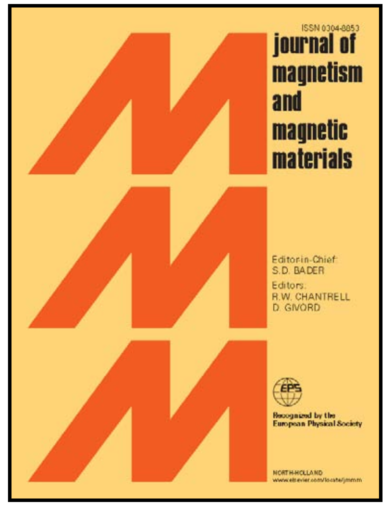

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Bias Voltage Dependence of Inverted Magnetoresistance on the Annealing Temperature in MgO-based Magnetic Tunnel Junctions

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Abstract

MgO-based magnetic tunnel junctions (MTJs) with a layer sequence $Ir_{22}Mn_{78}$ or $Fe_{50}Mn_{50}$ (10 nm) /CoFe (2 nm) /Ru (0.85 nm) /CoFeB ($0.5 \leq t < 2$ nm) /MgO (2.5 nm) /CoFeB (3 nm) have been fabricated. The bias voltage dependence of TMR is given as a function of the annealing temperature for these MTJs, which shows the TMR ratio changes its sign from inverted to normal at a critical bias voltage (V_C) when an unblanced synthetic antiferromagnetic stack CoFe/Ru/CoFeB is used. V_Cs change with the thickness of the pinned CoFeB and annealing temperature, which implies one can achieve different V_Cs by artificial control. The asymmetric V_C values suggest that a strong density-of-states modification occurs at bottom oxide/ferromagnet interface.

PACS: 73.40.Rw; 73.40.Gk; 75.70.-I; 85.70Kh

Keywords: Magnetic tunnel junctions (MTJs); Tunneling magnetoresistance (TMR); Bias dependence

1. Introduction

Spin polarized tunneling magnetoresistance (TMR) research has been much promoted since TMR was discovered in 1975 [1]. Subsequently slow progress was made with amorphous AlO_x-based magnetic tunnel junctions (MTJs) at room temperature [2]–[4], and high magnetic recording density was successfully realized in hard disk drives using AlO_x-based MTJs and the TMR effect few years ago [5]. In 2001, Butler and Mathon *et. al.* predicted the large TMR effect in single-crystal MgO-based MTJs [6], [7]. Soon after that, very large TMR ratios at room temperature were achieved in such MTJs [8], [9]. Nowadays well-oriented (001) MgO-based MTJs have given a TMR ratio of more than 1000% at low temperature and 500% at room temperature [10], which accords with the theory perfectly [6], [7]. From an application point of view, MgO-based MTJs are the better choice for next-generation magnetic recording and memory devices due to their higher sensitivity and lower noise compared to those of AlO_x-based MTJs.

The high TMR effect in MgO-based MTJs devices is due to the spin filter effect of the crystalline MgO barrier, which 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 ↑ and ↓ wavefunctions. Besides successfully achieving high TMR in MgO-based MTJs, many other results were introduced at the same time. For example, 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 TMR to change its sign from positive to negative above a critical bias voltage [11].

Sign reversal of TMR ratio as a function of bias has also been observed in AlO_x-based MTJs when a non-magnetic metal layer, e.g. Ru, Cu, and Au, is inserted between the magnetic electrode and the tunneling barrier [12]-[14]. The sign reversal usually depends on the thickness of non-magnetic metal layer. Quantum wells were considered as the reason for the sign reversal of the TMR [12]-[13]. Inversion of the TMR *at zero bias* can also arise when an unbalanced synthetic antiferromagnet (SAF) is used as the pinned layer

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[15].

In this work, we fix the thickness of the non-magnetic metal Ru (0.85 nm) in the sandwich CoFe/Ru/CoFeB, and change the thickness of the pinned CoFeB electrode in the SAF from 0.5 to 1.5 nm. Because of the single-crystal requirement for MgO to get high TMR in MgO-based MTJs, an amorphous CoFeB alloy is a good choice to achieve it. To obtain a single-crystal MgO, a thin CoFeB layer on Ru is selected, which also can realize sign reversal of the TMR as discussed in the work. The diagram of the critical bias voltage (V_C) versus the pinned CoFeB thickness and annealing temperature shows that one can control V_C as well.

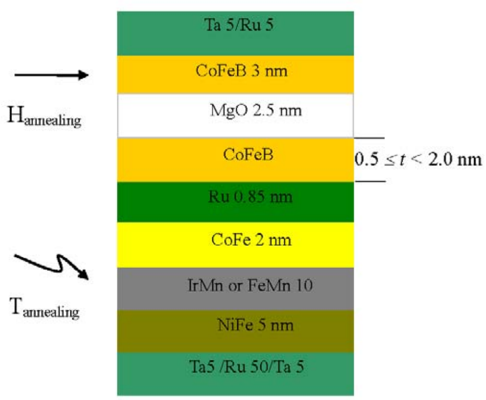

Fig. 1 The cross-sectional view of MgO-based MTJs. $H_{annealing}$ and $T_{annealing}$ imply the magnetic field and annealing temperature were applied during the annealing process.

