Surface Sensitive X-ray Scattering

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Introduction
- Concepts of surfaces
- Scattering (Born approximation)

Crystal Truncation Rods
- The basic idea
- How to calculate
- Examples

Reflectivity
- In Born approximation
- Exact formalism (Fresnel)
- Examples

Grazing Incidence Diffraction
- The basic idea
- Penetration depth
- Example

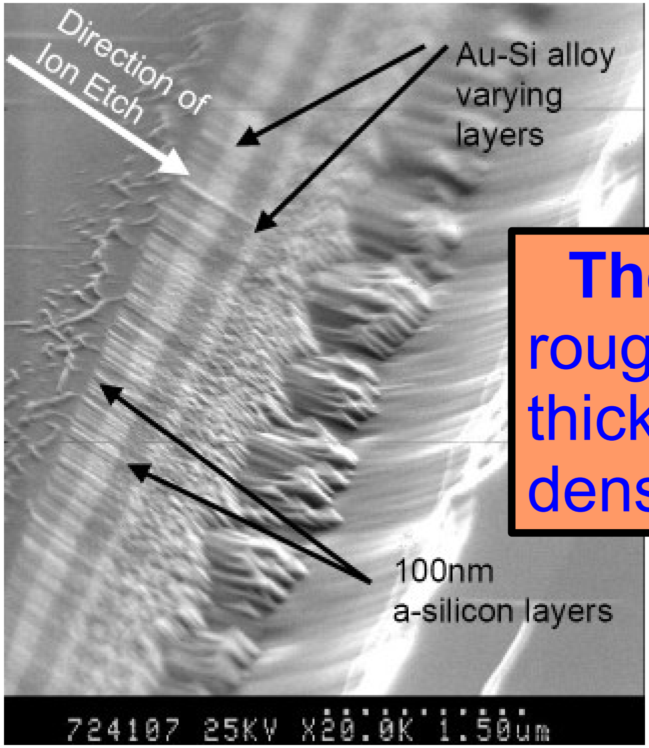
With x-ray and neutron reflectivity surfaces, buried interfaces and the properties of thin film systems can be investigated on a micro- and nanoscale.

Fundamental science, e.g.:

- layer growth
- roughness evolution

Industrial applications, e.g.:

- semiconductor devices
- storage devices / harddisks
- coatings
- lubricants
- catalysts



The layers'
roughnesses ?
thicknesses ?
densities ?

Advantages of x-ray and neutron reflectometry:

- Resolution in the Å-regime
- Gives a lot of information with just one measurement
- Usually non-destructive
- Highly element specific
- No special preparation of the sample
- (Averaged information over whole sample area)

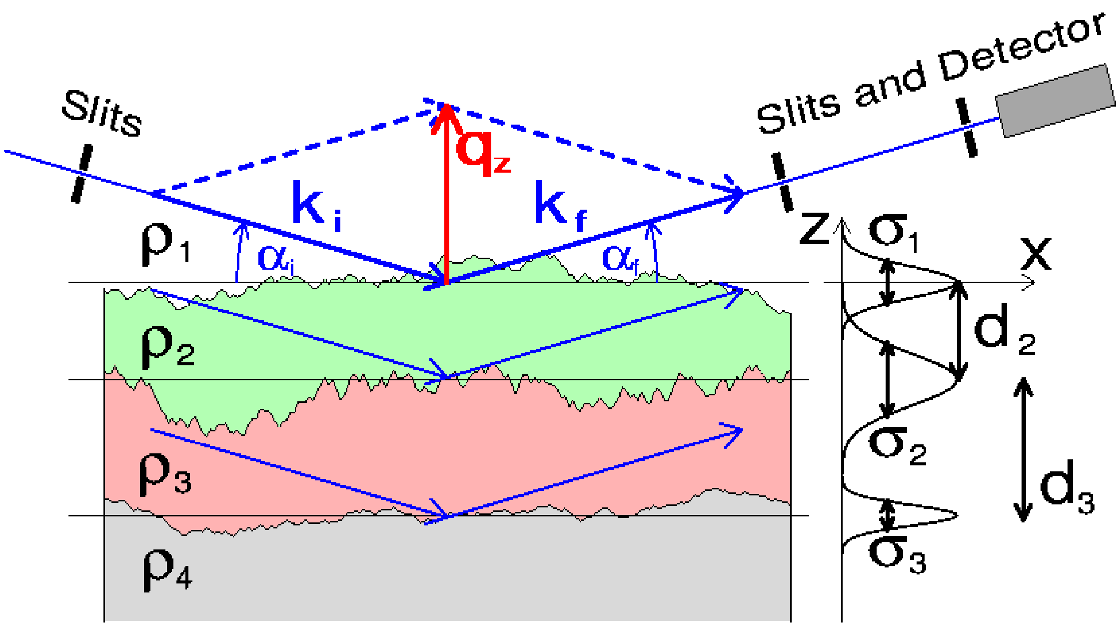
Disadvantages of x-ray and neutron reflectometry:

- No unique results without preknowledge
- No fast results
- Interpretation/analysis often not easy
- (No local information)

Theoretical Part

a) General Considerations

Photons with wavelength λ (or neutrons with $\lambda = h/\sqrt{2mE}$)
are scattered elastically (no energy change: $\lambda_i = \lambda_f$) at the surface.
The incident angle α_i equals the exit angle α_f .

