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Neutron study on the subject of giant magnetoresistance effect

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Abstract

Giant magnetoresistance is often discussed with the interfacial roughness in magnetization and/or the structural disorder. Polarized neutrons are particularly powerful for probing such a disordered state through the magnetic and lattice structure analysis. We renew in this paper our polarized-neutron experiments carried out on the TOP spectrometer installed at the pulsed neutron facility, KENS, investigating the magnetic structure relating to giant magnetoresistance. We confirmed that giant magnetoresistance occurs when the magnetic moment is aligned antiparallel, with every other layer separated by a nonmagnetic spacer layer. The diffusive off-specular reflection of magnetic origin suggested that the magnetic disorder near the interface might be related to the appearance of the magnetoresistance effect.

Keywords: Neutron reflection; Polarised neutrons; Gadrinium ytteribium

1. Introduction

On this occasion, we briefly describe the background of the magnetic neutron-scattering studies on magnetic superlattices, and also explain how polarized neutrons contribute to the magnetic structure analysis of magnetic superlattices as well as how it is important for the best understanding of the giant magnetoresistance (GMR) effect, specifically in magnetic superlattices. The paper by Sato et al. [1] was the first neutron-scattering experiment aiming to detect the interfacial effect on ferromagnetism, using multilayered films of stacking bilayers of one ferromagnetic film and another nonmagnetic film. The idea was that the local magnetic density distribution along the perpendicular direction can be derived by the intensity analysis of neutron-diffraction patterns from a multilayered film consisting of perfectly regular structure [2]. This is a direct method to investigate the effect of the interface or surface on the magnetic properties.

The first experiment was done with unpolarized neutrons, and the magnetic scattering was extracted by quantitative comparison of a data set of one with the magnetic field applying perpendicular to the scattering vector ($H \perp Q$) and the other with the magnetic field parallel to the scattering vector ($H // Q$). Magnetic scattering can only be seen in the condition where the magnetic moment has a finite fraction in the plane perpendicular to the scattering vector $Q(M\perp)$. This method should work, if the magnetic moment saturates in the applied field H.

$$I_{H\perp Q}/I_{H\parallel Q} = 1 + (p/b)^2 \qquad (1)$$

where p and b are respectively the magnetic and nuclear scattering lengths.

Polarized-neutron diffraction could determine the magnetic density distribution more accurately by taking a flipping ratio, $R = I^+/I^-$, since polarized-neutron scattering gives polarization-dependent magnetic scattering, as shown in the next equation [2]. In other words, the linear term of p/b is included in the polarized-neutron diffraction intensity, whereas this term is averaged out in unpolarized-neutron diffraction:

$$R = \frac{I^+}{I^-} = \left(\frac{1 + p/b}{1 - p/b}\right)^2 \qquad (2)$$

By using pulsed polarized-neutron beams, we have systematically studied the surface or interface ferromagnetism by using multilayered films on the TOP

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spectrometer installed at KENS, the neutron scattering facility at KEK, Tsukuba [3]. The TOP spectrometer provides pulsed polarized beams, in which polychromatic polarized neutrons of more than 3 Å can be used. Though the quality of the multilayered films in the microscopic structure was not as good as those currently used in neutron experiments, mainly owing to the fact the the growing method was not sophisticated enough in the early 198Os, superlattice reflections were observed up to third-order reflections in the small-angle polarized-neutron diffraction pattern. Thus the effect of the superlattice structure, or the effect on the interfacial magnetism, could be derived, if the intensities were strong enough [4,5].

A short time after the interface or surface magnetism was studied on the TOP spectrometer by using magnetic multilayers, a detailed magnetic structure analysis was made in Brookhaven National Laboratory by using rare-earth metallic superlattices grown by the method of molecular-beam epitaxy (MBE) [6]. Since the crystal quality is like a good single crystal of three dimensions, detailed structure analysis could be made for many satellite reflections around the fundamental reciprocal reflections. In contrast to our experiments measuring small-angle scattering, the conventional diffraction method can be studied for the lattice coherency as well [7]. For instance, ferromagnetic Gd moments were found to be uniformly lined up in parallel way in a Gd layer in Gd/Y superlattices.

