Orbital ordering in undoped manganites via a generalized Peierls instability

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We study the ground state orbital ordering of LaMnO₃, at weak electron-phonon coupling, when the spin state is A-type antiferromagnet. We determine the orbital ordering by extending to our Jahn-Teller system a recently developed Peierls instability framework for the Holstein model. By using two-dimensional dynamic response functions corresponding to a mixed Jahn-Teller mode, we establish that the Q₂ mode determines the orbital order.

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I. INTRODUCTION

Undoped manganites like LaMnO₃ are the parent systems for the colossal magnetoresistive materials. It is well known that orbital ordering occurs around 780 K resulting in a C-type orbital structure with two kinds of orbitals alternating on adjacent sites in the xy plane while like orbitals are stacked in the z direction. As the temperature is further lowered to 140 K, an A-type spin antiferromagnetic order sets in wherein the spins are ferromagnetically aligned in the xy plane with the spin coupling in the z direction being antiferromagnetic. To explain the observed order several studies have been reported. These studies fall into two broad classes based on the dominant cause for the observed order. One class corresponds to electron-electron (Coulombic) interaction, being the main cause while the other class treats the cooperative Jahn-Teller (JT) interaction as the more important one. Lin and Millis have made a quantitative analysis of the effects of both interactions, generally concluding that both pieces of physics are important, but with many subtleties. There is further controversy about the strength of the electron-phonon interaction with extended X-ray absorption fine structure and pulsed neutron diffraction measurements pointing to strong interaction while some electron microscopy measurements have inferred weak coupling in the charged ordered phases. In that regime optical measurements often infer small electronic gaps, and measurements of nonlinear transport have been interpreted as due to sliding motion of a density wave.

Without addressing ab initio the issues of the quantitative strength of the interactions it is worth understanding how in principle a weak coupling theory might possibly work. The notion of JT is a molecular one, and the linear splitting of levels by a local distortion a useful principle only if the induced gap is much larger than the bandwidth (which it is not). Nonetheless, oxides are generally viewed as a template for strong interaction physics, both of the electronic and phononic variety. In this paper, we step back from the complexities of the full many-body theories to point out that the canonical model for LaMnO₃ has a weak-coupling generalized Peierls instability that reproduces qualitatively the ordering observed. One advantage of the simplification introduced by our approach is that we can study effects of adiabaticity that turn out to enter logarithmically in the ratio of electronic bandwidth to phonon frequency.

Our observation follows straightforwardly from assuming A-type antiferromagnetic ordering. On account of strong Hund’s coupling, the transport is restricted to spin polarized electrons in two dimensions only, where furthermore the bands are strongly nested. The proximity to a nesting instability allows us to employ the weak-coupling framework developed earlier and analyze the orbital ordering by using a generalized Peierls instability approach. However, as compared to the one-dimensional Peierls charge density wave (CDW) approach, our higher dimensional orbital density wave (ODW) analysis is more complicated on account of there being two e_g orbitals (with inter-orbital hopping) and two response functions corresponding to the JT Q₂ and Q₃ distortions. The consequences of a nesting instability on the orbital ordering in LaMnO₃ were first discussed by Yarlagadda and Mitra and later qualitatively by Efremov and Khomskii.

In this paper, we study the Peierls instability condition by extending the recently developed reliable condition involving the dynamic susceptibility to a mixed JT mode. We find that Q₂ Jahn-Teller distortion, as observed experimentally, preempts other JT normal mode distortions at all values of adiabaticity and temperature. Furthermore, the condition of instability (i.e., functional dependence of critical coupling on adiabaticity) is qualitatively similar to that of the one-dimensional single-orbital Holstein model. Lastly, we also find that mean-field approximation (in spite of being crude) and static Peierls instability condition (albeit erroneous) indicate that Q₂ mode rather than Q₃ mode determines the orbital order.

