Correlation effects and the electronic structure of half-metallic feromagnets

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Most theoretical efforts for understanding half-metallic ferromagnets (HMF) are supported by first-principles calculations, based on density-functional theory (DFT). In fact the very discovery of HMF was due to such calculations [1]. DFT calculations are usually based on the Local Spin Density Approximation (LSDA) or the Generalized Gradient Approximation (GGA). These approximations have been proved very successful to interpret or even predict material properties in many cases, but they fail notably in the case of strongly-correlated electron systems. For such systems the so-called LSDA+U (or GGA+U) method is used to describe static correlations, whereas dynamical correlations can be approached within the LSDA+DMFT (Dynamical Mean-Field Theory). An important dynamical many-electron feature of half-metallic ferromagnets is the appearance of non-quasiparticle states [2,3] which can contribute essentially to the tunneling transport in heterostructures containing HMF [4,5].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. The density of these non-quasiparticle states vanishes at the Fermi level but increases drastically at the energy scale of the order of a characteristic magnon frequency ω_m, giving an important contribution in the temperature dependence of the conductivity due to the interference with impurity scattering [3].

Our study focuses on the half-metallic ferromagnets in which the gap is situated in the spin-down/minority channel. (The results are valid equally to the cases when the half-metallic gap is situated in the spin-up channel). In this extend abstract the examples of semi-Heuslers NiMnSb [6] and FeMnSb [7] will be presented, while the lecture presents also results for full-Heusler Co_2MnSi and zinc-blende half-metals like CrAs [8] and VAs [9].

The temperature dependence of the HMF electronic structure and stability of half-metallicity against different spin-excitations is crucial for practical applications in spintronics. A simple attempt to incorporate finite-temperature effects [10], leading to static non-collinear spin-configurations, shows a mixture of spin up and spin down density of states that destroys the half-metallic behavior. In our work we use a different more natural approach to investigate the proper dynamical spin fluctuations effect on the electronic structure at temperatures $T<T_c$, within the half-metallic ferromagnetic state.

Nonquasiparticle states: an illustrative example: Before investigating, *ab initio*, real materials it is worthwhile to illustrate the correlation effects on the electronic structures of HMFs using a simple ``toy" model. The one-band Hubbard model of a saturated ferromagnet can provide us the simplest model of a half-metallic state. Difficulties in solving the Hubbard model are well known. Fortunately there is an exact numerical solution in the limit of infinite dimensionality or large connectivity called Dynamical Mean Field Theory (DMFT) [11]. Following this approach we will consider the Bethe lattice with coordination $z \rightarrow \infty$ and nearest neighbor hoping $t_{ij} = t/z^{1/2}$. In this case a semicircular density of states is obtained as function of the effective hopping t: $N(\varepsilon)=1/(2\pi t^2)\ (4t^2 - \varepsilon^2)^{1/2}$. In order to stabilize our "toy" model in the HMF state, we add an external magnetic spin splitting term Δ=0.5eV, which mimics the local Hund polarization originating from other orbitals in real materials. This HMF state corresponds to a mean-field (HF) solution with an LSDA-like DOS, denoted in Fig.1 as a dashed line.

Our model allows the study the magnon spectrum through the two-particle correlation function which is obtained using the QMC procedure. We calculate the local spin-flip susceptibility[6]:

$$\chi_{loc}^{+-}(\tau-\tau') = \langle S^{+}(\tau)\, S^{-}(\tau')\rangle$$
$$\chi_{loc}^{+-}(\tau-\tau') = \langle T_{\tau}\, c_{\uparrow}^{+}(\tau)\, c_{\downarrow}(\tau)\, c_{\downarrow}^{+}(\tau')\, c_{\uparrow}(\tau')\rangle_{Seff}$$

which gives us information about the integrated magnon spectrum [3,12]. The DMFT results are presented in Fig.1. In comparison with a simple Hartree-Fock solution one can see an additional well-pronounced feature appearing in the spin-down gap region, just above the Fermi level. This new many-body feature corresponds to the so-called non-quasiparticle states in HMF's and represents the spin-polaron process [2,3]

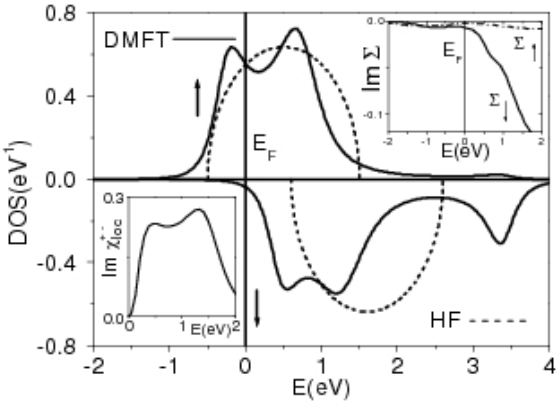

Fig.1. Density of states for HMF in the Hartree-Fock (HF) approximation (dashed line) and in QMC solution of DMFT problem for semi-circular model (solid line) with the band-width W=2eV,Coulomb interaction U=2 eV, Δ=0.5 eV, chemical potential μ=-1.5 eV and temperature T=0.25 eV. Insets: imaginary part of the the spin-flip susceptibility (left) and imaginary part of self energy (right).

