Boson representation of two-exciton correlations: An exact treatment of composite-particle effects

Satoru Okumura
*Department of Physics, Osaka University, Toyonaka Osaka 560-0043, Japan
and Department of Physics, Tohoku University, Aoba-ku, Sendai 980-8578, Japan*

Tetsuo Ogawa
*Department of Physics, Osaka University, Toyonaka, Osaka 560-0043, Japan
Department of Physics, Tohoku University, Aoba-ku, Sendai 980-8578, Japan
and "Structure and Transformation," PRESTO, JST, Kawaguchi 332-0012, Japan*

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We derive a bosonized Hamiltonian describing two-exciton correlation of semiconductor-photon coupled systems with a bosonization method, which takes into full account an effect of deviation of the excitons from ideal bosons. This deviation effect stems from the fact that excitons are composite particles, whose character appears clearly in the case where the excitons overlap each other. We call this effect a composite-particle effect (CPE). To our knowledges this effect was not considered completely in previous theoretical works on exciton-exciton interaction. The Hamiltonian introduced in this paper includes the results of the previous works as low-order terms of the CPE. After the introduction of a general theory of the bosonization method for arbitrary dimension and electron-hole mass ratio, we also demonstrate an application to a semiconductor bulk system coupled with a photon field in the heavy-hole limit. The bosonized Hamiltonian shows that the CPE brings about an enhancement of the exciton-exciton scattering strength and a qualitative change of the photo transition amplitude. It is also shown that the Hamiltonian describes two-exciton bound and scattering states.

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I. INTRODUCTION

Since semiconductors show various phenomena depending on the density of the carriers excited by light, they have attracted much notice for many years. In the low-density regime, third-order nonlinear optical responses, particularly four-wave-mixing signals near 1*s* exciton resonance, have been intensively investigated because they yield information not only on the transverse-relaxation time of excitons but also on two-exciton correlations, which are the most fundamental correlations in many-exciton correlations.[1–8] In the high-density regime, conversely, the Bose-Einstein condensation of the excitons, the electron-hole BCS states, the exciton Mott transition, the electron-hole droplet formation, etc., have been studied as interesting phenomena.[9–13]

For an analysis of these phenomena, theoretical methods are roughly classified into two kinds of frameworks. One is "fermionic methods," which directly treat the semiconductors as interacting electron-hole systems, and the other is "bosonic methods," which describe the systems by interacting ideal bosons corresponding to excitons (and the exciton molecules in some cases). Fermionic methods are of great generality because the electron-hole Hamiltonian describes the Coulomb interaction among the fermionic carriers and the carrier-light coupling. These methods have been developed in wide range of carrier densities by many workers.[12–17] Bosonic methods, on the other hand, comprise an effective theory for systems in which the excitons are well defined. Since these methods represent excitons as bosons, they give simple pictures to systems where the bosonic properties are useful in understanding the phenomena. For this reason, bosonic methods have often been employed phenomenologically for both the low-density systems[18,19] and the high-density systems.[10,20] Therefore, it is important to strengthen the microscopic bases of the bosonic methods, i.e., how to obtain the bosonic Hamiltonian by a bosonization of the excitons. Since two-exciton correlations are the most important many-exciton correlations not only for the low-density regime but also for the high-density regime, the bosonic Hamiltonian is required to describe two-exciton correlations. For this purpose, the Usui transformation[21–23] and the point-boson approximation[24,25] have been employed to estimate two-exciton correlation terms, i.e., the interaction between two bosonized excitons and the nonlinear coupling between two bosonized excitons and light.[22] Moreover, the exciton-exciton interaction described with the same formalism as the above bosonization methods have also been obtained, though these interactions are not evaluated as the result of the bosonization of the excitons.[26,27]

Although excitons have often been considered as bosons, strictly speaking they are *not* bosons satisfying typical boson commutation relations. This is because an exciton is a composite particle of an electron and a hole. In general, only when excitons are spatially separated enough from one another in comparison to the exciton Bohr radius *a* can they be regarded as bosons, whose internal structures are negligible. Conversely, when excitons come close to and overlap one another, composite characters of the excitons appear, and hence the excitons deviate from the bosons. Since the exciton correlations are determined by this deviation effect and the Coulomb interaction among the carriers, the deviation effect is indispensable for a correct estimation of the exciton correlations. We call the deviation effect a composite-particle effect (CPE). Bosonization methods should include the CPE in the boson-boson interaction and the boson-light nonlinear coupling. However, this effect has been less frequently considered thus far. In later sections, we will show that the boson-boson interaction and the boson-light nonlinear coupling derived in previous works[21–24,26,27] do not include the CPE completely. We classify those works as ''incomplete CPE theories.''

In this paper, we report a bosonized Hamiltonian describing correct two-exciton correlations in which the CPE is completely taken into account. To clarify the difference between the previous incomplete CPE theories and our ''complete CPE theory,'' we define the ''order'' of the CPE, and then derive a bosonized Hamiltonian including the full order of the CPE completely with the use of a bosonization technique.[28] This Hamiltonian includes the results of the incomplete CPE theory as low-order terms of the CPE. To keep generality, we assume that the material under consideration has two bands with arbitrary effective mass, spin degrees of freedom, and spatial dimension. Moreover, we generalize the quantum statistics of the particles in these bands. Although we implicitly suppose these particles as conduction electrons and valence holes in semiconductors, such an assignment is not necessarily required in our theory. We need to assume only that they are interacting particles with the same statistics and make energy eigenstates corresponding to bound states of the two-particle composite, e.g., a Wannier exciton. Its relative motion is assumed to be extended in the region of a characteristic length a (e.g., an exciton Bohr radius).[29]

In this paper, we confine ourselves to a process in which up to two two-particle composites are created by an external field. This corresponds to third-order nonlinear optical processes in semiconductors.[30] Here we should note that this restriction does *not* mean that our theory is not applicable to many-exciton systems, because the Hamiltonian describes complete two-exciton correlation terms in the ideal-boson space, e.g., $\hat{B}^\dagger\hat{B}^\dagger\hat{B}\hat{B}$, where $\hat{B}$ is a boson annihilation operator, irrespective of the constraint for the boson space. Even when we extend the system to a many-exciton system, the two-exciton correlations are unchanged and we obtain simply the correction of more than two-exciton correlations, e.g., $\hat{B}^\dagger\hat{B}^\dagger\hat{B}^\dagger\hat{B}\hat{B}\hat{B}$. The reason for this is that our bosonization technique gives a *normal-ordering* expansion of the boson operators. Therefore, the obtained Hamiltonian is also useful for many-exciton systems where more than two-exciton correlations are negligible. Only if we consider interactions between excitonic molecules, up to four-exciton correlation terms become necessary at least. This is a future problem, and we do not consider it in this paper.

This paper is organized as follows. In Sec. II, we introduce the starting Hamiltonian and the definition of the exciton, and then discuss the properties of the excitons including the CPE. The bosonization method we employ in this paper is formulated with the Tamm-Dancoff (TD) representation,[28] which represents a model space with exciton operators. Although, in general, bosonization methods need some restrictions to the ideal-boson space to obtain a one-to-one correspondence between the starting fermion space and the ideal-boson space, we need no restriction to the wave numbers of the component particles; this was a nuisance in the Usui transformation, but can put constraints on the energy of the relative motion of an exciton by using a TD representation. Moreover, we explicitly take into full account the exciton center-of-mass motion and the spin degrees of freedom of the component particles. In Sec. III, our bosonization method is elucidated, and an orthonormalized two-exciton state is introduced. Section IV is devoted to a calculation of matrix elements of the model Hamiltonian. In Sec. IV A, we examine the ''loose matrix elements,'' which stand for the matrix elements represented with the two-exciton states which are nonorthonormal basis. These loose matrix elements have often been interpreted as interactions among excitons.[21–24,26,27] Since the CPE is, however, not completely included in these elements, they lead to incorrect interactions among excitons and photons in the region that two excitons overlap each other. In Sec. IV B, we show *exact* matrix elements, which include the CPE completely. We also discuss a relation between the CPE and the phase-space filling. A general form of the bosonized Hamiltonian is given in Sec. IV C. The Hamiltonian contains four nontrivial interaction terms: two kinds of scattering among the bosonized excitons, and two kinds of nonlinear coupling between the bosonized excitons and the external photon field. In Sec. V we apply the general result to a semiconductor bulk system. In the limit of heavy holes, we can obtain exact, analytical forms of, e.g., boson-boson and boson-photon interactions. We find that the CPE enhances the scattering strength between the bosonized excitons, and the CPE results in a qualitative change of the nonlinear coupling between bosonized excitons and photons. We also show that our bosonization theory can explain both the bound and unbound states of two bosonized excitons.

II. STARTING HAMILTONIAN AND NONBOSON CHARACTERS OF EXCITONS

We start with a Hamiltonian consisting of three parts:

$$\hat{H}=\hat{H}_{cd}+\hat{H}_{a}+\hat{H}_{cd\text{-}a}, \tag{2.1}$$

where

$$\begin{aligned}\hat{H}_{cd}=&\sum_{\mu\mathbf{k}}\left(E_0+\frac{\hbar^2\mathbf{k}^2}{2m_c}\right)\hat{c}^\dagger_{\mu\mathbf{k}}\hat{c}_{\mu\mathbf{k}}+\sum_{\nu\mathbf{k}}\frac{\hbar^2\mathbf{k}^2}{2m_d}\hat{d}^\dagger_{\nu\mathbf{k}}\hat{d}_{\nu\mathbf{k}}\\&-\sum_{\mu\nu}\sum_{\mathbf{qkk}'}V_{cd}(\mathbf{q})\hat{c}^\dagger_{\mu\mathbf{k}+\mathbf{q}}\hat{d}^\dagger_{\nu\mathbf{k}'-\mathbf{q}}\hat{d}_{\nu\mathbf{k}'}\hat{c}_{\mu\mathbf{k}}\\&+\frac{1}{2}\sum_{\mu\mu'}\sum_{\mathbf{qkk}'}V_{cc}(\mathbf{q})\hat{c}^\dagger_{\mu\mathbf{k}+\mathbf{q}}\hat{c}^\dagger_{\mu'\mathbf{k}'-\mathbf{q}}\hat{c}_{\mu'\mathbf{k}'}\hat{c}_{\mu\mathbf{k}}\\&+\frac{1}{2}\sum_{\nu\nu'}\sum_{\mathbf{qkk}'}V_{dd}(\mathbf{q})\hat{d}^\dagger_{\nu\mathbf{k}+\mathbf{q}}\hat{d}^\dagger_{\nu'\mathbf{k}'-\mathbf{q}}\hat{d}_{\nu'\mathbf{k}'}\hat{d}_{\nu\mathbf{k}},\end{aligned} \tag{2.2}$$

$$\hat{H}_a=\sum_{\sigma\mathbf{K}}\hbar\omega_{\mathbf{K}}\hat{a}^\dagger_{\sigma\mathbf{K}}\hat{a}_{\sigma\mathbf{K}}, \tag{2.3}$$

$$\hat{H}_{cd\text{-}a} = \tilde{g} \sum_{\sigma} \sum_{\mathbf{K}} \hat{p}^{\dagger}_{\sigma\mathbf{K}} \hat{a}_{\sigma\mathbf{K}} + (\mathrm{H.c.}). \tag{2.4}$$

The first term, $\hat{H}_{cd}$, describes two kinds of interacting particles, whose annihilation operators are $\hat{c}_{\mu\mathbf{k}}$ and $\hat{d}_{\nu\mathbf{k}}$. We call these particles a c particle and a d particle, respectively. To investigate effects of the quantum statistics of the components, we shall not assume their statistics (fermionic or bosonic) for the moment, but assume only that both particles have identical statistics. Although we implicitly suppose these particles as a conduction electron and a valence hole, such an assignment is not necessarily required in this paper. The subscripts μ and ν are the indices of internal degrees of freedom of each particle, e.g., spins, and $\mathbf{k}$ is a D-dimensional wave vector defined as $\mathbf{k} = 2\pi/L(n_1\mathbf{e}_1 + n_2\mathbf{e}_2 + \cdots + n_D\mathbf{e}_D)$, where L is the system size, $\mathbf{e}_i$ is a unit vector of the direction i, and n_i is an integer. At least an energy E_0 is needed for a c particle to be created. We also assume that the interactions between different kinds of particles is attractive, while that among the same kinds is repulsive. The latter condition means that two particles of the same kind are not bound to one another. Here the summation on $\mathbf{q}$ excludes $\mathbf{q}=0$ and m_c (m_d) is the effective mass of the c (d) particle. The second term $\hat{H}_a$ is a free Hamiltonian for an a particle, corresponding to a photon, whose operators are $\hat{a}_{\sigma\mathbf{K}}$ and $\hat{a}^{\dagger}_{\sigma\mathbf{K}}$. The subscript σ stands for an internal degree of freedom, e.g., the polarization. The third term $\hat{H}_{cd\text{-}a}$ expresses the interaction between the a particle and a pair of the c and d particles, where $\hat{p}^{\dagger}_{\sigma\mathbf{K}}$ is defined as

$$\hat{p}^{\dagger}_{\sigma\mathbf{K}} \equiv \sum_{\mu\nu} \delta^{\mu+\nu}_{\sigma} \sum_{\mathbf{k}} \hat{c}^{\dagger}_{\mu\alpha\mathbf{K}+\mathbf{k}} \hat{d}^{\dagger}_{\nu\beta\mathbf{K}-\mathbf{k}}. \tag{2.5}$$

Here $\delta^{\mu+\nu}_{\sigma}$ is the Kronecker delta describing a selection rule of the internal degrees of freedom of these particles, and α and β are m_c/M and m_d/M, respectively, with $M = m_c + m_d$. Thus the total Hamiltonian $\hat{H}$ can describe semiconductors with a radiation field in an arbitrary dimension.

