PHYSICAL REVIEW B VOLUME 42, NUMBER 10 1 OCTOBER 1990

Discontinuous change of superconducting transition temperature from BCS type to bipolaron type in strongly coupled electron-phonon systems

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(Received 10 April 1990)

A unified theory for the superconducting transition temperature T_c of an electron-phonon coupled system has been derived, so as to clarify the nature of the intermediate region of coupling strength, as well as the weak (BCS) region and the strong (bipolaronic) one. This theory is mainly based on the polaron picture and takes the thermal fluctuation of pairing order into account within the coherent-potential approximation. In the weak-coupling limit, it reduces to the Eliashberg theory and gives a T_c that increases as the coupling increases, while in the strong-coupling region, it reduces to a pseudospin-type theory of bipolarons, and T_c decreases as the coupling increases. Thus, T_c takes its maximum in the intermediate region. It is shown, however, that this change of T_c from the BCS region to the bipolaronic one becomes discontinuous when the bandwidth of electron is greater than the phonon energy, while in the opposite case it changes continuously. The calculated results of the former case are discussed in connection with the insulator-superconductor transition of $Ba_{1-x}K_xBiO_3$.

I. INTRODUCTION

Theoretical problems related to very strong electron-phonon (*e*-ph) coupling have been subjects of considerable interest in recent years. In the case of one- and two-body problems of electrons, they are closely related to the optical properties of insulating solids, such as alkali metal halides, rare-gas solids, and semiconductors.[1] On the other hand, in the case of many-body problems, they are closely related to the superconductivity and the charge-density-wave (CDW) states of various metallic solids, such as the transition-metal oxides and low-dimensional conductors.[1–14] In the early stage of studies for these strong-coupling phenomena, the one- (or two-) body problem and the many-body problem have been developed rather independently of each other, since these two kinds of problems correspond to two quite different states of solids: insulators and conductors. At present, however, they are combined, so as to clarify various new phenomena coming from this strong coupling.[4,5]

In the one-body problem, as is well known, the very strong *e*-ph coupling causes a spontaneous breakdown of the translational symmetry of an electron. When the *e*-ph coupling is weak, the electron is in a wavelike state with the same translational symmetry as the crystal. However, in the case of a very strong *e*-ph coupling, it is in a localized state, being trapped by a self-induced local lattice distortion. This change of electron occurs almost discontinuously as the coupling increases, provided the energy-band width of electron is much greater than the phonon energy.[1] On the other hand, this change is continuous, when the width is of the same order as that of the phonon energy.

In the case of the many-electron system, we have two types of competing instabilities coming from the strong *e*-ph coupling: the superconducting (SP) state[2] and the CDW state.[3] The CDW state mainly comes from the adiabatic nature of the *e*-ph coupling, since we have a frozen lattice distortion, that results in the structural change of the crystal. On the other hand, the SP state comes from the inverse-adiabatic nature of *e*-ph coupling, since the local lattice distortion is always moving together with the electron, resulting in no frozen lattice distortion. The relative stability between these two states changes from material to material, according to the strength of *e*-ph coupling, the energy of phonons, the geometry of the Fermi surface, and the total number of electrons.[4,5]

Even though we have restricted ourselves only within the SP state, we still have two possibilities of the pairing, which are quite different from each other in their natures. The first one is the conventional BCS-type,[2] wherein the electrons only around the Fermi surface make pairs called Cooper pairs, while other electrons far from the Fermi surface remain unpaired. The second is the bipolaronic pairing,[6] wherein all the electrons in the conduction band make the singlet pairs. The interrelation between these two kinds of states is a matter of considerable interest in recent years, since it is closely related to the mechanism of the high-temperature superconductivity and the CDW states in various transition metal oxides, such as $LiTi_2O_4$,[7] $Ba_{1-x}K_xBiO_3$,[8–11] La-Sr-Cu-O,[12] Ti_4O_7,[13] and $Na_{0.33}V_2O_5$.[14]

In connection with these problems, Nasu[4,5] has studied the SP state coming from very strong *e*-ph couplings, using the polaron-type theory combined with the coherent-potential approximation for the fluctuation of pairing orders. In this study, it is shown that the nature of pairing changes from the conventional BCS-type to the bipolaronic one as the coupling increases. The transition temperature ($\equiv T_c$) is also shown to take its maximum value, neither in the BCS region nor in the bipolaronic one, but in the intermediate region between them.

In this theory, however, the nature of the intermediate region is not studied in detail, except the fact that this region becomes narrow as the ratio of the electron energy bandwidth to the phonon energy increases. Moreover, this theory does not reduce to the Eliashberg theory[15] in the weak-coupling limit.

In the present paper, we will derive a new unified theory for the superconducting transition temperature of a strongly coupled electron-phonon system, so as to reinforce our previous theory and clarify the nature of intermediate region. It will be shown that the transition from the BCS type to the bipolaronic one is almost discontinuous when the ratio of the bandwidth to the phonon energy is large, while it is continuous when this ratio is small.

Results of the former case will be discussed in connection with the newly discovered high-temperature superconductor $Ba_{1-x}K_xBiO_3$, especially in connection with its insulator-superconductor transition at around $x=0.375$.

II. HAMILTONIAN OF MANY-POLARON SYSTEM

Let us consider a model system composed of N lattice sites and N_e electrons ($N>N_e\gg 1$) with the following Hamiltonian ($\equiv H$),

$$H\equiv -T\sum_{\langle l,l'\rangle}\sum_{\sigma}\eta^{\dagger}_{l\sigma}\eta_{l'\sigma}+\sqrt{S\omega/2}\sum_{l,\sigma}(\zeta^{\dagger}_l+\zeta_l)n_{l\sigma} +\omega\sum_{l}\zeta^{\dagger}_l\zeta^{\dagger}_l \; . \tag{2.1}$$

Here T is the transfer energy of an electron between neighboring two lattice sites l and l' of a simple cubic crystal, and $\sum_{\langle l,l'\rangle}$ means the summation over all neighboring pairs of sites. $\eta^{\dagger}_{l\sigma}$ is the creation operator of an electron localized at site l with spin σ ($=\alpha$, β, where α denotes the up-spin and β denotes the down one). S is the site-diagonal e-ph coupling energy. $\zeta^{\dagger}_l$ is the creation operator of a phonon localized at site l with the energy ω, and $n_{l\sigma}\equiv\eta^{\dagger}_{l\sigma}\eta_{l\sigma}$. The interelectron Coulombic repulsion is completely neglected for simplicity. It is expedient to cast all quantities into dimensionless forms as

$$h\equiv H/\omega,\quad t\equiv T/\omega,\quad s\equiv S/\omega\;, \tag{2.2}$$

and this notation will be used hereafter.

As is already well known, each electron in the electron-phonon coupled system forms a polaron with a phonon cloud around itself, and the thickness of this cloud is of great importance. In the case of the one-electron problem ($N_e=1$), it is mainly determined by the competition between t and s. However, in the case of the many-electron system ($N_e\gg 1$), it is also determined by the electron density $c\equiv(N_e/2N)$, as well as by the nature of the Fermi surface and the crystal structure. In order to take these effects into account, we introduce a displacement operator ($\equiv M$) for phonon as

$$M=\exp\left[\sqrt{s/2}q\sum_{l,\sigma}(\zeta^{\dagger}_l-\zeta_l)n_{l\sigma}\right]\;, \tag{2.3}$$

where q is the variational parameter that denotes the thickness of the phonon cloud around the electron. At the present stage, it is unknown but will be determined later. As inferred from Migdal's theorem, it will be very small $q\ll 1$ in the weak-coupling case $S\ll T$ or in the adiabatic case $T\gg\omega$, while it will become maximum $q=1$ in the strong-coupling case $S\gg T$ or in the inverse-adiabatic case $T\ll\omega$. By using M, thus defined, we can transform the electron and the phonon into a polaron $a^{\dagger}_{l\sigma}$ and a new phonon $d^{\dagger}_l$, which are defined as

$$\begin{aligned}a^{\dagger}_{l\sigma}&\equiv M^{-1}\eta^{\dagger}_{l\sigma}M\\&=\exp[-\sqrt{s/2}q(\zeta^{\dagger}_l-\zeta_l)]\eta^{\dagger}_{l\sigma}\;,\end{aligned} \tag{2.4}$$

$$d^{\dagger}_l\equiv M^{-1}\zeta^{\dagger}_lM=\zeta^{\dagger}_l+\sqrt{s/2}q\sum_{\sigma}n_{l\sigma}\;, \tag{2.5}$$

where $a^{\dagger}_{l\sigma}$ is the creation operator of a polaron with a phonon cloud around itself, and $d^{\dagger}_l$ denotes the creation operator of a new phonon whose equilibrium position has been already displaced because of the e-ph coupling. In terms of the polaron and new phonon, we can write h as

$$\begin{aligned}h=&-t\sum_{\langle l,l'\rangle}\sum_{\sigma}a^{\dagger}_{l\sigma}a_{l'\sigma}\exp[\sqrt{s/2}q(d^{\dagger}_l-d_l-d^{\dagger}_{l'}+d_{l'})]\\&-u\sum_{l}n_{l\alpha}n_{l\beta}+h_{\text{ph}}-uN_e/2\\&+\sqrt{s/2}(1-q)\sum_{l}(d^{\dagger}_l+d_l)n_{l\sigma}\;,\end{aligned} \tag{2.6}$$

$$h_{\text{ph}}\equiv\sum_{l}d^{\dagger}_ld_l,\quad u\equiv s(2q-q^2)\;, \tag{2.7}$$

where the first term of Eq. (2.6) denotes the transfer of a polaron, the second one denotes the attraction between two polarons, and the third one denotes the Hamiltonian of new phonon. The last term [Eq. (2.7)] denotes the residual linear interaction between the new phonon and the polaron, which is not taken into account by the transformation M.

