

# Interactions of X-rays with Matter - Scattering

NX School 2026, Argonne National Lab  
14 September 2026

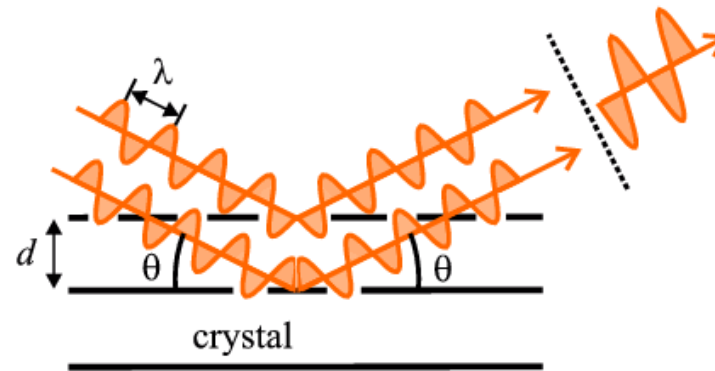
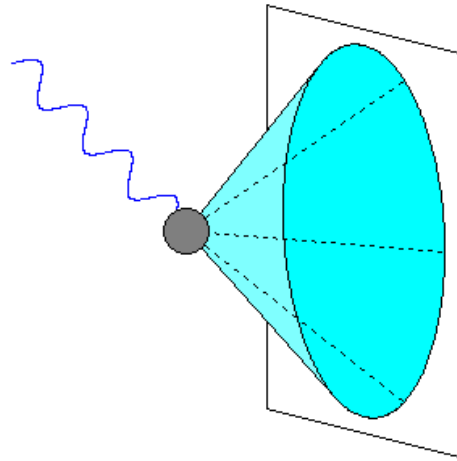
Ian Robinson

London Centre of Nanotechnology  
University College London

# X-ray Scattering Outline

- Scattering from Matter
- Powder diffraction methods “XRD”
  - Cora Lind-Kovacs’ lecture
- Surface diffraction and surface structure
  - Paul Miceli’s lecture
- Extension to Coherent X-ray Diffraction
  - Xiaojing Huang’s lecture
- XFEL study of thin film melting
  - Meng Liang’s lecture

# X-ray Scattering vs Diffraction



$$2d \sin \theta = \lambda$$

Bragg law

**Elements of Modern X-ray Physics**  
**Second Edition**

**Chapter 5**  
**Kinematical scattering II: crystalline order**

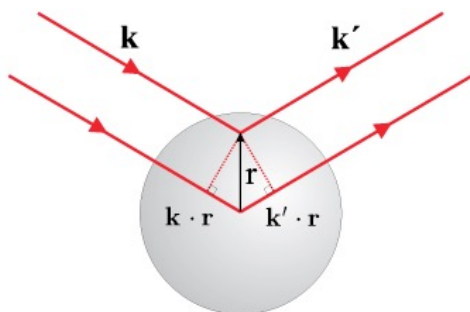
Jens Als-Nielsen and Des McMorrow

John Wiley & Sons Ltd

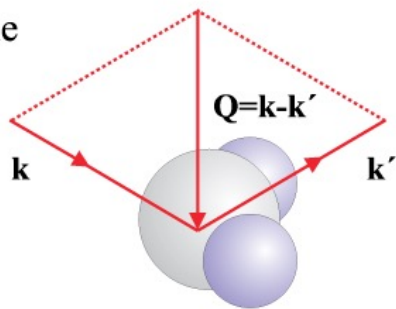
Chinese translation by Donglai Feng <dlfeng@fudan.edu.cn>

# Assemblies of Atoms

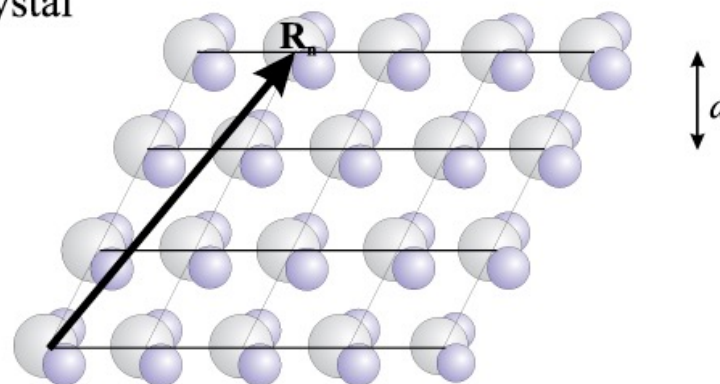
(a) One atom



(b) One molecule

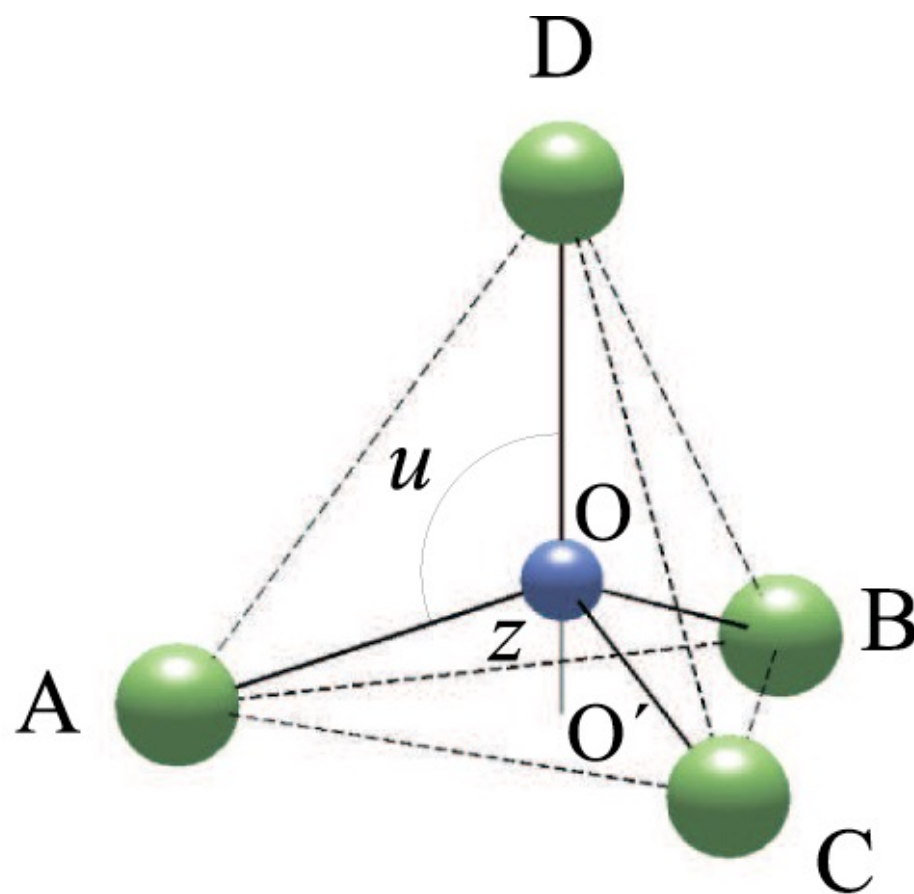


(c) A crystal

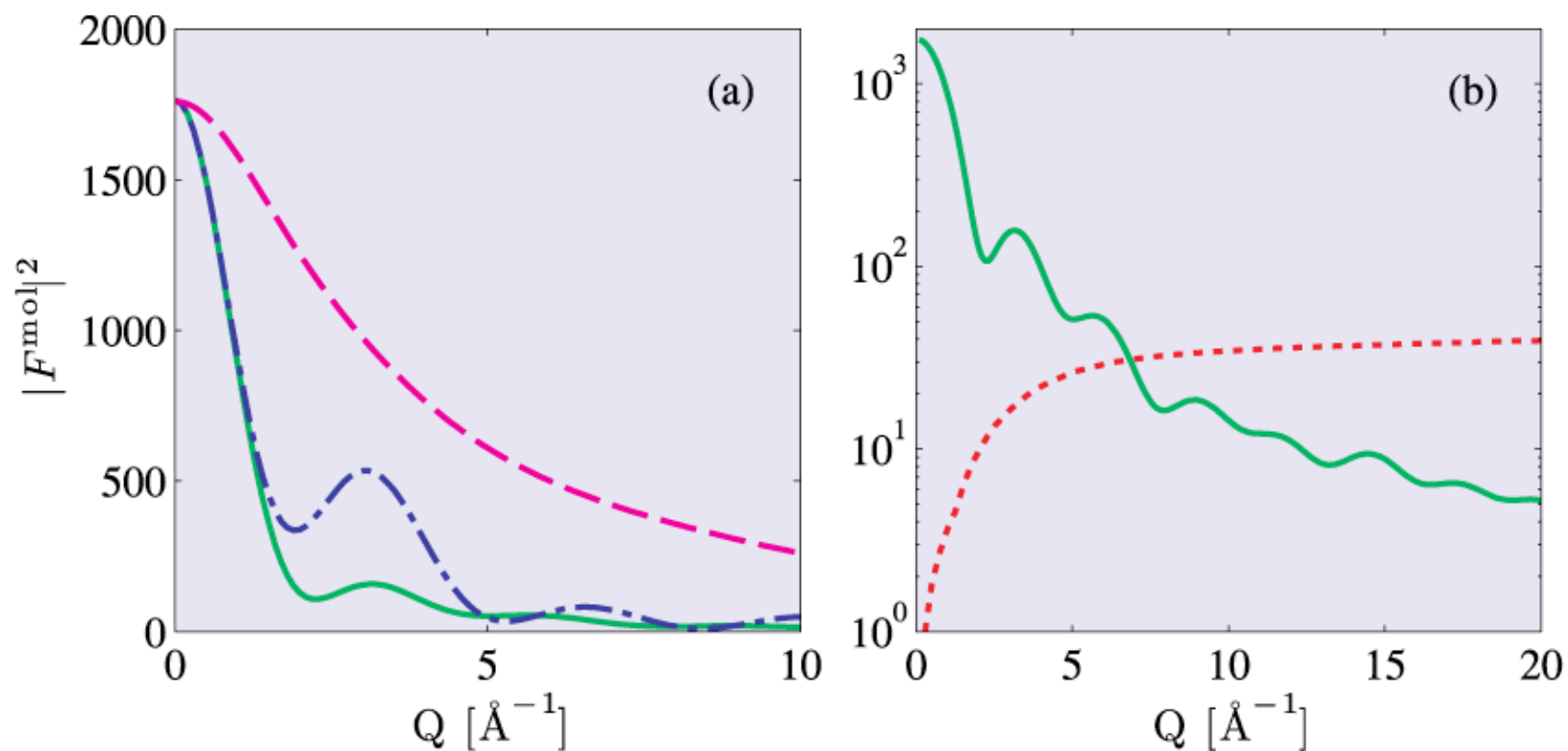


**Fig. 1.7** (a) Scattering from an atom. An X-ray with a wavevector  $\mathbf{k}$  scatters from an atom to the direction specified by  $\mathbf{k}'$ . The scattering is assumed to be elastic, i.e.  $|\mathbf{k}| = |\mathbf{k}'| = 2\pi/\lambda$ . The difference in phase between a wave scattered at the origin and one at a position  $\mathbf{r}$  is  $(\mathbf{k} - \mathbf{k}') \cdot \mathbf{r} = \mathbf{Q} \cdot \mathbf{r}$ . This defines the wavevector transfer  $\mathbf{Q}$ . (b) The scattering from a molecule. Here the scattering triangle is shown which relates  $\mathbf{k}$ ,  $\mathbf{k}'$  and  $\mathbf{Q}$ . (c) Scattering from a molecular crystal. The molecules are organized on a lattice with position vectors  $\mathbf{R}_n$  and a lattice plane spacing of  $d$ .

## Molecular scattering

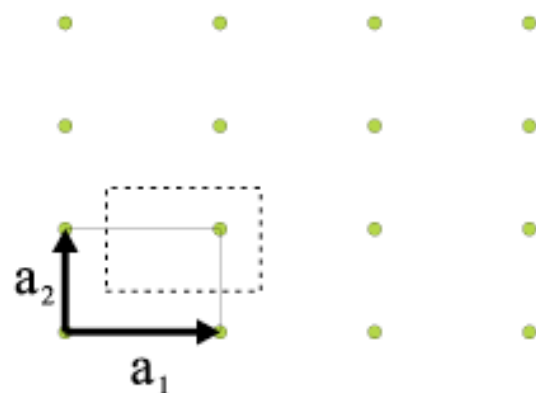


**Fig. 4.6** The  $\text{CF}_4$  molecule. The C-F bond length is  $1.38 \text{ \AA}$ , and the geometry is such that the ratio of the length of OD to  $\text{OO}'$  is 3:1.

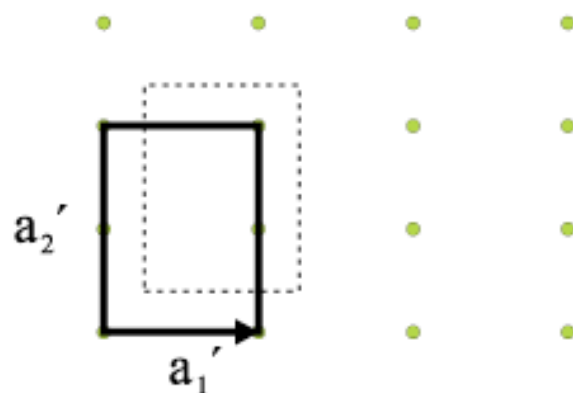


**Fig. 4.7** The calculated molecular structure factor squared of the  $\text{CF}_4$  molecule. (a) Dashed-dotted blue line is from Eq. (4.13); solid green line is calculated from a spherical average of the structure factor (Eq. (4.14)); magenta line is the square of the form factor of atomic molybdenum which has the same number of electrons as the  $\text{CF}_4$  molecule. (b) Solid green line is the spherically averaged structure factor of  $\text{CF}_4$ ; red dashed line is the Compton scattering calculated from the information given in the *International Tables of Crystallography*.

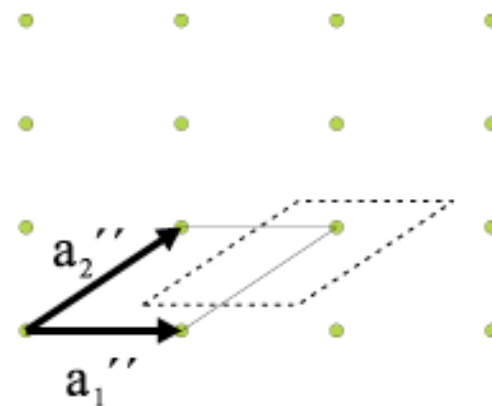
(a) primitive



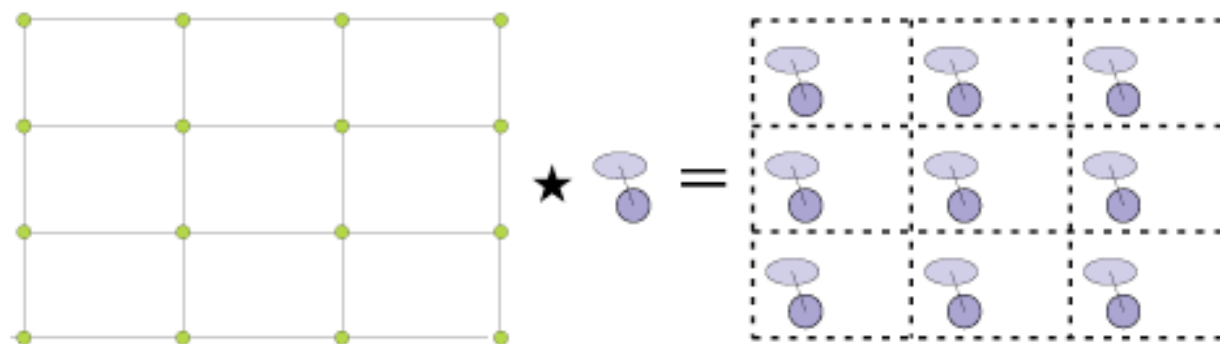
(b) non-primitive



(c) non-conventional



(d) Crystal=lattice  $\star$  basis

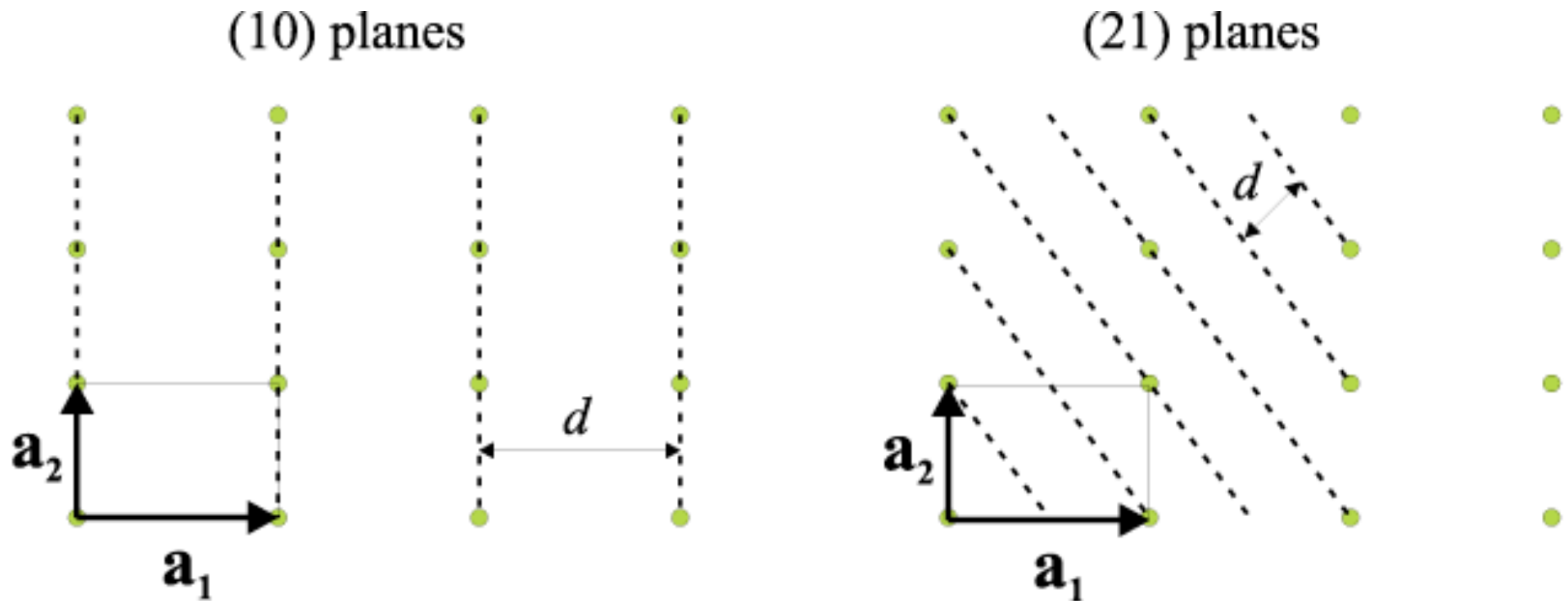


Lattice

Basis

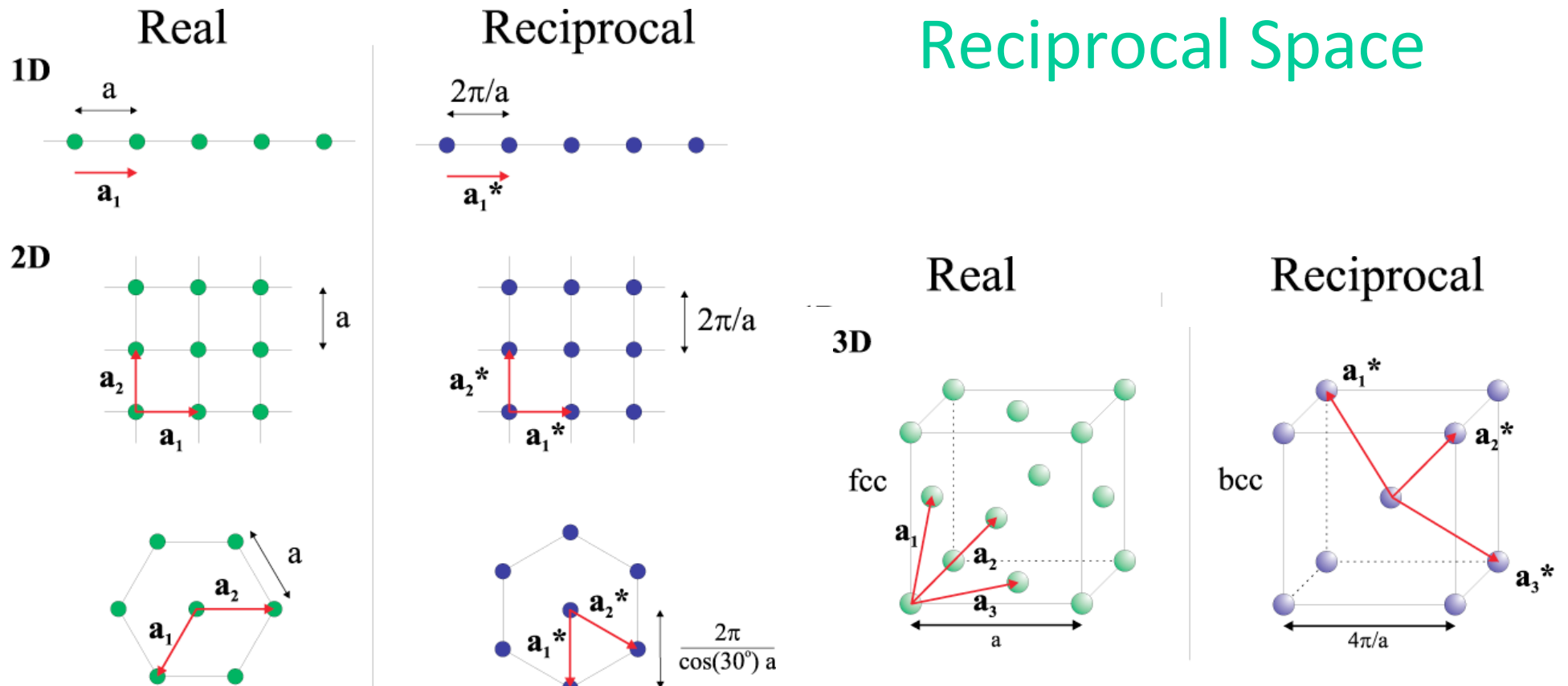
Crystal

# Miller Indices (hkl)



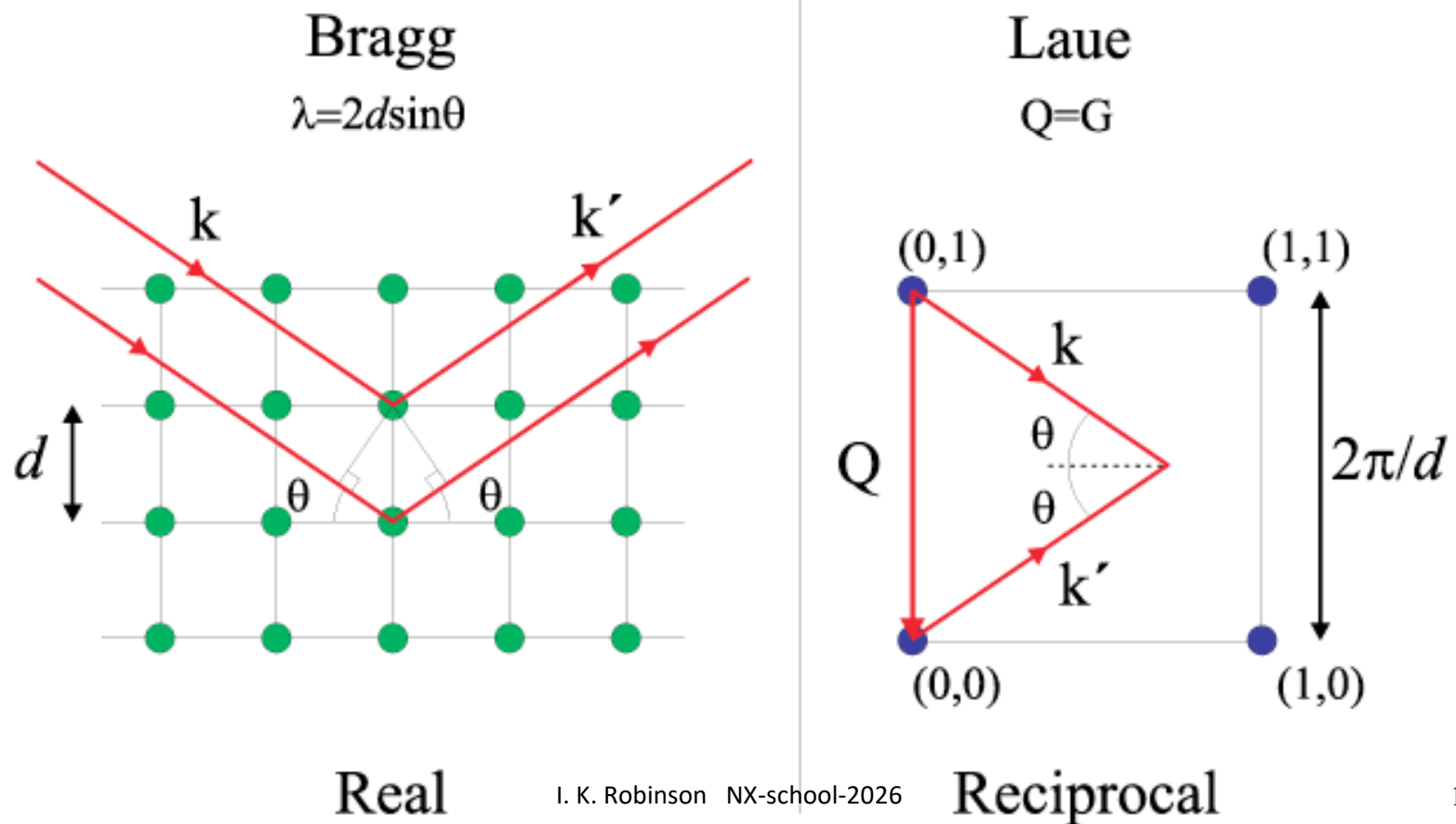
**Fig. 5.2** Lattice planes and Miller indices for the 2D rectangular lattice: (a) the (10) planes; (b) the (21) planes. In both cases the  $d$  spacing of the planes is indicated.

