Single bipolaronic Transition in Jahn-Teller Model

Reza Nourafkan

Department of Physics, Sharif University of Technology, P.O.Box: 11155-9161, Tehran, Iran

Massimo Capone

SMC, CNR-INFM and Dipartimento di Fisica, Università Sapienza, P.le Aldo Moro 2, I-00185, Roma, Italy

Nasser Nafari

Institute for Research in Fundamental Sciences (IPM), 19395-5531, Tehran, Iran

We investigate the bipolaronic crossover and the pairing transition for a two-orbital model with Jahn-Teller coupling to a two-fold degenerate phonon mode. The evolution from weak to strong coupling is reminiscent of the behavior of the single-band Holstein model: The polaron crossover in which the electrons and phonons become strongly entangled occurs for a weaker coupling than the binding of bipolarons, which gives rise to a metal-insulator transition. Interestingly, a single bipolaronic transition takes place also when the two bands have significantly different bandwidths, as opposed to the case of repulsive Hubbard-like interactions for which an orbital-selective Mott transition has been reported. This behavior is related to the inter-orbital nature of the Jahn-Teller coupling.

PACS numbers: 71.38.-k, 71.30.+h, 71.38.Ht, 71.10.Fd

I. INTRODUCTION

The interplay between spin, charge and orbital degrees of freedom plays an essential role in the description of the complex phase diagrams characterizing strongly correlated materials. One of the important mechanisms describing the properties of these systems is the electron-phonon (e-ph) interactions. Most of the studies of e-ph interactions are limited to the Holstein model, in which the electronic state forms a non-degenerate band and the phonons couple with the on-site electronic charge. However, many strongly correlated materials involve nearly degenerate $d$ or $f$ orbitals. In this work we move a step towards a more realistic description of the e-ph interaction in these compounds by considering a degenerate electronic manifold coupled with the proper degenerate phononic models. For these narrow-band systems the phonon dynamics can play a role, therefore the Born-Oppenheimer approximation is hardly justified and the decoupling of the electronic orbital states from the vibrational modes is not allowed.

A typical family of compounds involving such a spin-charge-orbital complex are the colossal magnetoresistance (CMR) manganites. While the double-exchange mechanism is the basis of the CMR phenomenon, relating the magnetic behavior to conduction properties, its interplay with the coupling between the degenerate $e_g$ electrons and the Jahn-Teller (JT) distortions of the MnO$_6$ octahedra is crucial to describe the properties of these materials leading to a variety of experimentally observed charge and/or orbital orders.

The Jahn-Teller model is also relevant to superconductivity in alkali-doped $A_x$C$_{60}$ molecular solid, where C$_{60}$ is the fullerene molecule and A stands for alkali cations K, Rb, or Cs. In the alkali-doped $A_x$C$_{60}$, the threefold degenerate $t_{1u}$ molecular level is partly occupied and couples strongly to eight $H_2$ intra-molecular Jahn-Teller phonons. In these systems the JT character of relevant phonons has important consequences in the presence of strong Coulomb repulsion. Indeed the JT-driven interaction between the electrons only involves spin and orbital degrees of freedom which are still free to fluctuate when the charge excitations are frozen by the Coulomb repulsion. This gives rise to a correlation-driven enhancement of phonon-driven superconductivity.

In both cases the interplay between electron-electron and e-ph interactions which may lead to a rich variety of physical phenomena. In this paper we focus on the pure e-ph interaction term in order to highlight the specific properties introduced by orbital degeneracy before considering the explicit role of electronic interactions, in the same spirit of previous investigations of the Holstein model. For the same reason, i.e., capturing the basic aspects of the electron-phonon interaction we do not allow for superconductivity or charge-density-wave ordering, limiting ourselves to the normal state. In the half-filled Holstein model it has indeed been shown that two related but distinct processes occur by increasing the e-ph coupling. The first effect is the well-known polaron formation, i.e., the progressive entanglement between the electronic motion and the lattice degrees of freedom. Polaron formation occurs indeed as a continuous crossover and it can be pinpointed by analyzing the phonon displacement distribution function. The attractive interactions also induce an attraction between the fermions. Even if superconductivity is explicitly inhibited, this gives rise to a binding of fermionic carriers, eventually leading to a pairing transition which for polaronic carriers becomes a bipolaronic metal-insulator transition.

