

# Ordered Phases and Magnetic Excitations in Doped Manganites

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We present theoretical models concerning magnetic and charge ordering in half-doped manganites. The new approach to microscopic description of this group of compounds, taking into consideration the oxygen degrees of freedom, is discussed. The spin wave dispersion relations are shown both in the model of effective spins on Mn ions and in the new approach when two Mn ions form a large effective spin  $S = 7/2$ .

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## 1. Introduction

The perovskite manganites of  $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$  type are characterized by rich phase diagrams. The microscopic mechanism responsible for the coexisting spin, charge, and orbital order, in particular the stability of so-called *CE*-phase (Fig. 1) with zigzag ferromagnetic (FM) chains and antiferromagnetic (AF) couplings between them, is still under debate. It is now well established, however, that not only strong Coulomb interactions, but also the Jahn–Teller (JT) effect play an important role in stabilizing the *CE*-phase [1].

Recent experiments suggest that realistic theoretical models should include explicitly oxygen degrees of freedom. One of them, supported by the structural data [2], assumes the formation of Mn–O–Mn Zener polarons. The unrestricted Hartree–Fock calculations support the above picture, predicting a strong magnetic coupling within polarons which leads to large effective  $S = 7/2$  spins; the values of exchange constants imply the ground state with FM (AF) order for the directions

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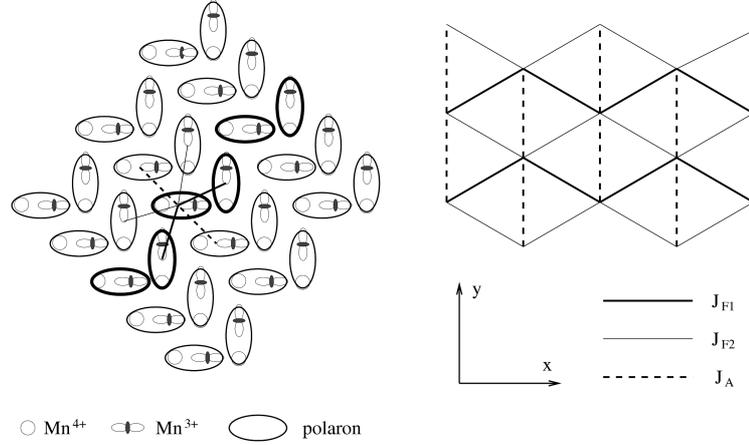


Fig. 1. The orbital scheme of the charge ordered  $CE$ -phase and the polaron model of this phase (left); spins along zigzag chains (one zigzag is marked bold) are ferromagnetically ordered, and there is an antiferromagnetic spin arrangement between chains, the planes are stacked in  $z$  direction. The right part shows the triangular lattice resulting from the polaron model.

corresponding to  $(a, b)$  planes ( $c$  axis) [3]. The picture of Zener polarons competes with another model, in which a lattice of effective Mn spins is considered. Recently, the spin wave spectrum has been studied [4] within the above approach. Both charge ordered (CO) phase and the “dimer” one were considered. The dimer model may be seen as intermediate between the conventional microscopic model of the charge ordered  $CE$ -phase and the Zener polarons picture: there is no charge ordering and Mn spins form pairs with strong interaction within the pair (similarly to the polaron model), however, the Mn ions are still treated as separate effective spins with  $S = 7/4$ .

## 2. Results and discussion

The linear spin waves spectrum has been calculated for the charge ordered phase and the dimer model, as well as for the polaron triangular lattice (Fig. 1). We use the Holstein–Primakoff transformation of spin operators:

$$S_i^z = S - a_i^+ a_i, \quad S_i^+ \simeq \sqrt{2S} a_i, \quad S_i^- \simeq \sqrt{2S} a_i^+ \quad (1)$$

and diagonalize the Heisenberg Hamiltonian:

$$H = \sum_{ij} J_{ij} \mathbf{S}_i \mathbf{S}_j, \quad (2)$$

in quadratic approximation [5].

## 2.1. The model of effective spins on Mn ions

We consider an effective Mn spins lattice with nearest neighbors (n.n.) ferromagnetic coupling along the zigzag chains and antiferromagnetic coupling between n.n. spins belonging to different zigzags both in plane and along  $z$  direction (the planes are stacked along  $z$ , and above a zigzag with spins “up” there is a zigzag with spins “down”). For the charge ordered phase, all values of coupling constants are equal  $J_F = J_A = 1$  ( $J_A$  is taken as an energy unit) and values of spins

