Investigation of Phase Objects through Coherent X-ray Diffraction

- Ian Robinson
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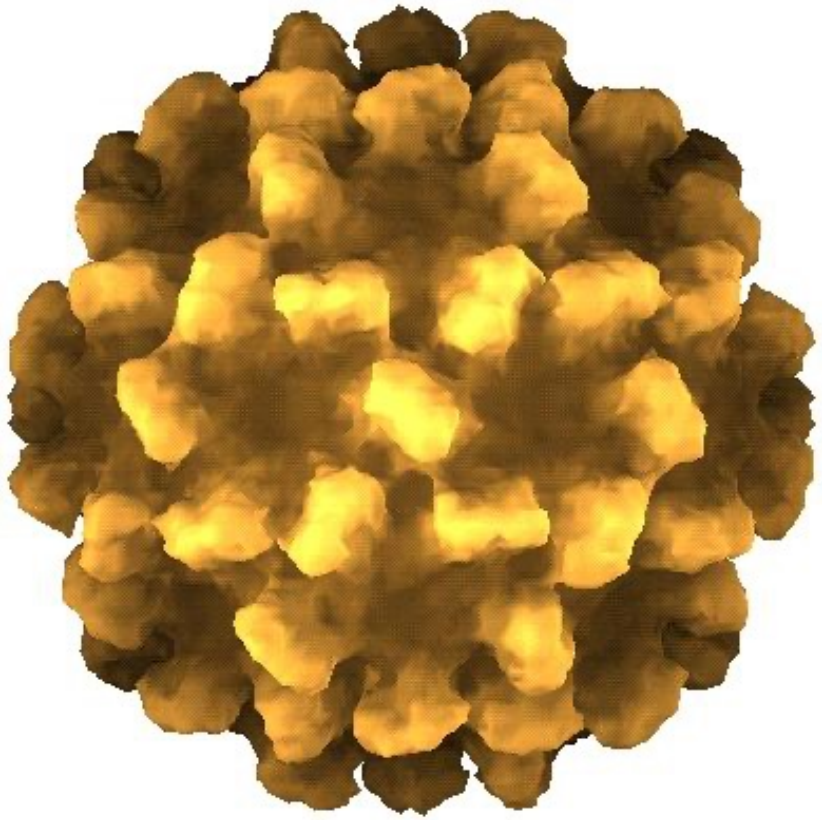
MPI Retreat,
Schloss Ringberg,
January 2005

Tomato Bushy Stunt Virus
1980



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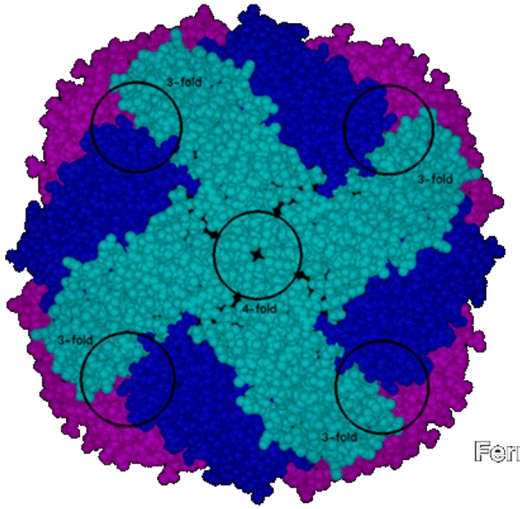
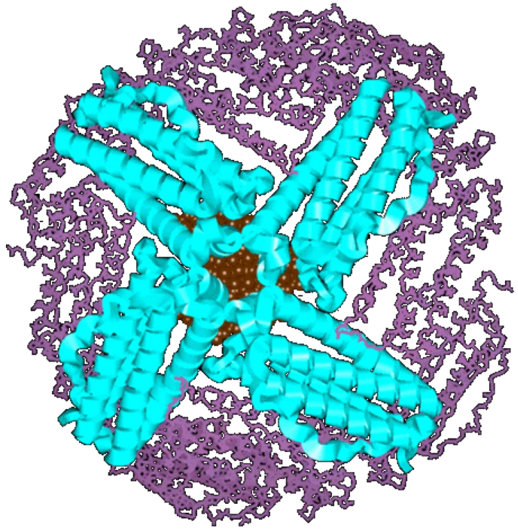
Horse Spleen Ferritin

- Ferritin used in almost all living things for Iron Storage
- Made of 24 identical protein subunits arranged in 432 symmetry
- Crystallizes as FCC (I23)
- Inner shell diameter 80Å
- Outer shell diameter 130Å
- Iron core: ferrihydrite form
- With Iron : Holoferritin
- Empty Shell : Apoferritin





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.

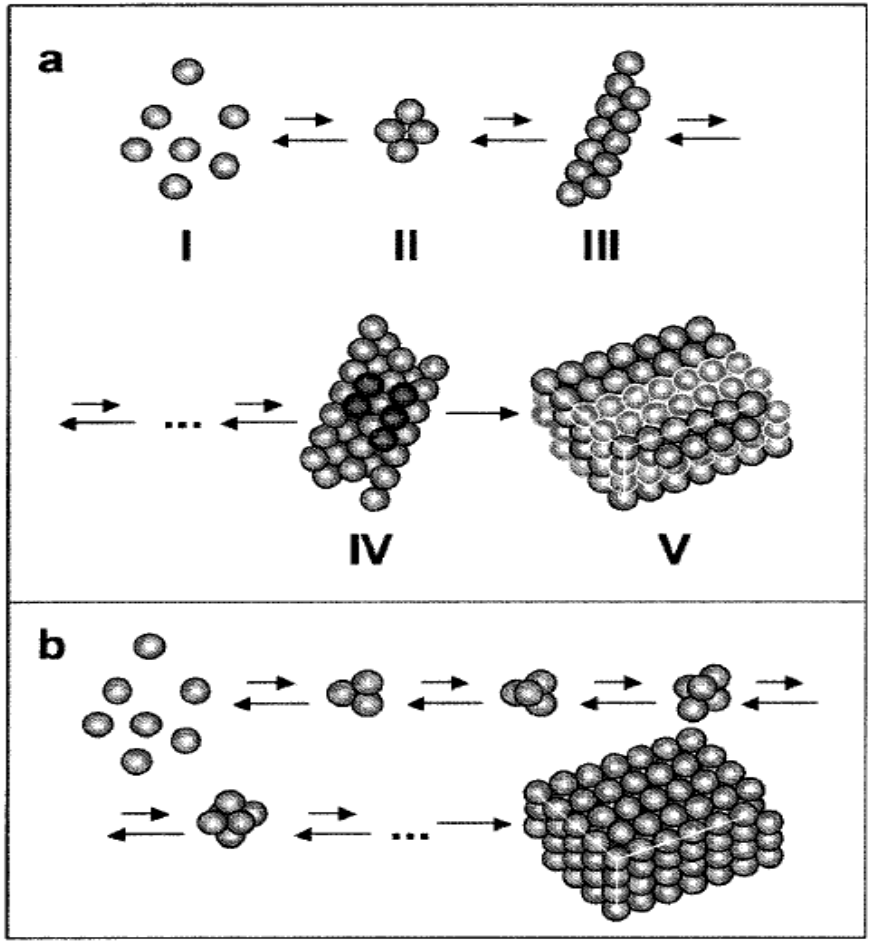
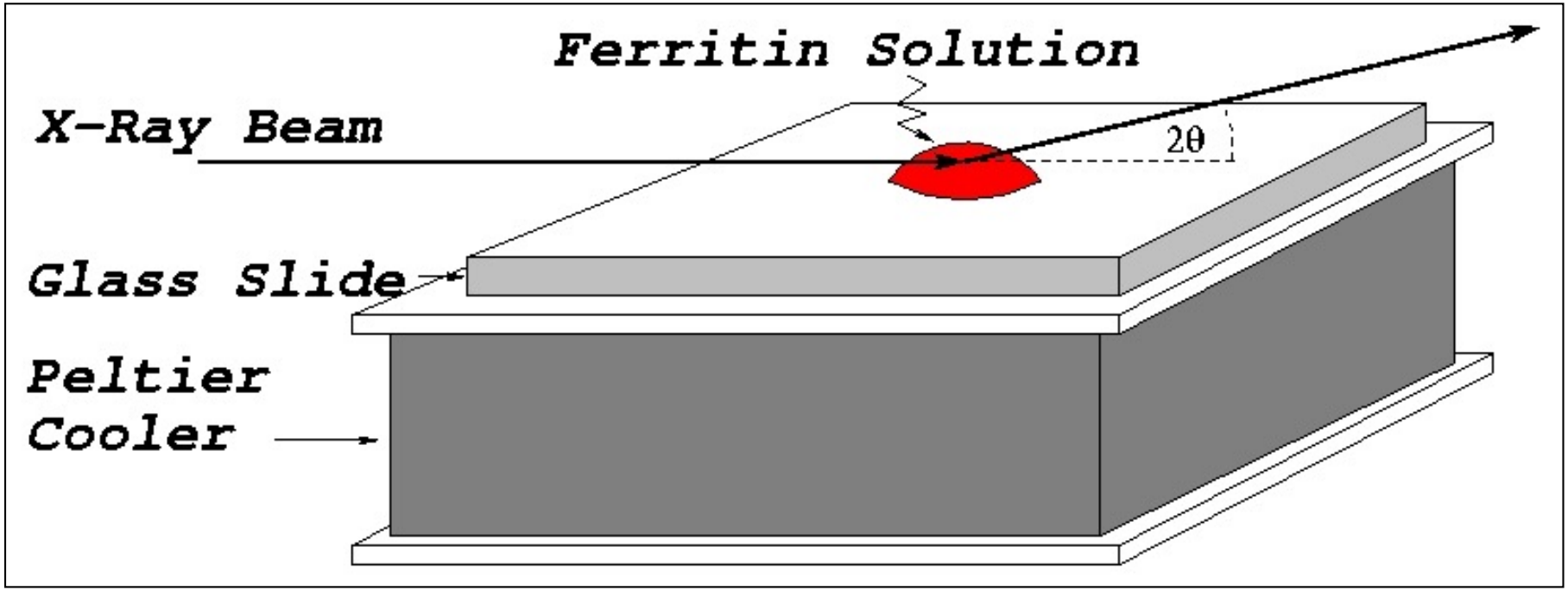
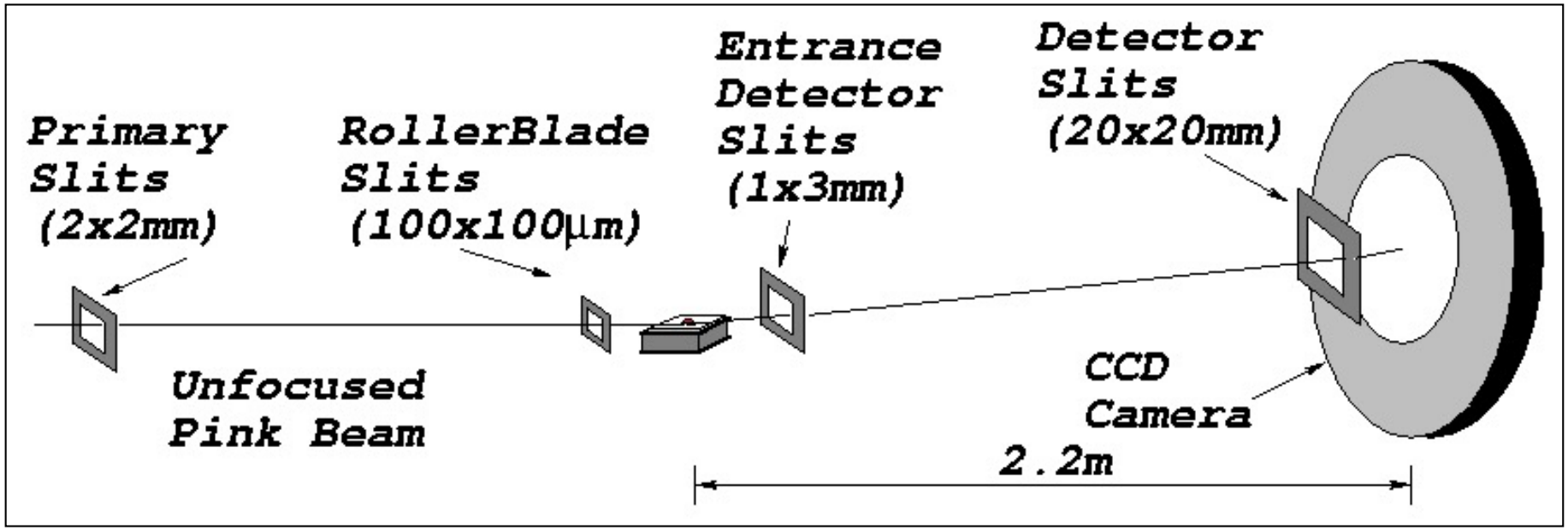


Figure 10. Schematic illustration of two nucleation pathways: (a) via a planar critical cluster (in IV molecules belonging to the second layer are shown in a lighter shade; in V the (110) layers that stack up to form this crystal are delineated by lighter and darker contours) and (b) via compact critical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

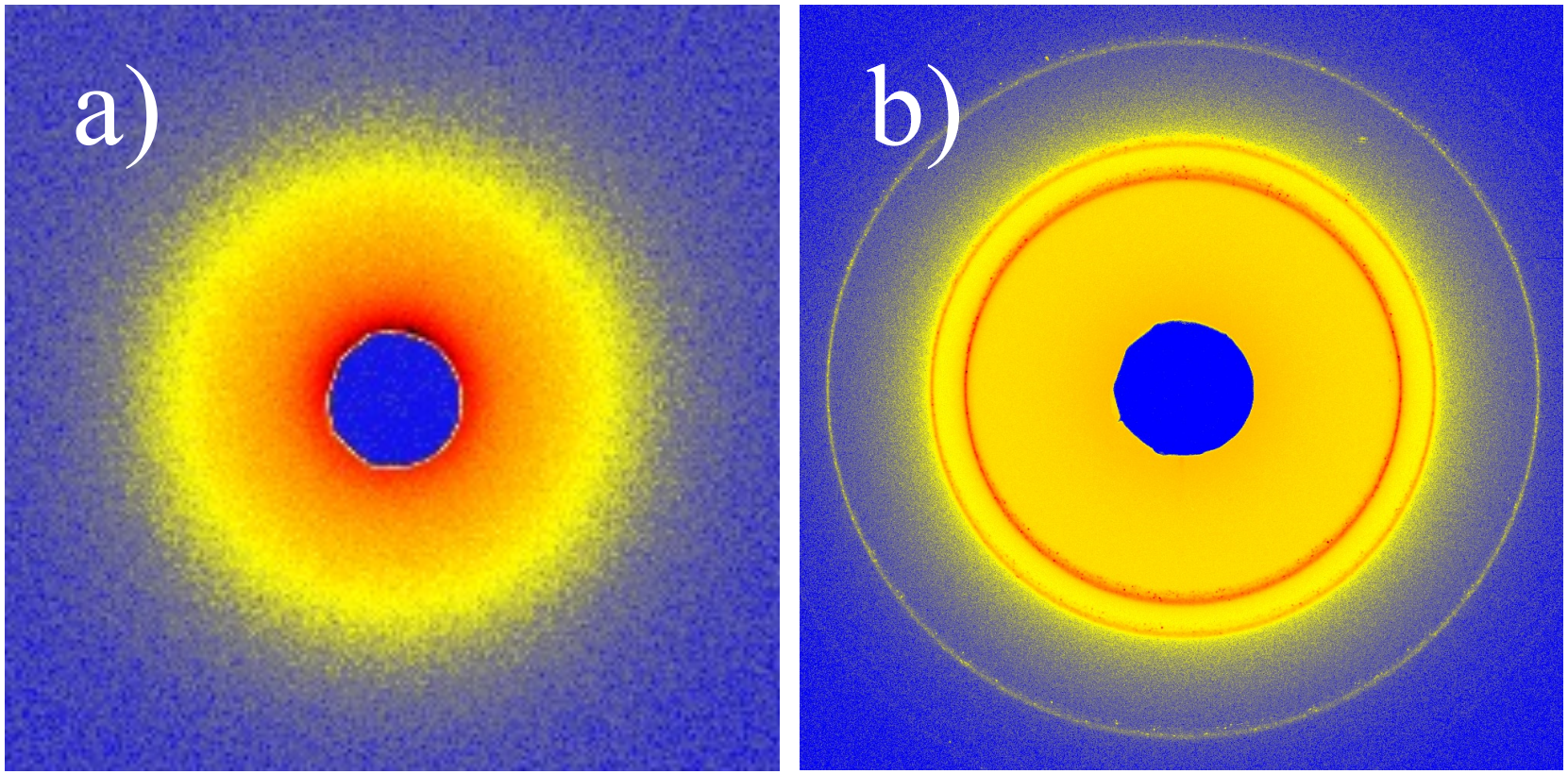
In-situ Study of Crystallization



Experiment at APS Sector 34



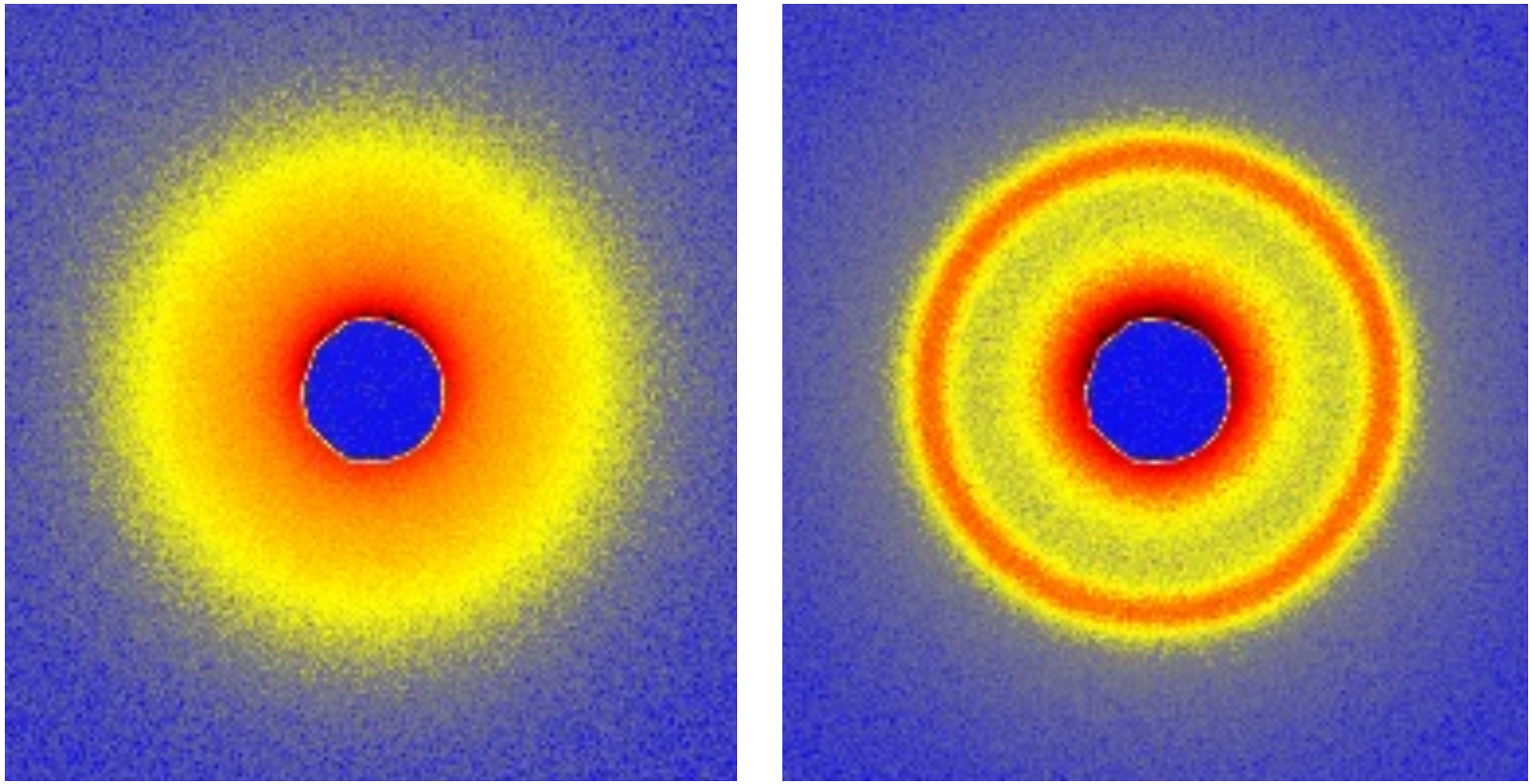
Crystallization of Holoferritin: SAXS



a) SAXS pattern of holoferritin at 10°C.

b) SAXS pattern after adding Cadmium salt

Effect of Temperature



SAXS pattern at T=+10°C and -10°C.

