Coherent X-ray Diffraction as a Materials Research Tool

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Diamond Light Source
University of Illinois

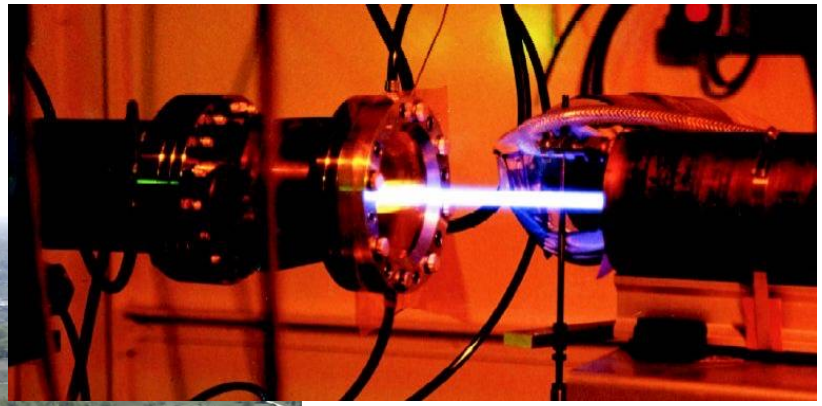
Condensed Matter Seminar,
Durham University,
December 2006

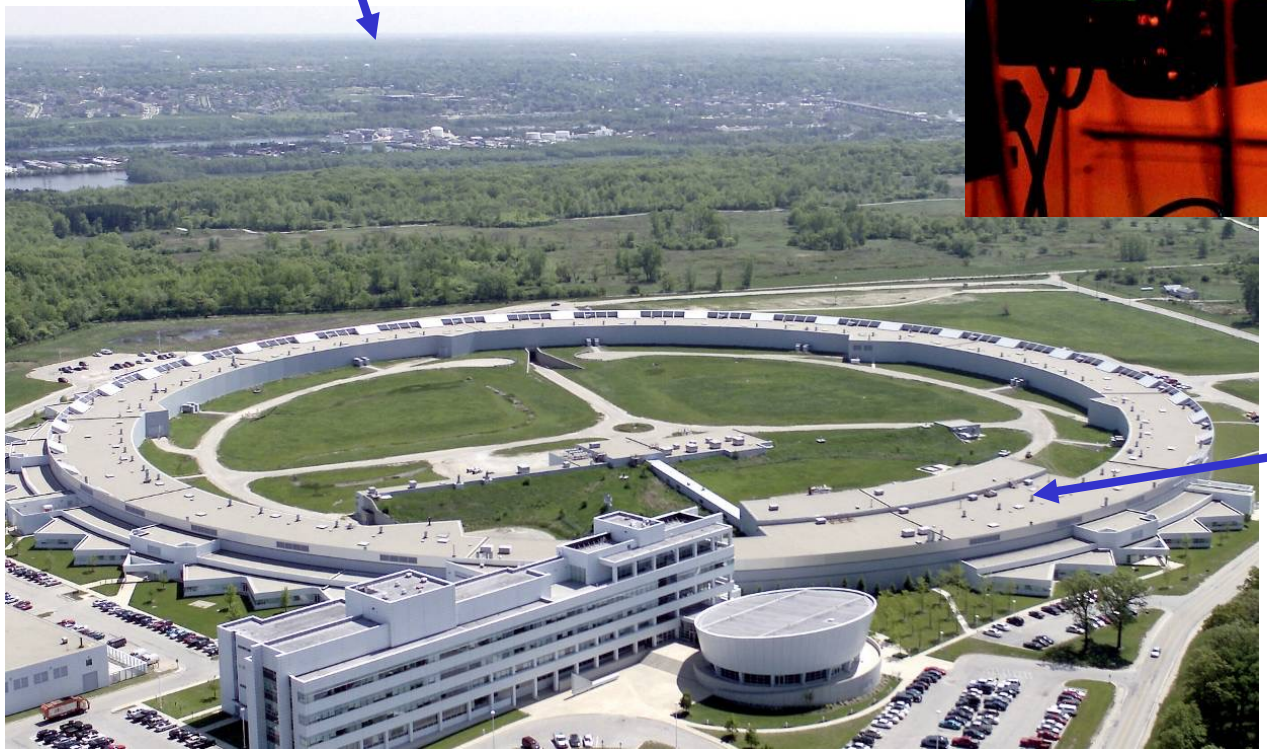
Outline

- Coherent X-ray Diffraction
- How to Solve the Phase Problem
- Nanocrystal Shapes
- Extension to Phase Objects
- Interfacial Contact Forces
- “Anomalous” Thermal Expansion

Synchrotron Radiation

Urbana





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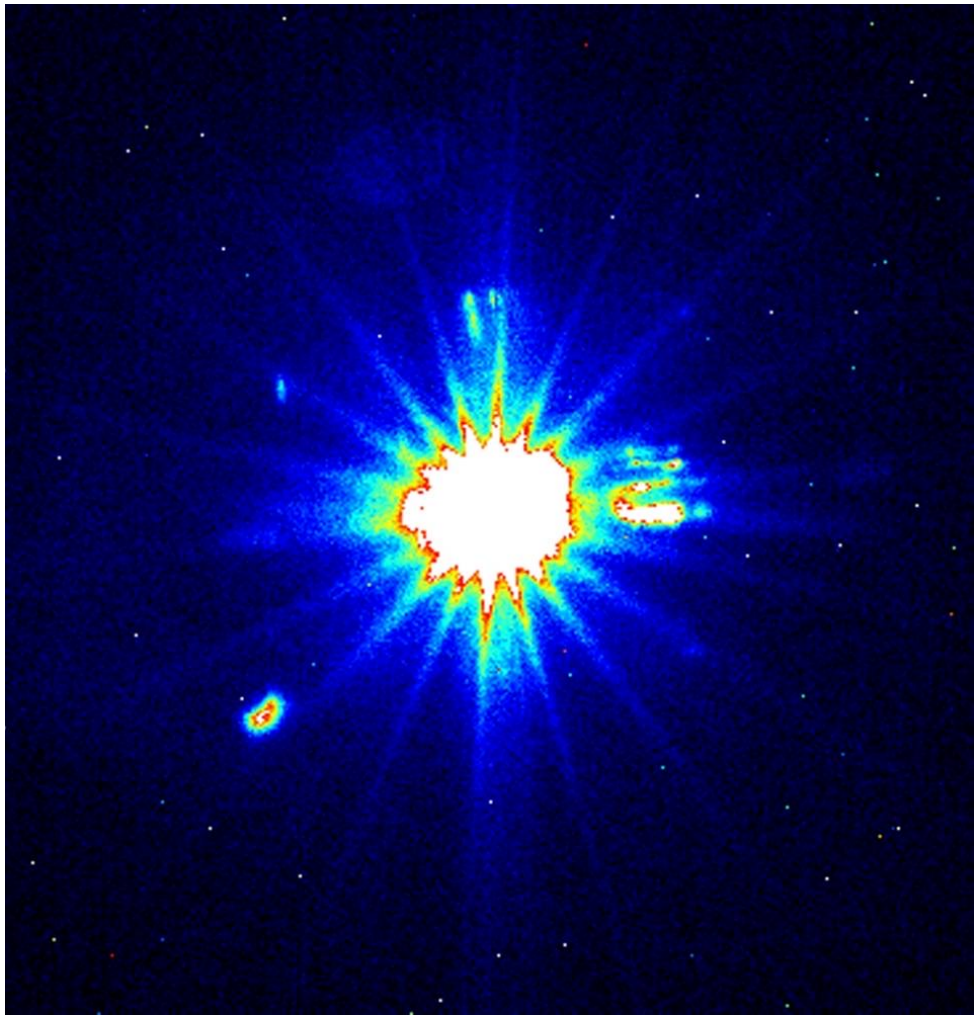
Diamond Light Source (RAL)

Last Girder – March 2006

“First Light” - May 2006

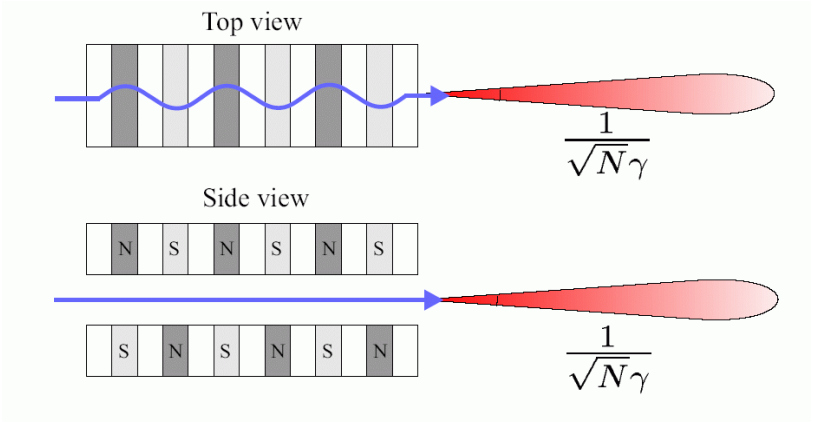
ramped to 3GeV – Sept 2006



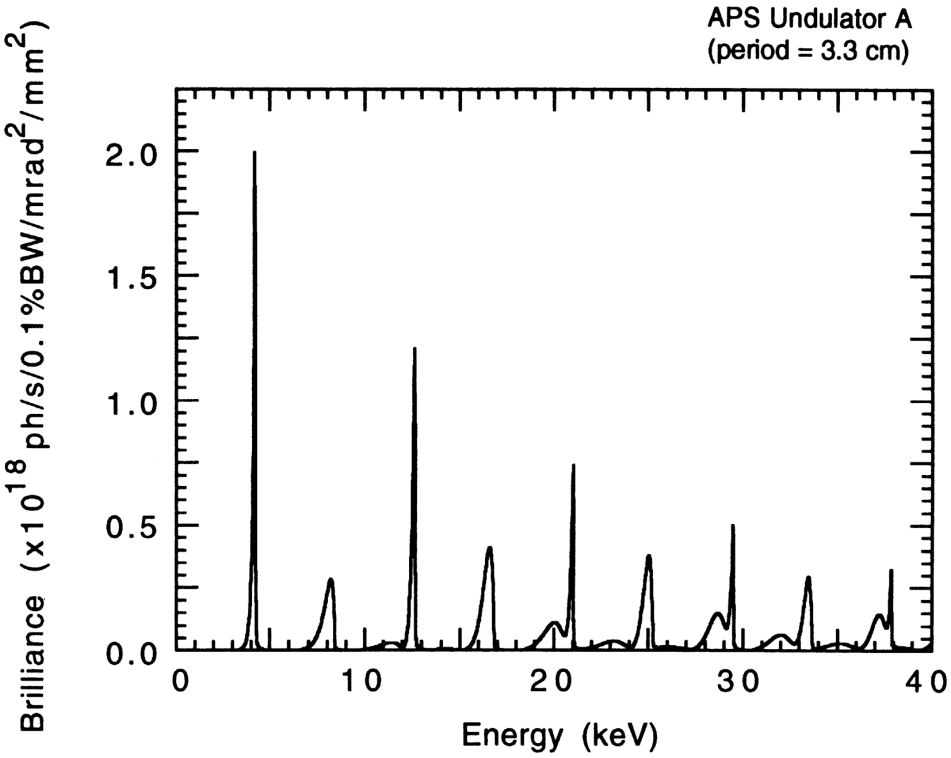
Typical beamline: I-22

Diamond in-vacuum X-ray Undulator

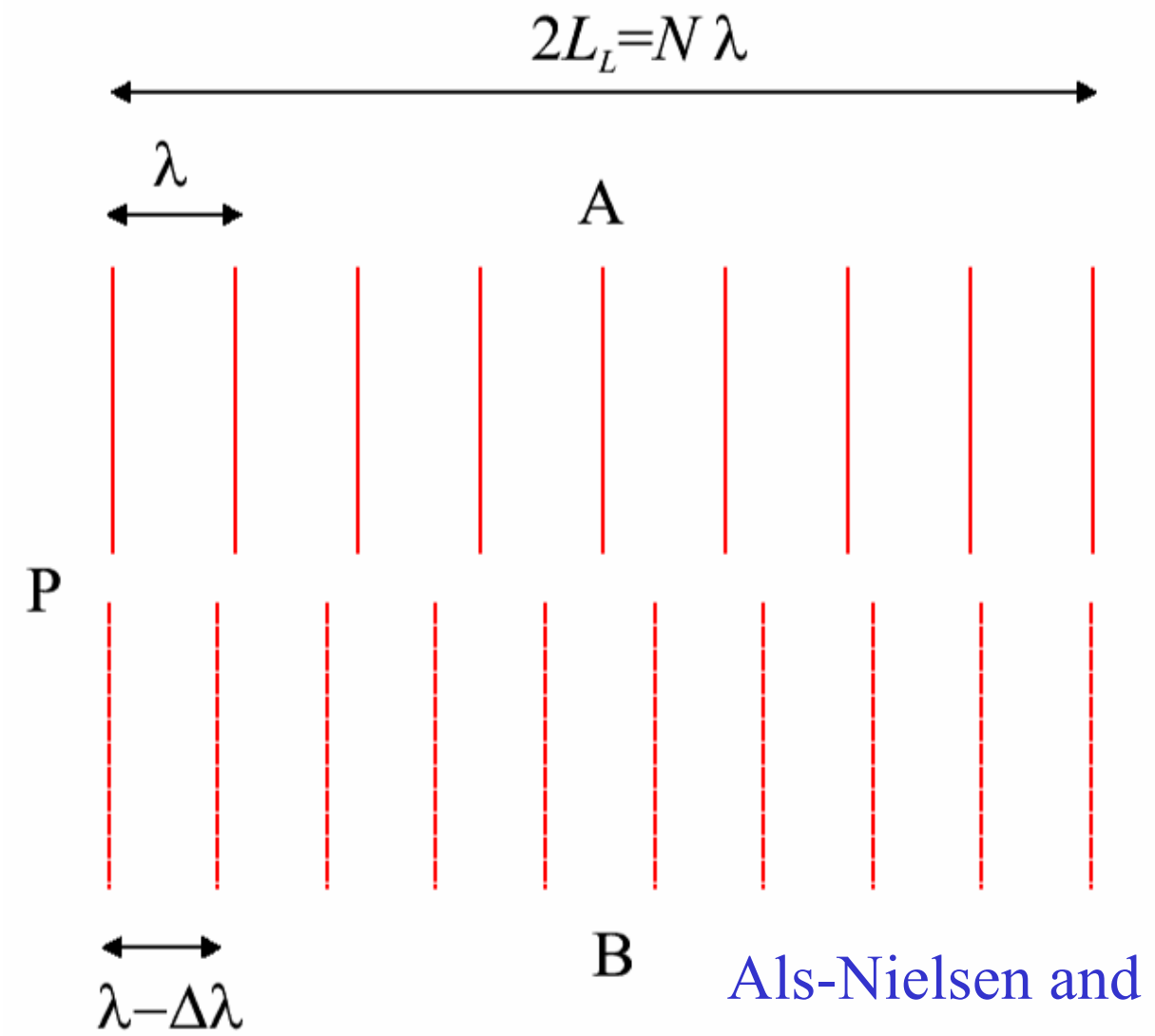
X-ray Undulator Principle



$$\lambda_X = \frac{\lambda_U}{2\gamma^2}\left\{1 + \frac{K^2}{2} + (\gamma\theta)^2\right\}$$



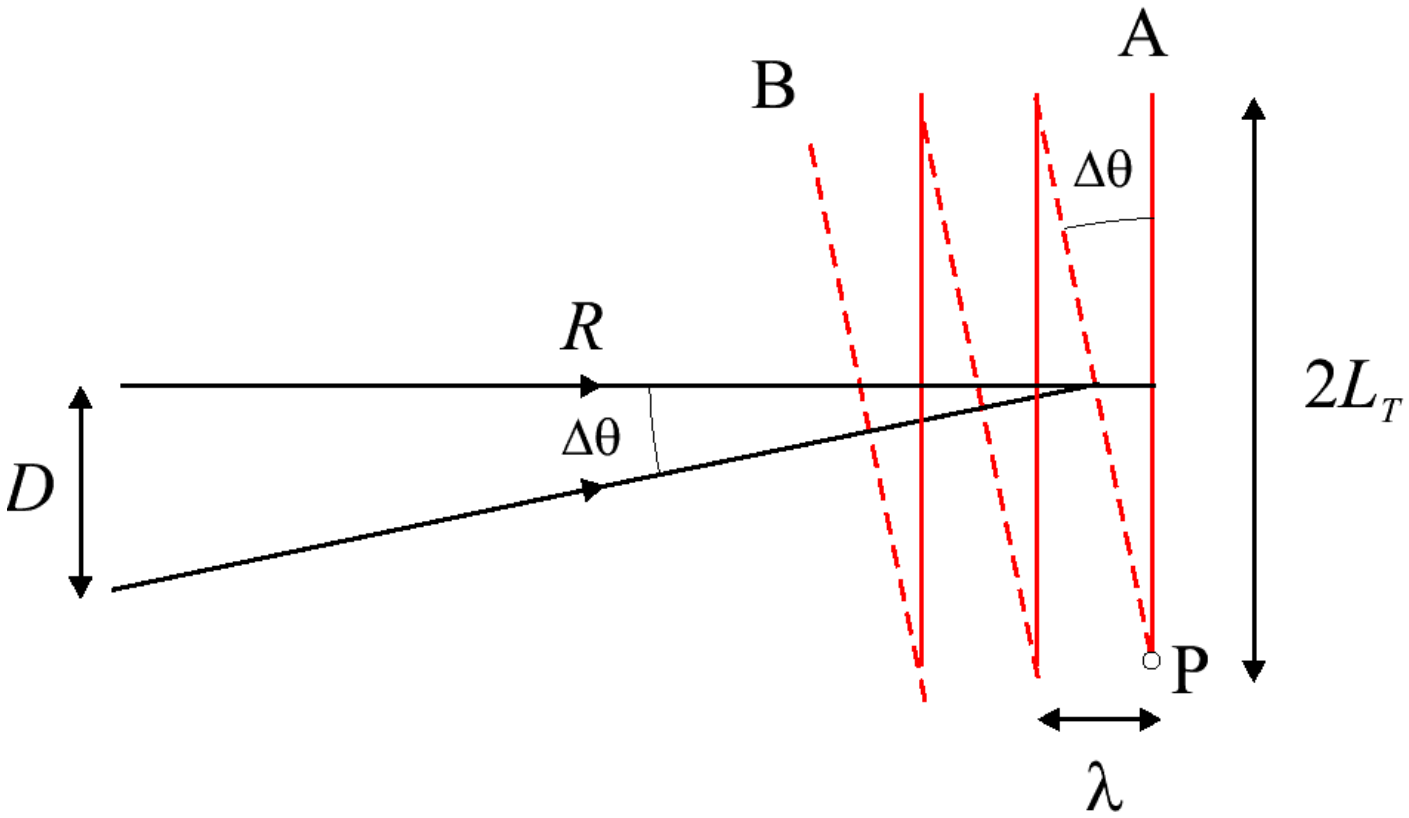
Longitudinal Coherence



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

Als-Nielsen and McMorrow (2001)

Lateral (Transverse) Coherence



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

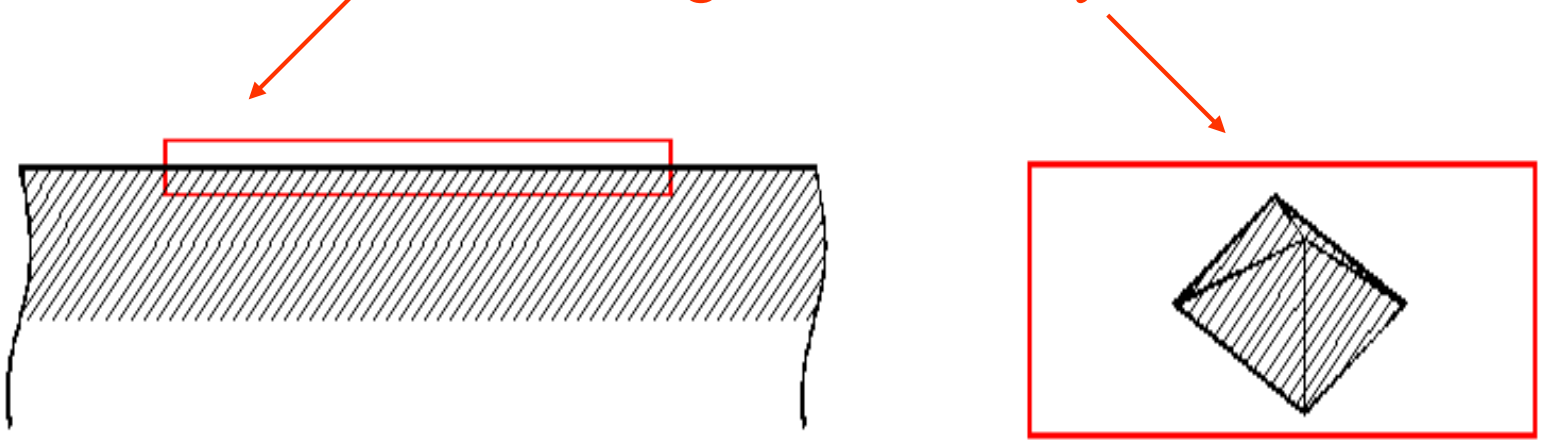
Als-Nielsen and McMorrow (2001)