Here we introduce a composite of c and d particles. The creation operator of the composite particle is defined as

$$\hat{b}^{\dagger}_{\mu\nu\mathbf{R}} = \int d^D\mathbf{r}\, \varphi(\mathbf{r}) \hat{c}^{\dagger}_{\mu\mathbf{R}+\beta\mathbf{r}} \hat{d}^{\dagger}_{\nu\mathbf{R}-\alpha\mathbf{r}}, \tag{2.6}$$

where $\hat{c}^{\dagger}_{\mu\mathbf{r}}$ and $\hat{d}^{\dagger}_{\nu\mathbf{r}}$ are the field operators of the c and d particles related to $\hat{c}^{\dagger}_{\mu\mathbf{k}}$ and $\hat{d}^{\dagger}_{\nu\mathbf{k}}$ via the Fourier transformation. We assume only that $\varphi(\mathbf{r})$ damps rapidly for $|\mathbf{r}| \gg a$ to describe a bound state, where a is a characteristic length of the spatial extent of $\varphi(\mathbf{r})$. This satisfies the eigenvalue equation

$$\left[-\frac{\hbar^2\nabla^2}{2m} - V_{cd}(\mathbf{r}) \right] \varphi(\mathbf{r}) = E\varphi(\mathbf{r}), \tag{2.7}$$

with $\int d^D\mathbf{r} |\varphi(\mathbf{r})|^2 = 1$, where $m = m_c m_d/M$, and $V_{cd}(\mathbf{r})$ is the Fourier transform of $V_{cd}(\mathbf{q})$. Thus the operator $\hat{b}^{\dagger}_{\mu\nu\mathbf{R}}$ expresses the creation of a composite particle at $\mathbf{R}$, whose binding energy is E. According to convention, let us call this composite particle an "exciton." The Fourier transform of the exciton creation operator $\hat{b}^{\dagger}_{\mu\nu\mathbf{R}}$, defined as

$$\begin{aligned} \hat{b}^{\dagger}_{\mu\nu\mathbf{K}} &= \frac{1}{L^{D/2}} \int d^D\mathbf{R}\, \hat{b}^{\dagger}_{\mu\nu\mathbf{R}} \exp(i\mathbf{K}\cdot\mathbf{R}) \\ &= \sum_{\mathbf{k}} \phi(\mathbf{k}) \hat{c}^{\dagger}_{\mu\alpha\mathbf{K}+\mathbf{k}} \hat{d}^{\dagger}_{\nu\beta\mathbf{K}-\mathbf{k}}, \end{aligned} \tag{2.8}$$

satisfies the eigenvalue-equation of $\hat{H}_{cd}$:

$$\hat{H}_{cd} \hat{b}^{\dagger}_{\mu\nu\mathbf{K}} |0\rangle = \left(E_0 + E + \frac{\hbar^2\mathbf{K}^2}{2M} \right) \hat{b}^{\dagger}_{\mu\nu\mathbf{K}} |0\rangle. \tag{2.9}$$

Here

$$\phi(\mathbf{k}) = \frac{1}{L^{D/2}} \int d^D\mathbf{r}\, \varphi(\mathbf{r}) \exp(-i\mathbf{k}\cdot\mathbf{r}), \tag{2.10}$$

and $|0\rangle$ means the vacuum state of $\hat{H}_{cd}$. Thus the one-exciton state $\hat{b}^{\dagger}_{\mu\nu\mathbf{K}}|0\rangle$ is an eigenstate of $\hat{H}_{cd}$.

To investigate effects coming from the composite-particle nature of the excitons, we calculate the commutation relations of the exciton operators, i.e.,

$$[\hat{b}_{\mu\nu\mathbf{R}}, \hat{b}_{\mu'\nu'\mathbf{R}'}] = 0, \tag{2.11}$$

$$\begin{aligned} [\hat{b}_{\mu\nu\mathbf{R}}, \hat{b}^{\dagger}_{\mu'\nu'\mathbf{R}'}] = {} & \delta^{\mu}_{\mu'} \delta^{\nu}_{\nu'} \delta^D(\mathbf{R}-\mathbf{R}') \\ & + s\,\delta^{\nu}_{\nu'} \int \frac{d^D\mathbf{r}}{\alpha^{2D}} \varphi^*\left(\frac{\mathbf{r}+\mathbf{R}}{\alpha}\right) \varphi\left(\frac{\mathbf{r}+\mathbf{R}'}{\alpha}\right) \\ & \times \hat{c}^{\dagger}_{\mu'(\mathbf{R}'+\beta\mathbf{r}')/\alpha} \hat{c}_{\mu(\mathbf{R}+\beta\mathbf{r})/\alpha} \\ & + s\,\delta^{\mu}_{\mu'} \int \frac{d^D\mathbf{r}}{\beta^{2D}} \varphi^*\left(\frac{\mathbf{r}-\mathbf{R}}{\beta}\right) \varphi\left(\frac{\mathbf{r}-\mathbf{R}'}{\beta}\right) \\ & \times \hat{d}^{\dagger}_{\nu'(\mathbf{R}'-\alpha\mathbf{r}')/\beta} \hat{d}_{\nu(\mathbf{R}-\alpha\mathbf{r})/\beta}, \end{aligned} \tag{2.12}$$

where $s = +1$ when both the c and d particles are bosons and $s = -1$ when both are fermions. Equation (2.11) is a simple commutation relation of the bosonlike statistics, because an exciton consists of components of an even number, i.e., two in this case. On the other hand, Eq. (2.12) shows that the exciton cannot be regarded as one ideal boson due to the existence of the second and third terms, which are direct consequences of the composite-particle nature of the excitons. Since the inner structures of the exciton appear in the second and third terms, these terms are influenced by the statistics of the c and d particles. When a distance between two excitons, whose center-of-mass positions are $\mathbf{R}$ and $\mathbf{R}'$, is sufficiently larger than a, i.e., $|\mathbf{R}-\mathbf{R}'| \gg a$, Eq. (2.12) becomes

$$[\hat{b}_{\mu\nu\mathbf{R}}, \hat{b}^{\dagger}_{\mu'\nu'\mathbf{R}'}] \simeq \delta^{\mu}_{\mu'} \delta^{\nu}_{\nu'} \delta^D(\mathbf{R}-\mathbf{R}'), \tag{2.13}$$

which is a consequence of the point-boson approximation. This means that the exciton can be regarded as a boson only if they are sufficiently separated from each other.

The effects arising from the composite-particle nature of the excitons appear in various physical values in the region that inner structures of the excitons can be recognized. These are the CPE's. The deviation from the ideal bosonic commutation relation in Eq. (2.12) is an example of the CPE. The CPE also appears in the inner product between the two-exciton states. We first show the inner product between the one-exciton states, $\hat{b}^\dagger_{\mu\nu\mathbf{K}}|0\rangle$. Abbreviating the set $\{\mu_i,\nu_i,\mathbf{K}_i\}$ to i, the inner product between the one-exciton states is $\langle 0|\hat{b}_m\hat{b}^\dagger_i|0\rangle=\delta^m_i$, where $\delta^m_i\equiv\delta^{\mu_m}_{\mu_i}\delta^{\nu_m}_{\nu_i}\delta^{\mathbf{K}_m}_{\mathbf{K}_i}$. Since the one-exciton states are eigenstates of $\hat{H}_{cd}$, this orthogonality is a natural result. On the other hand, the inner product between the two-exciton states $\hat{b}^\dagger_i\hat{b}^\dagger_j|0\rangle$ is evaluated as

$$\langle 0|\hat{b}_n\hat{b}_m\hat{b}^\dagger_i\hat{b}^\dagger_j|0\rangle=2\mathcal{F}^0(n,m|i,j)+2s\mathcal{F}^1(n,m|i,j), \tag{2.14}$$

where

$$\mathcal{F}^0(n,m|i,j)=\tfrac{1}{2}[A^0(n,m|i,j)f^0(n,m|i,j)+(i\leftrightarrow j\ \text{ or }\ m\leftrightarrow n)]=\tfrac{1}{2}(\delta^m_i\delta^n_j+\delta^m_j\delta^n_i), \tag{2.15}$$

$$\mathcal{F}^1(n,m|i,j)=\tfrac{1}{2}[A^1(n,m|i,j)f^1(n,m|i,j)+(i\leftrightarrow j\ \text{ or }\ m\leftrightarrow n)]. \tag{2.16}$$

Here we use the following definitions:

$$A^0(n,m|i,j)=\delta^{\mu_m}_{\mu_i}\delta^{\nu_m}_{\nu_i}\delta^{\mu_n}_{\mu_j}\delta^{\nu_n}_{\nu_j}, \tag{2.17}$$

$$A^1(n,m|i,j)=\delta^{\mu_m}_{\mu_j}\delta^{\nu_m}_{\nu_i}\delta^{\mu_n}_{\mu_i}\delta^{\nu_n}_{\nu_j}, \tag{2.18}$$

$$f^0(n,m|i,j)=\delta^{\mathbf{K}_m}_{\mathbf{K}_i}\delta^{\mathbf{K}_n}_{\mathbf{K}_j}, \tag{2.19}$$

$$\begin{aligned}f^1(n,m|i,j)=\delta^{\mathbf{K}_m+\mathbf{K}_n}_{\mathbf{K}_i+\mathbf{K}_j}\sum_{\mathbf{k}}\ &(\phi^*\alpha(\mathbf{K}_m-\mathbf{K}_j)+\mathbf{k})\\ &\times\phi^*(\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k})\\ &\times\phi(\mathbf{k})\phi(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k}).\end{aligned} \tag{2.20}$$

In the above formulas, $(i\leftrightarrow j$ or $m\leftrightarrow n)$ means a term obtained by exchanging the indices i and j or m and n. Note that $A^0(n,m|i,j)f^0(n,m|i,j)=A^0(m,n|j,i)f^0(m,n|j,i)$ and $A^1(n,m|i,j)f^1(n,m|i,j)=A^1(m,n|j,i)f^1(m,n|j,i)$ but $A^0(n,m|i,j)f^0(n,m|i,j)\neq A^0(n,m|j,i)f^0(n,m|j,i)\neq A^0(m,n|i,j)f^0(m,n|i,j)$, and so on. The Kronecker deltas of $A^1(n,m|i,j)$ in the second term have an exchanged form of μ_i and μ_j. This term expresses how much the two-exciton state contains another two-exciton state with exchanged components. We shall take the limit of $|\mathbf{R}_i-\mathbf{R}_j|\gg a$ after the Fourier transformation to change the variables from $\mathbf{K}_i$ to $\mathbf{R}_i$. When this procedure, called the point-boson limit, is applied to Eq. (2.14), $f^1(n,m|i,j)$ disappears, and then the inner product is reduced to

$$\langle 0|\hat{b}_n\hat{b}_m\hat{b}^\dagger_i\hat{b}^\dagger_j|0\rangle\simeq 2\mathcal{F}^0(n,m|i,j)=\delta^m_i\delta^n_j+\delta^m_j\delta^n_i, \tag{2.21}$$

which is a result of the point-boson approximation. Additionally, the second term of Eq. (2.14) depends on s, i.e., the statistics of the components of an exciton. Thus the term $2\mathcal{F}^1(n,m|i,j)$ results from the CPE. To describe the CPE systematically, the order of the CPE is introduced here; we regard the first term $\mathcal{F}^0(n,m|i,j)$ and the second term $\mathcal{F}^1(n,m|i,j)$ of Eq. (2.14) as CPE's of zeroth and of first orders, respectively. Similarly, the CPE of the nth order, $\mathcal{F}^n(n,m|i,j)$, is defined for any positive integer n, whose definition will be shown in AppendixA.

In this paper, we concentrate our discussion on processes where at most two excitons are created, which is sufficient for the third-order nonlinear optical processes in semiconductors. Therefore, we can consider only the subspace spanned by the ground state $|0\rangle$, the one-exciton states $\hat{b}^\dagger_{\mu\nu\mathbf{K}}|0\rangle$, and the two-exciton states $\hat{b}^\dagger_{\mu\nu\mathbf{K}}\hat{b}^\dagger_{\mu'\nu'\mathbf{K}'}|0\rangle$. As described above, however, the two-exciton states do not form an orthonormal set due to the CPE. Hence we need to introduce orthonormalized two-exciton states in Sec. III.

III. ORTHONORMALIZED TWO-EXCITON STATES AND BOSON MAPPING

We define an orthonormalized two-exciton state $|i,j\rangle$ as

$$|i,j\rangle\equiv\sum_{l=0}^{\infty}\sum_{n,m}\hat{b}^\dagger_n\hat{b}^\dagger_m|0\rangle(-s)^l\xi_l\mathcal{F}^l(m,n|i,j), \tag{3.1}$$

where $\xi_l=(2l)!/(2^l l!)^2$ and the coefficient $\mathcal{F}^l(n,m|i,j)$ is defined in Appendix A. This state is a superposition of the two-exciton states with the infinite order of the CPE, and has the following desirable properties:

$$|i,j\rangle=|j,i\rangle, \tag{3.2}$$

$$\langle n,m|=\sum_{l=0}^{\infty}\sum_{m',n'}(-s)^l\xi_l\mathcal{F}^l(n,m|m',n')\langle 0|\hat{b}_{n'}\hat{b}_{m'}, \tag{3.3}$$

$$\langle n,m|i,j\rangle=2\mathcal{F}^0(n,m|i,j)=\delta^m_i\delta^n_j+\delta^m_j\delta^n_i. \tag{3.4}$$

The orthonormalized two-exciton state has an exchange symmetry with repect to the indices, as shown in Eq. (3.2). Equation (3.3) can be proved with the use of Eq. (A8) in Appendix A. The third relation [Eq. (3.4)], means that these states form an orthonormal set. It is obvious that the orthonormalized two-exciton states are also orthogonal to the vacuum and to any one-exciton states. Thus the ground state $|0\rangle$, the one-exciton states $\hat{b}^\dagger_{\mu\nu\mathbf{K}}|0\rangle$, and the orthonormalized two-exciton states $|i,j\rangle$ are the orthonormal bases spanning the subspace under consideration.

The bosonization procedure[28] is introduced here as a map from the fermion subspace to an ideal boson space as follows.

$$|0\rangle\leftrightarrow|0), \tag{3.5}$$

$$\hat{b}_i^\dagger|0\rangle \leftrightarrow \hat{B}_i^\dagger|0), \tag{3.6}$$

$$|i,j\rangle \leftrightarrow \hat{B}_i^\dagger \hat{B}_j^\dagger |0). \tag{3.7}$$

Here $|0)$ is the vacuum state of the ideal boson space. The boson operators $\hat{B}_i$ and $\hat{B}_i^\dagger$ with an index $i=\{\mu_i,\nu_i,\mathbf{K}_i\}$ satisfy the boson commutation relations: $[\hat{B}_i,\hat{B}_j]=0$ and $[\hat{B}_i,\hat{B}_j^\dagger]=\delta_j^i$. The exchange symmetry for the bosons is guaranteed by Eq. (3.2). Hereafter, we call a boson described by $\hat{B}_{\mu\nu\mathbf{K}}$ a "bosonized exciton."

It is more useful to define a mapping operator $\hat{U}$ as

$$\hat{U}=|0\rangle(0|+\sum_i \hat{b}_i^\dagger|0\rangle(0|\hat{B}_i+\frac{1}{2!}\sum_{i,j}|i,j\rangle(0|\hat{B}_j\hat{B}_i. \tag{3.8}$$

With this operator, we can describe an arbitrary state $|\psi)$ and an operator $\hat{O}^B$ in the boson space in terms of the corresponding state $|\psi\rangle$ and the operator $\hat{O}$ in the fermion subspace as

$$|\psi)=\hat{U}^\dagger|\psi\rangle, \tag{3.9}$$

$$\hat{O}^B=\hat{U}^\dagger\hat{O}\hat{U}. \tag{3.10}$$

This method belongs to the Holstein-Primakoff (HP) type bosonization.[31] Hence the bosonized operator $\hat{O}^B$ is Hermitian if the fermion operator $\hat{O}$ is Hermitian. Moreover, it is obvious that the bosonized operator has a *normal-ordering* expansion form due to the mapping method [Eq. (3.10)], and an iteration method of the completeness relation $|0)(0|=1-\Sigma_i\hat{B}_i^\dagger\hat{B}_i+\cdots$. In general, the HP-type bosonization transforms Hermitian operators to Hermitian bosonized operators with infinite boson expansion terms. In the case under consideration, however, the bosonized operator in Eq. (3.10) has simply finite terms. The reason for this is as follows. Since we consider only optical processes in which up to two excitons are created from a vacuum, it is a necessary and sufficient condition for such processes that only the ground state, one-exciton states, and two-exciton states are taken into account. Moreover, a subspace spanned by two-exciton states is mapped to a subspace spanned by two-bosonized-exciton states. Therefore, two-bosonized-exciton states are also created from vacuum in the boson space for the processes under consideration. Thus we can safely neglect more than two-body scattering terms in the bosonized operator. An example of such processes is the third-order nonlinear optical process near $1s$-exciton resonance in semiconductors. The bosonized Hamiltonian for this case will be derived in Sec. V. However, we should note that irrespective of the constraint for the boson space, this restriction does *not* mean that our theory is not applicable to many-exciton systems. This is because the normal-ordering expansion form of the bosonization method enables the Hamiltonian to describe complete two-exciton correlation terms in ideal-boson space. Even when we extend the system to a many-exciton system, the two-exciton correlations are unchanged and we obtain simply the correction of more than two-exciton correlations, e.g., $\hat{B}^\dagger\hat{B}^\dagger\hat{B}^\dagger\hat{B}\hat{B}\hat{B}$.

