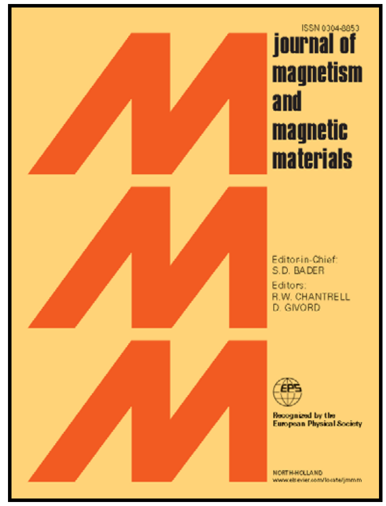
Author's Accepted Manuscript



www.elsevier.com/locate/jmmm

Annealing effect on tunneling magnetoresistance in MgO-based magnetic tunnel junctions with FeMn exchange-bias layer

Q.L. Ma, J.F. Feng, Gen Feng, K. Oguz, X.F. Han, J.M.D. Coey

PII:	S0304-8853(09)00876-2
DOI:	doi:10.1016/j.jmmm.2009.08.038
Reference:	MAGMA 55808

To appear in: *Journal of Magnetism and Magnetic Materials*

Received date:	15 July 2009
Revised date:	24 August 2009
Accepted date:	26 August 2009

Cite this article as: Q.L. Ma, J.F. Feng, Gen Feng, K. Oguz, X.F. Han and J.M.D. Coey, Annealing effect on tunneling magnetoresistance in MgO-based magnetic tunnel junctions with FeMn exchange-bias layer, *Journal of Magnetism and Magnetic Materials*, doi:10.1016/j.jmmm.2009.08.038

Journal of Magnetism and Magnetic Materials 00 (2009) 000–000

Journal of magnetism and magnetic materials

www.elsevier.com/locate/jmmm

Annealing effect on tunneling magnetoresistance in MgO-based magnetic tunnel junctions with FeMn exchange-bias layer

Q. L. Ma[a,b], J. F. Feng[a,b,*], Gen Feng[a], K. Oguz[a], X. F. Han[b], and J. M. D. Coey[a]

[a]School of Physics and CRANN, Trinity College, Dublin 2, Ireland

[b]Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, 100190 Beijing, China

Elsevier use only: Received date here; revised date here; accepted date here

Abstract

MgO-based magnetic tunnel junctions were fabricated, with a thin pinned CoFeB layer in the unbalanced synthetic antiferromagnet part of the stack FeMn/CoFe/Ru/CoFeB. Inverted and normal tunneling magnetoresistance (TMR) values occur at low and high annealing temperatures (T_a), respectively. The TMR ratio remains inverted up to T_a = 300 °C and it becomes normal around T_a = 350 °C. The exchange bias of FeMn disappears at high T_a. The sign reversal of the TMR ratio is mainly attributed to the disappearance of the exchange bias due to manganese diffusion during the annealing process.

PACS: 73.40.Rw; 73.40.Gk; 72.25.Mk

Keywords: Magnetic tunnel junctions (MTJs); Inverted and normal tunneling magnetoresistance (TMR); Elemental diffusion

1. Introduction

The prediction [1], [2] and recent observation [3] of tunnel magnetoresistance (TMR) ratios [$(R_{AP}-R_P)/R_P$] as high as 1000% in magnetic tunnel junctions (MTJs) with an MgO barrier have transformed the prospects for spin electronic devices. The resistances (R_P and R_{AP}) of an MTJ depend on the relative magnetization of the two ferromagnetic layers adjacent to the barrier: the resistance is usually low in the parallel state (P) and high in the antiparallel state (AP), giving a positive TMR ratio, while in the opposite case the TMR is negative. In these MTJ devices, antiferromagnetic materials with a high blocking temperature and large exchange-bias, such as IrMn [4] and PtMn [5], are generally used to pin one of the ferromagnetic layers. Otherwise, when parallel and antiparallel configurations are achieved by relying on different coercivities of the two ferromagnetic layers, the structure is known as pseudo-spin-valve [3], [6]. Exceptionally high TMR in some MgO-based MTJs is attributed to the possibility of annealing them at high temperature without a risk of diffusion of Mn from an antiferromagnetic pinned layer.

