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Formation and temperature evolution of correlated polarons in colossal magnetoresistive manganites

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Abstract

We investigate the temperature dependence of charge ordered clusters in manganites. In terms of an approximate treatment of the half-filled spinless Holstein model incorporating the double exchange effect and nearest-neighbor Coulomb interactions, we find that charge ordering and polaronic states appear simultaneously which naturally defines a correlated polaron state. Without the double exchange, the temperature evolution of the charge (polaron) ordering parameter exhibits a broad peak structure only in the strongly correlated quasiadiabatic regime. Including the double exchange, the peak becomes sharper and is observed both in the quasiadiabatic and antiadiabatic regimes. The peak structure in the quasiadiabatic regime provides an explanation for the temperature dependence of the polaron correlation in nanoscale charge ordered polaron clusters observed in the manganites with intermediate to narrow bandwidths.

1. Introduction

Electron–phonon interaction (EPI) is believed to be important in strongly correlated electronic systems such as high-T_C superconducting cuprates [1, 2] and colossal magnetoresistive manganites [3, 4]. When the EPI is strong enough, polarons usually form which are physical entities made of electrons self-localized by the strong lattice deformations that surround them. While it has been accepted that the Jahn–Teller type of EPI plays an essential role in various properties of the manganites [3, 4], recently it has been observed that not just polarons but, more importantly, their correlation and the resulting short range cluster structures are of paramount importance to the large magnitude of various colossal effects observed in the manganites such as the colossal magnetoresistance effect (CMR) [5–9] and the colossal electroresistance effect (CER) [10]. These up-to-date findings stress the importance of studying many-polaron systems in the presence of strong electronic correlations [11–15].

In neutron and x-ray scattering experiments on the CMR manganites [5–8, 16], short range polaron correlations that are consistent with the formation of charge ordered clusters with ordering wavevector the same as in the half-doped case appear as the Curie temperature is approached from below. The intensity of these correlations then decreases with further increasing the temperature and so peaks very close to T_C. This is in sharp contrast to the number of the polarons which also increases rapidly as the Curie temperature is approached from below but then is roughly constant. The polaron ordering is shown to be short range in nature with a correlation length of about 10 Å, which is nearly temperature-independent [5, 7]. Since the intensity of the short range polaron correlations and the resistivity have very similar temperature dependence, it has been argued that correlated polarons instead of uncorrelated randomly localized polarons are primarily responsible for the insulating character of the paramagnetic state and the amplified magnetoresistive effects [7, 11].

While the existence and the peak structure in the temperature evolution of short range polaron correlations have both been obtained in numerical simulations based on the microscopic model for manganites [11], it is still unclear whether the nonmonotonic peak structure of the polaron correlations is a proprietary property of the CMR manganites where the double-exchange (DE) effect is essential [17] or a common feature of strongly correlated electronic systems with substantial EPI. In an exact diagonalization study of the t–J model coupled with various different types of EPI on small clusters, charge ordered structures which are indicative of correlated polaron states are observed, but the temperature evolution behavior is unexplored [18]. In a work studying the

reentrant behavior of the charge ordering (CO) transition in the manganites, the temperature evolution of the CO is investigated but the DE effect is not adequately taken into account [19, 20]. An earlier work [21] discussed the temperature evolution of nanoscale polaron clusters in the form of bipolarons, but the possibility of charge ordering was not investigated.

In the present work, we study the above problem in terms of the single-band spinless Holstein model incorporating the DE and nearest-neighbor Coulomb interaction simultaneously. Since the correlation length of the charge ordered polaron clusters is only weakly temperature-dependent [5, 7], the peak structure observed in experiments is regarded as reflecting the strength of the CO parameter. Therefore, in this paper, we study long range CO states in half-filled systems as an idealization and approximation of the short range clusters.

Calculations based on unbiased numerical methods such as exact diagonalization are usually restricted to small systems [13, 22] due to the limitation of computer capacity. On the other hand, simpler approximate treatments are known to give both qualitatively correct behavior of the system and also intuitive analytical results [23–28]. We treat the electron–electron interaction and the DE in terms of a mean-field approximation and perform the conventional Lang–Firsov transformation [26, 29–31] to eliminate the EPI part of the Hamiltonian. We show that polarons form simultaneously with the CO structure. The CO state in the presence of EPI is essentially a correlated polaron state and the CO evolution also characterizes the evolution of the intensity of polaron correlation. We further investigate the temperature evolution of the CO parameter, or equivalently, polaron correlation strength. Without the DE, a peak structure is observed in the strongly correlated quasiadiabatic regime, while in the antiadiabatic and less correlated quasiadiabatic regimes, the CO parameter decreases monotonically with increasing temperature. When the DE is considered, a peak in the temperature evolution of the CO parameter appears both in the quasiadiabatic and antiadiabatic regimes when the nearest-neighbor Coulomb interaction is sufficiently strong.

