Quantum lattice fluctuations in the one-dimensional molecular-crystal model with Coulomb repulsion

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The effect of quantum lattice fluctuations on the ground state of a half-filled-band one-dimensional molecular-crystal model with on-site (U) and nearest-neighbor (V) Coulomb repulsion is investigated. The nonadiabatic effects due to finite phonon frequency ω are treated through a variational polaron wave function. The electronic correlations are decoupled by a Hartree-Fock approximation. Our variational approach gives a rather good description of the continuous variation of the dimerization, which is in good agreement with that of the Monte Carlo simulations, as functions of U, V, and ω. However, the Hartree-Fock theory predicts a fictitious long-range spin-density-wave order and a discontinuous transition between it and the charge-density-wave (CDW) state. These are somewhat different from the results of numerical simulations. Furthermore, we propose a valence-bonding (VB) state for describing the short-range antiferromagnetic correlation for intermediate values of U and V and determine the transition point between the VB state and the CDW state, which is in good agreement with that of the numerical simulations. The effect of quantum lattice fluctuations on the transition point is also discussed.

I. INTRODUCTION

In recent years, a great deal of theoretical effort has been devoted to understanding the competing instabilities in the one-dimensional many-electron and phonon coupling systems. Both in the adiabatic limit (where the phonon frequency ω equals zero) and the nonadiabatic limit (large-ω limit) the situation becomes clear, and a coherent picture has emerged.[1-4] However, the theoretical analysis becomes much more difficult in the presence of retardation (finite phonon frequency), and a few attempts have been made to evaluate the correction to the adiabatic scheme when the electronic energy is small enough so that quantum lattice fluctuations may play an important role.[4-9]

In a recent paper[10] (hereafter referred to as I) we studied the effect of quantum lattice fluctuations in the one-dimensional molecular-crystal model in the abscence of Coulomb repulsion by means of a variational approach, in which the nonadiabatic effects due to the finite phonon frequency ω are treated through a variational polaron wave function. In the treatment a part of the lattice distortion is carried over by the electrons and interferes with the static (frozen) lattice deformation. This leads to a weakening of the effective (adiabatic) potential stabilizing the dimerized state. Moreover, in the finite-ω case we showed that the lattice fluctuations are "squeezed" by the electron band motion, and the phonon subsystem is in the so-called two-phonon coherent state. Our results are in good agreement with both Monte Carlo simulations and renormalization-group analysis.

In this paper, we extend our previous variational treatment to the molecular-crystal model in the presence of on-site (U) and nearest-neighbor (V) Coulomb repulsion in the half-filled-band sector. The Hartree-Fock approximation will be used to decouple the electronic correlations, and besides the long-range charge-density-wave (CDW) order there may be another type of long-range order—spin-density-wave (SDW) order—in this approximation.[4,11,12] Furthermore, as it was proved exactly that the one-dimensional Hubbard model cannot develop the long-range SDW order,[13] we will propose a valence-bonding state for describing the short-range antiferromagnetic correlation for intermediate values of U and V, and then discuss the phase transition between the long-range CDW ordering state and the algebraic SDW ordering one.

The model considered in this paper was studied by Hirsch in a previous paper[4] by means of the Monte Carlo simulations on finite-size lattices. His main results are as follows: (1) the transition between CDW and SDW regions deviates slightly from $U=U_c=2V+\lambda^2/K$ (λ denotes electron-phonon coupling, K denotes the string constant) towards smaller U for intermediate correlation; (2) the phonon frequency plays a relatively minor role, and its main effect is to reduce the size of both CDW and SDW correlations, with respect to the $\omega=0$ limit; (3) the transition between CDW and SDW phases becomes discontinuous for large values of U. In this paper, we will compare our results with his.

In a recent paper Nasu[14] studied the same model as ours through a variational approach close to ours. But,

as was said in I, he decoupled the polaron effect and the frozen dimerization, not allowing for any interference between the two effects. We believe that this does not permit us to correctly treat the effect of quantum lattice fluctuations and makes it impossible to reproduce the numerical results of the Monte Carlo simulations.

II. THEORETICAL ANALYSIS

We start from the one-dimensional extended Hubbard model with on-site electron-phonon coupling in which the local electron density couples to the intramolecular vibration mode,

$$H=\sum_i\left[\frac{1}{2M}p_i^2+\frac{K}{2}q_i^2\right]-t\sum_{i,\sigma}(c_{i\sigma}^\dagger c_{i+1\sigma}+c_{i+1\sigma}^\dagger c_{i\sigma})-\lambda\sum_i q_i(n_i-1)+\sum_i Un_{i\uparrow}n_{i\downarrow}+\sum_i Vn_in_{i+1}\ , \tag{1}$$

where $n_{i\sigma}=c_{i\sigma}^\dagger c_{i\sigma}$ and $n_i=n_{i\uparrow}+n_{i\downarrow}$. The other notations are as usual. We restrict ourselves to the half-filled-band sector. One can perform an ordinary coordinate and momentum transformation on operators q_i and p_i:

$$p_i=i\left[\frac{\hbar\omega M}{2}\right]^{1/2}(b_i^\dagger-b_i),\quad q_i=\left[\frac{\hbar}{2M\omega}\right]^{1/2}(b_i^\dagger+b_i)\ , \tag{2}$$

where $\omega=\sqrt{K/M}$. Thus H becomes

$$H=\hbar\omega N/2+\sum_i\hbar\omega b_i^\dagger b_i-\lambda\left[\frac{\hbar}{2M\omega}\right]^{1/2}\sum_i(b_i^\dagger+b_i)(n_i-1)-t\sum_{i,\sigma}(c_{i\sigma}^\dagger c_{i+1\sigma}+c_{i+1\sigma}^\dagger c_{i\sigma})+\sum_i Un_{i\uparrow}n_{i\downarrow}+\sum_i Vn_in_{i+1}\ . \tag{3}$$

