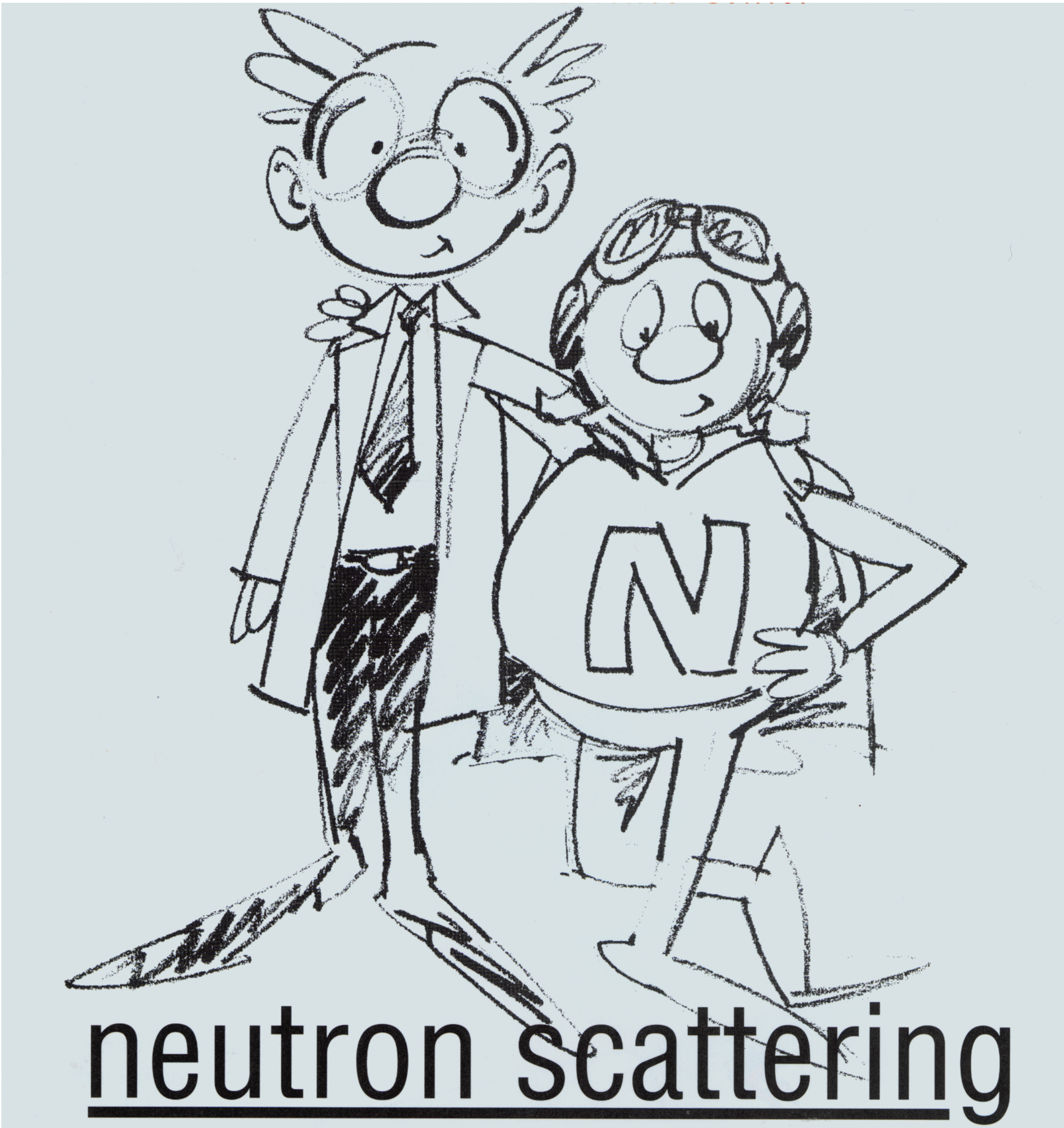

An Introduction to Surface Reflection

by

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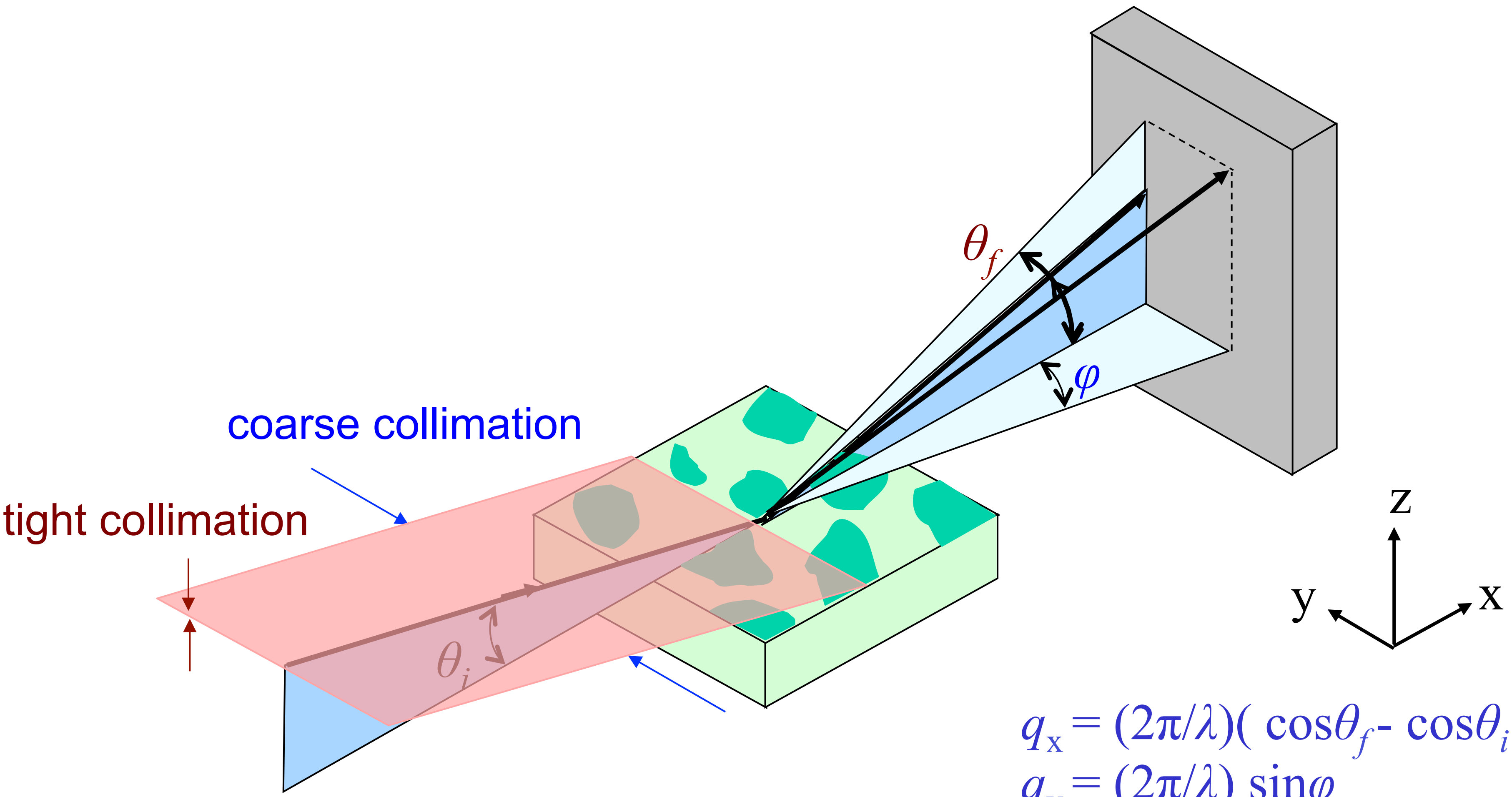
This Lecture

