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Ion-beam irradiation of Co/Cu nanostructures: Effects on giant magnetoresistance and magnetic properties

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We have studied the effects of ion irradiation at low doses ($<5\times10^{14}$ ions/cm^2) on the structural properties, giant magnetoresistance (GMR), and interlayer magnetic coupling in Co/Cu multilayers. X-ray analysis combined with magnetic and resistivity measurements reveal that intermixing is promoted by ion irradiation while the periodic structure and crystallographic properties of the multilayers are not significantly altered. The GMR ratio of a multilayer decreases *monotonically* with ion dose. However, thermal annealing on an irradiated multilayer results in sharp recovery of the reduced GMR, and can be associated with a backdiffusion process in metastably intermixed regions. Hence, using ion irradiation and subsequent annealing, the GMR of a *single* multilayer can be altered *reversibly* over a wide range. The variation of GMR upon irradiation (or annealing) is accompanied by significant suppression (or improvement) of the antiferromagnetic interlayer coupling. The correlation between GMR and AF coupling, as well as the role of enhanced electron scattering at interfaces during these processes are discussed. [DOI: 10.1063/1.1636525]

I. INTRODUCTION

Giant magnetoresistance (GMR) is observed in a variety of inhomogeneous materials such as metallic multilayers, spin valves, and granular structures and finds applications in magnetic recording heads and sensors.[1] Among these systems, sputtered polycrystalline Co/Cu multilayers are of particular interest due to its large GMR ratio at room temperature. In this context, the properties of the interfacial regions are of major importance. On the theoretical side, Inoue and Maekawa have reduced the interface scattering process to the classical case of magnetic impurities and attributed the large GMR observed in Co/Cu multilayers to random exchange potentials at interfaces.[1] On the experimental side, while the existence of intermixing between Co and Cu in Co/Cu multilayers prepared by sputtering techniques has been suggested by nuclear magnetic resonance (NMR) experiments,[2,3] the role it plays in GMR remains controversial. On the one hand, various research groups have revealed the interface origin of GMR in Co/Cu multilayers through planar doping experiments.[4,5] On the other hand, annealing[6] or artificial intermixing through codeposition[7] has indicated that the scattering centers causing the GMR are in the Co layers.

In order to clarify the role of interfacial mixing in GMR, techniques to modify the interfacial structure and the degree of intermixing in a systematic way are required. During deposition, interfaces in a multilayer structure can be modified by carefully controlling the deposition conditions. However, for every new interface condition, a new sample has to be made. A direct comparison between different samples may introduce further complexity, since changes in the deposition conditions not only modify the interfacial structure, but also affect the growth of subsequent layers and therefore the crystallographic properties of a film. For instance, it has been reported[7] that even a very small degree of artificial mixing by codeposition can lead to a drastic change in film texture, which is expected to influence interlayer magnetic coupling enormously.[8] Another interface modifying technique is postannealing. For a miscible system such as Fe/Cr multilayers, it has been found that annealing can promote mixing across interfaces and change GMR significantly.[9] Such behavior is not expected for Co/Cu multilayers since the elements are immiscible in equilibrium. Studies have shown that the interfacial structure and GMR of Co/Cu are scarcely affected by annealing at moderate temperatures,[10,11] while at higher temperatures, annealing leads to atomic segregation, the formation of island structures, and finally the breakdown of the multilayer.[3]

In a previous paper,[12] hereafter referred to as Paper I, it has been shown that low temperature ion-beam irradiation at low doses provides a very effective nonequilibrium technique to introduce short-range interface mixing in Co/Cu multilayers. Several advantages of this technique have been found. First, the ballistic nature of ion beam mixing makes it possible to reach a large degree of intermixing even in the immiscible system of Co/Cu. Second, the degree of intermixing can be well controlled by varying ion dose in a *single* multilayer. The fact that a single multilayer is used eliminates the effects of random sample–sample variations. Third, at such low ion doses, the crystallographic properties (texture, vertical coherence length, etc.) of a multilayer are nearly unaffected, thus any changes in the GMR of a multilayer are due to the blurring of interfaces only. In this paper, we will further correlate the GMR and the interfacial mixing induced by ion irradiation, in an effort to clarify the role of the interface in the GMR of Co/Cu multilayers.

II. EXPERIMENT

Co/Cu multilayers with various configurations were prepared by rf triode sputtering onto oxidized silicon (100) or glass (Corning 7059) substrates, at a pressure of 3 mTorr of argon gas, starting from a base pressure before sputtering below 1×10^{-7} Torr. The deposition rates, which had been calibrated with a quartz-crystal monitor, were about 2.2 Å per second for copper and 0.8 Å per second for cobalt. Total thickness of each multilayer is typically between 1000 and 2000 Å. Nominal thicknesses were confirmed by surface profilometry and low-angle x-ray reflectivity measurements. All the multilayers were deposited on a 50 Å copper buffer layer, and some of the multilayers also have a copper cap layer of 50 Å. Sample magnetization was measured using a vibrating sample magnetometer with a resolution better than 10^{-4} e.m.u. Transport properties were determined using a standard 4-point method on a high-resolution ac bridge. Low-angle x-ray reflectivity measurements were performed using a high-resolution triple-axis diffractometer with a Cu $K\alpha$ source. High-angle x-ray diffraction measurements were carried out on a conventional powder diffractometer, also with Cu $K\alpha$ radiation.

Ion irradiation experiments at normal incidence were performed in a vacuum of 10^{-7} Torr with 1 MeV Si^+ ions at currents of 0.1 μA/cm^2. To limit heating effects during irradiation, the samples were placed in thermal contact with a copper block kept at 77 K. Irradiation doses typically ranged from 10^{13} to 5×10^{14} ions/cm^2 and result in 0.01–0.5 displacements per atom (dpa), as estimated by TRIM simulations. The energy loss of the 1 MeV Si^+ in a multilayer with thickness around 1000 Å is less than 200 keV so that only a small fraction ($<0.1\%$) of the implanted ions comes to rest in the multilayers. The damage profile throughout a multilayer of about 1000 Å is expected to be relatively uniform as the energy of the MeV ions shows little variation over its thickness. Some of the irradiated multilayers were annealed up to 325 °C in vacuum of 1×10^{-5} Torr.

