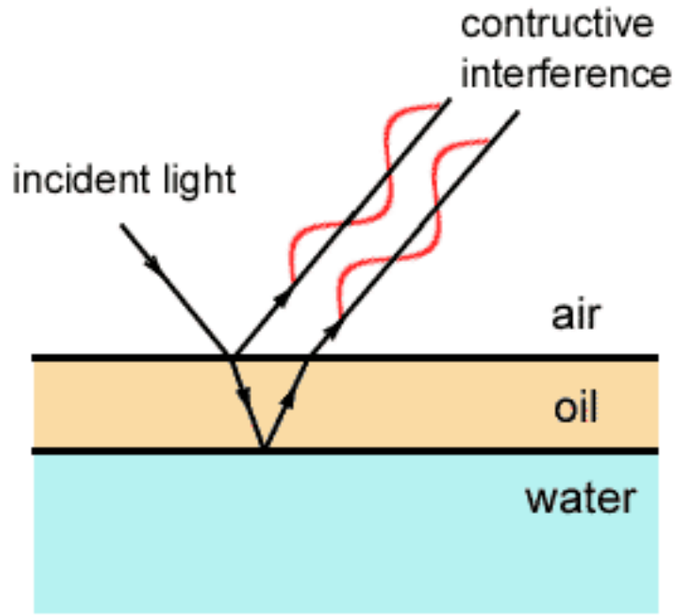
Oil spill in front of the reactor building..



Light and neutrons

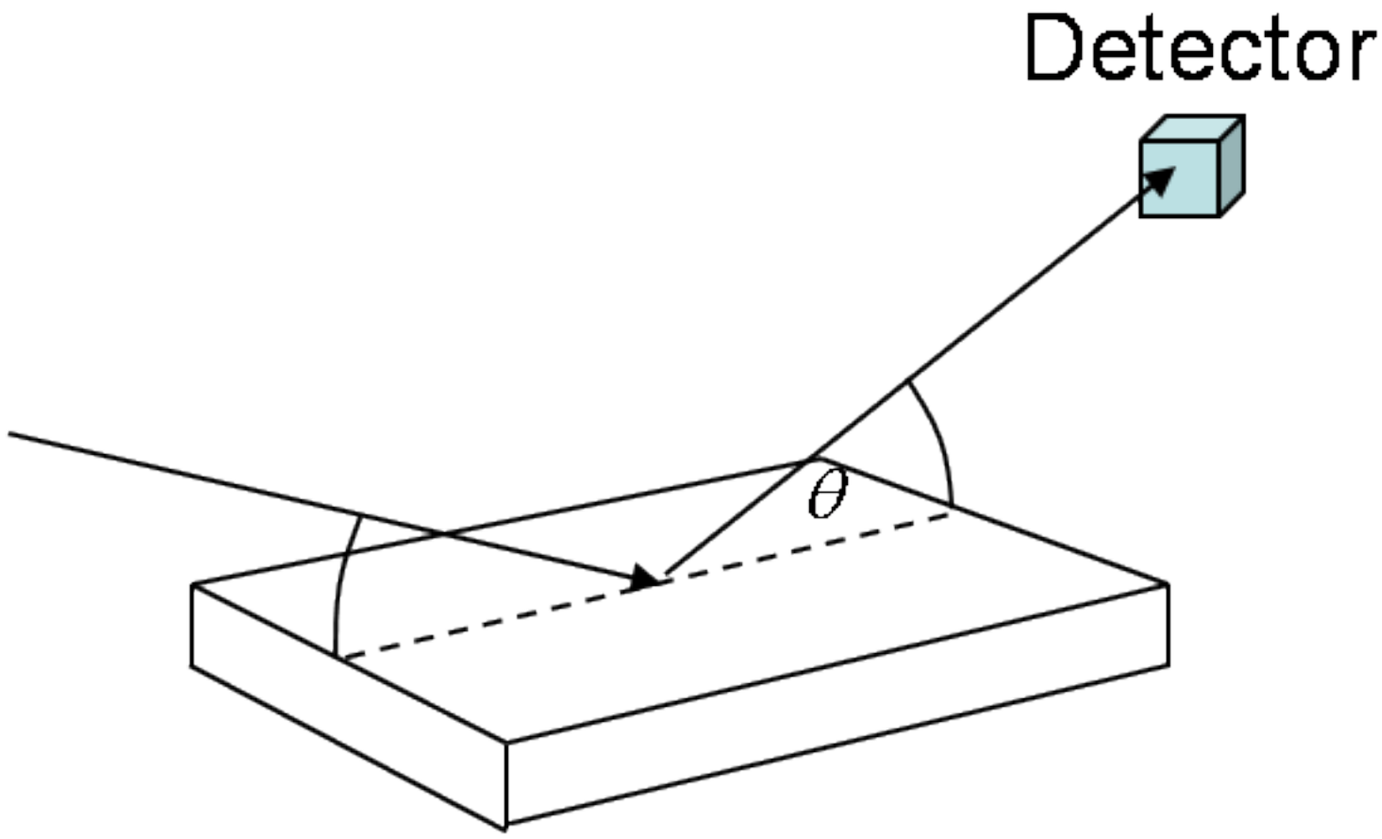
- Neutrons reflect from mirrors in a similar way as light.
- Manifestation of neutron's wave-particle nature!
- Light "sees" electrons while neutrons "see" nuclei

The underlying reason for these commonalities

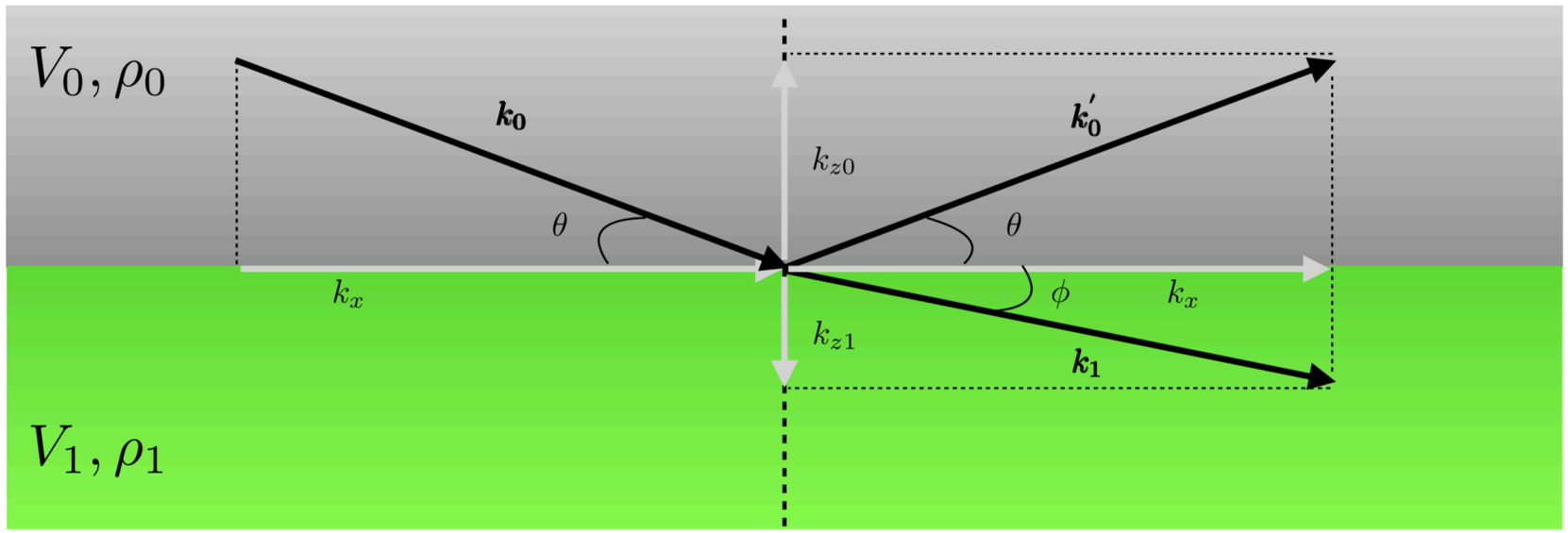
$$(\vec{\nabla}^2 + k^2)A = 0$$

both types of radiation their propagation obey a second-order partial differential equation of the same form (Helmholtz equation)

