Effect of quantum-lattice fluctuations in one-dimensional fluctuating-valence systems

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We have developed a variational approach to treat the nonadiabaticity, that is, the quantum-lattice fluctuations, of the electron-phonon interactions in the one-dimensional half-filled spinless Anderson lattice model including Coulomb interaction between both types of electrons. The nonadiabaticity due to finite phonon frequency is treated through a variational polaronic-type wave function, in which two variational parameters δ and τ^2 are used to take into account the dynamical distortion and the squeezing effect of phonon modes. We have found that the quantum-lattice fluctuations gradually smoothes the valence transitions when the polaronic level $\varepsilon_f - v$ changes. We have shown that conditions somewhat different from those of Hewson and Newns should be satisfied for the occurrence of a significant reduction of the effective hybridization. The effect of the Coulomb repulsion U is to suppress the quantum-lattice fluctuations, that is, to suppress the reduction of the fluctuating-valence frequency and the relaxation shift. We have also discussed the valence-density-wave ordering in the symmetric case. Our results show that the quantum-lattice fluctuations disfavor the ordering and the lattice dimerization parameter decreases with increasing phonon frequency. We have pointed out the possibility of an order-disorder transition in such systems.

I. INTRODUCTION

The theory of fluctuating-valence (FV) solids has been a subject of considerable investigation for decades,[1] but several important issues still remain open. An area of special interest, with some controversies and unanswered questions, is the full description of the electron-phonon interaction in FV systems. Usually, the Anderson-impurity model[2] or the periodic Anderson lattice (PAL) model is used to investigate the physical properties of FV systems, in which the f electrons are coupled to the conducting electrons via an on-site hybridization V. As V is usually assumed to be comparable to the relevant phonon energy, and because there is a large difference in the ionic radii associated with the two valence states, strong and nonadiabatic electron-phonon coupling may arise[3-9] and this might considerably reduce the frequency of valence fluctuations and thus effectively increase the lifetime of the individual configurations. This nonadiabatic electron-phonon coupling so far mostly been dealt with within a model of linear on-site coupling of the f-electron density to the local phonon mode for both the PAL model[3,5] and the single Anderson-impurity limit.[4,6-9]

There is a controversy about the role played by the quantum-lattice fluctuations in reducing the frequency of valence fluctuations and increasing the lifetime of the individual configurations. Sherrington and von Molnar[3] concluded that, for low-temperature properties there is a large polaronic reduction of the effective hybridization:

$$V \rightarrow V \exp(-v/2\omega) = V' , \tag{1}$$

if the electron-phonon coupling g is sufficiently large that the polaronic binding energy $v = g^2/\omega$ is larger than V and ω. This result is obtained by using perturbation theory in the hybridization V. Hewson and Newns[4] proposed a variational ansatz to deal with the Anderson-impurity model coupled to the local phonon mode and found more restrictive conditions than Sherrington and von Molnar for the renormalization of V.

Hanke and Schmeltzer,[5] by using the real-space renormalization-group method to the one-dimensional half-filled spinless PAL model coupled to phonons, concluded that when the Coulomb interaction between f and d electrons goes to zero the long-range quantum fluctuations of the boson field occur and they suppress the standard polaronic effects, i.e., the reduction of the FV frequency and the relaxation shift.

The large-degeneracy limit for infinitely large f-f repulsion of the Anderson-impurity model coupled to the local phonon mode was studied by several authors[6-8] because in this case some exact results can be obtained by diagonalizing a tridiagonal matrix. By using this method Schonbammer and Gunnarsson[6] concluded that the large polaronic reduction of the effective hybridization, $V \rightarrow V'$ [Eq. (1)], does not occur.

Zheng and Nasu[9] proposed a variational approach—the squeezed polaron state—to study the spinless Anderson-impurity model coupled to the local phonon mode. The nonadiabaticity effect due to finite phonon frequency are treated through a variational wave function, in which the softening of the phonon frequency as a result of electron-phonon interaction is taken into consideration by means of the squeezing transformation. Their results show that a significant reduction of the

effective hybridization do not occur even if the condition of Hewson and Newns[4] for its occurrence are satisfied.

The aim of this work is to present, by means of the squeezed polaron variational approach,[9,10] a consistent description of the electron-phonon interaction in one-dimensional spinless PAL model for the parameter range where ω, v, and V are comparable, which makes any rigorous analysis of the problem extremely difficult and variational results very useful. In Sec. II, two unitary transformations are used to take into account the static and dynamical distortion of the lattice, respectively. Then, a squeezed-phonon state is introduced as the ground-state wave function of the phonon subsystem, which favors the quantum lattice fluctuations and improves the energy estimate of the ground state. We divide our discussion of the ground-state properties into two parts: the unsymmetric (Sec. III) and symmetric (Sec. IV) cases. For the unsymmetric case only the uniform phase will be discussed. Conditions somewhat different from those of Hewson and Newns[4] for the occurrence of a significant reduction of the effective hybridization will be derived from our numerical results.

In this paper, only the spinless electron case will be discussed and in this case the PAL model is nothing but the famous Falicov-Kimball (FK) model plus an on-site f-d hybridization V. For a special choice of parameters (the symmetric case), that is, ϵ_f (the bare f level) = 0 and N_e (total number of spinless electrons)$=N$ (the total number of cells), it was shown[11-13] that the exact ground state of the FK model is structural. [In the two-dimensional case people call the structure a chessboard ordering phase[11,12] but, as we deal with the one-dimensional case, it will be called a valence-density-wave (VDW) ordering[13,14] in this paper.] The nonzero f-d hybridization V should suppress the ordering since it induces the finite lifetime of local f state. Inclusion of the electron-phonon interaction makes the situation more complicated. Adiabatic electron-phonon coupling favors the VDW ordering, as was shown by the famous Peierls' theorem.[15] However, the quantum-lattice fluctuations should make the ordering state less stable. These will be discussed in Sec. IV.

