Coherent lensless imaging with present and future light sources

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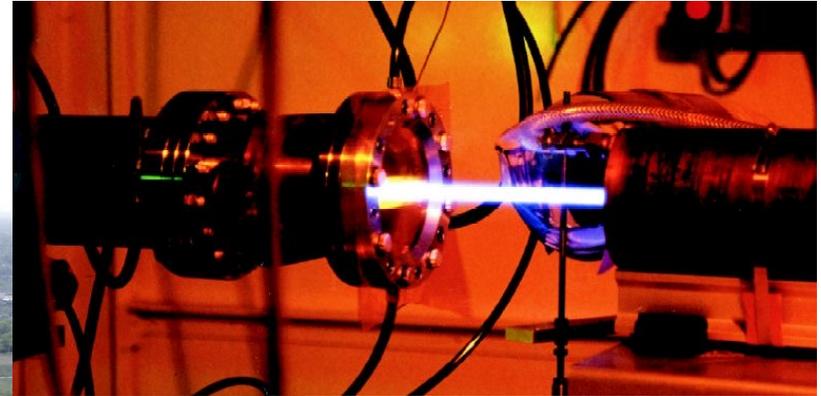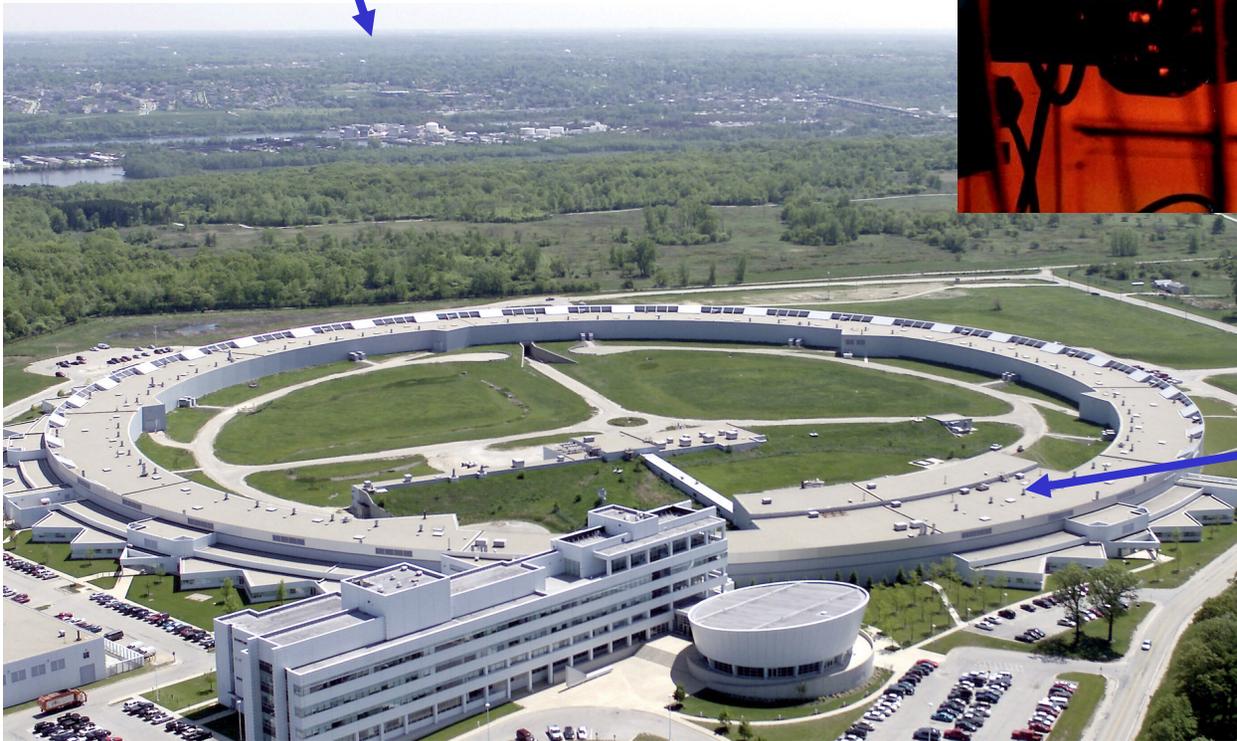SLS-Soleil Workshop

"The Full Spectrum"
Villigen, September 2006

Outline

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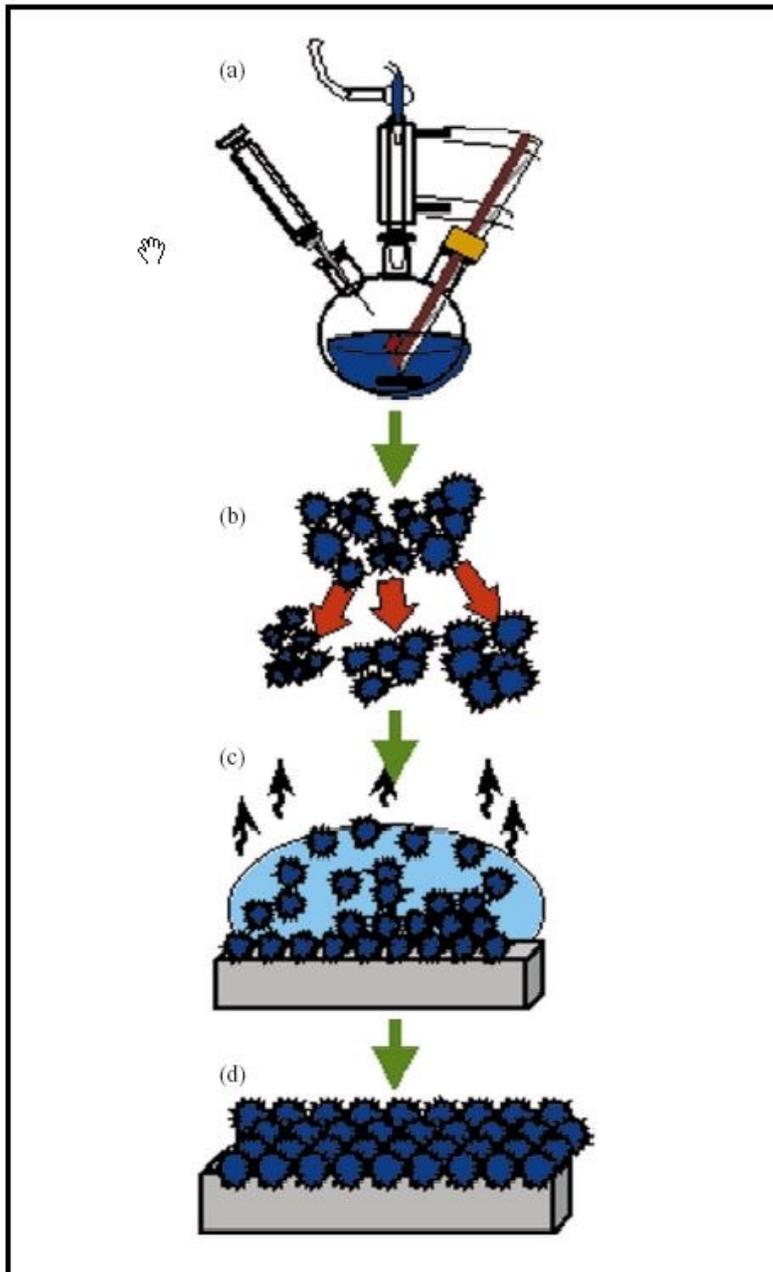- Opportunities with Electrons and FELs
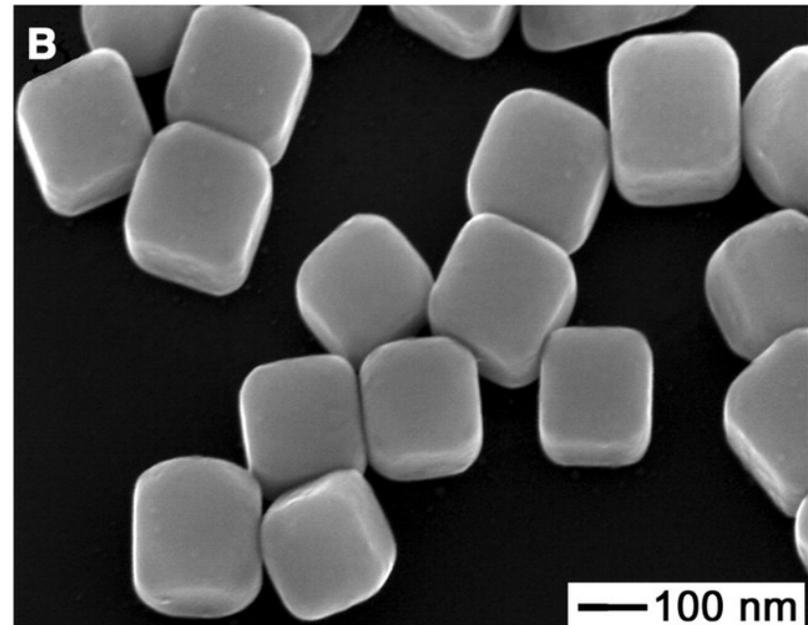
Synchrotron Radiation

