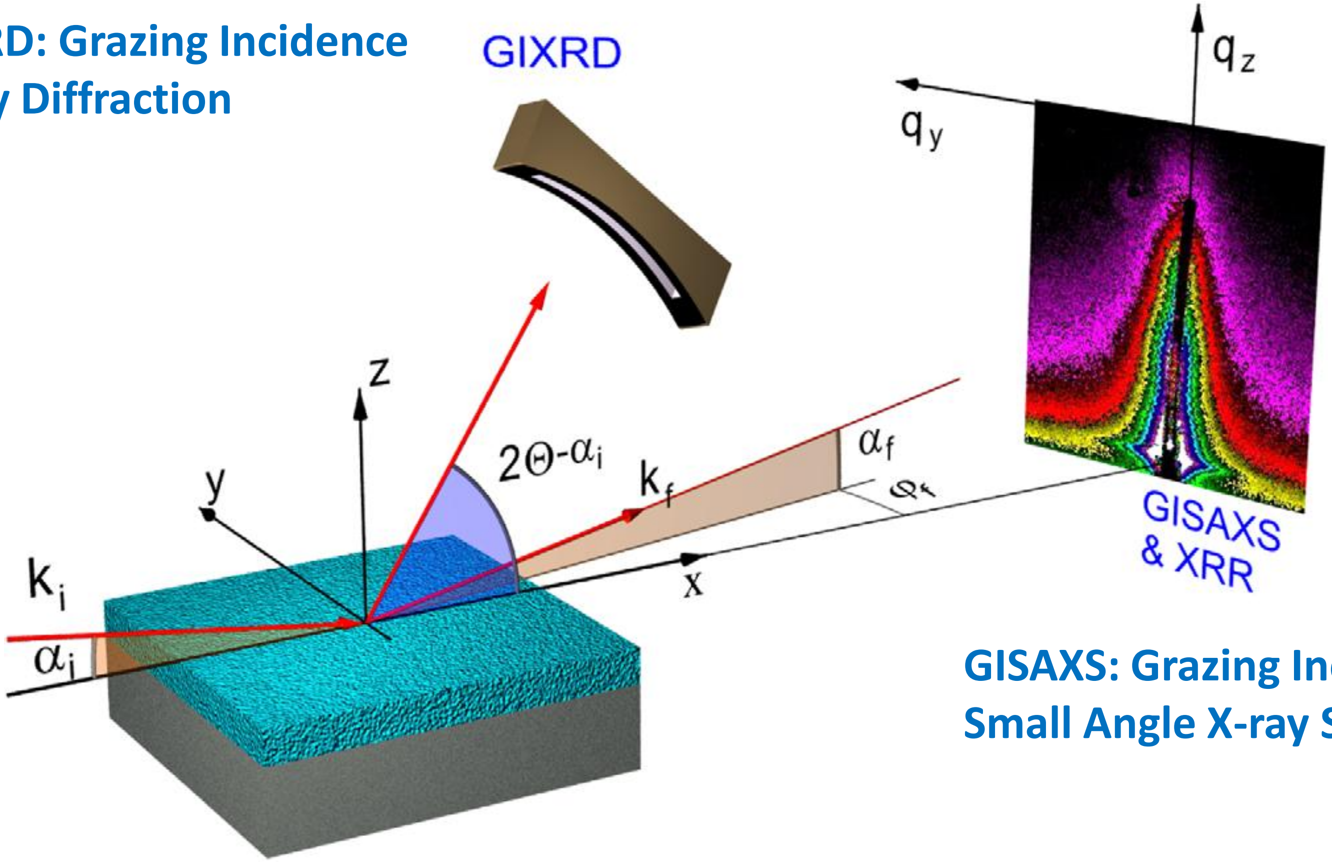
Introduction of X-ray Reflectivity

X-ray Techniques

GIXRD: Grazing Incidence X-ray Diffraction

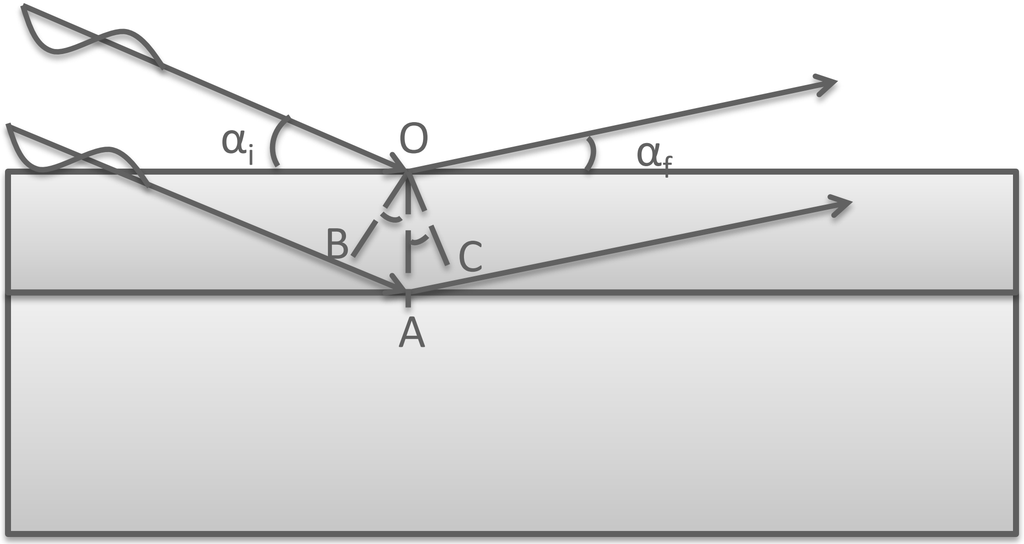


GISAXS: Grazing Incidence Small Angle X-ray Scattering

In GISAXS, the angle α_i is very small (<0.5^o) for GISAXS, X-ray penetrates the sample and reflection is very strong, beam stopper is required to protect detector.
In our experiment, α_i =1.8^o, beam intensity is reduced dramatically, no stopper.

