MgO-based double barrier magnetic tunnel junctions with thin free layers

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(Presented 11 November 2008; received 22 September 2008; accepted 24 November 2008; published online 9 March 2009)

The free layer thickness (t_{free}) in double barrier magnetic tunnel junctions (DMTJs) based on crystalline MgO barriers and CoFeB ferromagnetic layers has been varied from 0.5 to 3.0 nm in order to investigate its effect on the magnetic and electrical properties. One obvious feature of DMTJs with $t_{\mathrm{free}} \leq 1$ nm is the absence of sharp free layer switching in the TMR curves, which can be explained by the superparamagnetic nature of discontinuous CoFeB layer, which breaks into nanodots when it is very thin. Normal free layer switch is observed when $t_{\mathrm{free}}=2.0$ and 3.0 nm. Another difference is a rapid increase in junction resistance and tunnel magnetoresistance at low temperature for DMTJs with thin t_{free}, which is attributed to the Coulomb blockade effect. We also observed a small conductance peak in the dI/dV curve at low bias only in the parallel configuration and at temperatures below 100 K. This is related to the Kondo scattering process on the nanodots, which constitutes the discontinuous free layer. We found no Coulomb staircase existing in the I-V curves; this may be due to the microsize of the junctions. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3072474]

I. INTRODUCTION

Crystalline MgO thin film has been one of the most promising barriers since large tunnel magnetoresistance (TMR) was reported in MgO-based magnetic tunnel junctions (MTJs).[1,2] Double barrier MTJ (DMTJ), which consists of three ferromagnetic electrodes and two barriers, can be thought of as two single-barrier tunnel junctions (SMTJs) connected in series. The advantage for DMTJs is that the voltage across each junction can be much lower relative to the breakdown voltage, so the lifetime of the device is likely to be significantly increased.[3–5] In DMTJs, if the middle electrode is thin enough and the charging energy of these thin ferromagnetic layers is comparable to the applied voltage, then the Coulomb blockade phenomena may be observed. In the Coulomb blockade regime, sequential tunneling, which is the case in SMTJs and means the electrons tunnel through both barriers in an uncorrelated fashion, is strongly suppressed; cotunneling, where each electron tunnels into the middle electrodes and another electron must simultaneously leave the middle electrode, dominates the tunnel process. Theoretical calculations for DMTJs in the Coulomb blockade regime predict an enhanced TMR due to cotunneling,[6–8] which has been verified experimentally in the AlO_x-based MTJ system.[9–12] In this paper we will present the transport properties of MgO-based DMTJ with thin t_{free}, which can be related to the Coulomb blockade effect.

II. EXPERIMENT

DMTJs consisting of a 5 Ta/50 Ru/5 Ta/5 $\mathrm{Ni_{81}Fe_{19}}$/10 $\mathrm{Ir_{22}Mn_{78}}$/2 $\mathrm{Co_{90}Fe_{10}}$/0.85 Ru/3 $\mathrm{Co_{40}Fe_{40}B_{20}}$/t MgO/3 $\mathrm{Co_{40}Fe_{40}B_{20}}$/0.85 Ru/2 $\mathrm{Co_{90}Fe_{10}}$/10 $\mathrm{Ir_{22}Mn_{78}}$/5 Ta (t=0.5, 1.0, 2.0, and 3.0, thickness in nm) were grown by magnetron sputtering on thermally oxidized Si substrates in a Shamrock deposition tool. The MgO barrier was fabricated by rf sputtering from two MgO targets in a target-facing-target (TFT) gun. UV lithography was used to pattern the multilayer stacks into junctions with areas between 20×20 $\mu\mathrm{m}^2$ and 100×100 $\mu\mathrm{m}^2$. After fabrication, DMTJs were annealed in vacuum for 1 h at different temperatures. During annealing a magnetic field of 0.8 T was applied to establish a uniform exchange bias in the bottom electrode. The transport properties were then characterized by a standard four-point method. Magnetic properties were measured by alternating gradient force magnetometer (AGFM) from unpatterned samples directly.

