Pressure-Induced Enhancement of Giant Magnetoresistance due to Crossover of Interlayer Exchange Coupling in Fe/Cr Multilayers

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We report the first observation of a large pressure-induced enhancement of giant magnetoresistance (GMR) in magnetic multilayers (MML). In Fe/Cr MMLs with the Cr layer thickness of ~30 Å, a crossover from biquadratic to bilinear interlayer exchange coupling (IEC) was observed by applying pressure, and simultaneously the GMR under high pressure (>2 GPa) was enhanced to be twice as large as that at ambient pressure. The enhanced GMR is attributed to the suppression of the biquadratic IEC by applying pressure, and the electrical resistivity in parallel alignment of magnetization also showed a crossover behavior, suggesting an electronic origin for the observed pressure effects.

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Interlayer exchange coupling (IEC) is a novel phenomenon in layered magnetic nanostructures, which is closely related with the electronic structures of spacer materials and is not fully understood [1–3]. Particularly, the mechanism of the so-called biquadratic coupling is still an open question [4]. Fe/Cr multilayer is one of the most important systems because both IEC and giant magnetoresistance (GMR) were first discovered in Fe/Cr [5,6] and a great number of studies have been done to understand the IEC and GMR in this system. The IEC of metallic multilayers (MMLs) generally shows an oscillatory behavior as a function of spacer-layer thickness. Ferromagnetic (F) and antiferromagnetic (AF) coupling appear alternately as a function of the thickness of spacer layers [7,8]. Moreover, the biquadratic coupling often appears associated with the normal AF (bilinear) coupling and the strength of the biquadratic coupling is sometimes comparable to that of the bilinear one [9,10]. The biquadratic coupling stabilizes the spin arrangement with relative angle of 90° between magnetization of two neighboring ferromagnetic layers separated by a spacer layer, while normal AF (bilinear) coupling stabilizes that having the angle of 180° between them. Although GMR and IEC have different physical origins, GMR reflects the properties of IEC. Since the magnetoresistive change is related to the relative angle between magnetization of ferromagnetic layers, GMR also shows an oscillatory behavior with the same periodicity as IEC.

Application of high pressure is a unique method to get a deep insight into physical properties of materials, e.g., heavy-fermion systems, manganese perovskites, and so forth [11,12], because the electronic states, e.g., density of state, Fermi surface, and bandwidth, etc. can be continuously varied under high pressure as well as the volume of the materials. In the case of MMLs, the pressure dependence of GMR has been investigated for Fe/Cr and other MMLs [13,14]. However, dramatic pressure effects, such as an enhancement of GMR, were not observed in the previous experiments. In the present work, we have performed extended studies for Fe/Cr MMLs with a wider range of t_{Cr}, and have discovered novel pressure effects on GMR, IEC, and electrical resistivity in the Fe/Cr MML with $t_{Cr} \sim 30$ Å.

$[Fe(20 Å)/Cr(t_{Cr} Å)]_{20}$ MMLs with $t_{Cr} = 8$–36 Å were prepared on Si (111) substrates by using dc magnetron sputtering with a base pressure of $\sim 10^{-9}$ Torr. The electrical resistivity under hydrostatic pressures was measured with a standard dc four-probe method. The typical size of samples is about $0.3 \times 0.7 \times 0.2$ mm^3 including a Si substrate. The direction of applied magnetic field between −2 T and +2 T is parallel to the plane of Fe/Cr MMLs. The magnetoresistance (MR) ratio is defined as $\Delta\rho/\rho_s = (\rho(H) - \rho_s)/\rho_s$, where ρ_s and $\rho(H)$ are the electrical resistivities in the saturated and unsaturated states, respectively. An approximate saturation field H_s is defined as a magnetic field at which the MR extrapolated linearly from lower field intersects with that from high field as shown in an after-mentioned Fig. 2. High pressure up to 8.0 GPa was generated using a piston-cylinder and cubic-anvil type apparatus utilizing the conventional Teflon-cell technique. The pressure inside the cell was kept constant by controlling the load of hydraulic press. The details of the high pressure apparatus were described previously [15,16].