Urbana

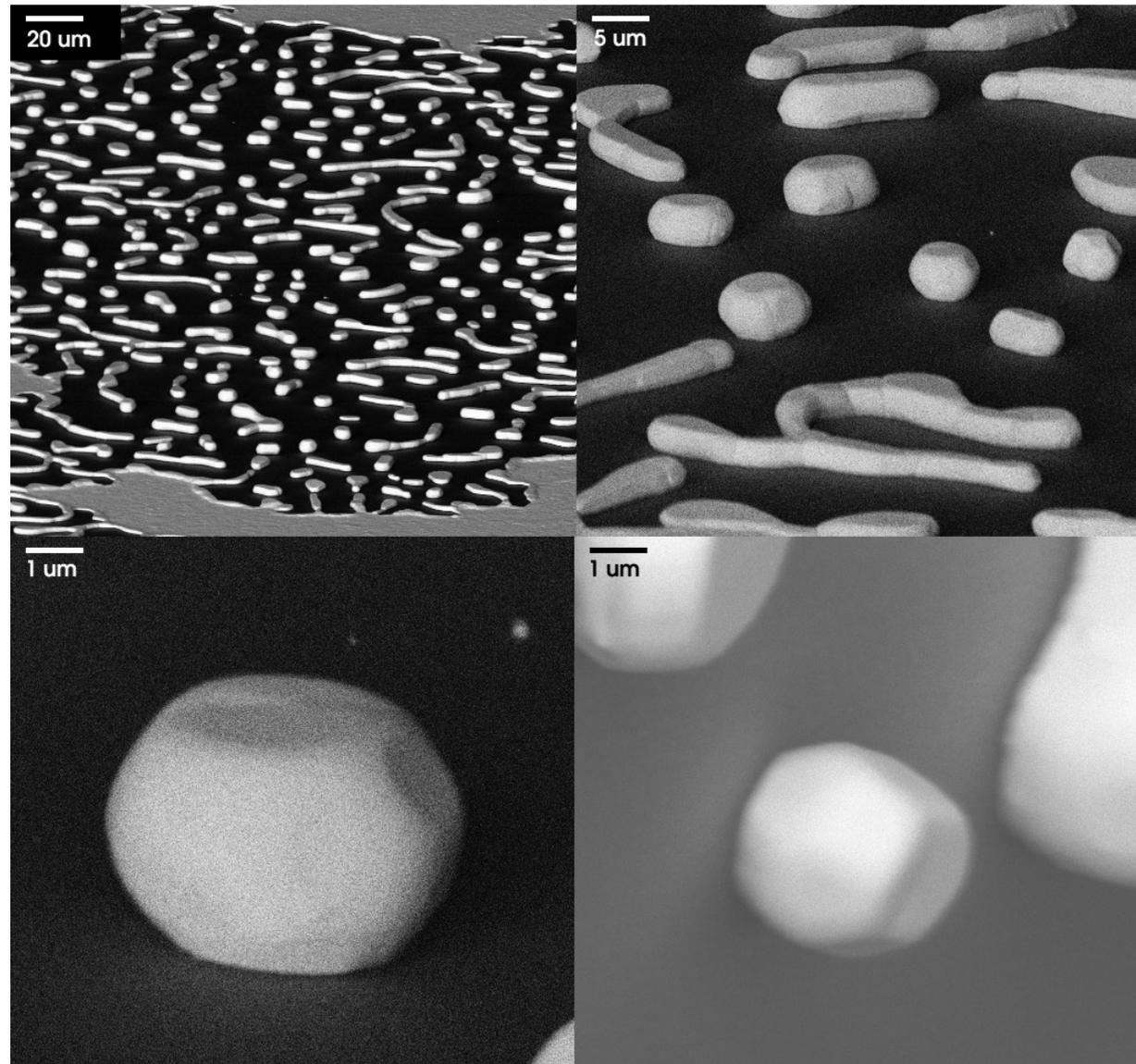**34-ID-C**

Chemical Synthesis of Nanocrystals

- Reactants introduced rapidly
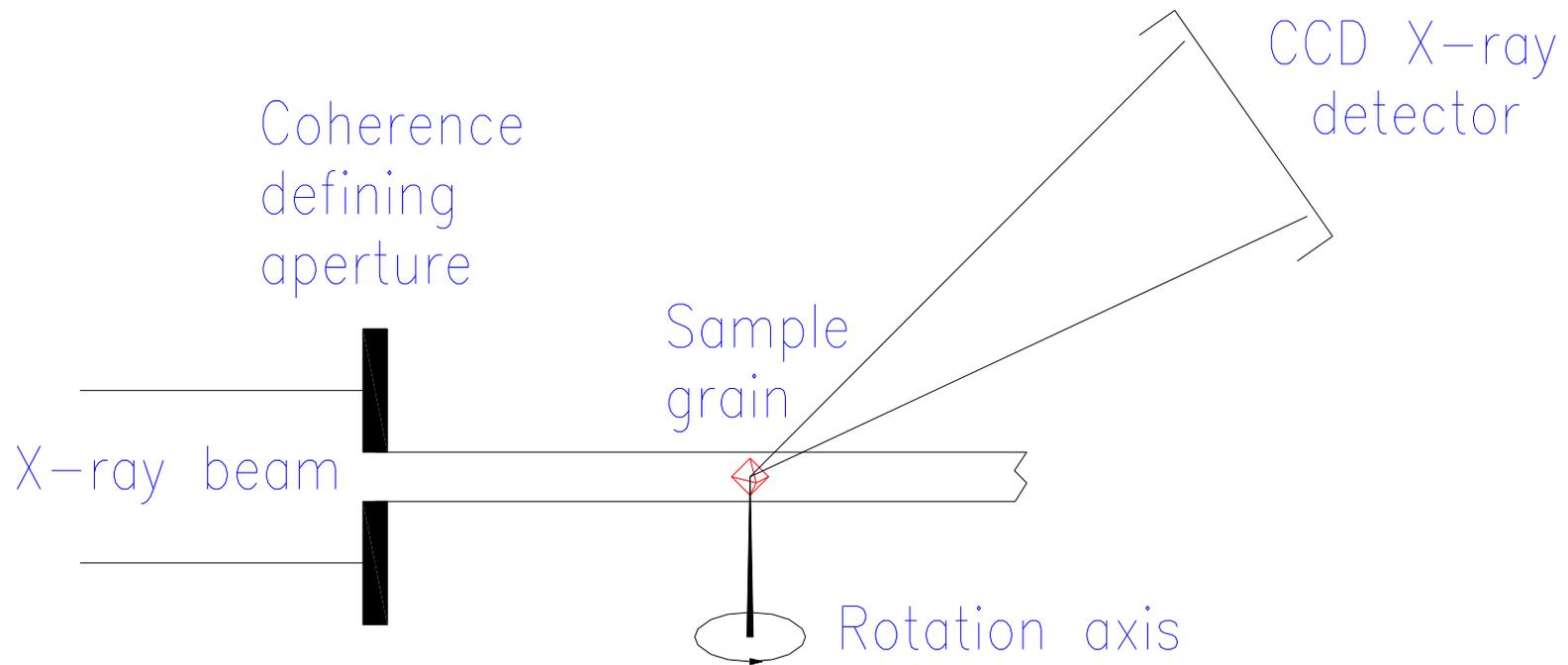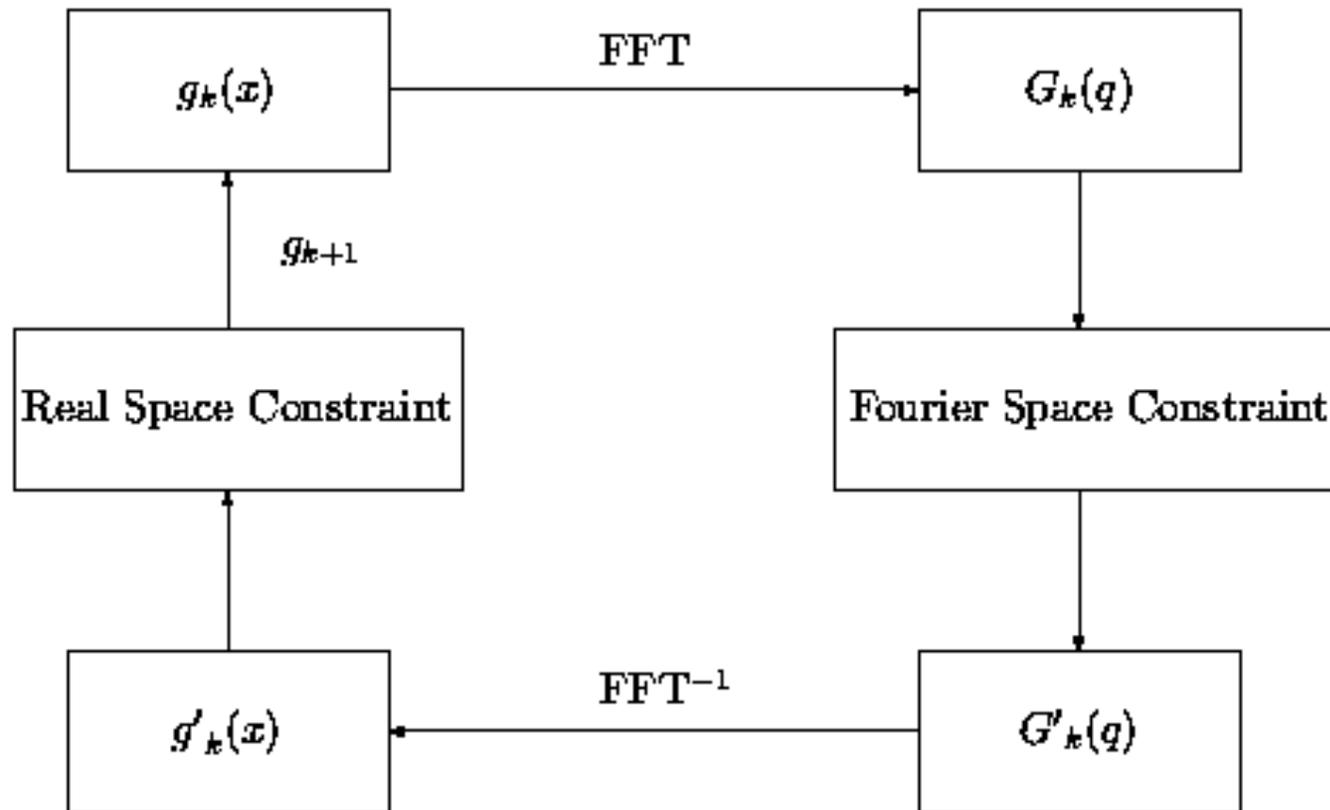- High temperature solvent
- Surfactant/organic capping agent

SEMS

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

Lensless X-ray Microscope

Coherence defining aperture

CCD X-ray detector

Sample grain

X-ray beam

Rotation axis

Generic "Error Reduction" method

J. R. Fienup Appl. Opt. <u>21</u> 2758 (1982)
R. W. Gerchberg and W. O. Saxton Optik <u>35</u> 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)
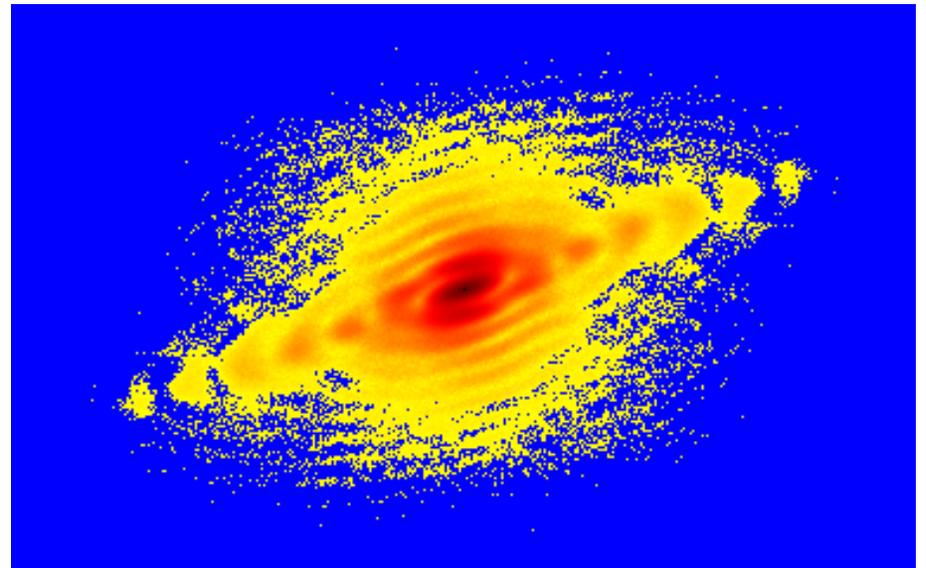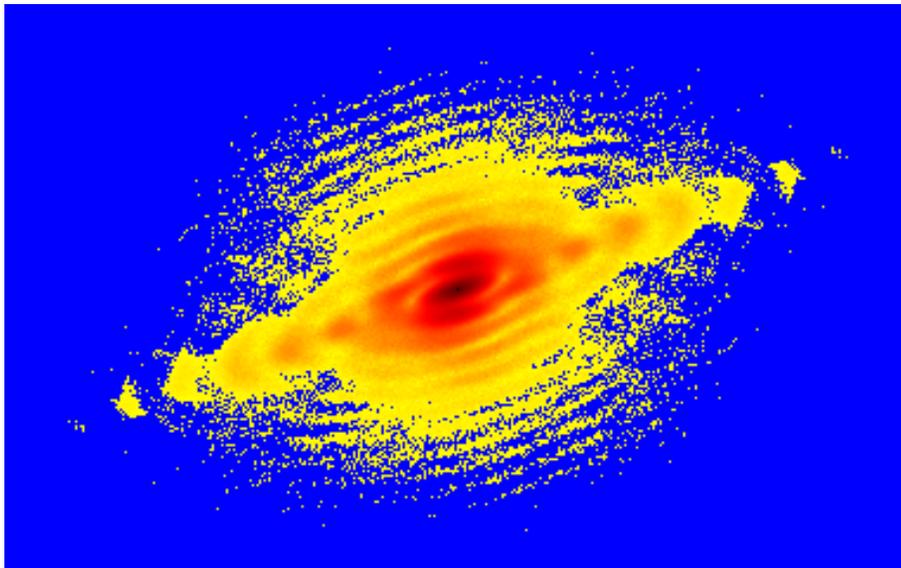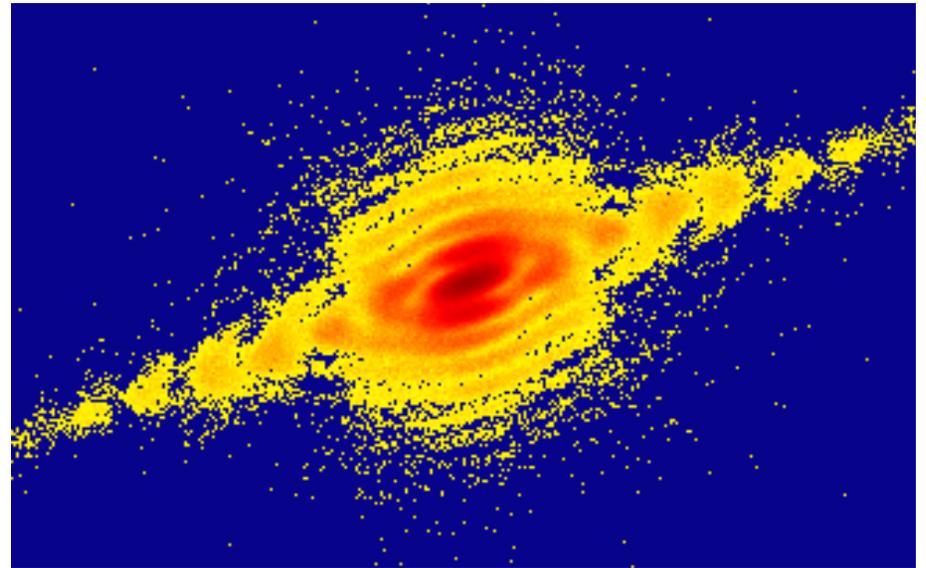
Phase Problem: Finite-size Effect

