

Three-dimensional Imaging by Inversion of Coherent X-ray Diffraction

- Ian Robinson
- Ivan Vartanyants
- Franz Pfeiffer
- Mark Pfeifer
- Garth Williams
- Meng Liang
- Ross Harder

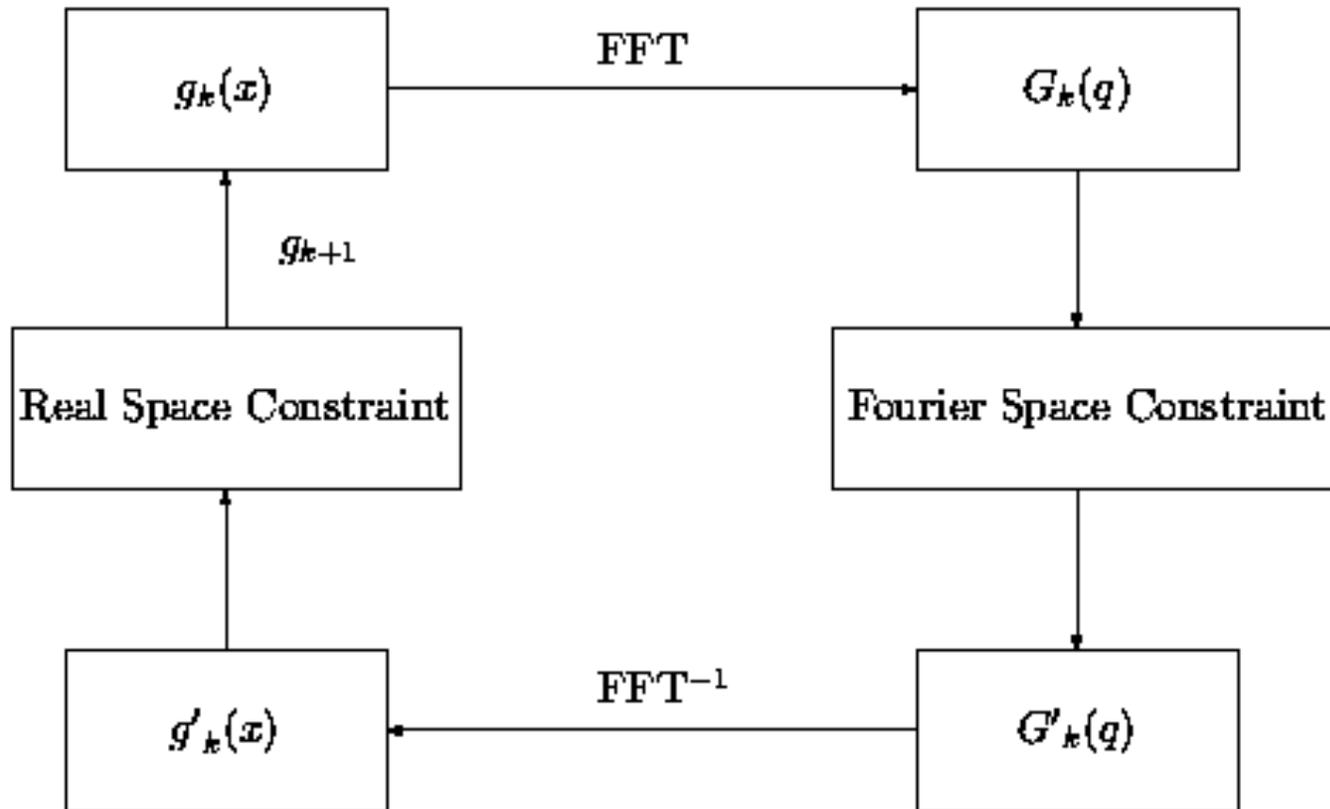
Department of Physics
University of Illinois

APS-CNM Workshop 3
Hard X-ray Nanoprobe
3 May 2005

Outline

- Coherence can be used for Structure
- The **Phase** Problem
- Nanocrystal Shapes
- Imaging of **Phase** Objects
- How small can we go?

Generic “Error Reduction” method



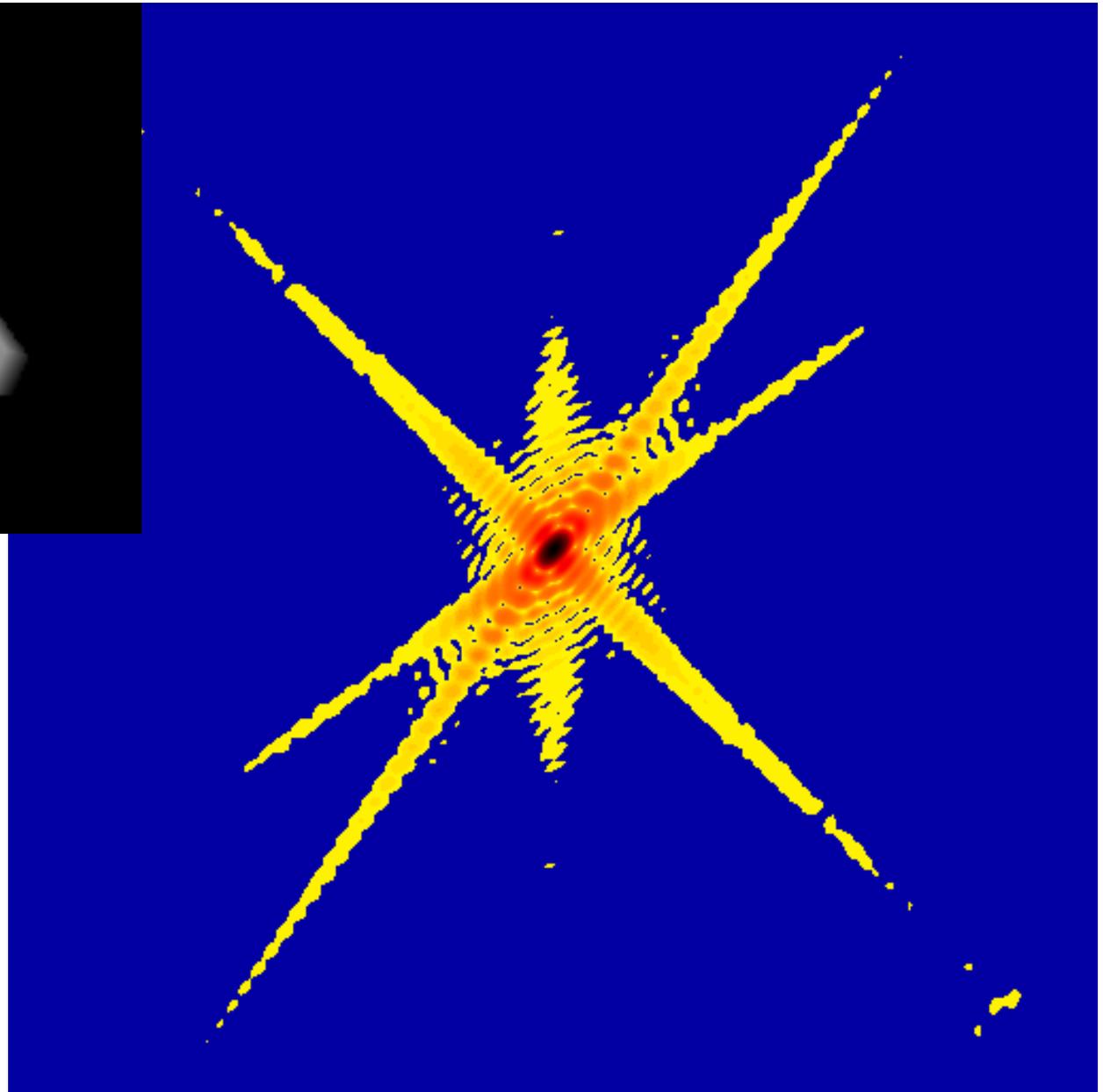
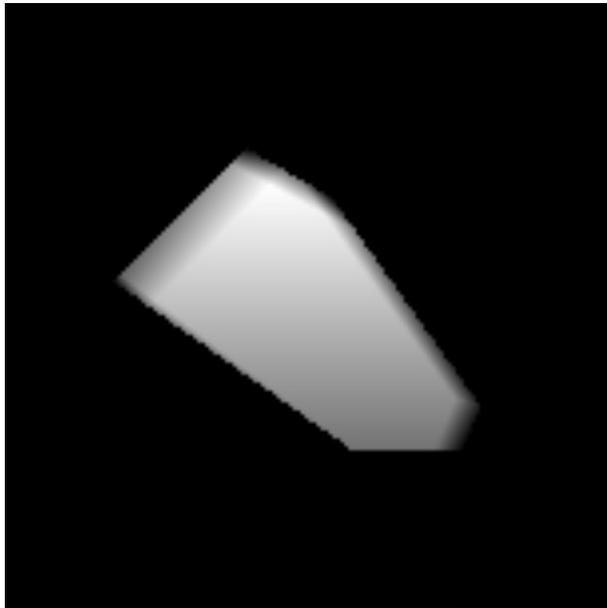
J. R. Fienup *Appl. Opt.* 21 2758 (1982)

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

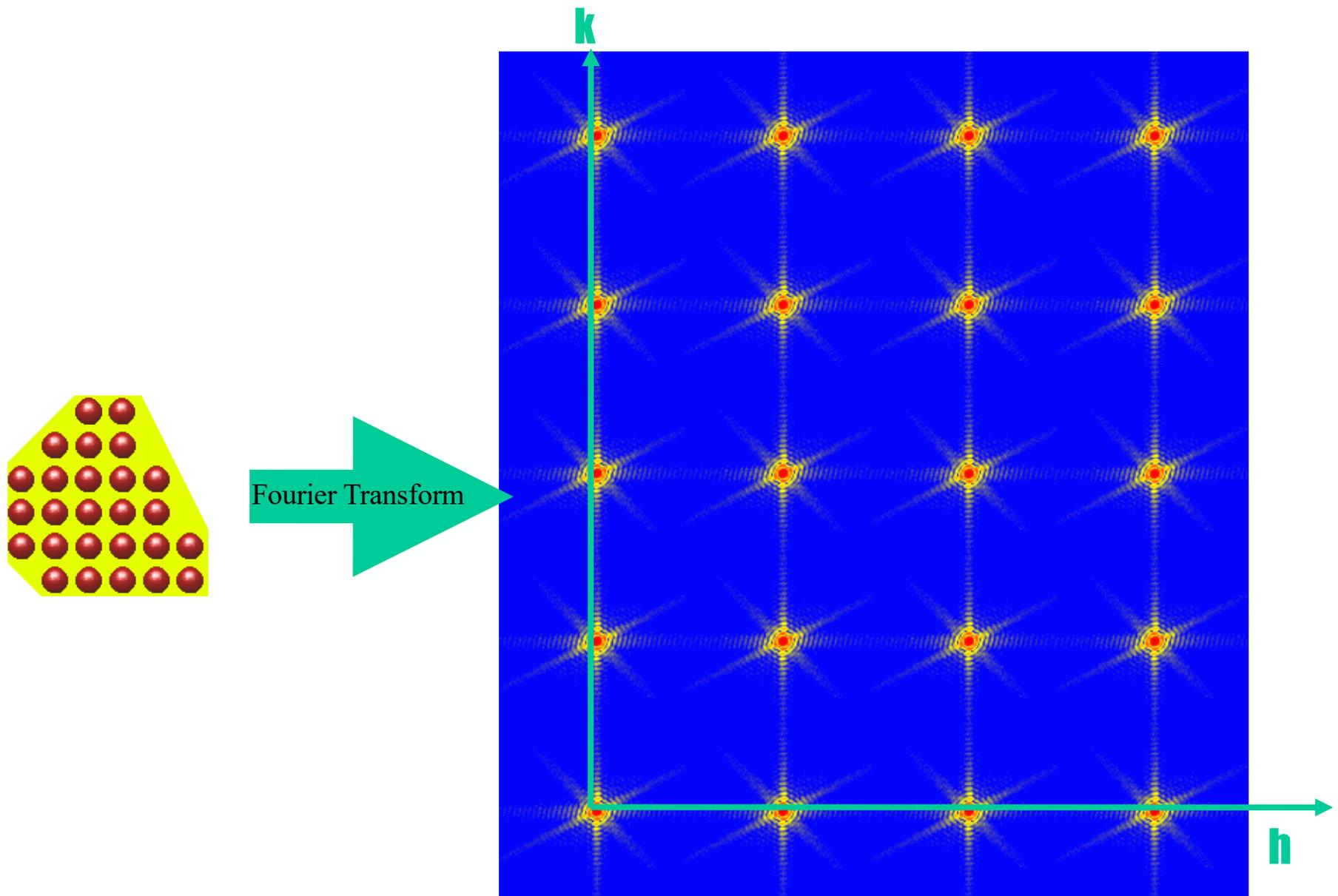
Real-space Constraints in Crystallography