# Reciprocal Space

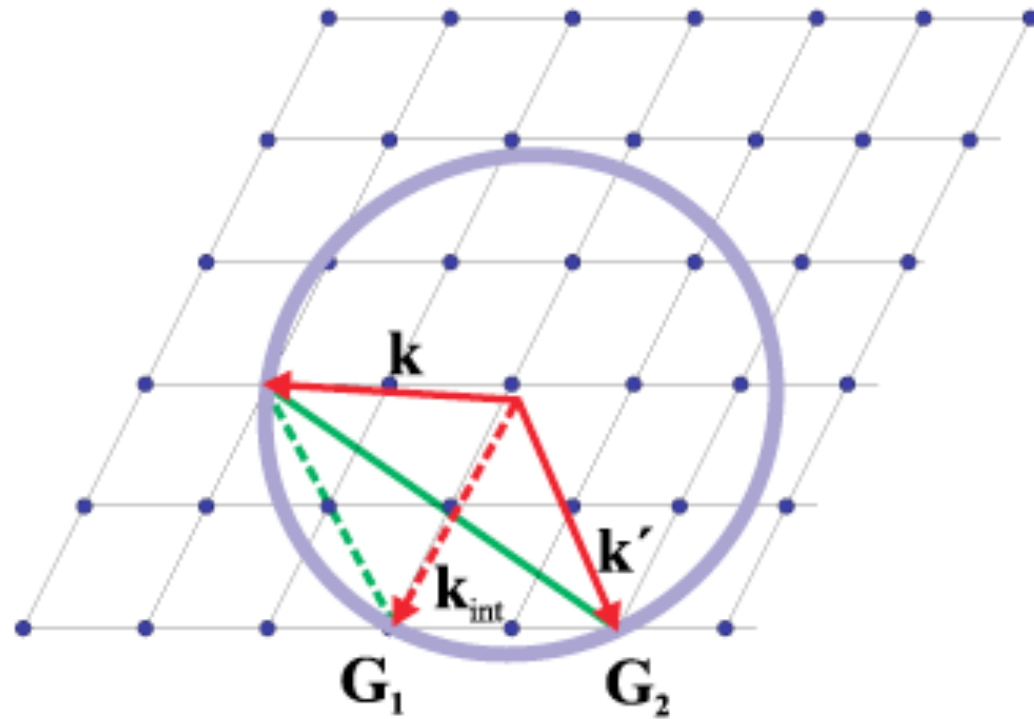


**Fig. 5.3** Example of the construction of reciprocal lattices in one, two, and three dimensions.

# Equivalence of Bragg and Laue geometries

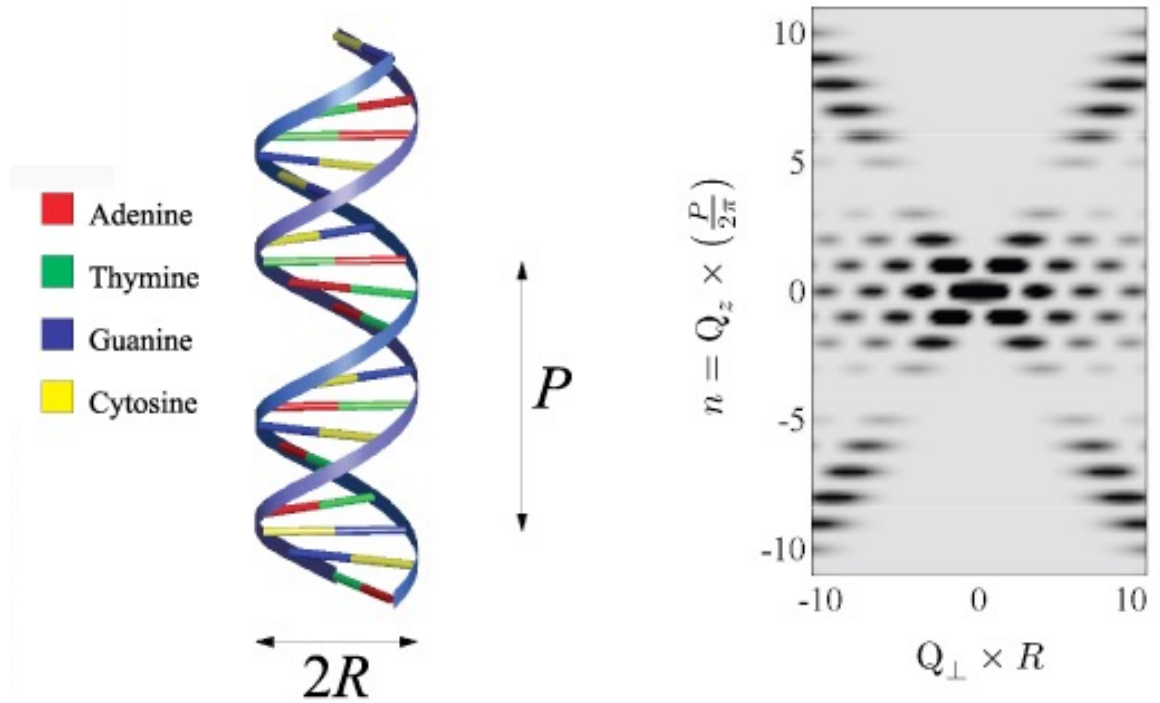
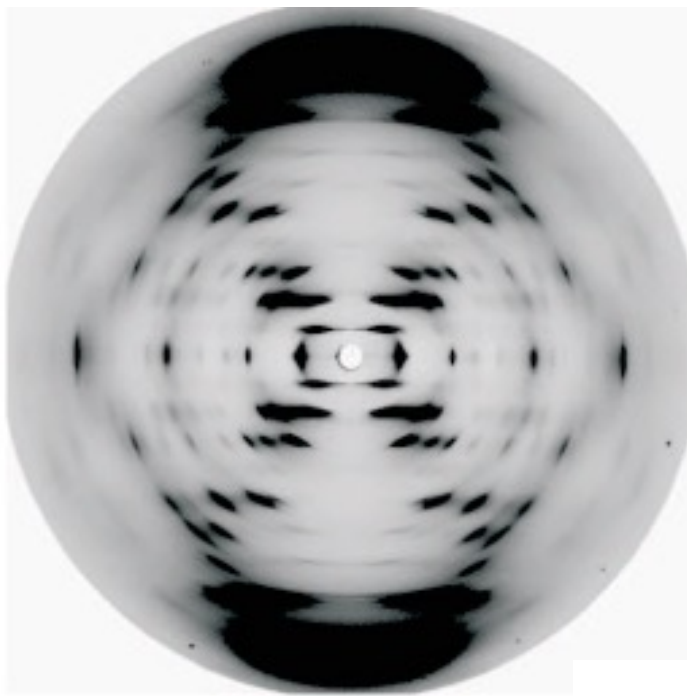


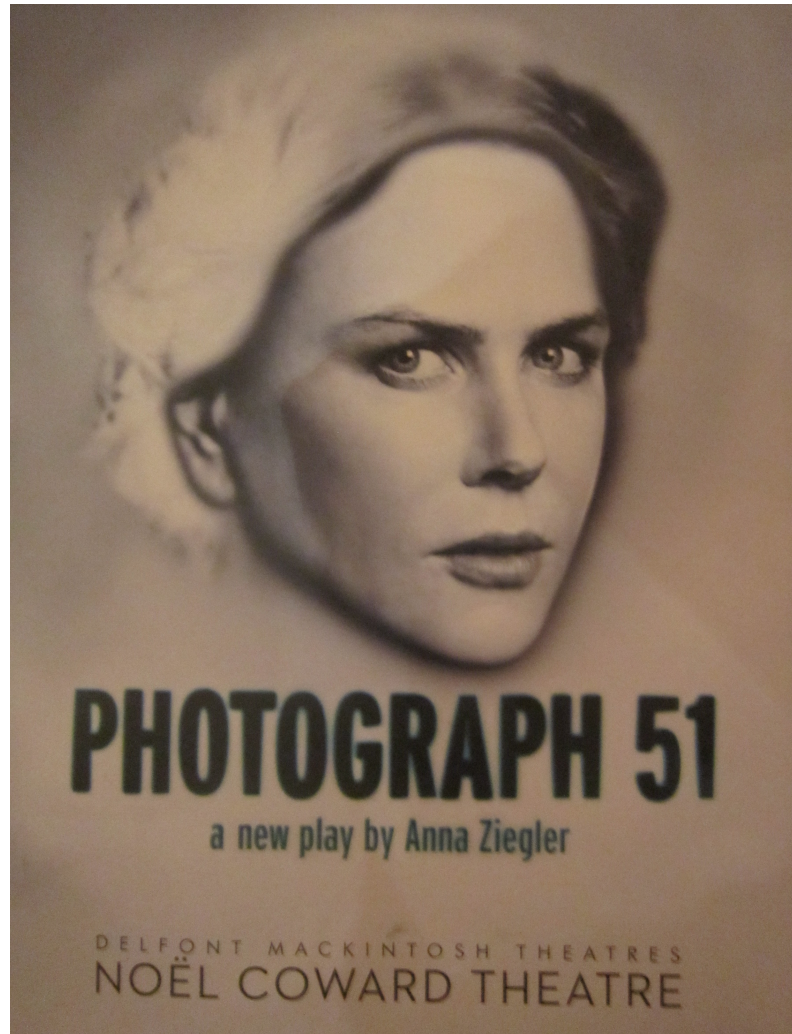
# Ewald sphere construction (2-beam)



# “Photograph 51” of DNA

Rosalind Franklin, Francis Crick, James Watson (1953)

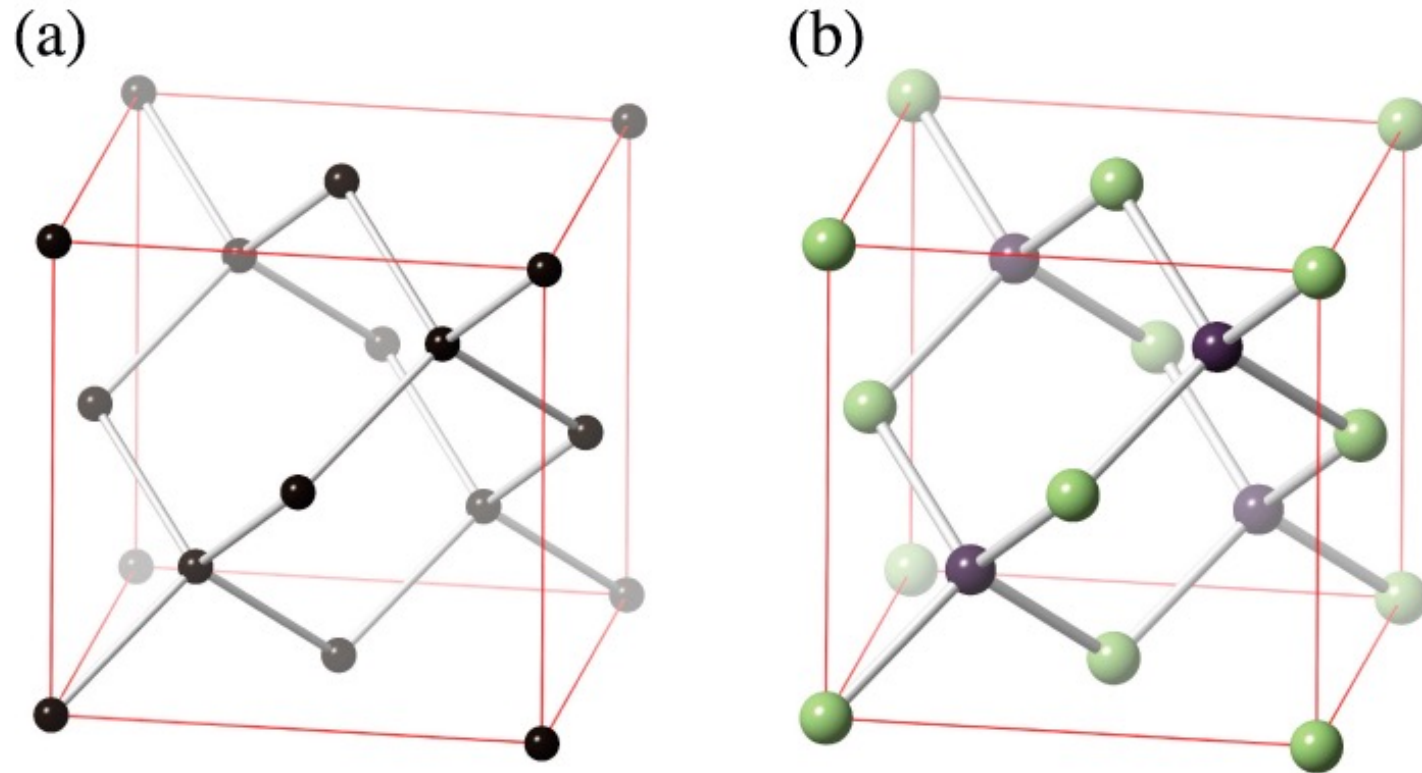




Nicole  
Kidman  
as  
Rosalind  
Franklin  
2015

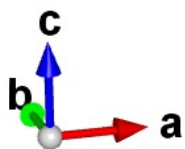
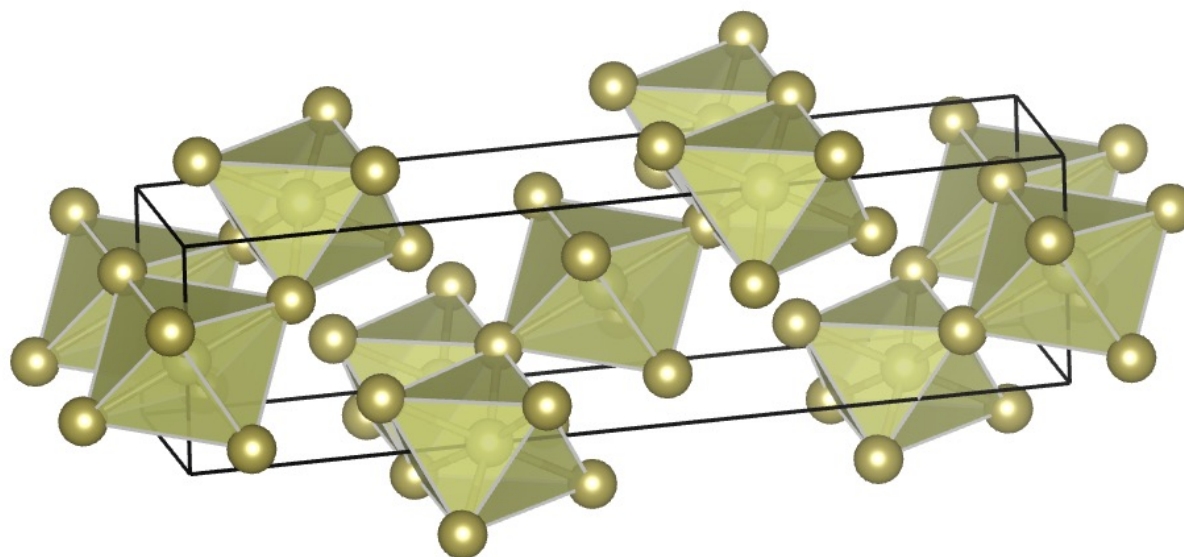
2026

14

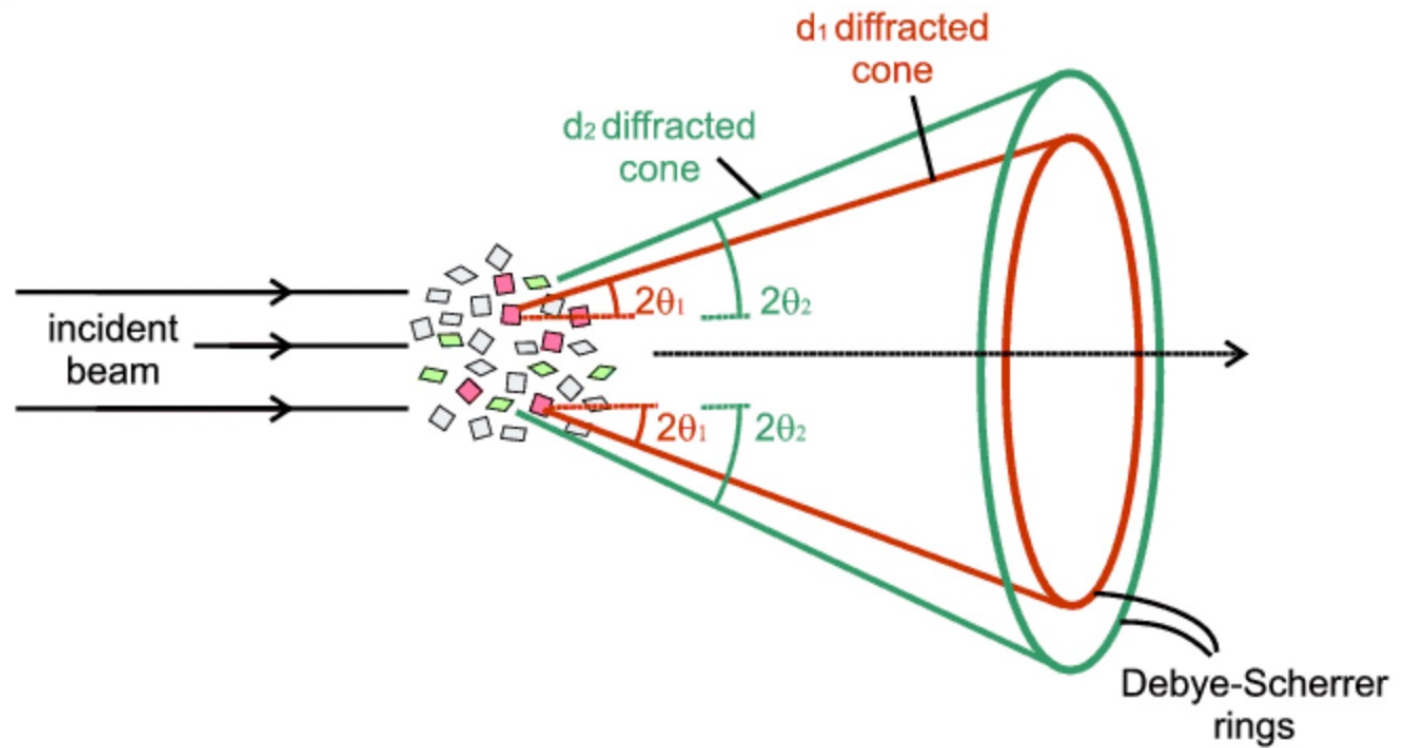


**Fig. 5.5** The diamond lattice (a) can be formed from two inter-penetrating *fcc* lattices displaced by  $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})$  with respect to each other. For the zinc sulfide (also known as zinc blende) structure (b) the two lattices are occupied by different types of atom.

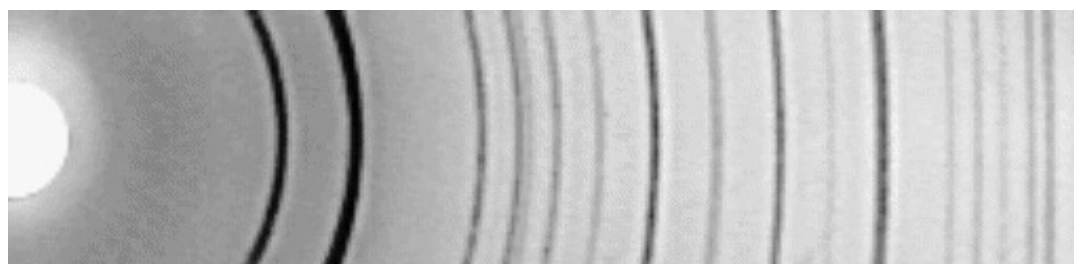
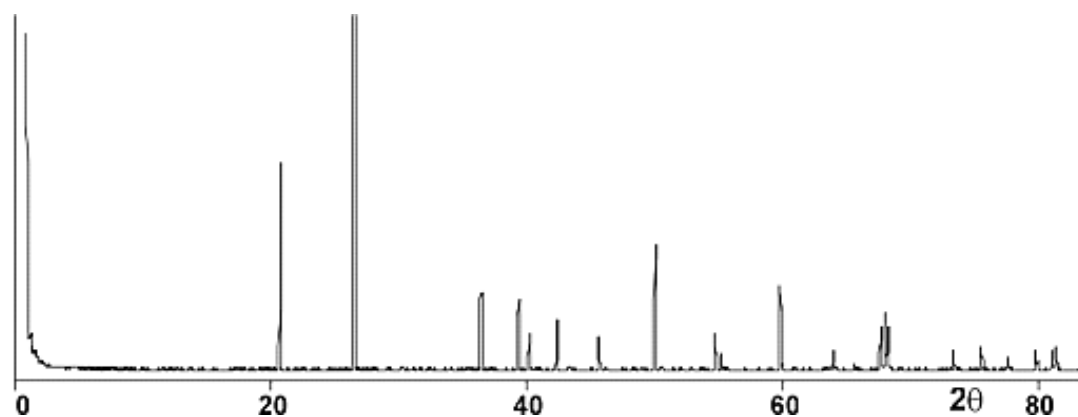
# VESTA software: IrTe<sub>2</sub>



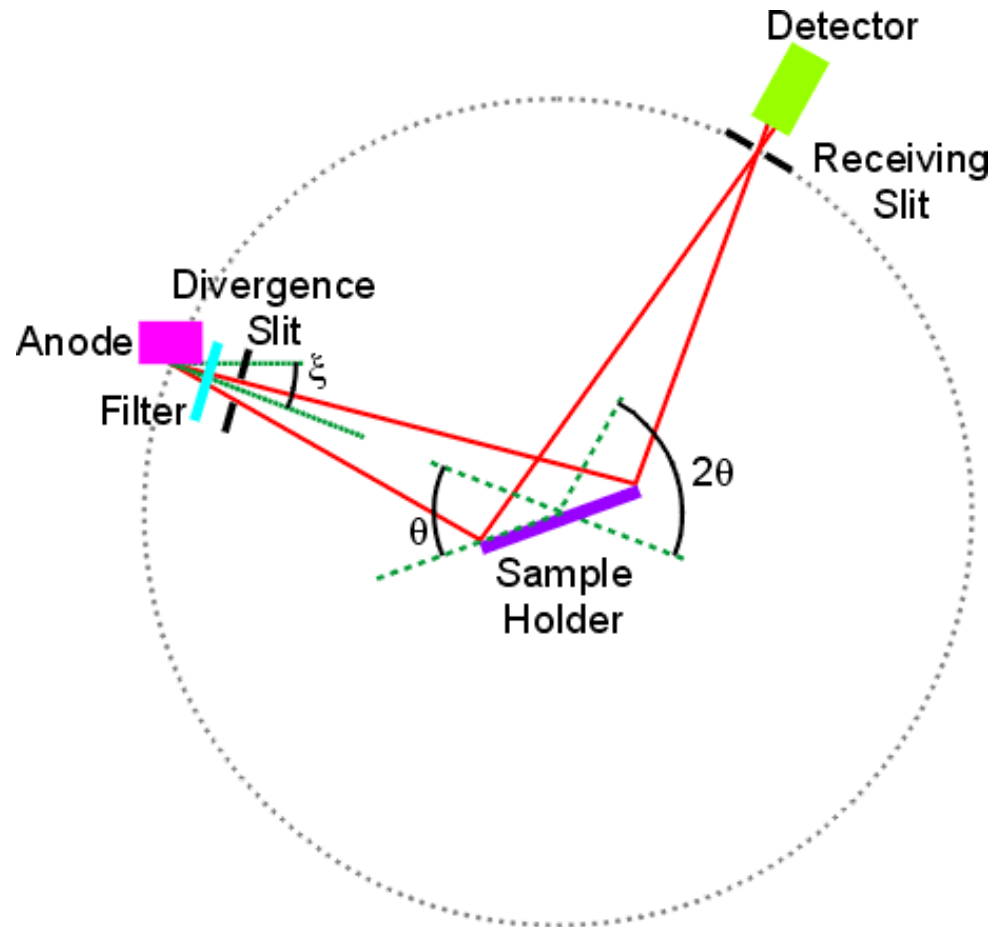
# Powder diffraction



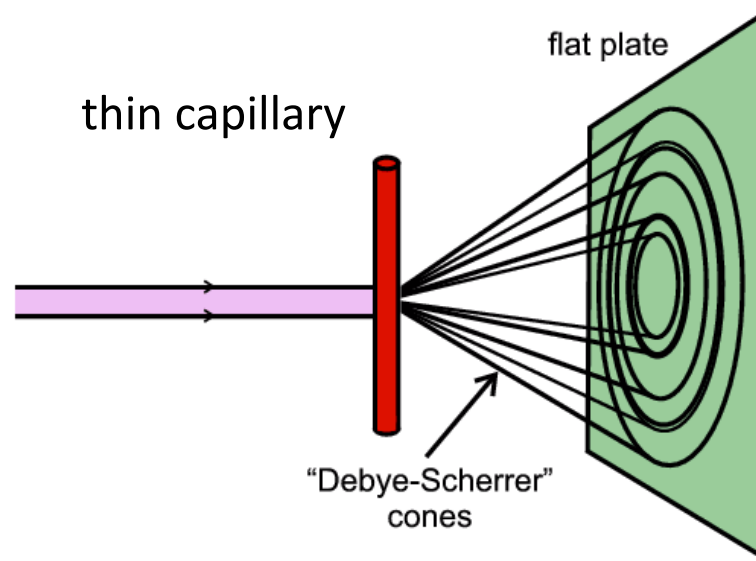
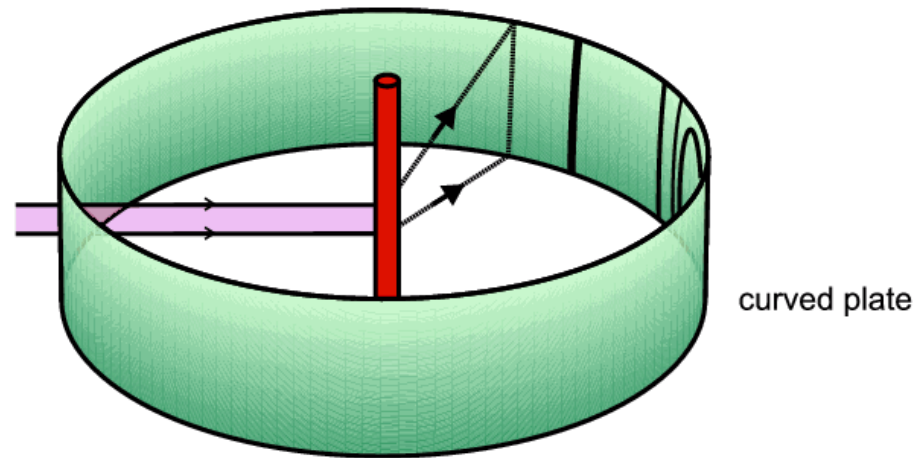
# Quartz powder pattern



