

**Inverse giant magnetoresistance in Fe/Cu/Gd<sub>1-x</sub>Co<sub>x</sub> spin-valves**

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For bottom spin-valves of NiO/Fe/Cu/Gd<sub>1-x</sub>Co<sub>x</sub>, giant magnetoresistance has been measured as a function of thickness and composition of GdCo layers as well as temperature. For all spin-valves involved here, the giant magnetoresistance has been attributed to contributions of spin-dependent scattering at interfaces and in bulk. The interfacial contribution produces positive giant magnetoresistance ratio for various compositions of GdCo alloys. However, the bulk contribution produces negative one for the Co contents from 50 at. % to a critical value  $x_C(R)$  (between 69 at. % and 77 at. %) and positive one for higher Co contents. It is suggested that the interfacial asymmetric factor of spin-dependent scattering is larger than 1.0 at the Cu/GdCo for various alloy compositions of GdCo and that the bulk asymmetric factor in GdCo layer is smaller and larger than 1.0 for Co contents below and above  $x_C(R)$ , respectively. For spin valves with Co contents below and/or above  $x_C(R)$ , the giant magnetoresistance ratio varies nonmonotonically and/or monotonically as a function of the GdCo layer thickness (temperature) for a specific temperature (a specific GdCo layer thickness), respectively. The change of the bulk asymmetric factor in the GdCo layer with the alloy composition can be attributed to the variation of either the spin alignment of Co and Gd atoms or the spin polarization of the GdCo layer.

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**I. INTRODUCTION**

Since the discovery of giant magnetoresistance (GMR) in magnetic multilayers and spin-valves, extensive experimental and theoretical investigations have been performed because of its important applications in magnetic storage devices.<sup>1-4</sup> In normal GMR, the resistivity is larger for antiparallel alignments of magnetizations in neighboring ferromagnetic layers than that of parallel configuration at saturation states, that is to say, the GMR ratio is positive. In inverse GMR effect, the resistivity for the antiparallel alignment of magnetization is smaller than that of the parallel alignment, that is to say, the GMR ratio is negative. The inverse GMR has been observed in many multilayers and spin valves.<sup>4-9</sup>

The GMR effect is attributed to originate from a competition of interfacial and bulk spin-dependent scattering, both of which are strongly related to the spin polarization of ferromagnetic layers. The spin polarization  $P_N$  is usually defined as  $[N(\uparrow) - N(\downarrow)] / [N(\uparrow) + N(\downarrow)]$ , where  $N(\uparrow)$  and  $N(\downarrow)$  are the density of states (including  $s$  and  $d$  electrons) at Fermi level for majority spins and minority ones, respectively.<sup>10</sup> The  $P_N$  can be determined by spin-polarized photoemission experiments. In contrast, tunneling spin polarization  $P_{TSP}$  can be determined by magnetic tunneling junction and is mainly contributed by conduction electrons,<sup>11</sup> where the  $s$  electrons are spin-polarized due to  $s$ - $d$  hybridization. For Co, the spin polarization  $P_N$  is negative. The resistivity of spin-down electrons is about one order larger

than that of spin-up electrons and the asymmetry factor of spin-dependent scattering  $\alpha = D(\downarrow) / D(\uparrow)$  is much larger than 1.0,<sup>7</sup> where  $D(\uparrow)$  and  $D(\downarrow)$  are the scattering possibility of spin-up and spin-down electrons, respectively. Although the spin polarization  $P_N$  of bulk Fe is positive due to its bcc structure, its magnitude is very small, in comparison with those of Co and Ni.<sup>12</sup> Nevertheless, the asymmetry factor  $\alpha$  has been shown to be still larger than 1.0.<sup>7</sup> Moreover, for most of interfaces at ferromagnetic (Fe, Co, and Ni)/noble metals (Au, Ag, and Cu), the  $\alpha$  is larger than 1.0, which might be related to the modification of density of the states of the ferromagnetic surface atomic layer by the noble spacer.<sup>13</sup> Therefore, the global asymmetric factor from Co and Fe layers is larger than 1.0 and no inverse GMR effect can be observed in Co/Cu/Fe spin valves. As the contents of Cr and V impurity in the Fe layers increase, the spin polarization  $P_N$  turns more positive<sup>9</sup> and the  $\alpha$  is smaller than 1.0, whereas the interfacial asymmetric factor at FeV(Cr)/Cu is larger than 1.0. For thin Fe alloy layers, the contribution of the interfacial scattering dominates and the global asymmetric factor from FeV is larger than 1.0 and thus the GMR ratio is positive. For thick Fe alloy layers, the contribution of the bulk scattering overcomes the interfacial one and the global asymmetric factor is smaller than 1.0 and then the GMR ratio is negative. Therefore, the contribution of the bulk scattering in FeV(Cr) layers to the GMR ratio of Co/Cu/FeV spin-valve is negative and that of interfacial scattering at the FeV(FeCr)/Cu is positive.

