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Magnetic and orbital excitations in manganese oxides

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Abstract

The magnetic and orbital structures in manganese oxides with perovskite structure are examined by using the exact diagonalization method on finite-size clusters. The orbital degeneracy in the e_g states is taken into account based on the effective Hamiltonian derived in the insulating state with strong Coulomb interaction. It is shown that A-, C- and G-type antiferromagnetic orderings occur as a result of competition or cooperation of the superexchange interaction of t_{2g} spins and the coupling between spins and orbitals. The spin and orbital excitations are also examined.

Keywords: Spin and orbital wave; $LaMnO_3$; Exact diagonalization

Since the discovery of colossal magnetoresistance (CMR) in manganese oxides with perovskite structure, the electronic and magnetic properties have attracted much attention in recent years [1]. Among them is the A-type antiferromagnetic (A-AF) ordered state in which the alignment of Mn spins in the c-direction is antiferromagnetic (AF) and that in the ab-plane is ferromagnetic (F) in the mother compound of the CMR systems, i.e., $LaMnO_3$. Several attempts to describe the anisotropic ordering have been made [2, 3]. A key to derive the A-AF state is the orbital ordering of e_g states in Mn ions. In $LaMnO_3$, the $3d(3x^2 - r^2)$ and $3d(3y^2 - r^2)$ orbitals order alternately in the ab-plane. Such an orbital ordering is accompanied with Jahn–Teller distortion [4]. However, the interplay of spin and orbital orderings has not been examined in detail in the insulators with strong Coulomb interaction.

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In this paper, we study the interplay, starting with the effective Hamiltonian of spin and orbital degrees of freedom derived in the insulating states with strong Coulomb interaction. By using exact diagonalization method of finite-size clusters, we find that the A-, C- and G-type AF orderings occur as a result of competition or cooperation of the superexchange interaction of neighbouring t_{2g} spins and the coupling between spin and e_g orbits. Our numerical results show that the orbital degeneracy is lifted by the spins. Then, the anisotropic AF orderings such as A-AF emerge. We also calculate the spin and orbital excitation spectra and find that the orbitals induce the continuous spectra in the spin excitation.

Let us first write the effective Hamiltonian in the e_g states as

$$H_I = -\frac{1}{U' - J'} \sum_{\langle ij \rangle} \left(\tfrac{3}{4} n_i n_j + S_i \cdot S_j\right) \times 2\left[\left(t_{ij}^{aa^2} + t_{ij}^{bb^2}\right)\left(\tfrac{1}{4} n_i n_j - T_i^z T_j^z\right)\right.$$

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$$
\begin{aligned}
&- 2t_{ij}^{aa}t_{ij}^{bb}(T_i^+ T_j^- + T_i^- T_j^+) \\
&+ 2(t_{ij}^{ab^2} + t_{ij}^{ba^2})(\tfrac{1}{4}n_i n_j + T_i^z T_j^z) \\
&- 2t_{ij}^{ab}t_{ij}^{ba}(T_i^+ T_j^+ + T_i^- T_j^-) \\
&- 2(t_{ij}^{ab}t_{ij}^{aa} - t_{ij}^{ba} - t_{ij}^{bb})(T_i^z T_j^+ + T_i^z T_j^-) \\
&- 2(t_{ij}^{ba}t_{ij}^{aa} - t_{ij}^{ab}t_{ij}^{bb})(T_i^+ T_j^z + T_i^- T_j^z)].
\end{aligned} \tag{1}
$$

Here, U' and J' are the Coulomb interaction and the Hund coupling between two e_g orbitals in a Mn site, respectively. S_i is spin-$\frac{1}{2}$ operator in e_g orbitals at site i. 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. The two eigenstates of T_i^z correspond to the two e_g orbitals. t_{ij}^{ab}'s are the transfer integrals between neighboring e_g orbitals where a and b denote the orbital states, i.e., t_{ij}^{ab} is the transfer integral between a orbital at site i and b at j. Eq. (1) induces the F interaction between neighboring spins. The effective Hamiltonian also includes the AF term between e_g spins. However, since the term may be absorbed in the AF interaction between t_{2g} spins, the term is not given here. The t_{2g} spins couple to the e_g spins through the strong Hund coupling. Since we are interested in the ground states and the low-energy excitation, we introduce the AF interaction J_{AF} between neighboring e_g spins with $S = \frac{1}{2}$ and neglect t_{2g} spins. Therefore, J_{AF} is about ten times larger than the superexchange interaction between neighboring t_{2g} spins with $S = \frac{3}{2}$. Then, the Hamiltonian is written as

$$
H = H_I + H_{AF},
$$

$$
H_{AF} = J_{AF} \sum_{\langle ij \rangle} S_i \cdot S_j. \tag{2}
$$

This Hamiltonian consists of the competitive two terms: H_I and H_{AF} cause the F and AF interactions between neighboring spins, respectively. This competitive relation depends on the orbital degree of freedom, resulting in a variety of magnetic structures.

Let use next examine the magnetic and orbital structures. We employ the Lanczös exact-diagonalization technique and calculate the correlation functions of spins and orbitals in a small cluster of 8 site die. The ground state of the Hamiltonian (2) is characterized by a 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. For $J_{AF} = 0$, the total spin quantum number takes the maximum value, which correspond to F state. With increasing J_{AF}, the value of total spin quantum number changes to zero around $J_{AF} \sim 1$.