Neutron reflection at a planar surface



Neutron reflection at a planar surface



$$V = \frac{2\pi\hbar^2}{m}\rho$$

$$\rho = \sum_j N_j b_j$$

elastic specular reflection $|\vec{k_0}| = |\vec{k_0'}| = k = 2\pi/\lambda$

$$E = \hbar^2 k^2 / 2m$$

wave-vector transfer: $Q = |\vec{Q}| = |\vec{k_0}' - \vec{k_0}| = 2k\sin\theta = 2k_z$

Starting point for calculations…

time independent Schröndinger equation for a particle in a constant potential

$$\frac{\hbar^2}{2m}\vec{\nabla}^2\psi(\vec{r}) + (E - V)\,\psi(\vec{r}) = 0$$

the interface is homogeneous in the xy-plane and reflection or refraction may take place only in the z-direction, thus problem reduced to one dimension

$$\frac{\hbar^2}{2m}\frac{d^2}{dz^2}\psi(z) + (E_z - V)\,\psi(z) = 0$$

$$E_{z0} = \frac{\hbar^2 k_0^2 \sin^2\theta}{2m}$$

Neutron energy vs potential…

at small angles $V - E_z > 0$

large angles $V - E_z < 0$

for a critical angle we have $V - E_z = 0$

using $V = \frac{2\pi\hbar^2}{m}\rho$ and $E_{z0} = \frac{\hbar^2 k_0^2 \sin^2\theta}{2m}$ we get

$$\cos\theta_c = \sqrt{1 - \frac{\lambda^2}{\pi}\rho} \qquad \text{or} \qquad \theta_c \approx \lambda\sqrt{\frac{\rho}{\pi}}$$

We solve the wave equation for the two cases…

for $E_z < V_1 (\theta < \theta_c)$

$$\frac{\hbar^2}{2m}\frac{d^2}{dz^2}\psi_0(z) + E_{z0}\psi_0(z) = 0 \qquad \text{upper medium}$$

$$-\frac{\hbar^2}{2m}\frac{d^2}{dz^2}\psi_1(z) + \kappa^2\psi_1(z) = 0 \qquad \text{lower medium}$$

with $\kappa^2 = \frac{2m}{\hbar^2}(V_1 - E_{z0})$

solutions:

$$\psi_0(z) = e^{ik_{z0}z} + re^{-k_{z0}z}$$

$$\psi_1(z) = te^{-\kappa z}$$

Boundary conditions…

wave function continuity: $\psi_0(0) = \psi_1(0)$

momentum conservation: $\dfrac{d\psi_0(0)}{dz} = \dfrac{d\psi_1(0)}{dz}$

$$\left.\begin{aligned} 1 + r &= t \\ ik_{z0}(1 - r) &= -\kappa t \end{aligned}\right\}$$

so $r = \dfrac{k_{z0} - i\kappa}{k_{z0} + i\kappa}$ and $R = |r|^2 = \dfrac{|k_{z0} - i\kappa|^2}{|k_{z0} + i\kappa|^2} = 1$

total reflection!

also note: $|\psi_1(z)|^2 = |t|^2 e^{-2\kappa z}$ evanescent wave

Second case…

for $E_z > V_1(\theta > \theta_c)$

$$\frac{\hbar^2}{2m}\frac{d^2}{dz^2}\psi_{z0}(z) + \left(\frac{\hbar^2 k_{z0}^2}{2m}\right)\psi_{z0}(z) = 0 \qquad \text{upper medium}$$

$$\frac{\hbar^2}{2m}\frac{d^2}{dz^2}\psi_{z1}(z) + \left(\frac{\hbar^2 k_{z0}^2}{2m} - \frac{2\pi\hbar^2}{m}\rho\right)\psi_{z1}(z) = 0 \qquad \text{lower medium}$$

$$\left[\frac{d^2}{dz^2} + k_{z0}^2\right]\psi_0(z) = 0$$

or $$\left[\frac{d^2}{dz^2} + \left(k_{z0}^2 - 4\pi\rho\right)\right]\psi_1(z) = \left[\frac{d^2}{dz^2} + k_{z1}^2\right]\psi_1(z) = 0$$

with $$k_{z1}^2 = k_{z0}^2 - 4\pi\rho = k_{z0}^2\left[1 - (\lambda^2/\pi)\rho\right]$$

solutions of wave equation…

$$\psi_0(z) = e^{ik_{z0}z} + re^{-ik_{z0}z}$$
$$\psi_1(z) = te^{ik_{z1}z}$$

plane waves moving in and out of the interface…

and by applying the same boundary conditions as previously we get

$$\left.\begin{aligned} 1 + r &= t \\ k_{z0}(1 - r) &= k_{z1}t \end{aligned}\right\} \rightarrow \left.\begin{aligned} r &= \frac{k_{z0} - k_{z1}}{k_{z0} + k_{z1}} \\ t &= \frac{2k_{z0}}{k_{z0} + k_{z1}} \end{aligned}\right\} \rightarrow \left.\begin{aligned} R &= |r|^2 = \left|\frac{k_{z0} - k_{z1}}{k_{z0} + k_{z1}}\right|^2 \\ T &= |t|^2 = \left|\frac{2k_{z0}}{k_{z0} + k_{z1}}\right|^2 \end{aligned}\right.$$

we can work the last result a bit more…

$$k_{z0,c} = \frac{2\pi}{\lambda}\sin\theta_c$$

$$k_{z0,c}^2 = \frac{4\pi^2}{\lambda^2}\sin^2\theta_c$$

remembering that $\cos\theta_c = \sqrt{1 - \frac{\lambda^2}{\pi}\rho}$

$$k_{z0,c}^2 = \frac{4\pi^2}{\lambda^2}(1 - \cos^2\theta_c)$$

we get $k_{z0,c}^2 = 4\pi\rho$ and together with $k_{z1}^2 = k_{z0}^2 - 4\pi\rho$

we obtain $k_{z1}^2 = k_{z0}^2 - k_{z0,c}^2$

$$R = |r|^2 = \left|\frac{k_{z0} - k_{z1}}{k_{z0} + k_{z1}}\right|^2 \Bigg\} \rightarrow R = \left|\frac{k_{z0} - \sqrt{k_{z0}^2 - k_{z0,c}^2}}{k_{z0} - \sqrt{k_{z0}^2 - k_{z0,c}^2}}\right|^2$$

and finally we get…

since $Q = 2k_{z0}$ → $R = \left| \frac{1 - \sqrt{1 - (Q_c/Q)^2}}{1 + \sqrt{1 - (Q_c/Q)^2}} \right|^2$ exact expression

using the Taylor series $\sqrt{1 - (Q_c/Q)^2} = 1 - \frac{1}{2}\left(\frac{Q_c}{Q}\right)^2 + ...$