The inverse GMR phenomenon was also observed in CoFe/Ag/CoFeGd and Tb<sub>20.5</sub>Fe<sub>80</sub>Co<sub>20</sub>/Cu/Co/Gd<sub>22.4</sub>Fe<sub>80</sub>Co<sub>20</sub> spin-valves, and Co/Dy/Co/Cu/Co/Cu multilayers.<sup>14–17</sup> However, no information was given on the interfacial and bulk spin-dependent scattering while it is very important to understand the nature of the inverse GMR in rare-earth elements based spin-valves. Moreover, very recently, the tunneling spin polarization  $P_{\text{TSP}}$  of GdCo alloys has been measured and found to vary with the alloy composition and temperature.<sup>18</sup> Therefore, it is essential to study the spin-dependent scattering of alloys consisting of transition metals and rare-earth elements employing spin-valves or multilayers.

In this work, we will study the spin-dependent scattering of GdCo alloys measuring the GMR curves of Fe/Cu/GdCo spin-valves with various compositions and thickness of GdCo layers as well as temperature. As discussed above, the global asymmetric factor of spin-dependent scattering from the Fe layer is larger than 1.0 although the spin polarization of Fe is positive.<sup>12,19</sup> It is of crucial importance to study the bulk spin-dependent scattering of GdCo layers and interfacial one at the Cu/GdCo in the current-in-plane (CIP) configuration. As a typical ferrimagnet, the magnetization of GdCo can be controlled modifying the alloy composition. When the magnetization of Gd atoms is larger (smaller) than that of the Co atoms, the magnetization of Co atoms is antiparallel (parallel) to that of GdCo layers. Therefore, normal and inverse GMR effects can be expected for Fe/Cu/GdCo spin-valves with high and low Co contents, respectively. Inversely, the asymmetric factor of spin-dependent scattering in the GdCo layer is proved to vary with the alloy composition.

## II. EXPERIMENTS

Bottom spin-valves of glass/Ta(8 nm)/NiO(31 nm)/Fe(2 nm)/Cu(3 nm)/GdCo( $t$  nm)/Ta(1 nm) were fabricated by a magnetron sputtering system. The base pressure was  $2 \times 10^{-5}$  Pa and the argon pressure was 0.33 Pa during deposition. The metallic and NiO layers were sputtered by dc and rf sputtering, respectively. The GdCo layer was deposited from a composite target with setting small pieces of Gd on the Co target. The deposition rates of metallic and NiO layers were 0.1–0.2 nm/s and 0.04 nm/s, respectively. The wedge shape of GdCo layers was used to avoid run-to-run error. A magnetic field of about 133 Oe was applied parallel to the film plane and along the wedge direction to induce a uniaxial anisotropy in the ferromagnetic layers. The compositions of GdCo alloys were analyzed by x-ray fluorescence. The GMR ratio was measured using standard dc four-point probe method with the current and magnetic field perpendicular to each other and both parallel to the film plane, i.e., CIP configuration. GMR curves at low temperatures were performed in superconducting quantum interference device (SQUID). For all samples, the possibility that the inverse GMR comes from anisotropic magnetoresistance (AMR) effect is excluded measuring the resistance at various orientations of the external magnetic field with respect to the sensing current.

