Effect of orbital degeneracy on magnetic phases of electron-doped manganites

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Abstract

The total energies are evaluated for magnetic phases of electron doped manganites using energy bands E(**k**) accounting for the orbital degeneracy of e_g-band of manganese for the main A-, C-, G- and F-types of spin and orbital order. To determine the magnetic and orbital configuration, a minimization of the ground state energy with respect to the cant angle between the spins of Mn^{4+} ions and e_g orbital angles of the two magnetic sublattices has been performed. A phenomenological account of the small splitting of e_g-level stabilizes the collinear spin structures with the corresponding ferromagnetic orbital order.

Key words: manganite; electron doping; magnetic structure; orbital ordering; phase diagram

1. Introduction

A variety and originality of ground states of the manganites $R_{1-x}A_xMnO_3$ (R is a rare-earth and A is an alcali-earth element) essentially results from strong correlations between the intrinsic degrees of freedom. To understand the connection between the spin and orbital ordering in the electron-doping region $0.5 < x < 1$, the complicated models oriented mainly on the hole-doping region have been extended [1,2].

We involve the model conventionally used for the region $0.5 < x < 1$ which contains the double exchange of degenerate e_g orbitals of Mn ions and the isotropic intersite exchange of t_{2g} local spins. A splitting of the e_g level reasonably introduced regardless of the particular mechanism lifts the orbital degeneracy due to anisotropy of the hopping integals and stabilizes both the magnetic and orbital order (OO). Our calculations accounting self-consistently the spin and orbital degrees of freedom result in the phase diagram well compatible with the experimental observations [3,4].

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2. Model

We use the following effective Hamiltonian:

$$\begin{aligned} H &= H_{DE} + H_m \\ H_{DE} &= \sum_{\langle ij\rangle\alpha\beta\sigma} (t_{ij}^{\alpha\beta} a_{i\alpha\sigma}^{+} a_{j\beta\sigma} + \text{H.c.}) \\ &\quad - J_H \sum_{i\alpha\sigma\sigma'} \mathbf{S}_i \cdot a_{i\alpha\sigma}^{+} \sigma_{\sigma\sigma'} a_{i\alpha\sigma'} \qquad (1) \\ H_m &= \sum_{\langle ij\rangle} J_{ij} \mathbf{S}_i \mathbf{S}_j \,. \end{aligned}$$

The indices α and β denote the double-degenerate e_g states. The term H_m corresponds to the Heisenberg exchange between localized spins. The matrix elements $t_{ij}^{\alpha\beta}$ are the corresponding nearest-neighbour hopping integrals.

The possible OO is accounted by the conventional transformation:

$$\begin{pmatrix} |\alpha\rangle \\ |\beta\rangle \end{pmatrix} = \begin{pmatrix} \cos\frac{\varphi}{2} & \sin\frac{\varphi}{2} \\ -\sin\frac{\varphi}{2} & \cos\frac{\varphi}{2} \end{pmatrix} \begin{pmatrix} |3z^2 - r^2\rangle \\ |x^2 - y^2\rangle \end{pmatrix} \qquad (2)$$

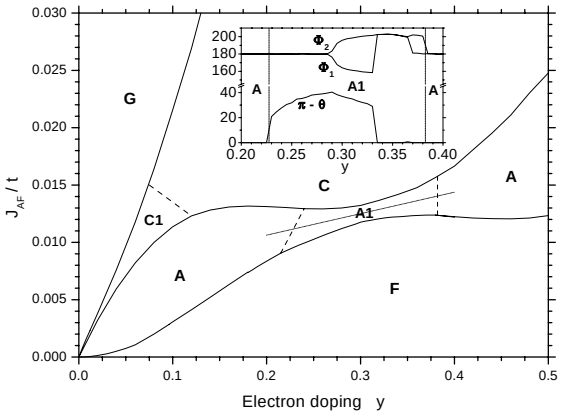

Fig. 1. Phase diagram, $J_H = 2.5t$ and $\Delta = 1.5ty$. On the inset is the behaviour of the cant angle and orbital angles in (deg.) along the dotted line on the diagram.

where $|\alpha\rangle$ and $|\beta\rangle$ are the occupied and empty states, respectively, and φ is the characteristic angle.

