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Enhanced Spin Polarization by An Extra Co or CoFe Layer in FM/Insulator/FM Structures

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Abstract - Spin-dependent tunnel magnetoresistance of Co(10nm)/Al_2O_3(2.6nm)/M(tnm)/ NiFe(10nm) with M = Co or CoFe was studied as a function of the thickness of M_t. The thickness (t) of the doped Co or CoFe was varied between 0 and 3 nm. We have experimentally showed that the polarization of tunneling electrons can be varied by modifying the interfacial condition between the insulator and magnetic layers. The enhancement of the tunneling magnetoresistance in samples of Co(10nm)/ Al_2O_3(2.6nm)/CoFe(tnm)/NiFe(10nm) is greater than that in samples of Co(10nm)/ Al_2O_3(2.6nm)/Co(tnm)/ NiFe(10nm) for t between 0.8 and 2.0 nm. The enhanced tunneling magnetoresistance may be attributed to the increase in the effective polarization of the tunnel electrons due to the spin-filtering effect from the additional magnetic layer M. Besides, from the oxidation process study of the Al-O insulator layer on Corning glass, we have shown experimentally that the optimal condition for the oxidation of the Al-O insulator layer in FM/Al-O/FM system can be obtained under the pressure of $5x10^{-2}$ torr for 50% of Ar + 50% of O_2 environment with both natural and plasma oxidations. For optimal oxidation of the Al-O insulator layer in a spin-dependent tunnel system, its thickness should be less than 3 nm

Key words: TMR, tunnel magnetoresistance, spin-filtering effect,

1. Introduction

The studies of spin-dependent tunneling junctions (SDT) are currently an active field of research because of its possible technological applications in data storage [1,2]. In general, the large tunneling magnetoresistance (TMR) ratio between two ferromagnetic metal layers (FM1 and FM2) separated by a thin insulator (I) can be explained by the Julliere model [3], that polarization of magnetic layers plays the dominant role. However, many essential factors such as the interfacial effect in TMR have not been clearly understood due to the fabrication difficulties of SDT junctions. One of the important interfacial effects is the spin-flip scattering by metallic impurities at the interface between the insulator and ferromagnetic electrodes. It is very interesting to investigate the exchange mechanism in FM1/I/FM2 system by adding a magnetic or a nonmagnetic metal interface layer into the insulating layer or between the insulating and magnetic layers as well as by the quality of the insulating layer. The presence of metallic impurities, such as Co, Ni, Cu, or Pd in the tunnel barrier of Co/Al_2O_3/$Ni_{80}Fe_{20}$ SDT junction have been reported to result in a reduction of TMR ratio due to the spin-flip in the spin-scattering process [4-6]. An enhancement of TMR ratio at high bias voltage has been experimentally studied in Co/Al_2O_3/Co/Al_2O_3/$Ni_{80}Fe_{20}$ system [7]; and in the Coulomb blockade regime has been theoretically studied in insulating Co-Al-O granular films [8]. In this paper, we report an experimental study on the enhancement of the polarization of the tunneling electron by the addition of an extra Co or CoFe layer in the interface of the Al_2O_3 and NiFe layers in the Co/Al_2O_3/NiFe system. We also report oxidation effect on an aluminum oxide insulator.

2. Experimental

The NiFe(10 nm)/M(t)/Al-O(2.6 nm)/Co(10 nm) SDT junctions with M being either Co or CoFe, and t varying from 0 to 3 nm were grown by a rf and dc magnetron sputtering system on 7059 corning glass substrates. The base pressure was lower than 3×10^{-7} torr. Co and NiFe were deposited by DC magnetron sputtering with a deposition rate of 0.3 ~ 0.4 nm/s and the insulator is made by RF glow discharge (50% of Ar + 50% of O_2) of the Al deposited on the bottom Co electrode. The oxidization of the aluminum layer was performed by both plasma and natural oxidization. Cross strip junctions with 1mm×1mm dimension was designed to measure junction resistance by current perpendicular to plane (CPP) and 4-probe methods. Electrical properties including TMR and current-voltage relation (I-V curve) were measured by using a DC source. AFM (atomic force microscopy) was used to check the morphology of films. All of measurements were made at room temperature.

