Inversion methods for Imaging by Coherent X-ray Diffraction

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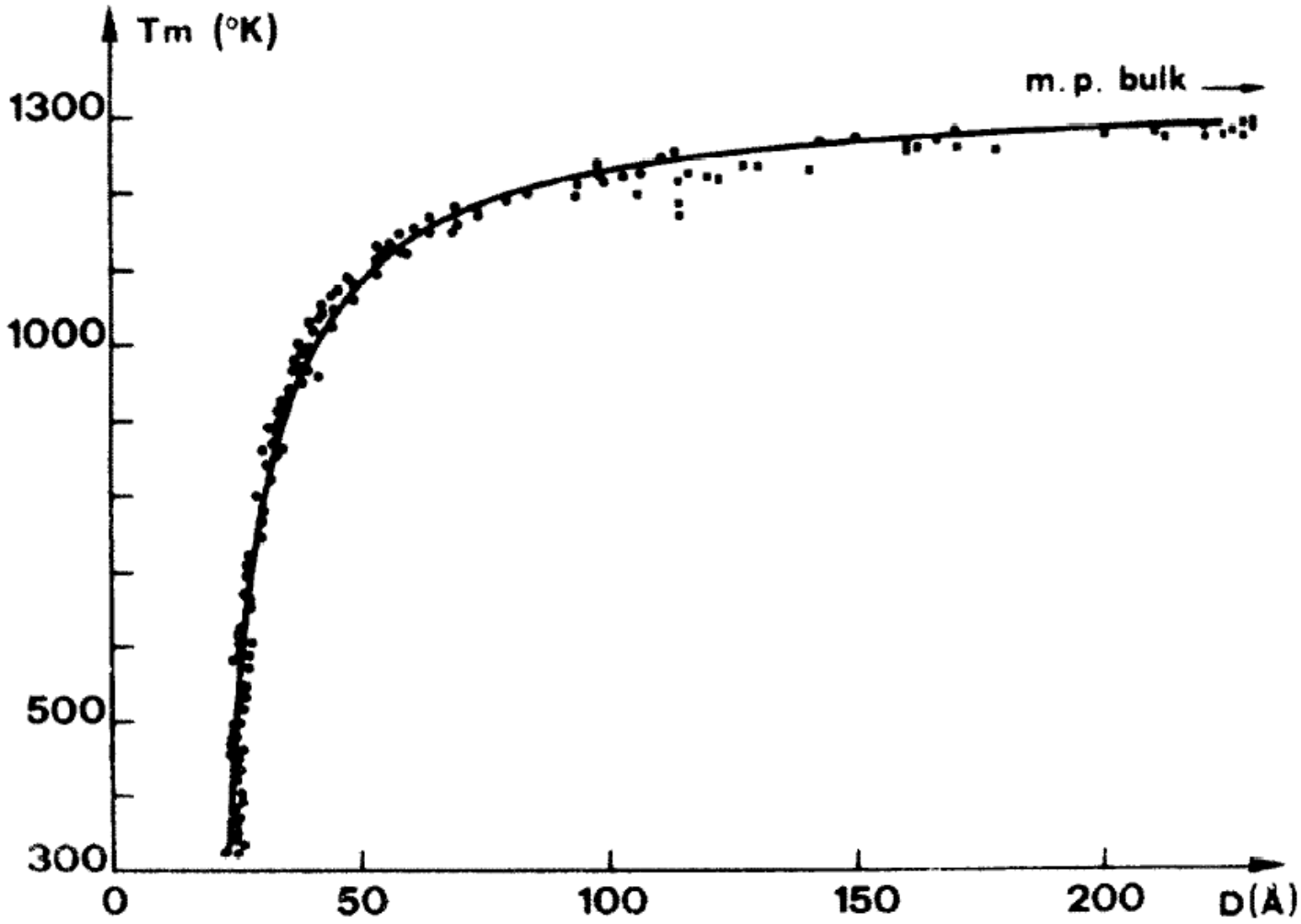
Outline

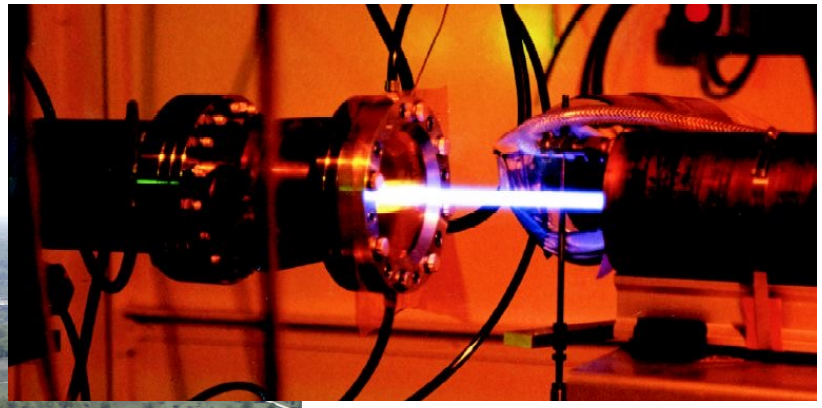
- Coherent X-ray Diffraction
- How to Solve the Phase Problem
- Nanocrystal Shapes
- Extension to Phase Objects
- Opportunities with Electrons and FELs

Size-dependent Melting of Au Particles

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



Synchrotron Radiation

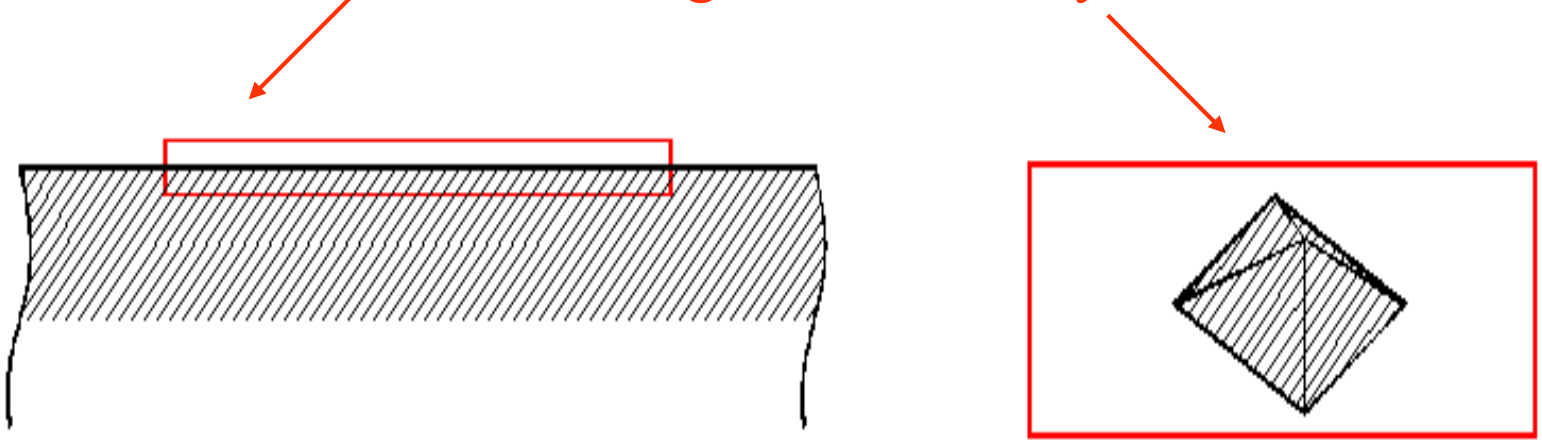


Coherence at APS, ESRF or DLS

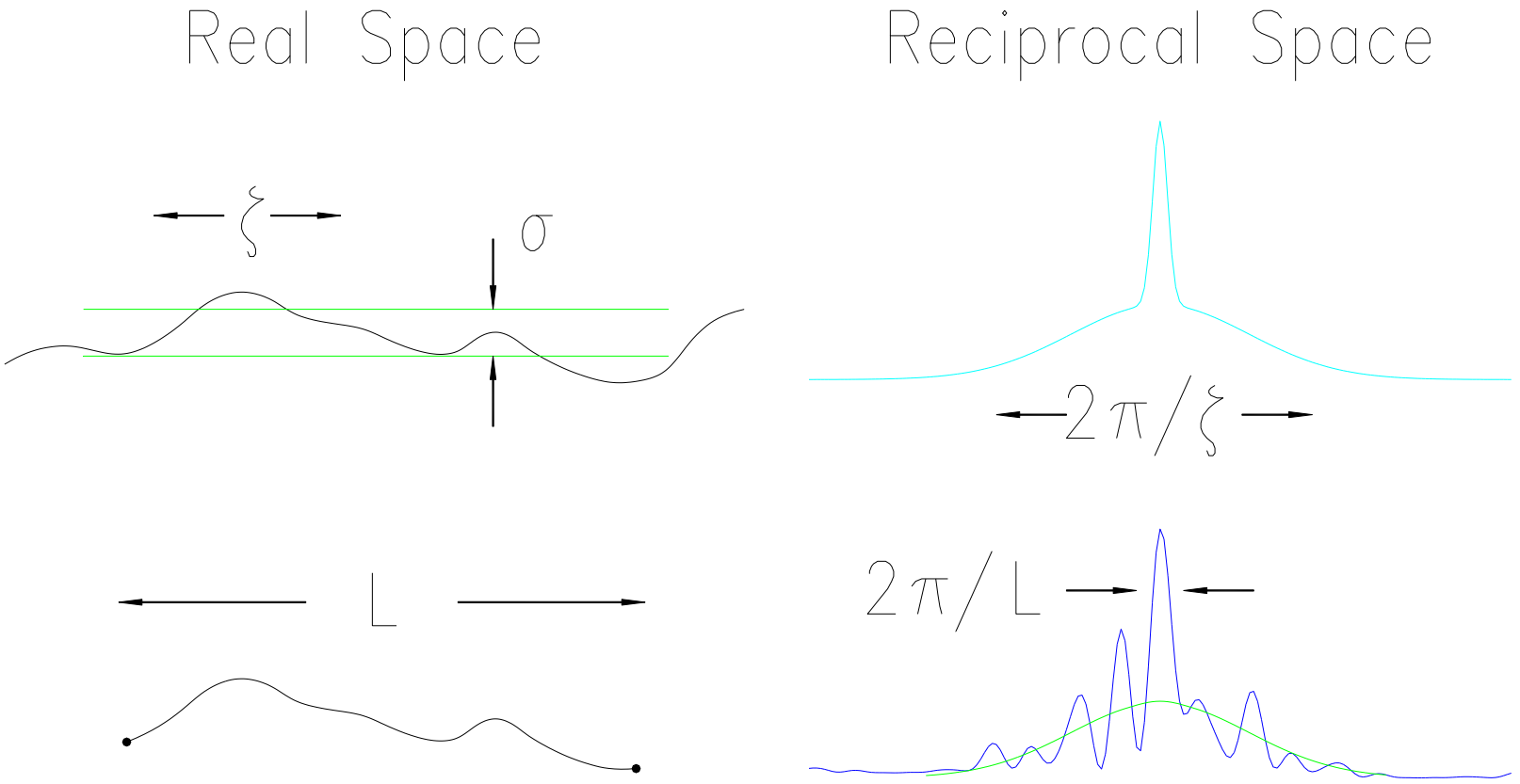
Typical of 3rd Generation (undulator) Synchrotron Source

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}

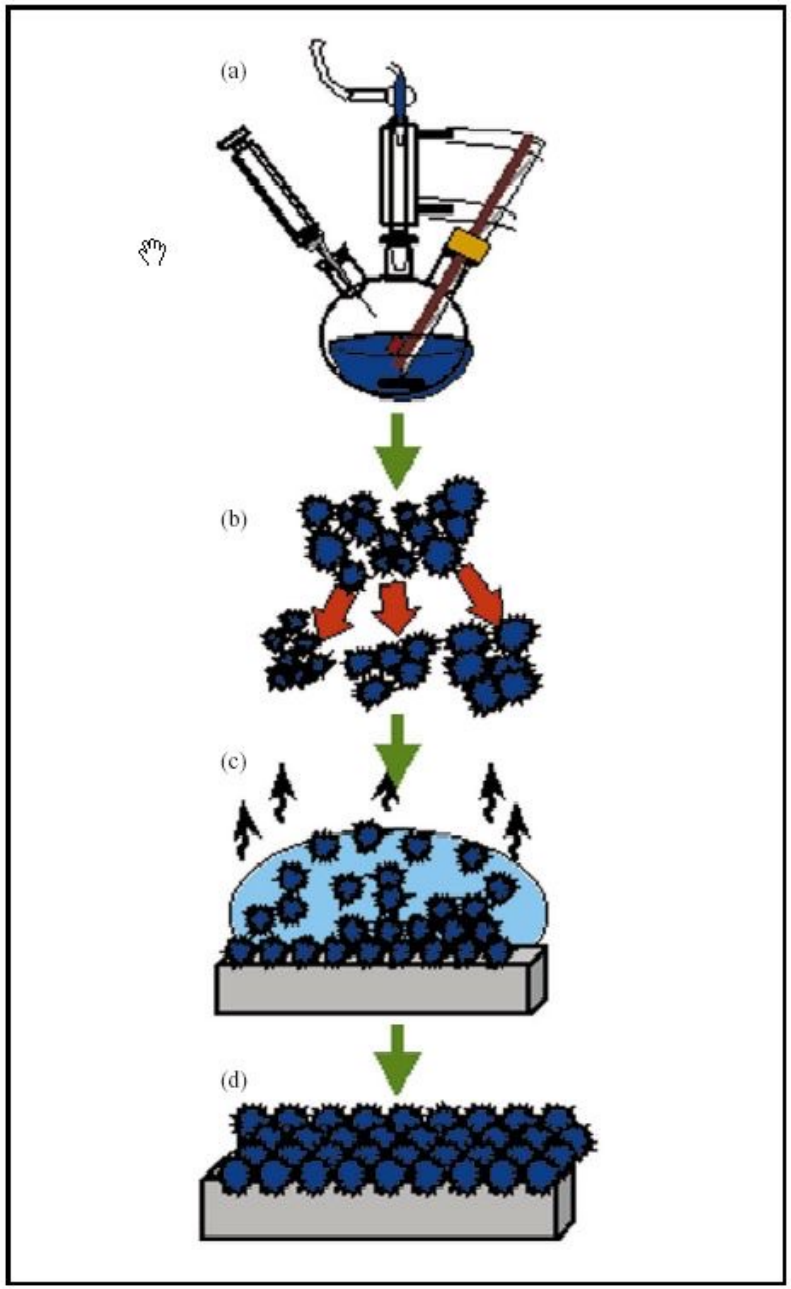
Coherent region defined by slits



Diffuse Scattering acquires Structure using CXD

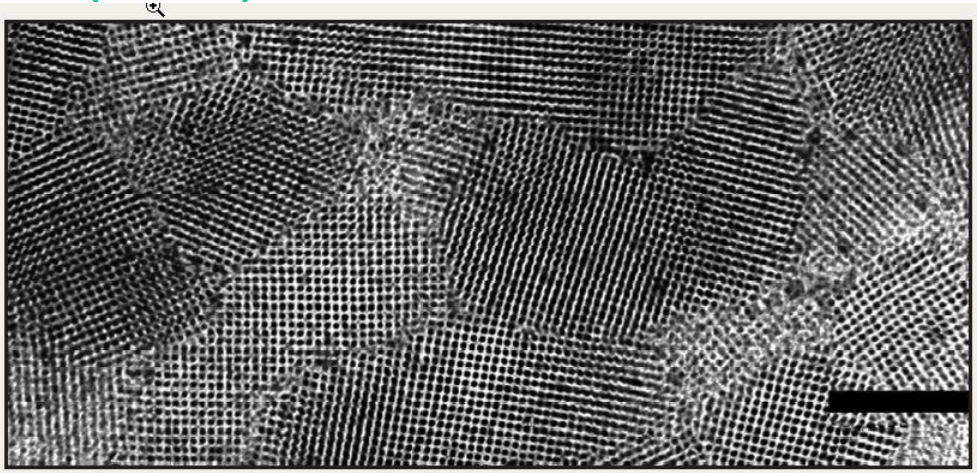


Chemical Synthesis of Nanocrystals



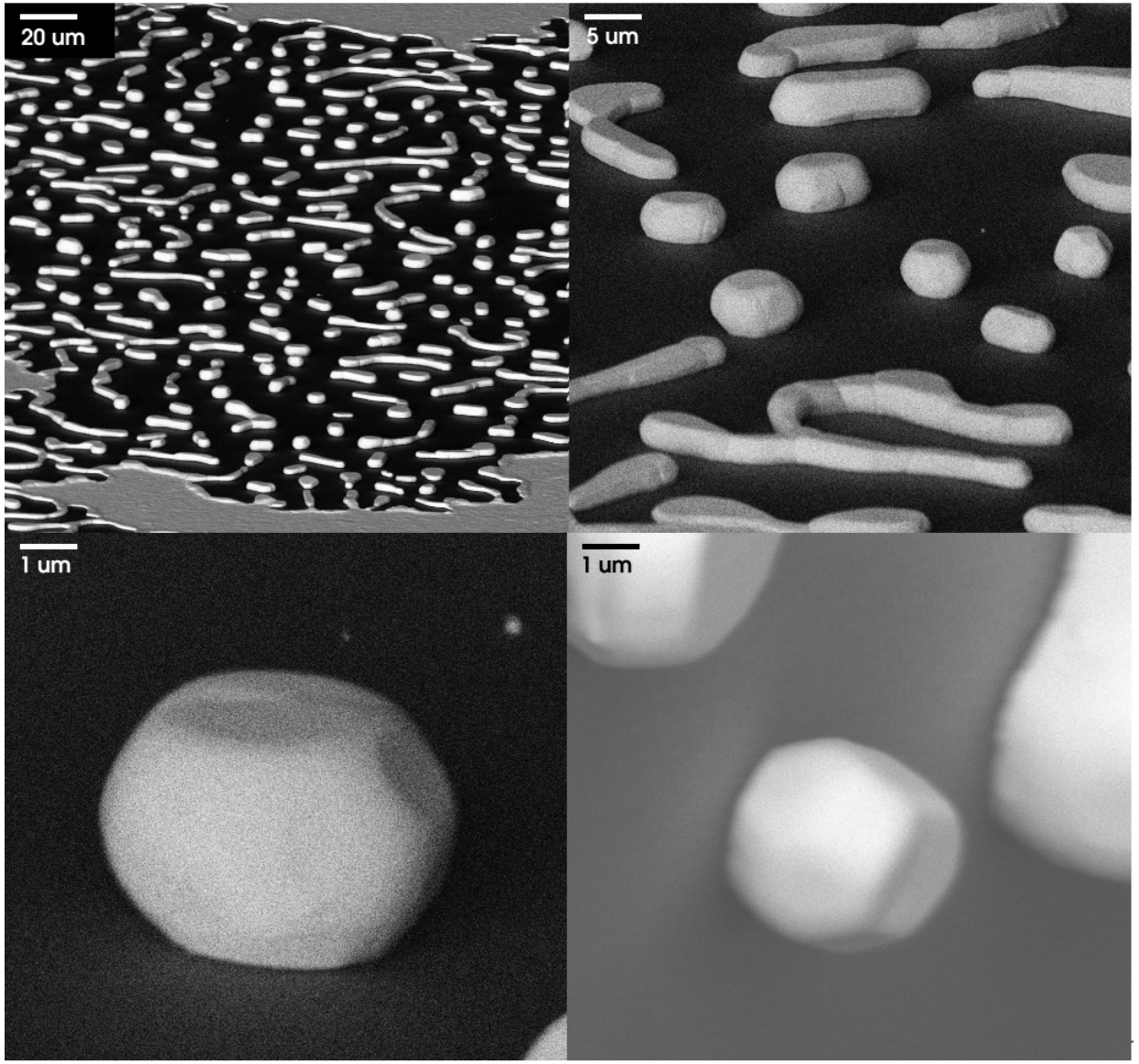
- Reactants introduced rapidly
- High temperature solvent
- Surfactant/organic capping agent
- Square superlattice (200nm scale)

