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Spin and orbital orderings in perovskite manganites

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Abstract

By using the exact diagonalization method on finite-size clusters, the interplay of spin and orbital in the undoped insulating and doped charge-ordered states of perovskite manganites is examined. The spins lift the orbital degeneracy. As a result, the anisotropic antiferromagnetic states are obtained.

Keywords: Orbital wave; Manganese oxides; Exact diagonalization; Spin waves; Transition metal oxides

In recent years, the transport and magnetic properties of perovskite manganites have attracted much attention in connection with the colossal magnetoresistance [1, 2] (CMR). A typical compound which shows CMR is $(La, X)MnO_3$ with X = Sr or Ca. The mother compound, $LaMnO_3$, is a Mott insulator with a layered antiferromagnetic (AF) ordering called A–AF state [3–5]. CMR is also observed in the doped charge-ordered compounds such as $La_{0.5}Ca_{0.5}MnO_3$ and $Pr_{0.5}Ca_{0.5}MnO_3$ [2]. Their charge-ordered phase is accompanied with an AF ordering and is called CE structure [4, 5]. However, it has not been understood why the anisotropic AF orderings such as A–AF state and CE structure emerge, although a key ingredient is considered to be the orbital degree in Mn e_g-orbitals [5–7].

In this paper, we study the interplay of spin and orbital starting with the effective Hamiltonian for the strongly correlated system with orbital degeneracy [7]. By using the exact diagonalization method on finite-size clusters, we obtain the A-, C- and G-type AF states. We also examine the interplay in the 50% doped charge-ordered states. Our results show that the spins lift the orbital degeneracy, resulting in the anisotropic magnetic AF states.

We take the effective Hamiltonian [7] as follows:

$$
\begin{aligned}
H = {} & V \sum_{\langle ij \rangle} n_i n_j + K \sum_i \boldsymbol{S}_i \cdot \hat{\boldsymbol{S}}_i + \sum_{\langle ij \rangle, ab, \sigma} t_{ij}^{ab} \tilde{c}_{ia\sigma}^{\dagger} \tilde{c}_{jb\sigma} \\
& + J_{AF} \sum_{\langle ij \rangle} \hat{\boldsymbol{S}}_i \cdot \hat{\boldsymbol{S}}_j - \frac{1}{U' - J'} \sum_{\langle ij \rangle} \left(\tfrac{3}{4} n_i n_j + \boldsymbol{S}_i \cdot \boldsymbol{S}_j\right) \\
& \times [2(t_{ij}^{aa^2} + t_{ij}^{bb^2})(\tfrac{1}{4} n_i n_j - T_i^z T_j^z) \\
& - 2t_{ij}^{aa} t_{ij}^{bb} (T_i^+ T_j^- + T_i^- T_j^+) \\
& + 2(t_{ij}^{ab^2} + t_{ij}^{ba^2})(\tfrac{1}{4} n_i n_j + T_i^z T_j^z) \\
& - 2t_{ij}^{aa} t_{ij}^{bb} (T_i^+ T_j^- + T_i^- T_j^+) \\
& - 2(t_{ij}^{ab} t_{ij}^{aa} - t_{ij}^{ba} t_{ij}^{bb})(T_i^z T_j^+ + T_i^z T_j^-) \\
& - 2(t_{ij}^{ba} t_{ij}^{aa} - t_{ij}^{ab} t_{ij}^{bb})(T_i^+ T_j^z + T_i^- T_j^z)]. \qquad (1)
\end{aligned}
$$

Here, V is the nearest-neighbor Coulomb repulsion, and K is the Hund coupling between e_g spin

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($\boldsymbol{S}_i$) and t_{2g} spin ($\hat{\boldsymbol{S}}_i$). t_{ij}^{ab}'s are the transfer integrals between neighboring e_g orbitals where a and b denote the orbital states. $\tilde{c}_{ia\sigma}$ denotes the annihilation operator of an electron with spin σ at the a orbital of the site i where the double occupancy at the site is excluded. J_{AF} is the superexchange interaction between t_{2g} spins. U' and J' are the Coulomb interaction and the Hund coupling between e_g orbitals in a Mn site, respectively. T_i^z, T_i^+, and T_i^- are the pseudo-spin operators which denote the orbital degree of freedom and are expressed as spin-$\frac{1}{2}$ operator.