I. K. Robinson, The Full Spectrum, Sept 2006
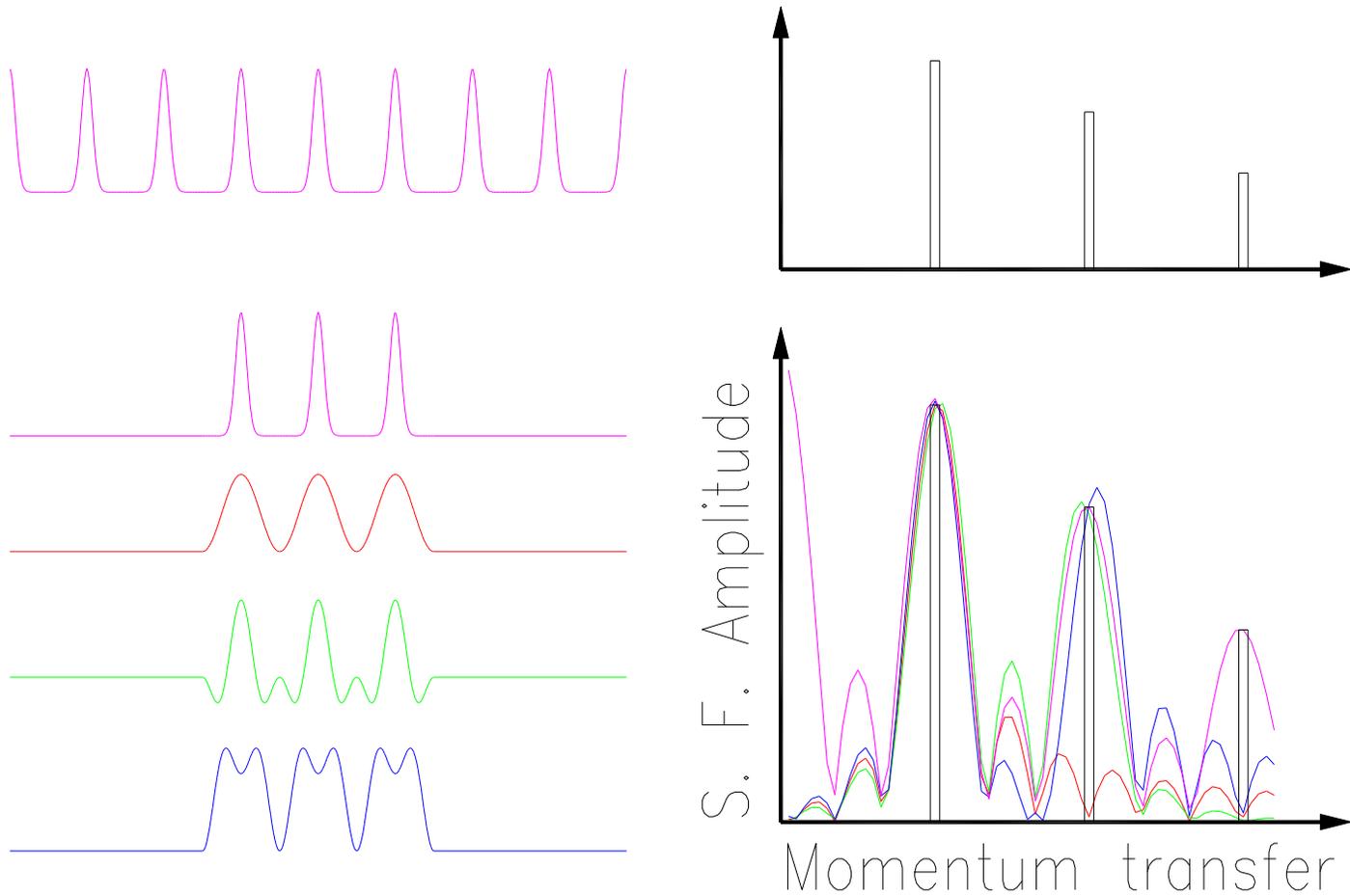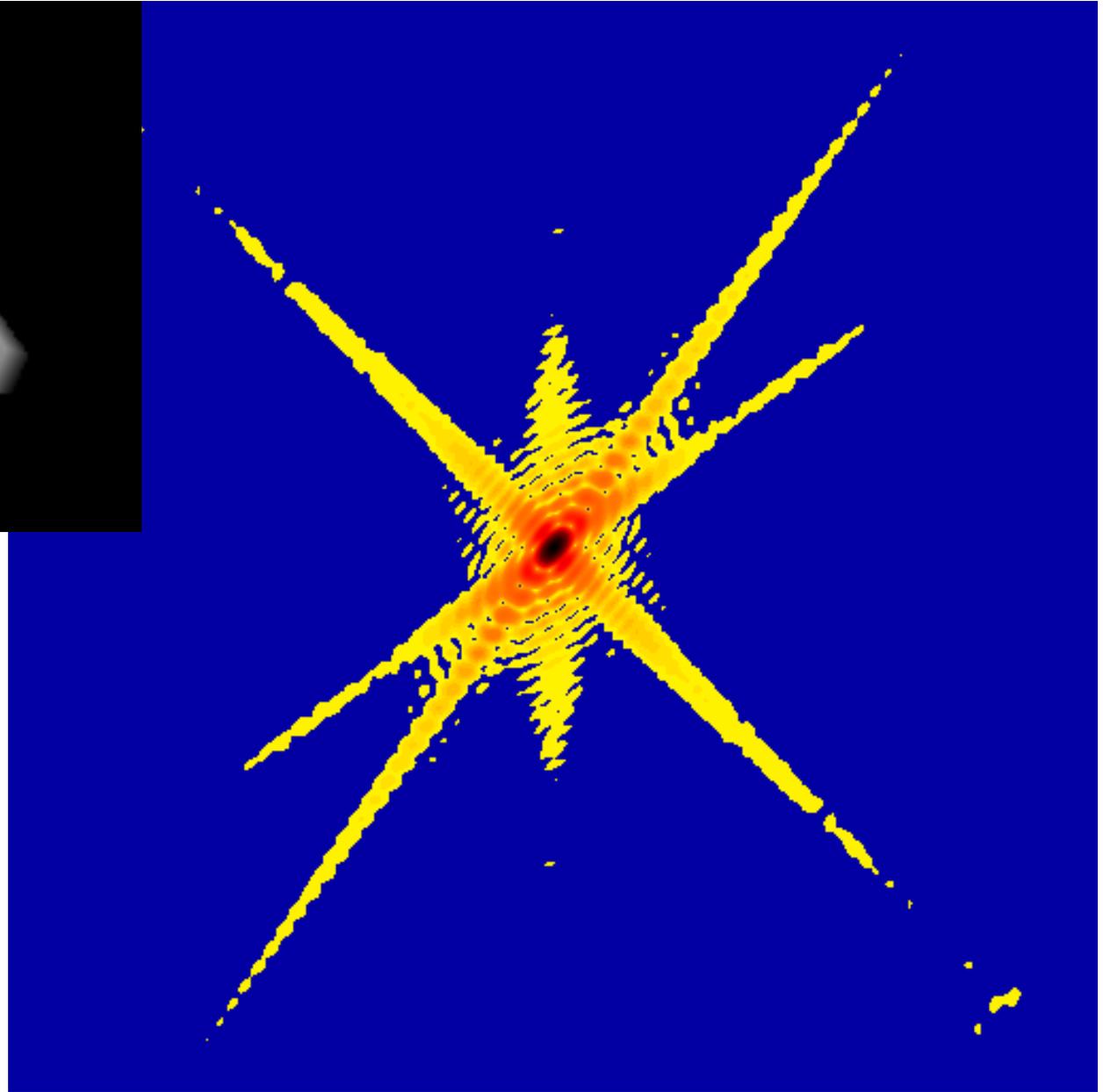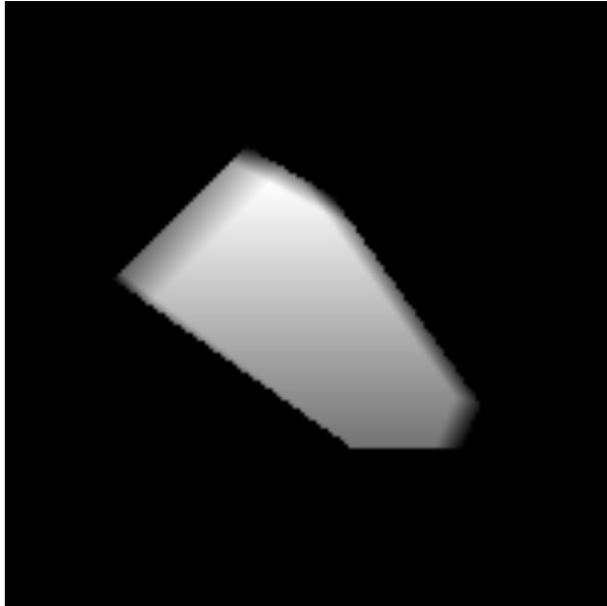
Coherent Diffraction from Crystals
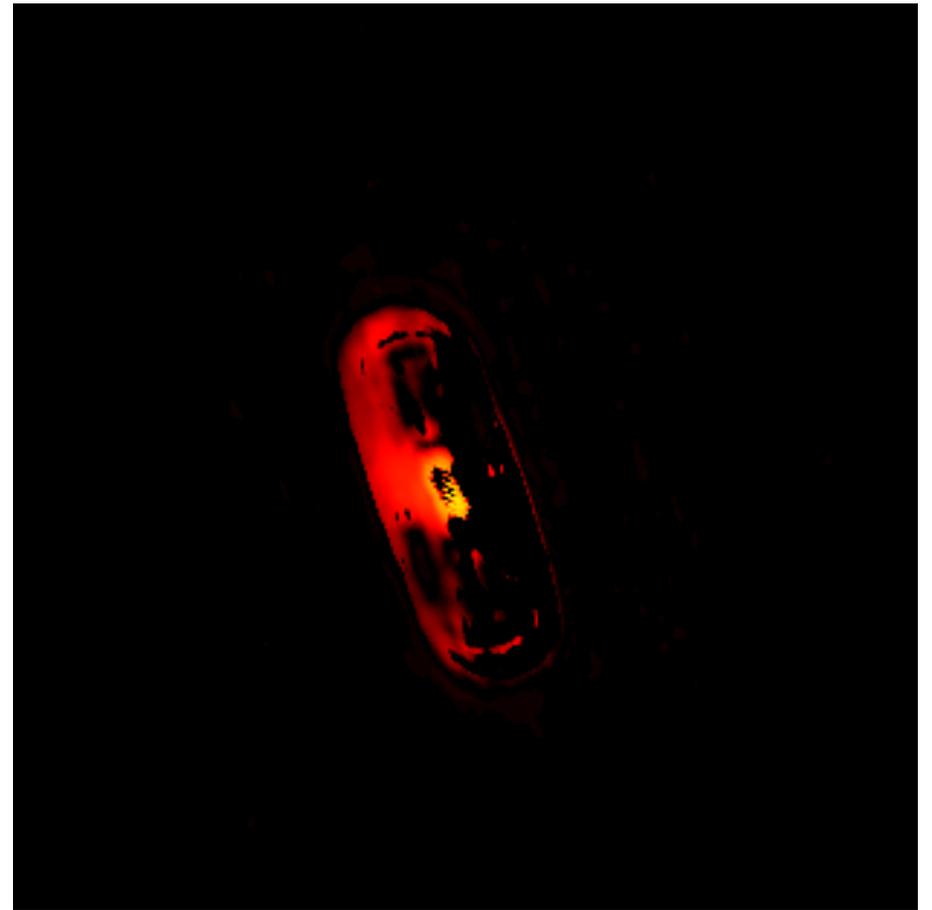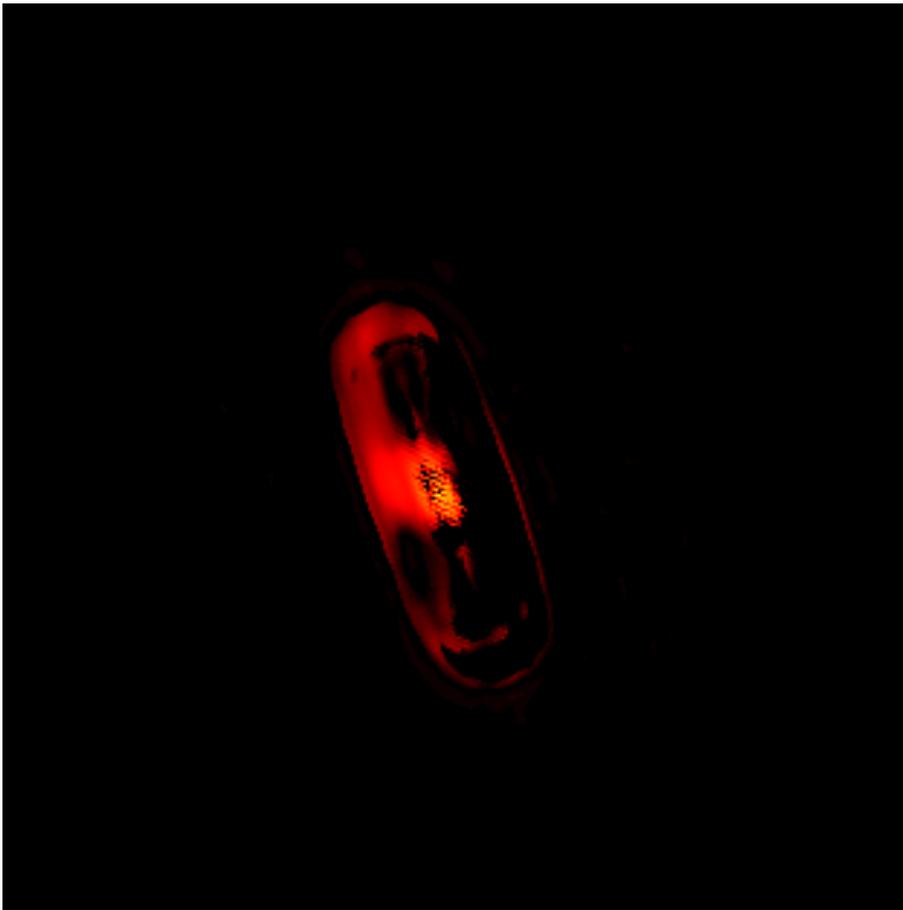
Fourier Transform