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

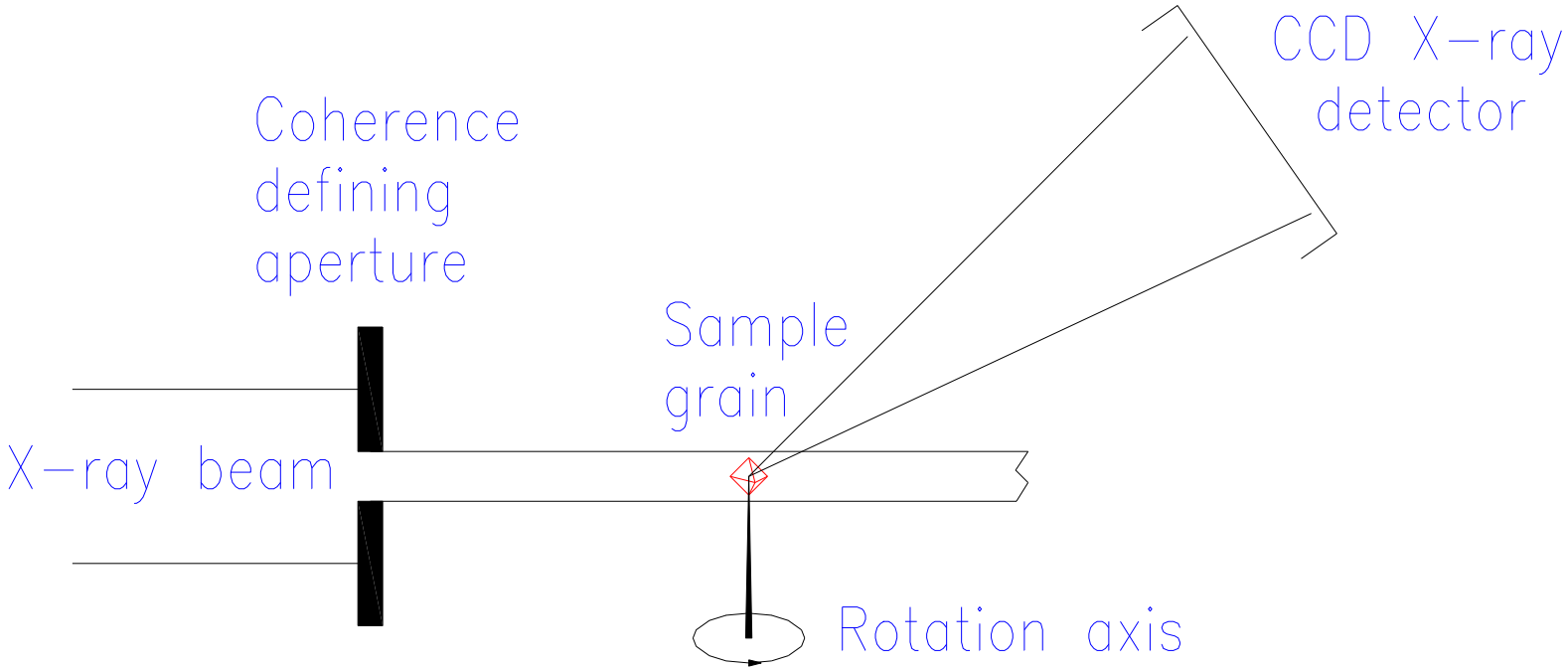


SEMS

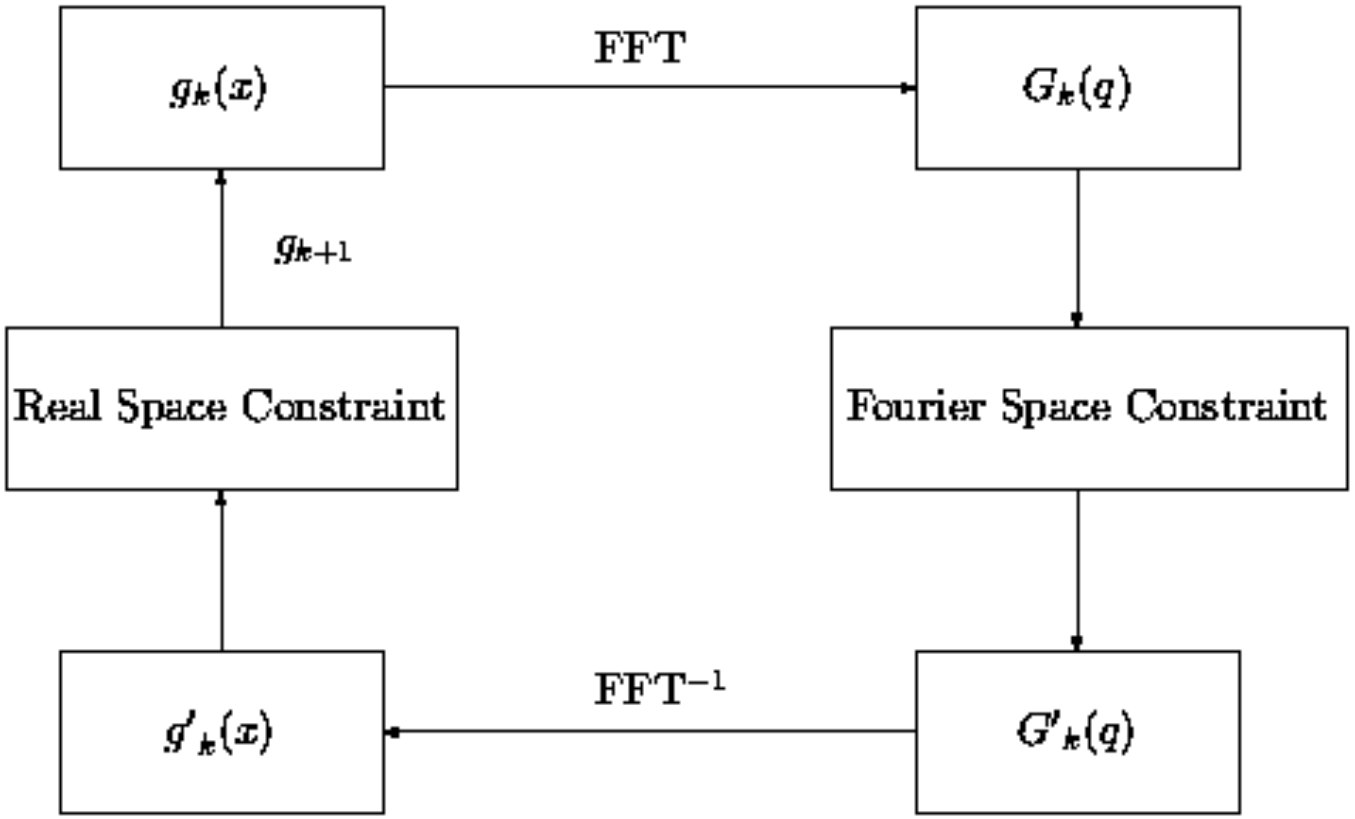
- Au blanket film
- Quartz substrate
- Annealed at 950°C for 70 hrs.



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)

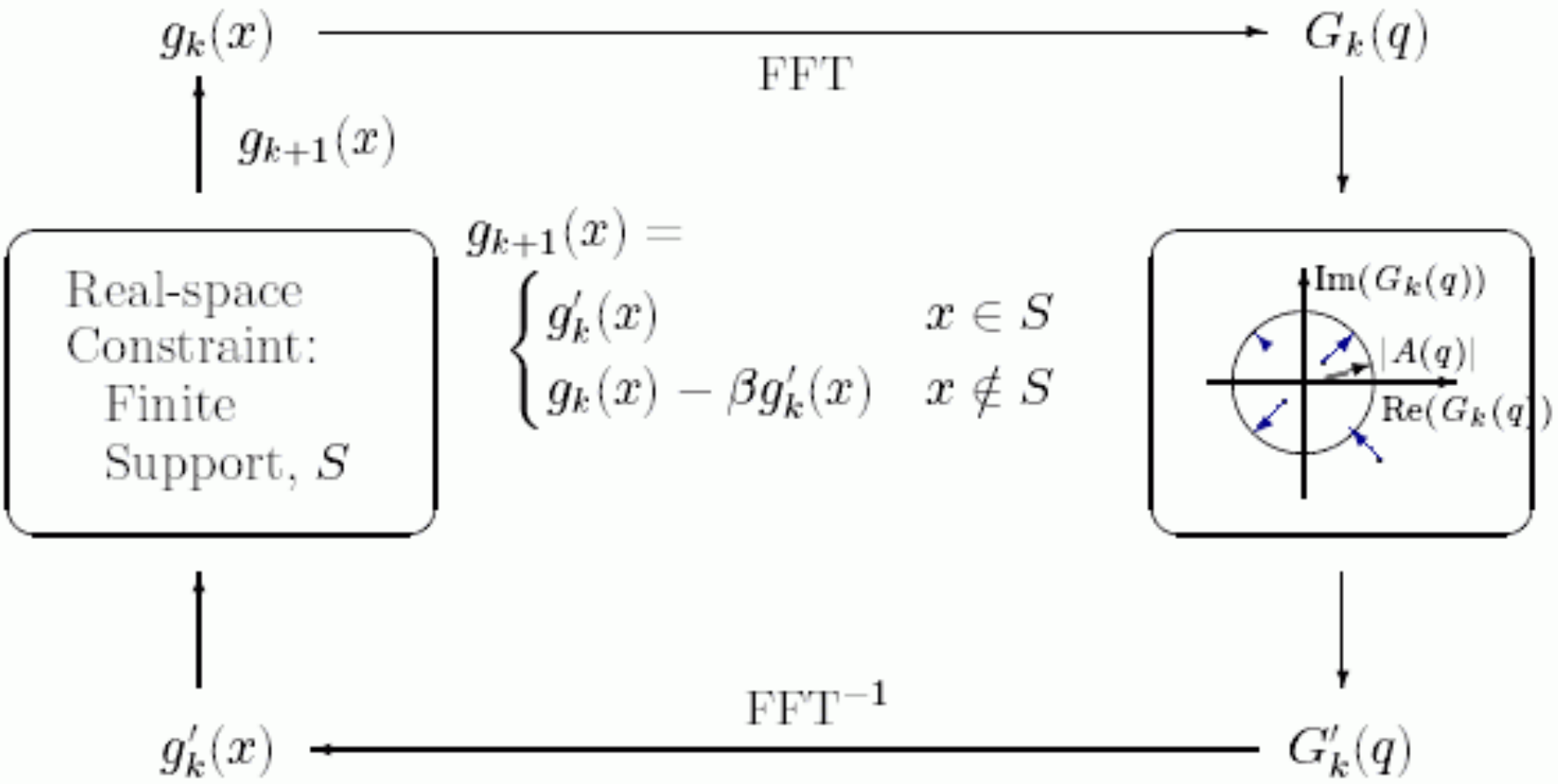
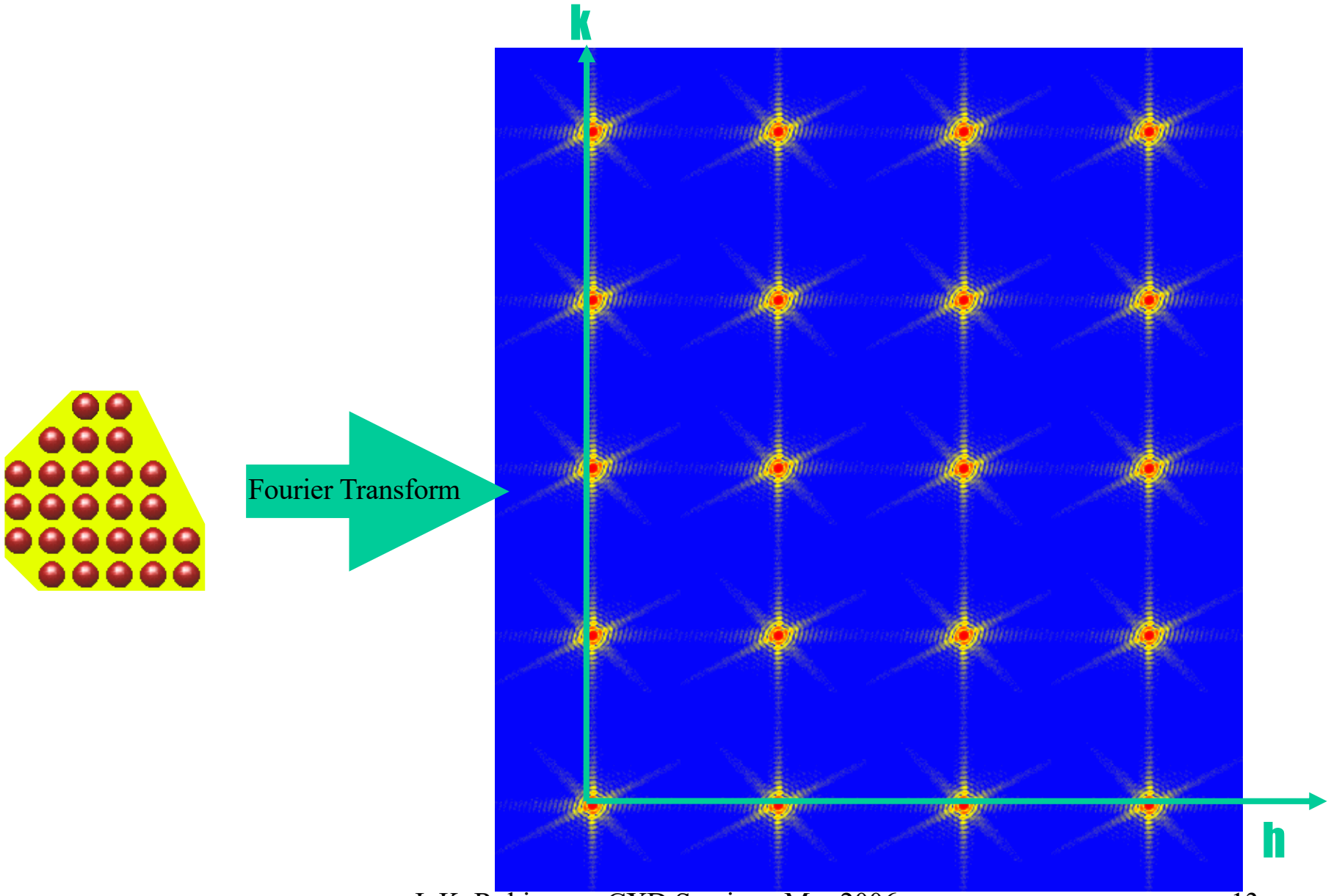
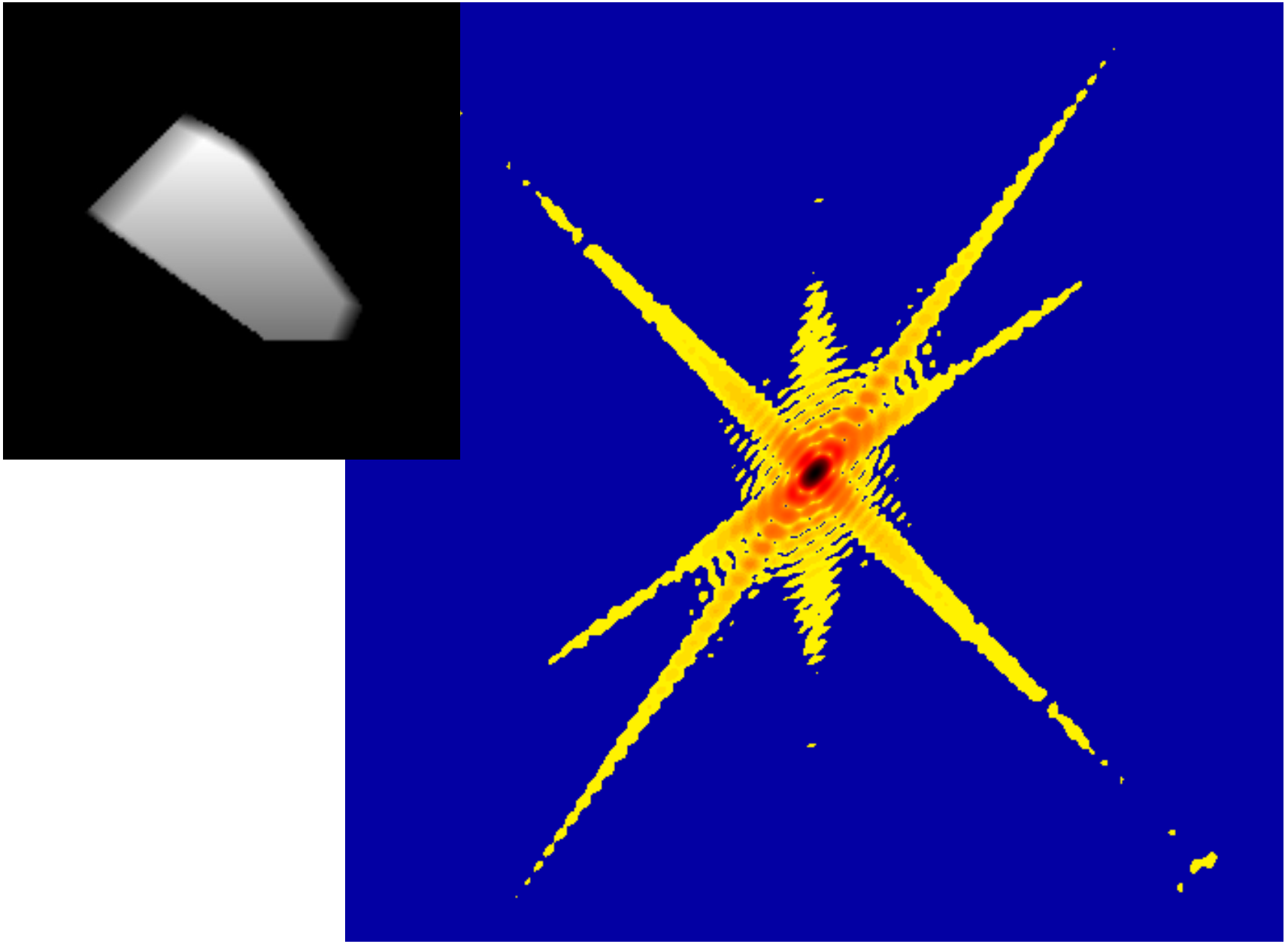


Figure 2.3: Fienup HIO

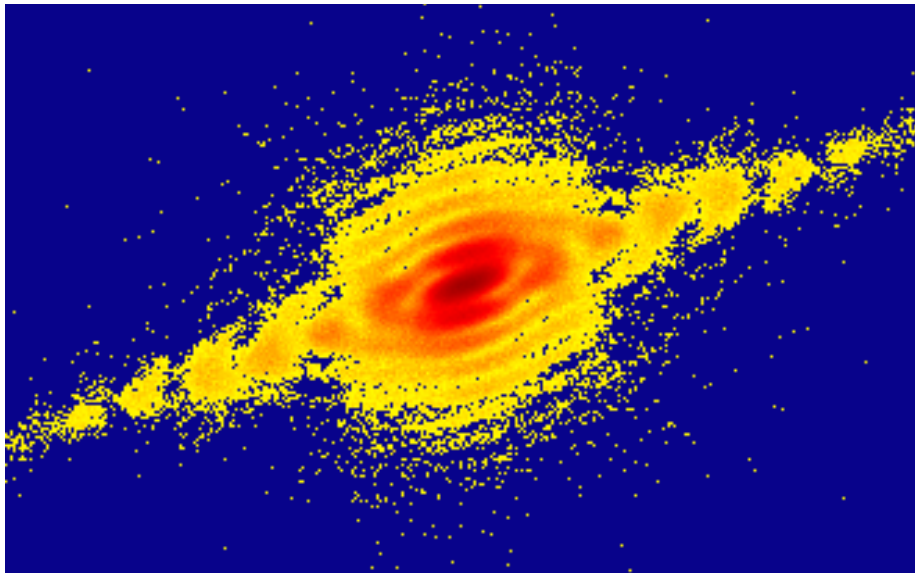
Coherent Diffraction from Crystals

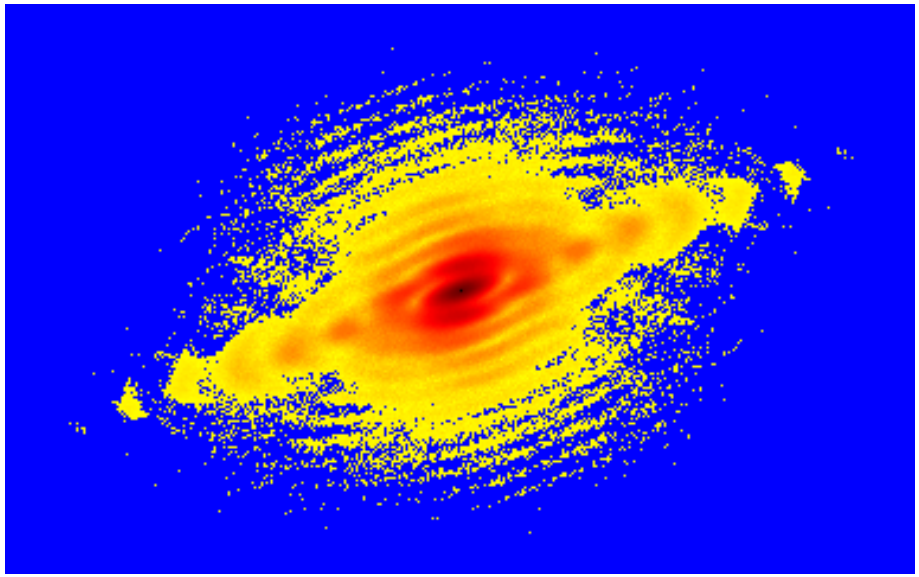


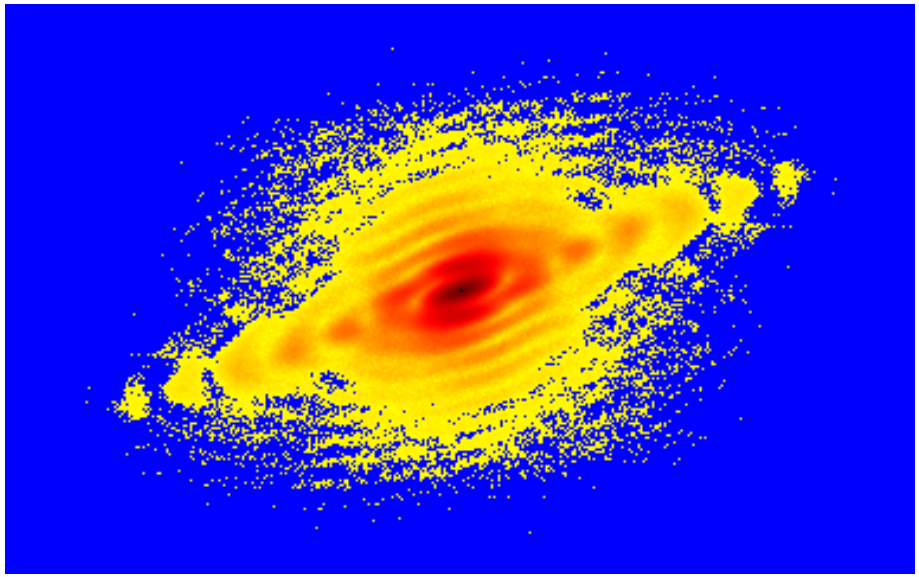


Symmetrized Data and two best fits

Chisq=0.0005







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

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

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

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

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

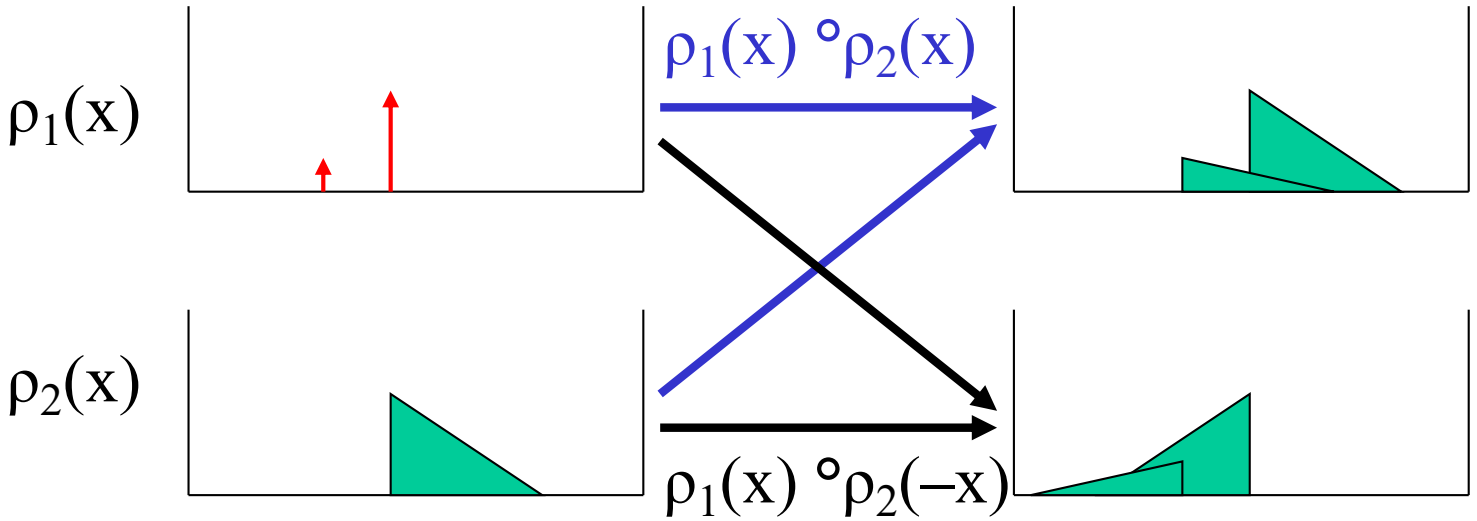
$$I(Q) = |\int \rho_1(-x) \circ \rho_2(x)e^{iqx}dx|^2$$