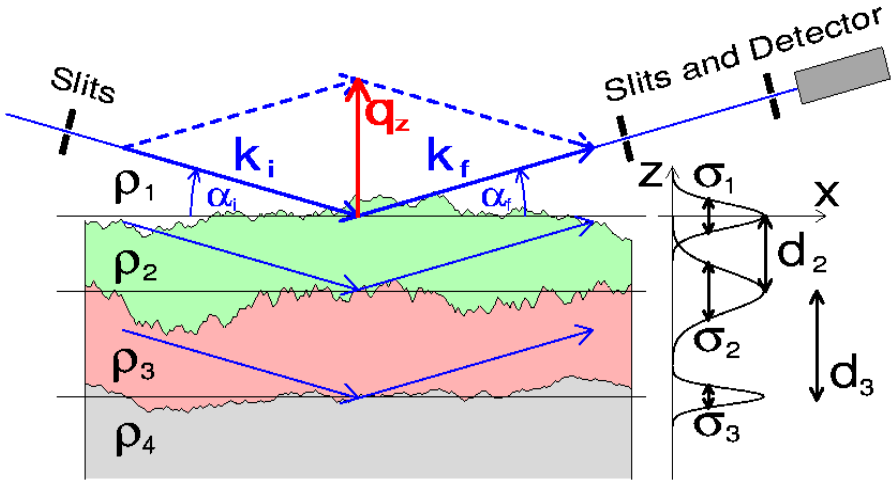


The density ρ_j means:

- Electron density for x-rays
- Scattering length density for neutrons

Wave vector transfer

$$q_z = \frac{4\pi}{\lambda}\sin(\alpha_f) = 2k_0 \sin(\alpha_f)$$

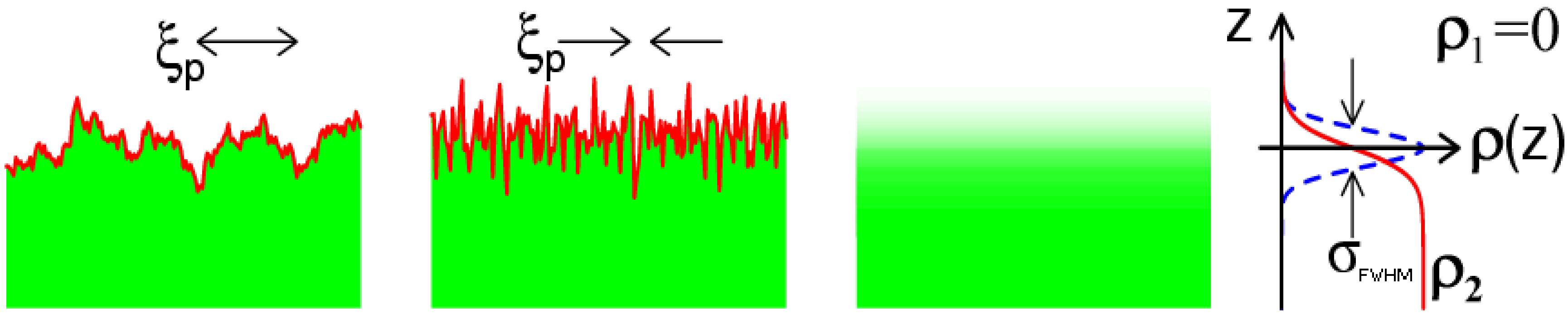


q_z **is perpendicular to the surface**

$\Rightarrow$

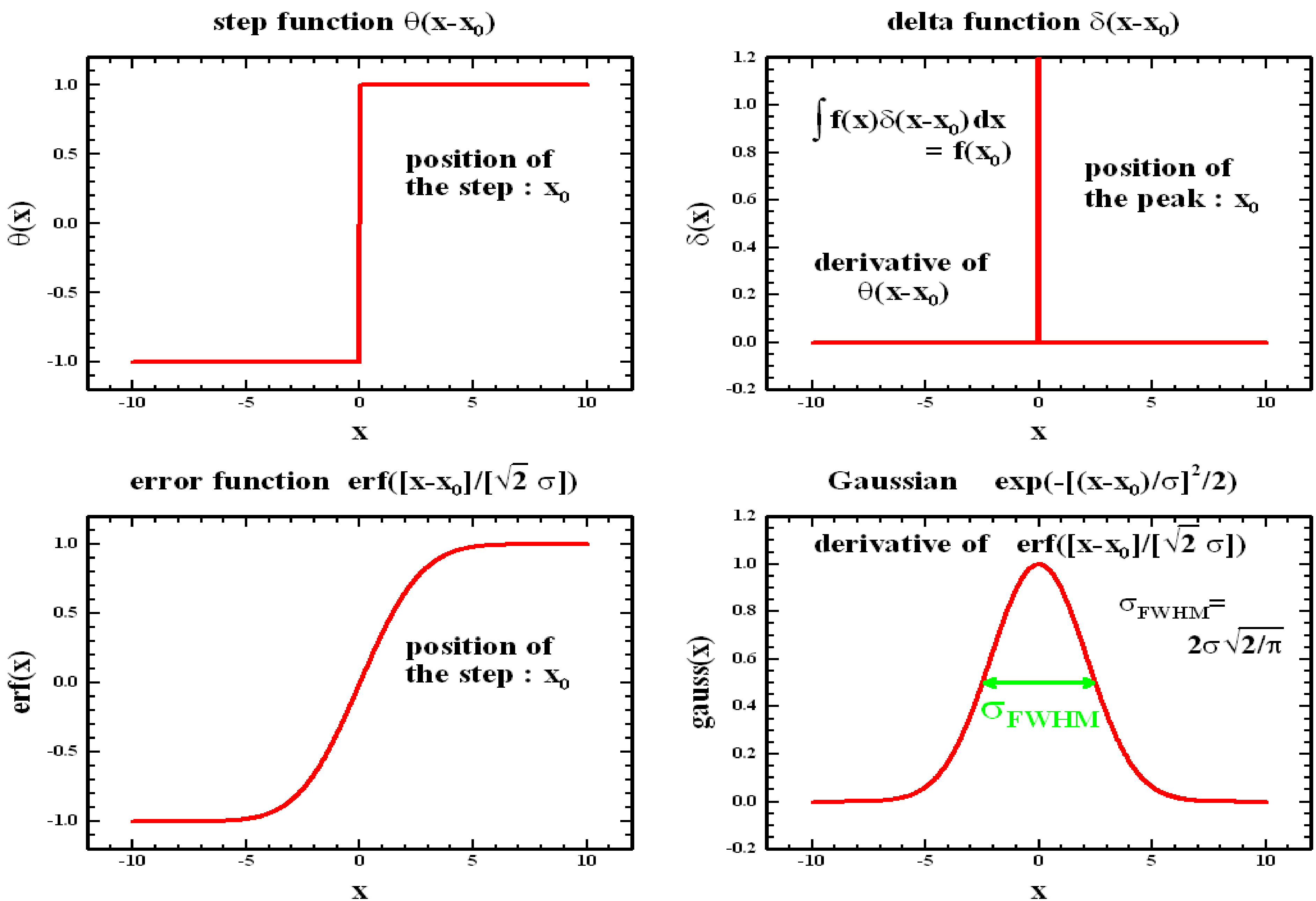
only sensitive to information perpendicular to the surface : electron (scattering length) density profile $\langle \rho(x,y,z) \rangle_{(x,y)} = \rho(z)$.

