

Coherent Diffraction from Surfaces and Interfaces

I. K. Robinson

University College London

Diamond Light Source

Winter school on Synchrotron Radiation
Hanse Wissenschaftskolleg, Delmenhorst

First UHV Experiments

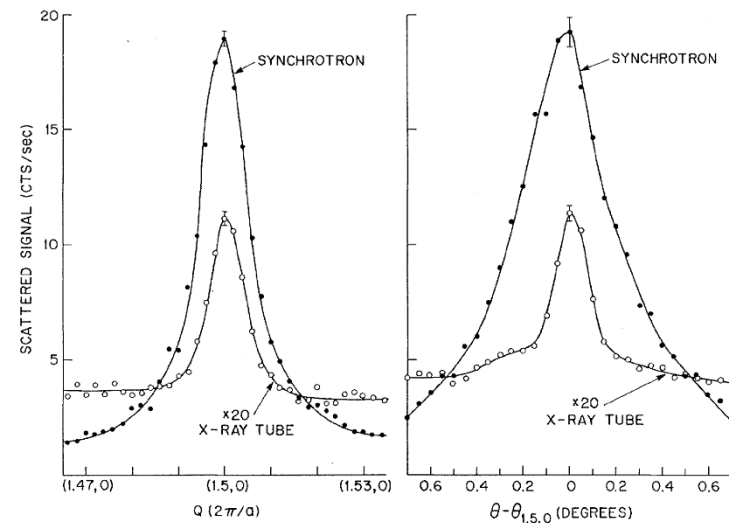
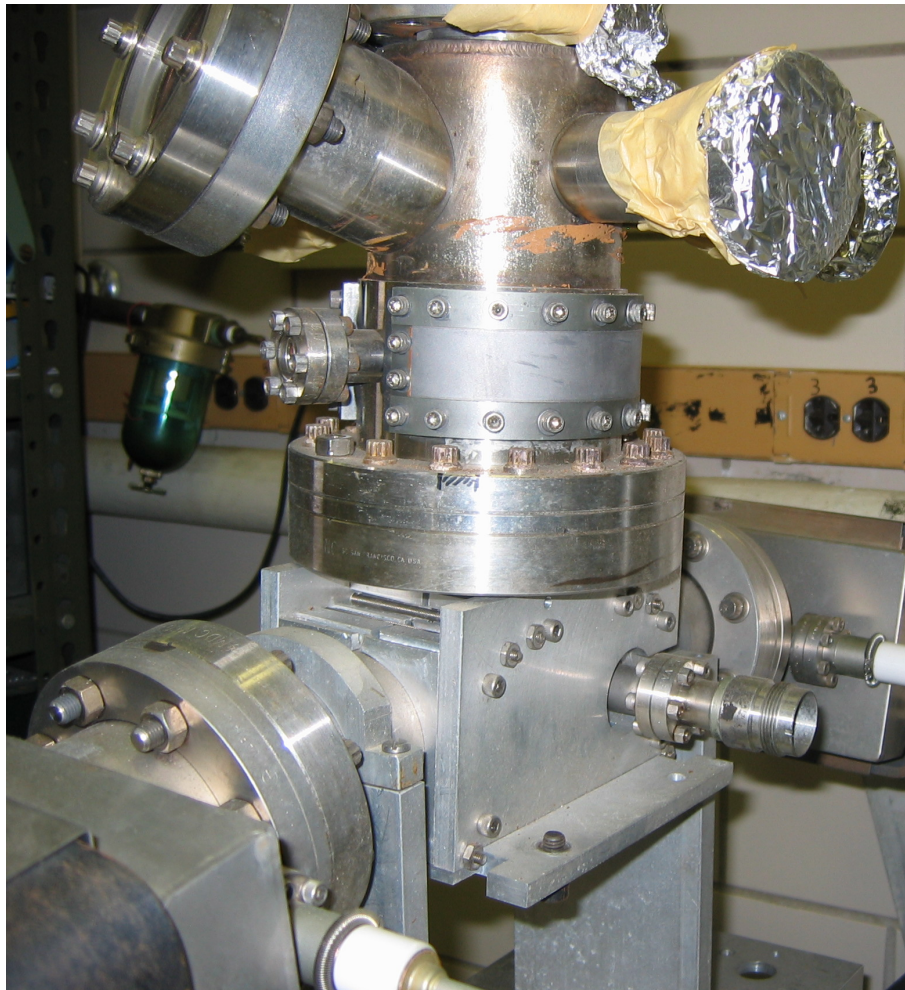
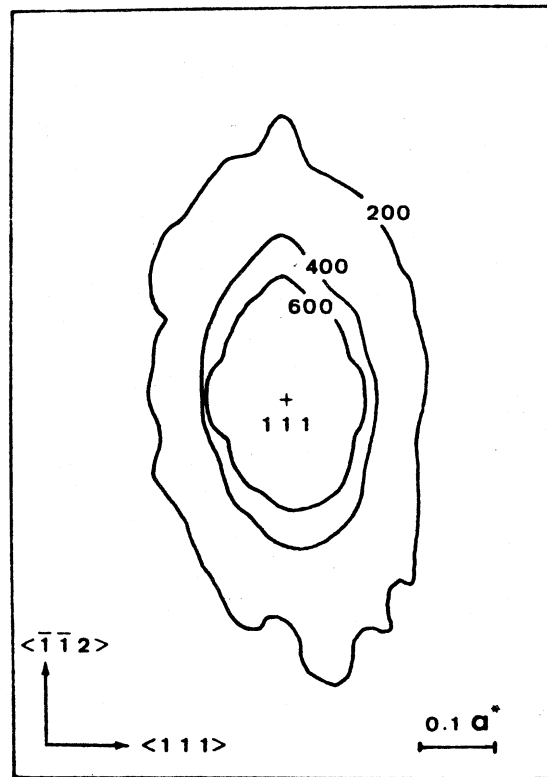


FIG. 1. A plot of the $(\frac{3}{2}, 0)$ Bragg reflection as a function of the momentum transfer $Q(2\pi/a)$ and the crystal's mosaic spread (deg).

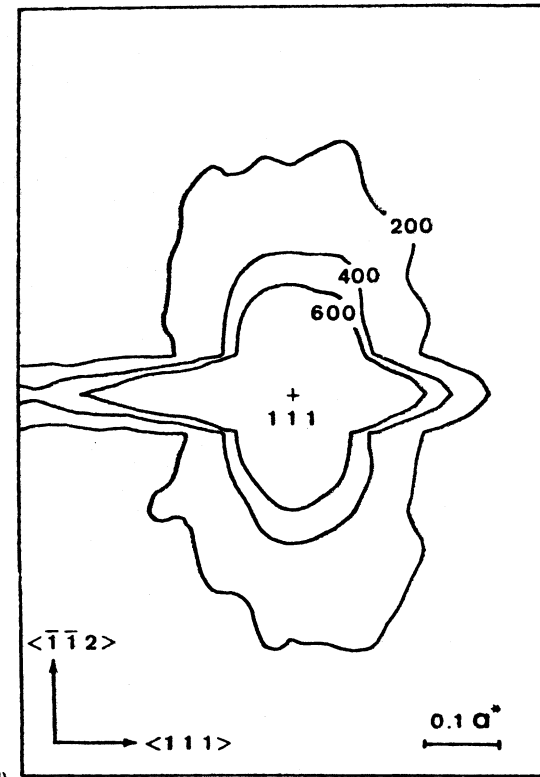
P. Eisenberger and W. C. Marra,
PRL 46 1081 (1981)
experiments done at SSRL

Diffuse Scattering from Si Wafer



(c)

Unpolished wafer



(d)

40 microns removed

N. Kashiwagara, J. Harada and M. Ogino, J. Appl. Phys 54 2706 (1983)

Diffraction as a Surface Integral

**Die äußere Form der Kristalle
in ihrem Einfluß auf die Interferenzerscheinungen
an Raumgittern**

Von M. v. Laue

Annalen der Physik [5] 26 55 (1936)

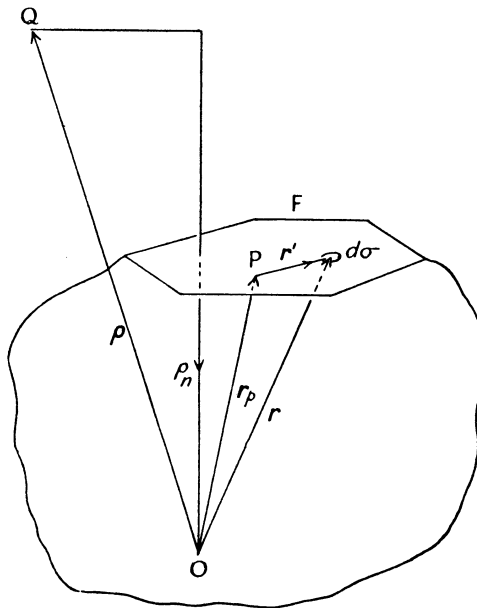
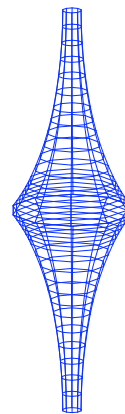


FIG. 200



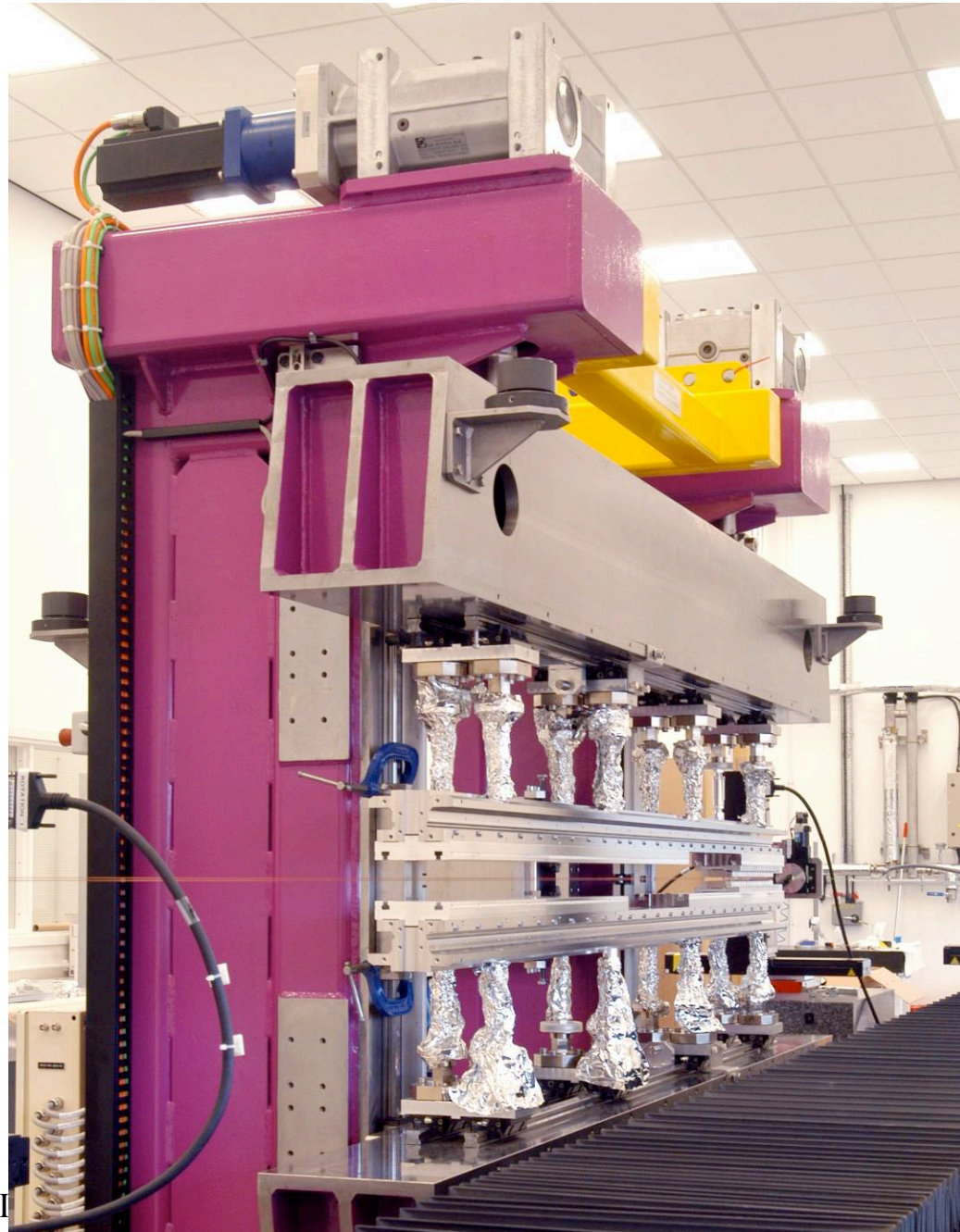
“Stacheln”

Diamond Light Source (RAL)

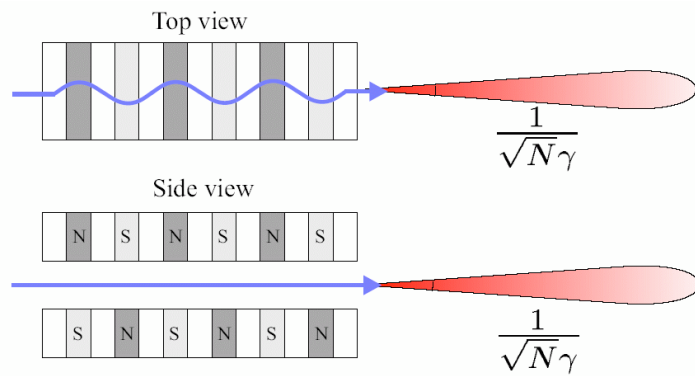


I. K. Robinson Bremen Winter School

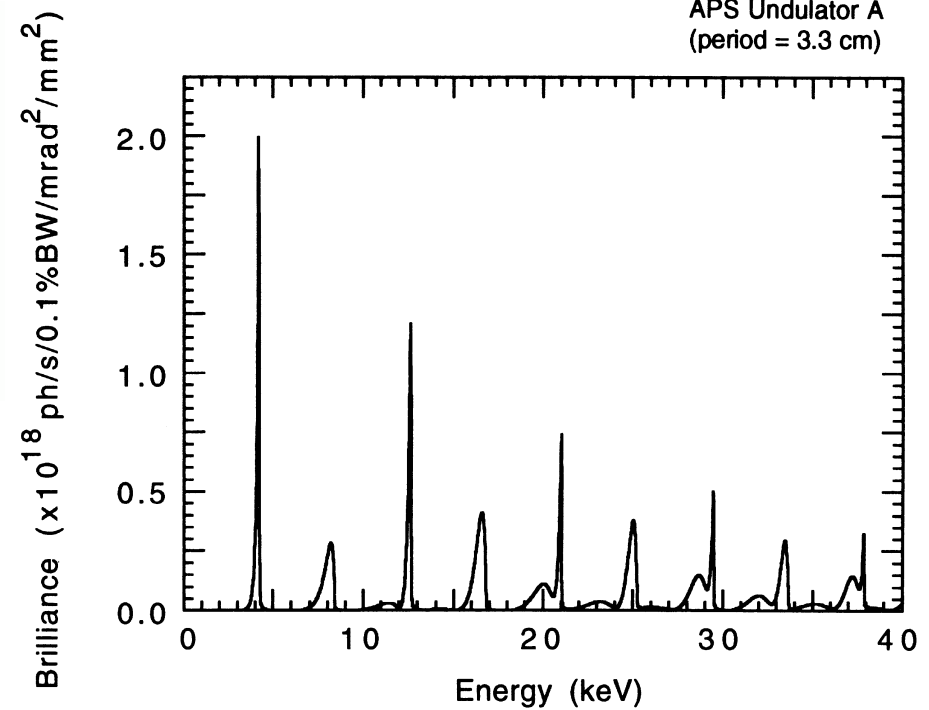
Diamond
in-vacuum
X-ray
Undulator



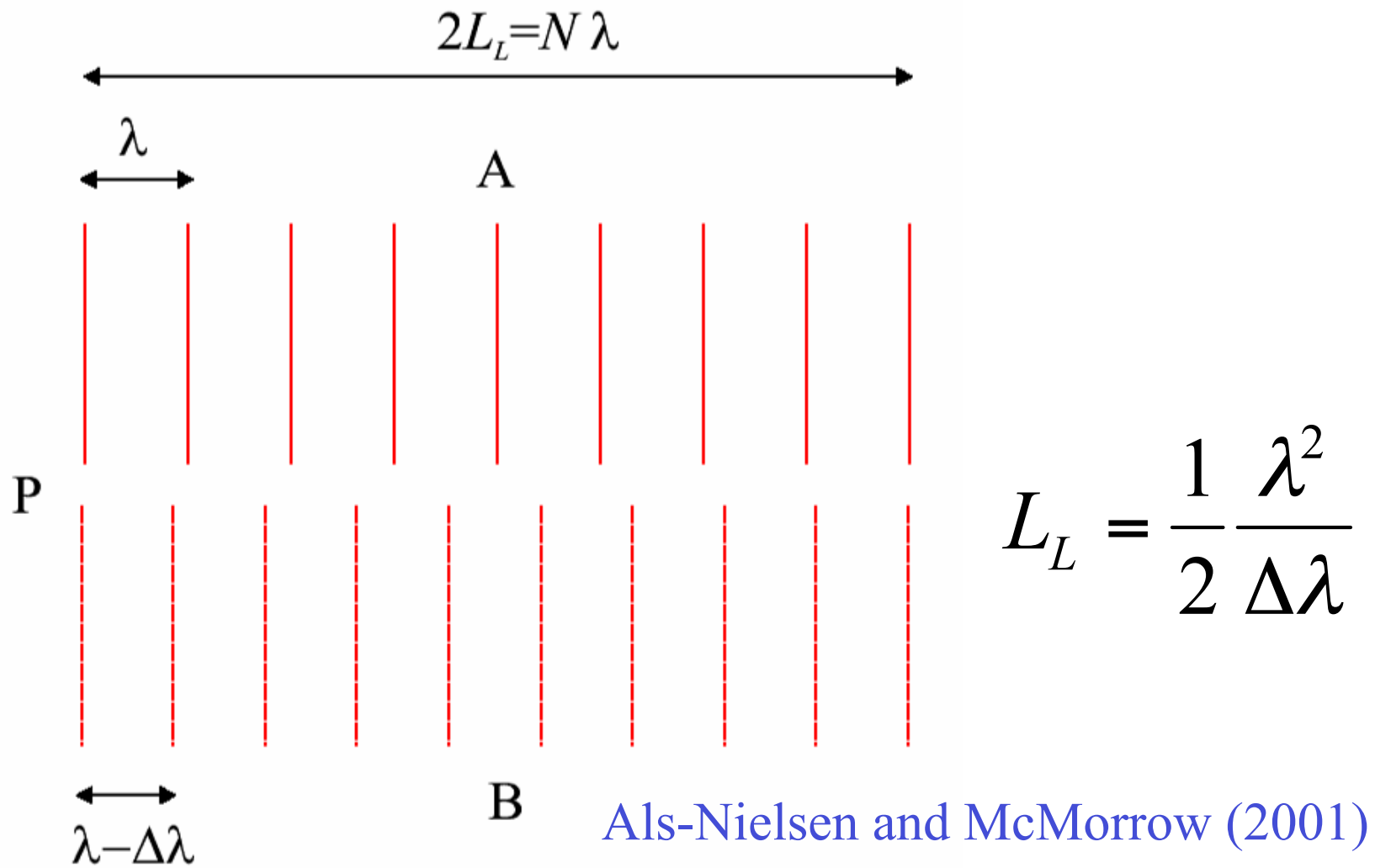
X-ray Undulator Principle



$$\lambda_X = \frac{\lambda_U}{2\gamma^2} \left\{ 1 + \frac{K^2}{2} + (\gamma\theta)^2 \right\}$$

