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Spin-orientation dependence in neutron reflection from a single magnetic film

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We demonstrate that the magnetic state of a thin magnetic film can be probed using polarized neutron reflection even when the magnetization in the film plane is at 90° to the polarization direction of the incident neutron beam. For this special case, no magnetic interaction is expected classically, thus the effect is entirely quantum mechanical in origin. We show both theoretically and experimentally that it arises from the independent interaction of each spin component with the magnetization in the film. We demonstrate this effect for a single Co/GaAs(001) film and discuss its application to magnetic measurements using an unpolarized neutron beam.

Polarized radiation techniques such as neutron reflection[1,2] (PNR) and circular x-ray dichroism,[3] are of increasing interest as quantitative probes of the magnetization in ultrathin magnetic films and multilayers. In the case of PNR, the well-defined neutron-solid interaction and the wave-vector dependence of the reflected intensity make it possible to determine quantitatively both the artificial magnetic and nonmagnetic structure associated with the individual layers and interfaces. Neutron reflection has been used successfully to determine the magnetic moments in ultrathin films[4,5] and can be extended to study the magnetization profile in multilayers typically on nm-length scales.[6] It has also been shown that vectorial information can be obtained in magnetically nonaligned multilayer structures.[7–9]

In this paper, we show both theoretically and experimentally that information on the magnetization of a single magnetic film can also be obtained if the magnetization vector is perpendicular to the spin orientation of the incident neutron beam. Thus the reflection process is entirely quantum mechanical in origin since in this case no magnetic interaction would be expected classically. First we theoretically analyze the reflectivity matrix for an arbitrary orientation of the spin polarization with respect to the magnetization vector in the film plane and show how the reflectivity depends on the orientation. We then present experimental results for the reflection of polarized neutrons from a single ferromagnetic film where the magnetization direction is aligned perpendicularly to the spin polarization. For this particular geometry, the measured reflected intensity is the same as for an unpolarized incident neutron beam in exact agreement with calculations. We also suggest how the effect could be exploited to obtain magnetic information using an unpolarized neutron beam.

In order to illustrate the difference between the conventional geometry used in PNR experiments and the case of an arbitrary orientation of the magnetization in the film plane, we first recall that the interaction of neutrons with a multilayer sample can be described simply by an effective potential which is different in each layer.[10] It consists of the sum of only two terms, one representing the nuclear component of the interaction, the other representing the magnetic component, the Zeeman interaction. In the jth layer, the interaction potential V_j is given by

$$V_j=\frac{\hbar^2}{2\pi m_n}\rho_j b_j-\boldsymbol{\mu}_n\cdot\mathbf{B}_j, \tag{1}$$

where m_n and $\boldsymbol{\mu}_n$ denote the neutron mass and magnetic moment, and ρ_j, b_j, and $\mathbf{B}_j$ denote the atomic density, coherent nuclear scattering length, and magnetic induction associated with the jth layer. In this paper, we will consider only the case of a single magnetic layer in which the magnetization lies in the plane of the film.

We now describe the spin-dependent neutron reflectivity quantum mechanically to see how the magnetization vector affects the measured reflectivity. The coordinates for the neutron spin and the magnetization vector are defined in Fig. 1. The magnetization lies in the plane of the sample and is oriented along the z' axis. The scattering plane is the xn

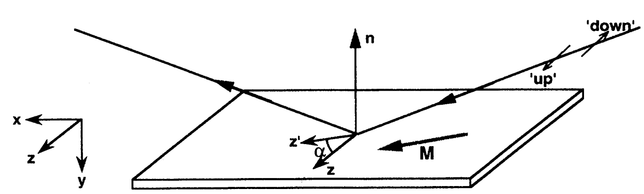

FIG. 1. Scattering geometry for a polarized neutron reflection experiment. The spin polarization of the incident neutron beam is fixed and parallel to the z axis. The magnetization vector **M** in the film plane is parallel to the z' axis and can be rotated around the surface normal (n axis). For clarity the incident angle is greatly exaggerated.

plane and the z axis is the polarization direction of the incident neutron spins. Spins pointing in the positive and negative z direction are referred to as "up" ($\uparrow$) and "down" ($\downarrow$) spins, respectively. The angle α defines the direction of the magnetization (z' axis) with respect to the spin quantization axis (z direction).

In PNR experiments, the spin polarized neutrons are reflected at grazing incidence from the magnetic sample for which the magnetization is aligned by an applied magnetic field. The reflected intensity is then measured for two orthogonal spin states of the incident beam as a function of the incident perpendicular wave vector q. In the conventional geometry, the direction of the applied field which aligns the magnetization is chosen so that the spin polarization of the incident neutron beam is parallel or antiparallel to the magnetization in the film plane (α=0).

