Assisting polarised neutron experimentalists: Extracting magnetic depth profiles from ab-initio calculations

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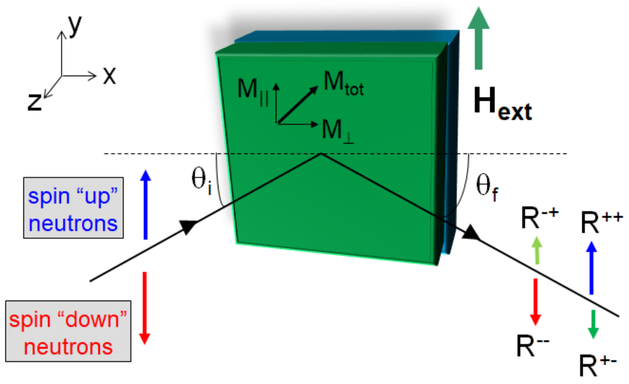
Neutron reflectometry: An overview



Neutron beams can be spin polarised with spin up (+) or down (-) states, which undergo reflection (R) from a magnetic film or mirror-surface at low incident angles. The neutron beam interacts near the surface of a material via a 1D quantum potential involving a nuclear and magnetic contribution. This leads to the phenomena of neutron birefringence where the two spin states reflect differently.

$$V_{\pm} = \frac{2\pi\hbar}{m_n} N\langle b \rangle \pm \bar{\mu}_n \cdot \bar{B}$$

Nuclear Scattering | Magnetic Scattering

$$n_{\pm} = \sqrt{1 - V_{\pm}/E}$$

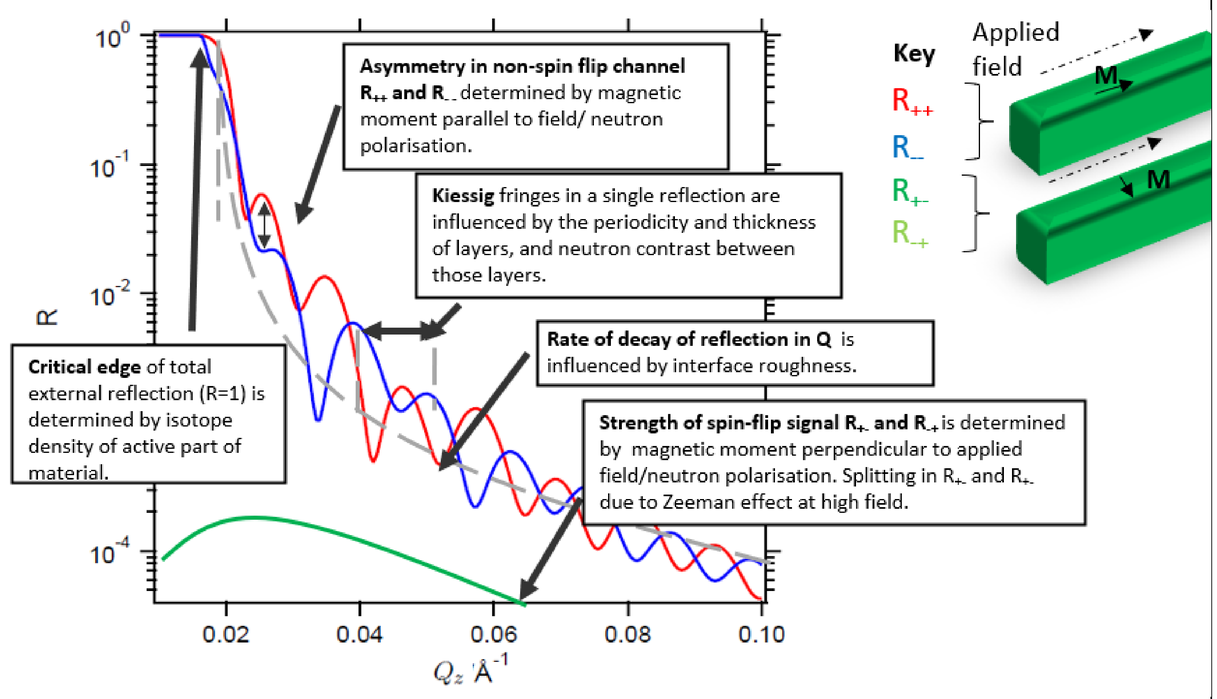
Neutron Refractive index depends on magnetisation!

N is the number of atoms / m_3
<b> is the average nuclear scattering length
m_n is the mass of the neutron

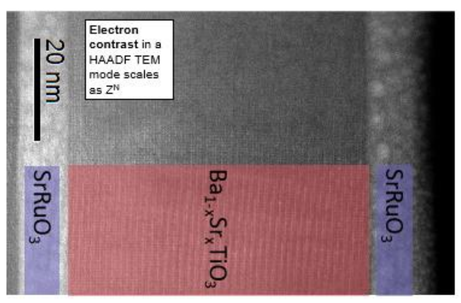
μ_N is the neutron magnetic moment
B is the magnetic induction field

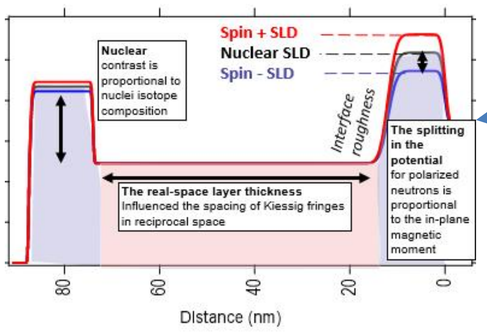
Interpreting PNR patterns

There are some qualitative guidelines for interpreting PNR data as shown below. In general, as phase information is lost during the measurement, the data needs to be fitted to extract quantitative details of the 1D potential (scattering length density).