II. MODEL HAMILTONIAN

We will now consider manganite systems with two e_g orbitals per site and ignore spin. The Hamiltonian con-
The electron-phonon interaction term is given by

$$H_{\text{e-ph}} = g \omega_0 \sqrt{2M \omega_0} \sum_{\vec{q}, \omega} \left| Q_{\vec{q}, \omega} \right| n_{\vec{q}} n_{-\vec{q}}$$

where $Q_{\vec{q}, \omega}$ is the dominant mode defined as $Q_{\phi} = Q_0 \cos(2\phi) + Q_2 \sin(2\phi)$ where only orbitals $\psi_{xz-y^2} \cos(\phi) + \psi_{yz} \sin(\phi)$ or their orthonormal orbital states $-\psi_{xz-y^2} \sin(\phi) + \psi_{yz} \cos(\phi)$ are occupied. The order parameter corresponding to phonons is given by $\langle f_{\phi \vec{q}} \rangle = |\langle f_{\phi \vec{q}} \rangle|^2$. Using reflection symmetry, we first note that $\chi_{\phi}(\vec{Q}, \omega) = \chi_{\phi}(-\vec{Q}, \omega)$, $n_{\vec{q}} = n_{-\vec{q}}$, $\langle f_{\phi \vec{q}} \rangle = \langle f_{\phi \vec{q}} \rangle$, and $\langle f^{\dagger}_{\phi \vec{q}} \rangle = \langle f^{\dagger}_{\phi \vec{q}} \rangle$. For $g > 0$, free energy minimum occurs at $\Theta = \pi$. Minimizing $F$, with respect to $|\langle f_{\phi \vec{q}} \rangle|$, yields $|\langle f_{\phi \vec{q}} \rangle| = g n_{\vec{q}}$. Thus, we get

$$F = - \frac{1 + 2g^2 \omega_0 \Re \chi_{\phi}(\vec{Q}, \omega)}{\Re \chi_{\phi}(\vec{Q}, \omega)} \left[ 1 + 2g^2 \omega_0 \Re \chi_{\phi}(\vec{Q}, \omega) \right] n_{\vec{q}} n_{-\vec{q}}$$

On defining the effective susceptibility as

$$\chi_{\phi}^{\text{eff}} = \frac{\Re \chi_{\phi}}{1 + 2g^2 \omega_0 \Re \chi_{\phi}}$$

the Peierls instability condition is given by

$$1 + 2g^2 \omega_0 \Re \chi_{\phi}(\vec{Q}, \omega_0) = 0$$

and leads to the divergence of $\chi_{\phi}^{\text{eff}}(\vec{Q}, \omega_0)$. We take $\omega = \omega_0$ in $\chi_{\phi}^{\text{eff}}(\vec{Q}, \omega)$ because $\omega_0$ is the natural frequency for lattice distortion. A better explanation for choosing $\omega = \omega_0$ is given in Appendix A.

We need to determine at what value of $\phi$ one gets the largest value of $\Re \chi_{\phi}(\vec{Q}, \omega_0)$. Then one can determine...
The dynamic susceptibility is given by

\[ \chi_\phi(\vec{q}, \omega) = \sum_n \left\{ \frac{|\langle n|Q\phi(\vec{q})|0\rangle|^2}{\omega - \xi_n + i\eta} - \frac{|\langle 0|Q\phi(\vec{q})|n\rangle|^2}{\omega + \xi_n + i\eta} \right\}, \tag{8} \]

where

\[ \rho Q\phi(\vec{q}) = \sum_k \left[ b^\dagger_{1k+\vec{q}} b^\dagger_{1k} - b^\dagger_{2k+\vec{q}} b^\dagger_{2k} \right] \cos(2\phi) \]

\[ + \left[ b^\dagger_{1k+\vec{q}} b_{2k} + b^\dagger_{2k+\vec{q}} b_{1k} \right] \sin(2\phi). \tag{9} \]