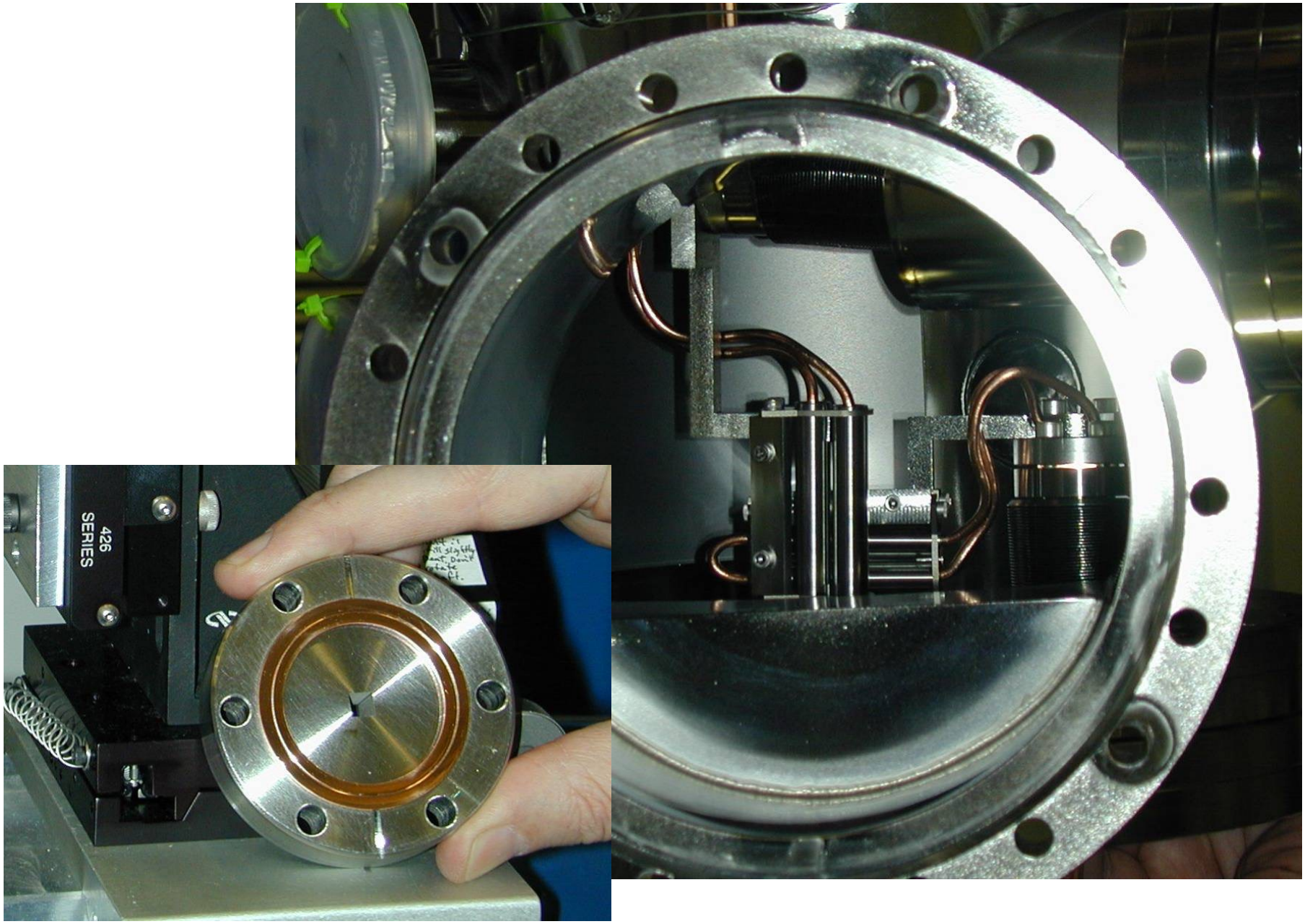
Coherence at APS, ESRF or DLS

Typical of 3rd Generation (undulator) Synchrotron Source

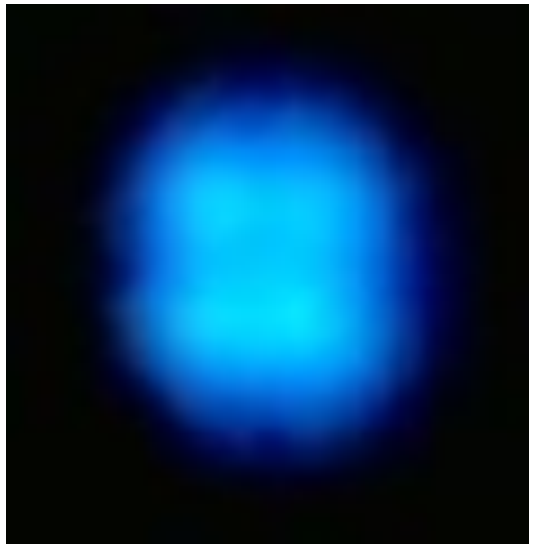
Coherence of	ξ_{VER}	ξ_{HORIZ}	ξ_{LONG}	Flux
Raw Undulator	35μm	9μm	0.004μm	2×10^{12}
Si(111) Monochromator	35μm	9μm	1μm	1×10^{10}
C(111) Monochromator	35μm	9μm	3μm	3×10^{9}

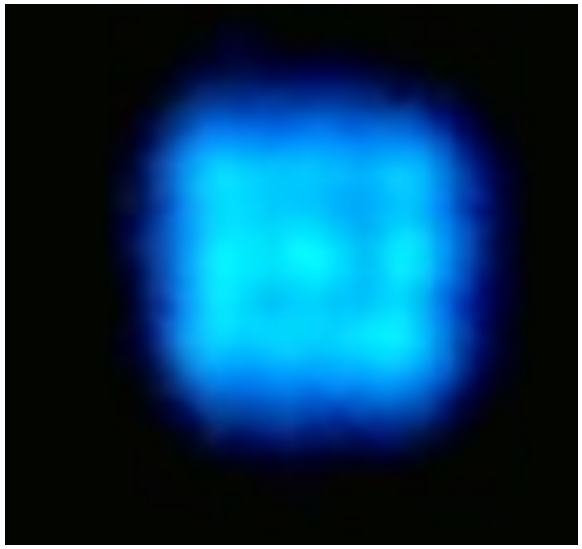
Coherent region defined by slits



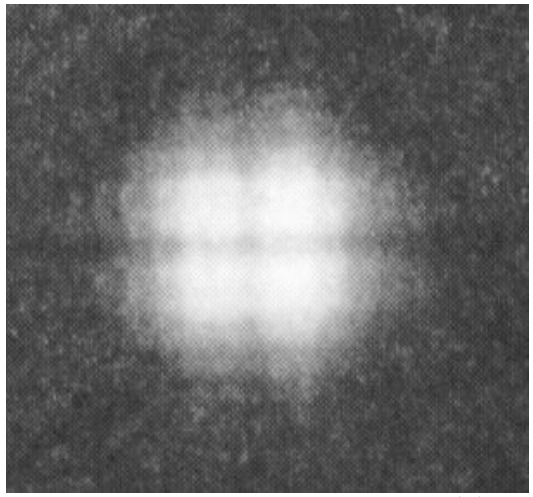

426
SERIES

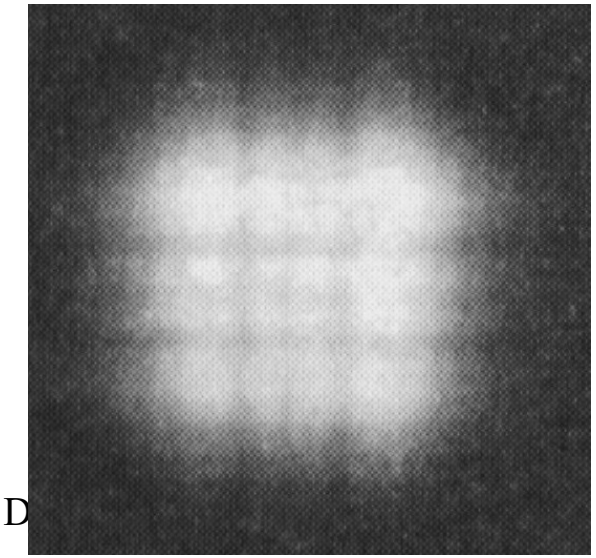
Fresnel Diffraction when $d^2 \sim \lambda D$





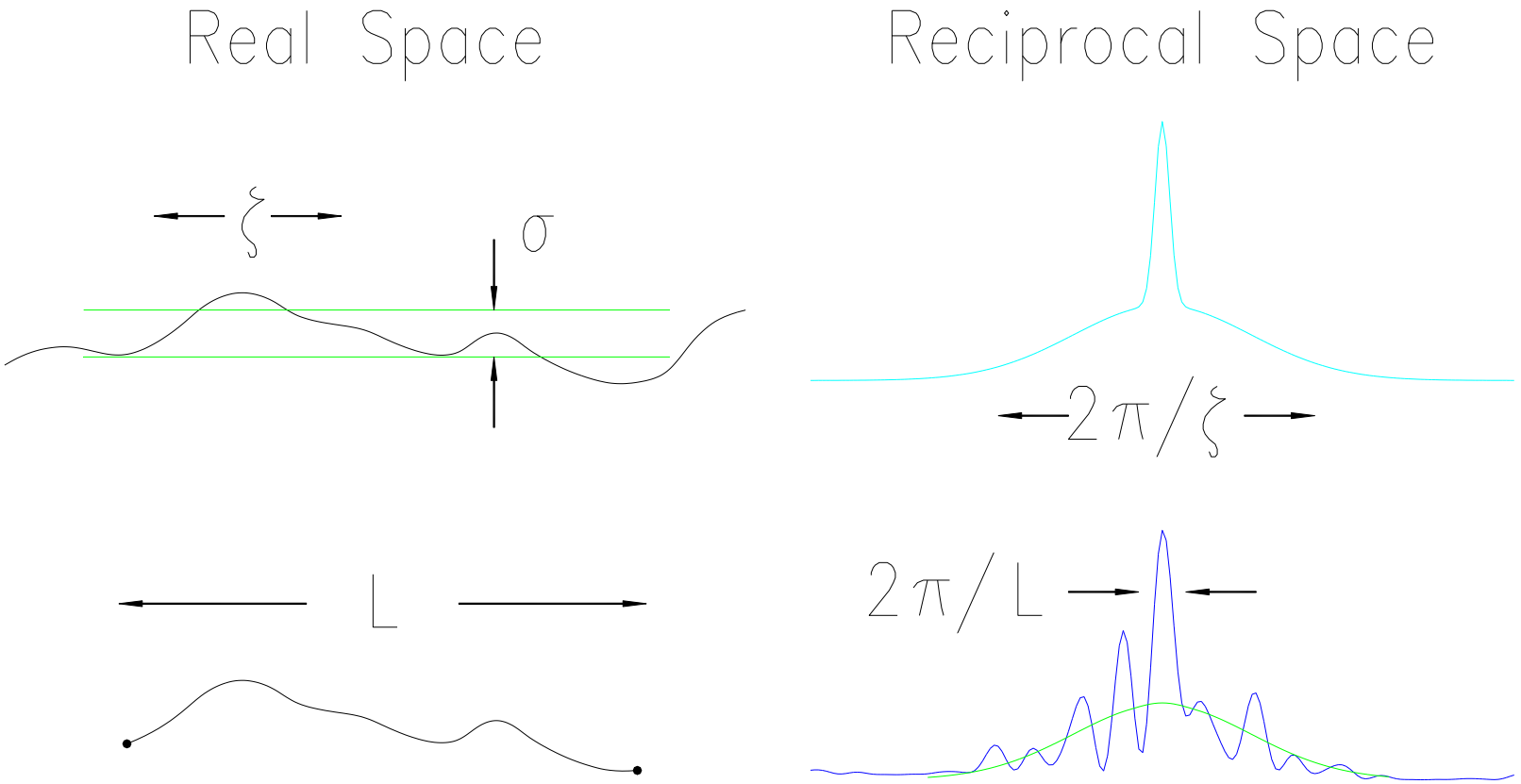
X-ray beam defined by RB slits





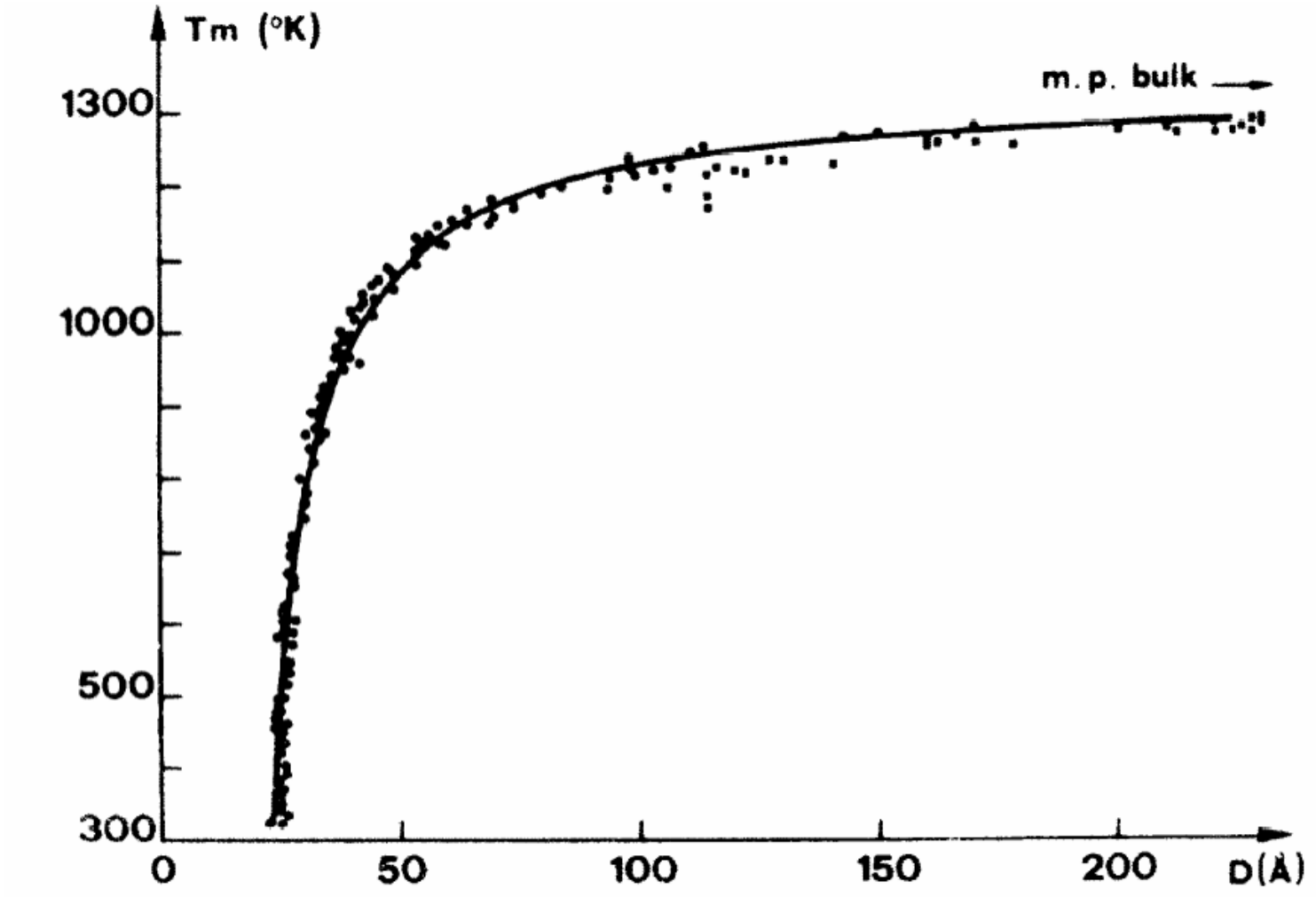
Visible Fresnel diffraction from Hecht "Optics"

