Nonquasiparticle States in Co₂MnSi Evidenced through Magnetic Tunnel Junction Spectroscopy Measurements

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We investigate the effects of electronic correlations in the full-Heusler Co₂MnSi, by combining a theoretical analysis of the spin-resolved density of states with tunneling-conductance spectroscopy measurements using Co₂MnSi as electrode. Both experimental and theoretical results confirm the existence of so-called nonquasiparticle states and their crucial contribution to the finite-temperature spin polarization in this material.

Next generation electronic devices will profit from technologies that control the spin degrees of freedom. Therefore, half-metallic ferromagnets (HMF), in which the majority-spin-density of states (DOS) crosses the Fermi level (E_F) while the minority-spin DOS shows a semiconducting behavior at E_F (or vice versa), can be seen as essential components for tunneling magnetoresistance (TMR) devices. In HMF-based TMR devices the magnetoresistance should, ideally, diverge if the conduction electron spins are 100% spin polarized. It is further expected that if such a HMF could be epitaxially grown on a semiconducting surface, fully polarized (100%) electrons could be possibly injected into the semiconductor.

First band-structure calculations performed on the half-Heusler, NiMnSb ($C1_b$-type structure), predicted a 100% spin polarization [1]. Subsequently, the studies were extended to other half-metallic systems. Of particular interest for realizing magnetic tunneling junctions (MTJ) appears to be the full-Heusler alloy Co₂MnSi ($L2_1$-type structure). A minority-spin band gap of 0.4 eV has been predicted and a Curie temperature of $T_c = 985$ K and a saturation magnetization of $5\mu_B$ was reported [2].

Some of us [3] recently fabricated magnetic tunnel junctions (MTJ) consisting of highly ordered Co₂MnSi epitaxial bottom electrode, Al-O tunnel barrier, and Co₇₅Fe₂₅ top electrode. A TMR ratio of 159%, at low temperatures and a value of 70% at room temperature, was determined. More recently, MTJ structures consisting of Co₂MnSi/Al-O/Co₂MnSi were fabricated, having a TMR ratio of 570%, at 2 K, the largest one reported to date for an Al-O amorphous tunneling barrier [4]. These experiments reveal the HMF character of Co₂MnSi with a minority-spin band gap and a high decrease of TMR ratio with temperature [5].

In order to understand the large temperature variation of the TMR ratio in Co₂MnSi, it is important to investigate the temperature dependence of its half-metallic density of states in the presence of electron-electron interaction. Zero-temperature band-structure calculations within the framework of density functional theory (DFT) were reported by Galanakis *et al.* [6]. According to these calculations, the half-metallic character of Co₂MnSi is determined by the existence of a minority-spin gap formed between the triply degenerate Co-Co antibonding t_{2g} and the double degenerate Co-Co antibonding e_g [6] bands. However, standard DFT—local density approximation (LDA) calculations are in general insufficient to describe some important many-body features of HMF, at zero or finite temperatures. One of these effects is the appearance of so-called nonquasiparticle (NQP) states, i.e., states appearing within the minority-spin band gap just above the Fermi level. These states describe low-energy electron excitations for minority spins, which turn out to be possible as superpositions of spin-up electron excitations and virtual magnons [7–9] ("spin-polaron") processes. Therefore, their description require an appropriate treatment of dynamical many-body effects. NQP states have been studied in several half-metals [10–14] by using a combined LDA + DMFT (dynamical mean field theory) approach (for a review, see Ref. [15]). NQP states contribute significantly to the tunneling transport in heterostructures based on HMF [16–18]. At $T = 0$ K, the density of NQP states vanishes at the Fermi level, E_F (in the presence of spin anisotropy at a slightly higher energy $E_F + \hbar\omega_m$, see below), while for $T > 0$ tails of the NQP states cross the Fermi energy and contribute to the depolarization.