- Why use neutron reflectivity?
- Refractive index for neutrons
- Neutron reflection by a smooth surface
- Neutron penetration depth
- Effect of surface roughness on specular reflection
- Reflection from a surface covered by a thin film
- Diffuse scattering due to surface roughness

Geometry at grazing incidence



$q_x = (2\pi/\lambda)(\cos\theta_f - \cos\theta_i)$
$q_y = (2\pi/\lambda)\sin\varphi$

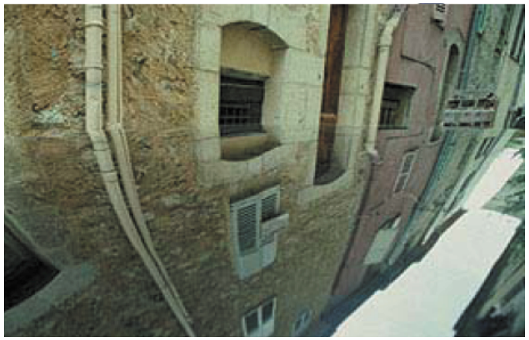
Surface Reflection Is Very Different From Most Neutron Scattering

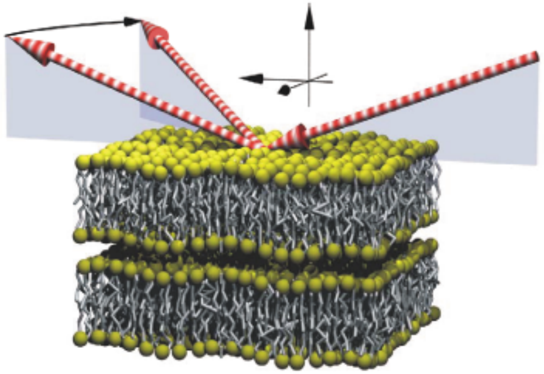
- We worked out the neutron cross section by adding scattering from different nuclei
  - We ignored double scattering processes because these are usually very weak
- This approximation is called the Born Approximation

- Below an angle of incidence called the critical angle, neutrons are perfectly reflected from a smooth surface
  - This is NOT weak scattering and the Born Approximation is not applicable to this case
- Specular reflection is used:
  - In neutron guides
  - In multilayer monochromators and polarizers
  - To probe surface and interface structure in layered systems

Why Use Neutron Reflectivity?

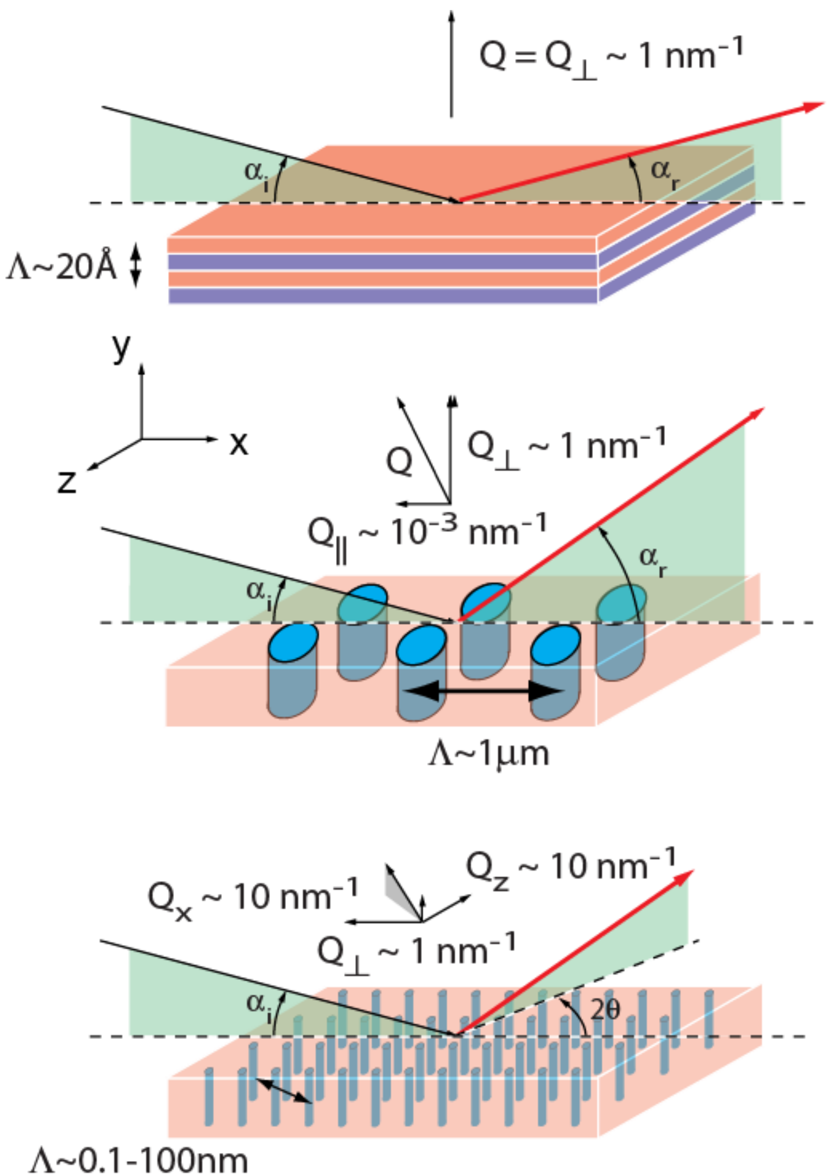
- Neutrons are reflected from most materials at grazing angles
- If the surface is flat and smooth the reflection is specular
  - Perfect reflection below a critical angle
  - Above the critical angle reflectivity is determined by the variation of scattering length density perpendicular to the surface
  - i.e. we can determine the "average" density profile normal to the surface of a film on the surface