Some concluding remarks will be given in Sec. V. In this work only the one-dimensional case will be discussed because the quantum-lattice fluctuation plays the most important role in one dimension. Besides, we put $\hbar=1$ within this paper.

II. THEORY

We start from the following one-dimensional spinless periodic Anderson model with local d-f Coulomb interaction plus an electron-lattice interaction on a local f level,

$$H=-\sum_i t(d_i^\dagger d_{i+1}+d_{i+1}^\dagger d_i)+\sum_i \epsilon_f f_i^\dagger f_i + \sum_i V(f_i^\dagger d_i+d_i^\dagger f_i)+\sum_i U d_i^\dagger d_i f_i^\dagger f_i + \sum_i g(b_i^\dagger+b_i)f_i^\dagger f_i+\sum_i \omega(b_i^\dagger b_i+\tfrac{1}{2}) . \quad (2)$$

It includes spinless conduction d electrons with nearest-neighbor hopping parameter t, localized f electrons which have an energy level ϵ_f (the zero point of energy scale is fixed at $\epsilon_d=0$), and an on-site hybridization V and an on-site Coulomb repulsion U between d and f electrons. The total number of spinless electrons is N in the case of half filling, where N is the total number of sites. In H, $b_i^\dagger(b_i)$ creates (destroys) the local Einstein phonon mode of frequency ω, which couples linearly to the occupancy of the local f level. Similar model Hamiltonians have been used previously in the discussions of the effect of quantum-lattice fluctuations in valence-fluctuating systems.[3-9]

In the following, two unitary transformations will be used to introduce variational parameters. The first one is a "shift" transformation

$$H_1=\exp(S_1)H\exp(-S_1) , \quad (3)$$

$$S_1=\sqrt{M\omega/2}\sum_i (b_i^\dagger-b_i)[x_0+(-1)^i m_0] , \quad (4)$$

where M is the mass parameter of the phonon mode. x_0 denotes the in-phase ($Q=0$) displacement but m_0 the out-of-phase ($Q=\pi$) displacement of the equilibrium position of oscillators. Here we use a nonzero m_0 to take into account the possibility of the staggered ordering of lattice. The second transformation is a modified Lang-Firsov transformation which introduces the dynamical distortion of the phonon modes related to the hopping on and off of the f electrons:

$$H_2=\exp(S_2)H_1\exp(-S_2) , \quad (5)$$

$$S_2=\frac{g\delta}{\omega}\sum_i (b_i^\dagger-b_i)[f_i^\dagger f_i-\sqrt{K/2v}\,x_0] , \quad (6)$$

where $v=g^2/\omega$ and $K=M\omega^2$. δ measures the dynamical distortion of the phonon modes and will be treated as a variational parameter. It is essential here to associate the polaronic distortion to the number fluctuation of f electrons, $f_i^\dagger f_i-\sqrt{K/2v}\,x_0$, but not to $f_i^\dagger f_i$ [later we shall show by variational principle that $\sqrt{K/2v}\,x_0=(1/N)\sum_i\langle f_i^\dagger f_i\rangle$]. The average of the number operator $\langle f_i^\dagger f_i\rangle$ is coupled to the average distortion of the phonon mode, which has been taken into account by our first transformation.

Now we decouple the electron and the phonon subsystems in the transformed Hamiltonian H_2 by averaging it over the squeezed-phonon state[9,10] instead of the vacuum state of phonons,

$$|\mathrm{ph},s\rangle=\exp(-S_3)|\mathrm{ph},0\rangle , \qquad S_3=\sum_i \alpha(b_i b_i-b_i^\dagger b_i^\dagger) , \quad (7)$$

where $|\mathrm{ph},s\rangle$ is the squeezed-phonon state and $|\mathrm{ph},0\rangle$ the vacuum state of phonons. Here α will also be treated as a variational parameter. After the separation of the electron and the phonon subsystems, we get an effective Hamiltonian for electrons:

$$H_{\rm eff}=\langle {\rm ph},s|H_2|{\rm ph},s\rangle$$
$$=\frac{K}{2}x_0^2(1-\delta)^2N+\frac{K}{2}m_0^2N+\frac{\omega}{4}(\tau^2+\tau^{-2})N-\sum_i t(d_i^\dagger d_{i+1}+d_{i+1}^\dagger d_i)+\sum_i \epsilon_f' f_i^\dagger f_i$$
$$-\sqrt{2vK}\,m_0(1-\delta)\sum_i(-1)^i f_i^\dagger f_i+\sum_i V\rho(f_i^\dagger d_i+d_i^\dagger f_i)+\sum_i U d_i^\dagger d_i f_i^\dagger f_i\ , \tag{8}$$

where

$$\epsilon_f'=\epsilon_f-v+v(1-\delta)^2[1-\sqrt{2K/v}\,x_0]\ , \tag{9}$$

$$\rho=\exp\left[-\frac{v}{2\omega}\delta^2\tau^2\right]\ ,\quad \tau=\exp(-2\alpha)\ . \tag{10}$$

We make Hartree-Fock approximation to the interaction term:

$$d_i^\dagger d_i f_i^\dagger f_i\approx n_{di}f_i^\dagger f_i+n_{fi}d_i^\dagger d_i-n_{di}n_{fi}-d_i^\dagger f_i\langle f_i^\dagger d_i\rangle-f_i^\dagger d_i\langle d_i^\dagger f_i\rangle+\langle d_i^\dagger f_i\rangle\langle f_i^\dagger d_i\rangle\ , \tag{11}$$

where $n_{di}=\langle d_i^\dagger d_i\rangle$, $n_{fi}=\langle f_i^\dagger f_i\rangle$. Then the effective Hamiltonian is approximated as

$$H_{\rm eff}\approx\frac{K}{2}x_0^2(1-\delta)^2N+\frac{K}{2}m_0^2N+\frac{\omega}{4}(\tau^2+\tau^{-2})N-\sum_i Un_{di}n_{fi}+U\Delta^2N-\sum_i t(d_i^\dagger d_{i+1}+d_{i+1}^\dagger d_i)$$
$$+\sum_i Un_{fi}d_i^\dagger d_i+\sum_i(V\rho-U\Delta)(f_i^\dagger d_i+d_i^\dagger f_i)+\sum_i(\epsilon_f'+Un_{di})f_i^\dagger f_i-\sqrt{2vK}\,m_0(1-\delta)\sum_i(-1)^i f_i^\dagger f_i\ , \tag{12}$$