In the following, three unitary transformations, similar to those in I, will be used to introduce three variational parameters. The first one is a coherent-state transformation for every phonon mode,

$$S_1=-\sum_i(-1)^i m_0\left[\frac{M\omega}{2\hbar}\right]^{1/2}(b_i^\dagger-b_i)\ , \tag{4}$$

$$\begin{aligned}H_1&=\exp(S_1)H\exp(-S_1)\\&=\hbar\omega N/2+\sum_i\hbar\omega b_i^\dagger b_i+Km_0^2N/2+\sum_i(-1)^i m_0\hbar\omega\left[\frac{M\omega}{2\hbar}\right]^{1/2}(b_i^\dagger+b_i)-\lambda\sum_i\frac{\hbar}{2M\omega}(b_i^\dagger+b_i)(n_i-1)\\&\quad-\sum_i(-1)^i\lambda m_0 n_i-t\sum_{i,\sigma}(c_{i\sigma}^\dagger c_{i+1\sigma}+c_{i+1\sigma}^\dagger c_{i\sigma})+\sum_i Un_{i\uparrow}n_{i\downarrow}+\sum_i Vn_in_{i+1}\ ,\end{aligned} \tag{5}$$

where m_0 is the first variational parameter that measures the phonon-staggered ordering. After the transformation, H_1 represents a new electron-phonon coupling system in which the equilibrium position of the phonon mode q_i is $-(-1)^i m_0-\lambda/K$, $-(-1)^i m_0$, and $-(-1)^i m_0+\lambda/K$, when $n_i=0$, 1, and 2, respectively. In other words, the polaron effect, which will be taken into account by the following two transformations, is referred to the phonon-staggered ordering state, not to the original disordered state, so long as $m_0\neq 0$.

The second one is a modified Lang-Firsov-type transformation[15] that introduces into consideration the well-known polaron effect,

$$S_2=-\sum_i g\delta(b_i^\dagger-b_i)(n_i-1)\ , \tag{6}$$

where we have defined

$$g^2=\varepsilon_p/\hbar\omega,\quad \varepsilon_p=\lambda^2/2K\ , \tag{7}$$

ε_p is the polaron binding energy. As we said in I, it is esential here to associate the effective polaronic distortion, measured by δ, to the density fluctuation n_i-1, but not to n_i. Actually, the homogeneous part of the charge density $\sum_i n_i=N$ is coupled to the homogeneous deformation, which carries a "full" polaron effect independent of δ and decoupled from the following transformed Hamiltonian. This is one of the differences between our treatment and that of Ref. 14.

$$H_2=\exp(S_2)H_1\exp(-S_2)=\hbar\omega N/2+\sum_i\hbar\omega b_i^\dagger b_i+Km_0^2N/2-\sum_i(-1)^i(1-\delta)\lambda m_0 n_i$$
$$+\sum_i(-1)^i m_0\hbar\omega\left[\frac{M\omega}{2\hbar}\right]^{1/2}(b_i^\dagger+b_i)-\sum_i\lambda\left[\frac{\hbar}{2M\omega}\right]^{1/2}(1-\delta)(b_i^\dagger+b_i)(n_i-1)$$
$$-t\sum_{i,\sigma}(c_{i\sigma}^\dagger c_{i+1\sigma}\exp\{-g\delta[(b_i^\dagger-b_i)-(b_{i+1}^\dagger-b_{i+1})]\}+\text{H.c.})$$
$$+[U-(2\delta-\delta^2)\lambda^2/K]\sum_i n_{i\uparrow}n_{i\downarrow}+\sum_i Vn_in_{i+1}\ . \tag{8}$$

δ is the second dimensionless variational parameter, and it measures the degree of the polaron effect. When $\delta=0$, there is no polaron effect. When $\delta=1$, we should have $m_0=0$ in Eq. (8), and the polaron effect suppresses the frozen phonon-staggered ordering. When $0<\delta<1$, we expect that there would be a nonzero frozen phonon-staggered ordering but reduced by the polaron effect. Of course, the practical value of δ should be determined by the variational approach.[10]

The third transformation is the so-called "squeezing" one that is used to partly offset the polaronic narrowing effect on the bandwidth, as can be seen from the second to the last term of Eq. (8), and to increase the possibility of the polaron tunneling.

$$S_3=-\sum_i\alpha(b_i^\dagger b_i^\dagger-b_ib_i)\ , \tag{9}$$

$$H_3=\exp(S_3)H_2\exp(-S_3)=\hbar\omega N/2+\sum_i\hbar\omega(b_i^\dagger\cosh2\alpha+b_i\sinh2\alpha)(b_i^\dagger\sinh2\alpha+b_i\cosh2\alpha)$$
$$+Km_0^2N/2-\sum_i(-1)^i(1-\delta)\lambda m_0n_i-\sum_i\lambda\left[\frac{\hbar}{2M\omega}\right]^{1/2}(b_i^\dagger+b_i)(n_i-1)e^{2\alpha}$$
$$+\sum_i(-1)^i m_0\hbar\omega\left[\frac{M\omega}{2\hbar}\right]^{1/2}(b_i^\dagger+b_i)e^{2\alpha}$$
$$-t\sum_{i,\sigma}(c_{i\sigma}^\dagger c_{i+1\sigma}\exp\{-g\delta[(b_i^\dagger-b_i)-(b_{i+1}^\dagger-b_{i+1})]e^{-2\alpha}\}+\text{H.c.})$$
$$+[U-(2\delta-\delta^2)\lambda^2/K]\sum_i n_{i\uparrow}n_{i\downarrow}+\sum_i Vn_in_{i+1}\ , \tag{10}$$

where α is the third variational parameter. This transformation generates a new type of coherent lattice state, the two-phonon state.[16] When $\alpha\to\infty$ ($e^{-2\alpha}\to0$) we can see that there would be no polaronic narrowing effect on the bandwidth. On the other hand, if $\alpha\to\infty$, the zero-point energy of phonons goes to infinity. So we should choose a proper value for α.

