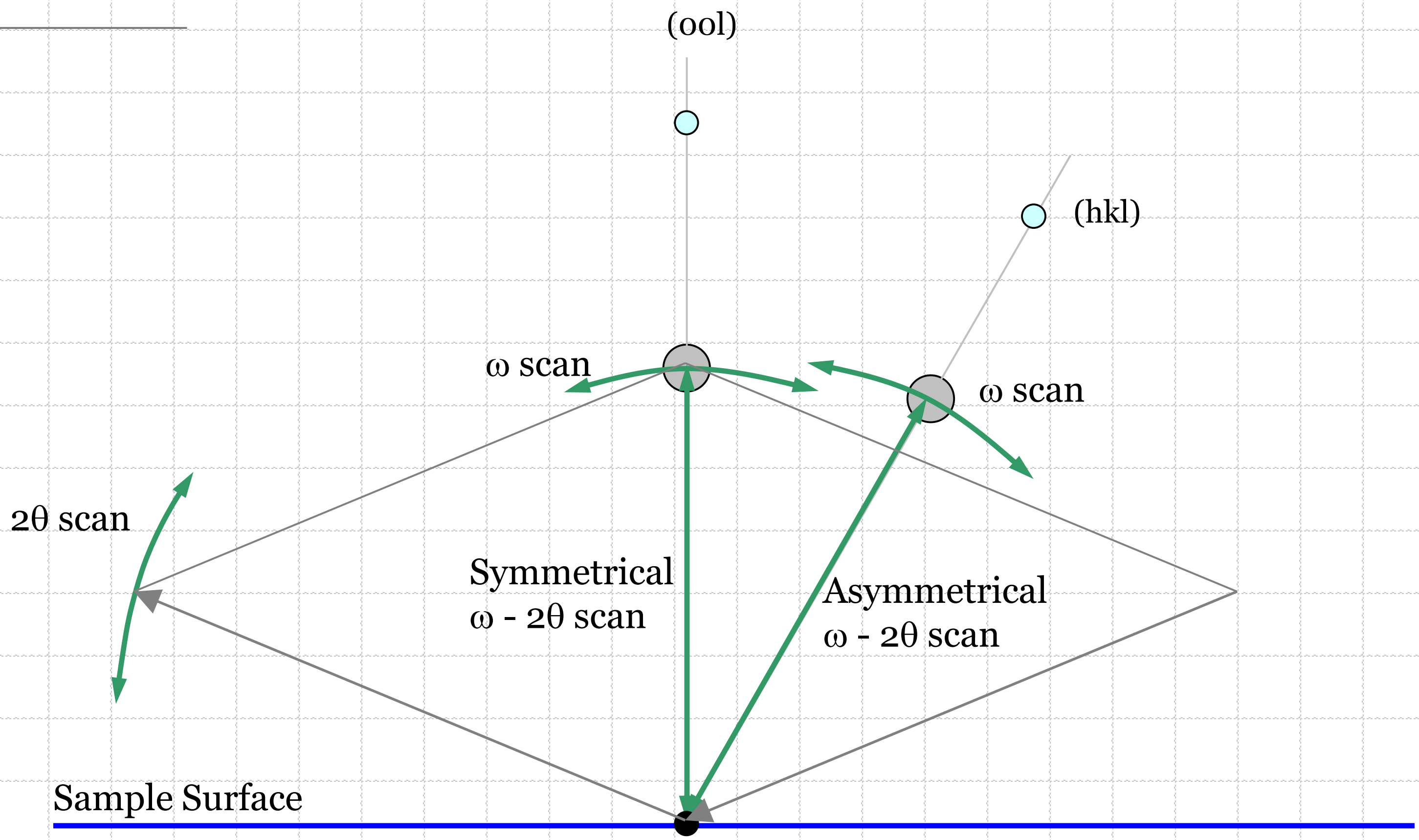
High-Resolution XRD – 2

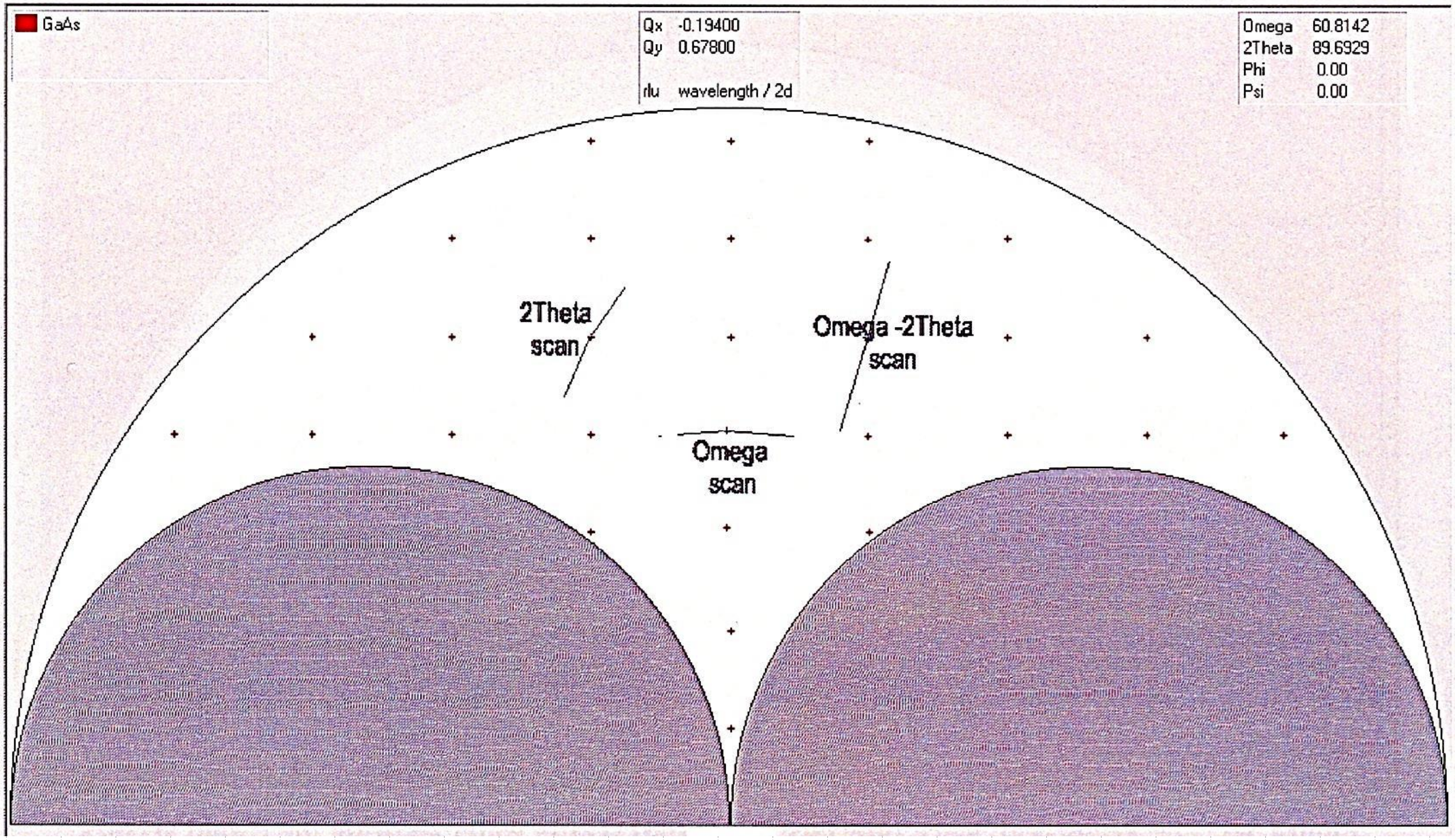
Reciprocal Lattice Mapping

Superlattices

Scan directions



Reciprocal Space

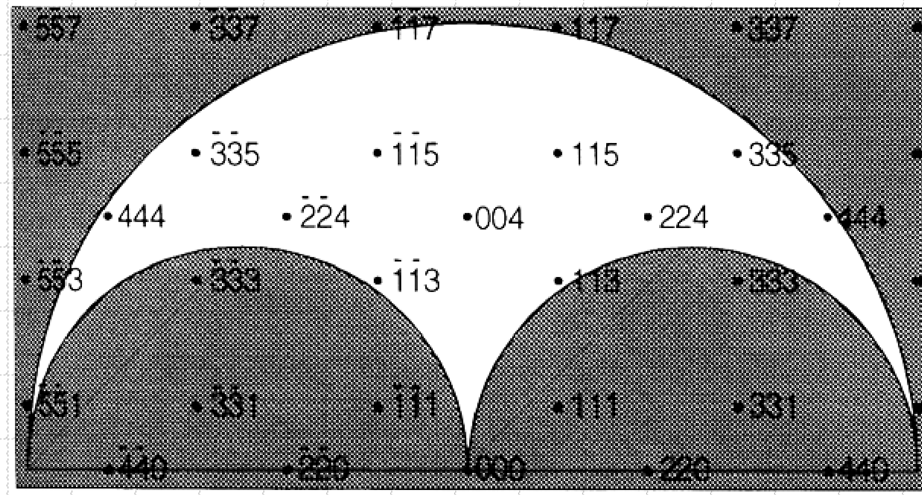


ω-scan is in the direction of an arc centered on the origin
2θ-scan is an arc along Ewald sphere circumference
ω-2θ scan is always strait line pointing away from the origin of the reciprocal space

Reciprocal Space

$$Q_x = \frac{1}{2}\{\cos\omega - \cos(2\theta - \omega)\}$$

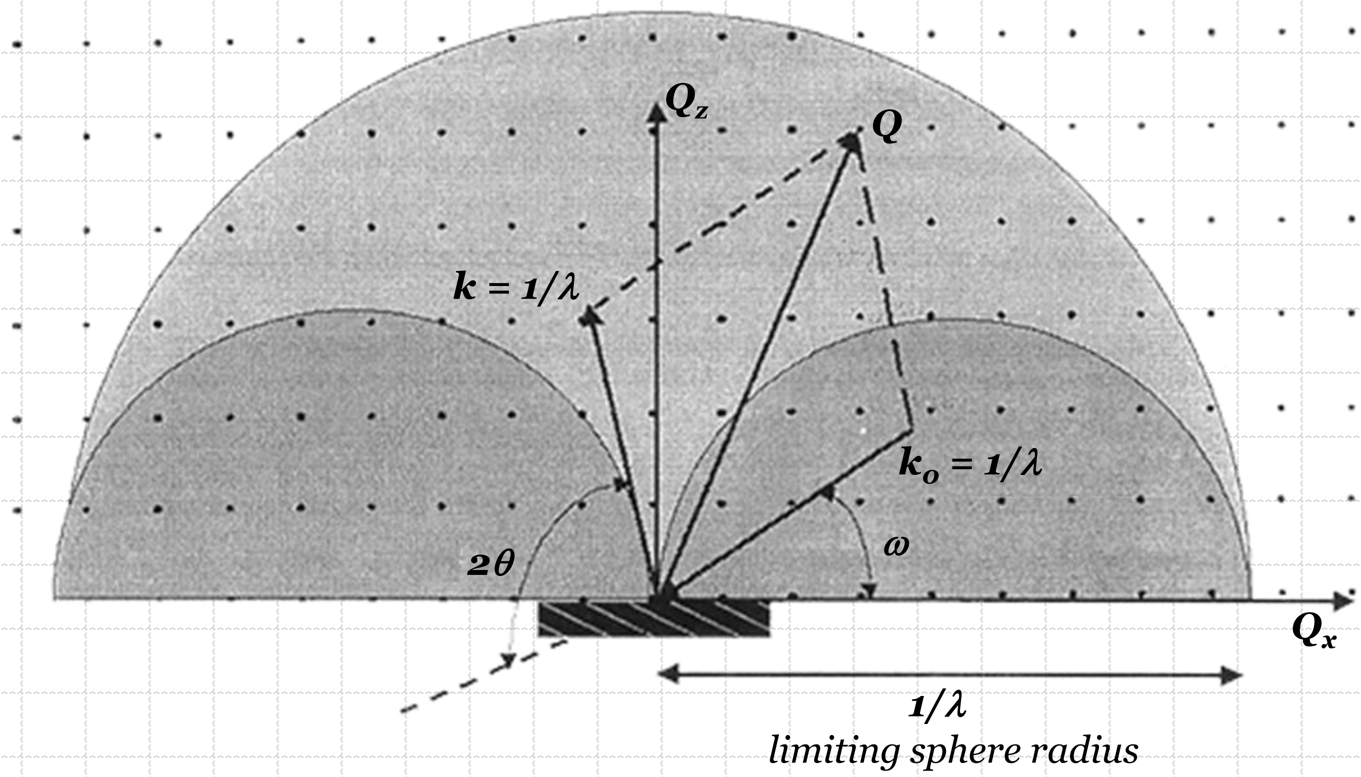
$$Q_z = \frac{1}{2}\{\sin\omega + \sin(2\theta - \omega)\}$$



$$q = \frac{4\pi sin\theta}{\lambda}$$

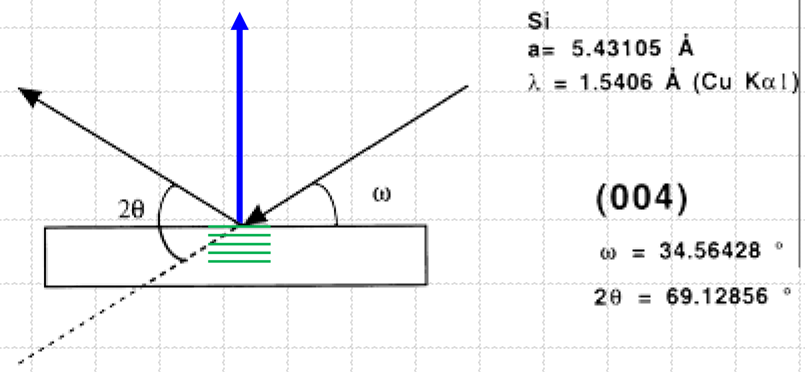
$$q_x = \frac{4\pi Q_x}{\lambda}$$

$$q_z = \frac{4\pi Q_x}{\lambda}$$

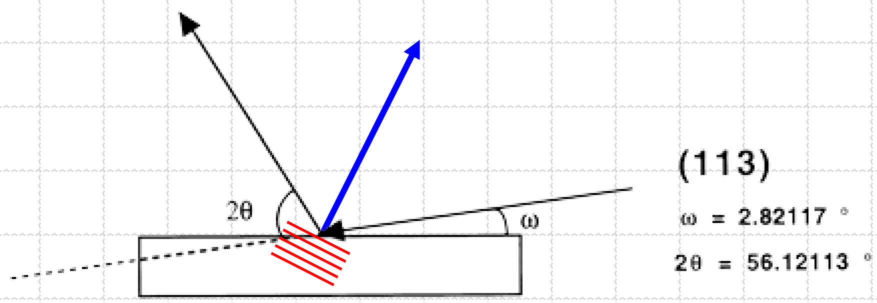


Symmetric and Asymmetric Scans

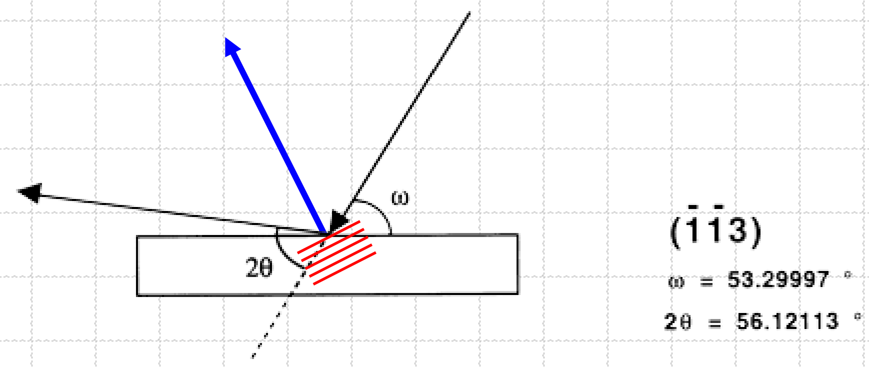
- Symmetric scan



- Asymmetric scan. Low angle of incidence.

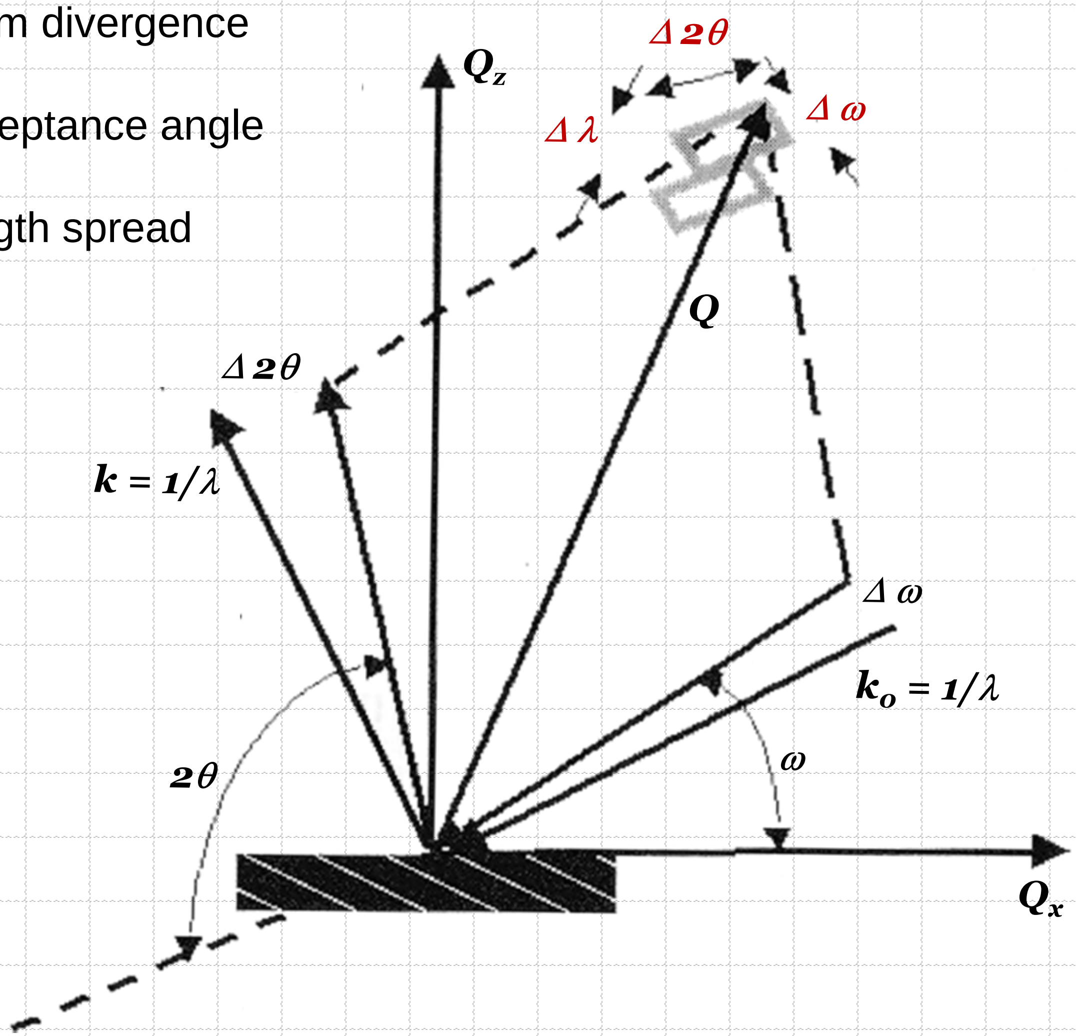


- Asymmetric scan. High angle of incidence.



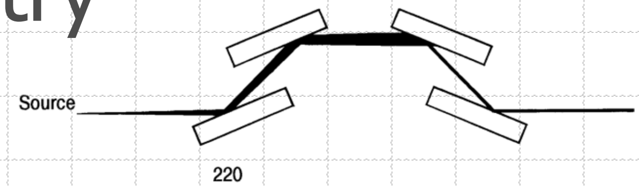
Reciprocal Space – instrument resolution

- Influence of incident beam divergence
- Influence of detector acceptance angle
- Influence of the wavelength spread

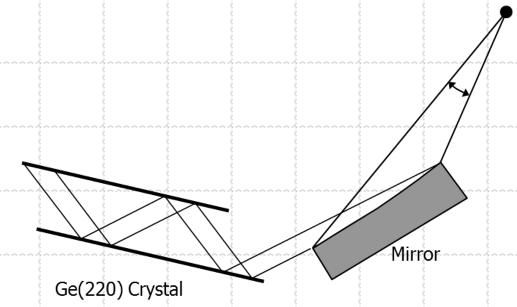


High-Resolution Diffractometry

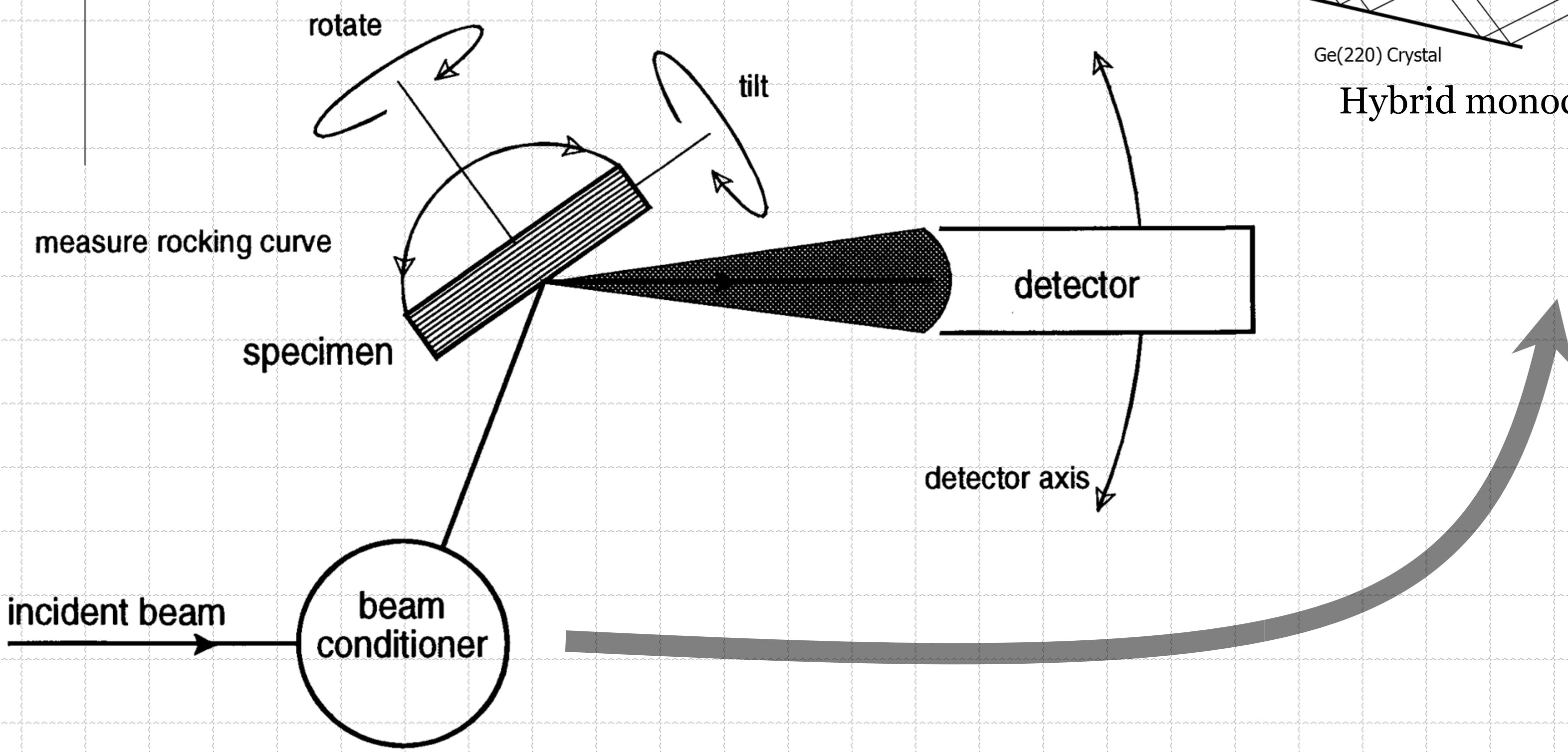
- Schematic of high resolution double-axis instrument