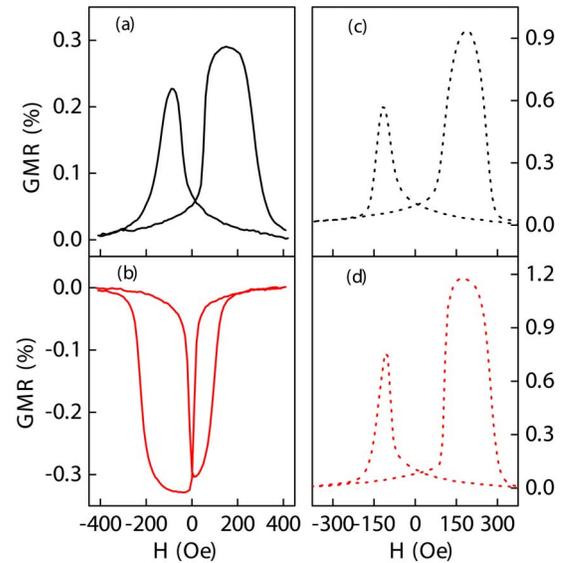


FIG. 1. (Color online) Typical GMR curves at room temperature for spin-valves Ta (8 nm)/NiO (31 nm)/Fe (2 nm)/Cu (3 nm)/Gd<sub>0.38</sub>Co<sub>0.62</sub>/Ta (1 nm) with 2.3 nm (a) and 4.2 nm (b) thick GdCo layers, Ta (8 nm)/NiO (31 nm)/Fe (2 nm)/Cu (3 nm)/Gd<sub>0.23</sub>Co<sub>0.77</sub>/Ta (1 nm) with 2.3 nm (c) and 4.2 nm (d) thick GdCo layers.

## III. RESULTS AND DISCUSSION

Figures 1(a) and 1(b) show typical curves of resistivity versus magnetic field at room temperature for Fe (2 nm)/Cu(3 nm)/Gd<sub>0.38</sub>Co<sub>0.62</sub> with 2.3 nm and 4.2 nm thick GdCo layers, respectively. The wide plateau in the curves hints the antiparallel alignment of the magnetizations in the Fe and GdCo layers. Apparently, the resistance change is induced by switching of magnetizations, and normal and inverse GMR effects originate from the spin-dependent scattering. The sign of the GMR ratio is strongly related to the thickness of GdCo layers for a specific alloy composition. Figures 1(c) and 1(d) show the GMR curves of spin valves Fe (2 nm)/Cu (3 nm)/Gd<sub>0.23</sub>Co<sub>0.77</sub> with 3.2 nm and 4.2 nm thick GdCo layers, respectively. For these two samples, the GMR ratio is 0.9% and 1.2%. With the same GdCo layer thickness of 3.2 nm, the GMR ratio at room temperature is –0.3% and 0.7% for contents of 62 at. % and 77 at. % Co, respectively. Therefore, the GMR ratio is also related to the composition of the GdCo layer for a specific thickness of the GdCo layer.

Figure 2(a) shows the dependence of the room temperature magnetization on the composition of GdCo alloys, which was provided by Hansen.<sup>20</sup> One can find that for Co contents lower than 50 at. %, the magnetization of GdCo alloys is almost equal to zero, exhibiting paramagnetic properties. The compensation composition  $x_C(M)$  is located at 80 at. % Co. For the Co contents from 50 at. % to the compensation composition  $x_C(M)$ , the magnetization of Gd atoms dominates and the net magnetization is parallel and/or antiparallel to the magnetization of Gd/Co atoms. At Co contents above the compensation composition  $x_C(M)$ , the magnetization of the GdCo alloy is parallel and/or antiparal-

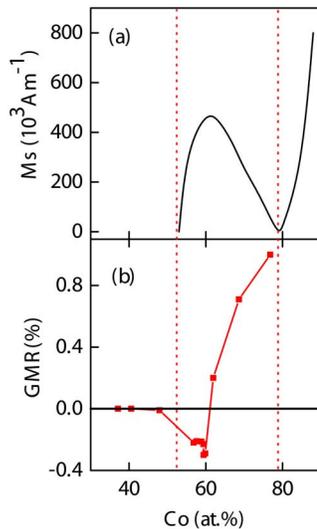


FIG. 2. (Color online) Dependence of magnetization at room temperature for GdCo alloy (Ref. 20) (a) and GMR ratio of the spin-valves Fe(2 nm)/Cu(3 nm)/GdCo (3.2 nm) (b) on the Co content of GdCo alloys. Measurements were performed at room temperature. The lines serve as a guide to the eye.