In general, an ideal boson space including N bosons is wider than the corresponding fermion space because of $N!$ kinds of permutations of N pairs of fermions, that is, a one-to-$N!$ correspondence. Therefore, we need a restriction on the ideal boson space to create a one-to-one correspondence. This restricted space is called a physical subspace. Since the Usui transformation[32] realizes the physical subspace by a restriction to the momenta of components of excitons, scatterings among excitons are hard to describe exactly. The bosonization method used here, on the other hand, can lead to a physical subspace by a restriction to the energy of an exciton relative motion (no restriction to the center-of-mass motion of the excitons). Moreover, we will see that interaction terms in our bosonized Hamiltonian have *no* boson-number dependence, which is in striking contrast to the Marumori mapping.[33] These properties, therefore, enable us to describe the physical picture that bosons with a definite internal energy are created, annihilated, and scattered via interactions. In this paper, we adopt the only one relative motion with energy E.

IV. MATRIX ELEMENTS AND BOSONIZED HAMILTONIAN

In this section, we derive a bosonized Hamiltonian with our bosonization method described in Sec. III. With the use of the mapping operator $\hat{U}$ defined in Eq. (3.8), the starting Hamiltonian $\hat{H}$ is transformed to

$$\begin{aligned}\hat{H}^B=\hat{U}^\dagger\hat{H}\hat{U}=&\sum_i\left(E_0+E+\frac{\hbar^2\mathbf{K}_i^2}{2M}\right)\hat{B}_i^\dagger\hat{B}_i\\&+\left(\frac{1}{2!}\right)^2\sum_{n,m,i,j}[\langle n,m|\hat{H}_{cd}|i,j\rangle-4(E_0+E)\\&\times\mathcal{F}^0(n,m|i,j)-\mathcal{T}^{(0)}(n,m|i,j)]\hat{B}_m^\dagger\hat{B}_n^\dagger\hat{B}_j\hat{B}_i\\&+\tilde{g}\sqrt{L^D}\varphi^*(0)\sum_i\delta_{\sigma_i}^{\mu_i+\nu_i}\hat{B}_i^\dagger\hat{a}_i+(\text{H.c.})\\&+\frac{1}{2!}\sum_{n,m,i,j}[\tilde{g}\langle n,m|\hat{p}_i^\dagger\hat{b}_j^\dagger|0\rangle\\&-\mathcal{P}^{(0)}(n,m|i,j)]\hat{B}_m^\dagger\hat{B}_n^\dagger\hat{B}_j\hat{a}_i+(\text{H.c.}),\end{aligned} \tag{4.1}$$

where we use the definition

$$\mathcal{T}^{(0)}(n,m|i,j)\equiv\frac{\hbar^2}{2M}(\mathbf{K}_n^2+\mathbf{K}_m^2+\mathbf{K}_i^2+\mathbf{K}_j^2)\mathcal{F}^0(n,m|i,j), \tag{4.2}$$

and the matrix elements

$$\langle 0|\hat{H}_{cd}|0\rangle=0, \tag{4.3}$$

$$\langle 0|\hat{b}_m\hat{H}_{cd}\hat{b}_i^\dagger|0\rangle=\delta_i^m\left(E_0+E+\frac{\hbar^2\mathbf{K}_i^2}{2M}\right), \tag{4.4}$$

$$\tilde{g}\langle 0|\hat{b}_m\hat{p}_i^\dagger|0\rangle=\delta_{\sigma_i}^{\mu_m+\nu_m}\delta_{\mathbf{K}_i}^{\mathbf{K}_m}\tilde{g}L^{D/2}\varphi^*(0). \tag{4.5}$$

The coefficient $\mathcal{P}^{(0)}(n,m|i,j)$ in Eq. (4.1) is defined by Eq. (B11) in AppendixB. An iteration method of the completeness relation $|0)(0|=1-\Sigma_i\hat{B}_i^\dagger\hat{B}_i+\cdots$ is also used. In Eq. (4.1), an index i of the a-particle operator stands for $i=\{\sigma_i,\mathbf{K}_i\}$, and that of the bosonized exciton operators are $i=\{\mu_i,\nu_i,\mathbf{K}_i\}$. In the following, we shall calculate the matrix elements such as $\langle n,m|\hat{H}_{cd}|i,j\rangle$ and $\tilde{g}\langle n,m|\hat{p}_i^\dagger\hat{b}_j^\dagger|0\rangle$ to clarify contribution of the original Coulomb interactions and the CPE.

A. Loose matrix elements

Before evaluating the matrix elements $\langle n,m|\hat{H}_{cd}|i,j\rangle$ and $\tilde{g}\langle n,m|\hat{p}_i^\dagger\hat{b}_j^\dagger|0\rangle$, we first consider $\langle 0|\hat{b}_n\hat{b}_m\hat{H}_{cd}\hat{b}_i^\dagger\hat{b}_j^\dagger|0\rangle$ and $\tilde{g}\langle 0|\hat{b}_n\hat{b}_m\hat{p}_i^\dagger\hat{b}_j^\dagger|0\rangle$. We call these ''loose matrix elements'' in this paper because these are not elements of the matrix representation of operators, i.e., $\hat{b}_i^\dagger\hat{b}_j^\dagger|0\rangle$ is not orthonormalized. These loose matrix elements have often been interpreted as exciton-exciton scattering amplitudes or interactions.[21–24,26,27] However, when the loose matrix elements are used as interactions, qualitative problems appear in the region that two excitons overlap with each other. We will show examples of such problems in Sec. V.

We first evaluate the loose matrix element of $\hat{H}_{cd}$:

$$\begin{aligned}\langle 0|\hat{b}_n\hat{b}_m\hat{H}_{cd}\hat{b}_i^\dagger\hat{b}_j^\dagger|0\rangle &=4(E_0+E)[\mathcal{F}^0(n,m|i,j)+s\mathcal{F}^1(n,m|i,j)]\\&\quad+\mathcal{T}^{(0)}(n,m|i,j)+s\frac{\hbar^2}{2M}(\mathbf{K}_n^2+\mathbf{K}_m^2+\mathbf{K}_i^2+\mathbf{K}_j^2)\\&\quad\times\mathcal{F}^1(n,m|i,j)+\mathcal{U}^{(0)}(n,m|i,j)-s\mathcal{V}^{(1)}(n,m|i,j),\end{aligned} \tag{4.6}$$

where $\mathcal{U}^{(0)}(n,m|i,j)$ and $\mathcal{V}^{(1)}(n,m|i,j)$ are defined by Eqs. (B1) and (B6) in Appendix B. The superscripts of $\mathcal{U}^{(0)}(n,m|i,j)$ and $\mathcal{V}^{(1)}(n,m|i,j)$ represent the order of the CPE. In the above equation, the fourth term $\mathcal{U}^{(0)}(n,m|i,j)$ and the fifth term $-s\mathcal{V}^{(1)}(n,m|i,j)$ result from the interaction terms of $\hat{H}_{cd}$. The fourth term expresses the interaction energy among the density distributions of the c and d particles, while the fifth term is one of the CPE's expressing the interaction with exchange of the components.[27] If we take the point-boson limit, Eq. (4.6) is reduced to

$$\begin{aligned}\langle 0|\hat{b}_n\hat{b}_m\hat{H}_{cd}\hat{b}_i^\dagger\hat{b}_j^\dagger|0\rangle\simeq 4(E_0+E)\mathcal{F}^0(n,m|i,j)\\+\mathcal{T}^{(0)}(n,m|i,j)+\mathcal{U}^{(0)}(n,m|i,j),\end{aligned} \tag{4.7}$$

where the first-order CPE terms, $\mathcal{F}^1(n,m|i,j)$ and $\mathcal{V}^{(1)}(n,m|i,j)$, disappear.

Next the loose matrix element $\tilde{g}\langle 0|\hat{b}_n\hat{b}_m\hat{p}_i^\dagger\hat{b}_j^\dagger|0\rangle$ is evaluated as

$$\tilde{g}\langle 0|\hat{b}_n\hat{b}_m\hat{p}_i^\dagger\hat{b}_j^\dagger|0\rangle=\mathcal{P}^{(0)}(n,m|i,j)-s\mathcal{Q}^{(1)}(n,m|i,j). \tag{4.8}$$

Here $\mathcal{P}^{(0)}(n,m|i,j)$ is given in Eq. (B11) and $\mathcal{Q}^{(1)}(n,m|i,j)$ is defined by Eq. (B12) in Appendix B. An index i of $p_i^\dagger$ denotes $\{\sigma_i,\mathbf{K}_i\}$. In Eq. (4.8), $\mathcal{P}^{(0)}(n,m|i,j)$ and $-s\mathcal{Q}^{(1)}(n,m|i,j)$ result from zeroth-order and first-order CPE's, respectively. In the point-boson limit, $\mathcal{Q}^{(1)}(n,m|i,j)$ disappears, to lead to

$$\tilde{g}\langle 0|\hat{b}_n\hat{b}_m\hat{p}_i^\dagger\hat{b}_j^\dagger|0\rangle\simeq\mathcal{P}^{(0)}(n,m|i,j). \tag{4.9}$$

When components of an exciton are fermionic, i.e., $s=-1$, $-s\mathcal{Q}^{(1)}(n,m|i,j)$ has been interpreted as the PSF, which expresses the reduction of transition amplitudes between the one- and two-exciton states by Pauli blocking. However, we should recall that the two-exciton state $\hat{b}_i^\dagger\hat{b}_j^\dagger|0\rangle$ is *not* orthonormalized, as shown in Eq. (2.14). In fact, the norms of some two-exciton states become less than unity if the components are fermions. The reduction of the transition amplitude, therefore, also comes from the norm reduction as well as the Pauli blocking. Thus we should discuss PSF with exact transition amplitudes (or the exact matrix elements) from a one-exciton state to an *orthonormalized* two-exciton state. In this sense, our bosonization method leads to the correct transition amplitudes.

B. Matrix elements

With the use of the loose matrix elements in Sec. IV A, we can obtain matrix elements $\langle n,m|\hat{H}_{cd}|i,j\rangle$ and $\tilde{g}\langle n,m|\hat{p}_i^\dagger\hat{b}_j^\dagger|0\rangle$ in Eq. (4.1). These matrix elements give an exact matrix representation of the operator. In the following, we show analytical formulas of the matrix elements, and discuss the contribution of the CPE.

The matrix element of $\hat{H}_{cd}$ between the orthonormalized two-exciton states is obtained as

$$\begin{aligned}\langle n,m|\hat{H}_{cd}|i,j\rangle &=4(E_0+E)\mathcal{F}^0(n,m|i,j)\\&\quad+\sum_{l=0}^{\infty}(-s)^l\mathcal{T}^{(l)}(n,m|i,j)\\&\quad+\sum_{l=0}^{\infty}(-s)^l\mathcal{U}^{(l)}(n,m|i,j)\\&\quad+\sum_{l=1}^{\infty}(-s)^l\mathcal{V}^{(l)}(n,m|i,j).\end{aligned} \tag{4.10}$$

Here the second term results from the free part of the model Hamiltonian,[34] and $\mathcal{T}^{(l)}(n,m|i,j)$ expresses the lth-order CPE. Its definition is given by Eq. (C1) in Appendix C. According to the definition, $\mathcal{T}^{(1)}(n,m|i,j)$ vanishes. Therefore, there is no first-order term of the CPE in the second term of Eq. (4.10). In the above, $\mathcal{U}^{(l)}(n,m|i,j)$ in the third term and $\mathcal{V}^{(l)}(n,m|i,j)$ in the fourth term are extensions of $\mathcal{U}^{(0)}(n,m|i,j)$ and $\mathcal{V}^{(1)}(n,m|i,j)$ in Eq. (4.6), respectively. Their definitions are also given as Eqs. (C4) and(C5) in Appendix C. Note that $\mathcal{U}^{(l)}(n,m|i,j)$ is defined for $l\geqslant 0$, while

$\mathcal{V}^{(l)}(n,m|i,j)$ is for $l\geqslant 1$. In the point-boson limit, the matrix elements [Eq. (4.10)], becomes

$$\langle n,m|\hat{H}_{cd}|i,j\rangle \simeq 4(E_0+E)\mathcal{F}^0(n,m|i,j)+\mathcal{T}^{(0)}(n,m|i,j)+\mathcal{U}^{(0)}(n,m|i,j), \tag{4.11}$$

which is the same form as Eq. (4.7). This results from the fact that the orthonormalized two-exciton state $|i,j\rangle$ is reduced to the two-exciton state $\hat{b}_i\hat{b}_j|0\rangle$ in the point-boson approximation.

