

Inversion of Diffraction from Objects with Complex Density

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Phase Retrieval and
Coherent Scattering Workshop
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Outline

- Overview of Coherent Diffraction
- The **Phase** Problem
- Imaging of **Phase** Objects
- Strain fields within Nanocrystals
- Symmetry of Diffraction Images

Capabilities of Coherent Diffraction

- Thermodynamic fluctuations
 - remove ensemble averaging in CXD
- Probe of structure on **nm** scale
 - 1D, 2D and 3D
 - non-periodic object gives **continuous** $F(\mathbf{q})$
- **Oversampling** (in reciprocal space) permits solution of the **phase** problem

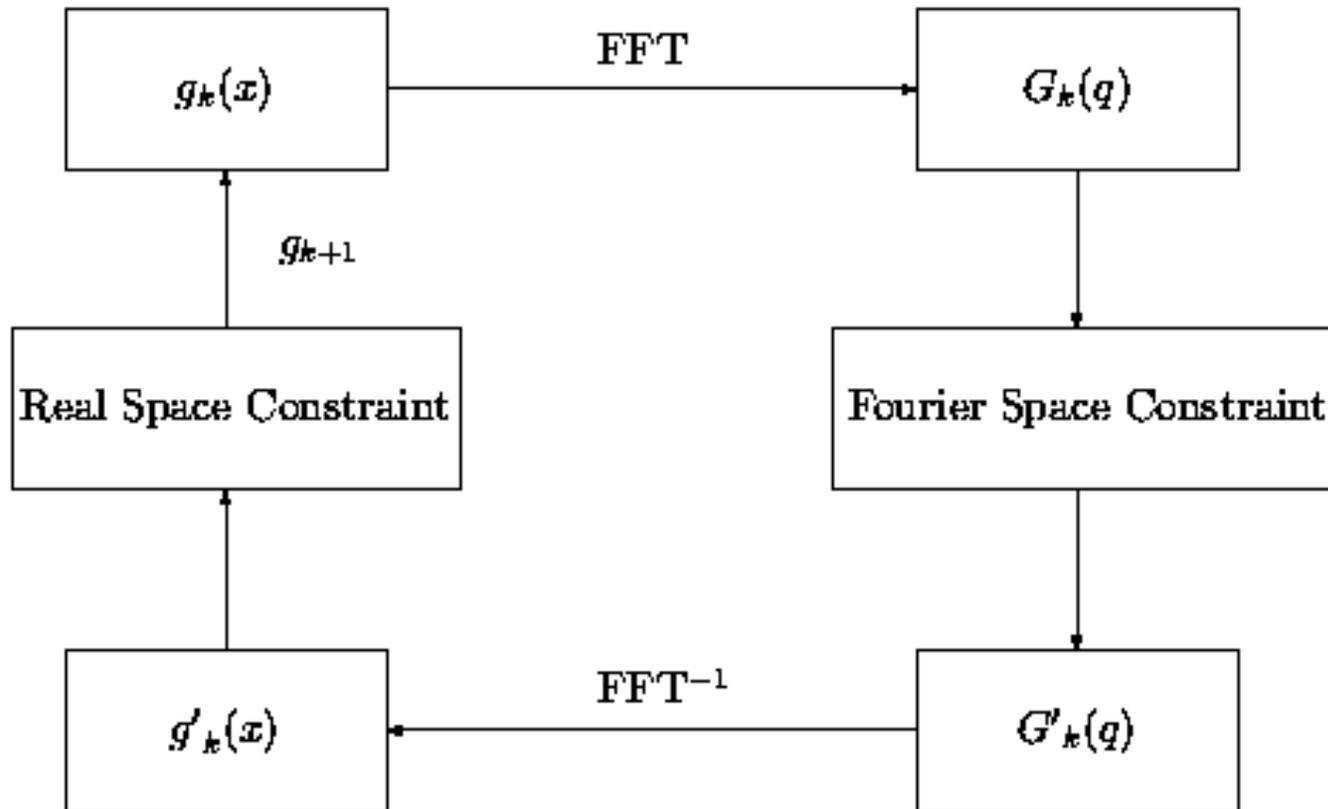
“Methods” Issues for CXD

- Control of Support (**Stadler, Zuo**)
 - remote *vs* integral aperture?
- Reference wave (**Eisebitt, Shen, Nugent**)
 - use of spherical wave illumination?
- Algorithm (**Chapman, Jacobsen, Zuo**)
- Detectors (**Nishino, Chapman**)
- Radiation Damage (**Chapman, Jacobsen, Zuo**)
 - electrons *vs* X-rays, imaging *vs* diffraction?

“Opportunities” for CXD

- Real-time data analysis (**Nishino**)
- Sample holders (**Spence**)
 - how to beat Brownian motion?
- Registration (**Vartanians, Spence**)
- Complimentarity (**Chesnel, Eisebitt**)
 - use of other microscopy methods
- Resonance (**Chesnel, Vartanians, Eisebitt**)
- Phase contrast (**Stadler, Robinson**)

Generic “Error Reduction” method



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

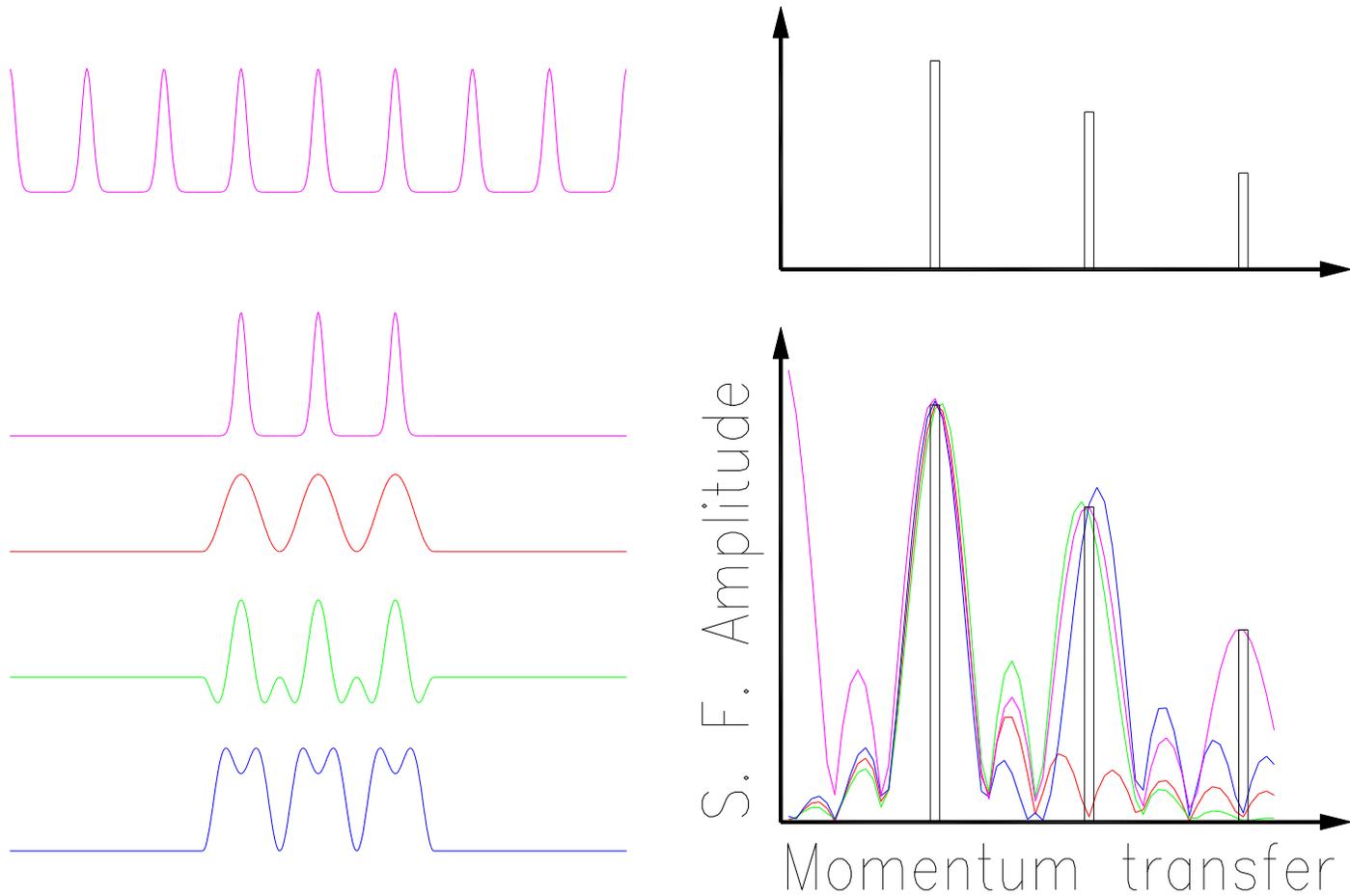
R. W. Gerchberg and W. O. Saxton *Optik* 35 237 (1972)

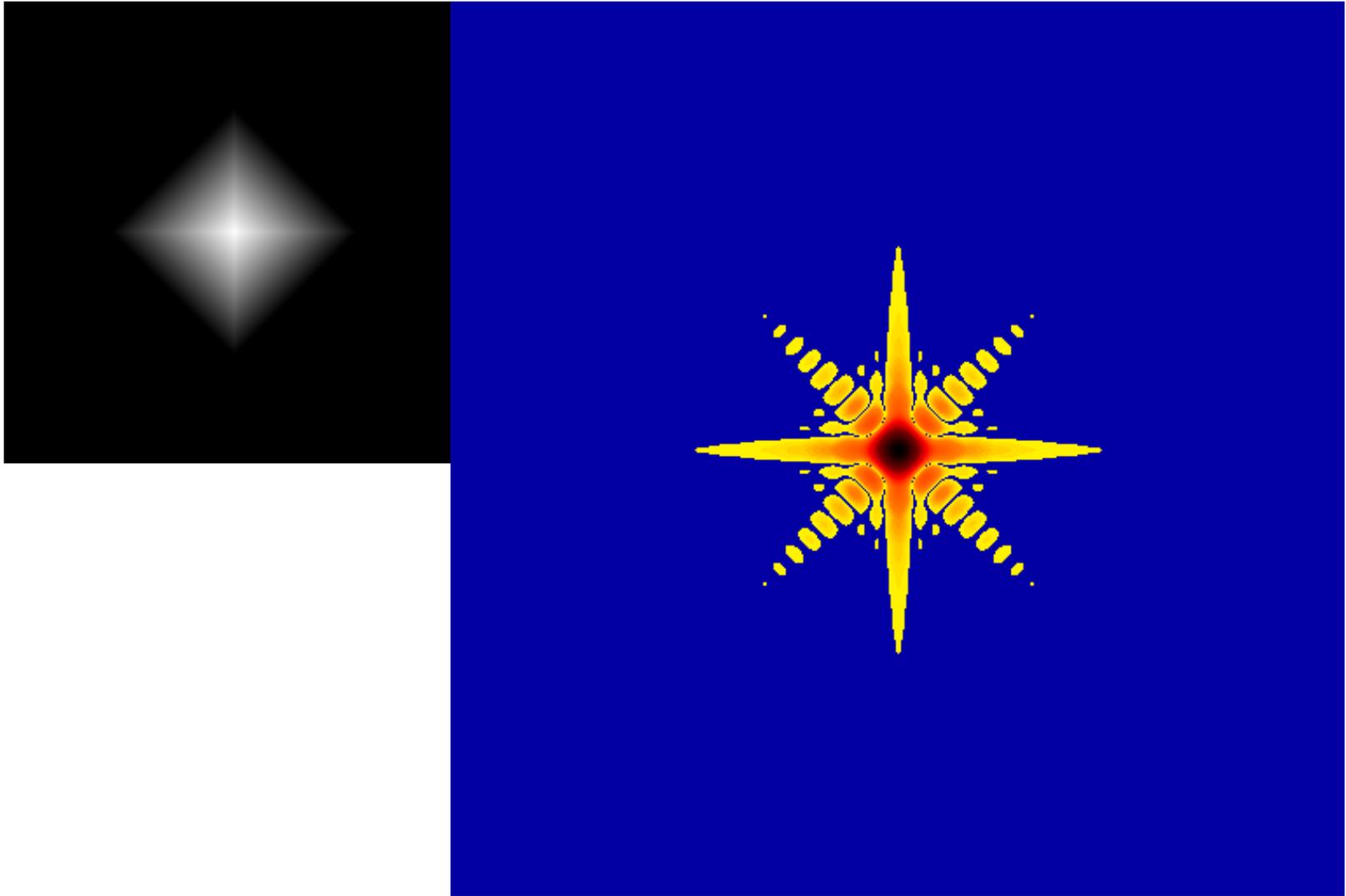
Real-space Constraints in Crystallography

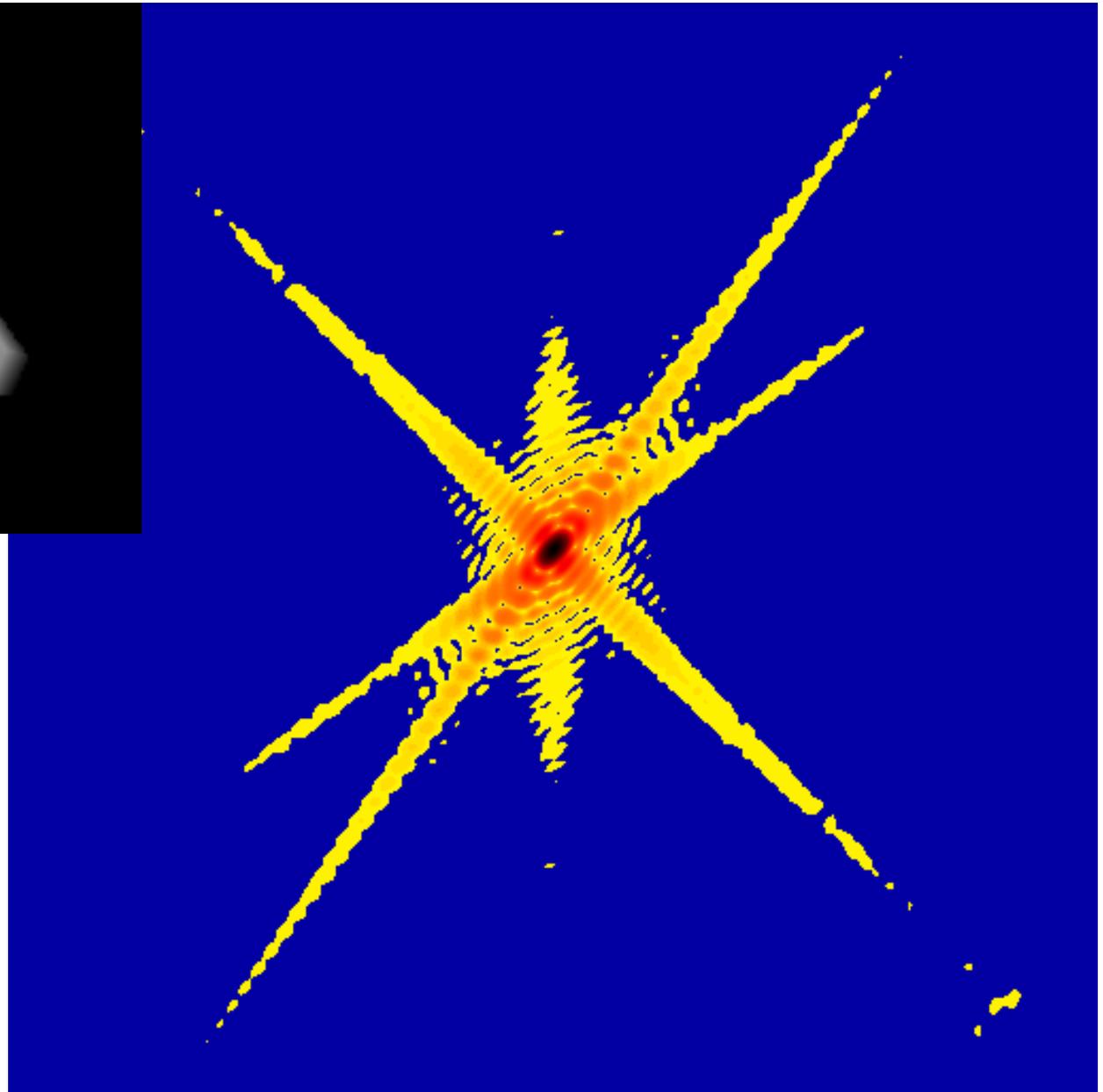
R. P. Millane, J. Opt. Soc Am. A **13** 725 (1996)

