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Superconducting transition temperatures of strongly coupled electron-boson systems: A self-consistent method for fluctuation from the Bardeen-Cooper-Schrieffer limit to the bipolaronic limit

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Superconducting transition temperatures T_c of a many-electron system coupling strongly with bosonic excitations are studied in the context of polaron theory. The boson clouds that move with electrons are taken into account within the variational method. The effect of fluctuation of the pairing order is also taken into account by the coherent-potential approximation. The resultant theory covers the whole region of the three basic parameters characterizing this system: the intersite transfer energy of the electron, T, the site-diagonal electron-boson coupling energy, S, and the energy of the boson, ω. T_c is found to become maximum in the transition region $S \approx T$, which lies between the weak (Bardeen-Cooper-Schrieffer) region $S \ll T$ and the strong (bipolaronic) region $S \gg T$. The width of this region is narrow in the adiabatic case $\omega \ll T$, but wide in the inverse-adiabatic case $\omega \gg T$. This region is characterized by the large ratio of the energy gap to $k_B T_c$. These results are applied to the Cu-O–type new ceramics, and the breathing mode of oxygen is shown to be able to give a high T_c of about 100 K. The effects of other quasibosons with high frequencies, such as plasmons and excitons, are also studied.

I. INTRODUCTION

An electron, coupling strongly with quasibosonic excitations such as phonons, plasmons, or excitons in a crystal, forms a polaron,[1] composed of the original electron and the boson cloud that moves along with the electron. In the case of phonons, we usually call it a lattice polaron, while in the case of quasibosons of electronic origins such as excitons and plasmons, we usually call it an electronic polaron. In both cases, the boson cloud enhances the effective mass of the polaron. Between two polarons an attraction acts through the overlap of their boson clouds, and if it is strong enough to exceed the direct Coulombic repulsion between two polarons, we can get a bound state called a singlet bipolaron.[2] Such singlet bipolarons, once formed, can give rise to the conductive charges with no spin already observed in amorphous solids[3] and conducting polymers.[4] Thus, the one- and two-body natures of the polaron have already been well clarified, and the purpose of the present paper is to extend these concepts to many-body systems.[5] As is well known, a metallic state of a many-electron system is always unstable with respect to the electron-boson coupling and becomes a superconducting state.[6]

Generally speaking, the superconducting transition temperature T_c of the electron-boson coupled system is mainly determined by three basic parameters, the intersite transfer energy of electron, T, the electron-boson (e-b) coupling energy S, and the energy quantum of bosonic excitation ω. At present, however, we have no systematic theory for T_c that can cover the whole region of these basic three parameters; T, S, and ω. The traditional Bardeen-Cooper-Schrieffer (BCS) theory,[6] which is useful only in the weak-coupling region $S \ll T$, tells us that T_c increases as S increases. In the strong-coupling region $S \gg T$, however, T_c is expected to decrease as S increases.[7,5] In the weak region, only electrons around the Fermi level make singlet pairs called Cooper pairs, while, in the strong limit, almost all electrons in the conduction band make strongly bound pairs, which are nothing but the singlet bipolarons mentioned before. Such bipolarons, once formed, can hardly be broken thermally at low temperatures. T_c in this case corresponds to the temperature at which only the coherence between bipolarons is broken by thermal fluctuation, with no change in each bipolaron.[5] In this case, T_c will decrease as the effective mass of the bipolaron increases, and according to the polaron theory,[1] this effective mass increases as S increases. Thus, we have seen the qualitative nature of the competition between S and T, and this competition is expected to make T_c maximum in the transition region $S \approx T$, which lies between the two extreme regions; the BCS region and the bipolaronic region.

In addition to this T-S competition, there is another important competition in the e-b coupled system. That is the competition between T and ω. As mentioned before, the boson cloud around the electron causes the enhancement of its mass, and also causes the mutual attraction between two electrons. In the adiabatic case, $T \gg \omega$, however, the oscillatory motion of the boson is too slow to follow the itinerant motion of the electron, resulting in a very thin boson cloud that can move with each electron. The electron, in this case, is almost bare, and the attraction is also very weak. In the case of the inverse-adiabatic limit $T \ll \omega$, on the other hand, the boson can follow the electron without retardation, and, hence, it causes no mass enhancement. The e-b coupling of this case fully acts as an attraction. When the conducting electrons have a wide energy band of about 10 eV, and are coupling with phonons of about 10 meV, the situation is almost adiabatic. On the other hand, when electrons with a narrow energy band are coupling with plasmons or interband excitonic excitations, the situation is almost inverse adiabatic. To describe these various situations of superconductivity realized in *e*-*b* coupled systems, a systematic theory is required that can cover the whole region of these three basic parameters; T, S, and ω. This problem is closely related to the high-T_c superconductivity of Cu-O–type new ceramics,[8] which has been a matter of worldwide interest recently.

In the present paper, we will clarify the nature of T_c as a function of T, S, and ω, making use of the polaron theory. The fluctuation of the order parameter will be taken into account by the single-site coherent potential approximation (CPA).[9] These results will be applied to the case of Cu-O–type new ceramics. Possible mechanisms of high-T_c superconductivity will be considered in connection with observed bosonic excitations in this material, such as the breathing-mode phonon,[10] the excitonic excitations,[11] and the plasmons.[12]

II. HAMILTONIAN OF MANY-POLARON SYSTEM

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

$$H = -\sum_{l,l',\sigma} T(l-l')\eta_{l\sigma}^{\dagger}\eta_{l'\sigma} + (S\omega/2)^{1/2}\sum_{l,\sigma}(\zeta_l^{\dagger}+\zeta_l^{\dagger})n_{l\sigma} + \omega\sum_{l}\zeta_l^{\dagger}\zeta_l \,. \qquad (2.1)$$

Here $T(l-l')$ is the transfer energy of the electron between two lattice sites, which are specified by two vectors l and l'. The chemical potential of electron is taken to be zero. $\eta_{l\sigma}^{\dagger}$ is the creation operator of electron at site l with spin σ ($=\alpha,\beta$, where α denotes an up spin and β denotes a down spin). S is the site-diagonal *e*-*b* coupling energy. $\zeta_l^{\dagger}$ is the creation operator of the boson at site l with the energy ω, and $n_{l\sigma} \equiv \eta_{l\sigma}^{\dagger}\eta_{l\sigma}$. This boson can correspond to the breathing-mode phonon, the plasmon, or the excitonic excitations. The interelectron Coulombic repulsion is completely neglected for simplicity.

It is expedient to cast all quantities into dimensionless forms: $h \equiv H/\omega$, $t(l-l') \equiv T(l-l')/\omega$, $s \equiv S/\omega$. Then we get

$$h = -\sum_{l,l',\sigma} t(l-l')\eta_{l\sigma}^{\dagger}\eta_{l'\sigma} + (s/2)^{1/2}\sum_{l,\sigma}(\zeta_l^{\dagger}+\zeta_l)n_{l\sigma} + \sum_{l}\zeta_l^{\dagger}\zeta_l \,. \qquad (2.2)$$

As mentioned in Sec. I, each electron in this electron-boson coupled system forms a polaron with a boson cloud around itself. In order to describe this effect, we introduce a displacement operator (M) for the equilibrium position of oscillatory motion of bosons as,

$$M = \exp\left[(s/2)^{1/2}q\sum_{l,\sigma}(\zeta_l^{\dagger}-\zeta_l)n_{l\sigma}\right], \qquad (2.3)$$

where q is the variational parameter that denotes the thickness of the boson 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 boson into a polaron $a_{l\sigma}^{\dagger}$ and a new boson $d_l^{\dagger}$ as

$$a_{l\sigma}^{\dagger} \equiv M^{-1}\eta_{l\sigma}^{\dagger}M = \exp[-(s/2)^{1/2}q(\zeta_l^{\dagger}-\zeta_l)]\eta_{l\sigma}^{\dagger}\,, \qquad (2.4)$$

$$d_l^{\dagger} \equiv M^{-1}\zeta_l^{\dagger}M = \zeta_l^{\dagger} + (s/2)^{1/2}q\sum_{\sigma}n_{l\sigma}\,, \qquad (2.5)$$

where $a_{l\sigma}^{\dagger}$ is the creation operator of a polaron with the boson cloud around itself, and $d_l^{\dagger}$ denotes the creation operator of new boson whose equilibrium position of oscillatory motion is already displaced because of the *e*-*b* coupling. In terms of the polaron and new boson, we can rewrite h as

$$h = -\sum_{l,l',\sigma} t(l-l')a_{l\sigma}^{\dagger}a_{l'\sigma}\exp[-(s/2)^{1/2}q(d_l^{\dagger}-d_l-d_{l'}^{\dagger}+d_{l'})] - \frac{s}{2}(2q-q^2)\sum_{l,\sigma,\sigma'}n_{l\sigma}n_{l\sigma'} + \sum_{l}d_l^{\dagger}d_l + (s/2)^{1/2}(1-q)\sum_{l,\sigma}(d_l^{\dagger}+d_l)n_{l\sigma}\,, \qquad (2.6)$$

where the first term denotes the transfer of a polaron, the second one denotes the attraction between two polarons, the third one denotes the Hamiltonian of new boson, and the last one denotes the interaction between the new boson and the polaron.