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

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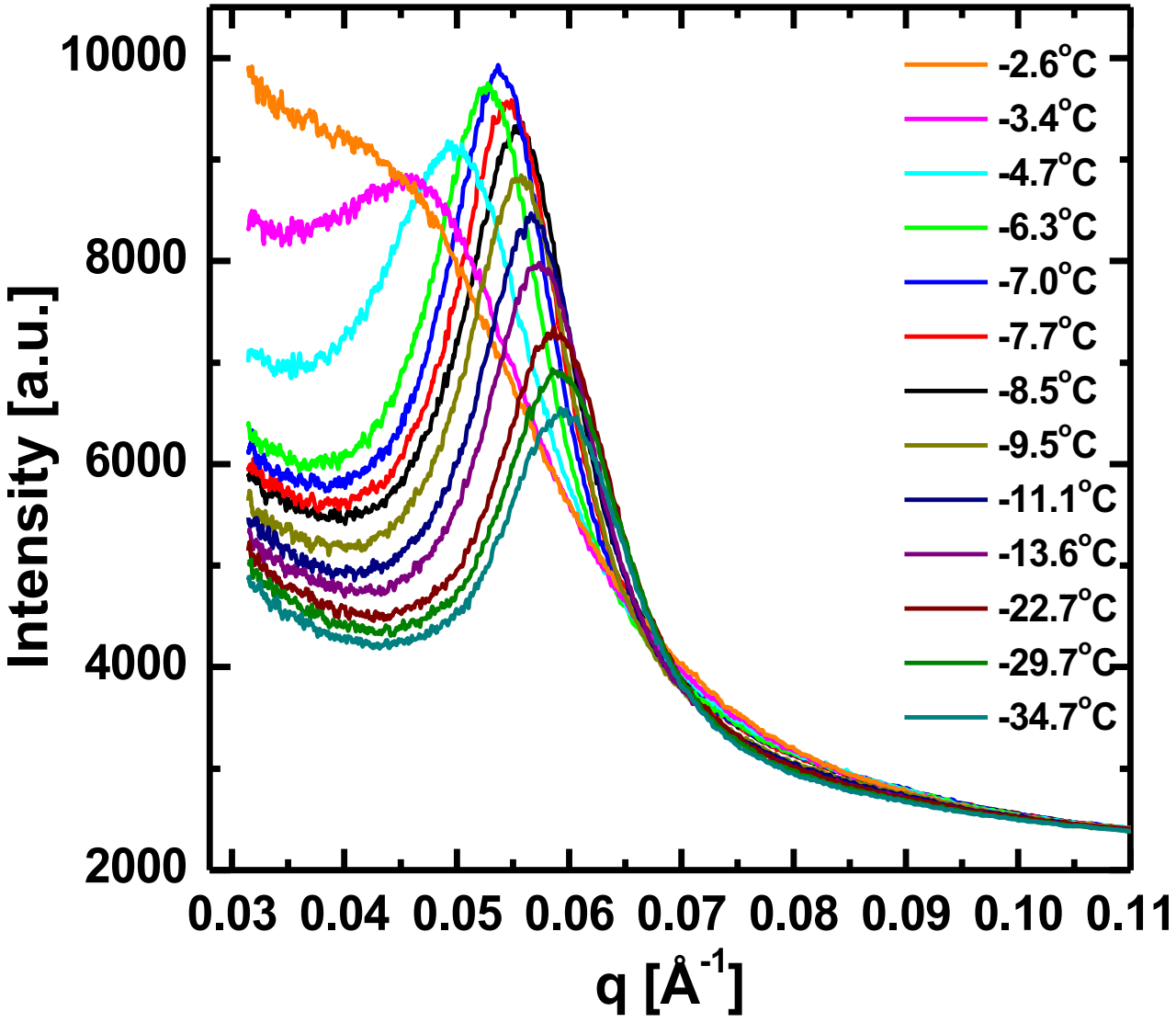
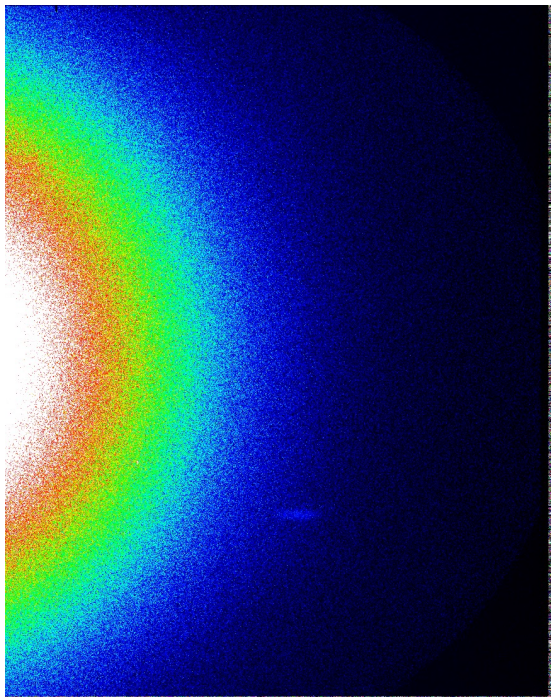
Integrated SAXS Data

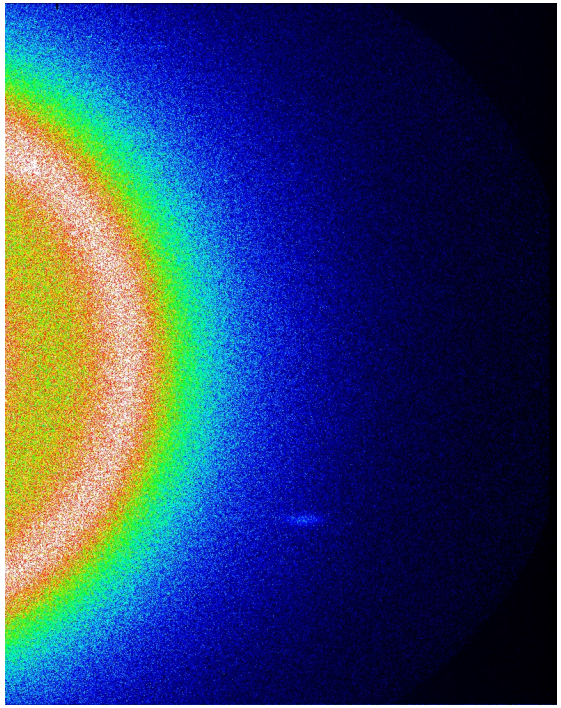


- The SAXS pattern shows a broad peak which shifts in position and changes in width as a function of temperature

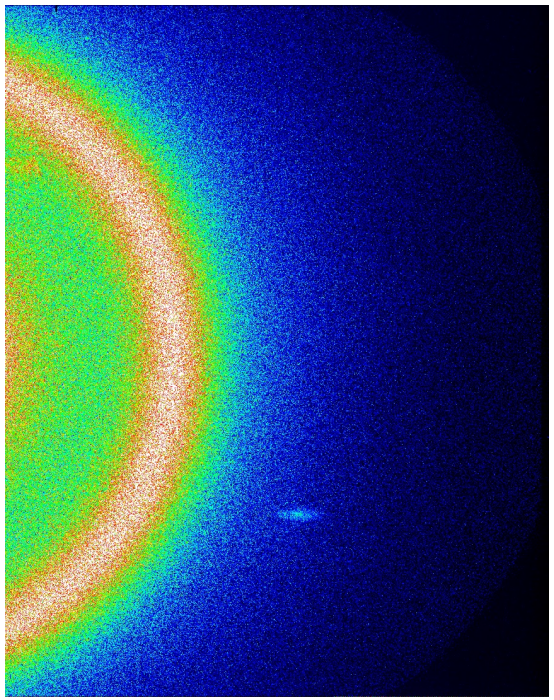
Ferritin Solution upon Freezing



T=13.3



T=-4.7



T=-34.7

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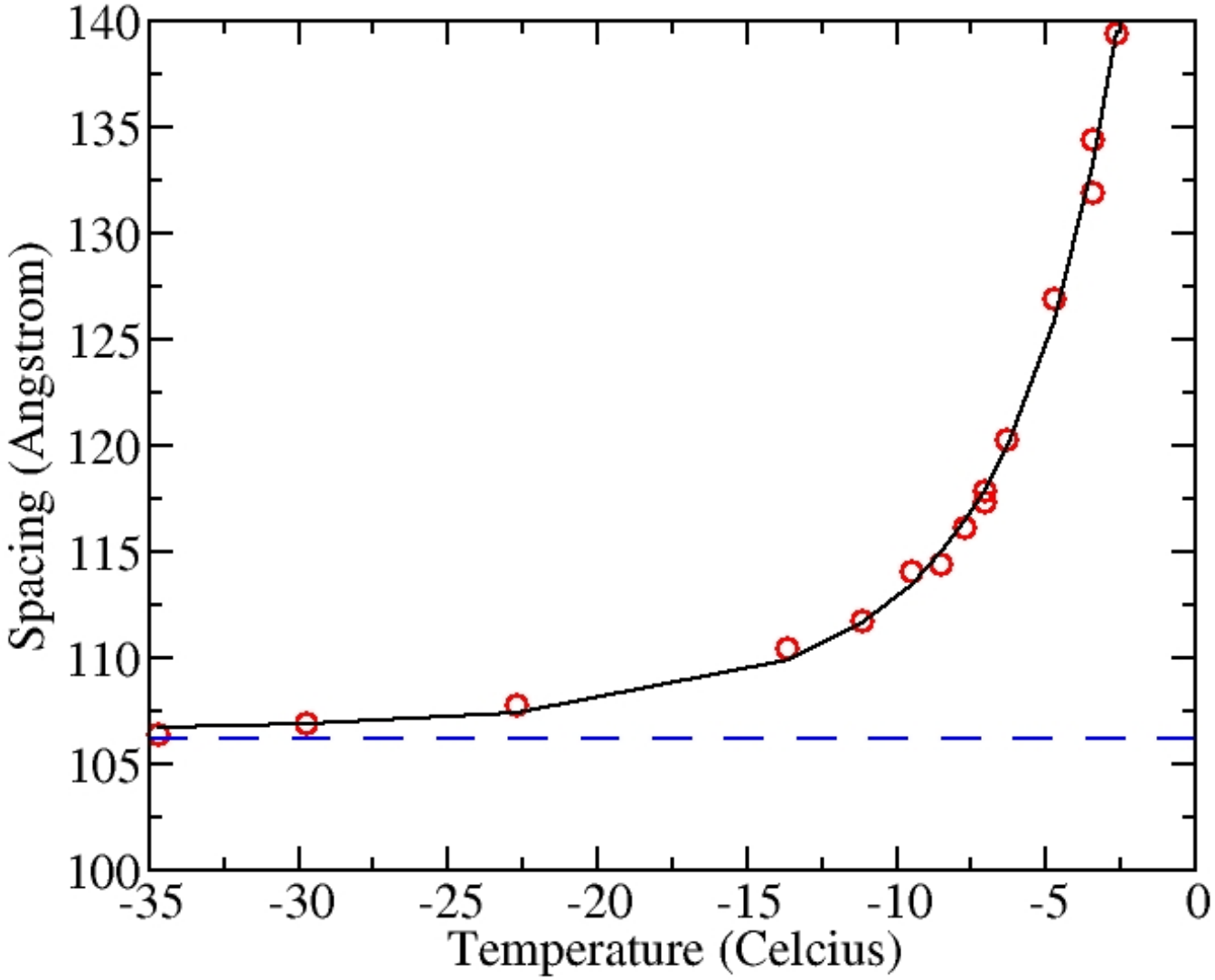
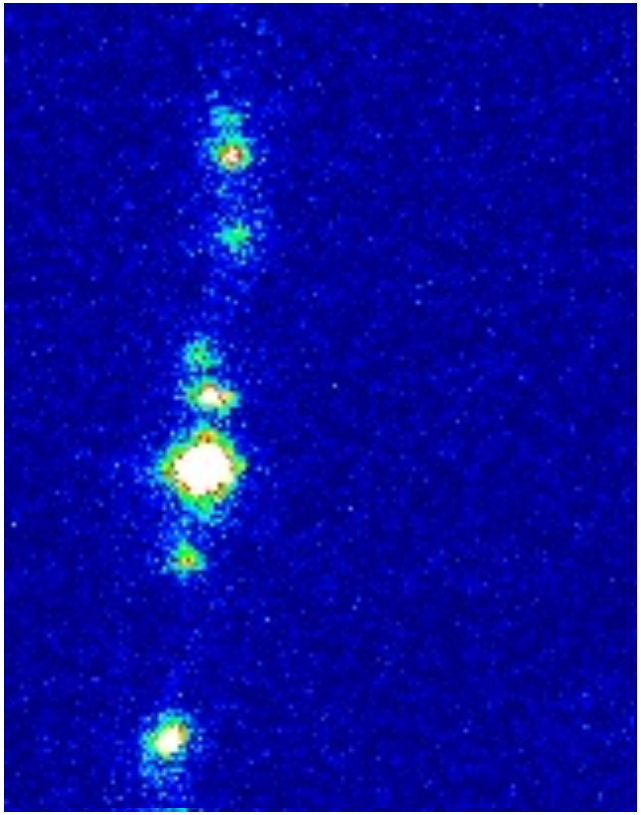
Temperature Variation of Peak Position

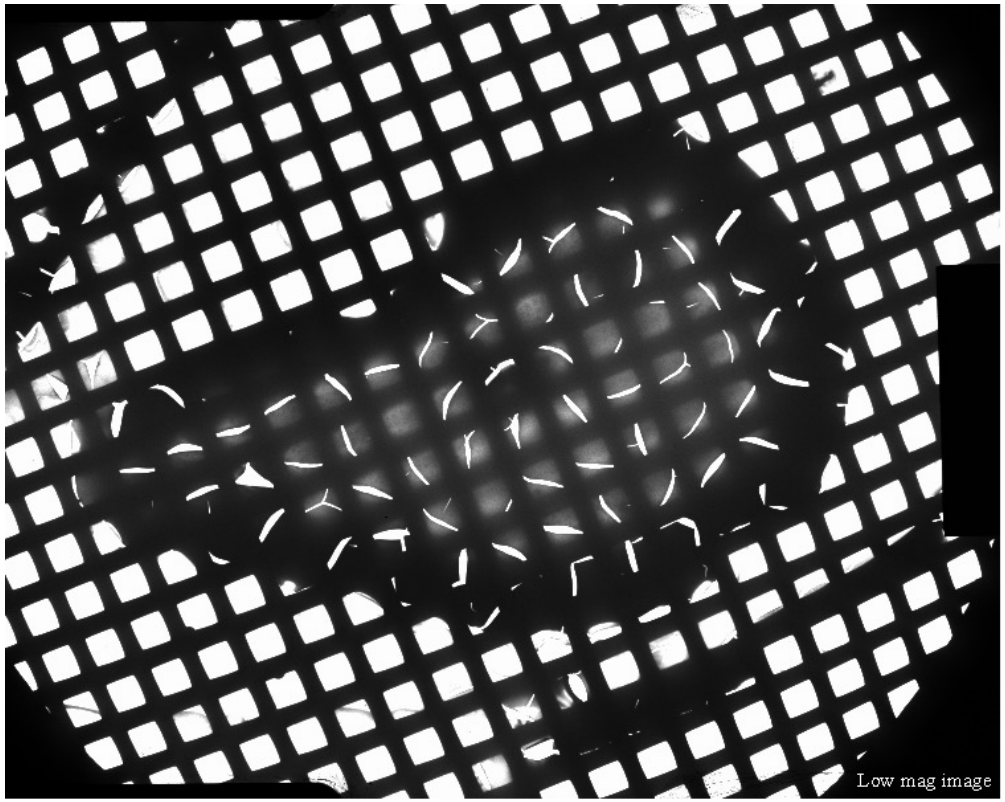


Ferritin (111) Powder Ring

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

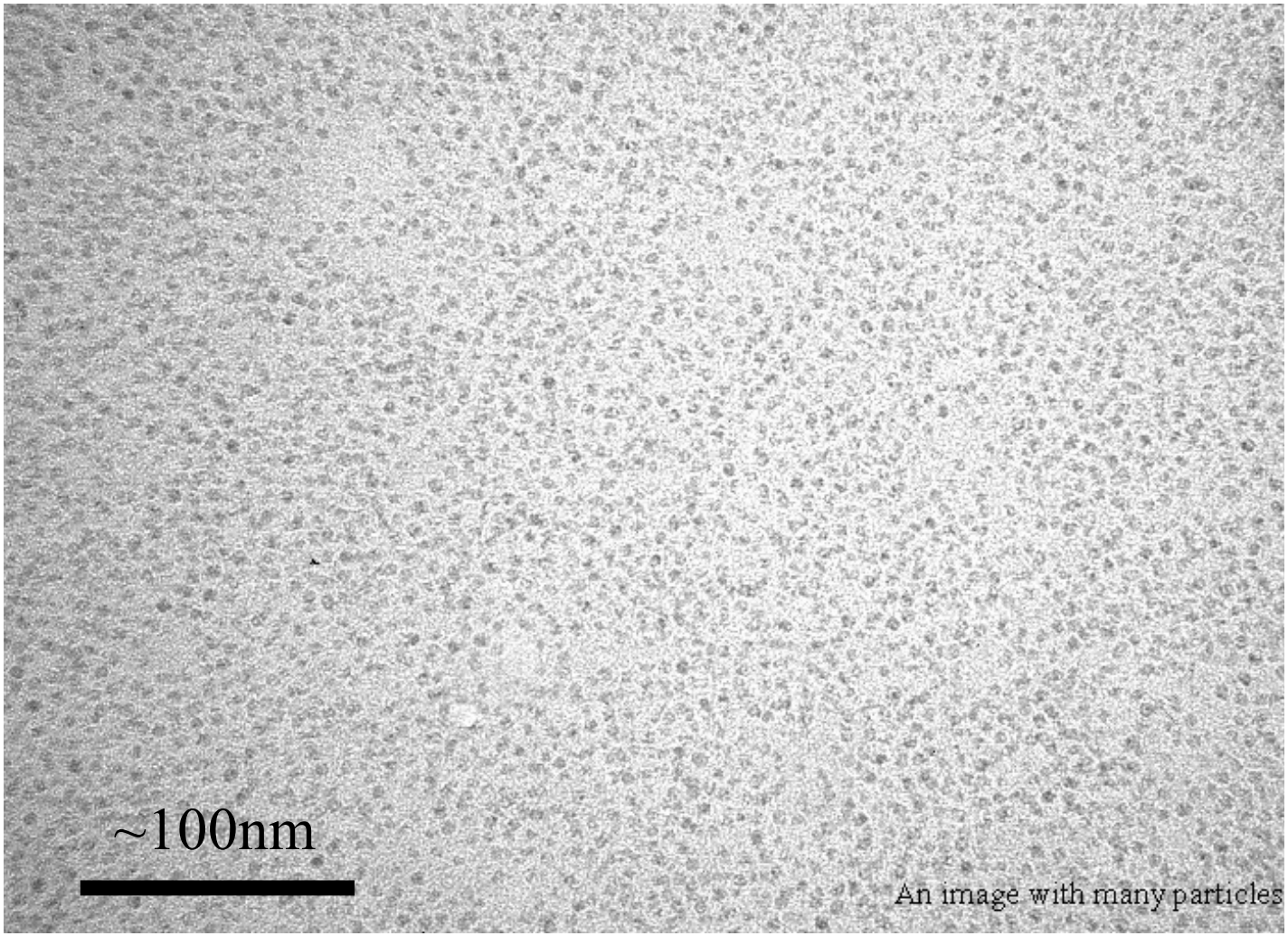


Overview of EM grid



- 3mm diam grid
- a-carbon + “Formvar”
- 10mg/ml Ferritin
- 10mg/ml NaCl
- dipped grid
- dried in air
- no cryo-cooling
  - ▷three regions of different density