Now we can average H_3 over the vacuum state $|0\rangle$ of the transformed phonon subsystem, and thus the electron and the phonon subsystems are decoupled and we obtain an effective Hamiltonian for the electron subsystem,

$$H_{\text{eff}}=\langle0|H_3|0\rangle=\hbar\omega(\tau^2+1/\tau^2)N/4+Km_0^2N/2-\sum_i(-1)^i(1-\delta)\lambda m_0n_i-\rho t\sum_{i,\sigma}(c_{i\sigma}^\dagger c_{i+1\sigma}+c_{i+1\sigma}^\dagger c_{i\sigma})$$
$$+[U-(2\delta-\delta^2)\lambda^2/K]\sum_i n_{i\uparrow}n_{i\downarrow}+\sum_i Vn_in_{i+1}\ , \tag{11}$$

where

$$\tau=\exp(-2\alpha)\quad\text{and}\quad\rho=\exp\left[-\frac{\lambda^2}{2K\hbar\omega}\delta^2\tau^2\right]\ . \tag{12}$$

We will use the Hartree-Fock approximation[11,12]

$$n_{i\uparrow}n_{i\downarrow}\simeq n_{i\uparrow}\langle n_{i\downarrow}\rangle+\langle n_{i\uparrow}\rangle n_{i\downarrow}-\langle n_{i\uparrow}\rangle\langle n_{i\downarrow}\rangle\ ,$$

$$(n_{i\uparrow}+n_{i\downarrow})(n_{i+1\uparrow}+n_{i+1\downarrow})\simeq(n_{i\uparrow}+n_{i\downarrow})\langle n_{i+1\uparrow}+n_{i+1\downarrow}\rangle+\langle n_{i\uparrow}+n_{i\downarrow}\rangle(n_{i+1\uparrow}+n_{i+1\downarrow})-\langle n_{i\uparrow}+n_{i\downarrow}\rangle\langle n_{i+1\uparrow}+n_{i+1\downarrow}\rangle\ ,$$

and set

$$\langle n_{i\sigma}\rangle=\tfrac{1}{2}[1+(-1)^i\sigma S+(-1)^i m_e]\ , \tag{13}$$

where $\sigma=+1$ for spin up and -1 for spin down. S and m_e are the long-range SDW and CDW ordering parameters, respectively,

$$S=\frac{1}{N}\sum_i(-1)^i(\langle n_{i\uparrow}\rangle-\langle n_{i\downarrow}\rangle)\ , \tag{14}$$

$$m_e=\frac{1}{N}\sum_{i,\sigma}(-1)^i\langle n_{i\sigma}\rangle\ . \tag{15}$$

We are in the half-filled case, so that

$$1=\frac{1}{N}\sum_{i,\sigma}\langle n_{i\sigma}\rangle\ . \tag{16}$$

In the Hartree-Fock approximation $H_{\rm eff}$ reads as follows:

$$H_{\rm eff}\simeq\hbar\omega(\tau^2+1/\tau^2)N/4+Km_0^2N/2+(V-\tfrac{1}{4}U_e)m_e^2N+VN+\tfrac{1}{4}U_eN+\tfrac{1}{4}U_eS^2N-\rho t\sum_{i,\sigma}(c_{i\sigma}^\dagger c_{i+1\sigma}+c_{i+1\sigma}^\dagger c_{i\sigma})$$
$$-\sum_i(-1)^i[(1-\delta)\lambda m_0+(2V-\tfrac{1}{2}U_e)m_e+\tfrac{1}{2}U_eS]n_{i\uparrow}-\sum_i(-1)^i[(1-\delta)\lambda m_0+(2V-\tfrac{1}{2}U_e)m_e-\tfrac{1}{2}U_eS]n_{i\downarrow}\ , \tag{17}$$

where

$$U_e=U-(2\delta-\delta^2)\lambda^2/K\ . \tag{18}$$

This Hartree-Fock Hamiltonian can be solved easily. The ground-state energy is

$$E_g(\delta,\tau^2)/N=\hbar\omega(\tau^2+1/\tau^2)/4+Km_0^2/2+V+\tfrac{1}{4}U_e+(V-\tfrac{1}{4}U_e)m_e^2+\tfrac{1}{4}U_eS^2$$
$$-\frac{1}{N}\sum_{k>0}\{4\rho^2t^2\cos^2k+[(1-\delta)\lambda m_0+(2V-\tfrac{1}{2}U_e)m_e+\tfrac{1}{2}U_eS]^2\}^{1/2}$$
$$-\frac{1}{N}\sum_{k>0}\{4\rho^2t^2\cos^2k+[(1-\delta)\lambda m_0+(2V-\tfrac{1}{2}U_e)m_e-\tfrac{1}{2}U_eS]^2\}^{1/2}\ , \tag{19}$$

in which the variational parameter m_0 is determined self-consistently by

$$Km_0=\frac{(1-\delta)}{N}\sum_{k>0}\frac{(1-\delta)\lambda m_0+(2V-\frac{1}{2}U_e)m_e+\frac{1}{2}U_eS}{\{4\rho^2t^2\cos^2k+[(1-\delta)\lambda m_0+(2V-\frac{1}{2}U_e)m_e+\frac{1}{2}U_eS]^2\}^{1/2}}$$
$$+\frac{(1-\delta)}{N}\sum_{k>0}\frac{(1-\delta)\lambda m_0+(2V-\frac{1}{2}U_e)m_e-\frac{1}{2}U_eS}{\{4\rho^2t^2\cos^2k+[(1-\delta)\lambda m_0+(2V-\frac{1}{2}U_e)m_e-\frac{1}{2}U_eS]^2\}^{1/2}}\ . \tag{20}$$

