230% room-temperature magnetoresistance in CoFeB/MgO/CoFeB magnetic tunnel junctions

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Magnetoresistance (MR) ratio up to 230% at room temperature (294% at 20 K) has been observed in spin-valve-type magnetic tunnel junctions (MTJs) using MgO tunnel barrier layer fabricated on thermally oxidized Si substrates. We found that such a high MR ratio can be obtained when the MgO barrier layer was sandwiched with amorphous CoFeB ferromagnetic electrodes. Microstructure analysis revealed that the MgO layer with (001) fiber texture was realized when the MgO layer was grown on amorphous CoFeB rather than on polycrystalline CoFe. Since there have been no theoretical studies on the MTJs with a crystalline tunnel barrier and amorphous electrodes, the detailed mechanism of the huge tunneling MR effect observed in this study is not clear at the present stage. Nevertheless, the present work is of paramount importance in realizing high-density magnetoresistive random access memory and read head for ultra high-density hard-disk drives into practical use. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1871344]

Tunneling magnetoresistance (TMR) effect in magnetic tunnel junctions (MTJs) has been extensively studied in the last decade due to the large potential applications of MTJs in magnetoresistive random-access memory (MRAM) and magnetic sensors.[1,2] MTJs using amorphous Al-oxide tunnel barrier layer have been the most frequently studied. Using this barrier material, a magnetoresistance (MR) ratio up to about 70% has been achieved at room temperature (RT).[3,4] Here, the MR ratio is defined as $(R_{ap}-R_p)/R_p$, where R_{ap} and R_p are the tunneling resistance when the magnetizations of the two electrodes are aligned in antiparallel and parallel, respectively. The MR ratio in such conventional MTJs with an Al–O tunnel barrier can be relatively well explained by the Julliere's model[5] when experimentally observed spin polarizations are used.

Recently, *ab initio* calculations predicted an extremely large MR ratio in Fe(001)/MgO(001)/Fe(001) single-crystalline MTJs, due to coherent tunneling.[6,7] When the coherency of electron wave functions is conserved during tunneling, only conduction electrons whose wave functions are totally symmetrical with respect to the barrier normal axis are connected to the electronic states in the barrier region and have significant tunneling probability. In the case of Fe(001), the Δ_1 band in the Fe(001) electrode has totally symmetrical characteristics with the majority spin Δ_1 subband crossing the Fermi energy E_F, whereas the minority spin Δ_1 band has no states at the E_F. Furthermore, barriers, such as MgO(001), promote efficient bonding and tunneling transmission of Δ_1 states, compared to other electronic symmetries. While electrons of all symmetries approach the Fe/MgO interface with unity density of states, evanescent wave function coupling introduces discrimination, as the density of Δ_1 states is much less affected within the barrier. This results in a R_{ap} much higher than R_p, leading to a huge MR ratio.[6] Several experimental studies have been performed in epitaxial Fe(001)/MgO(001)/Fe(001) MTJs.[8–12] The MR ratio was found to surpass the value obtained by the conventional MTJs (Ref. 11) and has reached up to 188% at RT.[12] However, all of these studies have been performed using single-crystal substrates, which limits its feasibility into practical use. Recently, 220% of MR ratio at RT has been presented in MTJs of the form $FeCo(001)/MgO(001)/(Fe_{70}Co_{30})_{80}B_{20}$.[13] In this case, the FeCo layer is a polycrystalline body centered cubic with a (001) texture. In this letter, we report an even higher MR ratio of 230% at room temperature achieved in spin-valve type magnetic tunnel junctions using a MgO barrier layer sandwiched with amorphous CoFeB ferromagnetic electrodes. The amorphous CoFeB electrodes are of great advantage to the polycrystalline FeCo electrodes in achieving a high homogeneity in 100 nm-sized small MTJs.

