Role of interfacial roughness in the giant magnetoresistance in Co/Cu superlattices

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We have measured both the magnetization and the temperature dependence of the giant magnetoresistance (GMR) in Co/Cu superlattices with different interfacial roughness. The magnetization dependence of the magnetoresistance (MR) has precluded the existence of a strong spin dependence in the potential not only for the bulk but also for the interfacial scattering. The temperature dependence of the GMR is hardly influenced by the interfacial roughness, while the residual resistivity changes significantly. The residual MR ratio decreases with increasing interfacial roughness. This reveals that the spin-dependent *s-d* scattering in the bulk is crucial for the GMR in Co/Cu superlattices. The interfacial roughness mainly contributes to the residual resistivity, and the spin dependence in the scattering at interfaces is weaker than that in the bulk.

I. INTRODUCTION

A great number of attempts have been made to clarify the origin of the giant magnetoresistance (GMR) observed in various magnetic multilayers.[1–3] Most of them have focused on the spin-dependent scattering of conduction electrons. Some theoretical studies attribute the GMR to the spin-dependent potential at interfaces.[4–6] In these theories, interfacial roughness is required as the scattering center. The strong spin-dependent scattering also comes from the spin-split density-of-states (SSDOS) for majority- and minority-spin *d* bands in the magnetic layers and occurs both in the bulk and at the interfaces.[7]

On the other hand, many experimental studies deal with the correlation between the GMR and the interfacial roughness to understand the role of interfaces.[8–13] For Fe/Cr superlattices,[8,9] the enhancement of both the magnitude and temperature dependence of the GMR due to interfacial roughness has been reported, so that the origin of the GMR in Fe/Cr is attributed to the interfacial scattering.[5] However, for transition metal/Cu superlattices, no one has reported that the interfacial roughness enhances the GMR.[10–13] Nevertheless, the importance of interfacial scattering has been pointed out indirectly in studies on the layer thickness dependence of the GMR.[14,15] The mechanism of the GMR in transition metal/Cu superlattices still remains unclear.

This lack of understanding lies in the difficulty of quantitativly understanding the relationship between the interfacial structure and the transport properties since the interfacial structure is difficult to control and analyze. As reported in our previous paper,[16] we succeeded in preparing Co/Cu superlattices with well-controlled interfacial roughness. In these samples, only the atomic interfacial roughness has been modified, while the morphology and the crystallinity remain unchanged. This enables us to study the effect of interfacial roughness on the magnetization and the temperature dependence of the GMR. In the present work, our experimental data are analyzed by the SSDOS model based on the theory proposed by Xing *et al.*[7] The results suggest that the GMR in Co/Cu superlattices mainly comes from the spin-dependent *s-d* scattering in the Co layers.

II. EXPERIMENT

Since the details of the sample preparation and characterization have been reported elsewhere,[16] we only briefly describe them here. The Co/Cu superlattices were deposited on an Fe buffer layer of a thickness of 5.0 nm prepared on a surface-oxidized Si substrate in a magnetron-sputtering system. The number of Co/Cu bilayers was 16, and the thicknesses of Co and Cu in a period were kept constant at 1.0 and 2.2 nm, respectively. Interfaces between Co and Cu layers were modified by the codeposition. The nominal structure of a sample was as follows: substrate/Fe(5.0)/Cu(2.2 − t_{mix}/2)/CoCu(t_{mix})/[Co(1.0 − t_{mix})/CoCu(t_{mix})/Cu(2.2 −t_{mix})/CoCu(t_{mix})]$_{15}$/Co(1.0 − t_{mix})/Cu(2.2 − t_{mix}/2), where t_{mix} is the nominal thickness of the codeposited regions, and the values in parentheses are the thicknesses of respective layers in units of nm. The value of t_{mix} was varied between 0 and 0.25 nm. The thickness fluctuation of each layer, characterized using x-ray diffraction, was less than 0.1 nm for all samples independently of t_{mix}. On the other hand, ^{59}Co NMR revealed that Co and Cu atoms were atomically mixed at the interfaces and that the amount of interfacial mixing increased according to the value of t_{mix}.