In this work, we use FeMn as the antiferromagnet in exchange-biased MgO-based MTJs. A pinned CoFeB layer is next to the MgO barrier in the synthetic antiferromagnetic (SAF) CoFe/Ru/CoFeB part of the stack. A sign reversal of the TMR ratio was observed in these MTJs when the pinned CoFeB layer is thin. The TMR ratio varies from inverted to normal with annealing temperature (T_a). The blocking temperature of FeMn is low [7] compared to IrMn and PtMn, and it is found that the elemental diffusion is much enhanced at high T_a. This diffusion causes the disappearance of the antiferromagnetic effect of FeMn, resulting in a sign change of the TMR. Results are compared with those for MgO-based IrMn-pinned MTJs using the same fabrication process [8], [9].

2. Experiment

FeMn-pinned MTJ (FeMn-MTJ) samples with a complete layer sequence Si/SiO_2(substrate) /Ta(5) /Ru(25) /Ta(5) / $Ni_{81}Fe_{19}$(5) /$Fe_{50}Mn_{50}$(10) /$Co_{90}Fe_{10}$(2) /Ru(0.85) /CoFeB (t) /MgO(2.5) /CoFeB(3) /Ta(5) /Ru(5) was grown by magnetron sputtering (thickness is given in nanometers). Further details on the growth process can be found in Ref. [9]. The thickness (t) of the pinned CoFeB ($Co_{40}Fe_{40}B_{20}$) layer was varied from 0.5 to 3.0 nm. After deposition of the multilayer stacks, micro-scale square-shaped junctions were fabricated using conventional UV lithography. They are usually 12 × 12 and $16 \times 16\ \mu m^2$. Thermal annealing of the patterned junctions was carried out at temperatures ranging from 150 to 400 ºC under vacuum for one hour in an applied magnetic field of 800 mT. All magnetotransport measurements of the patterned MTJs were performed by a four-probe method *near zero bias* (≤ 2 mV). The magnetic properties of unpatterned MTJ samples were measured with a vibrating sample magnetometer (VSM).

3. Results and Discussions

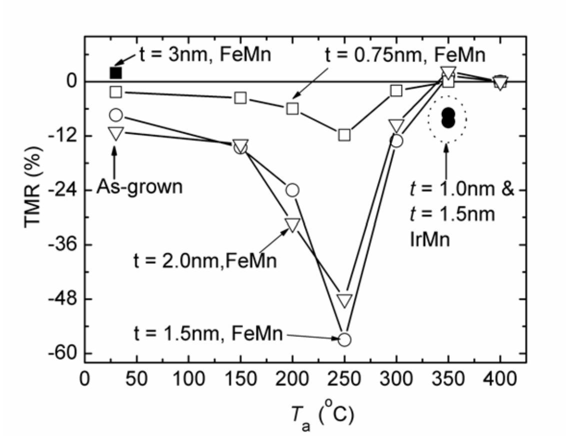


Figure 1 The TMR ratios at 300 K after annealing at various temperatures for FeMn-MTJs with different pinned CoFeB layer thickness t = 0.75, 1.5 and 2.0 nm. The TMR ratios for FeMn-MTJs with t = 3.0 nm in the as-grown state, and for IrMn-MTJs with t = 1.0 and 1.5 nm at T_a = 350 °C are also plotted as solid points [9].

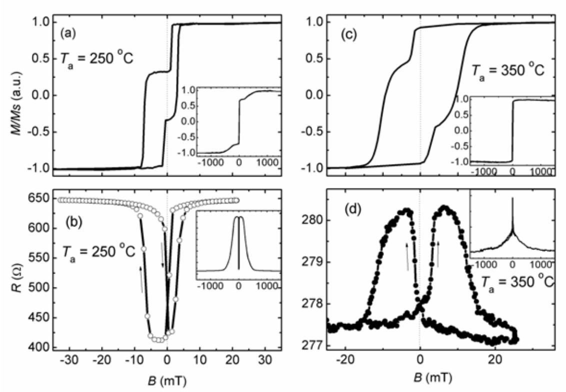


Figure 2 (a) and (b) the *M-H* loops, and (c) and (d) the *R-H* loops with t = 1.5 nm at T_a = 250 °C and 350 °C, measured at low magnetic field by VSM and four-probe technique. The insets show the corresponding data at large magnetic field.

