The original CTR beamline: X16A

Ian Robinson
Brookhaven National Lab
University College London

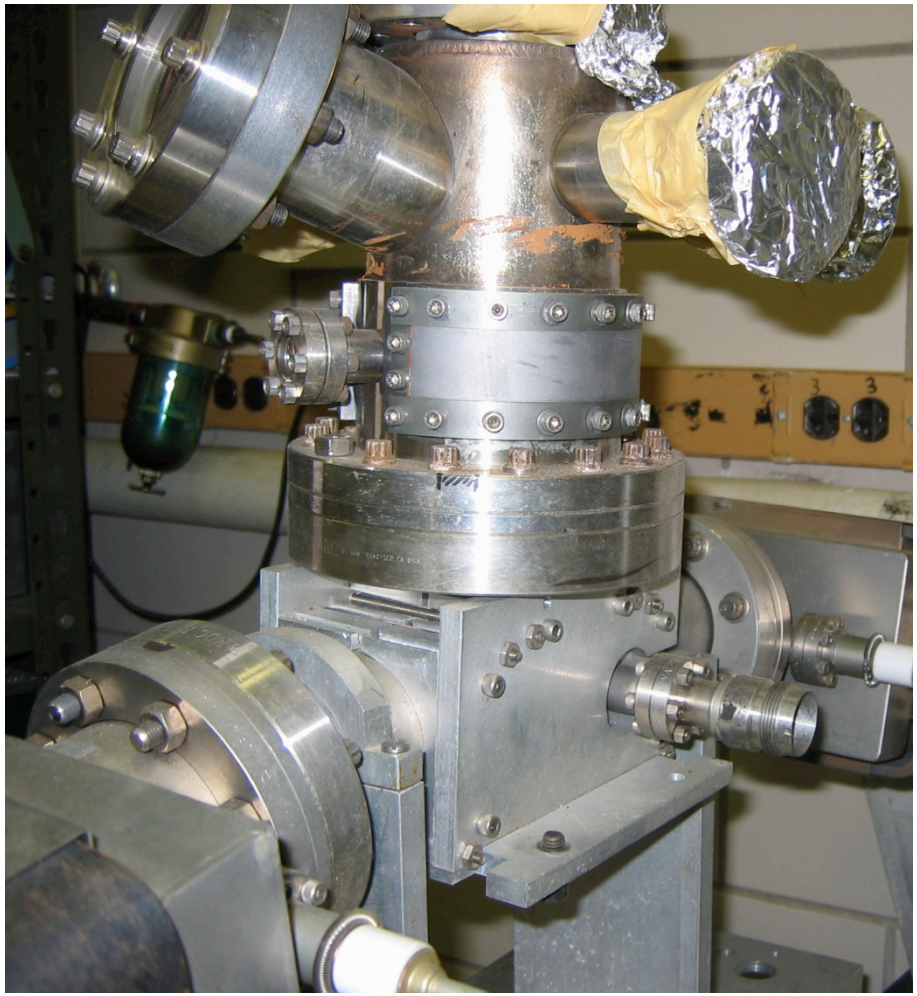
30 years of surface X-ray scattering at the NSLS

SXNS-14, Stony Brook

Generations of Synchrotron

- 2nd: Storage Ring Dedicated to X-rays
  - NSLS Surface X-ray Diffraction
- 3rd: Undulators in "Straight Sections"
  - APS Coherent X-ray Diffraction
  - Diamond Light Source
- X-ray Free Electron Lasers
  - LCLS Ultrafast Coherent Diffraction

First UHV Experiments (1981)



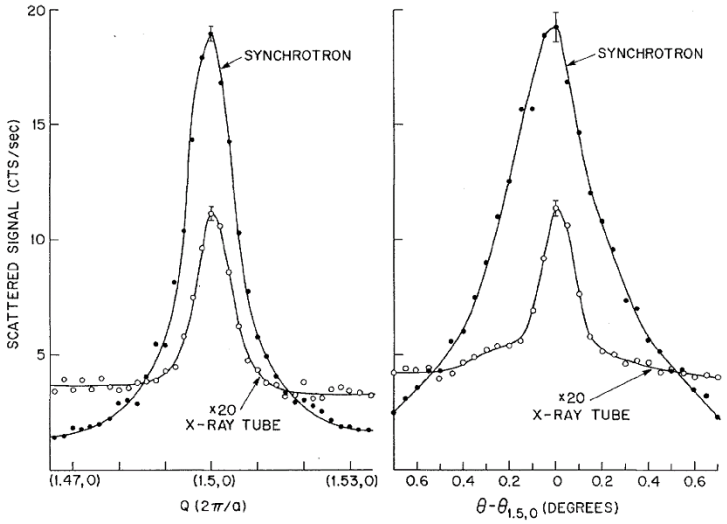
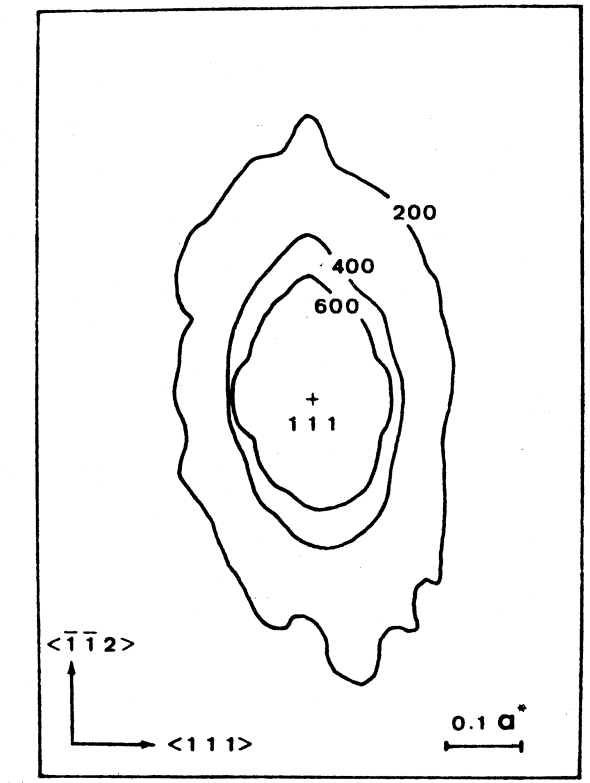


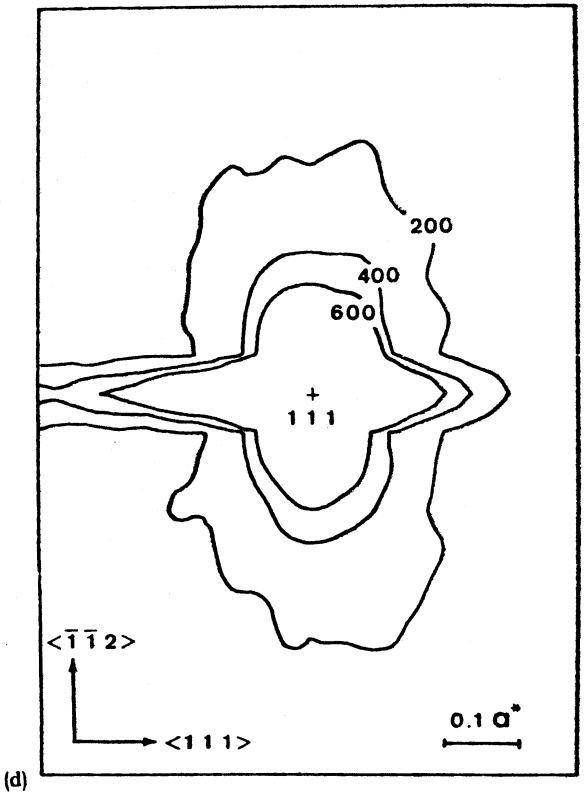
FIG. 1. A plot of the $(\frac{3}{2}, 0)$ Bragg reflection as a function of the momentum transfer $Q(2\pi/a)$ and the crystal's mosaic spread (deg).

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

Diffuse Scattering from Si Wafer



Unpolished wafer



40 microns removed

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

Surfaces in Dynamical Diffraction

A. M. Afanas'ev *et. al.*, Acta Cryst **A40** 352 (1984)

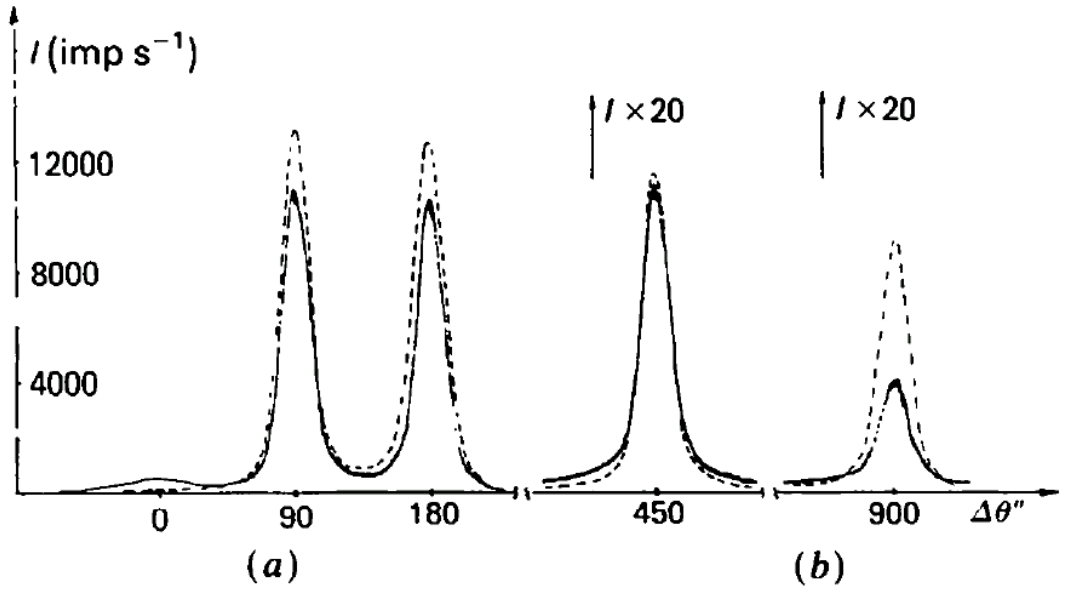


Fig. 2. TDC spectra from Ge(111) crystals, $(n, -n, n)$ arrangement, symmetric Bragg diffraction for (a) $\alpha = 90''$, (b) $\alpha = 450''$. Dashed line for ideal crystal, solid line for crystal first polished with diamond paste and then partially etched.

Effect of etching seen in triple-axis measurement of rocking curve

Scattering of X-rays From Crystal Surfaces

S.R.Andrews & R.A.Cowley JPCM **18** 6427 (1985)

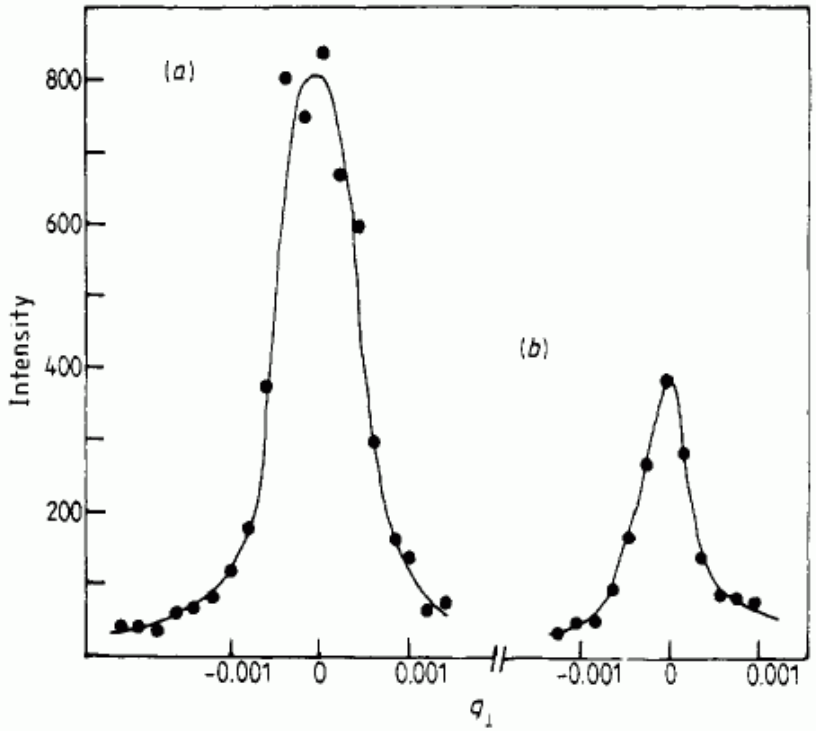
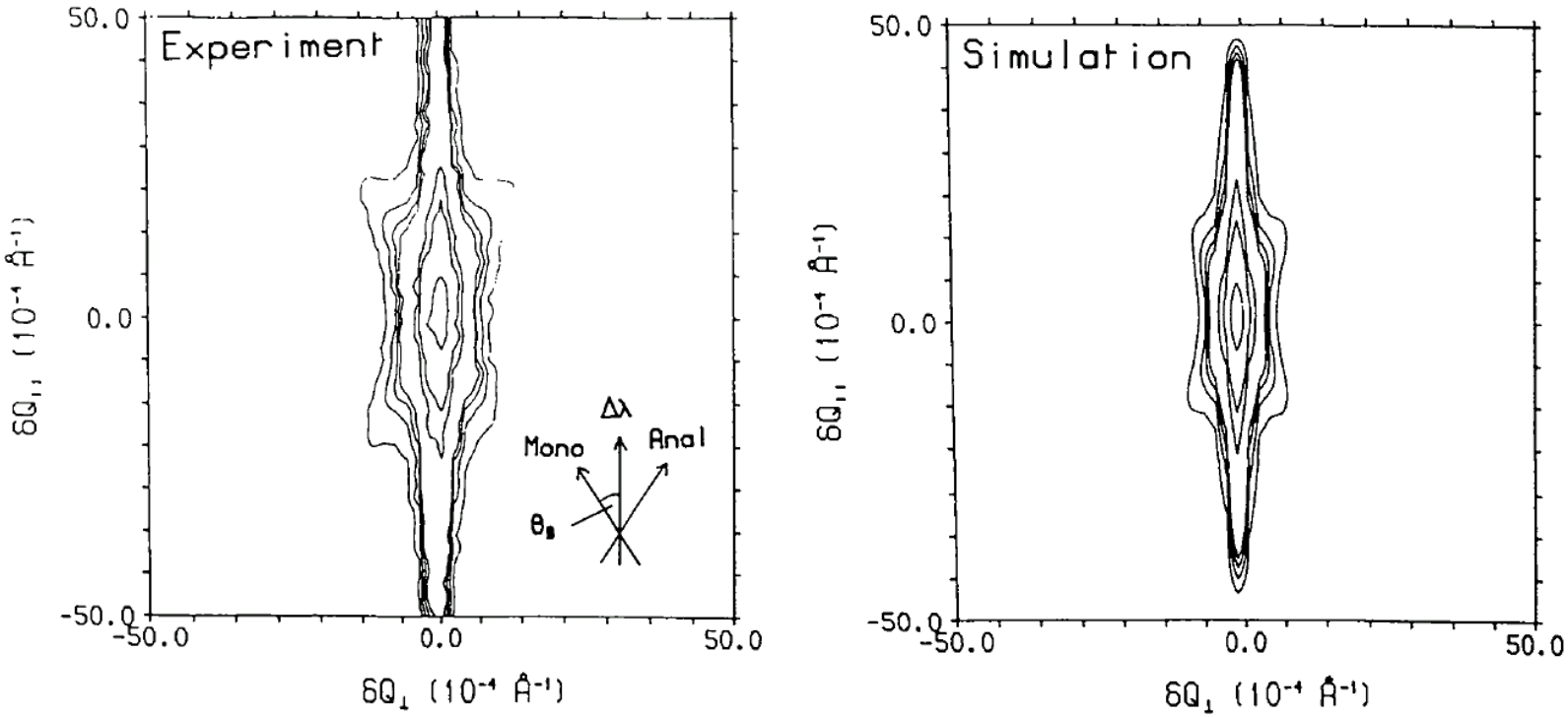


Figure 1. The intensity of scattering, as a function of q , (*a*) for $\boldsymbol{Q}$ = (0, 0, 4.005) (vertical scale in counts per 2 s) and (*b*) $\boldsymbol{Q}$ = (0, 0, 4.025) (vertical scale in counts per 300 s) in the GaAs sample with an (001) surface corresponding to figure 2, curve B.

Resolution Function of the Triple-Axis Spectrometer

R. A. Cowley *et. al.*, Acta Cryst **A45** 415 (1989)



Diffraction as a Surface Integral

Die äußere Form der Kristalle
in ihrem Einfluß auf die Interferenzerscheinungen
an Raumgittern
Von M. v. Laue

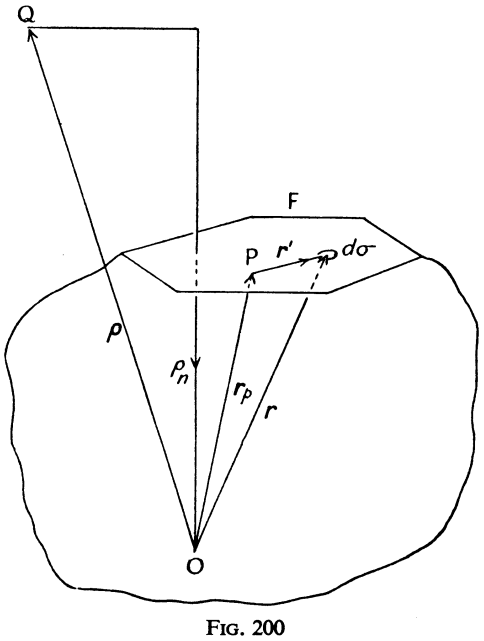
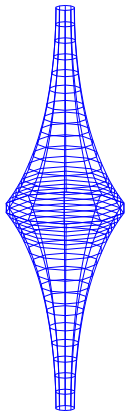


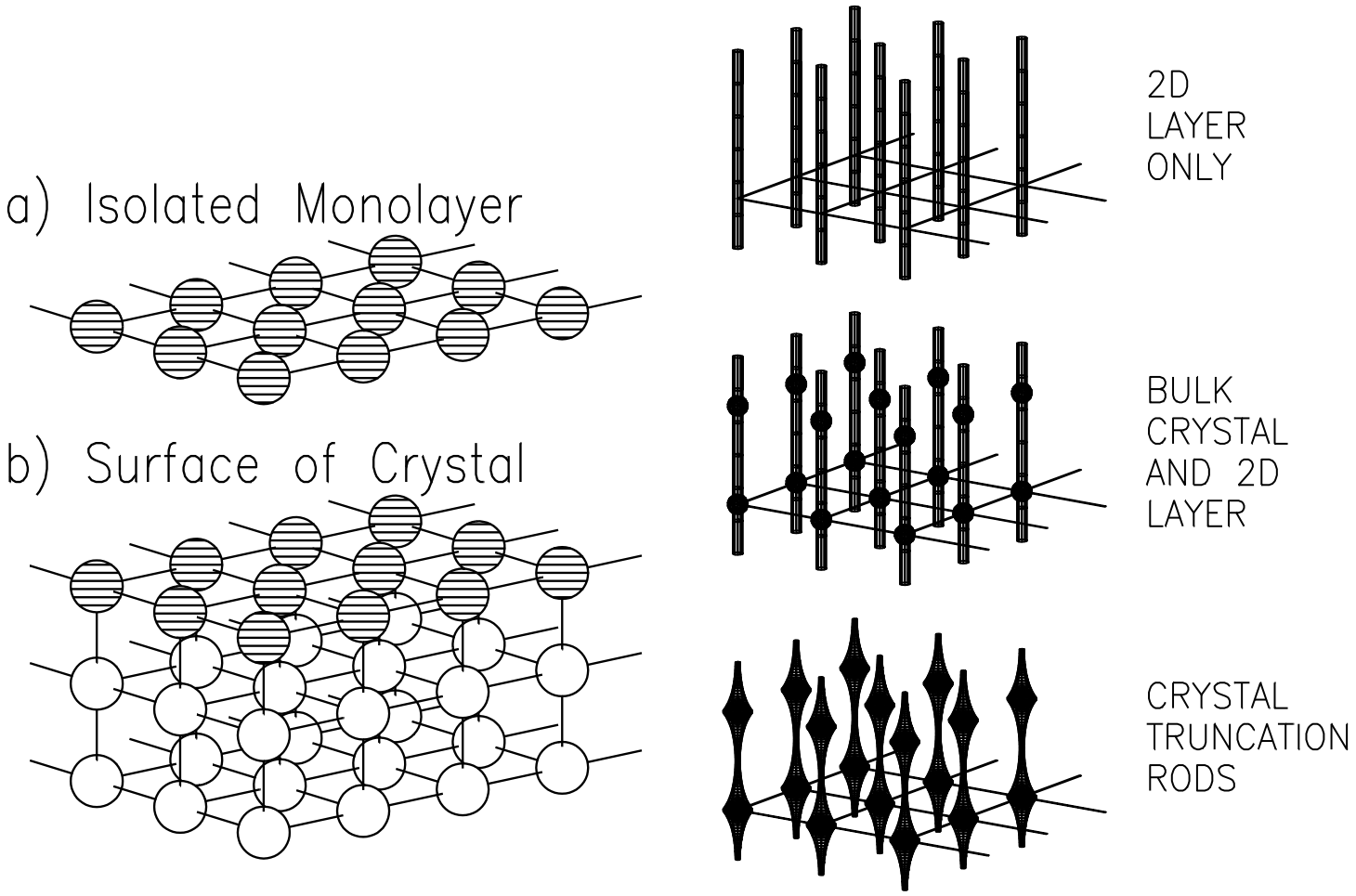
FIG. 200

Annalen der Physik [5] 26 55 (1936)

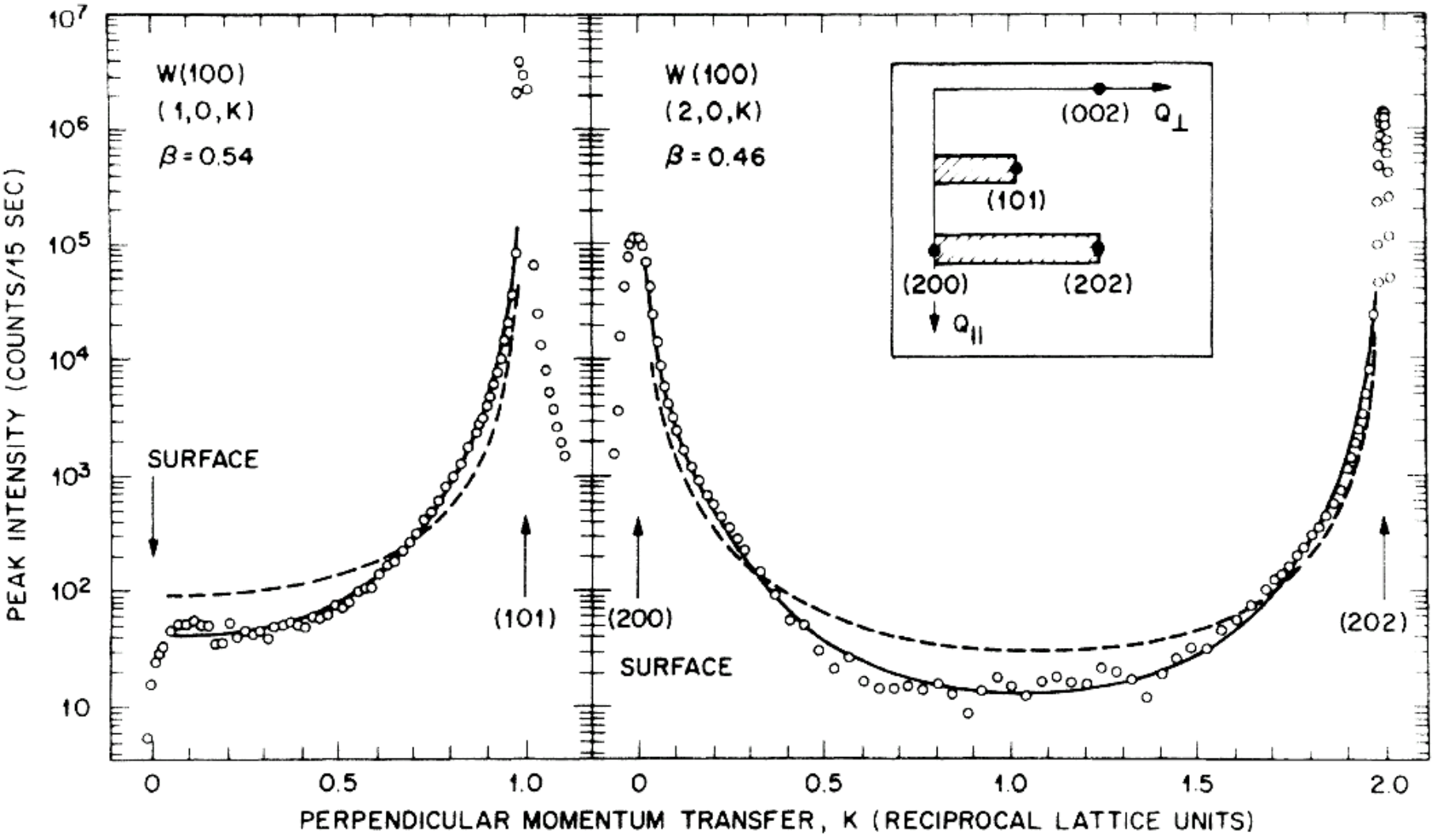


"Stacheln"

Crystal Truncation Rods (1986)

