Onset and melting of local orbital order

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Abstract

The onset and melting of locally staggered charge/orbital correlations is investigated within a two-orbital correlated electron model with inter-orbital and inter-site Coulomb interactions. The CE-type orbital correlation exhibits a sharp onset close to the Curie temperature and rapid thermal melting thereafter, which provides quantitative understanding of the $(\pi/2, \pi/2, 0)$ feature observed in neutron scattering experiments on $La_{0.7}(Ca_{x}Sr_{1-x})_{0.3}MnO_{3}$ single crystals. In the zig-zag AF state, the CE-type orbital correlations are found to be even more readily stabilized, but only within a narrow doping regime around $x = 0.5$.

(Some figures may appear in colour only in the online journal)

1. Introduction

The orbital degree of freedom of the electron has attracted considerable attention in recent years due to the rich variety of electronic, magnetic and transport properties exhibited by orbitally degenerate systems such as the ferromagnetic manganites, which have highlighted the interplay between spin and orbital degrees of freedom in these correlated electron systems [1, 2]. Orbital fluctuations, correlations and orderings have been observed in Raman spectroscopic studies [3] of orbiton modes in LaMnO$_3$, polarization-contrast-microscopy studies [4] of La$_{0.5}$Sr$_{1.5}$MnO$_4$, magnetic susceptibility and inelastic neutron scattering studies [5] of La$_x$Ru$_2$O$_{4\alpha}$, and resonant inelastic soft x-ray scattering studies [6] of YTiO$_3$ and LaTiO$_3$. A new detection method for orbital structures and ordering based on spectroscopic imaging scanning tunnelling microscopy is of strong current interest [7] in orbitally active metallic systems such as strontium ruthenates and iron pnictide superconductors.

A composite charge–orbital ordering is exhibited by half-doped narrow-bandwidth manganites such as La$_{0.5}$Ca$_{0.5}$MnO$_3$, with nominally Mn$^{3+}$ and Mn$^{4+}$ atoms on alternate lattice sites in a checkerboard pattern, a staggered orbital ordering of the Mn$^{3+}$electron between the two $e_g$ orbitals corresponding to the wavevector $(\pi/2, \pi/2, 0)$ and a CE-type AF ordering of the Mn core spins arranged in ferromagnetic zig-zag chains [8, 9].

Such CE-type orbital correlations persist in the metallic ferromagnetic phase of the colossal magnetoresistive (CMR) manganites, as revealed in neutron and x-ray scattering experiments in ferromagnetic bilayer [10, 11] and pseudo-cubic manganites [12, 13]. Short-range (10–20 Å) charge and orbital correlations associated with the CMR in the orthorhombic paramagnetic phase were observed as diffuse peaks at wavevector $(\pi/2, \pi/2, 0)$ in the same positions as superlattice peaks in the CE-type charge–orbital structure.

Sharp onset of CE-type dynamical charge/orbital correlations near the Curie temperature were also indicated in Raman scattering studies of Sm$_{1-x}$Sr$_x$MnO$_3$ $(x = 0.45)$ single crystals [14]. Systematic enhancement of this feature was observed with bandwidth reduction, along with the enhancement of CMR. CE-type dynamical charge/orbital correlations and accompanying collective and dynamical lattice distortions were suggested as being responsible for the steep metal–insulator transition and CMR near the charge/orbital ordering instability.

Recent neutron scattering studies of La$_{1-x}$$(Ca_{x}Sr_{1-x})_{3}$ MnO$_3$ crystals have also revealed a sharp onset of the $(\pi/2, \pi/2, 0)$ diffuse peak near the Curie temperature [15], which gradually diminishes in intensity and sharpness with decreasing Ca concentration. This behaviour of CE-type orbital correlations is remarkably similar to the resistivity temperature profile of these crystals with varying Ca concentration [16], and the sharp rise in resistivity and the metal–insulator transition observed near the Curie temperature.