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

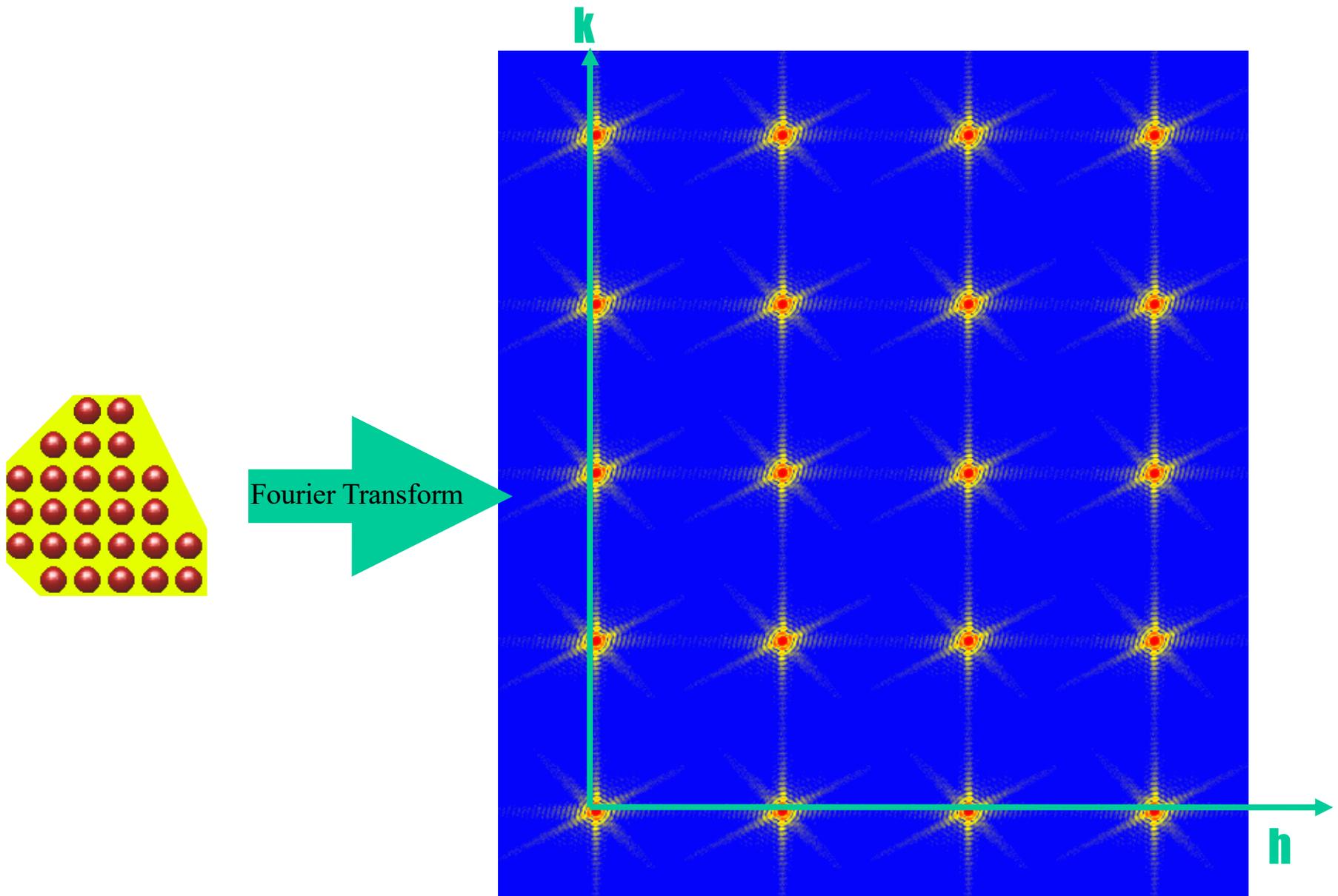
Phase Problem: Finite-size Effect







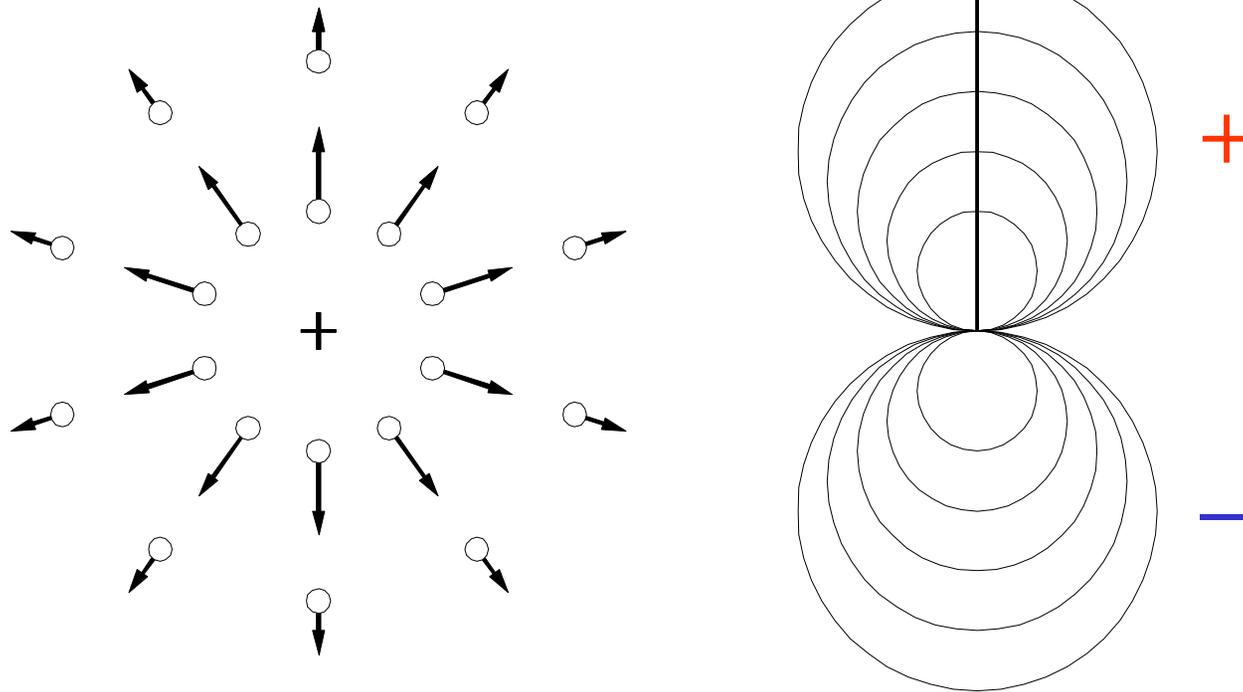
Coherent Diffraction from Crystals



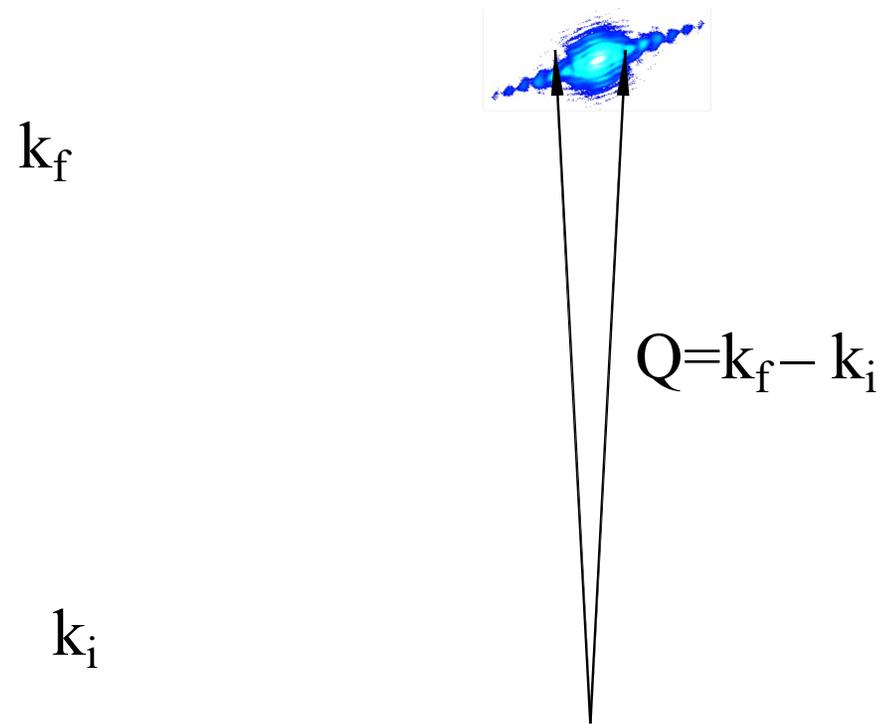
Diffraction by Strain of Point Defect

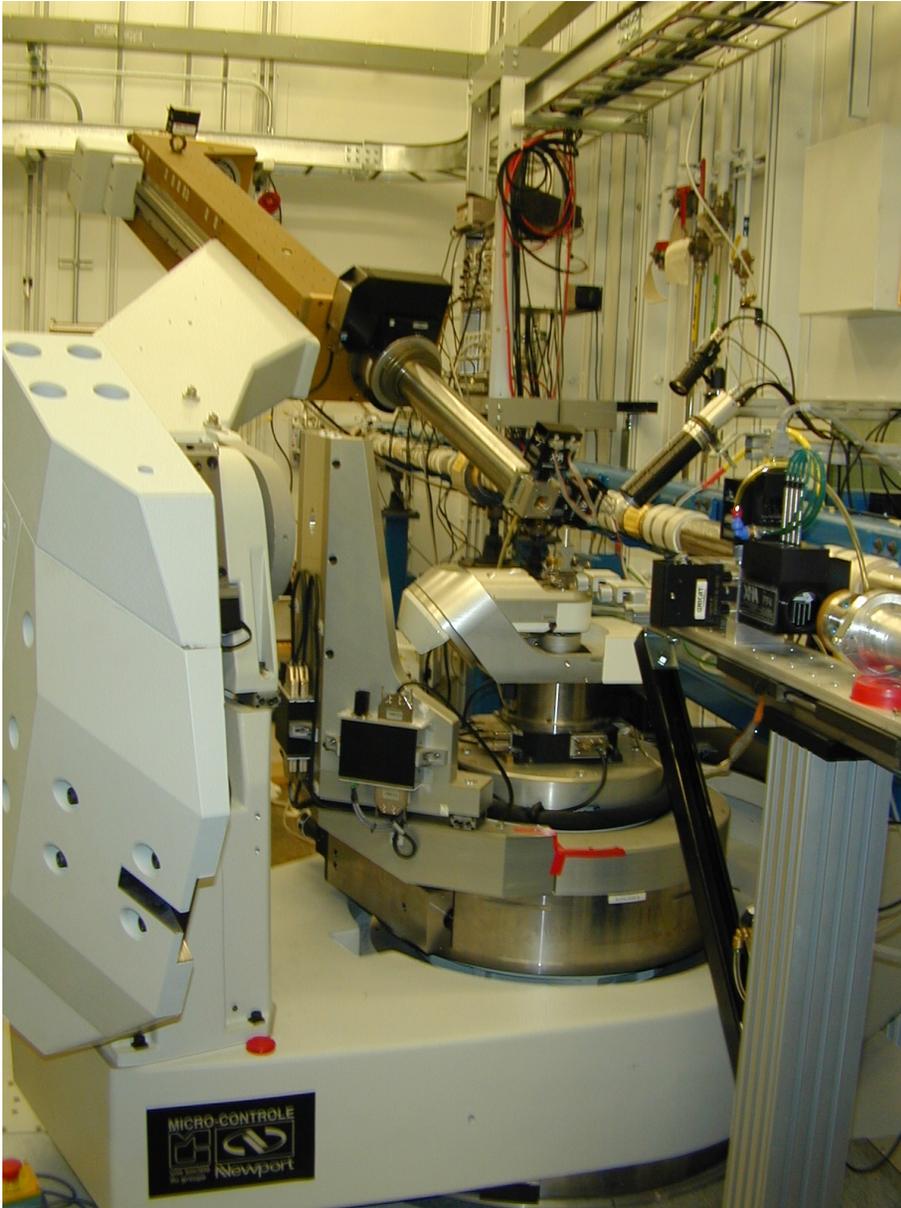
$$A \sim \sum e^{i\mathbf{Q}\cdot(\mathbf{R}_j+\mathbf{u}_j)}$$
$$\approx \sum e^{i\mathbf{Q}\cdot\mathbf{R}_j} (1+i\mathbf{Q}\cdot\mathbf{u}_j)$$