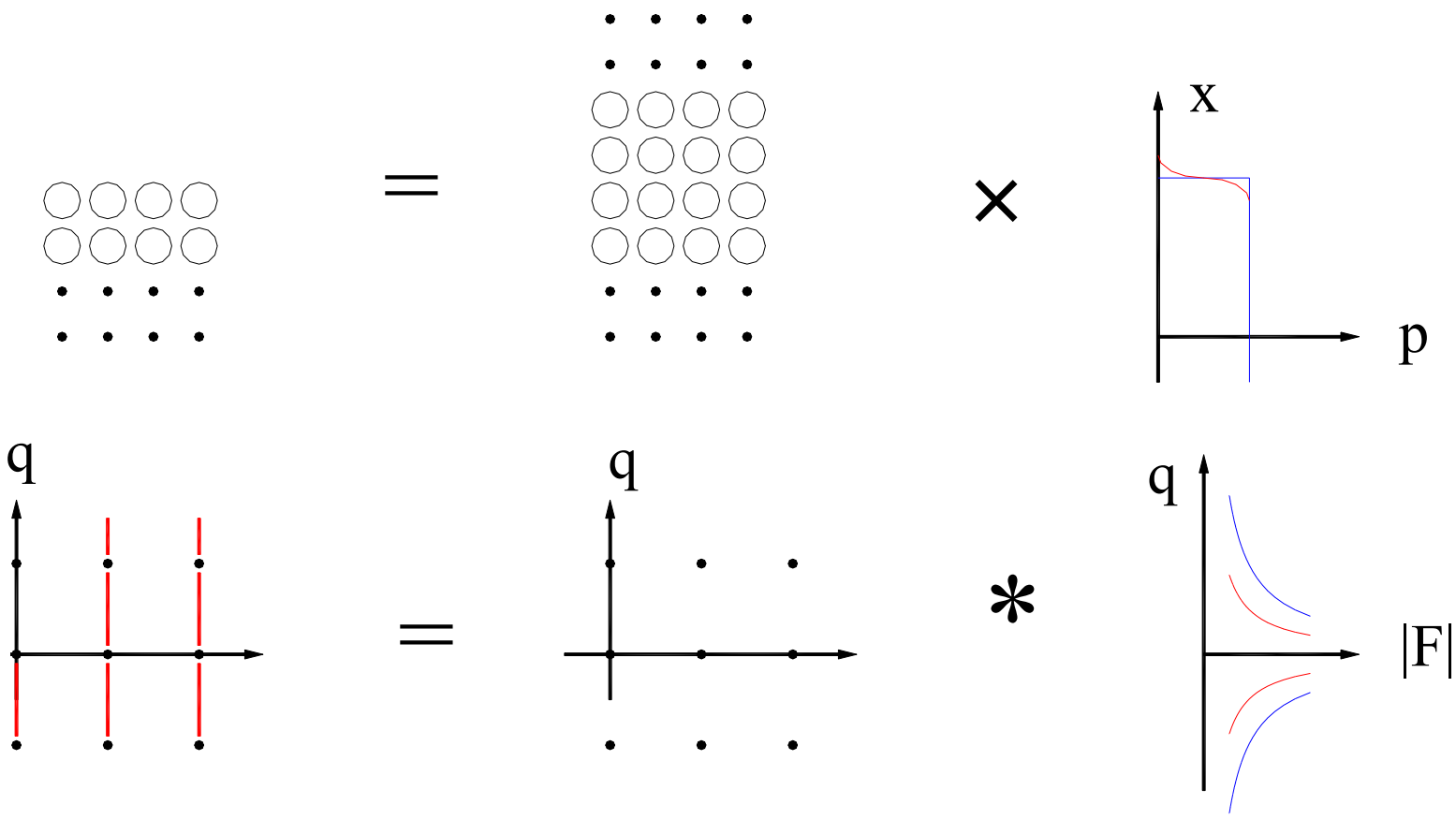


CRYSTAL TRUNCATION RODS AND SURFACE ROUGHNESS

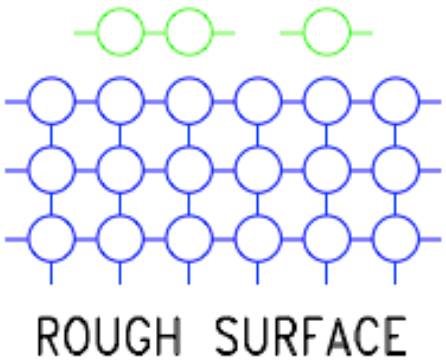


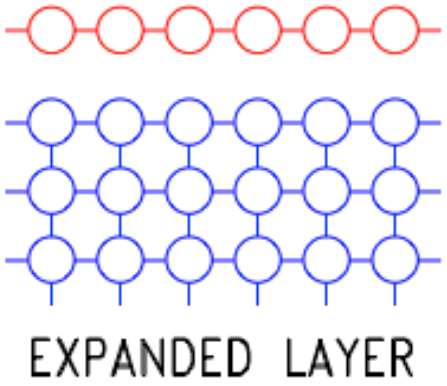
I. K. Robinson SXNS-14 2016

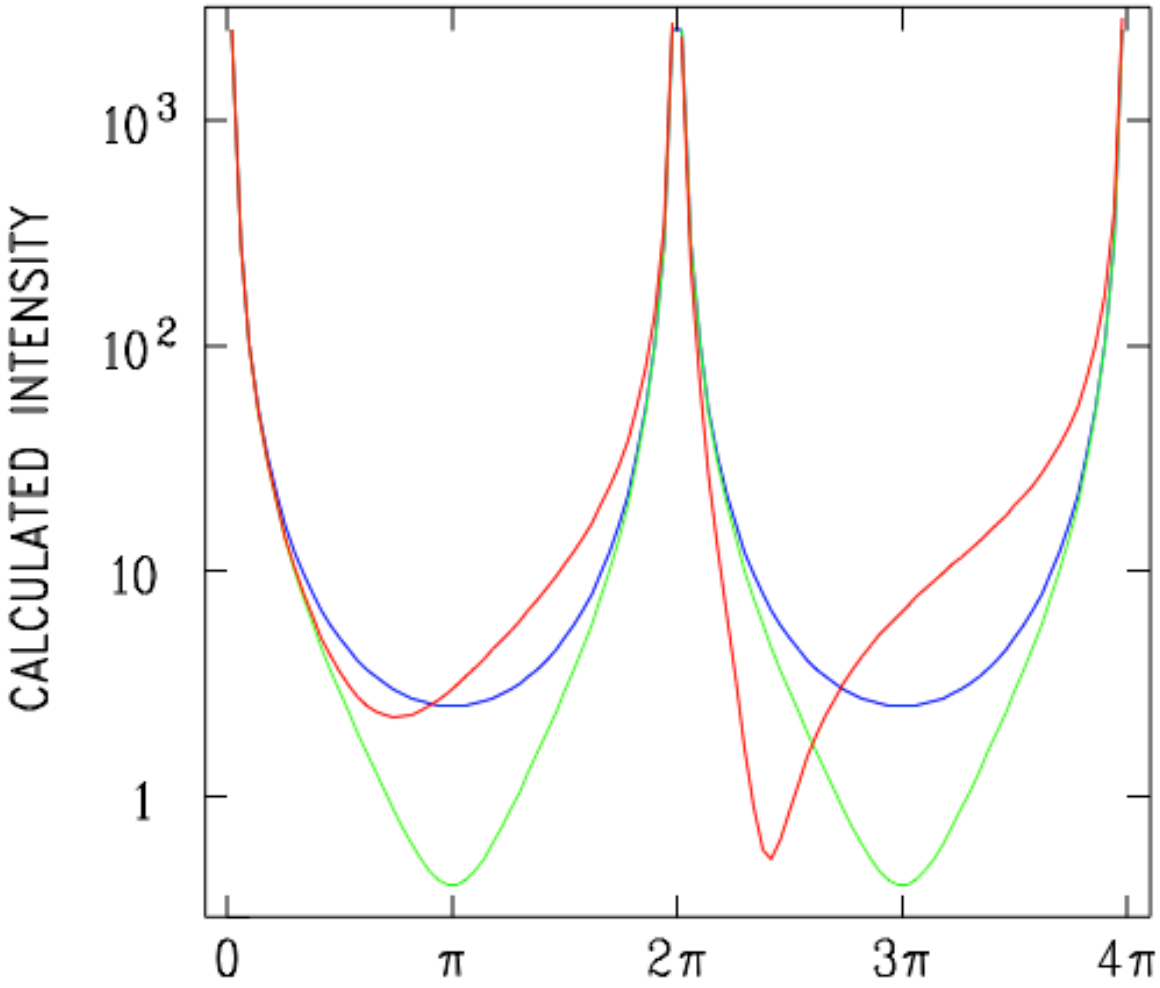
CTR as Convolution



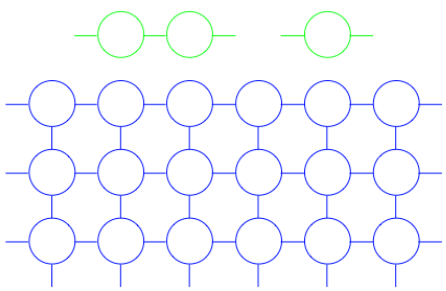
CTR is Sensitive to Surface Structure

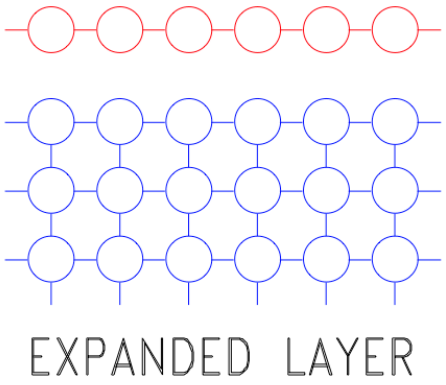


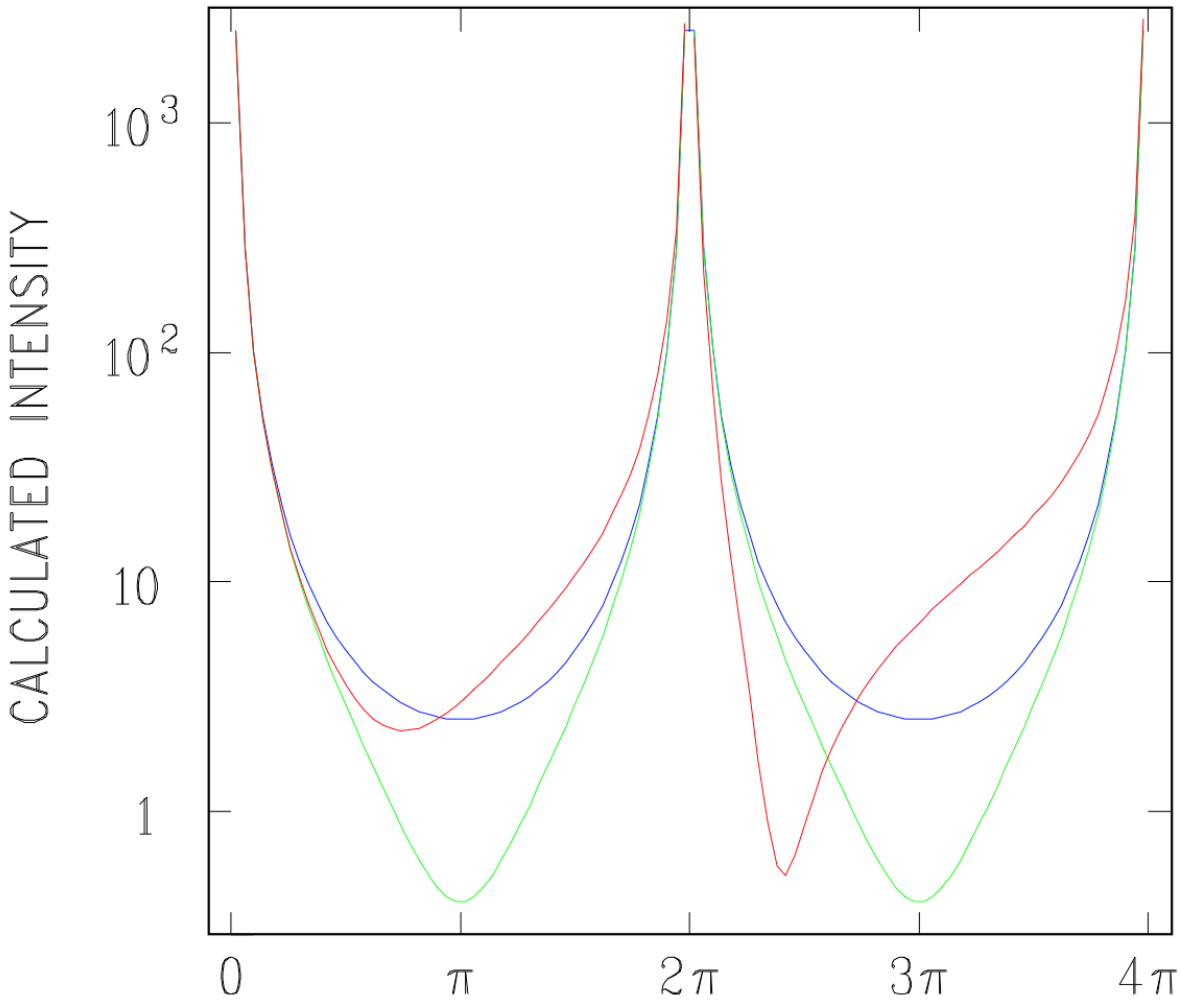




CTR is Sensitive to Surface Structure

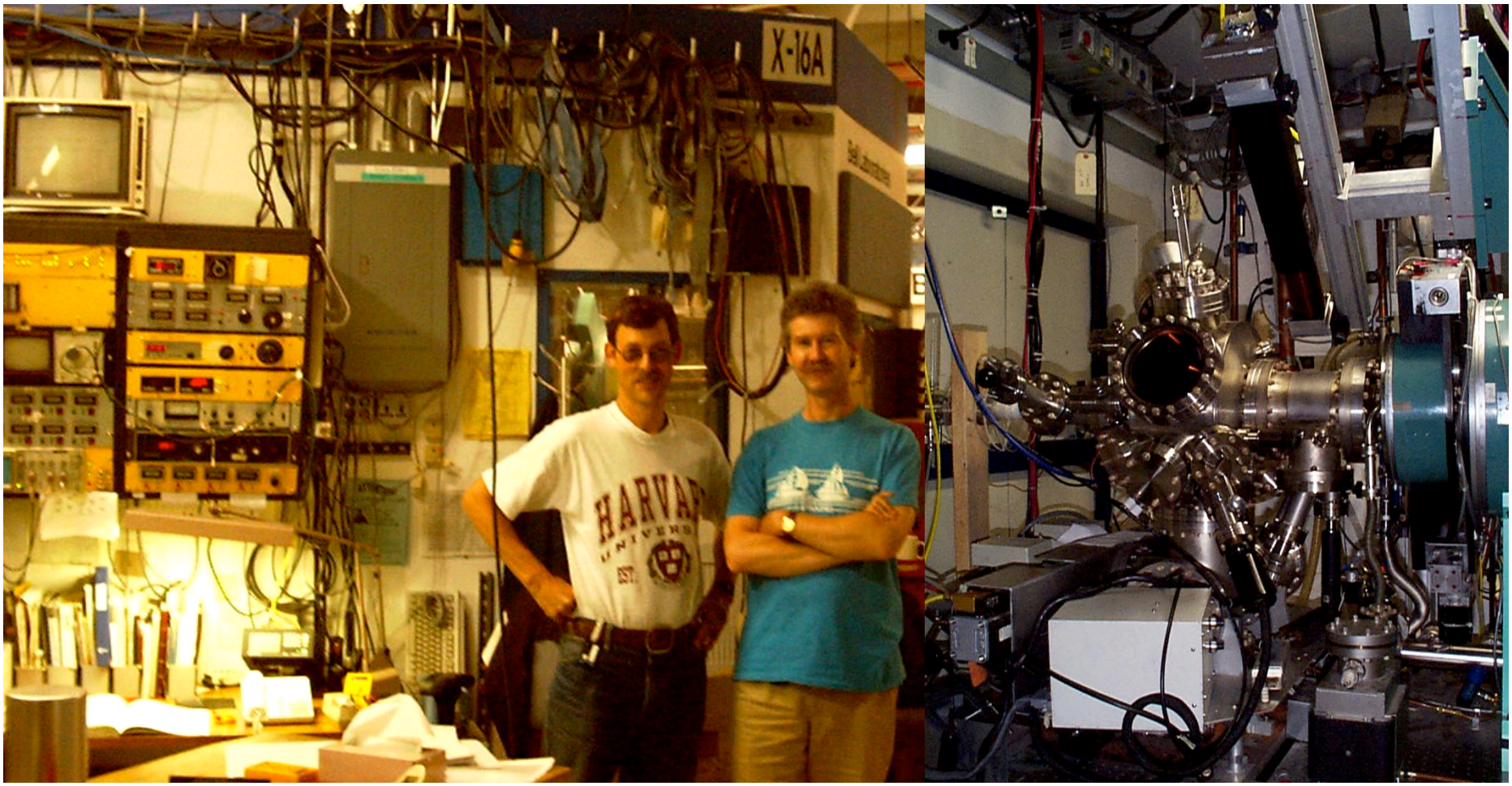


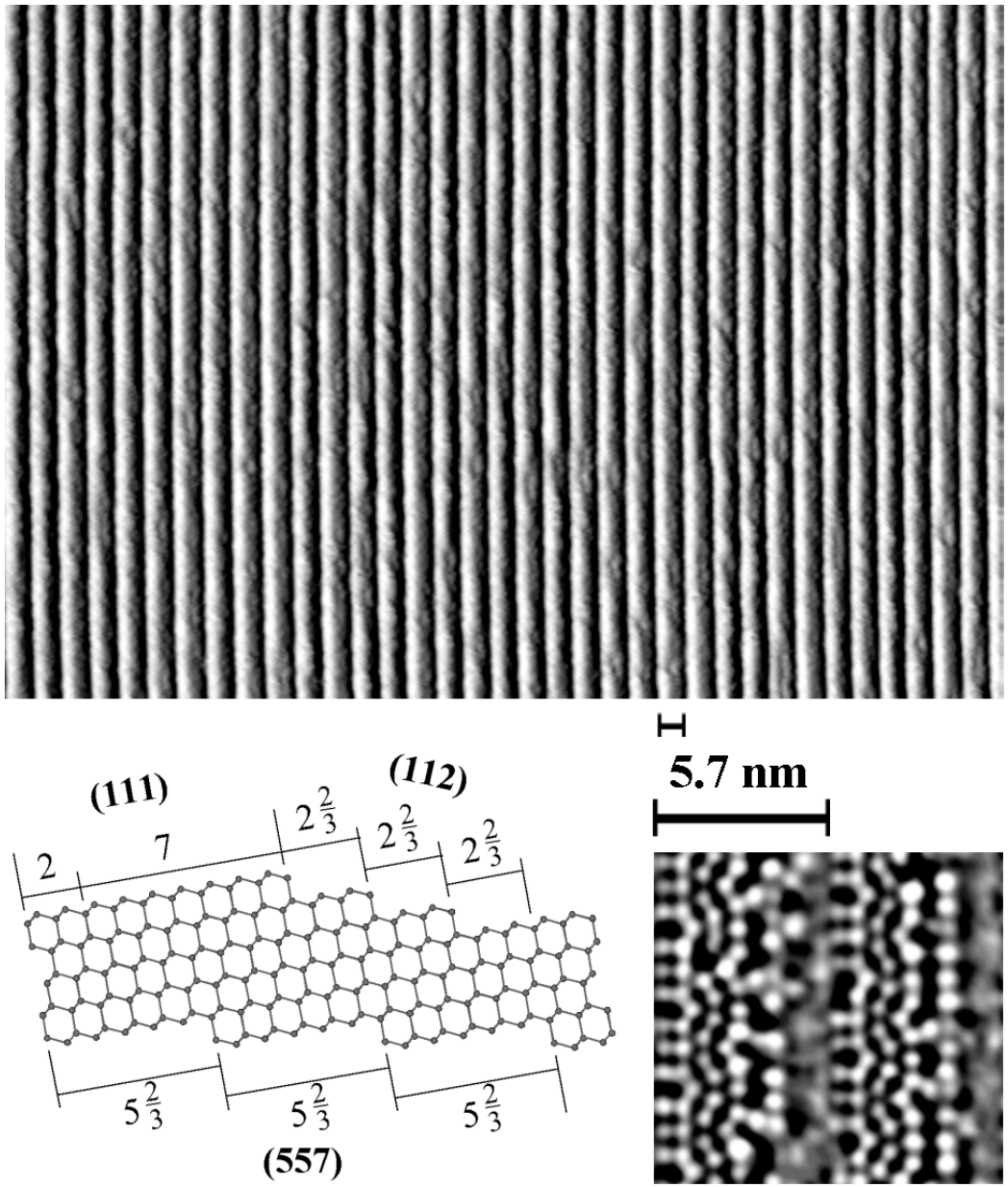




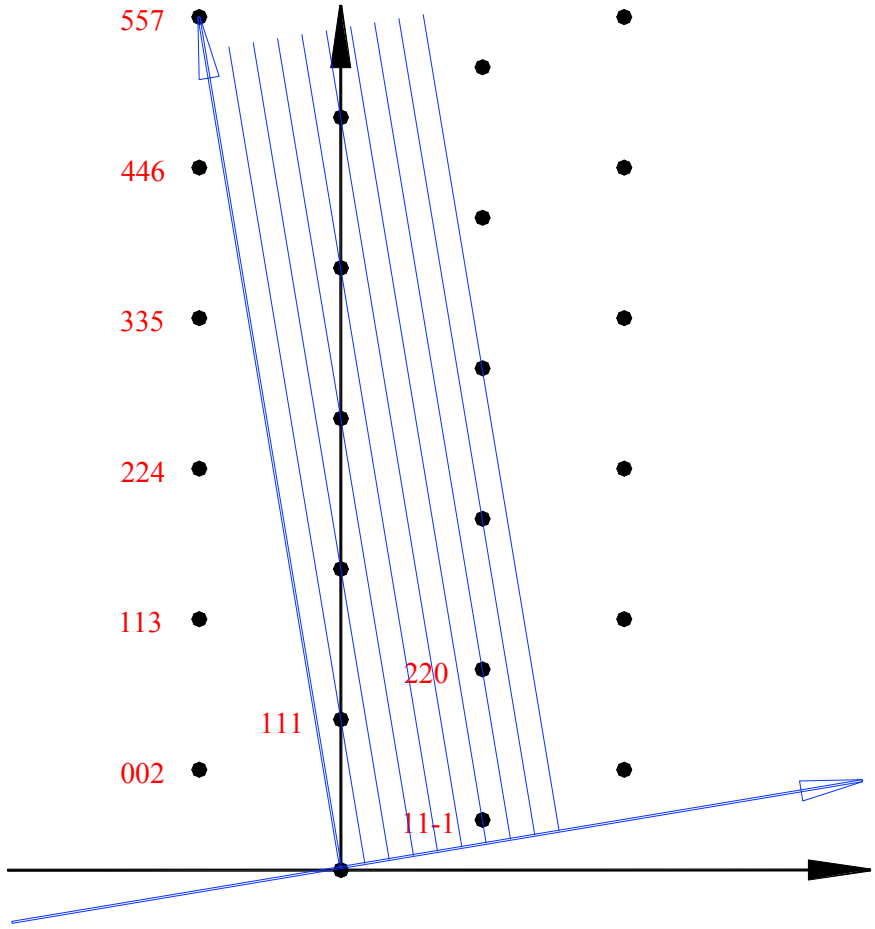
X16A Surface X-ray Diffraction

Brookhaven National Lab (1987 – 2004)

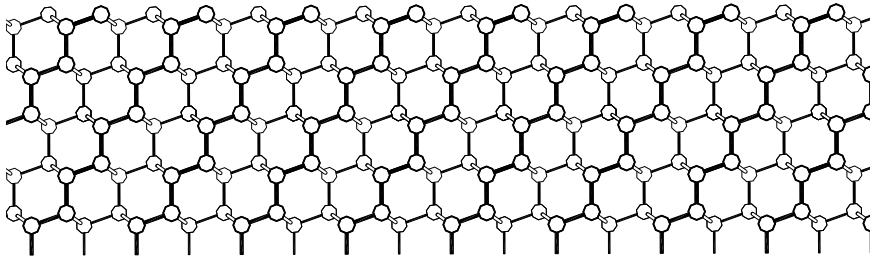


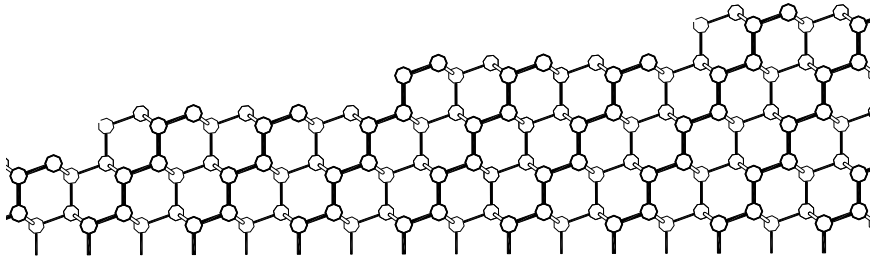

5.7 nm
(111)
(112)
2
7
$2\frac{2}{3}$
$2\frac{2}{3}$
$2\frac{2}{3}$
$5\frac{2}{3}$
$5\frac{2}{3}$
$5\frac{2}{3}$
(557)

Crystallography of Stepped Surfaces



Silicon (111)

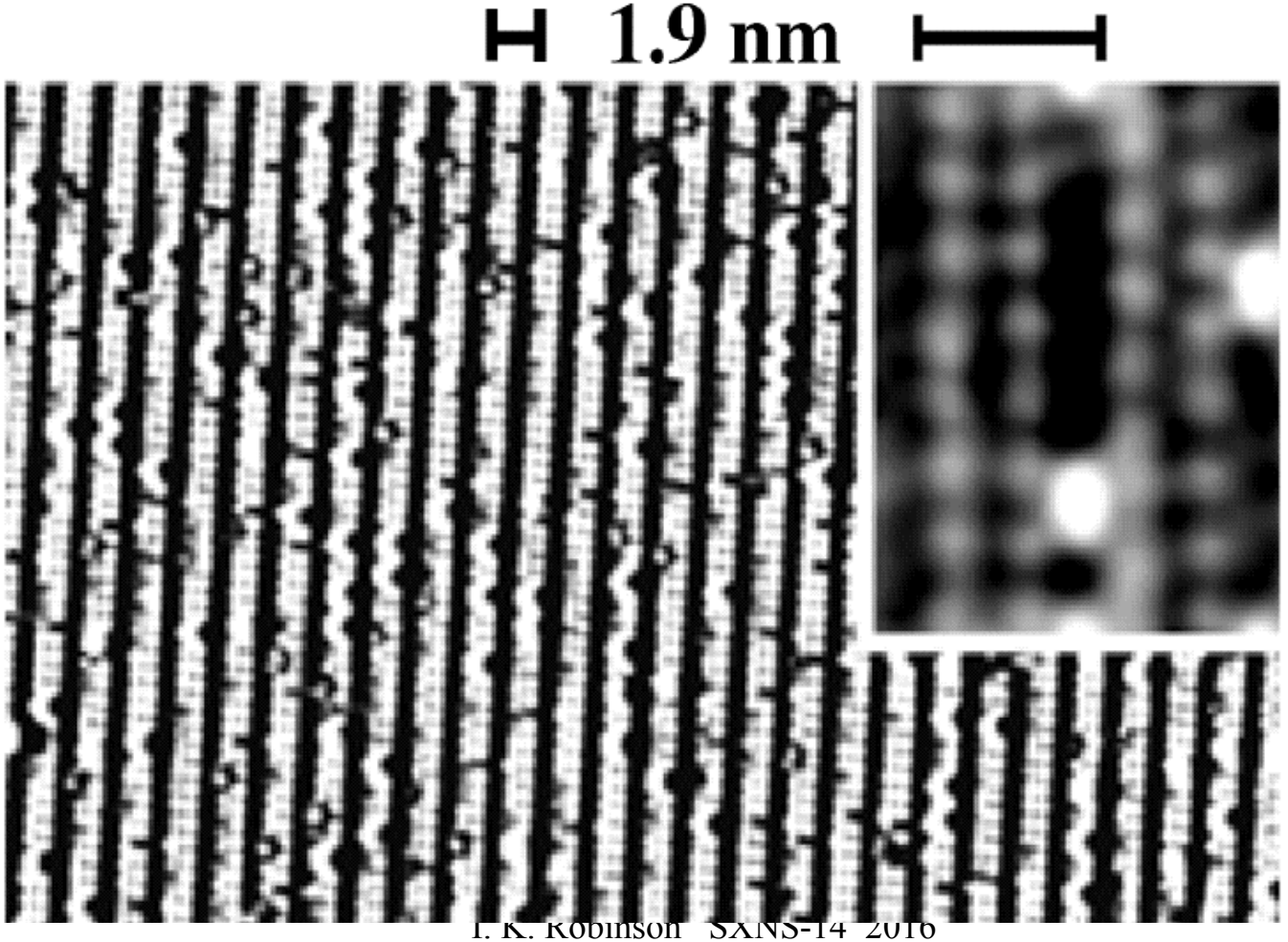


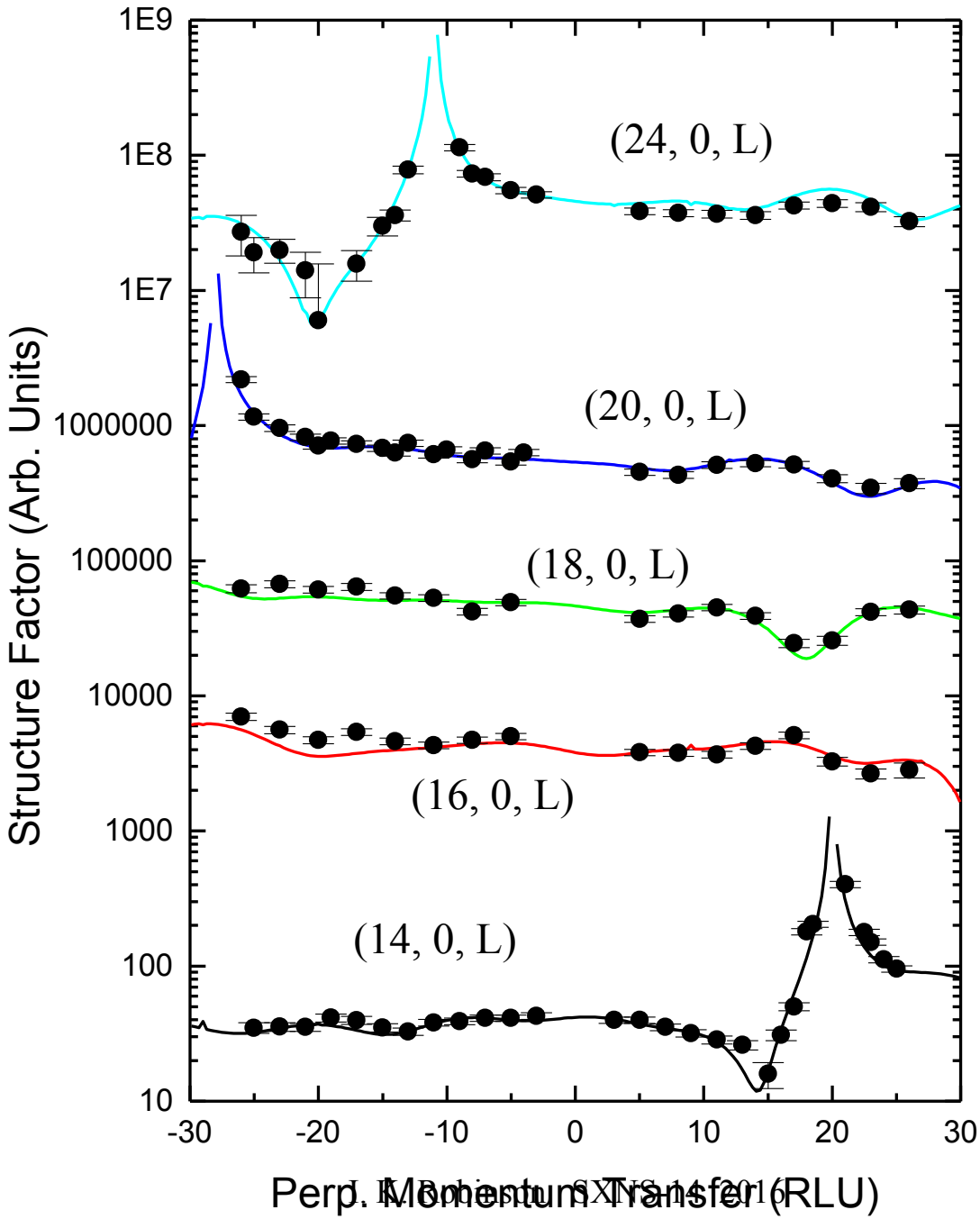


Si(557) surface

Si(557) coated with Au

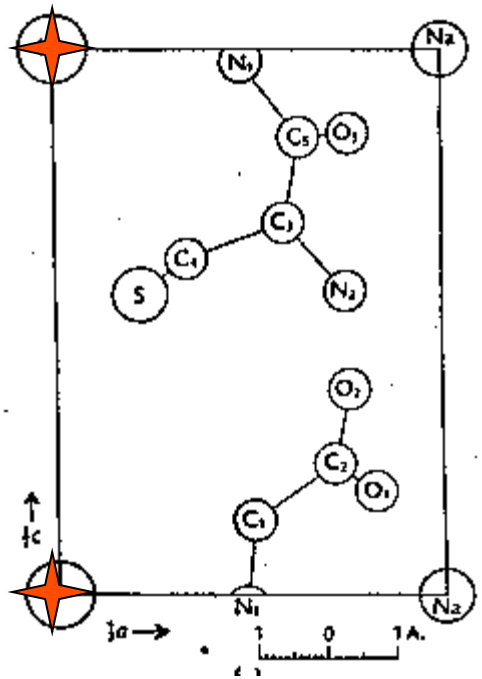
R. Losio, et. al., Phys. Rev. Lett. 86 4632 (2001)



