introduction to
reflectometry
thin films
and
hetero structures
outline

- heterostructures
  - magnetic layers
  - membrane systems

- reflectometry
  - (few formulae)

- ... derivation
  - (lots of formulae)

- experimental examples
  - Fe/Si
  - FeSi/GaAs interfaces
  - bio-membrane

- relevance for imaging
  - YES, there is some!
outline

• heterostructures
  → magnetic layers
  → membrane systems

• reflectometry
  → (few formulae)

• ... derivation
  → (lots of formulae)

• experimental examples
  → Fe/Si
  → FeSi/GaAs interfaces
  → bio-membrane

• relevance for imaging
  → YES, there is some!
magnetic films and heterostructures

...the damn magnetically dead layers...

- down-scaling ⇒ thin magnetic films
e.g. magnetic data storage

- spin polarised electron injection
e.g. spin-injection in a spin-transistor

- conflicting properties at interfaces
e.g. interface ferro – magnet superconductor

[Diagram of spin polarised electron injection]

[Image of magnetic films and heterostructures]
conflict of interests at superconductor / ferromagnet interfaces

(1) no interaction:
magnetic heterostructures

conflict of interests at superconductor / ferromagnet interfaces

(2) suppression of magnetism:
magnetic heterostructures

conflict of interests at superconductor / ferromagnet interfaces

(3) reality: induced magnetism within SC!

Habermaier, Physica C 364, 298 (2001); Holden, PRB 69, 064505 (2004); Stahn, PRB 71, 140509(R) (2005)
compression of self-organising polyglycerol-ester films

model-system for foams used for stabilising food products e.g. yogurt
trough to investigate membranes at the liquid/air interface
- heterostructures
  → magnetic layers
  → membrane systems

- reflectometry
  → (few formulae)

- ... derivation
  → (lots of formulae)

- experimental examples
  → Fe/Si
  → FeSi/GaAs interfaces
  → bio-membrane

- relevance for imaging
  → YES, there is some!
analogy to visible light

*flat* surfaces partly reflect light
→ picture of the boot

some media also transmit light
→ ground below the water

parallel interfaces
→ colourful soap bubbles

\[ q_Z = 2|k_0| \sin \omega \]

\[ |k| = \frac{2\pi}{\lambda} \]
measurement schemes

\[ R = R(q_z) = R(\lambda, \omega) \quad q_z = 4\pi \frac{\sin \omega}{\lambda} \]

angle-dispersive set-up

variation of \( \omega \) with fixed \( \lambda \)
detection under \( 2\omega \)

energy-dispersive set-up

variation of \( \lambda \) with fixed \( \omega \)
detection via time-of-flight
angle-dispersive set-up

neutron reflectometer

instrument: Morpheus at SINQ
**sample environment**

cooling with a *closed cycle refrigerator*  
$8 \text{ K} < T < 300 \text{ K}$

application of an external magnetic field with  
*Helmholtz coils*  
$-1000 \text{ Oe} < H < 1000 \text{ Oe}$

*sample*  
	tilt- and translation stages for alignment
data acquisition

typical quantities:
- angular range $0^\circ \ldots 10^\circ$
- $\lambda$ range $3\,\text{Å} \ldots 15\,\text{Å}$
- measurement time $10\,\text{min} \ldots 12\,\text{h}$

example:

Fe/Si multilayer on glass
polarised neutrons
$1\,\text{h per spin state}$
from the sample to $\rho(z)$
**outline**

- heterostructures
  - → magnetic layers
  - → membrane systems

- reflectometry
  - → (few formulae)

- ... derivation
  - → (lots of formulae)

- experimental examples
  - → Fe/Si
  - → FeSi/GaAs interfaces
  - → bio-membrane

- relevance for imaging
  - → YES, there is some!
analogy to visible light

flat surfaces partly reflect light
→ picture of the boot

some media also transmit light
→ ground below the water

parallel interfaces
→ colourful soap bubbles

scientist’s explanation:

- index of refraction,
- Fresnel reflectivity,
- transmittance,
- interference,
- bla bla bla . . .
plane wave in a medium $i$:

\[
\frac{\hbar^2}{2m} \frac{d^2}{dr^2} A e^{ik_i r} + (E - V_i) A e^{ik_i r} = 0
\]

\[
\frac{\hbar^2}{2m} (-k_i^2) e^{ik_i r} + (E - V_i) e^{ik_i r} = 0
\]

\[\Rightarrow k_i^2 = (E - V_i) \frac{2m}{\hbar^2}\]

\[n_i^2 = \frac{k_i^2}{k_0^2}\]

\[= \frac{E - V_i}{E}\]

by definition

\[n_i = \sqrt{1 - V_i/E}\]

\[\approx 1 - V_i/2E\]

for $V_i \ll E$

\[:= 1 - \delta\]

\[n_i - 1 \propto V_i\]

\[\Rightarrow \text{what is } V_i?\]
**What is $V_i$ for x-rays?**

Interaction $\gamma / \text{electron}$

(Off-resonance)

\[
V^e = \frac{2\pi \hbar^2}{m_e \text{vol}} \sum_i Z_i
\]

\[
= \frac{2\pi \hbar^2}{m_e} r_e \rho^e
\]

With:

- $Z_i = \text{electron number of atom } i$
- $r_e = \text{electron radius}$
- $m_e = \text{electron mass}$

\[
\delta = \frac{\lambda^2}{2\pi} r_e \rho^e
\]

With absorption: complex $n$

\[
n = 1 - \delta - i\beta
\]

At resonances:

\[
\delta = \delta(B)
\]
what is $V_i$ for neutrons?

interaction neutron / nucleus $j$

with $\lambda \gg r_{\text{nucleus } j}$

interaction neutron magnetic moment $\mu$

/ magnetic induction $B$

$\mu \uparrow \uparrow B \Rightarrow V^m = +\mu B$

$\mu \uparrow \downarrow B \Rightarrow V^m = -\mu B$

$\mu \perp B \Rightarrow$ spin-flip scattering

reflectometry 19

$V_j^\text{Fermi} = b_j \frac{2\pi \hbar^2}{m_n} \delta(r)$

$V^n = \frac{1}{\text{vol}} \int_j V_j^\text{Fermi} \, dr$

$= \frac{2\pi \hbar^2}{m_n} \frac{1}{\text{vol}} \sum_j b_j$

$:= \frac{2\pi \hbar^2}{m_n} \rho^b$

$V^m = \mu B_{\perp}$

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

$m_n = \text{neutron mass}$
what is the reflected intensity?

assumptions:
– one interface, only
– ideally flat and sharp
– homogeneous in x and y
⇒ only normal (z) components are relevant

continuity conditions for a plane wave impinging on the interface \(i, i+1\):

\[
\psi_{z,i} = \psi_{z,i+1}
\]

\[
\frac{d}{dz}\psi_{z,i} = \frac{d}{dz}\psi_{z,i+1}
\]

with

\[
\psi_{z,j} = A_j^\uparrow e^{ik_{z,j}z} + A_j^\downarrow e^{-ik_{z,j}z}
\]

\[
k_{z,j} = k_j \sin \omega_j = n_j k_0 \sin \omega_j
\]

reflectance

\[
r_{i,i+1} = \frac{A_i^\uparrow}{A_i^\downarrow}
\]

\[
= \frac{n_i \sin \omega_i - n_{i+1} \sin \omega_{i+1}}{n_i \sin \omega_i + n_{i+1} \sin \omega_{i+1}}
\]
Fresnel reflectivity

reflectance

\[ r_{i,i+1} = \frac{n_j \sin \omega_i - n_{i+1} \sin \omega_{i+1}}{n_j \sin \omega_i + n_{i+1} \sin \omega_{i+1}} \]

for \( \omega_{i+1} > 0 \)

transmittance

\[ t_{i,i+1} = \frac{2 n_j \sin \omega_i}{n_j \sin \omega_i + n_{i+1} \sin \omega_{i+1}} \]

for \( \omega_{i+1} > 0 \)

air/solid interface

\[ q_z = 2 \cdot \frac{2\pi}{\lambda} \cdot \sin \omega \]

for \( q_z > q^c \)

\[ r_{0,1} = \frac{1 - \sqrt{1 - (q^c/q_z)^2}}{1 + \sqrt{1 - (q^c/q_z)^2}} \]

\[ R(q_z) = |r_{0,1}(q_z)|^2 \]
reflected intensity of a multilayer

several parallel interfaces:
interference of all waves

\[ R(q_z) = |r(q_z)|^2 \]

what is \( r(q_z) \) of a multilayer?