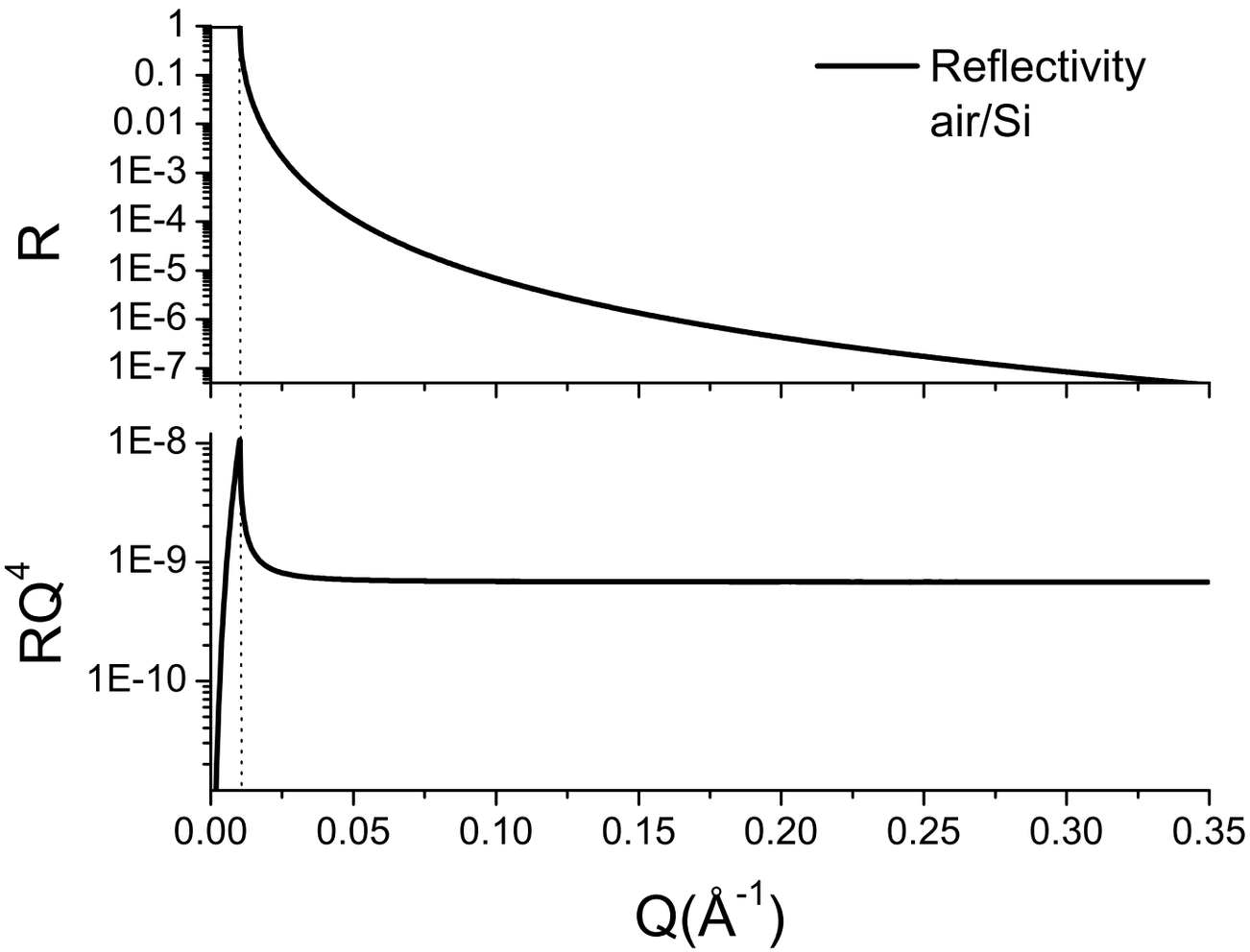
for $Q >> Q_c$ $R \approx \frac{1}{16}\frac{Q_c^4}{Q^4}$

Fresnel reflectivity

Summing up: neutron reflection at an interface

$$Q < Q_c \longrightarrow R = 1$$

$$Q >> Q_c \longrightarrow R \approx \frac{1}{16}\frac{Q_c^4}{Q^4}$$



Another approach: Born approximation

Lippmann-Schwinger integral equation for a scattering process

$$\psi(\vec{r}) = e^{ik\vec{r}} - \int \frac{e^{ik|\vec{r}-\vec{r'}|}}{|\vec{r}-\vec{r'}|}\rho(\vec{r'})(e^{i\vec{k}\vec{r'}} + \psi_s(\vec{r'}))d^3\vec{r'}$$

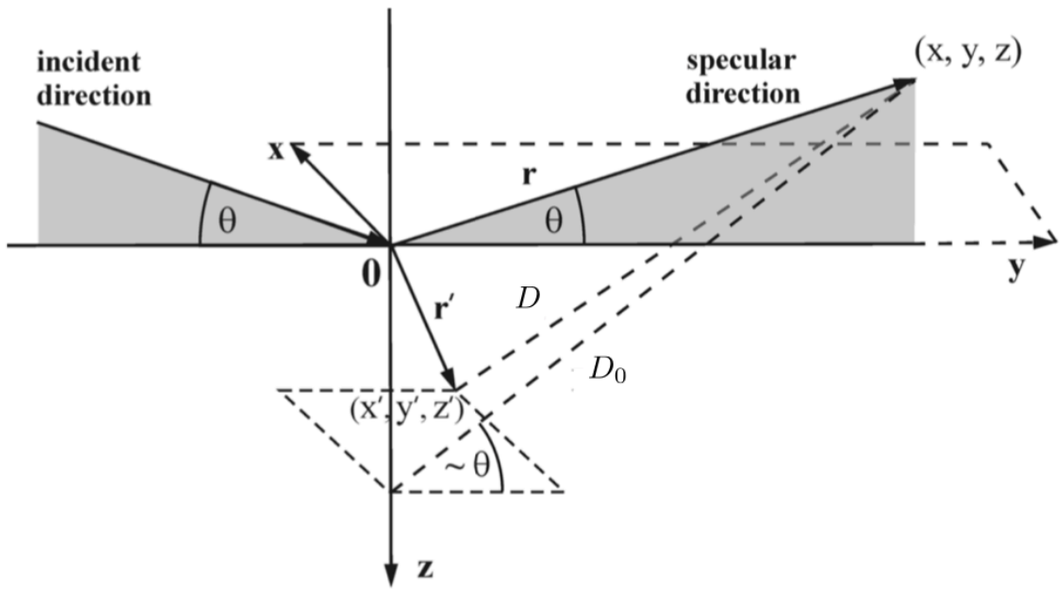
$$\psi_s(\vec{r}) = -\int \frac{e^{ik|\vec{r}-\vec{r'}|}}{|\vec{r}-\vec{r'}|}\rho(\vec{r'})(e^{i\vec{k}\vec{r'}} + \psi_s(\vec{r'}))d^3\vec{r'} = \quad \text{Born approximation}$$

$$= -\int \frac{e^{ik|\vec{r}-\vec{r'}|}}{|\vec{r}-\vec{r'}|}\rho(\vec{r'})e^{i\vec{k}\vec{r'}}\, d^3\vec{r'} - \int \frac{e^{ik|\vec{r}-\vec{r'}|}}{|\vec{r}-\vec{r'}|}\rho(\vec{r'})\psi_s(\vec{r'})d^3\vec{r'}$$

Another approach: Born approximation

$$\psi_s(\vec{r}) = -\int \frac{e^{ik|\vec{r}-\vec{r'}|}}{|\vec{r}-\vec{r'}|}\rho(\vec{r'})e^{i\vec{k}\vec{r'}}\,d^3\vec{r'}$$

For the reflection geometry we get…



$$r = -\frac{4\pi i}{Q}\int_{-\infty}^{+\infty} dz'\,\rho(z')e^{iQz'}$$

$$R = |r|^2 = \frac{16\pi^2}{Q^2}\left|\int_{-\infty}^{+\infty} dz'\,\rho(z')e^{iQz'}\right|^2$$

$$R = \frac{16\pi^2}{Q^4}\left|\int_{-\infty}^{+\infty} \frac{d\rho(z')}{dz'}e^{iQz'}\,dz'\right|^2$$

Reflectivity related to the Fourier transform of the sld profile!