Figure 1 shows the magnetization (M-H) curves at 4.5 K and ambient pressure for $[Fe(20 Å)/Cr(t_{Cr} = 10 Å)]_{20}$ (a) and $[Fe(20 Å)/Cr(t_{Cr} = 30 Å)]_{20}$ (b) MMLs [hereafter, abbreviated as Fe/Cr(10) etc.], where the MMLs with $t_{Cr} = 10$ Å and 30 Å correspond approximately to the first and second peaks of IEC and GMR, respectively, in Fe/Cr MMLs. The saturation fields H_s are much larger than that expected from the magneto-crystalline anisotropy of Fe ($H_s < 0.01$ T), and they indicate the presence of AF coupling in both Fe/Cr MMLs. However, the shapes of the

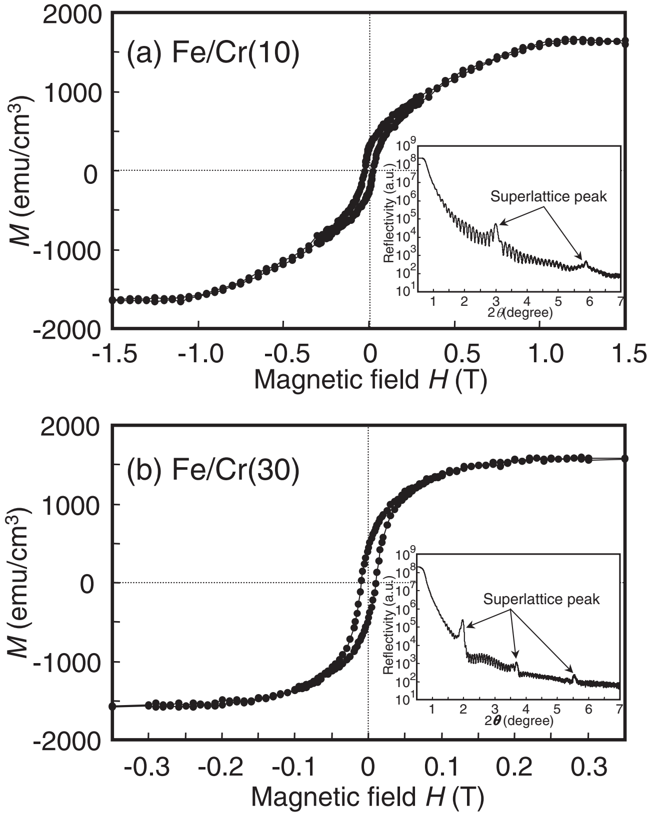

FIG. 1. Magnetization M as a function of magnetic field H at 4.5 K for (a) Fe/Cr(10) MML and (b) Fe/Cr(30) MML. The insets show low angle x-ray diffraction pattern spectra at room temperature.

M-H curves are different: the M-H curve for Fe/Cr(30) is more rounded than that for Fe/Cr(10). From the model calculations on biquadratic IEC [17,18], this fact shows that the contribution of the biquadratic coupling is comparable to that of bilinear coupling in the Fe/Cr(30) MML while the bilinear coupling is dominant in Fe/Cr(10). The insets of Fig. 1 show low angle x-ray diffraction pattern for the Fe/Cr MMLs. For Fe/Cr(10), higher frequency Kiessig [19] fringes due to the total thickness and two superlattice Bragg peaks are observed at $2\theta = 3°$ and 5.8°. On the other hand, for Fe/Cr(30), three Bragg peaks due to superlattice are observed at $2\theta = 2°$, 3.7°, and 5.5°. As shown in the insets of Figs. 1(a) and 1(b), for Fe/Cr(30), the decrease in amplitude of higher frequency Kiessig fringes is observed in comparison with Fe/Cr(10). The roughness of Fe/Cr(30) is somewhat larger than that of Fe/Cr(10) because the reduction of amplitude of Kiessig fringes represents the increase in the surface and interface roughness [20].