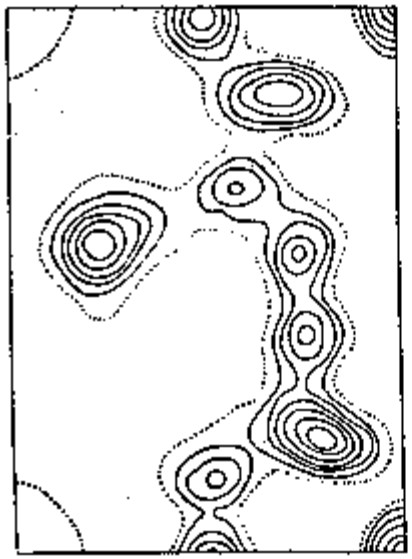

1E9
1E8
1E7
1000000
100000
10000
1000
100
10
-30
-20
-10
0
10
20
30
(24, 0, L)
(20, 0, L)
(18, 0, L)
(16, 0, L)
(14, 0, L)
Structure Factor (Arb. Units)
Perp. Momentum Transfer (RLU)

Phasing by a Single Heavy Atom

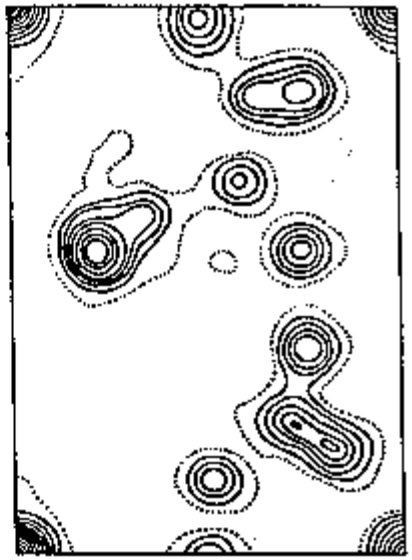
H. B. Dyer, Acta Cryst. 4 42 (1951)



Cysteinylglycine sodium iodide

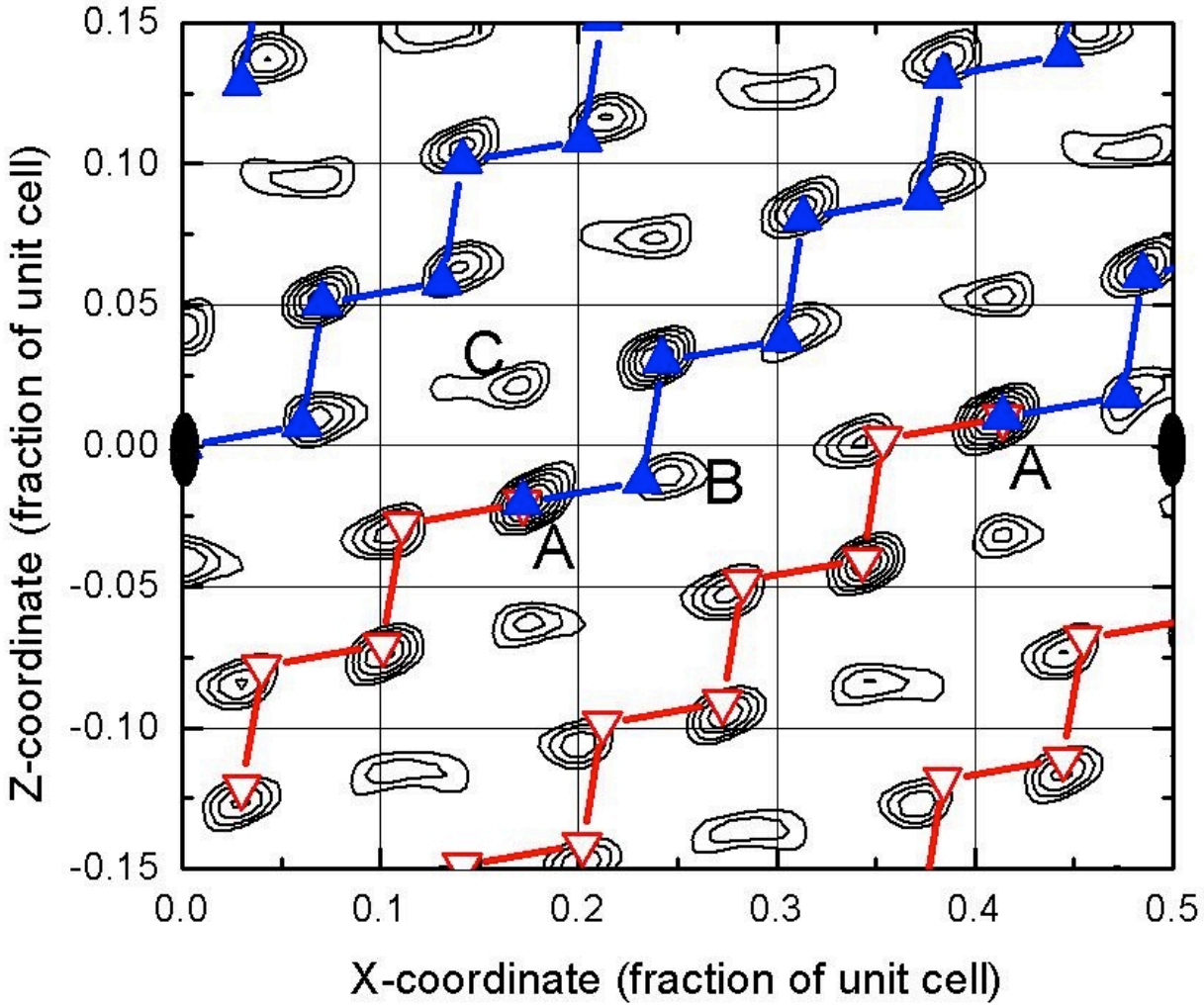


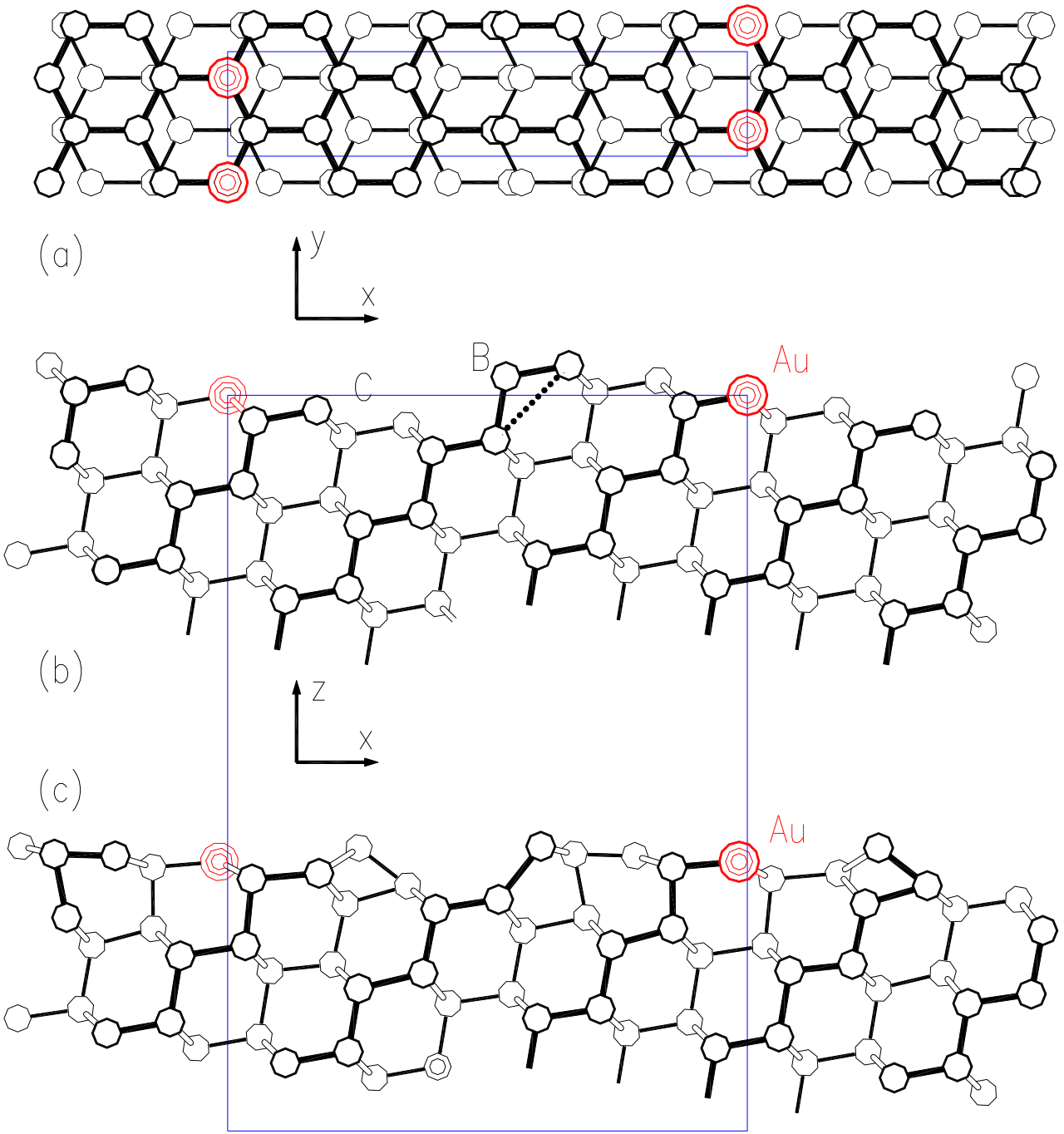
Patterson



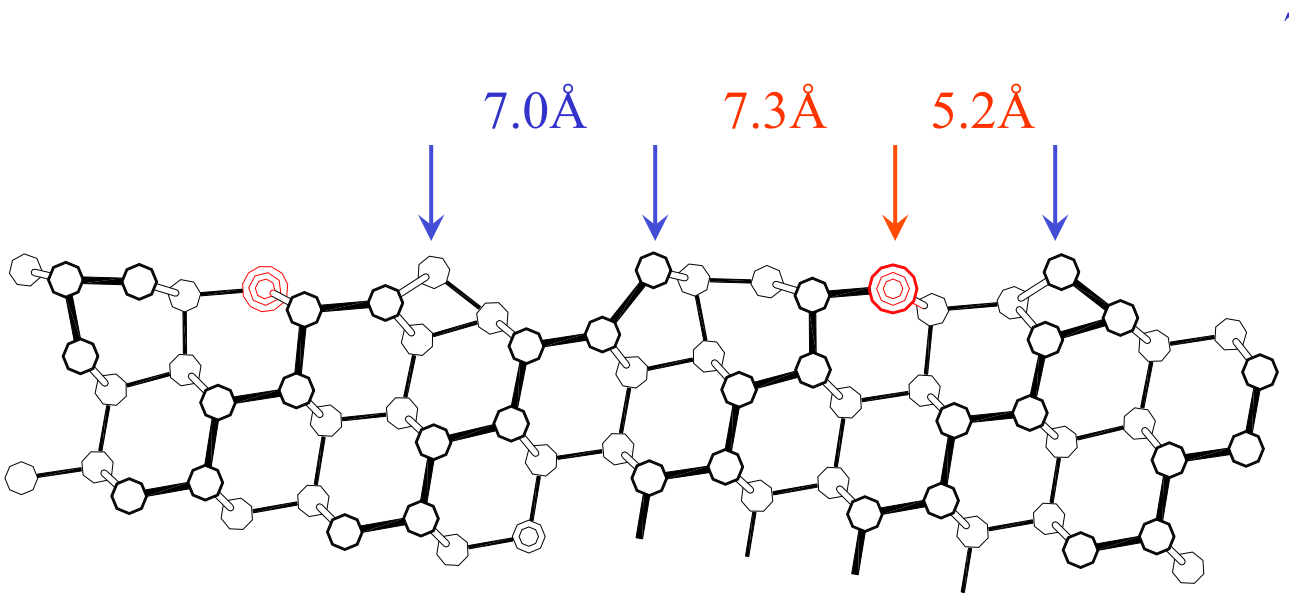
Electron density

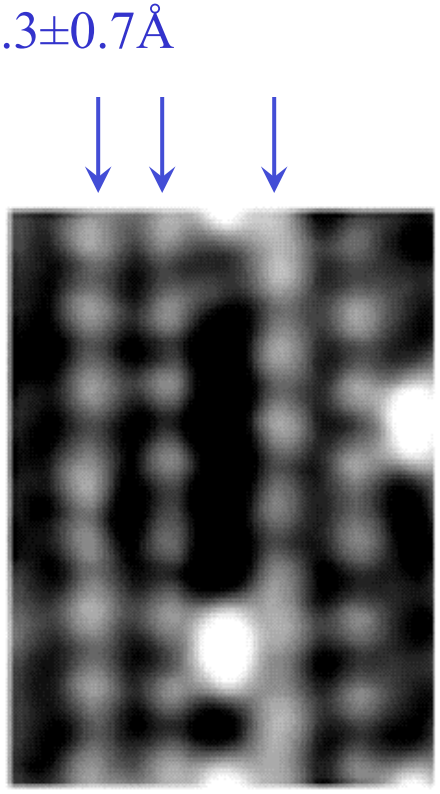
X-Z Patterson of Au/Si(557)




(a)
y
x
B
C
Au
(b)
z
x
(c)
Au

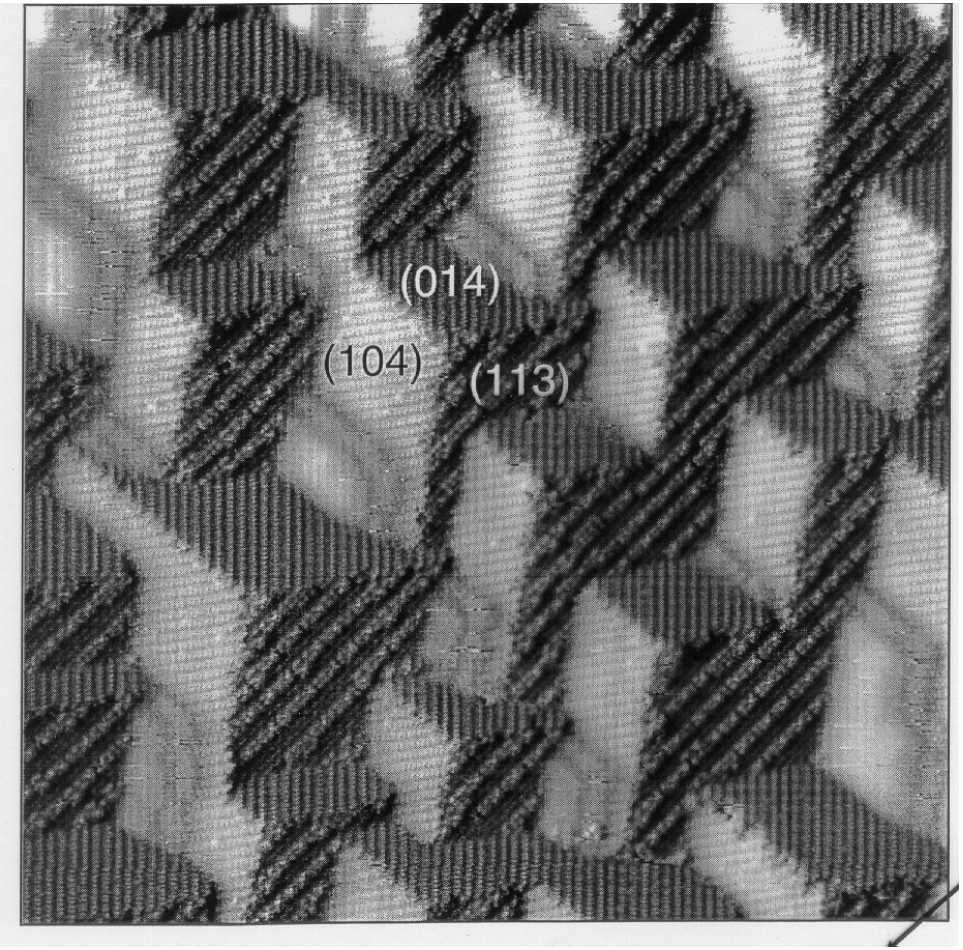
Comparison with STM

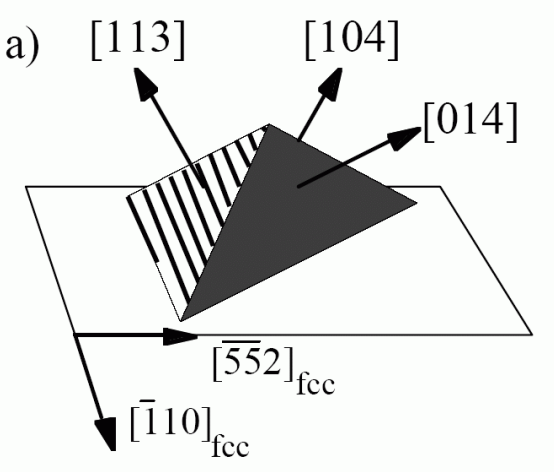


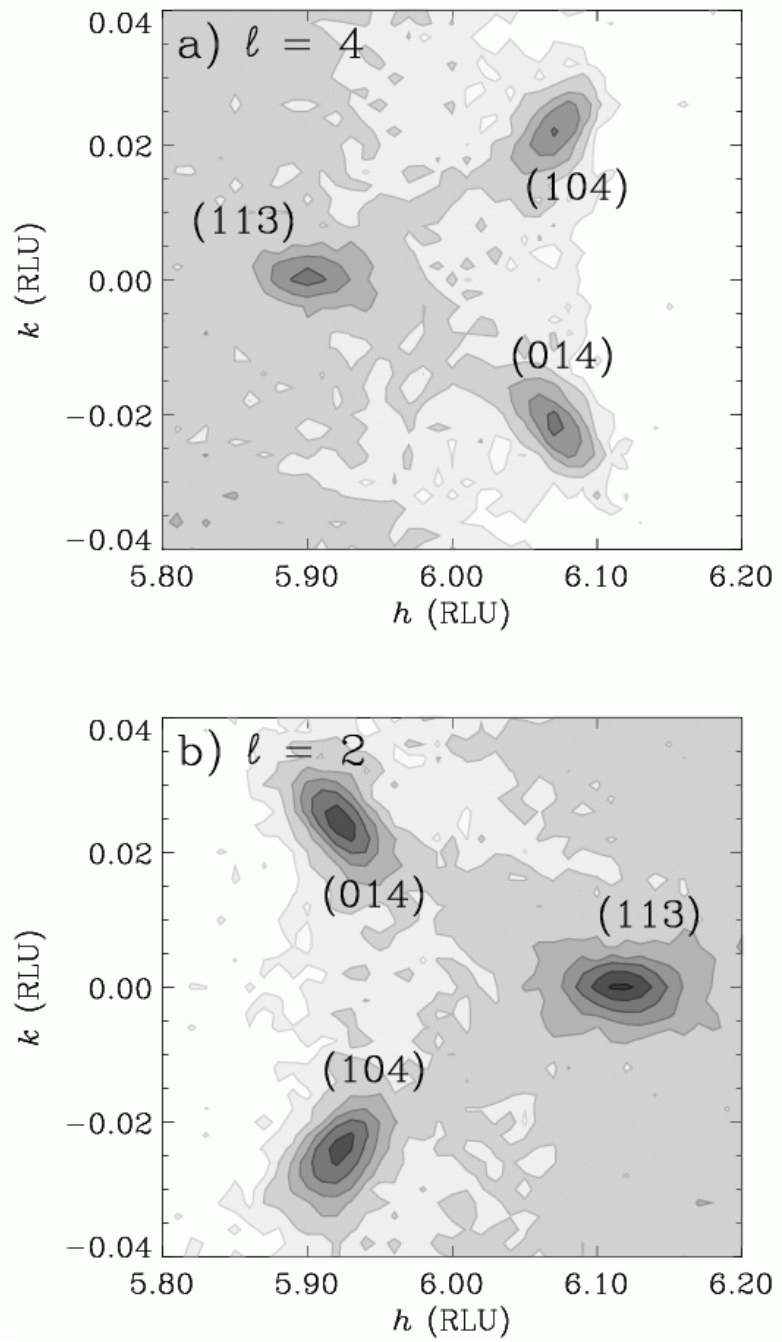


Cu(115) after Oxidation: STM

S. Reiter and E. Taglauer, Surf. Sci. **367** 33 (1996)



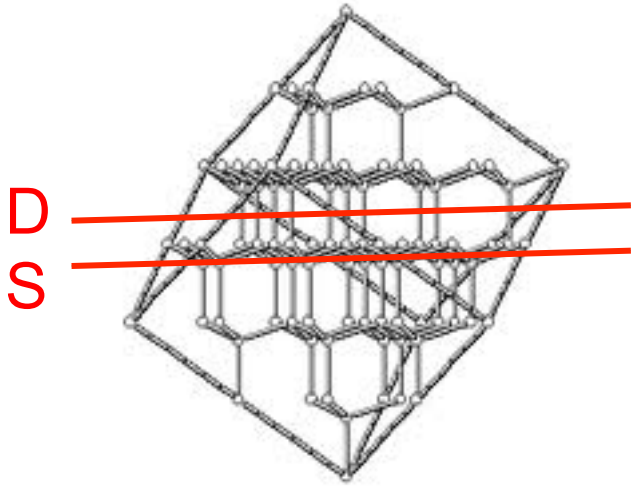


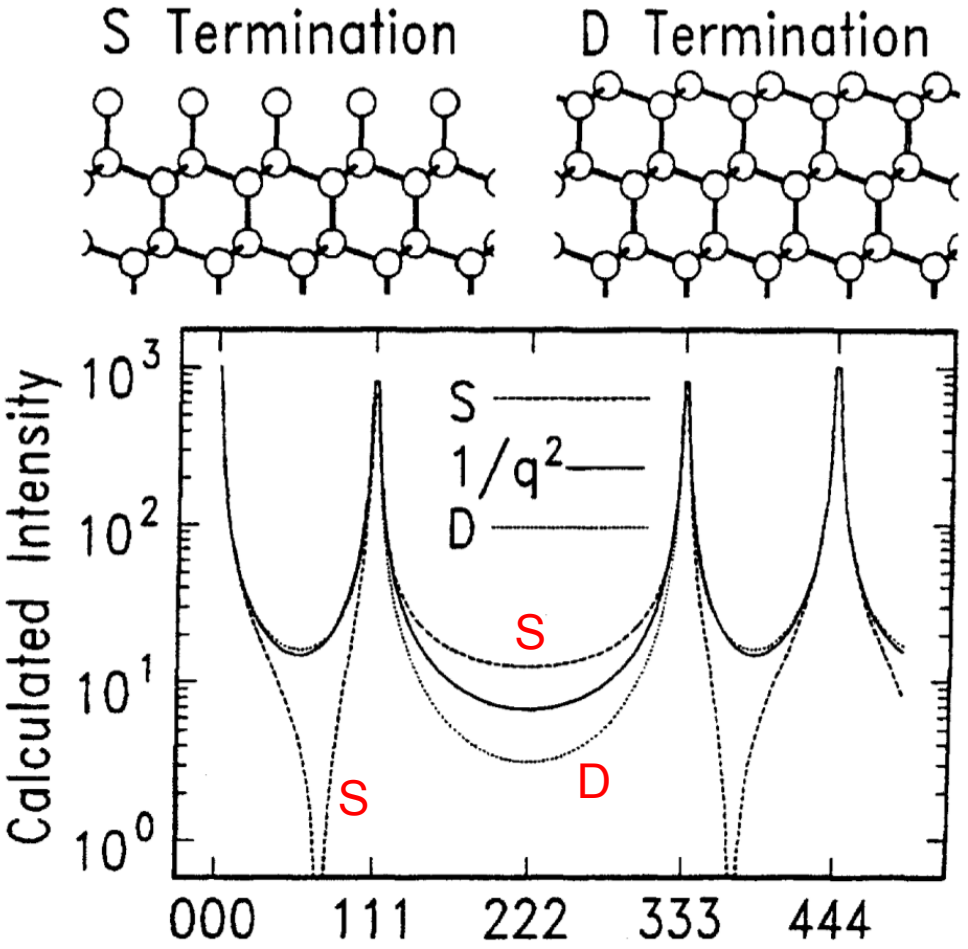


Cu(115) after Oxidation: X16A

Don Walko, UIUC PhD Dissertation (2000)