Born approximation: simple interface reflectivity

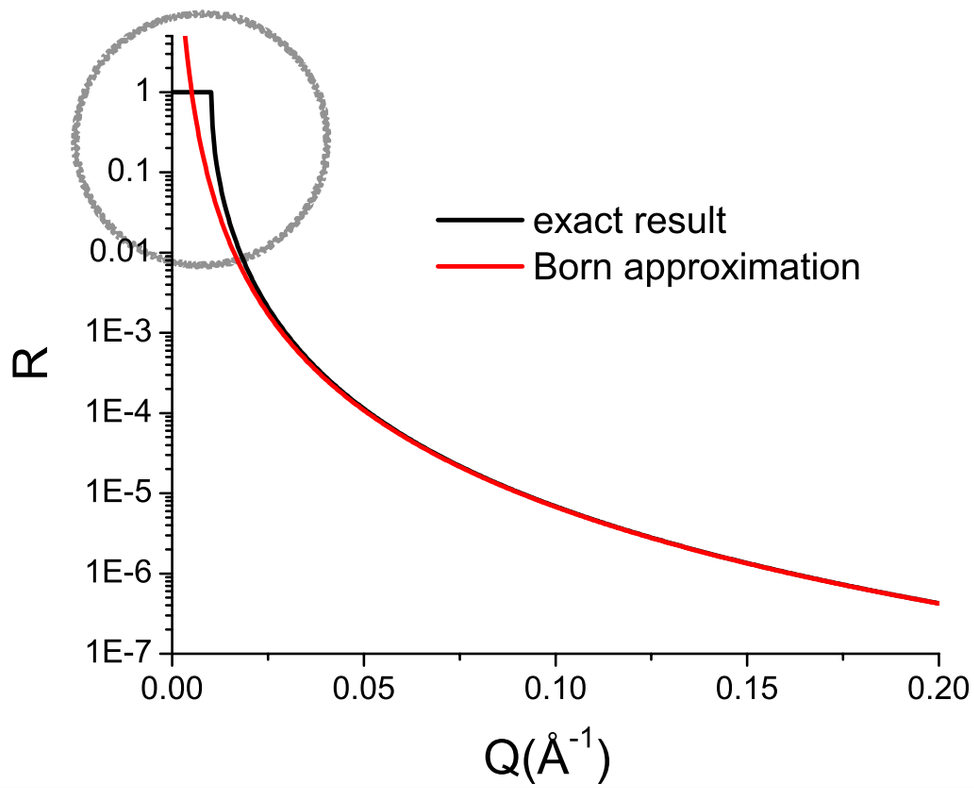
sld profile at an interface

$$\frac{d\rho(z)}{dz} = \rho\delta(z)$$

Fourier transform of the profile

$$\int_{-\infty}^{+\infty} \rho\delta(z)e^{iQz}dz = \rho e^0 = \rho$$

$$R_{Born} = \frac{16\pi^2\ \rho^2}{Q^4}$$



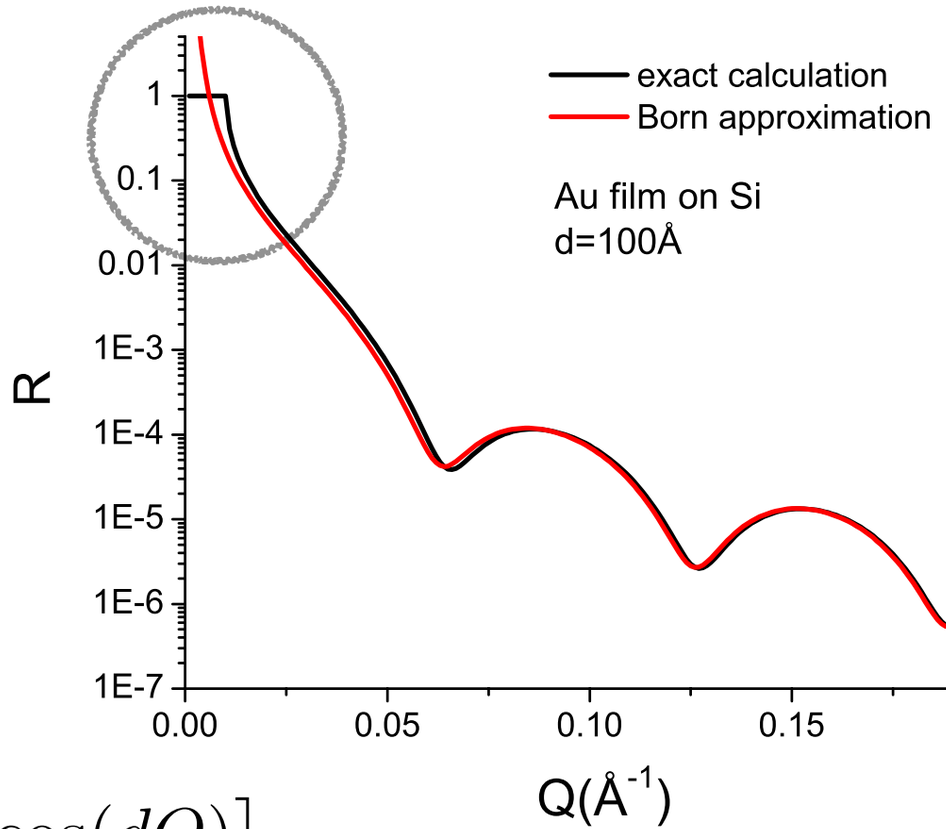
Born approximation: single film reflectivity

$$\rho(z) = \begin{cases} \rho_s, & \text{for } z > d \\ \rho_f, & \text{for } d < z < 0 \\ 0, & \text{for } z < 0 \end{cases}$$

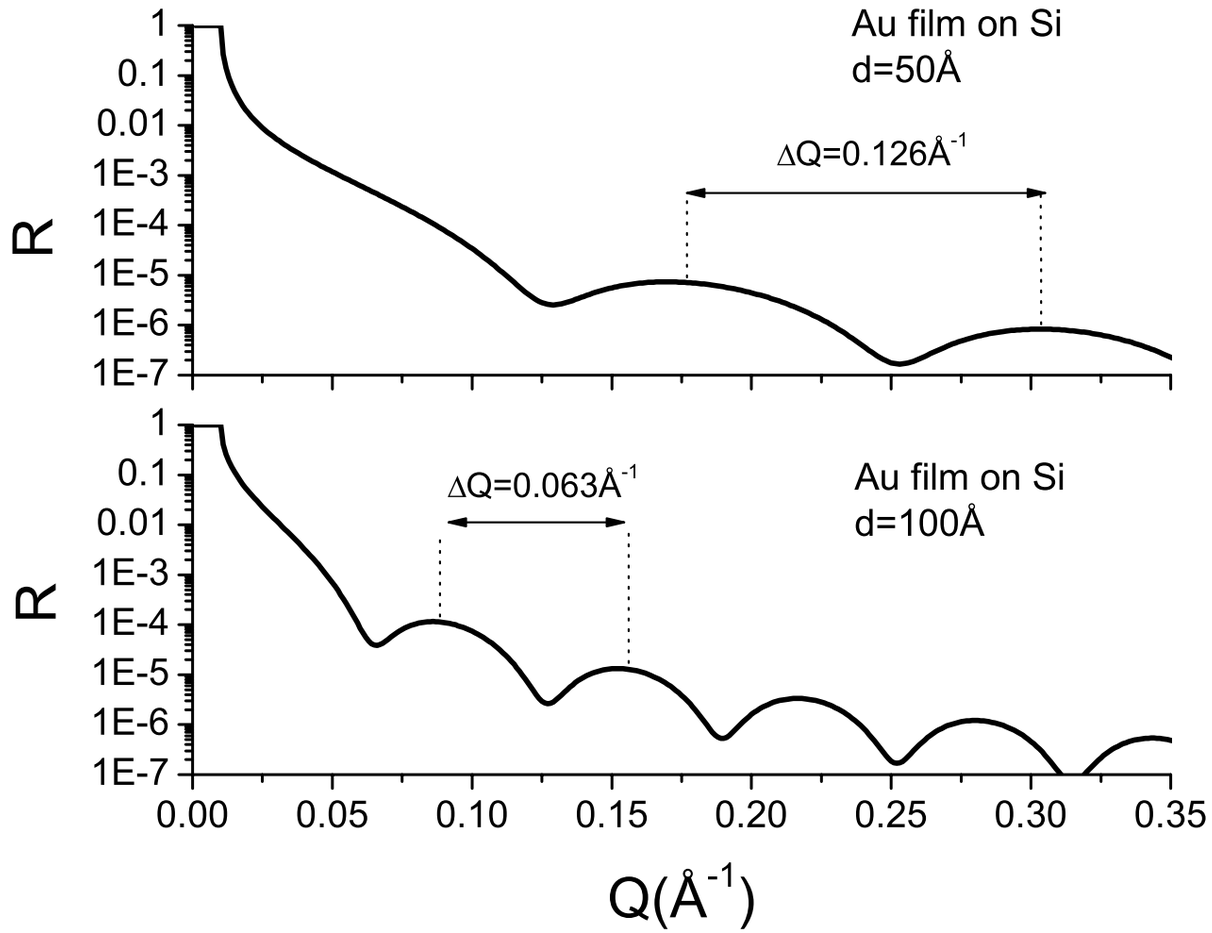
$$\frac{d\rho}{dz} = (\rho_f - \rho_s)\delta(z) - \rho_f \delta(z+d)$$

$$\int_{-\infty}^{+\infty} \frac{d\rho(z)}{dz} e^{iQz} dz = (\rho_f - \rho_s) - \rho_f e^{idQ}$$

$$R = \frac{16\pi^2}{Q^4}\left[\rho_f^2 + (\rho_f - \rho_s)^2 - 2\rho_f(\rho_f - \rho_s)\cos(dQ)\right]$$



