# Electron Correlations and the Minority-Spin Band Gap in Half-Metallic Heusler Alloys

L. Chioncel,[1,2] E. Arrigoni,[1] M. I. Katsnelson,[3] and A. I. Lichtenstein[4]

[1]*Institute of Theoretical Physics, Graz University of Technology, A-8010 Graz Austria*
[2]*Faculty of Science, University of Oradea, RO-410087 Oradea, Romania*
[3]*Radboud University Nijmegen, NL-6525 ED Nijmegen, The Netherlands*
[4]*Institute of Theoretical Physics, University of Hamburg, DE-20355 Hamburg, Germany*

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Electron-electron correlations affect the band gap of half-metallic ferromagnets by introducing nonquasiparticle states just above the Fermi level. In contrast with the spin-orbit coupling, a large asymmetric nonquasiparticle spectral weight is present in the minority-spin channel, leading to a peculiar finite-temperature spin depolarization effects. Using recently developed first-principle dynamical mean-field theory, we investigate these effects for the half-metallic ferrimagnetic Heusler compound FeMnSb. We discuss depolarization effects in terms of strength of local Coulomb interaction $U$ and temperature in FeMnSb. We propose $Ni_{1-x}Fe_xMnSb$ alloys as a perspective materials to be used in spin-valve structures and for experimental search of nonquasiparticle states in half-metallic materials.

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The realization of physical systems whose electronic states can be easily manipulated by external actions is a very promising issue from both the fundamental and applied sides. For example, spintronics materials in which charge and spin degrees of freedom can be used simultaneously in order to produce devices with new functionality have become subjects of growing interest.

A family of extensively studied spintronics materials are the half-metallic compounds (HM). First-principles electronic structure calculations for HM showed unusual properties in their spin-resolved band structure: while the electronic states for one spin projection have a metallic character with a nonzero density of states at the Fermi level $E_F$, the states with the other spin projection demonstrate a band gap around $E_F$ [1,2]. As a result, HM can in principle conduct a fully spin-polarized current, and therefore attract much attention due to potential applications in the field of spintronics [2,3]. These calculations, based on the density-functional theory with in the local density approximation (LDA) or the generalized gradient approximation, are very successful in many cases to describe or predict material properties. However, they fail notably for the case of strongly correlated electron systems. For such systems the LDA + DMFT (dynamical mean-field theory) method has been designed [4–6] and currently is used very extensively for various applications [7].

One of the dynamical many-electron features of a half-metallic ferromagnet (HMF), the nonquasiparticle (NQP) states [2,8,9], contribute significantly to the tunneling transport in heterostructures containing HMF [10,11], even in the presence of arbitrary disorder [12]. The origin of these states is connected with ''spin-polaron'' processes: the spin-down low-energy electron excitations, which are forbidden for HMF in the one-particle picture, turn out to be possible as superpositions of spin-up electron excitations and virtual magnons [8,9]. In previous publications, we applied the LDA + DMFT approach to describe the nonquasiparticle states in NiMnSb [13] and CrAs [14] and discussed the possible experimental investigation (bremsstrahlung isochromat spectroscopy, nuclear magnetic resonance, scanning tunneling microscopy, and Andreev reflection) techniques to clarify the existence of these states [13].

Here, we investigate the distinction between static (i.e., spin-orbit) and dynamic (correlation) effects leading to a finite-temperature depolarization. According to our results, for the class of Heusler HM, the strong depolarization at finite temperatures is essentially due to correlation effects, while the spin-orbit interaction gives a negligible effect. In addition, we identify characteristic features of the density of states (DOS) spectrum that should help distinguishing between static and dynamic depolarization effects.