Next, we show the matrix element of $\tilde{g}\hat{p}_i^\dagger$ between the one-exciton and orthonormalized two-exciton states,

$$\tilde{g}\langle n,m|\hat{p}_i^\dagger\hat{b}_j^\dagger|0\rangle=\sum_{l=0}^{\infty}(-s)^l\mathcal{P}^{(l)}(n,m|i,j)+\sum_{l=1}^{\infty}(-s)^l\mathcal{Q}^{(l)}(n,m|i,j), \tag{4.12}$$

where $\mathcal{P}^{(l)}(n,m|i,j)$ and $\mathcal{Q}^{(l)}(n,m|i,j)$ are given as Eqs. (C8) and (C9) in Appendix C. It should be noted that $\mathcal{P}^{(l)}(n,m|i,j)$ is defined for $l\geqslant 0$ and $\mathcal{Q}^{(l)}(n,m|i,j)$ is for $l\geqslant 1$. In the point-boson limit for Eq. (4.12), we obtain the same result as Eq. (4.9), i.e.,

$$\tilde{g}\langle n,m|\hat{p}_i^\dagger\hat{b}_j^\dagger|0\rangle\simeq\mathcal{P}^{(0)}(n,m|i,j). \tag{4.13}$$

We arrange the forms of the matrix elements [Eqs. (4.10) and (4.12)] with the use of Eq. (A3) in Appendix A. Classifying terms of the lth-order CPE into even and odd orders, we obtain

$$\langle n,m|\hat{H}_{cd}|i,j\rangle=4(E_0+E)\mathcal{F}^0(n,m|i,j)+\mathcal{T}^{(0)}(n,m|i,j)+\mathcal{I}_d(n,m|i,j)-s\mathcal{I}_x(n,m|i,j), \tag{4.14}$$

$$\tilde{g}\langle n,m|\hat{p}_i^\dagger\hat{b}_j^\dagger|0\rangle=\mathcal{P}^{(0)}(n,m|i,j)+\mathcal{G}_d(n,m|i,j)-s\mathcal{G}_x(n,m|i,j), \tag{4.15}$$

where

$$\mathcal{I}_d(n,m|i,j)=A^0(n,m|i,j)I_d(n,m|i,j)+(i\leftrightarrow j \text{ or } m\leftrightarrow n), \tag{4.16}$$

$$\mathcal{I}_x(n,m|i,j)=A^1(n,m|i,j)I_x(n,m|i,j)+(i\leftrightarrow j \text{ or } m\leftrightarrow n), \tag{4.17}$$

$$\mathcal{G}_d(n,m|i,j)=B^0(n,m|i,j)G_d(n,m|i,j)+(m\leftrightarrow n), \tag{4.18}$$

$$\mathcal{G}_x(n,m|i,j)=B^1(n,m|i,j)G_x(n,m|i,j)+(m\leftrightarrow n). \tag{4.19}$$

Here

$$B^0(n,m|i,j)=\delta_{\sigma_i}^{\mu_m+\nu_m}\delta_{\mu_j}^{\mu_n}\delta_{\nu_j}^{\nu_n}, \tag{4.20}$$

$$B^1(n,m|i,j)=\delta_{\sigma_i}^{\mu_n+\nu_m}\delta_{\mu_j}^{\mu_m}\delta_{\nu_j}^{\nu_n}, \tag{4.21}$$

$$I_d(n,m|i,j)=u^{(0)}(n,m|i,j)+\sum_{l=1}^{\infty}[t^{(2l)}(n,m|i,j)+u^{(2l)}(n,m|i,j)+v^{(2l)}(n,m|i,j)], \tag{4.22}$$

$$I_x(n,m|i,j)=\sum_{l=1}^{\infty}[t^{(2l-1)}(n,m|i,j)+u^{(2l-1)}(n,m|i,j)+v^{(2l-1)}(n,m|i,j)], \tag{4.23}$$

$$G_d(n,m|i,j)=\sum_{l=1}^{\infty}[p^{(2l)}(n,m|i,j)+q^{(2l)}(n,m|i,j)], \tag{4.24}$$

$$G_x(n,m|i,j)=\sum_{l=1}^{\infty}[p^{(2l-1)}(n,m|i,j)+q^{(2l-1)}(n,m|i,j)], \tag{4.25}$$

where $u^{(0)}(n,m|i,j)$ is given in Eq. (B2) in Appendix B and $t^{(l)}(n,m|i,j)$, $u^{(l)}(n,m|i,j)$, $v^{(l)}(n,m|i,j)$, $p^{(l)}(n,m|i,j)$, and $q^{(l)}(n,m|i,j)$ are defined by Eqs. (C2), (C6), (C7), (C11), and (C12) in Appendix C. From these formulas, it turns out that $I_d(n,m|i,j)$ and $I_x(n,m|i,j)$ include the CPE and the interactions V_{cd}, V_{cc}, and V_{dd}, while $G_d(n,m|i,j)$ and $G_x(n,m|i,j)$ contain the CPE and the coupling $\tilde{g}$. With respect to the order of the CPE, $I_d(n,m|i,j)$ contains all the order, $I_x(n,m|i,j)$ and $G_x(n,m|i,j)$ have more than zeroth-order CPE's, and $G_d(n,m|i,j)$ results from more than first-order CPE's. Therefore, only $I_d(n,m|i,j)$ remains in the point-boson limit.

C. Bosonized Hamiltonian

We shall define some functions for the bosonized Hamiltonian. Due to the translational symmetry, a total momentum conserves via the interactions. This momentum conservation appears in $I_d(n,m|i,j)$, $I_x(n,m|i,j)$, $G_d(n,m|i,j)$, and $G_x(n,m|i,j)$ as $\delta_{\mathbf{K}_i+\mathbf{K}_j}^{\mathbf{K}_m+\mathbf{K}_n}$. Therefore, introducing $I_d(\mathbf{Q},\mathbf{K})$, $I_x(\mathbf{Q},\mathbf{K})$, $G_d(\mathbf{Q},\mathbf{K})$, and $G_x(\mathbf{Q},\mathbf{K})$, we rewrite these four functions as

$$I_d(n,m|i,j)=\sum_{\mathbf{Q}}\delta_{\mathbf{K}_i+\mathbf{Q}}^{\mathbf{K}_m}\delta_{\mathbf{K}_j-\mathbf{Q}}^{\mathbf{K}_n}I_d\left(\mathbf{Q},\frac{1}{2}(\mathbf{K}_i-\mathbf{K}_j)\right), \tag{4.26}$$

$$I_x(n,m|i,j)=\sum_{\mathbf{Q}}\delta_{\mathbf{K}_i+\mathbf{Q}}^{\mathbf{K}_m}\delta_{\mathbf{K}_j-\mathbf{Q}}^{\mathbf{K}_n}I_x\left(\mathbf{Q},\frac{1}{2}(\mathbf{K}_i-\mathbf{K}_j)\right), \tag{4.27}$$

$$G_d(n,m|i,j)=\sum_{\mathbf{Q}}\delta_{\mathbf{K}_i+\mathbf{Q}}^{\mathbf{K}_m}\delta_{\mathbf{K}_j-\mathbf{Q}}^{\mathbf{K}_n}G_d\left(\mathbf{Q},\frac{1}{2}(\mathbf{K}_i-\mathbf{K}_j)\right), \tag{4.28}$$

$$G_x(n,m|i,j)=\sum_{\mathbf{Q}}\delta_{\mathbf{K}_i+\mathbf{Q}}^{\mathbf{K}_m}\delta_{\mathbf{K}_j-\mathbf{Q}}^{\mathbf{K}_n}G_x\left(\mathbf{Q},\frac{1}{2}(\mathbf{K}_i-\mathbf{K}_j)\right), \tag{4.29}$$

where $I_d(\mathbf{Q},\mathbf{K})$, $I_x(\mathbf{Q},\mathbf{K})$, $G_d(\mathbf{Q},\mathbf{K})$, and $G_x(\mathbf{Q},\mathbf{K})$ are independent of μ and ν, and $\frac{1}{2}(\mathbf{K}_i-\mathbf{K}_j)$ stands for the relative momentum of two bosonized excitons labeled by i and j. With these functions, we write the final forms of the bosonized Hamiltonian:

$$\hat{H}^B=\sum_{\mu\nu\mathbf{K}}\left(E_0+E+\frac{\hbar\mathbf{K}^2}{2M}\right)\hat{B}^\dagger_{\mu\nu\mathbf{K}}\hat{B}_{\mu\nu\mathbf{K}}+\sum_{\sigma\mathbf{K}}\hbar\omega_{\mathbf{K}}\hat{a}^\dagger_{\sigma\mathbf{K}}\hat{a}_{\sigma\mathbf{K}}+g\sum_{\sigma\mathbf{K}}\sum_{\mu\nu}\delta^{\mu+\nu}_{\sigma}\hat{B}^\dagger_{\mu\nu\mathbf{K}}\hat{a}_{\sigma\mathbf{K}}+(\mathrm{H.c.})$$
$$+\frac{1}{2}\sum_{\mu\nu\mu'\nu'}\sum_{\mathbf{QKK}'}I_d\left(\mathbf{Q},\frac{1}{2}(\mathbf{K}-\mathbf{K}')\right)\hat{B}^\dagger_{\mu\nu\mathbf{K}+\mathbf{Q}}\hat{B}^\dagger_{\mu'\nu'\mathbf{K}'-\mathbf{Q}}\hat{B}_{\mu'\nu'\mathbf{K}'}\hat{B}_{\mu\nu\mathbf{K}}$$
$$+\frac{(-s)}{2}\sum_{\mu\nu\mu'\nu'}\sum_{\mathbf{QKK}'}I_x\left(\mathbf{Q},\frac{1}{2}(\mathbf{K}-\mathbf{K}')\right)\hat{B}^\dagger_{\mu'\nu\mathbf{K}+\mathbf{Q}}\hat{B}^\dagger_{\mu\nu'\mathbf{K}'-\mathbf{Q}}\hat{B}_{\mu'\nu'\mathbf{K}'}\hat{B}_{\mu\nu\mathbf{K}}$$
$$+\sum_{\sigma\mu'\nu'}\sum_{\mathbf{QKK}'}\sum_{\mu\nu}\delta^{\mu+\nu}_{\sigma}G_d\left(\mathbf{Q},\frac{1}{2}(\mathbf{K}-\mathbf{K}')\right)\hat{B}^\dagger_{\mu\nu\mathbf{K}+\mathbf{Q}}\hat{B}^\dagger_{\mu'\nu'\mathbf{K}'-\mathbf{Q}}\hat{B}_{\mu'\nu'\mathbf{K}'}\hat{a}_{\sigma\mathbf{K}}+(\mathrm{H.c.})$$
$$+(-s)\sum_{\sigma\mu'\nu'}\sum_{\mathbf{QKK}'}\sum_{\mu\nu}\delta^{\mu+\nu}_{\sigma}G_x\left(\mathbf{Q},\frac{1}{2}(\mathbf{K}-\mathbf{K}')\right)\hat{B}^\dagger_{\mu'\nu\mathbf{K}+\mathbf{Q}}\hat{B}^\dagger_{\mu\nu'\mathbf{K}'-\mathbf{Q}}\hat{B}_{\mu'\nu'\mathbf{K}'}\hat{a}_{\sigma\mathbf{K}}+(\mathrm{H.c.}), \tag{4.30}$$

where $g\equiv\tilde{g}\sqrt{L^D}\varphi^*(\mathbf{r}=0)$. Note that the subscripts $\{\mu\nu\}$ and $\{\mu'\nu'\}$ of the bosonized excitons before and after the interaction are exchanged in I_x and G_x. This means an exchange of the components of excitons via these interactions, as shown in Fig. 1. We call these terms the "component-exchange scattering term" and the "component-exchange nonlinear-coupling term," respectively. On the other hand, the interactions I_d and G_d do not exchange these subscripts. We call these the "component-direct scattering term" and the "component-direct nonlinear-coupling term."

The exchange of the components is related to the order of the CPE. Since I_d and G_d consist of even orders of the CPE, an exchange of the components takes place even times, resulting in no exchange for the pairs. On the other hand, I_x and G_x contain odd orders of the CPE, leading to the exchange. We also find that only I_d is dominant in the region that the distance between two bosonized excitons is larger than a. This corresponds to the fact that only the interaction I_d has a zeroth-order term of the CPE [see Eq. (4.22)]. In such a region, G_d damps quite rapidly because it has only more than the second-order terms of the CPE [see Eq. (4.24)]. Since only up to the first-order of the CPE is estimated in the incomplete CPE theory, there is no corresponding term of G_d. Moreover, it should be noted that only the component-exchange interaction I_x and the component-exchange nonlinear coupling G_x depend on the statistics of the c and d particles.

Our bosonization method exhibits a normal-ordering expansion of the boson operators, and no boson-number dependence in bosonized operators.[28] Consequently, when we consider a system with more than two excitons, the two-body interactions in Eq. (4.30) remain unchanged, although many-body scattering terms have to be taken into account. Noting that two-body interactions are also of primary importance in

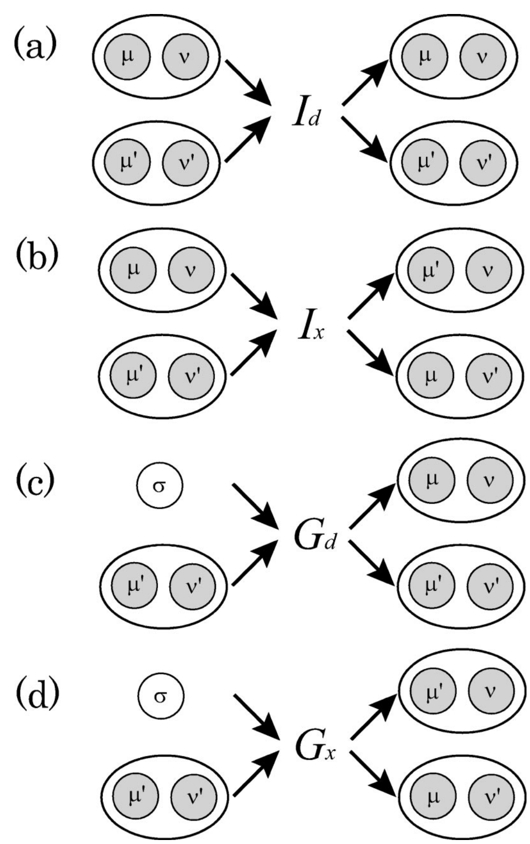

FIG. 1. Schematic diagrams of four kinds of interactions: (a) the component-direct boson-boson interaction I_d, (b) the component-exchange boson-boson interaction I_x, (c) the component-direct boson-photon nonlinear coupling G_d, and (d) the component-exchange boson-photon nonlinear coupling G_x. A pair with μ, ν, μ', and ν' stands for an exciton, and a circle with σ means a photon (an a particle).

many-exciton systems, our theory provides an effective step forward to many-exciton systems.[35] As an example of the above general formulation, we will apply it to a semiconductor bulk system with a photon field in Sec. V.

V. APPLICATION TO A SEMICONDUCTOR BULK SYSTEM

In this section, a semiconductor bulk system with a photon field is considered to derive concrete forms of the bosonized Hamiltonian. We pay special attention to the heavy-hole limit. We also study the formation of bound and unbound states of two bosonized excitons, which correspond to an excitonic molecule and to scattering states of two excitons.

A. Bosonized Hamiltonian for a semiconductor bulk system

We consider a III-V semiconductor bulk system, which consists of two conduction bands and two heavy-hole valence bands denoted by total-spin indices $\mu=\pm 1/2$ and $\nu=\pm 3/2$, respectively. Photons have a polarization of $\sigma=\pm 1$. Light-hole bands are not considered here. This system is equivalent to the model [Eq. (2.1)], by regarding the c particle as a conduction electron, the d particle as a valence hole, and the a particle as a photon. The energy E_0 corresponds to the band-gap energy E_g. We further assume that $V_{cd}=V_{cc}=V_{dd}\equiv V$ and $V(\mathbf{r})=e^2/(\varepsilon|\mathbf{r}|)$, where e is an elementary electric charge and ε is a dielectric constant. For simplicity, let us take a heavy-hole limit, $m_c/m_d \to 0$.[29,30] In this section, we employ a real-space representation, which is suitable for the heavy-hole limit. We consider a three-dimensional $1s$ exciton, whose creation operator and relative energy are

$$\hat{b}^\dagger_{\mu\nu\mathbf{R}}=\int d^3\mathbf{r}\,\varphi_{1s}(\mathbf{r})\hat{c}^\dagger_{\mu\mathbf{R}+\mathbf{r}}\hat{d}^\dagger_{\nu\mathbf{R}}, \tag{5.1}$$

$$E\equiv E_{1s}=-\frac{me^4}{2\varepsilon^2\hbar^2}, \tag{5.2}$$

respectively. Here $\varphi_{1s}(\mathbf{r})$ is the three-dimensional $1s$ exciton wave function for the relative motion,

$$\varphi_{1s}(\mathbf{r})=\frac{1}{\sqrt{\pi a^3}}\exp\left(-\frac{|\mathbf{r}|}{a}\right), \tag{5.3}$$

with the three-dimensional exciton Bohr radius $a=\varepsilon\hbar^2/(me^2)$. Indices $\{\mu_i,\nu_i,\mathbf{R}_i\}$ are abbreviated to i in this section.

