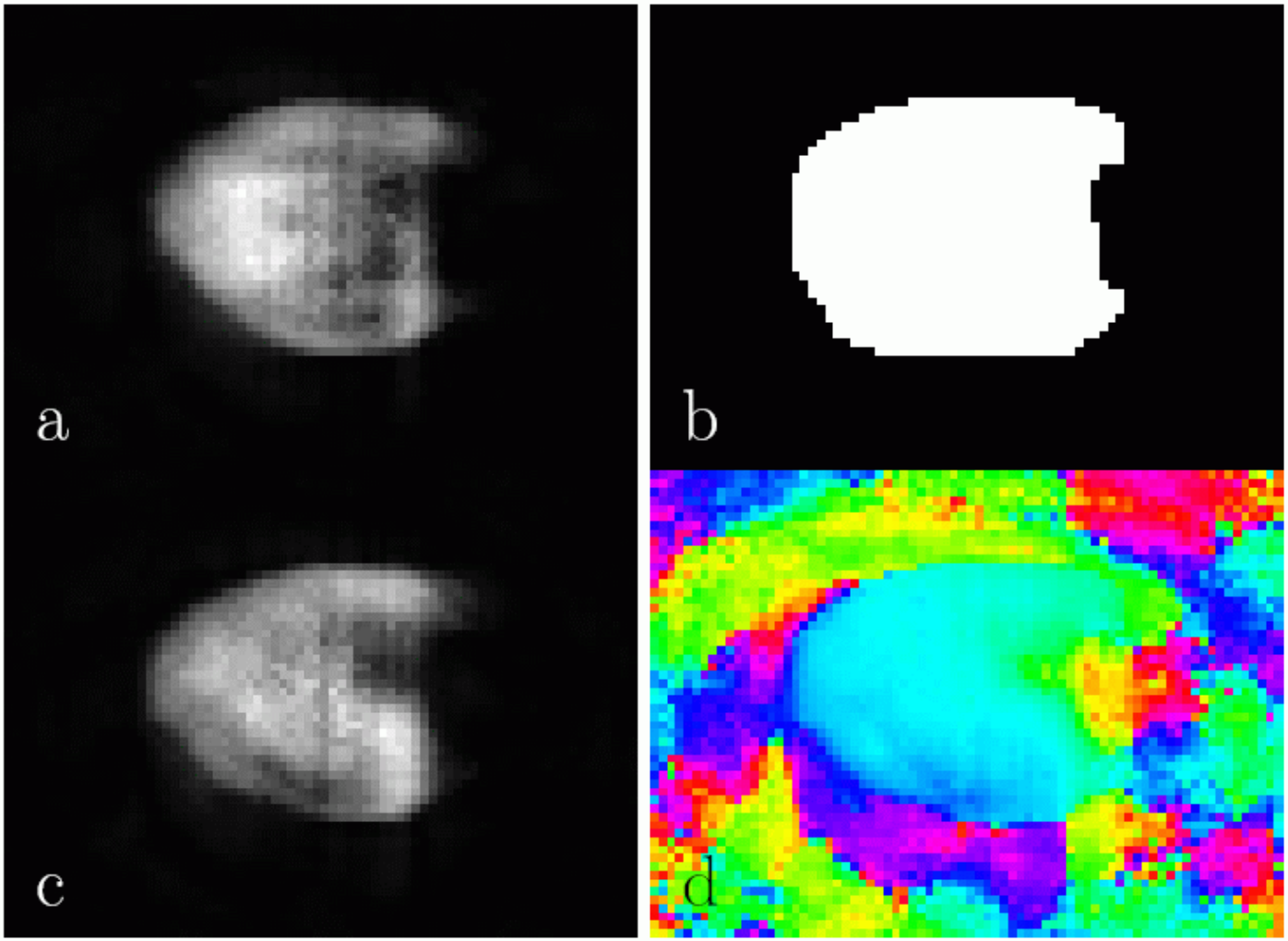


Figure 4.12: Center slices from 3D CXD pattern from Pb sample, on a log scale. Data file 296 from 10/03.