Under high pressure, generally, M-H curves of thin film samples with only a small magnetization are difficult to measure. Thus, magnetization process of thin films under high pressures has hardly been investigated to date. However, MR measurements make it possible to evaluate the magnetization process of thin films even under high pressure. Figure 2 shows MR curves of the (a) Fe/Cr(10) and (b) Fe/Cr(30) MMLs under different pressures. For Fe/Cr(10) MML, it was found that the peak value of MR ratio, i.e., $(\Delta\rho/\rho_s)_{\max}$ at 2.5 GPa is slightly smaller than that at 0.1 GPa, while H_s at 2.5 GPa is larger than that at 0.1 GPa [21]. $(\Delta\rho/\rho_s)_{\max}$ is defined as $(\Delta\rho/\rho_s)_{\max} = (\rho(H)_{\max} - \rho_s)/\rho_s$, where $\rho(H)_{\max}$ is the maximum value of resistivity in applied field at H near zero tesla. In contrast, for Fe/Cr(30) MML, the most striking feature of the measured MR curves is the large enhancement of GMR at high pressure: the MR ratio observed at 2 GPa (=31%) is twice as large as that at ambient pressure (=15%). Such a drastic change in MR ratios due to structural modifications has never been reported so far. Furthermore, it is also found that the shape of MR curves is changed significantly by applying pressure. At ambient pressure, the MR curve has sharp pointed peaks around

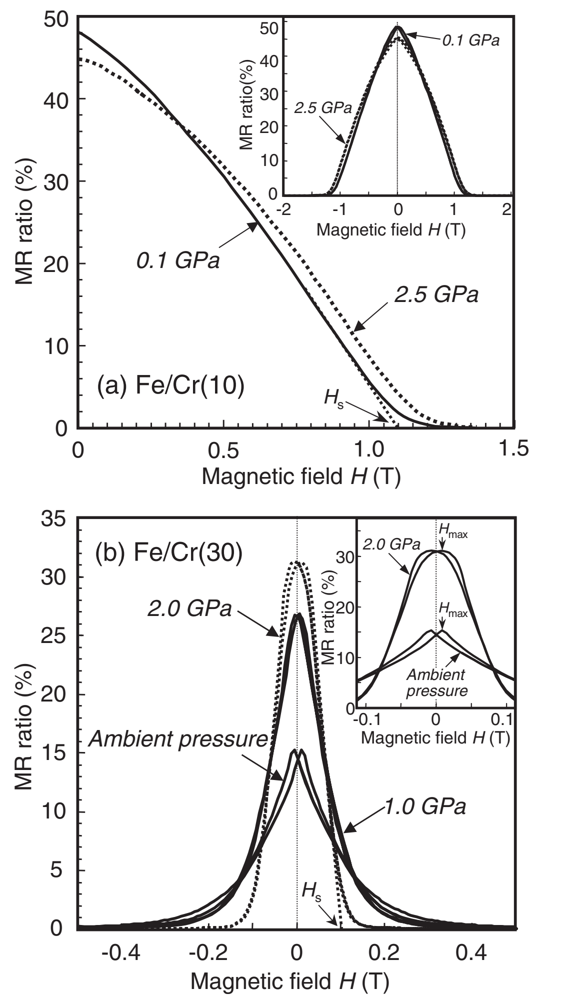

FIG. 2. (a) Magnetoresistance curves of Fe/Cr(10) MML at 4.2 K as a function of the magnetic field at various pressures. (b) Magnetoresistance curves of Fe/Cr(30) MML at 4.2 K as a function of the magnetic field at various pressures. $H_{\max}$ indicates the magnetic field at the maximum value of MR curve.