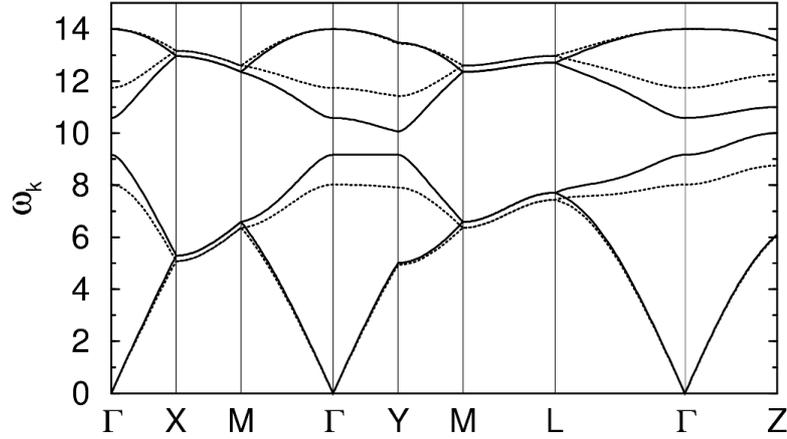


Fig. 2. Spin wave dispersion for the effective Mn spins lattice; the charge ordered (CO) phase (solid) and the dimer model (dotted). High symmetry points:  $\Gamma = (0, 0, 0)$ ,  $X = (\pi/\sqrt{8}, 0, 0)$ ,  $M = (\pi/\sqrt{8}, \pi/\sqrt{8}, 0)$ ,  $Y = (0, \pi/\sqrt{8}, 0)$ ,  $L = (\pi/\sqrt{8}, \pi/\sqrt{8}, \pi/2)$ , and  $Z = (0, 0, \pi/2)$  (the distance between n.n. Mn spins is a unit, zigzags are oriented along  $x$  axis). Parameters: the CO phase:  $J_F = J_A = 1$ ,  $S_b = 2$ ,  $S_c = 3/2$ ; the dimer phase:  $J_{Fs} = 1.5$ ,  $J_{Fw} = 0.5$ ,  $J_A = 1$ ,  $S = 7/4$ .

alternate along zigzags: the “corner” spins ( $\text{Mn}^{4+}$ )  $S_c = 3/2$  and the “bridge” spins ( $\text{Mn}^{3+}$ )  $S_b = 2$ . In the dimer phase all spins are equal  $S = 7/4$ , but the values of ferromagnetic coupling constants alternate along zigzags: the interaction is stronger within the “polarons”. We assume the values of the stronger ( $J_{Fs}$ ) and weaker ( $J_{Fw}$ ) coupling constant equal to 1.5 and 0.5, respectively, and compare the spin wave spectrum obtained to that resulting from the CO model. There are four Mn spins in a unit cell and four spin wave excitation energies were obtained for each  $\mathbf{k}$  vector. The results (Fig. 2) for these two spin arrangements are similar (especially for low energy excitations).

## 2.2. The polaron model

The polaron model predicts a triangular lattice of effective spins, in which each spin in the  $(a, b)$  plane is coupled with its four neighbors by FM exchange

constants, and with the other two by AF ones (Fig. 1). The two-dimensional (2D) lattice is therefore frustrated, and we show below that either collinear AF or spiral ground states are possible. The model has already been analyzed for equal FM exchange couplings  $J_{F1} = J_{F2}$  [6]. We investigate its phase diagram varying  $J_{F2}$  and  $J_A$  ( $J_{F1}$  is taken as an energy unit). The classical magnetic energy per site is given by:

$$E = -J_{F1} \cos \theta_1 - J_{F2} \cos \theta_2 + J_A \cos(\theta_1 + \theta_2), \quad (3)$$

where  $\theta_1$  and  $\theta_2$  are the angles between pairs of spins interacting by  $J_{F1}$  and  $J_{F2}$  exchange, respectively. If  $J_{F2} = J_{F1} = 1$  one finds that for  $J_A < 0.5$  the ground state is FM (in plane), and for  $J_A > 0.5$  it changes to an incommensurate spiral [6]. The angle between spins along  $y$  axis,  $\theta_A = \theta_1 + \theta_2$ , increases and reaches  $120^\circ$  for  $J_A = J_{F1}$ . If  $J_A \rightarrow \infty$  the orthogonal state described by Efremov et al. [7] is found, with AF ordered chains along  $y$ , and with angles  $\theta_1 = \theta_2 = 90^\circ$  between spins along the FM bonds. For small values of  $J_{F2}$  one finds a region of stability of the zigzag  $CE$ -phase ( $\theta_1 = 0, \theta_2 = 180^\circ$ ). The interaction  $J_{F2}$  frustrates this

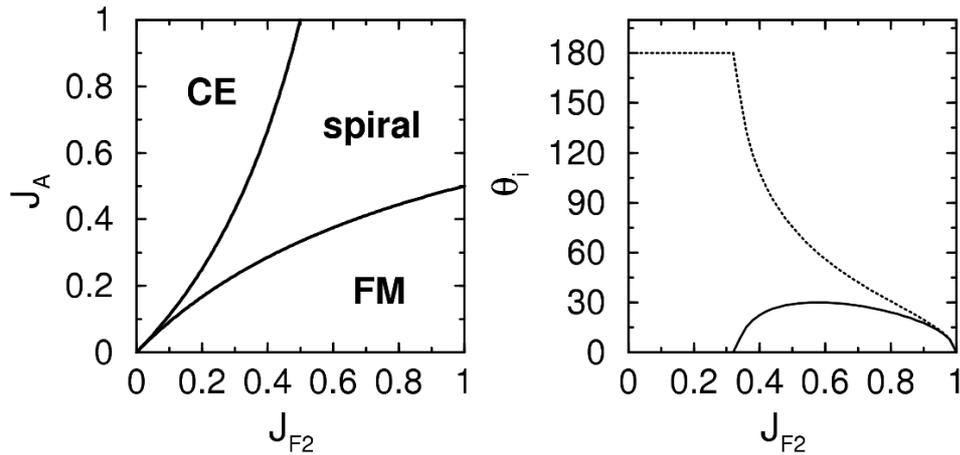


Fig. 3. The classical phase diagram of the polaron model (left), and angles between spins interacting by  $J_{F1}$  ( $\theta_1$ , solid) and  $J_{F2}$  ( $\theta_2$ , dotted) for  $J_A = 0.5$  (right).  $J_{F1} = 1$  everywhere.