Then, after some algebra, one gets

\[ \text{Re} \chi_\phi(\vec{q},\omega_0) = \sum_{\vec{k},\alpha,\beta} \left[ \frac{\langle \alpha|c_k^\dagger \beta_k^\dagger \rangle - \langle \beta|c_k^\dagger \alpha_k^\dagger \rangle}{\omega_0 - \epsilon_{\alpha_k} + \epsilon_{\beta_k}} \right] \times \]

\[ \cos^2 \left[ \frac{\theta_{\alpha_k+\vec{q}} + \theta_{\beta_k} + (\alpha + \beta)\pi}{2} + 2\phi \right]. \tag{10} \]

where \( \alpha = 1, 2; \beta = 1, 2; (\epsilon_{1k}^1, \epsilon_{2k}^2) = (b_{1k}^\dagger, b_{2k}^\dagger) \cdot \mathbf{M}, \mathbf{M} \) is the diagonalizing matrix for the kinetic matrix \( \mathbf{T} \) with \( \mathbf{M}_{1,1} = \sin(\theta_{k}/2), \mathbf{M}_{2,2} = -\sin(\theta_{k}/2), \text{and} \mathbf{M}_{1,2} = \cos(\theta_{k}/2). \) It is interesting to note that, for symmetric wavevectors \( \vec{q} = (q,q), \) there is no coupling between the density operators corresponding to \( Q_2 \) and \( Q_3 \) modes because the inter-orbital hopping \( T_{1,2} = 0.5\sqrt{3}(\cos p_x - \cos p_y) \) is asymmetric with respect to interchanging of momenta \( p_x \) and \( p_y. \) Thus for \( \vec{q} = (q,q), \) we obtain

\[ \chi_\phi(\vec{q},\omega_0) = \chi_3(\vec{q},\omega_0) \cos^2(2\phi) + \chi_2(\vec{q},\omega_0) \sin^2(2\phi), \tag{11} \]

where \( \chi_2,3 \) correspond to JT modes \( Q_2,3 \)

### A. Static Instability Case

Now, although the static Peierls instability condition

\[ 1 + 2g^2\omega_0\chi_\phi(\vec{Q},0) = 0 \]

erroneously predicts instability even for vanishing values of \( g, \) it can still help identify which normal mode produces the Jahn-Teller instability. We will first present results for the static susceptibilities \( \chi_{2,3}(\vec{Q},0). \) From the plot of \( \chi_{2,3}(\vec{Q},0) \) (shown in Fig. 2) as a function of scaled temperature \( \alpha T \) (with \( \alpha \) being a scaling parameter and hopping term \( cT \) set equal to 1.0 eV) we see that they diverge logarithmically as \( T \to 0 \) with \( \chi_2 \) diverging faster than \( \chi_3. \) At 0 K, both \( \chi_2(Q,0) \) and \( \chi_3(Q,0) \) produce a divergence because of the fact that \( \lambda_{Q+\vec{Q}} = -\lambda_2 \) and that the Fermi energy is zero. Furthermore, the ratio \( \chi_2(Q,0)/\chi_3(Q,0) = 3 \) at 0 K (see Appendix B for details). As can be seen from Fig. 3, we find that \( \chi_{2,3}(\vec{Q},0) \) vary logarithmically with \( k_B T/t \) for \( t/k_B T > 2 \) and thus have the form

\[ \text{Re}[-t\chi_{2,3}(\vec{Q},0)] = m_{2,3} \ln(t/k_B T) + \kappa_{2,3}. \tag{12} \]

We find that \( m_2 \approx 12.6 \) (\( m_3 \approx 4.2 \)) and \( \kappa_2 \approx 18.3 \) (\( \kappa_3 \approx 18.5 \)) with the ratio \( m_2/m_3 \) taking the expected value 3. Thus it appears that \( Q_2 \) mode is likely to dictate the orbital ordering.

