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Larmor precession of polarised neutrons as a probe of the magnetisation of ultrathin epitaxial films

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Abstract. We demonstrate that a large and readily measurable rotation of the spin orientation occurs for polarised neutrons critically reflected from epitaxial ferromagnetic films. For a monolayer thickness BCC Fe film, a spin polarisation of ~5% is induced in the direction normal to the incident polarisation vector at the critical reflection condition. For buried films, this polarisation change is found to be reduced only slightly. By measuring the polarisation rotation of the reflected beam in the critical reflection region, the absolute value of the magnetic moment per atom in the epitaxial film can be inferred with an accuracy which compares favourably with that which can be obtained using conventional polarised neutron reflection techniques.

1. Introduction

The problem of a spin-$\frac{1}{2}$ particle tunnelling through or reflected from a potential barrier has received considerable attention throughout the history of quantum mechanics (Collins *et al* 1987). Buttiker (1983) has emphasised that a particle tunnelling through a barrier in a magnetic field does not actually perform true Larmor precession and that the main effect of the magnetic field is to partially align the spin with the field. This phenomenon is used to polarise neutrons on reflection or transmission from or through saturated ferromagnetic films (Bloch 1936). Similarly, polarised electron beams can be produced by field emission from metals coated with a ferromagnetic semiconductor film which presents a spin dependent potential barrier to the tunnelling electrons (Muller 1972). More recently polarised neutron reflection (PNR) has been used to deduce the magnetisation of sandwiched epitaxial ferromagnetic films down to monolayer thickness (Bland *et al* 1987).

For angles of grazing incidence greater than that at which total external reflection occurs, the intensity of the reflected beam is modified as a result of interaction with the magnetic film. As noted by Felcher (1981), spatial variations of the magnetisation profile close to the surface of the reflecting medium can be detected as small changes in the spin-dependent neutron reflectivity. However, such changes are largest precisely where the absolute reflectivity is small, and hence this promising technique, while providing a means of determining the absolute value of the magnetic moment per atom in ultrathin films, is limited in its accuracy by the low reflected signal. The long counting times

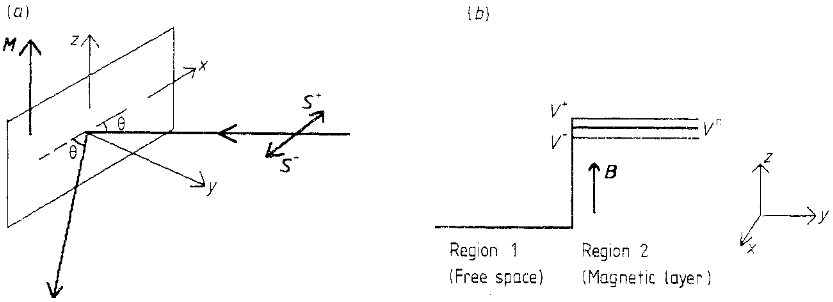

Figure 1. (*a*) Diagram showing the geometry of the physical situation under consideration. The polarisation direction of the incident beam is parallel or antiparallel to the *x* axis; the magnetisation of the film is along the *z* axis. (*b*) A simple 1D step potential to represent the reflecting medium (see § 2 of text).

required, even for intense cold neutron sources, impose severe constraints on such experiments.

In this paper we consider an alternative geometry for the neutron reflection experiment in which the change in polarisation, rather than the spin dependent intensity of the reflected neutron beam, is used to deduce the magnetisation of epitaxial films. We find that single Fe monolayers induce large and measurable changes in the polarisation of a beam launched with its polarisation transverse to the sample magnetisation. Such measurements can be made within the critical reflection region (i.e. the reflectivity is unity), so offering an important advantage over conventional PNR measurements.