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

$$h_0 \equiv \mathrm{Tr}_b[\exp(-\Theta h_b)h]/\mathrm{Tr}_b[\exp(-\Theta h_b)]\,, \qquad h_b \equiv \sum_{l} d_l^{\dagger}d_l \qquad (2.7)$$

where Tr_b means the trace in the boson space, and Θ denotes the reciprocal temperature $\Theta \equiv \omega/k_B T_{\mathrm{temp}}$ (T_{temp} is the temperature). Using h_0, thus defined, we can divide h into four parts as

$$h = h_0 + h_b + \Delta h_1 + \Delta h_2 \,. \qquad (2.8)$$

h_0 in Eq. (2.8) is given by

$$h_0=-x\sum_{l,l',\sigma} t(l-l')a^{\dagger}_{l\sigma}a_{l'\sigma} -s(2q-q^2)\sum_{l} n_{l\alpha}n_{l\beta}-s(2q-q^2)N/2\,, \tag{2.9}$$

where x is the reduction factor of the transfer energy due to the overlap integral of the boson clouds between two sites l and l',

$$x=\exp[-sq^2\coth(\Theta/2)/2]\,. \tag{2.10}$$

Its reciprocal denotes the enhancement of mass due to the polaron effect. The last term of Eq. (2.9) denotes the self-energy of the polarons. Δh_1 denotes the linear-interaction term between polarons and new bosons

$$\Delta h_1\equiv(s/2)^{1/2}(1-q)\sum_{l,\sigma}(d_l^{\dagger}+d_l)n_{l\sigma}\,, \tag{2.11}$$

which is not included in the polaron effect. The last term Δh_2 denotes the difference between the real transfer and the averaged one,

$$\Delta h_2\equiv-\sum_{l,l',\sigma} t(l-l')\{\exp[-(s/2)^{1/2} \times q(d_l^{\dagger}-d_l-d_{l'}^{\dagger}+d_{l'})]-x\}\,. \tag{2.12}$$

Let us now neglect Δh_1 and Δh_2, and calculate the total thermodynamic potential ($\equiv\Omega$),

$$\Omega\equiv-\Theta^{-1}\ln \mathrm{Tr}(e^{-\Theta h_0})\,, \tag{2.13}$$

given by h_0, using the path-integral formalism.[13–15] The effects of Δh_1 and Δh_2 will be discussed in the later sections.

To calculate Ω, we rewrite the interpolaron attraction term of Eq. (2.9) into the following quadratic form with respect to the pairing operator; $a^{\dagger}_{l\alpha}a^{\dagger}_{l\beta}$:

$$\sum_l n_{l\alpha}n_{l\beta}=\tfrac{1}{2}\sum_l (a^{\dagger}_{l\alpha}a^{\dagger}_{l\beta}+a_{l\beta}a_{l\alpha})^2\,. \tag{2.14}$$

As is well known, there are various other ways to rewrite this term into a quadratic form.[13] However, in the present paper we are interested only in the superconductivity which originates from h_0, and we will not be concerned with instabilities of the charge-density wave (CDW) type and the spin-density wave type. Hence, this way of rewriting is most appropriate. In this case, an asymmetric electron-hole picture is very convenient, with the up-spin electrons represented as electrons and the down-spin ones represented as holes:

$$A_l^{\dagger}\equiv a^{\dagger}_{l\alpha},\quad B_l^{\dagger}\equiv a_{l\beta}\,, \tag{2.15}$$

where $A_l^{\dagger}$ and $B_l^{\dagger}$ are the creation operators of electron and hole, respectively. In terms of them, h_0 can be written as

$$h_0=h_K+h_I+\sum_{\mathbf{k}} e_{\mathbf{k}}-s(2q-q^2)N/2\,, \tag{2.16}$$

where h_K denotes the one-body energy of electrons and holes

$$h_K\equiv\sum_{\mathbf{k}} e_{\mathbf{k}}(A^{\dagger}_{\mathbf{k}}A_{\mathbf{k}}-B^{\dagger}_{\mathbf{k}}B_{\mathbf{k}})\,, \tag{2.17}$$

with the following definitions:

$$A_{\mathbf{k}}\equiv\sum_{l} N^{-1/2}e^{-i\mathbf{k}\cdot\mathbf{l}}A_l\,,$$

$$B_{\mathbf{k}}=\sum_{\mathbf{k}} N^{-1/2}e^{-i\mathbf{k}\cdot\mathbf{l}}B_l\,, \tag{2.18}$$

$$t_{\mathbf{k}}\equiv\sum_{l} e^{-i\mathbf{k}\cdot\mathbf{l}}t(\mathbf{l})\,,$$

$$e_{\mathbf{k}}\equiv-xt_{\mathbf{k}}\,, \tag{2.19}$$

and $\mathbf{k}$ is the wave vector. The second term of Eq. (2.16) denotes the interaction between electrons and holes defined as

$$h_I\equiv-s(2q-q^2)\sum_{l}(A_l^{\dagger}B_l+B_l^{\dagger}A_l)^2/2\,. \tag{2.20}$$

III. FUNCTIONAL INTEGRAL FORM

From Eqs. (2.13) and (2.16), the thermodynamic potential Ω can now be written as

$$\Omega=\sum_{\mathbf{k}} e_{\mathbf{k}}-\Theta^{-1}\ln \mathrm{Tr}\{\exp[-\Theta(h_K+h_I)]\} -s(2q-q^2)N/2\,, \tag{3.1}$$

and the second term can be cast in the form of a time-ordered exponential, $\exp_+$, as

$$\mathrm{Tr}[e^{-\Theta(h_K+h_I)}] =\mathrm{Tr}\left[e^{-\Theta h_K}\exp_+\left[-\int_0^{\Theta}du\, h_I(u)\right]\right]\,, \tag{3.2}$$

where the time evolution of an operator O is defined as

$$O(u)\equiv\exp(uh_K)O\exp(-uh_K)\,. \tag{3.3}$$

Using the quadratic form of Eq. (2.20), we can formally rewrite the argument of this time-ordered exponential into the double-time integral form as

$$-\int_0^{\Theta}du\, h_I(u)=s(2q-q^2)\sum_l\int_0^{\Theta}du\int_0^{\Theta}du'\,\delta(u-u')[A_l^{\dagger}(u)B_l(u)+B_l^{\dagger}(u)A_l(u)] \times[A_l^{\dagger}(u')B_l(u')+B_l^{\dagger}(u')A_l(u')]/2\,. \tag{3.4}$$

The δ function on the right-hand side can be expanded by the Fourier series $f_m(u)$ as

$$\delta(u-u')=\sum_m f_m(u)f_m(u'),\quad 0\le(u,u')\le\Theta\,, \tag{3.5}$$

where m is an integer and $f_m(u)$ is defined by

$$f_m(u)\equiv\begin{cases}2^{1/2}\Theta^{-1/2}\cos(2\pi mu/\Theta) & \text{for } m=-1,-2,\dots, \\ 2^{1/2}\Theta^{-1/2}\sin(2\pi mu/\Theta) & \text{for } m=1,2,\dots, \\ \Theta^{-1/2} & \text{for } m=0 .\end{cases}$$

