High inverted tunneling magnetoresistance in MgO-based magnetic tunnel junctions

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Inverted tunneling magnetoresistance, where resistance decreases as the free layer in a magnetic tunnel junction flips its direction of magnetization after saturation, has been observed at zero bias in magnetic tunnel junctions with a thin CoFeB layer in the pinned synthetic antiferromagnetic CoFe/Ru/CoFeB stack. Magnetoresistance values as high as −55% at room temperature are measured in MgO-based tunnel junctions when the thickness of the pinned CoFeB layer is 1.5 nm. The inverted magnetoresistance is associated with imbalance of the synthetic antiferromagnetic pinned layer. Asymmetric bias dependence with a magnetoresistance sign change is observed for a 0.5 nm pinned CoFeB layer. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2779241]

MgO-based magnetic tunnel junctions (MTJs) have transformed the prospects for spin electronic devices due to their remarkably high magnetoresistance at room temperature.[1–4] Tunneling magnetoresistance (TMR), defined as $(R_{AP}-R_P)/R_P$, can be as large as 360% in exchange-biased MTJs (Ref. 5) and 405% in unpinned pseudospin-valve MTJs.[6] Here R_P and R_{AP} are the resistances of the tunnel junctions when the magnetizations of the ferromagnetic electrodes in contact with tunnel barrier are parallel and antiparallel, respectively. High magnetoresistance in these devices is due to the spin filter effect of the crystalline MgO barrier, which is relatively transparent for majority spin electrons injected from an oriented bcc Fe or Fe–Co electrode but attenuates minority spin electrons, as result of the different symmetries of the ↑ and ↓ wave functions. Besides high TMR values, linear and hysteresis-free switching has also been observed in MgO-based MTJs when the thickness of the free CoFeB layer is less than 1.0 nm.[7] Moreover, an interfacial resonance state located in the minority band of Fe (001), which has been probed by spin-polarized tunneling in epitaxial Fe/MgO/Fe MTJs, causes the TMR to change sign from positive to negative above a critical voltage.[8]

Here we report the observation of a high inverted magnetoresistance when the free layer in MgO-based MTJs switches at zero bias. A switch in the sign of the resistance change near zero applied field occurs as a function of the thickness of the pinned CoFeB layer next to the MgO barrier in an MTJ stack with a synthetic antiferromagnet. The effect is not found when a single pinned layer is used.

MTJs with a complete layer sequence Si/SiO_2(substrate)/Ta(5)/Ru(50)/Ta(5)/$Ni_{81}Fe_{19}$(5)/$Ir_{22}Mn_{78}$(10)/$Co_{90}Fe_{10}$(2)/Ru(0.85)/CoFeB(t)/MgO(2.5)/CoFeB(3)/Ta(5)/Ru(5) were grown using a Shamrock sputtering tool, where the numbers in parentheses are layer thicknesses in nanometers. The thickness (t) of the pinned CoFeB ($Co_{40}Fe_{40}B_{20}$) layer was varied from 0 to 3.0 nm. The MgO was grown in a separate zone of our sputtering system, from a target-facing-target source. Similar MTJs with AlO_x barriers were also prepared for a comparative test. Well-oriented (001) MgO barrier layers were confirmed by x-ray diffraction.[9] After deposition of the stack, square-shaped junctions with an area from 12×12 to 24×24 μm^2 were fabricated using conventional ultraviolet lithography. Typical resistance-area products were around 5×10^6 Ω μm^2, which is close to that reported by Jiang *et al.*[7] for barriers of the same thickness. Thermal annealing of the patterned junctions was carried out at temperatures ranging from 150 to 400 °C under vacuum for 1 h in an applied magnetic field of 800 mT. All magnetotransport measurements were performed by the standard four-probe technique. Electrons flowing from the top to the bottom electrode are defined as a positive current.