Let us now set up an effective Hamiltonian of a many-polaron system. Since the coordinate of the phonon has already been displaced by M so as to take the e-ph coupling into account, the equilibrium state of the new phonon acts as our reference state. The effective Hamiltonian of our many-polaron system ($\equiv h_0$) in this equilibrium state can be defined as

$$h_0\equiv\text{Tr}_{\text{ph}}(e^{-\Theta h_{\text{ph}}})/\text{Tr}(e^{-\Theta h_{\text{ph}}}),\quad \Theta\equiv\omega/k_BT_{\text{temp}}\;, \tag{2.8}$$

where Tr_{ph} means the trace in the phonon space, and Θ denotes the reciprocal temperature (T_{temp} is the temperature). h_0 is given as

$$h_0=-tX\sum_{\langle l,l'\rangle}\sum_{\sigma}a^{\dagger}_{l\sigma}a_{l'\sigma}-ucN-u\sum_{l}n_{l\alpha}n_{l\beta}\;, \tag{2.9}$$

where X is the reduction factor of the transfer energy due to the overlap integral of the phonon clouds between two neighboring sites l and l':

$$X\equiv\exp(-sq^2/2)\;. \tag{2.10}$$

Here, we have neglected the thermal excitation of the phonon, assuming the temperature is low; $\omega\gg k_BT_{\text{temp}}$. Using h_0, thus defined, we can divide h into four parts as

$$h = h_0 + h_{\mathrm{ph}} + h_1 + h_2 \ , \tag{2.11}$$

$$h_1 \equiv \sqrt{s/2}(1-q) \sum_{l,\sigma} (d_l^\dagger + d_l) n_{l\sigma} \ , \tag{2.12}$$

$$h_2 = -t \sum_{\langle l,l' \rangle} \sum_{\sigma} a_{l\sigma}^\dagger a_{l'\sigma} \times \{ \exp[\sqrt{s/2}q(d_l^\dagger - d_l - d_{l'}^\dagger + d_{l'})] - X \} \ , \tag{2.13}$$

where h_1 denotes the aforementioned residual interaction and h_2 denotes the difference between the real transfer and the averaged one.

As shown in our previous paper,[4] the unitary transformation M must also take a frozen part of lattice distortion into account when we want to study the possibility of a CDW state. However, throughout the present paper, we will completely neglect this possibility, and concentrate only on the SP state for simplicity. In order to study the SP state, it is rather convenient to describe all the energies from the chemical potential μ. Hence, we define Hamiltonians $\tilde{h}$ and $\tilde{h}_0$, which are referenced from μ, as

$$\tilde{h} \equiv h - \mu N_e \ , \tag{2.14}$$

$$\tilde{h}_0 \equiv h_0 - \mu N_e \ , \tag{2.15}$$

and we also transform them into the following Nambu representation:

$$a_{l\alpha} \rightarrow A_{l1}, \quad a_{l\beta} \rightarrow A_{l2}^\dagger \ , \tag{2.16}$$

where A_{li} $(i=1,2)$ is a new fermion operator. In terms of them, $\tilde{h}_0$ is written as

$$\tilde{h}_0 = \sum_k E_k (A_{k1}^\dagger A_{k1} - A_{k2}^\dagger A_{k2}) - u \sum_l A_{l1}^\dagger A_{l2} A_{l2}^\dagger A_{l1} + \sum_k E_k - ucN \ , \tag{2.17}$$

$$E_k \equiv e_k - \mu, \quad e_k \equiv -tX \sum_{\langle l,0 \rangle} e^{-ikl} \ , \tag{2.18}$$

$$A_{ki} \equiv \sum_l N^{-1/2} e^{-ikl} A_{li} \ , \tag{2.19}$$

where k is the wave vector of electron. h_1 and h_2 are also rewritten as

$$h_1 = \sqrt{s/2}(1-q) \sum_l (d_l^\dagger + d_l)(A_{l1}^\dagger A_{l1} - A_{l2}^\dagger A_{l2}) \ , \tag{2.20}$$

$$h_2 = -t \sum_{\langle l,l' \rangle} (A_{l1}^\dagger A_{l'1} - A_{l2}^\dagger A_{l'2}) \times \{ \exp[\sqrt{s/2}q(d_l^\dagger - d_l - d_{l'}^\dagger + d_{l'})] - X \} \ . \tag{2.21}$$

In Eq. (2.20) we have neglected the occurrence of the uniform displacement of a phonon.

III. TWO MEAN-FIELD THEORIES

Let us now study the superconducting transition temperature T_c given by $\tilde{h}_0$ within the mean-field theories, neglecting h_1 and h_2. The effects of h_1 and h_2 will be studied later in detail. We have two types of mean-field theories. The one is the conventional BCS theory,[2] being useful in the weak-coupling region $s \ll t$, and the other is the pseudospin-type mean-field theory[5,6] for bipolarons, being useful in the inverse-adiabatic and strong-coupling limits $1 \gg s \gg t$.

In the former mean-field theory, the attraction term in Eq. (2.17) is decoupled as

$$-u \sum_l A_{l1}^\dagger A_{l2} A_{l2}^\dagger A_{l1} \rightarrow -uc^2 N - u\bar{Q} \sum_l (A_{l1}^\dagger A_{l2} + A_{l2}^\dagger A_{l1}) + u\bar{Q}^2 N \ , \tag{3.1}$$

where the first term of the right-hand side comes from the diagonal decoupling and the second one comes from off-diagonal decoupling. $\bar{Q}$ in Eq. (3.1) is the order parameter defined as

$$\bar{Q} \equiv \sum_l N^{-1} \langle A_{l1}^\dagger A_{l2} \rangle \ , \tag{3.2}$$

where $\langle \cdots \rangle$ is the expectation value of $\cdots$. This $\bar{Q}$, at present stage, is unknown, and should be determined self-consistently. Using this approximated form for the attraction term, we can reduce $\tilde{h}_0$ into a mean-field-theory Hamiltonian h_{MF} which has a 2×2 matrix form

$$\tilde{h}_0 \rightarrow \tilde{h}_{\mathrm{MF}} = \sum_k (A_{k1}^\dagger, A_{k2}^\dagger) \begin{bmatrix} E_k & -F \\ -F & E_k \end{bmatrix} \begin{bmatrix} A_{k1} \\ A_{k2} \end{bmatrix} + uN\bar{Q}^2 - uc(c+1)N + \sum_k E_k, \quad F \equiv u\bar{Q} \ . \tag{3.3}$$

This 2×2 matrix can be diagonalized by the following unitary transformation from A_{ki} to a new fermion operator Z_{ki}:

$$\begin{bmatrix} Z_{k1}^\dagger \\ Z_{k2} \end{bmatrix} = \begin{bmatrix} \cos(\phi_k) & \sin(\phi_k) \\ -\sin(\phi_k) & \cos(\phi_k) \end{bmatrix} \begin{bmatrix} A_{k1} \\ A_{k2} \end{bmatrix} \ , \tag{3.4}$$

$$\phi_k \equiv \arctan[(Y_k + E_k)^{1/2} / (Y_k - E_k)^{1/2}] \ , \quad Y_k \equiv (E_k^2 + F^2)^{1/2} \ . \tag{3.5}$$

Here F in Eqs. (3.3) and (3.5) denotes the energy gap, and Y_k is the energy of new quaiparticles created by $Z_{k1}^\dagger$ and $Z_{k2}^\dagger$. In terms of F and Y_k, $\tilde{h}_{\mathrm{MF}}$ is rewritten as

$$\tilde{h}_{\mathrm{MF}} = \sum_k Y_k (Z_{k1}^\dagger Z_{k1} + Z_{k2}^\dagger Z_{k2}) + u\bar{Q}^2 N - uc(c+1)N + \sum_k E_k - \sum_k Y_k \ . \tag{3.6}$$

Using this form of $\tilde{h}_{\mathrm{MF}}$, we can evaluate the thermal average in Eq. (3.2), and can get the gap equation as

$$F = u \sum_k N^{-1} \frac{F}{2Y_k} \tanh(\Theta Y_k / 2) \ . \tag{3.7}$$

Since the chemical potential μ itself is still unknown, we must determine it from the following condition:

$$N_e/2=\sum_l \langle A_{l1}^\dagger A_{l1}\rangle \ . \tag{3.8}$$

After taking this average using $\tilde{h}_{\mathrm{MF}}$, we get an equation for μ as

$$c=\tfrac{1}{2}\sum_k N^{-1}\left[1-\frac{E_k}{Y_k}\tanh\left[\frac{\Theta Y_k}{2}\right]\right] . \tag{3.9}$$

The thickness parameter q is also still unknown, and we determine it so as to minimize the total thermodynamic potential. The thermodynamic potential ($\equiv\Omega_{\mathrm{MF}}$) within this mean-field theory is given by

$$\Omega_{\mathrm{MF}}\equiv-\Theta^{-1}\ln[\mathrm{Tr}(e^{-\Theta\tilde{h}_{\mathrm{MF}}})] , \tag{3.10}$$

and, in terms of Y_k and F, it is written by

$$\Omega_{\mathrm{MF}}=-2\Theta^{-1}\sum_k \ln(1+e^{-\Theta Y_k}) -uc(c+1)N+\sum_k (E_k-Y_k)+NF^2/u \ . \tag{3.11}$$

Using this form, we can determine q from the following condition:

$$\frac{\partial\Omega_{\mathrm{MF}}}{\partial q}=0 \ . \tag{3.12}$$

In this extremization of Ω_{MF} with respect to q, we can assume that F and μ are independent of q since the extremizations with respect to F and μ have been already finished by Eqs. (3.7) and (3.9). Thus, we get the following equation for q:

$$q=\frac{c(c+1)+\bar{Q}^2}{c(c+1)+\bar{Q}^2+\mu(\frac{1}{2}-c)+\sum_k N^{-1}\tanh(\Theta Y_k/2)E_k^2/Y_k} \ . \tag{3.13}$$

These three equations for F, μ, and q can be solved numerically, and their qualitative results at $T_{\mathrm{temp}}=0$ are summarized as follows. q increases from 0 to 1 as s/t increases,

$$q=0\sim 1 \ . \tag{3.14}$$

μ changes from the Fermi energy ($\equiv\varepsilon_f$) of a noninteracting electron to $-s(\frac{1}{2}-c)$,

$$\mu=\varepsilon_f\sim -s(\tfrac{1}{2}-c) \ . \tag{3.15}$$

F increases from zero to $s\sqrt{c(1-c)}$,

$$F=0\sim s\sqrt{c(1-c)} \ . \tag{3.16}$$

T_c given by this theory increases as s/t increase, just like F.

