Single-molecule X-ray Diffraction?

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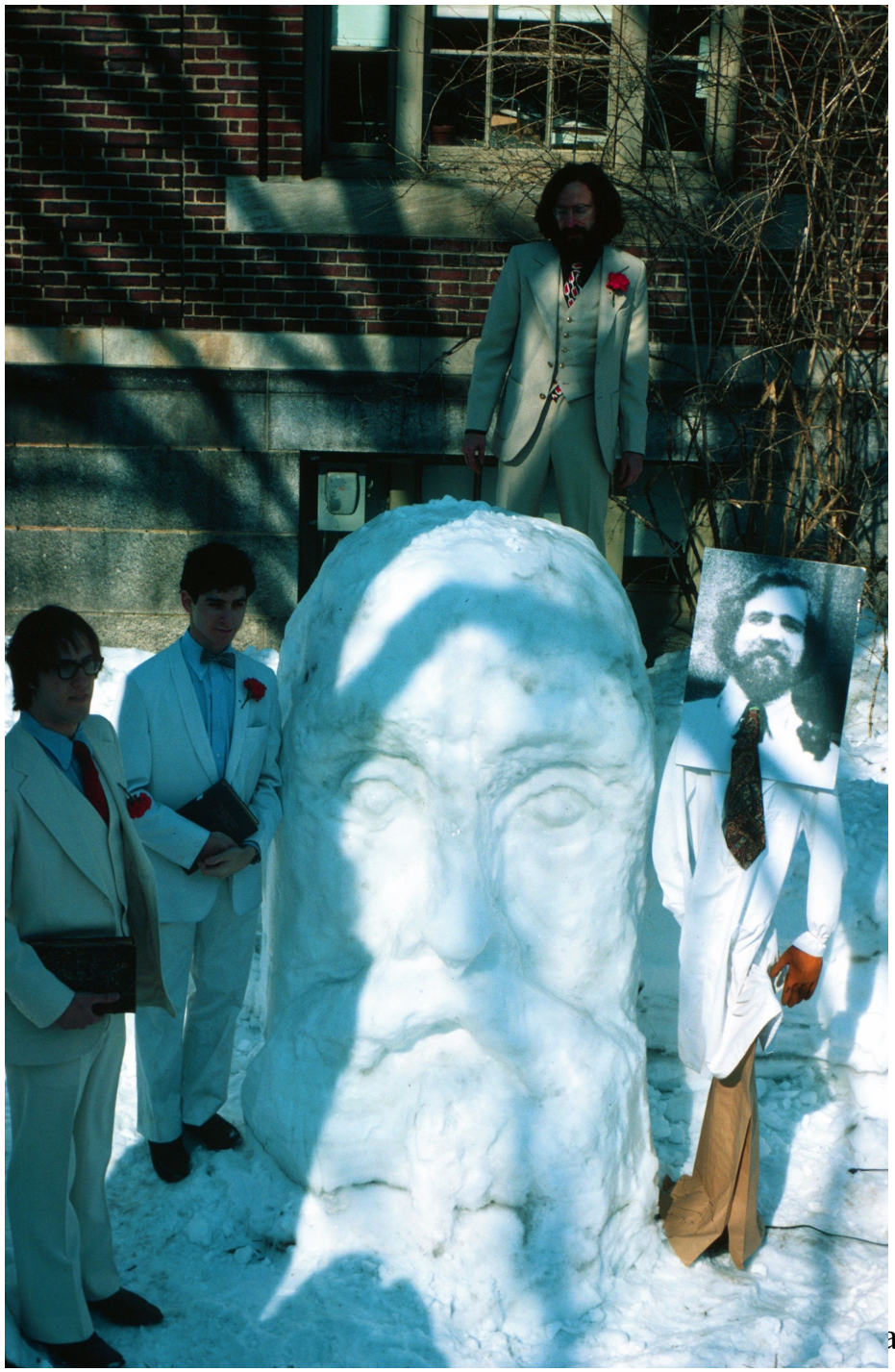


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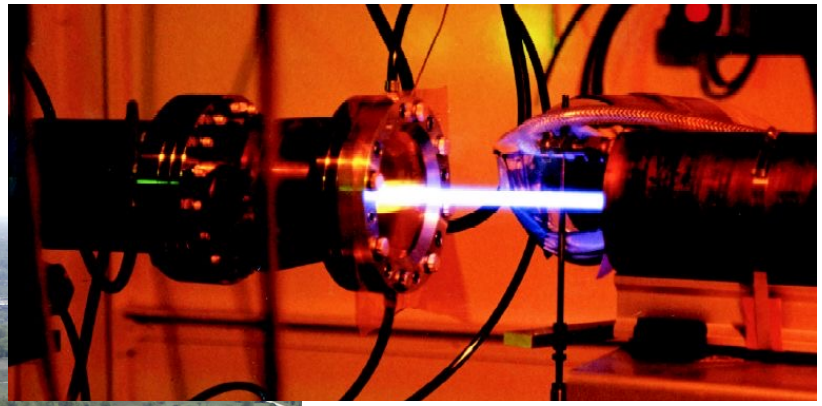
Dos Equis XX 1.00
Harp 1.00
Kirin 1.00

Outline

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

Synchrotron Radiation

Urbana



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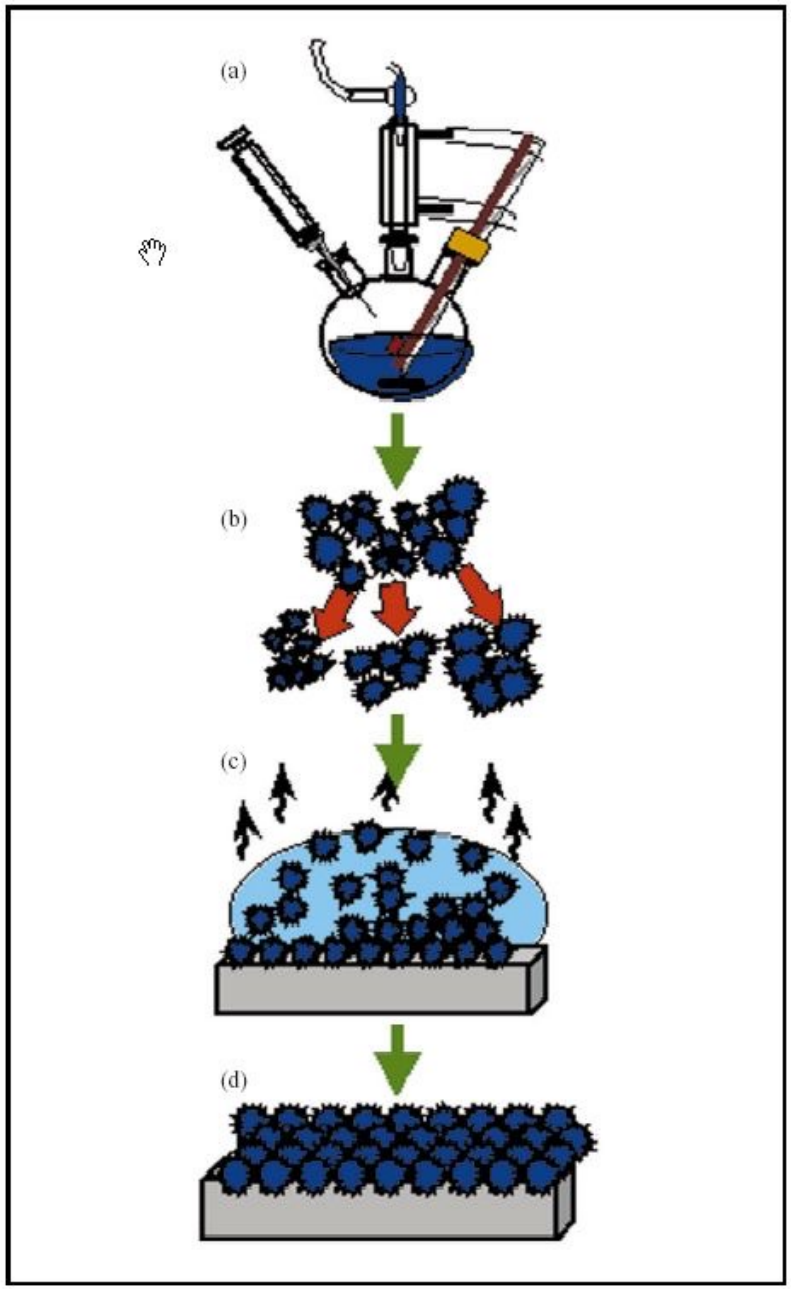
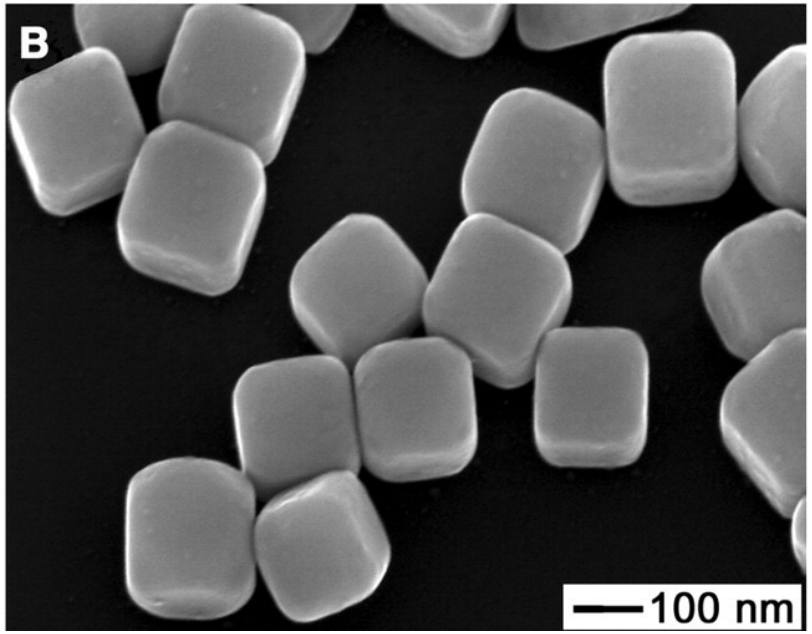
Chemical Synthesis of Nanocrystals