where we have assumed that

$$\Delta=\langle d_i^\dagger f_i\rangle=\langle f_i^\dagger d_i\rangle \tag{13}$$

is a uniform quantity even in the structural phase. By using the variational principle to the parameter x_0, one can show that

$$\sqrt{K/2v}\,x_0=\frac{1}{N}\sum_i\langle f_i^\dagger f_i\rangle\ . \tag{14}$$

In the following we shall discuss the solutions of this Hamiltonian in the case of $\epsilon_f\neq v$ (unsymmetric case) and that of $\epsilon_f=v$ (symmetric case) separately.

III. UNSYMMETRIC CASE

In unsymmetric case $\epsilon_f\neq v$, we assume that the system is uniform and there is no structural phase,

$$m_0=0\ ,\quad n_{fi}=n_f\ ,\quad n_{di}=n_d=1-n_f\ . \tag{15}$$

Thus, the ground-state energy of coupling system is

$$E_g/N=v(1-\delta)^2n_f^2+\frac{\omega}{4}(\tau^2+\tau^{-2})+\frac{U}{2}-U(1-n_f)n_f$$
$$+U\Delta^2-\frac{1}{2N}\sum_k\sqrt{(E_d+\epsilon_k-E_f)^2+4V'^2}\ , \tag{16}$$

where $E_d=Un_f$, $E_f=Un_d$,

$$\Delta=\frac{-1}{N}\sum_k\frac{V'}{\sqrt{(E_d+\epsilon_k-E_f)^2+4V'^2}}\ , \tag{17}$$

$$n_f=\frac{1}{2}+\frac{1}{2N}\sum_k\frac{E_d+\epsilon_k-E_f}{\sqrt{(E_d+\epsilon_k-E_f)^2+4V'^2}}\ , \tag{18}$$

$$\epsilon_k=-2t\cos k,\quad V'=V\rho-U\Delta\ . \tag{19}$$

The model system (2), as far as the ground-state properties are concerned, are defined by five input parameters which we can take to be the phonon frequency ω, the polaron binding energy v, the bare f level ϵ_f, the hybridization V, and the Coulomb repulsion U. The parameter t, the hopping integral of electrons, can be set equal to 1 by redefining the overall energy scale. In the numerical calculations we should, first of all, for every input set of parameters adjust δ and τ to let E_g/N reach at a stable minimum. (We shall denote the optimum values of δ and τ as δ_m and τ_m, respectively.) Then other physical quantities can be determined. In the following we give out some numerical results for the unsymmetric case.

In Fig. 1 we plot n_f versus polaronic level ϵ_f-v relations in the case of $V=0.2t$, $U=4t$, $v=t$, and $\omega=0$, $0.02t$, $0.1t$, $0.2t$, respectively. Here the input values of U and v are relatively large. When $\omega\to 0$, i.e., in the adiabatic limit, the valence transition from $\epsilon_f-v<0$ to

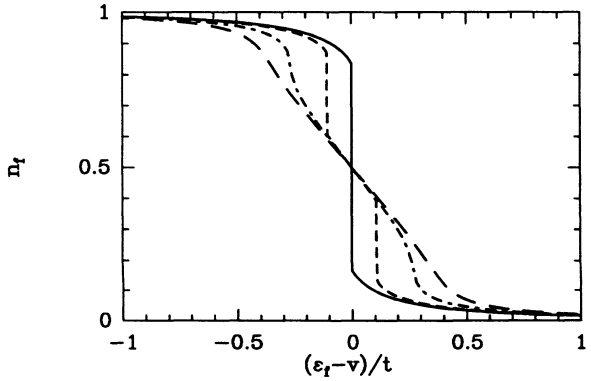

FIG. 1. n_f vs ϵ_f-v relation in the case of $V=0.2t$, $U=4t$, $v=t$, and $\omega=0$ (solid line), $0.02t$ (short-dashed line), $0.1t$ (dash-dotted line), and $0.2t$ (long-dashed line).

$\epsilon_f - v > 0$ is discontinuous. Note that with approximation (11) for the U term the valence transition is always continuous when $v = 0$.[16,17] The same result is also obtained with an alloy analog approximation.[18] For $U = 0$ discontinuous transitions are possible for $v > v_c$ either in the impurity[9] or the lattice case.[17] We can see that a finite phonon frequency can smear out this discontinuity. When ω is small ($\omega = 0.02t$ in this figure) n_f has some sharp transition at $|\epsilon_f - v| > 0$ ($|\epsilon_f - v| \sim 0.1t$ in this figure). Further increase of the phonon frequency ($\omega = 0.1t$ and $0.2t$ in this figure) makes the transition smoother and smoother.

Figure 2 shows the optimum values of the variational parameters δ_m, τ_m^2, and the corresponding reduction factor ρ as functions of $\epsilon_f - v$ in the case of $V = 0.2t$, $U = 4t$, $v = t$, and $\omega = 0.1t$. These curves are symmetric about $\epsilon_f - v = 0$. Around $|\epsilon_f - v| \sim 0$, δ_m arrives at its maximum and τ_m^2 at its minimum; this indicates that around $|\epsilon_f - v| \sim 0$ both the dynamical distortion and the squeezing of the phonon mode are the strongest. We note that the minimum value of the squeezing parameter in this figure is $\tau_m^2 = 0.356$, which corresponds to $\alpha_m = -\frac{1}{4}\ln\tau_m^2 = 0.258$ and indicates a strong squeezing effect.[9,10] ρ also has a minimum around $|\epsilon_f - v| \sim 0$, where $\rho = 0.561$. This value is much larger than what the reduction factor in Eq. (1) would give:

$$V'/V = \exp(-v/2\omega) = 0.0067 , \tag{20}$$

although the similar conditions as those of Hewson and Newns[4] for a significant reduction factor (20) of the effective hybridization in the Anderson-impurity model is to be observed, that is,

$$v > V, v > \omega > |\epsilon_f - v| , \omega > V' , \tag{21}$$

are all satisfied in the case of Fig. 2 when $|\epsilon_f - v| \sim 0$.