Let us now proceed to the inverse-adiabatic and the strong coupling limits, $1\gg s\gg t$. In this case, we can take the effects of t within the second-order perturbation, and can get the so-called pseudospin theory. In this case, all the electrons in the energy band form singlet pairs, called bipolarons, each of which is localized almost within a single site. We can describe this many-polaron system, using $\frac{1}{2}$ pseudospin assigned for each site, wherein up-spin corresponds to the presence of a bipolaron, while down-spin corresponds to absence. Thus, we get an effective pseudospin Hamiltonian $\underline{H}_s$ as

$$\begin{aligned}\underline{H}_s=-2J\sum_{\langle l,l'\rangle}[&(0.5+\underline{S}_l^z)(0.5-\underline{S}_{l'}^z)\\ &+(0.5-\underline{S}_l^z)(0.5+\underline{S}_{l'}^z)\\ &+\underline{S}_l^\dagger\underline{S}_{l'}^- +\underline{S}_l^-\underline{S}_{l'}^+], \quad J\equiv t^2/s \ ,\end{aligned} \tag{3.17}$$

$$\underline{S}_l^\pm\equiv\underline{S}_l^x\pm i\underline{S}_l^y \ , \tag{3.18}$$

where J is the pseudoexchange energy and $\underline{S}_l^\delta$ ($\delta=x$, y, and z) is the Pauli spin matrix. The last term in the square brackets in Eq. (3.17) denotes the real transfer of a bipolaron from l to l' through the second-order perturbation of t, while the first two terms denote the virtual transfer. To describe the superconducting state given by $\underline{H}_s$, we define a new effective Hamiltonian $\underline{\tilde{H}}_s$, referenced from the new chemical potential ($\equiv\mu_s$) of this system as

$$\underline{\tilde{H}}_s\equiv\underline{H}_s-\mu_s\left[2\sum_l \underline{S}_l^z\right] , \tag{3.19}$$

and within the mean-field theory, we replace $\underline{S}_l^z$, $\underline{S}_l^x$, and $\underline{S}_l^y$ in $\underline{H}_s$ by their averages as

$$\underline{S}_l^z\rightarrow\sum_l N^{-1}\langle\underline{S}_l^z\rangle=c-\tfrac{1}{2} , \tag{3.20}$$

$$\underline{S}_l^x\rightarrow\sum_l N^{-1}\langle\underline{S}_l^x\rangle=F_s, \quad \underline{S}_l^y=0 \ . \tag{3.21}$$

Here, Eq. (3.20) is nothing but the equation for the new chemical potential μ_s, and it is determined so as to be consistent with the total number of bipolarons. F_s in Eq. (3.21) is the order parameter of superconductivity. By this approximation, $\underline{\tilde{H}}_s$ is reduced as

$$\underline{\tilde{H}}_s\rightarrow-6NJ+24J\sum_l[(c-\tfrac{1}{2}-\mu_s')\underline{S}_l^z-F_s\underline{S}_l^x] , \qquad \mu_s'\equiv\mu_s/12J \ , \tag{3.22}$$

and using this form, we get the following self-consistency equations for μ_s and F_s:

$$c-0.5=-\frac{(c-\frac{1}{2}-\mu_s')}{2[(c-\frac{1}{2}-\mu_s')^2+F_s^2]^{1/2}} \times\tanh\{12\Theta J[(c-\tfrac{1}{2}-\mu_s')^2+F_s^2]^{1/2}\} , \tag{3.23}$$

$$F_s=\frac{F_s}{2[(c-\frac{1}{2}-\mu_s')^2+F_s^2]^{1/2}}\times\tanh\{12\Theta J[(c-\tfrac{1}{2}-\mu_s')^2+F_s^2]^{1/2}\}\ . \tag{3.24}$$

k_BT_c is determined from the following condition:

$$\left.\frac{\partial\langle S_l^x\rangle}{\partial F_s}\right|_{F_s=0}=1\ , \tag{3.25}$$

and its result becomes as

$$\Theta_c^{-1}\equiv k_BT_c/\omega=12J(0.5-c)/\mathrm{arctanh}(1-2c)\ . \tag{3.26}$$

In contrast to the conventional BCS-type mean-field theory, this T_c decreases as s increases, and hence, T_c takes its maximum in the intermediate-coupling region.

IV. FUNCTIONAL INTEGRAL FORMALISM

In order to derive a unified theory which can combine the aforementioned two mean-field theories, in this section we will develop a functional-integral formalism for the one-electron Green's function

$$\underline{G}(\tau-\tau')\ .$$

It has two time variables τ and τ', and has a $2N\times2N$ matrix form in the space spanned by the Wannier functions of electron $|li)$, $(i=1,2)$, with up $(i=1)$ or down $(i=2)$ spin at each lattice site l. Its element can be defined as

$$(ki|\underline{G}(\tau-\tau')|k'i')\equiv-\mathrm{Tr}\{e^{-\Theta\tilde{h}}T_+[A_{ki}(\tau)A_{k'i'}^\dagger(\tau')]\}/\mathrm{Tr}(e^{-\Theta\tilde{h}})\ , \tag{4.1}$$

$$A_{ki}(\tau)\equiv e^{\tau\tilde{h}}A_{ki}e^{-\tau\tilde{h}}\ , \tag{4.2}$$

$$|ki)\equiv\sum_l N^{-1/2}e^{ikl}|li)\ , \tag{4.3}$$

where T_+ is the time-ordering operator.

In order to rewrite Eq. (4.1) into a functional-integral form, we cast $\tilde{h}_0$ in Eq. (2.17) into the following form:

$$\tilde{h}_0\rightarrow h_K+h_I\ , \tag{4.4}$$

where unimportant constant terms are dropped. h_K is given as

$$h_K\equiv\sum_k E_k(A_{k1}^\dagger A_{k1}-A_{k2}^\dagger A_{k2})\ , \tag{4.5}$$

while h_I denotes the attraction with the following quadratic form:

$$h_I\equiv-u\sum_l[(A_{l1}^\dagger A_{l2}+A_{l2}^\dagger A_{l1})^2-(A_{l1}^\dagger A_{12}-A_{l2}^\dagger A_{l1})^2]/4\ . \tag{4.6}$$

Using this form, we can rewrite Eq. (4.1) into the following perturbation expansion form with respect to h_I, h_1, and h_2:

$$(ki|\underline{G}(\tau-\tau')|k'i')=-\mathrm{Tr}\{e^{-\Theta(h_K+h_{\mathrm{ph}})}T_+[Z_0(\Theta)A_{ki}(\tau)A_{k'i'}^\dagger(\tau')]\}/\mathrm{Tr}\{e^{-\Theta(h_K+h_{\mathrm{ph}})}T_+[Z_0(\Theta)]\}\ , \tag{4.7}$$

$$Z_0(\Theta)\equiv\exp\left[-\int_0^\Theta d\tau[h_I(\tau)+h_1(\tau)+h_2(\tau)]\right]\ . \tag{4.8}$$

Here, instead of Eq. (4.2), the time evolution of an operator O is now given as

$$O(\tau)\rightarrow e^{\tau(h_K+h_{\mathrm{ph}})}Oe^{-\tau(h_K+h_{\mathrm{ph}})}\ . \tag{4.9}$$

The time integral of $h_I(\tau)$ in Eq. (4.8) can be formally rewritten into the double-time integral as

$$\begin{aligned}-\int_0^\Theta d\tau\,h_I(\tau)=u\sum_l\int_0^\Theta d\tau\int_0^\Theta d\tau'\delta(\tau-\tau')\{&[A_{l1}^\dagger(\tau)A_{l2}(\tau)+A_{l2}^\dagger(\tau)A_{l1}(\tau)][A_{l1}^\dagger(\tau')A_{l2}(\tau')+A_{l2}^\dagger(\tau')A_{l1}(\tau')]\\&-[A_{l1}^\dagger(\tau)A_{l2}(\tau)-A_{l2}^\dagger(\tau)A_{l1}(\tau)][A_{l1}^\dagger(\tau')A_{l2}(\tau')-A_{l2}^\dagger(\tau')A_{l1}(\tau')]\}/4\ ,\end{aligned} \tag{4.10}$$

and by using the sinusoidal function $f_m(\tau)$,

$$f_m(\tau)\equiv\begin{cases}\sqrt{2/\Theta}\cos(\nu_m\tau), & m\le-1,\quad \nu_m\equiv2\pi m/\Theta,\\ \sqrt{2/\Theta}\sin(\nu_m\tau), & m\ge1,\\ \Theta^{-1/2}, & m=0\ ,\end{cases} \tag{4.11}$$

$\delta(\tau-\tau')$ in Eq. (4.10) can be expanded as

$$\delta(\tau-\tau')=\sum_m f_m(\tau)f_m(\tau'),\quad m=0,\pm1,\pm2,\ldots\ . \tag{4.12}$$

Substituting Eq. (4.12) into Eq. (4.10), we can get a quadratic form of the attraction term

$$-\int_0^\Theta d\tau\,h_I(\tau)=u\sum_{l,m}\left[\left[\int_0^\Theta d\tau f_m(\tau)[A_{l1}^\dagger(\tau)A_{l2}(\tau)+A_{l2}^\dagger(\tau)A_{l1}(\tau)]\right]^2-\left[\int_0^\Theta d\tau f_m(\tau)[A_{l1}^\dagger(\tau)A_{l2}(\tau)-A_{l2}^\dagger(\tau)A_{l1}(\tau)]\right]^2\right]\Bigg/4\ , \tag{4.13}$$

and using the following Gaussian-integral formula for an operator O:

$$e^{O^2/2}=(2\pi)^{-1/2}\int dy\, e^{Oy-y^2/2}\ , \tag{4.14}$$

we can now linearize the quadratic form as

$$T_+\exp\left[-\int_0^\Theta d\tau\, h_I(\tau)\right]=\prod_{l,m,i=1,2}\left[(2\pi)^{-1/2}\int dQ_{lmi}\right]\exp\left[-\sum_{l,m,i=1,2}Q_{lmi}^2/2\right]$$
$$\times T_+\exp\left[\sqrt{u/2}\sum_{l,m}\int_0^\Theta d\tau\, f_m(\tau)[(Q_{lm1}+iQ_{lm2})A_{l1}^\dagger(\tau)A_{l2}(\tau)+\text{H.c.}]\right]\ . \tag{4.15}$$

Thus, the interelectron attraction is rewritten into a linear interaction between electrons and a randomly fluctuating field Q_{lmi} $(i=1,2)$, which is nothing but the real $(i=1)$ or the imaginary $(i=2)$ part of the order parameter at site l with frequency ν_m. From Eq. (4.15), we get the functional-integral form for the Green's function as

$$(ki|\underline{G}(\tau-\tau')|k'i')=-\frac{\prod_{l,m,i=1,2}\left[\int dQ_{lmi}\right]\exp\left[-\sum_{l,m,i}Q_{lmi}^2/2\right]\text{Tr}\{e^{-\Theta(h_K+h_{\text{ph}})}T_+[Z(\Theta)A_{ki}(\tau)A_{k'i'}^\dagger(\tau')]\}}{\prod_{l,m,i=1,2}\left[\int dQ_{lmi}\right]\exp\left[-\sum_{l,m,i}Q_{lmi}^2/2\right]\text{Tr}\{e^{-\Theta(h_K+h_{\text{ph}})}T_+[Z(\Theta)]\}}\ , \tag{4.16}$$

$$Z(\Theta)\equiv\exp\left[-\int_0^\Theta d\tau\left[h_1(\tau)+h_2(\tau)-\sqrt{u/2}\sum_{l,m}f_m(\tau)[(Q_{lm1}+iQ_{lm2})A_{l1}^\dagger(\tau)A_{l2}(\tau)+\text{H.c.}]\right]\right]\ . \tag{4.17}$$