The most fascinating nature of the long-range magnetic order is the existence of antiferromagnetic structure in which the aligned Gd moments are in alternating directions with every other layer separated by a Y layer. The antiferromagnetic long-range order depends on the thickness of the intervening Y layer, which was interpreted by the long-range RKKY interaction through the conduction electrons in the Y layer. In this case, the detailed magnetic structure could be well determined by adopting the polarization analysis of the scattered neutrons for emitting polarized neutrons, with spin either up (+) or down (−) with respect to the scattering plane: the spin flip scattering cross-section I^{+-} or I^{-+} contains a component perpendicular to the applied magnetic field, at the same time along neutron polarization, in the plane perpendicular to the scattering vector **Q**. On the other hand, the non-spin flip cross-section I^{++} or I^{--} only detects a component parallel to the neutron spin polarization. Therefore the combined data of both I^{+-} (I^{-+}) or I^{++} (I^{--}) yield an accurate spin structure model, or the magnetic moment direction.

$$I^{+-\ (-+)} = A|\Sigma p_j(q_{xj} + \mathrm{i}q_{yj})\mathrm{e}^{\mathrm{i}\mathbf{Q}\mathbf{r}_j}|^2 \tag{3a}$$

$$I^{++\ (--)} = A|\Sigma(b_j + p_j q_{zj})\mathrm{e}^{\mathrm{i}\mathbf{Q}\mathbf{r}_j}|^2 \tag{3b}$$

where $\mathbf{r}_j$ is the position vector of the jth atom in the unit cell, $\boldsymbol{q}_j = \mathbf{Q}_j(\mathbf{Q}_j\mathbf{M}_j) - \mathbf{M}_j$ and suffixes x, y and z represent the Cartesian coordinates, taking $z \| \boldsymbol{P}$.

It was also found from the polarization analysis that the phase slip occurs at the interface boundary in Ho/Y superlattices [8]. Nevertheless, such an interesting modulation in the long-range magnetic ordered state and in the local phase or amplitude of the magnetic moment is accompanied by strain in rare-earth superlattices. This experimental fact shed light on the important role of magneto-elastic coupling in magnetic superlattices induced by the superlattice structure of the coherent multilayer stacking.

It must be emphasised that these interesting phenomena can be observed not only from the high quality of the superlattices grown by mainly MBE techniques but by the modern technique of polarised neutron diffraction with polarization analysis. The existence of magnetic coupling through thick nonmagnetic metallic layers as well as the interface effect on the magnetism attracted very much as a modern subject in the field of magnetism by the indispensable contribution of such detailed neutron-scattering experiments. It is also believed that this evidence became a trigger to the discovery of the GMR effect of the multilayers [9].

The recent development of the neutron reflection method also contributes to the experimental study on the effect of the interface or surface on ferromagnetism [10]. We believe that there will appear in the near future many important neutron reflection results investigating the interface roughness, in particular of magnetic density fluctuations at the interface by using polarized neutrons. In this text, we only show our experimental studies on the subject related to the GMR effect [11], since the principles of the polarized-neutron reflection method have already been discussed extensively [10,12].

2. Small-angle polarized-neutron diffraction from GMR superlattices

When we started small-angle neutron-diffraction experiments on Fe/Cr superlattice film which showed the GMR effect, we focused on excellent experimental results showing the external magnetic field dependence of both the electrical resistivity and magnetisation [13]. The experimental results indicate that the magnetoresistance effect ($-\Delta\rho$) is approximately proportional to the magnetisation squared (M^2). Although this experimental fact seems to be universal, at least in any Fe/Cr superlattice samples, there was no theoretical interpretation at the initial stage of our studies. We therefore tried to understand the physical origin of this specific relation in terms of the magnetic structure.

Polarized-neutron diffraction in GMR films of Fe/Cr superlattice presented distinct magnetic diffraction peaks of up to third-order superlattice reflections, corresponding to a long-range antiferromagnetic alignment of the Fe bulk moment in the small-angle scattering. The structure analysis was made for the first two intense superlattice reflections [14] in the superlattice film of [Fe(2.7 nm)/Cr(1.2 nm)]$_{30}$, which was grown on a float-glass substrate by slow deposition under ultra-high vacuum (10^{-9} Torr) at elevated temperature of about 300“ ”°C. The neutron-scattering cross-section contains the neutron polarization dependence, and therefore the magnetic structure could readily be determined for the present case of antiferromagnetic long-range order.

$$d\sigma(Q)/d\Omega = B(Q)^2 + P(Q)^2 + \boldsymbol{P}[B(Q)P^*(Q) + B^*(Q)P(Q)] \qquad (4)$$

where $B(Q)$ and $P(Q)$ are respectively nuclear and magnetic structure factors, $\boldsymbol{P}$ is the polarization vector of neutrons and Q is the scattering vector. Note that the dynamic scattering formula is more accurate for small-angle scattering, but the kinematic diffraction theorem was applied for the intensity analysis. We think the formula presented in Eq. (4) is applicable for our purpose.