It is argued that the novel temperature dependence of the polaron correlations is a manifestation of the competition between the entropy of the polarons tending to destroy the CO (correlated polaron) structure and the bandwidth narrowing effect due to polaron formation and the DE effect, which tends to enhance the CO and polaron correlations. The above results provide an explanation for the experimentally observed peak structure in the temperature evolution of polaron correlations [5–8, 16] in the CMR manganites, which belong to the strongly correlated quasiadiabatic regime [3, 32, 33]. By explicitly including the double exchange mechanism, the obtained temperature evolution of the polaron correlation fits the experimental data for manganites better than previous works [19, 20]. Our result also indicates that a similar effect should be observable in other strongly correlated electronic systems without DE but with EPI in the quasiadiabatic regime.

The rest of this paper is organized as follows. In section 2, we introduce the model and illustrate the methods used to treat it. In section 3, we show that polarons always appear simultaneously with the CO. Then we study the temperature dependence of the charge (polaron) ordering for some typical parameter sets, and give qualitative explanations for the results. In the end, a brief summary is given in section 4.

2. Model and method

The Hamiltonian of the single-band spinless Holstein model with the DE and nearest-neighbor Coulomb interaction is written as [20]

$$H = -t\sum_{\mathbf{i},\boldsymbol{\delta}}\cos\left(\frac{\theta_{\mathbf{i},\mathbf{i}+\boldsymbol{\delta}}}{2}\right)c_{\mathbf{i}}^{\dagger}c_{\mathbf{i}+\boldsymbol{\delta}} + \frac{V}{2}\sum_{\mathbf{i},\boldsymbol{\delta}} n_{\mathbf{i}}n_{\mathbf{i}+\boldsymbol{\delta}} + \lambda\sum_{\mathbf{i}} n_{\mathbf{i}}Q_{\mathbf{i}} + \tfrac{1}{2}\sum_{\mathbf{i}}(\kappa Q_{\mathbf{i}}^2 + P_{\mathbf{i}}^2/M). \quad (1)$$

$c_{\mathbf{i}}$ ($c_{\mathbf{i}}^{\dagger}$) annihilates (creates) a spinless fermion on lattice site $\mathbf{i}$ and $n_{\mathbf{i}} = c_{\mathbf{i}}^{\dagger}c_{\mathbf{i}}$ is the fermion number operator on site $\mathbf{i}$. The first term of the Hamiltonian describes the electron hopping between nearest-neighboring lattice sites with $\boldsymbol{\delta}$ denoting the hopping directions. The factor $\cos(\frac{\theta_{\mathbf{i},\mathbf{i}+\boldsymbol{\delta}}}{2})$ accounts for the DE with $\theta_{\mathbf{i},\mathbf{i}+\boldsymbol{\delta}}$ representing the angle between the local spins residing on sites $\mathbf{i}$ and $\mathbf{i}+\boldsymbol{\delta}$, respectively [3, 17]. The second term stands for the Coulomb interaction between electrons residing on nearest-neighboring sites with strength V. Earlier works indicate that in manganites the nearest-neighbor Coulomb interaction functions similarly in stabilizing the CO state to the Jahn–Teller effect which is rooted in the twofold degeneracy of the e_g orbit [34–36]. In our simplified spinless single-orbit model, the nearest-neighbor Coulomb interaction is the only mechanism that may induce the CO. The third term represents a local coupling between the lattice displacement $Q_{\mathbf{i}}$ and the electron occupation. The last term denotes the elastic and kinetic energy of the phonons. The suppression of the spin degrees of freedom can be ascribed either to strong Hund coupling [3] or to strong on-site Coulomb correlation [37] in the manganites.

When the phonon operators of the Hamiltonian are written in the second quantization form, the Hamiltonian becomes

$$H = -t\sum_{\mathbf{i},\boldsymbol{\delta}}\cos\left(\frac{\theta_{\mathbf{i},\mathbf{i}+\boldsymbol{\delta}}}{2}\right)c_{\mathbf{i}}^{\dagger}c_{\mathbf{i}+\boldsymbol{\delta}} + \frac{V}{2}\sum_{\mathbf{i},\boldsymbol{\delta}} n_{\mathbf{i}}n_{\mathbf{i}+\boldsymbol{\delta}} + \sqrt{E_{\mathrm{P}}\omega_0}\sum_{\mathbf{i}} n_{\mathbf{i}}(b_{\mathbf{i}}^{\dagger}+b_{\mathbf{i}}) + \omega_0\sum_{\mathbf{i}} b_{\mathbf{i}}^{\dagger}b_{\mathbf{i}}, \quad (2)$$

where the constant energy of the zero-point motion of the lattice is neglected and $b_{\mathbf{i}}$ ($b_{\mathbf{i}}^{\dagger}$) is the annihilation (creation) operator of a phonon on site $\mathbf{i}$. Throughout this paper, we set $\hbar = k_{\mathrm{B}} = 1$. The relationship between the parameters in the two forms of the Hamiltonian, equations (1) and (2), are $\omega_0 = \sqrt{\kappa/M}$ and $E_{\mathrm{P}} = \lambda^2/(2\kappa)$. ω_0 is the energy of the single-mode phonon and E_{P} is the energy for polaron formation. There are three normalized characteristic energy scales in the problem. The first is V/t, measuring the strength of electron correlation favoring charge ordered phases. Another is ω_0/t which determines the degree of adiabaticity of the system [38]. The last is E_{P}/t which measures the strength of the EPI.

