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Tunneling processes in asymmetric double barrier magnetic tunnel junctions with a thin top MgO layer

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Dynamic conductance dI/dV and inelastic electron tunneling spectroscopy (IETS) d^2I/dV^2 have been measured at different temperatures for double barrier magnetic tunnel junctions with a thin top MgO layer. The resistance in the antiparallel state exhibits a normal tunnel-like behavior, while the resistance in the parallel state shows metallic-like transport, indicating the presence of pinholes in the thin top MgO layer. Three IETS peaks are the zero-bias anomaly, interface magnons, and barrier phonons in both the parallel and antiparallel states. The zero-bias anomaly is the strongest peak in the parallel state and its intensity decreases with temperature. The magnon has the largest intensity in the antiparallel state and its intensity also decreases with temperature. The origins of the dips and peaks in the dI/dV - V curve are also discussed. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4838116>]

INTRODUCTION

High tunneling magnetoresistance (TMR) is now well known in Fe/MgO/Fe and CoFe(B)/MgO/CoFe(B) magnetic tunnel junctions (MTJs).^{1–5} Besides single barrier MTJs (SBMTJs), double barrier MTJs (DBMTJs) have also been extensively studied for the novel physical phenomena and potential applications in spintronic devices.^{6–14} With a thin ferromagnetic (FM) layer sandwiched by two MgO layers, quantum well oscillations and Coulomb blockade phenomena have been reported.^{7,8,12–14} Compared with SBMTJs, DBMTJs have larger $V_{1/2}$ (voltage where TMR decreases to its half maximum value),^{6,9–11} which is desirable for practical applications. Although a lot of studies have been made, the tunneling mechanisms in both SBMTJs and DBMTJs are still unclear.^{15–19}

In this work, we have investigated the tunneling processes in asymmetric DBMTJs with a thin top MgO layer. Tunneling resistances in two magnetic states, dynamic conductance dI/dV - V and inelastic electron tunneling spectroscopy (IETS) d^2I/dV^2 - V , have been measured as a function of temperature to clarify the tunneling mechanisms. IETS has proven to be an effective method to detect elementary excitations, including the zero-bias anomaly, magnons at ferromagnet/insulator interface, and barrier phonons in MTJs.^{20–22}

EXPERIMENT

Asymmetric DBMTJ stacks were deposited on thermally oxidized Si wafers using the high vacuum Shamrock cluster deposition tool: Ta (5)/Ru (30)/Ta (5)/Ni₈₁Fe₁₉ (5)/Ir₂₂Mn₇₈ (10)/Co₉₀Fe₁₀ (2.5)/Ru (0.9)/Co₄₀Fe₄₀B₂₀ (CoFeB) (3)/MgO (2)/Co₅₀Fe₅₀ (CoFe) (2)/CoFeB (1)/MgO (0.8)/CoFeB (3)/Ru

(0.9)/Co₉₀Fe₁₀ (2.5)/Ir₂₂Mn₇₈ (10)/Ni₈₁Fe₁₉ (5)/Ta (5)/Ru (5) (all thicknesses in nm). For reference, symmetric DBMTJs with the structure of Ta (5)/Ru (30)/Ta (5)/Ni₈₁Fe₁₉ (5)/Ir₂₂Mn₇₈ (10)/Co₉₀Fe₁₀ (2.5)/Ru (0.9)/CoFeB (3)/MgO (2)/CoFe (2)/CoFeB (1)/MgO (2)/CoFeB (3)/Ru (0.9)/Co₉₀Fe₁₀ (2.5)/Ir₂₂Mn₇₈ (10)/Ni₈₁Fe₁₉ (5)/Ta (5)/Ru (5) (all thicknesses in nm) were also prepared, see Fig. 1. All samples were patterned into junctions with the size of 5×10 or $10 \times 10 \mu\text{m}^2$ using ultraviolet photolithography and Ar ion beam etching, and then annealed at 350 °C under an external in-plane magnetic field of 800 mTorr for half an hour. The magnetoelectric transport properties were measured in a Quantum Design Physical Properties Measurement System (PPMS) using the conventional four-terminal DC method. The dynamic conductance dI/dV and inelastic electron tunneling spectroscopy d^2I/dV^2 were measured using the standard lock-in method with an AC modulation voltage of 4 mV and frequency of 30.79 Hz.