Thin films for MTJs were deposited onto thermally oxidized Si wafer (100) using magnetron sputtering system (ANELVA C7100) with base pressure better than 5×10^{-9} Torr. To obtain flat surface morphology, the gas pressure during sputtering was kept low at about $1.5–2.3 \times 10^{-4}$ Torr. The MTJ stack fundamental structure was substrate/Ta(10)/PtMn(15)/$Co_{70}Fe_{30}$(2.5)/Ru(0.85)/reference layer/MgO(1.8)/free layer/Ta(10)/Ru(7)(in nm). Two kinds of reference and free layer combinations were examined; CoFeB reference and free layers (each 3 nm), and $Co_{70}Fe_{30}$(2.5 nm) reference and $Co_{70}Fe_{30}$(1.5 nm)/$Ni_{83}Fe_{17}$(3 nm) free layers. CoFeB was deposited using $Co_{60}Fe_{20}B_{20}$ (at. %) alloy target. All the metal films were deposited by dc sputtering, while the MgO layer was depos-
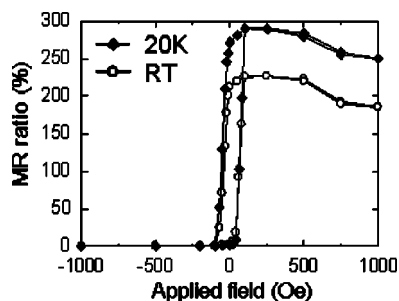
FIG. 1. MR curves of CoFeB/MgO/CoFeB MTJs evaluated at RT and 20 K.

FIG. 3. Cross-section HRTEM image of CoFe/MgO/CoFe/NiFe MTJs.

ited using a rf sputtering method. The MTJs were annealed in a high vacuum furnace chamber at 360 °C for 2 h in a magnetic field of 8 kOe. Cross-sectional high-resolution transmission electron microscopy (HRTEM) analysis was done to observe the film microstructure. The magnetotransport properties were evaluated by a dc four-point probe method in patterned MTJ with a junction size of 1 μm $\times$ 1 μm fabricated by photolithography and ion-milling method.

Figure 1 shows the MR curves of CoFeB/MgO/CoFeB MTJs at RT and 20 K. The MR ratio and resistance area product R_pA were, respectively, 230% and 420 $\Omega\mu$m^2 at RT and 294% and 440 $\Omega\mu$m^2 at 20 K. This is a rather high room-temperature TMR effect. On the other hand, the CoFe/MgO/CoFe/NiFe MTJs showed only a low MR ratio of about 62% at RT.

To understand the origin of the difference of MR ratio in those MTJs, structural analysis has been performed. Figure 2(a) shows a HRTEM image of the MTJs using CoFeB as the ferromagnetic electrodes. From Fig. 2(a), we can see that the morphology of the MTJs was very smooth and that the interfaces between the CoFeB and MgO layer were very sharp. The enlarged HRTEM image of the CoFeB/MgO/CoFeB layers is shown in Fig. 2(b). We found that the CoFeB layers were essentially amorphous, although some parts of the CoFeB layers were crystallized. The partial crystallization may have been caused by the high annealing temperature. Surprisingly, the MgO layer, grown on the amorphous
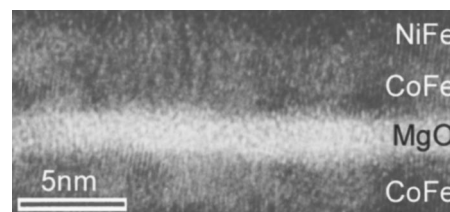
CoFeB layer, showed a good crystalline structure with a (001) fiber texture. The lattice image of MgO(001) can be clearly identified from the lower interface to the upper interface, indicating that the MgO(001) texture developed from the initial stage of growth, i.e., the nucleation stage. Note that MgO layer grown on amorphous Si$_3$N$_4$ substrates by ion beam assisted deposition is also (001) textured right at the inception of the deposition.[14]

