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Orbital Order and Electronic Correlations in a Planar Model for Manganites

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Understanding orbital ordered (OO) Mott insulating states lies at the heart of a consistent resolution of the colossal magneto-resistance (CMR) observed in manganites, where its melting induces a low-T insulator-metal transition upon hole doping for $0.25 \leq x \leq 0.45$. Motivated thereby, we study the OO states in a planar model for bilayer manganites using dynamical-mean-field-theory (DMFT) and finite-size diagonalisation methods. We derive the OO ground states in manganites, for $x = 0, \frac{1}{2}, \frac{2}{3}, \frac{3}{4}$ in agreement with observations, including the charge-orbital-magnetic ordered stripe phases for $x > \frac{1}{2}$. These OO states are shown to be associated with an alloy ordering of the $d_{3x^2-r^2}/d_{3y^2-r^2}$ orbitals on each Mn^{3+} site.

I. INTRODUCTION

Colossal magnetoresistance (CMR) materials have received much attention [1–3], due to their extreme sensitivity to minute perturbations [4]. The parent (cubic perovskite) materials are Mott-Hubbard insulators with G-type (anti-ferro, AF) orbital order of $d_{3x^2-r^2}/d_{3y^2-r^2}$ orbitals and A-type AF spin order [4]. Upon hole doping, x (divalent ion substitution) in $La_{1-x}Ca_xMnO_3$, for example, they evolve through ferromagnetic, orbital ordered (OO) Mott insulators with unusual properties [5], to a ferromagnetic metal (FM) at low-T. A transition to a paramagnetic insulator (PI), dependent upon cation-dopant type, is seen for $T > T_c$ [4]. A small magnetic field suppresses this insulator-metal (I-M) transition, leading to CMR. These phenomena are also seen in bilayer manganites. Further, more strange OO states are found in overdoped (with Ca) manganites. The half-doped manganites show a charge, orbital and AF order that is very sensitive to small perturbations [6, 7] ($H_{ext} = 5-7\ T$ gives a ferromagnetic (F) metal with no CO/OO). The overdoped manganites with $x = \frac{1}{2}, \frac{2}{3}, \frac{3}{4}, \frac{4}{5}$ show extremely stable pairs of $Mn^{3+}O_6$ Jahn-Teller distorted stripes having periods between $2-5a$ (a=unit cell length); for other values of x, a mixture of the two adjacent commensurate configurations is found [8]. For $x = 1$, $CaMnO_3$ is again an AF ($S = \frac{3}{2}, t_{2g}$) Mott insulator. Finally, the correlated nature of manganites is shown by dynamical spectral weight transfer (SWT) over large energy scales $O(4.0\ eV)$ in various [9–11] studies as a function of doping (x), temperature (T) and external magnetic fields (B_{ext}) – this can only result from strong electronic correlations. The importance of the Jahn-Teller (JT) [12, 13] coupling is evidenced by the large isotope effects [14] and by I-M transitions driven by $O^{18} \rightarrow O^{16}$ isotope substitution [15] (see, however, Ref. [16], where the JT coupling is argued to be much weaker than in [14, 15]). Thus, understanding CMR is inextricably linked to understanding how these strongly coupled orbital-spin-charge correlations are modified by small perturbations as a function of x. A unified description of these unusual observations in one picture is a formidable challenge for theory.

The CMR problem has been extensively tackled in literature [17–19] using a variety of numerical and analytic methods, for double exchange (DE) models, with/without Jahn-Teller phonons, as well as with strong multi-orbital (MO) Coulomb interactions with static/dynamic JT phonons [18, 19]. For OO states, the full MO Hubbard model has been studied by mapping it to a Kugel-Khomskii (KK) model [20]. However, a controlled treatment (semiclassical analyses [21–23] indicates an order-by-disorder mechanism) is hard: even the type of order is unclear there, and the results sensitively depend on the approximations used [24].