The Hamiltonian matrices of the size 8×8 for A, C and G antiferromagnetic (AF) structures are presented in [5]. Here, they are generalized by the transformation (2).

We restrict our model by two-sublattice structures of the types A, C and G. They are characterized by the angle θ between the spins of different ferromagnetic (FM) sublattices and by the orbital angles φ_1 and φ_2 (2) on the ions of the two sublattices. The FM spin order corresponds to the case $\theta = 0$ and the FM OO implies $\varphi_1 = \varphi_2$.

As in electron-doped manganites the e_g orbitals are partially occupied, the splitting of the e_g level depending on doping may occur. We assume the splitting Δ proportional to the electron doping and such that for $LaMnO_3$ $2\Delta = 0.3 - 0.5$eV [6].

The total energy for each AF structure has been minimized by simultaneous variation of the three angles θ, φ_1 and φ_2. The densities of states have been evaluated by summation over the Brillouin zone for the spectra $E(\mathbf{k})$ found by diagonalization of the corresponding matrices of the Hamiltonian (1).

In the calculations we accepted $t = 0.1$eV, $J_H = 0.25$eV and the AF coupling $J_{AF} = 1.5$meV evaluated from the Néel temperature $T_N = 141$K of undoped $CaMnO_3$.

3. Results

The phase diagram calculated for $J_H = 2.5t$ and $\Delta = 1.5ty$, $y = 1 - x$ (Fig.1) exhibits a set of magnetic phases differing by spin and orbital orders. The G phase is a spin-canted AF structure with the FM OO $|3z^2 - r^2\rangle$ ($\varphi_1 = \varphi_2 = 0$), the cant angle $(\pi - \theta)$ is nearly proportional to the doping and monotonously decreases with increasing the exchange coupling. Due to the cubic symmetry this structure retains a triple orbital degeneracy, the FM OO $|3y^2 - r^2\rangle$ ($\varphi_{1,2} = 120^\circ$) and $|3x^2 - r^2\rangle$ ($\varphi_{1,2} = 240^\circ$) are also possible. The AF C phase has a collinear spin arrangement with the FM OO $|3z^2 - r^2\rangle$, the $C1$ area corresponds to a canted C-type structure ($\pi - \theta \sim 20^\circ$) with the same FM OO. Two A areas correspond to the collinear spin structure with the FM OO $|x^2 - y^2\rangle$ ($\varphi_{1,2} = 180^\circ$), they are connected with an A-type isthmus $A1$ where the spin and orbital distortions compete with each other. The inset shows the behaviour of the cant angle $\pi - \theta$ and the orbital angles $\varphi_{1,2}$ with doping along the dotted line on the diagram. It is characterized by different combinations of spin canting $\pi - \theta$ up to 40° and FM OO with $\varphi_{1,2} = 180^\circ$ and 200°, as well as by the admixtures of AF OO with $\varphi_{1,2} \sim 180^\circ \pm 20^\circ$ and $\sim 190^\circ \pm 10^\circ$. F is a FM spin structure with the triple-degenerate FM OO of the type $|3z^2 - r^2\rangle$ similar to that of the phase G.

For the realistic values $J_{AF} = 0.015t$ and $J_H = 2.5t$, the phases G, C, A with the appropriate FM OO replace each other with increasing the doping from 0 to 0.5 which is in good agreement with experimental observations. Any deviation of the Hund coupling from its meaning $J_H = 2.5t$ only slightly shifts the interphase boundaries of Fig.1 and does not violate the general structure of the phase diagram.

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