l to the moment of Co/Gd atoms. Figure 2(b) shows the compositional variation of the GMR ratio in Fe/Cu/GdCo spin-valves within the regime from 30 at. % to 77 at. %, where the GdCo layer thickness is fixed at 3.2 nm. The GMR ratio is equal to zero for Co contents below 50 at. %. As the Co content is increased, the GMR ratio becomes negative. Finally, the GMR ratio becomes positive for high Co contents. Apparently, the sign and the magnitude of the GMR ratio depend on the composition of GdCo layer.

In order to get deep insight into the nature behind the results in Fig. 2(b), the GMR curves were measured as a function of the GdCo layer thickness at various compositions of the GdCo layers. Figure 3 shows the GMR ratio versus the GdCo layer thickness for the spin valves of Fe (2 nm)/Cu (3 nm)/GdCo(0–6 nm). Remarkably, with increase of the GdCo layer thickness, spin-valves with dif-

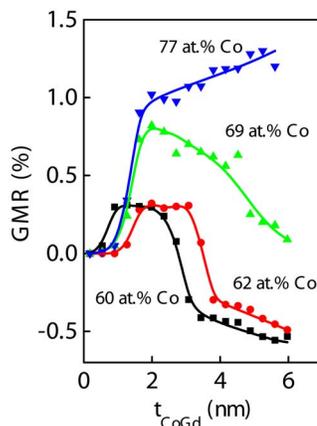


FIG. 3. (Color online) Dependence of GMR ratio at room temperature on the GdCo layer thickness for various compositions of GdCo. The lines serve as a guide to the eye.

ferent Co contents manifest various behaviors. For all four GdCo compositions, the GMR ratio initially increases to reach a positive maximum as the GdCo thickness is increased from zero to 2.0 nm. When the Co content is 77 at. %, the GMR ratio continues to increase with further increase of the GdCo layer thickness. For 69 at. % Co, the GMR ratio begins to decrease with further increase of the GdCo layer thickness. For 62 at. % and 60 at. % Co contents, the GMR ratio decreases sharply to negative values. A crossover for the GMR ratio from positive to negative has been observed during the variation of the GdCo layer thickness for low Co contents.

From the above results, three conclusions can be drawn for the present Fe/Cu/GdCo spin-valves. First of all, the GMR effect is contributed from the interfacial spin-dependent scattering and the bulk one. Then, for various compositions of the GdCo layer, the interfacial scattering at Cu/GdCo is larger than 1.0. This is in agreement with previously reported results that at ferromagnetic (Fe, Co, and Ni)/noble (Cu, Ag, and Au), the asymmetric factor is always larger than 1.0.<sup>4</sup> It might be induced by modification of electronic band structure of the surface atomic layer in the ferromagnetic layer through the spacer.<sup>13</sup> The exact reason is unclear that the asymmetric factor  $\alpha$  at the Cu/GdCo interface is always larger than 1.0 for either low or high Co contents, which needs further theoretical investigation. Accordingly, the positive GMR ratio can be induced by the interfacial spin-dependent scattering. Finally, in contrast, the magnitude and the sign of the contribution from the bulk scattering depend on the composition. For high and low Co contents, the contribution from the bulk scattering is positive and negative, and the asymmetric factor  $\alpha$  in the GdCo layer is larger and smaller than 1.0, respectively. There is a compensation composition  $x_c(R)$  between 69 at. % to 77 at. % Co, at which the sign of the contribution of the bulk scattering changes. It is noted that there is a difference between the  $x_c(R)$  and  $x_c(M)$ , which might be induced by an error of the composition measurements.