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’ ($d > 1$)
- Uses memory to avoid stagnation (Fienup HIO)

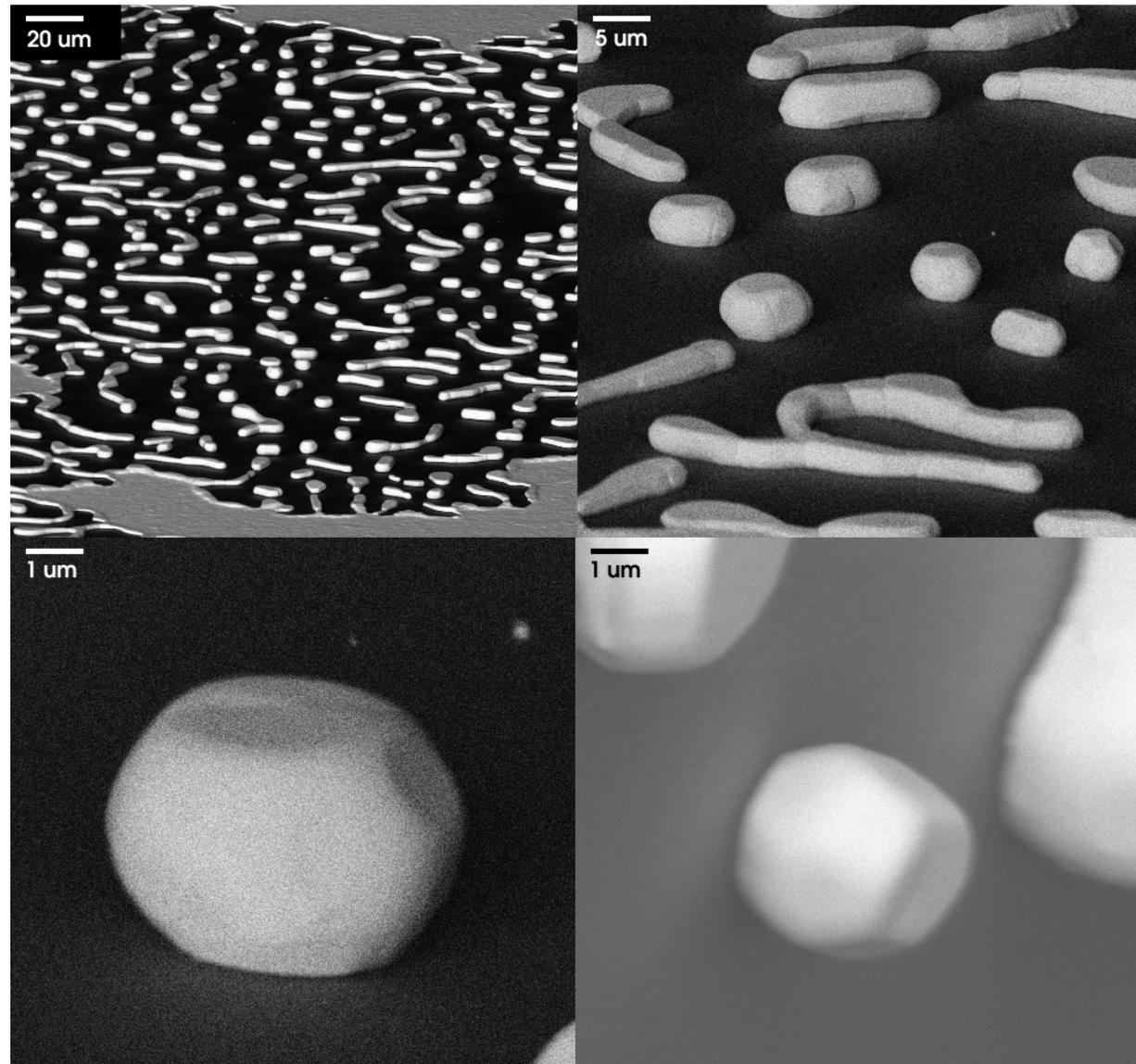


Coherent Diffraction from Crystals



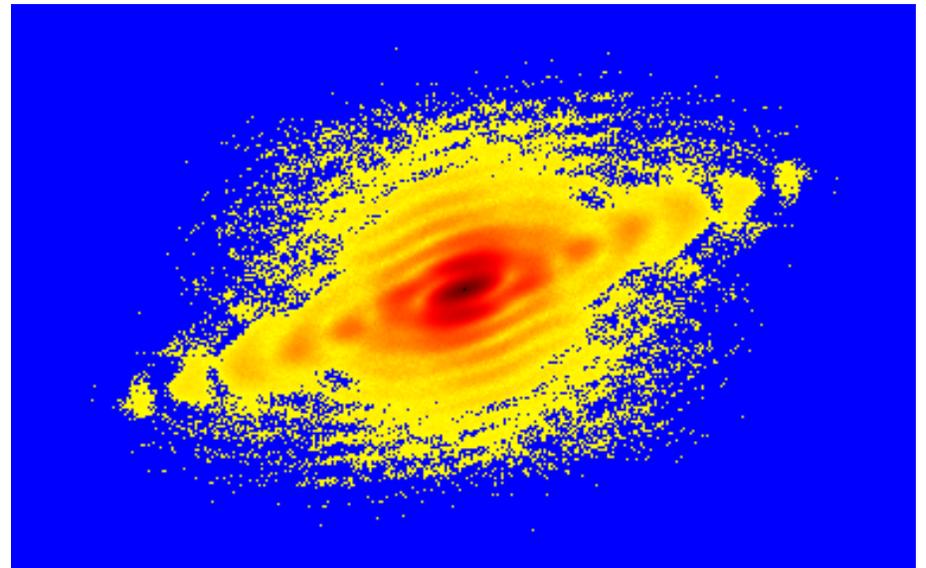
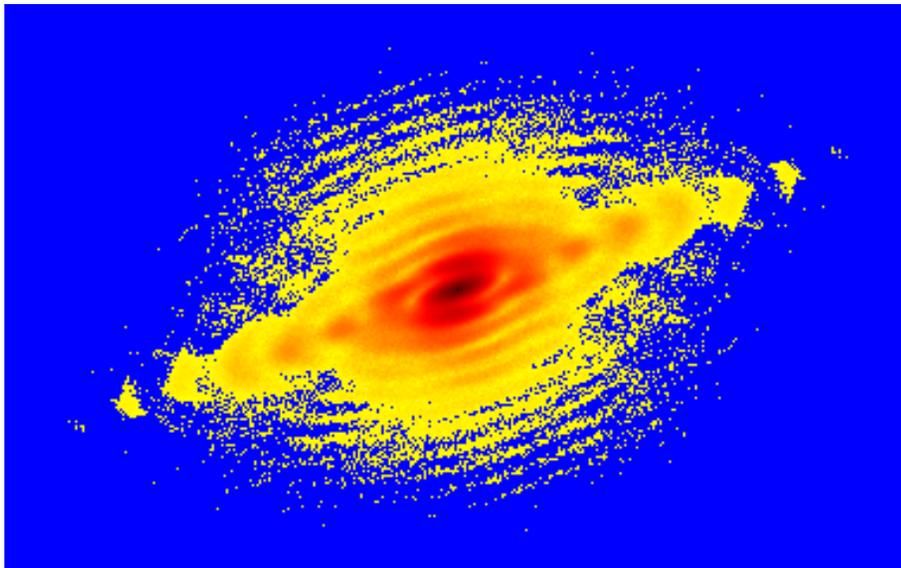
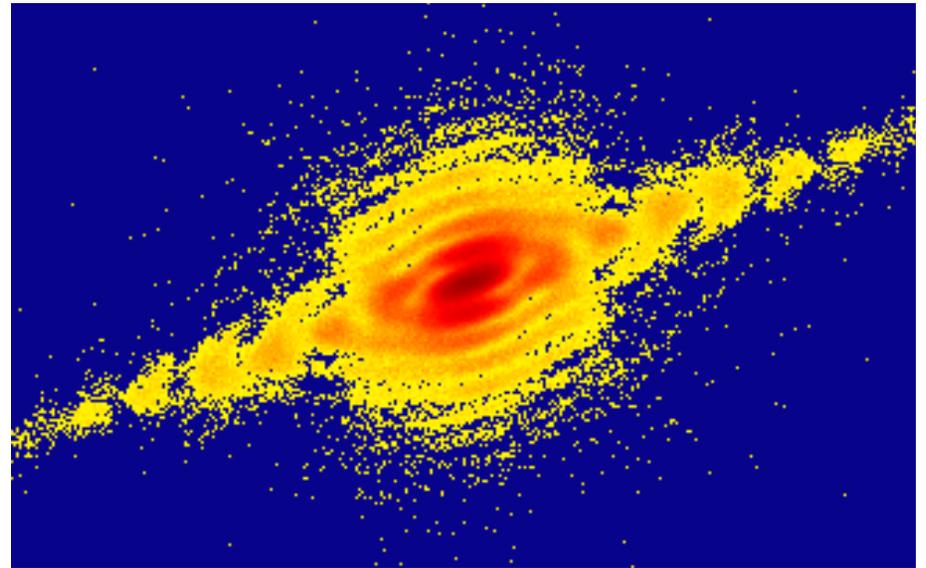
SEMS