Following standard procedures we make approximations to the above model to arrive at a tractable theoretical framework. First we make the well-known Lang–Firsov

transformation [20, 26, 29–31] $\bar{H} = \mathrm{e}^{-S} H \mathrm{e}^{S}$ to eliminate the EPI term, where

$$S = \sqrt{\frac{E_\mathrm{P}}{N\omega_0}} \sum_{\mathbf{i},\mathbf{q}} \mathrm{e}^{\mathrm{i}\mathbf{q}\cdot\mathbf{R_i}} c_\mathbf{i}^\dagger c_\mathbf{i} (b_\mathbf{q}^\dagger - b_{-\mathbf{q}}). \tag{3}$$

N denotes the number of lattice sites which is also the number of wavevectors in the first Brillouin zone. The result of the transformation is

$$\bar{H} = -t \sum_{\mathbf{i},\boldsymbol{\delta}} \cos\left(\frac{\theta_{\mathbf{i},\mathbf{i}+\boldsymbol{\delta}}}{2}\right) c_\mathbf{i}^\dagger c_{\mathbf{i}+\boldsymbol{\delta}} X_\mathbf{i}^\dagger X_{\mathbf{i}+\boldsymbol{\delta}} + \frac{V}{2} \sum_{\mathbf{i},\boldsymbol{\delta}} n_\mathbf{i} n_{\mathbf{i}+\boldsymbol{\delta}} - E_\mathrm{P} \sum_\mathbf{i} n_\mathbf{i} + \omega_0 \sum_\mathbf{i} b_\mathbf{i}^\dagger b_\mathbf{i}, \tag{4}$$

where

$$X_\mathbf{i} = \exp\left[\sqrt{\frac{E_\mathrm{P}}{N\omega_0}} \sum_\mathbf{q} \mathrm{e}^{\mathrm{i}\mathbf{q}\cdot\mathbf{R_i}} (b_\mathbf{q} - b_{-\mathbf{q}}^\dagger)\right]. \tag{5}$$

The factor $X_\mathbf{i}^\dagger X_{\mathbf{i}+\boldsymbol{\delta}}$ describes the influence of the EPI on the electron hopping processes. In the lowest order approximation, we will replace it by its thermodynamic average, which turns out to be

$$\langle X_\mathbf{i}^\dagger X_{\mathbf{i}+\boldsymbol{\delta}} \rangle = \mathrm{e}^{-S_T}, \tag{6}$$

where

$$S_T = \sum_\mathbf{q} \frac{E_\mathrm{P}}{N\omega_0} (1 - \cos(\mathbf{q}\cdot\boldsymbol{\delta})) \coth\frac{\beta\omega_0}{2} = \frac{E_\mathrm{P}}{\omega_0} \coth\frac{\beta\omega_0}{2}, \tag{7}$$

with no dependence on either $\mathbf{i}$ or $\boldsymbol{\delta}$.

Next, we treat the Coulomb interaction term in the mean-field approximation by making the replacement $n_\mathbf{i} n_{\mathbf{i}+\boldsymbol{\delta}} \simeq n_\mathbf{i} \langle n_{\mathbf{i}+\boldsymbol{\delta}} \rangle + \langle n_\mathbf{i} \rangle n_{\mathbf{i}+\boldsymbol{\delta}} - \langle n_\mathbf{i} \rangle \langle n_{\mathbf{i}+\boldsymbol{\delta}} \rangle$. In this work, we focus on the half-filled case. The ansatz for the mean-field occupation number of the spinless fermions is taken to be

$$\langle n_\mathbf{i} \rangle = n + \delta n \cos(\mathbf{Q_0}\cdot\mathbf{R_i}), \tag{8}$$

where $\mathbf{Q_0}$ is the modulation wavevector which is determined by the filling fraction of the spinless fermions in the system. In half-doped bulk manganites, the CO state is of CE type characterized by a charge modulation wavevector of $(\pi, \pi, 0)$ with concomitant ferromagnetic zigzag chains coupled antiferromagnetically [3, 39, 40]. The nearest-neighbor antiferromagnetic couplings between local t_{2g} spins are shown to be crucial to stabilize this structure [40]. In this work aimed at providing a qualitative interpretation of the temperature evolution behavior of small half-doped clusters concomitant with the ferromagnetic to paramagnetic transition in less than half-doped manganites [5, 7], we will only consider ferromagnetic and paramagnetic CO states and avoid the above complications by considering the CO state characterized by $\mathbf{Q_0} = (\pi, \pi, \pi)$. This type of charge modulation is also most natural when the DE is absent.