III. RESULTS AND DISCUSSIONS

A. Structural properties

In order to understand the effects of ion bombardment on GMR and the interlayer magnetic coupling, a systematic study of the structural evolution as a function of ion dose has been carried out using both low-angle and high-angle x-ray diffraction measurements for a series of multilayers before, during, and after annealing.

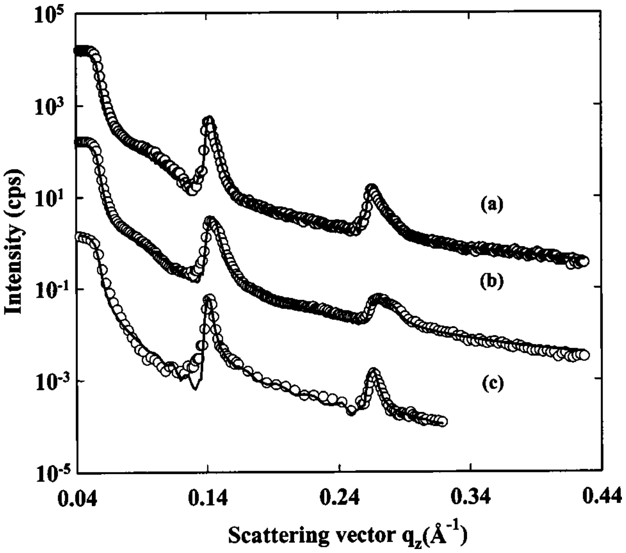

FIG. 1. Low-angle reflectivity spectra of a [Co(17 Å)/Cu(34 Å)]×30 multilayer, (a) as-deposited, (b) after irradiation with a dose of 2×10^{14} ions/cm^2, and (c) after subsequent annealing for 4 h at 250 °C.

1. Specular low-angle x-ray reflectivity

In Paper I, we were able to determine the intermixing width at various mixing stages as well as the efficiency of ion-beam mixing. The results agree well with the ballistic model which states that the mixing width Ω increases with ion dose according to the dependence $\Omega\sim\sqrt{\phi}$, where ϕ is the ion dose. Furthermore, the results also confirm that for ion doses lower than 5×10^{14} ions/cm^2, the mixing width at Co/Cu interfaces is less than 10 Å. Note that such minor intermixing at initial mixing stages has also been observed in many other multilayer systems.[13,14]

In order to examine closely the effects of ion irradiation at these low dose levels on a multilayer with a relatively small wavelength, Fig. 1 presents the small-angle x-ray spectra of a [Co(17 Å)/Cu(34 Å)]×30 multilayer, (a) as-deposited, (b) after irradiation with a dose of 2×10^{14} ions/cm^2. The as-deposited multilayer shows clear first- and second-order superlattice peaks. After irradiation, both peaks are retained, confirming that the multilayer structure is intact, but their intensities are reduced due to a blurring of the interfaces. The solid lines in Fig. 1 show the curves fitted to the spectra. Considering the low dose (and thus low dpa, displacements per atom) involved, the fits were obtained by assuming that in the simulation that only two extra slices are introduced at each interface by the radiation, corresponding to a Co-rich region ($\mathrm{Co}_{1-x}\mathrm{Cu}_x$, with $x<0.5$) and a Cu-rich region ($\mathrm{Cu}_{1-x}\mathrm{Co}_x$), respectively. Taking advantage of the fact the spectra have been taken on a single multilayer sample, other parameters, such as the layer thickness and roughness were kept fixed for simplicity. While it is found that the intermixing width and the effective

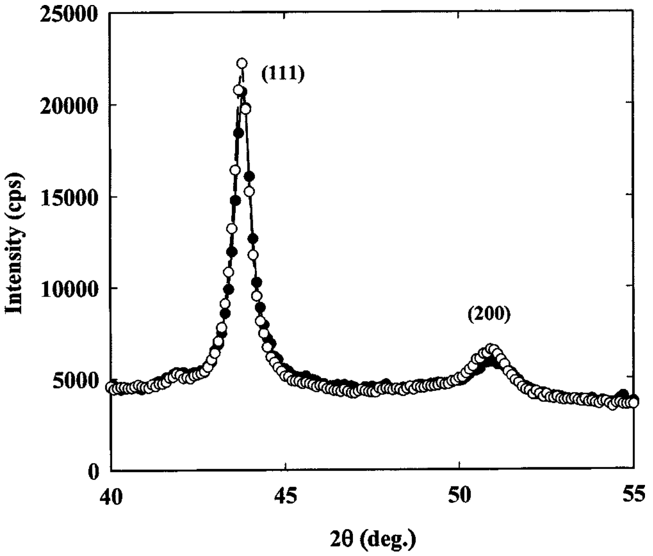

FIG. 2. High-angle x-ray diffraction spectra for a [Co(17 Å)/Cu(22 Å)]×30 multilayer before irradiation (solid circles), and after irradiation at 2 $\times 10^{14}$ ions/cm^2 (open circles).

composition x are somehow correlated, for fixed x in the range 0.1–0.3 (which is comparable to dpa), the intermixing width varies from 6 to 11 Å, values that are small compared with the wavelength of the multilayer. This analysis leads us to conclude that the principal structural effect has been to blur the Co/Cu interface over a range of ~10 Å without significantly altering the periodic structure of the multilayer.