Figure 3 shows a typical cross-sectional HRTEM of CoFe/MgO/CoFe/NiFe MTJs. Contrary to the MTJs with CoFeB electrodes, the interfaces between the MgO and CoFe layers looks rough. The CoFe layers showed a polycrystalline structure, although the lattice image was not clearly observed in some parts of the layers. The MgO layer grown on CoFe showed indistinct crystal structure. The poor epitaxial growth of MgO on CoFe may have been due to the difference of fiber texture for both layers. On the other hand, the fact that MgO with good crystallinity and (001) preferred orientation can be grown on amorphous CoFeB is considered to be due to the amorphous nature of the CoFeB layer, which may eliminate the lattice mismatch issue, and (001) fiber texture is the preferred orientation growth for MgO (a rock-salt structure).[15]

Although previous theoretical studies only consider coherent spin-polarized tunneling in fully epitaxial Fe(001)/MgO(001)/Fe(001) MTJs, our current result shows that a very high MR ratio could also be achieved when the MgO(100) barrier layer was sandwiched between amorphous electrodes. This implies that the correct symmetry of the MgO(001) barrier may be an essential factor to obtain a high MR ratio. However, there is also a probability that the high MR ratio is attributed to the smooth and sharp interfaces between the CoFeB and MgO. It should also be noted that at the present stage, we can not exclude a possibility of local crystallization of a few monolayers of CoFeB at the electrode/barrier interfaces, which could not be detected in the HRTEM observation. The crystalline MgO(001) layer might act as a template to locally crystallize the CoFeB electrodes. Note also that when the amorphous CoFeB electrodes were combined with an amorphous Al–O tunnel barrier, the MTJs exhibited the MR ratio of 108% at 20 K,[4] showing that the spin polarization of CoFeB was about 0.6, if we apply the Julliere's model.[5] The CoFeB electrodes combined with the MgO(001) tunnel barrier exhibited the MR ratio of 300%, indicating the spin polarization of about 0.8. The CoFeB/MgO(001) interface seems to enhance the spin polarization of evanescent electronic states. Since there have been no theoretical studies on the MTJs with a crystalline tunnel barrier and amorphous electrodes, the detailed mechanism of the huge TMR effect observed in this study is not clear at the present stage.

In Fig. 4, the bias voltage dependence of normalized MR ratio at RT in CoFeB/MgO/CoFeB MTJs is shown. Slightly
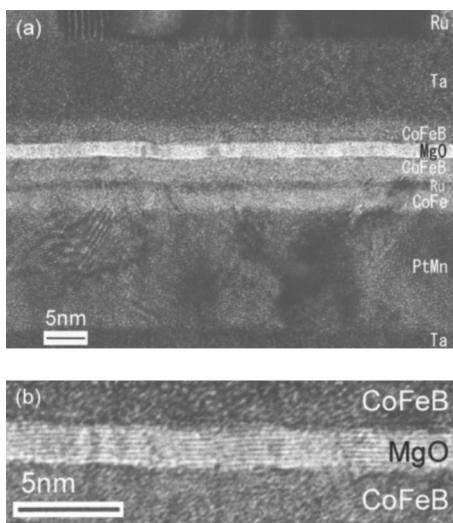

FIG. 2. (a) Cross-section HRTEM image of CoFeB/MgO/CoFeB MTJs. (b) Enlarged cross-section HRTEM image of the MTJs shown in (a). Only the CoFeB/MgO/CoFeB layers are shown.
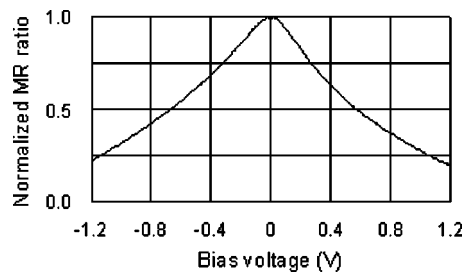
FIG. 4. Bias voltage dependence of normalized MR ratio evaluated at RT in CoFeB/MgO/CoFeB MTJs.

