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Magnetic multilayers studied by polarised neutron reflection

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Abstract

The capabilities of polarized neutron reflection (PNR) for directly determining the magnetic and non-magnetic structure in magnetic multilayers and superlattices are reviewed. We discuss examples of studies of the layer-dependent moments and spin orientations in various single films, exchange coupled trilayers and superlattices based on transition metal ferromagnetic layers.

Keywords: Polarized neutron reflection; Magnetic multilayers; Exchange coupling; Magnetometry

1. Introduction

There has recently been a renaissance of interest in the problem of interface-induced magnetic moments [1,2] for two principal reasons. Firstly, it is recognised that the interface magnetic moment is a fundamental quantity, intimately related to such key properties of magnetic multilayers as the interface anisotropy, interlayer exchange coupling and giant magnetoresistance ratio [2]. Secondly, interest is now rapidly growing in the application of polarised radiation techniques in general, such as X-ray Magnetic Circular Dichroism (XMCD) [3], second harmonic generation [4], polarised neutron reflection (PNR) [5] and diffraction which give access to the magnetic structure at interfaces.

In general, a variation in the magnetisation as a function of layer position can arise in chemically homogeneous layers due to, for example, to interface strain effects or interface anisotropies. In the case of magnetic superlattices composed of ultrathin ferromagnetic films, each magnetic layer adopts a spin configuration which is determined by the competing energies associated with the (layer-dependent) interlayer coupling fields and the internal fields of each layer. Complex spin configurations result and phase transitions between, for example, artificial antiferromagnetic, ferromagnetic or more complex phases can occur [6]. Essential to the underlying physics involved is the fact that each layer is non-equivalent – in particular, the outermost (surface and substrate) layers have configurations which differ from those of the interior. An important example is the spin-flop state predicted in Fe/Cr-type multilayers [7]. PNR is particularly important in this context since it is able to provide measurements of the *layer-selective* vector magnetic moment in ultrathin structures. Moreover, PNR is self-calibrating since the spin dependence of the reflectivity yields the total magnetic moment of the layer while the layer thickness in the nm range can be determined independently from the wave vector dependence of the reflectivity. We shall discuss the application of PNR to the study of magnetic moment distributions in ultrathin magnetic structures and to the structural characterisation of the interface.

2. Polarised neutron reflection

In PNR the partially reflected neutron intensity is measured as a function of the incident spin state and

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incident wave vector, either with or without polarisation analysis of the scattered beam. The incident wave vector k_{inc} is varied either by rotating the sample with fixed incident wavelength λ_{inc} or by employing a time-of-flight method with a fixed incidence angle θ [5]. The reflecting medium is treated as a stratified medium or multilayer as in conventional optics. To a good approximation the specular reflectivity is purely a function of the perpendicular wave vector component of k_{inc} given by q_{inc} (i.e. resolved along the sample normal in the vacuum region). For the jth medium the perpendicular component of wave vector is given by

$$q_j = \sqrt{q_i^2 + q_{ci}^2 - q_{cj}^2} \tag{1}$$

and with the critical wave vector q_{ci} for the ith medium given by

$$V_i = \frac{2\pi\hbar^2}{m_{\mathrm{n}}}\rho_i b_i - \mu_{\mathrm{n}} B_i, \tag{2}$$

where m_{n} is the neutron mass, ρ_i is the atomic density, b_i is the bound coherent neutron scattering length of the material [5], μ_{n} is the neutron magnetic moment and B_i is the total magnetic induction in the medium. Only the component of the magnetic induction in the plane of the sample contributes to refraction. In the simplest experimental arrangement, the sample magnetisation is aligned by an applied field in the plane of the sample and perpendicular to the scattering plane. Typically, for solids, the critical angle for 12 Å neutrons is of the order of 1°. The solution to the 1D Schrodinger equation for the optical potential of Eq. (1) for the ith medium is given by the sum of a forward (amplitude A_i) and backward travelling (amplitude B_i) wave. Methods for calculating the reflectivity coefficients have been described elsewhere [5, 8]. The flipping ratio $F = R^+/R^-$, where the superscripts correspond to the incident spin parallel (+) or antiparallel (−) to the applied field or the spin asymmetry $S = (F-1)/(F+1)$ is determined as a function of wave vector thus yielding magnetometric information.