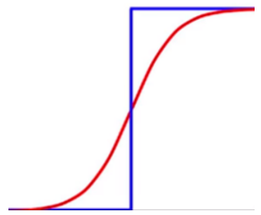
Single film reflectivity



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

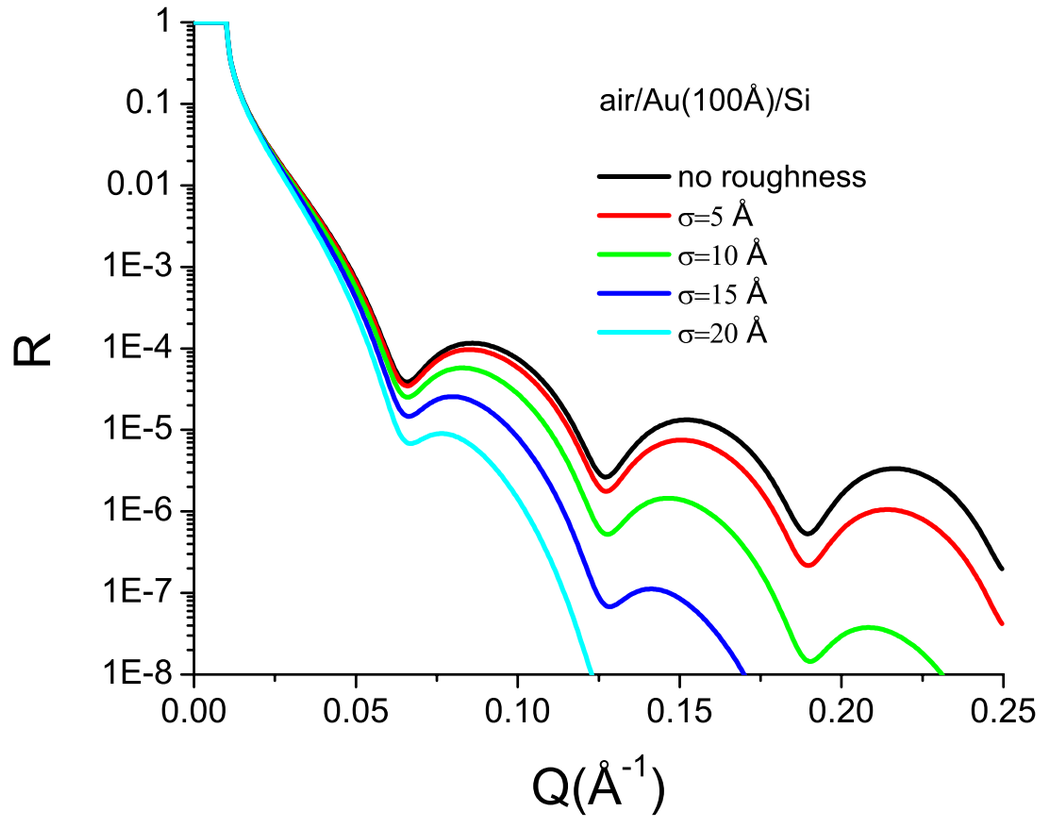
Born approximation: Effects of Roughness

$$\frac{d\rho}{dz} = \frac{\rho}{\sigma\sqrt{2\pi}} e^{-\frac{z^2}{2\sigma^2}}$$

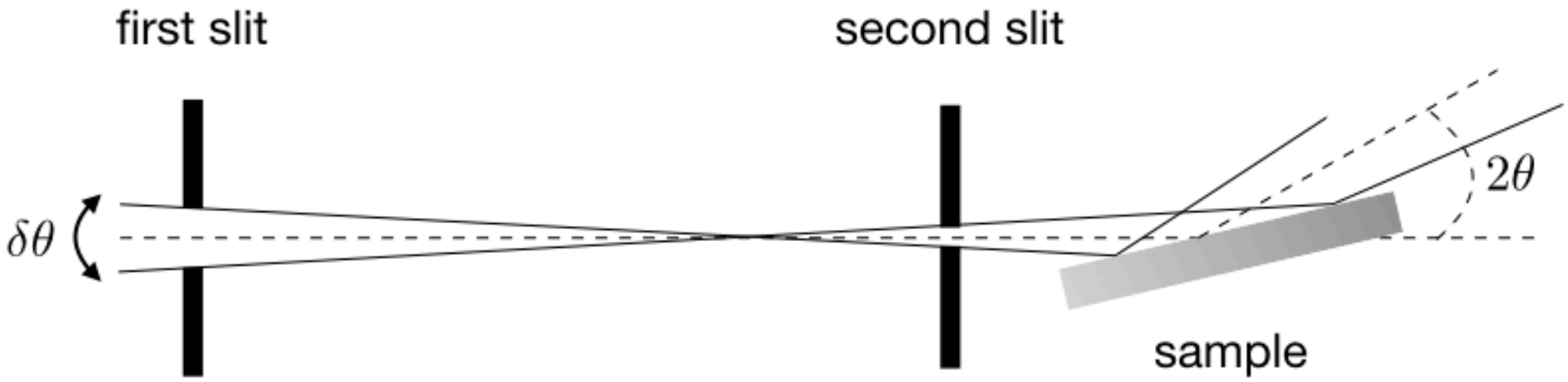


$$\int_{-\infty}^{+\infty} \rho\delta(z) e^{iQz} dz = \rho e^{-\frac{\sigma^2 Q^2}{2}}$$

$$R = \frac{16\pi^2\ \rho^2}{Q^4} e^{-\sigma^2 Q^2}$$



Experimental geometry



Two general types of reflectometers

- single wavelength (angle dispersive)
- time of flight (energy dispersive)

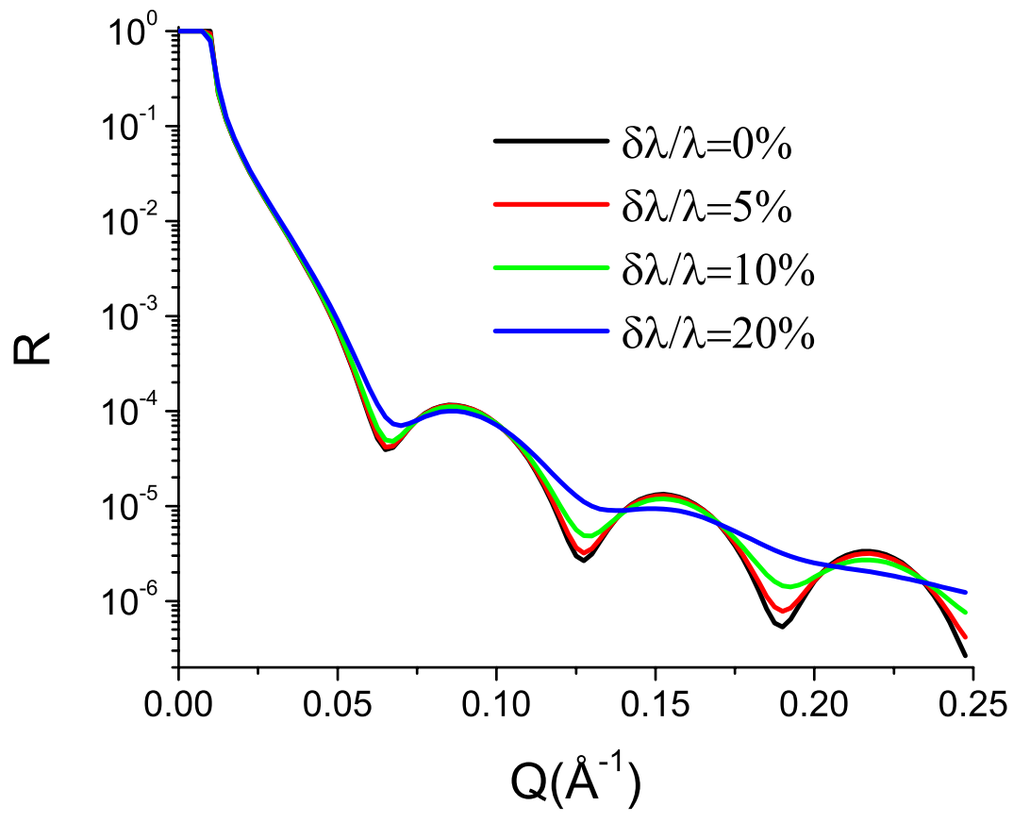
Experimental Q-resolution

$$\frac{Q}{4\pi} = \frac{1}{\lambda}\sin\theta \approx \frac{1}{\lambda}\theta \longrightarrow \frac{\delta(Q/4\pi)}{Q/4\pi} = \frac{\delta Q}{Q} \approx \sqrt{\left(\frac{\delta\lambda}{\lambda}\right)^2 + \left(\frac{\delta\theta}{\theta}\right)^2}$$

