Magnetoresistance of spin-dependent tunnel junctions with composite electrodes

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Spin-dependent tunnel junctions, $Co/Al_2O_3/Co$ (CoFe)/NiFe, were fabricated to investigate the effect of the additional Co (CoFe) interlayer on tunneling magnetoresistance. The quality of the junction was examined with a cross-sectional image generated by high-resolution transmission electron microscopy, and an electron energy loss spectra map. For junctions with a Co (CoFe) interlayer in the top electrode thinner than 0.8 nm (1.0 nm), the tunneling magnetoresistance ratio increases with interlayer thickness. For junctions with a 0.8–2.0 nm Co (1.0–2.0 nm CoFe) interlayer in the top electrode, the tunneling magnetoresistance ratio reaches the maximum value of 2.16 (4.45) times that without any Co (CoFe) interlayer in the top electrode. The increase in the tunneling magnetoresistance ratio may be attributed to the increased effective ferromagnetic electrode polarization and the various spin-flip scattering factors. [DOI: 10.1063/1.1419259]

I. INTRODUCTION

Spin-dependent tunnel (SDT) behavior between a pair of ferromagnetic layers separated by an insulator layer are currently the subject of much research.[1] These behaviors are fundamentally important in understanding spin-polarized electron tunneling and applications in digital devices, such as sensors and magnetic random access memory (MRAM). The tunneling magnetoresistance (TMR) ratio is defined as $(R_{\uparrow\downarrow}-R_{\uparrow\uparrow})/R_{\uparrow\uparrow}$, where $R_{\uparrow\uparrow}$ and $R_{\uparrow\downarrow}$ are the resistances for parallel and antiparallel spin alignment states of two ferromagnetic layers in the SDT junction, respectively. The ratio can be generally described by Julliere's model:[2]

$$\text{TMR ratio} = \frac{2P_1P_2}{1-P_1P_2}\times 100\%. \tag{1}$$

Where, P_1 and P_2 are spin polarization values of conduction electrons in two ferromagnetic electrodes (FM electrodes). Many details affecting TMR, such as the interfacial effect, are not yet been fully understood due to difficulties in fabricating and controlling the quality of SDT junctions. The spin-flip scattering between tunnel electrons and impurities in the insulator or at the insulator–ferromagnet interface represent an important effect. The presence of impurities, such as Co-, Ni-, Cu-, and Pd-based ions, in the insulator layer of $Co/Al_2O_3/Ni_{80}Fe_{20}$ SDT junctions has been reported to cause a reduction of the TMR ratio, due to the spin scattering.[3] Another important interfacial effect is the increased polarization of FM electrodes by adding high-polarization materials at the insulator–ferromagnet interface. This increase follows from the influence of the ferromagnet-insulator coupling on the effective polarization of the FM electrode, as indicted in an earlier theoretical work.[4] A higher polarization value of the additional interlayer yields a higher effective polarization value of the FM electrode, and therefore a higher TMR ratio. Jansen *et al.* have reported an increase in the TMR ratio by up to 1.25 times by adding Fe-based ions in the insulator of SDT junctions.[5]

This article presents the influence on TMR, as a function of thickness, of different additional interlayers, Co and CoFe, deposited at the interface between the insulator and the FM electrode of SDT junctions. The TMR ratio was dramatically increased by both Co and CoFe interlayers in the low interlayer thickness range, but was reduced at higher coverages of FM interlayer.

II. EXPERIMENTAL DETAILS

All SDT junctions were prepared in a high-vacuum magnetron sputtering system with a base pressure below 3×10^{-7} Torr. Three contact masks were employed to deposit the rectangular bottom Co electrode layer (10 nm), the circular insulator Al_2O_3 (2.3 nm), and the rectangular NiFe electrode layer (10 nm) on 7059 Corning glass substrates. Co and NiFe were deposited by dc power with a deposition voltage of around 300 V in 5 mTorr of Ar ambient. The insulator layer was formed using RF glow discharge (64% Ar + 36% O_2) with a −350 V bias voltage on the Al film. The additional interlayers Co and CoFe were grown at a slow deposition rate (0.02–0.05 nm/s) on the Al_2O_3 layer. A 50:50 CoFe alloy was used here.