Since Co and Cu are strongly immiscible in equilibrium, intermixing achieved by ion irradiation is metastable. Consequently, thermal treatment may restore the equilibrium state of this system through phase separation. For multilayers irradiated at very low ion dose, such a demixing process may lead to the restoration of a sharp interface structure. The spectrum shown in Fig. 1(c) shows that, after annealing of the previously irradiated sample [Fig. 1(b)] at 250 °C for 2 h, the intensities and linewidths of the superlattice peaks have almost fully recovered. As a result, this spectrum can be fitted without introducing the mixing layers provided that the interfacial roughness is raised slightly to 6.5±0.3 Å. In contrast, x-ray reflectivity spectra of virgin Co/Cu multilayers are little affected by annealing at the same temperature.[10,11]

2. High-angle x-ray diffraction

Figure 2 presents the high angle x-ray diffraction spectra for the same [Co(17 Å)/Cu(34 Å)]×30 multilayer before irradiation and after irradiation with a dose of 2 $\times 10^{14}$ ions/cm^2. Before irradiation, all multilayers mentioned in this paper are textured principally in the fcc (111) direction with a relatively weak fcc (200) component. Using the Scherrer formula, we estimate a grain size normal to the surface of about 130 Å, which is much larger than the individual layer thickness, and suggests good structural coherence across the interfaces. After irradiation, the linewidth of the (111) Bragg peak is nearly unchanged so that no sign of significant grain growth is detected. As shown in Fig. 2, the relative intensity of the (200) peak increases only slightly so that the film texture is also largely preserved. These results remain for ion doses up to 5×10^{14} ions/cm^2, and reveal that low-dose irradiation can be used to obtain very subtle interfacial modifications. Indeed, it provides a most effective method to study the role of the interfacial structure on GMR.

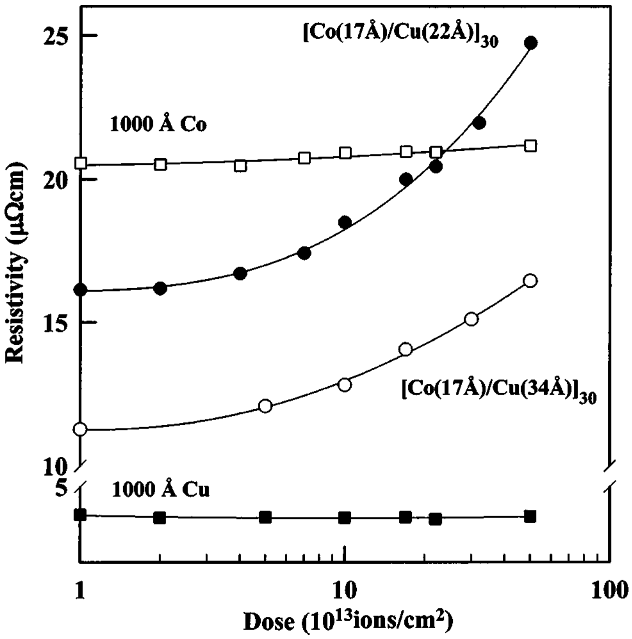

FIG. 3. Resistivities of [Co(17 Å)/Cu(22 Å)]×30 (solid circles) and a [Co(17 Å)/Cu(34 Å)]×30 (open circles) multilayers as functions of the ion dose. Also shown are corresponding data for 1000 Å films of Cu (solid squares) and Co (open squares).

B. Ion irradiation effects on electron scattering

Figure 3 presents the variations of the saturation resistivities (ρ_s) with ion dose for the [Co(17 Å)/Cu(22 Å)]×30 and the [Co(17 Å)/Cu(34 Å)]×30 multilayers; also shown are the corresponding variations of the resistivities of 1000 Å Cu and Co films. Here we concentrate on ρ_s to eliminate GMR contribution. For ion-beam doses up to 10^{13} ions/cm^2, no change in the resistivity of the multilayers is observed. At ion doses higher than 10^{13} ions/cm^2, the resistivity of the multilayers increases noticeably, well beyond that measured for pure films. We therefore suggest that the present large increase of resistivity in the Co/Cu multilayers is connected with enhanced interface electron scattering resulting from ion-beam mixing across the interfaces.

In order to convert the increase in resistivity of a multilayer to the enhancement in electron scattering near its interfaces, we adopt the semiclassical method, initially worked out by Camley and Barnas,[15] which is based on the Boltzmann equation in the relaxation time approximation. Suppose the electrical field **E** is applied in the film plane, along the x axis, and the z axis is normal to the film plane, the Boltzmann equation is written as

$$\frac{\partial g}{\partial z}+\frac{g}{\tau v_z}=\frac{eE}{mv_z}\frac{\partial f_0}{\partial v_x}, \qquad (1)$$

where $g(v,z)$ is the deviation of the electron distribution from the equilibrium Fermi–Dirac distribution $f_0(v)$. Then g

is further divided into two parts: one for electrons with positive v_z, $g_+(v,z)$, and another one for negative v_z, $g_-(v,z)$. The general solution of the Eq. (1) is

$$g_\pm(z,v)=\frac{eE\tau}{m}\frac{\partial f_0(v)}{\partial v_x}\left[1\pm F_\pm(v)\exp\left(\frac{\mp z}{\tau|v_z|}\right)\right], \tag{2}$$

where $F_\pm(v)$ are arbitrary functions of the electron velocity v, which are to be determined from the boundary conditions at an interface described as follows:

$$g_-(A)=T_{g_-}(B)+R_g(A), \tag{3}$$

$$g_+(B)=T_{g_+}(A)+R_g(B), \tag{4}$$

where *A* and *B* represent the two layers separated by the interface, *T* is the transmission coefficient at the interface and is related to the probability of electron scattering at the interface *D* by 1-*T*, and *R* represents the reflection coefficient at the interface and is negligible for Co/Cu interface. By incorporating the appropriate boundary conditions into Eq. (2), $F_\pm(v)$ and thus $g_\pm(v,z)$ are obtained. The current density in the direction of the field is then calculated by

$$J(z)=\int v_x g(v,z)d^3v. \tag{5}$$

Finally, the current in the entire structure is found by integrating $J(z)$ over the coordinate *z*, leading to the calculation of the effective resistivity of the entire structure.