In this Letter, we show the crucial importance of nonquasiparticle states for the finite-temperature depolariza-

tion in Co_2MnSi. This is achieved by combining a theoretical analysis of the spin-resolved density of states with tunneling-conductance spectroscopy measurements at different temperatures in a Co_2MnSi—based MTJ. Our combined experimental and theoretical analysis confirms the presence of NQP states and emphasizes their contribution to the finite-temperature polarization in this material.

Band-structure calculations were performed for the half-metallic Co_2MnSi alloy for the experimental lattice constant $a = 5.65$ Å. Calculations were performed using a recently developed LSDA + DMFT scheme [19]. Correlation effects in the valence Co and Mn d orbitals are included via an on-site electron-electron interaction in the form $\frac{1}{2}\sum_{i\{m,\sigma\}} U_{mm'm''m'''} c^\dagger_{im\sigma} c^\dagger_{im'\sigma'} c_{im'''\sigma'} c_{im''\sigma}$. The interaction is treated in the framework of dynamical mean field theory (DMFT) [15], with a spin-polarized T-matrix fluctuation exchange type of impurity solver [20]. Here, $c_{im\sigma}/c^\dagger_{im\sigma}$ destroys/creates an electron with spin σ on orbital m on-site i. The Coulomb matrix elements $U_{mm'm''m'''}$ are expressed in the usual way [21] in terms of three Kanamori parameters U, U', and J. Since the static part of the correlation effects is already included in the local spin-density approximation (LSDA), "double counted" terms must be subtracted. To obtain this, we replace $\Sigma_\sigma(E)$ with $\Sigma_\sigma(E) - \Sigma_\sigma(0)$ [22] in all equations of the LSDA + DMFT procedure [15]. Physically, this is related to the fact that DMFT only adds *dynamical* correlations to the LSDA result. For this reason, it is believed that this kind of double-counting subtraction "$\Sigma(0)$" is more appropriate for a DMFT treatment of metals than the alternative static "Hartree-Fock" (HF) subtraction [23].

Figure 1 shows the results of DOS calculations using the LSDA and LSDA + DMFT schemes at different temperatures. Realistic values of U for all $3d$ metals are predicted to vary between 2 and 6–7 eV [15]. We have checked that for values of the U parameters between 2 and 4 eV the spectral weight of NQP states is not significantly changed in agreement with recent calculations [11]. Here, we present results for $U = 3$ eV and $J = 0.9$ eV. In order to resolve the LSDA minority-spin gap, a broadening of around 15 K and a k-point mesh of 2304 points was used in the reduced Brillouin zone. The LSDA density of states, i.e., the $T = 0$ K result, confirms the existence of a minority-spin gap at E_F, in agreement with previous results [6]. In contrast, finite-temperature results obtained within LSDA + DMFT display a broadening of the NQP states across the Fermi energy (Fig. 1) in the minority-spin channel, and a spectral weight redistribution for majority spins.

Static noncollinear spin configurations at finite temperatures produce a homogeneous mixture of spin-up and spin-down density of states, in such a way that the proportionality relation between polarization and magnetization is roughly maintained as a function of temperature $\delta P(T) \propto \delta\langle S^z \rangle$ [24]. Previously, this proportionality relation was reproduced in magnetic semiconductors [25], in qualitative
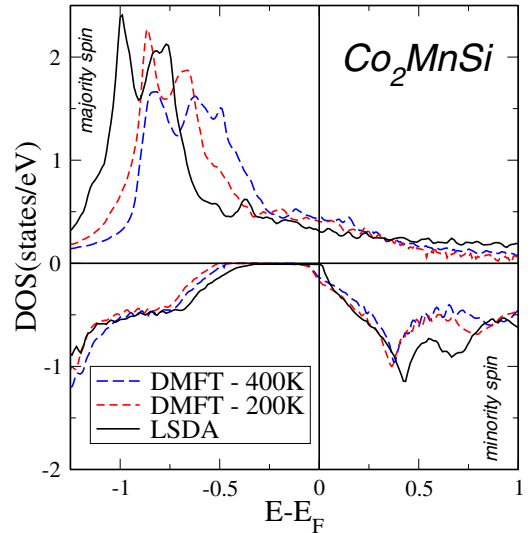