Diffuse Scattering acquires Structure using CXD

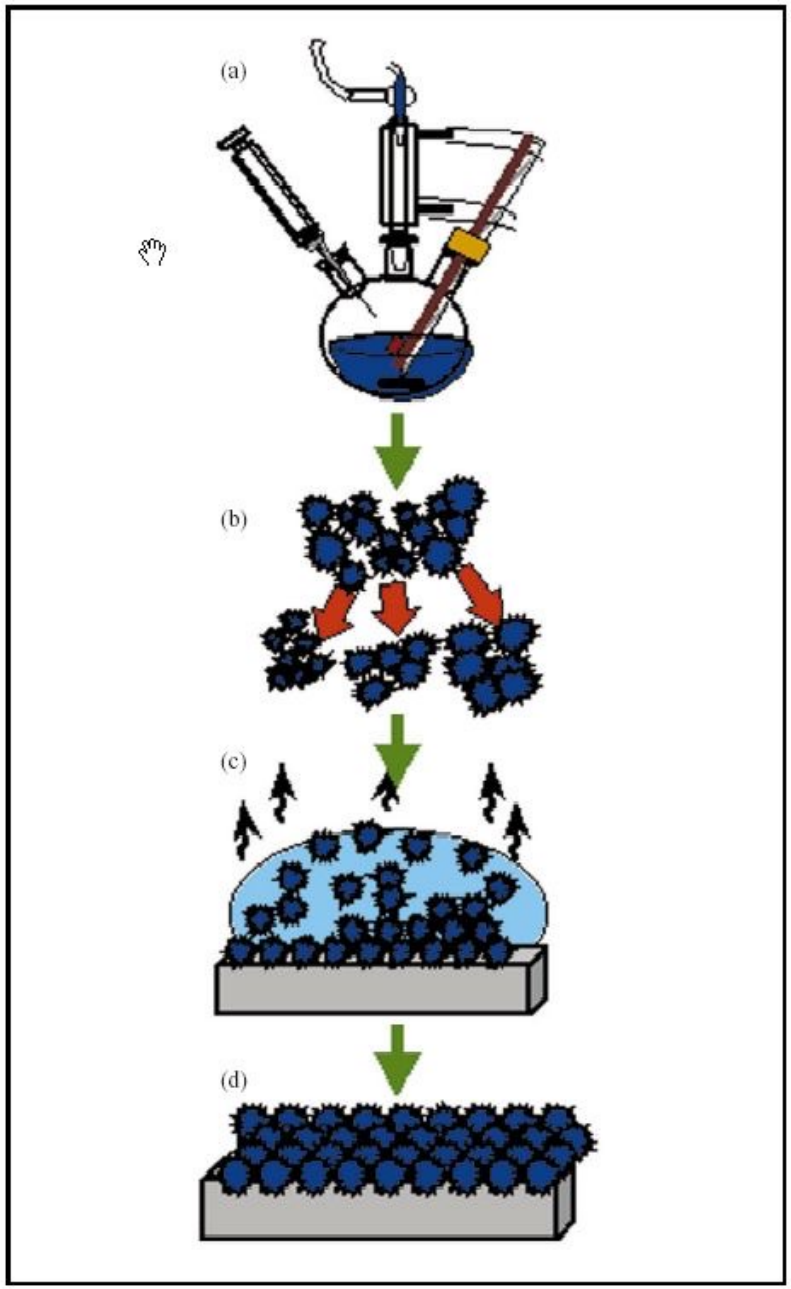


Size-dependent Melting of Au Particles

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

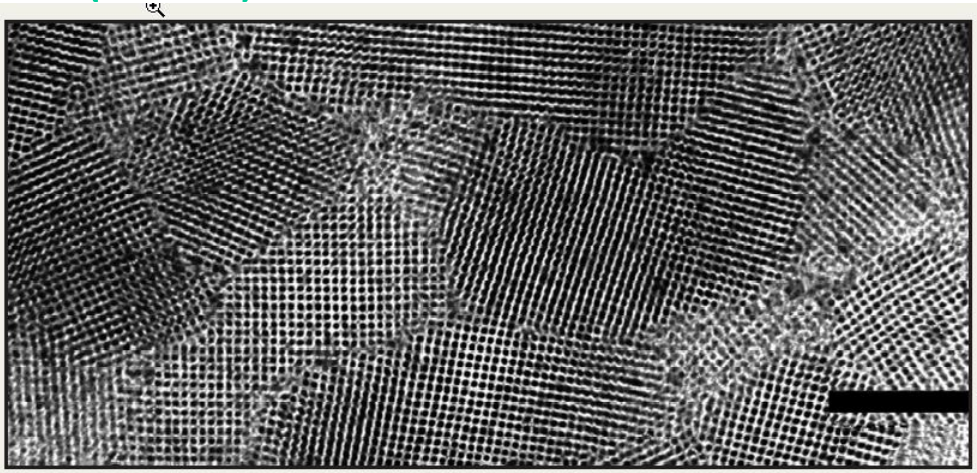


Chemical Synthesis of Nanocrystals

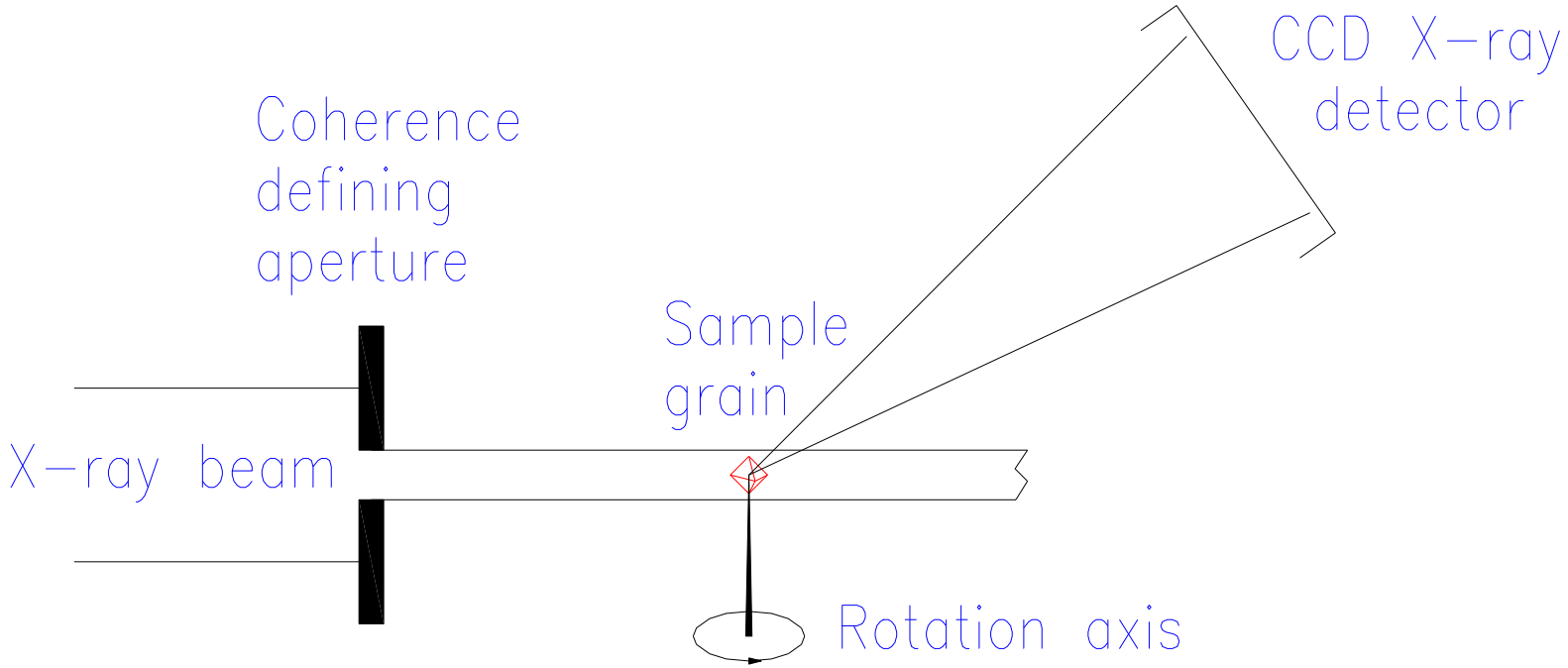


- Reactants introduced rapidly
- High temperature solvent
- Surfactant/organic capping agent
- Square superlattice (200nm scale)

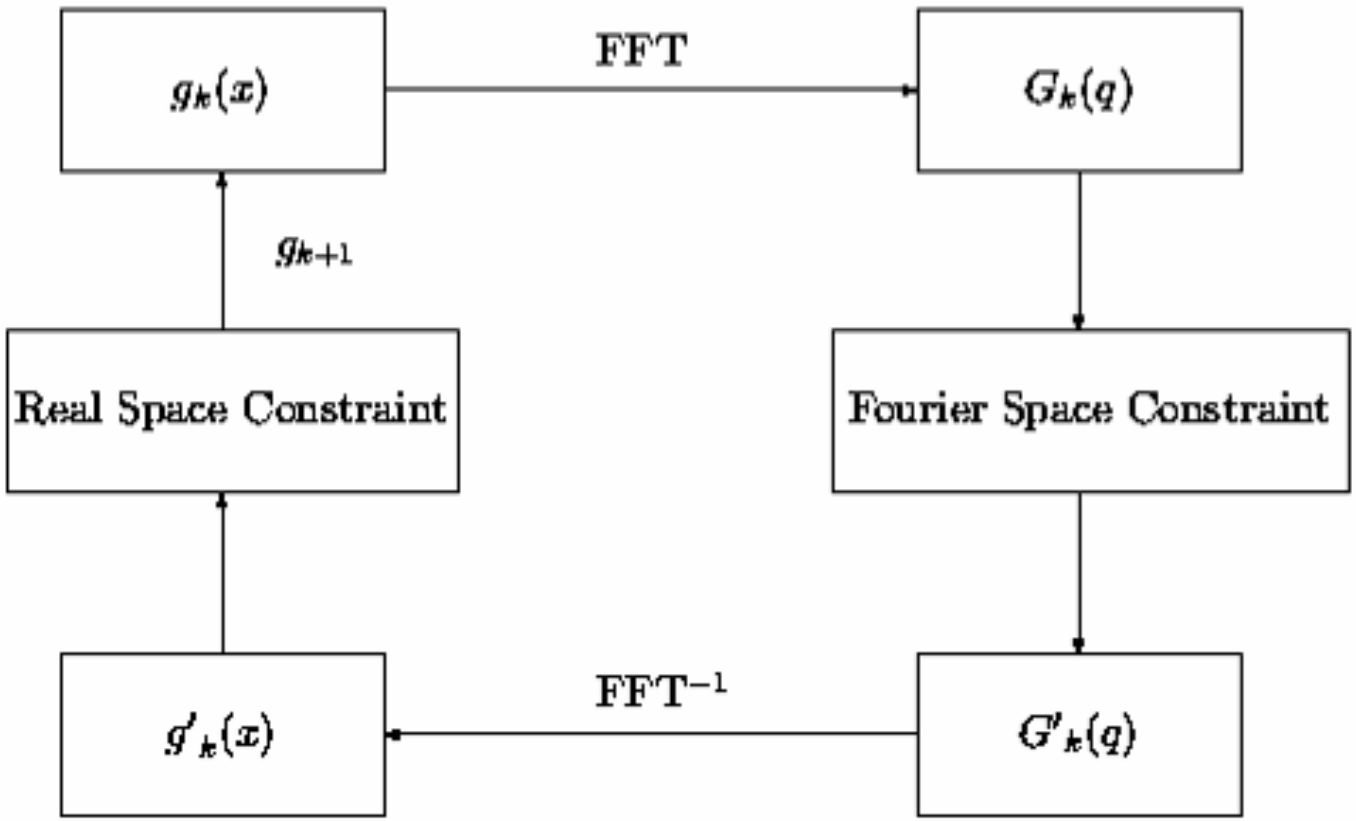
C. B. Murray, IBM J. Res. & Dev. **45** 47 (2001)



Lensless X-ray Microscope



Generic "Error Reduction" method



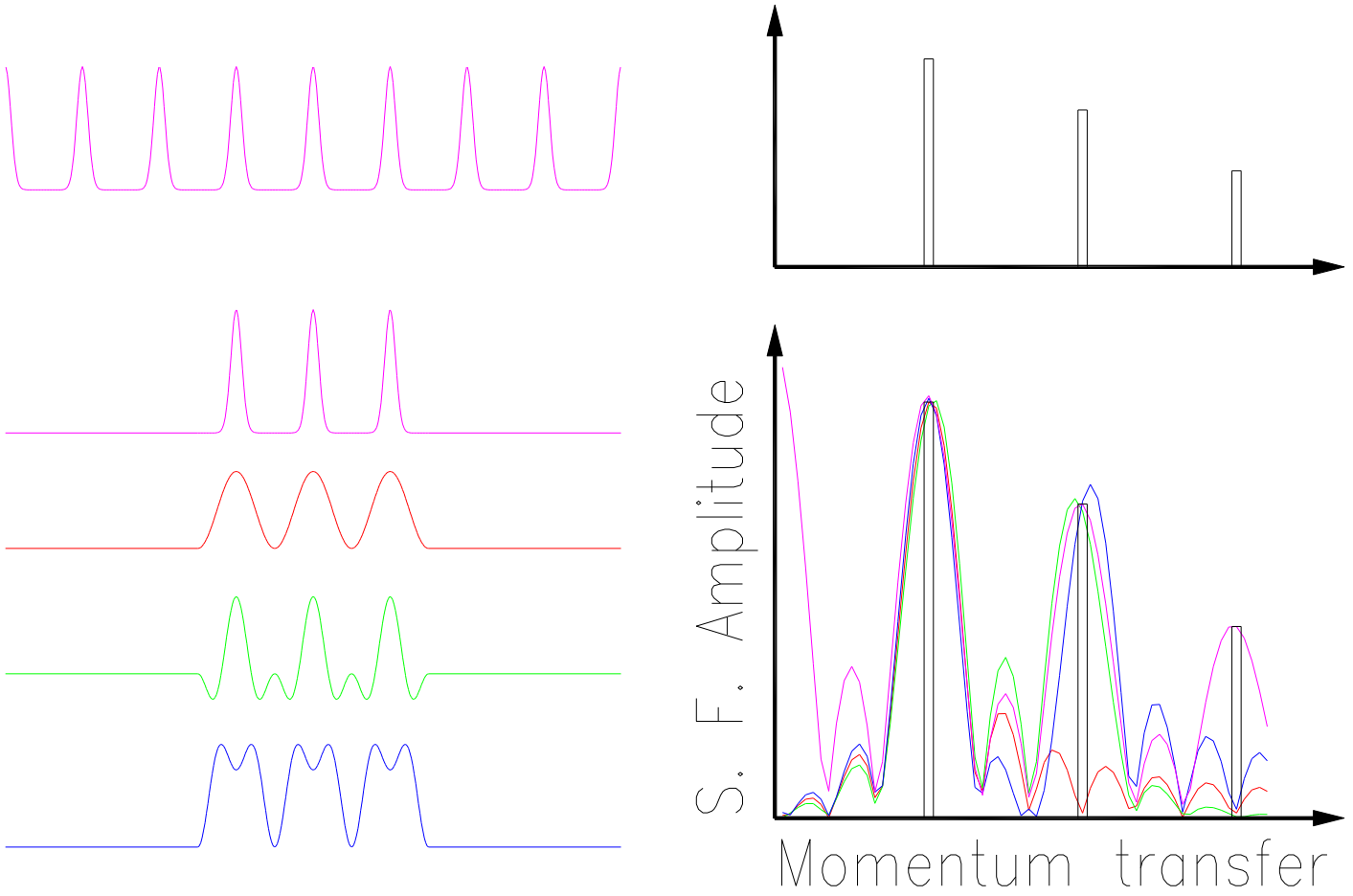
J. R. Fienup Appl. Opt. 21 2758 (1982)
R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)

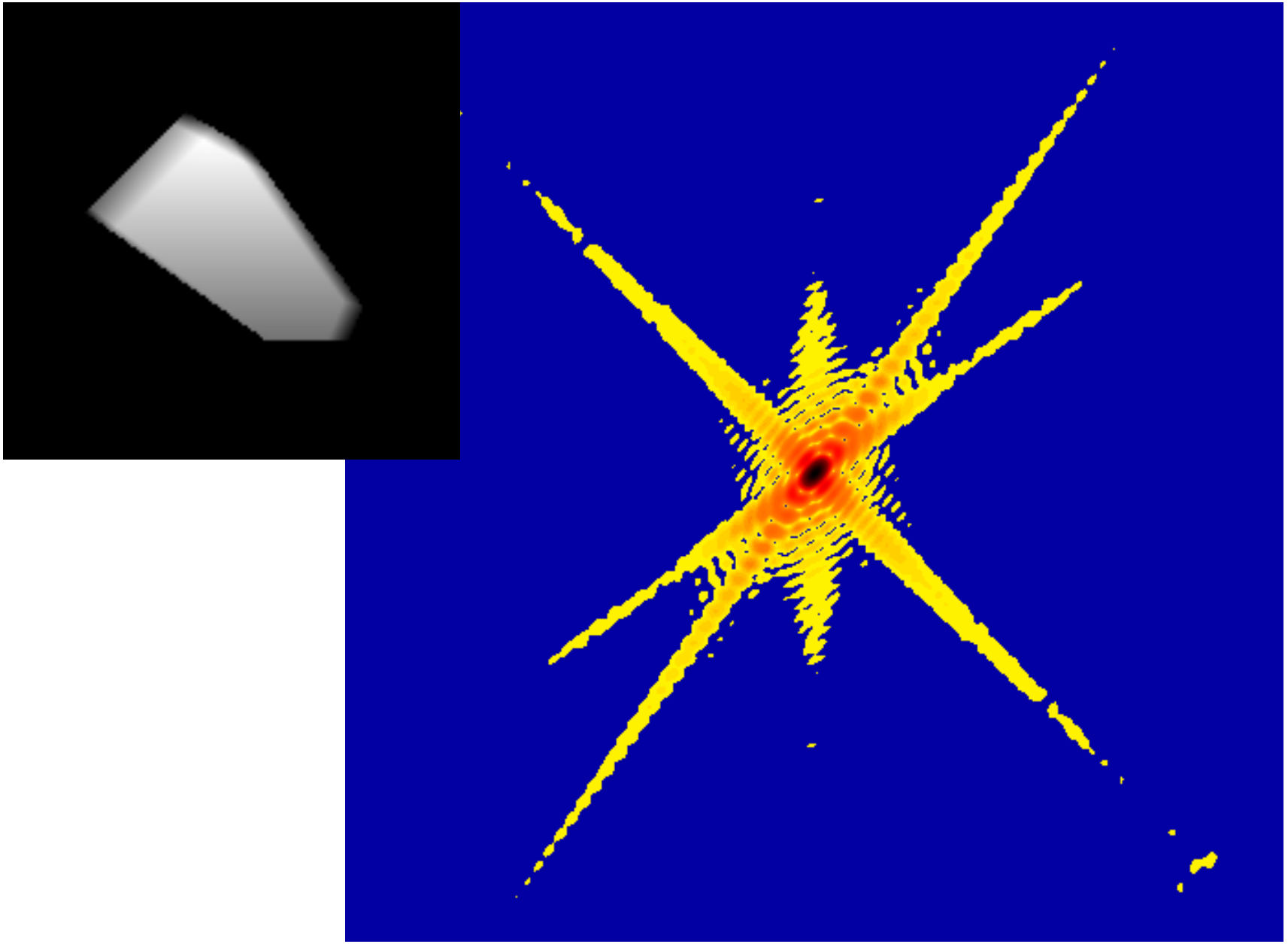
Real-space Constraints in Crystallograhy

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

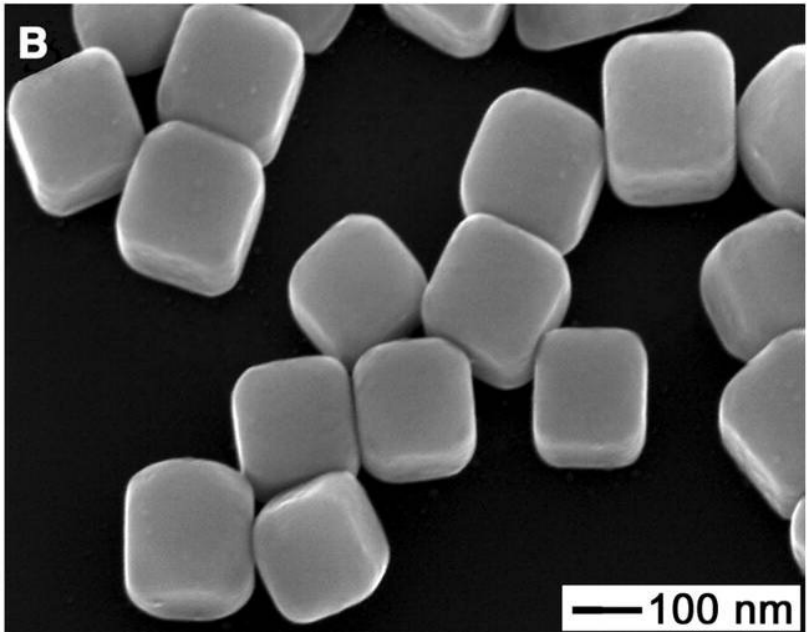
- 'Positivity' and 'Atomicity' constraints (Sayre)
- Finite support, molecular envelope
- Solvent flattening/Molecular replacement
- Non-crystallographic symmetry
- Non-uniqueness is 'pathologically rare' (d>1)
- Uses memory to avoid stagnation (Fienup HIO)