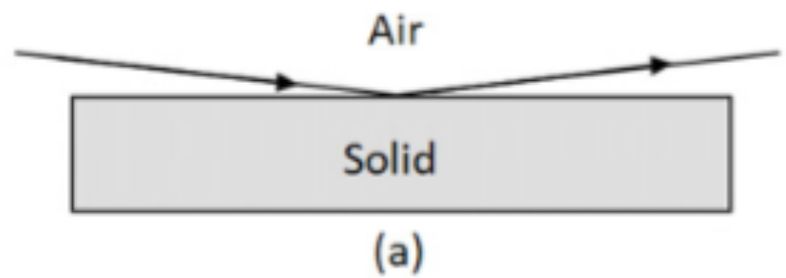
uncertainty propagation theory

$$R_{res}(Q) = \sum_{a=-p}^{p} w_a R(Q + \frac{a}{p}\frac{\delta Q}{Q}) / \sum_{a=-p}^{p} w_a$$

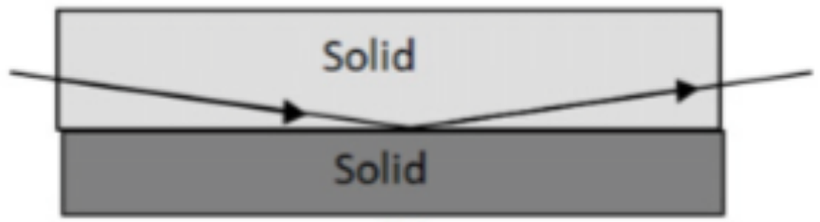
$$w_a = \exp(-2(a/p)^2)$$



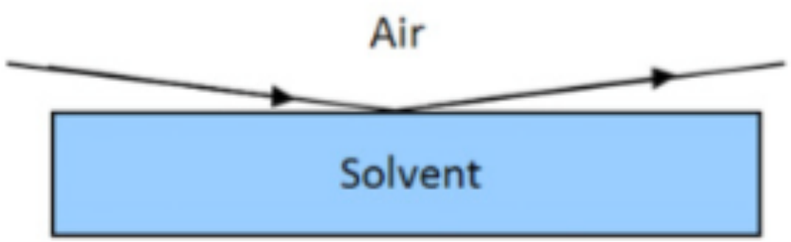
Experimental Setups and Sample environments



(a)



(b)



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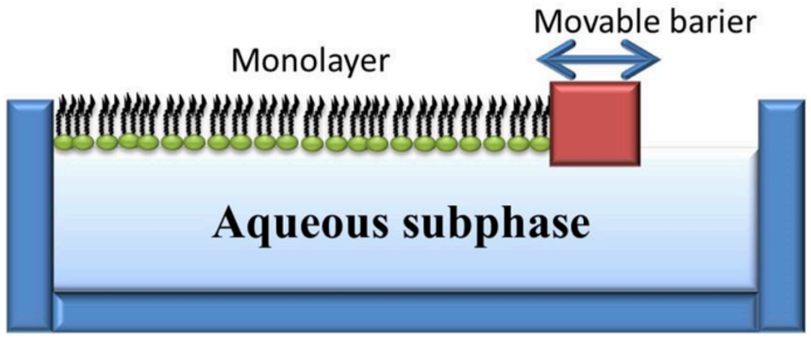

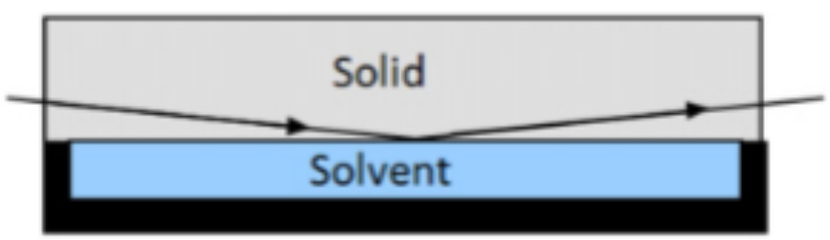

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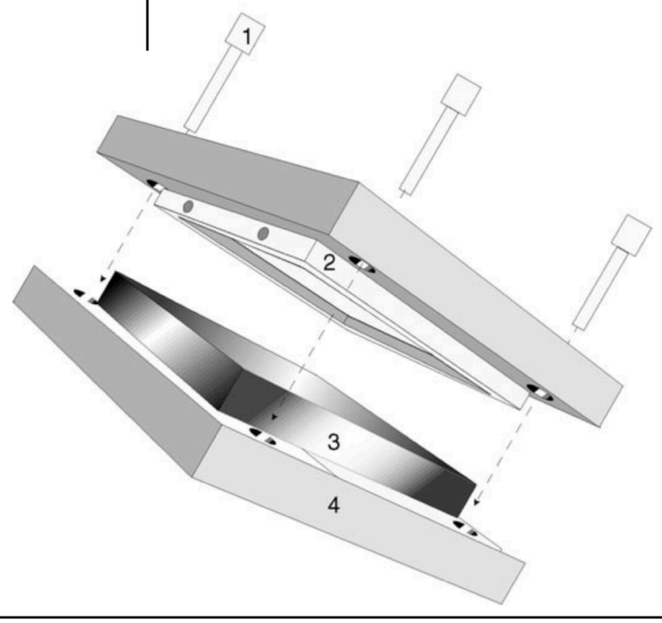
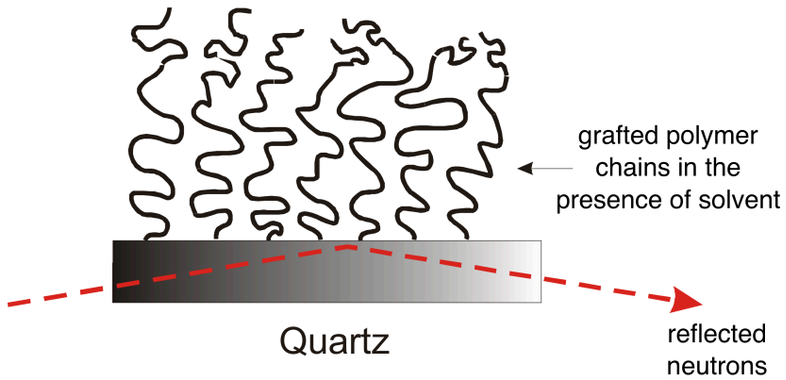
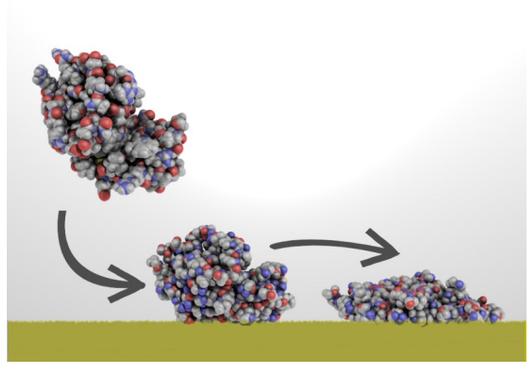


