Effects of quantum lattice fluctuations on the charge-density wave of halogen-bridged mixed-valence transition-metal linear complexes

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The effects of quantum lattice fluctuations on the charge-density wave (CDW) of a one-dimensional one-band model for halogen-bridged mixed-valence linear chain complexes are studied by a functional integral approach. Equations for the phonon order parameter and the phonon excitations are derived within a one-loop approximation. They can be applied to any value of the ionic mass ranging from the $M = 0$ (antiadiabatic limit) to the $M = \infty$ (adiabatic) limit. We find that the dimerized CDW survives the quantum lattice fluctuations for arbitrarily small electron-lattice coupling. In the case of finite ionic mass, the low-energy properties of the system are governed by the limit of $M = 0$ for a weak electron-phonon interaction, whereas they are governed by the adiabatic limit ($M = \infty$) for a stronger electron-phonon interaction.

I. INTRODUCTION

In a one-dimensional system, the metallic state of electrons is unstable against the electron-phonon interaction, and it results in the Peierls transition.[1] A dimerized lattice will be formed at band half-filling. This is true for an arbitrarily small coupling between the electrons and the lattice in the mean-field adiabatic approximation, which treats the phonon degree of freedom classically. In the case of polyacetylene,[2] the archetype of conducting polymers, the ground state is a bond order wave (BOW). The elementary excitations on the BOW are solitons and polarons, which play an important role in the explanation of the peculiar properties of conducting polymers.[3] For halogen-bridged mixed-valence transition-metal linear complexes (HMMC or MX chains), the symmetry broken ground states exhibit many possibilities, such as BOW, the charge-density wave (CDW), the spin-density wave (SDW), and their competition and coexistence.[4,5] On these symmetry broken ground states, there exists a variety of localized excitations such as solitons, polarons, bipolarons, and excitons.[6–8] The contributions of these localized excitations to various observable effects are essential in the understanding of experimental results such as optical absorption and resonance Raman scattering.

Although the mean-field theory of the one-dimensional electron-phonon models has achieved great success, the quantum lattice fluctuations are believed to be important in the understanding of these linear and nonlinear optical properties.[9–13] A prime question is whether the symmetry broken ground state survives the quantum lattice fluctuations. For the Su-Schrieffer-Heeger (SSH) model,[2] it has been shown that the ground state is of a dimerized lattice for the spin degree of freedom $N \geq 2$.[14–17] Furthermore, by employing renormalization-group arguments, Fradkin and Hirsch[16] showed that the low-energy behavior of the system is governed by the $M = 0$ limit, i.e., by the Gross-Neveu model, and not by the adiabatic $M = \infty$ limit. However, in the one-dimensional one-band model for MX complexes, the symmetry broken ground state is a CDW state, not a BOW state, will the situation be different? It is the purpose of this paper to investigate the effects of quantum lattice fluctuations on the CDW of MX complexes. We find that the dimerized CDW survives the quantum lattice fluctuations for an arbitrarily small electron-lattice coupling as the BOW in the SSH model. For the case where the ionic mass is finite, it agrees with the renormalization-group analysis by Fradkin and Hirsch[16] that the low-energy properties of the system are governed by the $M = 0$ limit for a weak electron-phonon interaction. However, for a strong electron-phonon interaction, the quantum lattice fluctuation is suppressed and the system's behavior is governed by the adiabatic limit.

This paper is organized as follows. In Sec. II we describe the model Hamiltonian which we use in this work and derive an effective phonon action by integrating out the electronic variables. In Sec. III we studied the quantum effects on the phonon excitations of the MX model at the CDW state in a similar way by treating the σ- and π-modes of the Hubbard model.[18] In Sec. IV the equations for the phonon order parameter are derived within a one-loop approximation, and the dependences of quantum lattice fluctuations on the ionic masses and the electron-phonon couplings are discussed. In the last section, a brief summary is given.

