Magnetic phase diagram and electronic phase separation in manganites

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Abstract

A detailed study on magnetic phase diagram and electronic phase separation has been performed in a mean field approximation for low carrier density manganites. It is shown that the phase separation is rather dependent of the values of Hund coupling, and it washes away the canted G-type AF. The C-type AF and CE-type AF, however, are stable against the phase separation. Observed electron-density dependence of magnetization may well be explained by taking into account weaker Hund coupling or electron-density dependence of J.

Key words: Perovskite manganites; Double-Exchange model; Orbital ordering; Jahn-Teller distortion

Perovskite manganites $R^{+3}_{1-x}A^{+2}_{x}\mathrm{MnO_3}$ have attracted much interest in the magnetic and transport properties.[1] In particular, discovery of the colossal magnetoresistance in room temperature has intensified experimental and theoretical efforts to understand and quantify the correlation between spin, charge and orbital orderings.

Recently, it is reported that electronic phase separation (PS) occurs in low electron-density manganites with the peculiar electron-density $n(=1-x)$ dependence of magnetization $M(n)$ in which the magnetization increases first, with increasing n, but vanishes around $n \sim 0.2$. [2] On the other hand, quarter filled manganites show a complex ordering called CE-type AF state. In order to understand the stability of the CE-type AF phase, several theoretical studies have been carried out, although their role in the stability of the CE-type AF state may be still controversial. The purpose of the present work is to calculate the magnetic phase diagram by taking into account the orbital ordering, Coulomb interaction, canted spin ordering and PS simultaneously, and to obtain deeper understanding on the magnetic properties of low electron-density manganites. In particular, we focus our attention on the peculiar $M(n)$ and stabilization of CE phase of quarter filled manganites. The Hamiltonian that we adopt is written as;

$$
\begin{aligned}
H = & \sum_{<ij>\mu\nu\sigma} t_{ij}^{\mu\nu} c_{i\mu\sigma}^{\dagger} c_{j\nu\sigma} - K \sum_{i\mu\sigma\sigma'} \mathbf{S}_i \cdot c_{i\mu\sigma}^{\dagger} \mathbf{s} c_{i\mu\sigma'} \\
& + U \sum_{i\mu} n_{i\mu\uparrow} n_{i\mu\downarrow} + U' \sum_{i\sigma\sigma'} n_{i\mu\sigma} n_{i\nu\sigma'} \\
& + V \sum_{<ij>} n_i n_j + J \sum_{<ij>} \mathbf{S}_i \cdot \mathbf{S}_j + g \sum_{i\mu\nu\sigma} c_{i\mu\sigma}^{\dagger} c_{i\nu\sigma}. \quad (1)
\end{aligned}
$$

Here $t_{ij}^{\mu\nu}$ represents the hopping of e_g electrons between orbital μ on site i and orbital ν on site j; K, U, U', and V stand for the Hund coupling between e_g electrons and localized t_{2g} spins, intra-, inter-orbital Coulomb repulsion, and n.n. Coulomb repulsion, respectively; J is the antiferromagnetic (AF) interaction between localized spins; and g characterizes the effect of Jahn-Teller distortion on the orbitals. $\mathbf{S}$ stands for the spins of localized t_{2g} spins, $\mathbf{s} = \boldsymbol{\sigma}/2$.

Below, we carry out the numerical calculations for 2D systems, keeping in mind the experimental results for layered manganites.[3,4] We treat Coulomb interaction U, U', V terms only quarter filled manganites, because they are less relevant for low electron-density

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manganites. When we consider the low electron-density manganites, we further assume a usual alternative type (G-), stripe type (C-) and their canting spin ordering, and their propagating vectors are along [1,1] and [1,0] directions of the square lattice, respectively. In quarter filled manganites, we assume that localized spins align ferromagnetic (F) along zigzag chains and form a spin-spiral structure between zigzag chains. The propagating vector of the spiral state is in the [1,1] direction of the square lattice.