FIG. 1 (color online). Spin-resolved density of states of the Co_2MnSi full-Heusler alloy obtained by LSDA and LSDA + DMFT. The LSDA + DMFT results are obtained for $U = 3$ eV and $J = 0.9$ eV (in both Co and Mn), and different temperatures.

agreement with experimental data [26]. In contrast, for some HMF materials, it was shown by model considerations [18] as well as direct numerical calculations [10,11,14], that the polarization displays a completely different temperature behavior in comparison with the magnetization.

In order to confirm experimentally the existence of NQP states in Co_2MnSi and to investigate their temperature dependence, we carried out tunneling spectroscopy measurements for Co_2MnSi-based MTJ. Specifically, the measurements were first carried out for Co_2MnSi/Al-O/Co_2MnSi-MTJ, for which a TMR ratio of 570% at 2 K was achieved in previous studies [5]. A positive bias voltage was applied to the upper Co_2MnSi electrode, in order to control the tunneling of electrons from the lower electrode to the upper one.

Figures 2(a) and 2(b) show the bias-voltage dependence of the differential tunneling conductance (dI/dV) in the antiparallel (AP) configuration, and the normalized TMR ratio, respectively. The measurements were conducted at temperatures between 2 and 300 K. At $T = 2$ K, only a very small tunneling conductance is observed below the lowest bias voltage ($V \approx 1$ mV). This is due to the half-metallicity of Co_2MnSi. However, the tunneling conductance rapidly increases in the low-bias region (10 mV $< |V| <$ 150 mV), producing a rapid decrease of the TMR ratio, as can be seen in Fig. 2(b). With increasing temperature, the tunneling conductance around the zero-bias region gradually increases and the steep structure at low bias is gradually lost.

These results are consistent with the temperature dependence of the TMR ratios and the saturation magnetization for Co_2MnSi plotted in Fig. 3. Here, the theoretical TMR
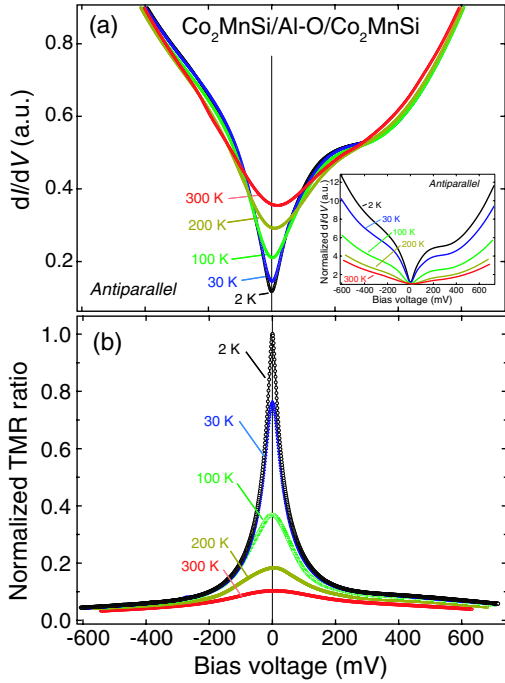
FIG. 2 (color online). (a) Tunnel conductance for the
Co$_2$MnSi/Al-O/Co$_2$MnSi antiparallel magnetic tunnel junction
and (b) TMR ratio (normalized to $T = 2K$, $V = 0$), as a function
of voltage for different temperatures. The inset of panel (a)
shows the conductance normalized to zero bias voltage.