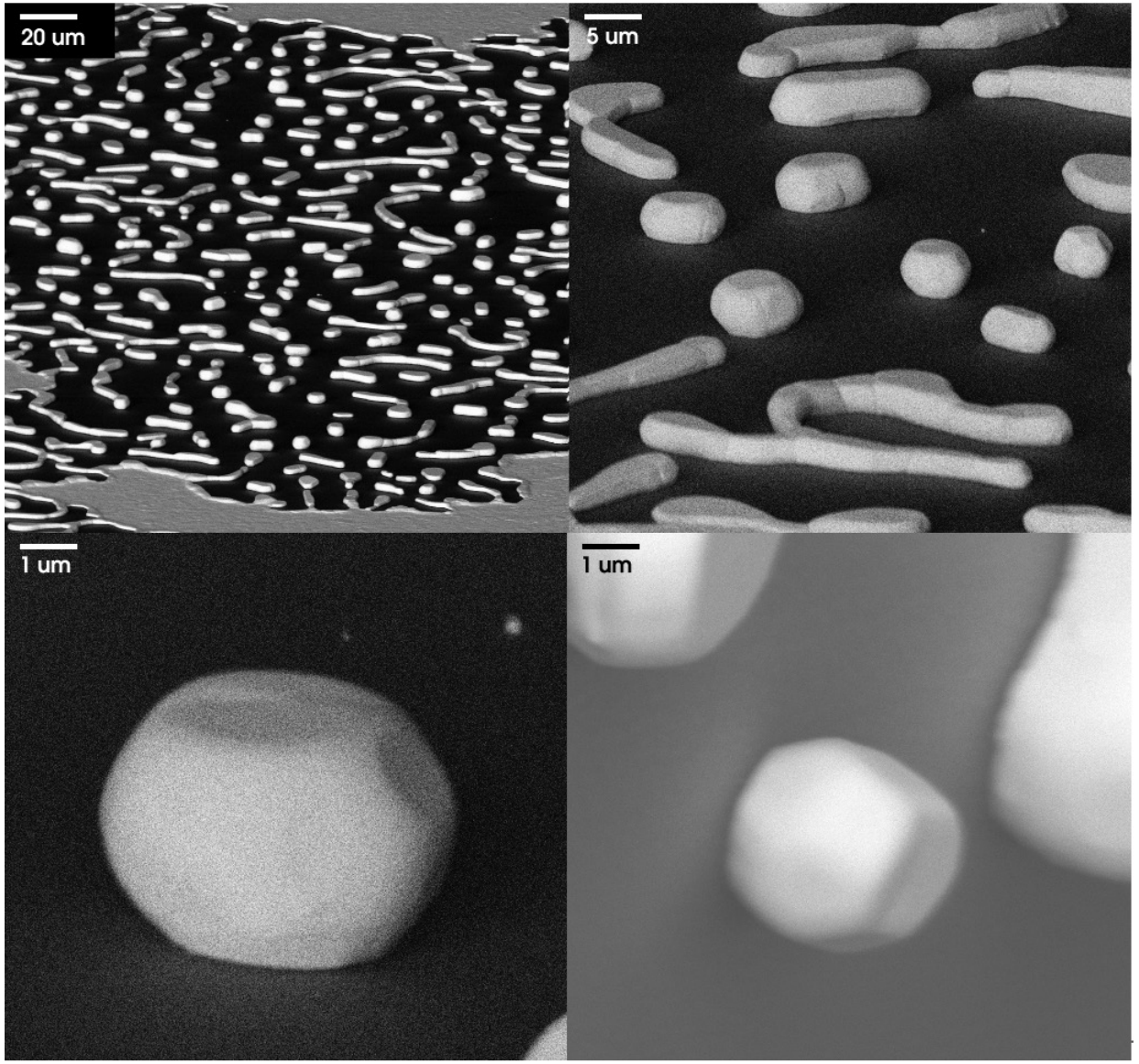


- Reactants introduced rapidly
- High temperature solvent
- Surfactant/organic capping agent