The in-plane magnetoresistance (MR) was measured with a standard dc four-terminal geometry as a function of the temperature in the range $2 \leq T \leq 300$ K. The dimension of the samples for the measurement was 2×15 mm^2, and the current used was 1 mA. This confirms that the resolution of the measured resistance is better than 10^{-3} Ω. In order to minimize the error due to the thermoelectric power in the measurement circuit, we averaged two sequential measurements with different polarities of the current. This sequence was repeated more than 50

times keeping the temperature constant for 10 min, and the collected data were averaged. The standard deviation for the data was smaller than the order of 10^{-4} Ω. The resistance-to-resistivity conversion was performed by scaling using the resistivity measured with samples at 300 K large enough to obtain an accuracy of 10^{-2} $\mu\Omega$ cm. As a result, for one sample, the accuracy of the absolute value of resistivity was 10^{-2} $\mu\Omega$ cm, while the resolution was better than 10^{-3} $\mu\Omega$ cm. Furthermore, the scattering of the data due to the sample reproducibility, which was obtained from measurements for five series of samples, was within ± 1 $\mu\Omega$ cm.

The magnetization was also measured with a superconducting quantum interference device (SQUID) magnetometer.

III. RESULTS AND DISCUSSION

A. Relationship between GMR and magnetization

Figure 1 shows the MR curves measured at 5 K for one of the series of samples with different t_{mix}. The MR ratio is defined as $(\rho - \rho_s)/\rho_s$, where ρ is the resistivity in an arbitrary field and ρ_s is the saturation resistivity. In general, the resistivity of our samples decreases from the initial value with increasing magnetic field H and saturates at the value of ρ_s in a field larger than the saturation field H_s. After saturation, the resistivity has a peak near the coercive field. Since the value of the initial resistivity is larger than that of the peak one, the antiferromagnetic (AF) alignment of the magnetization of the Co layers is more perfect at the initial state than that in the field where the resistivity has the peak.

Figure 2 shows the relationship between ρ and $(M/M_s)^2$ during the initial magnetization process, where M is the total magnetization and M_s is the total saturation magnetization of the samples. In this figure, the magnetization of the Fe buffer layer is corrected. The linear dependence on $(M/M_s)^2$ at 5 K is clearly observed

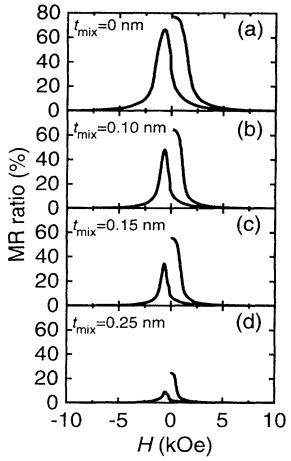

FIG. 1. The magnetoresistance curves measured at 5 K for samples with different interfacial roughness.

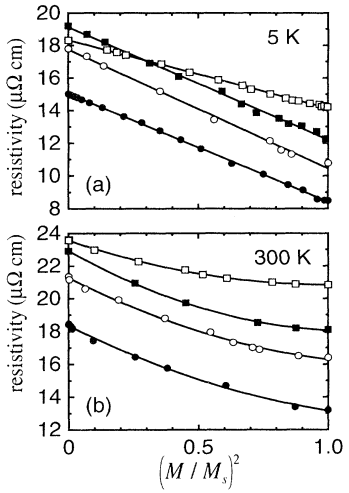

FIG. 2. The relationship between $\rho(m)$ and $(M/M_s)^2$ during the initial magnetization process measured at 5 K (a) and 300 K (b) for the samples of $t_{\mathrm{mix}} = 0$ (•), 0.10 (○), 0.15 (■), and 0.25 (□) nm. The magnetization of the Fe buffer layer is corrected for. The lines indicate a guide for the eyes.