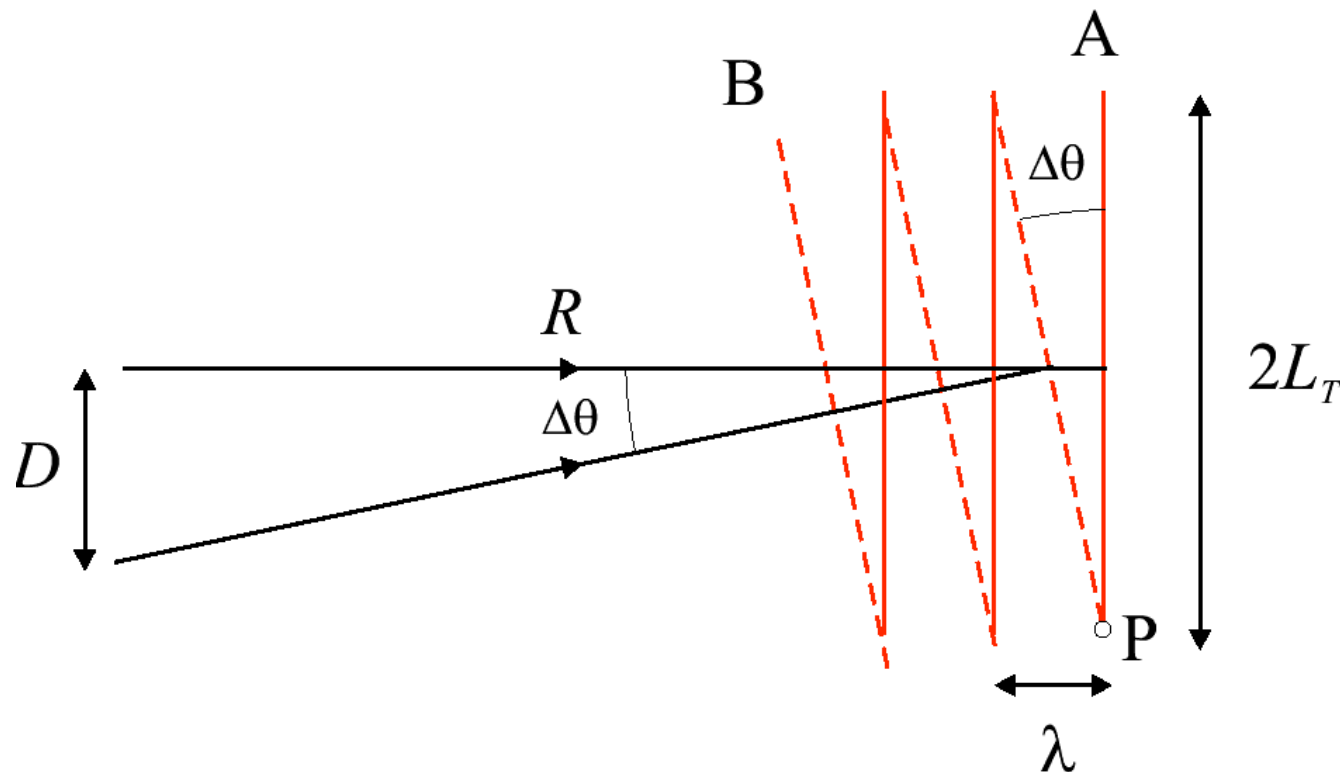


Longitudinal Coherence



Als-Nielsen and McMorrow (2001)

Lateral (Transverse) Coherence



$$L_T = \frac{\lambda R}{2 D}$$

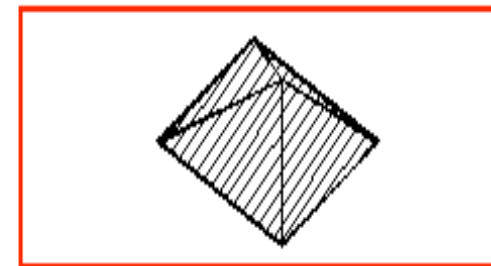
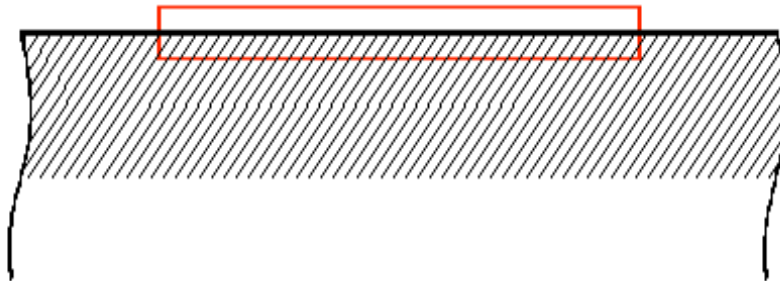
Als-Nielsen and McMorrow (2001)

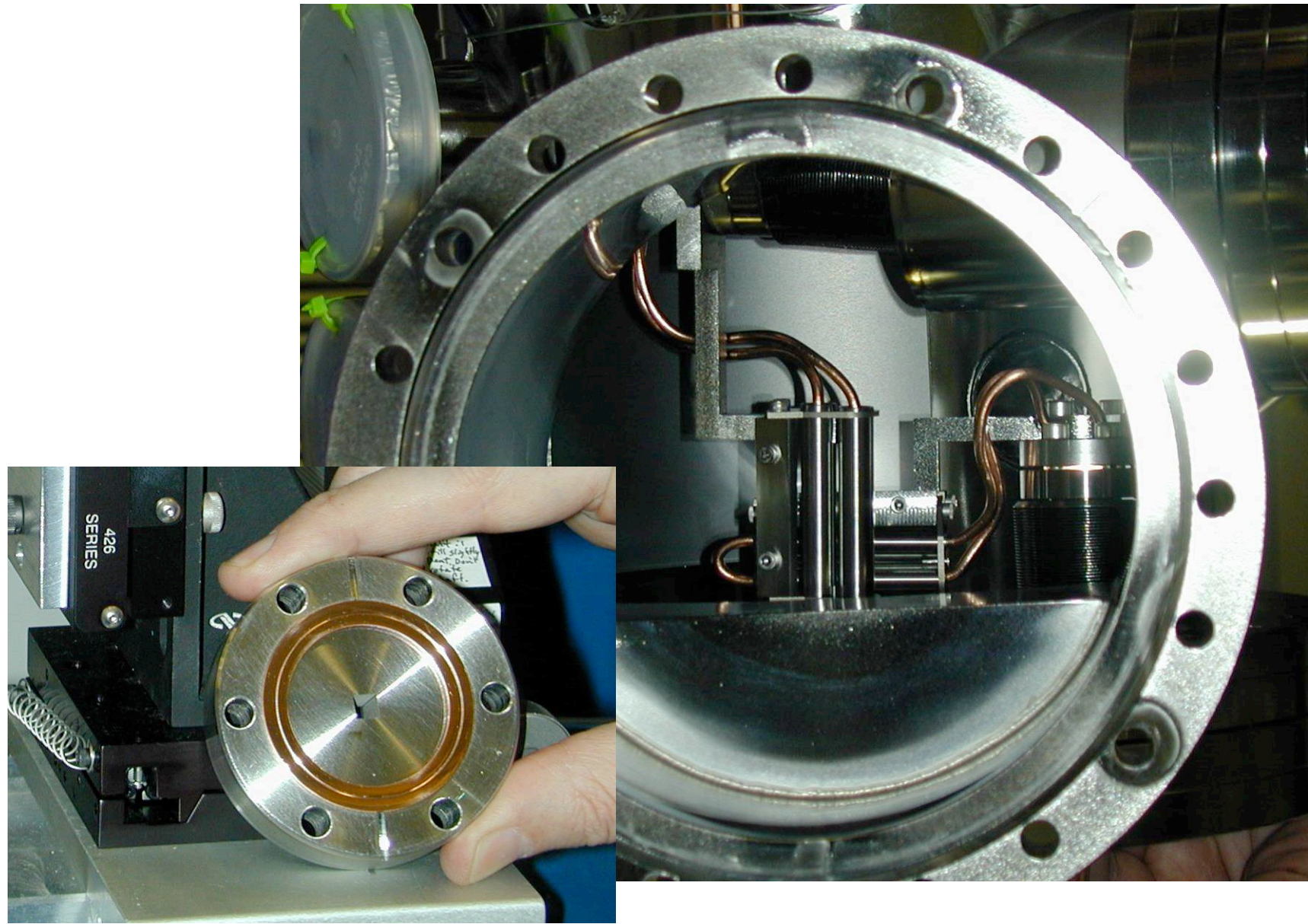
Coherence at APS, ESRF or DLS

Typical of 3rd Generation (undulator) Synchrotron Source

Coherence of	ξ_{VER}	ξ_{HORIZ}	ξ_{LONG}	Flux
Raw Undulator	$35\mu\text{m}$	$9\mu\text{m}$	$0.004\mu\text{m}$	2×10^{12}
Si(111) Monochromator	$35\mu\text{m}$	$9\mu\text{m}$	$1\mu\text{m}$	1×10^{10}
C(111) Monochromator	$35\mu\text{m}$	$9\mu\text{m}$	$3\mu\text{m}$	3×10^9

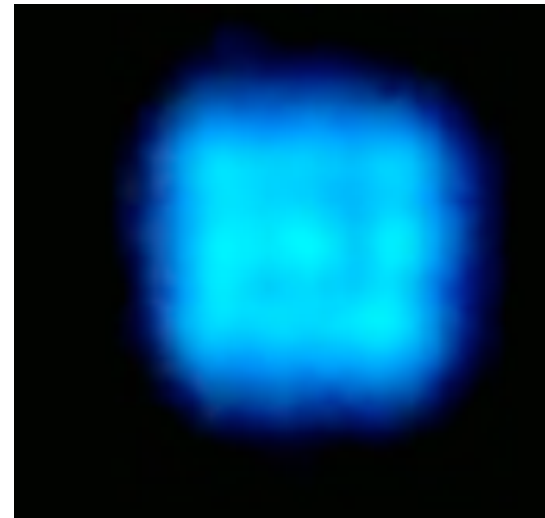
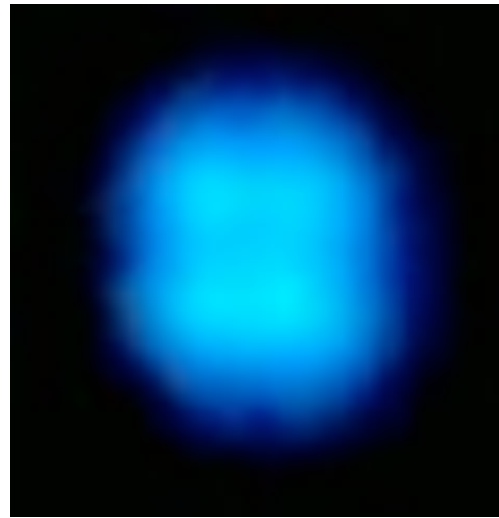
Coherent region defined by slits



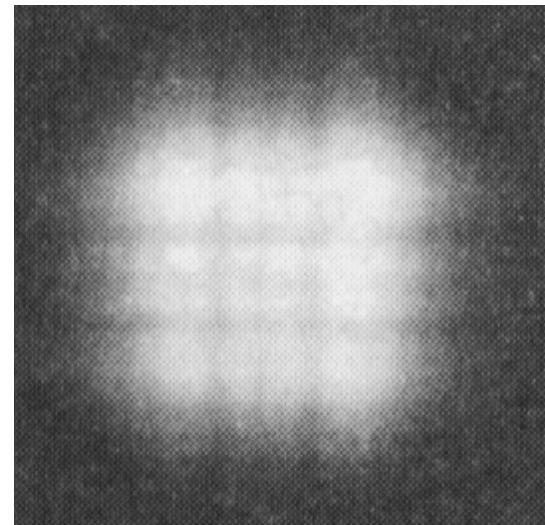
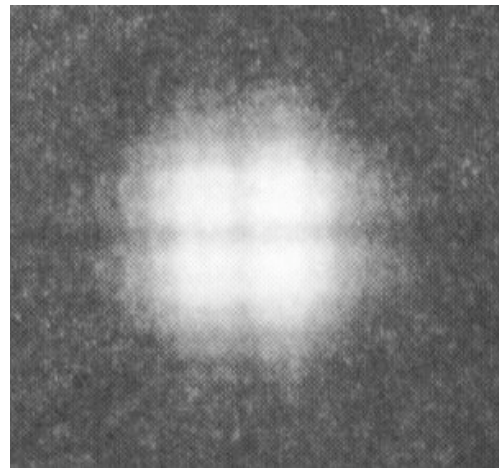


I. K. Robinson Bremen Winter School

Fresnel Diffraction when $d^2 \sim \lambda D$



X-ray
beam
defined
by RB
slits

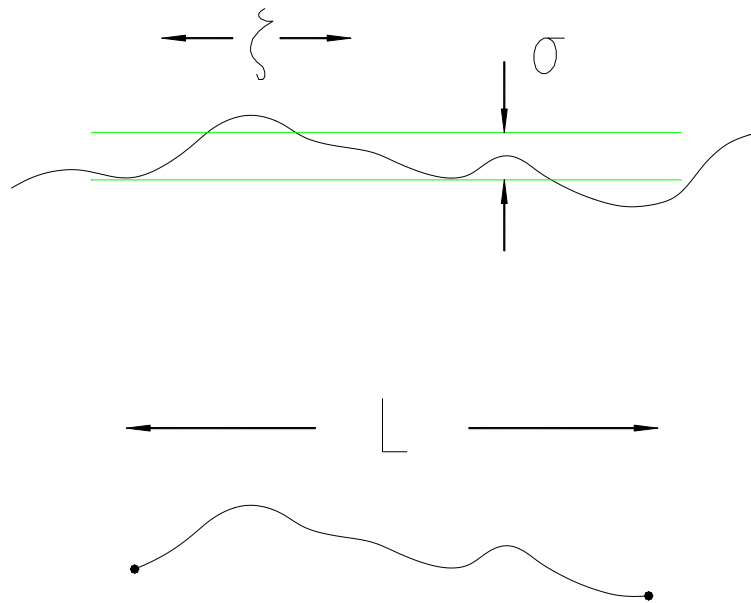


Visible
Fresnel
diffraction
from
Hecht
“Optics”