Macroscopic surface waviness gives rise to long-range fluctuations in the surface flatness equivalent to an increase in the angular spread of the incident beam. Following Ref. [10] for an interface exhibiting a random Gaussian roughness distribution, the specular reflectivity r_{ij} is attenuated to become $r_{ij}\exp(-W_{ij})$, where $W_{ij} = 2q_i q_j \sigma_{ij}$ and where $\sigma_{ij} = \langle \Delta y_{ij}^2 \rangle$ defines the variance of the local fluctuation in interface position. In practice roughness correlations occur and diffuse reflection results [11]. The diffuse scattering intensity accepted by the detector scales with the solid angle accepted and for measurements on single-crystal substrates its contribution must be accounted for at large wave vector. All early experiments on Fe films, supported by vicinal Ag substrates with very large diffuse scattering, led to an underestimate of the magnetic moment [12]. Spin disorder at the interface of magnetic multilayers has also been measured as a function of field in Fe/Cr [13] and Co/Ru multilayers [14].

3. Vector magnetometry

For the case of non-spin aligned (non-colinear) layers it is necessary to use a 4-component vector of the neutron wave within each medium of the form $(A_i^+, B_i^+, A_i^-, B_i^-)$, where the superscripts refer to the spin component with respect to the applied field [8]. In the case of non-colinear structures the + and − reflectivities are both dependent on both the in-plane components of the magnetisation vector as described by a reflectivity matrix. Accordingly, the flipping ratio versus wave vector curve is changed dramatically according to the spin configuration of the structure. In this case the non-parallel magnetic spins can be thought of as exerting a torque on the neutron spins with the result that a partial 'flipping' of the neutron spin takes place. The neutron beam is then described as a superposition of up- and down-spin states defined with respect to the original eigenstate in the guide field region. In PNR, the layer selectivity results from the spatial variation of the wave within the solid which arises due to the standing intensity waves which develop due to the reflection at each interface within the structure. This is distinct from the case of diffraction which occurs at higher wave vector, where the Fourier component of the spin configuration is probed [8]. Thus, in the case of two fully antiparallel ferromagnetic layers, PNR is able to determine the absolute orientation of each layer with respect to the applied field and not only the antiparallel ordering [5].

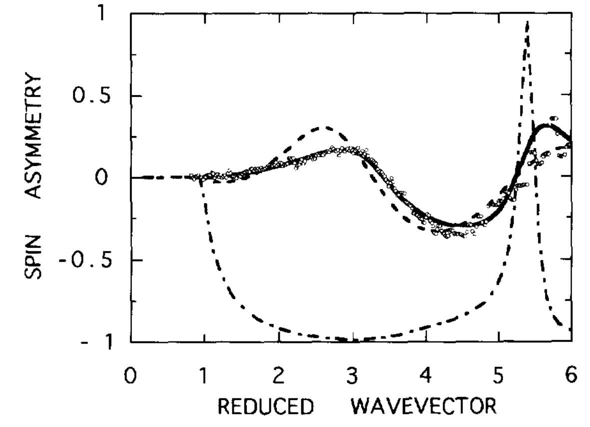

Fig. 1. The spin asymmetry observed for a sputtered Cr/Fe/Cr/Fe/Si structure with antiferromagnetic coupling compared with simulations (dashed and dot-dashed lines) which assume the two possible antiparallel orientations of the full layer moments with respect to the applied field direction [5, 15]. The full line is a best fit in which the moment magnitude and orientation are adjusted in each layer (see text).

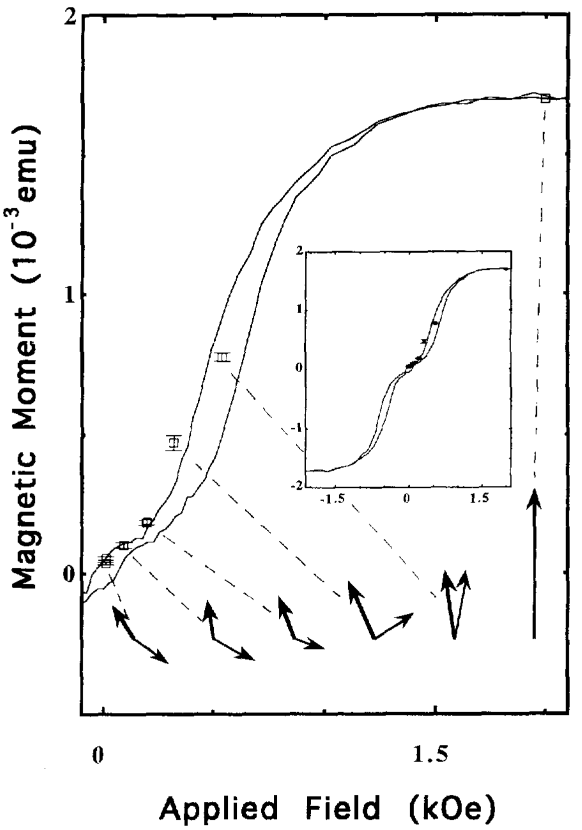

Fig. 2. The magnetic hysteresis loop for the Cr/Fe/Cr/Fe/Si structure (positive fields only) compared with the parallel component of magnetisation determined for specific field values together with the spin configurations of the top (full arrow) and the bottom (thin arrow) layer moments determined from PNR. The inset shows the full loop.