for all the samples. On the other hand, at 300 K, ρ no longer depends linearly on $(M/M_s)^2$, and no significant enhancement of the deviation from $(M/M_s)^2$ dependence due to the interfacial roughness is observed. In order to describe the magnetization dependence of the GMR in Co/Cu, we use a two-current model with spin mixing.[17] It gives the electrical resistivity

$$\rho = \frac{\rho_\uparrow \rho_\downarrow + \rho_{\uparrow\downarrow}(\rho_\uparrow + \rho_\downarrow)}{\rho_\uparrow + \rho_\downarrow + 4\rho_{\uparrow\downarrow}}, \tag{1}$$

where $\rho_\uparrow$, $\rho_\downarrow$, and $\rho_{\uparrow\downarrow}$ are the resistivity of spin-↑ and spin-↓ channels and spin mixing, respectively. In the temperature range where $\rho_\uparrow$ and $\rho_\downarrow$ are much larger than $\rho_{\uparrow\downarrow}$, one derives the following from Eq. (1):

$$\rho = \frac{\rho_\uparrow \rho_\downarrow}{\rho_\uparrow + \rho_\downarrow} + \frac{(\rho_\uparrow - \rho_\downarrow)^2}{(\rho_\uparrow + \rho_\downarrow)^2}\rho_{\uparrow\downarrow}. \tag{2}$$

When the mean free path of conduction electrons is longer than the superlattice period, $\rho_\uparrow$ and $\rho_\downarrow$ are given as[7,18]

$$\rho_\uparrow(m) = \frac{\eta}{2}\left\{\frac{2(1-\eta)}{\eta}\rho_N + \rho_+ + \rho_-\right\} + \frac{\eta}{2}(\rho_+ - \rho_-)m, \tag{3}$$

$$\rho_\downarrow(m) = \frac{\eta}{2}\left\{\frac{2(1-\eta)}{\eta}\rho_N + \rho_+ + \rho_-\right\} - \frac{\eta}{2}(\rho_+ - \rho_-)m, \tag{4}$$

where ρ_N is the resistivity of the nonmagnetic layer, ρ_+ and ρ_- are the resistivities of the magnetic layer for majority- and minority-spin electrons, respectively, and η is the thickness fraction of the magnetic layer in one

superlattice period. The value of m is determined by the geometric configuration of the magnetization of the magnetic layers. For completely AF coupled superlattices, $m = M/M_s$.[7,19] We write here the resistivities for both spin channels as $\rho_\uparrow(m)$ and $\rho_\downarrow(m)$ to express the dependence on m explicitly. Substituting Eqs. (3) and (4) into Eq. (2), the resistivity in a field is given by

$$\rho(m) = \rho_{\mathrm{AF}} - \left\{\frac{\eta\,(\rho_+ - \rho_-)}{4}\right\}^2 \left(1 - \frac{\rho_{\uparrow\downarrow}(m)}{\rho_{\mathrm{AF}}}\right) \frac{m^2}{\rho_{\mathrm{AF}}}, \quad (5)$$

where ρ_{AF} is the resistivity when the magnetization of the magnetic layers is perfectly AF aligned and is written as

$$\rho_{\mathrm{AF}} \equiv \rho(m=0) = \frac{\eta}{4}\left\{\frac{2\,(1-\eta)}{\eta}\rho_N + \rho_+ + \rho_-\right\}. \quad (6)$$

Here, we allow the spin mixing to depend on m. As can be seen from Eq. (5), $\rho(m)$ changes linearly with m^2, when $\rho_{\uparrow\downarrow}(m)$ is negligible. If the AF alignment of the magnetization of Co is perfect, $\rho(m)$ changes linearly with $(M/M_s)^2$.[19] Therefore the magnetic configuration of our samples is very close to the perfect AF alignment at the initial state (see Fig. 2). Nevertheless, the magnitude of the saturation MR ratio decreases with increasing t_{mix} as shown in Fig. 1. This indicates that the spin dependence in the scattering of conduction electrons from interfacial roughness is weaker than that in the Co layers.