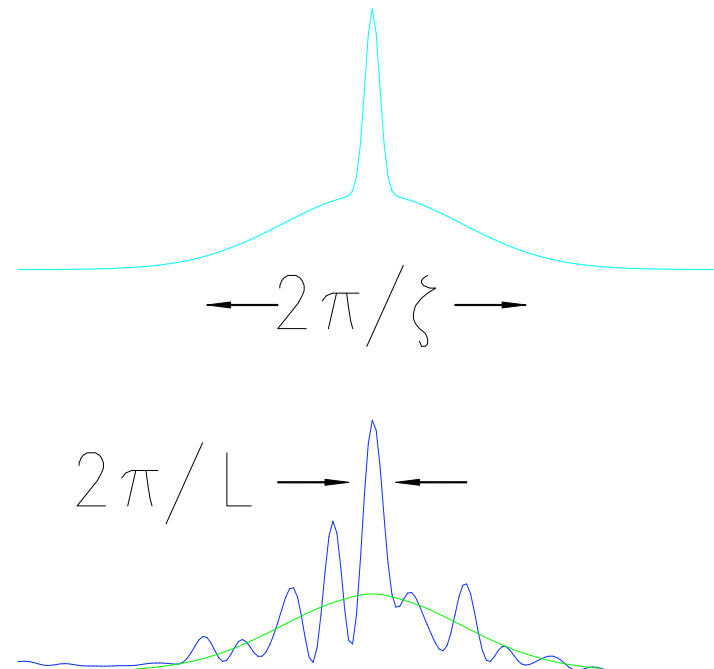
I. K. Robinson

Diffuse Scattering acquires Structure using CXD

Real Space



Reciprocal Space

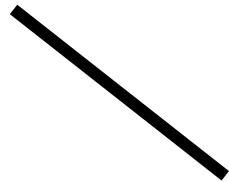


Rods, sheets and dots

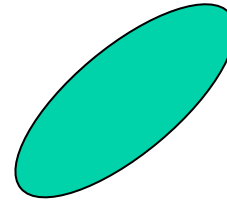
0D



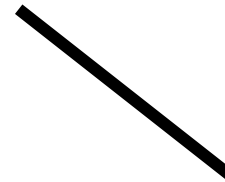
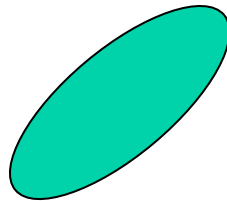
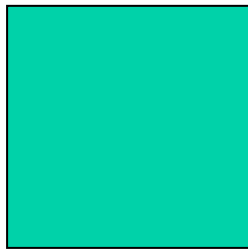
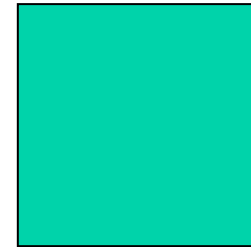
1D



2D



3D



Finite rods, sheets and dots

0D



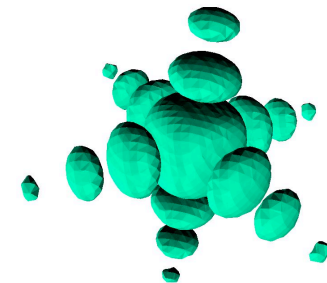
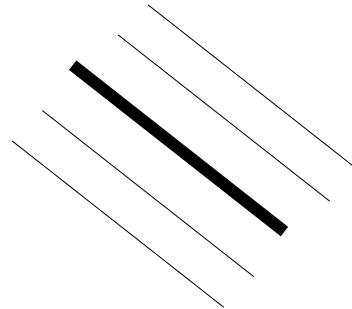
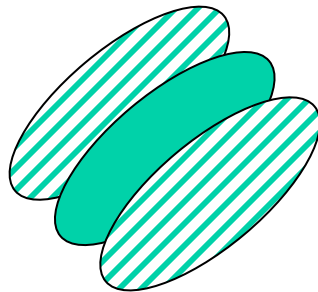
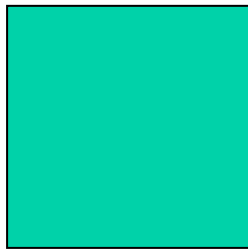
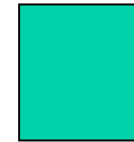
1D



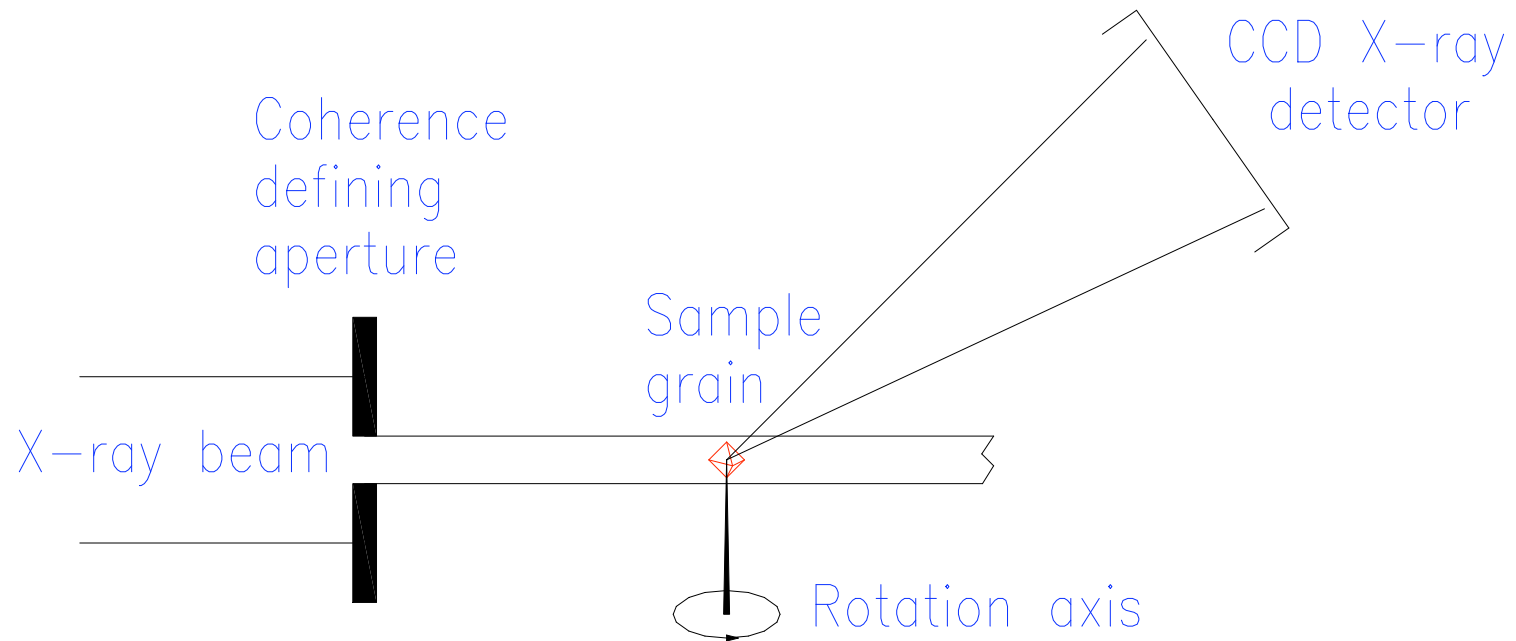
2D



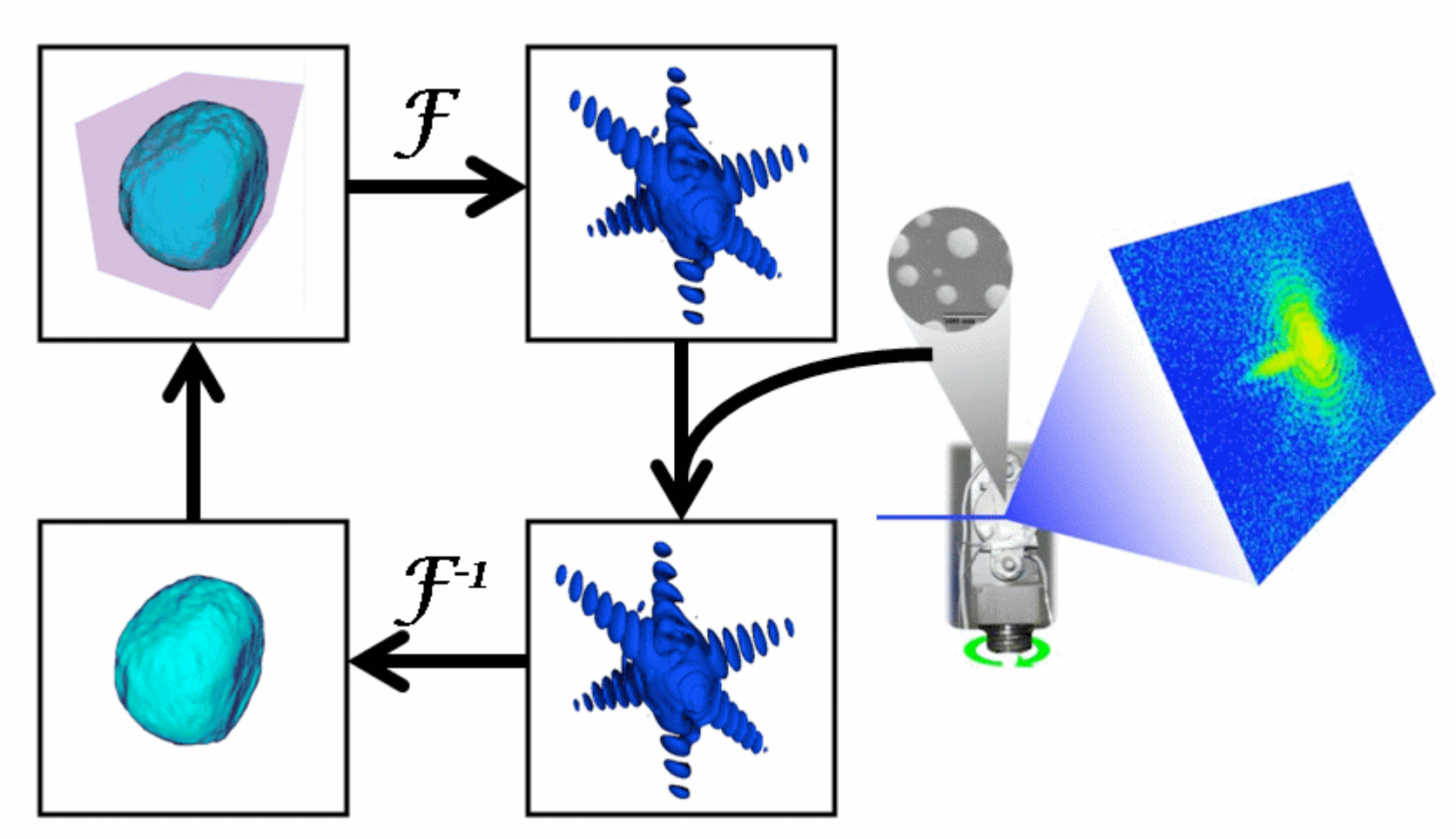
3D



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)

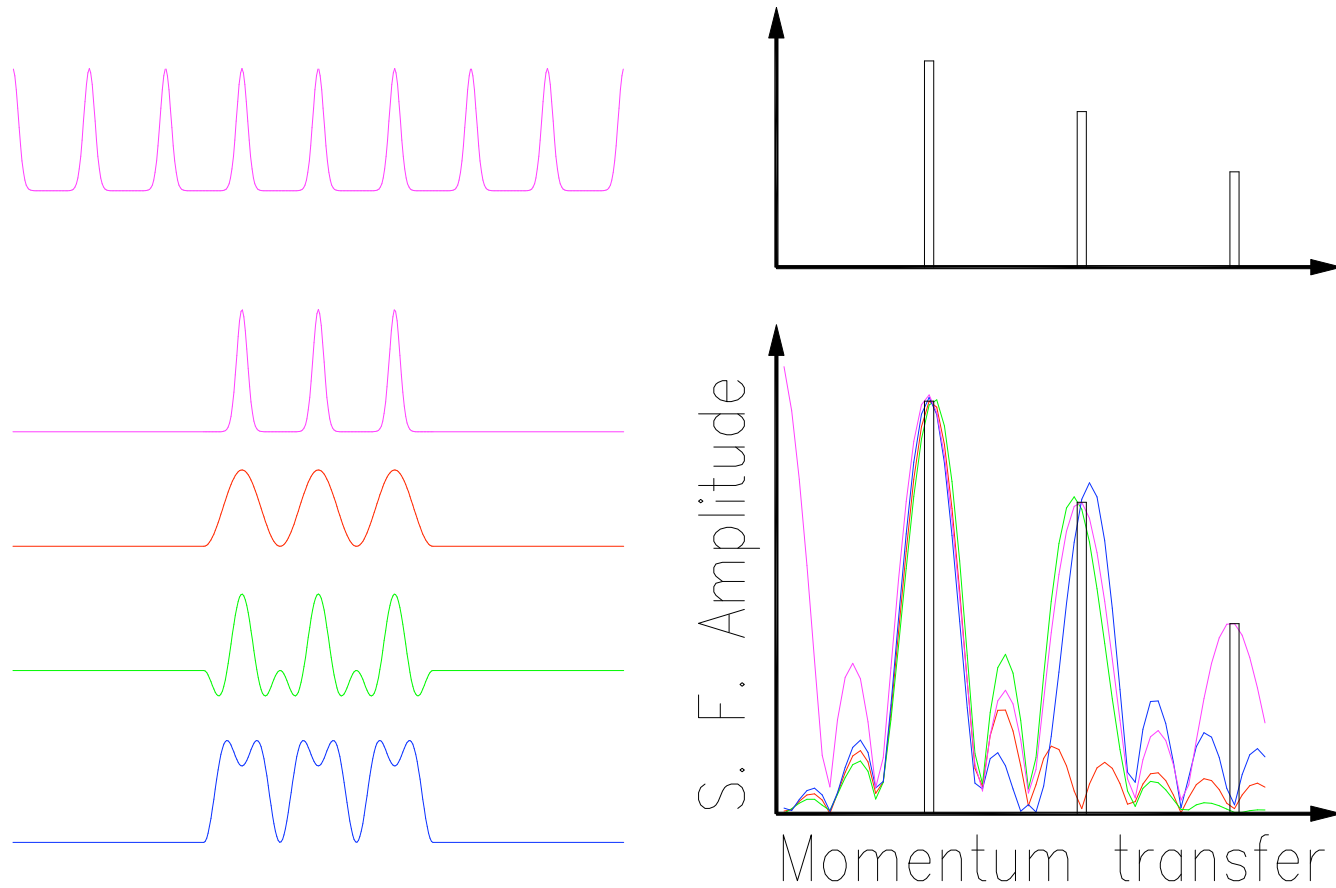
I. K. Robinson Bremen Winter School

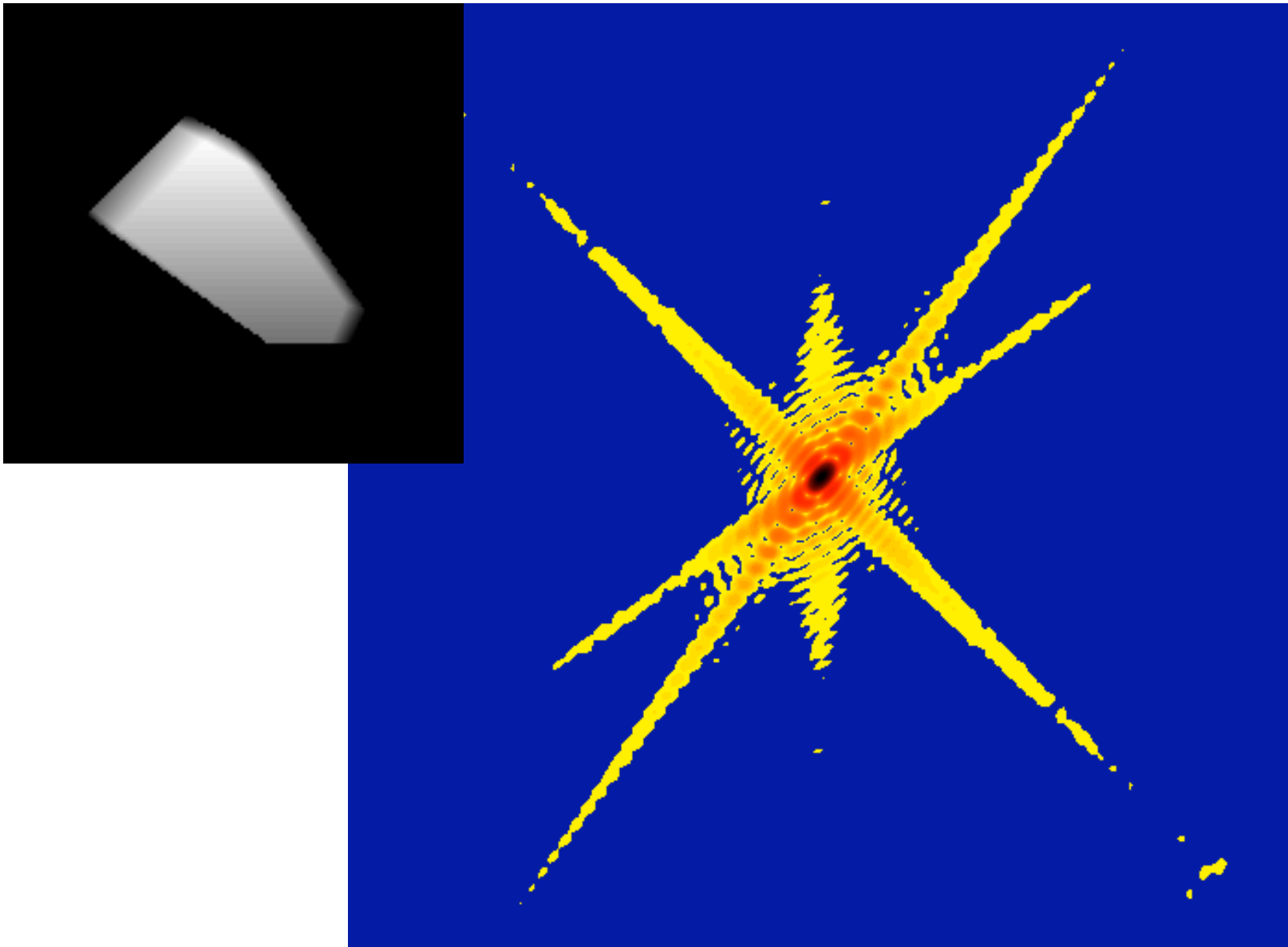
Real-space Constraints in Crystallography