In this paper we discuss how this physics is modified for a two-orbital $e \times E$ Jahn-Teller model. For this model we can also consider some perturbations, like the possibility of different bare widths for the two electronic bands, in analogy with the much discussed possibility of an orbital-selective Mott transition in the case of repulsive Hubbard-like interactions. The paper is organized as follows. After briefly introducing the model and the method of solution in the next section (Sec. II), we begin to investigate the JT model at half-filling.
in Sec. III and discuss how the e-ph coupling affects the electronic properties of the model. In Sec. IV, we then address the question of whether orbital-selective bipolaron transition occur in the system with different band-width. The concluding remarks is given in Sec. V.

II. MODEL AND METHOD

The $e \times E$ Jahn-Teller model consists of two degenerate electron orbitals and two degenerate phonon modes,

$$H = H_t + H_{ph} + H_{JT}$$

$$H_t = - \sum_{\langle ij \rangle \gamma \sigma} t_{ij} c_{i \gamma \sigma}^\dagger c_{j \gamma \sigma} + c_{j \gamma \sigma}^\dagger c_{i \gamma \sigma}$$

$$H_{ph} = \Omega_0 \sum_i (a_i^\dagger a_i + b_i^\dagger b_i)$$

$$H_{JT} = g \sum_{i \sigma} (n_{i1\sigma} - n_{i2\sigma})(a_i^\dagger + a_i) +$$

$$+ (c_{i1\sigma}^\dagger c_{i2\sigma} + c_{i2\sigma}^\dagger c_{i1\sigma})(b_i^\dagger + b_i),$$

where $a_i^\dagger (b_i^\dagger)$ and $a_i (b_i)$, respectively, create and annihilate the dispersionless phonons of type $a (b)$ on site $i$. These are the Jahn-Teller modes with the same symmetry and the same phonon frequency $\Omega_0$. $g$ is the related electron-phonon coupling strength. While phonons of type $a$ couple to the density unbalance between the two orbitals, the phonons of type $b$ are coupled to the hybridization between electronic orbitals that are orthogonal in the absence of these phonons. $c_{i \gamma \sigma}^\dagger$ and $c_{i \gamma \sigma}$ are the creation and annihilation operators for electrons at site $i$ in orbital $\gamma$ ($\gamma = 1, 2$) with spin $\sigma$, $n_{i \gamma \sigma} = c_{i \gamma \sigma}^\dagger c_{i \gamma \sigma}$ and $\langle \langle ij \rangle \rangle$ denotes nearest neighbors. $t_{ij} > 0$ are the nearest-neighbor hopping integrals.

We solve the model by means of dynamical mean field theory (DMFT)\cite{DMFT}. The method maps the lattice model onto a quantum impurity model in which an interacting site is embedded into a non-interacting bath, whose spectral function has to be determined self-consistently in such a way that the impurity Green’s function of the quantum impurity model coincides with the local Green’s function of the lattice model under consideration in order to enforce a quantum dynamical mean-field theory. This requirement leads to a self-consistency condition, which contains the information about the original lattice through the non-interacting density of states. A particularly popular and useful choice is a semi-circular density of states of half-bandwidth $D$, i.e.,

$$N(\omega) = \frac{1}{\pi D^2} \sqrt{D^2 - \omega^2},$$

which corresponds to an infinite coordination Bethe lattice. For this system the self-consistency equation takes the following simple form:

$$\frac{D^2}{4} G(i \omega_n) = \sum_k \frac{V_k^2}{i \omega_n - \epsilon_k},$$

where $G(i \omega_n)$ is the local Green’s function of the system, $\epsilon_k$ and $V_k$ are the energies and the hybridization parameters of the impurity model. To solve the impurity model at zero temperature ($T = 0$) we use the exact diagonalization method\cite{ExactDiag}, which works equally well at any value of the parameter and only involves a discretization of the bath function, which is described in terms of a finite and small set of levels $n_s$ (conventionally $n_s$ includes also the impurity site and the number of bath sites is $n_b = n_s - 1$) in order to limit the Hilbert space to a solvable size.

In principle, an infinite number of vibrons can be excited at each lattice site. In DMFT, however, the phonon degrees of freedom are limited to the impurity site. Since the diagonalization can only be carried out for a finite dimension, a truncation of the local phonon subspace is required. Here we use the basis state

$$|\nu\rangle_{ph}; \quad \nu_{\nu} = \left( \sum_{i=1}^{N_{mode}} n_{i,\nu} \right) \leq N_{ph}$$

leading to $(N_{ph} + N_{mode})!/(N_{ph}!N_{mode}!)$ allowed phonon configurations. Here $n_{i,\nu}$ is the number of $i$-type phonon in the basis state $|\nu\rangle_{ph}$ and $N_{mode} = 2$. Typical values we considered for the bath levels are $n_s = 5 - 6$ (which corresponds to $n_s = 10 - 12$ for a single-band model) and $N_{ph} \sim 30 - 40$. We tested that these numbers provide essentially converged results.