Figure 1 shows the room-temperature TMR ratio as a function of T_a for FeMn- MTJs with t = 0.75, 1.5, and 2.0 nm. A similar T_a dependence of TMR has been observed for all MTJs regardless of the CoFeB layer thickness. To show the junction resistance with magnetic field, the inverted TMR effect is defined as $(R_P - R_{AP})/R_P$, where R_P and R_{AP} correspond to the parallel and antiparallel states between the pinned and free CoFeB layers for these MTJs (also see Fig. 3). From this point of view, the inverted TMR has an opposite sign compared to the positive TMR, which can be seen from the *R-B* loops shown in Fig. 2 (b) and (d). The TMR effect remains inverted up to T_a = 300 ºC, and it becomes normal around T_a = 350 ºC. However, the TMR ratio is zero at T_a = 350 ºC for FeMn-MTJs with t = 0.75 nm. The bottom CoFeB/MgO interface may break down by elemental diffusion at high T_a when the pinned CoFeB layer is very thin [10], [11]. It is clear that the bottom CoFeB/MgO interface is rough for FeMn-MTJs, which enhances the element diffusion at high T_a (see below). No similar results were found for IrMn-MTJs [9]. Furthermore, the TMR effect almost disappears around T_a = 400 ºC for all FeMn-MTJs.

The MgO-based FeMn-MTJs with t = 1.5 nm were selected for detailed study. It is found that the magnetic reversals in the unpatterned *M-H* and patterned *R-H* samples show little difference in this stack. This is similar to the result of another group [12]. It is reasonable that the coercivity (H_c) and the exchange field (H_{ex}) increase somewhat after micro-fabrication process due to the change of shape anisotropy. Besides, the free switching of the magnetization in the free and pinned CoFeB electrodes occurs. The free switching of the magnetization and the squareness of the hysteresis loops create well-defined parallel and antiparallel states.

As shown in Fig. 2, the TMR effect at T_a = 250 ºC is inverted because of a change in switching sequence caused by the imbalance of the synthetic antiferromagnet CoFe/Ru/CoFeB [9]. A return to the low resistance state is observed at large magnetic field. An explanation of the inverted TMR behavior in Fig. 2 (b) is presented in Fig. 3. The high field behavior shown in the inset is due to overcoming the antiferromagnetic coupling across the Ru layer in the SAF stack, in fields in excess of $H_{ex}^{SAF} \approx 300$ mT. In smaller fields, the antiferromagnetic coupling stabilizes itself, but the SAF is unbalanced, in that the moment of the CoFe layer is always greater than that of the CoFeB layer. When the reverse applied field exceeds the exchange bias H_{ex} (~ 6 mT), the SAF stack reverses, giving the low resistance state. The switching field of the free CoFeB layer is H_c (~ 1.1 mT), which is similar to that reported by Wei *et al* [13]. The resistance of the stack is high or low, according to the relative alignments of the two CoFeB layers, as indicated in Fig. 3. The magnetization curve in Fig. 2 (a) indicates the additional switch of the NiFe seed layer, which has no influence on the resistance.

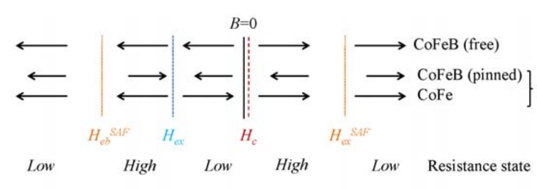


Figure 3 The switching sequence of the magnetic layers, which produces the inverted TMR. H_{ex} is the exchange bias field acting on the pinned CoFe layer. H_{ex}^{SAF} is the exchange field coupling the two layers of the SAF.

Almost no exchange bias was found in *M-H* and *R-H* curves when T_a was 350 ºC, which indicates disappearance of the exchange bias of the antiferromagnet. Instead of the exchange-biased reversal magnetization process at low T_a, a two-step reversal is observed for these FeMn-MTJs (see Fig. 2 (c) and (d)). Two different H_cs appear in both *M*-*H* and *R*-*H* curves, one H_c is 2.3 mT and the other is 10 mT. The small H_c belongs to the free CoFeB layer, the large one belongs to the synthetic antiferromagnetic CoFe/Ru/CoFeB stack. Here the H_c of the free layer increases a little compared to that at T_a = 250 ºC. The change of the H_c of the free CoFeB is mainly due to the crystallization of CoFeB film [14]. Moreover, elemental diffusion may occur in the free layer during the annealing process [15], which can also influence the H_c of the free CoFeB. Due to the thin pinned CoFeB layer, the annealing procedure may change the composition of the pinned CoFeB, as Mn enters it and B comes out [14], [15]. From this point of view, elemental diffusion is the important factor for the change of coercivity.