The spin asymmetry has been measured near remanence (12 Oe applied field) for a sputter-grown Cr/Fe/Cr/Fe/Si trilayer structure with antiferromagnetic interlayer coupling and the results compared with simulations which assume two possible purely antiparallel configurations of the full layer magnetic moments as shown in Fig. 1: (i) top layer parallel to the applied field (dashed line) and top layer antiparallel to the applied field (dot-dashed line) [9]. Clear differences between the asymmetry calculated for each case are seen. However, an almost exact fit to the data can be achieved in a model in (i) the moment in each layer is reduced from the bulk value due to the formation of magnetic domains on a scale much smaller than the coherence length of the neutron in-plane (~100 μm [5]) and (ii) that canting of the in-plane moment orientations of each layer with respect to each other occurs at low fields with inequivalent orientations of each moment with respect to the applied field (see Fig. 2). The systematics of the spin configuration has been studied as a function of applied field strength, as shown in Fig. 2. The total net moment along the applied field direction determined from these fits to the PNR data agree well with the corresponding values determined from vibrating sample (VSM) magnetometry. These measurements emphasise the inequivalence of the magnetic moment orientations in each layer. The canting at low field may indicate the presence of biquadratic coupling or, alternatively and most probably, that pinning processes differ in the two Fe layers, resulting in different moments and orientations. This study thus demonstrates the capability of PNR in carrying out layer-selective measurements. It also illustrates the need for careful studies of trilayer structures in testing the assumption that the magnetic layers are magnetically equivalent. This assumption frequently provides the basis for the analysis of polarized neutron diffraction from multilayers and superlattices [15–17].

Epitaxial spin valve structures of the form Cu/Co/50 ÅCu/FeNi/Cu/Si (0 0 1) were recently studied by PNR [18]. Room-temperature SQUID magnetometry loops for fields applied along the easy axis show saturation fields for the Co layers of ~200 Oe and reveal abrupt reversal of the FeNi layers at low fields with the Co layers reversing at higher fields due to the presence of a cubic magnetocrystalline anisotropy [19]. In fitting the spin-dependent reflectivity data obtained from the sample fully saturated by a sufficiently large applied field (i.e. layers aligned parallel), additional intermixed FeNi–Cu and Co–Cu layers of 9–11 Å thickness and variable composition are introduced at the interfaces [18]. If the interface regions are not included, the simulated reflectivities yield values which are not consistent with the total moment measured by SQUID magnetometry. This is important in showing that PNR is capable of giving the magnetic moment of very thin interface regions selectively and is therefore quite distinct from element-specific information provided by XMCD for example. It also illustrates the importance of combining the magnetic measurements from PNR with independent measurements (in this case SQUID) which are sensitive to the total thickness or moment of the sample. The presence of intermixed regions in the spin valve structures is consistent with a recently proposed model of the temperature dependence of the giant magnetoresistance amplitude in spin valve structures in which a spin-flip scattering of a strength dependent on the interface sharpness (determined principally by the level of chemical interdiffusion) is invoked [20]. Layer-selective vector magnetometry measurements using PNR have been made under conditions in which the relative alignment of the FeNi and Co layers in the single domain state is controlled using an external field [21].

4. Absolute magnetometry in single ferromagnetic films

Epitaxial samples of the form X/Fe/Ag (0 0 1) with X = Cu, Au, Ag and Pd were studied by PNR [22, 23]. In Fig. 3 we show the observed spin asymmetry (solid circles) corrected for partial incident beam polarisation, background intensity and diffuse scattering for

