Non-quasiparticle states in Co$_2$MnSi evidenced through magnetic tunnel junction spectroscopy measurements

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We investigate the effects of electronic correlations in the full-Heusler Co$_2$MnSi, by combining a theoretical analysis of the spin-resolved density of states with tunneling-conductance spectroscopy measurements using Co$_2$MnSi as electrode. Both experimental and theoretical results confirm the existence of so-called non-quasiparticle states and their crucial contribution to the finite-temperature spin polarisation 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 semiconductor surface, fully polarized (100%) electrons could be possibly injected into the semiconductor.

First band-structure calculations performed on the half-Heusler, NiMnSb (C$_1$$_t$-type structure), predicted a 100% spin polarization [1]. Subsequently, the studies were extended to other half-metallic systems. Of particular interest for realizing magnetic tunneling junctions (MTJ) appears to be the full-Heusler alloy Co$_2$MnSi (L2$_1$-type structure). A minority-spin band-gap of 0.4eV has been predicted and a Curie temperature of $T_C$ = 985 K and a saturation magnetisation of $5\mu_B$ was reported [2].

Some of us [3] recently fabricated magnetic tunnel junctions (MTJ) consisting of highly ordered Co$_2$MnSi epitaxial bottom electrode, Al-O tunnel barrier, and Co$_{75}$Fe$_{25}$ top electrode. A TMR ratio of 150% at low temperatures and a value of 70% at room temperature, was determined. More recently, MTJ structures consisting of Co$_2$MnSi/Al-O/Co$_2$MnSi were fabricated, having a TMR ratio of 570%, at 2K, the largest one reported to date for an Al-O amorphous tunneling barrier [4]. These experiments reveal the HMF character of Co$_2$MnSi with a minority spin band gap and a high decrease of TMR ratio with temperature [4].

In order to understand the large temperature variation of the TMR ratio in Co$_2$MnSi, 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. [5]. According to these calculations, the half-metallic character of Co$_2$MnSi is determined by the existence of a minority spin gap formed between the triply degenerate Co-Co anti-bonding $t_{2g}$ and the double degenerate Co-Co anti-bonding $e_g$ 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 non-quasiparticle (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 [6,7,8] (“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, 11, 12, 13, 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, 17, 18]. At $T = 0K$, 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 non-quasiparticle states for the finite-temperature depolarisation in Co$_2$MnSi. 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$_2$MnSi - 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$_2$MnSi alloy for the experimental lattice constant $a = 5.65\,\text{Å}$. Calculations were performed using a recently developed LSDA+DMFT scheme [19]. Correlation effects in the valence Co and Mn orbitals are included via an on-site electron-electron interaction in the form $\sum_{\{m,\sigma\}}^{} U_{mm'\sigma\sigma'} c_{im\sigma}^\dagger c_{im'\sigma'}^\dagger c_{im\sigma} c_{im'\sigma'}$. The interaction is treated in the framework of dynamical mean field theory (DMFT) [15], with a spin-polarized T-matrix Fluctuation Exchange (SPTF) type of impurities solver [21]. Here, $c_{im\sigma}/c_{im\sigma}^\dagger$ destroys/creates an electron with spin $\sigma$ on orbital $m$ on site $i$. The Coulomb matrix elements $U_{mm'\sigma\sigma'}$ 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 [22].

![Figure 1](image1.png)

**FIG. 1:** (Color online) Spin resolved density of states of the Co$_2$MnSi full-Heusler alloy obtained by LSDA and LSDA+DMFT. The LSDA+DMFT results are obtained for $U=3\,\text{eV}$ and $J=0.9\,\text{eV}$ (in both Co and Mn), and different temperatures.

Fig. 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 $U=3\,\text{eV}$ and $J=0.9\,\text{eV}$. In order to resolve the LSDA minority-spin gap, a broadening of around $16\,\text{K}$ and a k-point mesh of 2304 points was used in the reduced Brillouin zone. The LSDA density of states, i.e. the $T=0\,\text{K}$ 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 non-collinear 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 $\delta P(T) \propto S^z > 2\,\text{eV}$ [24]. Previously, this proportionality relation was reproduced in magnetic semiconductors [25], in qualitative 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$_2$MnSi and to investigate their temperature dependence, we carried out tunneling spectroscopy measurements for Co$_2$MnSi-based MTJ. Specifically, the measurements were first carried out for Co$_2$MnSi/Al-O/Co$_2$MnSi-MTJ, for which a TMR ratio of 570% at 2K was achieved in previous studies [3]. A positive bias voltage was applied to the upper Co$_2$MnSi electrode, in order to control the tunneling of electrons from the lower electrode to the upper one.

Figs. 2 (a) and (b) show the bias-voltage dependence of the differential tunneling conductance ($dI/dV$) in the anti-parallel (AP) configuration, and the normalized TMR ratio, respectively. The measurements were conducted at temperatures between 2K and 300K. At $T = 2\,\text{K}$, only a very small tunneling conductance is observed below the lowest bias voltage ($V \approx 1\,\text{mV}$). This is due to the half-metallicity of Co$_2$MnSi. However, the tunneling conductance rapidly increases in the low-bias region ($10\,\text{mV} < |V| < 150\,\text{mV}$), 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$_2$MnSi plotted in Fig. 3. Here, the theoretical TMR 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 polarisation of FeCo, as obtained independently from other measurements [27]. Despite its approximate character, this formula reproduces correctly the temperature dependence of the spin-polarization as was previously seen in detailed theoretical investigation of the tunneling current in the Half-metallic ferromagnets [17, 18]. Moreover, this is the standard 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 $2K \leq T \leq 300K$ (since $T_c \approx 985K$ 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 magnetisation is in rather good agreement with our LSDA+DMFT calculation. Moreover, for $T \lesssim 200K$, the experimental polarisation 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 $\delta P(T)/P(0) \propto T \ln T/T^*$ behavior is expected. $\Delta = 0.4eV$ is the minority spin gap, $W \approx 8eV$ the bandwidth and $T_c \approx 985K$. The value of $T^*$ estimated by this expression ($T^* \approx 2.5K$) is in good agreement with the value $T^* \approx 2.7K$ obtained from a fit to the experimental data. These facts, combined with the strong temperature dependence of the differential tunneling conductance shown in Fig. 2 clearly support the existence 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 $T \approx 0K$, 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. 4a). Here, $\omega_m$ is the anisotropy gap in the magnon spectrum, below which the density of non-quasiparticle 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. 4. Therefore, a finite tunneling conductance occurs even at vanishing bias voltage (Fig. 4b), producing a rapid decrease of spin polarization.

The NQP picture suggests that in order to improve the performances of Co$_2$MnSi-based MTJ, a 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 chan-
The behavior of the finite-temperature conductance demonstrates the important role played by NQP states in inducing depolarization, an effect which should be carefully considered in designing Co$_2$MnSi-based MTJ devices. The drastic reduction of the TMR ratio from low to room temperatures is a 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$_2$MnSi-based MTJ by increasing the magnon anisotropy was also discussed.

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