DMFT has already been applied to the study of strongly correlated electron-phonon systems and has emerged as one of the most reliable tools for the analysis of these systems\cite{DMFT1,DMFT2,DMFT3,DMFT4}. Studies on the normal phase of the Holstein model show that for e-ph couplings, the ground state is metallic with Fermi liquid characteristic, while increasing the coupling a first-order metal-insulator transition takes place. This phase displays a gap in the one-electron spectrum, but, in contrast to metal-insulator Mott transition, no hysteresis and no preformed gap is observed for the transition from the metallic to the bipolaronic insulating phase, at least for the relatively small values of phonon frequency that have been used. The pairing transition is accompanied by, but it does not coincide with, the progressive entanglement between electronic and vibronic degrees of freedom, or in other words the polaron crossover. For relatively small phonon frequencies, polarons are formed before the pairing transition occurs. Since the latter takes transition binds two polarons, it can be defined as a bipolaronic transition\cite{Bipolaron1}. One of the aims of this paper is to understand whether and to what extent the symmetry of the e-ph coupling can influence the electronic properties and the dynamic quantities of a given system.

To analyze the metal-insulator transition we study the quasi-particle weights $z_i = 1/[1 - dR \epsilon_i \Sigma_i(\omega)/d\omega|_{\omega=0}]$, $\Sigma_i$ being the self-energy for band $i$. Due to the momentum independence of the self-energy in DMFT, $z$ is proportional to the inverse effective mass ($z \propto m/m^*$), so that a vanishing $z$ implies a divergent effective mass and a metal-insulator transition.

We will also use the double occupancy $d_i = \langle n_i \rangle$, and the phonon displacement probability distribution function (PDF), defined by:

$$P(x) = \langle \psi_0 | x \rangle \langle x | \psi_0 \rangle,$$
as a marker of the polaron crossover. In Eq. (4), $|\psi_0\rangle$ is the ground state vector, and $|x\rangle\langle x|$ is the projection operator on the subspace where the phonon displacement value $\hat{x}$ has a given value $x$. This quantity, therefore, is a measure of the distribution of the local distortions. In the exact diagonalization approach we can evaluate $P(x) = \sum_{n,m} \phi_n(x)\phi_m(x)|\psi_0\rangle\langle n|\psi_0\rangle$, where $|n\rangle$ and $|m\rangle$ are the eigenstates of the harmonic oscillator and $|\phi_n(x)\rangle$ are the corresponding eigenfunctions.

III. RESULTS

We first focus on the isotropic systems consisting of identical bands, $D_1 = D_2 = D$. Furthermore, we confine ourselves to the adiabatic regime with a small ratio of the phonon frequency to the semi-bandwidth ($\Omega_0 = 0.2D_1$) in which the dynamics of phonons is slow as compared to the typical kinetic energy of electrons. The results obtained for this case at half-filling are shown in Figs. (1) and (2). In Fig. (1), panel (a) we report the quasi-particle weight $z_1 \equiv z$ as a function of electron-phonon coupling constant. In a system with e-ph interaction, by increasing the coupling constant, the system enters a strong-coupling polaronic regime in which the presence of an electron is associated with a finite lattice distortion. In the polaronic regime, the system is characterized by low-energy excitations which, although strongly renormalized, are still coherent. Also, the same e-ph coupling can cause a pair of polarons with opposite spins to attract each other forming a bound state in real space, called bipolaron. In DMFT those pairs are unable to move, and the bipolaron formation causes a metal-insulator transition associated with the divergence of the effective mass and the disappearance of a coherent peak at the Fermi level at a critical coupling $g_c$. This process is evident in our DMFT results from the enhancement of the electron effective masses, and the corresponding reduction of the quasi-particle weights ($z_1 = z_2 = z$) as the e-ph coupling constant $g$ increases, which eventually reaches the bipolaronic metal-insulator transition (MIT) shown in panel (a) of Fig. (1). Panel(b) of Fig. (1) shows the double occupancies ($d_1 = d_2$) as a function of $g$. Around the critical coupling, $g_c$, the double occupancies increase suddenly, showing a first order transition to bipolaronic phase.