That means: a reflectivity cannot distinguish different in-plane structures.



These different surfaces have the same reflectivity !

The following functions are important in the following:



Specularly Reflected Intensity in Born Approximation ($I_{scatt} << I_0$)

$$I(q_z) \propto \frac{1}{q_z^4} \left| \int \frac{d\rho(z)}{dz} \exp(iq_z z)\, dz \right|^2$$

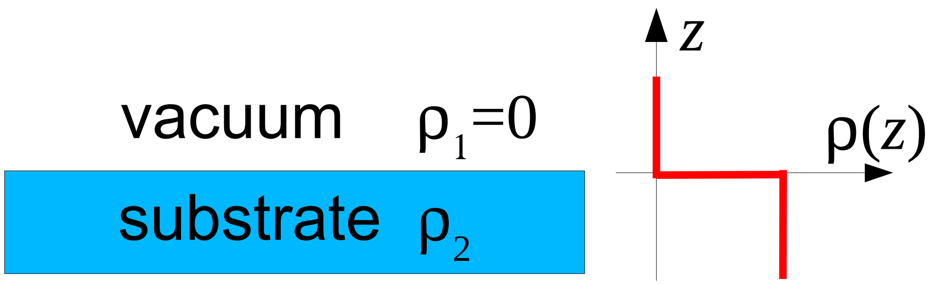
Given by the absolute square of the Fouriertransformation of the derivative of the density/(scattering length) profile and divided by q_z^4.

Consequences:

- Reflected intensity drops fast with increasing angle : $1/q_z^4$
- Only differences in density can be seen (contrast) : Derivative
- Only sensitive to density properties in z-direction : Density profile
- No direct picture visible : Fourier space
- Phase information gets lost $\Rightarrow$ no unique solution : Absolute square

Examples

1) single smooth surface at $z = 0$

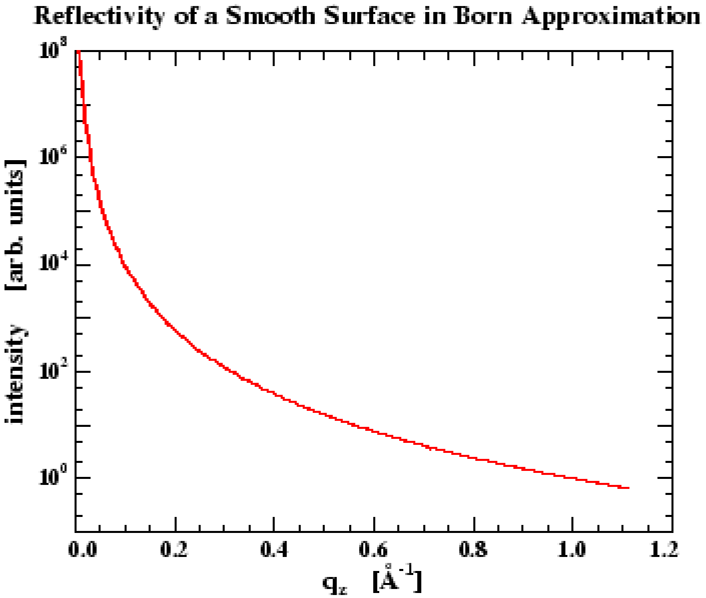


Density profile: $\rho(z)=\frac{\rho_2}{2}(1-\Theta[z]) \Rightarrow \frac{d\rho}{dz}\propto\delta(z)$

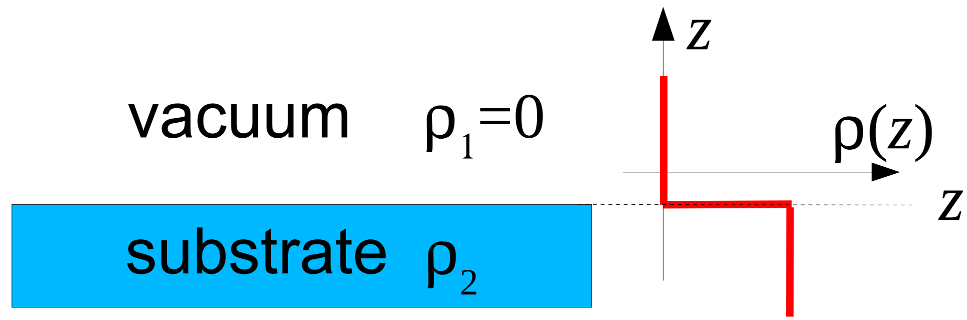
$$I(q_z)\propto\frac{1}{q_z^4}\left|\int\frac{d\rho(z)}{dz}\exp(iq_z z)\,dz\right|^2$$

$$=\frac{1}{q_z^4}\left|\int\delta(z)\exp(iq_z z)\,dz\right|^2$$

$$=\frac{1}{q_z^4}\left|\exp(iq_z\cdot 0)\right|^2=\frac{1}{q_z^4}\cdot|1|^2=\frac{1}{q_z^4}$$