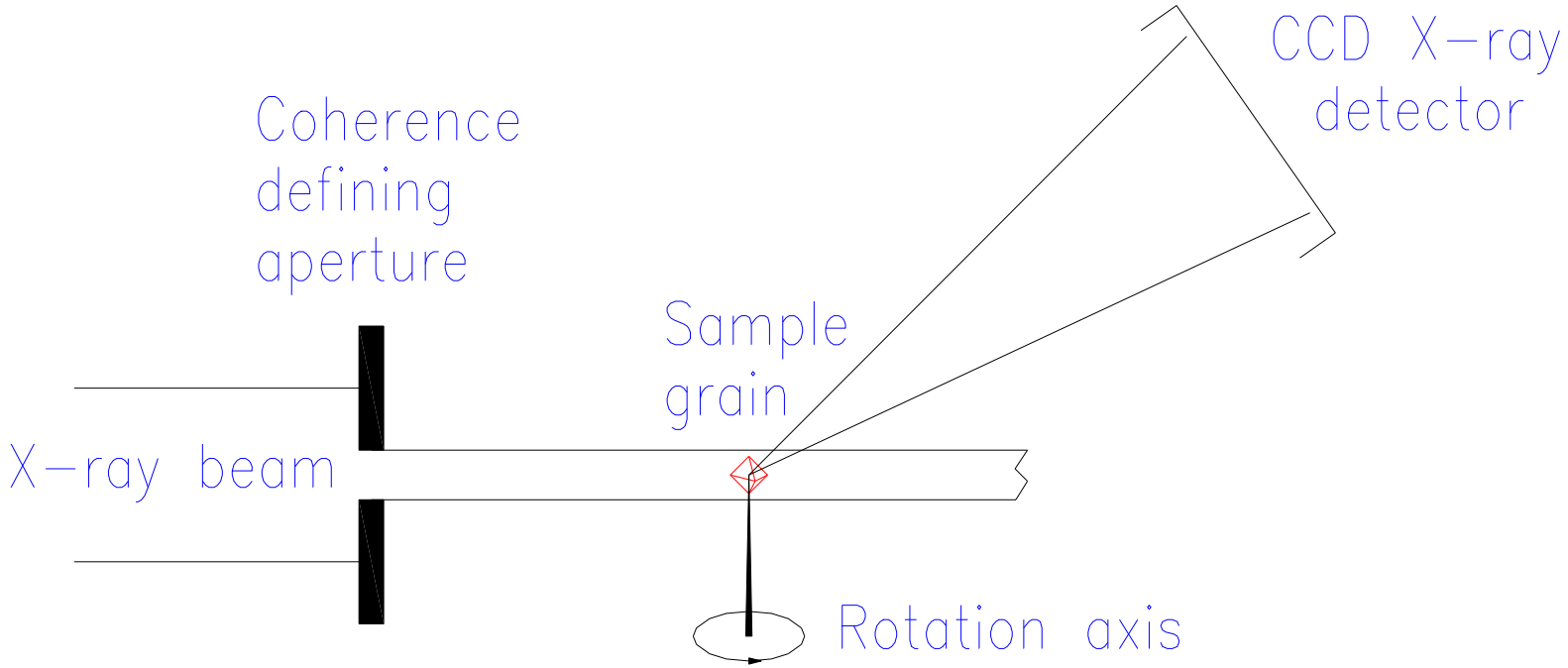


SEMS

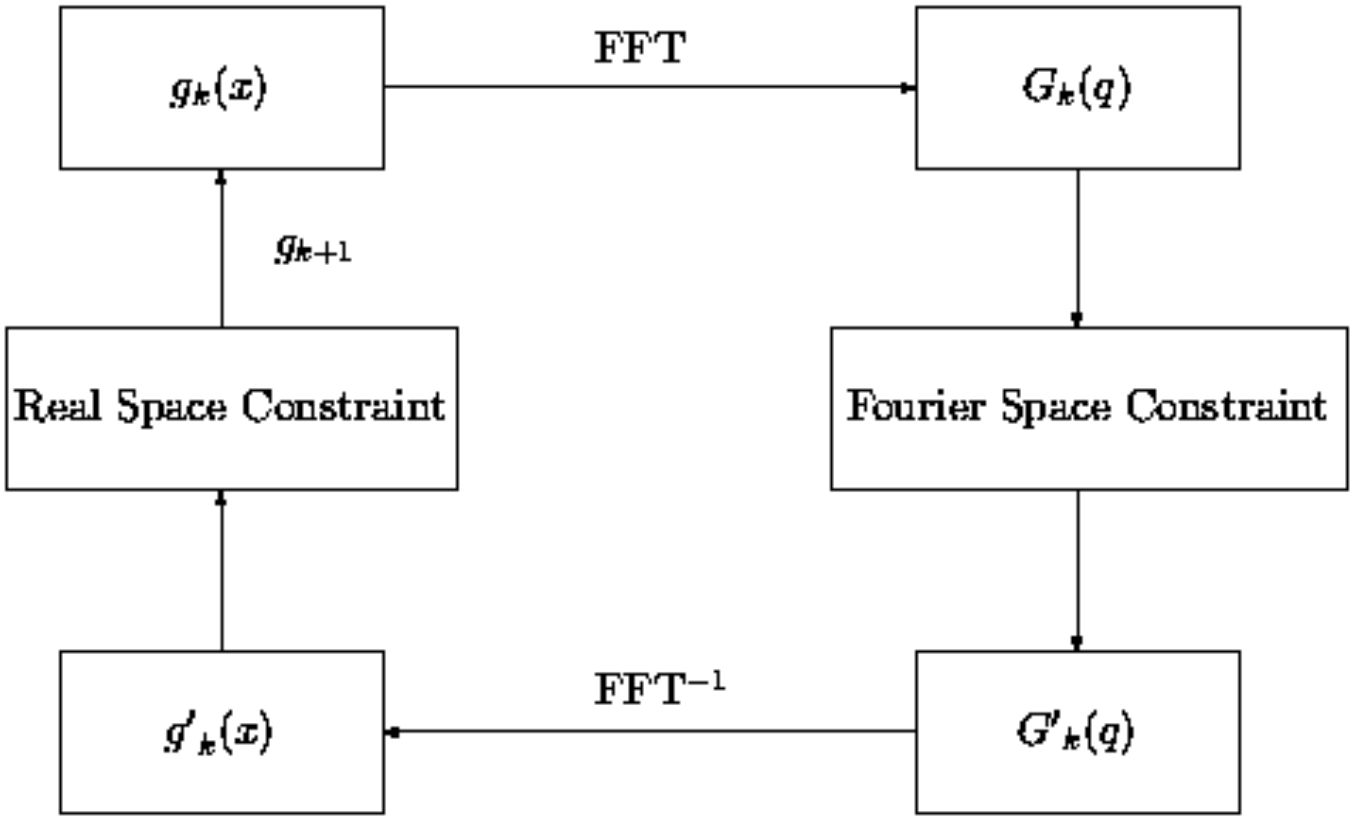
- Au blanket film
- Quartz substrate
- Annealed at 950°C for 70 hrs.



Lensless X-ray Microscope



Generic "Error Reduction" method



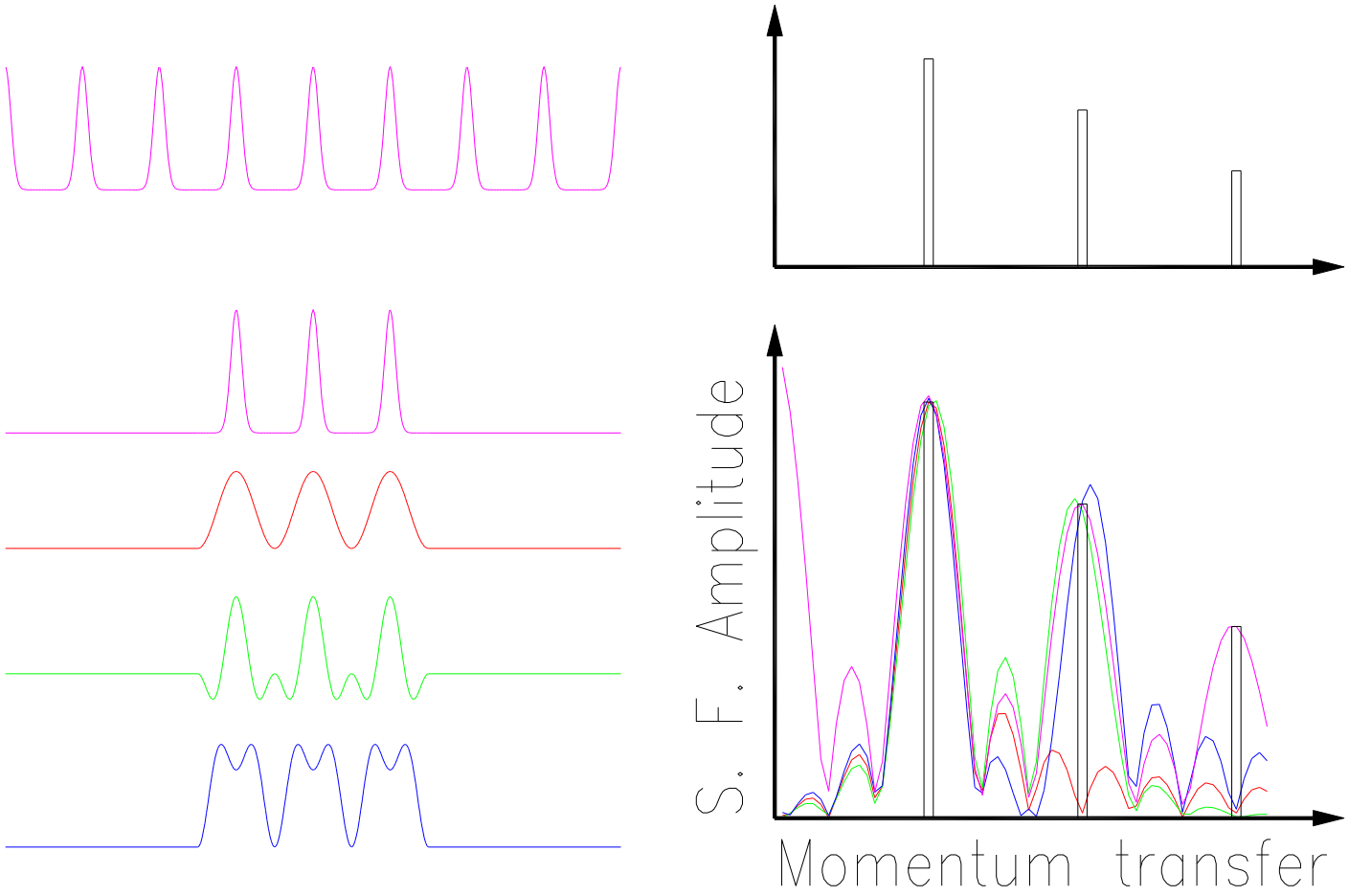
J. R. Fienup Appl. Opt. 21 2758 (1982)
R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)