Crystal imperfections [15], interfaces [16], and surfaces [17] constitute important examples of static perturbations of the ideal, periodic potential which affect the states in the half-metallic gap. It was shown recently [18], that, due to finite temperatures, the static noncollinear spin-configurations show a mixture of spin-up and spin-down density of states that destroy the half-metallic behavior. The noncollinearity could exist at zero temperature as well, in anisotropic structures, due to the spin-orbit coupling. In such cases static spin-flip scattering will introduce states in the half-metallic gap. It is the purpose of this Letter to use a many-body approach to investigate dynamical spin fluctuation effects on the electronic structure at temperatures below the Curie temperature, $T < T_C$, within the half-metallic state. In order to illustrate the specific differences between the many-body and the static noncollinear effects we extend here our previous LDA + DMFT calculations [13,14] to a different half-metallic Heusler alloy, that is, ferrimagnetic FeMnSb. One of the motivations to study the hypothetical FeMnSb material is to explore the effects of Fe doping in the host NiMnSb compound.

In our approach, correlation effects are treated in the framework of DMFT [7], with a spin-polarized $T$-matrix

 

fluctuation exchange type of the DMFT solver [6], the on-site Coulomb interaction being described by full four-indices orbital matrix [5]. The essential quantity is the LDA + DMFT bath Green function $\mathcal{G}^{\sigma}_{0,m,m'}$, where 0 denotes the impurity site. The DMFT self-consistency equation $\mathcal{G}^{-1}_{0,mm'} = G^{-1}_{\mathrm{LDA},mm'} - \Sigma_{0,mm'}$ is used to combine the LDA Green function with the solution of the impurity model $\Sigma_{0,mm'} = \Sigma_{0,mm'}[\mathcal{G}_{0,mm'}]$. Further computational details are described in Refs. [13,14,19].

In the simplest case of neglecting the dispersion of the magnon frequency, $\omega_{\mathbf{q}} \approx \omega_m$, in comparison with the electron hopping energy $t_{\mathbf{k}}$, the electronic self-energy becomes local [2,9]: For such a self-energy one can calculate the one-electron Green functions $G_{\mathbf{k},\downarrow}(E) = [E - t_{\mathbf{k},\downarrow} - \Sigma^{\mathrm{loc}}_{\downarrow}(E)]^{-1}$, which allow us to evaluate the additional contributions to the density of states. For illustrative purposes we write the lowest-order contribution:

$$\delta N_{\downarrow}(E) = \sum_{\mathbf{k}} \mathrm{Re}\Sigma_{\downarrow}(E)\delta'(E - t_{\mathbf{k}\downarrow}) - \frac{1}{\pi}\sum_{\mathbf{k}} \frac{Im\Sigma_{\downarrow}(E)}{(E - t_{\mathbf{k}\downarrow})^2}. \quad (1)$$

The second term of Eq. (1) is formally connected with the branch cut of the Green function due to the electron-magnon scattering processes [2,9].

In general, one should take into account spin-orbit coupling effects connecting the spin-up and spin-down channels through the scalar product between the angular momentum $\boldsymbol{l}$ and the spin $\mathbf{s}$ operators. The strength of this interaction is proportional to the spatial derivatives of the crystal potential $V(\mathbf{r})$: $V_{\mathrm{SO}} \propto \mathrm{grad}V(\boldsymbol{l} \cdot \mathbf{s})$ with nonzero off-diagonal elements $V^{\sigma,\sigma'}$, $\sigma = \uparrow, \downarrow$. For HM with a gap in the minority-spin (spin-down) channel, one could construct the wave function for spin-down electrons based on the general perturbation consideration so that the DOS in the gap has a quadratic dependence of the spin-orbit coupling strength [20]: $\delta n^{\mathrm{SO}}_{\downarrow}(E) \propto (V^{\downarrow,\uparrow})^2$. As one can see there is an obvious qualitative distinctions between the many-body [Eq. (1)] and the spin-orbit contribution in the minority-spin channel, in addition to the fact that the latter are orders in magnitude smaller [20]. In the former case the strong temperature dependence of the residues of the Green function and the ''tail'' of the NQP states give rise to a strong temperature dependence of the spin polarization, while the spin-orbit term is very weakly temperature dependent.

