Coherent X-ray Diffraction analysis of Semiconductor Quantum Dot structures

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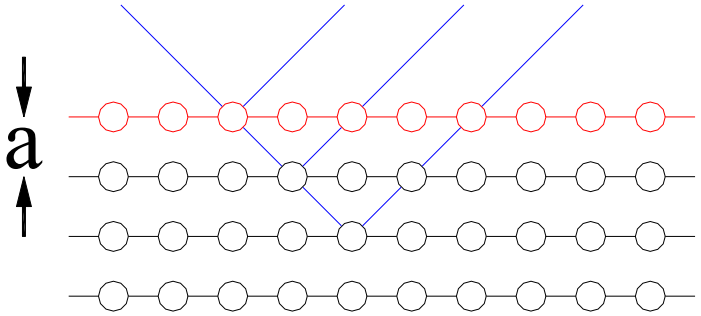
Outline

- Direct analysis of CTRs
- Coherence in Diffraction
- Solving the Phase Problem
- Nanocrystal Shapes
- Quantum Dot structures
- How small can we go?

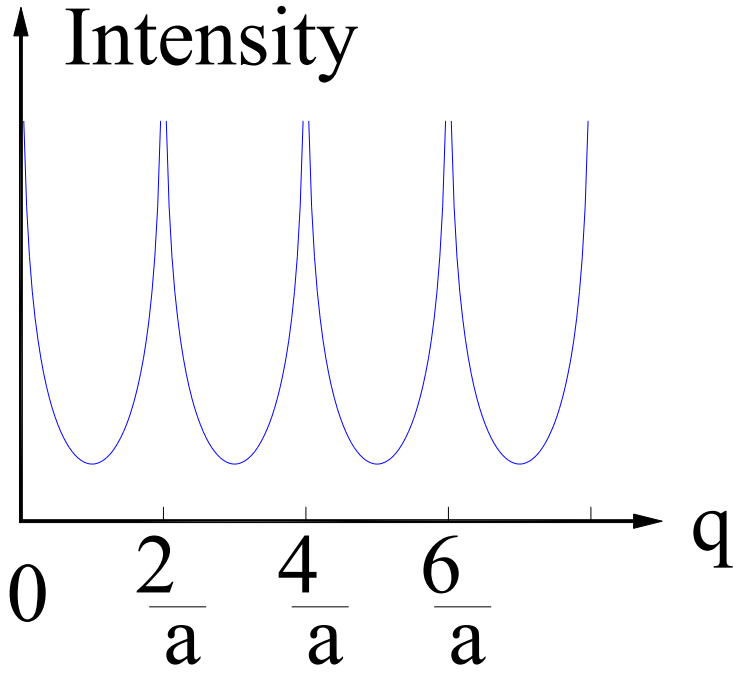
Origin of Truncation Rods



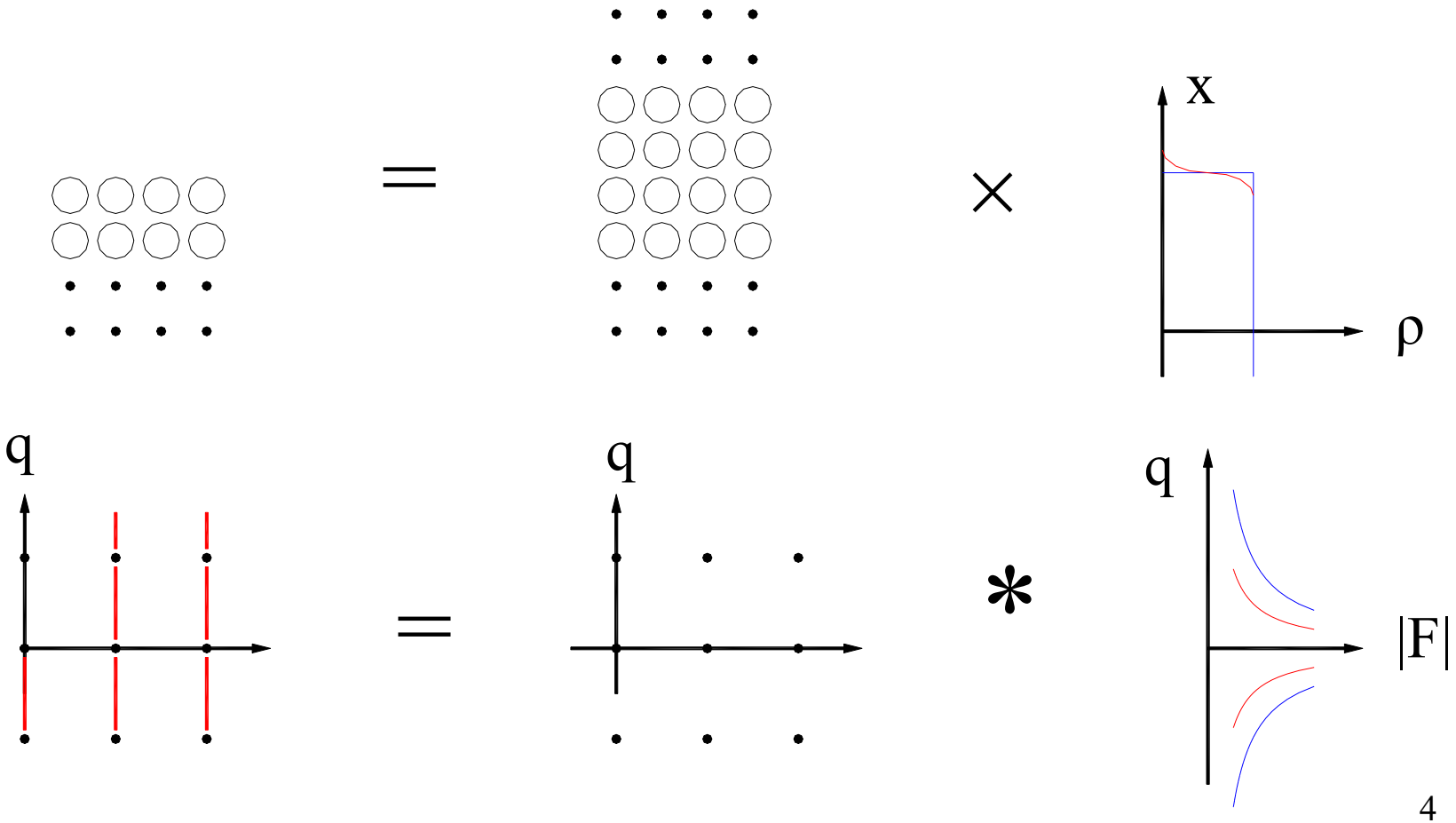
$$F_{CTR} = \sum_{n=0}^{\infty} A_n$$

$$= \sum_{n=0}^{\infty} f_L \; e^{inqa}$$

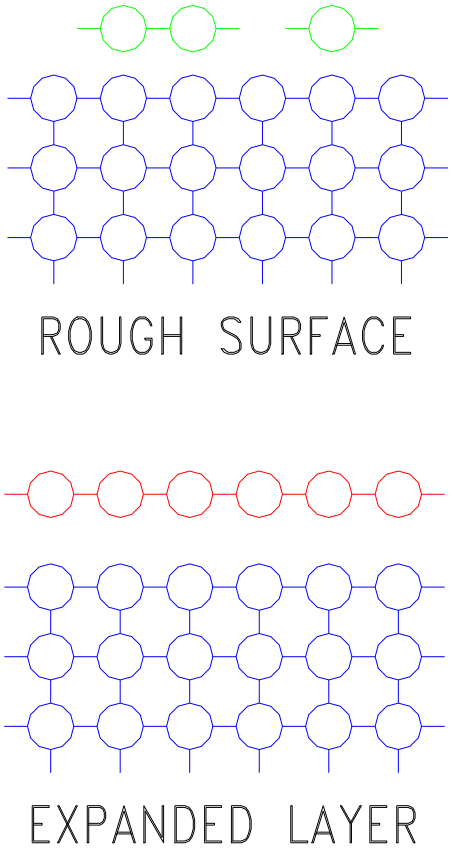
$$= \frac{f_L}{1 - e^{iqa}}$$

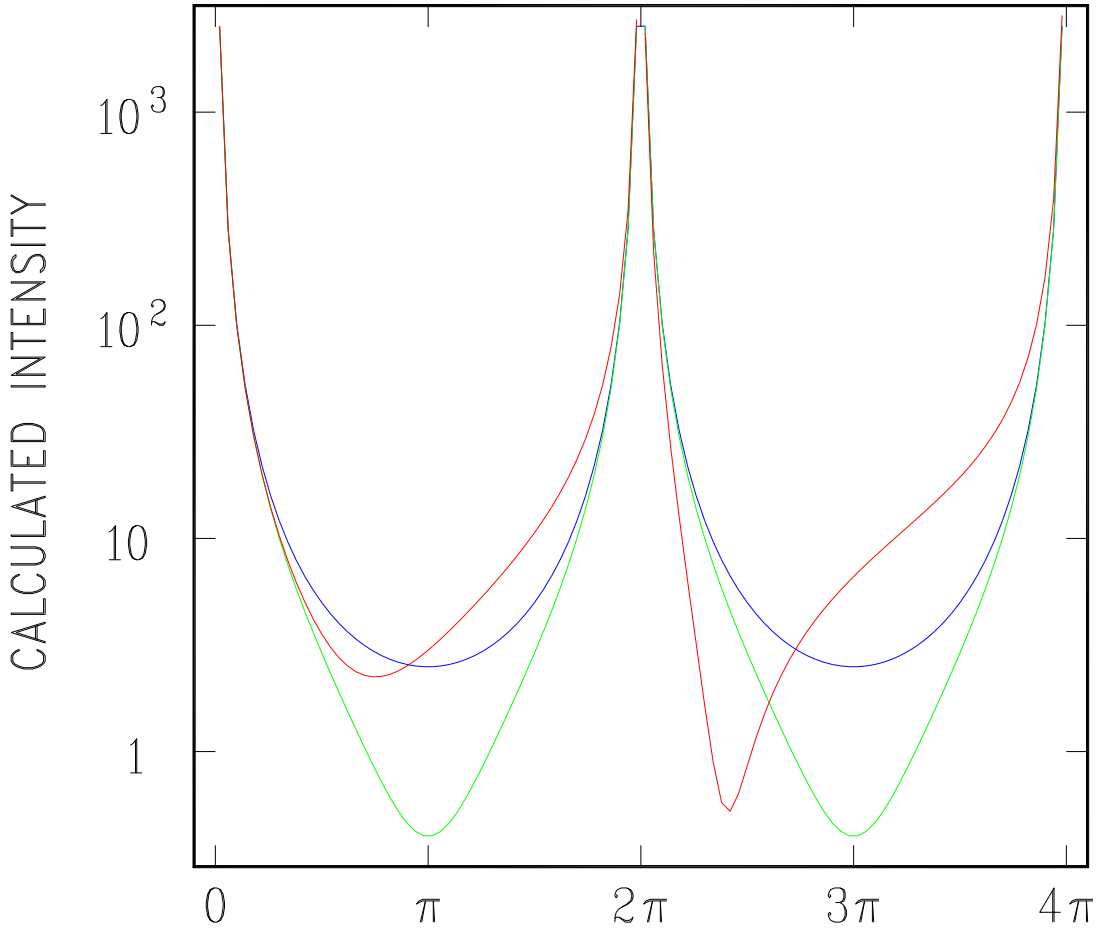


CTR as Convolution

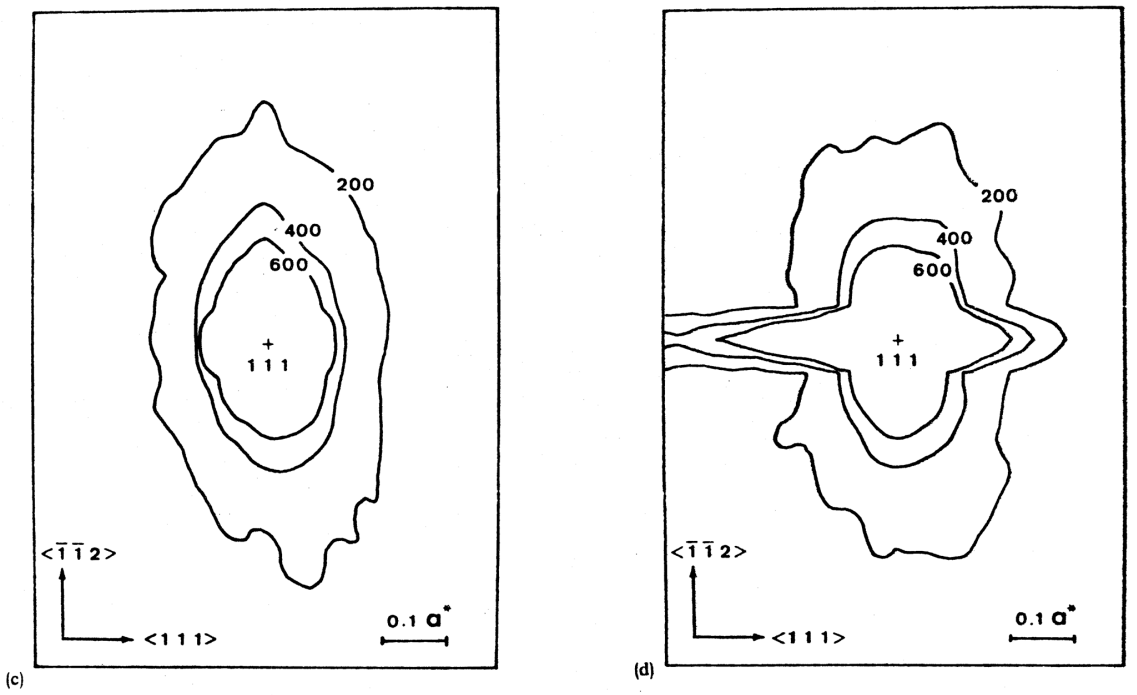


CTR is Sensitive to Surface Structure





Diffuse Scattering from Si Wafer



Unpolished wafer 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)

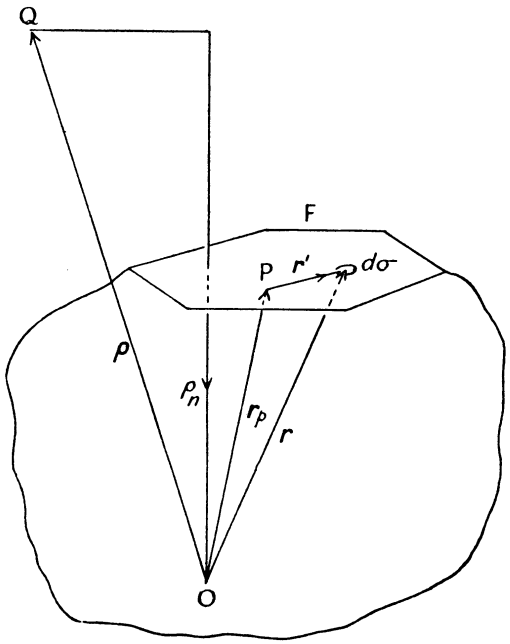
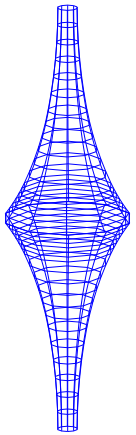
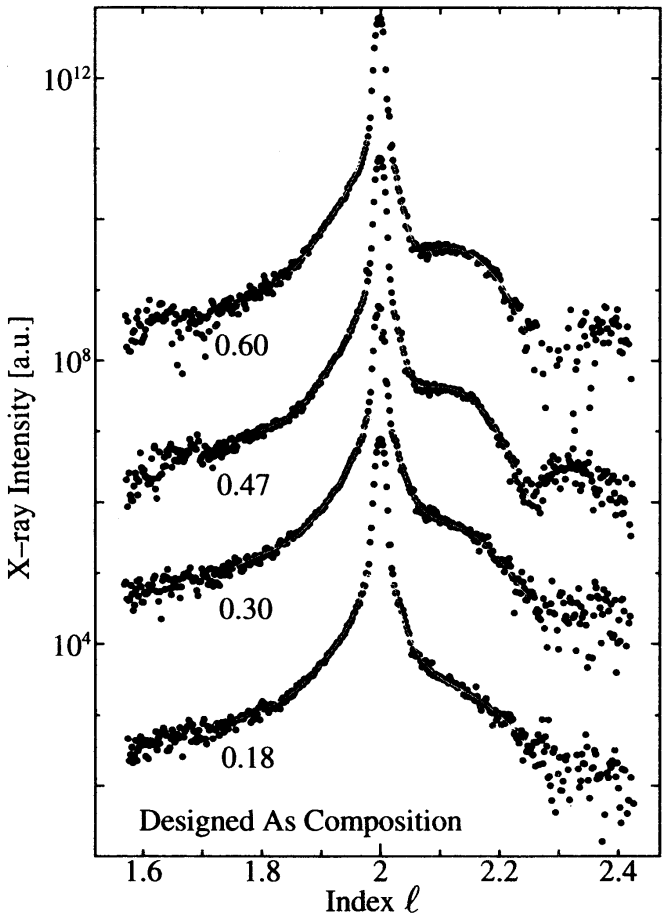


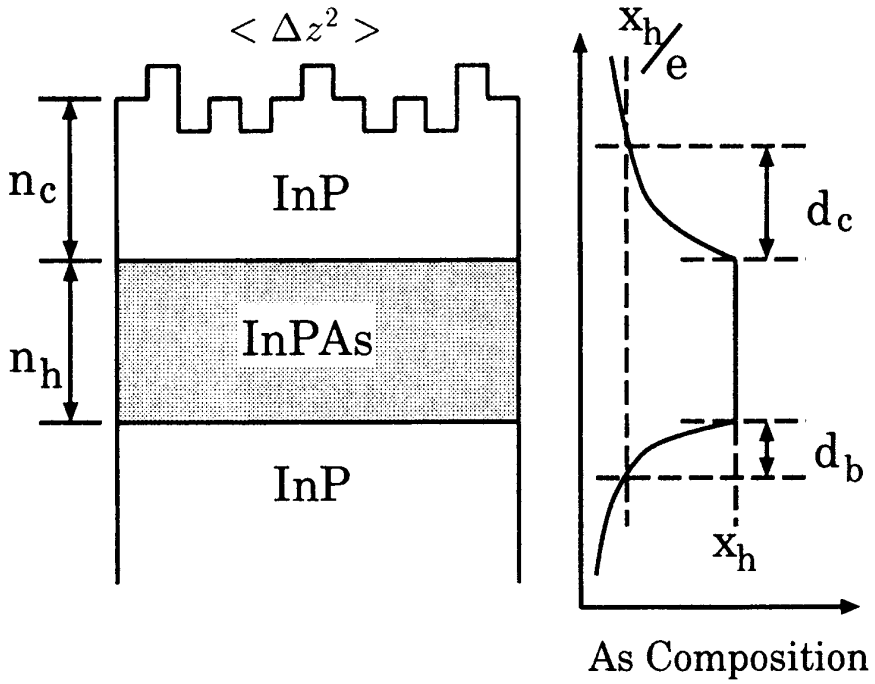
FIG. 200



"Stacheln"

