# Critical role of interface states for spin-dependent tunneling in half-metallic $Co_2$MnSi-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 $Co_2MnSi/MgO/Co_2MnSi$ 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 $Co_2$MnSi 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 ($Co_2YZ$) (Refs. 2–4) attracted much interest for use as ferromagnetic electrodes of spintronic devices. One Co-based full-Heusler alloy in particular, $Co_2$MnSi (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/$AlO_x$/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)/$Co_{90}Fe_{10}$ (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 $Co_{2.0}Mn_{0.91}Si_{0.93}$, as determined by inductively coupled plasma analysis. The fabricated junction size was $10 \times 10$ $\mu m^2$. 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 $\pm 4$ 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 $\pm 0.36$ 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 $\pm 0.36$ V for both polarities. The sharp increases in $G_{AP}$ and $G_P$ at $V_{C1}$ of $\pm 0.20$ V and $V_{C2}$ of $\pm 0.36$ 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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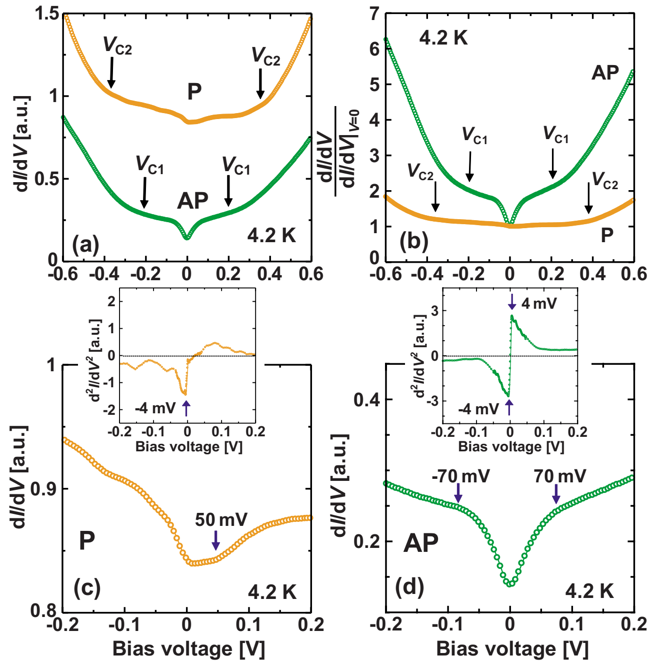


FIG. 1. (Color online) Typical $dI/dV$ ($=G$) vs $V$ characteristics ($G$ spectra) and corresponding $d^2I/dV^2$ spectra for the parallel (P) and antiparallel (AP) alignments at 4.2 K for a $Co_2MnSi/MgO$ (2.2 nm)/$Co_2MnSi$ MTJ. The bias voltage ($V$) is defined with respect to the lower $Co_2MnSi$ electrode: (a) $G_P$ and $G_{AP}$ spectra for $-0.6\ V<V<0.6\ V$. (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 $d^2I/dV^2$ spectrum. (d) $G_{AP}$ spectrum for $-0.2\ V<V<0.2\ V$. 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 $|V|<50$ 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 $\sim -30$ mV, but the $G_P$ spectrum for $V>0$ was almost completely independent of $V$ up to $V\sim 50$ 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 $|V|<100$ 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 $\pm 0.36$ V suggests that the tunneling conductance for P is quite high

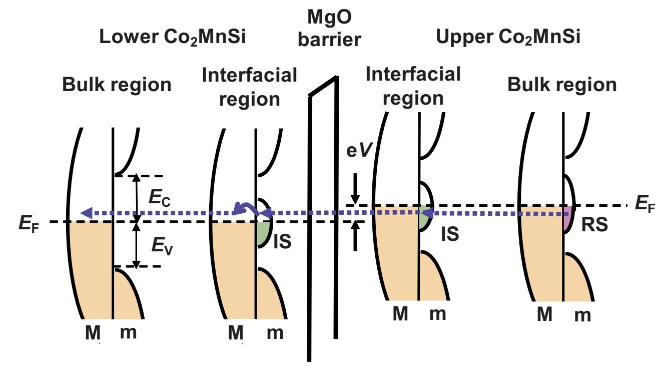


FIG. 2. (Color online) Schematic of a model of spin-dependent electronic structures for the lower and upper $Co_2MnSi$ (CMS) electrodes in CMS/MgO/CMS MTJs. Here, IS represents interface states for m-spins and RS represents residual states for m-spins in the bulk region of the upper CMS electrode. M 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) $\Delta_1$ 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 $k_z$ 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 $V\sim -30$ 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 $V\sim 50$ 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 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 $E_C$ 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

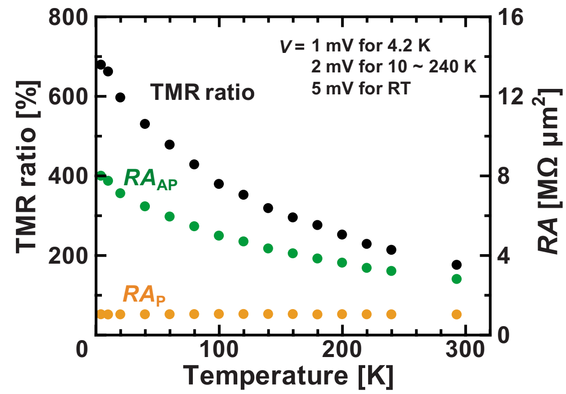


FIG. 3. (Color online) $RA_P$ and $RA_{AP}$, as well as the TMR ratio, as a function of temperature from 4.2 K to RT for a $Co_2MnSi$/MgO (2.2 nm)/$Co_2MnSi$ MTJ (the same MTJ as for Fig. 1), where $RA_P$ and $RA_{AP}$ are the respective resistance-area products for P and AP.

$G_P$ 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 $Co_2MnSi$ 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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