The resistivity of a multilayer has thus been calculated using parameters characterizing the probabilities of electron scatterings within layers or near interfaces. These include λ_{Co} and λ_{Cu}, the electron mean free paths for bulk cobalt and copper, respectively, and the electron transmission coefficient at interfaces (*T*). Conversely, if λ_{Co} and λ_{Cu} are given, the transmission coefficient (*T*) can be estimated from the electrical resistivity. It can be pointed out that, in ferromagnets such as Co, electron transport properties should be better described by the two-channel image, corresponding to spin-up and spin-down electrons, respectively. Unfortunately, little information on the scattering asymmetry of the two current channels can be obtained solely from the resistivity data. As a result, only the average values for all the parameters over the two spin channels are considered. Bearing this simplification in mind, we are able to obtain values of λ_{Co}, λ_{Cu}, and *T* by fitting the Cu-layer thickness dependence of the multilayer resistivity for a series of $[\mathrm{Co}(17\,\text{Å})/\mathrm{Cu}(t_{\mathrm{Cu}}\,\text{Å})]\times 30$ multilayers with t_{Cu} ranging from 10 to 50 Å: the interface transmission coefficient before irradiation is found to be around 0.8, and the λ_{Co} and λ_{Cu} obtained are 30 and 150 Å, respectively, very close to those of the pure 1000 Å Co and Cu films. Next, since the resistivity of pure Co or Cu films does not change upon irradiation, the λ_{Co} and λ_{Cu} are fixed as constants and the transmission coefficients of the irradiated films are estimated from the corresponding resistivity data shown in Fig. 3. The results are presented in Fig. 4 for the two multilayers. As can be seen, the transmission coefficients are very close and follow similar variations upon irradiation: each is reduced by a factor of two as the ion dose reaches 5×10^{14} ions/cm^2. This behavior is interpreted as a large enhancement of diffuse electron scattering near the interfaces as they are blurred by ion irradiation.

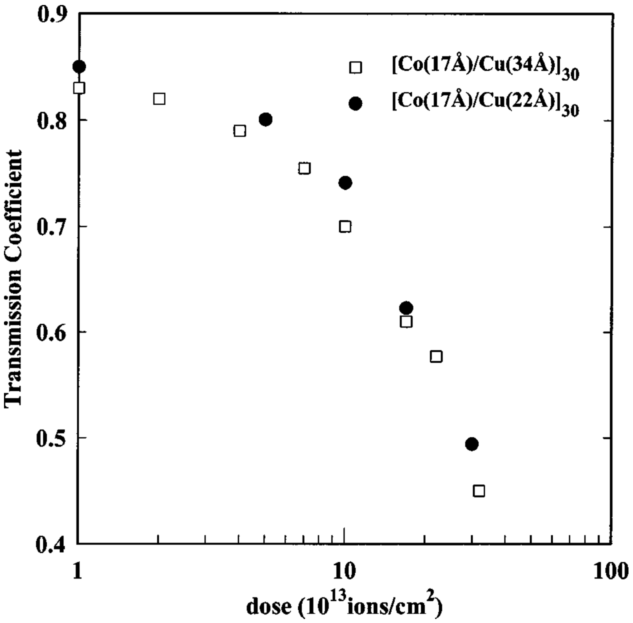

FIG. 4. Interface transmission coefficient as a function of the ion dose for [Co(17 Å)/Cu(22 Å)]×30 (solid circles) and [Co(17 Å)/Cu(34 Å)]×30 (open circles) multilayers.

The thermal demixing effect suggested by x-ray reflectivity measurements in Fig. 1 also affects electron scattering. To show this, three identical [Co(17 Å)/Cu(22 Å)]×30 multilayers were annealed. Before annealing, two of the multilayers had been subjected to irradiation doses of 1.3×10^{14} and 2.6×10^{14} ions/cm^2, respectively; the third multilayer had not been irradiated, and was included as a reference. The annealing temperature ranged from 200 to 325 °C, and the annealing periods were 4 h at temperatures below 250 °C and 2 h above this temperature. The evolutions of the resistivity, the GMR, and the magnetic properties were measured for the three samples after each annealing step.

Figure 5 shows the variation in resistivity for the multilayers. As shown, the resistivities of the irradiated multilayers decrease with temperature more rapidly than that of the nonirradiated sample. This behavior is consistent with the structural evolution observed by x-ray measurements; both phenomena can be explained by the thermal demixing driven by the equilibrium immiscibility of Cu and Co, and the resulting reformation of relatively abrupt interfaces.

C. Ion irradiation effects on GMR

Figure 6 shows the variations of the GMR with ion dose for three multilayers (a) [Co(10 Å)/Cu(10 Å)]×30, (b) [Co(17 Å)/Cu(22 Å)]×30, (c) [Co(17 Å)/Cu(34 Å)]×30. The Cu thickness of the first sample is close to the first oscillation peak of GMR, while the other two samples correspond to the second and the third peaks of GMR, respectively. For all three samples, no change in GMR is observed for ion dose below 10^{13} ions/cm^2; above this level, GMR decreases progressively with increasing dose. For the

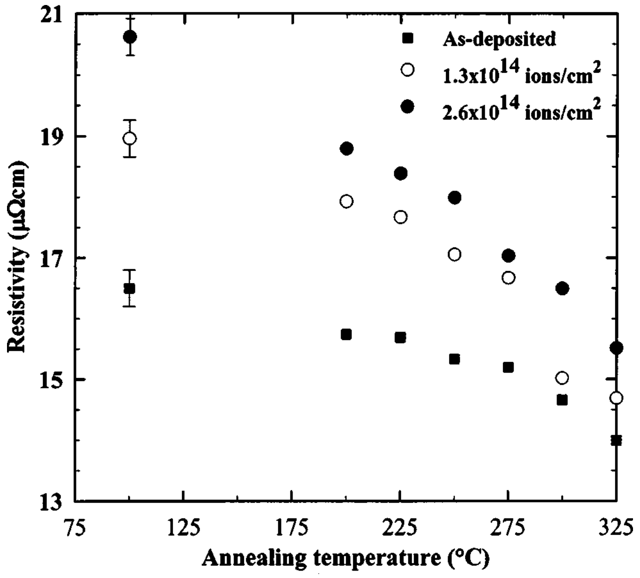

FIG. 5. Resistivity vs annealing temperature for three [Co(17 Å)/Cu(22 Å)]×30 multilayers: as-deposited (solid squares), irradiated at 1.3×10^{14} ions/cm^2 (open circles), and at 2.6×10^{14} ions/cm^2 (solid circles). The error bars indicate the uncertainty in the absolute resistivities of the samples.

multilayer near the first peak, the drop of GMR is particularly abrupt, probably due to its very thin Cu layers, whereas GMR falls more gradually for the other two multilayers. The rest of the discussion focuses on the multilayers at the second and third peaks.