As a matter of fact, in the nonrelativistic approximation (without spin-orbit coupling) there are two essentially different sources for states in the gap at finite temperatures. First, there is the simple classical effect of band filling due to disorder, that is, due to scattering on static (classical) spin fluctuations. This kind of the gap filling is symmetric with respect to the Fermi energy, that is, there is no essential difference between electrons and holes. In contrast, the correlation effects result in an asymmetry in the gap filling [2], namely, NQP states appear in spin-down channel just above the Fermi level [2,8,9,13,14], and in the spin-up channel below the Fermi level [2,9]. This asymmetry is a purely quantum effect connected with the Pauli principle and with the quantum character of spins; it disappears in the classical limit [2]. For example, in the case under consideration (minority-spin gap) from a consequent quantum point of view, the conduction-electron spin projection is not a good quantum number and real electron states are superpositions of the minority-electron states and majority-electron states plus magnon, with the same projection of the *total* spin of the crystal. However, the majority-electron states *below* the Fermi energy cannot participate in this superposition since they are already completely occupied, therefore these quantum (spin-polaronic) effects below the Fermi energy is totally suppressed at zero temperature by the Pauli principle. As a result, the states in the minority-spin gap at finite temperatures are formed from these spin-polaron states existing only above the Fermi energy plus the disorder-smeared states filling the gap more or less uniformly.

Early theoretical studies showed that the gap of the minority-spin channel is stable with respect to differently chosen $3d$ atom $X$ = Fe, Co, Ni in the XMnSb compounds [21,22]. A notable difference between Ni- and Fe-based Heusler alloys is that NiMnSb is a ferromagnetic half-metal, with a very small value of Ni magnetic moment ($0.2\mu_B$), whereas in FeMnSb the ferrimagnetic coupling between Fe($-1\mu_B$) and Mn($3\mu_B$) moments stabilizes the gap and the half-metallic electronic structure [22].

In the calculations we considered the standard representation of the $C1_b$ structure with a fcc unit cell containing three atoms: Fe(0, 0, 0), Mn(1/4, 1/4, 1/4), Sb(3/4, 3/4, 3/4), and a vacant site E(1/2, 1/2, 1/2). Unfortunately the ternary compound FeMnSb does not exist; however, indications concerning magnetic and crystallographic properties were obtained by extrapolating the series of $Ni_{1-x}Fe_xMnSb$ [22], to high Fe concentration. In this case we chose a lattice parameter of $a = 5.882$ Å for FeMnSb the same as in the recent LDA + SO calculation of Mavropoulos *et al.* [20]. To illustrate the differences between the static and dynamic effects we plot the DOS of the LDA + DMFT calculations which should be compared with recent results including SO coupling [20,22].

Note that depending on the character of chemical bonding, the value of $U$ for all $3d$ metals is predicted to vary between 2 and 6–7 eV [7] A relatively weak dependence of the nonquasiparticle spectral weight, on the $U$ value, (Fig. 1) is evidenced for both NiMnSb and FeMnSb compounds. A ''saturation'' of the spectral weight of FeMnSb takes place for almost the same value, $U^* \approx 1$ eV as in the case of NiMnSb. This effect is understood in terms of the $T$-matrix renormalization of the Coulomb interactions [6]. The spectral weight values for FeMnSb are larger in comparison with the ones obtained for NiMnSb [13], which can be attributed to the fact that at the Fermi level a larger DOS is present in the ferrimagnetic FeMnSb than in the ferromagnetic NiMnSb.