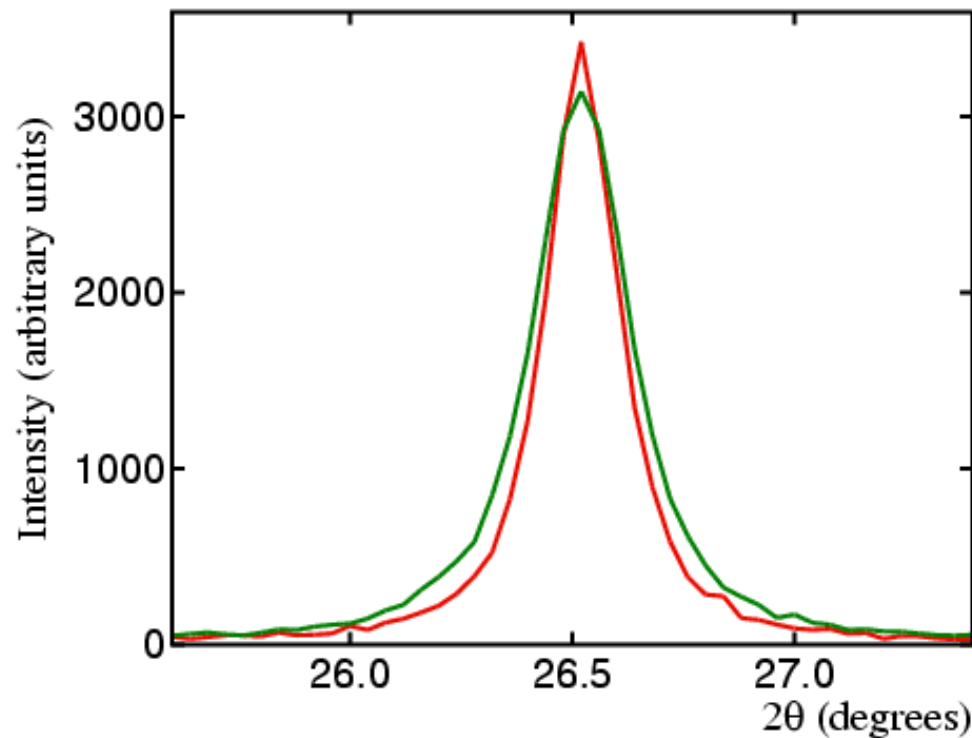
# Bragg-Brentano geometry



# Debye Scherrer method



## Size effect on lineshape



The Scherrer equation

$$\tau = \frac{K\lambda}{\beta \cos \theta}$$

$\tau$  = crystal size (nm)

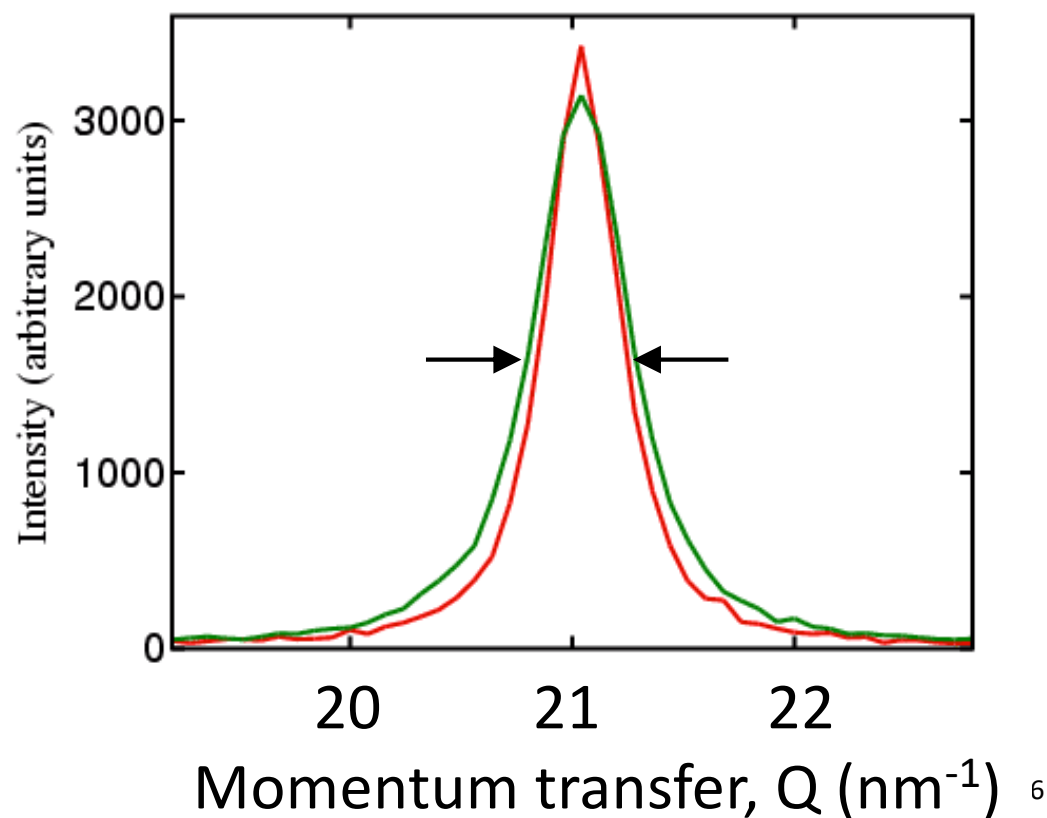
$\lambda$  = wavelength (nm)

$\beta$  = full width half max (rad)

$\theta$  = Bragg angle

$K$  = Scherrer shape factor  
= 0.94 for spheres

## “Physics” Notation may be Simpler



$$Q = k_f - k_i$$
$$= 4\pi/\lambda \sin \theta$$

$$\tau = 2\pi / \Delta Q$$

$\tau$  = crystal size (nm)

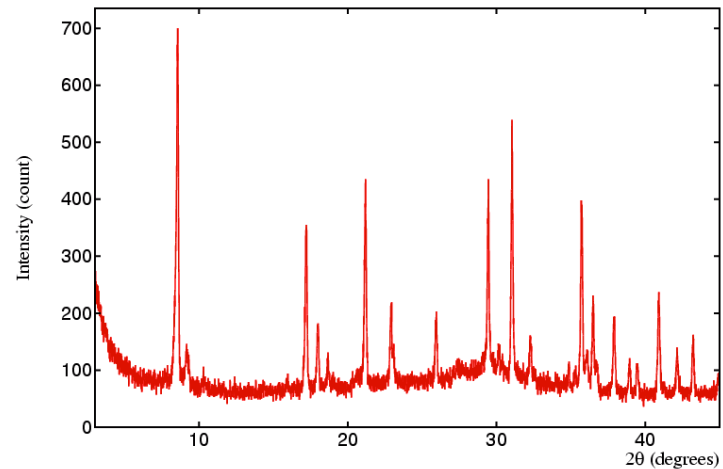
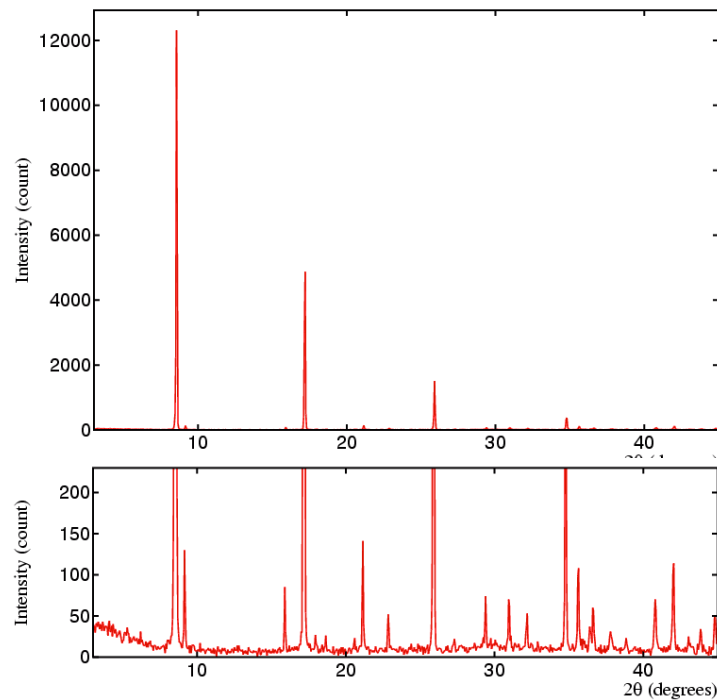
$\lambda$  = wavelength (nm)

$\theta$  = Bragg angle

$\Delta Q$  = full width half max

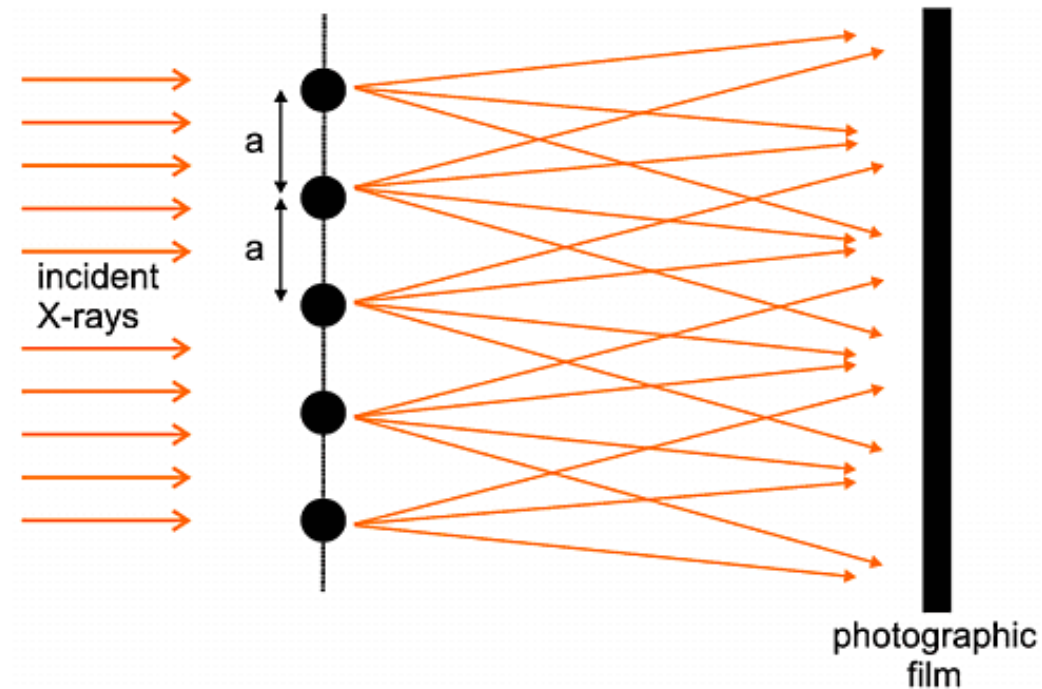
# Preferred orientation = texture

hydrated cement phase  $\text{Ca}_4\text{Al}_2(\text{SO}_4)\text{O}_6 \cdot 16\text{H}_2\text{O}$



left: flat plate holder  
right: capillary holder

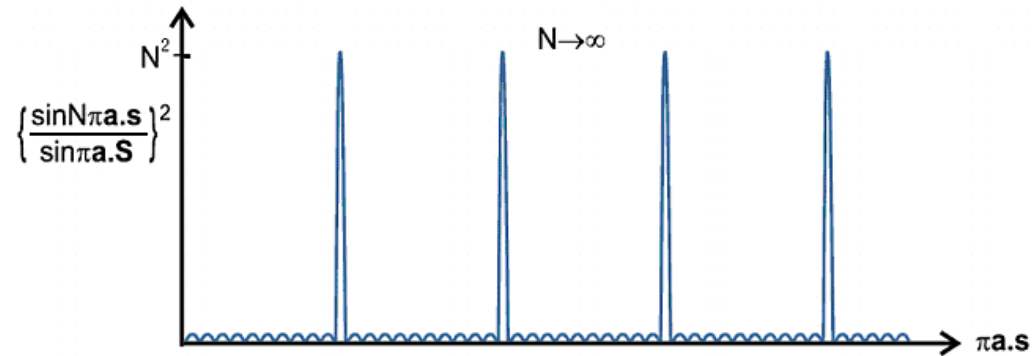
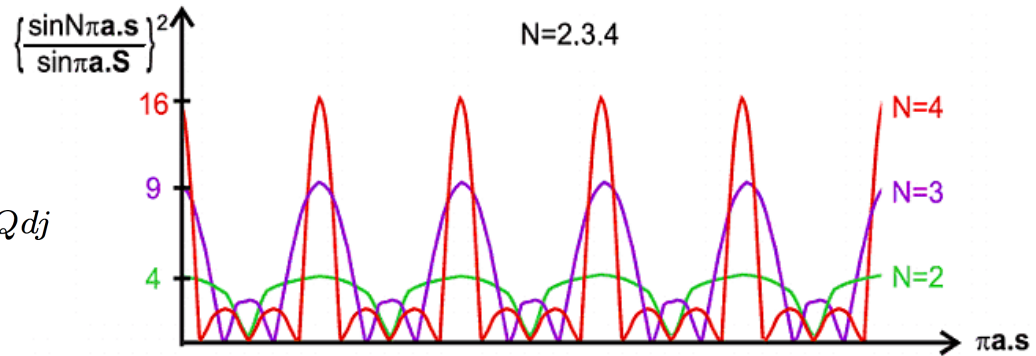
# Scattering from array of unit cells



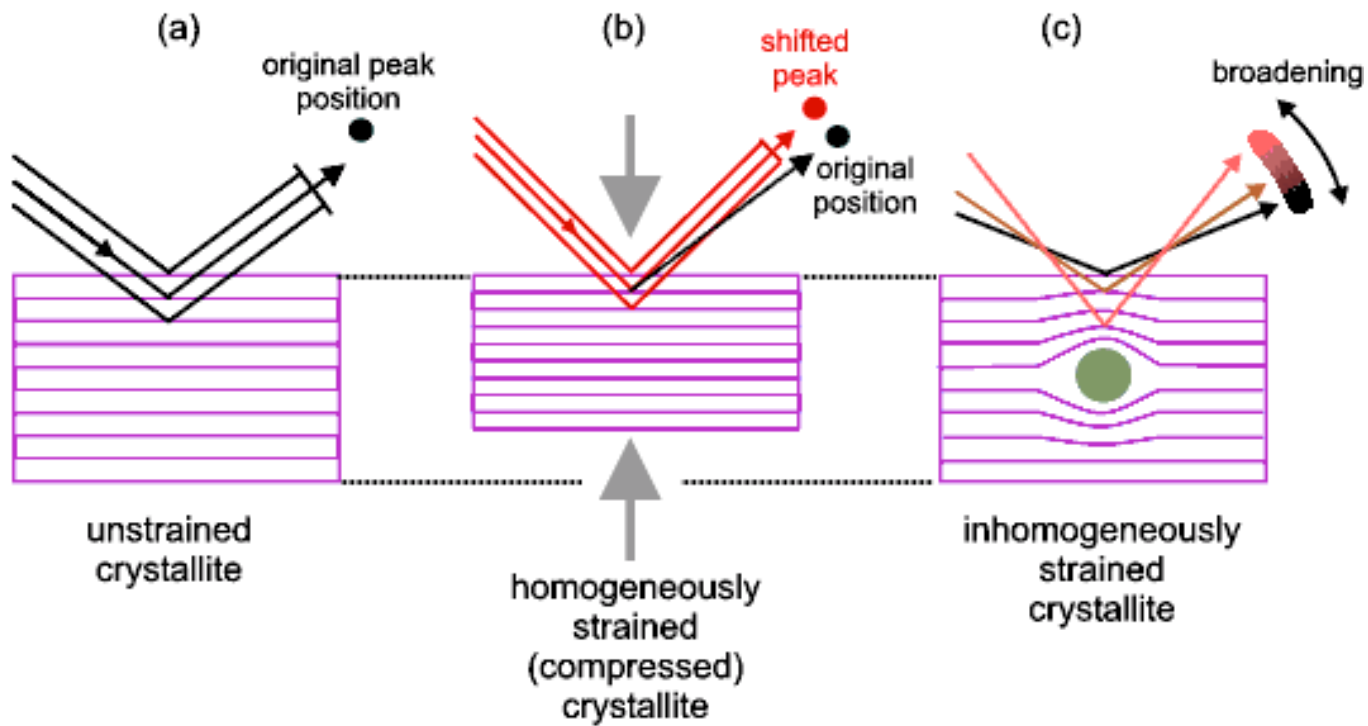
# Interference sum over unit cells

$$R_N(Q) = A \sum_{j=0}^{N-1} e^{iQdj}$$

$$= A \frac{1 - e^{iQdN}}{1 - e^{iQd}}$$



# Effect of Strain on Diffraction



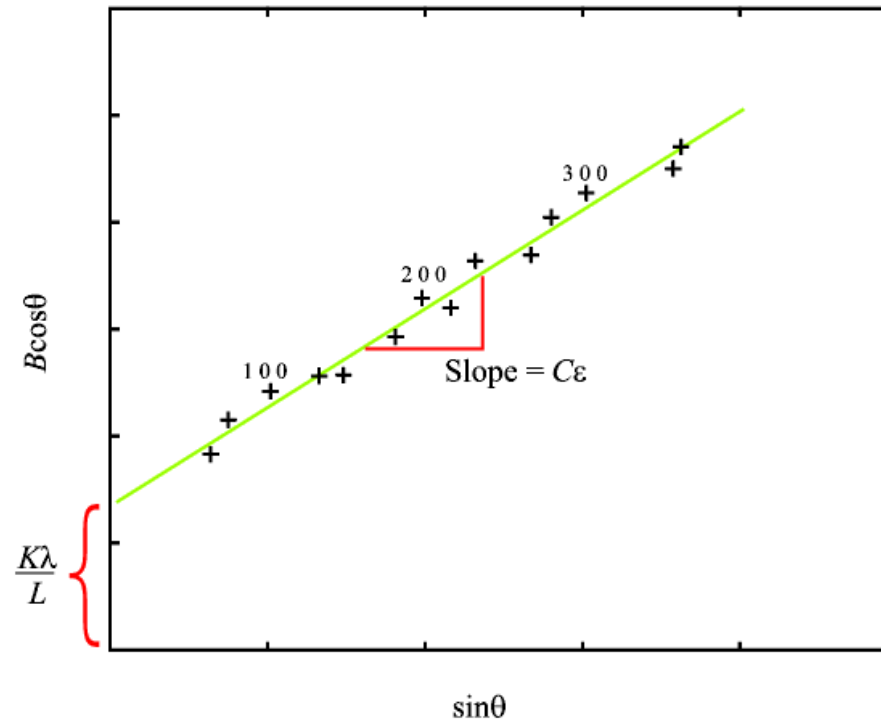
# G.K.Williamson and W.H.Hall Acta Metall. 1, 22-31 (1953)

Scherrer equation

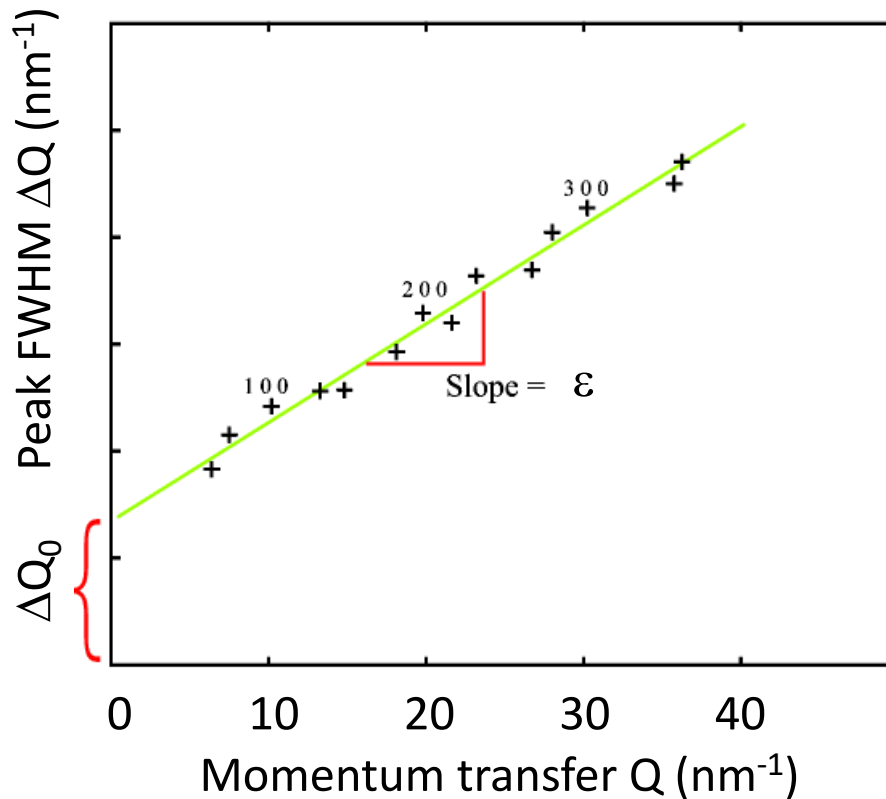
$$\beta_L = \frac{K\lambda}{L \cos\theta}$$

$$\beta_e = C\epsilon \tan\theta$$

$$\beta_{\text{tot}} = \beta_e + \beta_L = C\epsilon \tan\theta + \frac{K\lambda}{L \cos\theta}$$



## W-H Size-Strain in “Physics” Notation



$$Q = k_f - k_i$$

$$= 4\pi/\lambda \sin \theta$$

$$\tau = 2\pi / \Delta Q_0 = \text{size}$$

$\tau$  = crystal size (nm)

$\lambda$  = wavelength (nm)

$\theta$  = Bragg angle

$\Delta Q$  = full width half max

$\epsilon = \delta d/d$  = “microstrain”  
= range of local strains

# Scattering Outline

- Powder diffraction methods “XRD”
- Surface diffraction and surface structure
- Extension to Coherent X-ray Diffraction
- XFEL study of thin film melting

# First UHV Experiments (1981)

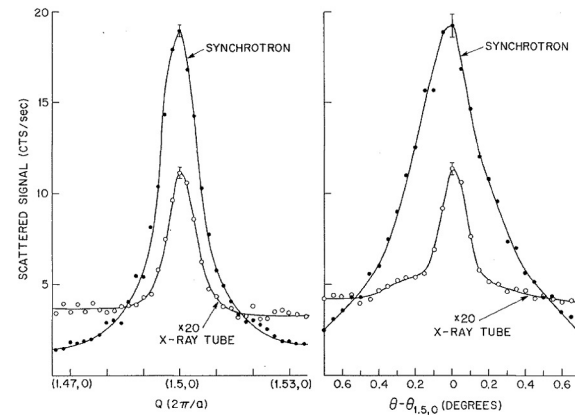
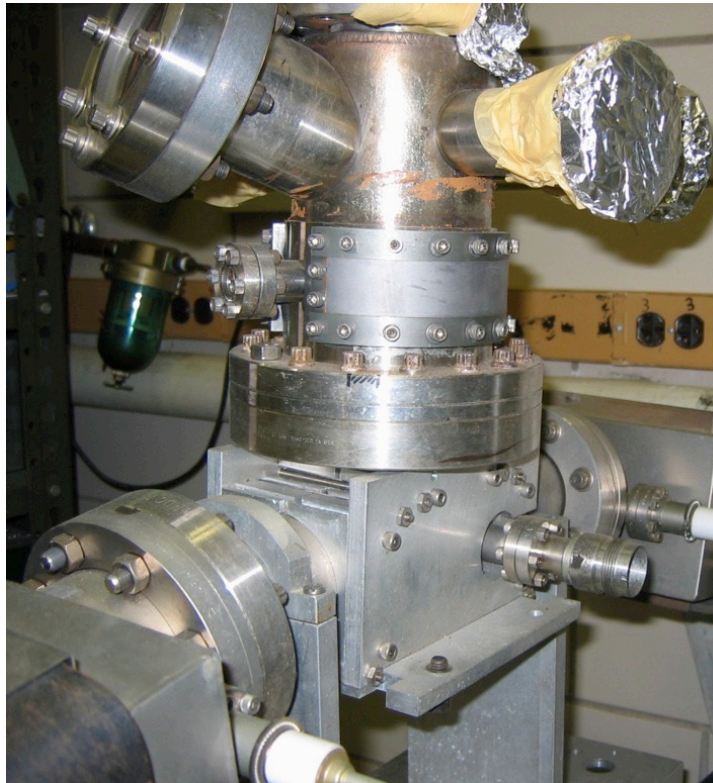
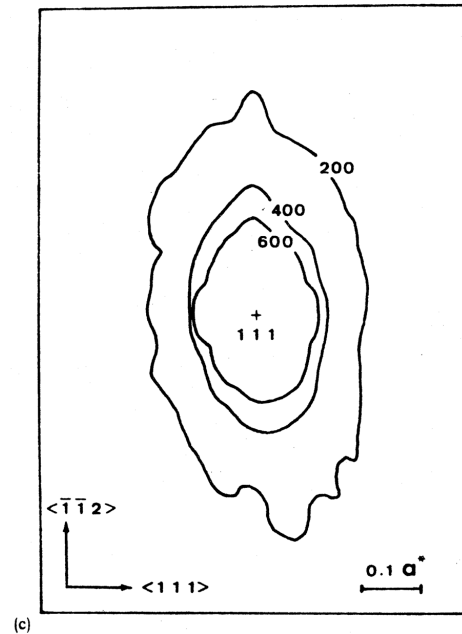


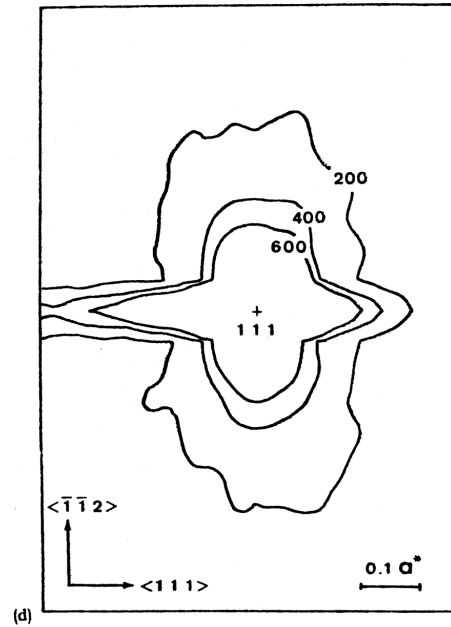
FIG. 1. A plot of the  $(\frac{3}{2}, 0)$  Bragg reflection as a function of the momentum transfer  $Q(2\pi/a)$  and the crystal's mosaic spread (deg).