Since this notation for the Green's function is exact, it includes both dynamical and static fluctuations of the order parameter. However, our purpose in the present paper is to look for an interpolation theory that can unify the aforementioned two mean-field theories. For this reason, we take only static (or thermal) fluctuation of the real part of the order parameter, $m=0$ and $i=1$. Hence, we define the static part Q_l as

$$Q_l\equiv Q_{l01}/\sqrt{2\Theta u}\ , \tag{4.18}$$

and in terms of it, the Green's function is rewritten as

$$(ki|\underline{G}(\tau-\tau')|k'i')\to-\frac{\prod_l\left[\int dQ_l\right]\text{Tr}\{e^{-\Theta h_0'(\mathbf{Q})}T_+[Z'(\Theta)A_{ki}(\tau)A_{k'i'}^\dagger(\tau')]\}}{\prod_l\left[\int dQ_l\right]\text{Tr}\{e^{-\Theta h_0'(\mathbf{Q})}T_+[Z'(\Theta)]\}}\ , \tag{4.19}$$

$$Z'(\Theta)\equiv\exp\left[-\int_0^\Theta d\tau[h_1(\tau)+h_2(\tau)]\right]\ . \tag{4.20}$$

Here, $h_0'(\mathbf{Q})$ is the unperturbed Hamiltonian defined by

$$h_0'(\mathbf{Q})\equiv h_K+h_I'(\mathbf{Q})+h_{\text{ph}}+u\sum_l Q_l^2\ , \tag{4.21}$$

and $h_I'(\mathbf{Q})$ denotes the linear interaction between Q_l and electrons,

$$h_I'(\mathbf{Q})\equiv -u\sum_l Q_l(A_{l1}^\dagger A_{l2}+\text{H.c.})\ . \tag{4.22}$$

In these formulas, $\mathbf{Q}$ symbolically denotes an N-dimensional vector

$$\mathbf{Q}\equiv(Q_1,Q_2,\ldots,Q_N)\ , \tag{4.23}$$

and the time evolution of an operator O is given by $h_0'(\mathbf{Q})$ as

$$O(\tau)\to e^{\tau h_0'(\mathbf{Q})}Oe^{-\tau h_0'(\mathbf{Q})}\ . \tag{4.24}$$

Thus, our system is now formally reduced to a one-electron problem under statistically fluctuating random field Q_l. However, the statistical distribution of $\mathbf{Q}$ should be determined self-consistently with the resultant Green's function.

In order to make this self-consistency clear, we define from Eq. (4.19), a componential Green's function

$$\underline{G}(\tau-\tau',\mathbf{Q})$$

for a given configuration of $\mathbf{Q}$. Its element is given as

$$(ki|\underline{G}(\tau-\tau',\mathbf{Q})|k'i')=-\frac{\text{Tr}\{e^{-\Theta h_0'(\mathbf{Q})}T_+[Z'(\Theta)A_{ki}(\tau)A_{k'i'}^\dagger(\tau')]\}}{\text{Tr}\{e^{-\Theta h_0'(\mathbf{Q})}T_+[Z'(\Theta)]\}}\ . \tag{4.25}$$

On the other hand, we can also define the componential thermodynamic potential $\Omega(\mathbf{Q})$ for a given configuration $\mathbf{Q}$,

$$\Omega(\mathbf{Q})\equiv-\Theta^{-1}\ln\text{Tr}\{e^{-\Theta h_0'(\mathbf{Q})}T_+[Z'(\Theta)]\}\ , \tag{4.26}$$

and, in terms of $\underline{G}(\tau-\tau',\mathbf{Q})$ and $\Omega(\mathbf{Q})$ thus defined,

$\underline{G}(\tau-\tau')$ is given as

$$\underline{G}(\tau-\tau')=\prod_l \left[\left(\frac{\Theta u}{\pi}\right)^{1/2}\int dQ_l\right] e^{-\Theta[\Omega(\mathbf{Q})-\Omega]} \times\underline{G}(\tau-\tau',\mathbf{Q}) . \tag{4.27}$$

Here Ω is the thermodynamic potential,

$$\Omega\equiv-\Theta^{-1}\ln\left\{\prod_l \left[\left(\frac{\Theta u}{\pi}\right)^{1/2}\int dQ_l\right] e^{-\Theta\Omega(\mathbf{Q})}\right\} , \tag{4.28}$$

and we can see from Eq. (4.27) that the factor $\exp[-\Theta\Omega(\mathbf{Q})]$ gives the statistical weight of each configuration.

The zero-order Green's function in the expansion in Eq. (4.25) is of great importance, since the polaron effects have already been included in it. This zero-order componential Green's function

$$\underline{G}_0(\tau-\tau',\mathbf{Q})$$

can be defined in the following way:

$$(ki|\underline{G}_0(\tau-\tau',\mathbf{Q})|k'i') \equiv-\frac{\mathrm{Tr}\{e^{-\Theta h_0'(\mathbf{Q})}T_+[A_{ki}(\tau)A^\dagger_{k'i'}(\tau')]\}}{\mathrm{Tr}(e^{-\Theta h_0'(\mathbf{Q})})} , \tag{4.29}$$

and the corresponding zero-order componential thermodynamic potential $\Omega_0(\mathbf{Q})$ can also be defined as

$$\Omega_0(\mathbf{Q})\equiv-\Theta^{-1}\ln\mathrm{Tr}(e^{-\Theta h_0'(\mathbf{Q})}) . \tag{4.30}$$

Since $h_0'(\mathbf{Q})$ is the one-electron Hamiltonian, we can formally rewrite it as

$$\Omega_0(\mathbf{Q})=u\sum_l Q_l^2+\Theta^{-1}N\ln(1-e^{-\Theta}) -\Theta^{-1}\sum_\lambda \ln(1+e^{-\Theta\tilde{E}_\lambda(\mathbf{Q})}) . \tag{4.31}$$

Here, the second term denotes the free energy of phonons, and it will be neglected hereafter, while $\tilde{E}_\lambda(\mathbf{Q})$ is the λth eigenvalue of an electron given by the following equation:

$$[\underline{h}_K+\underline{h}_l'(\mathbf{Q})]|\lambda(\mathbf{Q}))=\tilde{E}_\lambda(\mathbf{Q})|\lambda(\mathbf{Q})) . \tag{4.32}$$

In this formula, $\underline{h}_K$ and $\underline{h}_l'(\mathbf{Q})$ are the one-electron versions of h_K and $h_l'(\mathbf{Q})$;

$$\underline{h}_K\equiv\sum_k \tilde{E}_\lambda(\mathbf{Q})[|k1)(k1|-|k2)(k2|] , \tag{4.33}$$

$$\underline{h}_l'(\mathbf{Q})\equiv-u\sum_l \underline{h}_{ll}'(Q_l) , \tag{4.34}$$

$$\underline{h}_{ll}'(Q_l)\equiv Q_l[|l1)(l2|+|l2)(l1|] , \tag{4.35}$$

and $|\lambda(\mathbf{Q}))$ in Eq. (4.32) is the eigenvector corresponding to $E_\lambda(\mathbf{Q})$. Using $\underline{h}_K$ and $\underline{h}_l'(\mathbf{Q})$, thus defined, we can write the Fourier transform of the zero-order componential Green's function $\underline{G}_0(\tau-\tau',\mathbf{Q})$ as

$$\underline{G}_0(i\omega_m,\mathbf{Q})=\{i\omega_m-[\underline{h}_K+\underline{h}_l'(\mathbf{Q})]\}^{-1} , \qquad \omega_m\equiv\pi(2m+1)/\Theta , \tag{4.36}$$

and Eq. (4.31) can also be rewritten as

$$\Omega_0(\mathbf{Q})=u\sum_l Q_l^2 +\Theta^{-1}\int dE\,\ln(1+e^{-\Theta E})\frac{\mathrm{Im}}{\pi}\mathrm{Tr}[\underline{G}_0(E+i\epsilon,\mathbf{Q})] . \tag{4.37}$$

The zero-order thermodynamic potential Ω_0, the Green's function, and its Fourier component can all be obtained from their componential ones by integrating them over $\mathbf{Q}$ as

$$\Omega_0\equiv-\Theta^{-1}\ln\left\{\prod_l \left[\left(\frac{\Theta u}{\pi}\right)^{1/2}\int dQ_l\right] e^{-\Theta\Omega_0(\mathbf{Q})}\right\} , \tag{4.38}$$

$$\underline{G}_0(\tau-\tau')\equiv\prod_l \left[\left(\frac{\Theta u}{\pi}\right)^{1/2}\int dQ_l\right] e^{-\Theta[\Omega_0(\mathbf{Q})-\Omega_0]} \times\underline{G}_0(\tau-\tau',\mathbf{Q}) , \tag{4.39}$$

$$\underline{G}_0(i\omega_m)\equiv\int_0^\Theta d\tau\,\underline{G}_0(\tau)e^{i\omega_m\tau} , \tag{4.40}$$

where $\exp[-\Theta\Omega_0(\mathbf{Q})]$ is the statistical weight of each componential Green's function. However, as shown in Eq. (4.37), this weight itself depends on the Green's function, and hence all the quantities must be determined self-consistently.

