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Complimentary Use of Neutron and X-ray Reflection in Study on a Giant Magnetoresistance (GMR) Effect of Magnetic Multilayers

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Neutron and X-ray reflectivity measurements have been now widely used to obtain the structural information on surfaces, interfaces, thin films, layered films and multilayers of all kind of materials. Both neutron and X-ray are important as a probe of reflectivity measurements that have the nanoscale spatial resolution. Since each probe has both merits and demerits in comparison with each other, the complimentary use of both probes in reflectivity measurements gives very important and useful information especially on magnetic structures of magnetic multilayer. In this report we focus on recent results of investigation on Fe/Cr multilayers with different interfacial roughness by Neutron and X-ray reflectivity measurements, which shows the complimentary use is a very promising technique to study magnetic interfacial roughness in magnetic multilayers.

Key words: Reflectivity, Neutron, X-ray, Specular and off-specular reflection, a giant magnetoresistance effect, interfacial roughness

1. INTRODUCTION

A giant magnetoresistance (GMR) effect is first discovered in Fe/Cr multilayers. The effect is the phenomenon that resistivity is changed up to 50% by external magnetic fields [1]. The magnitude is very large compared with that of the conventional bulk magnets (less than 5%). Now the GMR effect is known to occur also in the other magnetic multilayers which consists of alternative stacked magnetic and nonmagnetic bilayers such as Co/Cu, NiFe/Cu, Co/Ag and so on [2-6]. In the multilayers ferromagnetic moments in the magnetic layers are coupled antiferromagnetically across the nonmagnetic layers in the absence of magnetic fields. It has been revealed that the resistivity strongly depends on the alignment of the adjacent ferromagnetic (F) layer. When the antiferromagnetic (AF) alignment is forced to the F one by external fields, the resistivity is effectively reduced.

The GMR effect can be qualitatively explained by the two-current model. In the transition metal like Fe the resistivity of up-spin (ρ_+) and down-spin (ρ_-) conduction electrons are not identical, and one of them is much larger than another one because of spin-dependent scattering. In the F alignment electrons of one of spin state travel in the channel of large resistivity, on the other hand, electrons of another spin state go through in the channel of small resistivity. In the case of the AF alignment electrons of both spin state experience the alternative large and small resistivity channel. Thus total resistivity in the AF alignment is larger than that in the F alignment.

The GMR effect can be understood by the spin-dependent scattering, however, the origin of the scattering is still unclear. In Fe/Cr multilayers it is reported that the interfacial roughness enhances the GMR effect [7]. Therefore it is now concluded that the interfacial roughness plays a key role in the GMR effect in Fe/Cr multilayers [8]. The interfacial roughness, the atomic disorder at the interface, is expected to induce magnetic disorder at the interfaces. Such magnetic disorder may directly affect the GMR effect because the spin-dependent scattering of conduction electrons at the interface is believed to be essential to the GMR effect. However, little attention has been paid to the magnetic disorder at the interface in studying the GMR [9]. Neutron and X-ray specular and off-specular reflectivity measurements are very powerful techniques to investigate such interfacial roughness quantitatively [10]. In order to clarify whether the magnetic roughness governs the GMR effect in Fe/Cr multilayers or not, we have performed neutron and X-ray off-specular reflectivity measurements of epitaxial Fe/Cr(001) multilayers with different interfacial roughhouses that was induced by annealing under the different condition.

2. NEUTRON AND X-RAY AS THE PROBE OF REFLECTIVITY MEASUREMENTS

Because of very small cross section available to magnetic moments, X-ray has not been intensively used for the study of magnetism. The resonant X-ray scattering enables to detect magnetic moments, however, interaction between neutron and magnetic moments are much larger than X-ray. In addition, neutron has much less absorption cross sections to the most of materials than X-ray, which means that neutrons deeply penetrate into material and that the damage of irradiation to specimen, which is significant in the case of hard X-ray, is negligibly small. Thus neutron is potentially an ideal non-destructive probe of magnetism in comparison with X-ray.

However, the luminosity of present neutron sources is much less than that of X-ray emitted by synchrotron sources. The insufficient intensity of incident neutrons makes it difficult to do experiments with well-collimated beams, which are essential to observe fine structures appearing in the profile of off-specular reflection. Therefore the complimentary use of neutron and X-ray is always very useful and sometimes indispensable to investigation on surface and interfacial magnetism of magnetic multilayers.

