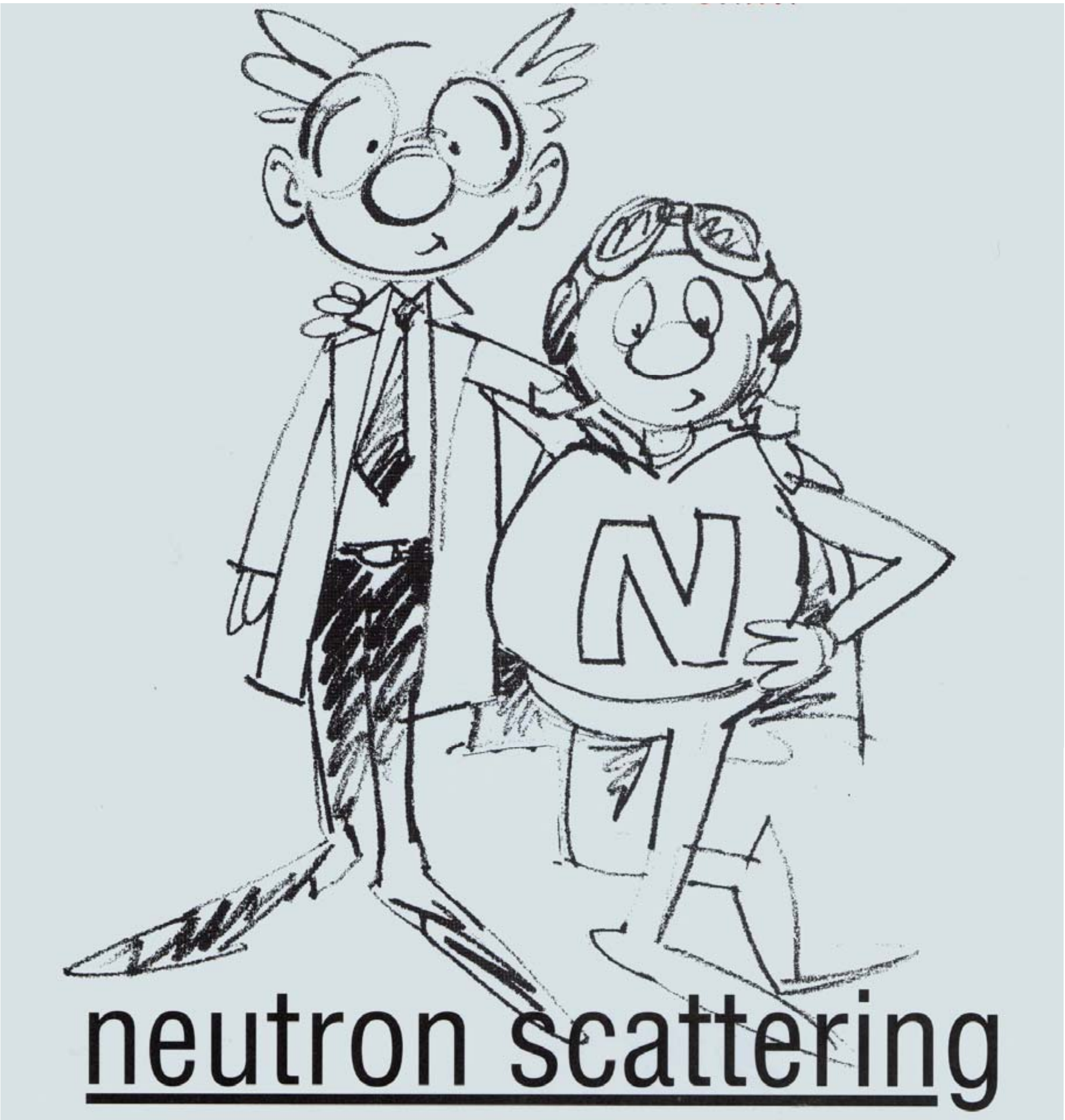


Introduction to Surface Reflection

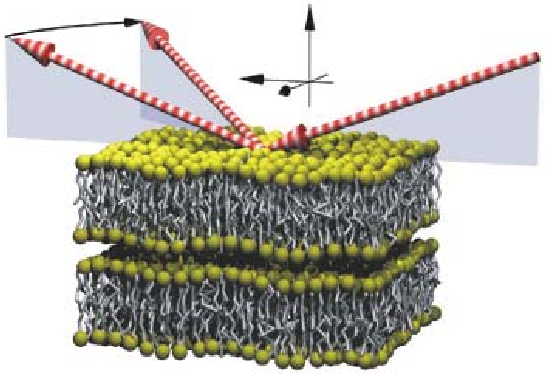
by

Roger Pynn

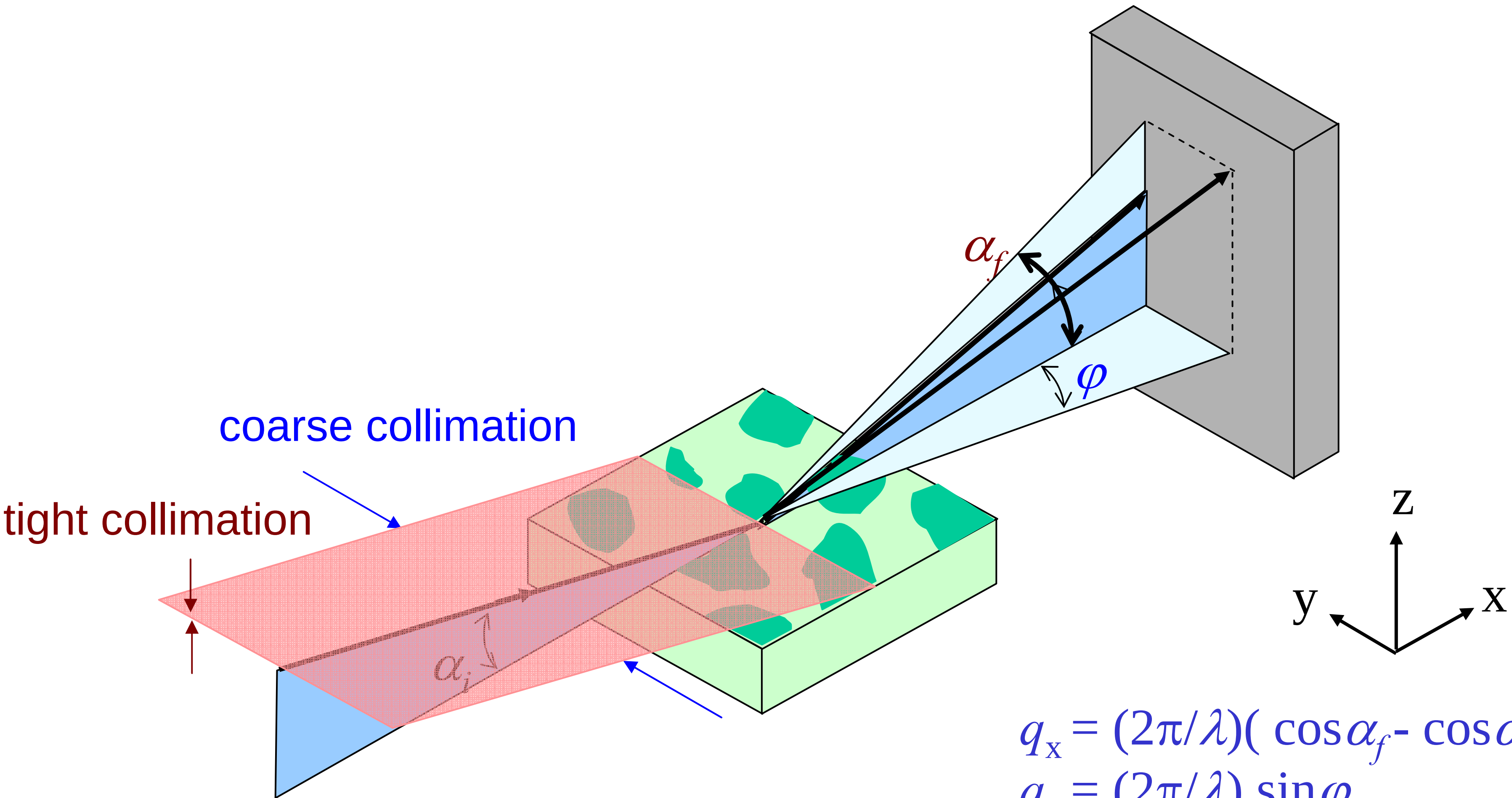
Indiana University
&
Spallation Neutron Source

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 (just like light reflecting from a soap film)
  - i.e. we can determine the "average" density profile normal to the surface of a film on the surface