In addition to the imperfect AF alignment, the spin mixing[7] and Hasegawa's valve effect[20] cause deviation from the $(M/M_s)^2$ dependence of $\rho(m)$. Thus the existence of significant spin mixing and the valve effect are precluded for our samples at 5 K. Xing *et al.*[7] indicated that the spin mixing at low temperature comes from the spin-dependent diagonal matrix elements of the potential for the scattering of electrons with each spin direction. They showed that it made the resistivity larger than the $(M/M_s)^2$ dependence in any field of $0 < H < H_s$. The present result suggests that the spin dependence in the scattering potential for both bulk and interfacial scattering is not very strong. Therefore the GMR in Co/Cu arises from the spin-dependent s-d scattering rate due to the different density of states (DOS) for majority- and minority-spin d bands in the magnetic layer.

The deviation from $(M/M_s)^2$ dependence at 300 K is likely to be due to the spin mixing. Since $\rho(m)$ deviates downwards from the linear dependence on $(M/M_s)^2$ in a field of $0 < H < H_s$, this does not come from the spin dependence of the scattering potential. We believe that the spin mixing at 300 K is due to thermal excitation of magnons and we have to take account of not only the diagonal but also the off-diagonal matrix elements. However, the interfacial roughness does not play an important role in the spin mixing, since no significant difference in the magnitude of the deviation from the $(M/M_s)^2$ dependence between the samples with different t_{mix} is observed.

B. Temperature dependence of GMR

We examined the effect of the interfacial roughness on the temperature dependence of the GMR with our attention on the initial resistivity ρ_0 and the saturation resistivity ρ_s. As mentioned above, the samples in the initial state have almost perfect AF alignment. Furthermore, we can keep the magnetic configuration constant during the measurements of the temperature dependence of ρ_0 and ρ_s. Figure 3 shows the temperature dependence of ρ_0 and ρ_s together with that of $\Delta\rho_0 = \rho_0 - \rho_s$ for the samples of various t_{mix}. With increasing temperature, both ρ_0 and ρ_s increase due to electron-phonon, electron-magnon, or other scattering processes. The residual resistivity of ρ_s increases with increasing t_{mix} due to the increase in interfacial scattering, though the difference in the temperature coefficient is small. Since the temperature coefficient of ρ_s is larger than that of ρ_0, $\Delta\rho_0$ decreases with increasing temperature. The deviations of ρ_0 and ρ_s from their values at 2 K are shown in Fig. 4. A minimum in ρ_0 is found at around 15 K for the samples of $t_{\mathrm{mix}} \leq 0.15$ nm, while ρ_s increases monotonically with increasing temperature. As indicated in Ref. 21, the minimum in ρ_0 is the characteristic feature for AF coupled Co/Cu superlattices. However, the minimum is not found for the sample of $t_{\mathrm{mix}} = 0.25$ nm.

Since the spin mixing does not contribute to ρ_0 explicitly [see Eq. (5)], the spin mixing in ρ_s is one of the processes reducing the value of $\Delta\rho_0$ with increasing temperature. However, the minimum in ρ_0 [Fig. 4(a)] indicates that there exist other processes of reducing the GMR. The difference in the scattering mechanism would be reflected in the power law for the temperature dependence of the resistivity. Thus we focus here on the power law for $\Delta\rho_0$ and ρ_s. Before we discuss the effect of

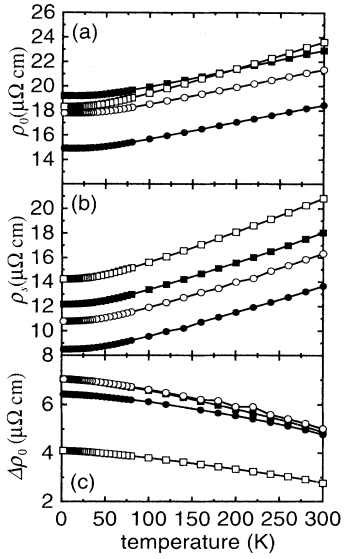

FIG. 3. Temperature dependence of (a) ρ_0, (b) ρ_s, and (c) $\Delta\rho_0$ for the samples of $t_{\mathrm{mix}} = 0$ (•), 0.10 (○), 0.15 (■), and 0.25 (□) nm. Note that the vertical scale for (c) is different from the others. The lines indicate a guide for the eyes.