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

# Diffuse Scattering from Si Wafer



(c) Unpolished wafer



(d) 40 microns removed

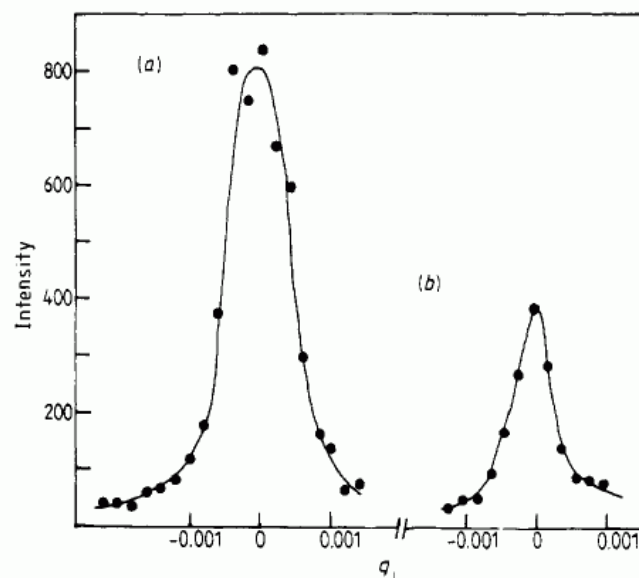
N. Kashiwagara, J. Harada and M. Ogino, J. Appl. Phys 54 2706 (1983)

# Scattering of X-rays From Crystal Surfaces

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

*Scattering of x-rays from crystal surfaces*

6433



**Figure 1.** The intensity of scattering, as a function of  $q_1$ , (a) for  $\mathbf{Q} = (0, 0, 4.005)$  (vertical scale in counts per 2 s) and (b)  $\mathbf{Q} = (0, 0, 4.025)$  (vertical scale in counts per 300 s) in the GaAs sample with an (001) surface corresponding to figure 2, curve B.

# 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)

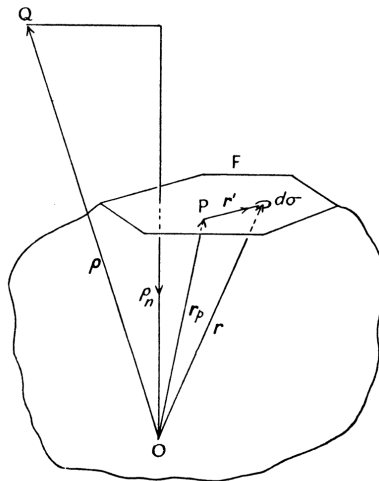
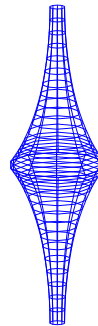


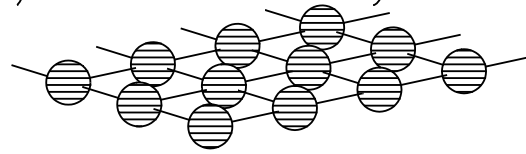
FIG. 200



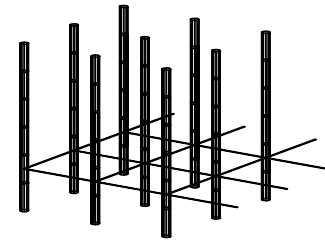
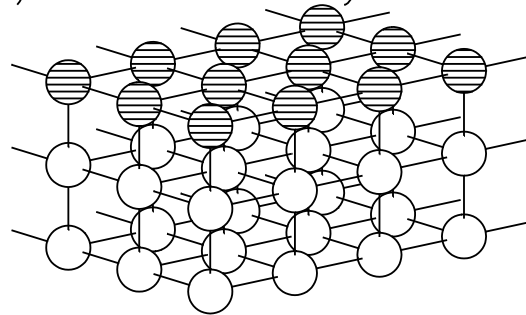
“Stacheln”

# Crystal Truncation Rods (1986)

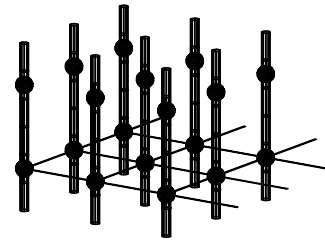
a) Isolated Monolayer



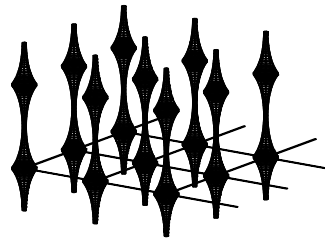
b) Surface of Crystal



2D  
LAYER  
ONLY

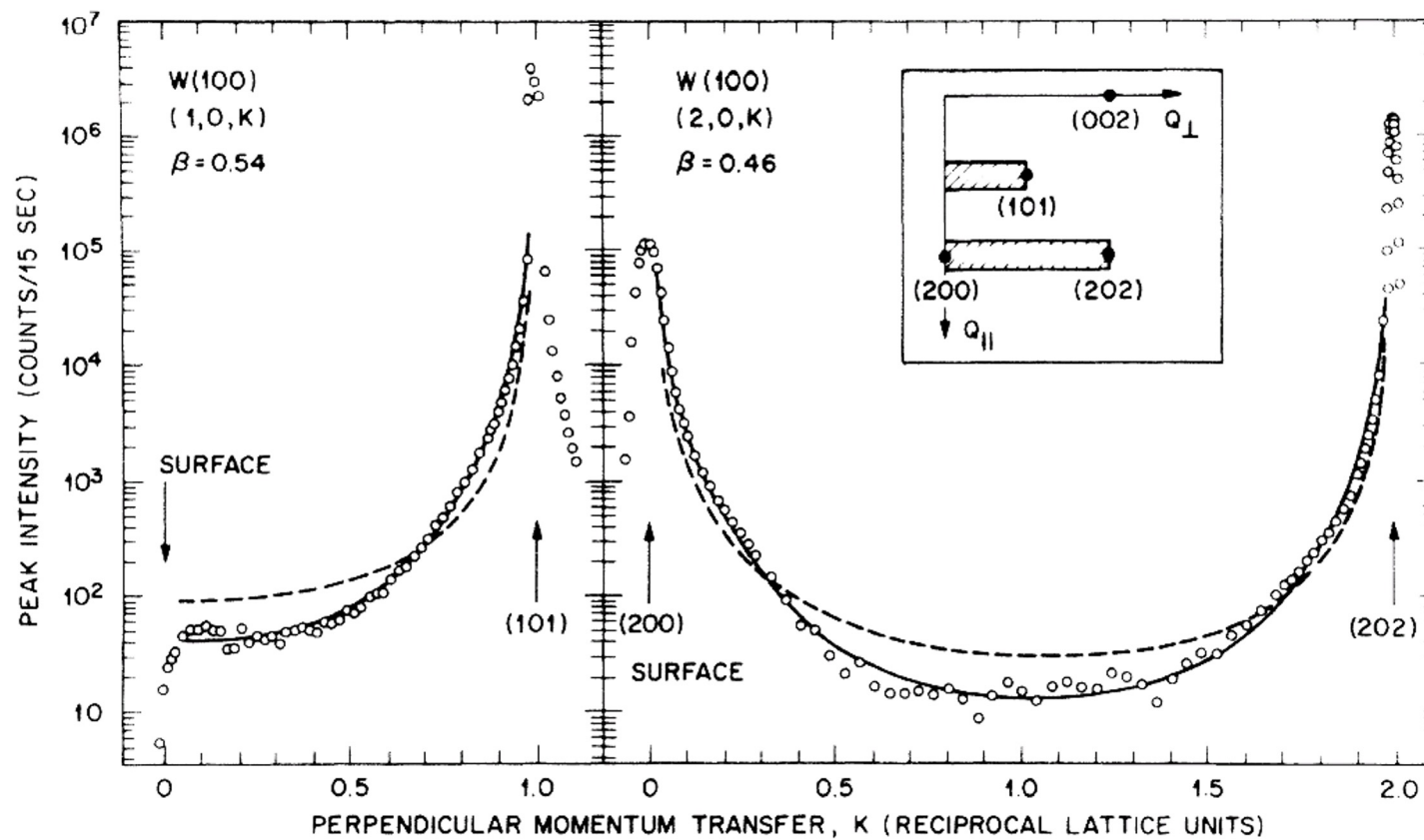


BULK  
CRYSTAL  
AND 2D  
LAYER

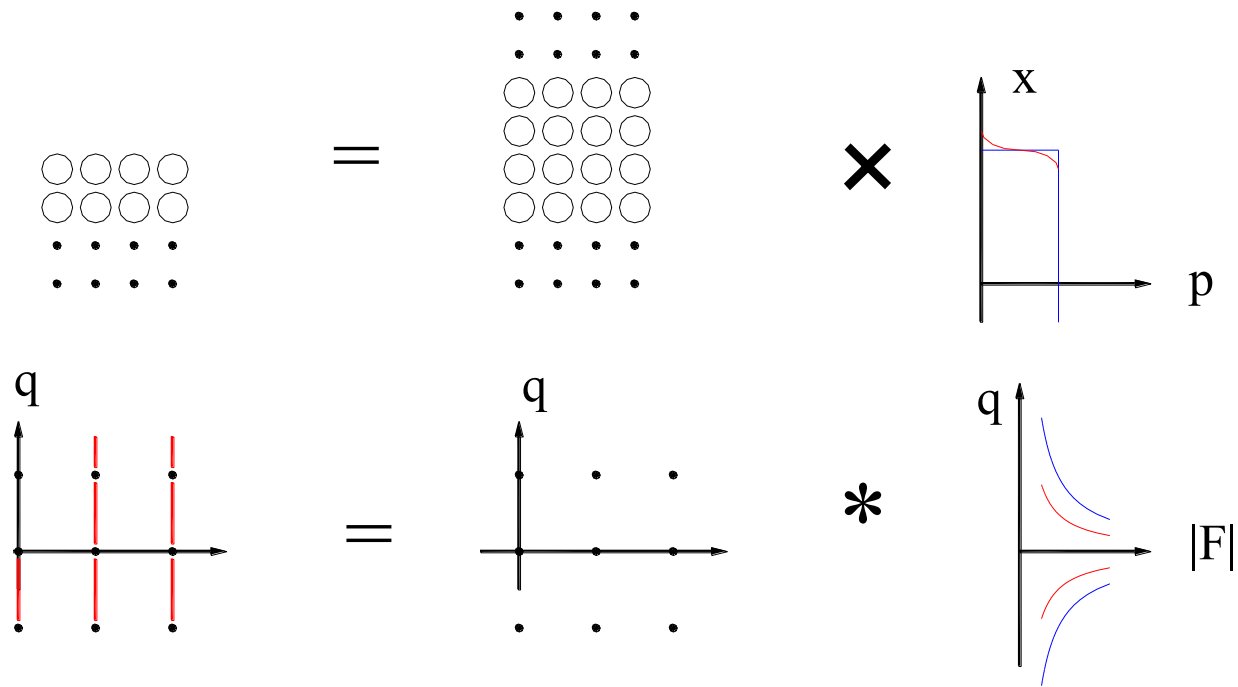


CRYSTAL  
TRUNCATION  
RODS

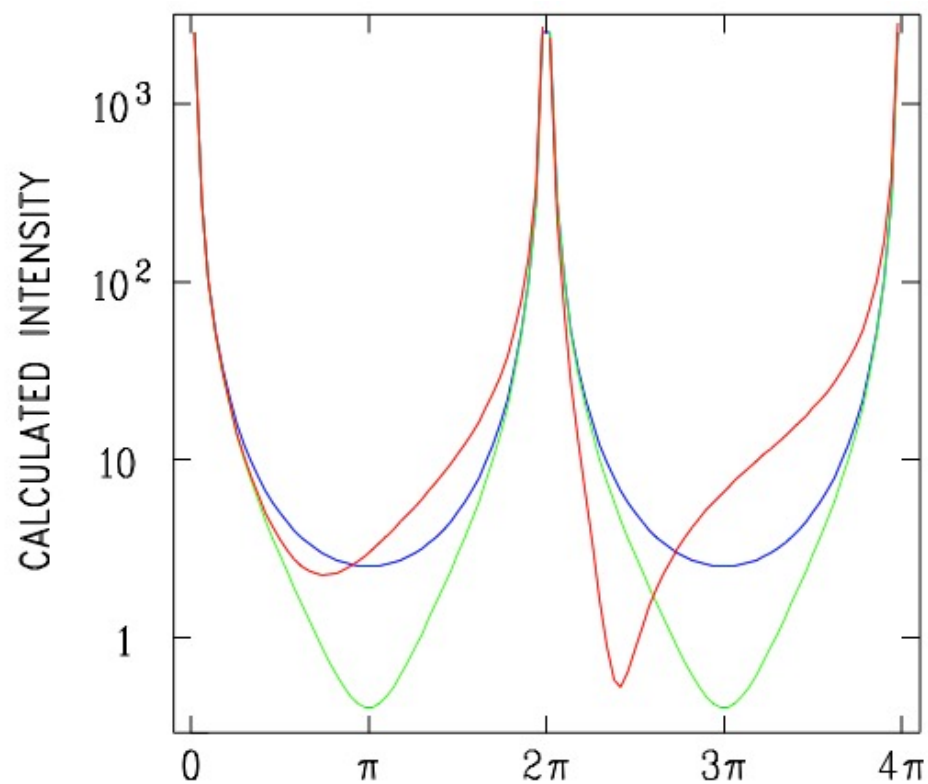
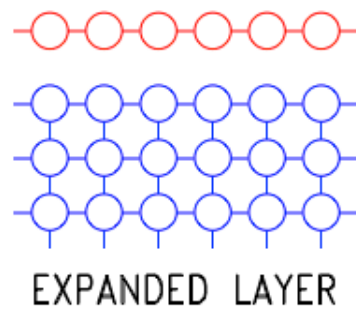
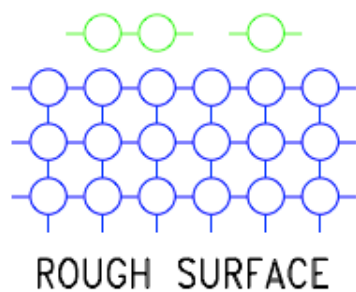
# CRYSTAL TRUNCATION RODS AND SURFACE ROUGHNESS