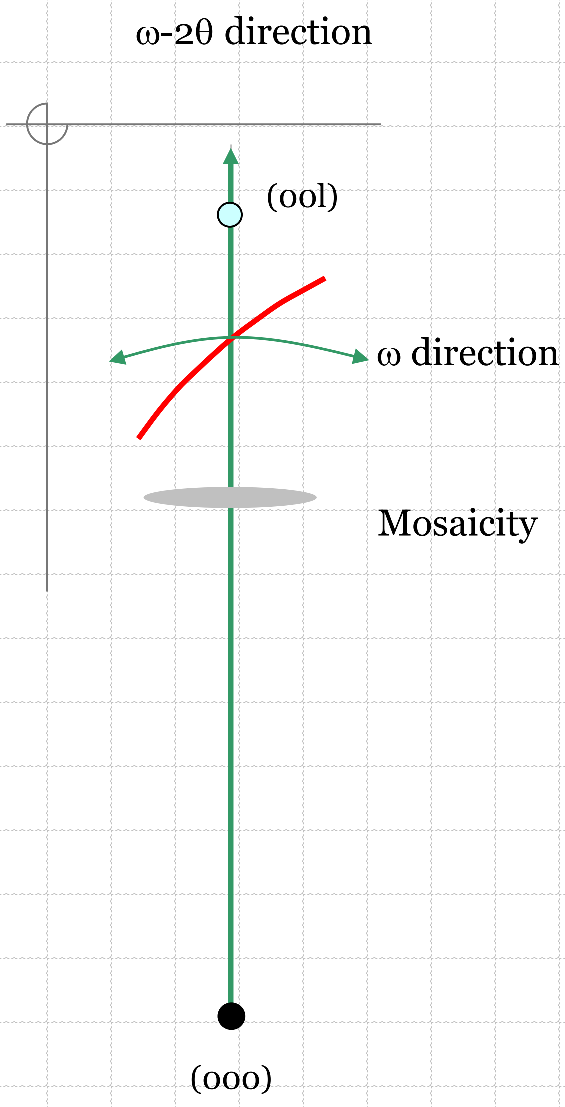


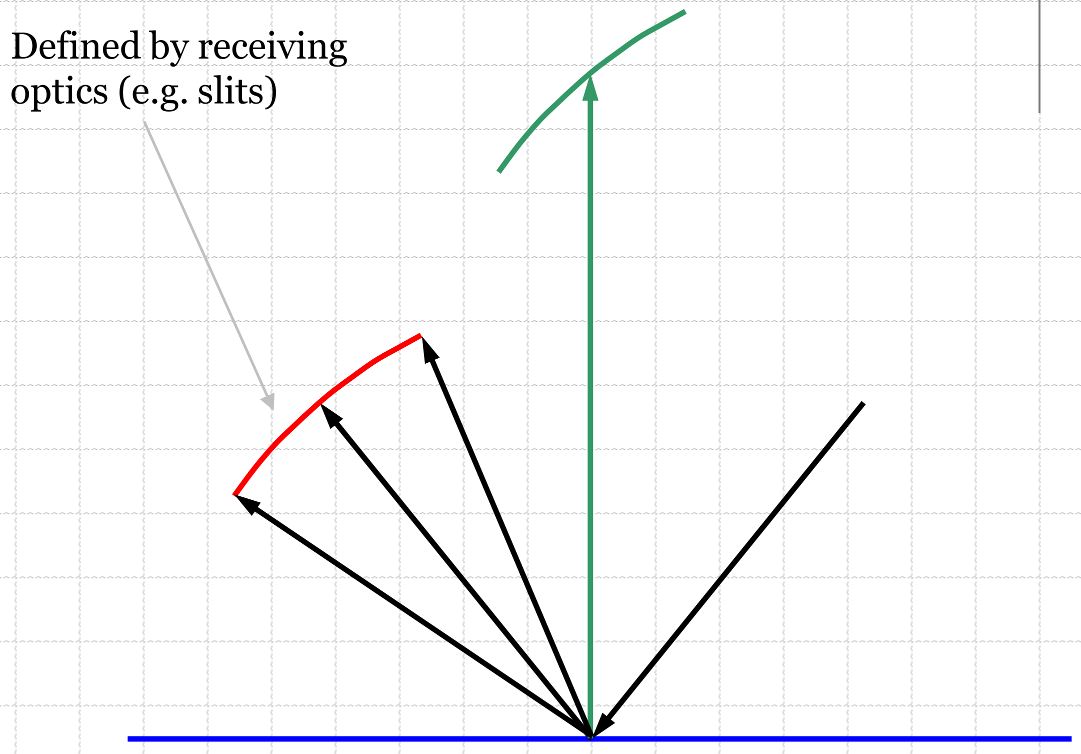
Bartels monochromator



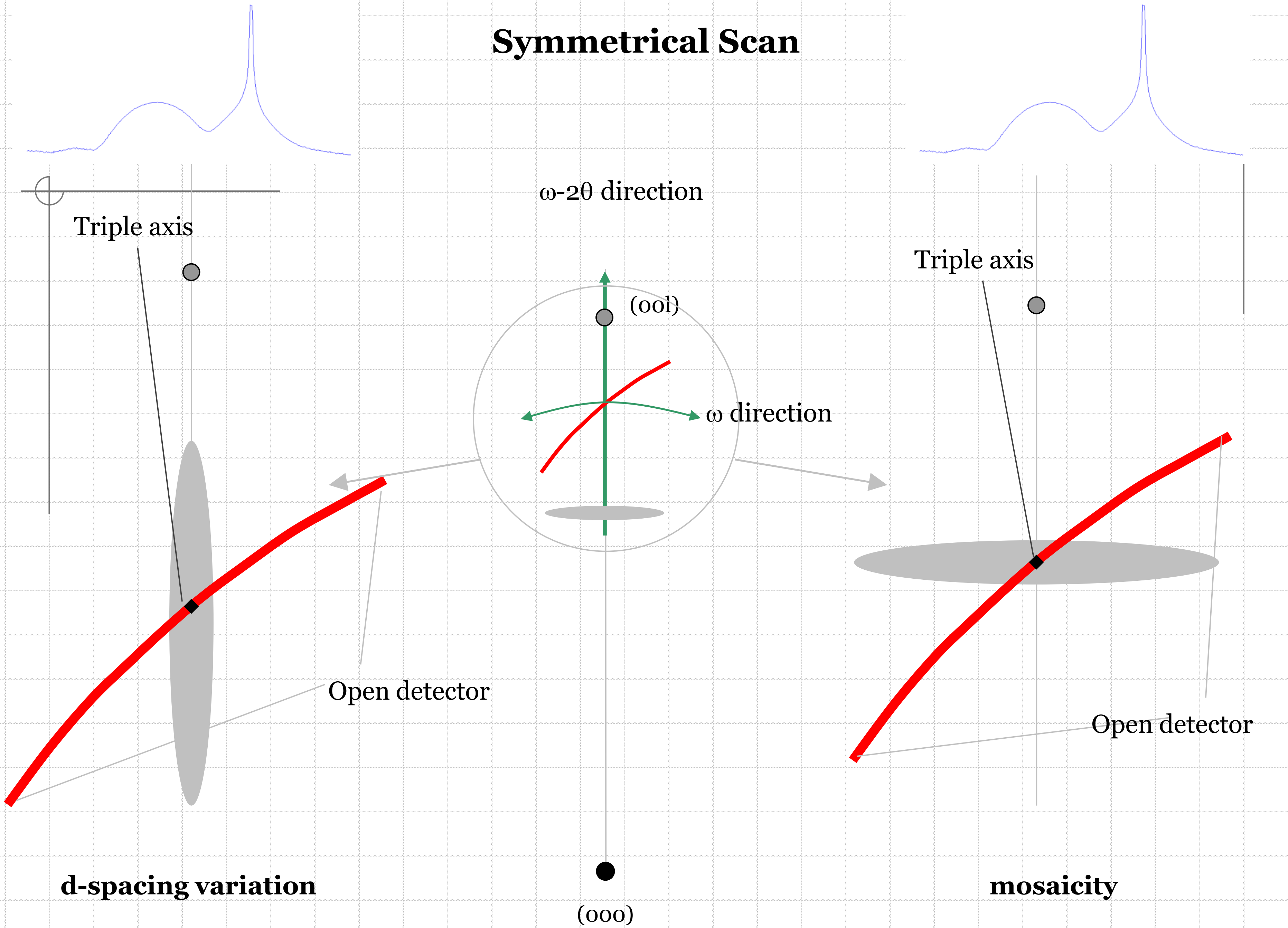
Hybrid monochromator




ω-2θ direction
(ool)
ω direction
Mosaicity
(ooo)

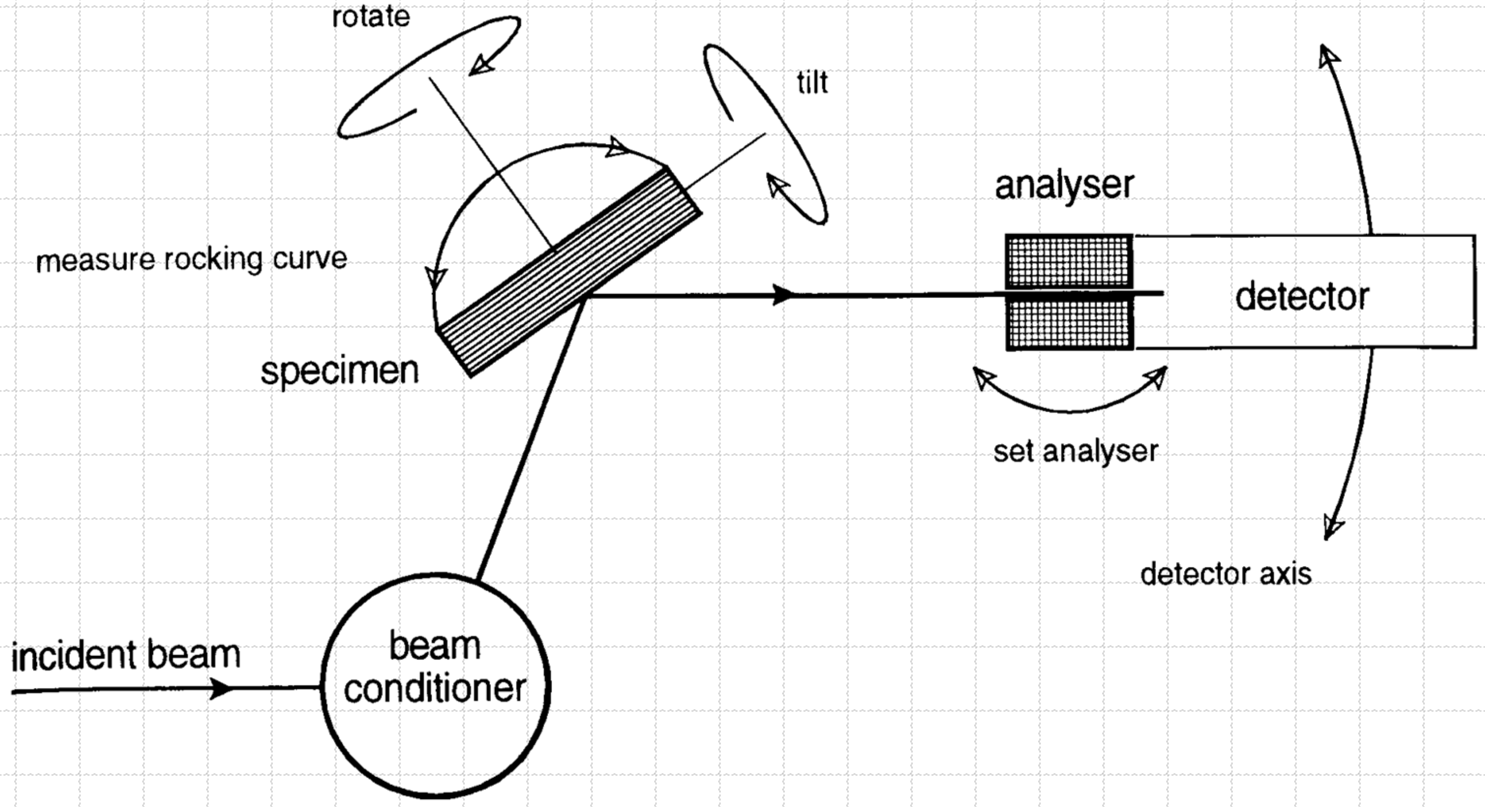

Defined by receiving
optics (e.g. slits)

Symmetrical Scan



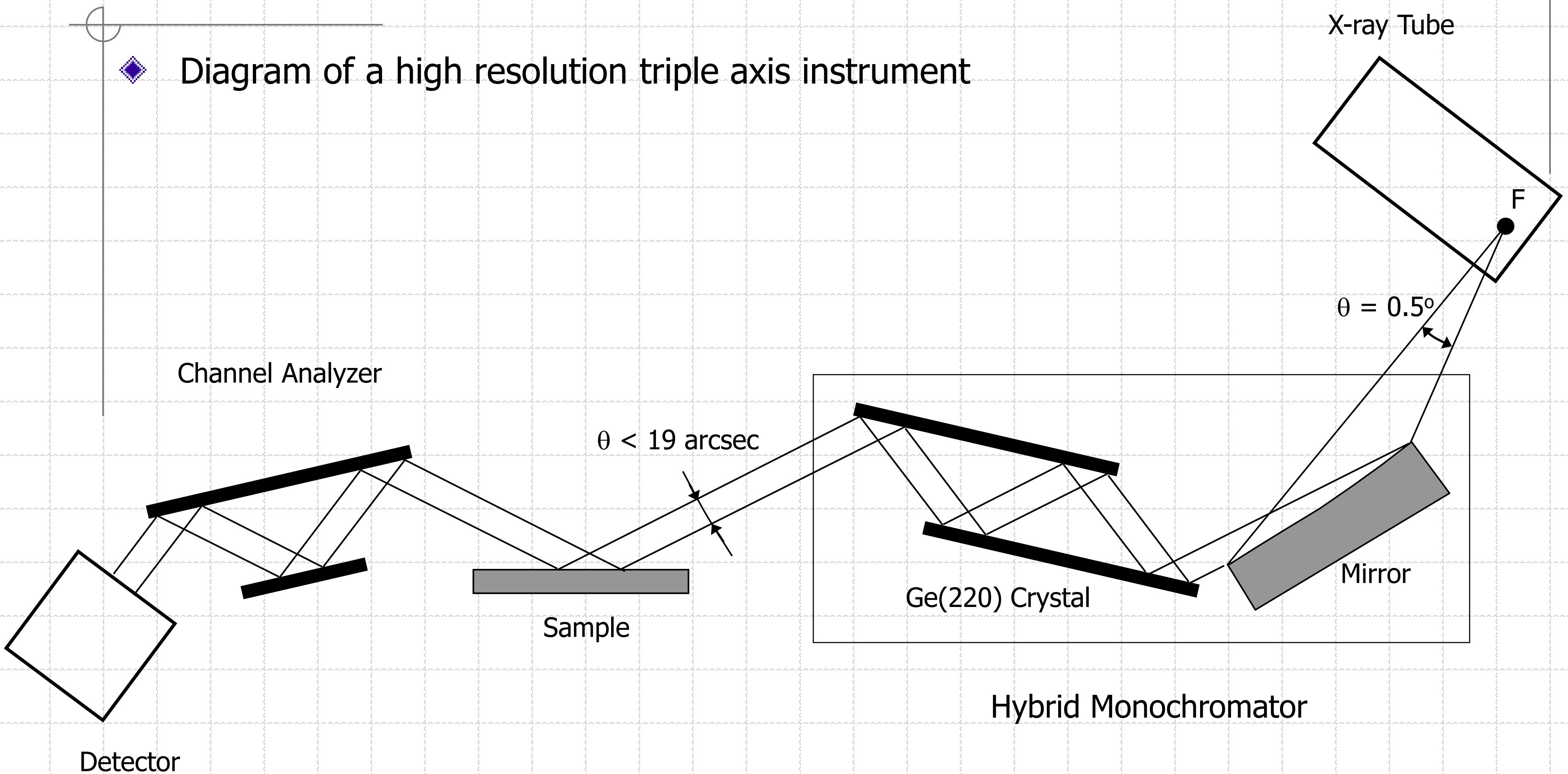
Triple Axis Geometry

- Schematic of high resolution triple-axis instrument

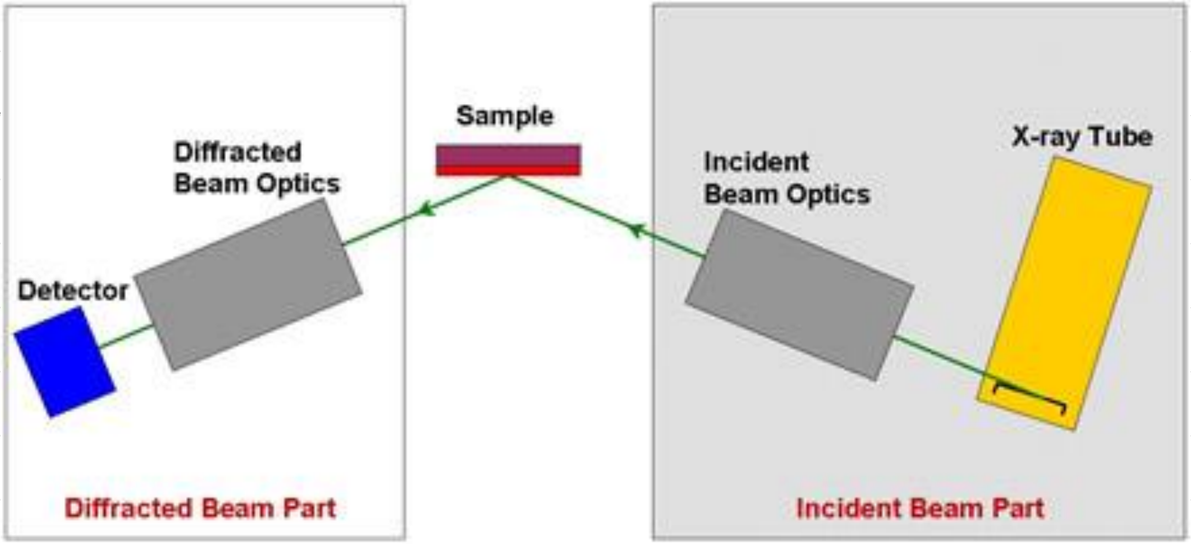


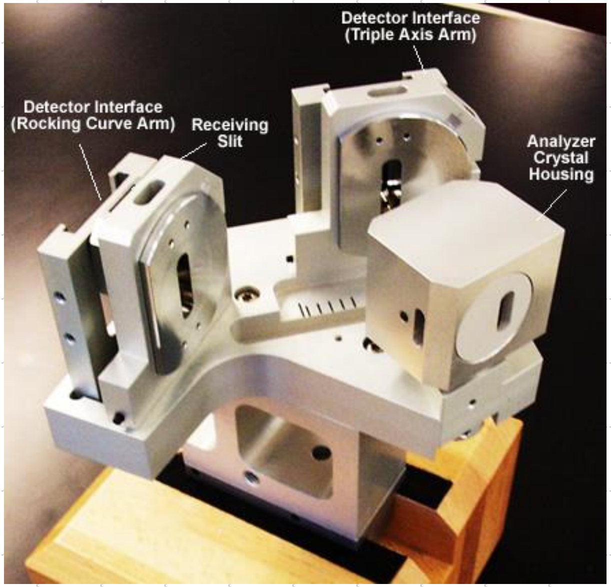
Triple Axis Geometry

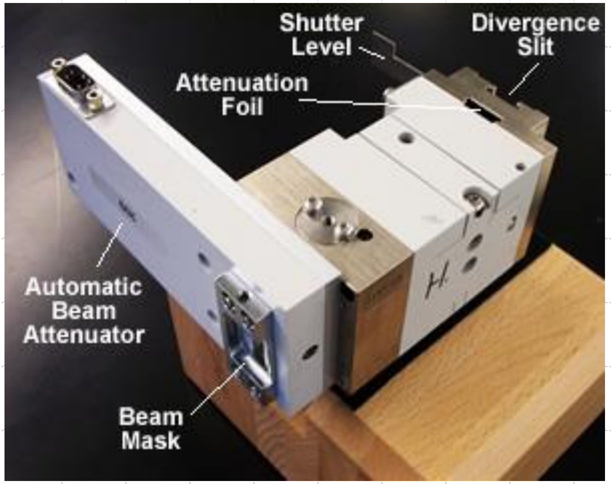
- Diagram of a high resolution triple axis instrument