Quantifying the neutron interaction potential





There are many complementary techniques (e.g. high-resolution TEM, X-ray reflectometry) that can be used to measure the structural parameters in thin films, and therefore constrain the nuclear scattering term for neutron reflectometry experiments to reduce the number of free parameters.

e.g. Direct comparison with cross-sectional TEM and neutron nuclear SLD is possible.

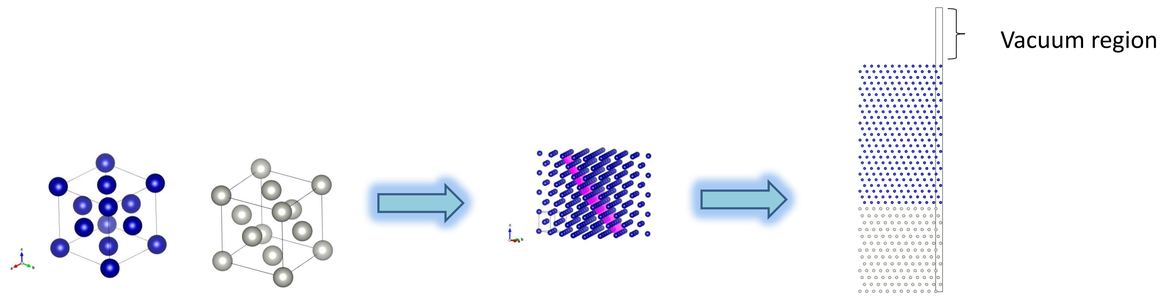
Check out our book chapter!
S. Callori, D. L. Cortie, T. Saerbeck, K.-W. Lin Solid State Physics: SSP Volume 71: AID 18 (2020)

In contrast, the magnetic sensitivity is unique to PNR, as no other technique can fully quantify the nanoscale magnetic profile in absolute units. Therefore, in order to constrain the magnetic term, and assist PNR experimentalists, it is advantageous to use complementary theoretical

Modelling virtual thin film systems with DFT

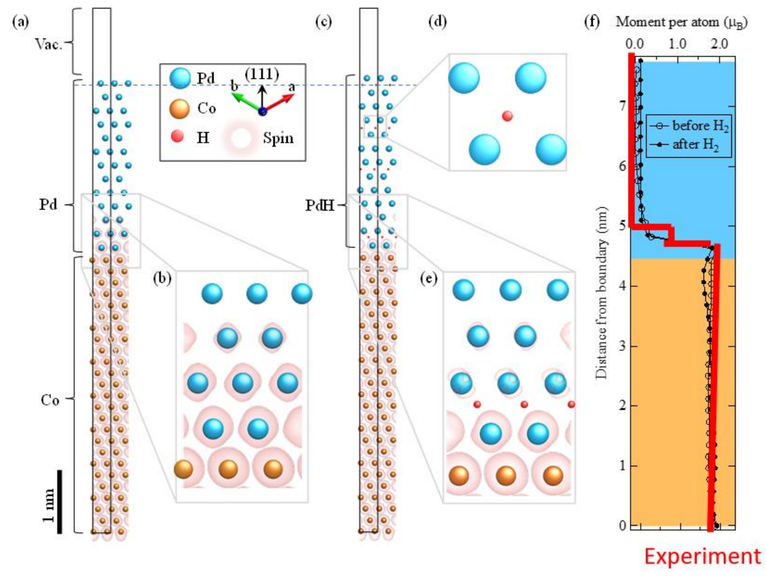
Our group has developed the capability to calculate the magnetic depth profile of single layers and multilayers using spin-polarised density functional theory for slab models implemented in VASP. The steps involved are:

1) DFT optimization of the single phases based on their bulk crystal structure cif files
2) Virtual cleavage of the optimised phases to form thin films surfaces
3) Modify the boundary conditions in the z-direction (out-of-plane) by introducing a vacuum region
4) Perform ionic relaxation / damped molecular dynamics to optimise the interface
5) Perform collinear spin-polarized calculations to generate the wave functions, and calculate the spin density
6) Optional: High quality NCSF cycle using the wave functions with spin orbit added



Extracting the magnetic depth profile in Co:Pd layers

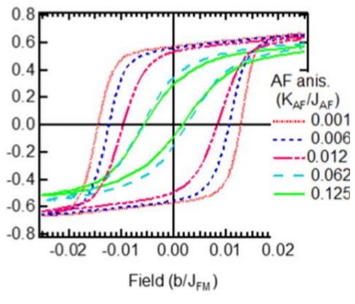
Using this DFT technique we calculated the spin density of Co/Pd bilayers, and compared this with experimental PNR SLD profiles. This helped to explain the functionality of magnetic hydrogen sensors.



G. L. Causer, D. L Cortie *et al.,* ACS Appl. Mat. & Int. **11**, 35420-35428 (2019)

Calculating magnetic properties using Model Hamiltonians

To simulate other magnetic properties (e.g. ordering temperatures, susceptibility, exchange bias), we also use model Hamiltonians solved by Monte Carlo and Micromagnetic calculations. For example we can calculate the unidirectional anisotropy (exchange bias) in nanocrystalline, bilayer coupled systems:



G. Causer, D. L. Cortie, Japanese Journal of Applied Physics 59 (SA), SAAC03 (2019)