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A Simple Method to Examine Individual Magnetizations in Fe/Rare Earth Multilayer Films by Polarized Neutron Diffraction

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Magnetic/magnetic multilayer films consisting of Fe and rare earth (RE) metal elements show complex magnetic properties. To investigate these magnetic properties, it is important to estimate magnetic moments of individual layers separately. A simple model to determine the Fe and RE moments in multilayer films from total magnetization and polarized neutron diffraction measurements is introduced. The model is applied to Fe/Pr and Fe/Tm multilayer films. Magnetic properties of these films are discussed.

KEY WORDS: Polarized neutron diffraction/ Multilayer/ Fe/Mg/ Fe/Pr/ Fe/Tm/ Rare earth metal

Introduction

Novel properties are expected in multilayer films which consist of two kinds of metal layers periodically stacked in an atomic scale.[1] In the case of multilayer films consisting of Fe and rare earth (RE) elements, two magnetic layers with dissimilar magnetic properties interact at interfaces. This magnetic interaction modifies the inherent magnetic properties of the constituent metals, leading to various magnetic structures and behaviors.[2,3] Generally speaking, magnetic interactions of Fe and light RE moments at interfaces cause a ferromagnetic alignment of Fe and RE magnetizations, and those of Fe and heavy RE moments cause an antiferromagnetic alignment. Furthermore perpendicular magnetic anisotropies often result from RE atoms at interfaces. Measurements of total magnetic moments by VSM or SQUID are insufficient to investigate such complicated magnetic behaviors of Fe/RE multilayer films which change with applied magnetic fields and temperatures. It is indispensable to separate the contributions of the Fe and RE layer moments to the total magnetization in the study of Fe/RE multilayer films.

Neutron diffraction technique is widely used to elucidate magnetic structures of crystals. In multilayer films discussed so far, the periodic structures of both chemical and magnetic are established by alternative stackings of two metal layers. Therefore neutron diffraction technique is applicable to investigate the magnetic structures of multilayer films. We have developed the experimental methods to investigate magnetic properties of multilayer films by polarized neutron diffraction. In the case of Fe/RE multilayer films, we have successfully separated the magnetic moments of Fe and RE layers in various Fe/RE combinations with combining polarized neutron diffraction and total magnetization

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measurements.[4,5] In this paper, we will introduce a simple model for the separation of the Fe and RE layer contributions to the total moments and show some examples.

Experimental

Fe/RE multilayer films are prepared by ultrahigh vacuum deposition. Fe and RE metals are heated by electron beam guns (E-guns) in the vacuum of 10^{-9}Torr range. The films are grown on kapton (polyimide) and glass substrates cooled down to -50°C at deposition rates of 0.2–0.3Å/s. The film thickness is measured by a quartz thickness monitor and controlled by shutters on E-gun crucibles. Total magnetic moments of the films are measured by a SQUID magnetometer. The films grown on kapton substrates are used for magnetization measurements. The polarized neutron diffraction measurements are carried out with the TOP spectrometer installed at the BSF of the National Laboratory for High Energy Physics (KEK). The films grown on glass substrates are used for measurements. The time-of-flight method is used to measure the diffraction profiles. The direction of the scattering vector is parallel to the film stacking direction. Measuring temperature is varied between 20 K and 300 K by a He refrigerator-type cryostat. External magnetic fields up to 9.4 kOe are applied parallel to the film plane and perpendicular to the direction of the scattering vector by an electromagnet. The diffraction intensities with neutron polarization parallel and antiparallel to the external field are determined from the diffraction profiles.

Model

In this section, we will develop a model which derives magnetic moments of individual magnetic layers in the magnetic/magnetic multilayer films. From experiments, we can obtain the total magnetic moments of the film and neutron polarization P which is determined from neutron diffraction intensities. The definition of P is $P = (I_+ - I_-)/[P_B(I_+ + I_-)]$, where I_+ (I_-) is a neutron diffraction intensity with a neutron polarization vector $\boldsymbol{P}_B$ parallel (antiparallel) to the external field. The polarization P is defined for every diffraction order. Hereafter, however, we will discuss only the first order diffraction.