Salamon *et al,* J. Phys. D: Appl. Phys. 46 (2013) 095304

Simple Explanation - consider as diffraction of scattered x-ray



$$AB = AO \bullet \sin\alpha_i, AC = AO \bullet \sin\alpha_f$$

In order to get interference pattern,

$$AB + AC = m\lambda (m = 1,2,3 \ldots)$$

$$d\sin\alpha_i + d\sin\alpha_f = m\lambda$$

$$\frac{\sin\alpha_i + \sin\alpha_f}{\lambda} = \frac{m}{d}$$

Given wave-vector transfer

$$q_z = \frac{2\pi}{\lambda}\left(\sin\alpha_i + \sin\alpha_f\right)$$

$$q_z = \frac{2\pi m}{d}$$

$$q_{z,1} = \frac{2\pi}{d}$$

$$q_{z,2} = \frac{2\pi * 2}{d}$$

.....

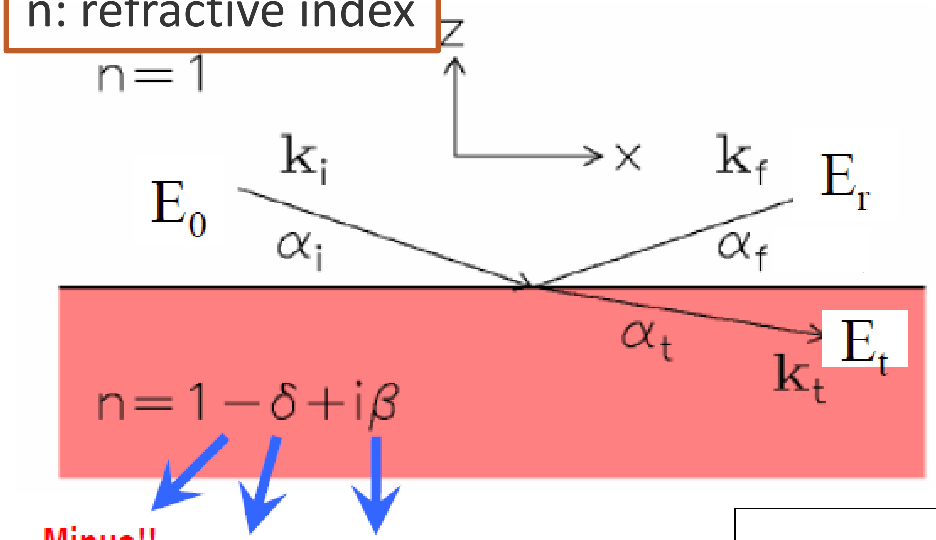
$$q_{z,j} = \frac{2\pi * j}{d}$$

$$\Delta q_z = \frac{2\pi}{d}$$

$$\boxed{d = \frac{2\pi}{\Delta q_z}}$$

Reflection and Transmission at Single Surface

n: refractive index



Minus!!

Dispersion

Absorption

$$\delta = \frac{\lambda^2}{2\pi} r_0 \rho \approx 10^{-4}..10^{-6}$$

$$\beta = \frac{\lambda}{4\pi} \mu \approx 10^{-6}..10^{-9}$$

Snell-Descartes law: $\cos\alpha_i = n \cos\alpha_t$

∃ transmitted wave only if $\cos(\alpha_t) \le 1$, i.e. $\alpha_i \ge \alpha_c$

If $\alpha_i \le \alpha_c$,
- Incident wave totally externally reflected.
- Transmitted wave exponentially damped with z.

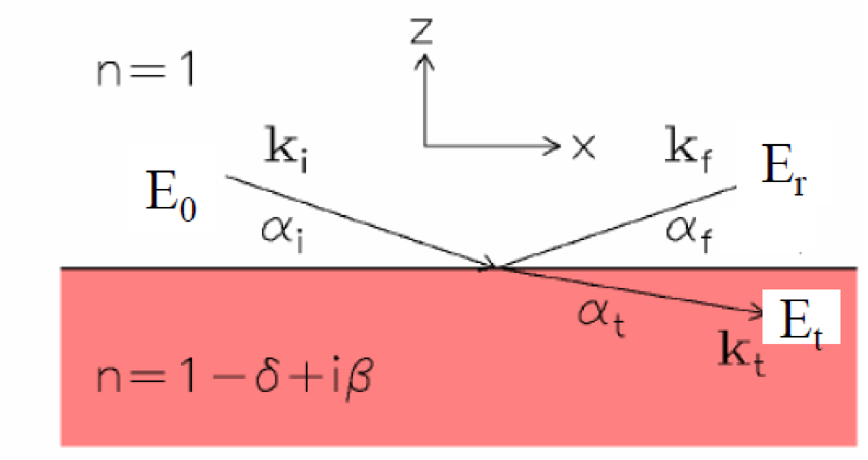
α_c critical angle for total external reflection of X-rays

$$\alpha_c = \sqrt{2\delta} = \sqrt{\frac{r_0}{\pi}} \times \lambda \times \sqrt{\rho} \approx 0.1 \text{ to } 0.5^\circ$$

Reflection and Transmission at Single Surface

- Fresnel equations:

Relationships between the amplitudes of incident, transmitted and reflected beam.



wave-vector transfer

$$q_z = \frac{2\pi}{\lambda}\left(\sin\alpha_i + \sin\alpha_f\right)$$

Amplitude

Intensity

Reflection

$$r(q_z) = \frac{E_r}{E_0} = \frac{q_z - \sqrt{q_z^2 - q_c^2}}{q_z + \sqrt{q_z^2 - q_c^2}}$$