In the context of this functional-integral formalism, let us reformulate the previous BCS-type mean-field theory shown in Eqs. (3.1) and (3.2). In the calculation of Ω_0 using Eqs. (4.21), (4.22), (4.30), and (4.38), we can divide Q_l into its mean value $\bar{Q}$ and the deviation ΔQ_l therefrom as

$$Q_l=\Delta Q_l+\bar{Q} , \tag{4.41}$$

and in terms of ΔQ_l and $\bar{Q}$, Ω_0 is given as

$$e^{-\Theta\Omega_0}=\prod_l \left[\left(\frac{\Theta u}{\pi}\right)^{1/2}\int d\Delta Q_l\right]\exp\left[-u\Theta\sum_l \Delta Q_l^2\right]\mathrm{Tr}\left\{\exp\left[-\Theta\left[\tilde{h}_{\mathrm{MF}}-u\sum_l \Delta Q_l[(A^\dagger_{l1}A_{l2}-\bar{Q})+\mathrm{H.c.}]\right]\right]\right\} . \tag{4.42}$$

Here, $\tilde{h}_{\mathrm{MF}}$ is the same as shown in Eq. (3.3) except constant terms. When the following condition is satisfied:

$$\frac{\mathrm{Tr}[e^{-\Theta\tilde{h}_{\mathrm{MF}}}(A^\dagger_{l1}A_{l2}-\bar{Q})]}{\mathrm{Tr}(e^{-\Theta\tilde{h}_{\mathrm{MF}}})}=0 , \tag{4.43}$$

we can neglect the linear deviation of Q_l from its mean-value $\bar{Q}$, and this condition is equal to Eq. (3.2). Thus, our theory can reproduce the BCS-type mean-field theory.

V. TWO-STATE APPROXIMATION AND COHERENT-POTENTIAL APPROXIMATION

In order to derive a unified theory in the context of the functional-integral form given in the preceding section, let us return to Eq. (4.38) and consider its physical significance in connection with the broken symmetry and the phase transition. It can be rewritten as

$$e^{-\Theta\Omega_0}=\prod_l \left[\int dQ_l P_l(Q_l)\right]\exp\left[-\frac{\mathrm{Im}}{\pi}\int dE\,\mathrm{Tr}[\underline{G}_0(E+i\epsilon,\mathbf{Q})]\ln(1+e^{-\Theta E})\right], \tag{5.1}$$

$$P_l(Q_l)\equiv\left[\frac{\Theta u}{\pi}\right]^{1/2}e^{-\Theta u Q_l^2}. \tag{5.2}$$

The quantity in the large parenthesis in Eq. (5.1) denotes the thermodynamic potential of electrons at a given configuration **Q**, and the summation over all possible configuration with the weight

$$\prod_l P_l(Q_l)$$

gives the total thermodynamic potential. When a broken symmetry occurs, however, the multidimensional configuration space spanned by $Q_1, Q_2, \ldots, Q_N$ will be separated into small regions which are independent of each other. In each region, there is a central configuration around which the local thermodynamic potential takes an extreme value. As for the order parameter, it takes a nonzero value in each local region, although its summation over all local regions becomes zero. How to find such a local region appropriately is a basic problem in the theory for phase transition. However, we can put this problem in a somewhat different way. The summation over **Q** within such a local region can also be described by Eq. (5.1) if we replace $P_l(Q_l)$ by a new weight function $P_l'(Q_l)$ that effectively reflects this conditional summation over **Q**. In this case, $P_l'(Q_l)$ has a polarization in Q_l space so as to give a nonzero order parameter, although the original weight function $P_l(Q_l)$ has not. Thus the problem to find a local region is now converted into the problem to find P_l'. For such $P_l'(Q_l)$, we assume the following simple form:

$$\begin{aligned}&P_l'(Q_l)=e^{-\Theta u Q_l^2}p_l(Q_l)\,,\\&p_l(Q_l)\equiv y_+\delta(Q_l-\bar{Q})+y_-\delta(Q_l+\bar{Q})\,,\\&y_++y_-=1\,,\end{aligned} \tag{5.3}$$

where Q_l takes only two values, $\bar{Q}$ and $-\bar{Q}$ with statistical weights y_+ and y_-, which are unknown at the present stage and will be determined later, self-consistently.

Although this two-state approximation for P' is very simple, it can easily be seen covering the two limiting cases mentioned before. We can get the BCS theory, if we take $y_-=0$ and $y_+=1$, while the aforementioned mean-field theory for bipolarons is nothing but the two-state approximation. Thus, our theory is an interpolation between the two kinds of mean-field theories; the BCS type and the pseudospin type. In this sense, the local order parameter Q_l need not be a complex number. In order to determine this $P_l'(Q_l)$ self-consistently, we also define another distribution function $P_l''(Q_l)$ as

$$P_l''(Q_l)=P_l(Q_l)\prod_{l'(\neq l)}\left[\int dQ_{l'}P_{l'}'(Q_{l'})\right]\exp\left[-\frac{\mathrm{Im}}{\pi}\int dE\,\mathrm{Tr}[\underline{G}_0(E+i\epsilon,\mathbf{Q})]\ln(1+e^{-\Theta E})\right]. \tag{5.4}$$

It gives the polarization at site l, induced by the polarization $P_{l'}'(Q_{l'})$ of its surrounding sites l' ($\neq l$). Since all the sites must be equivalent, the origin of the polarization and the resultant polarization should be equal. Hence, we can assume as

$$P_l''(Q_l)\propto P_l'(Q_l)\,, \tag{5.5}$$

and it gives a self-consistency equation for $y_\pm$.

Let us now consider the nature of this equation in detail. From Eq. (5.4), we define an effective thermodynamic potential $\Omega''(Q_l)$ of a site l under the polarized circumstance as

$$\Omega''(Q_l)\equiv-\Theta^{-1}\ln P_l''(Q_l)\,. \tag{5.6}$$

It can be easily seen that when $T_{\mathrm{temp}}=0$ and $T=0$, $\Omega''(Q_l)$ has two minima with equal depths at $Q_l=\pm\bar{Q}$, since the site l is not suffered by the polarization of surrounding sites even if we take $y_-=0$ in Eq. (5.3). As T increases, however, the two depths become unequal as schematically shown by curves *a* and *b* in Fig. 1, since the electrons at site l can move to l' and see the polarization therein. If T increases further, it will finally become a single-well curve like *c*. In this single-well situation, the original assumption for $P_l'(Q_l)$ described by Eq. (5.3) is of no sense, and Eq. (5.5) cannot give a self-consistent solution except $y_-=0$. Thus, we must return to the BCS-type simple solution when the two points $Q_l=\bar{Q}$ and $Q_l=-\bar{Q}$ are not separated by a potential barrier. This is the limitation for us to use Eq. (5.5).

Even if we restricted ourselves within this simplified distribution of $P_l'(Q_l)$, we, in the next step, have to perform the integral over Q_l in Eq. (5.1), which can be rewritten as

$$e^{-\Theta\Omega_0}=e^{-\Theta uN\bar{Q}^2}\left\langle\!\!\left\langle \exp\left[-\frac{\mathrm{Im}}{\pi}\int dE\,\mathrm{Tr}[\underline{G}_0(E+i\epsilon,\mathbf{Q})]\ln(1+e^{-\Theta E})\right]\right\rangle\!\!\right\rangle , \tag{5.7}$$

$$\langle\!\langle \cdots \rangle\!\rangle=\prod_l \left[\int dQ_l p_l(Q_l)\right]\cdots . \tag{5.8}$$

In order to perform this average over Q_l, we take the following two approximations: the neglect of higher-order cumulants more than the first, and the coherent-potential approximation. Within the first approximation, we get as

$$\Omega_0 \to uN\bar{Q}^2 + \Theta^{-1}\int dE\,\ln(1+e^{-\Theta E})\frac{\mathrm{Im}}{\pi}\mathrm{Tr}\langle\!\langle \underline{G}_0(E+i\epsilon,\mathbf{Q})\rangle\!\rangle . \tag{5.9}$$

This approximation gives the exact result in the strong-coupling limit ($T=0, S\neq 0$), and also in the weak-coupling limit ($T\neq 0, S=0$), and hence it acts as an interpolation in intermediate cases. The second approximation is to replace the randomly fluctuating part $\underline{h}'_{ll}(Q_l)$ of the Green's function by a complex self-energy $\underline{V}_l(E+i\epsilon)$, called the coherent potential,[16]

$$\underline{h}'_{ll}(Q_l)\to \underline{V}_l(E+i\epsilon) , \tag{5.10}$$

$$\underline{V}_l(E+i\epsilon)\equiv \sum_{ii'} V_{ii'}(E+i\epsilon)|li)(li'| , \tag{5.11}$$

where $V_{ii'}$ is the element of $\underline{V}_l$, assumed to be independent of l. The averaged Green's function $\underline{G}_C(z)$ within this approximation is given as

$$\langle\!\langle \underline{G}_0(z,\mathbf{Q})\rangle\!\rangle \to \underline{G}_C(z)\equiv \left[z-\left[\underline{h}_K-u\sum_l \underline{V}_l\right]\right]^{-1} ,$$

$$z=i\omega_m \text{ or } E+i\epsilon . \tag{5.12}$$

This self-energy is determined under the condition that the single-site scattering due to this replacement Eq. (5.10) is zero,

$$\underline{G}_{Cl}(E+i\epsilon)=\langle\!\langle \underline{G}_{Cl}(E+i\epsilon)/\{1+u[\underline{h}'_{ll}(Q_l)-\underline{V}_l(E+i\epsilon)]\underline{G}_{Cl}(E+i\epsilon)\}\rangle\!\rangle , \tag{5.13}$$

where $\underline{G}_{Cl}$ is a 2×2 matrix of $\underline{G}_C$ at a site l,

$$\underline{G}_{Cl}(E+i\epsilon)\equiv \sum_{ii'} |li)(li|\underline{G}_C(E+i\epsilon)|li')(li'| . \tag{5.14}$$

As shown in our previous paper,[5] in the practical calculations we have to solve the following nonlinear equations for $V_{ii'}$:

$$V_{ii}=u[(V_{ii'}V_{i'i}-\bar{Q}^2)G_{i'i'}+v_{ii}^2G_{ii} + V_{ii}(G_{ii'}V_{i'i}+G_{i'i}V_{ii'})] , \tag{5.15}$$

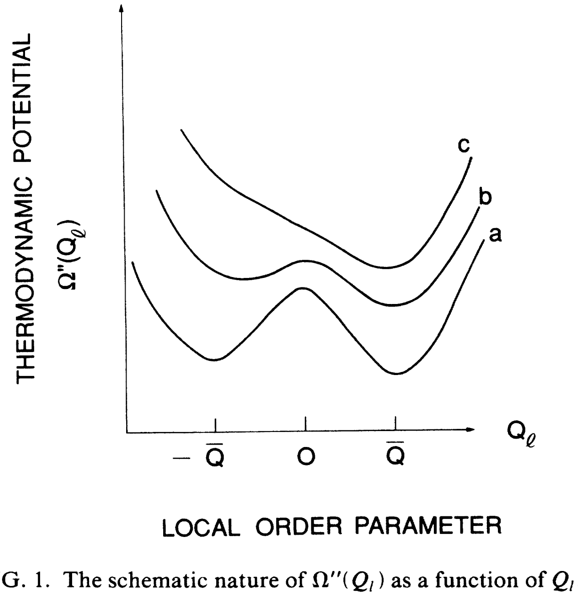

FIG. 1. The schematic nature of $\Omega''(Q_l)$ as a function of Q_l.

$$V_{ii'}=\tilde{Q}+u[(V_{ii'}^2-\bar{Q}^2)G_{i'i}+V_{ii}V_{i'i'}G_{ii'} + V_{ii'}(G_{ii}V_{ii}+G_{i'i'}V_{i'i'})] , \tag{5.16}$$

$$\tilde{Q}\equiv(y_+-y_-)\bar{Q},\quad (i,i')=(1,2) \text{ or } (2,1) \tag{5.17}$$

where $G_{ii'}$ in Eqs. (5.15), (5.16), and (5.17) denotes the element of $\underline{G}_{Cl}$ given by Eq. (5.14).

