

Reflectivity Study of Magnetic Thin Films by Complementary Use of X-ray and Polarized Neutrons

Recently, the subjects of magnetic coupling and magnetoresistance in ultrathin film structures have attracted a lot of attention. An antiferromagnetic (AFM) exchange coupling between two ultrathin ferromagnetic (FM) films usually 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 many structural parameters, especially interface structure and rms roughness. Recently, a larger biquadratic coupling resulting in a smaller MR has been reported in a few systems such as Fe/FeSi, Co/Cu, and Co/Ir. In order to study the atomic and magnetic depth profile of the thin film with different growth temperatures, a Polarized Neutron Reflectivity (PNR) study has been performed, complementing to the synchrotron X-ray reflectivity method.

Unlike X-ray scattering which mainly interacts with electrons, polarized neutron interacts with atomic nucleus and magnetic moment. The expressions for X-ray reflectivity are also valid for the polarized neutron reflectivity as well; only the refractive index needs to be redefined. The refractive index for polarized neutrons is commonly written as $n_{\pm} = n_N \pm n_M = 1 - (N\lambda_n^2/2\pi)(b \pm C\mu)$ where n_N and n_M refers to the refractive indexes from the nuclear and magnetic contributions respectively. N is the atomic number density; λ_n is the neutron wavelength; b is the bound coherent scattering length; μ is the magnetic moment per atom and C is a constant ($0.265 \times 10^{-12} \text{ cm} / \mu_B$). The neutron spin dependent reflectivity, R^+ (spin up) and R^- (spin down) corresponds to the n_+ and n_- , respectively. By fitting the spin-up and spin-down neutron reflection curves, the magnetic depth profile can be uniquely determined. Furthermore, by studying the neutron spin flip of sample and the vector of magnetic moment, the biquadratic coupling between magnetic layers can be determined.

Permalloy(Py)/Cr multilayers, analogous to the well-known Fe/Cr system, do not exhibit GMR effect. The absence of the GMR is postulated to be due to the interdiffusion or alloy formation between Py and Cr. However, in this system, the AFM-like exchange coupling is still present, implying that a GMR effect should exist. Therefore, further studies aimed at clarifying the origin of missing GMR effect as well as the biquadratic coupling should be carried out.

Py/Cr/Py trilayers were epitaxially grown on Pt buffered (15 nm) sapphire Al_2O_3 ($\bar{1}102$) substrate with different Cr thickness and growth temperatures ($T_g = 150^\circ\text{C}$ and 200°C). The

samples were found to be AFM at a Cr thickness of about 2.0 nm using the longitudinal magnetic optical Kerr effect measurement. The saturation field of the sample grown at $T_g = 200^\circ\text{C}$ is smaller than that grown at $T_g = 150^\circ\text{C}$. Surface structure and epitaxial orientation of the thin films were monitored by reflection high energy electron diffraction and also X-ray diffraction (XRD). From the XRD result, the crystal orientation relation is Al_2O_3 ($1\bar{1}02$) //Pt(111)//Py(111)//Cr//Py(111). The X-ray reflectivity experiments were carried out at wiggler BL17A/B beamline of Taiwan Light Source (TLS) and the PNR was measured with a POSY1 reflectometer of IPNS in Argonne National Lab. Due to the fact that the electron densities for the Py and Cr layer are very close, it is hard to determine the atomic roughness by X-ray reflectivity measurement alone. PNR served as a complementary tool to determine the interface roughness.

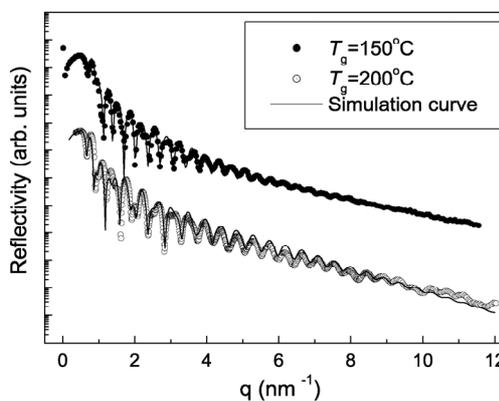


Fig. 1: Measured and simulated X-ray reflectivity curves. $T_g = 150^\circ\text{C}$ (solid circle) and $T_g = 200^\circ\text{C}$ (open circle). The solid line is the simulation curve.