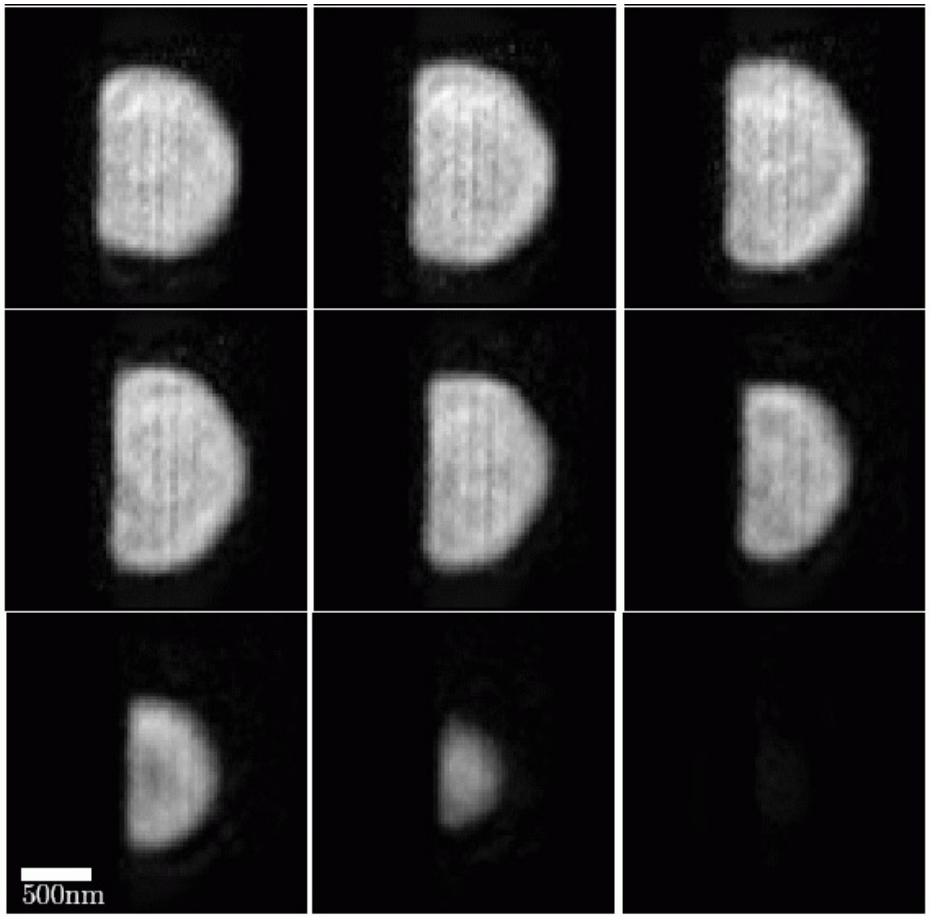
3D data along special directions

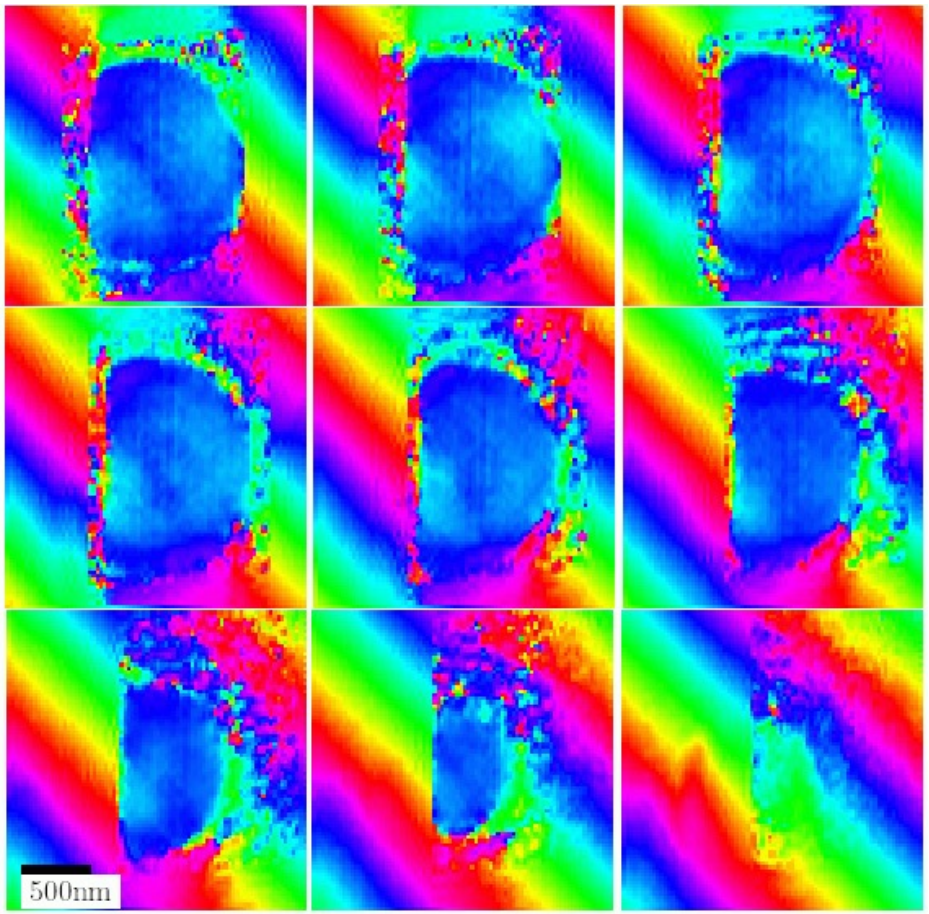


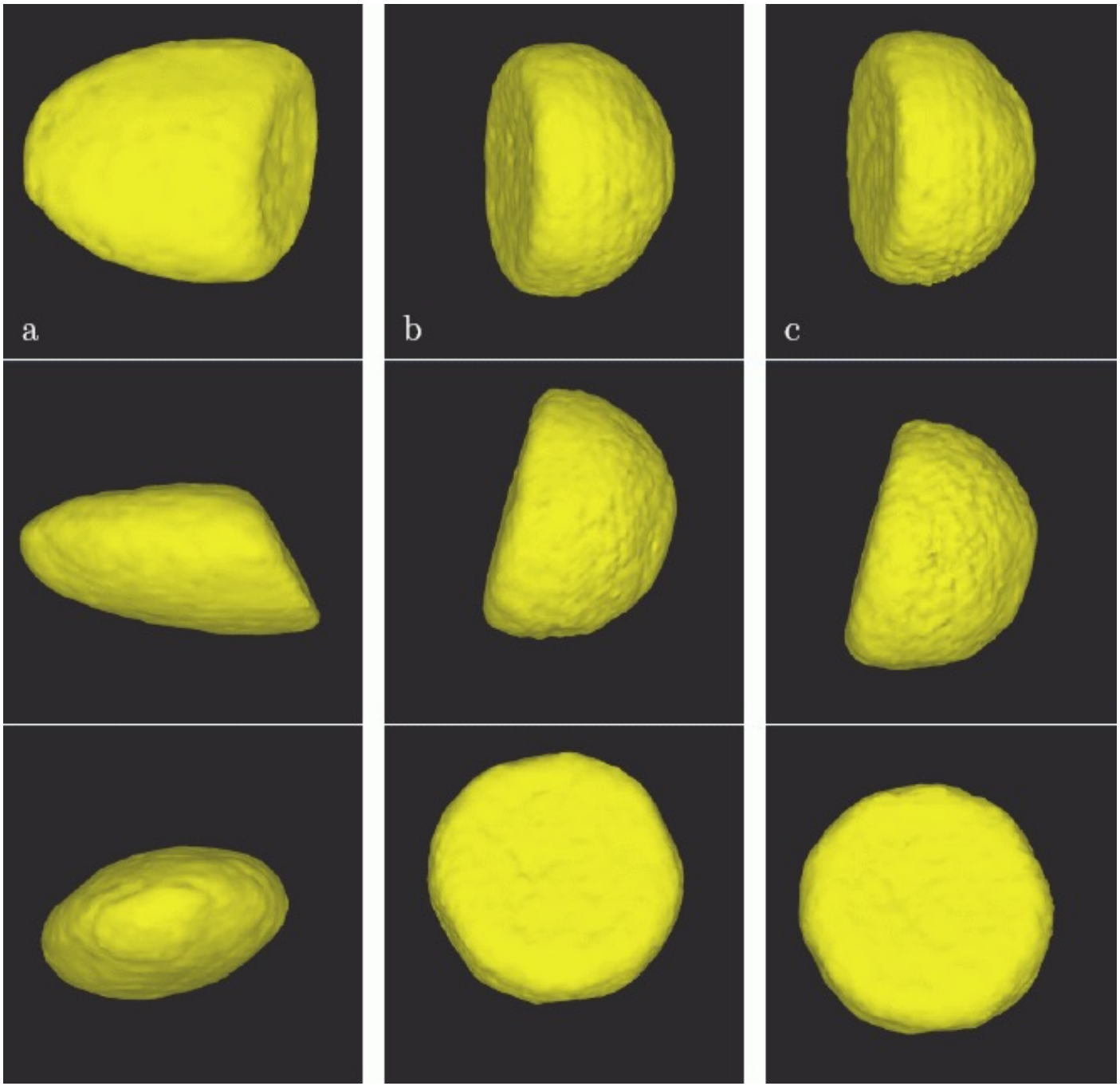
Learn shape of “tight” support



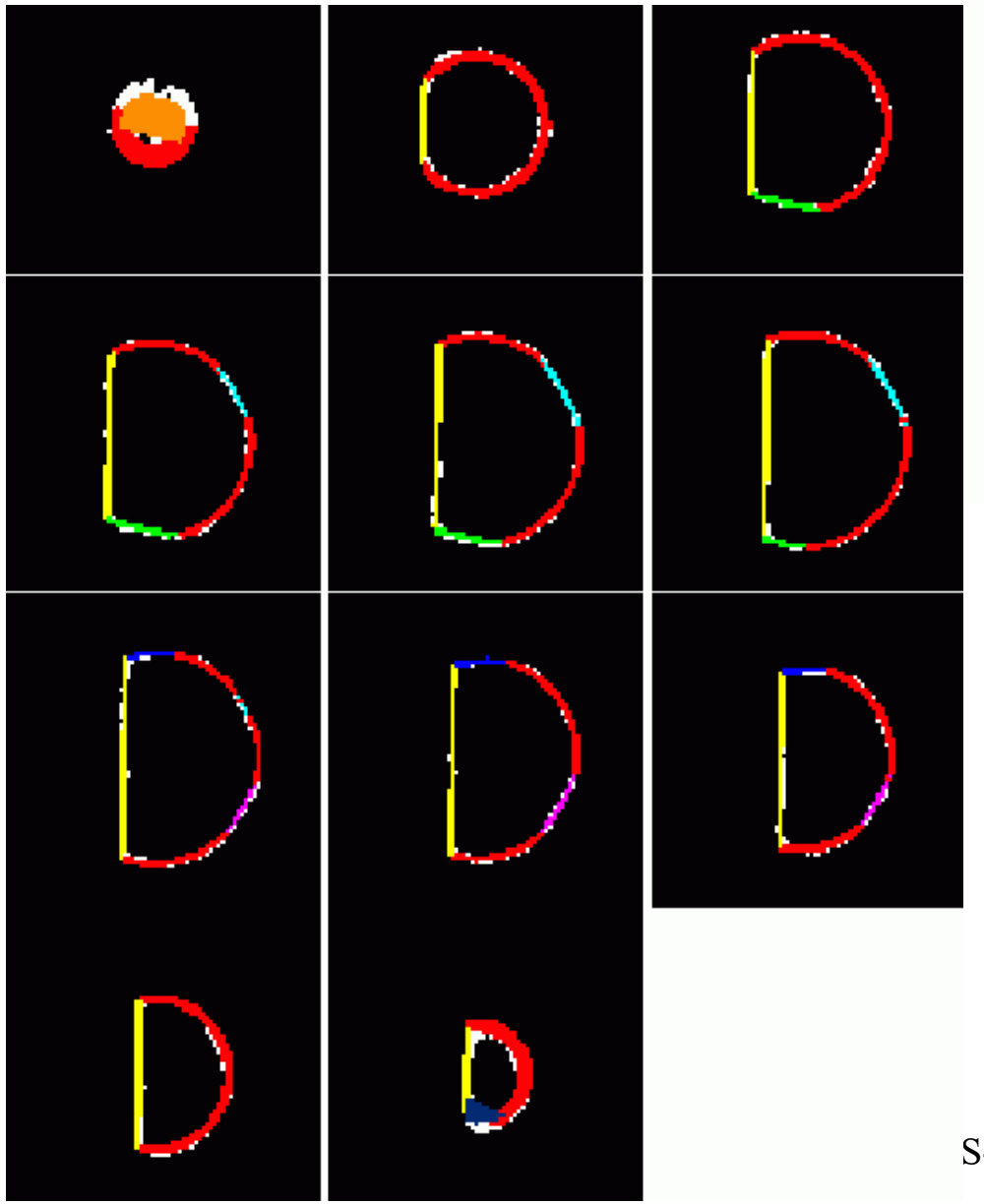
Then refine amplitude *and* phase





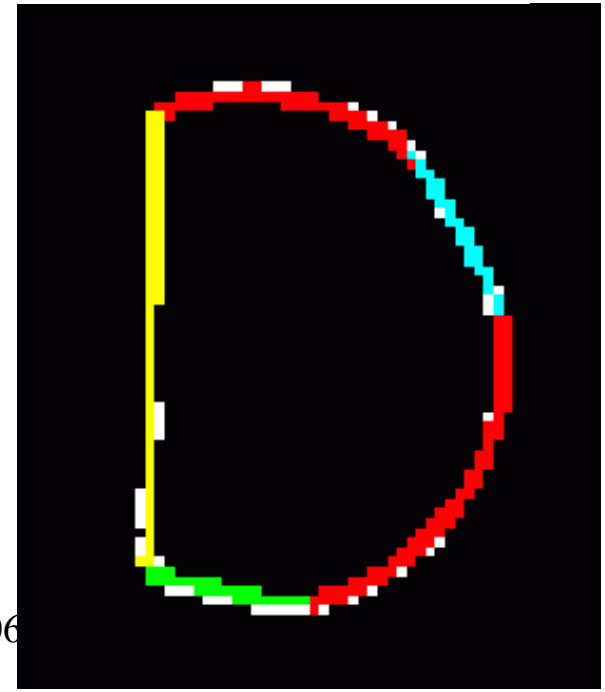