3. SAMPLES

50 bilayers of [Fe(3.0 nm)/Cr(1.0 nm)] were stacked on the R-plane of sapphire substrate by molecular beam epitaxy [11,12]. The combination of bilayer thickness was optimized so as to obtain the largest magnetoresistance (MR) ratio, $(R_0 - R_f) / R_0$ [1]. Here R_0 is the resistance in the absence of a magnetic filed, and R_f the resistivity in saturation fields higher than approximately 1.0 T.

The multilayer was cut into several pieces whose dimensions were 10×10 mm^2. Changing the annealing time and temperature for each piece, as grown, 6 hours at 623 K, 1 hour at 723 K and 6 hours at 723 K in a vacuum chamber, modified the intrinsic interfacial roughness. Magnetoresistance was measured by a conventional four-probe method at ambient temperature.

The MR ratio is reduced by the annealing procedure, and the MR ratio for each sample is 11.2 %, 9.5 %, 7.1 % and 1.1 %, respectively. The saturation field of the MR was also reduced with the annealing. This indicates that the annealing has enhanced the interfacial roughness and consequently made the AF coupling of the ferromagnetic Fe layers. The enhancement of the interfacial roughness is also suggested by the increase of R_f by the annealing. The reduction of the MR ratio was caused by both the increase of R_f and the decrease of $(R_0 - R_f)$ in all samples.

4. NEUTRON AND X-RAY SPECTROMETERS

Neutron specular and off-specular reflectivity measurements were performed on two spectrometers. CRISP is a pulsed neutron reflectometer, at the Rutherford Appleton Laboratory (RAL) [13]. HER (C11) is a triple axis spectrometer at JRR-3M at the Japanese Atomic Energy Research Institute of Tokai Establishment (JAERI) [14]. The wavelength of the incident neutrons was 0.415 nm by a PG(002).

X-ray specular and off-specular reflectivity measurements were performed on the 3C2 X-ray diffractometer at Pohang Accelerator Laboratory (PAL) in Pohang University of Science and Technology [15]. The photon energy was 8.05 keV tuned by a Si(111) monochromator.

5. RESULTS

5.1 Evidence of correlated magnetic interfacial roughness

A contour map of the unpolarized neutron reflection of the as-grown sample in the $Q_x - Q_z$ reciprocal plane is displayed in Fig. 1. The map was drawn by the measurements on CRISP by using one-dimensional-position-sensitive detector (1D-PSD) as schematically illustrated in Fig. 2. Here the sample plane is defined as the x - y plane and the direction normal to the sample plane as the z axis. No crystallographic and magnetic anisotropy is assumed in the plane, and the arbitrary axis in the plane is defined as x axis. The line along $Q_x = 0.0$ corresponds to the specular ridge of reflectivity.

The peak at $(Q_x, Q_z) = (0.0, 0.8)$ is the Bragg peak from the AF alignment of the ferromagnetic Fe layers and that at (0.0, 1.6) from the Fe/Cr bilayer itself. This figure clearly displays off-specular reflection that spreads along the constant Q_z line running through the AF Bragg peaks. The off-specular reflection indicates

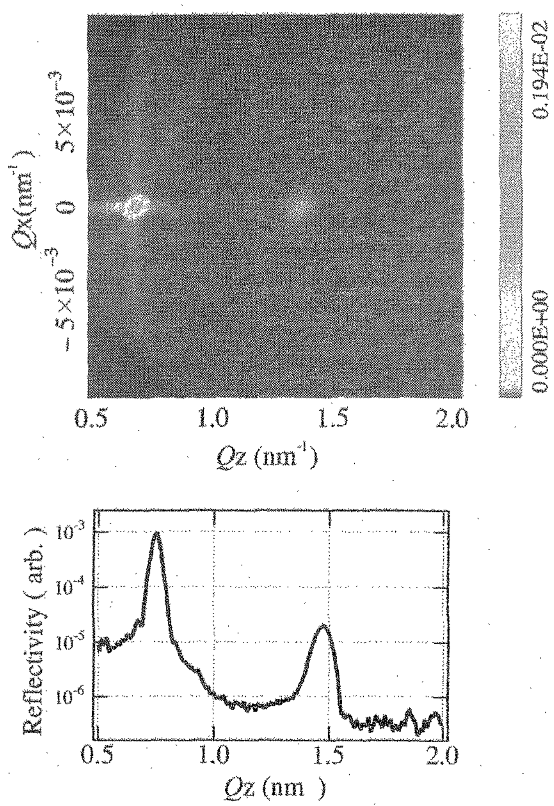


Fig. 1 Upper panel: The intensity map of neutron reflection of the as-grown sample in the $Q_x - Q_z$ plane. Lower Panel: The specular reflectivity for the as-grown sample: The cross section of the upper panel along the line of $Q_x = 0.0$.