The inner product between two-exciton states has the same form as Eq. (2.14),

$$\langle 0|\hat{b}_n\hat{b}_m\hat{b}^\dagger_i\hat{b}^\dagger_j|0\rangle=2\tilde{\mathcal{F}}^0(n,m|i,j)+2s\tilde{\mathcal{F}}^1(n,m|i,j), \tag{5.4}$$

with

$$\tilde{\mathcal{F}}^0(n,m|i,j)=\tfrac{1}{2}[A^0(n,m|i,j)\tilde{f}^0(n,m|i,j)+(i\leftrightarrow j \text{ or } m\leftrightarrow n)], \tag{5.5}$$

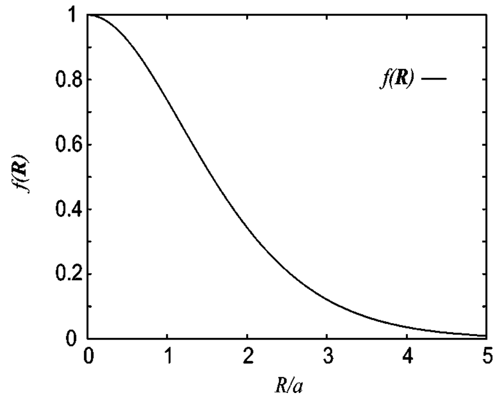

FIG. 2. The function $f(\mathbf{R})$ in Eq. (5.9) is plotted as a function of $|\mathbf{R}|/a$. It becomes unity at the origin, and damps rapidly for $|\mathbf{R}|\gg a$.

$$\tilde{\mathcal{F}}^1(n,m|i,j)=\tfrac{1}{2}[A^1(n,m|i,j)\tilde{f}^1(n,m|i,j)+(i\leftrightarrow j \text{ or } m\leftrightarrow n)], \tag{5.6}$$

$$\tilde{f}^0(n,m|i,j)=\delta^3(\mathbf{R}_m-\mathbf{R}_i)\,\delta^3(\mathbf{R}_n-\mathbf{R}_j), \tag{5.7}$$

$$\tilde{f}^1(n,m|i,j)=\delta^3(\mathbf{R}_m-\mathbf{R}_i)\,\delta^3(\mathbf{R}_n-\mathbf{R}_j)\,f(\mathbf{R}_i-\mathbf{R}_j), \tag{5.8}$$

where $s=-1$ in the present case and $f(\mathbf{R})$ is defined as

$$f(\mathbf{R})=\left[1+\frac{R}{a}+\frac{1}{3}\left(\frac{R}{a}\right)^2\right]^2\exp\left(-\frac{2R}{a}\right), \tag{5.9}$$

with $R=|\mathbf{R}|$. As shown in Fig. 2, the function $f(\mathbf{R})$ becomes unity at the origin and behaves asymptotically as $f(\mathbf{R})\sim\frac{1}{9}(R/a)^4\exp(-2R/a)$ for $|\mathbf{R}|\gg a$. The fact $f(\mathbf{R}=0)=1$ means that two excitons cannot occupy the same place, i.e., the excitons cannot be regarded as bosons, corresponding to $\langle 0|\hat{b}_n\hat{b}_m\hat{b}^\dagger_i\hat{b}^\dagger_j|0\rangle=0$. The asymptotic behavior for $|\mathbf{R}|\gg a$ reflects that the excitons behave as bosons when they separate from each other, resulting from that $\tilde{\mathcal{F}}^1(n,m|i,j)$ in Eq. (5.4), the first-order CPE, disappears for $|\mathbf{R}|\gg a$.

The orthonormalized two-exciton state [Eq. (3.1)] becomes simple in the present case as

$$|\mu_i\nu_i\mathbf{R}_i,\mu_j\nu_j\mathbf{R}_j\rangle=\hat{b}^\dagger_{\mu_i\nu_i\mathbf{R}_i}\hat{b}^\dagger_{\mu_j\nu_j\mathbf{R}_j}|0\rangle F_d(\mathbf{R}_i-\mathbf{R}_j)+\hat{b}^\dagger_{\mu_j\nu_i\mathbf{R}_i}\hat{b}^\dagger_{\mu_i\nu_j\mathbf{R}_j}|0\rangle(-s)F_x(\mathbf{R}_i-\mathbf{R}_j), \tag{5.10}$$

where

$$F_d(\mathbf{R})=\frac{1}{2}\left[\frac{1}{\sqrt{1-f(\mathbf{R})}}+\frac{1}{\sqrt{1+f(\mathbf{R})}}\right], \tag{5.11}$$

$$F_x(\mathbf{R})=\frac{1}{2}\left[\frac{1}{\sqrt{1-f(\mathbf{R})}}-\frac{1}{\sqrt{1+f(\mathbf{R})}}\right], \tag{5.12}$$

which are plotted in Fig. 3. Since the asymptotic forms of $F_d(\mathbf{R})$ and $F_x(\mathbf{R})$ for $|\mathbf{R}|\gg a$ are $F_d(\mathbf{R})\sim 1$

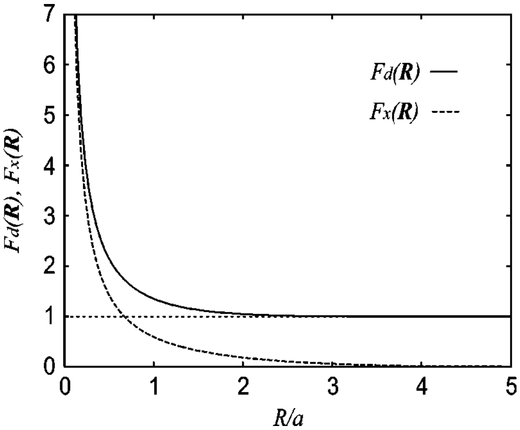

FIG. 3. Two functions $F_d(\mathbf{R})$ and $F_x(\mathbf{R})$ in Eqs. (5.11) and (5.12) are plotted as a function of $|\mathbf{R}|/a$. These approach asymptotically unity and zero, respectively, for $|\mathbf{R}|\gg a$.

$+\frac{1}{216}(R/a)^8\exp(-4R/a)$ and $F_x(\mathbf{R})\sim\frac{1}{18}(R/a)^4\exp(-2R/a)$, respectively, the orthonormalized two-exciton state becomes equivalent to the two-exciton state $\hat{b}^\dagger_{\mu_i\nu_i\mathbf{R}_i}\hat{b}^\dagger_{\mu_j\nu_j\mathbf{R}_j}|0\rangle$ in the case of $|\mathbf{R}_i-\mathbf{R}_j|\gg a$. The point-boson approximation $|\mu_i\nu_i\mathbf{R}_i,\mu_j\nu_j\mathbf{R}_j\rangle\simeq\hat{b}^\dagger_{\mu_i\nu_i\mathbf{R}_i}\hat{b}^\dagger_{\mu_j\nu_j\mathbf{R}_j}|0\rangle\leftrightarrow\hat{B}^\dagger_{\mu_i\nu_i\mathbf{R}_i}\hat{B}^\dagger_{\mu_j\nu_j\mathbf{R}_j}|0)$ is valid only in this case.

The loose matrix element [Eq. (4.6)] becomes

$$\langle 0|\hat{b}_n\hat{b}_m\hat{H}_{cd}\hat{b}_i^\dagger\hat{b}_j^\dagger|0\rangle=4(E_g+E_{1s})[\tilde{\mathcal{F}}^0(n,m|i,j)+s\tilde{\mathcal{F}}^1(n,m|i,j)]+\tilde{\mathcal{U}}^{(0)}(n,m|i,j)-s\tilde{\mathcal{V}}^{(1)}(n,m|i,j),\tag{5.13}$$

where

$$\tilde{\mathcal{U}}^{(0)}(n,m|i,j)=A^0(n,m|i,j)\tilde{u}^{(0)}(n,m|i,j)+(i\leftrightarrow j\ \text{ or }\ m\leftrightarrow n),\tag{5.14}$$

$$\tilde{\mathcal{V}}^{(1)}(n,m|i,j)=A^1(n,m|i,j)\tilde{v}^{(1)}(n,m|i,j)+(i\leftrightarrow j\ \text{ or }\ m\leftrightarrow n),\tag{5.15}$$

$$\tilde{u}^{(0)}(n,m|i,j)=\delta^3(\mathbf{R}_m-\mathbf{R}_i)\,\delta^3(\mathbf{R}_n-\mathbf{R}_j)\,u(\mathbf{R}_i-\mathbf{R}_j),\tag{5.16}$$

$$\tilde{v}^{(1)}(n,m|i,j)=\delta^3(\mathbf{R}_m-\mathbf{R}_i)\,\delta^3(\mathbf{R}_n-\mathbf{R}_j)\,v(\mathbf{R}_i-\mathbf{R}_j).\tag{5.17}$$

Two functions $u(\mathbf{R})$ and $v(\mathbf{R})$ are evaluated analytically[36] as

$$u(\mathbf{R})=\frac{e^2}{2\varepsilon a}\,\frac{2\,\exp(-2r)}{r}\left(1+\frac{5r}{8}-\frac{3r^2}{4}-\frac{r^3}{6}\right),\tag{5.18}$$

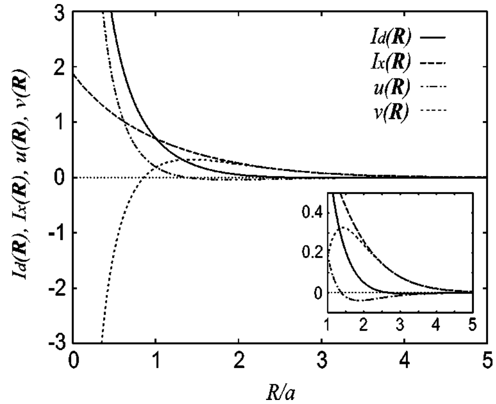

FIG. 4. The loose matrix elements $u(\mathbf{R})$ and $v(\mathbf{R})$, given in Eqs. (5.18) and (5.19), are plotted as a function of $|\mathbf{R}|/a$. The matrix elements $I_d(\mathbf{R})$ and $I_x(\mathbf{R})$ given in Eqs. (5.30) and (5.31) are also plotted. The vertical axis is normalized by a unit of $e^2/2\varepsilon a$. The inset is an enlargement of the main plot.

$$v(\mathbf{R})=\frac{e^2}{2\varepsilon a}\Bigg\{\left(-\frac{2}{r}-\frac{5}{4}+\frac{209r}{30}+\frac{26r^2}{5}+\frac{56r^3}{45}\right)\times\exp(-2r)-\frac{12}{5r}[\gamma S_+^2(r)+S_+^2(r)\ln(r)-2S_+(r)S_-(r)\mathrm{Ei}(-2r)+S_-^2(r)\mathrm{Ei}(-4r)]\Bigg\},\tag{5.19}$$

where $r\equiv|\mathbf{R}|/a$, γ is the Euler's constant, and

$$S_+(r)\equiv\left(1+r+\frac{r^2}{3}\right)\exp(-r),\tag{5.20}$$

$$S_-(r)\equiv\left(1-r+\frac{r^2}{3}\right)\exp(r),\tag{5.21}$$

$$\mathrm{Ei}(-r)\equiv-\int_r^\infty dt\,\frac{\exp(-t)}{t}.\tag{5.22}$$

These $u(\mathbf{R})$ and $v(\mathbf{R})$ are plotted in Fig. 4. The function $u(\mathbf{R})$, which expresses the Coulomb interaction between the charge distributions of excitons, diverges as $1/|\mathbf{R}|$ in the vicinity of the origin ($|\mathbf{R}|\sim 0$), and behaves asymptotically ($|\mathbf{R}|\gg a$) as $u(\mathbf{R})\sim -e^2/(6\varepsilon a)(R/a)^2\exp(-2R/a)$; near the origin the Coulomb repulsive interaction between holes becomes dominant, and for $|\mathbf{R}|\gg a$ the Coulomb interaction between the charge distributions *with no pole* is essential in three-dimensional $1s$ exciton systems. It should be noted that the exponential decay of $u(\mathbf{R})$ for $|\mathbf{R}|\gg a$ results not from the CPE but from the Coulomb interaction between the no-pole charge distributions. If we consider two-dimensional systems instead of three-dimensional ones, $u(\mathbf{R})$ is proportional to $1/|\mathbf{R}|^5$ for $|\mathbf{R}|\gg a$ due to the quadrupole-quadrupole Coulomb interaction.[37] On the other hand, $v(\mathbf{R})$ behaves as $-1/|\mathbf{R}|$ near the origin, and exhibits an exponential decay for $|\mathbf{R}|\gg a$ *due to the CPE*. In Fig. 5, $u(\mathbf{Q})$ and $v(\mathbf{Q})$, the Fourier transforms of $u(\mathbf{R})$ and $v(\mathbf{R})$, are drawn.

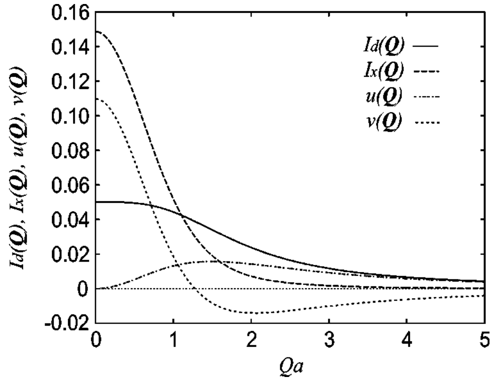

FIG. 5. The Fourier transforms of $u(\mathbf{R})$, $v(\mathbf{R})$, $I_d(\mathbf{R})$, and $I_x(\mathbf{R})$, denoted as $u(\mathbf{Q})$, $v(\mathbf{Q})$, $I_d(\mathbf{Q})$ and $I_x(\mathbf{Q})$, respectively, are plotted as a function of $|\mathbf{Q}|a$. The vertical axis is normalized by a unit of $(2\pi a/L)^3 e^2/2\varepsilon a$. We find that the interactions between two bosonized excitons $I_d(\mathbf{Q})$, and $I_x(\mathbf{Q})$ near $\mathbf{Q}\simeq 0$ are enhanced by the CPE in comparison with the incorrect interactions $u(\mathbf{Q})$ and $v(\mathbf{Q})$, which are estimated with the loose matrix elements.

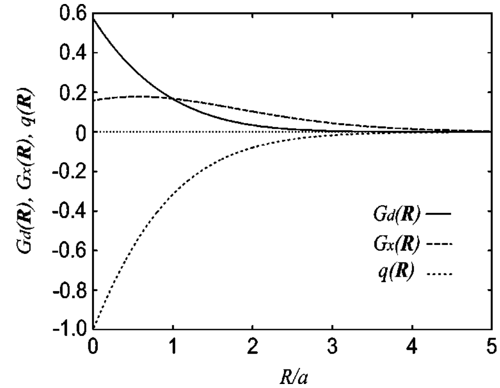

FIG. 6. The function $q(\mathbf{R})$ in Eq. (5.28) is plotted as a function of $|\mathbf{R}|/a$. The nonlinear couplings between the bosonized excitons and a photon, $G_d(\mathbf{R})$ and $G_x(\mathbf{R})$ in Eqs. (5.32) and (5.33), are also plotted. The vertical axis is normalized by a unit of $g=\tilde{g}\sqrt{L^3}\varphi_{1s}^*(0)$.

