

Magnetic surfaces and Thin Films (Polarized Neutron Reflectometry)

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Surfaces, Interfaces and Thin Films

For fundamental properties in the area of magneto- and spintronics



Exchange bias

F - pinned

Proximity effects and tunneling



Lateral structures







Surfaces, Interfaces and Thin Films

For fundamental properties in the area of magneto- and spintronics





Exchange bias







Content

1. X-ray and neutron reflectivity

2. Polarized neutron reflectivity (PNR)

- Methods
- Instrumentation
- Examples





1. X-ray and neutron reflectivity











Index of refraction



 $\sqrt{\varepsilon(\omega)} = n(\omega) = Brechungsindex \Rightarrow Optik$





X-ray refractive index

Refractive Index:

$$n^{2}(\omega) = 1 + \frac{4\pi\rho_{A}e^{2}}{m_{e}}\sum_{i}\frac{f_{i}}{\omega_{i}^{2} - \omega^{2} + i\gamma}$$

For x-ray's, the refractive index is always smaller than 1:

$$n(\lambda) = 1 - \frac{r_0}{2\pi} \rho_e \lambda^2$$
$$\approx 1 - 10^{-5} < 1$$

Adding dispersion and absorption correction:





Plot of δ and β for CoO





http://www-cxro.lbl.gov/optical_constants/getdb2.html 8



Snellius law for different media





Snellius law for different media



$$\frac{\cos\alpha}{\cos\alpha'} = n;$$





Critical angle for total reflection

Since n < 1, total reflection occurs at:

For all angles $\alpha < \alpha_c$ the wave can not penetrate into the medium, but at α_c there is an evanescent wave travelling along the interface







Critical angle for total reflection

Since n < 1, total reflection occurs at:

$$\frac{\cos\alpha_c}{\cos0} = \cos\alpha_c = n$$

For all angles $\alpha < \alpha_c$ the wave can not penetrate into the medium, but at α_c there is an evanescent wave travelling along the interface







Critical Scattering Vector

Scattering vector is defined as:

$$Q = \frac{4\pi}{\lambda} \sin \alpha = 2k \sin \alpha$$

Accordingly the critical scattering vector is:

$$Q_c = \frac{4\pi}{\lambda} \sin \alpha_c = 2k\sqrt{1 - \cos^2 \alpha_c} = \sqrt{4k^2(1 - n^2)}$$
$$\cong \sqrt{4k^2 2\delta} = \sqrt{16\pi r_0 \rho_e}$$

The critical scattering vector is no more a function of the wavelength. It is entirely determined by the property of the material and in particular by the electron density ρ_e .





Fresnel reflectivity

For $Q_z < Q_c$: R = 1, For $Q_z > Q_c$: R = R_f

The reflectivity drops with Q^4 , for scattering vectors $Q >> Q_c$. This applies for perfectly flat interfaces.







Fresnel reflectivity for Si



http://sergey.gmca.aps.anl.gov/





Reflectivity from a thin layer: Kiessig fringes







Reflectivity from a double layer









Reflectivity from a multilayer







Reflectivity and Bragg range







In-situ control of layer thickness and oxide growth



Layer thickness, electron density



Out -of-plane lattice constant, Number of coherently scattering lattice planes







Electron density profile from reflectivity data





Back transformation from reflectivity data to electron densities and thickness profiles is the ultimate goal. However, the back transformation is not always uniquely possible.









Smooth and rough surfaces







Smooth and rough surfaces







Smooth and rough surfaces







Reflectivity of rough surface

Master formular yields for a Gaussian roughness a damped Fresnel reflectivity:

$$R(Q_z) = R_F(Q_z) \exp(-Q_z^2 \sigma^2)$$

 $R_F(Q_z)$ is the Fresnel reflectivity of the ideal surface. Roughness adds a damping factor, similar to the Debye-Waller factor:







Off-specular diffuse scattering







Off-specular scattering from rough interfaces



Perfectly specluar surface, 100% reflection, mirror image



Perfectly rough surface, 100% diffuse scattering, projector wall



Partially reflecting and scattering from rough surface





Diffuse Scattering

Scattering function in the Born approximation:

$$S(\vec{Q}) = \int \langle \rho(0)\rho(R) \rangle e^{i\vec{Q} \cdot \vec{R}} d^3R$$