a
b
c

Fitting to facetted shape

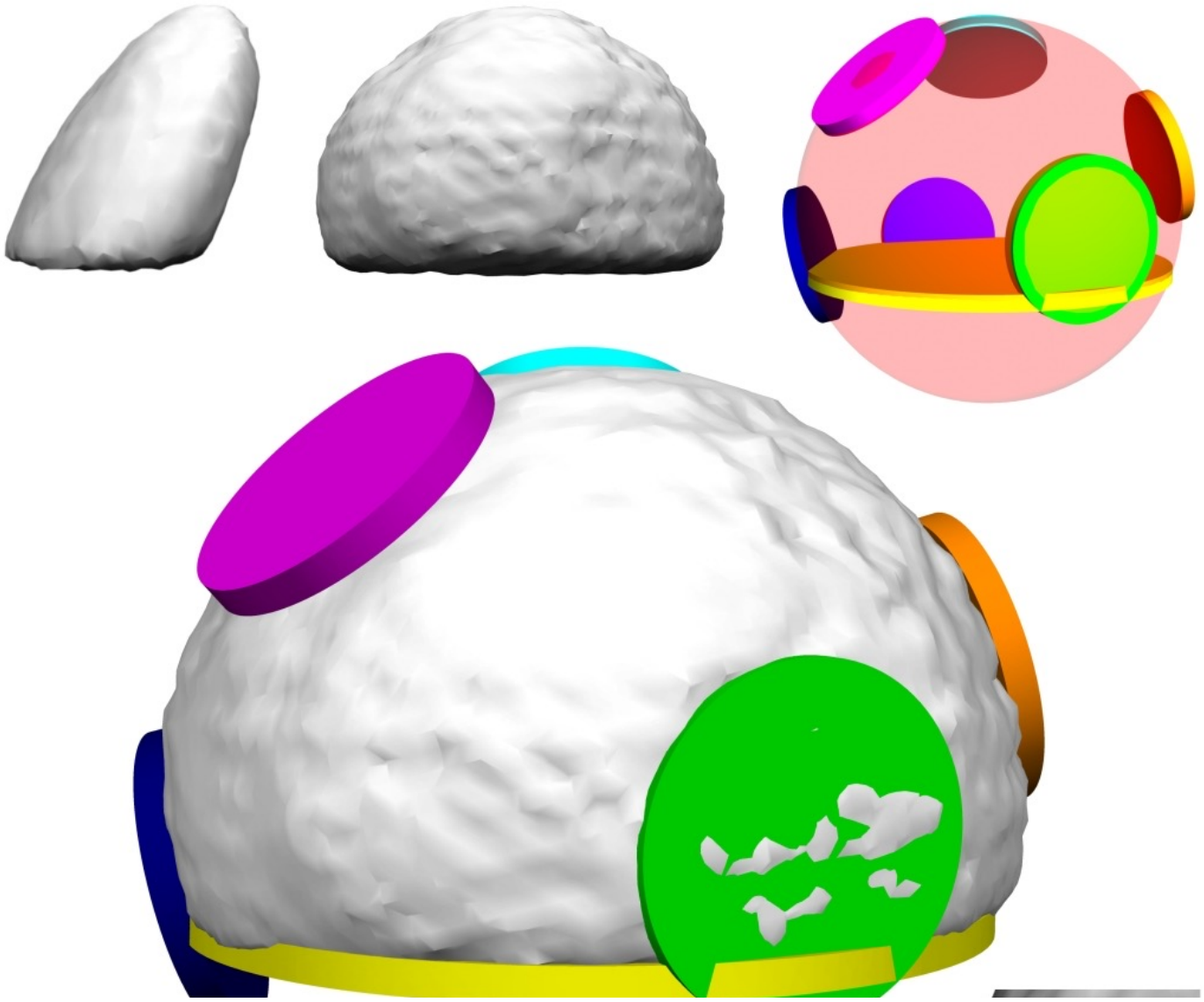


	$\|R\|$		P0	P1	P2	P3	P4	P5	P6
	9.4	P0	0	85	149	79	134	106	71
	25.7	P1		0	123	164	83	76	102
	25.1	P2			0	72	67	74	110
	25.9	P3				0	111	106	76
	25.4	P4					0	113	68
	25.4	P5						0	176
	26.0	P6							0 0