CE-type orbital correlations are also important for magnetic couplings and excitations in the ferromagnetic manganites, as investigated recently by including self-energy and vertex corrections within a systematic Goldstone-mode-preserving scheme. A coupling between spin fluctuations and CE-type orbital fluctuations was shown to yield strong intrinsically non-Heisenberg $(1 - \cos q)^2$ magnon self-energy corrections, resulting in no spin stiffness reduction but...
strongly suppressed zone-boundary magnon energies in the Γ–X direction [17, 18]. This spin–orbital coupling effect was shown to quantitatively account for the several zone-boundary anomalies observed in spin-wave excitation measurements on ferromagnetic manganites [19–24, 15].

Although the Jahn–Teller (JT) electron–phonon coupling is considered to be important in manganites, especially in the low and intermediate doping range [25], the relative importance of Coulomb interaction terms has been of considerable interest. While the CE-type phase was found to be stabilized by JT distortion using mean-field [26] and Monte Carlo [27] studies, the role of inter-site and inter-orbital Coulomb interactions has been emphasized in stabilizing various charge and orbital ordered states [28–31]. Inter-orbital Coulomb interaction has even been suggested to be much stronger than the electron–phonon coupling in order to account for the observed insulating behaviour in undoped manganites above the Jahn–Teller transition and the bond length changes below it [32, 33].

In a recent study of undoped manganites using density functional theory, Coulomb interaction was found to play an important role not only in inducing orbital ordering but also in facilitating the JT distortion via enhanced localization, and electron–lattice coupling alone was found insufficient to stabilize orbital ordering [34]. Similarly, at half-doping, the inter-site interaction was found to be equally important in stabilizing the CE-type state [35]. However, these investigations neglected the important inter-orbital interactions.

Generally, Coulombic and Jahn–Teller-phononic approaches for manganites have been shown to be qualitatively similar [36]. Indeed, a mean-field treatment of the Jahn–Teller term [37] yields an electronic exchange field in orbital space proportional to the orbital magnetization \(\langle n_{i\sigma\alpha} - n_{i\sigma\beta}\rangle\), exactly as would be obtained from the inter-orbital interaction term, suggesting that a purely JT or purely Coulombic approach with effective couplings can approximately describe the combined physical effects. Since orbital correlations are driven by both inter-orbital Coulomb interaction as well as Jahn–Teller electron–phonon coupling, local orbital correlations are therefore generally accompanied by local lattice distortion.

While the role of inter-orbital Coulomb interaction has been investigated for the undoped (\(x = 0\)) parent compound LaMnO\(_3\) [32, 33], and at half-doping (\(x = 0.5\)) [36], the thermal onset and melting of different types of local orbital correlations in the full doping range 0 < \(x\) < 0.5 for ferromagnetic manganites has not been studied within correlated electron models. This should be of great interest in view of the importance of CE-type orbital correlations on spin dynamics due to the spin–orbital coupling effect [17, 18], and their observation close to \(T_c\) in neutron, x-ray and Raman scattering experiments, and their likely role in the observed finite-temperature metal–insulator transition and the CMR effect.

In this paper, we will therefore investigate the doping dependence of local orbital correlations and their sensitivity on the inter-orbital (\(V\)) and inter-site (\(V'\)) Coulomb interactions within a two-orbital interacting electron model. Due to band narrowing in the correlated ferromagnet, a strong sensitivity on the ratio \(V'/t\) would result in a sharp onset of local orbital correlations near the Curie temperature. A detailed comparison of the overall shape with observed neutron scattering features can provide a fundamental insight into onset and melting of local orbital correlations in manganites. We will consider staggered (\(\pi, \pi, \pi\)) orbital correlations, CE-type orbital correlations in the ferromagnetic state and CE-type orbital correlations in the zig-zag AF state, which are of interest in different doping regimes of manganites. We will take the same manganite parameters as used in our recent comparison of spin dynamics results with experiments [17, 18], thus providing a unified picture of both spin and charge–orbital correlations. The values of \(V\) and \(V'\) considered in the present work are in agreement with other investigations [29, 31, 35].