II. MODEL AND RESULTS

Here, we take the first step to study the OO, Mott insulating phases observed in CMR manganites within a $2D$, MO Hubbard model incorporating the above-mentioned strongly coupled correlations. Our conclusions apply, with small additional modifications (to be treated separately) to bilayer manganites. We show that a $2D$ model suffices to capture the correct OO states observed as a function of doping, x, and leave the full $3D$ problem for a separate work. Going beyond previous studies [18, 19, 24], we show how incorporation of the realistic structure of a single MnO_4 layer explicitly in the one-electron hopping integrals introduces new, unanticipated features, making a qualitative difference to the physical results for all x. Further, we show how the strange stripe-ordered phases in the global phase diagram are naturally rationalised from our effective model.

We start with a model that explicitly includes orbital degeneracy of the e_g orbitals in manganites [2, 3],

$$\begin{aligned} H = & -\sum_{<ij>a,b} t_{ij}^{ab}(a_{i\sigma}^{\dagger} b_{j\sigma} + h.c) \\ & + U \sum_{i,\beta=a,b} n_{i\beta\uparrow} n_{i\beta\downarrow} + U' \sum_{i\sigma\sigma'} n_{ia\sigma} n_{ib\sigma'} \\ & - J_H \sum_{i\sigma\sigma'} \mathbf{S_i^c} \cdot \sigma_{\mathbf{i}} (a_{i\sigma}^{\dagger} a_{i\sigma'} + b_{i\sigma}^{\dagger} b_{i\sigma'}) + H_{JT}\ , \end{aligned} \quad (1)$$

where $< i, j >$ denote first neighbors sites, $a_{i\sigma}$ and $b_{i\sigma}$ are fermion annihilation operators in the doubly degenerate e_g orbitals, t_{ij}^{ab} $(a, b = d_{3x^2-r^2}, d_{3y^2-r^2})$ is a 2×2 matrix in orbital

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space incorporating realistic features of the basic $Mn-O$ perovskite structure [1, 3] and $\mathbf{S_i^c}$ is the core-spin due to localized t_{2g} electrons. U, U' are the on-site, intra- and inter-orbital Hubbard interactions, and J_H is the Hund's rule coupling giving rise to the FM state as in the usual DE model. Polaronic effects are described by H_{JT} (see below).

At strong coupling, setting $U, J_H >> t$ in Eq. (1) we find, in absence of U' and J_{JT}, the following effective Hamiltonian

$$H_0 = -\sum_{ij,a,b,\mu} t_\mu^{ab} \gamma_{ij}(\mathbf{S})(a_i^\dagger b_j + h.c). \qquad (2)$$

Here, $\mu = xy$, $t_x^{ab} = \frac{t}{4}[3,\sqrt{3},\sqrt{3},1]$ and $t_y^{ab} = \frac{t}{4}[3,-\sqrt{3},-\sqrt{3},1]$ define the one-electron hopping matrix for a single manganite layer and $\gamma_{ij}(\mathbf{S})$ is the usual DE projection factor [4]. One is effectively dealing with spinless fermions, but now with an orbital index. Clearly, this model ($U'=0$) cannot access the interplay between magnetism and OO in manganites. We now turn on U'. With U' and the JT coupling terms, H becomes

$$H_{eff} = H_0 + U' \sum_{i,a\neq b} n_{ia} n_{ib} + H_{JT} \, . \qquad (3)$$

Defining the operators $c_{i\alpha\uparrow} = (a_i + (-1)^\alpha \sqrt{3} b_i)/\sqrt{2}$, $c_{i\alpha\downarrow} = ((-1)^\alpha \sqrt{3} a_i - b_i)/\sqrt{2}$ with $(-1)^\alpha \equiv +1$ $(\alpha || x)$ and $\equiv -1$ $(\alpha || y)$, the effective Hamiltonian, Eq. (3), yields a Falicov-Kimball model (FKM) [25, 26] where only the $c_{i\alpha\uparrow}$ hop; the $c_{i\alpha\downarrow}$ are strictly immobile as long as no JT distortions are included. (Notice that within this definition the $c_{i\alpha\sigma}$ transform exactly like $d_{3x^2-r^2}(\uparrow), d_{3y^2-r^2}(\downarrow)$). Thus,

