Composite-operator approach for the p-d-mixing model of oxide superconductors

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A formulation of the p-d-mixing model of oxide superconductors is presented by introducing composite operators that describe electronic excitations associated with the Cu-O covalent bond. A conduction band developing inside the charge-transfer gap with hole doping can be understood as the band formed by mixing among p and d electrons and these composite electronic excitations.

I. INTRODUCTION

The high-T_c superconductivity of cuprate oxide superconductors appears only in an intermediate metallic region close to the metal-insulator transition.[1] The electronic state realized in this region is one of the central issues to be clarified in order to understand the mechanism of the high-T_c superconductivity. From experimental analysis of photoemission,[2-6] electron-energy-loss spectroscopy,[7] x-ray absorption spectroscopy,[8,9] and infrared optical spectroscopy,[10-12] a highly correlated p-d-mixing model is known to be suitable[13] to describe the electronic state of these cuprate oxides. There are three important interactions to determine the electronic state of those systems, a strong intra-atomic Coulomb interaction at the Cu site (U term), a large p-d mixing (t term), and a short distant Coulomb interaction between p and d electrons (V term). The strong intra-atomic Coulomb repulsion[2-5,14-18] U induces the Hubbard splitting in the d level. The insulator gap is a charge-transfer gap[2,5,10-12,14-18] between p and the upper Hubbard d levels. Starting from the p-d-mixing model, there are many theoretical works[19-23] discussing the nature of the electronic state in oxide superconductors and the effects of the above-mentioned interactions. It has been argued that the model may be reduced into a simple Hubbard model,[24,25] a t-J model,[26] or a Heisenberg-Kondo model.[27,28] It is also argued that the realized normal state may be a conventional Fermi-liquid state,[29,30] a resonating-valence-bond state,[31] or a marginal Fermi-liquid state.[32] The nature of the electronic state still remains as one of the controversial subjects in the oxide superconductor.

In the previous papers[19] we investigated the doping dependence of the electronic state by starting from p and d electron atomic levels and by including the p-d mixing and dynamical corrections due to fluctuations. The analysis indicates that the electron density of states is transferred from the original p- and d-electron levels to the Fermi level inside the charge-transfer gap by forming a highly correlated coherent electron state which is a bound state of a p electron and fluctuations. However, the approach presented in Ref. 19 confronted a difficulty in the appearance of the negative density of states with increasing carrier doping because of insufficient inclusion of contributions from the coherent states developed. In this paper, we propose to deal with this bound state from a slightly different viewpoint.

The analysis of Ref. 19 indicates that the p-electron excitation near the Fermi level is very much influenced by charge and spin states of neighboring Cu ions due to large covalency between p and d electrons. The energy of a p-electron excitation at an O site may largely fluctuate, depending on states of neighboring Cu ions. In such a situation, it may be more suitable to introduce electronic excitations relating to both Cu and O ions, instead of treating only p- and d-electron excitation at each lattice as primarily independent ones. Then we are led to introduce composite electronic operators constructed from p- and d-electron operators in order to describe such electronic excitations associated with Cu-O bonds. The introduction of these composite operators does not mean adding operator degrees of freedom in the formalism but corresponds to a procedure of choosing suitable asymptotic fields, in the field-theoretical language, for bound states which appear in addition to fundamental particle states. Observed quasiparticles are expressed by linear combinations of those field operators. One may regard such electronic operators as to describe excitations from states constructed by configuration mixing, since they describe the excitations from the ground state composed of p-d covalent mixing states and surrounding charge and spin states.

Once one is allowed to introduce such composite electronic excitations, we immediately have several p electronic excitations associated with a Cu-O bond (or Cu-O cluster). In addition to the original p-electron excitation, we have p-electron excitations accompanied by charge and spin fluctuations of d electrons and those accompanied by charge-transfer fluctuations (i.e., p-electron–d-hole pair creations (or vice versa)). That is, near the p level, several possible excitation levels overlap and mix each other. We can expect a variety of phenomena due to change of mixing strength of each component according to physical parameters such as carrier intensity, temperature and gap energy, etc.

This paper aims at presenting the treatment of the above-mentioned composite electronic excitations in the framework of the operator formalism. Since operators describing composite excitations do not follow a simple algebra of the creation and annihilation operators, a simple diagram method based on Wick's theorem cannot be

applied. We will present a general method to identify the self-energy of composite excitations. There are several requirements which the self-energy must satisfy. The Hermiticity of the real part of electron propagators and the positive definiteness of the spectral functions are necessary requirements. If the above conditions are not satisfied, we have the violation of unitarity and easily have the negative density of states. The difficulty in improving the Hubbard approximation[33] by higher-order correction lies in the fact that one may easily confront the appearance of the negative density of states in the calculation. In our scheme, such a difficulty is handled by introducing a generalized self-energy. In the subsequent paper,[34] we will present results of detailed numerical calculation based on the formulation of this paper. We will see that complicated change of electronic states near the metal-insulator transition of oxide superconductors is more easily understood as mutual interactions among p and d electrons and composite electronic excitations on Cu-O bonds. It will be shown that the band at the Fermi level has, in fact, a large component of composite electronic excitations.

In the next section, the general framework is presented for the treatment of composite operators. The p-d-mixing model is used. A notion of generalized self-energies and generalized mean fields for composite excitations is introduced. In Sec. III, explicit forms of static parts of the self-energies (i.e., mean-field parts) are obtained in the p-d-mixing model. Dynamical corrections for the self-energies are evaluated. In Sec. IV, fluctuations which are necessary in the formula in Sec. IV are evaluated. In order to satisfy sum rules required from the operator algebra, the formulation by use of relaxation functions is developed. Section V is devoted to concluding remarks. There we shortly discuss what kinds of phenomena are expected from the present formula and clarify why we have to include the processes presented in Sec. III. The detailed numerical calculation using the present formalism will be presented in the subsequent paper.[34] The analysis including the V term will be presented elsewhere.[35,36]

II. COMPOSITE OPERATOR AND GENERALIZED SELF-ENERGY

Let us consider the p-d-mixing model of oxide superconductors without the V term. The model is given by the following Hamiltonian:

$$H=\int d\mathbf{x}[\tfrac{1}{2}\varepsilon_\eta \eta^\dagger(x)\cdot\eta(x)+\varepsilon_p p^\dagger(x)\cdot p(x) +p_t^\dagger(x)\cdot\eta(x)+\eta^\dagger(x)\cdot p_t(x)] , \tag{2.1}$$

where the electron operators are in the spinor notation, the operator $\eta(x)$ is given by the d electron operator as[37]

$$\eta_\sigma(x)=d_\sigma(x)d^\dagger_{-\sigma}(x)d_{-\sigma}(x) \tag{2.2}$$

with σ being a spin index, and

$$\varepsilon_\eta=\varepsilon_d+U \tag{2.3}$$

with ε_d being the energy of the d level and U being the intra-atomic Coulomb repulsion. The operator η represents the $n_d=1\leftrightarrow n_d=2$ restricted d electronic transition. We neglect the transition to the lower Hubbard level. The operator p_t is an abbreviation,

$$p_t(x)=t(-i\nabla)p(x) , \tag{2.4}$$

with

$$t^2(\mathbf{k})=4t^2\gamma^2(\mathbf{k}) , \tag{2.5a}$$

$$\gamma^2(\mathbf{k})=1+\gamma_1^2(\mathbf{k}) , \tag{2.5b}$$

$$\gamma_1^2(\mathbf{k})=-\tfrac{1}{2}(\cos k_x a+\cos k_y a) . \tag{2.5c}$$

The equations of motion for p and d electrons are obtained from the Heisenberg equation $i(\partial/\partial t)p_\sigma(x)=[p_\sigma(x),H]$ and $i(\partial/\partial t)\eta_\sigma(x)=[\eta_\sigma(x),H]$ by use of the anticommutation relation,

$$\{p_s(\mathbf{x}),p_{s'}^\dagger(\mathbf{y})\}=\delta(\mathbf{x}-\mathbf{y})\delta_{s's}$$

and

$$\{\eta_s(\mathbf{x}),\eta_{s'}^\dagger(\mathbf{y})\}=-\tfrac{1}{2}\sum_\mu(\sigma^\mu)_{ss'}\eta_{s'}(\mathbf{x})\delta(\mathbf{x}-\mathbf{y}) ,$$

$$i\frac{\partial}{\partial t}p(x)=\varepsilon_p p(x)+\eta_t(x) , \tag{2.6}$$

$$i\frac{\partial}{\partial t}\eta(x)=\varepsilon_\eta\eta(x)-\tfrac{1}{2}\sigma^\mu n_\mu(x)p_t(x) . \tag{2.7}$$

Here

$$\sigma^\mu=(-1,\boldsymbol{\sigma}),\quad \sigma_\mu=(1,\boldsymbol{\sigma}) \tag{2.8}$$

and

$$n_\mu(x)=d^\dagger(x)\sigma_\mu d(x) . \tag{2.9}$$

The component with $\mu=0$ represents the charge-density operator of the d electron and that with $\mu=1,2,3$ represents the spin-density operator. In Eq. (2.7), the summation with respect to μ is understood. In Eqs. (2.6) and (2.7), the spinor notation is used for electron fields and $\sigma^\mu p_t(x)$ means, for example,

$$(\sigma^\mu p_t(x))_s=\sum_{s'}(\sigma^\mu)_{ss'}p_{ts'}(x) .$$

We introduce a composite operator $p_\mu(x)$ defined by

$$p_\mu(x)=p_\gamma(x)\delta n_\mu(x) , \tag{2.10}$$

where $\delta n_\mu(x)=n_\mu(x)-\langle n_\mu(x)\rangle$ with $\langle\ \rangle$ being the thermal average and

$$p_\gamma(x)=\gamma(-i\nabla)p(x) . \tag{2.11}$$

In the previous papers,[19] we have shown that repetition of the p electron and fluctuation loops gives rise to a state at the Fermi level. We can regard the electronic excitation described by $p_\mu(x)$ as the p electronic excitation accompanying charge ($\mu=0$) and spin ($\mu=1,2,3$) fluctuations of neighboring d electrons. We treat, in the following, $p(x)$, $\eta(x)$, and $p_\mu(x)$ on an equal footing.