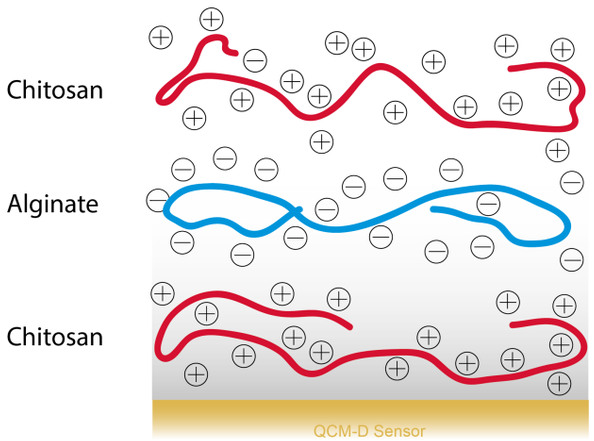
Typical systems that are studied by neutron reflectometry



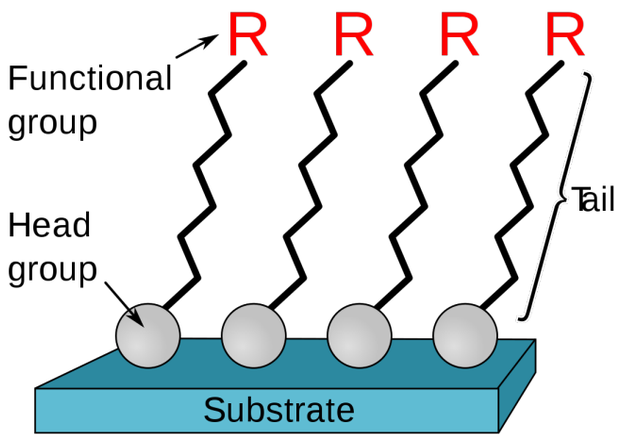
polymer brushes



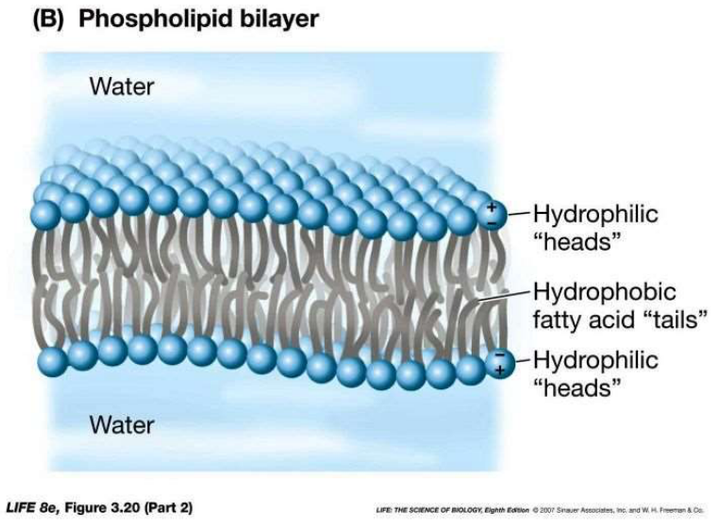
adsorbed proteins



polyelectrolyte
multilayers

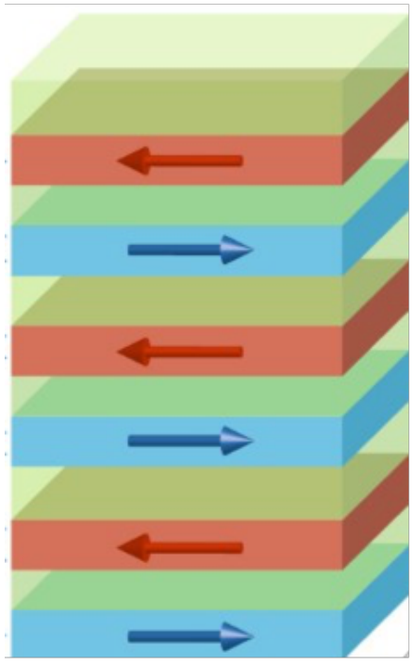


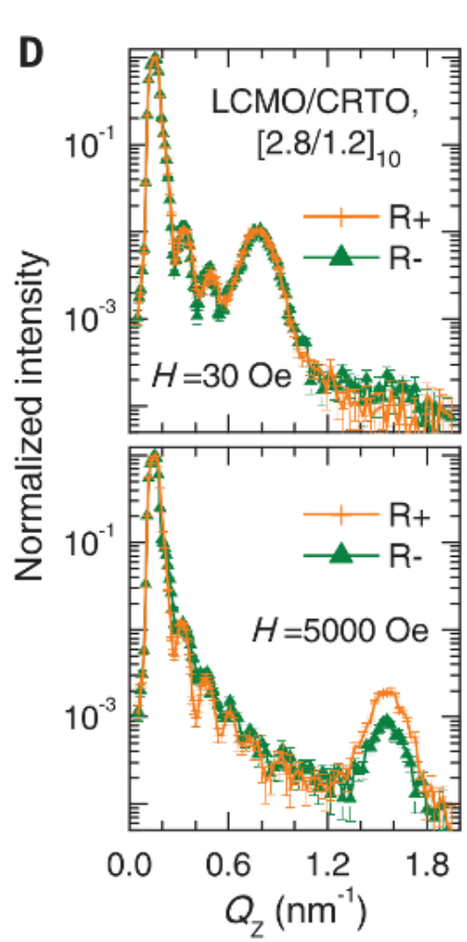
self assembled
monolayers



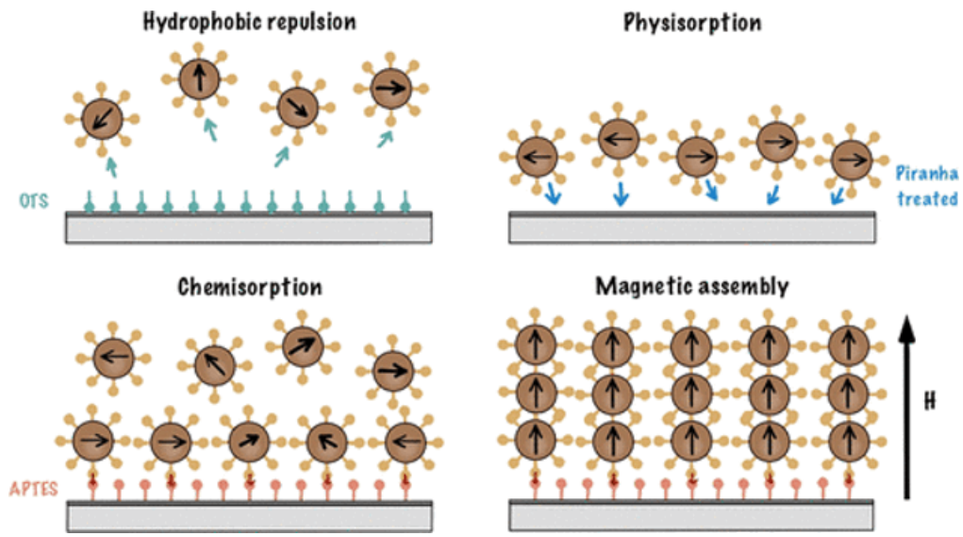
biomembranes

Typical systems that are studied by neutron reflectometry

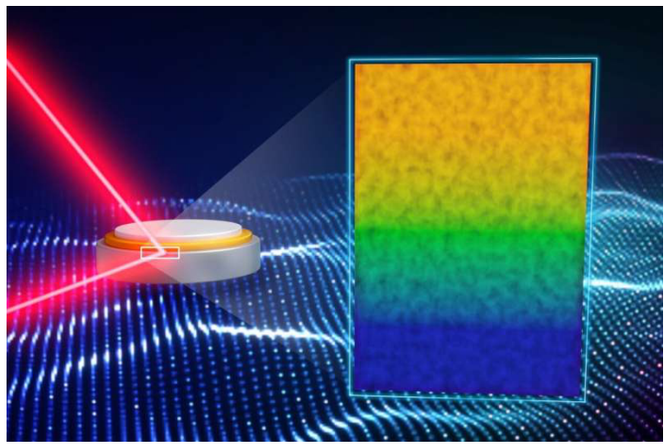




Nano-magnetism

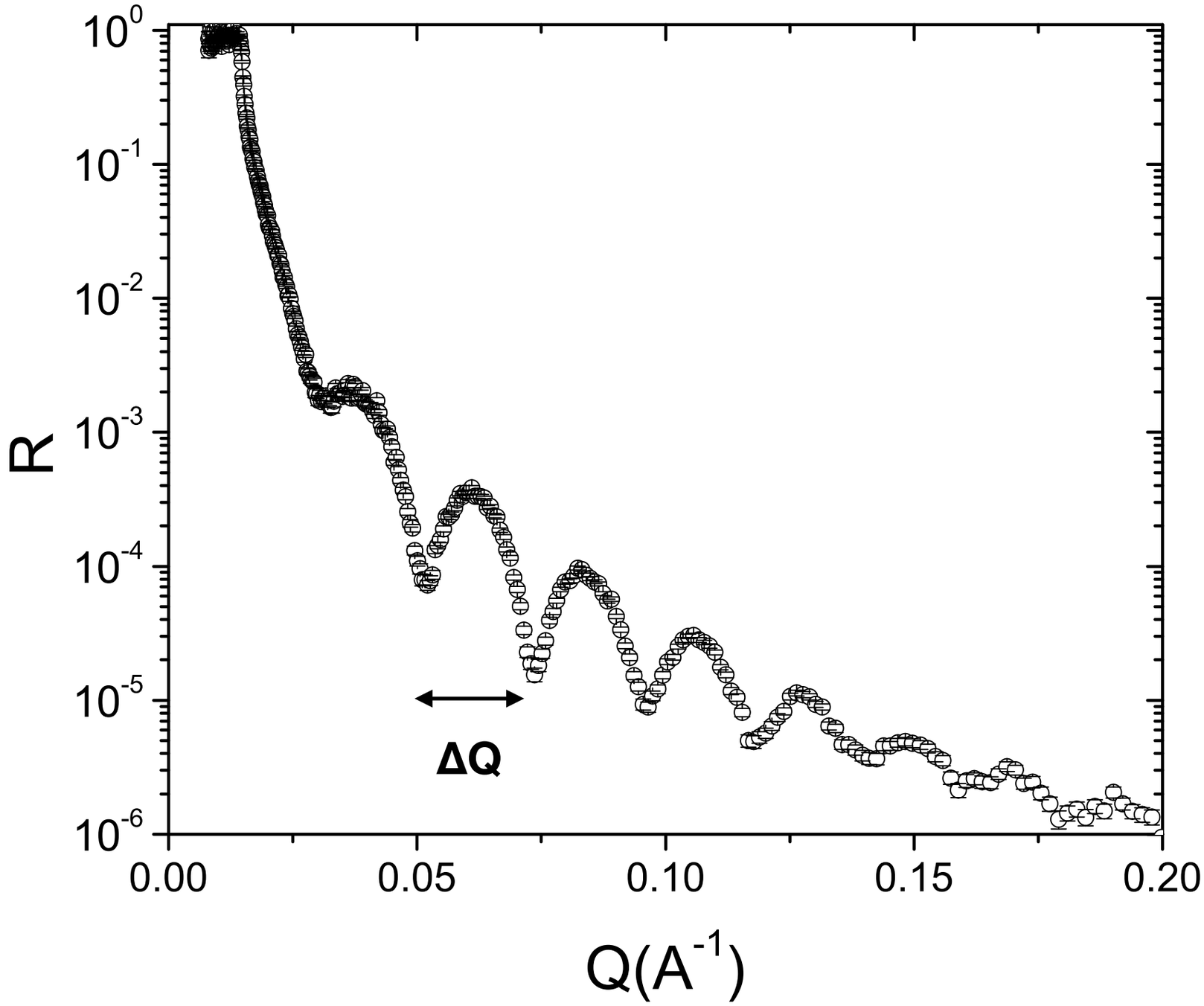


Nanoparticles on surfaces



Battery research

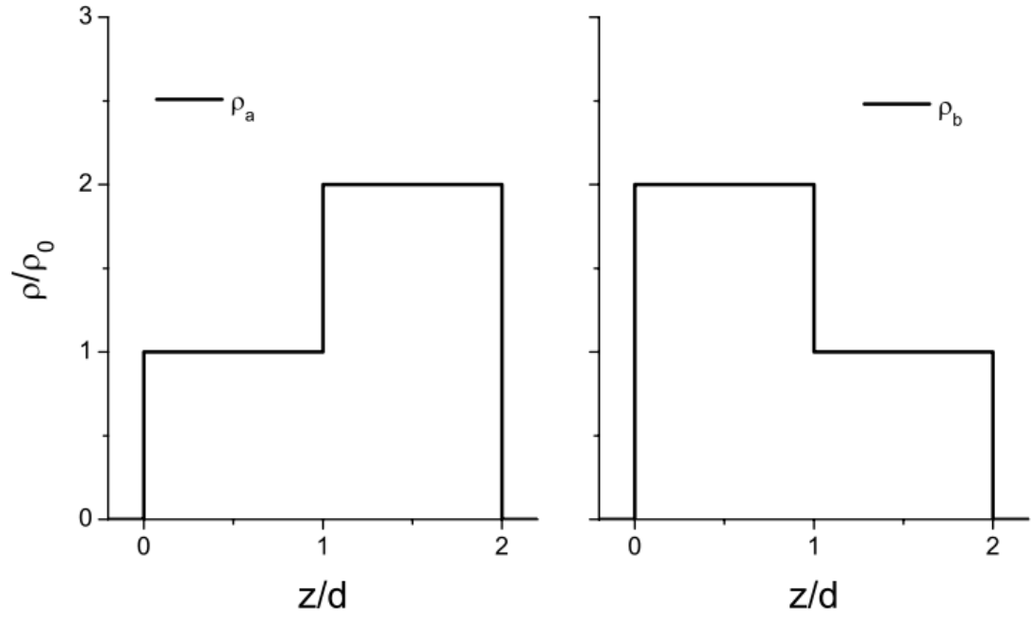
What do we do with acquired data?



A small “excursion” to the phase problem

$$\rho_a(z) = \begin{cases} 0, & \text{for } z > 2d \\ 2\rho_0, & \text{for } d > z > 2d \\ \rho_0, & \text{for } 0 > z > d \\ 0, & \text{for } z < 0 \end{cases}$$