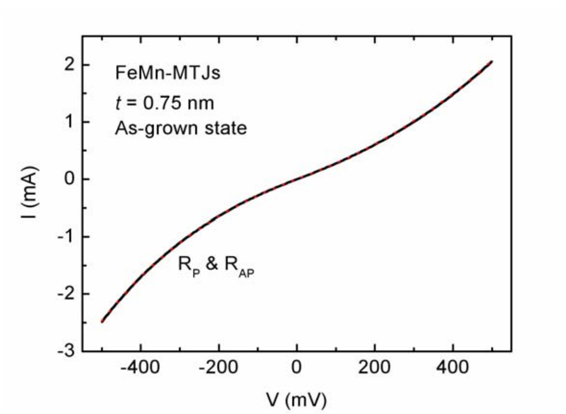

Fig. 2 The current-voltage curves in the as-grown state for MgO-based FeMn-MTJs with t = 0.75 nm, taken T = 300 K.

2. Experiment

The sequential sputtering method was used for sample preparation. Typical MTJ multilayer design is Ta (5) /Ru (50) /Ta (5) / $Ni_{81}Fe_{19}$ (5) /$Ir_{22}Mn_{78}$ (10) /$Co_{90}Fe_{10}$ (2) /Ru (0.85) / $Co_{40}Fe_{40}B_{20}$ (t) /MgO (2.5) / $Co_{40}Fe_{40}B_{20}$ (3) /Ta (5) /Ru (5), and the order is from the bottom electrode to the top, with thickness given in nanometers [15]. The thickness (t) of the pinned $Co_{40}Fe_{40}B_{20}$ (CoFeB) layer was varied from 0.5 to 1.5 nm. Moreover, MTJ stack with $Fe_{50}Mn_{50}$ (10 nm) was also prepared for comparison. All multilayers were grown under high vacuum and at room temperature in a Shamrock sputtering tool. A well-oriented (001) MgO barrier can be achieved and high TMR ratio was observed in MTJs with a thick pinned CoFeB [15], [16].

After deposition of the MTJ stack, square-shaped junctions with an area from 12 × 12 to 24 × 24 μm^2 were fabricated using conventional UV lithography. After checking them in the as-grown state, thermal annealing of the patterned junctions was carried out at temperatures ranging from 150 to 400ºC, under vacuum for one hour in an applied magnetic field of 800 mT (see Fig. 1). All magnetotransport measurements were performed by the standard four-probe technique at room temperature, with the magnetic field applied in the plane of MTJs. Electrons flowing from the top to the bottom electrode are defined as a positive current.

3. Results and Discussions

These MgO-based MTJs with a thin pinned CoFeB are of good quality, and their current-voltage characteristics are nonlinear (Fig. 2), as expected for spin polarized tunneling across the MgO insulator layer. Fig. 3 (a) shows the TMR ratio changes with the bias voltage in the as-grown state for FeMn-MTJs. The inverted TMR ratio is about -2.2% *at zero bias* and room temperature. After annealing at a temperature (T_a), its value increases greatly. For these MTJs, an inverted TMR as high as -55% has been observed when T_a = 250 °C and t = 1.5 nm [15]. Here the TMR ratio is defined as (R_{AP}-R_P)/R_L, R_P and R_{AP} are the resistances of MTJs when the magnetizations of the ferromagnetic electrodes in contact with tunnel barrier are parallel and antiparallel, and R_L is the lower one. It is found that the TMR ratio decreases rapidly when a non-magnetic metal layer is inserted between the magnetic electrode and the tunnel barrier in AlO_x-based MTJs [11]-[14]. However, the TMR ratio in MgO-based MTJs with a thin pinned CoFeB does not decrease so fast.

We find that the typical characteristic for these inverted MgO-based MTJs is that their TMR ratio changes its sign from inverted to normal as a function of bias, even without any annealing, as shown in Fig. 3 (a). For as-grown MTJs with t = 0.75 nm, V_C appears at about -280 mV. Here V_C is defined as the bias voltage at which TMR = 0. The detailed inverted and normal TMR curves at V_C = -400 and +200 mV are shown in Fig. 3 (b) and (c), respectively. A gradual magnetization reversal is observed in the *R* - *B* curves due to the thin pinned CoFeB electrode.

A possible discontinuous CoFeB film may form on Ru when the pinned CoFeB layer in the synthetic antiferromagnetic stack is thin (t ≤ 1.0 nm), and the non-magnetic metal Ru was in direct contact with the MgO barrier at some interface, which may cause the sign reversal of TMR [12]-[14]. However, it seems that a Ru quantum well cannot be the main reason for our observations [15]. In addition, no obvious TMR can be seen in the as-grown state or for T_a = 150 °C for MTJs with t = 0.5 nm. At T_a > 150 °C, the TMR effect appears, which may be due to the annealing process. The annealing effect has a key influence for MTJs, which can effectively improve the quality of the oxide/ferromagnet interface and increase the TMR ratio.