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)

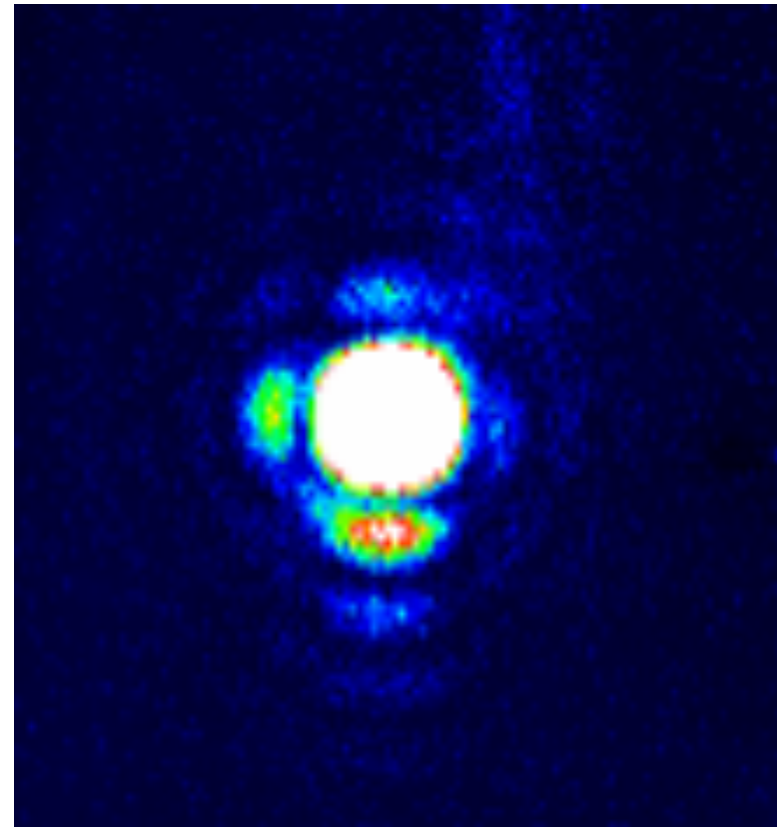
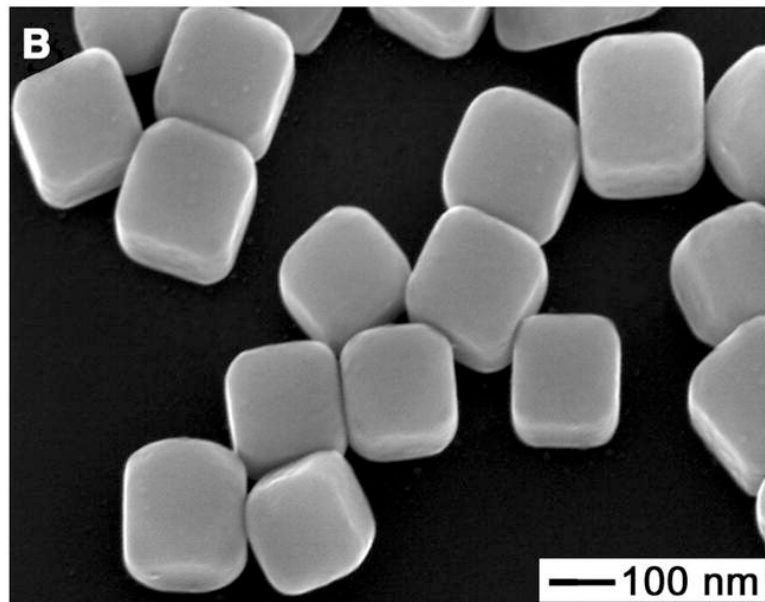
Oversampling solves Phase Problem





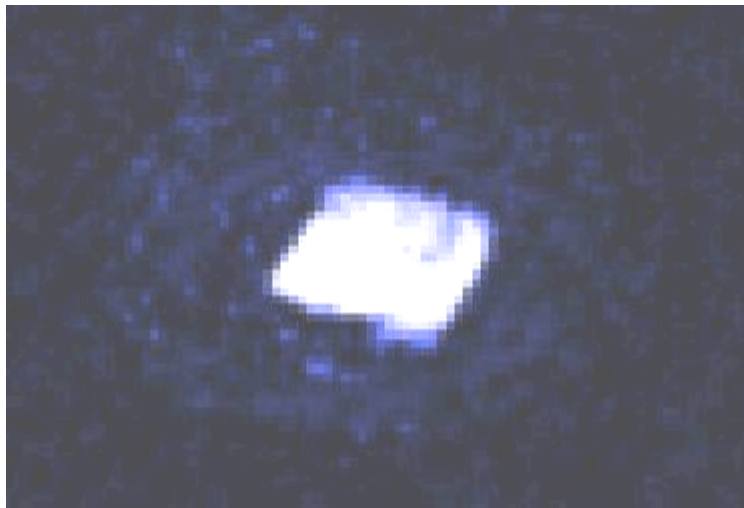
I. K. Robinson Bremen Winter School

Chemically Synthesized Silver Nanocubes

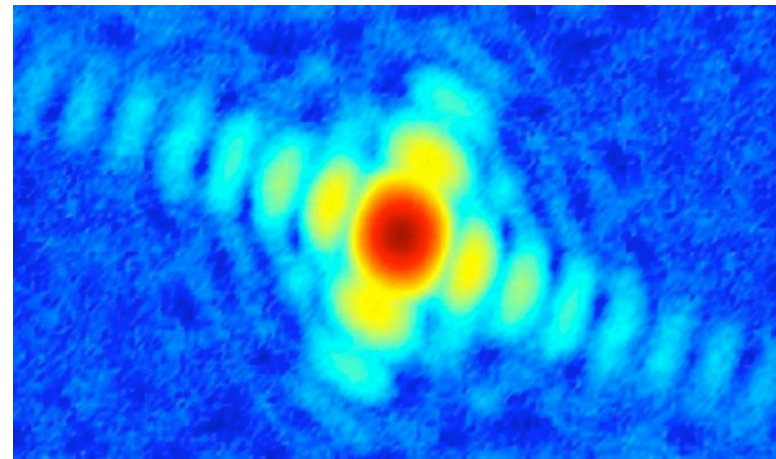
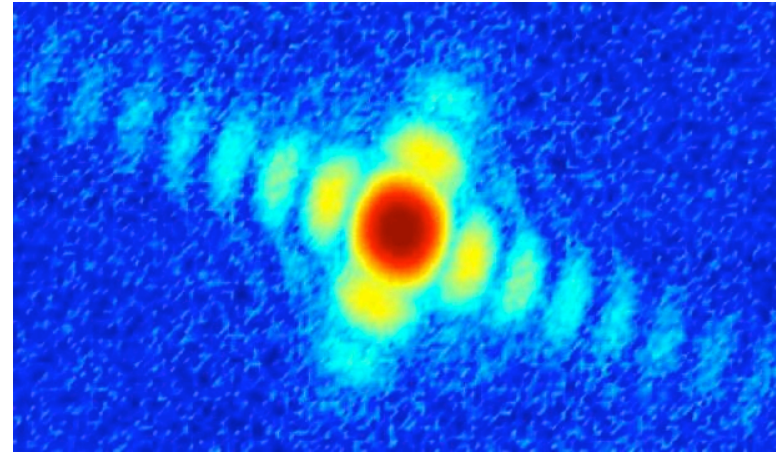


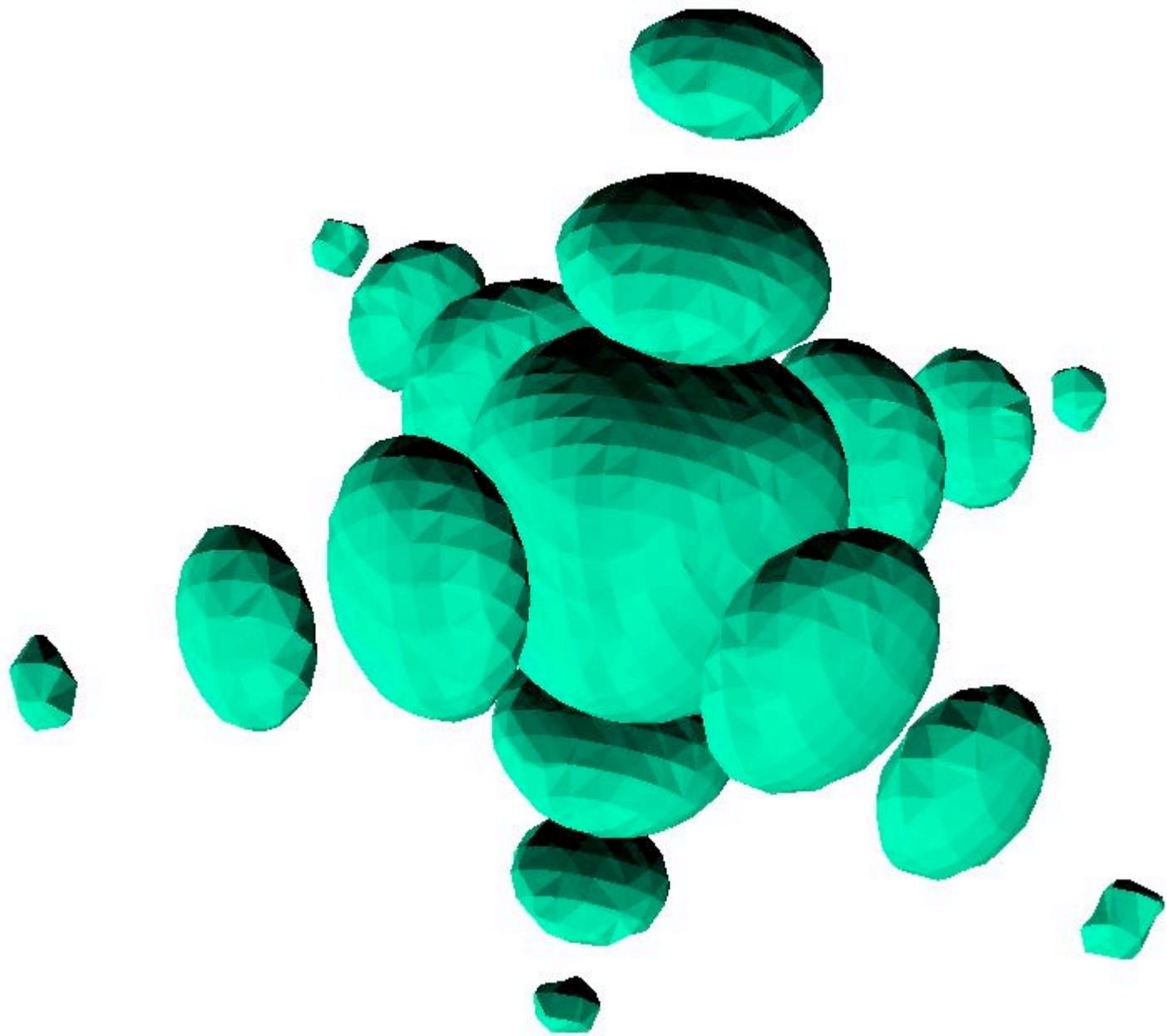
Yugang Sun and Younan Xia,
Science 298 2177 (2003)

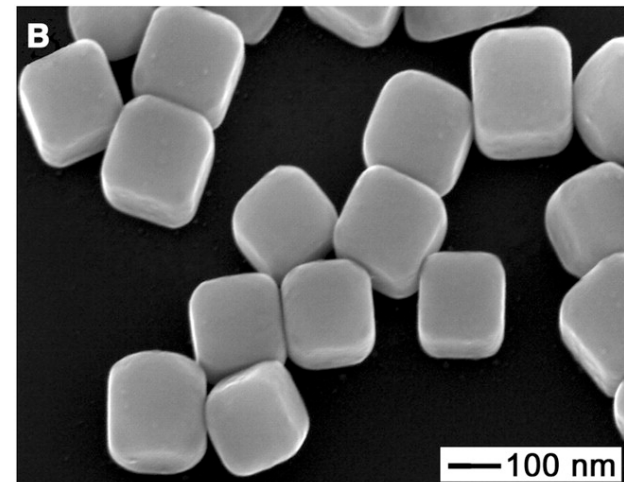
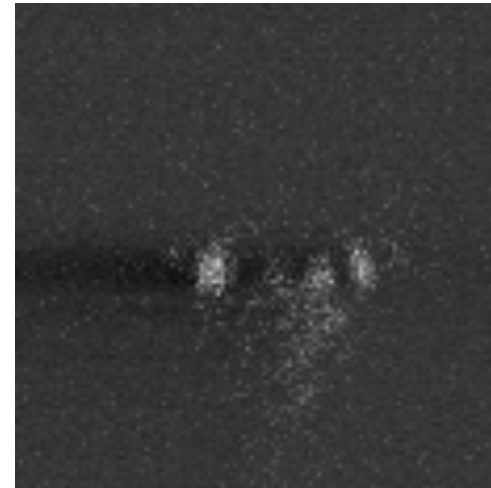
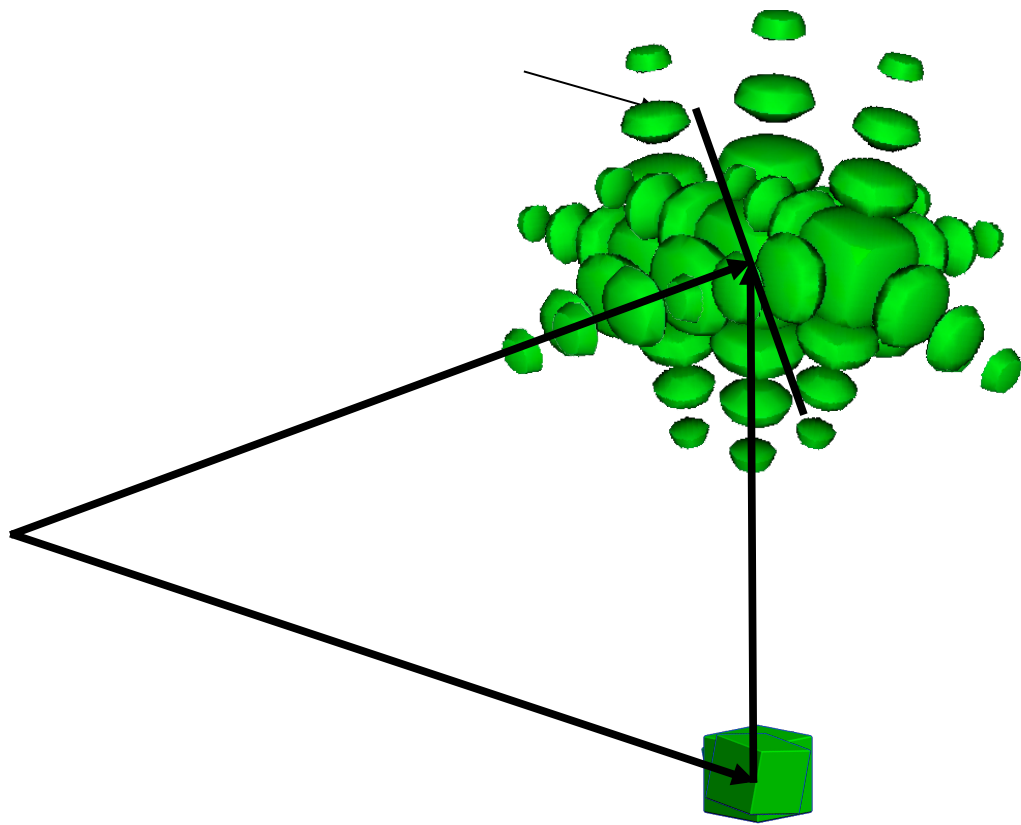
Reconstruction of Ag Nanocrystal