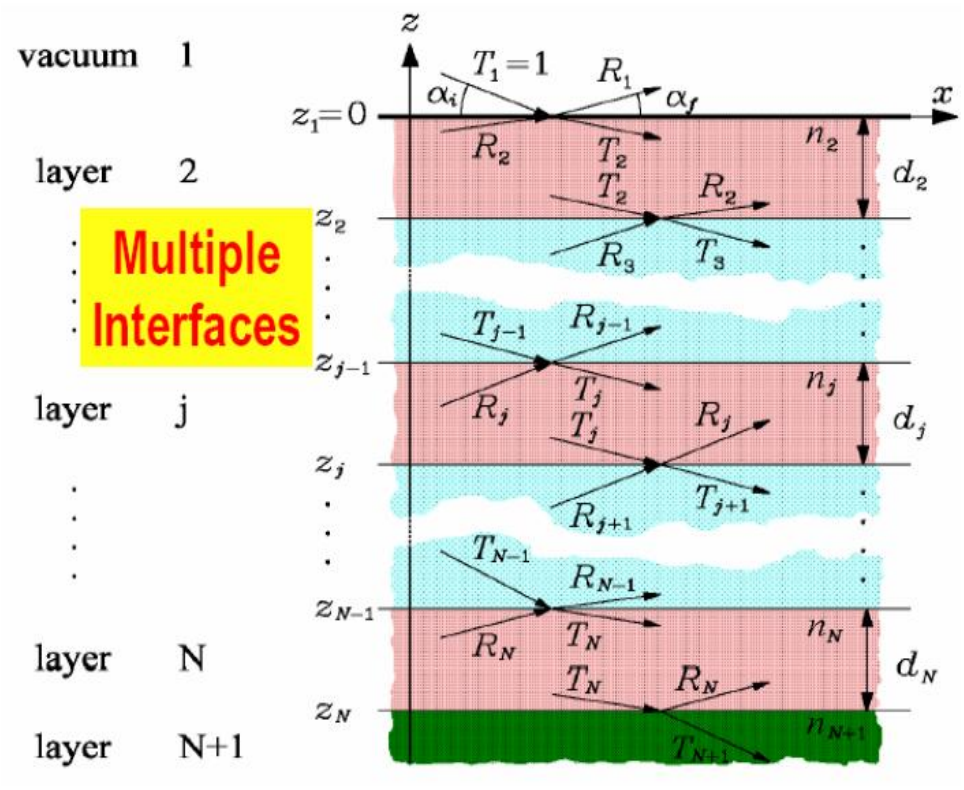
$$R = rr^* = |r|^2 = \left|\frac{E_r}{E_0}\right|^2$$

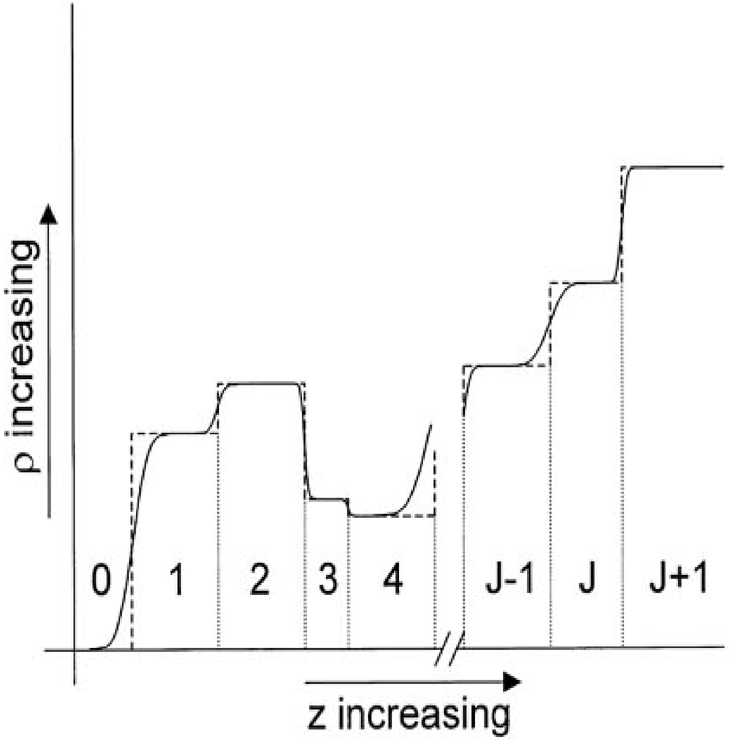
Transmission

$$t(q_z) = \frac{E_t}{E_0} = \frac{2q_z}{q_z + \sqrt{q_z^2 - q_c^2}}$$

$$T = tt^* = |t|^2 = \left|\frac{E_t}{E_0}\right|^2$$

Reflectivity from Multiple Layers





$$q_{z,j} = \sqrt{q_z^2 - q_{c,j}^2}.$$

$$r_{j,j+1} = \frac{q_{z,j} - q_{z,j+1}}{q_{z,j} + q_{z,j+1}},$$

$q_{c,j}$ is the wave-vector transfer in medium j at critical angle

$$\boxed{r = r_{0,1} + r_{1,2}e^{iq_{z,1}d_1} + r_{2,3}e^{i(q_{z,1}d_1 + q_{z,2}d_2)} + \cdots + r_{j,j+1}e^{i\sum_{k=0}^{j} q_{z,k}\, d_k} + \cdots}$$

Approximation

$$r = r_{0,1} + r_{1,2}e^{iq_{z,1}d_1} + r_{2,3}e^{i(q_{z,1}d_1+q_{z,2}d_2)} + \cdots + r_{j,j+1}e^{i\sum_{k=0}^{j} q_{z,k}\, d_k} + \cdots \tag{1}$$

$$R(q_z) = \left| \sum_{j=0}^{n} r_{j,j+1} e^{iq_z z_j} \right|^2 \text{ with } r_{j,j+1} = \frac{q_{z,j} - q_{z,j+1}}{q_{z,j} + q_{z,j+1}}.$$

A further approximation consists in neglecting the refraction and the absorption in the material in the phase factor in Eq. (1):

$$r = \sum_{j=0}^{n} r_{j,j+1}\, e^{iq_z \sum_{m=0}^{j} d_m}.$$

A final approximation consists in assuming that the wave vector q_z does not change significantly from one medium to the next so that the sum in the denominator of $r_{j,\, j+1}$ may be simplified:

$$r_{j,j+1} = \frac{q_{z,j}^2 - q_{z,j+1}^2}{(q_{z,j} + q_{z,j+1})^2} = \frac{q_{c,j+1}^2 - q_{c,j}^2}{4q_z^2} = \frac{4\pi r_e(\rho_{j+1} - \rho_j)}{q_z^2} \tag{2}$$

Where $q_{c,j} = \sqrt{16\pi r_e \rho_j}$

r_e is the classical radius of the electron

ρ_j is the electron density of layer j

Approximation

$$r_{j,j+1} = \frac{q_{z,j}^2 - q_{z,j+1}^2}{(q_{z,j} + q_{z,j+1})^2} = \frac{q_{c,j+1}^2 - q_{c,j}^2}{4q_z^2} = \frac{4\pi r_e(\rho_{j+1} - \rho_j)}{q_z^2} \qquad (2)$$

Thus,

$$r = 4\pi\, r_e \sum_{j=1}^{n} \frac{(\rho_{j+1} - \rho_j)}{q_z^2}\, e^{iq_z \sum\limits_{m=0}^{j} d_m}.$$