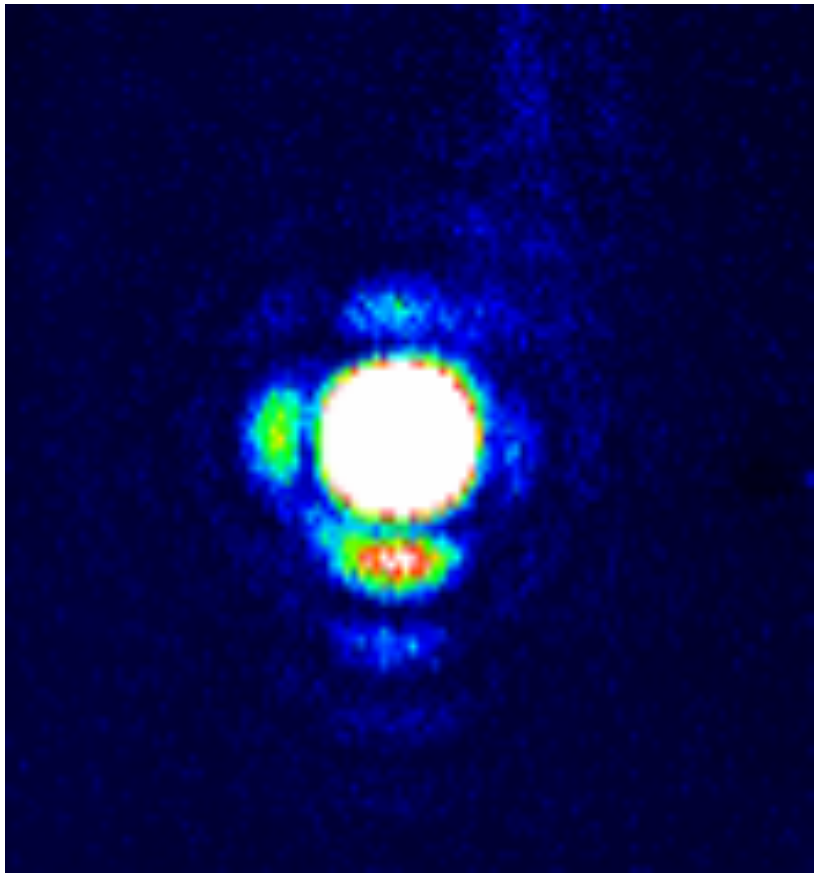
Oversampling solves Phase Problem





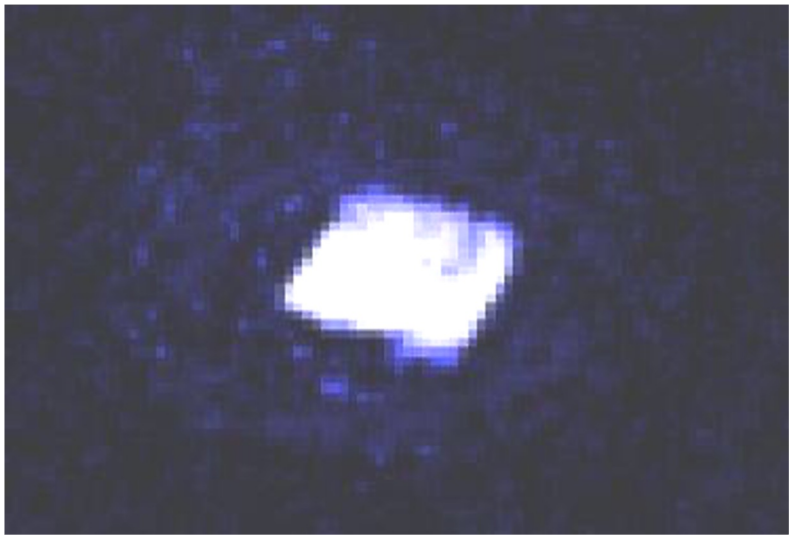
Chemically Synthesized Silver Nanocubes



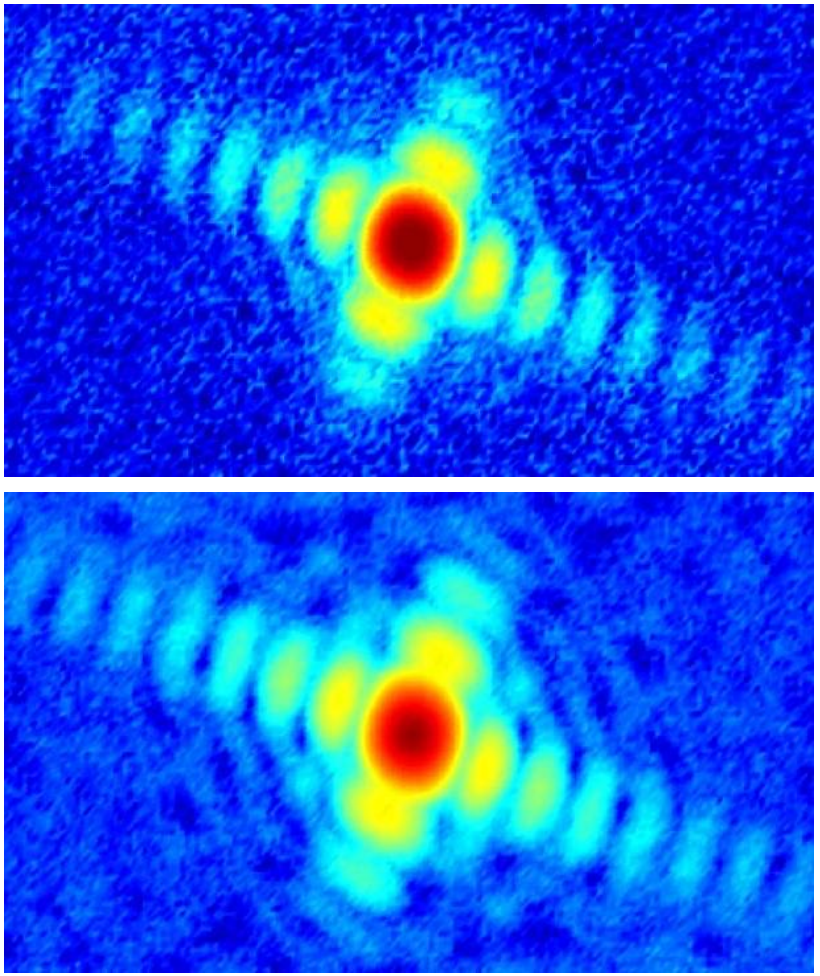


Yugang Sun and Younan Xia,
Science 298 2177 (2003)

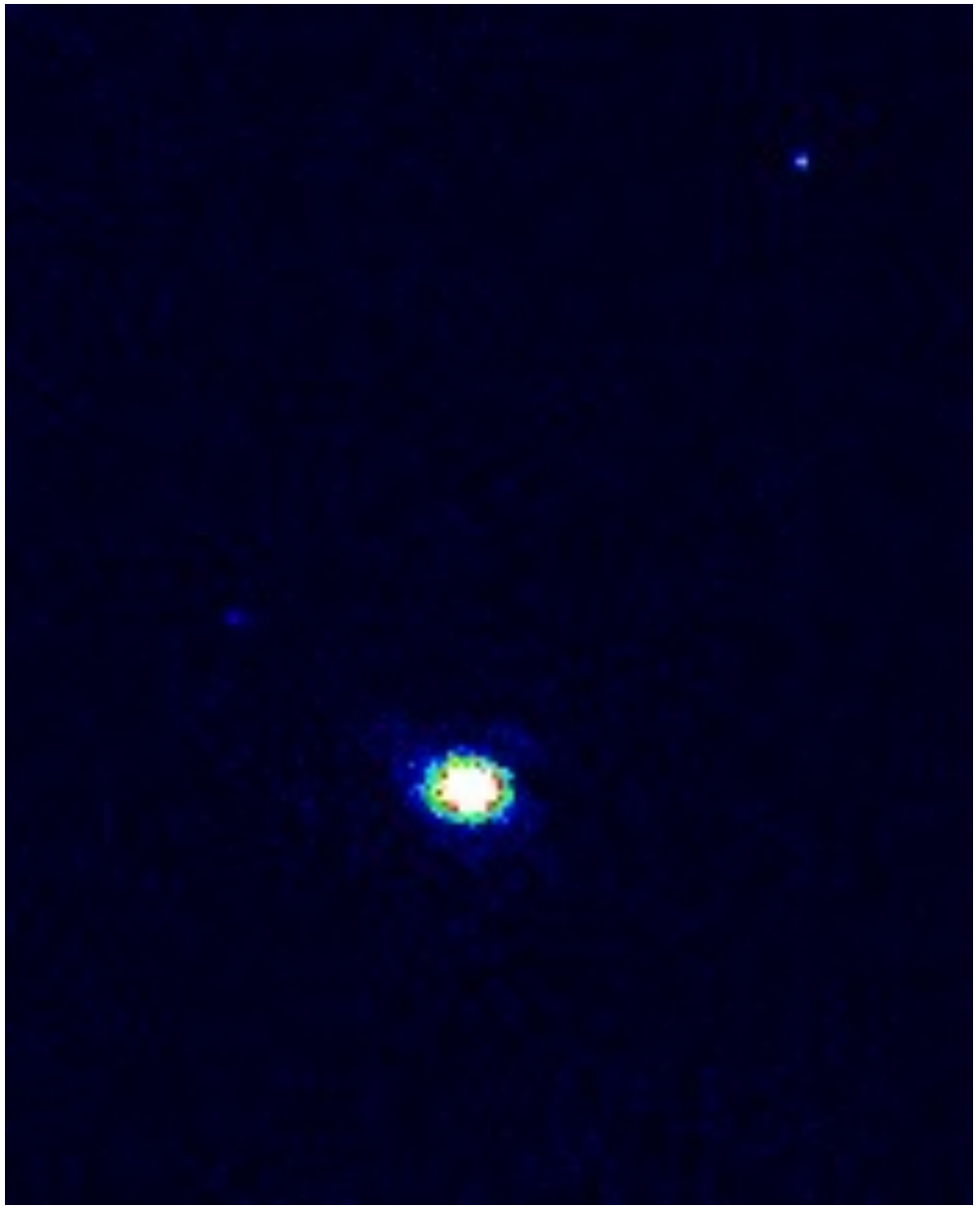
Reconstruction of Ag Nanocrystal

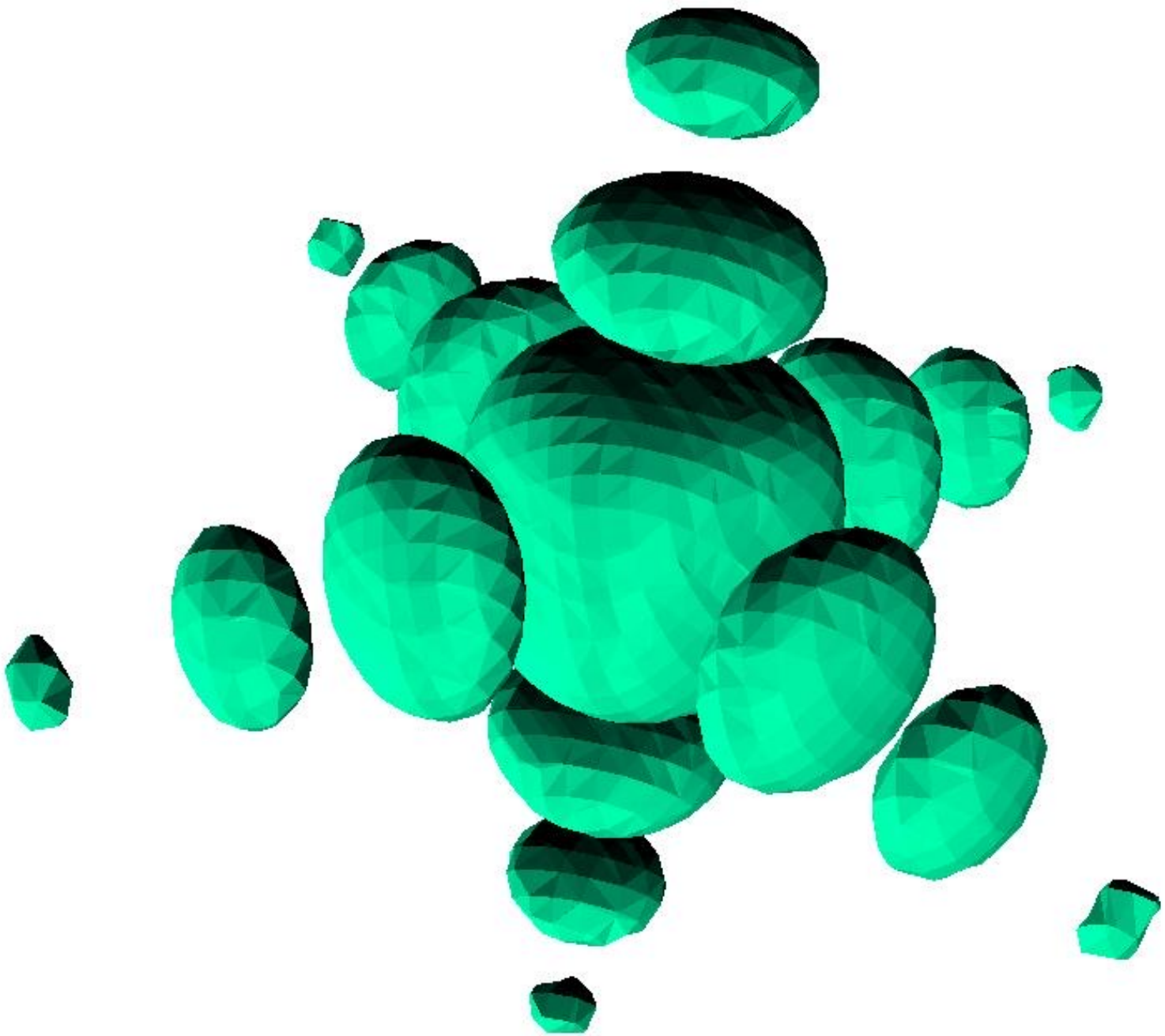


200nm

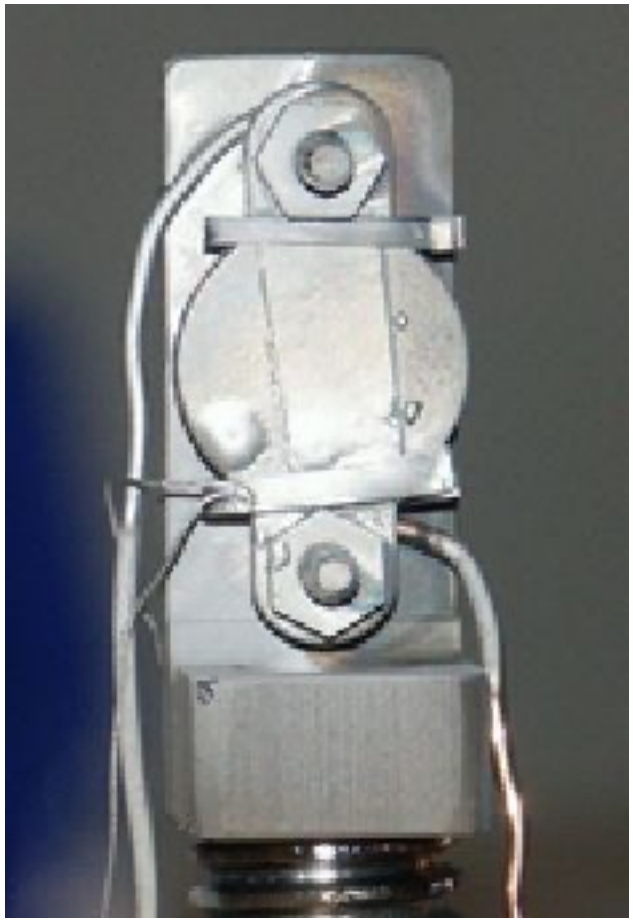


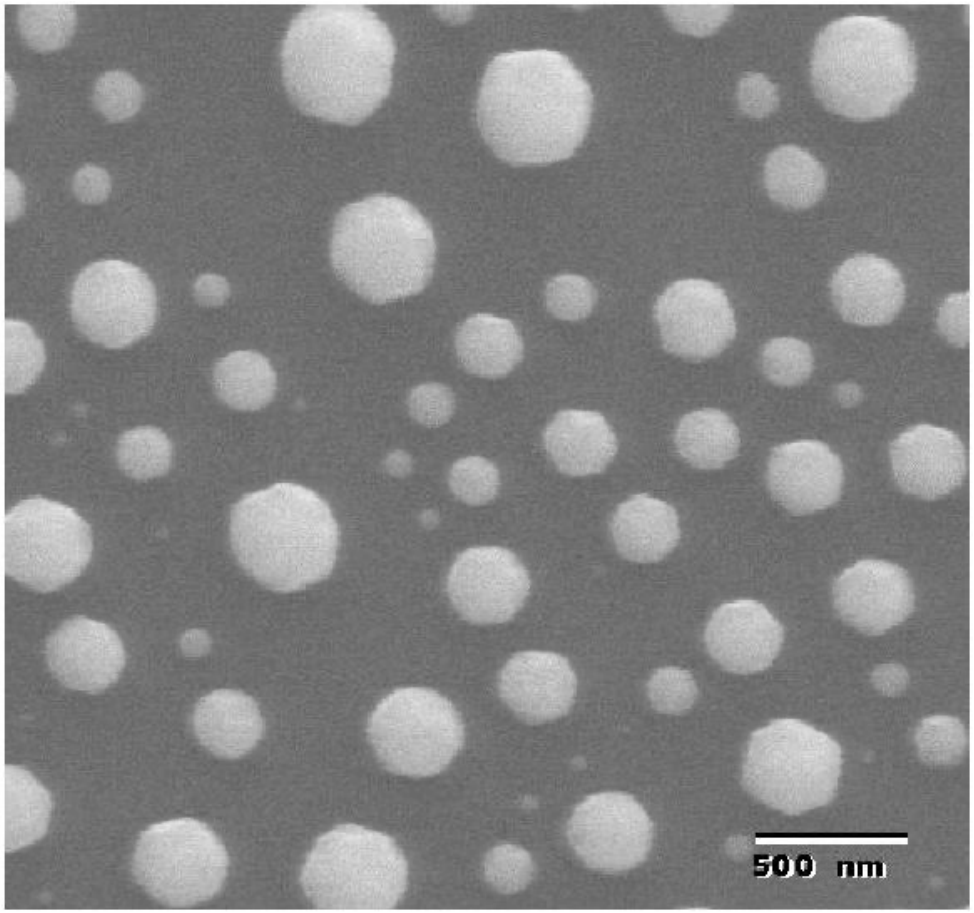
Rocking scan of Ag cubes with 0.01° steps