Finally, we also treat the DE factor in terms of a mean-field approximation [41, 42]. The DE factor is rewritten in terms of the local spin vectors:

$$\cos\left(\frac{\theta_{\mathbf{i},\mathbf{i}+\boldsymbol{\delta}}}{2}\right) = \sqrt{\frac{S^2 + \mathbf{S_i}\cdot\mathbf{S}_{\mathbf{i}+\boldsymbol{\delta}}}{2S^2}}. \tag{9}$$

Near the Curie temperature, the DE factor can be approximately expanded as $\frac{1}{\sqrt{2}}(1 + \frac{\mathbf{S_i}\cdot\mathbf{S}_{\mathbf{i}+\boldsymbol{\delta}}}{2S^2})$, where higher-order terms of the expansion are neglected. Substituting this expansion into the Hamiltonian, the effective Hamiltonian for the local spins is obtained as

$$\bar{H}_S = -\frac{\sqrt{2}t}{4S^2} \mathrm{e}^{-S_T} \sum_{\mathbf{i},\boldsymbol{\delta}} c_\mathbf{i}^\dagger c_{\mathbf{i}+\boldsymbol{\delta}} \mathbf{S_i}\cdot\mathbf{S}_{\mathbf{i}+\boldsymbol{\delta}}. \tag{10}$$

In order to study the temperature evolution of the local spin system, we replace the electronic bond operator $c_\mathbf{i}^\dagger c_{\mathbf{i}+\boldsymbol{\delta}}$ by its average. Because we consider a long range CO phase, these averages should be independent of either $\mathbf{i}$ or $\boldsymbol{\delta}$. We have

$$c_\mathbf{i}^\dagger c_{\mathbf{i}+\boldsymbol{\delta}} \longrightarrow \langle c_\mathbf{i}^\dagger c_{\mathbf{i}+\boldsymbol{\delta}} \rangle = \frac{1}{Nz} \sum_{\mathbf{i},\boldsymbol{\delta}} \langle c_\mathbf{i}^\dagger c_{\mathbf{i}+\boldsymbol{\delta}} \rangle = \frac{-1}{zt} \epsilon_\mathrm{B}, \tag{11}$$

where z is the coordination number and ϵ_B is the band energy per site [41]. Defining the effective coupling constant between local spins as $J = \frac{\sqrt{2}}{4zS^2} \mathrm{e}^{-S_T}(-\epsilon_\mathrm{B})$, $\bar{H}_S$ becomes

$$\bar{H}_S \approx -J \sum_{\mathbf{i},\boldsymbol{\delta}} \mathbf{S_i}\cdot\mathbf{S}_{\mathbf{i}+\boldsymbol{\delta}}. \tag{12}$$

Applying the standard Curie–Weiss type of mean-field approximation to the above effective Hamiltonian, the magnetization per site is determined by the following self-consistent relationship:

$$\langle S_z \rangle / S = \frac{2S+1}{2S} \coth\left(\frac{2S+1}{2S} x\right) - \frac{1}{2S} \coth\left(\frac{1}{2S} x\right), \tag{13}$$

where $x = \frac{2zJS}{T} \langle S_z \rangle$. The Curie temperature T_C is determined by the following self-consistent relationship:

$$T_\mathrm{C} = \frac{2zJ(T_\mathrm{C})}{3} S(S+1) = \frac{\sqrt{2}(S+1)}{6S} \mathrm{e}^{-S_{T=T_\mathrm{C}}}(-\epsilon_\mathrm{B}(T_\mathrm{C})). \tag{14}$$

In order to study the CO state of the electrons, we approximately substitute the DE factor in equation (4) by [39]

$$\sqrt{\frac{1 + \mathbf{S_i}\cdot\mathbf{S_j}/S^2}{2}} \approx \sqrt{\frac{1 + \langle \mathbf{S_i}\cdot\mathbf{S_j} \rangle / S^2}{2}} = \sqrt{\frac{1 + (\langle S_z \rangle / S)^2}{2}} = f(T). \tag{15}$$