If the origin of the *z* axis is chosen to be at the upper surface (medium 0 at a depth of $z_1 = 0$), consider that the material is made of an infinite number of thin layers, the sum may then be transformed into an integral over *z*, and the reflection coefficient becomes:

$$\boxed{r = \frac{4\pi\, r_e}{q_z^2} \int_{-\infty}^{+\infty} \frac{d\rho(z)}{dz}\, e^{iq_z z}\, dz} \qquad (3)$$

$\rho(z)$ is the electron density at z altitude

Replacing $(4\pi r_e \rho_s)^2 / q_z^4$ by $R_F(q_z)$:

$$\boxed{R(q_z) = r.r^* = R_F(q_z) \left| \frac{1}{\rho_s} \int_{-\infty}^{+\infty} \frac{d\rho(z)}{dz} e^{iq_z z} dz \right|^2}$$

and

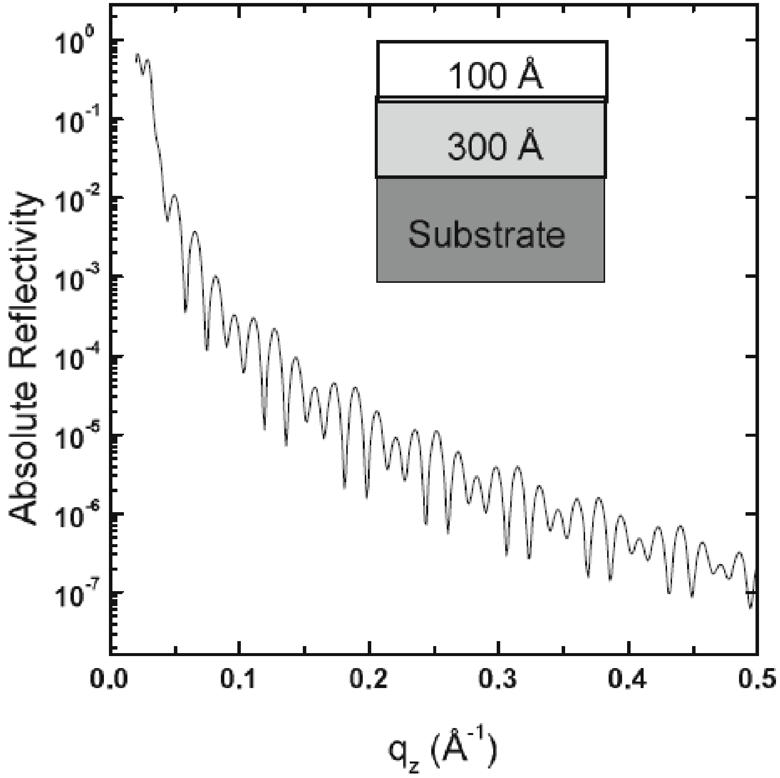
$$\boxed{\frac{R(q_z)}{R_F(q_z)} = \frac{1}{\rho_s^2} TF\left[\rho'(z) \otimes \rho'(z)\right]}$$

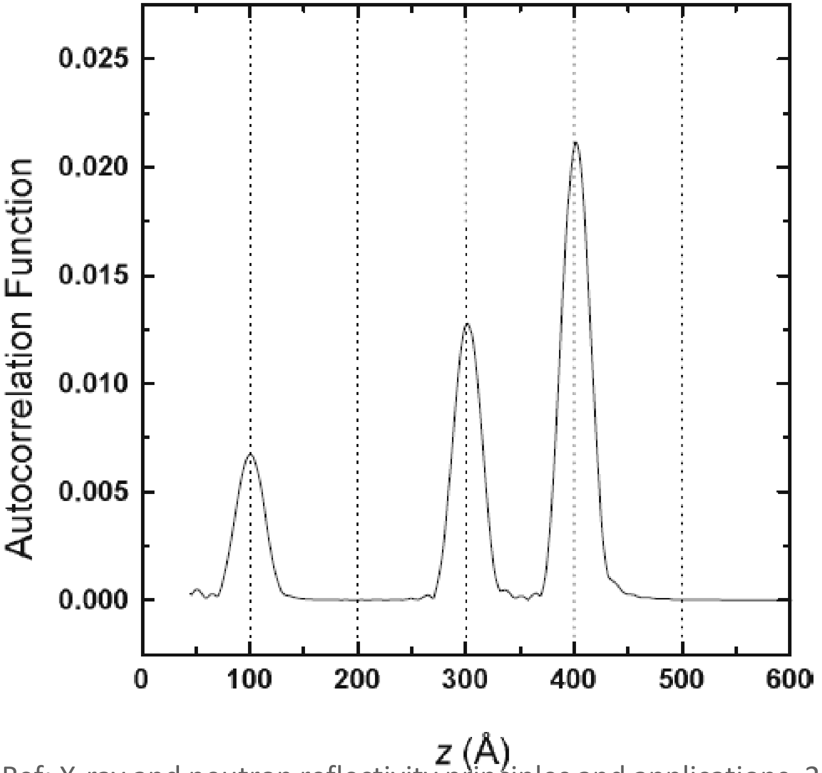
Examples

The data inversion gives the autocorrelation function of the first derivative of the electron density

$$\boxed{\frac{R(q_z)}{R_F(q_z)} = \frac{1}{\rho_s^2} TF\left[\rho'(z) \otimes \rho'(z)\right]}$$

R_F: Fresnel reflectivity of the substrate



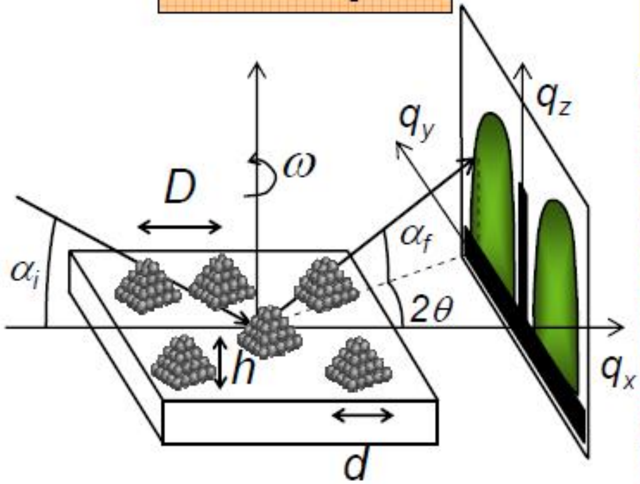


Ref: X-ray and neutron reflectivity principles and applications, 2009

Examples

Grazing Incidence Small Angle X-ray Scattering (GISAXS)