The loose matrix elements about $\tilde{g}\hat{p}_{\sigma\mathbf{R}}^\dagger$ [Eq. (4.8)] is evaluated as

$$\tilde{g}\langle 0|\hat{b}_n\hat{b}_m\hat{p}_i^\dagger\hat{b}_j^\dagger|0\rangle=\tilde{\mathcal{P}}^{(0)}(n,m|i,j)-s\,\tilde{\mathcal{Q}}^{(1)}(n,m|i,j), \tag{5.23}$$

where

$$\tilde{\mathcal{P}}^{(0)}(n,m|i,j)=B^0(n,m|i,j)\tilde{p}^{(0)}(n,m|i,j)+(m\leftrightarrow n), \tag{5.24}$$

$$\tilde{\mathcal{Q}}^{(1)}(n,m|i,j)=B^1(n,m|i,j)\tilde{q}^{(1)}(n,m|i,j)+(m\leftrightarrow n), \tag{5.25}$$

$$\tilde{p}^{(0)}(n,m|i,j)=\delta^3(\mathbf{R}_m-\mathbf{R}_i)\,\delta^3(\mathbf{R}_n-\mathbf{R}_j)\tilde{g}\sqrt{L^3}\varphi_{1s}^*(0), \tag{5.26}$$

$$\tilde{q}^{(1)}(n,m|i,j)=\delta^3(\mathbf{R}_m-\mathbf{R}_i)\,\delta^3(\mathbf{R}_n-\mathbf{R}_j)q(\mathbf{R}_i-\mathbf{R}_j). \tag{5.27}$$

Here the index i of $\hat{p}_i$ means $\{\sigma_i,\mathbf{R}_i\}$ and $\hat{p}_{\sigma\mathbf{R}}^\dagger=L^{-3/2}\Sigma_{\mathbf{K}}\hat{p}_{\sigma\mathbf{K}}^\dagger\exp(-i\mathbf{K}\cdot\mathbf{R})$. The analytical form of $q(\mathbf{R})$ is

$$q(\mathbf{R})=-g\left[1+\frac{R}{a}+\frac{1}{3}\left(\frac{R}{a}\right)^2\right]\exp\left(-\frac{2R}{a}\right), \tag{5.28}$$

where $g\equiv\tilde{g}\sqrt{L^3}\varphi_{1s}^*(0)$. The function $q(\mathbf{R})$ damps exponentially in the region of $|\mathbf{R}|\gg a$ due to the CPE, as shown in Fig. 6. The behavior of $q(\mathbf{R})$ near the origin indicates that an exciton cannot be created at the position where the other exciton has already stayed. The Fourier transform $q(\mathbf{Q})$ is shown in Fig. 7.

The matrix elements of the Hamiltonian are calculated with the orthonormalized two-exciton states [Eq. (5.10)]. Details of the matrix elements are given in Appendix D. With use of these matrix elements, we finally derive the bosonized Hamiltonian for the semiconductor bulk system in the heavy-hole limit:

$$\begin{aligned}\hat{H}^B=&\sum_{\mu\nu}\int d^3\mathbf{R}(E_g+E_{1s})\hat{B}_{\mu\nu\mathbf{R}}^\dagger\hat{B}_{\mu\nu\mathbf{R}}+\sum_{\sigma\mathbf{K}}\hbar\omega_{\mathbf{K}}\hat{a}_{\sigma\mathbf{K}}^\dagger\hat{a}_{\sigma\mathbf{K}}+g\sum_{\sigma}\int d^3\mathbf{R}\sum_{\mu\nu}\delta_\sigma^{\mu+\nu}\hat{B}_{\mu\nu\mathbf{R}}^\dagger\hat{a}_{\sigma\mathbf{R}}+(\text{H.c.})\\&+\frac{1}{2}\sum_{\mu\nu\mu'\nu'}\int d^3\mathbf{R}d^3\mathbf{R}'I_d(\mathbf{R}-\mathbf{R}')\hat{B}_{\mu\nu\mathbf{R}}^\dagger\hat{B}_{\mu'\nu'\mathbf{R}'}^\dagger\hat{B}_{\mu'\nu'\mathbf{R}'}\hat{B}_{\mu\nu\mathbf{R}}\\&-\frac{s}{2}\sum_{\mu\nu\mu'\nu'}\int d^3\mathbf{R}d^3\mathbf{R}'I_x(\mathbf{R}-\mathbf{R}')\hat{B}_{\mu'\nu\mathbf{R}}^\dagger\hat{B}_{\mu\nu'\mathbf{R}'}^\dagger\hat{B}_{\mu'\nu'\mathbf{R}'}\hat{B}_{\mu\nu\mathbf{R}}\\&+\sum_{\sigma\mu'\nu'}\int d^3\mathbf{R}d^3\mathbf{R}'\sum_{\mu\nu}\delta_\sigma^{\mu+\nu}G_d(\mathbf{R}-\mathbf{R}')\hat{B}_{\mu\nu\mathbf{R}}^\dagger\hat{B}_{\mu'\nu'\mathbf{R}'}^\dagger\hat{B}_{\mu'\nu'\mathbf{R}'}\hat{a}_{\sigma\mathbf{R}}+(\text{H.c.})\\&-s\sum_{\sigma\mu'\nu'}\int d^3\mathbf{R}d^3\mathbf{R}'\sum_{\mu\nu}\delta_\sigma^{\mu+\nu}G_x(\mathbf{R}-\mathbf{R}')\hat{B}_{\mu'\nu\mathbf{R}}^\dagger\hat{B}_{\mu\nu'\mathbf{R}'}^\dagger\hat{B}_{\mu'\nu'\mathbf{R}'}\hat{a}_{\sigma\mathbf{R}}+(\text{H.c.}).\end{aligned} \tag{5.29}$$

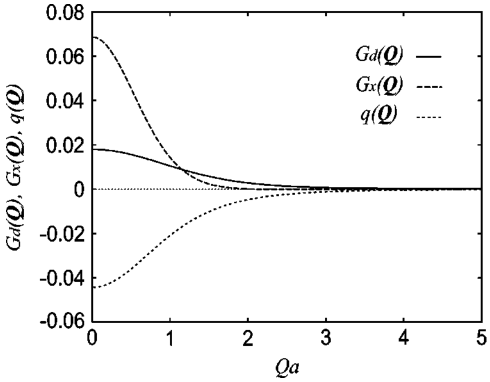

FIG. 7. The Fourier transforms of $q(\mathbf{R})$, $G_d(\mathbf{R})$, and $G_x(\mathbf{R})$, denoted as $q(\mathbf{Q})$, $G_d(\mathbf{Q})$, and $G_x(\mathbf{Q})$, respectively, are plotted as a function of $|\mathbf{Q}|a$. The vertical axis is normalized by a unit of $(2\pi a/L)^3 g$.

The first three terms of $\hat{H}^B$ are the free Hamiltonian for the bosonized excitons, the photons, and a linear coupling among them. The other four terms express the boson-boson scattering terms, whose coefficients are $I_d(\mathbf{R})$ and $I_x(\mathbf{R})$, and the boson-photon nonlinear-coupling terms, whose coefficients are $G_d(\mathbf{R})$ and $G_x(\mathbf{R})$. All of the coefficients are functions only of $\mathbf{R}$, and given as

$$I_d(\mathbf{R})=F_1(\mathbf{R})u(\mathbf{R})+F_2(\mathbf{R})v(\mathbf{R}), \tag{5.30}$$

$$I_x(\mathbf{R})=F_2(\mathbf{R})u(\mathbf{R})+F_1(\mathbf{R})v(\mathbf{R}), \tag{5.31}$$

$$G_d(\mathbf{R})=g[F_d(\mathbf{R})-1]+F_x(\mathbf{R})q(\mathbf{R}), \tag{5.32}$$

$$G_x(\mathbf{R})=gF_x(\mathbf{R})+F_d(\mathbf{R})q(\mathbf{R}). \tag{5.33}$$

The scattering coefficients $I_d(\mathbf{R})$ and $I_x(\mathbf{R})$ are plotted in Fig. 4, and the nonlinear coupling coefficients $G_d(\mathbf{R})$ and $G_x(\mathbf{R})$ are in Fig. 6. Here $u(\mathbf{R})$, $v(\mathbf{R})$, $F_d(\mathbf{R})$, $F_x(\mathbf{R})$, and $q(\mathbf{R})$ are given in Eqs. (5.18), (5.19), (5.11), (5.12), and (5.28), respectively, and

$$F_1(\mathbf{R})=\frac{1}{2}\left[\frac{1}{1-f(\mathbf{R})}+\frac{1}{1+f(\mathbf{R})}\right], \tag{5.34}$$

$$F_2(\mathbf{R})=\frac{1}{2}\left[\frac{1}{1-f(\mathbf{R})}-\frac{1}{1+f(\mathbf{R})}\right]. \tag{5.35}$$

Since the CPE disappears for $|\mathbf{R}|\gg a$, $I_d(\mathbf{R})$ and $I_x(\mathbf{R})$ are reduced respectively to $u(\mathbf{R})$ and $v(\mathbf{R})+f(\mathbf{R})u(\mathbf{R})$, which are the lowest-order CPE terms of the $I_d(\mathbf{R})$ and $I_x(\mathbf{R})$. Since the lowest-order of the CPE in $G_d(\mathbf{R})$ is second order, $G_d(\mathbf{R})$ damps quite rapidly for $|\mathbf{R}|\gg a$. On the other hand, the asymptotic behavior of $G_x(\mathbf{R})$ can be reduced to $q(\mathbf{R})+f(\mathbf{R})/2$, which is the first-order CPE in $G_x(\mathbf{R})$. These reduction forms correspond to the results of the incomplete CPE theory, whose scattering and nonlinear-coupling coefficients are described as $I_d(\mathbf{R})=u(\mathbf{R})$, $I_x(\mathbf{R})=v(\mathbf{R})$, and $G_x(\mathbf{R})=q(\mathbf{R})$. There is no corresponding term of $G_d(\mathbf{R})$.[21–24,26,27] Correspondence between the coefficients in the complete- and incomplete CPE theory is summarized in Table I.

The Fourier transforms of $I_d(\mathbf{R})$, $I_x(\mathbf{R})$, $G_d(\mathbf{R})$, and $G_x(\mathbf{R})$, which are denoted, respectively, as $I_d(\mathbf{Q})$, $I_x(\mathbf{Q})$, $G_d(\mathbf{Q})$, and $G_x(\mathbf{Q})$, are shown in Figs. 5 and 7. In these figures, the behavior around $\mathbf{Q}=0$ is remarkable. The strength of the scattering terms has often been estimated by the value at $\mathbf{Q}=0$. Compared with the previous results,[21–24,26,27] the scattering strength between the bosonized excitons are enhanced by the CPE, as shown in Fig. 5. Moreover, the behavior of the nonlinear couplings, $G_d(\mathbf{Q})$, and $G_x(\mathbf{Q})$, near $\mathbf{Q}=0$ is qualitatively different from the previous results using the Usui transformation.[21,22] A reason for this disagreement is that in our theory the strength of the couplings is estimated with the use of the matrix elements, which include the CPE correctly, while in the Usui transformation it is estimated with the loose matrix elements.

As shown in Eq. (5.29), the scatterings described by $I_x(\mathbf{R})$ and $G_x(\mathbf{R})$ exchange the indices of the bosonized excitons. Therefore, it is possible to create the bosonized excitons, to which the dipole transition is forbidden, i.e., $\mu+\nu=\pm 2$. For example, two bosonized excitons of $(\mu=-1/2,\nu=3/2)$ and $(\mu=1/2,\nu=-3/2)$ change to those of $(\mu=1/2,\nu=3/2)$ and $(\mu=-1/2,\nu=-3/2)$ after I_x scattering, and a bosonized exciton of $(\mu=-1/2,\nu=3/2)$ is transformed to two bosonized excitons of $(\mu=1/2,\nu=3/2)$ and $(\mu=-1/2,\nu=-3/2)$ after the nonlinear interaction G_x with a photon of $\sigma=-1$.

B. Bound and unbound states of the two bosonized excitons

We show that our boson theory can explain both bound and unbound states of the two bosonized excitons, which correspond to excitonic molecules and scattering states of two excitons, respectively. We first define the "triplet states," $|t;1)$, $|t;-1)$, and $|t;0)$, and the "singlet state," $|s)$ as

TABLE I. Correspondence between scattering and nonlinear-coupling coefficients in the complete CPE theory and the incomplete CPE theory. Coefficients up to the first order of the CPE are also listed for comparison.

Complete CPE theory (exact)	Complete CPE theory (up to the first order of the CPE)	Incomplete CPE theory
$I_d(\mathbf{R})$	$u(\mathbf{R})$	$u(\mathbf{R})$
$I_x(\mathbf{R})$	$v(\mathbf{R})+f(\mathbf{R})u(\mathbf{R})$	$v(\mathbf{R})$
$G_d(\mathbf{R})$	0	none
$G_x(\mathbf{R})$	$q(\mathbf{R})+f(\mathbf{R})/2$	$q(\mathbf{R})$

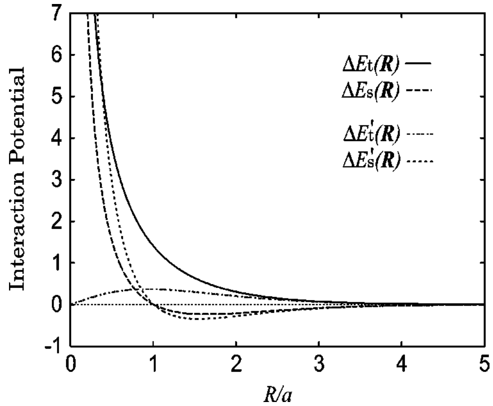

FIG. 8. The interaction potentials for the triplet states and the singlet state, $\Delta E_t(\mathbf{R})$ and $\Delta E_s(\mathbf{R})$ in Eqs. (5.41) and (5.42), are plotted as a function of $|\mathbf{R}|/a$ for $s=-1$. The vertical axis is normalized by a unit of $e^2/2\varepsilon a$. The incorrect interaction potentials, $\Delta E_t'(\mathbf{R})$ and $\Delta E_s'(\mathbf{R})$ are also drawn for comparison.

$$|t;\pm 1)\equiv \hat{B}^{\dagger}_{\pm(1/2)\nu\mathbf{R}}\hat{B}^{\dagger}_{\pm(1/2)\nu'\mathbf{R}'}|0), \tag{5.36}$$

$$|t;0)\equiv \frac{1}{\sqrt{2}}[\hat{B}^{\dagger}_{(1/2)\nu\mathbf{R}}\hat{B}^{\dagger}_{-(1/2)\nu'\mathbf{R}'}+\hat{B}^{\dagger}_{-(1/2)\nu\mathbf{R}}\hat{B}^{\dagger}_{(1/2)\nu'\mathbf{R}'}]|0), \tag{5.37}$$

$$|s)\equiv \frac{1}{\sqrt{2}}[\hat{B}^{\dagger}_{(1/2)\nu\mathbf{R}}\hat{B}^{\dagger}_{-(1/2)\nu'\mathbf{R}'}-\hat{B}^{\dagger}_{-(1/2)\nu\mathbf{R}}\hat{B}^{\dagger}_{(1/2)\nu'\mathbf{R}'}]|0), \tag{5.38}$$

where ν and ν' are arbitrary. For two-photon absorption processes, $|\mathrm{t};\pm 1)$ are allowed for the cocircular polarization configuration, while $|t;0)$ and $|s)$ are allowed for the counter circular configuration. We shall calculate an energy expectation value for these four states as a function of $\mathbf{R}-\mathbf{R}'$, that is,

$$(t;\zeta|\hat{H}^B_{cd}|t;\zeta)=2(E_g+E_{1s})+\Delta E_t(\mathbf{R}-\mathbf{R}'), \tag{5.39}$$

$$(s|\hat{H}^B_{cd}|s)=2(E_g+E_{1s})+\Delta E_s(\mathbf{R}-\mathbf{R}'), \tag{5.40}$$

where $\zeta=-1$, 0, or 1 and the interaction potentials, $\Delta E_t(\mathbf{R})$ and $\Delta E_s(\mathbf{R})$, are given as

$$\Delta E_t(\mathbf{R})=I_d(\mathbf{R})+(-s)I_x(\mathbf{R}), \tag{5.41}$$

$$\Delta E_s(\mathbf{R})=I_d(\mathbf{R})-(-s)I_x(\mathbf{R}). \tag{5.42}$$

These $\Delta E_t(\mathbf{R})$ and $\Delta E_s(\mathbf{R})$ are drawn in Fig. 8 for the case of $s=-1$. We find that $\Delta E_t(\mathbf{R})$ is positive for arbitrary $|\mathbf{R}|$, while $\Delta E_s(\mathbf{R})$ becomes negative around $|\mathbf{R}|=a$. Therefore, the triplet and singlet states correspond to unbound states and a bound state of the two bosonized excitons, respectively. In a fictitious case of $s=+1$, i.e., the components of an exciton are bosons, the triplet states correspond to bound states, and the singlet state corresponds to the unbound state. Our treatment on the interaction potentials is a modified version of the Heitler-London theory from the viewpoint of the bosonized excitons. In fact, these interaction potentials agree with the result of the Heitler-London theory, which treats *a* as a variational parameter.