The decrease of GMR upon irradiation is partially due to the increase in resistivity (ρ), as shown in Fig. 3 although most of the decrease of GMR is caused by the reduction of $\Delta\rho$ (the field-induced change in resistivity). It is interesting to note that in Fe/Cr multilayers, Kelly *et al.*[16] found that ion irradiation led to an increase in $\Delta\rho$ in spite of a significantly reduced AF coupling, and they explained this behavior in terms of the enhanced spin-dependent electron scattering at the Fe/Cr interfaces. In contrast, for the AF-coupled Co/Cu multilayers discussed here, no increase in $\Delta\rho$ upon irradiation is observed at any dose level, suggesting that the role of spin-dependent interface scattering in GMR might be quite different for Co/Cu and Fe/Cr multilayers. This point will be discussed further.

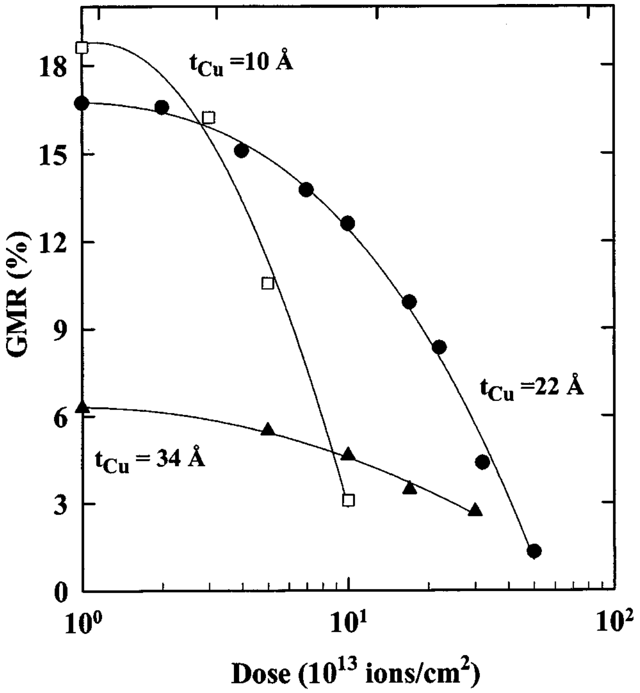

FIG. 6. GMR ratios of [Co(10 Å)/Cu(10 Å)]×30 (open squares), [Co(17 Å)/Cu(22 Å)]×30 (solid circles), and [Co(17 Å)/Cu(34 Å)]×30 (open circles) multilayer as functions of ion dose.

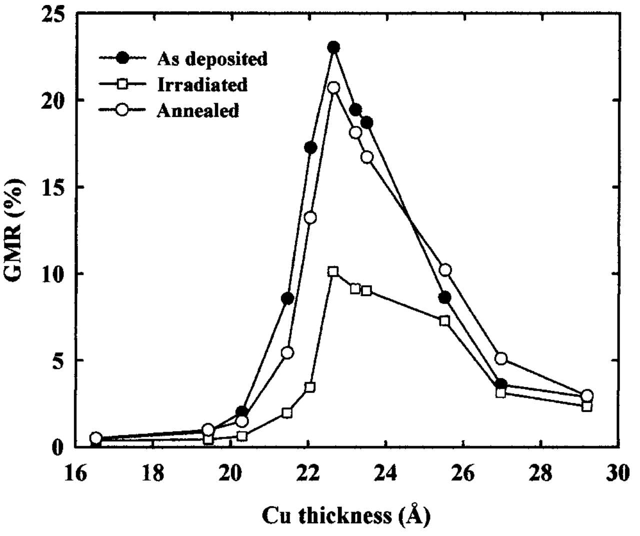

FIG. 7. GMR of a series of [Co(17 Å)/Cu(t Å)]×30 multilayers with 16 Å$<t<$30 Å before irradiation (solid circles), after irradiation with a dose of 2×10^{14} ions/cm^2 (open squares).

Figure 7 shows the GMR of a series of multilayers near the second peak before and after irradiation to a dose of 2×10^{14} ions/cm^2. (The GMR of the sample at the oscillation peak is somewhat larger than that of the sample described in Fig. 6, due to slightly different deposition conditions and a 50 Å Cu cap layer added to the latter sample.) Upon irradiation, GMR decreases for all multilayers studied. A closer look, however, reveals that the multilayers with Cu thickness slightly less than the peak position are more sensitive to the irradiation than those on the other side of the peak, leading to a plateau of GMR for a Cu-layer thickness between 23 and 27 Å. Such an asymmetry seems a bit surprising, given the fact that the half width of the original peak is only about 4 Å. Several factors might be responsible for this behavior. First, intermixing may reduce the effective thickness of nonmagnetic spacers; second, the mixing of Co into Cu may contract the Fermi surface of the Cu, which in turn, alters the oscillation period of the interlayer exchange coupling.[17]

Noticeably, the drop of GMR occurs at very low ion doses. For the [Co(10 Å)/Cu(10 Å)]×30 multilayer, the GMR is quenched at the dose of 10^{14} ions/cm^2, while for the other multilayers, the GMR has completely disappeared when the dose is increased to about 5×10^{14} ions/cm^2. These ion doses typically correspond to the initial stages of ion-beam mixing at the interfaces.[18–20]

Figure 7 also shows that GMR has almost completely recovered (even enhanced in several instances) following