Principle

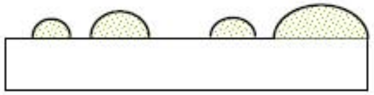


2D image around direct beam:
Fourier transform of objects

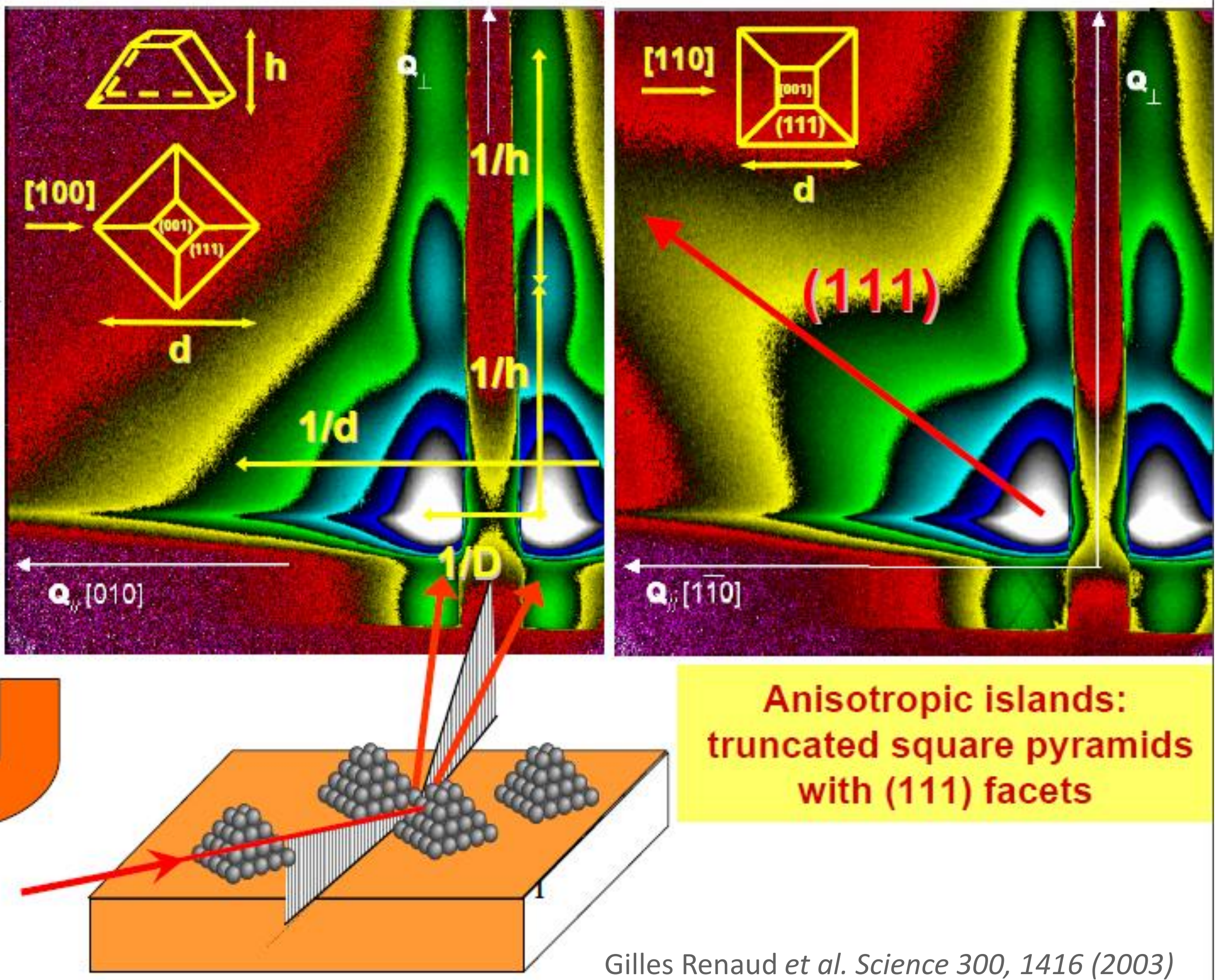
Morphology

- Shape
- Sizes
- Size distributions
- Particle-particle pair correlation function

Standard 3D growth (Volmer-Weber)