Let us introduce the following notation:

$$\psi_n(x)=\begin{bmatrix} p(x) \\ r(x) \\ p_s(x) \\ p_0(x) \end{bmatrix} , \tag{2.12}$$

where

$$r(x)=\sqrt{2/n}\,\eta(x) \tag{2.13}$$

with the normalization being determined to satisfy

$$\langle\{r(x),r^\dagger(y)\}\rangle=\delta(\mathbf{x}-\mathbf{y}) , \tag{2.14}$$

and

$$p_s(x)=\sigma p_\gamma(x)\mathbf{n}(x) \tag{2.15}$$

and

$$p_0(x)=p_\gamma(x)\delta n(x) . \tag{2.16}$$

The operator $p_s(x)$ represents the p electron excitation accompanying the Cu-spin fluctuation and $p_0(x)$ represents the p-electron excitation accompanying the Cu-charge fluctuation. In this section we develop a general scheme to obtain the propagator of $\psi_n(x)$. We will define the self-energy for the operator $\psi_n(x)$.

Let us suppose that we identify electronic excitations described by $\psi_n(x)$, in which certain composite operators are included. The choice of $\psi_n(x)$ arises from the physical consideration of which kinds of components describe the main distribution of the electron density of states. By use of the Heisenberg equation, we may have equations of motion in the following form:

$$i\frac{\partial}{\partial t}\psi_n(x)=j_n(x) . \tag{2.17}$$

Let us consider a retarded propagator $\langle R\psi_n(x)\psi_l^\dagger(y)\rangle$, where R indicates the retarded time order product and $\langle\ \rangle$ is the thermal average. The propagator $\langle R\psi_n(x)\psi_l^\dagger(y)\rangle$ satisfies the equation

$$i\frac{\partial}{\partial t_x}\langle R\psi_n(x)\psi_l^\dagger(y)\rangle=i\delta(t_x-t_y)\langle\{\psi_n(\mathbf{x}),\psi_l^\dagger(\mathbf{y})\}\rangle + \langle Rj_n(x)\psi_l^\dagger(y)\rangle . \tag{2.18}$$

Define a generalized self-energy by

$$\langle Rj_n(x)\psi_l^\dagger(y)\rangle=\Sigma_{nn'}(-i\partial_x)\langle R\psi_{n'}(x)\psi_l^\dagger(y)\rangle . \tag{2.19}$$

Here the summation over n' is understood. By denoting the Fourier transform of the propagator as $S_{nl}(\omega,\mathbf{k})$ and that of $\langle\{\varepsilon\psi_n(\mathbf{x}),\psi_l^\dagger(\mathbf{y})\}\rangle$ by $I_{nl}(\mathbf{k})$, we have

$$(\omega-\Sigma(\omega,\mathbf{k}))_{nn'}S_{n'l}(\omega,\mathbf{k})=I_{nl}(\mathbf{k}) . \tag{2.20}$$

Similarly, by use of the equation of motion for $\psi_l^\dagger$,

$$-i\frac{\partial}{\partial t_y}\langle R\psi_n(x)\psi_l^\dagger(y)\rangle=i\delta(t_x-t_y)\langle\{\psi_n(\mathbf{x}),\psi_l^\dagger(\mathbf{y})\}\rangle + \langle R\psi_n(x)j_l^\dagger(y)\rangle , \tag{2.21}$$

together with the definition

$$\langle R\psi_n(x)j_l^\dagger(y)\rangle=\Sigma_{l'l}^\dagger(i\partial_y)\langle R\psi_n(x)\psi_{l'}^\dagger(y)\rangle , \tag{2.22}$$

we have

$$S_{nl'}(\omega,\mathbf{k})(\omega-\Sigma^\dagger(\omega,\mathbf{k}))_{l'l}=I_{nl}(\mathbf{k}) . \tag{2.23}$$

The matrix $I_{nl}(\mathbf{k})$ determines the normalization of the electron propagator. Let us call the matrix I as the normalization matrix. Since the components of $\psi_n(x)$ contain composite operators, $I_{nl}(\mathbf{k})$ is not the identity matrix. Therefore, although $\Sigma(\omega,\mathbf{k})$ and $\Sigma^\dagger(\omega,\mathbf{k})$ satisfy the relation

$$\Sigma^\dagger(\omega,\mathbf{k})_{nl}=\{\Sigma(\omega^*,\mathbf{k})_{ln}\}^* , \tag{2.24}$$

Eq. (2.24) does not guarantee the positivity and hermiticity of the spectral functions. This indicates that, when one applies successive use of equations of motion to define certain mean fields and to include dynamical corrections, one easily violates the positivity of spectral functions, unless approximations are properly done. Specially the violation of Hermiticity in the real part of propagators in successive use of equations of motion is the serious problem.

Instead of calculating the self-energy itself, we consider more symmetrical functions with respect to field operators. First notice that, by applying Eq. (2.23) to Eq. (2.19) of the self-energy, we have

$$(\Sigma(\omega,\mathbf{k})I(\mathbf{k}))_{nl}=(\{j_n,\psi_l^\dagger\})+(Rj_n\delta j_l^\dagger) , \tag{2.25}$$

where the notation ( ) indicates the Fourier transform of the vacuum expectation value for the corresponding operator product and the variables ω and $\mathbf{k}$ are omitted. Here

$$\delta j_l^\dagger(y)=j_l^\dagger(y)-\Sigma_{l'l}^\dagger(i\partial_y)\psi_{l'}^\dagger(y) . \tag{2.26}$$

Similarly to Eq. (2.25), we have

$$(I(\mathbf{k})\Sigma^\dagger(\omega,\mathbf{k}))_{nl}=(\{\psi_n,j_l^\dagger\})+(R\delta j_n j_l^\dagger) , \tag{2.27}$$

where

$$\delta j_n(x)=j_n(x)-\Sigma_{nn'}(-i\partial_x)\psi_{n'}(x) . \tag{2.28}$$

By applying the time derivative from the left- and right-hand sides of the propagator $\langle R\psi_n(x)\psi_l^\dagger(y)\rangle$, we have

$$\omega S_{nl}(\omega,\mathbf{k})\omega=\omega I_{nl}+(\{j_n,\psi_l^\dagger\})+(Rj_nj_l^\dagger) \tag{2.29a}$$

$$=\omega I_{nl}+(\{\psi_n,j_l^\dagger\})+(Rj_nj_l^\dagger) , \tag{2.29b}$$

where Eq. (2.29a) is obtained by applying the right-hand-side time derivative first and Eq. (2.29b) vice versa. Therefore we have

$$(\{j_n,\psi_l^\dagger\})=(\{\psi_n,j_l^\dagger\})\ \ [\equiv(m_0)_{nl}] . \tag{2.30}$$

Further, since

$$(Rj_n\delta j_l^\dagger)=(R\delta j_n\delta j_l^\dagger)=(R\delta j_nj_l^\dagger) , \tag{2.31}$$

we have, from Eqs. (2.25), (2.27), and (2.30),

$$\Sigma(\omega,\mathbf{k})I(\mathbf{k})=I(\mathbf{k})\Sigma^\dagger(\omega,\mathbf{k})\ \ [\equiv m(\omega,\mathbf{k})] . \tag{2.32}$$

Now we have a set of functions which satisfy Hermiticity

requirements,

$$(I^{\dagger}(\mathbf{k}))_{kl}=(I(\mathbf{k}))^{*}_{lk} , \tag{2.33a}$$

$$(m_0^{\dagger}(\mathbf{k}))_{kl}=(m_0(\mathbf{k}))^{*}_{lk} , \tag{2.33b}$$

$$(m(\omega,\mathbf{k})^{\dagger})_{kl}=(m(\omega^{*},\mathbf{k}))^{*}_{lk} . \tag{2.33c}$$

Since the propagator is given in a form

$$S_{nl}(\omega,\mathbf{k})=\left[I(\mathbf{k})\frac{1}{\omega I(\mathbf{k})-m(\omega,\mathbf{k})}I(\mathbf{k})\right]_{nl} , \tag{2.34}$$

the propagator satisfies the symmetric requirement. Since I_{nl} is the positive definite matrix from its construction, the hermiticity of the real part and the positive definiteness of the spectral function is guaranteed from that of the dynamical part of $m(\omega,\mathbf{k})$, i.e.,

$$\delta m(\omega,\mathbf{k})_{nl}=(R\,\delta j_n \delta j_l^{\dagger}) . \tag{2.35}$$

Summarizing, the propagator of electron operators including composite operators is obtained from the normalization matrix $I(\mathbf{k})_{nl}$ given by

$$I(\mathbf{k})_{nl}=(\{\psi_n,\psi_l^{\dagger}\}) , \tag{2.36a}$$

the generalized mean field m_0 given by

$$(m_0(\mathbf{k}))_{nl}=(\{j_n,\psi_l^{\dagger}\})=(\{\psi_n,j_l^{\dagger}\}) \tag{2.36b}$$

and the dynamical correction

$$(\delta m(\omega,\mathbf{k}))_{nl}=(R\,\delta j_n \delta j_l^{\dagger}) \tag{2.36c}$$

as

$$S_{nl}(\omega,\mathbf{k})=\left[I(\mathbf{k})\frac{1}{\omega I(\mathbf{k})-[m_0(\mathbf{k})+\delta m(\omega,\mathbf{k})]}I(\mathbf{k})\right]_{nl} . \tag{2.37}$$

In the following sections, we evaluate the matrices $I(\mathbf{k})$, $m_0(\mathbf{k})$, and $\delta m(\omega,\mathbf{k})$ in the p-d-mixing model of oxide superconductors. In the Appendix, we present an exactly solvable example in the framework of this section.