In Fig. 3, we plot n_f versus $\epsilon_f - v$ relation in the case of $V = 0.2t$, $U = 0.3t$, $v = 0.6t$, and $\omega = 0$, $0.1t$, respectively. Here the input value of U is small. When $\omega = 0$, i.e., in the adiabatic limit, the valence transition from $\epsilon_f - v < 0$ to $\epsilon_f - v > 0$ is already continuous. A finite

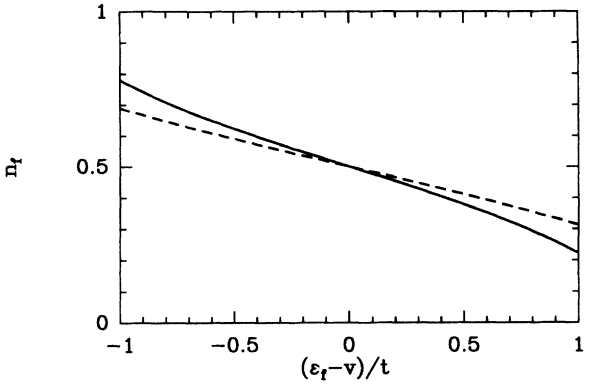

FIG. 3. n_f vs $\epsilon_f - v$ relation in the case of $V = 0.2t$, $U = 0.3t$, $v = 0.6t$, and $\omega = 0$ (solid line), $0.1t$ (dashed line).

phonon frequency ($\omega = 0.1t$ in this figure) smooths it further.

Figure 4 shows the optimum values of the variational parameters δ_m, τ_m^2, and the corresponding reduction factor ρ as functions of $\epsilon_f - v$ in the case of $V = 0.2t$, $U = 0.3t$, $v = 0.6t$, and $\omega = 0.1t$. These curves are also symmetric about $\epsilon_f - v = 0$ and are very smooth. The dynamical distortion δ_m, related to the relaxation shift, is large (~ 0.97) and the squeezing effect is small. A significant reduction of the effective hybridization appears for all values of $e_f - v$ ($\rho \sim 0.07$ in this figure). We note that the reduction factor of Eq. (1) in this case is

$$\exp(-v/2\omega) = 0.0498 .$$

Another quantity of interest is the fluctuation of the oscillator coordinates $q_i = (b_i^\dagger + b_i)/\sqrt{2M\omega}$. The part of the lattice deformation, the dynamical distortion which follows the electronic motion, is proportional to the f-electron density, thus fluctuates as the electrons move. The anomalous fluctuations can be much larger than the ordinary zero-point fluctuations if $g > 1$. The fluctuation in every phonon mode q_i can be expressed as

$$\langle \Delta q_i^2 \rangle = \langle q_i^2 \rangle - \langle q_i \rangle^2 , \tag{22}$$

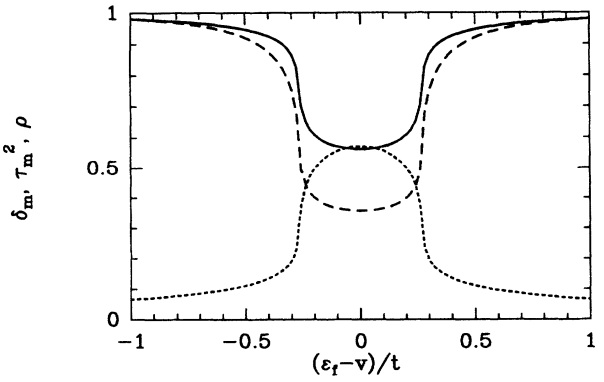

FIG. 2. δ_m (dotted line), τ_m^2 (dashed line), and ρ (solid line) as functions of $\epsilon_f - v$ in the case of $V = 0.2t$, $U = 4t$, $v = t$, and $\omega = 0.1t$.

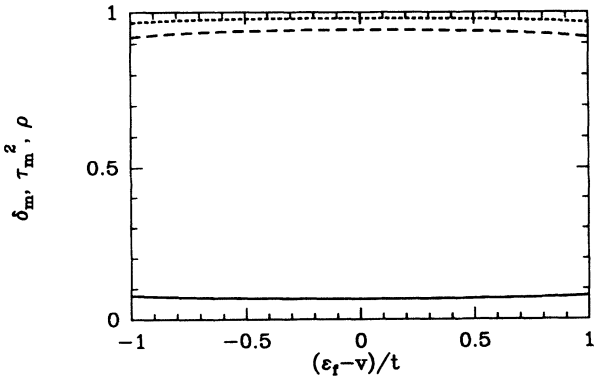

FIG. 4. δ_m (dotted line), τ_m^2 (dashed line), and ρ (solid line) as functions of $\epsilon_f - v$ in the case of $V = 0.2t$, $U = 0.3t$, $v = 0.6t$, and $\omega = 0.1t$.

where $\langle \cdots \rangle$ represents an average over the ground state of the total system. After the two unitary transformations [Eqs. (3) and (5)] and making an average over the squeezed-phonon state $|\mathrm{ph},s\rangle$ we have

$$\langle \Delta q_i^2 \rangle = \frac{1}{2M\omega}\left[\tau_m^{-2} + 4\frac{g^2}{\omega^2}\delta_m^2 n_f(1-n_f)\right] . \quad (23)$$

The ordinary zero-point fluctuation of a harmonic oscillator is

$$\langle \Delta q_i^2 \rangle_0 = \frac{1}{2M\omega} .$$

We define the dimensionless anomalous fluctuation as

$$f = (\langle \Delta q_i^2 \rangle - \langle \Delta q_i^2 \rangle_0)/\langle \Delta q_i^2 \rangle_0$$
$$= (\tau_m^{-2}-1) + 4\frac{g^2}{\omega^2}\delta_m^2 n_f(1-n_f) . \quad (24)$$

Here the first term $\tau_m^{-2}-1$ reflects the squeezing effect, and the second term, proportional to both f- and d-electron density n_f and $n_d = 1-n_f$, reflects the anomalous fluctuations due to the electron motion. The solid line in Fig. 5 shows f as a function of $\epsilon_f - v$ in the same case as that of Fig. 2, in which the Coulomb repulsion U is relatively large. One can see that the anomalous fluctuation is the strongest around $|\epsilon_f - v| \sim 0$. The dashed line in Fig. 5 shows f as a function of $\epsilon_f - v$ in the same case as that of Fig. 4, in which the Coulomb repulsion U is small. In this case the anomalous fluctuation is large for all values of $\epsilon_f - v$.