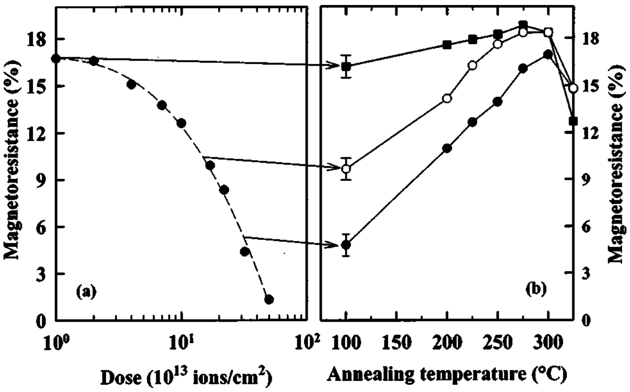

FIG. 8. (a) GMR of [Co(10 Å)/Cu(10 Å)]×30 (open squares), [Co(17 Å)/Cu(22 Å)]×30 (solid circles), and [Co(17 Å)/Cu(34 Å)]×30 multilayer (open circles) multilayers as functions of ion dose; (b) variations in GMR with annealing temperature for three [Co(17 Å)/Cu(22 Å)]×30 multilayers subjected to ion doses of 0 (solid squares), 1.3×10^{14} (open circles), and 2.6×10^{14} (solid circles) ions/cm^2, respectively.

postradiation heat treatment (first for 4 h at 240 °C, and then for 2 h at 260 °C). While irradiation suppresses and distorts the GMR peak, annealing has essentially restored the oscillation peak. Combined with the data from Fig. 1 that the multilayer structure has also been restored, it can be concluded that GMR does not originate from a granular-like contribution; rather it originates from the AF interlayer exchange coupling.

Figure 8 presents a composite image of the effects of ion bombardment and subsequent heat treatment for three [Co(17 Å)/Cu(22 Å)]×30 multilayers. Initially, the bombardment suppresses monotonically GMR but it can be entirely restored at any point in the process (up to a threshold ion dose beyond that used here). As a reference, the left-hand side of Fig. 8 plots the decrease of GMR with ion dose. No changes were observed up to 10^{13} ions/cm^2. The increase in GMR for the as-deposited multilayer results from the small decrease in the resistivity related to grain growth, as mentioned in the previous section. The more significant rise in the GMR of the irradiated multilayer is primarily due to the increase in $\Delta\rho$ as a result of the demixing process. GMR of the multilayer irradiated at higher dose increases nearly a factor of three, from about 4% to about 12% upon annealing at 225 °C. The MR of the multilayer irradiated at the lower dose has fully recovered to the as-deposited value after annealing at 250 °C. For annealing temperaturess over 300 °C, GMR falls off for all the multilayers, as the multilayer structure begins to break down. At each annealing step below this temperature, the GMR increases systematically, an effect that has been found for all the multilayers near the second peak of the GMR oscillation for ion doses below 6×10^{14} ions/cm^2. However, for the multilayers near the first peak, or those irradiated at heavy doses ($\geq10^{15}$ ions/cm^2), no clear effect is observed.

D. Ion irradiation effects on antiferromagnetic interlayer coupling

In order to understand the effects of ion irradiation on GMR, it is necessary to examine how this bombardment affects the AF interlayer coupling. Figure 9(a) shows the effect of ion radiation on the magnetization curve for the [Co(17 Å)/Cu(22 Å)]×30 multilayer before and after irradiation to a dose of 2.6×10^{14} ions/cm^2. Initially, the magnetization curve exhibits a small remanence and a slow approach to saturation, indicative of significant interlayer AF coupling. After irradiation, the magnetization curve becomes characteristic of ferromagnetic (FM) coupling, with high remanence and a rapid approach to saturation. Figure 9(b) further details the increase in the relative remanence M_r/M_s and the decrease in the normalized saturation field H_s/H_{s0} (where H_{s0} is the saturation field before irradiation) as a function of ion dose, parameters related to the degree of AF coupling between magnetic layers. Taking $(1-M_r/M_s)$ as an estimate of the volume fraction of AF coupled regions and H_s as indicative of the AF coupling strength, these results suggest that both the net AF coupled fraction and the AF coupling strength are systematically reduced by irradiation.

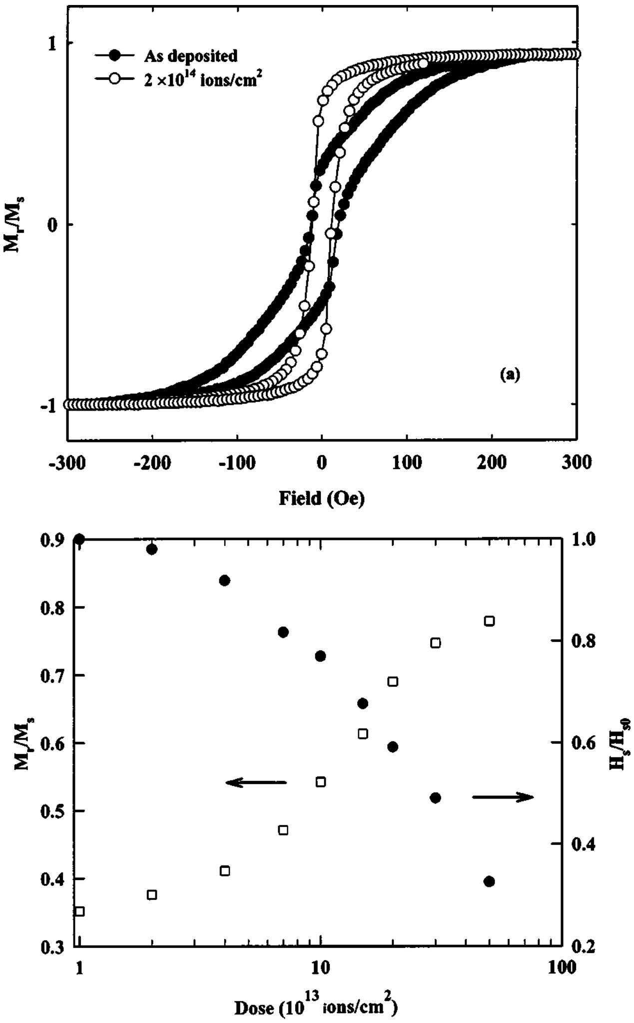

FIG. 9. (a) Magnetization curves for the [Co(17 Å)/Cu(22 Å)]×30 multilayer before irradiation (solid circles) and after irradiation with a dose of 2.6×10^{14} ions/cm^2 (open circles); (b) relative remanence magnetization (M_r/M_s), left scale, and normalized saturation magnetic field (H_s/H_{s0}), right scale, for the same multilayer as functions of the ion dose. H_{s0} is the saturation field of the as-deposited multilayer.