k

h
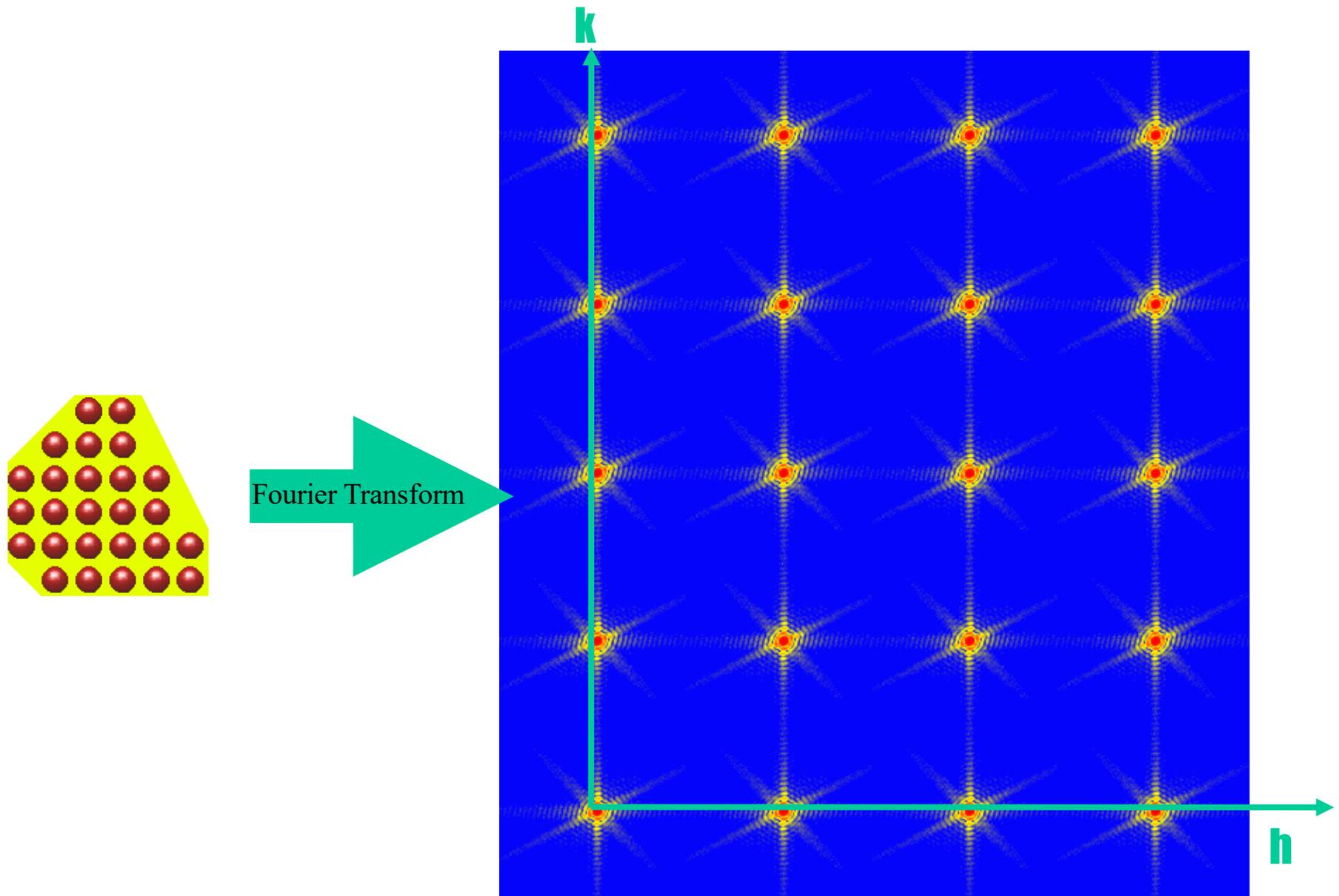
Symmetrized Data and two best fits

Chisq=0.0005

2D Reconstructions
chisquare = 0.0005

Facets of Equilibrium Crystal Shape

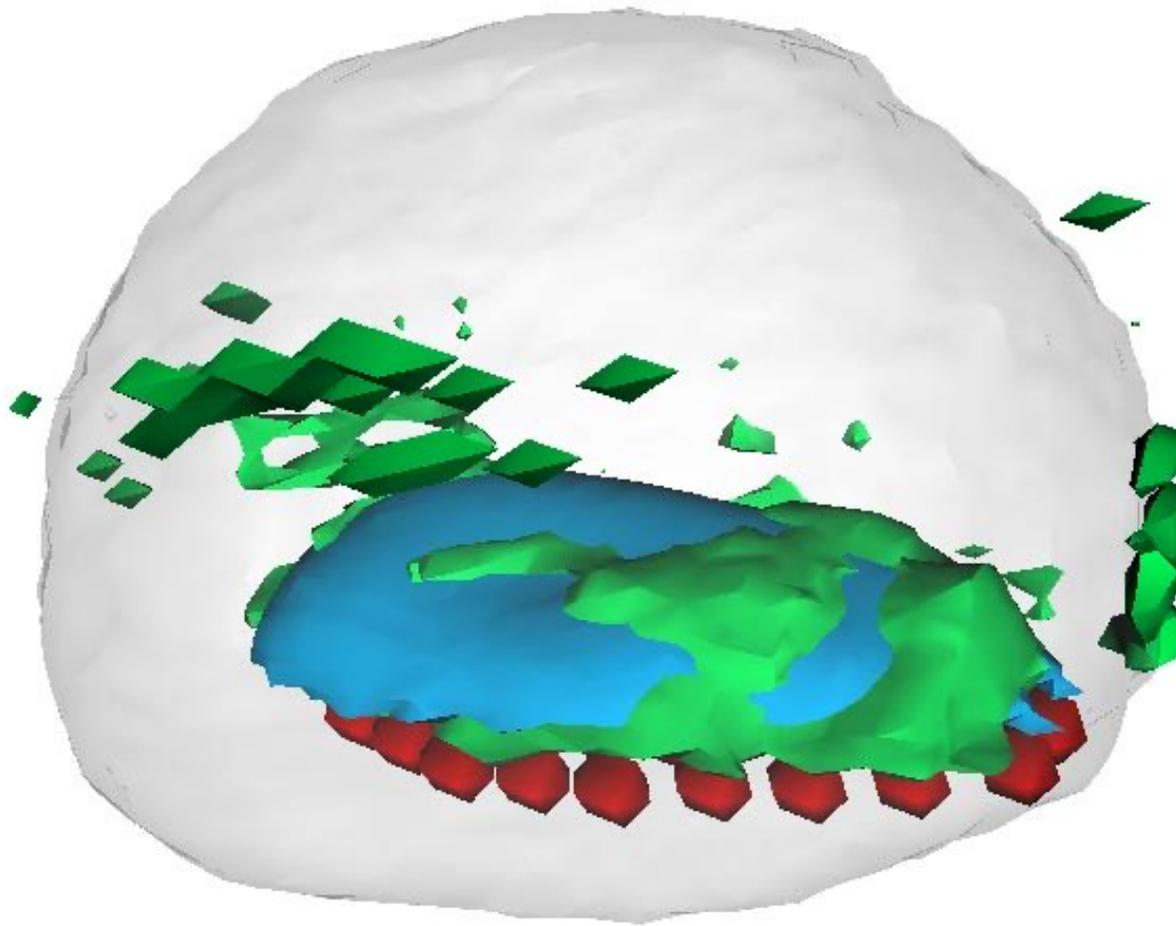# Real space image in phase too
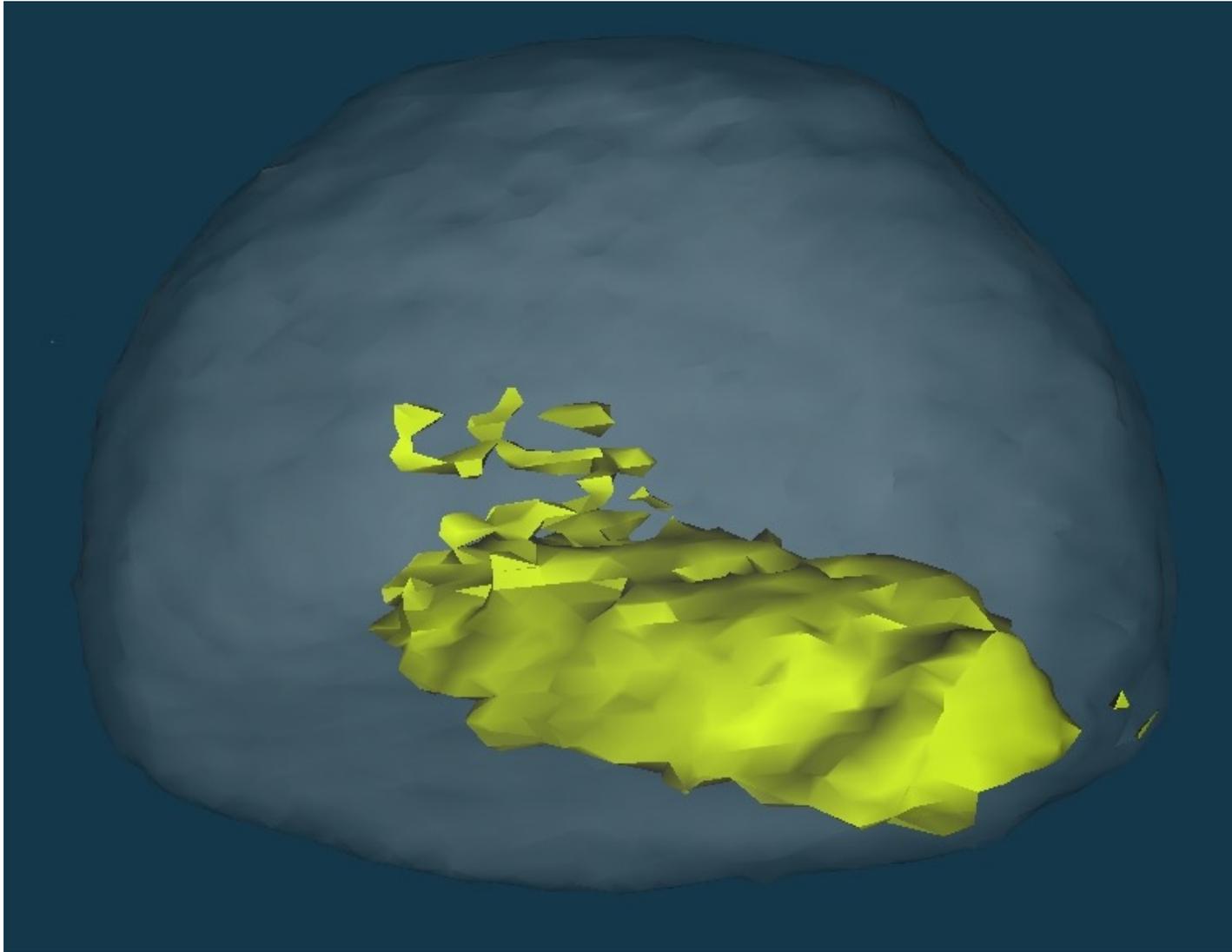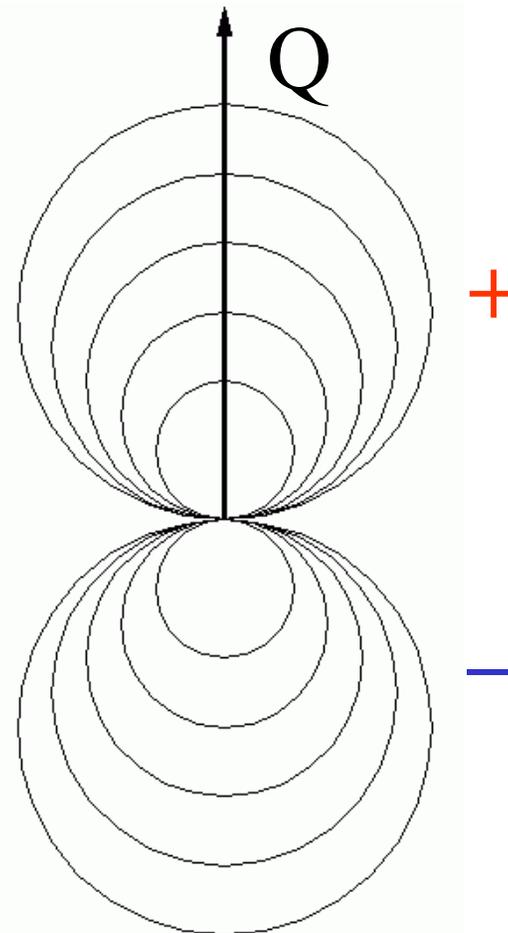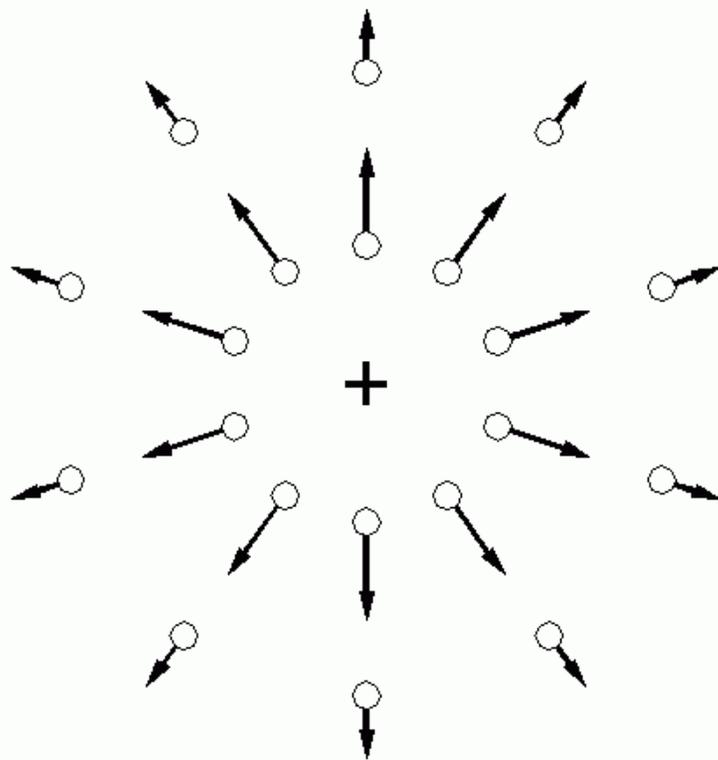
Fitted Phase Isosurface