II. THE EFFECTIVE ACTION

The one-dimensional one-band model for the MX complexes can be written as[19]

$$H = \sum_l \left[\frac{1}{2M} p_l^2 + K u_l^2 \right] - \sum_{l,s} t_0 (c_{l,s}^\dagger c_{l+1,s} + c_{l+1,s}^\dagger c_{l,s}) + \sum_{l,s} \alpha (u_l - u_{l+1}) c_{l,s}^\dagger c_{l,s}, \quad (1)$$

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where $c^{\dagger}_{l,s}$ and $c_{l,s}$ are the creation and annihilation operators of electrons sitting at the lth metal ion (M) with spin s, u_l (its conjugated momentum is p_l) is the displacement of the lth halogen ion (X), t_0 is the supertransfer integral between the nearest-neighbor metal ions, the coefficient α denotes the coupling strength parameter, and K is the elastic constant between the halogen ion and its neighboring metal ones. M is the mass of halogen ions. In this model, we have assumed the metal ions are much heavier than that of halogen ions, therefore the effects of quantum fluctuation are mainly induced by the halogen ions.

It should be noticed that the above Hamiltonian for MX complexes differs from the SSH model for polyacetylene by two points. In the SSH model, the displacement u_l modifies the transfer integral t_0. Such an interaction is called site-off-diagonal. In Eq. (1), on the other hand, u_l is coupled to the energies of electrons on the metal ions. The interaction is site-diagonal. Another is the bare phonon dispersion. The bare phonon excitations are correlated in the SSH model while they are independent in Eq. (1). These two points will be smeared out in their continuum versions,[20] but will they lead to a qualitatively different result on the quantum fluctuations?

To take into account the quantum lattice fluctuations, we employ a functional integral approach. The partition function for the Hamiltonian (1) can be written as a functional integral over both complex and Grassmann variables:

$$Z = \int \mathcal{D}[u]\mathcal{D}[\phi^*\phi]e^{-S}, \tag{2}$$

where the action S is

$$\begin{aligned} S = \int_0^\beta d\tau \Bigg\{ & \sum_l [K u_l^2 + \tfrac{1}{2} M (du_l/d\tau)^2] \\ & + \sum_{l,s} \phi^*_{l,s}(\partial_\tau - \mu)\phi_{l,s} \\ & - \sum_{l,s} t_0(\phi^*_{l,s}\phi_{l+1,s} + \phi^*_{l+1,s}\phi_{l,s}) \\ & - \sum_{l,s} \alpha(u_l - u_{l-1})\phi^*_{l,s}\phi_{l,s} \Bigg\}. \end{aligned} \tag{3}$$

By making Fourier transformations on the integral variables

$$u_l(\tau) = \frac{1}{\sqrt{\beta L}} \sum_{q,\Omega} e^{i(ql+\Omega\tau)} u_q(\Omega), \tag{4}$$

and similarly for $\phi^*_{l,s}$ and $\phi_{l,s}$, then for the phonon variables we have two branches on a dimerized lattice, that is, the acoustic phonon $a_q(\Omega)$ [$\equiv u_q(\Omega)$] and the optical phonon $b_q(\Omega)$ [$\equiv u_{q+Q}(\Omega)$], $Q = 2k_F$ ($k_F = \pi/2$ for the band half-filling, the lattice constant has been set as unit). For the Grassmann variables we could introduce a spinor notation via[9]

$$\Phi_{k,s} = \begin{pmatrix} \phi_{k+k_{F,s}} \\ \phi_{k-k_{F,s}} \end{pmatrix}, \tag{5}$$

which are right- and left-moving electrons, respectively, we could rewrite the partition function as

$$Z = \int \mathcal{D}[a^*a, b^*b, \Phi^*\Phi]e^{-S}, \tag{6}$$

and the corresponding action could be written as a sum of five terms,

$$S = S_a + S_b + S_e + S_{a-e} + S_{b-e}, \tag{7}$$

where the acoustic- and optical-phonon parts of the action are, respectively,

$$S_a = \sum_{q,\Omega} [K + \tfrac{1}{2} M\Omega^2] a_q(\Omega) a_{-q}(-\Omega), \tag{8a}$$