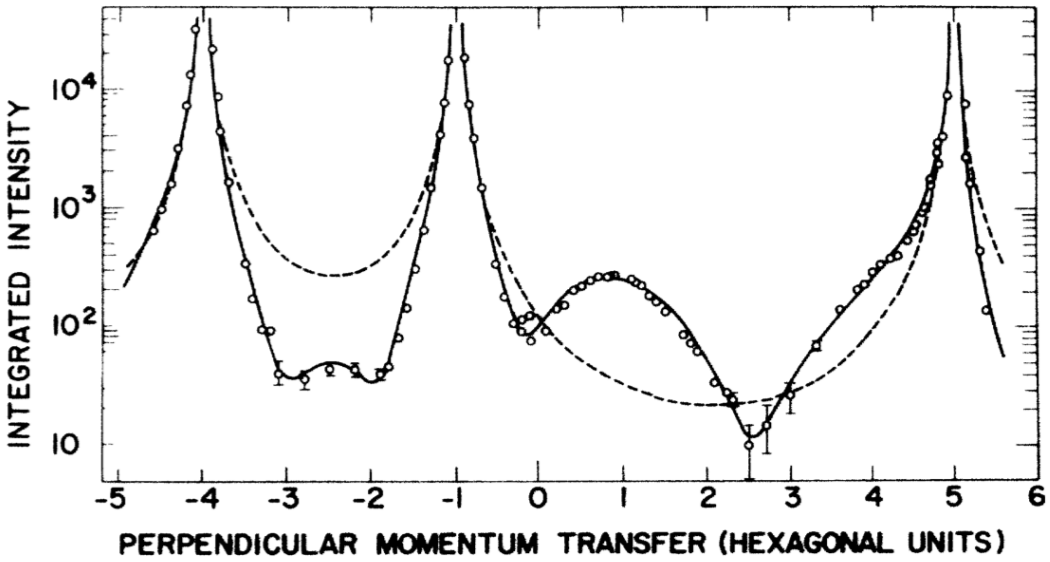
(111) Surface of Diamond Lattice

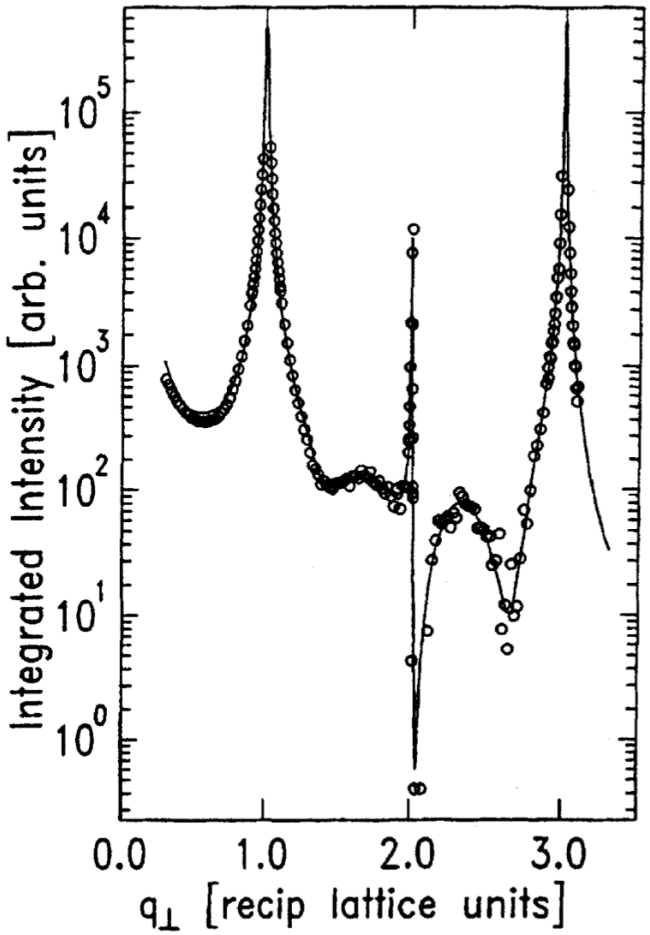




(111) Surface of Diamond Lattice

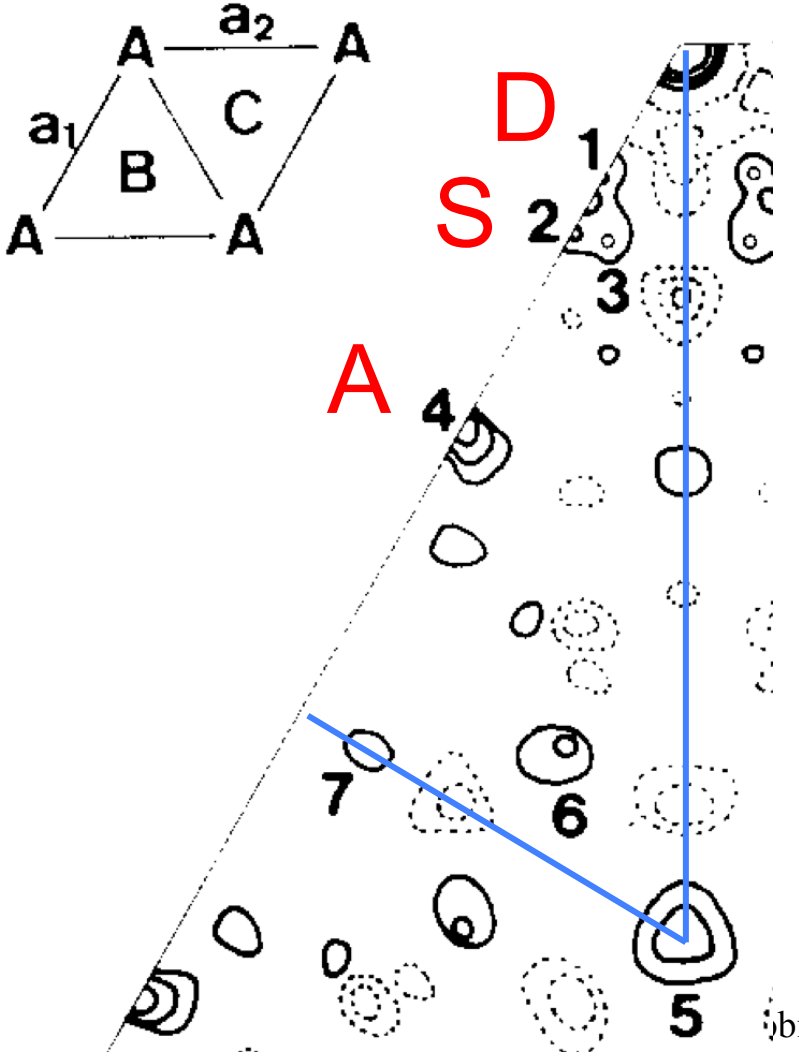
Si(111) 7x7 buried under a-Si
PRL 57 2714 (1986)

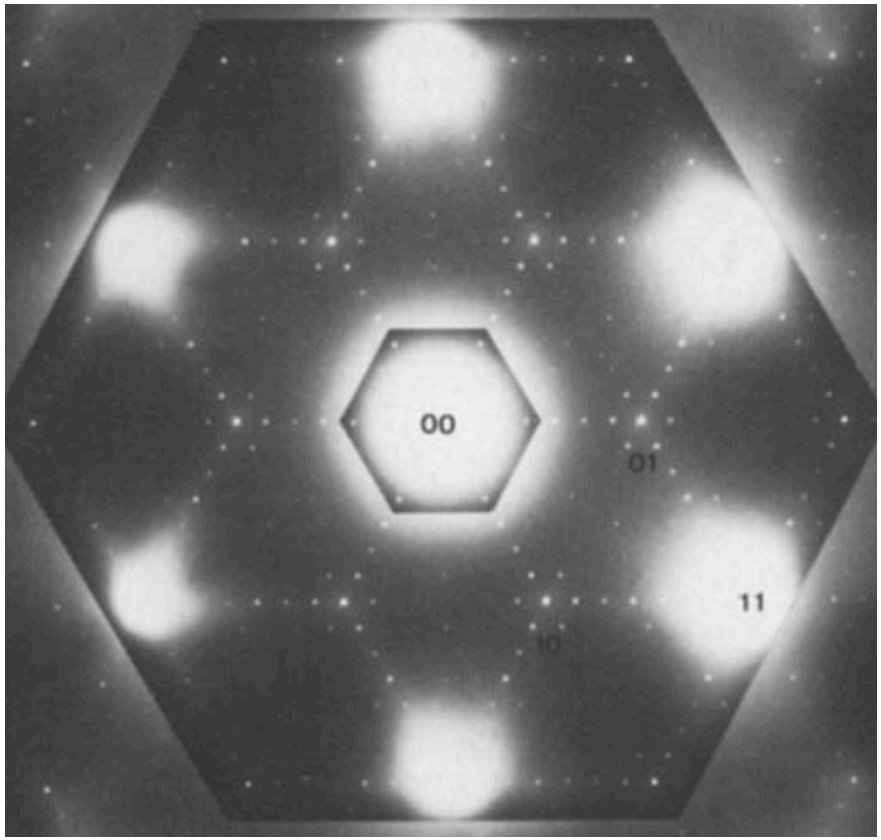




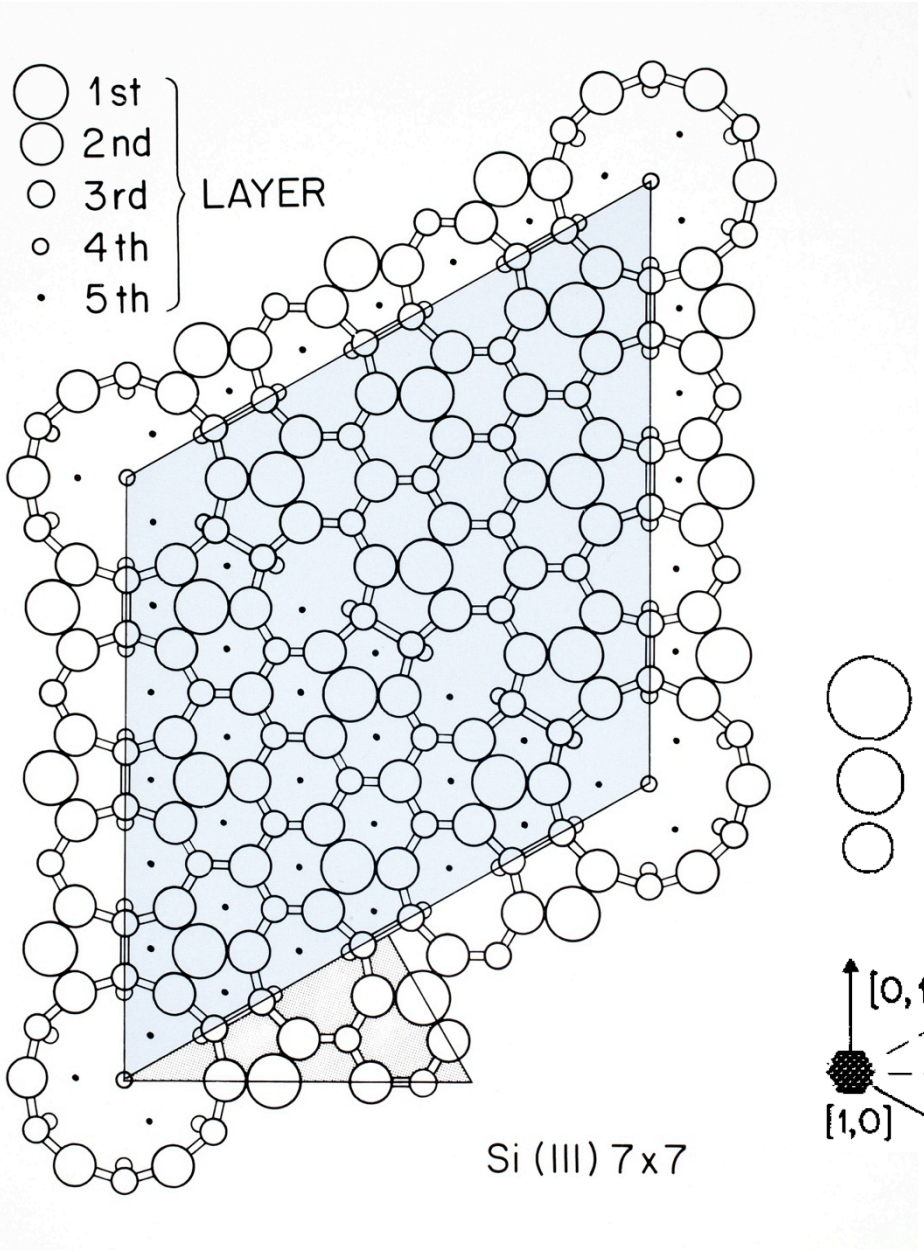
Patterson: Dimer-Adatom-Stacking-Fault

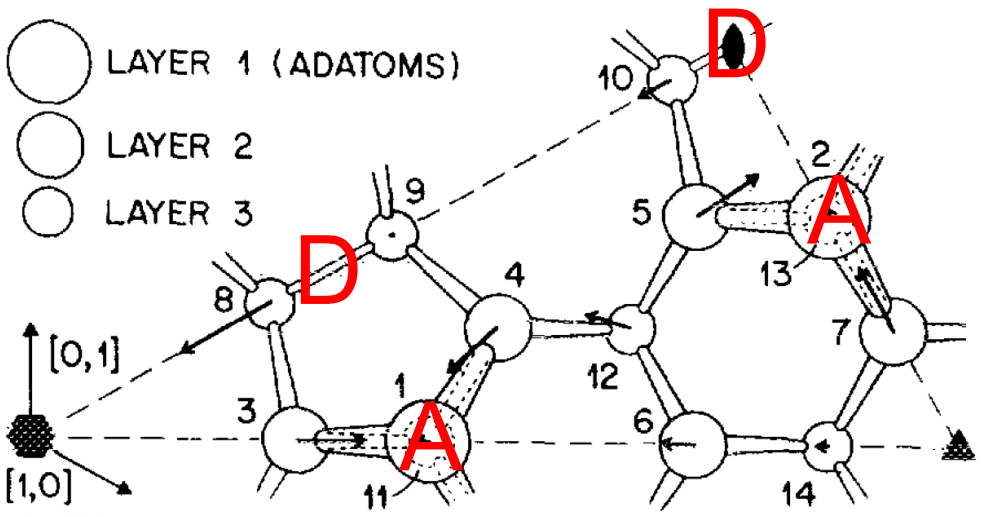
K. Takayanagi et al, Surf. Sci. 164 367 (1985)





DAS Model
Si(111) 7x7





7x7 Mosaic Tile

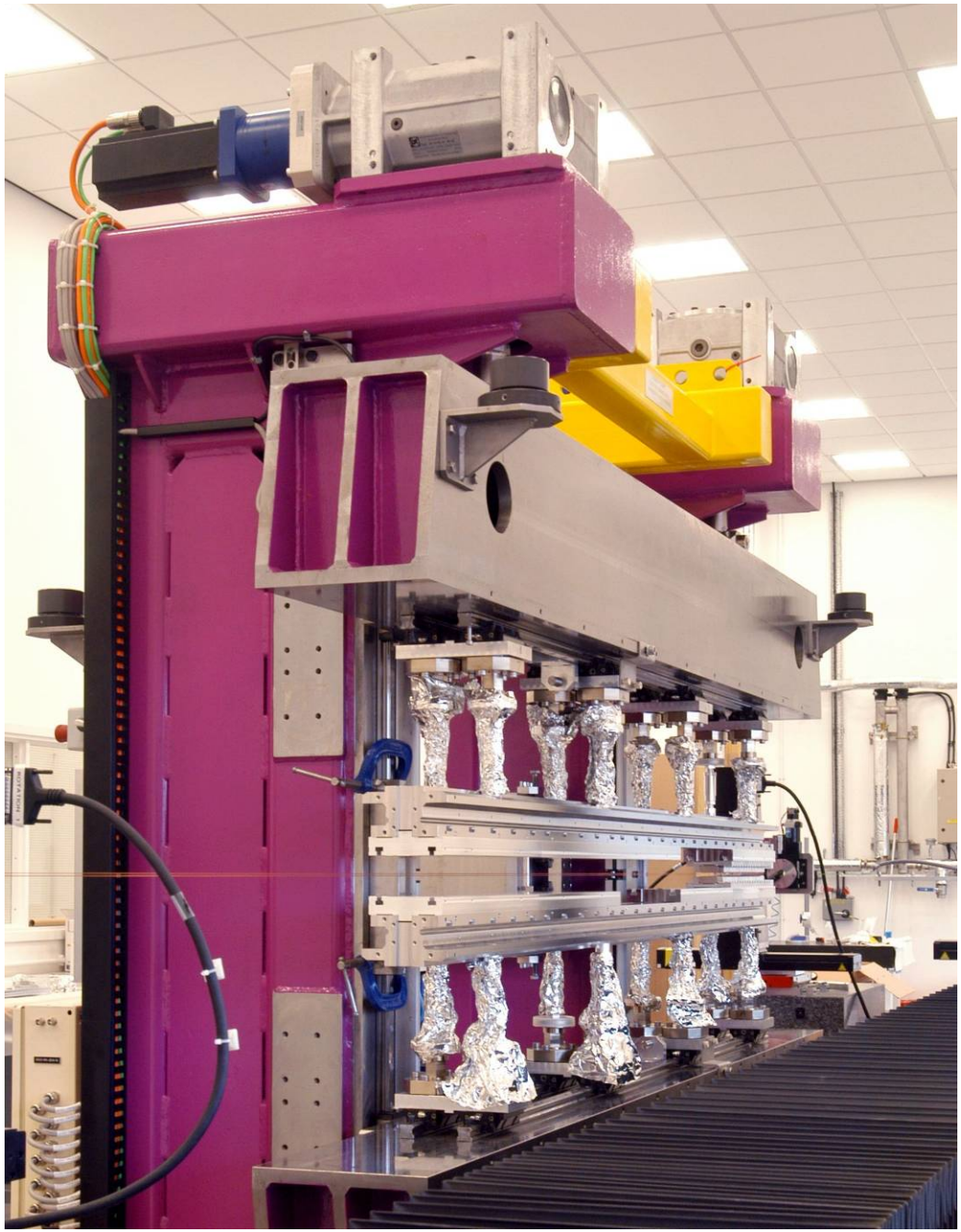
I. K. Robinson SXNS-14 2016

Generations of Synchrotron

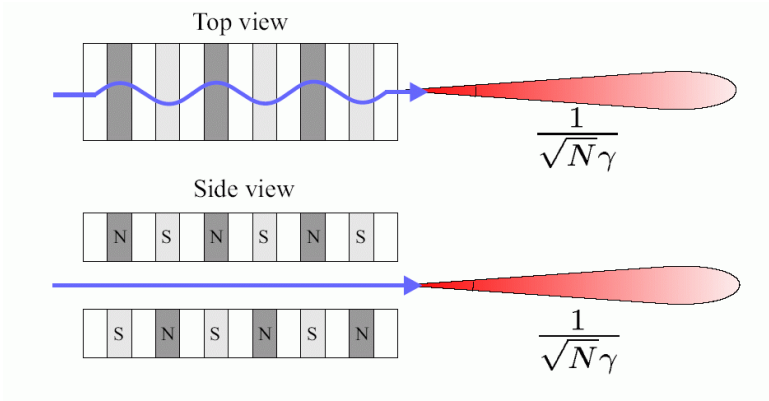
- 2nd: Storage Ring Dedicated to X-rays
  - NSLS Surface X-ray Diffraction
- 3rd: Undulators in "Straight Sections"
  - APS Coherent X-ray Diffraction
  - Diamond Light Source
- X-ray Free Electron Lasers
  - LCLS Ultrafast Coherent Diffraction

Advanced Photon Source (ANL)

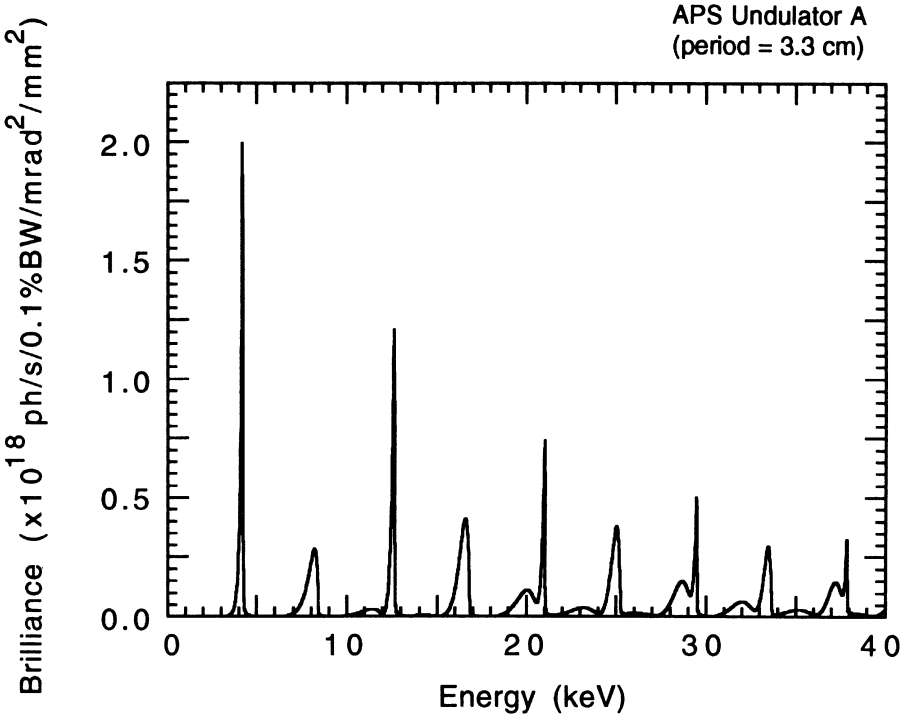
Diamond in-vacuum X-ray Undulator



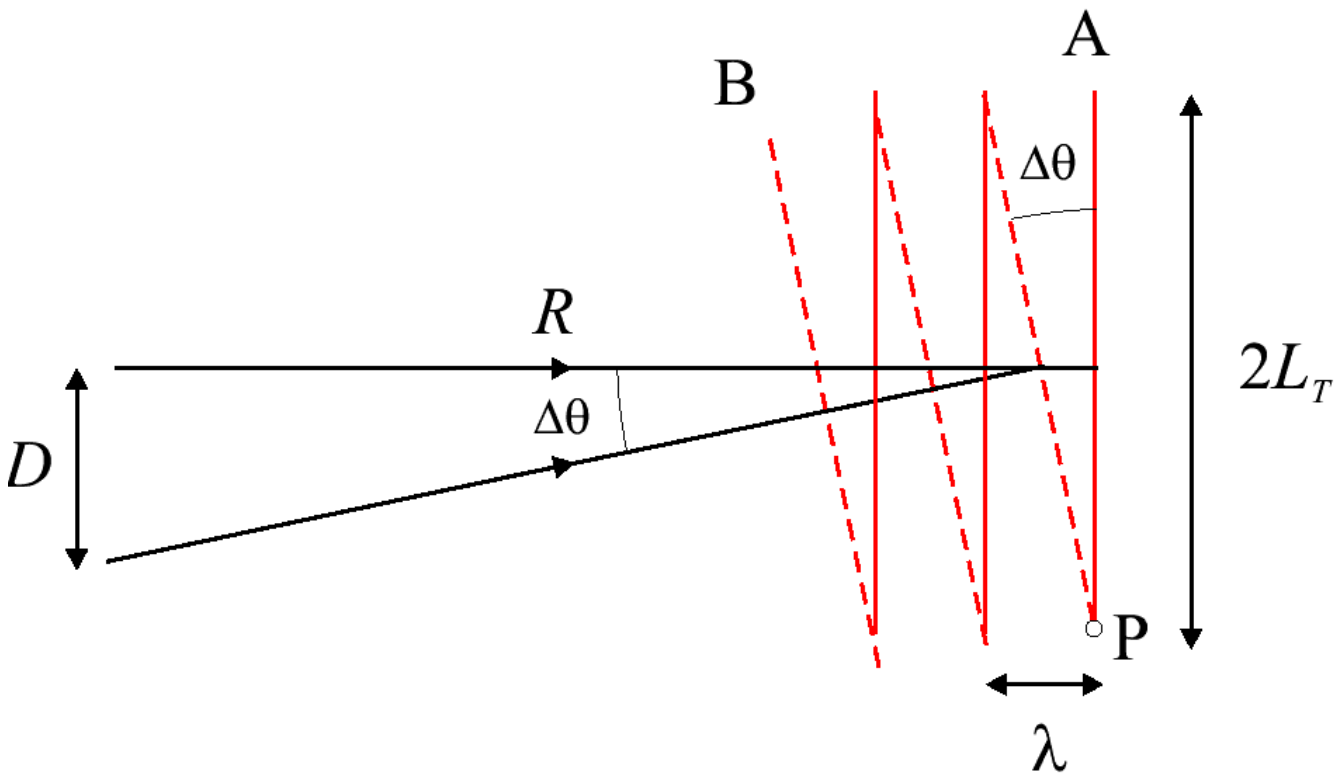
X-ray Undulator Principle



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



Lateral (Transverse) Coherence

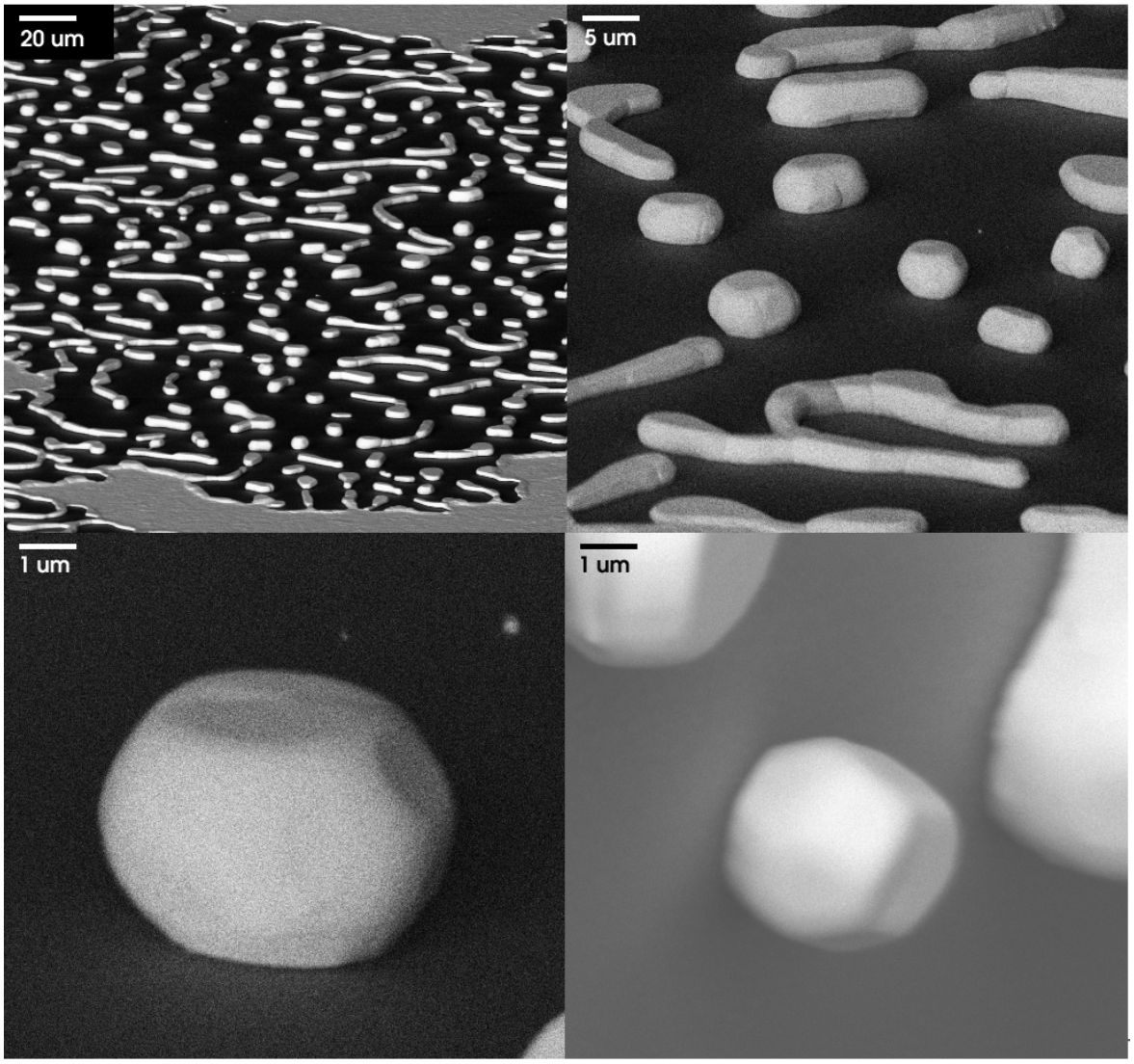


$$L_T = \frac{\lambda}{2}\frac{R}{D}$$

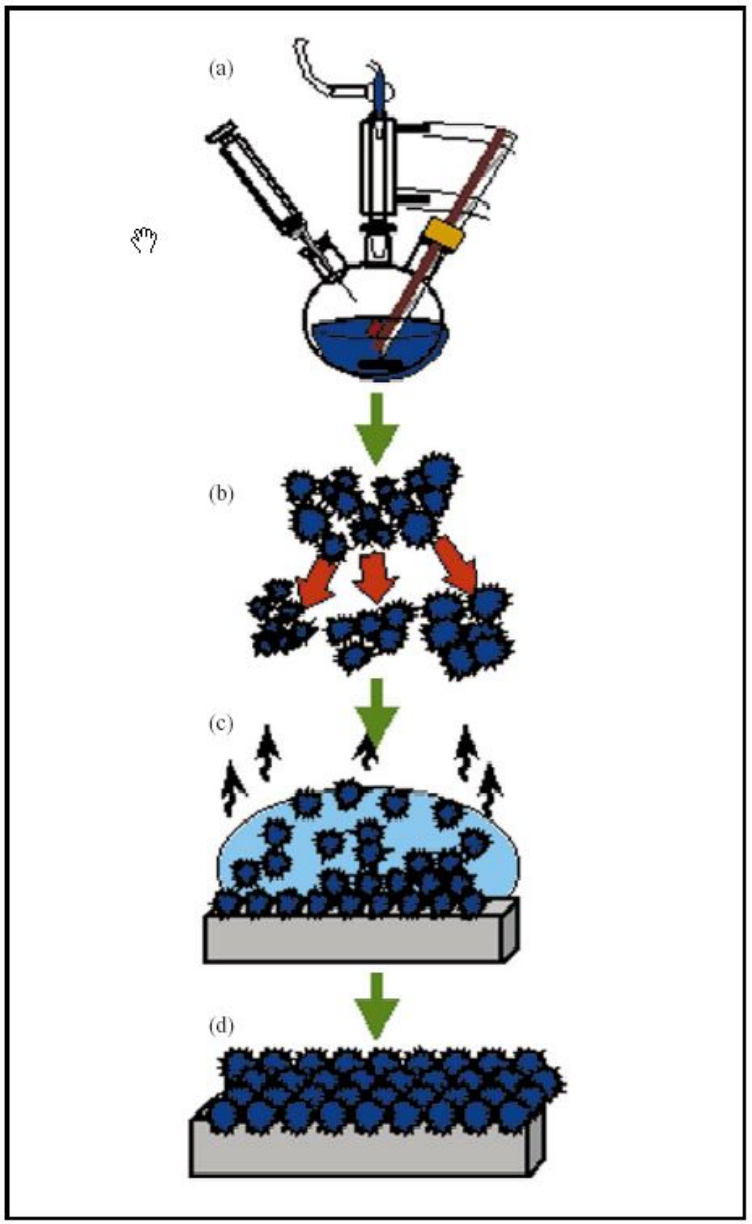
Als-Nielsen and McMorrow (2001)