Now we will consider the relation of the polarization P and the total magnetization M to the magnetic structure of the multilayer film. The definition of the total magnetization M is magnetic moments of the multilayer film normalized to the film volume. As we deal with the first order diffraction of the multilayer period (typically several tens Å), the film is approximated as a continuous scatterer of neutrons. In this approximation, each layer has a nuclear scattering amplitude density of $b_j\rho_j$ (j = Fe or RE) and a magnetic scattering amplitude density of $p_o\rho_j\mu_j(x)\boldsymbol{q}_j(x)$*, where b_j is a nuclear scattering amplitude of j atom, ρ_j a number density of j layer, p_o a magnetic scattering amplitude for a unit magnetic moment (2.69×10^{-13} cm), $\mu_j(x)$ magnetic moment (in Bohr magneton) of j layer at a position specified by x, and $\boldsymbol{q}_j(x)$ a projection vector of $\mu_j(x)$ to the film plane. Here we assume that the chemical modulation is a square wave and the magnetic modulation is an arbitrary form which is expressed by $\mu_j(x)\boldsymbol{q}_j(x)$. From these scattering amplitude densities, the calculation

* Magnetic form factors of atoms are regarded as unity because the scattering vector is small.

of diffraction intensities is straightforward. The nuclear and magnetic scattering amplitudes of j layer, B_j and $\boldsymbol{P}_j$, are calculated as

$$B_j = \int_0^{d_j} b_j \rho_j \exp(iQx)dx \tag{1}$$

and

$$\boldsymbol{P}_j = \int_0^{d_j} p_o \rho_j \mu_j(x) \boldsymbol{q}_j(x) \exp(iQx)dx, \tag{2}$$

where d_j is a thickness of j layer and Q is a scattering vector, $2\pi/(d_{Fe} + d_{RE})$ for the first order diffraction.

The scattering cross-section $d\sigma/d\Omega$ is given as

$$\frac{d\sigma}{d\Omega} = \sum_{j_1}\sum_{j_2} \{B_{j_1}B_{j_2}^* + \boldsymbol{P}_{j_1}\boldsymbol{P}_{j_2}^* + \boldsymbol{P}_B(B_{j_1}\boldsymbol{P}_{j_2}^* + \boldsymbol{P}_{j_1}B_{j_2}^* - i\boldsymbol{P}_{j_1} \times \boldsymbol{P}_{j_2}^*)\} \times \exp[iQ(R_{j_1} - R_{j_2})], \tag{3}$$

where R_j is a position of j layer.

The intensity I_+ (I_-) is proportional to $d\sigma/d\Omega$ with $\boldsymbol{P}_B = +P_B\mathbf{z}$ ($-P_B\mathbf{z}$), where $\mathbf{z}$ is a unit vector parallel to the applied field.

The equations (1)–(3) indicate a general relationship between the magnetic structure of the multilayer $\mu_j(x)\boldsymbol{q}_j(x)$ and the diffraction intensities $I_\pm$ or the polarization P. On the other hand, total magnetization M is related to $\mu_j(x)\boldsymbol{q}_j(x)$ by

$$M = \frac{\mu_B}{(d_{Fe} + d_{RE})} \sum_j \int_0^{d_j} \mu_j(x)\rho_j(\boldsymbol{q}_j(x)\cdot\mathbf{z})dx, \tag{4}$$

where μ_B is the Bohr magneton (9.27×10^{-21} erg/gauss).