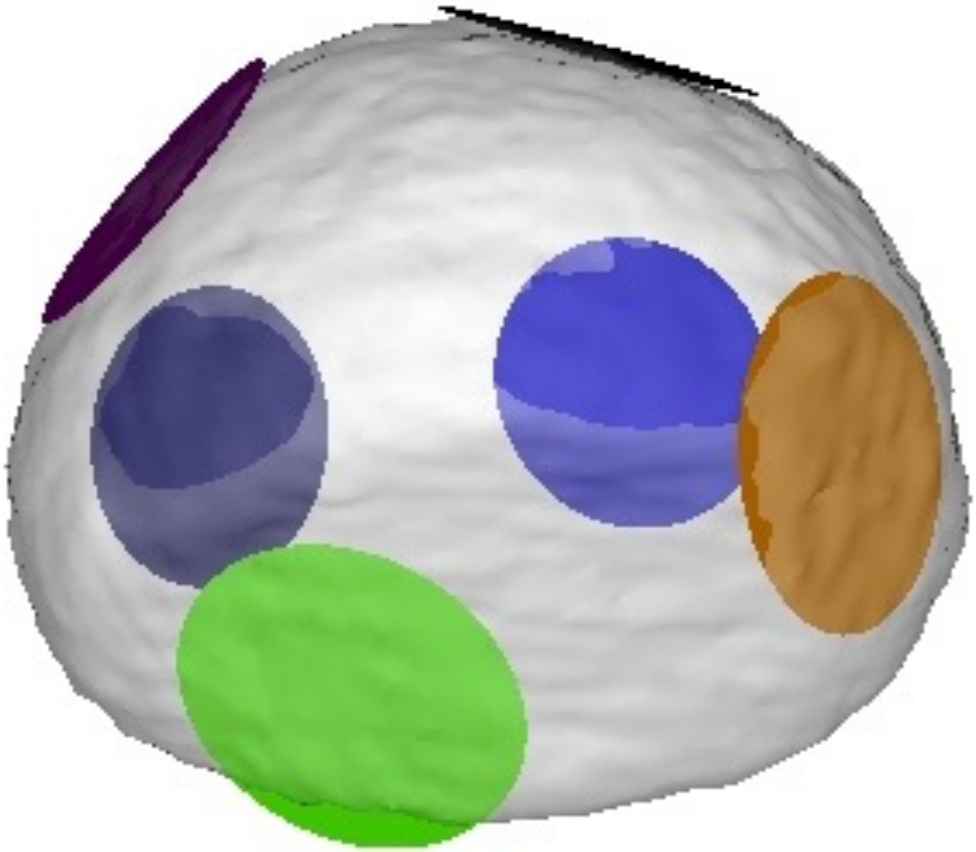
Angles between facets

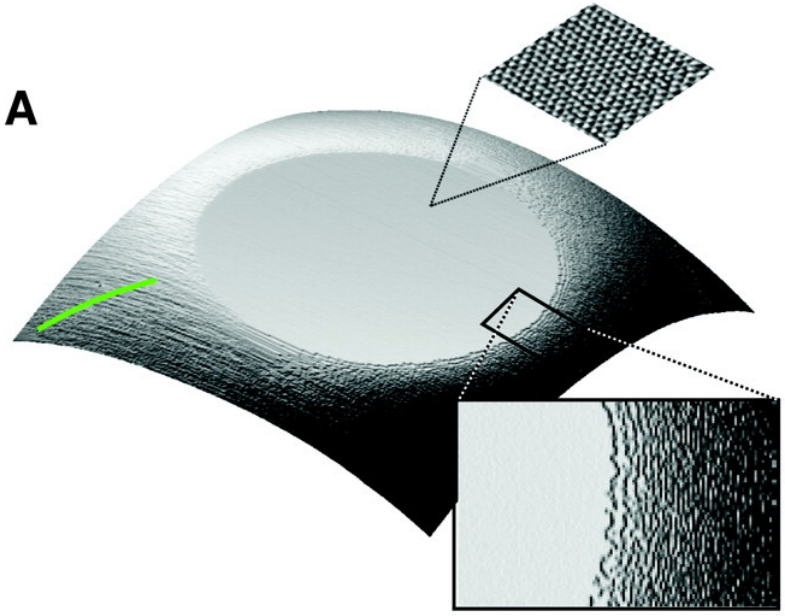


Fitting of crystal shape



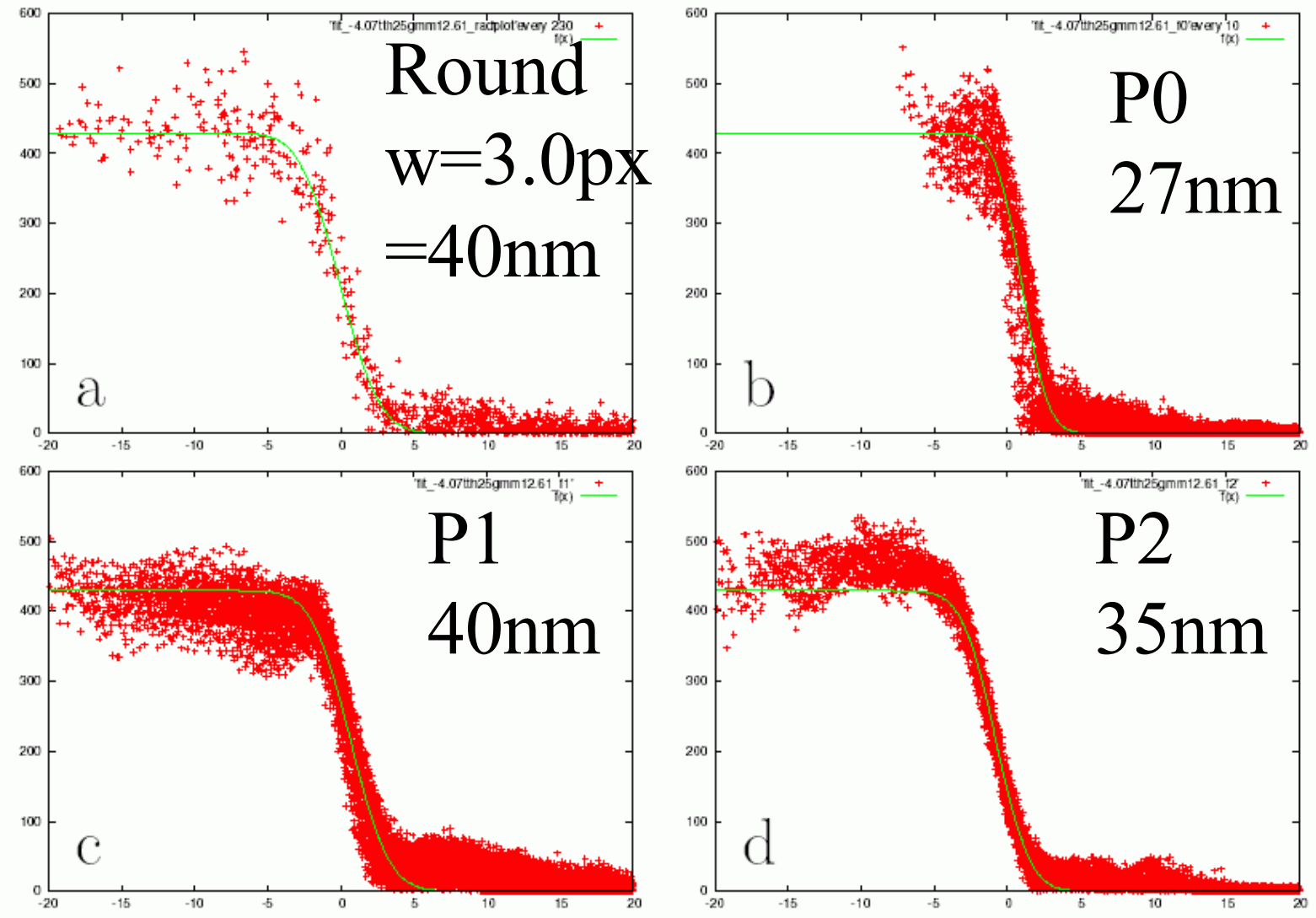
Facets of Equilibrium Crystal Shape



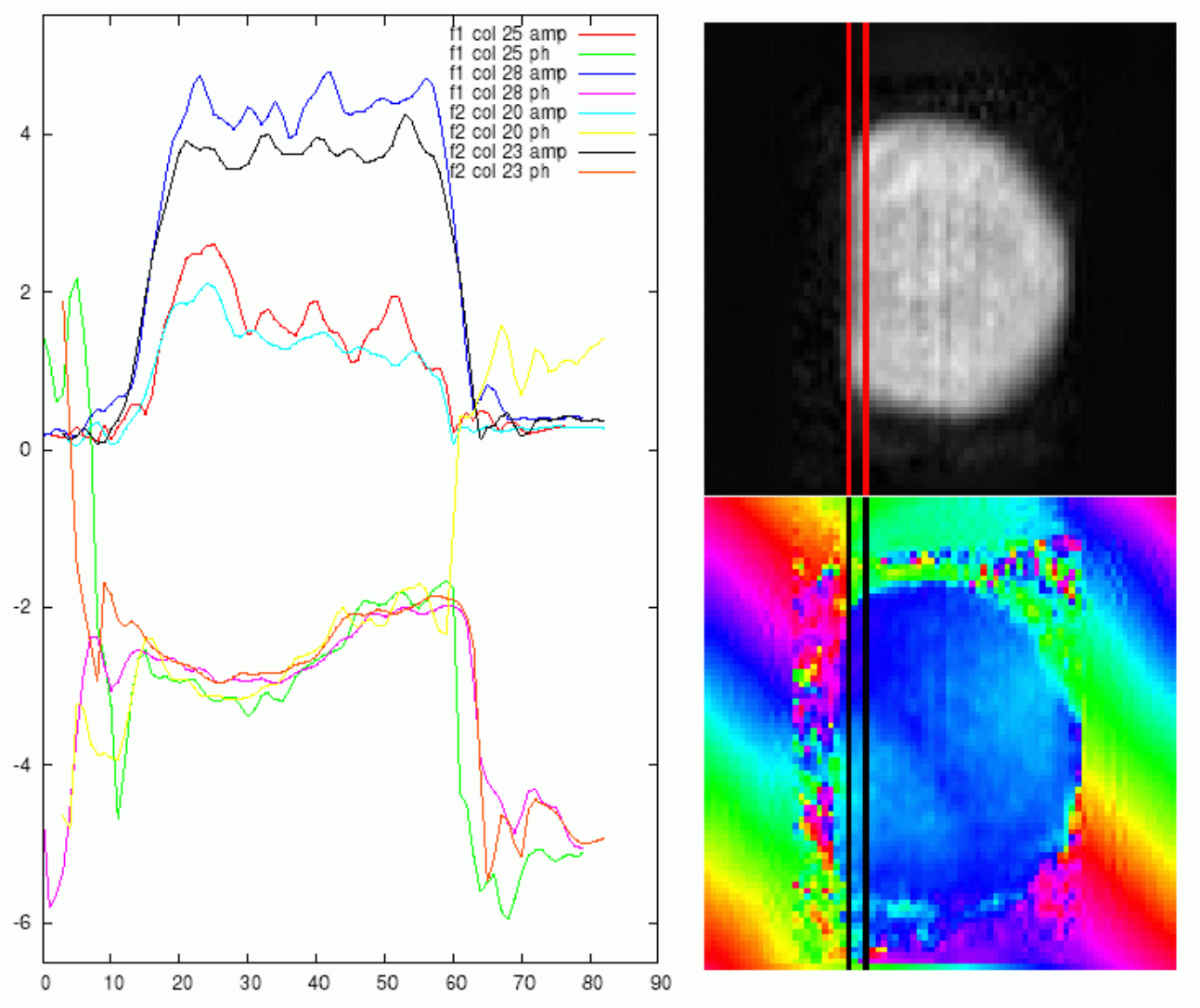


Thurmer K, Williams E, Reutt-Robey J
Science **297** 2033 (2002)