3. Result and Discussion

The tunnel resistance of the fabricated SDT junctions, glass substrate // Co (10 nm) / Al-O (2.6 nm) /Co or CoFe (t nm) / NiFe (10 nm), ranges from $2x10^2$ to 10^3 Ω. This resistance is much larger than the resistance of ferromagnetic electrodes (~ 10 to 30 Ω). Hence, the effect of the non-uniform current distribution in SDT junctions [9] during measurement of the resistance of SDT junctions by the 4-probe method can be excluded. The product of the resistance and the junction area (RA) was found in the same range of 2-10 Ω-cm^2 for all the junctions. As an example, Figs. 1(a) and (b) show the TMR loop for SDT of the Co (10 nm) / Al-O (2.6 nm) /M (t nm) / NiFe (10 nm) system. In Fig. 1(a), M = Co, for sample with t = 0.0 nm, the tunneling resistivity which is defined as the product of the tunnel resistance and junction area and the TMR ratio are 3.6 Ω-cm^2 and 3.5 %, respectively. The coercivity of the top and bottom electrodes is approximately 6 and 23 Oe, respectively. A TMR of approximately 9 % for the sample with t = 1.2 nm is shown in the middle part of Fig. 1(a). In the lower part of Fig. 1(a), the TMR ratio for the sample with t = 3.0 nm is ~ 4%, however, under this condition, the coercivity of the top and bottom electrodes is approximately the same, because the inserted 3.0 nm thick Co layer will be enough to dominate the top electrodes. Fig. 1(b) presents similar TMR behavior for samples with M = CoFe. A TMR of approximately 14 % for the samples with t = 1.3 nm is shown in the middle part of Fig. 1(a). An enhancement of the TMR for samples with t less than 2 nm were observed. This may be attributed to the increasing of the spin polarization of the top NiFe layer due to the additional Co or CoFe layer between the insulator and the NiFe layers. For samples with t larger than 2.0 nm, its TMR decreases manifestly as shown in the lower part of Fig. 1. It is not clear yet that the polarization is a very step function of t. Further quantitative analyses are in progress now and will be reported later.

The I-V curves for these junctions showed approximately the same behavior, with a slight variation of the barrier width and height. We can fit the curves by Simmon's appropriate formula [10]: $J = \alpha V + \gamma V^3$ to obtain the effective barrier width (d) and height (ϕ), where J is the current density in the SDT junction and V is the bias between the two ferromagnetic electrodes. Here, two coefficients, α and γ, are functions of d and ϕ. The effective barrier width and barrier height by fitting I-V curves from Simmon's formula as show in Fig. 2 are 2.1 nm and 1.2 eV for undoped samples, 2.6 nm and 1.3 eV for samples doped with Co, and 2.4 nm and 1.3 eV for samples doped with CoFe, respectively. These values are consistent with those of the previous studies [11,12], indicating an insulating behavior.

From the AFM measurement, the root-mean-square roughness is ≤ 1 nm for all the samples. In general, it is smaller than the thickness of the Al-O insulator layer. To further understand the insulator layer, the oxidation effect of an oxidized aluminum layer formed by either natural or plasma oxidization of aluminum films was studied by *in-situ* resistance measurement of oxide

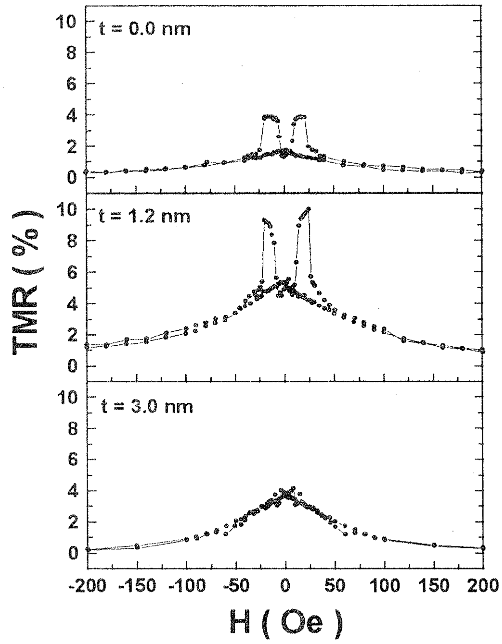

Fig. 1a TMR loops of the SDT junctions with the thickness of Co doped layer between NiFe and Al-O layers as 0.0 nm, 1.2 nm, and 3.0 nm.

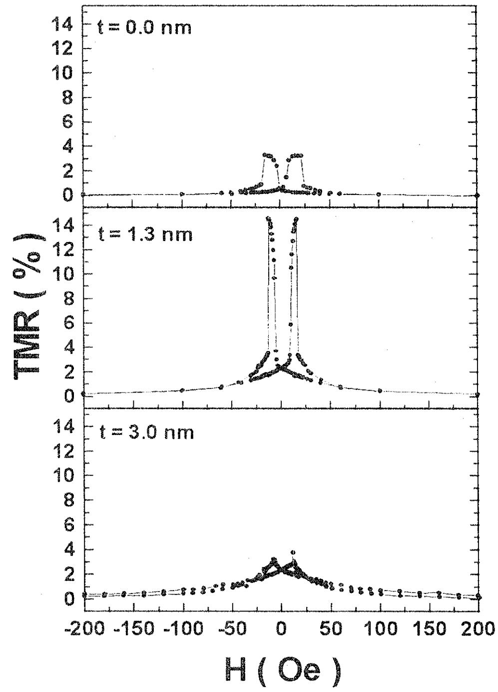

Fig. 1b TMR loops of the SDT junctions with the thickness of CoFe doped layer between NiFe and Al-O layers as 0.0 nm, 1.3 nm, and 3.0 nm.