Border Region (low density)



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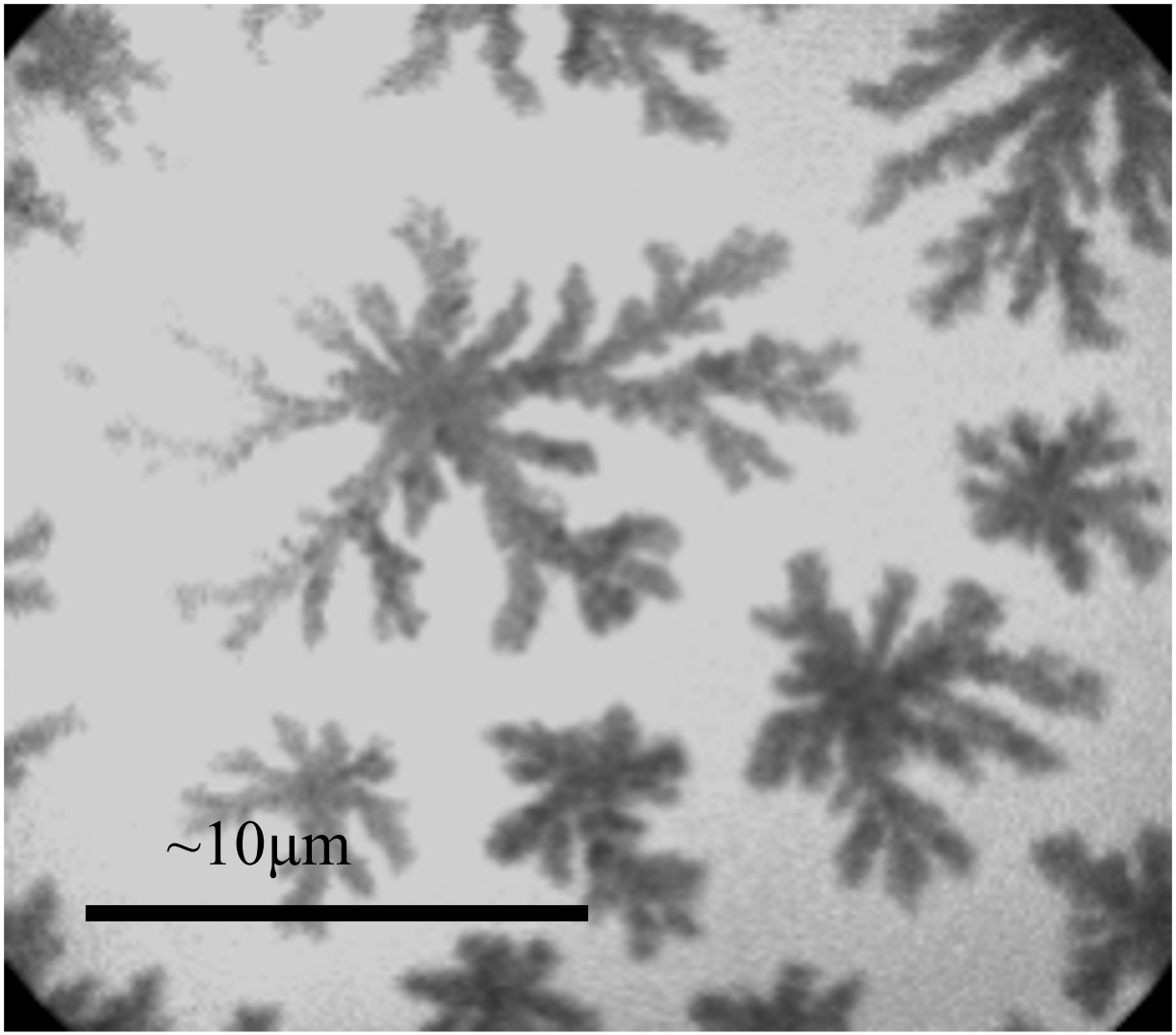
High Density Ring



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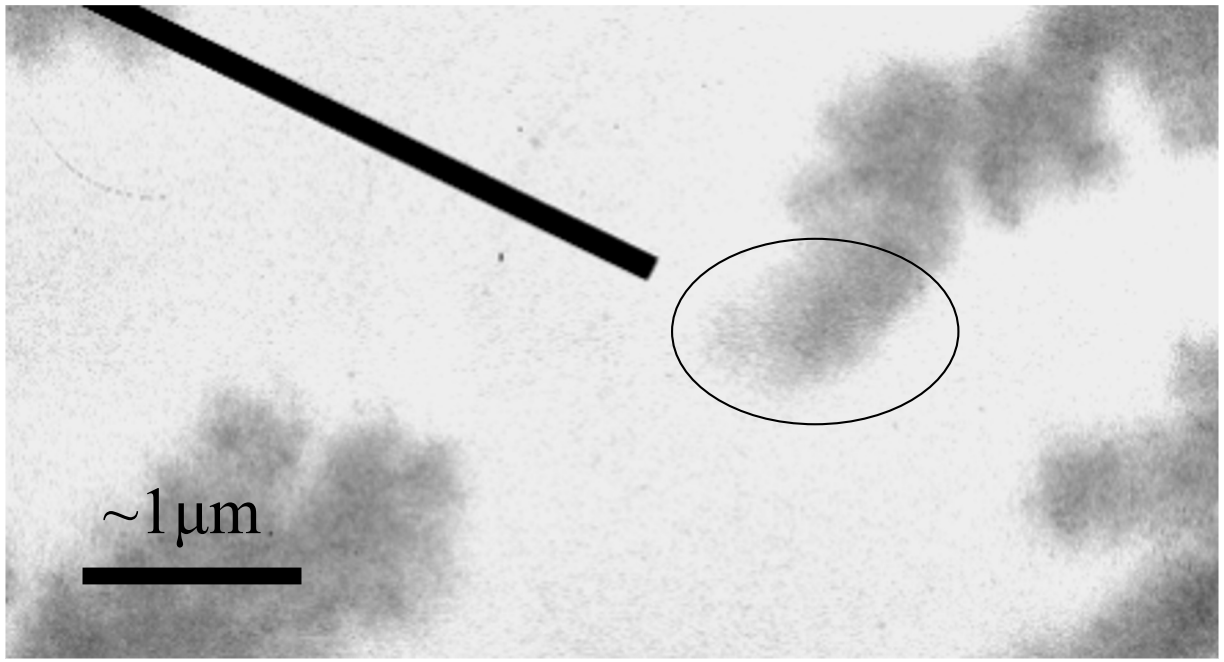
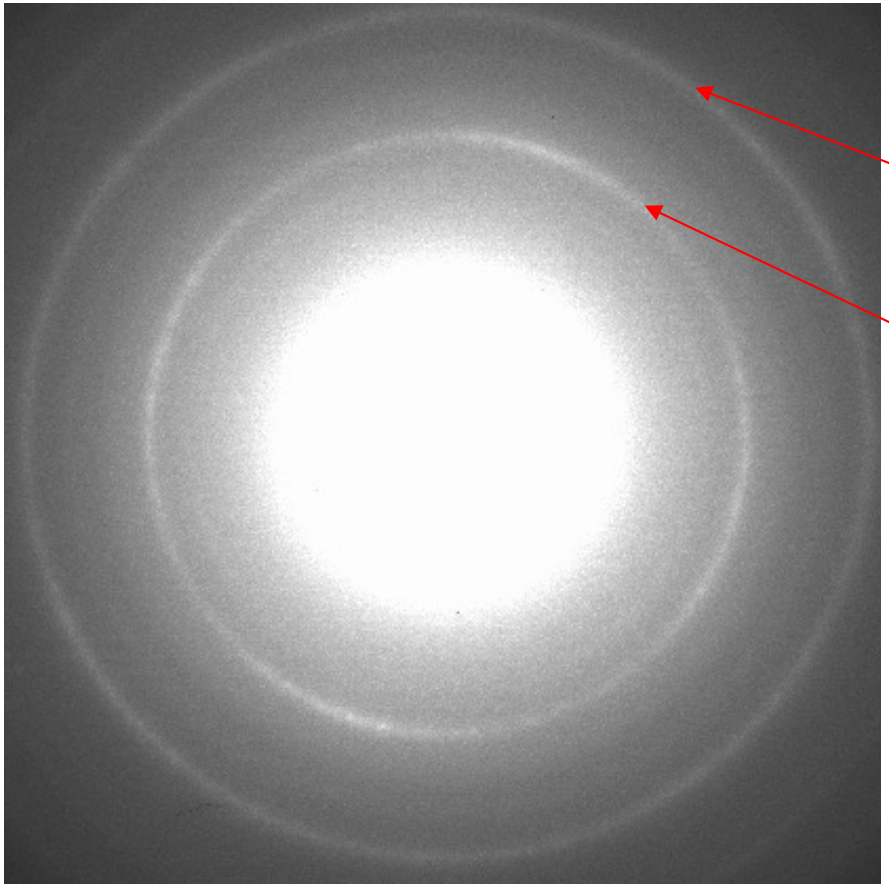
Select Area around Dendrimer Tip



Selective Area Diffraction



$Q=6.32Å^{-1}$

$Q=4.44Å^{-1}$

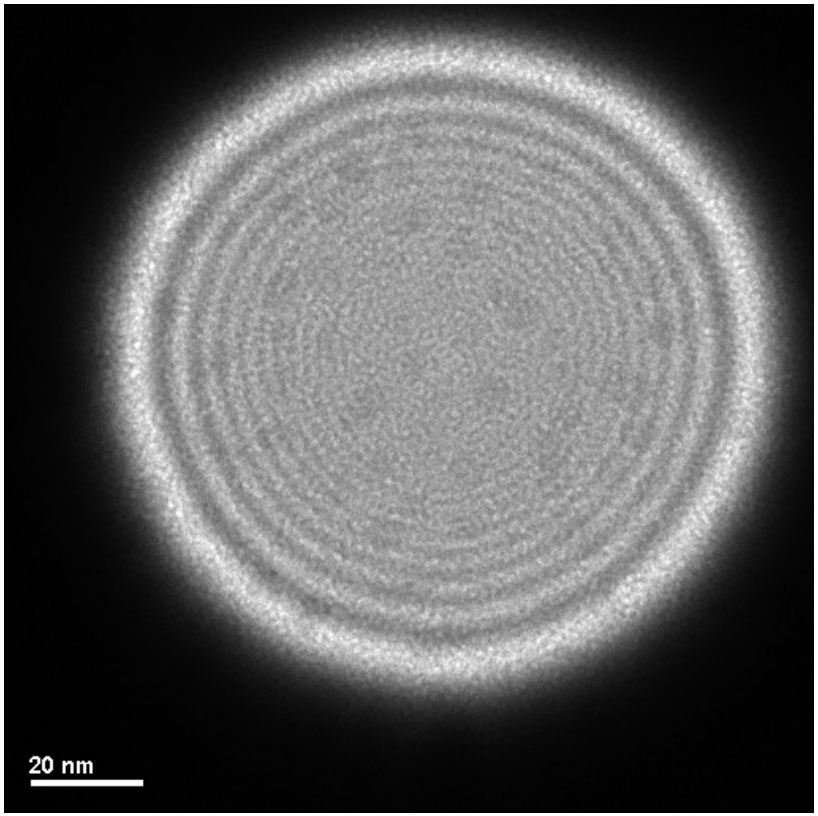
$\lambda = 0.025Å$

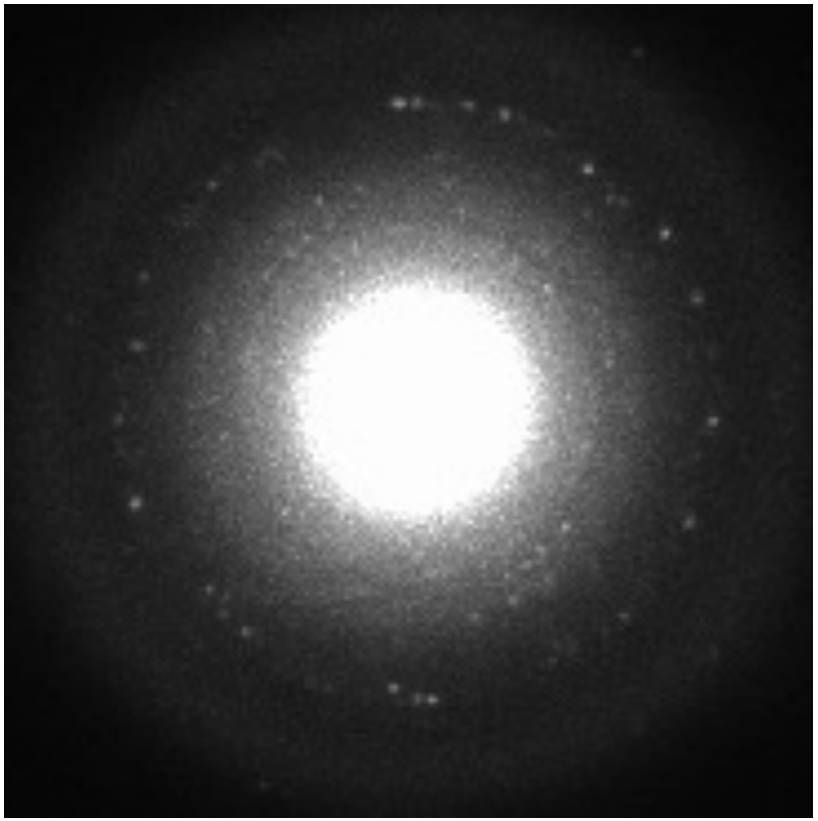
$D = 2.0m$

$R_1=35.4mm$

$R_2=50.4mm$

Diffraction from Border Region





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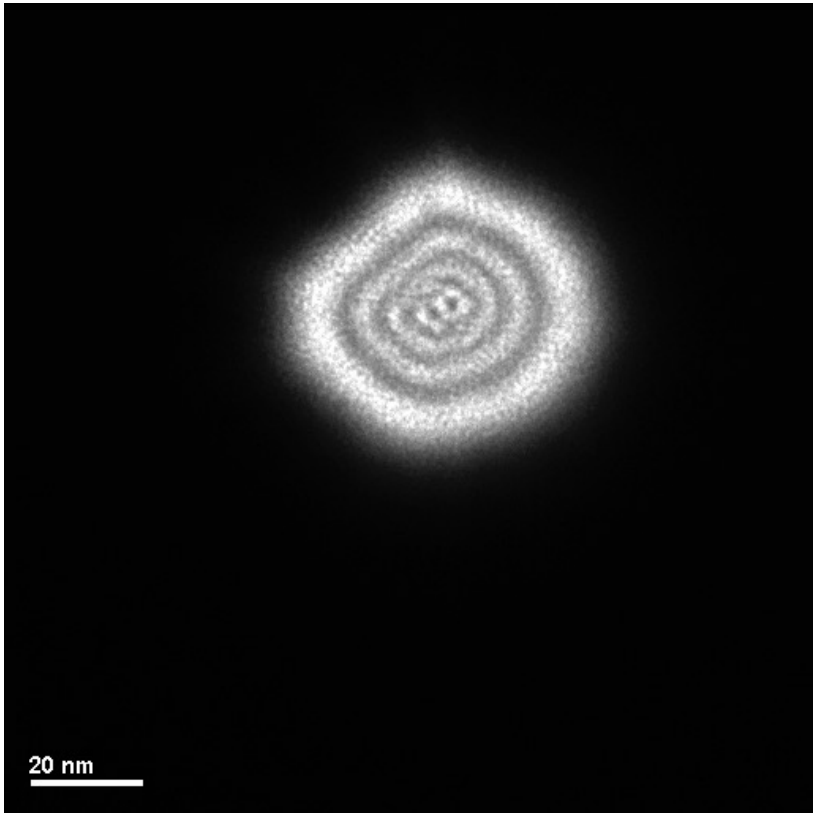
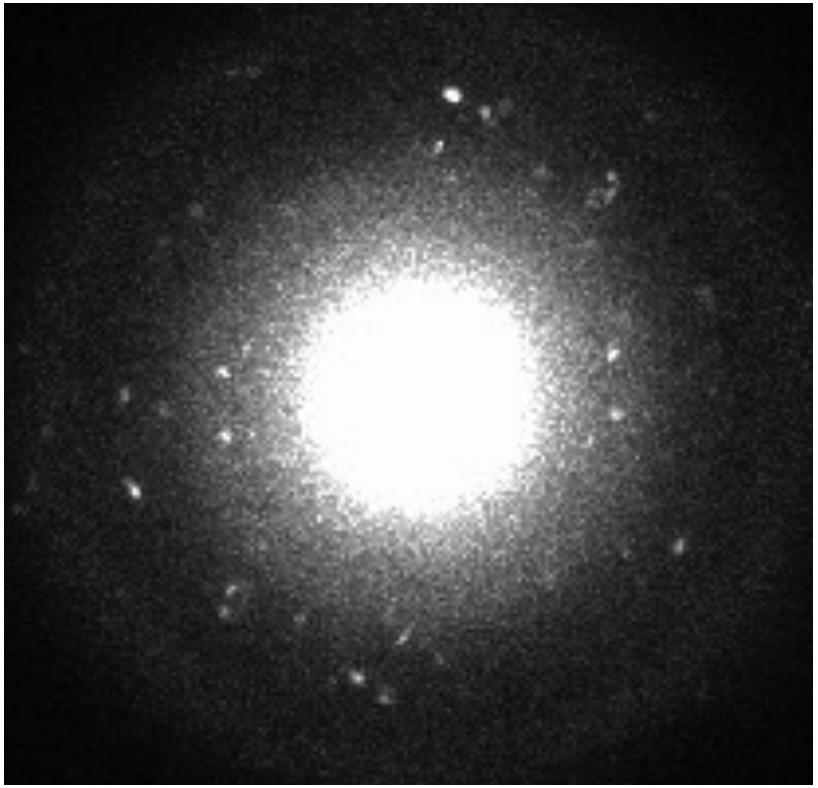
Smaller Area Diffraction