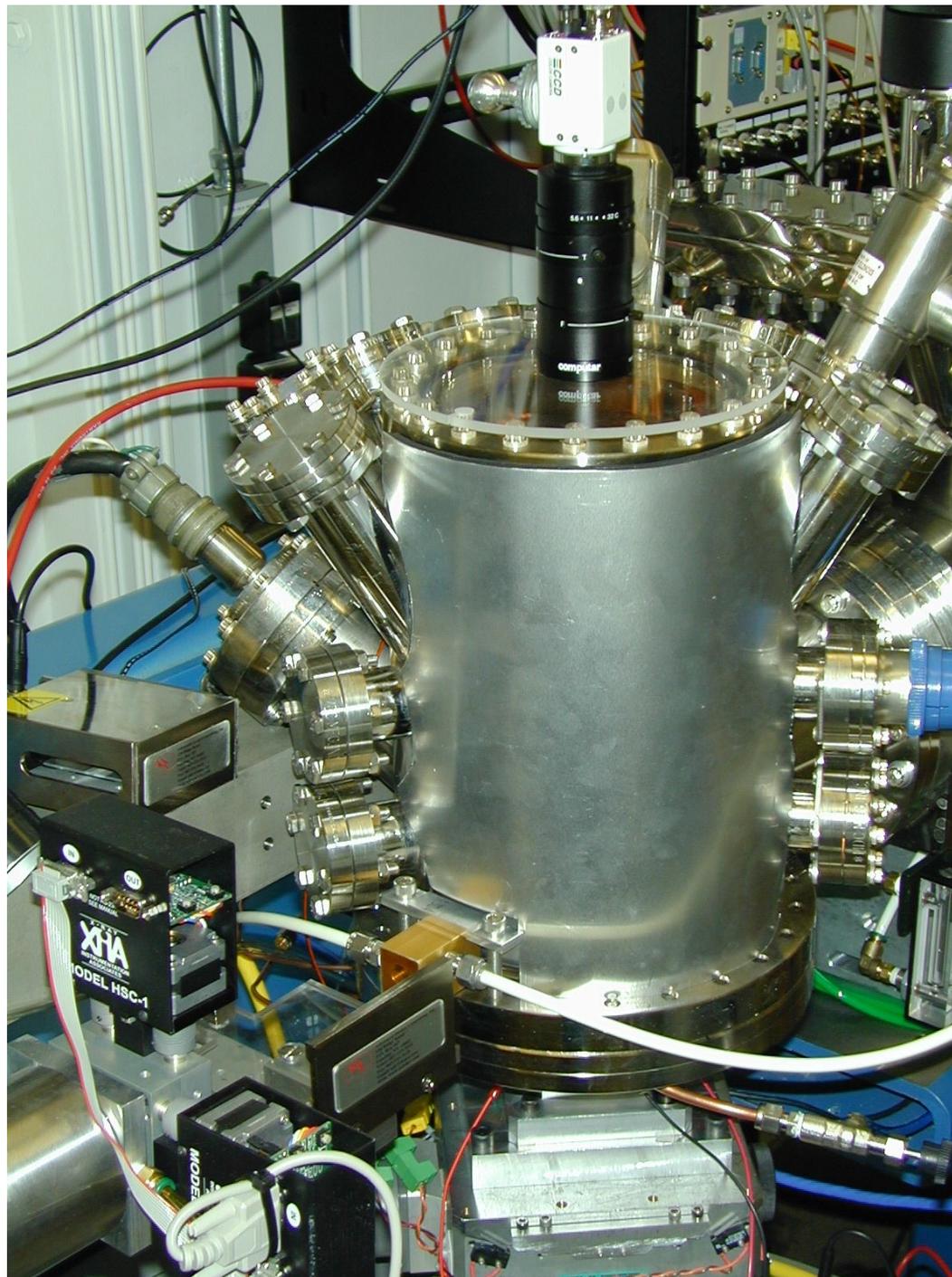
Imaginary density



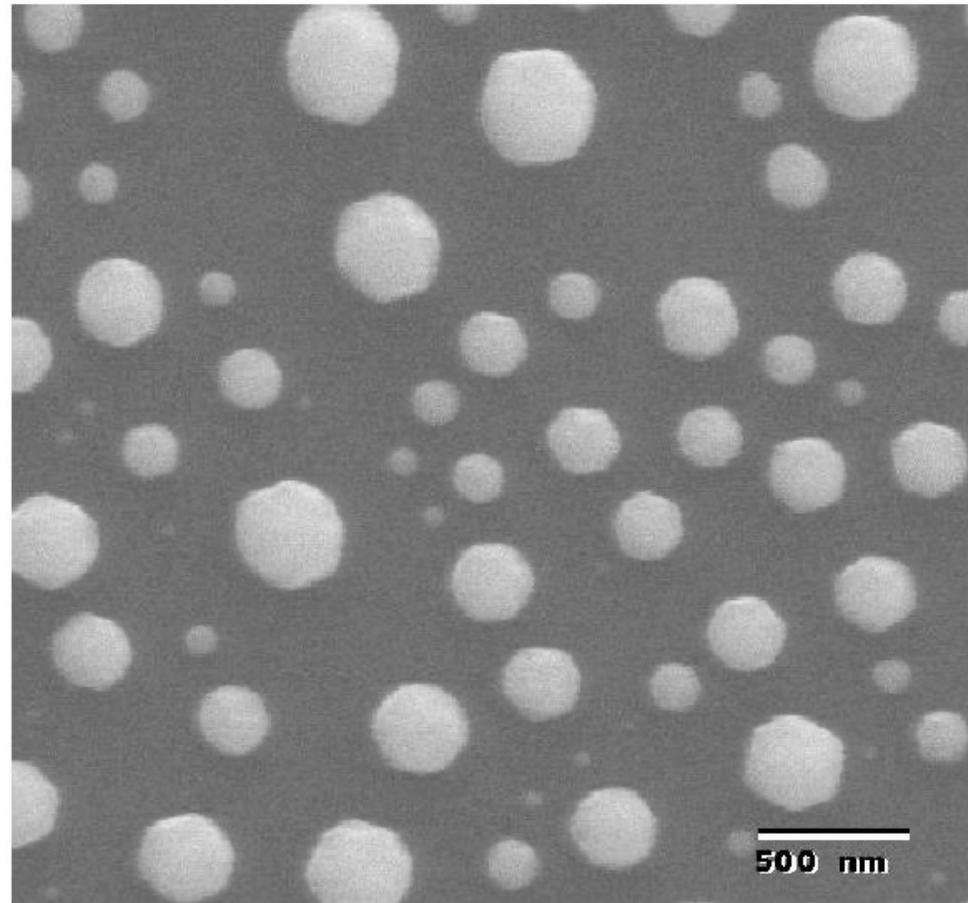
3D Diffraction Method







In situ growth of Pb crystals



Good statistics, 3D diffraction data

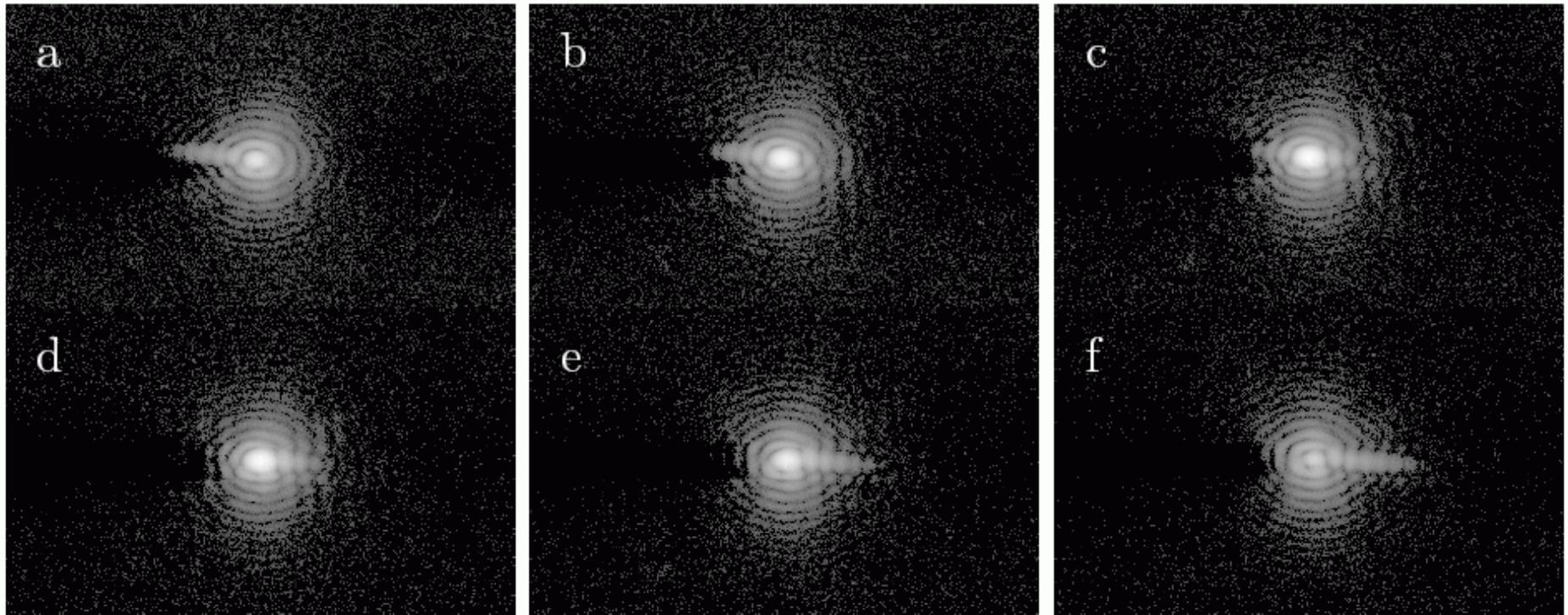
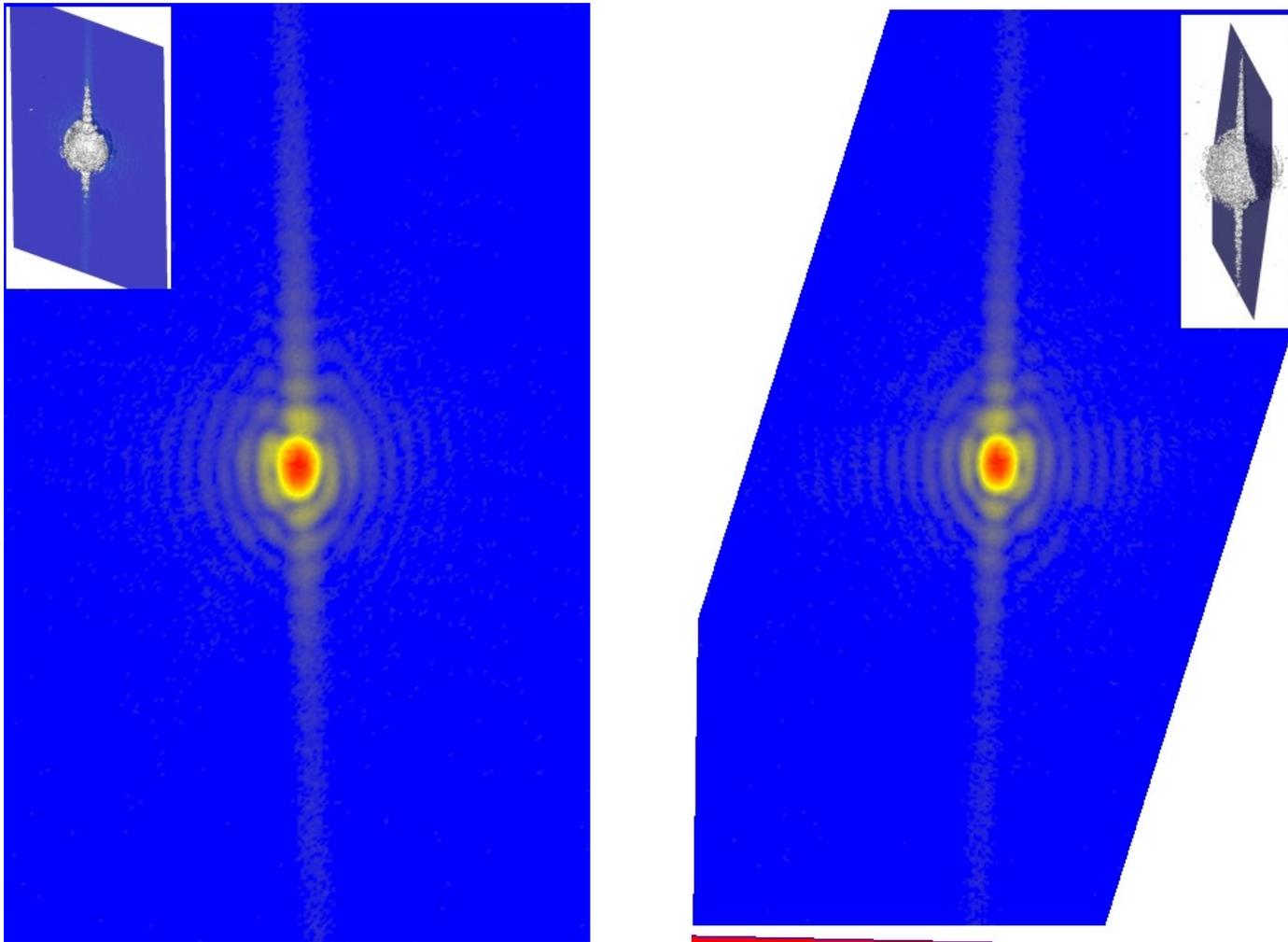
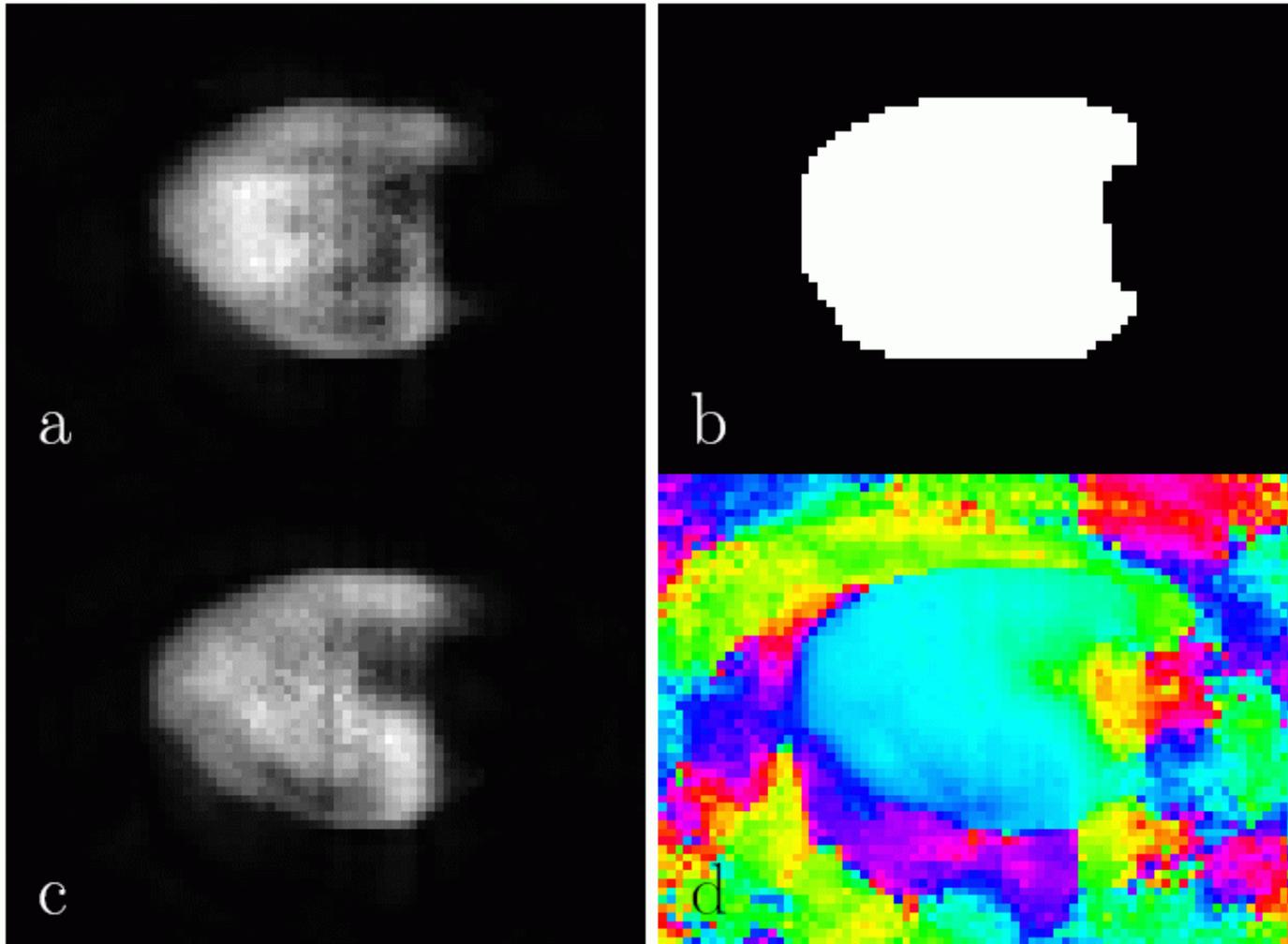


Figure 4.12: Center slices from 3D CXD pattern from Pb sample, on a log scale. Data file 296 from 10/03.