The equations to determine m_e and S, Eqs. (14) and (15), are

$$m_e=\frac{1}{N}\sum_{k>0}\frac{(1-\delta)\lambda m_0+(2V-\frac{1}{2}U_e)m_e+\frac{1}{2}U_eS}{\{4\rho^2t^2\cos^2k+[(1-\delta)\lambda m_0+(2V-\frac{1}{2}U_e)m_e+\frac{1}{2}U_eS]^2\}^{1/2}}$$
$$+\frac{1}{N}\sum_{k>0}\frac{(1-\delta)\lambda m_0+(2V-\frac{1}{2}U_e)m_e-\frac{1}{2}U_eS}{\{4\rho^2t^2\cos^2k+[(1-\delta)\lambda m_0+(2V-\frac{1}{2}U_e)m_e-\frac{1}{2}U_eS]^2\}^{1/2}}\ , \tag{21}$$

$$S=\frac{1}{N}\sum_{k>0}\frac{(1-\delta)\lambda m_0+(2V-\frac{1}{2}U_e)m_e+\frac{1}{2}U_eS}{\{4\rho^2t^2\cos^2k+[(1-\delta)\lambda m_0+(2V-\frac{1}{2}U_e)m_e+\frac{1}{2}U_eS]^2\}^{1/2}}$$
$$-\frac{1}{N}\sum_{k>0}\frac{(1-\delta)\lambda m_0+(2V-\frac{1}{2}U_e)m_e-\frac{1}{2}U_eS}{\{4\rho^2t^2\cos^2k+[(1-\delta)\lambda m_0+(2V-\frac{1}{2}U_e)m_e-\frac{1}{2}U_eS]^2\}^{1/2}}\ . \tag{22}$$

Compared with Eq. (21), Eq. (20) can be rewritten as

$$Km_0=(1-\delta)\lambda m_e \text{ or } m_0=(1-\delta)\frac{\lambda}{K}m_e\ . \tag{23}$$

Because of equations

$$Km_0^2/2+(V-\tfrac{1}{4}U_e)m_e^2=\tfrac{1}{2}[(1-\delta)^2\lambda^2/K+2V-\tfrac{1}{2}U_e]m_e^2=\tfrac{1}{2}J_em_e^2$$

and

$$(1-\delta)\lambda m_0+(2V-\tfrac{1}{2}U_e)m_e=[(1-\delta)^2\lambda^2/K+2V-\tfrac{1}{2}U_e]m_e=J_em_e\ , \tag{24}$$

where

$$J_e=(1-\delta)^2\lambda^2/K+2V-\tfrac{1}{2}U_e \ ,$$

we have, from Eqs. (19), (21), and (22),

$$E_g(\delta,\tau^2)N=\hbar\omega(\tau^2+1/\tau^2)/4+V+\tfrac{1}{4}U_e+\tfrac{1}{2}J_e m_e^2+\tfrac{1}{4}U_e S^2 - \frac{1}{N}\sum_{k>0}[4\rho^2t^2\cos^2k+(J_e m_e+\tfrac{1}{2}U_e S)^2]^{1/2}-\frac{1}{N}\sum_{k>0}[4\rho^2t^2\cos^2k+(J_e m_e-\tfrac{1}{2}U_e S)^2]^{1/2}, \quad (25)$$

$$m_e=\frac{1}{N}\sum_{k>0}\frac{J_e m_e+\frac{1}{2}U_e S}{[4\rho^2t^2\cos^2k+(J_e m_e+\frac{1}{2}U_e S)^2]^{1/2}}+\frac{1}{N}\sum_{k>0}\frac{J_e m_e-\frac{1}{2}U_e S}{[4\rho^2t^2\cos^2k+(J_e m_e-\frac{1}{2}U_e S)^2]^{1/2}} \ , \quad (26)$$

$$S=\frac{1}{N}\sum_{k>0}\frac{J_e m_e+\frac{1}{2}U_e S}{[4\rho^2t^2\cos^2k+(J_e m_e+\frac{1}{2}U_e S)^2]^{1/2}}-\frac{1}{N}\sum_{k>0}\frac{J_e m_e-\frac{1}{2}U_e S}{[4\rho^2t^2\cos^2k+(j_e m_e-\frac{1}{2}U_e S)^2]^{1/2}} \ . \quad (27)$$

As the ground-state energy, Eq. (25), can be rewritten as

$$E_g(\delta,\tau^2)N=\hbar\omega(\tau^2+1/\tau^2)/4+V+\tfrac{1}{4}U_e+(1/8J_e+1/4U_e)[(J_e m_e+\tfrac{1}{2}U_e S)^2+(J_e m_e-\tfrac{1}{2}U_e S)^2] +2(1/8J_e-1/4U_e)(J_e m_e+\tfrac{1}{2}U_e S)(J_e m_e-\tfrac{1}{2}U_e S)-\frac{1}{N}\sum_{k>0}\{[4\rho^2t^2\cos^2k+(J_e m_e+\tfrac{1}{2}U_e S)^2]^{1/2} +[4\rho^2t^2\cos^2k+(J_e m_e-\tfrac{1}{2}U_e S)^2]^{1/2}\} \ , \quad (28)$$

when $(1/8J_e-1/4U_e)>0$ we must let

$$J_e m_e+\tfrac{1}{2}U_e S=-(J_e m_e-\tfrac{1}{2}U_e S), \quad m_e=0 \quad (29)$$

for $E_g(\delta,\tau^2)/N$ to be minimized; but when $(1/8J_e-1/4U_e)<0$ we must let

$$J_e m_e+\tfrac{1}{2}U_e S=J_e m_e-\tfrac{1}{2}U_e S, \quad S=0 \ . \quad (30)$$

Thus, we know that the CDW and the SDW order could not coexist, and the transition between the two phases takes place at

$$U_e=2J_e, \quad U-\lambda^2/K=2V \ . \quad (31)$$

In the Hartree-Fock approximation a long-range SDW order results when $U-\lambda^2/K>2V$, but a long-range CDW order results when $U-\lambda^2/K<2V$. This fact is not influenced by quantum lattice fluctuations in the approximation.