ratio was obtained from the LDA + DMFT polarization by
using Julliere's formula and assuming a temperature-
independent value of 0.5 for the spin polarization of
FeCo, as obtained independently from other measurements
[27]. Despite its approximate character, this formula re-
produces correctly the temperature dependence of the
spin polarization as was previously seen in detailed theo-
retical investigation of the tunneling current in the half-
metallic ferromagnets [17,18]. Moreover, this is the stan-
dard formula used to extract the spin polarization from
TMR data [28]. The saturation magnetization (M_S) was
measured using a SQUID magnetometer for a
MgO-sub./Cr/Co$_2$MnSi film having the same bottom
electrode structure as the MTJs. As one can see, M_S values
change little in the temperature range 2 K $\leq T \leq$ 300 K
(since $T_c \simeq$ 985 K this corresponds to $0.002 \leq T/T_c \leq$
0.3). In contrast, the spin polarization (and thus the TMR
ratio) decrease drastically in the same temperature range.
As can be seen from Fig. 3 the experimental temperature
dependence of the TMR ratio and of the magnetization is in
rather good agreement with our LSDA + DMFT calcula-
tion. Moreover, for $T \lesssim$ 200 K, the experimental polariza-
tion curve (obtained from the TMR data by inversion of
Julliere's formula) is quite well reproduced by the analytic
expression $1 - P(T)/P(0) \propto T \ln T/T^*$ predicted in
Ref. [18]. Here, T^* represents a crossover temperature $\approx$
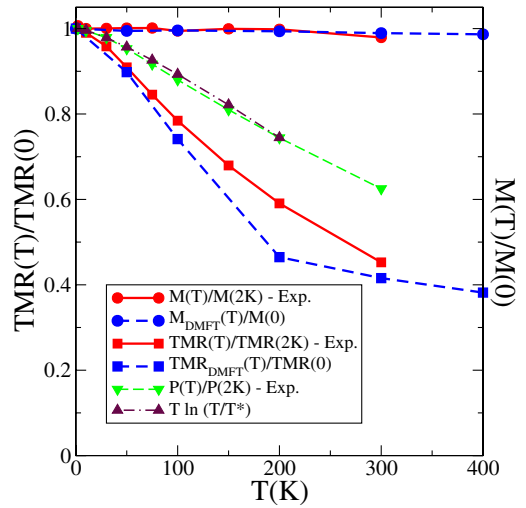$(\Delta/W)^2 T_c$, below which the $\delta P(T) \propto \delta \langle S^z \rangle$ behavior is

FIG. 3 (color online). Comparison of the TMR ratio and
magnetization (M) as a function of temperature for a
FeCo/Al-O/Co$_2$MnSi tunnel junction as obtained from experi-
mental data and from LSDA + DMFT. Triangle-down/up rep-
resents the measured polarization and the fit to the analytic
expression $1 - P(T)/P(0) \propto T \ln T/T^*$ [18].

expected. $\Delta = 0.4$ eV is the minority-spin gap, $W \approx$
8 eV the bandwidth, and $T_c \approx$ 985 K. The value of T^*
estimated by this expression ($T^* \approx$ 2.5 K) is in good
agreement with the value $T^* = 2.7$ K obtained from a fit
to the experimental data. These facts, combined with the
strong temperature dependence of the differential tunnel-
ing conductance shown in Fig. 2, clearly support the ex-
istence of minority-spin NQP states above the Fermi level.

The behavior of dI/dV as a function of bias-voltage V
and temperature can be understood by using the schematic
picture shown in Fig. 4. Here, the crucial role of NQP states
is apparent. In the case of an ideal half-metal at $T \approx 0$ K,
no tunneling process can occur in the AP state, as there are
no electronic states which can contribute to the tunneling.
Upon applying a finite bias voltage ($e|\Delta V| > \hbar\omega_m$) to the
tunnel junctions, a conducting channel for the minority
spins opens due to the nonvanishing NQP density of states
[Fig. 4(a)]. Here, $\hbar\omega_m$ is the anisotropy gap in the magnon
spectrum, below which the density of nonquasiparticle
states vanishes [16]. At finite temperatures ($k_B T > \hbar\omega_m$),
NQP states are expected to broaden and to extend across
E_F as shown in Fig. 1. Therefore, a finite tunneling con-
ductance occurs even at vanishing bias voltage [Fig. 4(b)],
producing a rapid decrease of spin polarization.