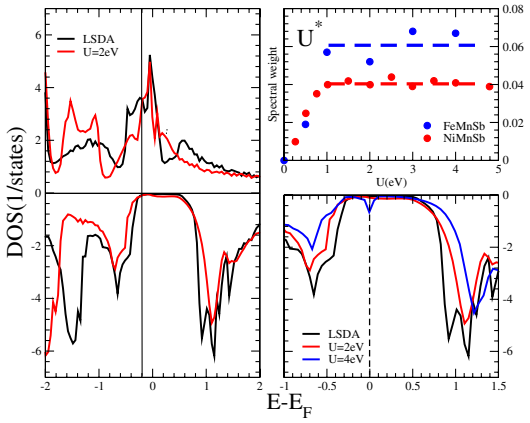


FIG. 1 (color online). Left: Density of states of half-metallic FeMnSb, LSDA (black line) and LSDA + DMFT [gray line (red online)], for the effective Coulomb interaction $U = 2$ eV, exchange parameter $J = 0.9$ eV, and temperature $T = 300$ K. Lower right panel: zoom around $E_F$ for different values of $U$. Upper right panel: Spectral weight of the NQP states calculated as function of $U$. The values obtained for NiMnSb [13] are plotted for comparison.

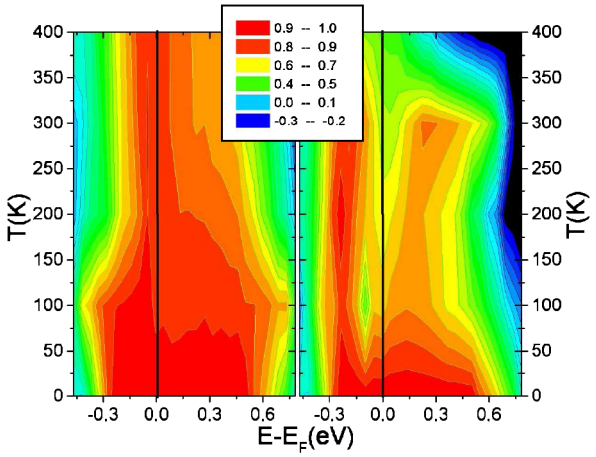


FIG. 2 (color online). Contour plots of polarization as a function of energy and temperature for different values of local Coulomb interaction $U$. Left: $U = 2$ eV, right: $U = 4$ eV. The LSDA polarization is plotted as the $T = 0$ K temperature result. The asymmetry of the NQP states is clearly visible for $U = 4$ eV.

The spin-orbit coupling produce a peak close to the Fermi level [20] in the minority-spin channel, which is an order of magnitude smaller than the spectral weight of the NQP states. According to the SO results [20], the polarization at the Fermi level for NiMnSb and FeMnSb are almost the same. In contrast, our calculation shows that the spectral weight of NQP states in FeMnSb is almost twice as large as the value calculated for NiMnSb.

In order to discuss the influence of temperature and local Coulomb interactions, on the polarization in FeMnSb compound, we present results of LDA + DMFT calculations for $T \leq 400$ K, and different $U$'s. Figure 2 presents the contour plot of polarization $P(E) = [N_\uparrow(E) - N_\downarrow(E)]/[N_\uparrow(E) + N_\downarrow(E)]$ as a function of temperature $T$ for $U = 2$ and 4 eV. The LDA value, plotted for convenience as the $T = 0$ result shows a gap of magnitude 0.8 eV in agreement with previous calculations [20].

One can see a peculiar temperature dependence of the spin polarization. The NQP features appears for $E - E_F \geq 0$, and is visible in Fig. 1, for $U = 2$ eV and the temperature $T = 300$ K. A strong depolarization effect is evidenced for the larger value of $U = 4$ eV. Already at 100 K, there is a strong depolarization of about 25%. Increasing the value of $U$, from 2 to 4 eV, the nonquasiparticle contribution is more significant, therefore the predominant factor in depolarization is played by the NQP. When the tail of these states crosses the Fermi level a drastic depolarization at Fermi level takes place. One can notice that for the case of $U = 4$ eV, the NQP is pinned at the Fermi level, and has a large contribution also due to a large value of DOS in the spin-up channel.

