

## Extraordinary interlayer coupling in Co/Cu(Mn) multilayers

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We have investigated the effect of Mn doping into Cu layers on the interlayer coupling in Co/Cu multilayers and found a partial change of the interlayer coupling from ferromagnetic to antiferromagnetic or vice versa with decreasing temperature. The change of the interlayer coupling occurs around a critical temperature which depends on both the Cu(Mn) layer thickness and Mn concentration. As an origin of the change of the interlayer coupling, a cooperative interaction among the Co layers and the Mn ions is discussed in correlation with a spin-glass-like state. [S0163-1829(99)06405-X]

### I. INTRODUCTION

There has been great interest in studying transport properties in magnetic multilayers since the discovery of the giant magnetoresistance (GMR) effect in the Fe/Cr system.<sup>1</sup> The oscillation of interlayer coupling strength as a function of nonmagnetic layer thickness has further promoted a widespread research interest in those systems.<sup>2-5</sup> Several theoretical models have been proposed to explain the mechanism of the oscillatory interlayer coupling,<sup>6-9</sup> however, the origin has not yet been fully understood. The theoretical models explaining the interlayer coupling can be roughly classified into two; a model based on Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and one based on the size quantization of magnetic carrier dispersion relation. In the RKKY model, we expect that the interlayer coupling is modified by magnetic impurities doped in nonmagnetic spacer layers.

In this paper, we report systematic measurements of magnetoresistance (MR) and magnetization in Co/Cu(Mn) multilayers where 4 and 13 at. % Mn is doped in the Cu spacer layers in order to investigate the effect of magnetic impurities on the interlayer coupling. The Cu(Mn) alloy is well known as a typical spin-glass material where Mn spins freeze randomly below a certain temperatures due to the RKKY interaction among them.<sup>10</sup> In such a system, we can expect a competition or cooperation between the interlayer coupling and the spin glass.

There have been several works which report the effect of the impurity doping in the spacer layers of Co/Cu multilayers on the MR magnitude<sup>11</sup> and on the oscillation period of the interlayer coupling due to the change of the nesting vector of the Fermi surface.<sup>11-14</sup> Very recently, Fullerton *et al.* have reported an enhanced biquadratic coupling between Fe layers in Fe/Cr(6 at. % Fe) superlattices with large Cr(Fe) layer thickness.<sup>15</sup> They inferred that the enhancement of biquadratic coupling is due to a local coupling between Fe layers and Cr(Fe) layers in which inhomogeneous antiferromagnetic ordering is expected. The present work modifies intentionally the interlayer coupling by magnetic interaction due

to Mn impurities in the spacer layers, as in our preliminary reports.<sup>16,17</sup>

### II. EXPERIMENT

All samples were prepared in an ultra-high-vacuum (UHV) compatible sputtering system at Michigan State University. The samples were deposited on chemically etched Si(100) substrates on which 50 Å of Fe buffer layer was first deposited. The substrate temperature during the deposition was between  $-15$  and  $-30$  °C. As sputtering targets, Cu(4 and 13 at. % Mn) alloys and Co were used. While the Co layer thickness was fixed at 15 Å, the Cu(Mn) layer thickness ( $d_{\text{Cu(Mn)}}$ ) were selected to span from the first to the second peaks of the MR oscillation reported for the undoped Co/Cu multilayers.<sup>5</sup> The number of bilayers was varied so that the total sample thickness becomes about 2400 Å. As a reference system to separate the effect due to the reduced mean free path of the conduction electron, we prepared 4 at. % Ge-doped reference samples in the same manner as Co/Cu(Mn) multilayers. Note that the resistivity for Ge-doped samples is larger than that for Mn-doped ones with the same spacer layer thickness though the residual resistivity per impurity concentration for Cu(Mn) alloys is larger than that for Cu(Ge) alloys,<sup>18</sup> which is possibly due to some deviation in the Ge and Mn concentrations.

The electrical resistivity was measured between 1.5 and 300 K by the standard dc four-probe method using a computer-controlled current source and KEITHLEY-181 nanovoltmeters. The magnetic field was applied in sample plane across the current direction, up to 15 kOe using a conventional iron core electromagnet and up to 50 kOe using a superconducting magnet. Magnetization measurement was performed using a Quantum Design Superconducting quantum interference device magnetometer up to 55 kOe.