Images courtesy of M. Tolan & T. Salditt

Various forms of small (glancing) angle neutron reflection



Specular reflectometry
Depth profiles
(nuclear and/or magnetic)

Off-specular (diffuse) scattering
In-plane correlated roughness
Magnetic stripes
Phase separation (polymers)

Glancing incidence diffraction
Ordering in liquid crystals
Atomic structures near surfaces
Interactions among nanodots

Viewgraph from M. R. Fitzsimmons

The Fermi Pseudo-Potential

$$\frac{d\sigma}{d\Omega} = \frac{1}{\Phi}\frac{1}{d\Omega}\sum_{\vec{k}'\,\mathrm{in}\,d\Omega} W_{\vec{k}\to\vec{k}'}$$ where the sum is over probabilities of all transitions

By Fermi's Golden Rule : $$\sum_{\vec{k}'\,\mathrm{in}\,d\Omega} W_{\vec{k}\to\vec{k}'} = \frac{2\pi}{\hbar}\rho_{\vec{k}'}\left|\left\langle\vec{k}'\middle|V\middle|\vec{k}\right\rangle\right|^2 = \frac{2\pi}{\hbar}\rho_{\vec{k}'}\frac{1}{Y^2}\left|\int_Y V(\vec{r})e^{i(\vec{k}-\vec{k}').\vec{r}}d\vec{r}\right|^2$$

where $\rho_{\vec{k}'}$ is # of momentum states in $d\Omega$, per unit energy, for neutrons in state $\vec{k}'$

Using standard "box normalization", the volume per k state is $(2\pi)^3/Y$ where Y = box volume

Final neutron energy is $E' = \frac{\hbar^2 k'^2}{2m} \Rightarrow dE' = \frac{\hbar^2 k'\,dk'}{m}$ so

$$\rho_{k'}dE' = \text{number of wavevector states in volume } k'^2 dk'\,d\Omega = \frac{Y}{(2\pi)^3}k'^2 dk'\,d\Omega$$

i.e. $$\rho_{\vec{k}'} = \frac{\text{number of wavevector states}}{\mathrm{dE'}} = \frac{Y}{(2\pi)^3}k'\frac{m}{\hbar^2}d\Omega$$

Further, $\Phi = \text{incident flux} = \text{density x velocity} = \frac{1}{Y}\frac{\hbar}{m}k$

Fermi pseudopotential

So, $$\frac{d\sigma}{d\Omega} = \frac{Y}{k}\frac{m}{\hbar}\frac{1}{d\Omega}\frac{2\pi}{\hbar}\frac{Y}{(2\pi)^3}k'\frac{m}{\hbar^2}d\Omega\left|\left\langle\vec{k}'\middle|V\middle|\vec{k}\right\rangle\right|^2 = \left(\frac{m}{2\pi\hbar^2}\right)^2\left|\int V(\vec{r})e^{i(\vec{k}-\vec{k}').\vec{r}}d\vec{r}\right|^2$$ so $$\boxed{V(\vec{r}) = \frac{2\pi\hbar^2}{m}b\delta(\vec{r})}$$

Refractive Index for Neutrons

The nucleus - neutron potential is given by : $V(\vec{r}) = \frac{2\pi\hbar^2}{m} b\delta(\vec{r})$ for a single nucleus.

So the average potential inside the medium is : $\bar{V} = \frac{2\pi\hbar^2}{m}\rho$ where $\rho = \frac{1}{volume}\sum_i b_i$

ρ is called the nuclear Scattering Length Density (SLD) - the same one we used for SANS

The kinetic (and total) energy of neutron in vaccuum is $E = \frac{\hbar^2 k_0^2}{2m}$

Inside the medium the total energy is $\frac{\hbar^2 k^2}{2m} + \bar{V}$

Conservation of energy gives $\frac{\hbar^2 k_0^2}{2m} = \frac{\hbar^2 k^2}{2m} + \bar{V} = \frac{\hbar^2 k^2}{2m} + \frac{2\pi\hbar^2}{m}\rho$ or $k_0^2 - k^2 = 4\pi\rho$

Since $k/k_0 = n =$ refractive index (by definition), and ρ is very small ($\sim 10^{-6}$ A^{-2}) we get :

$$\boxed{n = 1 - \lambda^2 \rho / 2\pi}$$

Since generally n < 1, neutrons are externally reflected from most materials.

Why do we Care about the Refractive Index?

- When the wavevector transfer **Q** is small, the phase factors in the cross section do not vary much from nucleus to nucleus & we can use a continuum approximation

- We can use all of the apparatus of optics to calculate effects such as:
  - External reflection from single surfaces (for example from guide surfaces)
  - External reflection from multilayer stacks (including supermirrors)
  - Focusing by (normally) concave lenses or Fresnel lenses
  - The phase change of the neutron wave through a material for applications such as interferometry or phase radiography
  - Fresnel edge enhancement in radiography

Only Neutrons With Very Low Velocities Perpendicular to a Surface Are Reflected

$k / k_0 = n$

The surface cannot change the neutron velocity parallel to the surface so:

$k_0 \cos\alpha = k \cos\alpha' = k_0 n \cos\alpha'$ i.e n = $\cos\alpha / \cos\alpha'$

$\boxed{\text{Neutrons obey Snell's Law}}$

Since $k^2 = k_0^2 - 4\pi\rho$ $\quad k^2(\cos^2\alpha' + \sin^2\alpha') = k_0^2(\cos^2\alpha + \sin^2\alpha) - 4\pi\rho$

i.e. $k^2 \sin^2\alpha' = k_0^2 \sin^2\alpha - 4\pi\rho$ or $k_z^2 = k_{0z}^2 - 4\pi\rho$

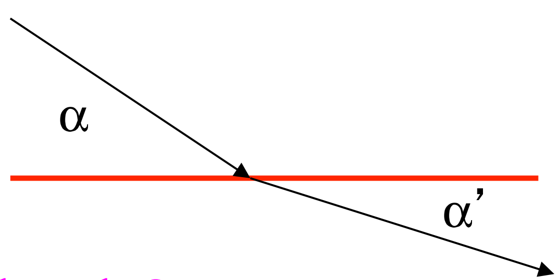
The critical value of k_{0z} for total external reflection is $k_{0z} = \sqrt{4\pi\rho}$

For quartz $k_{0zl}^{critical} = 2.05x10^{-3}$ A^{-1}

$(2\pi / \lambda)\sin\alpha_{\text{critical}} = k_{0zl}^{critical} \Rightarrow$

$\alpha_{\text{critical}}(^o) \approx 0.02\lambda(A)$ for quartz

Note: $\alpha_{\text{critical}}(^o) \approx 0.1\lambda(A)$ for nickel



How do we make a neutron bottle?