Let us first examine the spin and orbital structures in the undoped $LaMnO_3$. In the calculation, we introduce the AF interaction J_{AF} between neighboring e_g spins with $S = \frac{1}{2}$ and neglect t_{2g} spins since the t_{2g} spins strongly couple to the e_g spins through the Hund coupling. Then, it is possible to apply the Lanczös exact-diagonalization method and calculate the correlation functions of spins and orbitals in a cluster of 8 site die. The ground state is characterized by the parameter J_{AF} in units of $(t_0)^2/(U' - J')$ where t_0 is the transfer integral between neighboring $x^2 - y^2$ orbitals in the x-direction. The spin and orbital structures are obtained by examining the spin and pseudo-spin correlation functions. When J_{AF} increases, we find that the ferromagnetic (F), and A-, C- and G-type AF states occur successively. In the F-, and C- and G-type AF states, the $x^2 - y^2$ and $3z^2 - r^2$ orbitals are aligned alternately in all directions. On the other hand, in the A–AF state, the orbital state changes together with spins: The $3x^2 - r^2$ and $3y^2 - r^2$ ordering develops and the A–AF state is realized in a certain region of J_{AF} as seen in Fig. 1(a). We have also studied the phase diagram in the mean field theory, and found that the A–AF state is not stabilized in contrast with the numerical results. Note that in the exact diagonalization method, the A–AF state is obtained without lattice distortion. This fact shows that the quantum interplay of spin and orbitangular moments is important for the A–AF state.

Let us next study the spin and orbital orderings in the 50% doped charge-ordered states. In the calculation, the e_g and t_{2g} spins must be dealt with separately. Here, we assume that an t_{2g} spin is $\frac{1}{2}$, for simplicity. The parameter values are taken to be $t_0/(U' - J') = 0.1$, and $V = K = 10t_0$. We obtain

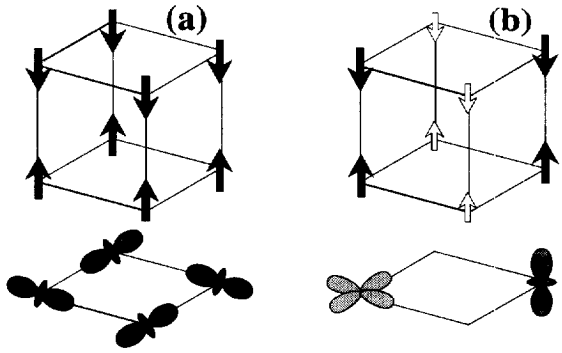

Fig. 1. Schematic figures of spin and orbital orderings in the undoped insulating (a) and 50% doped charge-ordered (b) states. For the orbital orderings, the in-plane structures are presented. In Fig. 1(b), white and black arrows indicate the Mn^{4+} and Mn^{3+} spins, respectively.

that F, AF and ferrimagnetic spin states occur with increasing J_{AF}. The ground states are almost degenerate due to the orbitals. However, we find that the degeneracy is lifted in the A–AF state as in the case of the undoped insulator. This is because of the coupling between spin and orbital. The charge, spin and orbital coupling orderings in the state are shown in Fig. 1(b).

Summarizing, we have examined the interplay of spin and orbital in the undoped insulating and doped charge-ordered states in the perovskite manganites. In the undoped insulating states, we found that the A-, C- and G-type AF orderings occur together with the orbital orderings. We also examined the interplay in the 50% doped charge-ordered states. Our results show that the coupling between spin and orbital lifts the degeneracy and derive the anisotropic magnetic AF states.

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