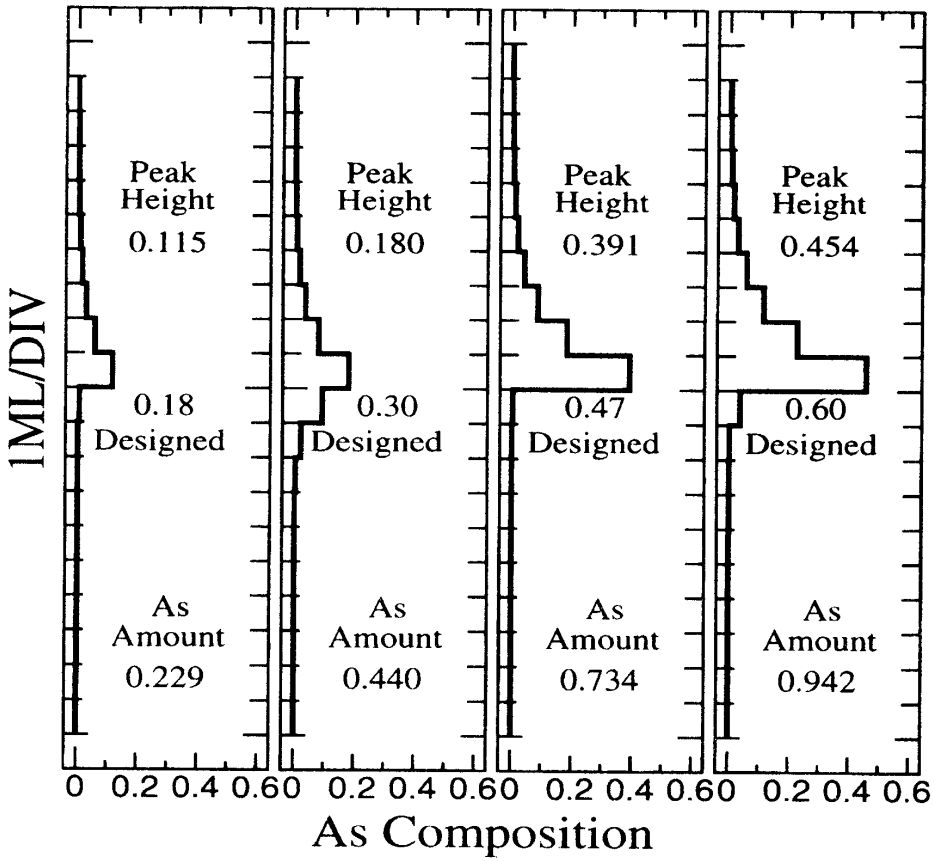
As in InP/InPAs heterostructure

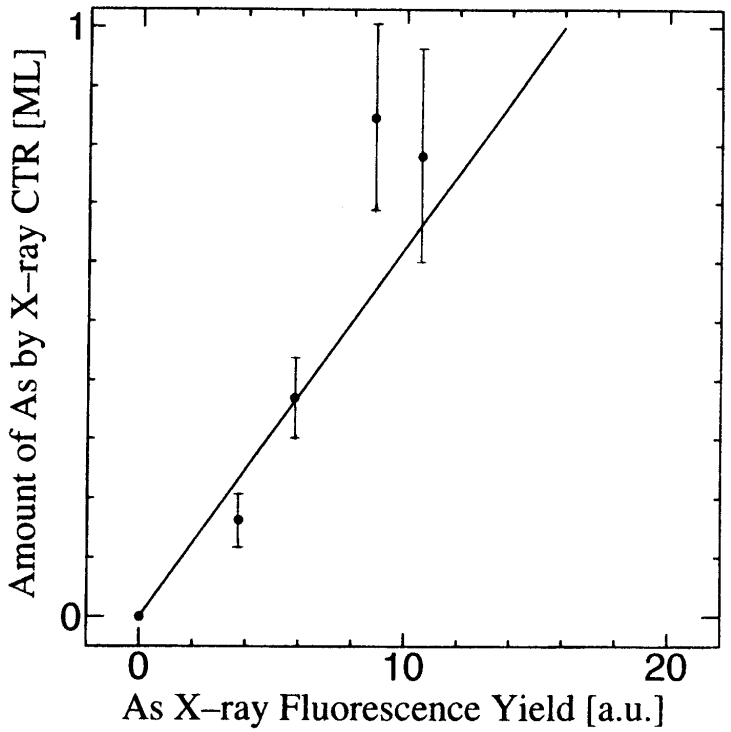




M. Tabuchi *et al* J. Appl. Phys 81 112 (1997)

CTR agrees with Fluorescence





M. Tabuchi *et al* J. Appl. Phys 81 112 (1997)

CTR of double-step structure

$$A_1(Q) = \sum_{j=0}^{\infty} \rho_1 e^{iQaj} + \sum_{j=n}^{\infty} \rho_2 e^{iQaj}$$
$$= (\rho_1 + \rho_2 e^{iQan}) \frac{1}{1-e^{iQa}}$$
$$= M_1(Q) \frac{1}{1-e^{iQa}}.$$

Perturbation limit: $\rho_2 << \rho_1$

$$|M_1(Q)| = |\rho_1 + \rho_2 e^{iQan}|$$

$$\simeq \rho_1 + \rho_2 \cos(Qan)$$

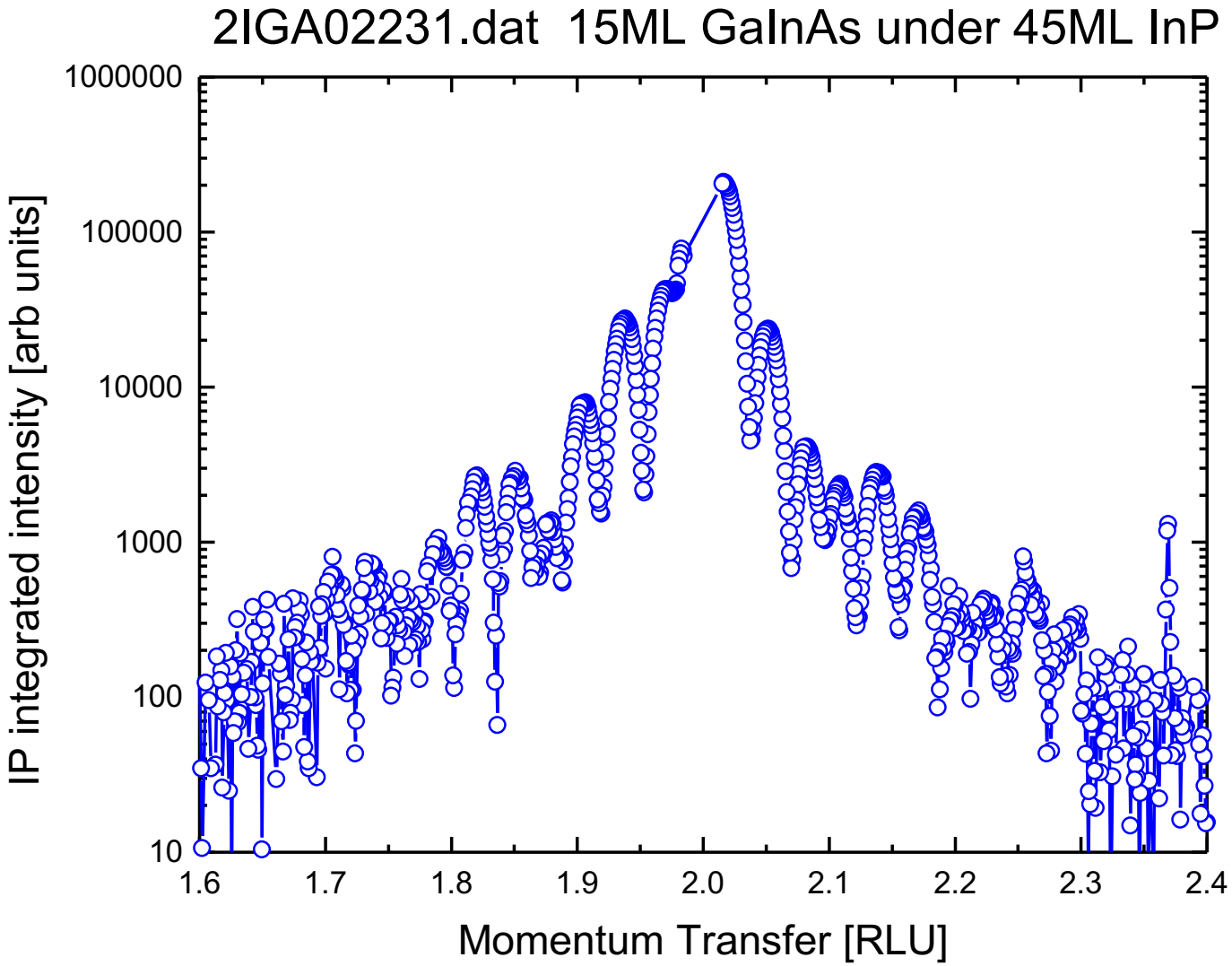
Data Analysis Procedure

$$M(Q) = \sqrt{I(Q)}/|A_0(Q)|$$

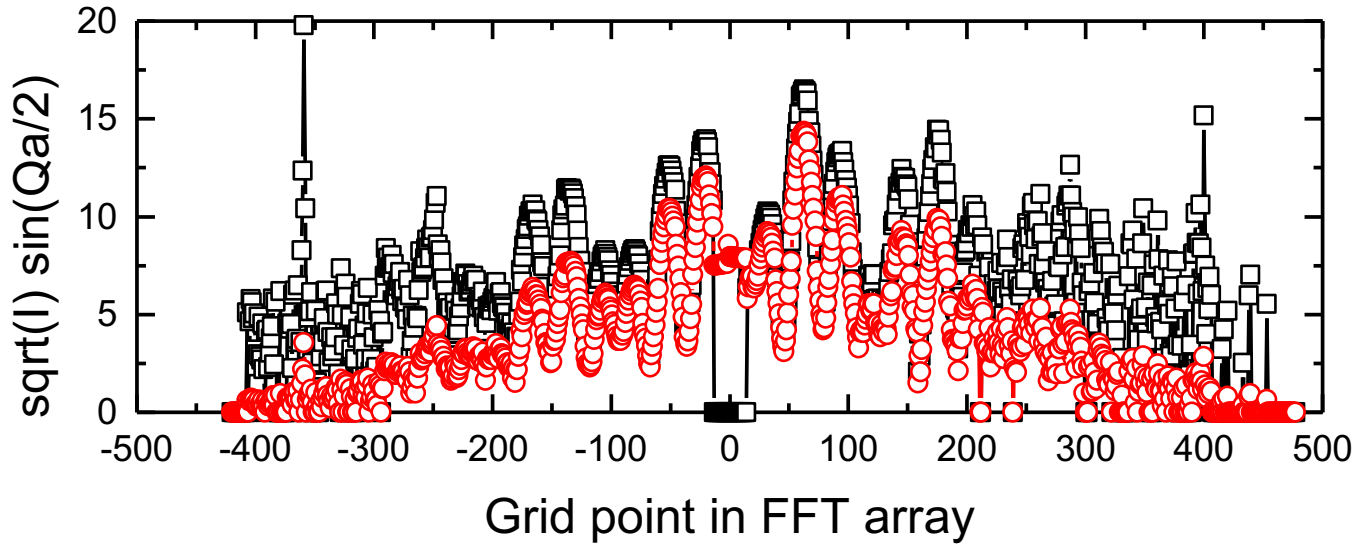
$$= \sqrt{I(Q)}\sin(Qa/2).$$

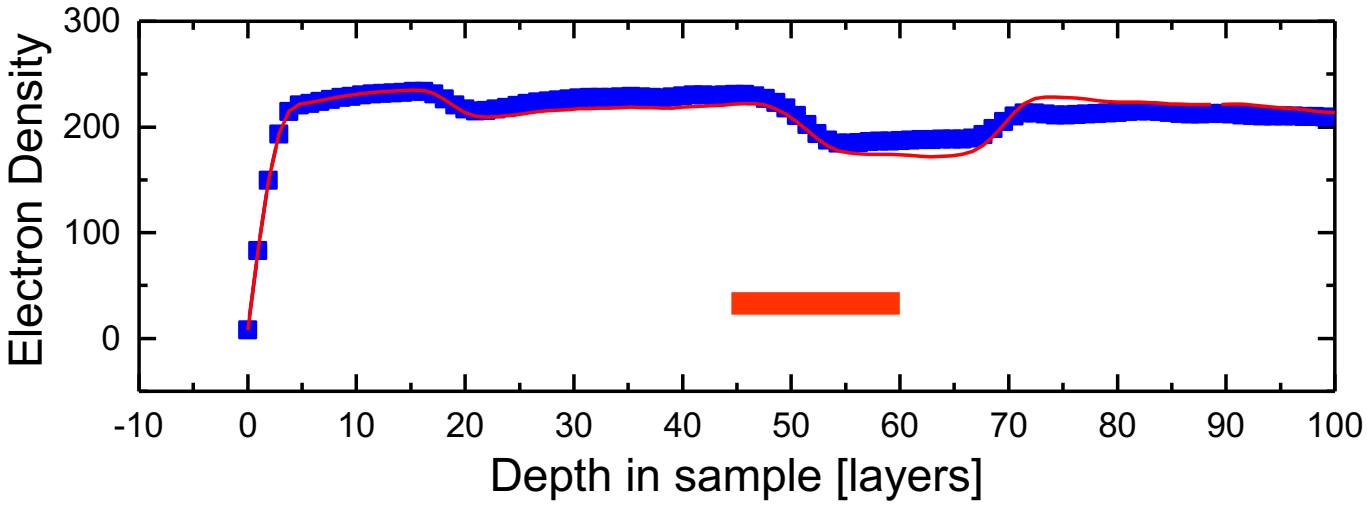
$$\rho(z) = \int_0^z \mathcal{F}\{\sqrt{I(Q)}\sin(Qa/2)\}dz'$$

Test InP/GaAs/InP heterostructure



Input and Output of FFT



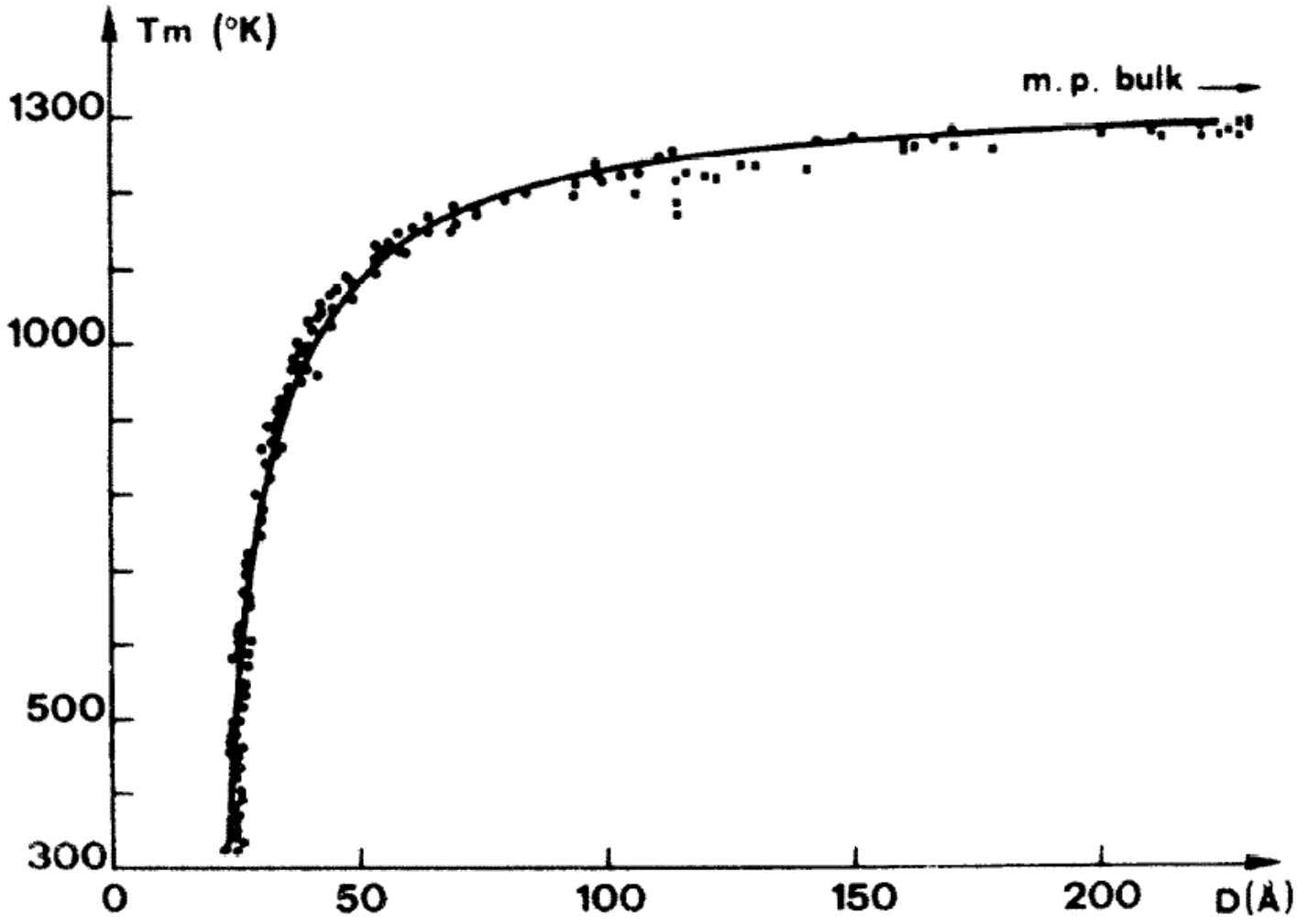


Goals of Coherent Diffraction

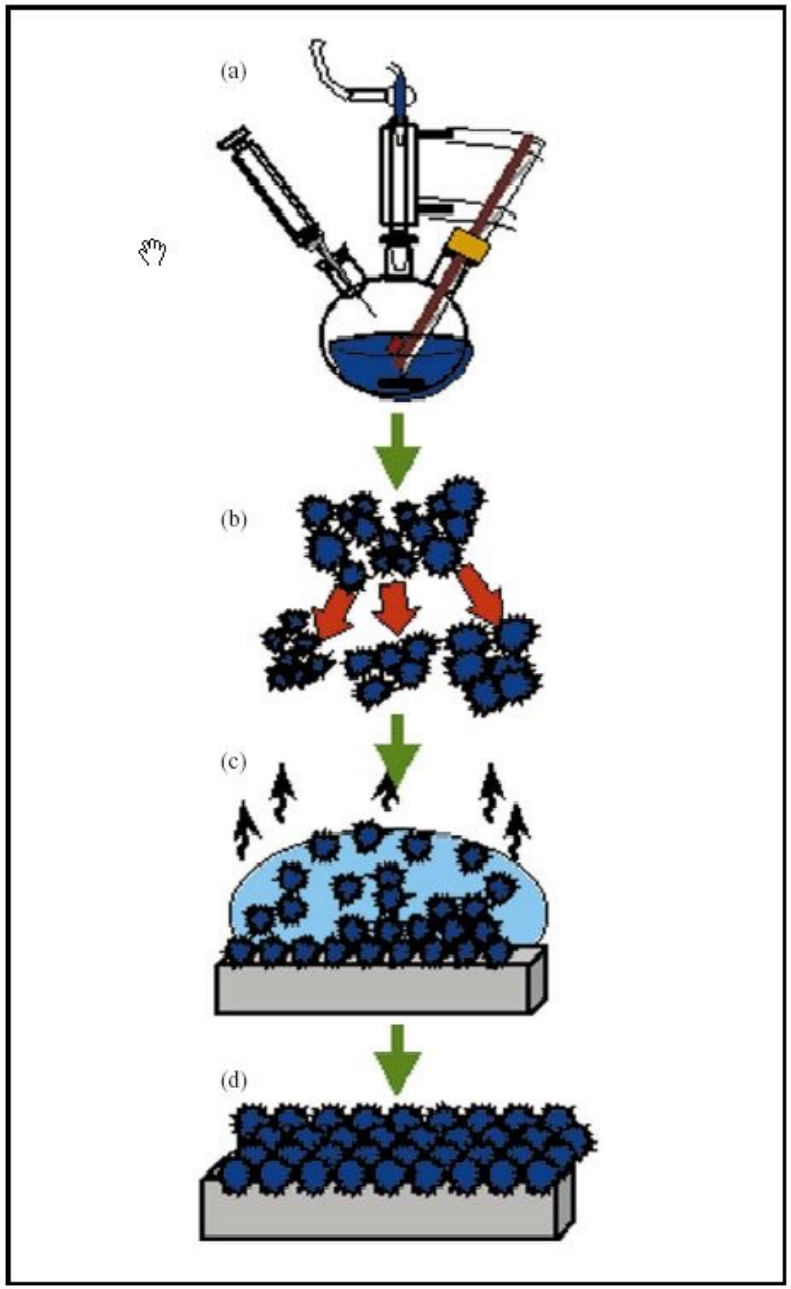
- Thermodynamic fluctuations
  - No ensemble average in CXD
- Probe of structure on nm scale
  - 1D, 2D and 3D
  - non-periodic object gives continuous F(**q**)
- Oversampling (in reciprocal space) permits solution of the phase problem