Diffraction by Strain of Point Defect

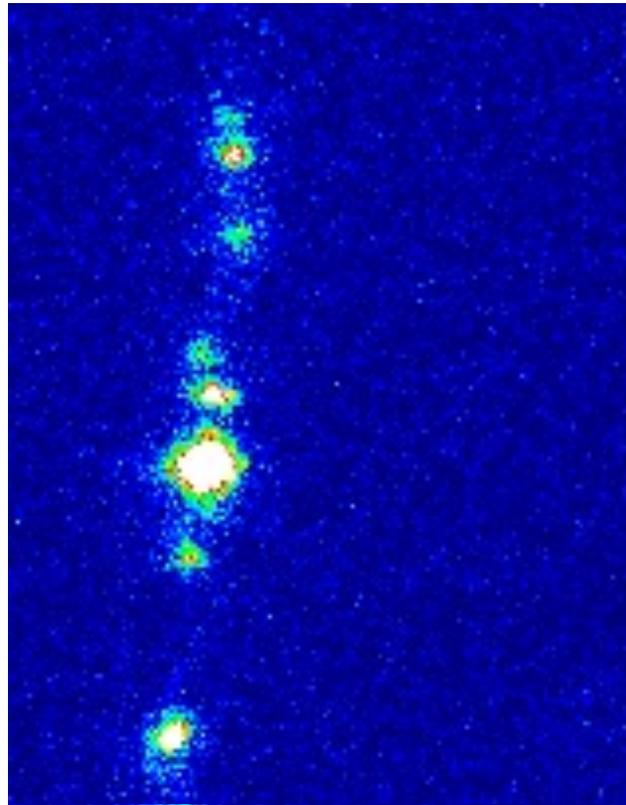$$A \sim \sum e^{i\mathbf{Q}\cdot(\mathbf{R}_j + \mathbf{u}_j)}$$
$$\approx \sum e^{i\mathbf{Q}\cdot\mathbf{R}_j}(1 + i\mathbf{Q}\cdot\mathbf{u}_j)$$

Imaginary density

Ferritin (111) Powder Ring

- 50 frames
- 30sec exposure
- 0.3sec playback
- 150x200 pixels
of 22.5μm

I. K. Robinson, The Full Spectrum, Sept 2006

Critical Nucleation

- a) Nucleation via planar 2D clustering
- b) Nucleation via compact close-packed 3D clustering
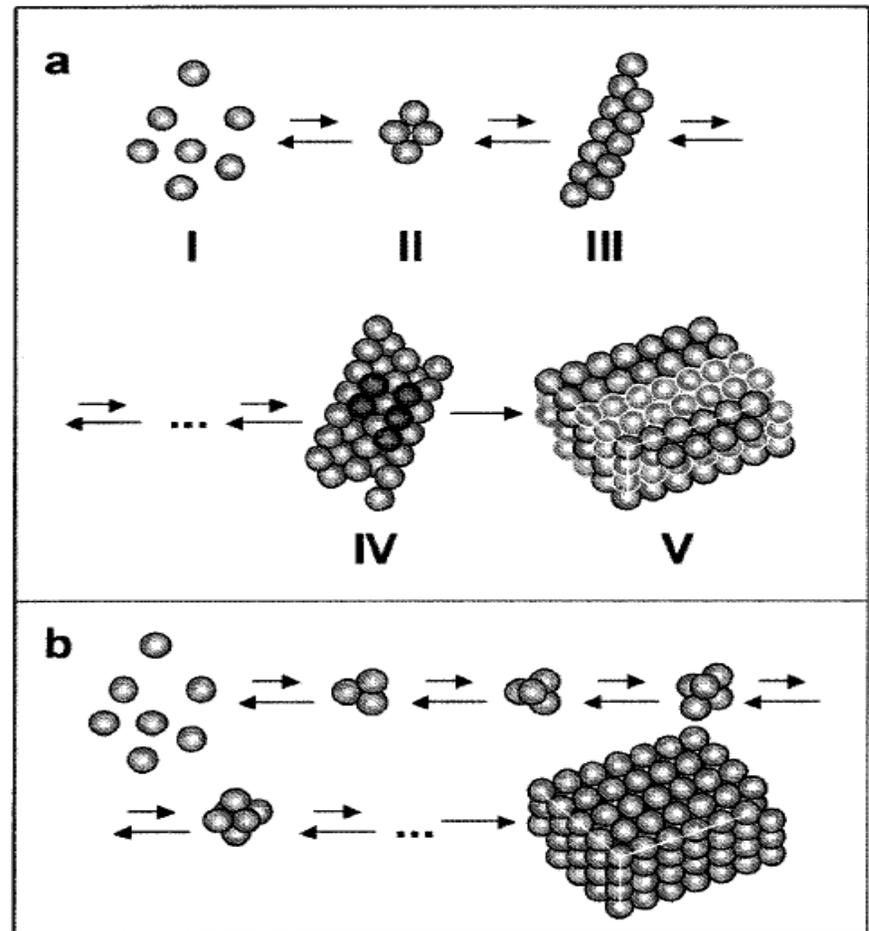- Possible also via non-crystalline (eg icosahedral) nuclei.

Figure 10. Schematic illustration of two nucleation pathways: (a) via a planar critical cluster (in IV molecules belonging to the second layer are shown in a lighter shade; in V the (110) layers that stack up to form this crystal are delineated by lighter and darker contours) and (b) via compact critical cluster.[5,6]