$$S_b = \sum_{q,\Omega} [K + \tfrac{1}{2} M\Omega^2] b_q(\Omega) b_{-q}(-\Omega), \tag{8b}$$

the electronic part of the action is

$$S_e = \sum_{k,\omega,s} \Phi^{\dagger}_{k,s}(\omega)[(i\omega - \mu) + 2t_0 \sin k\sigma_3]\Phi_{k,s}(\omega), \tag{8c}$$

and the acoustic-, and optical-phonon–electron interaction parts are the following:

$$\begin{aligned} S_{a-e} = & -\frac{2i\alpha}{\sqrt{\beta L}} \sum_{q,\Omega} \sum_{k,\omega,s} \sin(q/2) a_q(\Omega) \\ & \times \Phi^{\dagger}_{k,s}(\omega)\Phi_{k-q,s}(\omega - \Omega), \end{aligned} \tag{8d}$$

$$\begin{aligned} S_{b-e} = & -\frac{2\alpha}{\sqrt{\beta L}} \sum_{q,\Omega} \sum_{k,\omega,s} \cos(q/2) b_q(\Omega) \\ & \times \Phi^{\dagger}_{k,s}(\omega)\sigma_1\Phi_{k-q,s}(\omega - \Omega), \end{aligned} \tag{8e}$$

where σ_i are Pauli matrices.

As seen from Eq. (8d), the coupling between electrons and acoustic phonons vanishes for small q. Hence we will focus on the optical phonons although the effects of acoustic phonons can be readily taken into account, and the frequency of acoustic phonons will remain as the bare one $\omega_A(q) = \omega_Q \equiv \sqrt{2K/M}$, a dispersionless excitation. One can obtain an effective phonon action by integrating out the electronic variables of the partition since the action in Eq. (7) is bilinear in the Grassmann fields. The resulting partition function is

$$Z = \int \mathcal{D}[b^*b]e^{-S_{\text{eff}}}, \tag{9}$$

and the effective action is

$$S_{\text{eff}} = \sum_{q,\Omega} [K + \tfrac{1}{2} M\Omega^2] b_q(\Omega) b_{-q}(-\Omega) - N \ln \det(\mathcal{M}), \tag{10}$$

where N is the spin degree of freedom and det denotes the determinant of the matrix $\mathcal{M}$, which is defined by the actions $S_e + S_{b-e} = \sum_s \Phi^{\dagger}_s \mathcal{M} \Phi_s$ and has k, ω, and the indices of the Pauli matrices as labels.

III. THE PHONON EXCITATIONS

A. Saddle point approximation

By the functional derivation of the effective action S_{eff} with the phonon variable $b_q(\Omega)$ vanishing, we have

the lattice dimerization, i.e., the phonon condensation at zero momentum and zero frequency $\langle 2\alpha b_q(\Omega)\rangle = \sqrt{\beta L}\Delta\delta_{q,0}\delta_{\Omega,0}$, where the phonon order parameter Δ describes the lattice dimerization. Defining the unperturbed electronic Green's function $G_0(k,\omega)$ by

$$G_0(k,\omega) = -i(i\omega - \mu + 2t_0 \sin k\sigma_3 - \Delta\sigma_1)^{-1}, \quad (11)$$

and $b_q(\Omega) = \langle b_q(\Omega)\rangle + \tilde{b}_q(\Omega)$, we obtain the effective action by expanding the logarithm in order of $\tilde{b}$:

$$S_{\text{eff}} = S_{\text{eff}}^{(0)} + \sum_{n=1}^{\infty} S_{\text{eff}}^{(n)}. \quad (12)$$

$S_{\text{eff}}^{(0)}$ is the zeroth-order contribution in $\tilde{b}$ and is given by

$$S_{\text{eff}}^{(0)} = N\beta L\left\{\frac{\Delta^2}{8\pi\lambda t_0} - \int \frac{dkd\omega}{(2\pi)^2} \ln\det G_0^{-1}(k,\omega)\right\} \quad (13)$$

in the thermodynamic limit, and the dimensionless electron-phonon coupling constant λ is defined by $\lambda = N\alpha^2/2\pi K t_0$, the chemical potential $\mu = 0$ at the half-filling case. The electronic spectrum $E_k = \pm\sqrt{(2t_0 \sin k)^2 + \Delta^2}$, the electronic gap is 2Δ.