Example : **20 Å Ag/MgO(001) 500K**

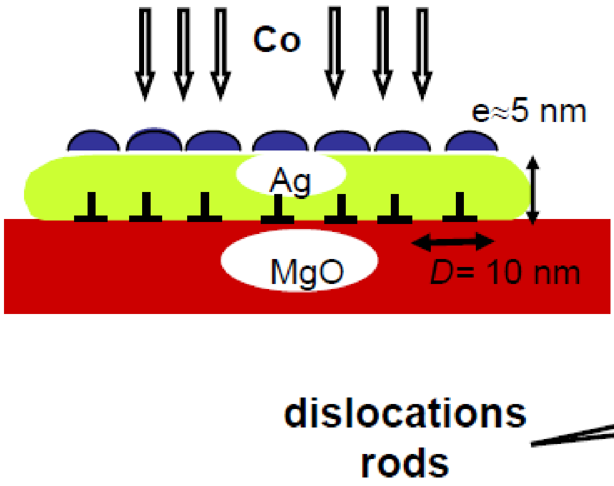


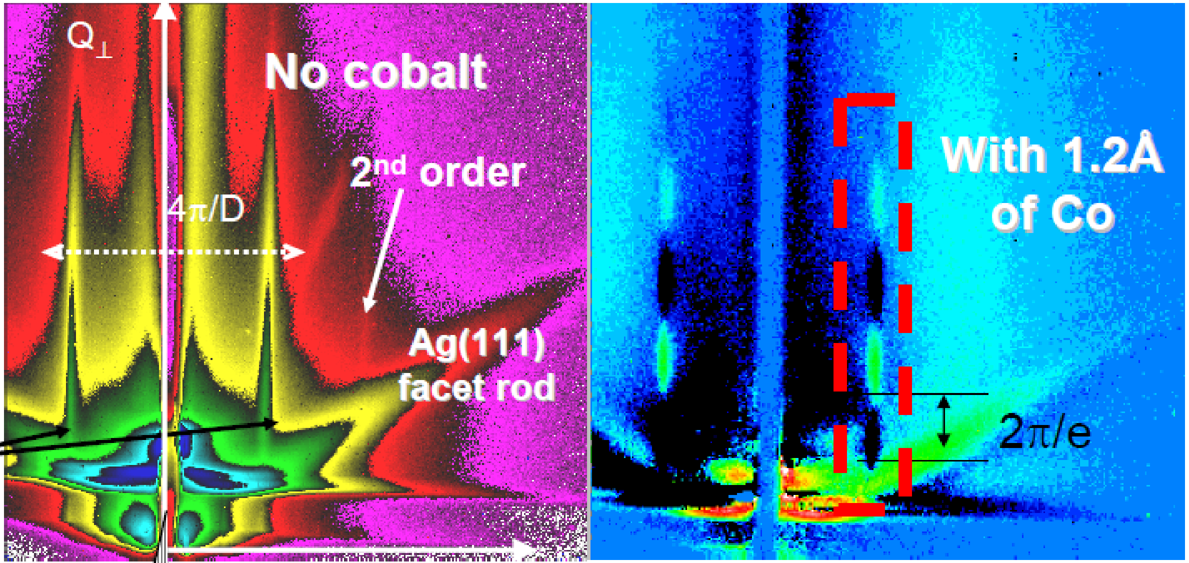
Anisotropic islands: truncated square pyramids with (111) facets

Gilles Renaud *et al. Science 300, 1416 (2003)*

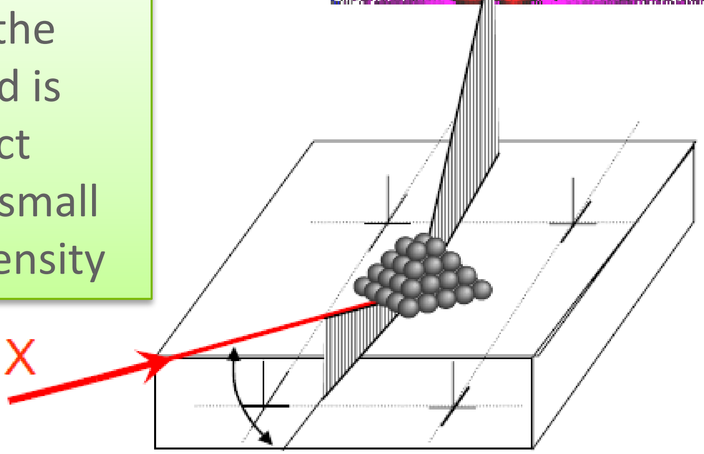
Examples

Self-organized growth of magnetic cobalt dots on an interfacial dislocation network : Co/Ag/MgO(100)





All the pictures, the center scatter rod is blocked to protect detector, due to small angle, strong intensity