Size-dependent Melting of Au Particles

P. Buffat and J-P. Borel, Phys. Rev. A 2287-97 (1975)

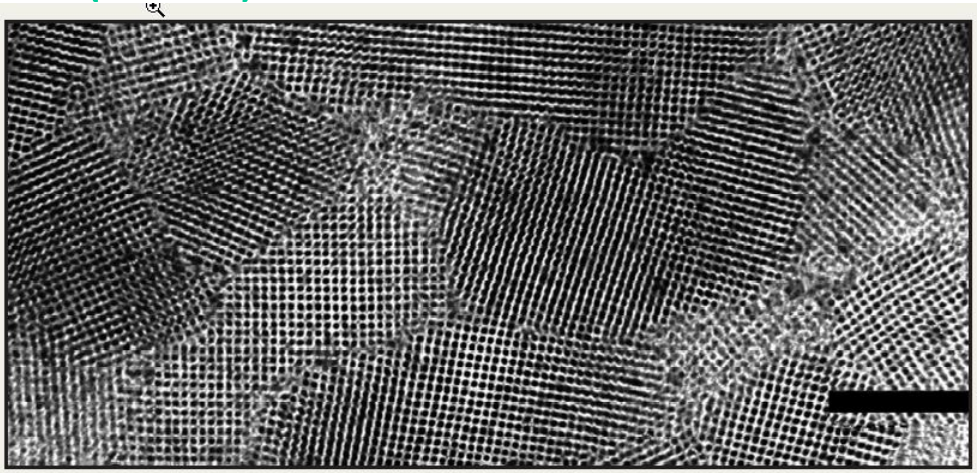


Chemical Synthesis of Nanocrystals

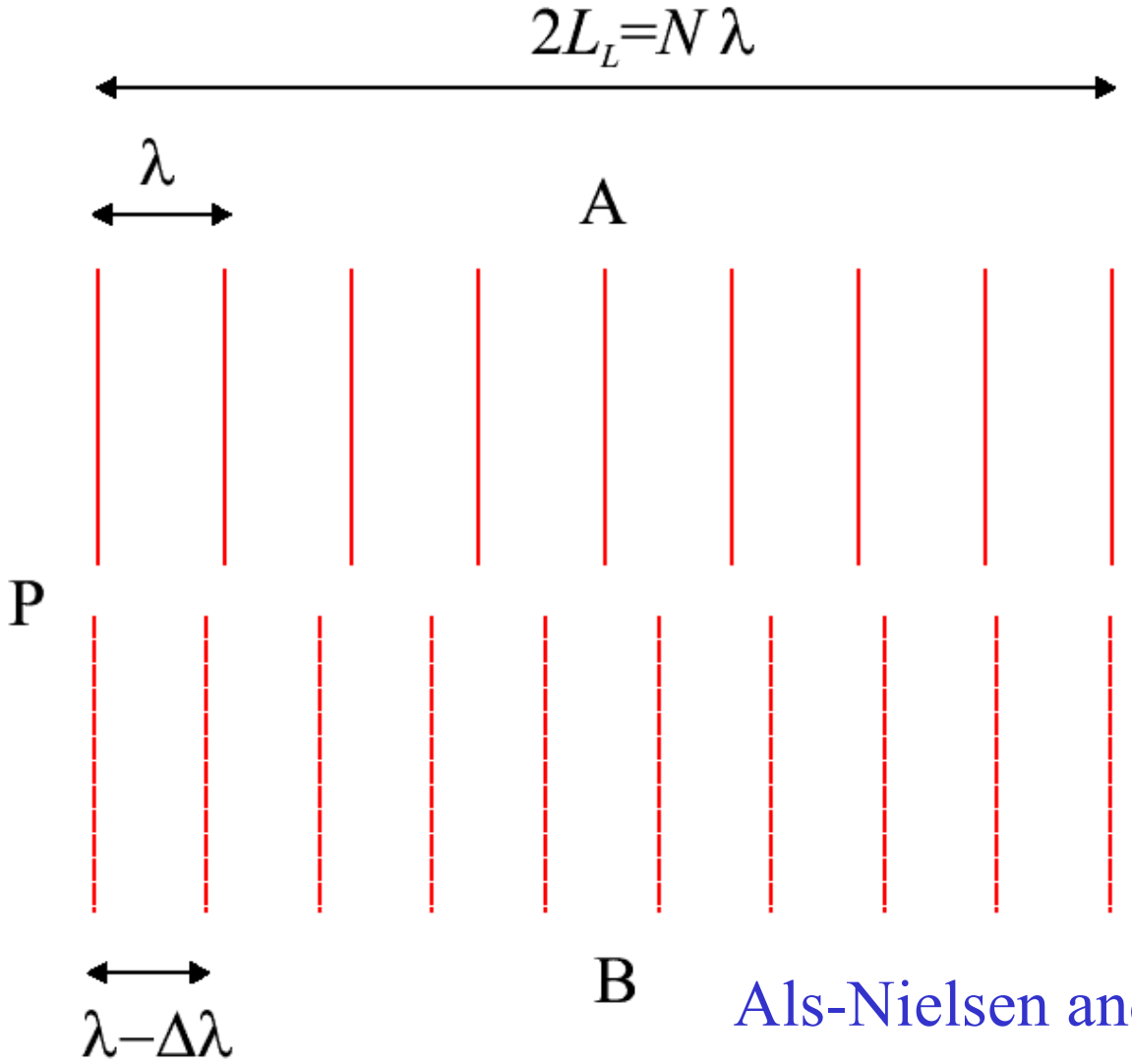


- Reactants introduced rapidly
- High temperature solvent
- Surfactant/organic capping agent
- Square superlattice (200nm scale)

C. B. Murray, IBM J. Res. & Dev. **45** 47 (2001)



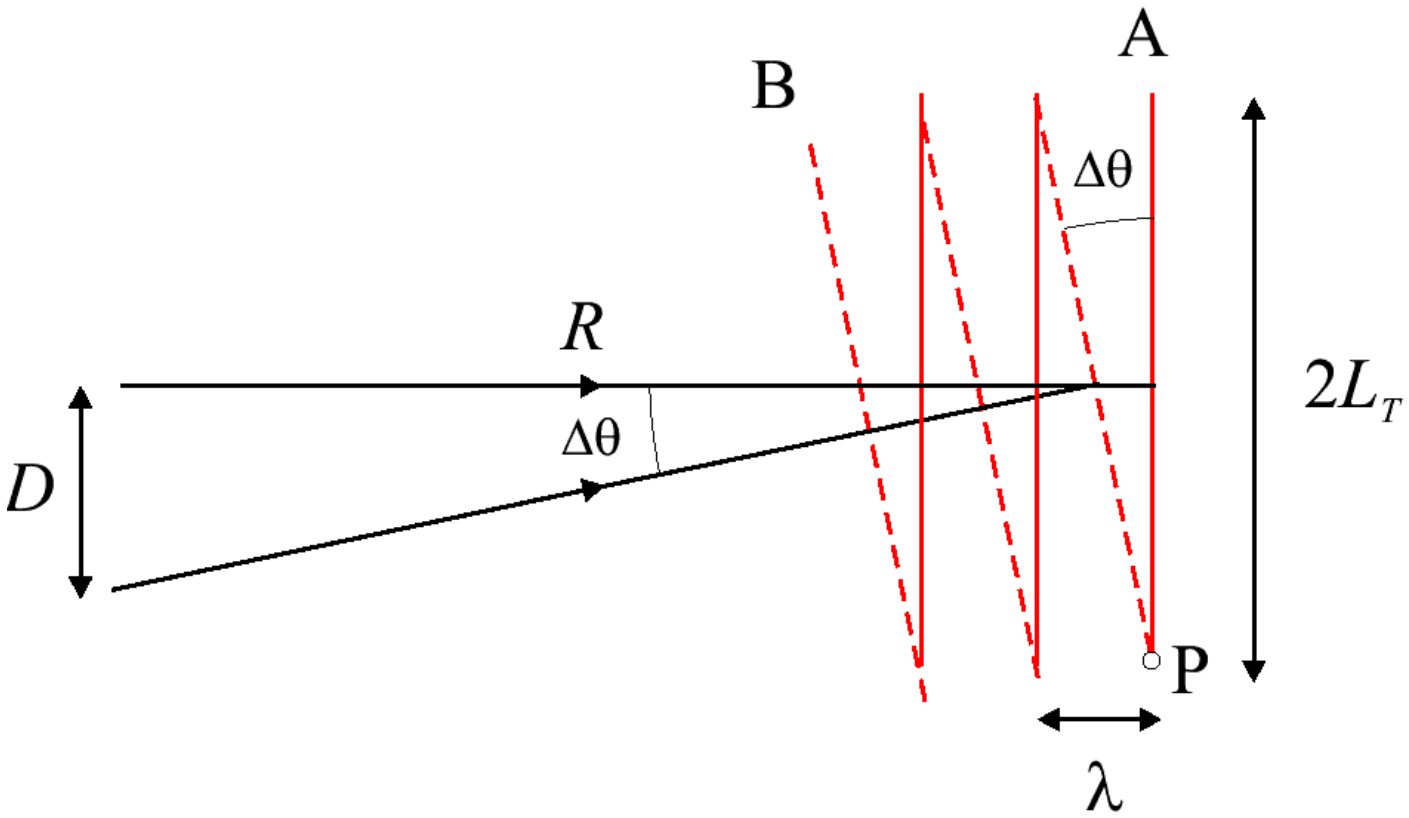
Longitudinal Coherence



$$L_L = \frac{1}{2}\frac{\lambda^2}{\Delta\lambda}$$

Als-Nielsen and McMorrow (2001)

Lateral (Transverse) Coherence



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

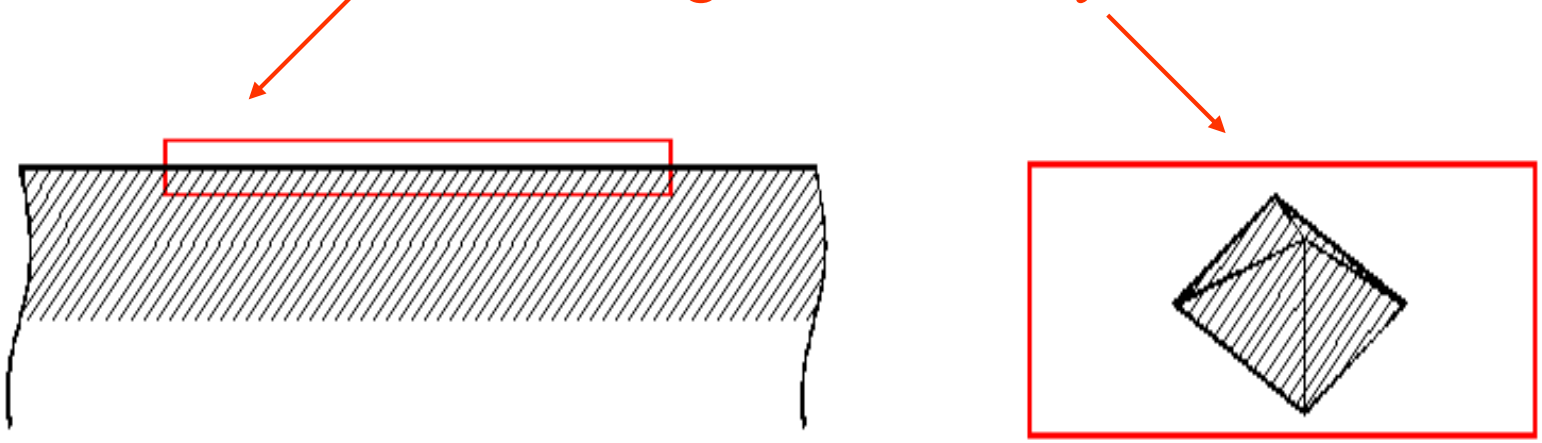
Als-Nielsen and McMorrow (2001)

Coherence at Spring8, APS or ESRF

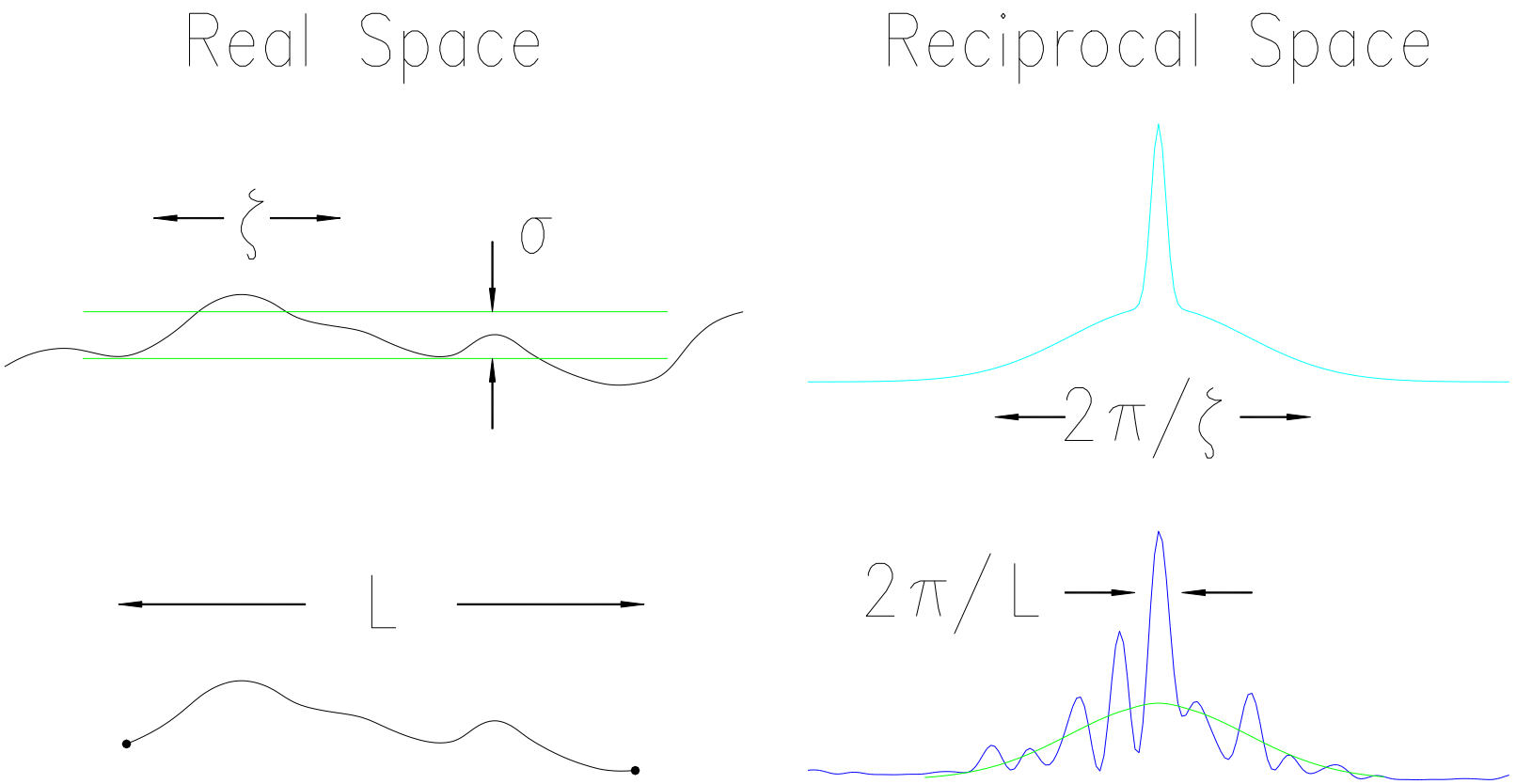
Typical of 3rd Generation (undulator) Synchrotron Source

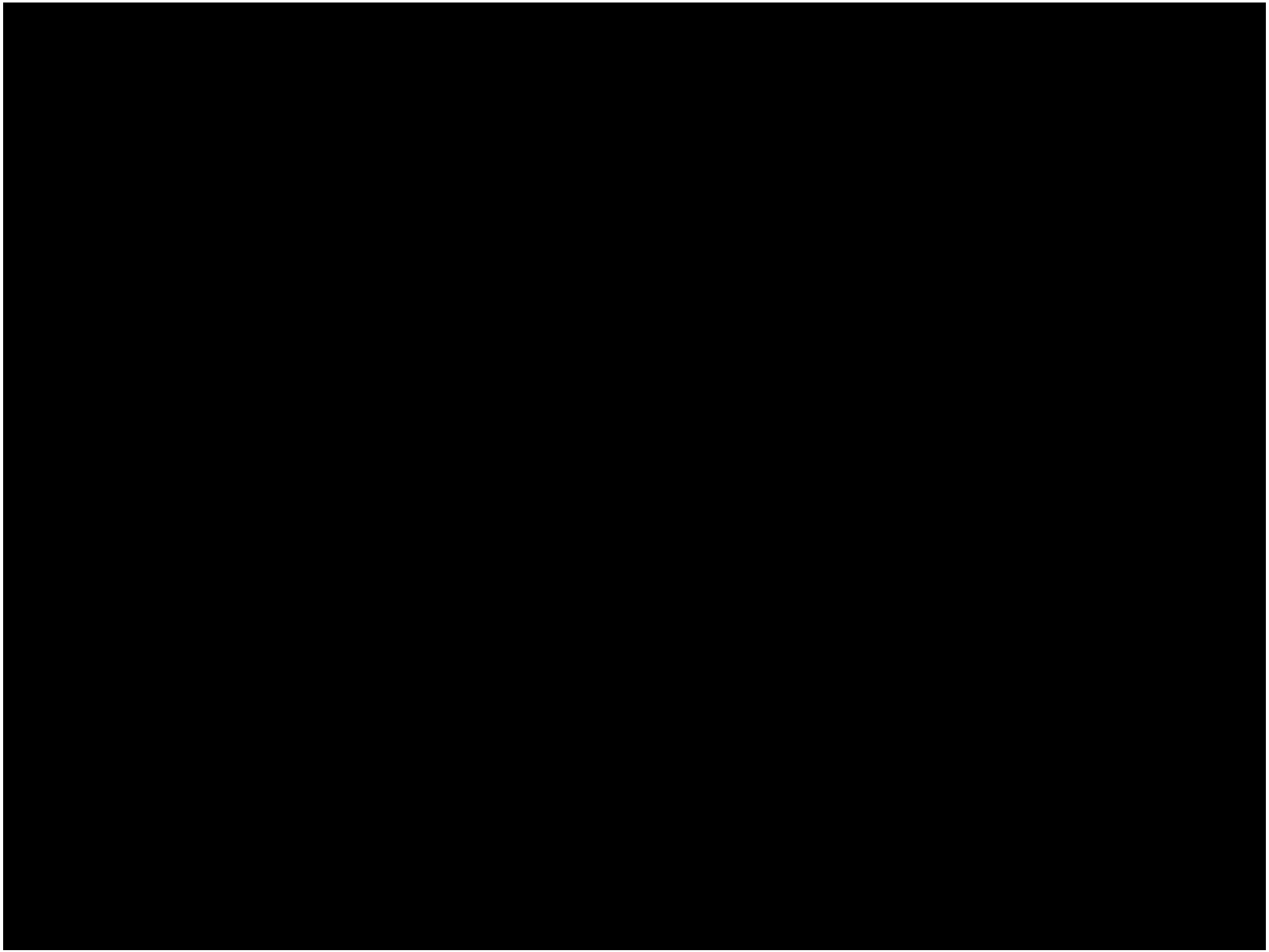
Coherence of	ξ_{VER}	ξ_{HORIZ}	ξ_{LONG}	Flux
Raw Undulator	35μm	9μm	0.004μm	2×10^{12}
Si(111) Monochromator	35μm	9μm	1μm	1×10^{10}
C(111) Monochromator	35μm	9μm	3μm	3×10^{9}