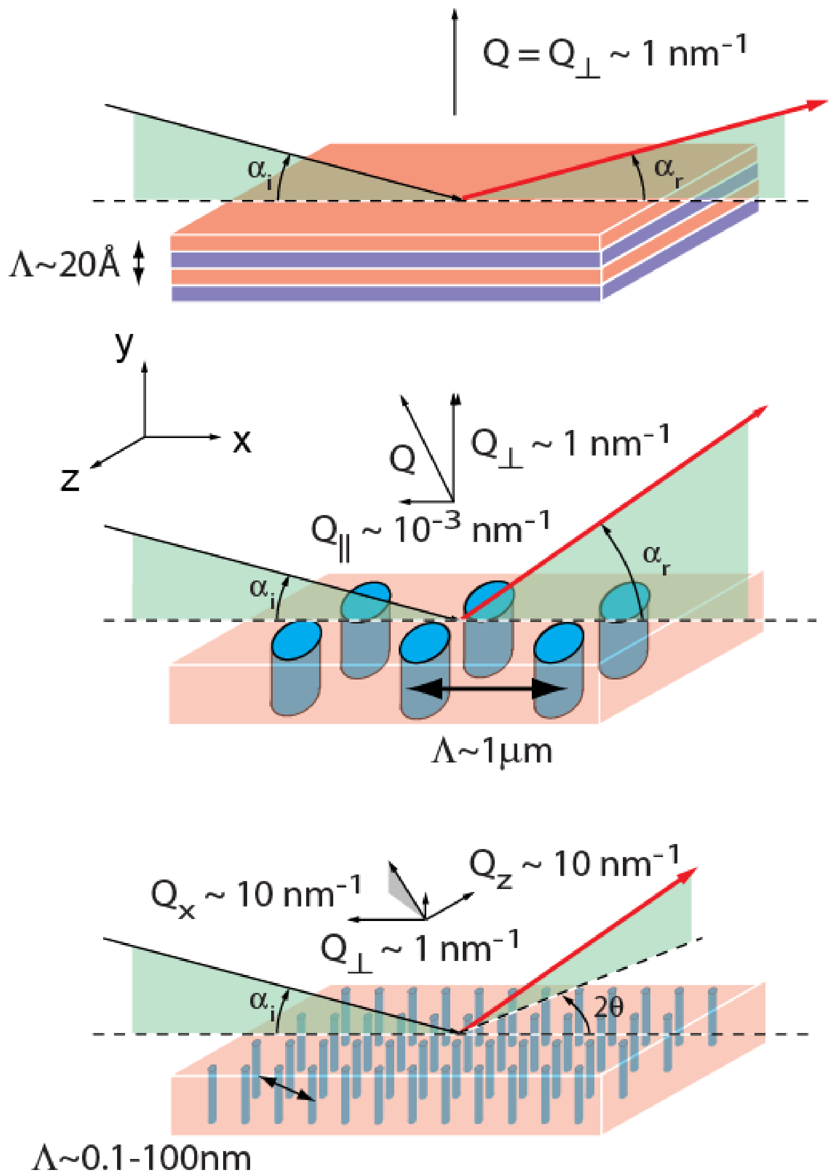
Geometry at grazing incidence



$$q_x = (2\pi/\lambda)(\cos\alpha_f - \cos\alpha_i)$$

$$q_y = (2\pi/\lambda)\sin\varphi$$

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)

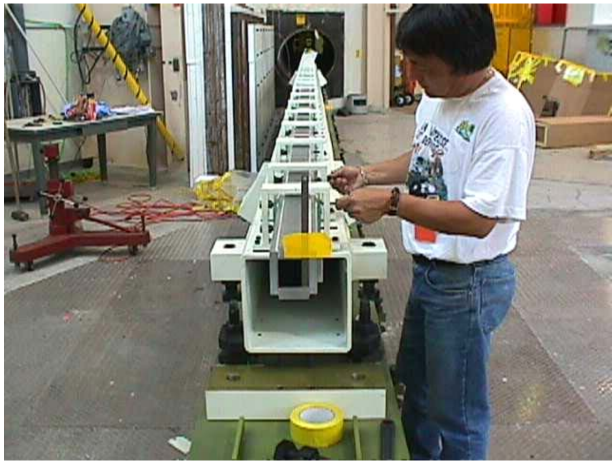
Glancing incidence diffraction
Ordering in liquid crystals
Atomic structures near surfaces
Interactions among nanodots

Viewgraph from M. R. Fitzsimmons

Surface Reflection Is Very Different From Most Neutron Scattering

- Usually we work out the neutron cross section by adding scattering from different nuclei
  - Double scattering processes were ignored 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 – reflectometry



What Is the Neutron Wavevector Inside a Medium?

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

where b is the nuclear scattering length.

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

ρ is called the nuclear Scattering Length Density (SLD)

The neutron obeys Schrodinger's equation :

$$\left[\nabla^2 + 2m(E - \overline{V})/\hbar^2\right]\psi(r) = 0$$

in vacuo $\psi(r) = e^{i\vec{k}_o.\vec{r}}$ so $k_0^2 = 2mE/\hbar^2$. Simlarly $k^2 = 2m(E - \overline{V})/\hbar^2 = k_0^2 - 4\pi\rho$

where k_0 is neutron wavevector *in vacuo* and k is the wavevector in a material

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

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

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

Only Those Thermal or Cold Neutrons With Very Low Velocities Perpendicular to a Surface Are Reflected

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

$k_0 \cos\alpha = k\cos\alpha' = k_0 n \cos\alpha'$ because $k/k_0 = \mathrm{n}$; i.e $\mathrm{n} = \cos\alpha / \cos\alpha'$

Neutrons obey Snell's Law

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

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 $k_{0z}^{critical} = \sqrt{4\pi\rho}$

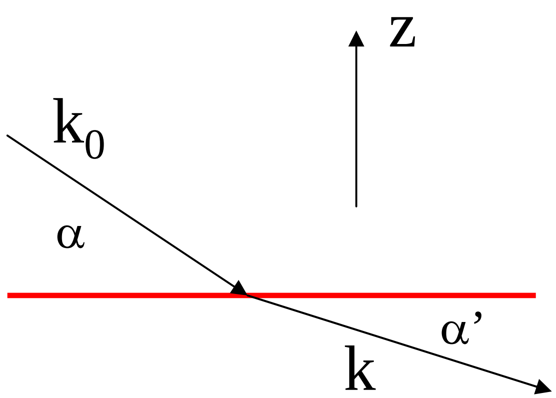
For quartz $k_{0z}^{critical} = 2.05x10^{-3}$ Å^{-1}

