# Phase Retrieval and Applications


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

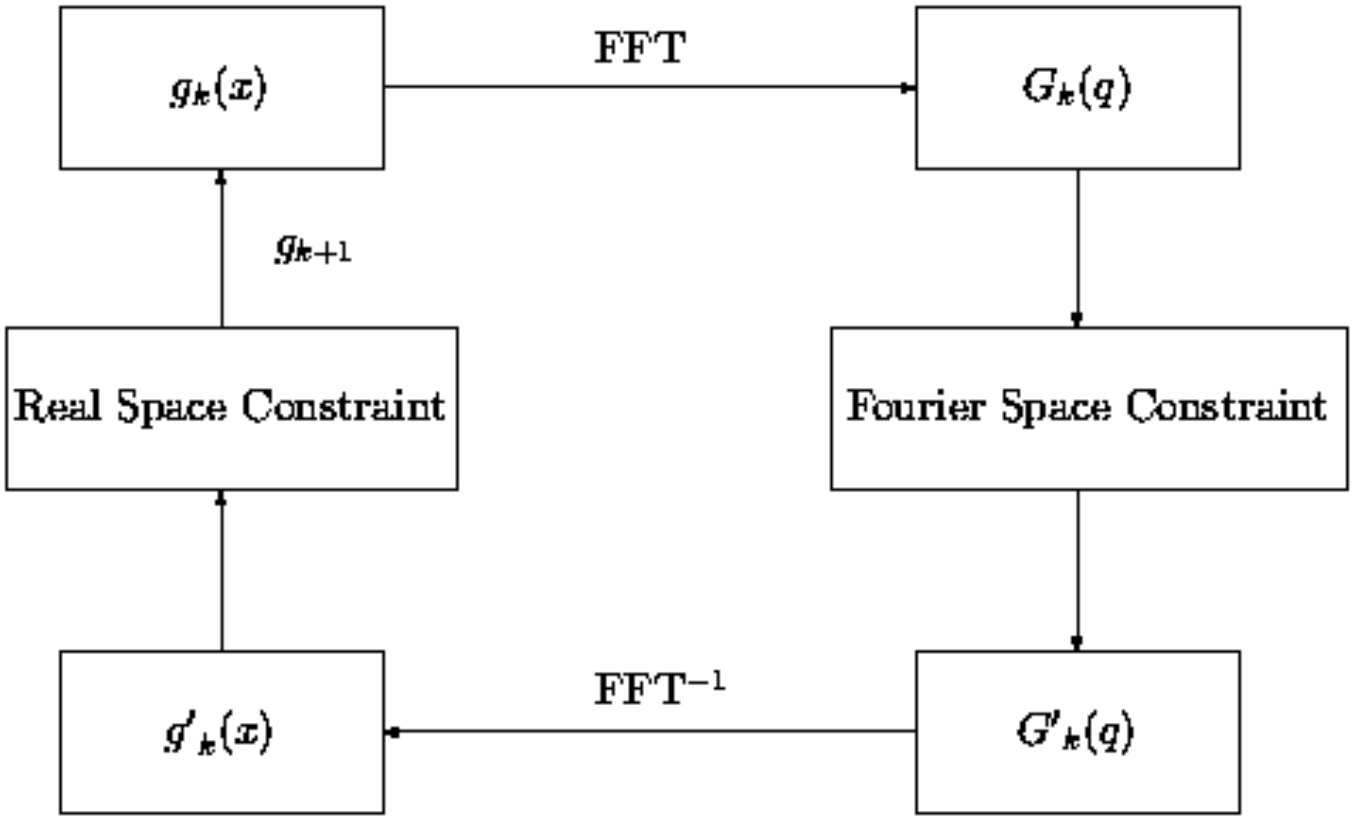
Department of Physics
University of Illinois


Euro-XFEL Workshop
Oct 28, 2005

# Outline

- Coherence + Phase Retrieval for Structure
- The Phase Problem
- Nanocrystal Shapes
- Imaging of Phase Objects
- Extrapolation and Technical Issues for XFEL
- “First” Euro-XFEL Experiment

# Generic "Error Reduction" method

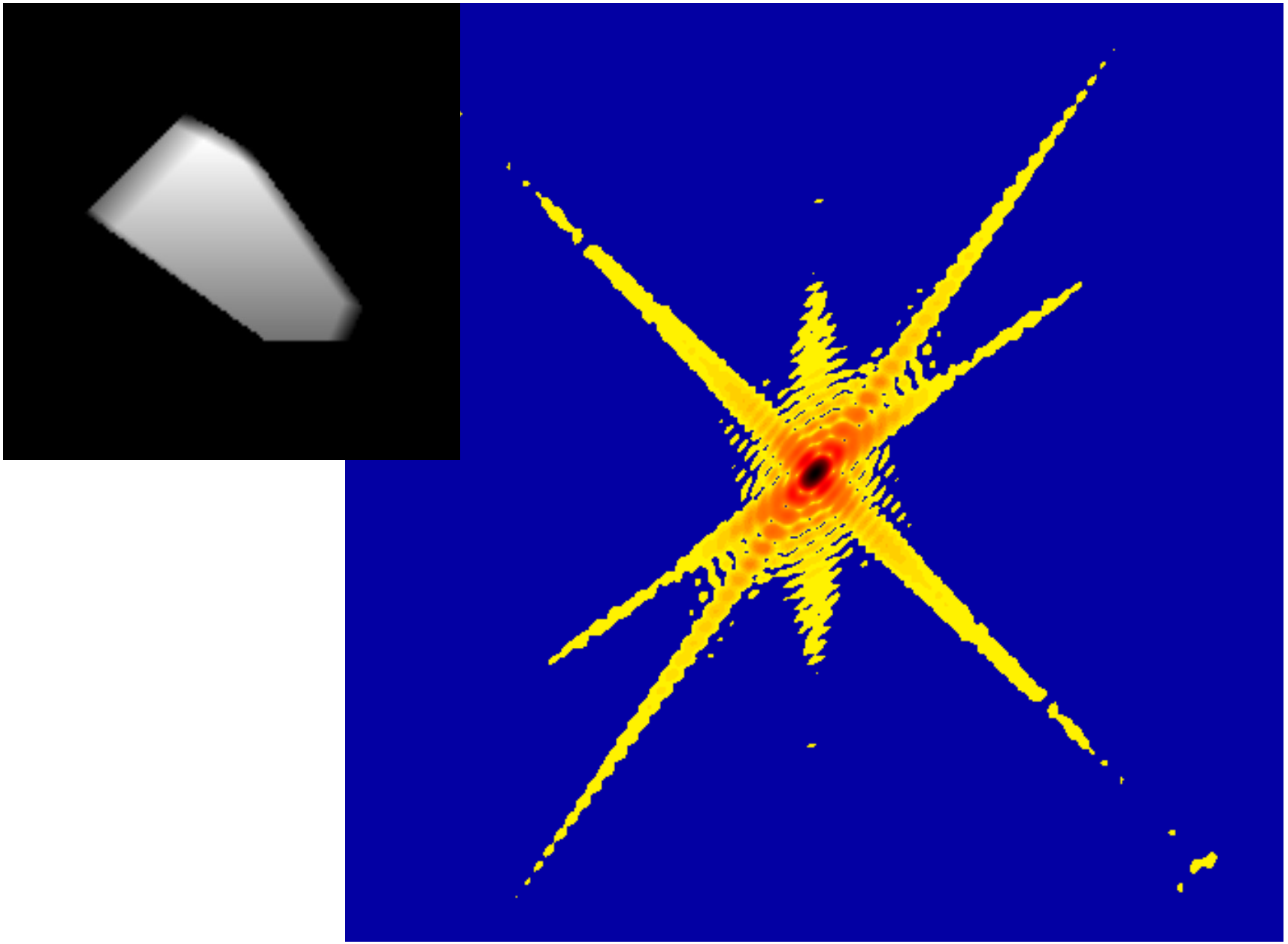



J. R. Fienup Appl. Opt. 21 2758 (1982)
R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)

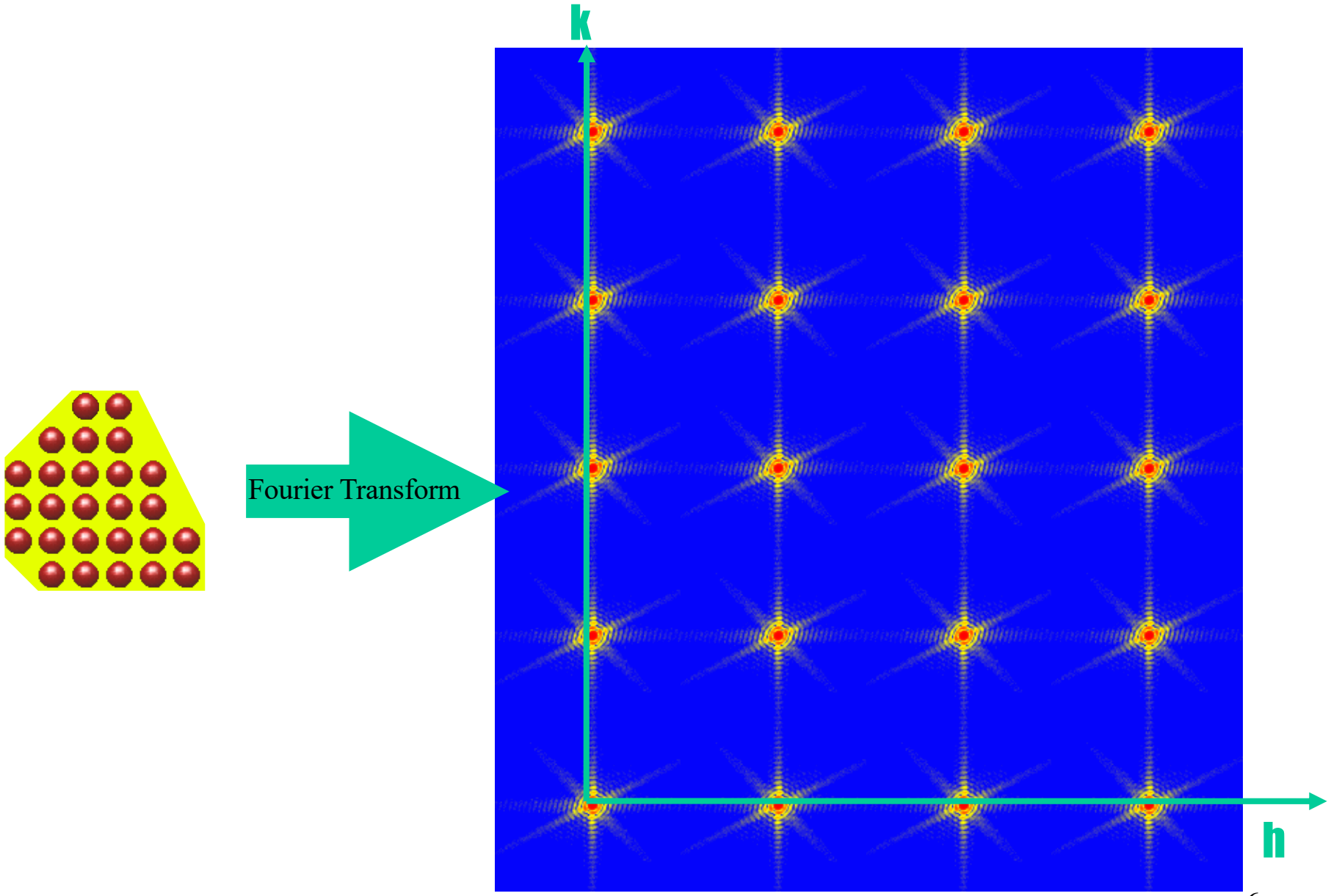
# Real-space Constraints in Crystallograhy

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)