Pair correlation function:

$$G(\vec{R}) = \left\langle \left(\rho(0) - \left\langle\rho(0)\right\rangle\right) \left(\rho(\vec{R}) - \left\langle\rho(\vec{R})\right\rangle\right) \right\rangle$$
$$= \left\langle\rho(0)\rho(\vec{R})\right\rangle - \left\langle\rho(0)\right\rangle \left\langle\rho(\vec{R})\right\rangle$$
$$= \left\langle\rho(0)\rho(\vec{R})\right\rangle - \left\langle\rho(0)\right\rangle^{2}$$

Inserting:

$$S_{tot}(\vec{Q}) = \left\langle \rho(0) \right\rangle^2 \int e^{i\vec{Q} \cdot \vec{R}} d^3 R + \int C(\vec{R}) e^{i\vec{Q} \cdot \vec{R}} d^3 R$$

 $S_{spec}(\vec{Q})$ + $S_{diff}(\vec{Q})$

Specular Reflection Diffuse Scattering



Height-height correlation function



Height-height correlation function for a single self-affine, fractal surface:

$$C(R) = \langle z(0)z(R) \rangle = \sigma^2 \exp[-(R / \xi)]^{2h}$$

 σ = rms roughness

$$\xi = cut-off length:$$

for $R > \xi$, interface appears smooth,

for R < ξ , interface appears rough, fractal behavior

$$S_{diff}(\vec{Q}) = \frac{\exp(-Q_z^2 \sigma^2)}{Q_z^2} \times \int [\exp(Q_z^2 C(R)) - 1] \exp(iQ_{\parallel}R) d^2R$$



S.K. Sinha, E.B. Sirota, S. Garoff, and H.B. Stanley, Phys. Rev. B 38 2297 (1988)⁶



Specular and off-specular scattering





J. Als-Nielsen and Des McMorrow, Wiley, 2001



Transverse scans

Transverse scan from an FePt film on GaAs





A. Nefedov et al. J. Phys.: Condens. Matter 14, 12273 (2002)



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Refractive index for neutrons

Snell's law for specular reflection:



$$n = \frac{\sin \gamma_0}{\sin \gamma_t} = \frac{\left|\vec{k}_t\right|}{\left|\vec{k}_i\right|}$$





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$$n = \frac{\sin \gamma_0}{\sin \gamma_t} = \frac{\left|\vec{k}_t\right|}{\left|\vec{k}_i\right|}$$

QM potential step for the z-component of the kinetic energy:






Combining both

$$n^{2} = \frac{\sin^{2} \gamma_{0}}{\sin^{2} \gamma_{t}} = \frac{\left|\vec{k}_{t}\right|^{2}}{\left|\vec{k}_{i}\right|^{2}} = \frac{E_{t}}{E_{i}} = \frac{E_{i} - V_{n}}{E_{i}} = 1 - \frac{4\pi}{k_{i}^{2}} N_{A} b_{coh}$$

 N_A = nuclei number density

 b_{coh} = coherent scattering length of nuclei A Notice that n ≤ 1, only for $b_{coh} \ge 0$

Total reflection only for $b_{coh} \ge 0$





Example: Neutron reflectivity from a non-magnetic, infinite thick and flat sample



For $Q_z < Q_c$: R = 1, only for b >0, i.e. for coherent scattering length.





Neutron Reflectivity







Neutron Reflectivity



- Film thickness
 Interface roughness
- Density profiles







$$V = V_n \pm V_m = \frac{2\pi\hbar^2}{m} N_A (b_n \pm p_m)$$







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Reflectivity from thin FM film







2. Polarized neutron reflectivity







































Initial state and polarization of neutron out side of the sample:







Initial state and polarization of neutron out side of the sample:



State and polarization of neutron inside of the sample:







$$V_{m,Y}(z) = -\left|\vec{\mu}_n\right| \left|\vec{B}(z)\right|$$
$$= -4\pi \left|\vec{\mu}_n\right| \left|\vec{M}(z)\right|$$
$$= -\frac{2\pi\hbar^2}{m_n} p_m(z)$$