I. K. Robinson, The Ful
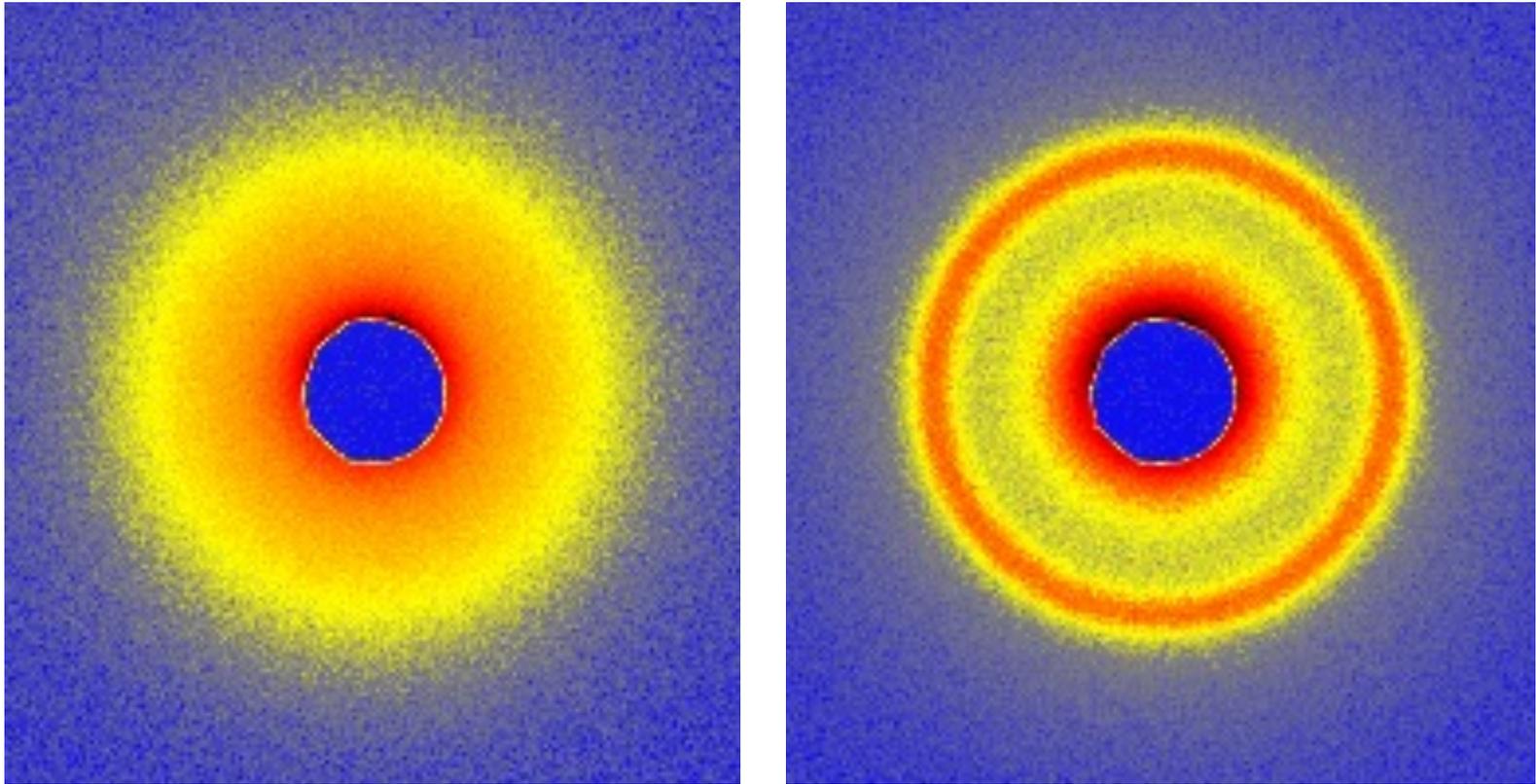
In-situ Study of Crystallization
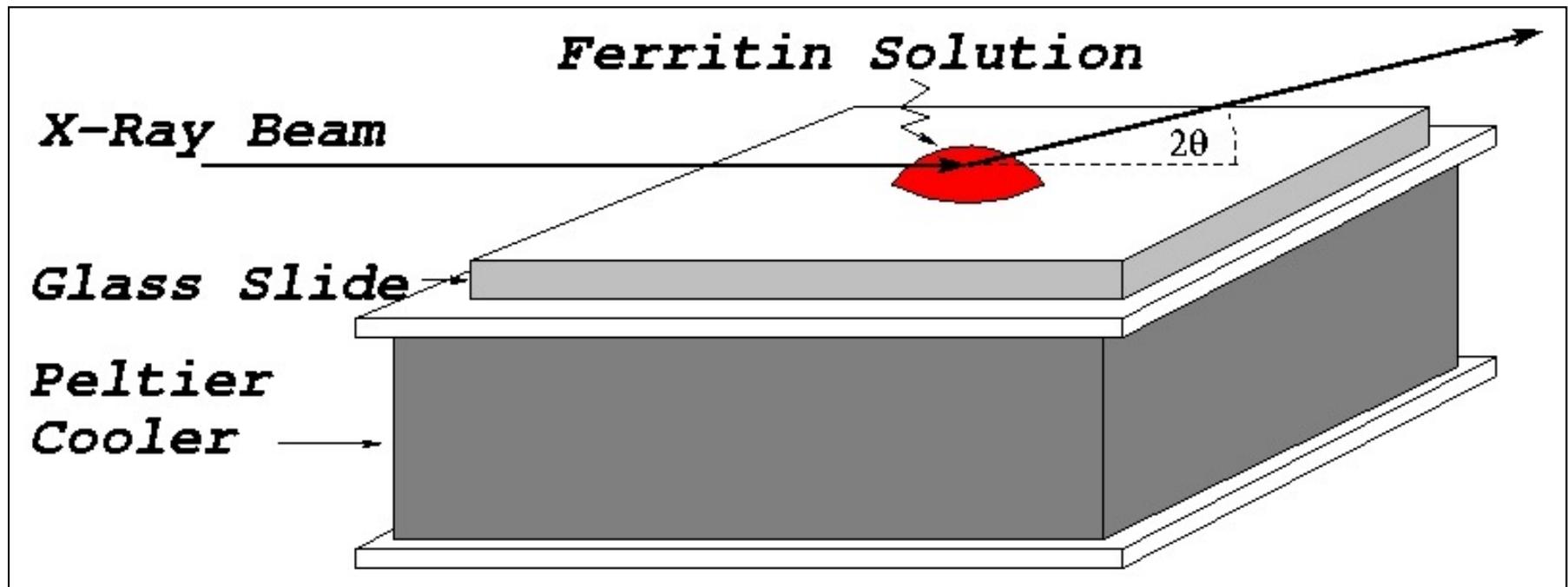
Crystallization of Holoferritin: SAXS
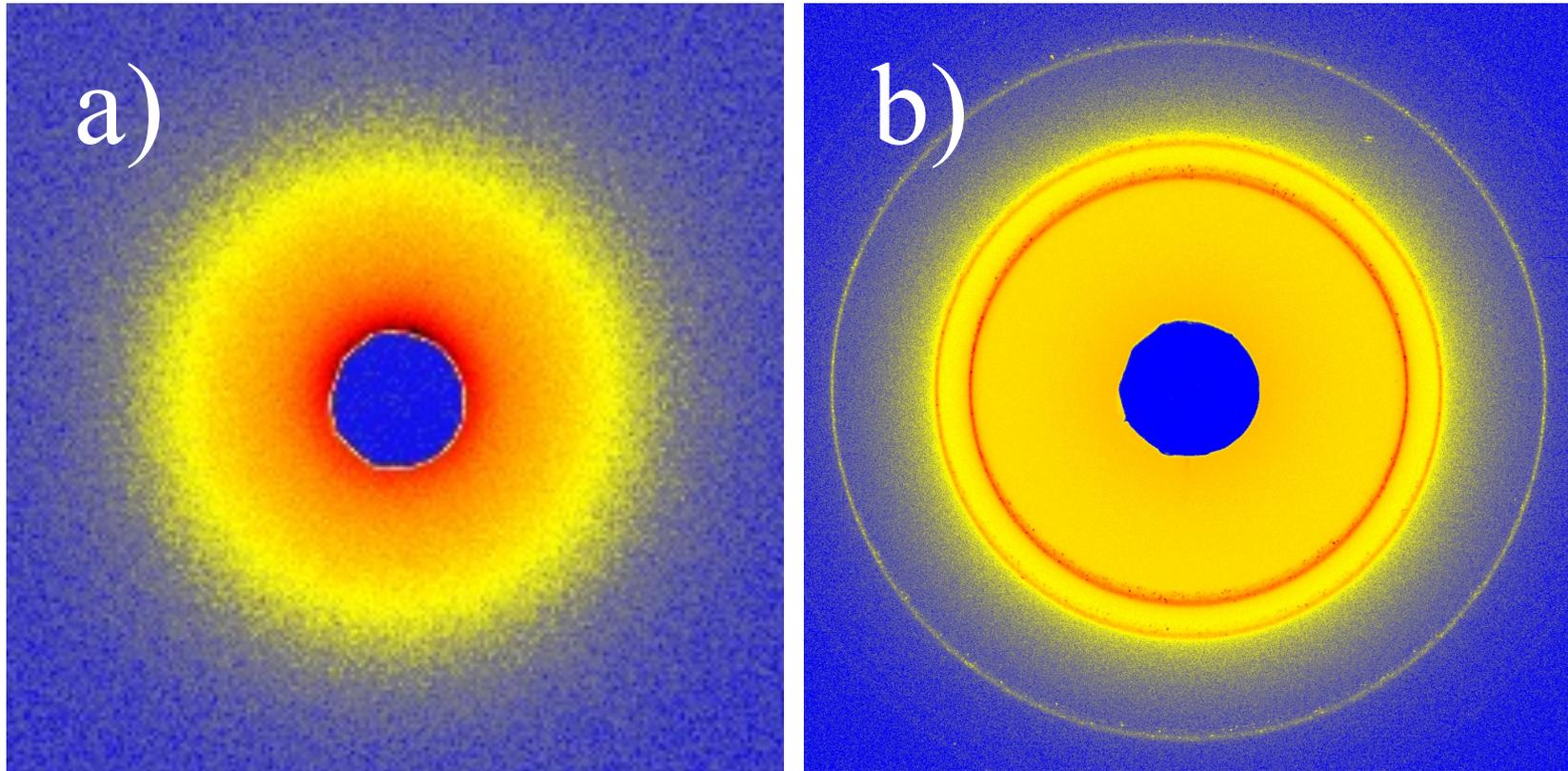
a) SAXS pattern of holoferritin at 10°C.
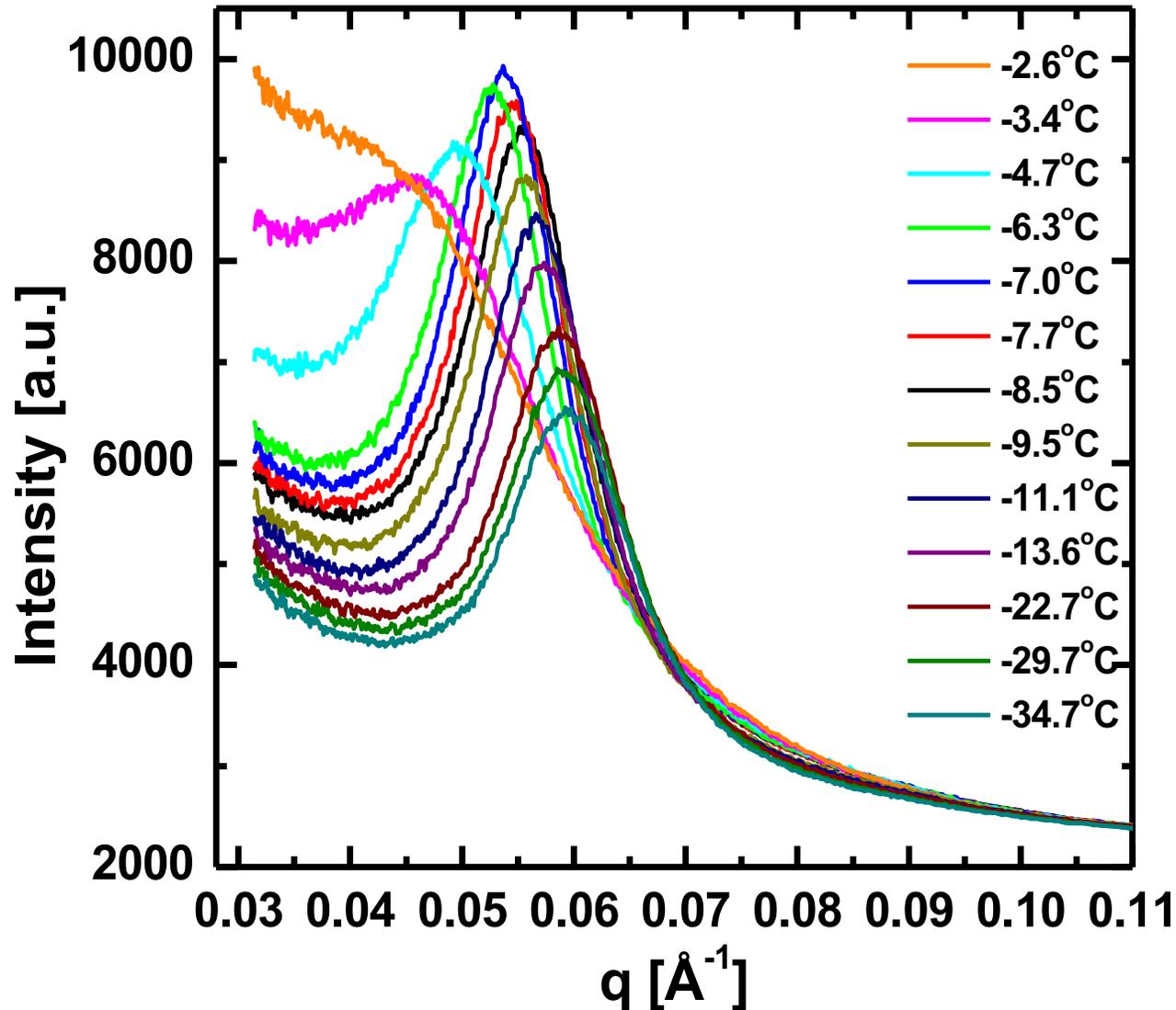
b) SAXS pattern after adding Cadmium salt

Effect of Temperature
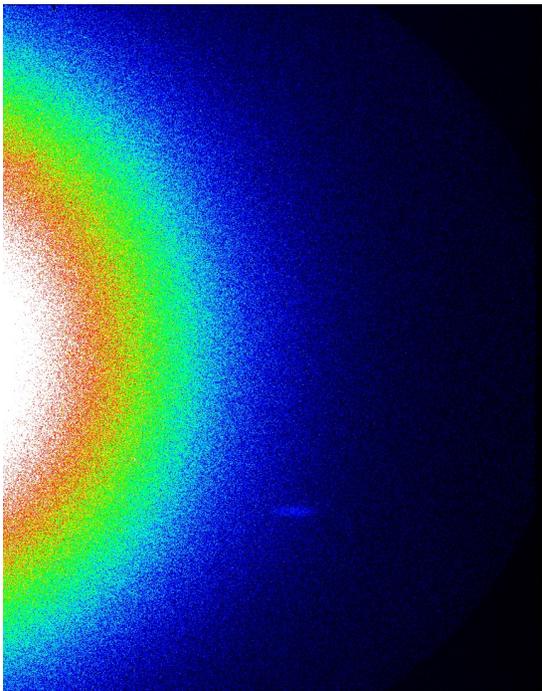

SAXS pattern at T=+10ºC and -10ºC.