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

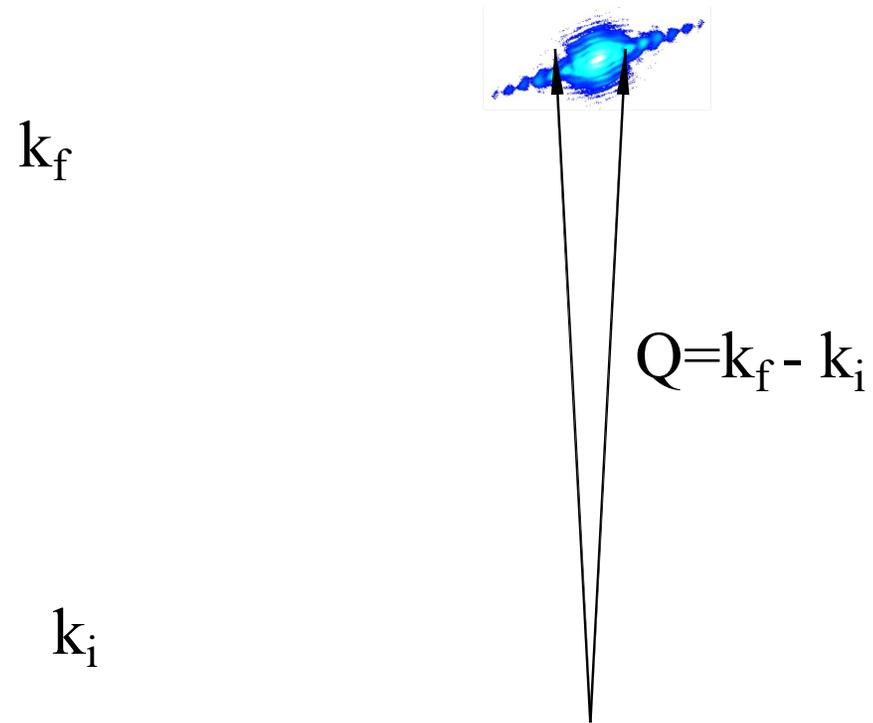


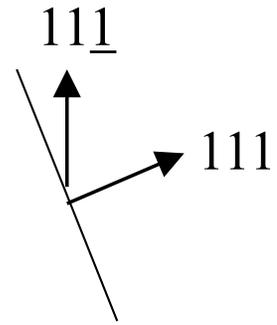
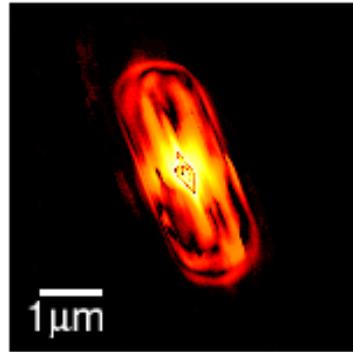
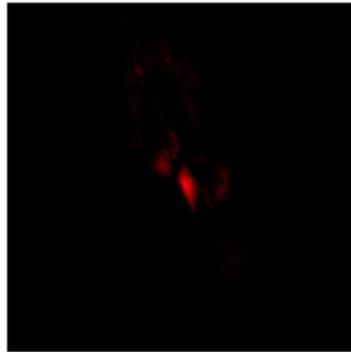
Symmetrized Data and two best fits

Chisq=0.0005

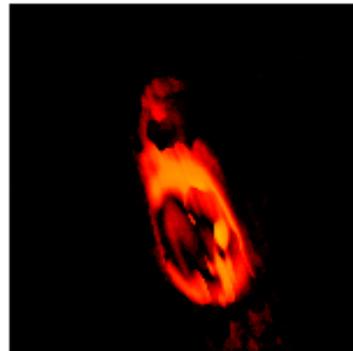
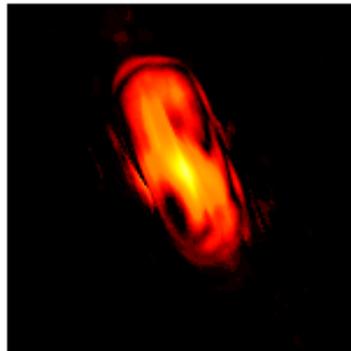
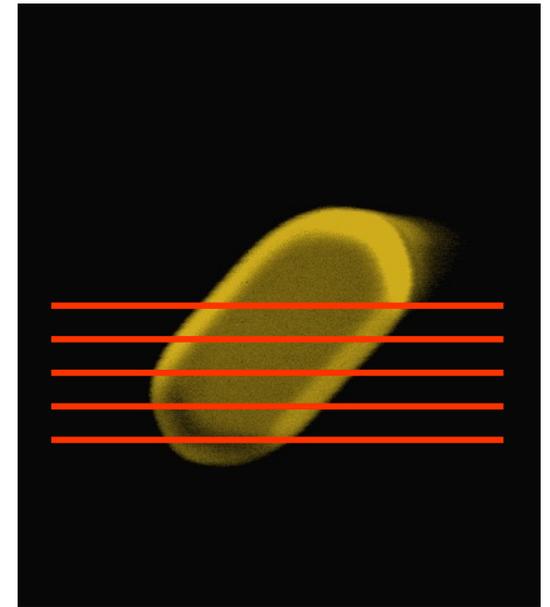
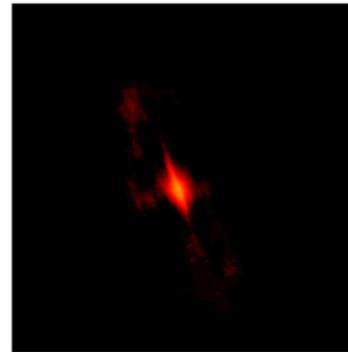
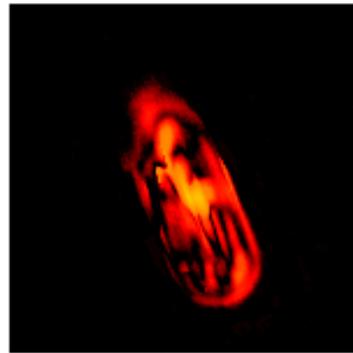
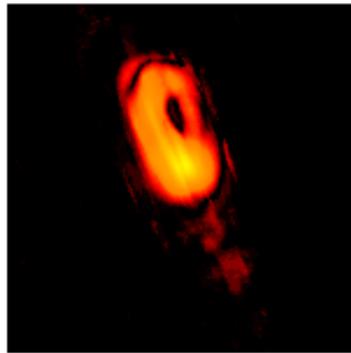
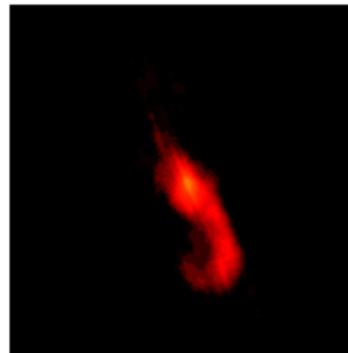
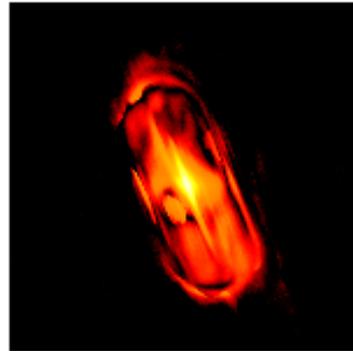
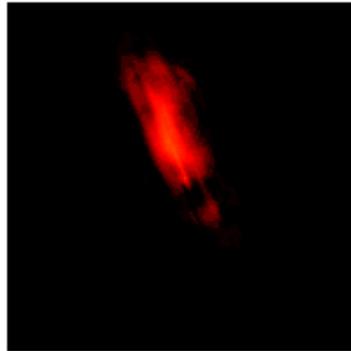


3D Diffraction Method

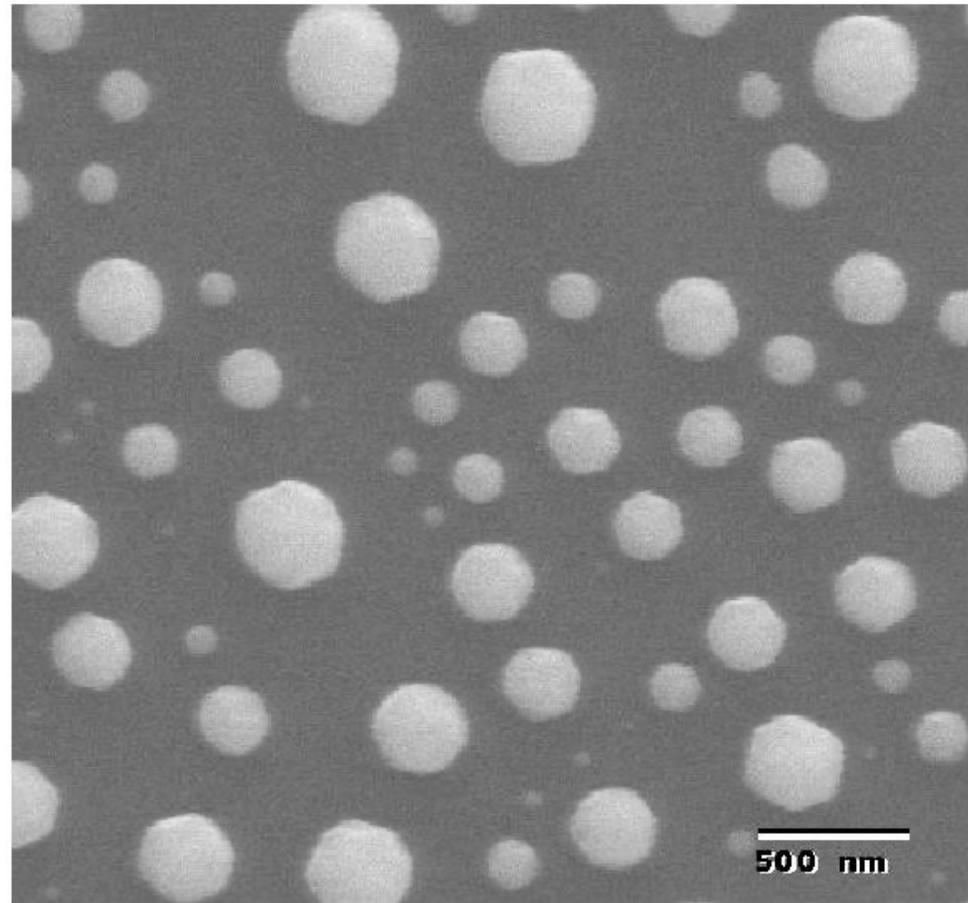




Slices through
plan view SEM:



In situ growth of Pb crystals



Good statistics, 3D diffraction data

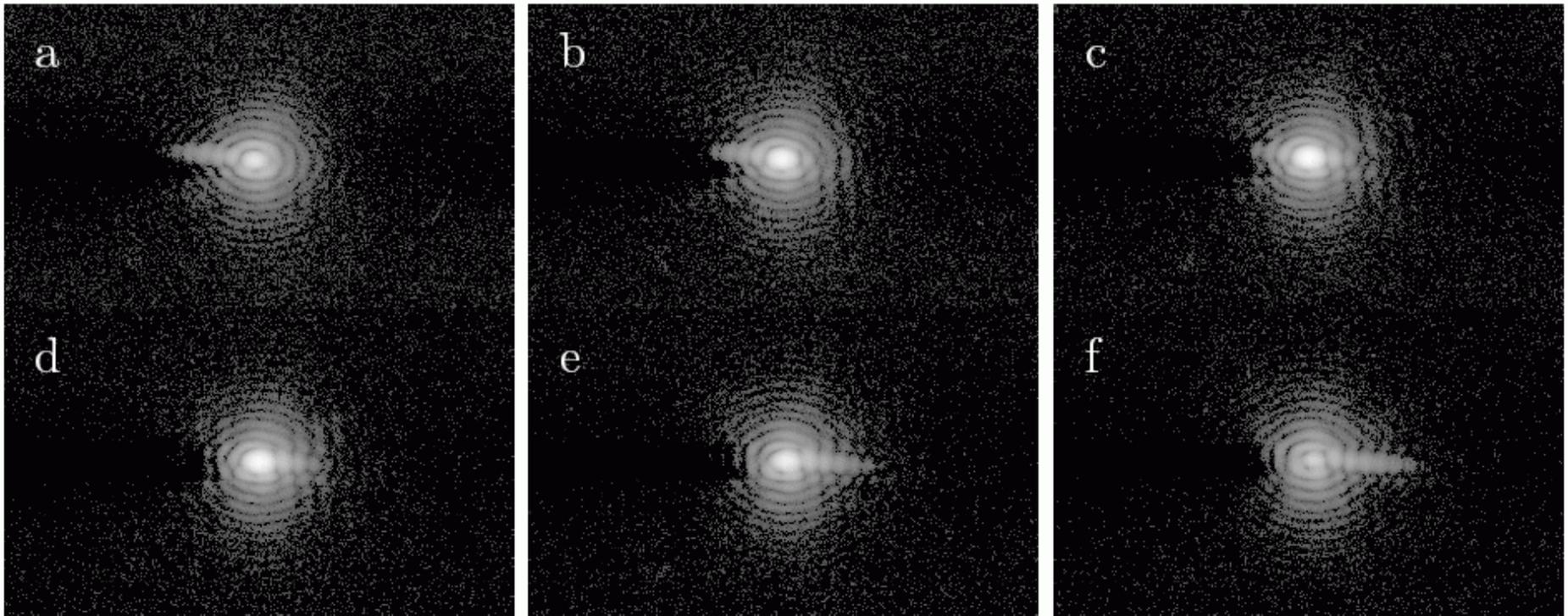
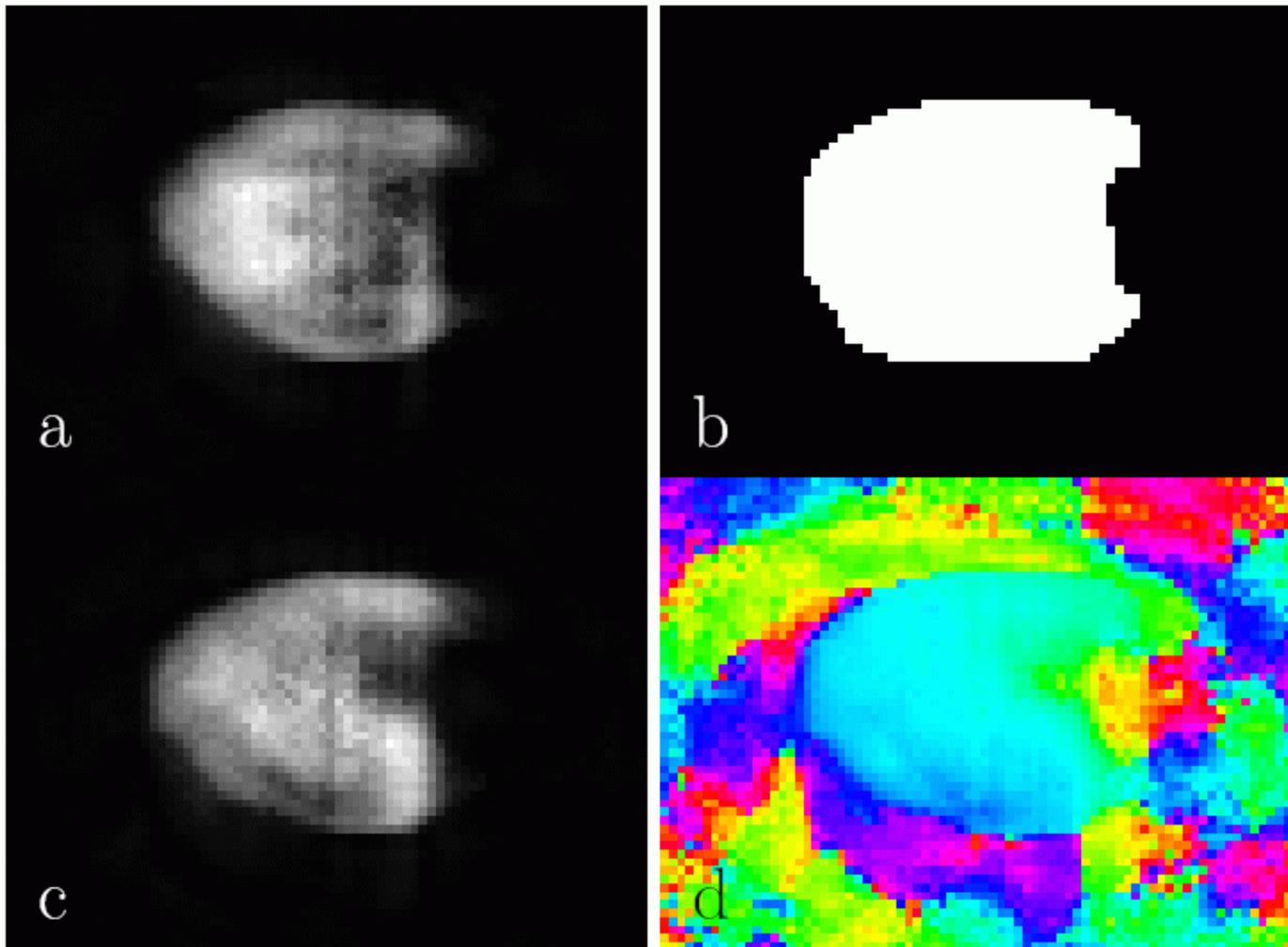
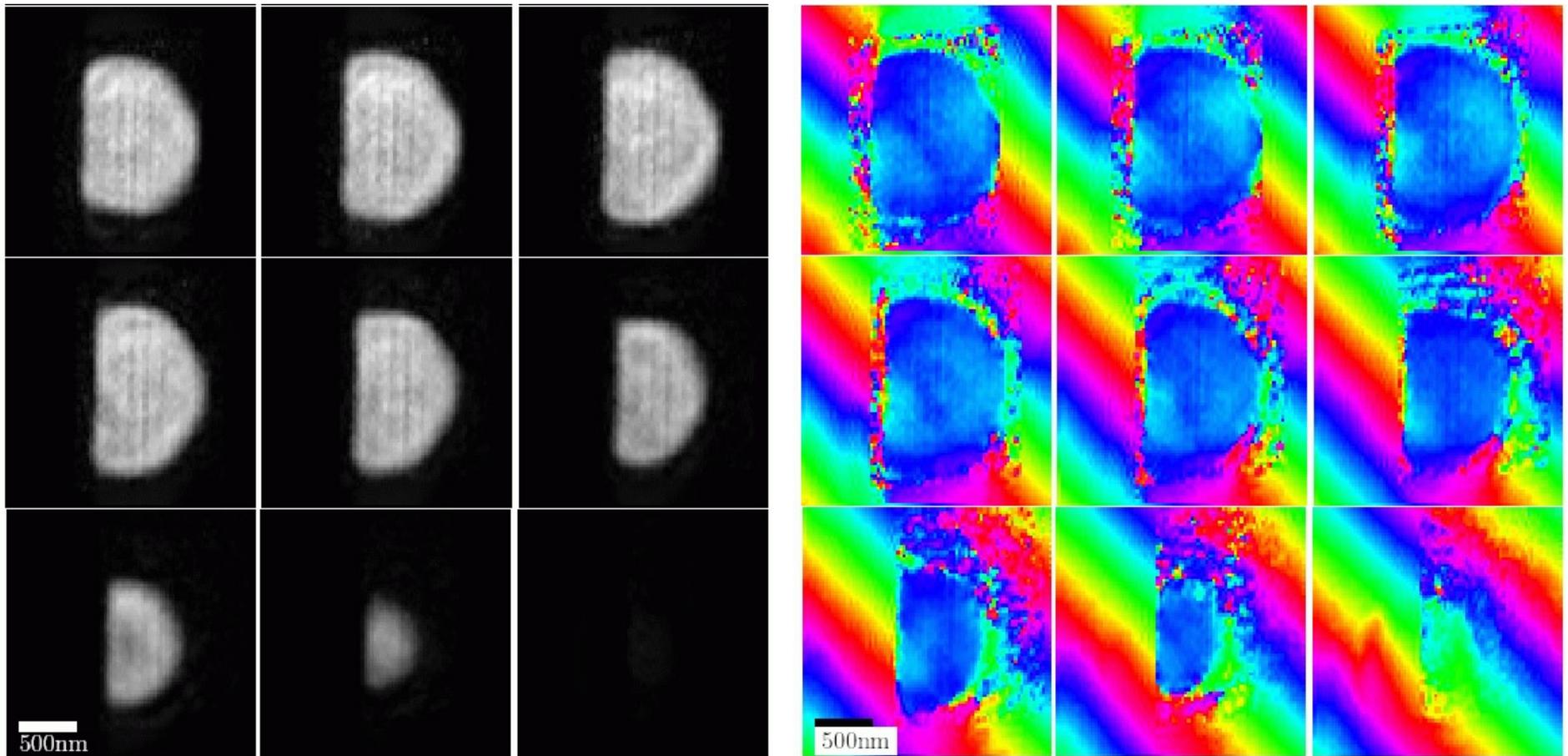


Figure 4.12: Center slices from 3D CXD pattern from Pb sample, on a log scale. Data file 296 from 10/03.