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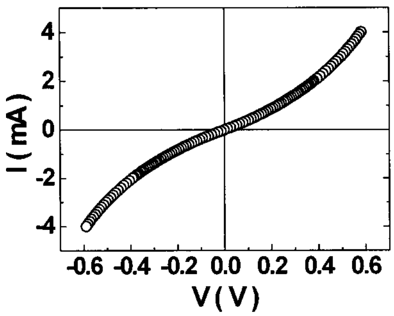

FIG. 1. $I-V$ curve for SDT junction, $Co/Al_2O_3/2.0$ nm Co/NiFe.

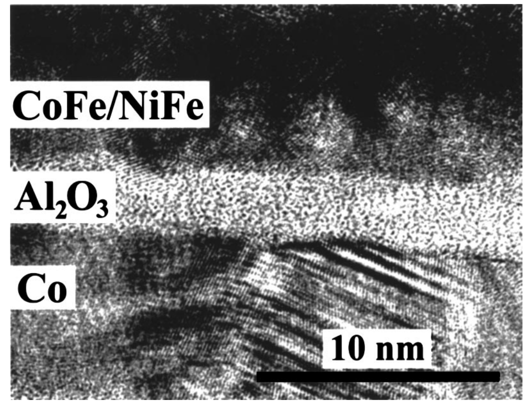

FIG. 2. Cross-sectional TEM image of SDT junction, $Co/Al_2O_3/1.2$ nm CoFe/NiFe.

A cross strip junction with 1 mm×1 mm area was fabricated for the four-probes measurement of the tunnel resistance in a current perpendicular to the film plane (CPP). Electrical properties were measured using a dc source at room temperature. Magnetic properties, such as the switching field, were determined by a longitudinal magneto-optical Kerr effect (MOKE) instrument.

The electron microscopic observation of the cross section of the SDT junction first followed standard thinning procedures.[6] The sample was mechanically polished, dimpled to a thickness of 3 μm, and then ion milled (usually at 5 keV, 1 mA) to perforation. The image of the cross-section image was taken by a JEOL 2010F field emission gun high-resolution electron microscope (HRTEM) equipped with an Oxford energy dispersive x-ray spectrometer (EDS). All images were obtained at an electron accelerating voltage of 200 kV.

Electron energy loss spectroscopy (EELS), in a reflective geometry, is a powerful tool with which for probing electron excitation at surfaces of ultra-thin films, with an element sensitivity which yields nanometer spatial resolution. The incident electron beam was normal to the film plane in TEM and EELS measurements. EDS and EELS were conducted with a 0.5 nm nanobeam probe.

III. RESULTS AND DISCUSSION

The tunnel resistivity, that is the *RA* value defined as the product of the tunnel resistance *R* and the junction area *A*, of the SDT junctions, glass substrate//Co (10 nm)/ Al_2O_3 (2.3 nm)/Co and CoFe/NiFe (10 nm), varied from 10 to 100 $M\Omega\,\mu m^2$. The tunneling properties of SDT junctions were examined by $I-V$ measurement. Figure 1 plots the $I-V$ curve of the SDT junction with the additional Co interlayer of 2.0 nm. This curve can be fitted by Simmons' formula,[7] $J=\alpha V+\gamma V^3$, to give the effective barrier width, d, and height, ϕ, where J is the current density through the SDT junction and V is the bias voltage across two FM electrodes. Two coefficients, α and γ, are functions of d and ϕ. The fitting results show that all of the SDT junctions considered here have a ϕ of 1–2 eV and a d of 2.3–2.5 nm. These values are consistent with those of earlier research,[8,9] implying sufficient insulator quality.