III. RESULTS AND DISCUSSION

Figure 1 shows the room temperature (RT) TMR as a function of magnetic field for both as-deposited [Figs. 1(a) and 1(b)] and annealed [Figs. 1(c) and 1(d)] DMTJs with $t_{\mathrm{free}}=1.0$ and 3.0 nm. Both as-deposited samples show the same TMR of 12% at RT and no exchange bias can be observed [Figs. 1(a) and 1(b)]. After annealing at 300 °C [Fig. 1(c)], DMTJs with $t_{\mathrm{free}}=3.0$ nm show a reasonable TMR ratio (120%) and well separated switches between the middle free layer and the two pinned layers. The TMR for DMTJs with $t_{\mathrm{free}}=1.0$ nm can also be enhanced by annealing, but it is smaller compared to that of $t_{\mathrm{free}}=3.0$ nm, around 39% at RT [Fig. 1(d)]. An obvious difference from TMR curves is the absence of free layer and top pinned layer switches for DMTJs with $t_{\mathrm{free}}=1.0$ nm [Fig. 1(d)]. For DMTJs with $t_{\mathrm{free}}=3.0$ nm [Fig. 1(c)], there is a sharp TMR increase around zero field, which corresponds to the free layer switch, and the first TMR drop at 21 mT indicates the magnetic moment flip of the top pinned CoFeB layer. Both of those TMR changes disappear on the curve for $t_{\mathrm{free}}=1.0$ nm

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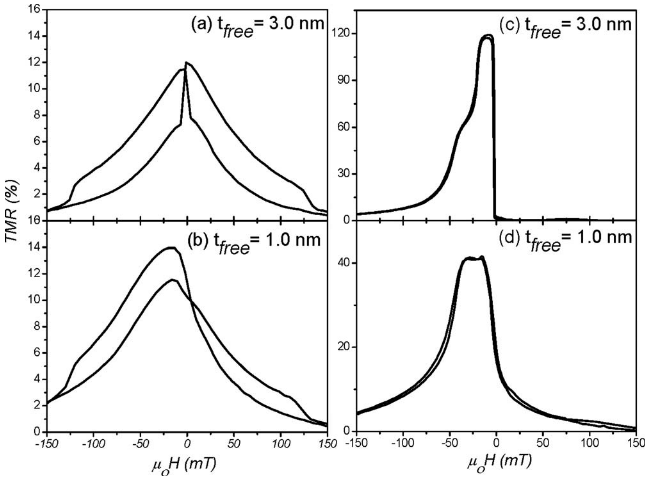

FIG. 1. TMR curves for DMTJs with $t_{\mathrm{free}}=1.0$ and 3.0 nm under [(a) and (b)] as-deposited state and [(c) and (d)] after annealing at 300 °C.

DMTJs. No transport data for DMTJs with $t_{\mathrm{free}}=0.5$ nm is shown in this paper because their resistance is larger than 10 GΩ, which is out of range of our magnetotransport properties measurement system. It is assumed that the two barriers are connected to each other and act as one single barrier with twice the thickness.

The reason for the unseparated switches between the free layer and top pinned layers in the DMTJ with thin t_{free} can be explained by the paramagnetic nature of thin CoFeB (≤1.0 nm), which is also the case in SMTJs.[13] The magnetization curves for unpatterned samples with three different t_{free} are shown in Fig. 2. A clear free layer switch can be observed only for $t_{\mathrm{free}}=3.0$ nm but not for $t_{\mathrm{free}}=0.5$ and 1.0 nm. The magnetic moment for a thin CoFeB (≈1.0 nm) layer orients randomly at zero field and can be gradually rotated along the applied magnetic field. However its whole magnetic moment cannot be totally aligned, even under a 100 mT field (not shown). This can be confirmed by the smooth change in resistance around zero field in the TMR curve. The top pinned layer starts to flip around 21 mT, which is not high enough to align the magnetic moment of middle layers.

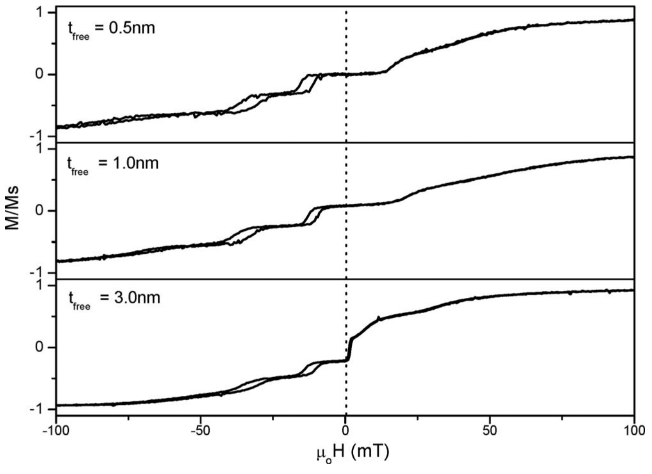

FIG. 2. The magnetization vs magnetic field curves for DMTJ with 0.5, 1.0, and 3 nm middle CoFeB layer.