2. Theory

We consider a spin-polarised neutron beam incident at a grazing angle θ to a thick magnetic film with the z axis normal to the plane of reflection (figure 1(*a*)). The polarisation direction of the incident beam is arranged to lie within the plane of reflection and for the purpose of our discussion is arbitrarily chosen to be parallel or antiparallel to the x axis. This geometry contrasts with the conventional arrangement in which the incident spin vector is aligned parallel or antiparallel to the magnetisation along the z axis. The refracting medium can be treated to a first approximation as invariant with respect to translation in the xy plane (i.e. neglecting roughness and surface height fluctuations which induce diffuse scattering) and the neutron–solid interaction can be modelled as a 1D potential $V(z)$ (Dietrich and Wagner 1985). Solving Schrodinger's equation allows the corresponding neutron wavefield to be calculated.

For a magnetic layer, i, the interaction potential is given by (Marshall and Lovesey 1971)

$$V_i = (h^2/2\pi m_\mathrm{n})\rho_i b_i - \boldsymbol{\mu}_\mathrm{n} \cdot \boldsymbol{B}_i \qquad (1)$$

where m_n denotes the neutron mass and where ρ_i, b_i and $\boldsymbol{B}_i$ denote the atomic density, nuclear scattering length and magnetic induction associated with the layer i. The term $-\boldsymbol{\mu}_n \cdot \boldsymbol{B}_i$ describing the magnetic part of the potential V^m has a dependence upon the neutron magnetic vector $\boldsymbol{\mu}_\mathrm{n}$ and thus upon the neutron spin orientation with respect to the magnetic induction.

A simple step potential is used to represent the reflecting medium (figure 1(b)). As neutrons enter the barrier they are subject to a magnetic field $\boldsymbol{B}$ which causes Larmor precession with frequency $\omega_L = (2\pi g\mu_n B)/h$, where g is the gyromagnetic ratio and μ_n the neutron magnetic moment. The effect on the neutron is to rotate the spin vector about the z axis resulting in a re-orientation of the neutron spin within the xy plane. Beyond the critical region the neutron penetrates the barrier and true Larmor precession no longer occurs, the effect of the magnetisation being to partially align the neutron spin parallel to the magnetic induction $\boldsymbol{B}$. The angle between incident and reflected spin states is assumed to be given by the Larmor frequency multiplied by a characteristic reflection time τ (Buttiker 1983). Considering the Zeeman contribution to the effective potential inside the barrier, we can re-express (1) as a combination of nuclear and magnetic contributions as

$$V^{\pm} = V^{n} \pm \tfrac{1}{2}\hbar\omega_L \tag{2}$$

where V^n represents the nuclear potential, and where $V^{\pm}$ indicates the interaction potential for parallel (+) and antiparallel (−) spins.

We assume that the external guide field required to maintain the incident beam polarisation is negligible compared with the coercive field of the film, so that the film remains remanently magnetised. In practice a guide field of ~ 1 G is sufficient to maintain the incident neutron polarisation, whereas the coercive field is likely to be substantially greater than this value (typically in the range 10–1000 G).

To calculate the precession angle $\omega_L\tau$, we begin by considering the Hamiltonian which describes the neutron reflected from the thick magnetic film. This Hamiltonian is given by

$$H_1 = (p^2/2m_n) \qquad \text{(in free space)} \tag{3}$$

$$H_2 = (p^2/2m_n + V^n) - (\tfrac{1}{2}\hbar\omega_L)\,\sigma_z \qquad \text{(in magnetic layer)} \tag{4}$$

where σ_z is the Pauli spin matrix along z. These Hamiltonians act on spinors of the form

$$\Psi = \begin{bmatrix} \psi_+(y) \\ \psi_-(y) \end{bmatrix} \tag{5}$$

which represent the spin state of the incident beam. For our specific geometry, where y is normal to the film and the incident beam is initially polarised in the x direction, we can write

$$\Psi_{\text{inc}} = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ 1 \end{bmatrix} \exp(iky). \tag{6}$$

Using the Hamiltonians H_1 and H_2 in the standard time-independent Schrodinger equation we can define a free space wavevector $\boldsymbol{k}$ and a wavevector within the barrier $\boldsymbol{\kappa}$

$$k = \sqrt{2m_n E}/\hbar \tag{7}$$

$$\kappa_{\pm}^2 = k^2 - (2m_n/\hbar^2)(V^n \pm \tfrac{1}{2}\hbar\omega_L). \tag{8}$$

The wavevector $\boldsymbol{\kappa}$ provides an exponentially decaying neutron-wave solution within the magnetic material.