### III. RESULTS

In this paper, the MR ratio is defined as  $(\rho_{\text{max}} - \rho_s)/\rho_s$  using the maximum resistivity  $\rho_{\text{max}}$  and the resistivity at

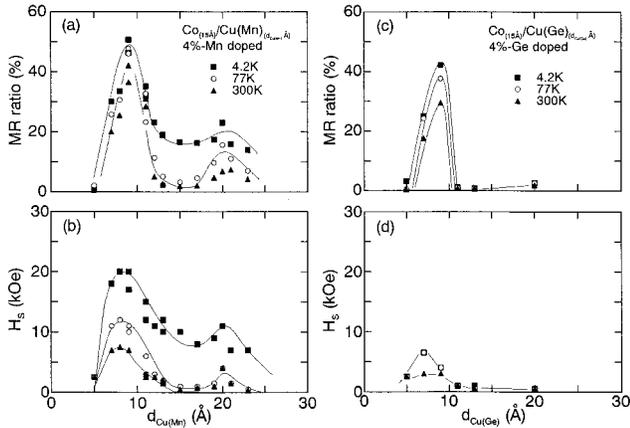


FIG. 1. The spacer layer thickness dependence of the magnetoresistance (MR) ratio and  $H_s$  determined from the MR measurements for Co/Cu(Mn) and Co/Cu(Ge) multilayers. The solid lines are guide to eyes.

maximum field  $\rho_s$ . The  $\rho_{\max}$  is replaced by the field-dependent resistivity  $\rho(H)$  when we discuss the field dependence. The saturation field ( $H_s$ ) for MR is defined as the field where the resistivity change becomes 99% of the total resistivity change  $\Delta\rho = (\rho_{\max} - \rho_s)$ . The  $H_s$  for magnetization ( $M$ ) is also defined as the field where  $M/M_s$  ( $M_s$  is the saturation magnetization) becomes 99%.

### A. Temperature dependence of MR oscillations in Co/Cu(Mn) multilayers

First, we show the dependence of MR ratio and  $H_s$  on  $d_{\text{Cu(Mn)}}$  in Fig. 1 for Co/Cu(4 at. % Mn) multilayers, along with those for Co/Cu(4 at. % Ge) multilayers. At 300 and 77 K, the MR oscillation is not much different from those reported for the undoped Co/Cu multilayers.<sup>4,5</sup> At 4.2 K, however, two striking characteristics become evident; (i) the MR ratio increases drastically around  $d_{\text{Cu(Mn)}} \sim 13 \text{ \AA}$  (oscillation minimum), and (ii) the  $H_s$  increases apparently for all  $d_{\text{Cu(Mn)}}$ . Similar characteristics were observed also for 13 at. % Mn-doped samples (not shown). The characteristic (i) suggests the growth of antiferromagnetic (AF) coupling component at low temperatures while the coupling is ferromagnetic at 300 and 77 K. The characteristic (ii) indicates that the strength of the interlayer coupling between adjacent Co layers is enhanced for all  $d_{\text{Cu(Mn)}}$ . For Ge-doped samples, only a slight enhancement in the MR ratio and the  $H_s$  has been observed at low temperatures, as is usually reported for undoped systems [see Figs. 1(c) and 1(d)]. Judging from these facts, the remarkable changes of interlayer coupling are undoubtedly due to the Mn doping.

### B. Field dependence of the MR and $M$ at fixed temperatures

Figure 2(a) shows the field dependence of transverse MR for a  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(4 \text{ at. \% Mn})_{(9 \text{ \AA})}$  multilayer. The spacer layer thickness 9 Å corresponds to the first peak of the MR oscillation as a function of Cu layer thickness.<sup>4,5</sup> The  $H_s$  increases remarkably with decreasing temperature, though it is not much different from that reported for Co/Cu multilayers<sup>4,5</sup> at 300 K. The enhancement of  $H_s$ , which is also reflected in