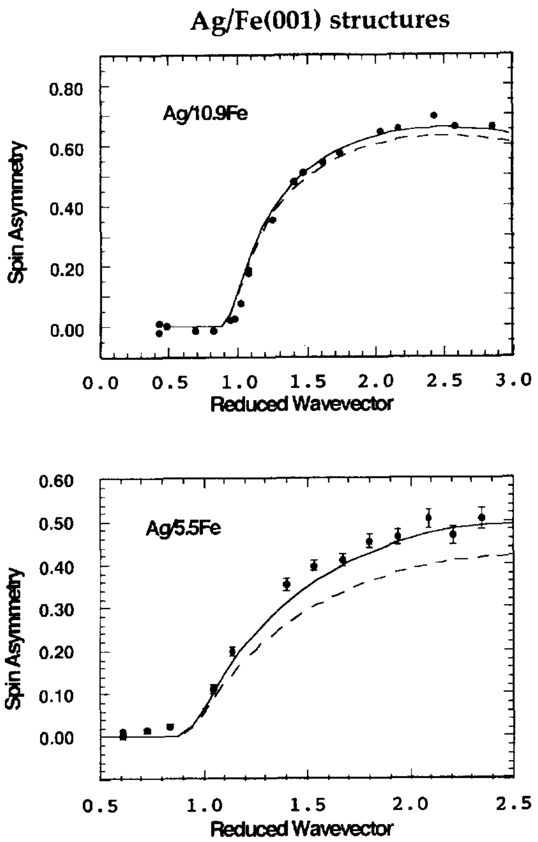

Fig. 3. The corrected spin asymmetry obtained at low temperature for: (top panel) 20 ML Au/7 ML Ag/10.9 MLFe/Ag (0 0 1) and (lower panel) 20 ML Au/7 ML Ag/5.5 ML Fe/Ag (0 0 1). The dashed and solid lines in the plots of the spin asymmetry refer to model fits for the bulk moment and assuming an enhanced moment, respectively (see text).

Ag/Fe/Ag structures [22, 23]. In each case the spin asymmetry was calculated using the known structural parameters adjusting only the moment per atom in order to best fit the data (shown as a solid line) and also assuming the bulk valve of $\mu_{Fe} = 2.2\mu_B$ (shown as a dashed line).

Results for structures with Cu, Au and Pd overlayers are shown in Fig. 4. For a Pd/5.6 Fe/Ag (0 0 1) sample an effective layer averaged moment of $2.66 \pm 0.05\mu_B$ (including the contribution from spin-polarised Pd) was determined, corresponding to a total enhancement of 20 ± 2% per Fe atom [23]. Augmented spherical wave (ASW) calculations in the LSDSA yield a moment for a Pd/5ML Fe sample of $2.59\mu_B$ for an ideal

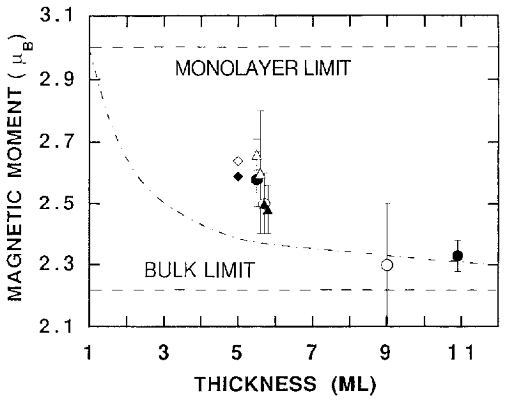

Fig. 4. The values of the layer averaged moment per Fe atom deduced from PNR measurements for structures of the form Au/X/Fe/Ag (0 0 1) where X = Cu (solid triangles), Au (open circles), Ag (solid circles) and Pd (open triangles) for the Fe thickness shown. The results of ASW calculations for a Pd/5 ML Fe/Ag (0 0 1) structures are shown for the case of no roughness (solid diamond) and approximately 1 ML roughness (open diamond).

interface in disagreement with the measured value. However, the calculated value increases to $2.64\mu_B$ when 25% intermixing is included across a 1 ML region at the interface. The average value of the layer-averaged moments for Cu/Fe, Ag/Fe and Au/Fe samples of Fe thickness approximately 5.5 ML is $2.5 \pm 0.05\mu_B$ (corresponding to an average enhancement of $14 \pm 2\%$) which is higher than the value of $2.4\mu_B$ predicted by Ohnishi et al. [25] for atomically sharp interfaces. Ferromagnetic resonance (FMR) line shape measurements of the total moment of the Fe films relative to a reference sample [23, 26] are found to agree with the PNR results within experimental error. The combined results clearly show that the enhancement occurs at the interface.

In thicker (> 20 ML) ferromagnetic films, the magnetic layer thickness can be directly determined by fitting the oscillatory wave vector dependence of the spin asymmetry. Thus, the magnetometry measurements are self-calibrating and magnetic profile effects can be probed [27, 28].

5. Summary and outlook

We have shown that PNR provides a valuable layer-selective magnetic probe of thin and ultrathin magnetic structures due to the special combination of magnetic and structural information that it provides. The capability of PNR in quantitatively and selectively probing the interface spin structure of layers with a common magnetic element is likely to provide an important complementary probe to XMCD techniques and to play an important part in unravelling the spin structure of magnetic interfaces in future.

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