The compositional dependence of the asymmetric factor in the GdCo layer can be understood by intuition in terms of spin arrangements of Fe and GdCo layers. Since the magnetic moment of Gd atoms comes from 4f electrons and is shielded by 5d and 6s electrons, the effect of spin-dependent scattering from atomic moment of Gd is expected to be very weak, in comparison with that of Co atoms, that is to say, the spin-dependent scattering of GdCo alloys originates mainly from Co atoms. As shown in Figs. 4(a) and 4(b), for low Co contents, the magnetization of Gd atoms dominates and the net magnetization is antiparallel to that of the Co atoms. This way, if the magnetization of the Fe layer is antiparallel to that of the GdCo layer at low magnetic fields, the magnetic moments of Fe and Co atoms are parallel to each other. Similar to Co/Cu/Fe spin-valves with parallel alignment of magnetic moments in Co and Fe layers, the sheet resistance at low magnetic fields is low in terms of the bulk scattering. When the magnetic moments of Fe and GdCo layers are parallel to each other at saturation fields, magnetic moments of Fe and Co atoms are antiparallel to each other and thus the sheet resistance is high at high magnetic fields. Therefore, the contribution of the bulk scattering is negative and the

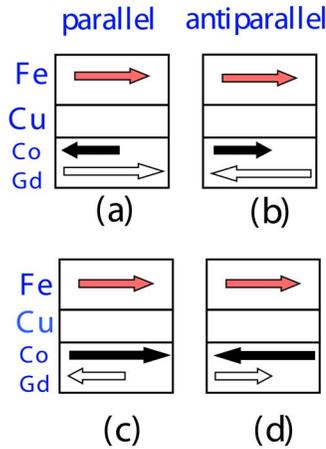


FIG. 4. (Color online) Schematic configurations of atomic magnetic moments of Co (solid black arrow), Gd (white open arrow) atoms in GdCo layers and Fe [red (gray) solid arrow] atoms. In (a) and (b), the magnetization of Gd atoms is larger than that of Co atoms, and vice versa in (c) and (d). The magnetizations of Fe and GdCo layers are parallel (left-hand column) and antiparallel (right-hand column) to each other.

inverse GMR effect occurs without any interfacial contribution. For Co contents of 60 at. %, 62 at. %, and 69 at. %, nonmonotonic variation of the GMR ratio can be observed as a function of the GdCo layer thickness, due to opposite contributions of the interfacial and bulk scattering. For high Co contents, the situation in Figs. 4(c) and 4(d) is similar to the Fe/Cu/Co spin-valves. Since the contributions of the bulk and interfacial scattering are both positive, spin-valve has a normal GMR effect and a monotonic variation of the GMR ratio occur for the content of 77 at. % Co.

The above results can also be explained as a result of variation of the spin polarization  $P_N$  of GdCo alloys with the alloy composition. As shown in Figs. 5(a) and 5(c), the spin polarization  $P_N$  of GdCo alloys for high Co contents is expected to be negative like pure Co. In this case, the asymmetric factor is larger than 1.0 for the GdCo layer. Since the asymmetric factor is larger than 1.0 at Fe/Cu and Cu/GdCo interfaces and in the GdCo and Fe layers,<sup>7</sup> no inverse GMR can be observed, as shown in Fig. 3. For low Co contents, the asymmetric factor  $\alpha$  in the GdCo layer is smaller than 1.0 and the contribution of the bulk scattering to the GMR ratio is negative. The spin polarization  $P_N$  of the GdCo layer is expected to be positive, opposite to that of Co. The schematic picture is shown in Figs. 5(b) and 5(d). Unambiguously, the spin polarization  $P_N$  of the GdCo layer changes with the alloy composition. It is noted that the tunneling spin polarization  $P_{TSP}$  of GdCo alloys can be adjusted modifying the alloy composition.<sup>18</sup>

In order to study the temperature dependence of the GMR ratio, GMR curves were measured at low temperatures. Figure 6 shows the typical GMR curve as well as the magnetization loop for the sample Fe(2 nm)/Cu(3 nm)/Gd<sub>31</sub>Co<sub>69</sub>(3.4 nm) at 10 K. Figures 7(a) and 7(b) show the GMR ratio as a function of temperature for Fe/Cu/Gd<sub>0.38</sub>Co<sub>0.62</sub> and Fe/Cu/Gd<sub>0.31</sub>Co<sub>0.69</sub> spin-valves with various thickness of GdCo layers. Apparently, the tem-