Homometric structures

Identical diffraction from two structures whenever the structure factor is factorizable

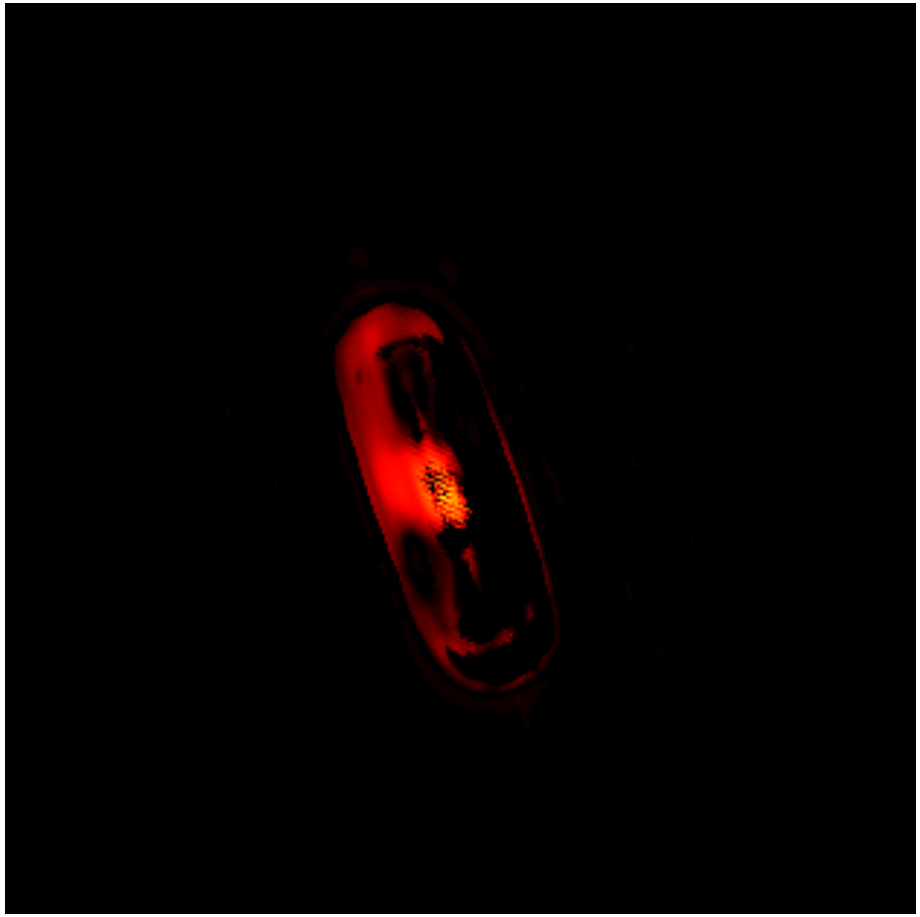
Homometric structures II

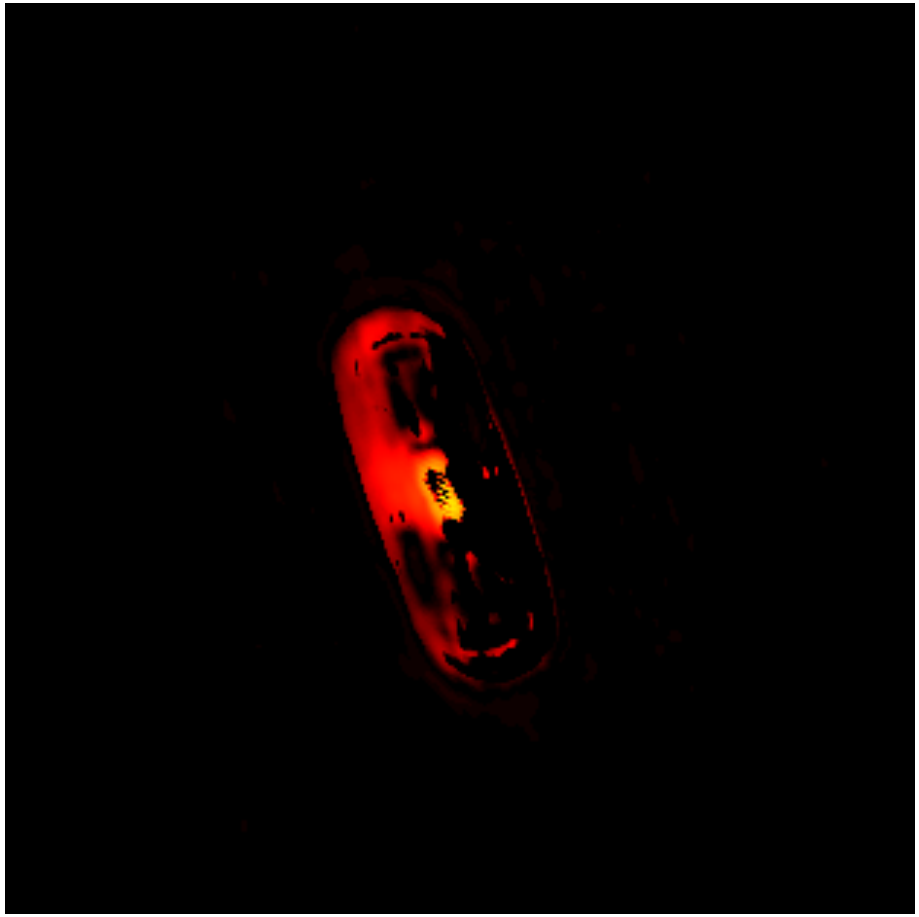
Convolution of two structures without mirror symmetry



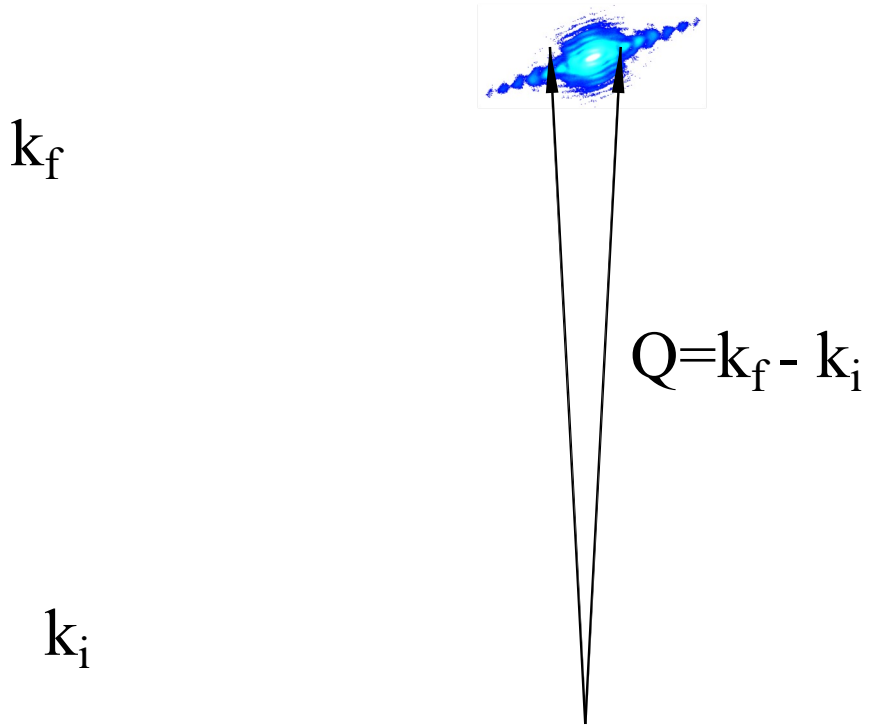
2D Reconstructions

chisquare = 0.0005

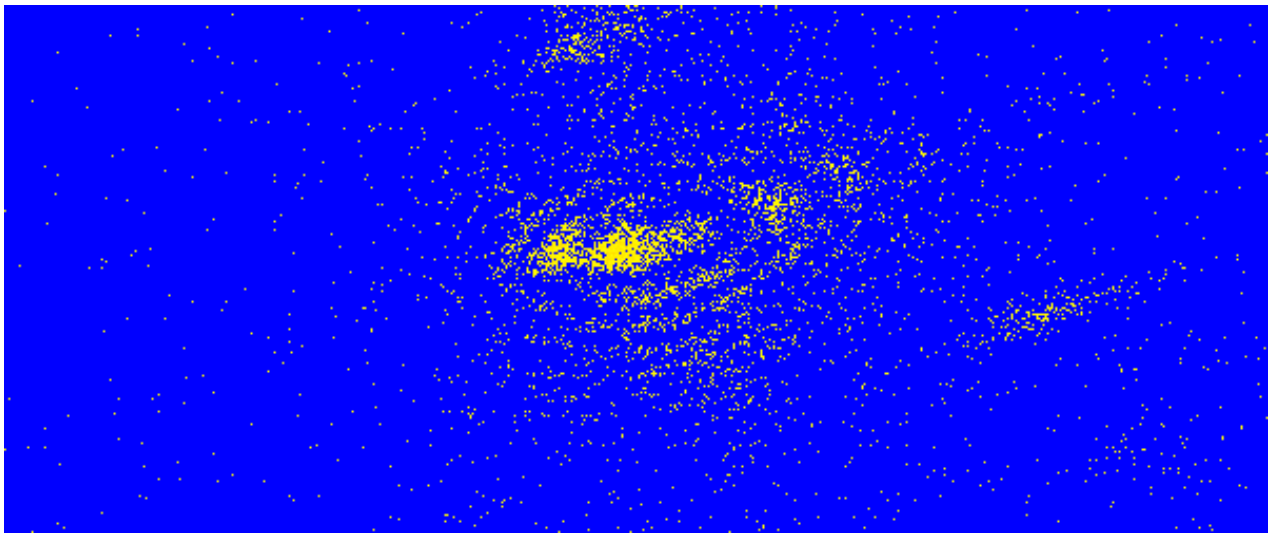


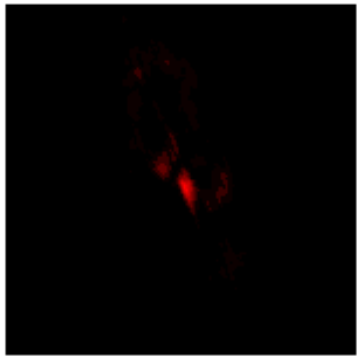
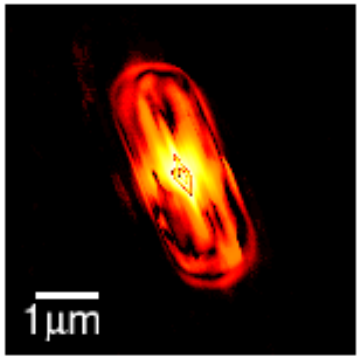


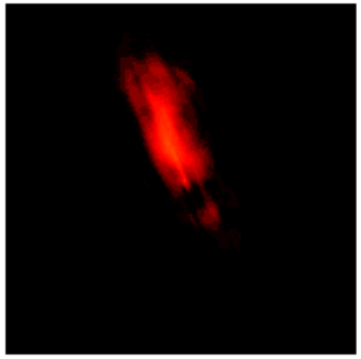
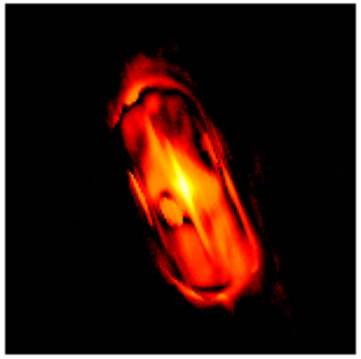
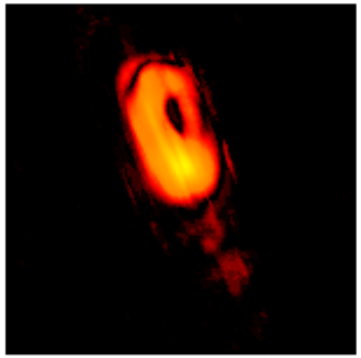
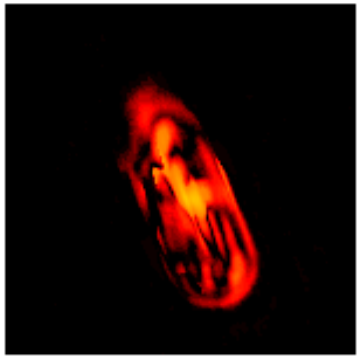
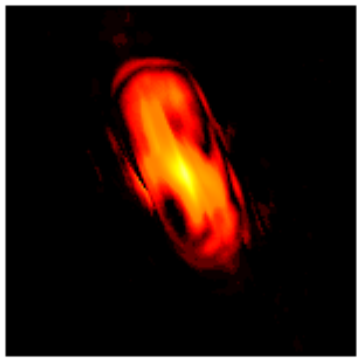
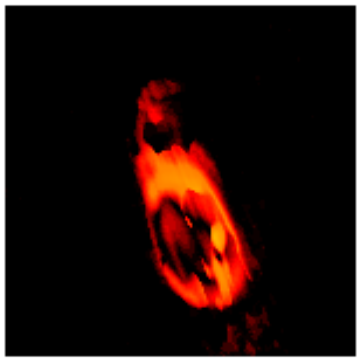
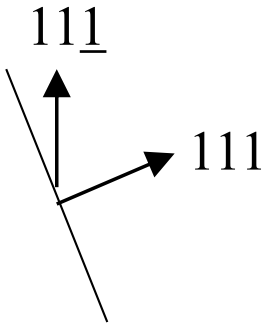
3D Diffraction Method

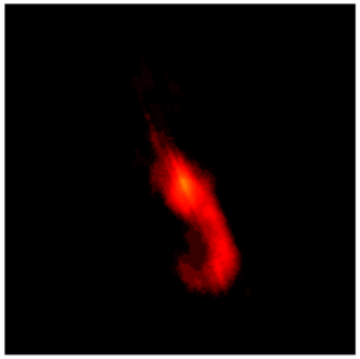
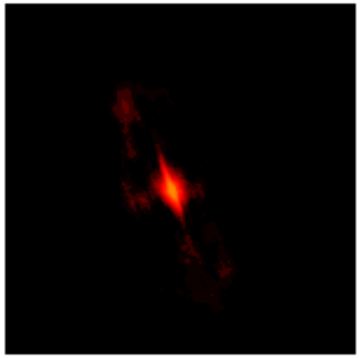


3D Diffraction Data
1 micron Au crystal

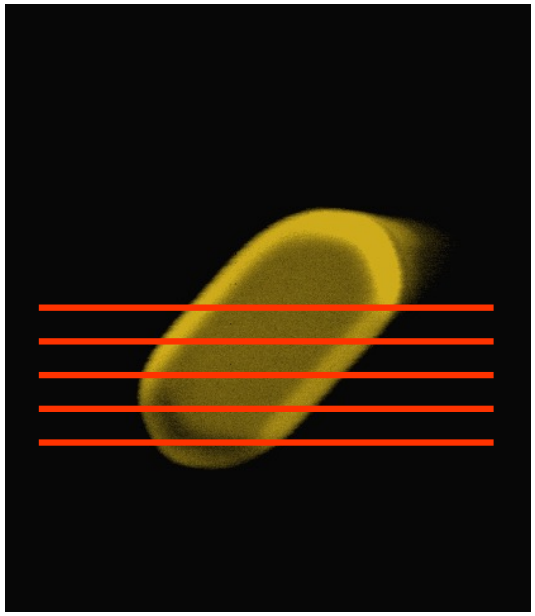


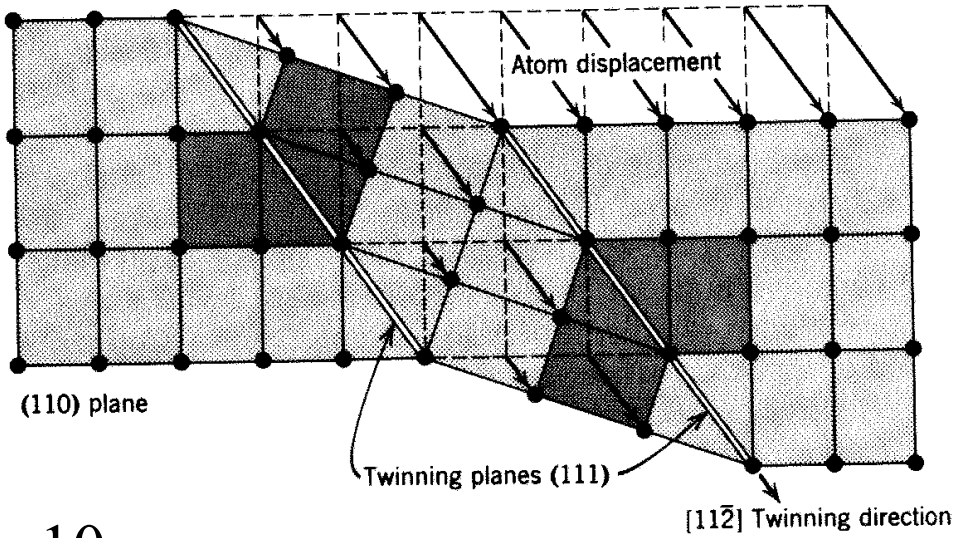



Slices through plan view SEM:



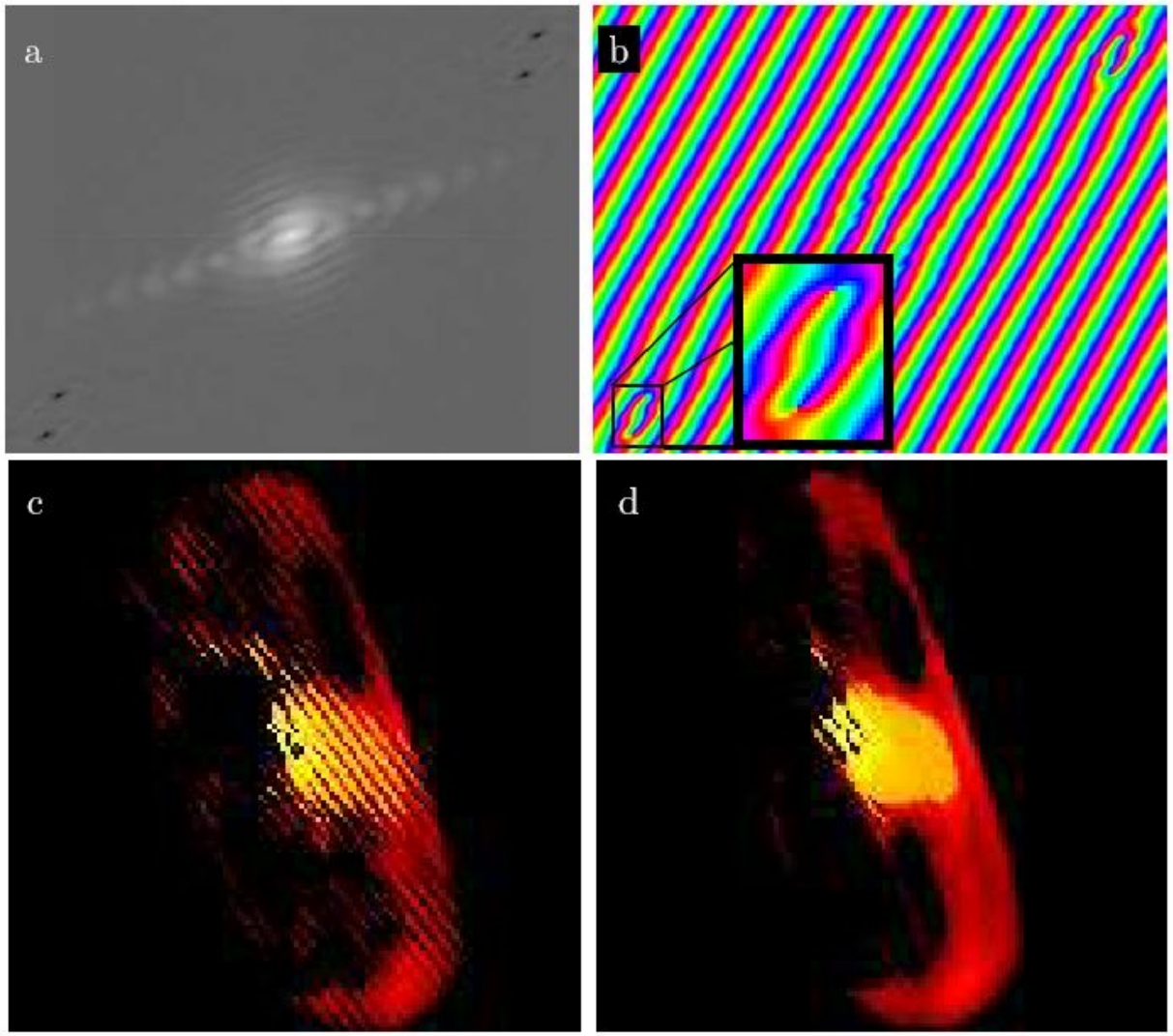
Twinning in deformed FCC metals

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

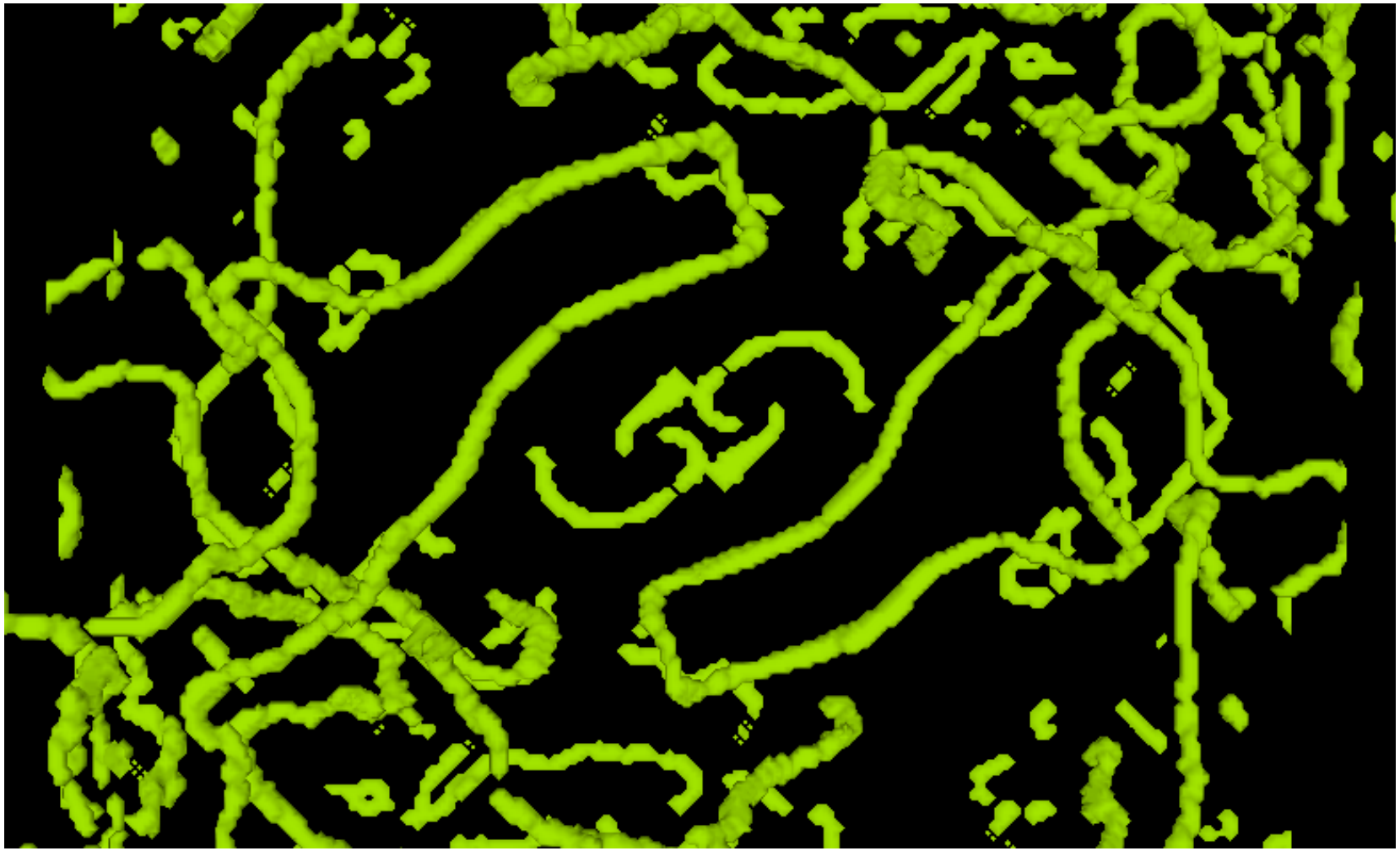


~10μm
Cu

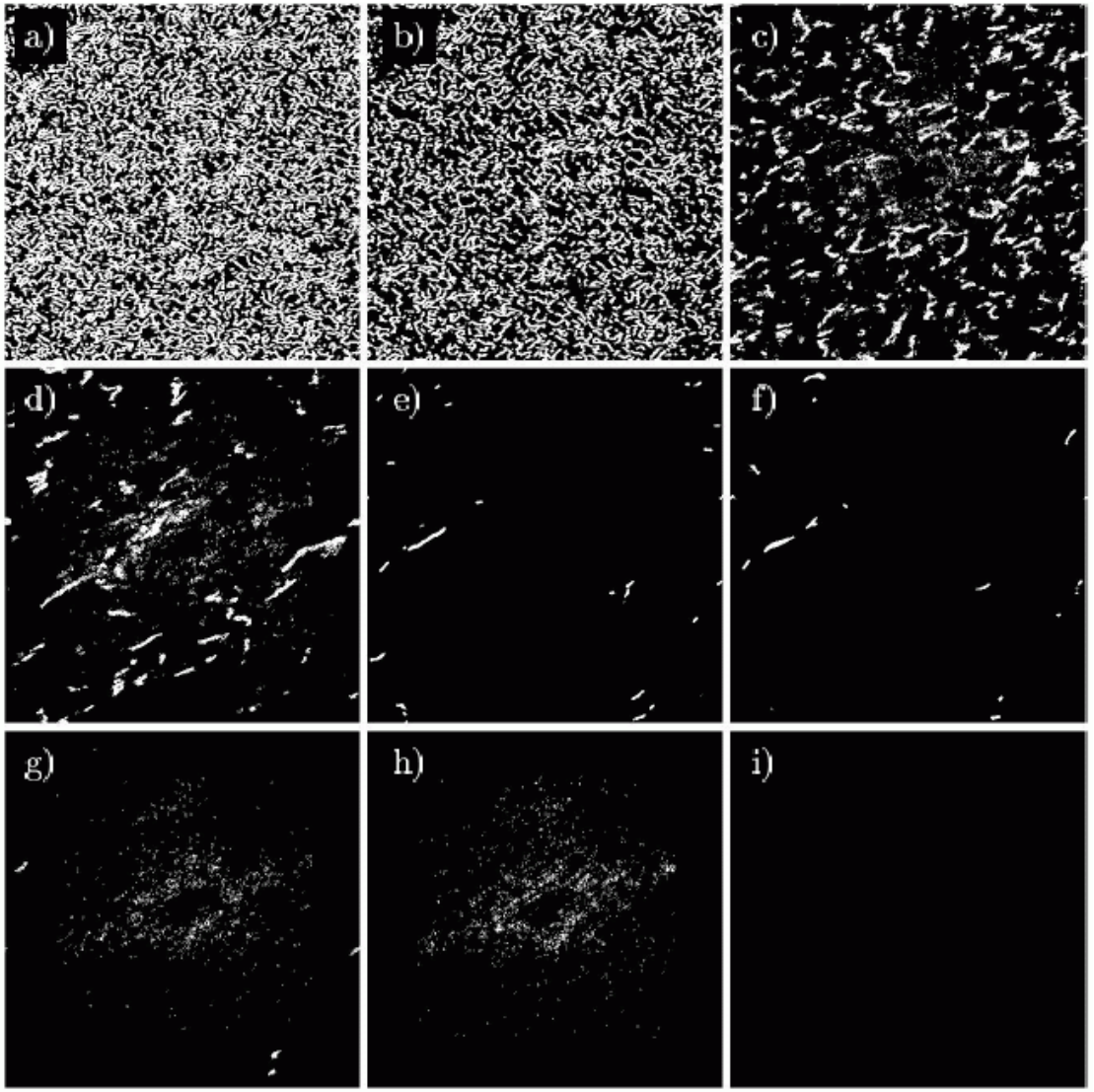
Phase Vortices and patching



‘Vortices’ Form Loops in 3D

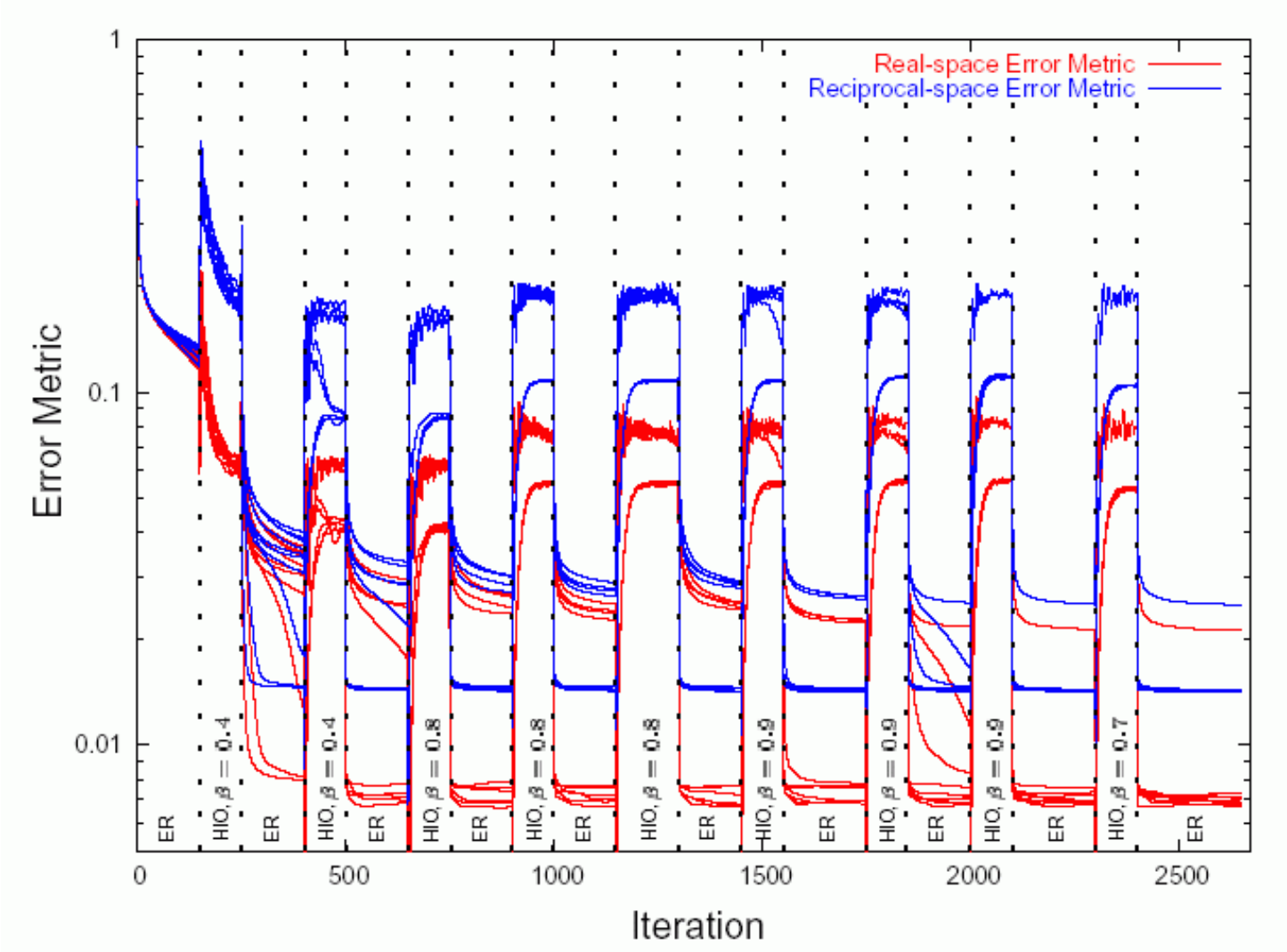


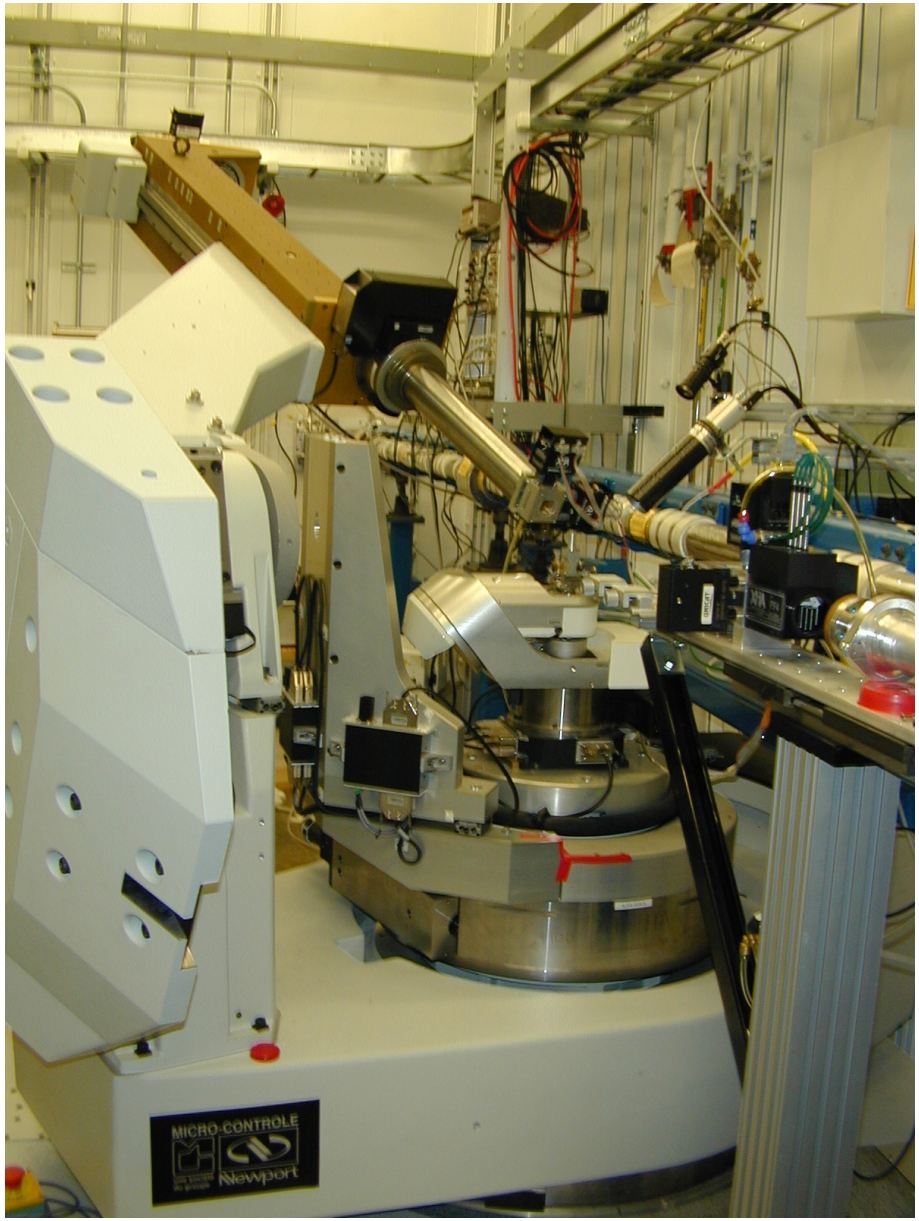
Clearing of Vortices during HIO

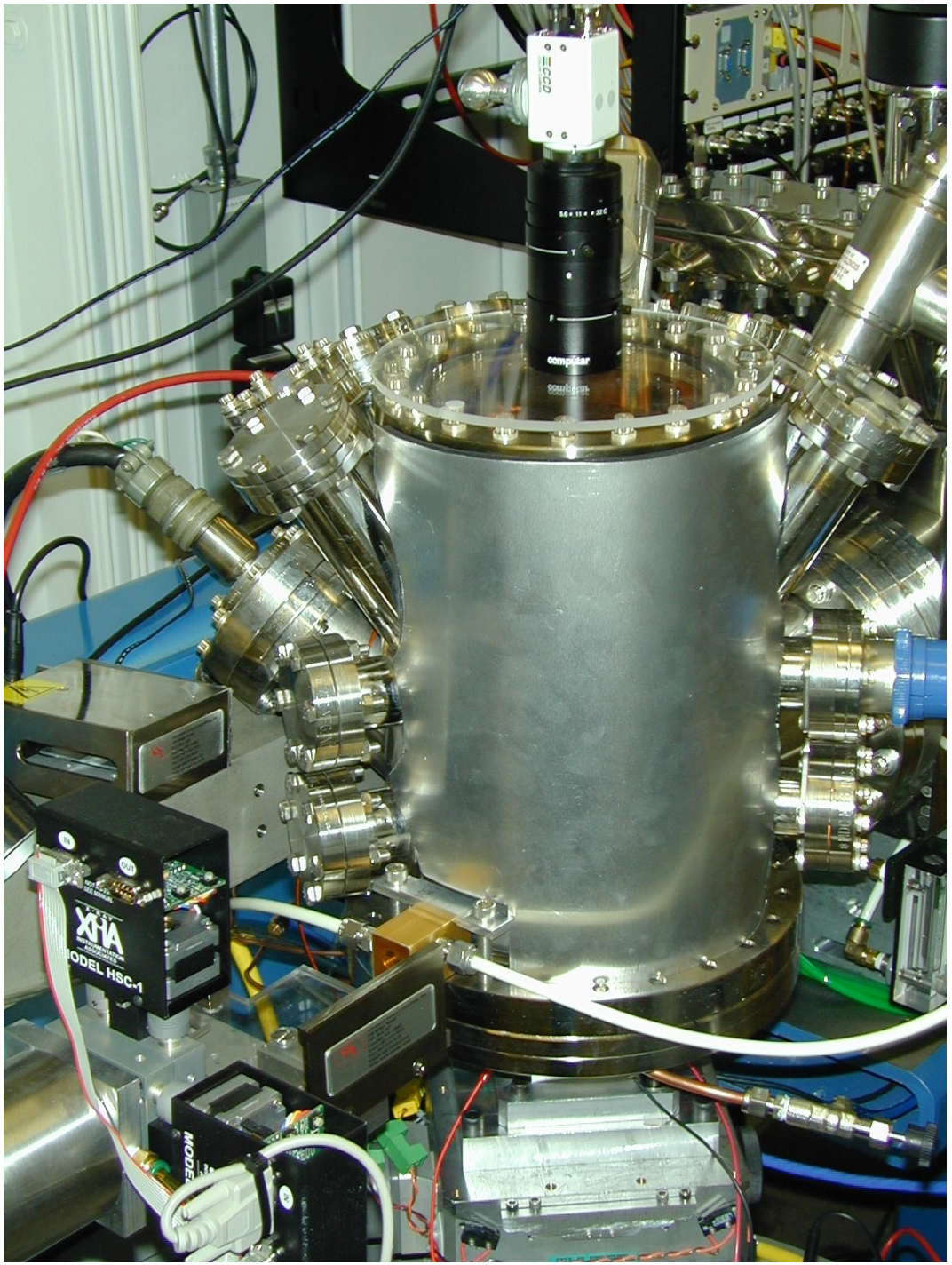


ER ER HIO
HIO ER ER
HIO HIO ER

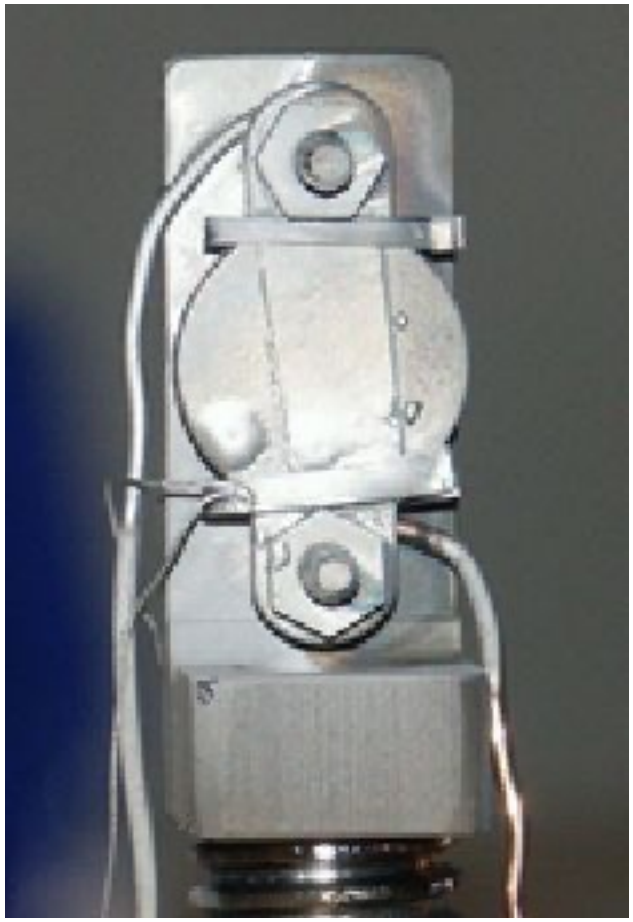
Progress of Phase Retrieval

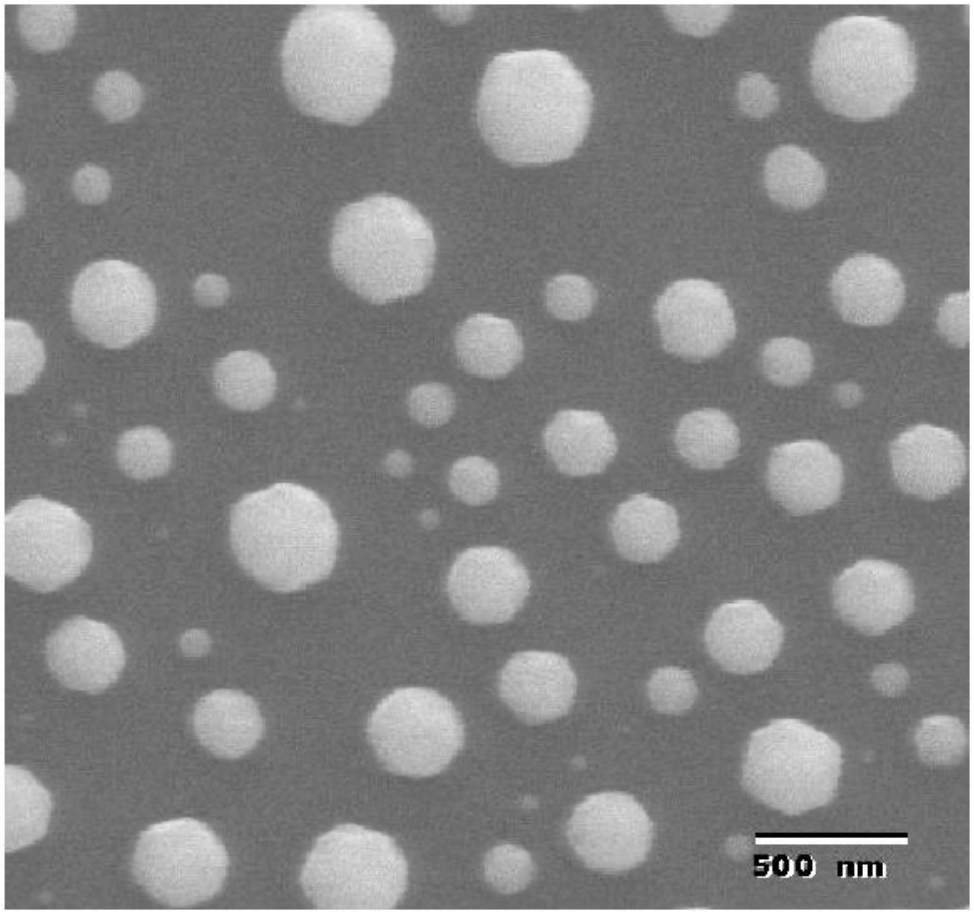



MICRO-CONTROLE
Newport