Nevertheless an important point in our studies is that the cross-term of $B(Q)P(Q)$ gives the fact that the magnetization in Fe layers rotates in the plane towards the magnetic field direction from the antiparallel alignment perpendicular to the external magnetic field (Fig. 1). This fact was also confirmed by the polarization analysis of scattered neutrons that the first-order reflection corresponding to the antiparallel long-range order of the alternating Fe layers is dominated by spin flip scattering. Then we determined the angle of the bulk Fe moment from the applied field in the film plane as a function of H [14].

This result immediately suggests an important conjecture of the proportionality between GMR and the antiferromagnetic component of the Fe magnetic moment, $M_{//}$. In other words, the resistivity change $\Delta\rho = \rho(H) - \rho(H_{\mathrm{sat}})$ is approximately proportional to $\sin^2\theta$, as shown in Fig. 1. $M// = M\sin\theta$. The term of $\sin\Theta$ corresponds to the staggered magnetisation. Except in fine detail, the proportionality seems to hold in all samples grown on different substrates. This point is very important.

We have extended the polarized-neutron diffraction studies to the so-called noncoupled GMR multilayered films of [Co/Cu/NiFe/Cu] structure [15]. This film shows a more pronounced GMR effect in lower magnetic field, which is expected to be applied to

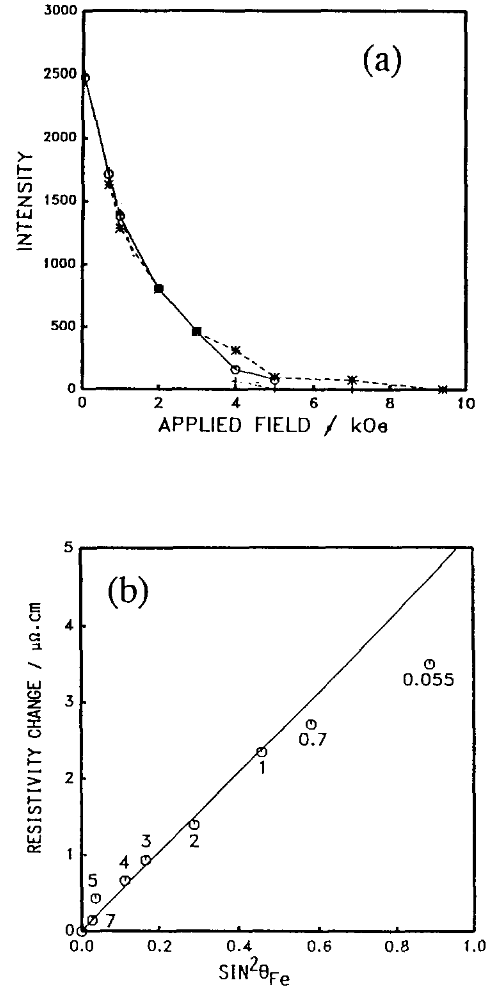


Fig. 1. (a) Peak intensities of the antiferromagnetic Bragg reflection corresponding to antiferromagnetic alignment of Fe bul moment vs. the magnetic field; (b) $\Delta\rho$ vs. calculated angle θ of bul Fe moment. θ is defined between H and M. (After Hosoito et al [14].)