Figure 2 depicts the cross-sectional TEM image of the SDT junction with the additional 1.2 nm thick CoFe interlayer. Both the top and bottom FM electrodes are polycrystalline, as commonly observed in sputtered thin films. The interfacial root-mean-square roughness of the individual layer is relatively low, only 0.22–0.34 nm, suggesting that the pinhole may not exist, and that no shunting current, other than the tunneling current, flows between the two FM electrodes. The average thickness of the Al_2O_3 layer is 2.9 nm. This value is compatible with the effective barrier width fitted above by the $I-V$ curve. The average ratio of the composition of Al–O is 2.00:3.06 from EDS analysis, revealing high insulator quality.

Although the metallic layers are polycrystalline, the in-plane anisotropy has a geometrically induced preferable orientation. In the TMR measurement, the applied magnetic field was along the easy axis. Figure 3 shows the typical TMR loop for the SDT junction with the additional 1.6 nm thick CoFe interlayer. This junction exhibits a saturated RA value of 36.7 $M\Omega\,\mu m^2$. The magnetization of the soft CoFe/NiFe layer switches first at a magnetic field of -10 Oe ($+10$ Oe) for decreasing (increasing) branch, while the bottom Co electrode switches at a field of -14 Oe ($+14$ Oe). The inset in Fig. 3 displays the corresponding MOKE response. The step feature of the hysteresis loop reveals the antiparallel magnetization state over the field range, 10–14 Oe. The maximum TMR ratios are 18.7% and 17.5% for decreasing and increasing branches of TMR loop, respectively, before the magnetization switching of the Co electrode. The highest tunnel resistance during the sweeping of the magnetic field may be missed since the tunnel resistance is very sensitive to

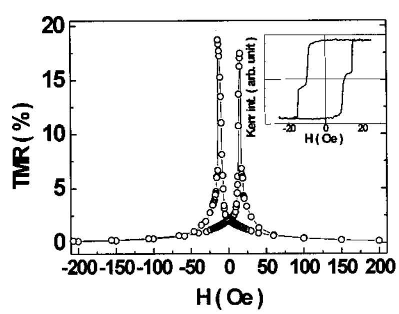

FIG. 3. Typical room temperature TMR loop of SDT junction, $Co/Al_2O_3/1.6$ nm CoFe/NiFe. The magnetic field is applied along the easy axis. The inset shows the corresponding hysteresis loop.

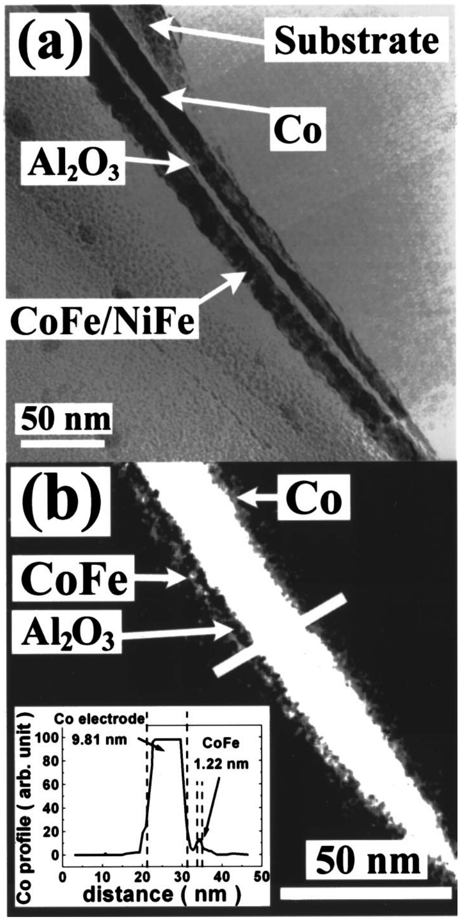

FIG. 4. EELS maps of SDT junction (Co/Al_2O_3/1.2 nm CoFe/NiFe). (a) Zero loss map. The tilted substrate is prepared by ion milling; (b) Co map. The energy loss is fixed at 779 eV of K edge for Co. The inset is the line scan (white straight line across the map) for the Co profile. The low Co signal of the additional 1.22 nm CoFe interlayer is due to the limit of the nanobeam probe.