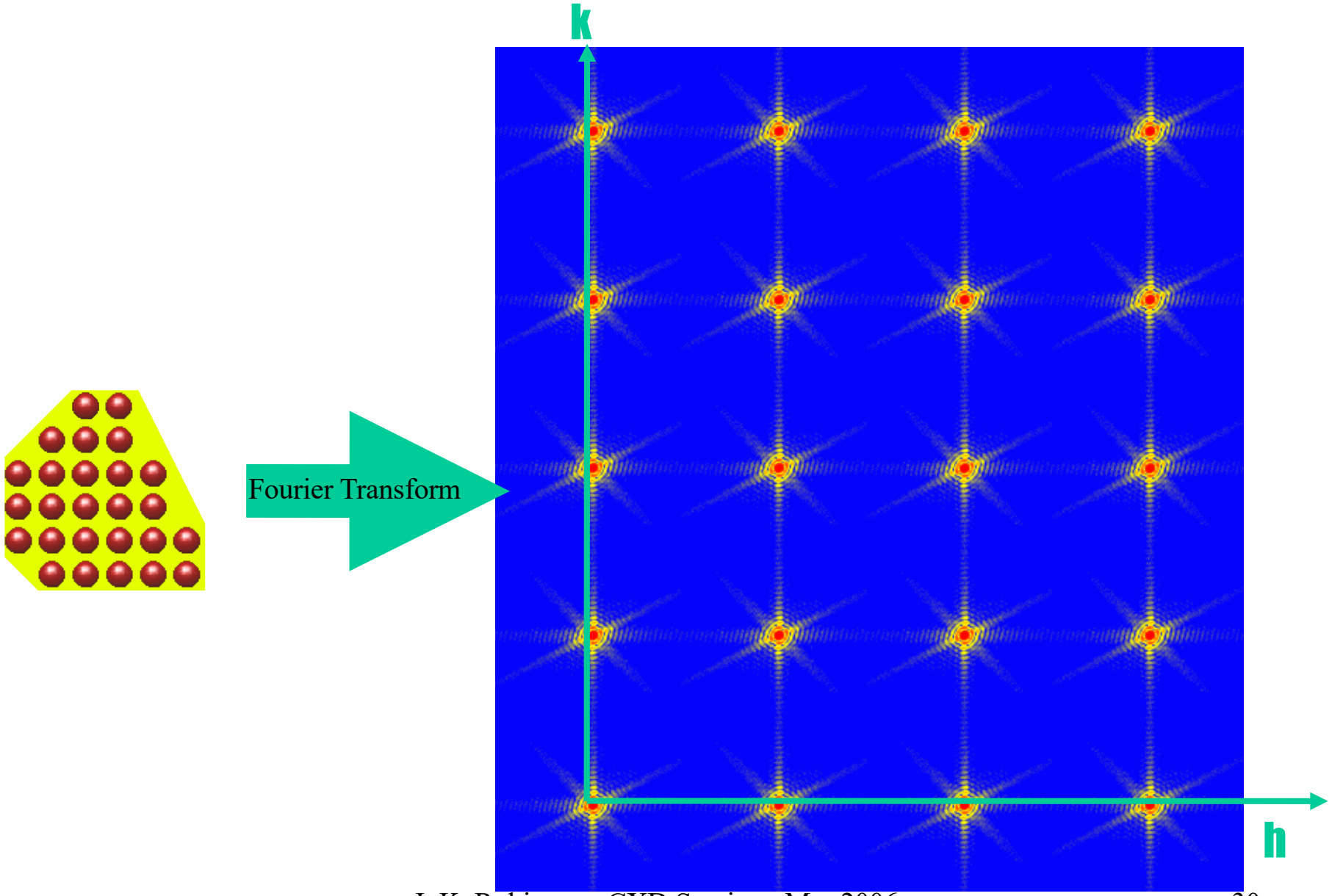

computar
MODEL HSC-1

In situ growth of Pb crystals





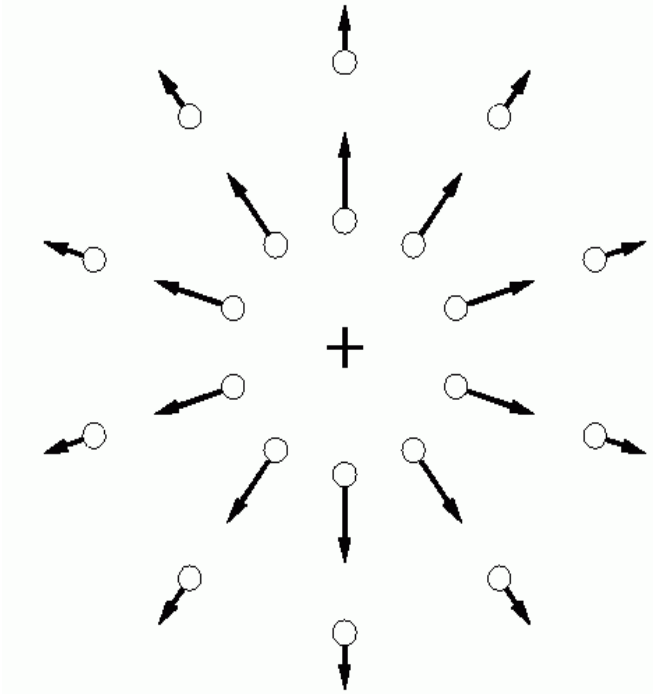
Coherent Diffraction from Crystals

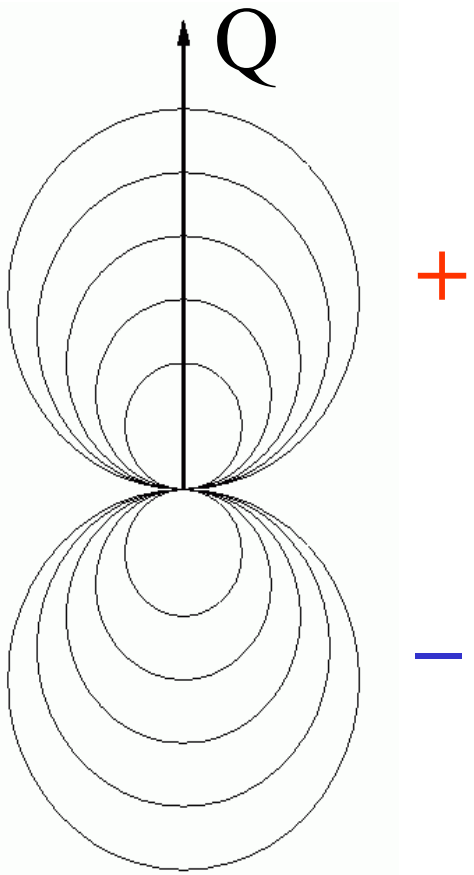


Diffraction by Strain of Point Defect

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

Imaginary density





Good statistics, 3D diffraction data

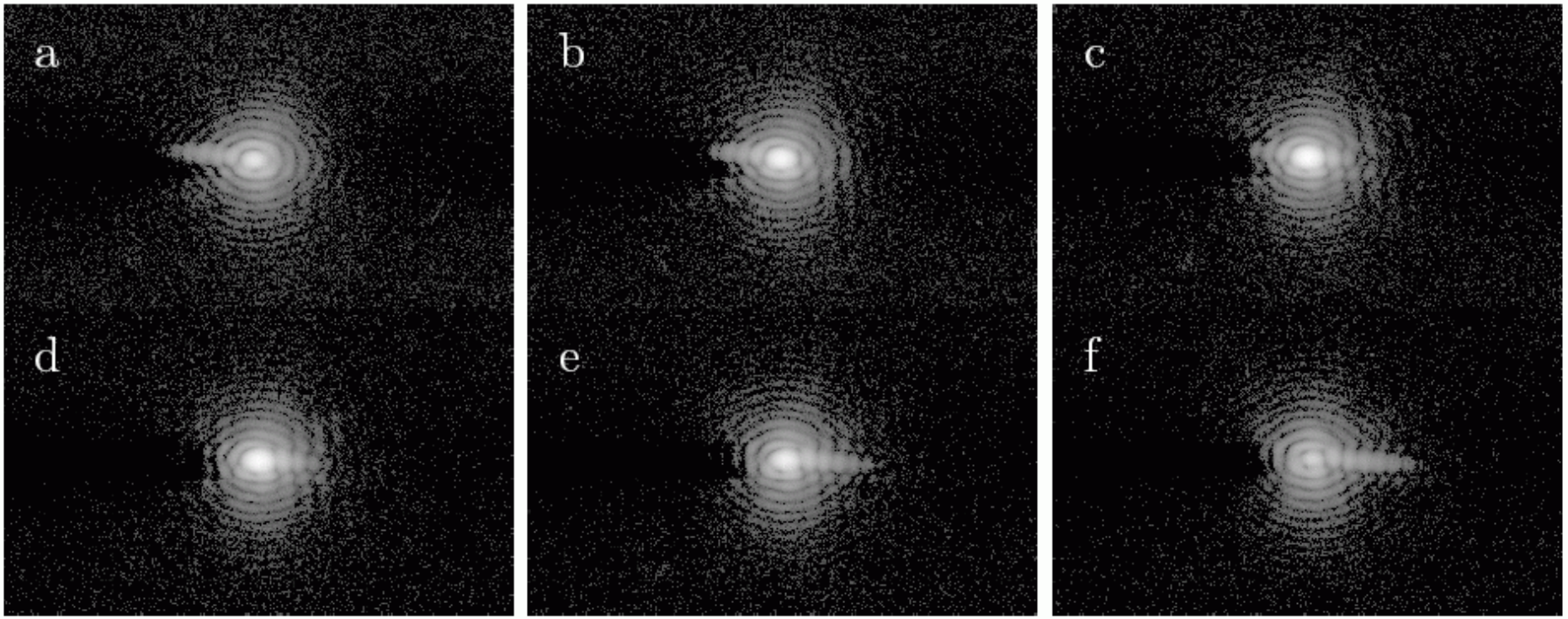
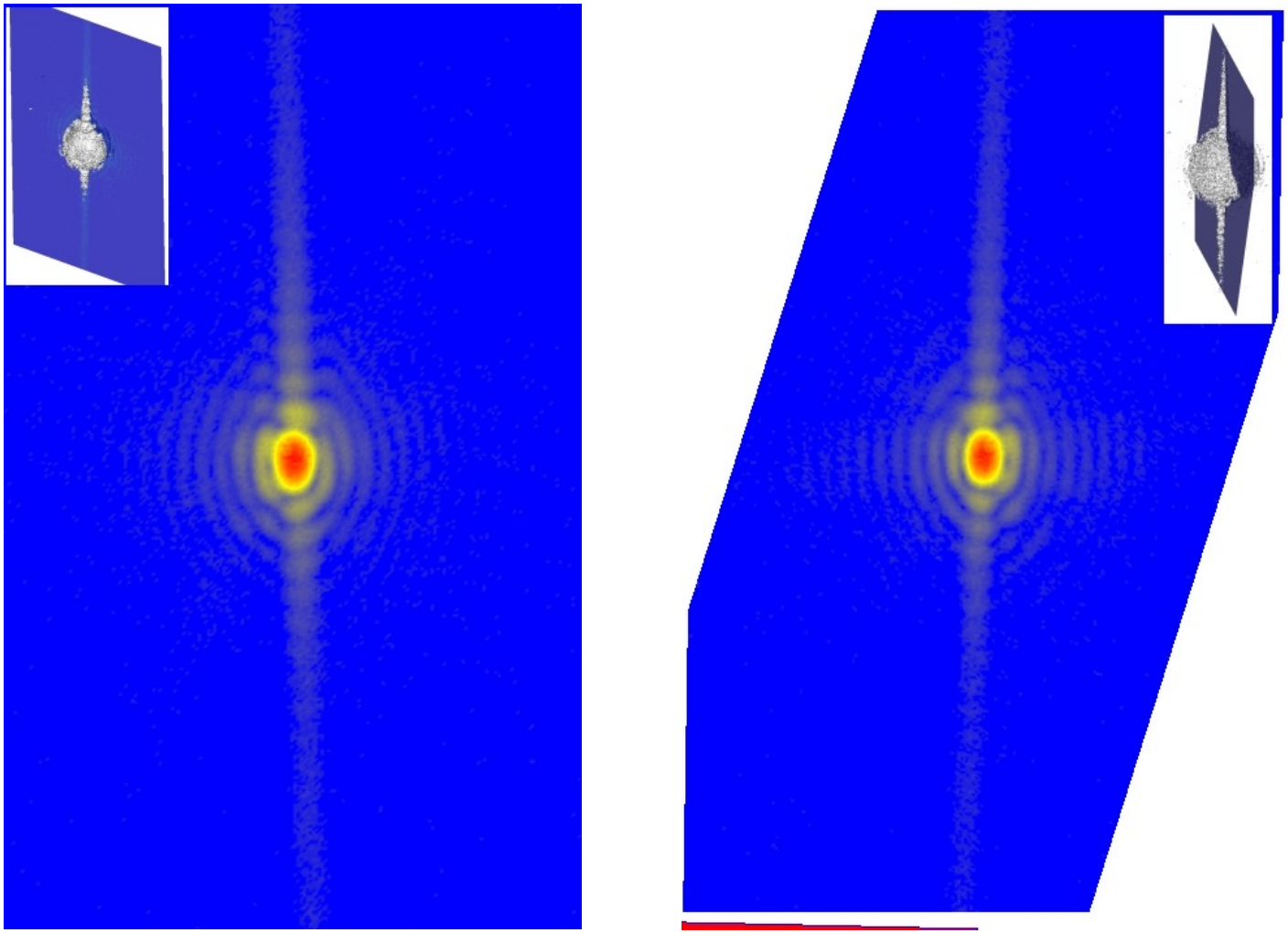
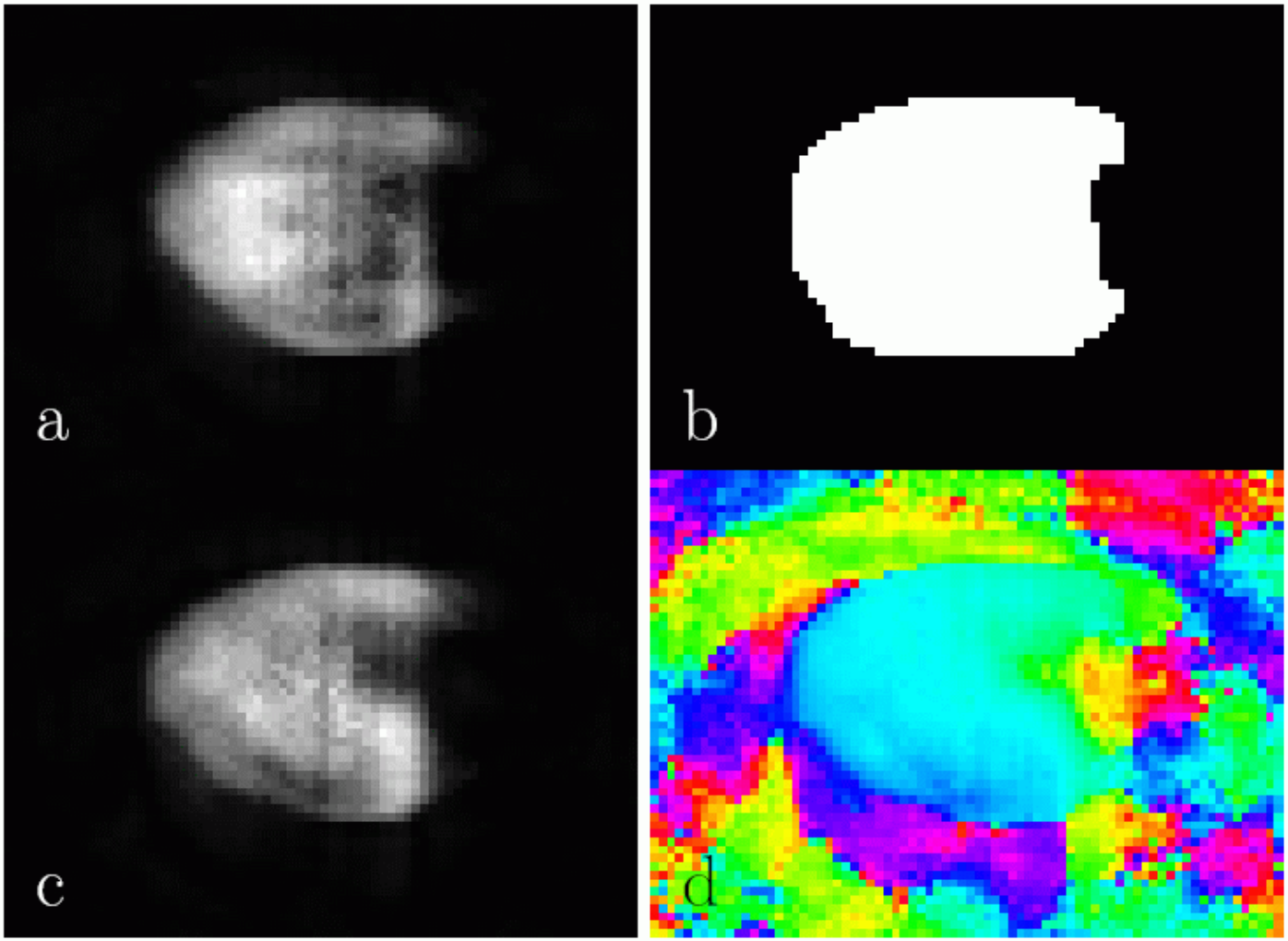


Figure 4.12: Center slices from 3D CXD pattern from Pb sample, on a log scale. Data file 296 from 10/03.