asymmetric bias-dependence curve was observed, despite the symmetric CoFeB/MgO/CoFeB films structure. This is probably due to a slight difference of the reference and free CoFeB/MgO interfaces. Note that the asymmetric bias dependence in our sample was much less than the fully epitaxial Fe(001)/MgO(001)/Fe(001) MTJs reported earlier.[11] The bias voltage at which the MR ratio reaches one-half of the zero-bias value V_{half} was 680 and 590 mV for negative and positive bias voltage, respectively. From V_{half} we can determine the output voltage $V_{out}(\equiv V_{half} \times (R_{ap} - R_p)/R_{ap})$, which was about 380 mV and 330 mV for the negative and positive bias, respectively. This value is comparable with the fully epitaxial Fe(001)/MgO(001)/Fe(001) MTJs (Ref. 11) and high enough to realize high-density MRAM.

In conclusion, we have fabricated polycrystalline spin-valve-type MTJs using the MgO barrier layer sandwiched with amorphous CoFeB ferromagnetic electrodes which shows a very high MR ratio of 230% and V_{out} of 380 mV at RT. The detailed mechanism of the huge TMR effect observed in this study is not clear at the present stage. The present work is of paramount importance in realizing high-density MRAM and read-head for ultra high-density hard-disk drives into practical use.

[1] T. Miyazaki and N. Tezuka, J. Magn. Magn. Mater. **139**, L231 (1995).
[2] J. S. Moodera, L. R. Kinder, T. M. Wong, and R. Meservey, Phys. Rev. Lett. **74**, 3273 (1995).
[3] D. Wang, C. Nordman, J. Daughton, Z. Qian, and J. Fink, IEEE Trans. Magn. **40**, 2269 (2004).
[4] K. Tsunekawa, D. D. Djayaprawira, H. Maehara, Y. Nagamine, M. Nagai, and N. Watanabe, *Proceedings of the Ninth Joint MMM-Intermag Conference*, Anaheim, CA, January 6, 2004, Abstract BD-03.
[5] M. Jullière, Phys. Lett. **54A**, 225 (1975).
[6] W. H. Butler, X.-G. Zhang, T. C. Schulthess, and J. M. MacLaren, Phys. Rev. B **63**, 054416 (2001).
[7] J. Mathon and A. Umersky, Phys. Rev. B **63**, 220403R (2001).
[8] W. Wufhenkel, M. Klaua, D. Ùllmann, F. Zavaliche, J. Kirschner, R. Urban, T. Monchesky, and B. Heinrich, Appl. Phys. Lett. **78**, 509 (2001).
[9] M. Bowen, V. Cros, F. Petroff, A. Fert, C. M. Boubeta, J. L. Costa-Krämer, J. V. Anguita, A. Cebollada, F. Briones, J. M. de Teresa, L. Morellon, M. R. Ibarra, F. Güell, F. Peiro, and A. Cornet, Appl. Phys. Lett. **79**, 1655 (2001).
[10] J. Faure-Vincent, C. Tiusan, E. Jouguelet, F. Canet, M. Sajieddine, C. Bellouard, E. Popova, M. Hehn, F. Montaigne, and A. Schuhl, Appl. Phys. Lett. **82**, 4507 (2003).
[11] S. Yuasa, A. Fukushima, T. Nagahama, K. Ando, and Y. Suzuki, Jpn. J. Appl. Phys., Part 2 **43-4B**, L588 (2004).
[12] S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, and K. Ando, Nat. Mater. **3**, 868 (2004).
[13] S. S. P. Parkin, C. Kaiser, A. Panchula, P. M. Rice, B. Hughes, M. Samant, and S.-H. Yang, Nat. Mater. **3**, 862 (2004).
[14] C. P. Wang, K. B. Do, M. R. Beasley, T. H. Geballe, and R. H. Hammond, Appl. Phys. Lett. **71**, 2955 (1997).
[15] E. A. Colbourn, Surf. Sci. Rep. **15**, 281 (1992).