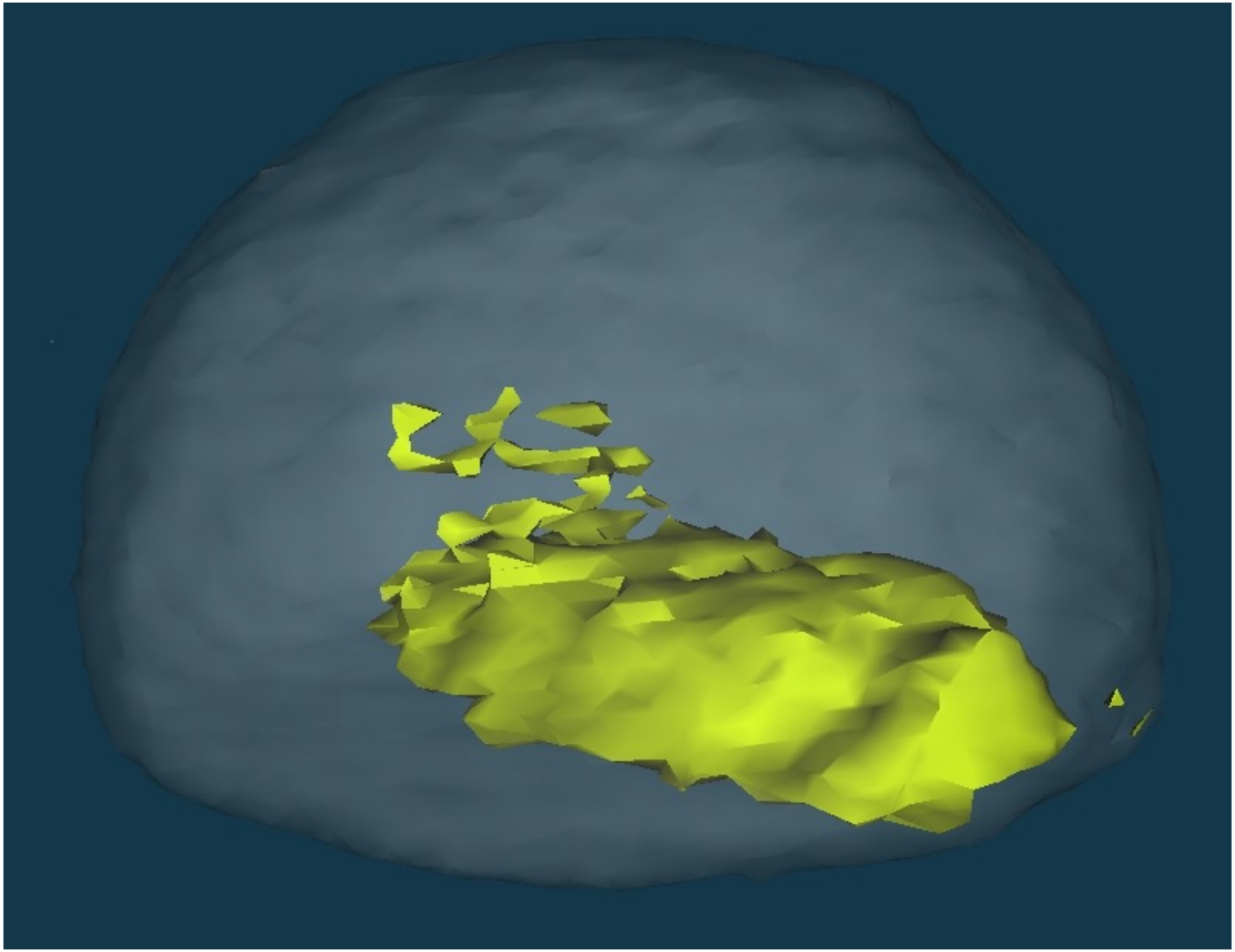
Density distribution across surface



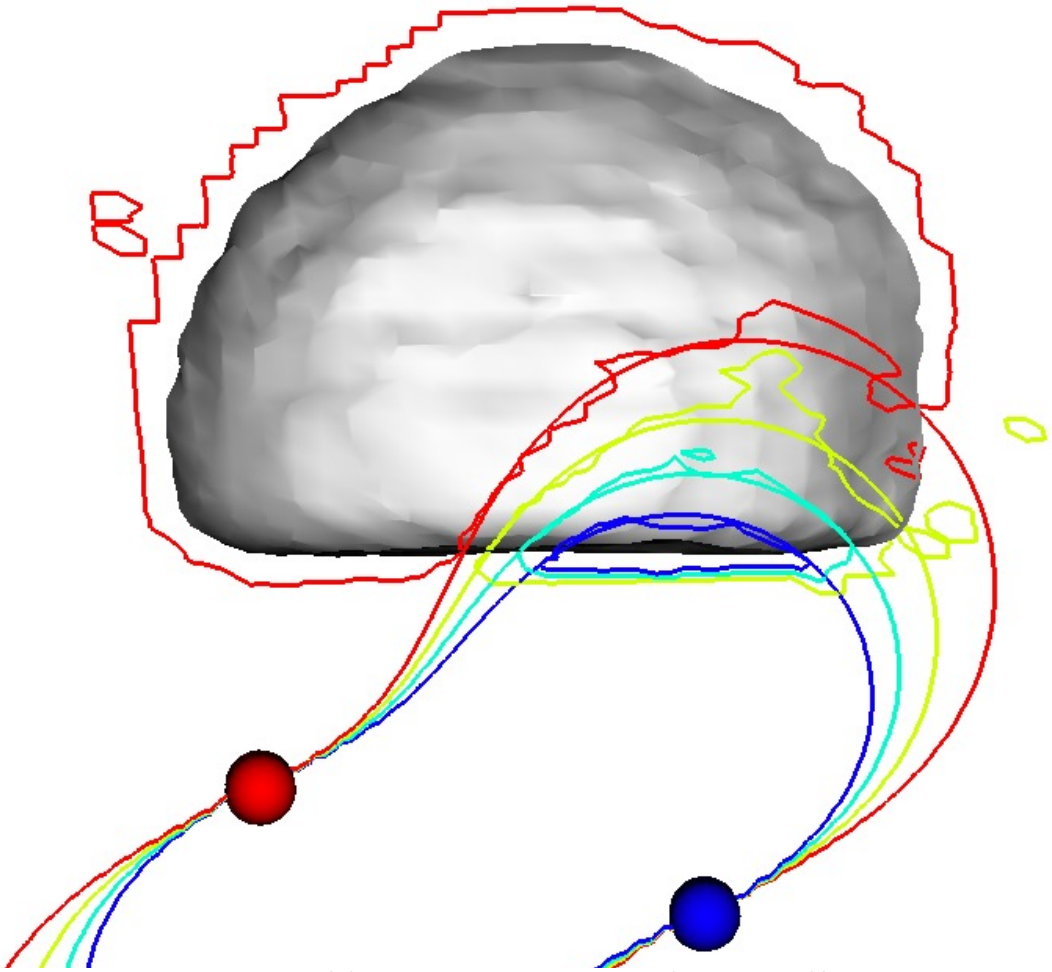
Phase structure near substrate interface