### B. Dynamic Instability Case

While both the static Peierls instability condition and the mean-field energy analysis (see Appendix C) depend only on the polaron size parameter \( (g^2\omega_0/t), \) here for the dynamical Peierls instability condition [of Eq. (7)] there are two relevant parameters — namely adiabaticity parameter \( t/\omega_0 \) and electron-phonon coupling \( g. \) We find that for any value of the adiabaticity parameter \( t/\omega_0 \) the maximum value of \( \text{Re} \chi_\phi(\vec{Q},\omega_0) \) occurs at \( \phi = \pi/4 \) which corresponds to \( Q_2 \) mode. In Fig. 3 using Eq. (10), a
variation of $\text{Re} \chi (\vec{Q}, \omega_0)$ (at 0 K) is plotted for a few representative values of $\phi = 0, \pi/12, \pi/6, \pi/4$. The curves for $\text{Re} \chi_{\pi/12}(\vec{Q}, \omega_0)$ and $\text{Re} \chi_{\pi/6}(\vec{Q}, \omega_0)$ (in Fig. 3) verify Eq. (11). Furthermore, we also found numerically that $\text{Re} \chi (\vec{Q}, \omega_0)$ [given by Eq. (10)] is symmetric about $\phi = \pi/4$ – a fact that follows from Eq. (11).

Quite strikingly, all the $\text{Re} \chi (\vec{Q}, \omega_0)$ vary logarithmically with the adiabaticity $t/\omega_0$ for $t/\omega_0 > 2$ and have the form

$$\text{Re} [-t \chi (\vec{Q}, \omega_0)] = m_\phi \ln (t/\omega_0) + \kappa_\phi.$$  \hspace{1cm} (13)

We find that $m_{\pi/4} \approx 12.6$ ($m_0 \approx 4.2$) and $\kappa_{\pi/4} \approx 25.5$ ($\kappa_0 \approx 20.9$). Interestingly, the slopes in Eq. (13) are the same as those in Eq. (12). The ratio of the slopes $m_{\pi/4}/m_0 = 3$ as expected from the fact that $\chi_2(\vec{Q}, 0)/\chi_3(\vec{Q}, 0) = 3$ at 0 K. Furthermore, this logarithmic dependence is quite like that for the Holstein model. Using the dynamic Peierls instability condition, similar to the Holstein model case, we are lead to an instability condition of the form $\omega_0 = a_1 t e^{-a_2 t/g \omega_0}$ where $a_{1,2}$ are constants. We also calculated the critical value of the electron-phonon coupling $g_c$, at which the instability occurs if only $Q_2$ mode or only $Q_3$ mode is excited. We find that the value of $g_c$ increases monotonically with the adiabaticity parameter (similar to the Holstein model) and that, as expected, the $g_c$ value is the smallest for $Q_2$ distortion (as can be seen in Fig. 4) at any value of $t/\omega_0$.

We have also studied the temperature dependence of the dynamical susceptibilities (as shown in Fig. 5) and find that at low temperatures the curves are constant with the extent of the constant region increasing as $t/\omega_0$ decreases. Such a behavior is consistent with the expectation that $\text{Re} \chi (\vec{Q}, \omega_0)$ is constant over the region $k_B T << \omega_0$. Furthermore, at higher temperatures the susceptibilities for various adiabaticities merge. For instance, when $\alpha T$ attains a value of around 300 K, curves for $t/\omega_0 = 100$ and $\infty$ merge (as can be seen from Figs. 2 and 4); and for $\alpha T$ around 2000 K, curves for $t/\omega_0 = 100$ and 5 merge. The high temperature behavior too is understandable because one expects the effect of non-zero value of $\omega_0$ to vanish when $k_B T >> \omega_0$. At the Jahn-Teller orbital ordering temperature of 780 K and for realistic values of both $t$ and $\omega_0$ (i.e., for $0.15 \text{ eV} \leq t \leq 0.38 \text{ eV}$ and for $0.06 \text{ eV} \leq \omega_0 \leq 0.07 \text{ eV}$), range of the critical coupling as obtained from Eq. (17) and Fig. 5 is $0.2 \leq g_c \leq 0.28$. For instance, at $T = 780$ K, $t = 0.2$ eV and hence $\alpha = 5$ in Fig. 5, and $\omega_0 = 0.07$ eV, we get $g_c \approx 0.21$. Lastly, for $k_B T >> \omega_0$ but $k_B T/t < 0.5$, the curves display a logarithmic dependence on $k_B T/t$ which is in tune with the logarithmic dependence on $\omega_0/t$ of the susceptibility of the Holstein model when $\omega_0/t < 0.5$ (see Ref. 1).