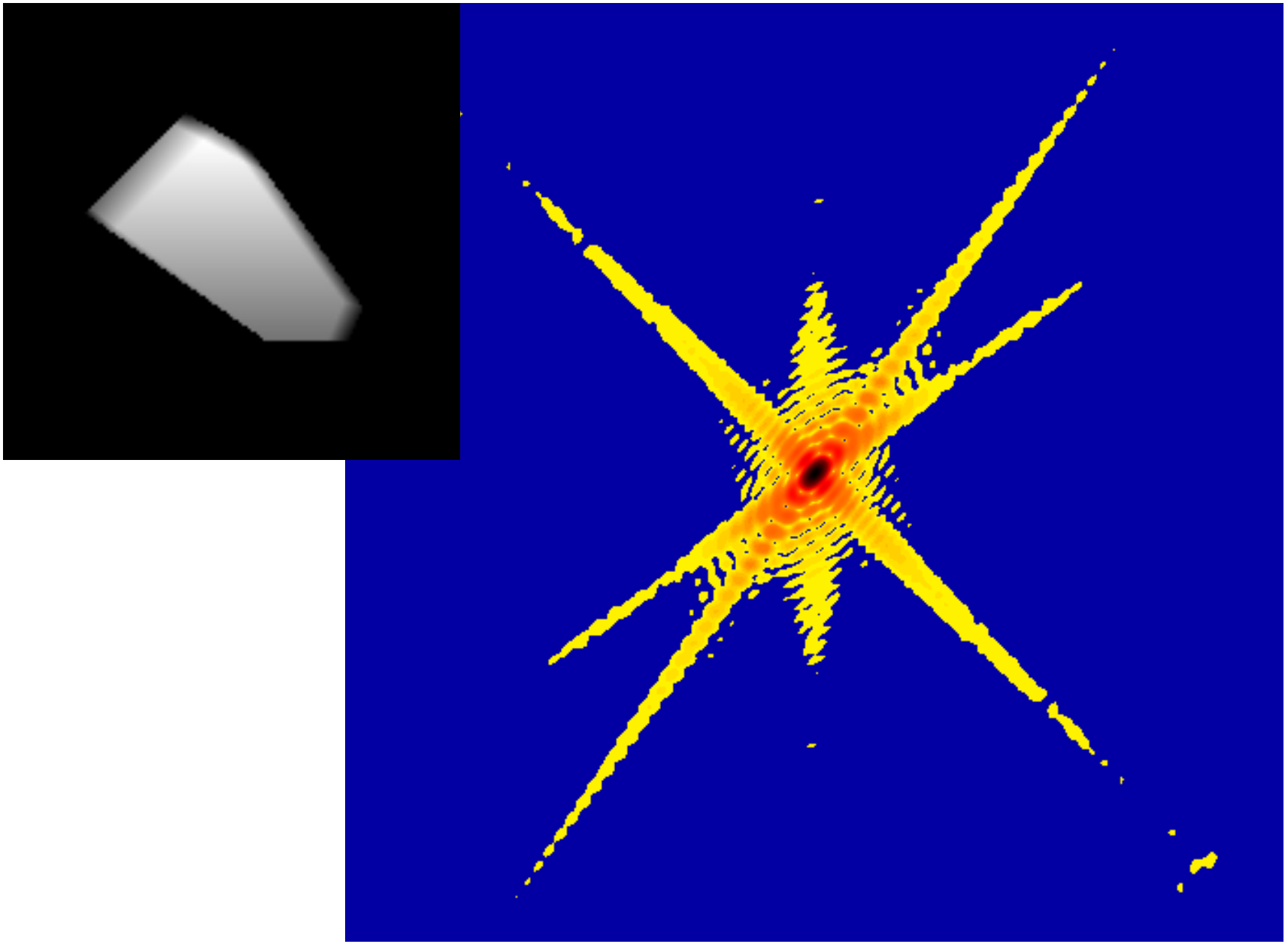
Real-space Constraints in Crystallograhy

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

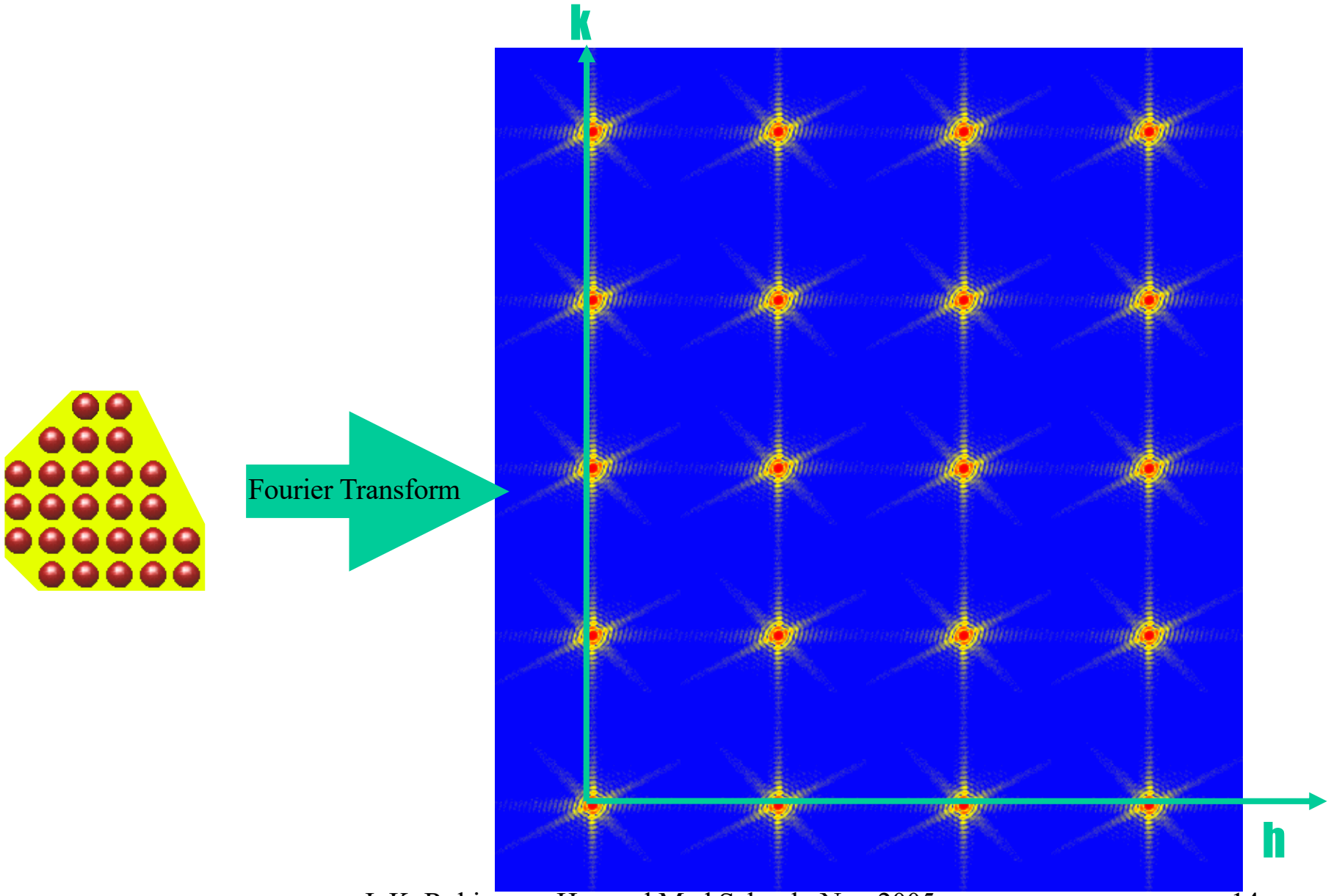
- 'Positivity' and 'Atomicity' constraints (Sayre)
- Finite support, molecular envelope
- Solvent flattening/Molecular replacement
- Non-crystallographic symmetry
- Non-uniqueness is 'pathologically rare' (d>1)
- Uses memory to avoid stagnation (Fienup HIO)