The mean-field gap equation

$$\Delta_{\text{ad}} = \Lambda e^{-1/2\lambda} \quad (14)$$

follows immediately from

$$\frac{\partial S_{\text{eff}}^{(0)}}{\partial\Delta} = 0, \quad (15)$$

where Λ is the integral cutoff. The first-order term $S_{\text{eff}}^{(1)}$ vanishes since $S_{\text{eff}}^{(0)}$ is obtained from the saddle point approximation. It can be seen that the solution Eq. (14) becomes exact in the limit of the spin degree of freedom $N \to \infty$ since the zeroth-order action is proportional to N.

B. The phonon excitations

The second-order contribution to the effective phonon action is

$$S_{\text{eff}}^{(2)} = \sum_{q,\Omega} \tilde{b}_q(\Omega)\tilde{b}_{-q}(-\Omega)[\tfrac{1}{2}M\Omega^2 + K\sin^2(q/2) + 2K\lambda f(q,\Omega)\cos^2(q/2)], \quad (16)$$

where the function $f(q,\Omega)$ is defined as

$$f(q,\Omega) = 1/2\lambda + 2\pi t_0 \int \frac{dkd\omega}{(2\pi)^2} \times \text{Tr}[G_0(k-q,\omega-\Omega)\sigma_1 G_0(k,\omega)\sigma_1]. \quad (17)$$

The frequencies of optical phonons are determined by the vanish of the coefficients in Eq. (16),[18] that is, the equation for the optical-phonon excitations is

$$\Omega^2 + \omega_Q^2 \sin^2(q/2) + \omega_0^2 f(q,\Omega)\cos^2(q/2) = 0, \quad (18)$$

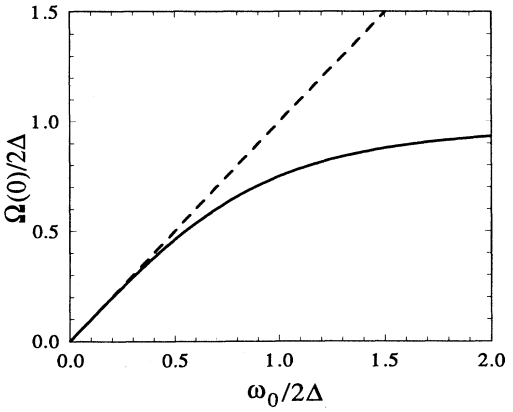

FIG. 1. The dependence of the optical-phonon frequency $\Omega(0)$ on the ionic mass M. ω_0 is defined in the text. The dashed line is the adiabatic result $\Omega(0) = \omega_0$. In the antiadiabatic limit, $M = 0$, $\omega_0 = \infty$, and $\Omega(0) = 2\Delta$.

which applies to all values of the halogen ionic mass ranging from the $M = 0$ (antiadiabatic limit) to the $M = \infty$ (adiabatic case) limit, and $\omega_0^2 \equiv 2\lambda\omega_Q^2$. The calculation of the function $f(q,\Omega)$ could be done in a straightforward way with the help of the mean-field gap equation (14), the result shown as a function of a single variable $\eta = \sqrt{4t_0^2q^2 + \Omega^2}/2\Delta$, that is, $f(q,\Omega) = F(\eta)$, with the function $F(\eta)$ as