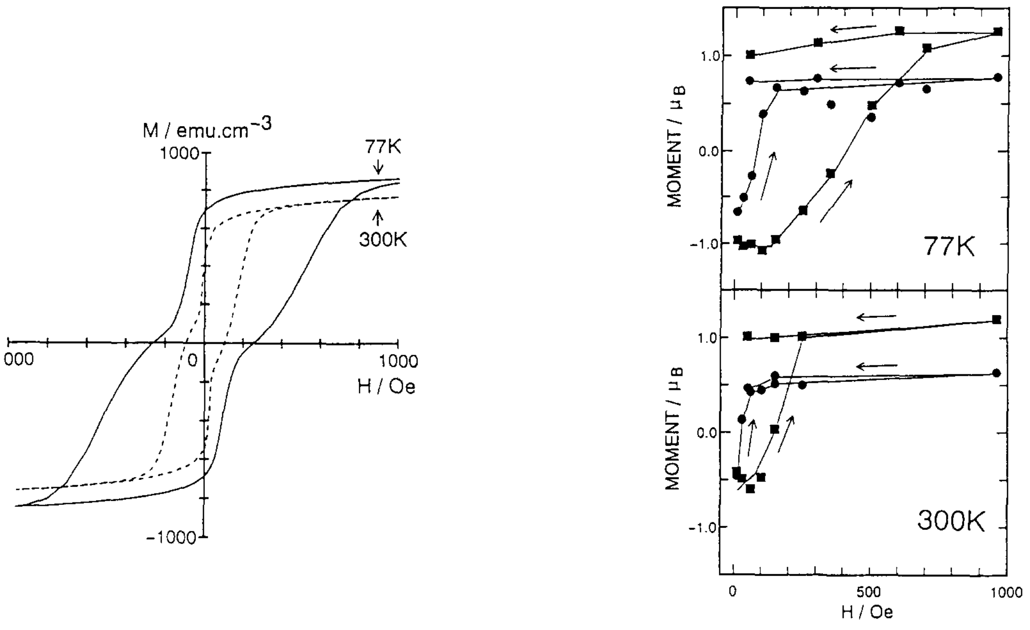
electronic devices such as a switch or a sensor detecting a very small magnetic field [16]. The bulk measurements are consistent with an expectation that th GMR effect may occur when the average Co and NiF magnetic moments seem to align antiparallel. Thi scenario may occur owing to the fact that the C magnetic moment on the Cu substrate shows a rela tively large exchange anisotropy and, on the othe hand, Permalloy or NiFe is well known as a so ferromagnet with very small magnetic coercive forc Then an antiparallel configuration of adjacent Co an NiFe moments occurs in a certain thickness of th intervening Cu layer.

Polarized-neutron diffraction studies were pe formed by using [Co(1 nm)/Cu(3.6 nm)/NiFe(1 nm Cu(3.6 nm)]$_{20}$ grown on a buffer layer of Cr on a gla substrate. A well-defined GMR effect occurs whe the resistance maximum coincides with the abru jump in the magnetisation process [17]. Then t reverse of only the NiFe moment, leaving the Co moment unchanged, is expected with the two-stepwise process of magnetization. However, this intriguing scenario must be experimentally confirmed, and it may be seen that the GMR holds a relation similar to what was found in the Fe/Cr cases. Another interesting feature in the present case of noncoupled GMR films is the large temperature dependence of the GMR effect: the GMR effect is over 40% at 77K, instead of about 10% at room temperature. Note that the magnetisation value only increases by as much as 12% when temperature is reduced below room temperature. We also intended to clarify this point in the neutron studies.

The magnetic field was first applied along the downward direction vertically such that the magnetisation saturated along the field direction prior to the experiment. Then polarized-neutron diffraction measurements were started from zero external field along the hysteresis loop. The experiments were done by counting I^+ and I^- separately. The magnetic field dependence was measured and then the results were analyzed by adopting a simple principle that the polarization-dependent scattering cross-section can be seen at the first-order superlattice reflection. The magnetization along the field direction contributes to the first-order superlattice reflection intensity, and therefore the average or coherent magnetic moment value in each Co, NiFe layer is extracted without any ambiguity.

The result is shown in Fig. 2, in which the bulk moment in each NiFe layer is shown to complete a flop process in a rather low field range up to 200 Oe, and, on the other hand, that more than about 1000 Oe is required to direct all the Co moment to the external field direction at 77 K. Since the coercive force to flip the bulk moment in the Co layer is distinctly higher at lower temperature, the reason why the GMR effect is larger at 77 K than at room temperature can be clarified for the first time by this experiment. This point is quantitatively seen in the intriguing plot in Fig. 3, where the values of ΔI are defined as the squared total moment of the antiparallel-aligned component; $[\mathbf{M}_{\mathrm{Co}}\text{-}\mathbf{M}_{\mathrm{NiFe}}]^2$ is plotted as the function defined as the difference between the measured field resistance and the saturated value; $\rho(H_m,T)-(H_{sat},T)$. The saturation field was postulated to be 0.1 T.