IV. CONCLUSIONS

We will now discuss the general features of the orbital-ordering instability and compare it with the Peierls instability in the Holstein model. For the Holstein model, at 0 K, the mean-field approximation gives a gap $\Delta$ of the form

$$\Delta = 8 t e^{-\pi t/g \omega_0}.$$  \hspace{1cm} (14)

In our case as well, we find that the gap is given by

$$\Delta_{2,3} = d^1_{2,3} t e^{-d^2_{2,3} t/g^2 \omega_0},$$  \hspace{1cm} (15)

where $d_{2,3}^1$ and $d_{2,3}^2$ are constants and $\Delta_{2,3} \approx g^2 \omega_0 |c_{2,3}|$ with $c_{2,3}$ being amplitudes of orbital density waves defined in Appendix C. It should however be noted that, when $\Delta/\omega_0 << 1$, mean-field gives erroneous results. For instance, it predicts a gap even when the electron-phonon coupling $g$ is small. Although, mean-field approximation is inaccurate at the transition, it can still help us figure out which of the two JT modes is dominant. As shown in Appendix C, mean-field correctly shows that $Q_2$ mode prevails over $Q_3$ mode.
Next, in the Holstein model \(15\), at 0 K and \(t/\omega_0 > 2\), the actual instability condition is given by

\[
\omega_0 = 8te^{-\pi t/g^2\omega_0}, \quad (16)
\]

For our JT system too, at \(k_BT << \omega_0 (k_BT >> \omega_0)\) and when \(t/\omega_0 > 2 \) (\(t/k_BT > 2\)), the instability is of the form

\[
\omega_0 (\gamma k_BT) = a_1te^{-a_2t/g^2\omega_0}, \quad (17)
\]

and thus, like the Holstein model, has an essential singularity at \(g = 0\). We also note that one cannot get the correct Peierls instability condition by the approximation \(P^2_\phi/(2M) + KQ^2_\phi/2 \approx KQ^2_\phi/2\) for the normal mode distortion even when \(t/\omega_0\) is large. This is because, when \(P^2_\phi/(2M) = 0\), the double commutator for the distortion \(Q_\phi\) becomes zero,

\[
\tilde{Q}_\phi = -\{[Q_\phi, H], H\} = 0, \quad (18)
\]

which implies that phase transition always occurs!

In summary, we observe that owing to the one-dimensional like Fermi surface at zero doping in manganites (as shown in Fig. 1), there are strong similarities of the above mentioned nature between our JT system and the one-dimensional Holstein model. The one-dimensionality of our manganite system is a result of the dimensionality of our manganite system. The dimensionality of the Fermi surface would be lost. How- ever, hopping in the third direction increases with temperature and the situation is different from that men- tioned in Ref. 24 where, since the hopping in the transverse direction decreases with increasing temperature, re-entrant behavior could occur. Lastly, electron-electron interactions can have an effect on the nesting conditions as pointed out by Kugel, Sheychakov, and Khomskii. These authors find that electron-electron interactions lead to the occurrence of nesting at a density of less than an electron per site. However, in this work, Luttinger’s theorem is violated [see Fig. 5(e) in Ref. 27] and implications of that should be investigated for non-Fermi liquid behavior. If, indeed in a full-fledged calculation, beyond the Hubbard I approximation, nesting (with flat Fermi surface) occurs at a lower density, then a corresponding ODW instability condition should be re-analyzed for such a situation.