III. RESULTS OF HARTREE-FOCK APPROXIMATION

Our model Hamiltonian (1) is defined by four parameters as far as ground-state properties are concerned, which we can take to be the phonon frequency ω, the polaron binding energy $\varepsilon_p=\lambda^2/2K$, the Coulomb repulsion U, and V. The transfer integral t can be set equal to 1 by redefining the overall energy scale.

Our variational procedure consists of two steps.[10] First, for every choice of the variational parameters δ and τ^2, Eq. (26) or (27) could be solved for m_e or S, and thus the corresponding ground-state energy $E_g(\delta,\tau^2)/N$ [Eq. (25)] could be determined. Second, one should search for optimum values of the variational parameters δ and τ^2, δ_m and τ_m^2, at which the ground-state energy $E_g(\delta,\tau^2)/N$ arrives at a stable minimum $E_g(\delta_m,\tau_m^2)/N$.

A. $\omega=0$ limit

In the $\omega=0$ limit we have found that $\delta_m=0$ and $\tau_m^2=1$ for any employed values of other parameters ε_p, U, and V. So in this limit we have the same solution as that of Chan and Heine.[11] A discontinuous transition takes place at $U-\lambda^2/K=2V$ between the CDW and SDW state.

$\delta_m=0$ is just as one might expect because the $\omega=0$ limit is the so-called adiabatic limit at which there should be no polaron effect.

B. $\omega=\infty$ limit

When $\omega=\infty$,

$$\rho=\exp(-\varepsilon_p\delta^2\tau^2/\hbar\omega)=1 \ ;$$

thus we can let $\tau_m^2=1$, that is, there is no "squeezing" effect in this limit. This leads to

$$E_g(\delta,1)/N-\tfrac{1}{2}\hbar\omega=V+\tfrac{1}{4}U_e+\tfrac{1}{2}J_e m_e^2 -\frac{2}{N}\sum_{k>0}(4t^2\cos^2k+J_e^2m_e^2)^{1/2} \ ,$$

$$1=\frac{2J_e}{N}\sum_{k>0}(4t^2\cos^2k+J_e^2m_e^2)^{-1/2} \ ,$$

when (32)

$$2V\geq U-\lambda^2/K \ ;$$

or

$$E_g(\delta,1)/N-\tfrac{1}{2}\hbar\omega=V+\tfrac{1}{4}U_e+\tfrac{1}{4}U_eS^2 -\frac{2}{N}\sum_{k>0}(4t^2\cos^2k+\tfrac{1}{4}U_e^2S^2)^{1/2}\ ,$$

$$1=\frac{U_e}{N}\sum_{k>0}(4t^2\cos^2k+\tfrac{1}{4}U_e^2S^2)^{-1/2}\ ,$$

when (33)

$$2V<U-\lambda^2/K\ .$$

By numerical calculations, we have found that the minimum of $E_g(\delta,1)/N$ is always achieved at $\delta_m=1$ no matter whether the other parameters ε_p, U, and V are large or small. If we substitute $\delta_m=1$ and $\tau_m^2=1$ into Eq. (11) we have the effective Hamiltonian

$$H_{\rm eff}=\hbar\omega N/2-t\sum_{i,\sigma}(c_{i\sigma}^{\dagger}c_{i+1\sigma}+c_{i+1\sigma}^{\dagger}c_{i\sigma}) +(U-\lambda^2/K)\sum_i n_{i\uparrow}n_{i\downarrow}+V\sum_i n_in_{i+1}\ ; \quad (34)$$

that is, in this limit the effective Hamiltonian becomes an extended Hubbard model. This is the same result as that of Hirsch,[4] who got it by means of the functional integration,[6] and discussed this limit in some detail.[4] For $V=0$, from the effective Hamiltonian (34) it is obvious that the transition, at $U=\lambda^2/K$, between the CDW state and SDW state, is continuous even if in the Hartree-Fock approximation.

C. General ω case

In our theory, m_0 is only a variational parameter, and we should define the phonon-staggered ordering parameter m_p, which can be measured by experiments or by Monte Carlo simulations, as follows:

$$m_p=\frac{1}{N}\sum_i(-1)^i\langle q_i\rangle\ , \quad (35)$$

where q_i is the position operator of the phonon mode and $\langle\ \rangle$ represents an average over the ground state of the total system. It is proved in I that

$$m_p=m_0+\frac{\lambda}{K}\delta m_e=\frac{\lambda}{K}m_e\ . \quad (36)$$

As $m_0=(\lambda/K)(1-\delta)m_e$, we have $m_p\geq m_0$; that is, we should include in m_p the contribution of the polaron effect apart from the static staggered ordering parameter m_0.

Figure 1 shows the dimerization parameter m_p and long-range SDW order parameter S as functions of U in the case of $\hbar\omega=0.1$ and $V=0$ or $V=U/2$ (we take the unit so that $t=1$, $k=0.25$, and $\varepsilon_p=0.81$). Figure 2 shows m_p and S as functions of U in the case of $\hbar\omega=1$ and $V=0$ or $V=U/2$. The qualitative behaviors of the curves in the two figures are the same: When $V=U/2$ there is always a long-range CDW order, but when $V=0$ there is a phase transition at $U=2\varepsilon_p=1.62$ between the CDW and the SDW state. We can see from the figures that when $\hbar\omega=0.1$ the transition is obviously discontinuous, but when $\hbar\omega=1$ the transition is quasicontinuous. That is to say that in the case of $V=0$ when the phonon frequency ω increases from 0 to ∞ the order of the transition between the CDW and SDW state changes from discontinuous to continuous in the Hartree-Fock approximation.