We will be concerned here only with local orbital correlations and not long-range orbital order, a proper treatment of which would require investigation of orbital fluctuations (orbitons) and the electron self-energy due to electron–orbiton coupling. We will therefore use the Hartree–Fock (HF) approximation which qualitatively describes the local orbital moment formation associated with the short time scale correlations (compared to the much longer orbital fluctuation timescales). Earlier HF studies of two-orbital interacting electron models have obtained a qualitatively correct phase diagram for electron-doped (\(x \geq 0.5\)) manganites [38].

2. CE-type orbital order in the ferromagnetic state

The undoped parent compound LaMnO\(_3\) has a staggered (\(\pi, \pi, 0\)) orbital structure with antiferro-orbital ordering in the ferromagnetic plane and ferro-orbital ordering in the perpendicular direction. Weakly doped manganites, which are ferromagnetic insulators, also exhibit an orbitally ordered state for \(x \lesssim 0.2\), as inferred from x-ray diffraction and neutron scattering experiments [39, 15]. The sharp suppression of calculated magnon energies and Curie temperature with the onset of staggered (\(\pi\)) orbital correlations at low hole doping is in good agreement with experiments [18]. In the following, we will focus on the CE-type orbital ordering near half-doping.

At half-doping, narrow-bandwidth manganites such as La\(_{0.5}\)Ca\(_{0.5}\)MnO\(_3\) exhibit a CE-type antiferromagnetic order consisting of ferromagnetic zig-zag chains with staggered charge and orbital ordering. Local CE-type orbital correlations also emerge in the ferromagnetic phase near the Curie temperature, coincident with the metal–insulator transition and the CMR effect, as mentioned earlier. While CE-type charge and orbital ordered ferromagnetic phase was studied at half-doping using cooperative phonons with large electron–phonon coupling [40–43], and recently at hole density \(x = 1/4\) using Monte Carlo investigations [44], and at several fractional hole densities using numerical simulations on finite 3d clusters [45], the doping and temperature dependence of CE-type orbital correlations
within an interacting electron model has not been investigated in the ferromagnetic phase.

In order to investigate the onset and melting of combined charge–orbital orderings in the ferromagnetic state, we therefore consider the following two-orbital model corresponding to the two \( e_g \) orbitals \( \mu = \alpha, \beta \) per site:

\[
H = -t \sum_{\langle ij \rangle, \sigma \mu} a_{i\sigma \mu}^\dagger a_{j\sigma \mu} - J \sum_{i\mu} S_{i\mu} \cdot \mathbf{a}_{i\mu} + V \sum_i n_{\alpha i\alpha \beta} + V' \sum_{\langle ij \rangle} n_{i\alpha j\beta}
\]

(1)

including inter-orbital (\( V \)) and inter-site (\( V' \)) Coulomb interactions which will induce local orbital and charge correlations. For simplicity, we consider a staggered charge–orbital ordering as shown in figure 1, corresponding to ordering wavevector \( \pi \) for charge order and \( \pi / 2 \) for orbital order in all directions, with (spin-) electronic densities:

\[
\langle n_{\alpha} \rangle_{\lambda} = (n_{\beta})_{\lambda} = (n + \delta n / 2 + \delta m) / 2
\]

\[
\langle n_{\alpha} \rangle_{\lambda} = (n_{\beta})_{\lambda} = (n + \delta n / 2 - \delta m) / 2
\]

\[
\langle n_{\alpha} \rangle_{B} = (n_{\alpha})_{D} = (n - \delta n / 2) / 2
\]

\[
\langle n_{\beta} \rangle_{B} = (n_{\beta})_{D} = (n - \delta n / 2) / 2
\]

(2)

where \( \delta n \) and \( \delta m \) represent charge and orbital density modulation. The spin-\( \downarrow \) electronic densities vanish at \( T = 0 \) as the spin-\( \downarrow \) bands are shifted above the Fermi energy by the exchange splitting \( 2J \). In a four-sublattice basis \( (\nu = A, B, C, D) \), with the electron field operator \( \Psi_{k\mu} \equiv (a_{k\mu}^\dagger a_{k\mu} a_{k\mu}^C a_{k\mu}^D) \), the HF-level Hamiltonian for spin-\( \uparrow \) electrons:

\[
H^{(0)} = \sum_{k, \mu} \Psi_{k\mu}^\dagger \left[ \begin{array}{cccc} \Delta_{\text{ch}} - \mu \Delta_{\text{orb}} & \delta_k & 0 & \delta_k^* \\
\delta_k^* & -\Delta_{\text{ch}} & \delta_k & 0 \\
0 & \delta_k^* & \Delta_{\text{ch}} + \mu \Delta_{\text{orb}} & \delta_k \\
\delta_k & 0 & \delta_k^* & -\Delta_{\text{ch}} \end{array} \right] \Psi_{k\mu}
\]