Phase Problem: Finite-size Effect



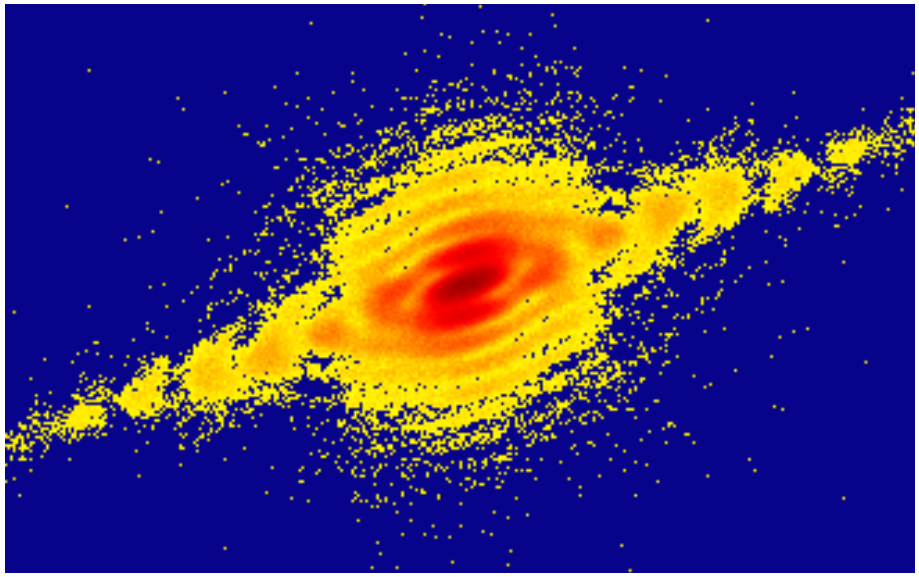


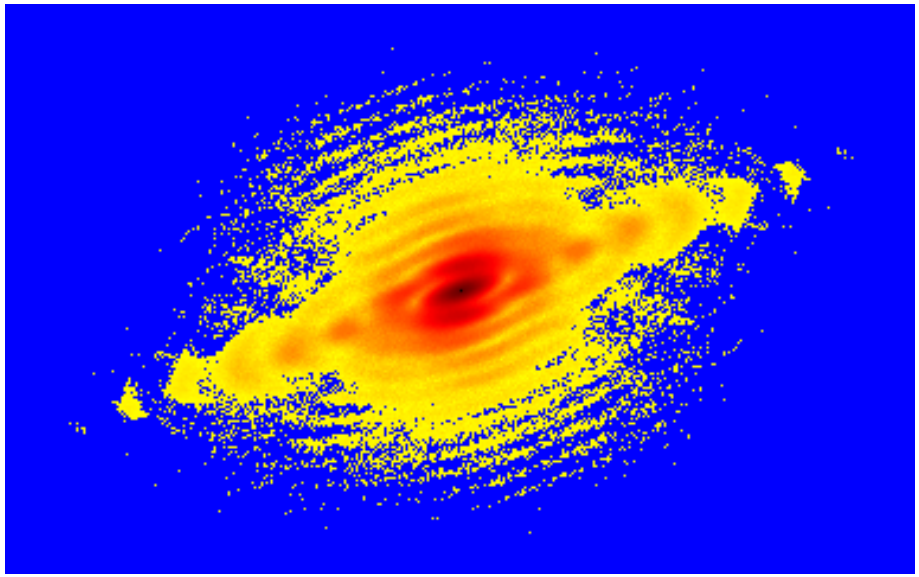
Coherent Diffraction from Crystals

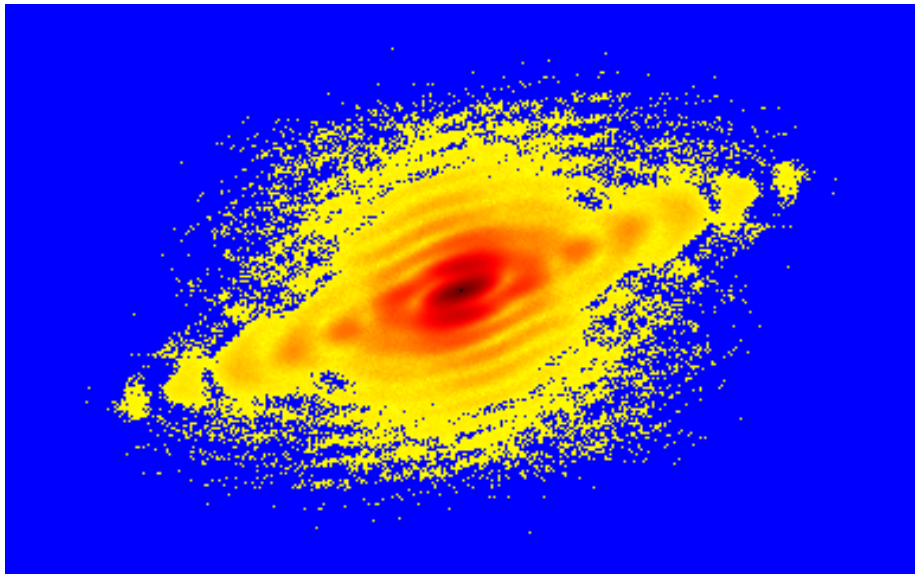


Symmetrized Data and two best fits

Chisq=0.0005

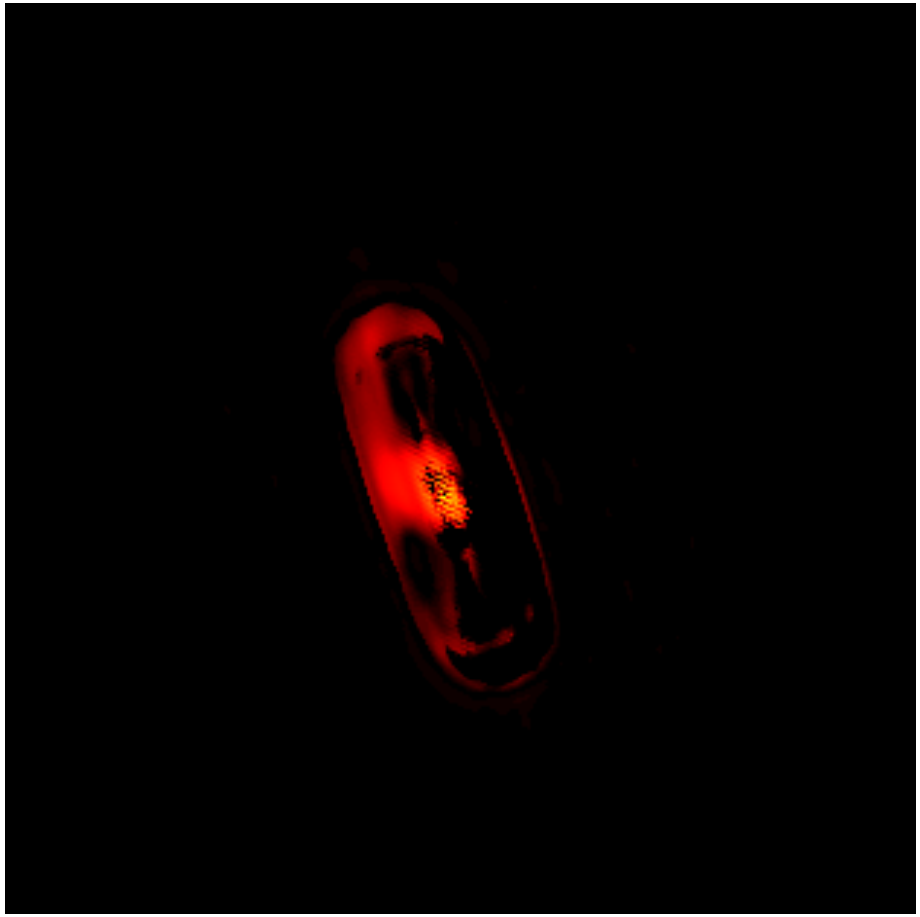


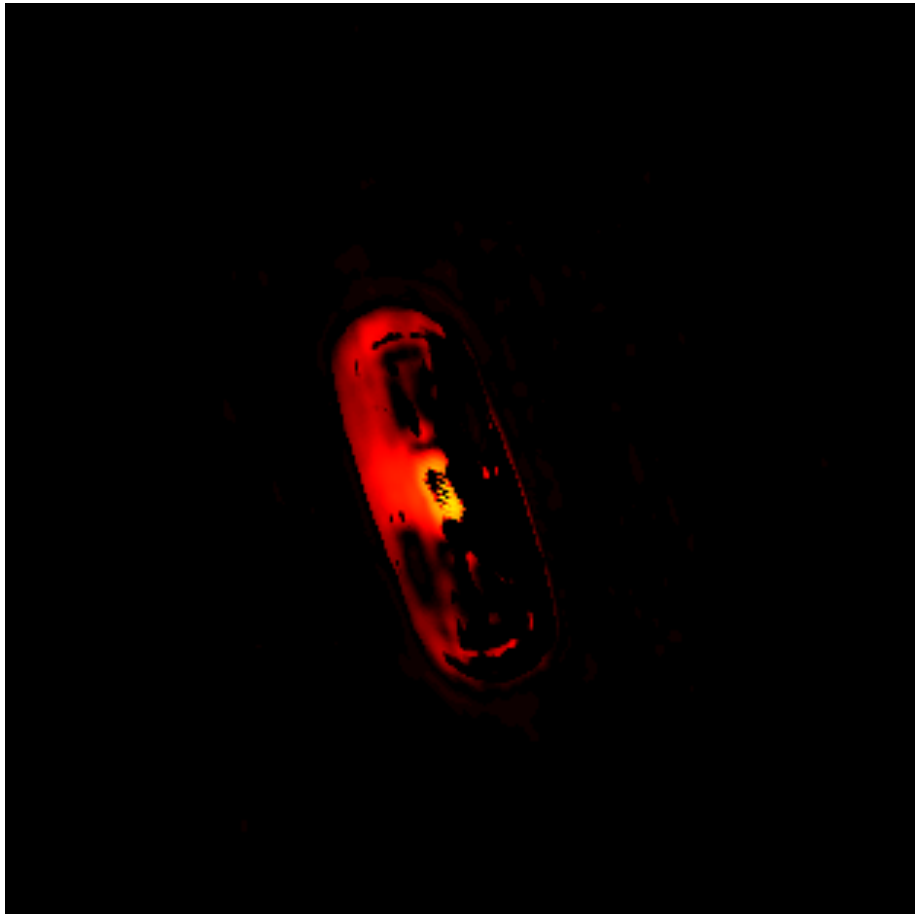