So far we treat the continuous model exactly. In Fe/RE multilayer films, some simplifications are reasonable. First $\boldsymbol{q}_j(x)$ is regarded, in most case, to be $\mathbf{z}$ if it is averaged over magnetic domains at the atom layer specified by x. Second $\mu_{Fe}(x)$ is μ_{Fe}^{unif} (independent of x). No detailed form is known about $\mu_{RE}(x)$. However at first approximation, we may put $\mu_{RE}(x) = \mu_{RE}^{unif}$. Under these conditions, the equations (1)–(3) lead to

$$P = \frac{2(b_{Fe}\rho_{Fe} - b_{RE}\rho_{RE})(p_o\rho_{Fe}\mu_{Fe}^{unif} - p_o\rho_{RE}\mu_{RE}^{unif})}{|b_{Fe}\rho_{Fe} - b_{RE}\rho_{RE}|^2 + |p_o\rho_{Fe}\mu_{Fe}^{unif} - p_o\rho_{RE}\mu_{RE}^{unif}|^2}, \tag{5}$$

where we assume b_j is a real number*. M is expressed as

$$M = \frac{\mu_B}{d_{Fe} + d_{RE}}(\mu_{Fe}^{unif}\rho_{Fe}d_{Fe} + \mu_{RE}^{unif}\rho_{RE}d_{RE}). \tag{6}$$

Equations (5) and (6) can be solved with respect to μ_{Fe}^{unif} and μ_{RE}^{unif} and one obtains two sets of solutions as

$$\mu_{Fe}^{unif} = \frac{M}{\mu_B\rho_{Fe}} + \frac{d_{RE}}{(d_{Fe} + d_{RE})} \frac{(b_{Fe}\rho_{Fe} - b_{RE}\rho_{RE})}{p_o\rho_{Fe}} \frac{(1 \pm \sqrt{1 - P^2})}{P} \tag{7}$$

and

$$\mu_{RE}^{unif} = \frac{M}{\mu_B\rho_{RE}} - \frac{d_{Fe}}{(d_{Fe} + d_{RE})} \frac{(b_{Fe}\rho_{Fe} - b_{RE}\rho_{RE})}{p_o\rho_{RE}} \frac{(1 \pm \sqrt{1 - P^2})}{P}. \tag{8}$$

Finally we note that if $\mu_{RE}^{unif} = 0$, one obtain from eq.(5)

* This condition is satisfied in most elements.

$$\mu_{Fe}^{unif} = \frac{(b_{Fe}\rho_{Fe} - b_{RE}\rho_{RE})}{p_0\rho_{Fe}} \frac{(1 \pm \sqrt{1 - P^2})}{P}. \quad (9)$$

This equation is used to check the validity of the model.

Results and Discussion

First we experimentally confirm the validity of the model discussed above. In this purpose, Fe/Mg multilayer films are one of the ideal systems, in which the condition $\mu_{Fe}(x) = \mu_{Fe}^{unif}$ is satisfied well.[6] As Mg metal has no magnetic moments, μ_{Fe}^{unif} is derived from the polarization P with eq.(9). The necessary constants for the calculation are summarized in Table I. On the other hand, μ_{Fe}^{unif} is independently obtained from the magnetization M with eq.(6). Comparisons at 8 and 300 K with changing applied field are shown in Fig. 1.[7]

Table I Numerical constants used in calculations of magnetic moments for individual layers

element	Nuclear magnetic amplitude/10^{-13} cm	number density/ 10^{22} cm^3
Fe	9.51	8.49
Mg	5.33	4.30
Pr	4.4	2.89
Tm	7.05	3.32

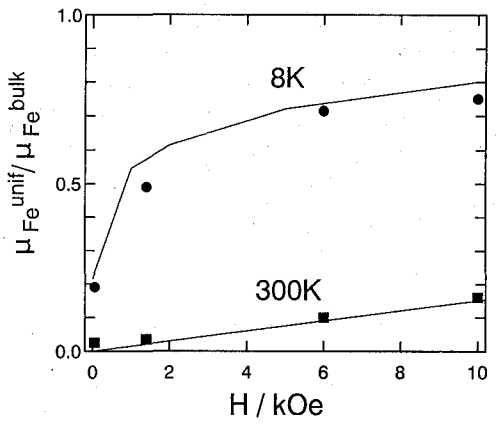

Fig. 1 Comparisons of Fe layer magnetic moments determined by neutron diffraction (● at 8 K and ■ at 300 K) and SQUID measurements (solid lines). The magnetic moment is scaled to the bulk value (2.2 μ_B). The sample is [Fe(8Å)/Mg(20Å)] × 100.

