Magnetic Neutron Reflectometry

Moses Marsh
Shpyrko Group
9/14/11
Outline

• Scattering processes
• Reflectivity of a slab of material
• Magnetic scattering
• Off-specular scattering
• Source parameters
• Comparison with x-rays
Neutrons are waves

- de Broglie wavelength: \( \lambda = \frac{h}{p} = \frac{h}{mv} \)
  - Example: \( \lambda = 2\text{Å}, \ v = 1978 \frac{m}{s}, \)
    \[ E = \frac{mv^2}{2} = 20.5 \text{ meV} \]
- Plane wave: \( \psi \sim e^{ik \cdot r}, \ k = \frac{2\pi}{\lambda} \)
Scattering from a single fixed nucleus

- Incident plane wave: \( \psi \sim e^{ikx} \)
- Potential from nucleus: \( V(r) = \frac{2\pi \hbar^2}{m} b \delta(r) \)
- Outgoing spherical wave \( \psi \sim -\frac{b}{r} e^{ikr} \)
- Differential cross section: \( \frac{d\sigma}{d\Omega} = b^2, b = \text{scattering length} \)
Intrinsic Cross Section: Neutrons

Source: Sinha lecture
Reflectometry: Neutrons in media

- Neutrons incident on a slab of material at grazing angle
- Average scattering length density:
  \[ \rho = \sum_i N_i b_i \]
  - Neutrons sample average in-plane density
  \[ \rho(z) = \langle \rho(x, y, z) \rangle_{xy} \]
- Elastic scattering
  - Energy in vacuum: \[ E = \frac{\hbar^2 k_0^2}{2m} \]
  - Energy in medium: \[ E = \frac{\hbar^2 k^2}{2m} + V = \frac{\hbar^2}{2m} (k^2 - 4\pi \rho) \]
- Index of refraction:
  \[ n = \frac{k}{k_0} = 1 - \frac{4\pi}{k_0^2} \rho = 1 - \frac{\lambda_0^2}{2\pi} \rho \]
- \( \rho \) is typically on the order of \( 10^{-6} \text{Å}^{-2} \)
Fresnel Reflectivity

- Specular reflection:
  \[ \alpha_r = \alpha_i \]
  \[ Q = 2k_{iz}\hat{z} = \frac{4\pi}{\lambda} \sin(\alpha_i)\hat{z} \]

- Reflection coefficient: ratio of reflected amplitude to incident amplitude
  \[ r = \frac{k_{iz} - k_{tz}}{k_{iz} + k_{tz}} = \frac{1 - \sqrt{1 - \left(\frac{Q_c}{Q}\right)^2}}{1 + \sqrt{1 - \left(\frac{Q_c}{Q}\right)^2}} \]

- Reflectivity:
  \[ R = |r|^2 \approx \left(\frac{Q_c}{2Q}\right)^4 \text{ for } Q \gg Q_c \]
  \[ Q_c^2 = 16\pi\rho \]
Fresnel Reflectivity

\[ R(Q) \sim Q^{-4} \]
Thin film on a substrate: Kiessig fringes

\[ d \left\{ \begin{array}{c} \rho_1 \\ \rho_2 \end{array} \right. \]

\[ \frac{2\pi}{d} > \rho_1 > \rho_2 \]

\[ \log \text{Reflectivity} \]

\[ Q (\text{Å}^{-1}) \]
Multilayer: Parratt formula

- Net reflectivity at the surface can be calculated *exactly* by enforcing continuity at each interface
- Dynamical: accounts for transmission and reflection at each interface
Reflectivity from an arbitrary $\rho(z)$

- Divide up $\rho(z)$ into slices of constant $\rho$ and apply Parratt formula
  - e.g.