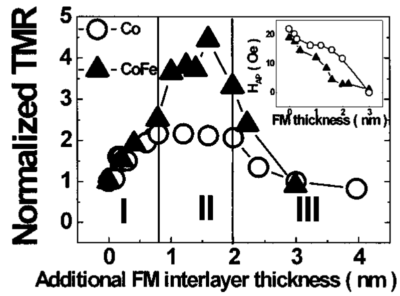

FIG. 5. Normalized TMR of SDT junctions as functions of the additional FM (Co and CoFe) interlayer thickness. The normalized TMR is defined as the ratio of the TMR ratio for SDT junctions to that for a junction without the additional FM interlayer (Co/Al_2O_3/NiFe). The inset shows the field range of the antiparallel magnetization state, H_{AP}, which is defined as the switching field difference between the top and the bottom electrodes, as a function of the additional FM interlayer.

the magnetic field near the bottom electrode switching field and the magnetic field resolution limit is 0.5 Oe. Missing the highest resistance slightly changes the maximum TMR ratios for decreasing and increasing branches of the TMR loop.

Figure 4 displays a EELS map of an SDT junction with the additional 1.2 nm thick CoFe interlayer. Figure 4(a) shows the zero loss map of four different layers. From top to bottom, they are milled substrate, the electrode Co, the insulator Al_2O_3, and the electrode with the additional FM interlayer CoFe/NiFe. This map is similar to the TEM image of Fig. 2. The ultra-thinness of CoFe is such that CoFe and NiFe layers can be distinguished neither by TEM nor the zero loss EELS map. However, as shown in Fig. 4(b), the weak but clear Co signal of CoFe layer next to the Al_2O_3 layer can be observed in the Co EELS map at the same place as the signal from Fig. 4(a), where the energy loss is fixed at 779 eV of K edge for Co. A complete CoFe layer is grown on the Al_2O_3 layer. Figure 4(b) shows the relative EELS intensity of the Co line scan profile. The thickness of CoFe is estimated as 1.22 nm.

Figure 5 summarizes the normalized TMR as a function of the additional Co and CoFe interlayer thicknesses. The normalized TMR is defined as the ratio of the TMR ratio of all SDT junctions, to that of the junction without the additional FM interlayer. For SDT junctions with the additional Co interlayer, the normalized TMR increases from 1 to 2.16 as Co thickness increases from 0 nm to 0.8 nm (region I). The normalized TMR of the SDT junctions with the additional CoFe interlayer increases from 1 to 3.80 with CoFe interlayer thickness, in a similar region (0–1.0 nm). According to the Julliere's model, this result implies that the effective polarization values of SDT junctions are increased by the presence of the additional FM interlayer. As claimed by Slonczewski,[4] the effective polarization of the tunnel electron is chiefly governed by the ferromagnet-insulator coupling. Thus, the polarization values of the additional FM interlayer near the insulator layer must be an important source of the increased TMR ratio found in SDT junctions with a Co or CoFe interlayer. For SDT junctions with the additional Co and CoFe FM interlayer thickness ranging from the end of region I to 2.0 nm (region II), the normalized TMR remains in the range, 2.06–2.16 (8.7%–9.1% of the TMR ratio), and 3.80–4.45 (16.0%–18.7% of the TMR ratio), respectively. Furthermore, in region III, the normalized TMR for both additional Co and CoFe interlayers decreases dramatically to approximately 1, almost equal to the TMR ratio of a junction without the additional FM interlayer. The inset in Fig. 5 presents the field range of the antiparallel magnetization state, H_{AP}, which is defined as the switching field difference between two electrodes, while varing the additional Co and CoFe interlayers. Overall, H_{AP} declines monotonically with increasing thickness of the additional FM interlayer. This result implies that the increasing TMR in region I and the variation observed in region II follow from a change in the transport behavior, rather than from changes in the coercive field which would enable a more stable or extended antiparallel magnetization state.