After the above approximations are made, we can Fourier transform the model Hamiltonian equation (4) into the wavevector space. The resulting Hamiltonian is written as

$$\bar{H} \approx \sum_{\mathbf{k}\in BZ} (-E_\mathrm{P} + \mathrm{e}^{-S_T} f(T) \epsilon_\mathbf{k}) c_\mathbf{k}^\dagger c_\mathbf{k} + \omega_0 \sum_\mathbf{q} b_\mathbf{q}^\dagger b_\mathbf{q} + \tfrac{1}{2} V \cdot \delta n \cdot \gamma_{\mathbf{Q_0}} \sum_{\mathbf{k}\in BZ} (c_\mathbf{k}^\dagger c_{\mathbf{k}+\mathbf{Q_0}} + c_{\mathbf{k}+\mathbf{Q_0}}^\dagger c_\mathbf{k}), \tag{16}$$

where $\gamma_{\mathbf{Q_0}} = \sum_\delta \mathrm{e}^{\mathrm{i}\mathbf{Q_0}\cdot\boldsymbol{\delta}}$ and $\epsilon_\mathbf{k} = -2t(\cos k_x + \cos k_y + \cos k_z)$. The temperature-dependent quantity e^{-S_T} defined in equation (7) is seen to be accounting for the effect of bandwidth narrowing by the formation of polarons. Since now the electronic and the phononic parts are independent, properties corresponding to the two systems can be treated separately. When finite temperature quantities are studied, a chemical

potential term which will be used to ensure the correct electron filling should be incorporated into the Hamiltonian which then becomes

$$\bar{K} = \bar{H} - \mu \sum_{\mathbf{k}\in BZ} c^{\dagger}_{\mathbf{k}} c_{\mathbf{k}}. \tag{17}$$

The chemical potential μ is determined by the condition

$$N_{\mathrm{e}} = \sum_{\mathbf{i}} \langle c^{\dagger}_{\mathbf{i}} c_{\mathbf{i}} \rangle_{\bar{K}} = \sum_{\mathbf{k}\in BZ} \langle c^{\dagger}_{\mathbf{k}} c_{\mathbf{k}} \rangle_{\bar{K}}, \tag{18}$$

where N_{e} is the total number of electrons in the system which is determined by the filling fraction.

3. Results and discussions

3.1. The correlated polaron state

According to the experiments on CMR manganites, polaron–polaron correlations lead to short range clusters of small lattice polarons with the ordering wavevector the same as that in the half-doped materials [6, 7]. The correlated polaron state [11, 43] can be defined to be a state composed of polarons distributed in an ordered pattern. The appearance of a polaronic state is indicated by the bimodal distribution of phonon displacement $\langle Q_{\mathbf{i}} \rangle$ [13, 15]. Because we have made a canonical transformation to the original Hamiltonian to eliminate the EPI term, the finite temperature quantum statistical averages should also be transformed accordingly. Taking an arbitrary operator $\hat{A}$ as an example

$$\begin{aligned}\langle \hat{A} \rangle_K &= \frac{1}{Z} \mathrm{Tr}(\mathrm{e}^{-\beta(H-\mu\hat{N})}\hat{A}) = \frac{1}{Z} \mathrm{Tr}(\mathrm{e}^{S}\mathrm{e}^{-\beta(\bar{H}-\mu\hat{N})}\mathrm{e}^{-S}\hat{A}) \\ &= \langle \mathrm{e}^{-S}\hat{A}\mathrm{e}^{S} \rangle_{\bar{K}} = \langle \hat{\tilde{A}} \rangle_{\bar{K}}, \end{aligned} \tag{19}$$

where $Z = \mathrm{Tr}(\mathrm{e}^{-\beta(H-\mu\hat{N})}) = \mathrm{Tr}(\mathrm{e}^{-\beta(\bar{H}-\mu\hat{N})})$ is the partition function. The lower labels K and $\bar{K}$ indicate that the quantum statistical averages are performed under $K(= H - \mu\hat{N})$ and $\bar{K}$, respectively. The definition of $Q_{\mathbf{i}}$ is

$$Q_{\mathbf{i}} = \sqrt{\frac{\omega_0}{2\kappa}}(b_{\mathbf{i}} + b^{\dagger}_{\mathbf{i}}) = \sqrt{\frac{\omega_0}{2N\kappa}} \sum_{\mathbf{k}\in BZ} (b_{\mathbf{k}} + b^{\dagger}_{-\mathbf{k}})\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}_{\mathbf{i}}}. \tag{20}$$