Substituting Eq. (3.5) into Eq. (3.4), we can get a quadratic form of the time-ordered exponential,

$$\exp_+\left[-\int_0^\Theta du\, h_I(u)\right]=\exp_+\left[s(2q-q^2)\sum_{l,m}\left[\int_0^\Theta du\, f_m(u)[A_l^\dagger(u)B_l(u)+B_l^\dagger(u)A_l(u)]\right]^2\Big/2\right], \tag{3.6}$$

and using the Gaussian integral formula for an operator O,

$$\exp(O^2/2)=(2\pi)^{-1/2}\int dy\, \exp(Oy-y^2/2), \tag{3.7}$$

we can linearize the quadratic form of Eq. (3.6) as

$$\exp_+\left[-\int_0^\Theta du\, h_I(u)\right]=\prod_{l,m}\left[(2\pi)^{-1/2}\int dQ_{lm}\right]\exp\left[-\sum_{l,m}Q_{lm}^2/2\right] \times\exp_+\left[[s(2q-q^2)]^{1/2}\sum_{l,m}\int_0^\Theta du\, Q_{lm}f_m(u)[A_l^\dagger(u)B_l(u)+B_l^\dagger(u)A_l(u)]\right]. \tag{3.8}$$

From this result, we finally get our path-integral form

$$\mathrm{Tr}[e^{-\Theta(h_K+h_I)}]=\prod_{l,m}\left[(2\pi)^{-1/2}\int dQ_{lm}\right]\exp\left[-\sum_{l,m}Q_{lm}^2/2\right] \times\mathrm{Tr}\left\{\exp_+\left[-\int_0^\Theta du\left[h_{K,u}-[s(2q-q^2)]^{1/2}\sum_{l,m}Q_{lm}f_m(u)(A_{l,u}^\dagger B_{l,u}+B_{l,u}^\dagger A_{l,u})\right]\right]\right\}, \tag{3.9}$$

where index u of operators $h_{K,u}$, $A_{l,u}^\dagger$, and $B_{l,u}^\dagger$ has no significance except for the time ordering between these operators, in contrast to case of Eq. (3.3).

In order to obtain the exact result of the thermodynamic potential given by h_0, we must perform the path integral with respect to Q_{lm}, which includes both dynamical and static fluctuations of the pairing order. However, our main purpose is to look for an interpolation method for T_c that is useful in all regions of T, S, and ω. For this reason, we retain only the Fourier component with $m=0$ in Eq. (3.9). Within this approximation, Eq. (3.9) is simplified as

$$\mathrm{Tr}[e^{-\Theta(h_K+h_I)}]=\prod_l\left[[\Theta s(2q-q^2)/2\pi]^{1/2}\int dQ_l\right]\exp\left[-\Theta s(2q-q^2)\sum_l Q_l^2/2\right]\mathrm{Tr}(\exp\{-\Theta[h_K+h_I'(\mathrm{Q})]\}), \tag{3.10}$$

where

$$Q_l\equiv Q_{l0}[\Theta s(2q-q^2)]^{-1/2}, \tag{3.11}$$

$$h_I'(\mathrm{Q})\equiv -s(2q-q^2)\sum_l Q_l(A_l^\dagger B_l+B_l^\dagger A_l), \tag{3.12}$$

and Q symbolically denotes an N-dimensional vector; $\mathrm{Q}\equiv(Q_1,Q_2,\dots,Q_N)$. Since only the static part of the fluctuation is retained, this is called static approximation.[13] However, it is exact in the two limiting cases ($T=0$, $S\neq0$) and ($S=0$, $T\neq0$), and can give an interpolation theory in the intermediate cases ($T\neq0$ and $S\neq0$). Moreover, it has no two-body interaction term, but has only a static randomness due to Q_l.

In the case of weak coupling $S\ll T$, the BCS-type mean-field approximation is expected to work well. Hence, let us now derive a solution that corresponds to this approximation, in the context of Eq. (3.10). One can easily infer that Q_l, in this equation, denotes the local-order parameter of the pairing, and if we replace Q_l by its average value $\bar{Q}$, we will get a result corresponding to the BCS theory.[6] For this reason, we replace Q_l by $\bar{Q}+\Delta Q_l$, where ΔQ_l denotes the deviation from the average. By this replacement we can rewrite Eq. (3.10) as

$$\mathrm{Tr}[e^{-\Theta(h_K+h_I)}]=\prod_l\left[[\Theta s(2q-q^2)/2\pi]^{1/2}\int d\Delta Q_l\right]\exp\left[-\Theta s(2q-q^2)\sum_l(\Delta Q_l-\bar{Q})^2/2\right] \times\mathrm{Tr}\left\{\exp\left[-\Theta\left[h_{\mathrm{MF}}-s(2q-q^2)\sum_l\Delta Q_l(A_l^\dagger B_l+B_l^\dagger A_l-2\bar{Q})\right]\right]\right\}, \tag{3.13}$$

where h_{MF} is given as

$$h_{\text{MF}} \equiv h_K - s(2q-q^2)\bar{Q}\sum_l (A_l^\dagger B_l + B_l^\dagger A_l) , \tag{3.14}$$

and the subscript "MF" denotes mean-field approximation. This Hamiltonian corresponds to the traditional BCS theory. If we require that $\bar{Q}$ satisfies the following condition:

$$\bar{Q} = \text{Tr}[\exp(-\Theta h_{\text{MF}}) A_l^\dagger B_l] / \text{Tr}[\exp(-\Theta h_{\text{MF}})] , \tag{3.15}$$

we can neglect the second term in the second line of Eq. (3.13), and the integration over ΔQ_l can be easily performed. Under this approximation, we can diagonalize h_{MF} as,

$$\begin{aligned} h_{\text{MF}} &= \sum_{\mathbf{k}} Y_{\mathbf{k}} (Z_{1\mathbf{k}}^\dagger Z_{1\mathbf{k}} + Z_{2\mathbf{k}}^\dagger Z_{2\mathbf{k}}) - \sum_{\mathbf{k}} Y_{\mathbf{k}} , \\ Y_{\mathbf{k}} &\equiv (e_{\mathbf{k}}^2 + F^2)^{1/2} , \\ F &\equiv s(2q-q^2)\bar{Q} , \end{aligned} \tag{3.16}$$

using the following transformation:

$$\begin{bmatrix} Z_{1\mathbf{k}}^\dagger \\ Z_{2\mathbf{k}} \end{bmatrix} = \begin{bmatrix} \cos(\phi_{\mathbf{k}}) & \sin(\phi_{\mathbf{k}}) \\ -\sin(\phi_{\mathbf{k}}) & \cos(\phi_{\mathbf{k}}) \end{bmatrix} \begin{bmatrix} A_{\mathbf{k}} \\ B_{\mathbf{k}} \end{bmatrix} , \tag{3.17}$$

$$\phi_{\mathbf{k}} \equiv \arctan[(Y_{\mathbf{k}} - e_{\mathbf{k}})/(Y_{\mathbf{k}} + e_{\mathbf{k}})]^{1/2} , \tag{3.18}$$

where $Z_{1\mathbf{k}}^\dagger$ and $Z_{2\mathbf{k}}$ are new fermion operators with the energy $Y_{\mathbf{k}}$. The thermodynamic potential ($\equiv \Omega_{\text{MF}}$) given by h_{MF} becomes

$$\Omega_{\text{MF}} \equiv \sum_{\mathbf{k}} e_{\mathbf{k}} - \sum_{\mathbf{k}} Y_{\mathbf{k}} - s(2q-q^2)N/2 - 2\Theta^{-1} \times \sum_{\mathbf{k}} \ln(1+e^{-\Theta Y_{\mathbf{k}}}) , \tag{3.19}$$

and from Eqs. (3.15) and (3.16) we can get the traditional gap equation,

$$\bar{Q} = \sum_{\mathbf{k}} N^{-1} \frac{F \tanh(\Theta Y_{\mathbf{k}}/2)}{2Y_{\mathbf{k}}} ,$$

or (3.20)

$$1 = s(2q-q^2) \sum_{\mathbf{k}} N^{-1} \frac{\tanh(\Theta Y_{\mathbf{k}}/2)}{2Y_{\mathbf{k}}} .$$

Using these results, we can determine q by the variational condition,

$$\frac{\partial \Omega_{\text{MF}}}{\partial q} = 0 \quad \text{for fixed } \bar{Q} ,$$

and we get

$$q = \left[1 + \coth(\Theta/2) \sum_{\mathbf{k}} N^{-1} e_{\mathbf{k}} [e_{\mathbf{k}} Y_{\mathbf{k}}^{-1} \tanh(\Theta Y_{\mathbf{k}}/2) - 1] / (1 + 4\bar{Q}^2) \right]^{-1} . \tag{3.21}$$

In the real calculation, Eqs. (3.20) and (3.21) are solved numerically by making use of an iteration method. As is already well known, T_c given by this theory increases infinitely as S increases, and it is quite unreasonable in the strong-coupling case. We cannot rely on such a theory when we want to discuss high-T_c problems.

