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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 \(\text{Co}_2\text{MnSi}/\text{MgO}/\text{Co}_2\text{MnSi}\) 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 \(\text{Co}_2\text{MnSi}\) electrodes facing a \(\text{MgO}\) 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 \(E_F\). Recently, potentially half-metallic Co-based full-Heusler alloys \((\text{Co}_2\text{YZ})\) (Refs. 2–4) attracted much interest for use as ferromagnetic electrodes of spintronic devices. One Co-based full-Heusler alloy in particular, \(\text{Co}_2\text{MnSi}\) (CMS), attracted interest5–8 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/\(\text{AlO}_x/\text{CMS}\) magnetic tunnel junctions (MTJs) (Ref. 6) to the 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 \(T_C\) ~ 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).10,12 Our purpose in the present study was to clarify the 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)/Co_{90}Fe_{10} (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 \(\text{Co}_2\text{Mn}_{0.93}\text{Si}_{0.07}\), 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 \(G_{\text{AP}}\) with increasing \(V\) was clearly observed in the small bias region of \(|V|\approx 70 \text{ mV}\) at 4.2 K [Figs. 1(a) and 1(d)]. This clear structure almost disappeared at RT. Corresponding to the marked increase in \(G_{\text{AP}}\) in the small bias region at 4.2 K, pronounced peak structures in the \(d^2I/dV^2\) spectrum with peaks at \(\pm 4 \text{ mV}\) 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 \(|V|\approx 70 \text{ mV}, G_{\text{AP}}\) showed a weak dependence on \(V\) for \(|V|\leq 0.20 \text{ V}\) and \(G_{\text{AP}}\) increased sharply at almost equal characteristic voltages \(V_{C_P}\) of \(\approx 0.20 \text{ V}\) for both polarities. The overall \(G_P\) spectrum shows a very weak dependence on \(V\) up to \(V\) of \(\approx 0.36 \text{ V}\) [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 \(V_{C_P}\) of \(\approx 0.36 \text{ V}\) for both polarities. The sharp increases in \(G_{\text{AP}}\) and \(G_P\) at \(V_{C_P}\) of \(\approx 0.20 \text{ V}\) and \(V_{C_P}\) of \(\approx 0.36 \text{ V}\), 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-
The bias voltage \( V \) is defined with respect to the lower Co\(_2\)MnSi electrode: (a) \( G_P \) and \( G_{AP} \) spectra for \(-0.6 \, V < V < 0.6 \, V \). (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 \(-0.2 \, V < V < 0.2 \, V \). The inset shows the corresponding \( dI/dV^2 \) spectrum. (d) \( G_{AP} \) spectrum for \(-0.2 \, V < V < 0.2 \, V \). The inset shows the corresponding \( dI/dV^2 \) spectrum.

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

FIG. 2. (Color online) Schematic of a model of spin-dependent electronic structures for the lower and upper Co\(_2\)MnSi (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) \( \Delta_1 \) 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 \( k \), along [001] from \( \Gamma \) 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 \( \Delta_1 \) channel to the M-spin \( \Delta_1 \) 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 \(-30 \, mV \), 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 \( V < -50 \, mV \). 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-
tunneling 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 states$^{11}$ 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 $\sim 50$ 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 $E_F$ 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 $G_P$ on $V$ up to $V_{C2}$ of $\pm 0.36$ V due to the coherent tunneling contribution from the M-spin $\Delta_1$ 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$_2$MnSi electrodes facing a MgO barrier. We demonstrated the critical role played by interface states for spin-dependent tunneling in these half-metallic Co$_2$MnSi-based MTJs, through spin-dependent tunneling spectroscopy.

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