In Fig. 3 one can see a clear distinction between the finite-temperature behavior of the polarization and magnetization, for different values of $U$. It is interesting to note that the reduced magnetization $M(T)/M(0)$ decreases slowly in the temperature range studied in Fig. 2. This reduction is a consequence of the finite-temperature excitations, i.e., spin-flip processes, affecting both spin channels. In the minority-spin channel, NQP states are formed, and in the majority channel a spectral weight redistribution around the Fermi level (Fig. 1) contributes to the depolarization. The corresponding depolarization increases with the strength of correlations. The density of NQP states displays a rather strong temperature dependence [9,10], resulting in the asymmetry that is visible in Figs. 1 and 2. Recently Dowben *et al.* [18] showed that noncollinearity results in a spin mixing which ultimately leads to a nonvanishing but symmetric DOS around the Fermi level in the gap of the insulating spin channel. Therefore, we suggest that the asymmetry in the DOS of the minority channel is the key feature that should help distinguishing whether correlation effects are responsible for the finite-temperature depolarization or not.

Our results show that at finite temperatures the NQP states appear in the gap of the minority-spin channel,

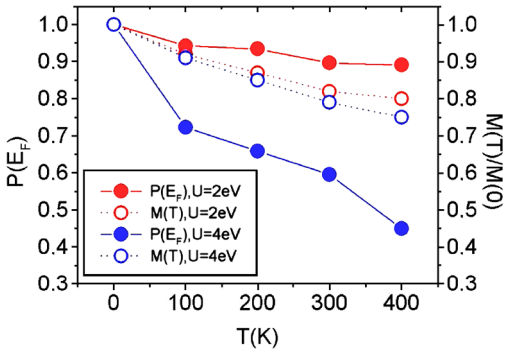


FIG. 3 (color online). Temperature dependent polarization at the Fermi level, $P(E = E_F, T)$ (solid line) and magnetization (dashed line) for different values of local Coulomb interaction $U$.

reducing the polarization significantly. In such a situation, the half-metallic state is depleted. On the other hand, recently Wang [23] suggested that states in the minority-spin gap of a half-metallic ferromagnet are localized due to disorder (Anderson localization). In such a case, the material behaves like a fully polarized HMF. The question whether NQP states are conducting or not has not been studied in the present work, and will be the subject of our future research.

Our work suggests that depolarization in this class of Heusler compounds is dominated by NQP states, while spin-orbit contributions are much smaller. In addition, many-body effects are more pronounced in FeMnSb than in NiMnSb. This is tightly connected to the larger DOS in the majority spin channel in the former material. Therefore, doping of NiMnSb by Fe could be an interesting issue to investigate the interplay between alloying and many-body effects. In this respect, we have carried out preliminary LDA + DMFT calculations [24] on NiMnSb supercell containing 25% Fe impurities, i.e., for $(Ni_3Fe)Mn_4Sb_4$. Our results show a half-metallic character at the LDA level, with similar strong correlation-induced depolarization effects as in pure FeMnSb. Therefore, for this material, many-body effects are of primary importance even in the presence of disorder. Correlation effects on surfaces of half-metals were discussed recently and it was shown that these states can be probed both directly and via their effect on surface states [12]. As a consequence in addition to the previously discussed experimental techniques [13], we propose the use of $Ni_{1-x}Fe_xMnSb$ alloys both in spin-valve structures and to investigate the existence of NQP states in half-metallic materials.

The discovery of giant magneto-resistance (GMR) has led to tremendous amount of activity to understand and develop technology based on high-density magnetic recording. The dominant mechanism leading to GMR is the spin-dependent *s-d* scattering. NiMnSb-based spin-valve structures using Mo spacer layers NiMnSb/Mo/NiMnSb/$SmCo_2$ were successfully produced [25]. The associated GMR exhibits a clear spin-valve contribution of around $\Delta R/R \approx 1\%$ [25]. One of the limiting factor for such a small value is the large resistivity of the Mo layer which determines limited flow of *active electrons* exchanged between the two ferromagnetic layers without being scattered. To improve on the GMR value the use a low-resistivity standard spacer such as Au or Cu was suggested [25]. On the other hand, having a larger value of DOS at the Fermi level which occurs in the majority spin channel (Fig. 1), the ferrimagnetic FeMnSb or $Ni_{1-x}Fe_xMnSb$ could increase the number of *active electrons* in such a spin-valve configuration. In addition, due to its ferrimagnetic properties, the spin-valve demagnetization field can be reduced. The reduced demagnetization is extensively exploited in synthetic ferrimagnet spin-valve heads and are known to have advantages over conventional spin-valve heads [26].