Coherent region defined by slits

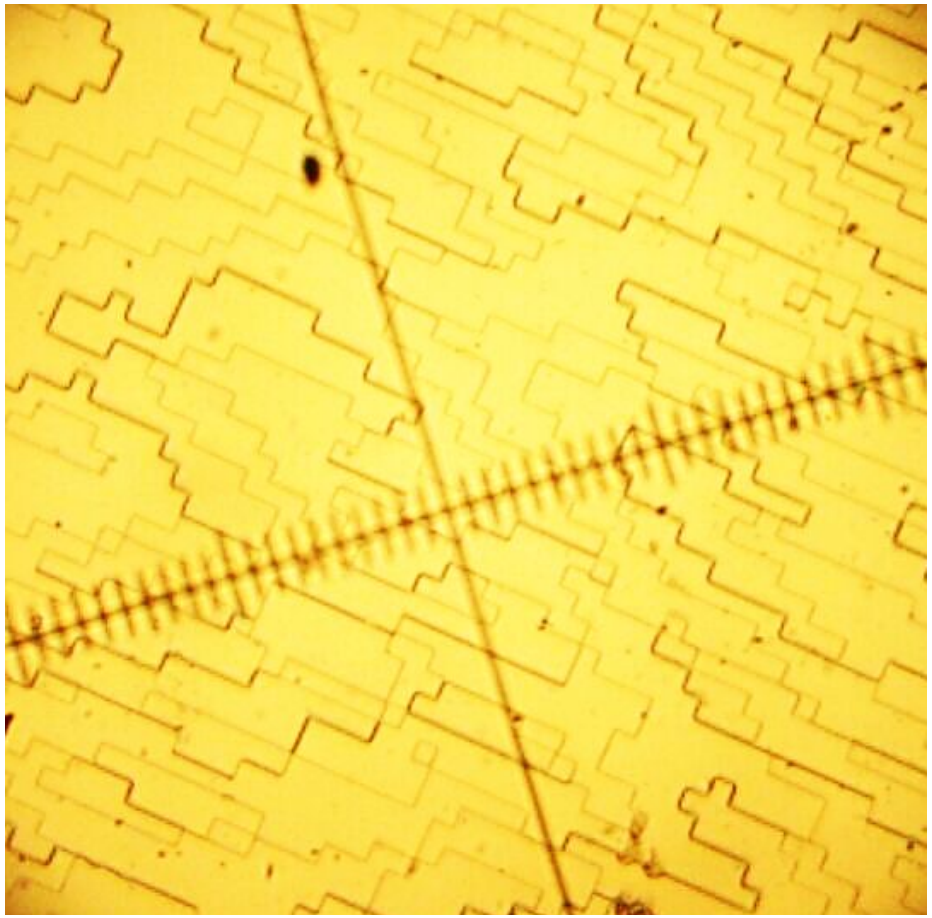


Diffuse Scattering acquires Structure using CXD

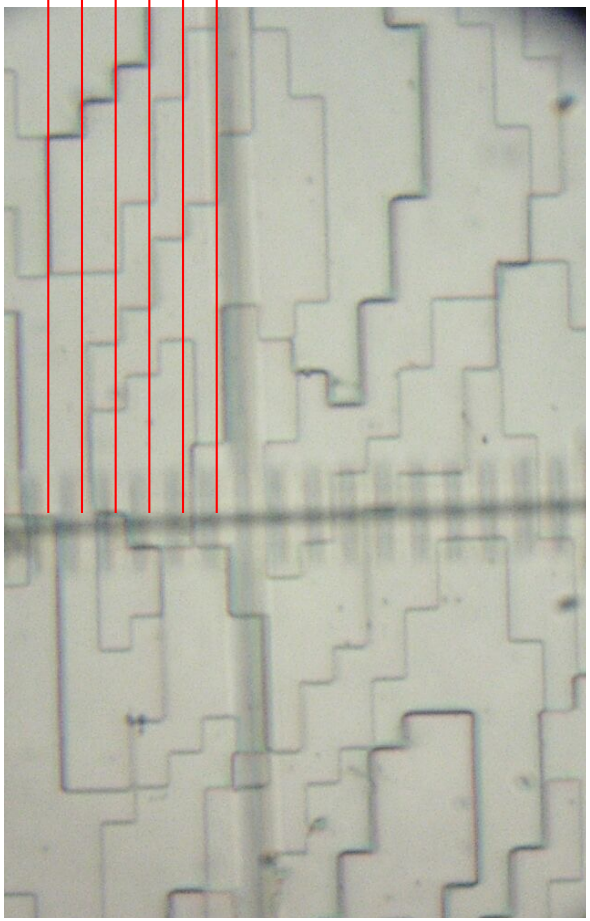




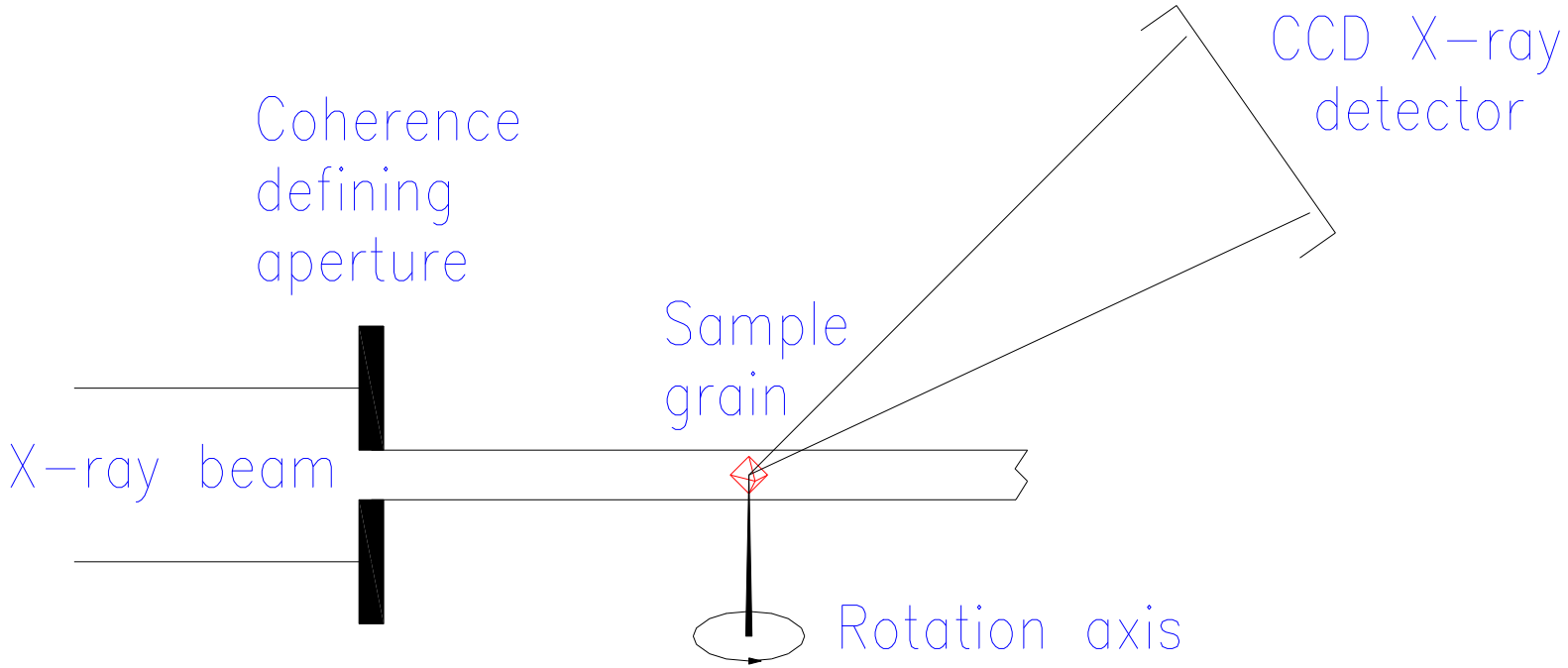
Microscope Images of Gratings



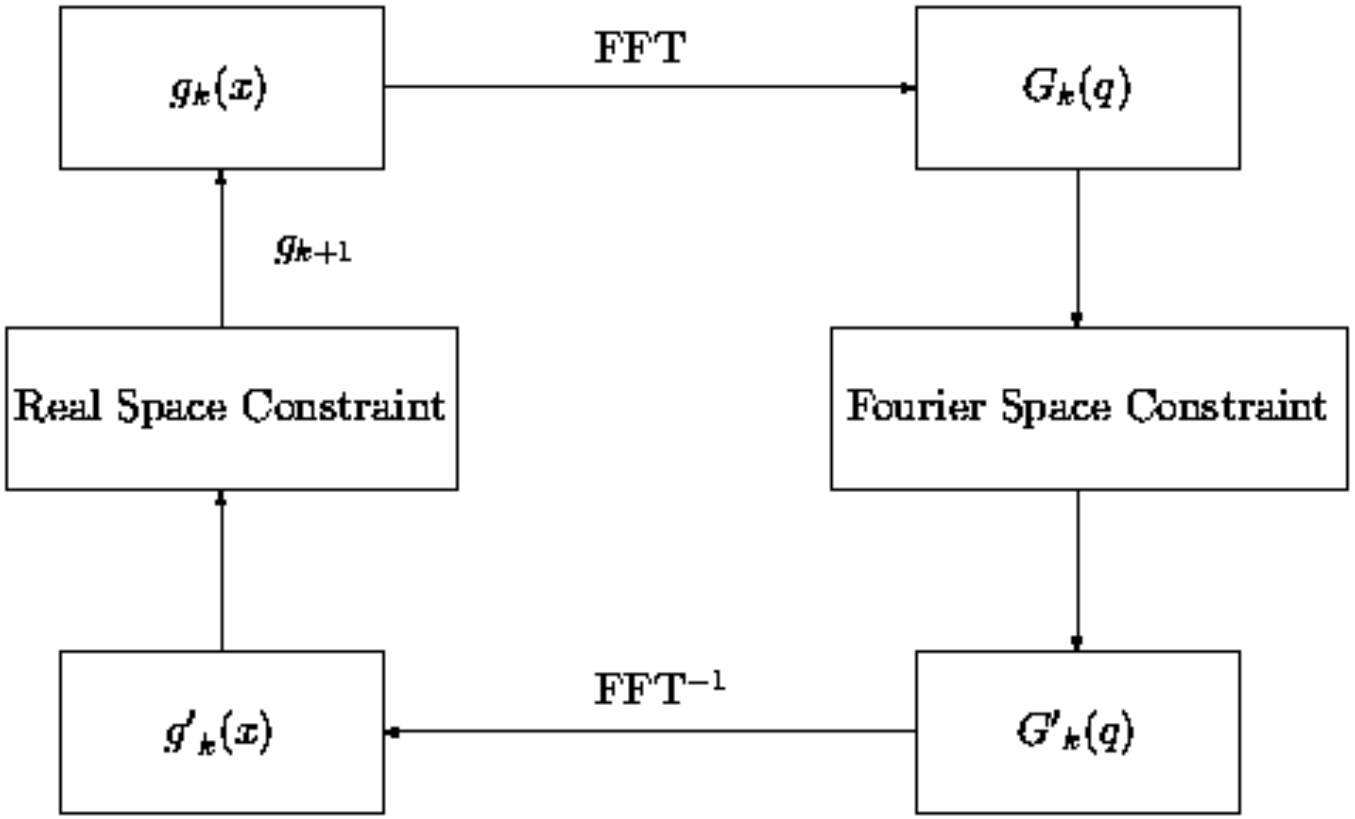
5μm grid lines



Lensless X-ray Microscope



Generic "Error Reduction" method



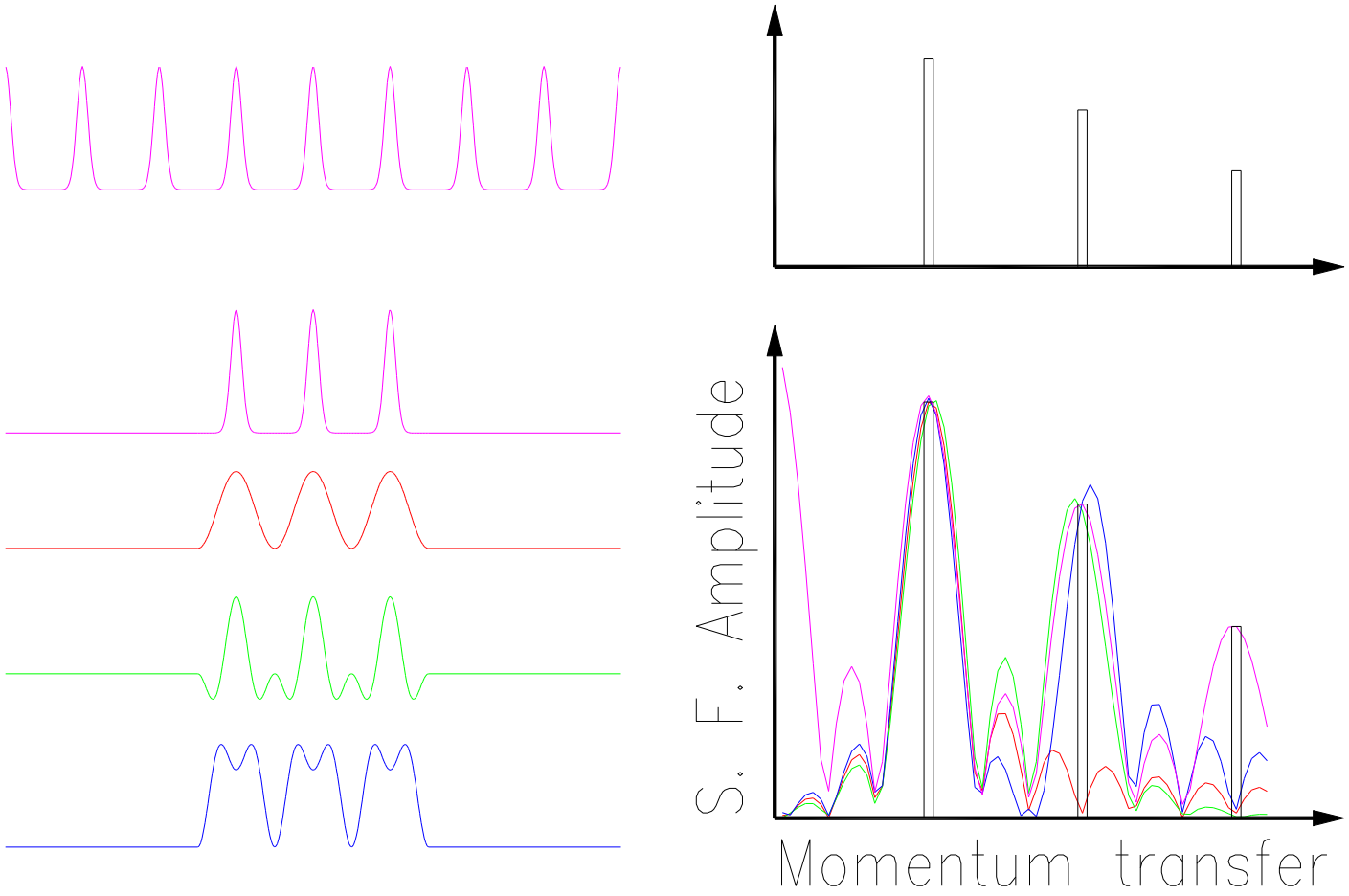
J. R. Fienup Appl. Opt. 21 2758 (1982)
R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)

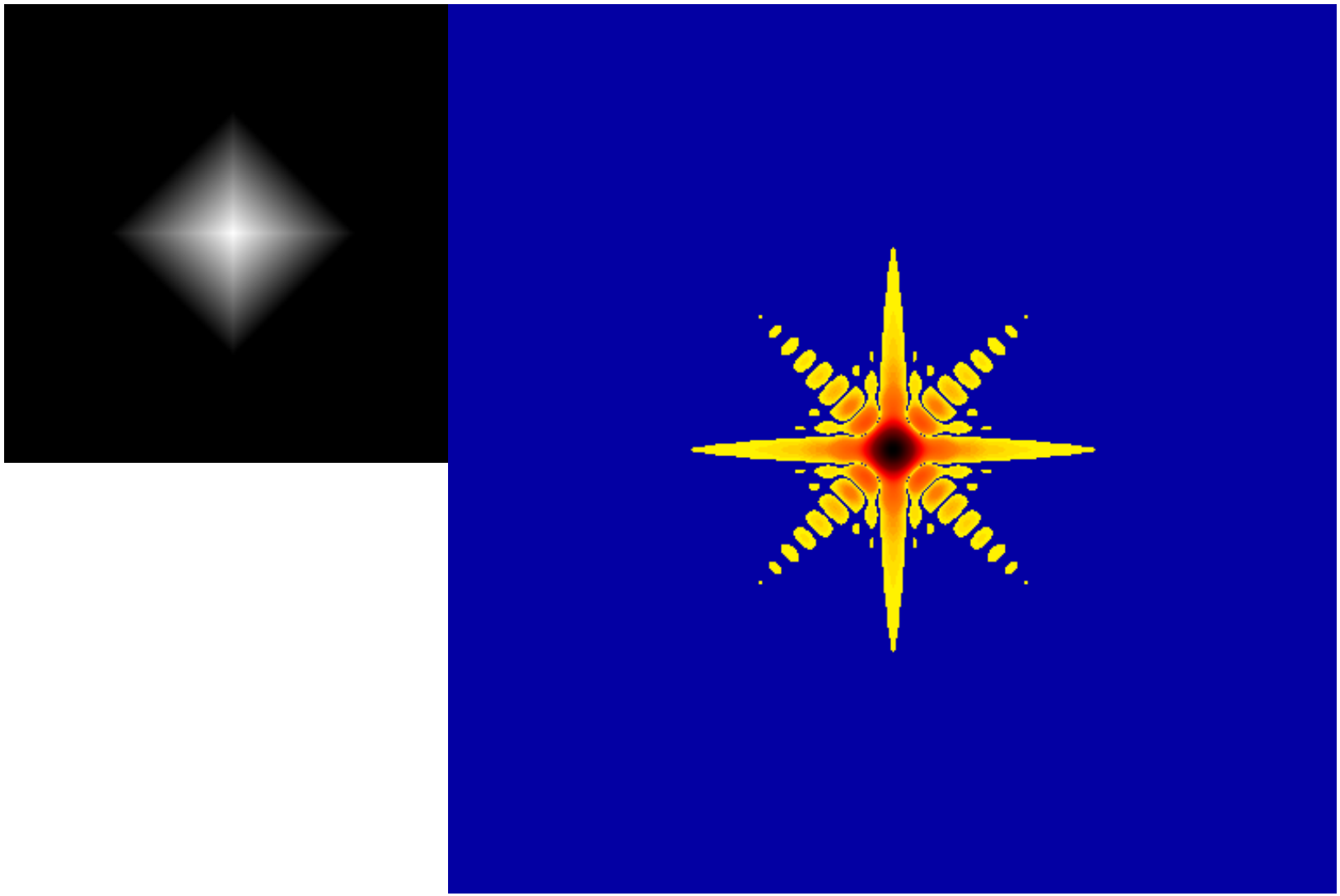
Real-space Constraints in Crystallograhy

