Coulomb Interaction and Instability of CE-Type Structure in Half-Doped Manganites

In their Letter, den Brink *et al.* [1] proposed theoretically that the one-dimensional ferromagnetic zigzag chains in the CE phase in half-doped manganites play an essential role in forming the orbital ordering, and, more surprisingly, the on-site Coulomb interaction U between electrons with different orbitals leads to experimentally observed charge ordering. In this Comment, I point out that the strong U will destroy the stability of CE-type phase.

The same notations as in Ref. [1] are employed here. In Ref. [1], the case of $U = 0$ was analyzed and the CE phase was found to be stable when the superexchange coupling $J > 0.1524t$ (t is the hopping integral). In the CE phase the one-dimensional zigzag chain consists of two types of lattice sites, the bridge and corner sites. The bridge site is occupied singly by electrons with certain orbitals while the corner sites can be occupied doubly. Without U, the charge distribution along the zigzag chain is uniform, i.e., $1/2$. Finite U will push the charge away from the corner sites to the bridge sites to form charge ordering. The theory explains the patterns of three orderings almost perfectly. We observe that U will also increase the energy of the CE phase. The rod-type (i.e., C-type) phase consists of one-dimensional ferromagnetic straight chains. The chains are occupied, at most, singly on each site. Thus U does not change the energy of C-type phase. This fact leads to the instability of the CE phase with respect to C-type phase for finite U. We evaluate the electronic energy for the CE phase by using an exact diagonalization method. The energies from the magnetic coupling are the same for both C-type and CE phases. At $U = 0$, $E_{\rm kin}^{\rm CE} = -0.6953t$ for CE-type phase and $E_{\rm kin}^{C} = -0.6366t$ for C-type phase. However, at $U = +\infty$, $E_{\rm kin}^{\rm CE} = -0.61t$ and the energy of C-type phase remains unchanged. In other words, the CE phase is unstable with respect to C-type phase when electron correlation becomes stronger. Numerical results for energies of the CE phase via U is shown in Fig. 1(a). The finite-size effect is very small [2]. At about $U \approx 5t$, CE-type phase has a higher energy than C-type phase. To establish a comprehensive phase diagram, we have to consider other possible phases. Except for conventional antiferromagnetic and ferromagnetic phases, a novel type of phase is taken into account. The phase consists of 2-site ferromagnetic valence bonds (VB), and the bonds couple antiferromagnetically. Electrons are localized within the bonds. The electronic part of the state in a valence bond is $\frac{1}{\sqrt{2}}(c_{i,\alpha,\sigma}^{\dagger} + c_{j,\alpha,\sigma}^{\dagger})\,|0\rangle$, and the local spins are parallel to the spin of the electron. The energy per site is $E_{\rm kin}^{\rm VB} + E_{\rm mag}^{\rm VB} = -0.5t - J$, which is independent of U (we take the magnetic energy as zero for the CE phase). The ferromagnetic phase has energy $-t + 2J$ for three dimensions and $-0.919t + 2J$. Thus, when $U = 0$ and $0.152t \le J \le 0.195t$ for three dimensions and $0.117t \le J \le 0.195t$ for two dimensions, the CE phase has a lower energy. The phase diagrams for $U = 0$ and $U = +\infty$ as shown in Figs. 1(b) and 1(c) are evaluated on a 4×4 lattice with a periodic boundary condition by means of a combination of mean field theory and exact diagonalization. We assume the magnetic structure and calculate the energy by exact diagonalization. FM and C-AF are assumed to contain two sublattices and evolve into G-AF when spins on two sublattices are not parallel for large J. Thus, the CE phase does not appear in the ground state for the model in Ref. [1] in the case of strong correlation.

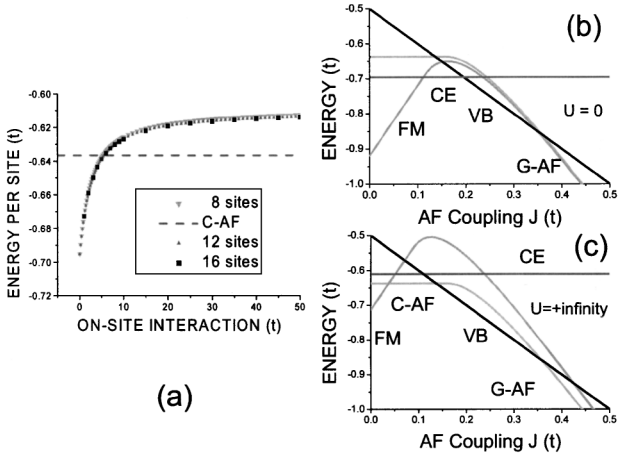

FIG. 1. (a) The electronic energy of the CE phase per site via the on-site interaction U; (b) and (c) the phase diagrams on a square lattice for $U = 0$ and $+\infty$, respectively.

In doped manganites, U is usually estimated to be much larger than t. In Ref. [1], $U \approx 10t$. The on-site U alone can produce the experimentally observed pattern of charge ordering, but cannot stabilize the CE phase. To solve this issue finally, we have to take into account other interactions. For example, the effect of finite large J_H leads to an attractive particle-hole interaction, which favors stabilization of the charge ordering [3].

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[1] J. van den Brink, G. Khaliullin, and D. Khomskii, Phys. Rev. Lett. **83**, 5118 (1999).
[2] The finite-size effect for $E_{\rm kin}^{C}$ is relatively large. $E_{\rm kin}^{C}(12) = -0.6220t$ while $E_{\rm kin}^{C}(+\infty) = -0.6366t$. Thus the calculated value U_c from Fig. 1(a) should be more reasonable than that from 16 sites.
[3] S. Q. Shen and Z. D. Wang, Phys. Rev. B **59**, 14 484 (1999); **61**, 9532 (2000).

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