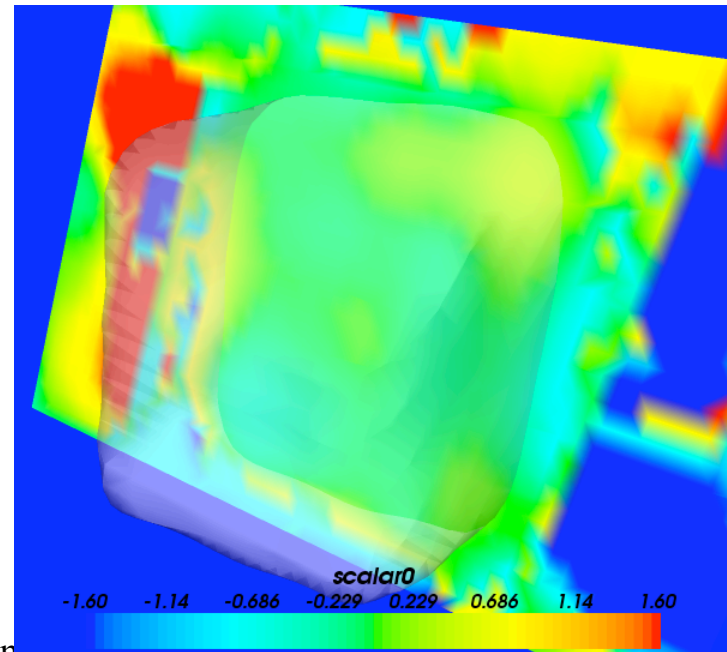
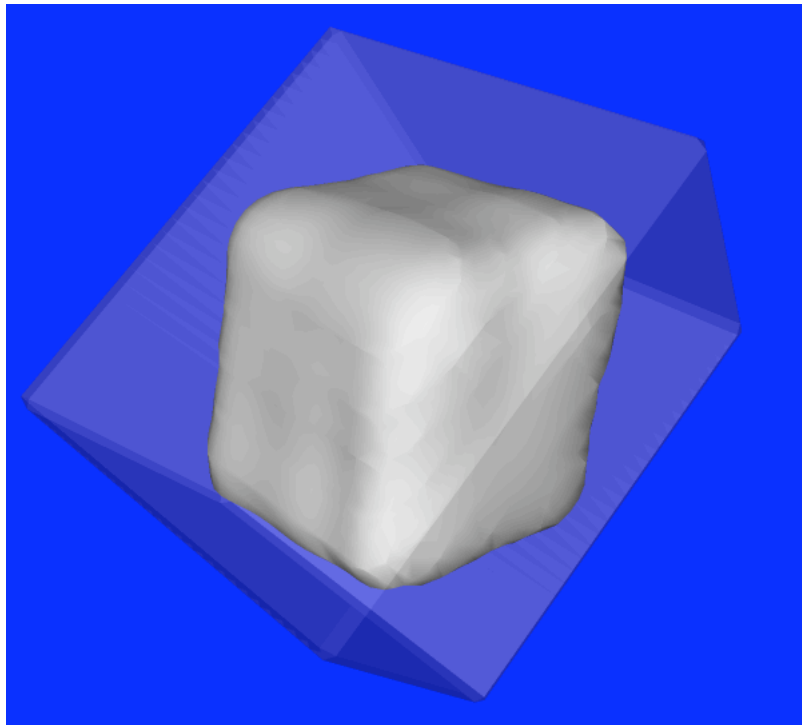
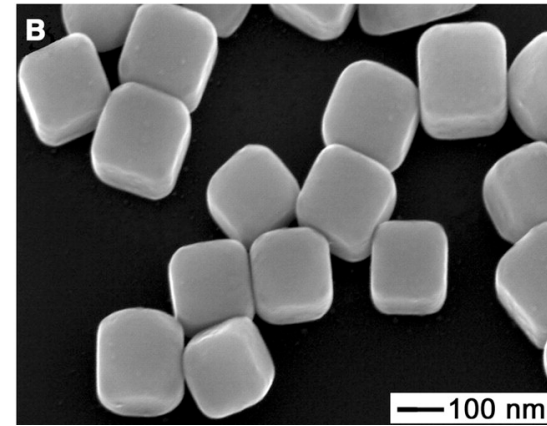
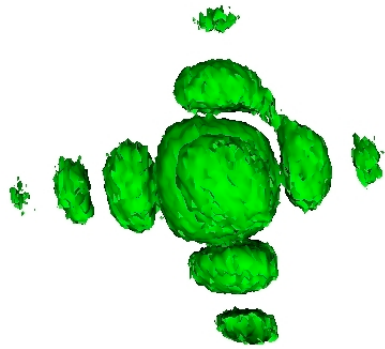
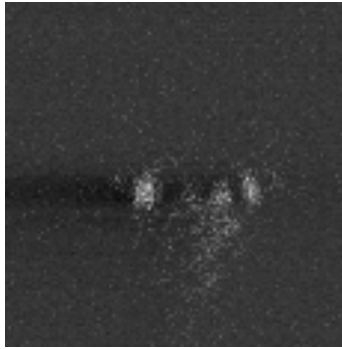


↔
200nm



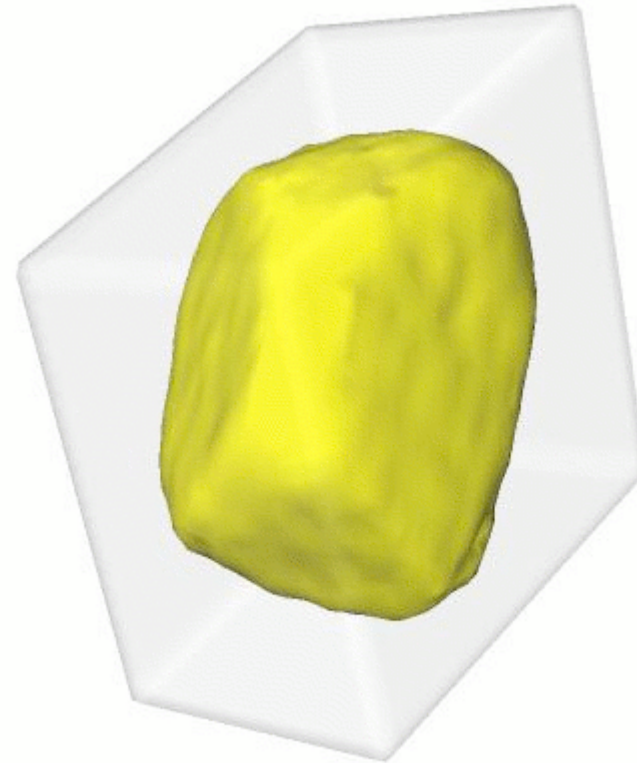
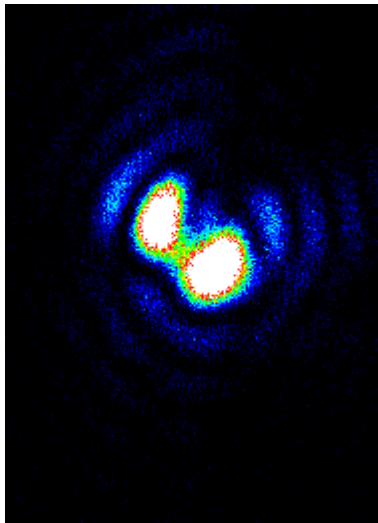






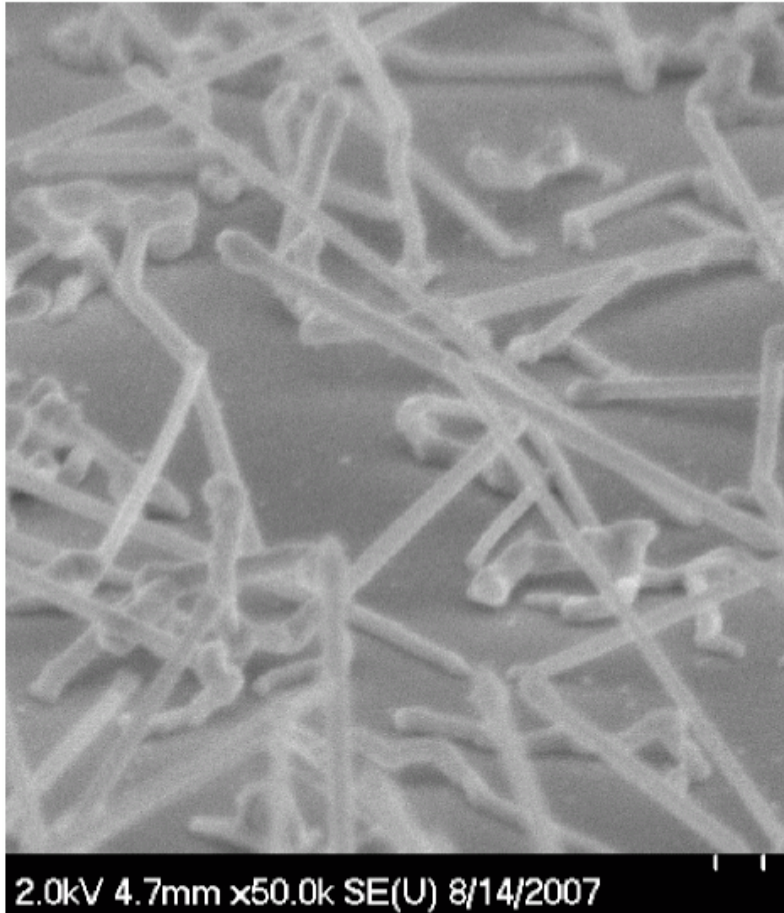
Gold nanocrystal reconstruction

showing support used for 20 HIO followed by 10 ER

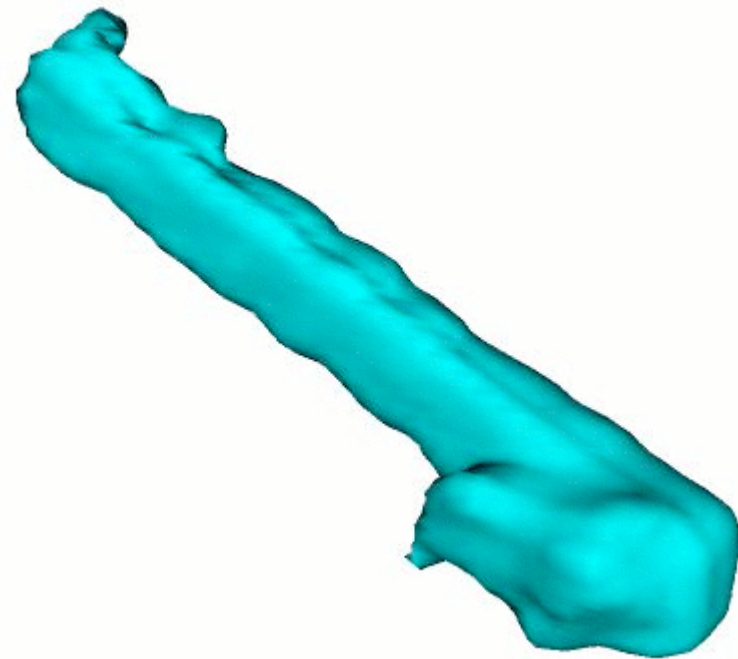


Reconstruction of InP nanowire

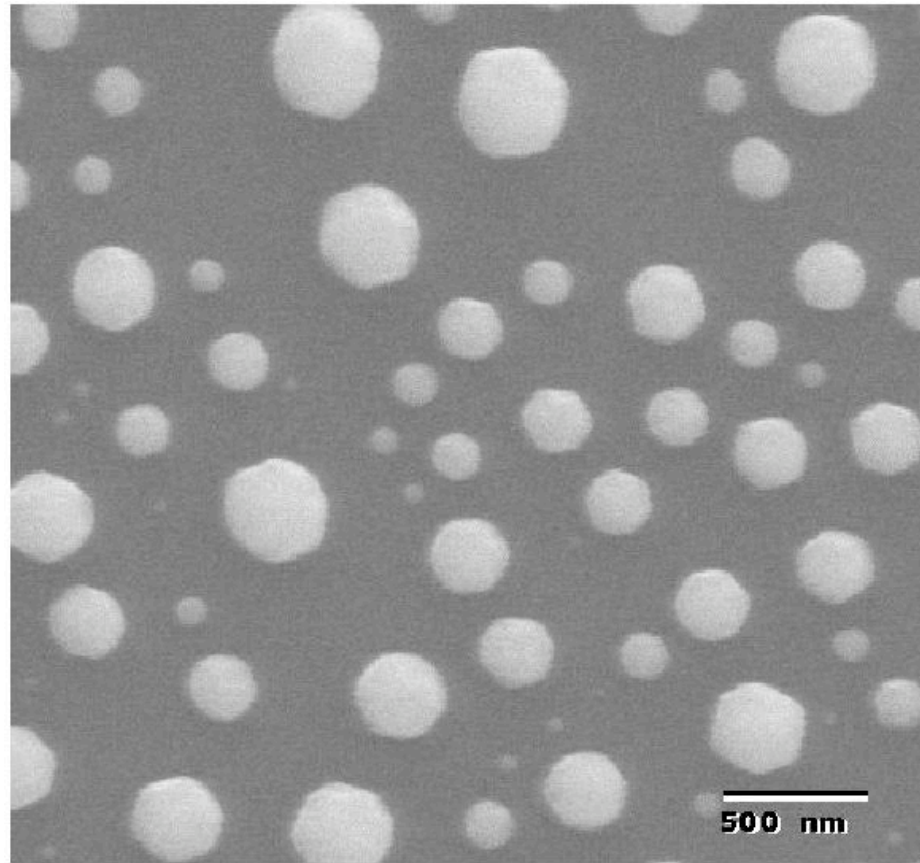
CVD on Si, Suneel Kodambaka, UCLA



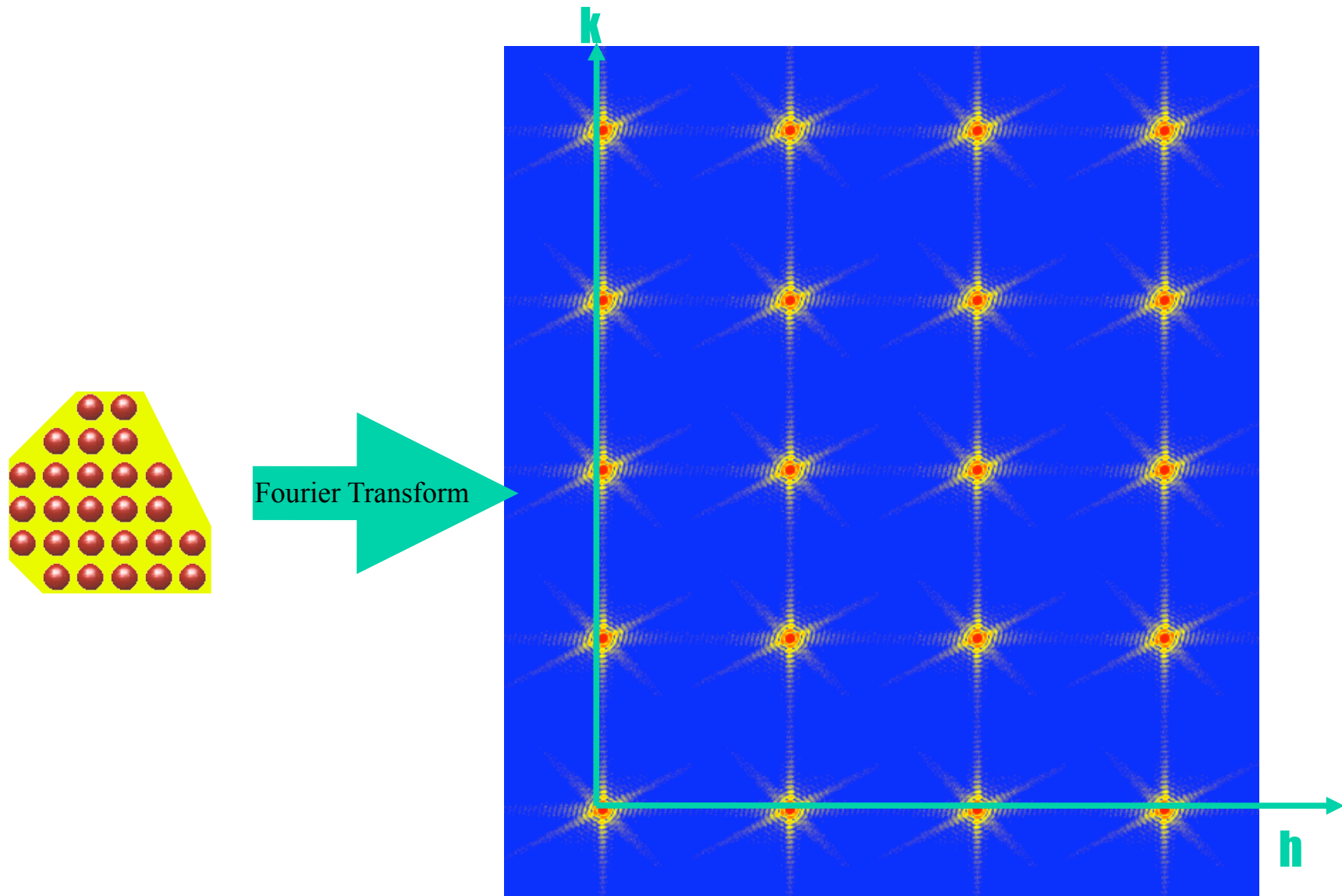
InP nanowires grown on Si (111)



