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Electronic correlation effects on the Curie temperature in double perovskites

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Abstract

In order to understand the role of electronic parameters controlling the half-metallic ground state and high Curie temperature in double perovskites we study the ferromagnetic compound Sr_2FeMO_6, where M is a transition metal. Here, we present a theoretical electronic approach to study the thermodynamic properties of these perovskites using the Green function technique and the renormalization perturbation expansion method. Our approach is based on an interacting picture of localized Fe-electrons and conduction M-electrons via a double-exchange-type mechanism and taking into account electronic correlation. Our results show an increase of the Curie temperature together with a shift to high band filling in good agreement with experimental results.
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Keywords: Half-metallic; Electronic correlation; Spintronics

1. Introduction

Recently, the physics of half-metallic materials in which only one-spin direction is present at the Fermi level, has been investigated in view of the potential spintronic applications [1], memory devices could be one of them due to their large magnetoresistance. The double perovskite compounds are one of those half-metallic materials, in

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particular the ferromagnetic oxide Sr_2FeMoO_6 is one of the most promised double perovskites since has a high Curie temperature $T_C=420K$ and colossal magnetoresistance at low-field [2]. In the double perovskite Sr_2FeMoO_6 and for the fully ordered case, Fe and Mo atoms occupy two interpenetrating sublattices α and β respectively. Oxygen atoms bridge the Fe and Mo ions and Sr lies on the middle of the cubic lattice.

The half-metallic character of the ferromagnetic oxide Sr_2FeMoO_6 may be modified by the introduction of disorder [3,4], experimental results of the saturation magnetization give values of the order of $3.2\mu_B$ lower than the saturation moment $4.0\mu_B$ expected for full polarization (S_{total}=5/2-1/2), this discrepancy is attributed to disorder in Fe and Mo ions and antiferromagnetic coupling between nearest-neighbors Fe [3,4]. In the present paper, we will consider Hubbard electronic correlation in both Fe and Mo sites for fully ordered systems in order to analyzed the Curie temperature behavior in double perovskites.

2. Theoretical Model

The model introduced here is based on a strongly correlated electron picture with localized Fe electrons (in the stable configuration d^5) with parallel spin and itinerant Mo electrons interacting with the local spins via a double-exchange type mechanism. The Hund´s coupling on Fe sites gives the high spin configuration for parallel spins at the t_{2g} and e_g orbitals of the $3d^5$ configuration, it means S=5/2. In addition, itinerant electrons arising from Mo d^1 configuration on t_{2g} orbital can hop on Fe sites only antiparallel to the localized spins due to the Pauli exclusion principle, providing the d^6 configuration with spin S=2. This is the basic ingredient for the kinetic mechanism of our electronic theory [5].
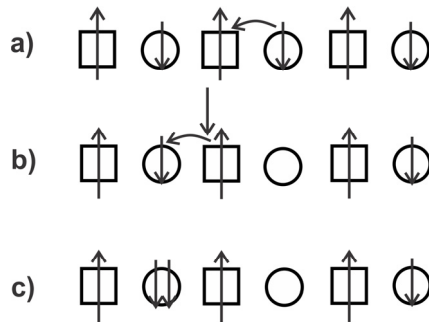

Fig. 1. Spin configuration scheme for Fe and Mo ions in the ferromagnetic state (a). Only spin down electrons can hop around, on Fe sites (b) and from Fe they can hop on Mo (c) due to the presence of degenerate orbitals. The localized Fe spin configuration is indicated by squares and circles give itinerant electrons spin configuration of Mo.