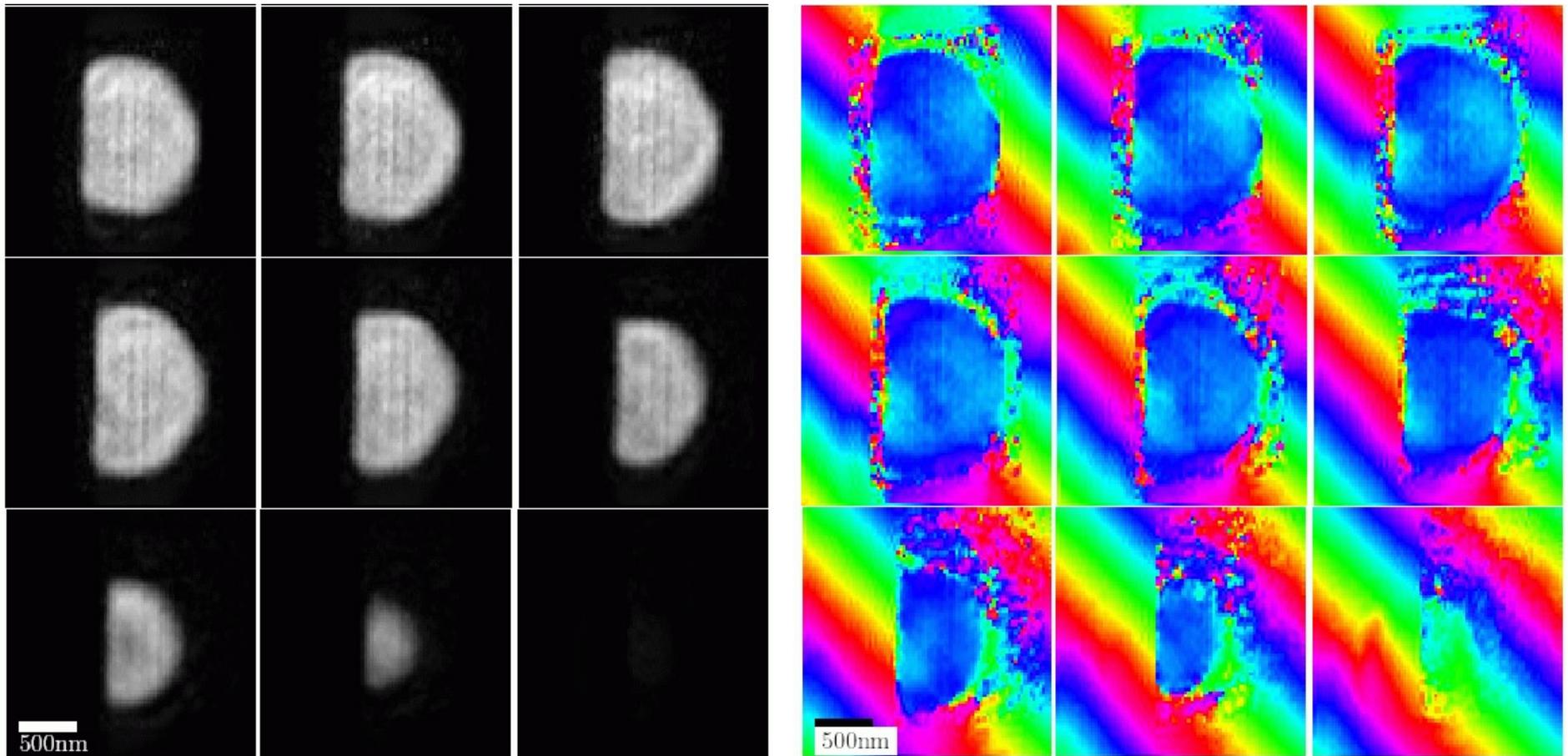
3D data along special directions



Learn shape of “tight” support

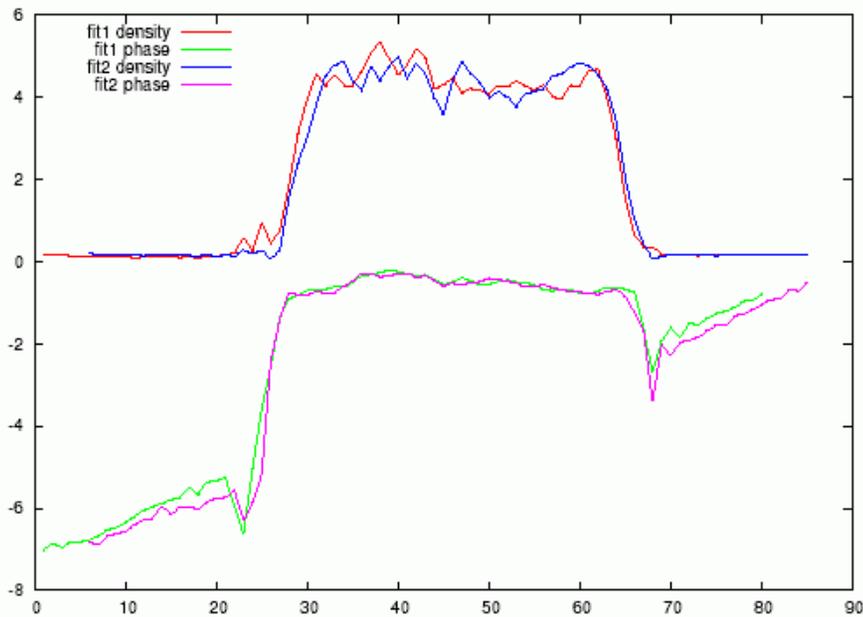


Then refine amplitude *and* phase

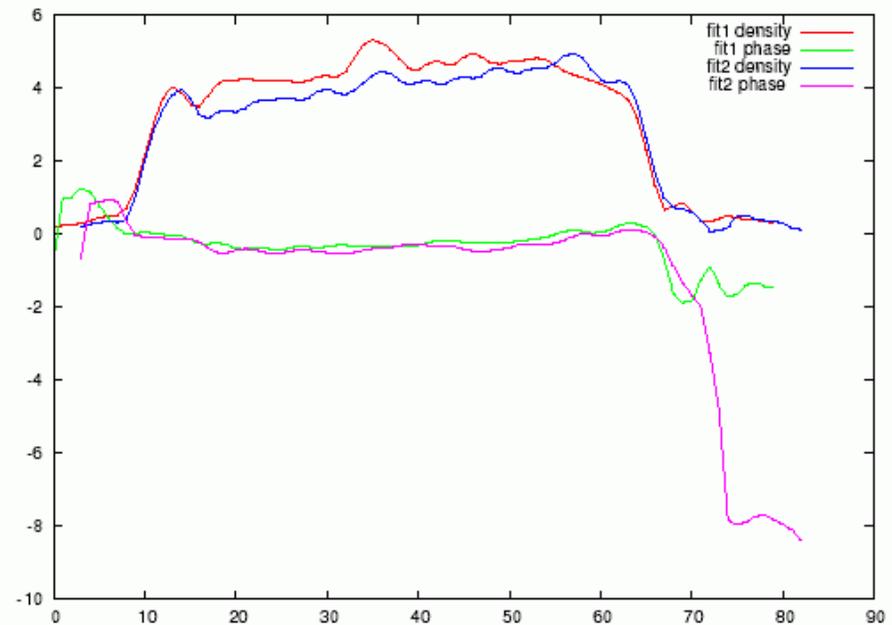


Amplitude/phase cross sections

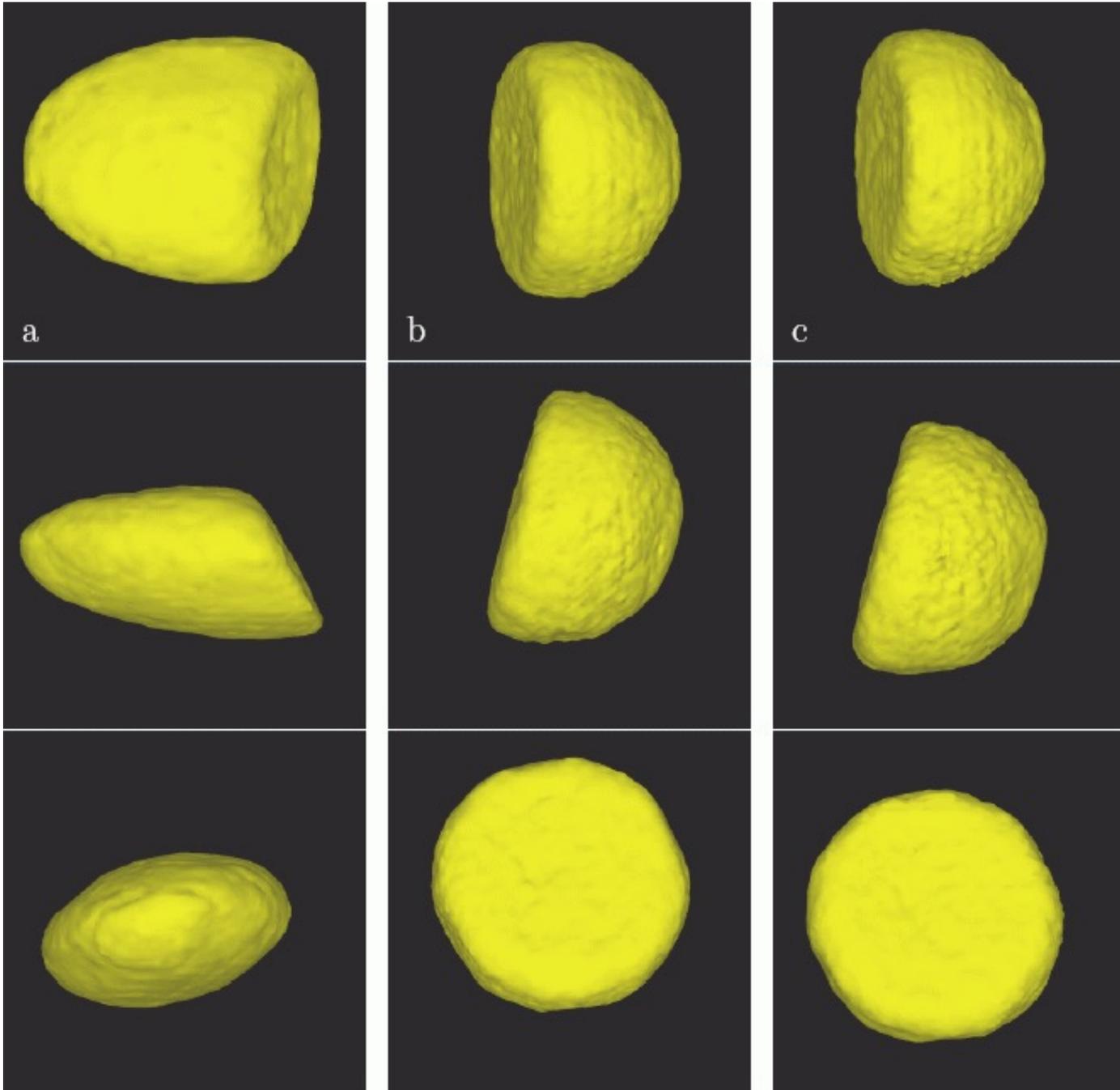
x-striations do not reproduce



x-coordinate