In situ growth of Pb crystals



Coherent Diffraction from Crystals



Good statistics, 3D diffraction data

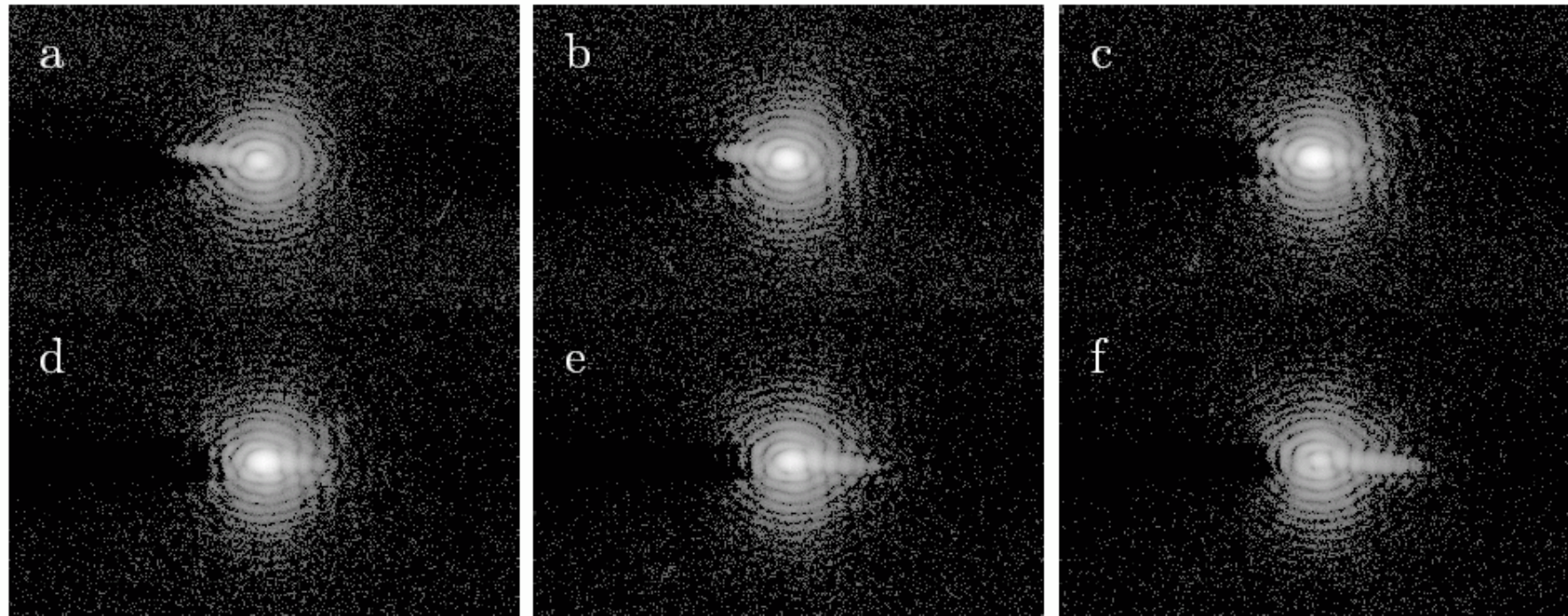
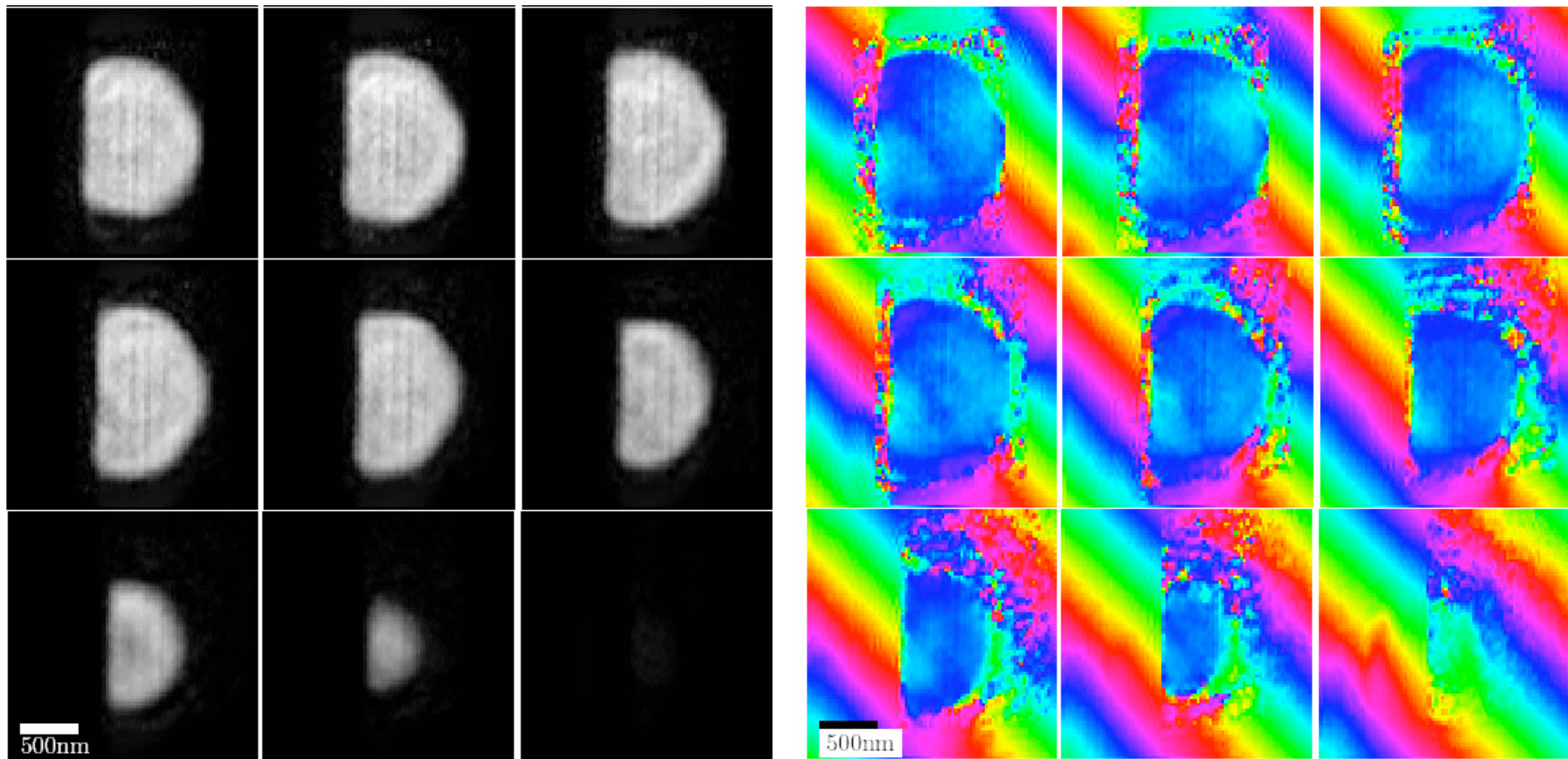
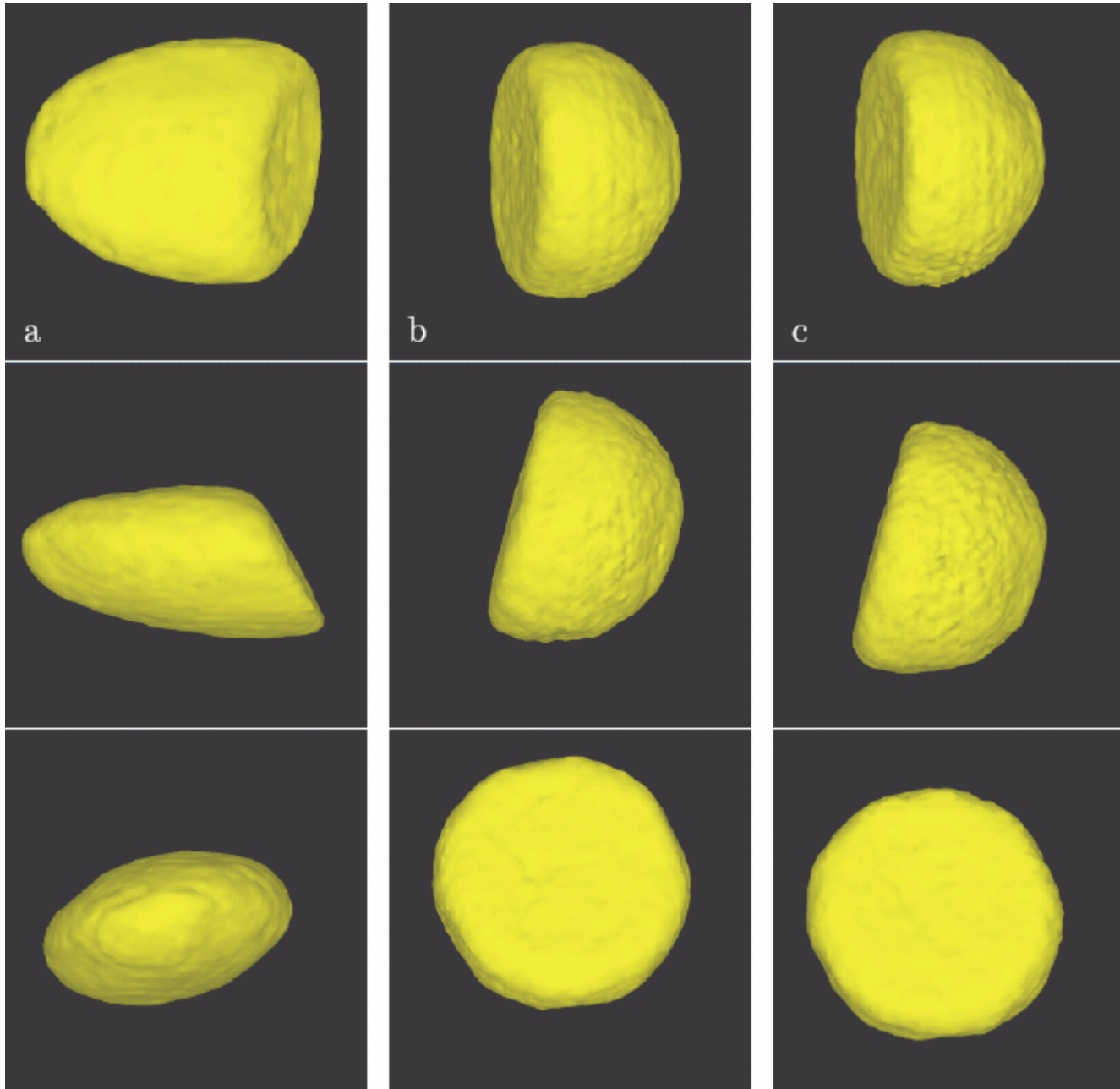


Figure 4.12: Center slices from 3D CXD pattern from Pb sample, on a log scale. Data file 296 from 10/03.

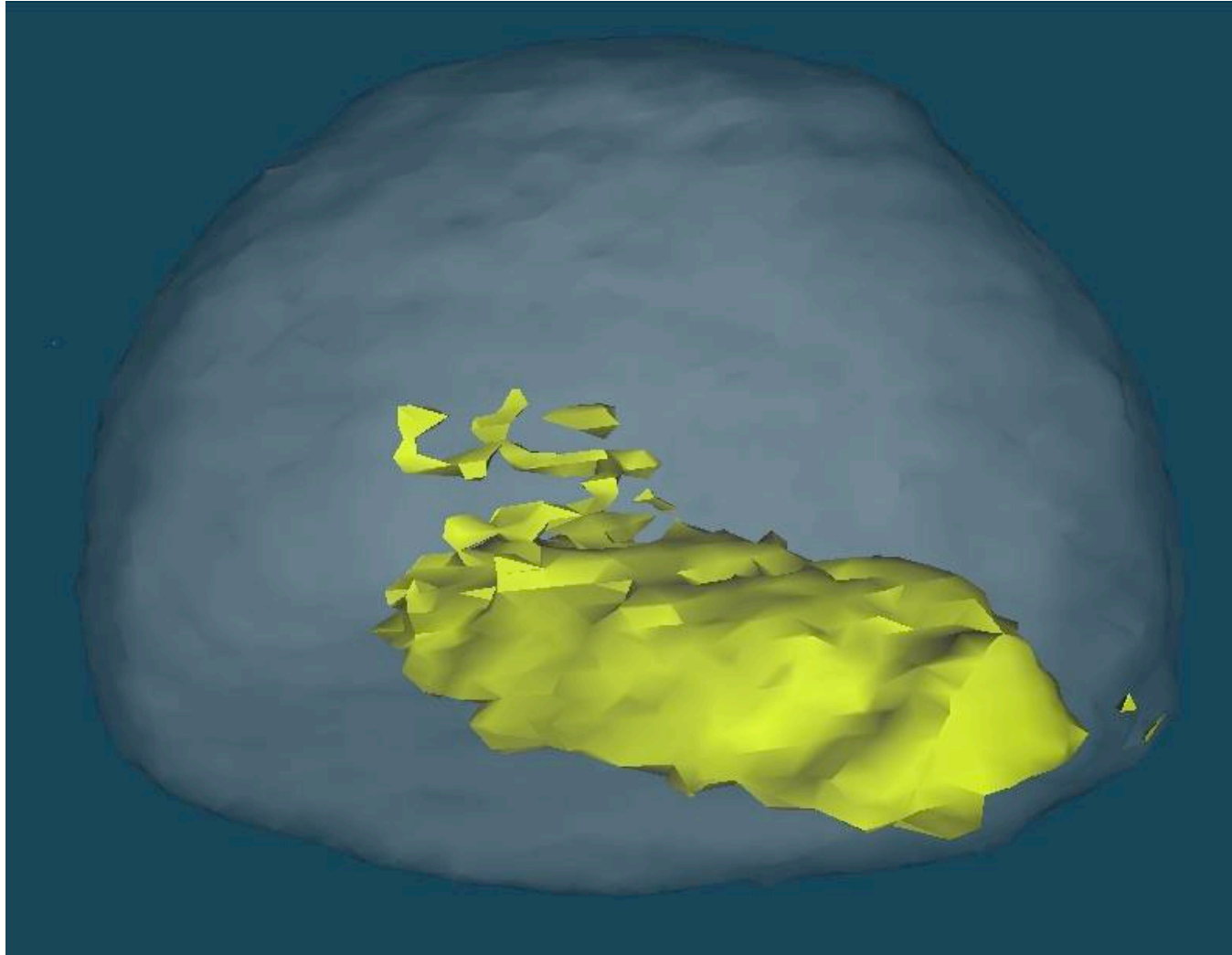
Complex Density (amplitude *and* phase)