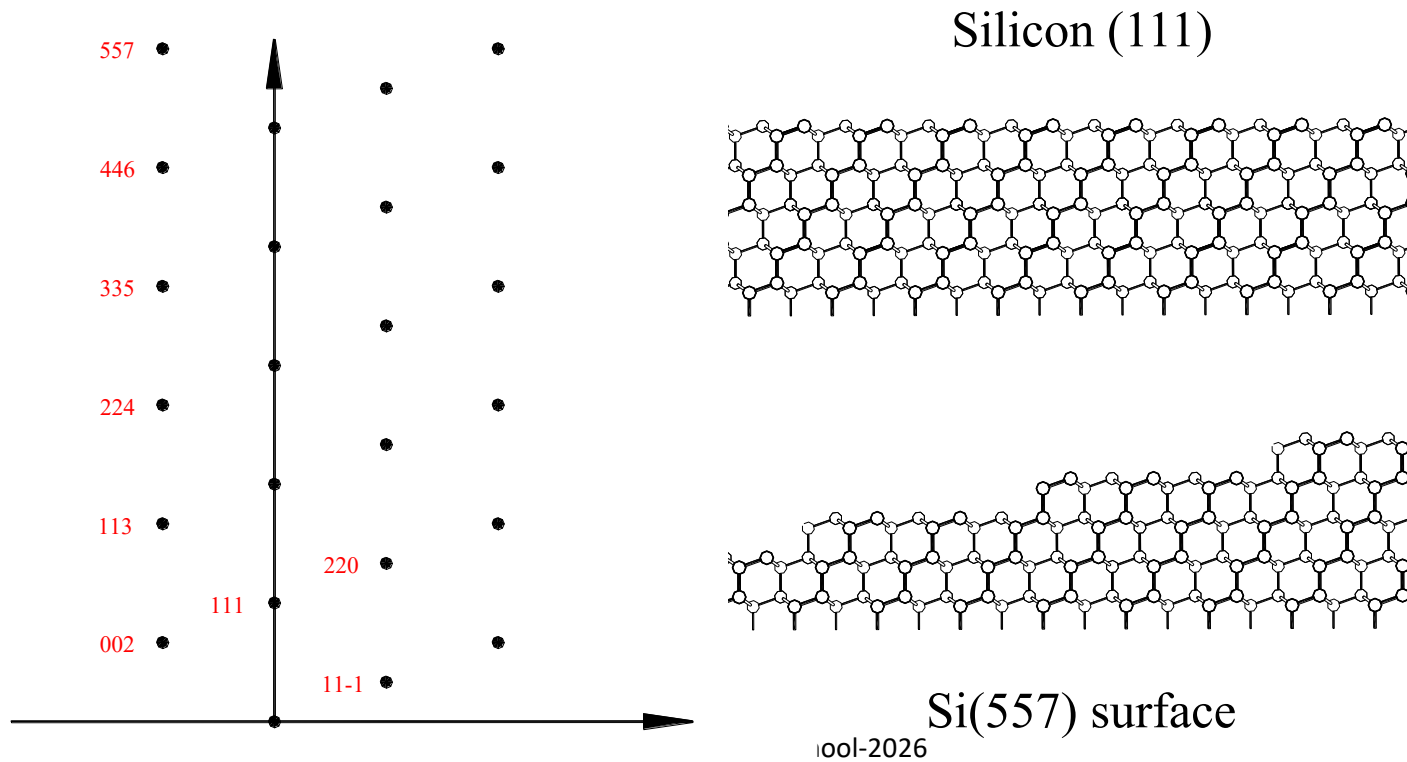
# CTR as Convolution



# CTR is Sensitive to Surface Structure

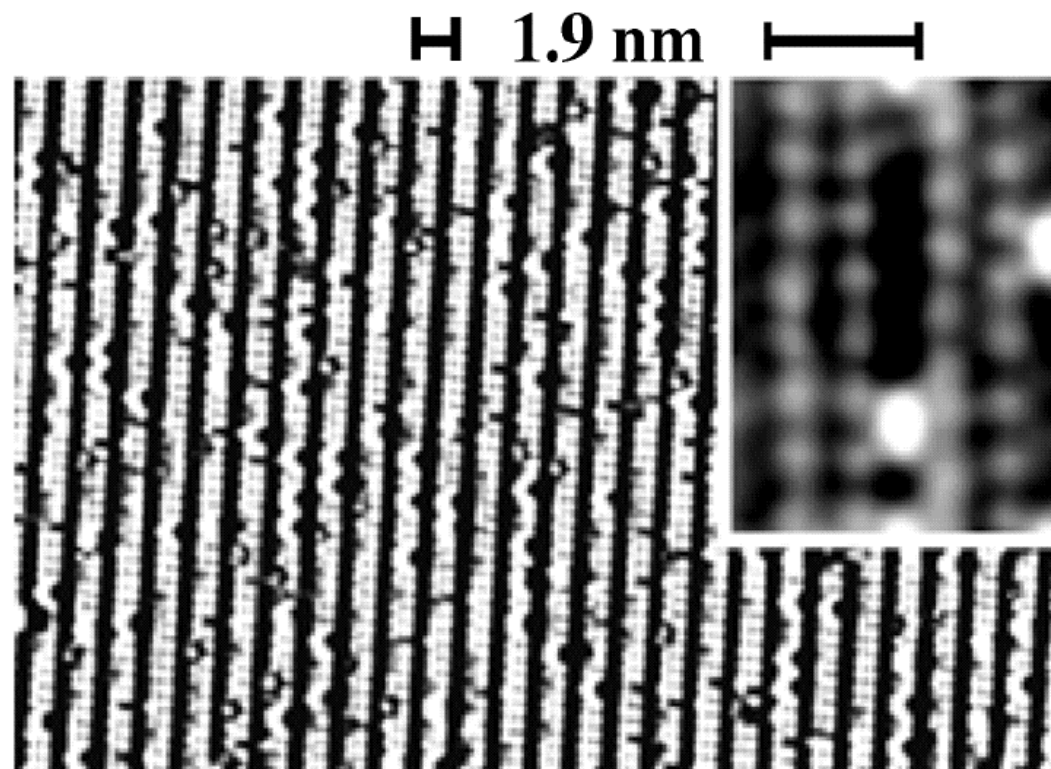


# Crystallography of Stepped Surfaces

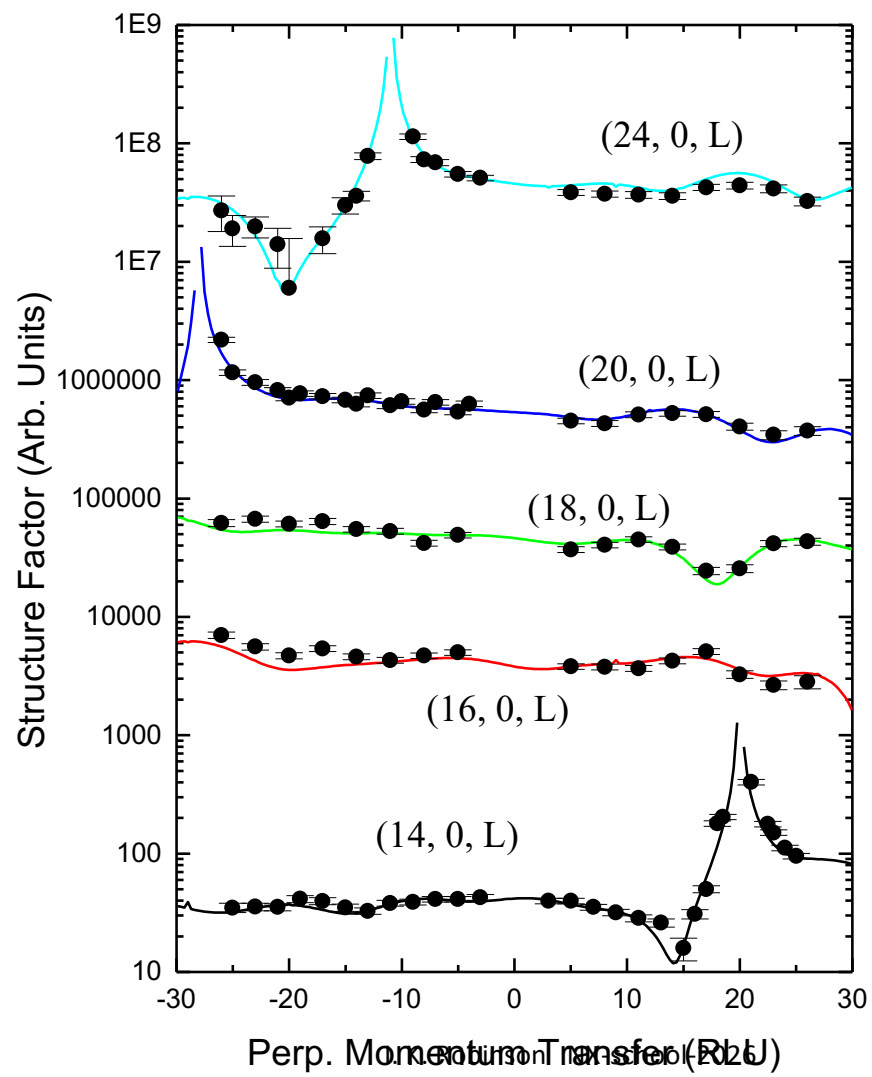


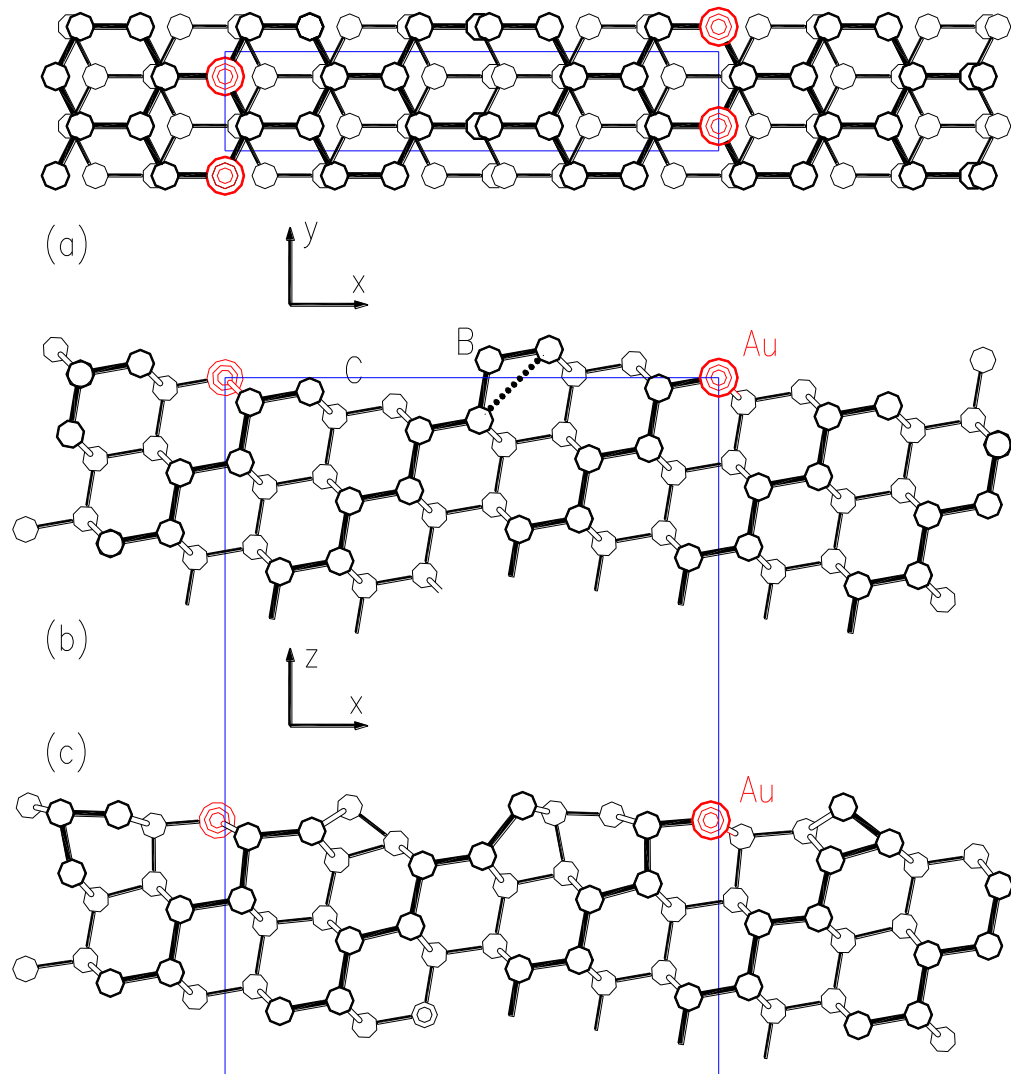
## Si(557) coated with Au

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

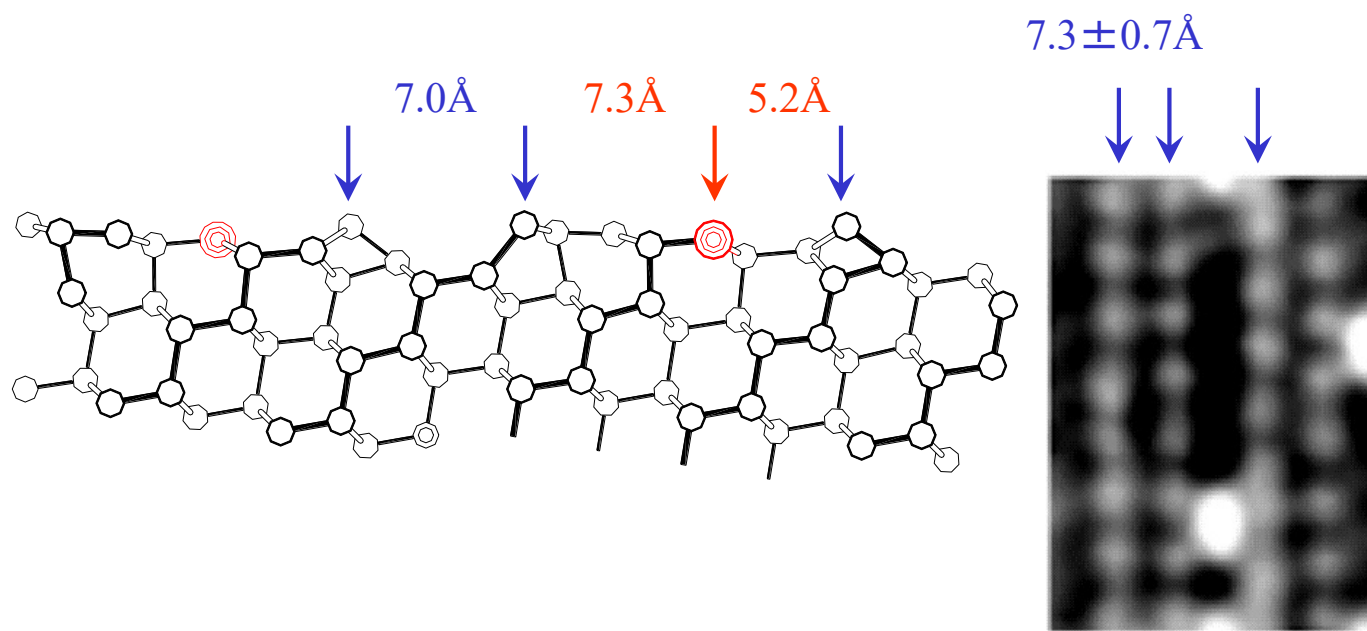


I. K. ROBINSON TX-SCH00F-2026

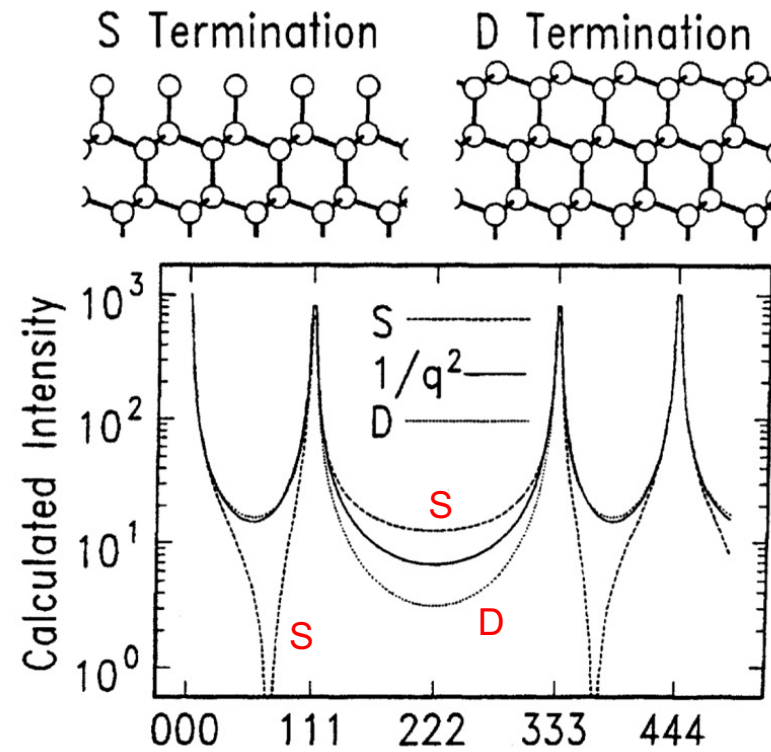
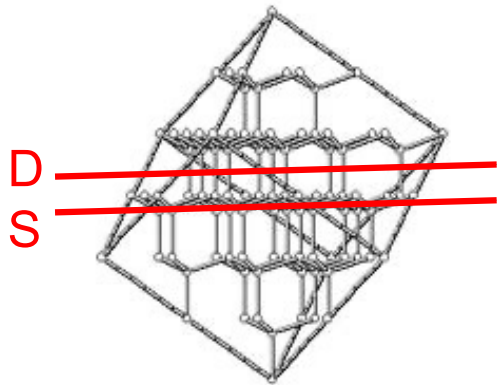




## Comparison with STM

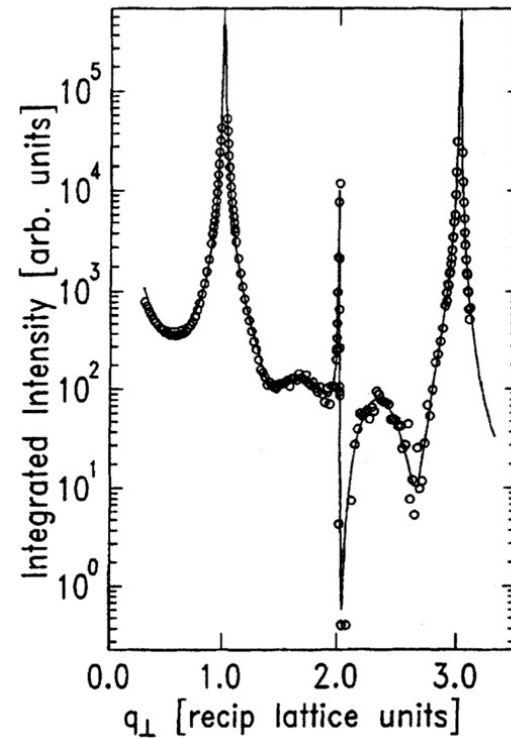
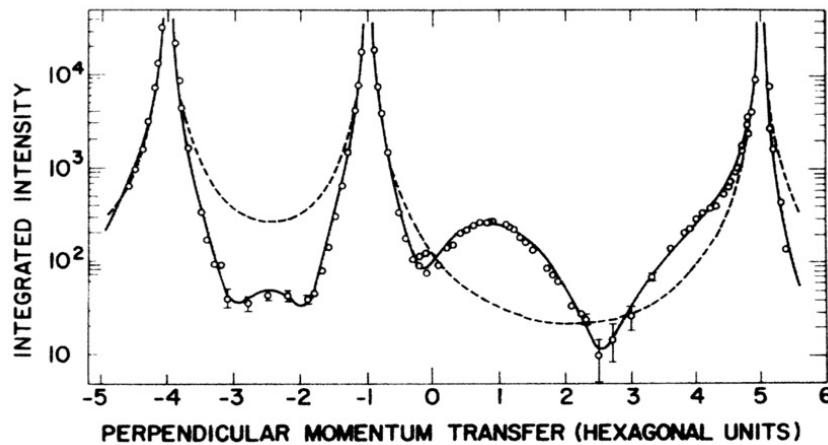


# (111) Surface of Diamond Lattice



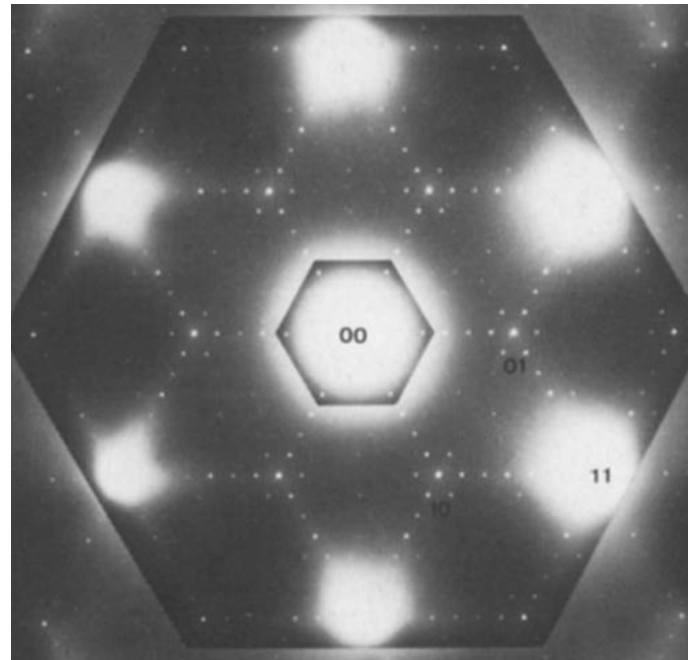
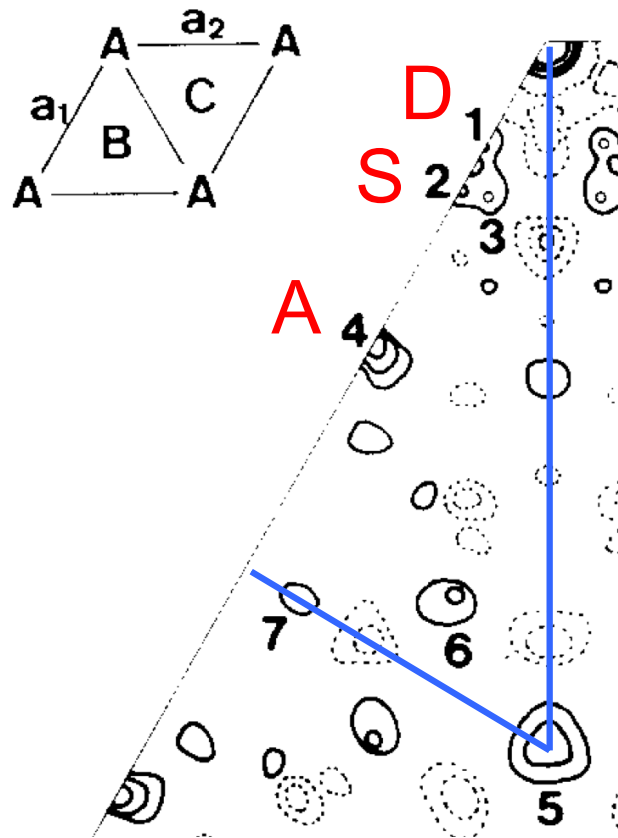
# (111) Surface of Diamond Lattice

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

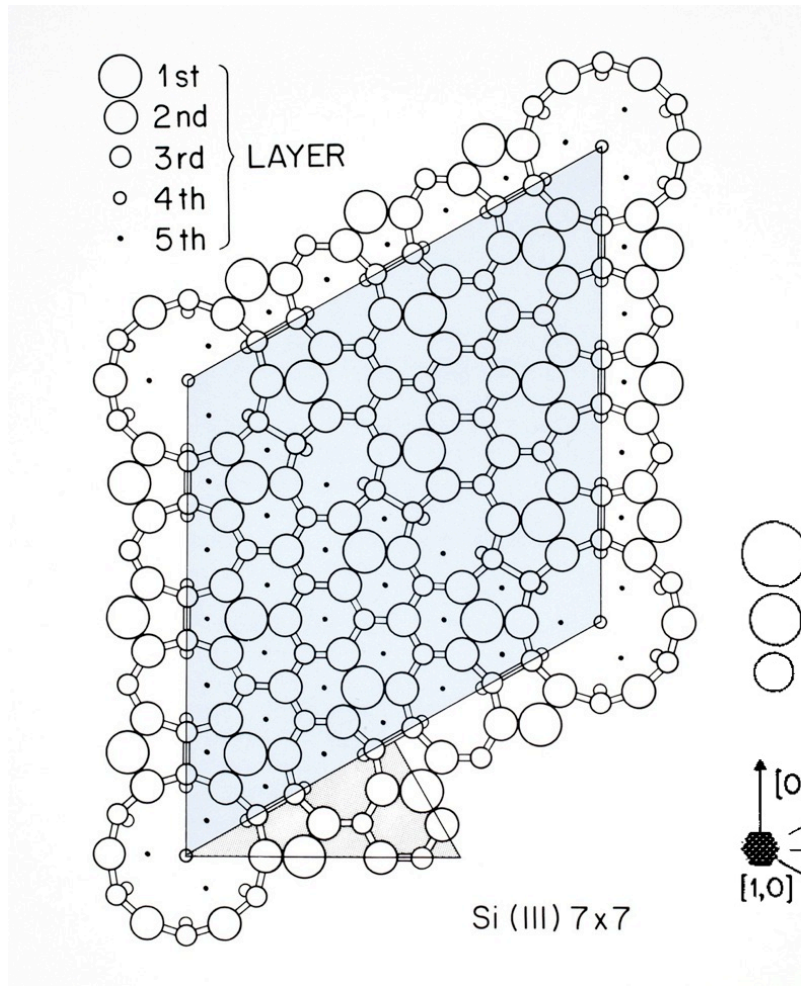


# Patterson: Dimer-Adatom-Stacking-Fault

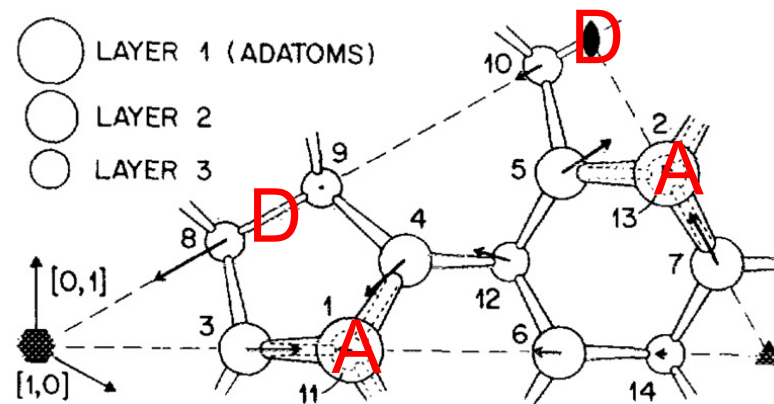
K. Takayanagi et al, Surf. Sci. 164 367 (1985)



Robinson NX-school-2026



## DAS Model Si(111) 7x7



# Surface X-ray Diffraction Collaborators

Peter Eisenberger

Paul Fuoss

Peter Bennett

Doon Gibbs

Ben Ocko

Peter Eng

Robert Feidenhans' 1

Jens Als-Nielsen

Jakob Bohr

Mike Altman

Elias Vlieg

Sunil Sinha

Don Walko

Franz Himpsel

Bell Labs

Stanford

Arizona

Brookhaven

Brookhaven

Chicago

Copenhagen

Copenhagen

Copenhagen

HKUST

Nijmegen

San Diego

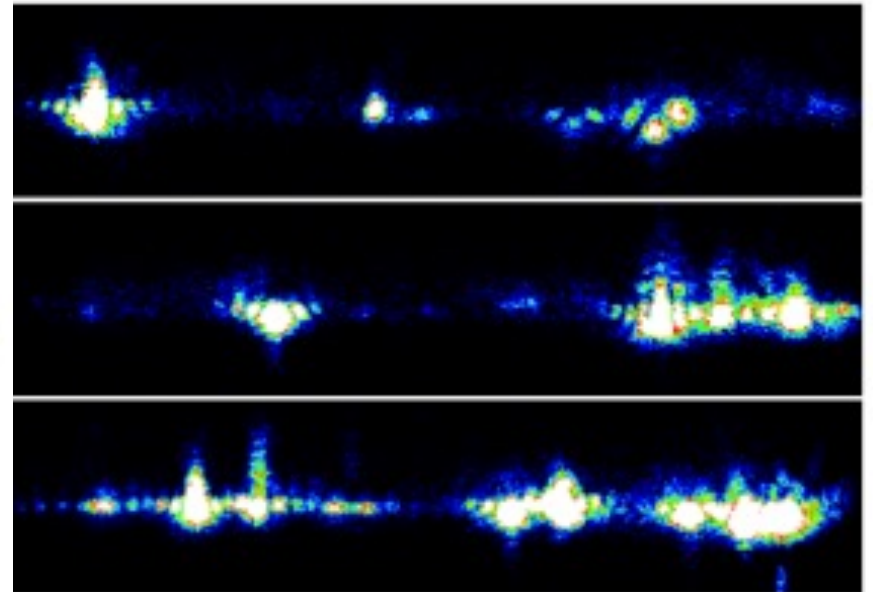
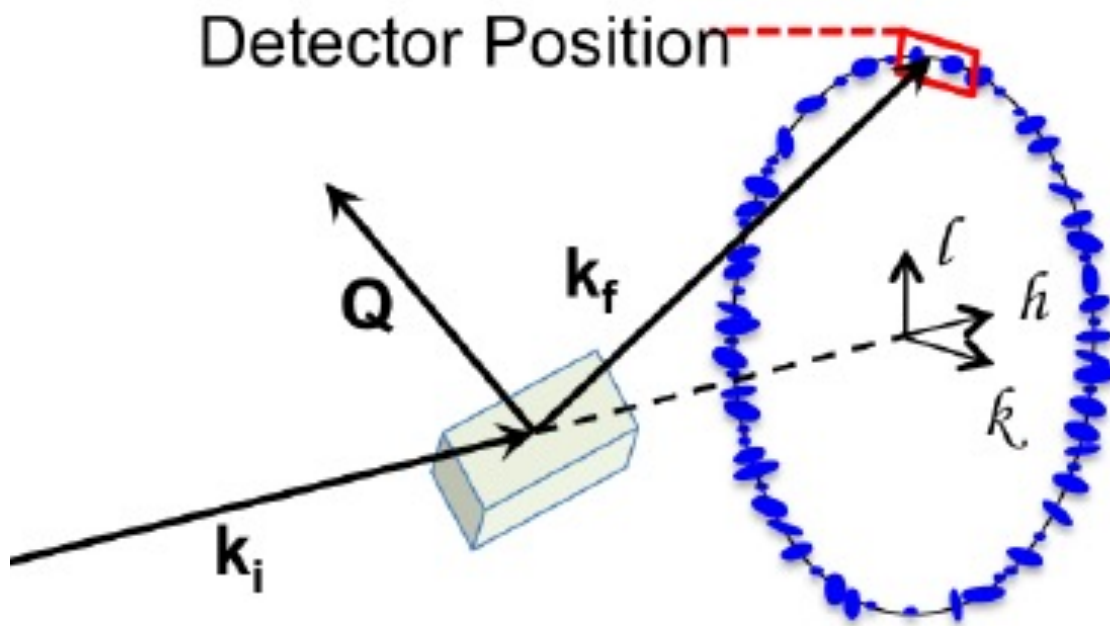
Urbana

Wisconsin

# Scattering Outline

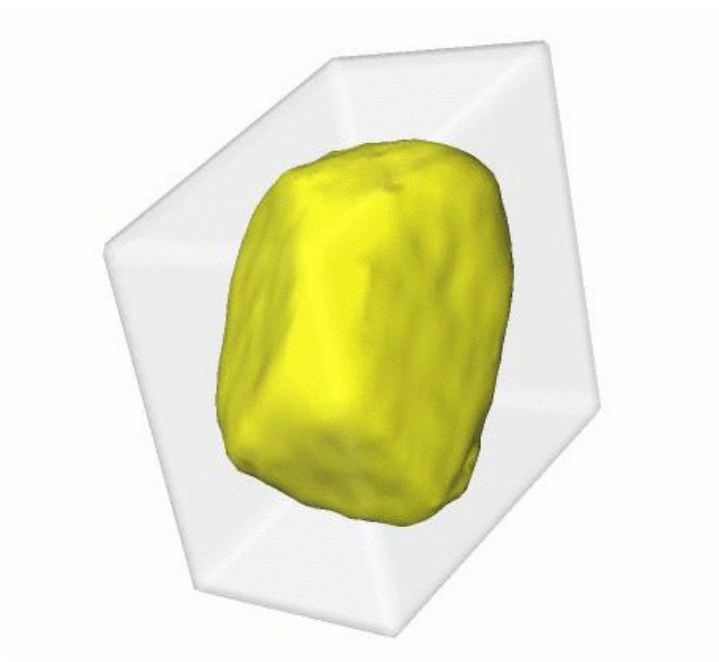
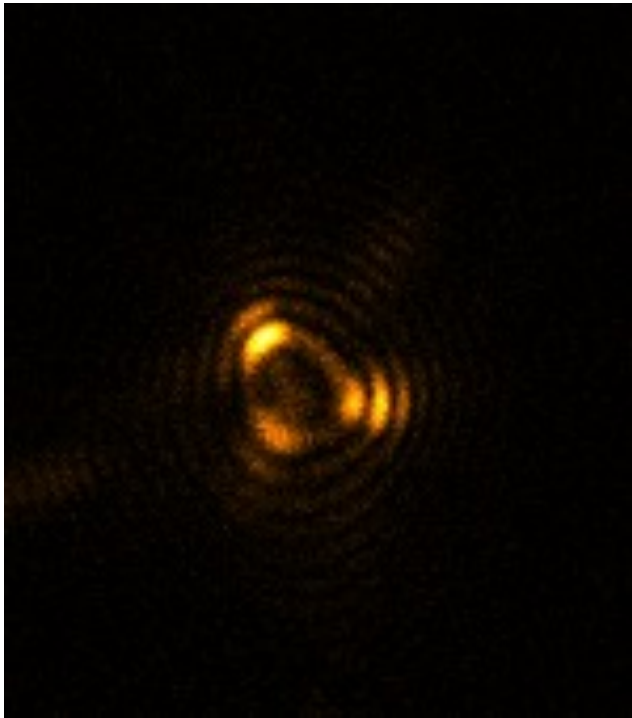
- Powder diffraction methods “XRD”
- Surface diffraction and surface structure
- Extension to Coherent X-ray Diffraction
- XFEL study of thin film melting