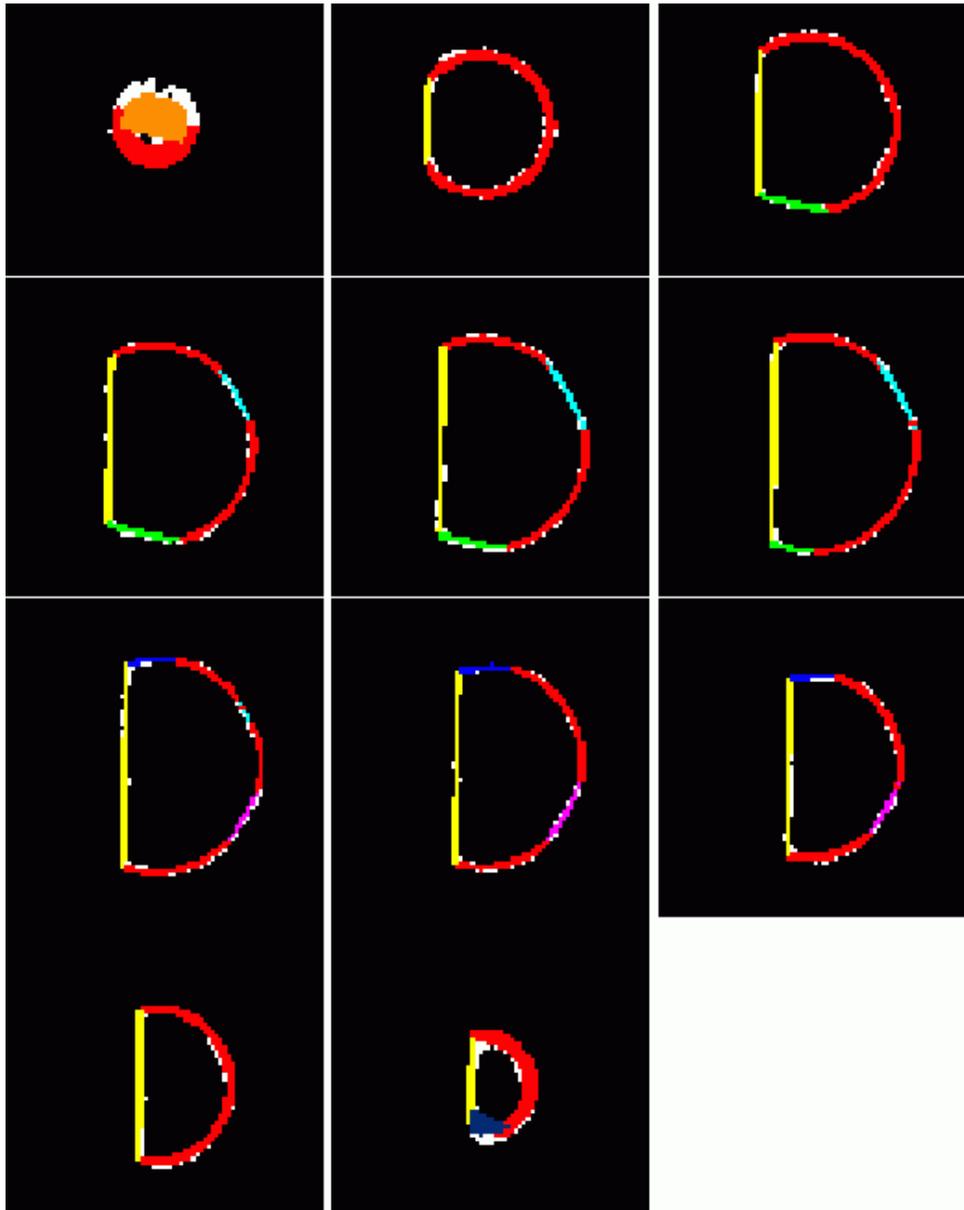


y-coordinate



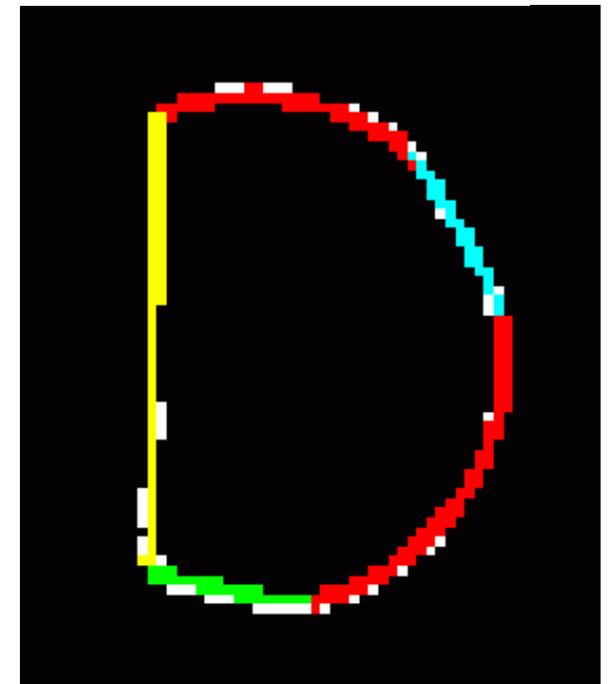
I. K. Robinson, Porquerolles

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

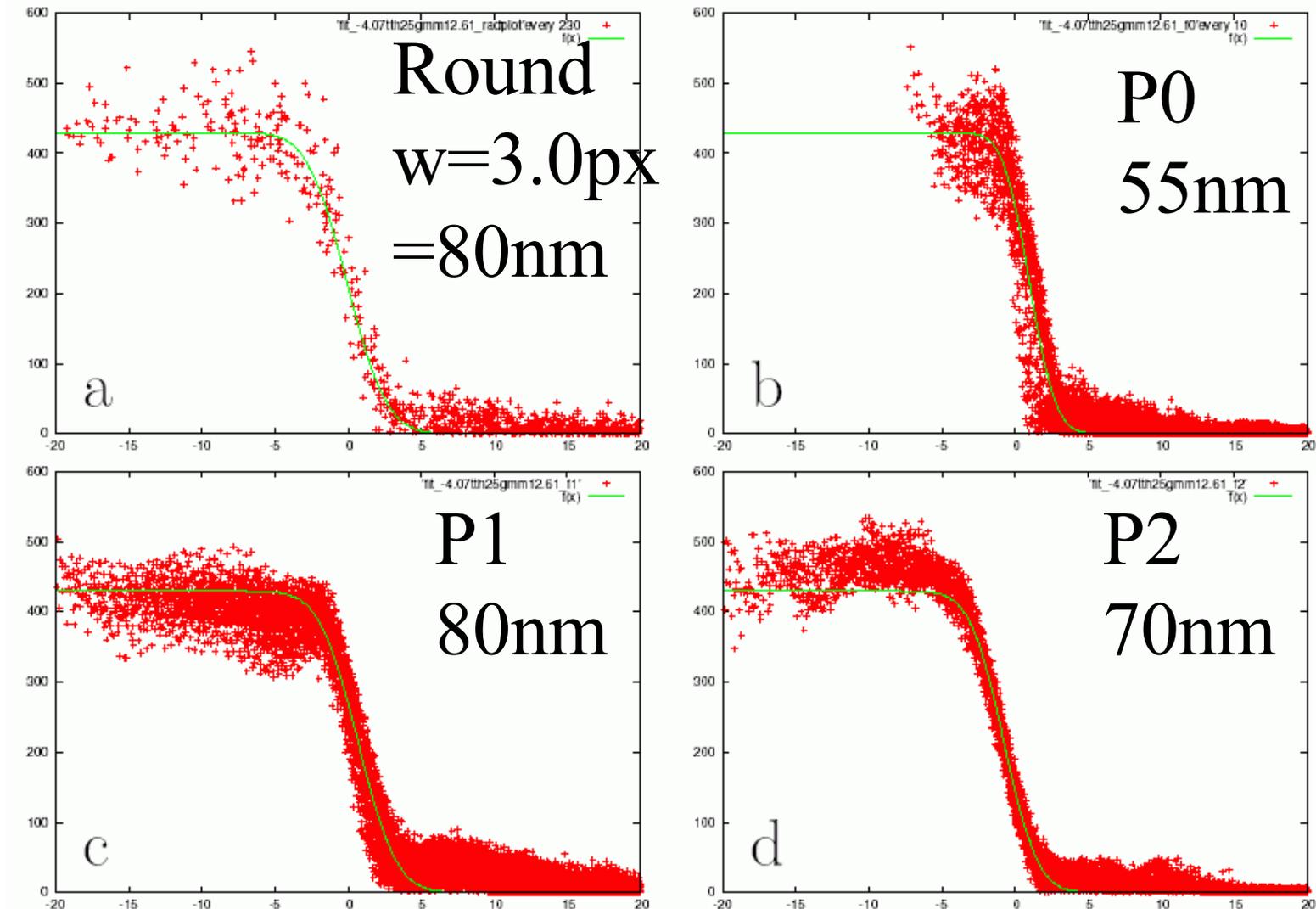


Porquerolles

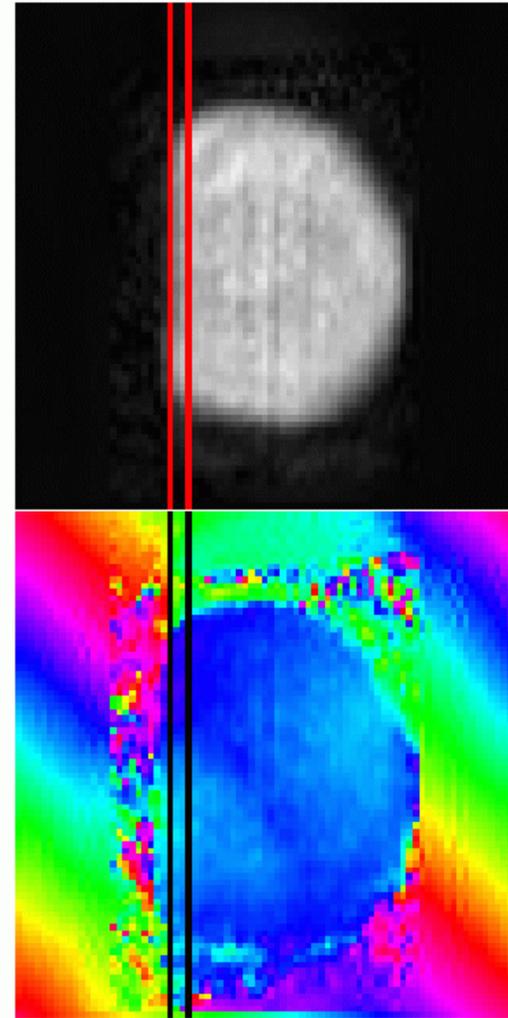
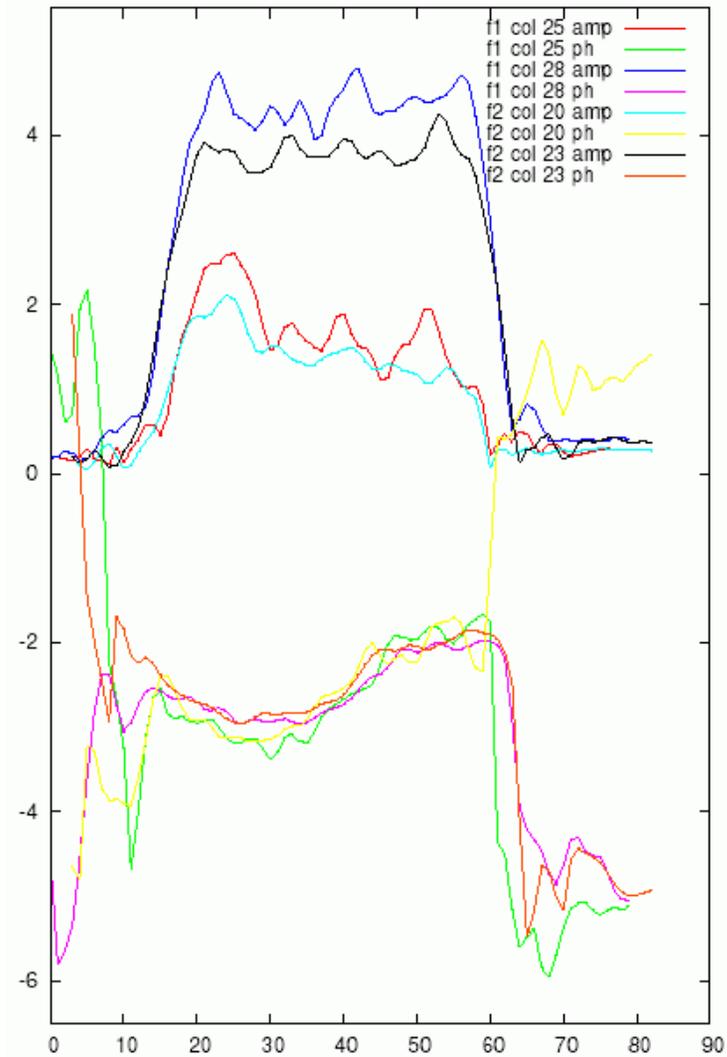
Facets of Equilibrium Crystal Shape