It has been reported that, in sputtered Co/Cu multilayers, a change in crystallographic texture has profound effects on interlayer magnetic coupling. However, at the present low ion doses, high-angle x-ray diffraction after each irradiation reveals only a subtle increase in the intensity of the (200) peak. While this change indicates an increase in the number of (200)-oriented grains, it cannot explain the dramatic suppression of the AF coupling upon irradiation: first, the change in texture is too subtle (see Fig. 2) considering the dramatic change in AF coupling; second, previous studies show that[8] in Co/Cu, the AF coupling is weakened as the grains are textured in the (111) direction and that the presence of (200)-oriented grains enhances AF coupling rather than suppresses it.

Interfacial mixing may also account for the change in interlayer magnetic coupling either indirectly or directly. Indirectly, the mixed interfacial layer effectively adds to the thickness of the nonmagnetic (Cu) spacer layer, at the expense of the magnetic (Co) layers. While this possibility cannot be totally ruled out, Fig. 7 shows that GMR decreases systematically for all the multilayers in the entire range of Cu thicknesses around the second AF-coupling peak with no shift in its position. This observation supports the conclusion that the shift in magnetic coupling period is at least not the dominant mechanism for the reduction in AF coupling with ion dose. On the other hand, AF interlayer coupling can be suppressed directly by crystalline disorder near interfaces. Through a first-principle calculation, Kudrnovsky *et al.*[21] concludes that a small amount of interfacial mixing may sharply reduce the AF coupling strength due to the strong disorder present in the Co–Cu alloy system, in particular, between Cu and down-spin Co states. This kind of disorder is expected to be generated when Co and Cu are metastably mixed by a nonequilibrium technique such as ion-beam irradiation. Furthermore, with weaker AF coupling, it is more difficult to nucleate a perfect AF alignment throughout a superlattice in zero magnetic field. Consequently, a decrease in AF coupling strength reduces the volume fraction of the AF coupled regions, as observed in Fig. 9(b).

We underscore the fact that changes in GMR are closely accompanied by similar variations in the AF coupling between Co layers. The features of AF interlayer coupling suppressed by ion irradiation are recovered by the annealing. Specifically, the magnetization curve after annealing exhibits low remanence and high saturation field similar to those of the magnetization before irradiation. Figure 10 presents details of the increase with annealing temperature of $(1-M_r/M_s)$:$(1-M_r/M_s)$ increases systematically with annealing temperature up to about 300 °C. This behavior parallels that of the magnetoresistance given in Fig. 8. For the multilayer irradiated at the lower dose, the remanence rises close to that of the as-deposited multilayer upon annealing at 250 °C. For the multilayer irradiated at the higher dose, though not fully recovered, $(1-M_r/M_s)$ significantly increases upon annealing. Such behavior reinforces the earlier conclusion that the AF exchange coupling between layers scales with the sharpness and the degree of atomic order of the interfacial regions. Finally, annealing at temperatures above 300 °C leads to a breakdown of the multilayer structure and also of GMR and the AF coupling.

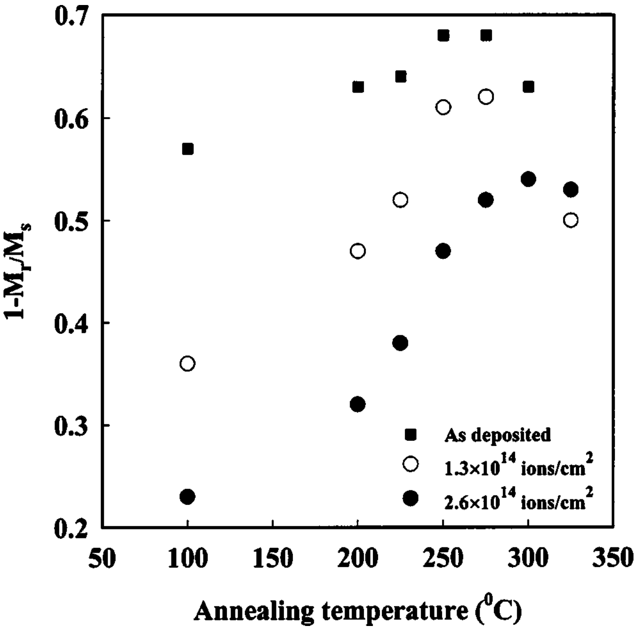

FIG. 10. Variations in $(1-M_r/M_s)$ with annealing temperature for three [Co(17 Å)/Cu(22 Å)]×30 multilayers subjected to ion doses of 0 (solid squares), 1.3×1014 (open circles), and 2.6×10^{14} (solid circles) ions/cm^2, respectively.

IV. FURTHER DISCUSSION

For the Co/Cu samples discussed here, it is interesting to note that the decrease of MR occurs at similar ion doses as the suppression of AF magnetic coupling, as is evident from Figs. 9(b) and 6, and suggests that the current reductions in GMR and the AF coupling upon irradiation are directly connected. Recently, a linear relationship between the GMR ratio and the AF coupling fraction has been deduced by Takahashi and Inomata,[22] in the form:

$$\mathrm{MR}=\mathrm{MR}_0[1-(p+\alpha)], \tag{6}$$

where p is the volume fraction of the AF coupled regions, α is a correction due to the local ferromagnetic coupling or magnetic anisotropy, and MR$_0$ is the MR of the multilayer should the AF coupling be perfect. Experimentally, this relationship was confirmed in a series of Co/Cu multilayers deposited under various conditions.[22] To our knowledge, no results have ever been reported regarding this relationship in a *single* Co/Cu multilayer with systematically modified interfaces, due to the difficulty of modifying the AF coupling fraction over a wide range in a single sample. The present results [specifically Fig. 9(b)], demonstrate that $(1-M_r/M_s)$ of a multilayer can be altered extensively between about 0.3 and 0.7 using ion irradiation. Figure 11 combines the data in Figs. 6 and 9(b), with the GMR of the [Co(17 Å)/Cu(22 Å)]×30 multilayer irradiated at various doses. Clearly, at lower doses, the GMR increases linearly with $(1-M_r/M_s)$. Fitting to Eq. (6) yields $\alpha\approx0$ (as expected) and MR$_0\approx26\%$. It can be concluded that the decrease of GMR upon irradiation at low doses ($\leqslant 2\times10^{14}$ ions/cm^2) is directly controlled by the suppression of the AF coupling. At higher doses, the GMR decreases more rapidly than the reduction in AF coupling fraction. One pos-