Also interesting is to see the effect of correlation term U on the quantum-lattice fluctuations. Figures 6(a) and 6(b) show the optimum values of the variational parameters δ_m and τ_m^2, respectively, as functions of U in the case of $V=0.2t$, $|\epsilon_f - v| = 0.05t$, and four different sets of values of v and ω. One can see that the dynamical distortion of phonon modes, proportional to δ_m, decreases with increasing U. This means that the on-site correlation between d and f electron suppresses the dynamical distortion, which is related directly to the relaxation shift.

Figure 6(c) shows the reduction factor ρ versus U relations in the same case. Here one can see that a significant reduction of the effective hybridization appears for $v=0.6t$, $\omega=0.1t$ and $U<v$. We note that this is the same case as that in Fig. 4. Combining the results shown in Figs. 4 and 6(c) we believe that in the spinless PAL model a significant reduction of the effective hybridization appears when

$$v > V \ , v > \omega \ , \omega > V' \ , \text{ and } U < v . \quad (25)$$

These conditions are somewhat different from those [Eq. (21)] of Hewson and Newns[4] for the occurrence of a significant reduction factor of the effective hybridization in the Anderson-impurity model.

Figure 7 shows the anomalous fluctuation f as a function of U in the same case as that of Fig. 6. The anomalous fluctuation decreases with increasing U, which means that the on-site Coulomb repulsion suppresses the quantum-lattice fluctuations. The physical picture of this suppression is that the on-site repulsion favors the single occupation of every site ($N_e = N$ in our calculation), which, in turn, obstructs the hopping of electrons on and off the localized f level and the related dynamical distortion of the phonon mode. We note that the anomalous fluctuation shown in this figure is the strongest when conditions in Eq. (25) are satisfied.

The correlation term U suppresses the quantum-lattice fluctuations, which is the same as what Hanke and

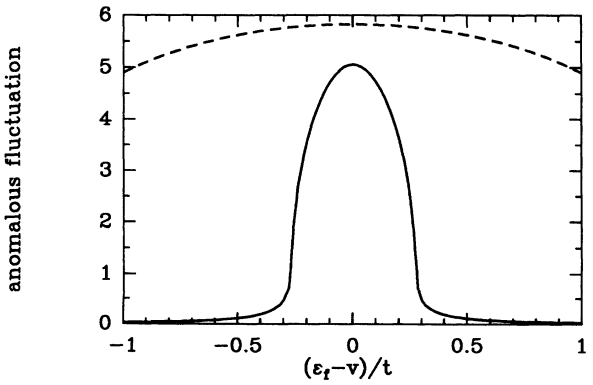

FIG. 5. Anomalous fluctuation f as a function of $\epsilon_f - v$ in the case of $V=0.2t$, $U=4t$, $v=t$, $\omega=0.1t$ (solid line), and $V=0.2t$, $U=0.3t$, $v=0.6t$, and $\omega=0.1t$ (dashed line).

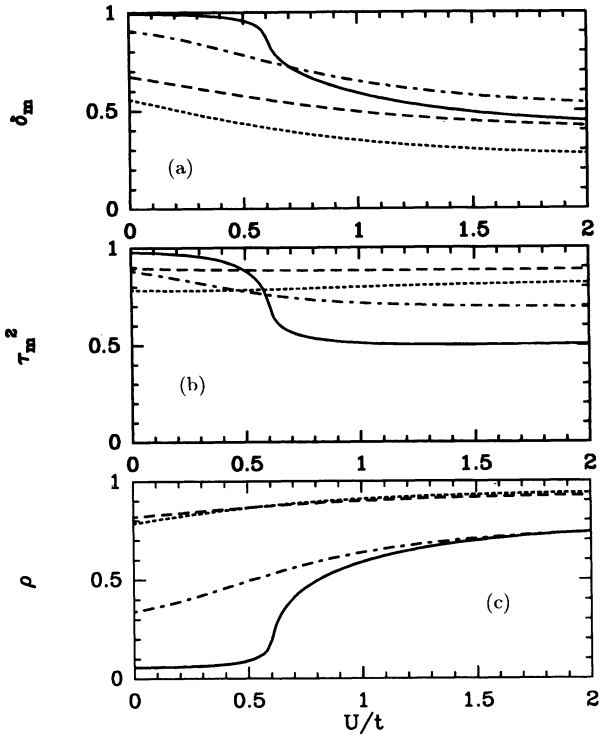

FIG. 6. (a) δ_m vs U relations in the case of $V=0.2t$, $|\epsilon_f - v| = 0.05t$, and $v=0.2t$, $\omega=0.1t$ (dotted line); $v=0.2t$, $\omega=0.2t$ (dashed line); $v=0.6t$, $\omega=0.1t$ (solid line); $v=0.6t$, $\omega=0.2t$ (dash-dotted line). (b) τ_m^2 vs U relations in the same case as that of (a). (c) ρ vs U relations in the same case as that of (a).