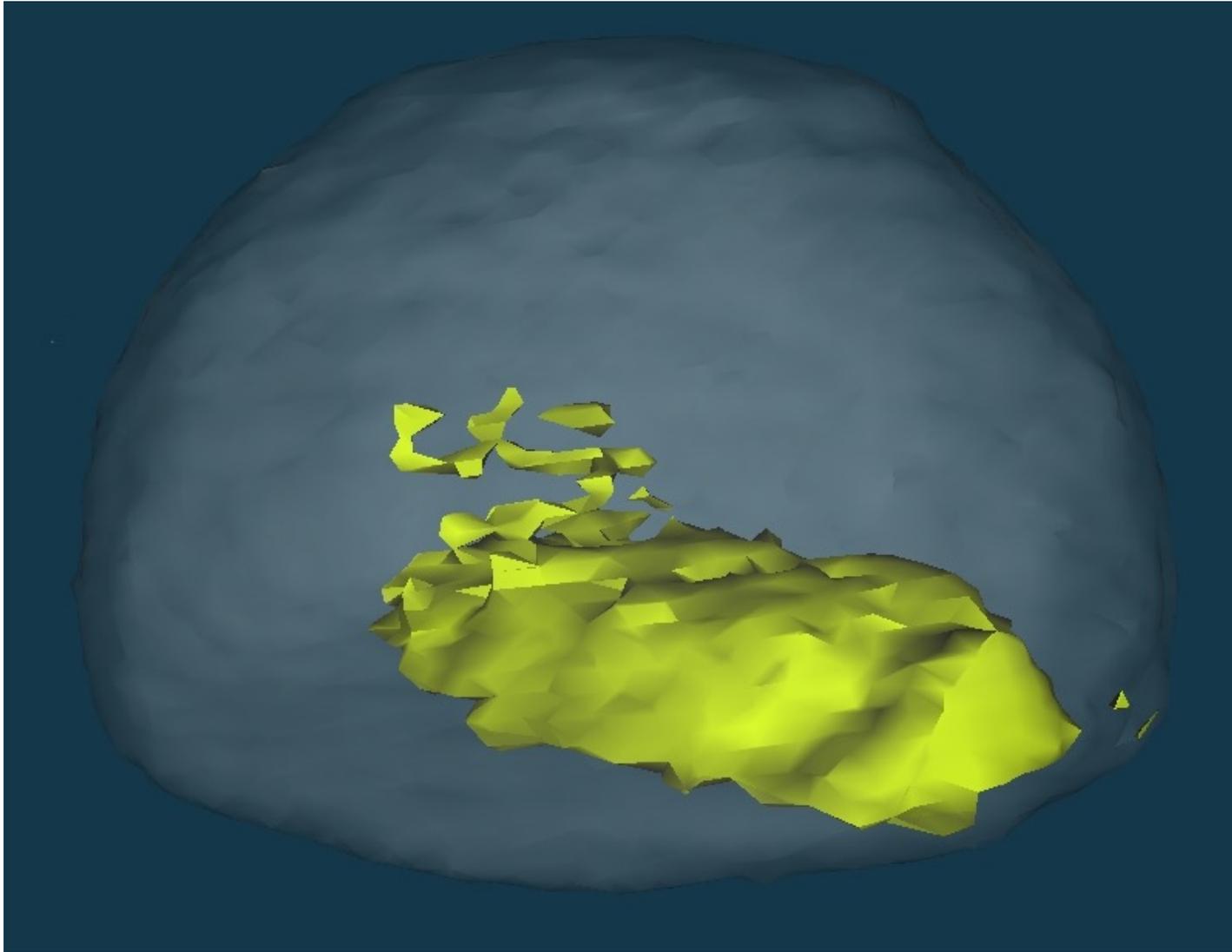
Density distribution across surface



Phase structure near substrate interface

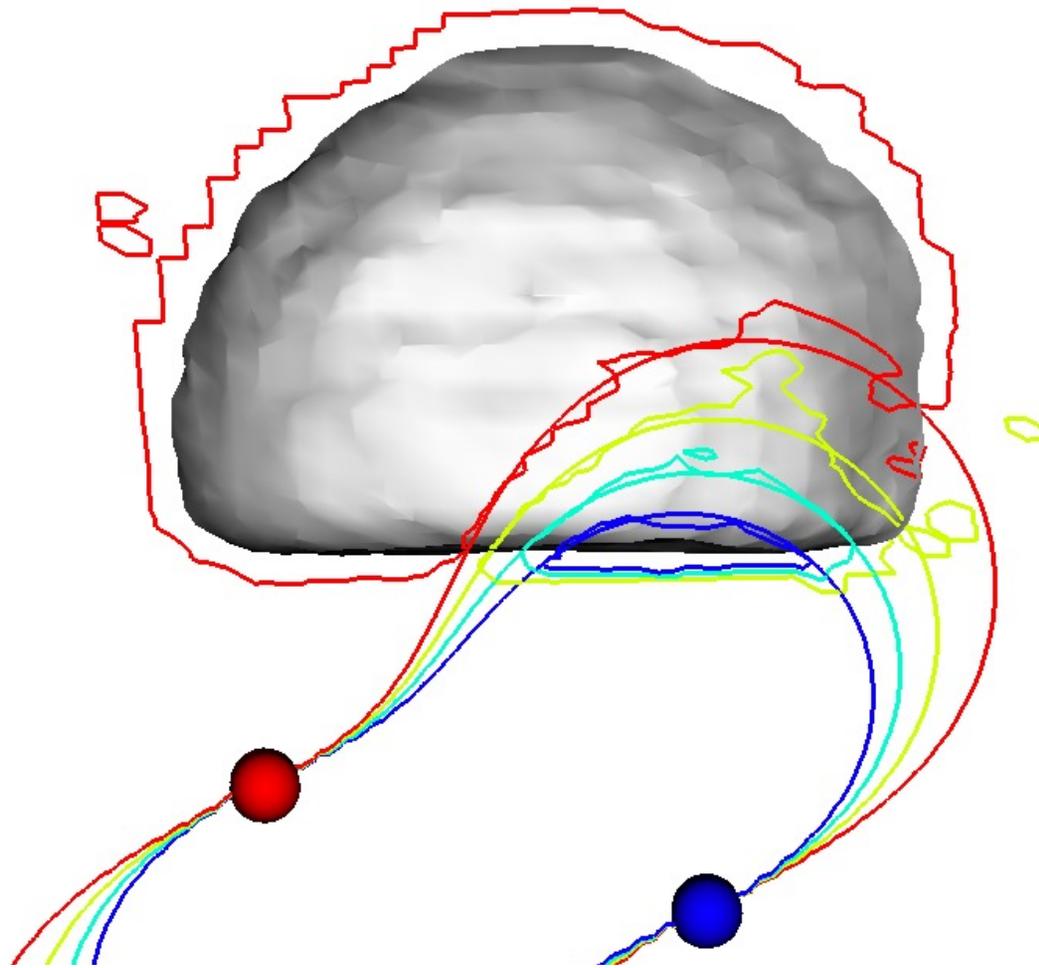


Modeling of 3D Phase Bump



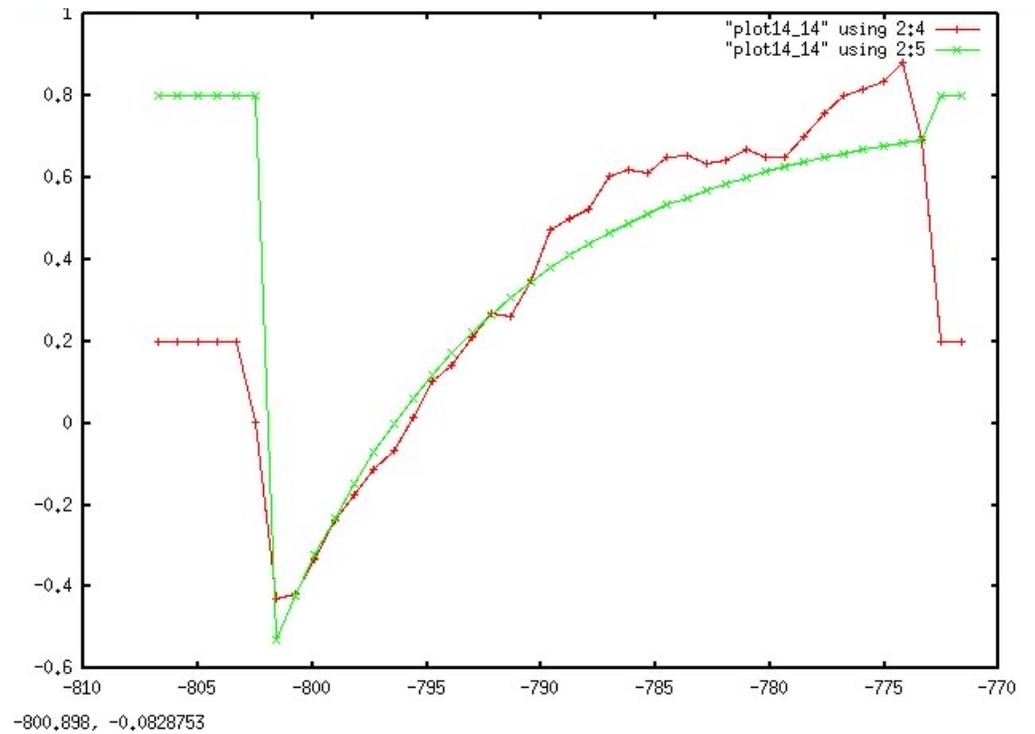
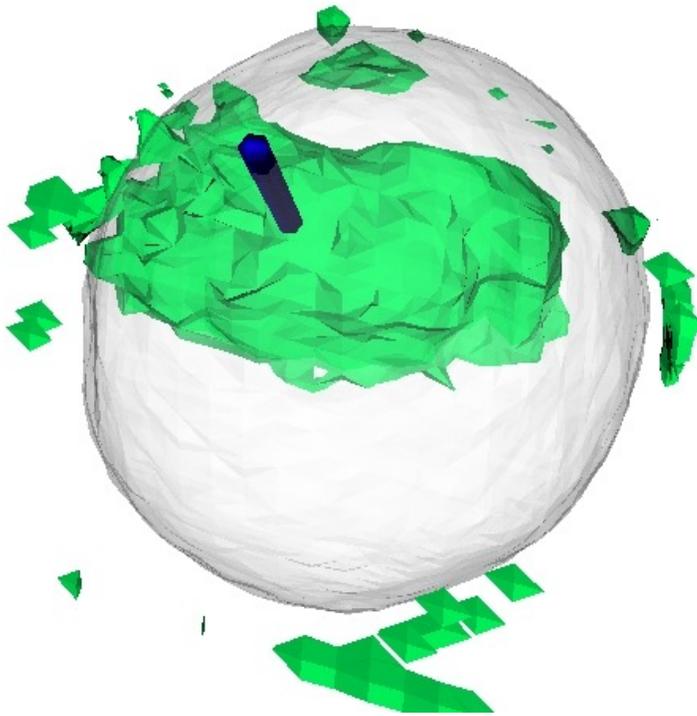
I. K. Robinson, Porquerolles

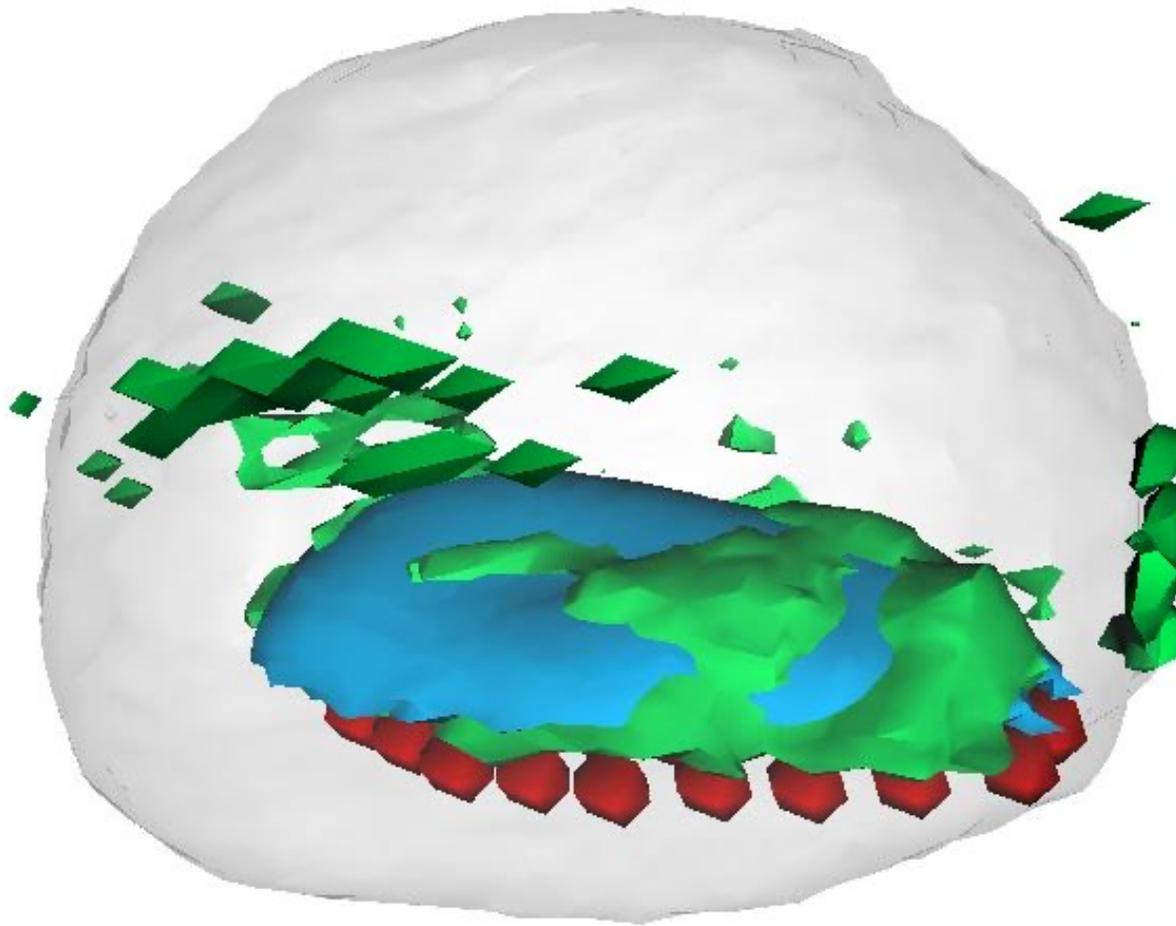
Field lines of Point Charges



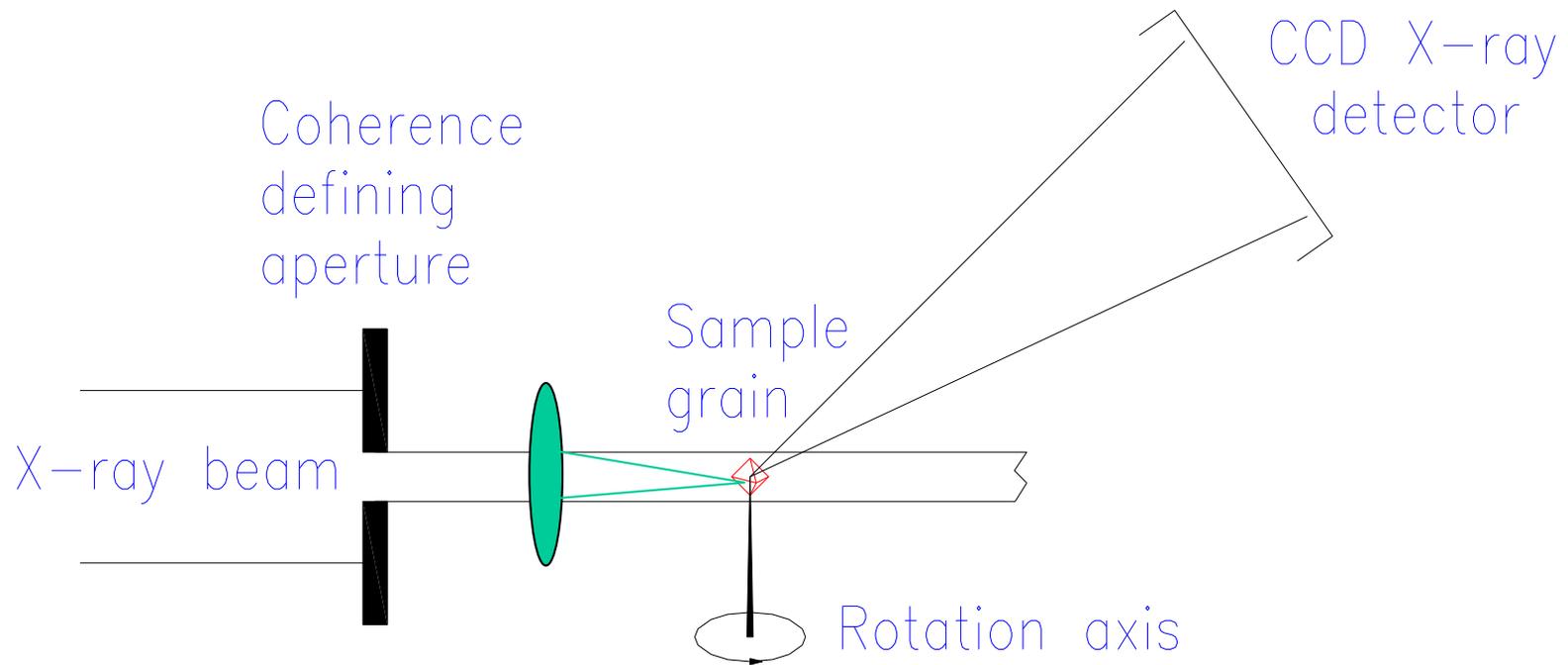
I. K. Robinson, Porquerolles

Line scan through phase

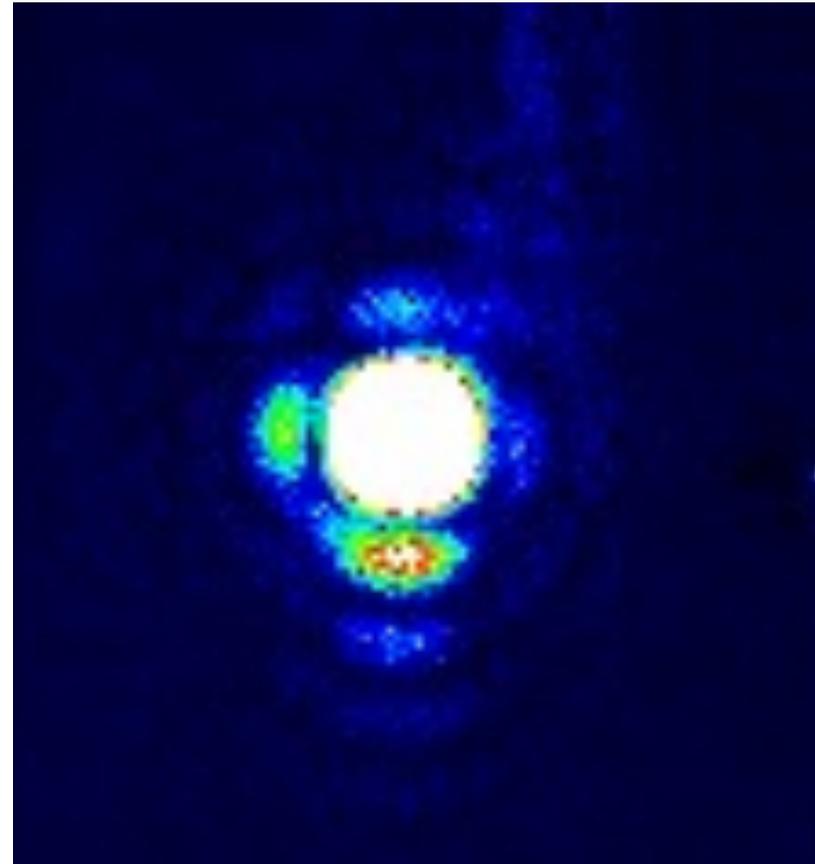
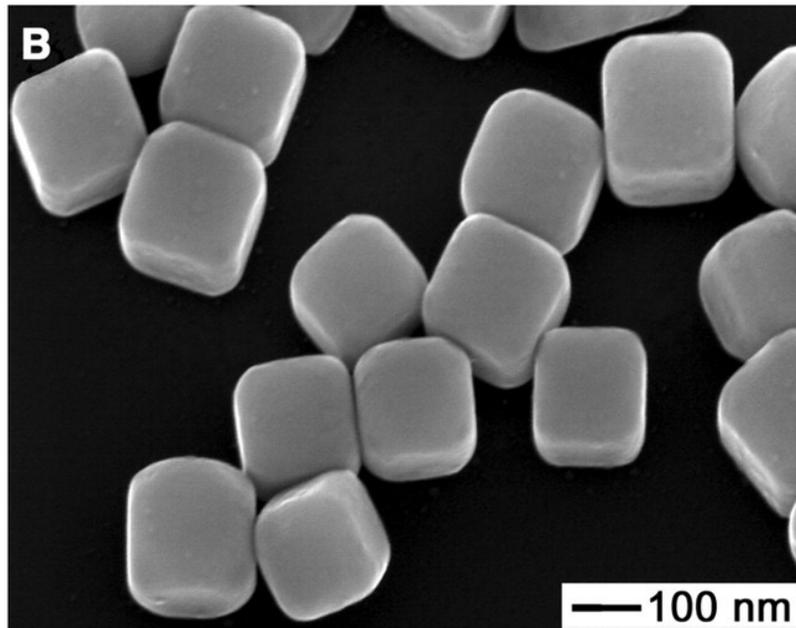