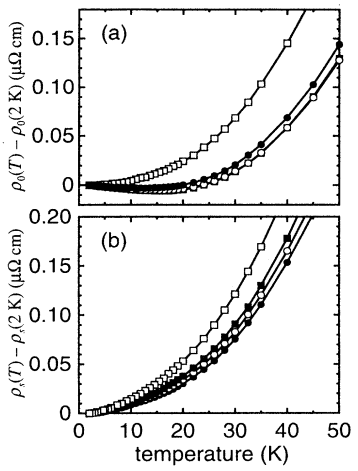

FIG. 4. Deviation of (a) ρ_0 and (b) ρ_s from their values at 2 K for the samples of $t_{\mathrm{mix}} = 0$ (•), 0.10 (○), 0.15 (■), and 0.25 (□) nm. The lines indicate a guide for the eyes.

interfacial roughness on the temperature dependence of $\Delta\rho_0$ and ρ_s, we deduce the general expression for them in any given field. Defining the MR in any given field as $\Delta\rho(m) \equiv \rho(m) - \rho(m=1) = \rho(m) - \rho_s$, Eq. (4) is written as

$$\Delta\rho(m) = \left\{\frac{\eta(\rho_+ - \rho_-)}{4}\right\}^2 \frac{1}{\rho_{\mathrm{AF}}} \times \left\{1 - m^2 - \frac{\rho_{\uparrow\downarrow}(m=1) - m^2\rho_{\uparrow\downarrow}(m)}{\rho_{\mathrm{AF}}}\right\}. \quad (7)$$

In the temperature range where $\rho_{\uparrow\downarrow} \ll \rho_{\mathrm{AF}}$,

$$\Delta\rho(m) = (1 - m^2)(\rho_{\mathrm{AF}} - \rho_s). \quad (8)$$

Equation (8) indicates that $\Delta\rho(m)$ is proportional to $\rho_{\mathrm{AF}} - \rho_s$, whenever the geometric factor m is kept constant. Note that this is correct even in the case that the AF alignment of magnetization of Co is imperfect (nonzero m). In the high temperature region, care must be taken to interpret the temperature dependence of $\Delta\rho(m)$, since the spin mixing comes into the expression for $\Delta\rho(m)$.

Figures 5 and 6 show double logarithmic plots of $\rho_s(T) - \rho_s(2\ \mathrm{K})$ and $\Delta\rho_0(2\ \mathrm{K}) - \Delta\rho_0(T)$. At temperatures lower than 100 K, ρ_s closely approximates a T^2 power law. The temperature dependence of ρ_s changes to T^n (n = 1–1.5) over 100 K. This temperature dependence cannot be attributed only to the spin mixing, since spin mixing obeys a $T^{3/2}$ power law at low temperatures and a T^2 power law at high temperatures.[17] The spin mixing found in Fig. 2(b) will be superimposed on the large temperature dependence due to other excitations. The electron-electron or electron-magnon scattering is a possible mechanism to explain the T^2 dependence, although details of the process have not been clarified. On the other hand, $\Delta\rho_0$ changes linearly with $T^{3/2}$ over the whole measurement temperature range. As can be seen from Eqs. (6) and (7), decrease in $\Delta\rho_0$ is caused by a decrease in $|\rho_+ - \rho_-|$ and increase in ρ_{AF}. Since $1/\rho_{\mathrm{AF}}$ does not simply depend on $T^{3/2}$, the term $|\rho_+ - \rho_-|$ plays an important role in the temperature dependence of $\Delta\rho_0$. Since $|\rho_+ - \rho_-|$ reflects the difference in population between the majority- and minority-spin d bands in Co, it should be closely related to the spontaneous magnetization. Saito *et al.*[21] indicated that $\Delta\rho_0$ and the spontaneous magnetization showed a similar temperature dependence. In fact, the spontaneous magnetization of our samples also shows the $T^{3/2}$ dependence. However, the relationship between them is more complicated than a simple linear relation.