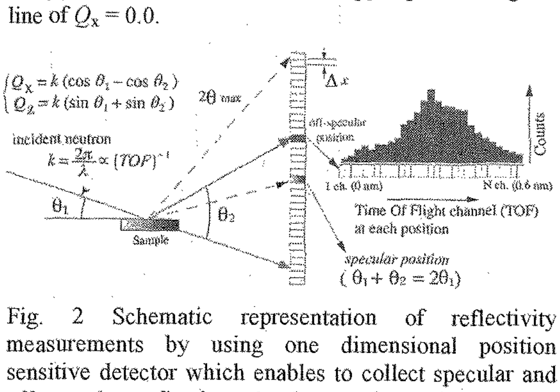


Fig. 2 Schematic representation of reflectivity measurements by using one dimensional position sensitive detector which enables to collect specular and off-specular reflection simultaneously with Time Of Flight (TOF) of pulsed neutrons.

the existence of correlated interfacial roughness from the bottom to the top layer [16, 17]. It should be mentioned that the off-specular reflection at Q_z = 0.8 nm^{-1} is purely magnetic in origin because the peak is an AF one. Such correlated roughness was not clearly observed at the 1st bilayer peak. The off-specular reflection from the correlated magnetic interfacial roughness was also observed at the AF peak in the other samples, however, the profile of off-specular reflection was strongly dependent on the annealing procedures as described in the next section.

5.2 Off-specular reflection from magnetic and atomic interfacial roughness

The profile of transverse scan, the scan along the constant Q_z line, at Q_z = 0.8 nm^{-1} was drastically changed by the annealing as shown in Fig. 3 (a) for the as-grown sample and (b) for the sample annealed at 723 K during 1 hour, respectively. The profile of off-specular reflection at Q_z = 1.6 nm^{-1} contrasted with that at 0.8 nm^{-1}: the annealing does not affect so much on the profiles as shown in Fig. 4.

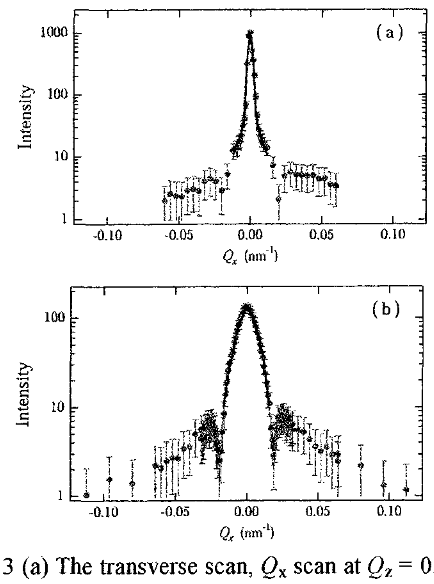
It is noted that the scan range is much wider as compared to the X-ray scans. Due to the strong attenuation of the X-ray beam by the sample there is an unreachable region in reciprocal plane outside of the limits at which the incident angle is zero or the sample plane points to the detector. On the other hand, neutrons are essentially transparent to the samples of interest. Thus signals are still detected beyond the limits as shown in Figs. 3 & 4. Only small dips appeared at the limits of $Q_x = \pm$ 0.02 nm^{-1} and $Q_x = \pm$ 0.072 nm^{-1} in these figures.

The profile of off-specular reflection at Q_z = 1.6 nm^{-1} contains the quantitative information on the atomic disorder at the interface, on the other hand, the profile at Q_z = 0.8 nm^{-1} independently gives us that on the magnetic disorder at the interface. Therefore we can separate the magnetic disorder at the interface from the atomic disorder and then know the correlation among atomic and magnetic interfacial roughness, and the GMR.

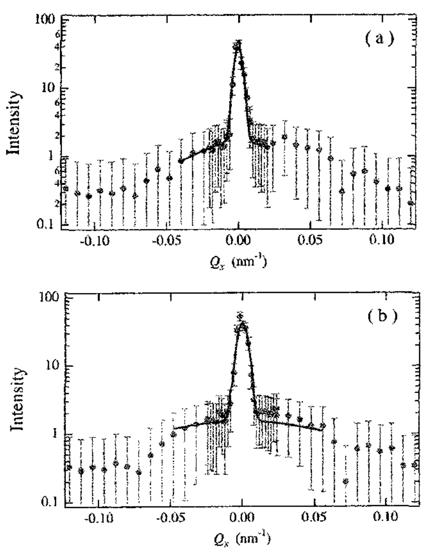
The profile of off-specular reflection from a rough surface can be described by a function in which the rough surface is treated as a self-affine Fractal surface [10]. This approach has been extended to the interfacial roughness in multilayers [16, 17]. Here interfacial roughness is dealt with as a structure factor of a single surface that modifies the reflected intensity from multilayers with a perfect stack as described in Ref. 18.