Learn shape of “tight” support

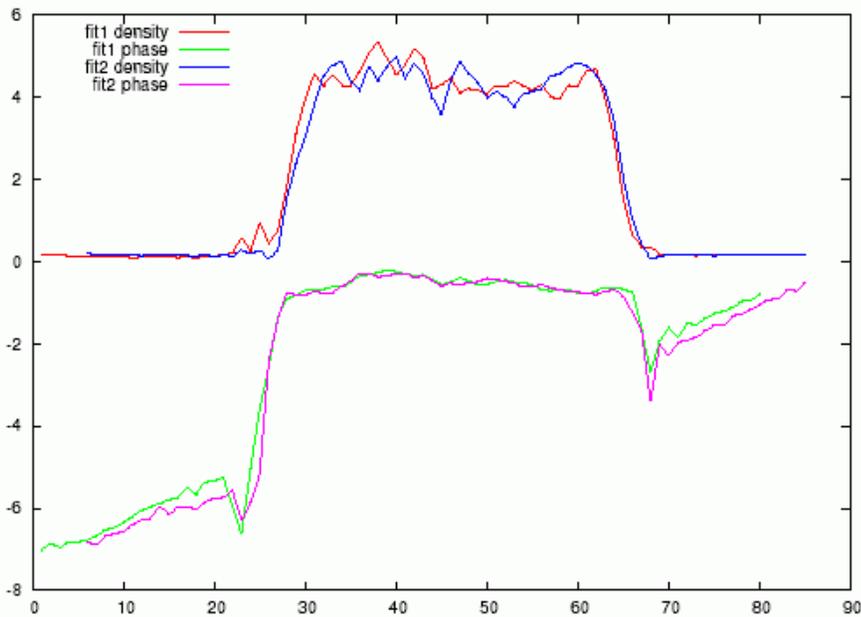


Then refine amplitude *and* phase

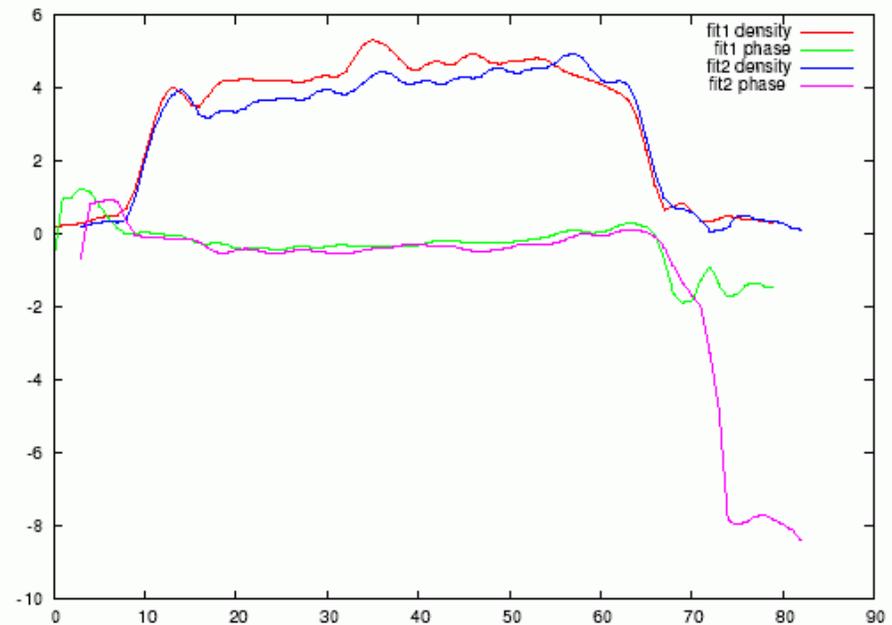


Amplitude/phase cross sections

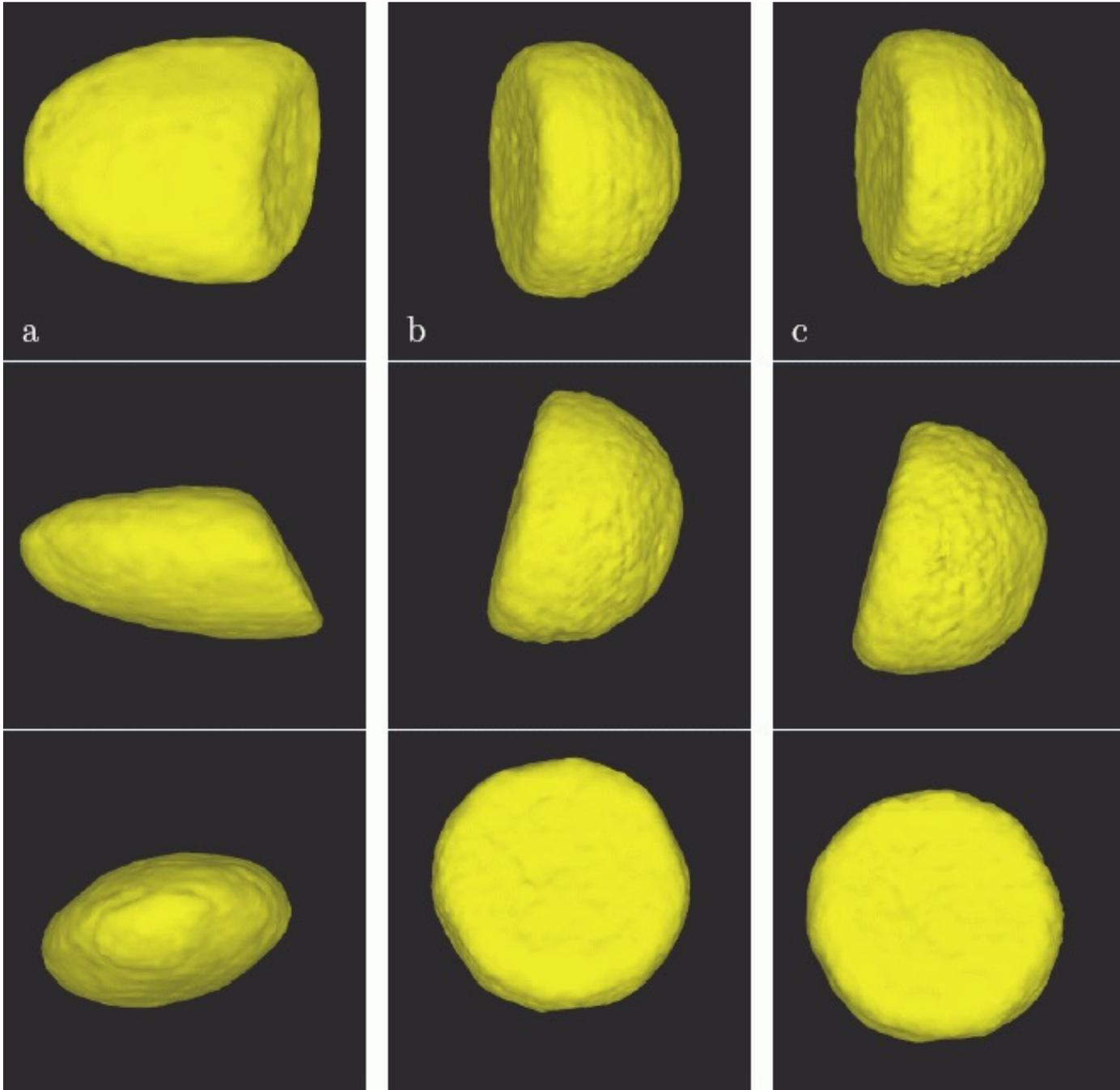
x-striations do not reproduce



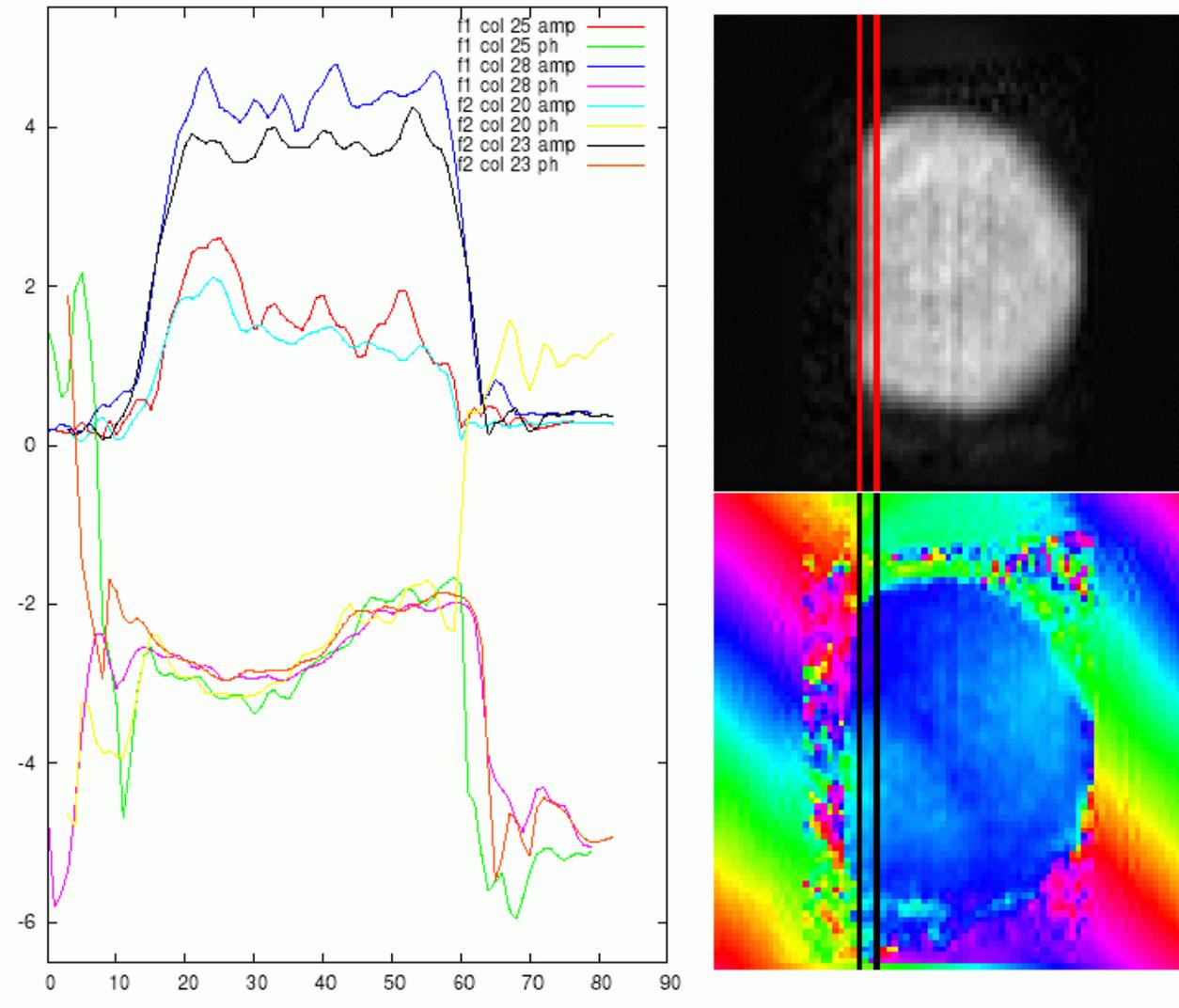
x-coordinate



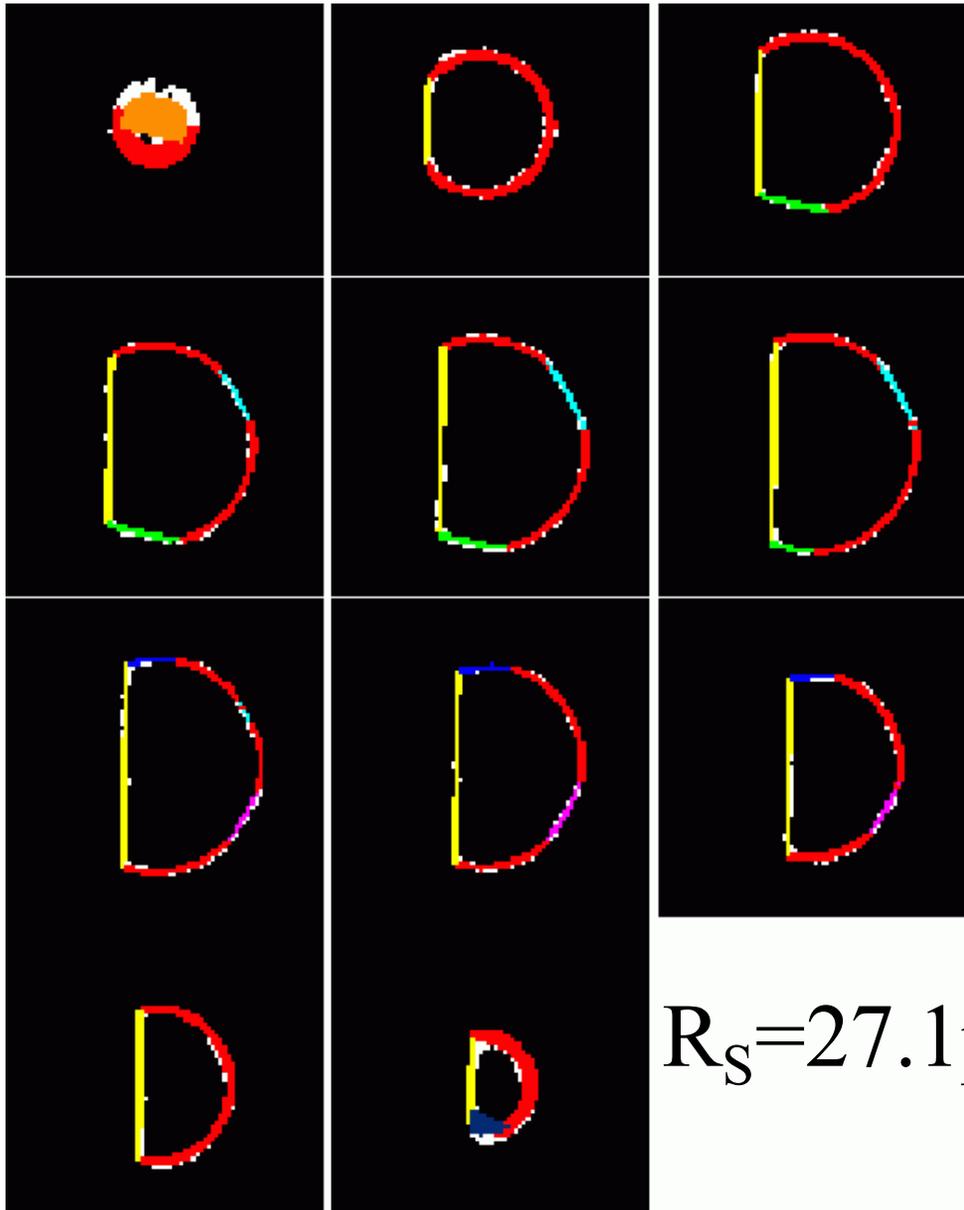