2) single smooth surface at $z = z_1$ (shifted)



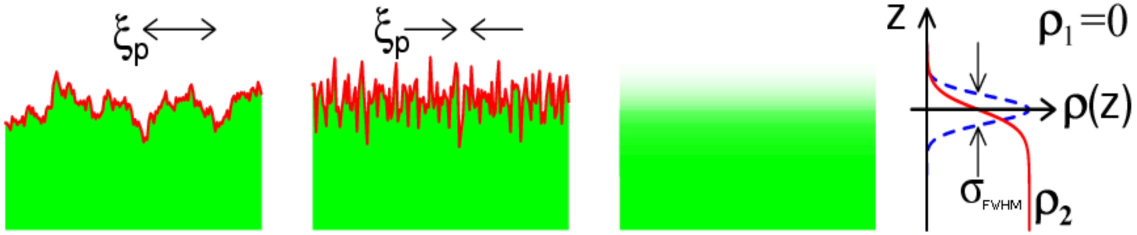
Density profile: $$\rho(z)=\frac{\rho_2}{2}\left(1-\Theta[z-z_1]\right) \quad \Rightarrow \quad \frac{d\rho}{dz}\propto\delta(z-z_1)$$

$$I(q_z)\propto\frac{1}{q_z^4}\left|\int\frac{d\rho(z)}{dz}\exp(iq_z z)\,dz\right|^2=\frac{1}{q_z^4}\left|\int\delta(z-z_1)\exp(iq_z z)\,dz\right|^2$$

$$=\frac{1}{q_z^4}\left|\exp(iq_z z_1)\right|^2=\frac{1}{q_z^4}\cdot 1^2=\frac{1}{q_z^4}$$

A shift of the sample does not change the reflectivity.

3) single rough surface with roughness σ



Density profile:

$$\rho(z) = \frac{\rho_2}{2}\left[1 - \mathrm{erf}\left(\frac{z}{\sqrt{2}\,\sigma}\right)\right] \Rightarrow \frac{d\rho}{dz} \propto \exp\left(\frac{-z^2}{2\sigma^2}\right)$$

$$I(q_z) \propto \frac{1}{q_z^4}\left|\int \frac{d\rho(z)}{dz}\exp(iq_z z)\,dz\right|^2$$

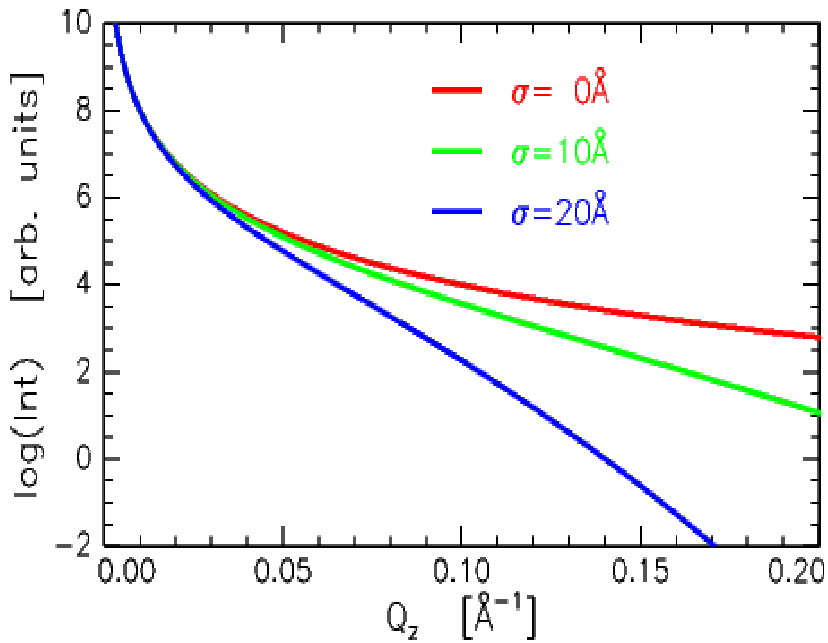
$$= \frac{1}{q_z^4}\left|\int \exp\left(\frac{-z^2}{2\sigma^2}\right)\exp(iq_z z)\,dz\right|^2$$

Fourier transformation is known!

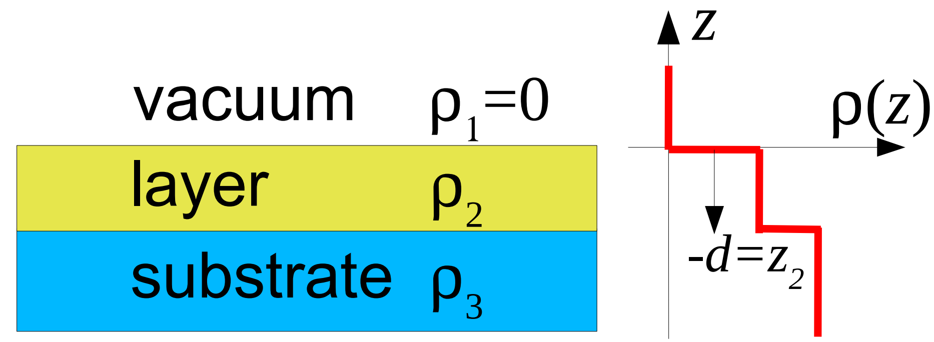
$$\propto \frac{1}{q_z^4}\left|\exp\left(\frac{-q_z^2\sigma^2}{2}\right)\right|^2 = \frac{1}{q_z^4}\exp(-q_z^2\sigma^2)$$