Images: picture2 and diffraction4





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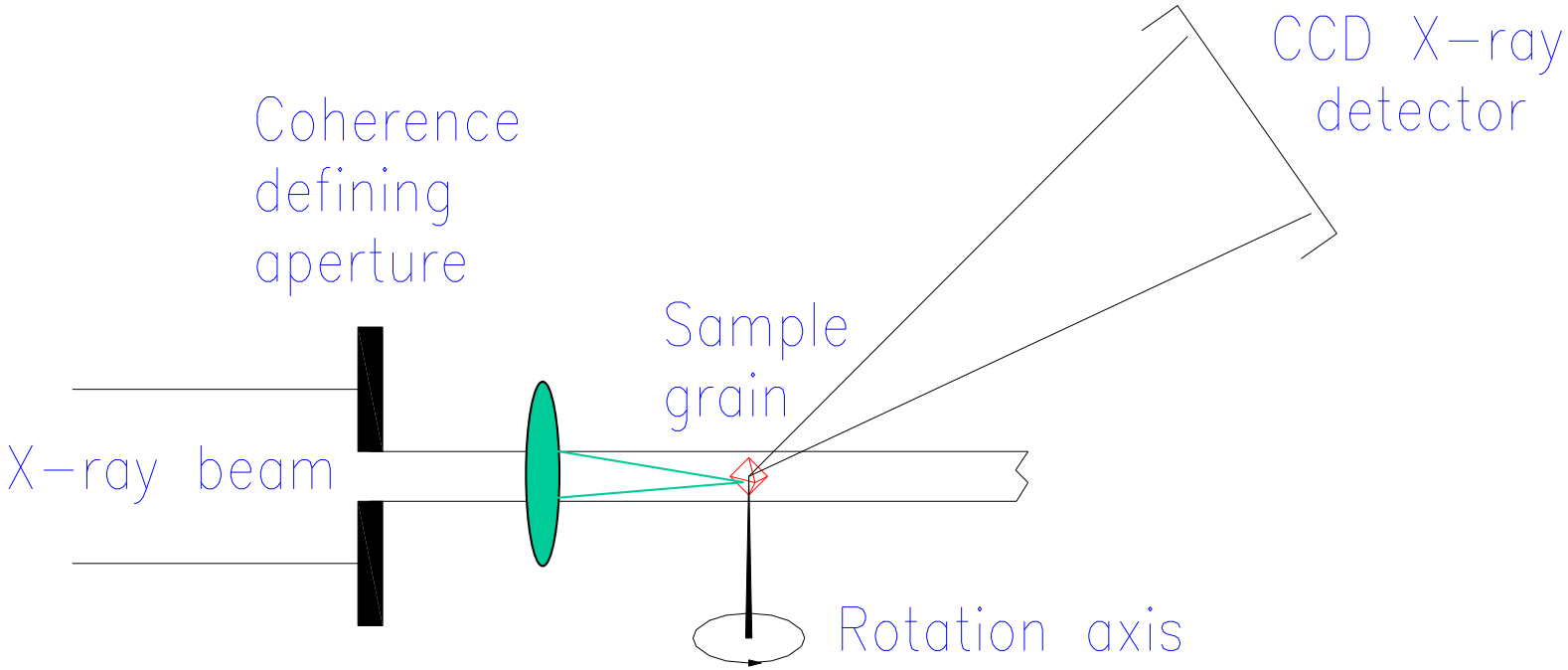
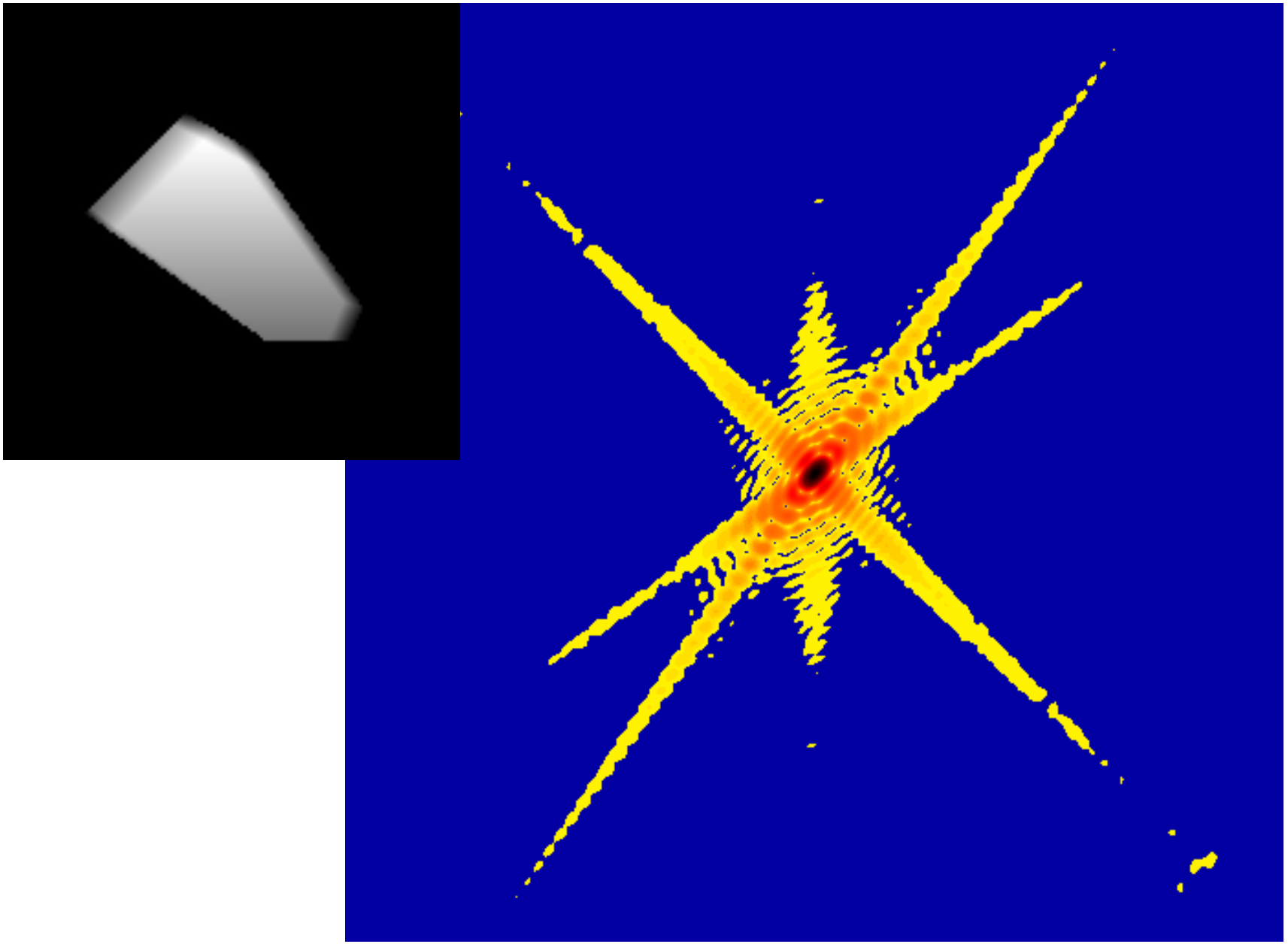
Lensless X-ray Microscope





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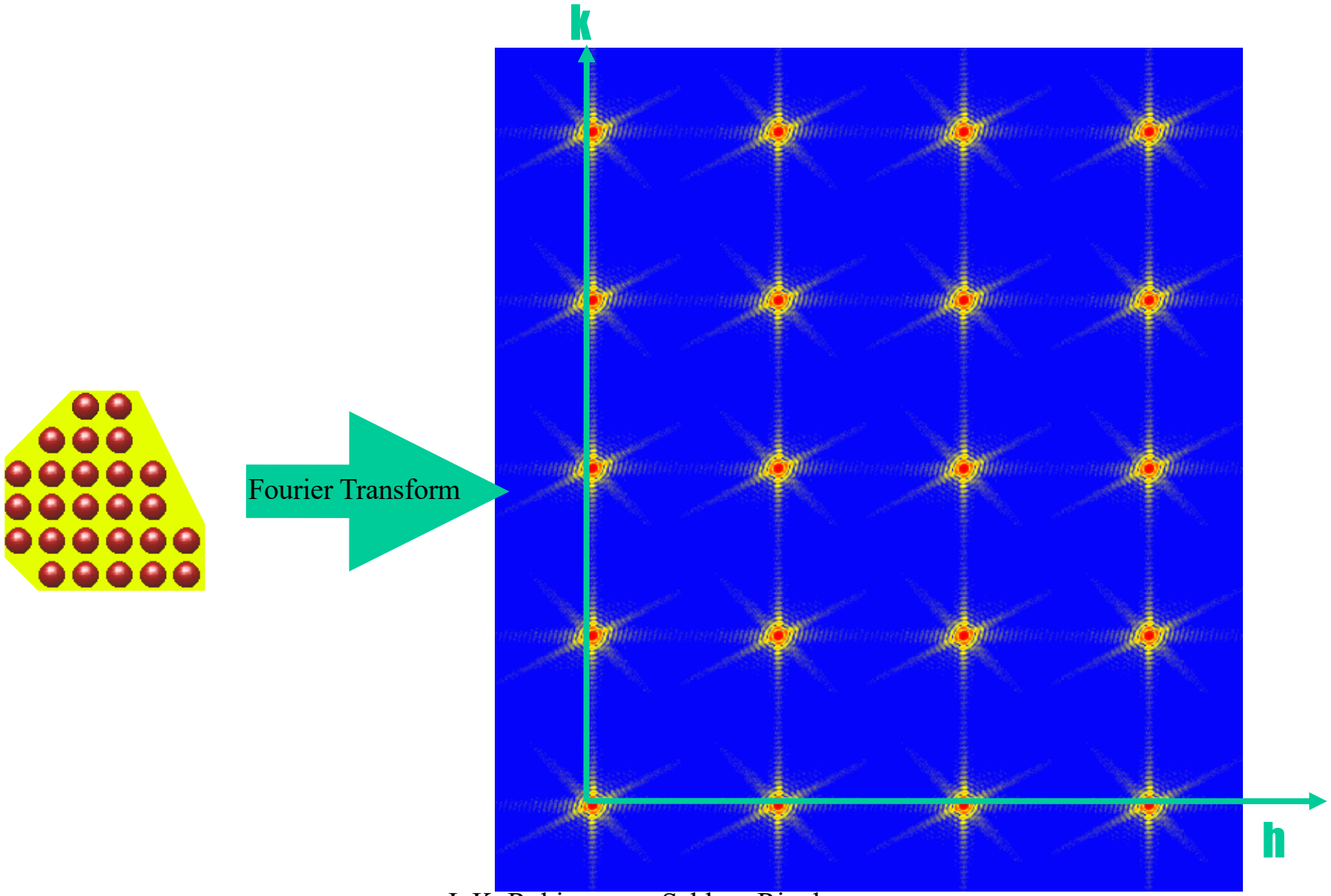
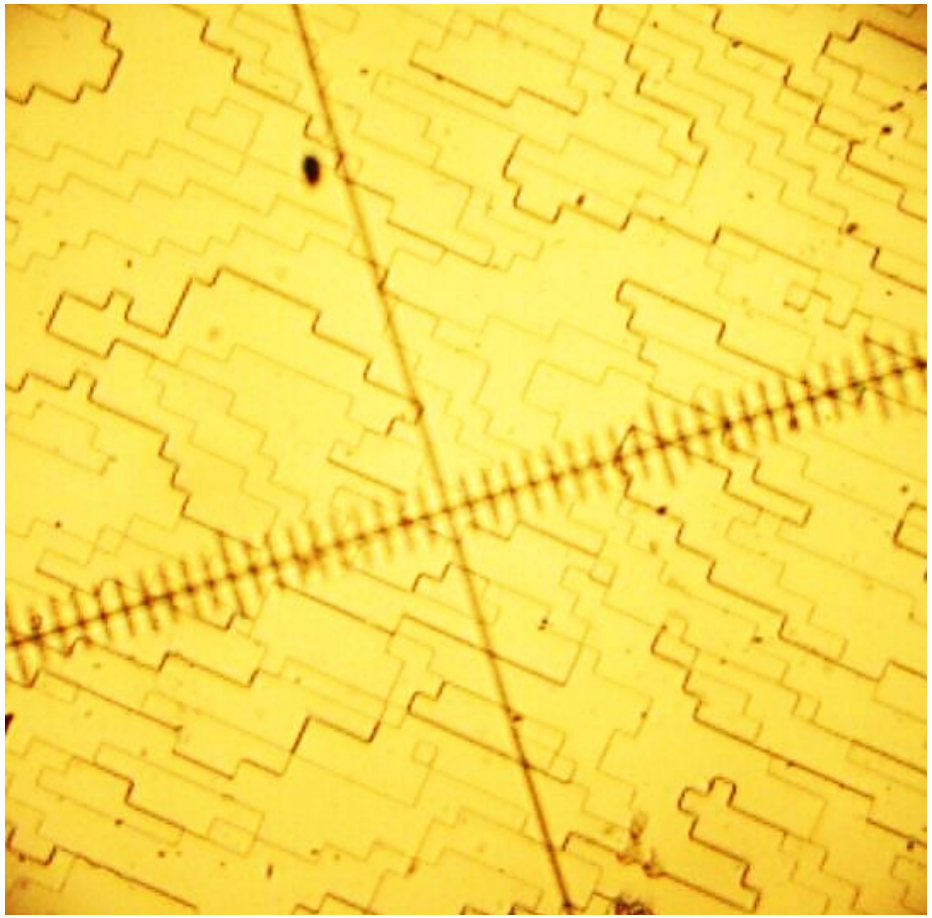
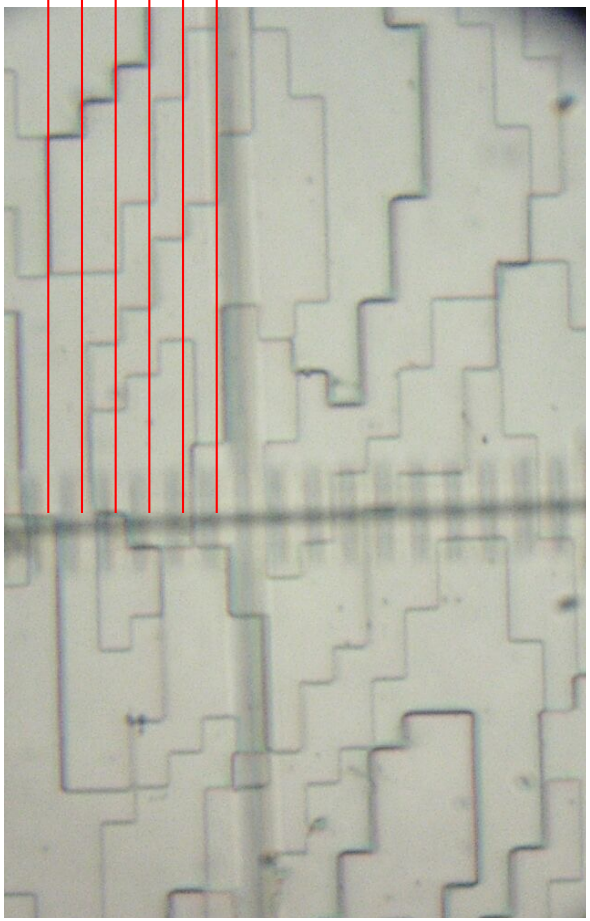
Coherent Diffraction from Crystals



Microscope Images of Gratings



5μm grid lines



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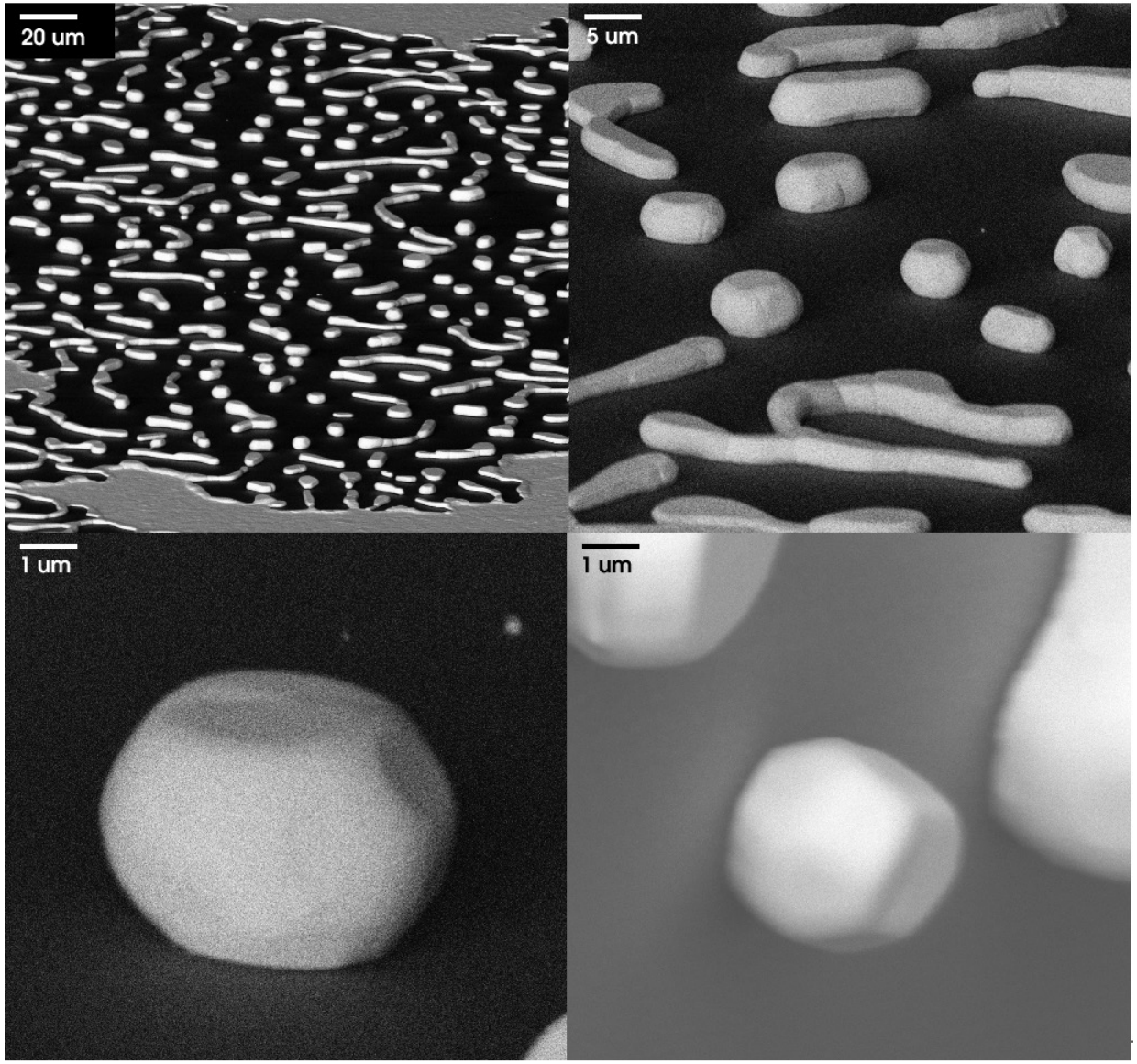
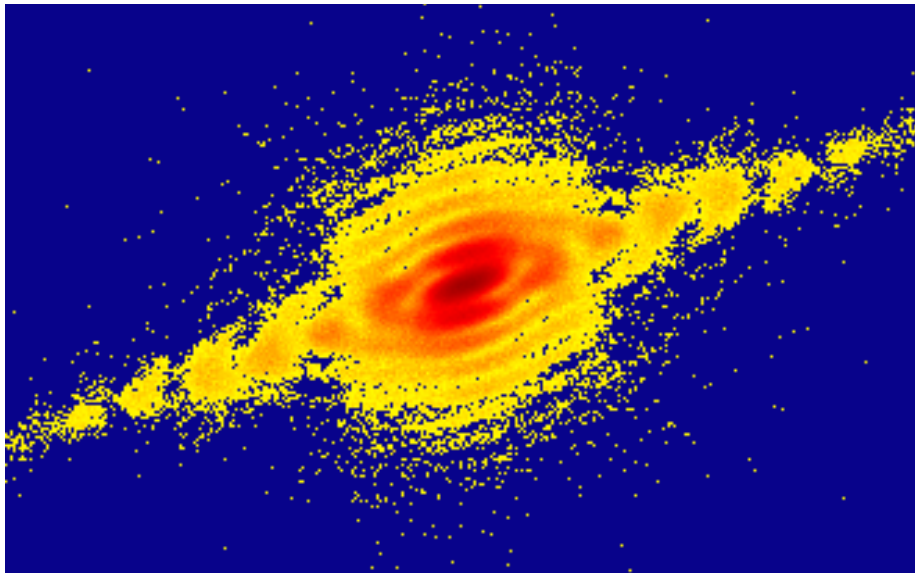
SEMS