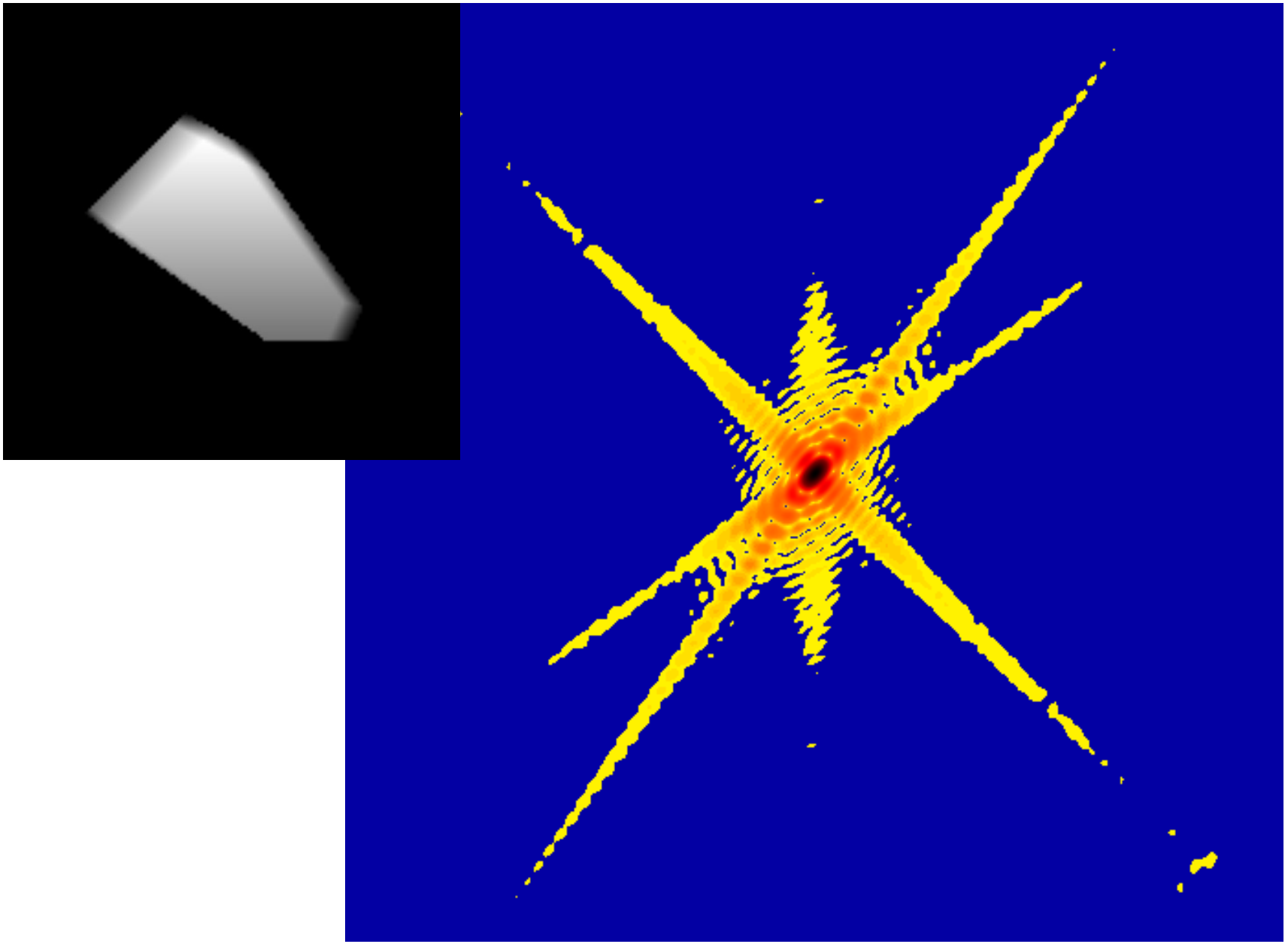
R. P. Millane, J. Opt. Soc Am. A **13** 725 (1996)

- '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)

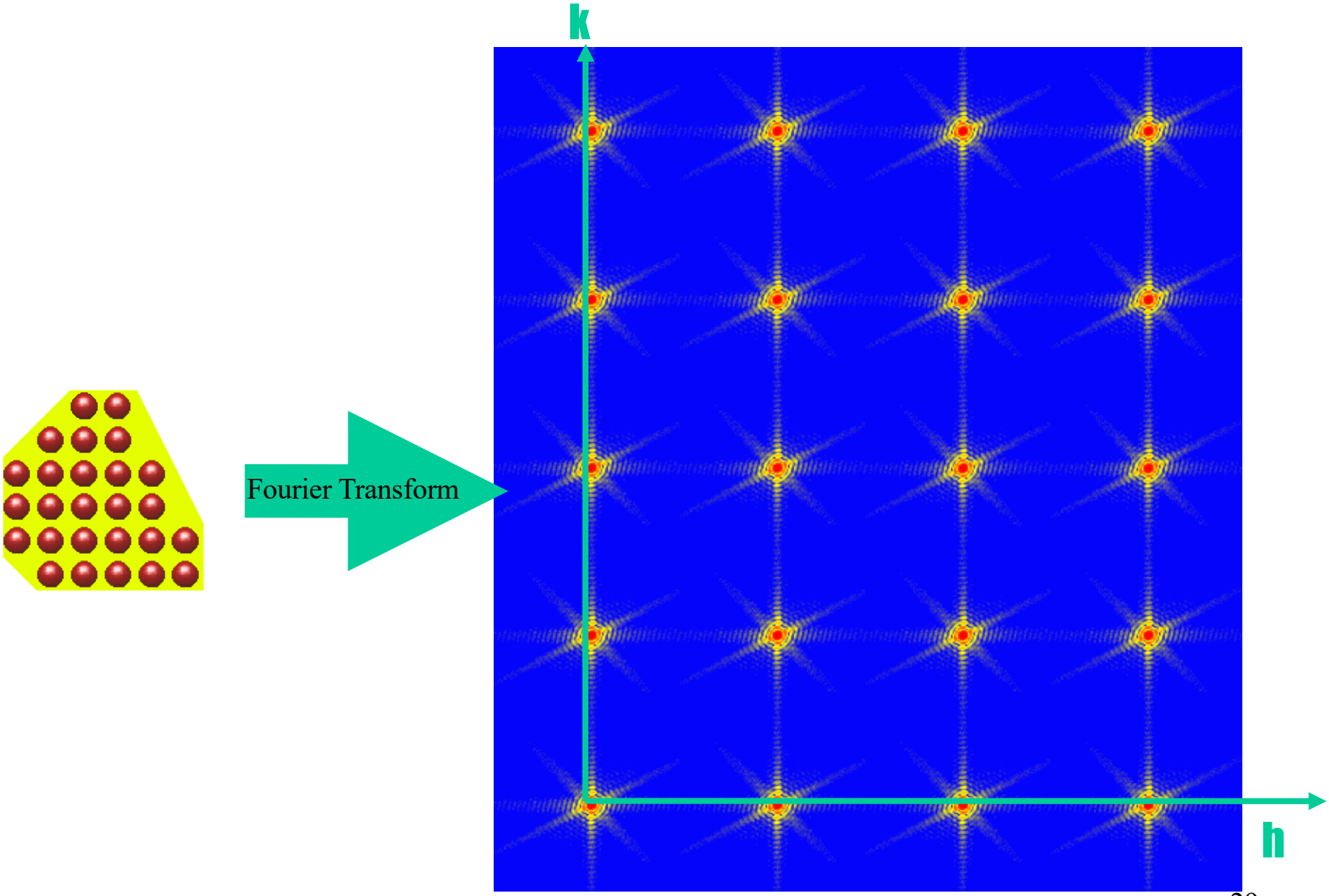
Phase Problem: Finite-size Effect





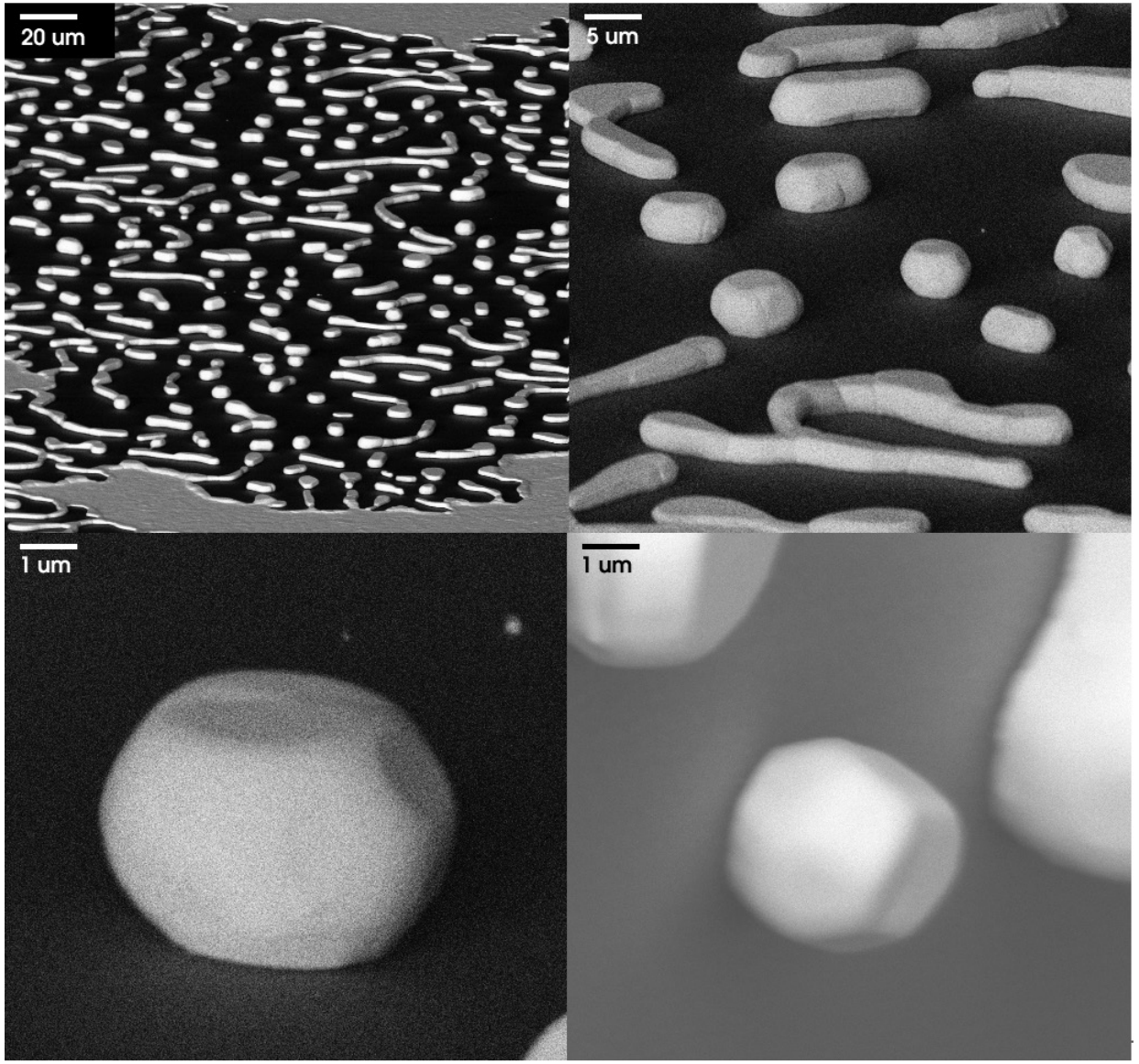


Coherent Diffraction from Crystals



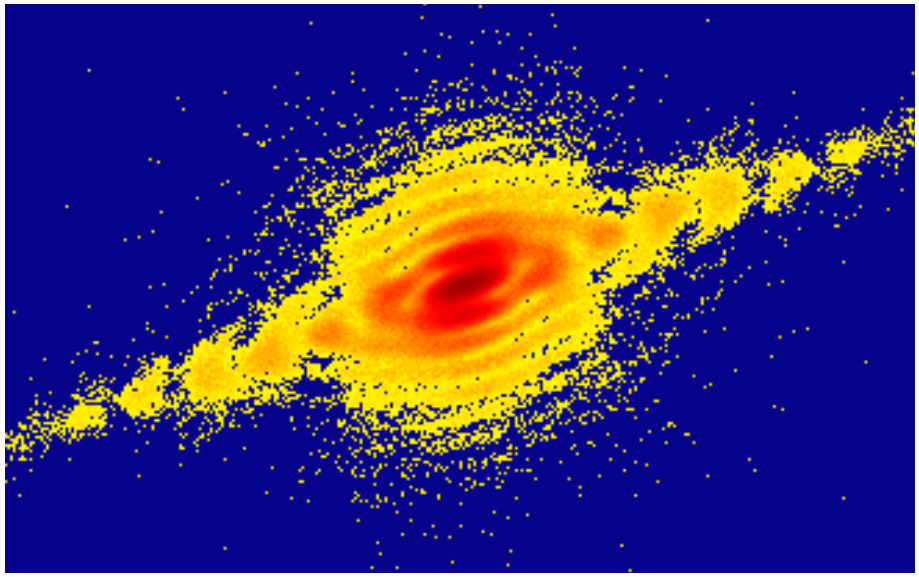
SEMS

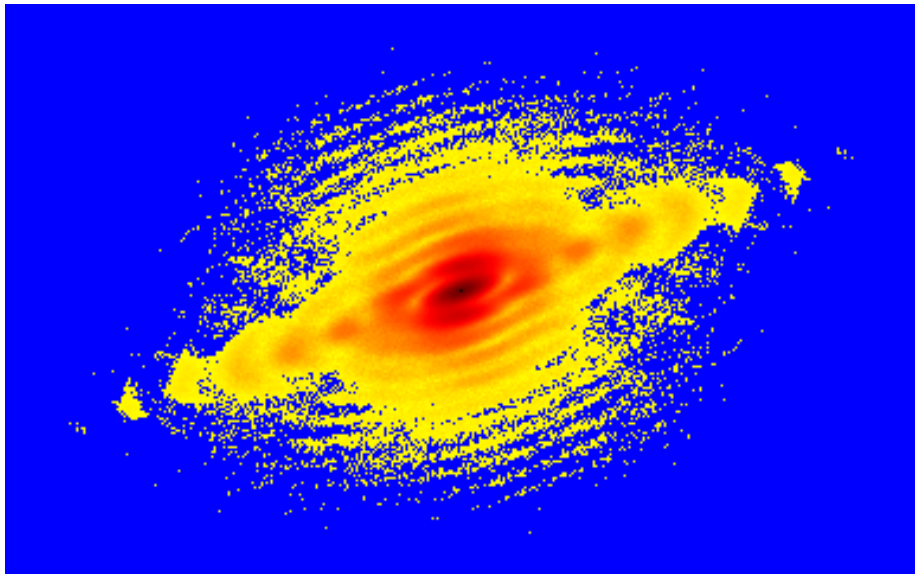
- Au blanket film
- Quartz substrate
- Annealed at 950°C for 70 hrs.