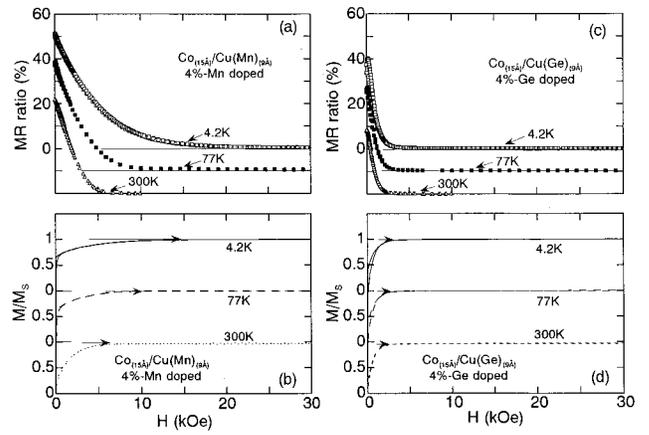


FIG. 2. The field dependence of the MR ratio and magnetization ( $M$ ) for  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(\text{Mn})_{(9 \text{ \AA})}$  and  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(\text{Ge})_{(9 \text{ \AA})}$  multilayers.

the  $M(H)$  as shown in Fig. 2(b), is far beyond what usually observed in ordinary ferromagnetic materials at low temperatures. In fact, for Ge-doped sample, such an enhancement of  $H_s$  with decreasing temperature was not observed both in the MR and the  $M$  [see Figs. 2(c) and 2(d), respectively].

The effect of Mn doping at low temperatures appears more drastically for  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(\text{Mn})_{(13 \text{ \AA})}$ . The spacer-layer thickness 13 Å corresponds to the first minimum of the MR oscillation for undoped Co/Cu multilayers; i.e., Co layers are ferromagnetically (F) coupled.<sup>4,5</sup> As expected, the MR ratio at 77 and 300 K is only a few percent as shown in Fig. 3(a). The ferromagnetic coupling is confirmed also by the  $M(H)$  [see Fig. 3(b)]. At 4.2 K, however, a spectacular enhancement of both the MR ratio and the  $H_s$  was observed, suggesting a growth of the AF-coupling component. That is also evidenced clearly in the  $M(H)$  at 4.2 K. Taking into account the fact that no anomaly in the MR and the  $H_s$  has been found for the Ge-doped samples as shown in Figs. 3(c) and 3(d), Mn impurities play an essential role in the growth of AF-coupling component at low temperatures.

### C. Temperature dependence of the interlayer coupling

In order to examine the growth of AF component at low temperatures, we have measured the temperature dependence of the resistivity ( $\rho$ ) for  $d_{\text{Cu(Mn)}} \sim 13 \text{ \AA}$ . As shown in Fig. 4,  $\rho$

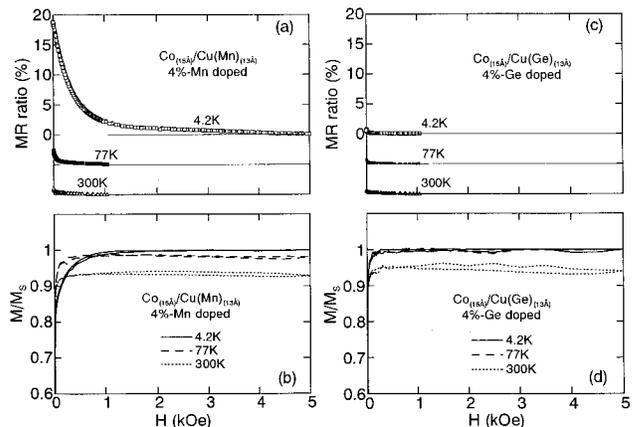


FIG. 3. The field dependence of the MR ratio and  $M$  for  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(\text{Mn})_{(13 \text{ \AA})}$  and  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(\text{Ge})_{(13 \text{ \AA})}$  multilayers.