\[
\Psi_0(0) = \begin{pmatrix}
A_0^{\uparrow} \\
A_0^{\downarrow}
\end{pmatrix}
\]

free choice of phase

\[
= \begin{pmatrix}
1/t_{0,1} & r_{0,1}/t_{0,1} \\
r_{0,1}/t_{0,1} & 1/t_{0,1}
\end{pmatrix}
\begin{pmatrix}
A_1^{\uparrow} \\
A_1^{\downarrow}
\end{pmatrix}
\]

continuity condition

\[
= I_{0,1} \begin{pmatrix}
e^{ik_{z,1}d_1} & 0 \\
0 & e^{-ik_{z,1}d_1}
\end{pmatrix}
\begin{pmatrix}
A_1^{\uparrow} e^{-ik_{z,1}d_1} \\
A_1^{\downarrow} e^{ik_{z,1}d_1}
\end{pmatrix}
\]

phase factor

\[
= I_{0,1} T_1 \begin{pmatrix}
1/t_{1,2} & r_{1,2}/t_{1,2} \\
r_{1,2}/t_{1,2} & 1/t_{1,2}
\end{pmatrix}
\begin{pmatrix}
A_2^{\uparrow} e^{-ik_{z,1}d_1} \\
A_2^{\downarrow} e^{ik_{z,1}d_1}
\end{pmatrix}
\]

\[
= I_{0,1} T_1 I_{1,2} \begin{pmatrix}
e^{ik_{z,2}d_2} & 0 \\
0 & e^{-ik_{z,2}d_2}
\end{pmatrix}
\begin{pmatrix}
A_2^{\uparrow} e^{-ik_{z,2}(d_1+d_2)} \\
A_2^{\downarrow} e^{ik_{z,2}(d_1+d_2)}
\end{pmatrix}
\]

\[\vdots\]

\[= M \begin{pmatrix}
A_{\text{substr}}^{\uparrow} e^{-ik_{z,\text{substr}} \sum_i d_i} \\
A_{\text{substr}}^{\downarrow} e^{ik_{z,\text{substr}} \sum_i d_i}
\end{pmatrix}\]
What is $r(q_z)$ of a multilayer?

$$
\psi_0(0) = \begin{pmatrix} A_0^\uparrow \\ A_0^\downarrow \end{pmatrix}
= M \begin{pmatrix} 0 \\ A_{\text{substr}}^\downarrow e^{i k_{z,\text{substr}} \sum_i d_i} \end{pmatrix}
$$

$$
r(q_z) = A_0^\uparrow / A_0^\downarrow
= \frac{M_{12} A_{\text{substr}}^\downarrow e^{i k_{z,\text{substr}} \sum_i d_i}}{M_{22} A_{\text{substr}}^\downarrow e^{i k_{z,\text{substr}} \sum_i d_i}}
= \frac{M_{12}(q_z)}{M_{22}(q_z)}
$$

The calculation of $M_{12}(q_z)$ and $M_{22}(q_z)$ is trivial . . .

. . . if all $n_i$ and $d_i$ are known!
reflected intensity of a multilayer

\[ R(q_z) = |r(q_z)|^2 \]

⇒ all phase information is lost

⇒ one way road:

⇒ calculation of \( R(q_z) \) using a model

and

comparison to measured curve(s)

real effects
to be taken into account:

– non-sharp interfaces
– inhomogeneous layers
– illumination of the sample
– resolution of the set-up

\( \Delta \omega, \Delta \lambda \)
simulated reflectivity

...of a surface

Critical edge $\Rightarrow V_{\text{substrate}}$

Snell's law: \[
\frac{\cos \omega_i}{\cos \omega_{i+1}} = \frac{n_{i+1}}{n_i}
\]

