Interacting excitons described by an infinite series of composite-exciton operators

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We revisit the approach proposed by Mukamel and co-workers to describe interacting excitons through infinite series of composite-boson operators for both the system Hamiltonian and the exciton commutator—which, in this approach, is properly kept different from its elementary-boson value. Instead of free-electron-hole operators, as used by Mukamel's group, we here work with composite-exciton operators which are physically relevant operators for excited semiconductors. This allows us to get *all* terms of these infinite series explicitly, the first terms of each series agreeing with the ones obtained by Mukamel's group when written with electron-hole pairs. All these terms nicely read in terms of Pauli and interaction scatterings of the composite-exciton many-body theory we have recently proposed. However, even if knowledge of these infinite series now allows us to tackle N-body problems, not just two-body problems like third-order nonlinear susceptibility $\chi^{(3)}$, the necessary handling of these two infinite series makes this approach far more complicated than the one we have developed and which barely relies on just four commutators.

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I. INTRODUCTION

Most particles known as bosons are composite particles made of even number of fermions. Proper treatment of the underlying Pauli exclusion principle between fermionic components of these particles has been a long-standing problem for decades.[1] Because many-body theories for quantum particles were, up to our work, valid for elementary particles only,[2] sophisticated "bosonization" procedures[3,4] have been proposed to replace composite bosons by elementary bosons. These elementary bosons then interact through effective scatterings constructed on interactions which exist between their fermionic components but dressed by "appropriate" fermion exchanges.[5] Although quite popular due to the fact that they allow calculations on problems otherwise unsolvable through known procedures, such bosonizations have an intrinsic major failure linked to the fact that, by replacing two free fermions by one boson, we strongly reduce degrees of freedom of the system. This shows up through the fact that, while closure relation for N elementary bosons is

$$\bar{I}=\frac{1}{N!}\sum \bar{B}^{\dagger}_{i_1}\cdots\bar{B}^{\dagger}_{i_N}|v\rangle\langle v|\bar{B}_{i_N}\cdots\bar{B}_{i_1}, \tag{1.1}$$

with $[\bar{B}_i,\bar{B}_j]=\delta_{ij}$, the one for N composite bosons made of two free fermions reads[6]

$$I=\frac{1}{(N!)^2}\sum B^{\dagger}_{i_1}\cdots B^{\dagger}_{i_N}|v\rangle\langle v|B_{i_N}\cdots B_{i_1}. \tag{1.2}$$

The huge prefactor change from $N!$ to $(N!)^2$ makes all sum rules for elementary and composite bosons, based on this closure relation, irretrievably different whatever effective scatterings are produced by bosonization procedures. Moreover indeed, through this prefactor difference in closure relations, we have explained[6] the factor 1/2 difference in the link between lifetime and sum of transition rates that we had found[7] for composite and bosonized excitons.

Besides bosonization, very few other approaches to interacting composite bosons have been proposed. In the late 1960s, Girardeau[8] suggested to introduce a set of "ideal-atom operators" in addition to fermionic operators for electrons and protons. These operators, which are bosonic by construction, represent all bound states of one atom but *not* its extended states. They are forced into the problem through a so-called Fock-Tani unitary transformation which, in an exact way, transforms *one* exact atom bound state into one ideal-atom state. Unfortunately, this nicely simple result does not hold for N-atom states with $N\geq 2$, the procedure turning quite complicated very fast. This is why, although not advocated by Girardeau,[8] we can be tempted by using his procedure as a bosonization procedure, i.e., by only keeping ideal-atom operators in transformed states and transformed Hamiltonian. We have however shown[9] that, with such a reduction, the obtained results for a few relevant physical quantities are at odd from the correct ones and even for the sign. The idea to add to fermionic operators for electrons and protons, a set of bosonic operators for atomic bound states, is, in fact, rather awkward because fermionic operators form a complete set in themselves so that Girardeau[8] artificially introduces an overcomplete set of operators in a problem already complex, this overcompleteness being obviously difficult to handle properly. Precise comparison of Girardeau's procedure[8] with the composite-boson many-body theory we have constructed can be found in Ref. 9.

Another approach, still currently used, has been proposed by Mukamel and co-workers[10–12] in the 1990s. It is based on the fully correct idea that the system Hamiltonian, when acting on fermion pairs, can be replaced by an infinite series of pair operators. In this approach, the fact that pairs of fermions differ from elementary bosons is kept exactly through commutators of pair operators which are also written as infinite series. The pair operators used by Mukamel and co-workers[10–12] are products of free fermion operators. However, as these are not physically relevant pair operators for problems dealing with excitons, their calculations turn out very complicated. This is probably why they have only derived the first term of the Hamiltonian and pair-commutator series. This thus makes their results of possible use for problems restricted to two excitons only. Moreover indeed, using them, they have successfully calculated[12] the third-order susceptibility $\chi^{(3)}$ which results from interactions of two unabsorbed photons through their coupling to two virtual excitons.