The magnetic moments of the Fe layer obtained by neutron diffraction (● at 8 K and ■ at 300 K) agree well with those obtained from the magnetization measurements by SQUID (solid lines). It is emphasized that there are no adjustable parameters in obtaining the Fe

moments from neutron or magnetization measurements. Figure 1 proves the validity and reliability of the model.

Now we apply the model for the separation of Fe and RE moments in multilayer films. The first example is an Fe/Pr multilayer film ([Fe(26Å)/Pr(18Å)] × 40). The temperature dependence of total magnetization normalized to the film volume and the polarization is plotted in Figs. 2 and 3. In this system, it is known by Mössbauer spectroscopy that the

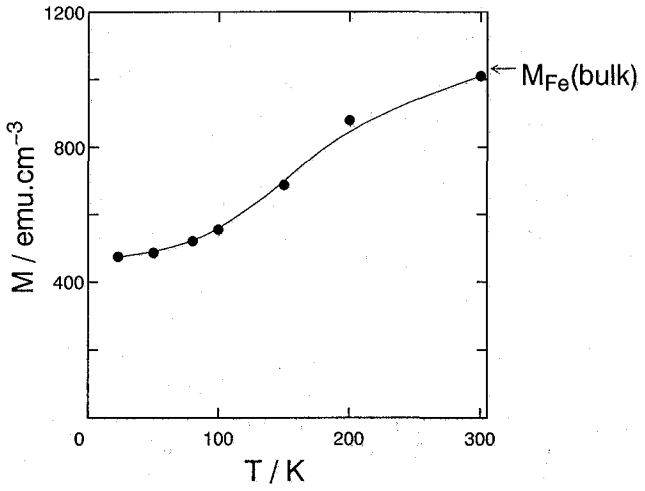

Fig. 2 Temperature dependence of total magnetization in the Fe/Pr multilayer film. M_{Fe}(bulk) corresponds to the magnetization with a full moment of Fe atoms (2.2 μ_B).

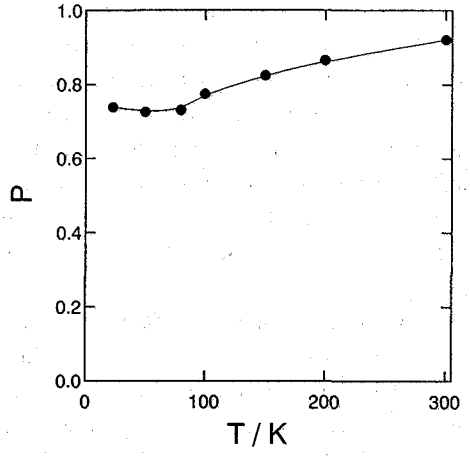

Fig. 3 Temperature dependence of polarization in the Fe/Pr multilayer film.

direction of the Fe moments changes with temperature; at room temperature in-plane and at low temperatures almost perpendicular to the film plane.[8] The total magnetization at

room temperature in the field of 8kOe is almost equal to the calculated Fe magnetization of the bulk α-Fe M_{Fe}(bulk). The magnetization decreases with cooling the sample. The magnetic moments of the Fe and Pr layers are determined from measured M and P. The temperature dependence of μ_{Fe}^{unif} (●) and μ_{Pr}^{unif} (■) is shown in Fig. 4. From eq. (5)

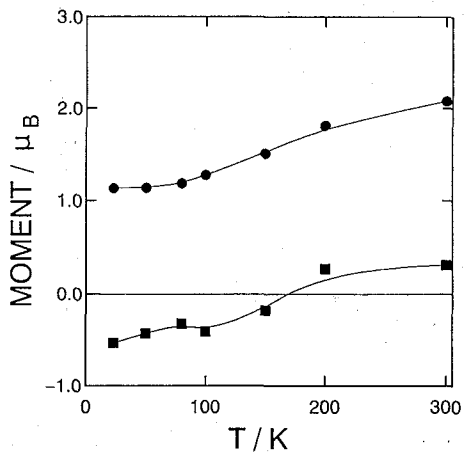

Fig. 4 Temperature dependence of magnetic moments for the Fe (●) and Pr (■) layers determined from polarization and total magnetization measurements.