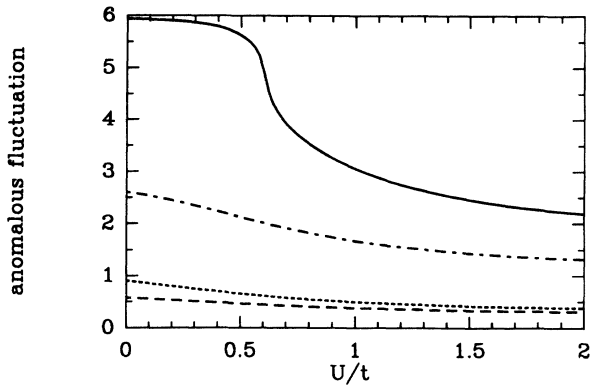

FIG. 7. Anomalous fluctuation f vs U relations in the same case as that of Fig. 6(a).

Schmeltzer[5] pointed out. However, from their real-space renormalization-group calculation they concluded that when the correlation term U goes to zero, the long-range electronic correlations and the related phonon quantum fluctuations suppress the standard polaronic effects and no reduction of the FV frequency with accompanying lifetime enhancement and no relaxation shift occur. This is different from our results. As one can see from our numerical results that the reduction of the FV frequency and the relaxation shift connected with the dynamical distortion δ_m are both the strongest when $U\to 0$ and $|\epsilon_f - v|\sim 0$. We believe that the difference between our results and those of Hanke and Schmeltzer[5] comes from that they fix the bare f level by $E_f = E_d$ ($\epsilon_f = 0$ in our notation). So in their calculations the polaronic level $\epsilon_f - v = -v$. When v is not small the effective f-level occupancy n_f should be near 1 and this could suppress the reduction of the FV frequency and the relaxation shift. We note that the quantum-lattice fluctuations are the cause of the reduction of the FV frequency and the relaxation shift in one-dimensional half-filled PAL model coupled to phonons because when $\omega = 0$ there is no quantum-lattice fluctuations, the lattice oscillations cannot follow the hopping of electrons on and off the localized f level and thus no reduction of the effective hybridization and no relaxation shift.

IV. SYMMETRIC CASE

When $\epsilon_f = v$ it is easy to prove that $x_0 = \sqrt{v/2K}$ and thus $\epsilon_f' = 0$ in H_{eff}. We assume that in this case the system can go into a structural phase (the so-called VDW phase) (Refs. 11–14) and define the f and d electron staggered ordering parameters m_f and m_d as follows:

$$\langle f_i^\dagger f_i \rangle = \tfrac{1}{2} + (-1)^i m_f \ , \tag{26}$$

$$\langle d_i^\dagger d_i \rangle = \tfrac{1}{2} + (-1)^i m_d \ . \tag{27}$$

Now the ground-state energy is

$$E_g/N = \frac{v}{4}(1-\delta)^2 + \frac{K}{2}m_0^2 + \frac{\omega}{4}(\tau^2+\tau^{-2}) + \frac{U}{4} - Um_d m_f + U\Delta^2$$
$$- \frac{1}{N}\sum_{k>0}\{\epsilon_k^2 + U^2 m_f^2 + (X-Um_d)^2 + 2V'^2 + 2\sqrt{(X-Um_d)^2\epsilon_k^2 + [(X-Um_d)Um_f + V'^2]^2}\}^{1/2} \ , \tag{28}$$

where

$$m_f = \frac{1}{N}\sum_{k>0}\left\{(X-Um_d) + \frac{(X-Um_d)\epsilon_k^2 + [(X-Um_d)Um_f + V'^2]Um_f}{\sqrt{(X-Um_d)^2\epsilon_k^2 + [(X-Um_d)Um_f + V'^2]^2}}\right\}$$
$$\times\{\epsilon_k^2 + U^2 m_f^2 + (X-Um_d)^2 + 2V'^2 + 2\sqrt{(X-Um_d)^2\epsilon_k^2 + [(X-Um_d)Um_f + V'^2]^2}\}^{-1/2} \ , \tag{29}$$

$$m_d = -\frac{1}{N}\sum_{k>0}\left\{Um_f + \frac{(X-Um_d)[(X-Um_d)Um_f + V'^2]}{\sqrt{(X-Um_d)^2\epsilon_k^2 + [(X-Um_d)Um_f + V'^2]^2}}\right\}$$
$$\times\{\epsilon_k^2 + U^2 m_f^2 + (X-Um_d)^2 + 2V'^2 + 2\sqrt{(X-Um_d)^2\epsilon_k^2 + [(X-Um_d)Um_f + V'^2]^2}\}^{-1/2} \ , \tag{30}$$

$$\Delta = -\frac{V'}{N}\sum_{k>0}\left\{1 + \frac{[(X-Um_d)Um_f + V'^2]}{\sqrt{(X-Um_d)^2\epsilon_k^2 + [(X-Um_d)Um_f + V'^2]^2}}\right\}$$
$$\times\{\epsilon_k^2 + U^2 m_f^2 + (X-Um_d)^2 + 2V'^2 + 2\sqrt{(X-Um_d)^2\epsilon_k^2 + [(X-Um_d)Um_F + V'^2]^2}\}^{-1/2} \ , \tag{31}$$

$$X = \sqrt{2vK}\,(1-\delta)m_0 = 2v(1-\delta)^2 m_f \ . \tag{32}$$

Here $m_0 \neq 0$ is treated as a variational parameter and the optimum value of it is proportional to m_f, as is shown in Eq.

(32). The physical phonon-staggered ordering parameter m_p, which can be measured in experiments, is defined as

$$m_p=-\frac{1}{N}\sum_i(-1)^i\langle q_i\rangle\ . \tag{33}$$

After the two unitary transformations [Eqs. (3) and (5)] and making an average over the squeezed-phonon state $|\mathrm{ph},s\rangle$ we have

$$m_p=m_0+\sqrt{2v/K}\,\delta m_f=\sqrt{2v/K}\,m_f\ , \tag{34}$$

where we have used Eq. (32). Obviously $m_p\geq m_0$, that is, it is included in m_p the contribution of the dynamical distortion of phonon modes associated with the f-electron motion apart from the static staggered ordering parameter m_0.