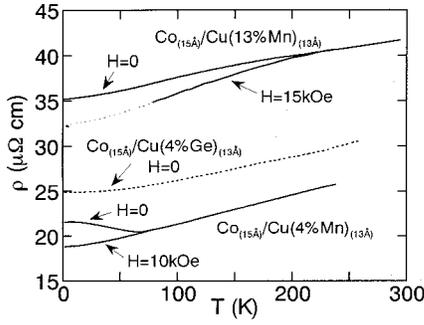


FIG. 4. The temperature dependence of resistivity ( $\rho$ ) for  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(\text{Mn})_{(13 \text{ \AA})}$  and  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(\text{Ge})_{(13 \text{ \AA})}$  multilayers. For the 13 at. % Mn-doped sample,  $\rho$  at 15 kOe was measured above 77 K except a fixed point at 4.2 K.

at zero field for the 4 at. % Mn-doped sample shows a minimum near 70 K. The curve at 10 kOe starts to deviate from that at zero field near almost the same temperature. The resistance minimum is not due to ordinary Kondo effect since the temperature at the resistance minimum is much higher than Kondo temperature expected for dilute Cu(Mn) alloy.<sup>19</sup> It should be also noted that Cu(4 at. % Mn) bulk alloy is reported to exhibit a spin-glass transition.<sup>10</sup> Most importantly, the minimum was observed only for the samples with the  $d_{\text{Cu}(\text{Mn})} \sim 13 \text{ \AA}$  which is near the first minimum of the MR oscillation for the undoped Co/Cu system.<sup>16,17</sup> The smallness of the  $\rho$  value excludes the possibility of the weak-localization effect. It is naturally inferred that the increase of  $\rho$  below about 70 K is due to the enhancement of the conduction-electron scattering responsible for the GMR resulting from the growth of AF-coupling component. For the 13 at. % Mn-doped sample, the difference between  $\rho$  at  $H=0$  and 15 kOe starts near 200 K, below which the AF component might grow (see Fig. 4).

Taking into account the fact that the interlayer coupling changes from ferromagnetic to antiferromagnetic in the originally F-coupled samples, the reverse is expected in originally AF-coupled samples. Such an expectation was confirmed by the temperature dependence of remnant magnetization normalized by the saturation magnetization ( $M_0/M_s$ ) in Fig. 5(a). The  $M_0/M_s$  shows an enormous enhancement with decreasing temperature for the Mn-doped samples near the first peak of MR oscillation ( $d_{\text{Cu}(\text{Mn})} = 7$  and  $9 \text{ \AA}$ ) in comparison with the slight increase for the Ge-doped sample. In contrast to the samples with  $d_{\text{Cu}(\text{Mn})} \sim 13 \text{ \AA}$ ,  $M_0/M_s$  exhibits a slight but apparent decrease with decreasing temperature, reflecting a decrease of F coupling at low temperatures. The growth of F coupling in originally AF-coupled samples is also reflected in the temperature dependence of  $\Delta\rho$ . For  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(\text{Ge})_{(9 \text{ \AA})}$ ,  $\Delta\rho$  increases slightly with decreasing temperature as reported for undoped Co/Cu multilayers,<sup>4</sup> while for  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(\text{Mn})_{(9 \text{ \AA})}$  it definitely decreases with decreasing temperature as shown in Fig. 5(b). These results could be understood if the interlayer coupling in some selected area changes from AF to F at low temperatures.

All these facts demonstrate the temperature-dependent switching of interlayer coupling due to Mn doping; i.e., the interlayer coupling partially changes from antiferromagnetic

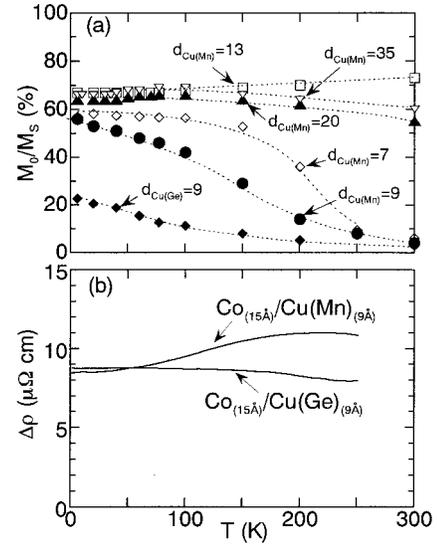


FIG. 5. (a) The temperature dependence of remnant magnetization normalized to saturation value ( $M_0/M_s$ ) for 4 at. % Mn-doped  $\text{Co}/\text{Cu}(\text{Mn})$  and  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(\text{Ge})_{(9 \text{ \AA})}$  multilayers, (b) The temperature dependence of  $\Delta\rho (= \rho_{\text{max}} - \rho_s)$  for  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(\text{Mn})_{(9 \text{ \AA})}$  and  $\text{Co}_{(15 \text{ \AA})}/\text{Cu}(\text{Ge})_{(9 \text{ \AA})}$  multilayers.

to ferromagnetic in AF-coupled samples and vice versa in F-coupled samples with decreasing temperature.