SEMS

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

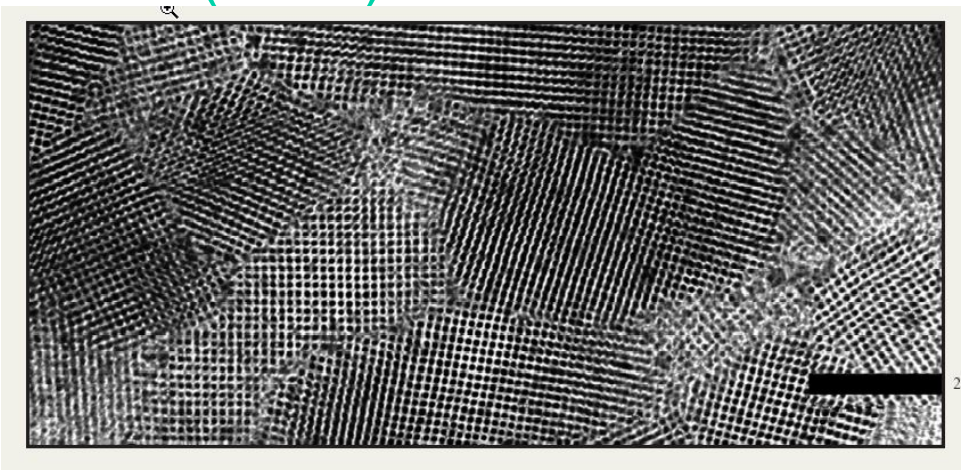


Chemical Synthesis of Nanocrystals



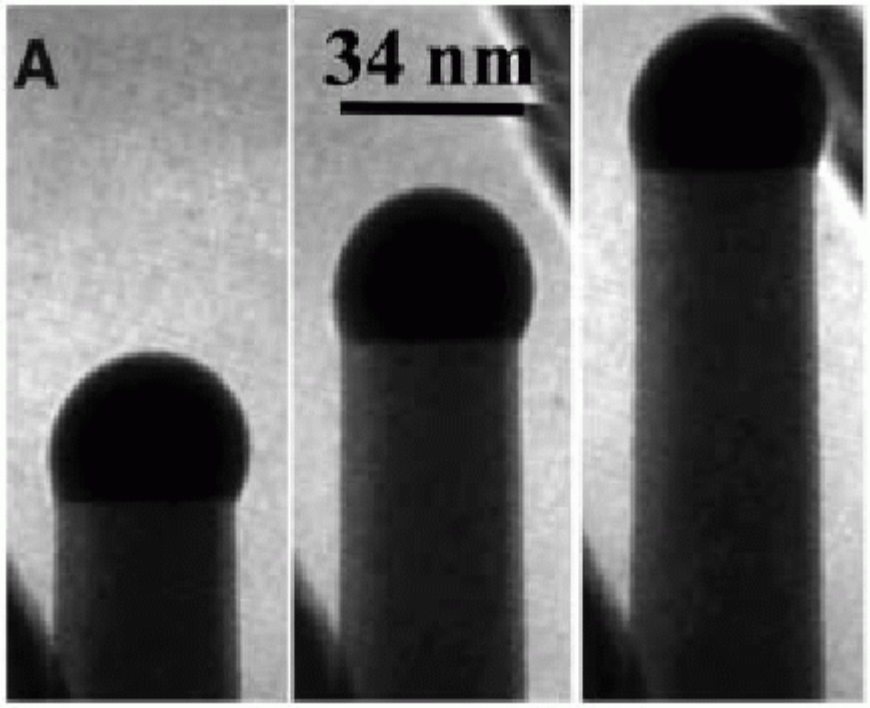
- Reactants introduced rapidly
- High temperature solvent
- Surfactant/organic capping agent
- Square superlattice (200nm scale)

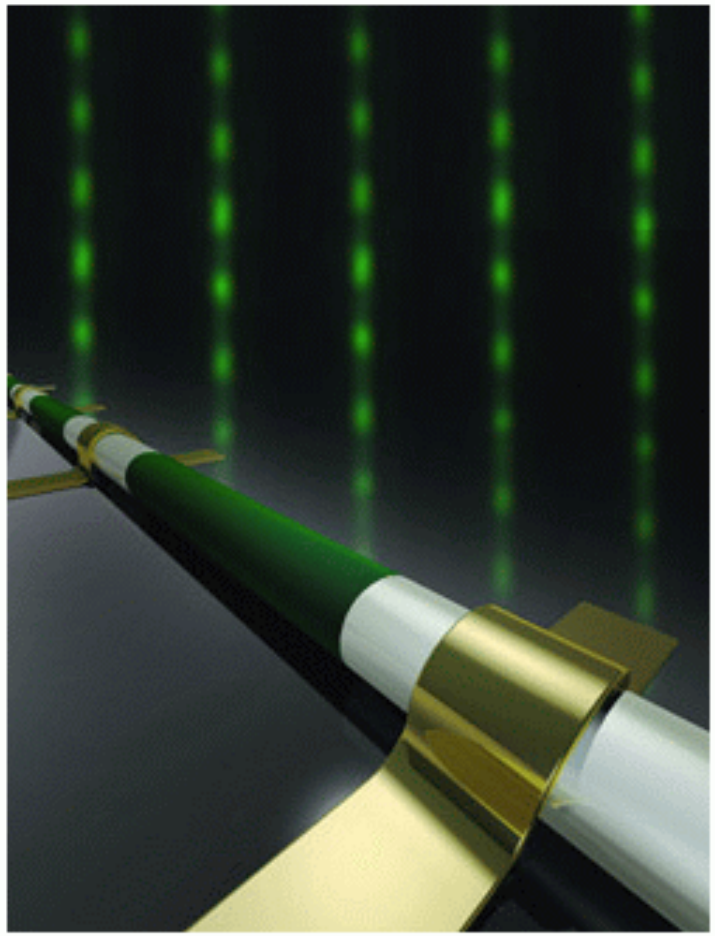
C. B. Murray, IBM J. Res. & Dev. **45** 47 (2001)



VLS growth of nanowires

S. Kodambaka et al., Science 316 729 (2007)

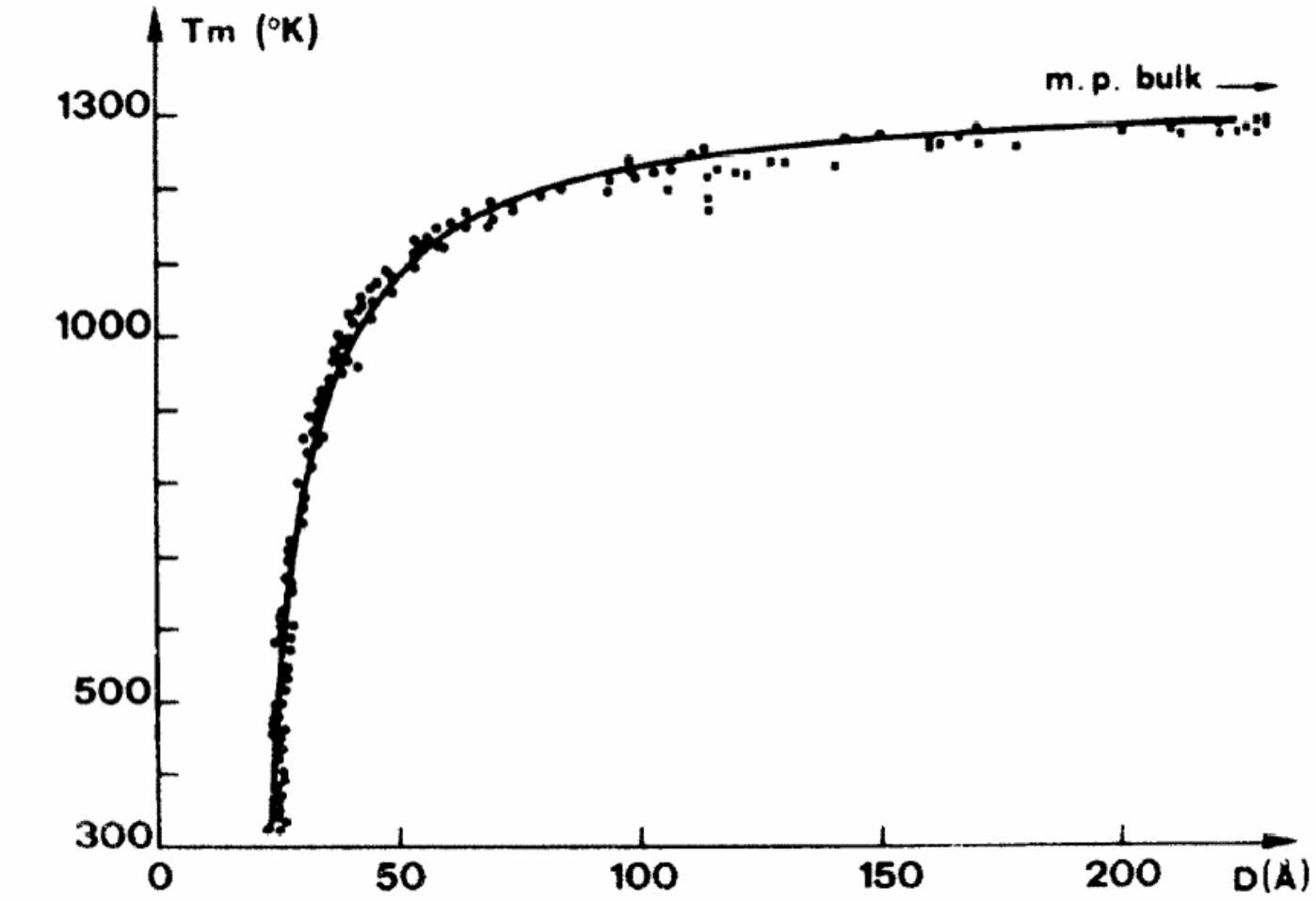




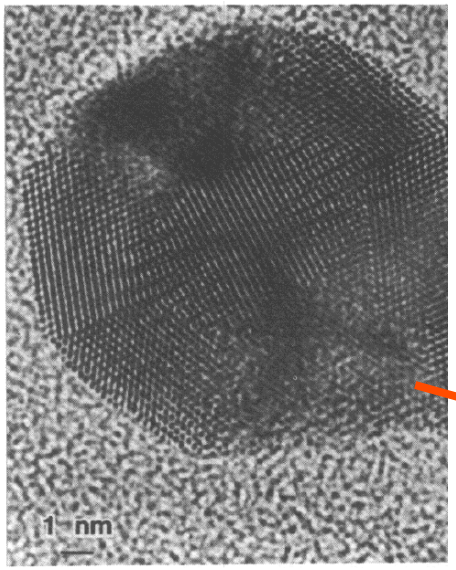
NiSi/Si nanowire heterostructure devices. *Nature* **430**, 61 (2004).

Size-dependent Melting of Au Particles

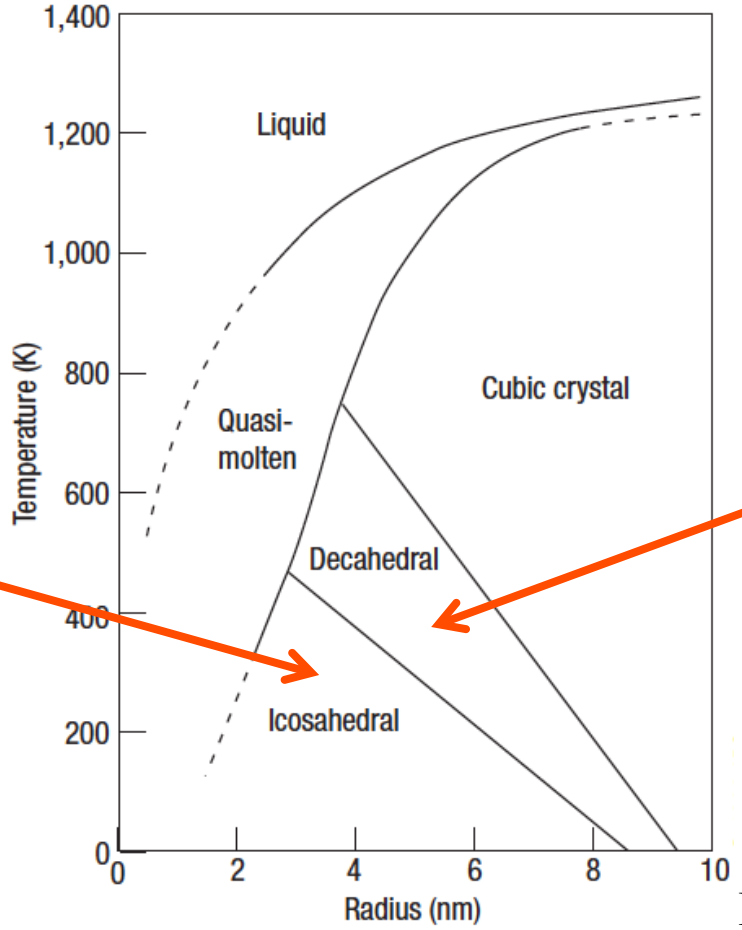
P. Buffat and J-P. Borel, Phys. Rev. A 2287-97 (1975)

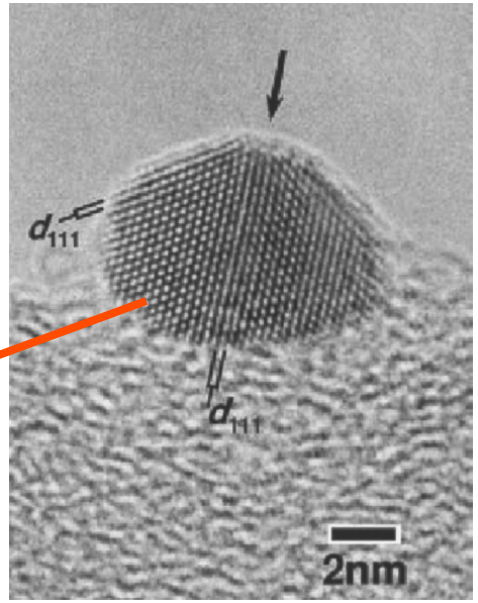


Structure of Gold vs Size



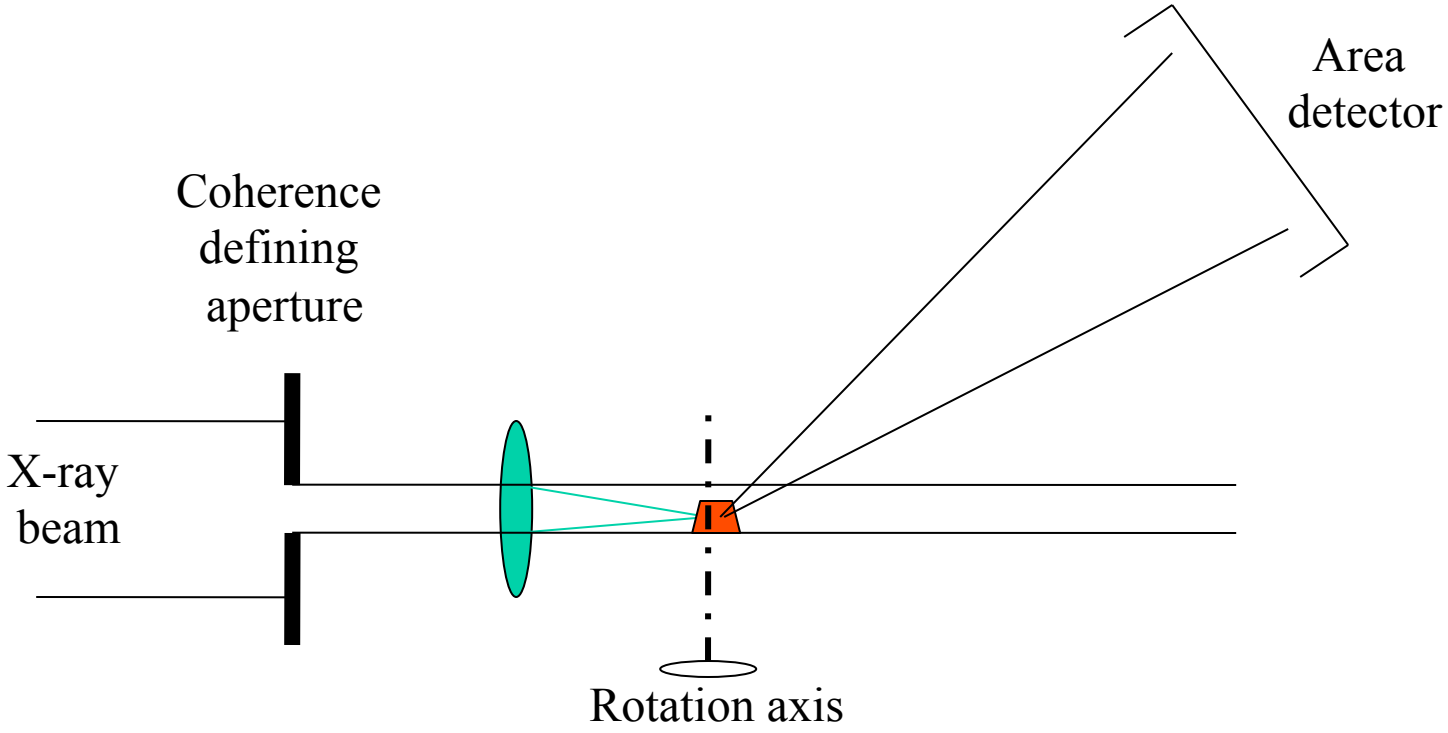
L. D. Marks, RPP (1994)

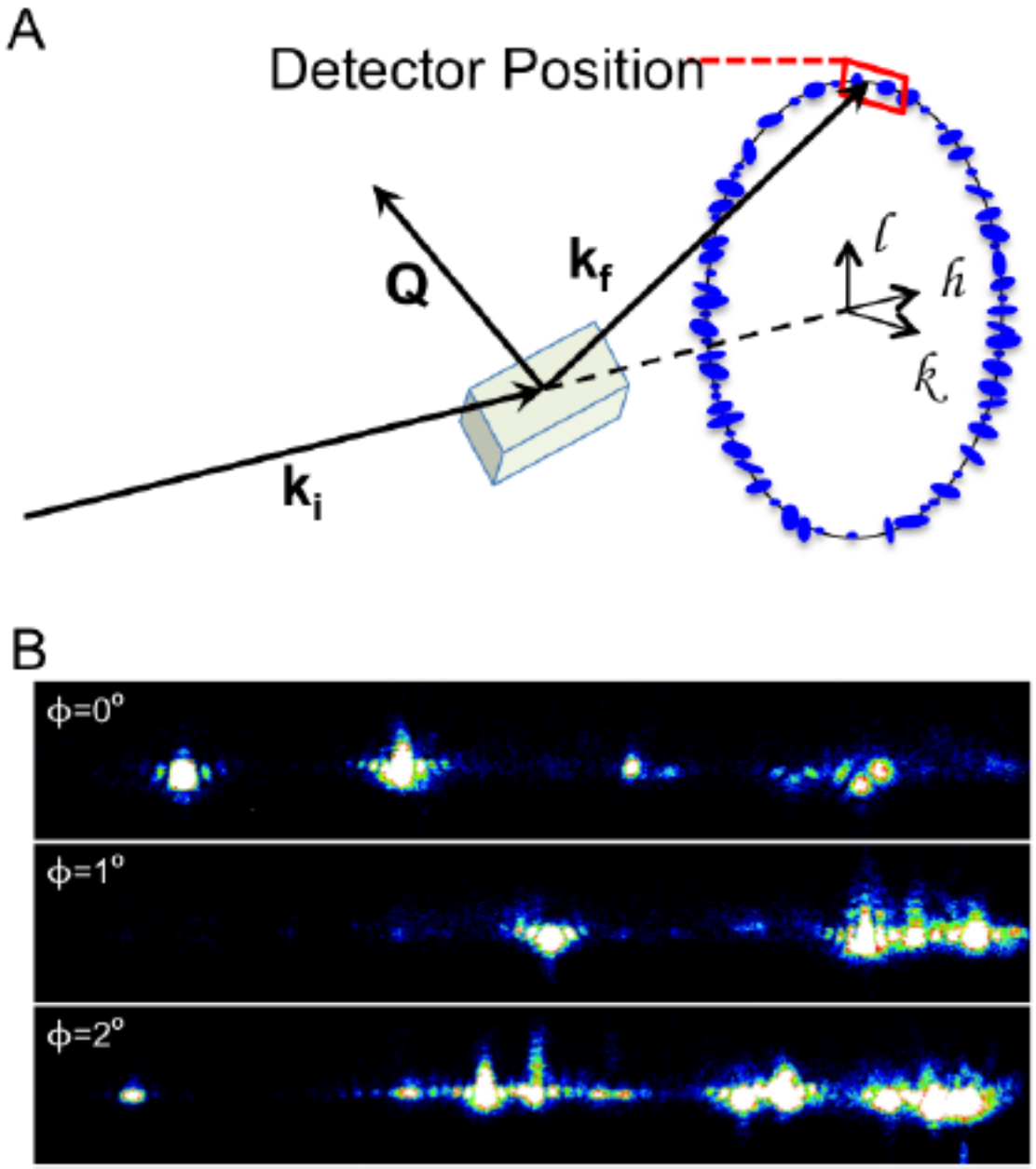




Koga and Sugawara (2003)

Lensless X-ray Microscope, 2003





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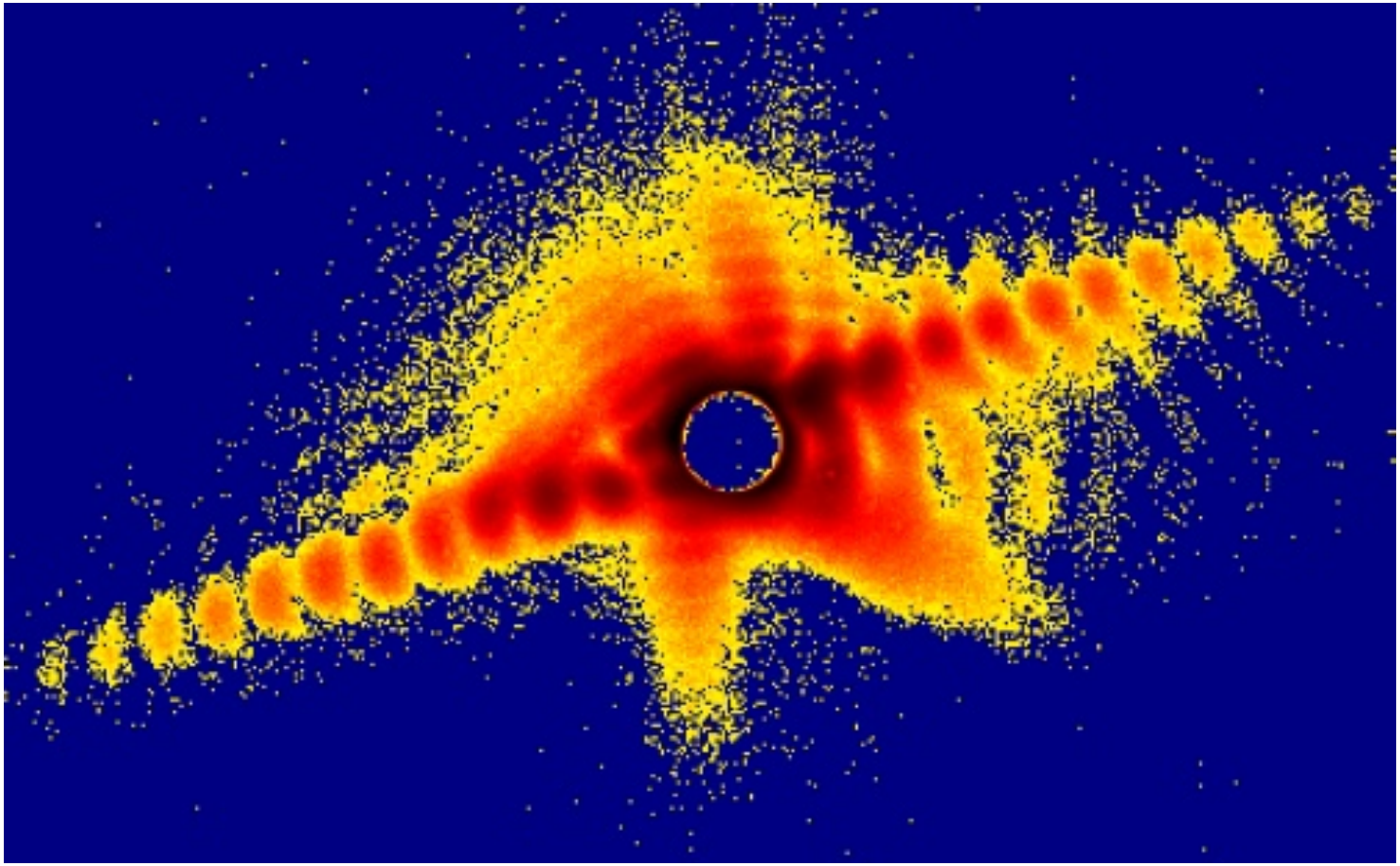
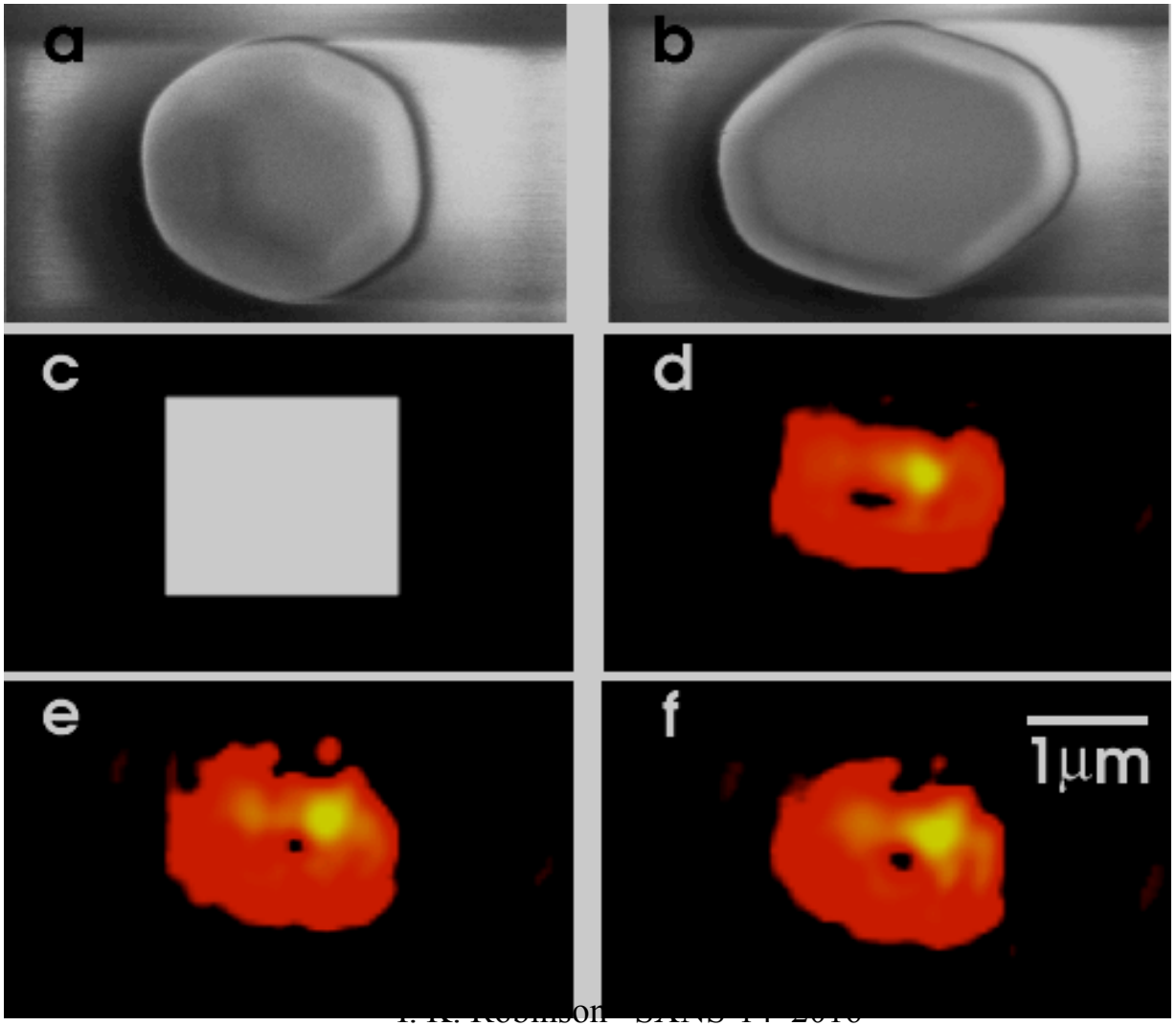
Coherent X-ray Diffraction from Gold Nanocrystal, (111) Bragg Peak