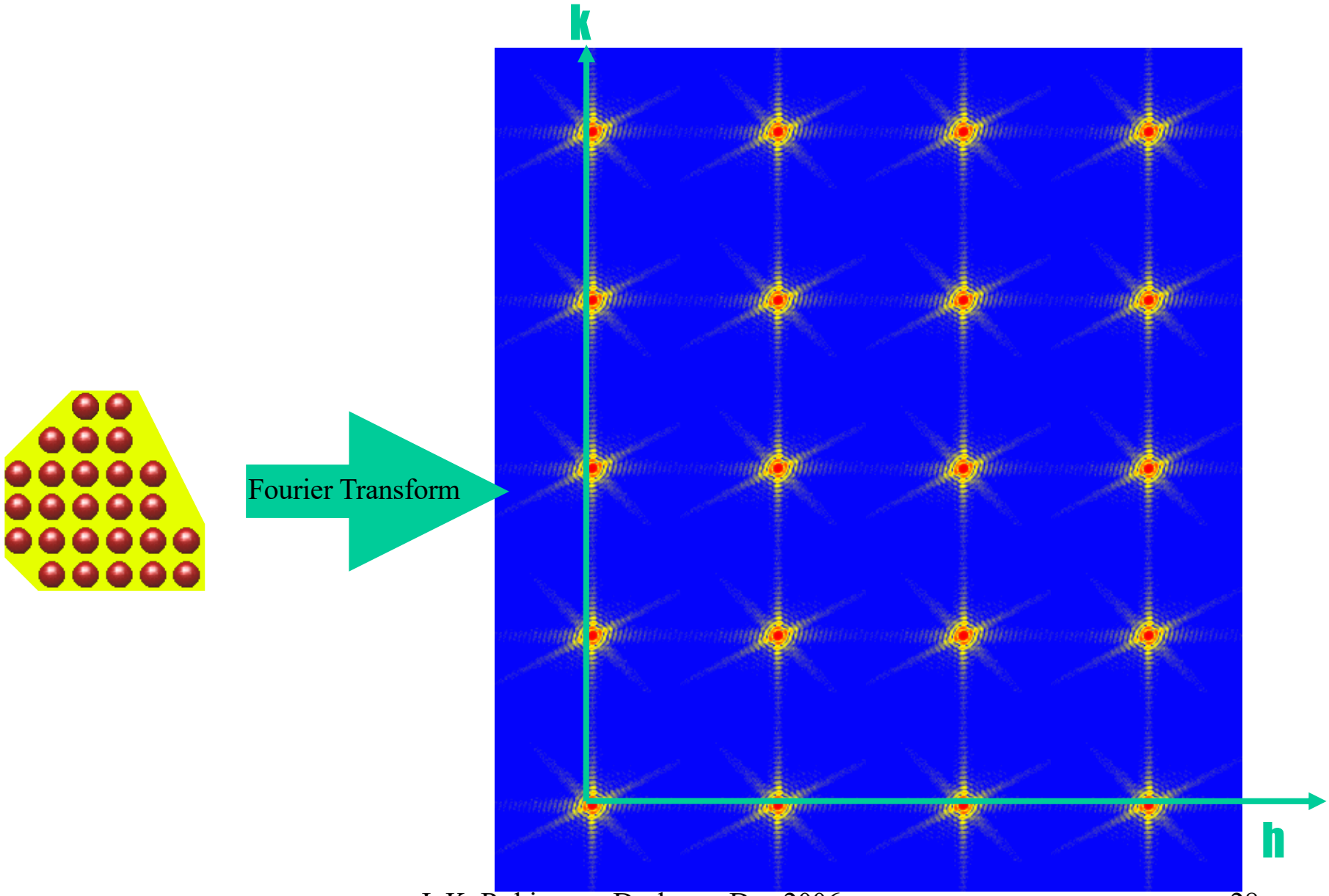


In situ growth of Pb crystals





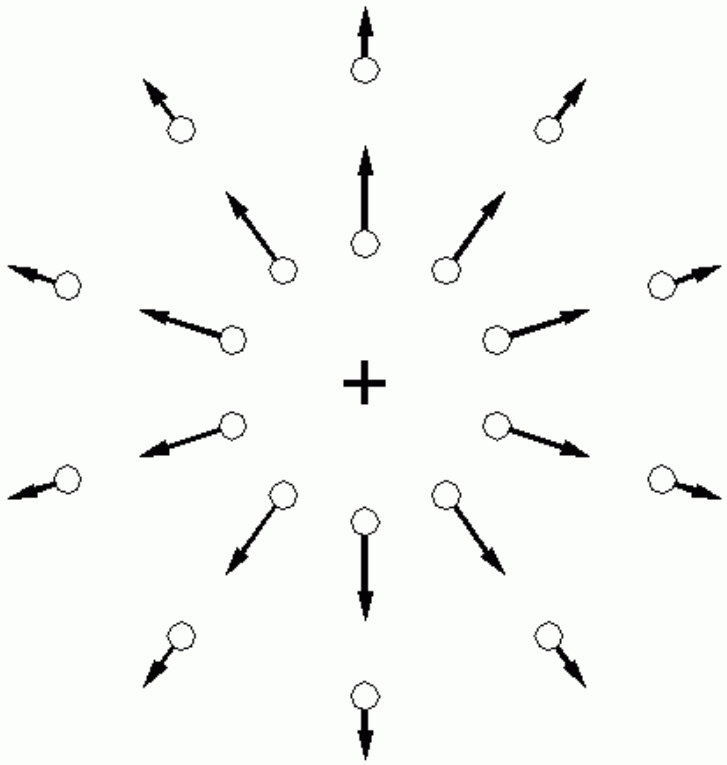
Coherent Diffraction from Crystals

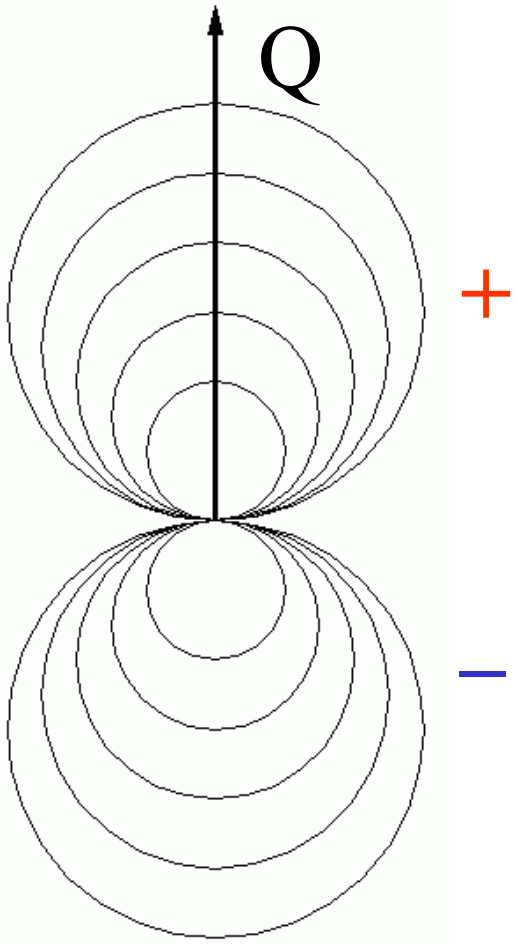


Diffraction by Strain of Point Defect

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

Imaginary density





Good statistics, 3D diffraction data

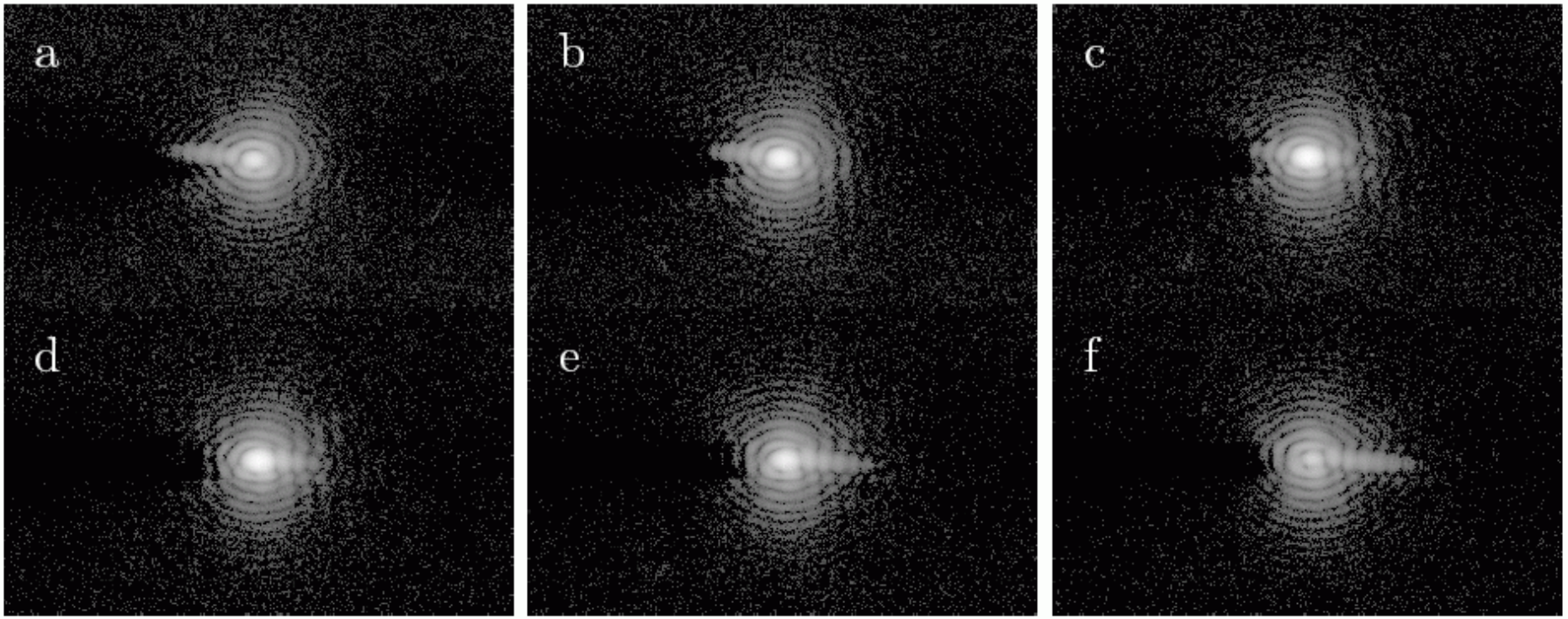
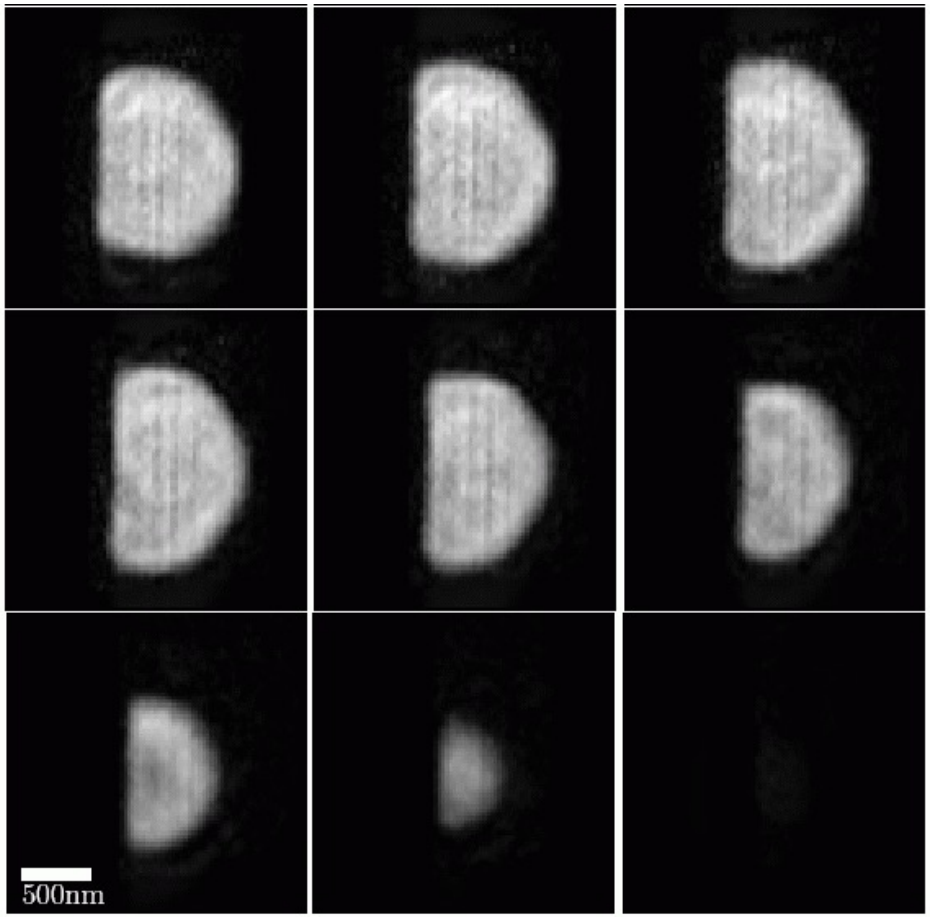
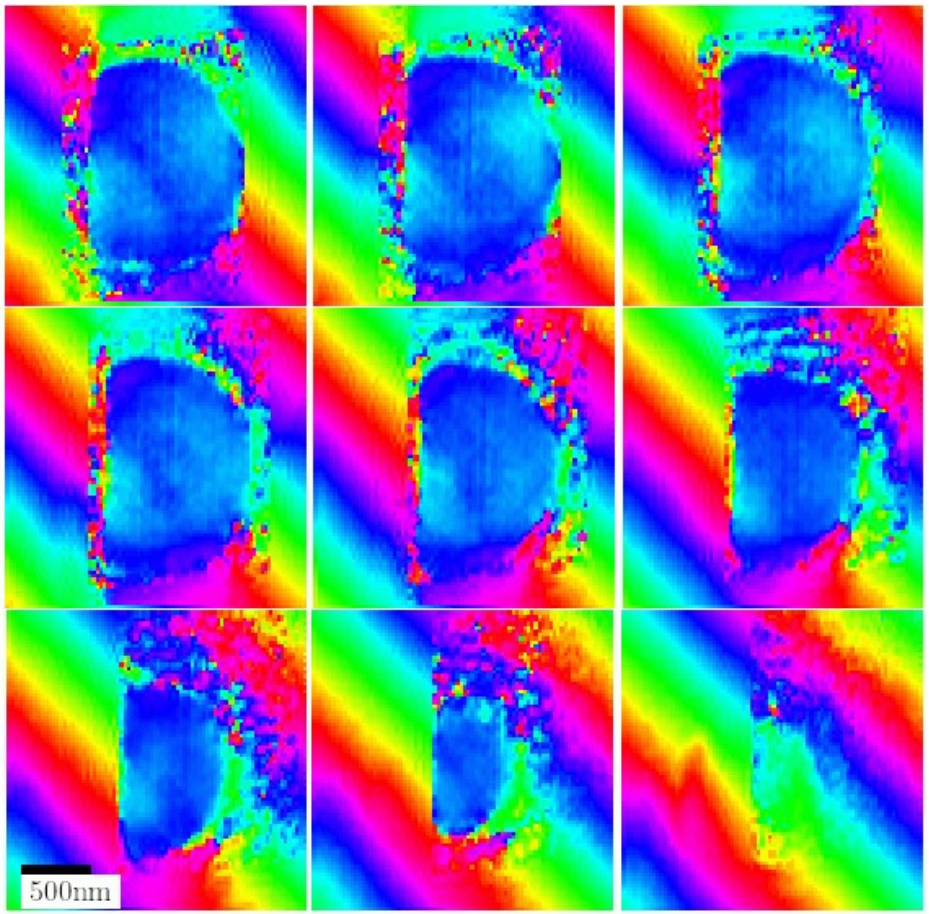
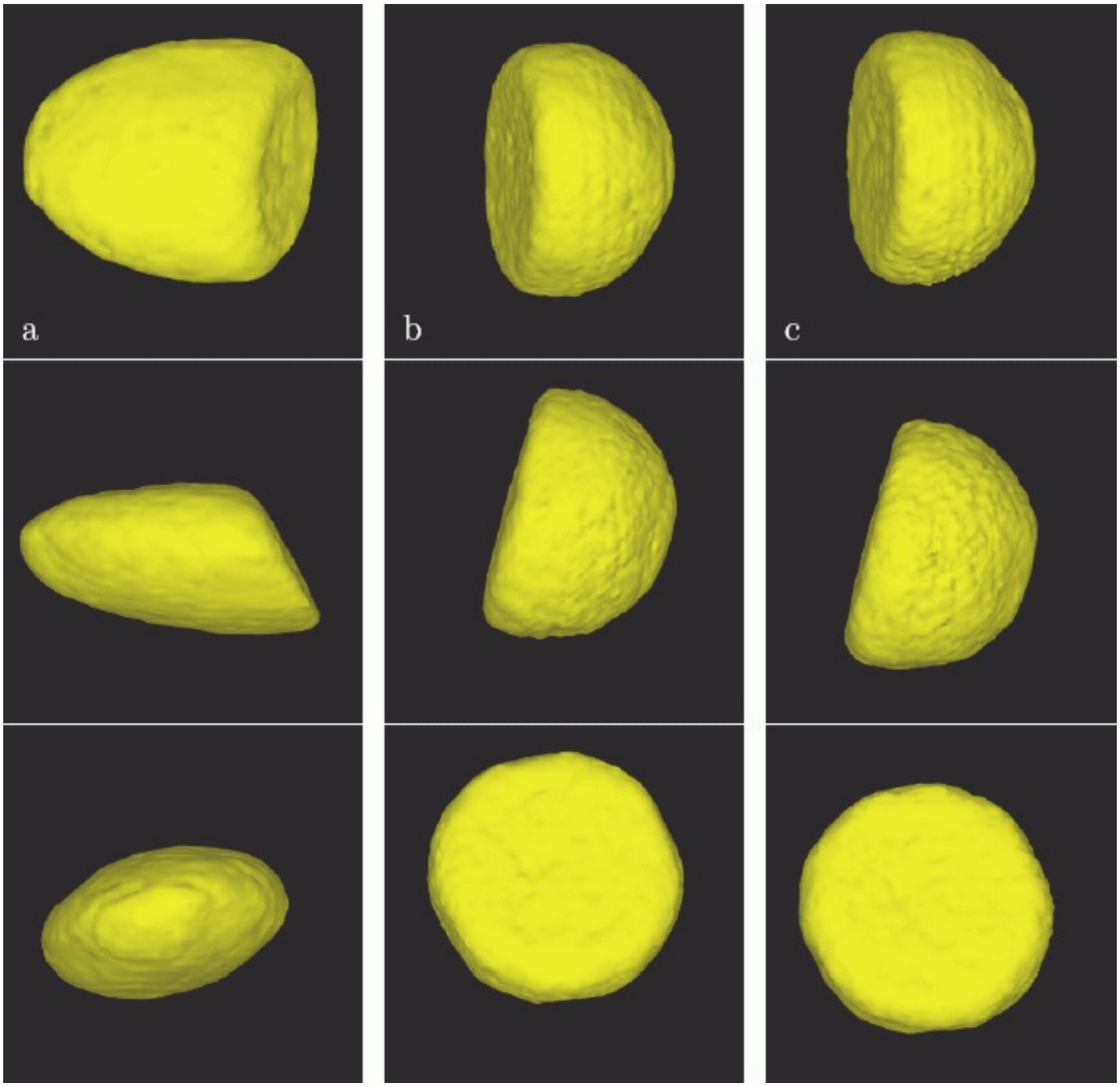


Figure 4.12: Center slices from 3D CXD pattern from Pb sample, on a log scale. Data file 296 from 10/03.

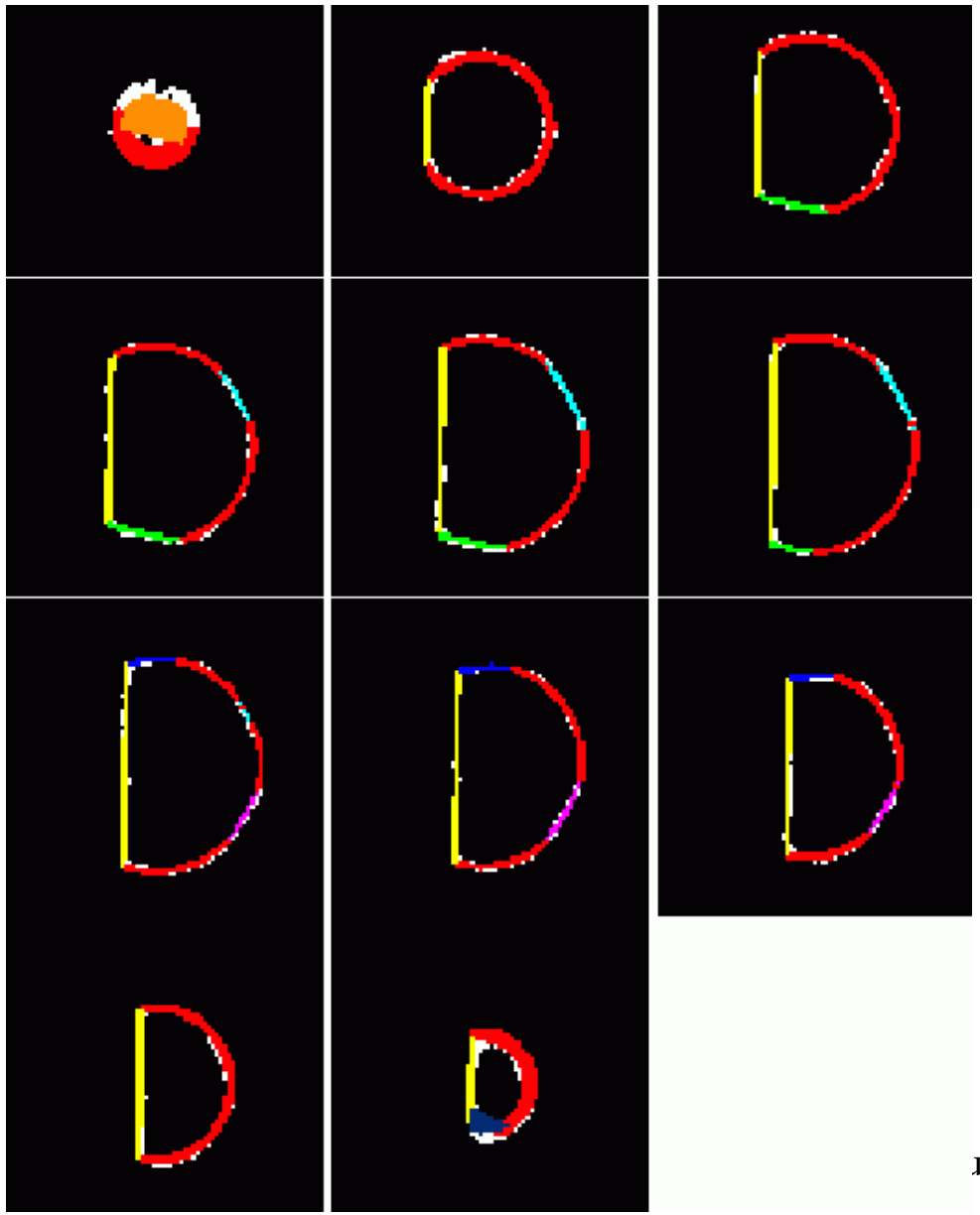
Complex Density (amplitude *and* phase)