High-resolution optics

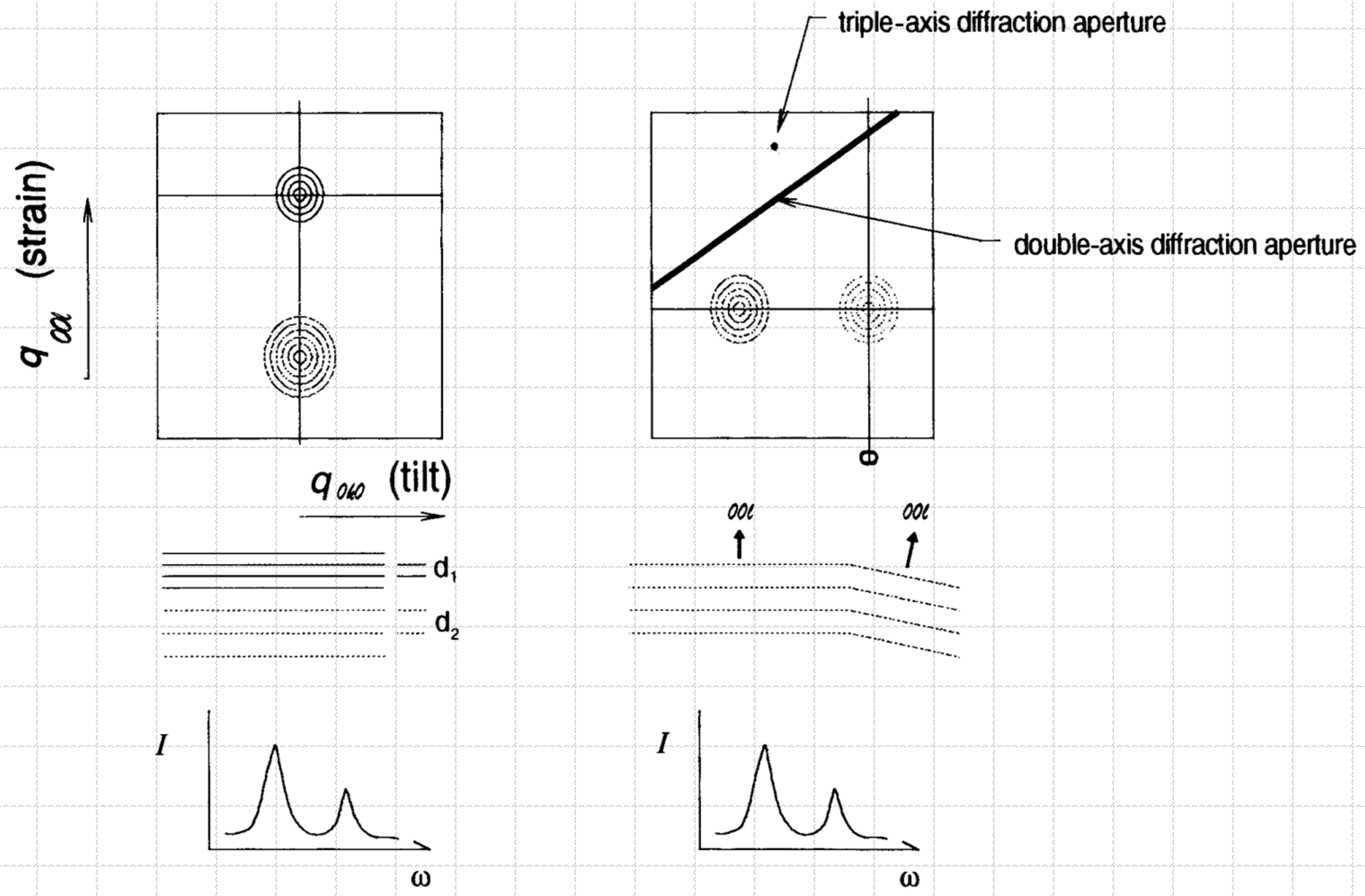


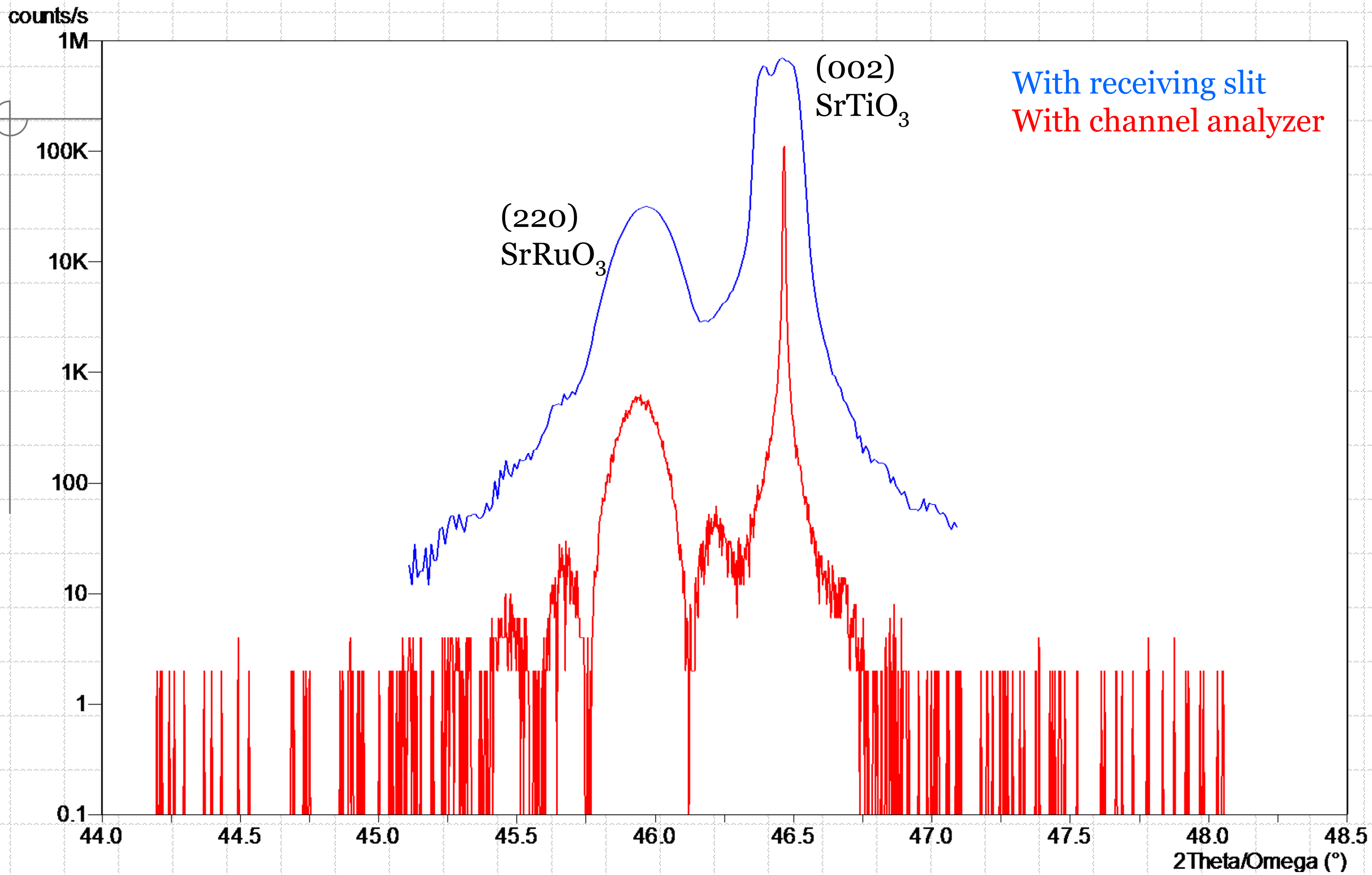




Separation of Lattice Tilts and Strains

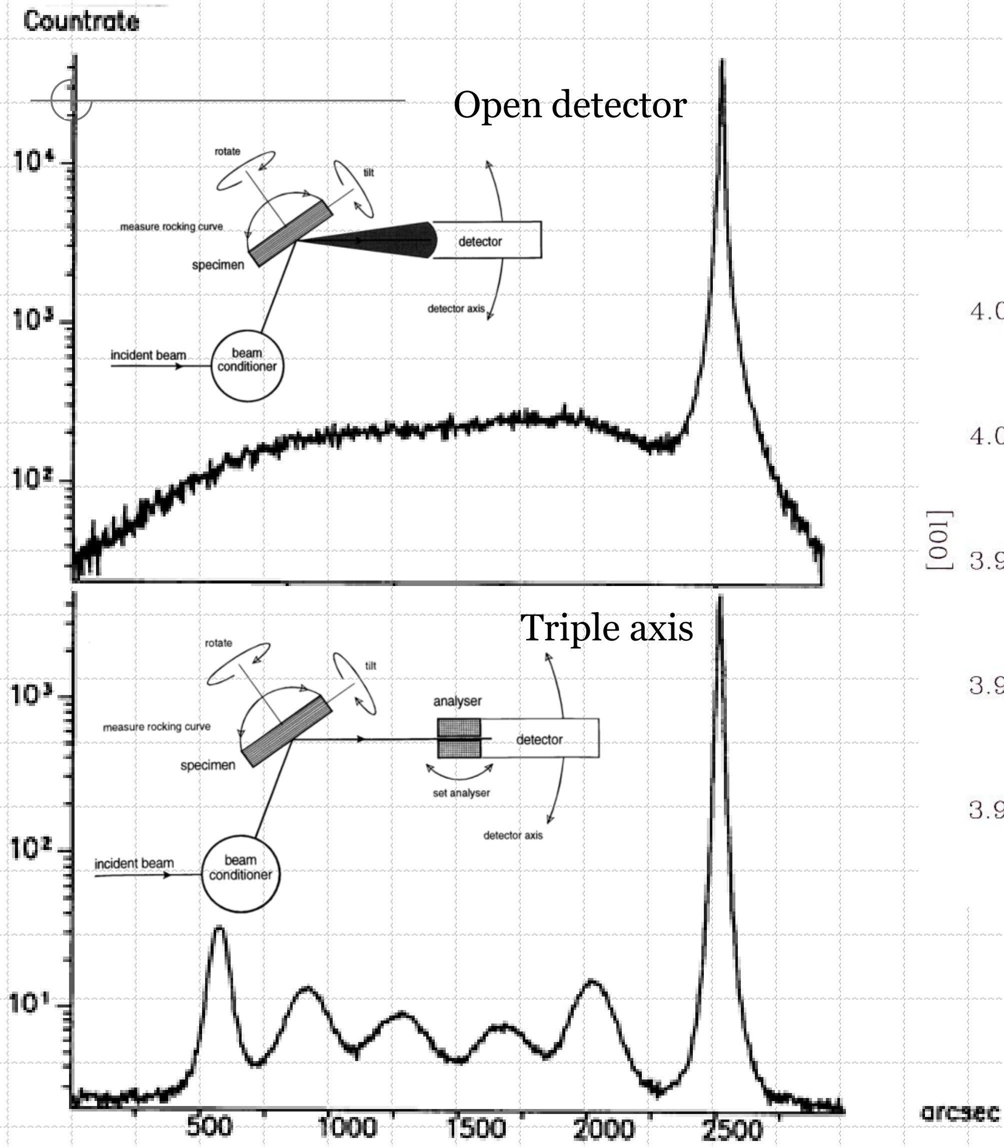
- Triple axis measurements: real and reciprocal representations



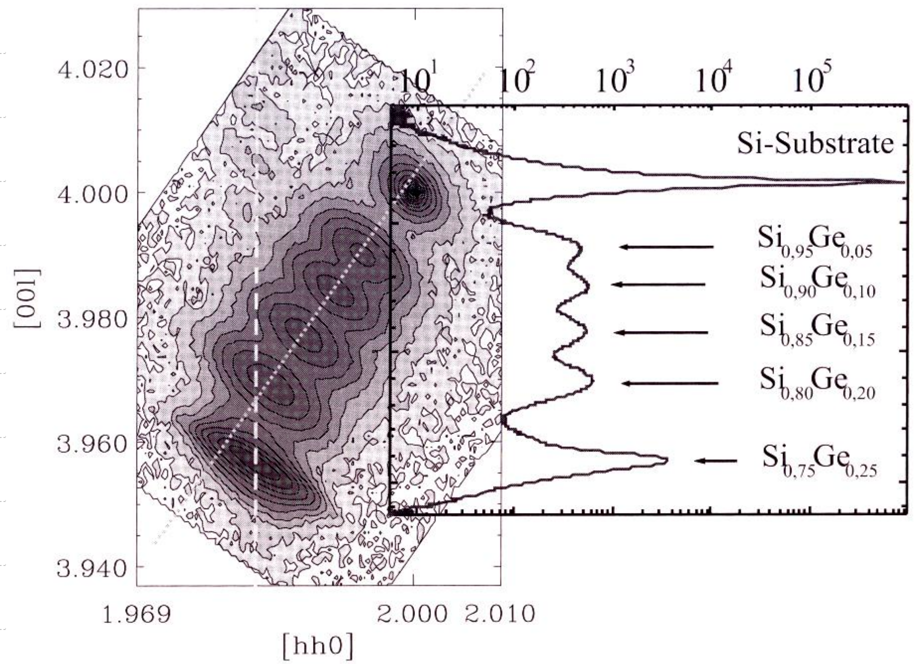


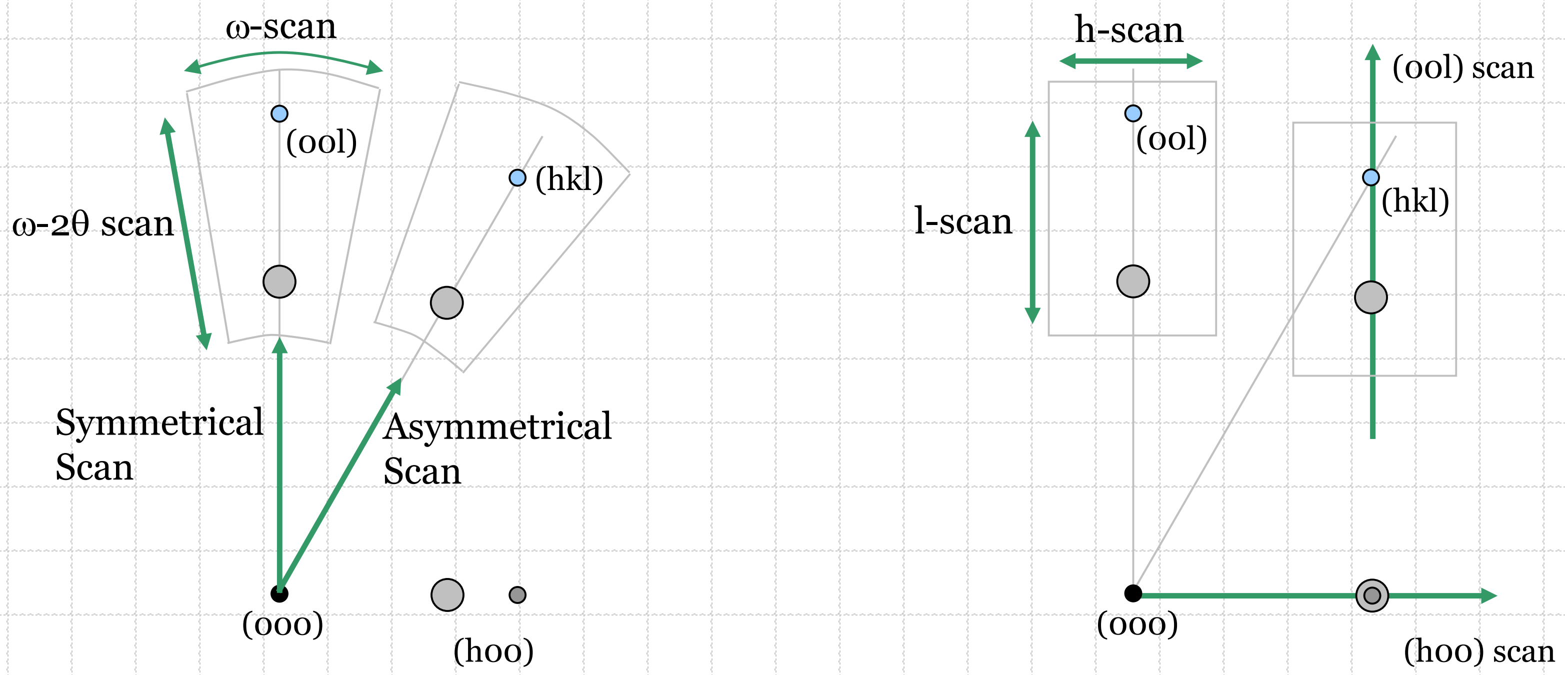
counts/s
1M
100K
10K
1K
100
10
1
0.1
44.0
44.5
45.0
45.5
46.0
46.5
47.0
47.5
48.0
48.5
2Theta/Omega (°)
(220)
$SrRuO_3$
(002)
$SrTiO_3$
With receiving slit
With channel analyzer

Triple-Axis Diffractometry

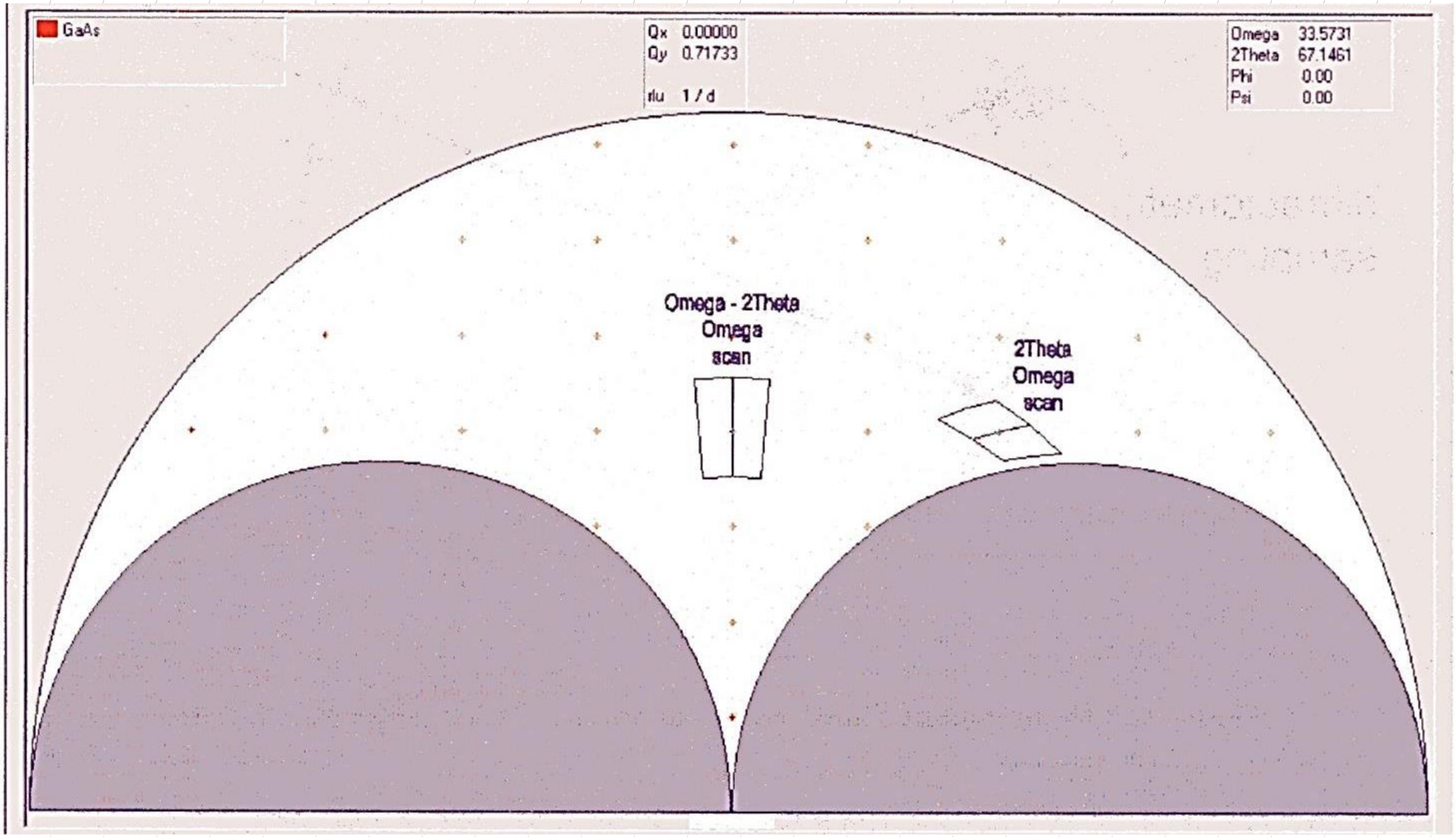


Ge content: 25% 20% 15% 10% 5%

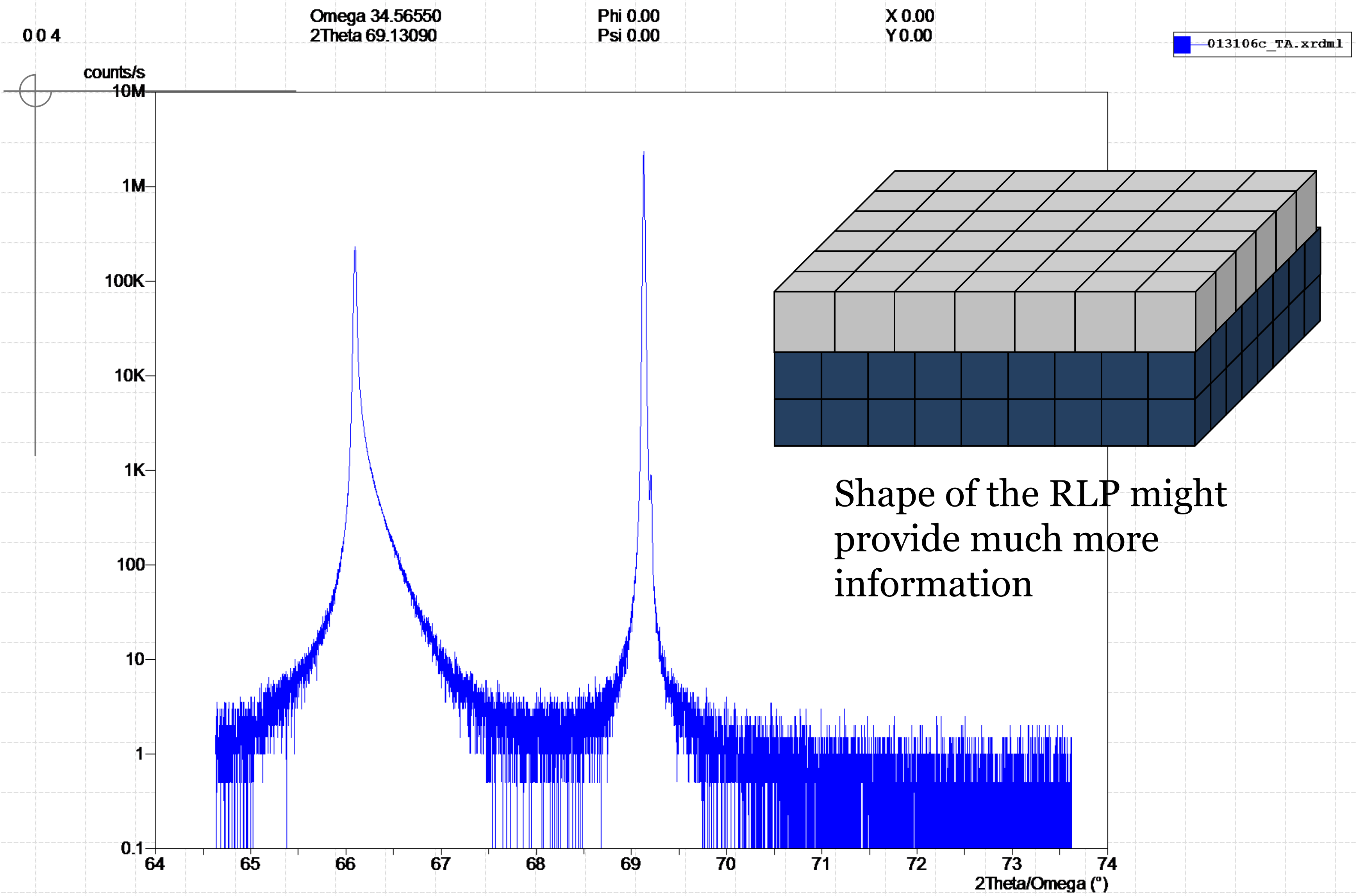



ω-scan
(ool)
(hkl)
ω-2θ scan
Symmetrical Scan
Asymmetrical Scan
(ooo)
(hoo)
h-scan
(ool) scan
(ool)
(hkl)
l-scan
(ooo)
(hoo) scan

Triple-Axis Diffractometry



Relaxed SiGe on Si(001)



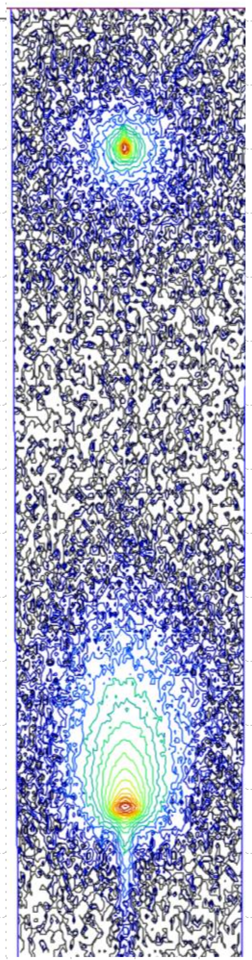
Shape of the RLP might provide much more information

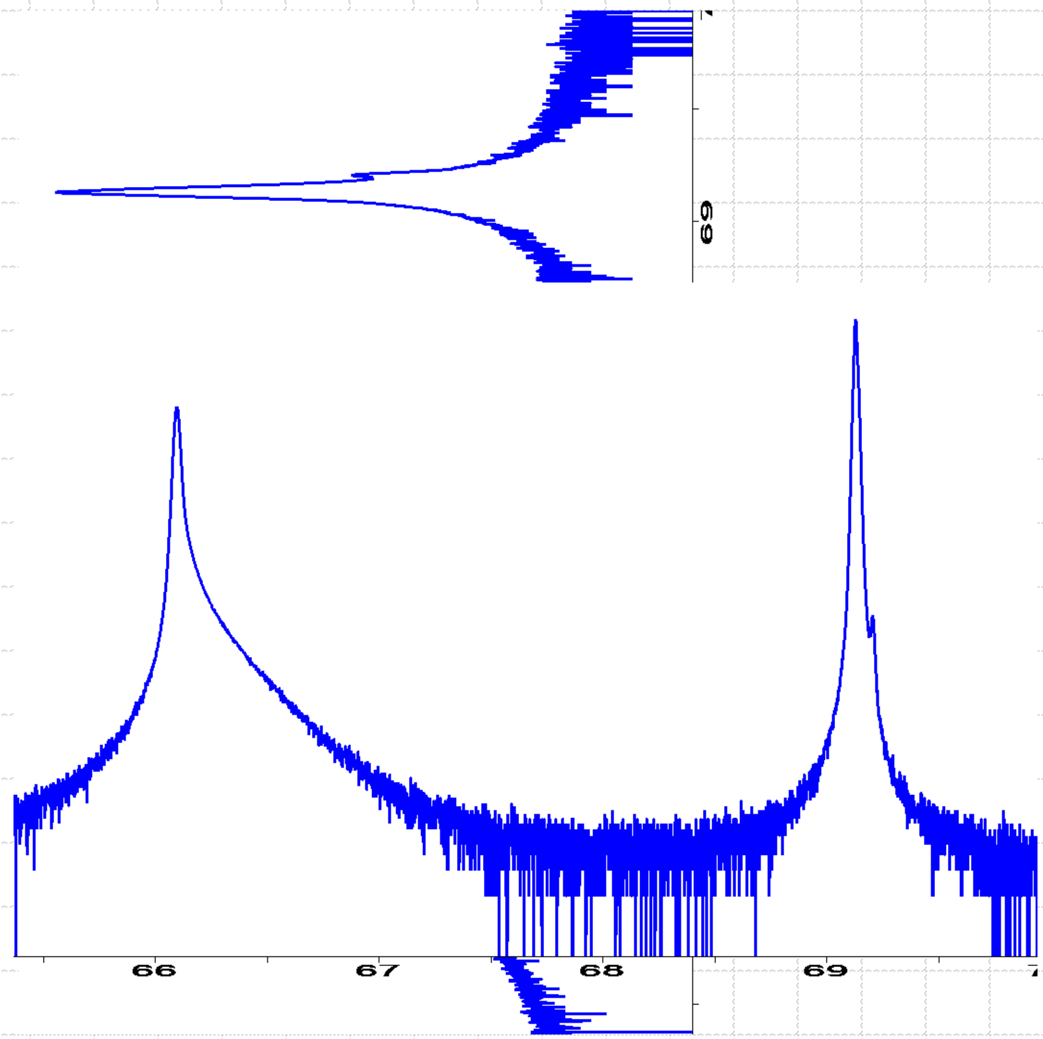
Relaxed SiGe on Si(001)