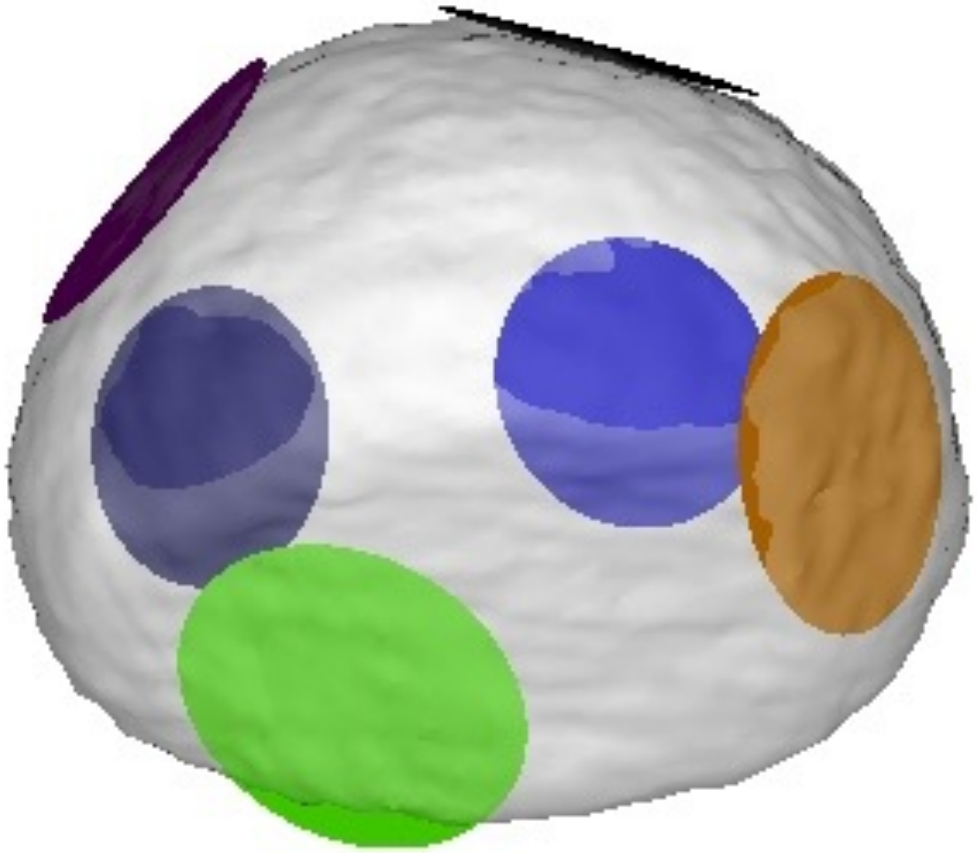
2D Reconstructions

chisquare = 0.0005

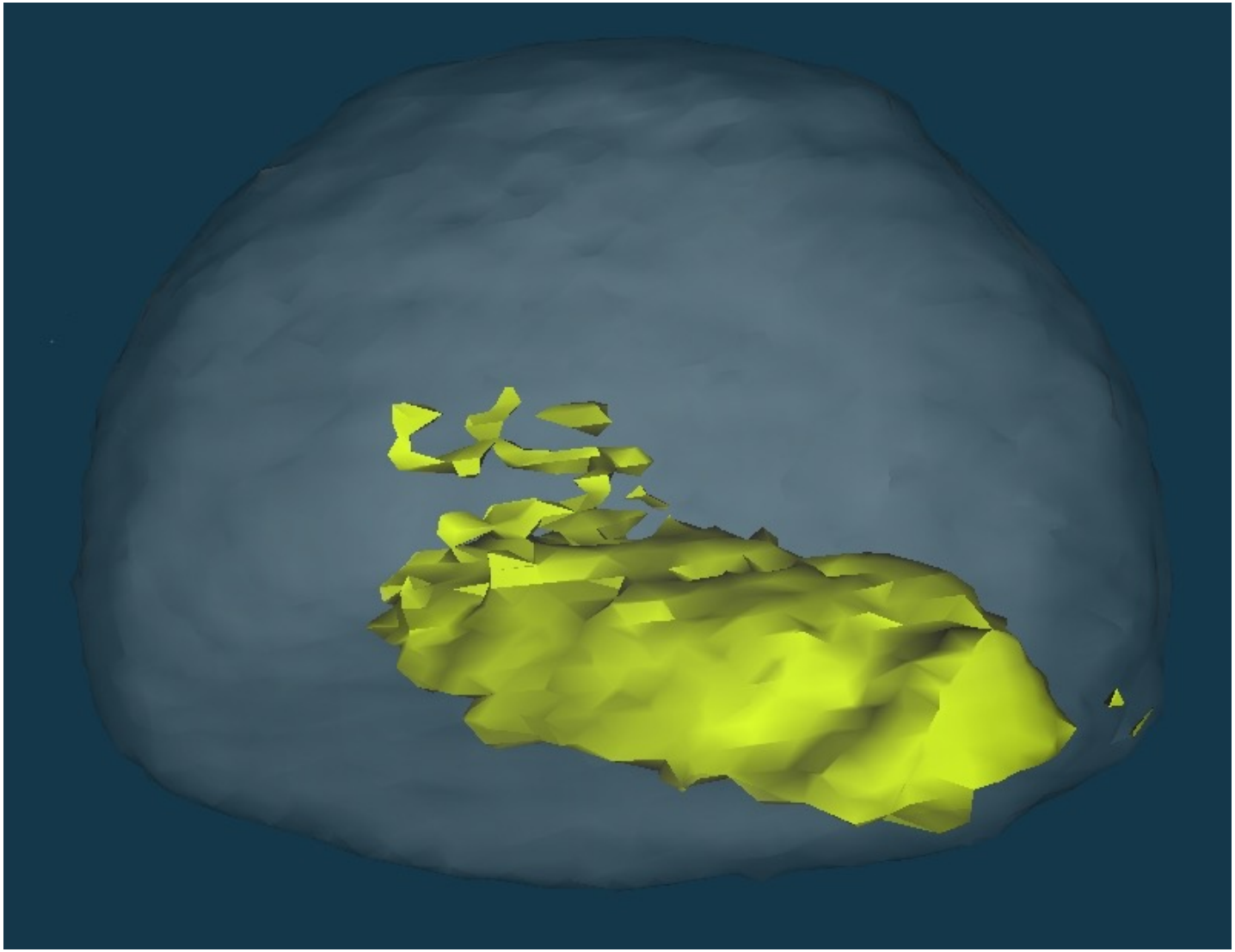




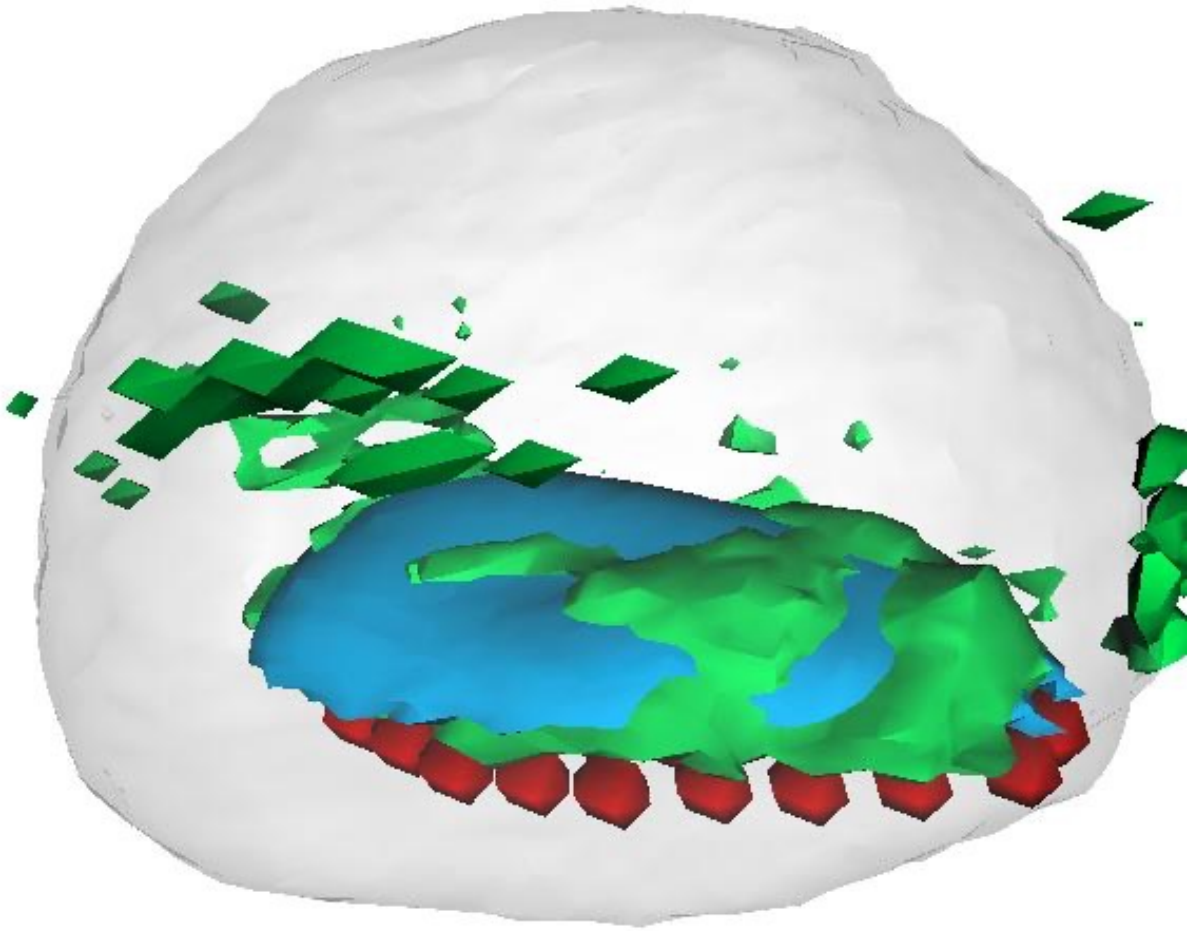
Facets of Equilibrium Crystal Shape



Real space image in phase too

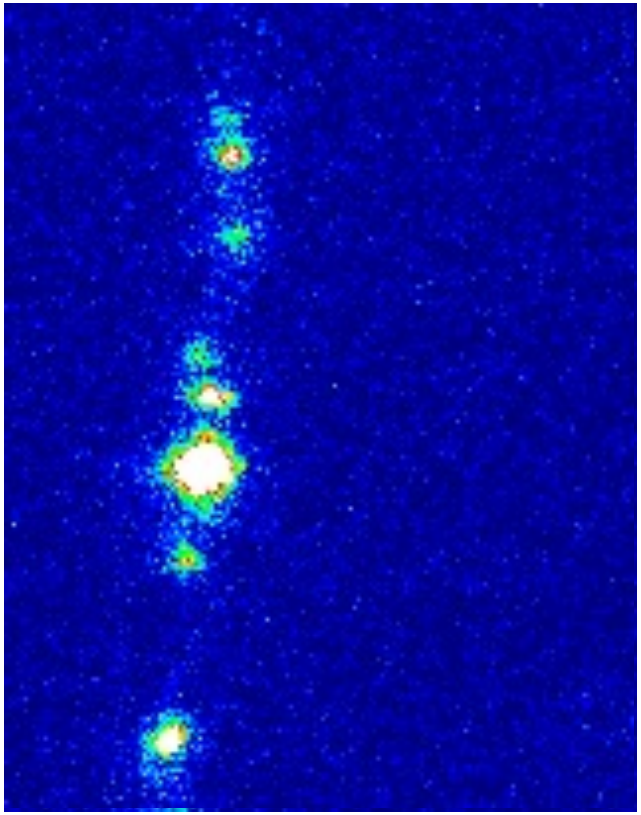


Fitted Phase Isosurface



Ferritin (111) Powder Ring