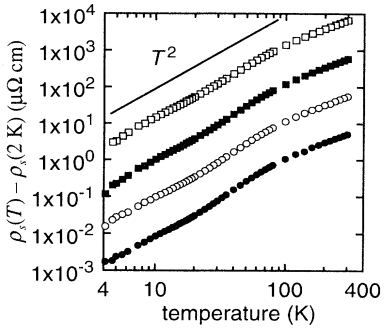

FIG. 5. Double logarithmic plot of $\rho_s(T) - \rho_s(2\ \mathrm{K})$ versus temperature for the samples of $t_{\mathrm{mix}} = 0$ (•), 0.10 (○), 0.15 (■), and 0.25 (□) nm. For the better understanding, the data for the samples of $t_{\mathrm{mix}} = 0.10$, 0.15, and 0.25 nm are multiplied by 10, 10^2, and 10^3, respectively.

As shown in Figs. 5 and 6, the power laws for $\Delta\rho_0$ and ρ_s are independent of t_{mix}, although the residual $\Delta\rho_0$ and ρ_s change significantly due to the increase in interfacial roughness (see Fig. 3). This suggests that the interfacial scattering mainly contributes to the residual resistivity, while the temperature dependence mainly comes from the bulk scattering. On the assumption that the bulk scattering is crucial for the GMR, the small value of $\Delta\rho_0$ for the sample of $t_{\mathrm{mix}} = 0.25$ nm is likely to be attributed to the decrease in the effective thickness of Co layers due to the significant interfacial mixing. As a result, the

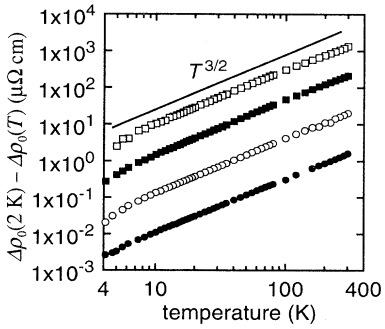

FIG. 6. Double logarithmic plot of $\Delta\rho_0(2\ \mathrm{K}) - \Delta\rho_0(T)$ versus temperature for the samples of $t_{\mathrm{mix}} = 0$ (•), 0.10 (○), 0.15 (■), and 0.25 (□) nm. For the better understanding, the data for the samples of $t_{\mathrm{mix}} = 0.10$, 0.15, and 0.25 nm are multiplied by 10, 10^2, and 10^3, respectively.

minimum in ρ_0 is only found for the samples of $t_{mix} \leq 0.15$ nm but not for the sample of $t_{mix} = 0.25$ nm.

In the SSDOS model, for GMR the scattering of s electrons to unfilled d bands with a large difference in the DOS is crucial.[7] In the Co/Cu superlattices, s electrons near the Fermi level do not experience a large potential difference at any of the interfaces,[18] since the s bands for Co and Cu are well aligned. Therefore s electrons have almost free-electron-like properties. On the other hand, the d band in Cu and the majority-spin d band in Co lie well below the Fermi level, while the minority-spin d band in Co has a large DOS at the Fermi level.[22] Thus the minority-spin d band may be localized in the Co layers. In this situation, the wave function of the electron in the minority-spin d band does not have a large amplitude near the Co/Cu interface, while that of s electrons shows no considerable change in the superlattice. Therefore the interfacial roughness will weaken the spin dependence in the scattering.

There are many studies attributing the origin of the GMR in Co/Cu superlattices to interfacial scattering.[14,15] Most of them are accomplished by measuring the dependence of the GMR on the thickness of the magnetic layers. However, the thickness dependence reveals only that the scattering centers are concentrated in a small region. We propose a hypothesis that the scattering centers causing the GMR are *in* the Co layer and concentrated *near* the interfaces. To verify the hypothesis, a detailed structural analysis to clarify the position and the kind of scattering centers will be required.

IV. CONCLUSION

We have measured the magnetization and temperature dependence of the GMR in Co/Cu superlattices with artificially modified interfaces. The magnetization dependence indicates that there is no significant spin dependence in the potentials for the bulk and interfacial scattering. The temperature dependence of the GMR is almost independent of the interfacial roughness, while the residual resistivity changes significantly. The residual MR ratio decreases with increasing interfacial roughness. A logical conclusion is, therefore, that the spin-dependent bulk s-d scattering is crucial for the GMR in Co/Cu superlattices.

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