Polarized neutron and X-ray reflectivity study of the structure and exchange coupling of permalloy(Ni$_{80}$Fe$_{20}$)/Cr/permalloy trilayers

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Received 26 September 2002; accepted 26 February 2003

Abstract

The magnetic coupling between two permalloy layers with a Cr separating layer was studied using LMOKE, polarized neutron reflectivity and XRD. The coupling between two permalloy layers is antiferromagnetic-like when the Cr thickness is near 2.0 nm along the easy axis. A strong biquadratic coupling term has been found from the polarized neutron reflectivity study. The strong biquadratic coupling may be caused by the rough interface between the permalloy and Cr layers. The missing GMR effect of this system might be due to this strong biquadratic coupling of permalloy layers.

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PACS: 68.35.Ct; 61.10.Kw; 61.12.Ha

Keywords: Polarized neutron reflectivity; Interlayer antiferromagnetic coupling; X-ray scattering; Permalloy/Cr multilayers

1. Introduction

Recently, the subjects of magnetic coupling and magnetoresistance in ultrathin film structures attract a lot of attention. Antiferromagnetic (AFM) exchange coupling between two ultrathin ferromagnetic (FM) films can be found for a non-magnetic spacer layer of a specific thickness. This is the basis of the phenomenon of giant magnetoresistance (GMR). The GMR effect is known to be highly sensitive to structural parameters such as rms roughness, in-plane correlation lengths, terrace size and shapes, profiles of atomic intermixing, atomic displacements and specific interface. Recently, for a few systems such as Fe/FeSi, Co/Cu, and Co/Ir it has been reported that larger biquadratic coupling results in smaller MR [1–4].
Typically, the exchange coupling per unit area can be expressed as $E = -J_1 \cos(\phi) + J_2 \cos^2(\phi)$, where $J_1$ and $J_2$ are the bilinear and biquadratic coupling constants, respectively, and $\phi$ is the angle between the magnetic moment of the FM layers. Usually, $J_1$ is much larger than $J_2$, but in some specific conditions $J_2/J_1$ can be quite large. For example, in Fe/Cr system $J_2/J_1$ increases from 0.1 to 1 [6]. Other systems such as Fe/Si [7] and Fe/Al [8] also have strong biquadratic contributions. In the calculation model for Fe/Cr system, Stoeffler reported that with interfacial imperfections $J_2$ could even be larger than $J_1$ [9]. The imperfect interface such as interdiffusion or periodic terraces might reduce the coupling strength of the magnetic layer and raise the $J_2/J_1$ ratio. Studying the interface structure is a way of understanding the biquadratic coupling.

The system of permalloy (Py)/Cr multilayers is similar to the well-known Fe/Cr but in the absence of the GMR effect [10]. The absence of the GMR is postulated due to the interdiffusion or alloy form between Py and Cr. However, in this system the AFM-like exchange coupling still appears which implies a GMR effect should exist. Further evidence should be provided to study the origin of the lack of the GMR effect. To study the biquadratic coupling in this system is essential.

2. Experimental and results

Py/Cr multilayers were epitaxially grown on Pt buffered (15 nm) sapphire $\text{Al}_2\text{O}_3(1\overline{1}0\overline{2})$ substrate with different Cr thicknesses and growth temperatures ($T_g$) [11]. These thin films were prepared in a molecular beam epitaxial system with a base pressure lower than $2 \times 10^{-7}$ Pa. The substrate was baked at 1000°C for 1 h before vacuum deposition. The deposition rate was about 0.01 nm/s and the substrate temperature was between 150°C and 200°C. Surface structure and epitaxial orientation of the thin films were monitored by reflection high-energy electron diffraction and also X-ray diffraction (XRD). Figs. 1 and 2 show the XRD result, and the crystal orientation relation is $\text{Al}_2\text{O}_3(1\overline{1}0\overline{2})$$\parallel$$\text{Pt}(1\overline{1}\overline{1})$$\parallel$$\text{Py}(1\overline{1}\overline{1})$$\parallel$$\text{Cr}$$\parallel$$\text{Py}(1\overline{1}\overline{1})$. Fig. 2 shows the orientation of $\{3\overline{1}\overline{1}\}$ of Py and Pt layers with different azimuthal angles. The result indicates that an epitaxy growth was obtained with a major crystalline structure (group A) in three-fold symmetry and a smaller number of crystalline structures (group B) rotated 60° azimuthally.

The magnetic coupling between two Py layers with a Cr separating layer was studied using longitudinal magneto-optical Kerr effect (LMOKE) and polarized neutron reflectivity (PNR). The alternative AFM and FM coupling between two Py layers as a function of Cr thickness can be found. A typical LMOKE result
is shown in Fig. 3(a), which reveals that the coupling between two Py layers is AFM-like when the Cr thickness is near 2.0 nm along the easy axis with two-fold symmetry. Fig. 3(b) shows the hysteresis loop of Py/Cr/Py trilayers at growth temperature of 150°C and 200°C. The saturation field $H_s$ is 190 Oe at $T_g = 150$°C which is obviously larger than that grown at $T_g = 200$°C (79 Oe).