The polarization value of the FM interlayer can be considered to be the effective polarization value of the FM electrode, since the additional FM interlayer with region II thickness entirely covers Al_2O_3 as shown in Fig. 4(b). Previous studies have reported polarization values of CoFe, Co, and NiFe are 47%–53%, 34%–45%, and 32%–48%.[10–12] The wide range of polarization values for these three materials may follow from the quality of samples and experimental methods employed. Possibly the highest normalized TMR can thus be estimated as 1.50 and 1.85 for the additional Co and CoFe interlayers, respectively, according to Julliere's model. However, these two values are much lower than the experimental results (2.16 and 4.45) presented in Fig. 5. Another influencing factor can be assumed to apply. The various spin-flip scattering factors due to various magnetic ions at the interface of Al_2O_3 and ferromagnetic layers, such as Ni^{+2}, Ni^{+3}, and Co^{+2} ions,[3] may influence the spin-dependent transport behavior, and thus the TMR ratio. The presence of magnetic ions may follow from the diffusion of oxygen ions from the Al_2O_3 layer to FM electrodes, due to the thermal stability and the activated energy difference between Al_2O_3 and FM electrodes.

In region I of Fig. 5, the TMR ratio increases monotonically with increasing additional FM interlayer thickness. Two causes may apply this phenomenon. The first is the cluster-like formation of the additional FM interlayer. Unfortunately, the limited EELS resolution ($\sim$1 nm) is such that the EELS map of the SDT junction in region I cannot be clearly determined. However, the assumption of the discrete ultrathin additional FM interlayer remains reasonable in the common sputtering growth process. Both the cluster-like additional FM interlayer and the NiFe layer contribute to the effective polarization value of the top FM electrode. Accordingly, the effective polarization exceeds that of the pure NiFe layer. Another possible reason is the size effect of the additional FM interlayer on the polarization value. Upadhyay *et al.*[12] elucidated the size effect on the polarization value of ultrathin Co films (under 1 nm thick), determined in a transport experiment. A highly sensitive and monotonic increase in the polarization with Co thickness was identified. This result suggests that the polarization values of the additional FM interlayers (both Co and CoFe), and in turn, the effective electrode polarization increase rapidly with the thickness of the additional FM interlayer over the low coverage range.

As indicated by the EELS data in Fig. 4(b), the additional FM interlayer with a thickness in region II exhibits complete layer formation. The polarization of the FM interlayer may reach a saturation value. Hence, the TMR ratio in region II is maintained almost constant. As mentioned above, the large difference between the results for the additional Co and CoFe interlayers in region II may follow from two possible causes. The first is the polarization behavior. The second is the presence of the ionized Fe at the interface between the insulator and the top electrode. Jansen *et al.* demonstrated the enhancement of TMR by Fe ions.[5]

The coercivity of the top FM electrode increases with the additional FM interlayer thickness due to the directive exchange coupling of the additional FM interlayer and NiFe as the coercivity of the additional FM interlayer exceeds that of NiFe. Thus, the field range of antiparallel spin alignment between the top and bottom FM electrodes decreases with an increasing additional FM interlayer thickness. For region III, the antiparallel field range approaches zero. The absence of a perfect antiparallel spin alignment between the top and bottom FM electrodes reduces the normalized TMR for both the additional Co and the additional CoFe interlayer.

IV. CONCLUSION

The TMR was demonstrated to be strongly influenced by adding the additional FM interlayer at the interface between the insulator layer and the FM electrode. An enhanced factor of 2.16 (4.45) times the TMR ratio of the SDT junctions was obtained by adding a 0.8–2.0 nm Co (1.0–2.0 nm CoFe) interlayer at the interface of the insulator and the FM electrode. The EELS map showed generation of a continuous additional FM interlayer in the ultrathin limit at $\sim$1.2 nm. The presence of an ultra-thin additional FM interlayer may change the detailed behavior of the electrode-insulator coupling at interface, possibly leading to a complex interplay between the effective polarization and the spin-flip scattering process, in turn greatly influencing the TMR.

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