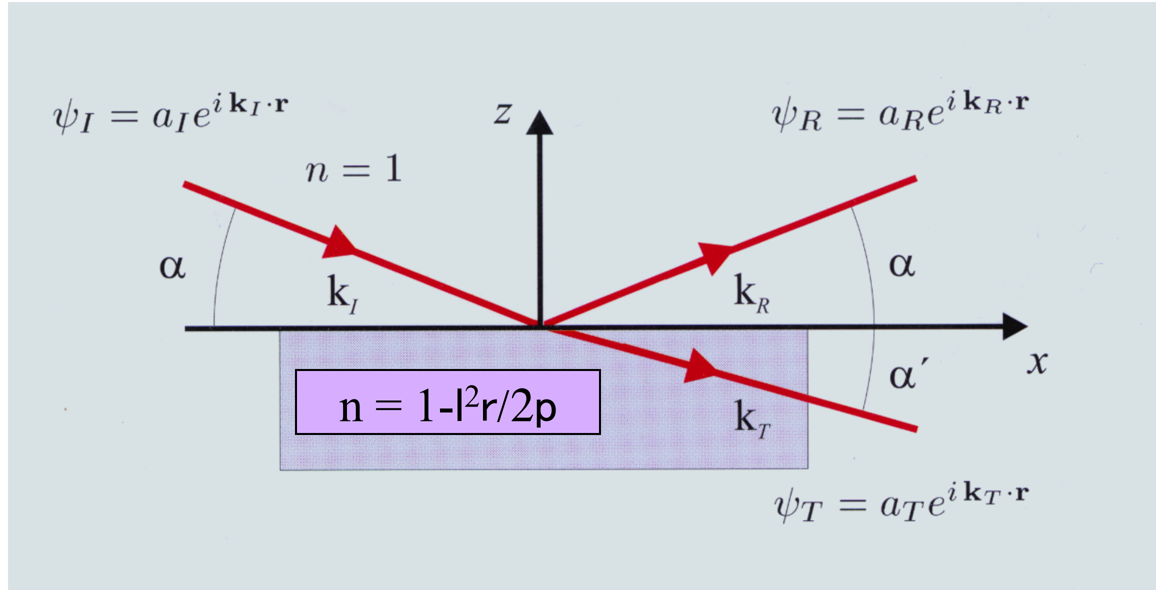
Reflection of Neutrons by a Smooth Surface: Fresnel’s Law

continuity

of ψ & $\dot{\psi}$ at z = 0 $\Rightarrow$

$$a_I + a_R = a_T \qquad (1)$$

$$a_I \vec{k}_I + a_R \vec{k}_R = a_T \vec{k}_T$$



components perpendicular and parallel to the surface:

$$a_I k\cos\alpha + a_R k\cos\alpha = a_T nk\cos\alpha' \qquad (2)$$

$$-(a_I - a_R)k\sin\alpha = -a_T nk\sin\alpha' \qquad (3)$$

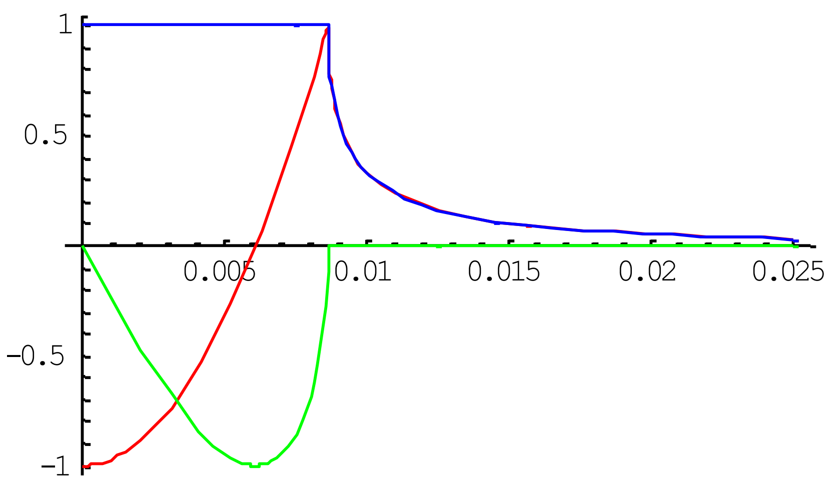
(1) & (2) => Snell's Law: $\boxed{\cos\alpha = n\cos\alpha'}$

$$(1)\ \&\ (3) \Rightarrow \frac{(a_I - a_R)}{(a_I + a_R)} = n\frac{\sin\alpha'}{\sin\alpha} \approx \frac{\sin\alpha'}{\sin\alpha} = \frac{k_{Tz}}{k_{Iz}}$$

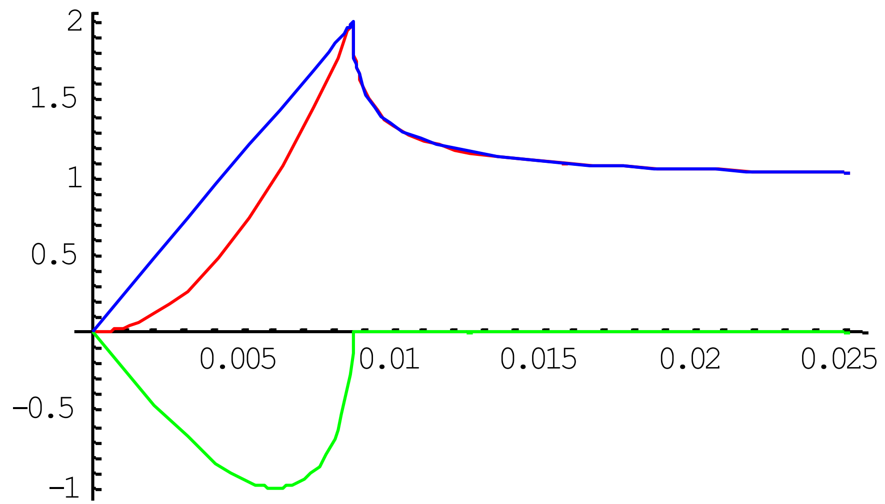
so reflectance is given by $\boxed{r = a_R / a_I = (k_{Iz} - k_{Tz})/(k_{Iz} + k_{Tz})}$

What do the Amplitudes a_R and a_T Look Like?