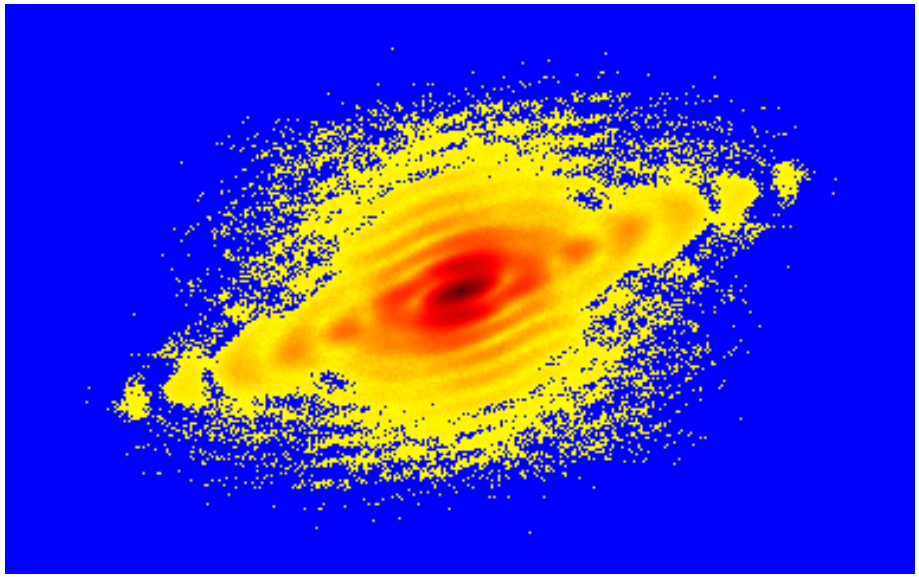


Symmetrized Data and two best fits

Chisq=0.0005

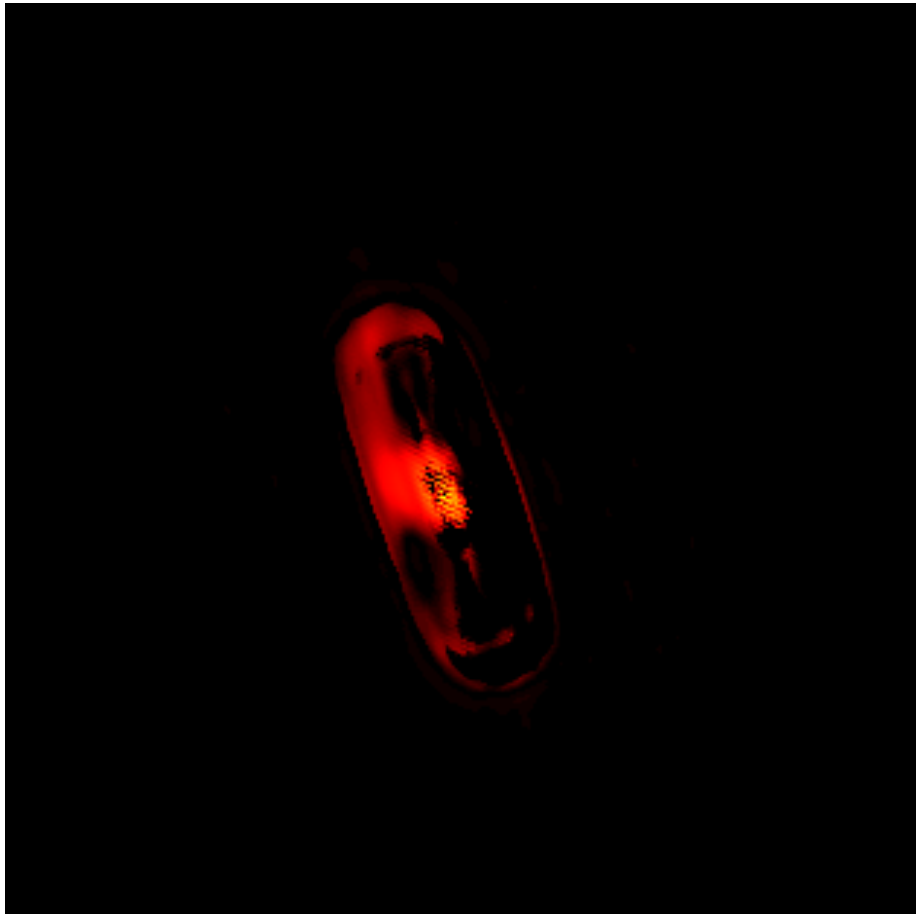


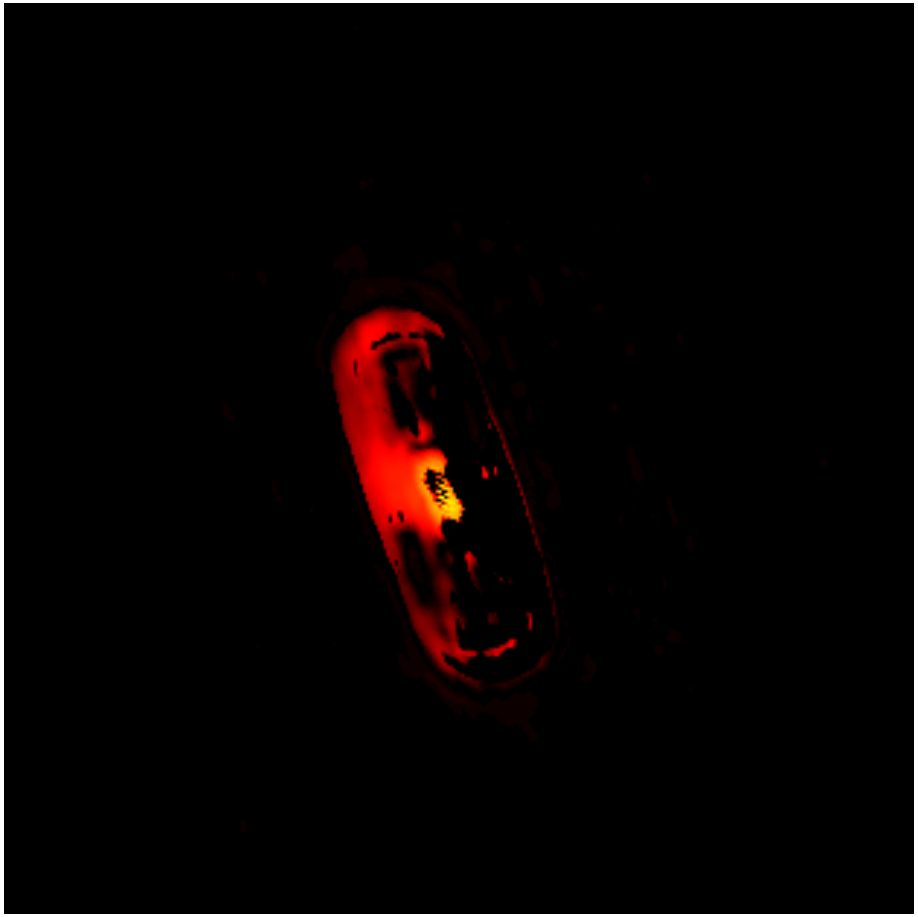




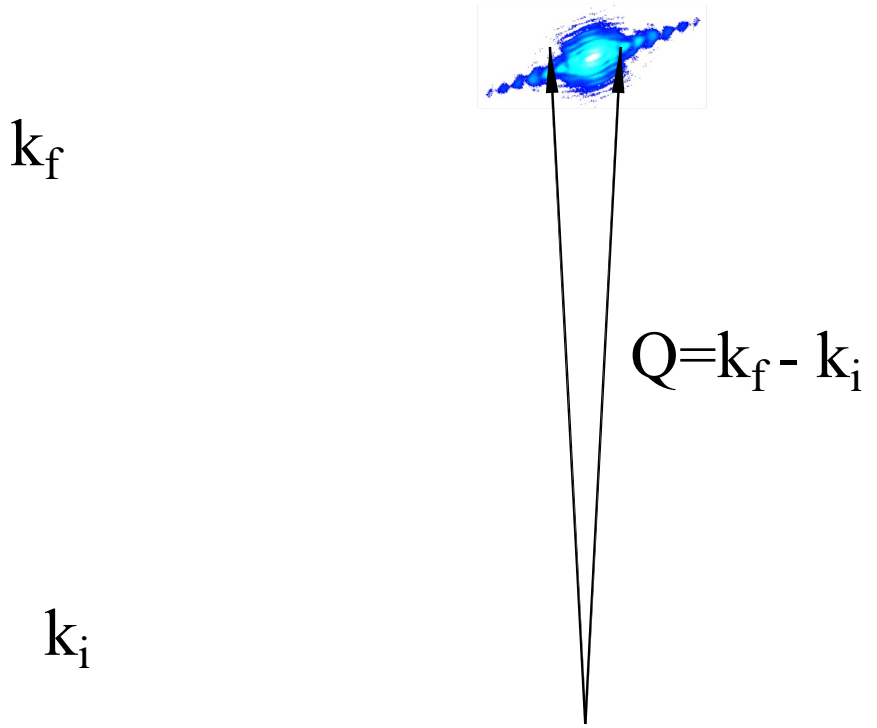
2D Reconstructions

chisquare = 0.0005

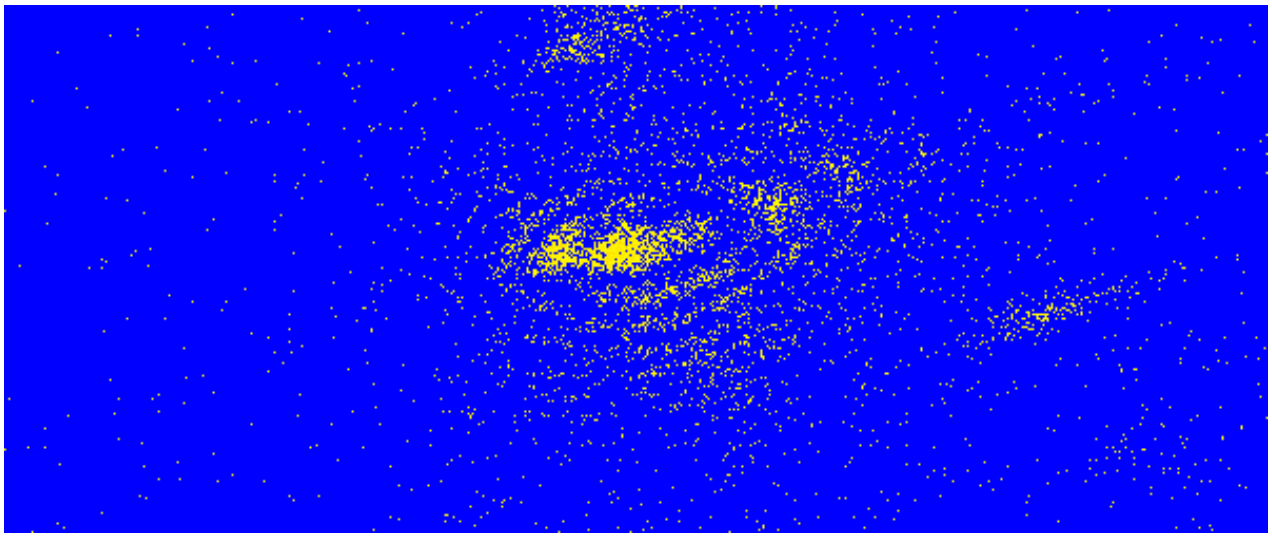




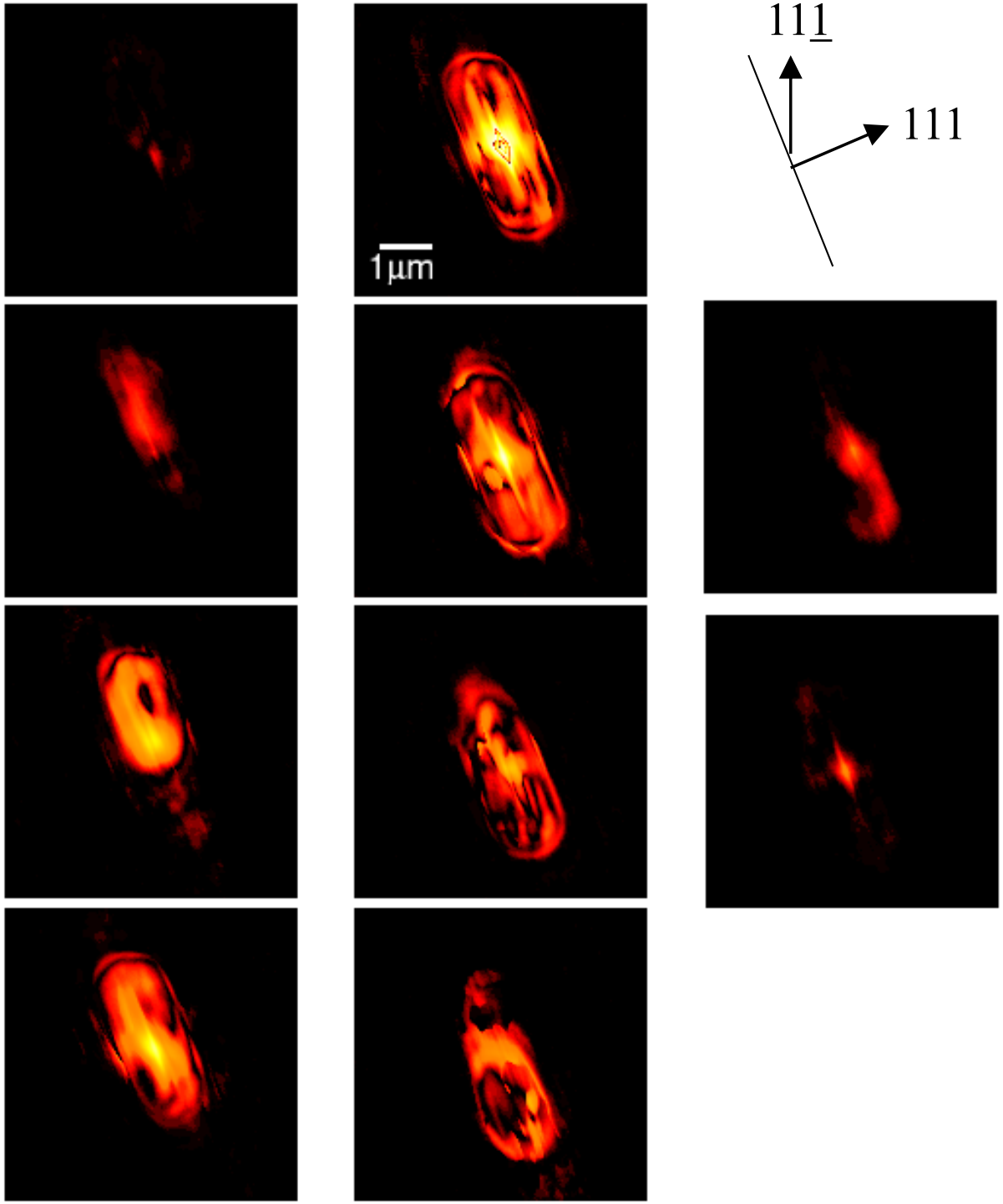
3D Diffraction Method



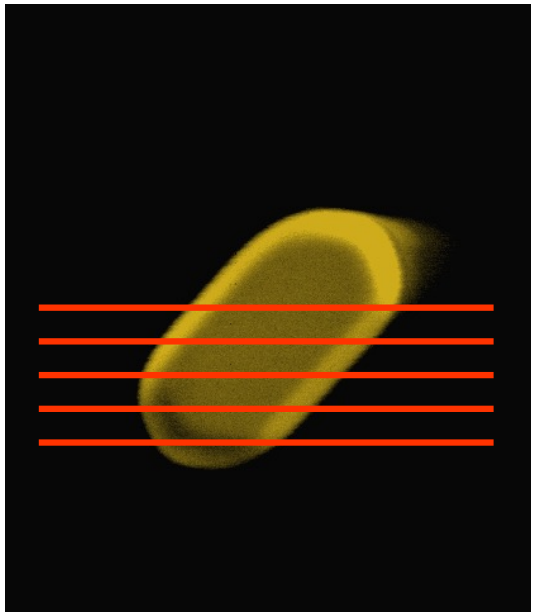
3D Diffraction Data
1 micron Au crystal



* Center is Symmetric *

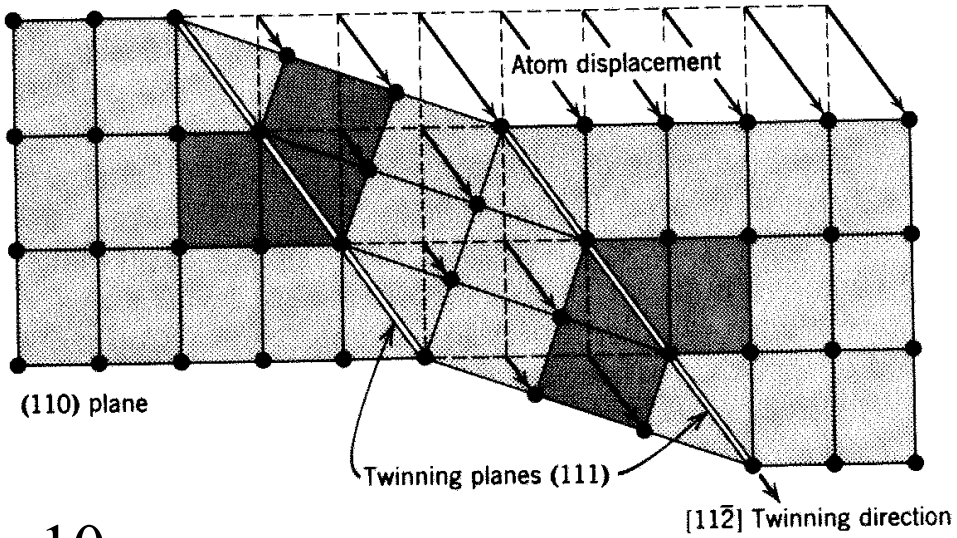


Slices through plan view SEM:

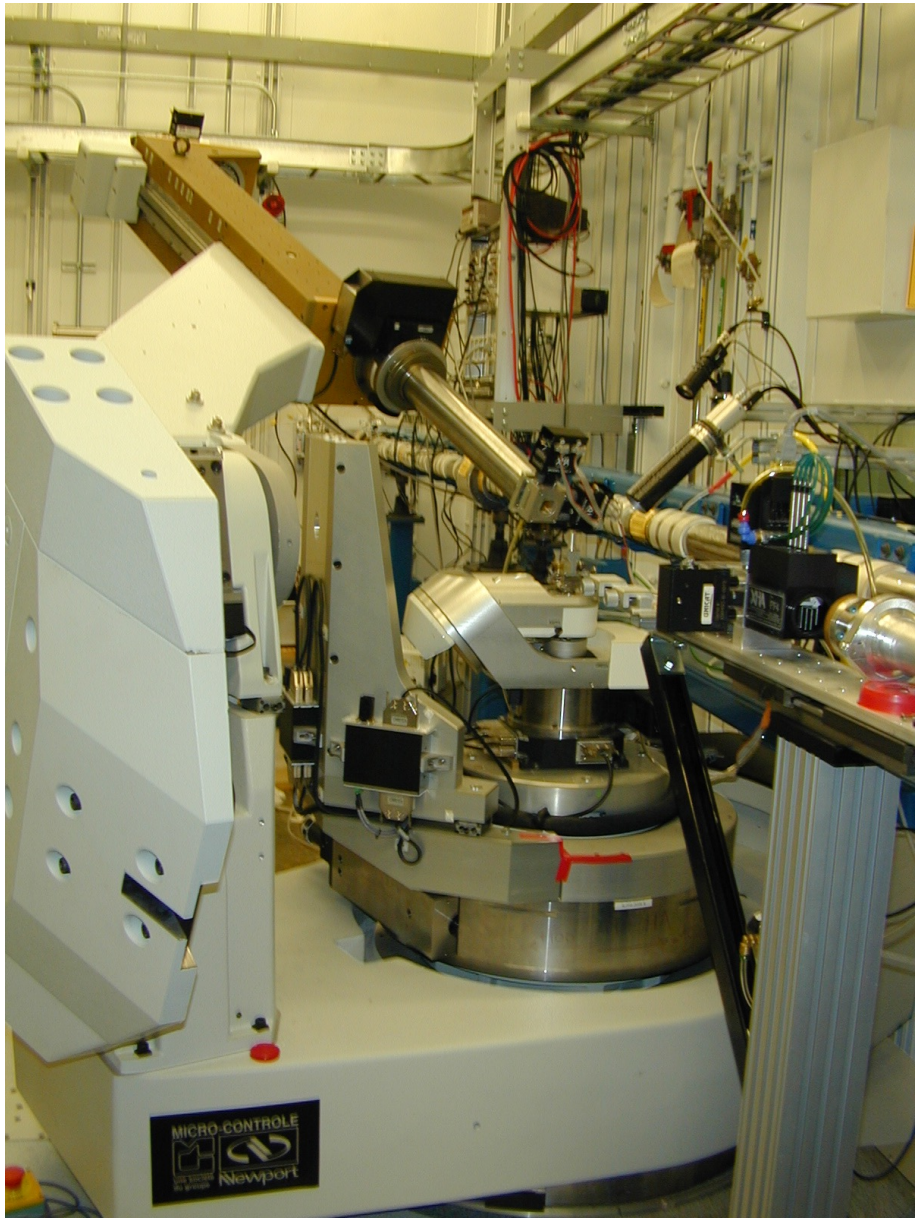


Twinning in deformed FCC metals

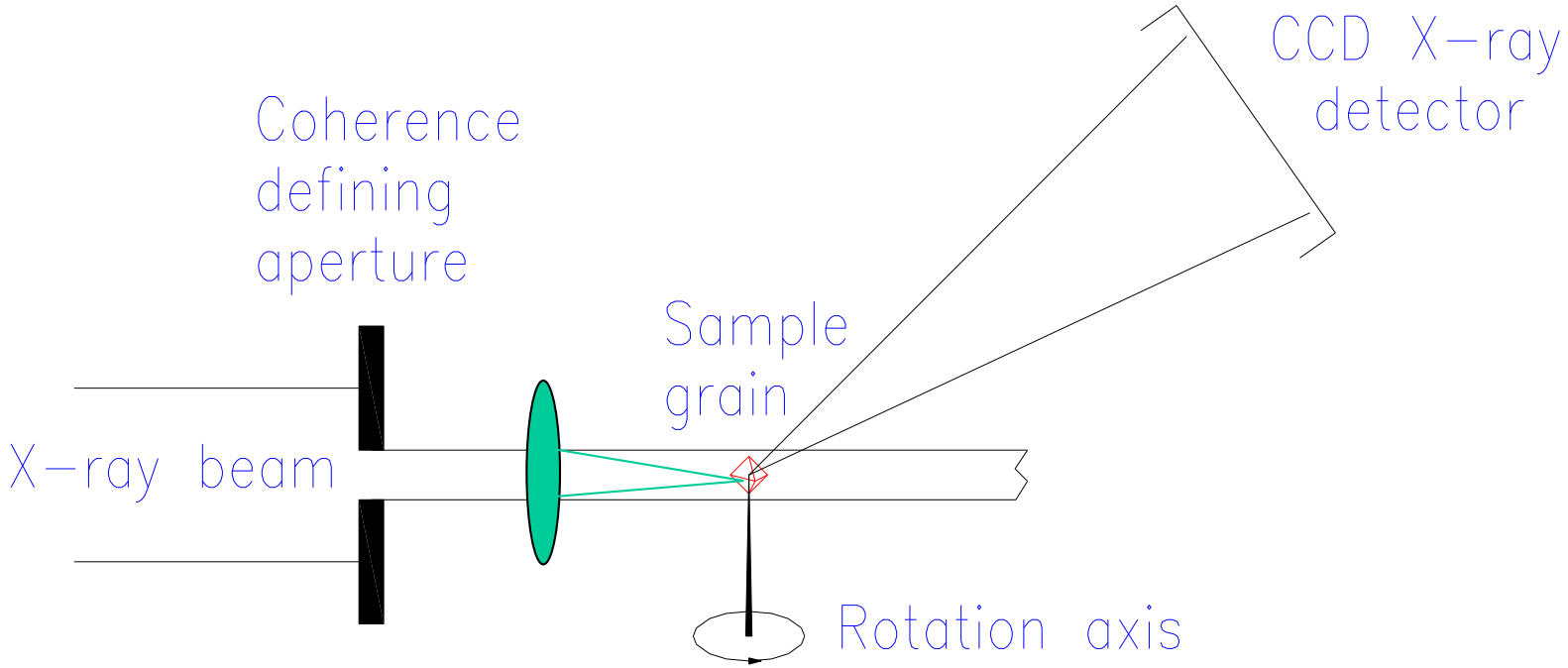
J. Wulff, "Structure and Property of Materials III" (1965)