$$\begin{aligned} H_{eff} &= -\sum_{<ij>,\alpha} t\gamma_{ij}(\mathbf{S})(c_{i\alpha\uparrow}^\dagger c_{j\alpha\uparrow} + h.c) \\ &\quad + U' \sum_{i,\alpha} n_{i\alpha\uparrow} n_{i\alpha\downarrow} + H_{JT} \\ &\equiv H_{FKM} + H_{JT} \, , \end{aligned} \qquad (4)$$

reflecting the correlation between the magnetic and orbital degrees of freedom described above.

In orbital space, the JT coupling corresponds to addition of external fields [27, 28],

$$H_{JT} = Q_2 \sum_i (n_{ia} - n_{ib}) + Q_3 \sum_i (a_i^\dagger b_i + h.c). \qquad (5)$$

In the rotated basis, this is,

$$H_{JT} = Q_{++} \sum_{i,\alpha} (n_{i\alpha\uparrow} - n_{i\alpha\downarrow}) + Q_{+-} \sum_{i,\alpha} (c_{i\alpha\uparrow}^\dagger c_{i\alpha\downarrow} + h.c) \, , \qquad (6)$$

where $Q_{++} = ((-1)^\alpha \sqrt{3} Q_2 - Q_3)/2$ and $Q_{+-} = (Q_2 + (-1)^\alpha \sqrt{3} Q_3)/2$ are staggered JT distortions which follow the orbital (electronic) variables. So $H_{eff} = H_{FKM} + H_{JT}$ is a FKM with a local, staggered hybridisation between the $c_{\alpha\uparrow}, c_{\alpha\downarrow}$ at each site. Inclusion of finite phonon frequency $(M\Omega^2(Q_2^2 + Q_3^2)/2)$ and intersite phonon coupling terms is required in a full analysis: we have not done this here.

For a half-filled band of spinless fermions, the (numerical) exact solution of H_{eff} in $D=2$ implies an anti-ferro orbital order (AF-OO) of $d_{3x^2-r^2}, d_{3y^2-r^2}$, exactly as required [26]. Such a FKM has been employed earlier [18, 19, 24] for mangnanites, but $c_{\alpha\uparrow} = d_{x^2-y^2}, c_{\alpha\downarrow} = d_{3z^2-r^2}$ there. This would lead to an AF-OO of $d_{x^2-y^2}/d_{3z^2-r^2}$, at variance with observations. Here, such a FKM follows from the realistic hopping structure. Moreover, the AF-OO (Mott insulating, see below) state is driven by large U', in contrast with band-based scenarios. We note that Yamasaki *et al.* [29] have derived an AF-OO Mott insulator for cubic $LaMnO_3$ (with $x=0$) using LDA+DMFT. Our work is thus complementary to theirs for $x=0$, but goes much further, permitting us to study the exotic OO states for $x \geq \frac{1}{2}$ as well (see below). Moreover, given our effective FK mapping [30], the OO state(s) are readily understood in terms of an alloy ordering of $d_{3x^2-r^2}, d_{3y^2-r^2}$ orbitals at *each Mn* site.