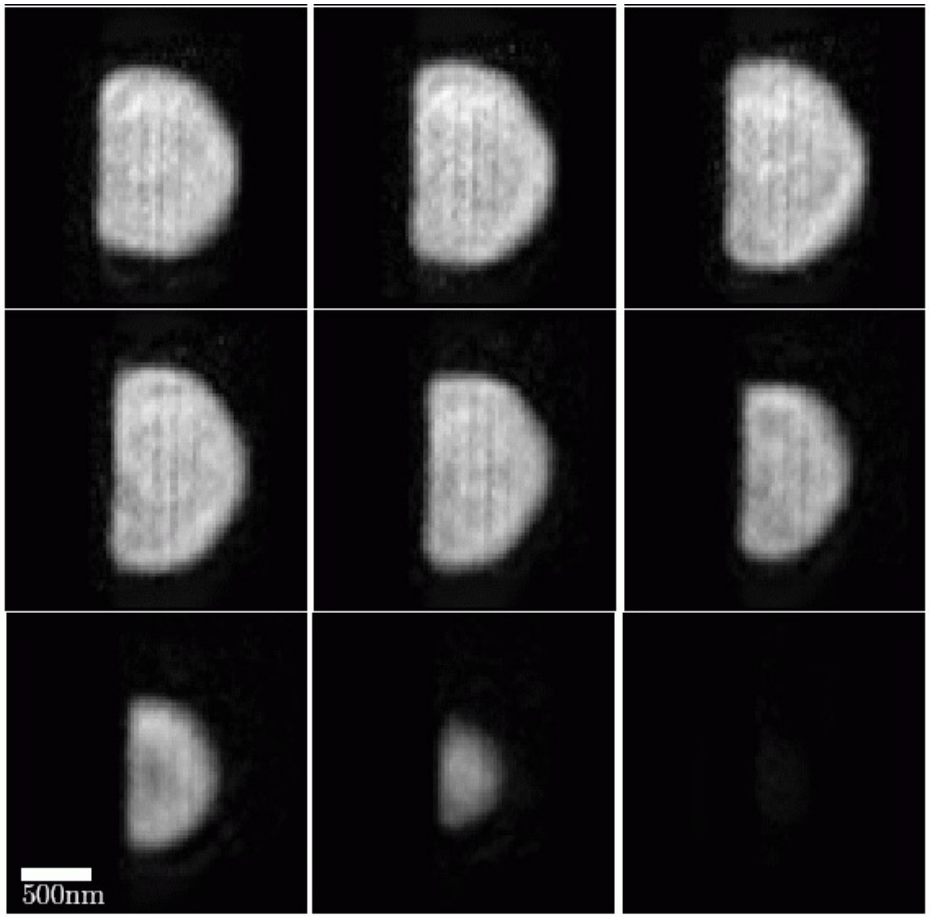
3D data along special directions

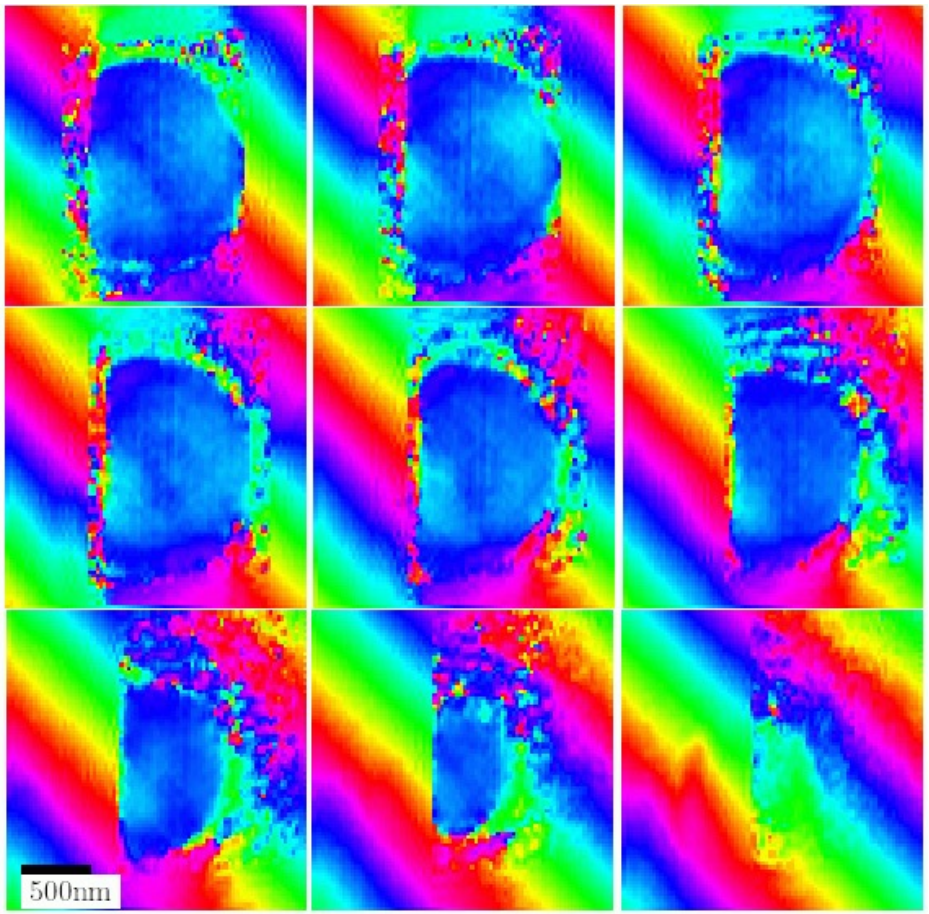


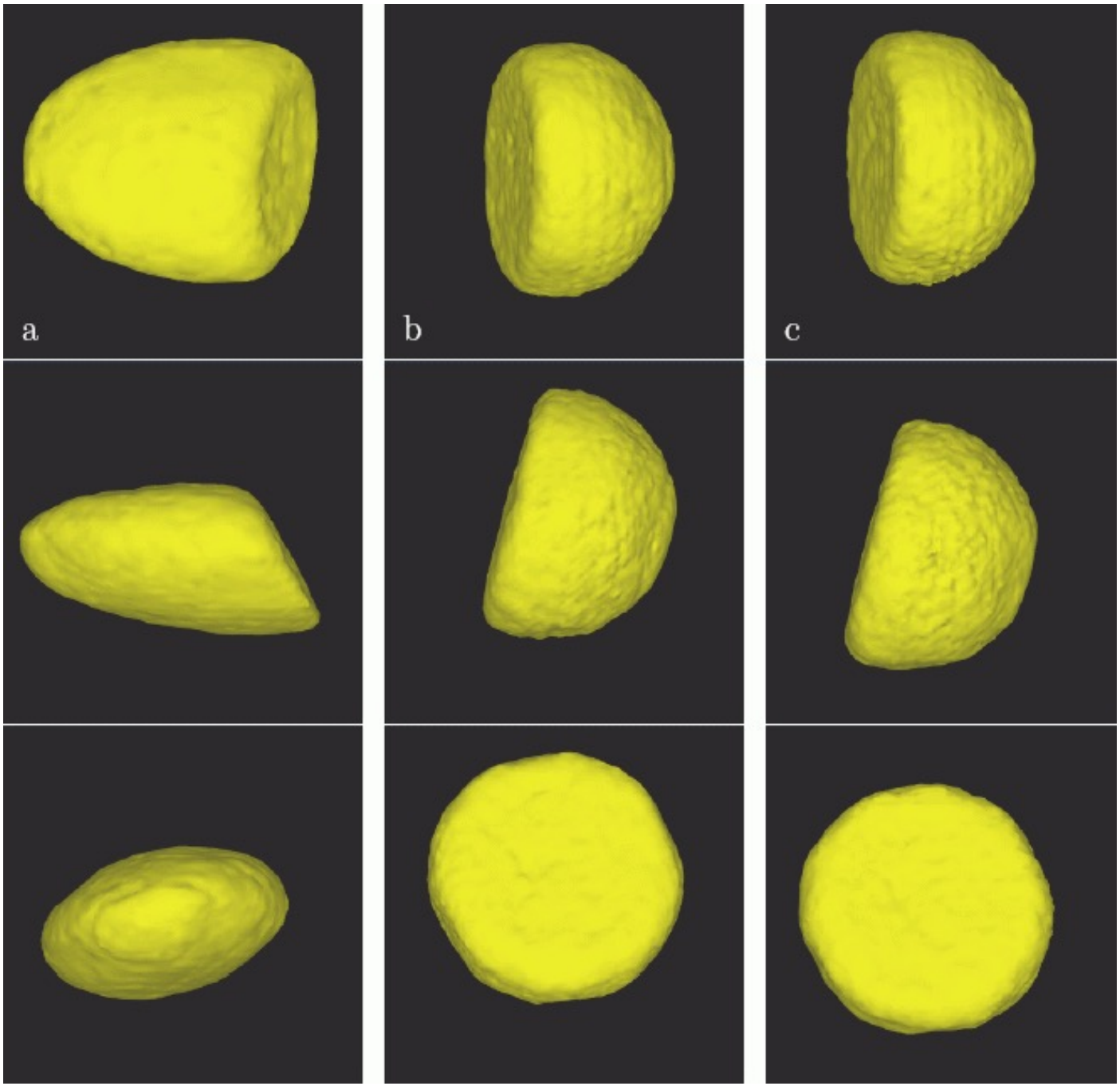
Learn shape of “tight” support



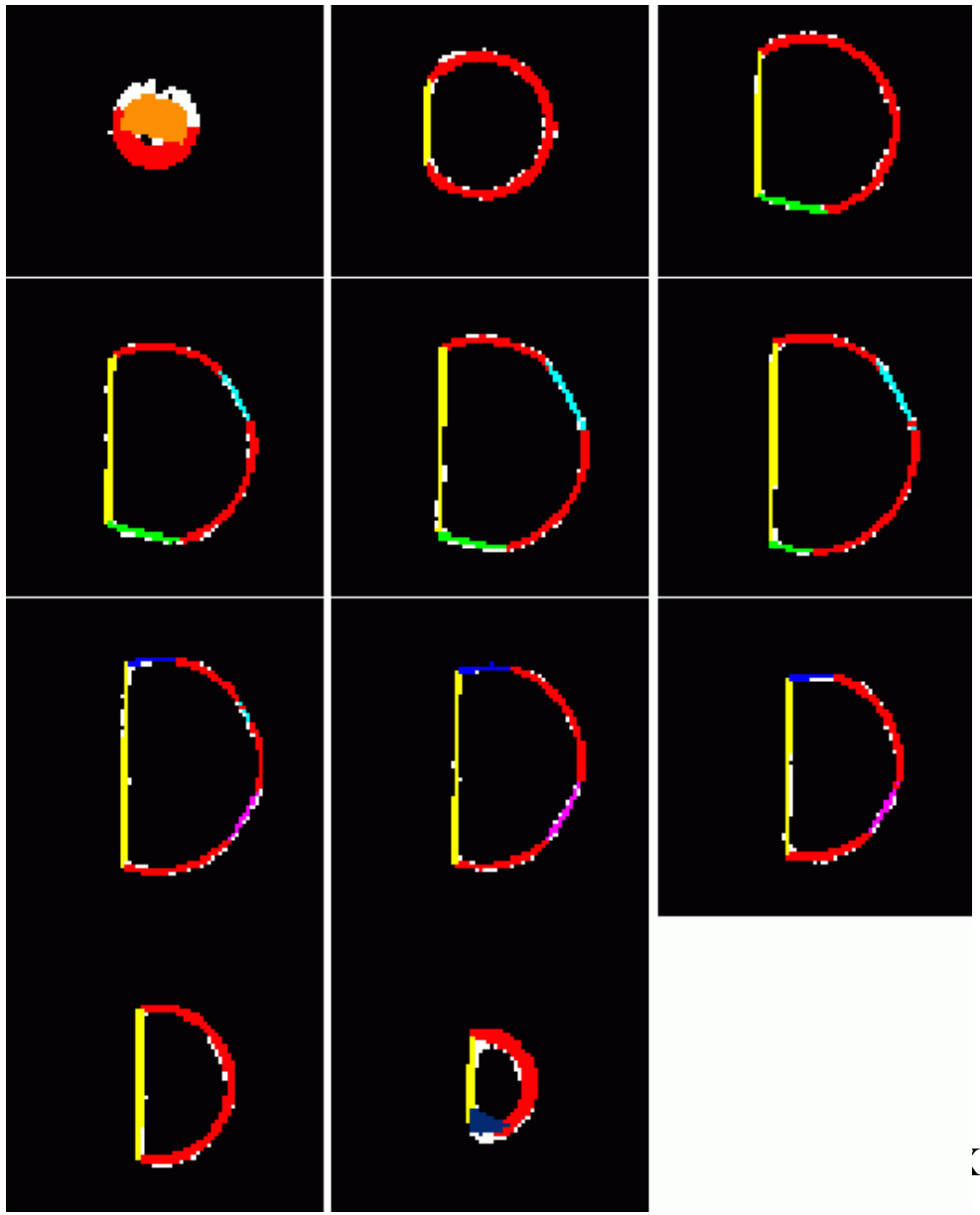
Then refine amplitude *and* phase





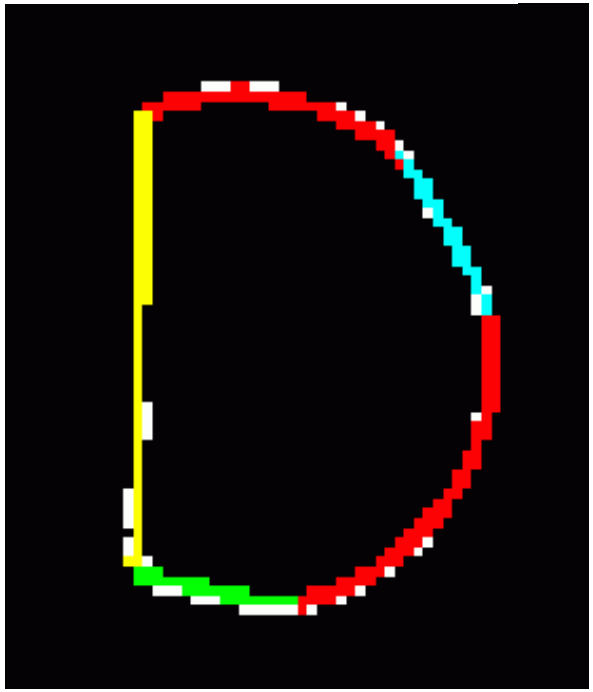

a
b
c

Fitting to facetted shape

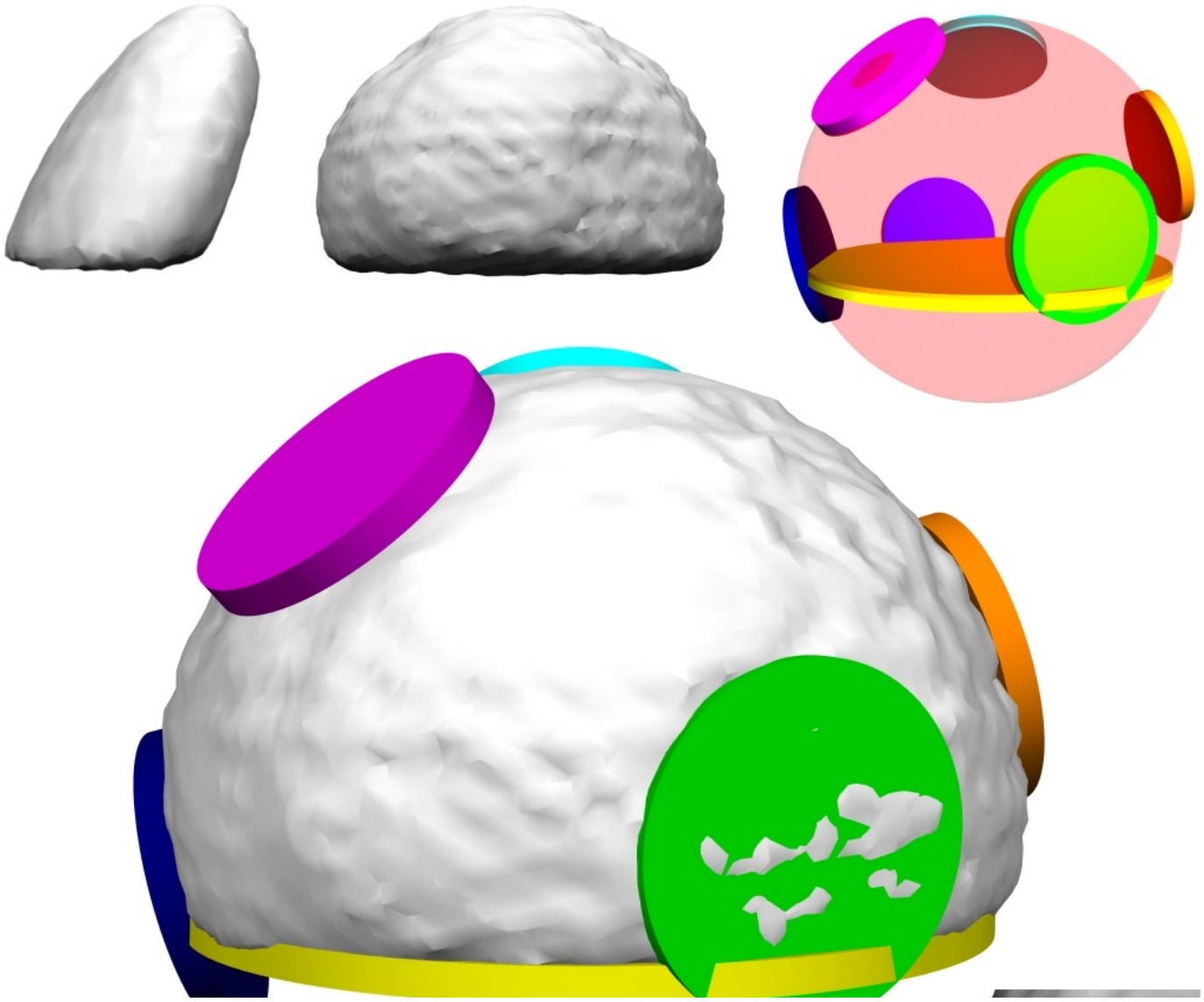


	$\|R\|$		P0	P1	P2	P3	P4	P5	P6
	9.4	P0	0	85	149	79	134	106	71
	25.7	P1		0	123	164	83	76	102
	25.1	P2			0	72	67	74	110
	25.9	P3				0	111	106	76
	25.4	P4					0	113	68
	25.4	P5						0	176
	26.0	P6							0 0

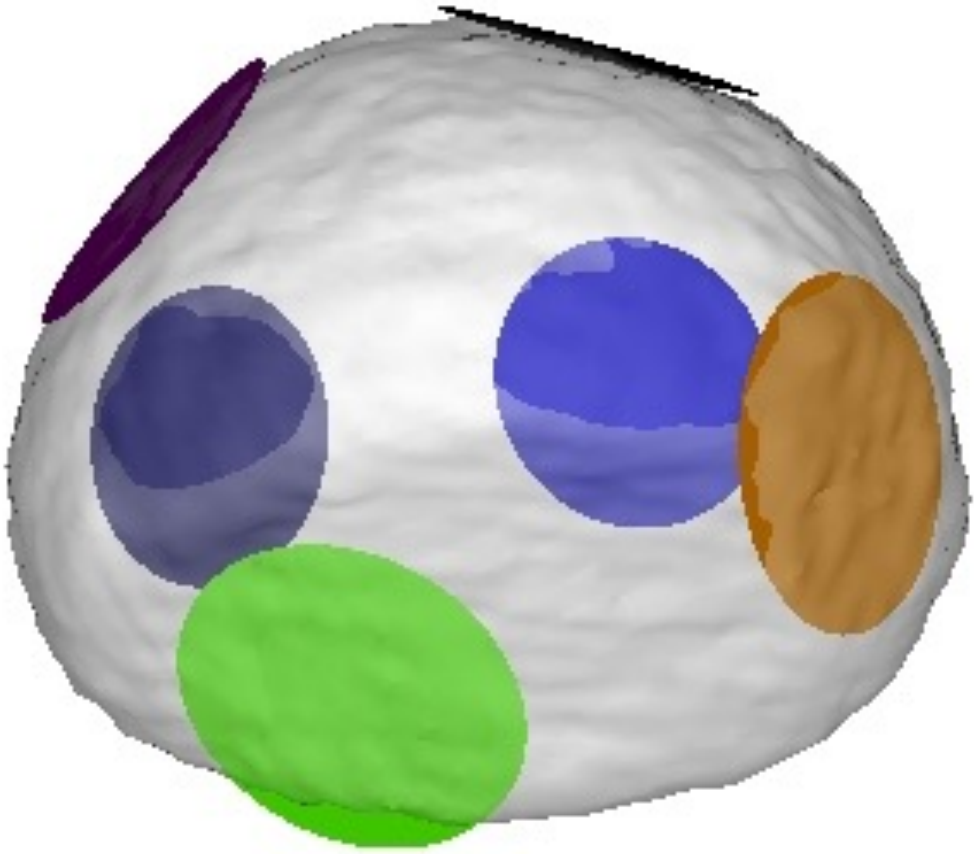
Angles between facets

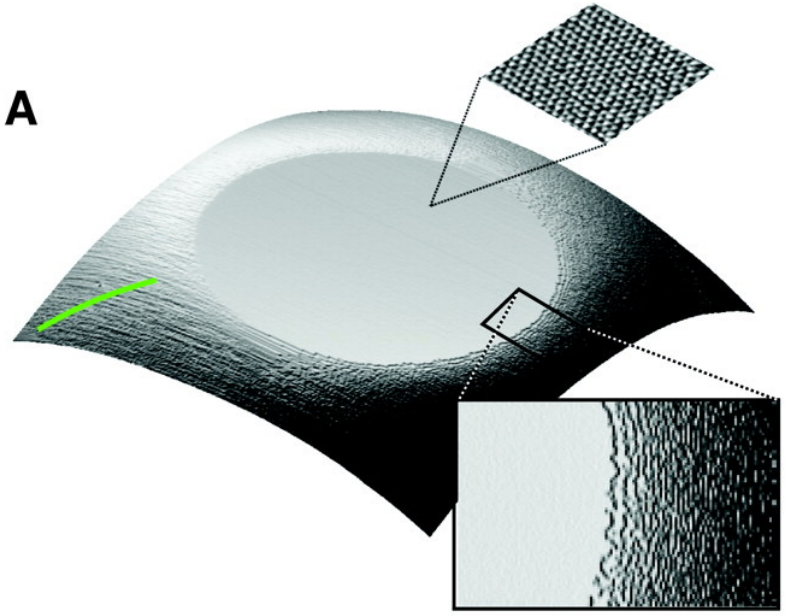


Fitting of crystal shape



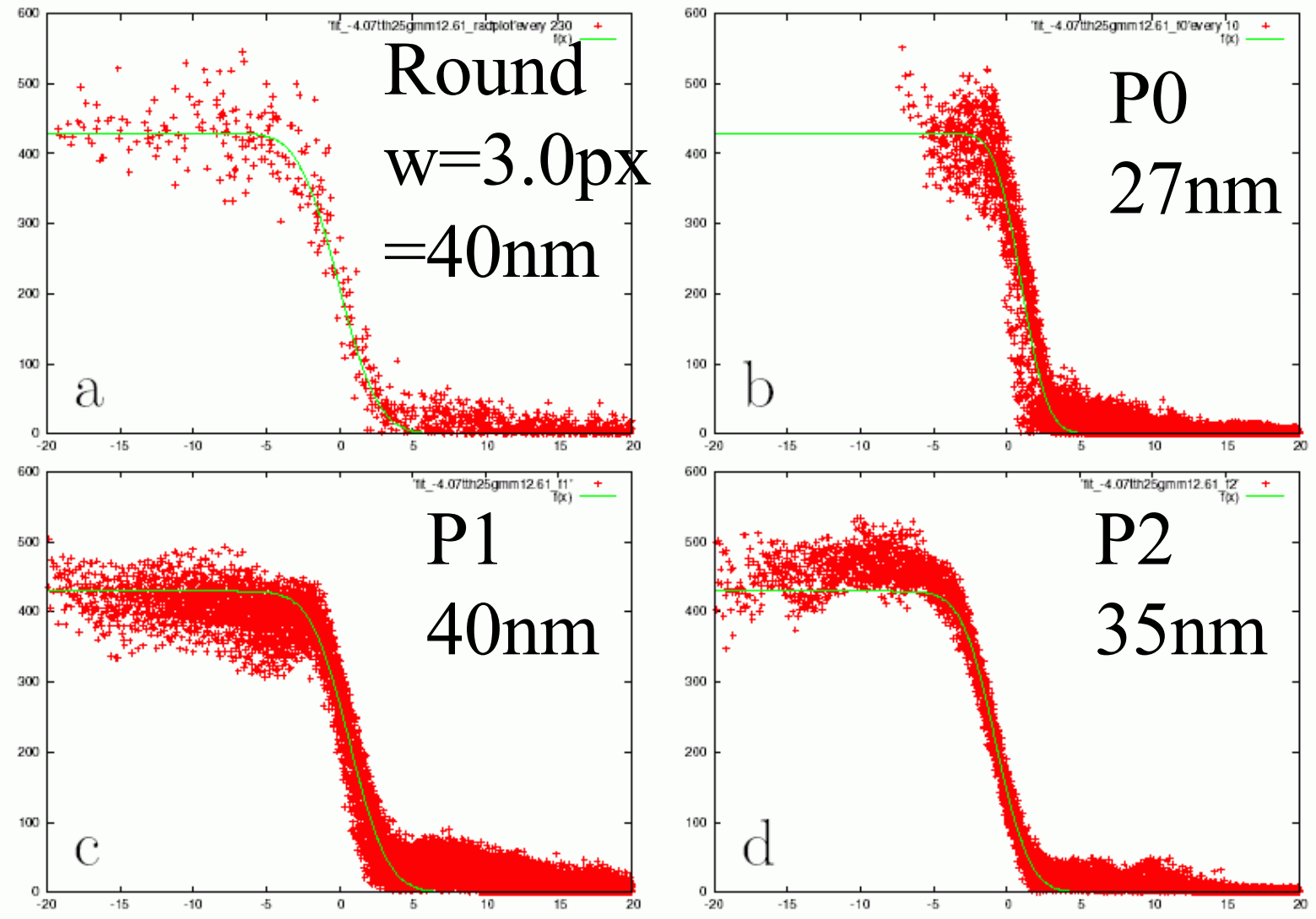
Facets of Equilibrium Crystal Shape



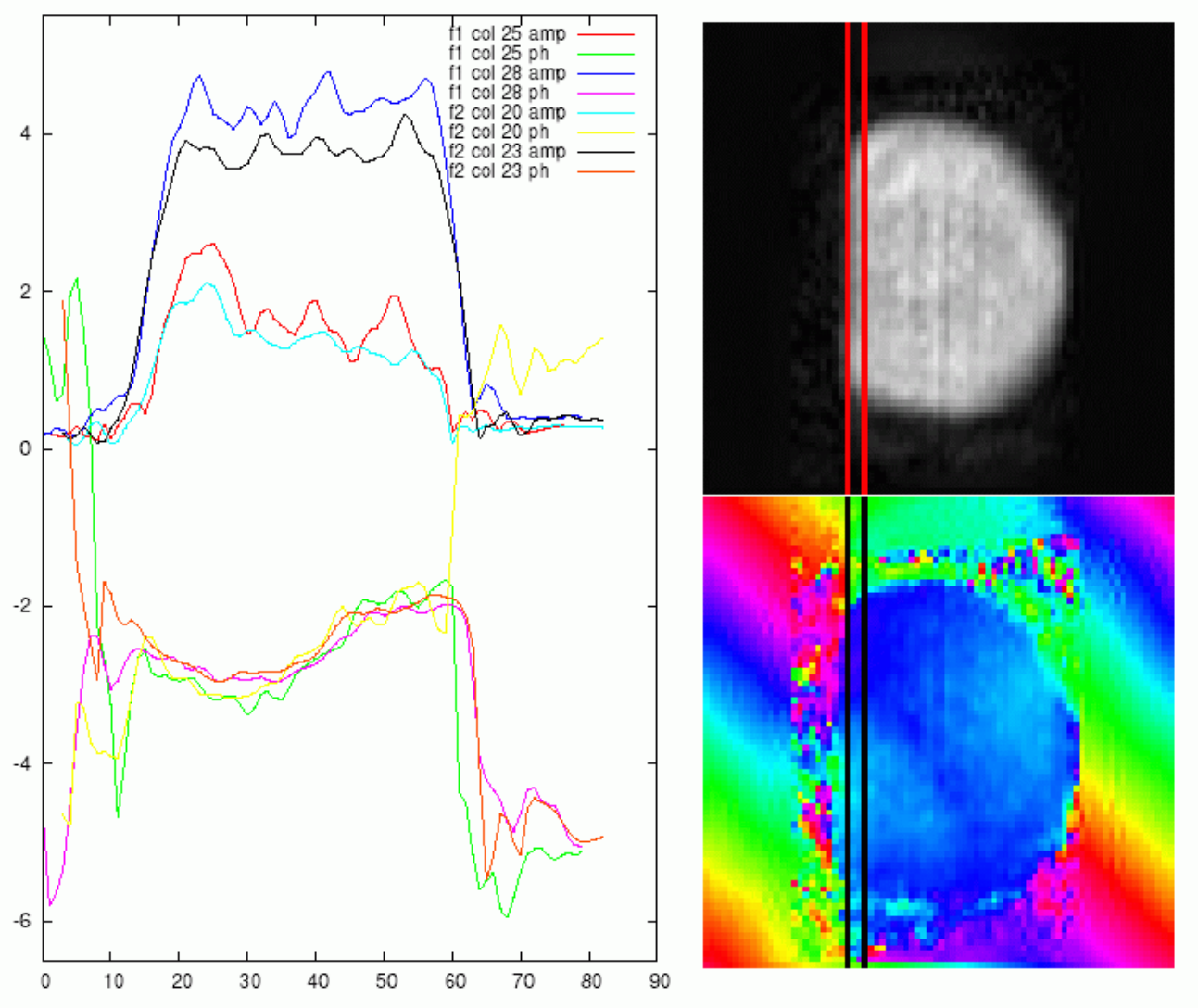


Thurmer K, Williams E, Reutt-Robey J
Science **297** 2033 (2002)