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APPENDIX A

We shall give a heuristic justification for the use of dynamic susceptibility in the Peierls instability condition

\[
1 + 2g^2\omega_0 \text{Re}_\phi(\tilde{Q}, \omega_0) = 0. \quad (A1)
\]

Let \(Q_\phi\) be the dominant normal mode distortion operator (at wavevector \(\tilde{Q}\)) in the Fourier transformed space. We know that the double time derivative of the operator \(Q_\phi\) is given by

\[
\tilde{Q}_\phi = -\{[Q_\phi, H], H\}. \quad (A2)
\]

Then on taking matrix elements we get

\[
\langle \Phi_1 | \tilde{Q}_\phi | \Phi_0 \rangle = -(E_{\Phi_1} - E_{\Phi_0})^2 \langle \Phi_1 | Q_\phi | \Phi_0 \rangle. \quad (A3)
\]

Before we close, a few general discussions are in order. Above the magnetic transition temperature \(T_N\), where orbital structure does not change much, transport is permitted in the third direction and the Fermi surface for three dimensions should be considered. Then, although the bands are not flat, we still have the nesting condition \(\lambda_1 = -\lambda_2^2\) for \(\tilde{Q} = (\pi, \pi, \pi)\) and hence the static susceptibilities will diverge. However, the experimental ordering wavevector is \((\pi, \pi, 0)\) and not \((\pi, \pi, \pi)\). To get the observed ordering one will have to incorporate additional physics such as octahedral tilting. Next, at non-zero temperatures below \(T_N\), hopping in the third direction is small but non-zero owing to the non-saturation in A-type antiferromagnetic order. Then flatness (one-dimensionality) of the Fermi surface would be lost. However, hopping in the third direction increases with temperature and the situation is different from that mentioned in Ref. 24 where, since the hopping in the transverse direction decreases with increasing temperature, re-entrant behavior could occur. Lastly, electron-electron interactions can have an effect on the nesting conditions as pointed out by Kugel, Sboychakov, and Khomskii. These authors find that electron-electron interactions lead to the occurrence of nesting at a density of less than an electron per site. However, in this work, Luttinger’s theorem is violated [see Fig. 5(e) in Ref. 27] and implications of that should be investigated for non-Fermi liquid behavior. If, indeed in a full-fledged calculation, beyond the Hubbard I approximation, nesting (with flat Fermi surface) occurs at a lower density, then a corresponding ODW instability condition should be re-analyzed for such a situation.
where $\Phi_n$ is an eigenstate with $n$ phonons all of which are in the state $\vec{Q}$. When $\omega_{eff}^2 = (E_{\Phi_1} - E_{\Phi_0})^2 \leq 0$, instability occurs for transition from $|\Phi_0\rangle$ to $|\Phi_1\rangle$ provided that $\langle \Phi_1 | Q_{\vec{Q}} | \Phi_0 \rangle \neq 0$. Now, at weak electron-phonon couplings (i.e., when $g\omega_0/t < 1$)

$$E_{\Phi_1} - E_{\Phi_0} = \omega_0 + Re\Sigma_0(\vec{Q},\omega_0) = \omega_0 + g^2\omega_0^2 Re\chi_0(\vec{Q},\omega_0),$$
(A4)

where $\Sigma_0$ is the self-energy corresponding to mode $Q_{\vec{Q}}$. Thus, when

$$\omega_{eff}^2 = \omega_0^2[1 + 2g^2\omega_0Re\chi_0(\vec{Q},\omega_0)] = 0,$$  
(A5)

CDW instability occurs. The above instability condition is exact up to second-order in perturbation theory. A more detailed and rigorous derivation of the dynamic Peierls instability condition is given in Ref. [1].

**APPENDIX B**

We will show analytically that $\chi_2(\vec{Q},0)/\chi_3(\vec{Q},0) = 3$ at 0 K. Understanding the susceptibilities is complicated because the eigenstates [corresponding to the eigenvalues $\lambda_{1,2}^k$] are a linear combination of the states $\psi_{k,x^2-y^2}$ and $\psi_{k,3z^2-r^2}$ with coefficients that are a function of the wavevector $\vec{k}$. More precisely, the eigenvectors for $\lambda_{1,2}^k$ are given by $(c_{1,k}^+, c_{2,k}^+)$ $= (b_{1,k}^{\dagger}, b_{2,k}^{\dagger}) \cdot M$, where $M$ is the diagonalizing matrix for the kinetic matrix $T$ with $M_{1,1} = \sin(\theta_k/2)$, $M_{2,2} = -\sin(\theta_k/2)$, and $M_{1,2} = \cos(\theta_k/2)$. Now, from the kinetic matrix $T$, we get

$$\cos(\theta_k) = \frac{0.5[\cos p_x + \cos p_y]}{\sqrt{\cos^2 p_x + \cos^2 p_y - \cos p_x \cos p_y}},$$
(B1)

and

$$\sin(\theta_k) = \frac{0.5\sqrt{3}[\cos p_x - \cos p_y]}{\sqrt{\cos^2 p_x + \cos^2 p_y - \cos p_x \cos p_y}}.$$  
(B2)