Figure 1 shows the measurement and simulation of X-ray reflectivity at $T_g=150\text{ }^\circ\text{C}$ (solid circle) and $T_g=200\text{ }^\circ\text{C}$ (open circle). The solid line is the simulation curve. Fig. 2 shows the typical results of polarized neutron reflectivity measurement for the sample grown at $150\text{ }^\circ\text{C}$. The measurements were done without a spin-flip analyzer and the applied field was along an easy axis with the field strength of 600 Oe (a), and 10 Oe (b). The inset of Fig. 2 shows the spin asymmetry, $S=(R^+-R^-)/(R^++R^-)$, under different neutron polarizations. The simulation parameters were listed in Table 1 ($T_g=150\text{ }^\circ\text{C}$) and Table 2 ($T_g=200\text{ }^\circ\text{C}$). With a spin flip analyzer added to the PNR experiments, the coupling angle of the magnetic moment on two different Py layers was determined. Both the coupling angles between magnetic moments are 0° or parallel if the applied field is larger than saturation field. However, at low applied field, the angle is about 60° for the sample grown at $T_g=200\text{ }^\circ\text{C}$ and at 14 Oe, and about 90° for the sample grown at $T_g=150\text{ }^\circ\text{C}$ and at 10 Oe. The coupling angle is neither 180° nor 0° which implies

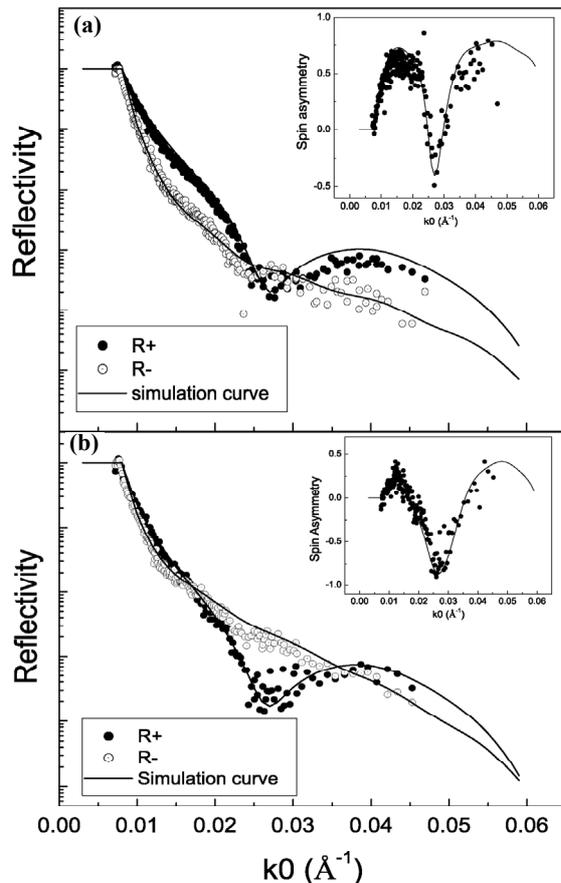


Fig. 2: Typical results of polarized neutron reflectivity measurements without spin-flip analyzer for the samples grown at $150\text{ }^\circ\text{C}$, with applied field along the easy axis. (a) applied field = 600 Oe; (b) applied field = 10 Oe. Inset: the spin asymmetry.

that a biquadratic coupling term should be included. This strong biquadratic coupling might be due to the rough interface of the sample.

For all experiments and theoretical predictions so far, the strength of the biquadratic coupling is larger for the thin film with a less sharp interface. 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 $200\text{ }^\circ\text{C}$ is larger than that grown at $150\text{ }^\circ\text{C}$. 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. 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. The magnetic moment of the samples that we determined is smaller than the bulk value ($1\text{ }\mu_B$), which indicates that the Py layer at interface is affected by the adjacent layer.

Table 1. The simulation parameters of reflectivity of Py/Cr/Py multilayers with growth temperature $150\text{ }^\circ\text{C}$

	Density g/cm ³	Thickness nm	Atomic roughness (by X-ray) (nm)	Interface roughness (by PNR) (nm)	Magnetic moment μ_b
Capping layer	7.74±2.0	0.8±0.4	1.54 ±0.3	0.6±0.3	0
Ni ₈₀ Fe ₂₀	8.3±0.4	4.0±0.2	0.5 ±0.2	0.5±0.4	0.80±0.3
Cr	7.19±0.3	2.5±0.3	---	2.0±0.4	<0.03
Ni ₈₀ Fe ₂₀	8.3±0.4	4.0±0.2	---	1.0±0.4	0.80±0.3
Pt	21.0±0.2	17.1±0.1	0.34 ±0.05	0.3±0.4	0
Al ₂ O ₃	3.987	-----	0.105±0.05	0.13±0.1	-----