$\Rightarrow \cos \omega_c^0 = n_1$
simulated reflectivity

... of a thin layer

\[ \log_{10}[R(\omega)] \]

amplitude \( \Rightarrow \Delta V_{\text{layer,substrate}} \)

minimum \( \Rightarrow d_{\text{layer}} \)
simulated reflectivity

...of a thick layer

\[
\log_{10}[R(\omega)]
\]

amplitude \( \Rightarrow \Delta V_{\text{layer, substrate}} \)

minima \( \Rightarrow d_{\text{layer}} \)
simulated reflectivity

... of a periodic multilayer

\[
\log_{10}[R(\omega)]
\]

maxima $\Rightarrow d_{\text{bilayer}}$

amplitudes $\Rightarrow \Delta V_{\text{layers}}$

$\Rightarrow \Delta d_{\text{layers}}$

minima $\Rightarrow d_{\text{film}}$

\[\omega/\text{deg}\]

\[0\ 1\ 2\ 3\ 4\ 5\]
\[ \delta = 1 - n = \frac{\lambda^2}{2\pi} (\rho^b + \rho^m) \text{ for neutrons} \]
\[ = \frac{\lambda^2}{2\pi} r_e \rho^e \text{ for x-rays} \]

Ni: \( \rho^b = 9.4 \cdot 10^{-6} \text{ Å}^{-2} \)
\[ \Rightarrow \delta^{\text{nuc}} = 3.75 \cdot 10^{-5}, \lambda = 5 \text{ Å} \]
\[ \Rightarrow \omega^c \approx 0.5^\circ \]
\( \delta \ll 1 \)
small angles of incidence!

Fe: \( \rho^m \approx 6 \cdot 10^{-6} \text{ Å}^{-2} \)
\[ \Rightarrow \delta^m \approx 2.4 \cdot 10^{-5}, \lambda = 5 \text{ Å} \]
\( \delta^m \sim \delta^b \)

Al: \( r_e \rho^e = 2.2 \cdot 10^{-5} \text{ Å}^{-2} \)
\[ \Rightarrow \delta^e = 8.7 \cdot 10^{-5}, \lambda = 5 \text{ Å} \]
\( \delta^e \sim \delta^b \)
<table>
<thead>
<tr>
<th>some numbers</th>
<th>reflectometry 31</th>
</tr>
</thead>
<tbody>
<tr>
<td>probed depth</td>
<td>100 nm → 1 µm</td>
</tr>
<tr>
<td>(less for strong absorbers)</td>
<td></td>
</tr>
<tr>
<td>depth resolution</td>
<td>0.2 nm → 400 nm</td>
</tr>
<tr>
<td>strongly model dependent</td>
<td></td>
</tr>
<tr>
<td>$t$ and $\delta$ might be strongly correlated</td>
<td></td>
</tr>
<tr>
<td>lateral coherence</td>
<td>1 µm → 100 µm</td>
</tr>
<tr>
<td>averaging laterally over all microstructures</td>
<td></td>
</tr>
</tbody>
</table>
• heterostructures
  → magnetic layers
  → membrane systems

• reflectometry
  → (few formulae)

• ... derivation
  → (lots of formulae)

• experimental examples
  → Fe/Si
  → FeSi/GaAs interfaces
  → bio-membrane

• relevance for imaging
  → YES, there is some!
spintronics

use not only the electron charge to carry information but also its spin

e.g. transistor based on spin / FM alignment:

spin-polarised currents exist in *half-metals* (e.g. Fe$_3$Si)

but

polarised spin injection into a semiconductor (e.g. GaAs) is inefficient

⇒ what happens at the interface?
Fe$_3$Si film on GaAs
search for a magnetically dead layer

\[ \text{Al} 300 \text{ Å} \]
\[ \text{Fe}_3\text{Si} 100 \text{ Å} \]
\[ \text{GaAs} \infty \]

sample size: $5 \times 5 \text{ mm}^2$
measurement time: 24 h neutron
1 h x-ray
Fe$_3$Si film on GaAs
search for a magnetically dead layer

\[ B = 2.0(2) \, \text{T in Fe}_3\text{Si} \]

no magnetically dead layer detectable
Fe/Si multilayer

ideal case:

\[ \rho_{Fe}^b + \rho_{Fe}^m \gg \rho_{Si} \]
\[ \rho_{Fe}^b - \rho_{Fe}^m = \rho_{Si} \]
neutron polariser