The data points are appreciably classified into two groups: one group is on the straight line of the linear relationship (closed symbols) and the other is a group deviating below the straight line (open symbols). The former group includes only the data in the field range where only the bulk moment in the NiFe layer flip, and the bulk Co moment is still in the antiparallel direction to the external field. Since the NiFe is



(a)

(b)

Fig. 2. (a) Magnetization curves of noncoupled [Co/Cu/NiFe/Cu] GMR film; (b) magnetic field dependence of each Co and NiFe moment derived from polarized small-angle neutron-diffraction measurement. (After Hosoito et al. [17].)

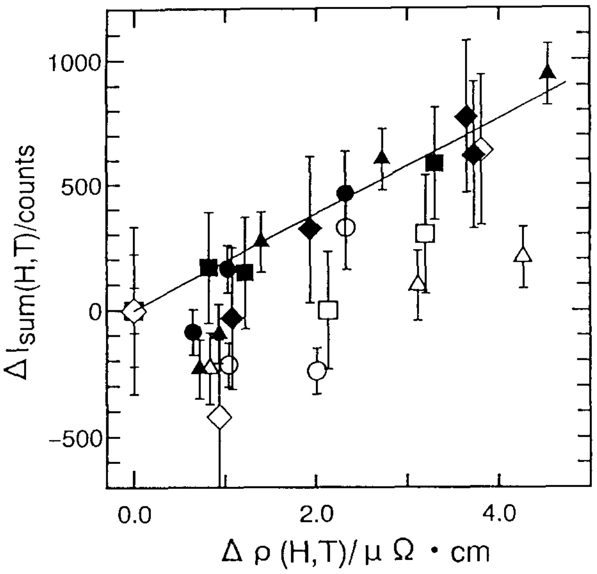


Fig. 3. $\Delta\rho$ vs. neutron diffraction intensities. Closed symbols mean data taken at the lower field range of NiFe moment reversed, and open symbols are in the higher field range, in which Co moments gradually direct along *H*. (After Hosoito et al. [17].)

magnetically soft, a uniform spin flip motion is expected in NiFe layer. On the other hand, each Co moment in the Co layer may not be uniformly lined up along the field direction until the external field reaches a saturated value, owing to the large exchange anlsotropy. Therefore the plot of the squared average total moment of the antiparallel-aligned component might only be meaningful in the lower field region, where the antiparallel alignment of the bulk Co and NiFe moments can be guaranteed. On the other hand, in the process of the rotation of bulk Co moments, the order parameter of $(M_{\mathrm{Co}} - M_{\mathrm{NiFe}})$ might be hard to define.

Nevertheless, examples presented here show important evidence that the GMR effect is closely related to the atomic moment distribution where the average moment is parallel in the ferromagnetic layer and, at the same time, the long-range ordered state exists antiparallel with each adjacent magnetic layer. Then the result suggests that the specific relationship of $\Delta\rho \approx M^2$ should hold even in the noncoupled GMR film of [Co/Cu/FeNi/Cu].

3. Polarized-neutron reflection and the GMR effect

Though polarized-neutron reflection is a simple experimental method, it provides rich information, in particular for the magnetic density profile perpendicular to the film plane of the multilayers [18]. The neutron refractive index $n^{+(-)}$, being the neutron-polarization dependence due to the nuclear and magnetic scattering being of almost same magnitude, gives a good sensitivity in the reflection profile. The refractive index is determined by the scattering potential, and therefore the interface and/or surface effect on the magnetic properties is readily detected. In the simplest case of a magnetic thin film deposited on a nonmagnetic substrate, neutron reflection $R^{+(-)}$ can be calculated in terms of each reflectivity coefficient at each interface, r_{01} or r_{12}. Suffixes 0, 1 and 2, represent respectively the air, magnetic film and nonmagnetic substrate. $k_1^{+(-)} = k_0[n_1^{+(-)} - \cos^2\theta_j]^{1/2}$. θ_j is the reflecting angle.