In Fig. 1, we illustrate the localized and itinerant electron spin configuration in the ferromagnetic state. We consider only nearest-neighbors hopping between Fe and Mo.
The electronic correlations among itinerant electrons are considered through the Hubbard on-site interaction on Mo and Fe sites

$$H_c^{Mo} = (U^{Mo} + 2J^{Mo})\sum_{i,\nu}n_{i\nu\uparrow}n_{i\nu\downarrow} + U^{Mo}\sum_{i,\nu,\nu'\neq\nu,\sigma}n_{i\nu\uparrow}n_{i\nu'\downarrow} + (U^{Mo} - J^{Mo})\sum_{i,\nu,\nu'\neq\nu,\sigma}n_{i\nu\sigma}n_{i\nu'\sigma},$$

$$H_c^{Fe} = (U^{Fe} - J^{Fe})\sum_{j,\nu,\nu'\neq\nu,\sigma}n_{j\nu\sigma}n_{j\nu'\sigma,}$$

i and j correspond to sites occupied by Mo and Fe respectively, ν and ν' label the three degenerated t_{2g} orbitals. H_c^{Mo} takes into account that both spin directions are possible on Mo sites while, on the contrary, H_c^{Fe} is restricted to correlations between parallel spins on different orbitals. Treating these terms in the mean-field approximation we obtain the effective Fe and Mo sites energies for itinerant electrons, $\tilde{E}_{Fe,\sigma}$ and $\tilde{E}_{Mo,\sigma}$ respectively. $\tilde{E}_{Fe,\uparrow}(\tilde{E}_{Fe,\downarrow})$

refers to Fe sites with localized spins – (+). These sites energies have to be determined self-consistently. To obtain the density of states for itinerant electrons we calculate the local Green´s functions in an interpenetrating Bethe lattice using the Renormalized Perturbation Expansion (RPE) [5]. These Green´s functions with spin down are given by

$$G_{\downarrow+}^{\alpha\,Fe} = \frac{1}{\omega - \tilde{E}_{Fe,\downarrow} - \frac{w^2}{4} G_{\downarrow}^{\beta\,Mo}},$$

$$G_{\downarrow}^{\beta\,Mo} = \frac{1}{\omega - \tilde{E}_{Mo,\downarrow} - \frac{w^2}{4} \nu_+ G_{\downarrow+}^{\alpha\,Fe}},$$

and those with spin up are

$$G_{\uparrow-}^{\alpha\,Fe} = \frac{1}{\omega - \tilde{E}_{Fe,\uparrow} - \frac{w^2}{4} G_{\uparrow}^{\beta\,Mo}},$$

$$G_{\uparrow}^{\beta\,Mo} = \frac{1}{\omega - \tilde{E}_{Mo,\uparrow} - \frac{w^2}{4} \nu_- G_{\uparrow-}^{\alpha\,Fe}},$$

Where $\nu_\pm = (1 \pm m)/2$ is the probability that an Fe site has its localized spin + or −. 2w being the band width.

4. Results and discussions

In general, in the absence of correlation, itinerant electrons have different site energies on Mo and Fe, E_{Mo} and E_{Fe} respectively, thus determining $\tilde{E}_{Fe,\sigma}$ and $\tilde{E}_{Mo,\sigma}$. $\Delta_0 = E_{Mo} - E_{Fe}$ is a key parameter for an electronic theory; we take E_{Fe} as the reference energy. In the following we will present results using $U_{eff}^{Fe} = U^{Fe} - J^{Fe} = 3w$, $U_{eff}^{Mo} = U^{FMo} - J^{Mo} = w$ and $J^{Mo} = 0.1w$ for the correlation parameters on Fe and Mo. In the ferromagnetic phase (m=1) it is convenient to define $\Delta = \tilde{E}_{Mo,\downarrow} - \tilde{E}_{Fe,\downarrow}$ as a parameter. It is straightforward to obtain Δ as function of Δ_0 [3]. In the particular case $\Delta=0$ this reduces to $\Delta = \Delta_0 - 2nw/3$ i.e. $\Delta_0 = 2nw/3$.