magnetic structure, and a crossover to a spiral phase is found at larger values of  $J_{F2}$  (Fig. 3). Spin spirals have the lowest energy for a broad range of parameters. For  $J_A > 1$  new classical phases with nontrivial spin order can be found, and the ground state approaches the  $CE$ -phase order for all  $J_{F2} < 1$ .

The antiferromagnetic coupling  $J_{Az}$  between polarons along  $z$  axis may be added to the model without changing the overall picture (spin order is always AF between planes). Calculating the spin waves dispersion relations we introduced two boson operators due to two inequivalent sites in the lattice. The dispersion

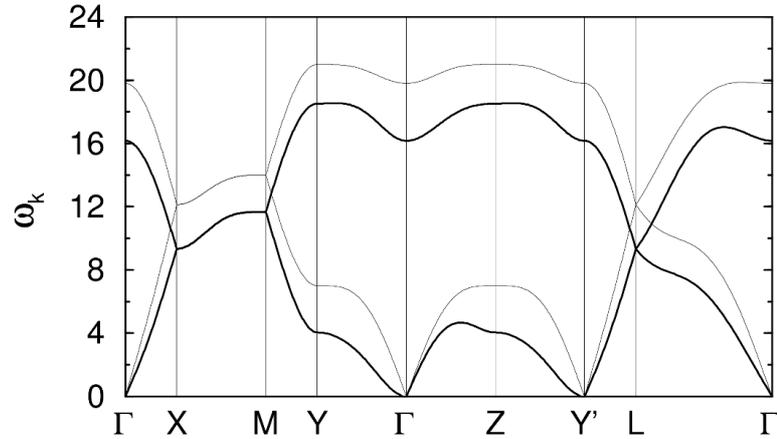


Fig. 4. Spin wave dispersion for the  $CE$ -phase in the effective polaron model. High symmetry points:  $\Gamma = (0, 0, 0)$ ,  $X = (\sqrt{3}\pi/3, 0, 0)$ ,  $M = (\sqrt{3}\pi/3, \pi, 0)$ ,  $Y = (0, \pi, 0)$ ,  $Z = (0, 0, \pi)$ ,  $Y' = (0, \pi, \pi)$ , and  $L = (\sqrt{3}\pi/3, \pi, \pi)$  (within this approach we assume equal distances between polarons, and treat them as a unit). Parameters:  $J_{F1} = 1$ ,  $J_A = 0.5$ ,  $J_{Az} = 0.5$ ,  $S = 7/2$  (both sets);  $J_{F2} = 0$  (dotted),  $J_{F2} = 0.33$  (solid).

relations are shown for the  $CE$ -phase order: without frustration ( $J_{F2} = 0$ ) and near the limit of stability of the collinear order for  $J_A = 0.5$  (Fig. 4). The acoustic mode softens along  $\Gamma - Y$  direction when the spiral phase is approached.

### 3. Conclusions

We analyzed the spin excitations for different spin models of the  $CE$ -phase: the models of interacting effective spins on Mn ions and of the Zener polarons. The last approach leads to frustrated triangular lattice with variety of magnetic ground states, including the  $CE$ -phase. The magnetic order in the ground state of half-doped manganites could be better understood by measuring spin wave dispersion.

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### References

- [1] S. Yunoki, T. Hotta, E. Dagotto, *Phys. Rev. Lett.* **84**, 3714 (2000); O. Sikora, A.M. Oleś, *Phys. Status Solidi B* **236**, 380 (2003); J. Bała, P. Horsch, F. Mack, *Phys. Rev. B* **69**, 094415 (2004).

- [2] A. Daoud-Aladine, J. Rodríguez-Carvajal, L. Pinsard-Gaudart, M.T. Fernández-Díaz, A. Revcolevschi, *Phys. Rev. Lett.* **89**, 097205 (2002).
- [3] G. Zeng, C.H. Patterson, *Phys. Rev. B* **67**, 220404 (2003).
- [4] C.I. Ventura, B. Alascio, *Phys. Rev. B* **68**, 020404 (2003).
- [5] L.R. Walker *Spin Waves and Other Magnetic Modes in Magnetism*, Academic Press, London 1963.
- [6] N.P. Konstantinidis, C.H. Patterson, *Phys. Rev. B* **70**, 064407 (2004).
- [7] D.V. Efremov, J. van den Brink, D.I. Khomskii, cond-mat/0306651 (2003).