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Critical role of interface states for spin-dependent tunneling in half-metallic Co₂MnSi-based magnetic tunnel junctions investigated by tunneling spectroscopy

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We investigated at 4.2 K the differential conductance (dI/dV) versus V characteristics of fully epitaxial Co₂MnSi/MgO/Co₂MnSi magnetic tunnel junctions (MTJs) featuring high tunnel magnetoresistance ratios of about 700% at 4.2 K (about 180% at room temperature). We developed a tunneling model to explain the observed tunneling spectra and showed the critical role played by interface states for minority spins existing around the Fermi level of Co₂MnSi electrodes facing a MgO tunnel barrier in the spin-dependent tunneling characteristics of these MTJs with half-metallic electrodes. © 2009 American Institute of Physics. [DOI: 10.1063/1.3083560]

Applying spin-polarized electrons is essential to realize spintronic devices. Half-metallic ferromagnets are characterized by an energy gap for one spin direction at the Fermi level (E_F), leading to a complete spin polarization at E_F .¹ Recently, potentially half-metallic Co-based full-Heusler alloys (Co₂YZ) (Refs. 2–4) attracted much interest for use as ferromagnetic electrodes of spintronic devices. One Co-based full-Heusler alloy in particular, Co₂MnSi (CMS), attracted interest^{5–8} because of its theoretically predicted half-metallic nature, with a large energy gap of 0.42 eV (Ref. 2) to 0.81 eV (Ref. 3) for its minority-spin (m-spin) band, and because of its high Curie temperature (T_C) of 985 K. Chioncel *et al.*⁹ attributed the strong temperature (T) dependence of the tunnel magnetoresistance (TMR) ratio observed for CMS/AIO_x/CMS magnetic tunnel junctions (MTJs) (Ref. 6) to a strong T dependence of the CMS spin polarization arising from so called nonquasiparticle states. On the other hand, Ležaić *et al.*¹⁰ took into consideration the dependence of the hybridization of states forming the half-metal gap on thermal spin fluctuations and predicted that the CMS spin polarization remained close to 100% up to $0.27 T_C \sim 270$ K, then quickly decreased beyond that. Mavropoulos *et al.*¹¹ pointed out, based on theoretical arguments, the crucial role of interface states of half-metallic electrodes facing a tunnel barrier for spin-dependent tunneling in half-metal-based MTJs.

We recently fabricated fully epitaxial MTJs with CMS thin films as both lower and upper electrodes and with a MgO barrier (CMS/MgO/CMS MTJs), and from these MTJs obtained high TMR ratios of up to 705% at 4.2 K and 182% at room temperature (RT).^{8,12} Our purpose in the present study was to clarify the key tunneling mechanism that dominates the spin-dependent tunneling characteristics of MTJs with potentially half-metallic electrodes, in particular, CMS/MgO/CMS MTJs, through tunneling spectroscopy measurements.

The fabricated MTJ layer structure was as follows: (from the substrate side) MgO buffer (10 nm)/CMS lower electrode (50 nm)/MgO tunnel barrier (2–3 nm)/CMS upper electrode

(5 nm)/Ru (0.8 nm)/Co₉₀Fe₁₀ (2 nm)/IrMn (10 nm)/Ru cap (5 nm), grown on a single-crystalline MgO (001) substrate. The preparation of the fully epitaxial CMS/MgO/CMS MTJs is described in detail elsewhere.⁸ The film composition of CMS films used in this study was Co_{2.0}Mn_{0.91}Si_{0.93}, as determined by inductively coupled plasma analysis. The fabricated junction size was $10 \times 10 \mu\text{m}^2$. The dI/dV spectrum was measured using a conventional lock-in method at 317 Hz with a typical modulation peak-to-peak voltage of 10 mV. The second derivative d^2I/dV^2 was obtained mathematically from the dI/dV spectrum. The bias voltage (V) was defined with respect to the lower CMS electrode.

Figure 1 shows typical dI/dV ($=G$) and corresponding d^2I/dV^2 spectra at 4.2 K for parallel (P) and antiparallel (AP) alignments for a fabricated CMS/MgO/CMS MTJ; this MTJ showed a high TMR ratio of 679% at 4.2 K (176% at RT). A marked increase in G_{AP} with increasing V was clearly observed in the small bias region of $|V| \leq 70$ mV at 4.2 K [Figs. 1(a) and 1(d)]. This clear structure almost disappeared at RT. Corresponding to the marked increase in G_{AP} in the small bias region at 4.2 K, pronounced peak structures in the d^2I/dV^2 spectrum with peaks at ± 4 mV were observed for AP [inset of Fig. 1(d)]; these indicated magnon excitation in the collector caused by tunnel electrons with excess energies above E_F (i.e., hot electrons).¹³ Beyond $|V| \sim 70$ mV, G_{AP} showed a weak dependence on V for $|V| \leq 0.20$ V and G_{AP} increased sharply at almost equal characteristic voltages V_{C1} of $\sim \pm 0.20$ V for both polarities. The overall G_P spectrum shows a very weak dependence on V up to V of ± 0.36 V [Fig. 1(a)]. This feature can be more clearly seen in the normalized G_P spectrum [Fig. 1(b)]. G_P also increased sharply at almost equal characteristic voltages V_{C2} of ± 0.36 V for both polarities. The sharp increases in G_{AP} and G_P at V_{C1} of ± 0.20 V and V_{C2} of ± 0.36 V, respectively, indicate the existence of half-metal gaps for the lower and upper CMS electrodes. We deduced from these characteristics an almost equal value of 0.20 eV for E_V (the energy difference from E_F to the top of the m-spin valence band) and E_C (that from the bottom of the m-spin conduction band to E_F) for the lower CMS and a value of 0.16 eV for both E_V and E_C for the upper CMS,¹² showing that the lower and upper CMS elec-