As for the average over $Q_{l'}$ ($l'\neq l$) in Eq. (5.4), we also use the same two approximations, and can get as

$$P_l''(Q_l)\to P_l(Q_l) \times\exp\left[\int dE\,\ln(1+e^{-\Theta E}) \times\left[-\frac{\mathrm{Im}}{\pi}\mathrm{Tr}\langle\!\langle \underline{G}_0(E+i\epsilon,\mathbf{Q})\rangle\!\rangle_l\right]\right] , \tag{5.18}$$

$$\langle\!\langle \cdots \rangle\!\rangle_l\equiv \prod_{l'(\neq l)} \left[\int dQ_{l'}p_{l'}(Q_{l'})\right]\cdots , \tag{5.19}$$

$$\langle\!\langle \underline{G}_0(E+i\epsilon,\mathbf{Q})\rangle\!\rangle_l \to \underline{G}_C/\{1+u[\underline{h}'_{ll}(Q_l)-\underline{V}_l]\underline{G}_C\} . \tag{5.20}$$

Here the average over $Q_{l'}$ at surrounding sites l' ($\neq l$) is to replace $\underline{h}'_{ll'}$ by $\underline{V}_{l'}$. After the integration by parts over E in Eq. (5.18), we can get a well-known form for the thermodynamic potential as

$$\int dE \ln(1+e^{-\Theta E})\left[-\frac{\mathrm{Im}}{\pi}\mathrm{Tr}\langle\langle \underline{G}_0\rangle\rangle_l\right]=-\Theta\int dE\, n(E)\left[\frac{\mathrm{Im}}{\pi}\mathrm{Tr}(\ln\langle\langle \underline{G}_0\rangle\rangle_l)\right],\quad n(E)=(e^{\Theta E}+1)^{-1}, \tag{5.21}$$

and using this result, we get our final equation for $y_\pm$ as

$$y_-/y_+=\exp\left[-\Theta\frac{\mathrm{Im}}{\pi}\int dE\, n(E)\mathrm{Tr}\left[\ln\frac{\{1+u[\underline{h}'_{ll}(-\bar{Q})-\underline{V}_l]\underline{G}_{Cl}\}}{\{1+u[\underline{h}'_{ll}(\bar{Q})-\underline{V}_l]\underline{G}_{Cl}\}}\right]\right], \tag{5.22}$$

wherein the trace of the 2×2 matrix becomes

$$\begin{aligned}\mathrm{Tr}(\ln\{1+u[\underline{h}'_{ll}(\pm\bar{Q})-\underline{V}_l]\underline{G}_{Cl}\})=\ln\{&1-u[V_{11}G_{11}+V_{22}G_{22}+(V_{12}\mp\bar{Q})G_{21}+(V_{21}\mp\bar{Q})G_{12}]\\&+u^2[V_{11}V_{22}-(V_{12}\mp\bar{Q})(V_{21}\mp\bar{Q})(G_{11}G_{22}-G_{21}G_{12})]\}\ .\end{aligned} \tag{5.23}$$

The transition temperature T_c is determined from Eq. (5.22) as the highest temperature that can give a nontrivial solution,

$$y_+\neq y_-\ . \tag{5.24}$$

Let us now see how this theory can reproduce the aforementioned two mean-field theories shown in Sec. III. Since we have already mentioned the BCS-type theory occasionally, we will now examine our result only in the strong-coupling and the inverse-adiabatic limits, $1\gg s\gg t$, in comparison with Eq. (3.26). In this case we can neglect t as the zero-order approximation and get two levels at each site, as schematically shown in Fig. 2. When $Q_l=\bar{Q}$, we denote these two levels $|+)$ and $|-)$, and they are given as

$$|+)=\sqrt{c}\,|l1)+\sqrt{1-c}\,|l2)\ , \tag{5.25}$$

$$|-)=-\sqrt{1-c}\,|l1)+\sqrt{c}\,|l2)\ . \tag{5.26}$$

In this calculation, we have used the limiting values of q, μ, and F, which are shown in Eqs. (3.14), (3.15), and (3.16). When $Q_{l'}=\bar{Q}$ $(l'\neq l)$, we also have the same two levels in the surrounding sites, while, when $Q_{l'}=-\bar{Q}$, we get another two levels denoted $|+)'$ and $|-)'$. They are given as

$$|-)'=-\sqrt{c}\,|l'1)+\sqrt{1-c}\,|l'2)\ , \tag{5.27}$$

$$|+)'=\sqrt{1-c}\,|l'1)+\sqrt{c}\,|l'2)\ . \tag{5.28}$$

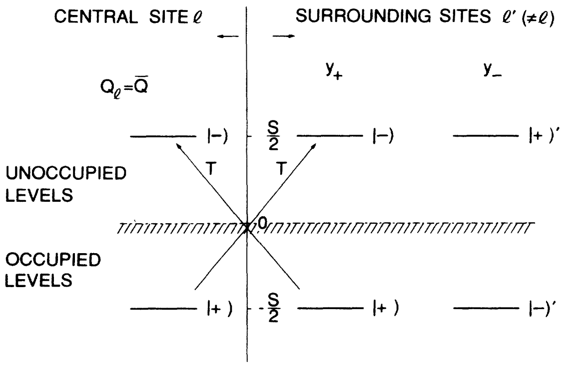

FIG. 2. Localized energy levels at each site, $T=0$, $Q_l=\bar{Q}$. y_+ and y_- are the statistical weights of each level. l is the relevant site and l' is its environmental sites. The long arrows denote the mixing between levels through T.

Using this energy-level scheme, we can calculate the energy gain due to the second-order perturbation of t, which causes the virtual intersite mixing between occupied levels and unoccupied levels, as shown in Fig. 2. We can easily see that the mixing between $|+)$ and $|+)'$ is zero, because the up-spin electrons and the down-spin ones have opposite signs of the transfer with each other in Eq. (2.17). In taking the average over $Q_{l'}$, we can see that these two sets of levels exist with a statistical weight y_+ and y_- as shown in Fig. 2, and using this result we can calculate the total energy gain due to the second-order perturbation of t. When $Q_l=-\bar{Q}$, the situation is entirely the same, and we finally get from Eq. (5.22),

$$y_\pm\propto e^{24J\Theta\sqrt{c(1-c)}y_\pm}\ . \tag{5.29}$$

It gives a T_c defined by

$$\Theta_c^{-1}\equiv k_BT_c/\omega=12J\sqrt{c(1-c)}\ . \tag{5.30}$$

Equations (5.30) and (3.26) give almost same results as shown in Fig. 3, and they are equal when $c=0.5$ as shown in our previous paper.[5]

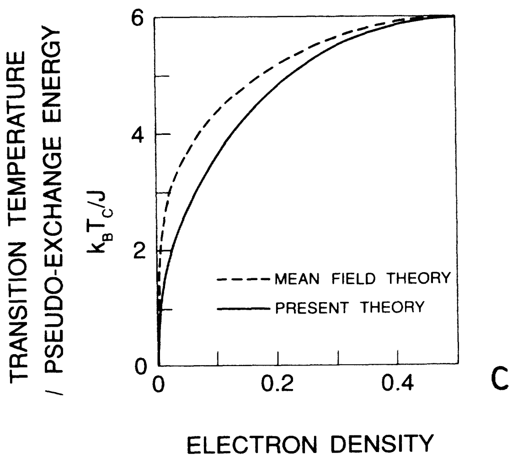

FIG. 3. T_c as a function of the electron density c $(=N_e/2N)$ in the inverse-adiabatic and strong-coupling limits, $\omega\gg S\gg T$. $J\equiv T^2/S\omega$. The dashed line is the mean-field theory: Eq. (3.26).

VI. RESIDUAL INTERACTIONS

As the effective Hamiltonian for our many-polaron system, we have used $\tilde{h}_0$ instead of $\tilde{h}$, neglecting h_1 and h_2 given by Eq. (2.20) and (2.21). Therefore, in the weak-coupling limit, $s \ll 1 \ll t$, our theory does not coincide with the Eliashberg equation,[15] which is based on the second-order perturbation with respect to s.

In order to remedy this apparent defect, we take the effects of h_1 into account by the perturbation, using the present framework of the theory. In this case, the order parameter $\langle A_{l1}^{\dagger} A_{l2} \rangle$, the thickness of the phonon cloud q, and the chemical potential μ will all change from their old values. However, the effects of h_1 become zero in the strong-coupling limit because of the factor $(1-q)$ included in it, and hence, we assume such change occurs only in the order parameter $\bar{Q}$ without any changes of q and μ.