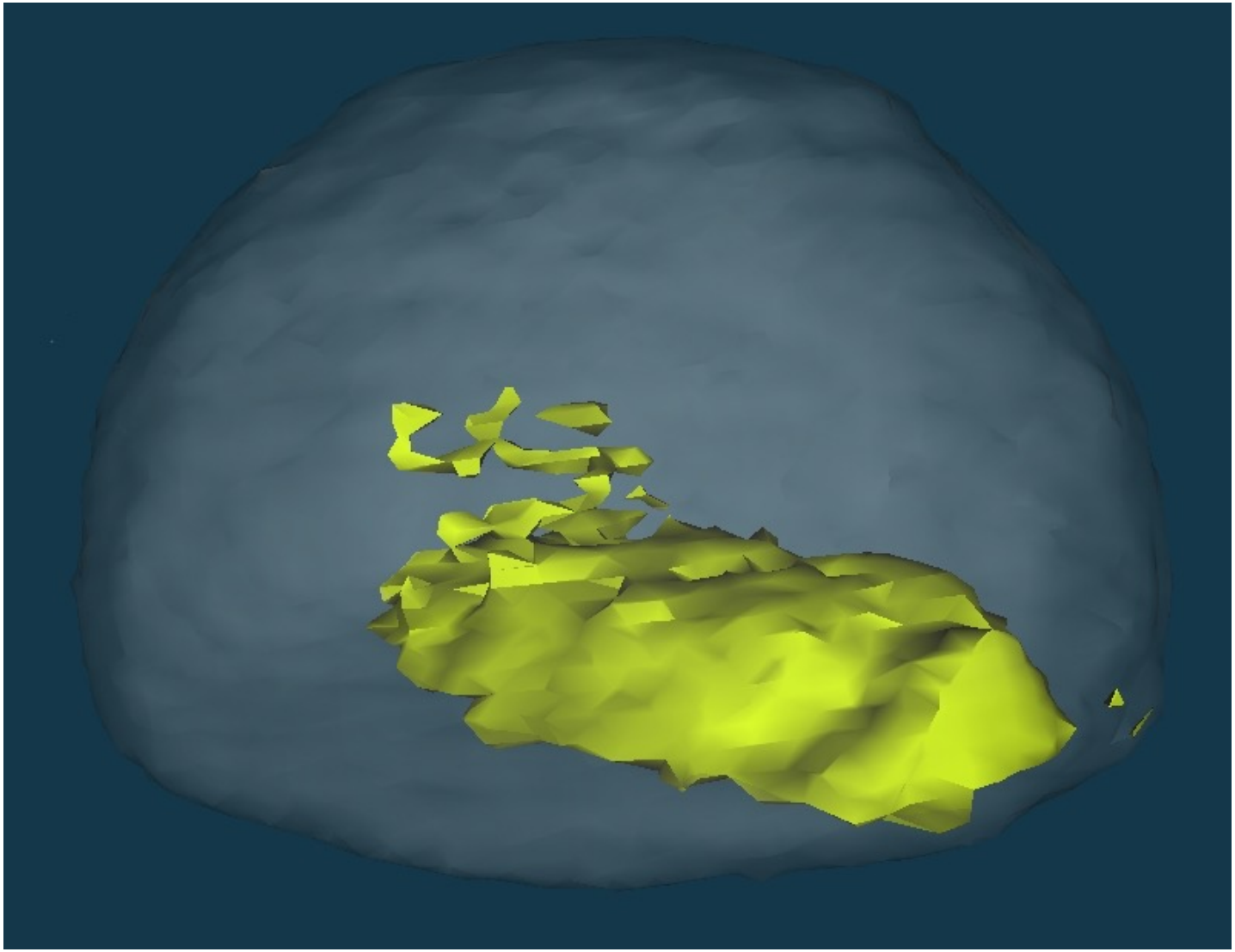
Density distribution across surface



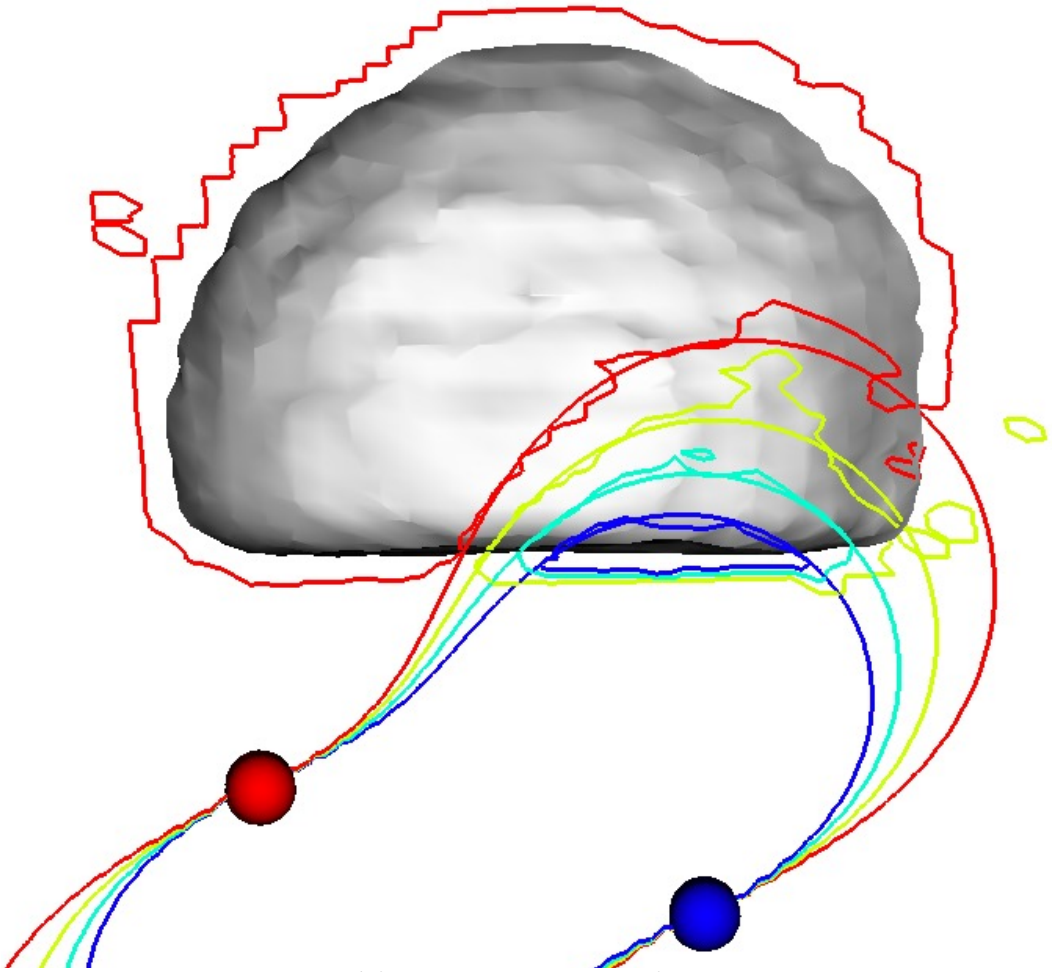
Phase structure near substrate interface