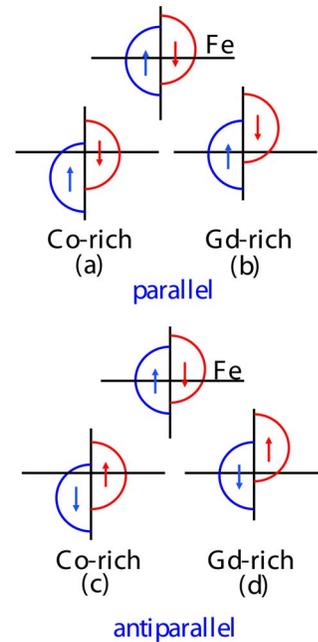


FIG. 5. (Color online) Schematic illustration of density of states in GdCo and Fe layers. The magnetizations of Fe and GdCo layers are parallel (a) and (b) and antiparallel (c) and (d) to each other, with (a) and (c) for Co rich and (b) and (d) for Gd rich.

perature dependence of the GMR ratio is strongly related to the GdCo layer thickness. For large thickness of the GdCo layer, such as 3.4 nm and 3.8 nm, the GMR ratio increases monotonically from more negative values to less negative ones and even to positive ones in the temperature regime from 5 K to 300 K. This is because the contribution of the

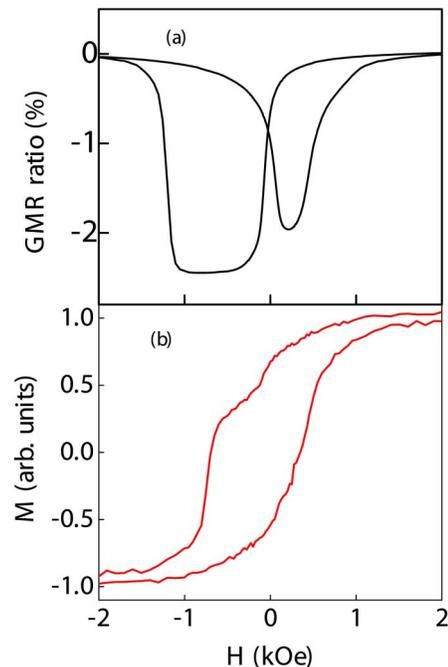


FIG. 6. (Color online) The GMR curve (a) and magnetization loop (b) for Fe(2 nm)/Cu(3 nm)/Gd<sub>31</sub>Co<sub>69</sub>(3.4 nm) at 10 K.

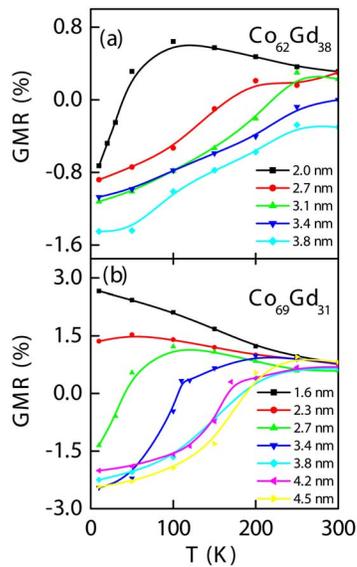


FIG. 7. (Color online) Dependence of GMR ratio on temperature for various GdCo layer thickness and  $\text{Gd}_{38}\text{Co}_{62}$  (a) and  $\text{Gd}_{31}\text{Co}_{69}$  (b). The lines serve as a guide to the eye.