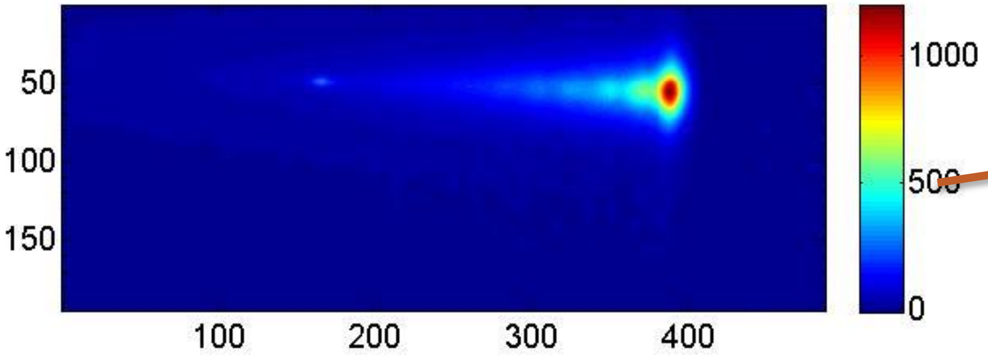
Interferences

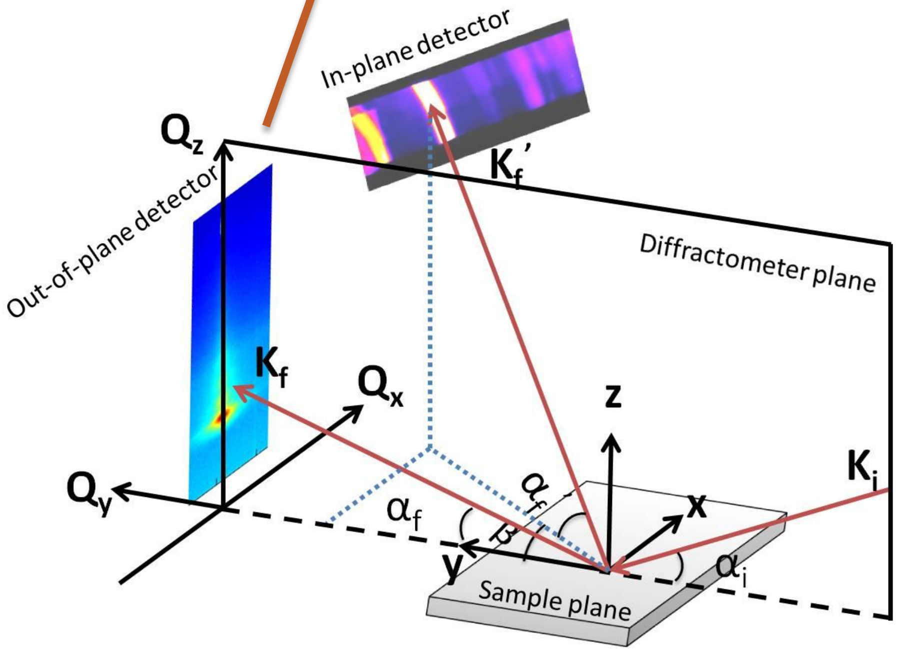
⇩

Co islands are ordered

F. Leroy et al, PRL 95, 185501 (2005)

Schematic of BNL Experiment Geometry

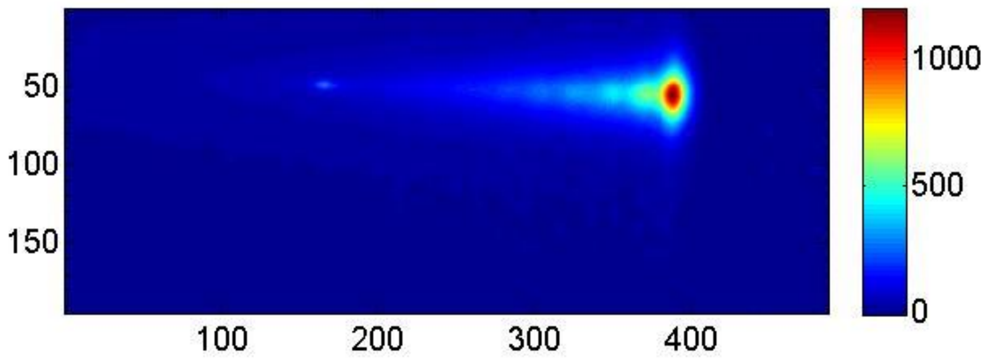


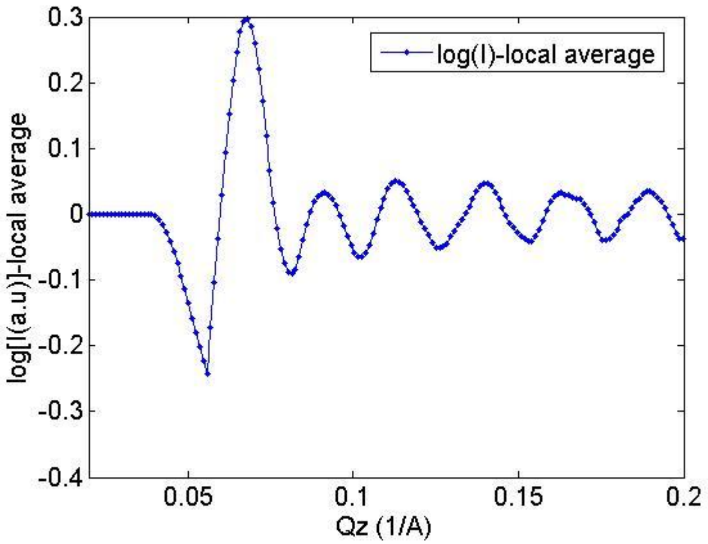


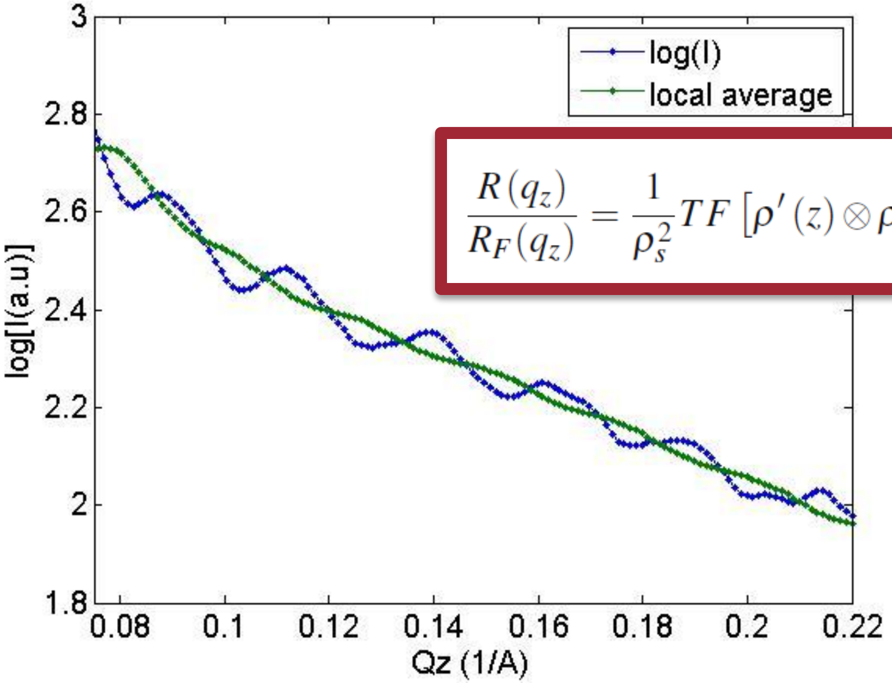
Pilatus 100K Detector System	
Pixel size	172 x 172 μm^2
Format	487 x 195 = 94 965 pixels
Active area	83.8 x 33.5 mm^2
Counting rate	> $2x10^6$ counts/s/pixel
Energy range	3 – 30 keV
Readout time	< 2.7 ms
Framing rate	> 200 Hz

K_i is the direction of incident X-ray, pointing to sample.
The recorded image is the reflected beam intensity image