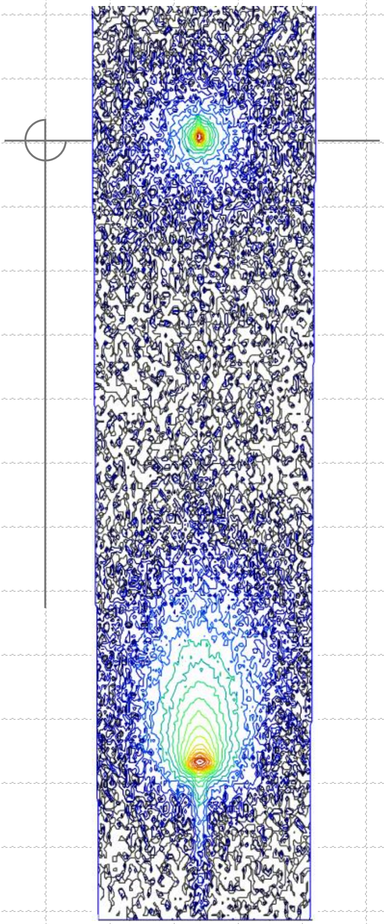
(004) RLM

Si(004)

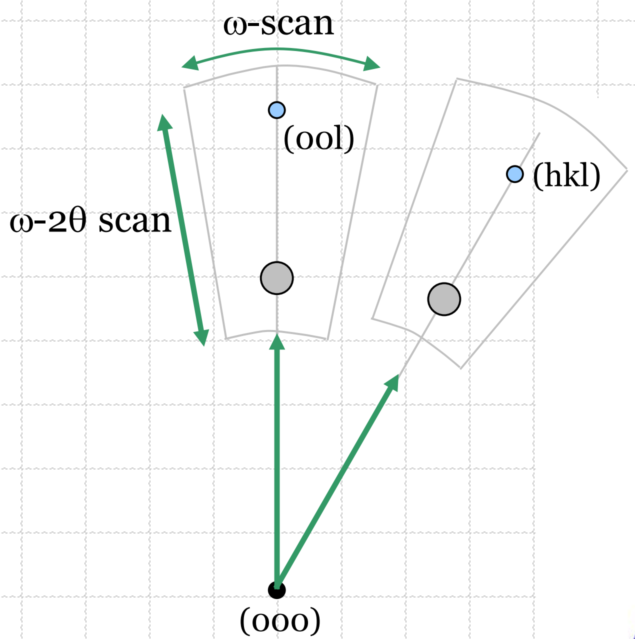
SiGe(004)

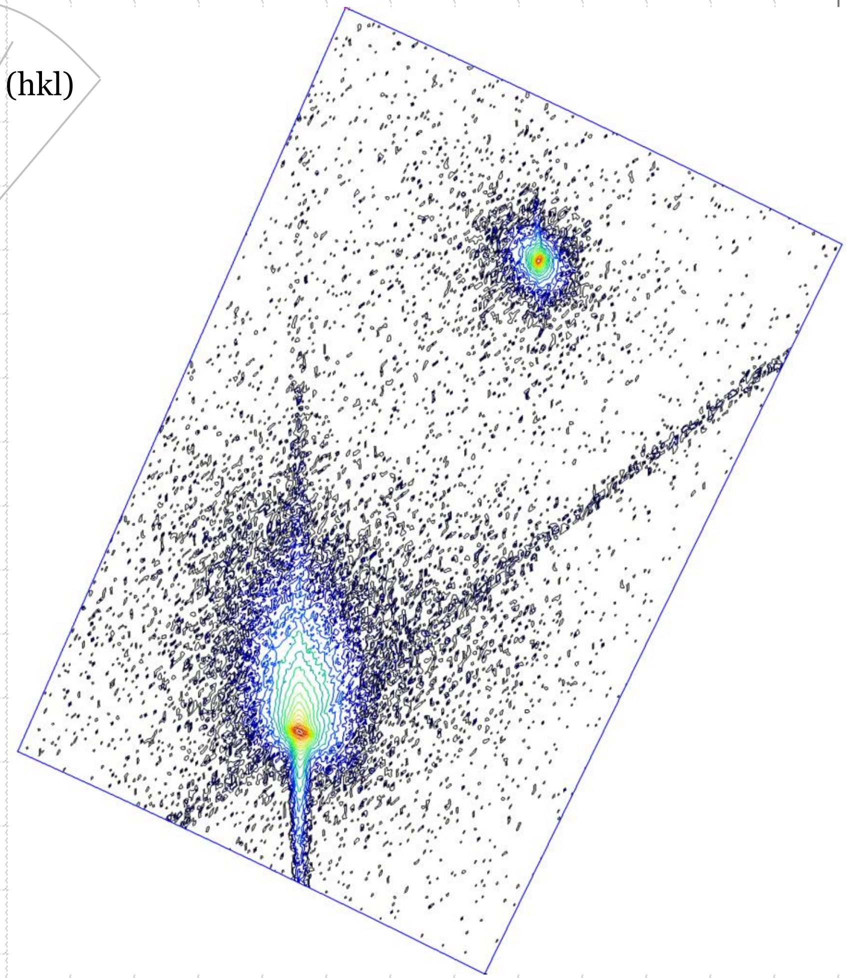






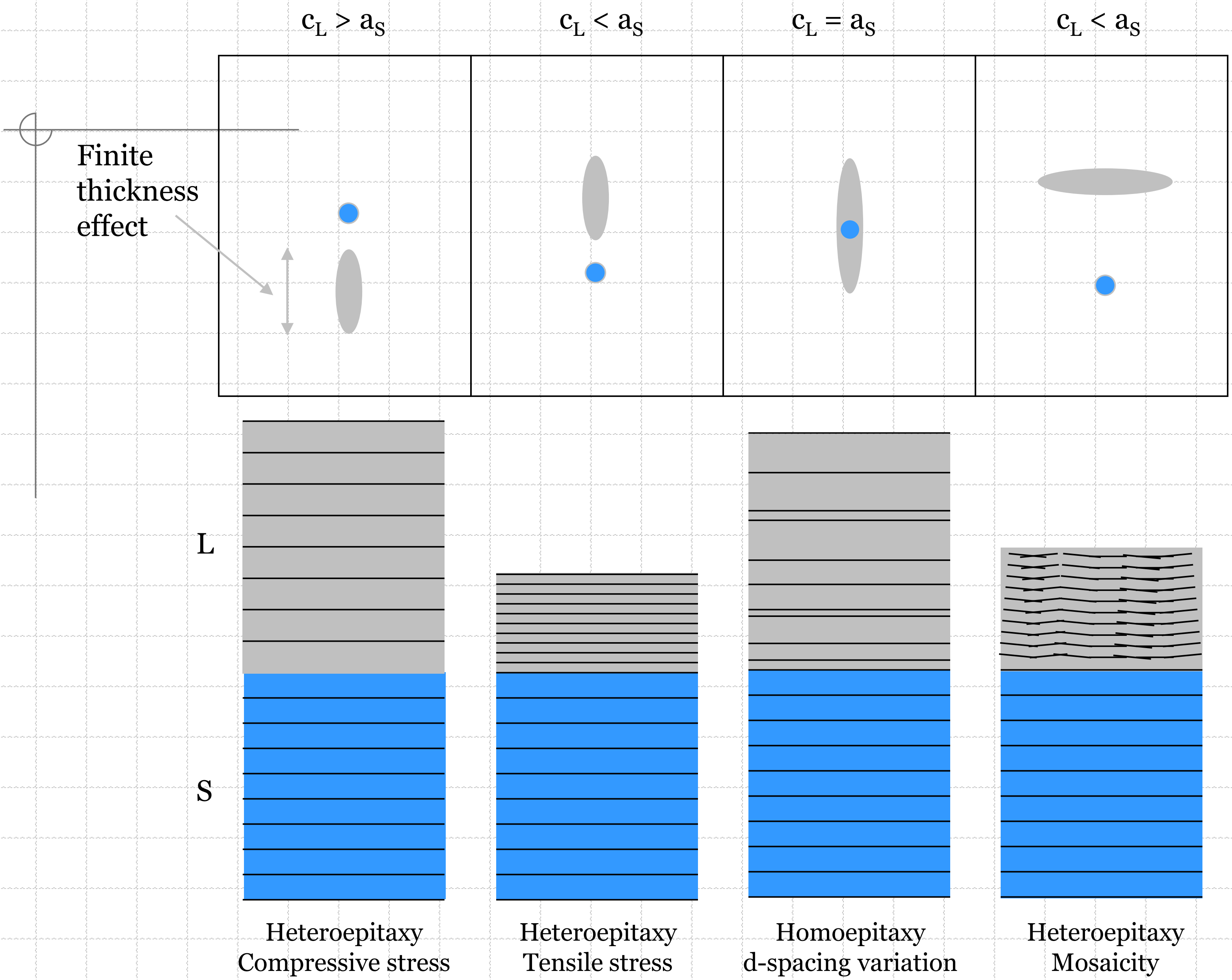
(004)

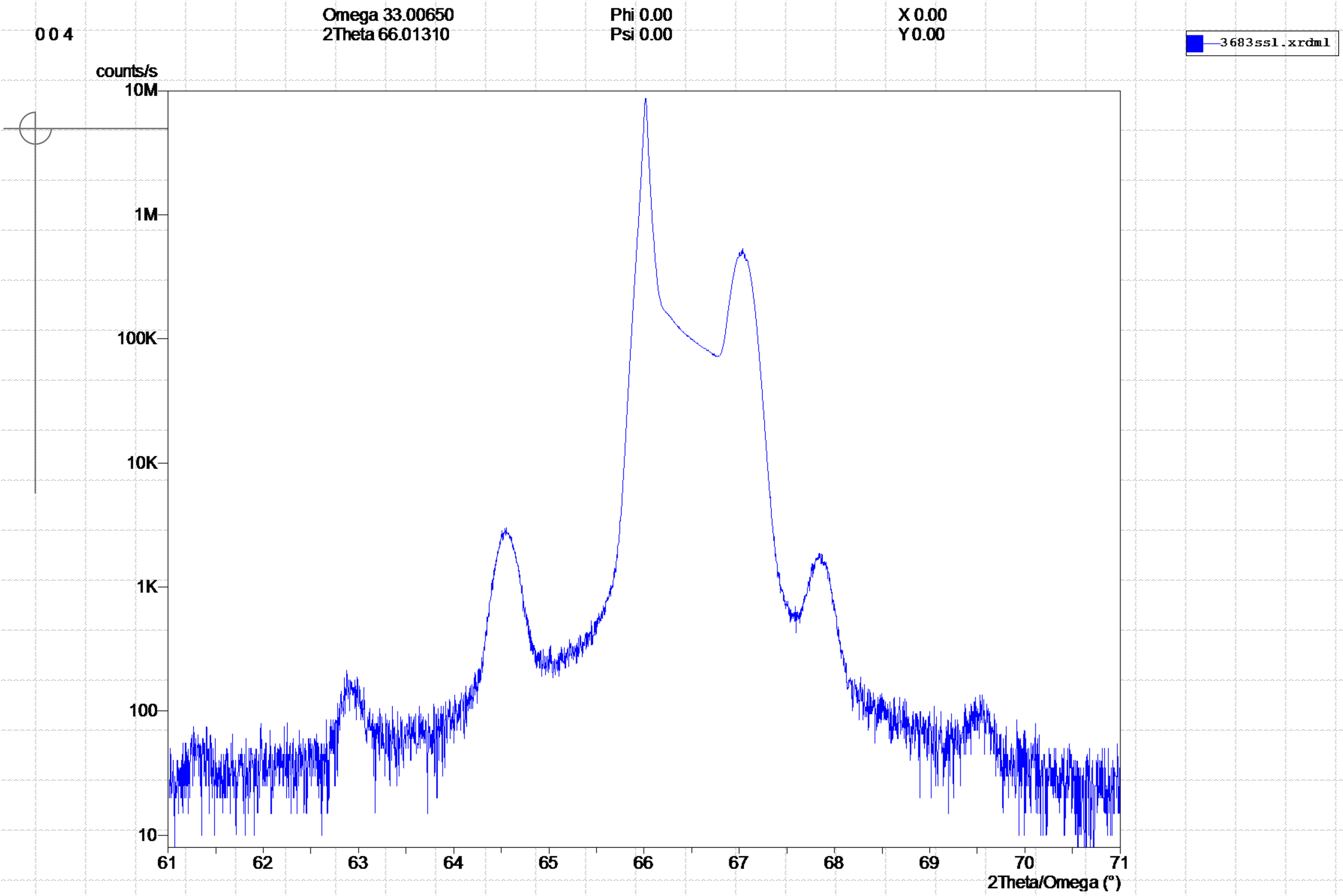




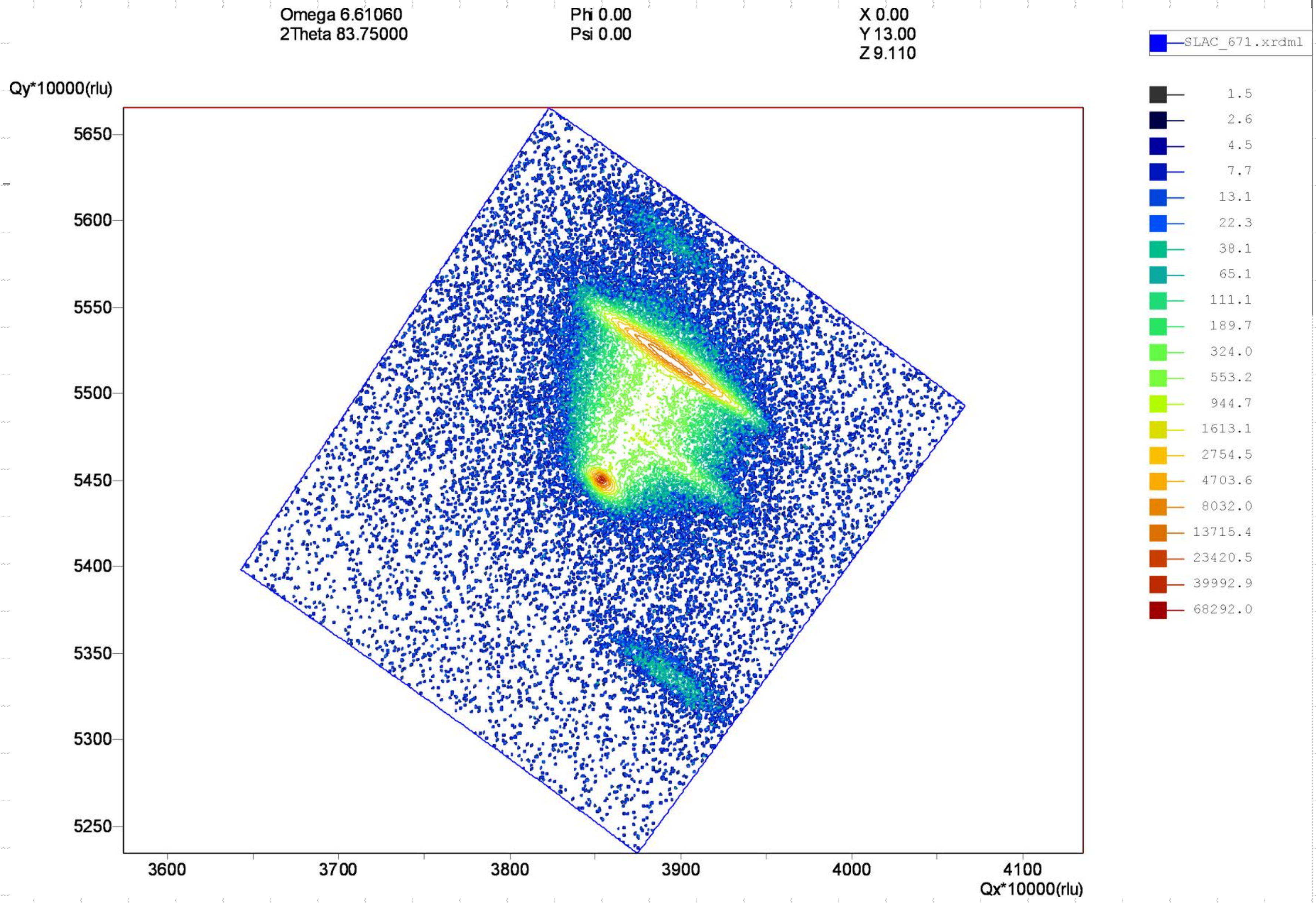
(113)

Real RLP shapes





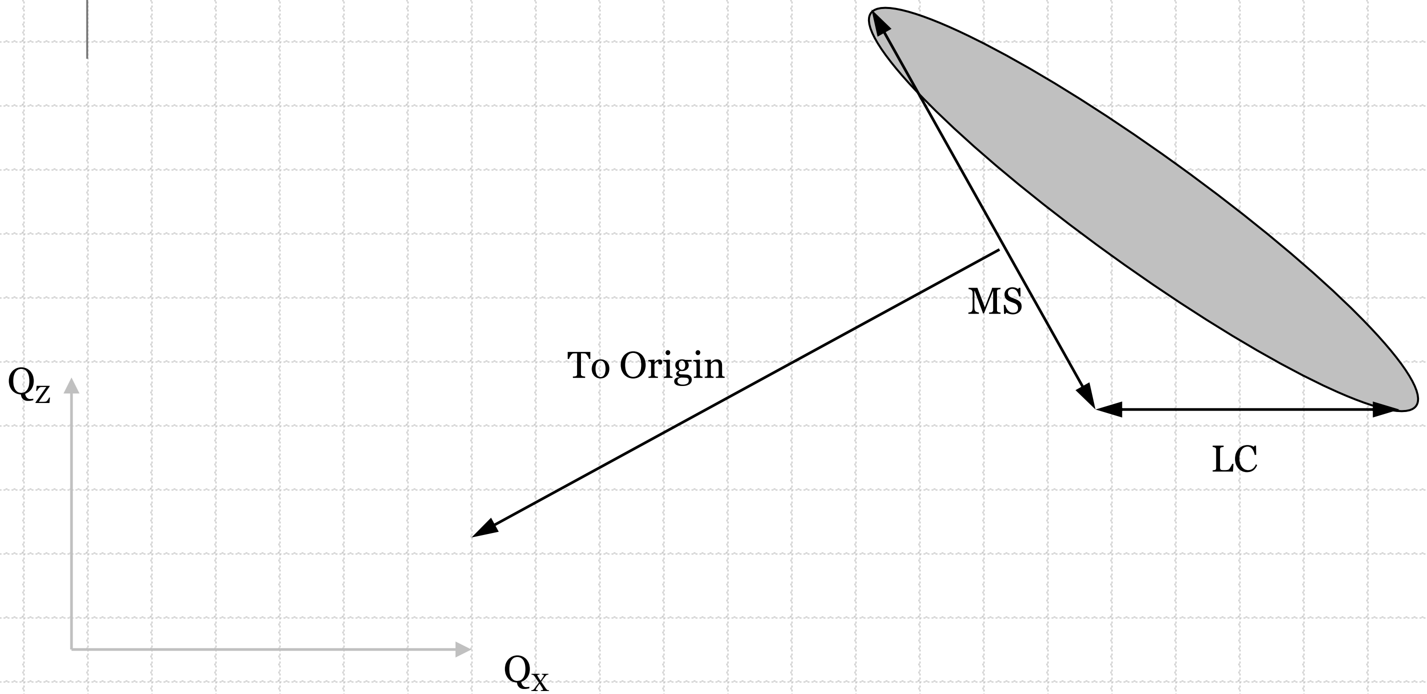
0 0 4
Omega 33.00650
2Theta 66.01310
Phi 0.00
Psi 0.00
X 0.00
Y 0.00
3683ssl.xrdml
counts/s
10M
1M
100K
10K
1K
100
10
61
62
63
64
65
66
67
68
69
70
71
2Theta/Omega (°)


Omega 6.61060
2Theta 83.75000
Phi 0.00
Psi 0.00
X 0.00
Y 13.00
Z 9.110
SLAC_671.xrdml
Qy*10000(rlu)
5650
5600
5550
5500
5450
5400
5350
5300
5250
3600
3700
3800
3900
4000
4100
Qx*10000(rlu)
1.5
2.6
4.5
7.7
13.1
22.3
38.1
65.1
111.1
189.7
324.0
553.2
944.7
1613.1
2754.5
4703.6
8032.0
13715.4
23420.5
39992.9
68292.0

Mosaic Spread and Lateral Correlation Length

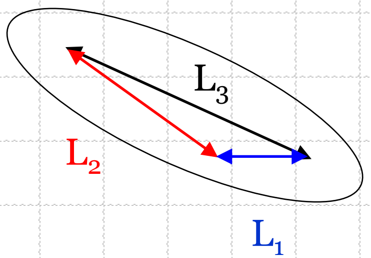
The mosaic spread of the layer is calculated from the angle that the layer peak subtends at the origin of reciprocal space measured perpendicular to the reflecting plane normal.

The lateral correlation length of the layer is calculated from the reciprocal of the FWHM of the peak measured parallel to the interface.