The NQP picture suggests that in order to improve the
performances of Co$_2$MnSi-based MTJ, special attention
should be paid to magnon excitations. This suggests a
strategy to improve the performances of Co$_2$MnSi-based
MTJ. The idea is to modify the magnon excitations, while
at the same time preserving the electronic properties, i.e.,
maintaining the gap in the minority channel. One possibil-
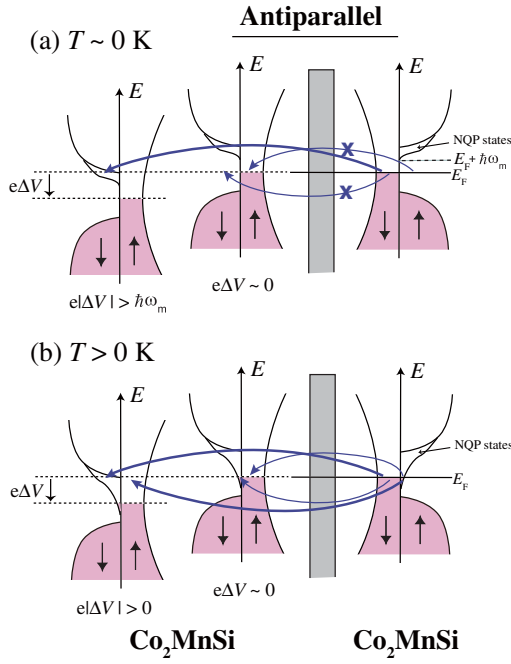ity would be to increase the magnetic anisotropy and thus

FIG. 4 (color online).   Schematic representation of an electron tunneling process at $\Delta V \approx 0$ and at finite bias voltage in a Co_2MnSi/Al-O/Co_2MnSi magnetic tunnel junction. At $T \approx 0$ K (a), NQP states vanish at E_F, while for $T > 0$ (b) (see text) NQP states extend across E_F and an additional tunneling channel opens.

the magnon gap [29]. This can be achieved by a proper doping with rare-earth atoms in the half-metallic material [29]. Experimental work is in progress.

Finally, let us comment on alternative depolarization mechanisms. Dowben *et al.* [24] showed that finite-temperature noncollinearity produces a spin mixing which ultimately leads to a nonvanishing but symmetric DOS around the Fermi level in the gap of the insulating spin channel. In contrast, as discussed above, the low-energy DOS within the minority gap induced by many-body effects (NQP states) appears only above the Fermi level, and is thus strongly asymmetric. In addition, its low-energy part is strongly temperature dependent, and much larger in magnitude than the one produced by noncollinear or spin-orbit effects. Therefore, we expect depolarization due to NQP states to be dominant in comparison with other effects, such as the static noncollinearity [24] or spin-orbit coupling.

In summary, we carried out a combined theoretical and experimental study of depolarization effects in half-metallic Co_2MnSi. Our tunneling-conductance measurements in Co_2MnSi-based magnetic tunnel junctions showed for the first time the existence of NQP states above the Fermi level in the minority-spin channel. The behavior of the finite-temperature conductance demonstrates the important role played by NQP states in inducing depolar-

ization, an effect which should be carefully considered in designing Co_2MnSi-based MTJ devices. The drastic reduction of the TMR ratio from low to room temperatures is clear evidence of the detrimental effect played by the NQP states on the electron spin polarization. A possible strategy to improve the performances of the Co_2MnSi-based MTJ by increasing the magnon anisotropy was also discussed.

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