Therefore, at first we calculate this new value $(\equiv \bar{Q}')$ of $\langle A_{l1}^{\dagger} A_{l2} \rangle$ within the conventional BCS theory. For this aim, we will expand the Green's function given by Eq. (4.7) straightforwardly with respect to the attraction h_I and h_1 as

$$
\begin{aligned}
(ki|\underline{G}(\tau-\tau')|k'i') = {} & (ki|\underline{G}_{00}(\tau-\tau')|k'i') \\
& + \sum_{l} \int_0^{\Theta} d\tau_1 (ki|\underline{G}_{00}(\tau-\tau_1)|li)(li'|\underline{G}_{00}(\tau_1-\tau')|k'i')[-u(1-\delta_{ii'})(li|\underline{G}(+0)|li')] \\
& + \sum_{l} \int_0^{\Theta} d\tau_1 \int_0^{\Theta} d\tau_2 (ki|\underline{G}_{00}(\tau-\tau_2)|li)(li'|\underline{G}_{00}(\tau_1-\tau')|k'i') \\
& \times \left[-\frac{s}{2}(1-q)^2(1-\delta_{ii'})(li|\underline{G}(\tau_2-\tau_1)|li')D(\tau_2-\tau_1) \right] + \cdots ,
\end{aligned} \tag{6.1}
$$

where G_{00} is the Green's function given only by h_K, and the second term denotes the first-order perturbation of the attraction. The third term is the second-order perturbation of h_1, wherein $D(\tau_2-\tau_1)$ denotes the phonon Green's function

$$
D(\tau_2-\tau_1) \equiv \frac{\mathrm{Tr}(e^{-\Theta h_{\mathrm{ph}}} T_+ \{[d_l^{\dagger}(\tau_2)+d_l(\tau_2)][d_l^{\dagger}(\tau_1)+d_l(\tau_1)]\})}{\mathrm{Tr}(e^{-\Theta h_{\mathrm{ph}}})} . \tag{6.2}
$$

In the above expansion, we have taken into account only the off-diagonal part of the interaction and the diagonal part is neglected, since the polaron effect has already been included in h_K. As for higher-order terms not written explicitly in Eq. (6.1), we take only the diagrams which are reducible into these lowest two terms. Taking the Fourier component of this equation, we can get the Green's function $\underline{G}_{\mathrm{MF}}$ based on the BCS-type mean-field theory instead of the exact Green's function $\underline{G}$ as

$$
\underline{G}(i\omega_m) \to \underline{G}_{\mathrm{MF}}(i\omega_m) \equiv [i\omega_m - \underline{h}_K - \underline{W}_{\mathrm{MF}}(i\omega_m)]^{-1} , \tag{6.3}
$$

where $\underline{W}_{\mathrm{MF}}$ denotes the off-diagonal self-energy, coming from the two origins shown in Eq. (6.1), and it is defined as

$$
\begin{aligned}
\underline{W}_{\mathrm{MF}}(i\omega_m) \equiv {} & \sum_{l,i,i'} |li)(1-\delta_{ii'}) \\
& \times [W_{ii'}^{A}(i\omega_m) + W_{ii'}^{B}(i\omega_m)](li'| .
\end{aligned} \tag{6.4}
$$

$W_{ii'}^{A}$ comes from the second term of Eq. (6.1) and is given as

$$
W_{ii'}^{A}(i\omega_m) \equiv -u\Theta^{-1}N^{-1} \sum_{l,m'} e^{+i\omega_{m'}0}(li|\underline{G}_{\mathrm{MF}}(i\omega_{m'})|li') , \tag{6.5}
$$

while $W_{ii'}^{B}$ comes from the third term of Eq. (6.1) and is given as

$$
\begin{aligned}
W_{ii'}^{B}(i\omega_m) = {} & -\frac{s}{2}(1-q)^2\Theta^{-1}N^{-1} \\
& \times \sum_{l,m'} D(i\nu_{m-m'})(li|\underline{G}_{\mathrm{MF}}(i\nu_{m'})|li') , \\
D(i\nu_m) \equiv {} & (1+i\nu_m)^{-1} + (1-i\nu_m)^{-1} .
\end{aligned} \tag{6.6}
$$

Here $D(i\nu_m)$ is the Fourier component of $D(\tau)$. Equations (6.3)–(6.6) form a set of simultaneous equations, since $W_{ii'}^{A,B}$ is given by $\underline{G}_{\mathrm{MF}}$ itself, and if $W_{ii'}^{B}=0$, our theory becomes equal to the previous BCS-type mean-field theory. In order to perform the summation over m' in Eqs. (6.5) and (6.6), we define the imaginary part of the off-diagonal element as

$$
\rho_{ii'}^{\mathrm{MF}}(E) \equiv -\frac{\mathrm{Im}}{\pi}(li|\underline{G}_{\mathrm{MF}}(E+i\epsilon)|li') , \tag{6.7}
$$

and in terms of it, we can formally rewrite $\underline{G}_{\mathrm{MF}}$ as

$$
(li|G_{\mathrm{MF}}(i\omega_m)|li') = \int dE' \frac{\rho_{ii'}^{\mathrm{MF}}(E')}{i\omega_m - E'} . \tag{6.8}
$$

We substitute this new form into Eqs. (6.5) and (6.6), and can get as

$$W^A_{ii'}=-u\int dE'\rho^{\mathrm{MF}}_{ii'}(E')n(E')\ , \tag{6.9}$$

$$\begin{aligned}W^B_{ii'}(E+i\epsilon)=&-\frac{S}{2}(1-q)^2\\&\times\int dE'\rho^{\mathrm{MF}}_{ii'}(E')\\&\times\left[\frac{1-n(E')}{E+i\epsilon-(E'+1)}\right.\\&\left.+\frac{n(E')}{E+i\epsilon-(E'-1)}\right]\ ,\end{aligned} \tag{6.10}$$

which are essentially same as the Eliashberg equation when $q \ll 1$. In practical calculations, we can use the following form of the off-diagonal element:

$$\begin{aligned}(li|G_{\mathrm{MF}}(E+i\epsilon)|li')&=\sum_k N^{-1}(W^A_{ii'}+W^B_{ii'})/2Y'_k\\&\quad\times[(E+i\epsilon-Y'_k)^{-1}-(E+i\epsilon+Y'_k)^{-1}]\ ,\\Y'_k&\equiv[E_k^2+(W^A_{12}+W^B_{12})(W^A_{21}+W^B_{21})]^{1/2}\ ,\end{aligned} \tag{6.11}$$

and using these results, we finally get the new value $\bar{Q}'$ of $\langle A^\dagger_{l1}A_{l2}\rangle$ as

$$\bar{Q}'=-u\int dE'[\rho^{\mathrm{MF}}_{12}(E')+\rho^{\mathrm{MF}}_{21}(E')]n(E')/2\ . \tag{6.12}$$

As mentioned occasionally, this value becomes different from $\bar{Q}$ only in the weak-coupling case, since this difference comes from h_1.

Next let us take into account the effects of thermal fluctuation of this order parameter by the method developed in the previous sections. Similar to Eq. (5.3), we approximate that Q_l fluctuates only between two values $\bar{Q}'$ and $-\bar{Q}'$, with the weight y'_+ and y'_- ($y'_+ + y'_- = 1$). In this case, the Green's function is also given by the average of the componential one given by Eq. (4.25). It is written as

$$\underline{G}(\tau-\tau')=\langle\langle G(\tau-\tau',\mathbf{Q})\rangle\rangle'\ , \tag{6.13}$$

where $\langle\langle\cdots\rangle\rangle'$ denotes the above-mentioned new average of $\cdots$ over Q_l, similar to Eq. (5.8). By expanding this Green's function with respect h_1, we get

$$\begin{aligned}(ki|\underline{G}(\tau-\tau')|k'i')&=-\langle\langle\mathrm{Tr}\{e^{-\Theta h'_0(\mathbf{Q})}T_+[A_{ki}(\tau)A^\dagger_{k'i'}(\tau')]\}/\mathrm{Tr}(e^{-\Theta h'_0(\mathbf{Q})})\rangle\rangle'\\&\quad-\tfrac{1}{2}\int_0^\Theta d\tau_1\int_0^\Theta d\tau_2\langle\langle\mathrm{Tr}\{e^{-\Theta h'_0(\mathbf{Q})}T_+[A_{ki}(\tau)h_1(\tau_1)h_1(\tau_2)A^\dagger_{k'i'}(\tau')]\}/\mathrm{Tr}(e^{-\Theta h'_0(\mathbf{Q})})\rangle\rangle'+\cdots\ ,\end{aligned} \tag{6.14}$$

where the first term denotes the free propagation in this random fluctuation and the second term denotes the second-order scattering due to h_1. To be consistent with Eq. (6.1), we take only the off-diagonal element of this second-order effect, and as for the free propagation we replace them by the Green's function given by the CPA as

$$\begin{aligned}(ki|\underline{G}(\tau-\tau')|k'i')&=(ki|\underline{G}'_C(\tau-\tau')|k'i')+\sum_l\int_0^\Theta d\tau_1\int_0^\Theta d\tau_2(ki|\underline{G}'_C(\tau-\tau_2)|li)(li'|\underline{G}'_C(\tau_1-\tau')|k'i')\\&\quad\times\left[-\frac{S}{2}(1-q)^2(1-\delta_{ii'})(li|\underline{G}(\tau_2-\tau_1)|li')D(\tau_2-\tau_1))\right]+\cdots\ ,\end{aligned} \tag{6.15}$$

where G'_C is the Green's function given by the new CPA. It should be noted that this G'_C is not equal to G_C of the previous section, since the distribution of Q_l has now become different from the previous section. We can denote it in terms of the new self-energy $V'_{ii'}$ as

$$\underline{G}'_C(z)=\left[z-\left[\underline{h}_K-u\sum_l\underline{V}'_l(z)\right]\right]^{-1}\ , \tag{6.16}$$

$$\underline{V}'_l(z)\equiv\sum_{i,i'}V'_{ii'}(z)|li)(li'|\ . \tag{6.17}$$

This self-energy $V'_{ii'}$ must be determined by a similar equation as that of Eq. (5.13), and it will be mentioned later. After taking the Fourier transform of Eq. (6.15), we can get a new CPA Green's function $\underline{G}_{CC}$ instead of the exact one $\underline{G}$ as

$$\underline{G}(i\omega_m)\rightarrow\underline{G}_{CC}(i\omega_m)\equiv[\underline{G}'_C(i\omega_m)^{-1}-\underline{W}_c(i\omega_m)]^{-1}\ , \tag{6.18}$$

$$\underline{W}_c(i\omega_m)\equiv\sum_{l,i,i'}|li)(1-\delta_{ii'})W^c_{ii'}(i\omega_m)(li'|\ , \tag{6.19}$$

where $W^c_{ii'}$ is the off-diagonal element coming from the second term of Eq. (6.15) and is defined by

$$\begin{aligned}W^c_{ii'}(i\omega_m)\equiv&-\frac{S}{2}(1-q)^2\Theta^{-1}N^{-1}\\&\times\sum_{m',l}D(i\nu_{m-m'})(li|G_{CC}(i\omega_{m'})|li')\ .\end{aligned} \tag{6.20}$$

Similar to Eq. (6.3) given by the mean-field theory, Eqs. (6.18), (6.19), and (6.20) form a set of simultaneous equations, and can be solved by the same method as shown in Eqs. (6.7), (6.8), (6.9), and (6.10).