I. K. Robinson SXNS-14 2016

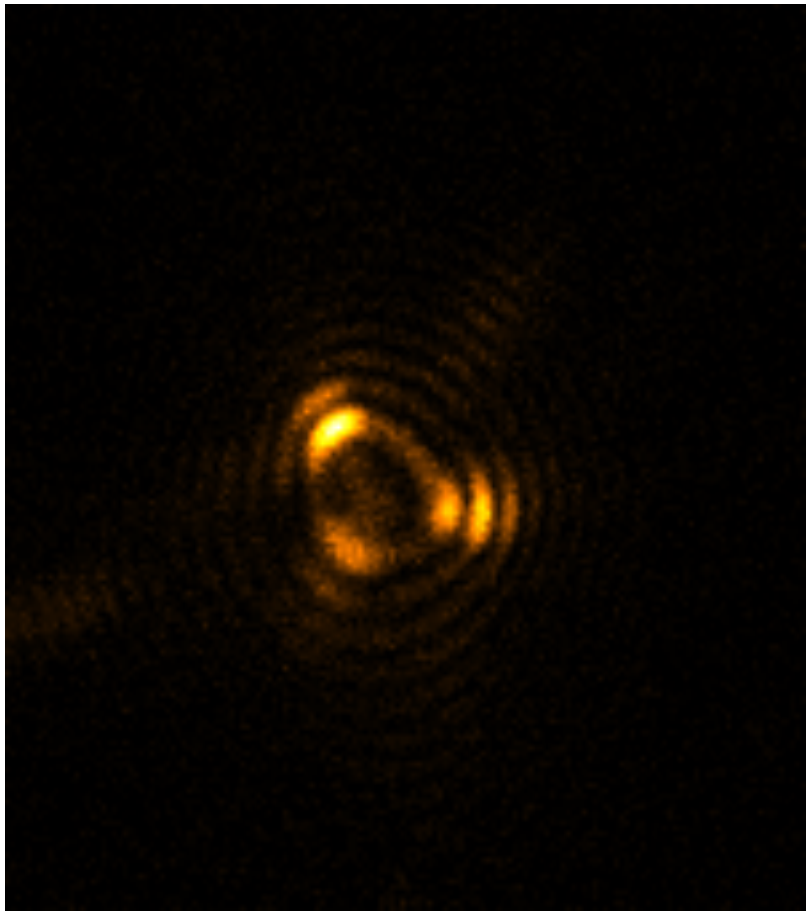
First Reconstruction of Gold Nanocrystals

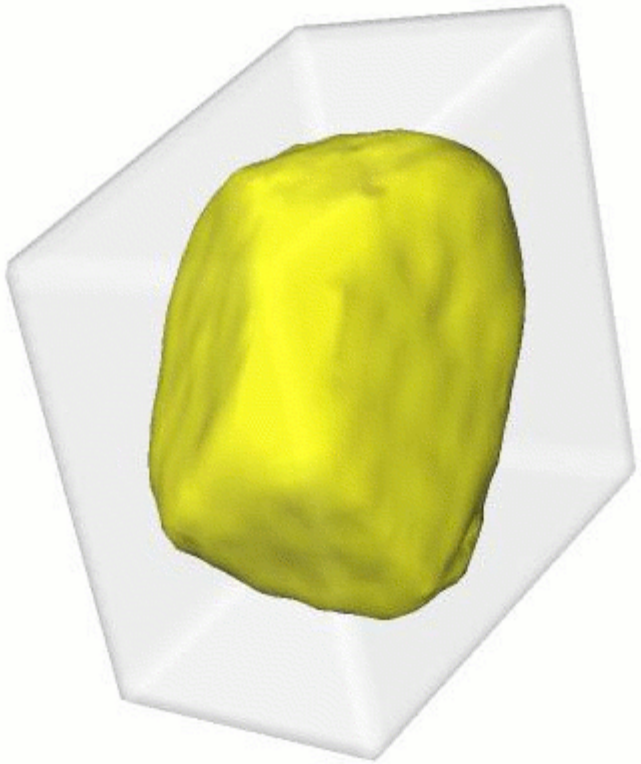
PRL 87 195505 (2001)



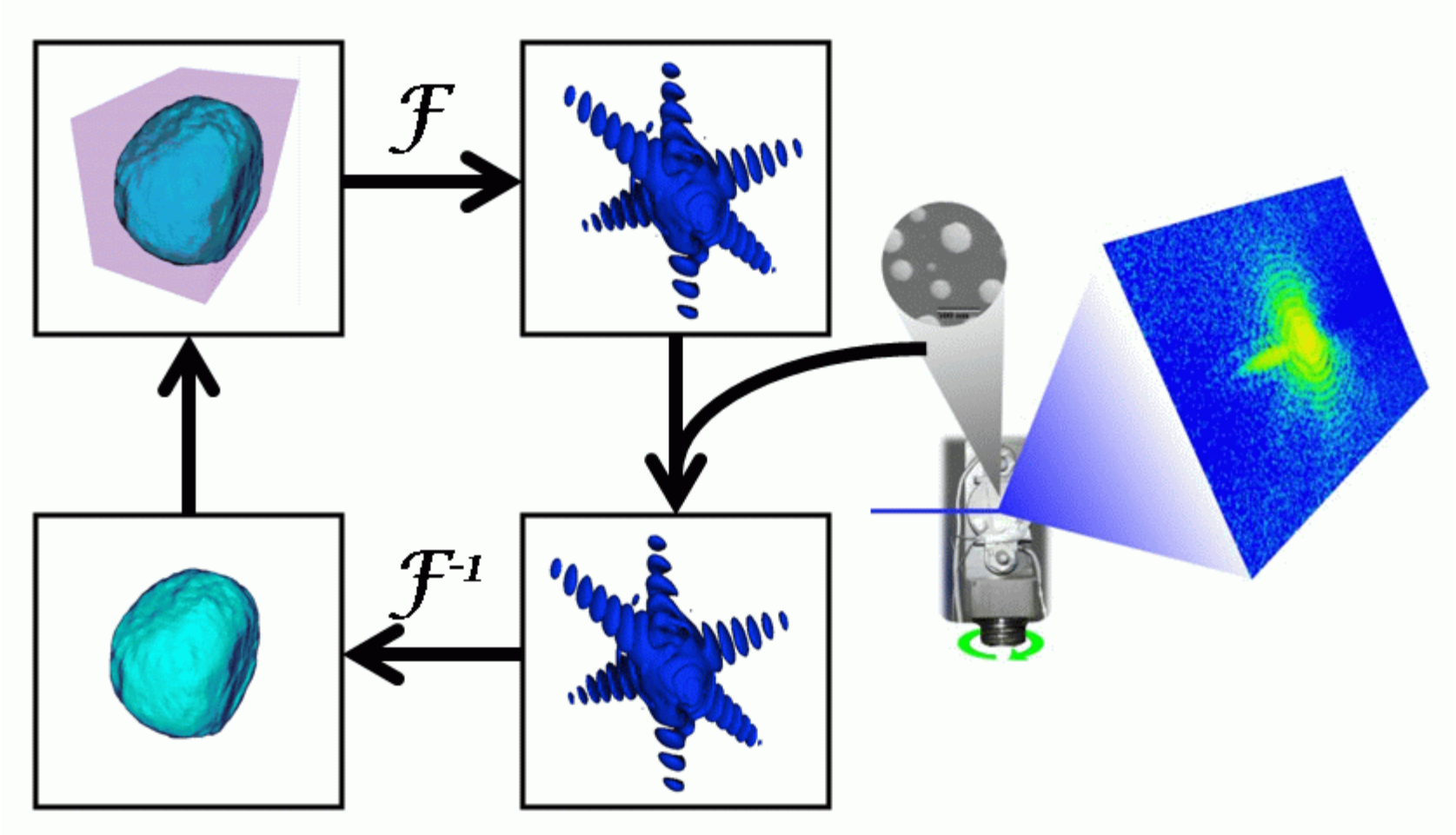
Gold nanocrystal reconstruction

showing support used for 20 HIO followed by 10 ER



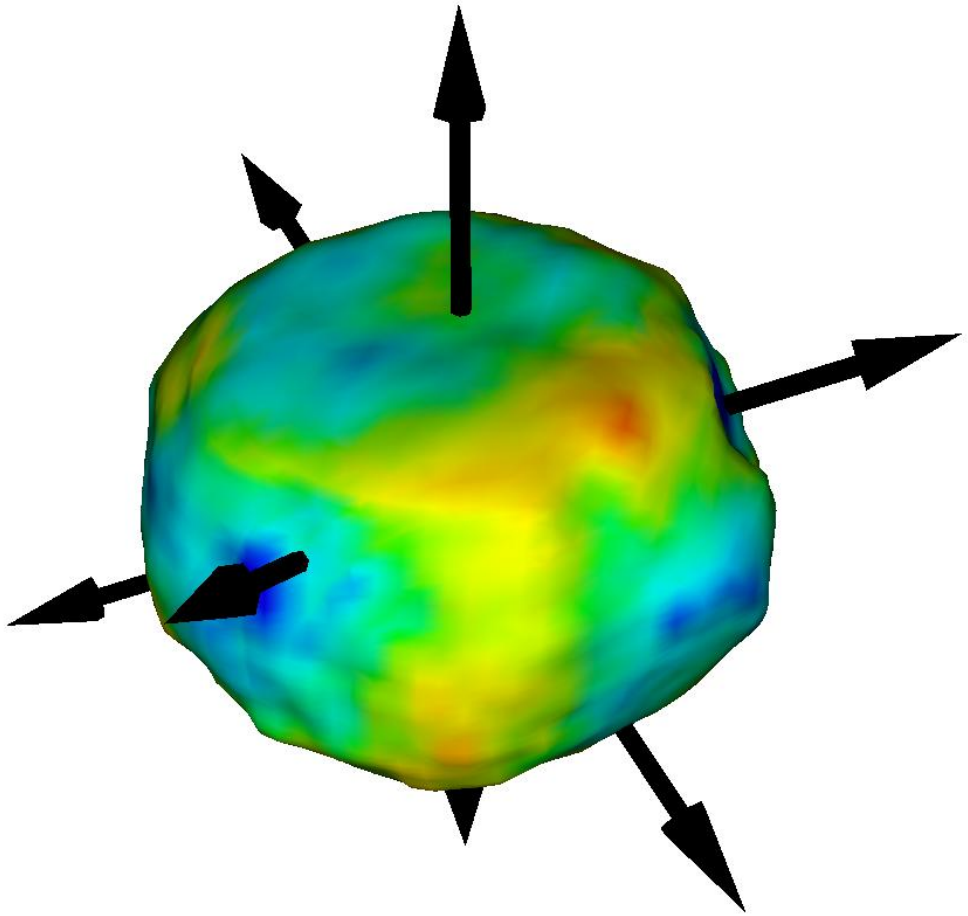


Generic "Error Reduction" method



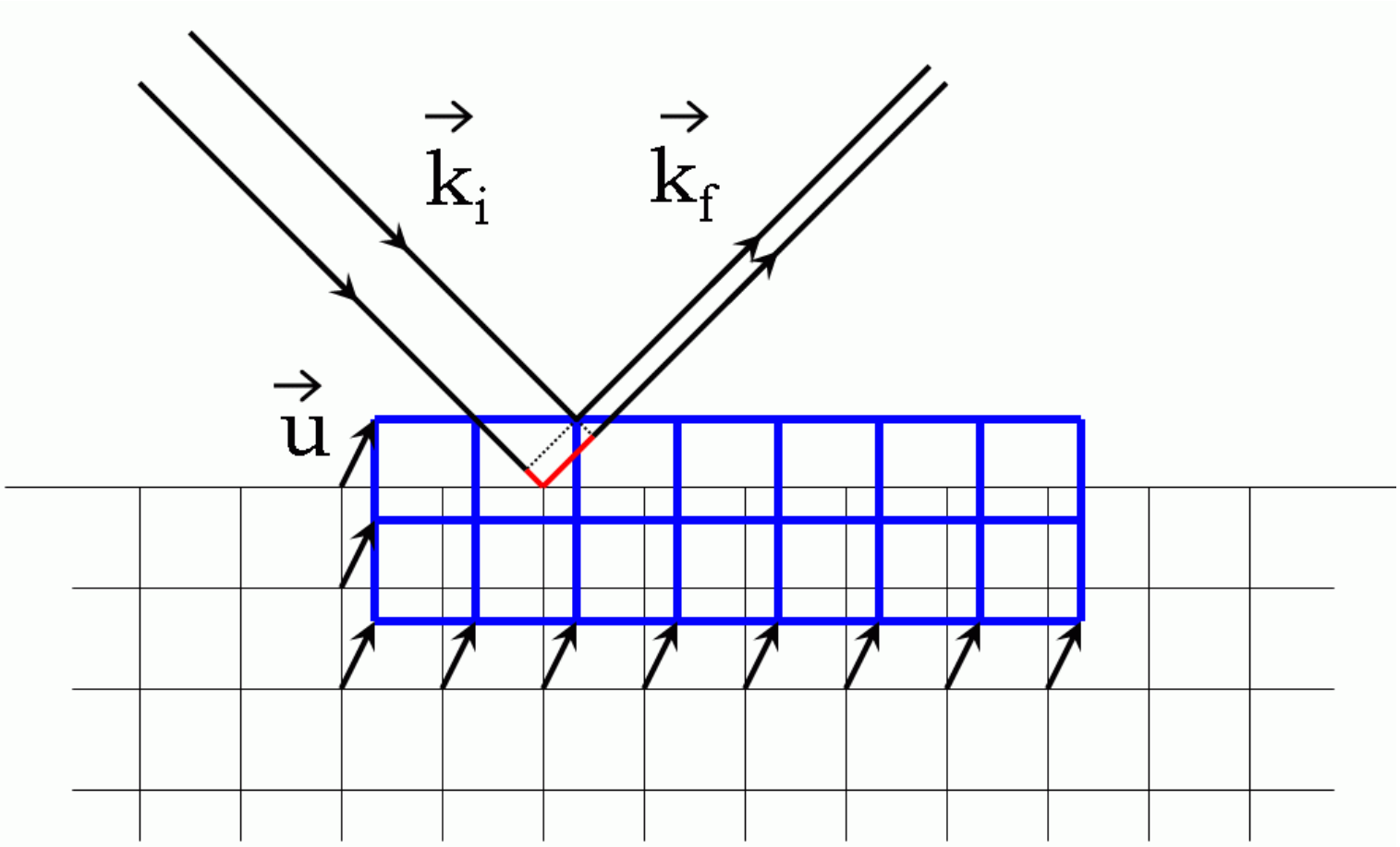
J. R. Fienup Appl. Opt. 21 2758 (1982)
R. W. Gerchberg and W. O. Saxton Optik 35 237 (1972)
I. K. Robinson SXNS-14 2016

Phase isosurface of residual strain



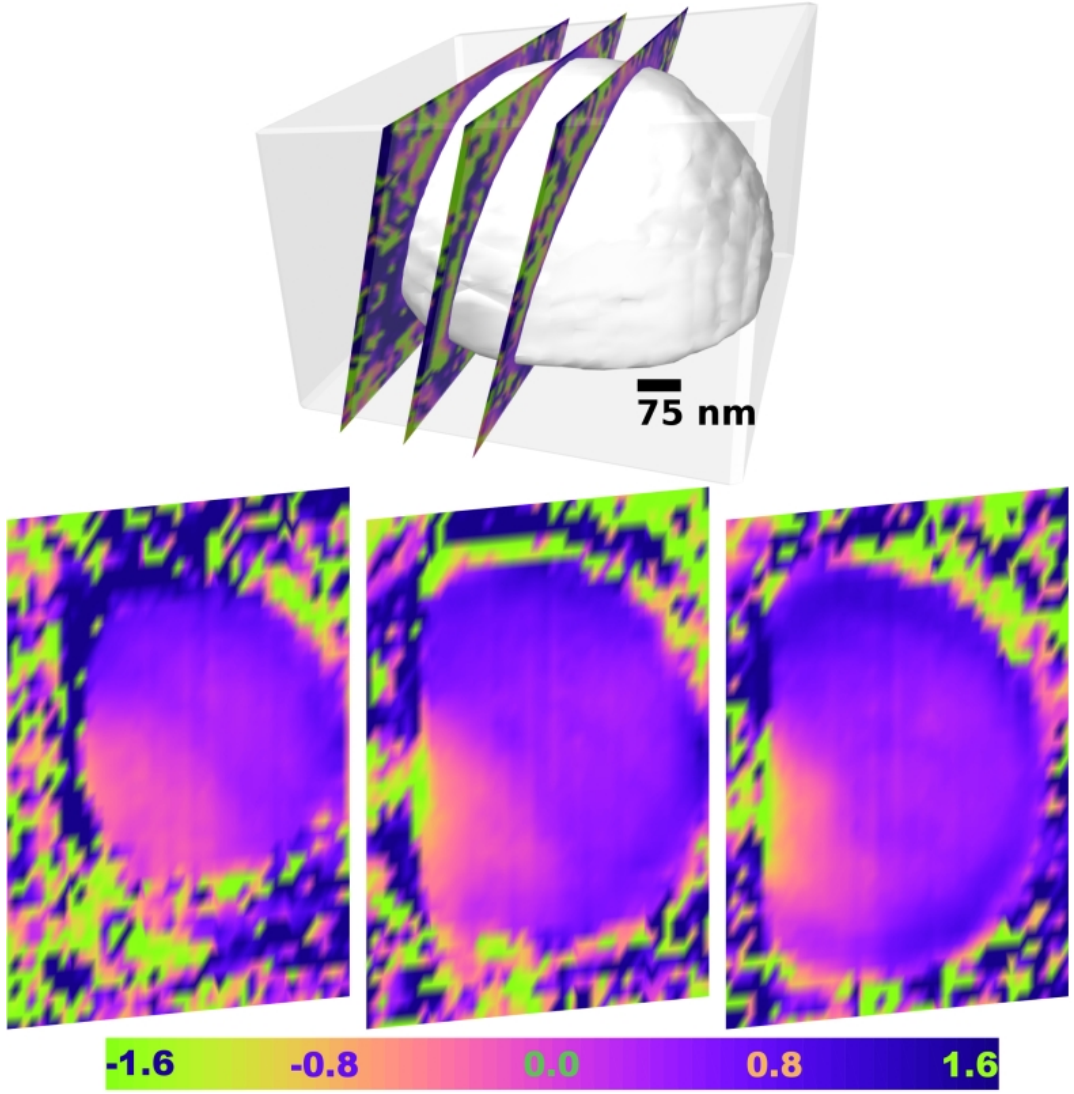
Sensitivity to strain

$$\Delta\varphi = \mathbf{k}_f \cdot \mathbf{u} - \mathbf{k}_i \cdot \mathbf{u} = \mathbf{Q} \cdot \mathbf{u}$$



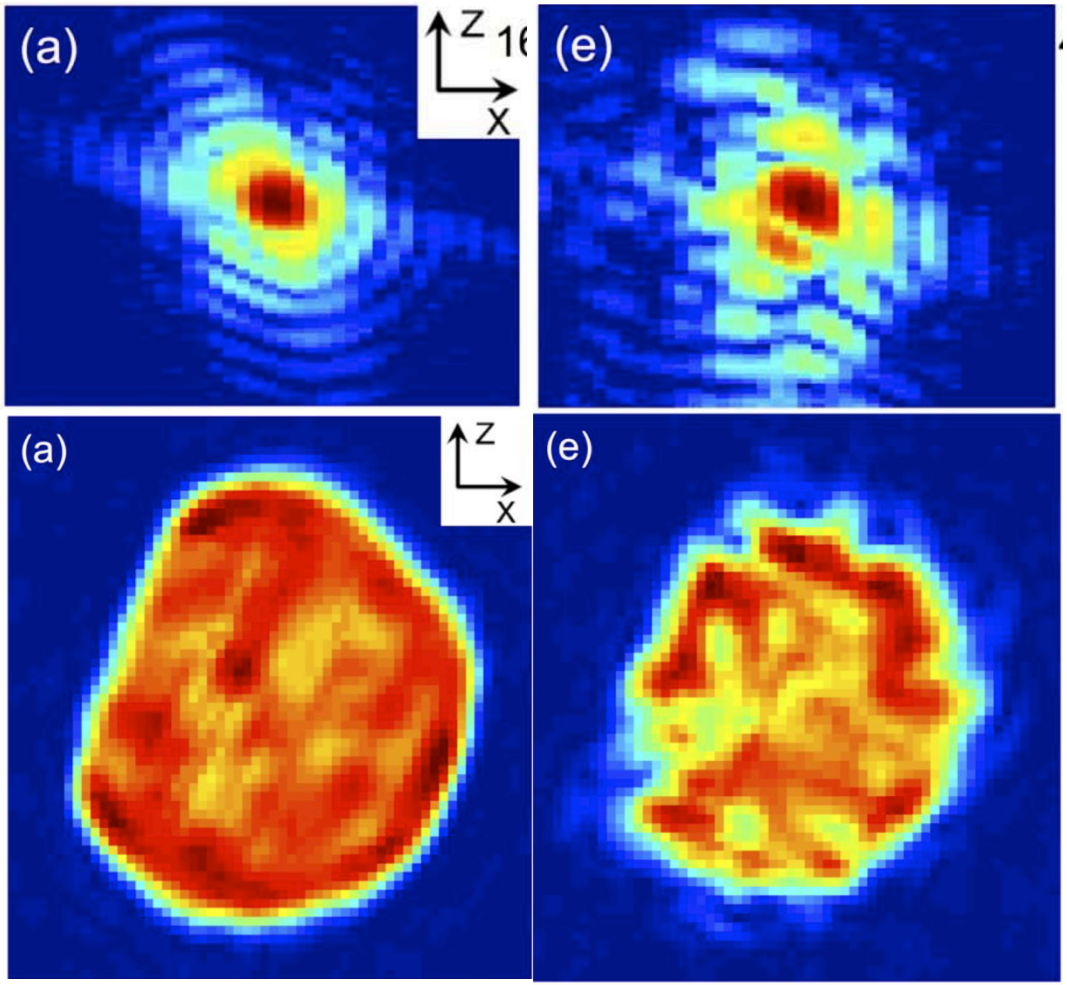
Contact Forces cause Strain in Pb

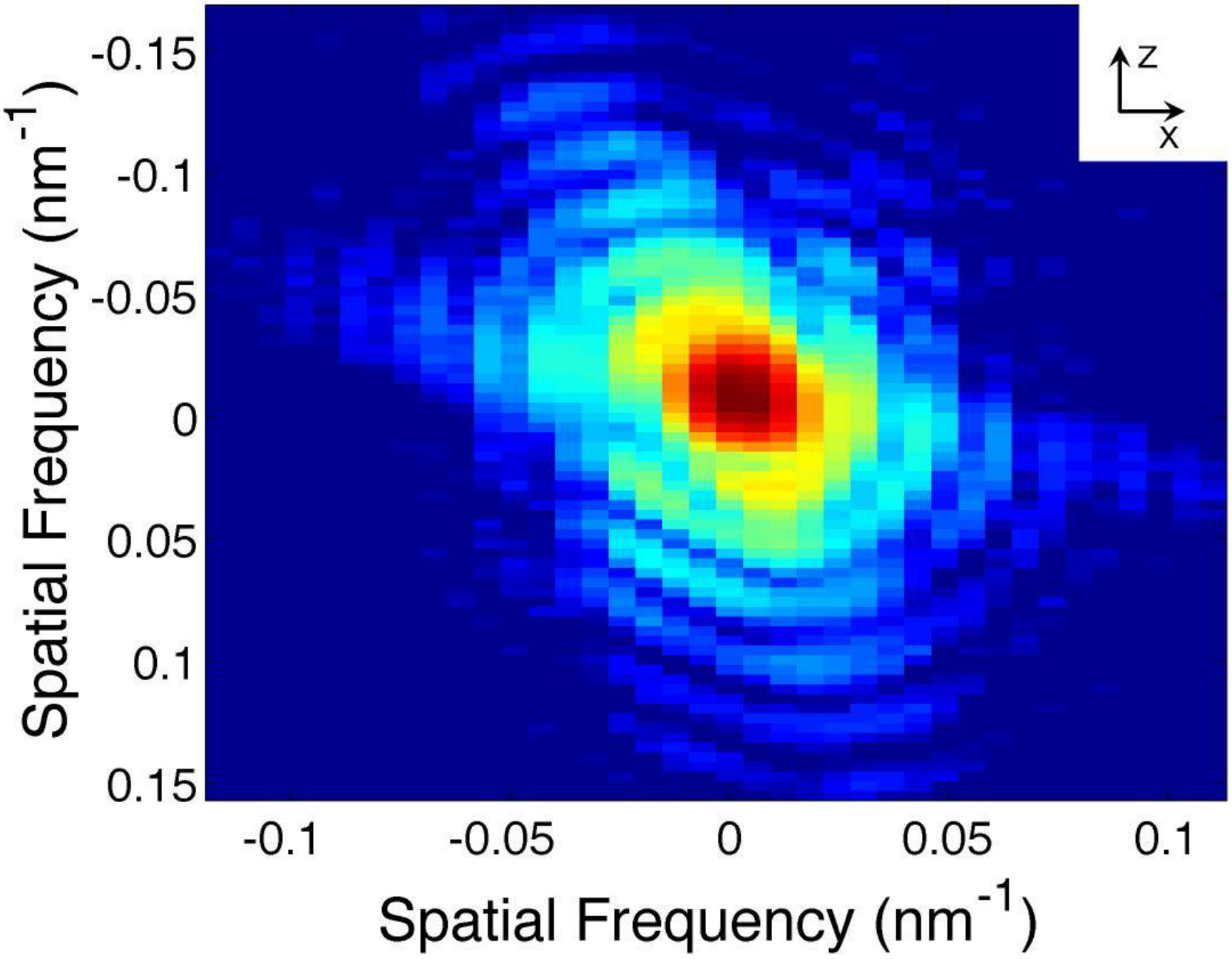
M. Pfeifer et at, Nature 442 63 (2006)