III. NORMALIZATION MATRIX, MEAN FIELDS, AND DYNAMICAL CORRECTION

Let us identify electron operators of electronic excitations in the Cu-O cluster as $p(x)$, $r(x)[=\sqrt{2/n}\,\eta(x)]$, $p_s(x)$ $[=\sigma p_\gamma(x)\mathbf{n}(x)]$, and $p_0(x)$ $[=p_\gamma(x)\delta n(x)]$. Here the spinor notation is used for electron fields and $\sigma p_\gamma(x)$, for example, indicates

$$(\sigma p_\gamma(x))_s=\sum_{s'}(\sigma)_{ss'}p_{\gamma s'}(x) .$$

They satisfy the equation of motion obtained from the Hamiltonian (2.1),

$$i\frac{\partial}{\partial t}p(x)=\varepsilon_p p(x)+t_n r_\gamma(x) , \tag{3.1a}$$

$$i\frac{\partial}{\partial t}r(x)=t_n p_\gamma(x)+\varepsilon_\eta r(x)-\frac{t_n}{n}(p_s(x)-p_0(x)) , \tag{3.1b}$$

$$i\frac{\partial}{\partial t}p_s(x)=\varepsilon_p p_s(x)+t_n(h_s(x)-\psi_s(x)+\varphi_s(x)) , \tag{3.1c}$$

and

$$i\frac{\partial}{\partial t}p_0(x)=\varepsilon_p p_0(x)+t_n\langle 2-n\rangle r(x)+t_h(h_0(x)-\psi_0(x)+\varphi_0(x)) , \tag{3.1d}$$

where

$$t_n=2t_0\sqrt{n/2} , \tag{3.2}$$

$$h_s(x)=\sigma r_{\gamma_1^2}(x)\delta\mathbf{n}(x),\quad h_0(x)=r_{\gamma_1^2}(x)\delta n(x) , \tag{3.3a}$$

$$\psi_s(x)=\sigma p_\gamma(x)\delta\mathbf{n}_+(x),\quad \psi_0(x)=p_\gamma(x)\delta n_+(x) , \tag{3.3b}$$

and

$$\varphi_s(x)=\sigma p_\gamma(x)\delta\mathbf{n}_-(x),\quad \varphi_0(x)=p_\gamma(x)\delta n_-(x) \tag{3.3c}$$

with

$$\begin{aligned}\delta n_{+\mu}(x)&=p_\gamma^{\dagger}(x)\sigma_\mu r(x)-\langle p_\gamma^{\dagger}(x)\sigma_\mu r(x)\rangle ,\\ \delta n_{-\mu}(x)&=r^{\dagger}(x)\sigma_\mu p_\gamma(x)-\langle r^{\dagger}(x)\sigma_\mu p_\gamma(x)\rangle .\end{aligned} \tag{3.4}$$

It should be noted that, to obtain Eq. (3.1), any operator product of d-electron operators at the equal site must be reduced to independent freedom by use of the algebraic relations,

$$r_\sigma r_s^{\dagger}=\left[-1+\frac{2}{\langle n\rangle}-\frac{1}{\langle n\rangle}\delta n-\frac{1}{\langle n\rangle}\mathbf{n}\cdot\sigma\right]_{\sigma s} , \tag{3.5a}$$

$$r_s^{\dagger}r_\sigma=2\left[1-\frac{1}{\langle n\rangle}+\frac{1}{\langle n\rangle}\delta n\right]_{\sigma s} , \tag{3.5b}$$

$$r_\sigma r_s=r_\sigma^{\dagger}r_s^{\dagger}=0 , \tag{3.5c}$$

$$\delta n r_\sigma=(1-\langle n\rangle)r_\sigma,\quad r_\sigma\delta n=r_\sigma(2-\langle n\rangle) , \tag{3.6a}$$

$$n_i r_\sigma=-\sigma_i r_\sigma ,\quad r_\sigma n_i=0 , \tag{3.6b}$$

$$r_\sigma^{\dagger}\delta n=r_\sigma^{\dagger}(1-\langle n\rangle),\quad \delta n r_\sigma^{\dagger}=(2-\langle n\rangle)r_\sigma^{\dagger} , \tag{3.6c}$$

$$r_\sigma^{\dagger}n_i=-r_\sigma^{\dagger}\sigma_i,\quad n_i r_\sigma^{\dagger}=0 , \tag{3.6d}$$

$$n^2=n+2n_\uparrow n_\downarrow , \tag{3.7a}$$

$$n n_i=n_i , \tag{3.7b}$$

and

$$n_i n_j=\delta_{ij}n+i\epsilon_{ijk}n_k=2\delta_{ij}n_\uparrow n_\downarrow . \tag{3.7c}$$

We have an additional restriction

$$n_\uparrow n_\downarrow=n-1 \tag{3.8}$$

in the present approximation, i.e., in the restriction on $n=1\leftrightarrow 2$ transition $[(n-1)(n-2)=0]$.

As for the components of the electronic operator ψ_n, we have identified the operators in Eqs. (3.3a)–(3.3c). Inclusion of more composite operators means enlargement of the would-be quasitable excitations in the cluster under consideration. In the case to solve a finite-size cluster exactly, it is necessary to include all possible independent

electronic composite operators which mix in equations of motion. Since operators describe transitions among states, the number of operators is larger than that of states. Then the exact diagonalization with respect to state vectors may be more economical. However, when the lattice case is considered and intersite interactions become important, the present description may be more powerful, since concerned excitations can be traced lattice by lattice. It is suitable to describe competition between locality and itinerancy of electrons. In the present approximation, we assume that the units up to Cu, O, and Cu-O bond play main roles to determine the electronic state and assume that a unit O-Cu-O, for example, is expressed by a combination of them. In the case that the local nature is strong, one needs to introduce more composite operators in the present scheme.

A. Normalization matrix I

Once the electronic operator ψ_n is identified, it is straightforward to obtain the matrix I given by $\langle\{\psi_n,\psi_l^\dagger\}\rangle$. In this calculation, the algebraic relations (3.5)–(3.8) must be fully taken into account. We have

$$I_{nl}=\begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 3b & b \\ 0 & 3b & 4a_s+3(\chi_s+\chi_s'\gamma_1^2(\mathbf{k})) & 0 \\ 0 & b & 0 & \chi_0+\chi_0'\gamma_1^2(\mathbf{k}) \end{bmatrix}, \tag{3.9}$$

where

$$b=\langle p_\gamma r^\dagger\rangle=\langle rp_\gamma^\dagger\rangle\ , \tag{3.10}$$

$$a_s=\langle p_\gamma p_s^\dagger\rangle=\langle p_\gamma p_s^\dagger\rangle\ , \tag{3.11}$$

$$\chi_s=\langle\delta n_i\delta n_i\rangle=\langle 2-n\rangle\ , \tag{3.12}$$

$$\chi_0=\langle\delta n\delta n\rangle=\langle 2-n\rangle\langle n-1\rangle\ , \tag{3.13}$$

$$\chi_s'=-\langle(\gamma_1^2\delta n_i)\delta n_i\rangle\ , \tag{3.14}$$

and

$$\chi_0'=-\langle(\gamma_1^2\delta n)\delta n\rangle\ . \tag{3.15}$$

Here we have used the fact that

$$\gamma_1^2[\mathbf{x}-\mathbf{y}]=\frac{\Omega}{(2\pi)^2}\int d^2k\,\gamma_1^2(\mathbf{k})e^{i\mathbf{k}\cdot(\mathbf{x}-\mathbf{y})}$$

with

$$\gamma_1^2(\mathbf{k})=-\tfrac{1}{2}(\cos k_x a+\cos k_y)$$

is nonvanishing only at the nearest neighbor.