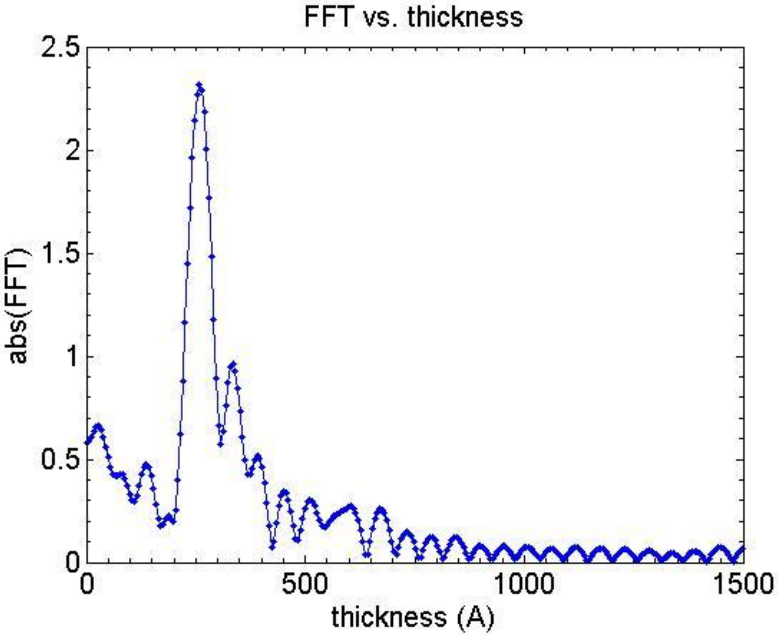
Sample: spec_start_S144_00190





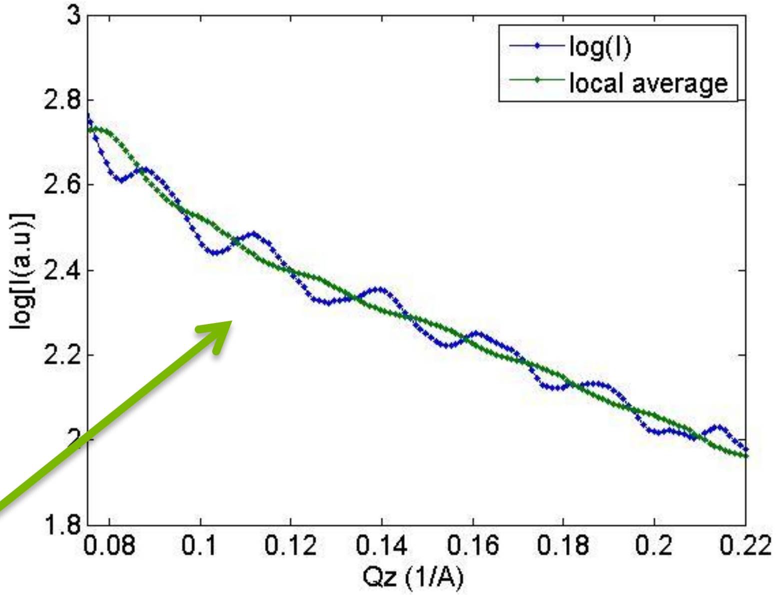


$$\frac{R(q_z)}{R_F(q_z)} = \frac{1}{\rho_s^2} TF\left[\rho'(z) \otimes \rho'(z)\right]$$



Local Average

$$\boxed{\frac{R(q_z)}{R_F(q_z)} = \frac{1}{\rho_s^2} TF\left[\rho'(z)\otimes\rho'(z)\right]}$$



$$\boxed{\log[I_0 \bullet R(q_z)]} - \boxed{\log[I_0 \bullet R_F(q_z)/\rho_s^2]} = \log\left|TF[\rho'(z)\otimes\rho'(z)]\right|$$

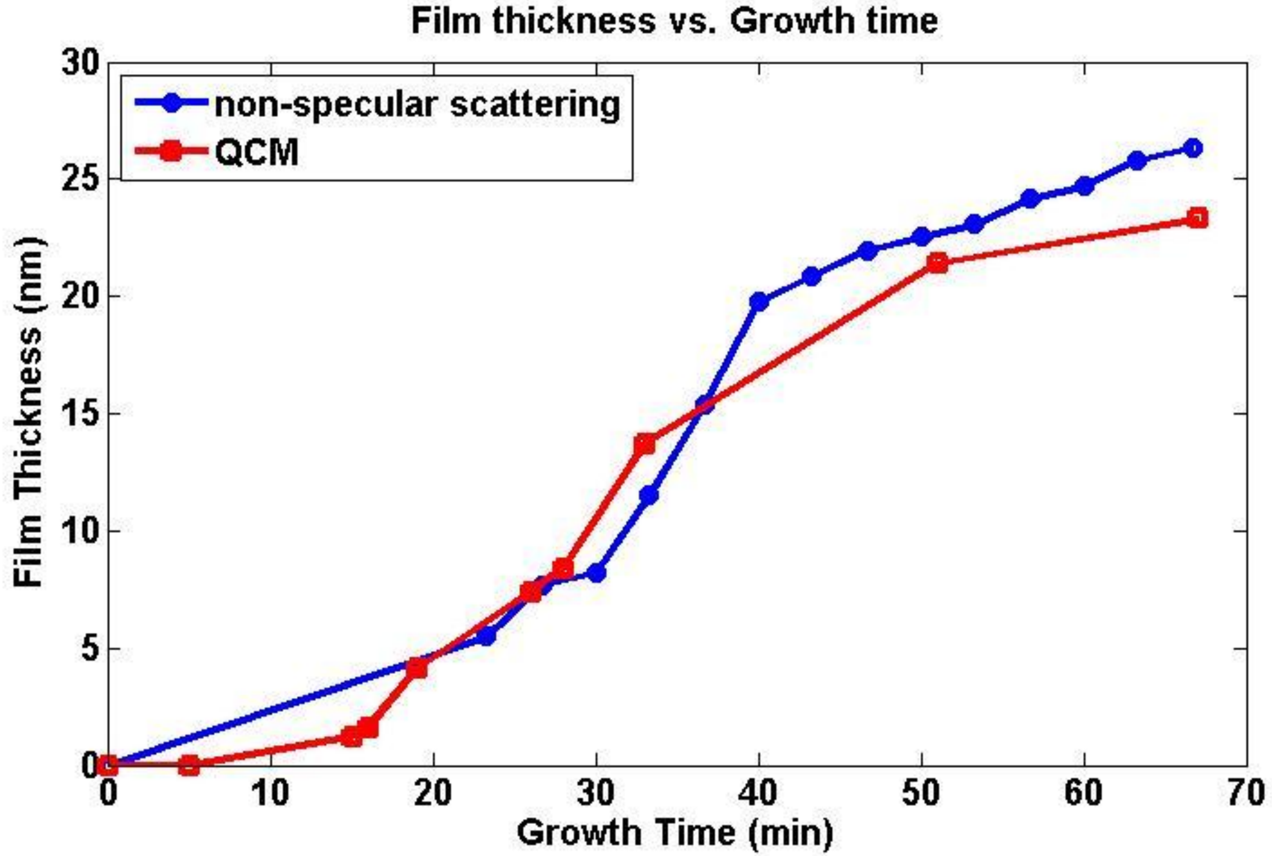
Local average (Green curve) is defined as:

$$\log[R_F(q_z)/\rho_s^2] \approx \frac{1}{N}\sum_{q_z=q_{z1}}^{q_{z2}} \log[R(q_Z)]$$

$$\Delta q_z = q_{z2} - q_{z1} > \text{oscillation period}$$

Sample: spec_start_S144

Sb film only deposited on Si (100)



Two methods get the similar result for Sb deposition on Si (100).