Debye-Waller factor

Effect of the roughness



4) single smooth layer with thickness d



Density profile:

$$\rho(z)=\frac{\Delta\rho_1}{2}[1-\Theta(z)]+\frac{\Delta\rho_2}{2}[1-\Theta(z+d)]$$

Derivative of $\rho(z)$:

$$\frac{d\rho}{dz}\propto\Delta\rho_1\,\delta(z)+\Delta\rho_2\cdot\delta(z+d)$$

with: $\Delta\rho_1=\rho_2-\rho_1$

$\Delta\rho_2=\rho_3-\rho_2$

$$I(q_z)\propto\frac{1}{q_z^4}\left|\int\frac{d\rho(z)}{dz}\exp(iq_z z)\,dz\right|^2=\frac{1}{q_z^4}\left|\int[\Delta\rho_1\delta(z)+\Delta\rho_2\delta(z+d)]\exp(iq_z z)\,dz\right|^2$$

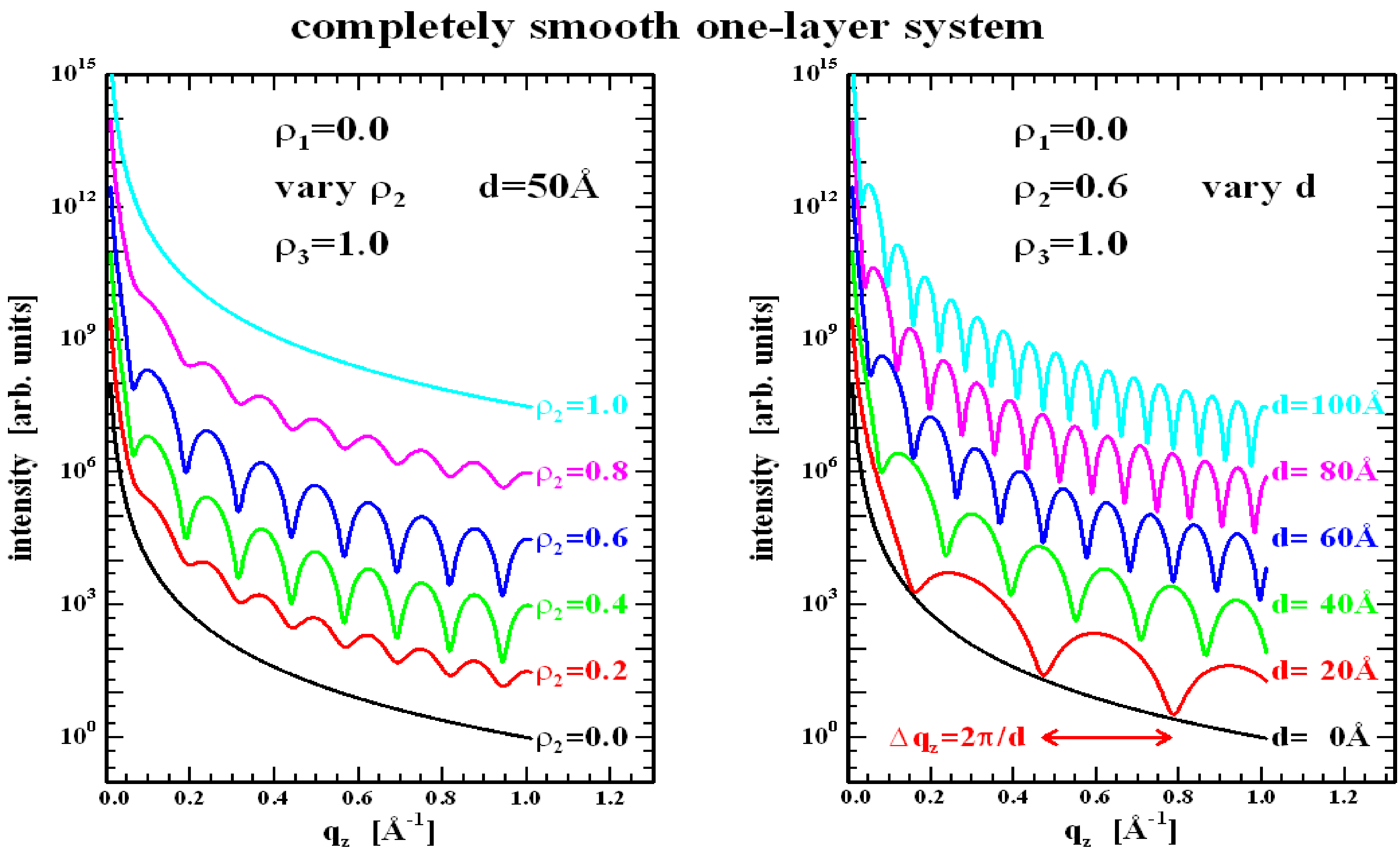
$$=\frac{1}{q_z^4}\left|\Delta\rho_1+\Delta\rho_2\exp(-iq_z d)\right|^2=\frac{1}{q_z^4}[\Delta\rho_1+\Delta\rho_2\exp(iq_z d)]\cdot[\Delta\rho_1+\Delta\rho_2\exp(-iq_z d)]$$

$$=\frac{1}{q_z^4}(\Delta\rho_1^2+\Delta\rho_2^2+\Delta\rho_1\Delta\rho_2[\exp(iq_z d)+\exp(-iq_z d)])$$