B. Mean field

The mean field m_0 is calculated from $\langle\{j_n,\psi_l^\dagger\}\rangle$. Although m_0 should be a Hermitian matrix, an expression of the vacuum expectation value does not obviously satisfy this condition. This is because the Hermiticity of m_0 is satisfied only after taking the vacuum expectation value for the correct ground state. In fact, when we evaluate m_0 by the anticommutator $\langle\{j_n,\psi_l^\dagger\}\rangle$, we have for $(m_0)_{rp_s^\dagger}$,

$$(m_0)_{rp_s^\dagger}=3\epsilon_\eta b-\frac{t_n}{n}[4a_s+3(\chi_s+\chi_s'\gamma_1^2(\mathbf{k}))] \tag{3.16}$$

and for $(m_0)_{p_sr^\dagger}$,

$$(m_0)_{p_sr^\dagger}=3\varepsilon_p b-t_n\left[3a+\frac{3}{n}a_0+\frac{1}{n}a_s+3a_r'+\frac{3}{n}\chi_s'\gamma_1^2(\mathbf{k})\right], \tag{3.17}$$

where

$$a=\langle p_\gamma p_\gamma^\dagger\rangle\ , \tag{3.18}$$

$$a_r'=-\langle(\gamma_1^2 r)r^\dagger\rangle\ , \tag{3.19}$$

and

$$a_0=\langle p_\gamma p_0^\dagger\rangle=\langle p_0 p_\gamma^\dagger\rangle\ . \tag{3.20}$$

This fact may be one of the reasons why the inclusion of higher-order corrections is difficult in a straightforward decoupling method in successive use of equations of motion. In the course of solving the equations, there is no guarantee for m_0 to be Hermitian, when one uses straightforwardly the expression of m_0 obtained from the equations of motion. When one deals with a non-Hermitian matrix, one easily has a nonunitary transformation in the procedure of diagonalization and the unitarity is violated. In order to avoid this situation, we use the hermiticity of m_0 positively as the requirement. For example, since $(m_0)_{rp_s^\dagger}$ and $(m_0)_{p_sr^\dagger}$ should be complex conjugate to each other for the correct solution, we use only the expression for $(m_0)_{rp_s^\dagger}$. The rule of the approximation is as follows. By successive use of equations of motion, there appear composite operators and its source term j_n composed of higher-order operator products. The evaluation of the mean field becomes less accurate due to an approximation introduced to evaluate such higher-order operator products. By using the Hermiticity relation, the mean field m_0 is evaluated from source terms with lower-order operator products. For any operator product of d-electron operators at the equal site, the algebraic relations (3.5)–(3.8) are considered. Then we have

$$(m_0)_{pp^\dagger}=\varepsilon_p\ , \tag{3.21}$$

$$(m_0)_{pr^\dagger}=t_n\gamma(\mathbf{k})\ , \tag{3.22}$$

$$(m_0)_{pp_s^\dagger}=3t_n b\gamma(\mathbf{k})\ , \tag{3.23}$$

$$(m_0)_{pp_0^\dagger}=t_n b\gamma(\mathbf{k})\ , \tag{3.24}$$

$$(m_0)_{rr^\dagger}=\varepsilon_\eta-2b\frac{t_n}{n}\ , \tag{3.25}$$

$$(m_0)_{rp_s^\dagger}=3\varepsilon_\eta b-\frac{t_n}{n}[3(\chi_s+\chi_s'\gamma_1^2(\mathbf{k}))+4a_s]\ , \tag{3.26}$$

$$(m_0)_{rp_0^\dagger}=\varepsilon_\eta b+\frac{t_n}{n}(\chi_0+\chi_0'\gamma_1^2(\mathbf{k}))\ , \tag{3.27}$$

$$(m_0)_{p_sp_s^\dagger}=\varepsilon_p[3(\chi_s+\chi_s'\gamma_1^2(\mathbf{k}))+4a_s]$$
$$+t_n(4b_s'+6b(a-1)-\tfrac{3}{2}(\chi'_{+0s}-\chi'_{-0s})\gamma_1^2(\mathbf{k}))\ , \tag{3.28}$$

$$(m_0)_{p_sp_0^\dagger}=t_n(-6ab+(2-n)3b-\tfrac{3}{2}(\chi'_{+0s}+\chi'_{0-\rho})\gamma_1^2(\mathbf{k}))\ , \tag{3.29}$$

and

$$(m_0)_{p_0p_0^\dagger}=\varepsilon_p(\chi_0+\chi_0'\gamma_1^2(\mathbf{k}))$$
$$+t_n[\chi_s b+b(2a-n)$$
$$-\tfrac{1}{2}(3\chi'_{0+\rho}+\chi'_{0-\rho})\gamma_1^2(\mathbf{k})]\ , \tag{3.30}$$

where

$$b_s'=-\langle r_{\gamma_1^2}p_s^\dagger\rangle=-\langle p_s r_{\gamma_1^2}^\dagger\rangle\ , \tag{3.31}$$

$$\chi'_{+0\mu}=-\langle\gamma_1^2\delta n_{+\mu}\delta n_\mu\rangle\ , \tag{3.32}$$

and

$$\chi'_{-0\mu}=-\langle\gamma_1^2\delta n_{-\mu}\delta n_\mu\rangle\ . \tag{3.33}$$

The other half of the components in the matrix m_0 are given by the Hermiticity relations in Eq. (2.33b). It should be noted that the k dependence of the mean fields appears from the nearest-neighbor spin and charge fluctuations. This may be understood as the process in which a Cu-O covalent electronic state rearranges into that of the nearest-neighbor Cu-O bond, leaving spin and charge fluctuations. One may regard this process as a covalent electronic excitation hopping around the crystal.

C. Dynamical correction

The dynamical correction is obtained from

$$\delta m(\omega,\mathbf{k})_{nl}=(R\,\delta j_n\delta j_l^\dagger)\ . \tag{3.34}$$

In the present scheme, we have assumed that the quasistable electronic excitations are composed of p, r, and p_μ and that they produce the main peak structures in the intensity distribution of the density of states. The dynamical correction is given by decay processes of those electronic excitations. Here we evaluate it by the one-loop contributions composed of electronic excitations under consideration and fluctuations. Since δm contains no reducible ψ_n lines, it has only components for p_s and p_0. Loops of the dynamical correction are composed from the source in Eq. (3.1),

$$:j_s(x):=t_n:h_s(x)-\psi_s(x)+\varphi_s(x):\tag{3.35a}$$

and

$$:j_0(x):=t_n:h_0(x)-\psi_0(x)+\varphi_0(x):\ , \tag{3.35b}$$

where the normal product indicates the irreducible components. These interactions describe decays to continuums of electronic excitations and fluctuations. We have

$$\delta m_{\mu\nu}(x-y)=t_n^2\{\langle R:h_\mu(x)::h_\nu^\dagger(y):\rangle-\langle R:h_\mu(x)::\psi_\nu^\dagger(y):\rangle+\langle R:h_\mu(x)::\varphi_\nu^\dagger(y):\rangle$$
$$-\langle R:\psi_\mu(x)::h_\nu^\dagger(y):\rangle+\langle R:\psi_\mu(x)::\psi_\nu^\dagger(y):\rangle-\langle R:\psi_\mu(x)::\varphi_\nu^\dagger(y):\rangle$$
$$+\langle R:\varphi_\mu(x)::h_\nu^\dagger(y):\rangle-\langle R:\varphi_\mu(x)::\psi_\nu^\dagger(y):\rangle+\langle R:\varphi_\mu(x)::\varphi_\nu^\dagger(y):\rangle\}\ . \tag{3.36}$$

We neglect the interface among different decay processes (i.e., only diagonal processes are taken) and use the one-loop approximation. By use of the notation

$$R(\langle A_1(x)B_1(y)\rangle\cdots\langle A_n(x)B_n(y)\rangle)\equiv\Theta(t_x-t_y)\langle A_1(x)B_1(y)\rangle\cdots\langle A_n(x)B_n(y)\rangle$$
$$\pm\Theta(t_y-t_x)\langle B_1(y)A_1(x)\rangle\cdots\langle B_n(y)A_n(x)\rangle\ , \tag{3.37}$$

with + for a bosonic $A_1\cdots A_n$ and − for a fermionic $A_1\cdots A_n$, the contributions from $\langle R:h_\mu(x)::h_\nu^\dagger(y):\rangle$, $\langle R:\psi_\mu(x)::\psi_\nu^\dagger(y):\rangle$, and $\langle R:\varphi_\mu(x)::\varphi_\nu^\dagger(y):\rangle$ are evaluated as

$$\langle R:h_\mu(x)::h_\nu^\dagger(y):\rangle\approx R(\langle r_{\gamma_1^2}(x)r_{\gamma_1^2}^\dagger(y)\rangle\langle\delta n_\mu(x)\delta n_\nu(y)\rangle)\ , \tag{3.38}$$

$$\langle R:\psi_\mu(x)::\psi_\nu^\dagger(y):\rangle\approx R(\langle p_\gamma(x)p_\gamma^\dagger(y)\rangle\langle\delta n_{-\mu}(x)\delta n_{+\nu}(y)\rangle)+R(\langle p_\gamma(x)r^\dagger(y)\rangle\sigma_\nu\langle\delta n_{-\mu}(x)p_\gamma(y)p_\gamma^\dagger(y)\rangle)\ , \tag{3.39}$$

and

$$\langle R:\varphi_\mu(x)::\varphi_\nu^\dagger(y):\rangle\approx R(\langle p_\gamma(x)p_\gamma^\dagger(y)\rangle\langle\delta n_{+\mu}(x)\delta n_{-\nu}(y)\rangle)-\tfrac{1}{2}R(\langle p_\gamma(x)p_\gamma^\dagger(y)\rangle\sigma_\mu\sigma_\lambda\langle\delta n_{+\mu}(x)\delta n_{-\lambda}(y)\rangle)$$
$$-\tfrac{1}{2}R(\langle p_\gamma(x)p_\gamma^\dagger(y)\rangle\langle\delta n_{+\lambda}(x)\delta n_{-\nu}(y)\rangle)\sigma_\lambda\sigma_\mu$$
$$+\tfrac{1}{4}R(\langle p_\gamma(x)p_\gamma^\dagger(y)\rangle\sigma_\lambda\sigma_\mu\sigma_\nu\sigma_{\lambda'}\langle\delta n_{-\lambda}(x)\delta n_{+\lambda'}(y)\rangle)$$
$$+R(\langle p_\gamma(x)p_\gamma^\dagger(y)\rangle\langle p_\gamma(x)p_\gamma^\dagger(y)\rangle\langle r(y)r^\dagger(x)\rangle)(2\delta_{\mu\nu}-\sigma_\nu\sigma_\mu)\ , \tag{3.40}$$

where the last term in Eq. (3.40) arises to adjust double counting of a p-p-r three-body Fermion loop. We have

$$\delta m_{ss}=\sigma_i \delta m_{ij}\sigma_j \ , \tag{3.41a}$$

$$\delta m_{s0}=\sigma_i \delta m_{i0} \tag{3.41b}$$

and δm_{00} given by $\delta m_{\mu\nu}$ with $\mu=\nu=0$.