In this paper, we follow the idea of Mukamel and co-workers[10–12] but with exciton operators $B_i^\dagger$ instead of products of free-electron and free-hole operators $a_{\mathbf{k}_e}^\dagger b_{\mathbf{k}_h}^\dagger$, these exciton operators being the ones which create one-electron-hole-pair eigenstates of the system Hamiltonian,

$$(H-E_i)B_i^\dagger|v\rangle = 0. \tag{1.3}$$

Thanks to the closure relation for N composite excitons [Eq. (1.2)], it is easy to derive all terms of the series for the composite-boson commutator and for the system Hamiltonian in an exact way. As expected, prefactors in these infinite series read in terms of the two key parameters of the composite-boson many-body physics, namely, Pauli scatterings for fermion exchanges in the absence of fermion interaction and interaction scatterings for fermion interactions in the absence of fermion exchange.

However, even with these two infinite series at hand explicitly, so that problems dealing with many-body effects between N excitons could now be tackled, this approach turns out to be definitely far more complicated than the composite-boson many-body theory we have recently constructed.[13,14] Indeed, in this theory, calculations dealing with many-body effects between any number N of excitons simply reduce to performing a set of commutations between exciton operators, according to two commutators for fermion exchanges [see Eq. (5) of Ref. 15 or Eq. (14) of Ref. 13], namely,

$$[B_m, B_i^{\dagger N}] = NB_i^{\dagger N-1}(\delta_{m,i} - D_{mi}) - N(N-1)B_i^{\dagger N-2}\sum_n \lambda\begin{pmatrix} n & i \\ m & i \end{pmatrix} B_n^\dagger, \tag{1.4}$$

$$[D_{mi}, B_j^{\dagger N}] = NB_j^{\dagger N-1}\sum_n \left\{ \lambda\begin{pmatrix} n & j \\ m & i \end{pmatrix} + \lambda\begin{pmatrix} m & j \\ n & i \end{pmatrix} \right\} B_n^\dagger, \tag{1.5}$$

and two commutators for fermion interactions [see Eq. (5) of Ref. 16 or Eq. (13) of Ref. 13], namely,

$$[H, B_i^{\dagger N}] = NB_i^{\dagger N-1}(E_i B_i^\dagger + V_i^\dagger) + \frac{N(N-1)}{2} B_i^{\dagger N-1}\sum_{m,n} \xi\begin{pmatrix} n & i \\ m & i \end{pmatrix} B_m^\dagger B_n^\dagger, \tag{1.6}$$

$$[V_i^\dagger, B_j^{\dagger N}] = NB_j^{\dagger N-1}\sum_{m,n} \xi\begin{pmatrix} n & j \\ m & i \end{pmatrix} B_m^\dagger B_n^\dagger. \tag{1.7}$$

In these equations, D_{mi} is the exciton "deviation-from-boson operator" defined through Eq. (1.4) taken for $N=1$, namely,

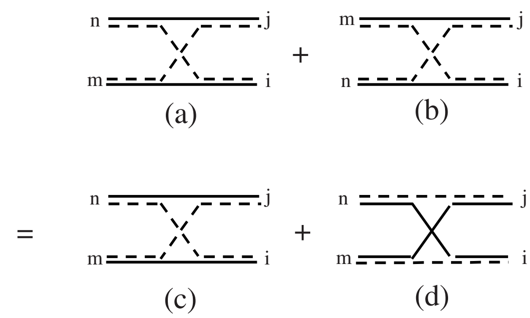

FIG. 1. Shiva diagrams for $\Lambda_{mi}(n,j)$ defined in Eq. (1.12). $\lambda(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix})$, represented by diagram (a), is identical to $\lambda_e(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix})$, represented by diagram (c), in which m and i have the same electron. $\lambda(\begin{smallmatrix} m & j \\ n & i \end{smallmatrix})$, represented by diagram (b), is identical to $\lambda_h(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix})$, represented by diagram (d), in which m and i have the same hole.

$$D_{mi} = \delta_{m,i} - [B_m, B_i^\dagger], \tag{1.8}$$

while Pauli scattering $\lambda(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix})$ of two "in" excitons (i,j) toward two "out" excitons (m,n) follows from Eq. (1.5) taken for $N=1$ [also see Eq. (4) of Ref. 17], namely,

$$[D_{mi}, B_j^\dagger] = \sum_n \left\{ \lambda\begin{pmatrix} n & j \\ m & i \end{pmatrix} + \lambda\begin{pmatrix} m & j \\ n & i \end{pmatrix} \right\} B_n^\dagger. \tag{1.9}$$