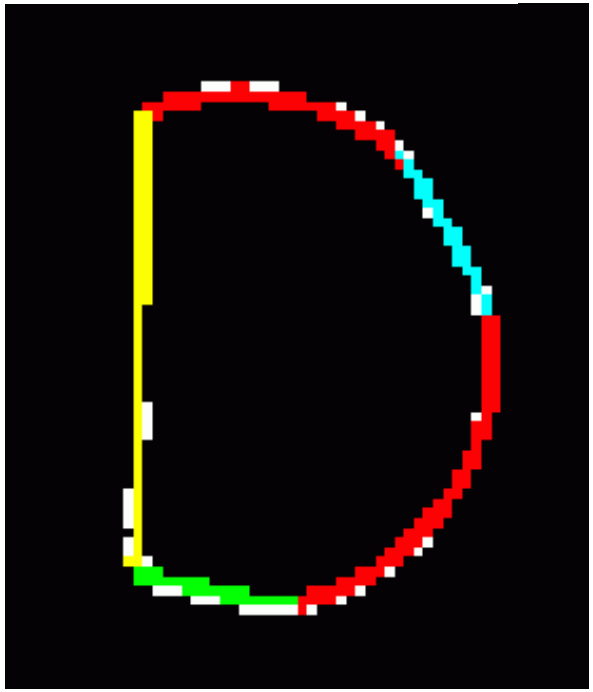

a
b
c

Fitting to facetted shape

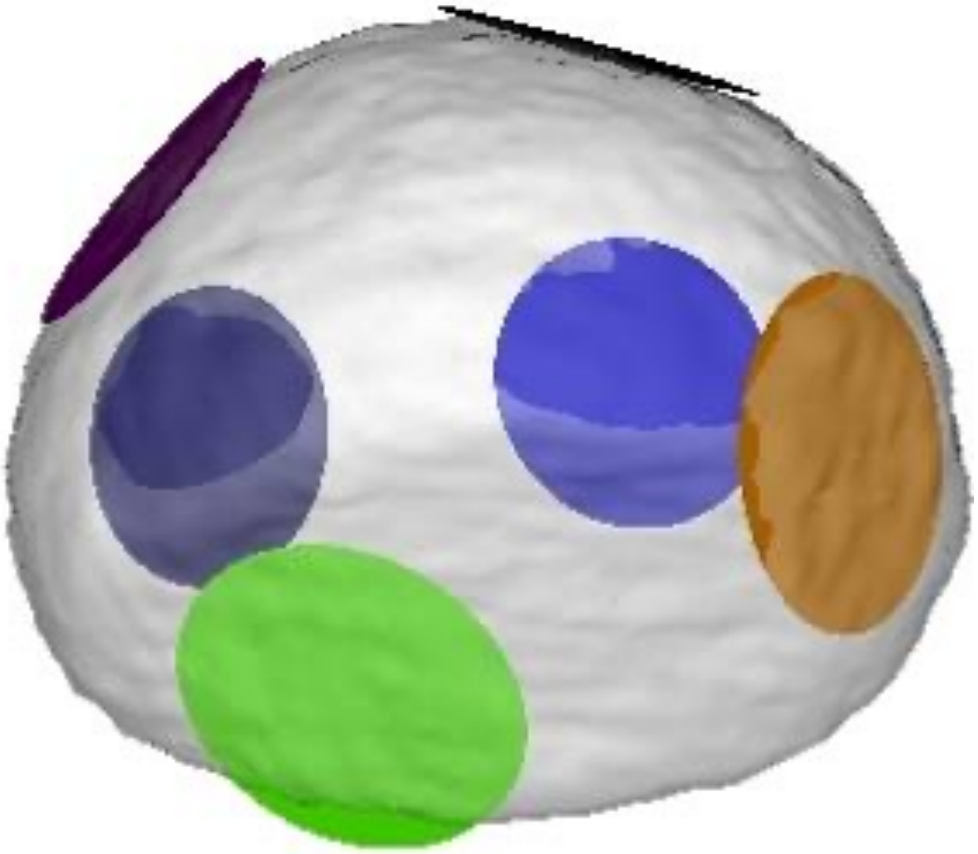


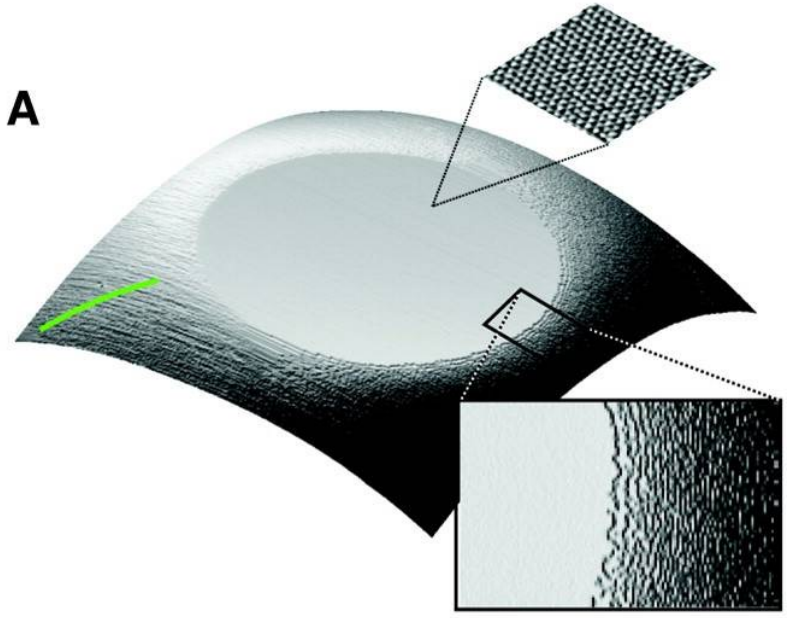
	$\|R\|$		P0	P1	P2	P3	P4	P5	P6
	9.4	P0	0	85	149	79	134	106	71
	25.7	P1		0	123	164	83	76	102
	25.1	P2			0	72	67	74	110
	25.9	P3				0	111	106	76
	25.4	P4					0	113	68
	25.4	P5						0	176
	26.0	P6							0 0

Angles between facets



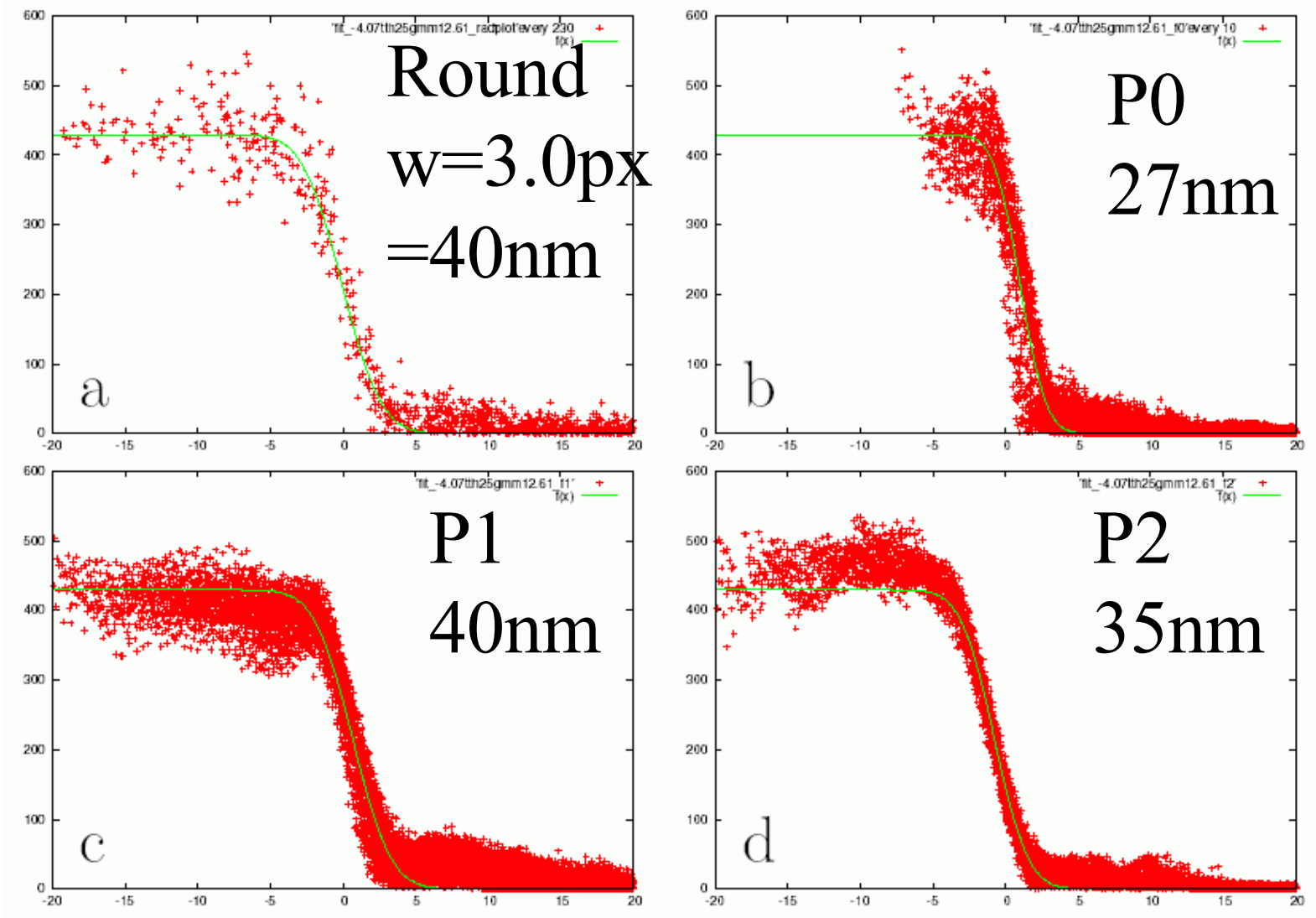
Facets of Equilibrium Crystal Shape



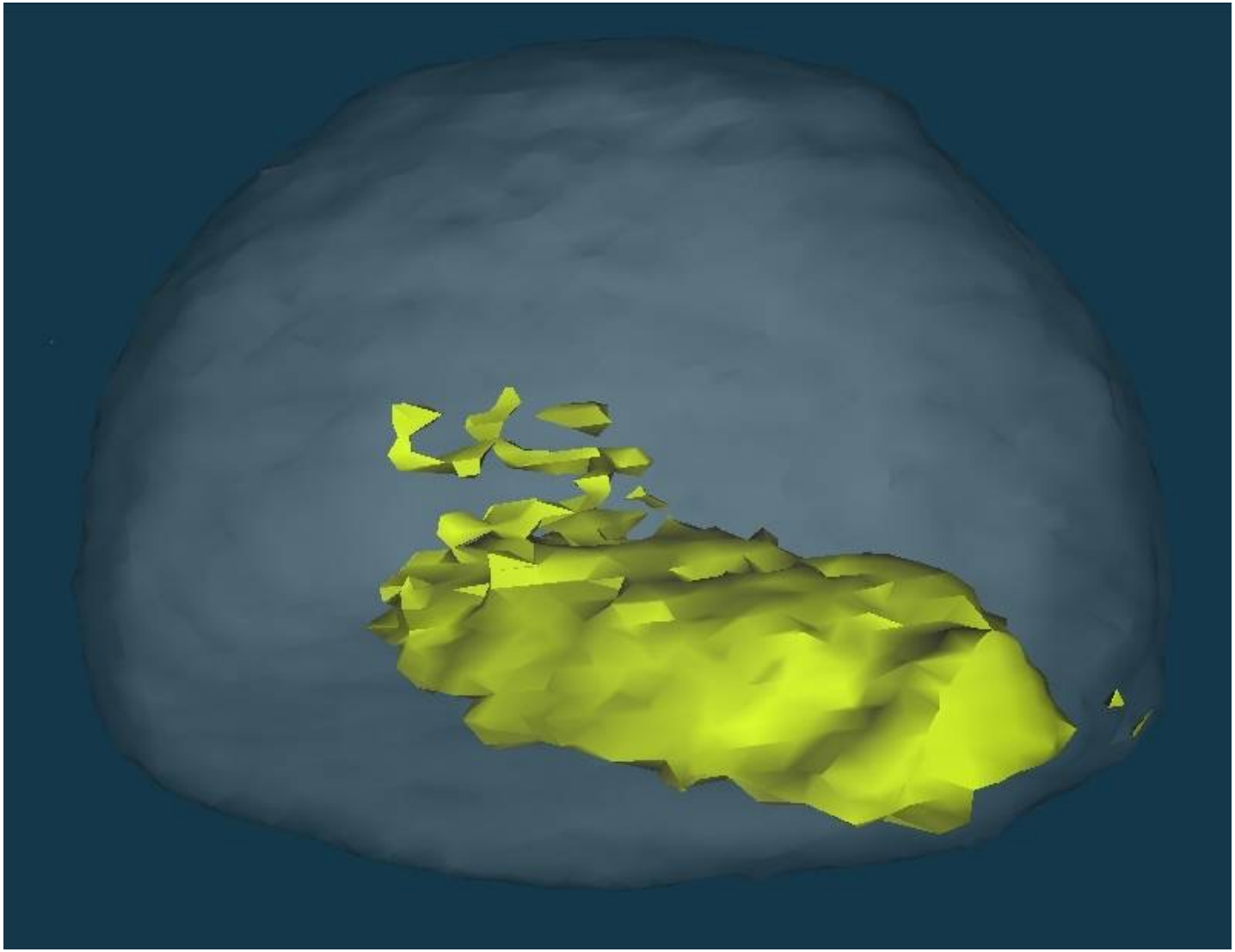


Thurmer K, Williams E, Reutt-Robey J
Science **297** 2033 (2002)