In the next section, the fluctuations $\langle R\,\delta n_\mu(x)\delta n_\nu(y)\rangle$ that appear in Eqs. (3.38)–(3.40) will be evaluated.

IV. FLUCTUATIONS

In the formulas presented in the previous sections, we have correlation functions of fluctuation

$$\delta n_{l_\mu}(x)=\begin{bmatrix}\delta n_\mu(x)\\ \delta n_{+\mu}(x)\\ \delta n_{-\mu}(x)\end{bmatrix} , \tag{4.1}$$

where $l=0,\pm$ and

$$\delta n_{+\mu}(x)=p_\gamma^\dagger(x)\sigma_\mu r(x)-\langle p_\gamma^\dagger(x)\sigma_\mu r(x)\rangle$$

and

$$\delta n_{-\mu}(x)=r^\dagger(x)\sigma_\mu p_\gamma(x)-\langle r^\dagger(x)\sigma_\mu p_\gamma(x)\rangle \ . \tag{4.2}$$

In this section, we derive the formula to obtain those fluctuations.

The equations of motion for $\delta n_{l_\mu}(x)$ are given by

$$i\frac{\partial}{\partial t}\delta n_\mu(x)=t_n(\delta n_{-\mu}(x)-\delta n_{+\mu}(x))\equiv j_{0\mu}(x) \ , \tag{4.3a}$$

$$i\frac{\partial}{\partial t}\delta n_{+\mu}(x)=(\varepsilon_\eta-\varepsilon_p)\delta n_{+\mu}(x)+c_\mu\delta n_\mu(x)+\delta j_{+\mu}(x)\equiv j_{+\mu}(x) \ , \tag{4.3b}$$

and

$$-i\frac{\partial}{\partial t}\delta n_{-\mu}(x)=(\varepsilon_\eta-\varepsilon_p)\delta n_{-\mu}(x)+c_\mu\delta n_\mu(x)+\delta j_{-\mu}(x)\equiv j_{-\mu}(x) \ , \tag{4.3c}$$

where

$$c_\mu=-2\frac{t_n}{n}(\delta_{\mu0}(1+a)+\delta_{\mu i}(1-a)) \ , \tag{4.4}$$

with

$$a=\langle p_\gamma p_\gamma^\dagger\rangle \ , \tag{4.5}$$

and

$$\delta j_{+\mu}(x)=t_n{:}\left[p_\gamma^\dagger(x)\sigma_\mu p_\gamma(x)-\frac{1}{n}p_\gamma^\dagger(x)\sigma_\mu\sigma^\lambda p_\gamma(x)\delta n_\lambda(x)-r_{\gamma_1^2}^\dagger(x)\sigma_\mu r(x)\right]{:} \ , \tag{4.6a}$$

$$\delta j_{-\mu}(x)=(\delta j_{+\mu}(x))^\dagger \tag{4.6b}$$

with the normal product indicating the subtraction of obvious paired vacuum expectation values.

A. Intersite correlations

In the mean fields, there appear correlations with the nearest-neighbor fluctuations $\delta n_\mu(x)$. We approximate those intersite correlations by a one-loop fermion loop. For example, χ'_μ in Eqs. (3.14) and (3.15),

$$\chi'_\mu=\langle \delta n_\mu(\mathbf{x}')\delta n_\mu(\mathbf{x})\rangle \ , \tag{4.7}$$

are evaluated as

$$\chi'_\mu=\int d\omega\frac{1}{\omega^2}\left[-\frac{1}{\pi}\right]\mathrm{Im}(Rj_{0\mu}(\mathbf{x})j_{0\mu}^\dagger(\mathbf{x}')) \tag{4.8}$$

with $\langle Rj_{0\mu}(x)j_{0\mu}^\dagger(x')\rangle$ being approximated as

$$\langle Rj_{0\mu}(x)j_{0\mu}^\dagger(x')\rangle=2t_n^2[R(\langle p_\gamma(x)p_\gamma^\dagger(x')\rangle\langle r^\dagger(x)r(x')\rangle)-R(\langle p_\gamma(x)r^\dagger(x')\rangle\langle r^\dagger(x)p_\gamma(x')\rangle)-R(\langle r(x)p_\gamma^\dagger(x')\rangle\langle p_\gamma^\dagger(x)r(x')\rangle)+R(\langle r(x)r^\dagger(x')\rangle\langle p_\gamma^\dagger(x)p_\gamma(x')\rangle)] \ . \tag{4.9}$$

Note that

$$\langle Rp_\gamma(x)p_\gamma^\dagger(x')\rangle=\frac{i}{2\pi}\int d\omega e^{-i\omega(t-t')}\frac{\Omega}{(2\pi)^2}\times\int d^2k\gamma^2(\mathbf{k})S_{pp}^\dagger(\omega,\mathbf{k}) \ . \tag{4.10}$$

B. Intrasite fluctuations

As for fluctuations appearing in calculations of dynamical corrections $\delta m(\omega,\mathbf{k})$, intensity distributions in the energy variable are more important, since composite electronic excitations are always with fluctuations and their energies are affected by those of fluctuations. In this paper, we approximate dynamical corrections by decays of local excitations, since primary effects to composite excitations are expected to be local. For local fluctuations, the spin and charge at a single site correlate strongly themselves. This is because there are constraints from the local algebra which restrict whole weights of fluctuations in forms of sum rules. In order to obtain dynamical behaviors under such restrictions of sum rules, it is convenient to use the relaxation function rather than the response function. For an operator A, let us use a notation $\int A$, defined by

$$\int A=\int_0^\beta d\lambda\, A(t+i\lambda) \ . \tag{4.11}$$

A relaxation function for arbitrary operators A and $B^\dagger$ is

given by

$$\left\langle A(t)\int B^{\dagger}(t')\right\rangle_{\Theta}=\Theta(t-t')\left\langle A(t)\int B^{\dagger}(t')\right\rangle . \tag{4.12}$$

By use of the spectral functions $\rho_{AB}(\omega)$ of the local response function $\langle R\,A(t)B^{\dagger}(t')\rangle$, we have

$$\left\langle A(t)\int B^{\dagger}(t')\right\rangle_{\Theta}=\frac{i}{2\pi}\int d\omega e^{-i\omega(t-t')} \times\int d\kappa\frac{\rho_{AB}(\kappa)}{\kappa}\frac{1}{\omega-\kappa+i\delta} . \tag{4.13}$$

It should be noted that we have for $t=t'$

$$\left\langle A\int B^{\dagger}\right\rangle^{*}=\left\langle B\int A^{\dagger}\right\rangle , \tag{4.14a}$$

$$\left\langle A\int B^{\dagger}\right\rangle=\left\langle B^{\dagger}\int A\right\rangle , \tag{4.14b}$$

and

$$\left\langle A\int A^{\dagger}\right\rangle\geq 0 . \tag{4.14c}$$

Also the spectral function $\rho_{AB}(\omega)$ is given by

$$\int d\omega e^{-i\omega(t-t')}\rho_{AB}(\omega)=\langle[A(t),B^{\dagger}(t')]\rangle , \tag{4.15}$$

and it satisfies the relation

$$\rho_{AB}(\omega)=\rho_{BA}(\omega)^{*}=-\rho_{B^{\dagger}A^{\dagger}}(-\omega)=-\rho_{A^{\dagger}B^{\dagger}}(-\omega)^{*} , \tag{4.16}$$

which gives similar relations among relaxation functions through Eq (4.13). Define operators $\delta N_{l_\mu}(x)$ $(l=0,\pm)$ as

$$\delta N_{l_\mu}(x)=\delta n_{l_\mu}(x)-\alpha_l^{\mu}\delta n_{\mu}(x) , \tag{4.17}$$

where $\alpha_0^{\mu}=0$ and $\alpha_{\pm}^{\mu}=\alpha_{\mu}$ are determined by

$$\left\langle\delta n_{+\mu}\int\delta n_{\mu}\right\rangle=\alpha_{\mu}\left\langle\delta n_{\mu}\int\delta n_{\mu}\right\rangle . \tag{4.18}$$

With this definition, we have

$$\left\langle\delta N_{\mu}\int\delta N_{+\mu}\right\rangle=\left\langle\delta N_{\mu}\int\delta N_{-\mu}\right\rangle=0 . \tag{4.19}$$