Its quantum statistical average is

$$\begin{aligned}\langle Q_{\mathbf{i}} \rangle_K &= \sqrt{\frac{\omega_0}{2N\kappa}} \sum_{\mathbf{k}\in BZ} [\langle \bar{b}_{\mathbf{k}} \rangle_{\bar{K}} + \langle \bar{b}^{\dagger}_{-\mathbf{k}} \rangle_{\bar{K}}]\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}_{\mathbf{i}}} \\ &= \sqrt{\frac{\omega_0}{2N\kappa}} \sum_{\mathbf{k}\in BZ} \Bigg[\langle b_{\mathbf{k}} \rangle_{\bar{K}} + \langle b^{\dagger}_{-\mathbf{k}} \rangle_{\bar{K}} \\ &\quad - 2\sqrt{\frac{E_{\mathrm{P}}}{N\omega_0}} \sum_{\mathbf{l}} \langle c^{\dagger}_{\mathbf{l}} c_{\mathbf{l}} \rangle_{\bar{K}} \mathrm{e}^{-\mathrm{i}\mathbf{k}\cdot\mathbf{R}_{\mathbf{l}}} \Bigg] \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}_{\mathbf{i}}} \\ &= -\frac{\lambda}{\kappa} \langle c^{\dagger}_{\mathbf{i}} c_{\mathbf{i}} \rangle_{\bar{K}} = -\frac{\lambda}{\kappa} \langle c^{\dagger}_{\mathbf{i}} c_{\mathbf{i}} \rangle_{K} = -\frac{\lambda}{\kappa} \langle n_{\mathbf{i}} \rangle_{K}. \end{aligned} \tag{21}$$

The above relationship is equivalent to those used in the exact diagonalization studies where the phonon operators are treated classically [13, 44, 45], and is valid no matter whether the DE is considered. It indicates that the polarons always form simultaneously with the appearance of the CO state, and the distribution of the polarons exactly follows the charge distribution. The strength of the polaron correlations is thus reflected by the CO parameter δn. Further works by more accurate numerical and analytical methods are favored to study the extent of the validity of the above approximate relationship.

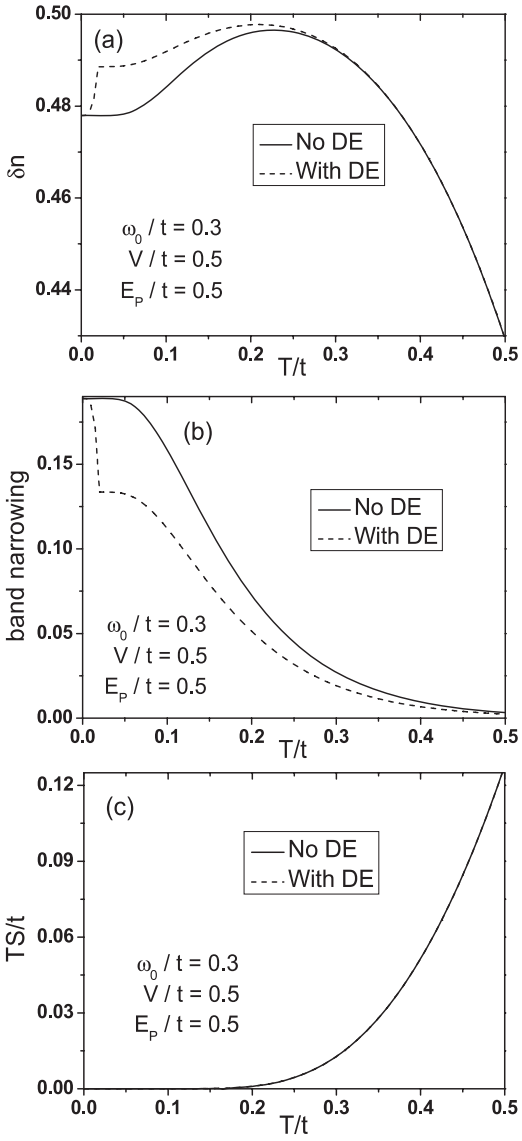

Figure 1. (a) Charge (polaron) ordering parameter, (b) band narrowing factor and (c) normalized entropy as functions of the normalized temperature with and without the DE, in the strongly correlated quasiadiabatic regime. Note: in (c), the two curves almost coincide and are hardly distinguishable.

3.2. Temperature evolution of the correlated polaron state

Figure 1(a) shows the temperature evolution of the charge (polaron) ordering parameter for $\omega_0/t = 0.3$, $V/t = 0.5$ and $E_{\mathrm{P}}/t = 0.5$. $\omega_0/t = 0.3$ corresponds to a typical quasiadiabatic phonon frequency [38]. The inter-site electron interaction $V/t = 0.5$ is strong compared to the typical values $V/t \sim 0.1$–0.2 estimated for the manganites due to a large dielectric constant [40]. The polaron binding energy $E_{\mathrm{P}}/t = 0.5$ characterizes a typical intermediate strength of the EPI [3]. Therefore, the present case corresponds to the strongly

correlated quasiadiabatic regime [38, 40]. A peak structure is observed in both the systems with and without the DE effect.