The amplitude reflection coefficient between the two media is given by the relation

$$r^{\pm} = r_{12}^{\pm} = (ik - \kappa_{\pm})/(ik + \kappa_{\pm}) \tag{9}$$

which is the familiar Fresnel reflection coefficient.

Thus for identical incident energies there exist two different reflectivities depending on the incident neutron spin state (+ or −). In the critical region both reflectivities are of magnitude unity and differ only in phase. For the spinor components we assume a superposition of incident and reflected waves, of the forms $\exp(\mathrm{i}ky) + r\exp(-\mathrm{i}ky)$ in region 1 and transmitted wave $t\exp(\mathrm{i}\kappa y)$ in region 2 (where t is the corresponding transmission coefficient).

The orientation of the spin of the reflected neutrons is now given by the spinor

$$\Psi_{\mathrm{r}} = \frac{r^+}{\sqrt{2}}\begin{bmatrix}1\\0\end{bmatrix} + \frac{r^-}{\sqrt{2}}\begin{bmatrix}0\\1\end{bmatrix} = \frac{1}{\sqrt{2}}\begin{bmatrix}r^+\\r^-\end{bmatrix}. \tag{10}$$

The expectation values of the components of reflected beam polarisation $\boldsymbol{P}^{\mathrm{r}}$ are given by (Buttiker 1983)

$$\langle P_y^{\mathrm{r}}\rangle = \langle\psi^*|\sigma_y|\psi\rangle/\langle\psi^*|\psi\rangle$$

(where σ_y is the Pauli spin matrix along y). Hence

$$\langle P_y^{\mathrm{r}}\rangle = \langle\psi^*|\begin{pmatrix}0 & -\mathrm{i}\\ \mathrm{i} & 0\end{pmatrix}|\psi\rangle\Big/\langle\psi^*|\psi\rangle = \mathrm{i}(r^+r^{-*} - r^-r^{+*})/(|r^+|^2 + |r^-|^2). \tag{11}$$

Similarly

$$\langle P_x^{\mathrm{r}}\rangle = (r^+r^{-*} + r^-r^{+*})/(|r^+|^2 + |r^-|^2) \tag{12}$$

and

$$\langle P_z^{\mathrm{r}}\rangle = (|r^+|^2 - |r^-|^2)/(|r^+|^2 + |r^-|^2). \tag{13}$$

It is clear from (13) that in the critical region, true Larmor precession does occur and that, as expected,

$$\langle P_z^{\mathrm{r}}\rangle = 0$$

since

$$|r^+|^2 = |r^-|^2 = 1.$$

The change in reflected spin states can be interpreted in terms of a Larmor precession time τ

$$\langle S_y^{\mathrm{r}}\rangle = -\tfrac{1}{2}\hbar\omega_{\mathrm{L}}\tau \tag{14}$$

$$\langle S_x^{\mathrm{r}}\rangle = \tfrac{1}{2}\hbar(1 - \tfrac{1}{2}\omega_{\mathrm{L}}^2\tau^2). \tag{15}$$

The precession time τ is associated with the amount of time the neutron spends inside the barrier, which is a function only of κ for an infinitely thick barrier. As we have already seen, κ relates directly to the magnetic induction within the film, as does ω_{L} (from equations (1) and (2)). If the magnetic film is assumed to be saturated then the magnetic induction B_0 within the film can be approximated by $\mu_0\boldsymbol{M}_{\mathrm{s}}$, where $\boldsymbol{M}_{\mathrm{s}}$ is the saturation magnetisation. A measurement of the reflected beam polarisation can therefore be used to estimate $\boldsymbol{M}_{\mathrm{s}}$.