Let us write the equations of motion of $\delta N_{l_\mu}(x)$ in the form

$$i\frac{\partial}{\partial t}\delta N_{l_\mu}(x)=\epsilon_l J_{l_\mu}(x) , \tag{4.20}$$

with $\epsilon_l=1$ (for $l=0,+$) and -1 (for $l=-$), where

$$J_{0\mu}(x)=t_n(\delta N_{-\mu}(x)-\delta N_{+\mu}(x)) , \tag{4.21a}$$

$$J_{+\mu}(x)=(c_{\mu}+(\varepsilon_{\eta}-\varepsilon_{p})\alpha_{\mu})\delta N_{\mu}(x) +(\varepsilon_{\eta}-\varepsilon_{p}+t_n\alpha_{\mu})\delta N_{+\mu}(x) -t_n\alpha_{\mu}\delta N_{-\mu}(x)+\delta j_{+\mu}(x) \tag{4.21b}$$

and

$$J_{-\mu}(x)=[J_{+\mu}(x)]^{\dagger} . \tag{4.21c}$$

Applying the equations of motion (4.20) to the relaxation function of $\delta N_{l_\mu}(x)$, we have

$$i\frac{\partial}{\partial t}\left\langle\delta N_{l_\mu}(t)\int\delta N_{k_\mu}^{\dagger}(t')\right\rangle_{\Theta}=i\delta(t-t')\left\langle\delta N_{l_\mu}\int\delta N_{k_\mu}^{\dagger}\right\rangle +\epsilon_l\left\langle J_{l_\mu}(t)\int\delta N_{k_\mu}^{\dagger}(t')\right\rangle_{\Theta} . \tag{4.22}$$

Define the decay rate $\Sigma^{\mu}(\omega)_{lk}$ as

$$\epsilon_l\left\langle J_{l_\mu}(t)\int\delta N_{k_\mu}^{\dagger}(t')\right\rangle_{\Theta} \equiv\Sigma^{\mu}(i\partial_t)_{ll'}\left\langle\delta N_{l'_\mu}(t)\int\delta N_{k_\mu}^{\dagger}(t')\right\rangle_{\Theta} . \tag{4.23}$$

Then we have the Fourier transform $(\delta N_{l_\mu}\int\delta N_{k_\mu}^{\dagger})$ of $\langle\delta N_{l_\mu}(t)\int\delta N_{k_\mu}^{\dagger}(t')\rangle_{\Theta}$ as

$$(\delta N_{l_\mu}\int\delta N_{k_\mu}^{\dagger})=\left[\frac{1}{\omega-\Sigma^{\mu}(\omega)}F^{\mu}\right]_{lk} \tag{4.24}$$

with F_{lk}^{μ} being given by

$$F_{lk}^{\mu}=\left\langle\delta N_{l_\mu}\int\delta N_{k_\mu}^{\dagger}\right\rangle . \tag{4.25}$$

The spectral relation (4.13) together with the relation (4.16) leads to the matrix structure of F^{μ} as

$$F^{\mu}=\begin{bmatrix}F_{00}^{\mu} & 0 & 0\\ 0 & F_{++}^{\mu} & F_{+-}^{\mu}\\ 0 & F_{+-}^{\mu *} & F_{++}^{\mu}\end{bmatrix} . \tag{4.26}$$

Similarly, defining

$$\left\langle\delta N_{l_\mu}(t)\int J_{k_\mu}^{\dagger}(t')\right\rangle_{\Theta}\epsilon_k \equiv\Sigma^{\mu\dagger}(-i\partial_{t'})_{k'k}\left\langle\delta N_{l_\mu}(t)\int\delta N_{k'_\mu}^{\dagger}(t')\right\rangle_{\Theta} , \tag{4.27}$$

we have

$$(\delta N_{l_\mu}\int\delta N_{k_\mu}^{\dagger})=\left[F^{\mu}\frac{1}{\omega-\Sigma^{\mu\dagger}(\omega)}\right]_{lk} . \tag{4.28}$$

From Eqs. (4.23) and (4.27), we have

$$(\Sigma^{\mu}(\omega)F^{\mu})_{lk}=\epsilon_l\left\langle J_{l_\mu}\int\delta N_{k_\mu}^{\dagger}\right\rangle+\epsilon_l(J_{l_\mu}\int\delta J_{k_\mu}^{\dagger})\epsilon_k , \tag{4.29a}$$

$$(F^{\mu}\Sigma^{\mu\dagger}(\omega))_{lk}=\left\langle\delta N_{l_\mu}\int J_{k_\mu}^{\dagger}\right\rangle\epsilon_k+\epsilon_l(\delta J_{l_\mu}\int J_{k_\mu}^{\dagger})\epsilon_k , \tag{4.29b}$$

where

$$\delta J_{k_\mu}^{\dagger}(t)\epsilon_k\equiv J_{k_\mu}^{\dagger}(t)\epsilon_k-\Sigma^{\mu\dagger}(-i\partial_t)_{k'k}\delta N_{k'_\mu}^{\dagger}(t) , \tag{4.30a}$$

$$\epsilon_l\delta J_{l_\mu}(t)\equiv\epsilon_l J_{l_\mu}(t)-\Sigma^{\mu}(i\partial_t)_{ll'}\delta N_{l'_\mu}(t) . \tag{4.30b}$$

In Eq. (4.30), Σ^{μ} and $\Sigma^{\mu\dagger}$ operate from outside of relaxation functions. By use of the commutativity of the time derivatives, we can show, following a similar procedure used in Sec. II,

$$\epsilon_l(J_{l_\mu}\int \delta N^{\dagger}_{k_\mu})=(\delta N_{l_\mu}\int J^{\dagger}_{k_\mu})\epsilon_k \ , \tag{4.31}$$

which defines the mean fields as

$$(M_0)^{\mu}_{lk}=\epsilon_l(J_{l_\mu}\int \delta N^{\dagger}_{k_\mu}) \ , \tag{4.32}$$

satisfying

$$(M_0)^{\mu}_{lk}=((M_0)^{\mu}_{kl})^* \ . \tag{4.33}$$

According to the spectral representation (4.13) and by using $(i\partial/\partial t)\delta N_{l_\mu}(x)=\epsilon_l J_{l_\mu}(x)$, we have the matrix structure of M^{μ}_{0lk} as

$$M^{\mu}_0=\begin{bmatrix} 0 & (M^{\mu}_0)_{0+} & -(M^{\mu}_0)^*_{0+} \\ (M^{\mu}_0)^*_{0+} & (M^{\mu}_0)_{++} & 0 \\ -(M^{\mu}_0)_{0+} & 0 & -(M^{\mu}_0)_{++} \end{bmatrix} . \tag{4.34}$$

The dynamical correction of the mean field, $\delta M^{\mu}(\omega)_{lk}$, is given by

$$\delta M^{\mu}(\omega)_{lk}=\epsilon_l(\delta J_{l_\mu}\int \delta J^{\dagger}_{k_\mu})\epsilon_k \ , \tag{4.35}$$

which satisfies the relation

$$\delta M^{\mu}(\omega)_{lk}=(\delta M^{\mu}(\omega^*)_{kl})^* \ . \tag{4.36}$$

Since δJ_{l_μ} should not contain any reducible $\delta N_{l'_\mu}$ components, we have

$$\delta M^{\mu}(\omega)=\begin{bmatrix} 0 & 0 & 0 \\ 0 & \delta M^{\mu}_{++}(\omega+i\delta) & \delta M^{\mu}_{+-}(\omega+i\delta) \\ 0 & \delta M^{\mu *}_{+-}(\omega+i\delta) & \delta M^{\mu}_{++}(-\omega+i\delta) \end{bmatrix} . \tag{4.37}$$

In order to determine F^{μ} and M^{μ}_0, we use the equal-time expectation values $\langle \delta N_{l_\mu}\delta N^{\dagger}_{k_\mu}\rangle$ and $\epsilon_l\langle J_{l_\mu}\int \delta N^{\dagger}_{k_\mu}\rangle$. In fact, from the spectral representation similar to Eq. (4.13), we have

$$\langle \delta N_{l_\mu}\delta N^{\dagger}_{k_\mu}\rangle=\int d\omega\left[-\frac{1}{\pi}\right]\mathrm{Im}\left[\frac{1}{\omega-\Sigma^{\mu}(\omega)}F^{\mu}\right]_{lk} \times\omega(1+f_B(\omega)) \ . \tag{4.38a}$$

and

$$\epsilon_l\langle J_{l_\mu}\delta N^{\dagger}_{k_\mu}\rangle=\int d\omega\left[-\frac{1}{\pi}\right]\mathrm{Im}\left[\Sigma^{\mu}(\omega)\frac{1}{\omega-\Sigma^{\mu}(\omega)}F^{\mu}\right]_{lk} \times\omega(1+f_B(\omega)) \ . \tag{4.38b}$$

From Eq. (4.38) and the relation

$$\Sigma^{\mu}(\omega)=(M^{\mu}_0+\delta M^{\mu}(\omega))F^{\mu-1} \ , \tag{4.39}$$

F^{μ}, α_{μ}, and M^{μ}_0 are self-consistently obtained.