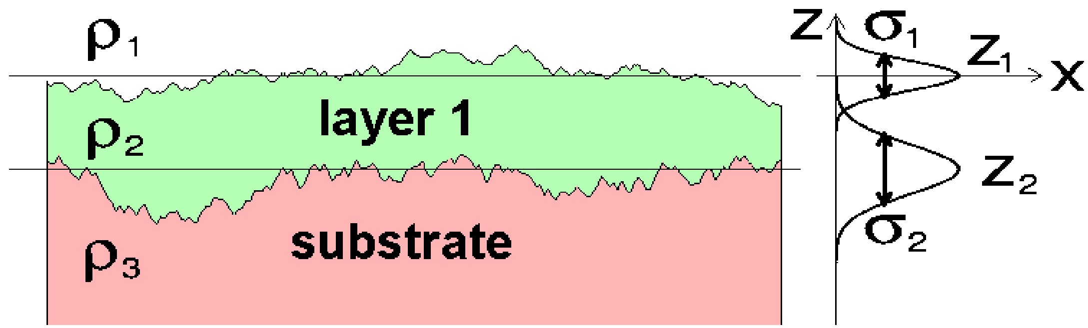
$$=\frac{1}{q_z^4}[\Delta\rho_1^2+\Delta\rho_2^2+2\Delta\rho_1\Delta\rho_2\cos(q_z d)]$$

oscillating function

- Contrasts $\Delta\rho_1$ and $\Delta\rho_2$ determine the visibility of the oscillations.
- Film thickness d determines the period via $\Delta q_z = 2\pi/d$.



5) single layer with rough interfaces and thickness $d = -z_2$



Density profile:

$$\rho(z) = \frac{\Delta\rho_1}{2}\left[1 - \mathrm{erf}\left(\frac{z - z_1}{\sqrt{2}\,\sigma_1}\right)\right] + \frac{\Delta\rho_2}{2}\left[1 - \mathrm{erf}\left(\frac{z - z_2}{\sqrt{2}\,\sigma_2}\right)\right]$$

Derivative of $\rho(z)$:

$$\frac{d\rho}{dz} \propto \frac{\Delta\rho_1}{\sigma_1}\exp\left(\frac{-(z - z_1)^2}{2\sigma_1^2}\right) + \frac{\Delta\rho_2}{\sigma_2}\exp\left(\frac{-(z - z_2)^2}{2\sigma_2^2}\right)$$

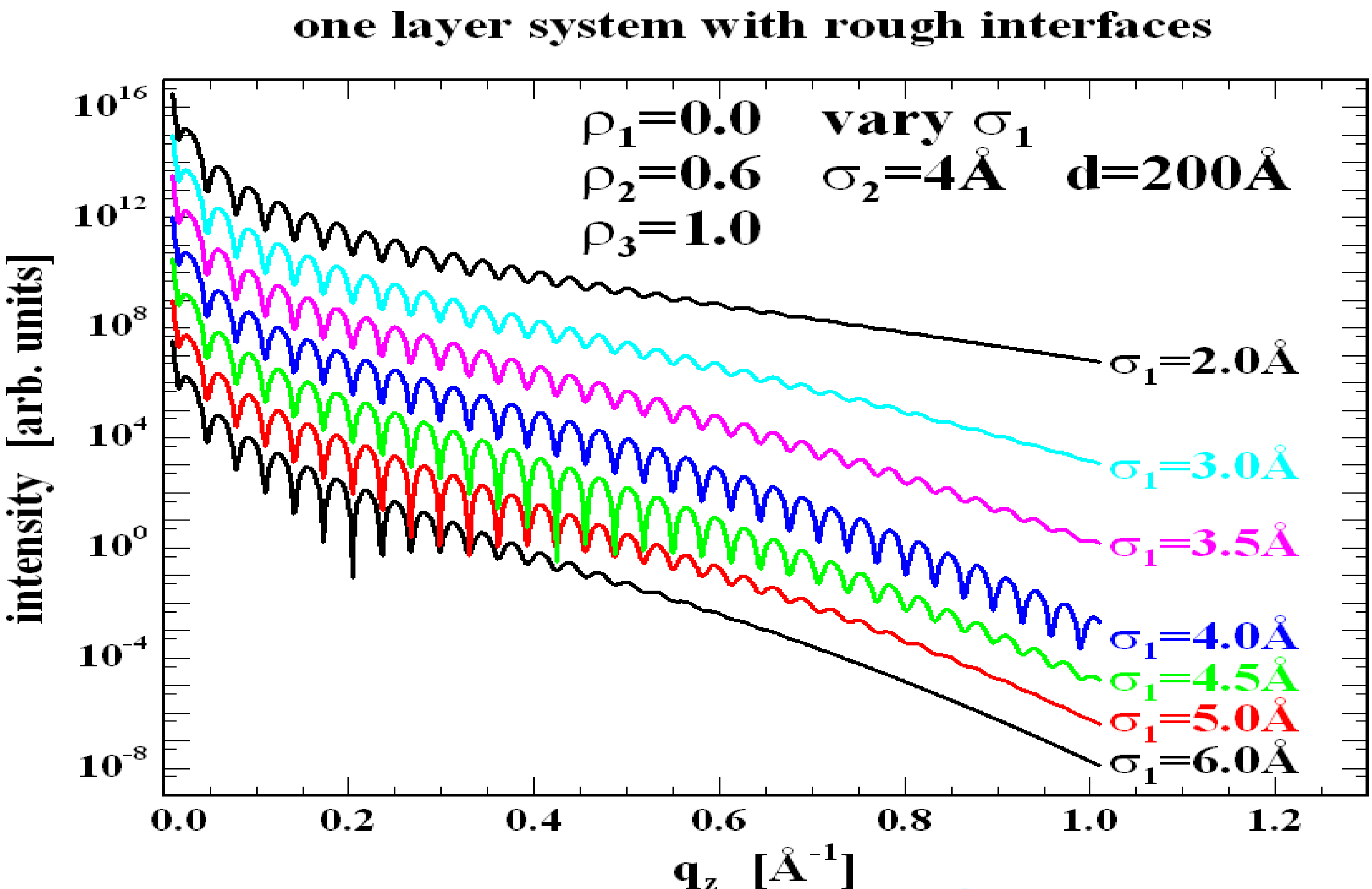
using :

$$\int \exp\left(\frac{-(z - z_1)^2}{2\sigma_1^2}\right)\exp(iq_z z)\,dz = \exp(iq_z z_1)\sqrt{2}\,\sigma_1 \exp\left(\frac{q_z^2\sigma_1^2}{2}\right)$$