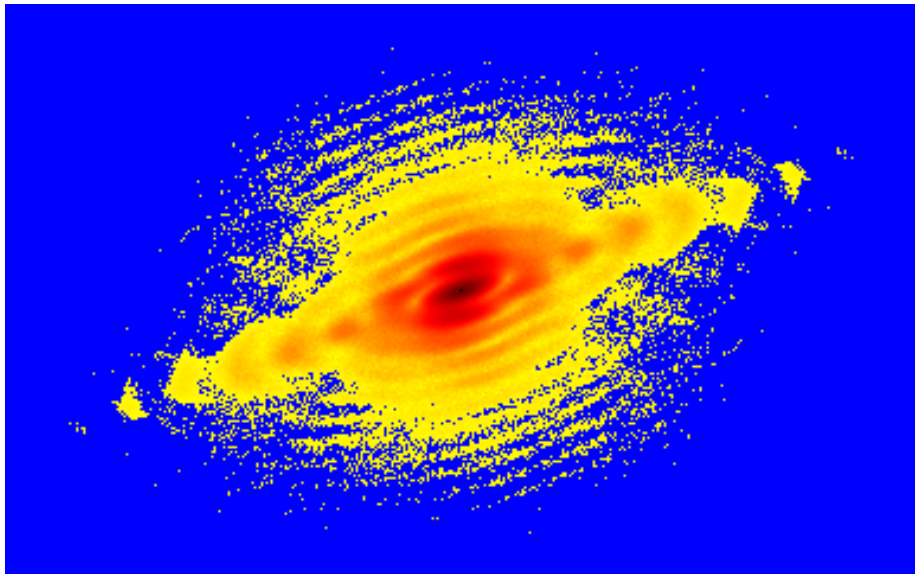
- Au blanket film
- Quartz substrate
- Annealed at 950°C for 70 hrs.

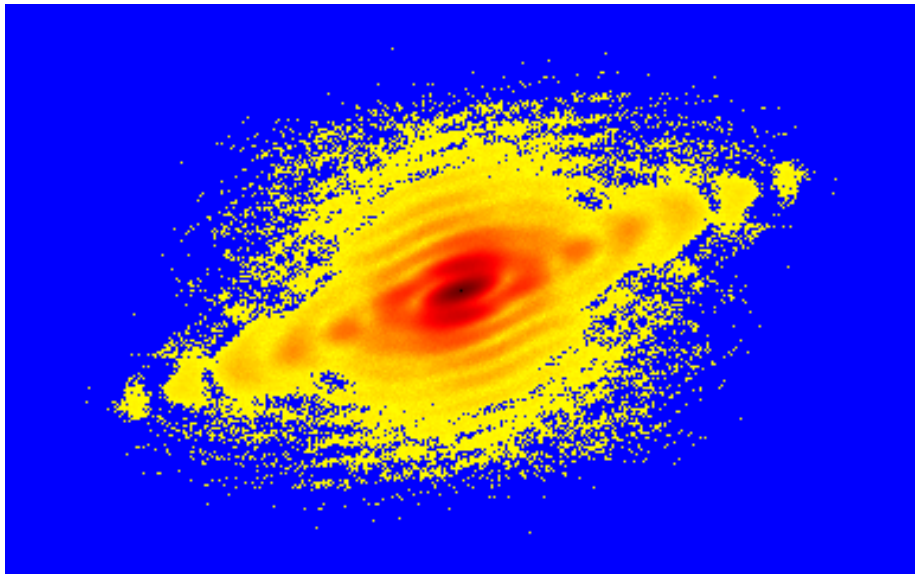


Symmetrized Data and two best fits

Chisq=0.0005







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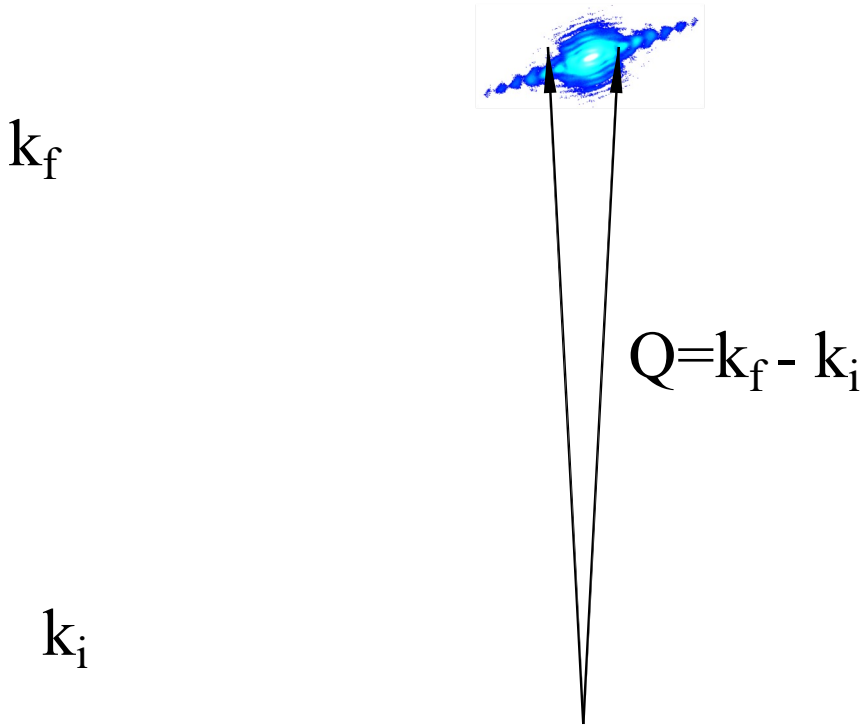
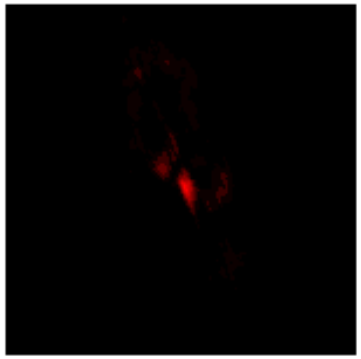
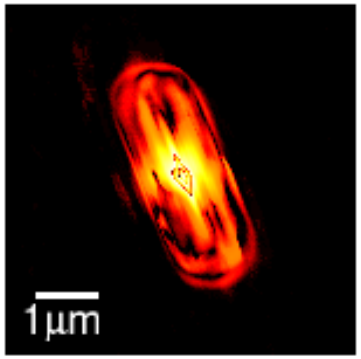
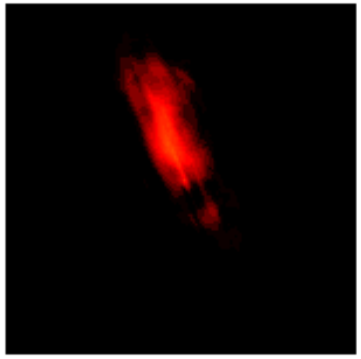
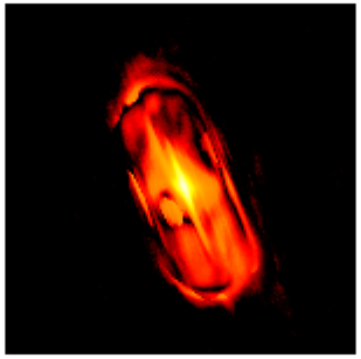
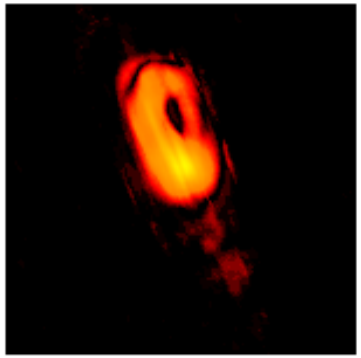
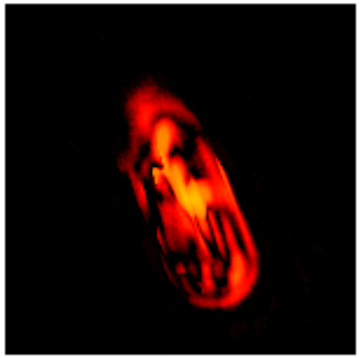
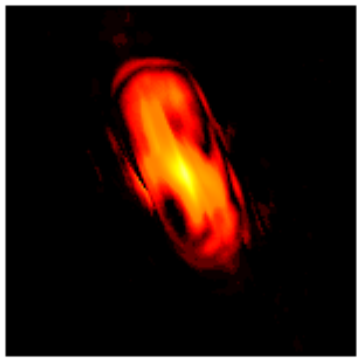
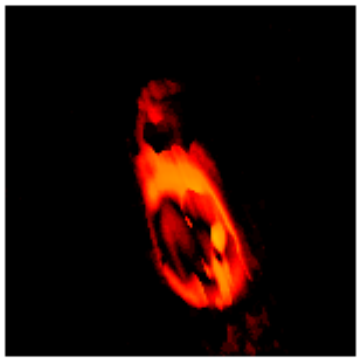
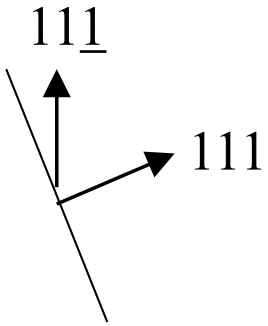
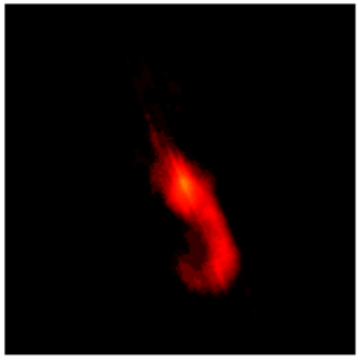
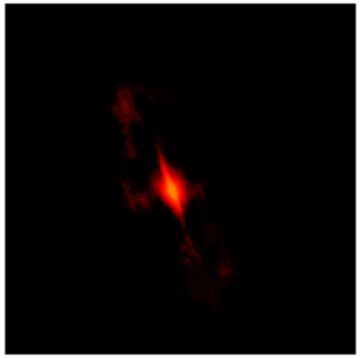
3D Diffraction Method

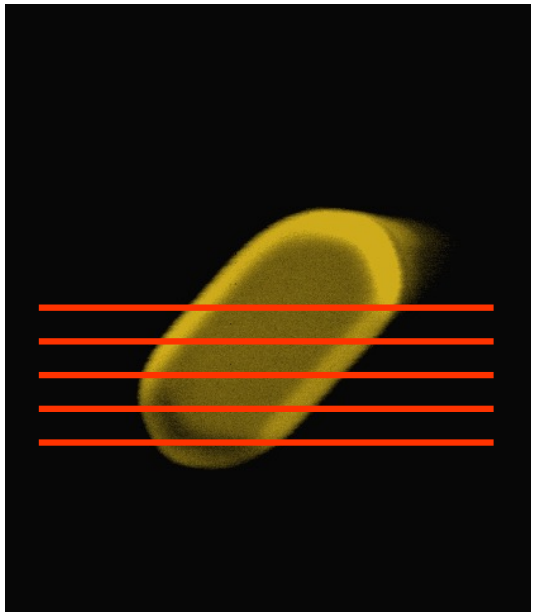


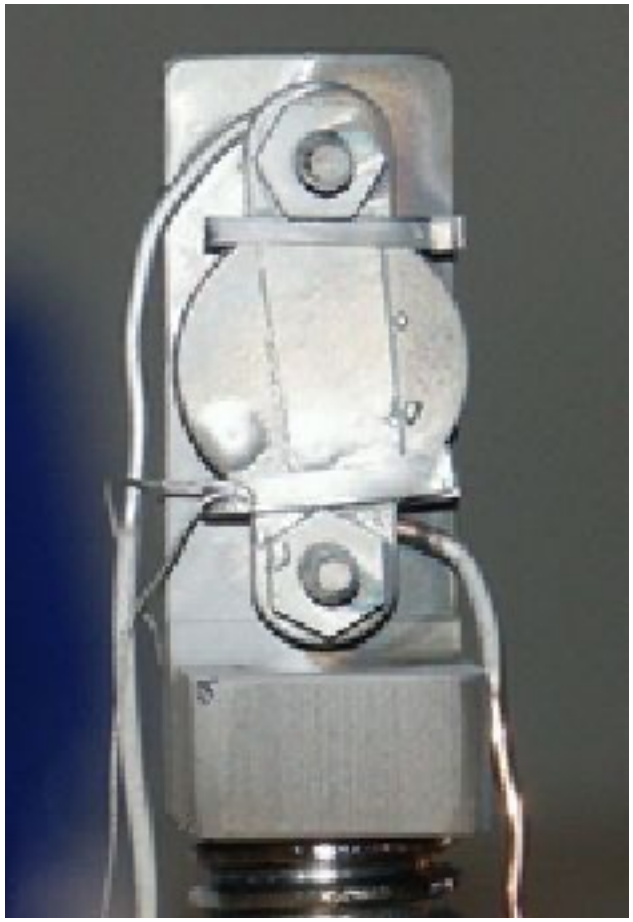








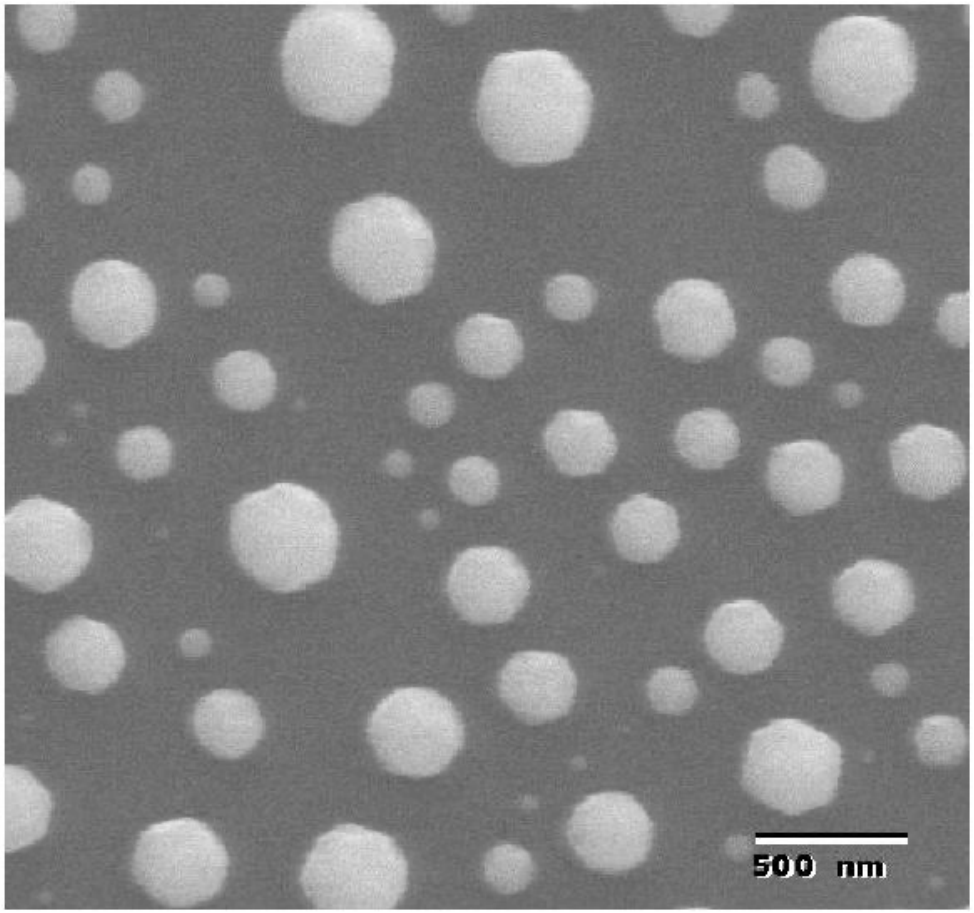



Slices through plan view SEM:



In situ growth of Pb crystals





Good statistics, 3D diffraction data

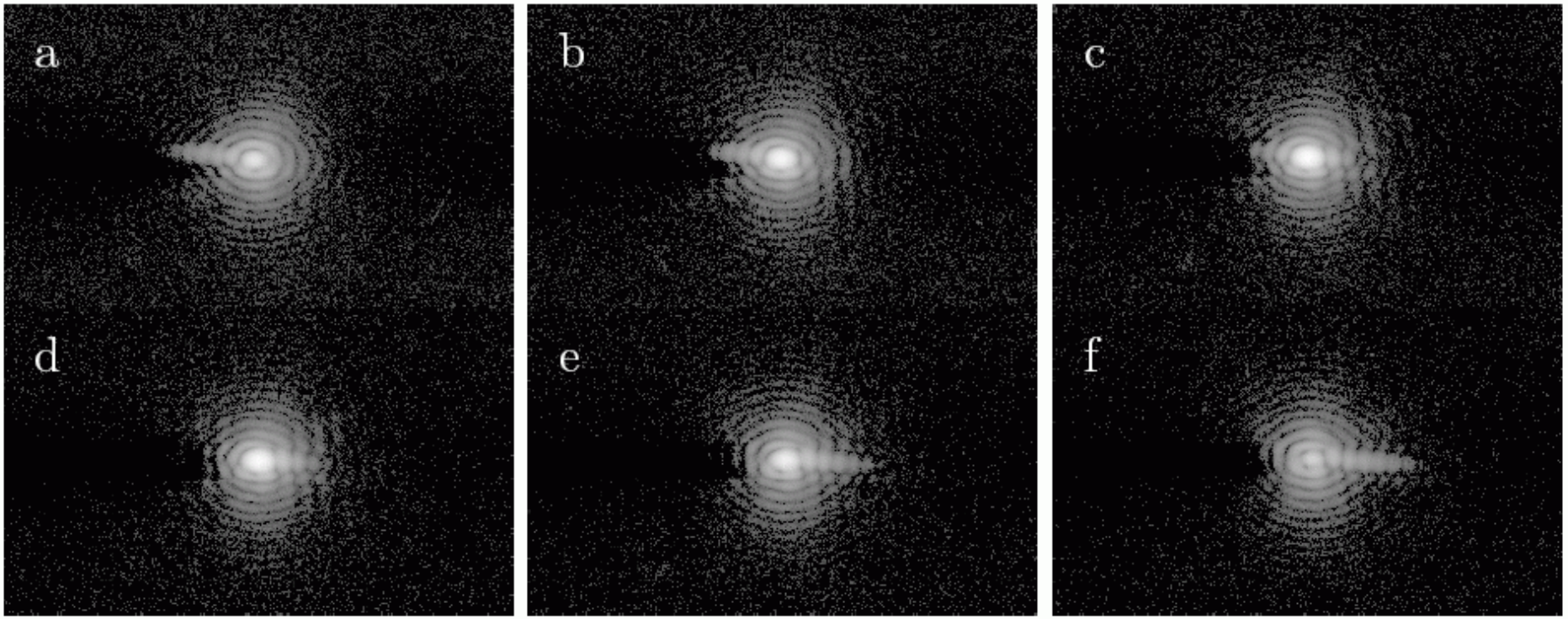
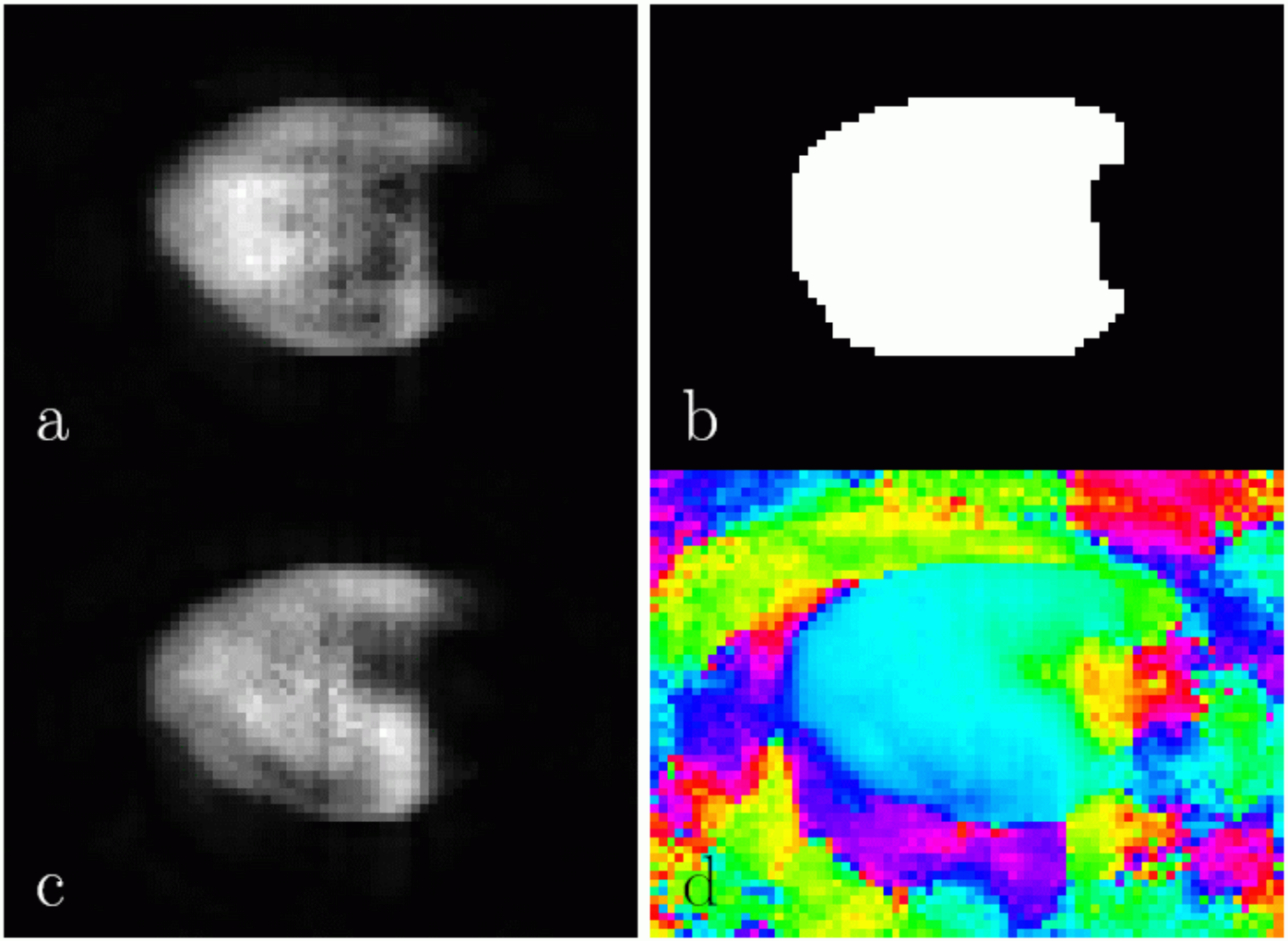


Figure 4.12: Center slices from 3D CXD pattern from Pb sample, on a log scale. Data file 296 from 10/03.

Learn shape of “tight” support



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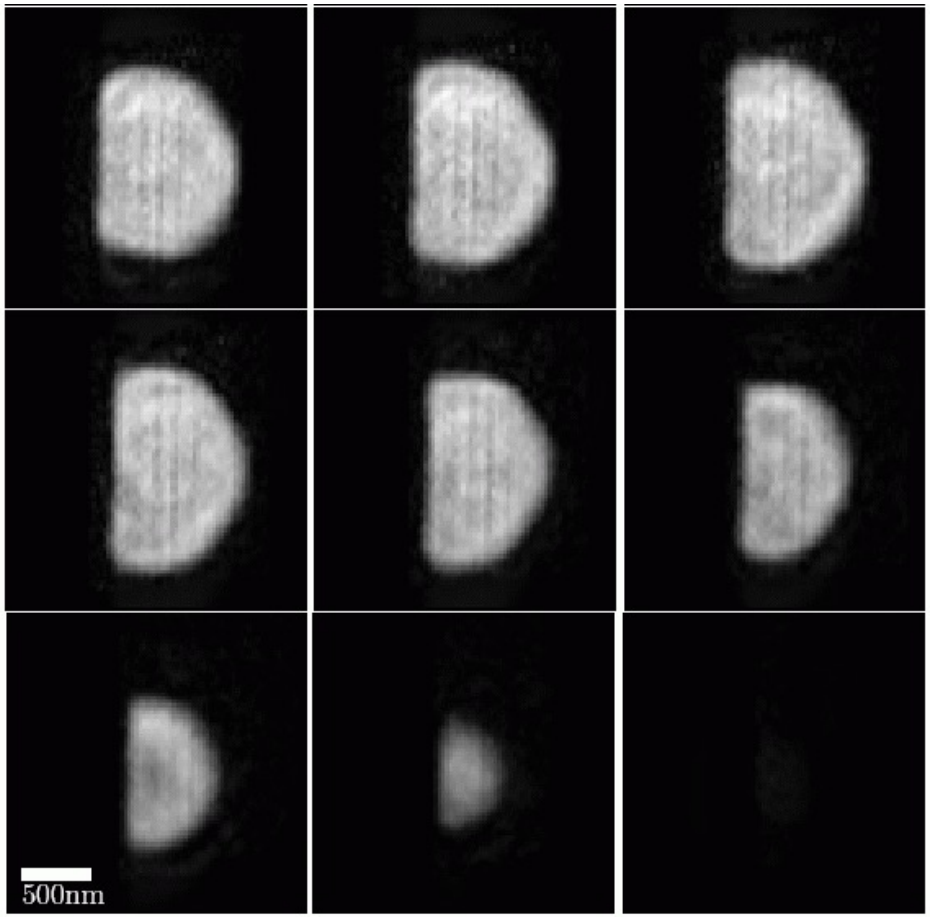
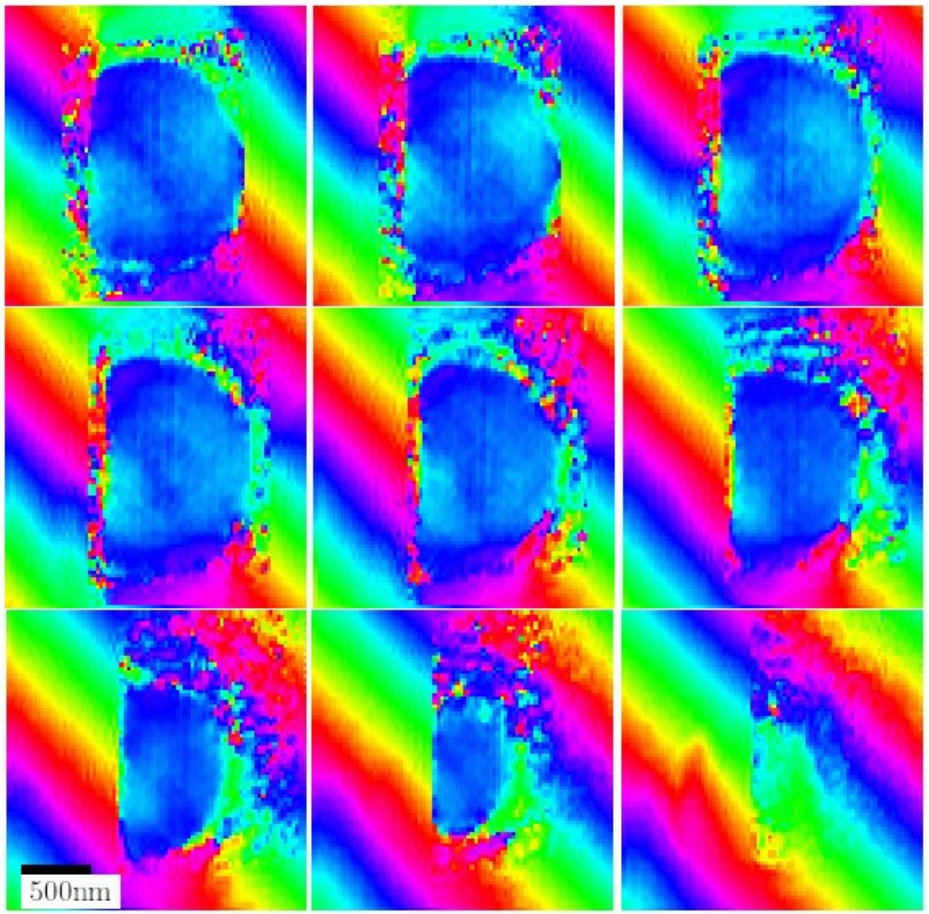
Then refine amplitude *and* phase





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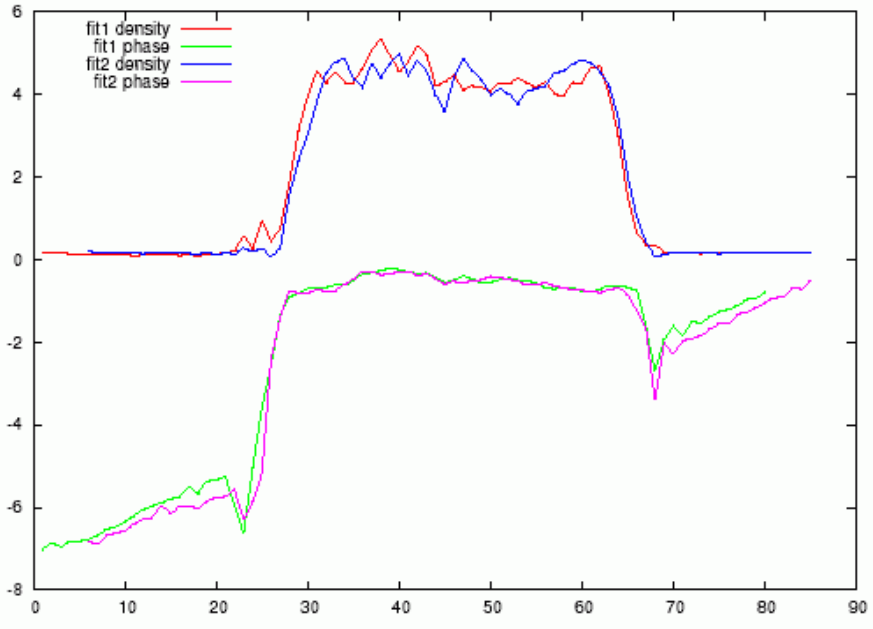
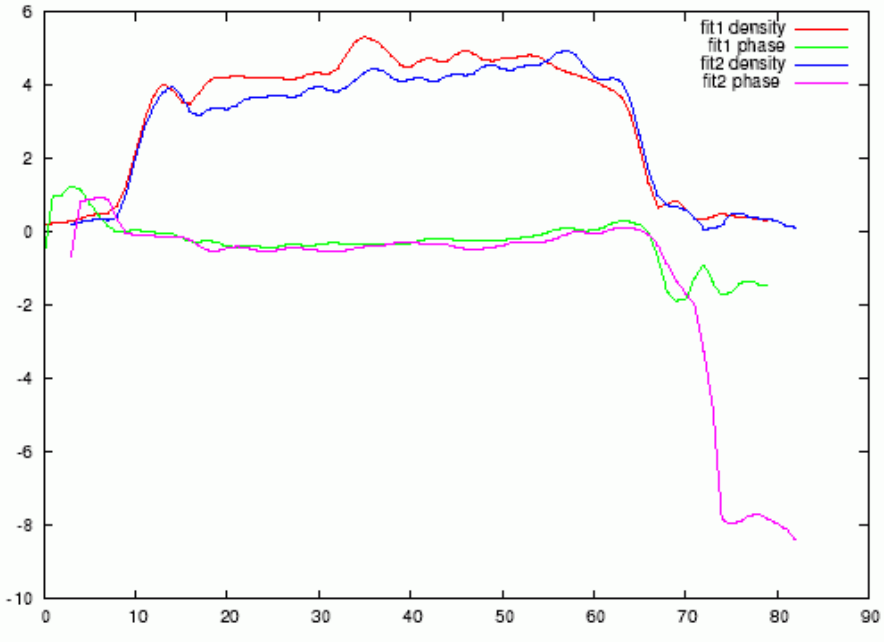
Amplitude/phase cross sections