Lensless X-ray Microscope

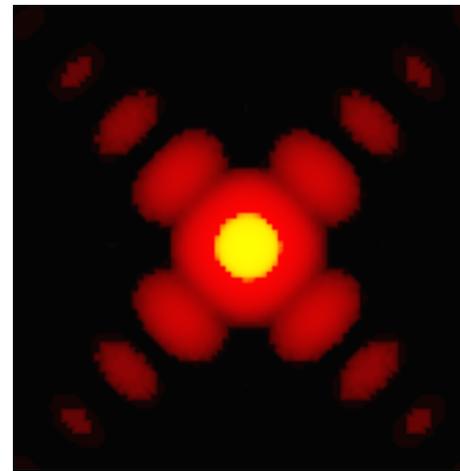
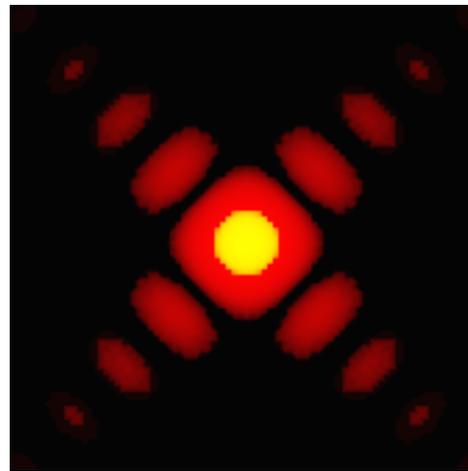


CXD from Silver Nanocubes



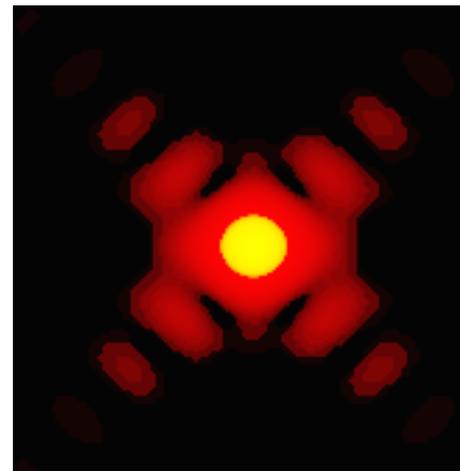
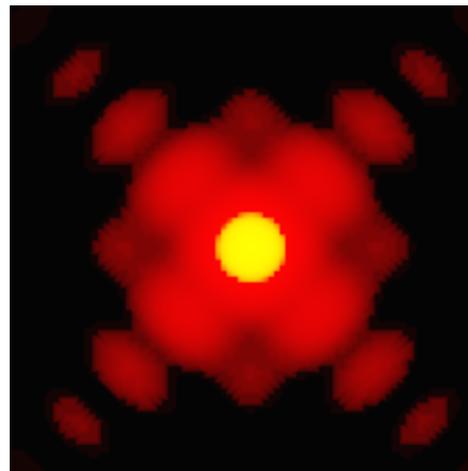
Yugang Sun and Younan Xia,
Science 298 2177 (2003)

Spherical and cylindrical waves



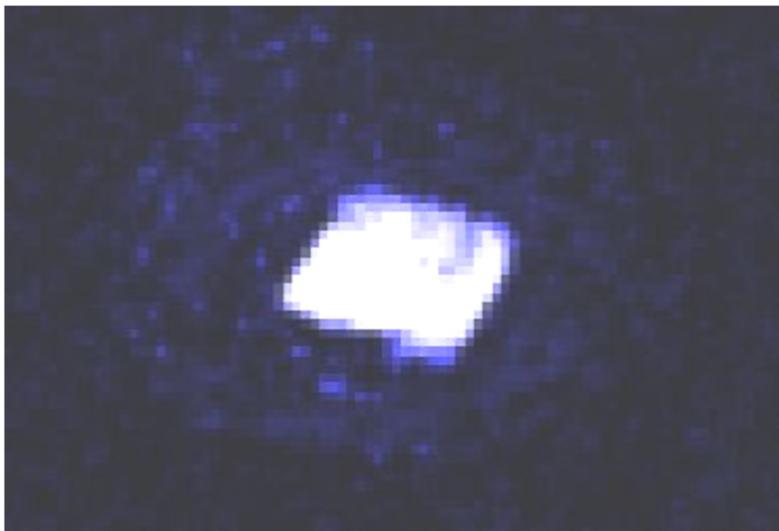
$\pi/4$

$\pi/2$

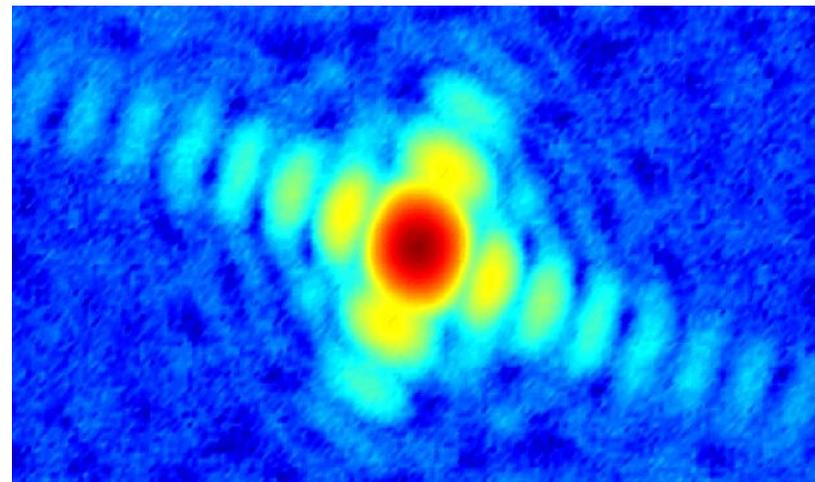
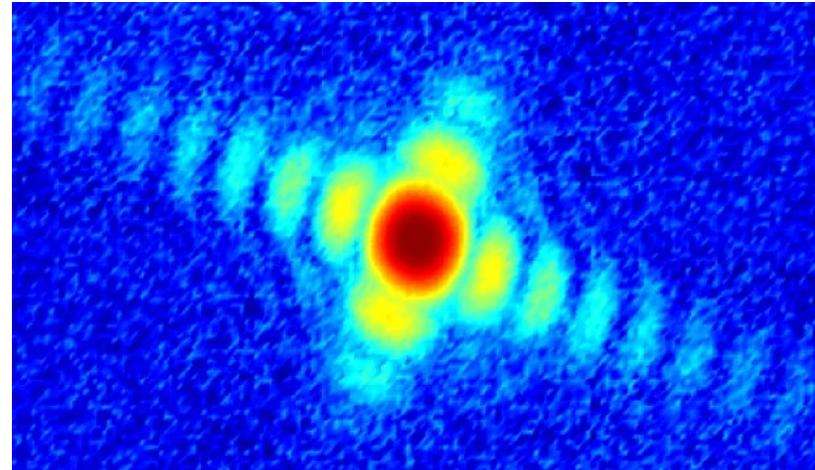


$\pi/4$ (x)