y-coordinate



Phase structure near substrate interface

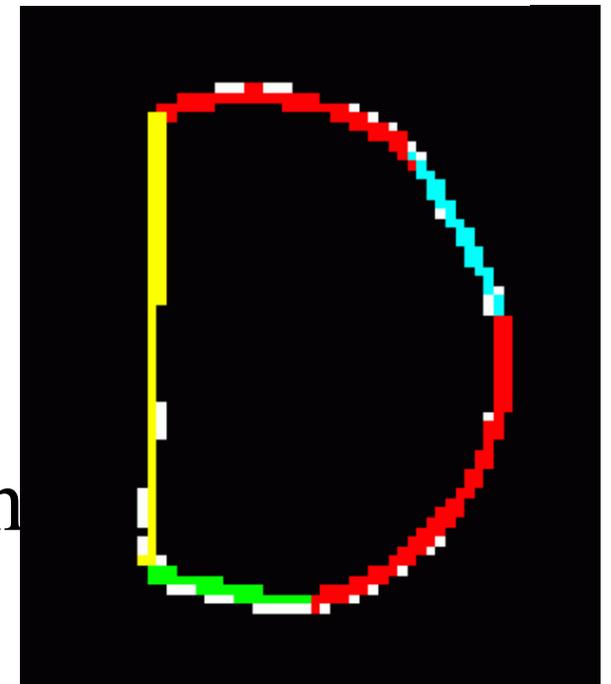


Fitting to faceted shape



	$ 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

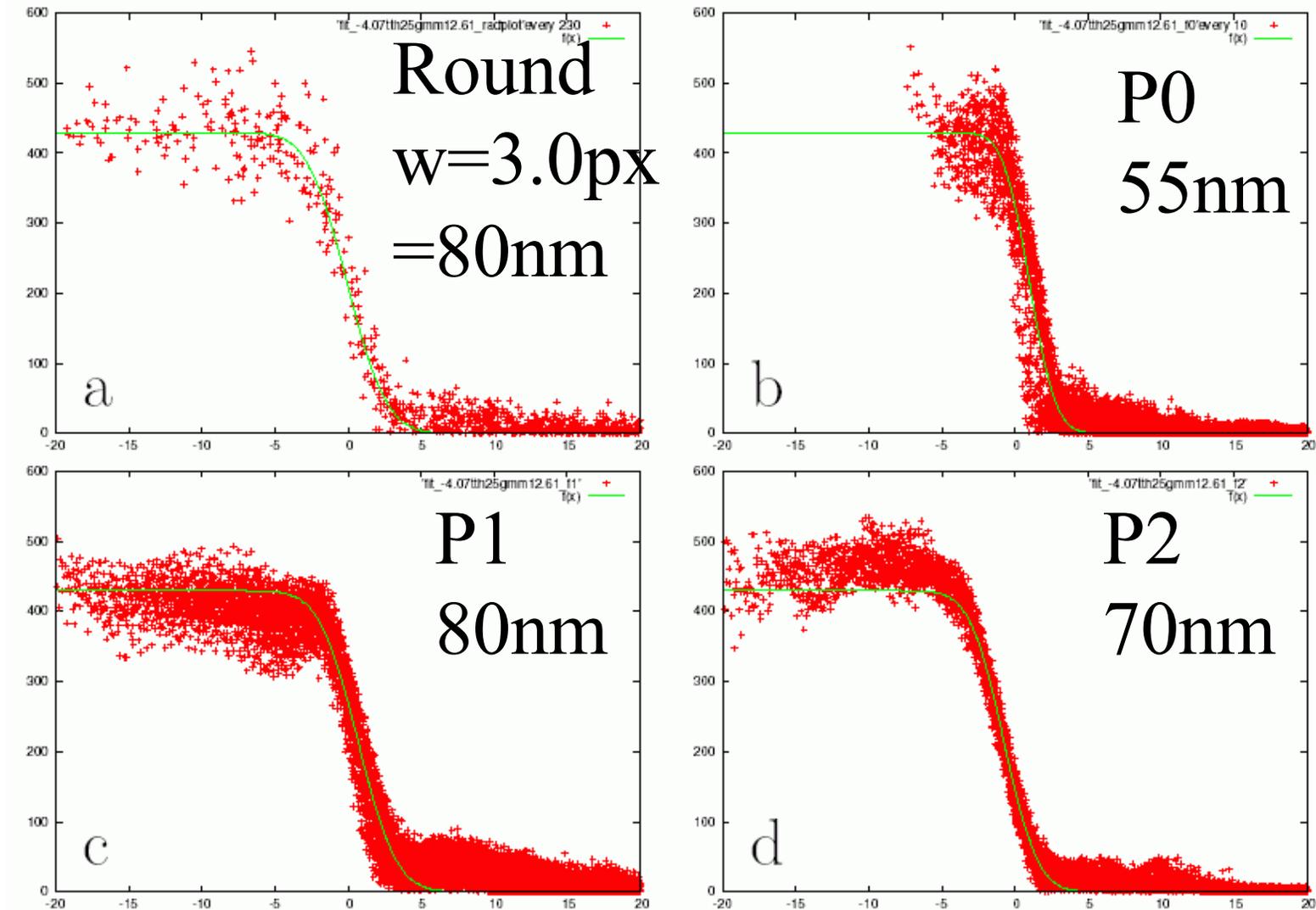
Angles between facets



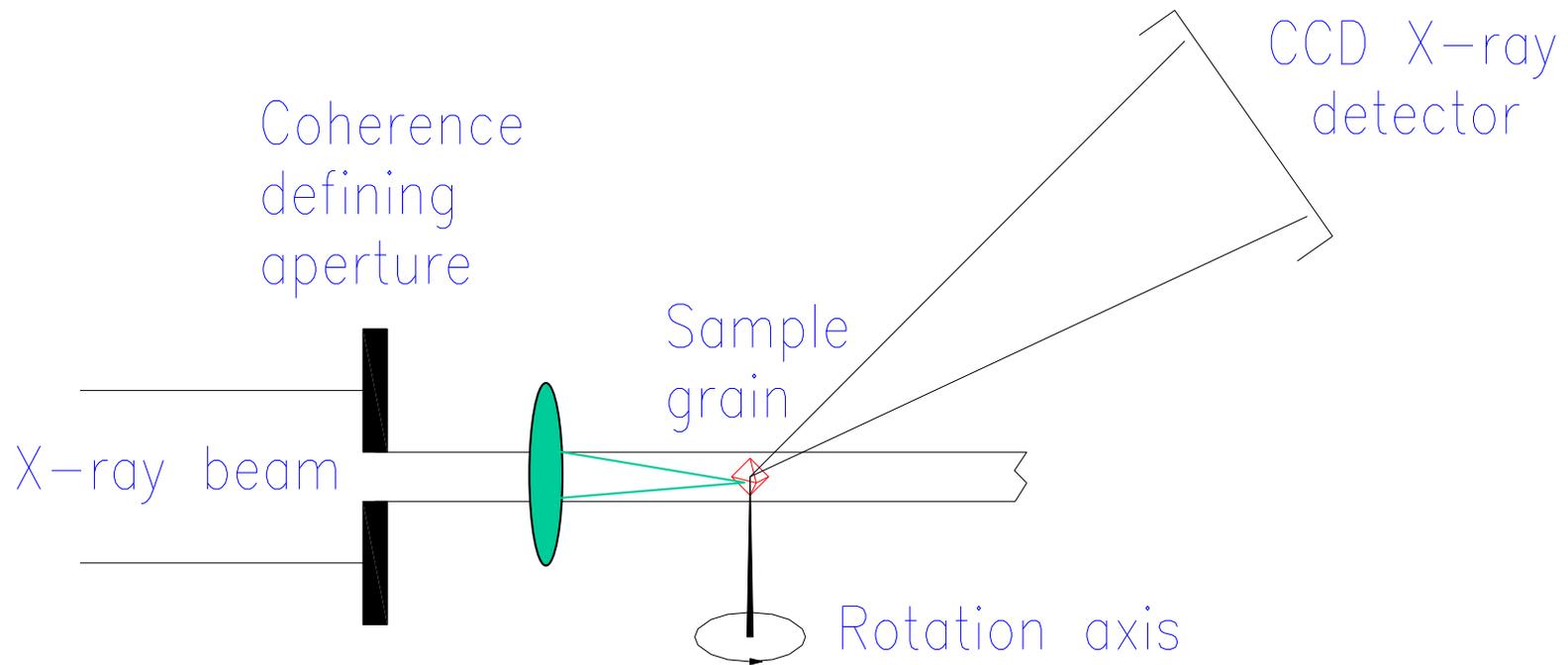
$$R_S = 27.1 \text{ px} = 724 \text{ nm}$$