bulk scattering is negative to the GMR ratio and plays a dominant role in the entire temperature regime. It is indicated that for the two series of samples the asymmetric factor of the GdCo layer is smaller than 1.0 from 5 K to 300 K. For very small GdCo layer thickness, such as 1.6 nm in Fig. 7(b), there is no contributions from the bulk scattering and the GMR ratio of the spin-valve is positive and increases with decreasing temperature. It is shown that the asymmetric factor at Cu/GdCo interface is larger than 1.0 from 5 K to 300 K. For an intermediate GdCo layer thickness, such as 2.0 nm in Fig. 7(a) and 2.7 nm in Fig. 7(b), the GMR ratio changes nonmonotonically with temperature. This phenomenon clearly hints that the mean free path in the GdCo layers changes with temperature.<sup>21</sup> At high temperatures, the mean free path is small, the contribution of the interfacial scattering dominates over the bulk contribution, and the GMR ratio is positive and its magnitude is increased with decreasing temperature. With further decreasing temperature, the mean free path is increased and the contribution of the bulk scattering starts to appear and finally dominates over the interfacial one and the GMR ratio becomes negative accordingly.

Figure 7 also shows that there is a crossover for the GMR ratio from positive to negative values as a function of the GdCo layer thickness for a fixed temperature. Alternatively, a crossover occurs as a function of temperature for a fixed (intermediate) GdCo layer thickness. This is because the contributions of the interfacial and the bulk spin-dependent scattering are opposite. More remarkably, the crossover value is shifted towards larger GdCo layer thickness at higher temperatures. Equivalently, the crossover temperature is shifted towards higher temperatures at larger GdCo layer thickness. Figure 8 shows the results for spin-valves Fe/Cu/ $\text{Gd}_{38}\text{Co}_{62}$ . It is clearly indicated that with increasing temperature the asymmetric factor of bulk scattering is decreased more sharply than that of the interfacial scattering. In order to explain the results in Fig. 8, the increase of the mean

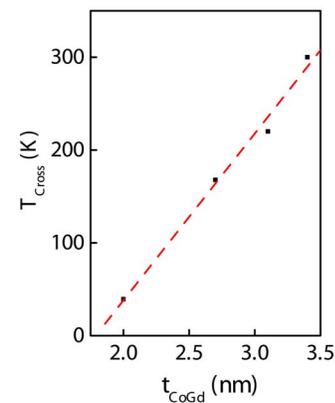


FIG. 8. (Color online) For Fe/Cu/ $\text{Gd}_{38}\text{Co}_{62}$  spin-valve, the crossover temperature increases with increase of the GdCo layer thickness. The lines serve as a guide to the eye.

free path of the GdCo layer with decreasing temperature should also be taken into account, in addition to the competition between the interface scattering and the bulk scattering.

#### IV. CONCLUSION

In summary, we have prepared glass/Ta/NiO/Fe/Cu/ $\text{Gd}_{1-x}\text{Co}_x$  spin-valves by magnetron sputtering. GMR curves have been measured as a function of alloy composition and thickness of GdCo layers as well as temperature. For spin-valves with fixed GdCo layer thickness of 3.2 nm, the room temperature GMR ratio is negative for Co contents from 50 at. % to 62 at. % and is positive for Co contents higher than 62 at. %. For Co contents below 69 at. %, it first increases to a positive maximum with increase of the GdCo layer thickness and then decreases to negative with further increase of the GdCo layer thickness. For high Co contents, it increases monotonically with the GdCo layer thickness. The variation of the GMR ratio with temperature depends on thickness of the GdCo layer, in addition to the alloy composition of the GdCo layers. At low Co contents, the GMR ratio increases from negative to positive with increasing temperature for thick GdCo layers, whereas it is positive and decreases with increasing temperature for thin GdCo layers. For intermediate GdCo layer thickness, it changes nonmonotonically with temperature. For high Co contents, the positive GMR ratio changes monotonically with temperature and GdCo layer thickness. It is suggested that the interfacial asymmetric factor of spin-dependent scattering at Cu/GdCo is larger than 1.0 for all compositions of the GdCo layer and that the bulk one in the GdCo layer is larger and smaller than 1.0 as the Co content is above and below the compensation composition, respectively. In order to explain the nonmonotonic variation of the GMR ratio with temperature and GdCo layer thickness, the effect of temperature on the mean free path should be considered. The variation of the bulk asymmetric factor in the GdCo layer with the alloy composition can be explained in terms of the variation of either the spin alignment of Co and Gd atoms or the spin polarization of the GdCo layer.

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