The magnetic structures are obtained by the spin correlation function $S(\boldsymbol{q})$. In the region of $1 < J_{AF} < 1.4$, the correlation function becomes the largest at $\boldsymbol{q} = (0, 0, \pi)$. (Of course, the symmetry of Hamiltonian (2) results in the relation $S(\pi, 0, ,0) = S(0, \pi, 0) = S(0, 0, \pi)$ and $S(\pi, \pi, 0) = S(0\,\pi, \pi) = S(\pi, 0, \pi)$.) This result denotes the A-AF structure. In the region of $1.4 < J_{AF} < 1.5$, on the other hand, the correlation function takes the largest at $\boldsymbol{q} = (\pi, \pi, 0)$. In this case, the AF spin alignment is the ab-plane and the F one is along the c-axis. In other words, the so-called C-type antiferromagnetic ordered structure appears in the region. In the region of $1.5 < J_{AF}$, the correlation function is the largest at $\boldsymbol{q} = (\pi, \pi, \pi)$, i.e. the G-type magnetic structure is obtained.

What kind of orbital structures are realized in these magnetic states? In order to examine the orbital structures, we calculate the pseudo-spin correlation functions in the exact diagonalization method. The orbital and magnetic structures derived from the calculation are shown schematically in Fig. 1. 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. The relation between spin and orbital orderings in these three states is understood as follows: In this orbital ordering, the F spin interaction is induced in H_I. The interaction, however, is

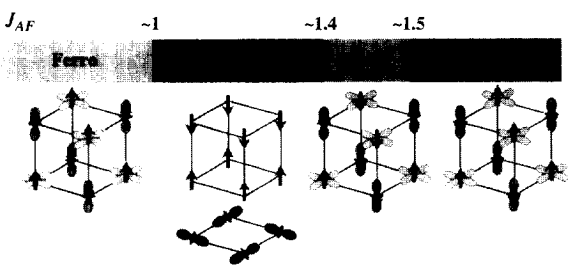

Fig. 1. Phase diagram of the magnetic and orbital structures. J_{AF} is in units of $(t_0)^2/(U' - J')$ where t_0 is the transfer integral between neighboring x^2–y^2 orbitals in the x-direction. In the A-AF state, the in-plane orbital state is illustrated.

weaker in the *ab*-plane than along the *c*-axis. Therefore, when J_{AF} increases, the F coupling in the *ab*-plane is overcome by J_{AF} and the C-type AF state appears. With further increasing J_{AF} keeping the orbital ordering, the G-type AF state appears. With further increasing J_{AF} keeping the orbital ordering, the G-type AF state is stabilized. It is also possible that the orbital state changes together with spins: When the $3x^2-r^2$ and $3y^2-r^2$ ordering develops, the A-AF state is realized in a certain region of J_{AF} as seen in Fig. 1. We have also studied the phase diagram in the mean field theory, and found that the A-AF state is not stabilized unless the lattice distortion due to the Jahn–Teller effect is introduced. Note that in the exact diagonalization method, the A-AF state obtained without the lattice distortion. Thus, the A-AF state may be considered to be the quantum one induced by the coupling between spins and orbitals.

Finally, let us consider the spin and orbital excitations. Fig. 2 shows the spin excitation spectrum $S(\boldsymbol{q}, \omega)$ *at* $J_{AF} = 1.2$ where the A-AF is stabilized. The solid and broken curves show the spin wave dispersion calculated in the spin wave approximation. The dispersion in the approximation agrees with the numerical results. The competition and

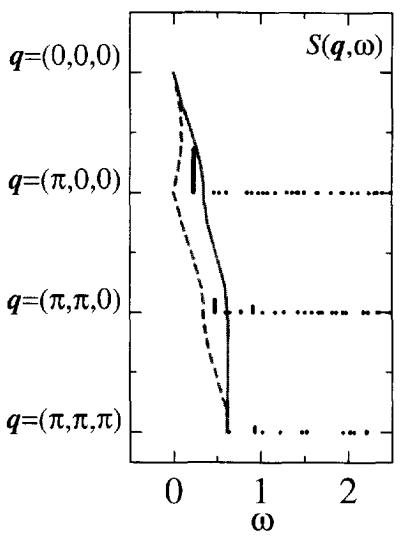

Fig. 2. Spin excitation spectrum $S(\boldsymbol{q}, \omega)$ at $J_{AF} = 1.2$ where the A-AF is stabilized. The solid and broken curves show the spin wave dispersion in the in-plane and out-of-plane directions, respectively, in the spin wave approximation.

cooperation of orbital and spin interactions depresses the dispersion. We also find the continuous spectra in the numerical results. They come from the coupling of spins and orbitals excitations.

Summarizing, we have examined the interplay of the spin and orbital orderings in the insulating state with strong Coulomb interaction. In our numerical results, we found that the A-, C- and G-type AF orderings occur as a result of competition of cooperation of the spin and orbital degrees of freedom. Especially, the A-AF state appears in cooperation of the orbital and spin ordering. This is because the coupling between spins and orbitals lifts the orbital degeneracy in the A-AF state. On the other hand, the state is not obtained in the mean field theory unless the strong Jahn–Teller distortion is introduced. We have also investigated the spin and orbital excitation spectra. The orbital excitation induces the continuous spectra in the spin excitation.

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