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The effect of magnetic annealing on barrier asymmetry in $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}/\text{MgO}$ magnetic tunnel junctions

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ABSTRACT

We demonstrate that high quality magnetic tunnel junctions with $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ electrodes and crystalline MgO barriers, which show a 230% tunnelling magnetoresistance ratio at room temperature, can be successfully fabricated by using a target-facing-target sputtering technique and present the results on the change of barrier asymmetry after the annealing at different temperatures. Bias voltage dependence of magnetoresistance ratio for $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}/\text{MgO}$ magnetic tunnel junctions is highly asymmetrical in the as-deposited states and becomes more symmetrical after magnetic annealing. It also exhibits a shift of the tunneling magnetoresistance maximum to a positive bias voltage under the low temperature annealing.

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1. Introduction

Since the significant TMR values were reported in 1995^[1–2], magnetic tunnel junctions (MTJs) emerged as a promising component for magnetic sensors and magnetic random access memory (MRAM). The application of MgO tunnel barriers in magnetic tunnel junctions has recently enhanced tunnel magnetoresistance (TMR) values to more than 200% at room temperature^[3–5]. With its high magnetic field sensitivity, good MTJs with MgO barriers are suitable MTJ applications. The high TMR in these devices is due to the spin filter effect of the crystalline MgO barrier. The barrier is relatively transparent for the majority spin electrons injected from an oriented bcc Fe or Fe-Co electrode but attenuates the minority spin electrons, as result of the different symmetry of the $\uparrow$ and $\downarrow$ wavefunctions^[6–7].

Usually two steps are essential in order to obtain high TMR ratio for $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}/\text{MgO}$ MTJs, the formation of crystallized MgO barrier and post magnetic annealing. Molecular beam epitaxy is the most effective way to fabricate MTJs with crystalline barrier. S. Yuasa et al has reported 180 % TMR in full epitaxial Fe (001) / MgO (001) / Fe (001) MTJs^[3]. However it is a challenge to produce highly oriented (001) MgO barrier just by sputtering. In 2004, Stuart Parkin's group from IBM^[4] demonstrated high TMR in sputtered MgO based MTJs for the first time by preparing crystallized MgO barrier using reactive magnetron sputtering⁴. Shortly after that, the Japanese company Anelva^[5] introduces another commercial solution to produce high quality MgO-based MTJs. In this technique, $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}$ produced by conventional DC sputtering is used as the ferromagnetic (FM) layer, and the MgO barrier is grown directly by RF sputtering from an MgO target. The sputtering system has a unique feature of large distance and offset configuration between substrates and targets. Furthermore, compared with conventional sputtering system, the sputtering gas pressure can be reduced by one order of magnitude in

their sputtering tools. By using this sputtering technique, a highly oriented thin (001) MgO layer can be grown on the top of the amorphous ferromagnetic layer. Here we report the successful fabrication of high TMR $\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}/\text{MgO}$ MTJs by using the target-facing-target (TFT) sputtering techniques and the change of barrier asymmetry after the annealing at different temperatures.

2. Experiment

Single barrier MTJs were grown by magnetron sputtering (DC and RF) on thermal-oxidized Si substrates in a Shamrock deposition system (base pressure around 3×10^{-7} Torr), which consists of two sputtering chambers. All the metal layers are deposited in the main sputtering chamber and the MgO barrier layer was deposited by RF sputtering from a target-facing-target (TFT) gun. In order to establish exchange bias, the wafers were placed in an in-plane magnetic field of 5 mT during the metal deposition. Junctions with the size of $100 \times 100 \mu\text{m}^2$ to $20 \times 20 \mu\text{m}^2$ were fabricated by UV-lithography and Ar ion milling techniques. The annealing process was performed in a vacuum furnace under a magnetic field of 800 mT. The typical temperature ramp-up rate and dwell time were $10^\circ\text{C}/\text{min}$ and 90 minutes. X-ray diffraction measurement was conducted for structural characterization by using a Philips analytical X-pert Pro diffractometer with Cu $K\alpha$ radiation. The magnetic property was measured by alternating gradient force magnetometer (AGM). The transport properties were measured by a standard four-point method.