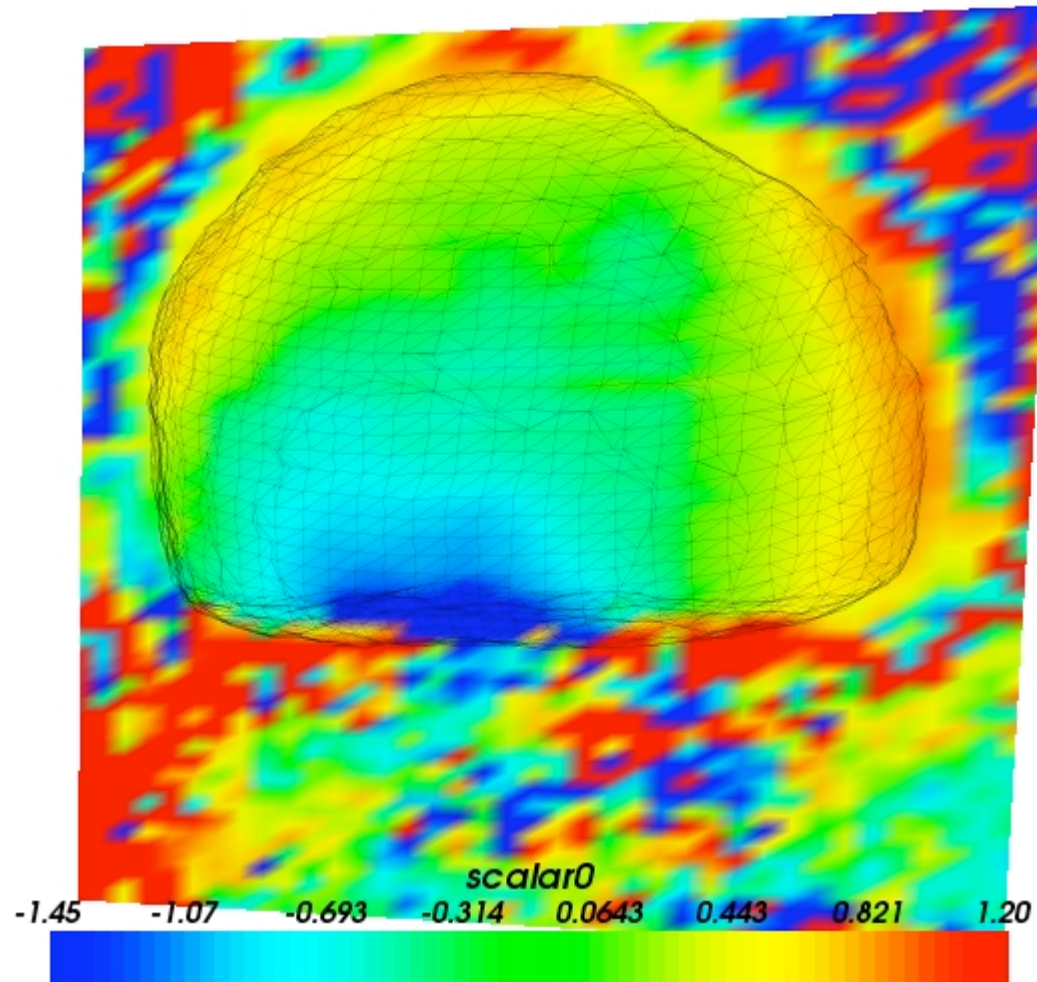
I. K. Robinson Bremen Winter School

Modeling of 3D Phase Bump



I. K. Robinson Bremen Winter School

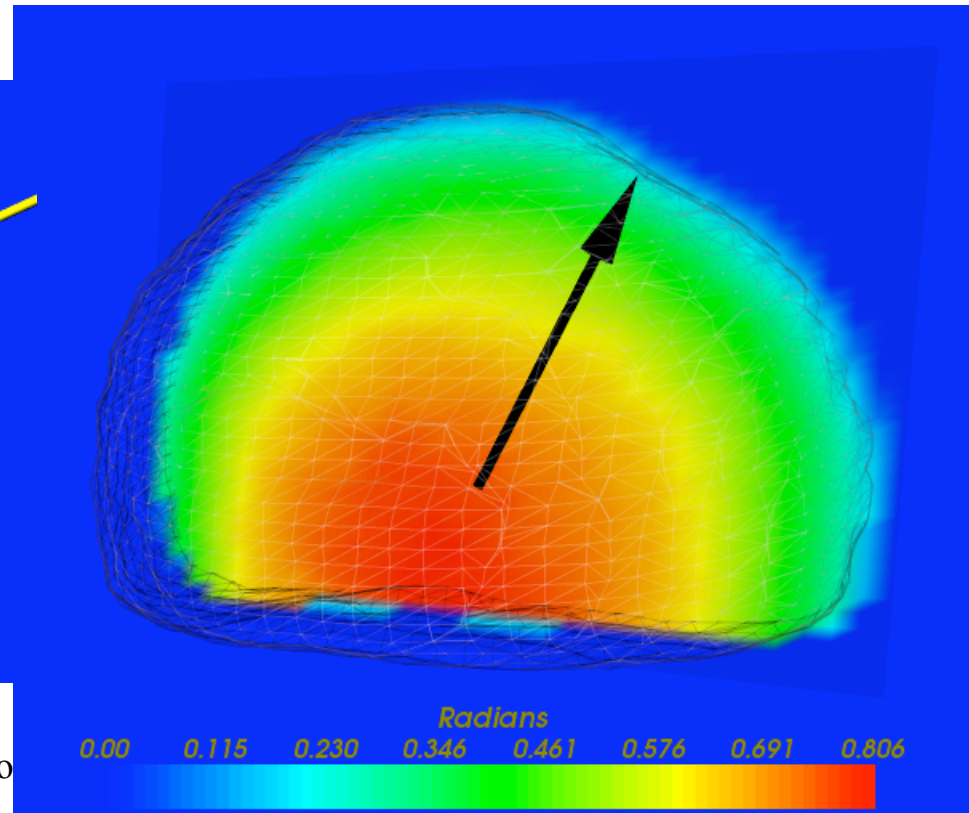
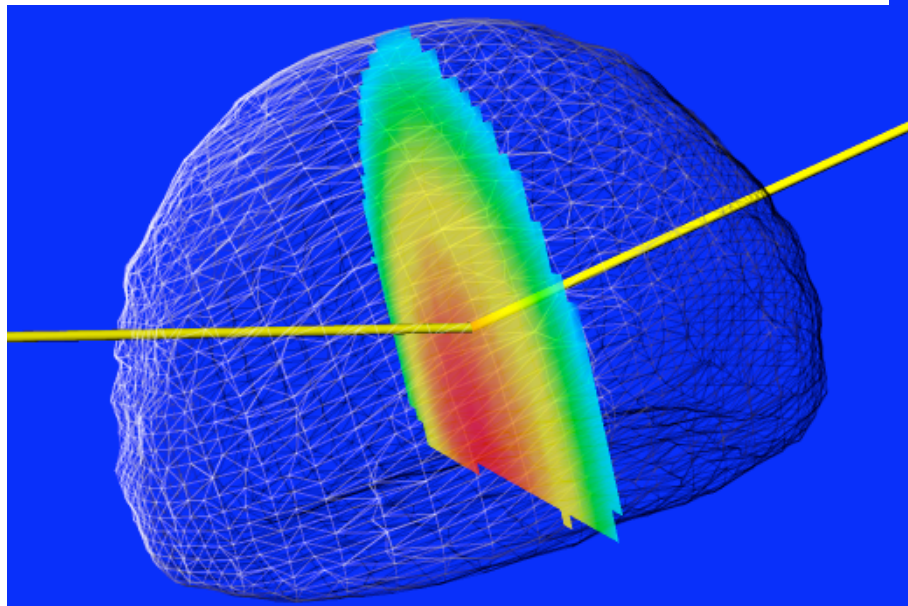
Contours showing Positive Phase



Refraction effects in Lead at 8.9keV

Phase accumulation due to refraction along scattering path
 $n=1-\delta+i\beta$

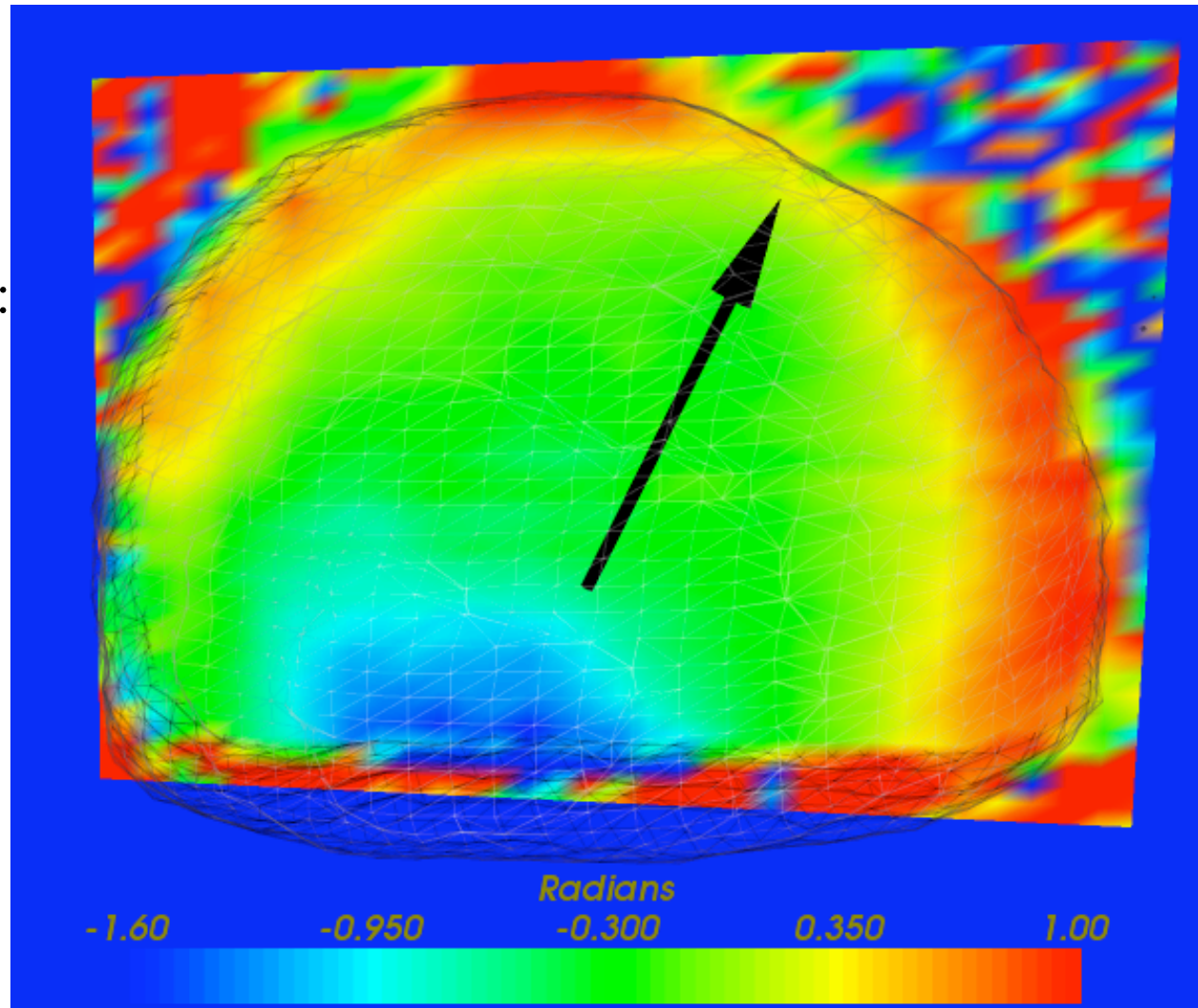
$$\delta=2.23\times 10^{-5}$$
$$\beta=2.19\times 10^{-6}$$



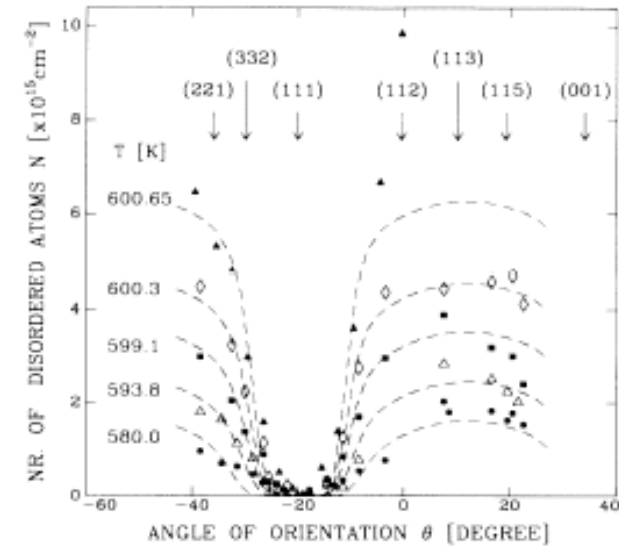
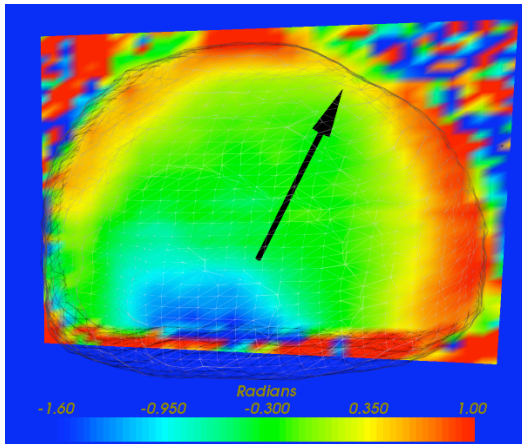
Refraction corrected phase map

Max phase = 1.15rad
= 0.052nm

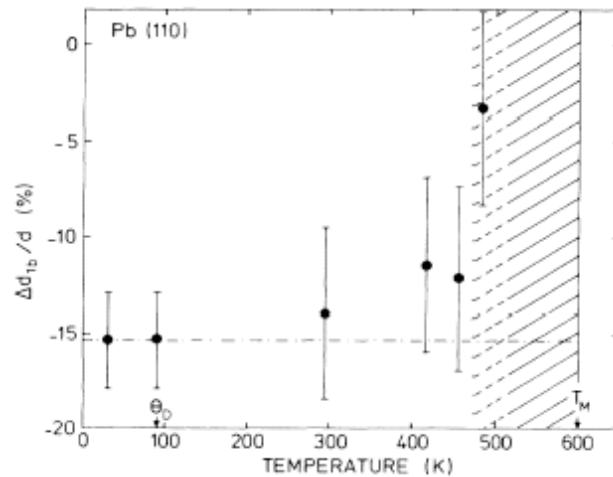
Phase on the (111) facet:
= 0.47 rad
= 0.02nm



“Anomalous” Thermal Expansion (of Surfaces)

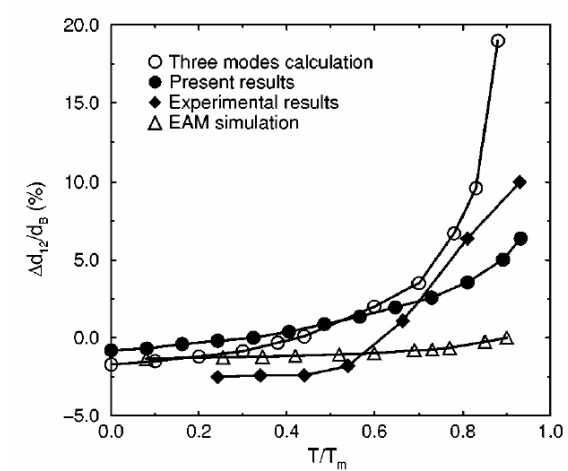


Pluis, van der Gon, Frenken, van der Veen
PRL 59 401(1987)



Frenken, Huusen, van der Veen
PRL 58 (1987)

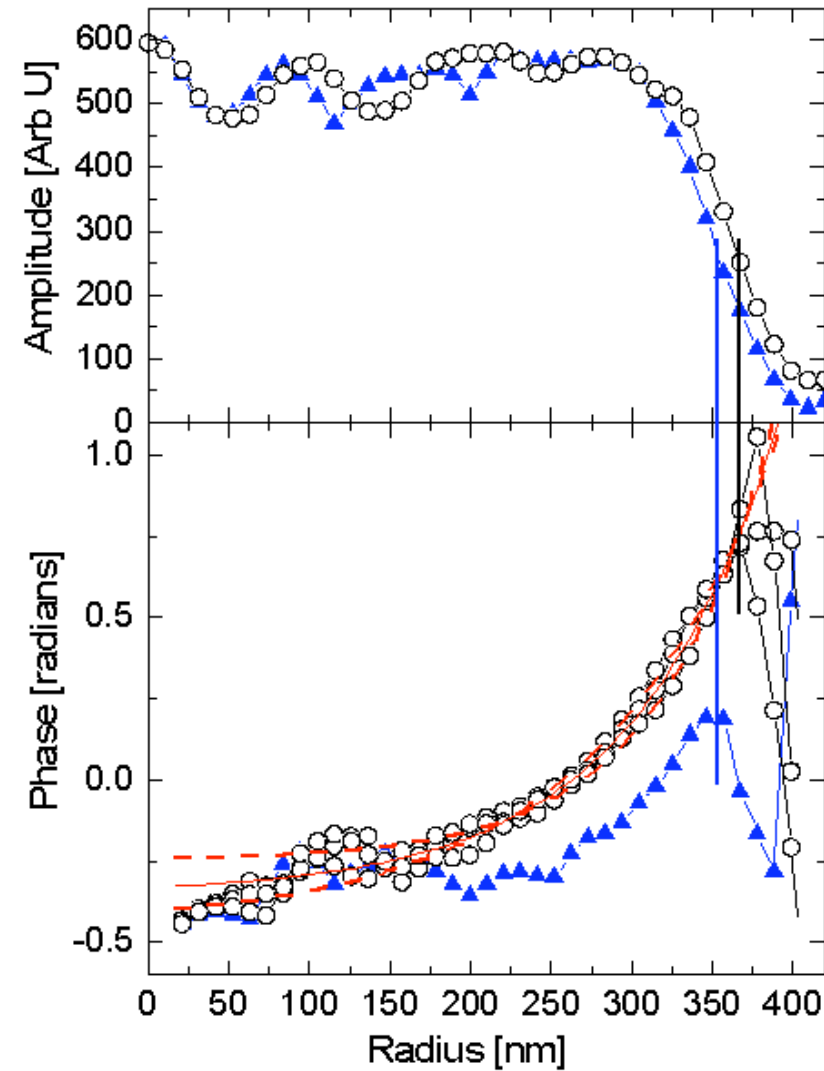
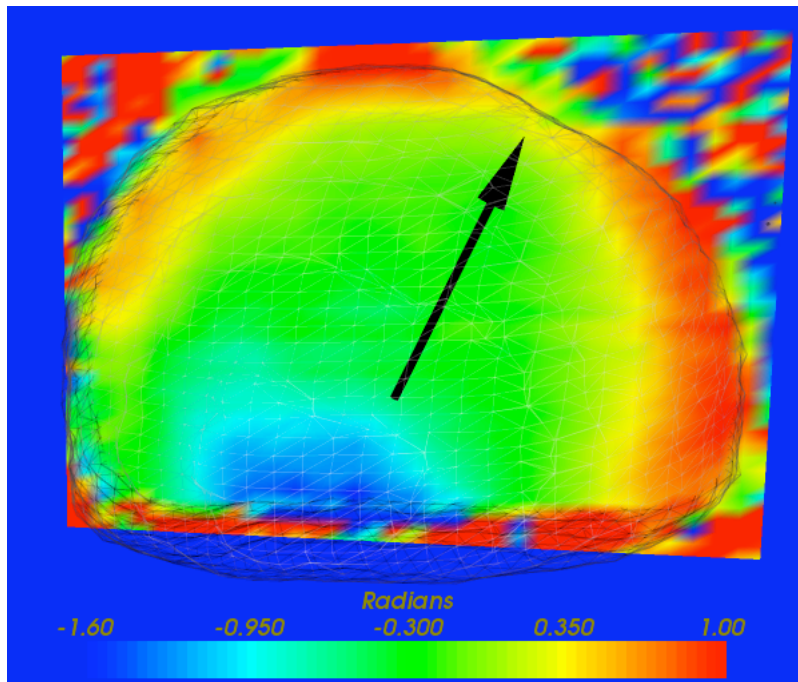
I. K. Robinson Bremen Win



Ag(111) theory J. Xie (Scheffler group)
PRB 59 970 (1999)