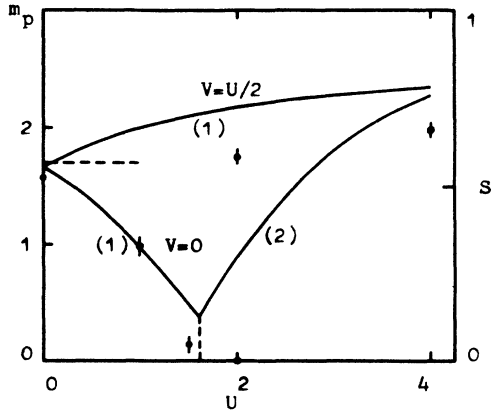

FIG. 1. (1) m_p and (2) S vs U relations, $\hbar\omega=0.1$, $V=0$, or $V=U/2$. The dashed line is the $\omega=0$ result for U, $V=0$. The filled circle with the line through it denotes Monte Carlo simulations (Ref. 4).

For comparison, in Figs. 1 and 2 we also show the results of Monte Carlo simulations.[4] We can see that our results are in good agreement with those of the numerical simulations, except the order of transitions in the case of $V=0$. Hirsch[4] predicted a continuous transition no matter whether the frequency ω is large or small. In addition, for $V=U/2$ the variational results, in agreement with Monte Carlo simulations,[4] indicate that the dimerization increases slowly with U, while the mean-field theory[4,11,12] (in which the effect of quantum lattice fluc-

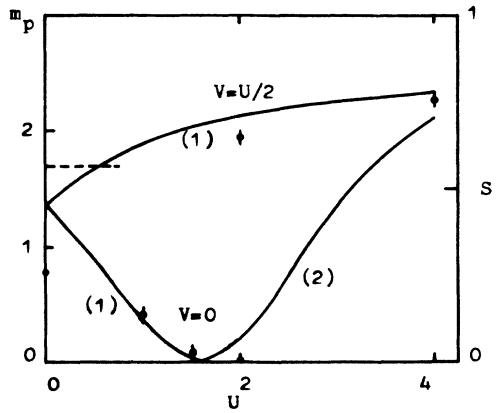

FIG. 2. (1) m_p and (2) S vs U relations, $\hbar\omega=1$, $V=0$, or $V=U/2$. The dashed line is the $\omega=0$ result for U, $V=0$. The filled circle with the line through it denotes Monte Carlo simulations (Ref. 4).

tuations are omitted) would predict no change in that case.

Figure 3 shows the optimum values of the variational parameters δ_m and τ_m^2 as functions of U in the case of $\hbar\omega=1$, $V=0$ or $V=U/2$. For $V=0$, when U increases δ_m increases, reaches a maximum at $U=1.62$, where the phase transition between the CDW and SDW state takes place, and then decreases. Besides, in the τ_m^2 versus U curve for the same case there is a minimum at the transition point. These facts indicate that at the phase transition point the polaron effect is strongest. For $V=U/2$, the CDW ordering dominates, and it can be seen from the figure that δ_m decreases and τ_m^2 increases monotonically with increasing U. We believe that it is the long-range order that reduces the polaron effect.[10]

Figure 4 shows the dimerization parameter m_p as functions of V in the cases of $\hbar\omega=1$ and $U=2,4,6$, and 8. For comparison the results of Monte Carlo simulations[4] are also shown. The phase trnsition for every case is discontinuous, and it takes place exactly at $V_c=\frac{1}{2}(U-\lambda^2/K)$ in our Hartree-Fock approximation. These are different from the results of numerical simulations that predict that the transition may be continuous for small values of U, and it can deviate slightly towards larger V for intermediate values of U. We will return to this problem and explain the meaning of the vertical dashed-dotted line in the figure in the next section.

Figure 5 shows the CDW ordering parameters m_e, δ_m, and τ_m^2 as functions of $\hbar\omega$ in the case of $U=4$ and $V=2$. Figure 6 shows the SDW ordering parameter S, δ_m, and τ_m^2 as functions of $\hbar\omega$ in the case of $U=4$ and $V=0$. From the two figures we can see that δ_m are always monotonically increasing functions of $\hbar\omega$ and in the large-ω limit $\delta_m\to 1$, as is discussed in Sec. III B. Because when ω increases from 0 to ∞ our model system goes from the adiabatic limit to the nonadiabatic limit, we can say that the nonadiabaticity in the system is connected with the well-known polaron effect.[15,17,18] In the two figures our variational treatment gives a good

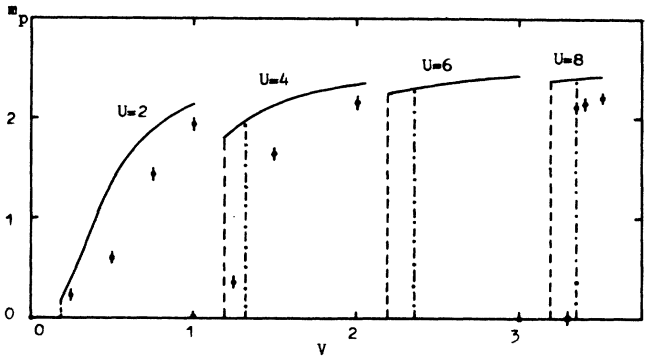

FIG. 4. m_p vs V relations for various U, $\hbar\omega=1$. The vertical dashed lines represent the transition points in the Hartree-Fock approximation. The filled circle with the line through it denotes Monte Carlo simulations (Ref. 4). See the text for further details.

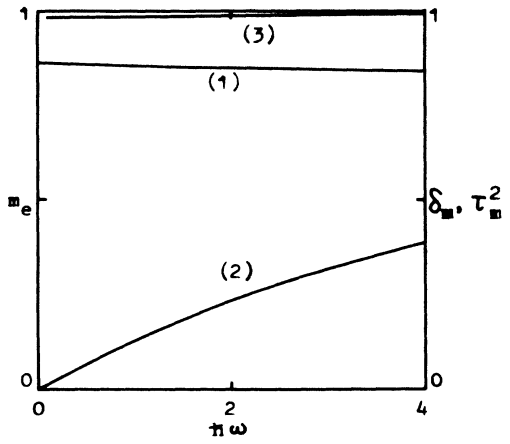

FIG. 5. (1) m_e, (2) δ_m, and (3) τ_m^2 vs $\hbar\omega$ relations, $U=4$ and $V=2$.