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

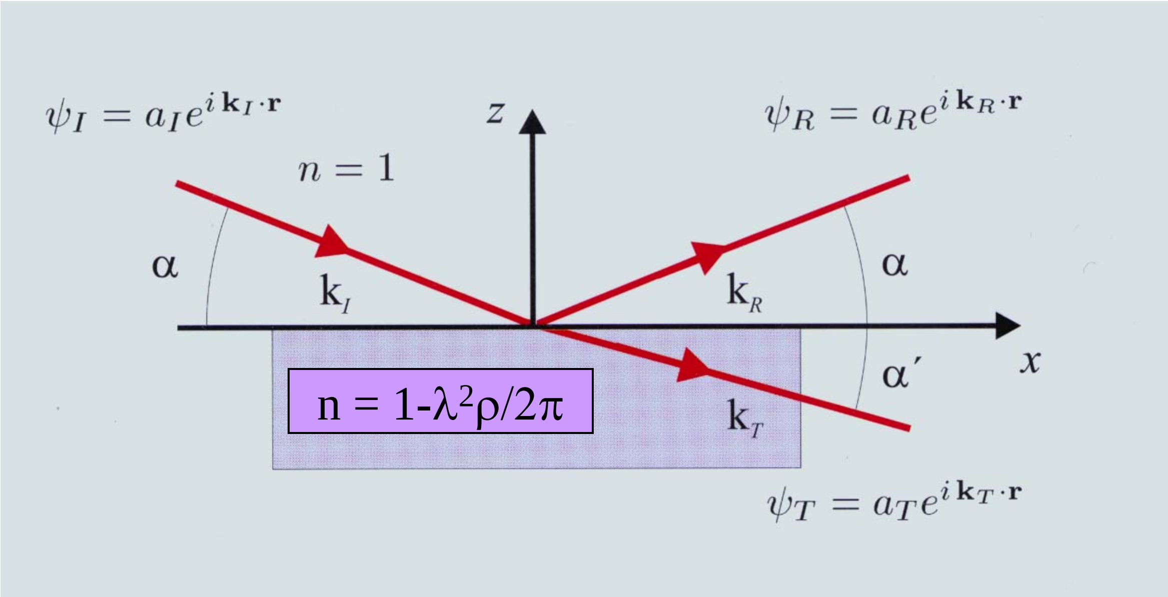
$\alpha_{\mathrm{critical}}(^o) \approx 0.02\lambda(\text{Å})$ for quartz

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

and $n = \sqrt{1 - \rho\lambda^2/\pi} \approx 1 - 0.005\lambda^2$ for Ni



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: $\cos\alpha = n\cos\alpha'$

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

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

Measured Reflectivity

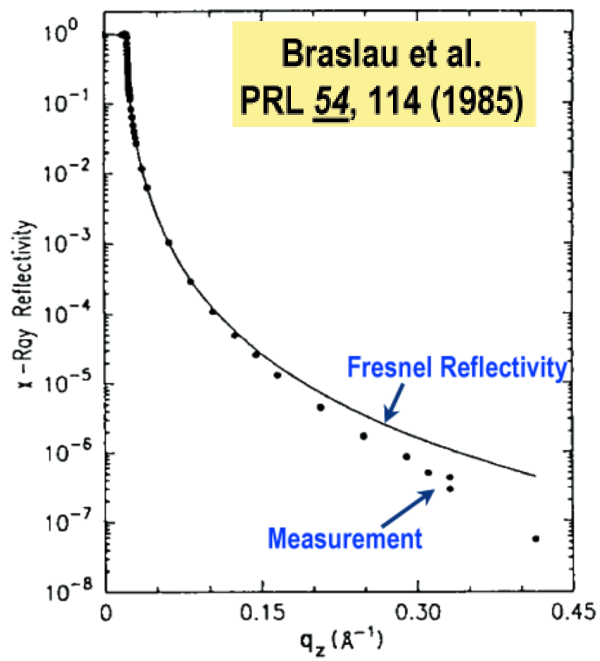
- We do not measure the reflectance, r, but the reflectivity, R given by:

  $$R = \frac{\text{\# of neutrons reflected at } Qz}{\text{\# of incident neutrons}} = r.r^*$$

  i.e., just as in diffraction, we lose phase information

- Notice, also, that the measurement averages the reflectivity over the surface of the sample: i.e. measured reflectivity depends on

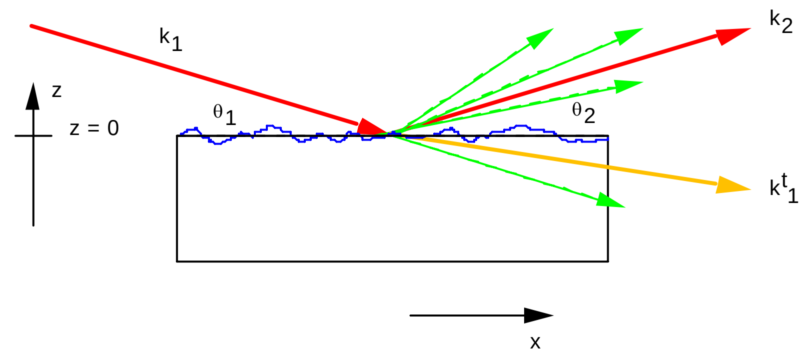
$$\overline{\rho}(z) = \frac{1}{S}\int dx \int dy \rho(x, y, z)$$



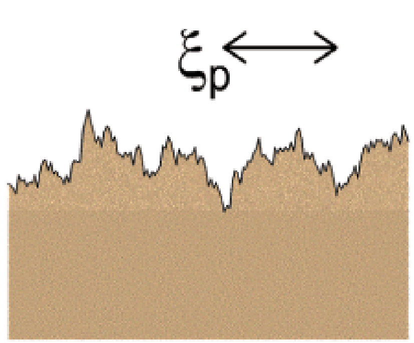
Measured and Fresnel reflectivities for water – difference is due to surface roughness

Surface Roughness

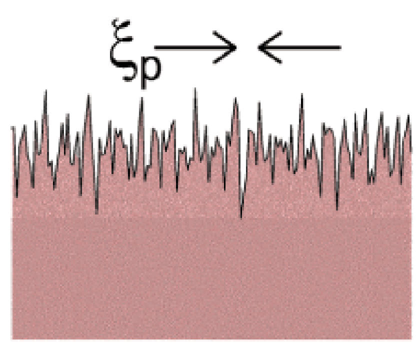
- Surface roughness causes diffuse (non-specular) scattering and so reduces the magnitude of the specular reflectivity