We argue that the peak structure appears as a result of the competition between the energy band narrowing effect and the entropy of the polaronic system. The energy band narrowing due to polaron formation and the DE effect reduces the effective hopping integral and so enhances the relative strength of the Coulomb interaction, which tends to promote the CO structure, while the entropy which increases with temperature tends to destroy any ordered structures. According to equation (16), the band narrowing effect is described by the dimensionless factor $e^{-S_T} \cdot f(T)$. The entropy per site from the electronic part of equation (17) can be calculated as

$$S = \frac{1}{2N} \sum_{\mathbf{k}\in BZ,\alpha} \left[\ln(1 + e^{-\beta\epsilon_{\mathbf{k}\alpha}}) + \frac{\beta\epsilon_{\mathbf{k}\alpha}}{e^{\beta\epsilon_{\mathbf{k}\alpha}} + 1} \right], \qquad (22)$$

where α labels the two eigenvalues obtained by diagonalizing the electronic part of equation (17) for a given $\mathbf{k}$. The temperature evolutions of the above two quantities are shown in figures 1(b) and (c). Both in the presence and in the absence of the DE effect, the bandwidth narrowing starts to show up at a much lower temperature than the entropy TS/t deviates from zero, which contributes to the initial increase of δn. The polaronic part of the bandwidth narrowing factor e^{-S_T} starts to decrease at $T/t \simeq 0.05$. The DE part of the bandwidth narrowing factor $f(T)$ begins to decrease at a much lower temperature and stops decreasing above the Curie temperature $T_C \simeq 0.02t$. When both the polaron formation and the DE are playing roles, the difference in the characteristic temperatures of the two factors results in a two-step increase of the CO parameter δn, as can be seen in figure 1(a). The entropy term is almost the same with or without the DE effect, as shown in figure 1(c). TS/t begins to increase rapidly at $T/t \simeq 0.22$, which leads to the decrease of δn. The competition between the bandwidth narrowing and the entropy thus gives rise to a peak structure in the temperature evolution of δn.

Figure 2 shows the result for $\omega_0/t = 3$, $V/t = 0.5$ and $E_P/t = 0.5$, which belongs to the strongly correlated antiadiabatic regime [38, 40]. From figure 2(a), only when the DE effect is considered can a peak structure be observed, even if $V/t = 0.5$ is large compared to realistic values for the CMR manganites, which are typically in the range $V/t \sim 0.1$–0.2 [40]. By comparing the two curves in figure 2(b), we can find that now the bandwidth narrowing, which induces the initial increase of δn, almost completely comes from the DE factor. The polaronic bandwidth narrowing will occur at a characteristic temperature proportional to ω_0, which is much higher than the temperature where the effect of the entropy term becomes significant, as can be seen from figures 2(b) and (c). This explains the absence of a peak structure in the absence of the DE effect. As compared to figure 1, where the polaronic bandwidth narrowing contributes dominantly to the appearance of the peak structure, the peak induced by the DE effect in figure 2 is sharper with a faster increase at low temperatures.

The temperature dependence of δn for several smaller values of V/t is shown in figure 3. For $V/t = 0.1$ and

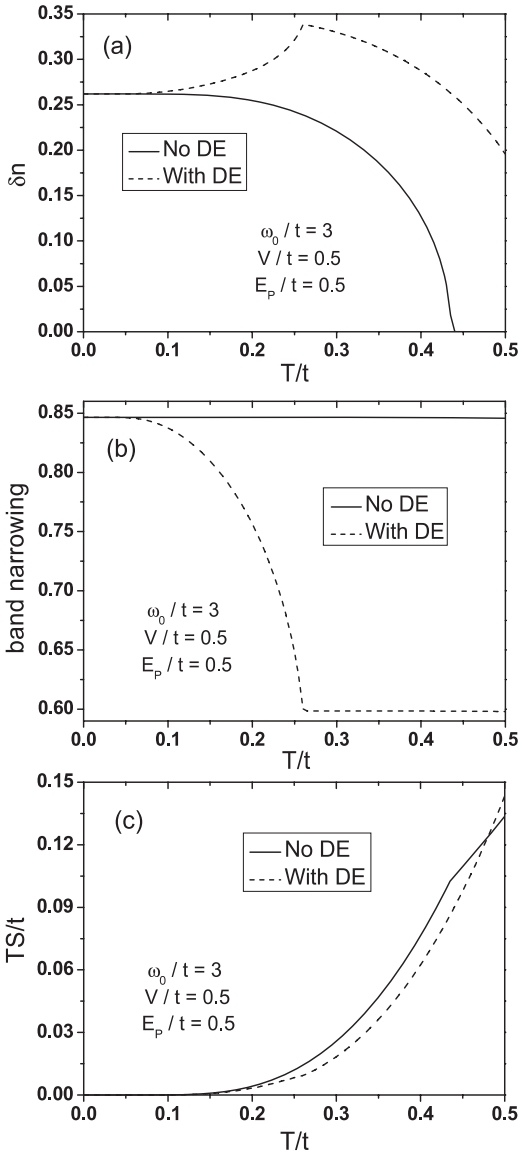

Figure 2. (a) Charge (polaron) ordering parameter, (b) band narrowing factor and (c) normalized entropy with and without the DE as functions of the normalized temperature in the strongly correlated antiadiabatic regime.