growth of Al films as a function of the film thickness. Five samples with the thickness of the Al layer as 67.4, 90, 150, 200, and 300 A have been studied systematically. The variation of Al thickness was calculated from the

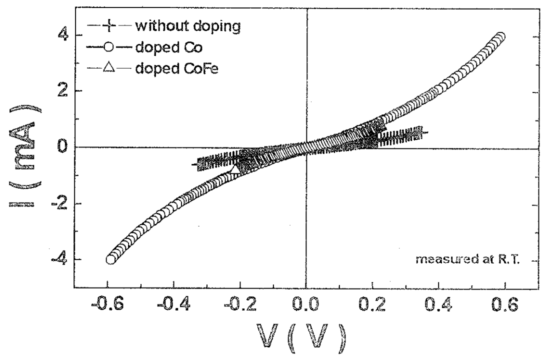

Fig. 2 I-V curves for all the SDT junctions.

sheet resistance of Al. In general, before oxidiation, the resistance of the films increased approximately from 1 to 20 Ω for films with thickness varied from 67.4 to 300 A as shown in the top part of Fig. 3. Under the pressure of $5x10^{-2}$ torr for 50% of Ar + 50% of O_2 environment, samples start to oxidize naturally, and the resistance increases abruptly within a few seconds. If we keep the same pressure condition, the resistance is not changed, which means that oxidation is saturated. After fitting these experimental data to the relation of the resistance as a function of the thickness of Al for all the samples, as shown in Fig. 3, the oxidation layer for all the Al films is approximately 6 A. An oxygen stable samples after natural oxidization could be oxidized again by plasma oxidization process. As shown in Fig. 3, only the data of Al films with thickness of 67.4 and 90 A were used to fit the relation. Because the change of their resistance to

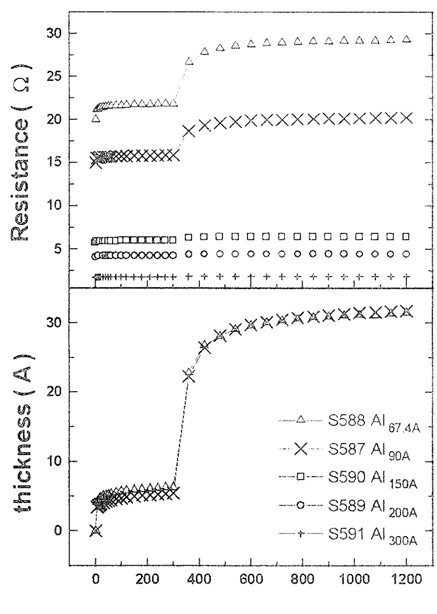

Fig. 3 The variation of the resistance and Al thickness of the oxidation as function of time for Al films with thickness of 67.4, 90, 150, 200, and 300 A.

the oxidation condition is sensitive to this analysis. The plasma oxidization starts roughly after 5 minutes of the natural oxidation and under the same pressure and gas conditions. We observed that the resistance increases again. This means oxidation continues for the Al films. In general, the Al thickness drops abruptly by ~ 4 times compared to that by natural oxidization. The total thickness of the oxidation of the Al films is ~ 30 A.

Fig. 4 shows the TEM picture of a cross section of an undoped sample on a Si substrate with a thin SiO_2 buffer layer. A Si substrate was used instead of a glass

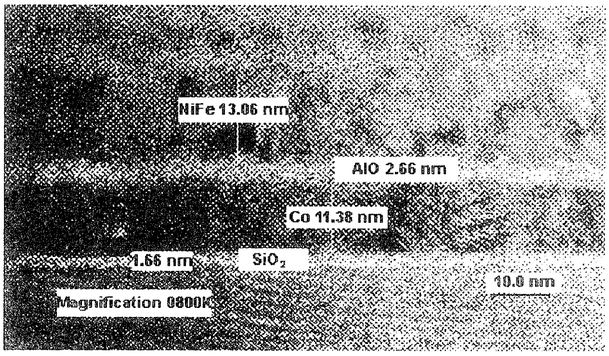

Fig. 4 TEM cross-section picture of a NiFe/Al-O/Co spin-dependent junction.

substrate to fabricate ample preparation. The thickness of the layers for NiFe, Al-O, Co, and SiO_2 are 13.66, 2.66, 11.38, and 1.66 nm, respectively (resolution is ±0.02 nm). The interfacial roughness of all the layers is (≤ 1 nm) quite small, if we compare it with the thickness of the individual layers. The Al-O layer was formed under both natural and plasma oxidization. The thickness of Al-O is quite uniform. Both TEM and AFM observations clearly show the good quality of the junctions.

