

Comment on “Charge-Orbital Stripe Structure in $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$ ($x = 1/2, 2/3$)”

The origin of charge and orbital ordering (COO) observed in manganites is currently under much discussion. In a recent Letter [1], Mutou and Kontani (MK) claimed that the origin of this COO was due to the long-range Coulomb interaction V . In this Comment, it is pointed out that the charge structure observed in experiments contradicts such a claim, and that the complicated nature of the phenomenon does not allow for an interpretation as simple as presented in their Letter.

(1) MK propose that COO in manganites originates from V . If this were correct, the NaCl-type charge ordering should occur. In the cubic perovskite manganite, however, COO in the a - b plane simply stacks along the c axis [2]. Thus, V cannot be the only origin of COO in manganites. In fact, it has been argued based on experimental results [3] that V is not a dominant driving force for COO formation even in the single-layered manganite.

(2) MK construct their Hamiltonian using $d_{3x^2-r^2}$ and $d_{3y^2-r^2}$ orbitals as e_g electronic basis. However, they are not orthogonal, as MK themselves mentioned. This choice of basis must be employed with care [4]. Corrections accounting for this nonorthogonality problem may be important [5].

(3) In MK's phase diagrams, $U \gg t_0$ and $V \geq t_0$ are needed to explain COO for both $x = 1/2$ and $2/3$. Here t_0 is the hopping amplitude between neighboring $d_{3x^2-r^2}$ orbitals and U is the on-site interorbital Coulomb repulsion. The former condition is indeed satisfied, since $U \sim 5$ eV and $t_0 \sim 0.5$ eV. As for V , if its bare value 3.6 eV is used, then $V/t_0 \gg 1$. With such a large V , however, it is hard to understand how manganites can have anything but charge-ordered states at all densities, and more specifically, the appearance of the metallic ferromagnetic (FM) phase would be excluded. However, in the real materials V is much smaller than the bare value. Simply using the bulk dielectric constant from experiments (20 ~ 50) [6] provides a lower limit for V/t_0 of the order of 0.1. When the Madelung energies between the NaCl-type and charge-stacked structures are compared [7], an estimate of this ratio is somewhat increased to 0.5, but still a reasonable estimate will be $V < t_0$.

(4) After a variety of calculations and experiments in manganites, it is becoming clear that the emergence of complex structures such as COO can be more satisfactorily explained through a delicate balance among two or more competing interactions. In fact, a model including Hund's rule coupling, Jahn-Teller distortion, and Coulomb interaction is already known to reproduce the so-called CE-type antiferromagnetic (AFM) phase with the charge-stacked COO at $x = 1/2$ [8]. This CE-state stabilization is induced by the kinetic energy of e_g electrons, whose motion is restricted by the t_{2g} -spin alignment through the double-

exchange mechanism. More precisely, at $x = 1/2$, the charge stacking is concomitant with the CE-type phase, while the NaCl-type charge ordering appears in the C-type AFM state. An essential difference between those two AFM phases is the “topology” of the one-dimensional FM paths of t_{2g} spins; straight path for C-type and zigzag path for CE-type. The special properties of the latter causes the energy gain of the charge-stacked CE-type phase [9]. This scenario provides a possible explanation for the stabilization of the CE phase that deserves further study.

In conclusion, the formation of the complex COO patterns in manganites is not simply understood by the MK's scenario based on V .

T. Hotta,¹ E. Dagotto,² H. Koizumi,³ and Y. Takada¹

¹Institute for Solid State Physics

University of Tokyo

5-1-5 Kashiwa-no-ha

Kashiwa, Chiba 277-8581, Japan

²National High Magnetic Field Laboratory

and Department of Physics

Florida State University

Tallahassee, Florida 32306

³Faculty of Science

Himeji Institute of Technology

Kamigori, Ako-gun, Hyogo 678-1297, Japan

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- [9] The topological idea can also explain the competition between the Wigner-crystal and bistrripe type structures in the a - b plane [T. Hotta *et al.*, Phys. Rev. Lett. **84**, 2477 (2000)], experimentally observed at $x = 2/3$ and $3/4$ [S. Mori *et al.*, Nature (London) **392**, 473 (1998)], and predict that the charge-stacking along the c axis occurs only in the bistrripe phase.