The left inset of Fig.1, represents the imaginary part of the local spin-flip susceptibility. One can see a well pronounced shoulder ~ 0.5eV, which is related to a characteristic *magnon* excitation [3]. In addition there is a broad maximum ~1eV corresponding to the Stoner excitation energy. The right inset of Fig.1, represents the imaginary part of self-energy. The spin up channel can be described by a Fermi-liquid type behavior, with a parabolic energy dependence Im $\Sigma_\uparrow \sim (E-E_F)^2$, whereas in the spin down channel, of $\Sigma_\downarrow$, the non-quasiparticle shoulder at 0.5 eV, is visible.

The existence of the non-quasiparticle states for this model has been proved by perturbation-theory arguments [2] (i.e. for a broad-band case) and in the opposite, infinite-U limit [3]. Physically, the appearance of these states can be considered as a kind of spin-polaron effect. According to the conservation laws, in the many-body theory the spin-down state with the quasimomentum **k** can form a superposition with the spin-up states with the qausimomentum **k-q** plus a *magnon* with the quasimomentum **q** running through the whole Brillouin zone. Taking into account the restrictions from the Pauli principle (an impossibility to scatter into occupied states) one can prove that this superposition can form only above the Fermi energy (here we consider the case where the spin-up electronic structure is metallic and the spin-down is semiconducting; oppositely, the nonquasiparticle states form only *below* the Fermi energy) [2,3]. If we neglect the *magnon* energy in comparison with the typical electron one, the density of non-quasiparticle states will vanish abruptly right at the Fermi energy; more accurate treatment shows that it vanishes continuously in the interval of the order of the *magnon* energy according to a law which is dependent on the *magnon* dispersion [2]. As a consequence the non-quasiparticle states are almost currentless [2,3]. Recently, some evidence of the existence of almost currentless states near the Fermi energy in the half-metallic ferromagnet CrO_2 have been obtained by X-ray spectroscopy [13].

NiMnSb and FeMnSb: In the previous section we illustrated the effect of many-body interactions on the Hubbard model for a half-metallic ferromagnet. In this section we show that *the same* many-body effects are captured in the realistic LDA+DMFT electronic structure calculation for NiMnSb and FeMnSb. Fig.2 represents the results for density of states using LDA and LDA+DMFT

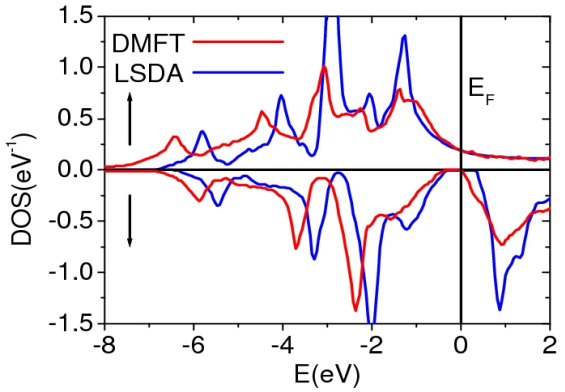

Fig. 2 Density of states for HMF NiMnSb in the LSDA scheme (blue line) and in LDA+DMFT scheme (red line) with effective Coulomb interaction U=3eV, exchange parameter J=0.9eV and temperature T=300 K. The NQP state is evidenced just above the Fermi level.

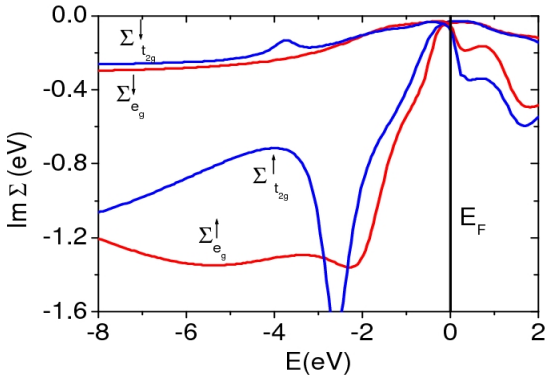

Fig.3 The imaginary part of self-energies ImΣ_d^dw for t_{2g} (solid line) and e_g (dotted line), ImΣ_d^{\up} for t_{2g} (dashed line) and e_g (dashed dotted line) respectively.