In the same way, "creation potential" $V_i^\dagger$ of exciton i and interaction scattering $\xi(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix})$ follow from Eqs. (1.6) and (1.7) taken for $N=1$ [also see Eq. (3) of Ref. 17], namely,

$$[H, B_i^\dagger] = E_i B_i^\dagger + V_i^\dagger, \tag{1.10}$$

$$[V_i^\dagger, B_j^\dagger] = \sum_{m,n} \xi\begin{pmatrix} n & i \\ m & i \end{pmatrix} B_m^\dagger B_n^\dagger. \tag{1.11}$$

Let us note that Eq. (1.8), which basically says that particles are not elementary but composite bosons, was known for quite a long time.[1,18] Equation (1.10) is more recent. It was introduced by one of us in her theory of exciton optical Stark effect.[19,20] On the contrary, Eqs. (1.9) and (1.11) which allow us to reach the two elementary scatterings of two excitons, namely, $\lambda(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix})$ and $\xi(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix})$, are fundamentally different. They are the keys of our composite-boson many-body theory.[17]

In this paper, we are going to use these four commutators to write the system Hamiltonian H and the deviation-from-boson operator D_{mi} as infinite series of exciton operators $B_i^\dagger$. This will allow us to generate physically relevant prefactors for these series. They are found to read in terms of exciton energies E_i, interaction scattering of two excitons $\xi(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix})$, and the following sum of Pauli scatterings:

$$\Lambda_{mi}(n,j) = \lambda\begin{pmatrix} n & j \\ m & i \end{pmatrix} + \lambda\begin{pmatrix} m & j \\ n & i \end{pmatrix} = \lambda_e\begin{pmatrix} n & j \\ m & i \end{pmatrix} + \lambda_h\begin{pmatrix} n & j \\ m & i \end{pmatrix}. \tag{1.12}$$

$\Lambda_{mi}(n,j)$, shown in Fig. 1, corresponds to processes in which excitons (i,j) exchange either a hole or an electron, excitons (m,i) having same electron in $\lambda_e \equiv \lambda$, while they have the same hole in λ_h. Due to electron-hole symmetry, it is quite reasonable to find these two processes on the same footing, in the $\Lambda_{mi}(n,j)$ factor.

II. DEVIATION-FROM-BOSON OPERATOR

Let us start with deviation-from-boson operator D_{mi} defined in Eq. (1.8). Since both D_{mi} and the product of exciton operators $B_m^\dagger B_i$ conserve number of pairs, we can look for D_{mi} as

$$D_{mi}=\sum_{n=1}^{\infty} D_{mi}^{(n)}, \tag{2.1}$$

where the most general form for $D_{mi}^{(n)}$ acting in the subspace made of states having $p \geq n$ pairs can be taken as

$$D_{mi}^{(n)}=\sum_{\{r\}} d_{mi}^{(n)}(r_1',\ldots,r_n';r_1,\ldots,r_n)B_{r_1'}^\dagger\cdots B_{r_n'}^\dagger B_{r_1}\cdots B_{r_n}. \tag{2.2}$$

We get this series by enforcing it to be such that, when acting on any N-exciton state $|\psi_N\rangle$ linear combination of $B_{j_1}^\dagger\cdots B_{j_N}^\dagger|v\rangle$, it gives the same result as the original operator D_{mi}, namely,

$$D_{mi}|\psi_N\rangle=\sum_{n=1}^{N} D_{mi}^{(n)}|\psi_N\rangle, \tag{2.3}$$

for any N-pair state $|\psi_N\rangle$. We are going to derive the various operators $D_{mi}^{(n)}$ by iteration, starting from $n=1$, as we now show.

Before going further, it is of importance to note that, due to carrier exchanges between two excitons, we do have [see Eq. (5) of Ref. 17]

$$B_i^\dagger B_j^\dagger=-\sum_{m,n}\lambda\begin{pmatrix} n & j\\ m & i\end{pmatrix}B_m^\dagger B_n^\dagger. \tag{2.4}$$

This equation, which comes from the two ways to construct two excitons out of the two electron-hole pairs, shows that N-exciton states $|\psi_N\rangle$ for $N\geq 2$, as well as operators like $D_{mi}^{(n)}$ for $n\geq 2$, can be written in many different ways, these various forms being related through the replacement of any $B^\dagger B^\dagger$ by a sum of $B^\dagger B^\dagger$ according to Eq. (2.4). Just as $B_{i_1}^\dagger\cdots B_{i_N}^\dagger|v\rangle$ states form an overcomplete set for N-pair states, $B_i^\dagger$'s form an overcomplete set of operators. This, in particular, allows us to guess that, among the various possible forms of $D_{mi}^{(n)}$, the one which has a physically relevant meaning is most probably the simplest one. We will come back to this fundamental indetermination, linked to exciton composite nature, at the end of this section.