In the expressions for $\chi_{2,3}(\vec{Q},0)$ given below

$$\chi_2(\vec{Q},0) = \sum_{\vec{k},\alpha,\beta} \left[ \frac{\langle c_{\vec{k}}^{\dagger} c_{\vec{k}}^\alpha \rangle - \langle c_{\vec{k}+Q}^{\dagger} c_{\vec{k}+Q}^\beta \rangle}{\lambda_{1,2}^k - \lambda_{2,1}^{\vec{k}+\vec{Q}}} \right] \times \sin^2 \left[ \frac{\theta_{\vec{k}+\vec{Q}} + \theta_{\vec{k}} + (\alpha + \beta)\pi}{2} \right],$$
(B3)

and

$$\chi_3(\vec{Q},0) = \sum_{\vec{k},\alpha,\beta} \left[ \frac{\langle c_{\vec{k}}^{\dagger} c_{\vec{k}}^\alpha \rangle - \langle c_{\vec{k}+Q}^{\dagger} c_{\vec{k}+Q}^\beta \rangle}{\lambda_{1,2}^k - \lambda_{2,1}^{\vec{k}+\vec{Q}}} \right] \times \cos^2 \left[ \frac{\theta_{\vec{k}+\vec{Q}} + \theta_{\vec{k}} + (\alpha + \beta)\pi}{2} \right].$$
(B4)

because $\lambda_{2,1}^{\vec{k}+\vec{Q}} = -\lambda_{1,2}^k$ and since on the Fermi surface (FS) $\lambda_{1,2}^k = 0$, the following term diverges

$$\left[ \frac{\langle c_{\vec{k}}^{\dagger} c_{\vec{k}}^\alpha \rangle - \langle c_{\vec{k}+Q}^{\dagger} c_{\vec{k}+Q}^\beta \rangle}{\lambda_{1,2}^k - \lambda_{2,1}^{\vec{k}+\vec{Q}}} \right].$$
(B5)

Furthermore, because $\cos(\vec{k}+\vec{Q}) = -\cos(\vec{k})$ and since $\lambda_{1,2}^k = 0$ on the FS, the following term also diverges

$$\left[ \frac{\langle c_{\vec{k}}^{\dagger} c_{\vec{k}}^\alpha \rangle - \langle c_{\vec{k}+Q}^{\dagger} c_{\vec{k}+Q}^\beta \rangle}{\lambda_{2,1}^k - \lambda_{1,2}^{\vec{k}+\vec{Q}}} \right].$$
(B6)

Then

$$\chi_2(\vec{Q},0)/\chi_3(\vec{Q},0) = \frac{\cos^2 \left[ \frac{\theta_{\vec{k}+\vec{Q}} + \theta_{\vec{k}}}{2} \right]}{\sin^2 \left[ \frac{\theta_{\vec{k}+\vec{Q}} + \theta_{\vec{k}}}{2} \right]}_{FS} = \frac{\sin^2(\theta_k)_{FS}}{\cos^2(\theta_k)_{FS}} = 3,$$  
(B7)

where use has been made of the fact that the FS is flat and one-dimensional like and that on the FS either $k_x = \pm \pi/2$ or $k_y = \pm \pi/2$.