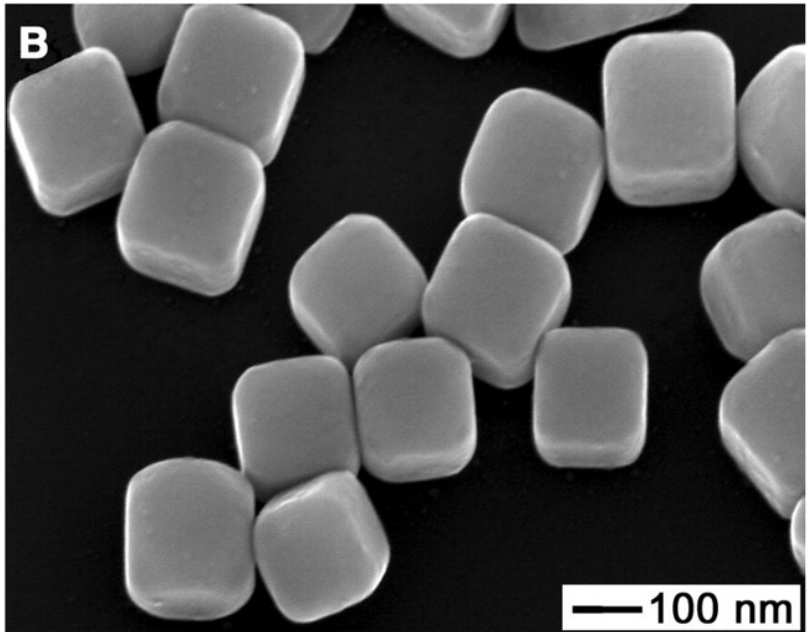
~10μm
Cu

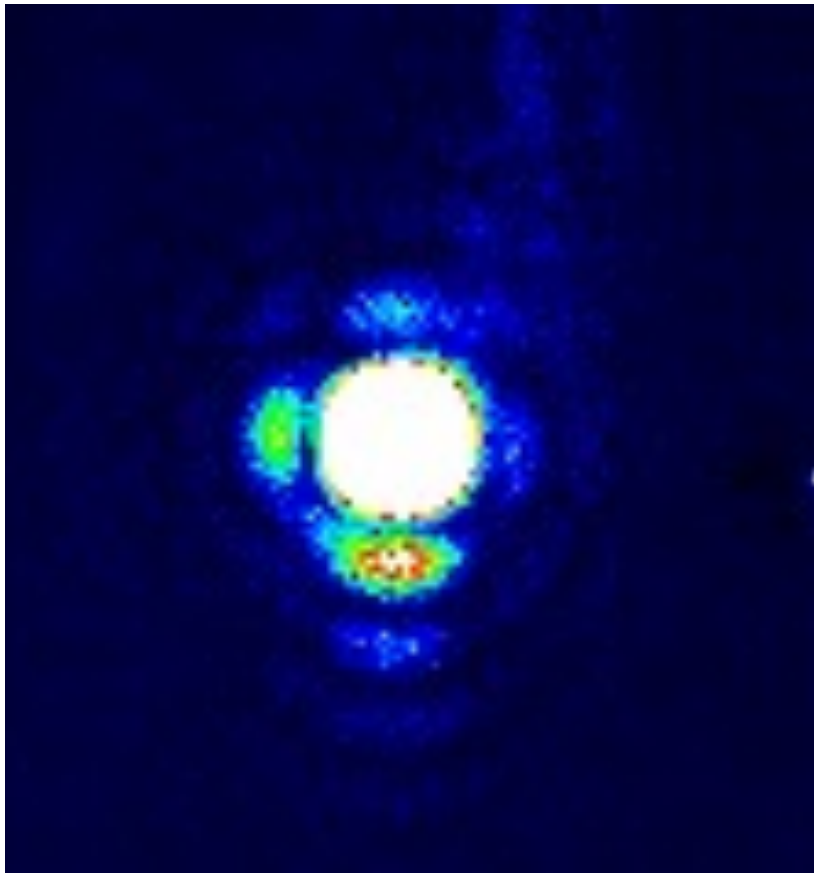

MICRO-CONTROLE
Newport

Lensless X-ray Microscope



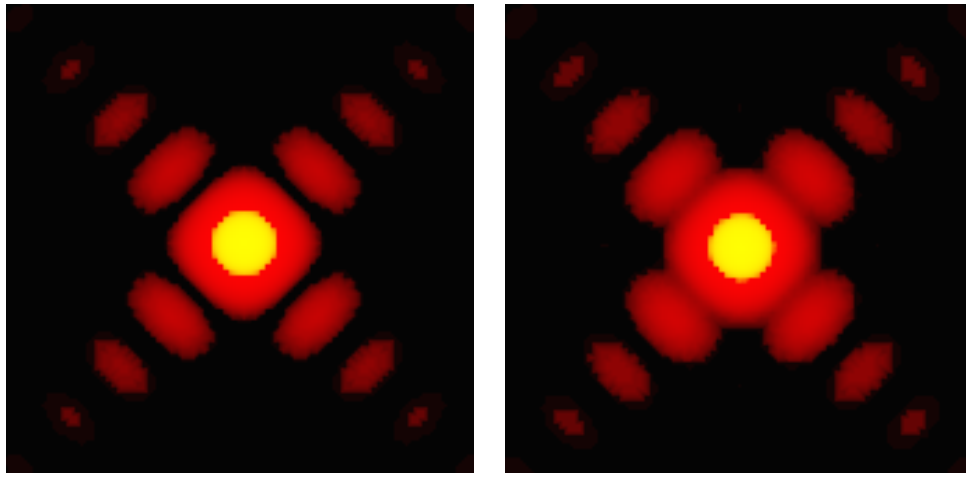
CXD from Silver Nanocubes



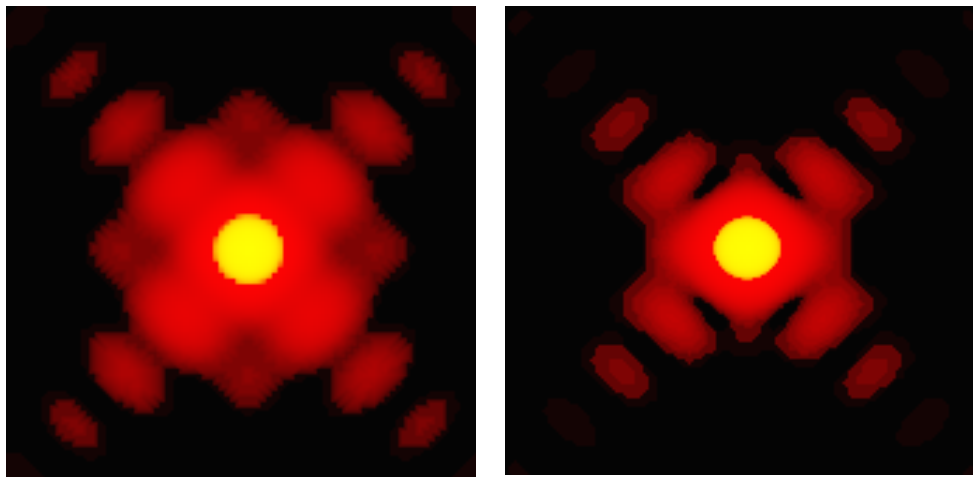


Yugang Sun and Younan Xia,
Science 298 2177 (2003)

Spherical and cylindrical waves



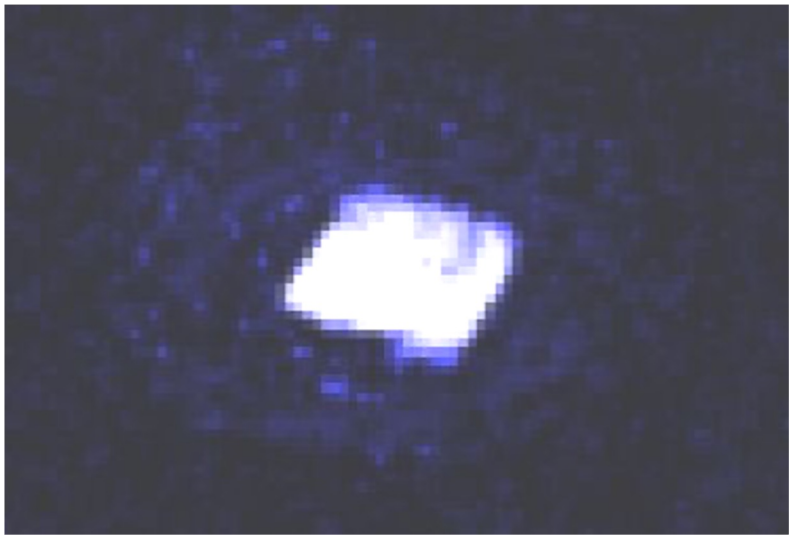
π/4



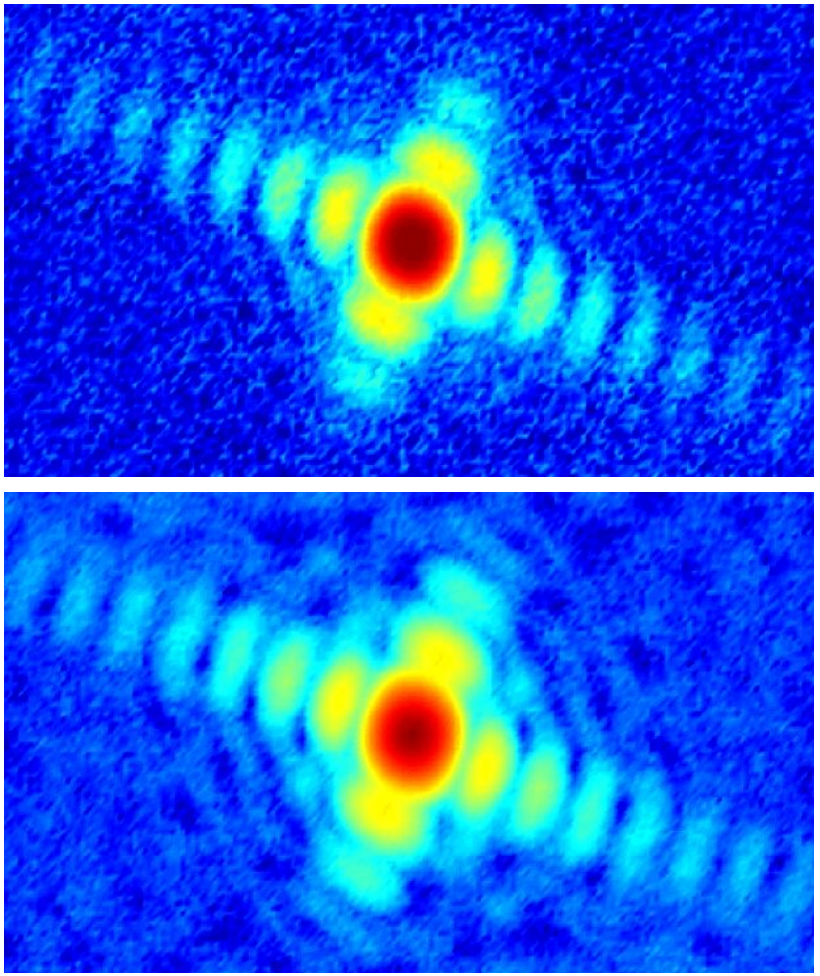
π/2

π/4 (x)

Reconstruction of Ag Nanocrystal

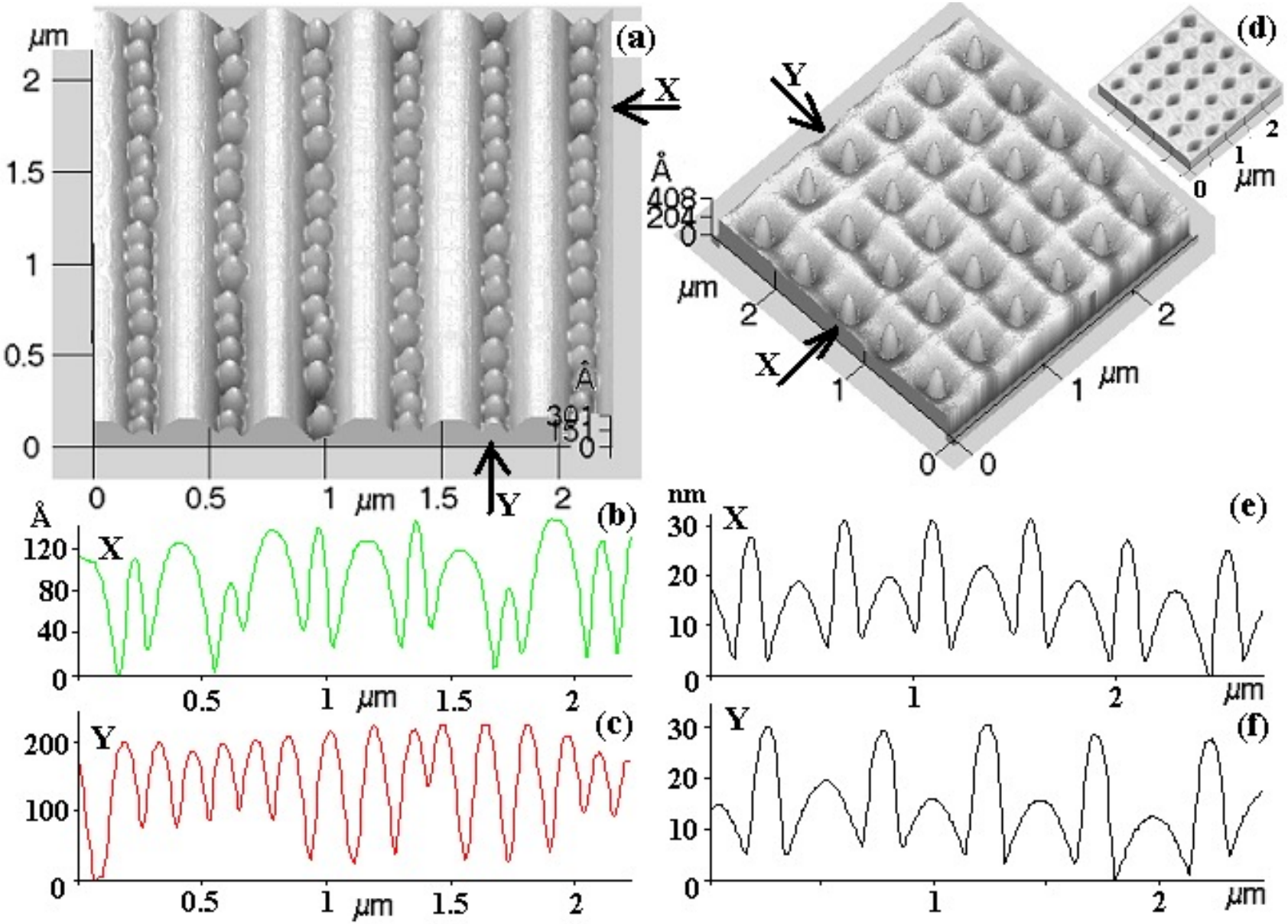


200nm



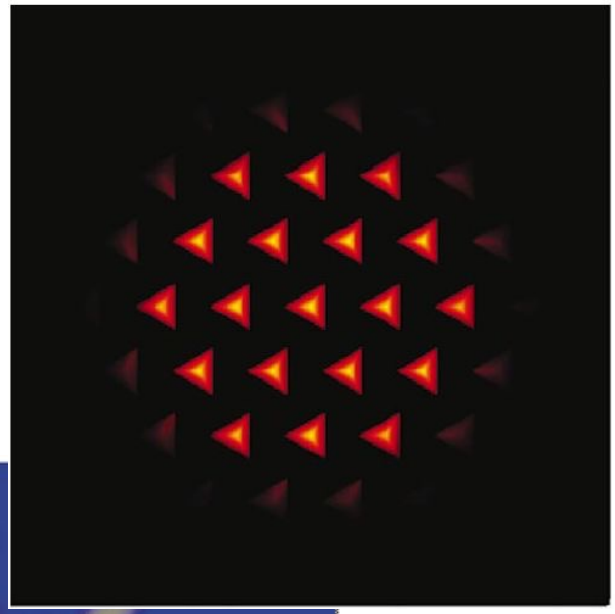
1D and 2D Quantum Dot Arrays

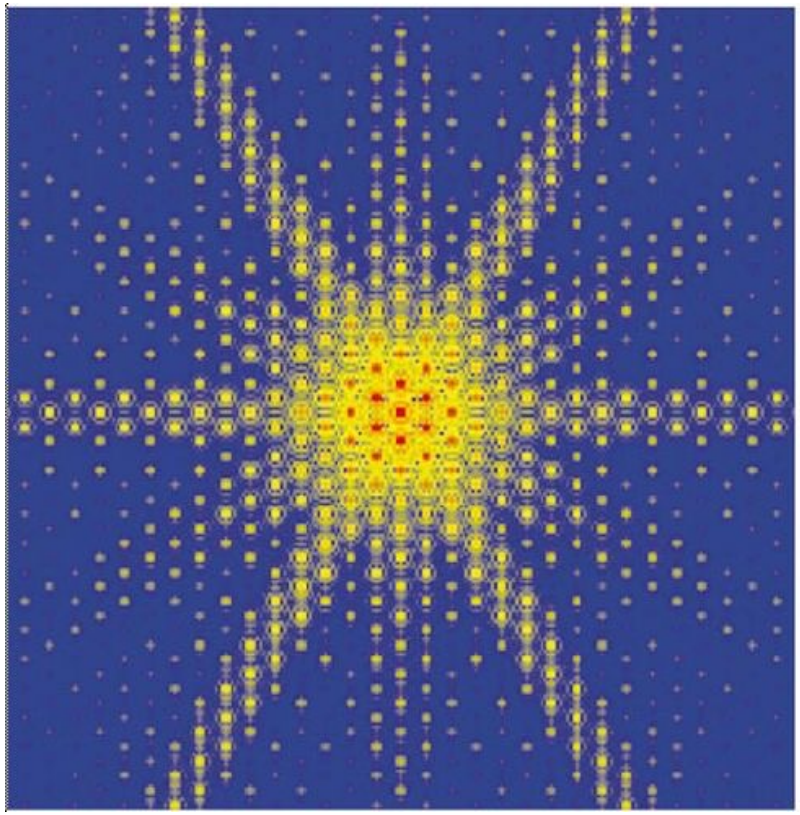
Zhenyang Zhong, G. Bauer, Johannes Kepler Universität Linz

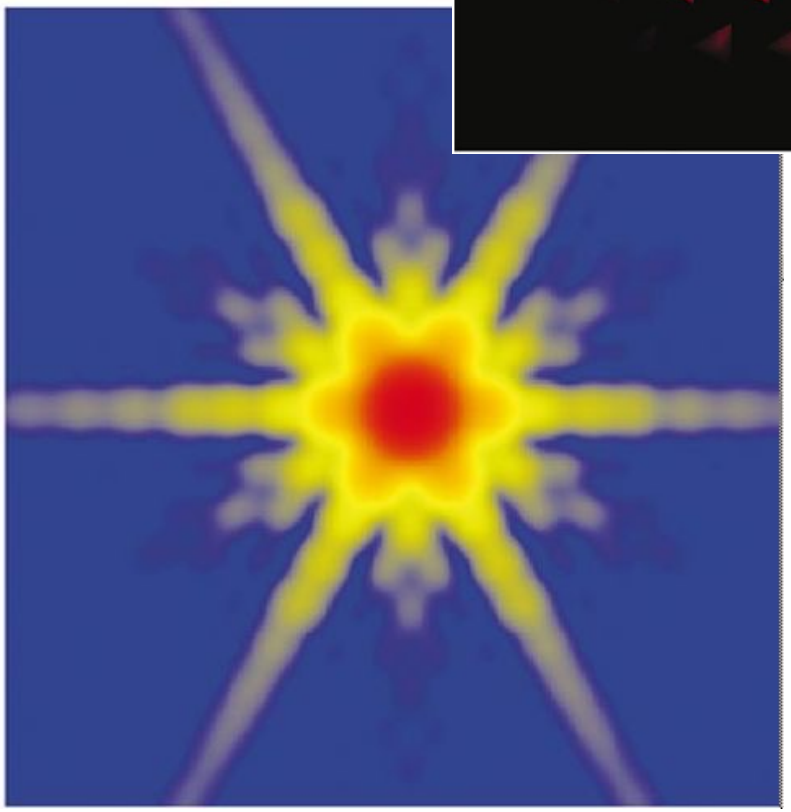


Vary Coherence (model)