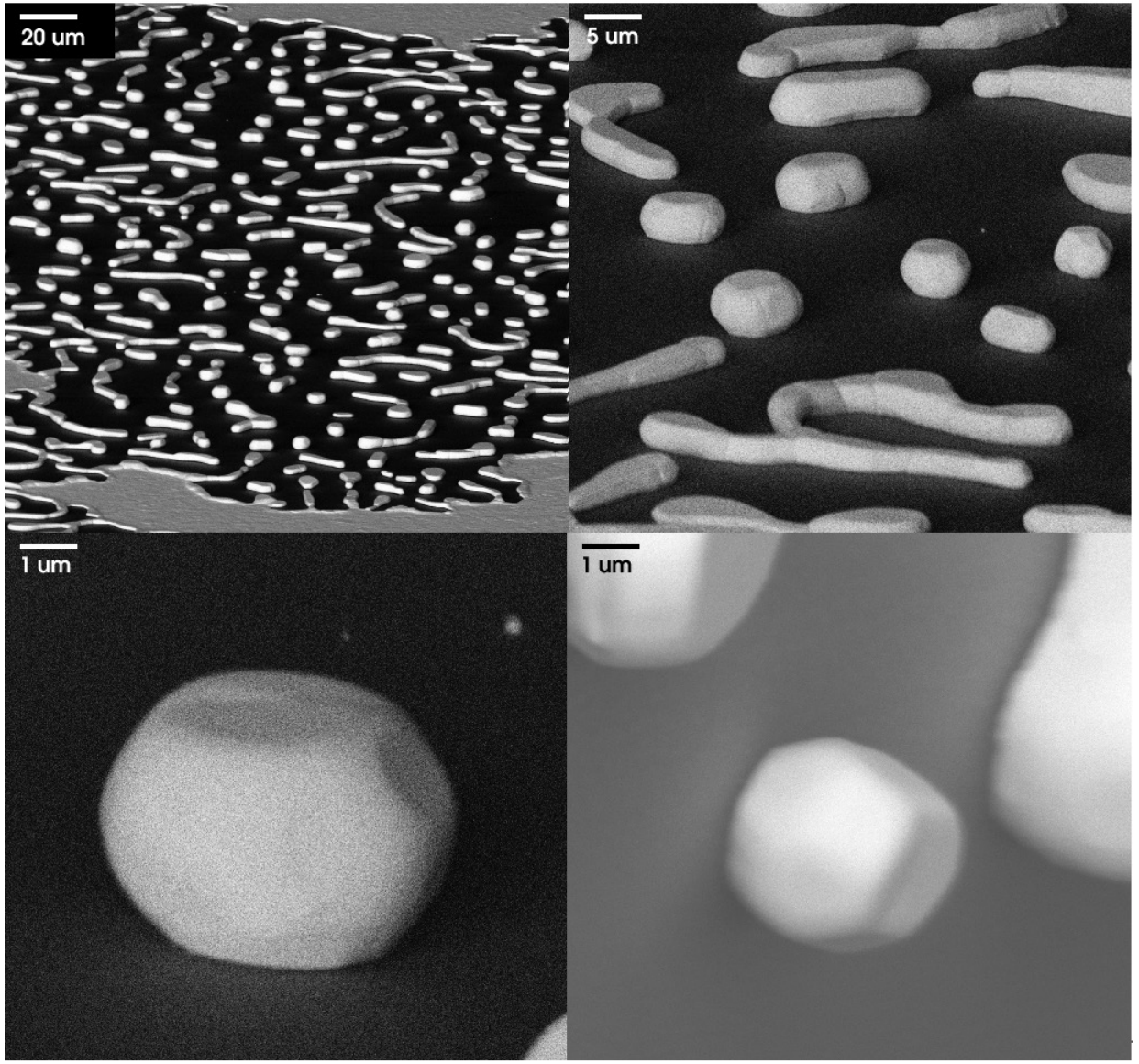


# Coherent Diffraction from Crystals



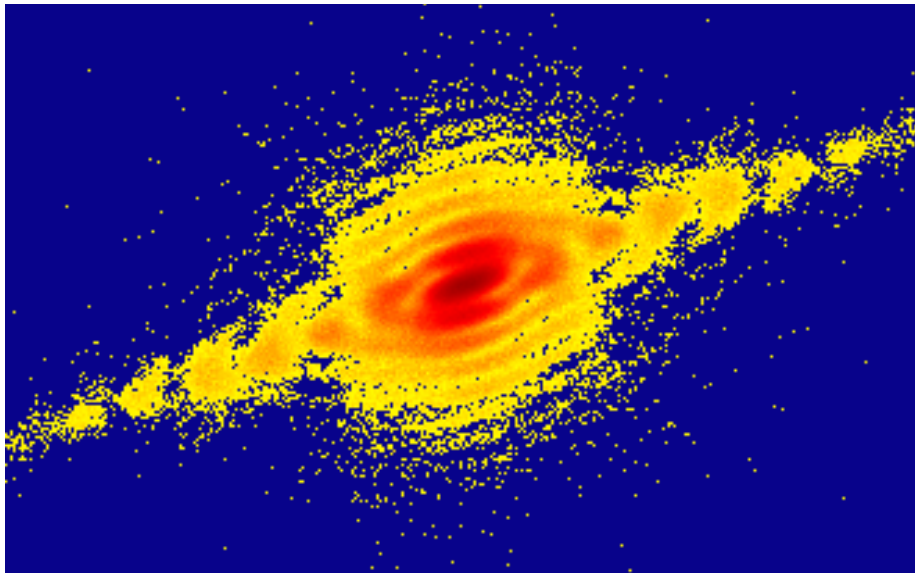
# SEMS

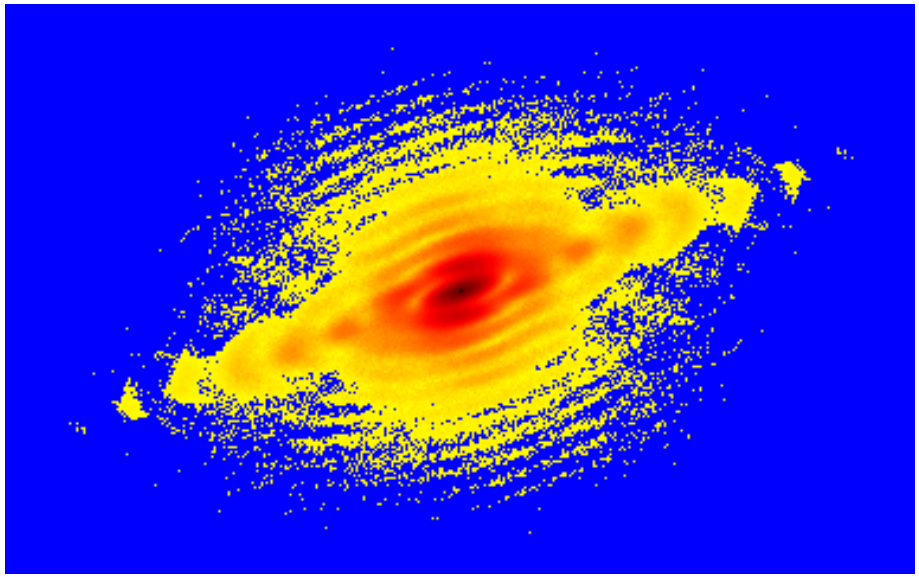
- Au blanket film
- Quartz substrate
- Annealed at 950°C for 70 hrs.

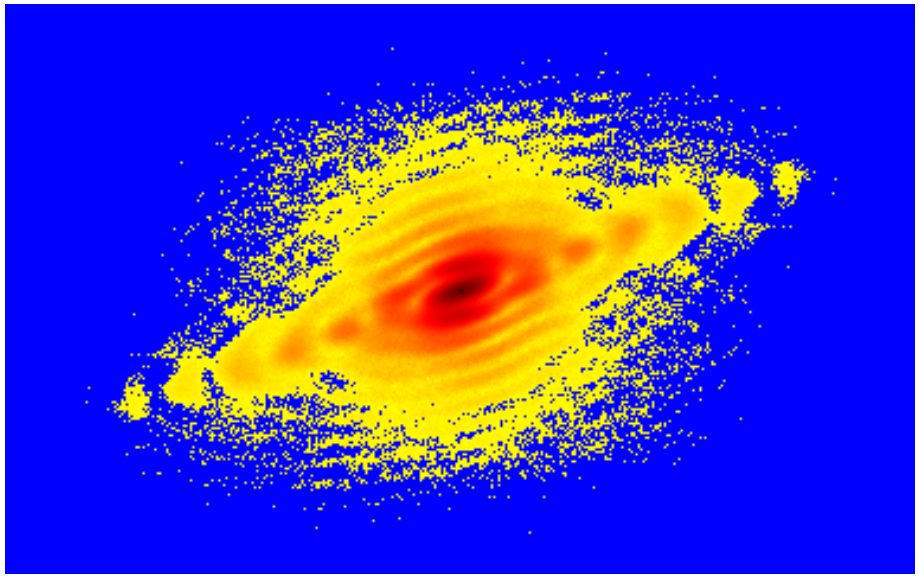


# Symmetrized Data and two best fits

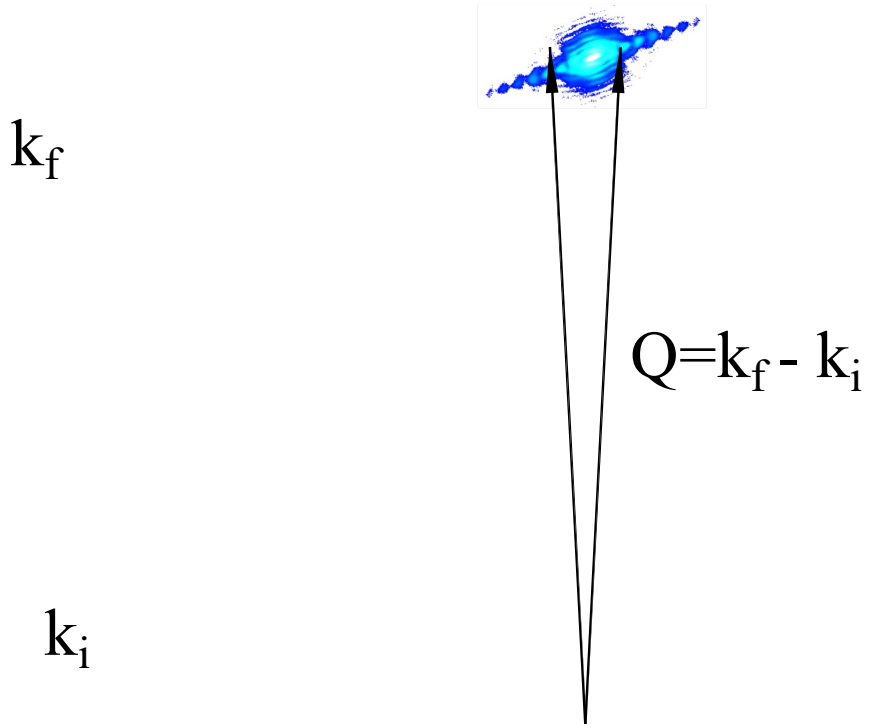
Chisq=0.0005

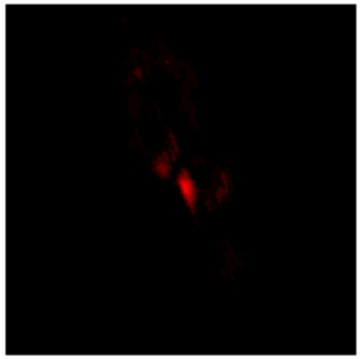
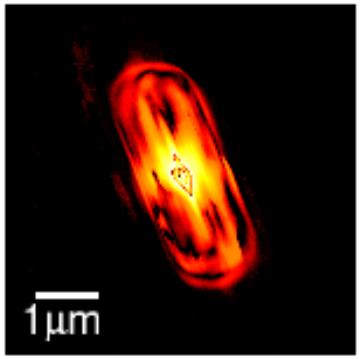


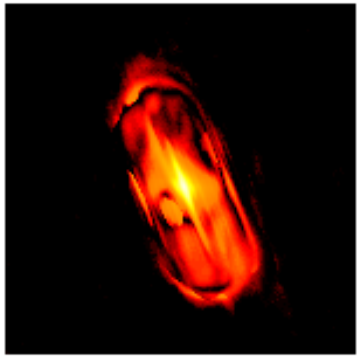
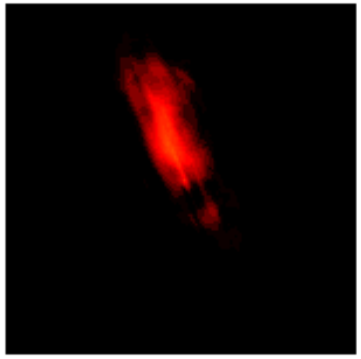
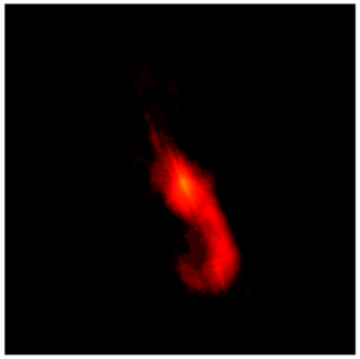
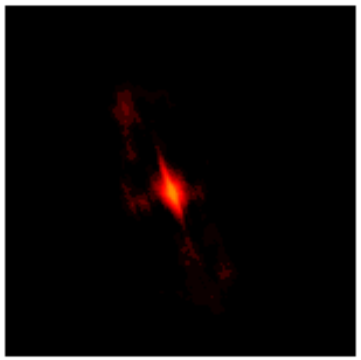
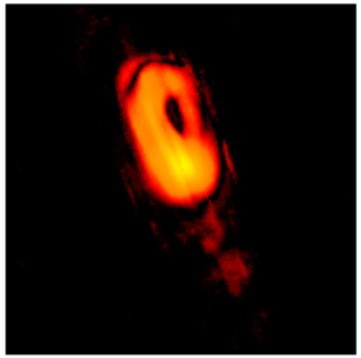
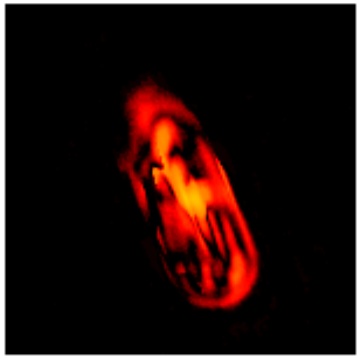
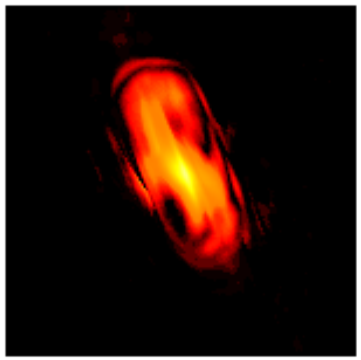
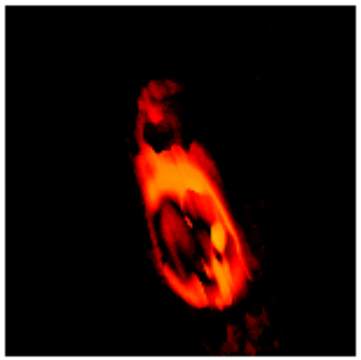
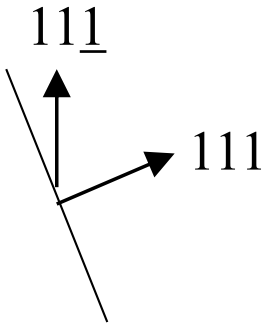