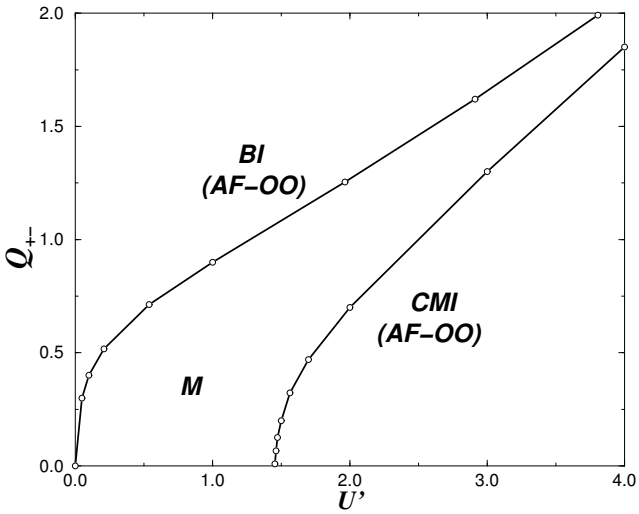

Figura 1: Phase diagram for the transformed multi-orbital model with $Q_{++}=0$ at half-filling. The charge Mott (CMI) and the band (BI) insulators, both with anti-ferro-orbital order (AF-OO) are separated by an incoherent, pseudogapped metal (M) phase.

We now study $H_{eff} = H_{FKM} + H_{JT}$ in the $D=\infty$ [31]. As shown earlier [26], the dynamical-mean-field-theory (DMFT) works surprisingly well for the $2D$ FKM. The FKM with/without Q_{+-} has an almost exact solution in the large lattice dimensional limit, $D=\infty$ [32]. The formalism is essentially the same as that used previously, which is based on a perturbative treatment of the hopping and the transverse field around the atomic limit, and gives very good agreement with Quantum Monte Carlo (QMC) results for the same model [33]. Keeping U'/t fixed and large, phase transitions from the Mott insulator with AF-OO to correlated (incoherent) metal with no OO, to a correlation-assisted band insulator, again with AF-OO, occur: this is indeed borne out in the $D=\infty$ solution, as shown in Fig. 1. Given that U' is much larger than $Q_{++,+-}$ in H above [18, 34], we conclude that manganites fall into the CMI class with AF-OO, and that the JT terms lead to additional stabilization of both. Finally, DMFT gives the full, correlated spectral functions of the model for arbitrary para-

meter values and band-fillings, at a very modest numerical cost. This allows us to study the filling driven Mott transition from an AF-OO Mott insulator to an incoherent metal (see below).

The relevant DMFT equations were derived earlier [32], so we do not repeat them here. Since the JT terms are staggered, but bilinear in the e_g basis, they are easily incorporated into the earlier DMFT structure. The Green function is now a (2×2) matrix in orbital space. The staggered, JT "external field" terms imply an averaging over their orientations, which is carried out within the DMFT equations to yield the density of states (DOS). We choose $U' = 2.6$ eV, $Q_{++} = 0.3$ eV, $Q_{+-} = 0.4$ eV as model parameters [34] along with a non-interacting DOS for the $2D$ square lattice with bandwidth, $W = 2.0$ eV and variable band-filling, $n = (1-x)$, in the DMFT solution. For $n = 1$, (see Fig. 2) we obtain an AFOO Mott insulator. This is obtained from the computed value of $D_{1\alpha} = (-1)^{\alpha}\langle(c^{\dagger}_{i\alpha\uparrow}c_{i\alpha\downarrow} + h.c)\rangle = C(\frac{U'}{W}, Q_{++,+-}) = 0.07$ and $D_{2\alpha} = (-1)^{\alpha}\langle(n_{i\alpha\uparrow} - n_{i\alpha\downarrow})\rangle = C'(\frac{U'}{W}, Q_{++,+-}) = 0.05$ (not shown), obtained directly from

$$D_{2\alpha} = -\frac{1}{\pi}\int \sigma Im G_{\alpha\sigma}(\omega)d\omega \qquad (7)$$

and

$$D_{1\alpha} = -\frac{1}{\pi}\int Im G_{\alpha\uparrow\downarrow}(\omega)d\omega \qquad (8)$$

from the DMFT equations. Away from $n = 1$, the DMFT equations have to be supplemented with the Friedel-Luttinger sum rule, $\langle n\rangle = -\frac{1}{\pi}\int_{-\infty}^{E_F}\sum_{\alpha,\sigma} Im G_{\alpha\sigma}(\omega)d\omega$. This is computed self-consistently within the DMFT.