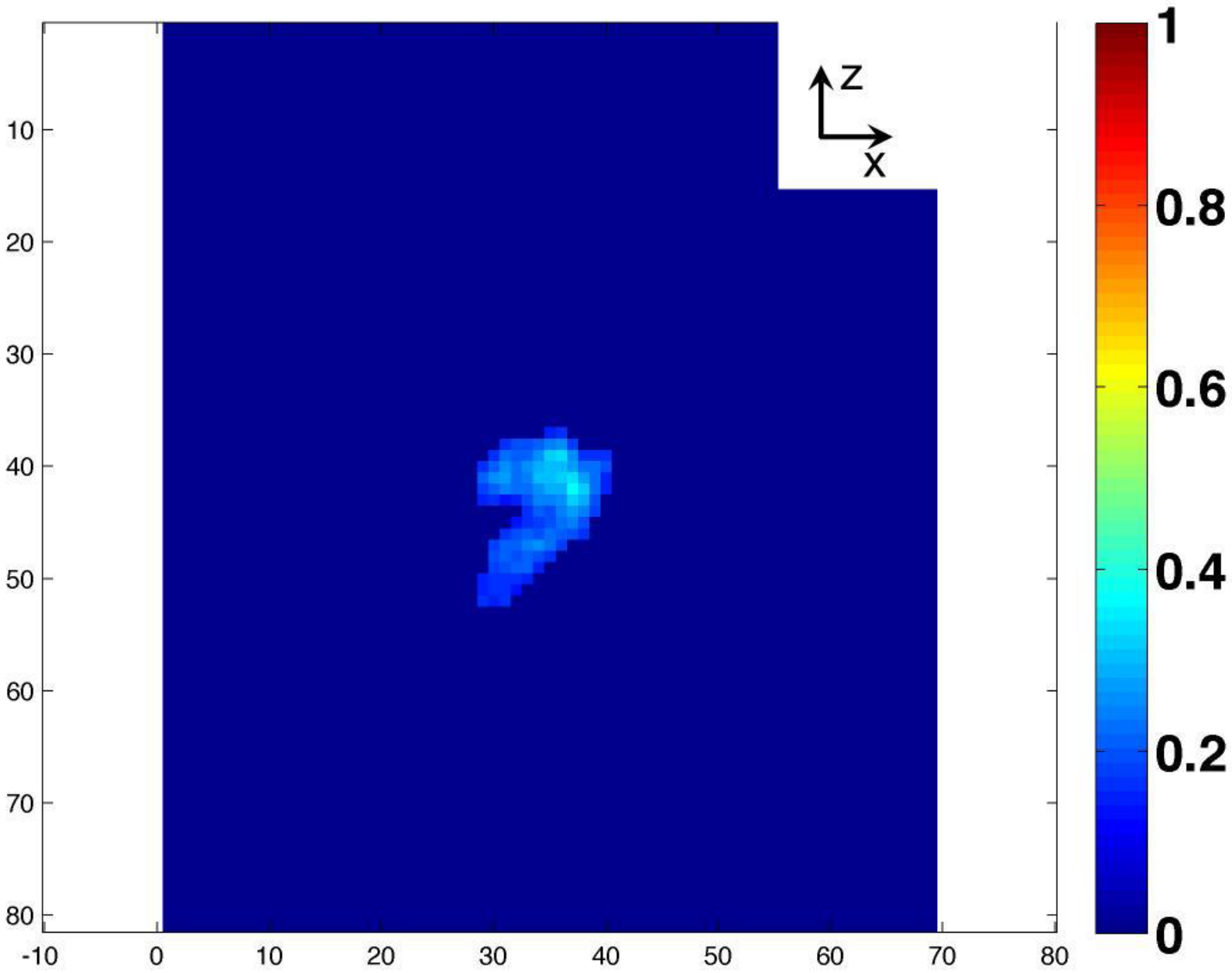


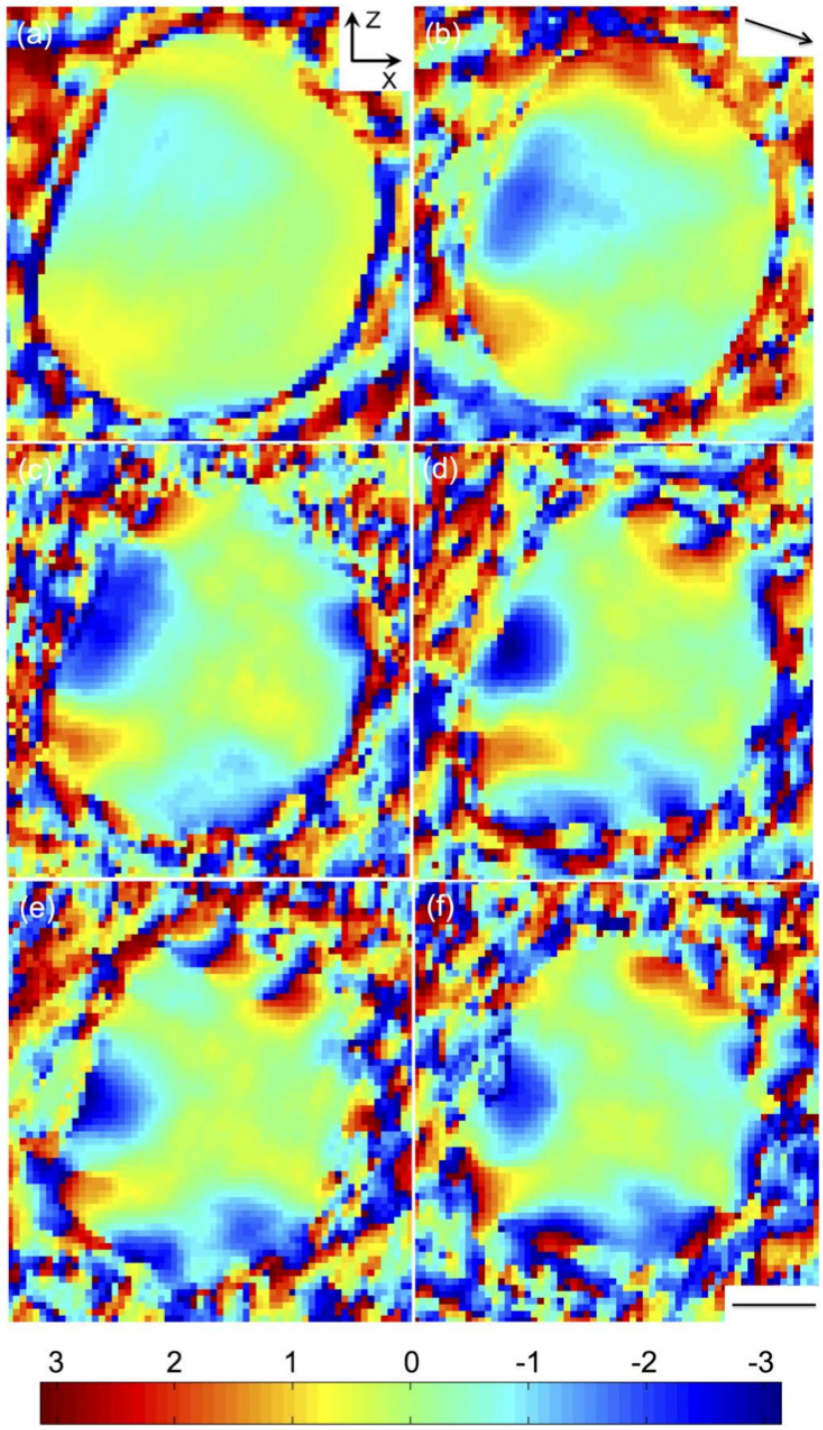
Copper Diffusion into Gold Nanocrystal

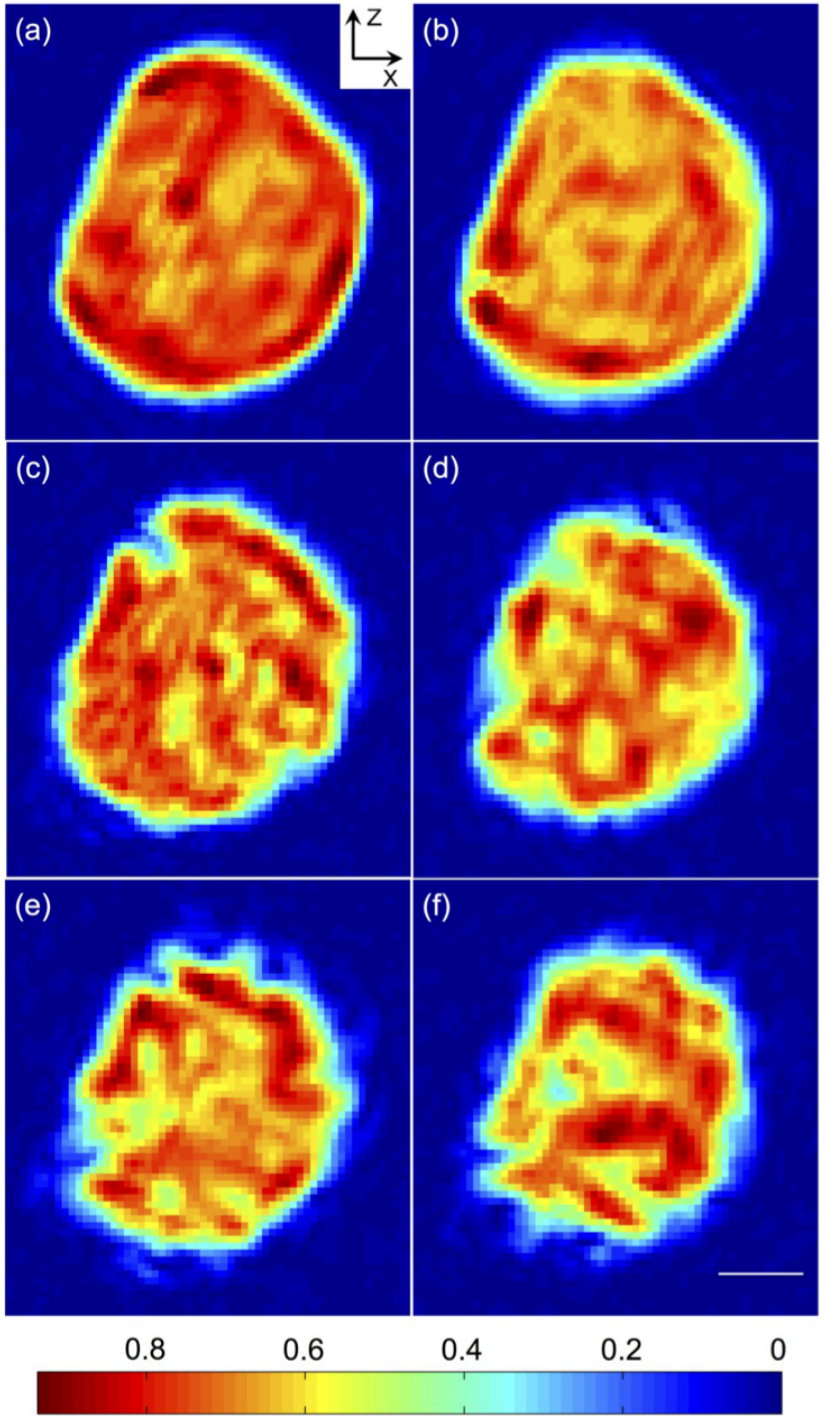
Gang Xiong, et al, Sci Rep 4 6765 (2013)




-0.15
-0.1
-0.05
0
0.05
0.1
0.15
Spatial Frequency (nm-1)
-0.1
-0.05
0
0.05
0.1
Spatial Frequency (nm-1)
z
x


Z
X
10
20
30
40
50
60
70
80
-10
0
10
20
30
40
50
60
70
80
1
0.8
0.6
0.4
0.2
0

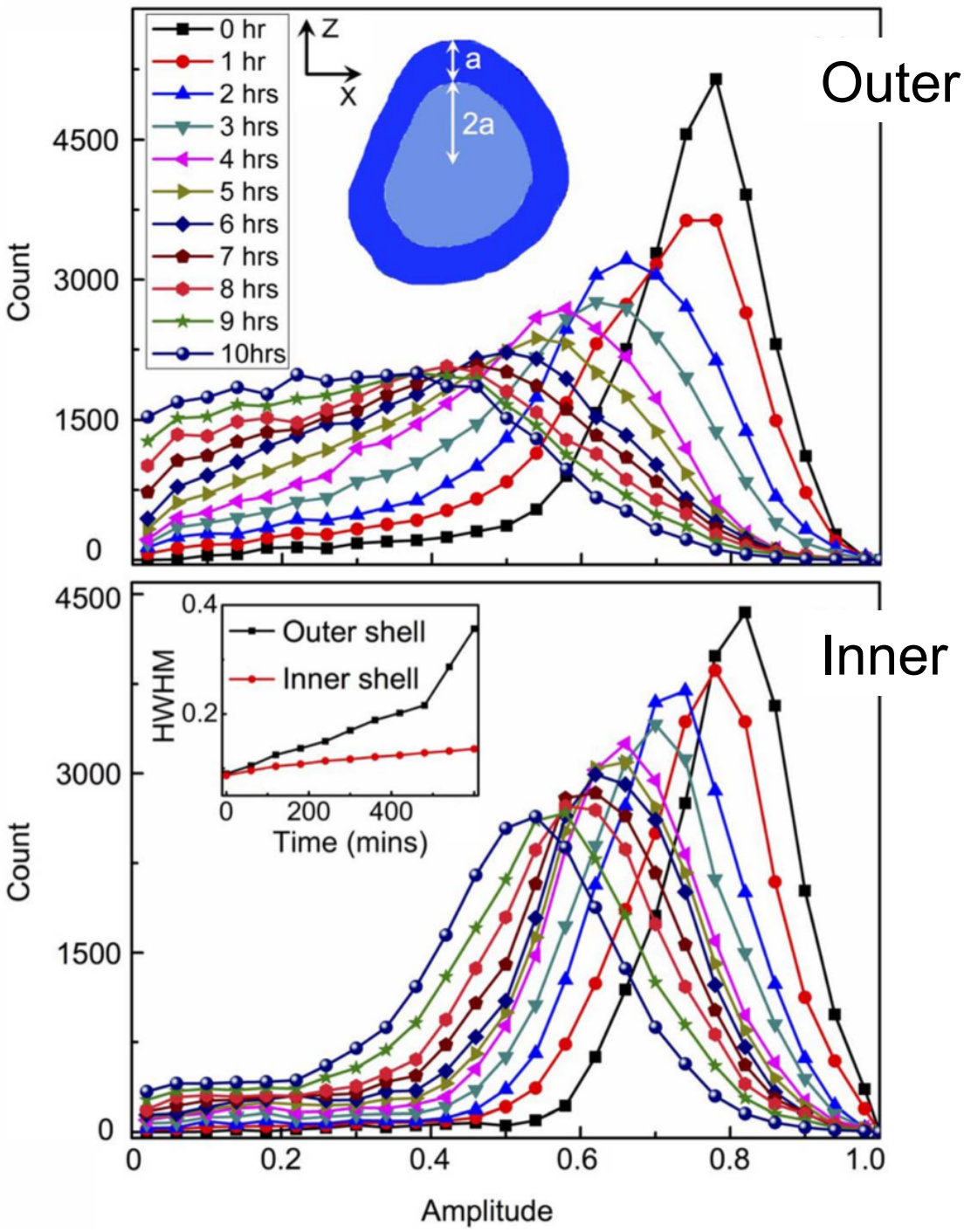

(a)
Z
X
(b)
(c)
(d)
(e)
(f)
3
2
1
0
-1
-2
-3


(a)
Z
X
(b)
(c)
(d)
(e)
(f)
0.8
0.6
0.4
0.2
0

Atomic Diffusion within Individual Gold Nanocrystal

Gang Xiong,
J. N. Clark,
C. Nicklin,
J. Rawle &
I. K. Robinson

Sci Rep **4** 6765 (2013)



Generations of Synchrotron

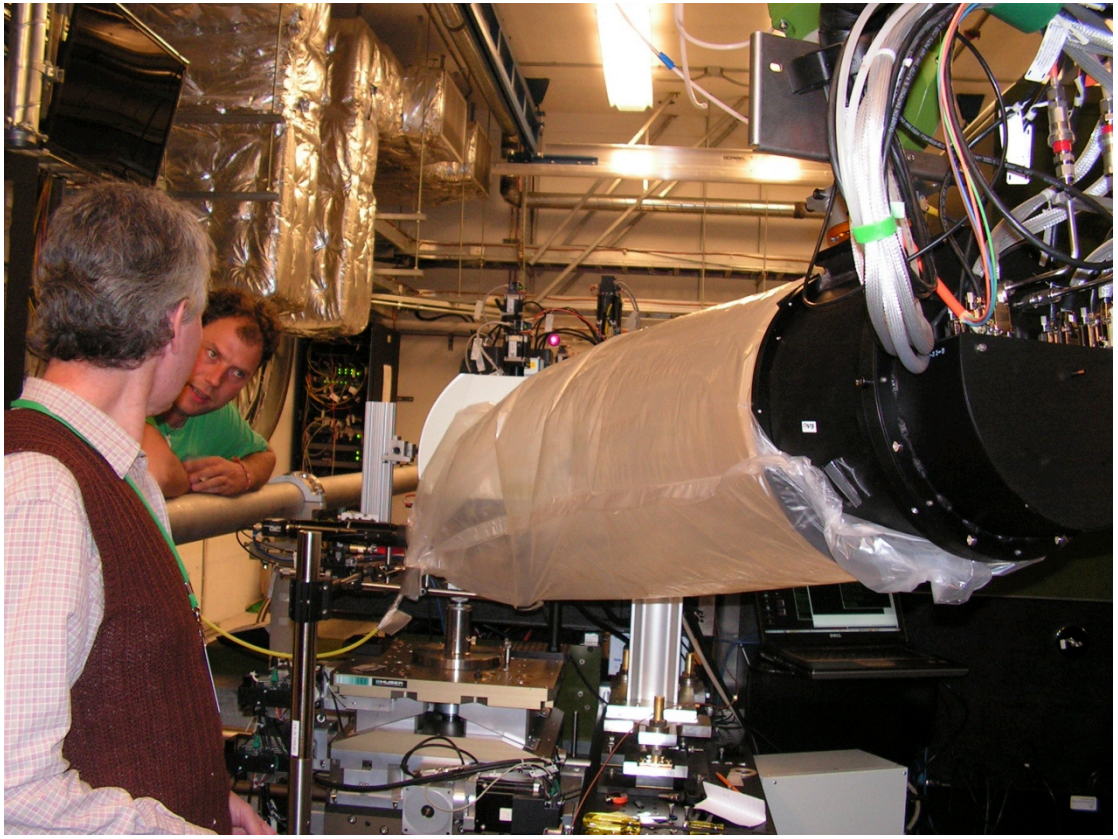
- 2nd: Storage Ring Dedicated to X-rays
  - NSLS Surface X-ray Diffraction
- 3rd: Undulators in "Straight Sections"
  - APS Coherent X-ray Diffraction
  - Diamond Light Source
- X-ray Free Electron Lasers
  - LCLS Ultrafast Coherent Diffraction

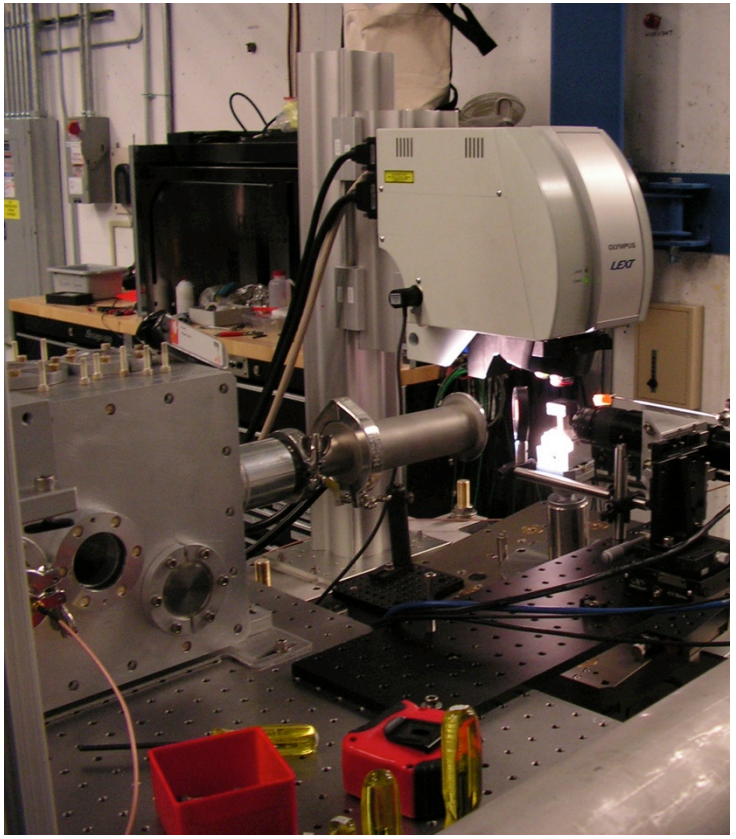
Linac Coherent Light Source

X-Ray Free-electron Laser, SLAC, Stanford, USA

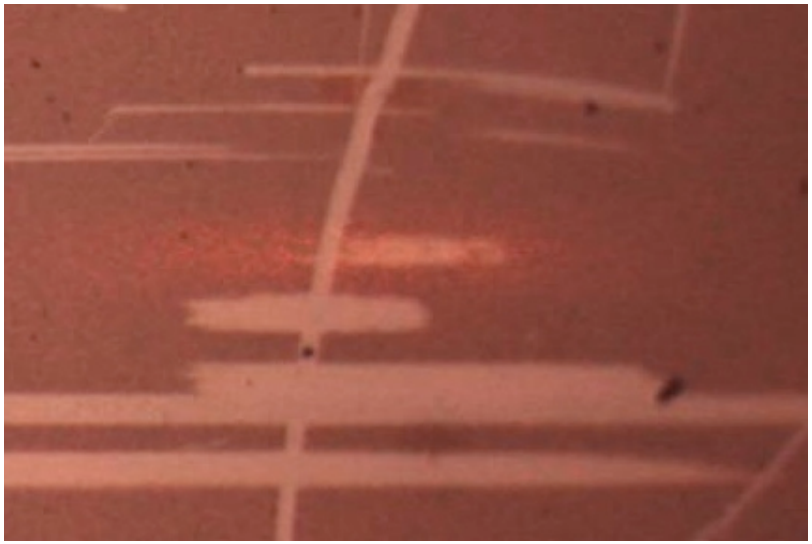
Pump-probe at LCLS (XPP)

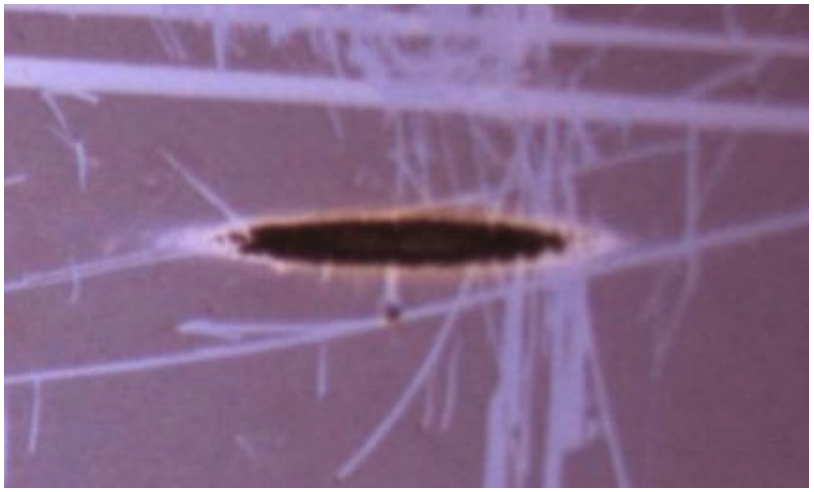
Justin Wark, Loren Beitra, Alexander Korsunsky, Ross Harder, David Fritz ,
Sebastien Boutet, Jesse Clark, Garth Williams, Brian Abbey, Andy Higginbotham,
Diling Zhu, Henrick Lemke, Mattieu Chollet, Marc Messerschmidt





Visible and Confocal microscopy





Pump-probe at LCLS (XPP)

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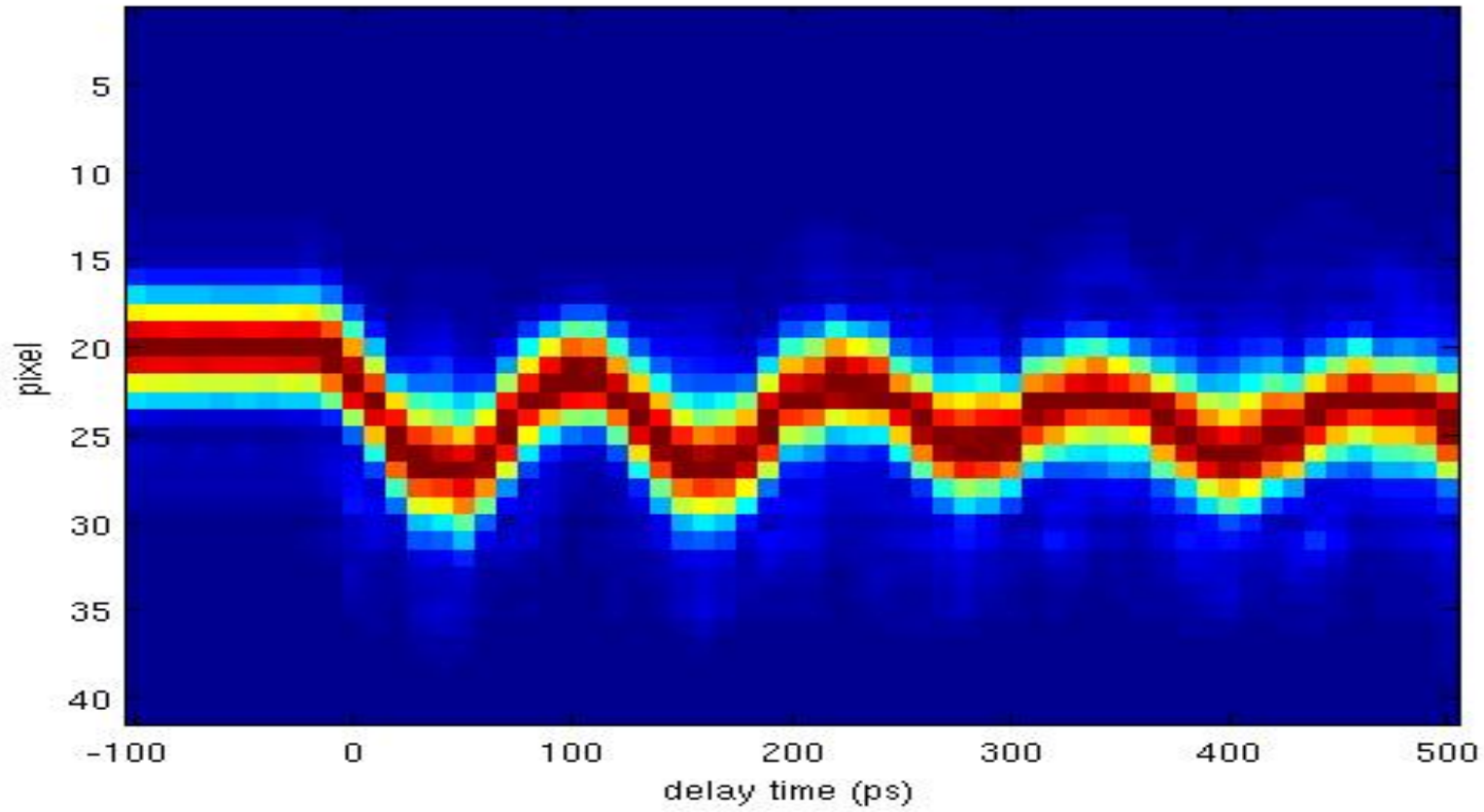
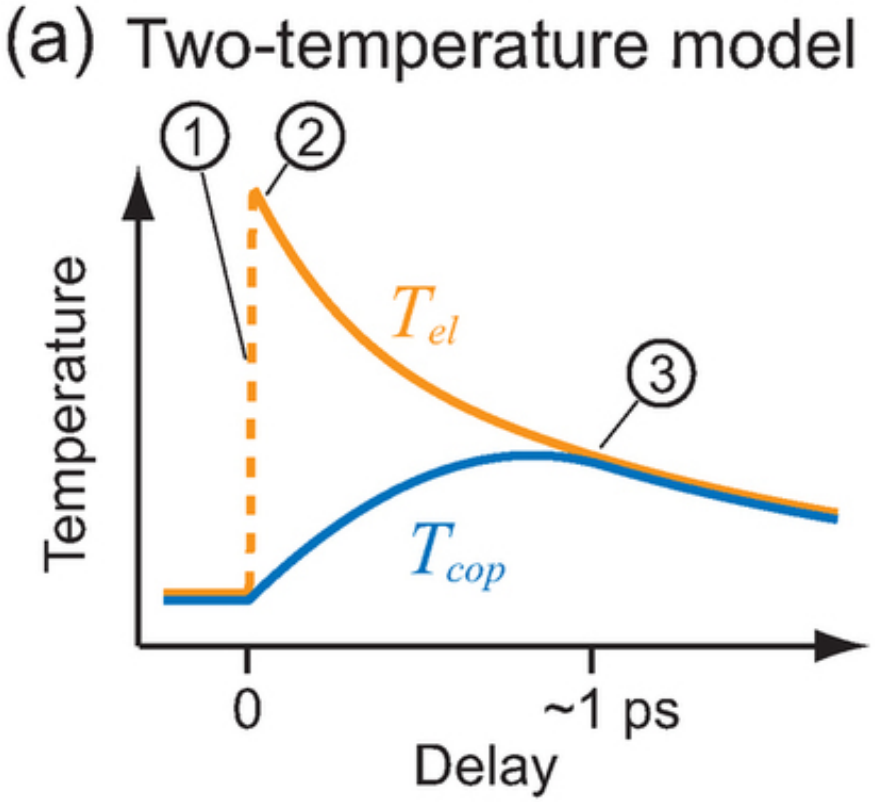
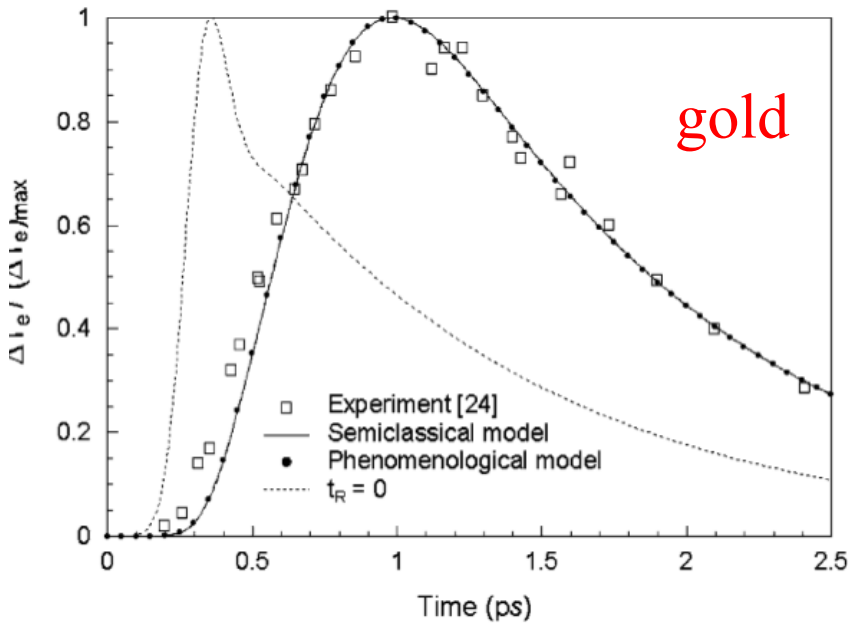