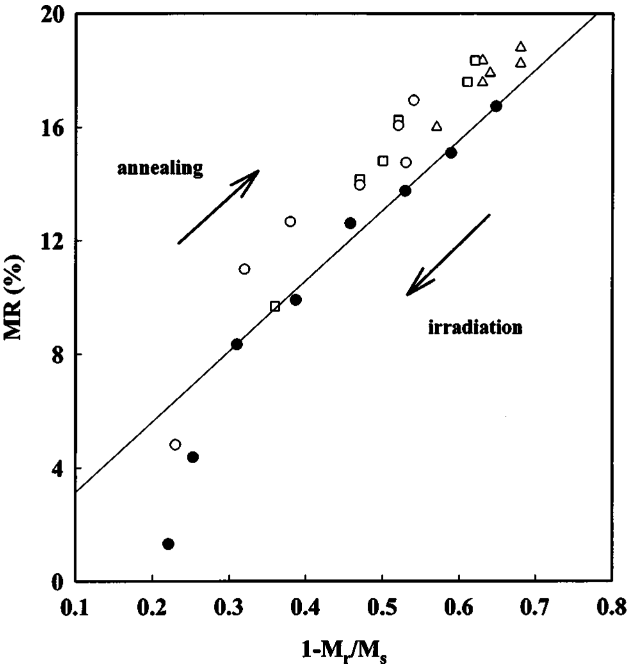

FIG. 11. GMR vs $(1-M_r/M_s)$ for the [Co(17 Å)/Cu(22 Å)]×30 multilayer (solid circles) of Figs. 8(a) and 9(b), and for the three multilayers of Figs. 8(b) and 10 which were subjected to ion doses of 0 (open triangles), 1.3 $\times 10^{14}$ (open squares), and 2.6×10^{14} (open circles) ions/cm^2, respectively.

sible explanation is that, as the interfaces are further mixed, strong electron scattering in the disordered intermixed regions may effectively confine the carriers of both spin channels within individual layers, a scenario which is consistent with the rapid decrease in the electron transmission coefficient at interfaces as seen in Fig. 4.

Figure 11 also correlates the increase of GMR and $(1-M_r/M_s)$ upon annealing by combining data from Figs. 8 and 10. For the nonirradiated multilayer, annealing below 300 °C alters the remanence only slightly, thus the corresponding data are only varied over a small range. For the irradiated multilayers, in contrast, the effects of annealing on GMR and AF coupling fraction are *amplified*, and both GMR and AF coupling are tuned over much wider ranges. As can be seen, the experimental points fall close to a single line, suggesting that the recovery of GMR upon annealing in the irradiated multilayers is the direct consequence of the improvement of AF coupling. For any fixed value of $(1-M_r/M_s)$, the GMR of the annealed multilayer is systematically larger, probably due to grain growth upon annealing.

No increase in either $\Delta\rho$ or GMR is ever observed for any of our Co/Cu multilayer at any ion dose. Such a result is inconceivable if the GMR in Co/Cu multilayers is dominated by *spin-dependent* impurity scattering at interfaces. Thus, we find no evidence that such scattering dominates GMR in Co/Cu multilayers. On this point, our results agree with those obtained from artificial interface mixing through codeposition.[7]

The possibility of interface origin of GMR in Co/Cu multilayers has been suggested by early experiments of planar doping.[4] While the major effect of ion irradiation is to introduce intermixing, the origin of GMR may be associated not only with spin-dependent impurity scattering due to intermixing, but also with other mechanisms. For example, the importance of "geometrical" interface roughness in GMR has been pointed out by Hood *et al.*[23] Recently, Stiles has noted[24] that spin-dependent reflection from interfaces resulting from the band structure of bulk Co may also contribute to a large GMR through channeling effects, even if there is no spin-dependent defect scattering. The existence of such contributions particularly cannot be excluded from our results. Nevertheless, our results suggest that if the GMR in Co/Cu multilayers is interface related, it is more likely related to some mechanisms other than the intermixing based impurity scattering.

V. CONCLUSIONS

Combining low-dose ion bombardment and subsequent thermal annealing provides an effective *ex situ* technique to modify the Co/Cu interface structure *controllably* and to tune GMR and the AF interlayer coupling *reversibly* for a *single* Co/Cu multilayer over a wide range. Structural analysis, resistivity, magnetoresistance, and magnetization measurements all point to the conclusion that low-dose ion irradiation is capable of generating nonequilibrium intermixing at the Co/Cu interfaces while subsequent annealing on irradiated multilayers provokes backdiffusion and restores abrupt interfaces. It is also concluded that GMR and AF interlayer coupling are directly connected and are strongly dependent on the interface sharpness and structure. A sharp and atomically ordered interface improves AF coupling and thus leads to larger GMR, while interfacial mixing suppresses AF coupling and reduces GMR. A linear relationship between GMR and the volume fraction of AF coupled regions is observed. The absence of enhanced GMR following ion-induced intermixing leads to the conclusion that spin-dependent electron scattering does not play a decisive role in the variation of GMR.

Noticeably, even at ion fluences where interface mixing was barely detectable by x-ray reflectivity, large changes in the magnetoresistivity could be observed. Hence, magnetoresistance measurements can be used as a very sensitive probe to detect subtle interfacial mixing induced by ion-beam bombardment, or interfacial demixing provoked by thermal annealing. Such a probe is particularly valuable for the Co/Cu system in which the atomic contrast between the elements is small, thus making it difficult to detect the initial stages of ion-beam modification by alternative techniques.

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