- Or: apply Born approx. (kinematical treatment)

$$r(Q) \approx \frac{4\pi}{iQ} \int_{-\infty}^{\infty} \rho(z)e^{iQz} \, dz$$

- Assumes scattering is weak. Fails near $Q_c$ where reflectivity approaches unity (strong scattering)
Inverting Reflectivity Data

• How do you get $\rho(z)$ from $R(Q)$?
  – Start with a good model and refine parameters
  – Inverse Fourier transform: phase can be recovered by adding reference layers of known density to the front & back of the film (Majkrzak 1995)
Magnetic reflectometry

• A magnetic field adds a spin-dependent potential to the Schrödinger equation

\[ V_m = -\mu \cdot B = -\mu \sigma \cdot B \]

– \( \mu \) is the neutron magnetic moment
– \( \sigma \) has the Pauli spin matrices as components

• We now have to treat the incoming plane wave as a spinor: \( \psi = \left( \begin{array}{c} \psi_+ \\ \psi_- \end{array} \right) \)

– Polarizers and analyzers allow control of which spin hits the sample as well as which spin is detected
SNS BL4a Schematic

collimating apertures

polarizer

spin flipper

magnetic surface

spin flipper

analyzer
detector
Magnetic reflectometry

- Let $z$ be the spin quantization axis, and let $x$ lie along $Q$.
- Only the components of $B$ perpendicular to $Q$ contribute to scattering.
- This gives rise to a set of coupled Schrödinger equations for $\psi_+$ and $\psi_-$

\[
[\partial_x^2 + k_{0x}^2 - 4\pi \rho_{++}]\psi_+ - 4\pi \rho_{+-}\psi_- = 0
\]
\[
[\partial_x^2 + k_{0x}^2 - 4\pi \rho_{--}]\psi_- - 4\pi \rho_{-+}\psi_+ = 0
\]

\[
\rho_{\pm\pm} = \rho_n \pm \frac{m}{2\pi\hbar^2} \mu B_z ; \quad \rho_{\pm\mp} = \frac{m}{2\pi\hbar^2} \mu (B_x \mp iB_y)
\]
Magnetic reflectometry

- $\rho_{+-}$ and $\rho_{-+}$ cause spin flips during scattering
- If $B$ is parallel to $\hat{z}$, then the equations decouple and there is no spin flipping. Then we can treat each polarization separately.
- For arbitrary $B(z)$ we can solve for the reflectivity numerically using an approach similar to the Parratt formalism (Felcher 1987)
- Corrections must be applied for incomplete beam polarization
Magnetic reflectometry

- Simulations: alternating Fe/Cr layers, each 4nm thick. 20 layers total.
- Magnetism only in Fe layers
Non-specular reflection

- Diffuse scattering from interfacial roughness (Sinha 1988)
- Off specular peaks from in-plane order
  - Also Yoneda peaks near the critical angle, where the transmission coefficient is maximized
- Sensitive to components of \( \mathbf{B} \) normal to plane

Zabel 1994
What about x-rays?

- Brighter, more coherent sources
- Element specificity
- Unaffected by external magnetic fields
- Lower penetration depth
- Insensitive to light elements
- Magnetic scattering much weaker than charge scattering
Spallation Neutron Source (ORNL)

- Proton accelerated into mercury target, neutrons knocked loose
- Neutrons come out in pulses (60 Hz)
- Moderator gives them a thermal distribution of velocities
- Detector uses time of flight methods to determine $Q$
SNS BL4a Fact Sheet

- Wavelength range: $1.8\text{Å} < \lambda < 14.0\text{Å}$
- Q range: $0 < Q < 0.4\text{Å}^{-1}$
- Minimum reflectivity: $10^{-8}$
- Magnetic field max
  - $1.2\ T$ with a gap of 50 $mm$
  - $3\ T$ with a gap of 10 $mm$
- Temperature range: $5 - 750\ K$
References


• Lectures by Majkrzak, Sinha, Pynn
UHV system for in-situ polarized reflectometry experiments on ultrathin magnetic films (for clarity reasons the sample magnet is suppressed).
Scattering from many nuclei

- $q = k' - k$
- $\frac{d\sigma}{d\Omega} = \sum_{ij} b_i b_j e^{i q \cdot (r_i - r_j)}$
- Ensemble average: $\frac{d\sigma}{d\Omega} = \langle b \rangle^2 S(q)$
- $S(q) = \langle \sum_{ij} e^{i q \cdot (r_i - r_j)} \rangle = \langle \hat{\rho}_n(q) \hat{\rho}_n^*(q) \rangle$
- $\hat{\rho}_n(q)$ is the Fourier transform of nuclear density