RESULTS AND DISCUSSION

The structures of asymmetric and symmetric DBMTJs were sketched in Figs. 1(a) and 1(b), respectively. For asymmetric DBMTJ, top MgO layer is so thin that it becomes discontinuous with inevitable pinholes^{17,23} forming in the MgO layer, where two CoFeB layers have direct contact, as shown in Fig. 1(a). However, when MgO layer becomes thick enough, it is continuous and therefore no pinholes emerge within MgO layer, as indicated in Fig. 1(b).

As indicated in Figs. 2(a) and 2(b), TMR ratio at 300 K for symmetric and asymmetric DBMTJs can reach as high as 215% and 176%, respectively. The middle free layer of asymmetric DBMTJ strongly couples with top CoFeB pinned layer due to thin top MgO layer and thus shows gradual switching. By contrast, the switching of the middle free layer of symmetric DBMTJ is very sharp. As shown in

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FIG. 1. The structures for (a) asymmetric and (b) symmetric DBMTJs.

Figs. 2(c) and 2(d), the resistance in the antiparallel state R_{AP} for both symmetric and asymmetric DBMTJs displays a rapid increase with decreasing temperature, while the resistance in the parallel state R_P is weakly temperature dependent, which indicates high quality MgO²⁴ results in the increasing of TMR with decreasing temperature. The temperature dependence of R_P and R_{AP} is a result of competition between magnetic disorder and thermal excitation. For symmetric DBMTJ, R_P shows tunnel-like transport. But for asymmetric DBMTJ, R_P exhibits metallic-like transport, indicating the presence of pinholes^{17,23} in the thin top MgO layer, as indicated in Fig. 1(a).

According to Shang's model,¹⁵ the dependence of G_P (conductance in the parallel state) and G_{AP} (conductance in the antiparallel state) on temperature in MTJs can be expressed as

$$G_{P,AP}(T) = G_T(1 \pm P_1(T)P_2(T)) + sT^{1.33}, \quad (1)$$

where + and – correspond to the parallel (P) and antiparallel (AP) configurations, respectively. The first term in Eq. (1) represents spin-dependent elastic tunneling conductance,

where $G_T = G_0(CT)/\sin(CT)$ with G_0 for zero-temperature conductance and $C = 1.387 \times 10^{-4}d/\phi^{1/2}$ for the strength of electronic thermal smearing near Fermi level related to barrier thickness d in Angstrom and barrier height ϕ in eV, and $P(T) = P_0(1 - \alpha T^{3/2})$ with P_0 for spin polarization of ferromagnetic electrode at $T = 0$ K, where α is a material-dependent constant. The second term in Eq. (1) denotes spin-independent hopping conductance through chains of two localized states in the barrier, where s is a prefactor associated with the density of two localized states in the barrier. In total, there are five parameters G_0 , C , P_0 , α , and s , which can be obtained by fitting the $G_{P,AP}(T)$ and TMR(T) curves. From the TMR(T) curve, P_0 and α can be simultaneously obtained. G_0 and C are determined from the $\Delta G(T)$ (defined as $\Delta G(T) = G_P(T) - G_{AP}(T)$) curve, while s is obtained by fitting the $G_{P,AP}(T)$ curves. The fitting results of G_0 , C , P_0 , α , and s for symmetric and asymmetric DBMTJs are summarized in Table I.