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



New routes to structure?

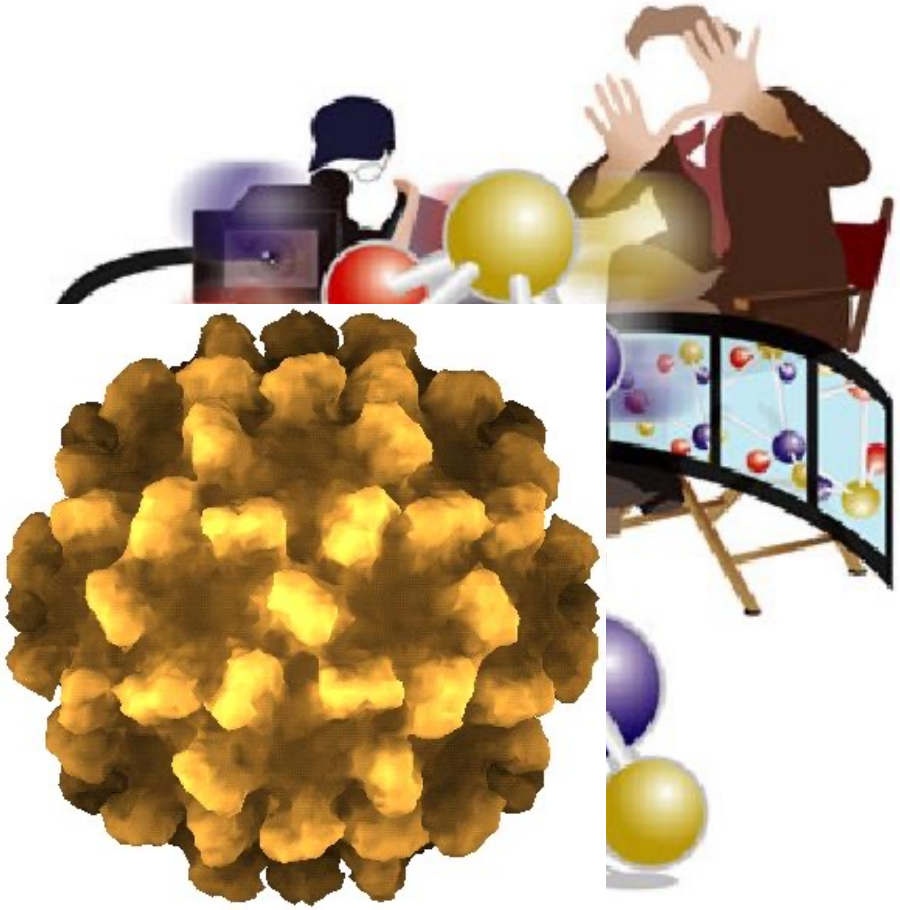
- Cryo-EM
  - real-space image, but not dose-efficient
- Inversion of diffraction
  - dose-efficient, especially for phase contrast
  - about equally efficient for photons or electrons
- How to beat the dose limit?