First of all, let us see the case where $\epsilon_f=v=0$, that is, no electron-phonon coupling. By choosing these special input parameters our model system (1) is the spinless PAL model in symmetric case,

$$E_g/N=\frac{U}{4}-Um_dm_f+U\Delta^2-\frac{1}{N}\sum_{k>0}\{\epsilon_k^2+U^2m_f^2+U^2m_d^2+2V'^2+2\sqrt{U^2m_d^2\epsilon_k^2+[-U^2m_dm_f+V'^2]^2}\}^{1/2}\ , \tag{35}$$

$$m_f=\frac{1}{N}\sum_{k>0}\left\{-Um_d+\frac{-Um_d\epsilon_k^2+[-U^2m_dm_f+V'^2]Um_f}{\sqrt{U^2m_d^2\epsilon_k^2+[-U^2m_dm_f+V'^2]^2}}\right\}$$
$$\times\{\epsilon_k^2+U^2m_f^2+U^2m_d^2+2V'^2+2\sqrt{U^2m_d^2\epsilon_k^2+[-U^2m_dm_f+V'^2]^2}\}^{-1/2}\ , \tag{36}$$

$$m_d=-\frac{1}{N}\sum_{k>0}\left\{Um_f+\frac{-Um_d[-U^2m_dm_f+V'^2]}{\sqrt{U^2m_d^2\epsilon_k^2+[-U^2m_dm_f+V'^2]^2}}\right\}$$
$$\times\{\epsilon_k^2+U^2m_f^2+U^2m_d^2+2V'^2+2\sqrt{U^2m_d^2\epsilon_k^2+[-U^2m_dm_f+V'^2]^2}\}^{-1/2}\ , \tag{37}$$

$$\Delta=-\frac{V'}{N}\sum_{k>0}\left\{1+\frac{[-U^2m_dm_f+V'^2]}{\sqrt{U^2m_d^2\epsilon_k^2+[-U^2m_dm_f+V'^2]^2}}\right\}$$
$$\times\{\epsilon_k^2+U^2m_f^2+U^2m_d^2+2V'^2+2\sqrt{U^2m_d^2\epsilon_k^2+[-U^2m_dm_f+V'^2]^2}\}^{-1/2}\ , \tag{38}$$

with $V'=V-U\Delta$. The solutions of these equations can be summarized by the solid line in the $V\sim U$ phase diagram of Fig. 8. When $V=0$ our model is the spinless Falicov-Kimball model and a VDW phase exists in the ground state of the symmetric case for all values of $U>0$, as was shown by some previous authors.[11–13] However, when $V>0$ is a finite quantity, a disordered (uniform) phase appears for smaller U. We can see from the phase diagram (Fig. 8) that when $V>V_c=0.256t$ the VDW phase disappears for any values of U.

The effect of the electron-phonon interaction on the VDW ordering phase is twofold. First, one can infer from the famous Peierls' theorem[15] that the adiabatic electron-phonon coupling favors the VDW ordering. We show this in Fig. 8 by the short-dashed ($v=0.04t$), dash-dotted ($v=0.12t$), and long-dashed ($v=0.2t$) line with nonzero coupling $v>0$ but vanished phonon frequency $\omega=0$. Obviously, the size of the VDW ordering region increases with increasing coupling v. We note that in the

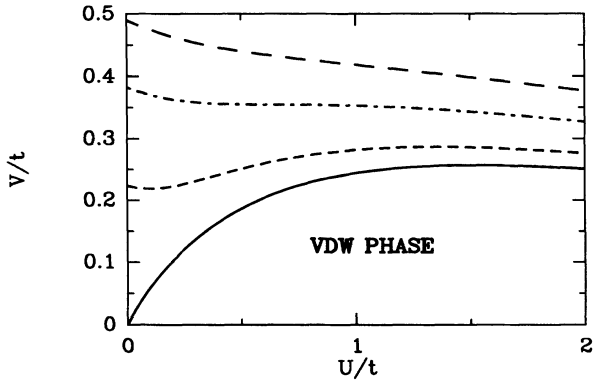

FIG. 8. Phase boundary in the symmetric case. $\omega=0$, $v=0$ (solid line), $0.04t$ (short-dashed line), $0.12t$ (dash-dotted line), $0.2t$ (long-dashed line). The phase transition is second order across the lines. The region under the lines is structural, but that above corresponds to uniform phase.

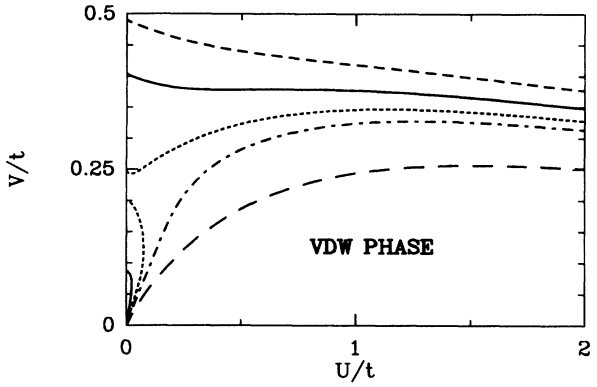

FIG. 9. Phase boundary in the symmetric case. $v=0.2t$, $\omega=0$ (short-dashed line), $0.1t$ (solid line), $0.2t$ (dotted line), $0.3t$ (dash-dotted line). The long-dashed line corresponds to the case $v=0$, which is included here for reference. Note that when $U/t \ll 1$ a small isolated disordering region exists between $V=0$ and $V\sim\omega$ for $\omega=0.1t$ and $0.2t$ in this figure.

adiabatic case ($\omega=0$), as long as $v>0$, an ordering state exists for small but finite hybridization $V>0$ even if the correlation $U=0$. This is different from the $v=0$ case where the ordering state cannot exist for $U=0$.