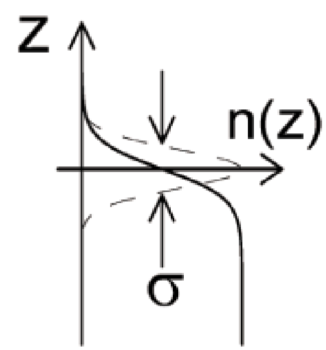
- The way in which the specular reflection is damped depends on the length scale of the roughness in the surface as well as on the magnitude and distribution of roughness



“sparkling sea”model -- specular from many facets



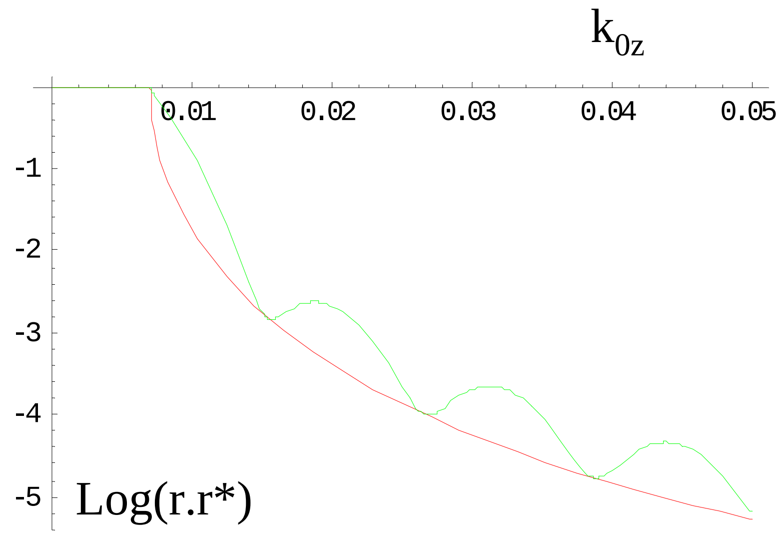
each piece of surface scatters independently -- Nevot Croce model



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

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

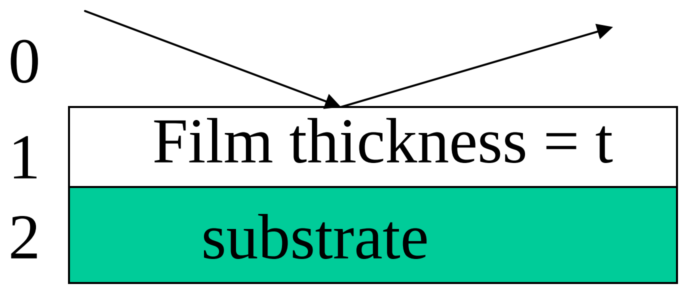
Fresnel's Law for a Thin Film



- $r=(k_{0z}-k_{1z})/(k_{1z}+k_{0z})$ 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 ~ π/t (a 250 A film was used for the figure)

Multiple Layers – Parratt Iteration (1954)

- The same method of matching wavefunctions and derivatives at interfaces can be used to obtain an expression for the reflectivity of multiple layers

$$X_j = \frac{R_j}{T_j} = e^{-2ik_{z,j}z_j} \frac{r_{j,j+1} + X_{j+1} e^{2ik_{z,j+1}z_j}}{1 + r_{j,j+1} X_{j+1} e^{2ik_{z,j+1}z_j}}$$

where $r_{j,j+1} = \dfrac{k_{z,j} - k_{z,j+1}}{k_{z,j} + k_{z,j+1}}$

Start iteration with

$R_{N+1} = X_{N+1} = 0$ and $T_1 = 1$

(i.e. nothing coming back from inside substrate & unit amplitude incident wave)

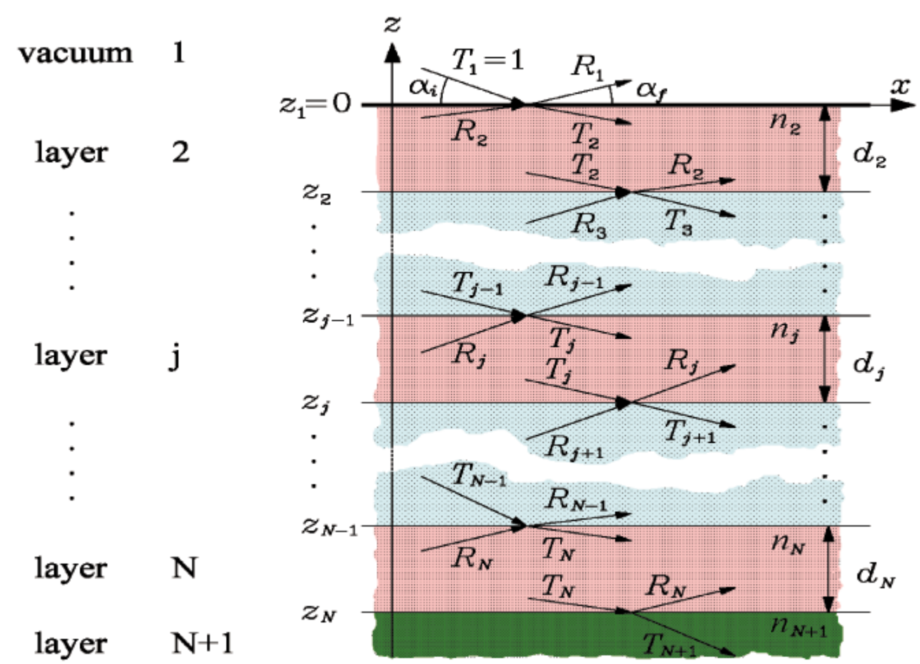


Image from M. Tolan

Dealing with Complex Density Profiles

- Any SLD depth profile can be "chopped" into slices
- The Parratt formalism allows the reflectivity to be calculated
- A thickness resolution of 1 Å is adequate – this corresponds to a value of Q_z where the reflectivity has dropped below what neutrons can normally measure
- Computationally intensive!!