Let us define

$$X^{\mu}_{lk}=\langle \delta N_{l_\mu}\delta N^{\dagger}_{k_\mu}\rangle \tag{4.40a}$$

and

$$Y^{\mu}_{lk}=\epsilon_l\langle J_l\delta N^{\dagger}_{k_\mu}\rangle \ . \tag{4.40b}$$

From the definition of δN_{l_μ} in Eq. (4.17), we have

$$X^{\mu}_{lk}=x^{\mu}_{lk}-\alpha^{\mu}_l x^{\mu}_{0k}-x^{\mu}_{l0}\alpha^{\mu}_k+\alpha^{\mu}_l x^{\mu}_{00}\alpha^{\mu}_k \tag{4.41a}$$

and

$$Y^{\mu}_{lk}=y^{\mu}_{lk}-\alpha^{\mu}_l y^{\mu}_{0k}-y^{\mu}_{l0}\alpha^{\mu}_k+\alpha^{\mu}_l y^{\mu}_{00}\alpha^{\mu}_k \tag{4.41b}$$

with

$$x^{\mu}_{lk}=\langle \delta n_{l_\mu}\delta n^{\dagger}_{k_\mu}\rangle \tag{4.42a}$$

and

$$y^{\mu}_{lk}=\epsilon_l\langle j_{l_\mu}\delta n^{\dagger}_{k_\mu}\rangle \ . \tag{4.42b}$$

The explicit form of x^{μ}_{lk} is given by

$$x^0_{lk}=\begin{bmatrix} (n-1)(2-n) & -2b(2-n) & -2b(1-n) \\ -2b(2-n) & 2\left\{\frac{2-n}{n}(1-a)+\frac{1}{n}(a_0+a_s)-2b^2\right\} & -4b^2 \\ -2b(1-n) & -4b^2 & 4\left\{\frac{n-1}{n}a+\frac{1}{n}a_0-b^2\right\} \end{bmatrix} \tag{4.43}$$

and

$$x^i_{lk}=\begin{bmatrix} 2-n & 0 & 2b \\ 0 & 2\left\{\frac{2-n}{n}(1-a)+\frac{1}{n}(a_0-a_s)\right\} & 0 \\ 2b & 0 & 4\left\{\frac{n-1}{n}a+\frac{1}{n}a_0\right\} \end{bmatrix} , \tag{4.44}$$

where $a=\langle p_\gamma p_\gamma^\dagger\rangle$, $b=\langle p_\gamma r^\dagger\rangle$, $a_0=\langle p_\gamma p_0^\dagger\rangle$, and $a_s=\langle p_\gamma p_s^\dagger\rangle$. Similarly, y_{lk}^μ is given by

$$y_{lk}^\mu=\begin{bmatrix} t_n(x_{-k}^\mu-x_{+k}^\mu) \\ (\varepsilon_\eta-\varepsilon_p)x_{+k}^\mu+c_\mu x_{0k}^\mu \\ -(\varepsilon_\eta-\varepsilon_p)x_{-k}^\mu-c_\mu x_{0k}^\mu \end{bmatrix}+\delta y_{lk}^\mu \ , \tag{4.45}$$

with

$$\delta y_{++}^\mu=\frac{t_n}{n}\{4(1-a)b-2(2-n)b' -\delta_{\mu 0}(b_0'+b_s')-\delta_{\mu i}(b_0'-\tfrac{1}{3}b_s')\} \ , \tag{4.46a}$$

$$\delta y_{+-}^\mu=t_n\left\{-2ab+2ab\left[-\frac{4-n}{n}\delta_{\mu 0}+\delta_{\mu i}\right]-2ba_r'\right\} \tag{4.46b}$$

and

$$\delta y_{--}^\mu=-\frac{t_n}{n}\{8ab+4(n-1)b'-2b_0'\} \ , \tag{4.46c}$$

where $a=\langle p_\gamma p_\gamma^\dagger\rangle$, $a_r'=-\langle r_{\gamma_1^2} r^\dagger\rangle$, $b=\langle p_\gamma r^\dagger\rangle$, $b'=-\langle p_\gamma r_{\gamma_1^2}^\dagger\rangle$, $b_0'=\langle p_0 r_{\gamma_1^2}^\dagger\rangle$, and $b_s'=\langle p_s r_{\gamma_1^2}^\dagger\rangle$. The spectral function of fluctuations $\rho_{lk}^\mu(\omega)$ defined by

$$\int d\omega\rho_{lk}^\mu(\omega)e^{-i\omega(t-t')}=\langle[\delta n_{l_\mu}(t),\delta n_{k_\mu}^\dagger(t')]\rangle \tag{4.47}$$

is obtained as

$$\rho_{lk}^\mu(\omega)=R_{lk}^\mu(\omega)+\alpha_l^\mu R_{0k}^\mu(\omega)+R_{l0}^\mu(\omega)\alpha_k^\mu+\alpha_l^\mu R_{00}^\mu(\omega)\alpha_k^\mu \tag{4.48}$$

with $R_{lk}^\mu(\omega)$ being given by

$$R_{lk}^\mu(\omega)=-\frac{\omega}{\pi}\mathrm{Im}\left[F^\mu\frac{1}{\omega F^\mu-M_0^\mu-\delta M^\mu(\omega)}F^\mu\right] . \tag{4.49}$$

The explicit form of the dynamical correction $\delta M_{lk}^\mu(\omega)$ is obtained from Eq. (4.35). Since δJ_{l_μ} should not contain reducible $\delta N_{l_\mu'}$ lines, the correction $\delta M_{lk}^\mu(\omega)$ arises from loop corrections formed by $\delta j_{\pm\mu}$ given in Eq. (4.6). We approximate this contribution by fermion loops. By use of the spectral relation (4.13), we can evaluate the contributions of such fermion loops to the relaxation function from the retarded function,

$$\mathrm{Im}\delta M_{lk}^\mu(\omega)\approx\frac{1}{\omega}\mathrm{Im}(\epsilon_l:\delta j_{l_\mu}:\,:\delta j_{k_\mu}^\dagger:\epsilon_k) \ , \tag{4.50}$$

where $:\delta j_{+\mu}(x):$ indicates the contribution from fermion loops. The source $:\delta j_{+\mu}(x):$ is treated as

$$:\delta j_{+\mu}(x):=t_n:p_\gamma^\dagger(x)\sigma_\mu p_\gamma(x) -\frac{1}{n}p_\gamma^\dagger(x)\sigma_\mu(p_s(x)-p_0(x)) -r_{\gamma_1^2}^\dagger(x)\sigma_\mu r(x): \tag{4.51}$$

and fermion loops are formed from all possible combinations composed of $p_\gamma(x)$, $p_s(x)$, $p_0(x)$, and $r(x)$ with subtraction of double counting.

V. CONCLUDING REMARKS

In this paper, we present a formulation to treat highly correlated electron systems by introducing the notion of composite electronic excitations. The method to define the self-energies for such composite excitation is developed. Equations of motion and the retarded formulation is fully used. The mean-field approximation gives an approximate spectral intensity distribution. Several peak structures may be obtained as the result of the energy shift and mixing among p and d levels and composite electronic excitation levels. In the first approximation there are p, p_0, and p_s electronic excitations at ε_p and the upper Hubbard level r at ε_η. The shifts of their energies and mixing strengths are given by mean fields which contain effects from different lattice points. There are mean fields given by a $(=\langle p_\gamma p_\gamma^\dagger\rangle)$, b $(=\langle p_\gamma r^\dagger\rangle)$, a_0 $(=\langle p_\gamma p_0^\dagger\rangle)$, and a_s $(=\langle p_\gamma p_s^\dagger\rangle)$. One may say that "$2a$" relates to the p-hole density in a CuO_2 cluster and "b" does to the mixing between p and r. Since $2a_1$ $[=2\langle p_\gamma p_\gamma^\dagger(2-n)\rangle]$ indicates the p-hole density with the $Cu^{2+}(n=1)$ state and $2a_2$ $[=2\langle p_\gamma p_\gamma^\dagger(n-1)\rangle]$ the p-hole density with the Cu^+ $(n=2)$ state, a_0 relates to the ratio of them. When a p_γ electron and Cu spin in a CuO_2 cluster form a singlet or triplet state, one has

$$p_\gamma^\dagger\sigma p_\gamma\mathbf{n}=\begin{cases}-3 & \text{(for a singlet)},\\ 1 & \text{(for a triplet)} .\end{cases} \tag{5.1}$$

Therefore the expectation value $p_\gamma^\dagger\sigma p_\gamma\mathbf{n}$ may be given as

$$\langle p_\gamma^\dagger\sigma p_\gamma\mathbf{n}\rangle=-3C_s+C_t \ , \tag{5.2}$$

where C_s and C_t are the p-hole densities coupled with a Cu spin in singlet and triplet states, respectively, in a CuO_2 cluster. Note also that

$$C_s+C_t=\langle p_\gamma^\dagger\cdot p_\gamma(2-p_\gamma^\dagger\cdot p_\gamma)(2-n)\rangle \ . \tag{5.3}$$

From Eqs. (5.2) and (5.3), C_s and C_t are evaluated. The mean field a_s relates to the difference between p-hole densities occupying singlet and triplet states. Roughly speaking, a quasiparticle state is constructed from a linear combination of p and r which is the covalent electronic state. By adding composite operators p_0 and p_s, the correct quasielectron excitation ψ is expressed as

$$\psi=c_p p+c_r r+c_0 p_0+c_s p_s+\cdots \ . \tag{5.4}$$

The components p_0 and p_s describe the situation where the p electronic excitation is strongly influenced by states of neighboring Cu ions. One may regard this procedure as the configuration mixing. The mixing with such states is induced due to the nature of the Hubbard split d level. Hopping of p_0 and p_s electronic excitations to neighboring lattices is strongly controlled by the states of the Cu ions. This can be seen in the mean-field level in the fact that the intersite contribution is mostly controlled by intersite fluctuations.