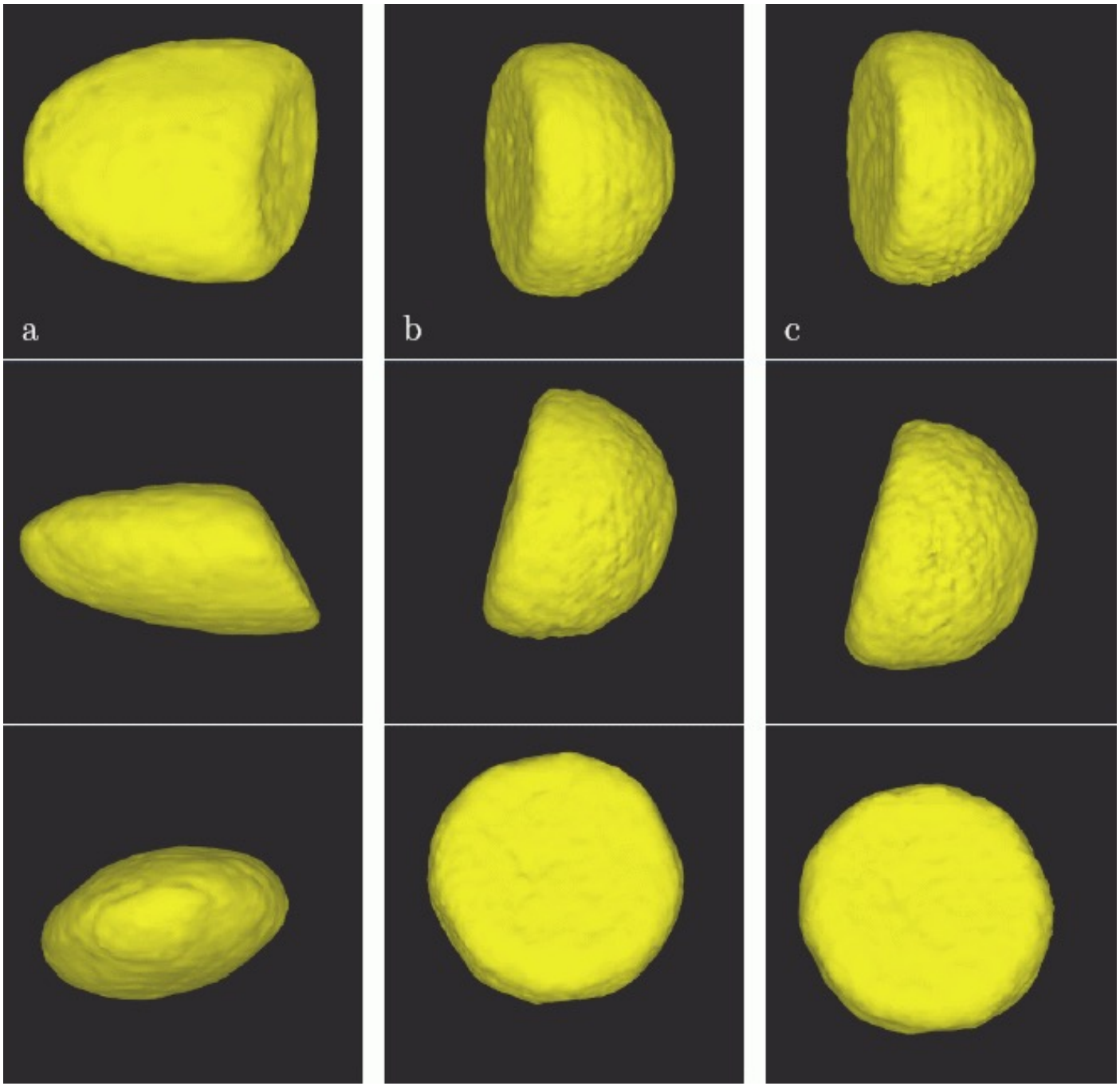
x-striations do not reproduce



x-coordinate



y-coordinate



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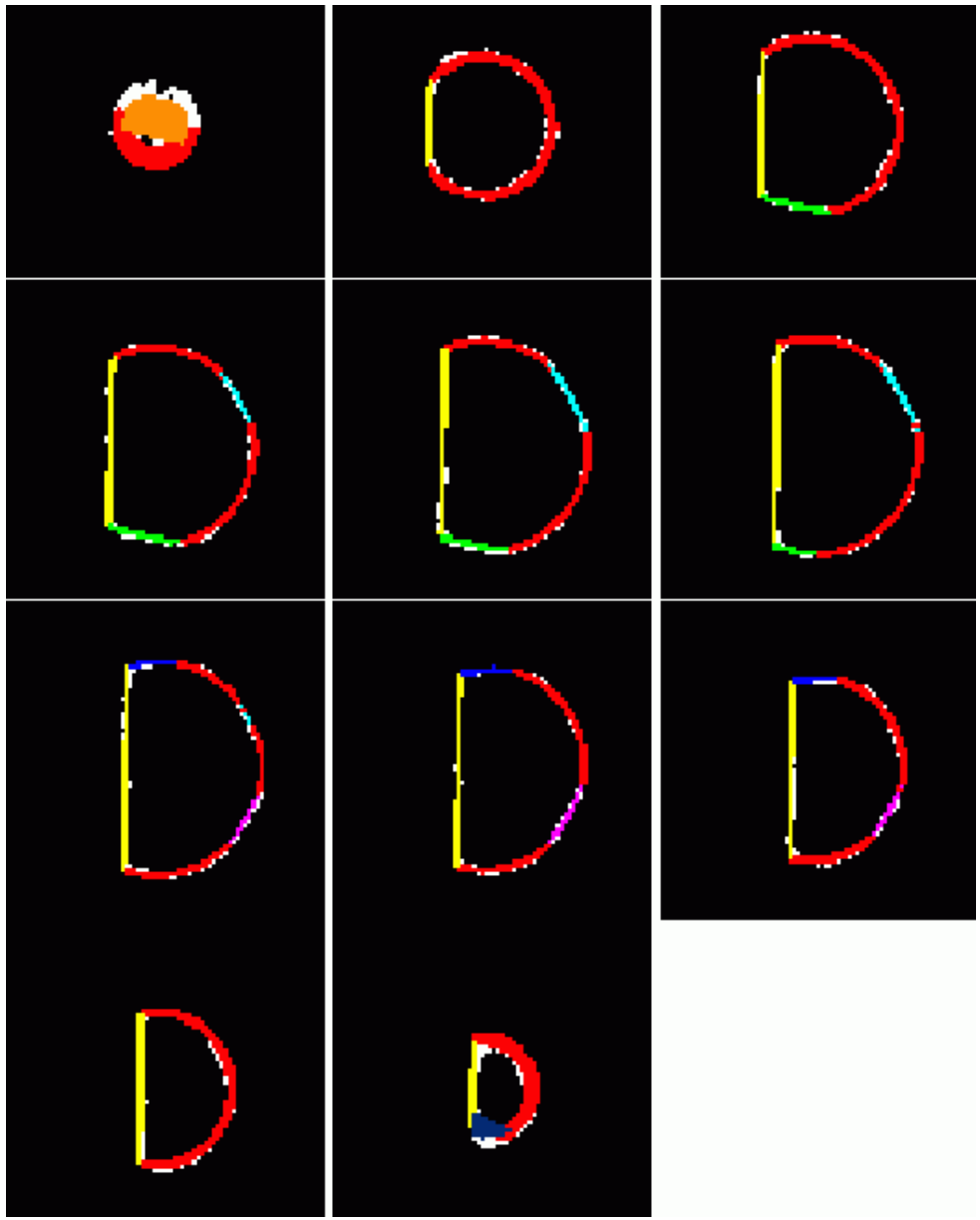
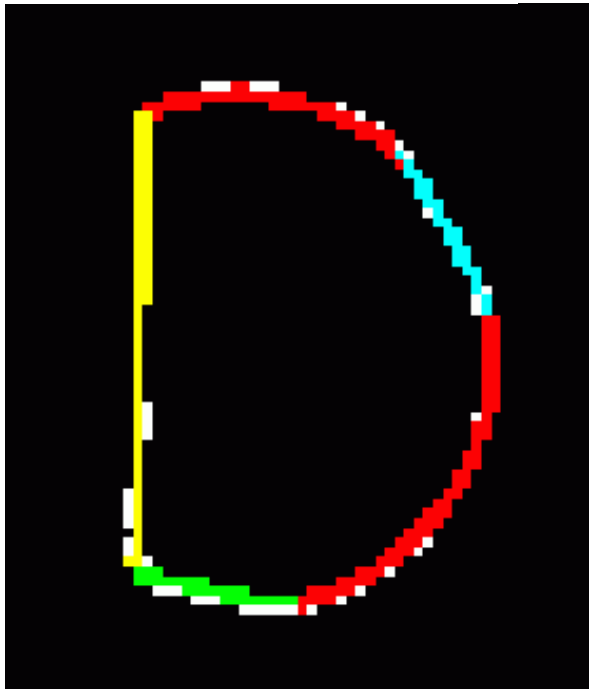
Fitting to facetted shape



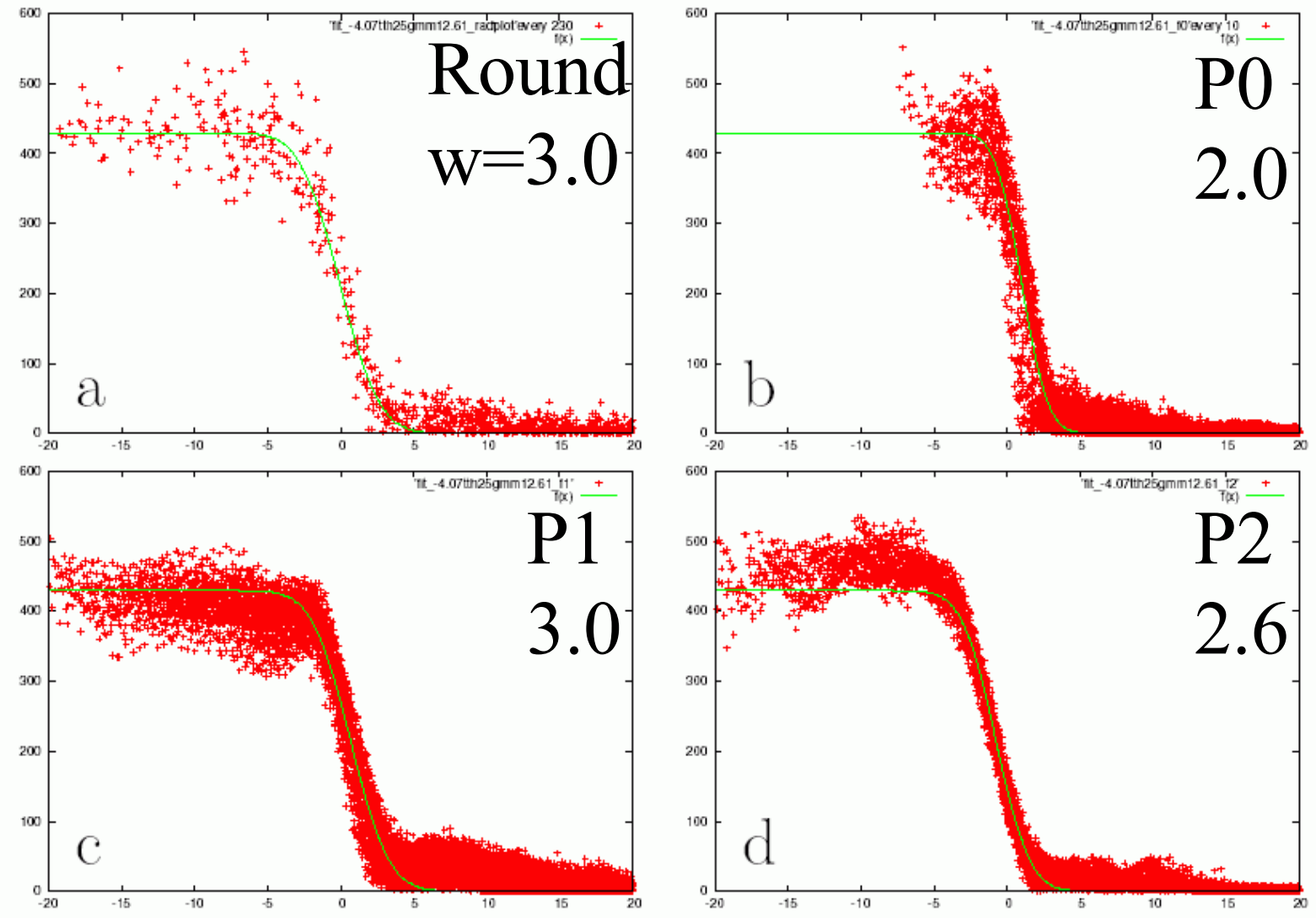
	$\|R\|$		P0	P1	P2	P3	P4	P5	P6
	9.4	P0	0	85	149	79	134	106	71
	25.7	P1		0	123	164	83	76	102
	25.1	P2			0	72	67	74	110
	25.9	P3				0	111	106	76
	25.4	P4					0	113	68
	25.4	P5						0	176
	26.0	P6							0 0

Angles between facets

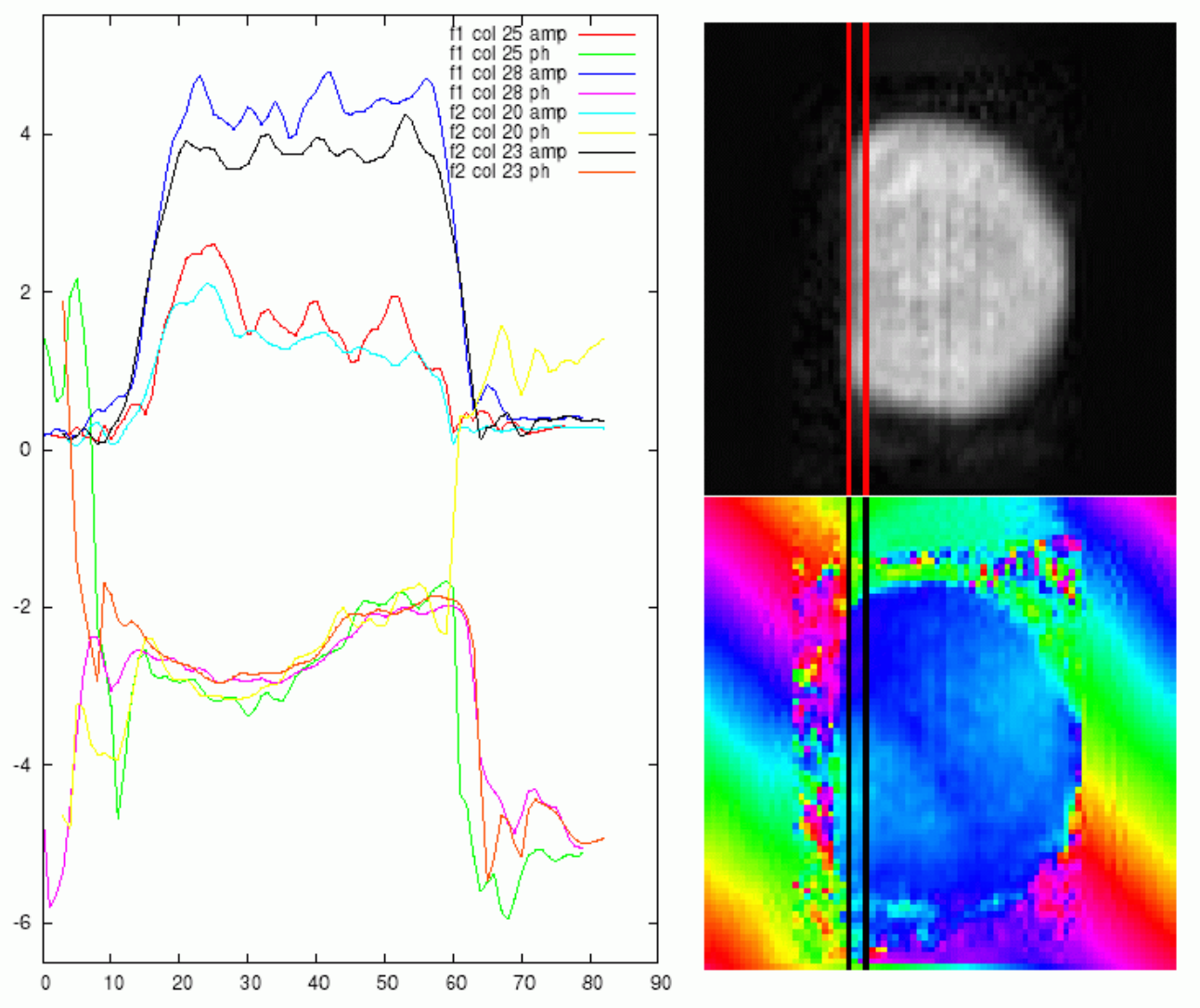


Schloss Ringberg

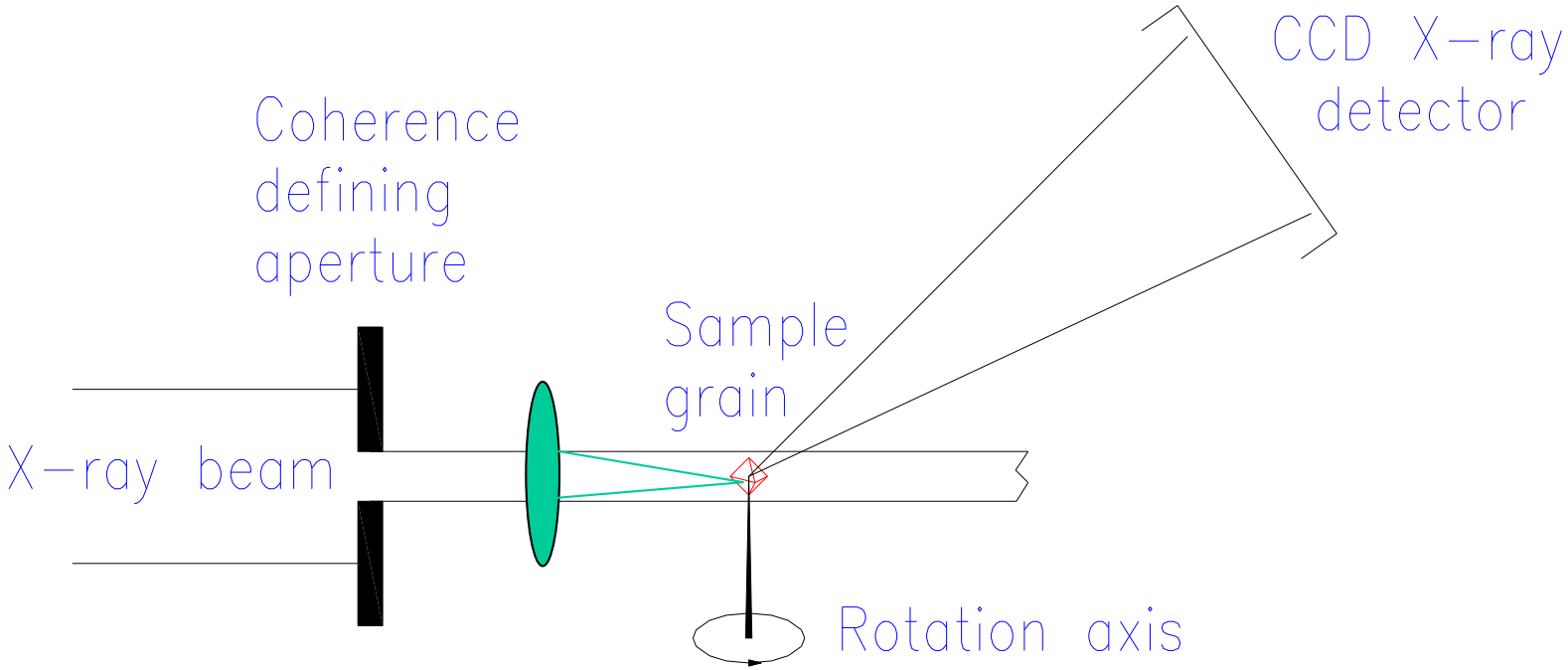
Density distribution across surface



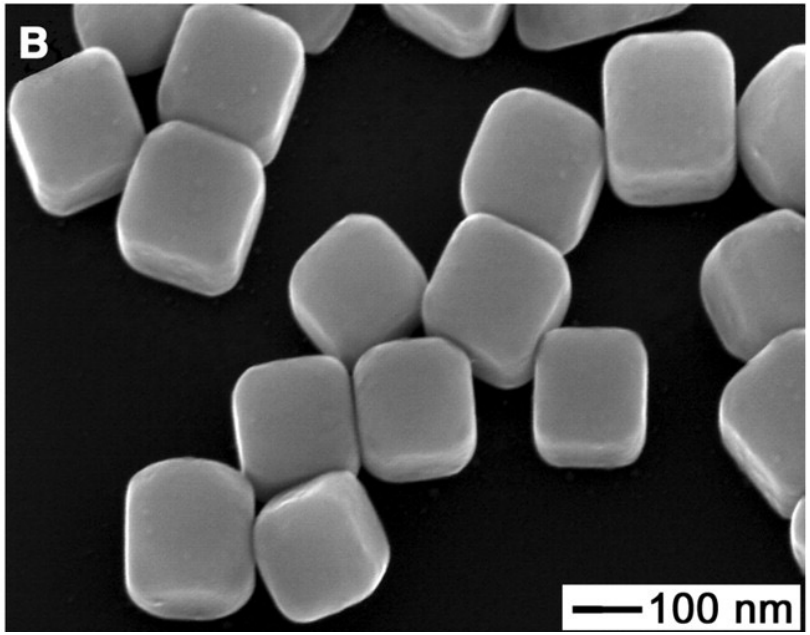
Phase structure near substrate interface

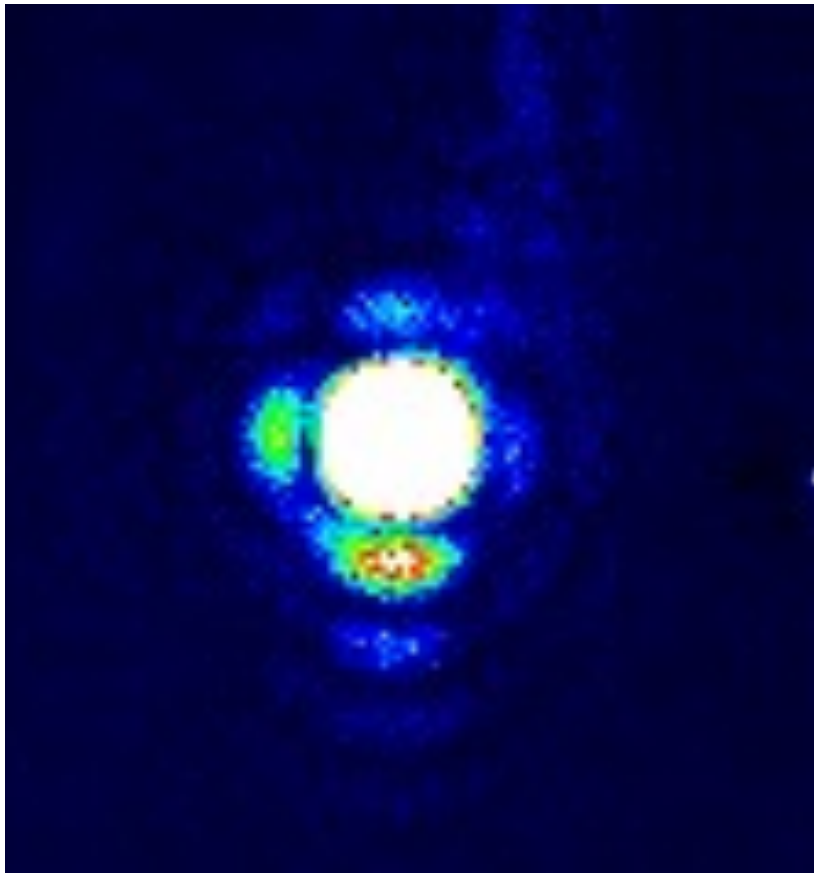


Lensless X-ray Microscope



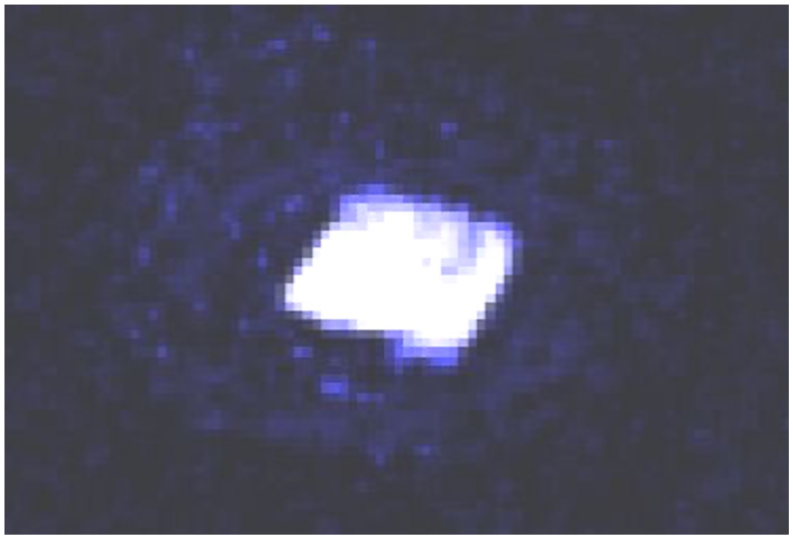
CXD from Silver Nanocubes



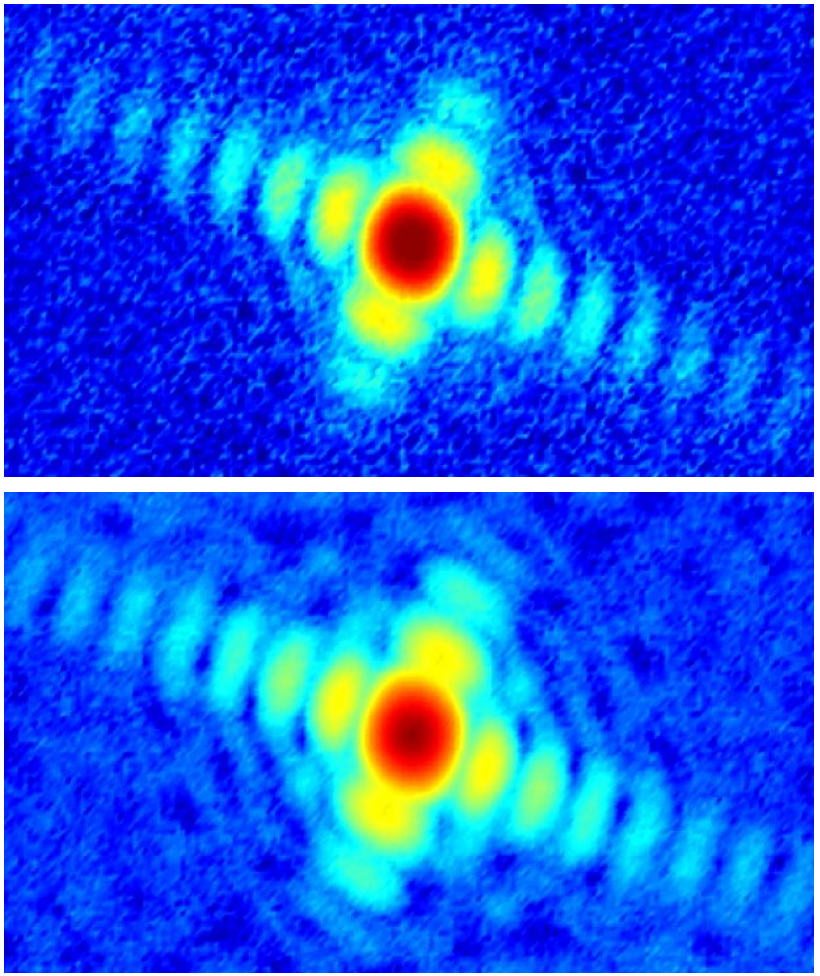


Yugang Sun and Younan Xia,
Science 298 2177 (2003)