- For reflection from a flat substrate, both a_R and a_T are complex when $k_0 < 4\pi\rho$ I.e. below the critical edge. For $a_I = 1$, we find:

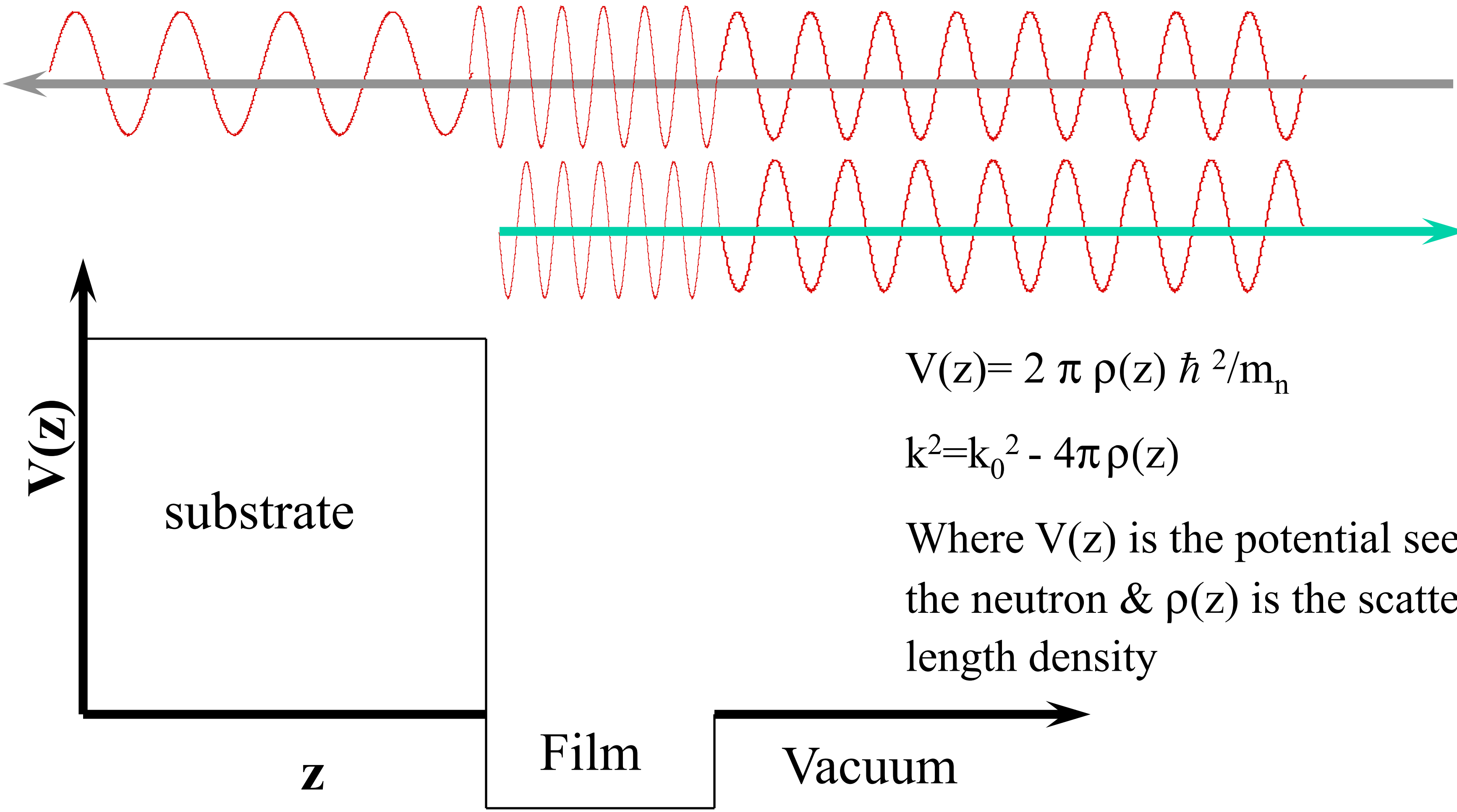


Real (red) & imaginary (green) parts of a_R plotted against k_0. The modulus of a_R is plotted in blue. The critical edge is at $k_0 \sim 0.009$ A^{-1} . Note that the reflected wave is completely out of phase with the incident wave at $k_0 = 0$



Real (red) and imaginary (green) parts of a_T. The modulus of a_T is plotted in blue. Note that a_T tends to unity at large values of k_0 as one would expect

One can also think about Neutron Reflection from a Surface as a 1-d Problem



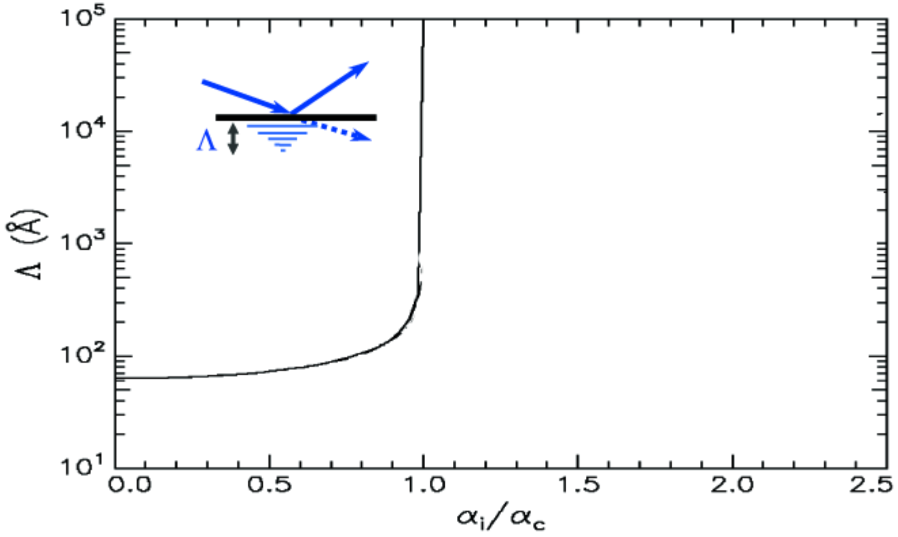
$V(z) = 2\pi\rho(z)\hbar^2/m_n$

$k^2 = k_0^2 - 4\pi\rho(z)$

Where V(z) is the potential seen by the neutron & ρ(z) is the scattering length density