PS Nanoprobe W/S

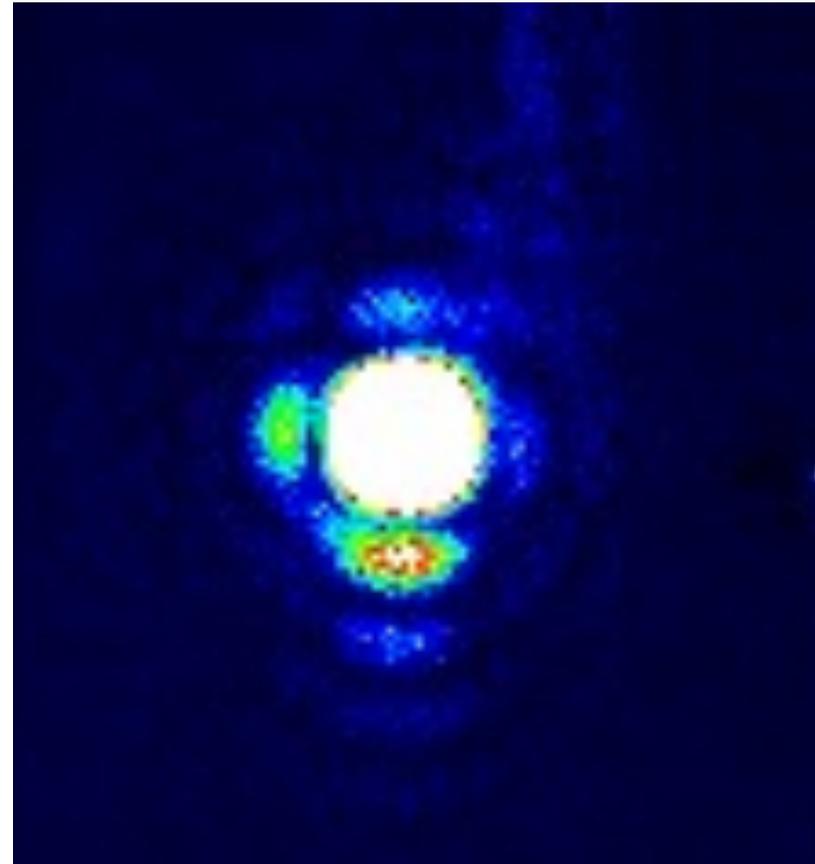
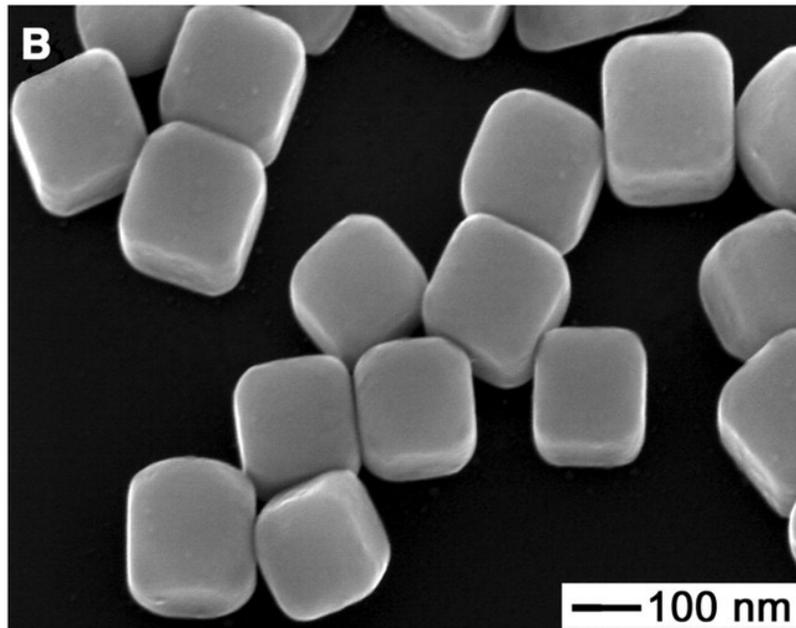
Density distribution across surface



Lensless X-ray Microscope

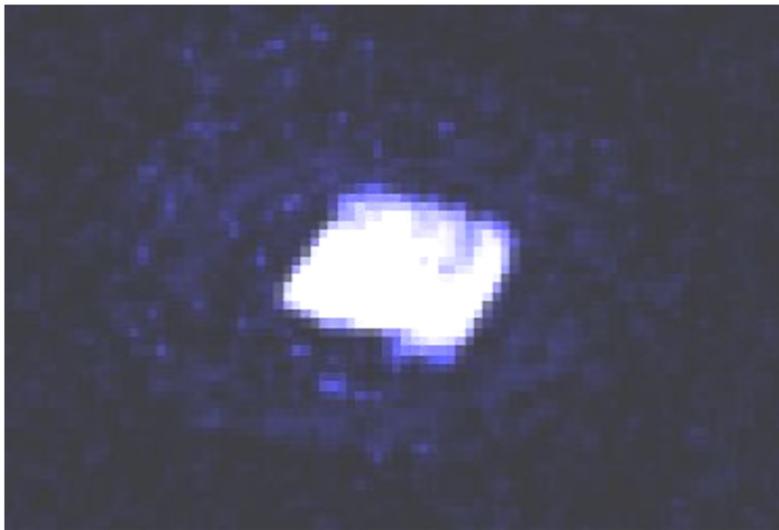


CXD from Silver Nanocubes

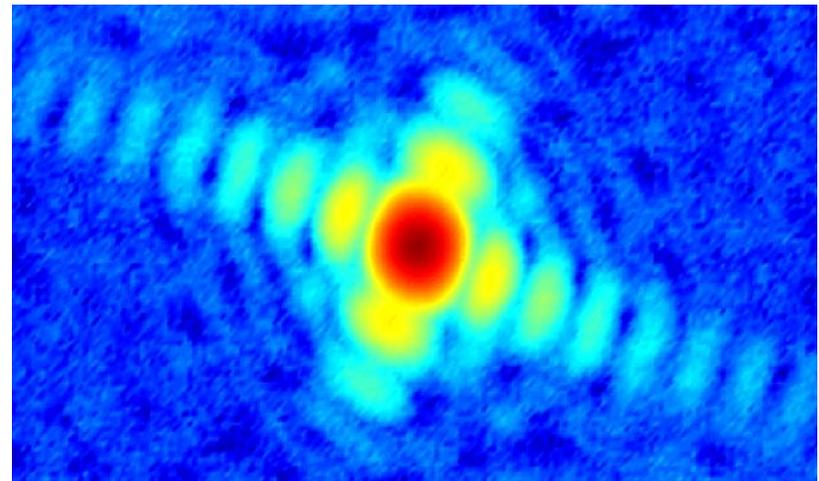
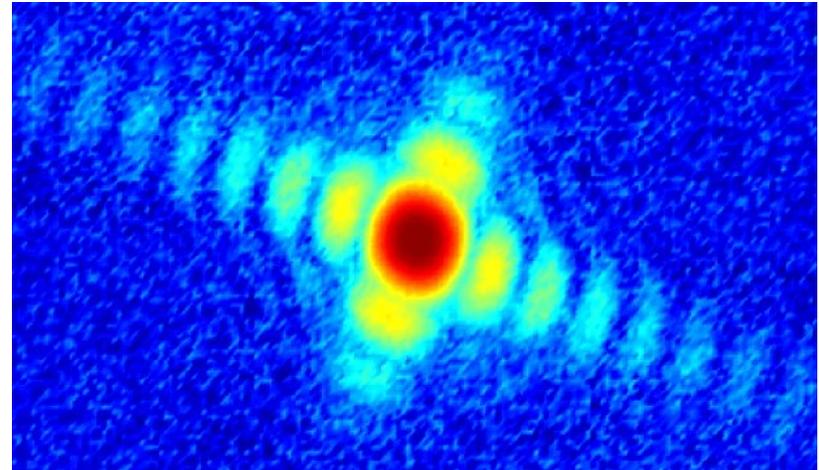


Yugang Sun and Younan Xia,
Science 298 2177 (2003)

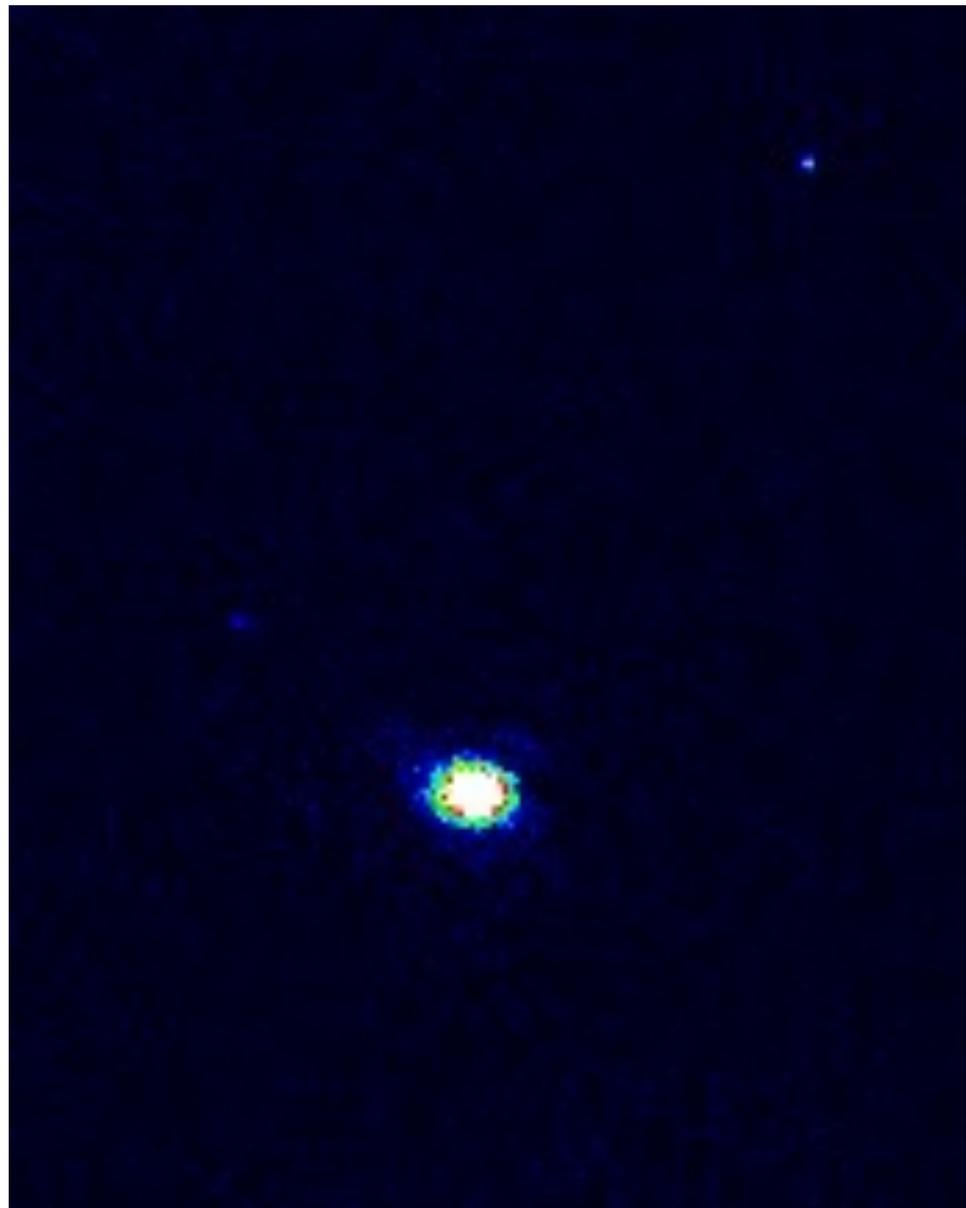
Reconstruction of Ag Nanocrystal



←→
200nm



Rocking
scan of Ag
cubes with
 0.01° steps



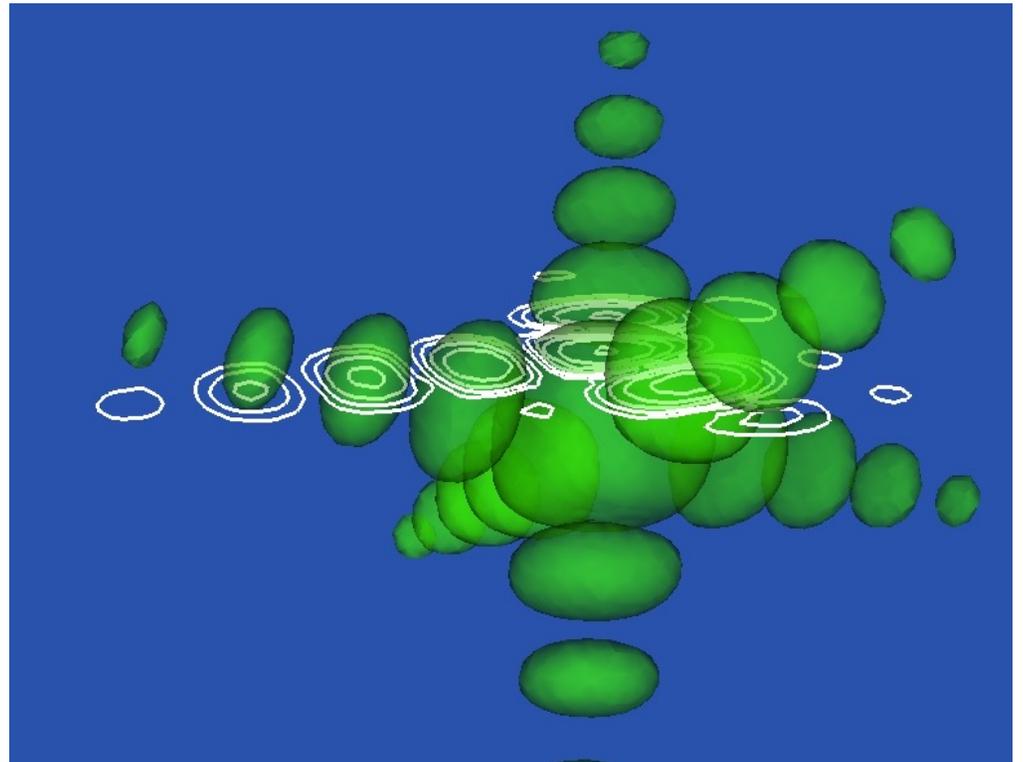
Miscentered Non-Centrosymmetric Reciprocal Space Slice