# Powder diffraction with Coherence

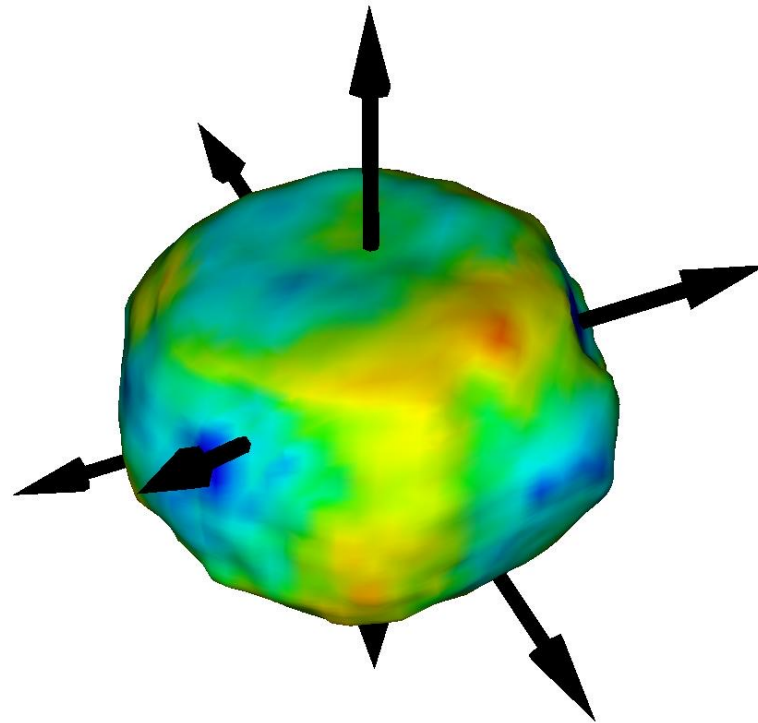


# Gold nanocrystal reconstruction

showing support used for 20 HIO followed by 10 ER

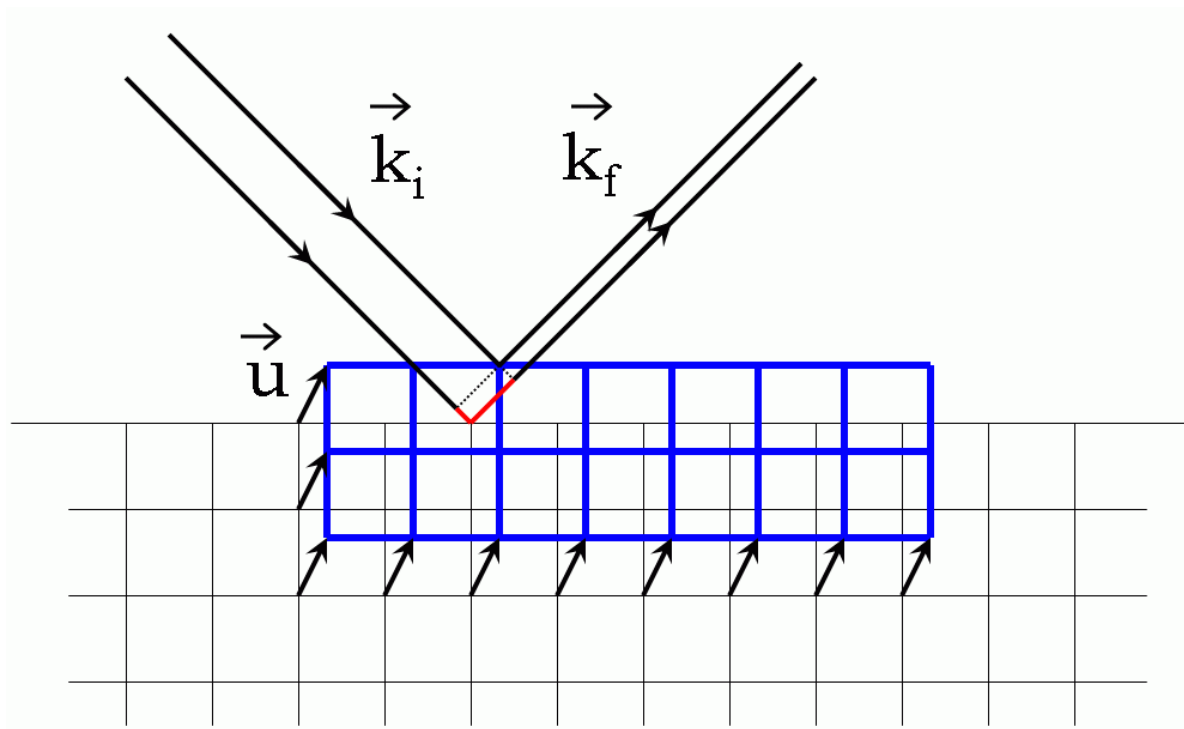


# Phase isosurface of residual strain

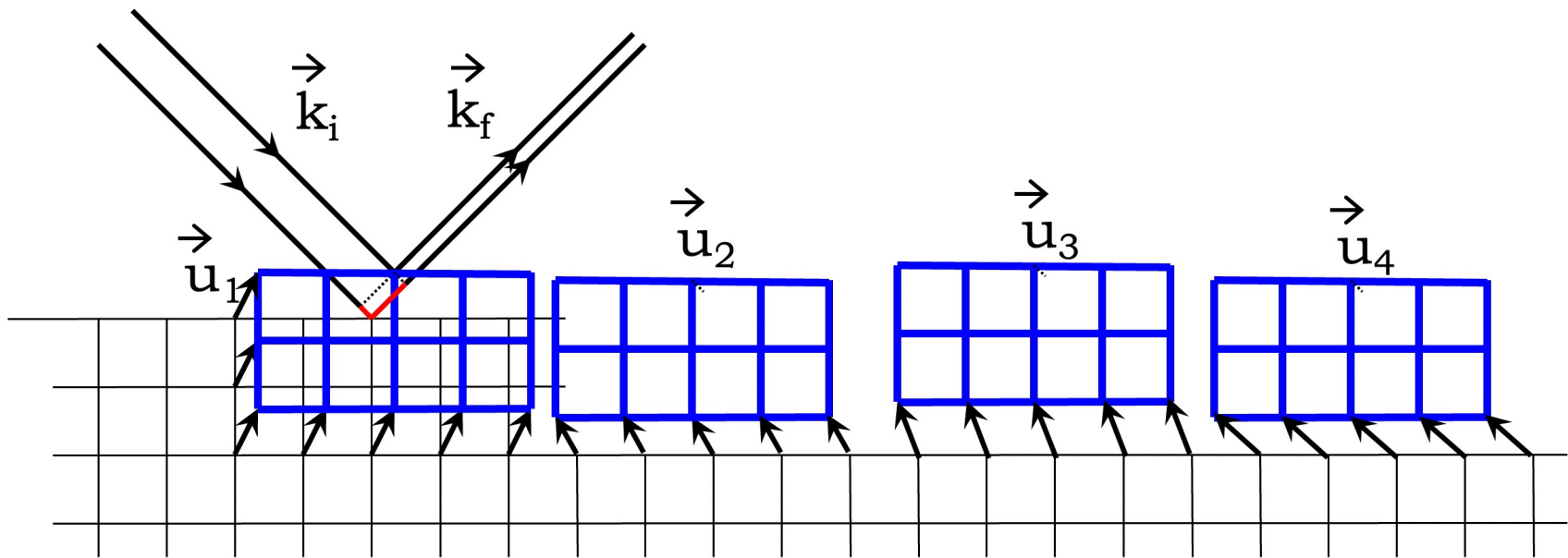


# Sensitivity to strain

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



# Generalization to Phase Domains

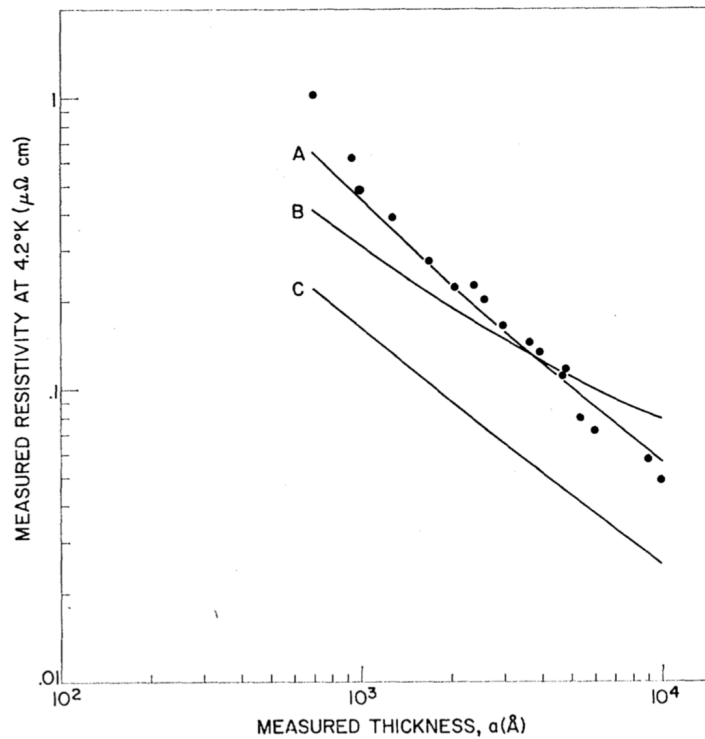


# Scattering Outline

- Powder diffraction methods “XRD”
- Surface diffraction and surface structure
- Extension to Coherent X-ray Diffraction
- XFEL study of thin film melting

# Resistivity of Al Thin Films

A. F. Mayadas and M. Shatzkes, PRB **1** 1382 (1970)



- “Universal curve” of MFP vs electron energy
- Thermal MFP removed at low temperature
- Grain size proportional to thickness (model)



# “Two-temperature” model (2TM)

I. K. Robinson et al, Journal of Optics **18** 054007 (2016)

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

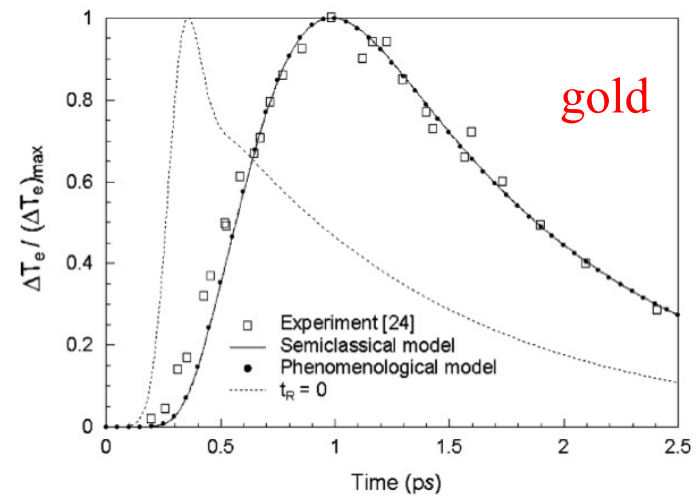
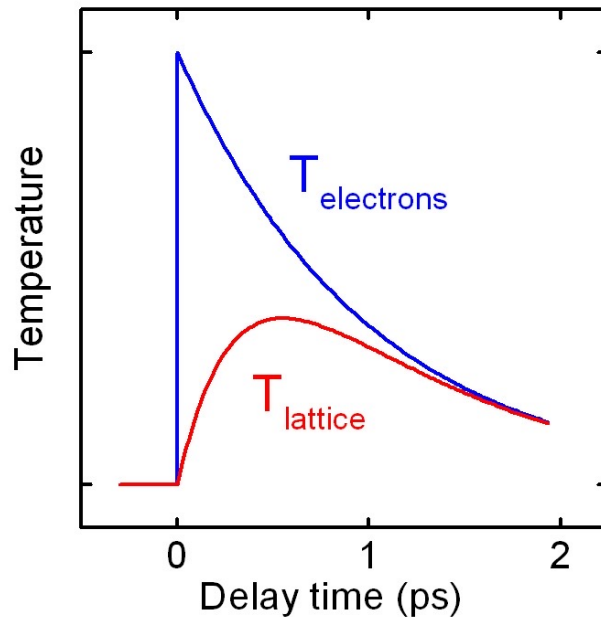
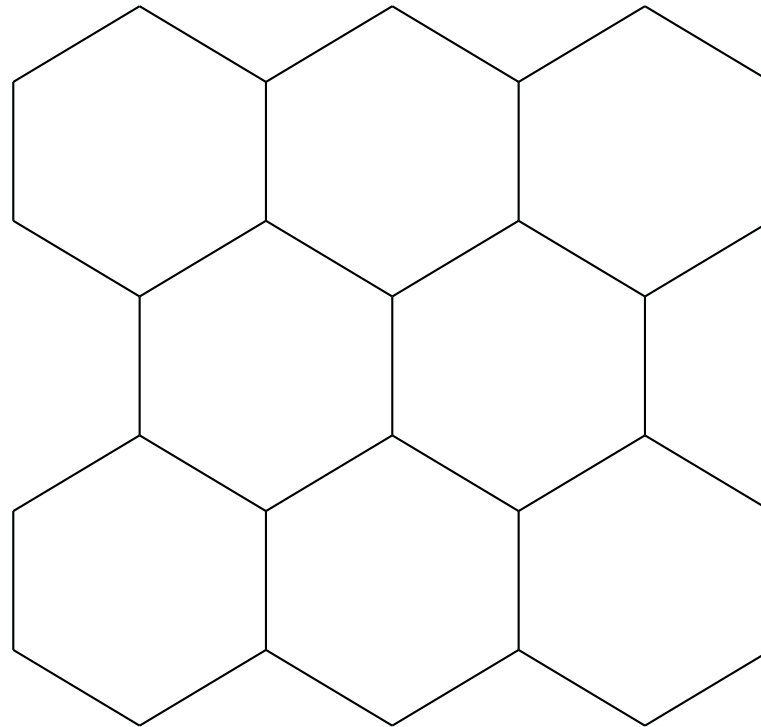
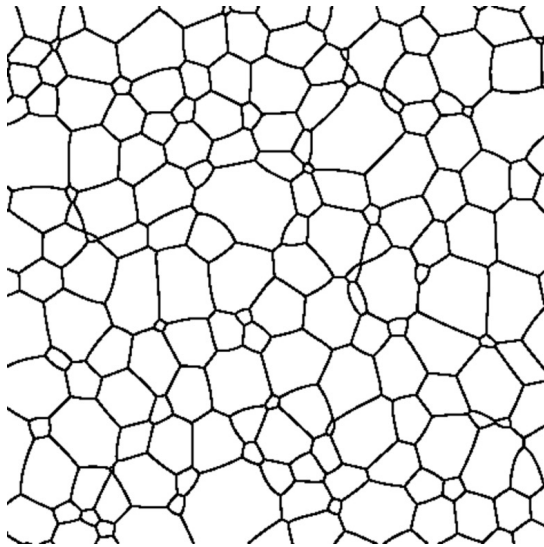


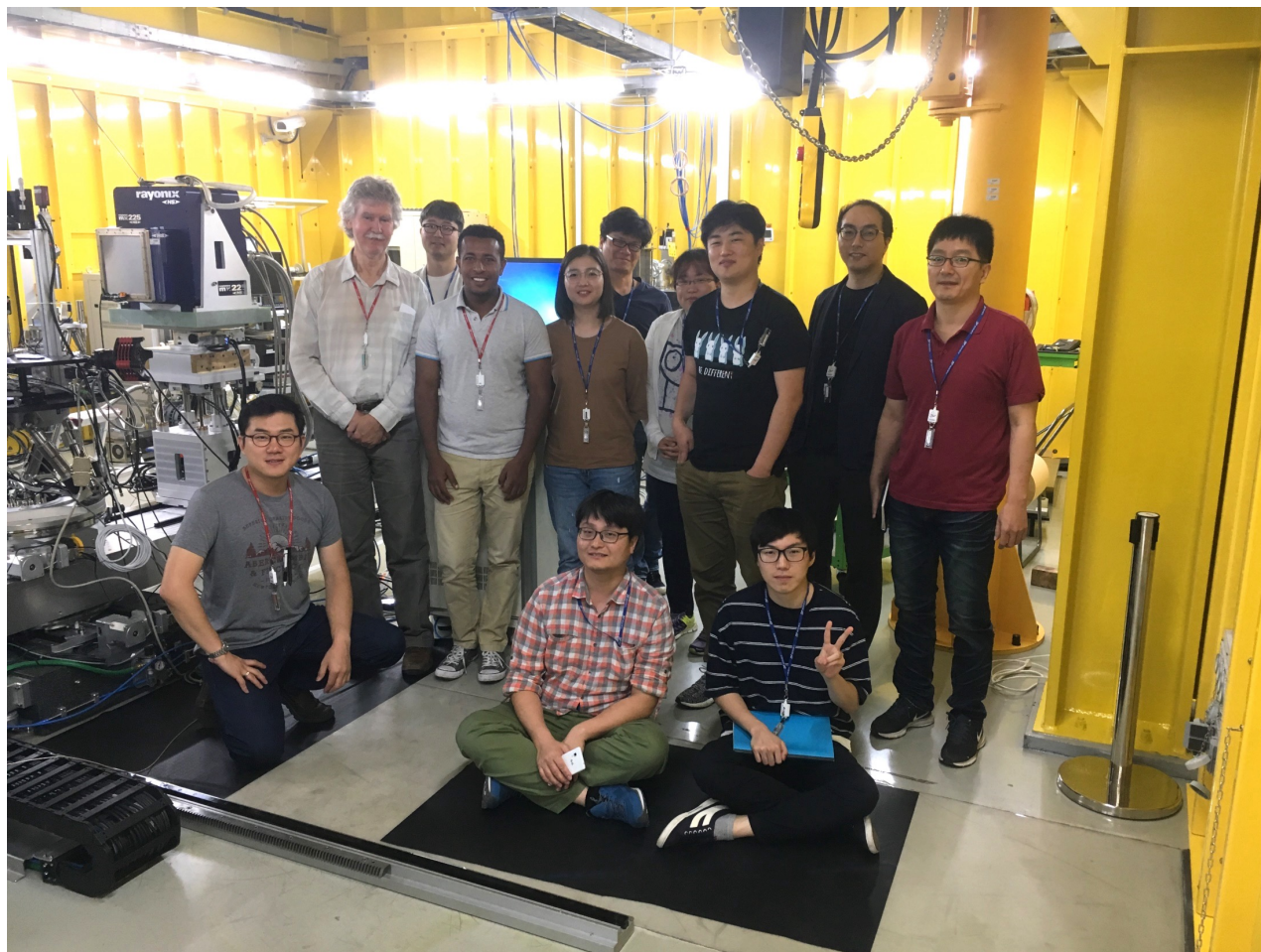
Fig. 2. Comparison of the change in electron temperature at the front surface of an 80-nm gold film irradiated by a 2.8 mJ/cm<sup>2</sup>, 800 nm, 150-fs laser pulse.

# XFEL Questions about Melting

- Where does the melting start?
- Where does the 2TM couple to the lattice?
- Role of sample geometry?
- Can we see the liquid phase?
- Are there transient liquid structures?
- How fast does melting take place?

## XFEL Grain Boundary Melting ?



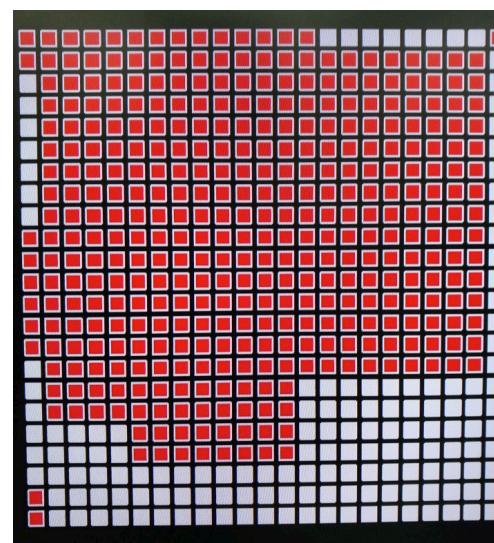
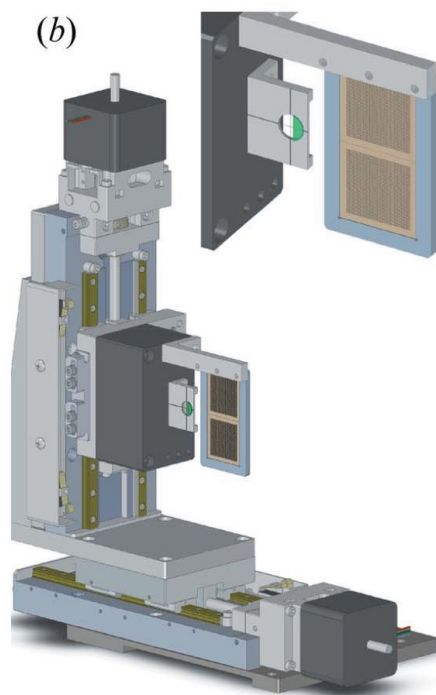


I. K. Robinson NX-school-2026

# Scan Stage for MAXIC chamber

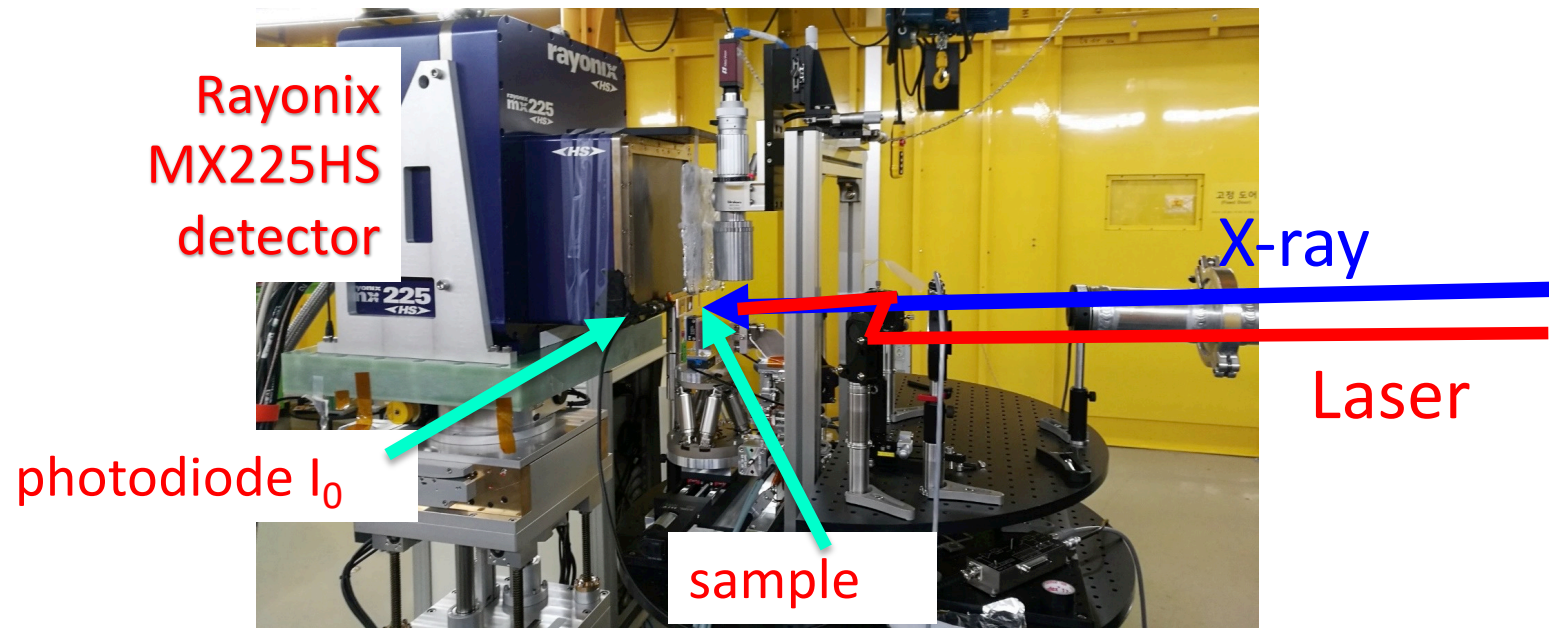
Changyong Song et al, J. Appl. Cryst. **47** 188 (2014)