"Two-temperature" model

Y. Ishida et al, Nature Scientific Reports 1 64 (2011)

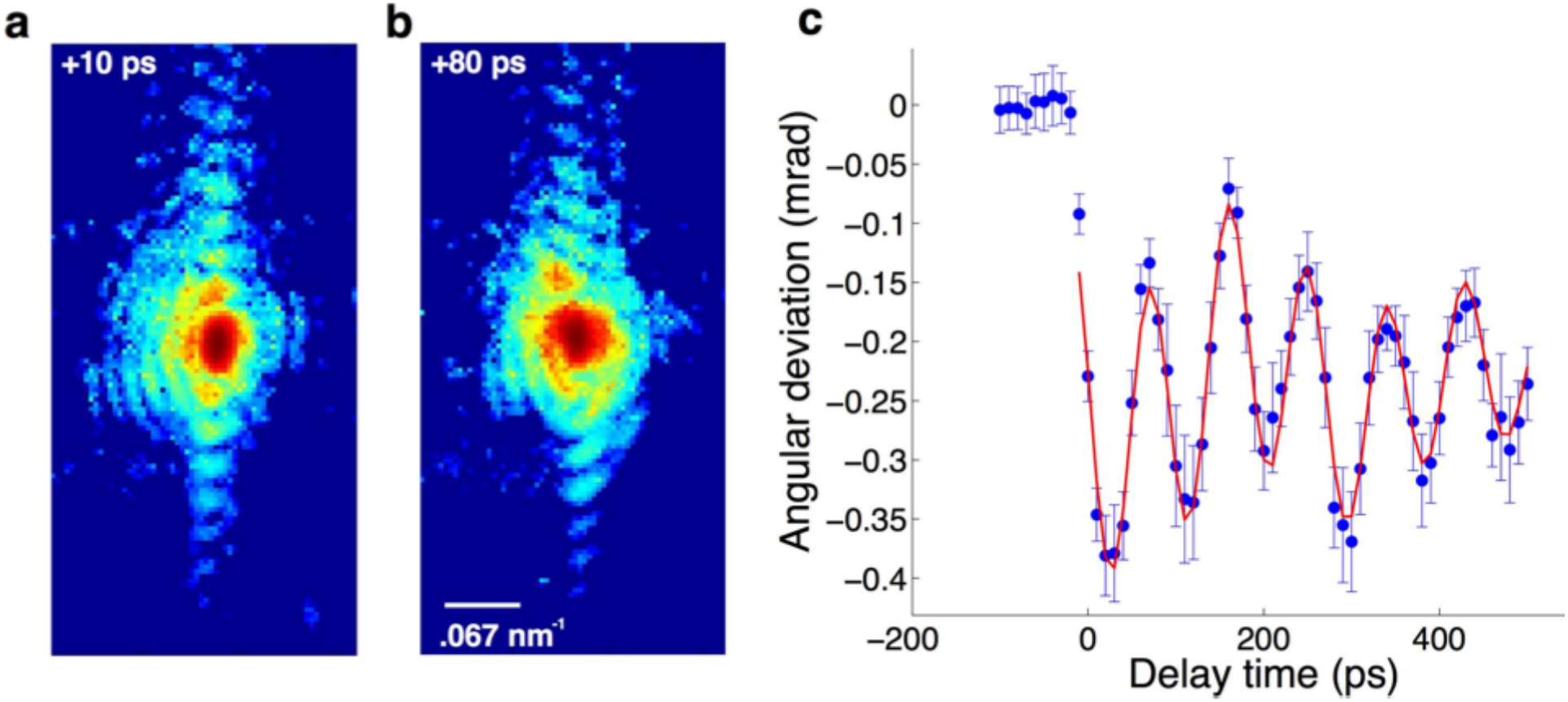
J.K. Chen et al, Int J. Heat Transfer 49 307 (2006)





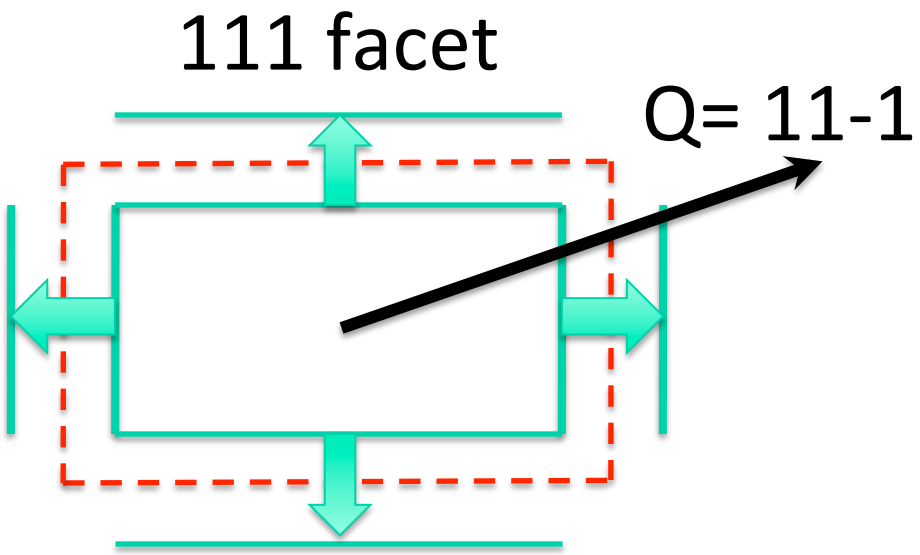
ig. 2. Comparison of the change in electron temperature at ıe front surface of an 80-nm gold film irradiated by a 2.8 mJ/ n^2, 800 nm, 150-fs laser pulse.

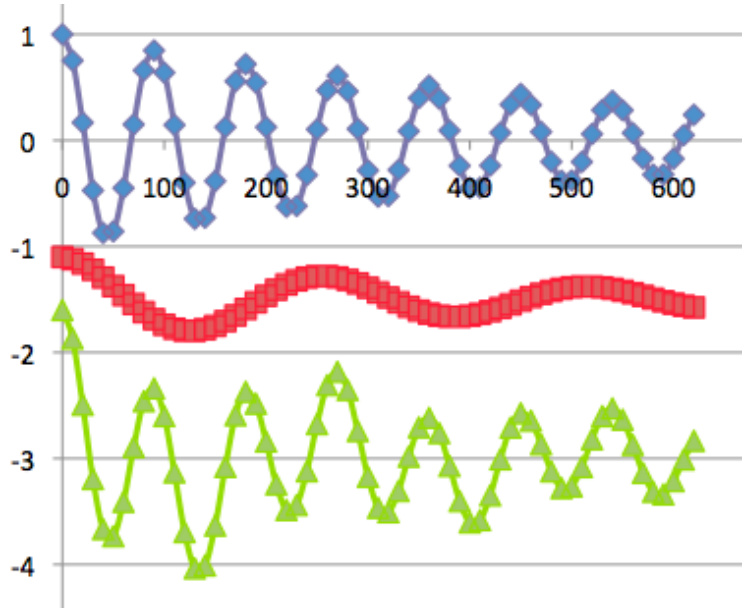
Time resolved Bragg peak position



Two Normal Modes of Vibration

$$S(\tau) = \sum_{n=1}^{N} A_n \exp\left[-(\tau/\tau_{d,n})^2\right] \cos(\omega_n \tau + \varphi_{0,n})$$





T_1 = 90ps h_1 = 145nm c_S = 3240 m/s

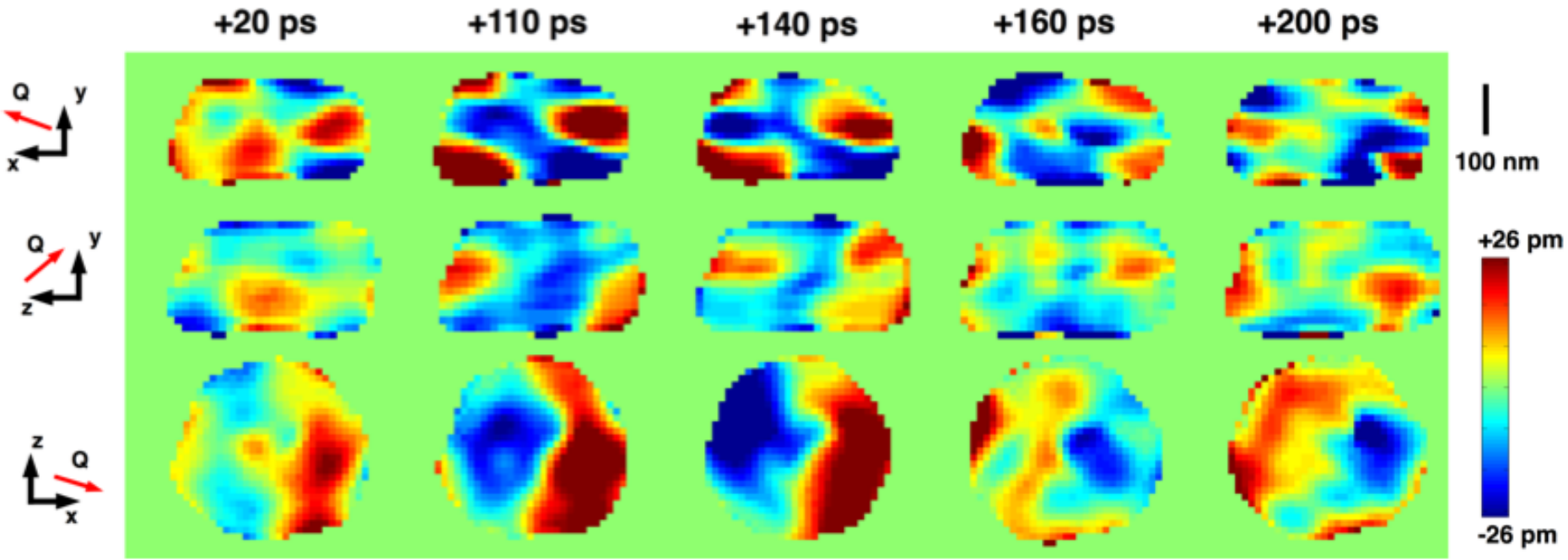
T_2 = 259ps h_2 = 420nm

Dynamic imaging of displacements

CDI inversion of 3D diffraction patterns

1000 frames averaged at each point of rocking curve

Jesse Clark et al, Science 341 56 (2013)

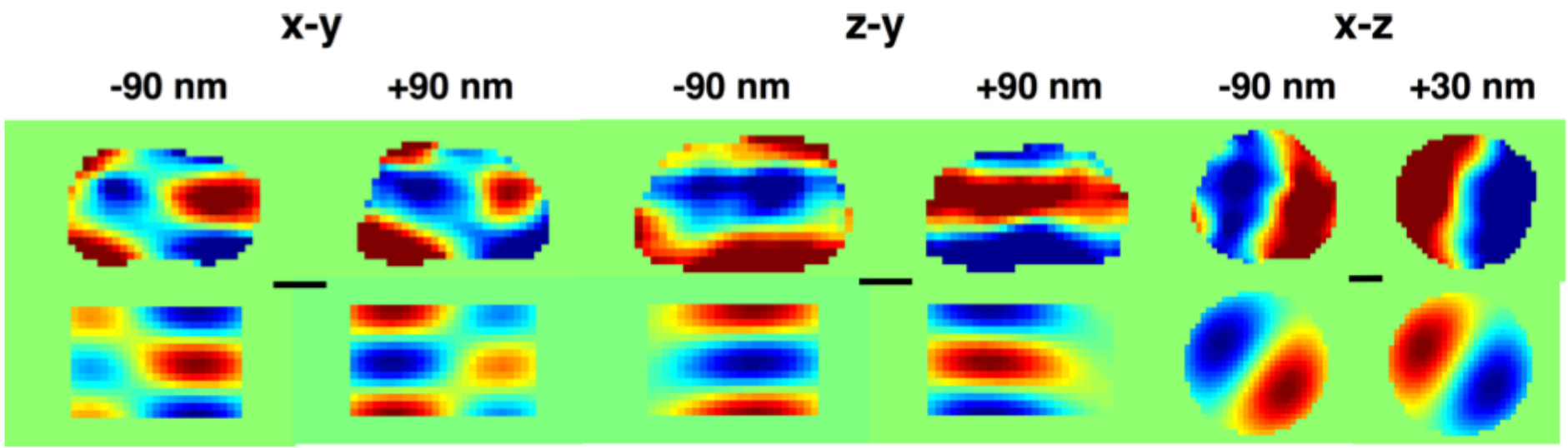


Dynamic imaging of displacements

CDI inversion of 3D diffraction patterns

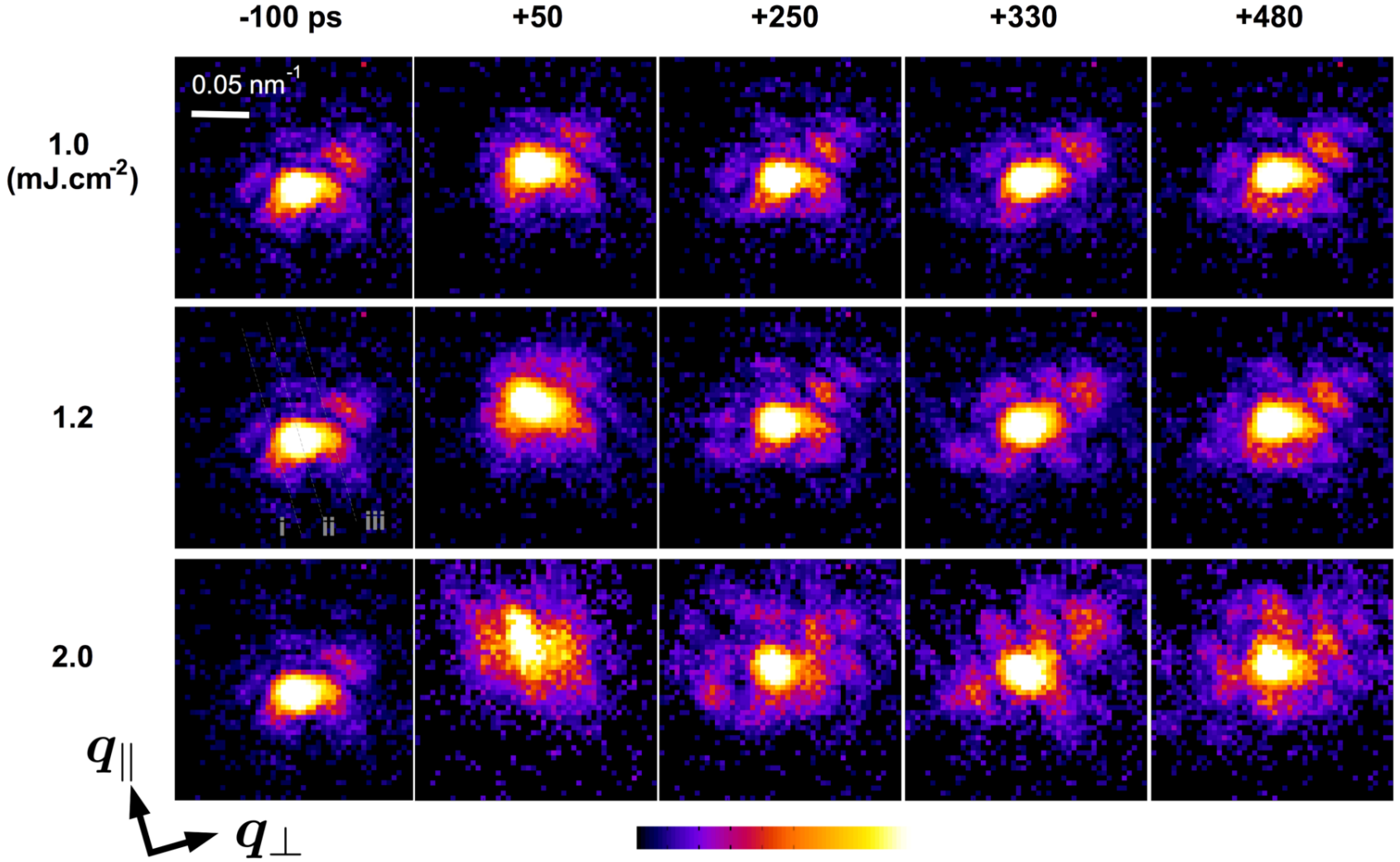
Comparison with (1,1) normal mode of cylinder

Jesse Clark et al, Science 341 56 (2013)



Dependence on Laser Fluence

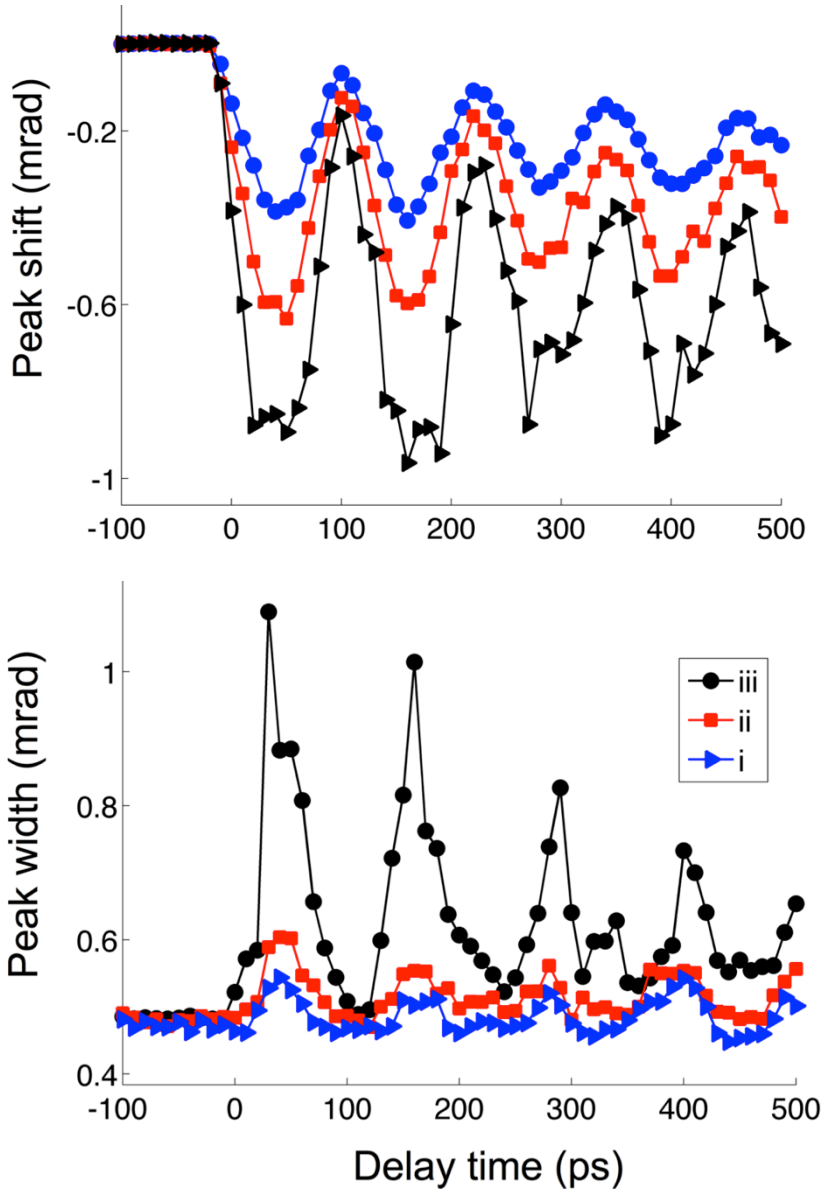
Jesse Clark et al, to be published



Dependence on Laser Fluence

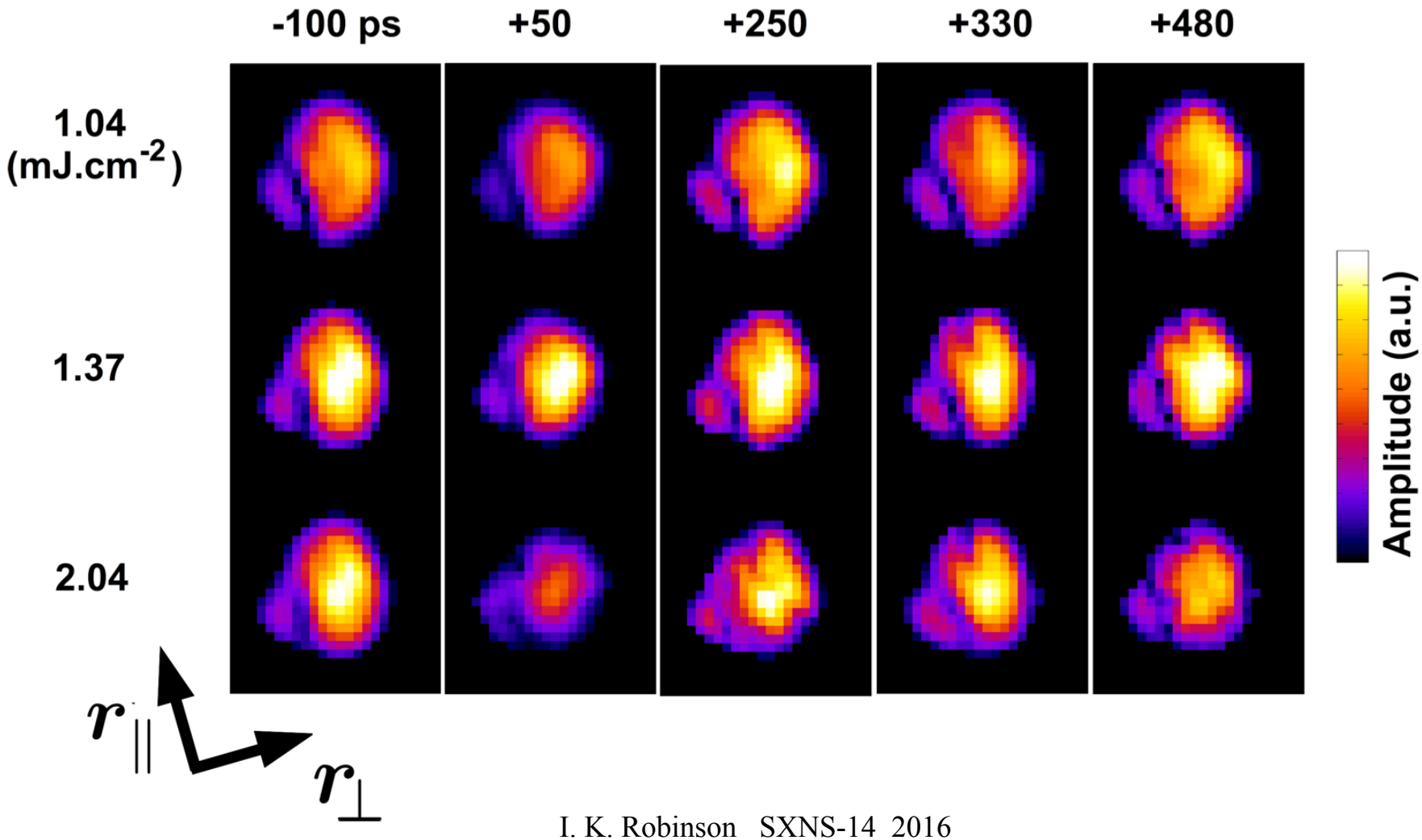
Jesse Clark et al, to be published

1.0 mJ cm^{-2}
1.2 mJ cm^{-2}
2.0 mJ cm^{-2}



Dependence on Laser Fluence

Jesse Clark et al, to be published



Surface X-ray Diffraction pioneers

Peter Eisenberger	Bell Labs
Paul Fuoss	Argonne
Peter Bennett	Arizona
Doon Gibbs	Brookhaven
Ben Ocko	Brookhaven
Peter Eng	Chicago
Robert Feidenhans'l	Copenhagen
Jens Als-Nielsen	Copenhagen
Jakob Bohr	Copenhagen
Mike Altman	HKUST
Elias Vlieg	Nijmegen
Sunil Sinha	San Diego
Don Walko	Urbana
Franz Himpsel	Wisconsin

Coherent X-ray Diffraction

Jesse Clark
Ross Harder
Gang Xiong
Garth Williams
Sebastien Boutet
Mark Pfeifer
Loren Beitra
Meng Liang
Xiaowen Shi
Steven Leake
Richard Bean
John Pitney
Ivan Vartaniants

Justin Wark (Oxford)
Alexander Korsunsky
Andy Higginbotham

Chris Nicklin (Diamond)
Jonathan Rawle
Brian Abbey (Melbourne)

David Fritz (SLAC)
Diling Zhu
Henrick Lemke
Mattieu Chollet
Marc Messerschmidt

Summary

- Crystal Truncation Rods
- Au Quantum Wires on Stepped Si
- Facetting of Cu(115) seen in CTRs
- Silicon (111) 7x7
- Nanocrystal: paradigm of Surface/Bulk
- Cu diffusion into Au
- Ultrafast snapshots of Vibrating Crystals