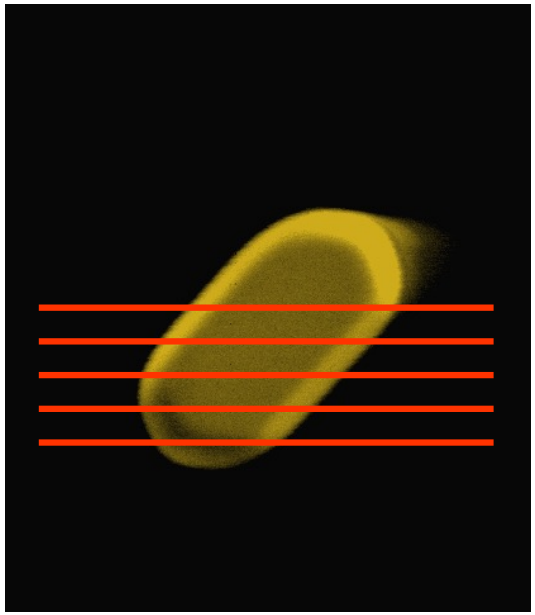
# 3D Diffraction Method



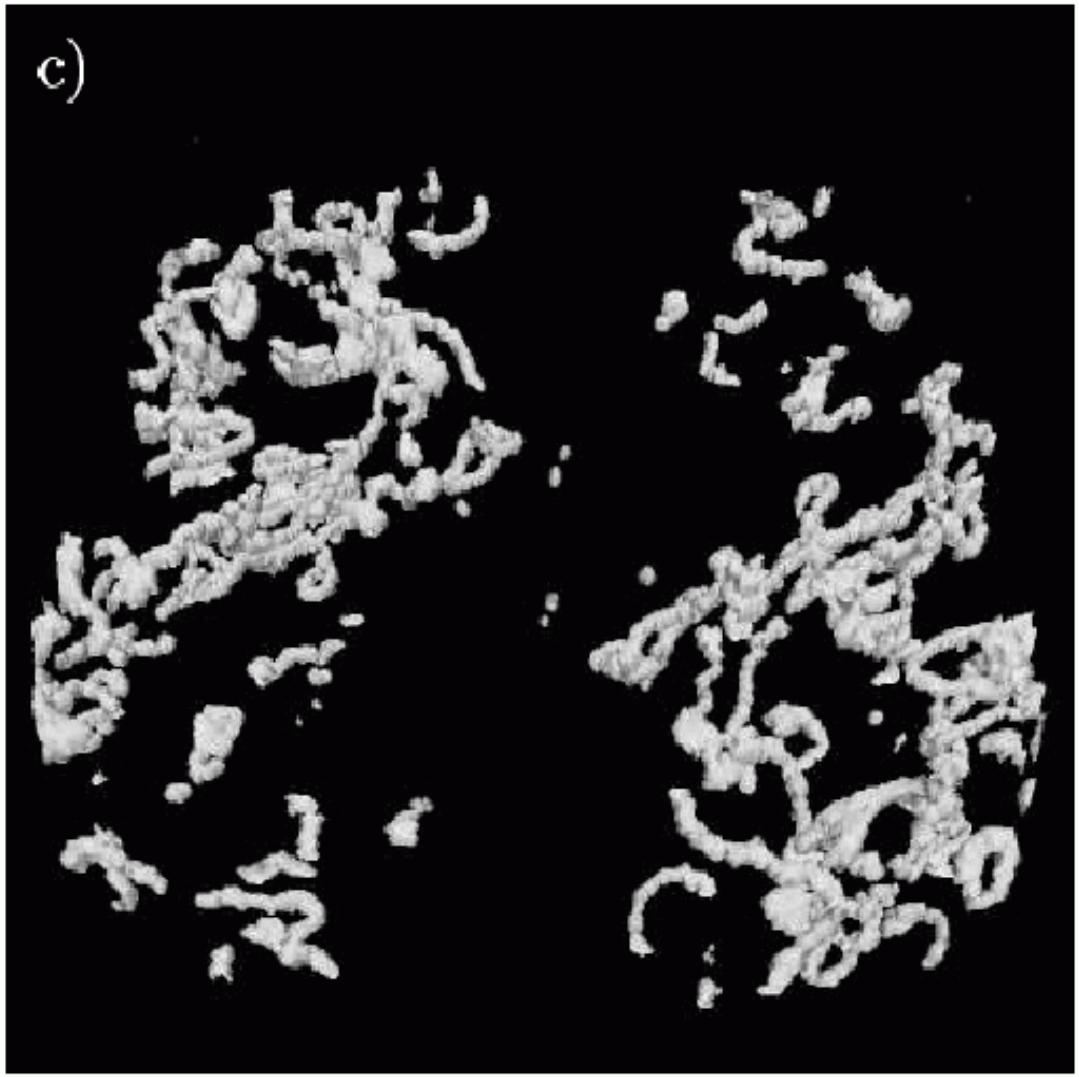



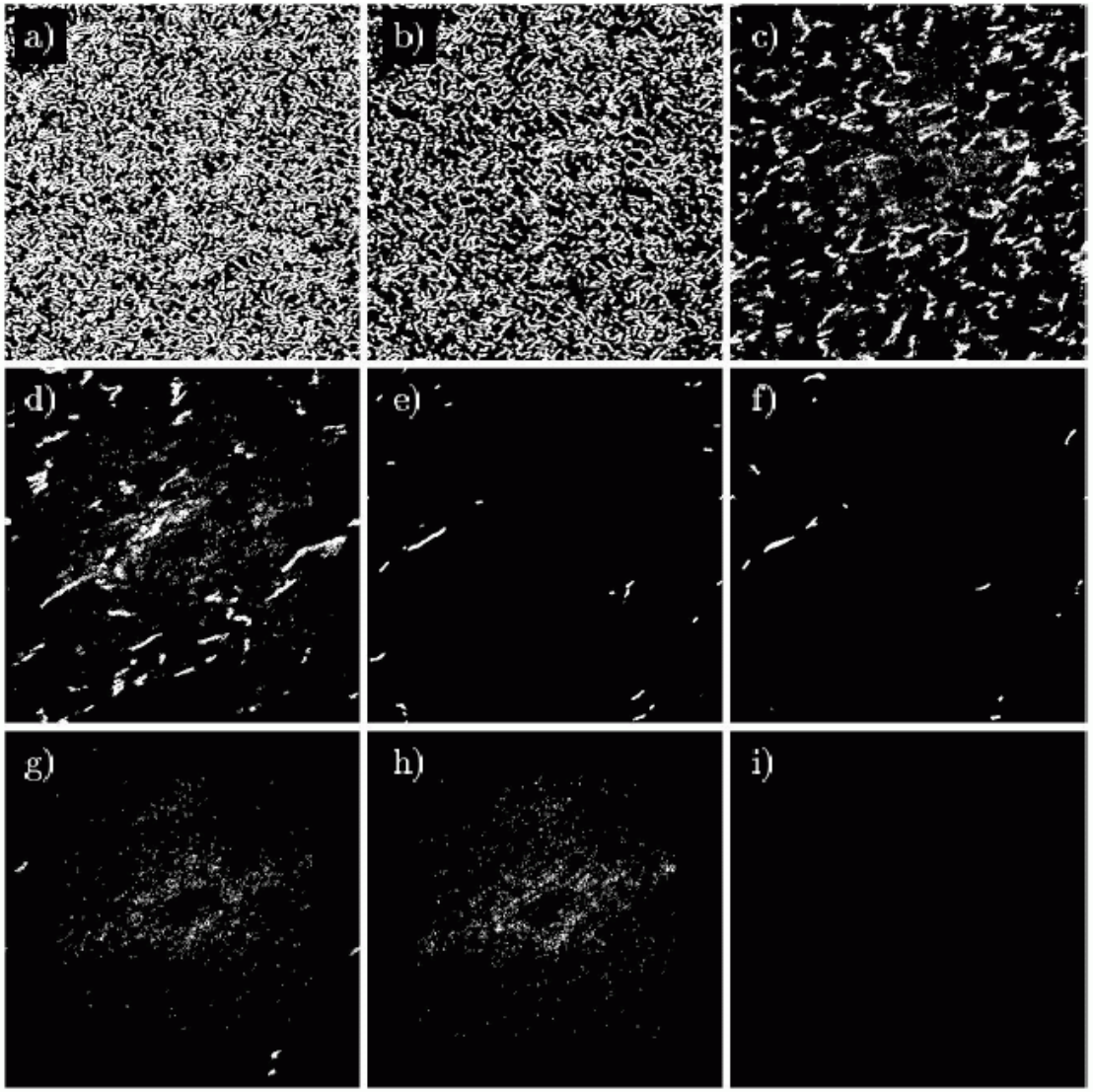

Slices through plan view SEM:



# ‘Vortices’ Form Loops in 3D

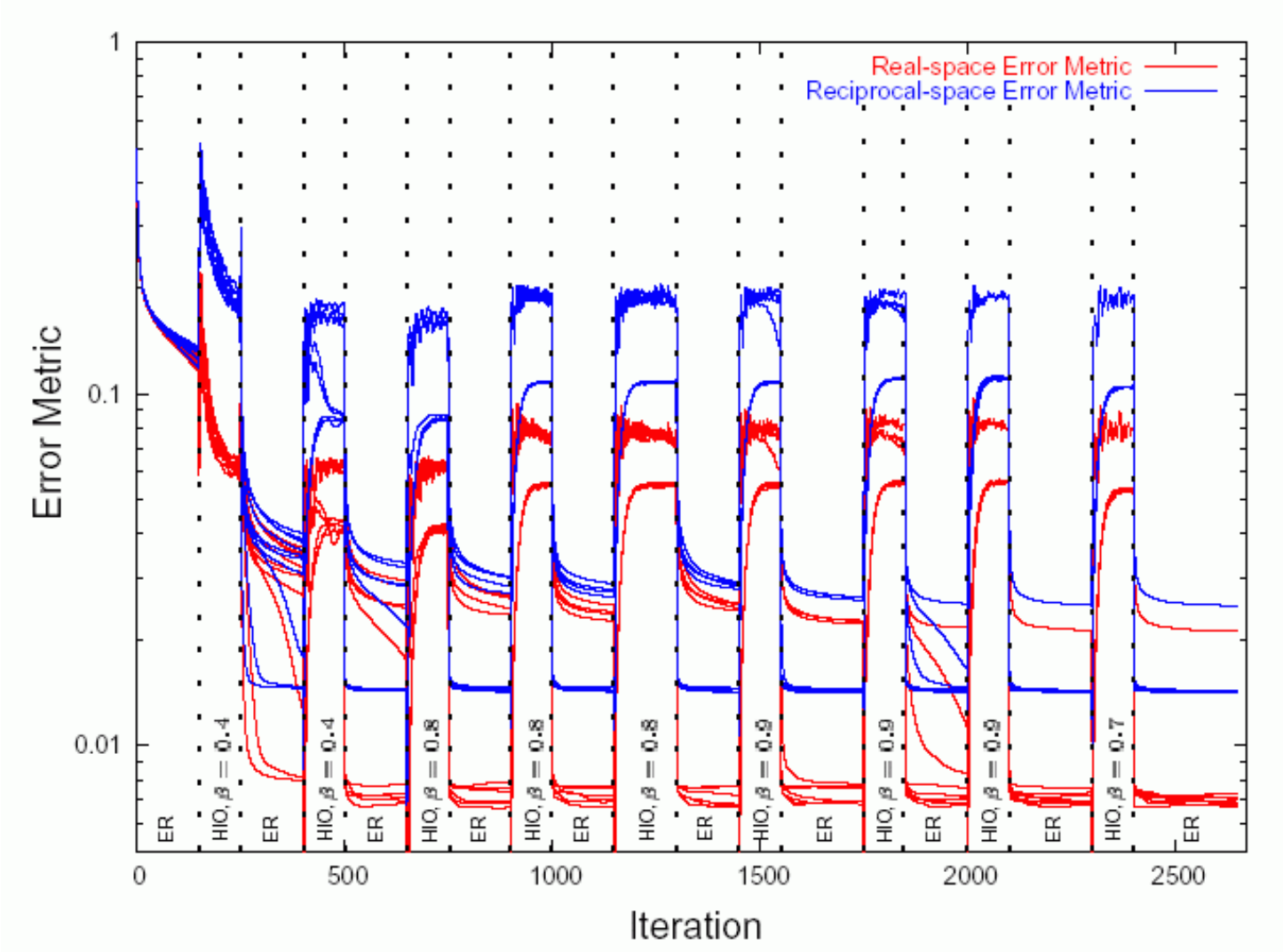


# Clearing of Vortices during HIO

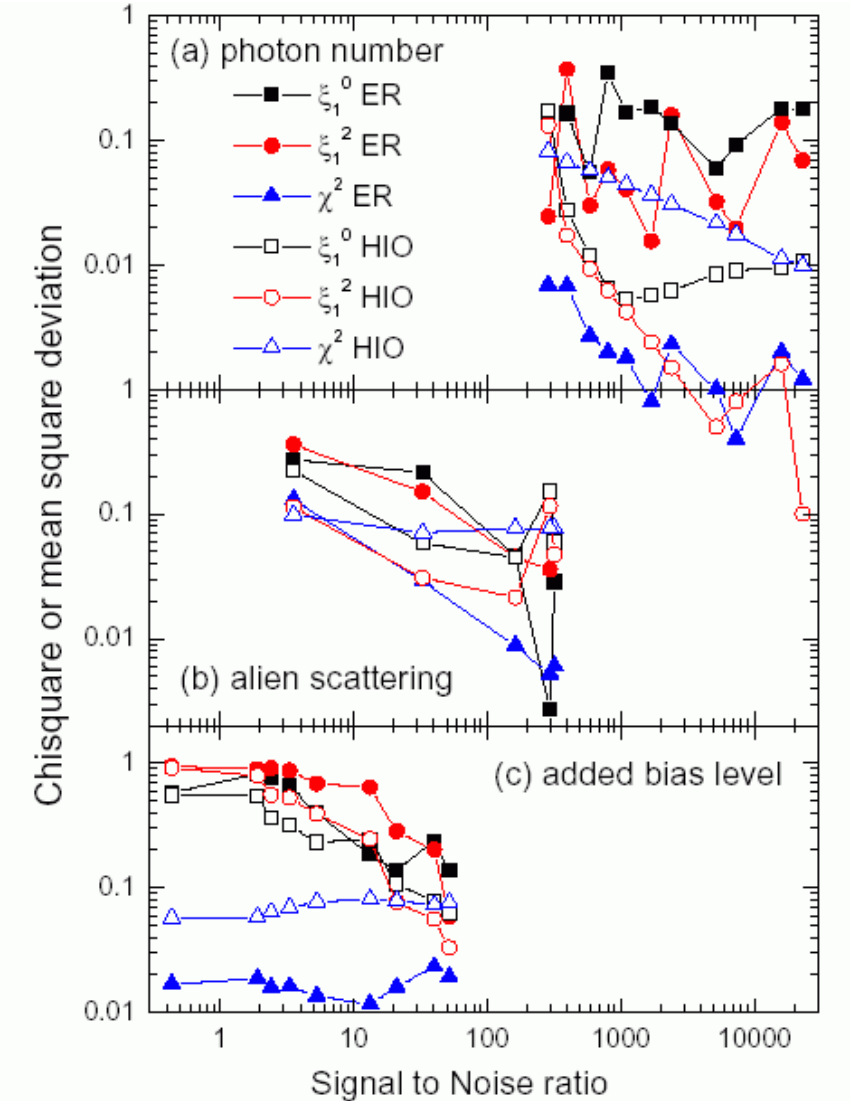


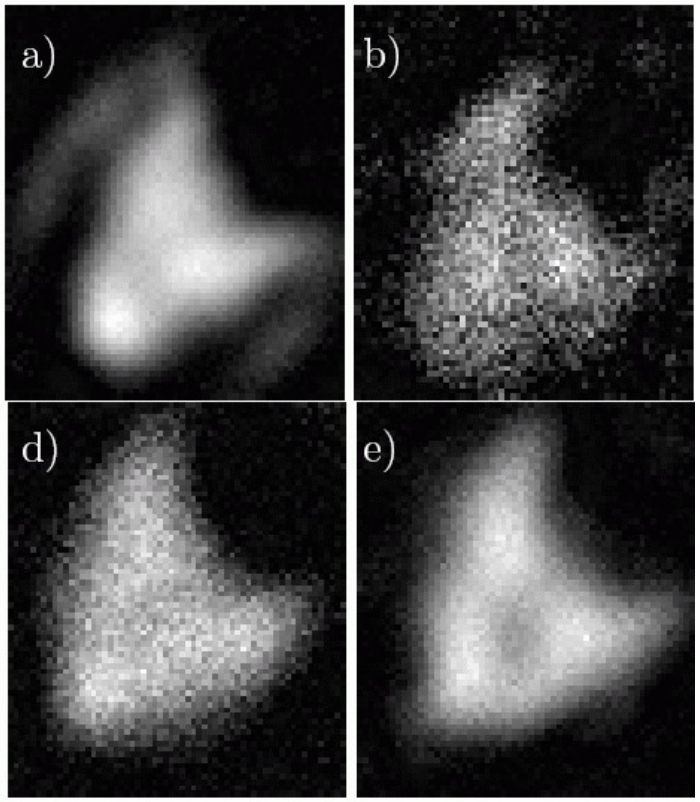

ER ER HIO
HIO ER ER
HIO HIO ER

# Progress of Phase Retrieval

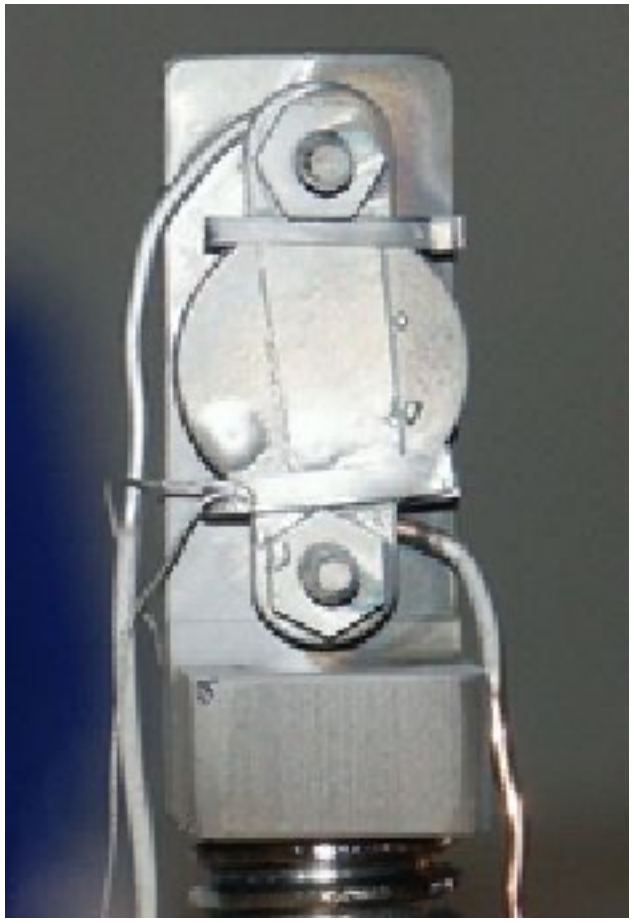


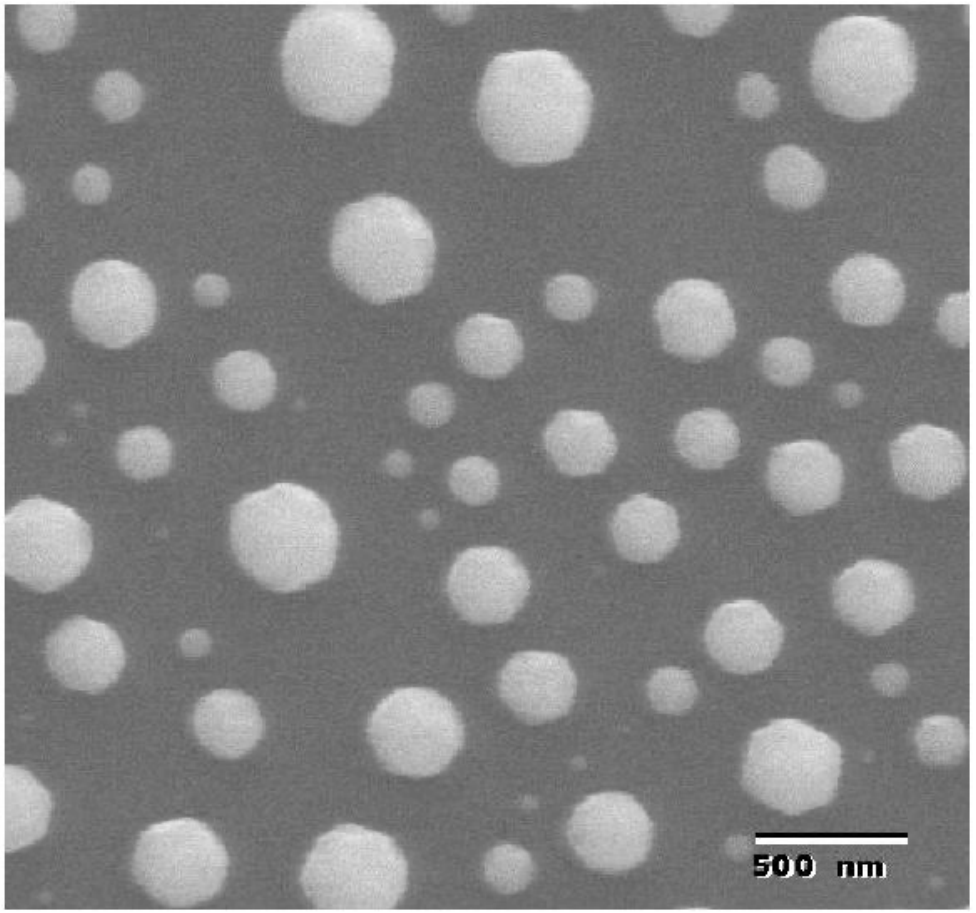
# Study of Convergence with Noise



# In situ growth of Pb crystals





# Good statistics, 3D diffraction data

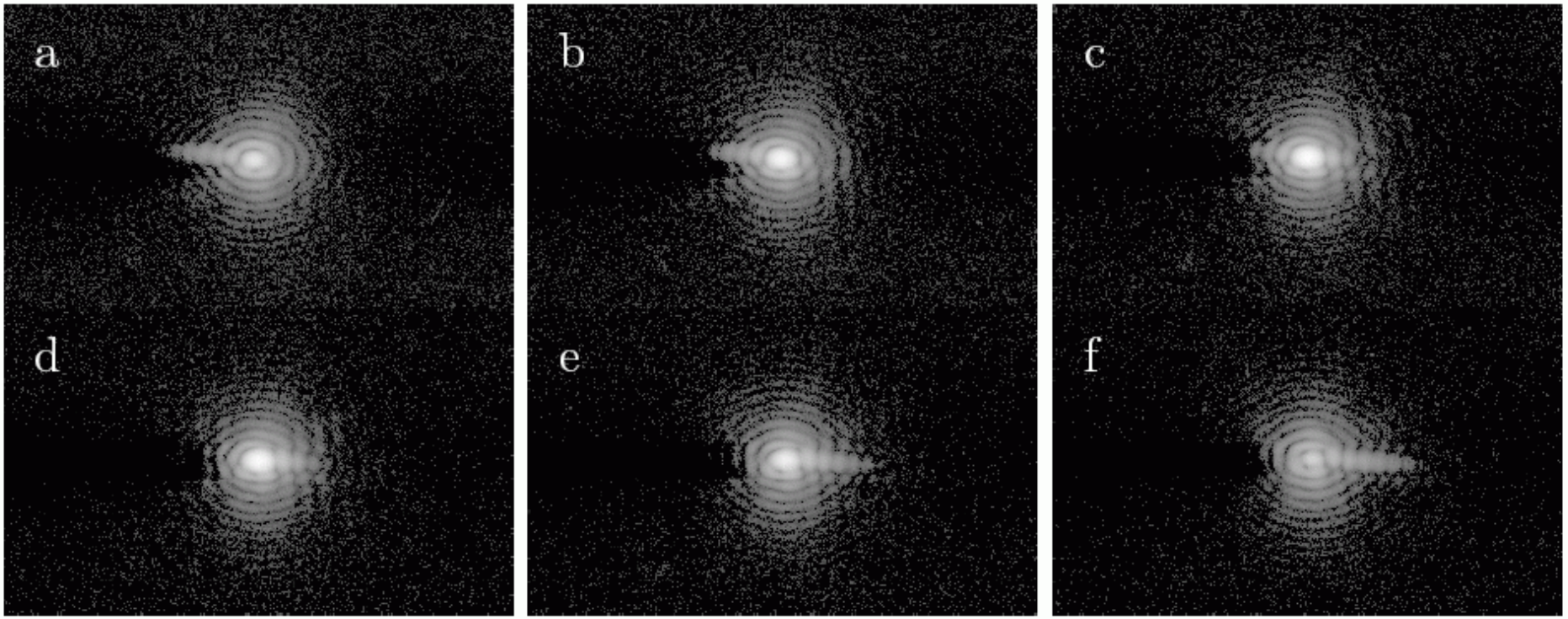
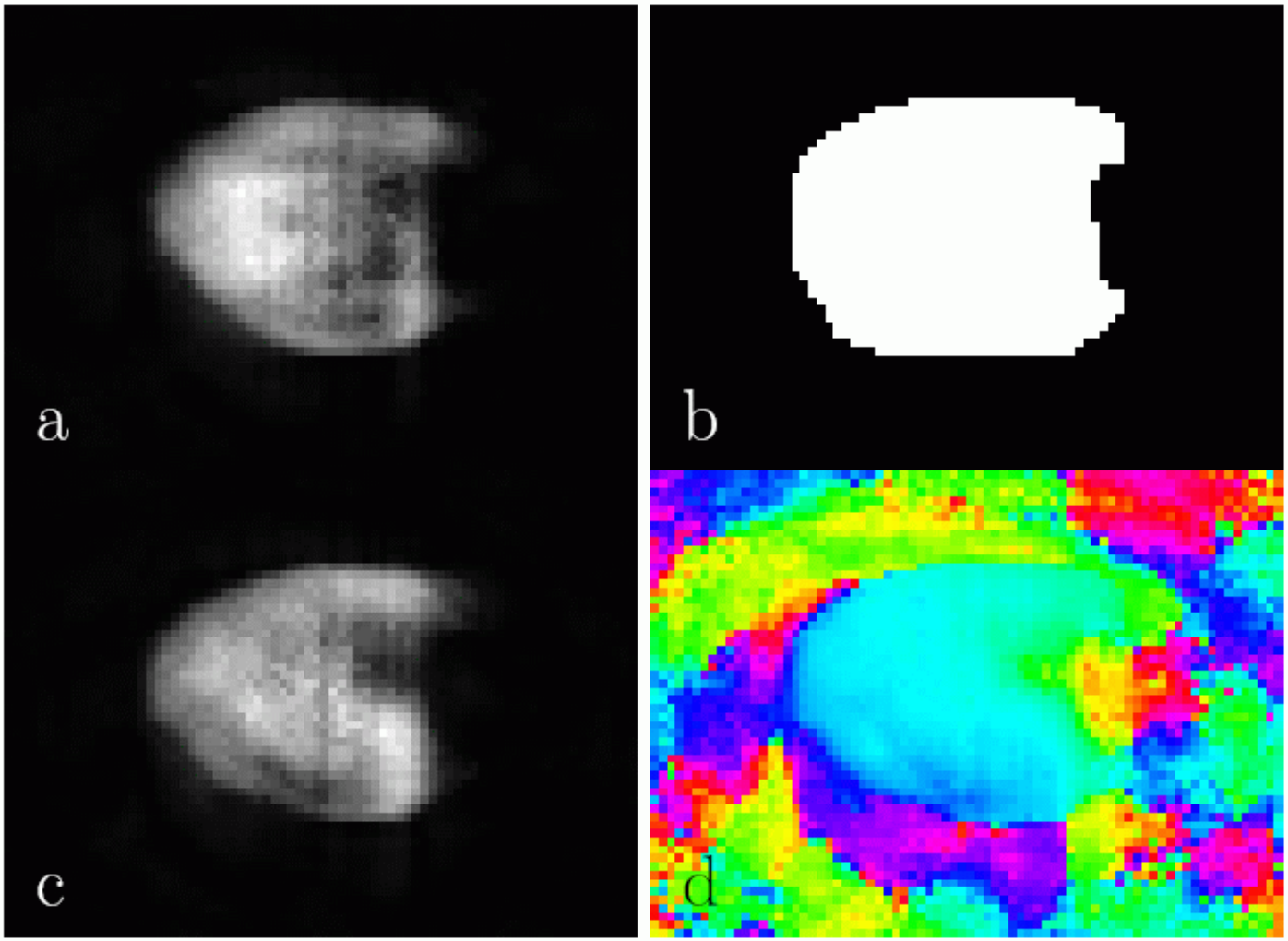
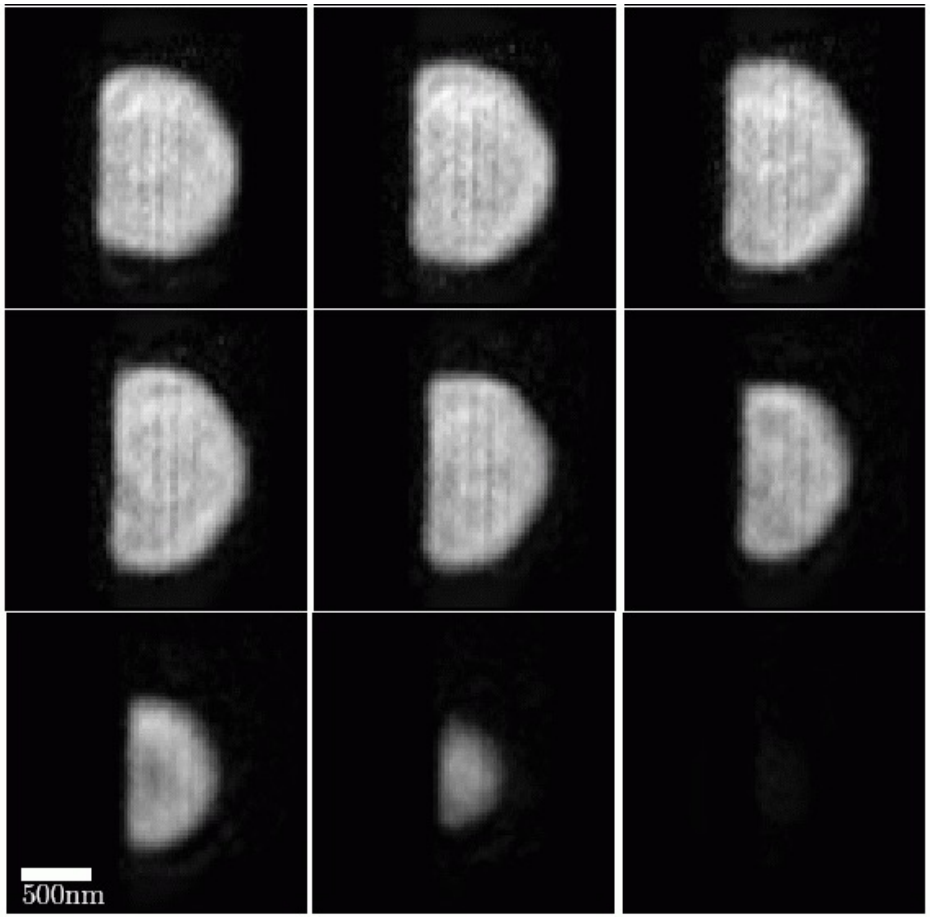