It is interesting to note that, due to its larger DOS in the majority spin channel, FeMnSb is expected on the one hand to provide a better performance in Half-metal based spin-valves in comparison with NiMnSb. On the other hand, our calculation shows that such a larger DOS is accompanied by an equally larger DOS of NQP that suppresses polarization. The conclusion, thus, is that correlation effects are important and should not be neglected precisely in putatively ''good'' HM materials, i.e., in materials with a large DOS in the majority spin channel.

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[1] R. A. de Groot *et al.*, Phys. Rev. Lett. **50**, 2024 (1983).
[2] V. Yu. Irkhin and M. I. Katsnelson, Usp. Fiz. Nauk **164**, 705 (1994). [Phys. Usp. **37**, 659 (1994)].
[3] I. Žutiċ, J. Fabian, and S. Das Sarma, Rev. Mod. Phys. **76**, 323 (2004).
[4] V. I. Anisimov *et al.*, J. Phys. Condens. Matter **9**, 7359 (1997).
[5] A. I. Lichtenstein and M. I. Katsnelson, Phys. Rev. B **57**, 6884 (1998).
[6] M. I. Katsnelson and A. I. Lichtenstein, J. Phys. Condens. Matter **11**, 1037 (1999); Eur. Phys. J. B **30**, 9 (2002).
[7] G. Kotliar and D. Vollhardt, Phys. Today **57**, No. 3, 53 (2004); G. Kotliar *et al.*, cond-mat/0511085 [Rev. Mod. Phys. (to be published)].
[8] D. M. Edwards and J. A. Hertz, J. Phys. F **3**, 2191 (1973).
[9] V. Yu. Irkhin and M. I. Katsnelson, Sov. Phys. Solid State **25**, 1947 (1983).
[10] V. Yu. Irkhin and M. I. Katsnelson, Eur. Phys. J. B **30**, 481 (2002).
[11] E. McCann and V. I. Fal'ko, Phys. Rev. B **68**, 172404 (2003).
[12] V. Yu. Irkhin and M. I. Katsnelson, Phys. Rev. B **73**, 104429 (2006).
[13] L. Chioncel *et al.*, Phys. Rev. B **68**, 144425 (2003).
[14] L. Chioncel *et al.*, Phys. Rev. B **71**, 085111 (2005).
[15] H. Ebert and G. Schütz, J. Appl. Phys. **69**, 4627 (1991).
[16] G. A. de Wijs and R. A. de Groot, Phys. Rev. B **64**, 020402 (2001).
[17] I. Galanakis, J. Phys. Condens. Matter **14**, 6329 (2002).
[18] P. A. Dowben and R. Skomski, J. Appl. Phys. **93**, 7948 (2003).
[19] L. Chioncel *et al.*, Phys. Rev. B **67**, 235106 (2003).
[20] Ph. Mavropoulos *et al.*, J. Phys. Condens. Matter **16**, S5759 (2004).
[21] J. Kubler, Physica (Amsterdam) **127B+C**, 257 (1984).
[22] R. A. de Groot, A. M. van der Kraan, and K. H. J. Buschow, J. Magn. Magn. Mater. **61**, 330 (1986).
[23] X. Wang and X.-G. Zhang, Phys. Rev. Lett. **82**, 4276 (1999).
[24] L. Chioncel *et al.* (to be published).
[25] C. Hordequin, J. P. Nozieres, and J. Pierre, J. Magn. Magn. Mater. **183**, 225 (1998).
[26] H. Kanai, K. Noma, and J. Hong, Fujitsu Sci. Tech. J. **37**, 174 (2001).