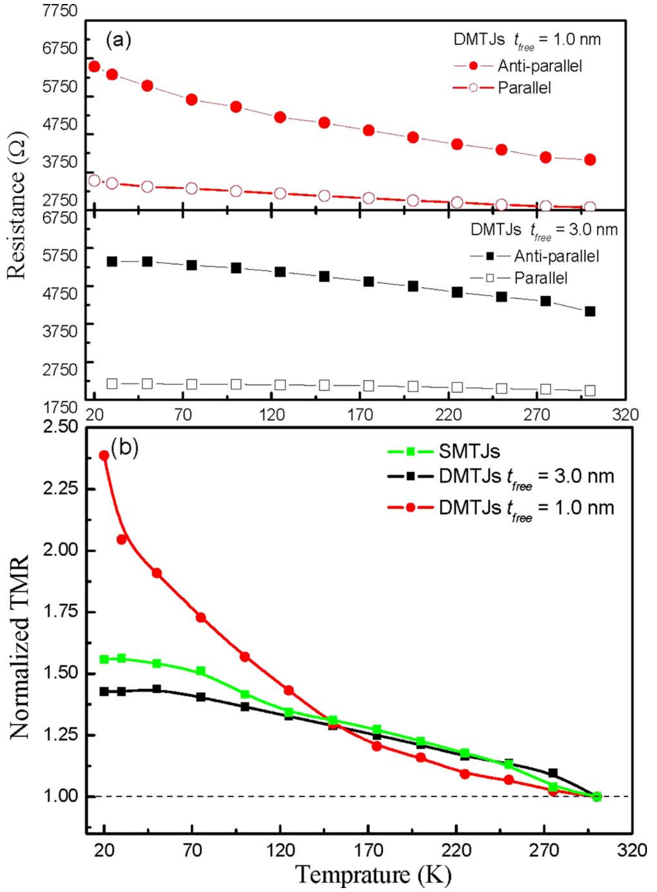

FIG. 3. (Color online) (a) Temperature dependence of the resistance of DMTJs with $t_{\mathrm{free}}=1.0$ and 3.0 nm (b) Temperature dependence of normalized TMR ratio for SMTJs and DMTJs with $t_{\mathrm{free}}=1.0$ and 3.0 nm.

The temperature dependence of resistance [in both parallel (P) and antiparallel (AP) configurations] for DMTJs with $t_{\mathrm{free}}=1.0$ and 3.0 nm has been shown in Fig. 3(a). The resistance of $t_{\mathrm{free}}=1.0$ nm shows a much stronger temperature dependence at low temperature regimes (<50 K) compared with that of $t_{\mathrm{free}}=3.0$ nm, which indicates the Coulomb blockade effect.[10,11] Very similar experimental result was recently reported by Yang *et al.*,[14] which shows that the Coulomb blockade effect can be observed at low bias and low temperature for t_{free} of 1.5 nm.

A comparison of the temperature dependence of normalized TMR between SMTJs and DMTJ ($t_{\mathrm{free}}=1.0$ and 3.0 nm) are shown in Fig. 3(b). At RT, DMTJ with $t_{\mathrm{free}}=1.0$ shows a TMR ratio of 39% and *RA* values of 3.6×10^{6} $\Omega\ \mu\mathrm{m}^2$ (AP) and 2.5×10^{6} (P)$\Omega\ \mu\mathrm{m}^2$. The two *RA* values reach 3.5×10^{7} $\Omega\ \mu\mathrm{m}^2$ (AP) and 2.5×10^{7} (AP)$\Omega\ \mu\mathrm{m}^2$ at 20 K. This gives a TMR of 104%, which is 2.6 times larger than the RT value. This temperature associated TMR enhancement is more remarkable than that of SMTJs and DMTJs with $t_{\mathrm{free}}=3.0$ nm. TMR increases 1.5 times, from 229% to 352%, for SMTJs when the temperature drops to 20 K; it increases 1.3 times for DMTJs with $t_{\mathrm{free}}=3.0$ nm, from 120% to 154%. The increase in the TMR for SMTJs and DMTJs with $t_{\mathrm{free}}=3.0$ nm follows the same tendency, but DMTJs with $t_{\mathrm{free}}=1.0$ nm behave differently. There is a big TMR increase when temperature decreases from 50 to 20 K. It is believed that this increase in TMR is due to the cotun-