In order to study the atomic and magnetic depth profile of the thin film with different growth temperatures, an X-ray reflectivity and PNR study has been performed at room temperature (shown in Figs. 4 and 5). The X-ray reflectivity experiments were carried out at wiggler BL17A/B beamline of Taiwan Light Source (TLS) and the PNR was measured with POSY1 reflectometer of IPNS in Argonne National Laboratory [12]. The simulation parameters were listed in Table 1 ($T_g = 150$°C) and Table 2 ($T_g = 200$°C). Due to the fact that electron density between the Py and Cr layers is too close, the atomic roughness is hard to determine by X-ray reflectivity alone. PNR served as a complementary tool to determine the interface roughness.

To study the orientation of the magnetic moment of the FM layers, a spin flip analyzer is added to the PNR experiments [13]. A similar study of spin orientation has been reported in Fe/Cr system [14]. However, due to the small magnetic moment of permalloy and poor flipping ratio (less than 16), the signal of the spin-flip neutron is embedded into the depolarized neutron. A correction of the flipping ratio is needed. The effective reflectivity $R^{++}_{\text{eff}}$ or $R^{+-}_{\text{eff}}$ can be obtained from

$$R^{++}_{\text{eff}} = \frac{R^{++}_{\text{exp}} (1 + P) - R^{+-}_{\text{exp}} (1 - P)}{2P},$$
$$R^{+-}_{\text{eff}} = \frac{-R^{++}_{\text{exp}} (1 - P) + R^{+-}_{\text{exp}} (1 + P)}{2P},$$

where the suffix “exp” refers to the experiment data and $P$ as polarization factor of the incident polarization. Typically, the coupling of the two Py

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Fig. 3. The hysteresis loop measured by LMOKE (a) as a function of the azimuthal angle of applied field. (b) $T_g = 150$ and 200°C.

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Fig. 4. The measured and simulation of X-ray reflectivity. $T_g = 150$°C (solid circle) and $T_g = 200$°C (open circle). The solid line is the simulation curve.
layers is FM when the applied field is higher than $H_s$. When the applied field is near 0, the moment of the Py layers is expected to be collinear along the easy axis in the opposite direction. As the applied field is lower than $H_s$, the angle between the magnetic moment of each Py layer is less than 180°. Fig. 6 shows a typical PNR result at an applied field of 14 Oe. The coupling angles between the two Py layers determined from the simulation of the spin asymmetry, $S = (R^{++} - R^{-+})/(R^{++} + R^{-+})$, are listed in Table 3. The coupling angle is neither 180° nor 0°, which implies that a biquadratic coupling term should be included.

3. Discussion

The simulation results, listed in Tables 1 and 2, indicate that the interface roughness between the Py and the Cr layer is nearly 2.0 nm, which is close to the Cr thickness. This implies that either a heavily interdiffused interface or an incomplete and jagged Cr layer was formed. The interface roughness for the sample grown at 200°C is larger than that grown at 150°C and the $H_s$ for the sample grown at 200°C is smaller than that grown at 150°C. Typically, the coupling strength for an ideal sharp boundary sample would be larger than that with an imperfect boundary and the saturation field would be larger [8]. In this work, the reducing of $H_s$ might be due to the larger interface roughness. The large interface roughness or interdiffusion might also reduce the magnetic moment of the FM layer. In Tables 1 and 2, the magnetic moment of the sample that we determined is smaller than the bulk value ($1 \mu_B$), which indicates the Py layer at interface is affected by the adjacent layer [15].

From Table 3, we can see that a large coupling angle between two Py layers indicates the biquadratic coupling is very strong. This strong

![Fig. 5.](image-url)  
Fig. 5. The typical results of PNR without spin-flip analyzer at growth temperature 200°C with applied field along easy axis: (a) applied field = 200 Oe, (b) applied field = 5 Oe. Inset: the spin asymmetry.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Simulation parameters of reflectivity of Py/Cr/Py multilayers with growth temperature 150°C</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Density (g/cm$^3$)</td>
</tr>
<tr>
<td>Capping layer</td>
<td>7.74 ± 2.0</td>
</tr>
<tr>
<td>Ni$<em>{80}$Fe$</em>{20}$</td>
<td>8.2 ± 0.3</td>
</tr>
<tr>
<td>Cr</td>
<td>7.19 ± 0.3</td>
</tr>
<tr>
<td>Ni$<em>{80}$Fe$</em>{20}$</td>
<td>8.2 ± 0.3</td>
</tr>
<tr>
<td>Pt</td>
<td>21.0 ± 0.2</td>
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<tr>
<td>Al$_2$O$_3$</td>
<td>3.987</td>
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</table>
biquadratic coupling might be due to the rough interface of the sample. For all experiments and theoretical predictions so far [16,6], the strength of the biquadratic coupling is larger for the thin film with a less sharp interface. As the previous

experiments show [1–4], this strong biquadratic coupling reduces the magnetoresistance. In this work, no GMR was found. The magnetoresistance of the sample has an anisotropic magnetoresistance of less than 3% [11], in agreement with the earlier report [10].