G_0 for asymmetric DBMTJ is smaller than that for symmetric DBMTJ because of the larger junction size, $10 \times 10 \mu\text{m}^2$ and $5 \times 10 \mu\text{m}^2$ for symmetric and asymmetric DBMTJs, respectively. Moreover, stronger scattering may occur for asymmetric DBMTJ due to the existence of pinholes in the thin top MgO layer. C for asymmetric DBMTJ is less than that for symmetric DBMTJ due to the thin top MgO layer in asymmetric DBMTJ. P_0 for symmetric DBMTJ is almost the same as that for asymmetric DBMTJ, which is nearly independent of the MgO layers. Generally, α is larger for surface than bulk, and sensitive to interface roughness.²⁵ α for asymmetric DBMTJ is larger than that for symmetric DBMTJ. This indicates that there is more disorder and the interface is much rougher in asymmetric DBMTJ caused by the discontinuous top MgO layer. s/G_0 represents the ratio of the spin-independent part to the spin-dependent part; its value is $3.34 \times 10^{-7} \text{ S}^{-1} \text{ K}^{-1.33}$ and $7.80 \times 10^{-7} \text{ S}^{-1} \text{ K}^{-1.33}$ for symmetric and asymmetric DBMTJs, respectively. The contribution from pinholes in the thin top MgO layer is spin-independent, causing the larger s/G_0 in asymmetric DBMTJ.

FIG. 2. TMR ratio versus magnetic field H for (a) symmetric and (b) asymmetric DBMTJs at 300 K. T dependence of R_P , R_{AP} , and TMR ratio for (c) symmetric and (d) asymmetric DBMTJs. Solid lines are fit results according to Shang's model (Ref. 15).

TABLE I. Fitting parameters G_0 , C , P_0 , α , and s for symmetric and asymmetric DBMTJs according to Eq. (1).

MTJ type	G_0 (10^{-3} S)	C (10^{-3} K $^{-1}$)	P_0 (%)	α (10^{-5} K $^{-3/2}$)	s (10^{-10} K $^{-1.33}$)
Symmetric DBMTJ	1.09	2.15	74.9	0.64	3.64
Asymmetric DBMTJ	0.69	1.38	76.7	2.06	5.38

FIG. 3. IETS d^2I/dV^2 versus voltage V for asymmetric and symmetric DBMTJs in the P (a) and AP (b) states at 10 K, and for asymmetric DBMTJ in the P (c) and AP (d) states at different T . ZBA, M, and Ph mark the positions of zero-bias anomaly, interface magnons, and barrier phonons, respectively.

Fig. 3 gives the IETS of asymmetric and symmetric DBMTJs. For MgO-based SBMTJs, the zero-bias anomaly (ZBA), magnon (M), and phonon (Ph) peaks are usually located at $V \leq 15$ mV, $V = 20$ – 35 mV, and $V \approx 81$ mV, respectively.^{21,22} The origins of these peaks are commonly considered to be magnetic impurities, interface magnons, and barrier phonons, respectively.^{21,22} In the P state of asymmetric and symmetric DBMTJs measured at 10 K, ZBA, M, and Ph peaks are at 5 meV/7 meV, 37 meV/58 meV, and 84 meV/144 meV for asymmetric/symmetric DBMTJ, as indicated in Fig. 3(a). The voltage values of these characteristic peaks of symmetric DBMTJ are about twice as great as those of SBMTJ, while the voltage values of these peaks of asymmetric DBMTJ are only a bit larger than those of SBMTJ due to the direct contact between the middle free layer and the top pinned CoFeB layer, as illustrated in Fig. 1(a). As shown in Fig. 3(b), the case for the AP state is similar to that for the P state. ZBA, M, and Ph peaks are at 8 meV/8 meV, 28 meV/34 meV, and 82 meV/120 meV for asymmetric/symmetric DBMTJ.