A. Derivation of $D_{mi}^{(1)}$

Let us first consider a one-exciton state $|\psi_1\rangle$. By inserting closure relation for one-exciton subspace, i.e., Eq. (1.2) taken for $N=1$, in front of this state, we find

$$D_{mi}|\psi_1\rangle=\sum_{r_1} D_{mi}B_{r_1}^\dagger|v\rangle\langle v|B_{r_1}|\psi_1\rangle. \tag{2.5}$$

As $D_{mi}B_{r_1}^\dagger|v\rangle=[D_{mi},B_{r_1}^\dagger]|v\rangle$ since $D_{mi}|v\rangle=0$ which follows from Eq. (1.8) acting on vacuum, we get from Eqs. (1.9) and (1.12)

$$D_{mi}|\psi_1\rangle=\sum_{r_1',r_1}\Lambda_{mi}(r_1',r_1)B_{r_1'}^\dagger|v\rangle\langle v|B_{r_1}|\psi_1\rangle, \tag{2.6}$$

where $\Lambda_{mi}(n,j)$ is the combination of Pauli scatterings introduced in Eq. (1.12).

We then note that projector $|v\rangle\langle v|$ can be removed from this equation since state $B_{r_1}|\psi_1\rangle$ has zero pair while identity operator reduces to $|v\rangle\langle v|$ for such a state. Consequently, Eq. (2.6) also reads

$$D_{mi}|\psi_1\rangle=\sum_{r_1',r_1}\Lambda_{mi}(r_1',r_1)B_{r_1'}^\dagger B_{r_1}|\psi_1\rangle. \tag{2.7}$$

Since this equation is valid for any state $|\psi_1\rangle$, we readily find that operator $D_{mi}^{(1)}$ such that $D_{mi}|\psi_1\rangle=D_{mi}^{(1)}|\psi_1\rangle$ can be taken as

$$D_{mi}^{(1)}=\sum_{r_1',r_1}\Lambda_{mi}(r_1',r_1)B_{r_1'}^\dagger B_{r_1}, \tag{2.8}$$

with $\Lambda_{mi}(r_1',r_1)$ given in Eq. (1.12). This result is the same as the one given by Mukamel and co-workers[12] [see Eqs. (11)–(13) of Ref. 12].

B. Derivation of $D_{mi}^{(2)}$

We now consider two-exciton state $|\psi_2\rangle$. By inserting closure relation [Eq. (1.2)] for two-exciton subspace, in front of $|\psi_2\rangle$, we get

$$D_{mi}|\psi_2\rangle=\left(\frac{1}{2!}\right)^2\sum_{r_1,r_2} D_{mi}B_{r_1}^\dagger B_{r_2}^\dagger|v\rangle\langle v|B_{r_2}B_{r_1}|\psi_2\rangle. \tag{2.9}$$

To go further, we note that, due to Eqs. (1.9) and (1.12),

$$\begin{aligned} D_{mi}B_{r_1}^\dagger B_{r_2}^\dagger|v\rangle &= ([D_{mi},B_{r_1}^\dagger]+B_{r_1}^\dagger D_{mi})B_{r_2}^\dagger|v\rangle\\ &=\sum_{r'} B_{r'}^\dagger[\Lambda_{mi}(r',r_1)B_{r_2}^\dagger+\Lambda_{mi}(r',r_2)B_{r_1}^\dagger]|v\rangle. \end{aligned} \tag{2.10}$$

We then insert this result into Eq. (2.9) and relabel bold indices. By noting that projector $|v\rangle\langle v|$ can again be removed from this equation since $B_{r_2}B_{r_1}|\psi_2\rangle$ also has zero pair, we end with

$$D_{mi}|\psi_2\rangle=\frac{2}{(2!)^2}\sum_{r_1',r_1,r_2}\Lambda_{mi}(r_1',r_1)B_{r_1'}^\dagger B_{r_2}^\dagger B_{r_2}B_{r_1}|\psi_2\rangle. \tag{2.11}$$