Figure 1(a) shows the TMR ratios in MgO- and AlO_x-based MTJs under a small bias voltage of 2 mV for the pinned CoFeB thickness dependence ($0\leq t\leq3.0$ nm). By changing t, a sign reversal of TMR has been obtained for *both kinds* of barriers. For as-grown AlO_x-based MTJs, inverted TMR values of −0.1%, −8.1%, and −9.5% were obtained when t=0.5, 1.0, and 1.5 nm, respectively. The sign relates to the sense of the resistance change near zero field. Moreover, an inverted TMR value of −0.3% was observed in AlO_x-based MTJs when Ru was in direct contact with the barrier (t=0), which is in agreement with results published by other groups.[10,11] When MgO is the tunneling barrier, much higher inverted TMR values are observed. However, no TMR effect is observed in MgO-based MTJs without the pinned CoFeB layer (t=0), which is consistent with the requirement for crystalline bcc Fe to be in contact with the MgO if the spin filter effect is to operate.[1,2] Figure 1(b) presents representative TMR curves of MgO-based MTJs with t=0.5, 1.0, and 2.0 nm. The magnitude of the TMR at 300 K increases up to −55% for t=1.5 nm, in that series of devices, and a sign reversal occurs between 1.5 and 2.0 nm. Positive TMR ratios of 115% and 130% were observed for t=2.0 and 3.0 nm, respectively.

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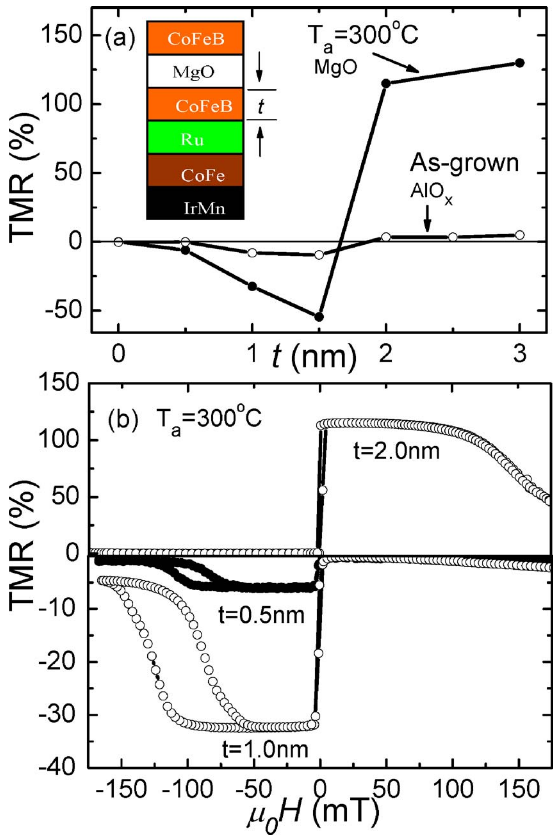

FIG. 1. (Color online) (a) Pinned CoFeB thickness dependence of the TMR ratios in MTJs with MgO and AlO_x barriers in the thickness range $0 \leq t \leq 3.0$ nm. Inset shows the structure of the MTJ stack. (b) Representative TMR curves of MgO-based MTJs with t=0.5, 1.0, and 2.0 nm which were annealed at T_a=300 °C and measured at T=300 K.