#### D. Switching temperature of the interlayer coupling

To clarify the characteristics of temperature dependence of the interlayer coupling,  $H_s$  estimated from  $M(H)$  is plotted as a function of temperature in Fig. 6. For  $\text{Co}/\text{Cu}(\text{Mn})$  multilayers, the  $H_s$  increases exponentially with decreasing temperature. The slope of  $\ln(H_s)$  versus  $T$  changes abruptly

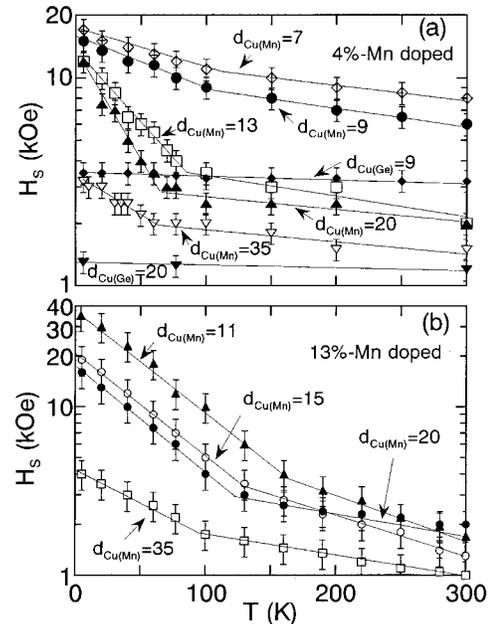


FIG. 6. (a) The temperature dependence of  $H_s$  determined from  $M(H)$  curves for 4 at. % Mn-doped  $\text{Co}/\text{Cu}(\text{Mn})$  multilayers along with that for  $\text{Co}/\text{Cu}(\text{Ge})$ , and (b) for 13 at. % Mn-doped  $\text{Co}/\text{Cu}(\text{Mn})$  multilayers.

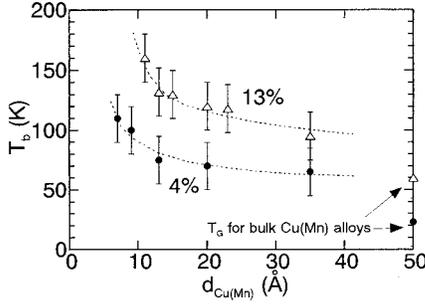


FIG. 7. The dependence of critical temperature ( $T_b$ ) on  $d_{\text{Cu(Mn)}}$  for 4 at. % and 13 at. % Mn-doped Co/Cu(Mn) multilayers along with spin-glass temperature ( $T_G$ ) of bulk Cu(Mn) alloys (Ref. 10).

around a critical temperatures ( $T_b$ ). For the 4 at. % Mn-doped sample with  $d_{\text{Cu(Mn)}} \sim 13 \text{ \AA}$ ,  $T_b$  is close to the temperature at the resistance minimum, reflecting the abrupt growth of the AF component below  $T_b$ . The  $H_s$  below  $T_b$  for 13 at. % Mn-doped samples is much larger than that for the 4 at. % Mn-doped one, suggesting that the larger Mn concentration yields the larger  $H_s$ .

The  $d_{\text{Cu(Mn)}}$  dependence of  $T_b$  is shown in Fig. 7 for both 4 and 13 at. % Mn-doped samples. For all the  $d_{\text{Cu(Mn)}}$ ,  $T_b$  of 13 at. % Mn-doped samples is larger than that of the 4 at. % Mn-doped ones. For small  $d_{\text{Cu(Mn)}}$  ( $< \sim 15 \text{ \AA}$ ),  $T_b$  increases sharply with decreasing  $d_{\text{Cu(Mn)}}$ , however, it depends only weakly on  $d_{\text{Cu(Mn)}}$  for large  $d_{\text{Cu(Mn)}}$ . For  $d_{\text{Cu(Mn)}} = 35 \text{ \AA}$ ,  $T_b$  is more than twice higher than the spin-glass temperature ( $T_G$ ) for bulk Cu(Mn) alloys ( $\sim 23 \text{ K}$  for 4 at. % and  $\sim 60 \text{ K}$  for 13 at. %<sup>10</sup>).