The TMR ratio measured as a function of t shows a maximum for SDT junctions, glass substrate // Co (10 nm) / Al-O (2.6 nm) /Co or CoFe (t nm) / NiFe (10 nm), with t between 0.0 and 3.0 nm are shown in Fig. 5. For thinner samples with t less than approximately 0.8 nm (region I in Fig. 5), the TMR increases with the thickness of the inserted Co or CoFe layer. This indicates that the spin-filtering ability of the inserted Co or CoFe layer increases with increasing its thickness from 0 to ~0.8 nm. The polarization of transmited electrons at the ferromagnet-insulator interface should be enhanced by a kind of spin filtering effect due to the existence of the inserted Co or CoFe layer. Upadhyay et al. [13] reported that the polarization of Co increases rapidly from 0 to 38 % with increasing the thickness of Co from 0 to 1 nm by fitting the spin up and down transmission coefficients of the transport experiment of superconductor (Pb)/ ferromagnetic material (Co)/normal metal(Cu). In region II, the spin filtering effect is dominated for SDT junction. This is because the inserted Co or CoFe layer is thick enough to play the spin-filtering mechanism.

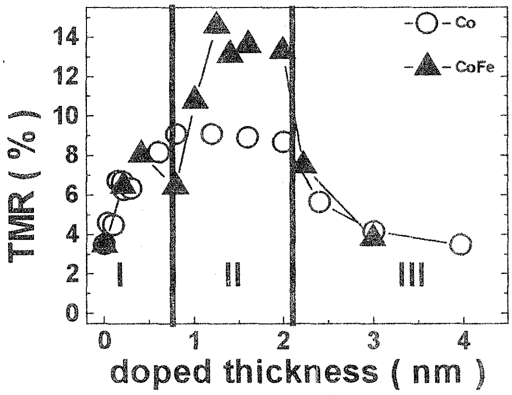

Fig. 5 The variation of the TMR as a function of the thickness of doped Co and CoFe layers.

The maximum TMR is reached 9 and 14 % for SDT junctions with 0.8 ~ 2 nm thickness of inserted Co and CoFe, respectively. This indicates that the spin filtering effect is larger for inserting CoFe than that of Co. In region III, the thickness of the inserted Co or CoFe layer is > 2.0 nm, which is thick enough to dominate the magnetic behavior of the top magnetic electrode in FM/Insulator/FM SDT junction. This can be demonstrated from the variation of the coercivity of the top and bottom magnetic layers. We have observed that the difference of the coercivity between the top and bottom magnetic layers decreases with the thickness of the inserted Co or CoFe layer. In other words, the anti-ferromagnetic interaction between the top NiFe and the bottom Co layers separated by a thin Al-O insulator with thickness < 3.0 nm is reduced by increasing the thickness of the inserted Co or CoFe layer. This can be also related to the shrinking of the plateau of the maximum TMR region as shown in Fig. 1. The plateau of the maximum TMR is ~ 20 Oe for sample with t = 0.0 nm, however, it is approximately zero for samples with t = 3.0 nm. The easily and hardly magnetized spins exist in both top and bottom electrodes. In the region with thicker inserted Co or CoFe layer, the switching magnetic field of easily magnetized spins of two ferromagnetic electrodes are almost the same. The hardly magnetized spins can be explained in the orange peel model by the presence of roughness of ferromagnet-insulator interface [14]. In region III, the formation of a perfect antiparallel magnetic alignment of the top and bottom electrodes is weaker than that in region II, and only the rotation of hardly magnetized spins contributes to tunneling magnetoresistance.

In summary, from the oxidation process study of the Al-O insulator layer and an extra magnetic layer in the interface of the Al-O and NiFe layers in Co/Al-O/NiFe system, we have shown experimentally that the optimal condition for the oxidation of the Al-O insulator layer in a spin dependent tunnel system and the enhanced spin polarization in SDT junctions by an extra Co or CoFe layer. The polarization of tunneling electrons can be varied by properly modifying the interfacial condition between the insulating and magnetic layers; and we have observed that this effect is greater for inserting CoFe layer than that of inserting Co layer.

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