(3)

where the effective charge and orbital exchange fields (\( z \) is the lattice coordination number):

\[
\Delta_{\text{ch}} = (V / 2 - zV')\delta n / 2 \quad \Delta_{\text{orb}} = V\delta m / 2
\]

(4)

and the NN hopping term (which mixes AB, BC, CD and DA sublattices):

\[
\delta_k = -t(e^{ikx} + e^{iky} + e^{ikz}).
\]

(5)

Figure 1. CE-type staggered charge/orbital correlations corresponding to ordering wavevectors \( \pi \) and \( \pi / 2 \) for charge and orbital orders, respectively.

The Hamiltonian matrix (3) was numerically diagonalized to obtain the four eigenvalues \( E_{k\lambda} \) corresponding to the four sub-bands \( \lambda \). The four-component eigenvectors \( \phi_{k\lambda} \) yield the electronic amplitude on sublattice \( \nu \). Evaluation of the new staggered charge and orbital order in terms of the electronic densities obtained by summing over occupied states yields the self-consistency conditions:

\[
\delta n = \sum_{k, \lambda} \left| \phi_{k\lambda}^A \right|^2 - \left| \phi_{k\lambda}^C \right|^2
\]

\[
\delta n = \sum_{k, \lambda} \left| \phi_{k\lambda}^B \right|^2 - \left| \phi_{k\lambda}^D \right|^2
\]

(6)

where the Fermi energy \( E_F \) is also determined self-consistently in terms of the average (spin-\( \uparrow \)) electronic density \( n = 1 - x \). The above self-consistent procedure provides an unrestricted HF scheme, with independent determination of charge and orbital order.

Figure 2 shows the behaviour of the self-consistent CE-type orbital order in the ferromagnetic state with hole doping \( x \) for different inter-orbital interaction strength \( V \). The orbital order is optimal around \( x = 0.4 \) and decreases sharply towards half-doping \( (x = 0.5) \). Due to the small overlap between the two lowest-energy sub-bands as shown in figure 3, the lowest sub-band is fully occupied not at \( x = 0.5 \) but at slightly lower \( x \). As the Fermi energy moves down with increasing \( x \) towards 0.5, the maximally orbital states at the top of the lowest sub-band get emptied, leading to a sharp suppression of the CE orbital order. For \( 0.3 < x < 0.35 \), the orbital order rises sharply for a small change in \( V \) from 4 to 4.4, rendering the system extremely sensitive to small changes in bandwidth and temperature.

The strong presence of CE-type orbital correlations found here in the ferromagnetic state near \( x = 0.45 \) is consistent with the observation of anomalous zone-boundary magnon softening [24], which has been ascribed to correlation-induced magnon self-energy correction arising from coupling of spin fluctuations with specifically CE-type orbital correlations [17, 18].

A strong electron–hole asymmetry around half-doping \( (x = 0.5) \) directly follows from the strongly asymmetric band structure and orbital character about the Fermi energy. While
Figure 2. Behaviour of the CE-type orbital order with hole doping $x$ for different inter-orbital interaction strength $V$, showing its strong sensitivity to small changes in $V'/t$. Here $V'/t = 0.73$.

Figure 3. The density of states showing the four sub-bands in the self-consistent CE-type orbitally ordered ferromagnetic state.

Figure 4. The sharp sensitivity of the CE-type orbital order to the inter-site charge interaction strength $V'$. The CE-type orbital order is strongly enhanced for $x < 0.5$ due to added electrons going in the maximally orbital states at the top of the lowest sub-band, it is rapidly suppressed due to further depletion of these states for $x > 0.5$. The CE-type orbital order peaks at $x \lesssim 0.5$ when the lowest sub-band gets completely filled and further electron filling of the opposite sublattice state at the bottom of the second sub-band results in slow decrease of orbital order with decreasing $x$.

