Polarised Neutron Reflectometry

a complementary method to RIXS and ARPES

spectroscopy workshop on novel materials
PSI - SYN
Beatenberg, 3.–7. May 2011
...did so far:

- chemistry studies
- $\gamma$-Compton spectroscopy on GaAs
- x-ray diffraction (resonant! HASYLAB, ESRF) on GaAs and ZnSe in electric fields
- neutron optics development
- instrument scientist, reflectometry
- PNR on layered magnetic films
  - YBCO/LCMO (C. Bernhard, B. Keimer)
  - div. (F. Miletto)
  - LSMO/YBCO (M. Radovic)
· CV
  → done.

· intro to PNR
  → reflectometry in general
  → ... with neutrons
  → ... on magnetic samples
  → experimental set-up

· experiments: LSMO / YBCO interfaces
  → bi-layers (Y. Sassa, M. Radovic)
  → multilayers (M. Radovic)
Analogy to visible light

Flat surfaces partly reflect light
→ picture of the boot

Some media also transmit light
→ ground below the water

Parallel interfaces
→ colorful soap bubbles
Fresnel reflectivity

- reflectivity of a sharp flat surface
- total external reflection for \( q_z < q^c \)
- exponential decay of \( R(q_z) \) for \( q_z > q^c \)

neutrons / x-rays:
\[
\lambda \in \{1 \ldots 20 \text{ Å}\}
\]
\[
\omega^c < 1^\circ
\]
refraction of transmitted beam
\( \Rightarrow \) dynamical scattering theory
reflected intensity of a multilayer

several parallel interfaces:
interference of all waves

⇒ complex reflectance

\[ r = r(q_z, n_0, n_1, n_2, \ldots, d_1, d_2, \ldots) \]

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

– illumination of the sample
– resolution of the set-up
  \( \Delta \omega, \Delta \lambda \)
– non-sharp interfaces
– inhomogeneous layers
**off-specular scattering**

Ni/Ti multilayer (non magnetic!)

In our cases:

- Resolution in $x$: $\approx 0.01^\circ$
- Resolution in $y$: $> 1^\circ$

$\Rightarrow$ Integrated over $y$
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)

J. Stahn:
*Introduction to polarised neutron and resonant x-ray reflectometry*
http://people.web.psi.ch/stahn/publications

... on magnetic systems

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

... of a surface

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

decay $\Rightarrow$ roughness

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

$\omega/\text{deg}$
simulated reflectivity

... of a thin layer

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

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

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

... of a thick layer

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

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

minima \( \Rightarrow t_{\text{layer}} \)

\( \omega / \text{deg} \)
simulated reflectivity

... of a periodic multilayer

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

maxima \(\Rightarrow t_{\text{bilayer}}\)

amplitudes \(\Rightarrow \Delta V_{\text{layers}}\)

\(\Rightarrow \Delta t_{\text{layers}}\)

minima \(\Rightarrow t_{\text{film}}\)
what is $V_i$ for neutrons?

interaction neutron / nucleus $j$ with $\lambda \gg r_{\text{nucleus}j}$

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

$$ V_i^n = \frac{1}{Vol} \int_j V_j^{\text{Fermi}} \, dr $$

$$ = \frac{2\pi \hbar^2}{m} \frac{1}{Vol} \sum_j b_j $$

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

interaction neutron magnetic moment $\mu / \text{magnetic induction } B$

$$ V^m = \mu B $$

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

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

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

$\mu \perp B \Rightarrow V^m = 0$
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
energy-dispersive set-up

neutron reflectometer

Amor at SINQ

time-of-flight / energy encoding
sample environment

cooling with a
*closed cycle refrigerator*  \( 8 \text{ K} < T < 450 \text{ K} \)

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

or

usage of a cryo-magnet  \( 1.4 \text{ K} < T < 300 \text{ K} \)
\[-50000 \text{ Oe} < H < 50000 \text{ Oe} \)

and *sample*

and sample tilt- and translation stages for alignment
from the sample to a profile
**pro / con**