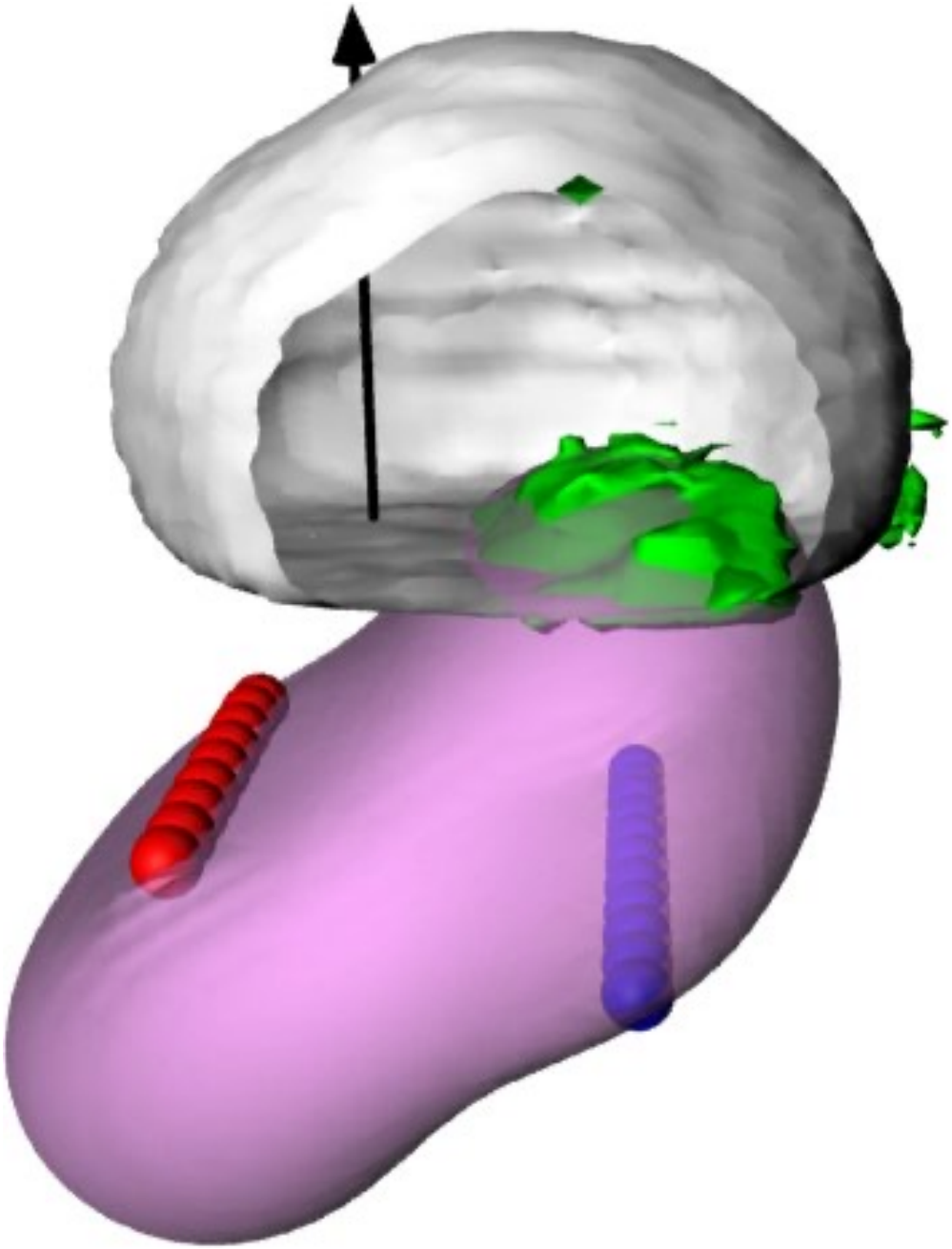


Modeling of 3D Phase Bump



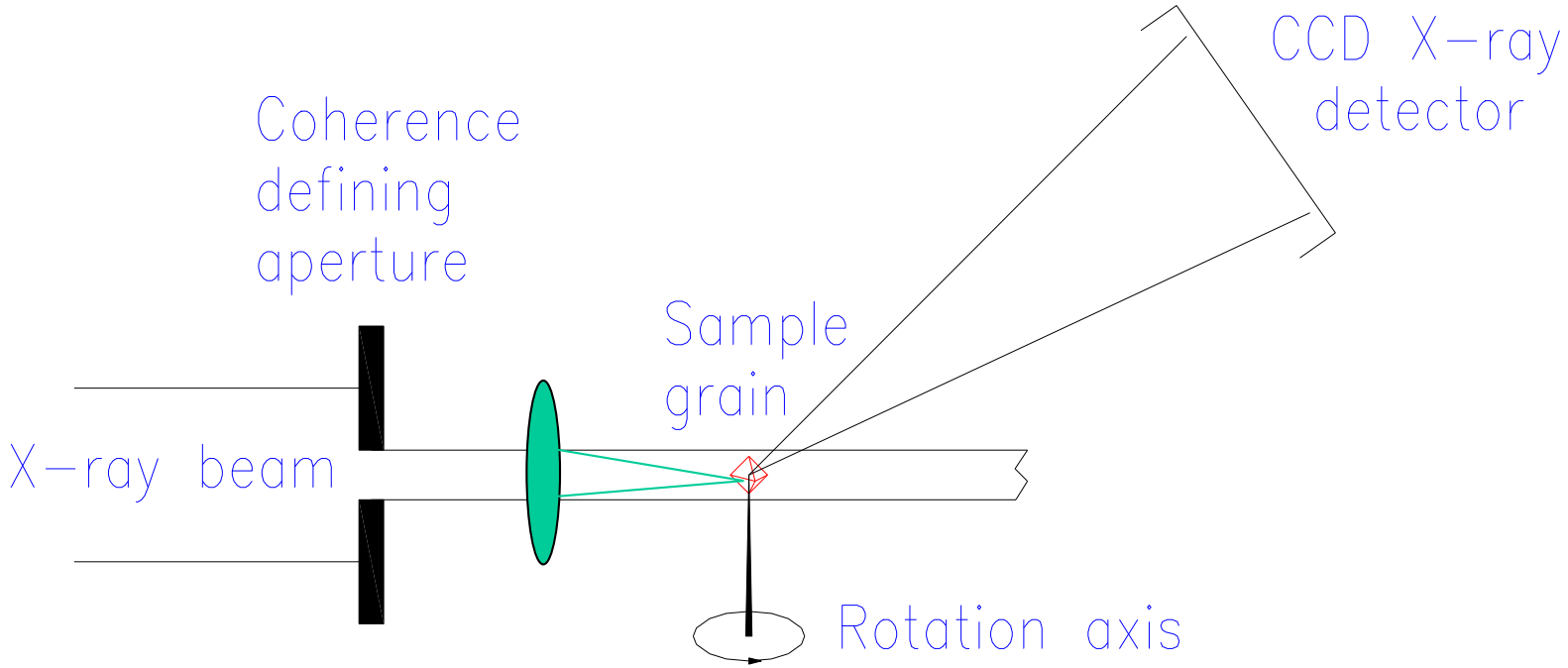
Field lines of Point Charges



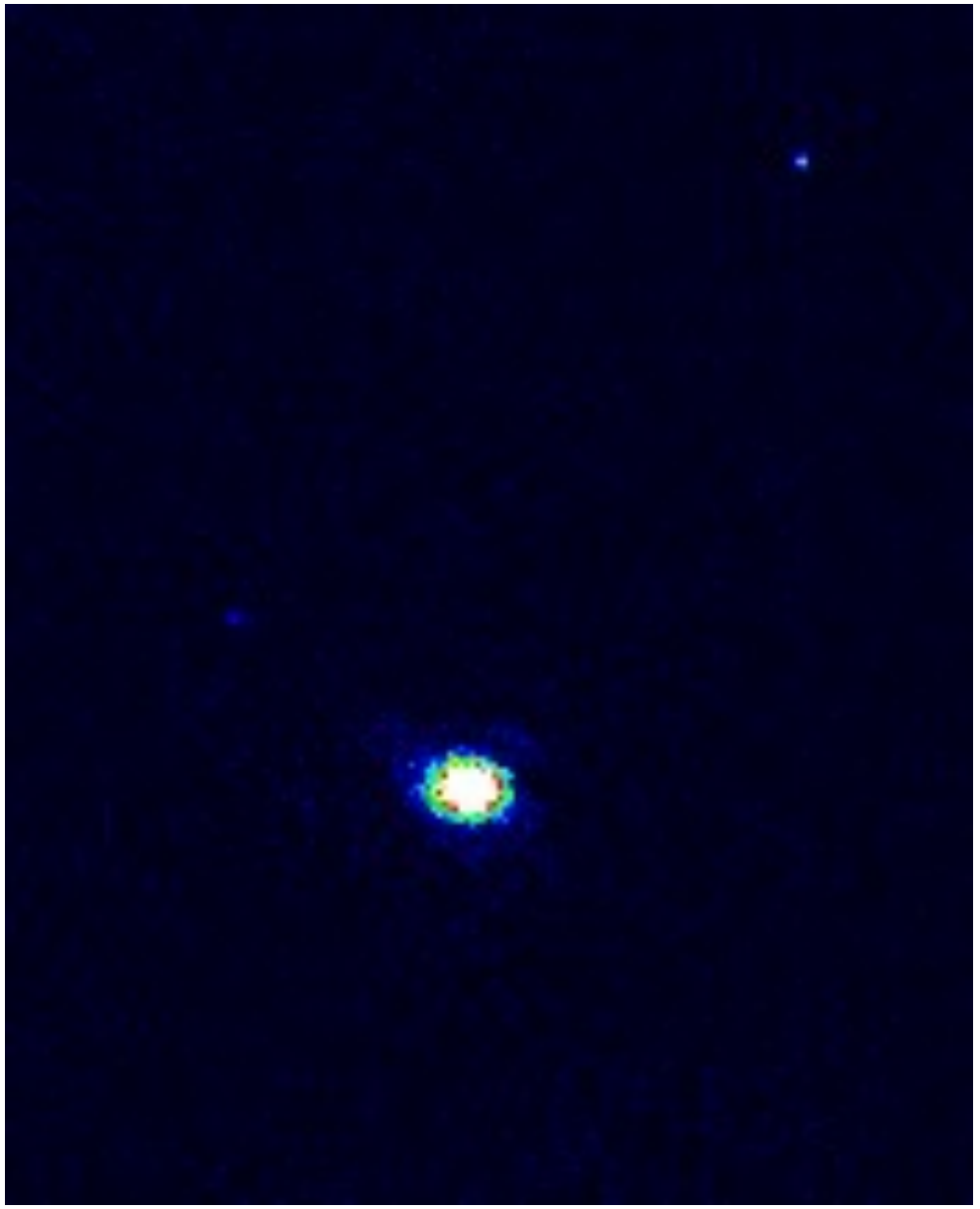


Future: Optimise using Optics

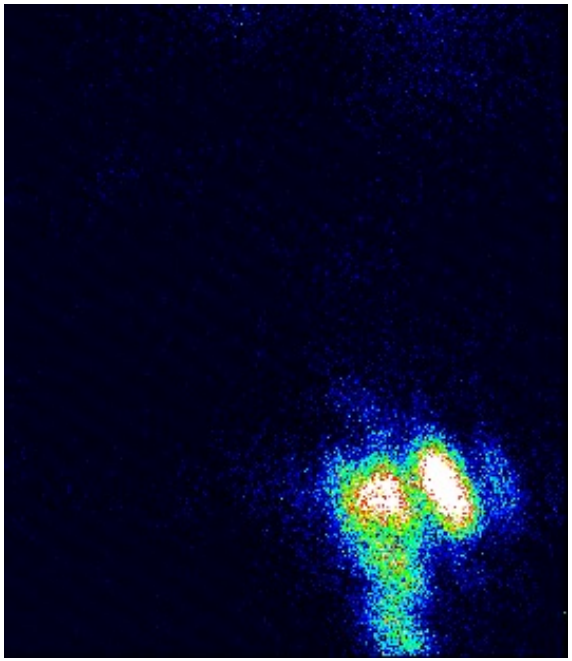
match available coherence to sample size



Rocking scan of Ag cubes with 0.01° steps



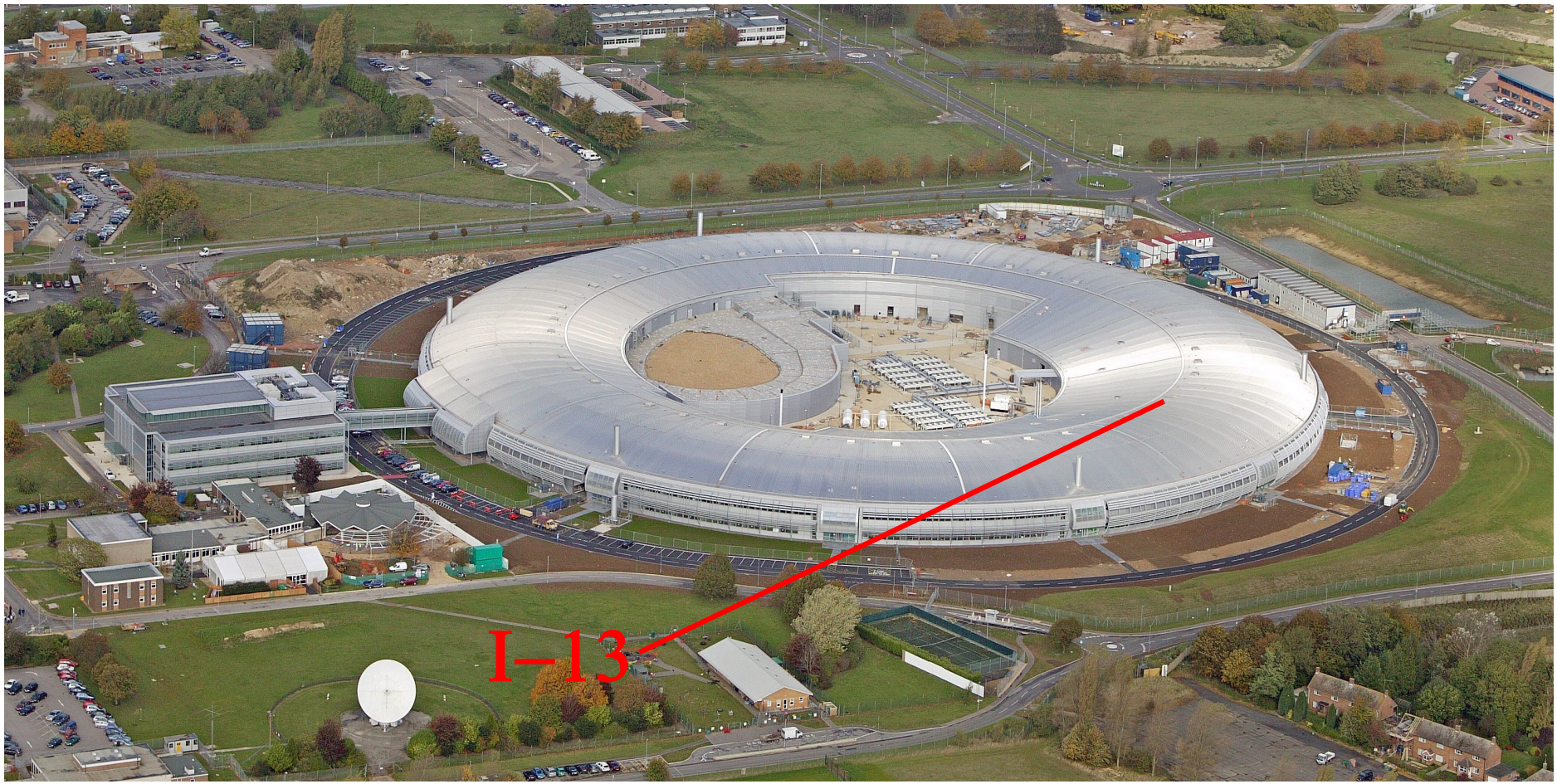