Another interesting feature is to understand the in-plane symmetry of the magnetic hysteresis loops. By comparing the data of crystal orientation (Figs. 1 and 2) and the magnetic anisotropy in Fig. 3, we can find that the in-plane two-fold symmetric magnetic hysteresis loops are along a three-fold symmetric crystallographic plane. The origin of the in-plane magnetic anisotropy is still a puzzle. The two-fold magnetic anisotropy might be induced by strain from the lattice mismatch of Py(111) on Cr(110), or by step-induced anisotropy. From our grazing incidence X-ray diffraction measurements, there is no detectable strain difference of the Py layer and Pt buffer layer in different in-plane directions. However, the Cr layer is too thin and disordered, so the structure of the Cr layer is hard to determine. The miscut angle of the substrate was also measured and no correlation was found with the magnetic hard-axis direction. Atomic force microscopy measurements were done and no surface steps were observed. The possibility of step-induced anisotropy is also slim. It is interesting to see that the magnetic hard axis is parallel to the vector of the projection of Al2O3(11 10 2) on the plane which is in two-fold symmetry. The influence of the substrate through a 16 nm of Pt buffer layer is quite peculiar. It is possible that the Pt seed layer deposited on the uniaxial Al2O3(1 10 2) substrate has a lattice mismatch and induces a very small strain beyond

<table>
<thead>
<tr>
<th></th>
<th>Density</th>
<th>Thickness</th>
<th>Atomic roughness by X-ray (nm)</th>
<th>Interface roughness by PNR (nm)</th>
<th>Magnetic moment µ₀</th>
</tr>
</thead>
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<tr>
<td>Capping layer</td>
<td>21.0 ± 2.0</td>
<td>0.8 ± 0.4</td>
<td>1.9 ± 0.2</td>
<td>0.6 ± 0.3</td>
<td>0</td>
</tr>
<tr>
<td>Ni80Fe20</td>
<td>8.4 ± 0.3</td>
<td>4.1 ± 0.2</td>
<td>0.5 ± 0.1</td>
<td>2.5 ± 0.4</td>
<td>0.86 ± 0.3</td>
</tr>
<tr>
<td>Cr</td>
<td>7.19 ± 0.3</td>
<td>2.0 ± 0.1</td>
<td>—</td>
<td>2.5 ± 0.4</td>
<td>&lt;0.03</td>
</tr>
<tr>
<td>Ni80Fe20</td>
<td>8.4 ± 0.3</td>
<td>4.1 ± 0.2</td>
<td>—</td>
<td>1.0 ± 0.4</td>
<td>0.86 ± 0.3</td>
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<tr>
<td>Pt</td>
<td>21.0 ± 0.2</td>
<td>13.9 ± 0.2</td>
<td>0.27 ± 0.1</td>
<td>0.3 ± 0.4</td>
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<tr>
<td>Al2O3</td>
<td>3.987</td>
<td>—</td>
<td>0.15 ± 0.1</td>
<td>0.15 ± 0.1</td>
<td>—</td>
</tr>
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</table>

Table 2
Simulation parameters of reflectivity of Py/Cr/Py multilayers with growth temperature 200°C

Table 3
Simulation results of the magnetic moment of the Py layer with different growth temperatures under different applied fields

<table>
<thead>
<tr>
<th>Growth temperature</th>
<th>Applied field (Oe)</th>
<th>Coupling angle (deg.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>200°C</td>
<td>14</td>
<td>60 ± 15</td>
</tr>
<tr>
<td>150°C</td>
<td>10</td>
<td>90 ± 30</td>
</tr>
<tr>
<td>—</td>
<td>100</td>
<td>45 ± 15</td>
</tr>
<tr>
<td>—</td>
<td>150</td>
<td>35 ± 15</td>
</tr>
</tbody>
</table>

Fig. 6. The typical results of PNR with spin-flip analyzer at growth temperature 150°C with applied field 10 Oe along easy axis. Inset: the spin asymmetry.
our measuring capability. Another possibility would be a shape anisotropy induced by the grain shape of the Pt seed layer of this system. Grazing incidence X-ray diffraction also shows this possibility as being negative.

In conclusion, a large biquadratic coupling between Py layers was found by the PNR measurement, which might cause the GMR to be extinguished in this system. The interface roughness plays an important role in the magnetic coupling of FM layers.

Acknowledgements

The hospitality of SRRC operating TLS is gratefully acknowledged. This work was supported by the National Science Council of the Republic of China under Contract No. NSC90-2112-M-007-053. Work at Argonne was supported by the US Department of Energy, Basic Energy Sciences-Materials Sciences, under Contact #W-31-109-ENG-38.

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