The inset of the Fig. (1), panel (b), shows orbital correlations between the two orbitals, $\langle n_1n_2 \rangle$. Upon increasing the e-ph coupling, the orbital correlation $\langle n_1n_2 \rangle = 1 - \langle n_1 \rangle \langle n_2 \rangle$ increases until bipolaron transition takes place.

Evolution of quasi-particle weights and the double occupancies do not clearly show the polaron crossover. Therefore, we have computed the phonon displacement probability distribution function to clarify this point. Fig. (2) shows the evolution of the phonon displacement probability distribution function, $P(x)$, as a function of $g$. In the case of isotropic systems, the PDFs for a- and b-modes are the same. Upon increasing the e-ph coupling, a smooth crossover occurs between a unimodal distribution and a bimodal distribution in complete analogy to the single-band Holstein model.

![FIG. 2: Phonon displacement distribution function $P(x)$. The various lines correspond to different values of $g$.](image)

Before entering the bipolaronic phase characterized by $z = 0$, a very narrow region exists in which polarons are formed, nonetheless, the value of quasi-particle weights, although being very small, are different from zero (see two dot-dashed curve of Fig. (2)). The existence of this region shows that, in the process of the metal-insulator bipolaronic transition, polarons are formed for those values of $g$ which are somewhat larger than the polaron crossover. While the qualitative result is similar to what found for the Holstein model, the region in which polarons exist without forming bipolarons turns out to be narrower in the Jahn-Teller model.

We have so far discussed the bipolaronic transition for the two-orbital electronic system with the same bandwidth. However, as a result of the geometric complexity of many transition metal oxides, the degeneracy of the valence bands is frequently lifted, giving rise to a coexisting partially filled narrow- and wide-bands. Actually, the two-band Hubbard model with orbitals of different widths has recently received considerable attention and it has been shown that in certain range of repulsive Coulomb interactions and Hund’s coupling the so-called orbital-selective phase occurs in which the narrow-band is insulating while the wide-band is still metallic.

In the case of a phonon interaction involving the total charge on the two orbitals, as in a generalized Holstein model, the situation would be most likely similar to the case of the repulsive Hubbard model. Indeed a Holstein coupling induces a charge-charge attraction which, in the antiadiabatic limit $\Omega_0 \rightarrow \infty$ reduces to an attractive Hubbard model, which in turn is equivalent to the repulsive one at half-filling. For the JT interaction the question is instead not obvious because the
phonon modes are not coupled to the total charge, and they are associated with the orbital degrees of freedom. In particular, the $b$-mode is directly associated with a hybridization between the two orbitals, an effect which is clearly against an orbital selective behavior. If we integrate out the phonons in our effective action in which the interaction part of (1) is present only on the impurity, we obtain a retarded attraction

$$V^{\text{eff}}(\omega) = g^2 \left[ (n_1 - n_2)^2 + \left( \sum_{\sigma} c_{1\sigma}^\dagger c_{2\sigma}^\dagger + h.c. \right)^2 \right] D(\omega)$$

where $D(\omega) = -2\Omega_0/(\Omega_0^2 - \omega^2)$ is the bare phonon propagator for each mode. In the antiadiabatic limit $\Omega_0 \rightarrow \infty$ we can drop the frequency dependence in the propagator obtaining an unretarded attraction which involves the orbital momentum of the electrons in the two orbitals and coincides with an inverted Hund’s rule term.

![Graph](image-url)