$$R^{+(-)} = \left|\frac{r_{01}^{+(-)} + r_{12}^{+(-)}\,\mathrm{e}^{2ik_1^{+(-)}t_1}}{1 + r_{01}^{+(-)}r_{12}^{+(-)}\,\mathrm{e}^{2ik_1^{+(-)}t_1}}\right|^2 \qquad (5)$$

The reflection from a multilayered film is extended similarly by means of retardation. We first demonstrated how the magnetic density profile was obtained by polarized-neutron reflection experiments by fitting the specular reflection data to the calculation of the reflectivity mentioned just above. In this analysis of the reflection data from a Ni/Ti multilayer specimen, taken on the TOP spectrometer, a simple square-well scattering potential was adopted in order to avoid any complexity. The multilayered film was grown by the magnetron sputtering method at PSI [18]. The observed wavelength dependence of the polarized-neutron reflection data as well as the flipping ratio are shown in Fig. 4, in which the Ni magnetic moment value and the magnetic-layer thickness were fitted to the experimental data. As shown in the figure, the observed data could be well fitted to the simple model calculation. The existence of the magnetic dead layer might be attributed to the interdiffusion of Ti and Ni near the interface layer.

Nevertheless this experiment demonstrated that the polarized-neutron reflection method is shown to be a promising tool, in particular for measuring the effect of the superlattice structure on ferromagnetism, i.e. the interfacial magnetic effect. In this way, further experiments have proceeded to the present subject on the specific relation of the GMR effect and superlattice structure by using Fe/Cr superlattices.

The specular reflection from the MBE-grown Fe/Cr revealed that the bulk Fe magnetic moment has the long-range-ordered structure of antiparallel alignment as expected. The samples were grown at the NEC laboratory by using a Cr(100) substrate, which was grown on an epitaxially grown Nb(100) buffer layer on either MgO(110) or $Al_2O_3(1102)$ single-crystal substrate [19]. The lattice structure is represented as $[\mathrm{Fe}(100)(3\ \mathrm{nm})/\mathrm{Cr}(100)(1\ \mathrm{nm})]_{80}$.

Besides specular reflection, intense off-specular reflection was observed. This fact was somewhat

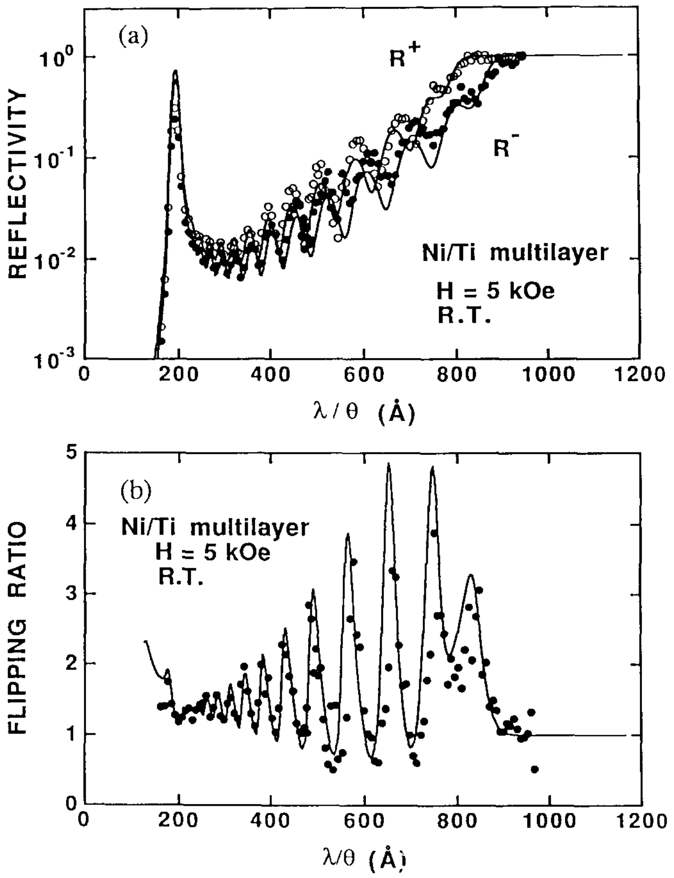


Fig. 4. (a) Polarized-neutron reflection data of Ni/Ti multilayered film, which is used for the neutron polarizer (applied field, 0.5 T); (b) flipping ratio of polarized-neutron reflection (the line was calculated as described in the text). (After Endoh [18].)