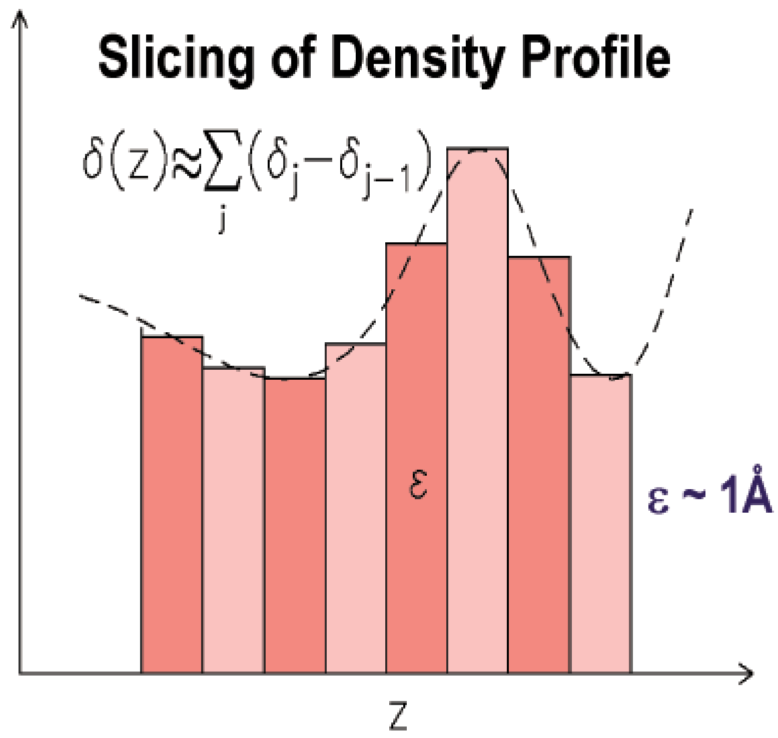
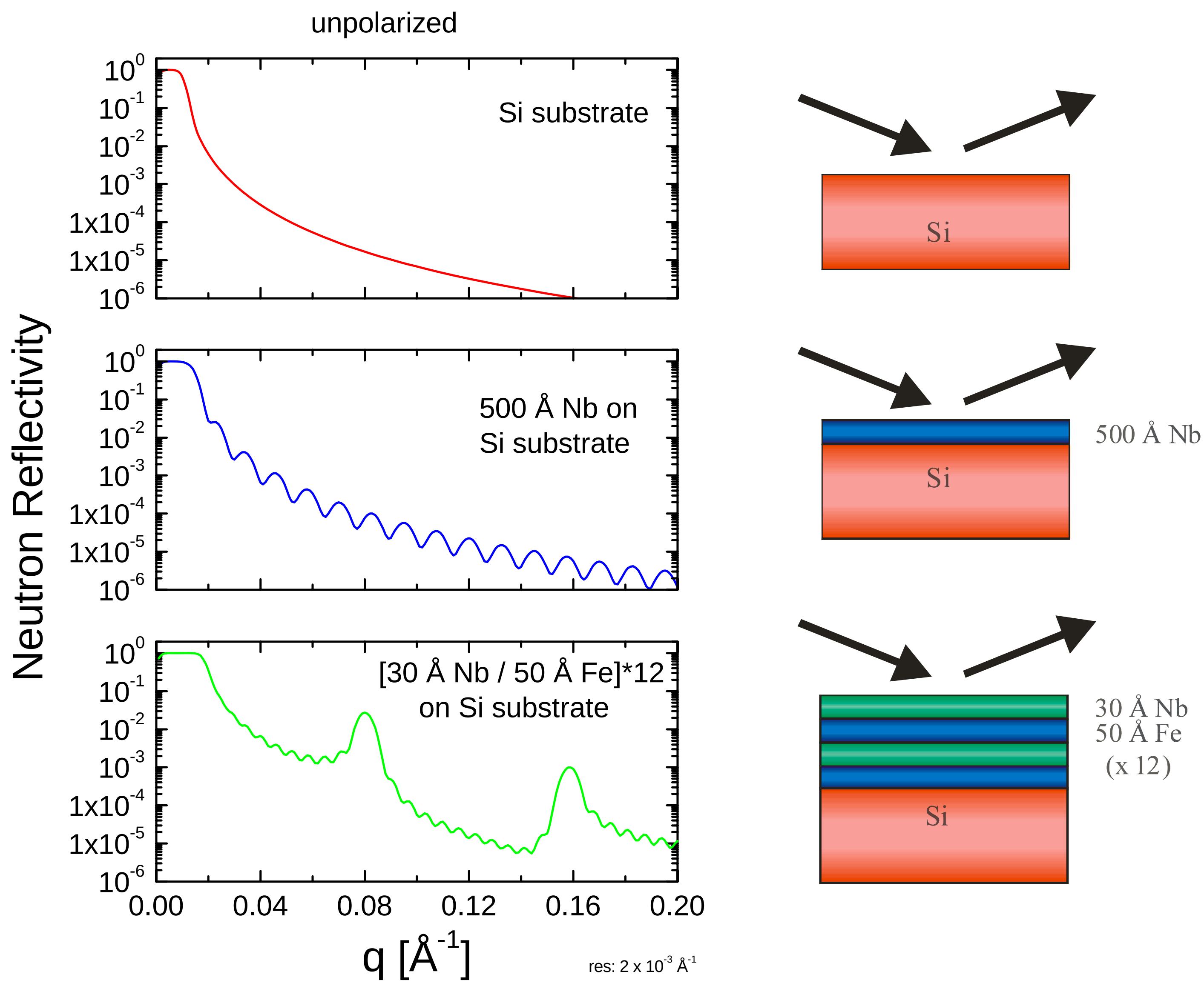