IV. TWO-STATE APPROXIMATION AND CPA

Our main purpose is to look for an interpolation theory for T_c which is useful from the weak limit to the strong limit. In order to obtain such a systematic theory, let us examine strong coupling and the inverse-adiabatic case $\omega \gg S \gg T$, since the BCS-type mean-field theory is expected to break down most typically in this case, although it works very well in the weak region. As shown in our preceding papers,[5] all the electrons in the conduction-band form bipolarons in this strong-coupling case, and the freedom of our system is reduced only to the presence or the absence of a bipolaron at each lattice site. This situation can be denoted by a pseudospin assigned for each site, where the up spin corresponds to the presence and the down spin corresponds to the absence of the bipolaron. The Hamiltonian in this case is written as

$$\begin{aligned} h_0 = -\sum_{l,l'} 2t^2(l-l')s^{-1} \{ & [(0.5+\underline{S}_l^z)(0.5-\underline{S}_{l'}^z) \\ & + (0.5-\underline{S}_l^z)(0.5+\underline{S}_{l'}^z)] \\ & + 2(\underline{S}_l^x \underline{S}_{l'}^x + \underline{S}_l^y \underline{S}_{l'}^y) \} , \end{aligned} \tag{4.1}$$

where we have omitted unimportant constant terms of h_0. $\underline{S}_l^z$, $\underline{S}_l^x$, and $\underline{S}_l^y$ are the Pauli spin operators of the 2×2 matrix forms. The first line $[\cdots]$ of Eq. (4.1) denotes the virtual transfer of a bipolaron through the second-order perturbation of $T(l-l')$, while the second line $(\cdots)$ denotes the real transfer. Since the averaged value of $\underline{S}_l^z$ is zero in the N-site N-electron system, we tilt the z axis of spin 90° in the zx plane, and transform $\underline{S}_l^z$, $\underline{S}_l^x$, and $\underline{S}_l^y$ into new spin operators $\tilde{S}_l^z$, $\tilde{S}_l^x$, and $\tilde{S}_l^y$, defined as

$$\tilde{S}_l^x \equiv -\underline{S}_l^z, \quad \tilde{S}_l^z \equiv \underline{S}_l^x, \quad \tilde{S}_l^y \equiv \underline{S}_l^y . \tag{4.2}$$

After this transformation, we get a Hamiltonian of a pseudospin system with a ferromagnetic exchange interaction as

$$h_0 = -N\sum_{l} t^2(l)s^{-1} - \sum_{l,l'} 4t^2(l-l')s^{-1}[\tilde{S}_l^z\tilde{S}_{l'}^z - (\tilde{S}_l^+\tilde{S}_{l'}^+ + \tilde{S}_l^-\tilde{S}_{l'}^-)/2] ,$$

$$\tilde{S}_l^\dagger \equiv \tilde{S}_l^x + i\tilde{S}_l^y, \quad \tilde{S}_l^- \equiv \tilde{S}_l^x - i\tilde{S}_l^y . \tag{4.3}$$

By applying the traditional mean-field theory for this pseudospin system, we can easily get T_c as

$$k_B T_c = \sum_{l} T^2(l)S^{-1} . \tag{4.4}$$

It gives a new T_c, that decreases as S increases in contrast to the BCS theory. It should be noted, that in this calculation based on the mean-field theory for the pseudospin system, we have been concerned only with the Boltzmann's distribution between two eigenstates $|l,\pm\rangle$ of the operator $\tilde{S}_l^z$. These states are written as

$$|l,\pm\rangle = 2^{-1/2}(A_l^\dagger \pm B_l^\dagger)|0\rangle \quad (|0\rangle \rightarrow \text{vacuum of electron and hole}) , \tag{4.5}$$

and the local-order parameter of these states takes the values $\pm\bar{Q}$ as

$$\langle l,\pm | A_l^\dagger B_l | l,\pm\rangle = \pm\bar{Q} , \tag{4.6}$$

because $\bar{Q}$ is 0.5 in the strong-coupling limit.

Keeping this result in mind, let us return to Eq. (3.10), and consider its physical significance in connection with the broken symmetry and the phase transition. The last factor of Eq. (3.10) gives the local thermodynamic potential associated with a given configuration $\mathbf{Q}$, and the summation over all possible configurations with the weight

$$\prod_{l} P(Q_l), \quad P(Q_l) \equiv [\Theta s(2q-q^2)/2\pi]^{1/2} \times \exp[-\Theta s(2q-q^2)Q_l^2/2] \tag{4.7}$$

gives the total thermodynamic potential of our system. 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 $\mathbf{Q}$ within such a local region can also be described by Eq. (3.10), if we replace $P(Q_l)$ by a new weight function $P'(Q_l)$ which effectively reflects this conditional summation over $\mathbf{Q}$. In this case $P'(Q_l)$ has a polarization in Q_l space so as to give a nonzero order parameter, although the original weight function $P(Q_l)$ has not. Thus, the problem to find a local region is now converted into the problem to find P'. For such $P'(Q_l)$, we assume the following simple form,

$$P'(Q_l) = y_+\delta(Q_l - \bar{Q}) + y_-\delta(Q_l + \bar{Q}), \quad y_+ + y_- = 1 , \tag{4.8}$$

where Q_l takes only two values, $\bar{Q}$ and $-\bar{Q}$, with statistical weights y_+ and y_-, which are unknown at 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 Eq. (3.14) if we take $y_- = 0$ and $y_+ = 1$, while Eq. (4.6) is nothing but the two-state approximations. 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.

Within this approximation, we can rewrite Eq. (3.10) as

$$\mathrm{Tr}[e^{-\Theta(h_K + h_I)}] = \langle\langle \mathrm{Tr}\{e^{-\Theta[h_K + h_I'(\mathbf{Q})]}\}\rangle\rangle , \tag{4.9}$$

where

$$\langle\langle \cdots \rangle\rangle \equiv \prod_{l} \left[\int dQ_l P'(Q_l)\right] \cdots , \tag{4.10}$$

and since we have no two-body terms in the Hamiltonian, the right-hand side can be easily rewritten in terms of the one-body Green's function $\underline{G}(E+i\varepsilon,\mathbf{Q})$ as

$$\langle\langle \mathrm{Tr}\{e^{-\Theta[h_K + h_I'(\mathbf{Q})]}\}\rangle\rangle = \left\langle\!\left\langle \exp\left[-\frac{\mathrm{Im}}{\pi}\left[\int dE \ln(1+e^{-\Theta E})\,\mathrm{Tr}[\underline{G}(E+i\varepsilon,\mathbf{Q})]\right]\right]\right\rangle\!\right\rangle , \tag{4.11}$$

where the Green's function is defined as

$$\underline{G}(E+i\varepsilon,\mathbf{Q}) \equiv \left[E + i\varepsilon - \left[\underline{h}_K + \sum_{l} \underline{h}_{Il}(Q_l)\right]\right]^{-1} . \tag{4.12}$$

In this equation, $\underline{h}_K$ and $\sum_l \underline{h}_{Il}(Q_l)$ are the one-body versions of h_K and $h_I'(\mathbf{Q})$, which are defined as

$$\underline{h}_{Il}(Q_l) \equiv -UQ_l[\,|l1)(l2| + |l2)(l1|\,], \quad U \equiv s(2q-q^2) \tag{4.13}$$

$$\underline{h}_K \equiv \sum_{\mathbf{k}} e_{\mathbf{k}}[\,|\mathbf{k}1)(\mathbf{k}1| - |\mathbf{k}2)(\mathbf{k}2|\,] \tag{4.14}$$

$$|\mathbf{k}i) \equiv \sum_{l} N^{-1/2} e^{-i\mathbf{k}\cdot l} |li), \quad i = 1,2 \tag{4.15}$$

where $|l1)$ and $|l2)$ are the Wannier functions of the electron and the hole at site l, respectively.