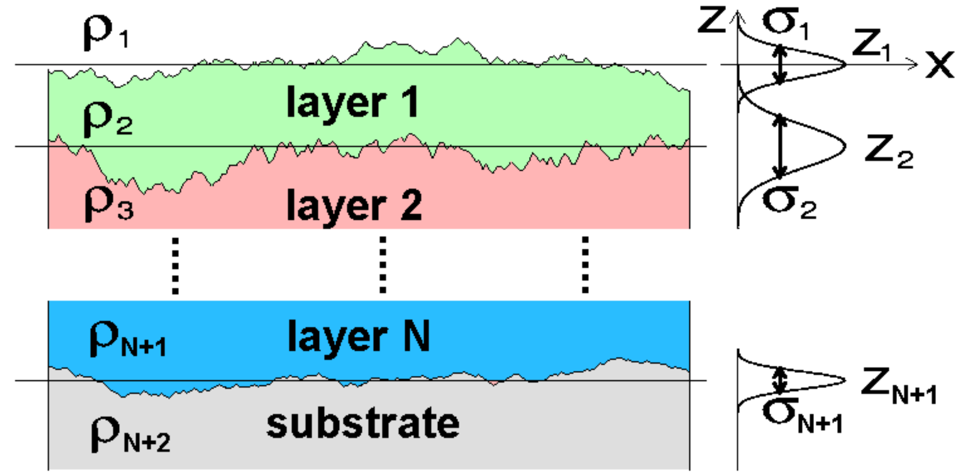
Result :

$$I(q_z) \propto \frac{1}{q_z^4}\left[\Delta\rho_1^2 \exp(-q_z^2\sigma_1^2) + \Delta\rho_2^2 \exp(-q_z^2\sigma_2^2) + 2\Delta\rho_1\Delta\rho_2 \exp\left(-q_z^2\frac{\sigma_1^2 + \sigma_2^2}{2}\right)\cos(q_z z_2)\right]$$

- At large q_z the scattering is dominated by the smoothest interface.
- The difference between the σ's of a layer determines the “die-out” of the oscillations.



5) general case: N rough layers



Density profile: $$\rho(z) = \frac{1}{2}\sum_{j=1}^{N+1} \Delta\rho_j \left(1 - \mathrm{erf}\left[\frac{z - z_j}{\sqrt{2}\,\sigma_j}\right]\right) \quad \text{with} \quad \Delta\rho_j = \rho_{j+1} - \rho_j$$

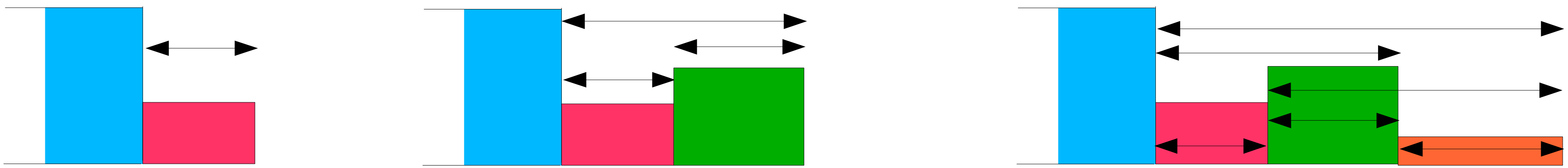
$$I(q_z) \propto \frac{1}{q_z^4}\left(\sum_{j=1}^{N+1} \Delta\rho_j^2 \exp(-q_z^2\sigma_j^2) + 2\sum_{j=1}^{N}\sum_{k=j+1}^{N+1} \Delta\rho_j \Delta\rho_k \exp\left(-q_z^2 \frac{\sigma_j^2 + \sigma_k^2}{2}\right) \cos[q_z(z_j - z_k)]\right)$$

Scattering terms from the single interfaces

Each distance z_j-z_k gives an oscillating term, scaled with the respective Debye-Waller factor and the contrasts at the interfaces.

For a first guess on reflectivity data: Fourier backtransformation of $q_z^4 \cdot I(q_z)$ will show distinct peaks for each oscillation ($\Leftrightarrow$ distance).