There are differences in the TMR effect between FeMn- and IrMn- MgO-based MTJs [9]. We have seen that the inverted TMR increases with increasing T_a, and reaches the maximum value at the same annealing temperature of 250 ºC for FeMn-MTJs with different pinned CoFeB thickness (0.75 nm, 1.5 nm, 2.0 nm). Then, the TMR ratio remains inverted up to t = 2.0 nm for FeMn-MTJs while it is normal for IrMn-MTJs at t = 2.0 nm. The TMR effect becomes normal for FeMn-MTJs when the thickness of the pinned CoFeB is 3 nm (see Fig. 1). Thirdly, the TMR ratio remains inverted at all annealing temperatures for IrMn-MTJs with $t \leq 1.5$ nm. As shown in Fig. 1, the TMR effect remains inverted around T_a = 350 ºC for IrMn-MTJs with t = 1.0 and 1.5 nm because of the higher blocking temperature of IrMn. Two factors may explain the different TMR behavior of FeMn-MTJs and IrMn-MTJs. One factor is that a rougher bottom CoFeB interface may appear during the stack growth when FeMn is chosen as the antiferromagnetic layer. Since the pinned CoFeB layer is thin, a much rougher bottom CoFeB/MgO interface occurs when a rougher FeMn is used, and a normal TMR effect could be obtained in FeMn-MTJs only with thicker CoFeB (see Fig. 1). This factor is responsible for the TMR effect for FeMn- and IrMn- MTJs at different CoFeB thickness and annealing temperature. The other factor is the tendency for Mn to diffuse more easily from FeMn as compared to the more chemically stable IrMn [16]. This factor influences the sign of TMR with the annealing temperature.

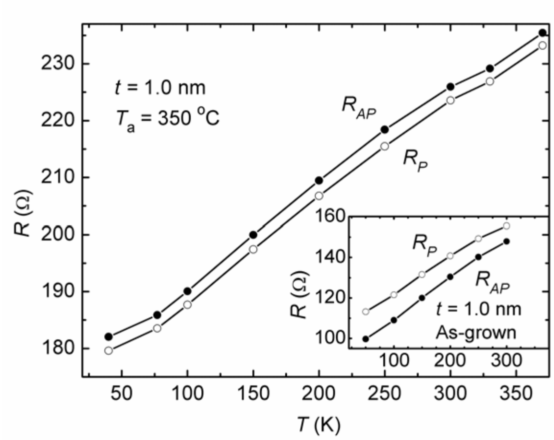


Figure 4 The temperature dependence of the junction resistance at T_a = 350 °C. The insets show the temperature dependence of the junction resistance in the as-grown state for another MTJ with t = 1.0 nm.

As shown in Fig. 4 and the inset, a metal-like junction resistance dependent on temperature is observed for these FeMn-MTJs in the as-grown and annealed states. In the as-gown state, the metal-like behavior may come from the locally thin regions in the MgO due to the roughness of the bottom CoFeB. The absence of similar results for IrMn-MTJs indicates that the FeMn layer has an important influence on the roughness. Furthermore, the thin regions in the MgO layer may become thinner at high T_a due to the diffusion. It is found that the metal-like behavior does not influence the sign of the TMR ratio for FeMn-MTJs, which can be seen from the TMR effect with T_a in Fig. 1 and 2. From this, one infers that most of tunneling occurs through the thinner regions in the MgO barrier. Furthermore, the current-voltage characteristics are nonlinear (Fig. 5), implying that these FeMn-MTJs have tunneling-type current-voltage characteristics. Despite roughness in the MgO barrier, the resistance-area (RA) products for FeMn-MTJs reach 10^5 Ω·μm^2. The barrier roughness lowers the RA value compared to that of IrMn-MTJs, which is an order of magnitude greater, 10^6 Ω·μm^2.

Because of the rough bottom CoFeB/MgO interface and then the rough MgO barrier, the spin filter effect of the crystalline MgO barrier is weak for these FeMn- MTJs. Only a relatively low TMR effect has been observed at room temperature (see Fig. 5). Besides, due to the elemental diffusion at the CoFeB/MgO interfaces at high T_a, the highest TMR value for these MgO-based FeMn-MTJs is even lower than that for the AlO_x-based MTJs [13]. Hence almost no coherent tunneling process takes place for these FeMn- MTJs. Diffusion reduces the TMR effect due to the decrease of the spin polarization at the interface. Besides the above-mentioned factors, the thinnest pinned CoFeB layer may form a discontinuous film, which has a strong inelastic tunneling effect that also much weakens the TMR.