According to the generalized cumulant expansion method,[16] Eq. (4.11) can be expanded as

$$\left\langle\!\left\langle \exp\left[-\frac{\mathrm{Im}}{\pi}\left[\int dE\,\ln(1+e^{-\Theta E})\,\mathrm{Tr}[\underline{G}(E+i\varepsilon,\mathbf{Q})]\right]\right]\right\rangle\!\right\rangle$$

$$=\exp\left[-\frac{\mathrm{Im}}{\pi}\left[\int dE\,\ln(1+e^{-\Theta E})\,\mathrm{Tr}[\langle\langle \underline{G}(E+i\varepsilon,\mathbf{Q})\rangle\rangle]\right]\right.$$

$$+\tfrac{1}{2}\int dE\int dE'\,\ln(1+e^{-\Theta E})\ln(1+e^{-\Theta E'})$$

$$\times\left[\left\langle\!\left\langle \frac{\mathrm{Im}}{\pi}\{\mathrm{Tr}[\underline{G}(E+i\varepsilon,\mathbf{Q})]\}\frac{\mathrm{Im}}{\pi}\{\mathrm{Tr}[\underline{G}(E'+i\varepsilon,\mathbf{Q})]\}\right\rangle\!\right\rangle\right.$$

$$\left.\left.-\left\langle\!\left\langle \frac{\mathrm{Im}}{\pi}\{\mathrm{Tr}[\underline{G}(E+i\varepsilon,\mathbf{Q})]\}\right\rangle\!\right\rangle\left\langle\!\left\langle \frac{\mathrm{Im}}{\pi}\{\mathrm{Tr}[\underline{G}(E'+i\varepsilon,\mathbf{Q})]\}\right\rangle\!\right\rangle\right]+\cdots\right], \tag{4.16}$$

where the first term in [· · ·] of the right-hand side denotes the first-order cumulant and the other terms correspond to the higher-order cumulants. However, we terminate this expansion up to the first order, and the thermodynamic potential is given as

$$e^{-\Theta\Omega}=\exp\left[-\Theta\left[\sum_{\mathbf{k}}e_{\mathbf{k}}-s(2q-q^2)N/2\right]+\int dE\,\ln(1+e^{-\Theta E})\left[-\frac{\mathrm{Im}}{\pi}\mathrm{Tr}[\langle\langle \underline{G}(E+i\varepsilon,\mathbf{Q})\rangle\rangle]\right]\right]. \tag{4.17}$$

Such a termination of the cumulant expansion gives the exact result in two limiting cases ($T=0$, $S\neq0$) and $S=0$, $T\neq0$), and, hence, it can give an interpolation theory for the intermediate cases ($T\neq0$ and $S\neq0$).

Our problem is now reduced to the averaged one-body Green's function $\langle\langle \underline{G}(E+i\varepsilon,\mathbf{Q})\rangle\rangle$, and we determine it within the single-site CPA.[9] That is, we approximate $\langle\langle \underline{G}(E+i\varepsilon,\mathbf{Q})\rangle\rangle$ by an effective Green's function $\underline{G}(E+i\varepsilon)$ in which $\underline{h}_{ll}(Q_l)$ is replaced by an energy-dependent coherent potential $\underline{V}_l(E)$,

$$\langle\langle \underline{G}(E+i\varepsilon,\mathbf{Q})\rangle\rangle\rightarrow\underline{G}(E+i\varepsilon)\equiv\left[E+i\varepsilon-\left[\underline{h}_K+\sum_l\underline{V}_l(E)\right]\right]^{-1}, \tag{4.18}$$

where $\underline{V}_l(E)$ is the 2×2 matrix with the element $V_{ij}(E)$ ($i,j=1,2$) which is defined as

$$\underline{V}_l(E)\equiv-U\sum_{i,j=1,2}V_{ij}(E)\,|\,li)(lj\,|\;. \tag{4.19}$$

$\underline{V}_l(E)$ is determined by the well-known condition, that the averaged site-diagonal scattering matrix, coming from the replacement $\underline{h}_{ll}(Q_l)\rightarrow\underline{V}_l(E)$, must be zero,

$$\langle\langle[\underline{h}_{ll}(Q_l)-\underline{V}_l(E)]/\{1-\underline{G}(E+i\varepsilon)[\underline{h}_{ll}(Q_l)-\underline{V}_l(E)]\}\rangle\rangle=0\;. \tag{4.20}$$

Since the two-state approximation is similar to the binary alloy problem,[9] it can be rewritten into the following simple form:

$$\underline{V}_l(E)=\underline{h}_{ll}(\tilde{Q})-\{[\underline{h}_{ll}(\bar{Q})-\underline{V}_l(E)]\underline{G}(E+i\varepsilon)[\underline{h}_{ll}(-\bar{Q})-\underline{V}_l(E)]$$

$$+[\underline{h}_{ll}(-\bar{Q})-\underline{V}_l(E)]\underline{G}(E+i\varepsilon)[\underline{h}_{ll}(\bar{Q})-\underline{V}_l(E)]\}/2\;, \tag{4.21}$$

$$\tilde{Q}\equiv(y_+-y_-)\bar{Q}\;. \tag{4.22}$$

$\underline{G}(E+i\varepsilon)$ defined in Eq. (4.18) is already diagonal with respect to **k**, because V_{ij} is independent of l, and its element can be written as

$$(\mathbf{k}i\,|\,\underline{G}(E+i\varepsilon)\,|\,\mathbf{k}'j)=\delta_{\mathbf{k}\mathbf{k}'}G_{\mathbf{k}ij}(E+i\varepsilon),\quad i,j=1,2 \tag{4.23}$$

where $G_{\mathbf{k}ij}(E+i\varepsilon)$ denotes the diagonal element, and is given as

$$\begin{bmatrix}G_{\mathbf{k}11} & G_{\mathbf{k}12}\\ G_{\mathbf{k}21} & G_{\mathbf{k}22}\end{bmatrix}=\begin{bmatrix}E+i\varepsilon+e_{\mathbf{k}}+UV_{22} & -UV_{12}\\ -UV_{21} & E+i\varepsilon-e_{\mathbf{k}}+UV_{11}\end{bmatrix}$$

$$\times([E+i\varepsilon+U(V_{11}+V_{22})/2]^2-\{[e_{\mathbf{k}}-U(V_{11}-V_{22})/2]^2+U^2V_{12}V_{21}\})^{-1}\;. \tag{4.24}$$

Using Eq. (4.23), we can define the lattice Green's function $G_{ij}(E+i\varepsilon)$ as

$$G_{ij}(E+i\varepsilon)=\sum_{\mathbf{k}}N^{-1}G_{\mathbf{k}ij}(E+i\varepsilon)\;, \tag{4.25}$$

which is independent of l. From Eqs. (4.25), (4.24), and (4.21) we get a set of equations for V_{ij} as

$$\left.\begin{aligned} V_{ii} &= U[V_{ii}^2 G_{ii} + (V_{12}V_{21} - \bar{Q}^2)G_{jj} + V_{ii}(V_{21}G_{12} + V_{12}G_{21})] \\ V_{ij} &= \tilde{Q} + U[V_{11}V_{22}G_{ij} + (V_{ij}^2 - \bar{Q}^2)G_{ji} + V_{ij}(V_{11}G_{11} + V_{22}G_{22})] \end{aligned}\right\}, \quad (i,j)=(1,2) \text{ or } (2,1) . \tag{4.26, 4.27}$$

The new order parameter R, which is defined as

$$R \equiv \sum_{l} (2N)^{-1} \frac{\langle\langle \mathrm{Tr}\{ e^{-\Theta[h_K + h_I'(\mathbf{Q})]} (A_l^\dagger B_l + B_l^\dagger A_l)\} \rangle\rangle}{\langle\langle \mathrm{Tr}\{ e^{-\Theta[h_K + h_I'(\mathbf{Q})]} \} \rangle\rangle} , \tag{4.28}$$

can also be calculated by using almost the same procedure as we did in getting Eq. (4.17):

$$R = -\frac{\mathrm{Im}}{\pi} \int dE\, (1+e^{\Theta E})^{-1} [G_{12}(E+i\varepsilon) + G_{21}(E+i\varepsilon)]/2 . \tag{4.29}$$