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FIG. 1. (Color online) Typical $dI/dV (=G)$ vs V characteristics (G spectra) and corresponding d^2I/dV^2 spectra for the parallel (P) and antiparallel (AP) alignments at 4.2 K for a $\text{Co}_2\text{MnSi}/\text{MgO}$ (2.2 nm)/ Co_2MnSi MTJ. The bias voltage (V) is defined with respect to the lower Co_2MnSi electrode: (a) G_P and G_{AP} spectra for $-0.6 \text{ V} < V < 0.6 \text{ V}$. (b) Corresponding normalized G_P and G_{AP} spectra, where G_P and G_{AP} are normalized by their respective values at $V=0$. (c) G_P spectrum for $-0.2 \text{ V} < V < 0.2 \text{ V}$. The inset shows the corresponding d^2I/dV^2 spectrum. (d) G_{AP} spectrum for $-0.2 \text{ V} < V < 0.2 \text{ V}$. The inset shows the corresponding d^2I/dV^2 spectrum.

trodes each featured a half-metal gap of 0.40 and 0.32 eV, respectively, with E_F located near the middle of the half-metal gap in both cases.¹²

In more detail, however, the G_P spectrum at a small bias, especially for $|V| < 50 \text{ mV}$, was notably asymmetric regarding the bias polarity [Fig. 1(c)]. The G_P spectrum for $V < 0$, where electrons tunnel from the upper CMS to the lower CMS, showed a clear increase with increasing V up to $\sim -30 \text{ mV}$, but the G_P spectrum for $V > 0$ was almost completely independent of V up to $V \sim 50 \text{ mV}$ (i.e., the d^2I/dV^2 values were nearly zero for this region). The corresponding d^2I/dV^2 spectrum for P demonstrated a clear peak structure with a peak at -4 mV only for $V < 0$ [inset of Fig. 1(c)], indicating magnon excitation caused by tunnel electrons with excess energies for P and $V < 0$.

We will now discuss the spin-dependent tunneling characteristics that appeared with a small bias of $|V| < 100 \text{ mV}$. The most important characteristics that should be clarified are the pronounced asymmetric behaviors of the G_P and d^2I/dV^2 spectra for P. The model of spin-dependent electronic structures for the lower and upper CMS electrodes that we propose to explain the G and d^2I/dV^2 spectra is illustrated in Fig. 2. The observed characteristics for P can reasonably be understood by assuming (1) the existence of interface states around E_F in the m-spin band gap in the interfacial region of the lower CMS facing a MgO barrier and (2) the existence of residual states around E_F in the m-spin band gap in the bulk region of the upper CMS in addition to interface states for the upper CMS.

First, we consider a possible tunneling process for P. The very weak dependence of G_P on V up to V_{C2} of $\pm 0.36 \text{ V}$ suggests that the tunneling conductance for P is quite high

FIG. 2. (Color online) Schematic of a model of spin-dependent electronic structures for the lower and upper Co_2MnSi (CMS) electrodes in CMS/MgO/CMS MTJs. Here, IS represents interface states for m-spins and RS represents residual states for m-spins in the bulk region of the upper CMS electrode. M and m refer to M- and m-spins, respectively. A schematic for a tunneling process for the P alignment at a small negative bias (electrons tunnel from the upper CMS to the lower CMS) is also plotted.

due to the coherent tunneling contribution^{14,15} from the majority-spin (M-spin) Δ_1 channel of CMS in CMS/MgO/CMS, which has been theoretically predicted,¹⁶ and so the contribution of tunneling involving the inelastic process in the electrodes would be relatively small. If we take into consideration the monotonic dispersion relation of the energy E versus the wave number k_z along $[001]$ from Γ to X near E_F for CMS,^{3,16} it is improbable that the asymmetric behavior observed in the G_P spectrum in the small bias region was due to tunneling from the M-spin Δ_1 channel to the M-spin Δ_1 channel. Furthermore, since E_F is near the middle of the half-metal gap for both the lower and upper CMS, we cannot attribute the increase of G_P for $V < 0$ with increasing V up to -30 mV to direct band-to-band tunneling from the m-spin valence band of the upper CMS to the m-spin conduction band of the lower CMS. Accordingly, we attribute the observed asymmetric behavior for P in the small V region to tunneling from the m- to m-spin states, both of which are located around E_F . The most probable candidate to explain these m-spin states located around E_F is interface states. The observed increase in G_P with increasing V for $V < 0$ up to $V \sim -30 \text{ mV}$, however, requires one or more sources to supply electrons to interface states in the upper CMS. There are two possible sources: one is possible residual states with a small density of states existing around E_F in the m-spin band gap in the bulk region of the upper CMS. Electrons would then be supplied from m-spin residual states to m-spin interface states. The other possible source is spin-flip scattering of electrons from the M-spin band to possible m-spin interface states in the upper CMS that is caused by thermally excited magnons. This second possibility is improbable, however, because if such a source is available in the upper CMS, it should be equally available in the lower CMS, which is contradictory to the experimental result of G_P being almost completely independent of V up to $V \sim 50 \text{ mV}$. Thus, the validity of the first possibility only is reasonably justified.