It is important to mention that the magnetic moment per formula unit is not sensitive to the U values. For a temperature equal to T=300K the calculated magnetic moment, μ=3.96 μ_B, is close to the integer LDA-value μ=4.00μ_B, which suggests that the half-metallic state is stable with respect to the introduction of the correlation effects. In addition, the DMFT gap in the spin down channel, defined as the distance between the occupied part and starting point of non-quasiparticle states' "tail", is also not very sensitive to the U values. For different U the slope of the "tail" is slightly changed, but the total DOS is weakly U-dependent due to the T-matrix renormalization effects. Thus different values of U, do not effect too strongly on a general picture of the LDA+DMFT electron energy spectrum (except the smearing of the density of states features which is due to different temperatures in our calculations). In comparison with the LDA result, a strong spectral weight transfer is present for the whole occupied part of the valence band. The most interesting new effect is the appearance of the non-quasiparticle states in the energy gap above the Fermi energy. Their spectral weight for realistic values of the parameters are not too small which means that they should be well-pronounced in the corresponding experimental data.

In general, one should take into account *spin-orbit coupling effects* connecting the spin-up and -down channels through the scalar product between the angular momentum **l** and the spin **s** operators. The strength of this interaction is proportional to the spatial derivatives of the crystal potential V(**r**): V_{SO} ~ grad V **ls** with non zero off-diagonal elements $V^{\sigma,\sigma'}$,σ=↑,↓. Our results [7] suggest 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. (see Fig.5). 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 calculation 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.

The nature of non-quasiparticle states: From the point of view of the many-body theory, the general approach in DMFT is to neglect the momentum-dependence in the electron self-energy. In many cases, such as the Kondo effect, the Mott metal-insulator transition, etc. the energy dependence of the self-

energy is obviously much more important than the momentum dependence and, therefore, the DMFT is adequate to consider these problems [11].

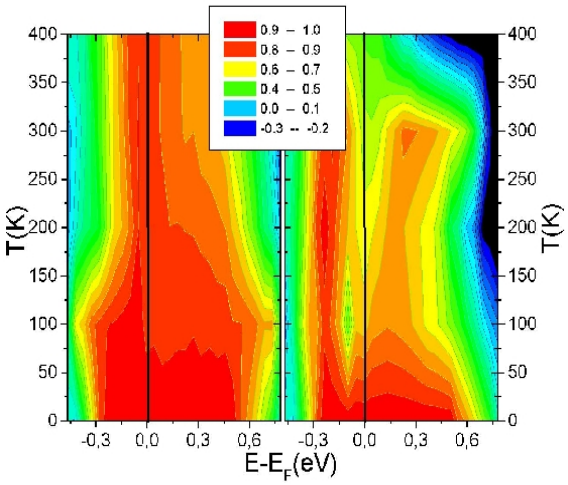

Fig.4 Contour plots of polarization as function of energy and temperature for different values of local Coulomb interaction U. Left U=2eV, right U=4eV. The LSDA polarization is plotted as the T=0K temperature result. The asymmetry of NQP states is clearly visible for U=4eV.

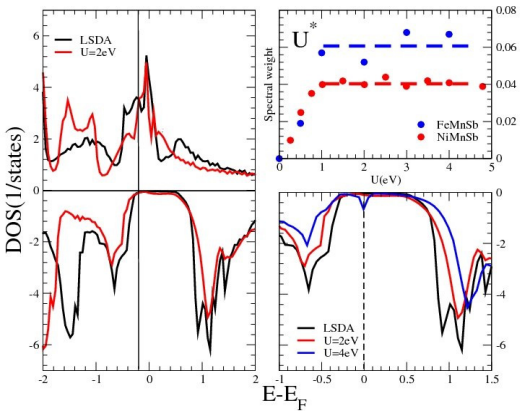

Fig.5 Left: Density of states of half-metallic FeMnSb, LSDA (black line) and LSDA+DMFT (red line), for the effective Coulomb interaction U=2eV exchange parameter J=0.9eV and temperature T=300K. Lower right panel: zoom around E_F for different values of U. Upper right panel: Spectral weight of the NQP states calculated as function of U. The values obtained for NiMnSb are plotted for comparison.

As for itinerant electron ferromagnetism, the situation is not completely clear. One can expect that in magnets with well defined local magnetic moments such as half-metallic ferromagnets the local approximation for the self-energy (i.e., the DMFT) should be even more accurate. In particular, as we discussed above, it can be used for the calculations of spin-polaronic (non-quasiparticle) effects in these materials.

Several experiments could be performed in order to clarify the impact of these non-quasiparticle states on spintronics. Experiments such as Bremsstrahlung Isohromat Spectroscopy (BIS) or Spin Polarized Scanning Tunneling Microscopy (SP-STM), Andreev reflection spectroscopy using the tunneling junction superconductor - HFM will be discussed during the lecture.

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