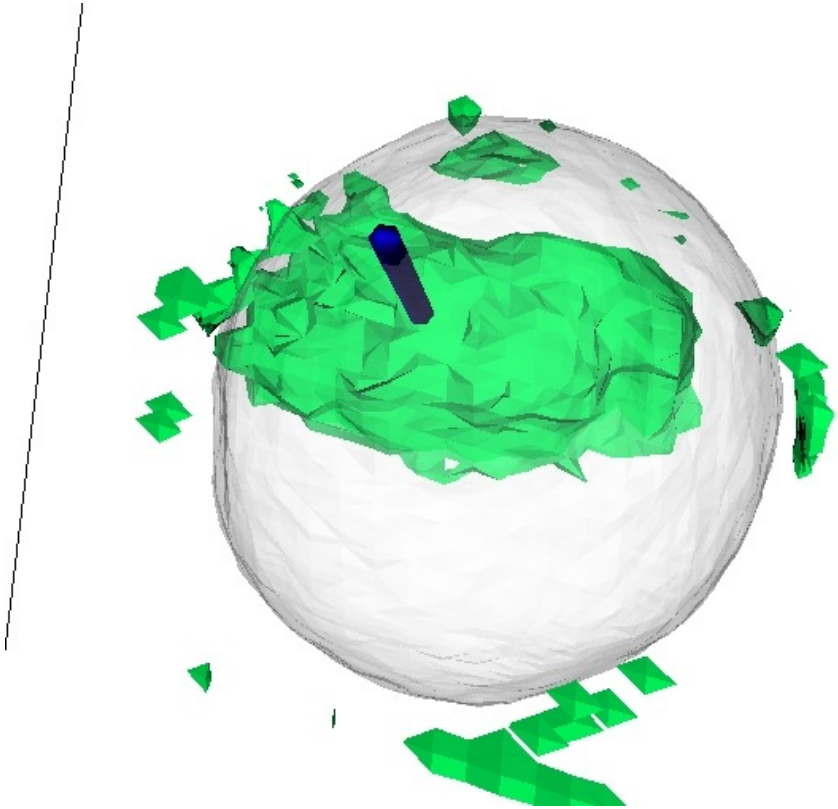
Modeling of 3D Phase Bump

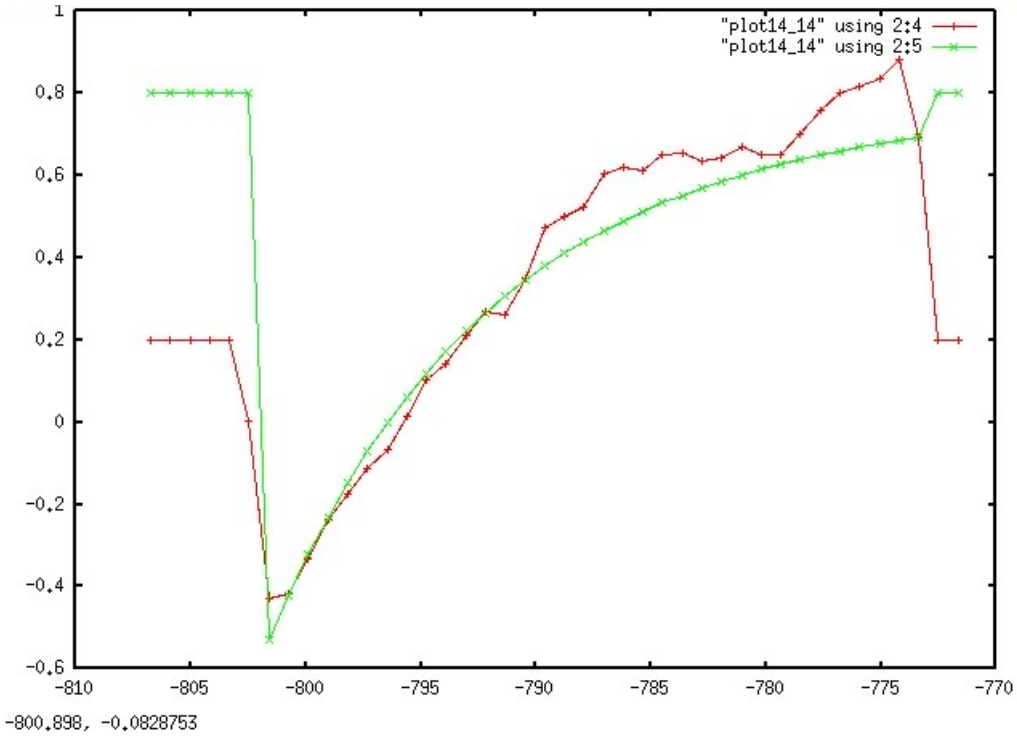


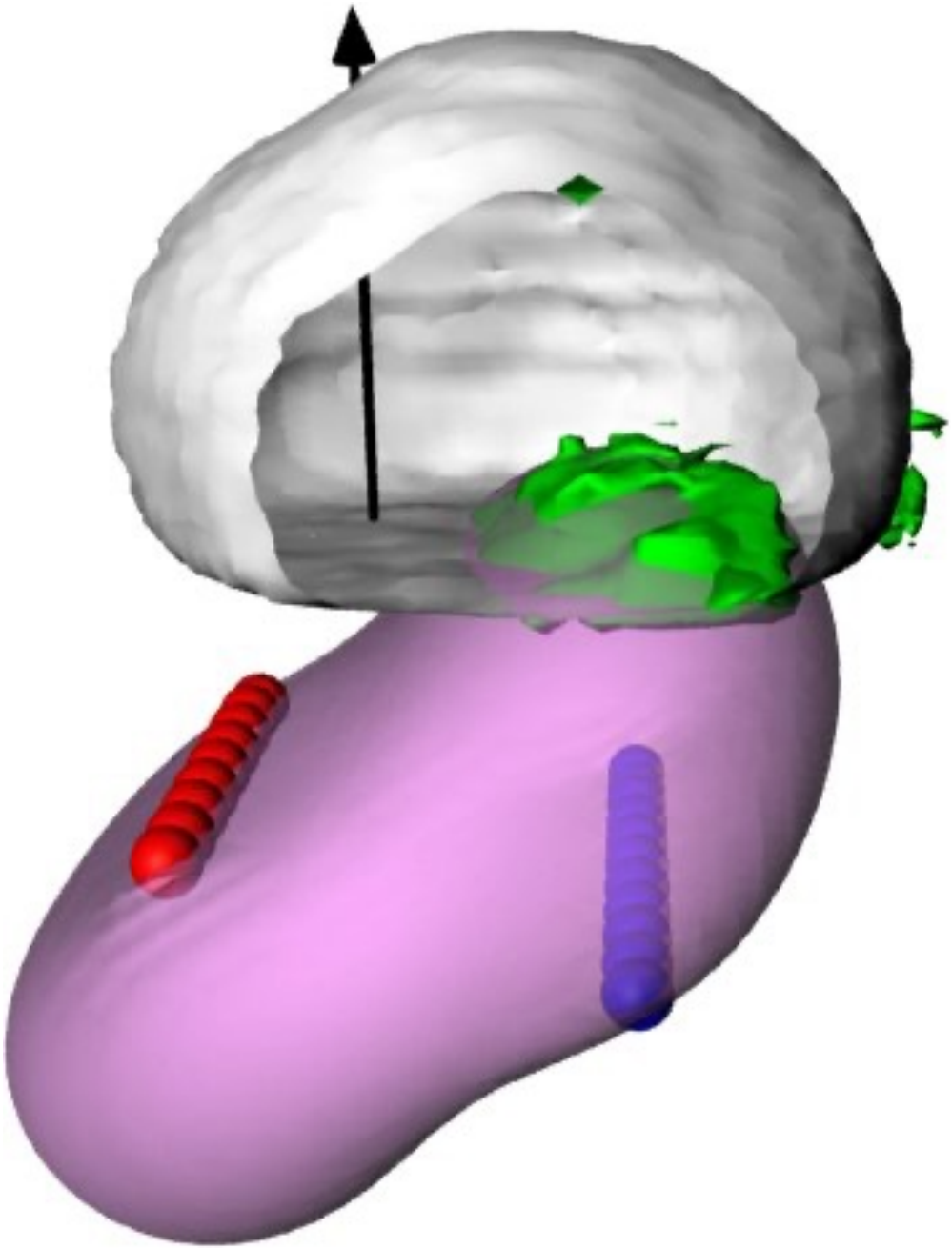
Field lines of Point Charges



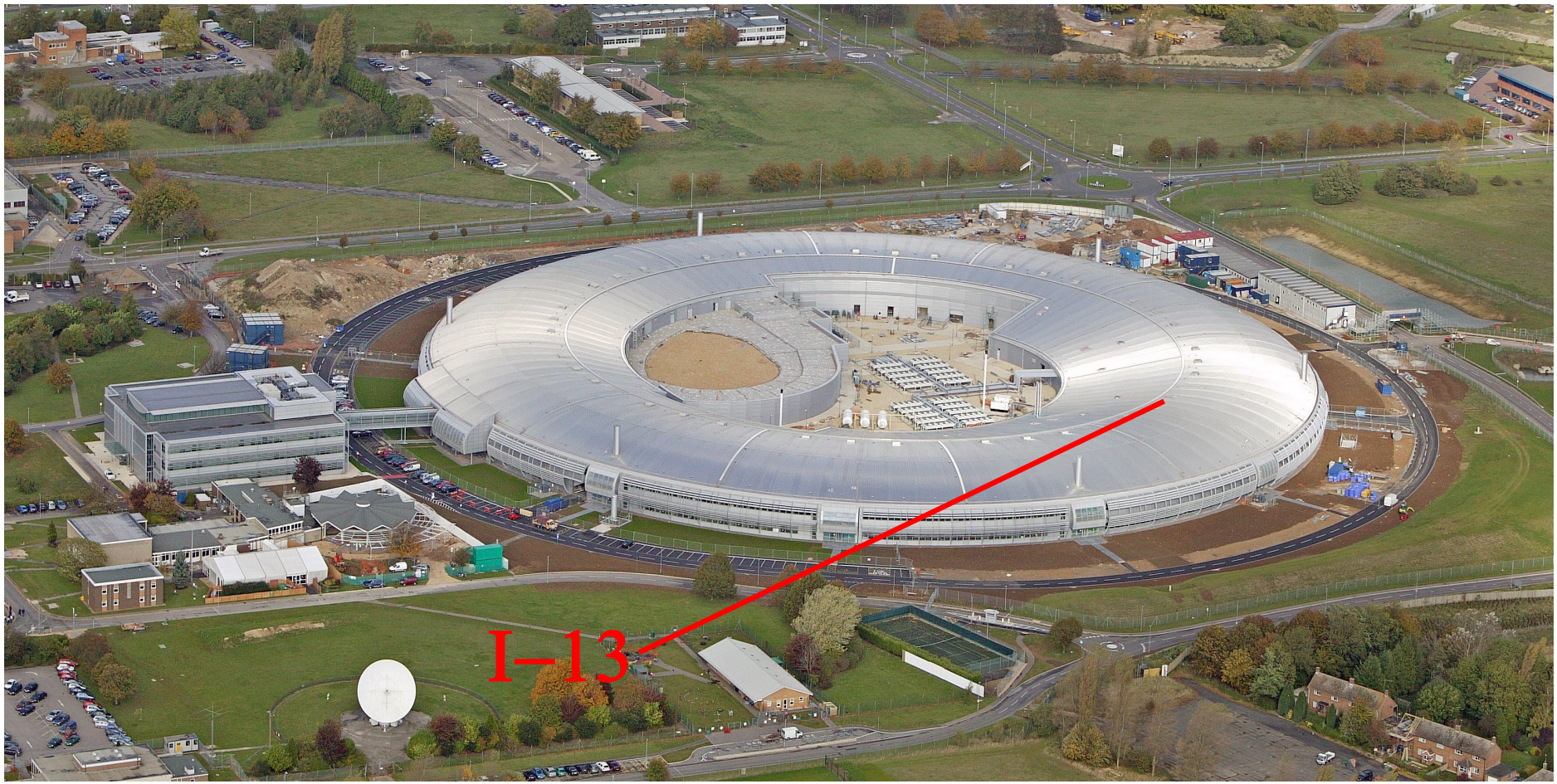
Line scan through phase





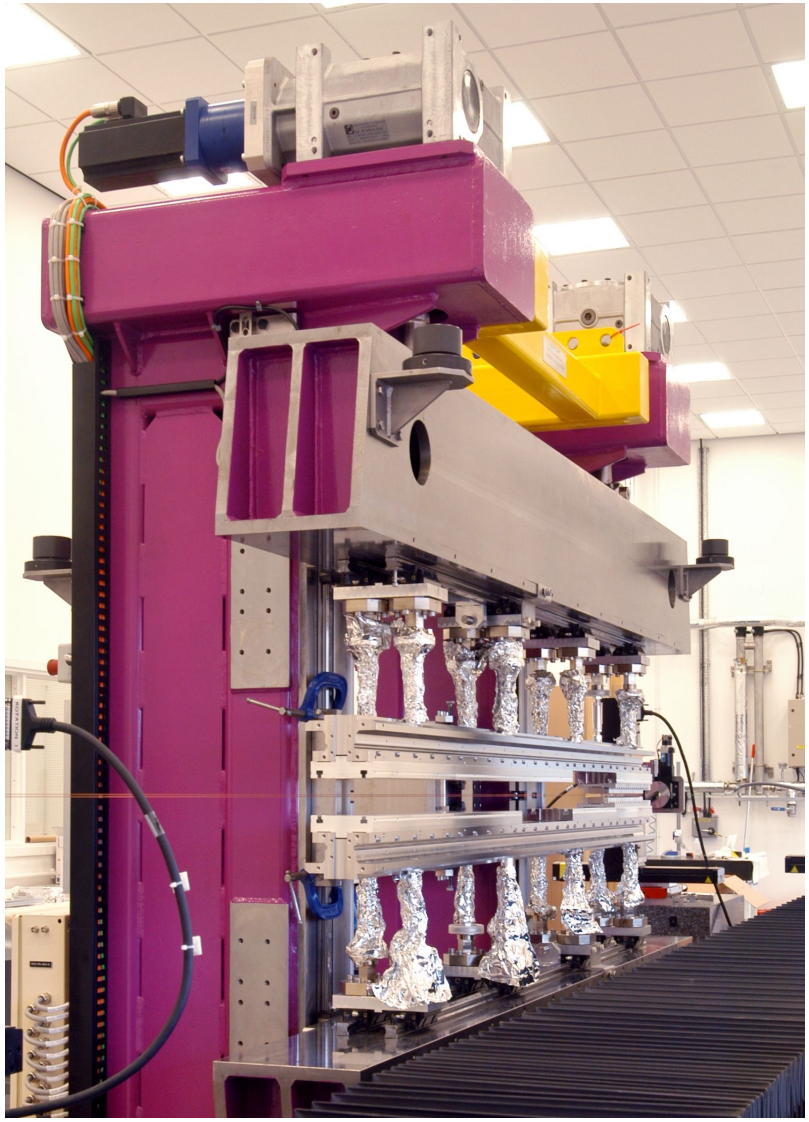


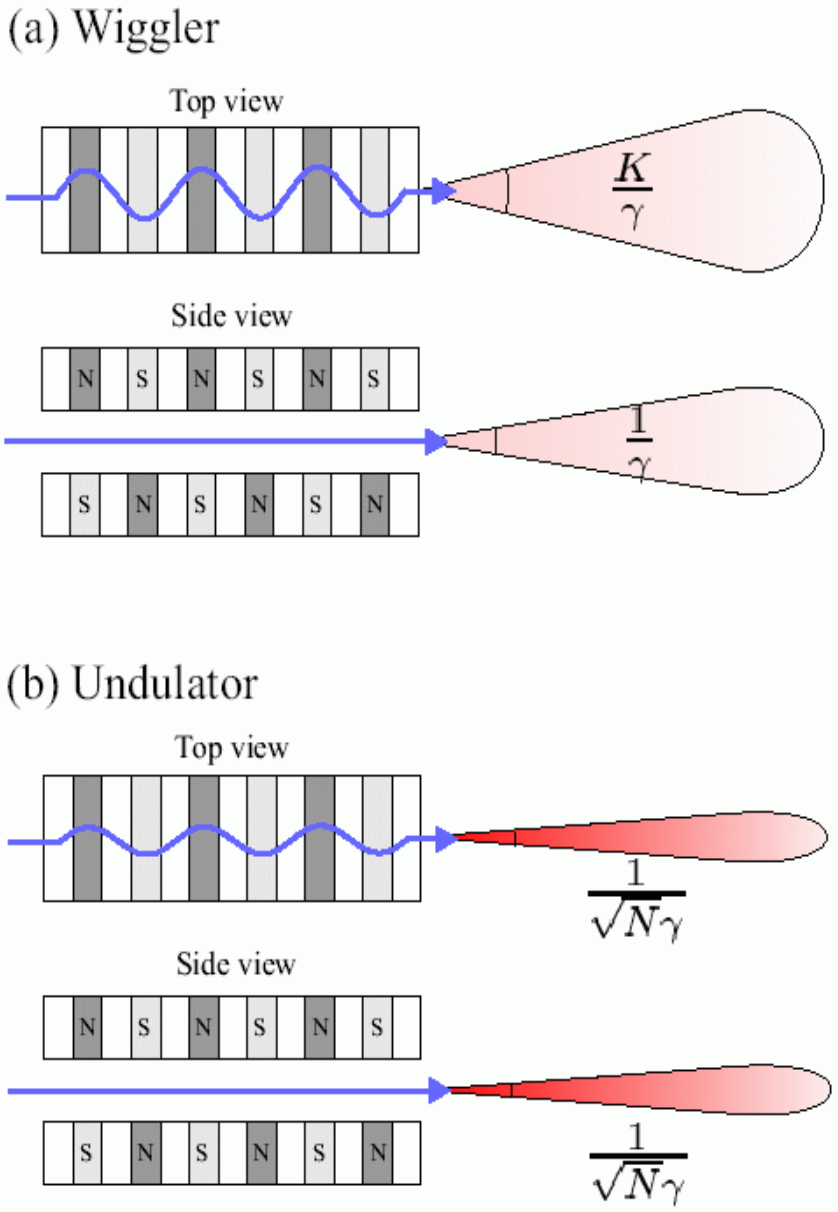
Diamond Light Source, Oct 2005



I16 Materials and Magnetism Beamline

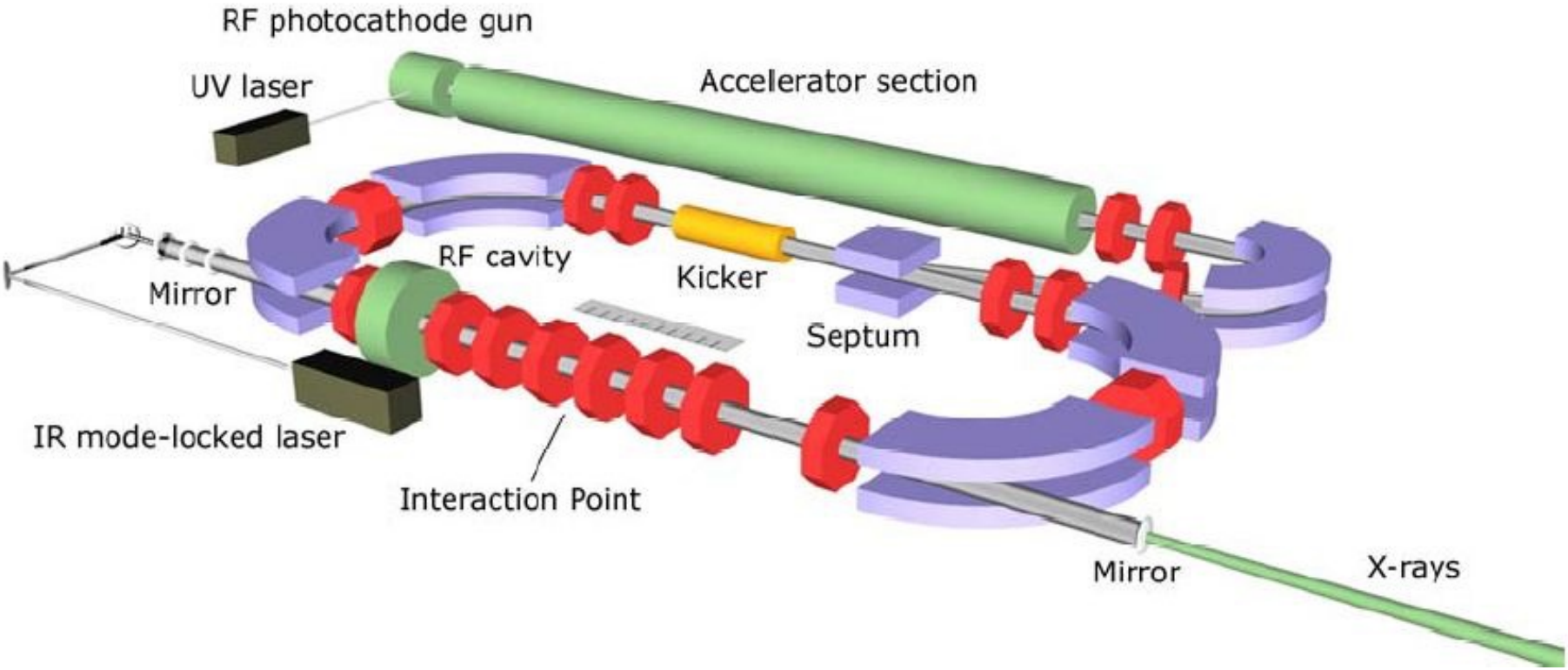
Undulator Radiation Source



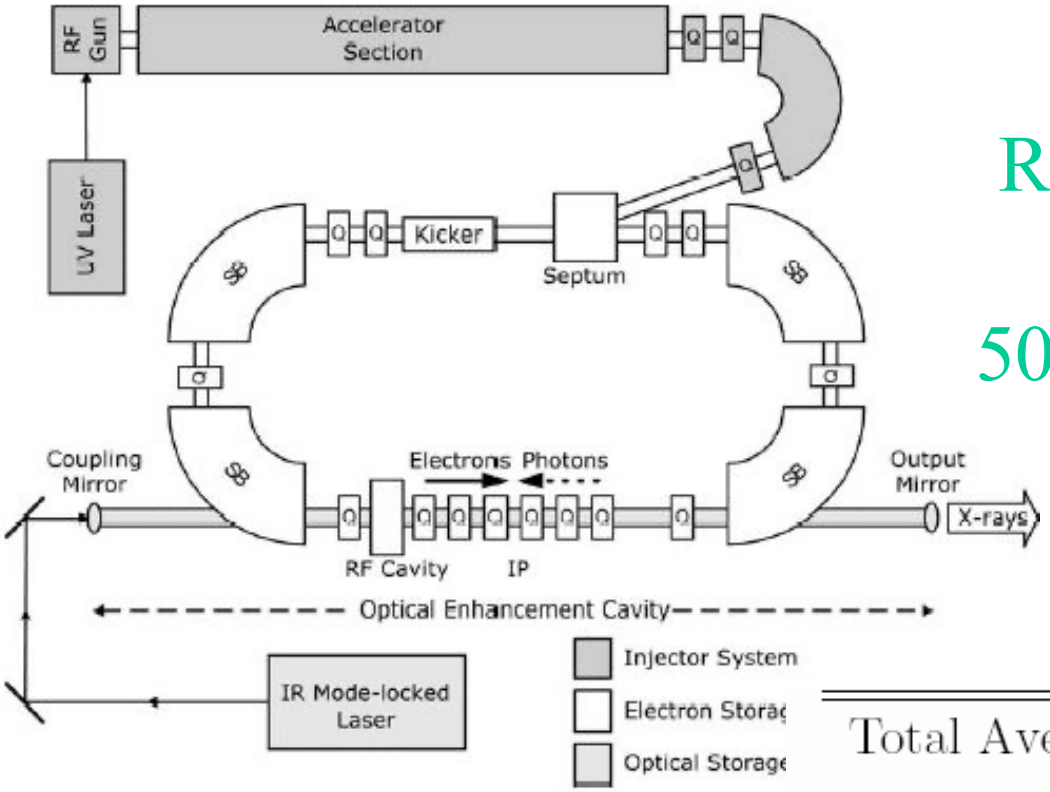


Compact Light Source

Ron Ruth, Lyncean Technologies



CLS Specifications
R. J. Loewen, Ph.D. Thesis
SLAC (2003)
50MeV (γ=100) + 1μm laser

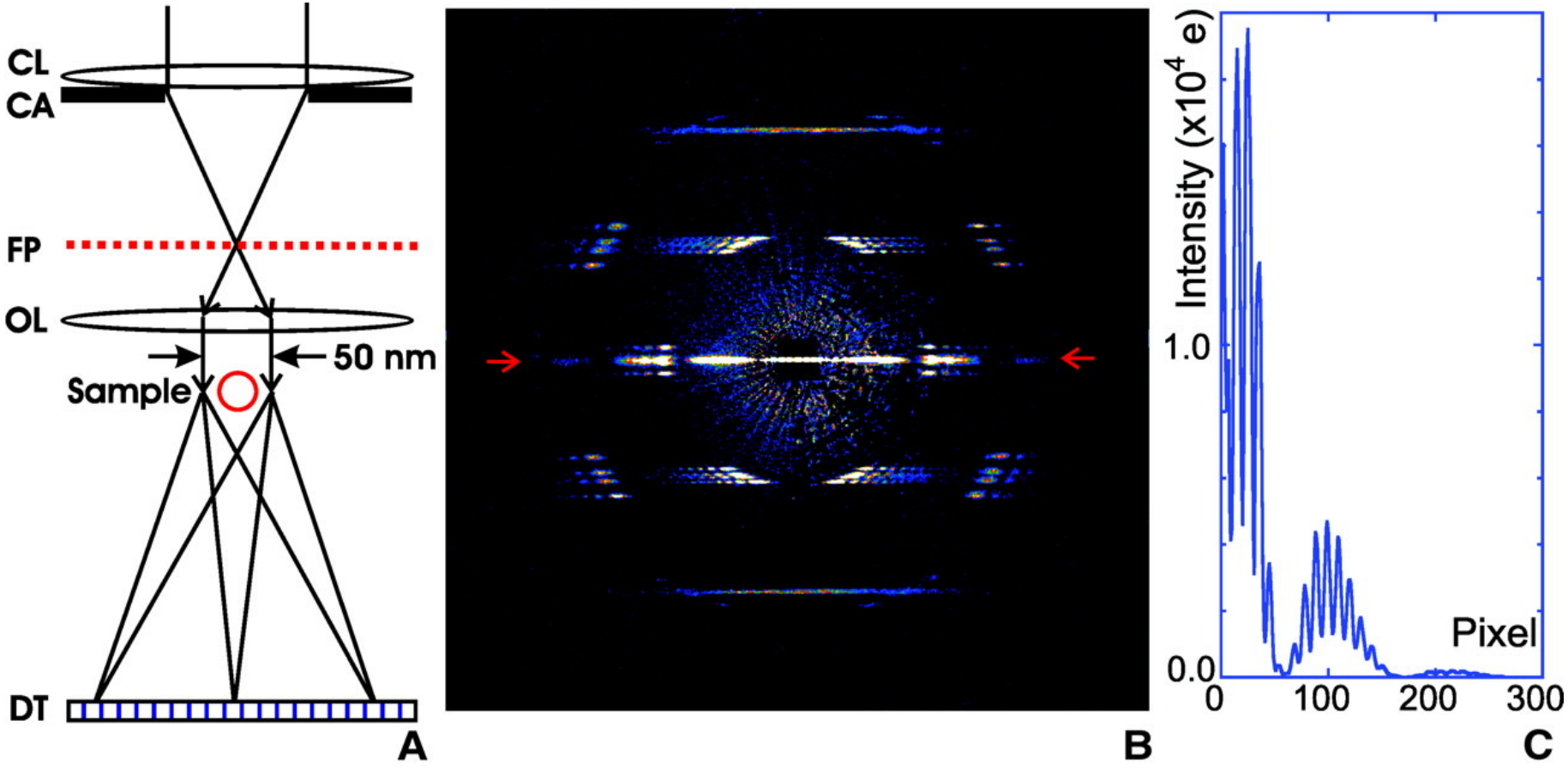


$$\omega = \frac{4\gamma^2}{(1+\gamma^2\theta^2)}\,\omega_0$$

Total Average Flux	$\gtrsim 10^{13}$ ph/s
Avg. "Monochromatic" Flux*	$\approx 10^{10}$ ph/s
Source Spot Size†	30 μm radius
Source Divergence†	$\approx$ 3 mrad
Source Brightness	$\gtrsim 10^{11}$
X-ray Energy Range‡	12 keV $\pm$ 6 keV

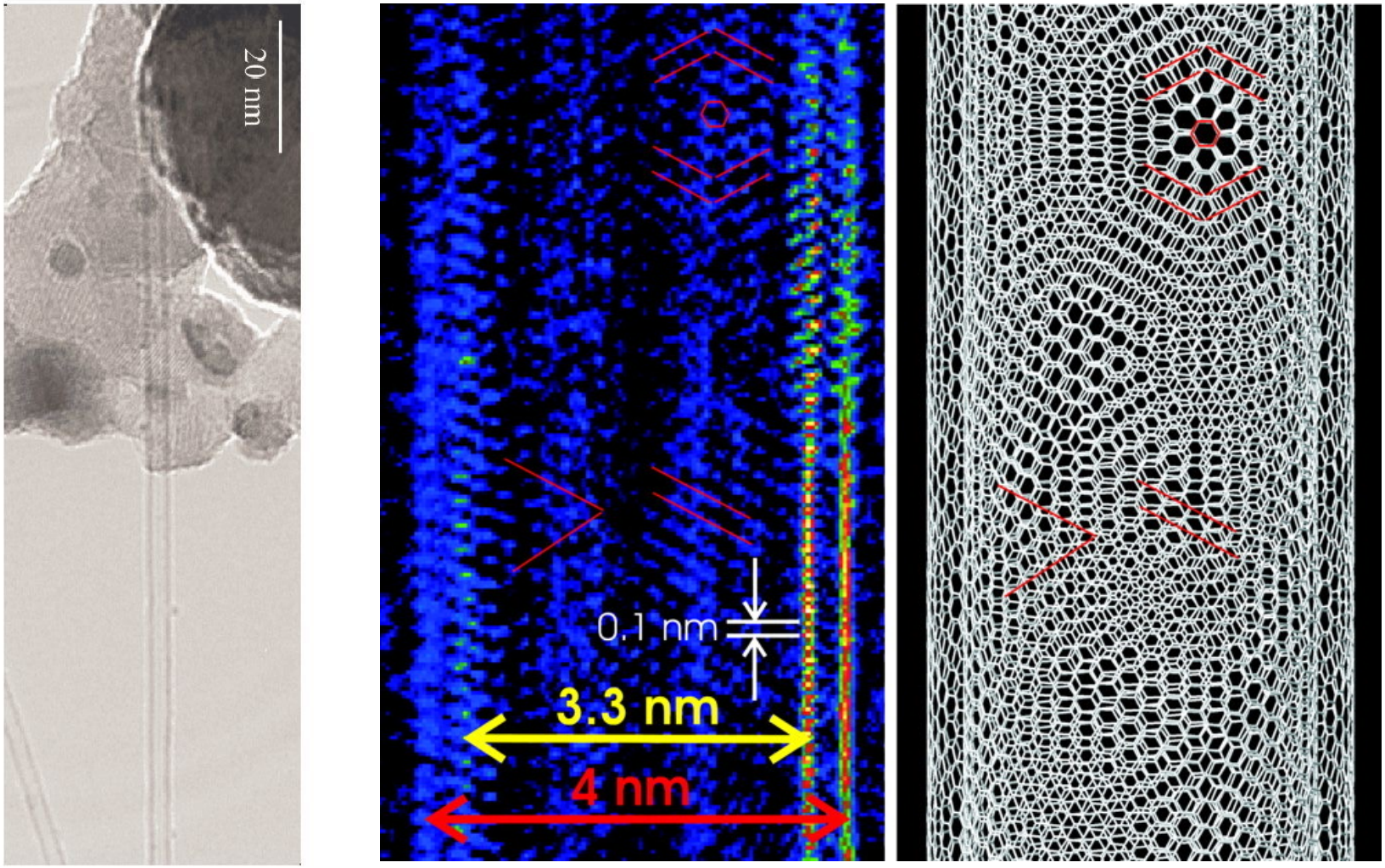
Electron Diffraction from C-nanotube

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

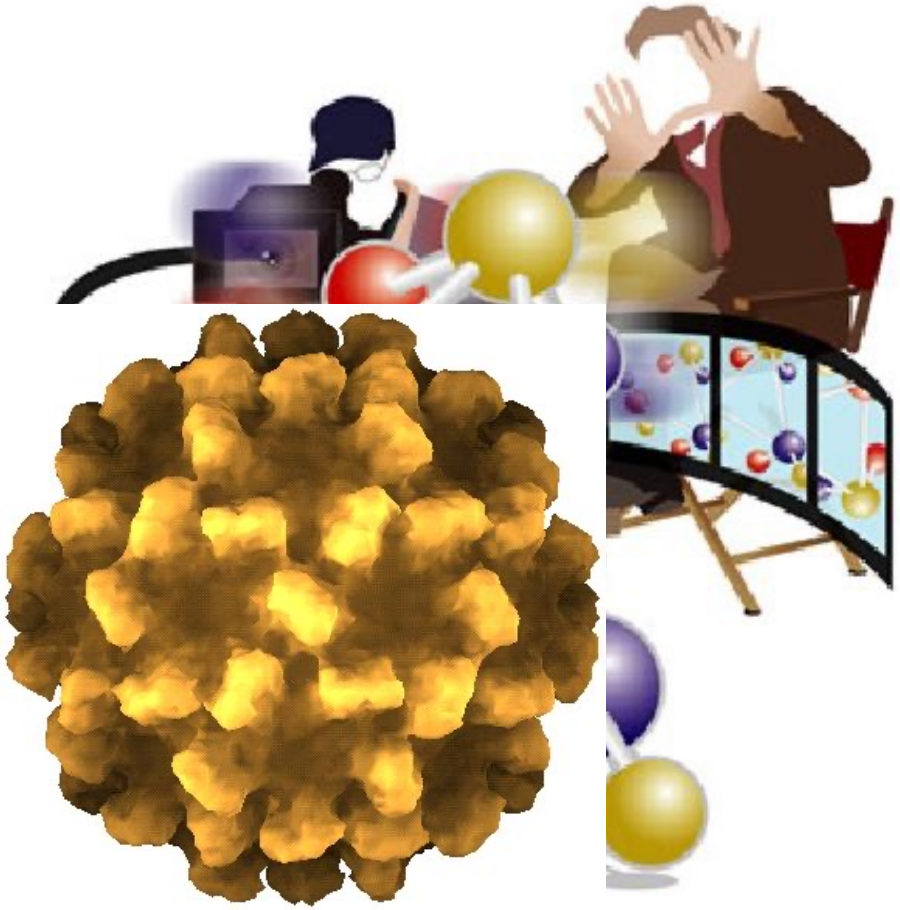


Images of DW C-nanotube

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