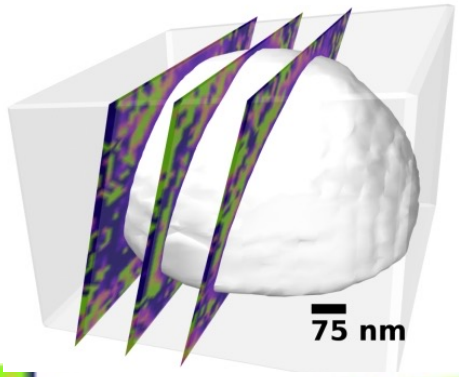
Density distribution across surface

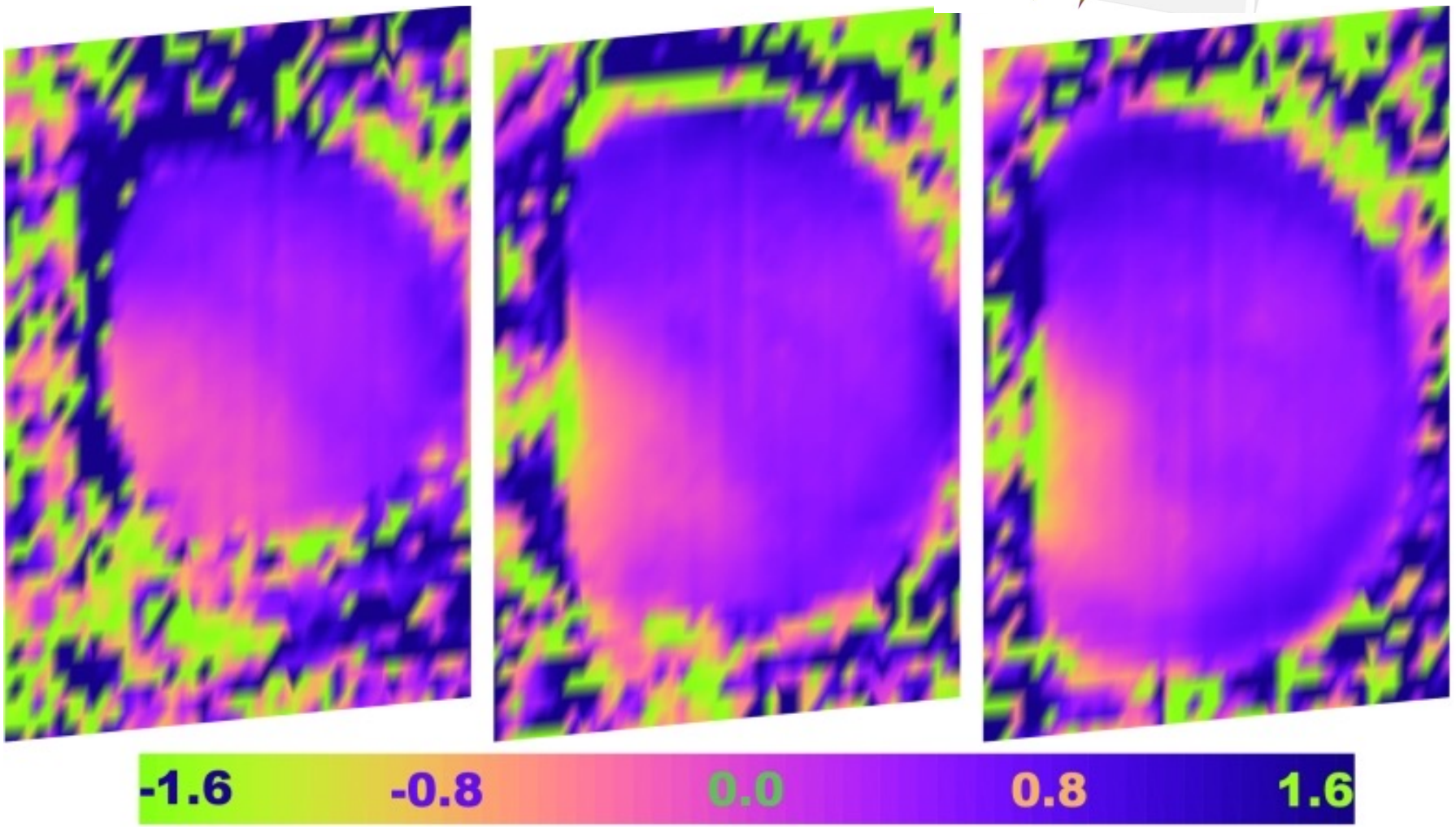


Modeling of 3D Phase Bump

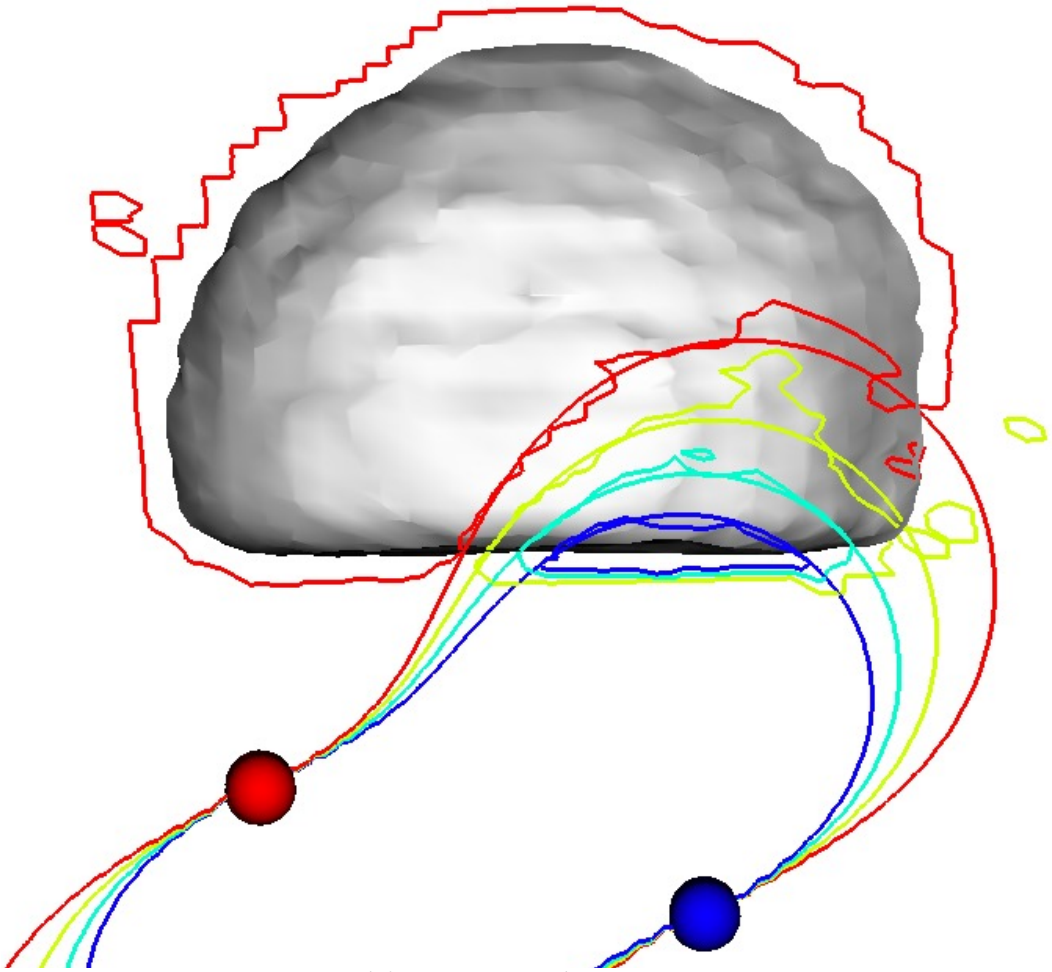


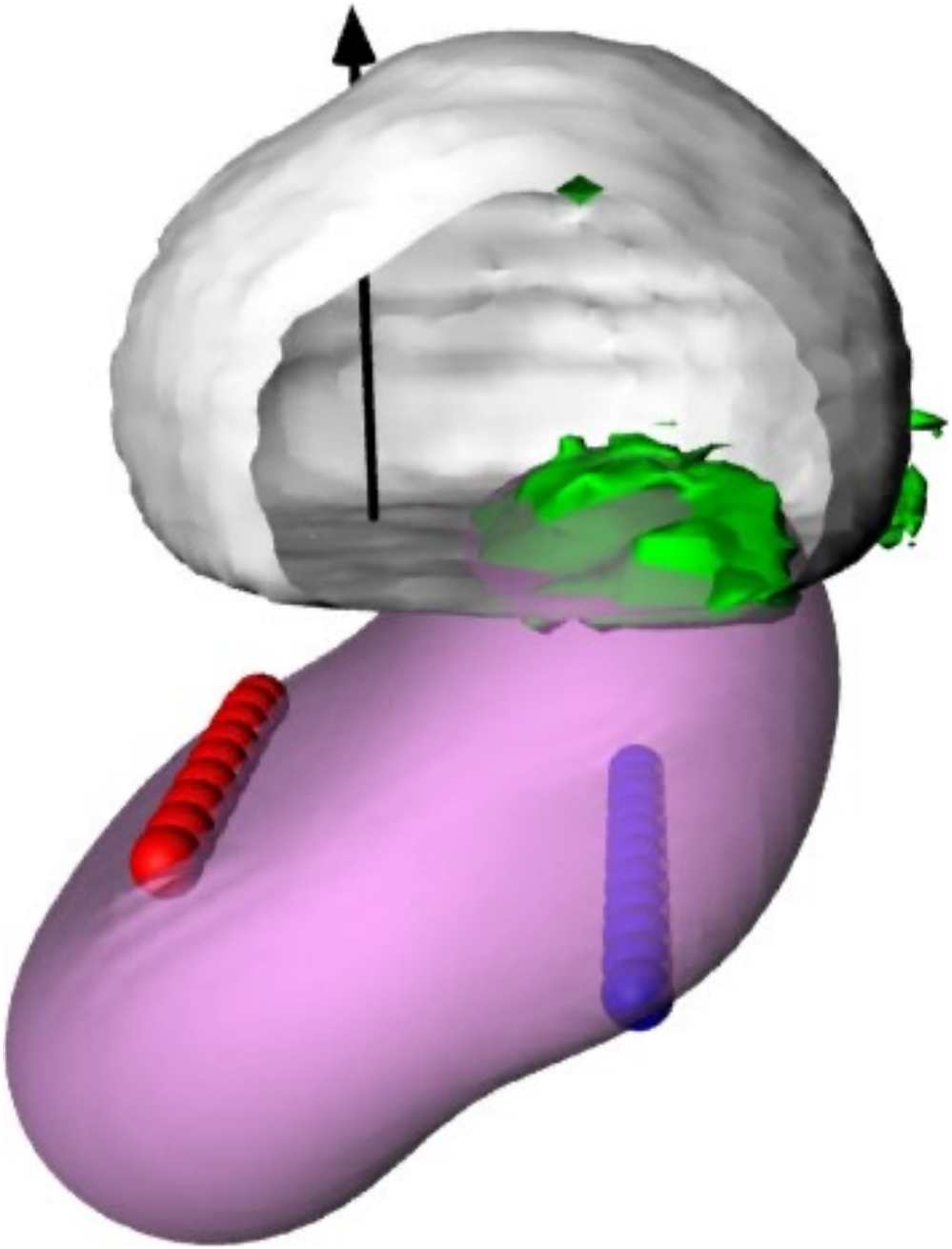
3D phase map sections





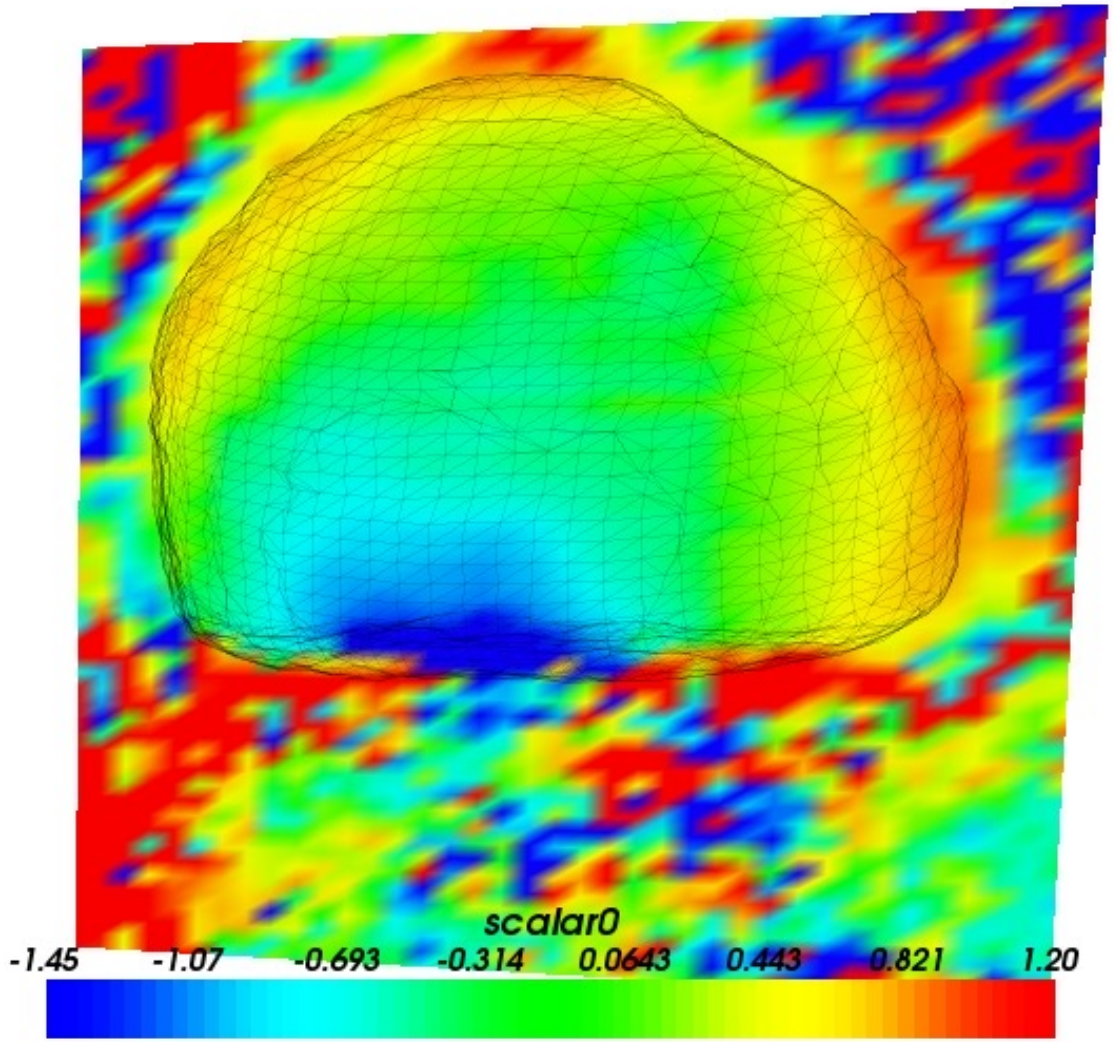
Field lines of Point Charges



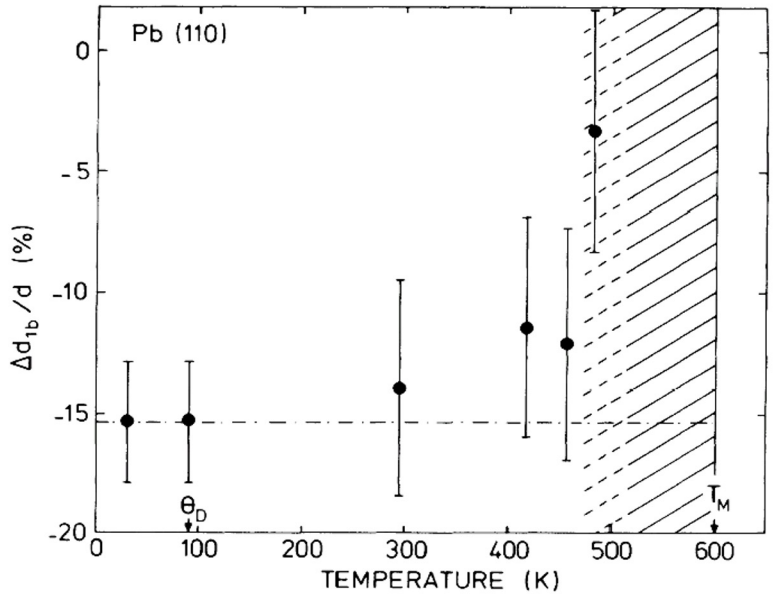


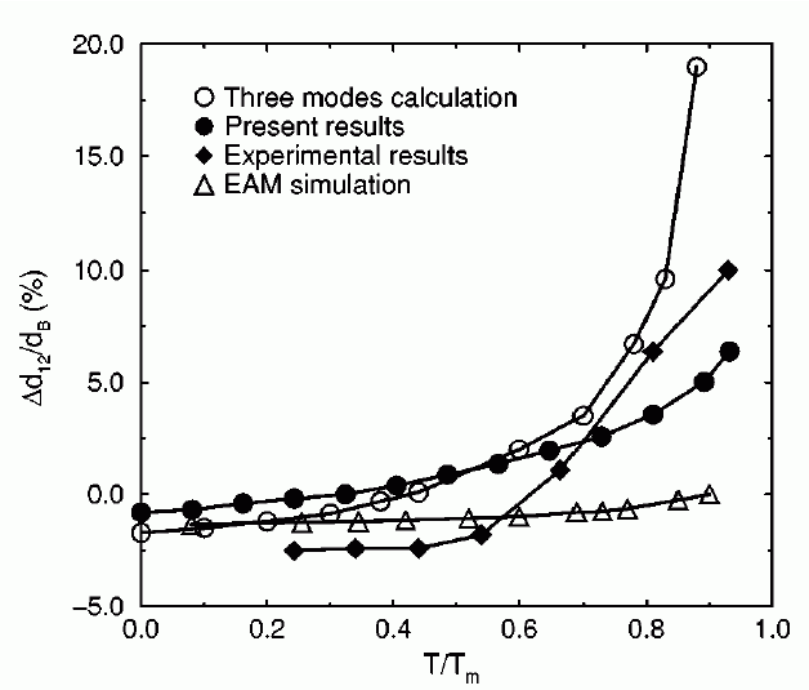
Contours showing Positive Phase

including correction for refraction by crystal



“Anomalous” Thermal Expansion of surfaces



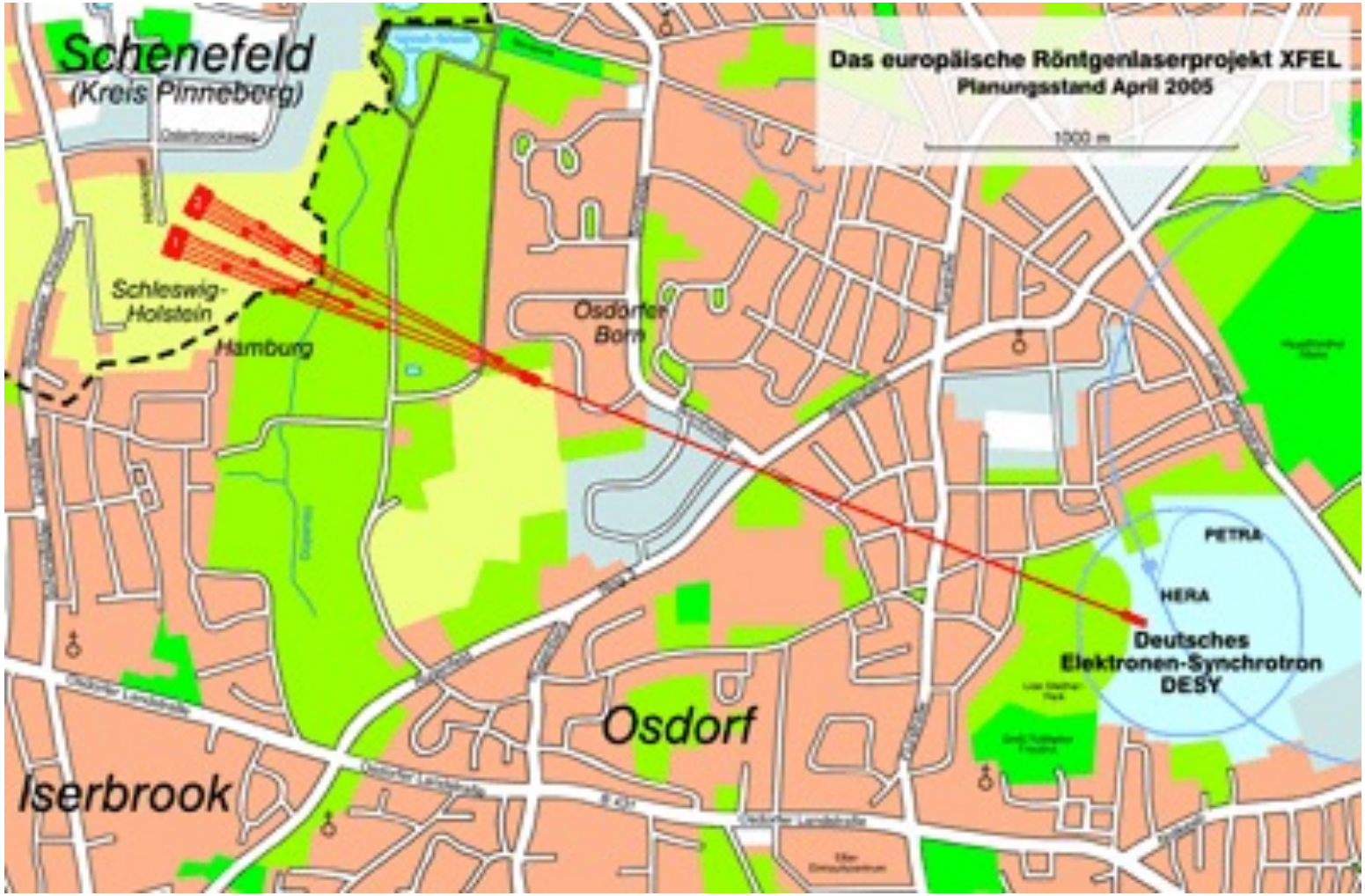


Pb(110) experiment
J. Frenken, F. Huussen and
J.F.van der Veen, PRL 58 401 (1987)

Ag(111) theory
J. Xie (Scheffler group)
PRB 59 970 (1999)