Initial state and polarization of neutron out side of the sample:



State and polarization of neutron inside of the sample:







$$V_{m,Y}(z) = -\left|\vec{\mu}_n\right| \left|\vec{B}(z)\right|$$
$$= -4\pi \left|\vec{\mu}_n\right| \left|\vec{M}(z)\right|$$
$$= -\frac{2\pi\hbar^2}{m_n} p_m(z)$$

Total neutron – sample potential (independent of the angle ϕ):

$$V = V_n \pm V_m = \frac{2\pi\hbar^2}{m_n} N_A (b_n \pm p_m)$$

Initial state and polarization of neutron out side of the sample:



State and polarization of neutron inside of the sample:







Schrödinger equation

Potential for polarized neutron scattering at magnetic samples:

$$V = \begin{pmatrix} V_{++} & V_{+-} \\ V_{-+} & V_{--} \end{pmatrix} = \frac{2\pi\hbar}{m_n} N_A \begin{pmatrix} b_{coh} + p_Y & p_X \\ p_X & b_{coh} - p_Y \end{pmatrix}$$

Inserting in Schrödinger equation:

$$\left(-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2}+V\right)\!\!\begin{pmatrix}\psi_+\\\psi_-\end{pmatrix}=E\!\begin{pmatrix}\psi_+\\\psi_-\end{pmatrix}$$

Yields the respective reflectivities R⁺⁺, R⁻⁻, R⁺⁻, R⁻⁺.





Reflectivity and Asymmetry

$$R^{+} = R^{++} + R^{+-}$$

$$R^{-} = R^{--} + R^{-+}$$

$$R^{+} - R^{-} = R^{++} - R^{--}$$

$$R^{+} + R^{-} = R^{++} + R^{--} + 2R^{+-}$$
Spin Asymmetry :

$$SA = \frac{R^{+} - R^{-}}{R^{+} + R^{-}} = \frac{R^{++} - R^{--}}{R^{++} + R^{--} + 2R^{+-}}$$





Spin asymmetry (SA), single surface







Reflectivity and Asymmetry for single thin ferromagnetic film (Fe)



F. Radu et al. submitted





Non-spin flip (NSF) scattering



NSF – scattering measures the Y-component of the magnetization vector: M_{Y} (longitudinal component)





Spin-flip (SF) scattering



SF – scattering measures the X-component of the magnetization vector: M_x (transverse component)





Vector Magnetometry











$$\phi = \arctan\left(\frac{M_y}{M_x}\right)$$
$$\left|\vec{M}\right| = \frac{M_y}{\sin\phi}$$





Selection rule for magnetic neutron scattering



Magnetization components parallel to the scattering vector are not visible to the neutrons!





Instrumention: two types





Instrumention: two types

- Angle
 dispersive
- monochromatic beam
- scanning of $\boldsymbol{\alpha}$







Instrumention: two types

- Angle
 dispersive
- monochromatic beam
- scanning of $\boldsymbol{\alpha}$
- Wavelength
 dispersive
- White beam
- TOF method,
- \bullet fixed α









Polarizer, analyzer, and spin flipper

transmission supermirror:







Polarizer, analyzer, and spin flipper







Schematics: neutron reflectometer with complete polarization analysis



This part is identical for angle dispersive and wavelength dispersive instruments





Schematics of an angle dispersive (fixed wavelength) reflectometer






The ADAM Reflectometer at the ILL



http://www.ill.fr/YellowBook/ADAM/





TOF - PNR - CRISP-ISIS









- a. Thin films
- b. Superlattices and roughness
- c. Exchange bias
- d. Magnetic patterns





PNR of 2 nm Fe film on 150 nm Nb on sapphire substrate



Amount of magnetic material: 10⁻³ emu





Nb cap 5nm
Fe 2 nm
Nb film 150 nm
Substrate





































Nuclear and magnetic density profile in (GaMn)As









Nuclear and magnetic density profile in (GaMn)As







Nuclear and magnetic density profile in (GaMn)As



G



PNR from model-spinstructures in magnetic multilayers







Reciprocal space maps in the small angle regime







Exchange coupling in [Fe_{0.43}Cr_{0.57}(24Å)/Cr(28Å)]