We describe the reflectivity process by a matrix $\mathbf{r}(q)$ which connects the incident and reflected spin wave function and is given by

$$\mathbf{r}(q)=\begin{pmatrix} r^{\uparrow\uparrow}(q) & r^{\uparrow\downarrow}(q) \\ r^{\downarrow\uparrow}(q) & r^{\downarrow\downarrow}(q) \end{pmatrix}. \tag{2}$$

The diagonal elements represent the spin conserving reflectivities and the off-diagonal elements the spin-flip reflectivities. They all depend on the polarization direction with respect to the orientation of the magnetization vector, i.e., the angle α. In all experiments without polarization analysis the measured reflected intensities for each incident spin state are $R^{\uparrow}(q)=|r^{\uparrow\uparrow}(q)|^2+|r^{\uparrow\downarrow}(q)|^2$ and $R^{\downarrow}(q)=|r^{\downarrow\uparrow}(q)|^2+|r^{\downarrow\downarrow}(q)|^2$.

In single magnetic films or a multilayer system for which the magnetization vectors of the individual layers are all parallel or antiparallel to each other, the common magnetization direction defines a natural quantization axis for the spins of the incident neutron beam. In this case a diagonal form of the reflectivity matrix in Eq. (2) can always be found which retains only the spin conserving elements. The absence of spin-flipping elements can be readily understood classically because the magnetic induction in the film induces no magnetic torque on the neutron spins when the magnetization and the spin quantization axis are aligned parallel or antiparallel.

For the coordinate system defined above with the magnetization vector along the z axis and parallel to the polarization direction (α=0) the reflectivity matrix can be written as

$$\mathbf{r}(q)=\begin{pmatrix} r^{+}(q) & 0 \\ 0 & r^{-}(q) \end{pmatrix}. \tag{3}$$

This corresponds to the case of the conventional PNR geometry.

When the magnetization in the film plane is oriented along the z' direction at an angle $\alpha\neq 0$, the incident spin wave function can always be decomposed in two components parallel and antiparallel with respect to the z' axis:

$$\begin{aligned} |\uparrow_z\rangle &= \cos\frac{\alpha}{2}\,|\uparrow_{z'}\rangle - \sin\frac{\alpha}{2}\,|\downarrow_{z'}\rangle, \\ |\downarrow_z\rangle &= \sin\frac{\alpha}{2}\,|\uparrow_{z'}\rangle + \cos\frac{\alpha}{2}\,|\downarrow_{z'}\rangle. \end{aligned} \tag{4}$$

The two spin orientations parallel and antiparallel to the z' axis then interact independently with the magnetization in the multilayer system and hence the measured intensities are always given by the weighted sums of the spin conserving reflectivities:

$$\begin{aligned} R^{\uparrow}(q) &= \cos^2\left(\frac{\alpha}{2}\right)|r^{+}|^2+\sin^2\left(\frac{\alpha}{2}\right)|r^{-}|^2, \\ R^{\downarrow}(q) &= \sin^2\left(\frac{\alpha}{2}\right)|r^{+}|^2+\cos^2\left(\frac{\alpha}{2}\right)|r^{-}|^2. \end{aligned} \tag{5}$$

In conventional PNR experiments (α=0) only one incident spin polarization contributes to the reflected intensity. In the other special case, when the polarization direction of the incident neutron spins is orthogonal to the sample magnetization ($\alpha=\pi/2$) the average intensity is measured which is also obtained for an unpolarized incident neutron beam:

$$\begin{aligned} R^{\uparrow}(q)=R^{\downarrow}(q) &= \tfrac{1}{2}(|r^{+}|^2+|r^{-}|^2) \\ &= \tfrac{1}{2}(R^{\uparrow}_{\text{conventional}}+R^{\downarrow}_{\text{conventional}})=R_{\text{unpolarized}}. \end{aligned} \tag{6}$$

In order to show how the spin orientation of the reflected neutrons depends on the wave vector, it is instructive to consider the polarization of the reflected beam for this case when the z' direction is parallel to the x axis ($\alpha=\pi/2$). Using spherical polar coordinates to describe the polarization direction of the reflected beam one obtains

$$\theta=2\tan^{-1}|A(q)| \quad \text{and} \quad \phi=-i\ln\left(\frac{A(q)}{|A(q)|}\right),$$