$$F(\eta) = \eta^{-1}(1+\eta^2)^{1/2}\sinh^{-1}\eta. \quad (19)$$

Actually, Eq. (18) is a self-consistent equation for the phonon excitations. Since we used a real temperature variable τ in the Fourier transformation in Eq. (4), the renormalized optical-phonon frequency $\Omega(q)$ should be related with Ω in Eq. (18) by $\Omega(q) = i\Omega$, with which we can get a real frequency $\Omega(q)$ by solving Eq. (18). Looking at the case $q = 0$, the phonon excitation energies are determined by the self-consistent equation

$$\xi^2 = g^2\sqrt{1-\xi^2}\frac{\sin^{-1}\xi}{\xi}, \quad (20)$$

where $\xi = \Omega(0)/2\Delta$ and $g = \omega_0/2\Delta$. The numerical result is given in Fig. 1. It can be seen that the adiabatic approximation (the dashed line in Fig. 1) is good for $\omega_0/2\Delta < 0.4$ and the phonon excitation energies $\Omega(0)$ are always lower than that of electronic exitations. In the limit of $M = 0$, $\Omega(0) = 2\Delta$, the gap of optical phonons equals that of electronic excitations.

IV. THE PHONON ORDER PARAMETER

A. One-loop approximation

By performing the integration in Eq. (9) over the fluctuations $\tilde{b}$ to the second-order term, we have

$$Z = e^{-\beta L \Gamma[\Delta]}, \tag{21}$$

where the free energy density $\Gamma[\Delta]$ is composed of two parts,

$$\Gamma[\Delta] = \Gamma_0[\Delta] + \Gamma_1[\Delta], \tag{22}$$

the $\Gamma_0[\Delta] = S_{\rm eff}^{(0)}/\beta L$ is the zeroth-order contribution, and the one-loop contribution $\Gamma_1[\Delta]$ is given by

$$\Gamma_1[\Delta] = \frac{1}{2}\int \frac{dqd\Omega}{(2\pi)^2} \ln[\tfrac{1}{2}M\Omega^2 + K\sin^2(q/2) + 2\lambda K f(q,\Omega)\cos^2(q/2)] \tag{23}$$

in the zero-temperature and thermodynamic limit. The equation for the phonon order parameter Δ is determined by $\partial\Gamma[\Delta]/\partial\Delta = 0$. Within the one-loop approximation, we have

$$1 = 8\pi\lambda t_0 \int \frac{dkd\omega}{(2\pi)^2}\left\{\frac{1}{\omega^2 + E_k^2} - \frac{1}{4N\Delta}\left[\frac{\omega^2 + \omega_Q^2\sin^2(k/2)}{\omega_0^2 f'(k,\omega)\cos^2(k/2)} + \frac{f(k,\omega)}{f'(k,\omega)}\right]^{-1}\right\}. \tag{24}$$

The derivation of the function $f(k,\omega)$ with the phonon order parameter can be obtained by Eq. (17). It is $f'(k,\omega) = 4\Delta f(k,\omega)/(\omega^2 + 4t_0^2k^2 + 4\Delta^2)$. Then the electronic gap Δ satisfies the equation

$$1 = 8\pi\lambda t_0 \int \frac{dkd\omega}{(2\pi)^2}\left\{\frac{1}{\omega^2 + 4t_0^2k^2 + \Delta^2} - \frac{1}{N}\frac{1}{d_1^2\omega^2 + d_2^2 4t_0^2k^2 + 4\Delta^2}\right\}, \tag{25}$$

where we have used the abbreviations

$$d_1^2 = 1 + \frac{4\Delta^2}{\omega_0^2}, \tag{26}$$

$$d_2^2 = 1 + \frac{4\Delta^2}{2\lambda(4t_0)^2}. \tag{27}$$

d_1 describes the effects of the finite ionic mass while d_2 is caused by the lattice elastic energies. By performing the integral of Eq. (25), we get the electronic gap

$$\Delta = \Lambda \exp\left[-\left(1 - \frac{2\lambda\ln 2}{Nd_1d_2}\right)\Big/ 2\lambda\left(1 - \frac{1}{Nd_1d_2}\right)\right]. \tag{28}$$