By analogy with optics the reflectivity of multilayer films can be easily calculated using etalon-type equations. A simple three-medium system (inset of figure 2(a)) has a reflection coefficient given by

$$r^{\pm}_{ijk} = \frac{r^{\pm}_{ij} + r^{\pm}_{jk}\exp(2\mathrm{i}q^{\pm}_j t_j)}{1 + r^{\pm}_{ij} r^{\pm}_{jk}\exp(2\mathrm{i}q^{\pm}_j t_j)}. \tag{16}$$

This analysis extends to an arbitrary number of layers, the reflectivity $r_{ij\ldots n}$ directly replacing the basic two media reflectivity in equations (11)–(13). Therefore by calculating the total spin-dependent reflectivity according to equation (16) (or its extension to many layers) and substituting the result in equations (11)–(13), the reflected beam polarisation can be evaluated exactly without invoking a precession time explicitly. Moreover, by fitting the wavevector-dependent polarisation to data obtained for an epitaxial film of known thickness, the effective induction within the ferromagnetic layer can be obtained, and hence an estimate of the magnetic moment per atom provided as in conventional PNR experiments.

3. Results

In figure 2(*a*) we show the polarisation of the reflected beam as a function of reduced wavevector q/q_c (where q_c is the critical wavevector for reflection from the substrate) calculated for 1 monolayer of Fe epitaxed to Ag (001). The equivalent 1D potential for this structure is shown in the inset of the figure. A moment of 2.2 μ_B per Fe atom is assumed with the Fe layer adopting the BCC structure with almost zero strain (i.e. perfect epitaxy) (Jonker *et al* 1986). The incident spin is aligned along the x axis as shown in figure 1. The z polarisation is unchanged (i.e. zero) in the critical region $q < q_c$ and so the changes induced in the x and y polarisation correspond to Larmor precession about the magnetic field within the medium. Beyond the critical region (i.e. for $q > q_c$) all components of the polarisation change, as noted by Buttiker (1983), and the partial alignment of the spin induced along the z direction provides the basis for polarising mirror applications, as we have already noted.

The changes in the x and y polarisation components increase monotonically with q. This can be understood as a result of the increased penetration of the neutron wave with increasing q, or alternatively as an increase in the precession time τ (equations (14) and (15)). Whereas the change in the x polarisation is small (of the order of 0.2% at $q = q_c$), the change in the y polarisation at $q = q_c$ is surprisingly large (of the order of 5%). Since the intensity reflectivity is unity in the critical region, both of these changes are in principle detectable using a high flux source. A precision of 0.1% in the critical reflectivity corresponds to a total count of 10^6 which is equivalent to determining the reflectivity at $q = 2q_c$ to within an accuracy of ~1.5%. Such a counting requirement is comparable with that of conventional PNR experiments on thin films where polarisation analysis of the reflected beam is not employed (Felcher 1981). However the substantial changes induced in the y polarisation require much shorter counting times and so can be readily detected with high accuracy. In figure 2(*b*), we repeat the calculations of figure 2(*a*) for the same epitaxial system overcoated with a 100 Å Ag layer. The equivalent 1D potential is shown in the inset. Such a structure is used in conventional PNR experiments since the z polarisation is enhanced beyond the critical region (Bland *et al* 1987) and the buried layer is chemically sealed, so preventing contamination. A reduced wavefield intensity at the magnetic layer arises due to the finite penetration of the neutron wavefield in the