$$\text{where } A(q)=\frac{r^{+}(q)-r^{-}(q)}{r^{+}(q)+r^{-}(q)}. \tag{7}$$

The azimuth angle ϕ and the polar angle θ are defined in the conventional way with respect to the xyz tripod (see Fig. 1). At low incident momentum, for wave vectors below the critical wave vector, $|r^{+}(q)|^2=|r^{-}(q)|^2=1$ and therefore $\phi=\pi/2$: thus the reflected neutron is polarized in the yz plane. However, it will in general be at some angle θ to the z axis because of the Larmor precession of the neutron in the magnetic layer due to the magnetization along the x axis. For wave vectors above the critical wave vector, both $|r^{+}(q)|^2$ and $|r^{-}(q)|^2$ fall from unity, although by different amounts, and ϕ will no longer take the value of $\pi/2$: the reflected neutron beam acquires a component of spin in the x direction, i.e., parallel to the magnetization vector. Indeed, if $|r^{+}(q)|^2\gg|r^{-}(q)|^2$, $\phi\rightarrow 0$ and the reflected beam becomes totally polarized in the x direction. These effects could be directly probed by using polarization analysis on the reflected beam.[11]

There are therefore two effects at work: first, the Larmor precession of the neutrons when they pass through a ferromagnetic layer,[11] which causes it to precess in the yz plane; second, the reorientation of the neutron spins which become partially polarized in the x direction above the critical wave vector due to reflection at the interfaces. This is analogous to the result of a study by Büttiker,[12] which was concerned with

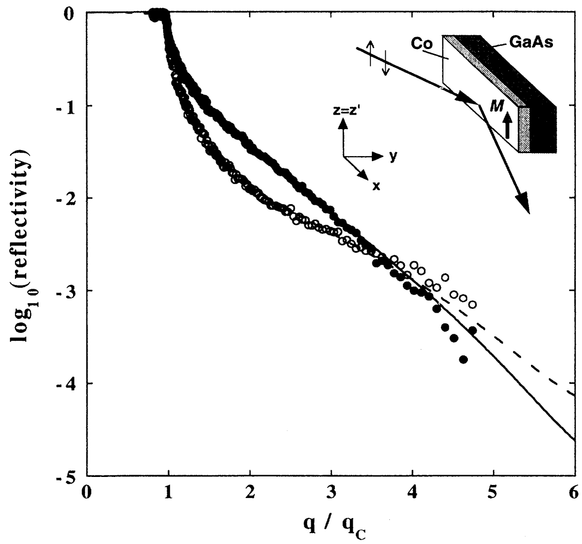

FIG. 2. Measured and calculated reflected intensities as a function of the reduced wave vector from a magnetic 80-Å-thick single Co film on GaAs for the conventional PNR geometry ($\alpha=0$). The critical wave vector $q_c=6.21\times10^{-3}$ Å^{-1} is determined by the substrate material. The solid (empty) circles are experimental data and the solid (dashed) line represents the calculated intensity $R^{\uparrow}(q)$ ($R^{\downarrow}(q)$) for up (down) spin polarization of the incident neutron beam. The inset illustrates the scattering geometry.

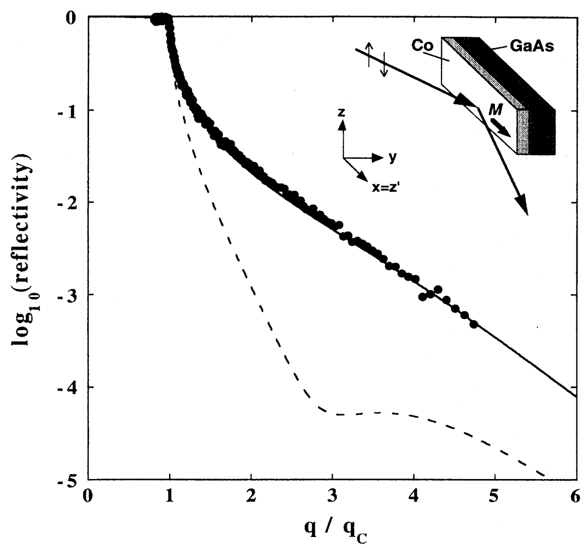

FIG. 3. Measured and calculated reflected intensities as a function of the reduced wave vector from a magnetic 80-Å-thick single Co film on GaAs for the spin polarization of the incident neutrons perpendicular to the magnetization in the film plane ($\alpha=\pi/2$). The critical wave vector $q_c=6.21\times10^{-3}$ Å^{-1} is determined by the substrate material. The dashed line represents the calculated intensity when only the nuclear potential is considered. The solid line has been obtained using the full quantum-mechanical expression. The inset illustrates the scattering geometry.

understanding the traversal time of a spin-$\frac{1}{2}$ particle in a potential barrier in which a magnetic field is confined.