In figure 2, results for the density of states as function of energy are shown for the ferromagnetic (m=1) (Fig. 2a) and paramagnetic (m=0) (Fig. 2b) states and different band filling n, using $\Delta_0 = 2nw/3$. As expected, in the ferromagnetic state, only down spin electron states exist and we obtain the semi-elliptic density of states with a bandwidth 2w. In the paramagnetic phase, the bandwidth is reduced and a gap appears within the equal up and down density of states. In both cases, the density of states shows a shift to higher energies with increasing n, due to the increase of both Δ_0 and electronic correlations.

The kinetic energy $E_{kin}(m)$ of the itinerant electrons as a function of magnetization is presented in Fig. 3. It is calculated from the total density of states,

$$E_{kin}(m) = 3 \sum_{\sigma} \int_{-\infty}^{E_F} \rho_\sigma \left(m, \omega, \tilde{E}_{Mo,\sigma}, \tilde{E}_{Fe,\sigma} \right) \omega \, d\omega$$

The Helmholtz free energy $F = E_{kin}(m,\Delta) - TS(m)$, is obtained directly with $S(m) = \ln(2) - \nu_+\ln(2\nu_+) - \nu_-\ln(2\nu_-)$ the entropy of the local spins consistent with our approximation that these spins are either up or down. The Curie temperature (T_C) is determined from the condition $m(T_C)=0$ [5], it means, the minimum of the free energy,

$$T_C = -\frac{1}{k_B}\left.\frac{\partial^2 E_{kin}(m)}{\partial m^2}\right|_{m=0}.$$

Figure 3 (see inset) shows this limit for the kinetic energy.

Figure 4 shows the Curie temperature T_C/w as function of the band filling for the correlated case (indicated by dots) using $\Delta_0 = 2nw/3$ and the above mentioned values for the correlation parameters, the non-correlated case $\Delta = \Delta_0 = 0$ (indicated by squares) is also included as a reference. For the non correlated case the maximum T_C is reached in the range 0.5-1 and the half-metallic character is nevertheless preserved for band filling up to n=2.5. Electronic correlations permit to raise T_C by increasing n, in particular in Mo compound (n=1) by doping Sr with trivalent ions like in qualitative agreement with the experimental behavior [6]. A complete work will be presented in a forthcoming publication. Our results for the Curie temperature are in qualitative agreement with those obtained by Brey *et al*. [7].
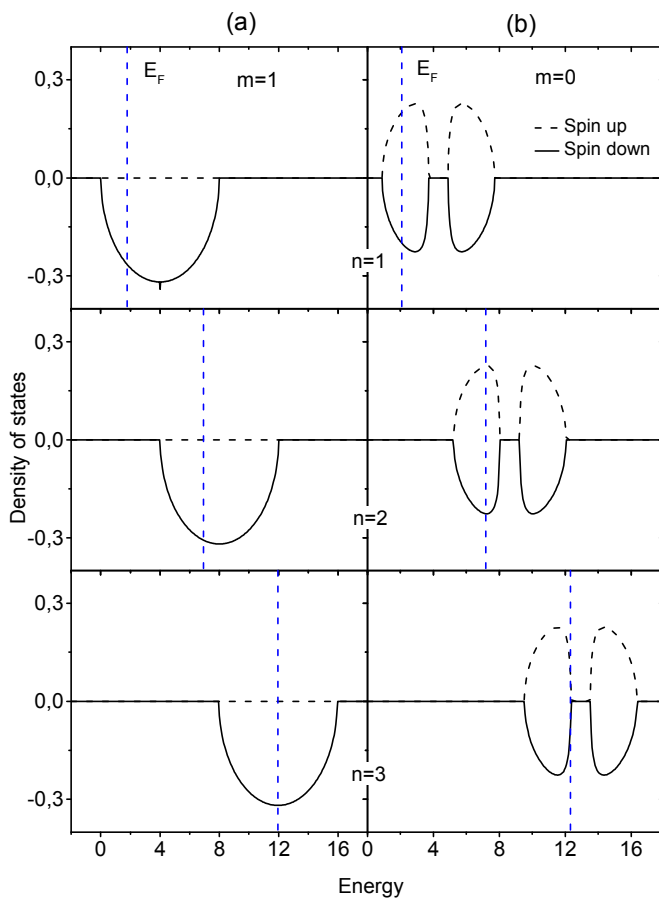