Image from M. Tolan

Reflectivity of Layered Structures



The Goal of Reflectivity Measurements Is to Infer a Density Profile Perpendicular to a Flat Interface

- In general the results are not unique, but independent knowledge of the system often makes them very reliable
- Frequently, layer models are used to fit the data
- Advantages of neutrons include:
  - Contrast variation (using H and D, for example)
  - Low absorption – probe buried interfaces, solid/liquid interfaces etc
  - Non-destructive
  - Sensitive to magnetism
  - Thickness length scale 10 – 5000 Å
- Issues include
  - Generally no unique solution for the SLD profile (use prior knowledge)
  - Large samples (~10 cm^2) with good scattering contrast are needed

Magnetic Properties of the Neutron

- The neutron has a magnetic moment of -9.649 x 10^{-27} JT^{-1}

$$\vec{\mu}_n = -\gamma \mu_N \vec{\sigma}$$

where $\mu_N = e\hbar / 2m_p$ is the nuclear magneton,

$m_p =$ proton mass, $e =$ proton charge and $\gamma = 1.913$

$\vec{\sigma}$ is the Pauli spin operator for the neutron. Its eignevalues are ± 1

- Note that the neutron's spin and magnetic moment are antiparallel
- Because of its magnetic moment, the neutron feels a potential given by:

$$V_m(\vec{r}) = -\vec{\mu}_n . \vec{B}(\vec{r}) \quad \text{where} \quad \vec{B}(\vec{r}) = \mu_0 \mu \vec{H}(\vec{r}) = \mu_0 [\vec{H}(\vec{r}) + \vec{M}(\vec{r})]$$

- Thus the neutron senses the distribution of magnetization in a material

Magnetic Scattering of the Neutron

- For nuclear scattering, the matrix element that appears in the expression for the scattering cross section is: $\sum_j b_j e^{i\vec{Q}.\vec{R}_j}$

- The equivalent matrix element for magnetic scattering is:

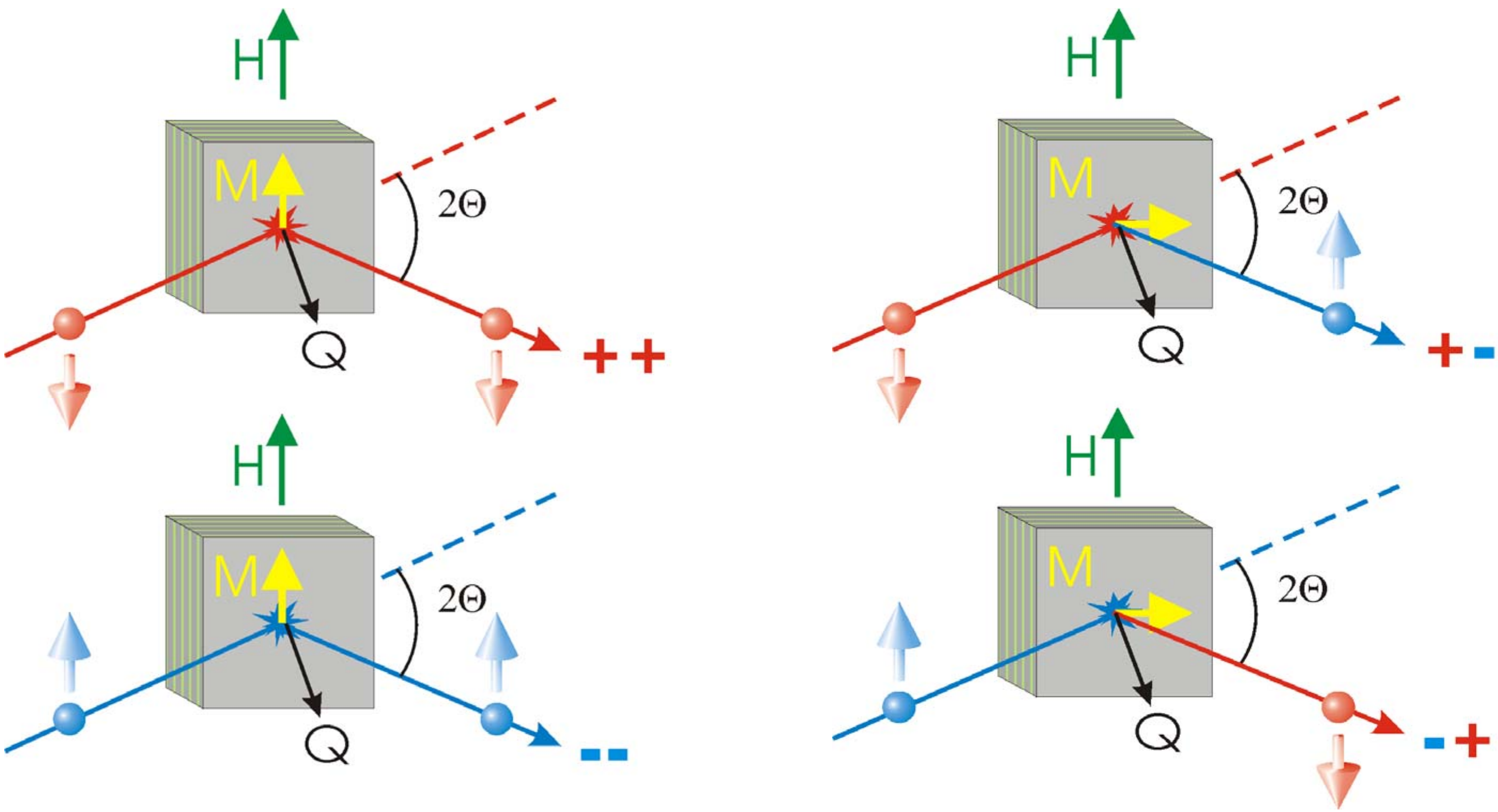
$$\gamma r_0 \frac{1}{2\mu_B} \vec{\sigma}.\vec{M}_\perp(\vec{Q}) \quad \text{where } \mu_B = \frac{e\hbar}{2m_e} \text{ is the Bohr magneton } (9.27 \times 10^{-24} \text{ JT}^{-1})$$