**APPENDIX C: MEAN-FIELD CDW ANALYSIS**

Assuming that the total wavefunction of the system is separable into a phononic part and an electronic part, after averaging the Hamiltonian over the phononic coordinates, we get the following effective Hamiltonian (with details given in Ref. [19]):

$$\bar{H} = \sum_\vec{p} B_\vec{p} \cdot T \cdot B_\vec{p}$$

$$-2g^2\omega_0 \sum_j \left[ \langle b_{1,j}^{\dagger} b_{2,j} \rangle \langle b_{1,j}^{\dagger} b_{2,j} + b_{2,j}^{\dagger} b_{1,j} \rangle + \langle b_{1,j} b_{1,j} - b_{2,j} b_{2,j} \rangle \rangle \right] + g^2\omega_0 \sum_j \left[ \langle b_{1,j}^{\dagger} b_{2,j} + b_{2,j}^{\dagger} b_{1,j} \rangle \langle b_{1,j}^{\dagger} b_{2,j} + b_{2,j}^{\dagger} b_{1,j} \rangle + \langle b_{1,j} b_{1,j} - b_{2,j} b_{2,j} \rangle \right]^2,$$  
(C1)

where $< \ldots >$ implies averaging over the relevant coordinates which here are electronic.

Based on the arguments that wavevector $\vec{Q}$ determines the orbital ordering in two-dimensions (as discussed in Sec. III), we compute the ground state energy using mean-field when only either $Q_{2}$ mode or $Q_{3}$ mode gets excited cooperatively in the system. The order parameters are given by $\langle b_{1,j}^{\dagger} b_{2,j} + b_{2,j}^{\dagger} b_{1,j} \rangle = c_2 \cos(\vec{Q} \cdot \vec{R}_j)$ and $\langle b_{1,j} b_{1,j} - b_{2,j} b_{2,j} \rangle = c_3 \cos(\vec{Q} \cdot \vec{R}_j)$ with $-1 \leq c_{2,3} \leq 1$ and $\vec{R}_j$ being the position vector. Here it should be...
pointed out that the order parameter \(\langle b^\dagger_j b_{j'} + b^\dagger_{j'} b_j \rangle\) corresponds to the density difference of electrons in the two orbitals \(\psi_X \equiv (\psi_{x^2-y^2} - \psi_{3z^2-r^2})/\sqrt{2}\) and \(\psi_Y \equiv -(\psi_{x^2-y^2} + \psi_{3z^2-r^2})/\sqrt{2}\) (as described in Ref. 10).

The unit cell needed to compute the ground state energy consists of two adjacent sites with the Brillouin zone being given by \(-\pi \leq (k_x + k_y) \leq \pi\) and \(-\pi \leq (k_x - k_y) \leq \pi\). We diagonalize a 4 \times 4 matrix at each momentum and integrate the lowest two eigenenergies over the Brillouin zone to obtain the ground state energy. The results of our calculations are shown in Fig. 6. From Fig. 6(a) we see that the ground state energy corresponds to the \(Q_2\) mode with the difference in energy between the \(Q_2\) only state and the \(Q_3\) only state peaking at intermediate values of the dimensionless polaronic energy \((g^2\omega_0/t)\). For zero values and infinite values of the polaronic energy both modes yield the same energy because zero value implies no phononic coupling effect while infinite value corresponds to localized polarons. Thus for large values of the polaronic energy, the ground state energy is only slightly smaller than the polaronic energy. Furthermore, from Fig. 6(b) we also see that, as the polaronic energy increases, the values of \(c_2,3\) increase and become unity around \(g^2\omega_0/t \sim 2\) implying that for the \(Q_3\) \((Q_2)\) mode \(\psi_{x^2-y^2}\) \((\psi_X)\) orbital is occupied fully at one site with the \(\psi_{3z^2-r^2}\) \((\psi_Y)\) orbital being fully occupied at the adjacent sites.

\[\text{FIG. 6: (a) Dependence of dimensionless ground state energy per site (}\mathcal{E}/t\text{) on dimensionless polaronic energy (}g^2\omega_0/t\text{) for cooperative }Q_2\text{ and }Q_3\text{ modes; (b) variation of coefficients }c_{2,3}\text{ of ODW order parameters for }Q_2\text{ and }Q_3\text{ distortions as a function of }g^2\omega_0/t\text{.} \]

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