Mosaic Spread and Lateral Correlation Length

The Mosaic Spread and Lateral Correlation Length functionality derives information from the shape of a layer peak in a diffraction space map recorded using an asymmetrical reflection



$$L_3 = \sqrt{\Delta q_x^2 + \Delta q_z^2}$$

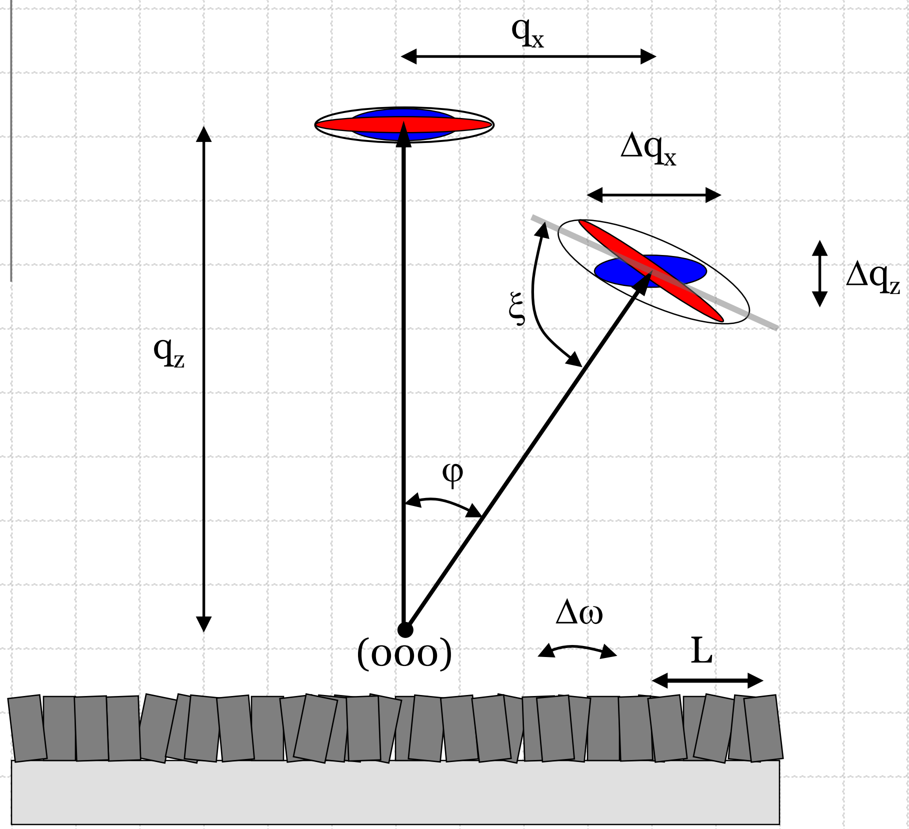
and

$$\varphi = \frac{1}{\tan\left(\frac{q_x}{q_z}\right)}$$

$$\xi = \frac{1}{\tan\left(\frac{\Delta q_x}{\Delta q_z}\right)}$$

$$\frac{L_1}{L_2} = -\frac{\cos\xi}{\cos(\varphi + \xi)}$$

$$\frac{L_3}{L_2} = -\frac{\sin\varphi}{\cos\xi}$$

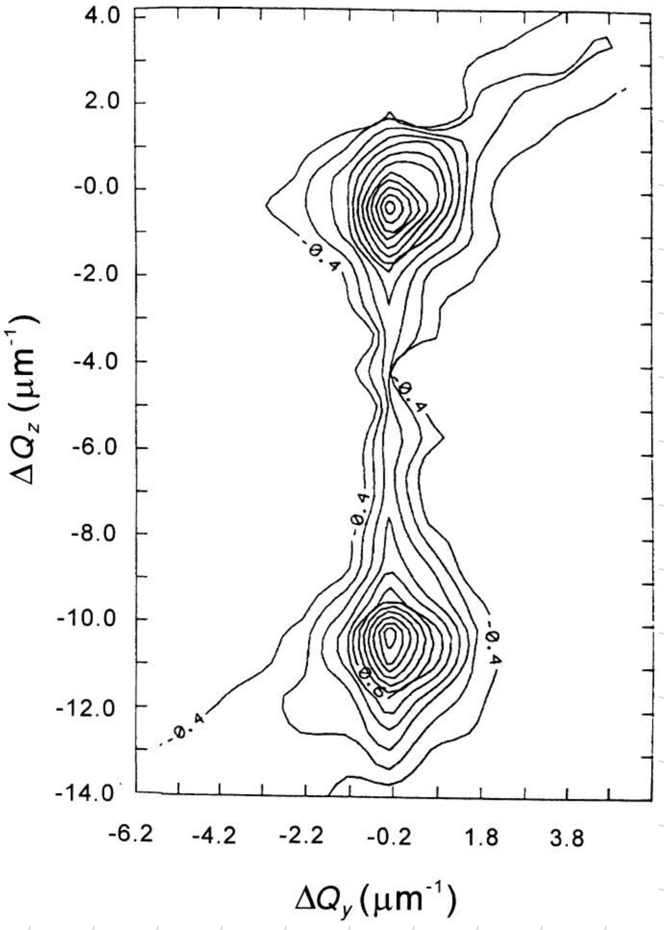


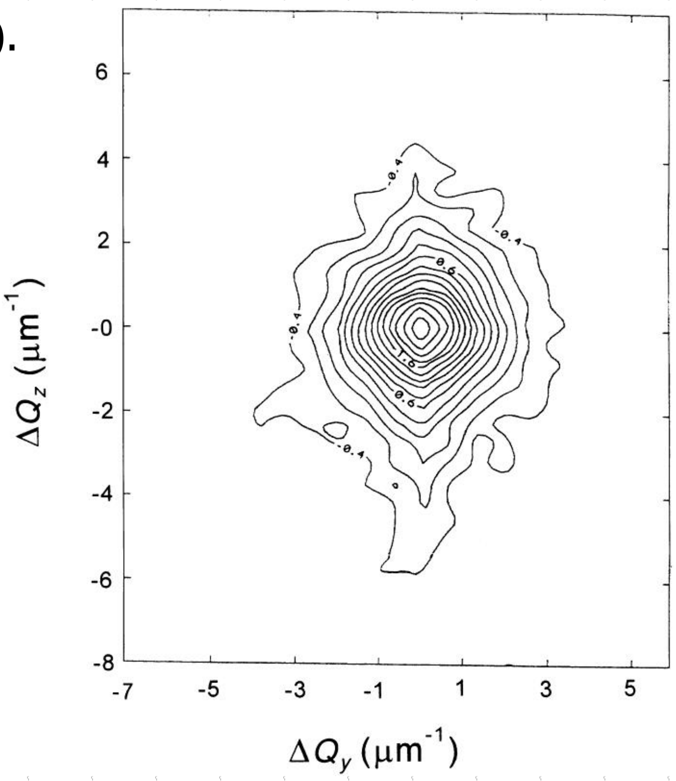
$$\text{Lateral correlation length} = \frac{1}{L_1}$$

$$\text{Microscopic tilt} = \frac{L_2}{\sqrt{q_x^2 + q_z^2}}$$

Applications of RLM

Thin epitaxial GaAs film on GaAs(001).
Film has more As.





RLM after annealing

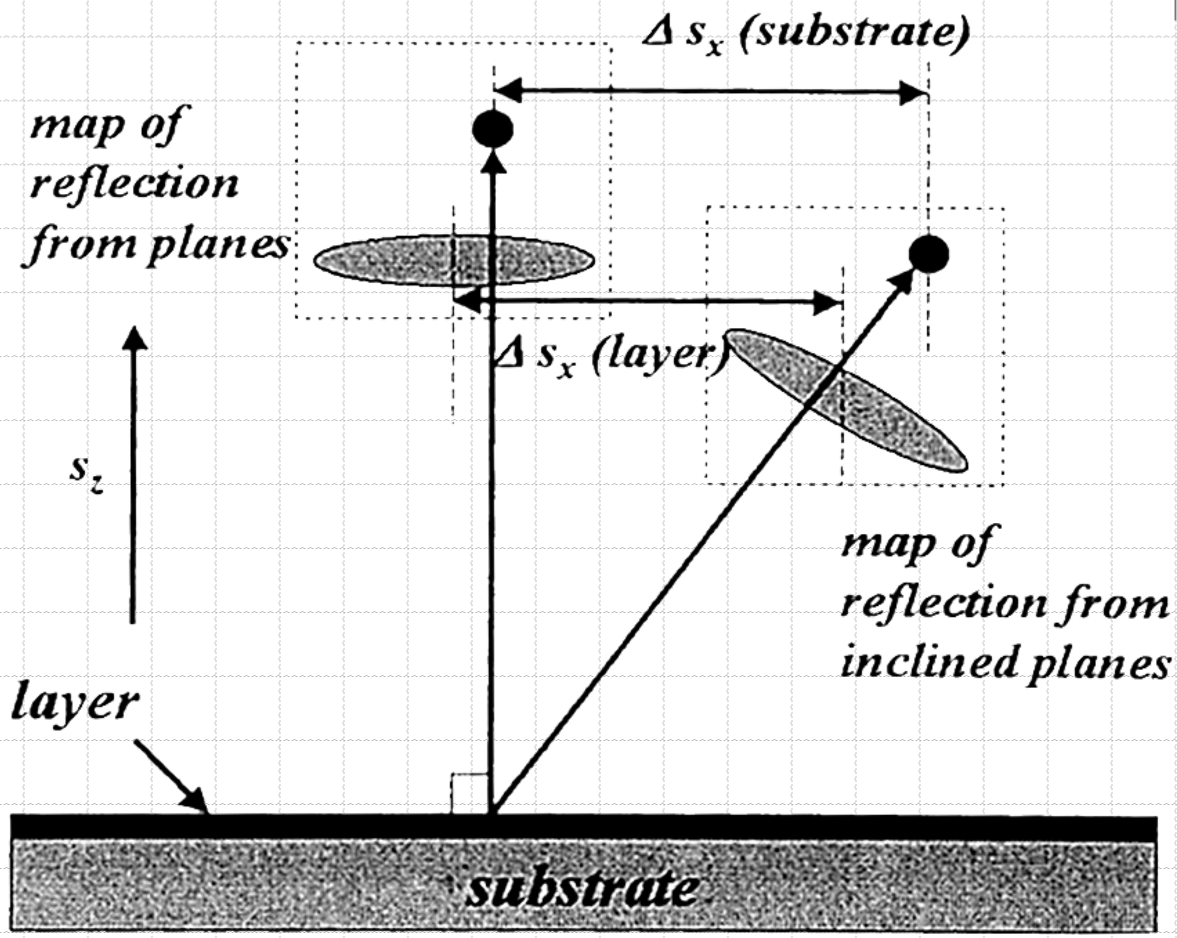
Strain in Partially Relaxed Layers

- A schematic of the measurements to be extracted from two RLM's to determine the relaxation of a layer on a substrate.

$$s_x = \frac{q_x}{2\pi} = \frac{1}{\lambda}\{\cos\omega - \cos(2\theta - \omega)\}$$

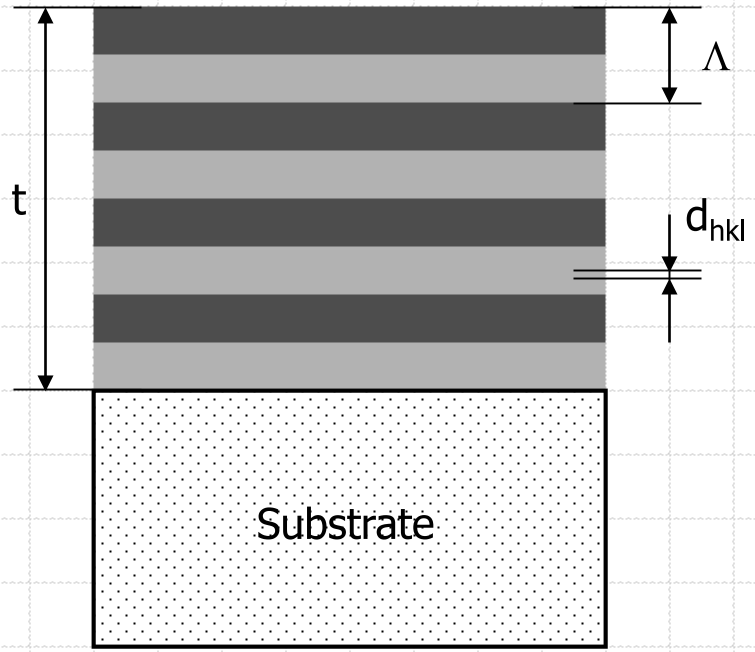
$$s_z = \frac{q_z}{2\pi} = \frac{1}{\lambda}\{\sin\omega - \sin(2\theta - \omega)\}$$

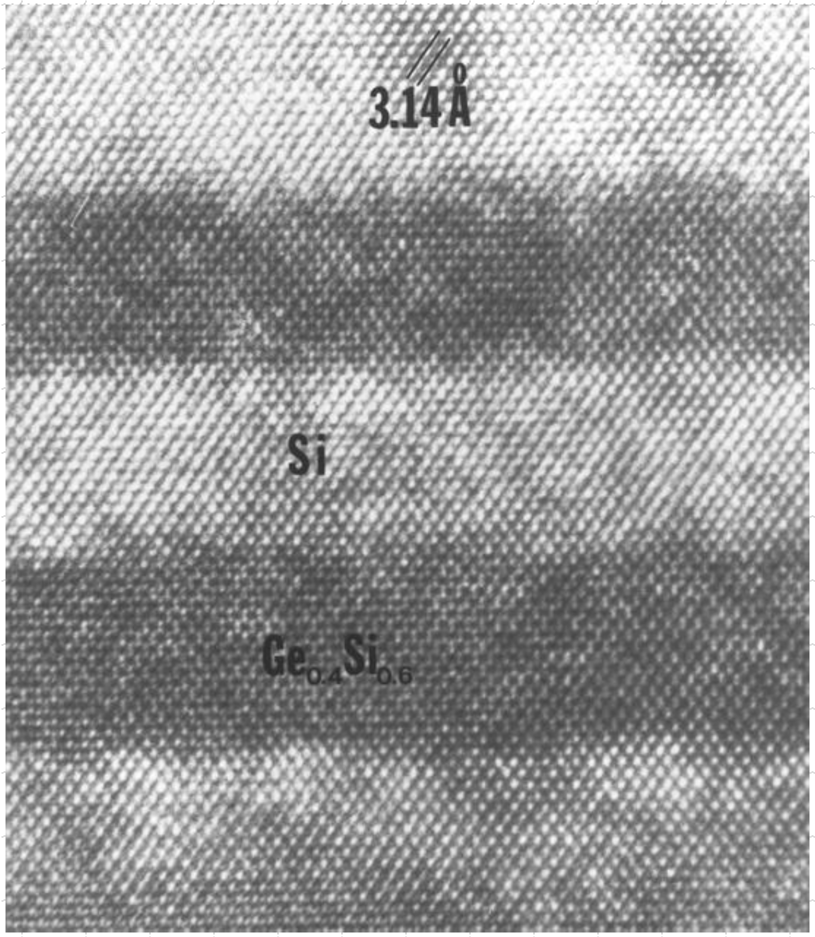
$$q = \frac{4\pi \sin\theta}{\lambda}$$



Superlattices and Multilayers

- Transmission electron micrograph depicts one of the earliest multilayer, GeSi / Si "superlattices." The dots are the actual images of individual columns of atoms.



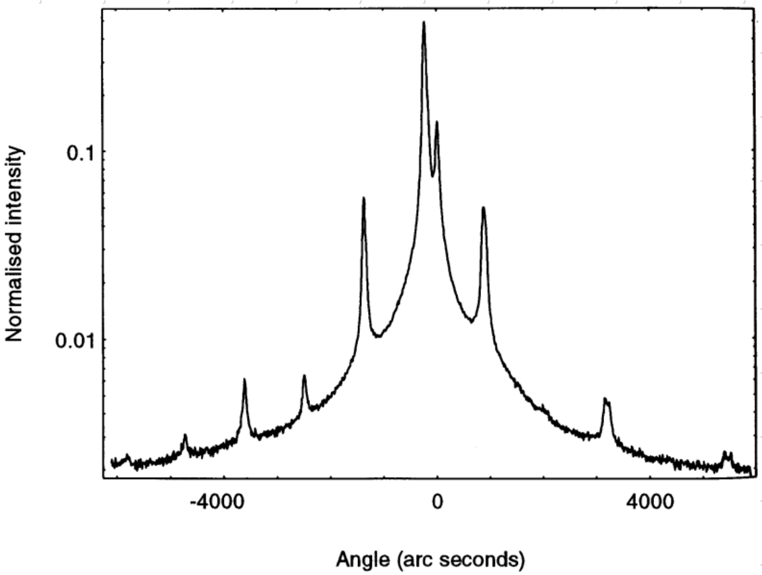


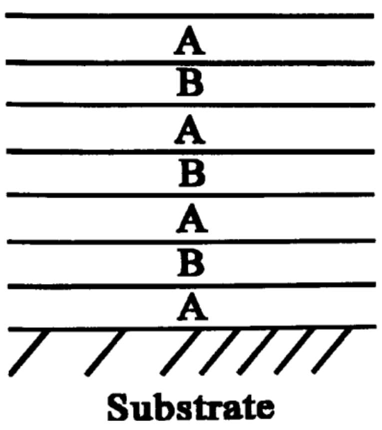
Superlattices and Multilayers

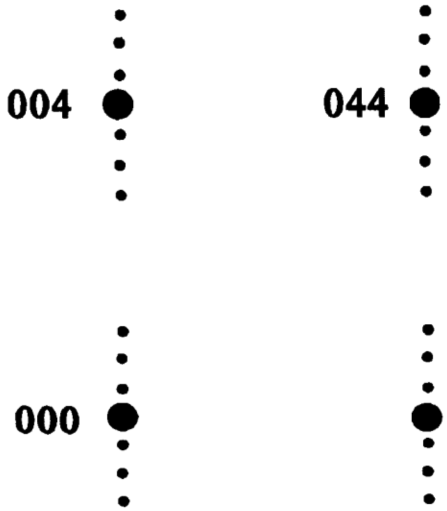
- General characteristics of large repeat superlattices
  - The spatial period of the structure
  - The thickness of the repeating unit
  - The composition of the layers
  - The dispersion in the repeating period
  - The interface roughness
  - The interface grading

Superlattices and Multilayers

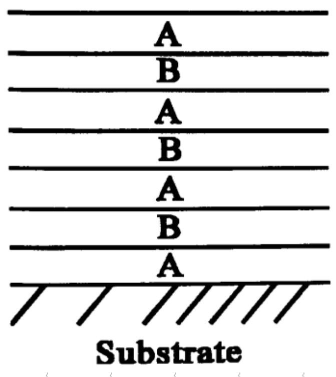
- Double-axis rocking curve from an AlGaAs/GaAs superlattice.
  - Cu Kα radiation, 004 reflection



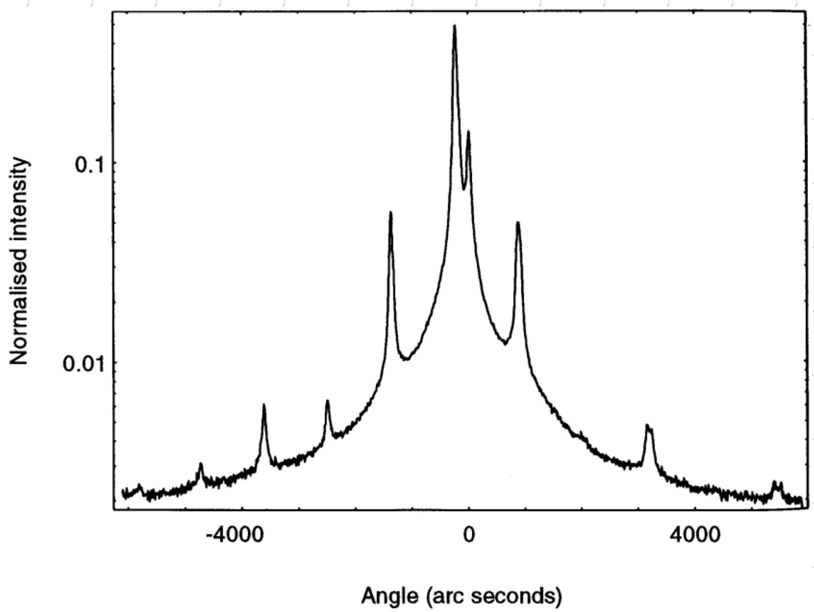




Superlattices and Multilayers



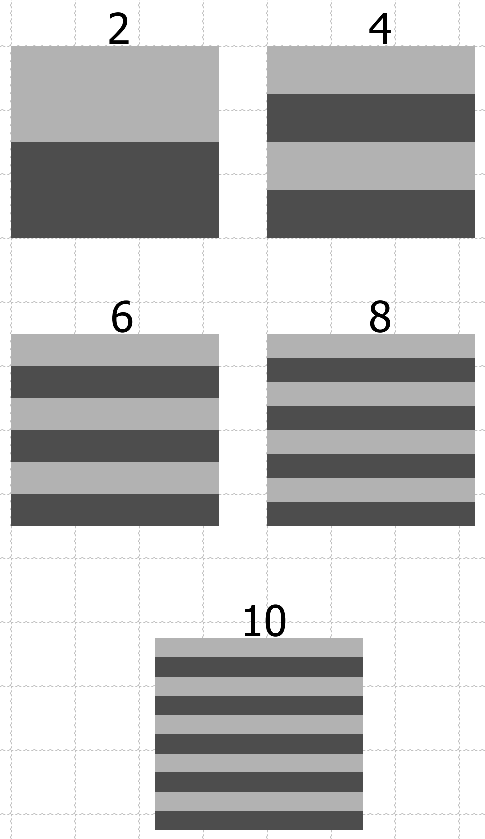
- The rocking curve shows the following features:
  - A substrate peak from the A substrate
  - A peak caused by the addition of Bragg reflections from the A and B components of the MQW. This is zero-order or average mismatch peak, from which the average composition of the A+B layers may be obtained by differentiation of Bragg's law.
  - A set of subsidiary "satellite" peaks symmetrically surrounding the zero-order peak, with spacing determined by the periodicity (total thickness of the repeating layers) of the MQW.

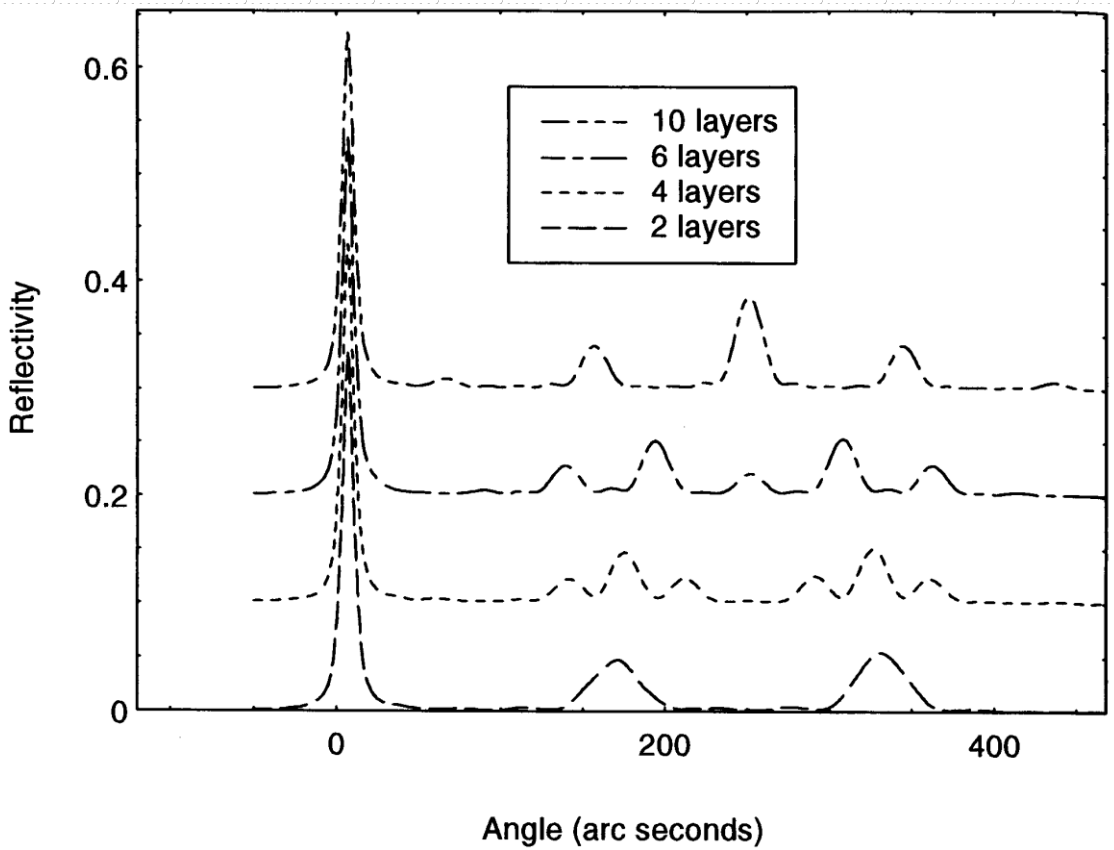


XRD pattern from multiple quantum well (MQW) with substrate A (GaAs) and stack of AB layers where B is alloy (e.g. $Ga_{1-x}Al_xAs$).

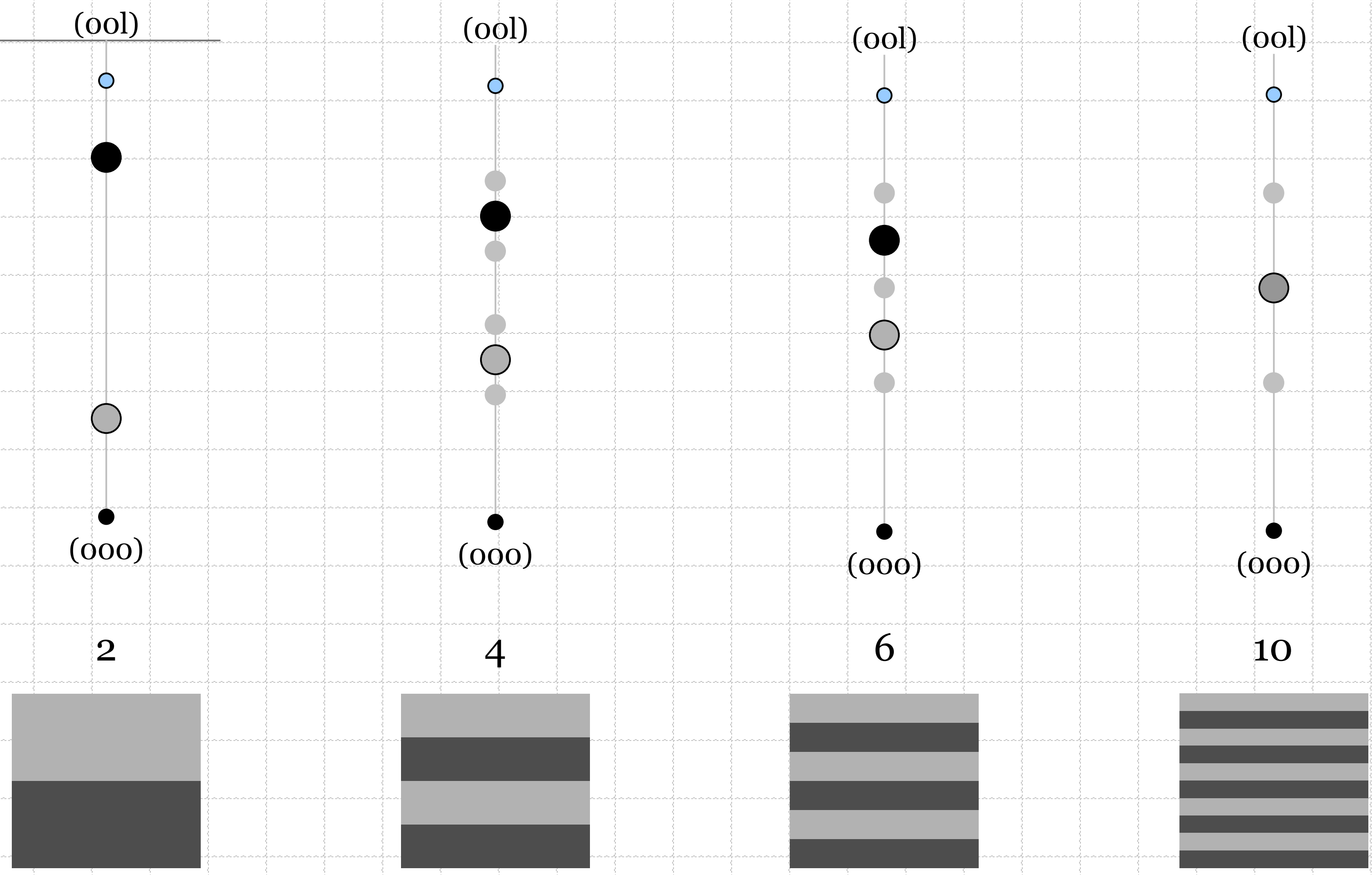
Superlattices and Multilayers

- Simulated rocking curves from an epilayer of total thickness 1μm, subdivided into 2, 4, 6 and 10 layers of alternate composition.