If we now turn to $D_{mi}^{(1)}$ acting on $|\psi_2\rangle$, we note that $B_{r_1}|\psi_2\rangle$ has one pair so that, if we insert closure relation for one-pair subspace in front of this state, we find

$$D_{mi}^{(1)}|\psi_2\rangle = \sum_{r_1',r_1} \Lambda_{mi}(r_1',r_1) B_{r_1'}^{\dagger}\left[\sum_{r_2} B_{r_2}^{\dagger}|v\rangle\langle v|B_{r_2}\right] B_{r_1}|\psi_2\rangle. \tag{2.12}$$

We can again remove projector $|v\rangle\langle v|$ from this equation since $B_{r_2}B_{r_1}|\psi_2\rangle$ has zero pair. This readily shows that operator $D_{mi}^{(2)}$ such that $D_{mi}|\psi_2\rangle=(D_{mi}^{(1)}+D_{mi}^{(2)})|\psi_2\rangle$ can be taken as

$$D_{mi}^{(2)} = -\frac{1}{2}\sum_{r_1',r_1,r_2} \Lambda_{mi}(r_1',r_1) B_{r_1'}^{\dagger} B_{r_2}^{\dagger} B_{r_2} B_{r_1}. \tag{2.13}$$

C. Derivation of $D_{mi}^{(3)}$

To better grasp how $D_{mi}^{(n)}$ can be constructed by iteration, let us calculate one more $D_{mi}^{(n)}$ explicitly. We consider three-pair state $|\psi_3\rangle$ and inject in front of it, closure relation for three-pair subspace. This leads to

$$D_{mi}|\psi_3\rangle = \frac{1}{(3!)^2}\sum_{r_1,r_2,r_3} D_{mi} B_{r_1}^{\dagger} B_{r_2}^{\dagger} B_{r_3}^{\dagger}|v\rangle\langle v|B_{r_3}B_{r_2}B_{r_1}|\psi_3\rangle. \tag{2.14}$$

To go further, we do like for Eq. (2.10) and use commutator $[D_{mi},B_r^{\dagger}]$ given in Eq. (1.9). This leads to

$$D_{mi}B_{r_1}^{\dagger}B_{r_2}^{\dagger}B_{r_3}^{\dagger}|v\rangle = \sum_{r'}\{\Lambda_{mi}(r',r_1)B_{r_2}^{\dagger}B_{r_3}^{\dagger} + \Lambda_{mi}(r',r_2)B_{r_1}^{\dagger}B_{r_3}^{\dagger} + \Lambda_{mi}(r',r_3)B_{r_1}^{\dagger}B_{r_2}^{\dagger}\}|v\rangle. \tag{2.15}$$

We then inject this result into Eq. (2.14), relabel bold indices, and remove projector $|v\rangle\langle v|$. This leads to

$$D_{mi}|\psi_3\rangle = \frac{3}{(3!)^2}\sum_{r_1',\{r\}} \Lambda_{mi}(r_1',r_1) B_{r_1'}^{\dagger}B_{r_2}^{\dagger}B_{r_3}^{\dagger}B_{r_3}B_{r_2}B_{r_1}|\psi_3\rangle. \tag{2.16}$$

We now turn to $D_{mi}^{(1)}|\psi_3\rangle$. Since $B_{r_1}|\psi_3\rangle$ has two pairs, we get, by using closure relation for two-pair subspace,

$$D_{mi}^{(1)}|\psi_3\rangle = \left(\frac{1}{2!}\right)^2 \sum_{r_1',\{r\}} \Lambda_{mi}(r_1',r_1)\; B_{r_1'}^{\dagger}B_{r_2}^{\dagger}B_{r_3}^{\dagger}\rangle\langle v|B_{r_3}B_{r_2}B_{r_1}|\psi_3\rangle. \tag{2.17}$$

We do the same for $D_{mi}^{(2)}|\psi_3\rangle$ in which $B_{r_1}B_{r_2}|\psi_3\rangle$ has one pair. By collecting all terms, we see that operator $D_{mi}^{(3)}$ such that $D_{mi}|\psi_3\rangle=(D_{mi}^{(1)}+D_{mi}^{(2)}+D_{mi}^{(3)})|\psi_3\rangle$ can be taken as

$$D_{mi}^{(3)} = \frac{1}{3}\sum_{r_1',\{r\}} \Lambda_{mi}(r_1',r_1) B_{r_1'}^{\dagger}B_{r_2}^{\dagger}B_{r_3}^{\dagger}B_{r_3}B_{r_2}B_{r_1}. \tag{2.18}$$

D. Derivation of $D_{mi}^{(n)}$

The above results lead us to think that operator $D_{mi}^{(n)}$ can be written as

$$D_{mi}^{(n)} = \gamma_n \sum_{r_1',\{r\}} \Lambda_{mi}(r_1',r_1) B_{r_1'}^{\dagger}B_{r_2}^{\dagger}\cdots B_{r_n}^{\dagger}B_{r_n}\cdots B_{r_2}B_{r_1}, \tag{2.19}$$

where γ_n is a numerical prefactor which, in spite of its values for $n=(1,2,3)$, does not reduce to $(-1)^{n-1}/n$.