**magnetic** signal almost as strong as nuclear one

only \( B_\perp \) is probed

no element sensitivity

high **depth resolution** (down to 0.1 nm)

strongly **model-based**

penetration depth 1 000 nm

resolution limit 500 nm

limited \( q_z \)-range accessible

extreme sample environments are **no** problem

**time-consuming** data analysis

supporting methods needed
measurements: LSMO/YBCO interfaces

bi-layer: STO/LSMO/YBCO

sample size: $10 \times 5 \text{ mm}^2$
measurement time: 6 h

reflectivity $\log_{10}[R(q_z)]$

$|+\rangle_{100 \text{ K}}$
$|\rangle_{100 \text{ K}}$

YBCO 7 nm
LSMO 30 nm
STO $\infty$
measurements: LSMO/YBCO interfaces

bi-layer: STO/LSMO/YBCO


sample size: 10 × 5 mm²
measurement time: 6 h
fit-time: 8 h

simulation
free parameters:
thicknesses
magnetisation

fit is not satisfactory!
measurements: LSMO/YBCO interfaces

multi-layer: STO/[LSMO/YBCO]$_4$/LSMO

M. Radovic, May 2010

sample size: $5 \times 5 \text{ mm}^2$
measurement time: 18 h
measurements: LSMO/YBCO interfaces

multi-layer: STO / [LSMO / YBCO]_4 / LSMO

M. Radovic, May 2010

sample size: 5 × 5 mm²
measurement time: 18 h
fit-time: 12 h

simulation
free parameters:
  thicknesses
  magnetisation
  magnetically dead layers

fit is good
measurements: LSMO/YBCO interfaces

multi-layer: STO/[LSMO/YBCO]$_4$/LSMO

M. Radovic, May 2010

- Top layer: density reduced by $\approx 20\%$
- Thicknesses increased by $\approx 10\%$
- YBCO: density reduced by $\approx 4\%$
- LSMO: magnetisation $2.0\,\mu_B$/Mn
- LSMO: magnetically dead layers $\approx 0.6\,\text{nm}$
measurements: LSMO/YBCO interfaces

multi-layer: STO/[LSMO/YBCO]_4/LSMO

M. Radovic, May 2010

LSMO: magnetically dead layers ≈ 0.6 nm
measurements: LSMO/YBCO interfaces

Multi-layer: STO/[LSMO/YBCO]$_4$/LSMO

M. Radovic, May 2010

LSMO: no magnetically dead layers

$\Rightarrow$ higher $q_Z$ & better statistics needed
measurements: LSMO/YBCO interfaces

multi-layer: STO/[LSMO/YBCO]₄/LSMO

M. Radovic, May 2010

sample size: 5 × 5 mm²
measurement time: 18 h

reflectivity \( \log_{10}[R(qz)] \)

structurally forbidden peak
⇒ magnetic profile breaks symmetry
magnetically dead layer of \( \approx 0.6 \) nm

to be fitted with a more complex model
to take along:

- PNR probes $\rho(z)$ where $\rho = \rho(\text{composition}, B_{\perp})$
  - atomic depth resolution
  - lateral integration over several $\mu$m

- data analysis via comparison with model
  - $\Rightarrow$ no unique solution
  - $\Rightarrow$ PNR is a team-player

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<tr>
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<th>probes</th>
<th>depth</th>
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<tbody>
<tr>
<td>ARPES</td>
<td>surface</td>
<td>1 nm</td>
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<tr>
<td>RIXS</td>
<td>bulk</td>
<td>100 nm</td>
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<tr>
<td>PNR</td>
<td>interfaces</td>
<td>1000 nm</td>
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<tr>
<td>XR</td>
<td>interfaces</td>
<td>100 nm</td>
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conventional TOF set-up on Amor:

- $\Delta q$ defined by flight-path length and slits
- energy-dispersive

selene set-up on Amor:

- $\Delta q$ defined by flight-path length and position-sensitive detector
- energy- and angle-dispersive