$H = 0$ T as is clearly seen in Fig. 2(b). With increasing pressure, on the other hand, the MR curve becomes rounded around $H = 0$ T. This remarkable change in the shape of MR curves is well understood by considering the variation of IEC from biquadratic to bilinear coupling by applying pressure: it is clearly shown by model calculations [17,18] that MR curves show pointed peaks in biquadratically coupled MMLs while MR curves show rounded shapes in bilinearly coupled MMLs. For comprehensive discussion, let us consider the angular dependence of MR, which is described by $\Delta\rho \sim \sin^2(\theta/2)$, where ρ is the resistivity and θ is the angle of spins between adjacent ferromagnetic layers [22]. In biquadratically coupled MMLs, this expression can be expanded as $\Delta\rho \sim 1/2 - (1/2)(\pi/2 - \theta)$ around $H = 0$ T, i.e., $\theta = \pi/2$. By using the linear dependence of θ on H around $H = 0$ T, we obtain the relation, $\Delta\rho \propto H$, indicating the linear dependence. On the other hand, in bilinearly coupled MMLs, the parabolic relationship $\Delta\rho \sim 1 - (1/2)(\pi - \theta)^2$ is obtained around $H = 0$ T, i.e., $\theta = \pi$. A similar calculation leads to $\Delta\rho \propto H^2$, indicating the rounded shape of MR curves. The observed enhancement of MR is naturally given by the variation of IEC. Because $\Delta\rho \sim \sin^2(\theta/2)$, MR in bilinearly coupled MMLs ($\theta = \pi$) should show that the magnitude of MR is twice as large as MR in biquadratically coupled MMLs ($\theta = \pi/2$).

Figure 3(a) shows the electrical resistivity at $H = 0$ T and above H_s, ρ_0, and ρ_s, as a function of pressure for Fe/Cr(10) and Fe/Cr(30), where ρ_s for Fe/Cr(10) and Fe/Cr(30) are $\rho(H = 2\text{ T})$ and $\rho(H = 1\text{ T})$, respectively. ρ_0 and ρ_s decrease with increasing pressure, which is the common behavior in metallic systems. However, for Fe/Cr(30), the pressure coefficients of ρ_0 and ρ_s are definitely changed around 2 GPa, suggesting that a crossover in the electronic state occurs. Figure 3(b) shows H_s as a function of pressure for these two MMLs, where it is noted that H_s roughly corresponds to the magnitude of nonferromagnetic, i.e., bilinear and biquadratic IECs. Up to ~2 GPa, H_s for Fe/Cr(30) decreases in accordance with the steep reduction in the biquadratic IEC, in other words, the crossover from biquadratic to weak bilinear IEC. The decreasing rate of H_s is much reduced when the bilinear IEC becomes dominant above 2 GPa. Figure 3(c) shows the MR ratio [approximately the difference between ρ_0 and ρ_s shown in Fig. 3(a)] for the present two MMLs at 4.2 K as a function of pressure, and Fig. 3(d) shows the MR ratio for Fe/Cr(30) at 4.2 K and 296 K. The pressure-induced enhancement of GMR as shown in Fig. 2 is clearly observed for Fe/Cr(30), and it is interpreted as a result of the crossover from biquadratic to bilinear IEC, as mentioned in the above paragraph. It is also found that pressure-induced GMR is observed at room temperature. The present result of Fe/Cr(10) is qualitatively in agreement with the previous experiments [13,14].

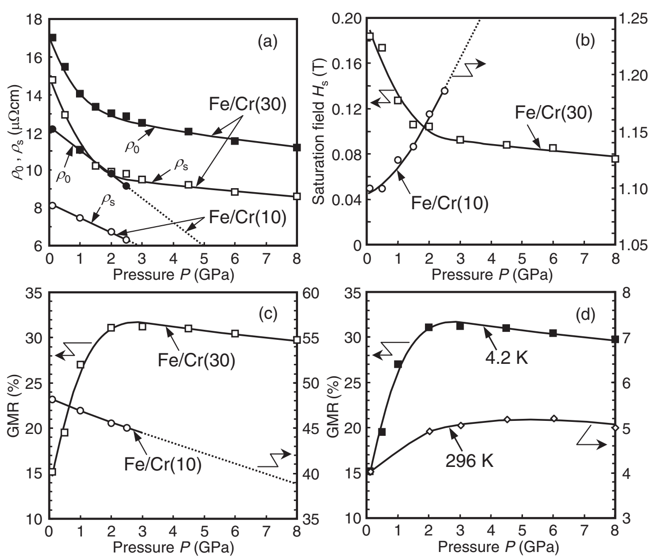

FIG. 3. ρ_0 ($H = 0$ T) and saturation resistivity ρ_s (a), saturation field H_s (b), and the GMR (c) at 4.2 K as a function of pressure for Fe/Cr(10) and Fe/Cr(30) MMLs. The solid lines serve only to connect the data and the dashed lines of Fe/Cr(10) MML is represented as the one expected data at higher pressure ($P > 2.5$ GPa). (d) The GMR at 4.2 K and 296 K as a function of pressure for Fe/Cr(30) MML.