3. Results and discussions

The room temperature magnetization and magneto-transport data for the as-deposited and annealed $\text{MgO}/\text{Co}_{40}\text{Fe}_{40}\text{B}_{20}/\text{CoFeB}$ MTJs

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are shown in Figure 1. For the as-deposited MTJs (Fig. 1 (a) and (c)), the switching of the pinned layer and free layers is not well defined due to the weak exchange bias after sputtering. The exchange field (H_{ex}) is only 23 mT and coercivity (H_c) of the free layer is 7 mT. The small difference between H_{ex} and H_c for the as-deposited MTJs leads to simultaneous magnetization reversal of the free and pinned ferromagnetic layers. This means that a full antiparallel configuration between free and pinned layers can not be realized before annealing, which results a small TMR ratio of only 18%. A huge increase of the TMR ratio from 18% to 230% can be achieved after annealing at 350°C for 1 hour under an 800 mT field, as shown in Fig. 1 (b). The H_c for the free layer is around 1 mT. A large H_{ex} around 40 mT was established after the annealing process. Therefore a well-defined and stable anti-parallel configuration between free and pinned layers was realized from 40 mT to 0 mT (Fig. 1 (d)).

The annealing temperature effect on the TMR ratio, exchange bias (H_{ex}) and coercivity of the free layer (H_c) are summarized in Fig. 2. By increasing annealing temperature (T_a) up to 375°C, the TMR ratio increases linearly from few percent in the as-deposited state to 230%. The TMR begins to drop at 400°C. This is mainly thought to be due to the diffusion of Mn and Ru into the MgO barrier^[8]. Also as we can see, H_{ex} decreases with the increasing annealing temperature, but the H_c of CoFeB layer increases. At low T_a , for example 200°C, the annealed samples show a H_{ex} of 103 mT and H_c of 0.77 mT. After annealing at 375°C, although H_{ex} decreases to 17 mT and H_c increases to 3 mT, the bottom pinned and top free CoFeB can still switch independently following the change of magnetic field. A H_{ex} of 5 mT and H_c of 3.5 mT can be seen for the samples annealed at 400°C. The small difference between those two values results in simultaneous magnetization reversal of pinned and free layer. No perfect anti-parallel configuration can be obtained in this case. This could be another reason for the drop of TMR ratio at high annealing temperature.

Fig. 1. The room temperature magnetoresistance and magnetization loop for as-deposited (a), (c) and annealed (b), (d) MgO-based magnetic tunnel junctions.

Fig. 2. Annealing temperature dependence of TMR ratio, Exchange bias field (H_{ex}) and free layer coercivity (H_c).

Fig. 3 (a) shows the normalized TMR ratio as a function of applied bias voltage for a 325°C annealed sample. The TMR ratio decreases with the increasing negative / positive bias voltage and dropped to half its value at 600 mV at negative bias ($-V_{1/2}$) and about 900 mV at positive bias ($+V_{1/2}$). A relative large $\Delta V_{1/2}$ of 300 mV, which is defined as the difference between $-V_{1/2}$ and $+V_{1/2}$, indicates a highly asymmetrical bias dependence for the samples annealed at this temperature. At the same time, the maximum value of the TMR wasn't obtained at zero bias but at +15 mV (as shown in the inset).

The asymmetric TMR bias dependence can be explained by considering the barrier asymmetry $\Delta\phi$, which is defined as the difference in barrier height between the top and bottom electrodes: $\Delta\phi = \phi_{top} - \phi_{bottom}$. Fig. 3 (b) shows a simple energy diagram for the MgO-based MTJs based on model developed by Zhang^[9]. The maximum value of the TMR can be obtained under an applied bias voltage of $\Delta\phi$ eV, where the barrier profile has a symmetrical shape. The asymmetric bias dependence in MgO-based MTJs has been reported in many papers. In epitaxial Fe(001)/MgO(001)/Fe(001) single crystal MTJs, this phenomena was attributed to dislocations at the interfaces and lattice distortions^[10]. In the case of CoFeB/MgO MTJs, it was believed that different structure at the top and bottom CoFeB/MgO interfaces also plays an important role. The annealing process, which can crystallize CoFeB layers and equalize the interface on both sides of MgO is supposed to promote more symmetric behavior.

Fig. 3. (a) Normalized TMR ratio as a function of bias voltage for a 325°C annealed sample (b) Energy diagram for the MgO-based MTJs with an asymmetrical barrier.

The bias dependence of the TMR ratio curves for the SMTJs annealed at different temperature have been put together in Fig. 4 (a) and (b). A clear change from asymmetric bias dependence to a more symmetric one can be observed with increasing annealing temperature. The samples annealed at 200°C show a $\Delta V_{1/2}$ of 620 mV (with a $-V_{1/2} = 480$ mV and $\Delta V_{1/2} = 1100$ mV). The value of $\Delta V_{1/2}$ decreases to 160 mV after annealing at 350°C. The bias dependence curve becomes more symmetric. Figure 4 (b) is compilation of bias dependence curves in the low bias region. The shifts of maximum MR (V_{bias}) were 42 mV and 0 mV for samples annealed at 200°C and 350°C respectively.