$\omega_0/t = 0.3$, as shown in figure 3(a), δn does not show a peak structure in the absence of the DE and a peak appears when the DE is switched on. This clearly indicates that the critical value of V/t for the appearance of the peak structure is reduced in the presence of the DE. In the presence of the DE, with a decrease of V/t to 0.08, the CO parameter exhibits a reentrant-like behavior [19, 20], which first decreases, then increases and finally decreases again with temperature, giving rise to a peak structure. The peaks observed in the above two cases are both due to the bandwidth narrowing effect caused by the DE effect, and the positions of the peaks are very close to the Curie temperatures. With a further decrease of V/t to 0.06, δn becomes monotonic with changing temperature without showing a peak even in the presence of the DE, as shown also in figure 3(b). In the antiadiabatic regime $\omega_0/t = 3$, δn decreases monotonically with temperature even for a relatively

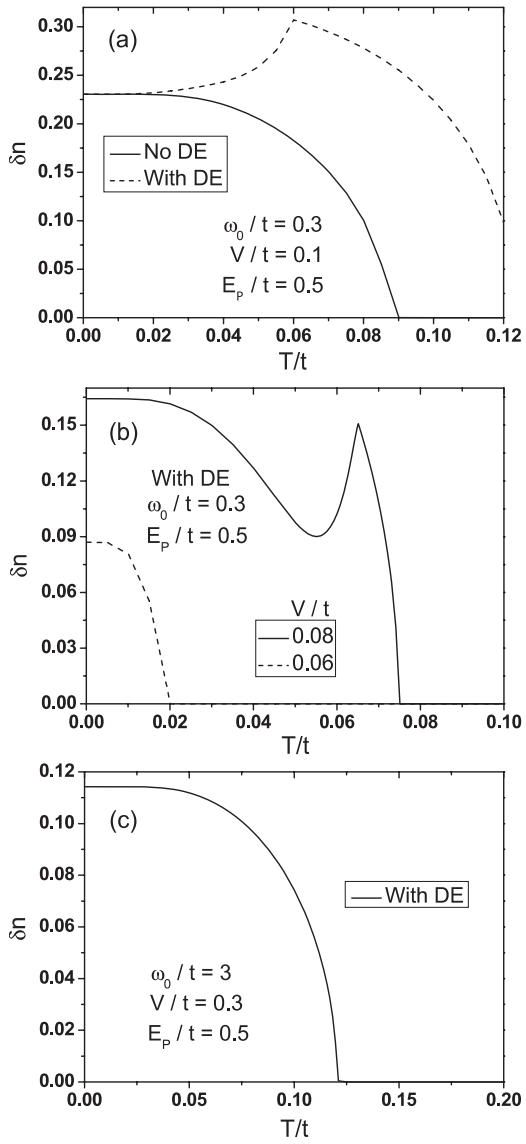

Figure 3. Temperature dependence of the charge (polaron) ordering parameter with and without the DE for some different parameter sets.

large $V/t = 0.3$ and in the presence of the DE, as shown in figure 3(c).

In the above calculations, we have fixed $E_P/t = 0.5$, and $\omega_0/t = 0.3$ and 3 are used as two characteristic phonon frequencies belonging respectively to the quasiadiabatic and antiadiabatic regimes. Changing the value of E_P/t will not change the qualitative features of the result. For the manganites, ω_0 is estimated to range from 0.06 to 0.09 eV, while the bare hopping integral t is estimated to be in the range of 0.2–0.5 eV [3]. Therefore, the CMR manganites with typical intermediate or narrow bandwidth characterized by relatively small t belong to the quasiadiabatic regime. Our results above indicate that the peak structure observed in the temperature evolution of the polaron correlations in the manganites should be a combined effect of both the DE and the EPI. Besides, our results also suggest that a similar peak structure may also occur in other strongly correlated electron systems without the DE in the quasiadiabatic regime.

4. Summary

Summarizing, we have studied the temperature evolution of the charge ordered state in terms of the half-filled spinless Holstein model including the DE and nearest-neighbor Coulomb interaction. We show that polarons always form simultaneously with the appearance of the charge order, which naturally defines a correlated polaron state. A formal relationship between the formation of polarons and the appearance of charge order in CMR manganites is provided and discussed for the first time. Our results demonstrate that, in the strongly correlated quasiadiabatic regime, the temperature evolution of the charge (polaron) ordering shows a peak structure both with and without the DE effect. In the strongly correlated antiadiabatic regime, only when the DE effect is included can a peak structure appear. In other parameter regimes where the charge order can appear, monotonic temperature dependence of the CO parameter is observed. The results are argued to arise from the competition between the band narrowing effect and the entropy of the polaronic system. Because the manganites usually belong to the strongly correlated quasiadiabatic regime, the above result gives an explanation to the peculiar peak structure observed in the temperature evolution of the intensity of the polaron correlations in many CMR manganites.

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