Move beam on to sample....



New Beamline Opportunities

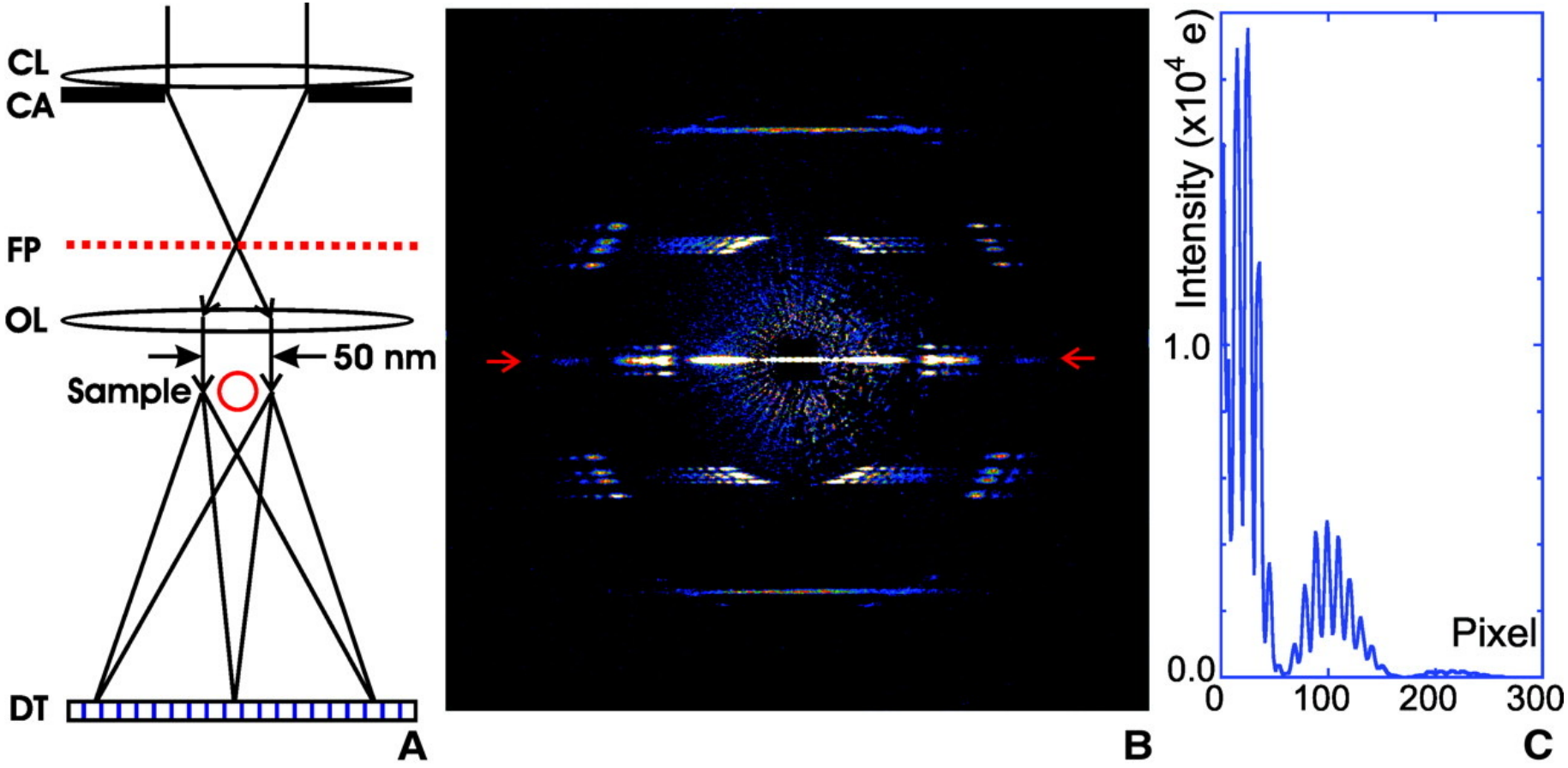
- I-13 phase 2 “Coherence” beamline at Diamond
- Principal Beamline Scientist in Aug 2006
- First Users in 2010-11
- Three branches on canted undulators:
  - USAXS and XPCS
  - Coherent Diffraction, lensless imaging
  - Full-field Imaging with hard X-rays