For $\langle n\rangle = 0.9,\ 0.8$, we obtain an *incoherent*, pseudogapped, metallic state (see Fig. 2) with a sharp reduction of local anti-ferro orbital (AFO) correlations ($D_{1\alpha} = 0.009$). Thus, appearance of the doping-driven (FM) metallic state is intimately linked to the melting of local AF orbital correlations of the Mott insulator with x. The non-Fermi liquid (non-FL) character of the FM contrasts with what is expected in the FKM with uniform hybridisation ($V = Q_{+-}$ in the usual FKM with hybridization), where a correlated FL metal is obtained whenever V is relevant [35]. In our model, the staggered "fields" $Q_{++,+-}$ produce a low-energy pseudogap, suppressing FL coherence. Chemical disorder will further reinforce incoherence [18]. Given the d-wave character of the staggered JT terms (note that both $Q_{++,+-}$ have components that change sign under a $\pi/2$ rotation in xy plane), as well as the (more important) fact that d-wave ground states are obtained near half-filling in a Hubbard-like (FKM) model [36], we predict that this incoherent FM-metal phase will exhibit a d-wave pseudogap.

In contrast to earlier FKM work [18, 19, 24], however, the ordered, insulating phases in un(doped) manganites arise naturally from our model. The checkerboard order of $d_{3x^2-r^2}, d_{3y^2-r^2}$ corresponds to an AF-OO insulator. The exotic bi-stripe states too are naturally predicted from the analysis of our FKM. In the insulating phases, the "hybridisation"(Q_{+-}) is irrelevant, and the resulting FKM ri-

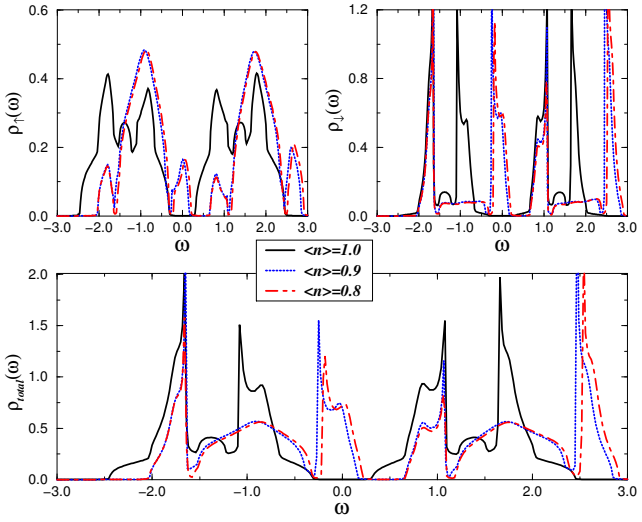

Figura 2: Partial orbital-resolved (top panels) and the total many-body DOS for the AF-OO phase of H_{eff} (see text) within DMFT for $U' = 2.6$ eV, and *staggered* $Q_{++} = 0.3$ eV, $Q_{+-} = 0.4$ eV, for various band-fillings. Off-diagonal components of the spectral fuction are not shown. For $\langle n\rangle = 0.9, 0.8$, the low-energy pseudogap at $\omega = 0$ is clearly seen in the DOS.

gorously undergoes phase separation into hole-rich (orbital disordered) and hole-poor (orbitally ordered) phases, as shown by Freericks *et al.* [30] by minimizing the total energy for various x. We have repeated their analysis for various $x \geq 0.5$. For $x = \frac{1}{2}, \frac{2}{3}, \frac{3}{4}, \frac{4}{5}$, we obtain stripe phases with periods $2, 3, 4, 5$, as observed by Mori *et al.* [8] using electron diffraction. In Fig. 3, we show only the OO ground states for $x = \frac{1}{2}, \frac{2}{3}$; these correspond to those observed in manganites for these hole dopings.