$$\text{and } r_0 = \frac{\mu_0}{4\pi} \frac{e^2}{m_e} \text{ is classical radius of the electron } (2.818 \times 10^{-6} \text{ nm})$$

- Here $\vec{M}_\perp(\vec{Q})$ is the component of the Fourier transform of the magnetization that is perpendicular to the scattering vector $\vec{Q}$. This form arises directly from the dipolar nature of the magnetic interaction.

- Unlike the neutron-nucleus interaction, the magnetic interaction of the neutron with a scattering system specifically depends on neutron spin

Polarized Neutron Reflection
Note: Arrows Represent Neutron Moments not Spins



Non-Spin-Flip

++ measures b + M_z

- - measures b - M_z

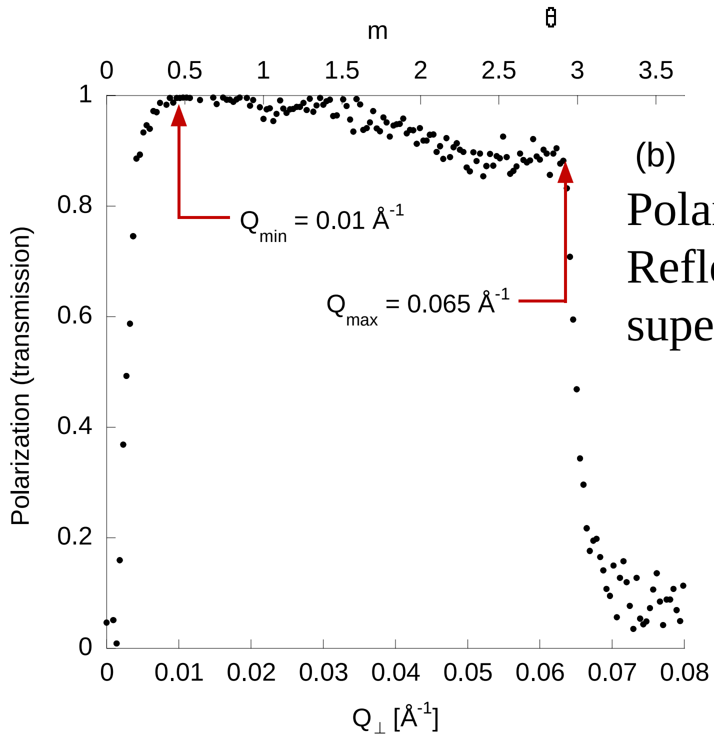
Spin-Flip

+- measures $M_x + i\,M_y$

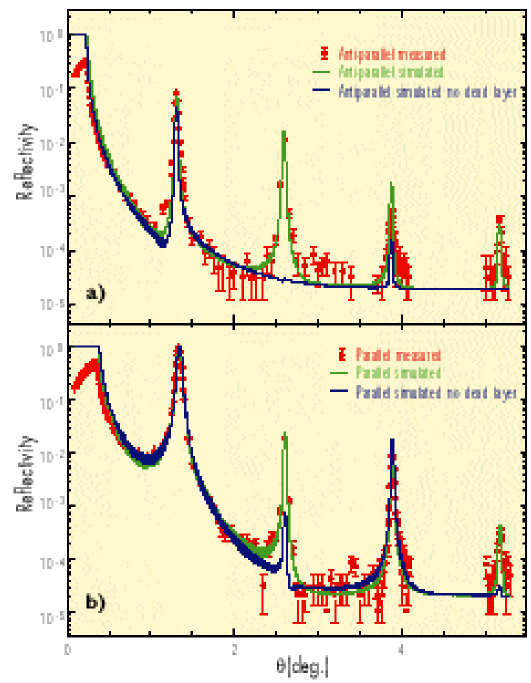
-+ measures $M_x – i\,M_y$

Magnetic Multilayers and Supermirrors

- By depositing many thin layers of contrasting SLD on a substrate we can make a Bragg reflecting device
  - With magnetic material in alternate layers, Bragg reflections can be polarized
- By tuning the layer thicknesses we can produce multiple Bragg peaks just above the mirror critical edge, effectively increasing the critical angle – called a supermirror
  - Can be made to polarize neutrons



Polarization of neutrons Reflected from an Fe-Si supermirror



50-bilayer Fe/Si multilayer

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