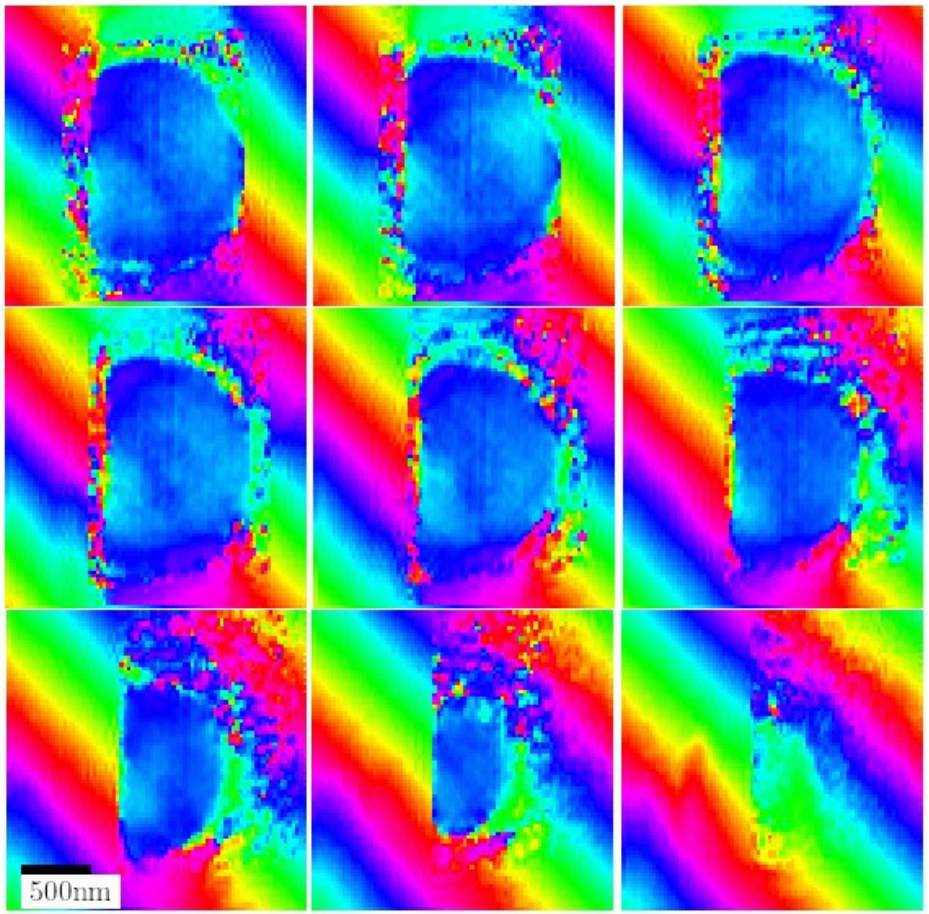

Figure 4.12: Center slices from 3D CXD pattern from Pb sample, on a log scale. Data file 296 from 10/03.