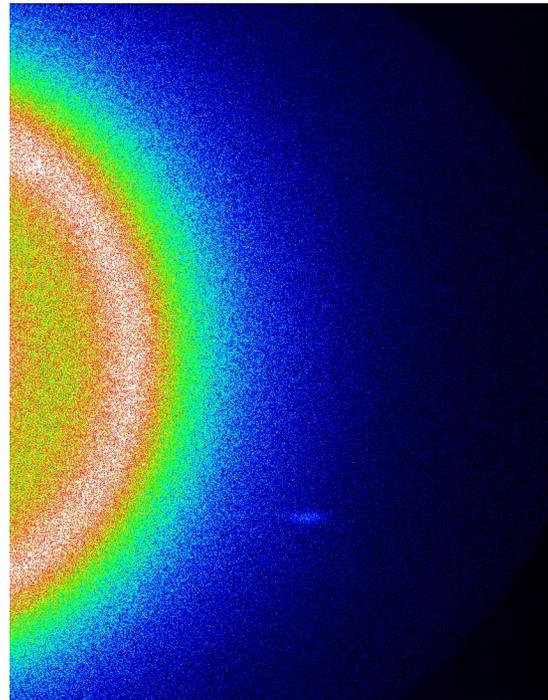Holoferritin in dilute (10mg/ml) NaCl only.

Integrated SAXS Data

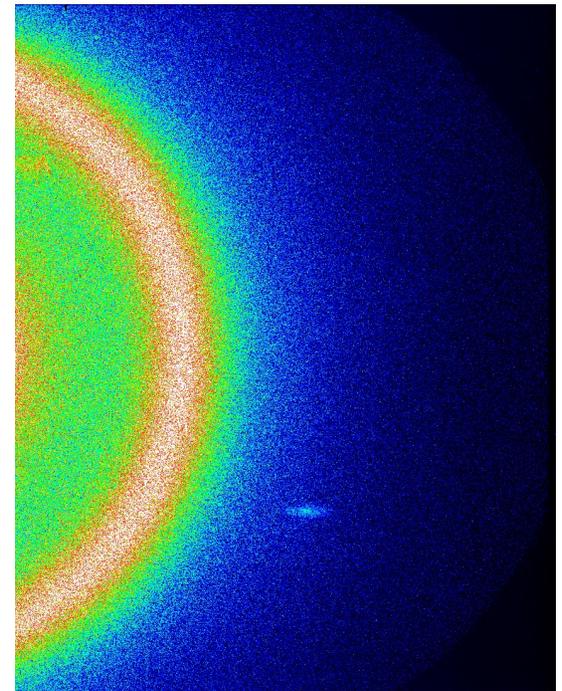- The SAXS pattern shows a broad peak which shifts in position and changes in width as a function of temperature

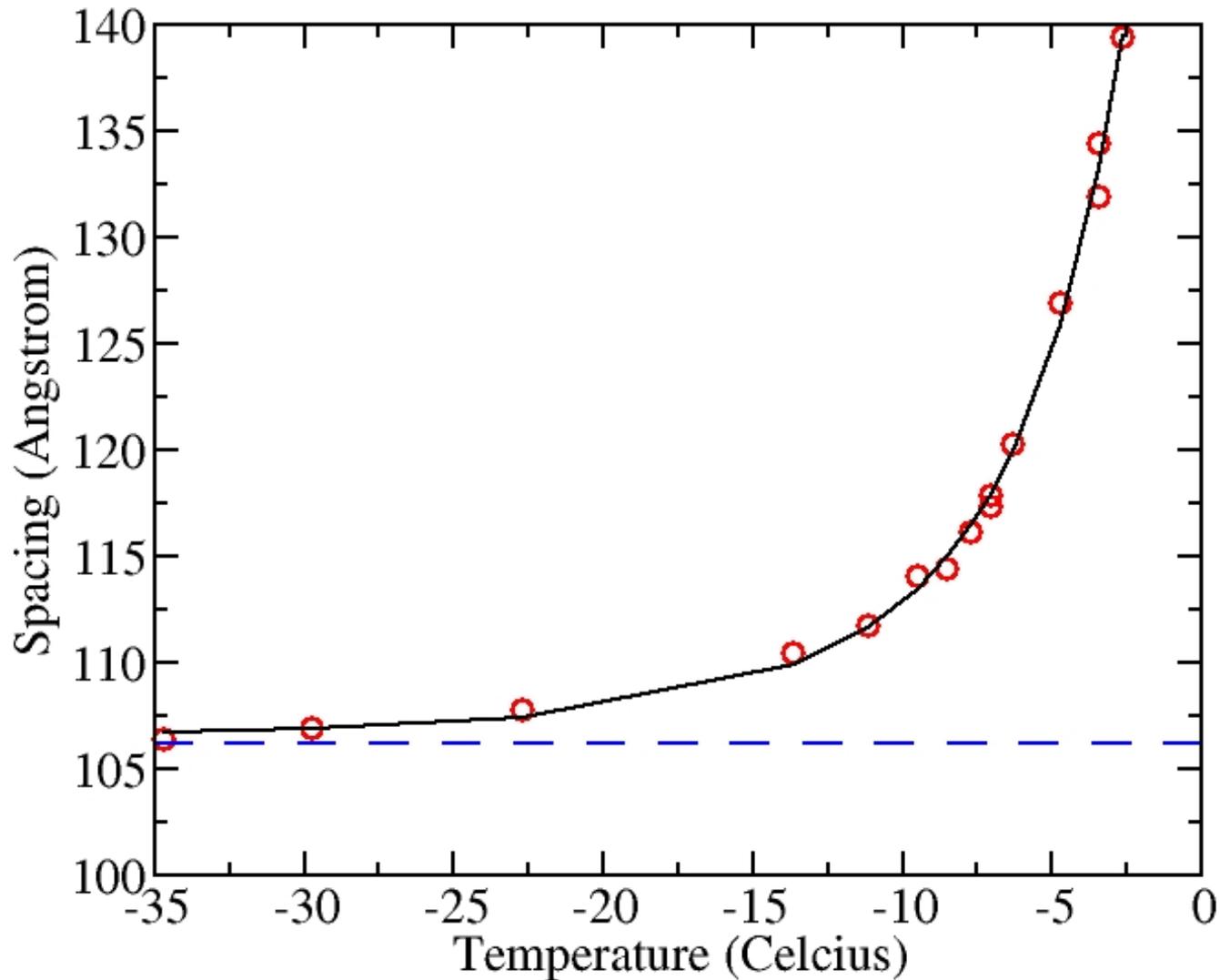# Ferritin Solution upon Freezing
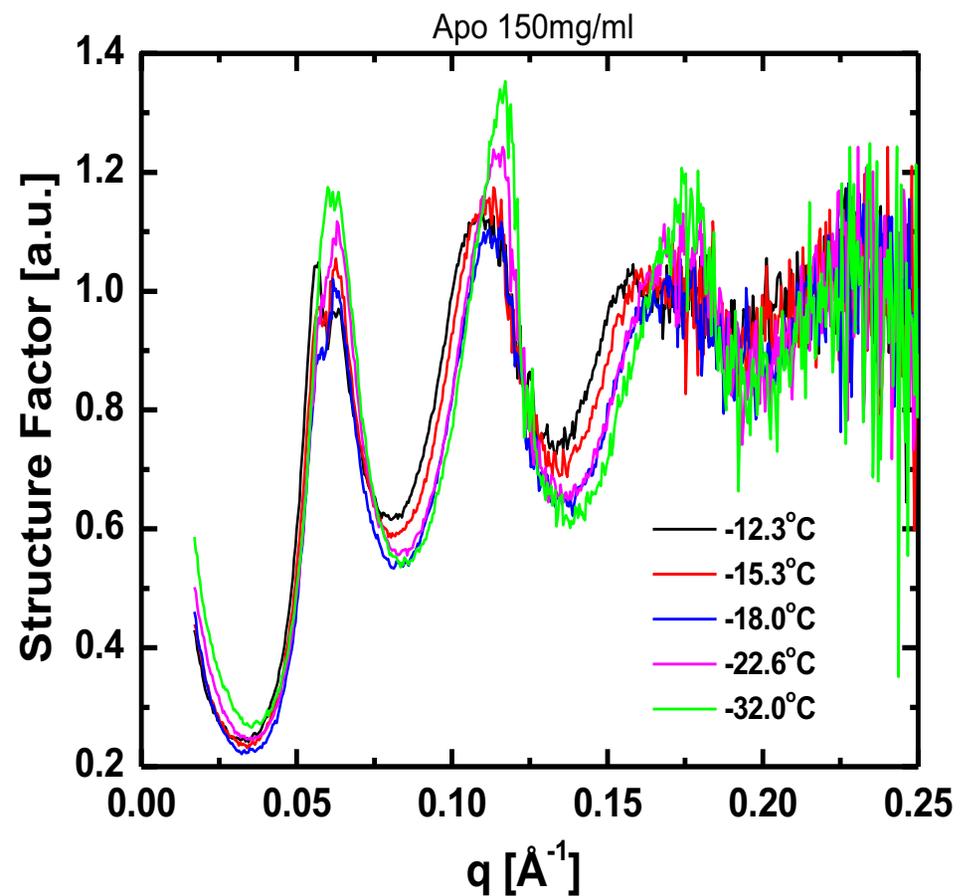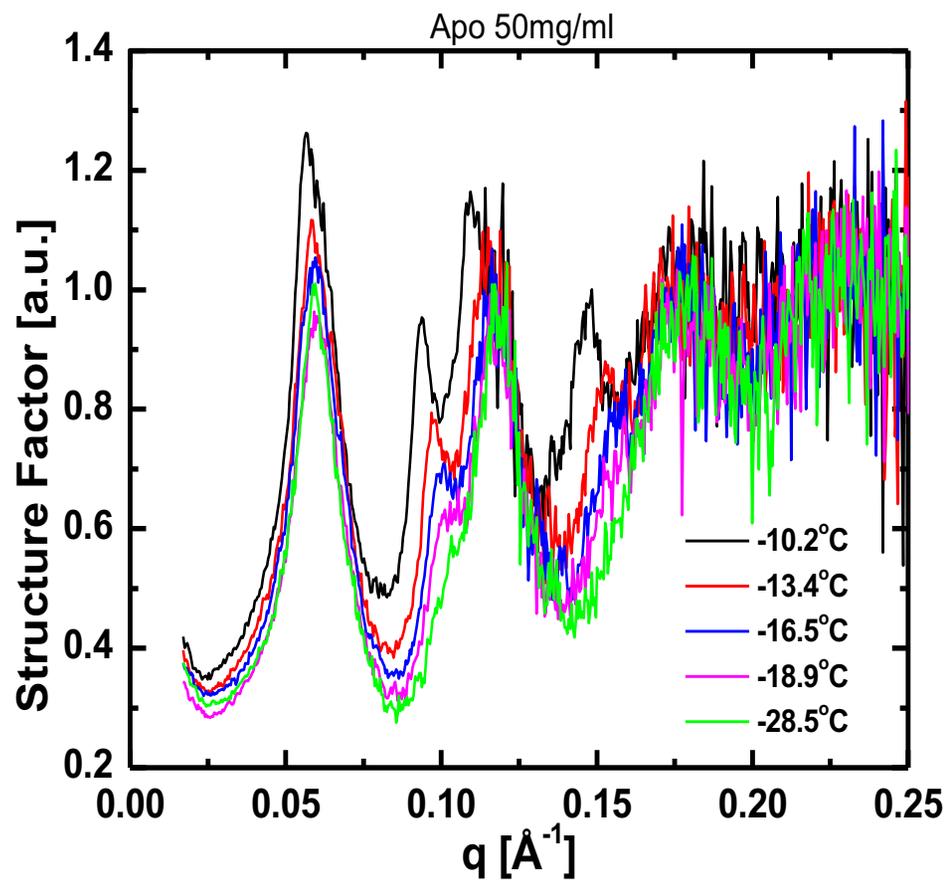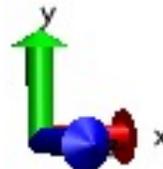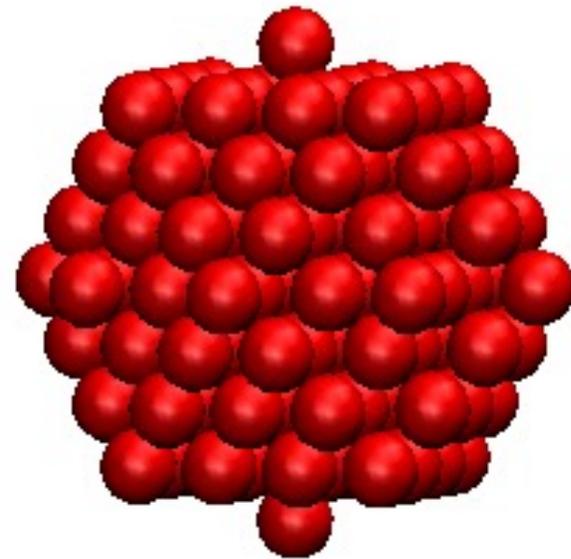