Second, the nonadiabaticity of the electron-phonon interaction should lead to instability of the VDW ordering state. We show the $V\sim U$ phase diagram for different $\omega\geq 0$ values in Fig. 9. One can see that with increasing ω, the VDW ordering region shrinks but the disordering (uniform phase) region expands. We note that when $U/t \ll 1$ a small isolated disordering region develops between $V=0$ and $V\sim\omega$ for $0<\omega\leq v$. For $\omega>v$, no ordering state can exist for $U=0$ and the phase boundary becomes similar to the case $v=0$.

Figure 10 shows the dimerization parameter m_p as a function of U in the case of $V=0.2t$, $v=0.2t$, and $\omega=0$, $0.1t$, $0.2t$, $0.3t$, respectively. Increasing ω significantly reduces the ordering (order parameter m_p), particularly for small U, eventually leading to its complete destruction. An order-disorder transition appears with increasing U. For $\omega=0.3t$, this takes place around $U=0.235t$.

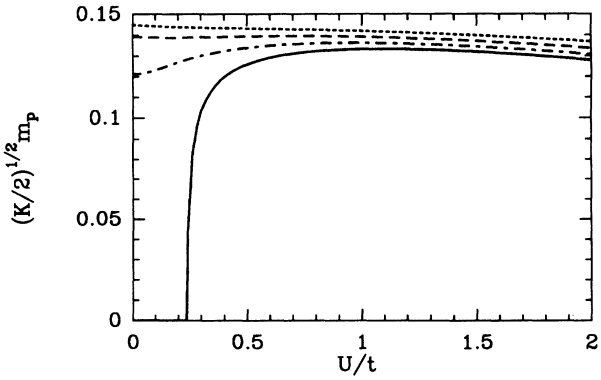

FIG. 10. The dimerization parameter m_p vs U relations in the case of $V=0.2t$, $v=0.2t$, and $\omega=0$ (dotted line), $0.1t$ (dashed line), $0.2t$ (dash-dotted line), and $0.3t$ (solid line).

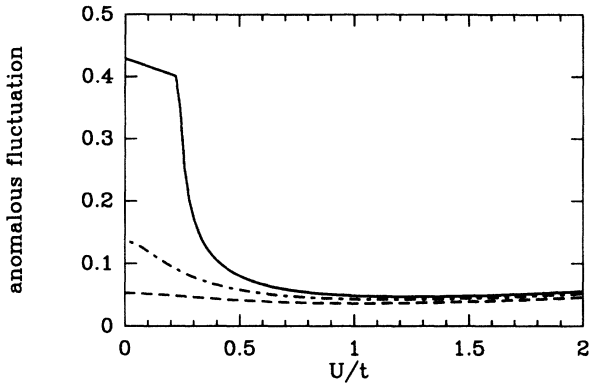

FIG. 11. Anomalous fluctuation f as functions of U in the case of $V=0.2t$, $v=0.2t$, and $\omega=0.1$ (dashed line), $0.2t$ (dash-dotted line), and $0.3t$ (solid line).

In symmetric case the dimensionless anomalous fluctuation of phonon modes associated with the motion of f electrons is

$$f=(\langle\Delta q_i^2\rangle-\langle\Delta q_i^2\rangle_0)/\langle\Delta q_i^2\rangle_0$$
$$=(\tau_m^{-2}-1)+4\frac{g^2}{\omega^2}\delta_m^2(\tfrac{1}{4}-m_f^2)\ . \tag{39}$$

We show in Fig. 11 the anomalous fluctuation f as a function of U in the case of $V=0.2t$, $v=0.2t$, and $\omega=0.1t$, $0.2t$, $0.3t$, respectively. Here one can see that the anomalous fluctuation is larger for smaller U. Besides, the value of the anomalous fluctuation is small for the VDW ordering state. This means that the ordering suppresses the quantum-lattice fluctuations.

V. CONCLUSION

In this paper we have developed a variational approach to treat the nonadiabaticity, that is, the quantum-lattice fluctuations, of the electron-phonon interaction in the one-dimensional half-filled spinless PAL model. The nonadiabaticity due to finite phonon frequency is treated through a variational polaronic-type wave function, in which two variational parameters δ and τ^2 are used to take into account the dynamical distortion and the squeezing effect of phonon modes. In this way we have found that the quantum-lattice fluctuation gradually smooths the valence transitions when the polaronic level ϵ_f-v changes. We have shown that conditions somewhat different from those of Hewson and Newns[4] should be satisfied for the occurrence of a significant reduction of the effective hybridization in the one-dimensional spinless PAL model. The effect of the Coulomb repulsion U is to suppress the quantum-lattice fluctuations, that is, to suppress the reduction of the FV frequency and the relaxation shift. This conclusion is different from that of Hanke and Schmeltzer.[5]

Our conclusion about the effect of U is obtained after the Hartree-Fock approximation, Eq. (11), is used, but that of Hanke and Schmeltzer[5] is based on the renormalization-group method. However, we believe

that our results reflect the effect of U correctly even if U is not small. This can be shown more clearly by the following qualitative argument. The reduction of the FV frequency and the finite relaxation shift result from the dynamical distortion of phonon modes (the quantum-lattice fluctuations). But a finite U favors the single occupation of each site ($N_e=N$ in our treatment), which in turn obstructs the hopping of electrons on and off the localized f level and suppress the related dynamical distortion of the phonon mode. Thus, the reduction of the FV frequency and the relaxation shift decreases with increasing U, as was shown by our numerical results. As our opinion, the difference between our results and those of Hanke and Schmeltzer[5] comes from that they fix the bare f level by $\epsilon_f=0$ and thus a nonzero polaronic level $-v$, when v is not small, may suppress the reduction of the FV frequency and the relaxation shift.

We have also discussed the symmetric case where a structural phase, the so-called VDW ordering, can occur. Our results show that the quantum-lattice fluctuations disfavor the VDW ordering and the lattice dimerization parameter m_p decreases with increasing ω. We have pointed out the possibility of an order-disorder transition in such systems. Besides, we show that VDW ordering makes the anomalous fluctuation of the phonon mode small.

This work can be extended easily to the two- and three-dimensional and to the spin-$\frac{1}{2}$ cases.

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