Dynamical corrections have the spectral weights around the energies ε_η and $2\varepsilon_p-\varepsilon_\eta$. These contributions push down or push up the energy levels of p_0 and p_s excitations. In addition, there occurs redistribution of the electron density of states through decay processes to p, r, p_0, and p_s electronic excitations and fluctuations. Through decay processes the spectral intensity may be transferred to a lower energy region, and the peak structures of the mean-field result may be broadened to have a tendency of accumulating their intensity to the Fermi level (FL). In the subsequent paper,[34] we will see in fact that such phenomena are observed and especially that the accumulation of spectral intensity at the FL is induced with hole doping. We will also see that low-energy excitations of spin and charge fluctuations play important roles for such transfer of spectral intensities.

Finally, we comment shortly on our approximation scheme in comparison with the Hubbard approximation.[33] Expression (2.20) or (2.37) is most general and is independent of the approximation. The approximation comes in the evaluation of $I(\mathbf{k})$, $m_0(\mathbf{k})$, and $\delta m(\omega,\mathbf{k})$. In the Hubbard approximation, $\Sigma(\omega,\mathbf{k})$ is directly evaluated by use of the equation of motion (2.17) combined with the point splitting method and certain decomposition rule such as the random-phase approximation. Since $I(\mathbf{k})$ is not the identity matrix, it is hard to see whether or not the obtained $\Sigma(\omega,\mathbf{k})$ satisfies the necessary requirements for the total propagators such as Hermiticity and positivity. In our scheme, this point of calculation is systematized by considering the mean field $m_0(\mathbf{k})$ and the dynamical correction $\delta m(\omega,\mathbf{k})$. We have evaluated $\delta m(\omega,\mathbf{k})$ from the one-loop contributions of fermion and bosonic fluctuations, following the loop expansion in terms of renormalized excitations. Although δm contains only one loop, the full propagators contain infinite order of repetition of many loops through the mixing scheme induced by $I(\mathbf{k})$ and $m_0(\mathbf{k})$ and the self-consistency among mean fields.

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APPENDIX: EXACTLY SOLVABLE MODEL

In order to demonstrate the validity of the formulation presented in Sec. II, a simple example is presented in this appendix. Let us consider the single-site Kondo model where a d-electron freedom is restricted to its spin freedom and the d spin couples with a p-electron spin antiferromagnetically.

The Hamiltonian is given by

$$H=\varepsilon_p p^\dagger p+Jp^\dagger\sigma p\mathbf{S}\ , \tag{A1}$$

where σ is the Pauli matrix. We take an insulating vacuum where there is no p hole and consider one p-electron excitation spectrum. The model (A1) is exactly solvable and gives the result for the p-electron Green function,

$$S_{pp^\dagger}(\omega)=\frac{1}{4}\frac{1}{\omega-(\varepsilon_p+\frac{3}{2}J)}+\frac{3}{4}\frac{1}{\omega-(\varepsilon_p-\frac{1}{2}J)}\ , \tag{A2}$$

where

$$S_{pp^\dagger}(t_x-t_y)\equiv\langle Rp(t_x)p^\dagger(t_y)\rangle =\frac{i}{2\pi}\int d\omega e^{-i\omega(t_x-t_y)}S_{pp^\dagger}(\omega)\ . \tag{A3}$$

The result (A2) is obtained by identifying the eigenvectors of the Hamiltonian H and by taking the zero-temperature limit from the thermal average. The p electronic excitation is strongly renormalized and splits into two excitation levels around $\omega=\varepsilon_p$. The first and second terms correspond to excitations to the spin singlet and triplet final states of the p hole and the Cu spin, respectively.

Now we apply to this model the method developed in Sec. II. The equation of motion for the p electron is

$$i\frac{\partial}{\partial t}p(t)=\varepsilon_p p(t)+J\sigma p(t)\mathbf{S}(t)\ . \tag{A4}$$

Let us introduce a composite operator

$$p_s(t)=\equiv\sigma p(t)\mathbf{S}(t)\ , \tag{A5}$$

and identify a generalized electronic field $\psi(t)$ as

$$\psi(t)=\begin{bmatrix}\psi_1(t)\\ \psi_2(t)\end{bmatrix}=\begin{bmatrix}p(t)\\ p_s(t)\end{bmatrix}\ . \tag{A6}$$

The equation of motion for ψ is expressed as

$$i\frac{\partial}{\partial t}\psi(t)=j(t)\ , \tag{A7}$$

where

$$j(t)=\begin{bmatrix}j_1(t)\\ j_2(t)\end{bmatrix}\ , \tag{A8}$$

$$j_1(t)=\varepsilon_p p(t)+Jp_s(t)\ , \tag{A9}$$

and

$$j_2(t)=3cJp(t)+(\varepsilon_p+J)p_s(t) +J\sigma_i p(t)i\varepsilon_{ijk}p^\dagger(t)\sigma_j p(t)S_k(t) \tag{A10}$$

with $c=S_i^2=\frac{1}{4}$. Then one gets the equation for the Green function $S_{\psi\psi^\dagger}(t_x-t_y)$ [$\equiv\langle R\psi(t_x)\psi^\dagger(t_y)\rangle$],

$$i\frac{\partial}{\partial t_x}S_{\psi\psi^\dagger}(t_x-t_y)=i\delta(t_x-t_y)I+\langle Rj(t_x)\psi^\dagger(t_y)\rangle\ , \tag{A11}$$

where

$$I=\langle\{\psi,\psi^\dagger\}\rangle=\begin{bmatrix}1 & 0\\ 0 & 3(c+\frac{2}{3}a_s)\end{bmatrix} \tag{A12a}$$

with

$$a_s=\langle pp_s\rangle\ . \tag{A12b}$$

The generalized self-energy $\Sigma(\omega)$ is defined by Eq. (2.25),

$$\Sigma(i\partial_{t_x})i\delta(t_x-t_y)I=i\delta(t_x-t_y)m_0+\delta m(t_x-t_y)\ , \tag{A13}$$

where

$$m_0=\langle\{j,\psi^\dagger\}\rangle \tag{A14a}$$

and

$$\delta m(t_x-t_y)=\langle R\delta j(t_x)j^\dagger(t_y)\rangle - \langle R\delta j(t_x)\psi^\dagger(t_y)\rangle\Sigma^\dagger(-i\partial_{t_y})\ . \tag{A14b}$$

Note that $\delta j_1(t)=0$ and

$$\delta j_2(t)=J\sigma_i p(t)i\varepsilon_{ijk}p^\dagger(t)\sigma_j p(t)S_k(t)\ ,$$

and therefore

$$\delta m(t_x-t_y)=\begin{bmatrix}0 & 0\\ 0 & \delta m_{22}(t_x-t_y)\end{bmatrix}\ . \tag{A15}$$

After straightforward calculations, we get the expression for m_0,

$$(m_0)_{11}=\varepsilon_p\ , \tag{A16a}$$

$$(m_0)_{12}=3J(c+\tfrac{2}{3}a_s)\ , \tag{A16b}$$

$$(m_0)_{22}=(\varepsilon_p+J)3(c+\tfrac{2}{3}a_s)+3J(-2ca+4\chi)\ , \tag{A16c}$$

where

$$a=\langle pp^\dagger\rangle \tag{A17a}$$

and

$$\chi=\langle S_1\tfrac{1}{2}p^\dagger\sigma_2 p\tfrac{1}{2}p^\dagger\sigma_2\sigma_1 p\rangle\ . \tag{A17b}$$

The component $(m_0)_{21}$ should be equal to $(m_0)_{12}$ according to the Hermiticity requirement. In the case of the ground state with no p hole, $a=0$, $a_s=\chi=0$. Then

$$I=\begin{bmatrix}1 & 0\\ 0 & 3c\end{bmatrix} \tag{A18a}$$

and

$$m_0=\begin{bmatrix}\varepsilon_p & 3cJ\\ 3cJ & 3c(\varepsilon_p+J)\end{bmatrix}\ . \tag{A18b}$$

By substituting Eqs. (A17) into Eq. (2.34), one finds

$$S_{\psi\psi^\dagger}(\omega)=\frac{1}{(\omega-\varepsilon_p)(\omega-\varepsilon_p-J)-3cJ^2}\times\begin{bmatrix}\omega-\varepsilon_p-J & 3cJ\\ 3cJ & 3c(\omega-\varepsilon_p)\end{bmatrix}\ , \tag{A19}$$

which reproduces the exact result of Eq. (A2).

In this approach, we can reinterpret the splitting of the p-electron excitations into the singlet and triplet states as the consequence of the mixing between p and p_s. In fact, Eq. (A2) can be reexpressed as

$$S_{pp^\dagger}(\omega)=\frac{1}{\omega-\varepsilon_p-\Sigma_p(\omega)}\ , \tag{A20}$$

where

$$\Sigma_p(\omega)=\frac{3cJ^2}{\omega-(\varepsilon_p+J)}\ . \tag{A21}$$

The function $\Sigma_p(\omega)$ is the p-electron self-energy, and it is also a one-p-electron irreducible component of $S_{p_sp_s^\dagger}(\omega)$. The energy (ε_p+J) is the shifted excitation energy of p_s.

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