V. SELF-CONSISTENCY CONDITION FOR $y_\pm$

Let us now derive a self-consistency equation for $y_\pm$. As we have mentioned, the last factor of Eq. (3.10) gives a local thermodynamic potential at the given configuration $\mathbf{Q}$, and we can interpret it as a statistical weight, which, using, we can calculate expectation values of various physical quantities. One example is seen in Eq. (4.28). As an extension of this way of thinking, we can define a new weight function $P''(Q_l)$ as

$$P''(Q_l) \equiv \langle\langle \mathrm{Tr}\{ e^{-\Theta[h_K + h_I'(\mathbf{Q})]} \} \rangle\rangle_l / \langle\langle \mathrm{Tr}\{ e^{-\Theta[h_K + h_I'(\mathbf{Q})]} \} \rangle\rangle ,$$

where

$$\langle\langle \cdots \rangle\rangle_l \equiv \prod_{\substack{l' \\ (l' \neq l)}} \int dQ_{l'} P'(Q_{l'}) \cdots , \tag{5.1}$$

which describes a statistical distribution of Q_l at site l, although it is not normalized. $P''(Q_l)$, thus defined, includes the effects of $P'(Q_{l'})$ of its environmental sites l' ($\neq l$), because polarons are moving between these sites through the transfer energy $T(l-l')$. Hence, if $P''(Q_l)$ has some polarization in the Q_l space, its origin is nothing but the polarization of $P'(Q_{l'})$. Since all of the sites must be equivalent, we get a self-consistency condition for $P'(Q_l)$ as

$$P''(Q_l) \propto P'(Q_l) . \tag{5.2}$$

That is, P' must be equal to P'' except for the normalization constant. Using the same procedure as we did in the previous section, we can rewrite $P''(Q_l)$ defined by Eq. (5.1) in terms of the Green's function as

$$P''(Q_l) = \exp\left[-\int dE \ln(1+e^{-\Theta E}) \frac{\mathrm{Im}}{\pi} \times \{ \mathrm{Tr}[\langle\langle \underline{G}(E+i\varepsilon,\mathbf{Q}) \rangle\rangle_l - \langle\langle \underline{G}(E+i\varepsilon,\mathbf{Q}) \rangle\rangle] \} \right] , \tag{5.3}$$

and within the CPA, $\langle\langle \underline{G}(E+i\varepsilon,\mathbf{Q}) \rangle\rangle_l$ can be replaced by $\underline{G}_l'$ which is defined as

$$\langle\langle \underline{G}(E+i\varepsilon,\mathbf{Q}) \rangle\rangle_l \rightarrow \underline{G}_l'(E+i\varepsilon, \pm\bar{Q}) \equiv \left[E + i\varepsilon - \left[\underline{h}_K + \underline{h}_{Il}(\pm\bar{Q}) + \sum_{\substack{l' \\ (l' \neq l)}} \underline{V}_{l'}(E) \right] \right]^{-1} . \tag{5.4}$$

Thus, we can write the equation for $y_\pm$ as

$$y_\pm \propto \exp\left[-\int dE \ln(1+e^{-\Theta E}) \frac{\mathrm{Im}}{\pi} \times \{ \mathrm{Tr}[\underline{G}_l'(E+i\varepsilon, \pm\bar{Q}) - \underline{G}(E+i\varepsilon)] \} \right] , \tag{5.5}$$

and using a well-known mathematical formula[17] based on the integration by parts with respect to E, we finally get an equation for y_+ alone

$$y_+ = \left[1+\exp \left[\Theta \int dE\,(1+e^{\Theta E})^{-1} \frac{\mathrm{Im}}{\pi} \{ \mathrm{Tr}[\ln(\{1-[\underline{h}_{ll}(-\bar{Q})-\underline{V}_l(E)]\underline{G}(E+i\varepsilon)\} \right.\right.$$
$$\left.\left. \times\{1-[\underline{h}_{ll}(\bar{Q})-\underline{V}_l(E)]\underline{G}(E+i\varepsilon)\}^{-1})]\} \right]\right]^{-1} . \tag{5.6}$$

To calculate the trace in Eq. (5.6), the following mathematical technique is convenient. We formally rewrite $\underline{h}_{ll}$, $\underline{V}_l$, and $\underline{G}$ in terms of $\underline{S}_l^z$, $\underline{S}_l^x$, and $\underline{S}_l^y$, and make use of the formula

$$\ln(1-O) = -\sum_{n=1}^{\infty} O^n \Big/ n$$

for an operator O. The result is given as

$$\mathrm{Tr}(\ln\{1-[\underline{h}_{ll}(\pm\bar{Q})-\underline{V}_l(E)]\underline{G}(E+i\varepsilon)\}) = \ln(1-U[V_{11}G_{11}+V_{22}G_{22}+(V_{12}\mp\bar{Q})G_{21}+(V_{21}\mp\bar{Q})G_{12}]$$
$$+U^2\{[V_{11}V_{22}-(V_{12}\mp\bar{Q})(V_{21}\mp\bar{Q})](G_{11}G_{22}-G_{12}G_{21})\}) . \tag{5.7}$$

By using these results, we can calculate T_c from the equation

$$y_+ = y_- . \tag{5.8}$$

Let us now consider the strong coupling limit $S \gg T$ and solve this equation within the second-order perturbation with respect to T. In this limit, as seen from Eqs. (3.21) and (3.20), $\bar{Q}=\frac{1}{2}$ and $q=1$. Hence, the zero-order Green's function $\underline{G}_l'(E+i\varepsilon,\pm\bar{Q})$ becomes as

$$\underline{G}_l'(E+i\varepsilon,\pm\bar{Q}) = |\,l,\pm\rangle(E+i\varepsilon+S/2)^{-1}\langle l,\pm\,| + |\,l,\mp\rangle(E+i\varepsilon-s/2)^{-1}\langle l,\mp\,|$$
$$+ \sum_{\substack{l' \\ (l'\neq l)}} \{\,|\,l',+\rangle[y_+(E+i\varepsilon+s/2)^{-1}+y_-(E+i\varepsilon-s/2)^{-1}]\langle l',+\,|$$
$$+\,|\,l',-\rangle[y_-(E+i\varepsilon+s/2)^{-1}+y_+(E+i\varepsilon-s/2)^{-1}]\langle l',-\,|\,\} . \tag{5.9}$$

In Fig. 1, we have schematically shown the localized energy levels corresponding to the poles of $\underline{G}_l'$, taking $Q_l=\bar{Q}$ as an example. We should note that the chemical potential of electron is taken to be zero. There are two levels at each site, and all these levels are mixed up with each other through the transfer energy, whose matrix element is given as

$$\langle l\mp\,|\,\underline{h}_K\,|\,l',\pm\rangle = -xt(l-l'), \quad l\neq l' . \tag{5.10}$$

In the case of $Q_l=\bar{Q}$, the energy lowering of the occupied level $|\,l,+\rangle$ due to the mixing with unoccupied levels $|\,l',-\rangle$ through the second-order perturbation is given as $-\sum_l t^2(l)s^{-1}x^2y_+$. On the other hand, the energy lowering of the occupied levels $|\,l',+\rangle$ due to the mixing with $|\,l,-\rangle$ is entirely same as the previous value. We can also easily see that the mixing between occupied levels causes no net energy difference when they are summed up. In the case of $Q_l=-\bar{Q}$, we can also calculate the energy lowering by the same procedure, and from Eq. (5.5) we obtain

$$y_\pm \propto \exp\left[2\Theta \sum_l x^2t^2(l)s^{-1}y_\pm\right] . \tag{5.11}$$

We should also note that the mixing between environmental sites causes no difference between the two cases $Q_l=\bar{Q}$ and $Q_l=-\bar{Q}$. From Eq. (5.11), T_c is given as

$$k_BT_c = \sum_l T^2(l)/[S\exp(S/\omega)] , \tag{5.12}$$

which is a more general form than Eq. (4.4). In the real calculation of T_c, $\sum_l T(l)^2$ can be obtained from the second moment of the state density of electron.

VI. POLARON–NEW-BOSON INTERACTION AND NUMERICAL RESULTS

Let us now take into account the effect of coupling between polarons and new bosons given by Eq. (2.11). The region of temperature in which we are mainly interested is $\Theta^{-1} \lesssim k_BT_c/\omega$, and k_BT_c is expected to be far smaller than ω; $k_BT_c \ll \omega$. In this sense, our system is mainly in the new-boson vacuum, and Δh_1 causes the small mixing between this vacuum and one-boson excited states. We take this effect into account by the second-order perturbation theory of Δh_1, and can eliminate the new-boson part as we usually do in the BSC theory. After this procedure, we can obtain an effective Hamiltonian ($\equiv h_e$) as

$$h \to h_e = h_0 - s(1-q^2)\sum_{l,\sigma,\sigma'} n_{l\sigma}(h_0+1-E_0)^{-1}n_{l\sigma'}/2 , \tag{6.1}$$

where E_0 of the energy denominator $(h_0+1-E_0)^{-1}$ is the lowest eigenvalue of h_0. In order to take the second term of Eq. (6.1) into account within the framework of the theory developed in preceding sections, we approximate as,

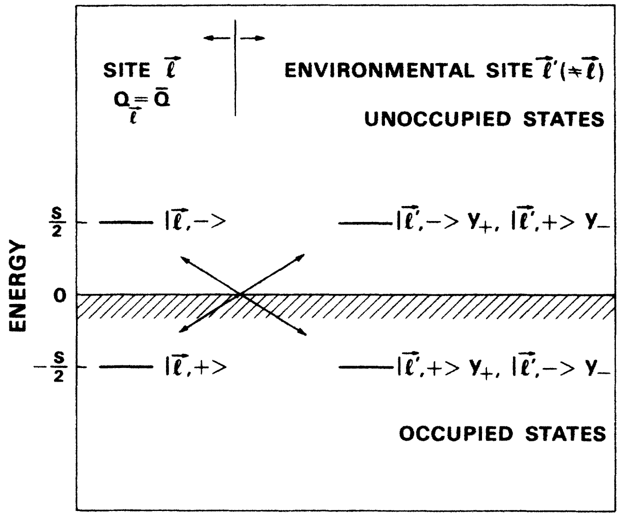

FIG. 1. Localized energy levels at each site, $T=0$, $Q_l=\bar{Q}$. y_+ and y_- are the statistical weights of each level. l is the relevant site and l' is its environmental site. The long arrows denote the mixing between levels through $T(l-l')$. The chemical potential of electron is taken to be zero.