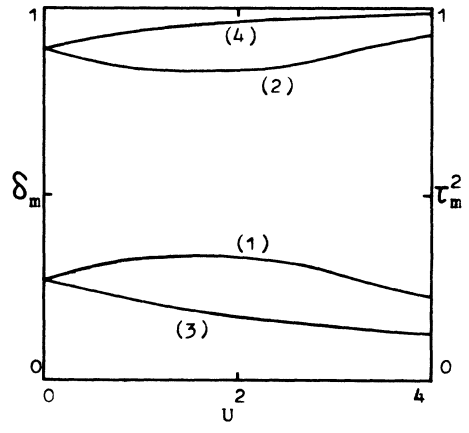

FIG. 3. [(1) and (3)] δ_m and [(2) and (4)] τ_m^2 vs U relations, $\hbar\omega=1$, [(1) and (2)] $V=0$, and [(3) and (4)] $V=U/2$.

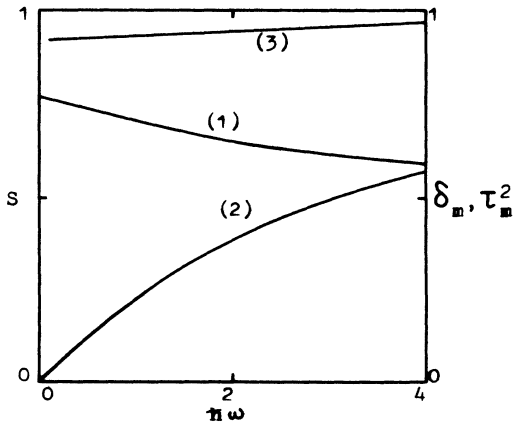

FIG. 6. (1) S, (2) δ_m, and (3) τ_m^3 vs $\hbar\omega$ relations, $U=4$ and $V=0$.

description for the continuous decrease of the long-range ordering parameter m_e and S with increasing $\hbar\omega$ between the adiabatic and the nonadiabatic limit. It is the polaron effect that reduces the long-range order, but it cannot be suppressed completely in our model system with nonzero U and V.

IV. FURTHER RESULTS FOR PHASE TRANSITION POINTS

In the Hartree-Fock approximation the phase transition between the CDW and SDW state occurs exactly at $U-\lambda^2/K=2V$, and in the region of $U-\lambda^2/K>2V$ it predicts a fictitious long-range SDW order. However, the results of Monte Carlo simulations[4] predict a transition between the long-range CDW ordering states and the short-range antiferromagnetic correlation state, which occurs at $V=V_c>\frac{1}{2}(U-\lambda^2/K)$ for intermediate values of U. In this section, we will propose a valence-bonding state for describing the short-range antiferromagnetic correlation for intermediate values of U and V, and then compare its energy with that of the CDW state to determine the phase transition point.

Let us come back to the effective Hamiltonian (11) and make the variational parameter m_0 equal to zero,

$$H_{\rm eff}=\hbar\omega(\tau^2+1/\tau^2)N/4-\rho t\sum_{i,\sigma}(c^\dagger_{i\sigma}c_{i+1\sigma}+c^\dagger_{i+1\sigma}c_{i\sigma}) +[U-(2\delta-\delta^2)\lambda^2/K]\sum_i n_{i\uparrow}n_{i\downarrow}+V\sum_i n_i n_{i+1} . \quad (37)$$

When $U-\lambda^2/K$ and V are larger than the hopping integral t, we can divide $H_{\rm eff}$ into two parts,

$$H_{\rm eff}=H_0+H_1 , \quad (38)$$

$$H_0=\sum_n\{-t\sum_\sigma(c^\dagger_{2n\sigma}c_{2n+1\sigma}+c^\dagger_{2n+1\sigma}c_{2n\sigma})+[U-(2\delta-\delta^2)\lambda^2/K](n_{2n\uparrow}n_{2n\downarrow}+n_{2n+1\uparrow}n_{2n+1\downarrow})+Vn_{2n}n_{2n+1}\} , \quad (39)$$

$$H_1=-t\sum_{n,\sigma}(c^\dagger_{2n-1\sigma}c_{2n\sigma}+c^\dagger_{2n\sigma}c_{2n-1\sigma}) , \quad (40)$$

and we will treat H_1 by perturbation theory. The ground state of H_0 in the half-filled-band case can be easily constructed,

$$|g\rangle=\prod_n\frac{1}{(1+v^2)^{1/2}}\left[\frac{1}{\sqrt{2}}(c^\dagger_{2n\uparrow}c^\dagger_{2n+1\downarrow}-c^\dagger_{2n\downarrow}c^\dagger_{2n+1\uparrow})+\frac{v}{\sqrt{2}}(c^\dagger_{2n\uparrow}c^\dagger_{2n\downarrow}+c^\dagger_{2n+1\uparrow}c^\dagger_{2n+1\downarrow})\right]|0\rangle , \quad (41)$$

where $|0\rangle$ is the vacuum state and

$$v=\left[\frac{[U-(2\delta-\delta^2)\lambda^2/K-V]^2}{16t^2}+1\right]^{1/2}-\frac{U-(2\delta-\delta^2)\lambda^2/K-V}{4t} . \quad (42)$$

The ground-state energy $E_g(\delta,\tau^2)$, which is the lowest eigenenergy of the zeroth-order Schrödinger equation

$$H_0|g\rangle=E_g|g\rangle , \quad (43)$$

and is related to the variational parameter δ and τ^2, is

$$E_g(\delta,\tau^2)/N=3V/4+\tfrac{1}{4}[U-(2\delta-\delta^2)\lambda^2/K] -\left[\frac{[U-(2\delta-\delta^2)\lambda^2/K-V]^2}{16}+t^2\right]^{1/2} . \quad (44)$$

The optimum values of δ and τ^2, δ_m and τ_m^2, and the stable minimum of $E_g(\delta,\tau^2)/N$, $E_g(\delta_m,\tau_m^2)/N$, can be determined by a similar manner as that illustrated at the beginning of Sec. III. In what follows, we take $E_g(\delta_m,\tau_m^2)/N$, which is thus derived, as a rough estimation for the ground-state energy of the short-range antiferromagnetic correlation state and then compare it with the ground-state energy of the long-range CDW ordering state to determine the phase transition point.