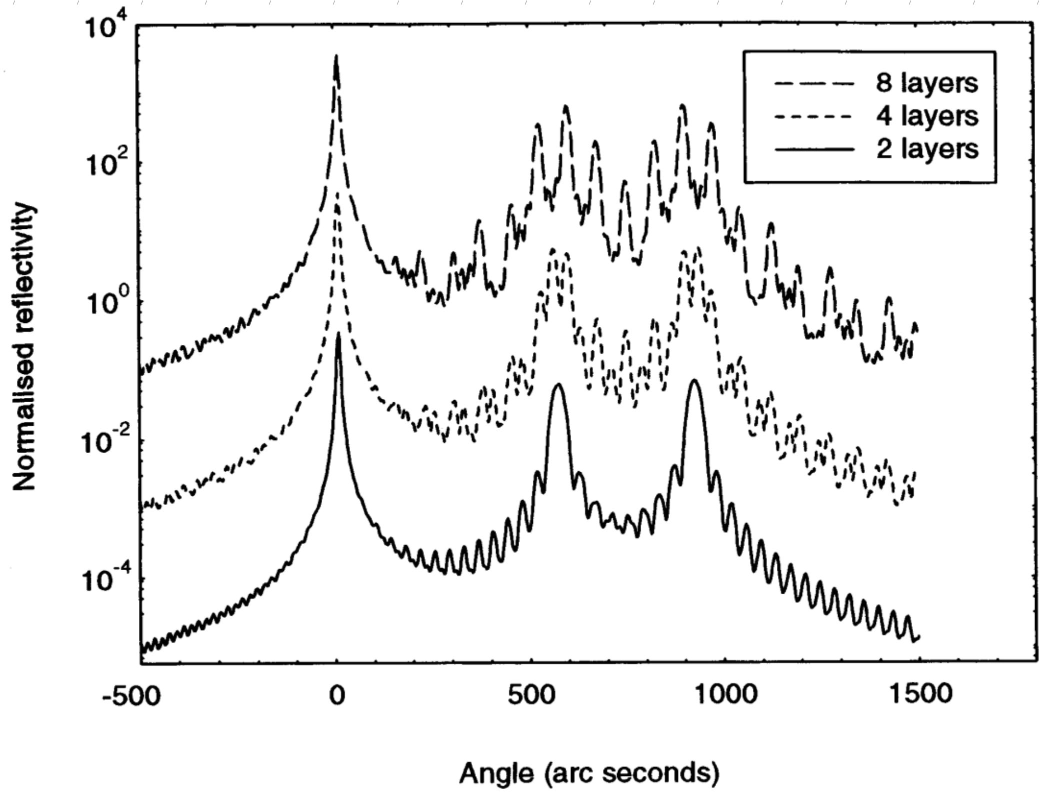


Superlattices and Multilayers



Superlattices and Multilayers

- Simulated rocking curve from sequence of layers of total thickness 1μm, divided into 2, 4 and 8 repeats of $In_xGa_{1-x}As$ layers of composition x = 0.5 and x = 0.43.

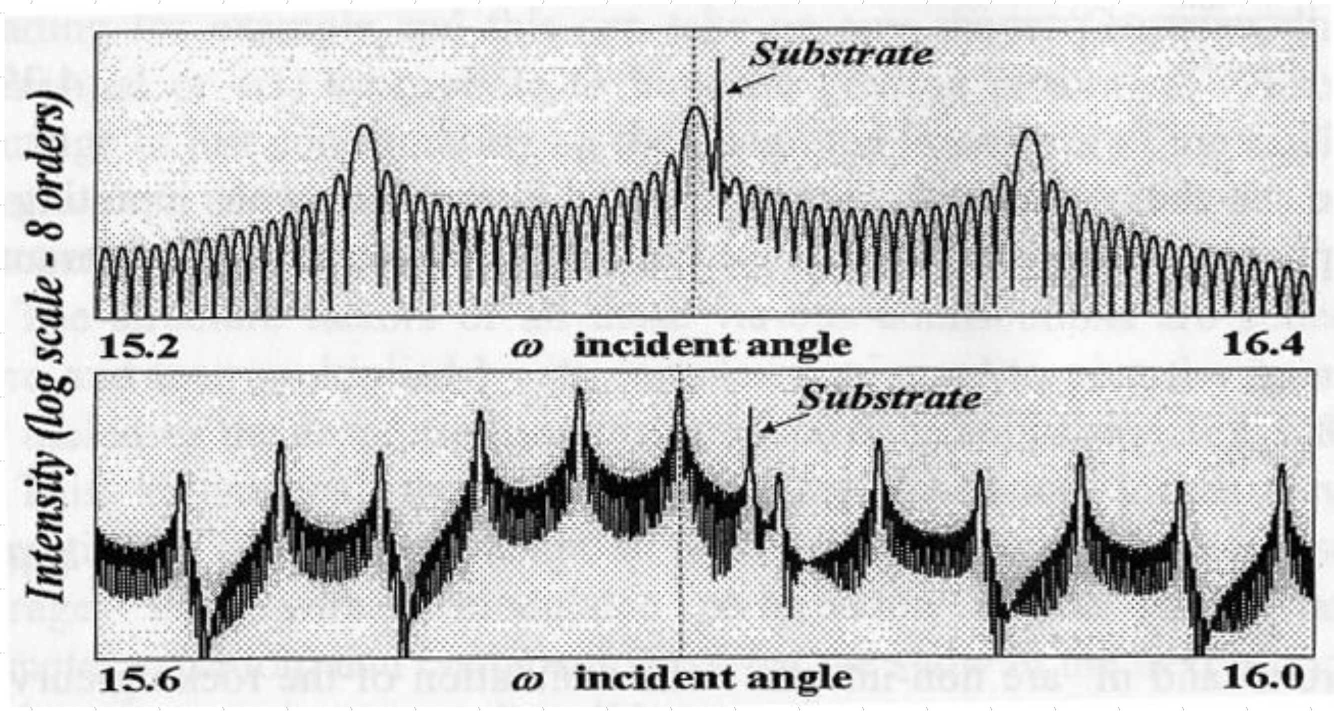


Superlattices and Multilayers

- The multilayer periodicity is defined as:

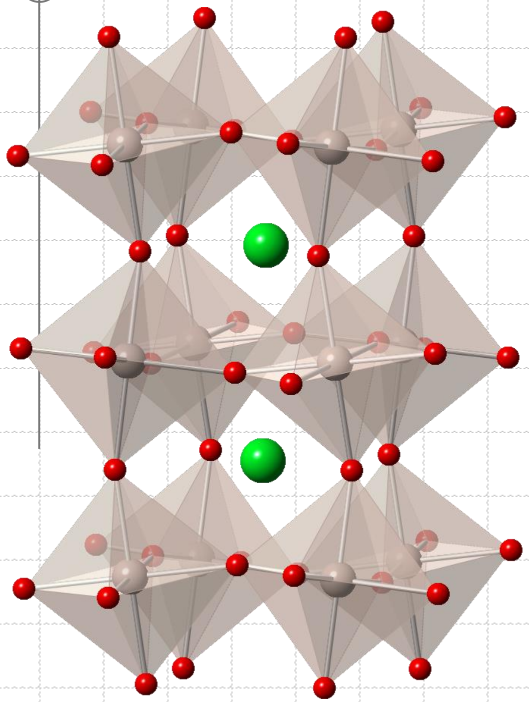
$$\Lambda = \frac{(n_i - n_j)\lambda}{2(\sin\omega_i - \sin\omega_j)}$$

equation defines satellite peak positions

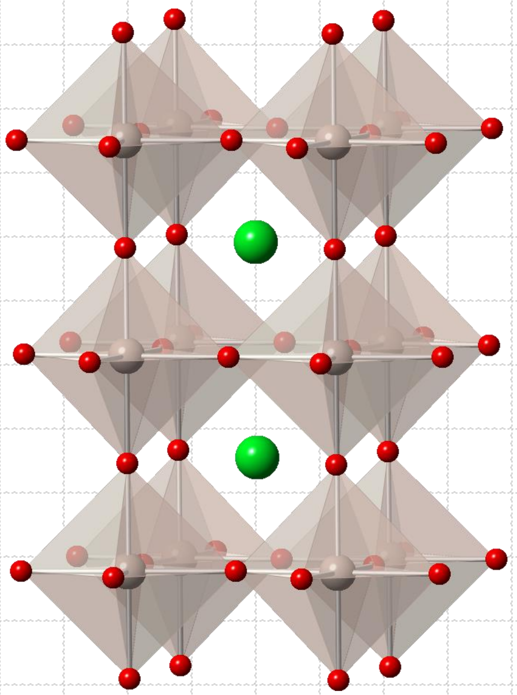


Structure of $SrRuO_3$

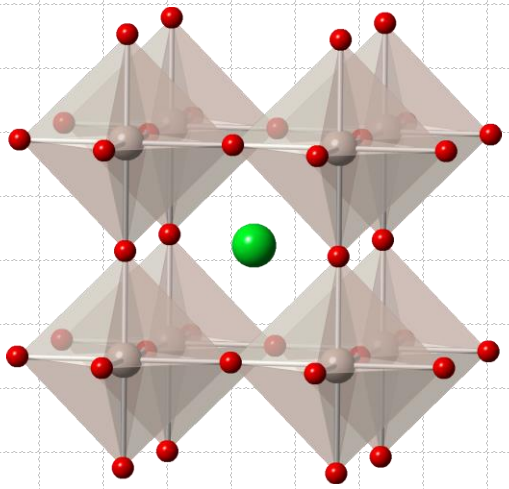
Orthorhombic



Tetragonal



Cubic



a = 5.567 Å
b = 5.530 Å
c = 7.845 Å

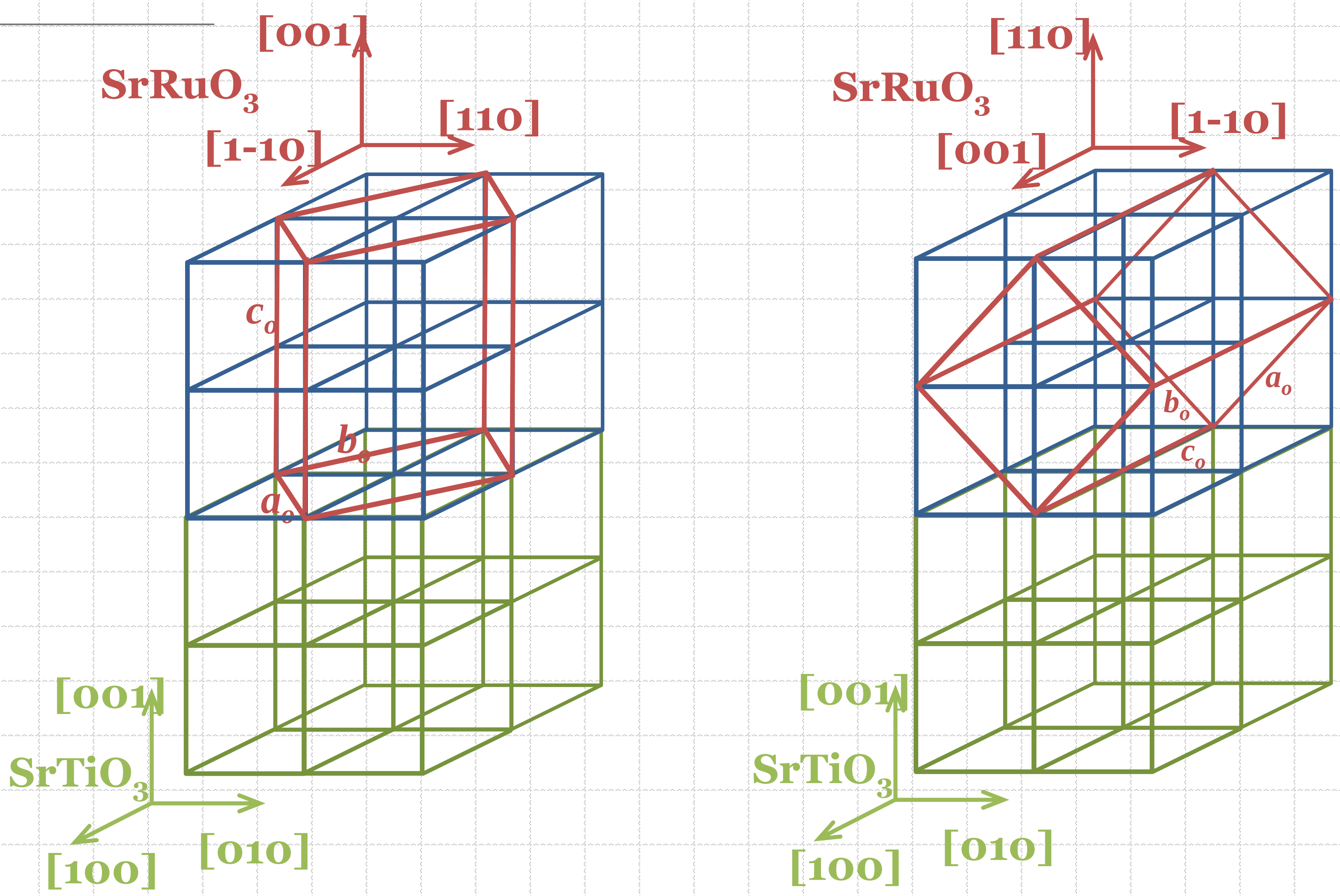
→ 275-550 °C

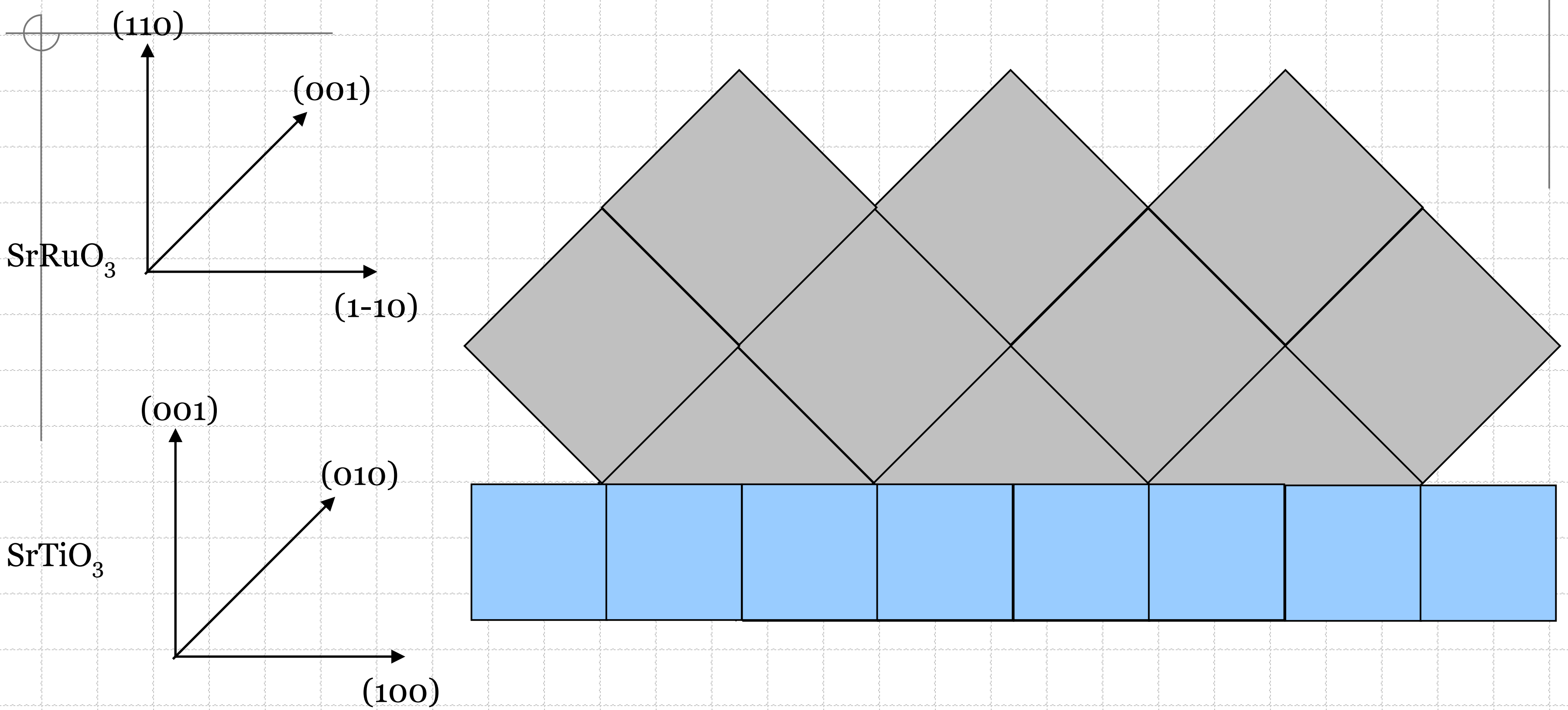
a = 5.578 Å
c = 7.908 Å

→ 510-702 °C

a = 3.956 Å

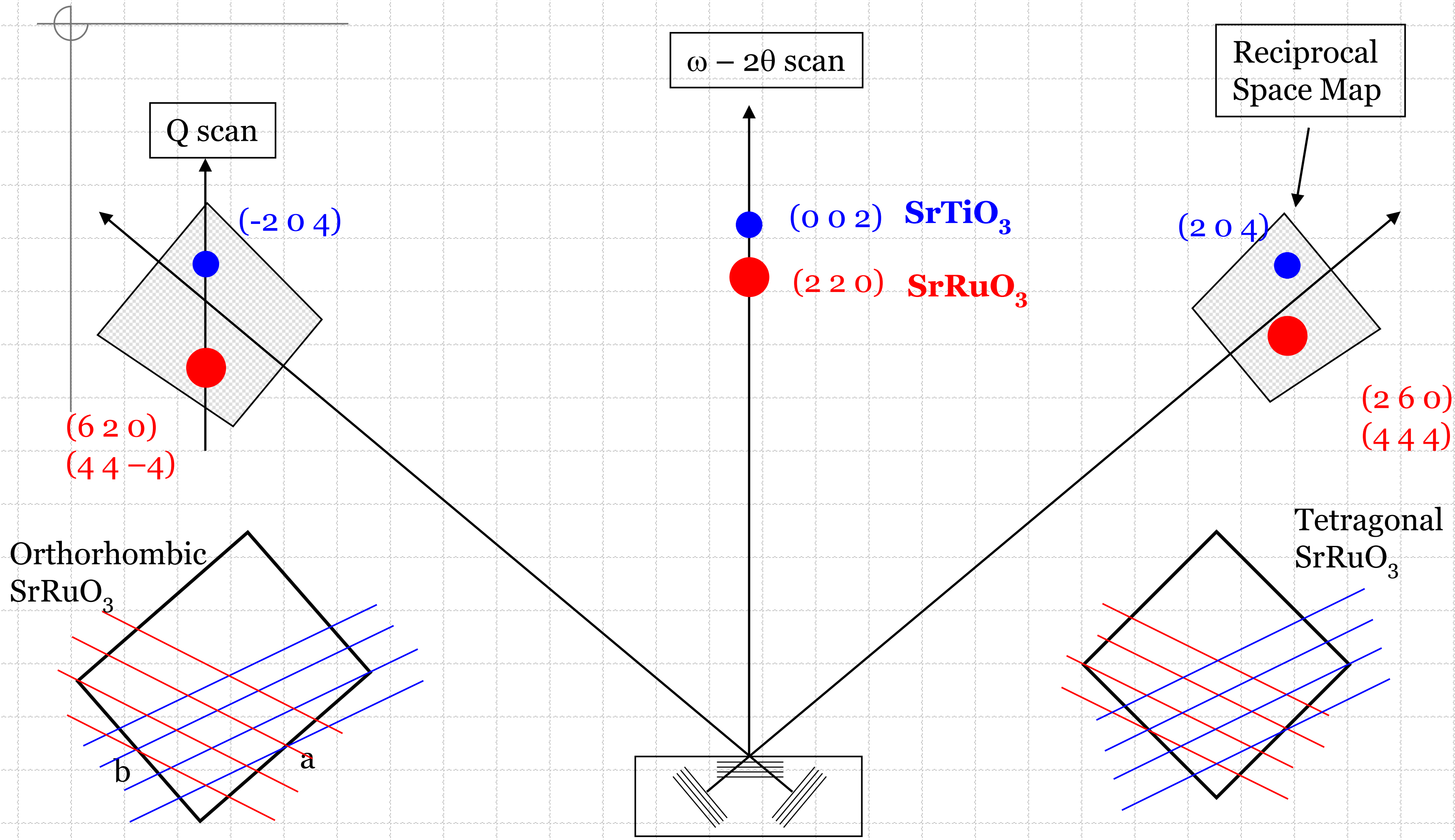
Structure of $SrRuO_3$





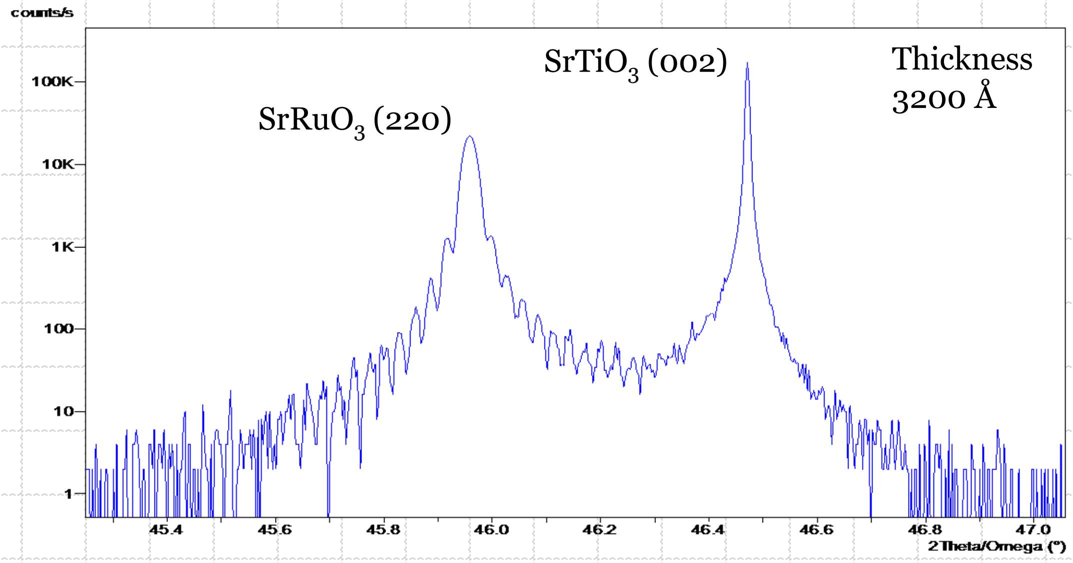
(110)
(001)
SrRuO3
(1-10)
(001)
(010)
SrTiO3
(100)