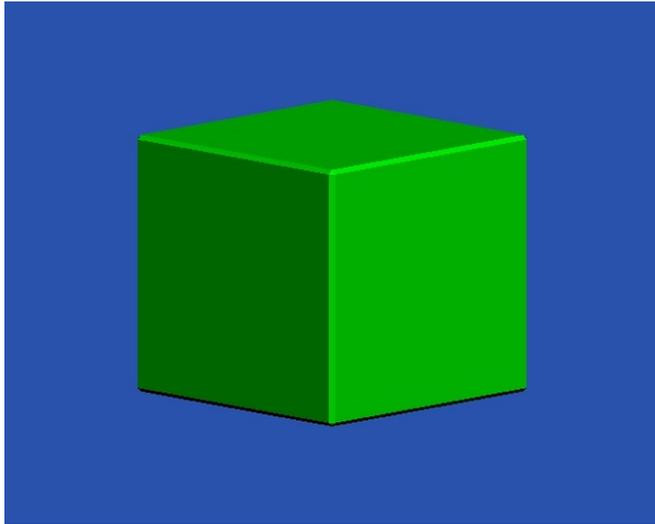
$$A_q \neq A_{\bar{q}}^*$$



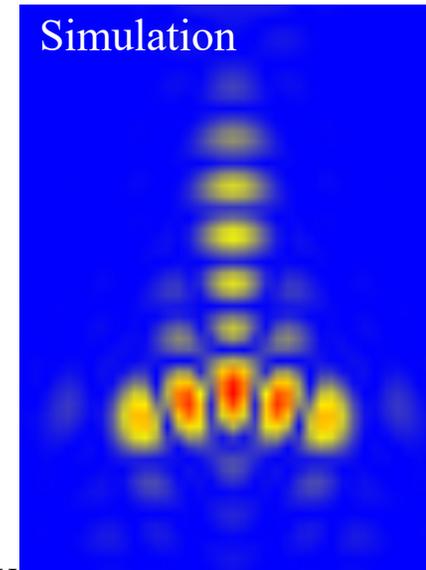
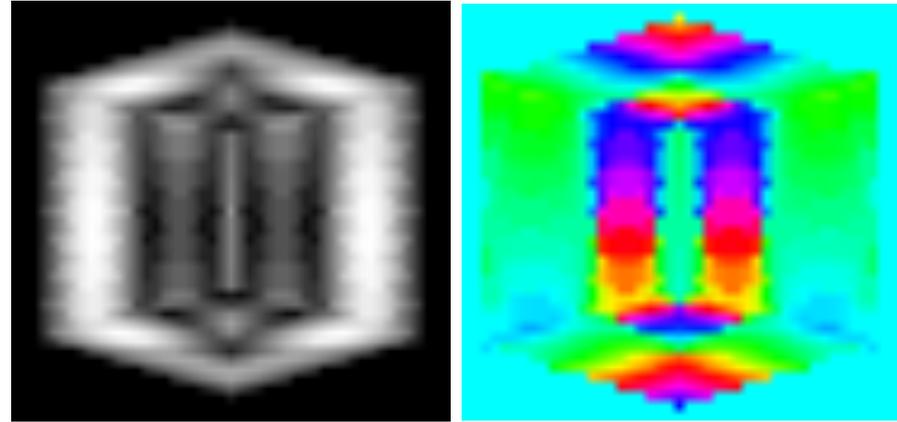
Real-space phase.



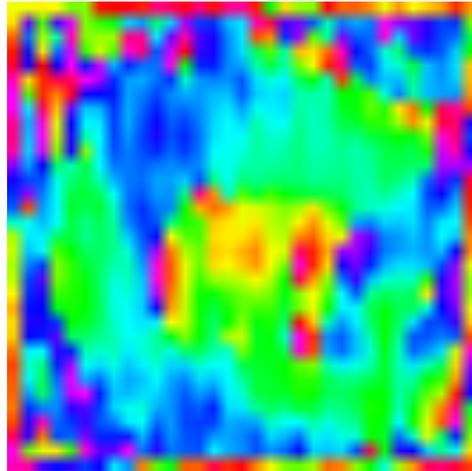
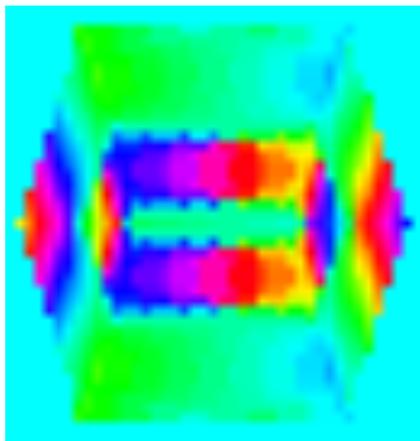
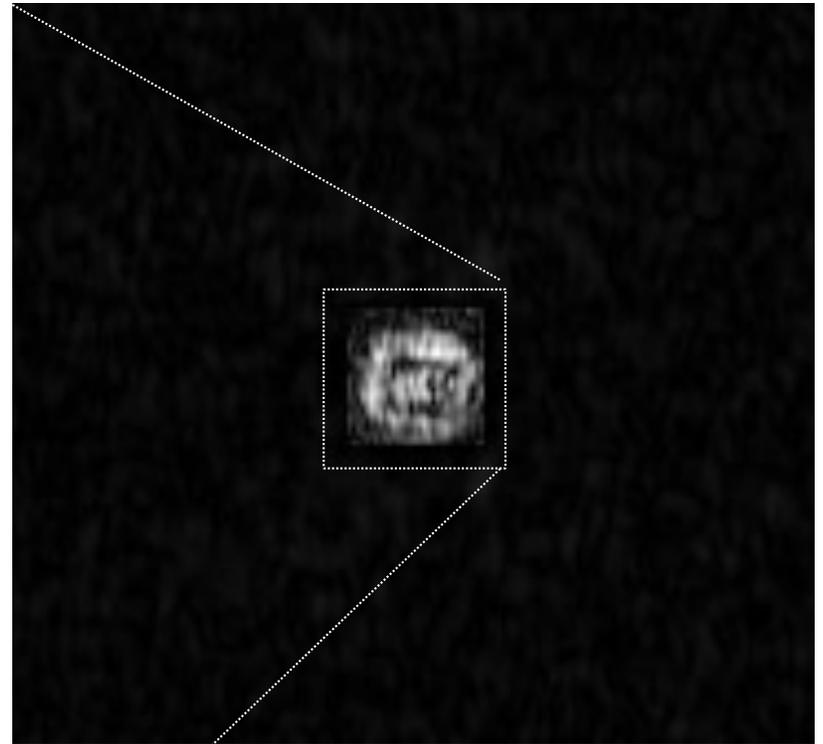
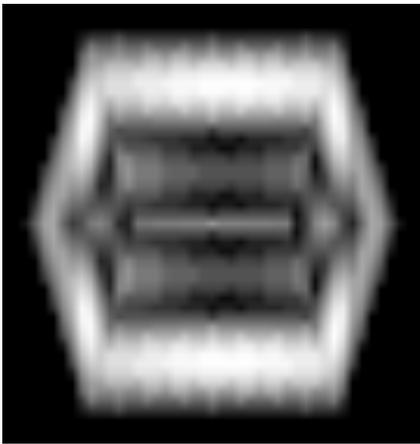
What can we expect to see?



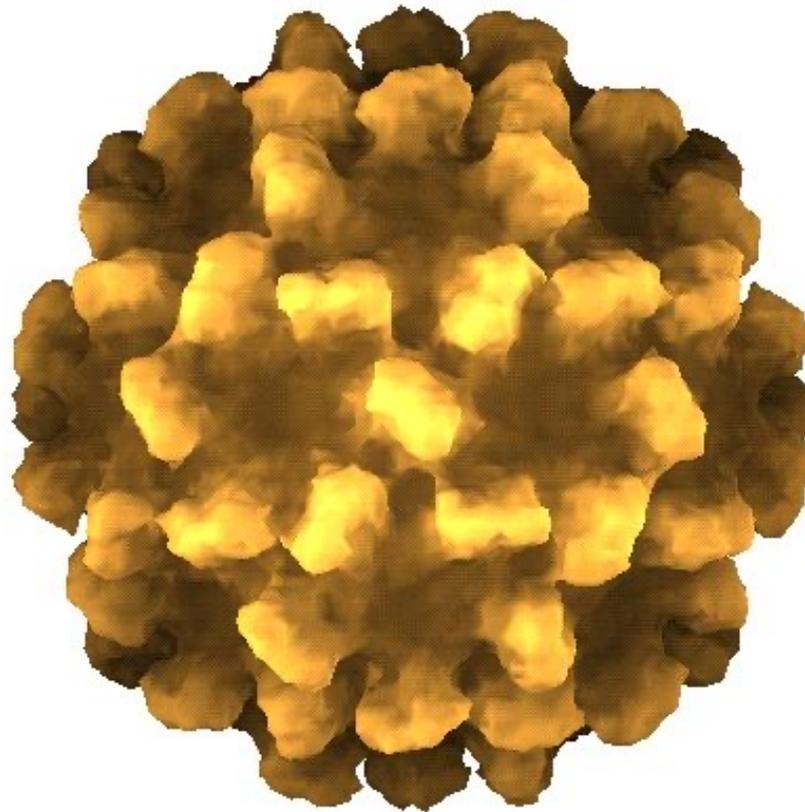
$$A(\bar{q}_o + \bar{q}_z) = \sum_i \underbrace{\sum_z \rho(x, y, z)}_{u_i(x, y)} e^{2\pi i \bar{q}_z \cdot \bar{z}_i} e^{2\pi i (\bar{q}_o) \cdot \bar{r}_i}$$



Direct Space Result (1000 Iterations)



Tomato Bushy Stunt Virus 1980



Conclusions and Outlook

- Inversion of CXD demonstrated
- Internal structure of Au Nanocrystals
- Phase structure visible in Pb Nanocrystals
- Off-center diffraction gives complex image
- Single molecules one day