Fig. 4. Normalized TMR ratio as a function of bias voltage for sample annealed at different temperatures (a) high bias region (b) low bias region

The shift of maximum TMR (V_{bias}) has been reported in single barrier AlO_x -based MTJs, which is explained in two different ways, but not in the case of MgO-based MTJs. Sato *et al* [11] consider Co-Al interdiffusion and a change in band structure after annealing as a possible cause; Oepets *et al* [12] believe the barrier asymmetry $\Delta\phi$ can be the reason for the asymmetric bias dependence and shift of maximum MR according to their observations. The calculations of the later group clearly showed the change from asymmetric to symmetric bias with decreasing $\Delta\phi$. They also assumed the change of the barrier height ($\Delta\phi$) was related to oxidation time in the preparation of the AlO_x barrier.

Fig. 5. Demonstration of the asymmetric bias dependence changing with the value of $\Delta\phi$ based on Oepets' calculations.

Fig. 5 is a demonstration of the asymmetric bias dependence changing with the value of $\Delta\phi$ based on Oepets' calculations. Of

course, the results should be interpreted only as an indication of the effect of the asymmetry of potential barrier on asymmetry of the TMR versus bias voltage in our MgO-based MTJs. The changing of the bias dependence curves with decreasing $\bar{A}\phi$ is very similar to its evolution with the increasing annealing temperature. So it is reasonable to speculate that the shift of maximum TMR (V_{bias}) could be a result of the change of $\bar{A}\phi$ during the annealing process. A large difference of barrier height between the bottom and top electrodes ($\bar{A}\phi$) for as-deposited or low-temperature annealed samples will result in a shift of the maximum TMR (V_{bias}) to positive bias. With the increasing annealing temperature ($T_1 > T_2 > T_3$ as indicated in Fig. 5), $\bar{A}\phi$ decreases. The maximum TMR (V_{bias}) will shift to zero bias as the bias dependence curves become more symmetric.

4. Conclusion

We observed the symmetry change of the bias dependence and the shift of maximum TMR in high TMR MgO/CoFeB MTJs during the annealing process. The junctions annealed at 200°C show a large $\bar{A}V_{1/2}$ of 620 mV. This value decreases to 160 mV after annealing at 350°C. The shifts of maximum MR (V_{bias}) also decrease from 42 mV to 0 mV when the annealing temperature increases from 200°C and 350°C respectively. The change of asymmetric bias dependence and shift of maximum MR can be attributed to the barrier asymmetry ($\bar{A}\phi$) change during the annealing process based on Opets' calculations.

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References

- [1] J.S. Moodera, L.R. Kinder, T.M. Wang, R. Merservey, Phys. Rev. Lett. 74 (1995) 3273.
- [2] T. Miyazaki, N. Tezuka, J. Magn. Magn. Mater. 139 (1995) L231.
- [3] S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki, and K. Ando, Nat. Mater. 3, (2004) 868.
- [4] S. S. P. Parkin, C. Kaiser, A. Panchula, P.M. Rice, B. Hughes, M. Samant, and S-H Yang, Nat. Mater. 3 (2004) 862.
- [5] D. D. Jayaprawira, K. Tsunekawa, M. Nagai, H. Maehara, S. Yuasa, Y. Suzuki, and K. Ando, Appl. Phys. Lett. 86 (2005) 092502.
- [6] W.H. Butler, X.-G. Zhang, T.C. Schulthess, and J.M. MacLaren, Phys. Rev. B 63 (2001) 054416.
- [7] J. Mathon and A. Umerski, Phys. Rev. B 63 (2001) 220403.
- [8] X. Y. Liu, D. Mazumdar, W. F. Shen, B. D. Schrag, and G. Xiao, Appl. Phys. Lett. 89 (2006) 023504.
- [9] X. Zhang, B. Z. Li, G. Sun, and F. C. Pu, Phys. Rev. B 56 (1997) 5484.
- [10] T. Noizaki et al., Phys. Rev. Lett, 96 (2006) 027208.
- [11] M. Sato, H. Kikuchi, and K. Kobayashi, IEEE Trans. Magn. 35 (1999) 2946.
- [12] W. Opets, M. F. Gillies, R. Coehoorn, R. J. M. van de Veerdonk, and W. J. M. de Jonge, J. Appl. Phys. 89 (2001) 8038.