To determine γ_n, we look for the recursion relation it obeys. To get this recursion relation, we follow the procedure we have used for $n\leq 3$, namely, we insert closure relation for N-pair subspace in front of state $|\psi_N\rangle$. This leads to

$$D_{mi}|\psi_N\rangle = \left(\frac{1}{N!}\right)^2 \sum_{\{r\}} D_{mi}B_{r_1}^{\dagger}\cdots B_{r_N}^{\dagger}|v\rangle\langle v|B_{r_N}\cdots B_{r_1}|\psi_N\rangle. \tag{2.20}$$

We then calculate $D_{mi}B_{r_1}^{\dagger}\cdots B_{r_N}^{\dagger}|v\rangle$ using commutator (1.9); we relabel bold indices and remove projector $|v\rangle\langle v|$. This gives

$$D_{mi}|\psi_N\rangle = \frac{N}{(N!)^2}\sum_{r_1',\{r\}} \Lambda_{mi}(r_1',r_1) B_{r_1'}^{\dagger}B_{r_2}^{\dagger}\cdots B_{r_N}^{\dagger}B_{r_N}\cdots B_{r_2}B_{r_1}|\psi_N\rangle. \tag{2.21}$$

We then turn to $D_{mi}^{(n)}$ acting on $|\psi_N\rangle$ for $n<N$. Since state $B_{r_1}\cdots B_{r_n}|\psi_N\rangle$ has $(N-n)$ pairs, closure relation for this subspace leads to

$$D_{mi}^{(n)}|\psi_N\rangle = \left(\frac{1}{(N-n)!}\right)^2 \gamma_n \sum_{r_1',\{r\}} \Lambda_{mi}(r_1',r_1) B_{r_1'}^{\dagger}B_{r_2}^{\dagger}\cdots B_{r_n}^{\dagger} \times B_{r_{n+1}}^{\dagger}\cdots B_{r_N}^{\dagger}B_{r_N}\cdots B_{r_{n+1}}B_{r_n}\cdots B_{r_1}|\psi_N\rangle. \tag{2.22}$$

By inserting these results into Eq. (2.3), it is easy to show that the form [Eq. (2.19)] for $D_{mi}^{(n)}$ is indeed valid provided that γ_n's are linked by

$$\gamma_N = \frac{N}{(N!)^2} - \sum_{n=1}^{N-1}\frac{\gamma_n}{[(N-n)!]^2}, \tag{2.23}$$

with $\gamma_1=1$. From this equation, it is easy to show that the first γ_n's are

$$\gamma_2 = -1/2,$$

$$\gamma_3 = 1/3,$$

$$\gamma_4 = -11/48,$$

$$\gamma_5 = 11/120, \tag{2.24}$$

and so on, with γ_N going to zero with increasing N.

E. Other forms of $D_{mi}^{(n)}$

As said at the beginning of this section, composite-boson operators $B_i^{\dagger}$ form an overcomplete set to describe electron-

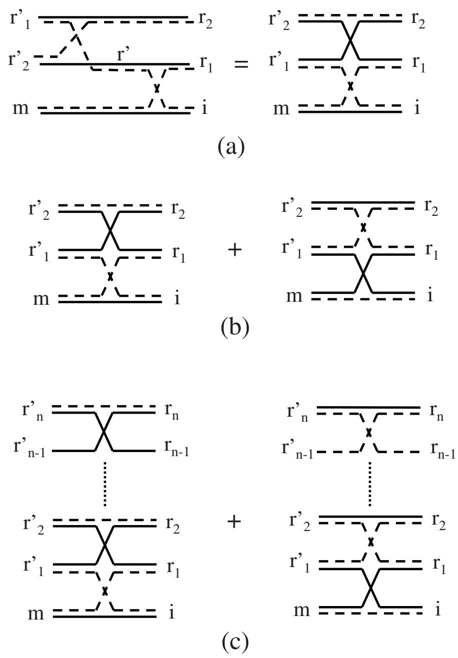

FIG. 2. (a) Shiva diagram representation of Eq. (2.27), the summation over the bold index r' being performed readily. (b) Shiva diagrams for the prefactor $\Lambda_{mi}(\begin{smallmatrix} r_2' & r_2 \\ r_1' & r_1 \end{smallmatrix})$ appearing in Eq. (2.28). This prefactor corresponds to carrier exchanges between (i,r_1,r_2) leading to (m,r_1',r_2'), in which the excitons m and i either have the same electron or the same hole. (c) Same as (b) for the prefactor appearing in Eq. (2.29).