Although it is consistently explained that the observed pressure dependence of H_s and MR originates from the crossover of IEC, the crossover behavior of ρ_s seems to be difficult to explain on the basis of a change in IEC. Since ρ_s is a physical quantity for the parallel magnetization state realized above H_s, it should not depend on which coupled state (90° or 180°) is stable at $H = 0$ T; i.e., the observed behavior of ρ_s should not be interpreted only as a result of the crossover of IEC. Because the electrical resistivity generally reflects electronic states at Fermi surfaces, we consider that the present result of ρ_s shows a possible crossover of the electronic states and its importance, although the change in the electronic structures of MMLs at high pressure has not been emphasized until now. If we assume that E_F of Fe/Cr(30) shift around some peaks of density of states (e.g., $3d$ virtual bound states) by applying pressure, depending on relative position of the peaks to E_F, the pressure may increase or decrease ρ_s.

The mechanism of the biquadratic IEC has been argued since its discovery. Slonczewski showed theoretically that the interface roughness causes biquadratic IEC [10]. However, the interface roughness is hardly considered to be changed by applying pressure as well as the IEC because the pressure-induced strain of Fe/Cr MMLs is estimated from the compressibilities of bulk Fe and Cr to be only 0.4% at 2 GPa. Because no plastic deformation is observed, the atomic configuration should not be significantly changed in the Fe/Cr MML and only elastic strain of ~0.4% can be considered under high pressure. The strain of ~0.4% leads to the change of 0.1%–1% in magnetostatic energy in the roughness-induced model. Therefore, the roughness is less suitable to be the main origin of the pressure effect of biquadratic IEC observed for Fe/Cr(30). Slonczewski also proposed the loose spin model for biquadratic IEC [4] and suggested that the mechanism of this model can be regarded as an intrinsic mechanism in electronic structures of MMLs [23] while the interface roughness gives an extrinsic mechanism. Edwards *et al.* [24] and Erickson *et al.* [25] studied the intrinsic mechanism of biquadratic IEC by calculating electronic structures of MMLs. By considering the intrinsic mechanisms based on the electronic structures, the whole observed results for Fe/Cr(30) shown in Fig. 3 may be consistently understood; i.e., if a crossover in the electronic state is induced by applying pressure, the resistivity also shows a change due to this crossover and furthermore the biquadratic IEC should show a significant variation through the intrinsic mechanism.

The large pressure effects of IEC and GMR were also observed for Fe/Cr(26) and Fe/Cr(34), showing that the crossover behavior of IEC and the enhanced GMR are characteristic for the Fe/Cr MMLs with t_{Cr} around the second peak of IEC and GMR [26]. Moreover, it is noted that the pressure effects of IEC and GMR depend not only on Cr layer thickness but also on crystallographic orientation: while GMR and IEC as a function of Cr layer thickness in epitaxial Fe/Cr MMLs are qualitatively the same as those in polycrystalline ones, their pressure dependence is different between epitaxial and polycrystalline ones [13,14].

In conclusion, we studied the pressure effect of IEC and GMR in Fe/Cr MMLs. In Fe/Cr MMLs with t_{Cr} of ~30 Å, a crossover from biquadratic to bilinear IEC was clearly observed by applying pressure, and simultaneously the GMR under high pressure was enhanced to be twice as large as that at ambient pressure. The observed results in this study provide evidence for the presence of an intrinsic mechanism of biquadratic IEC as well as the first observation of pressure-induced enhancement of GMR. The effects of pressure on GMR and IEC should be useful for further understanding of the physical origins of GMR and IEC in MMLs.

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