Fig. 1(a) shows the $J-n$ phase diagram with $K/t = 4$ and $g/t = 0$ (solid curves).[5] We see that the spin G-AF states is stabilized because the gain in the AF exchange energy between t_{2g} spins exceeds the gain in the band energy in the F state. With increasing n, the G-Cant state is stabilized as the gain in the band energy increases. When the gain in the band energy exceeds the AF exchange energy, the F state is stabilized. The C-AF state appears only in a region where $0.2 \leq n \leq 0.35$ with $J/t \geq 0.02$. Fig. 1(b) indicates that the G-Cant state, however, is unstable against the PS, and as a result, phase mixing of the G-AF and C-AF states and that of the G-AF and F states appear $J/t > 0.02$, $J/t < 0.02$, respectively. The line of CE-type AF states is taken into account Coulomb interaction. This state show a charge ordering of checker-board type due to anisotropic hopping of e_g electrons and strong on-site Coulomb repulsion.

By introducing lattice distortion g, the orbital ordering appears in such a way that the Jahn-Teller distortion is favorable to lower the kinetic energy. The orbital orderings for $g/t = 0.3$ are shown in Fig. 1 with new boundaries (broken curves) between the C-AF, G-Cant and F states. In contrast, the CE-type AF state appears in the region with $J/t > 0.02$, irrespective of the lattice distortion. When $K/t = 2$, the C-AF state extends to regions with smaller J as shown by a chained curve in Fig. 1(a). The phase diagram of ground states with $K/t = 2, g/t = 0.3$ is shown as Fig. 1(c).

$M(n)$ is calculated by using the canting angle between localized spins for various sets of parameter values, and the results are shown in Fig. 2. The observed magnetization increases first, with increasing n, reaches a maximum value of ~ 0.5 at $n \sim 0.08$, and vanishes around $n \sim 0.2$. Because value of J/t may be estimated to be ~ 0.01 from the Néel temperature 110K of $CaMnO_3$, the experimental results may be explained by taking smaller values of K/t and/or by assuming n-dependence of J/t.

In conclusion, the CE-type AF and C-AF are stable against the phase separation. Observed n-dependence of magnetization may well be explained by taking into account weaker Hund coupling or n-dependence of J. We have also discussed that the magnetic phases of manganites at quarter filling taking into account Coulomb interactions and found that the CE-type AF

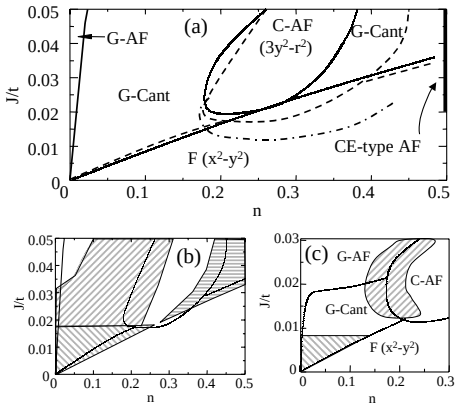

Fig. 1. (a) Phase diagram of the ground state with $K/t = 4, g = 0$ (solid curves), $K/t = 4, g/t = 0.3$ (broken curve) and $K/t = 2, g/t = 0.3$ (chained curve). (b) The shaded areas shows the PS regions of $K/t = 4, g/t = 0$. (c) A phase diagram with $K/t = 2, g/t = 0.3$ and the PS regions.

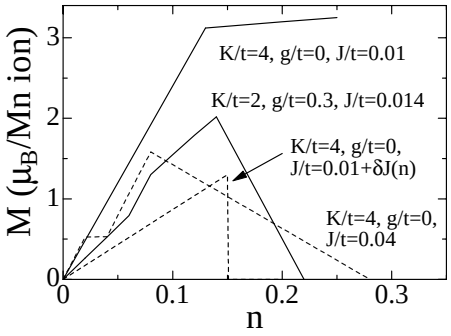

Fig. 2. Calculated results of $M(n)$ by taking into account PS.

state is stabilized in the region where J/t is large. The present results are qualitatively consistent with the observed phase diagram of the quarter-filled manganites.

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