Given that $Mn^{3+,4+}$ correspond to one/zero e_g electron on each Mn site, the $2D$ model automatically has charge-order (CO) of the correct types for these values of x. Also, the stripe OO of pairs of $Mn^{3+}O_6$ (distorted) octahedra automatically corresponds to a bi-stripe CO of e_g electrons with the periodicity determined by x [2, 3]. Given the bi-stripe OO states, Goodenough-Kanamori-Anderson rules directly imply that intersite interactions between the "core" t_{2g} spins ($S = 3/2$) will lead to AF-coupled ladders (Mn^{3+}) separated by strips of JT-undistorted (Mn^{4+}) regions. Given suppression of e_g hopping in an AF background, these stripe states will be insulators, as observed [1–3]. These states will be further stabilised upon inclusion of JT terms and longer range elastic interactions.

This fully corresponds to observations in bilayer manganites for $x > 0.5$ [2, 3]. Thus, stripe states in overdoped CMR result from an alloy ordering of a binary alloy of $Mn^{3+}(S = 2, d^4)$ and $Mn^{4+}(S = \frac{3}{2}, d^3)$ orbitals with $d_{3x^2-r^2}, d_{3y^2-r^2}$ symmetry. Phase separation/stripe phases have long been studied using the FKM (binary alloy disorder model) in alloy physics [37]. Here, we show how these phenomena in manganites arise from strong, MO electronic correlations, which are now

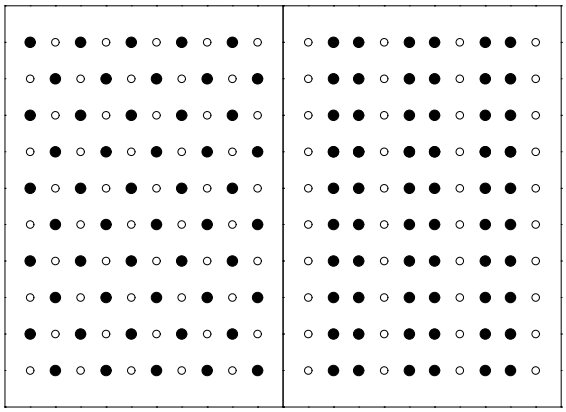

Figura 3: Two different charge-orbital ordered (COO) ground states of the effective Falicov-Kimball model for $x = \frac{1}{2}$ (left) and $x = \frac{2}{3}$ (right). These exactly correspond to the COO states observed in manganites for these x values [2, 3, 8].

representable as a binary alloy model. Since OO states spontaneously break discrete, Ising symmetries of H (Eq. (1)), the link to alloy ordering (described within an Ising model framework [37]) is readily apparent. Finally, it is worth noting that OO phases in a $3D$ model were derived within a static Hartree-Fock approximation [38]. In future, we shall make contact with these results.

III. CONCLUSION

To conclude, we have shown how consideration of the actual multi-orbital structure of the hopping matrix in the e_g sector within a multi-orbital correlated model results in an understanding of the various orbital-ordered (OO) insulating phases observed in CMR manganites, especially in bilayer cases, as a function of x. These are now understood simply as an alloy ordering of $d_{3x^2-r^2}, d_{3y^2-r^2}$ orbitals, driven predominantly by the inter-orbital correlations (U'). Our study shows that OO in overdoped ($x > 0.5$) manganites need not imply very strong JT coupling, in agreement with [16]: by itself, U' leads directly to such phases as a function of x. A moderate JT distortion will further stabilise these ordered phases. Within multi-orbital DMFT, we have shown how an AFOO/F Mott insulator turns into a correlated, incoherent, ferromagnetic bad metal upon hole doping. This goes hand-in-hand with a drop in local AFO correlations. These results are consistent with indications from a host of experiments probing various phases of doped bilayer manganites [39]. Interestingly, planar nickelates are also modelled by a similar Hamiltonian, and our work also naturally explains the OO/stripe phases observed there [40]. We expect our analysis to be broadly applicable to a variety of transition-metal oxide systems showing a variety of OO/magnetic ground states as function of suitable tuning parameters [41].

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