#### IV. DISCUSSION

Two possibilities are discussed in this section as origins of the partial sign change of the interlayer coupling below  $T_b$ . The first is the canted state of the adjacent Co layers, where F and AF components coexist apparently, due to a biquadratic coupling interaction. In order to explain the experimental reports on biquadratic coupling,<sup>20,21</sup> Slonczewski has proposed a theoretical model where localized-electron states with an unpaired spin (loose spin) located within or at the interfaces of the nonmagnetic metallic spacer layers enhance biquadratic interaction.<sup>22</sup> The Mn-doped system in the present experiment is one of the model systems to examine the loose spin theory. Rodmacq and co-workers have applied the scaling procedure between  $\rho$  and  $M^2$  in order to confirm the existence of biquadratic coupling in NiFe/Ag multilayers.<sup>23,24</sup> First, they assumed that the resistivity changes proportionally to  $(1 - \cos^2 \theta)/(1 - \cos^2 \theta_0)$ , where the magnetizations in magnetic layers alternately make an angle  $+\theta$  and  $-\theta$  with respect to the external field and the angle between the magnetizations at zero field is  $2\theta_0$ . For the canted state where the magnetizations are at an angle  $2\theta_0$  in zero field, one will have  $\cos \theta = M/M_s$  and  $\cos \theta_0 = M_0/M_s$ , therefore the resistivity changes proportionally to  $[M_s^2 - M^2(H)]/[M_s^2 - M_0^2]$ .<sup>23,24</sup> Applying the same procedure, we confirmed that the interlayer coupling in the present Co/Cu(Mn) system at lower temperatures cannot be explained by the canted state of the adjacent Co layers as shown in Fig. 8(a).<sup>25</sup> An alternative explanation is a change of the sign of

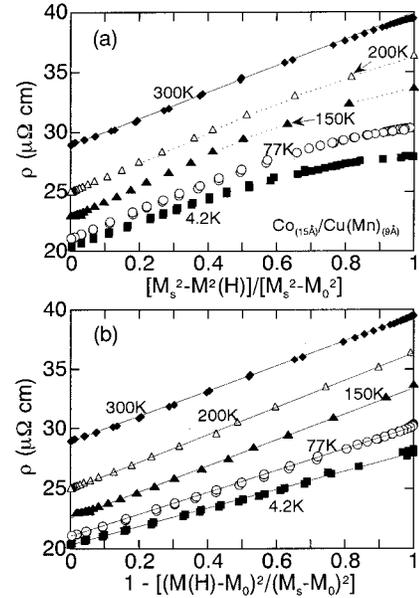


FIG. 8. (a)  $\rho$  versus  $[M_s^2 - M^2(H)]/[M_s^2 - M_0^2]$  and (b)  $\rho$  versus  $1 - [(M(H) - M_0)/(M_s - M_0)]^2$  plots for the  $\text{Co}_{(15 \text{ \AA})}/\text{Cu(Mn)}_{(9 \text{ \AA})}$  multilayer.

the interlayer coupling in some selected areas, leading to the coexistence of AF and F components. The effect of distributed Mn ions on the interlayer coupling cannot be canceled out by averaging over the sample since the samples in the present experiment are polycrystalline and composed of grains or columnar structures with finite sizes. The finite effect, which is left after averaging over within each grain below  $T_b$ , can alter the interlayer coupling in some grains from F coupling to AF coupling or vice versa. That explains the experimental observation; i.e., coexistence of AF and F components in a single sample below  $T_b$ . In the scaling procedure by Rodmacq *et al.*, the resistivity in the samples where both AF- and F-coupled areas coexist changes proportionally to  $1 - [(M(H) - M_0)/(M_s - M_0)]^2$ .<sup>23,24</sup> In fact, the scaling procedure between  $\rho$  and  $M^2$  supports this mechanism [see Fig. 8(b)].<sup>25</sup> Note that for the Ge-doped sample with  $d_{\text{Cu(Ge)}} = 9 \text{ \AA}$ , the scaling is roughly obeyed without taking into account the contribution of  $M_0$  (not shown), suggesting the ordinary AF coupling.<sup>25</sup>