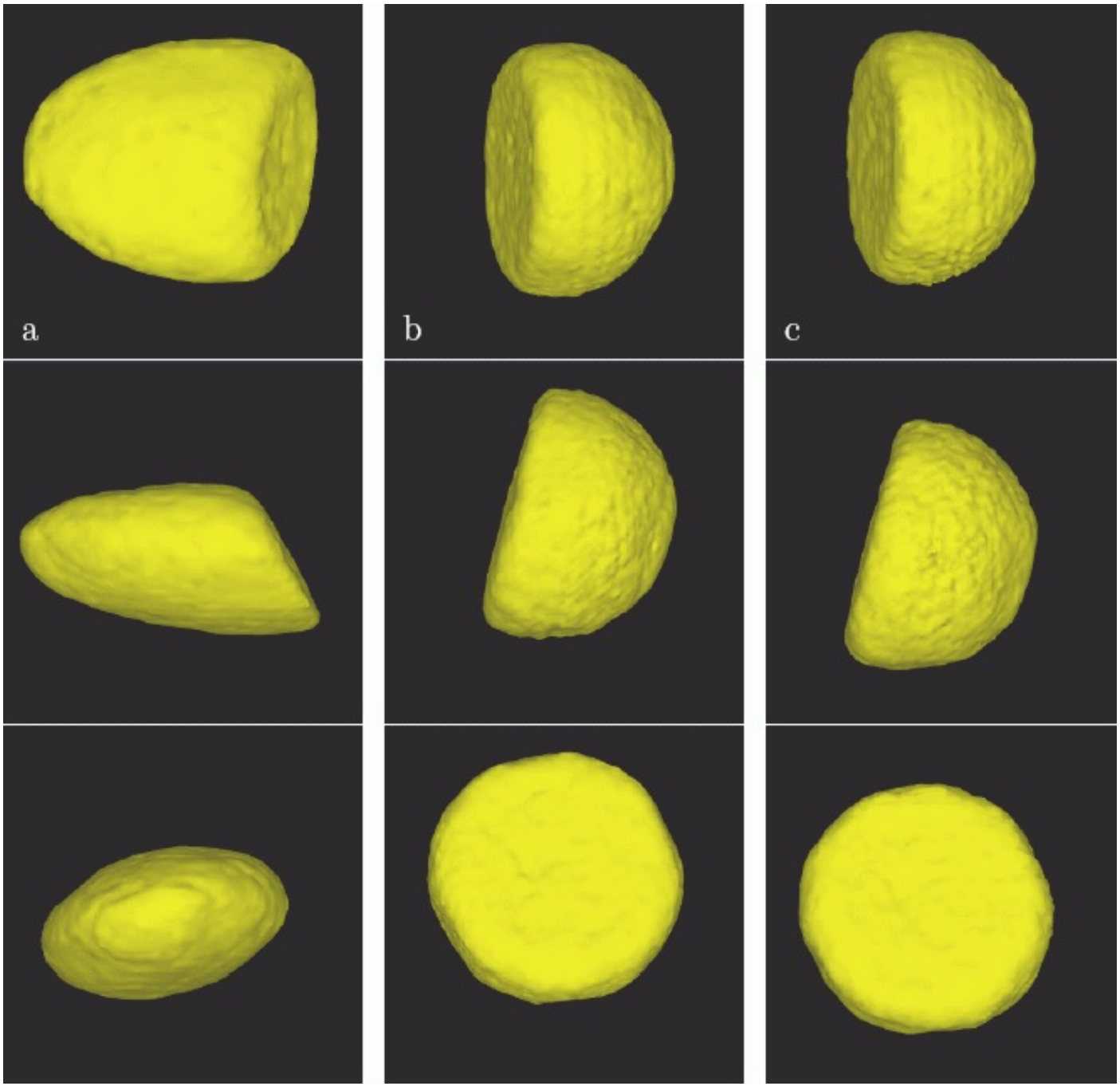
# Learn shape of “tight” support



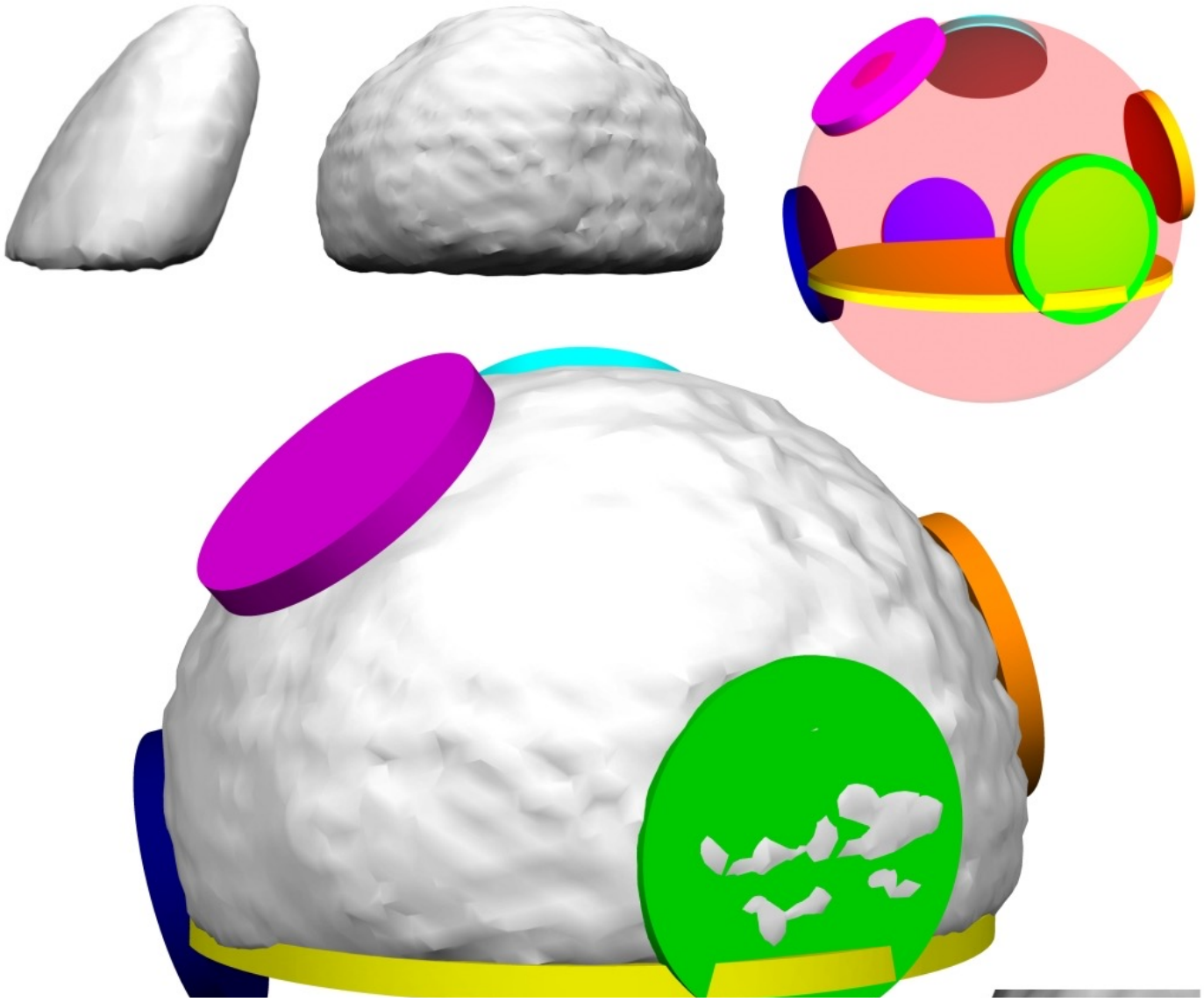
# Then refine amplitude *and* phase



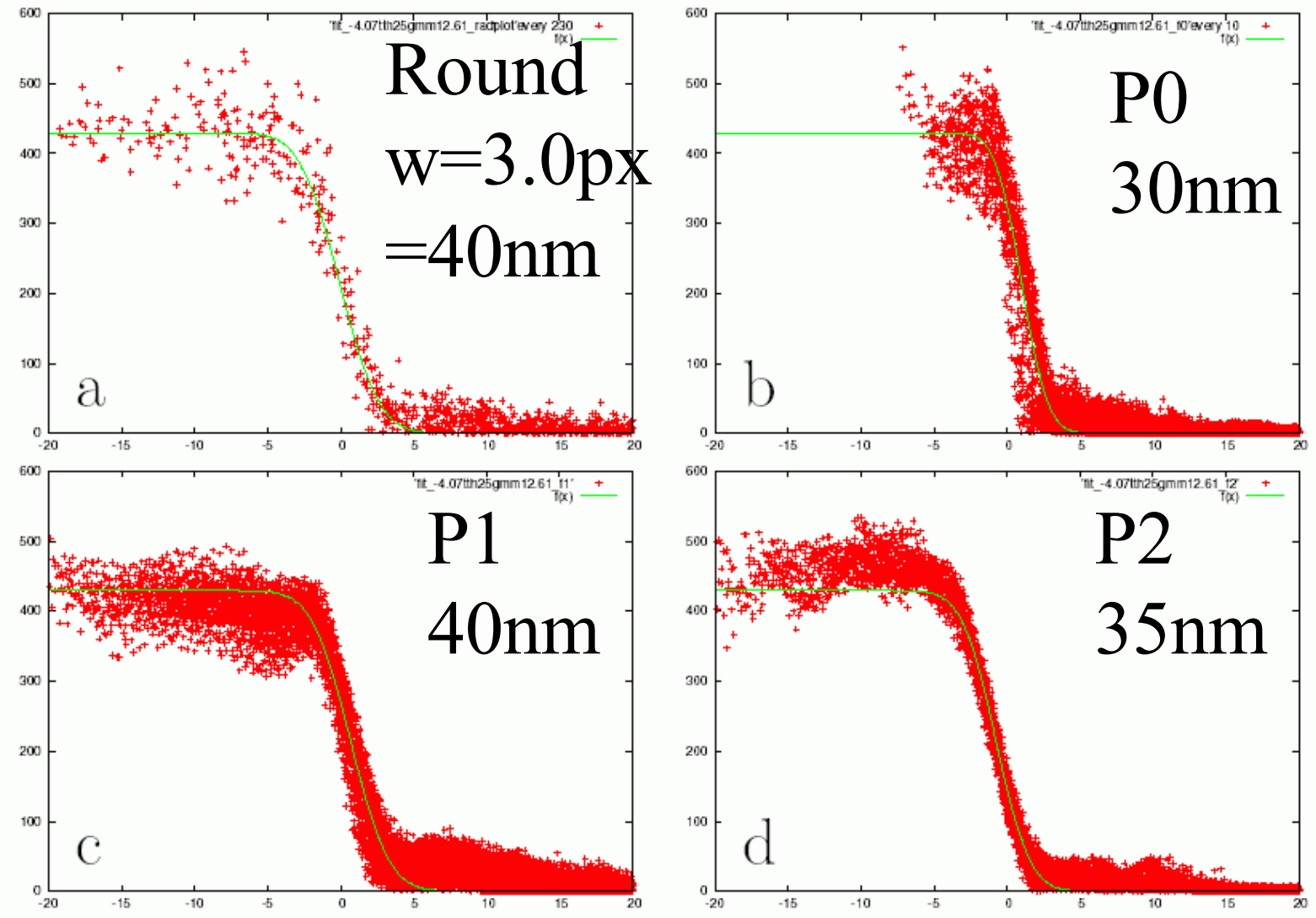




a
b
c

# Fitting of crystal shape



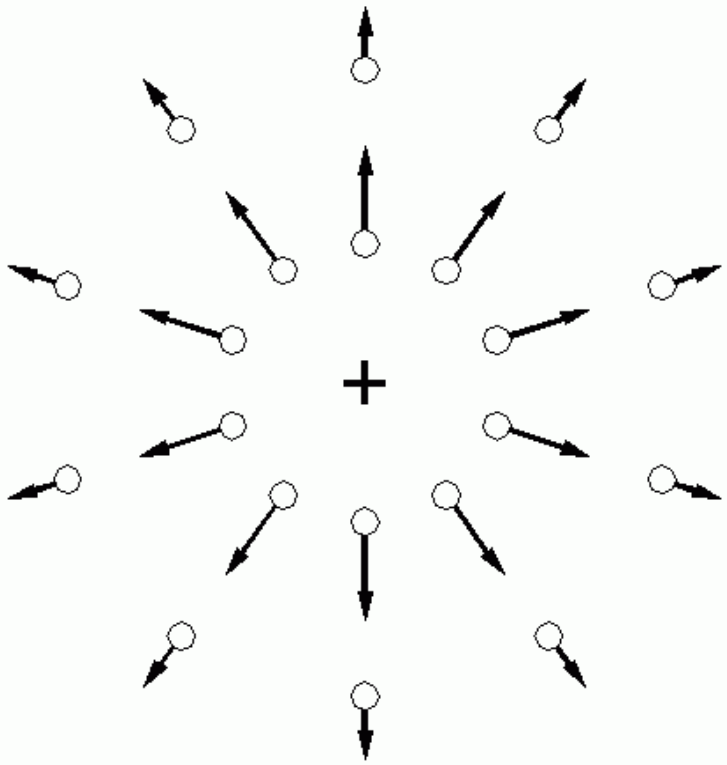
# Density distribution across surface

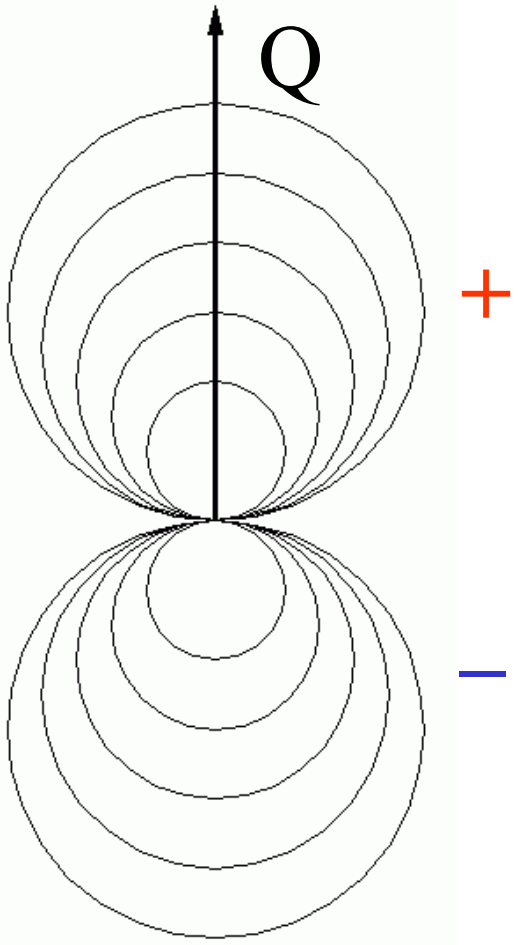


# Diffraction by Strain of Point Defect

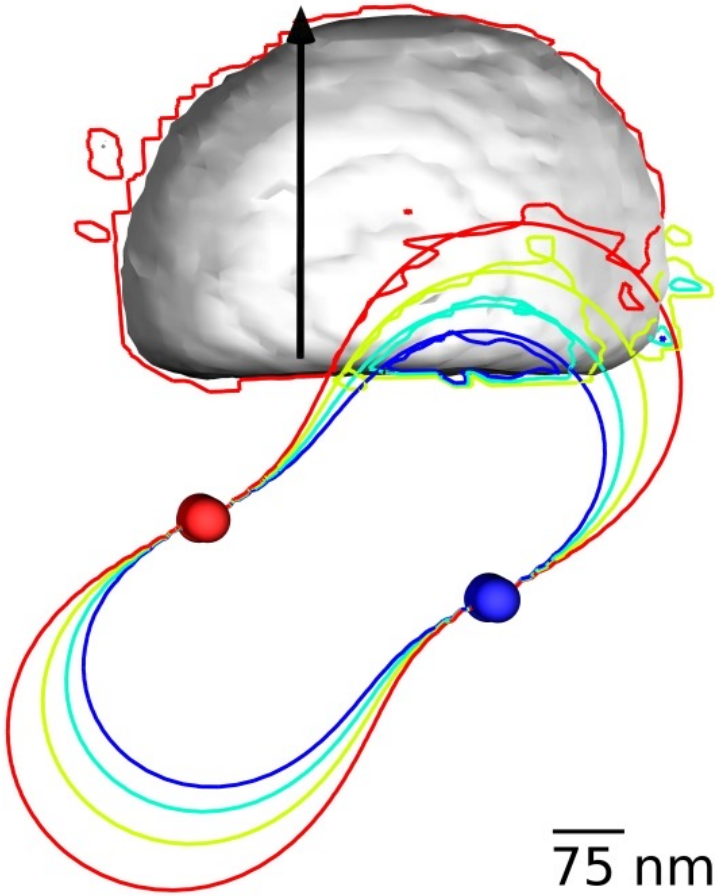
$$A \sim \sum e^{i\mathbf{Q}\cdot(\mathbf{R}_j+\mathbf{u}_j)}$$
$$\approx \sum e^{i\mathbf{Q}\cdot\mathbf{R}_j}(1+i\mathbf{Q}\cdot\mathbf{u}_j)$$