Penetration Depth

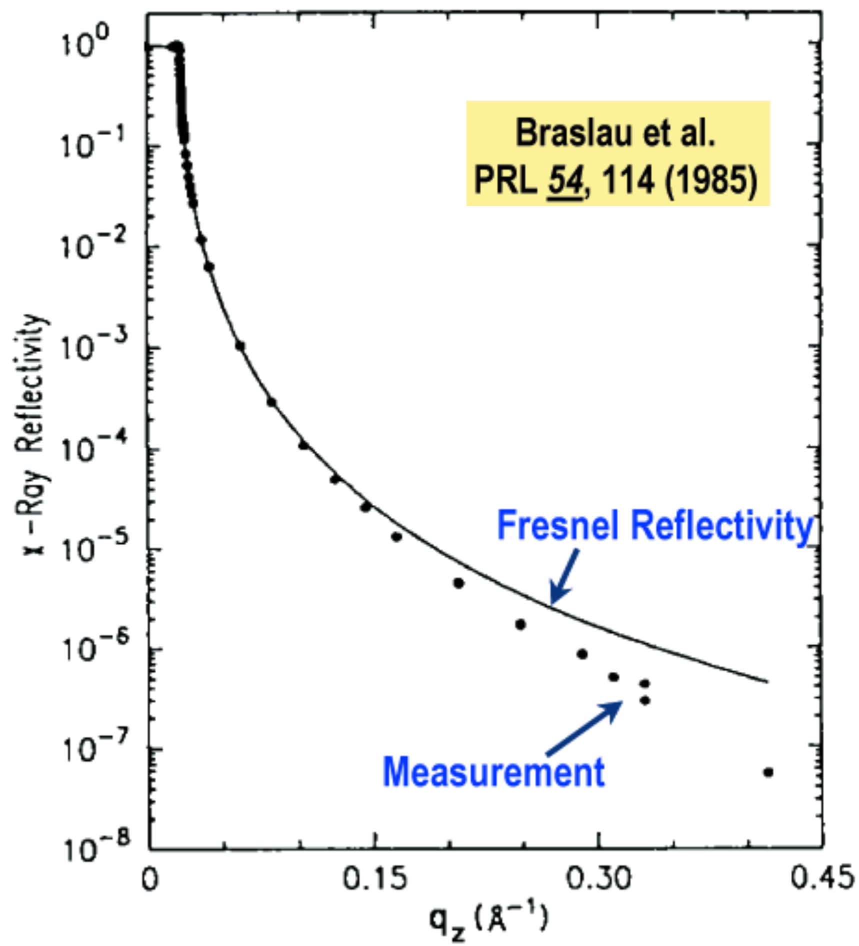
- In the absence of absorption, the penetration depth becomes infinite at large enough angles
- Because k_z is imaginary below the critical edge (recall that $k_z^2 = k_{0z}^2 - 4\pi\rho$), the transmitted wave is evanescent
- The penetration depth $\Lambda = 1/\operatorname{Im}(k)$
- Around the critical edge, one may tune the penetration depth to probe different depths in the sample



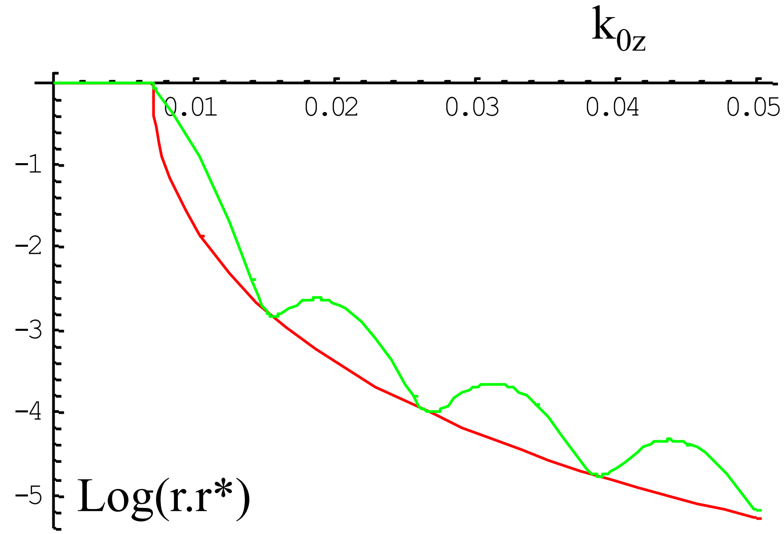
Measured Reflectivity

- We do not measure the reflectance, r, but the reflected intensity, r.r* i.e., just as in diffraction, we lose phase information

- Compare measured and Fresnel x-ray reflectivities for a water surface
  - Difference is due to surface roughness



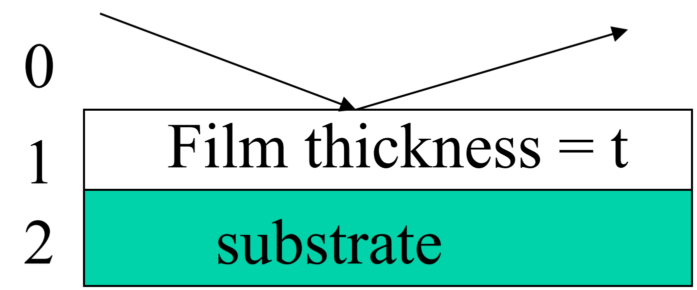
Fresnel's Law for a Thin Film



- $r=(k_{0z}-k_{1z})/(k_{1z}+k_{0z})$ is Fresnel's law
- Evaluate with $\rho=4.10^{-6}$ A^{-2} gives the red curve with critical wavevector given by $k_{0z} = (4\pi\rho)^{1/2}$
- If we add a thin layer on top of the substrate we get interference fringes & the reflectance is given by:

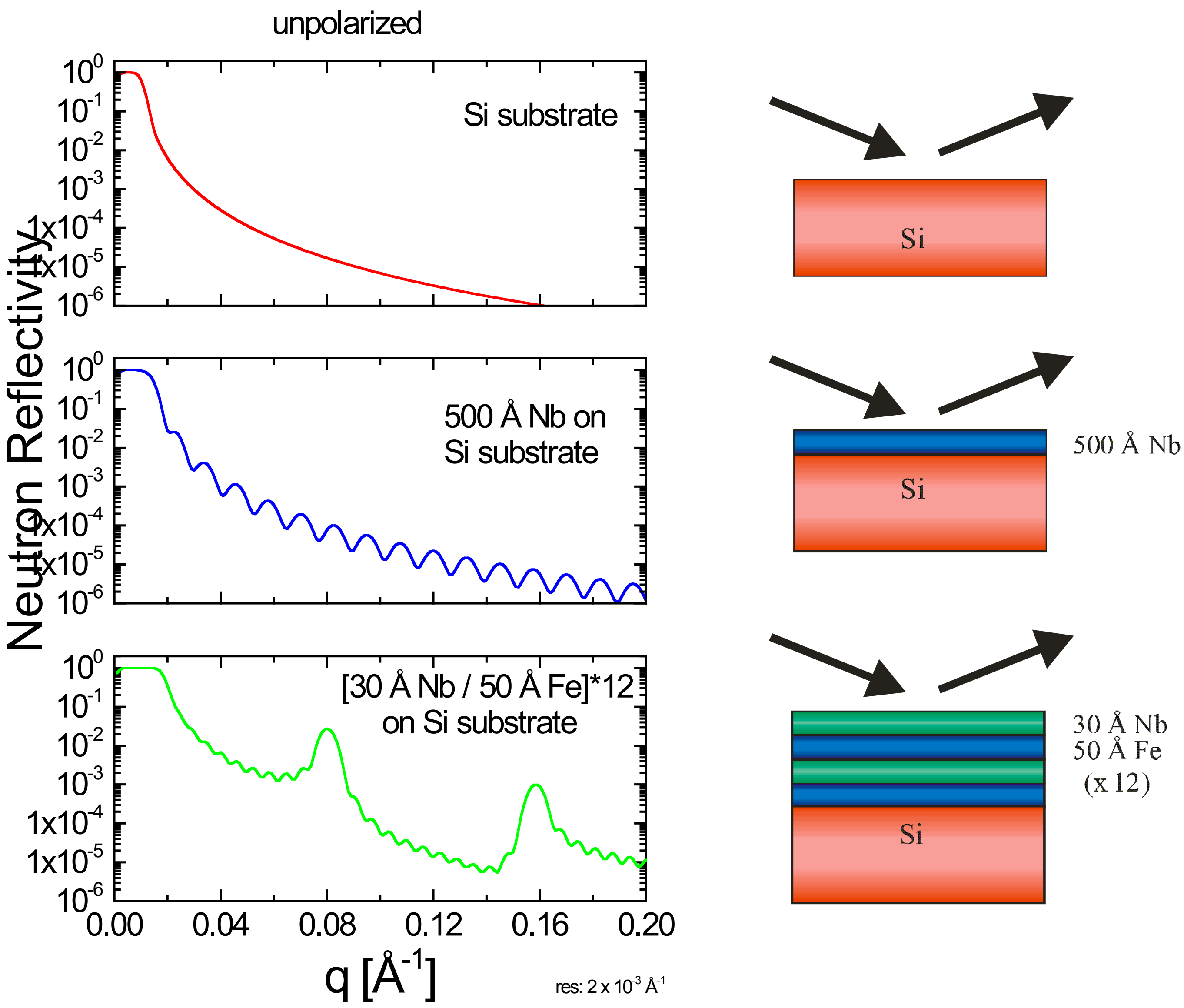
$$r = \frac{r_{01} + r_{12} e^{i2k_{1z}t}}{1 + r_{01} r_{12} e^{i2k_{1z}t}}$$

and we measure the reflectivity R = r.r*



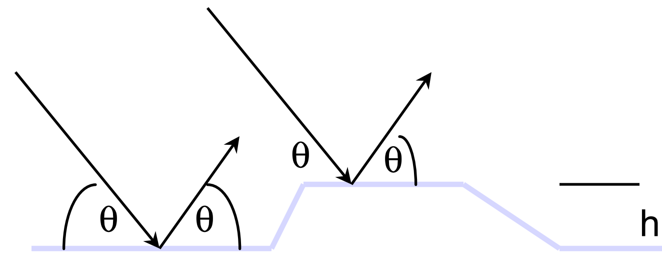
- If the film has a higher scattering length density than the substrate we get the green curve (if the film scattering is weaker than the substance, the green curve is below the red one)
- The fringe spacing at large k_{0z} is ~ pt (a 250 A film was used for the figure)

Reflectivity of Layered Structures



When Does a “Rough” Surface Scatter Diffusely?

- Rayleigh criterion

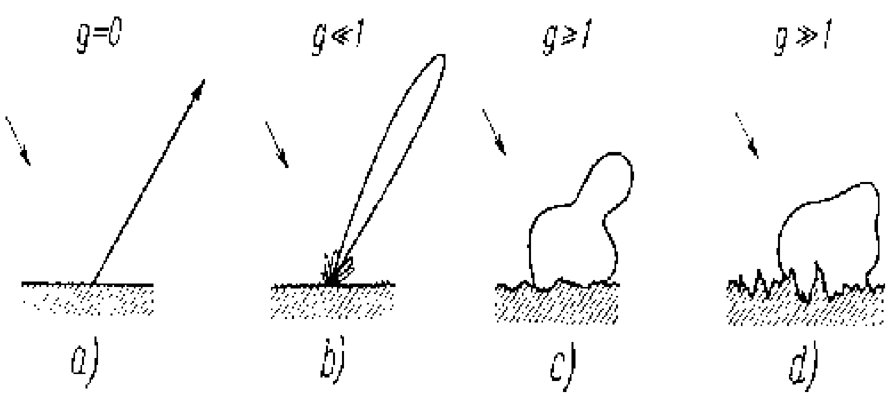


path difference: $\Delta r = 2\ h \sin\theta$

phase difference: $\Delta\Phi = (4\pi h/\lambda) \sin\theta$

boundary between rough and smooth: $\Delta\Phi = \pi/2$

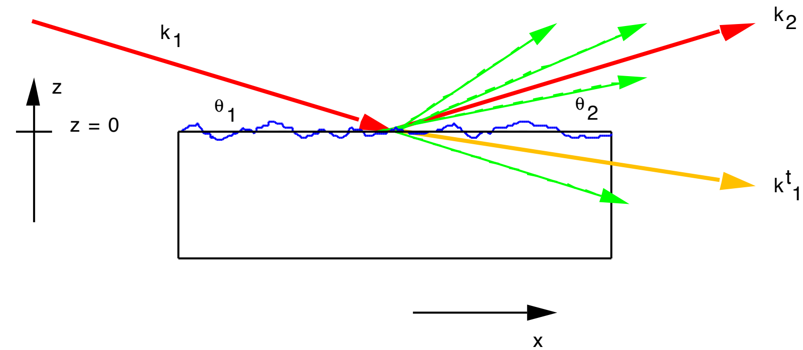
that is $h < \lambda/(8\sin\theta)$ **for a smooth surface**