**FIG. 3:** Quasi-particle weights, $z_1$, $z_2$, (left panels) and double occupancies, $d_1$, $d_2$, (right panels) at half-filling as a function of electron-phonon interaction for different bandwidths. Top panels: $D_2/D_1 = 0.5$, bottom panels $D_2/D_1 = 0.2$. The insets of right panels show the orbital correlations, $\langle n_1 n_2 \rangle$ as a function of $g$. In order to address the existence of the selective bipolaronic phase, we considered two different ratios between the bare bandwidths: $D_2/D_1 = 0.5$ and $0.2$. Fig. 3 shows the results for quasi-particle weights and double occupancies of the two bands in the two cases (The top panel is $D_2/D_1 = 0.5$ and the bottom one $D_2/D_1 = 0.2$). The results for the $z^*$'s are apparently similar to those shown in Fig. 1 for identical bandwidths. The two renormalization factors are indeed only slightly different, in sharp contrast with the results for the Hubbard model. Moreover, the difference in renormalization goes in the opposite direction than for repulsive models with pure charge interactions. Here the wider band is (slightly) more renormalized by the interactions, as if the effect of e-ph coupling is to make the two bandwidths closer with respect to their bare values. However, when the system approaches the polaron crossover, the difference between quasi-particle weights reduces, displaying a fast damping dependence on $g$. Therefore we have a single bipolaronic transition, at least within our numerical accuracy in a wide range of $D_2/D_1$ and the orbital-selective effects are extremely limited if compared with charge interactions. This is interesting also because reducing the value of $D_2$, the second band is not in the same adiabatic regime of the broad one, which could imply some different behavior. Nonetheless, the inter-orbital nature of the e-ph adiabatic regime overcomes also this effect to leading to a unique transition. The spectral functions of the two bands (not shown) confirm the uniqueness of the transition. At the bipolaronic transition, where the two $z^*$'s vanish simultaneously, the peak at the Fermi level disappears in both bands.

On the other hand, the growth in the number of doubly occupied sites (right panels in Fig. 3) depends on the band and on the value of the bandwidth ratio. The narrow band rapidly develops a larger number of doubly occupied sites, and the effect is more pronounced the narrower the band is. The critical value of $g$ for the metal insulator transition decreases as the ratio $D_2/D_1$ is reduced. This behavior is somewhat natural since we are keeping $D_1$ fixed and reducing $D_2$. Therefore the overall kinetic energy of the system is reduced, making polaron and bipolaron formation easier (both the polaron crossover and the bipolaron transition are expected to occur because the e-ph interaction overcomes the kinetic energy and its delocalizing effect, at least in the adiabatic regime in which the energetic convenience also implies a sizeable entanglement between electrons and phonons).

The phonon displacement distribution functions are shown at Fig. 4 for $D_2/D_1 = 0.5$. Breaking the symmetry between the two orbitals determines a different behavior of the two phonon modes. With increasing e-ph coupling, the PDF of the $a$ mode becomes bimodal before the bipolaronic transition occurs, while for the same values of the coupling the PDF of the $b$-mode maintains a unimodal shape with a single feature which broadens without showing bimodality all the way to the bipolaronic transition. The two peaks of the bimodal distribution for the $a$ mode turn out to have slightly different height. The width of the region of couplings between the polaron crossover for the $a$ mode and the metal insulator transition is larger than for the case of equal bandwidths.

![Graph](image-url)

**FIG. 4:** Phonon displacement distribution function $P(x)$. The left panel is for $a$ phonon mode which shows bimodality. The right panel is for $b$ phonon mode which does not show bimodality.
IV. CONCLUDING REMARKS

We considered the normal phase of a two-orbital model coupled with a two-fold degenerate phonon manifold via an $e \times E$ Jahn-Teller coupling. Solving the model at half-filling and zero temperature by means of dynamical mean-field theory with exact diagonalization as the “impurity solver”, we investigated the polaron crossover and the bipolaron transition which occur as a function of the coupling constant. In the standard case of identical bands, our results are qualitatively similar to those of a single-band Holstein model. In the adiabatic regime that we investigate, a polaron crossover takes place for weaker coupling with respect to a metal-insulator transition associated to the formation of bipolarons. The intermediate region between the two effect is found to be smaller than in the single-band case.

Interestingly, the situation is essentially unaltered if the two bands have different bandwidths. The system undergoes a single bipolaronic transition, which is preceded by a polaronic crossover for one of the two phononic modes.

Nonetheless, the effect of electron-electron interaction can be significantly different in the present two-orbital case. In the single-band case, a charge-charge Hubbard repulsion directly competes with the Holstein term which couples the phonons to the local charge. This gives rise to a phase diagram characterized by the competition between phonon-driven and correlation-driven localization effects. In the present case, the phonons couple to electronic degrees of freedom other than the total onsite charge. This makes the competition between the two terms much more subtle. In a purely electronic model which coincides with (1) if the antiadiabatic limit is taken for phonons it has been indeed shown that phonon-mediated superconductivity can be enhanced by correlations and several anomalous properties can arise. The investigation of the combined effects of correlation and JT coupling in the present model is therefore expected to lead to a rich and interesting physics.

Acknowledgments

M.C. acknowledges financial support of MIUR PRIN 2007 Prot. 2007FW3MJX003