hole pairs. This is why any given operator acting in N-pair subspace with $N\geq 2$, when written in terms of these $B_i^\dagger$'s, can appear through different expressions. Indeed, due to Eq. (2.4), it is possible to rewrite $B_{r_1'}^\dagger B_{r_2}^\dagger$ in Eq. (2.19) as

$$B_{r_1'}^\dagger B_{r_2}^\dagger = -\sum_{r_1'',r_2'} \lambda \begin{pmatrix} r_1'' & r_2 \\ r_2' & r_1' \end{pmatrix} B_{r_1''}^\dagger B_{r_2'}^\dagger, \tag{2.25}$$

since $B_m^\dagger B_n^\dagger = B_n^\dagger B_m^\dagger$. We then note that

$$\sum_{r_1'} \lambda \begin{pmatrix} r_1'' & r_2 \\ r_2' & r_1' \end{pmatrix} \lambda \begin{pmatrix} r_1' & r_1 \\ m & i \end{pmatrix} = \lambda_3 \begin{pmatrix} r_2' & r_2 \\ r_1'' & r_1 \\ m & i \end{pmatrix}, \tag{2.26}$$

where, according to Fig. 2(a), λ_3 is just the exchange scattering between three excitons (i,r_1,r_2). This allows us to replace the first factor of $D_{mi}^{(n)}$ in Eq. (2.19) by

$$\sum_{r_1'} \Lambda_{mi}(r_1',r_1) B_{r_1'}^\dagger B_{r_2}^\dagger = -\sum_{r_1'',r_2'} \Lambda_{mi} \begin{pmatrix} r_2' & r_2 \\ r_1'' & r_1 \end{pmatrix} B_{r_1''}^\dagger B_{r_2'}^\dagger. \tag{2.27}$$

While prefactor $\Lambda_{mi}(r_1',r_1)$ corresponds to carrier exchanges between two excitons (i,r_1) leading to (m,r_1') with excitons m and i having either same electron or same hole, prefactor $\Lambda_{mi}(\begin{smallmatrix} r_2' & r_2 \\ r_1' & r_1 \end{smallmatrix})$ corresponds to carrier exchanges between excitons (i,r_1,r_2) leading to (m,r_1'',r_2'), with excitons m and i also having either same electron or same hole [see Fig. 2(b)].

If we keep doing this procedure for $B_{r_2'}^\dagger B_{r_3}^\dagger$ with $B_{r_2'}^\dagger$ relabeled as $B_{r_1'}^\dagger$ and so on, we end with $D_{mi}^{(n)}$ written in a quite symmetrical form, although far more complicated than Eq. (2.19), namely,

$$D_{mi}^{(n)} = (-1)^{n-1}\gamma_n \sum_{\{r'\},\{r\}} \Lambda_{mi} \begin{pmatrix} r_n' & r_n \\ \cdot & \cdot \\ \cdot & \cdot \\ r_1' & r_1 \end{pmatrix} B_{r_1'}^\dagger \cdots B_{r_n'}^\dagger B_{r_n} \cdots B_{r_1}, \tag{2.28}$$

where the prefactor corresponds to carrier exchanges between $(n+1)$ excitons $(i,r_1,\ldots,r_n)$ leading to $(m,r_1',\ldots,r_n')$ in which excitons m and i either have same electron or same hole [see Fig. 2(c)].

III. SYSTEM HAMILTONIAN

Let us now turn to the system Hamiltonian originally written in terms of fermionic operators for free electrons and free holes. It contains kinetic electron and hole contributions. It also contains Coulomb interaction between electrons, between holes, and between electrons and holes. It is actually quite easy to write the electron-hole part of this Hamiltonian in terms of excitons. Indeed, by using the link between exciton operators and free-electron and free-hole operators, namely,

$$B_i^\dagger = \sum_{\mathbf{k}_e,\mathbf{k}_h} a_{\mathbf{k}_e}^\dagger b_{\mathbf{k}_h}^\dagger \langle \mathbf{k}_h,\mathbf{k}_e|i\rangle, \tag{3.1}$$

$$a_{\mathbf{k}_e}^\dagger b_{\mathbf{k}_h}^\dagger = \sum_i B_i^\dagger \langle i|\mathbf{k}_e,\mathbf{k}_h\rangle, \tag{3.2}$$

where $\langle \mathbf{k}_h,\mathbf{k}_e|i\rangle$ is i exciton wave function in momentum space, we readily find electron-hole Coulomb interaction as