and (6), two sets of magnetic moments are obtained. In this case, however, the one set, which is not plotted in Fig. 4, is meaningless because it gives unphysically large magnetic moments of both Fe and Pr layers. The magnetic moment μ_{Fe}^{unif} at room temperature is comparable to the bulk value (2.2 μ_B). It decreases with lowering the temperature. It is clear that the magnetic moment of the Fe layer is mostly responsible for the decrease of the total magnetization at lower temperature. The magnetic moment obtained from the model is a parallel component of the magnetic moment vector to the applied field. Therefore the decreased Fe moment at lower temperatures means that the direction of the Fe moment vector is in an out-of-plane direction, which is consistent with Mössbauer results. The magnetic moment of the Pr layer is small even at low temperatures compared to the full moment of Pr (3.2 μ_B). This suggests that the Pr layer has a strong perpendicular magnetic anisotropy.

The second example is an Fe/Tm multilayer film ([Fe(50Å)/Tm(15Å)] × 30). This film shows no perpendicular anisotropy. The temperature dependence of μ_{Fe}^{unif} and μ_{Tm}^{unif} in the applied field of 9.4 kOe is reproduced in Fig. 5. In this case, two sets of the Fe and Tm layer moments are physically acceptable. One is shown by ○ (Fe) and □ (Tm), and the other ● (Fe) and ■ (Tm). The former case indicates that the Fe moment is almost the same as the bulk value and the Tm moment of about 4 μ_B couples antiferromagnetically to the Fe moment. The latter case means that the Fe moment is reduced to about 80% of the bulk and the Tm layer has almost no moment compared to the full moment of Tm (7.0 μ_B). We emphasize that concerning the temperature dependence, several combinations of two sets of solutions at different temperatures are probable; for example (●,

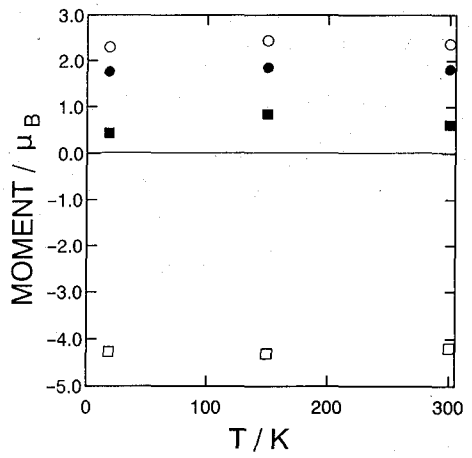

Fig. 5 Temperature dependence of magnetic moments for the Fe and Tm layers determined from polarization and total magnetization measurements. In this case, two reasonable sets of magnetic moments are obtained. One is expressed as ○ (Fe) and □ (Tm), and the other ● (Fe) and ■ (Tm).

■) at 300 and 150 K, and (○, □) at 20 K. Though (●, ■) is the most probable for all temperatures from the hyperfine field of Fe atoms obtained by Mössbauer spectra,[8] the other possibilities cannot be perfectly ruled out. More studies, for example with different applied fields, are necessary to get a conclusion.

Finally we briefly comment on the assumption of $\mu_{RE}(x) = \mu_{RE}^{unif}$. Indeed this is not a case especially at higher temperature. Even in the Fe layer, there is a slight difference in the magnetic moments of interface and inner layer atoms. However from numerical calculations, we find that

$$\int_0^{d_j} \mu_j(x)dx \simeq d_j\mu_j^{unif} \tag{10}$$

for $\mu_j(x)$ which reproduces the observed P and M. Therefore μ_j^{unif} is a good first approximation for the Fe and RE layer moments even if the assumption is not perfectly satisfied.

In summary we have developed a simple model to make an estimation of the magnetic moments of individual layers in Fe/RE multilayer films. The model is applied to several Fe/RE multilayer systems and important magnetic behaviors are indicated.

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