If the sample contains a number of magnetic layers, the magnetization of each of which lie in different directions, the situation becomes more complicated because it is no longer possible to find a basis of spin wave functions in which the reflectivity matrix is diagonal. Spin-flip processes at each interface must then be considered. Nevertheless, it is possible to calculate the reflectivity matrix in this general case using a transfer matrix method.[7,13]

We present results for a Co film deposited onto GaAs(001) under UHV conditions.[14] The PNR measurements were made at 300 K using the time-of-flight polarized neutron spectrometer (SPN-1) at the Joint Institute for Nuclear Research, Dubna. In Fig. 2, we show conventional PNR reflectivity measurements for the incident neutron spin state parallel and antiparallel to the film magnetization ($\alpha=0$). The sample was magnetically saturated in an applied field which was maintained during the reflection measurements. The abscissa in Fig. 2 is the "reduced wave-vector," that is the incident perpendicular wave vector q normalized by the critical wave vector q_c for GaAs, below which only total reflection occurs. In this geometry "up" spin means that the neutron spin is parallel to the film magnetization and hence the neutron moment is antiparallel to the film magnetization. The fitted reflectivity curves were obtained by varying the layer thickness of the Co film and an oxide layer, which is present on top, and by using the magnetic moment for bulk hcp Co of $1.7\mu_B$ for the Co layer. Figure 2 shows simulated reflectivity curves for $|r^+(q)|^2$ and $|r^-(q)|^2$ in the case $\alpha=0$ which correspond to the structure 8 Å CoO/80 Å Co/GaAs. For these thicknesses a good fit to the data is obtained throughout the studied wave-vector range. At large q, the low reflectivities prevent accurate measurements.

We reduced the applied field to a few gauss and then rotated the sample by 90° around its surface normal so that the in-plane magnetization which defines the z' direction is parallel to the x axis, i.e., $\alpha=\pi/2$. This magnetic field was sufficient to maintain the neutron polarization but too small to affect the magnetization in the sample. The spin orientation of the incident neutrons and the magnetization direction in the film are now at right angles (see inset of Fig. 3). The measured and calculated total reflected intensity is shown in Fig. 3. For the fit only the orientation of the sample was changed, all other parameters are identical to those deduced from the data in Fig. 2. The dashed line shows a simulation assuming that the magnetic interaction is switched off (i.e., $\mu_{Co}=0$) which clearly does not fit the data. This case corresponds to a classical treatment of the problem assuming that, since the magnetization and the neutron moment are at right angles ($\boldsymbol{\mu}_n\cdot\mathbf{B}=0$), the only interaction is due to the nuclear part of the interaction [see Eq. (1)]. One should note here that the nuclear reflectivity curve is lower than $R^{\uparrow}(q)$ and $R^{\downarrow}(q)$ due to the particular potentials for the investigated substrate-film system. In many multilayer systems the nuclear reflectivity curve lies between $R^{\uparrow}(q)$ and $R^{\downarrow}(q)$. The solid line in Fig. 3, which follows the experimental data extremely well, was obtained using the quantum mechanically derived Eq. (6). Since both $|r^+(q)|^2$ and $|r^-(q)|^2$ contribute equally to the reflectivity when $\alpha=\pi/2$, one also obtains the solid curve in Fig. 3 by averaging the two calculated reflectivities in Fig. 2 corresponding to an unpolarized incident neutron beam. For $0<\alpha<\pi/2$, the reflected intensity for a polarized and an unpolarized beam are different, however.

We have thus demonstrated that in PNR the magnetic interaction plays a key role even if the polarization direction of the incident neutron spins is parallel to the plane of a magnetic layer on a nonmagnetic substrate, but at right angles to the magnetization vector. For this geometry classically no effect from the magnetization would be expected and the reflected intensity obtained with a polarized or an unpolarized incident beam are equal. The effect arises because the incident neutron beam consists of both eigenstates defined with respect to the sample quantization axis parallel to the in-plane magnetization, and these eigenstates are reflected independently thus leading to an intensity sum for the total reflectivity. For an arbitrary spin orientation of the incident neutron beam with respect to the magnetization direction, polarization analysis of the reflected beam would allow us to determine the absolute orientation of the magnetization vector in the film plane which is not the aim of this work.

Finally, we suggest how the spin orientation dependence of the reflectivity could be exploited in magnetic measurements using an unpolarized beam. First, the reflectivity due to the nuclear potential (the dashed line of Fig. 3) can be measured by magnetizing the sample along the surface normal, since the neutrons are sensitive only to the in-plane component of the magnetization in each layer.[10] Second, the reflectivity due to the spin-dependent potential (the solid line of Fig. 3) can be obtained by magnetizing the film in plane. In this case, the measured reflected intensity is the average of the spin up and down reflectivities [see Eq. (6)], which is not the same that one would obtain if the magnetization is zero as we have demonstrated. Hence, by comparing the reflected intensities for the two magnetization orientations, perpendicular to the film plane and in the film plane, one can directly probe the magnetization with an unpolarized beam.

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