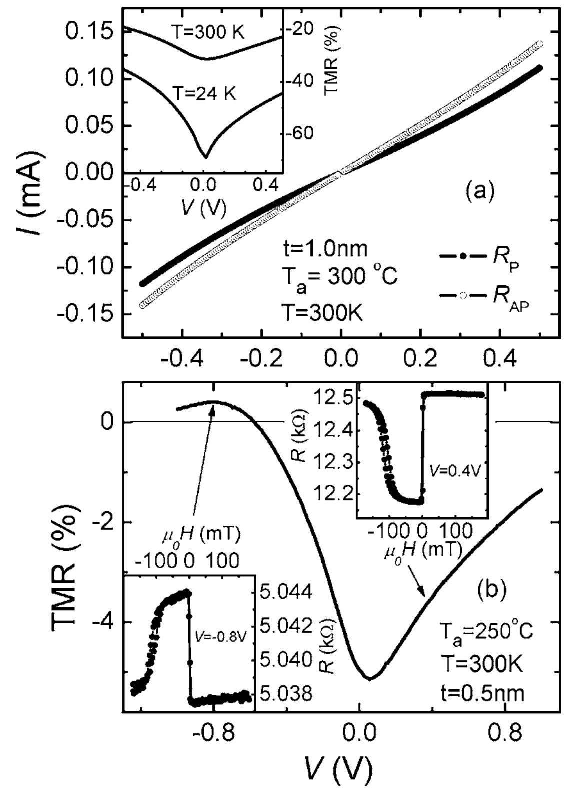

FIG. 3. (a) Representative current-voltage characteristics at 300 K in MgO-based MTJs with t=1.0 nm for parallel and antiparallel states. Inset shows the resistance in the parallel and antiparallel configurations at 24 K. (b) The bias voltage dependence of the TMR ratio in an MgO-based MTJs with t =0.5 nm at 300 K. Insets show the normal and inverted TMR curves at V =−0.8 and +0.4 V.

Figure 2 shows the inverted room temperature TMR ratios as a function of annealing temperature (T_a) for MgO-based MTJs with t=0.5, 1.0, and 1.5 nm. A similar T_a dependence has been observed in all MTJs with different pinned CoFeB thicknesses. The TMR increases with increasing T_a, reaching the highest value at 250–300 °C. After that, the inverted TMR decreases. The fall in TMR at high T_a might be attributed to Mn diffusion in the annealing process when a very thin pinned CoFeB layer is present. It is found that higher annealing temperatures can be used with a thicker pinned layer, which implies that the thicker CoFeB layer does reduce the thermal diffusion of Mn in the annealing process. The highest TMR ratios of −55% and +142% were found at T_a=300 and 325 °C for t=1.5 and 2.0 nm, respectively.

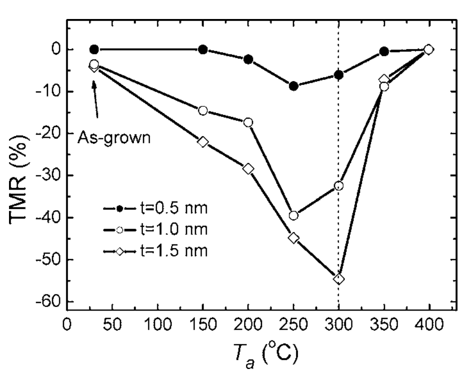

FIG. 2. TMR ratios at 300 K after annealed at various temperatures for MgO-based MTJs with t=0.5, 1.0, and 1.5 nm. The dotted line marks the TMR ratios at T_a=300 °C which are plotted in Fig. 1.

From the current-voltage characteristics measured at 300 K, shown in Fig. 3(a), it is clear that tunneling transport is taking place in these MTJs. The inset of Fig. 3(a) compares the bias dependence in the high and low resistance configurations at 24 K. A cusplike zero-bias anomaly is observed in the high resistance configuration, which is smeared out at 300 K. This is similar to MgO MTJs with $t \geq 2.0$ nm having the normal sign of magnetoresistance, where the zero bias anomaly which gives rise to the cusplike TMR appears also in the high resistance configuration. The low temperature characteristics have been discussed previously in terms of magnon-assisted inelastic tunneling.[12–14]

Figure 3(b) shows the bias voltage dependence of the TMR ratios in MgO-based MTJs with t=0.5 nm at 300 K. Although the TMR value for t=0.5 nm is small compared to that for t=1.0 nm, an interesting result appears. There is a sign change of TMR at negative bias. Typical positive and negative TMR curves at V=−0.8 and +0.4 V are shown in the insets of Fig. 3(b). Asymmetric bias dependence is associated with a physical asymmetry of the tunnel junction.[15–17] It may reflect an asymmetry between the top and bottom oxide/ferromagnet interfaces, with a strong density-of-states modification at one of them as previously proposed for AlO_x (Ref. 10) and for MgO.[15] The change of TMR sign with bias [Fig. 3(b)] suggests pronounced structure in the density of states within 0.6 eV of the Fermi level.[8,18]