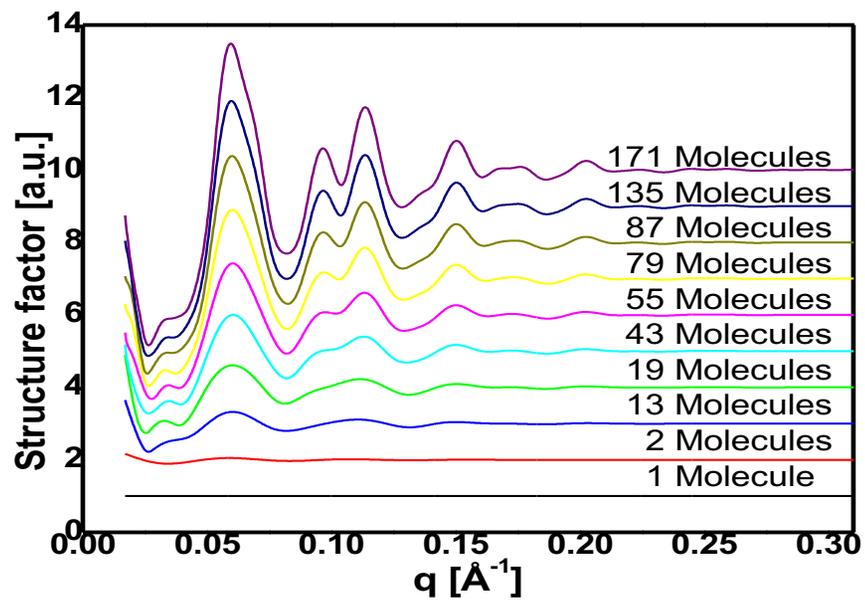T=13.3   T=-4.7   T=-34.7

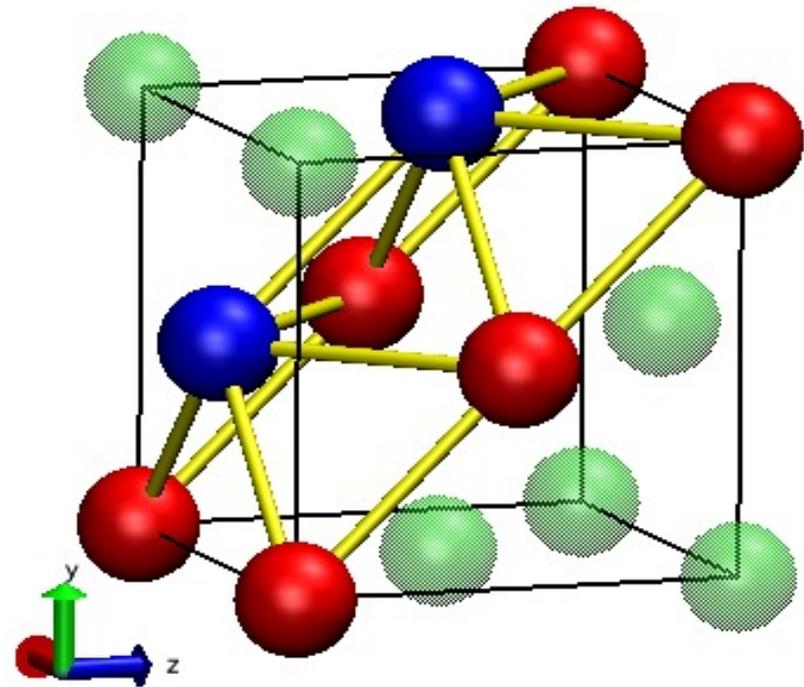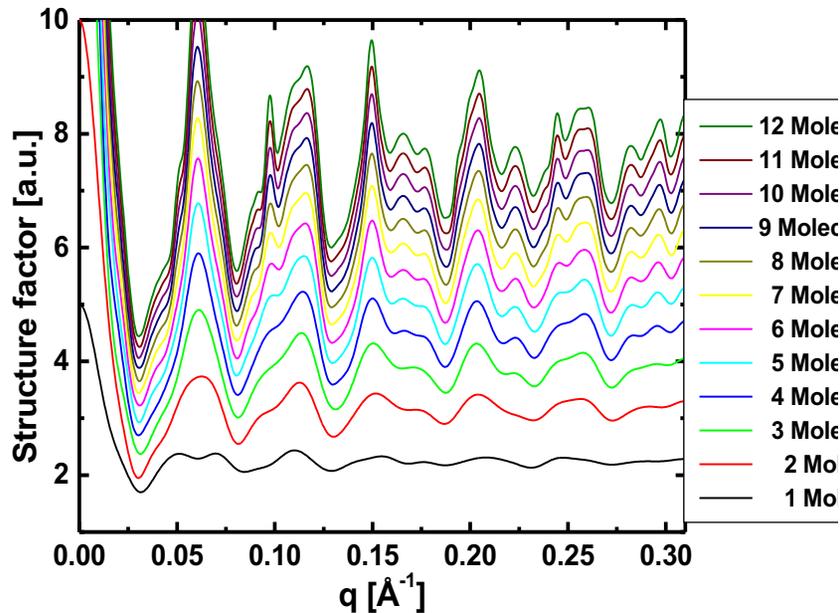# Temperature Variation of Peak Position

Structure Factors of Apoferritin

FCC Spherical Clusters

Planar Clusters of 110 Rods

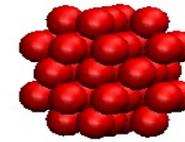## Structure factor simulation as 5 <110> rods with 1 to 12 molecules per rod

Best Fits

T=–10ºC
Cluster Size = 36
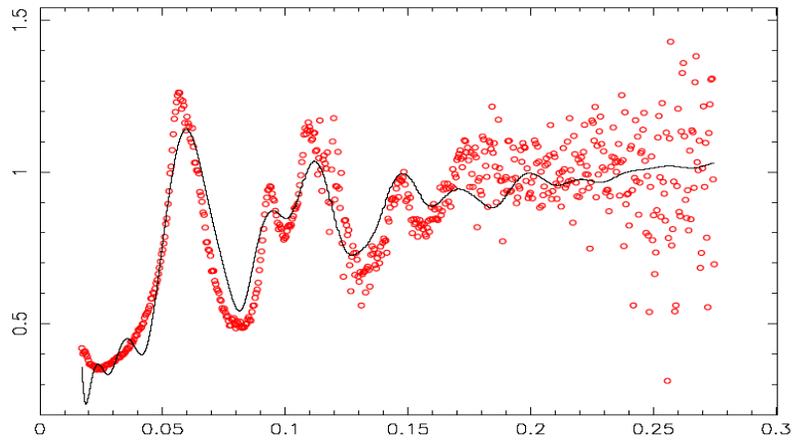
T=–13ºC
Cluster Size = 63
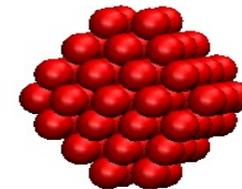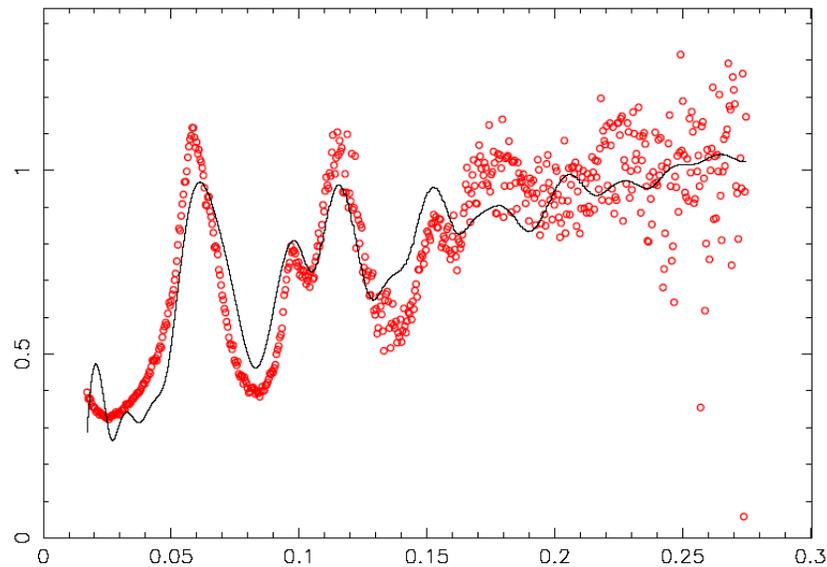
New routes to structure?

- Cryo-EM
  - real-space image, but not dose-efficient
- Inversion of diffraction
  - dose-efficient, especially for phase contrast
  - about equally efficient for photons or electrons
- How to beat the dose limit?

Do it faster!

How fast must we go?

- Radiation breaks bonds and ionizes matter
- Inertial response of nucleus to broken bond
- Structure is 'lost' when an atom moves by one bond-length
- Phonon vibration time: 10^{14} Hz = 10fs

- Photons better than electrons (space charge)
- Possible with X-ray free-electron laser (XFEL)

Molecular Movies using XFEL
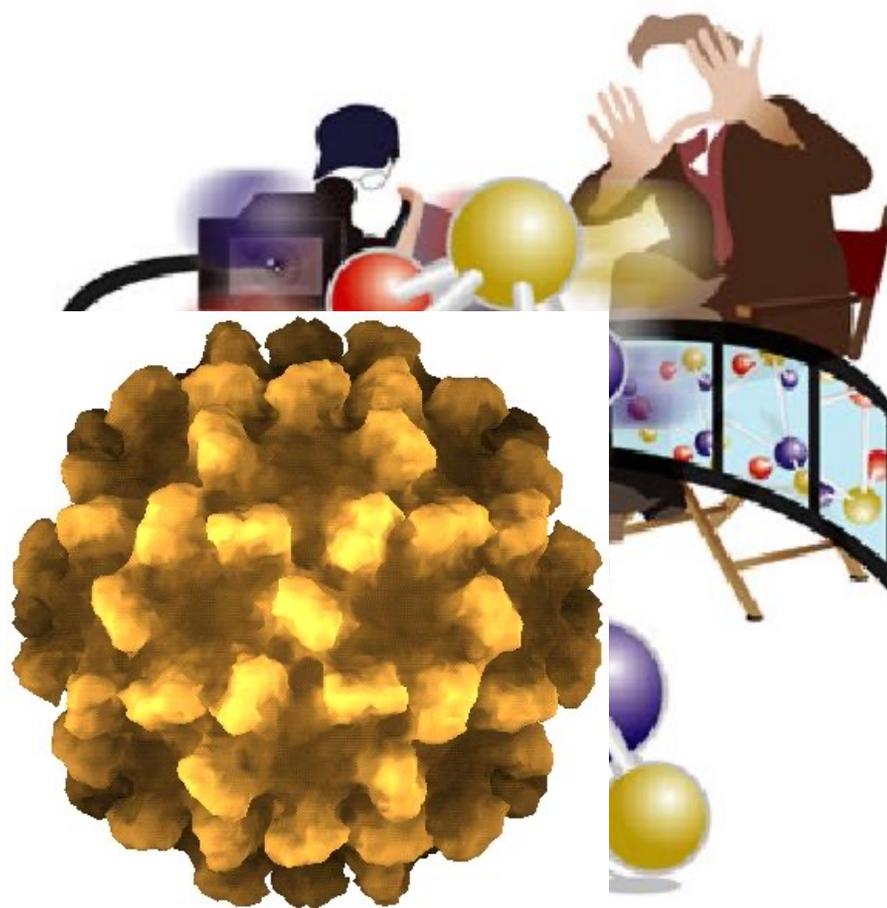
Conclusions and Outlook

- Internal structure of Au Nanocrystals

- 3D imaging practical for nanocrystals

- Imaging by computation instead of lens

- Strain fields imaged as complex density

- Single molecules using XFEL

Compact Light Source
Ron Ruth, Lyncean Technologies