Next we discuss the mechanism of the change of the interlayer coupling in some grains below  $T_b$ . As one of the possible scenarios, we propose a simple model based on the cooperation between interlayer exchange interaction and RKKY interaction due to the doped Mn spins. At higher temperatures above  $T_b$ , Mn spins are in the paramagnetic state and can freely rotate [see Fig. 9(a)]. In such a condition, the time average of the spin polarization of the conduction electrons due to Mn spins which mediates the RKKY interaction is almost zero. Therefore, the Mn spins might have only a minor influence on the interlayer coupling within each grain. Below  $T_b$ , the fluctuation of Mn spins ceases and the interlayer exchange interaction and the RKKY interaction among the Mn spins start to cooperate. The cooperation gives two influences on the interlayer coupling; i.e., a partial change of the sign of the interlayer coupling and an enhancement of  $H_s$ . Since the effect of distributed Mn ions on the interlayer coupling is not canceled out due to grains or co-

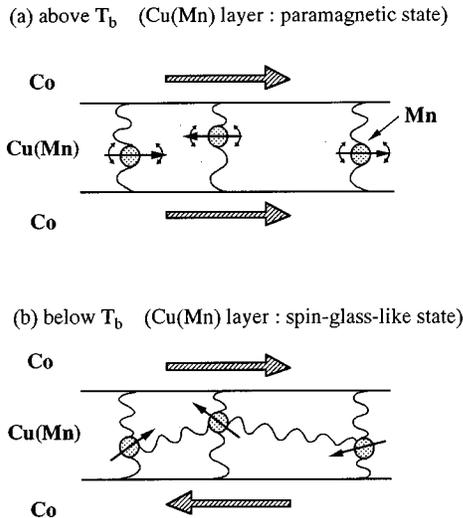


FIG. 9. Schematic illustrations of the switching of the coupling between Co layers above and below  $T_b$ . The solid wavy curves represent the RKKY interaction.

lumbar structures, the sign of the cooperative interaction depends on each grain. When the sign of the cooperative interaction below  $T_b$  is opposite from the interlayer exchange interaction above  $T_b$  in the grain, the coupling between Co layers changes [see Fig. 9(b)]. For  $d_{\text{Cu(Mn)}} \sim 13 \text{ \AA}$ , the ferromagnetically coupled Co layers are partially changed to AF coupling below  $T_b$ , while for  $d_{\text{Cu(Mn)}} \sim 9 \text{ \AA}$  where the Co layers are antiferromagnetically coupled, the F component grows below  $T_b$ . The cooperative interaction causes also a freezing of magnetizations of Co layers and Mn spins, which suppress the response of Co layers to the external field, leading to an enhancement of  $H_s$ . The influence of the cooperative interaction on the change of the interlayer coupling is also reflected in the dependence of  $H_s$  and  $T_b$  on Mn concentration. The  $H_s$  below  $T_b$  for the 13%-doped sample is

much larger than that for the 4%-doped one with same  $d_{\text{Cu(Mn)}}$  (see Fig. 6). The  $T_b$  for the 13%-doped samples is about twice higher than that for the 4%-doped one (see Fig. 7). Taking into account the fact that the RKKY interaction shows a damped oscillation as a function of the distance from a magnetic moment,<sup>26</sup> the stronger cooperative interaction is expected for the shorter interval among the Mn spins and Co layers, leading to a larger  $H_s$  for the larger Mn concentration. The higher  $T_b$  for 13 at. % Mn-doped samples mimics the concentration dependence of  $T_G$  for bulk Cu(Mn) alloy system.

## V. CONCLUSION

We found a drastic effect of Mn doping in the Cu spacer layers on the interlayer coupling between Co layers in Co/Cu multilayers; i.e., the interlayer coupling changes its sign from antiferromagnetic to ferromagnetic or vice versa partially with decreasing temperature. The change of the interlayer coupling becomes evident around a critical temperature. In order to explain the change of the interlayer coupling, we discussed a simple model based on the RKKY interaction cooperatively working among Mn spins and Co layers in correlation with spin-glass-like state in Cu(Mn) layers.

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