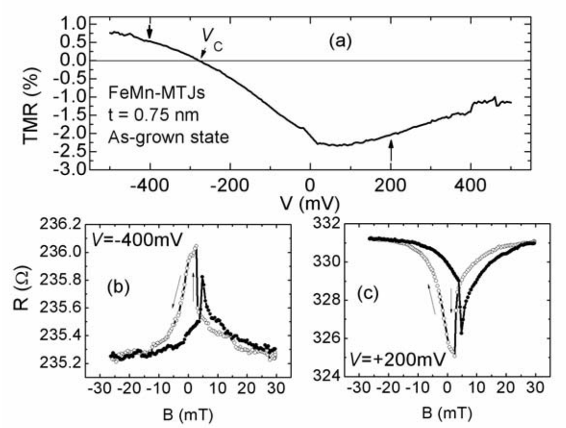

Fig. 3 (a) The bias voltage dependence of the TMR ratio in MgO-based FeMn-MTJs with t = 0.75 nm at 300 K. (b) and (c) The normal and inverted TMR curves at V = -400 and +200 mV; the arrow marks in (a) are the detailed bias positions in (b) and (c). The data in this figure and figure 2 are taken from the same junction.

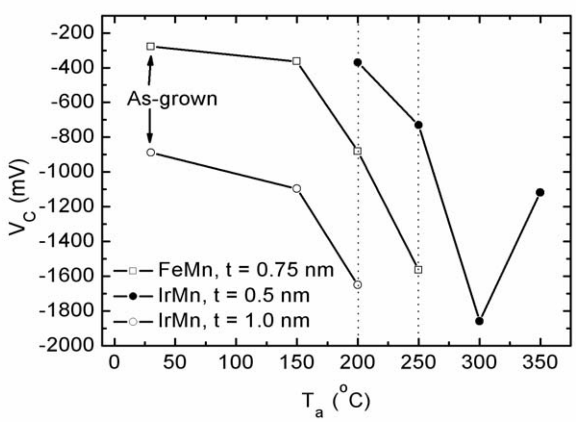

Fig. 4 The critical bias voltages (V_Cs) as a function of annealing temperature under different pinned CoFeB thicknesses in MgO-based FeMn- & IrMn- MTJs, taken at 300 K.

Figure 4 plots V_C versus annealing temperature at different pinned CoFeB thicknesses. The V_C value shown in Fig. 4 is an average one, which implies that the V_C value is not fixed at a fixed t and T_a due to the different physical position for MTJs of the same series [17]. V_C increases with increasing T_a and then starts to decrease around a certain T_a. A similar change is observed in TMR versus T_a, which suggests that the decrease of V_C may be due to Mn diffusion at higher T_a [15]. For example, V_C changes from about -245 mV at T_a = 200 °C to about -1860 mV at T_a = 300 °C for MTJs with t = 0.5 nm. For this reason, V_C at higher T_a was not attained in MTJs with $t \geq$ 0.75 nm since a larger bias voltage need to be applied and MTJs were easily destroyed! V_C increases with the increase of the thickness of the pinned CoFeB layer. For the same reason, no obvious V_C can be observed when $t \geq 1.5$ nm. Moreover, the pinned CoFeB film may then be continuous and no Ru is in direct contact with the MgO barrier.

As it known, the annealing process would much improve the quality of the electrode/barrier interfaces in a MTJ, and the TMR ratio can be increased largely by increasing T_a. For example, the TMR ratios are 6% and 171% in the as-grown state and at T_a = 375 °C for MgO-based MTJs with t = 3.0 nm [15]. Moreover, the TMR-V curve can become more symmetric (see Fig. 3 in Ref. [15]), which explains the increase of V_C with the increase of T_a in these MgO-based MTJs. In addition, it is found that the TMR-V curve also becomes more symmetric with the increase of t for these FeMn- or IrMn- MTJs.

Sign reversal of the TMR only appears at negative bias for MTJs with $t \leq 1.0$ nm [Fig. 3(a)]. This is thought to be evidence for physical asymmetry of the MTJ sandwich between the top and bottom oxide/ferromagnet interfaces [18], [19]. A strong modification of tunneling density-of-states, which is the pertinent parameter for tunneling, at bottom oxide/ferromagnet interface is likely and the V_C values in Fig. 4 for different pinned CoFeB thickness and T_a suggest pronounced structure in the density of states (DOS) near the Fermi level on one side [14], [17]. Due to the discontinuously thin pinned CoFeB in the SAF stack CoFe/Ru/CoFeB, the sign reversal of polarization may reflect the tunneling DOS provided by the system CoFe/Ru/CoFeB as a whole entity.

4. Conclusion

The diagram of sign reversal of TMR is given as a function of the pinned CoFeB thickness ($0.5 \leq t \leq 1.0$ nm) and annealing temperature ($30 \leq T_a \leq 350$ °C) in MgO-based MTJs. From it, one can achieve the inverted TMR ratio below the V_C and the normal one above the V_C for an MgO-based MTJ at a fixed t and T_a. This may be useful for designing the spintronics devices.

Acknowledgements

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