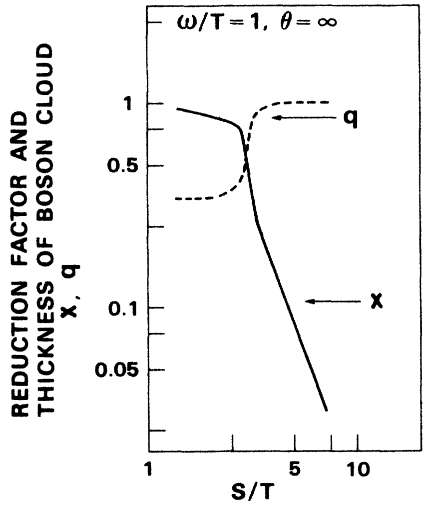

FIG. 2. The reduction factor x and the thickness of boson cloud q as a function of S/T. $\omega/T=1$, $\Theta=\infty$.

$$(h_0+1-E_0)^{-1}\to(2F+1)^{-1}\frac{F}{[F^2+(4Tx/\omega)^2]^{1/2}}\ , \qquad (6.2)$$

where the energy denominator is replaced by $(2F+1)$, $2F$ being the energy gap given by the BCS theory at absolute zero temperature. The factor $F/[F^2+(4Tx/\omega)^2]^{1/2}$ is introduced so that it represents the effective area in which $Y_{\mathbf{k}}$ can be replaced by F in the k space. $[F^2+(4Tx/\omega)^2]^{1/2}$ denotes the half-width of the energy band with the pairing order, since we have taken the constant density of states centered at zero and with a width $8T$ for noninteracting electrons. This simple density of states will be used in all calculations hereafter. The formal nature of h_e, thus obtained, is entirely the same as h_0 given by Eq. (2.9) except that U is replaced by U' defined by,

$$U\to U'\equiv s\{(2q-q^2)+(1-q^2)(2F+1)^{-1}$$
$$\times F[F^2+(4Tx/\omega)^2]^{-1/2}\}\ , \qquad (6.3)$$

although the values of q, F, and x have to be determined beforehand. One can easily infer that the difference between h_0 and h_e mainly appears in the BCS region with $q\ll 1$, and the bipolaronic region is not affected. Using h_e, thus obtained, we repeat the same procedure as in preceding sections and finally get a new set of equations for T_c, R, and G, which are the same as Eqs. (5.8), (4.29), and (4.18) except that U is replaced by U'.

As for Δh_2 in Eq. (2.12), we have also estimated its effects, but they are always smaller than that of Δh_1. Let us now proceed to the numerical results obtained by solving the set of equations above. Figure 2 shows q and x given by the BCS-type mean-field theory at absolute zero of temperature, and we can see that a continuous but sudden change occurs in q and x, as S/T increases from the BCS region to the bipolaronic region. Figure 3 shows the difference of the two order parameters R and $\bar{Q}$, obtained by the present theory and by the BCS-type mean-field theory. In this case, T_c is about a half of that of the BCS theory. Figure 4 shows the one-particle density of states, $-\mathrm{Im}[G_{11}(E+i\varepsilon)]/\pi$, as a function of temperature and energy. As mentioned before, the density of states of noninteracting electrons is assumed to be constant with the width $8T$ and centered at the chemical potential of the electron, that is zero. We can see the energy gap opens as temperature Θ^{-1} decreases. Figure 5 shows an example for the left- and the right-hand sides of Eq. (5.6), which are calculated to determine y_+ numerically. Figure 6 shows T_c as a function of S/T and ω/T. In the BCS region $S\ll T$, T_c increases as S increases, while in the bipolaronic region it decreases as S increases. The rapidity of this decrease depends also on $\exp(S/\omega)$ as shown in Eq. (5.12), and this factor denotes the mass enhancement. In the inverse-adiabatic case with a large

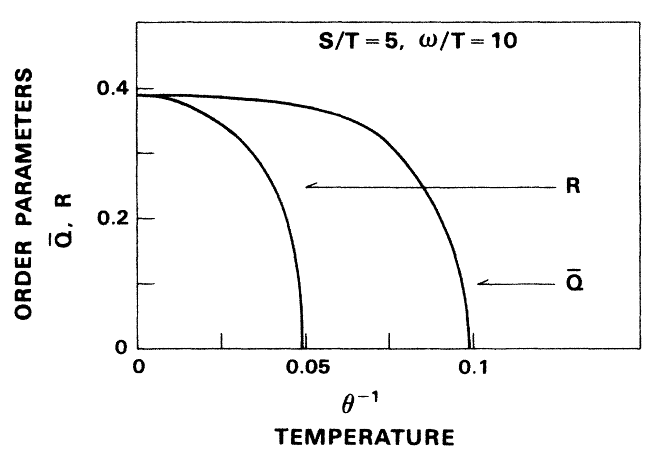

FIG. 3. Order parameters R and $\bar{Q}$ as a function of temperature Θ^{-1} obtained by Eqs. (4.28) and (3.20). $S/T=5$, $\omega/T=10$.

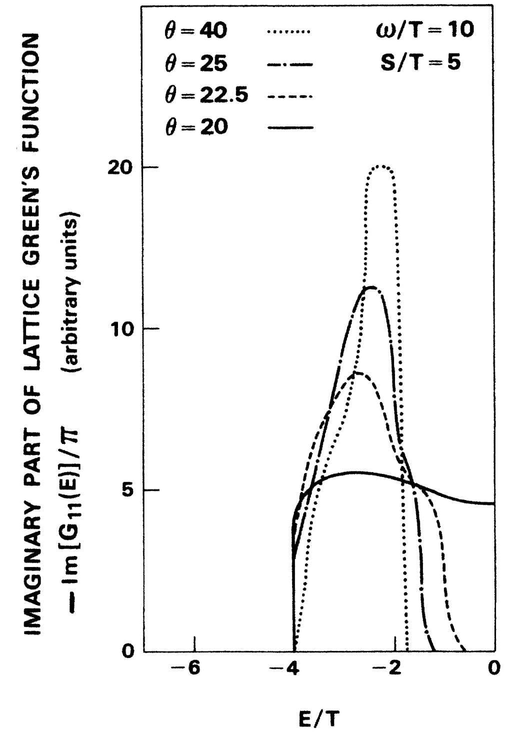

FIG. 4. The imaginary part of the lattice Green's function $-\mathrm{Im}(G_{11})/\pi$ as a function of E and the reciprocal temperature Θ. $\omega/T=10, S/T=5$.

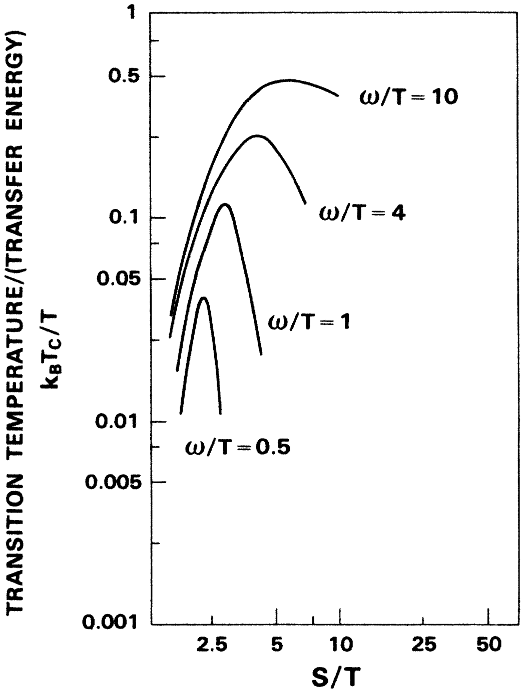

FIG. 6. The transition temperatures k_BT_c/T as a function of ω/T and S/T. T denotes the intersite transfer energy of the electron.