$$\rho_b(z) = \begin{cases} 0, & \text{for } z > 2d \\ \rho_0, & \text{for } d > z > 2d \\ 2\rho_0, & \text{for } 0 > z > d \\ 0, & \text{for } z < 0 \end{cases}$$



$$\int_{-\infty}^{+\infty} \frac{d\rho_a(z)}{dz} e^{iQz} dz = \int_{-\infty}^{+\infty} \left(\rho_0\delta(z) + \rho_0\delta(z+d) - 2\rho_0\delta(z+2d)\right) dz e^{iQz} dz =$$

$$= \rho_0(1 + e^{iQd} - 2e^{i2Qd})$$

$$\int_{-\infty}^{+\infty} \frac{d\rho_b(z)}{dz} e^{iQz} dz = \int_{-\infty}^{+\infty} \left(2\rho_0\delta(z) - \rho_0\delta(z+d) - \rho_0\delta(z+2d)\right) dz e^{iQz} dz =$$

$$= \rho_0(2 + e^{iQd} - e^{i2Qd})$$

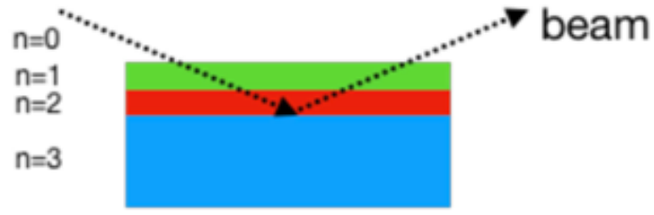
$$|\rho_0(2 + e^{iQd} - e^{i2Qd})|^2 = |\rho_0(1 + e^{iQd} - 2e^{i2Qd})|^2 = \\ = \rho_0[6 - 2\cos(dQ) - 2\cos(2dQ)]$$

$$R = \frac{16\pi^2\rho_0^2}{Q^4}\left[6 - 2\cos(dQ) - 2\cos(2dQ)\right]^2$$

Let us get some hands on experience with actual calculations

ANAKLASIS repository

https://github.com/alexandros-koutsioumpas/anaklasis



Building models…