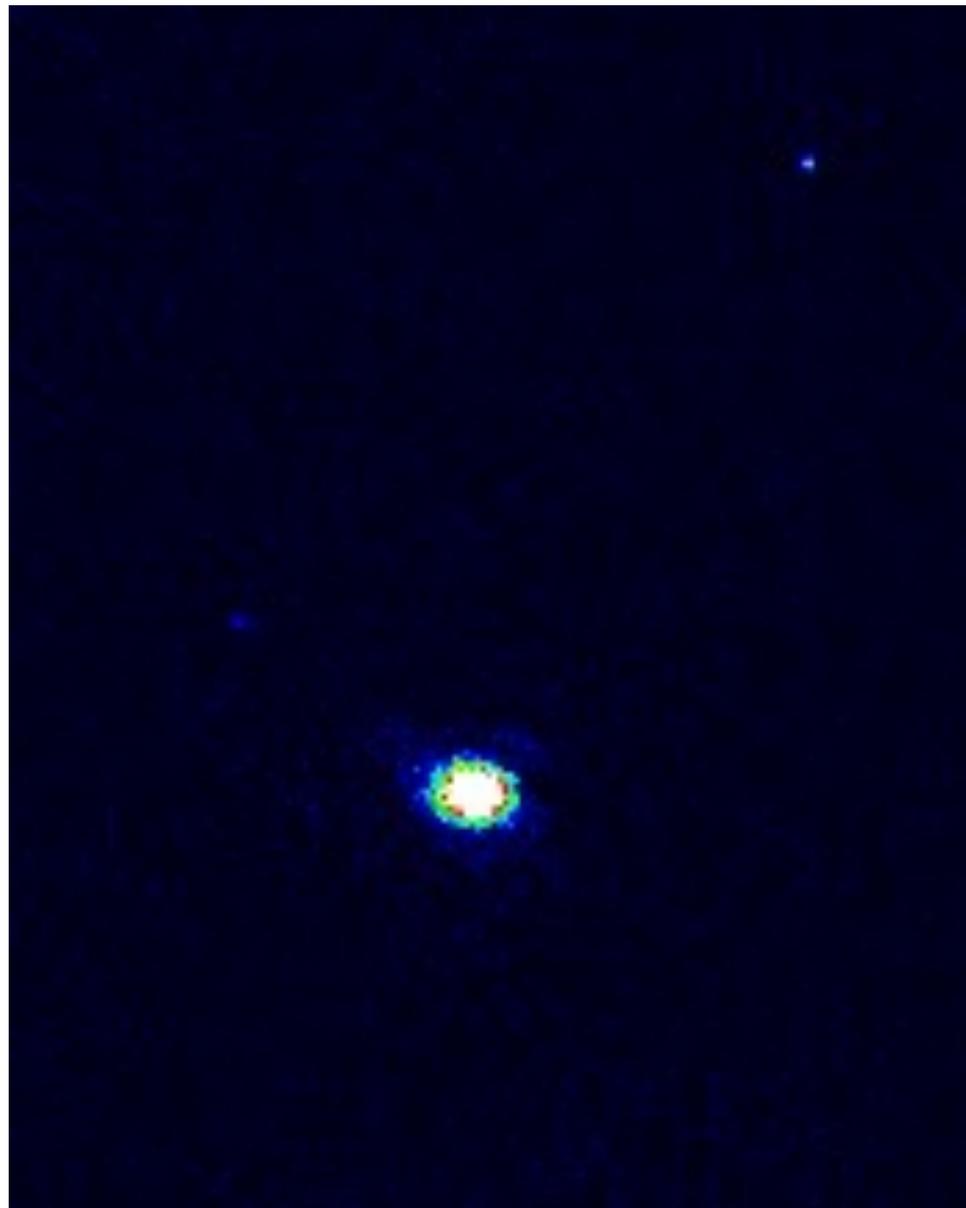
Reconstruction of Ag Nanocrystal

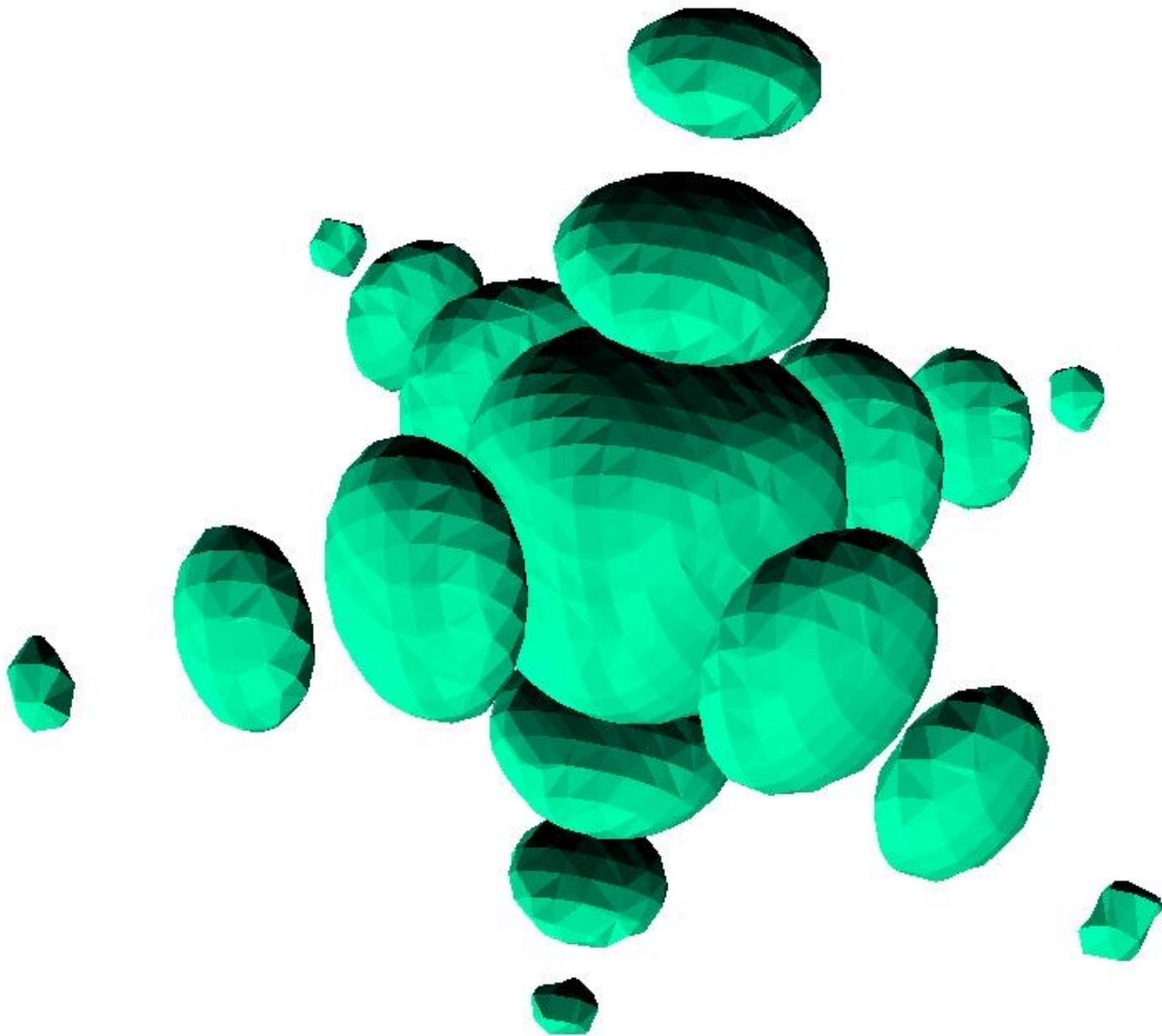


←→
200nm



Rocking
scan of Ag
cubes with
 0.01° steps





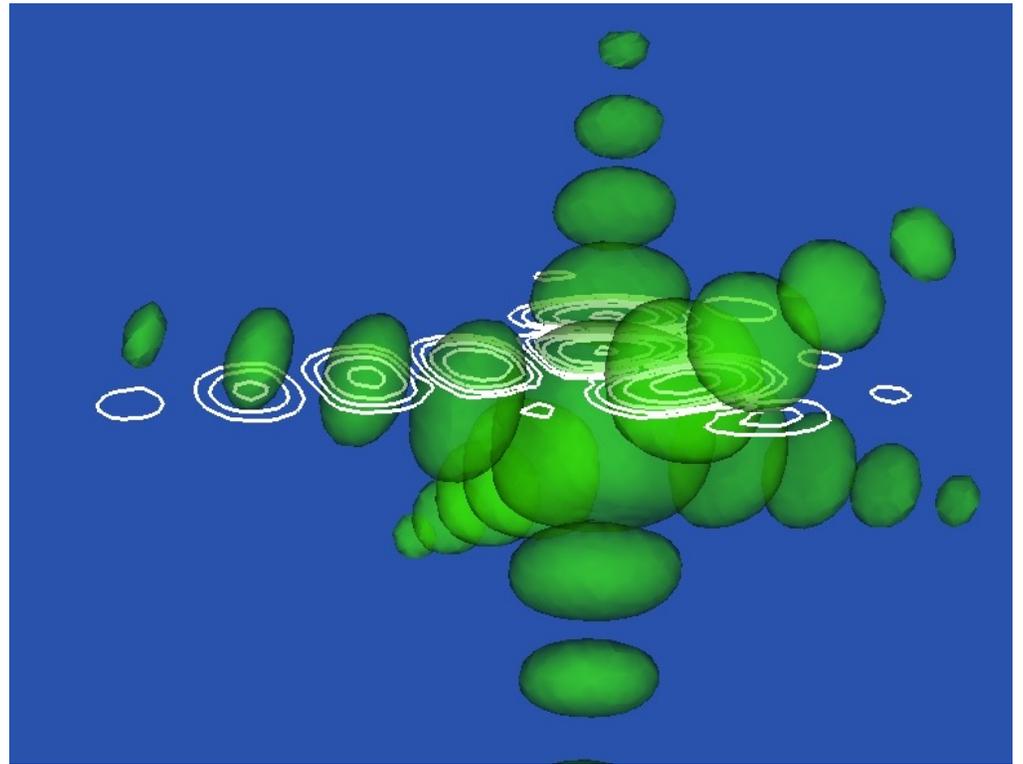
Miscentered Non-Centrosymmetric Reciprocal Space Slice



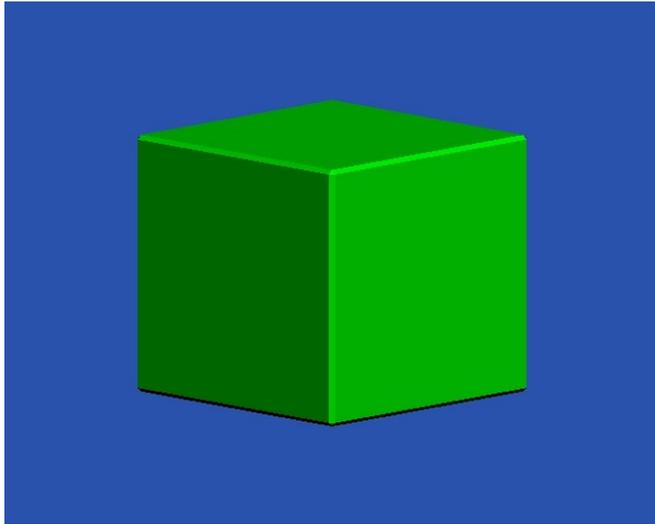
$$A_q \neq A_{\bar{q}}^*$$



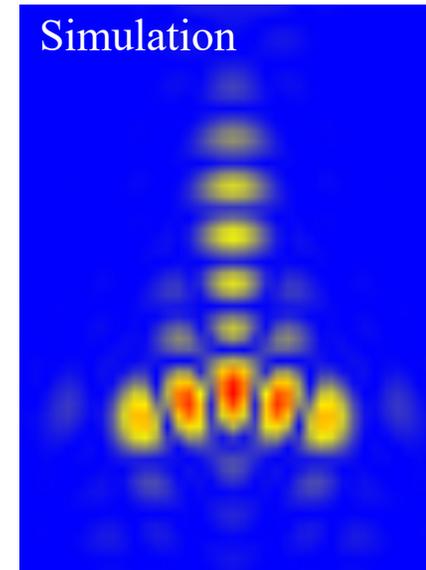
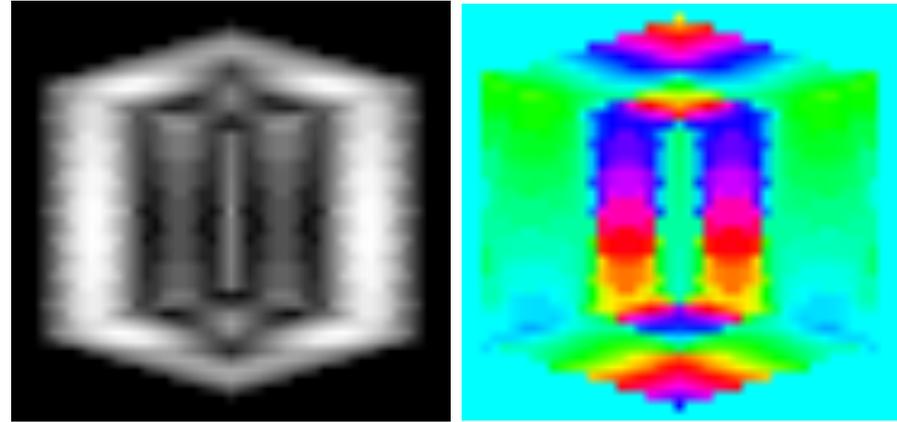
Real-space phase.



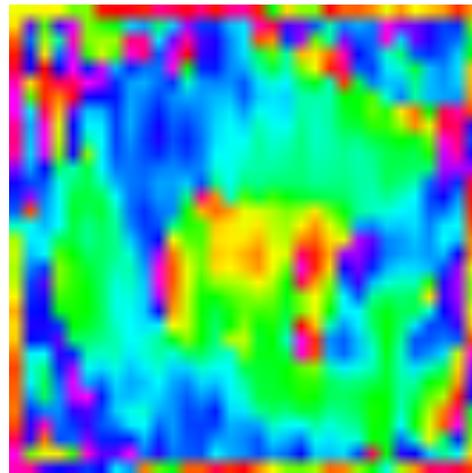
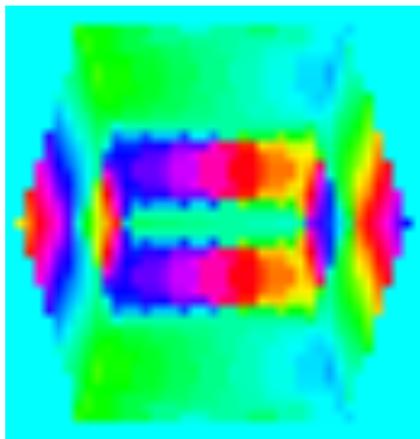
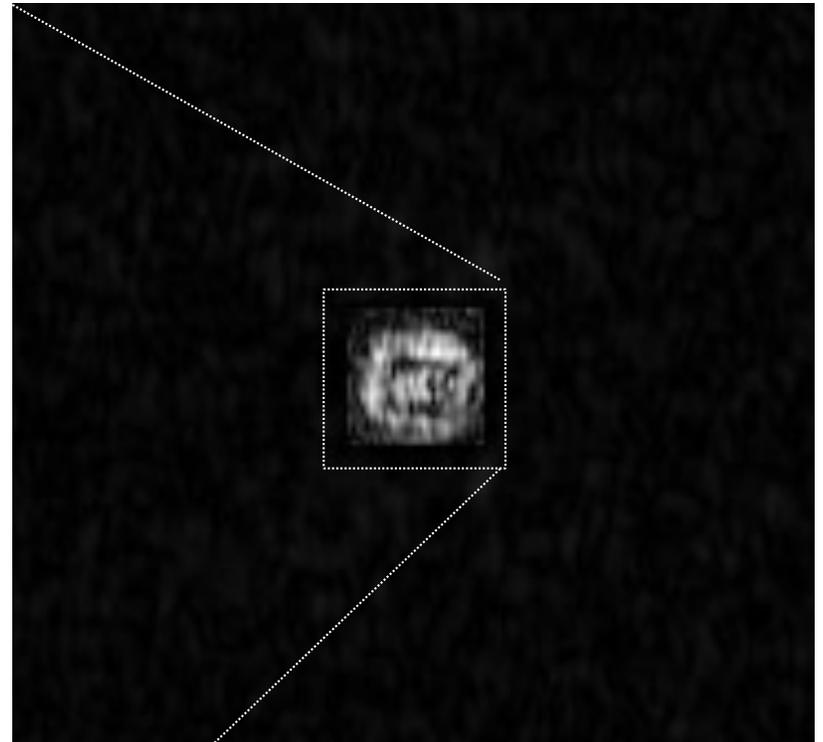
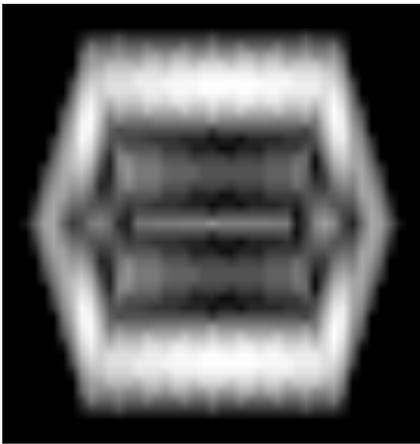
What can we expect to see?



$$A(\bar{q}_o + \bar{q}_z) = \sum_i \underbrace{\sum_z \rho(x, y, z)}_{u_i(x, y)} e^{2\pi i \bar{q}_z \cdot \bar{z}_i} e^{2\pi i (\bar{q}_o) \cdot \bar{r}_i}$$

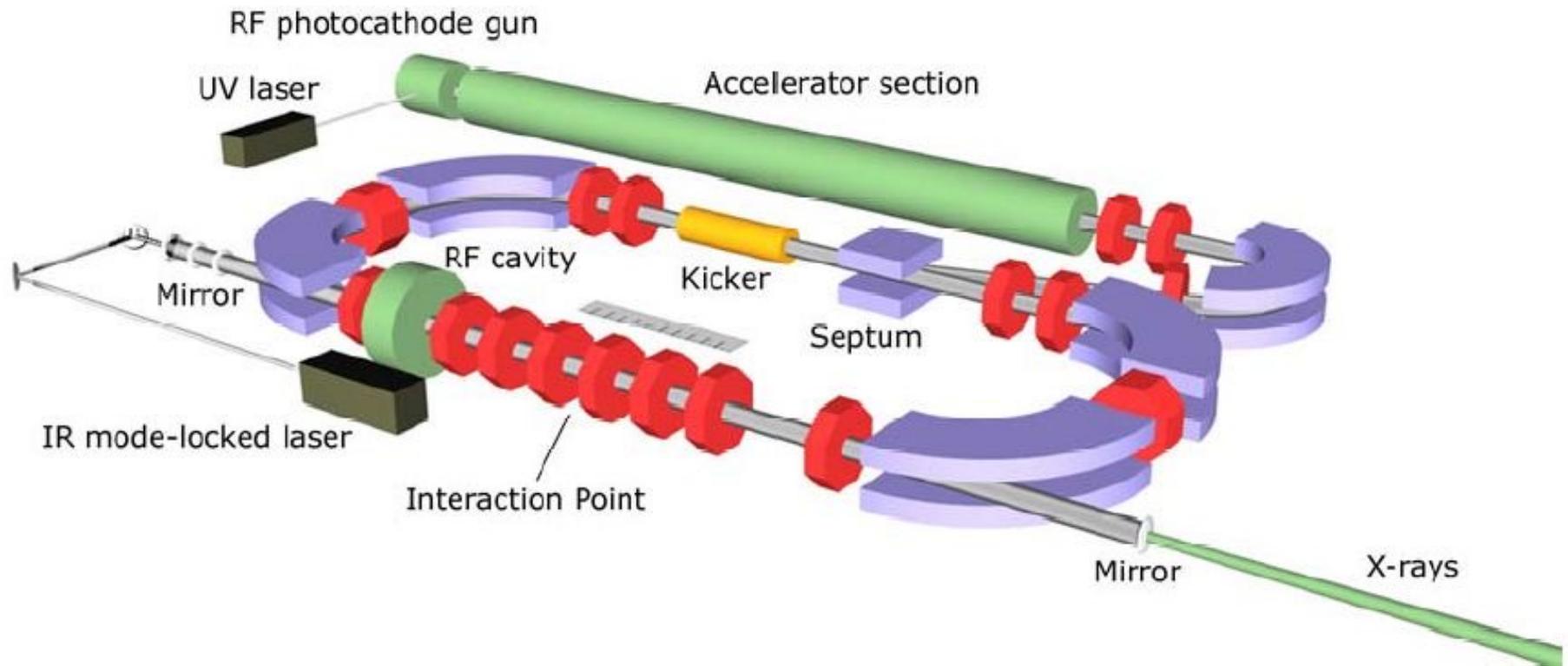


Direct Space Result (1000 Iterations)



Compact Light Source

Ron Ruth, Lyncean Technologies



Conclusions and Outlook

- Inversion of CXD demonstrated
- Spherical wave effects not yet seen
- Complex density represents strain
- Miscentered pattern inverts to complex object