Daewoong Nam, scanning software

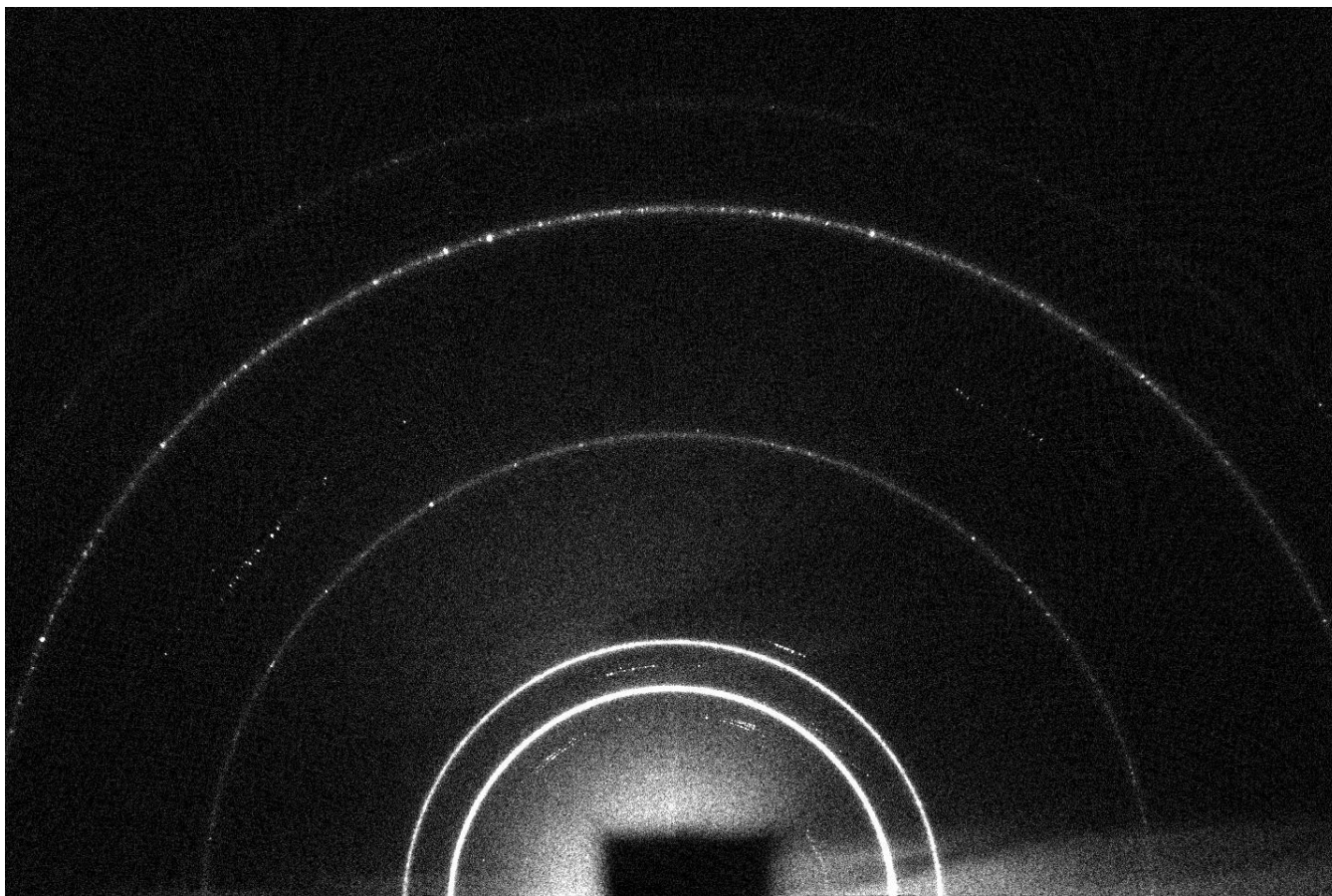


23x23 windows

# Experimental set-up at PAL-XFEL September 2017

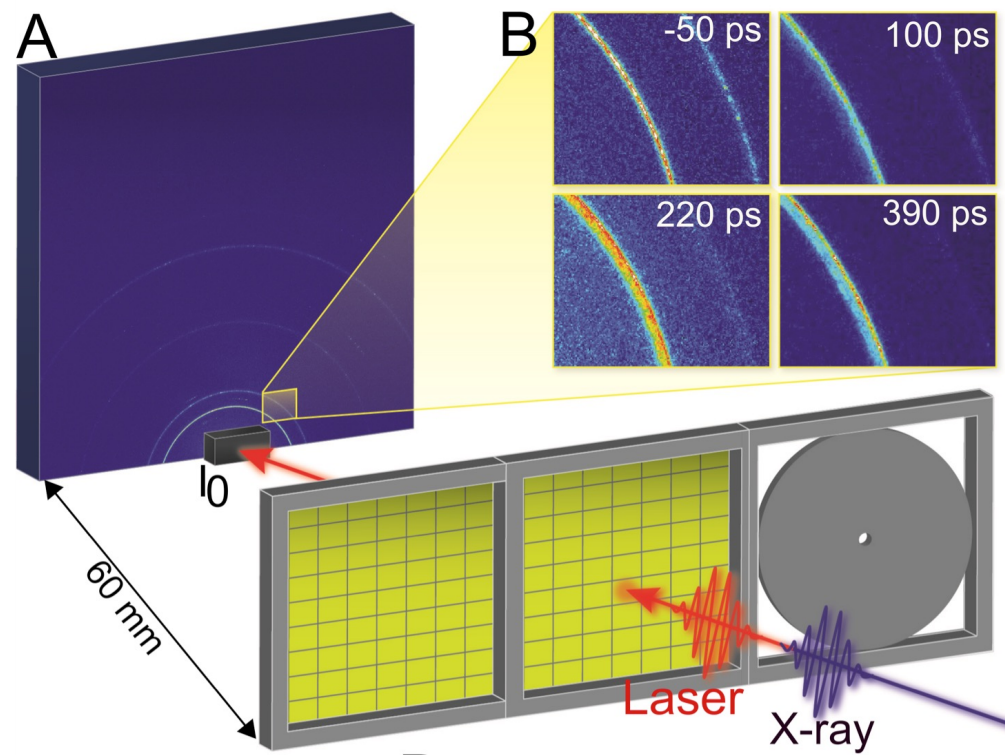


## Raw data #397 300nm film

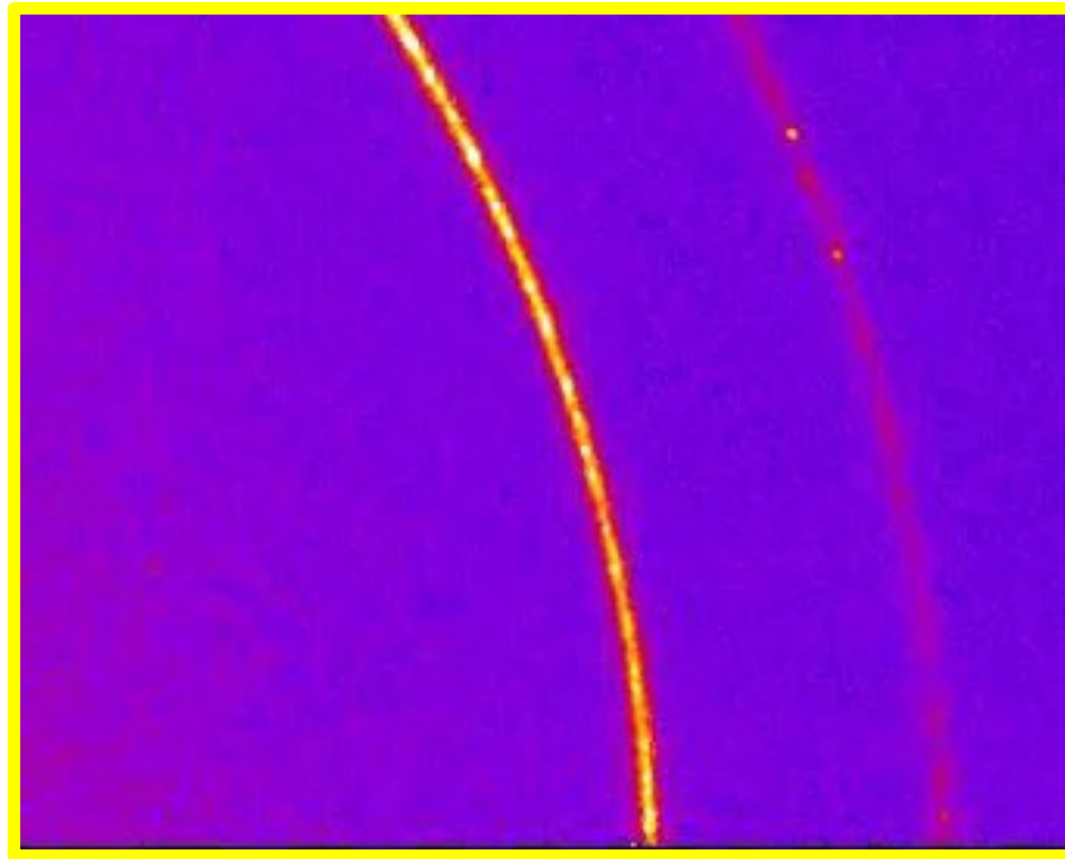


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# Powder Diffraction Geometry

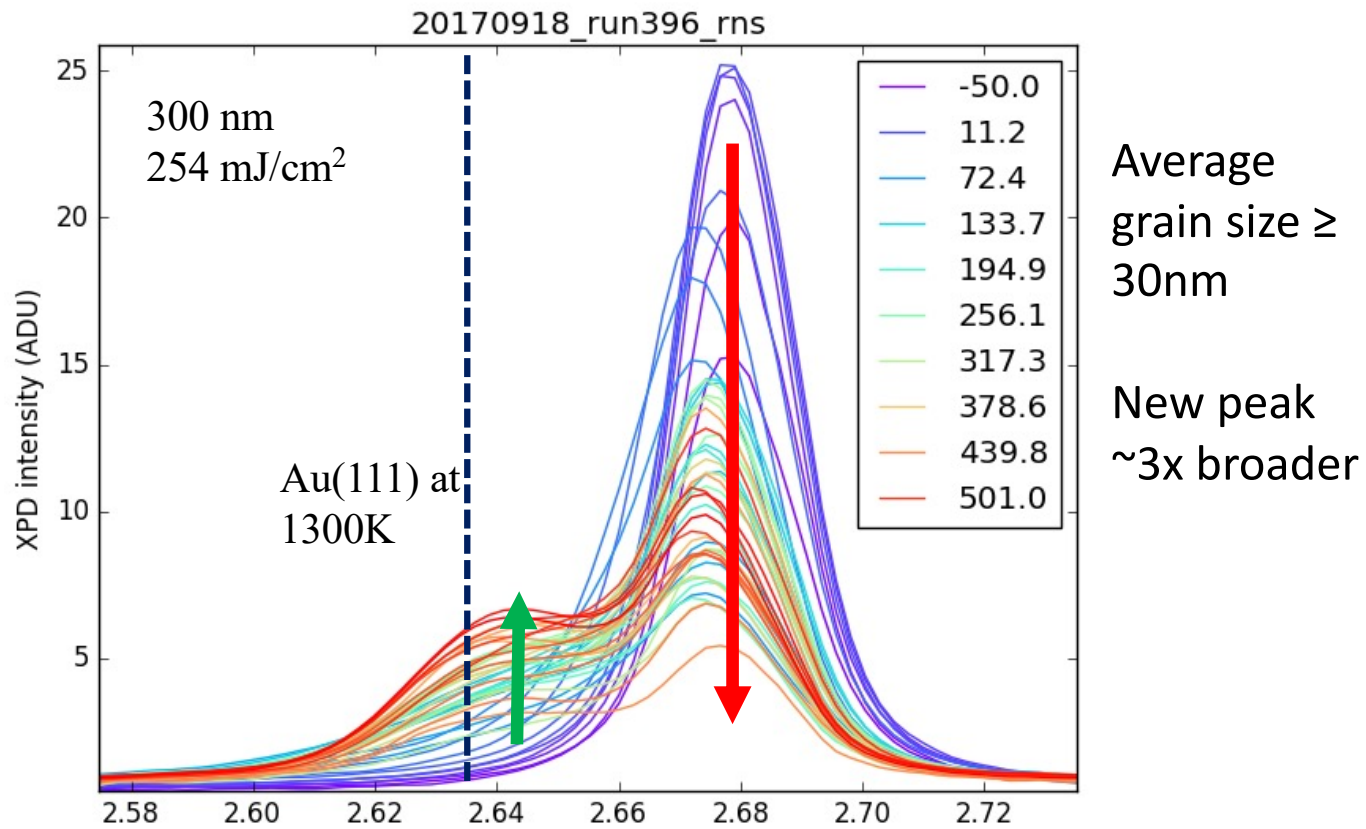


Raw data #396 300nm film, 254 mJ/cm<sup>2</sup>



I. R. ROBINSON - NIA-SCHOOL-2020

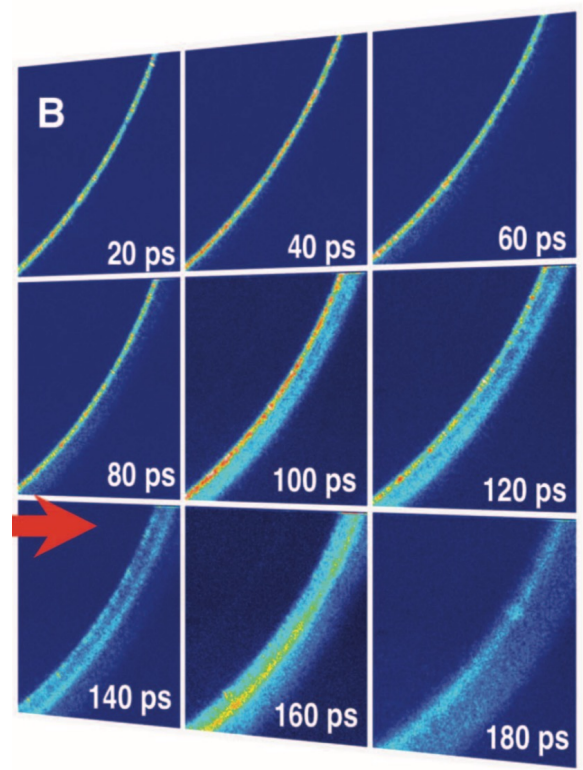
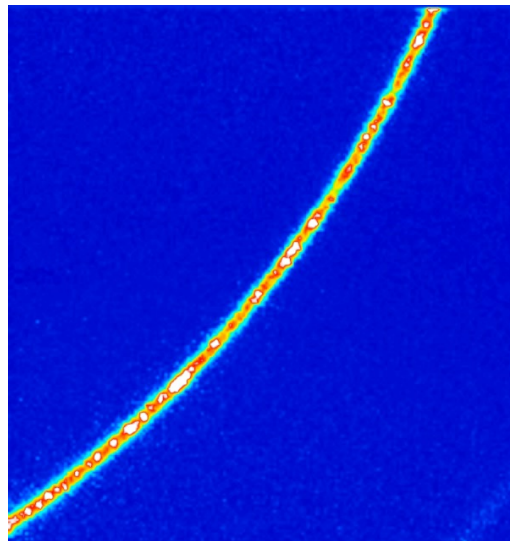
# PyFAI integration around ring



# Thin Film of Cu at CXI, LCLS

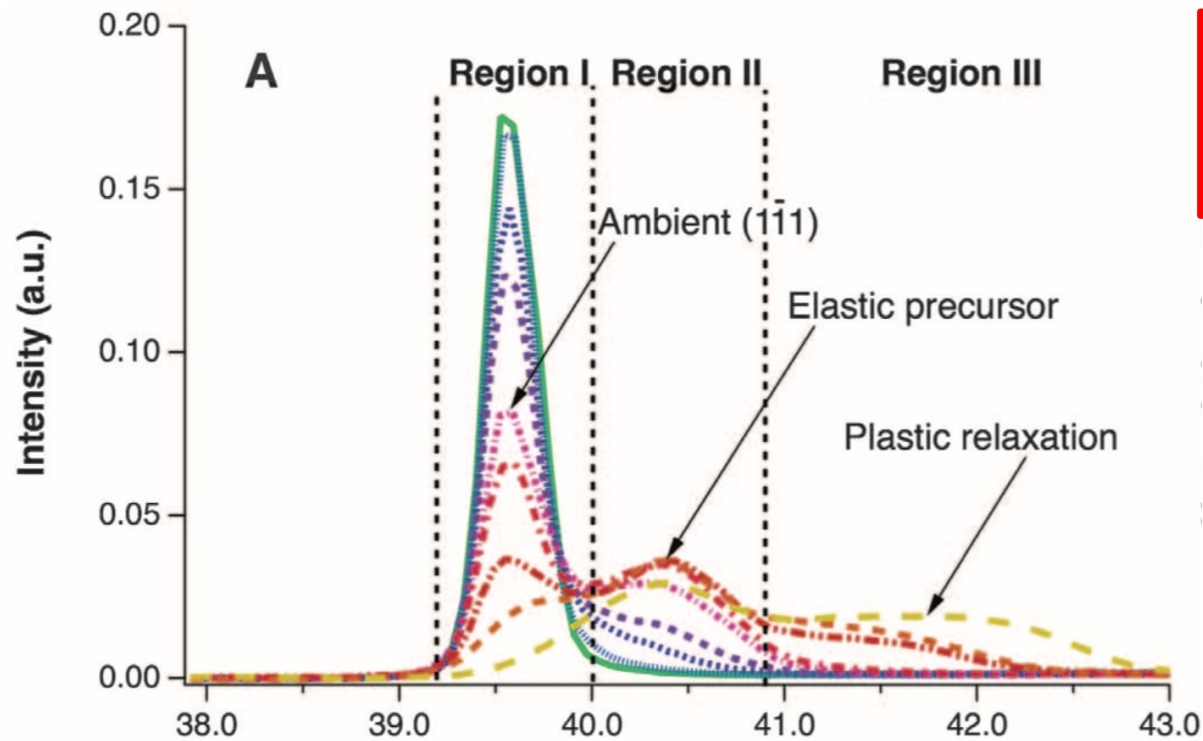
Despina Milathianaki et al, Science **342** 220 (2013)

Laser  
38 J/cm<sup>2</sup>  
170ps



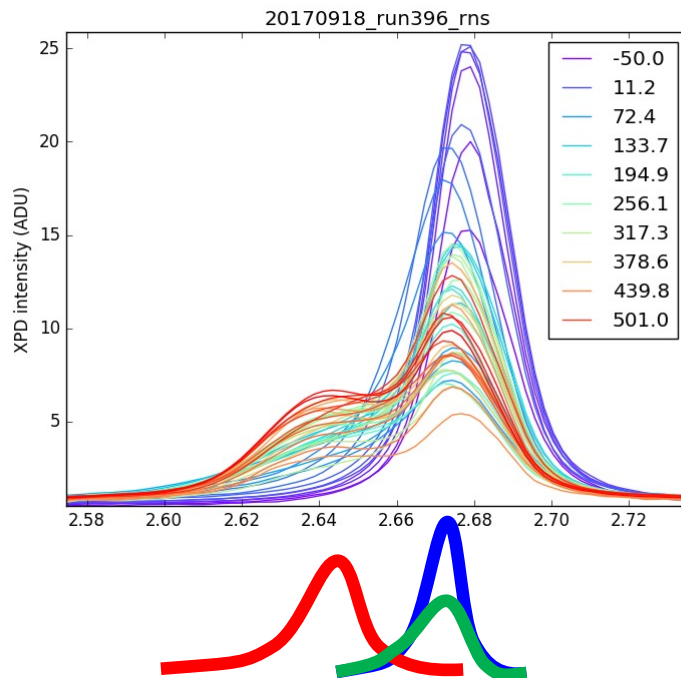
# Thin Film of Cu at CXI, LCLS

D. Milathianaki et al, Science **342** 220 (2013)



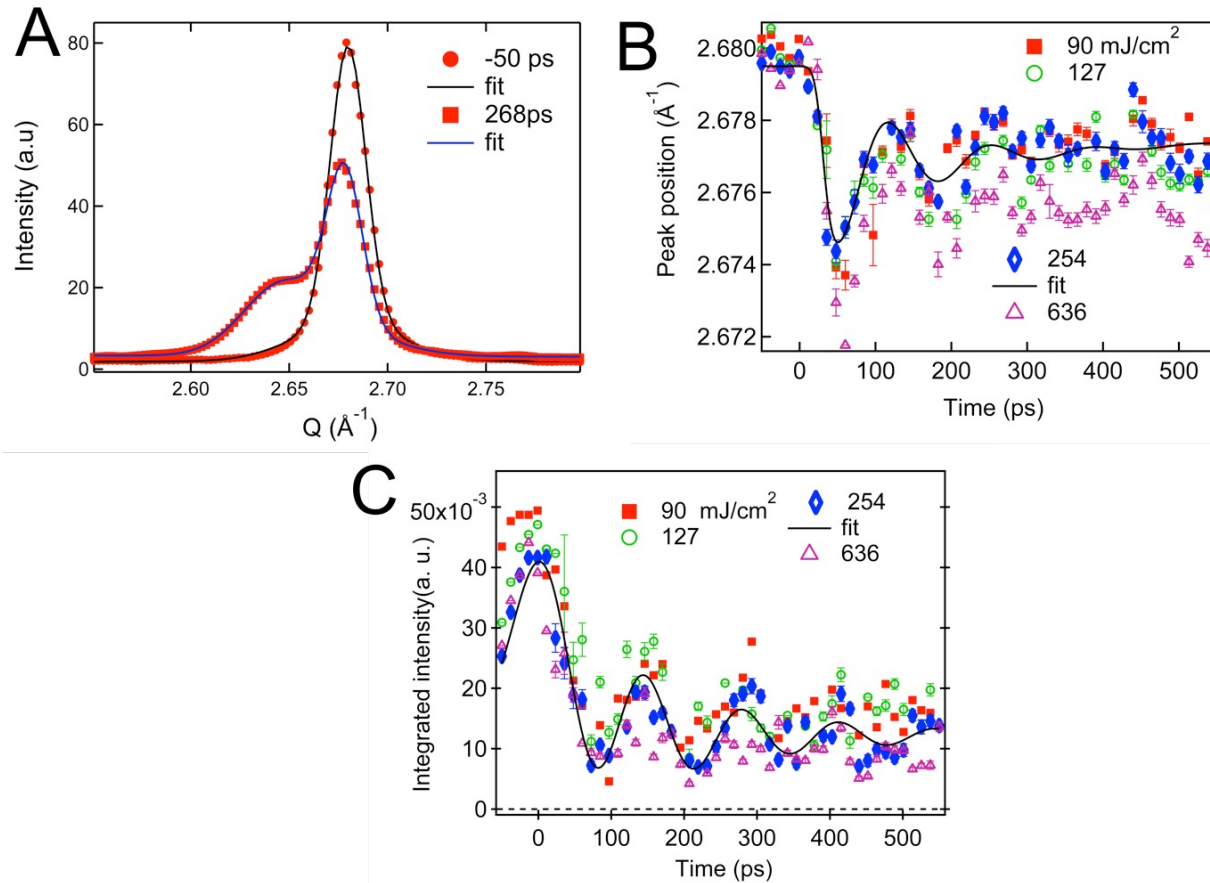
Laser  
38 J/cm<sup>2</sup>  
170ps

# Gaussian fitting procedure



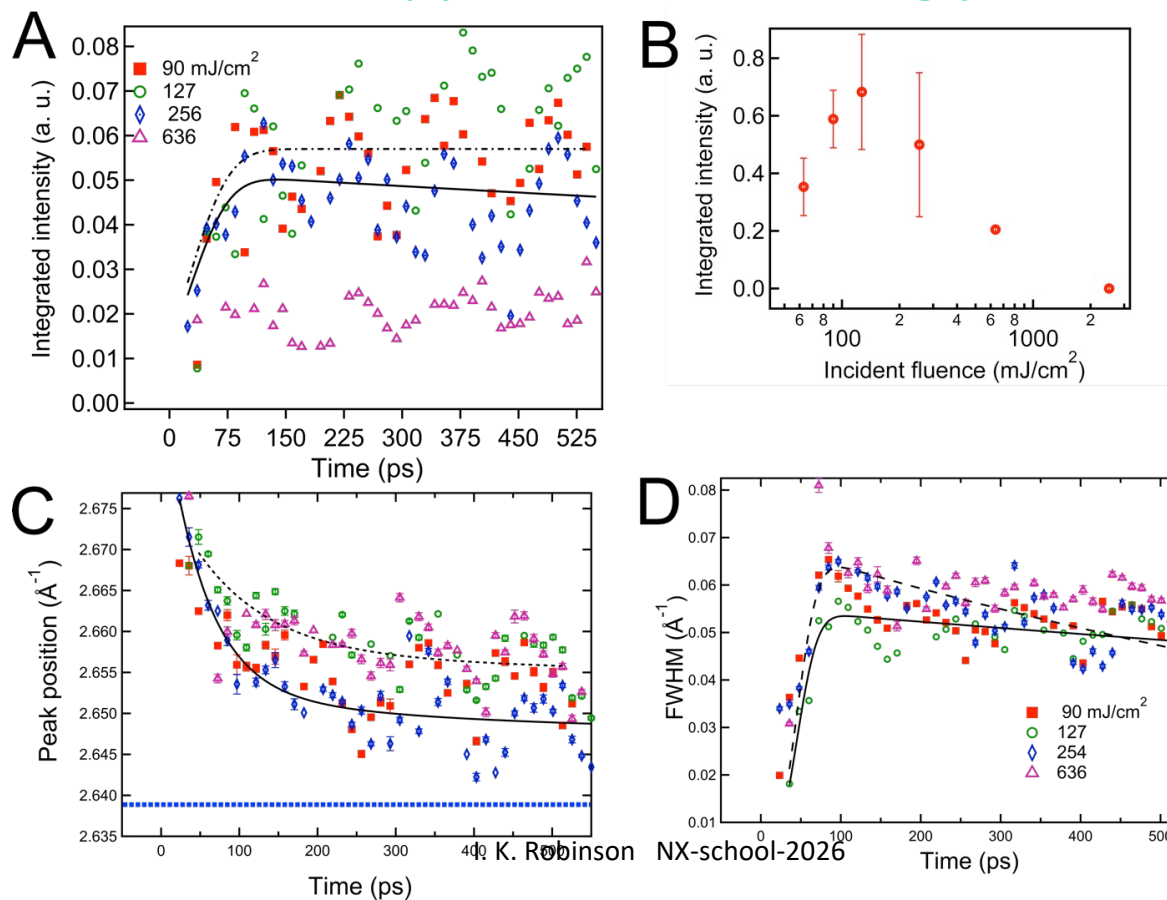
- Crystal Peaks (2x)
  - fixed widths
  - fixed height ratio
  - variable position
- “New” Peak
  - variable width
  - variable height
  - variable position

# Response of the Crystal 111 peak



# Response of New Melt-front Peak

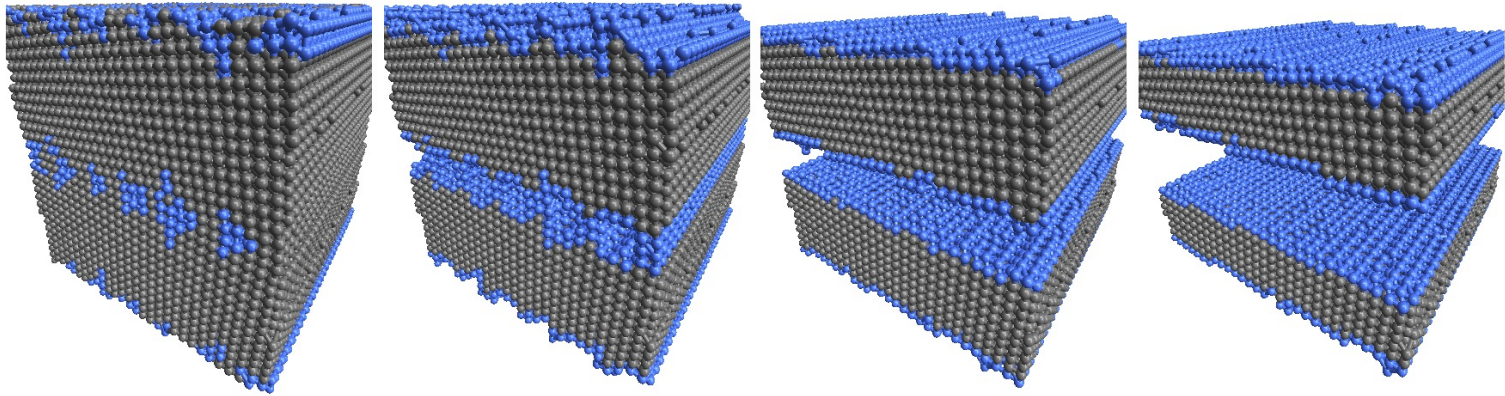
## Material trapped at the melting point