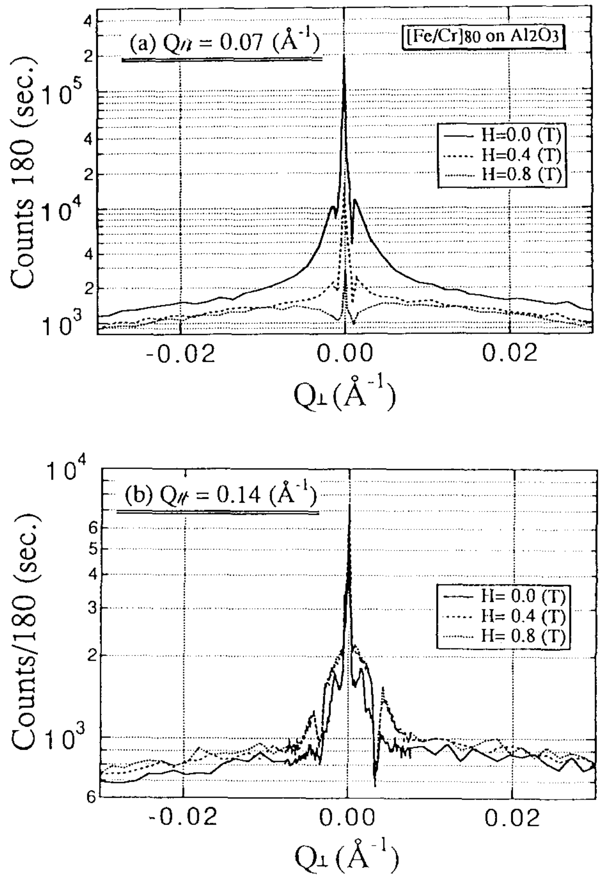


Fig. 5. Neutron diffuse scattering (a) across the antiferromagnetic superlattice point and (b) across the bilayer superlattice point. The various lines shown in the figures are data for different applied fields in the plane. (After Takeda et al. [19].)

surprising, because the lattice structure of the MBE-grown samples is highly coherent in structure, which was detected by the reflection high-energy electron diffraction patterns. Since the off-specular reflection is intense at around the wave vector corresponding to double the bilayer structure — in other words, the period of the antiferromagnetic long-range order of the Fe bulk moment — it might be magnetic in origin. In order to study this point more thoroughly, a specific scan to search the magnetic disorder in the plane was made on the TOPAN spectrometer installed at JRR3M of Tokai establishment of JAEPI. The scan was made along both the specular reflection ($Q\|$) and the perpendicular direction ($Q\perp$), crossing at the antiferromagnetic and bilayer reflection point in $Q\|$, in order to measure the off-specular line shape as shown in Fig. 5 [11].

The characteristic feature is, as seen in the figure, that the intense diffusive off-specular reflection is dependent on the external field, which coordinates with the intensities of the sharp peak of the antiferromagnetic Bragg reflection. The off-specular reflection at Q corresponding to the superlattice structure is less intense, and the intensities increase with the field strength. The result immediately indicates that the off-specular reflection is magnetic.

Except for a quantitative argument described in the previous literature [11], it is reasonably explained that the off-specular reflection component should be correlated with the GMR effect. Integrated intensities of the off-specular diffuse scattering are approximately M^2 of the staggered moment, which also is immediately correlated to the proportionality of $\Delta\rho \approx M^2$. It must be emphasized that the line shape of the diffuse scattering of the off-specular reflection does not show a significant change in the field, but only the scattering intensity is field-dependent. If the line shape is approximated by the lorentzian, the line width gives about 100 lattices of the correlation length, which is far smaller than the length scale of the rocking curve of the Bragg peak (20″ of arc). Therefore we speculated that the off-specular diffuse scattering is dominated by the disordered Fe spins at each interface, which must be responsible for the GMR effect in the exchange-coupled Fe/Cr film.

Finally it should be emphasized here that the present neutron-reflection measurements contain the first experimental evidence showing the strong correlation of GMR and the magnetic interface roughness. Furthermore we argue that the interfacial magnetic roughness observed is not extended more than about 100 lattices in a linear dimension in the plane, which is far smaller than the scale of the ferromagnetic domain size often discussed in this subject. Therefore future studies must clarify this point.

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