In this case, however, equations for $y'_\pm$ and $V'_{ii'}$ must also be solved simultaneously, and these equations are essentially the same as Eqs. (5.22) and (5.13), if we replace $\underline{G}_C$ and $\underline{V}$ by G_{CC} and $\underline{V}'$, respectively. It will be not necessary for us to repeat them again. We have thus obtained a new theory that can interpolate two mean-field theories: the BCS-type and the pseudospin type, and this new theory also becomes identical to the Eliashberg theory in the weak-coupling limit, because in this case we get $q \ll 1$.

We can also take the effects of h_2 into account, according to the similar principle given above. However, this term gives only small effects, since in both limits of weak and strong couplings it becomes zero.

VII. NUMERICAL RESULTS

Let us now see numerical results obtained by the theory developed in previous sections. In the inverse adiabatic case, $T/\omega < 1$, we cannot get any new results other than reported in our previous paper.[5] In this case, we get $q \approx 1$ irrespective of the ratio s/t, and hence h_1 is always zero. In our previous paper, however, we were concerned with only the N-site N-electron system ($c = 0.5$). In the present paper, we could clarify the c dependence of T_c. Its result in the strong-coupling limit, $s/t \gg 1$, is shown in Fig. 3. From this figure, we can see that the pseudospin type mean-field theory and our unified theory give almost the same results, although they are not exactly the same except for the case $c = 0.5$.

On the other hand, in the case of a rather large but finite value of T/ω, we get a discontinuous increase of q from a small value to 1, as s increases. As shown in Fig. 4, in the weak region, it is about 0.1 because of the Migdal's theorem; however, it increases abruptly to 1. This is nothing but the transition from the BCS-type state to the bipolaronic state. The origin of this change in the thickness of the phonon cloud is essentially the same as that of the one-body problem of the electron, as mentioned in Sec. I. However, the critical value of S/T is smaller than that of the one-body case because the total energy gain due to the e-ph coupling has a many-body nature. In fact, the critical value of S/T increases as c decreases, as shown in Fig. 5. In connection with such c dependence, we can also see from Fig. 5 that the back transition from the bipolaronic state to the BCS state occurs as we decrease c, keeping S constant. We will return to this point later.

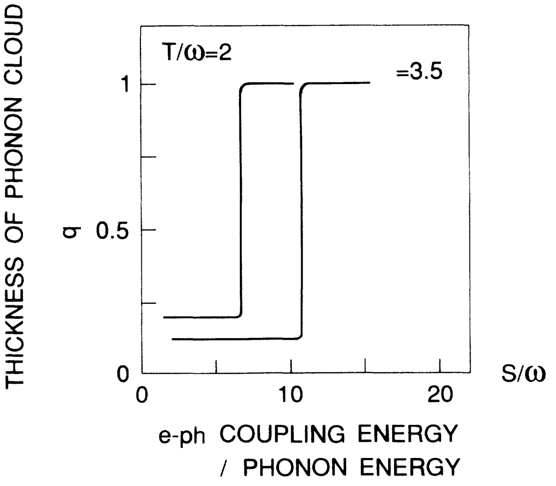

FIG. 4. Thickness of the phonon cloud q as a function of S. $c = 0.3$. $T/\omega = 2$ and 3.5.

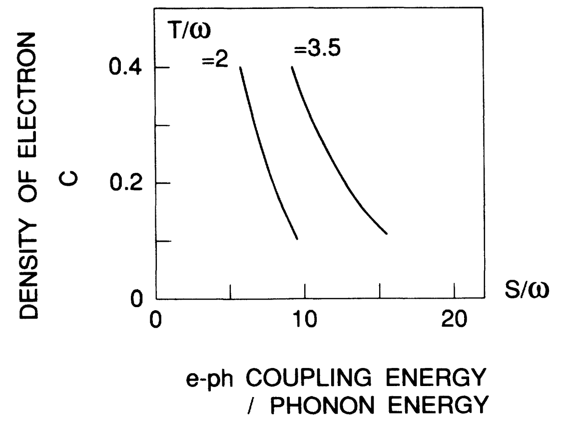

FIG. 5. The phase boundary between the BCS region and the bipolaronic region as a function of the electron density c and S. $T/\omega = 2$ and 3.5.

Figure 6 shows T_c as a function of S/ω at $c = 0.3$. In

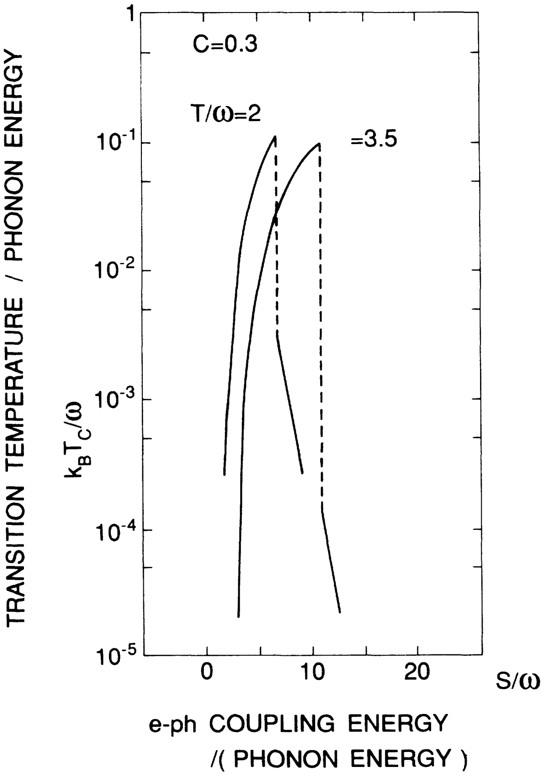

FIG. 6. T_c as a function of S. $c = 0.3$. $T/\omega = 2$ and 3.5.

the BCS region, T_c increases rapidly as S/ω increases, while it decreases as S/ω increases in the strong-coupling region. The transition between two regions is almost discontinuous and in the case of $T/\omega \approx 3.5$, T_c decreases in its two orders of magnitude. As seen from the difference between the case $T/\omega = 3.5$ and that of $T/\omega = 2.5$, this discontinuity decreases as T/ω decreases. This comes mainly from the reason that the retardation effect decreases as T/ω decreases.

In order to compare these theoretical results to the newly found high-temperature superconductors of transition metal oxides,[8-12] let us discuss the nature of the bipolaronic state. If we assume that the full bandwidth $12T$ is 4 eV and the phonon energy ω is 0.08 eV, we get $T/\omega = 3.5$. It is a typical case realized in these metal-oxides materials, and in this case, T_c of the bipolaronic state becomes too small to be observed. Thus, we can observe only the bipolaronic state above T_c, wherein there is no coherence between bipolarons, although the pairing within a single bipolaron is still well established. Such a state is usually called the charge glass or the bipolaron glass in analogy with the spin glass. It has no paramagnetic susceptibility, since the spin has already disappeared by the pairing. It also has no long-range spatial order of charge, but has an optical gap in the one-body excitation spectrum which corresponds to the pair breaking. The absence of long-range order allows us to distinguish this state from the CDW state. As for the electric conductivity, it will give small value, because the bipolaron is heavy.

According to the recent experimental studies[8-11] on $Ba_{1-x}K_xBiO_3$, the "insulating state" of this material in the region $x \approx 0.35$ has no long-range CDW-type order that can result in the strong mixed valence of Bi. However, it has a large optical gap in the visible region. On the other hand, when x becomes larger, $x > 0.375$, the electrons of this material are in the BCS-type superconducting state with T_c of 30 K. The increase of x in this material just corresponds to the decrease of the electron number in the 6s band of Bi, and $x = 0.375$ is nearly equal to the case $c = 0.3$ of our theory. Thus, the insulator-superconductor transition of this material is one of possible candidates for the bipolaron-glass to BCS transition predicted in Figs. 4–6.

VIII. CONCLUSION AND DISCUSSION

As mentioned occasionally, we have shown that T_c discontinuously changes from the BCS region to the bipolaronic region as the e-ph coupling s increases, provided the ratio T/ω is large. It is also shown that the back transition occurs as we decrease the electron number c.

Because of simplicity, the CDW-type instability is excluded from the beginning, and hence we have to clarify the relative stability between the CDW state and the bipolaronic state. It is our theme for future study. However, we can say that there will be many cases wherein the bipolaronic state becomes more stable than the CDW state, depending on the crystal structure, the dispersion relation of the electron energy band, the energy of the phonon, and the electron concentration. The presence of randomness and other inhomogeneous natures of crystals are also expected to favor the bipolaronic state rather than the CDW state.

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[1] Y. Toyozawa, in *Highlights of Condensed Matter Theory,* proceedings of the International School of Physics "Enrico Fermi," Course XXXX, edited by F. Bassani, F. Fumi, and M. Tosi (North-Holland, Amsterdam, 1985), p. 798.

[2] J. Shrieffer, *Theory of Superconductivity* (Benjamin, New York, 1964).

[3] P. Peierls, *Quantum Theory of Solids* (Oxford, New York, 1955), p. 108.

[4] K. Nasu, Phys. Rev. B **35**, 1748 (1987).

[5] K. Nasu, Phys. Rev. B **37**, 5075 (1988).

[6] A. Alexandrov and J. Ranninger, Phys. Rev. B **23**, 1796 (1981); **24**, 1164 (1981); A. Alexandrov, J. Ranninger, and S. Robaszkiewics, *ibid.* **33**, 4526 (1986).

[7] D. Johnston, J. Low Temp. Phys. **25**, 145 (1976).

[8] H. Sato, S. Tajima, H. Takagi, and S. Uchida, Nature (London) **338**, 241 (1989).

[9] S. Kondo, M. Sera, Y. Ando, and M. Sato, Physica C **157**, 469 (1989).

[10] S. Pei, J. Jorgensen, D. Hinks, B. Dabrowski, D. Richards, A. Mitchell, Y. Zheng, J. Newsam, S. Sinha, D. Varkin, and A. Jacobson, Physica C **162–164**, 556 (1989).

[11] G. Wertheim, J. Remeika, and D. Buchanan, Phys. Rev. B **26**, 2120 (1982).

[12] J. Bednorz and K. Müller, Z. Phys. B **64**, 199 (1986).

[13] C. Shelenker, *Physics of Disordered Materials,* edited by D. Adler, H. Fritzsche, and S. Ovshinsky (Plenum, New York, 1985), p. 369.

[14] M. Onoda and H. Nagasawa, J. Phys. Soc. Jpn. **52**, 223 (1983).

[15] G. Eliashberg, Zh. Eksp. Teor. Fiz. **38**, 966 (1960) [Sov. Phys.–JETP **11**, 696 (1960)].

[16] P. Soven, Phys. Rev. **156**, 809 (1967).