R. Siebrecht, et al. 2000





Exchange coupling in [Fe_{0.43}Cr_{0.57}(24Å)/Cr(28Å)]







Magnetic roughness in Co/Cu superlatties



S. Langridge, J. Schamlian, C.H. Marrows, D.T. Dekadjevi, B.J. Hickey, PRL 85, 4964 (2000).





Transverse scans across half-order AF peak



$$S_{Diff}(Q) = DW \int d^2 \vec{r} e^{i\vec{Q}_{\parallel}\cdot\vec{r}} \left[s + m + sm\right]$$

s = structural roughness m =domain distribution roughness sm =cross term contains magnetic roughness

Diffuse scattering due to:

- domain size distribution
- orientational domain distribution
 Diffuse scattering diminishes in high fields





New ³He Spin-Filter







New ³He Spin-Filter







New ³He Spin-Filter



³He spin-filter technique is very useful for the polarization analysis of offspecular scattering. Compared to solid-state analyzer, the spin-filter covers a wider angular range and is free of small angle scattering.





AF-coupled Co/CoO multilayer







Exchange bias effect

Exchange interaction of a F and AF layer across a common interface

























Magnetic field















F. Radu, M. Etzkorn, R. Siebrecht, T. Schmitte, K. Westerholt, and H. Zabel Phys. Rev. B 67, 134409 (2003)





























Magnetic hysteresis: neutrons tell the difference

1. Nucleation and domain wall movement:



2. Coherent Rotation:



3. Domain formation:









PNR results of CoO/Co bilayer



F. Radu, M. Etzkorn, R. Siebrecht, T. Schmitte, K. Westerholt, and H. Zabel Phys. Rev. B 67, 134409 (2003)





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Diffuse scattering from CoO/Co







After first field cooling: Single domain state, large H_{c1}







After first field cooling: Single domain state, large H_{c1}



After first reversal: domain wall motion, creating AF domains





After first field cooling: Single domain state, large H_{c1}



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After first field cooling: Single domain state, large H_{c1}



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After first field cooling: Single domain state, large H_{c1}



After first reversal: domain wall motion, creating AF domains



Spin glass type interfacial layer







Neutron reflectometers with polarization analysis

ADAM	ILL	SPN	Dubna
D17	ILL	REFLEX	Dubna
HADAS	FRJ	ROG	Delft
CRISP	ISIS	REFSANS	GKSS/FRM II
AMOR	PSI	N-REX	MPI/FRM II
MORPHEUS	PSI	MIRA	FRM II
V6	HMI	MARIA	Jülich/FRM II
EROS	LLB		
PRISM	LLB		
PNR	GKSS		
NeRo	GKSS		







Do we still need PNR?





X-Ray Magnetic Dichroism XMCD

XMCD experimental layout







XMCD and XRMS



- Element specific
- Spin and orbital moment analysis
- Magnetic scattering (XRMS)
- Vector magnetometry (similar to MOKE)
- Environment of magnetic ion (metallic versus ionic)
- High time resolution (ns-ps)
- High spatial resolution (x-ray microscopy: 10-20 nm)





YES, PNR is still required!

- The only method to provide depth resolved absolute magnetic moments
- Sensitive to magnetic induction (including stray fields in domain walls and screening fields in superconductors)
- No interference with optical terms, and no independent determination of optical parameters required
- Born approximation is sufficient for analysis at Q > Q_c
- Vector magnetometry is measured in the same field configuration
- Deep interfaces and layers are accessible
- Polarization analysis of diffuse scattering possible
- Coherence length of neutrons on the order of magnetic domain sizes (several μm), providing access to fluctuation terms.





Literature

- J.F. Ankner and G.P. Felcher, J. Magn. Magn. Mater. 200, 751 (1999)
- M.R. Fitzsimmons et al. J. Magn. Magn. Mater.
 271, 103 (2004)
- H. Zabel and K. Theis-Bröhl, J. Phys.: Condens. Matter 15, S505 (2003)
- H. Zabel, Materials Today, Jan. 2006
- Many further references are in these reviews







Thank you for your attention