A possible tunneling process from the m- to m-spin states for $V < 0$ is also illustrated in Fig. 2, where electrons in m-spin residual states located around E_F in the half-metal gap in the bulk region of the upper CMS are supplied to m-spin interface states located around E_F in the interfacial region of the upper CMS, and then tunnel into m-spin interface states located around E_F of the lower CMS. This tun-

neling process cannot contribute to the tunneling conductance without spin-flip scattering in the lower CMS because there is no available state for m-spins around E_F in the bulk region of the lower CMS. Tunnel electrons with excess energies, however, excite magnons in the interfacial region of the lower CMS electrode. The tunneling electrons are then spin-flip scattered to the M-spin band in the lower CMS electrode and contribute to the tunnel conductance. This picture combines the concept of electrons tunneling into interface states¹¹ with that of magnon excitation by hot electrons in the collector.¹³ The existence of residual states in the m-spin band gap for the upper CMS may be related to a possible lower structural quality of the 5-nm-thick upper CMS thin film that was grown on a 2–3-nm-thick MgO barrier.

On the other hand, the almost constant G_P values for a small positive bias region up to ~ 50 mV indicate that there was no residual state around E_F in the m-spin band gap in the bulk region of the lower CMS electrode. In the absence of such a state, electrons would not be supplied to m-spin interface states from the bulk region in the lower CMS and G_P values would be almost constant for the small positive bias region.

The origin of the sharp increase in G_{AP} with increasing V in the small bias region of $|V| \leq 70$ mV can be consistently explained with the same model of spin-dependent electronic structures for the lower and upper CMS electrodes. Note that E_F lies near the middle of the half-metal gap and the estimated values of E_C were 0.20 eV for the lower CMS and 0.16 eV for the upper CMS. Thus, it is highly unlikely that the marked increase in G_{AP} in the small bias region of $|V| \leq 70$ mV at 4.2 K was due to direct tunneling from the M-spin band to the unoccupied m-spin conduction band. It is more reasonable to ascribe the marked increase in G_{AP} to $|V| \leq 70$ mV at 4.2 K to the following tunneling process: (1) Electrons tunnel from the M-spin band of the upper CMS to m-spin interface states of the lower CMS (here, we consider a tunneling process for $V < 0$). (2) Tunnel electrons excite magnons in the interfacial region due to their excess energies above E_F in the lower CMS and simultaneously electrons are spin-flip scattered to the M-spin band in the lower CMS, contributing to the marked increase in G_{AP} . The tunneling process for $V > 0$ is essentially the same.

The existence of interface states for m-spins around E_F in the interfacial region of CMS electrodes facing a MgO barrier has recently been predicted theoretically through first-principles calculations of electronic structures,¹⁷ which is consistent with our experimental finding.

Figure 3 shows R_P and R_{AP} , as well as the TMR ratio, as a function of T from 4.2 K to RT for a CMS/MgO/CMS MTJ (the same MTJ as for Fig. 1), where R_P and R_{AP} are the respective tunnel resistances for P and AP. R_{AP} decreased with increasing T from 4.2 K to RT, while R_P showed almost complete independence on T from 4.2 K to RT. It is clear that the R_{AP} dependence on T determined the T dependence of the TMR ratio. With increasing T , spin-flip scattering by thermally excited magnons from the M-spin band to m-spin interface states in the emitter and from m-spin interface states to the M-spin band in the collector would increase the tunneling conductance for AP. This picture was originally discussed by Mavropoulos *et al.*¹¹ We also ascribe the almost complete independence of R_P with respect to T to the high tunnel conductance for P, as for the very weak dependence of

FIG. 3. (Color online) R_{AP} and R_{AP} , as well as the TMR ratio, as a function of temperature from 4.2 K to RT for a $\text{Co}_2\text{MnSi}/\text{MgO}$ (2.2 nm)/ Co_2MnSi MTJ (the same MTJ as for Fig. 1), where R_P and R_{AP} are the respective resistance-area products for P and AP.

G_P on V up to V_{C2} of ± 0.36 V due to the coherent tunneling contribution from the M-spin Δ_1 channel.

In conclusion, we experimentally found strong evidence for the existence of interface states for m-spins around E_F in the interfacial region of Co_2MnSi electrodes facing a MgO barrier. We demonstrated the critical role played by interface states for spin-dependent tunneling in these half-metallic Co_2MnSi -based MTJs, through spin-dependent tunneling spectroscopy.

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