GW study of half-metallic electronic structure of $La_{0.7}Sr_{0.3}MnO_3$

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Abstract

Half-metallic systems will be quite useful in future spin electronics. The *GW* approximation is employed to study electronic structure of half-metallic $La_{0.7}Sr_{0.3}MnO_3$, to show that the lowest quasiparticle energy of the unoccupied minority spin states is far above the Fermi energy compared to that in the LSDA. As a bulk, this system is predicted to be a fully spin-polarized half-metallic ferromagnet.

Key words: *GW* approximation ; colossal-magnetoresistive manganites ; half-metals

1. Introduction

Half-metallic systems[1] attract much attention for possible applications to future spin electronics. A lot of works are executed to find out promising materials both in experiments and theories. Among them, $La_{0.7}Sr_{0.3}MnO_3$ is a material in dispute. The point is the degree of the spin polarization, which is important for device performance. But the experimental values of the spin polarization varies from 35% to 100%[2].

In the LSDA, this system is calculated to be a spin-polarized half-metal, but has non-zero density of states in the minority spin component as well as the majority spin component.[3] However it is well known that the LSDA has difficulties in describing excited states: The LSDA underestimates band gaps in semiconductors and insulators. The *GW* approximation[4] is a method which takes account of electron correlations within the RPA to give larger band gaps than those in the LSDA in most cases. In general, the calculated *GW* band gaps are in good agreement with the experimental ones.

In this study, *GW* quasiparticle energies are calculated to theoretically clarify electronic structure of $La_{0.7}Sr_{0.3}MnO_3$ including electron-electron correlations beyond the LSDA treatment. It is shown that the lowest energy of the unoccupied minority spin state is far above the Fermi energy compared to that in the LSDA. As a bulk, this system is predicted to be a fully-polarized half-metal in the first-principles calculation. This is the first study for the half-metallic system in the *GW* approximation.

2. Method of the Calculation and Results

In the *GW* approximation, quasiparticle energy E_{kn} is calculated as

$$E_{kn} = \epsilon_{kn}^{\mathrm{LDA}} + \left(\langle \psi_{kn}^{\mathrm{LDA}} | \Sigma(E_{kn}) - V^{xc} | \psi_{kn}^{\mathrm{LDA}} \rangle \right)$$

where $\epsilon_{kn}^{\mathrm{LDA}}$ and $|\psi_{kn}^{\mathrm{LDA}}\rangle$ are the nth energy level and wave function at wave number k. They are given in the LMTO-ASA in the LSDA. $\Sigma(E)$ is the self-energy in the RPA and V^{xc} is the LSDA exchange-correlation potential. The virtual crystal approximation[5] is employed to simulate "Sr" doping to $LaMnO_3$. This is

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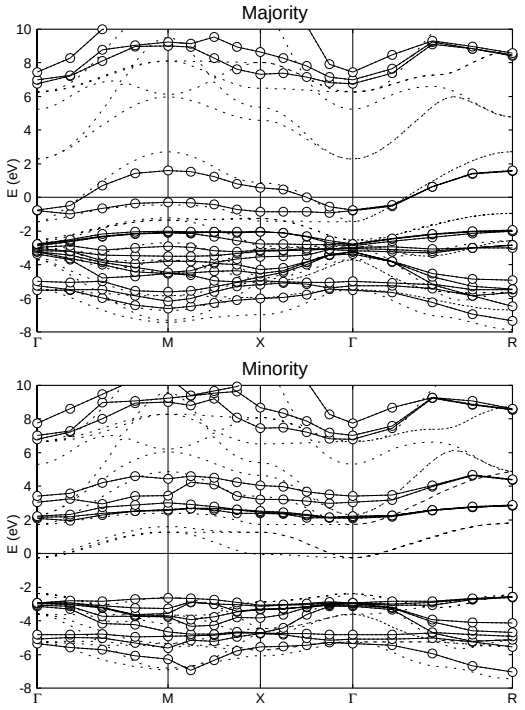

Fig. 1. LSDA eigen-energies (dotted lines) and *GW* quasiparticle energies (circles) for the majority and minority spin states of $La_{0.7}Sr_{0.3}MnO_3$. Solid lines are shown as guidelines.

the all-electron *GW* calculation that includes core electrons.

The calculated LSDA eigen-energies and *GW* quasiparticle energies are shown in Fig. 1.[6] In the LSDA, the Fermi level crosses the majority spin Mn e_g band. However, there is also a small portion of the minority spin Mn t_{2g} state located at the Fermi level, as it was already reported previously.[3] In the *GW* approximation, these states are pushed up by about 2 eV above the Fermi level. Besides, the oxygen $2p$ band is shifted down that additionally increases the bang gap in the minority spin channel. The electron-electron correlations also reduces the band widths. So, the valence majority spin Mn e_g band is shrunken by almost 50%. The conduction minority spin Mn t_{2g} band is shrunken by 68%, as the additional energy splitting between oxygen $2p$ and Mn t_{2g} levels suppresses the hybridization between these two states. Hence, we conclude that the system is a fully spin-polarized half-metallic ferromagnet, at least on the level of virtual crystal approximation.

The change of the Mn potential due to the random distribution of La and Ca atoms can be estimated (in LSDA) as 0.6eV,[7] which is substantially smaller than the new 2eV position of the edge of the minority spin Mn t_{2g} band relative to the Fermi level, obtained in our *GW* calculations. Therefore the system is expected to be fully spin-polarized even in the case of cation disorder.

Finally, our calculations have important implication for the analysis of magnetic phase diagram of $La_{1-x}Sr_xMnO_3$, which can be analyzed in terms of the Mn e_g bandwidth (w) and the intra-atomic exchange splitting at the Mn sites (Δ_{rmex}).[8] According to the *GW* calculations, w becomes smaller (in comparison with the LSDA), while Δ_{rmex} becomes larger due to the additional upward shift of the minority spin Mn t_{2g} and Mn e_g states. Therefore, the ratio w/Δ_{rmex} will further decrease in comparison with LSDA calculations. It will additionally stabilize the ferromagnetic phase for x=0.3 and justify the applicability of the double exchange limit, $w/\Delta_{rmex}\rightarrow 0$ for the analysis of other antiferromagnetic structures which typically appear for larger x.

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[5] Striktly speaking, the virtual crystal approximation can be applyed only to $La_{1-x}Ba_xMnO_3$, and corresponds to the replacement of (La,Ba) sites by pseudoatoms with the fractional atomic number Z=57−x. However, the exact chemical origin of the cation atoms seems to have only small effect on the electronic properties of perovskite manganites, and once the crystal structure is fixed, the alloys of the type (La,Ca), (La,Sr), and (La,Ba) exhibit very similar behavior.[8]

[6] The system is assumed to be cubic with the lattice parameter of 7.32 a.u. The msesh of 8×8×8 k-points is used both in LSDA and *GW* calculations.

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