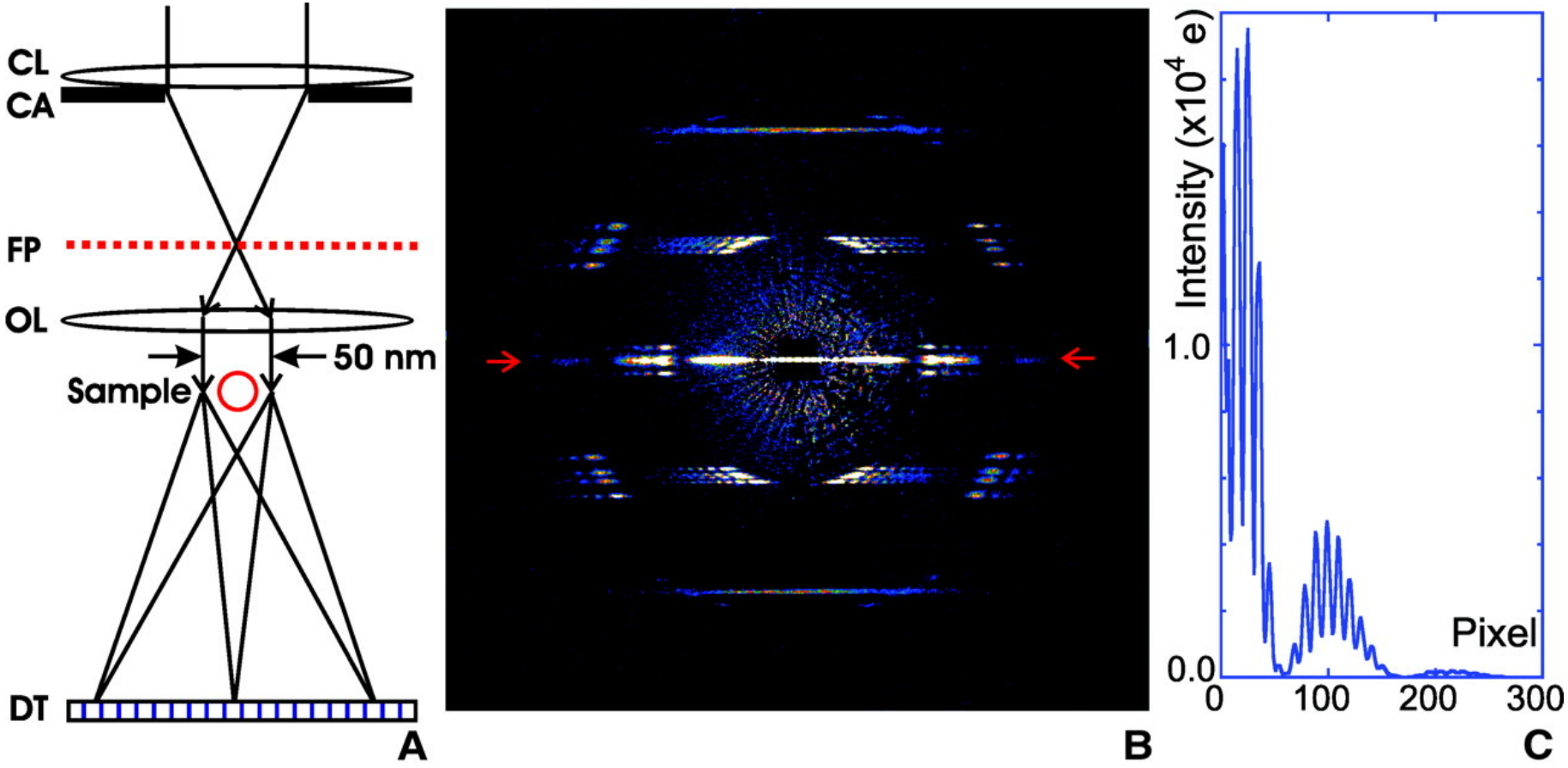
Future Euro-XFEL in Hamburg

should be operating in 2014



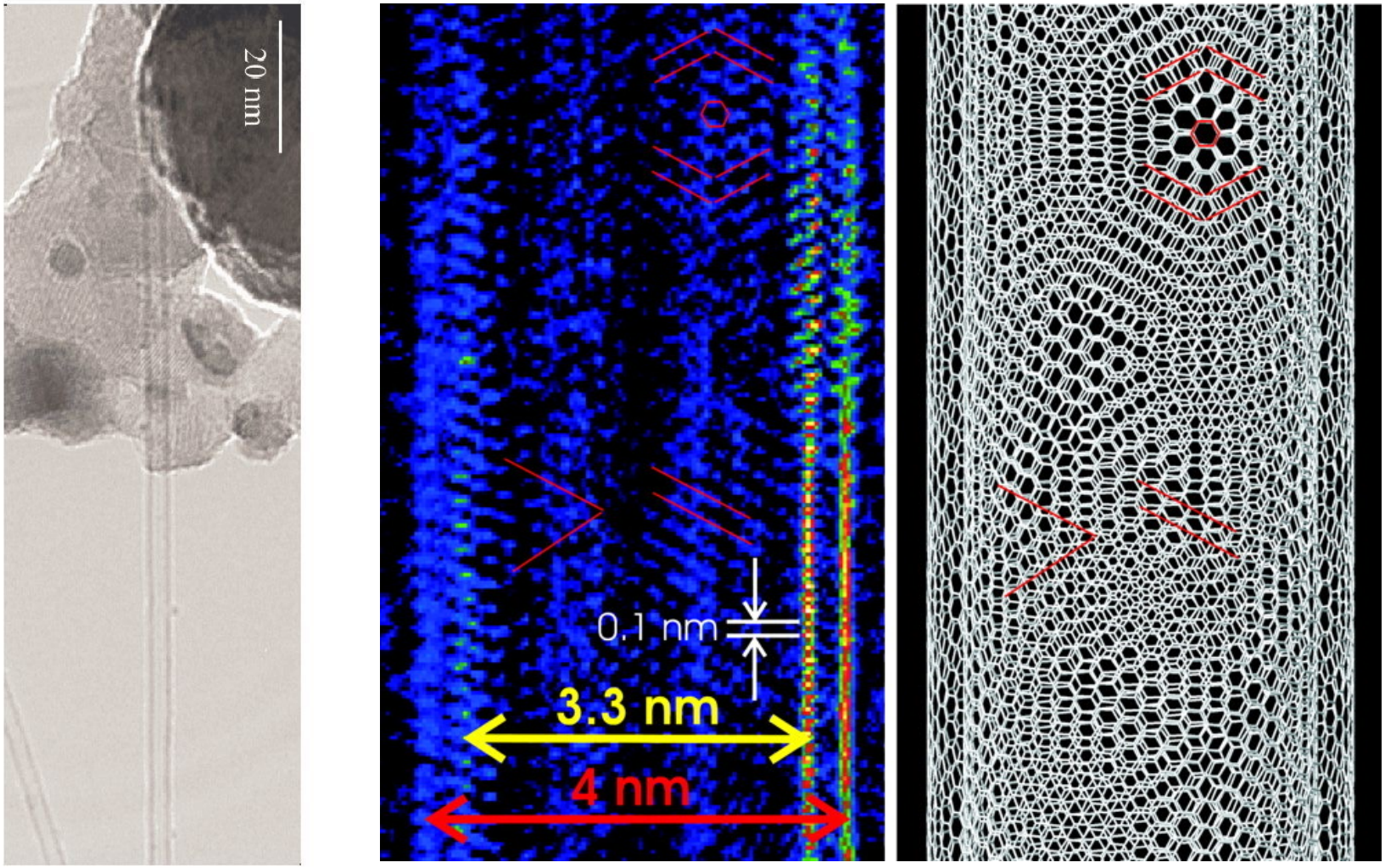
Electron Diffraction from C-nanotube

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

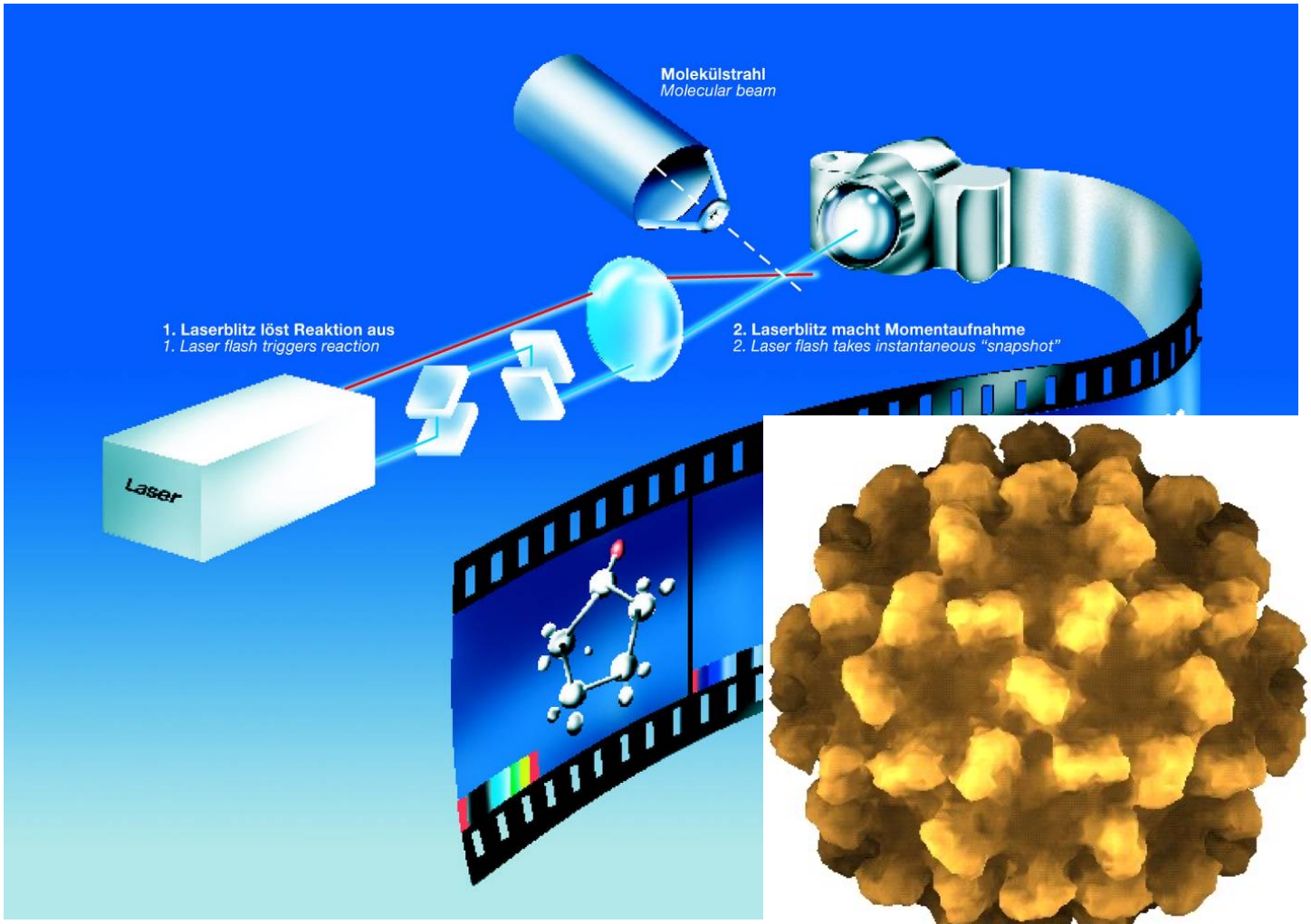


Images of DW C-nanotube

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



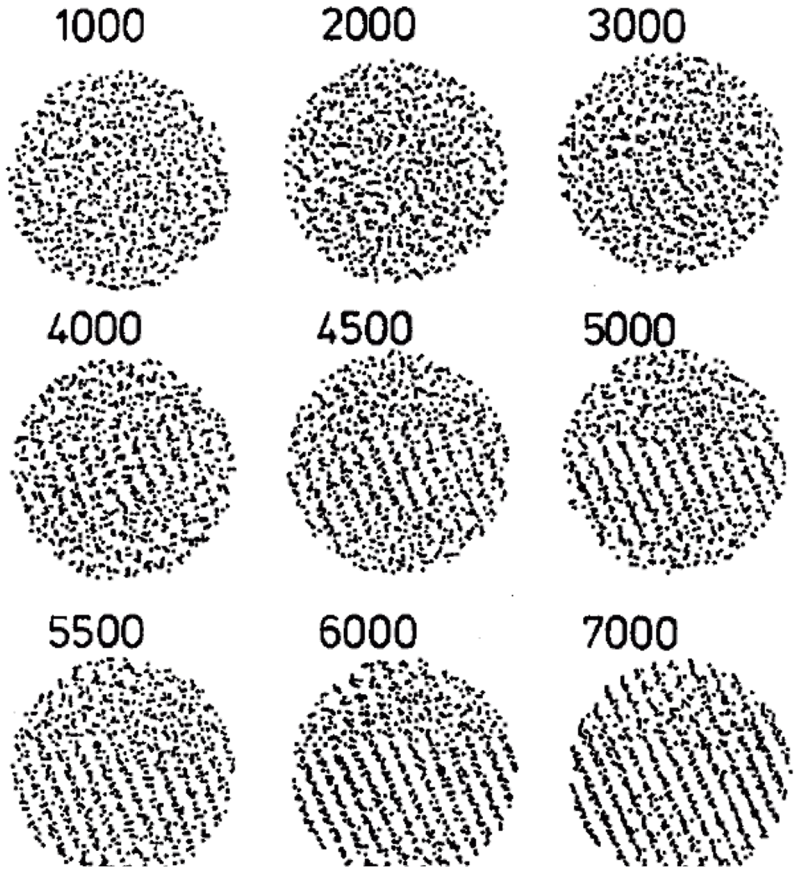
Molecular Movies using XFEL

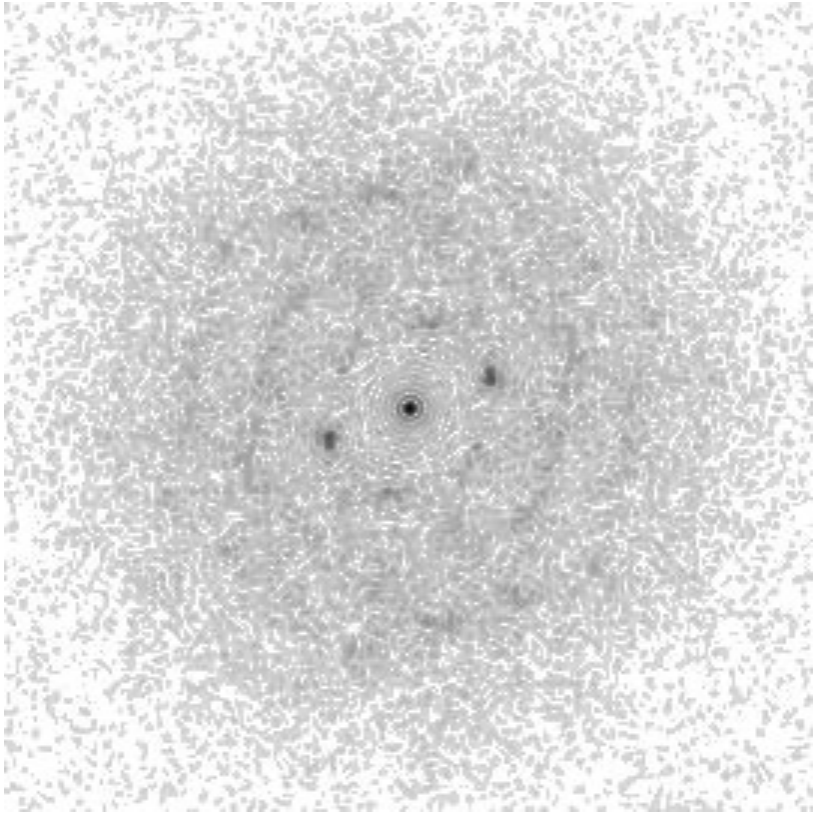


MD simulation of freezing

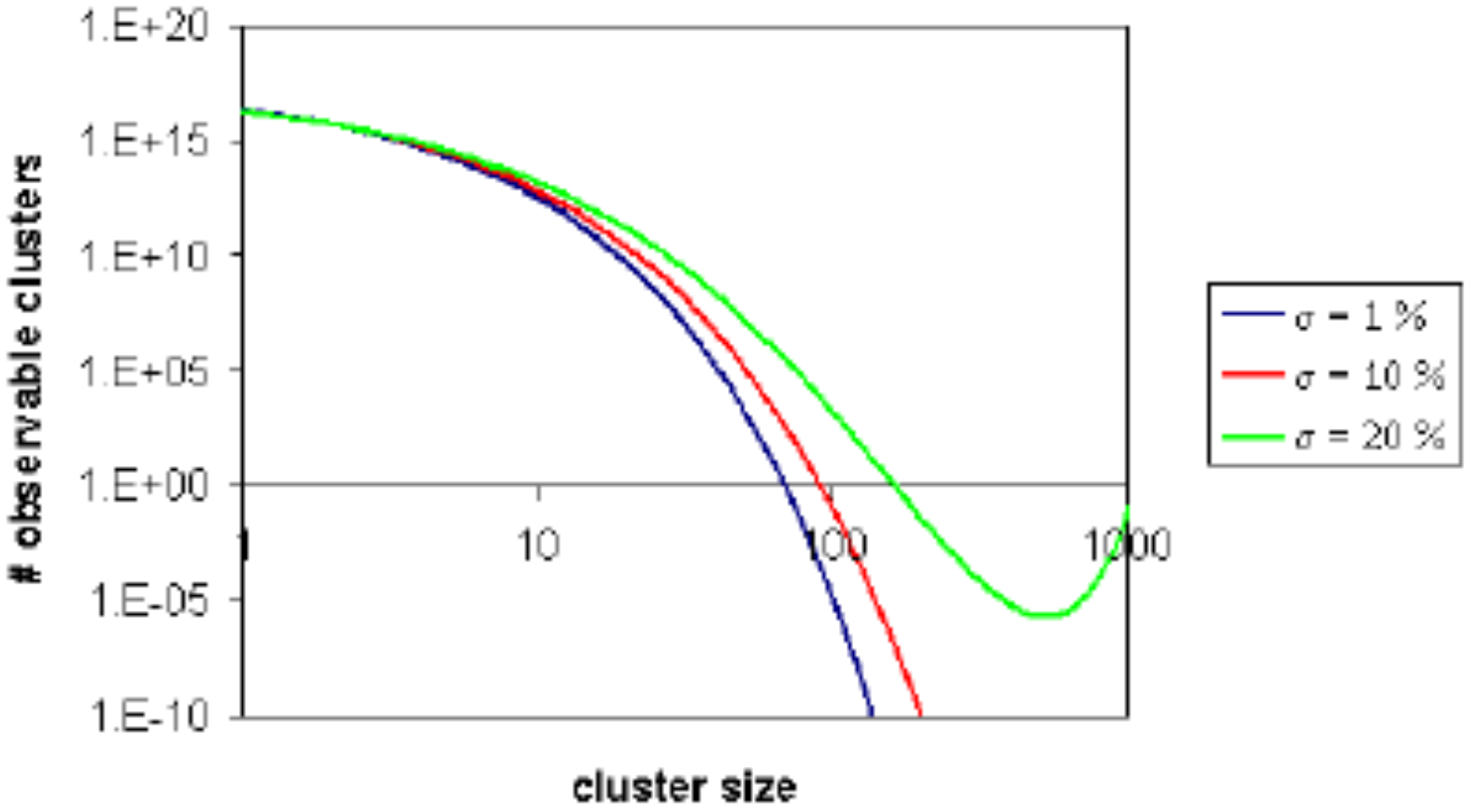
LJ liquid of 864 atoms. Time steps after T-Jump

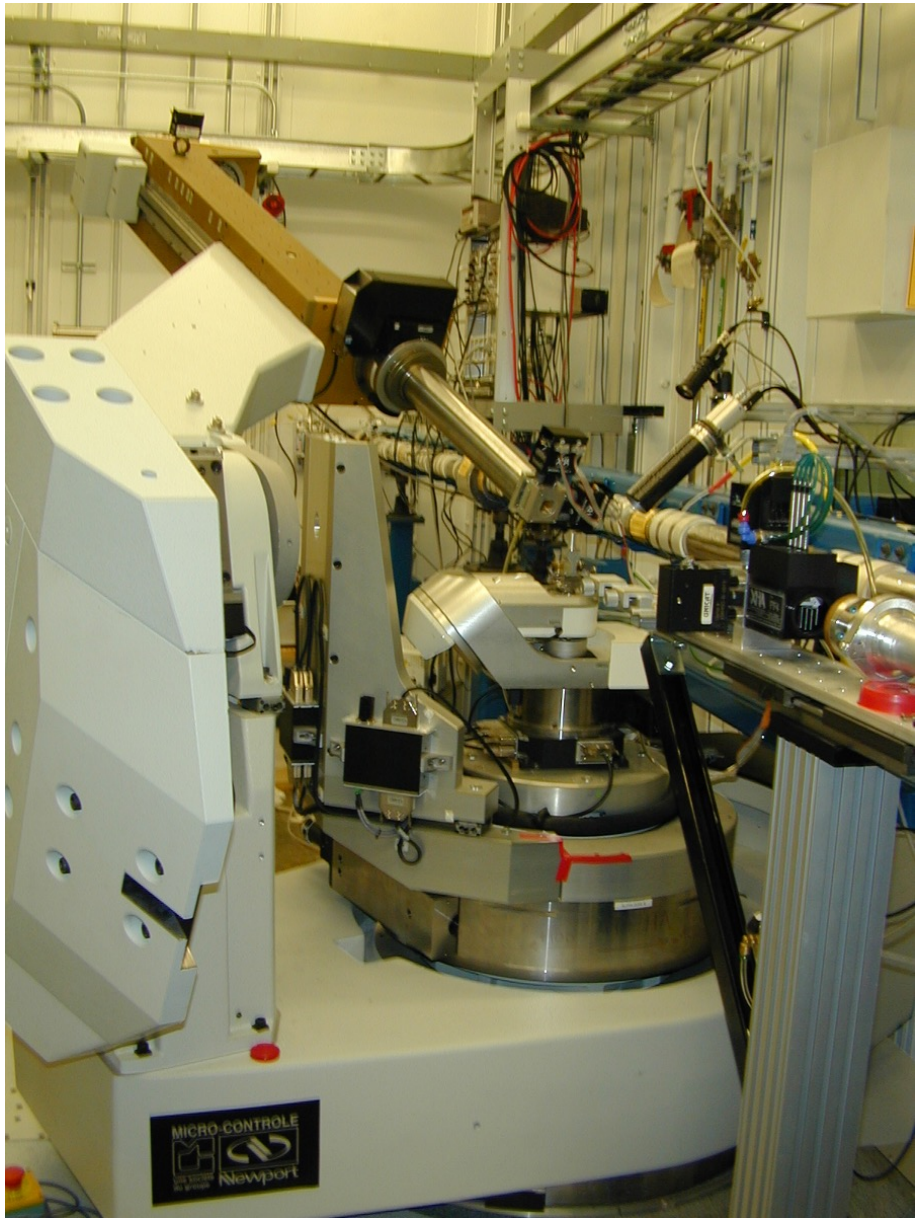
S. Nose and F. Yonezawa, JCP 84 1893 (1986)



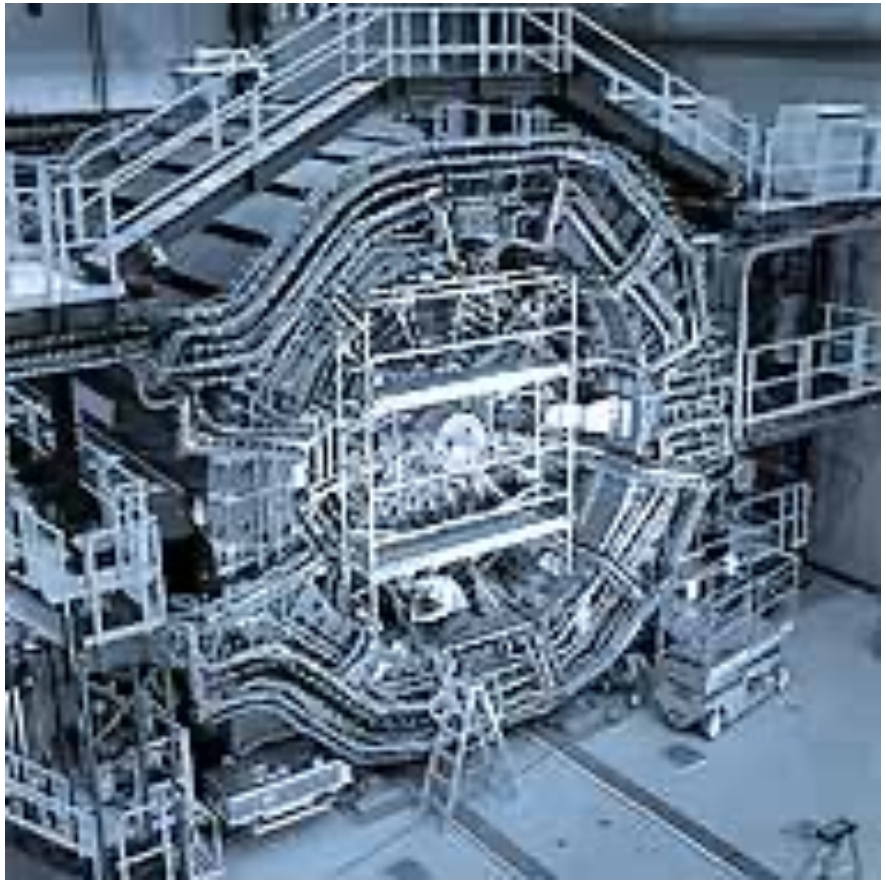


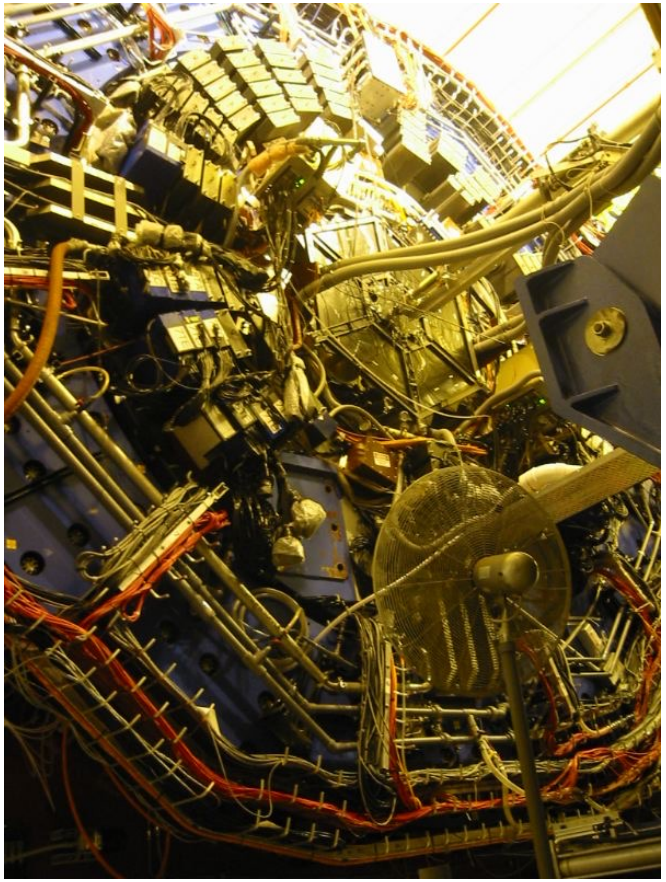
Nucleation cluster size distribution




MICRO-CONTROLE
Newport

The HEP approach: detectors drive the science





Conclusions

- Internal structure of Ag and Pb Nanocrystals
- 3D imaging practical for nanocrystals
- Phasing by computation instead of lens
- Strain fields imaged from asymmetric patterns
- Contact Forces cause strain inside crystal
- Surface strain has orientation dependence