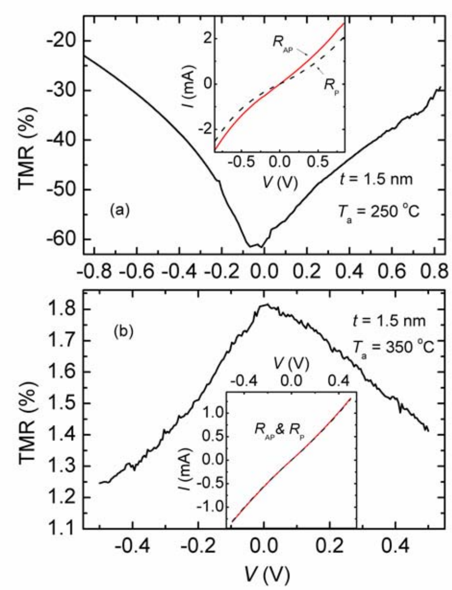


Figure 5 TMR ratios as a function of bias voltage at T_a = 250 and 350

°C. The insets in (a) and (b) are the *I-V* curves at the two annealing temperatures.

4. Conclusion

A sign reversal of the TMR effect in MgO-based FeMn-MTJs, changing from inverted to normal, has been studied as a function of the annealing temperature. The inverted TMR effect is due to the imbalance of the SAF stack. Elemental diffusion from the FeMn layer contributes to the change of TMR at high annealing temperature. From an application point of view, FeMn may not be a good choice as antiferromagnet because of the low thermal stability and the possibly increased roughness at the insulator/ferromagnet interface that destroys much of the TMR.

Acknowledgements

This work was supported by Science Foundation Ireland as part of MANSE project, and by the Ireland-China Scientific Exchange Scheme. Some of the work was supported by the EU as part of the BIOMAGSENS project. We are also grateful for support from the Chinese State Key Project of Fundamental Research of Ministry of Science and Technology (MOST, No.2006CB932200), and National Natural Science Foundation (NSFC No.10574156, 50528101 and 50721001).

References

[1] W. H. Butler, X.-G. Zhang, T. C. Schulthess, and J. M. MacLaren, Phys. Rev. B 63 (2001), 054416.

[2] J. Mathon and A. Umersky, Phys. Rev. B 63 (2001), 220403.

[3] S. Ikeda, J. Hayakawa, Y. Ashizawa, Y. M. Lee, K. Miura, H. Hasegawa, M. Tsunoda, F. Matsukura, and H. Ohno, Appl. Phys. Lett. 93 (2008), 082508.

[4] Y. M. Lee, J. Hayakawa, S. Ikeda, F. Matsukura, and H. Ohno, Appl. Phys. Lett. 89 (2006), 042506.

[5] Y. M. Jang, C. H. Nam, J. Y. Kim, B. K. Cho, Y. J. Cho, and T. W. Kim, Appl. Phys. Lett. 89 (2006), 163119.

[6] J. Hayakawa, S. Ikeda, Y. M. Lee, F. Matsukura and H. Ohno, Appl. Phys. Lett. 89 (2006), 232510.

[7] J. P. Nozières, S. Jaren, Y. B. Zhang, A. Zeltser, K. Pentek, and V. S. Speriosu, J. Appl. Phys. 87 (2000), 3920.

[8] G. Feng, S. van Dijken, and J. M. D. Coey, Appl. Phys. Lett. 89 (2006), 162501.

[9] J. F. Feng, G. Feng, J. M. D. Coey, X. F. Han, and W. S. Zhan, Appl. Phys. Lett. 91 (2007), 102505.

[10] J. H. Lee, C. S. Yoon, C. K. Kim, J. H. Yuh and Y. W. Kim, J. Appl. Phys. 91 (2001), 7472.

[11] M. Sato and K. Kobayashi, IEEE Trans. Magn. **33**, 3553 (1997).

[12] G. Hu and Y. Suzuki, Phys. Rev. Lett. 89 (2002), 276601.

[13] H. X. Wei, Q. H. Qin, M. Ma, R. Sharif, and X. F. Han, J. Appl. Phys. 101 (2007), 09B501.

[14] Y. H. Wang, W.C. Chen, S. Y. Yang, K.H. Shen, C. Park, M.J. Kao and M. J. Tsai, J. Appl. Phys. 99 (2006), 08M307.

[15] K. Komagaki, M. Hattori, K. Noma, H. Kanai, K. Kobayashi, Y. Uehara, M. Tsunoda, M. Takahashi, Intermag 2009 conference, DB-03.

[16] Mahesh G. Samant, Jan Lü ning, Joachim Stöhr, and Stuart S. P. Parkin, Appl. Phys. Lett. 76 (2000), 3097.