Our data demonstrate that inverted TMR at zero bias can be observed for both MgO- and AlO_x-based MTJs, which indicates that the effect does not depend on the nature of the barrier. MgO acts as spin filter when grown together with an epitaxial bcc Fe–Co layer,[1,2] whereas amorphous AlO_x has no wave vector selection for the transmitted electrons. The critical requirement is that the CoFeB layer is thin and that the Ru layer lies close to the barrier. One suggestion for negative TMR is formation of a Ru quantum well.[11,19,20] We found little or no effect when the ruthenium is in contact with the barrier oxide—a small negative TMR (−0.3%) in AlO_x-based MTJs with no pinned CoFeB layer ($t=0$), and no effect at all for MgO. If the negative TMR in this case could be attributed to a Ru quantum well, its contribution to the TMR effect would be expected to weaken when a thin CoFeB layer is inserted between the Ru and AlO_x. Instead, the negative TMR increases on increasing the thickness of the inserted CoFeB ($t>0$, in Fig. 1). It seems that a Ru quantum well cannot be the main reason for our observations.

Negative TMR can be observed when a tunnel junction is built from two different ferromagnetic electrodes with opposite spin polarization.[21] The scattering of majority-spin electrons by Ru impurities in Co or Fe is known to be several times greater than the scattering minority-spin electrons.[22] This is reflected in the negative scattering anisotropy parameter γ of the CoFeB/Ru interface.[23] Furthermore the Ru moment couples parallel to that of the $3d$ ferromagnet. Electrons transmitted at the CoFeB/Ru interface will be predominately minority spin. If the thickness t of the CoFeB pinned layer is insufficient to repolarize them, or if it is discontinuous, negative TMR can result.

The question is really whether we are observing true negative TMR, or whether our observations are due to a change in switching sequence caused by the imbalance of the synthetic antiferromagnet. The two possibilities can be distinguished by measurements in higher fields. If the TMR is intrinsically negative, switching the layers of the synthetic antiferromagnet from antiparallel to parallel will barely influence the resistance. However, if low resistance state is due to an inversion of the exchange bias, which leads to antiparallel alignment of the CoFeB and the free layers in small reverse fields, then the low resistance state should be recovered in fields that are large enough to overcome the exchange coupling σ_{af} of the synthetic antiferromagnet. This is exactly what we find in the high field measurement shown in Fig. 4. In fields of ±1000 mT the low-resistance state is recovered. The estimated value of σ_{af} is therefore 1 mJ m^{-2}.

In conclusion, the inverted TMR in MTJs with both AlO_x and MgO tunnel barriers is due to imbalance of the synthetic antiferromagnet. The asymmetry of the junction evident in Fig. 3(b) and the change of sign of the TMR appear to be related to diffusion of Ru to one side of the barrier.

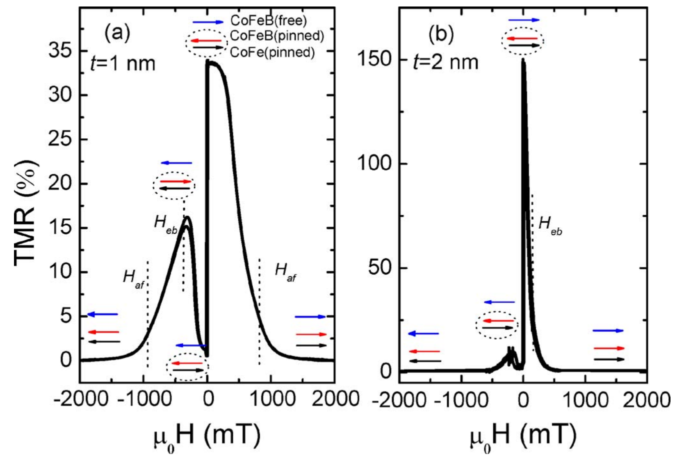

FIG. 4. (Color online) Tunnel magnetoresistance in high magnetic fields for MgO-based MTJs with $t=1.0$ and 2.0 nm at 300 K. The antiferromagnetic exchange field H_{af} and the exchange bias field H_{eb} are indicated.

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