It can be seen that in the adiabatic limit ($M \to \infty$) $d_1 \to \infty$, the quantum fluctuation is completely suppressed, and $\Delta = \Delta_{\rm ad}$ is the mean-field result (14). In the antiadiabatic limit ($M = 0$) $d_1 = 1$, although $d_2 > 1$, the behavior of Eq. (28) should be similar to $d_2 = 1$ in the weak electron-phonon interaction since it is governed by the $M = 0$ limit as shown by the renormalization-group arguments. The numerical calculations also show it.

We show the numerical results of Eq. (28) in Figs. 2 and 3. Figure 2 gives the dependence of the gap parameter Δ on the electron-phonon coupling λ for both the adiabatic and the antiadiabatic limits and for an intermediate ionic mass M. The behavior of the system for an intermediate ionic mass M is interesting. For a weak electron-phonon interaction, the electronic gap Δ is extremely close to the value of that in the antiadiabatic limit, i.e., the quantum fluctuation reaches its maximum. This result is in agreement with the renormalization-group analysis.[16] However, for a stronger electron-phonon coupling λ, Δ becomes close to $\Delta_{\rm ad}$, i.e., the quantum fluctuation is suppressed by the electron-phonon interaction. This behavior could be understood because we know that the electron-phonon interaction will induce the electronic cloud around the ions and then the effective mass of the polaron formed by an ion with its surrounding electronic cloud becomes very large for a strong electron-phonon interaction. It is

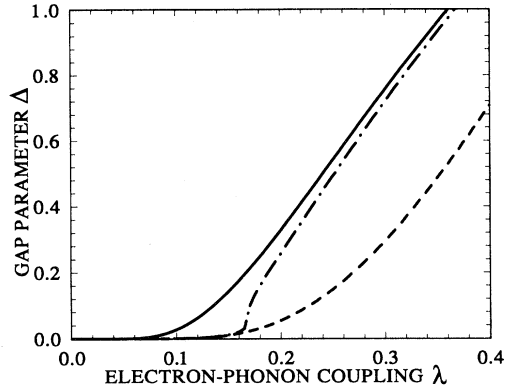

FIG. 2. The dependence of the gap parameter Δ on the electron-phonon coupling λ. The solid line is for $M \to \infty$ and the dashed line is for $M = 0$. The dot-dashed line is for the system with an intermediate ionic mass M ($\omega_Q = 0.2$). t_0 is taken as unit.

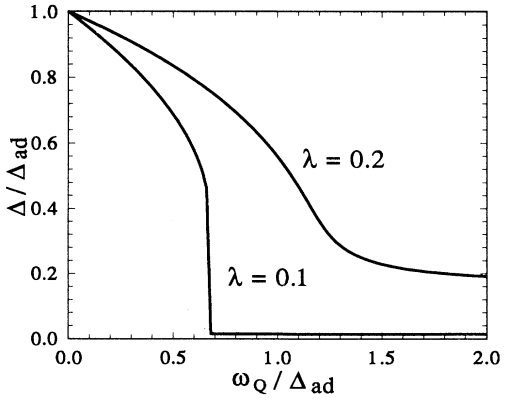

FIG. 3. The dependence of the phonon order parameter Δ on the ionic mass M (equivalently on the bare phonon frequency ω_Q) for different electron-phonon couplings λ. $\Delta_{\rm ad}$ is the value in the adiabatic limit.