Fig. 2. Density of states versus energy for the Sr_2FeMO_6 compound (M is a transition metal) in both, ferromagnetic (m=1) and paramagnetic (m=0), phases for different values of the band filling (n).
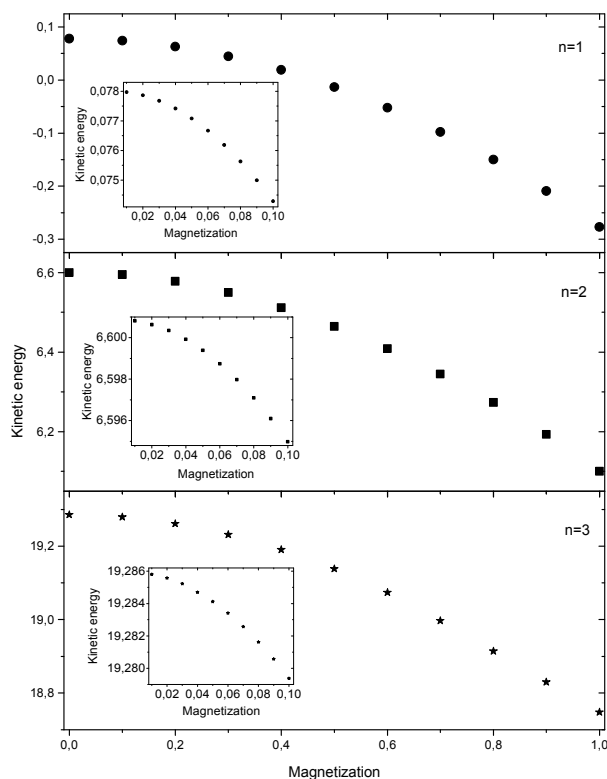
Fig. 3. Kinetic energy as function of the magnetization for different values of the band filling (n=1,2,3). Inset figure shows the limit of the kinetic energy when the magnetization goes to 0.
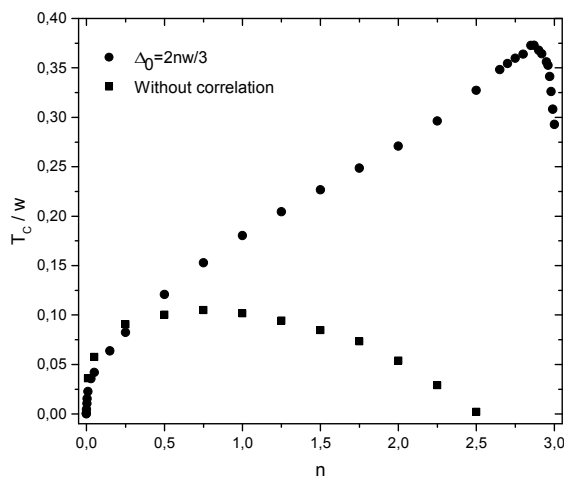

Fig. 4. Variation of the Curie temperature T_C as function of the band filling n, for the non-correlated (squares) and correlated (dots) cases, the electronic correlation in Fe and Mo ions induces a transfer energy given by $\Delta_0 = 2nw/3$.

Conclusions

Our results illustrate the behavior of the Curie temperature in double perovskites Sr_2FeMO_6 (M a transition metal) compounds, within a strongly correlated approach and the Green´s function formalism. The reasonable good agreement of our T_C with the experimental results confirms clearly that the electronic correlation effects should be incorporated in order to explain the experimental observation in these double perovskites.

Acknowledgements

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