Table 2. The simulation parameters of reflectivity of Py/Cr/Py multilayers with growth temperature $200\text{ }^\circ\text{C}$

	Density g/cm ³	Thickness nm	Atomic roughness (by X-ray) nm	Interface roughness (by PNR) nm	Magnetic moment μ_b
Capping layer	16.0±2.0	0.8±0.4	1.9±0.2	0.6±0.3	0
Ni ₈₀ Fe ₂₀	8.5±0.3	4.1±0.2	0.5±0.2	0.5±0.4	0.86±0.3
Cr	7.19±0.3	2.0±0.3	---	2.5±0.4	<0.03
Ni ₈₀ Fe ₂₀	8.5±0.3	4.1±0.2	---	1.0±0.4	0.86±0.3
Pt	21.0±0.2	13.9±0.1	0.27±0.05	0.3±0.4	0
Al ₂ O ₃	3.987	-----	0.15±0.05	0.15±0.1	-----

Although it is possible to discern an intensity variation covering eight orders of magnitude during a synchrotron X-ray reflectivity measurement, the low electron contrast forbids us to determine the interface structure between Py and Cr layers. In comparison, despite a modest capability of only discerning an intensity difference of four orders of magnitude in the present neutron reflectivity measurements, the interface roughness between Py/Cr can still be determined, although not with great accuracy. For a further study of the interface roughness between Py and Cr layers, anomalous X-ray reflectivity measurements were also performed at chosen incident photon energy near Cr, Fe and Ni absorption edges. The experimental spectra of the sample (Cr=2.7 nm) as a function of the incident photon energy were plotted in Fig. 3. The simulation is done under an assumption that the interface roughness profile is a Gaussian smeared step along the surface normal direction. The simulation parameters of the anomalous reflectivity are listed in Table 3. From the simulation result the roughness between the Cr and bottom perm alloy

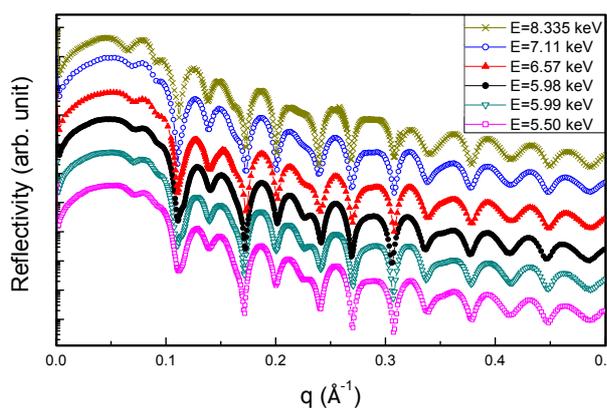


Fig. 3: The anomalous X-ray reflectivity scattering of Py/Cr/Py trilayer sample at different energies of incident beam.

Table 3. The simulation parameters of anomalous reflectivity of Py/Cr/Py multilayers with growth temperature 150 °C.

	Density (g/cm ³)	Thickness (nm)	Roughness (nm)
Pt	21	0.8±0.4	1.1
Ni ₈₀ Fe ₂₀	8.6	4.45±0.2	0.55
Cr	7.19	2.35±0.4	>0.4
Ni ₈₀ Fe ₂₀	8.6	4.45±0.2	<0.35
Pt	21	17.2±0.4	0.33
Al ₂ O ₃	3.987	1E9	0.135

layers is below than 0.35 nm. The roughness of upper interface of the Cr is larger than 0.4 nm.

The inconsistency of the roughness determination obtained by the neutron reflectivity and anomalous X-ray reflectivity is not totally surprising. The smaller dynamical range for a neutron reflectivity measurement seriously limits the resolution of depth profile, while the small scattering factor difference in anomalous scattering gives rise to a large error in determining the roughness. In addition, the X-ray reflectivity determines only the chemical roughness while the neutron reflectivity determines both the magnetic roughness and the chemical roughness. Actually, the magnetic roughness might not always be the same as chemical roughness at interfaces. In the future, three new spallation neutron sources will be commissioned in USA, Japan and Europe. Their higher neutron intensity is expected to ease the problems we face today in determining interface structure of the thin films.

BEAMLINES

17B W20 X-ray Scattering beamline

EXPERIMENTAL STATION

X-ray Scattering end station

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