Figure 5. The sharp onset and melting of the CE-type orbital order close to the Curie temperature in the correlated ferromagnet. The shape and temperature scale are quantitatively very similar to the observed behaviour of the $(\pi/2, \pi/2, 0)$ intensity in neutron scattering experiments [15].

The CE-type orbital order shown above was obtained using the unrestricted HF scheme of equation (6), extended to finite temperature by summing over all states with appropriate Fermi functions. The hopping reduction was included approximately as $t(T)/t(0) = 1 - (1/3)(1 - \langle S_z \rangle / S)$ in terms of the temperature-dependent magnetization $\langle S_z \rangle$; this yields a reduction from 1 in the ferromagnetic state to 2/3 in the paramagnetic state, which is similar to the hopping reduction by the factor $\langle \cos(\theta_{ij}/2) \rangle$ in the double-exchange model [25]. The corresponding thermal enhancements for the ratios $V/t$ and $V'/t$ considered in our self-consistent analysis were from 3.3–5.0 and 0.5–0.75, respectively.

Bandwidth (hopping) reduction in a correlated ferromagnet follows from electronic self-energy correction due to electron–magnon interaction, which becomes important near $T_c$ due to thermal excitation of local zone-boundary magnons. Due to this band narrowing near the Curie temperature, finite Jahn–Teller distortion and orbital correlations were shown to
be self-consistently generated in a two-orbital FKLM [37]. However, only ferro-orbital correlations were considered.

Figure 7 shows hysteresis in the onset of CE-type orbital order with increasing/decreasing $V/t$. For the same value of $V/t$ (in the range between 4.0 and 4.6), there are two distinct self-consistent states with and without orbital ordering, depending on the history. Bandwidth reduction with temperature will translate this into a hysteresis behaviour with temperature, indicating metastability and coexisting regions with and without local orbital correlations. This behaviour of CE-type orbital correlations should be important in view of recent observations of spin-glass behaviour, phase separation and evidence of metastability in manganites near half-doping [46].

3. CE-type orbital order in zig-zag AF state

Long-range charge and orbital ordering sets in at half-doping in bilayer and pseudo-cubic manganites where equal numbers of nominal Mn$^{3+}$/Mn$^{4+}$ ions form a checkerboard arrangement in a plane with ferromagnetic zig-zag chains coupled antiferromagnetically, which is repeated in the perpendicular direction. Evidence for such ordering comes from superstructure reflections at wavevector $(\pi, \pi, 0)$ and $(\pi/2, \pi/2, 0)$ in diffraction experiments corresponding to charge and orbital order, respectively.

To investigate the onset of local CE-type orbital ordering in such zig-zag AF states in two dimensions, we consider a $4 \times 4$ sublattice basis for the spin-up and spin-down ferromagnetic zig-zag chains. In the HF approximation, the effective single-particle Hamiltonian for spin-$\uparrow$ electrons is extended to

$$H^{(0)} = \sum_{k,\mu} \Psi_{k\mu}^+ \begin{pmatrix} H_{uu} & H_{ud} \\ H_{du} & H_{dd} \end{pmatrix} \Psi_{k\mu}$$  \hspace{1cm} (7)$$

where the intra-chain term for the spin-$\sigma$ ferromagnetic chain ($\sigma = \uparrow/\downarrow$) is given by

$$H_{\sigma\sigma}(k) = \begin{pmatrix} \Delta_{ch} - \mu \Delta_{orb} & \delta_{k_x} & 0 & \delta_{k_y}^* \\ \delta_{k_x}^* & -\Delta_{ch} & \delta_{k_y} & 0 \\ 0 & \delta_{k_y} & \Delta_{ch} + \mu \Delta_{orb} & \delta_{k_x} \\ \delta_{k_y}^* & 0 & \delta_{k_x} & -\Delta_{ch} \end{pmatrix} - \sigma JS1$$  \hspace{1cm} (8)$$