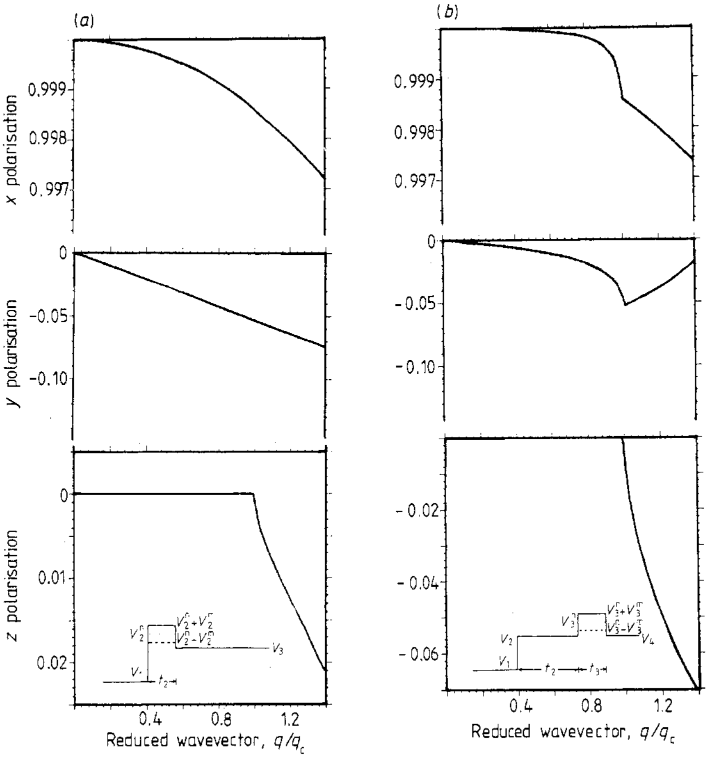

Figure 2. (*a*) The polarisation of the reflected beam as a function of reduced wavevector, for an Fe monolayer epitaxed to Ag (001). Inset: the 1D potential under consideration. (*b*) Repeat of the calculations for the epitaxial system of figure 2(*a*) overcoated with a 100 Å Ag layer.

critical region and this reduces the changes in P_x and P_y in the critical region. However, at the critical wavevector, the changes in P_x and P_y are comparable with those obtained for the free Fe film. Beyond the critical wavevector, P_x changes in a manner similar to the case of the free Fe film. The z polarisation, however, is much increased and, accordingly, to conserve the total polarisation, P_y is distorted. Nonetheless, as in the case of the free epitaxial Fe film, the polarisation changes along the x and y directions are sufficiently large to be readily measurable.

A complication from the viewpoint of designing an experiment to observe such changes in P_x and P_y is that the largest changes occur for P_y, whereas a small change occurs in a large overall polarisation for P_x. If the guide field is maintained along x, then clearly the reflected beam polarisation along y will precess as the neutron leaves the sample. One solution to this problem would be to include a spin flipper (Mezei 1972) between the sample and detector so that the spin component along y can be rotated onto the guide field orientation. A second difficulty is that the measurements are most conveniently performed with the film in the remanently magnetised state. Single-domain-like, ideal epitaxial magnetic films with an in-plane easy magnetisation direction exhibit quadrangular hysteresis loops (Liu *et al* 1988), and so this condition is likely to be met for epitaxial thin-film systems.

The proposed experiment could be readily performed with ultra-cold neutrons (UCN) since high incident beam intensities are not essential. High spin polarisation can be achieved with ultra-cold neutrons and the increased critical angle provides a useful advantage.

4. Summary and conclusions

We have demonstrated that a large and readily measurable rotation of the spin orientation occurs for polarised neutrons critically reflected from epitaxial ferromagnetic films. For a BCC Fe monolayer, a spin polarisation of approximately 5% is induced in the direction normal to the incident polarisation vector at the critical reflection condition. The polarisation change is found to be only slightly reduced for buried films. Measurements of the polarisation rotation of the reflected beam in the critical reflection region can be used to infer the absolute value of the magnetic moment per atom in the epitaxial film with an accuracy of roughly one order of magnitude greater than that which can be achieved with conventional PNR techniques.

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