easy to see that the quantum fluctuations are suppressed for the system with a large ionic mass. Figure 3 shows the dependence of the gap parameter Δ on the ionic mass M (or equivalently on the phonon frequency ω_Q, which is proportional to $1/\sqrt{M}$) for different electron-phonon couplings λ. It can be seen that for a weak electron-phonon coupling, such as $\lambda = 0.1$, the gap parameter Δ decreases with the increase of the phonon frequency ω_Q, and at a critical point (it is $\omega_Q/\Delta_{\rm ad} = 0.68$ for $\lambda = 0.1$) there is a sudden jump and then Δ becomes the value at the $M = 0$ limit. In other words, at a large region of the ionic mass from zero, the quantum fluctuation is important and the gap parameter Δ is reduced to a very small value, however when the ionic mass increases to a critical value, the quantum fluctuation is mostly suppressed and then Δ has a jump. For a stronger electron-phonon interaction, Δ decreases smoothly with the increase of the phonon frequency ω_Q. The parameter region, where the behavior of the system is governed by the antiadiabatic limit, decreases with the increase of the electron-phonon coupling λ.

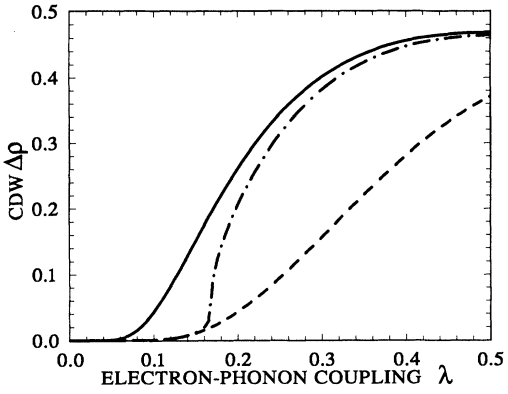

FIG. 4. The dependence of the charge-density wave $\Delta\rho$ on the electron-phonon coupling λ. The solid line is for $M \to \infty$ and the dashed line is for $M = 0$. The dot-dashed line is for the system with an intermediate ionic mass M ($\omega_Q = 0.2$). t_0 is taken as unit.

B. The charge-density wave

The CDW parameter $\Delta\rho$, which is defined by $\langle \sum_s c^\dagger_{ls} c_{ls} \rangle = 1 - (-1)^l \Delta\rho$, on the metal ions can be calculated as follows:

$$\Delta\rho = -\frac{1}{\beta L} \sum_{k,\omega,s} \langle \Phi^\dagger_{k,s}(\omega) \sigma_1 \Phi_{k,s}(\omega) \rangle = \frac{1}{\beta L} \frac{\partial \ln Z_e}{\partial \Delta}, \tag{29}$$

where the electronic partition function Z_e is defined by $Z = Z_e Z_k$, and $Z_k = \exp(-N\beta L \Delta^2 / 8\pi\lambda t_0)$ is the partition function caused by the elastic energies of the lattice. By using the equation $\partial\Gamma[\Delta]/\partial\Delta = 0$ for the determinaton of the phonon order parameter Δ, we have the charge disproportion

$$\Delta\rho = \frac{N}{4\pi\lambda t_0}\Delta. \tag{30}$$

The dependence of the charge-density wave $\Delta\rho$ on the electron-phonon coupling λ is shown in Fig. 4, from which a similar behavior with the gap parameter Δ can be seen.

V. SUMMARY

In this paper, we have studied the effects of quantum lattice fluctuations on the CDW of MX complexes by a functional integral approach. Both the phonon order parameter and the phonon excitations for any value of the ionic mass M are obtained. The calculation is performed by first integrating out the electronic variables and then expanding the effective action to the quadratic terms in the phonon variables. The equations for the phonon excitations are derived by vanishing the coefficients of the quadratic term of the effective action. Our results show that the phonon excitation has the electronic gap as its maximum in the antiadiabatic limit. The equation for the phonon order parameter (also the charge-density wave) is obtained by integrating the phonon variables up to the quadratic terms. For the system with an intermediate ionic mass the low-energy behavior is governed by the antiadiabatic ($M = 0$) limit, which is in agreement with the renormalization-group analysis of Fradkin and Hirsch. However, for a stronger electron-phonon interaction, the quantum lattice fluctuation is suppressed and the system's behavior is governed by the adiabatic limit.

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