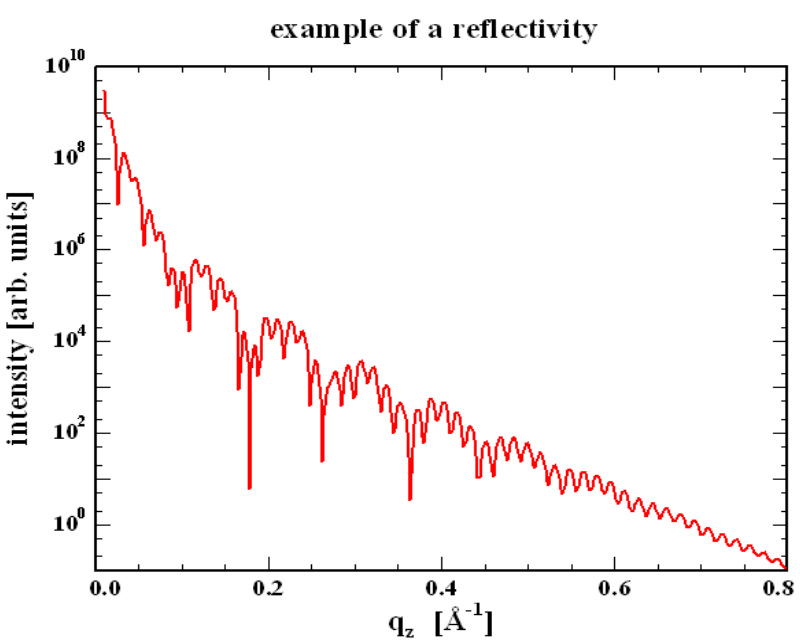
Maximum number of distances



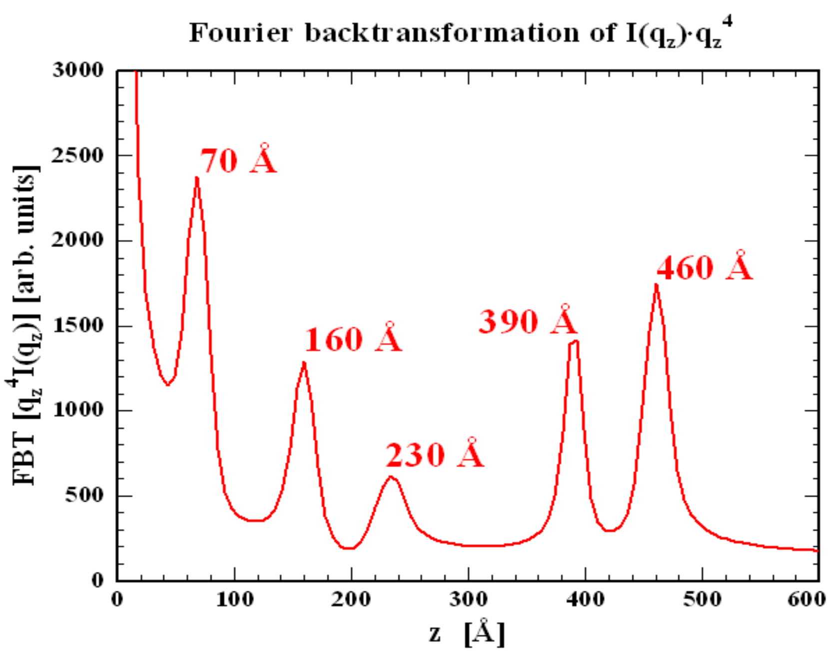
1 layer : 1

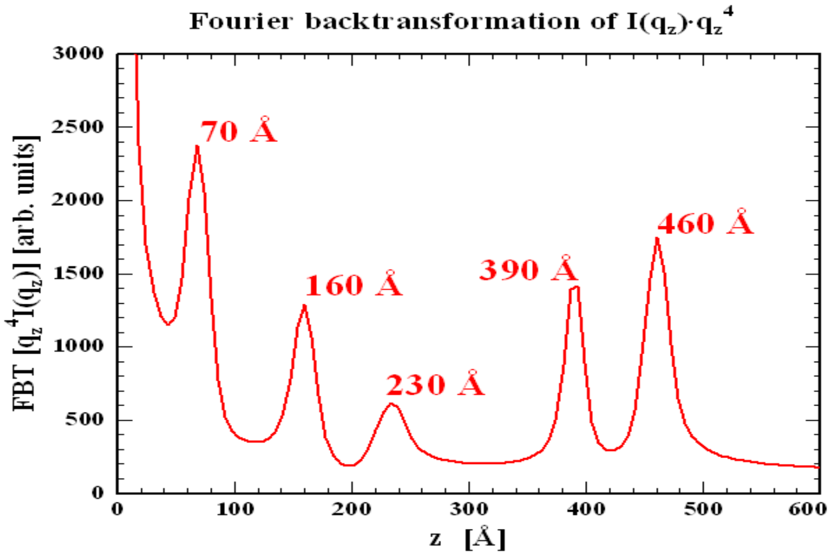
2 layers : 3

3 layers : 6



FTB →

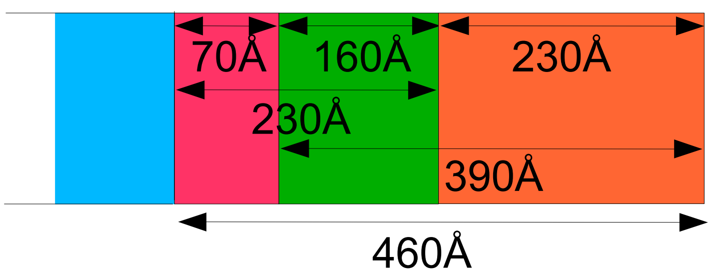




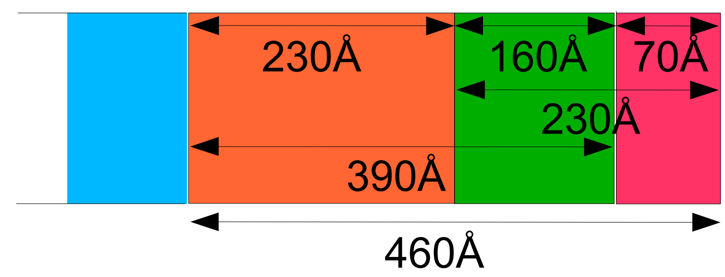
Only 5 peaks !

Likely a 3-layer system with one layer thickness matching the sum of two neighboring layers.

Two possibilities:



or



Result of swapping layers