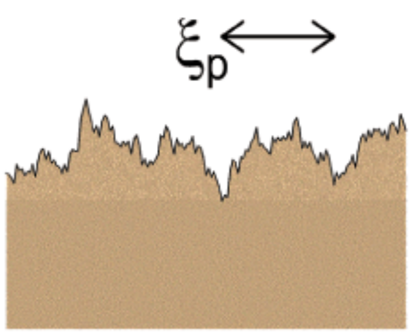
where $g = 4\ \pi\ h \sin\theta / \lambda = Q_z\ h$

Surface Roughness

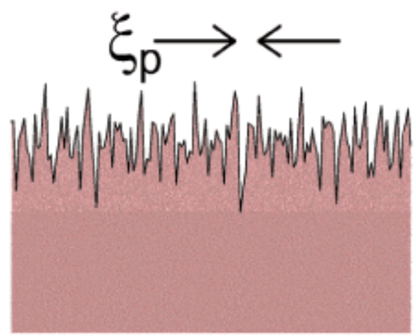
- Surface roughness causes diffuse (non-specular) scattering and so reduces the magnitude of the specular reflectivity



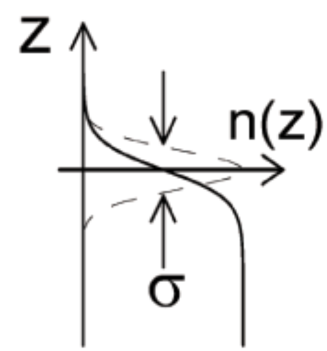
- The way in which the specular reflection is damped depends on the length scale of the roughness in the surface as well as on the magnitude and distribution of roughness



"sparkling sea" model
-- specular from many facets



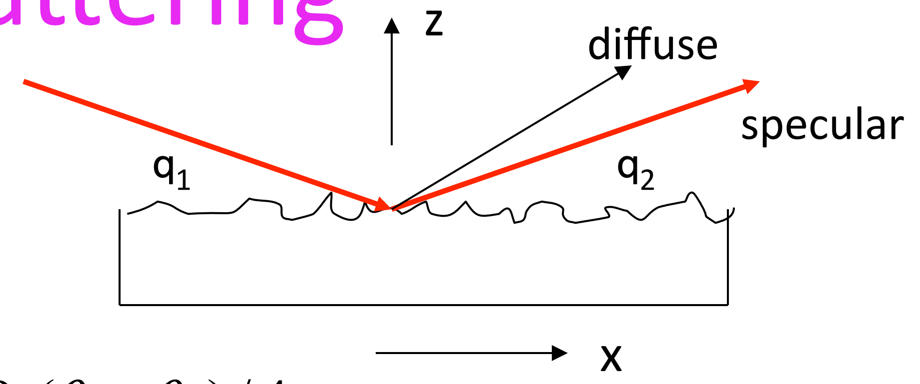
each piece of surface scatters indepedently
-- Nevot Croce model



Note that roughness introduces a SLD profile averaged over the sample surface

$$R = R_F e^{-2k_{1z}k^t_{1z}\sigma^2}$$

Diffuse Scattering



If an interface is rough it will scatter

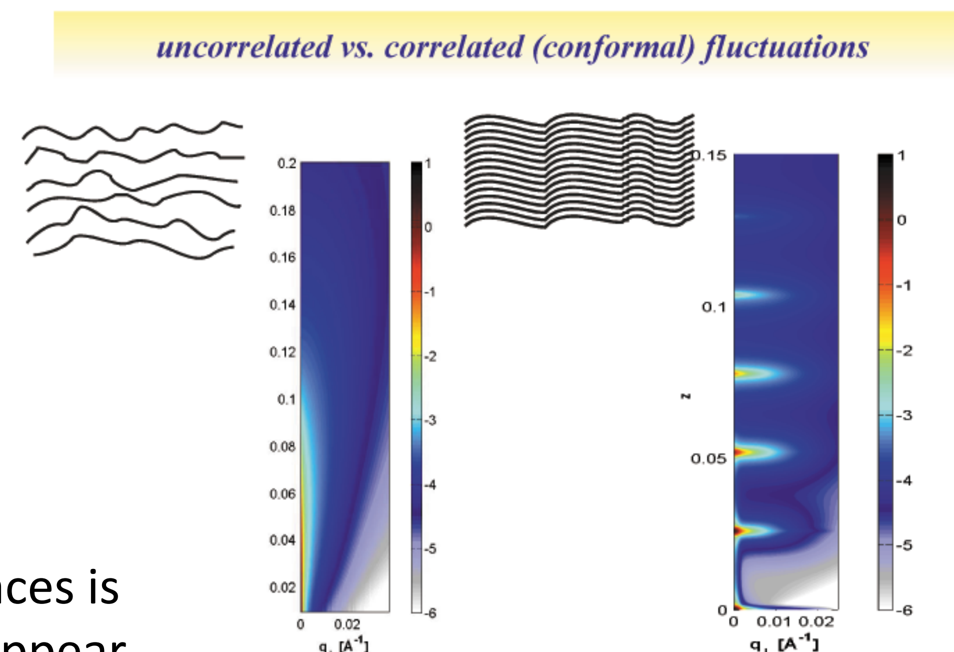
both specularly and diffusely

$$Q_x = k(\cos\theta_2 - \cos\theta_1) \approx \frac{k}{2}(\theta_1^2 - \theta_2^2) = Q_z(\theta_1 - \theta_2)/4$$

$$\text{If } \theta_1 - \theta_2 = 1^\circ \approx 0.02 \text{ radians}$$

$$Q_x \approx 0.005 Q_z \approx 10^{-3} \ \text{nm}^{-1}$$

$$\text{i.e. the in-plane length scale}$$
$$\text{probed can be} \sim 1 \text{ micron!!}$$



If the roughness of neighboring interfaces is correlated, the diffuse scattering will appear as constant-Q_z ridges extended in Q_x

Image from G.Brotons & L. Belloni (CEA/SACLAY).