ω/T, this decrease is rather slow, while in the adiabatic case with a small ω/T, it is very rapid. Thus, T_c takes its maximum in the transition region $S \approx T$, which lies between the BCS region and the bipolaronic region. As seen from Figs. 2 and 6, this transition region is narrow in the adiabatic case, while it is relatively wide in the inverse-adiabatic case. It is also interesting to see the ratio between the effective energy gap $2U'R$ to k_BT_c; $2U'R/k_BT_c$. In the BCS region it is about 3.5, but in the bipolaronic region it can take much bigger values, for example, it is 8 in the case of Fig. 3.

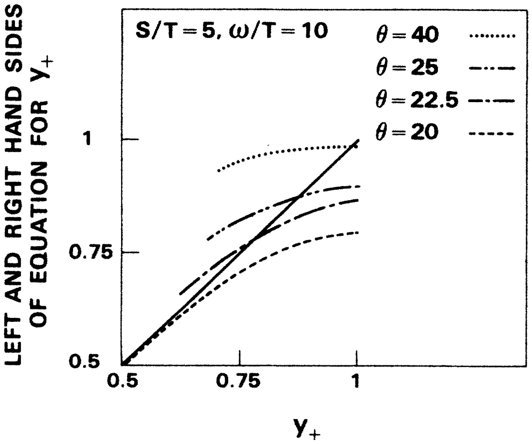

FIG. 5. The left- and right-hand sides of Eq. (5.6) calculated to determine y_+ numerically. The solid line denotes the left-hand side, and the other lines denote the right-hand side at various reciprocal temperatures. $S/T=5, \omega/T=10$.

These new results are firstly obtained by our unified theory that can cover the whole region of T, S, and ω. Finally, it should be noted that T_c has its upper limit for given values of T and ω, even though we have completely ignored the occurrence of the CDW-type instability[5] or other instabilities of the lattice. Consequently, such a unified theory is essential to study the high-T_c superconductivity, if it comes from the coupling between electrons and quasibosons.

VII. Cu-O–TYPE NEW CERAMICS

Using the numerical results obtained in preceding sections, let us now consider the nature of the superconductivity of Cu-O–type new ceramics. The high-temperature superconductivity of this type material is a matter of worldwide interest in these several months, and its T_c is now going to exceed 100 K. To clarify the microscopic mechanism of such a new phenomena, many theoretical and experimental studies have already been undertaken.[8,10–12,18] In spite of these efforts, however, the whole aspect of its mechanism is still left unclear. For this reason, in this section, we consider this problem, assuming that such a high T_c comes from the strong coupling between electrons and quasibosonic excitations with very high frequencies.

As is well known, the electrons relevant to this superconductivity are in the antibonding state between the d orbital of Cu and the p orbital of O. According to the energy-band calculation[19] the width of the conduction band is about 2 eV, and it corresponds to the case $T \approx 0.2$ eV in our theory. The number of the electrons of this material somewhat deviates from the half-filled case, while our theory is derived only for the half-filled case. However, this difference causes no serious change in using our theory, if we choose the value of T appropriately, since the density of state of the noninteracting electrons is assumed to be constant. The first candidate for the boson is the breathing mode phonon of oxygen atoms whose energy is about 0.1 eV.[10] In the optical spectra of this material, we have another high frequency mode with a energy of about 0.3 eV.[11] Judging from its energy, one can tentatively assign that it is an excitonic excitation. This is the second candidate, which seems to cooperate with the first candidate. The third candidate is the plasmon with the energy of about 0.8 eV.[12] Since its energy is very low compared with that of the alkali metals, we can expect that it couples with conducting electrons at the Fermi level.

From Fig. 6, we can see T_c's corresponding to these three candidates. The breathing mode can give T_c of about 100 K. The second candidate can give T_c of about 300 K, and the third candidate can give T_c of about 400 K. These high values of T_c should be understood as the upper limit of T_c for given ω and T, since we have neglected the pair breaking effects such as the randomness, the direct Coulombic repulsion and the CDW-type instability. As mentioned occasionally the high T_c always appears in the transition region between the BCS region and the bipolaronic region, and, hence, it is highly probable that the superconductivity of this material is in this transition region. As the characteristics of this region, we should note the following two points: The first is the ratio $2U'R/k_BT_c$. As mentioned before, it can become larger than that of the BCS region, and in some cases it takes a value of the order of 10. By the tunneling spectroscopy such a large ratio is observed.[20] The second is related with the isotope effect.[21] When the three parameters T, S, and ω are of the same order, $\partial F/\partial\omega$ becomes relatively smaller than that of the traditional BCS region, as already shown in our previous paper (see Fig. 4 of Ref. 5b). This also holds for $\partial T_c/\partial\omega$, and, hence, the isotope effect is expected to be relatively smaller than that of the BCS region.

Since we have completely neglected the interelectron Coulombic repulsion, we are now going to take this effect into account on the basis of the present theory. We hope to report the results in subsequent papers. We will also attempt to reinforce the static approximation by including dynamical effects.

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[1] C. Kuper and G. Whitfield, *Polarons and Excitons* (Oliver and Boyd, Edinburgh, 1963).

[2] V. Vinetskii, Zh. Eksp. Teor. Fiz. **40**, 1459 (1961) [Sov. Phys.—JETP **13**, 1023 (1961)].

[3] P. Anderson, Phys. Rev. Lett. **34**, 953 (1975).

[4] J. Bredas and G. Street, Acc. Chem. Res. **18**, 309 (1985).

[5] (a) K. Nasu, Phys. Rev. B **35**, 1748 (1987); (b) Physica **143B**, 229 (1986); (c) J. Phys. Soc. Jpn. **54**, 1933 (1985).

[6] J. Schrieffer, *Theory of Superconductivity* (Benjamin, New York, 1964).

[7] P. Pincus, P. Chaikin, and C. Coll, III, Solid State Commun. **12**, 1265 (1973); A. Alexandrov and J. Ranninger, Phys. Rev. B **23**, 1796 (1981); B **24**, 1164 (1981); A. Alexandrov, J. Ranninger, and S. Robaszkiewicz, *ibid.* **33**, 4526 (1986).

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

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

[10] S. Sugai, M. Sato, and S. Hosoya, Jpn. J. Appl. Phys. **26**, L495 (1987).

[11] K. Kamaras, C. Porter, M. Doss, S. Herr, D. Tanner, D. Bonn, J. Greedan, A. O'Reilly, C. Stager, and T. Timusk, Phys. Rev. Lett. **59**, 919 (1987).

[12] S. Tajima, S. Uchida, S. Tanaka, S. Kanbe, K. Kitazawa, and K. Fueki, Jpn. J. Appl. Phys. **26**, L432 (1987).

[13] W. Evenson, J. Schrieffer, and S. Wang, J. Appl. Phys. **41**, 1199 (1970).

[14] F. Gautier, in *Magnetism of Metals and Alloys*, edited by M. Cyrot (North-Holland, Amsterdam, 1982), p. 1.

[15] T. Moriya, *Spin Fluctuations in Itinerant Electron Magnetism* (Springer-Verlag, Berlin, 1985).

[16] R. Kubo, J. Phys. Soc. Jpn. **17**, 1100 (1962).

[17] S. Doniach and E. Sondheimer, *Green's Functions for Solid State Physicists* (Benjamin, London, 1974), p. 90.

[18] P. Anderson, Science **235**, 1196 (1987).

[19] K. Takegahara, Jpn. J. Appl. Phys. **26**, L437 (1987).

[20] I. Iguchi, H. Watanabe, Y. Kasai, T. Mochiku, A. Sugishita, and E. Yamaka, Jpn. J. Appl. Phys. **26**, L645 (1987).

[21] B. Batlogg, G. Kourouklis, W. Weber, R. J. Cava, A. Jayaraman, A. White, K. Short, L. Rupp, and E. Rietman, Phys. Rev. Lett. **59**, 912 (1987).