Here we see the interaction strength between the bosonized excitons with the use of the loose matrix elements in Sec. IV A. We stress again that the use of loose matrix elements instead of the matrix elements is an unreliable approximation in the bosonization procedure: the point-boson approximation. Under this approximation, $I_d(\mathbf{R})$ and $I_x(\mathbf{R})$ in the bosonized Hamiltonian become $u(\mathbf{R})$ and $v(\mathbf{R})$, respectively. With these interactions, the interaction potentials of the triplet states and the singlet state are evaluated as

$$\Delta E_t'(\mathbf{R})=u(\mathbf{R})+(-s)v(\mathbf{R}), \tag{5.43}$$

$$\Delta E_s'(\mathbf{R})=u(\mathbf{R})-(-s)v(\mathbf{R}), \tag{5.44}$$

which are shown also in Fig. 8 for $s=-1$. We can easily find that $\Delta E_t'(\mathbf{R})$ shows a qualitative difference from $\Delta E_t(\mathbf{R})$ in the region of $|\mathbf{R}|<a$. Thus employing loose matrix elements leads to incorrect results, in particular, in the region where the excitons overlap with each other.

VI. CONCLUSIONS

Starting with the Hamiltonian describing interband electron-hole excitations, excitons, in semiconductors coupled with a photon field, we derive a bosonized Hamiltonian for two-exciton optical processes, taking into full account deviations of the excitons from ideal bosons. In the bosonization procedure, we have clarified how the exciton-exciton and exciton-photon interactions depend on the quantum statistics of the component particles of an exciton, on the momenta of the exciton center-of-mass motion, and on the internal degrees of freedom of the components. In the case where two excitons overlap one another, deviations of excitons from ideal bosons become crucial, which affect the interactions among bosonized excitons and photons. We have called such effects the composite-particle effect (CPE). It is stressed again that our bosonization can map two-exciton systems with an *arbitrary* exciton Bohr radius *a* to the *ideal* boson systems even when the point-boson approximation is invalid, e.g., $|\mathbf{R}-\mathbf{R}'|\sim a$. In the bosonized Hamiltonian, there are four nontrivial interaction terms— the component-direct boson-boson scattering term denoted by I_d, the component-exchange boson-boson scattering term I_x, the component-direct nonlinear-coupling term G_d, and the component-exchange nonlinear-coupling term G_x—between the bosonized excitons and a photon. In terms of the CPE order, I_d starts from a zeroth-order CPE, I_x and G_x from more than a zeroth-order CPE and G_d from more than a first-order CPE. It has also been shown that the quantum statistics of the components affects only the signs of I_x and G_x. As an example, we have studied a semiconductor bulk system with a radiation field in the heavy-hole limit. The strength of $I_d(\mathbf{Q})$ and $I_x(\mathbf{Q})$ in the vicinity of $\mathbf{Q}=0$ is enhanced by the CPE. We have also shown that our theory can correctly explain the bound and unbound states of the two bosonized excitons.

The bosonization method used in this paper exhibits normal-ordering expansion of the boson operators, and no boson-number dependence in the bosonized operators.[28,33] Accordingly, when we consider more than two exciton systems, the two-body interactions I_d, I_x, G_d, and G_x obtained in this paper remain unchanged. Then we believe that our bosonization to two-exciton systems can be an important step forward in understanding excitonic nonlinear optical processes and the nature of many-exciton states.

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APPENDIX A: DEFINITION OF THE COEFFICIENT IN EQ. (3.1)

We shall give a definition of $\mathcal{F}^l(n,m|i,j)$ for any integer $l\geqslant 1$, which expresses the lth-order CPE. We first introduce two definitions $f^l(n,m|i,j)$ and $A^l(n,m|i,j)$,

$$A^l(n,m|i,j)\equiv\left(\prod_{k=1}^{l-1}\sum_{i^{(k)}j^{(k)}}\right)A^1(n,m|i^{(l-1)},j^{(l-1)})\times A^1(j^{(l-1)},i^{(l-1)}|i^{(l-2)},j^{(l-2)})\times\dots\times A^1(j^{(2)},i^{(2)}|i^{(1)},j^{(1)})A^1(j^{(1)},i^{(1)}|i,j),\tag{A1}$$

$$f^l(n,m|i,j)\equiv\left(\prod_{k=1}^{l-1}\sum_{i^{(k)}j^{(k)}}\right)f^1(n,m|i^{(l-1)},j^{(l-1)})\times f^1(j^{(l-1)},i^{(l-1)}|i^{(l-2)},j^{(l-2)})\times\dots\times f^1(j^{(2)},i^{(2)}|i^{(1)},j^{(1)})f^1(j^{(1)},i^{(1)}|i,j),\tag{A2}$$

where $i^{(k)}=\{\mu_{i^{(k)}},\nu_{i^{(k)}}\}$ in Eq. (A1), $i^{(k)}=\mathbf{K}_{i^{(k)}}$ in Eq. (A2), and $A^1(n,m|i,j)$ and $f^1(n,m|i,j)$ are given in Eqs. (2.18) and (2.20). Here $A^l(n,m|i,j)$ can be reduced to

$$A^l(n,m|i,j)=\begin{cases}A^0(n,m|i,j) & \text{for } l=\text{even}\\ A^1(n,m|i,j) & \text{for } l=\text{odd}\end{cases}\tag{A3}$$

On the other hand, $f^l(n,m|i,j)$ has a wave-number conservation rule of

$$f^l(n,m|i,j)=0,\quad \text{if } \mathbf{K}_m+\mathbf{K}_n\neq\mathbf{K}_i+\mathbf{K}_j.\tag{A4}$$

Next we define $\mathcal{F}^l(n,m|i,j)$ as

$$\mathcal{F}^l(n,m|i,j)\equiv\left(\prod_{k=1}^{l-1}\sum_{i^{(k)}j^{(k)}}\right)\mathcal{F}^1(n,m|i^{(l-1)},j^{(l-1)})\times\mathcal{F}^1(j^{(l-1)},i^{(l-1)}|i^{(l-2)},j^{(l-2)})\times\dots\times\mathcal{F}^1(j^{(2)},i^{(2)}|i^{(1)},j^{(1)})\mathcal{F}^1(j^{(1)},i^{(1)}|i,j),\tag{A5}$$

where $i^{(k)}=\{\mu_{i^{(k)}},\nu_{i^{(k)}},\mathbf{K}_{i^{(k)}}\}$ and $\mathcal{F}^1(n,m|i,j)$ is given in Eq. (2.16). We can rewrite this in terms of $f^l(n,m|i,j)$ and $A^l(n,m|i,j)$ as

$$\mathcal{F}^l(n,m|i,j)=\tfrac{1}{2}[A^l(n,m|i,j)f^l(n,m|i,j)+(i\leftrightarrow j\ \text{ or }\ m\leftrightarrow n)].\tag{A6}$$

Here we note some useful relations:

$$\mathcal{F}^l(n,m|i,j)=\mathcal{F}^l(n,m|j,i)=\mathcal{F}^l(m,n|i,j),\tag{A7}$$

$$\mathcal{F}^l(j,i|m,n)=[\mathcal{F}^l(n,m|j,i)]^*.\tag{A8}$$

APPENDIX B: DEFINITIONS OF SOME FUNCTIONS IN THE LOOSE MATRIX ELEMENTS

This appendix is devoted to definitions of some functions in Sec. IV A. The interaction energy among density distributions of the components of excitons, $\mathcal{U}^{(0)}(n,m|i,j)$, which is a zeroth-order CPE, is defined as

$$\mathcal{U}^{(0)}(n,m|i,j)=A^0(n,m|i,j)u^{(0)}(n,m|i,j)+(i\leftrightarrow j\ \text{ or }\ m\leftrightarrow n),\tag{B1}$$

where

$$u^{(0)}(n,m|i,j)=u_{cd}(n,m|i,j)+u_{cc}(n,m|i,j)+u_{dd}(n,m|i,j),\tag{B2}$$

with

$$u_{cd}(n,m|i,j)=-2\sum_{\mathbf{Q}}\delta^{\mathbf{K}_m}_{\mathbf{K}_i+\mathbf{Q}}\delta^{\mathbf{K}_n}_{\mathbf{K}_j-\mathbf{Q}}V_{cd}(\mathbf{Q})\times\left[\sum_{\mathbf{k}}\phi^*(\alpha\mathbf{Q}+\mathbf{k})\phi(\mathbf{k})\right]\times\left[\sum_{\mathbf{k}}\phi^*(\beta\mathbf{Q}+\mathbf{k})\phi(\mathbf{k})\right],\tag{B3}$$

$$u_{cc}(n,m|i,j)=\sum_{\mathbf{Q}}\delta^{\mathbf{K}_m}_{\mathbf{K}_i+\mathbf{Q}}\delta^{\mathbf{K}_n}_{\mathbf{K}_j-\mathbf{Q}}V_{cc}(\mathbf{Q})\times\left[\sum_{\mathbf{k}}\phi^*(\beta\mathbf{Q}+\mathbf{k})\phi(\mathbf{k})\right]^2,\tag{B4}$$

$$u_{dd}(n,m|i,j)=\sum_{\mathbf{Q}}\delta^{\mathbf{K}_m}_{\mathbf{K}_i+\mathbf{Q}}\delta^{\mathbf{K}_n}_{\mathbf{K}_j-\mathbf{Q}}V_{dd}(\mathbf{Q})\times\left[\sum_{\mathbf{k}}\phi^*(\alpha\mathbf{Q}+\mathbf{k})\phi(\mathbf{k})\right]^2.\tag{B5}$$

The interaction for the exchange of the exciton components, $\mathcal{V}^{(1)}(n,m|i,j)$, the first-order CPE, is

$$\mathcal{V}^{(1)}(n,m|i,j)=A^1(n,m|i,j)v^{(1)}(n,m|i,j) + (i\leftrightarrow j \text{ or } m\leftrightarrow n), \tag{B6}$$

where

$$v^{(1)}(n,m|i,j)=v_{cd}(n,m|i,j)+v_{cc}(n,m|i,j)+v_{dd}(n,m|i,j), \tag{B7}$$

with

$$\begin{aligned} v_{cd}(n,m|i,j)=\frac{1}{2}\delta^{\mathbf{K}_m+\mathbf{K}_n}_{\mathbf{K}_i+\mathbf{K}_j}\sum_{\mathbf{q}} V_{cd}(\mathbf{q})\sum_{\mathbf{k}} [&\phi^*(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\mathbf{k}-\mathbf{q})\phi^*(\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k})\\ &\times\phi(\mathbf{k})\phi(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k})+\phi^*(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\mathbf{k})\phi^*(\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k}-\mathbf{q})\\ &\times\phi(\mathbf{k})\phi(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k})+\phi^*(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\mathbf{k})\phi^*(\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k})\\ &\times\phi(\mathbf{k}-\mathbf{q})\phi(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k})+\phi^*(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\mathbf{k})\phi^*(\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k})\\ &\times\phi(\mathbf{k})\phi(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k}-\mathbf{q})], \end{aligned} \tag{B8}$$

$$\begin{aligned} v_{cc}(n,m|i,j)=-\delta^{\mathbf{K}_m+\mathbf{K}_n}_{\mathbf{K}_i+\mathbf{K}_j}\sum_{\mathbf{q}} V_{cc}(\mathbf{q})\sum_{\mathbf{k}} &\phi^*(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\mathbf{k})\phi^*(\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k}-\mathbf{q})\\ &\times\phi(\mathbf{k}-\mathbf{q})\phi(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k}), \end{aligned} \tag{B9}$$

$$\begin{aligned} v_{dd}(n,m|i,j)=-\delta^{\mathbf{K}_m+\mathbf{K}_n}_{\mathbf{K}_i+\mathbf{K}_j}\sum_{\mathbf{q}} V_{dd}(\mathbf{q})\sum_{\mathbf{k}} &\phi^*(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\mathbf{k}-\mathbf{q})\phi^*(\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k})\\ &\times\phi(\mathbf{k}-\mathbf{q})\phi(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k}). \end{aligned} \tag{B10}$$

Next we give the definitions of $\mathcal{P}^{(0)}(n,m|i,j)$ and $\mathcal{Q}^{(1)}(n,m|i,j)$ as

$$\mathcal{P}^{(0)}(n,m|i,j)=B^0(n,m|i,j)p^{(0)}(n,m|i,j)+(m\leftrightarrow n), \tag{B11}$$

$$\mathcal{Q}^{(1)}(n,m|i,j)=B^1(n,m|i,j)q^{(1)}(n,m|i,j)+(m\leftrightarrow n), \tag{B12}$$

where $B^0(n,m|i,j)$ and $B^1(n,m|i,j)$ are given in Eqs. (4.20) and (4.21), respectively, and

$$p^{(0)}(n,m|i,j)=\tilde{g}\,\delta^{\mathbf{K}_m}_{\mathbf{K}_i}\delta^{\mathbf{K}_n}_{\mathbf{K}_j}\sqrt{L^D}\varphi^*(0), \tag{B13}$$

$$\begin{aligned} q^{(1)}(n,m|i,j)=-\tilde{g}\sum_{\mathbf{k}} &\phi^*(\alpha(\mathbf{K}_m-\mathbf{K}_j)+\mathbf{k})\\ &\times\phi^*(\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k})\phi(\alpha(\mathbf{K}_m-\mathbf{K}_j)\\ &+\beta(\mathbf{K}_m-\mathbf{K}_i)+\mathbf{k}). \end{aligned} \tag{B14}$$