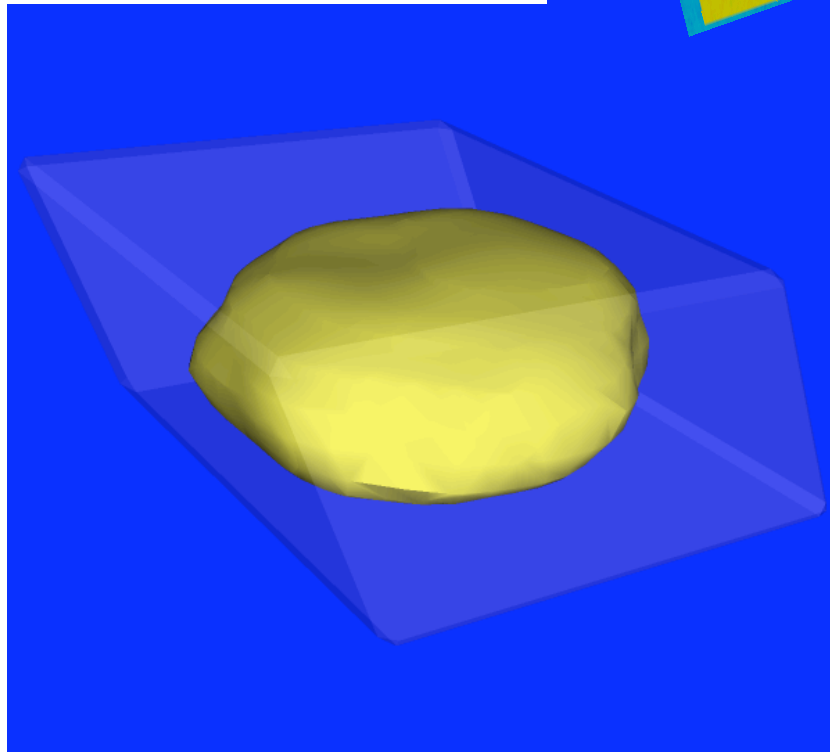
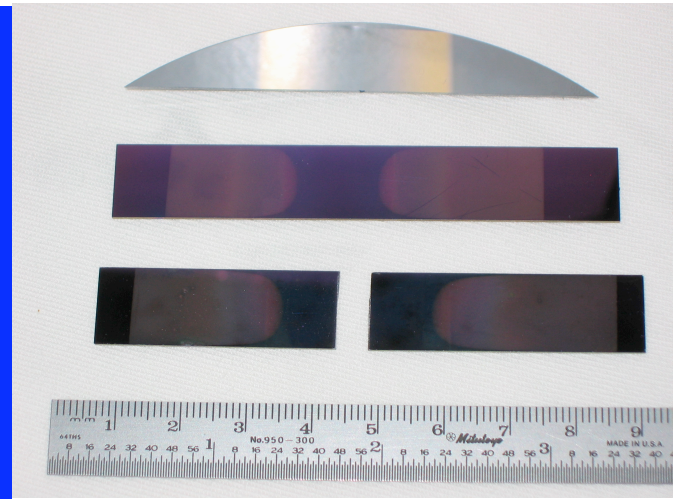
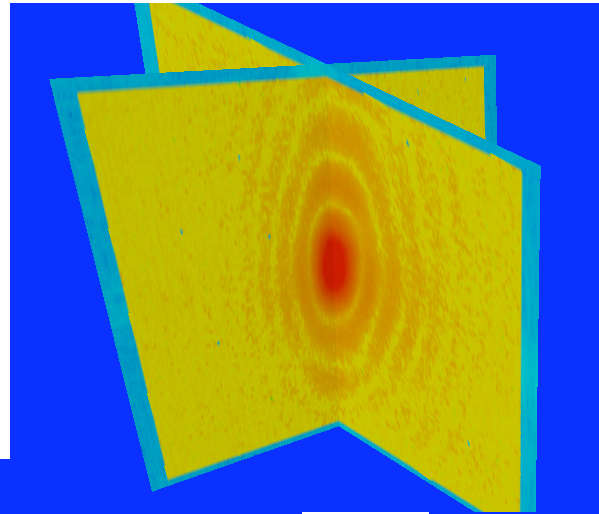
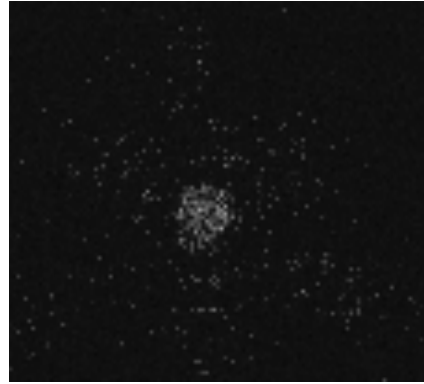
Facets subtend $18.6\text{deg} \pm 2.6$
ECS predicts 14deg

Depth measured $90\text{nm} \pm 20$
Expect about 75nm

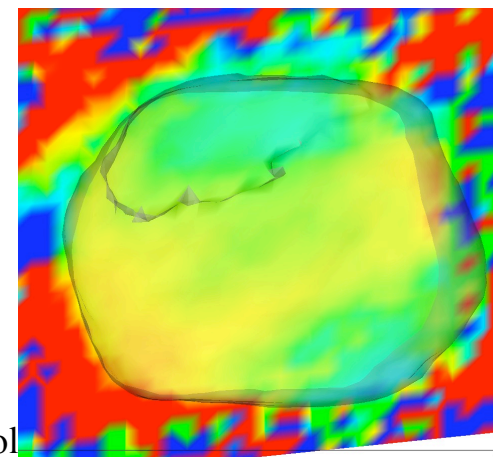
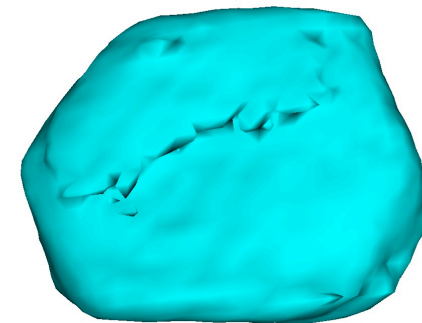
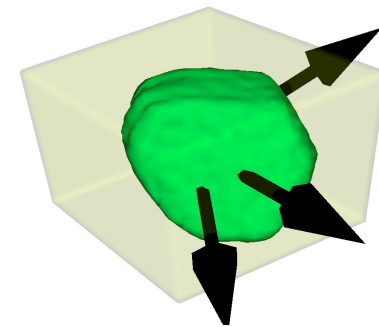
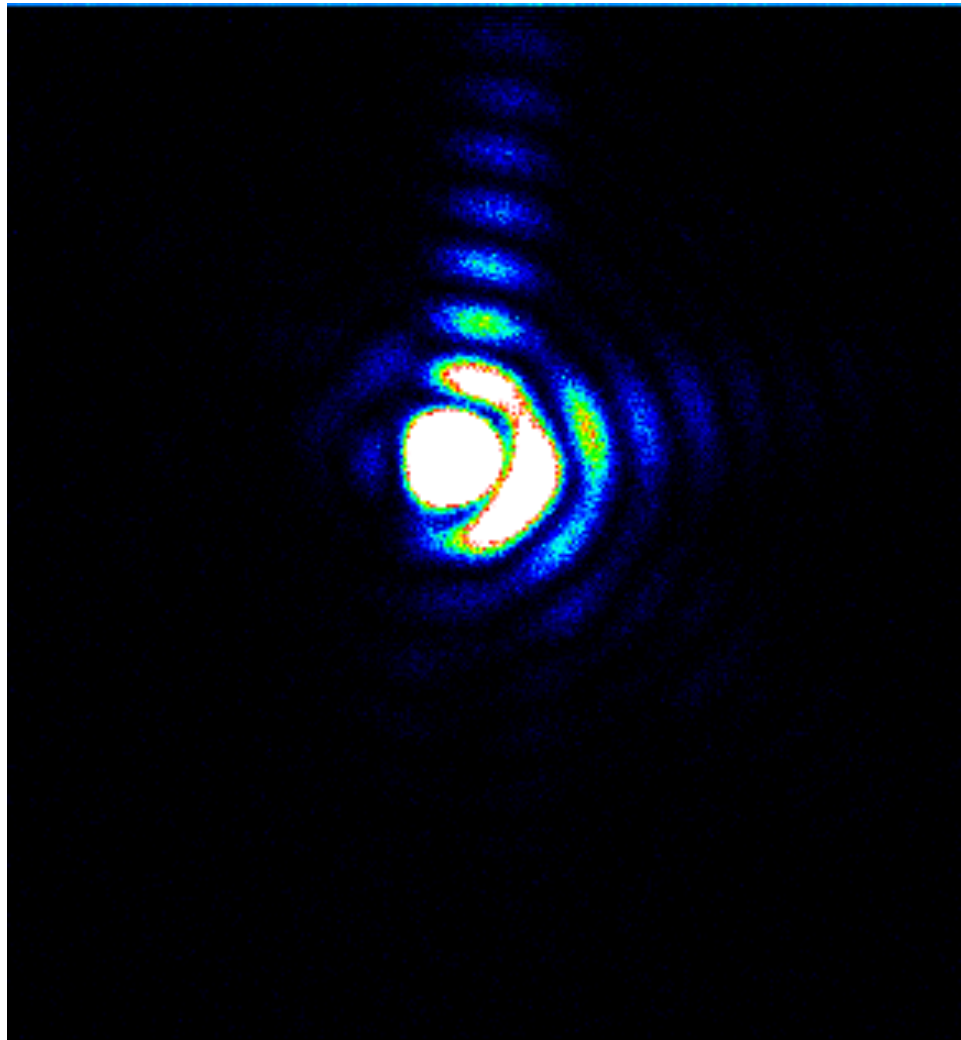


3D Au crystal

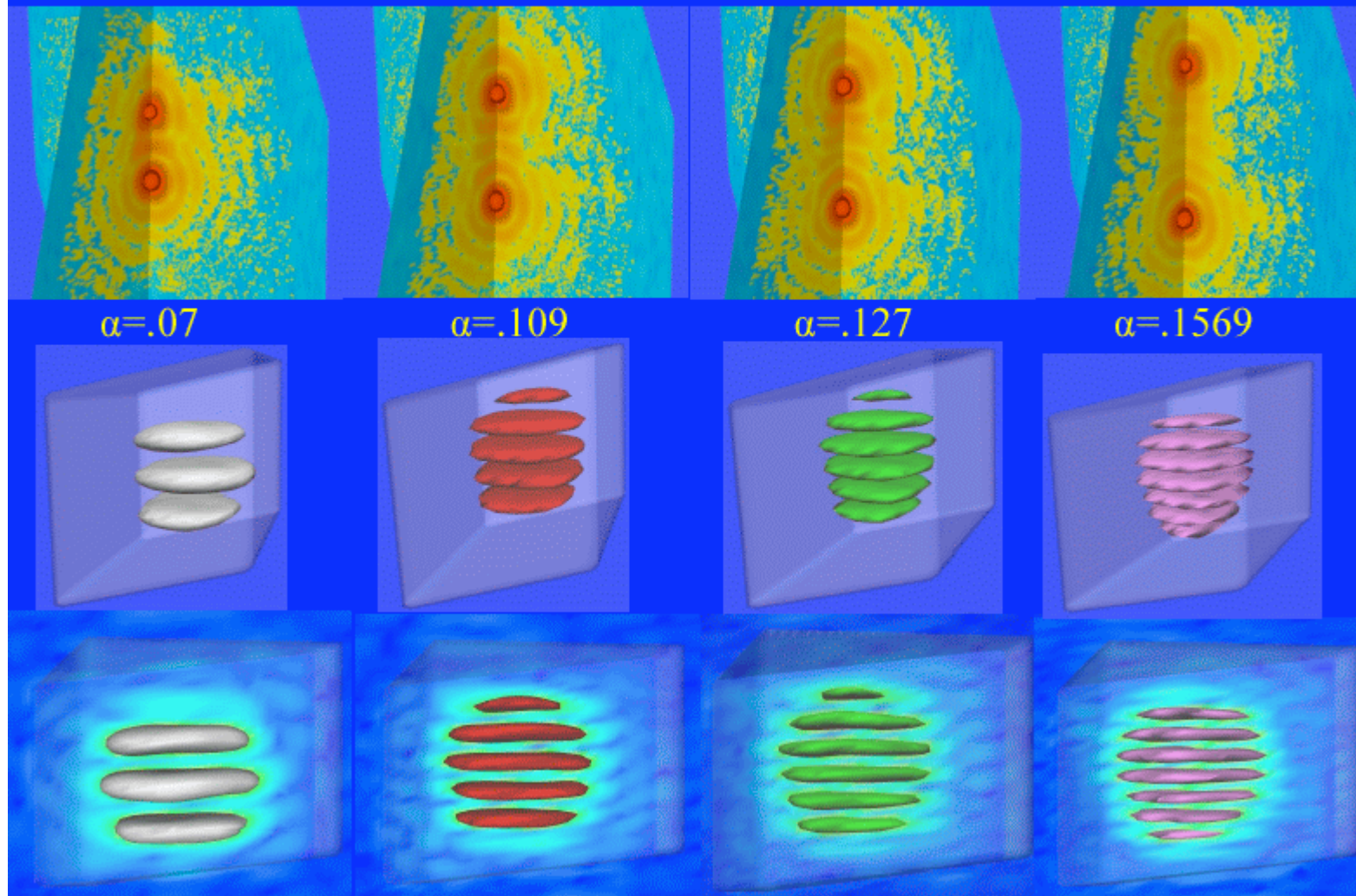
XFD-OFM Chian Liu, Ali Khounsary



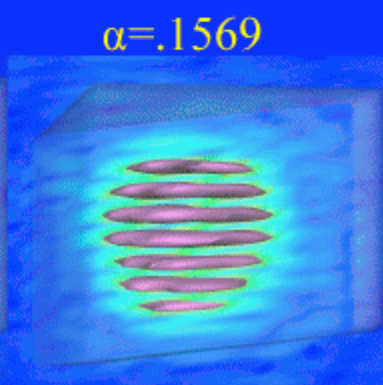
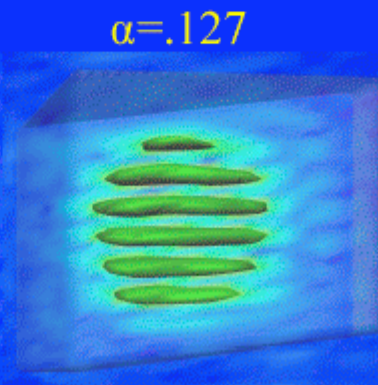
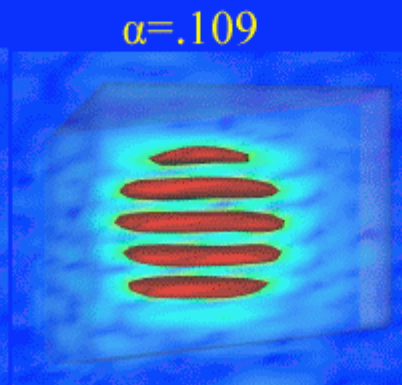
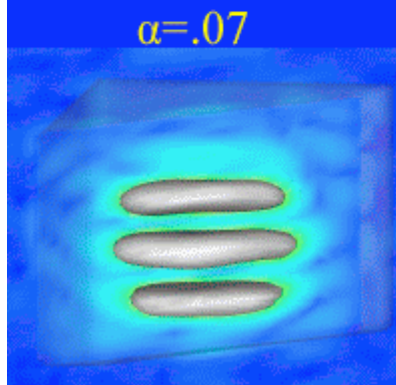
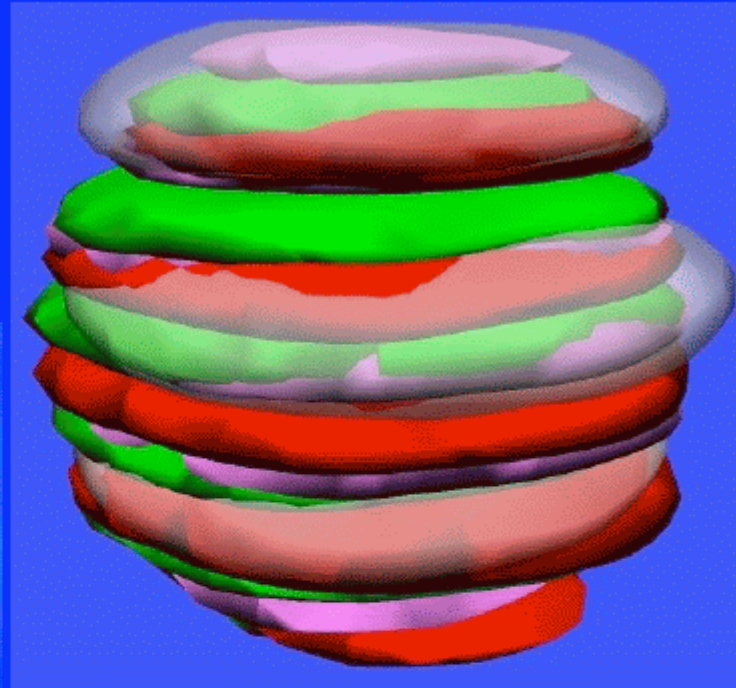
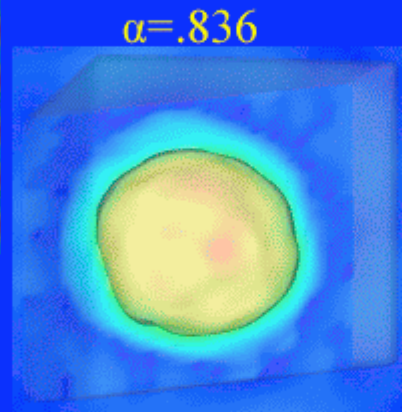
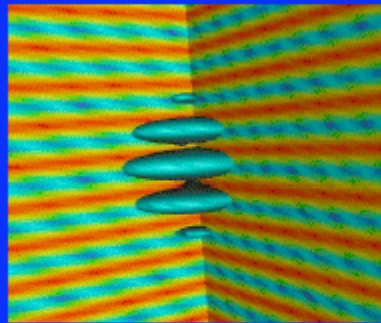
son Br



3D Standing Wave Diffraction from Au



Standing Wave Ptychography?



Important Contributions from:

Ross Harder	APS
Steven Leake	UCL
Piotr Gryko	UCL
Loren Beitra	UCL
Moyu Watari	UCL
Meng Liang	Illinois
Garth Williams	Melbourne
Mark Pfeifer	LaTrobe
Franz Pfeiffer	SLS
Ivan Vartanants	DESY

Conclusions

- Nanocrystal: paradigm of Surface/Bulk
- Chemically synthesized Ag cubes
- Dewetting of Pb and Au hemispheres
- Surface strains visible
- Coherence of SR essential
- No longer average over “typical” structure