Figs. 3(c) and 3(d) show the P and AP state IETS for asymmetric DBMTJ at different temperatures. For the P state, ZBA is the strongest peak, while M has the largest intensity for the AP state. The temperature dependence of ZBA voltage position and intensity for the P state of asymmetric DBMTJ, and M voltage position and intensity for the AP state of asymmetric DBMTJ are plotted in Fig. 4. The intensities of both ZBA and M decrease with increasing temperature, when the voltage positions of ZBA and M move towards high voltage. With increasing temperature from 2 K

to 150 K, ZBA voltage position and intensity for the P state vary from 5 meV to 53 meV and from 1.65 mA/V 2 to 0.26 mA/V 2 , while those of M for the AP state change from 27 meV to 82 meV and from 1.37 mA/V 2 to 1.15 mA/V 2 . With increasing temperature, both ZBA and M become weak and even disappear owing to thermal excitation considering

FIG. 4. T dependence of (a) ZBA voltage position V_{position} and intensity in the P state and (b) M voltage position V_{position} and intensity in the AP state for asymmetric DBMTJ.

FIG. 5. Dynamic conductance dI/dV versus voltage V for asymmetric and symmetric DBMTJs in the P (a) and AP (b) states at 10 K, and for asymmetric DBMTJ in the P (c) and AP (d) states at different T . D1 and D2 mark the positions of dI/dV minima in the P state, while P1 and P2 correspond to the positions of dI/dV maxima. D labels the position of dI/dV minimum in the AP state.

that $k_B T$ is equal to 26 meV at 300 K with k_B as Boltzmann constant.

The dynamic conductance dI/dV - V for asymmetric and symmetric DBMTJs is plotted in Fig. 5. Figs. 5(a) and 5(b) display dI/dV in the P and AP states for asymmetric and symmetric DBMTJs measured at 10 K, respectively. In the P state of asymmetric and symmetric DBMTJs, there are two dips marked by D1 and D2, and two peaks marked by P1 and P2, while there is only one dip located at $V=0$ meV in the AP state and marked by D. For asymmetric DBMTJ, D1, P1, P2, and D2 are at $V=0$, 44, 120, and 368 meV, while they are located at $V=0$, 61, 185, and 628 meV for symmetric DBMTJ. The voltage positions of D1, P1, and P2 for asymmetric and symmetric DBMTJs in the P state agree very well with those of ZBA, M, and Ph in the P state IETS, while D voltage position in the AP state coincides with ZBA voltage position in the AP state IETS. For the P state, D1 originates from magnetic impurities scattering and is the so-called ZBA. Interface magnons and barrier phonons-assisted inelastic tunneling enhances conductance and causes the maxima of dI/dV at P1 and P2. Considering that the top of majority spin Δ_2' , and Δ_5 lies 0.2 V above E_F for Fe and the top of minority spin Δ_2 lies 0.3 V above E_F for Co,^{19,26} electrons across the barrier need to overcome the energy of the top of the bands, and thus dI/dV in the P state decreases and shows a dip at D2. The band structure of CoFe electrode is therefore identified as the possible reason for D2. D in the AP state shown in Fig. 5(b) is identified as ZBA. As indicated in Figs. 5(c) and 5(d), D1 and D become wider with increasing temperature, P1 disappears above 50 K, and P2 and D2 only show weak temperature dependence.

CONCLUSIONS

In conclusion, tunneling processes in asymmetric and symmetric DBMTJs have been studied in this work. Unlike tunneled transport in symmetric DBMTJ, R_P presents a

metallic-like behavior revealing the influence of pinholes, where two CoFeB layers make direct contact in asymmetric DBMTJ due to the thin top MgO layer. Three peaks originating from zero-bias anomaly, interface magnons, and barrier phonons are simultaneously observed in the IETS at low temperature for both asymmetric and symmetric DBMTJs. For asymmetric DBMTJ, ZBA in the P state has the largest intensity and its voltage position shifts towards high voltage when its intensity decreases with increasing temperature; M in the AP state is the strongest peak, and its voltage position also moves to high voltage as its intensity weakens when temperature is increased. Two dips originating from zero-bias anomaly and band structure of CoFe electrode, and two peaks caused by interface magnons and barrier phonons are simultaneously observed in the P state, while there is only one dip identified as zero-bias anomaly in the AP state of the dI/dV - V curve at low temperature for both asymmetric and symmetric DBMTJs.

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