APPENDIX C: DEFINITIONS OF SOME FUNCTIONS IN THE MATRIX ELEMENTS

Here we give the definition of the functions in the matrix elements [Eqs. (4.10) and (4.12)]. The definition of $\mathcal{T}^{(l)}(n,m|i,j)$ is

$$\begin{aligned} \mathcal{T}^{(l)}(n,m|i,j)=&\frac{1}{2}\sum_{l'=0}^{l}(\xi_{l'}\eta_{l-l'}+\eta_{l'}\xi_{l-l'})\\ &\times\sum_{n'm'i'j'}\mathcal{F}^{l'}(n,m|m',n')\\ &\times\mathcal{T}^{(0)}(n',m'|i',j')\mathcal{F}^{l-l'}(j',i'|i,j)\\ =&A^l(n,m|i,j)t^{(l)}(n,m|i,j)\\ &+(i\leftrightarrow j \text{ or } m\leftrightarrow n), \end{aligned} \tag{C1}$$

where $\xi_l=(2l)!/(2^l l!)^2$, $\mathcal{T}^{(0)}(n,m|i,j)$ and $\mathcal{F}^l(n,m|i,j)$ are defined in Eqs. (4.2) and (A5), and

$$\begin{aligned} t^{(l)}(n,m|i,j)=&\frac{1}{2}\sum_{l'=0}^{l}(\xi_{l'}\eta_{l-l'}+\eta_{l'}\xi_{l-l'})\\ &\times\sum_{\mathbf{K}_{i'}\mathbf{K}_{j'}}f^{l'}(n,m|i'j')\frac{\hbar^2}{2M}(\mathbf{K}_{i'}^2+\mathbf{K}_{j'}^2)\\ &\times f^{l-l'}(j',i'|i,j), \end{aligned} \tag{C2}$$

$$\eta_l=\begin{cases}\xi_0 & \text{for } l=0\\ \xi_l-\xi_{l-1} & \text{for } l\geqslant 1.\end{cases} \tag{C3}$$

According to this definition, $\mathcal{T}^{(1)}(n,m|i,j)=0$ because of $\xi_0\eta_1+\eta_0\xi_1=0$.

Next, $\mathcal{U}^{(l)}(n,m|i,j)$ and $\mathcal{V}^{(l)}(n,m|i,j)$ are defined as

$$\mathcal{U}^{(l)}(n,m|i,j)=\sum_{l'=0}^{l}\xi_{l'}\xi_{l-l'}\sum_{n'm'i'j'}\mathcal{F}^{l'}(n,m|m',n')\times\mathcal{U}^{(0)}(n',m'|i',j')\mathcal{F}^{l-l'}(j',i'|i,j)=A^{l}(n,m|i,j)u^{(l)}(n,m|i,j)+(i\leftrightarrow j\ \text{ or }\ m\leftrightarrow n),\tag{C4}$$

$$\mathcal{V}^{(l)}(n,m|i,j)=\sum_{l'=0}^{l-1}\xi_{l'}\xi_{l-1-l'}\sum_{n'm'i'j'}\mathcal{F}^{l'}(n,m|m',n')\times\mathcal{V}^{(1)}(n',m'|i',j')\mathcal{F}^{l-1-l'}(j',i'|i,j)=A^{l}(n,m|i,j)v^{(l)}(n,m|i,j)+(i\leftrightarrow j\ \text{ or }\ m\leftrightarrow n),\tag{C5}$$

where $\mathcal{U}^{(0)}(n,m|i,j)$ and $\mathcal{V}^{(1)}(n,m|i,j)$ are given in Eqs. (B1) and (B6), and

$$u^{(l)}(n,m|i,j)=\sum_{l'=0}^{l}\xi_{l'}\xi_{l-l'}\sum_{\mathbf{K}_{m'n'i'j'}}f^{l'}(n,m|m',n')\times u^{(0)}(n',m'|i',j')f^{l-l'}(j',i'|i,j),\tag{C6}$$

$$v^{(l)}(n,m|i,j)=\sum_{l'=0}^{l-1}\xi_{l'}\xi_{l-1-l'}\sum_{\mathbf{K}_{m'n'i'j'}}f^{l'}(n,m|m',n')\times v^{(1)}(n',m'|i',j')f^{l-1-l'}(j',i'|i,j),\tag{C7}$$

with $u^{(0)}(n,m|i,j)$ and $v^{(1)}(n,m|i,j)$ given in Eqs. (B2) and (B7). The summation runs over $\mathbf{K}_{m'}$, $\mathbf{K}_{n'}$, $\mathbf{K}_{i'}$, and $\mathbf{K}_{j'}$. Here note that $\mathcal{U}^{(l)}(n,m|i,j)$ and $\mathcal{V}^{(l)}(n,m|i,j)$ are defined for $l\geqslant 0$ and $l\geqslant 1$, respectively.

In Eq. (4.12), $\mathcal{P}^{(l)}(n,m|i,j)$ and $\mathcal{Q}^{(l)}(n,m|i,j)$ are given as

$$\mathcal{P}^{(l)}(n,m|i,j)=\xi_l\sum_{m'n'}\mathcal{F}^{l}(n,m|m',n')\mathcal{P}^{(0)}(n',m'|i,j)=B^{l}(n,m|i,j)p^{(l)}(n,m|i,j)+(m\leftrightarrow n),\tag{C8}$$

$$\mathcal{Q}^{(l)}(n,m|i,j)=\xi_{l-1}\sum_{m'n'}\mathcal{F}^{l-1}(n,m|m',n')\times\mathcal{Q}^{(1)}(n',m'|i,j)=B^{l}(n,m|i,j)q^{(l)}(n,m|i,j)+(m\leftrightarrow n),\tag{C9}$$

where $\mathcal{P}^{(0)}(n,m|i,j)$ and $\mathcal{Q}^{(1)}(n,m|i,j)$ are given in Eqs. (B11) and (B12), and

$$B^{l}(n,m|i,j)=\sum_{\mu_i\nu_i}\delta_{\sigma_i}^{\mu_i+\nu_i}A^{l}(n,m|i,j)=\begin{cases}B^{0}(n,m|i,j) & \text{for } l=\text{even}\\ B^{1}(n,m|i,j) & \text{for } l=\text{odd},\end{cases}\tag{C10}$$

$$p^{(l)}(n,m|i,j)=\sum_{\mathbf{K}_{m'}\mathbf{K}_{n'}}\xi_l f^{l}(n,m|m',n')p^{(0)}(n',m'|i,j),\tag{C11}$$

$$q^{(l)}(n,m|i,j)=\sum_{\mathbf{K}_{m'}\mathbf{K}_{n'}}\xi_{l-1}f^{l-1}(n,m|m',n')\times q^{(1)}(n',m'|i,j),\tag{C12}$$

with $B^0(n,m|i,j)$ and $B^1(n,m|i,j)$ in Eqs. (4.20) and (4.21), and $p^{(0)}(n,m|i,j)$ and $q^{(1)}(n,m|i,j)$ in Eqs. (B13) and (B14). Here $\mathcal{P}^{(l)}(n,m|i,j)$ and $\mathcal{Q}^{(l)}(n,m|i,j)$ are defined for $l\geqslant 0$ and $l\geqslant 1$, respectively.

APPENDIX D: MATRIX ELEMENTS FOR A SEMICONDUCTOR BULK SYSTEM

The matrix elements of $\hat{H}_{cd}$ and $\tilde{g}\hat{p}^{\dagger}_{\sigma\mathbf{R}}$ for a semiconductor bulk system are given here in the heavy-hole limit. With the use of the orthonormalized two-exciton states [Eq. (5.10)] we obtain the matrix elements of $\hat{H}_{cd}$ as

$$\langle n,m|\hat{H}_{cd}|i,j\rangle=4(E_g+E_{1s})\mathcal{F}^{0}(n,m|i,j)+\tilde{\mathcal{I}}_d(n,m|i,j)-s\tilde{\mathcal{I}}_x(n,m|i,j),\tag{D1}$$

where

$$\tilde{\mathcal{I}}_d(n,m|i,j)=A^{0}(n,m|i,j)\delta^3(\mathbf{R}_m-\mathbf{R}_i)\delta^3(\mathbf{R}_n-\mathbf{R}_j)\times I_d(\mathbf{R}_i-\mathbf{R}_j)+(i\leftrightarrow j\ \text{ or }\ m\leftrightarrow n),\tag{D2}$$

$$\tilde{\mathcal{I}}_x(n,m|i,j)=A^{1}(n,m|i,j)\delta^3(\mathbf{R}_m-\mathbf{R}_i)\delta^3(\mathbf{R}_n-\mathbf{R}_j)\times I_x(\mathbf{R}_i-\mathbf{R}_j)+(i\leftrightarrow j\ \text{ or }\ m\leftrightarrow n).\tag{D3}$$

Here $I_d(\mathbf{R})$ and $I_x(\mathbf{R})$ are given in Eqs. (5.30) and (5.31). The matrix element of $\tilde{g}\hat{p}^{\dagger}_{\sigma\mathbf{R}}$ is

$$\tilde{g}\langle n,m|\hat{p}_i^{\dagger}\hat{b}_j^{\dagger}|0\rangle=\tilde{\mathcal{P}}^{(0)}(n,m|i,j)+\tilde{\mathcal{G}}_d(n,m|i,j)-s\tilde{\mathcal{G}}_x(n,m|i,j),\tag{D4}$$

where $\tilde{\mathcal{P}}^{(0)}(n,m|i,j)$ is given in Eq. (5.24) and

$$\tilde{\mathcal{G}}_d(n,m|i,j)=B^{0}(n,m|i,j)\delta^3(\mathbf{R}_m-\mathbf{R}_i)\delta^3(\mathbf{R}_n-\mathbf{R}_j)\times G_d(\mathbf{R}_i-\mathbf{R}_j)+(m\leftrightarrow n),\tag{D5}$$

$$\tilde{\mathcal{G}}_x(n,m|i,j)=B^{1}(n,m|i,j)\delta^3(\mathbf{R}_m-\mathbf{R}_i)\delta^3(\mathbf{R}_n-\mathbf{R}_j)\times G_x(\mathbf{R}_i-\mathbf{R}_j)+(m\leftrightarrow n).\tag{D6}$$

Here $G_d(\mathbf{R})$ and $G_x(\mathbf{R})$ are given in Eqs. (5.32) and (5.33), respectively.

[1] H. Haug and S.W. Koch, *Quantum Theory of the Optical and Electronic Properties of Semiconductors*, 3rd ed. (World Scientific, Singapore, 1994).
[2] B.F. Feuerbacher, J. Kuhl, and K. Ploog, Phys. Rev. B **43**, 2439 (1991).
[3] K.P. Pantke, D. Oberhauser, V.G. Lyssenko, J.M. Hvam, and G. Weimann, Phys. Rev. B **47**, 2413 (1993).
[4] T. Saiki, M. Kuwata-Gonokami, T. Matsusue, and H. Sakaki, Phys. Rev. B **49**, 7817 (1994).
[5] E.J. Mayer, G.O. Smith, V. Heuckeroth, J. Kuhl, K. Bott, A. Schulze, T. Meier, D. Bennhardt, S.W. Koch, P. Thomas, R. Hey, and K. Ploog, Phys. Rev. B **50**, 14 730 (1994).
[6] H. Wang, J. Shah, T.C. Damen, and L.N. Pfeiffer, Solid State Commun. **91**, 869 (1994).
[7] M. Kuwata-Gonokami, S. Inouye, H. Suzuura, M. Shirane, R. Shimano, T. Someya, and H. Sakai, Phys. Rev. Lett. **79**, 1341 (1997).
[8] T. Aoki, G. Mohs, M. Kuwata-Gonokami, and A.A. Yamaguchi, Phys. Rev. Lett. **82**, 3108 (1999).
[9] S. A. Moskalenko and D. W. Snoke, *Bose-Einstein Condensation of Excitons and Biexcitons* (Cambridge University Press, Cambridge, England, 2000).
[10] J.C. Kim and J.P. Wolfe, Phys. Rev. B **57**, 9861 (1998).
[11] M. Nagai, R. Shimano, and M. Kuwata-Gonokami, Phys. Rev. B **86**, 5795 (2001).
[12] H. Haug and S. Schmitt-Rink, Prog. Quantum Electron. **9**, 3 (1984).
[13] T.J. Inagaki, T. Iida, and M. Aihara, Phys. Rev. B **62**, 10 852 (2000).
[14] M. Lindberg and S.W. Koch, Phys. Rev. B **38**, 3342 (1988).
[15] V.M. Axt and A. Stahl, Z. Phys. B: Condens. Matter **93**, 195 (1994); **93**, 205 (1994).
[16] M. Lindberg, Y.Z. Hu, R. Binder, and S.W. Koch, Phys. Rev. B **50**, 18 060 (1994).
[17] Th. Östreich, K. Schönhammer, and L.J. Sham, Phys. Rev. B **58**, 12 920 (1998).
[18] H. Suzuura, Y.P. Svirko, and M. Kuwata-Gonokami, Solid State Commun. **108**, 289 (1998).
[19] H. Ishihara and K. Cho, Phys. Rev. B **53**, 15 823 (1996).
[20] O.M. Schmitt, D.B. Tran Thoai, L. Banyai, P. Gartner, and H. Haug, Phys. Rev. Lett. **86**, 3839 (2001).
[21] E. Hanamura, J. Phys. Soc. Jpn. **37**, 1545 (1974).
[22] T. Hiroshima, Phys. Rev. B **40**, 3862 (1989).
[23] A. Thilagam, Phys. Rev. B **63**, 045321 (2001).
[24] J. Inoue, T. Brandes, and A. Shimizu, J. Phys. Soc. Jpn. **67**, 3384 (1998).
[25] There are theoretical treatments where the exchange interaction between excitons was considered but deviations of excitons from ideal bosons were not taken into full account. These treatments are classified into the point-boson approximation in this paper.
[26] M. Hawton and D. Nelson, Phys. Rev. B **57**, 4000 (1998).
[27] C. Ciuti, V. Savona, C. Piermarocchi, A. Quattropani, and P. Schwendimann, Phys. Rev. B **58**, 7926 (1998).
[28] T. Kishimoto and T. Tamura, Phys. Rev. C **27**, 341 (1983).
[29] T. Ogawa and S. Okumura, Int. J. Mod. Phys. B (to be published).
[30] S. Okumura and T. Ogawa, J. Lumin. **87-89**, 238 (2000).
[31] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).
[32] T. Usui, Prog. Theor. Phys. **23**, 787 (1960).
[33] T. Marumori, M. Yamamura, and A. Tokunaga, Prog. Theor. Phys. **31**, 1009 (1964).
[34] M. Combescot and O. Betbeder-Matibet, cond-mat/0106346 (unpublished).
[35] E. Hanamura and H. Haug, Phys. Rep. **33**, 209 (1977).
[36] J. C. Slater, *Quantum Theory of Molecules and Solids* (McGraw-Hill, New York, 1963), Vol. 1.
[37] S. Okumura and T. Ogawa (unpublished).