Reconstruction of Ag Nanocrystal



200nm



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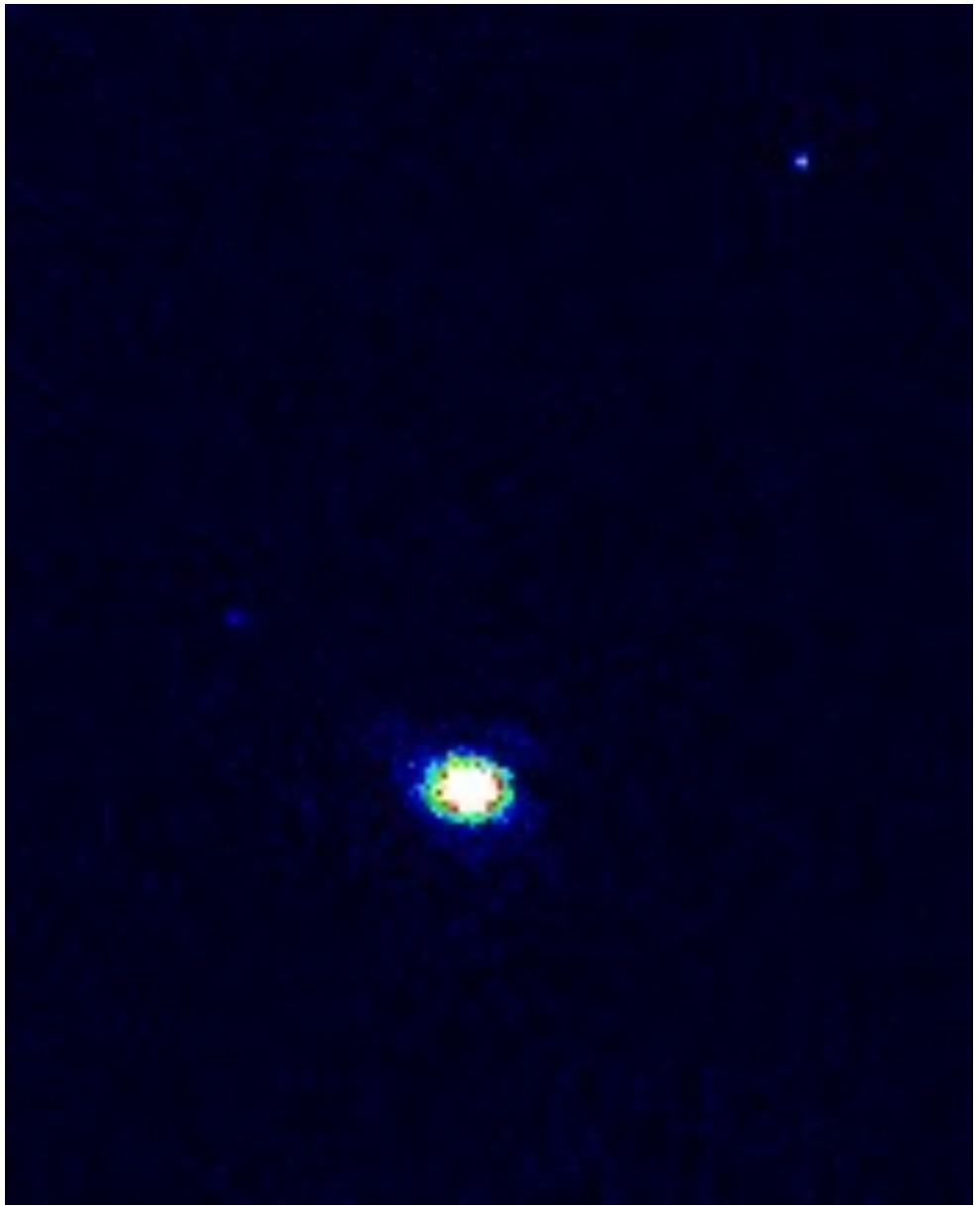
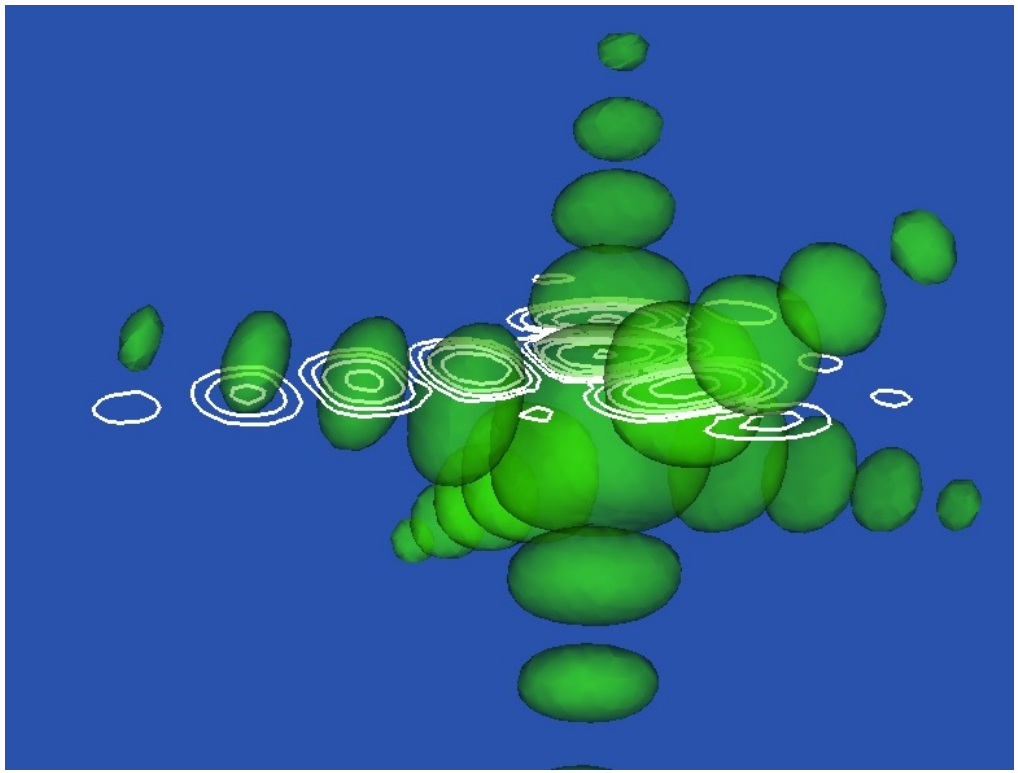
Rocking scan of Ag cubes with 0.01° steps



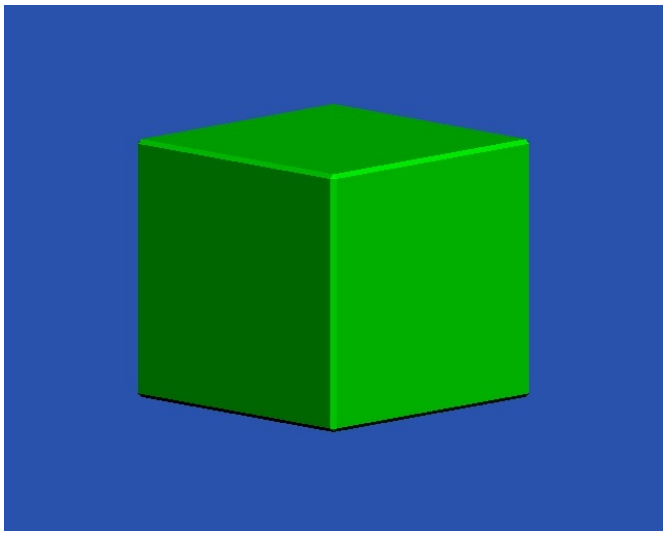
Miscentered Non-Centrosymmetric Reciprocal Space Slice

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

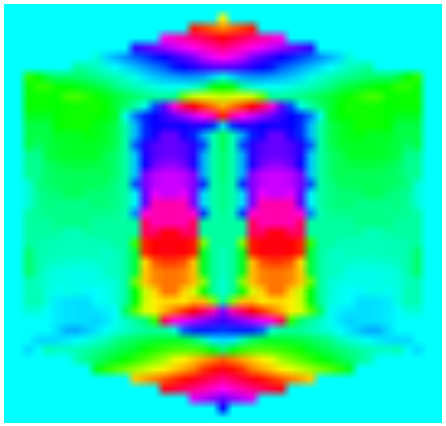


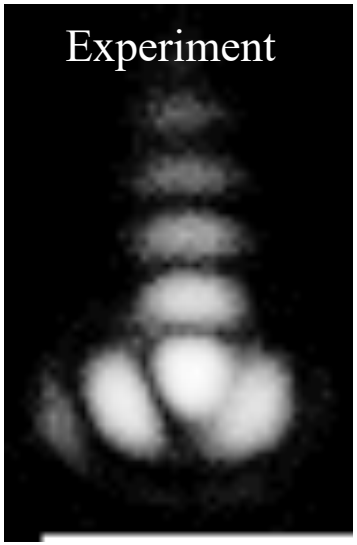
Real-space phase.

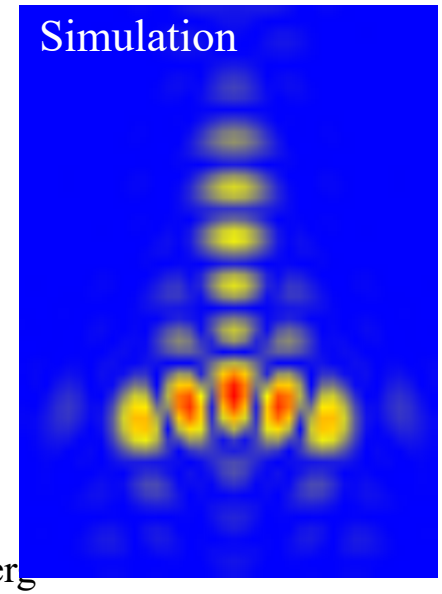
What can we expect to see?



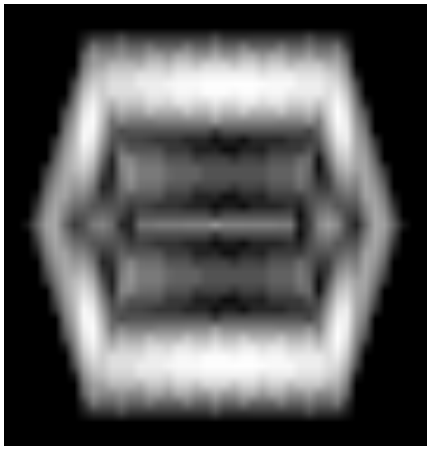
$$A(\bar{q}_o+\bar{q}_z)=\sum_i \sum_z \underbrace{\rho(x,y,z)}_{u_i(x,y)} e^{2\pi i \bar{q}_z\cdot\bar{z}_i} e^{2\pi i(\bar{q}_o)\cdot\bar{r}_i}$$

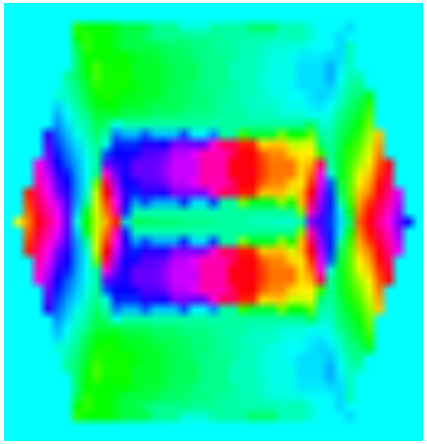


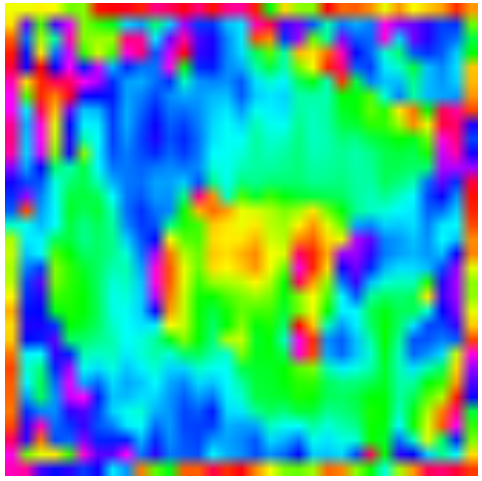


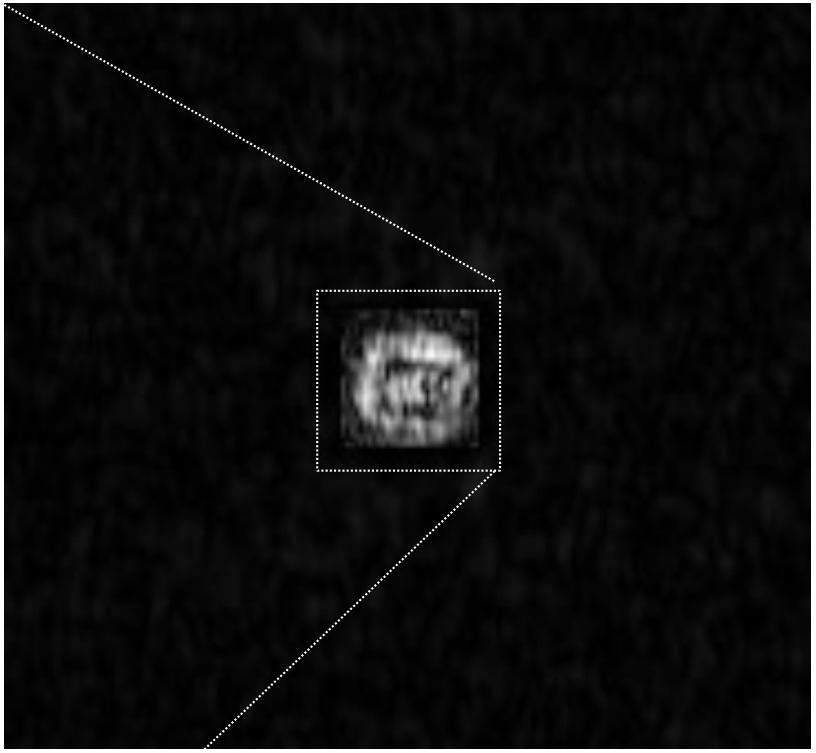


Direct Space Result (1000 Iterations)

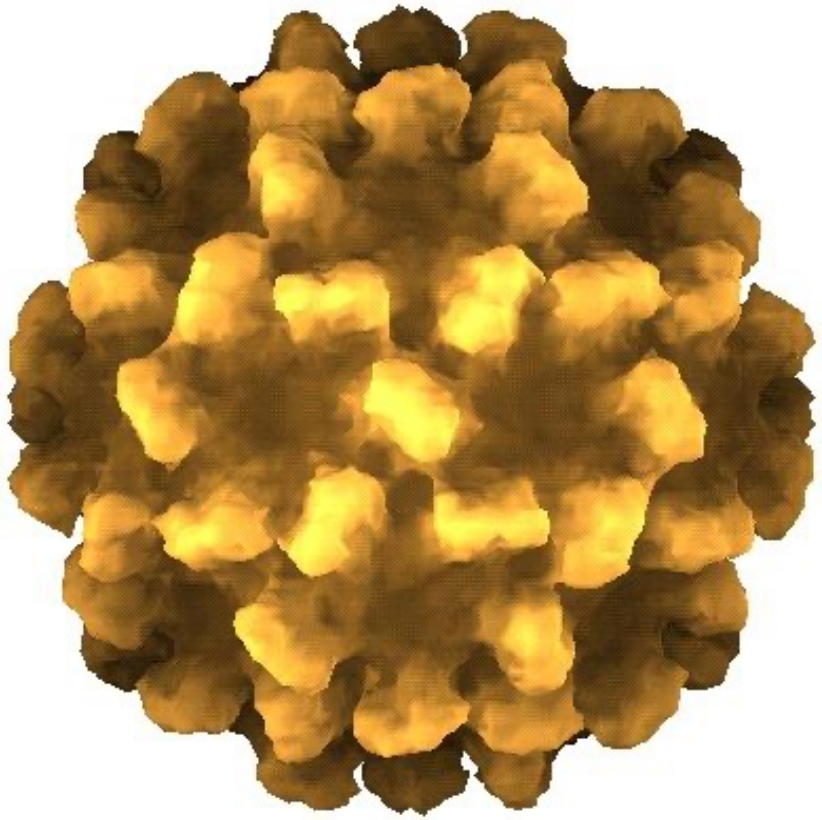








Tomato Bushy Stunt Virus
1980



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Conclusions

- Coherent X-ray Diffraction works even after focusing optics
- Smallest Ferritin crystals about 3 microns
- Implosion due to radiation damage
- Expanded aggregate state upon freezing
- Single molecule diffraction ideas can be tested by electron diffraction
- Phase objects ‘equally’ accessible