K. Robinson NX-school-2026

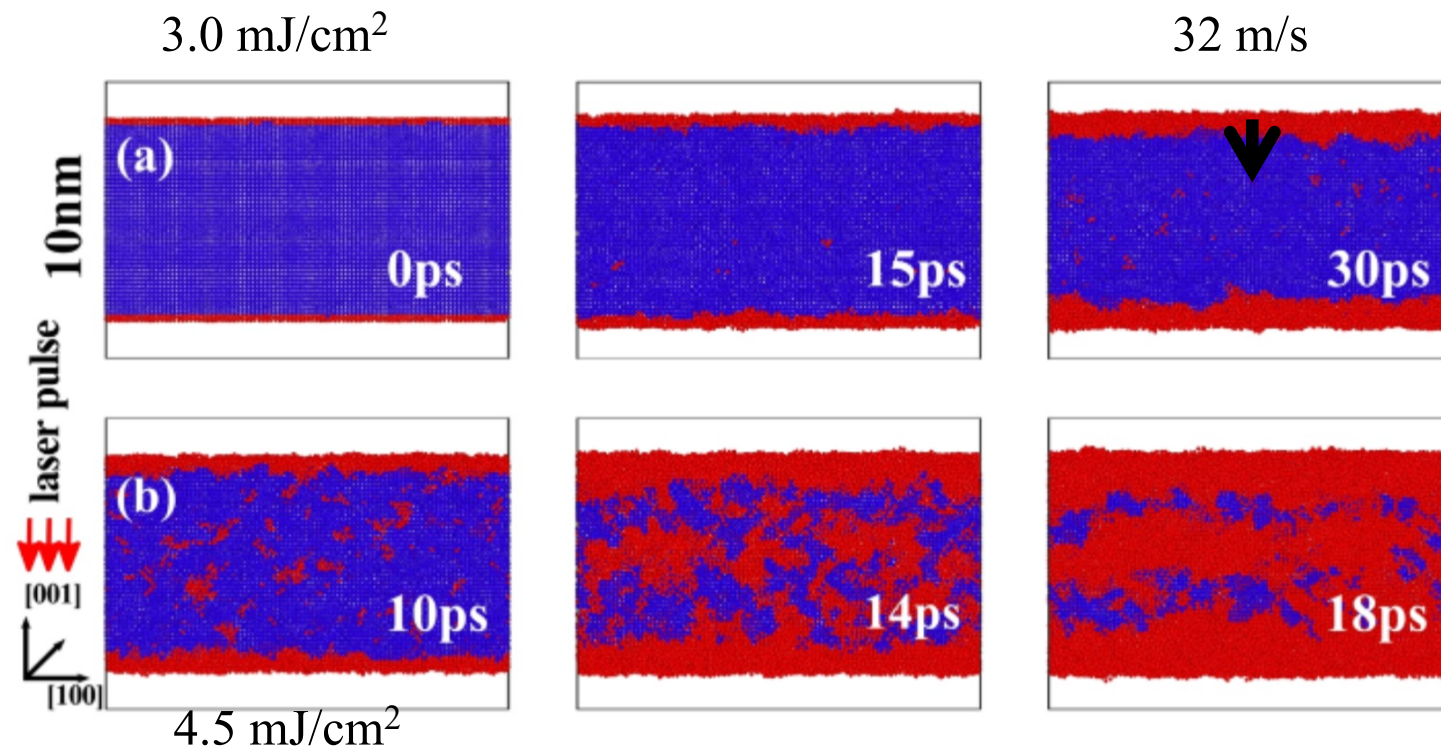
# Force-Field Simulation of GB melting

J. Berry, K. Elder and M. Grant, PRB 77 224114 (2008)

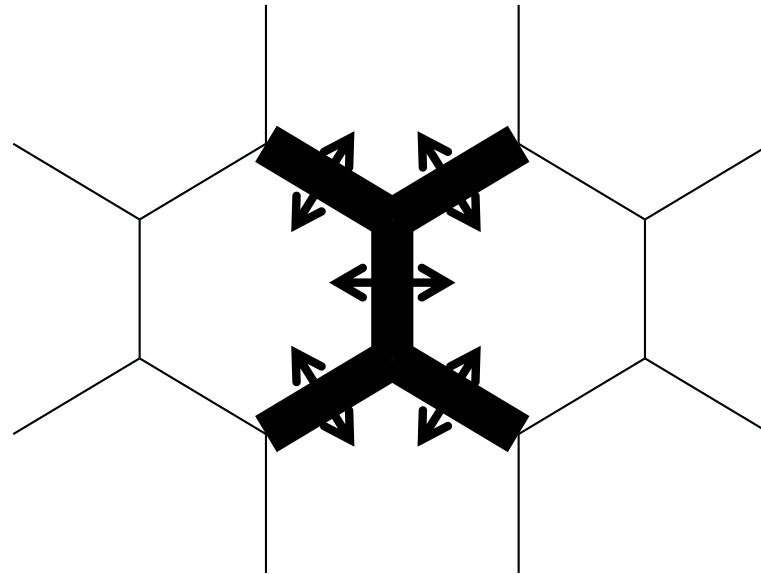
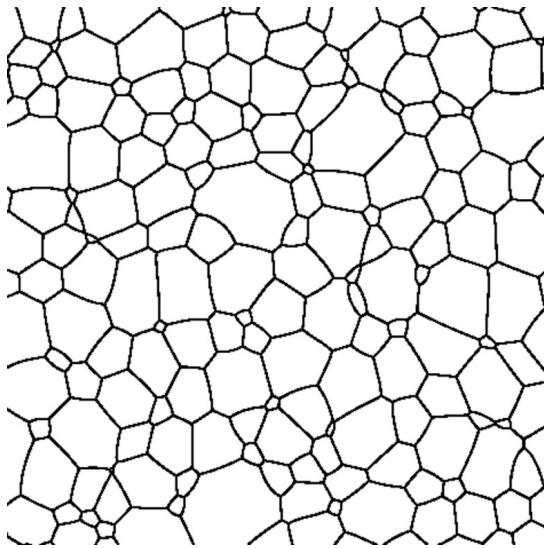


# 2TM-MD (EAM) simulation Au slab

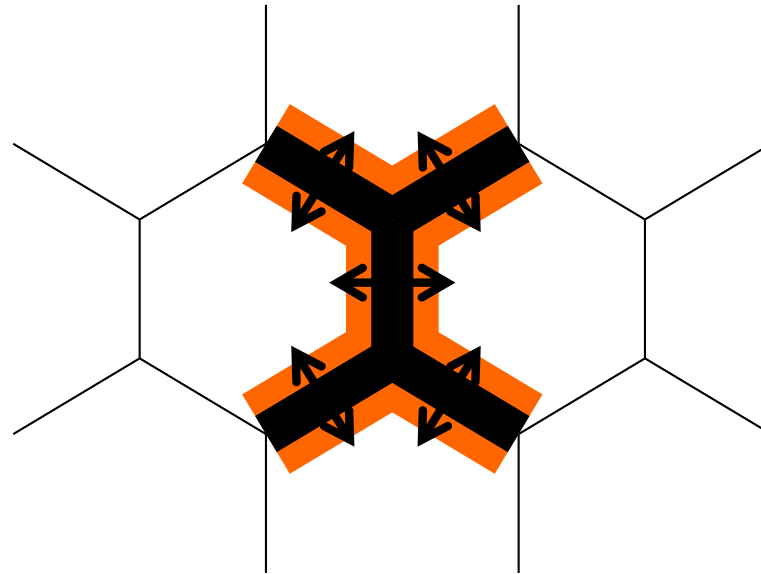
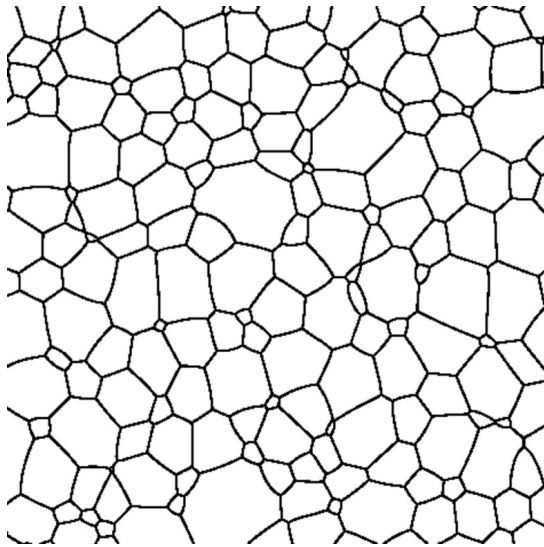
Giret et al, APL **103** 253107 (2013)



# Grain Boundary Melting

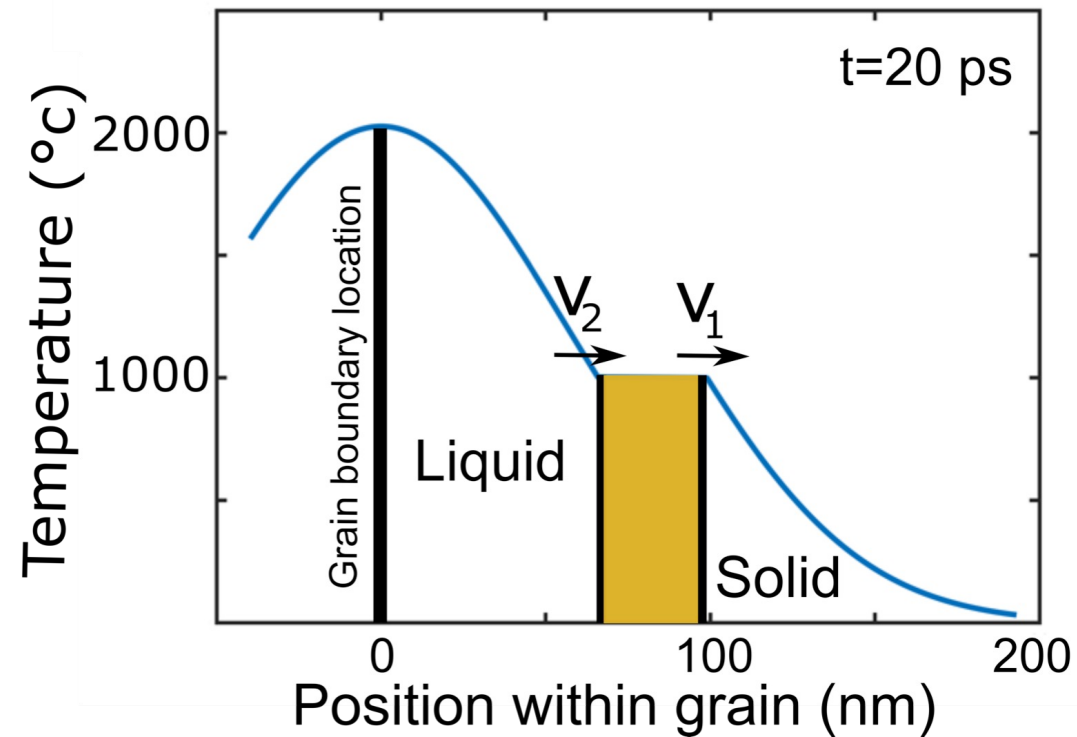


# Grain Boundary Melting



# Grain Boundary Induced Melting

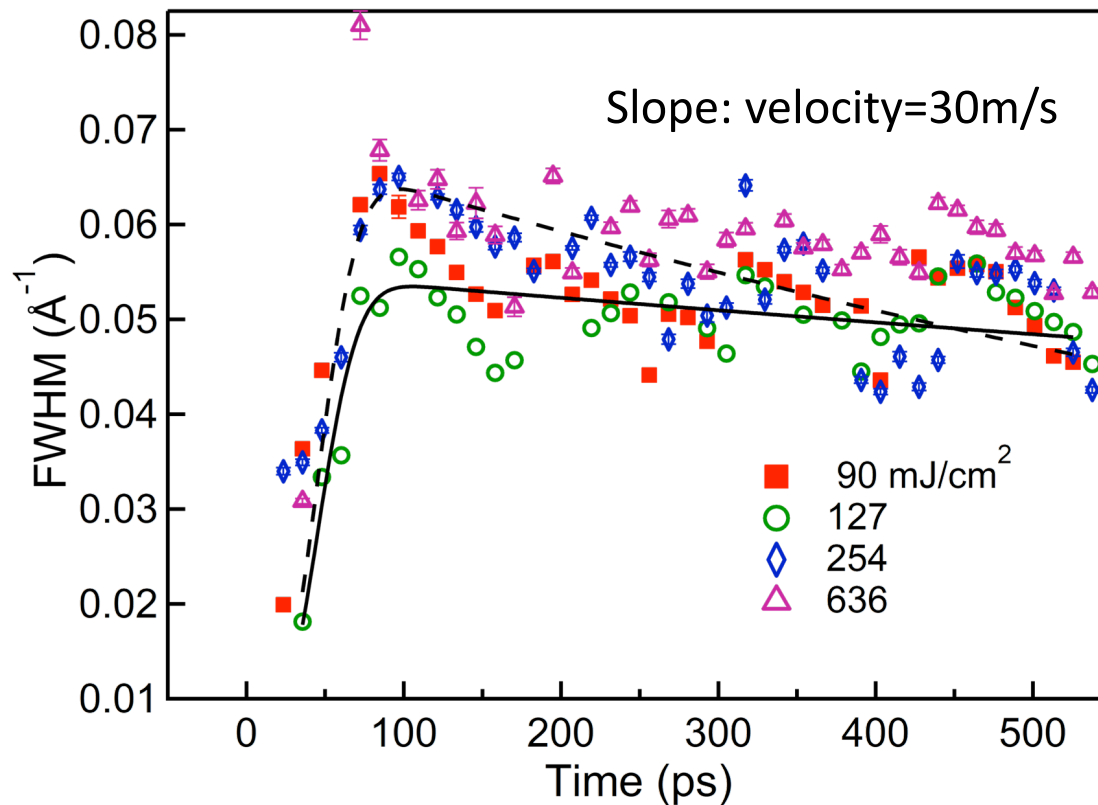
T. A. Assefa et al Science Advances 6 eaax2445 (2020)



I. K. Robinson NX-school-2026

# Width of new “Melt-Front” Peak

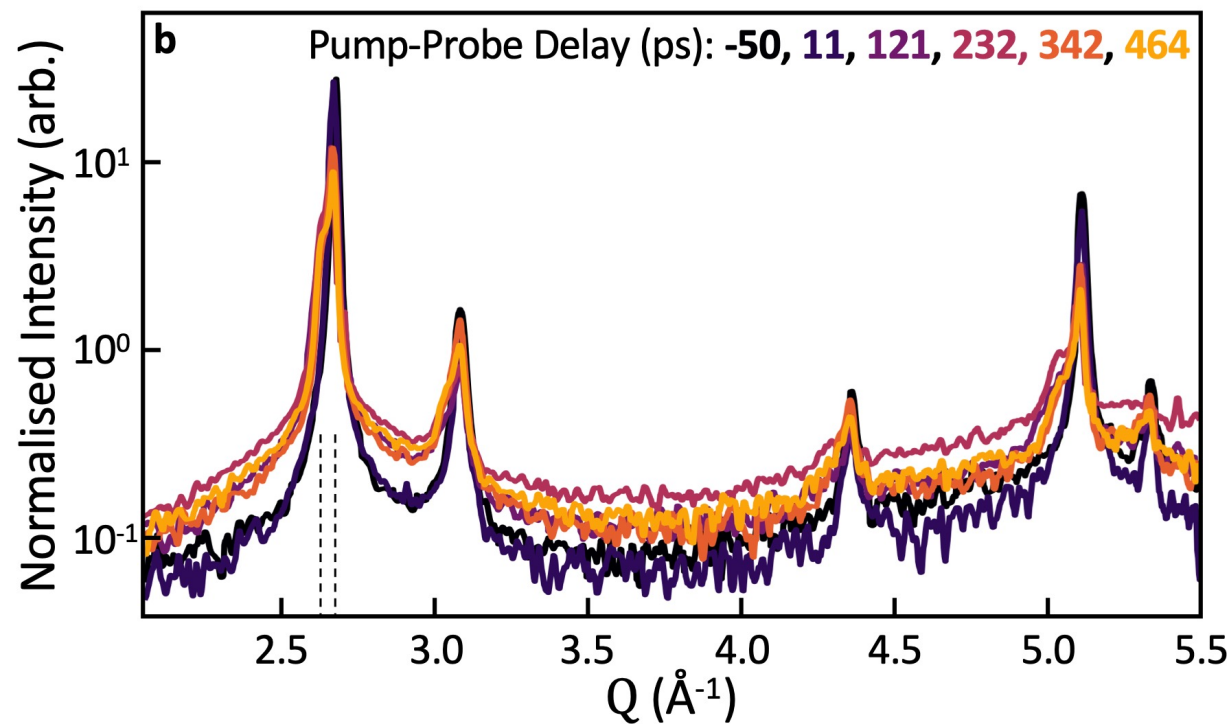
T. A. Assefa et al Science Advances 6 eaax2445 (2020)



$$\tau = 2\pi / \Delta Q$$
$$= 12 \text{ nm}$$

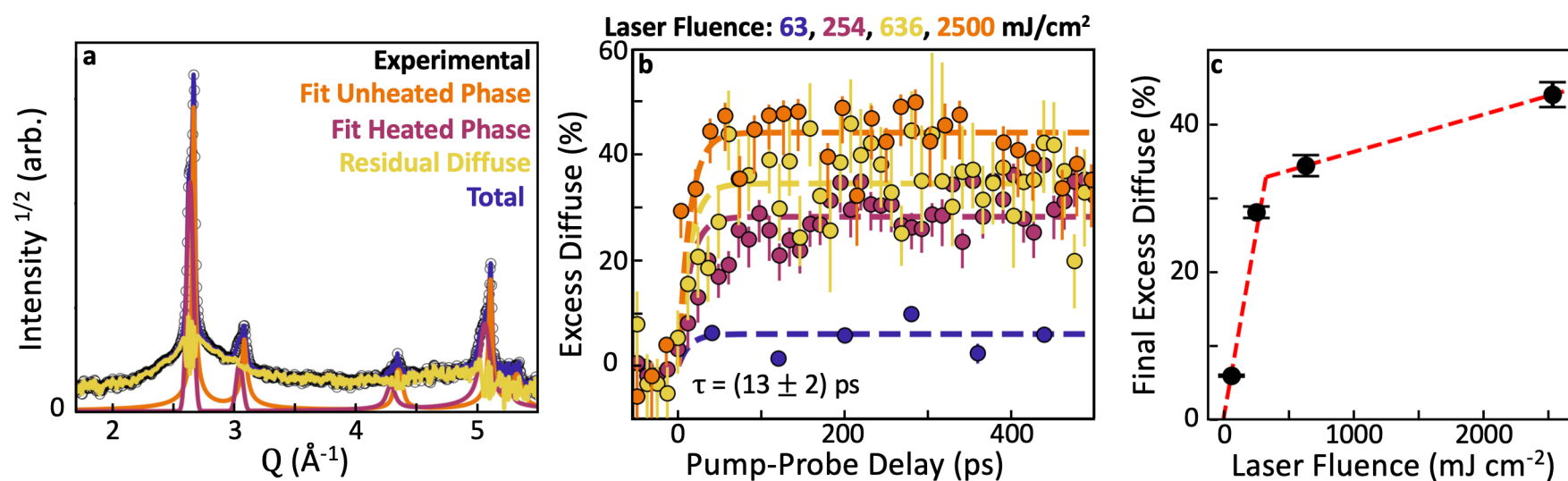
# Liquid structure factor

I. K. Robinson, et al, IUCrJ 10 656–661 (2023)

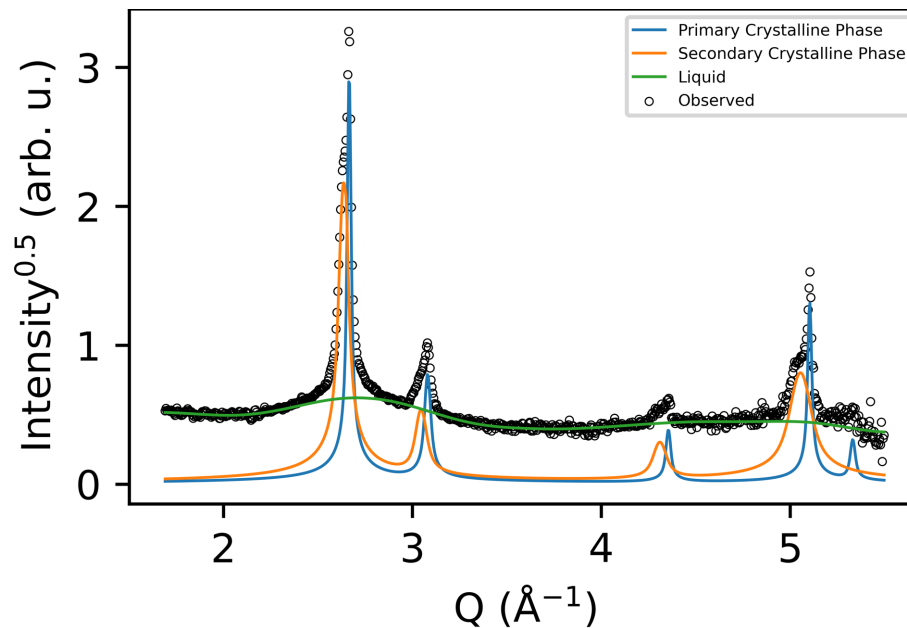


# Extracting Liquid structure factor

I. K. Robinson, et al, IUCrJ 10 656–661 (2023)



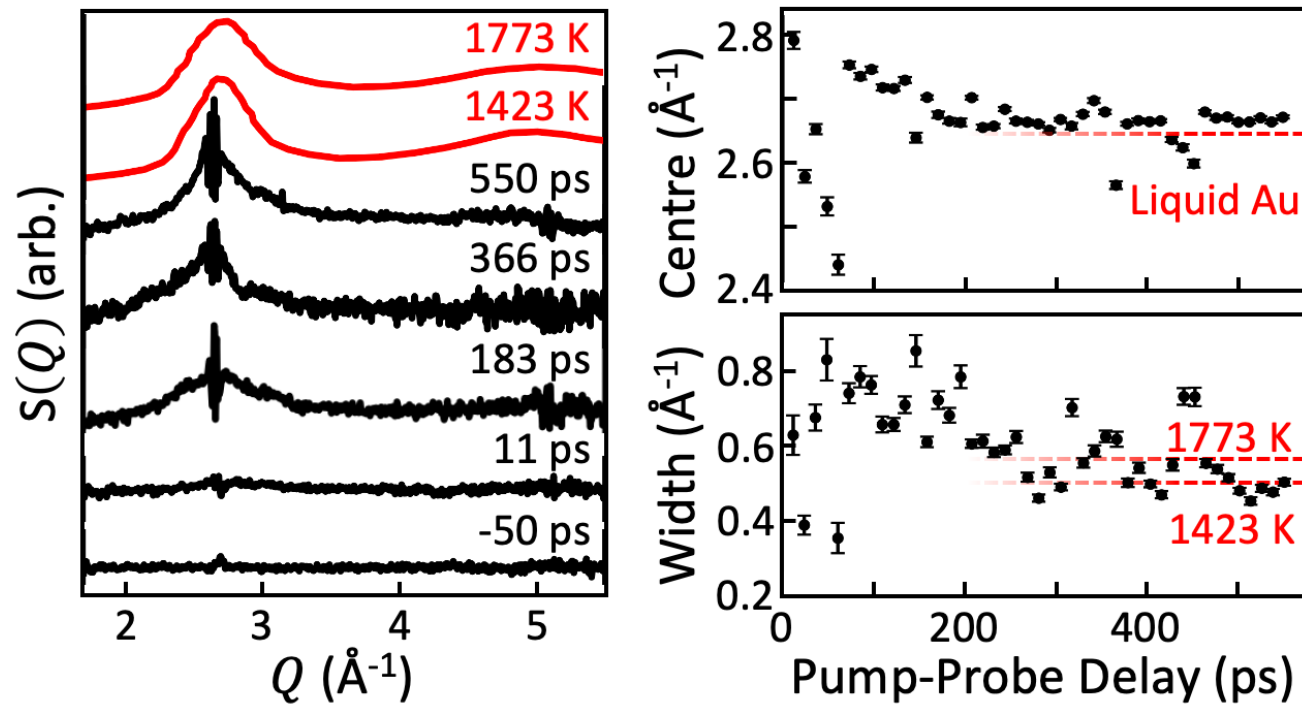
# Extracting the diffuse signal



- Raw data plotted, 231 ps, square root intensity scale
- Unconstrained peak fitting approach
- Remove Bragg components

# Liquid structure factor

I. K. Robinson, et al, IUCrJ 10 656–661 (2023)



# Melt-front description of melting in polycrystalline materials

- Laser induced disorder
- Three phases: liquid, solid, melt
- 2-phase inhomogeneous melting
- Energy transfer at Grain Boundaries
- Interface melting like nanoparticles
- Measured structure of Melt Front

# Thin-film melting collaborators

Tadesse Assefa  
Jack Griffiths  
Yue Cao  
Emil Bozin  
Rob Koch  
Pavol Juhas  
Simon Billinge

Dongjin Kim  
Sungwon Kim  
Hyunjung Kim  
Changyong Song

Sunam Kim  
Jae Hyuk Lee  
Yongsam Kim  
Jaeku Park  
Sang-Youn Park  
Intae Eom  
Hyojung Hyun  
Tae-Yeong Koo  
Jaehun Park  
Daewoong Nam  
Sang Soo Kim



# X-ray Scattering Summary

- Scattering from Matter
- Powder diffraction methods “XRD”
  - Cora Lind-Kovacs’ lecture
- Surface diffraction and surface structure
  - Paul Miceli’s lecture
- Extension to Coherent X-ray Diffraction
  - Xiaojing Huang’s lecture
- XFEL study of thin film melting
  - Meng Liang’s lecture