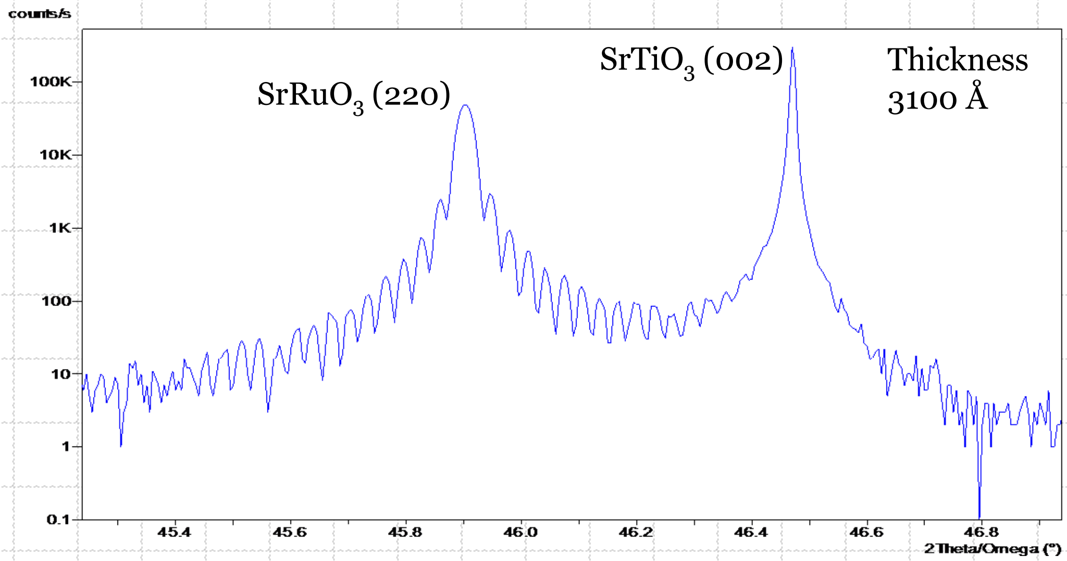
X-ray Diffraction Scan Types



$\omega - 2\theta$ symmetrical scans

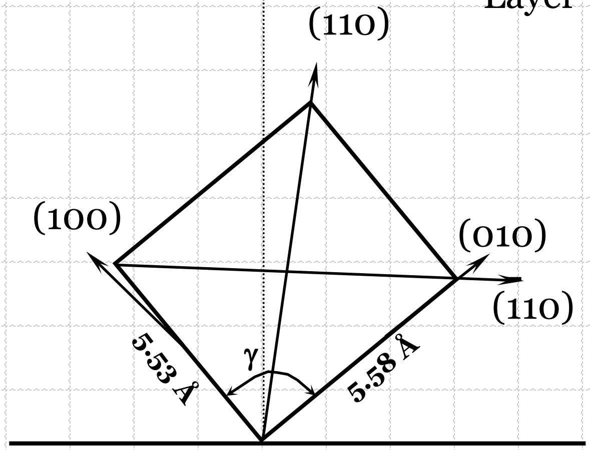
Finite size fringes indicate well ordered films

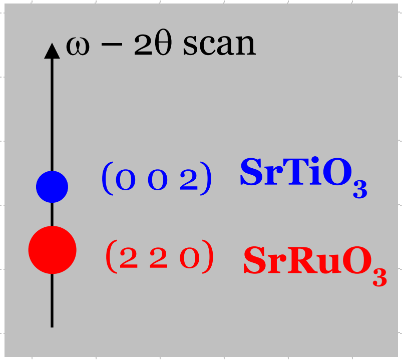


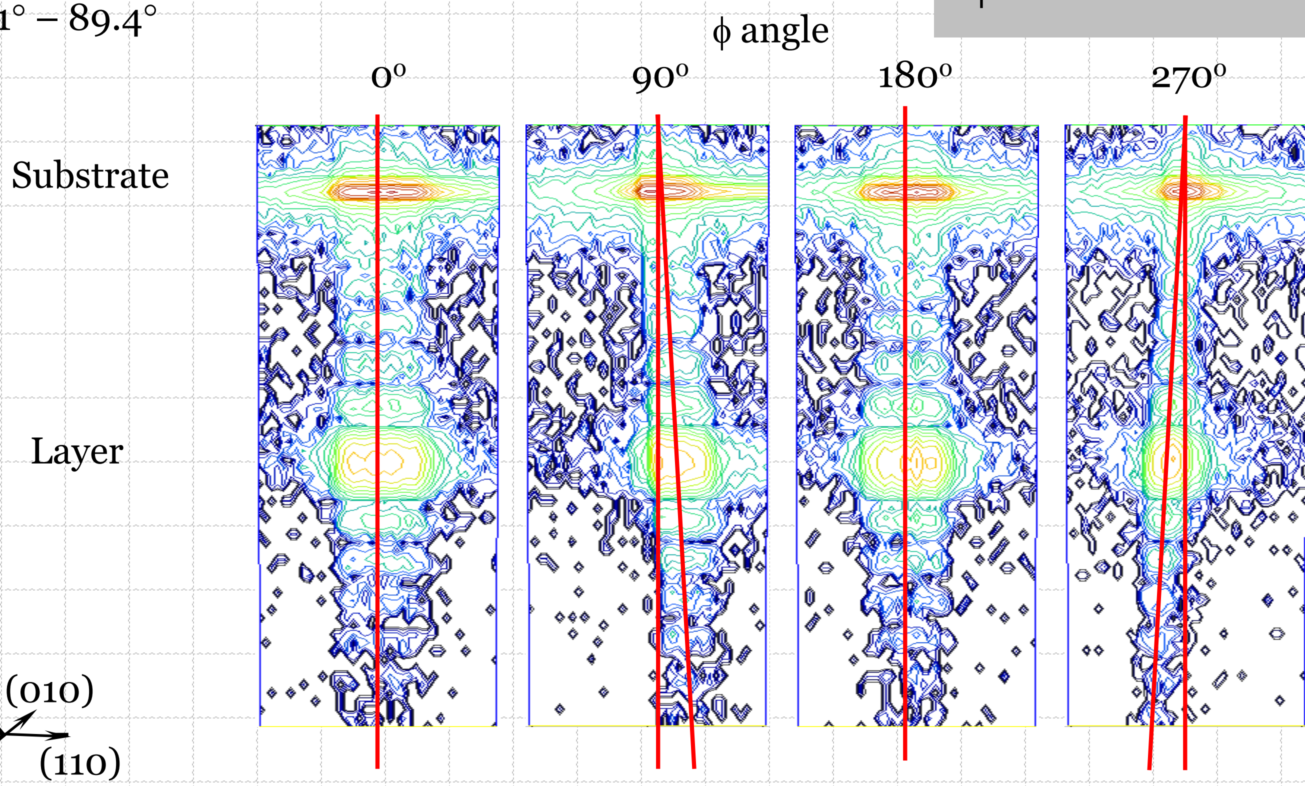


Reciprocal Lattice Map of SrRuO3 (220) and SrTiO3 (002)

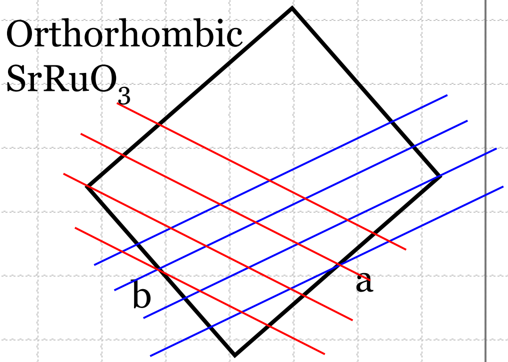
Distorted perovskite structure:
Films are slightly distorted from orthorhombic, $\gamma = 89.1° – 89.4°$

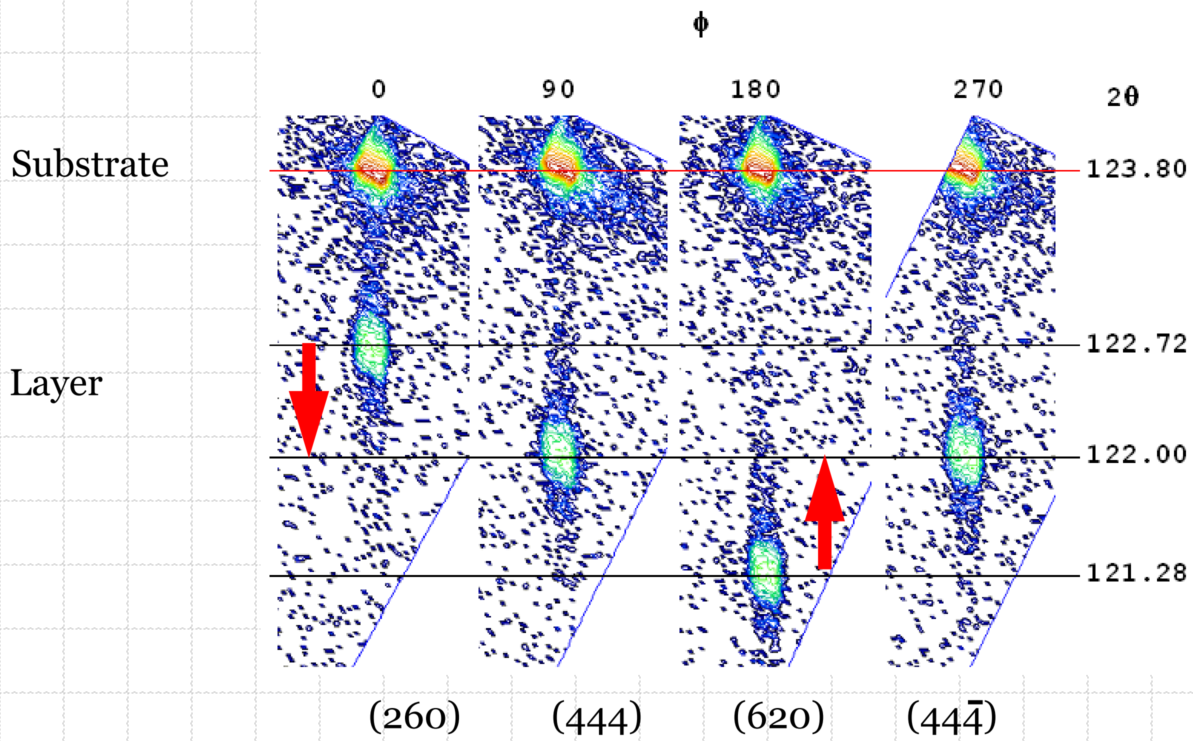




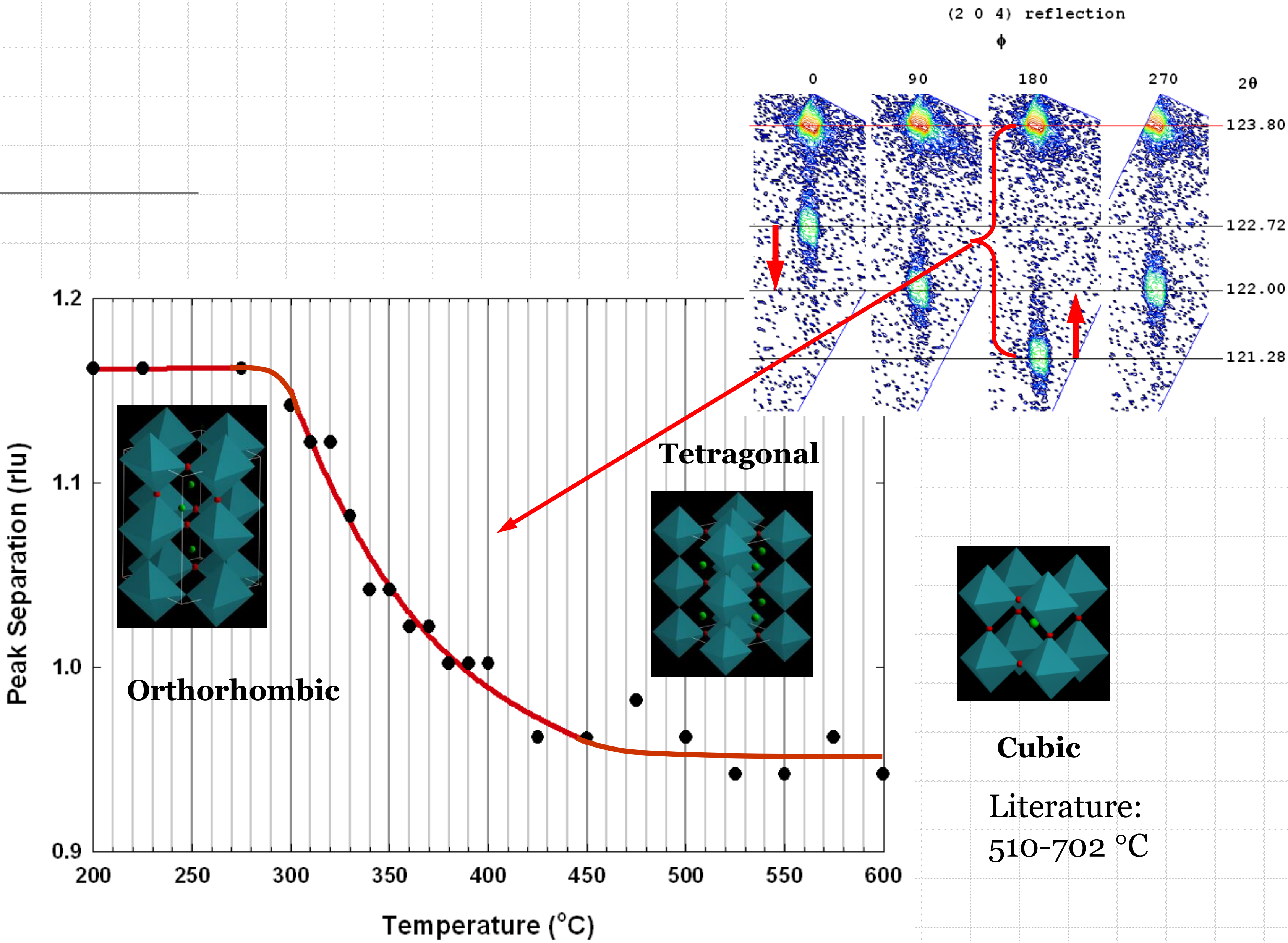


High-Resolution Reciprocal Area Mapping





Orthorombic to Tetragonal Transition

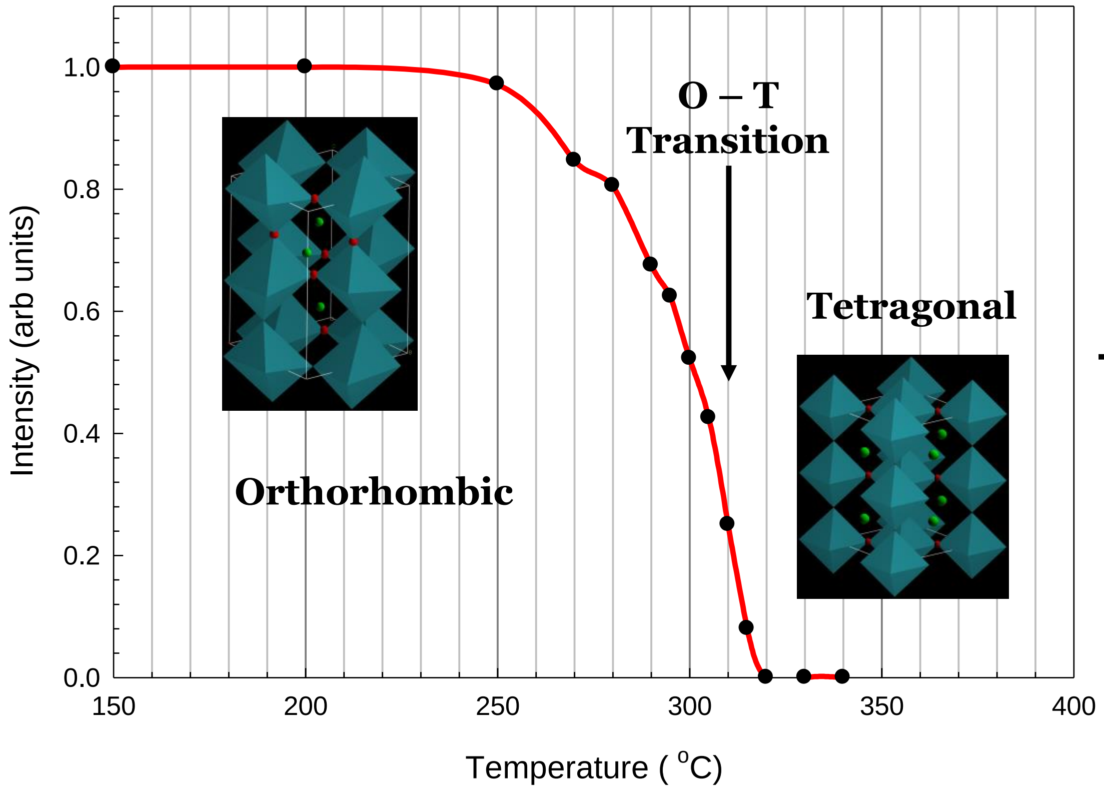


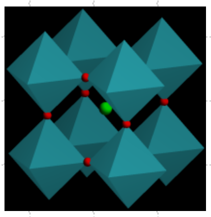
Literature:
510-702 °C

Transition Orthorhombic to Tetragonal ~ 350 °C

Structural Transition, (221) reflection

(221) Peak	
Orthorhombic	**Present**
Tetragonal	**Absent**





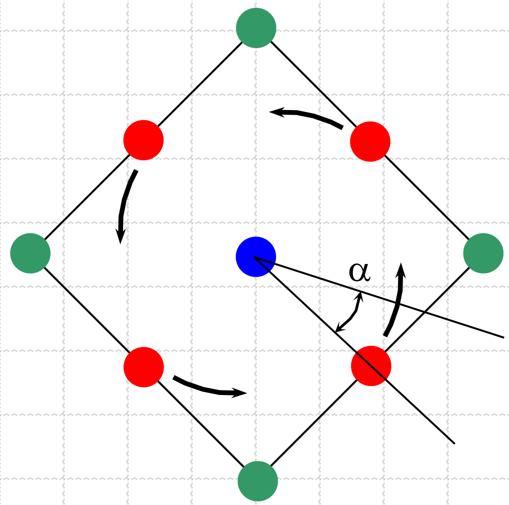
Cubic

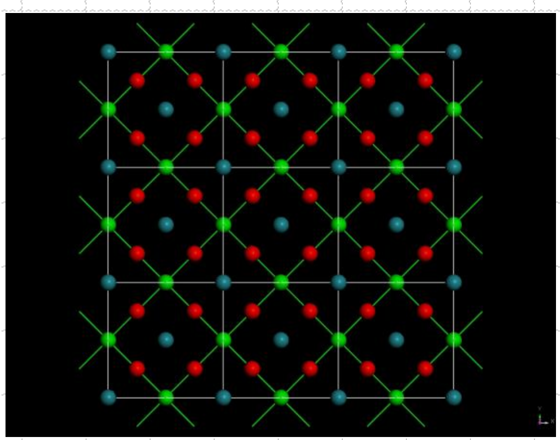
Literature:
510-702 °C

Transition Orthorhombic to Tetragonal ~ 310 °C

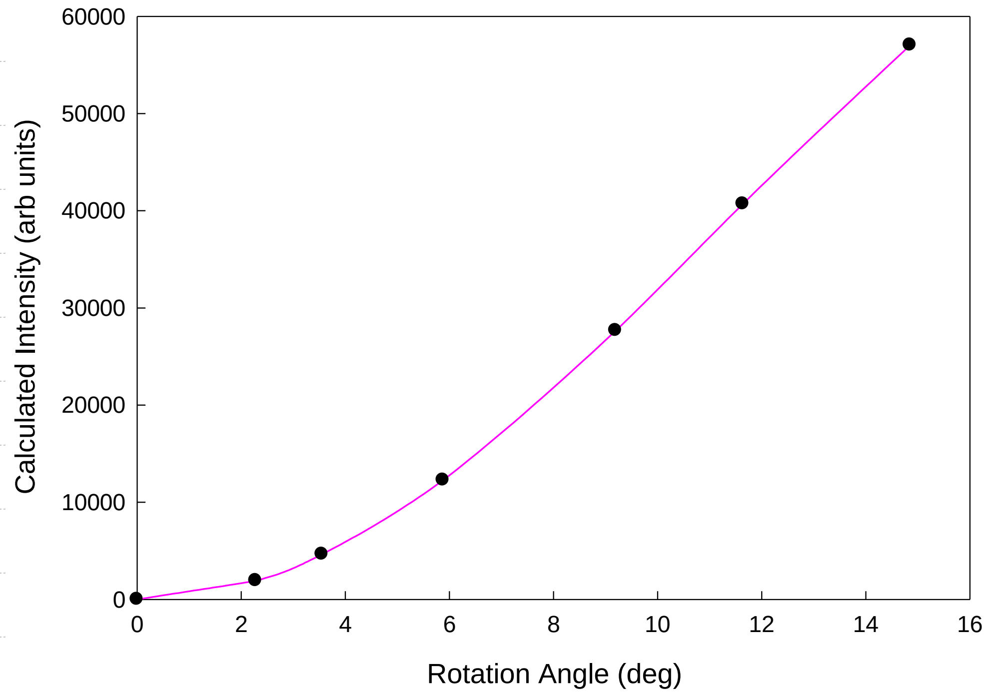
Transition Orthorhombic to Tetragonal ~ 310 °C

Structural Transition, (211) reflection

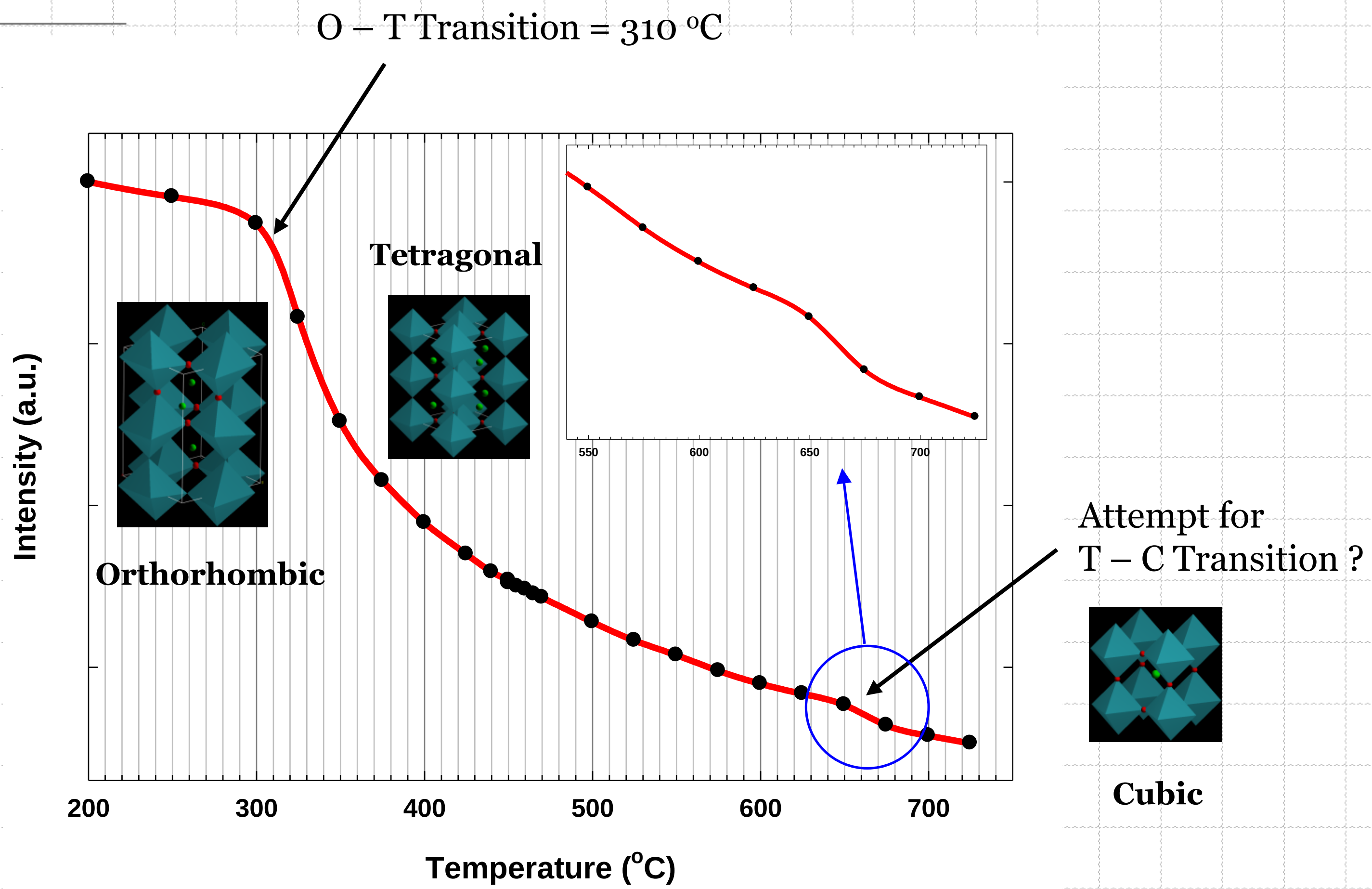




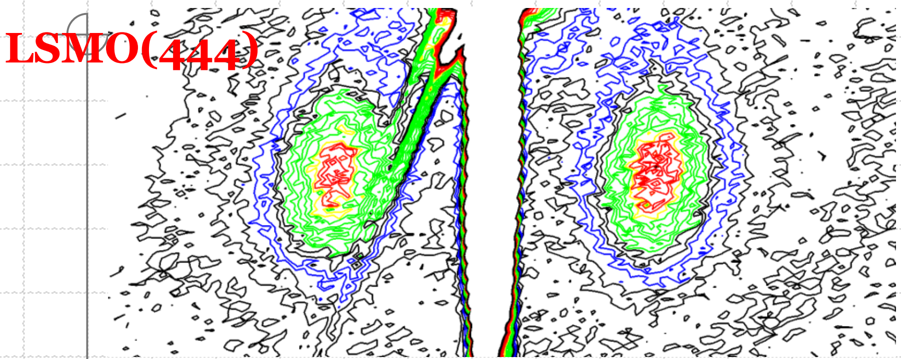
(211) peak is **absent** in cubic $SrRuO_3$