in the four-sublattice basis introduced earlier, and the inter-chain term:

$$H_{ud}(k) = \begin{pmatrix} 0 & \delta_{k_x} & 0 & \delta_{k_y}^* \\ \delta_{k_x}^* & 0 & \delta_{k_y} & 0 \\ 0 & \delta_{k_y}^* & 0 & \delta_{k_x} \\ \delta_{k_y} & 0 & \delta_{k_x}^* & 0 \end{pmatrix}$$  \hspace{1cm} (9)$$

where $\delta_{k_x}$, etc, are the corresponding components of the NN hopping term given in equation (5). The resulting $8 \times 8$ Hermitian matrix was diagonalized to obtain the eigenvalues and eigenvectors, and the charge–orbital ordering was obtained self-consistently as described in section 2.

As the CE-type orbital order involves a four-sublattice structure, figure 8 shows the corresponding four sub-bands in the low-energy part of the $e_g$ electronic DOS. There is a similar structure shifted up by the exchange splitting $2JS$ corresponding to the spin-down ferromagnetic chains. Out of the four sub-bands, the lowest one is fully occupied at $x = 0.5$ (corresponding to quarter-filling in the spin-$\uparrow$ electron sector).

Figure 9 shows that the CE-type orbital order in the zig-zag AF state is stabilized within a narrow doping range around $x = 0.5$. With increasing Hund’s coupling $J$ and Coulomb barrier in the AF state, the dimensionality of the mobile $e_g$ electrons decreases effectively from two to one. Due to this reduced dimensionality and delocalization, the CE-type orbital near half-doping is found to be more readily stabilized even for smaller interaction strength. Similar doping behaviour of the CE-type orbital order was recently obtained using the Jahn–Teller approach [42].
4. Conclusions

A strong sensitivity of local orbital order on doping concentration, interaction strengths and temperature is highlighted in our investigation of onset and melting of local orbital order within a two-orbital model with effective inter-orbital and inter-site Coulomb interactions.

In the ferromagnetic state, the CE-type orbital order was found to be optimal around $x = 0.4$ and to diminish rapidly at $x = 0.5$. In the doping regime $0.3 < x < 0.4$, the orbital order was found to rise sharply at $V' / t = 4$ and $V' / t = 0.75$, rendering the system extremely sensitive to small changes in bandwidth and temperature. A sharp thermal onset and melting of this orbital ordering was found, remarkably similar in shape and temperature scale to neutron scattering observations, when a small temperature-dependent reduction of the hopping term was included within a self-consistent treatment of the local orbital order, with similar values of $t$ and $V$ as used in a recent detailed comparison of calculated spin dynamics for manganites with experiments [18].

The onset of CE-type orbital order was found to exhibit hysteresis behaviour on increasing/decreasing $V' / t$ (temperature), with two distinct self-consistent states with and without orbital ordering. Indicating metastability and coexisting regions with and without local orbital correlations, this hysteresis behaviour should be important in view of recent observations of spin-glass behaviour, phase separation and evidence of metastability in manganites near half-doping [46].

In a two-dimensional CE-type AF state, with alternating zig-zag ferromagnetic chains, the CE-type orbital order was found to be stabilized only within a very narrow doping range around $x = 0.5$. Due to reduced dimensionality and delocalization of mobile $e_g$ electrons in the zig-zag AF state, the CE-type orbital order near half-doping is stabilized for even smaller interaction strengths. This strong sensitivity and its rapid melting away on either electron or hole doping implies that CE-type orbital order would be highly susceptible in a strong magnetic field as well, which is of interest in the context of magnetic-field-induced melting of CE-type orbital order in half-doped manganites [47].

While only local orbital correlations (local moments in pseudo-spin space) associated with short timescale correlations were considered here, including low-energy orbital fluctuations would allow for investigation of long-range orbital ordering features (orbital correlation length, orbital disordering temperature) and reduction of orbital order due to quantum and thermal excitation of orbitons. Also, renormalization of the CE-state electron spectral properties due to electron–orbiton coupling self-energy resulting from multiple orbiton emission/absorption processes would provide insight into correlated lattice polarons.

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