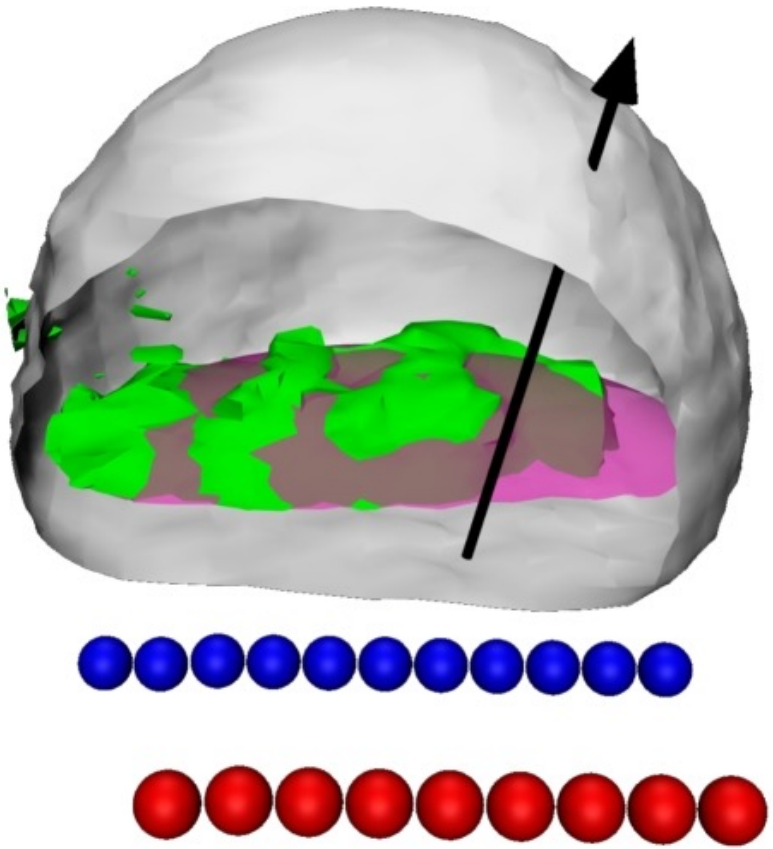
Imaginary density





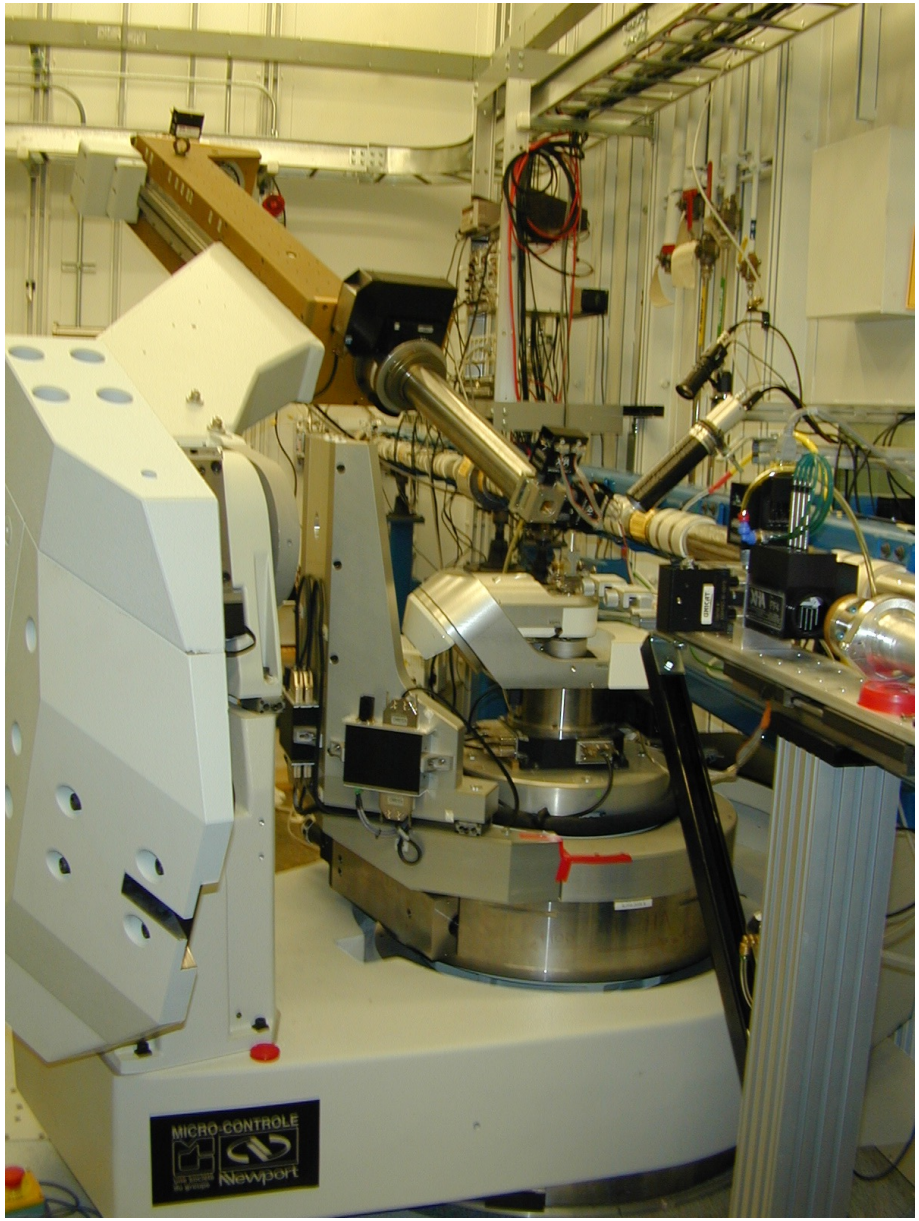
# Modeling of Deformation Field

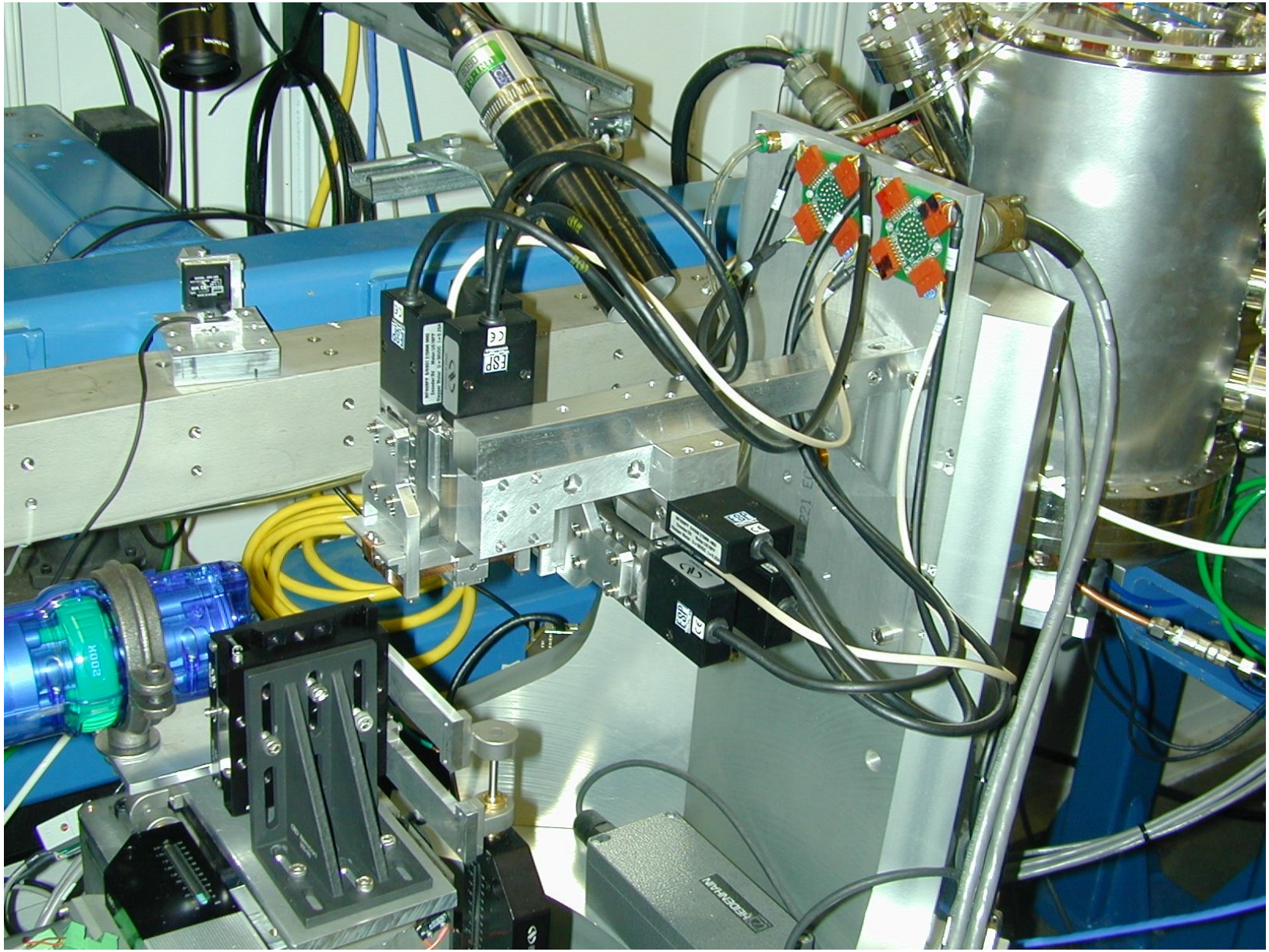


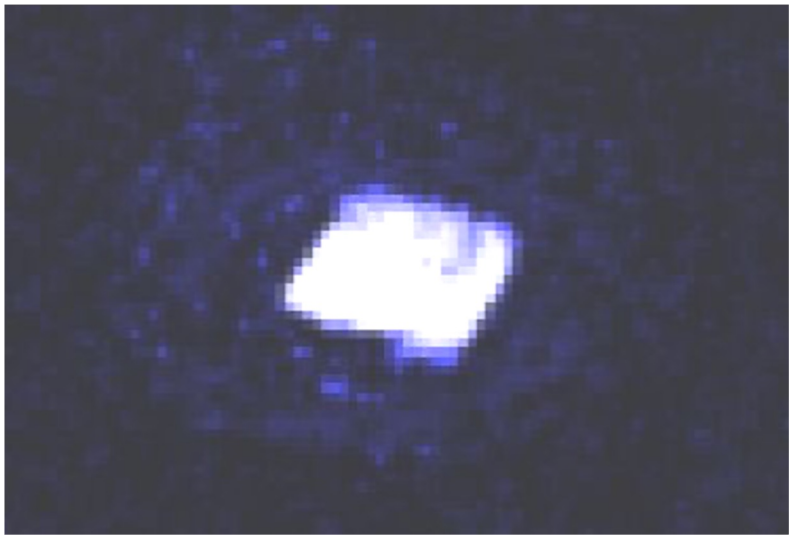
# Extrapolation to Euro-XFEL

- Steep coherent flux limit to accessible size scales: $a^6$ or $a^3$ ?
- $10^{12}$ photons/sec will be $10^{12}$ photons/shot
- Already at 100nm, sample stability important
- Smaller objects move faster: scales as $a^5$.