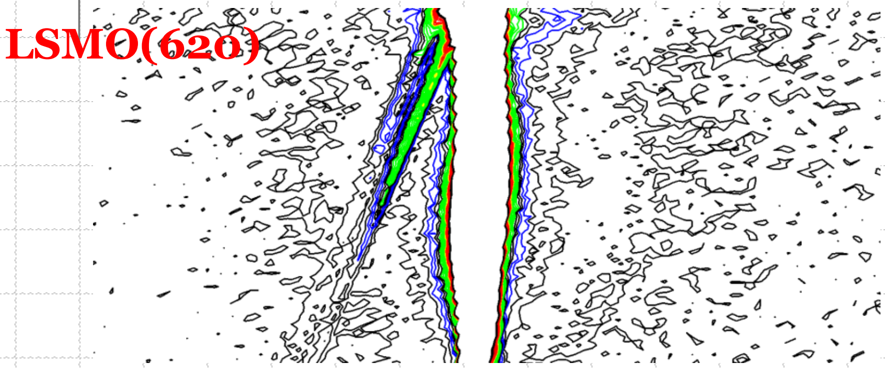
Structural Transition, (211) reflection

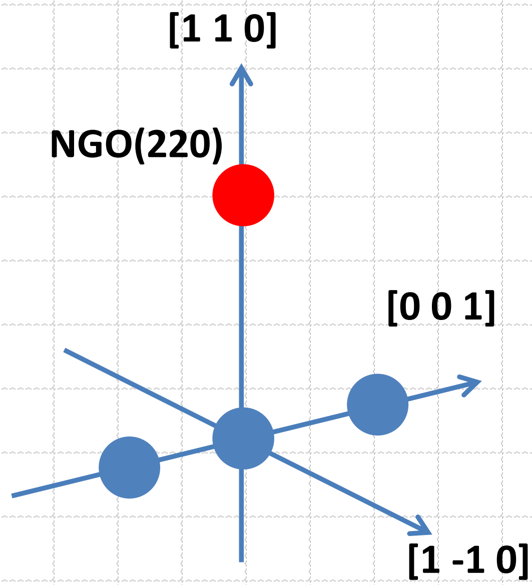


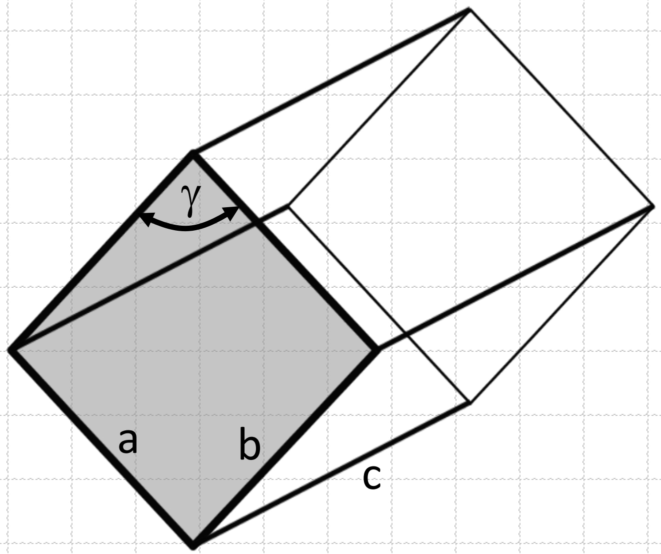
Beam along [001]



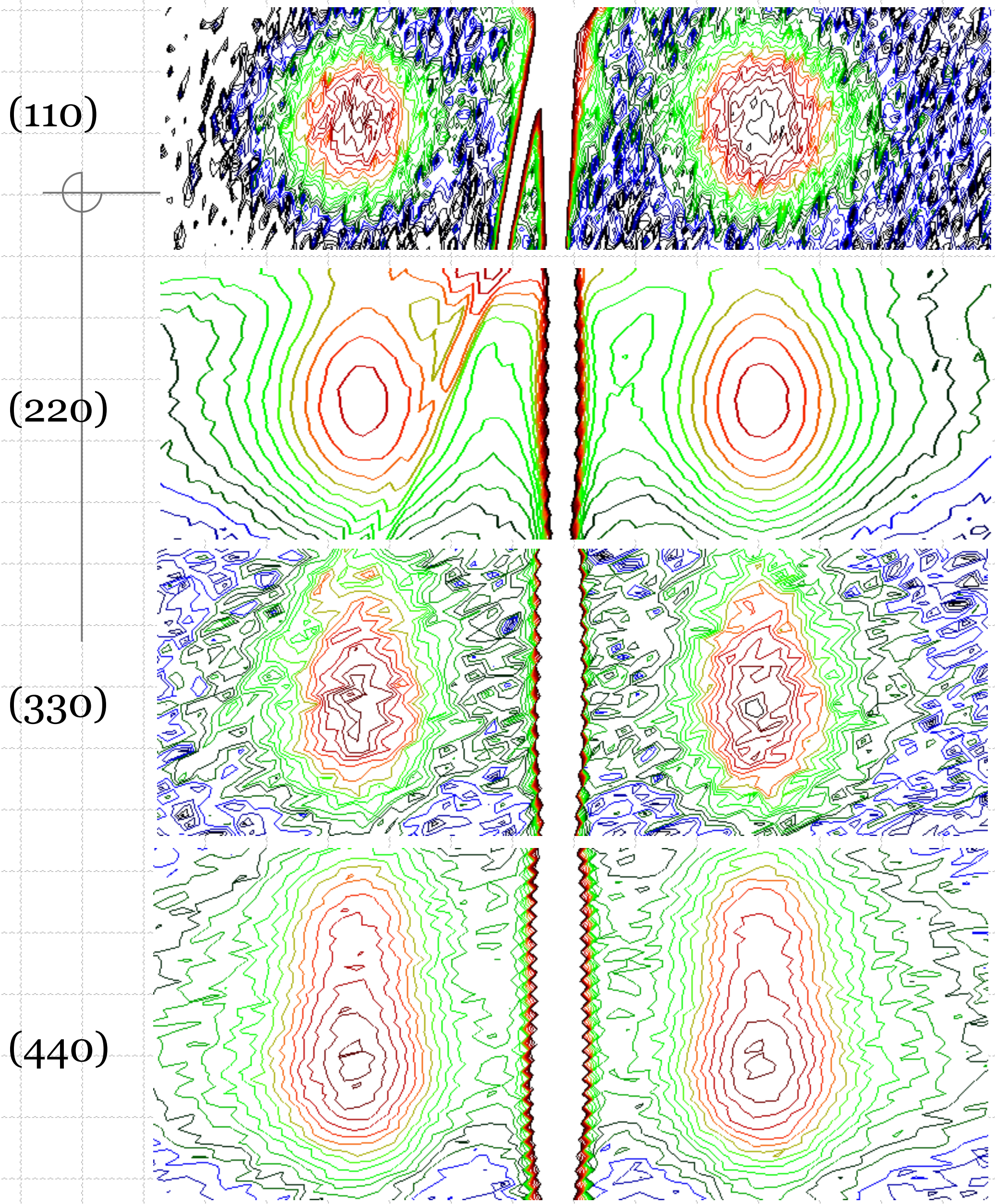
Beam along [1-10]



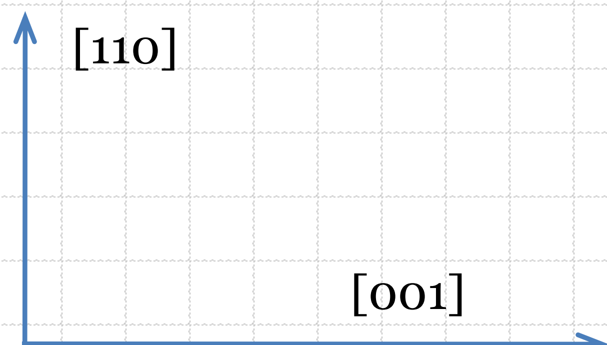


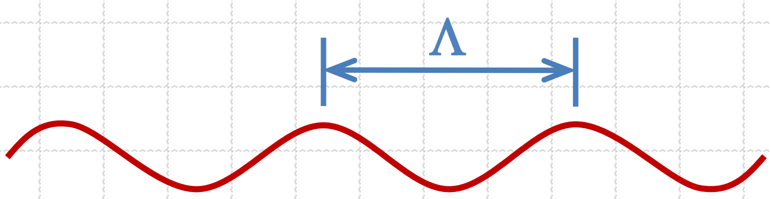


LSMO_NGO



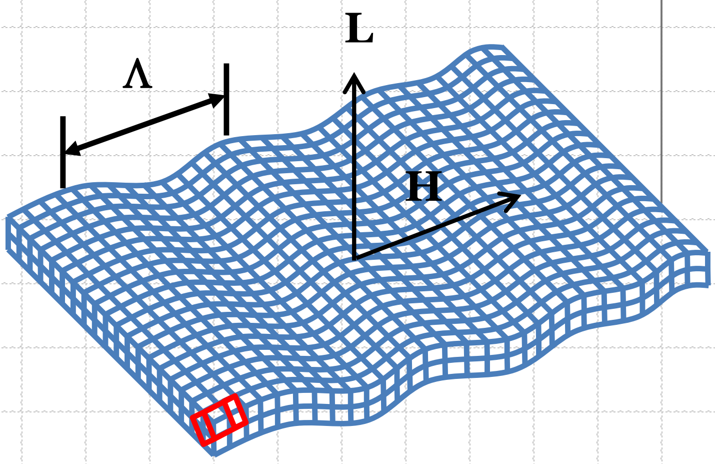
Satellite spacing does not change along out-of-plane direction





The lattice modulations described using kinematical x-ray diffraction.

We used a cubic LSMO unit cell with $a_{c||} = c/2$ and $a_{c\perp} = a^2 + b^2 - 2ab\cos(180-\gamma)$



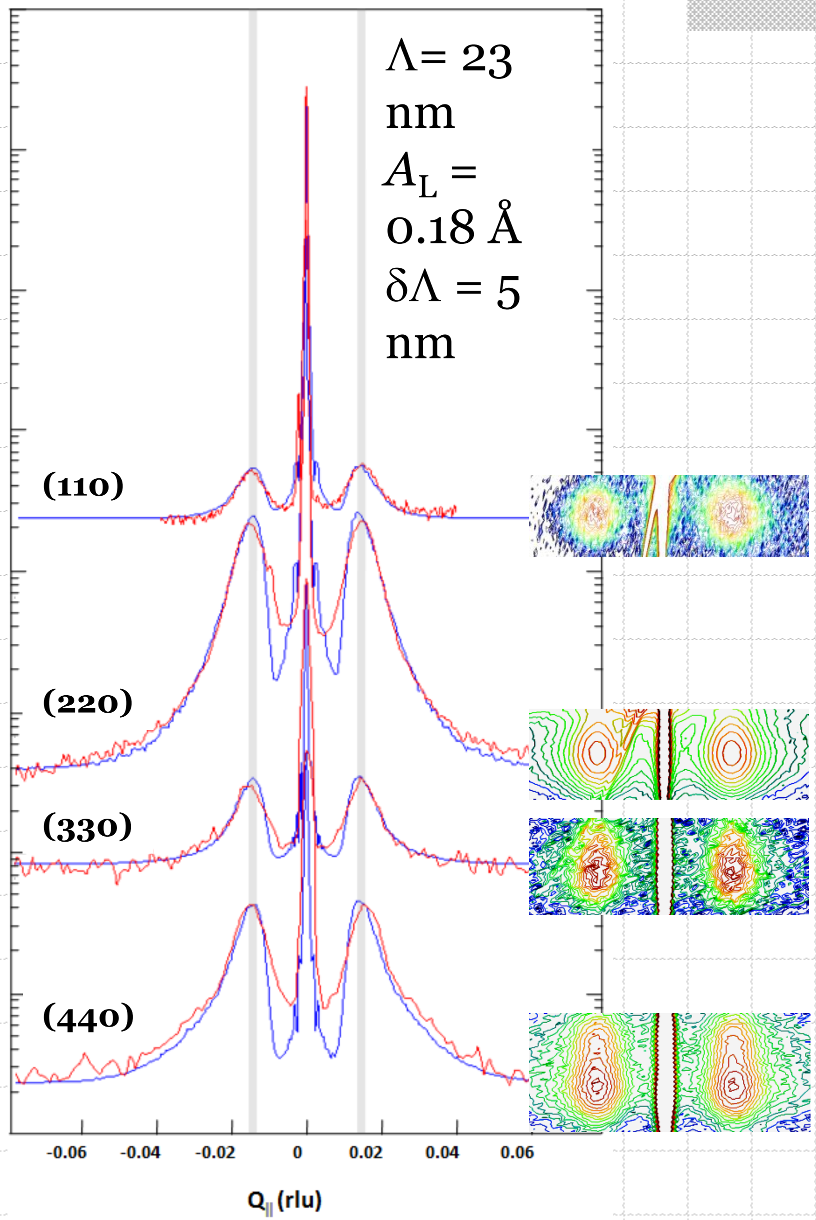
Unit cells have displacements along L-direction which are periodic along H-direction with periodicity, Λ. Then a one-dimensional complex structure factor along H-direction can be written as:

$$F_H = F_{uc}\sum_j e^{2\pi i(Hx_j+Lz_j)} = F_{uc}\sum_{n=1}^{N} e^{2\pi i\left(H\frac{x_n}{a_{c||}}+L\frac{z_n}{a_{c\perp}}\right)}$$

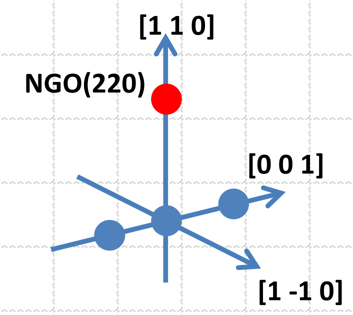
$F_{uc}, x_j = \frac{x_n}{a_{c||}}, z_j = \frac{z_n}{a_{c\perp}}$ unit cell structure factor and relative x- and z-positions along H and L directions of a LSMO cubic unit cell

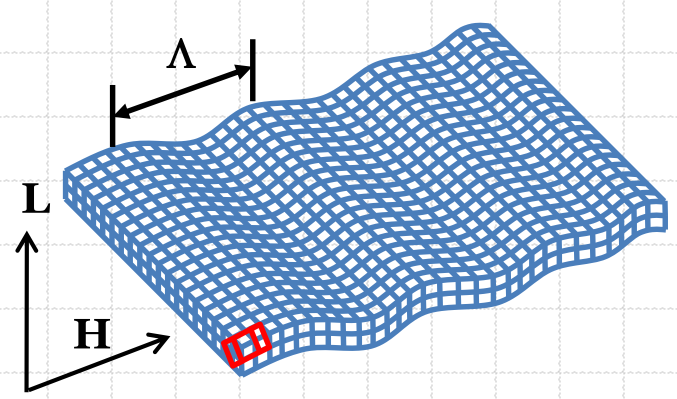
if $x_n = na_{c||}$ and $z_n = A_L\cos(k_H n a_{c||})$ then:

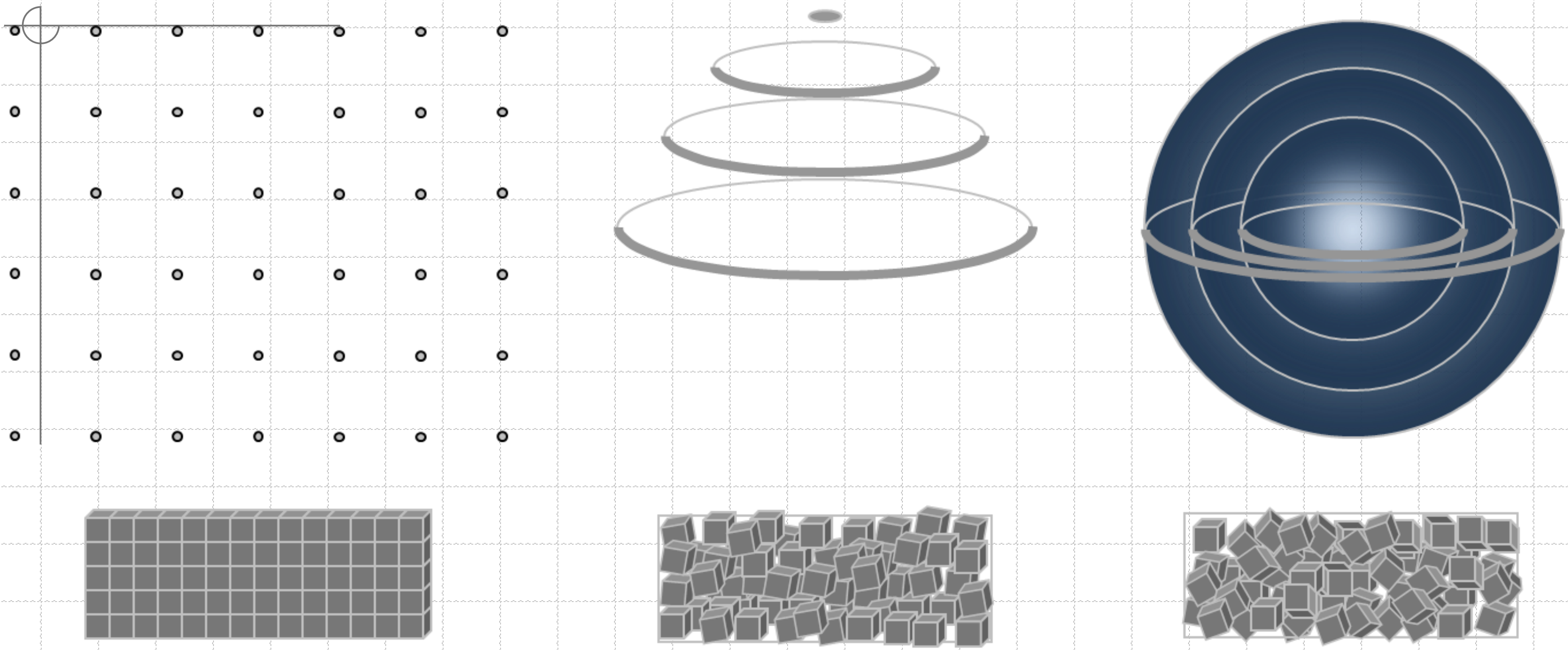
$$F_H = F_{uc}\sum_{n=1}^{N} e^{2\pi i\left[Hn+\frac{L}{a_{c\perp}}A_L\cos(k_H n a_{c||})\right]}$$



H-scans around LSMO(hk0) reflections with $h = k = 1,2,3,4$. The gray vertical lines are guides to the eye and show that satellite peaks originate from periodic atomic displacements and not from out-of-plane twinning.







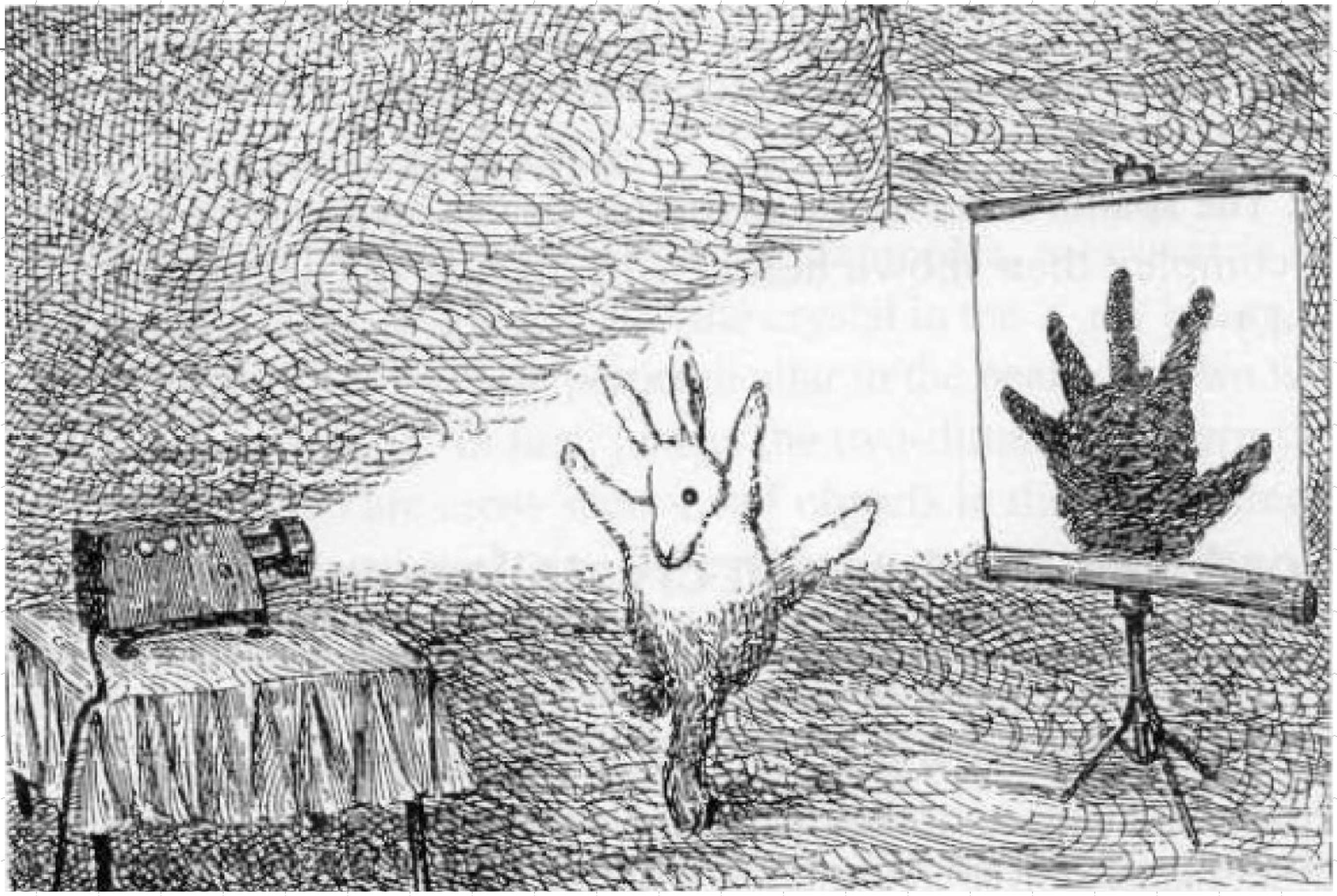
Single crystal | Preferred orientation | Polycrystalline

- Reciprocal space for epitaxial thin films is very rich.

- Shape and positions of reciprocal lattice points with respect to the substrate reveal information about:
  - Mismatch
  - Strain state
  - Relaxation
  - Mosaicity
  - Composition
  - Thickness ....

- Diffractometer instrumental resolution has to be understood before measurements are performed.

Things can look very different in reciprocal space than in real space…



Fun in reciprocal space.