I. A. Vartanyants and I. K. Robinson,
J. Sync. Rad. **10** 409 (2003)

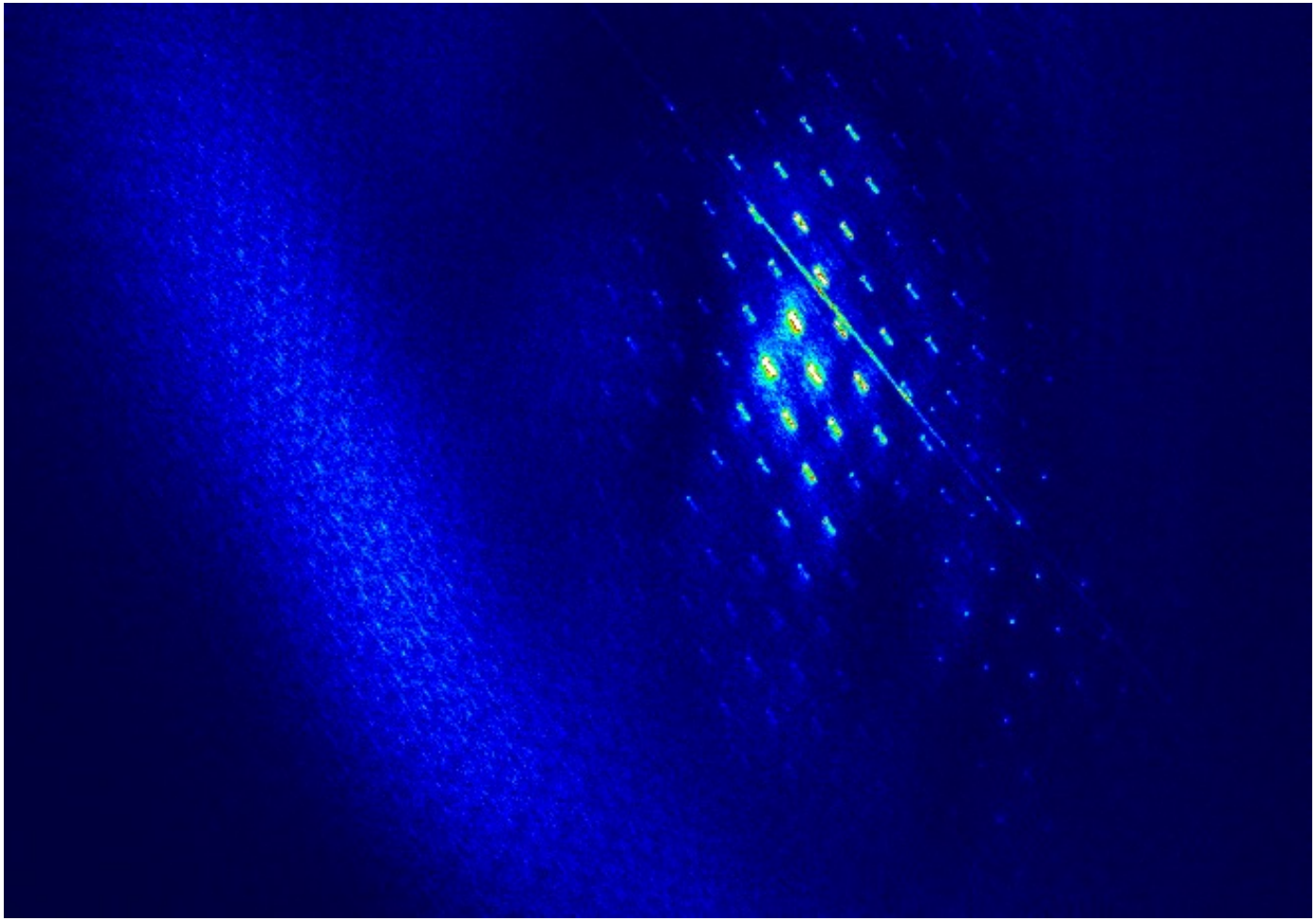




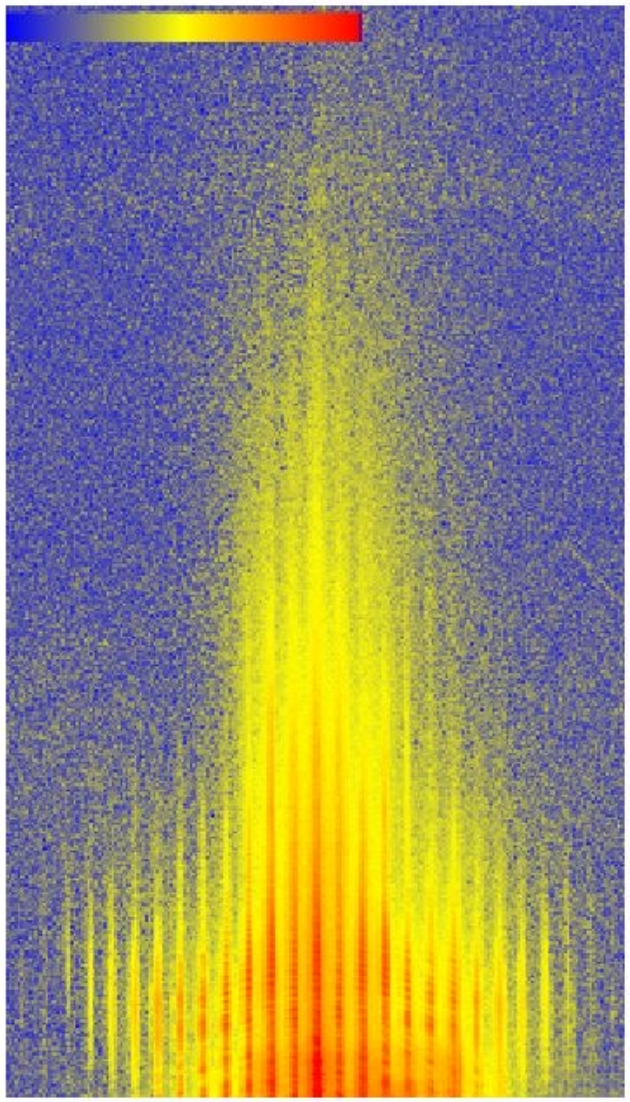


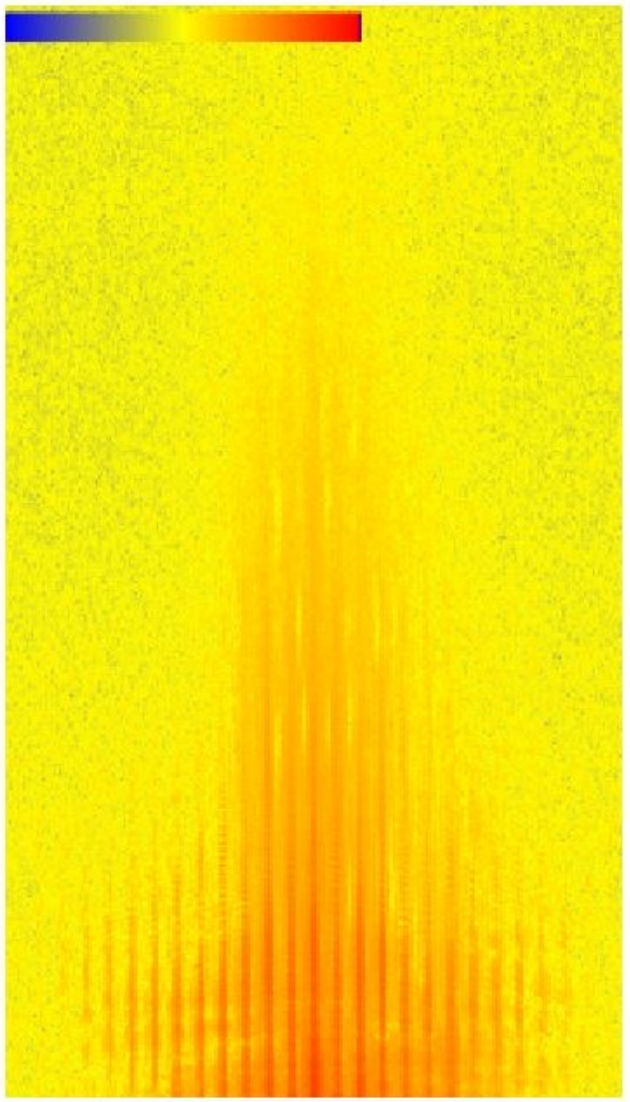
202 Diffraction from Ge QD array

with G. Bauer, Z. Zhong and T. H. Metzger



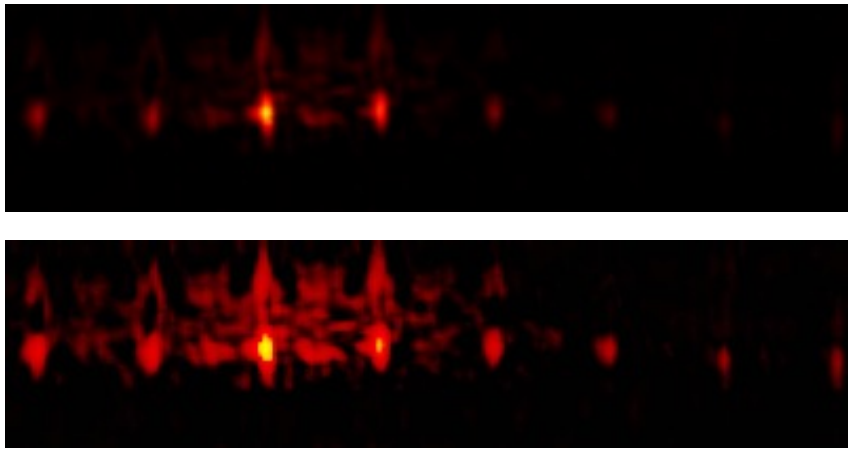
Result of fitting GISAXS data

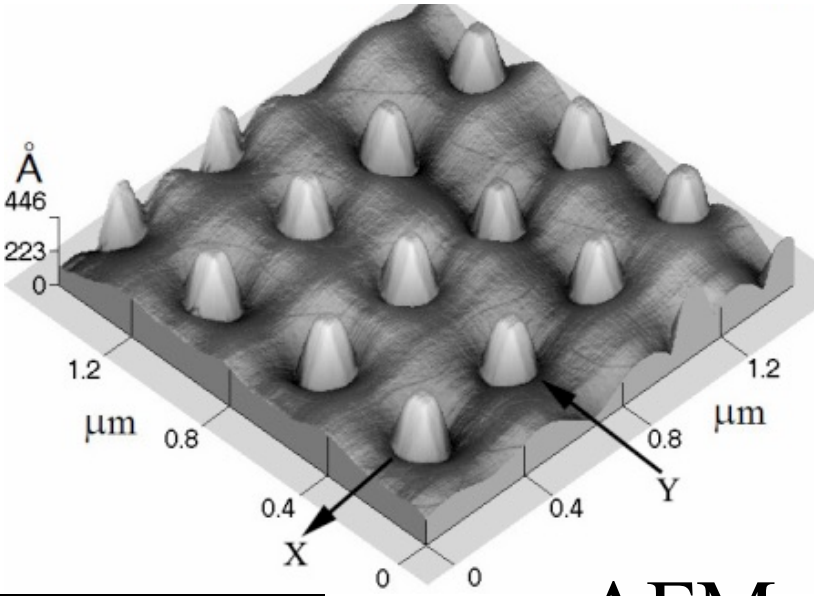




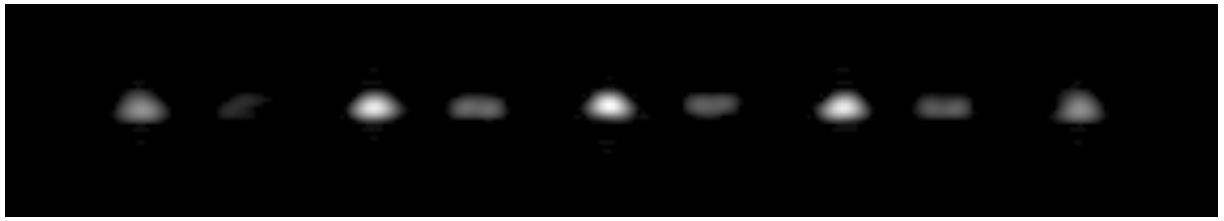
Images from Fitting GISAXS

with Jeff Onken



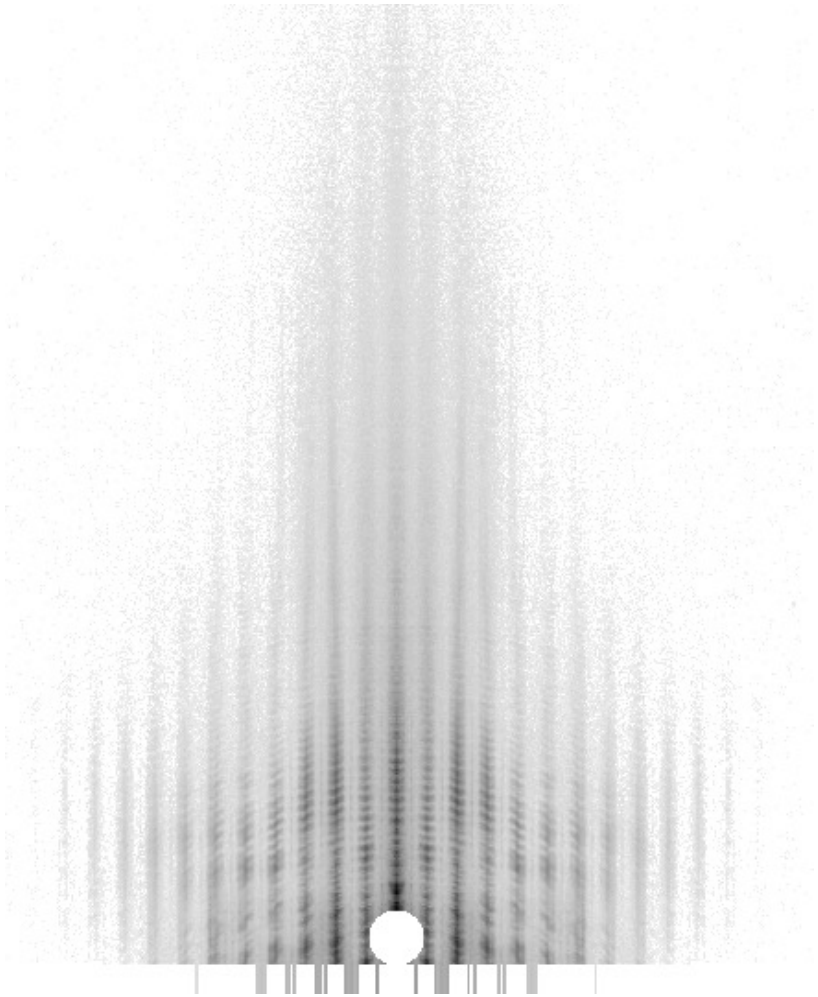


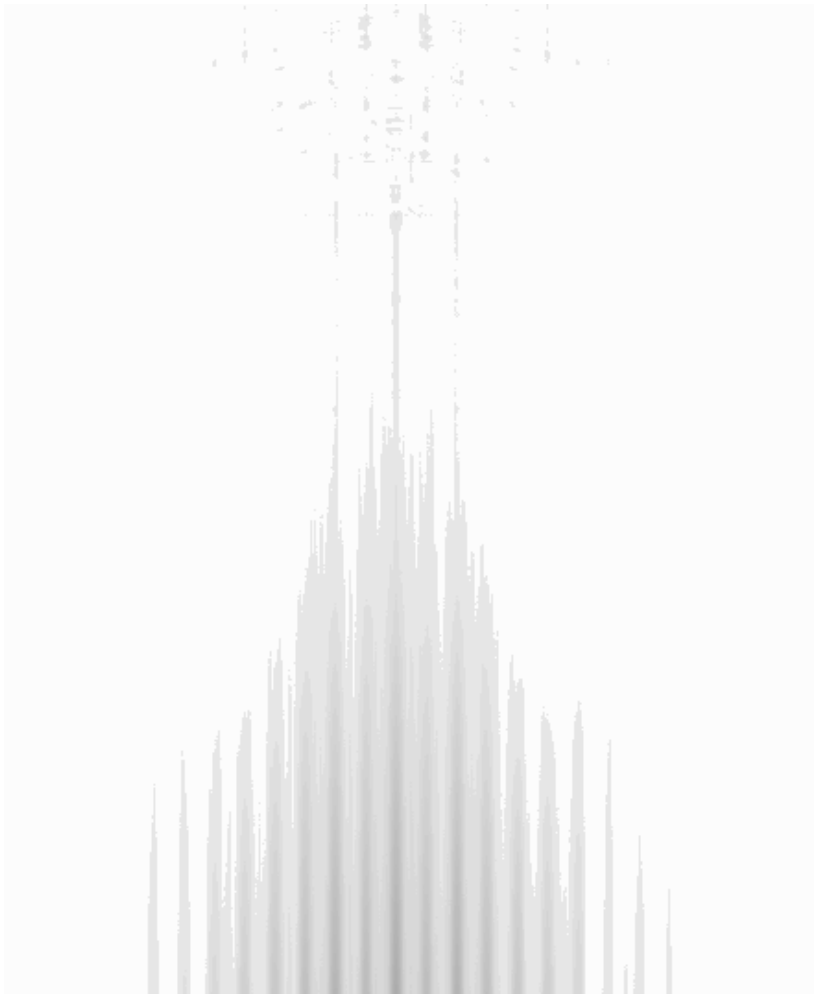
AFM



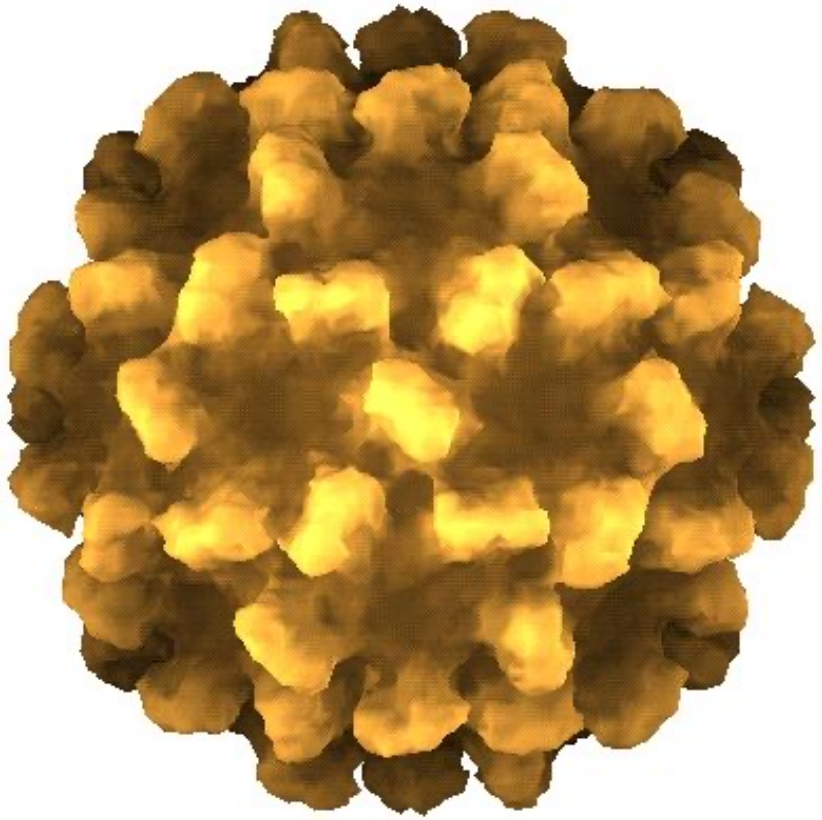
Mask used as support

Diffraction pattern under constraint





Tomato Bushy Stunt Virus
1980



Conclusions and Outlook

- Direct inversion of CTRs for heterostructures
- Inversion of CXD demonstrated also
- Internal structure of Au Nanocrystals
- Preservation of coherence upon focussing
- Simple Quantum Dot stuctures possible now
- Single molecules one day