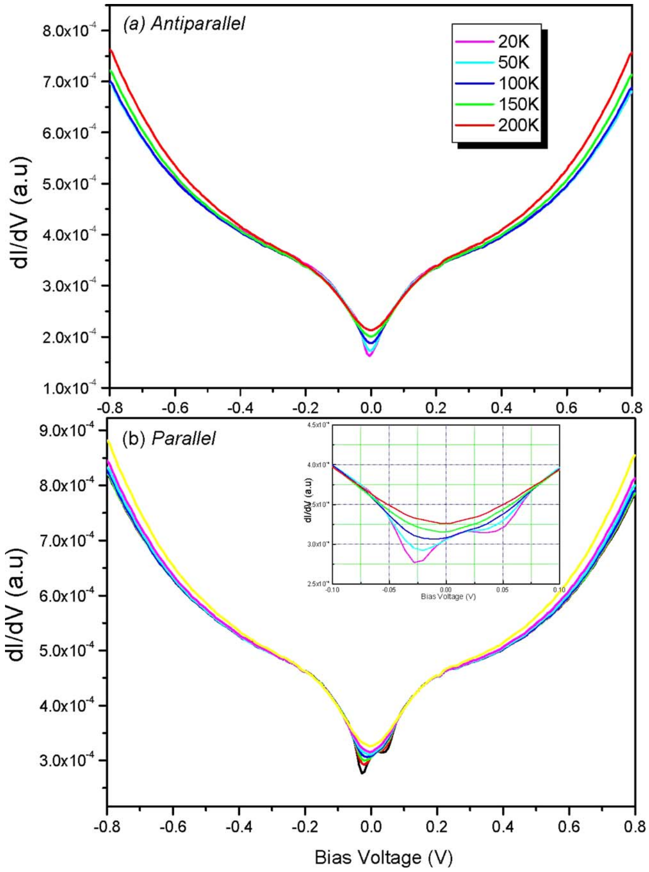

FIG. 4. (Color online) dI/dV curves for DMTJs with $t_{\text{free}}=1.0$ nm measured at different temperatures. (a) AP configuration. (b) P configuration.

neling in the Coulomb blockade regime. In this regime, sequential tunneling is strongly suppressed. The electrons must tunnel into the middle electrode after the simultaneous leaving of certain electrons also from the middle electrode. Thus the dramatic increasing in resistance in the AP configuration can be expected.

The TMR ratio of DMTJs with $t_{\text{free}}=1.0$ nm is lower than that of DMTJs with $t_{\text{free}}=3.0$ nm even under low temperature and low bias. This could be explained as follows. (1) The reduction in polarization due to the superparamagnetic nature of thin free layers could be one of the reasons. (2) The TMR can be reduced because a perfect AP configuration could not be achieved between the thin free layers and the two pinned layers. The sharpness and position of coercive field of the free layer strongly influence the TMR value of double barrier junction. (3) Another factor which affects the TMR ratio is the orientation of middle CoFeB free layers after postannealing. As we know, highly oriented CoFe (001)/MgO (001) interfaces are required to guarantee the larger TMR in MgO-based MTJs. However with (001) oriented MgO barrier on each side, to crystallize quasiepitaxially the middle free layer on both of them would be really difficult.

Figure 4 shows the conductance of DMTJs with $t_{\text{free}}=1.0$ nm measured at different temperatures. Dips appear under both the P and AP configurations, which is the so-called "zero-bias anomaly." The dips, existing at all the temperatures, indicate that this anomaly is not because of Coulomb blockade; it is usually explained by magnon and/or phonon assisted tunneling.[15,16] Similar characterization has also been reported for MgO/Fe DMTJs with the middle Fe less than 1.5 nm,[5] the disappearance of the dips for thicker Fe is similar with our results of $t_{\text{free}}=3.0$ nm. Another interesting observation is the appearance of a small peak from the conductance curve of the P configuration at 20 K [inset in Fig. 4(b)]. The difference seen in the curve is believed to be related to the electronic structure and tunneling processes that depend critically on the thickness of the middle CoFeB layers and its crystalline structure. Yang *et al.*[14] believe that the conductance peak is one of the characteristic features of Kondo-assisted tunneling. They observed a similar peak for a DMTJs with 0.5 nm CoFe middle layer at 2.5 K for both P and AP configurations and claimed that Kondo temperature is 10%–20% higher for AP configuration.

IV. CONCLUSIONS

The magnetic and transport properties for DMTJs with different middle CoFeB layer thickness have been characterized. The absence of free layer switch for DMTJs with thin t_{free} can be attributed to the superparamagnetic nature of the discontinuous CoFeB layers. The strong temperature dependence of junction resistance and TMR ratio are related to the Coulomb blockade effect at the low temperature regime.

ACKNOWLEDGMENTS

This work was supported by Science Foundation Ireland as part of the CINSE and MANSE projects, and the European BIOMAGSENS project. We are grateful to H. C. Wu for the AGFM measurement.

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