In this formalism the profile is characterized by three parameters, σ_c, ξ and h. Here σ_c is the rms value of the vertically correlated interfacial roughness from the bottom to the top layer, ξ is the lateral in-plane correlation length or effective cutoff length of the Fractal surface, and h is the Hurst parameter describing how jagged the roughness. The lines in the figures are the fitting curves using this structure factor in the case of h = 1/2.

We found the followings by the fitting. Bulk parameters, magnitude of AF coupling of ferromagnetic Fe layers across Cr layer and magnetic domain sizes, are directly connected to the MR ratio. On the other hand, roughness parameters, which characterized the magnetic interfacial



Fib. 3 (a) The transverse scan, Q_x scan at Q_z = 0.8 nm^{-1}, of the as grown sample. (b) The same scan for the sample annealed at 723 K during 1 hour.



Fib. 4 (a) The transverse scan at Q_z = 1.6 nm^{-1} of the as grown sample. (b) The same scan for the sample annealed at 723 K during 1 hour.

roughness, were very sensitive to the annealing process. However, those for the atomic interfacial roughness were less sensitive. This suggests that the magnetic interfacial roughness is dominant over the atomic interfacial roughness for the spin-independent electron scattering at the interface which is a disadvantage to the GMR effect.

Although we obtained the quantitative information on the roughness, the fitting is less reliable because of the poor statistics and resolution power as shown in Fig. 4. In order to obtain the more reliable parameters of the atomic interfacial roughness, we performed the specular and the transverse scans through the bilayer peak and the higher order peaks by using 3C2 X-ray spectrometer at PAL. Figure 5 shows the results of the as-grown (a) and the most annealed sample at 723 K during 6 hours (b). Because of much higher Q resolution, fine structures in specular reflectivity profiles appear in the figures, and the structures make it hard to fit both specular and off-specular profiles simultaneously. The fitting procedure is now in progress and the results will report elsewhere.

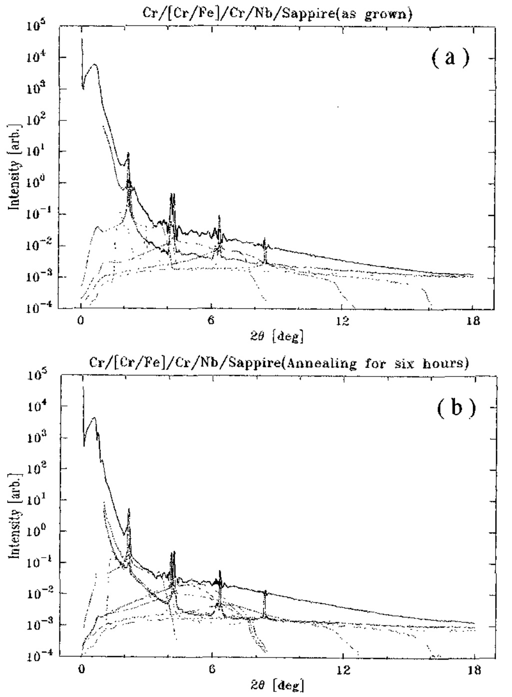


Fig. 5 X-ray specular and off-specular reflection of the as-grown sample (a) and the most annealed sample (b). Several offset scans are also displayed in the figures.

6. CONCLUSION

Specular and off-specular reflctivities of Fe/Cr multilayers with different interfacial roughness were measured using both neutron and X-ray in order to elucidate quantitative relation among atomic and magnetic interfacial roughness, and the GMR effect. It is revealed that bulk parameters such as magnitude of AF coupling and magnetic domain size, are directly connected to the MR ratio and that the roughness parameters, which characterized the magnetic interfacial roughness, were very sensitive to the annealing process. This suggests that the magnetic interfacial roughness is dominant over the atomic interfacial roughness for the spin-independent electron scattering at the interface which is a disadvantage to the GMR effect. However, complementary use of X-ray is indispensable to get more reliable quantitative data especially for the atomic interfacial roughness.

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