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



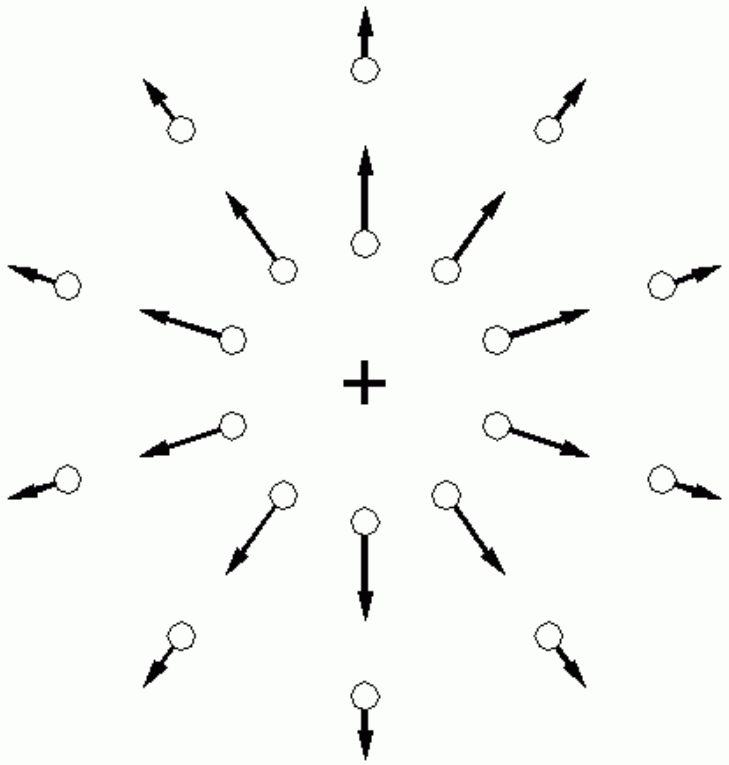
Conclusions and Outlook

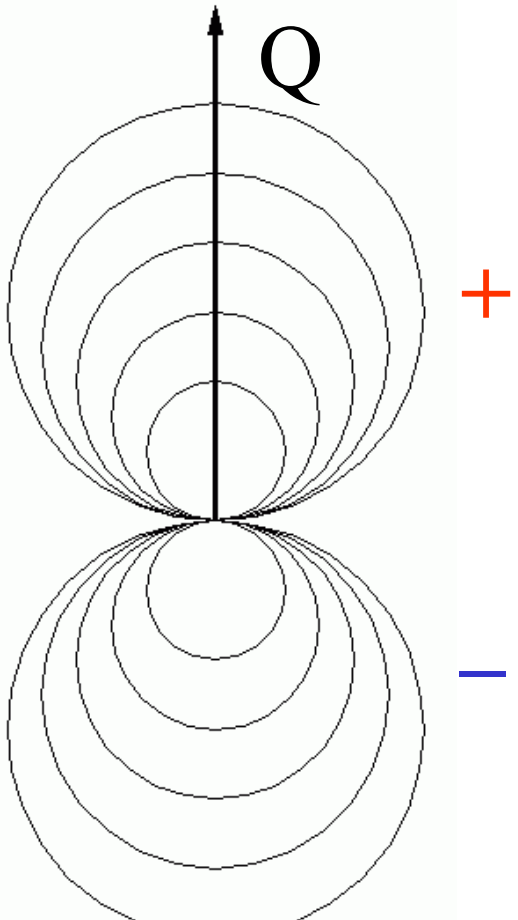
- Internal structure of Au Nanocrystals
- 3D imaging practical for nanocrystals
- Imaging by computation instead of lens
- Strain fields imaged as complex density
- Single molecules using XFEL

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





Critical Nucleation

Yau S.-T. and Vekilov P.G., *J. Am. Chem. Soc.* (**2001**), *123*, 1080-1089

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

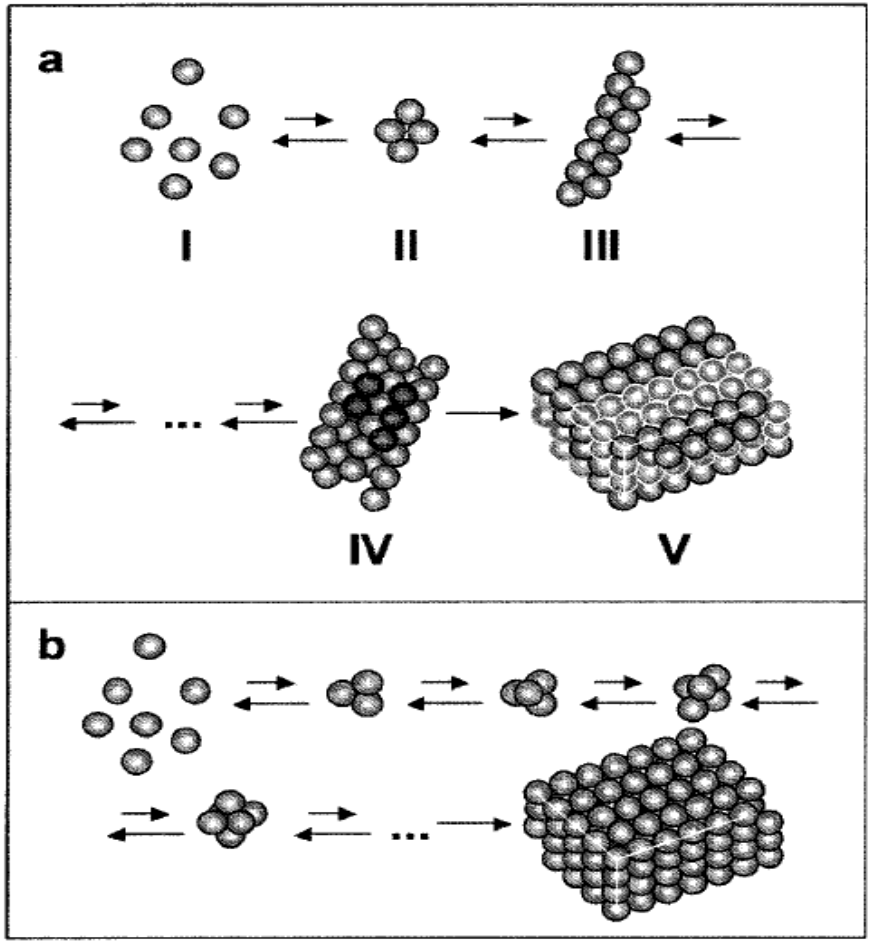


Figure 10. Schematic illustration of two nucleation pathways: (a) via a planar critical cluster (in IV molecules belonging to the second layer are shown in a lighter shade; in V the (110) layers that stack up to form this crystal are delineated by lighter and darker contours) and (b) via compact critical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

Compact Light Source

Ron Ruth, Lyncean Technologies