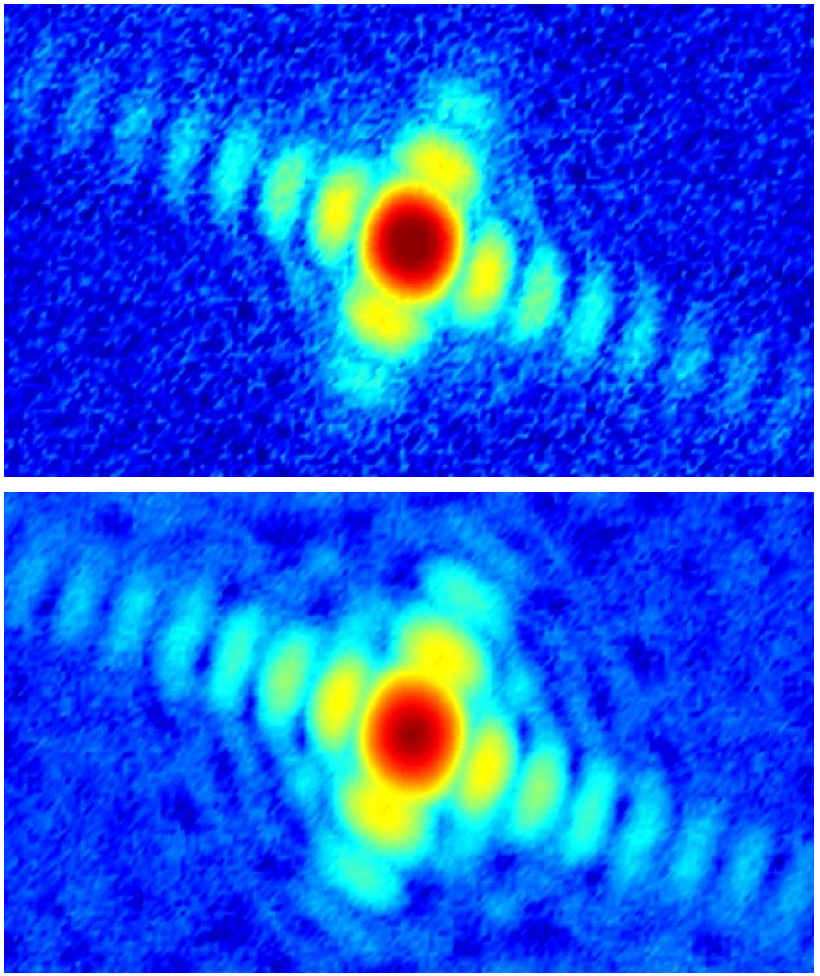

MICRO-CONTROLE
Newport



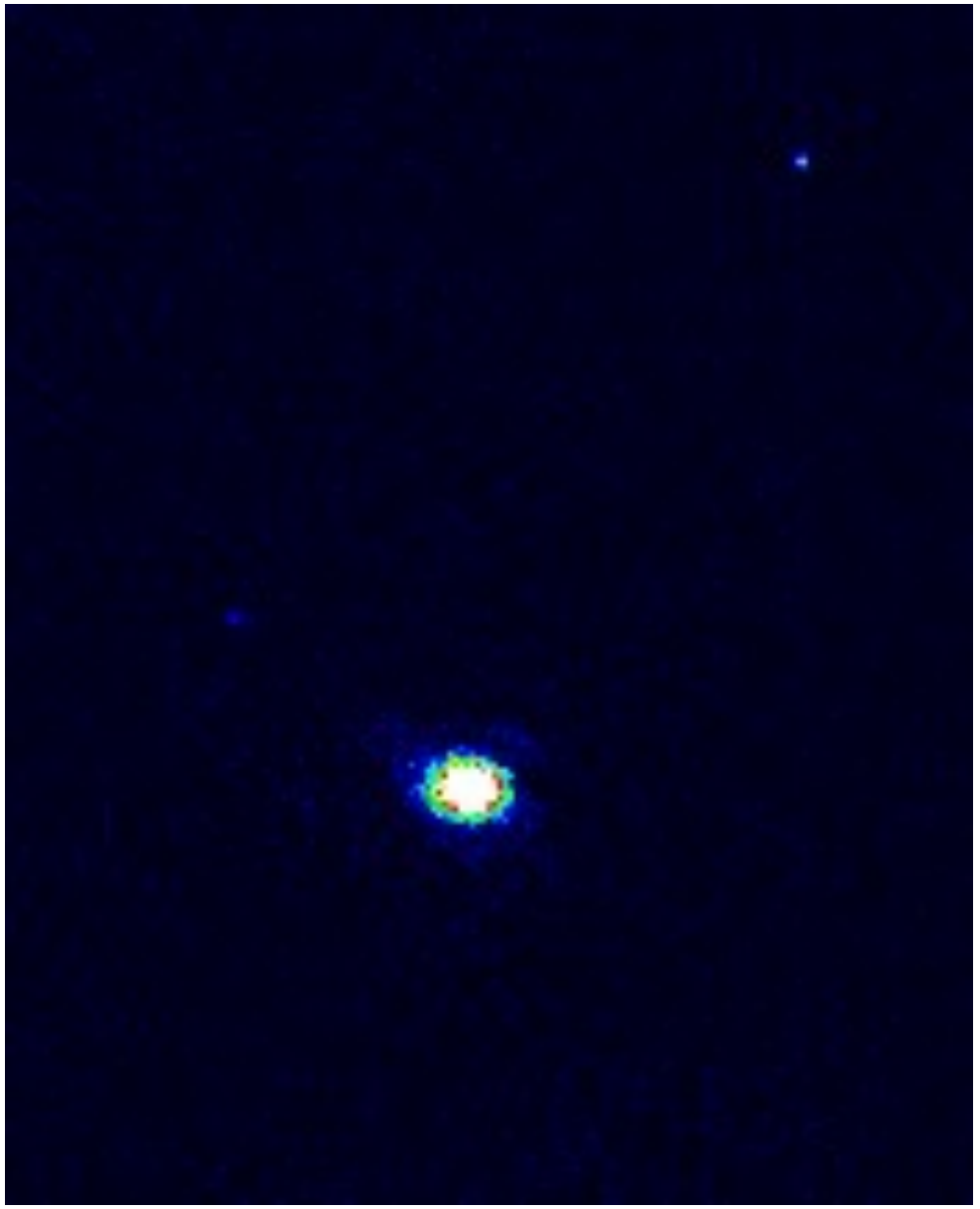
# Reconstruction of Ag Nanocrystal

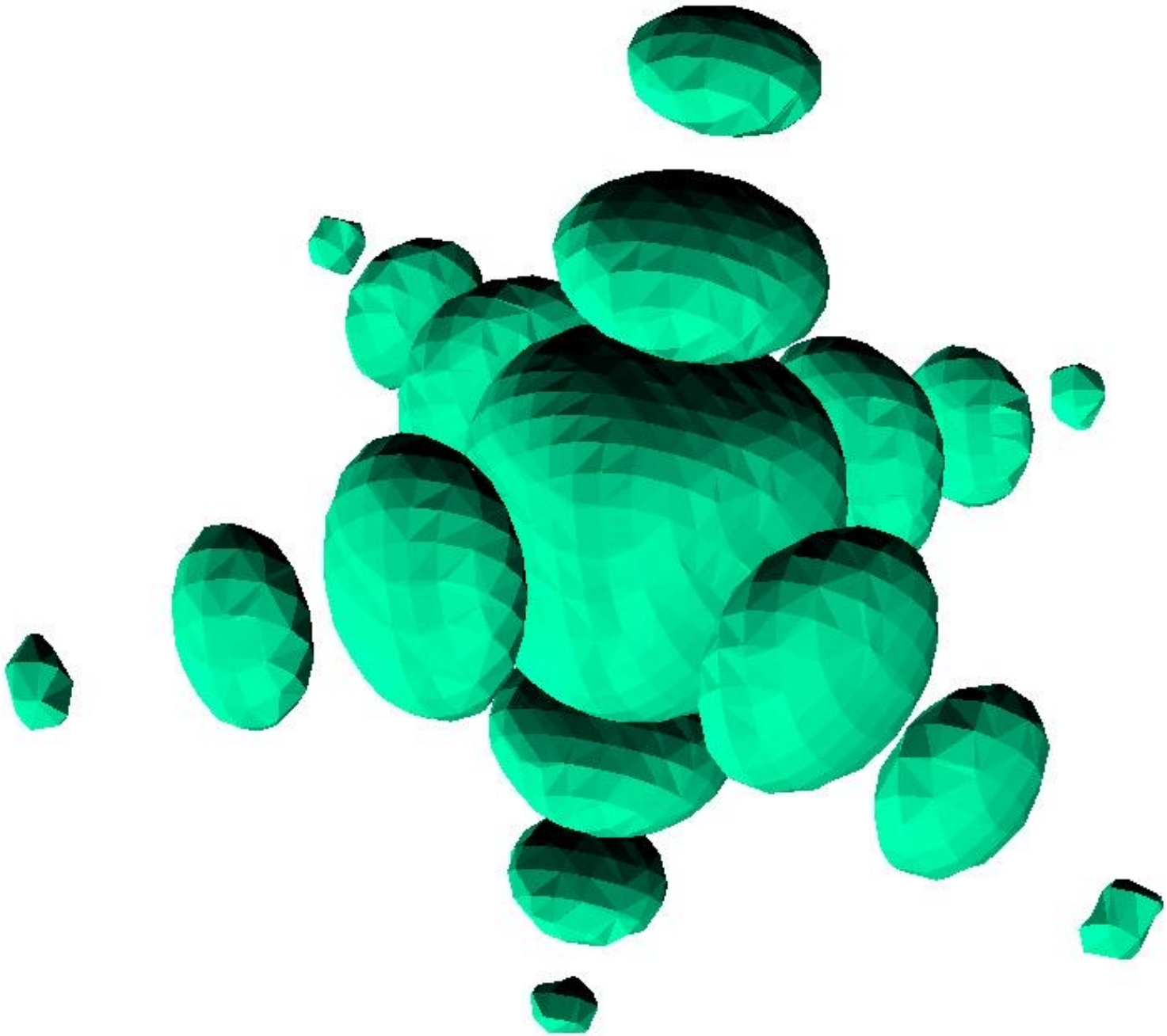


200nm



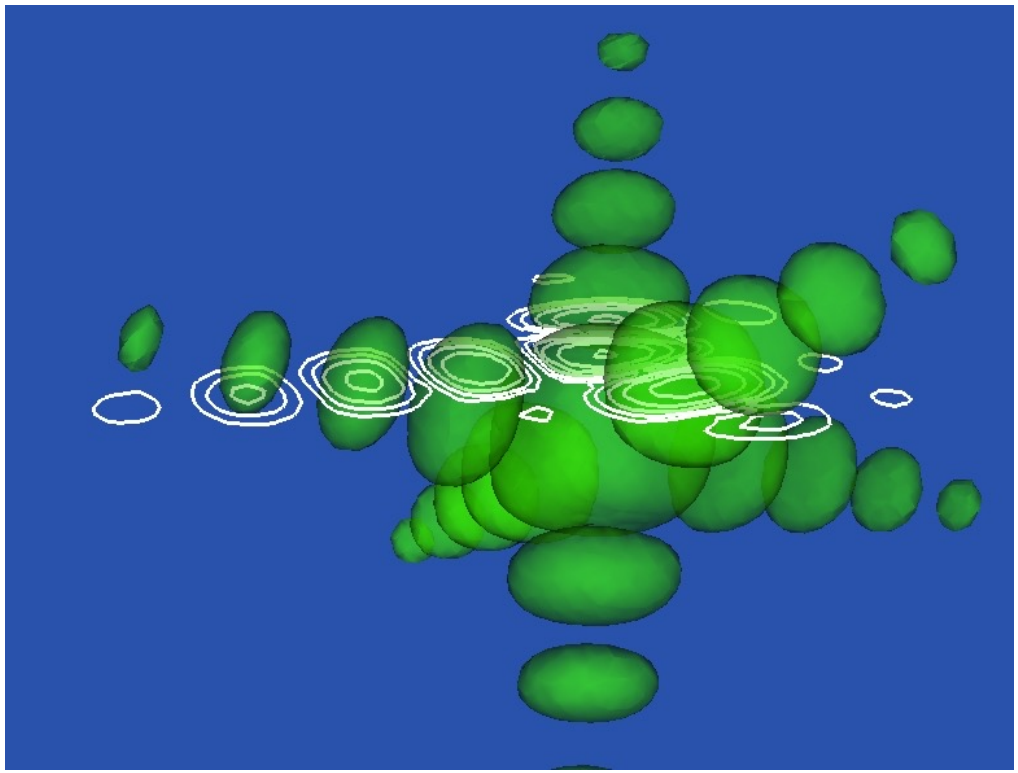
# Rocking scan of Ag cubes with 0.01° steps





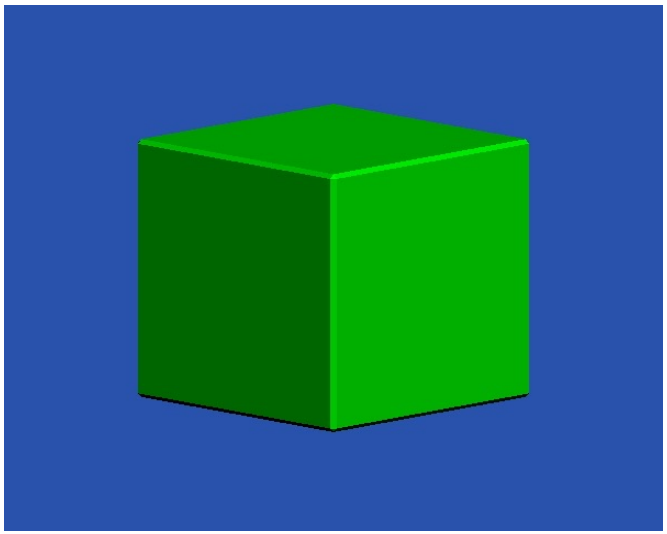
# Miscentered Non-Centrosymmetric Reciprocal Space Slice

$$A_q \neq A^*_{\bar{q}}$$

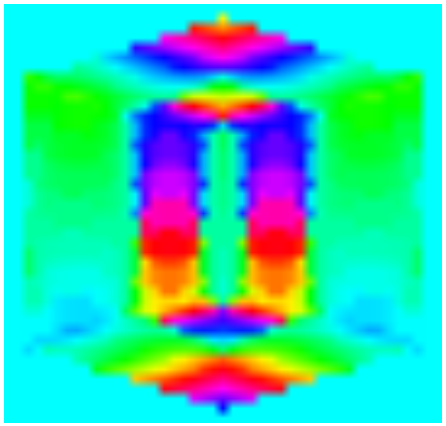


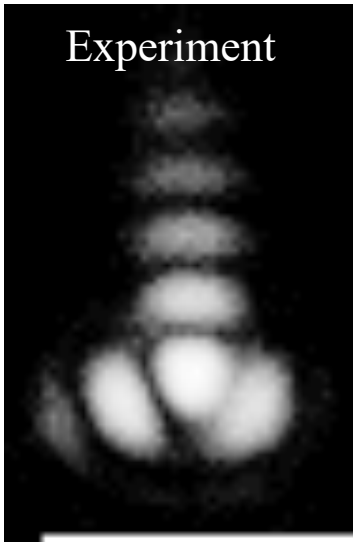
**Real-space phase.**

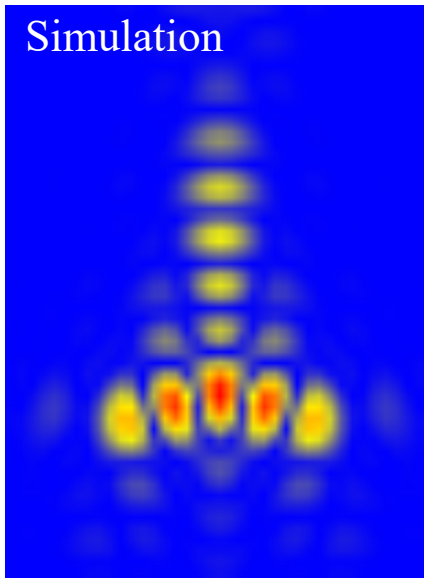
# What can we expect to see?



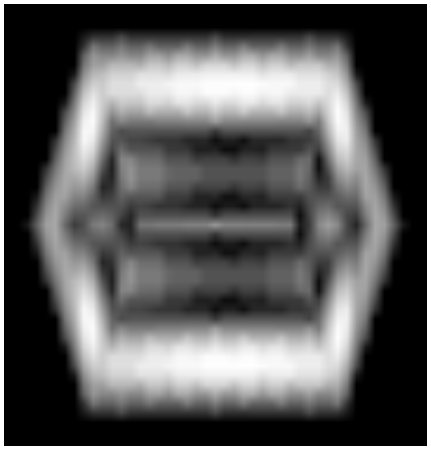
$$A(\bar{q}_o+\bar{q}_z)=\sum_i \sum_z \underbrace{\rho(x,y,z)}_{u_i\{x,y\}} e^{2\pi i \bar{q}_z\cdot\bar{z}_i} e^{2\pi i\{\bar{q}_o\}\cdot\bar{r}_i}$$

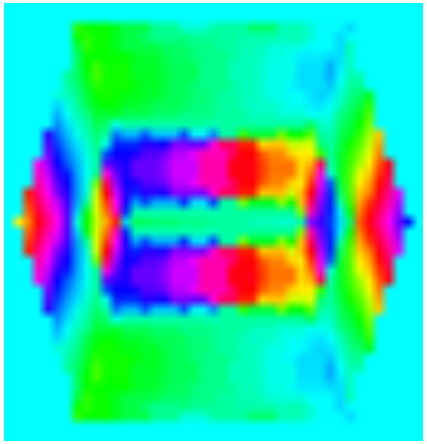


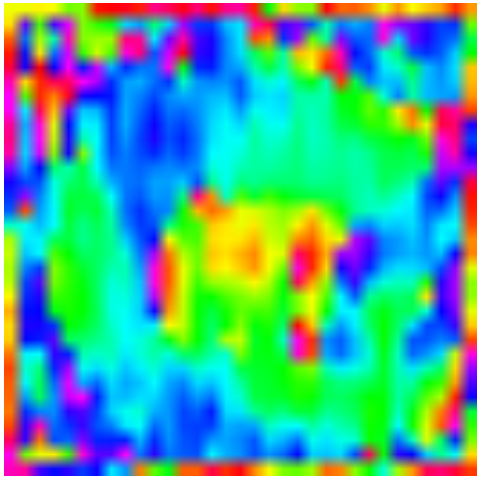


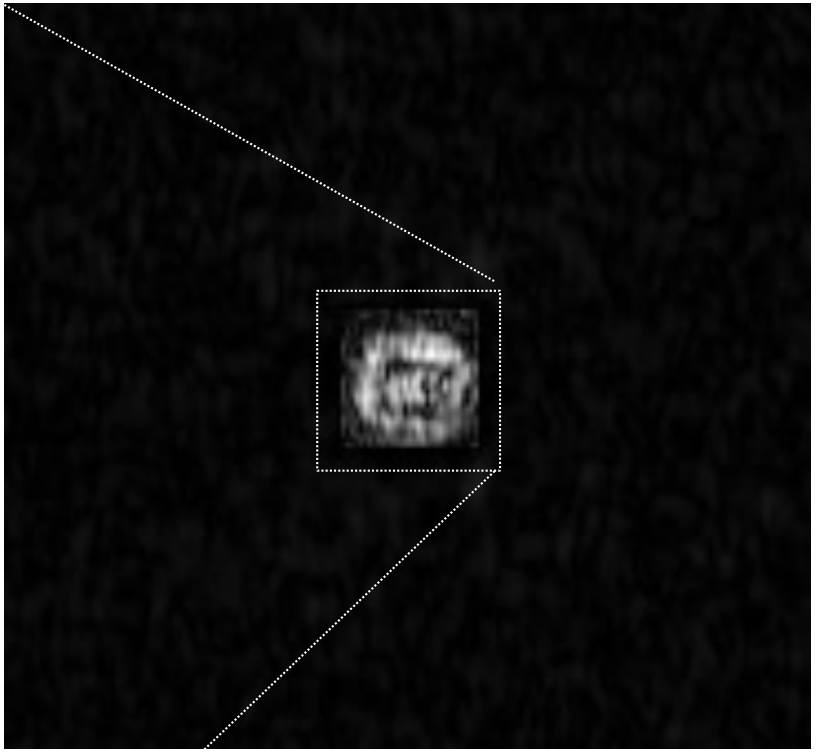



# Direct Space Result (1000 Iterations)





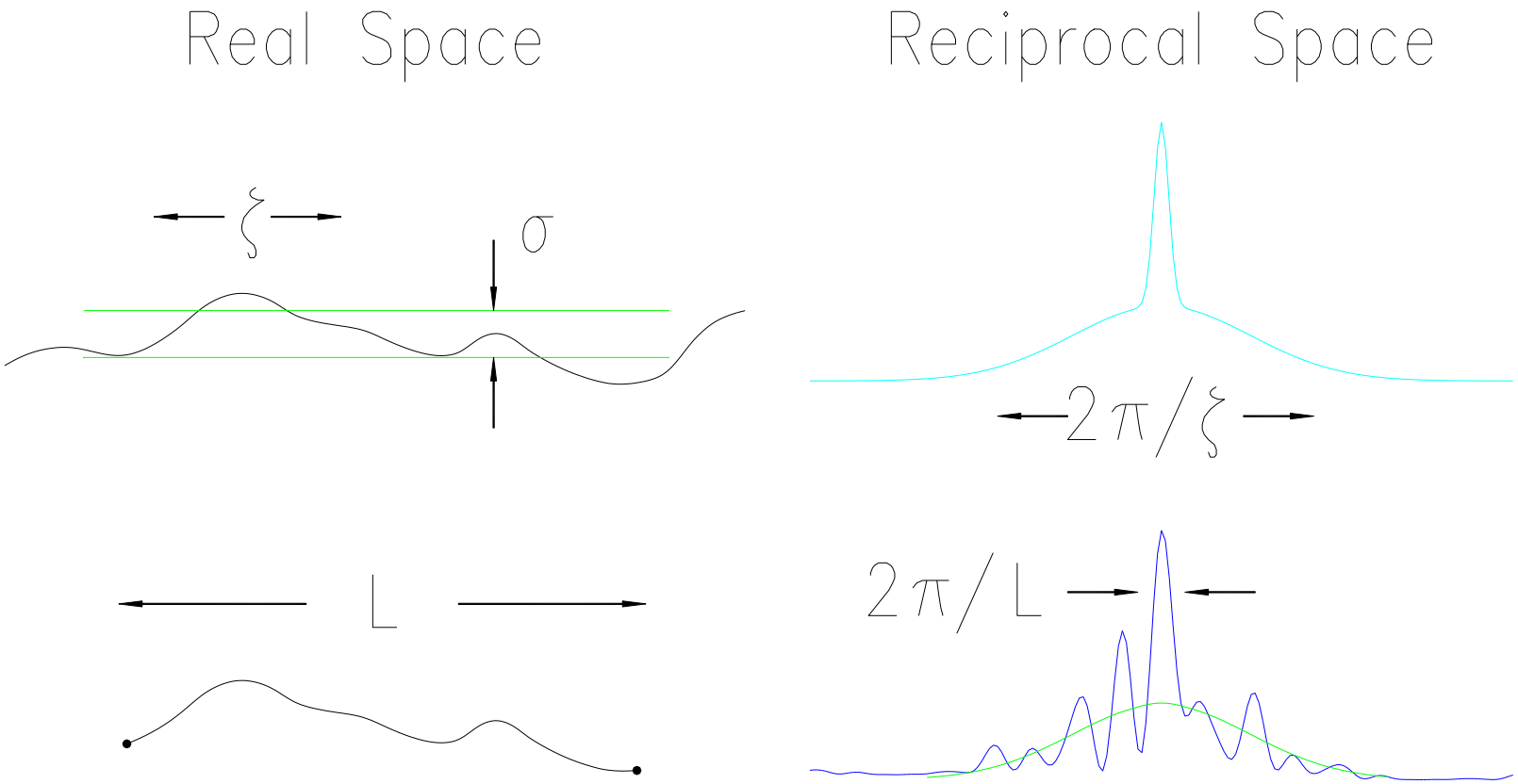




# “First” Euro-XFEL Experiment

- Critical nucleation at solid/liquid boundary
- Fluctuations of solid in liquid near MP
- Pre-critical nuclei during crystallisation
- Smaller nuclei harder to see: focus
- Smaller nuclei fluctuate more: snapshot

# Diffuse Scattering acquires Structure using CXD



# Conclusions and Outlook

- Phase Retrieval of CXD demonstrated
- Internal structure of Au Nanocrystals
- Phase structure visible in Pb Nanocystals
- Focusing for access to much smaller clusters
- Thermal motion of small clusters important
- Euro-XFEL can solve both problems