Fe/Si multilayer

reality: interdiffusion leads to 5 Å thin magnetically dead Fe : Si layers

sample size: 70 × 50 mm²
measurement time: 1 h
compression of self-organising polyglycerol-ester films

H$_2$O substituted by D$_2$O
⇒ strong contrast between solvent and film (essentially [CH$_2$]$_n$)
⇒ high critical edge

constant film thickness
laterally more homogeneous
⇒ less roughness
⇒ lower damping of $R(q_z)$
polymer brushes

applications:
- anti-fouling color
- coatin within ball bearings
- matrix for chemical sensors

where are the functional groups located?

Schüwer, Macromolecules 44, 6868-6874 (2011)
chemical sensor

polymer brushes
location of functional groups

341 Å
non-functionalised

1016 Å
leucine functionalised

867 Å
serine functionalised
off-specular scattering

\[ \rho = \rho(x, z) \Rightarrow R(q_x \neq 0) \neq 0! \]

Ni/Ti multilayer (non magnetic!)

resolution in \( x \): \( \approx 0.01^\circ \)
resolution in \( y \): \( > 1^\circ \)
\( \Rightarrow \) integrated over \( y \)
outline

- heterostructures
  → magnetic layers
  → membrane systems

- reflectometry
  → (few formulae)

- ... derivation
  → (lots of formulae)

- experimental examples
  → Fe/Si
  → FeSi/GaAs interfaces
  → bio-membrane

- relevance for imaging
  → YES, there is some!
relevance for imaging

total reflection and refraction change beam direction

⇒ important for *large* sample-detector distances

also (optically) rough interfaces show significant total reflection!
transmission of a slightly tilted square prism: 
\( n < 1 \Rightarrow \) total external reflection possible 
parallel, monochromatic beam
relevance for imaging

transmission of a slightly tilted square prism:
- no refraction
- no reflection
relevance for imaging

transmission of a slightly tilted square prism:
- refraction
- total reflection
transmission of a slightly tilted square prism:
- refraction
- partial reflection
transmission of a slightly tilted square prism: some numbers

Al prism with $\varnothing = 10 \times 10 \text{ mm}^2$

$n = 1 - 8.3 \cdot 10^{-6}, (5 \text{ Å})$

$\omega^c = 0.233^{\circ}, (5 \text{ Å})$

⇒ reflection (and refraction) can lead to detectable features like halos or shadows
measured transmission (Eberhard Lehmenn, PSI)

$\lambda \in [1.8, 5] \, \text{Å}$

$\Delta \omega \approx 0.1^\circ$

$10 \times 10 \, \text{mm}^2$

100 mm

Al cube has not perfectly flat and parallel surfaces

$\Rightarrow \omega = 0 \pm \Delta \omega_{\text{beam}} \pm \Delta \omega_{\text{surface}}$
Reflectometry probes depth-profile of some potential averages laterally ⇒ ideal for layered systems data analysis by modelling

With neutrons resolution: atom to sub-\(\mu\)m isotope selective detects in-plane magnetic induction

With x-rays resolution: atom to sub-\(\mu\)m detects electron density

...in resonance detects magnetic states of atoms

Radiography might be affected !!!
reflectometry, in general:
  J. Daillant, A. Gibaud:
  *X-ray and Neutron Reflectivity*

  U. Pietsch, V. Holý, T. Baumbach:
  *High-Resolution X-Ray Scattering*
  (Springer 2004)

... on magnetic systems
  F. Ott:
  *Neutron scattering on magnetic surfaces*
  C. R. Physique 8, 763-776 (2007)

... using resonant x-rays
  S. Brück:
  *Magnetic Resonant Reflectometry on Exchange Bias Systems*
  Dissertation, Stuttgart 2009