First of all, we consider the large-ω limit ($\omega=\infty$ limit), where the effective Hamiltonian (37) becomes that of Eq. (34). The transition line obtained from our work in this limit is shown in Fig. 7, where the dashed line denotes $U-\lambda^2/K=2V$. For comparison, the results of the Monte Carlo simulations[4] are also shown. It is evident that when $V>1$ and $U-\lambda^2/K>2$, that is, when H_1 in

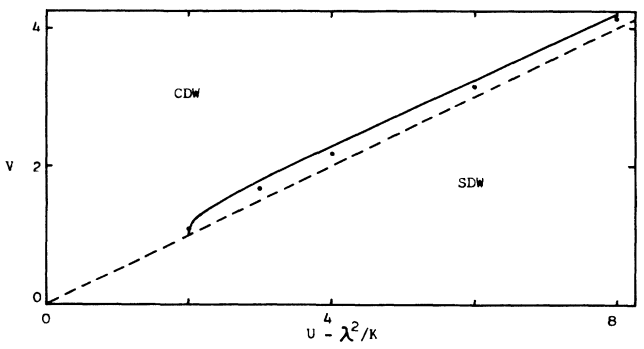

FIG. 7. Phase boundary between CDW and SDW regions in the $\omega=\infty$ limit. The dashed line denotes $U-\lambda^2/K=2V$. The solid line denotes the prediction of the valence-bonding state in Sec. IV. The closed circle denotes Monte Carlo simulations (Ref. 4).

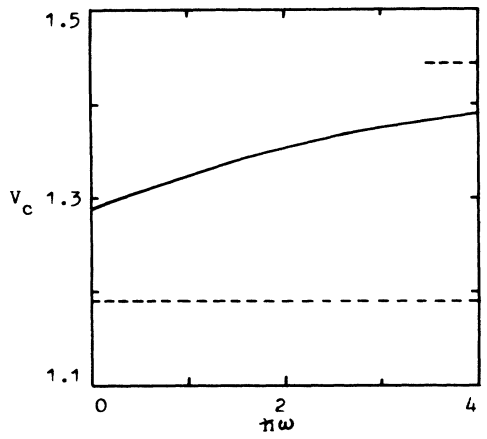

FIG. 8. Phase transition point V_c vs $\hbar\omega$ relation, $U=4$. The horizontal dashed line denotes $V_c=\frac{1}{2}(U-\lambda^2/K)=1.19$. The short-dashed line at the top-right corner is the $\omega=\infty$ limit: $V_c=1.4442$.

(40) could be treated as a perturbation, our results for the transition point are in good agreement with those of the numerical simulations.

Then we consider the general ω case. Figure 8 shows the transition point V_c as a function of $\hbar\omega$ in the case of $U=4$. The horizontal dashed line denotes

$$V_c=\tfrac{1}{2}(U-\lambda^2/K)=1.19\ ,$$

the result of the Hartree-Fock approximation. The short dashed line at the top-right corner of the figure is the $\omega=\infty$ limit: $V_c=1.4442$. We can see from the figure that V_c increases monotonically with increasing $\hbar\omega$, starting from $V_c=1.2884$ when $\omega=0$. We believe this is because the polaron effect, connected with the nonzero ω, is unfavorable to the long-range order.

We have also calculated the values of V_c in the cases of $\hbar\omega=1$ and $U=4$, 6, and 8, and the results are shown in Fig. 4 by the vertical dashed-dotted line. The result for $U=8$ is $V_c=3.3442$, which is in good agreement with that of Hirsch,[4] $V_c=3.32\pm0.05$.

V. CONCLUSION

We have continued our previous work in I to study the influence of quantum lattice fluctuations on the ground state of a half-filled-band one-dimensional molecular-crystal model with on-site and nearest-neighbor Coulomb repulsion. Our main results are summarized as follows.

(a) We have shown that our variational approach gives a rather good description, even if a Hartree-Fock approximation is used to decouple the Coulomb correlation, of the continuous variation of the dimerization as functions of the Coulomb repulsion U, V, and the frequency ω. We have compared our results with those of numerical simulations[4] and found that they are in good agreement. Our variational treatment might overestimate the long-range order because we start from a long-range ordering state with $m_0\neq0$, while the finite-size calculations would increase the disorder. We believe that these are the main factors that result in the deferences between our results and those of Ref. 4 when the long-range CDW order dominates.

(b) Our theory in the Hartree-Fock approximation always predicts a discontinuous transition between the CDW and the SDW state, exactly at $U-\lambda^2/K=2V$, except for $V=0$; in this case the order of the transition changes from discontinuous to continuous when ω increases from 0 to ∞.

(c) We have made a rough estimation for the ground-state energy of the short-range antiferromagnetic correlation state. By comparing it with the ground-state energy of the long-range CDW ordering state we have determined the phase transition point that is in good agreement with that of the numerical simulations for intermediate values of U and V. Furthermore, we have shown that the transition point V_c moves up (U and ε_p fixed) when the frequency ω increases, and this indicates that the polaron effect favors a disordered state.

In this paper, the superconducting correlations are totally disregarded because the parameter regime discussed is particularly unfavorable for them except for $V=0$; in this case we showed in I that in the $\omega=\infty$ limit if $U<\lambda^2/K$ the system is on the borderline between CDW and superconductivity.[4,6] If the system is a departure from the half-filled case the superconducting correlation might dominate.[4,19] This is a problem for further research.

ACKNOWLEDGMENTS

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