Diamond Light Source, Oct 2005



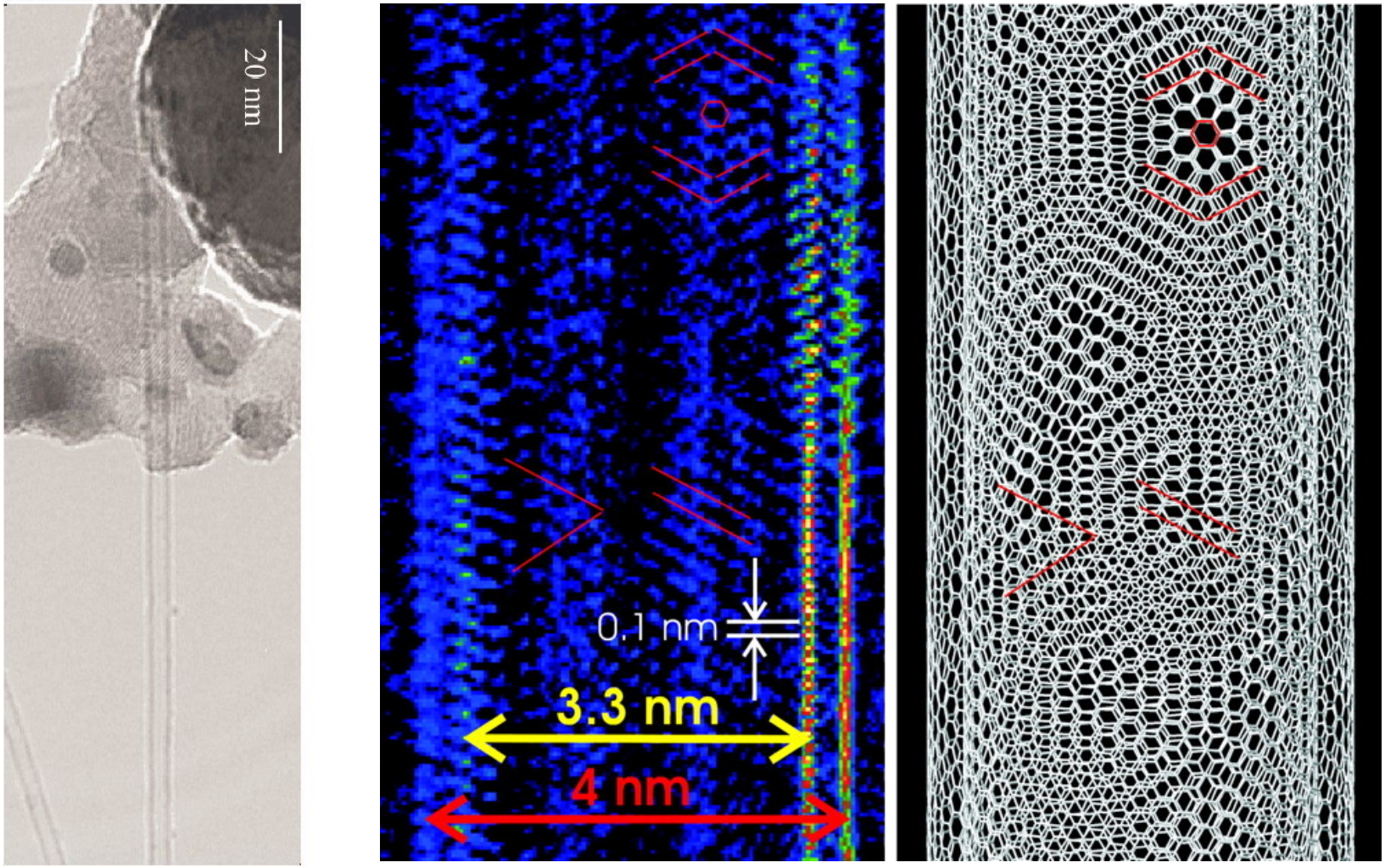
Electron Diffraction from C-nanotube

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

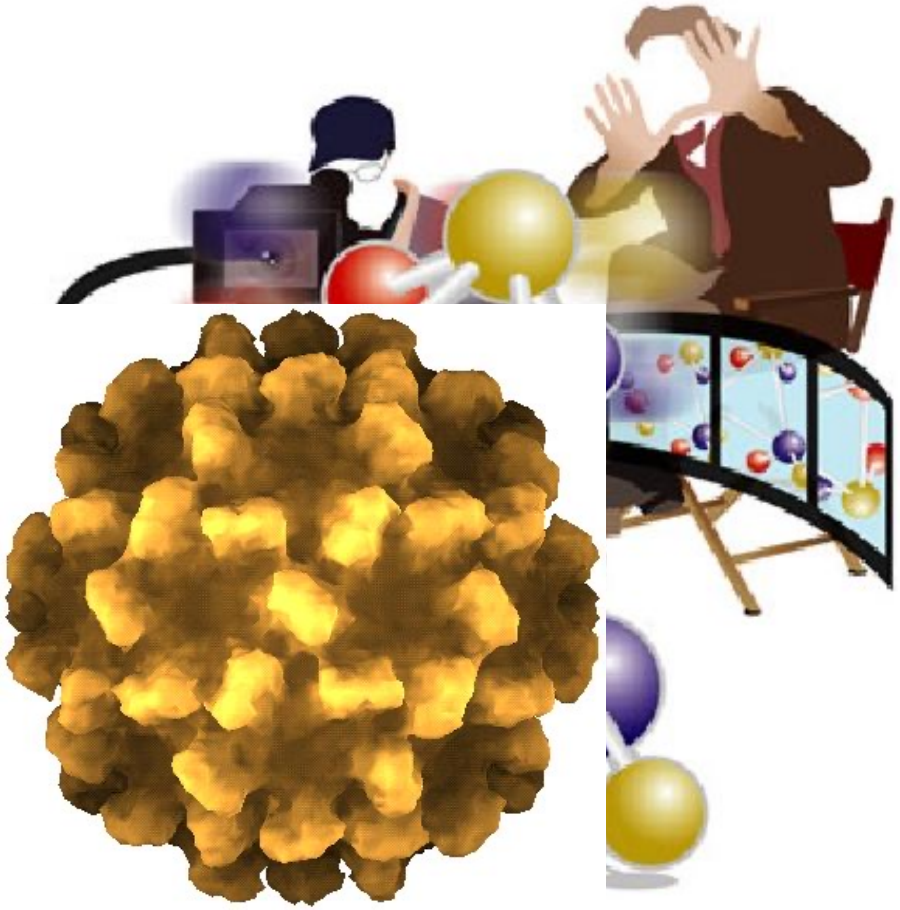


Images of DW C-nanotube

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