$$\begin{aligned} V_{eh} &= -\sum_{\mathbf{q},\mathbf{k}_e,\mathbf{k}_h} V_{\mathbf{q}} a_{\mathbf{k}_e+\mathbf{q}}^\dagger b_{\mathbf{k}_h-\mathbf{q}}^\dagger b_{\mathbf{k}_h} a_{\mathbf{k}_e} \\ &= -\sum_{i,j} B_i^\dagger B_j \sum_{\mathbf{q},\mathbf{k}_e,\mathbf{k}_h} V_{\mathbf{q}} \langle i|\mathbf{k}_e+\mathbf{q},\mathbf{k}_h-\mathbf{q}\rangle\langle \mathbf{k}_h,\mathbf{k}_e|j\rangle. \end{aligned} \tag{3.3}$$

On the contrary, this cannot be done for other parts of the Hamiltonian, namely, kinetic-energy terms in $a^\dagger a$ and $b^\dagger b$ and electron-electron and hole-hole Coulomb terms in $a^\dagger a^\dagger aa$ and $b^\dagger b^\dagger bb$. Nevertheless, since both operator H and product of exciton operators $B_m^\dagger B_i$ conserve the number of electron-hole pairs, it is *a priori* possible to write H as

$$H = \sum_{n=1}^{\infty} H^{(n)}, \tag{3.4}$$

$$H^{(n)} = \sum_{\{r\},\{r'\}} h^{(n)}(r_1',\ldots,r_n';r_1,\ldots,r_n) B_{r_1'}^\dagger \cdots B_{r_n'}^\dagger B_{r_n} \cdots B_{r_1}, \tag{3.5}$$

so that $H^{(n)}$ acts on states having $p\geq n$ pairs. This series is determined by enforcing

$$H|\psi_N\rangle = \sum_{n=1}^{N} H^{(n)}|\psi_N\rangle \tag{3.6}$$

for any state $|\psi_N\rangle$ having N electron-hole pairs. Here again, $H^{(n)}$ for $n\geq 2$ is expected to have various forms since any $B^\dagger B^\dagger$ can be replaced by sum of $B^\dagger B^\dagger$, according to Eq. (2.4). To get the various terms of $H^{(n)}$ expansion, we are again going to extensively use closure relation (1.2) for N-pair states. This will allow us to get one of these possible forms of H quite easily.

A. Derivation of $H^{(1)}$

To get $H^{(1)}$, we insert closure relation for one-pair states in front of $|\psi_1\rangle$ in $H|\psi_1\rangle$. This leads to

$$H|\psi_1\rangle = \sum_{r_1} HB^\dagger_{r_1}|v\rangle\langle v|B_{r_1}|\psi_1\rangle. \tag{3.7}$$

We first replace $HB^\dagger_{r_1}|v\rangle$ by $E_{r_1}B^\dagger_{r_1}|v\rangle$ as exciton operators create one-pair eigenstates of the system. We then note that $B_{r_1}|\psi_1\rangle$ has zero pair, so that we can remove projector $|v\rangle\langle v|$ from this equation. This leads to

$$H|\psi_1\rangle = \sum_{r_1} E_{r_1}B^\dagger_{r_1}B_{r_1}|\psi_1\rangle. \tag{3.8}$$

Since $H^{(1)}|\psi_1\rangle$ must be equal to $H|\psi_1\rangle$ for any one-pair state $|\psi_1\rangle$, we readily see that $H^{(1)}$ can be identified with

$$H^{(1)} = \sum_{r_1} E_{r_1}B^\dagger_{r_1}B_{r_1}. \tag{3.9}$$

B. Derivation of $H^{(2)}$

We now turn to two-pair subspace. By inserting closure relation for two-pair states in front of $|\psi_2\rangle$, we find

$$H|\psi_2\rangle = \left(\frac{1}{2!}\right)^2 \sum_{r_1,r_2} HB^\dagger_{r_1}B^\dagger_{r_2}|v\rangle\langle v|B_{r_2}B_{r_1}|\psi_2\rangle. \tag{3.10}$$

We then use Eqs. (1.10) and (1.11) to find

$$\begin{aligned}HB^\dagger_{r_1}B^\dagger_{r_2}|v\rangle &= (B^\dagger_{r_1}H + E_{r_1}B^\dagger_{r_1} + V^\dagger_{r_1})B^\dagger_{r_2}|v\rangle \\ &= (E_{r_1}+E_{r_2})B^\dagger_{r_1}B^\dagger_{r_2}|v\rangle + \sum_{r'_1,r'_2} \xi\begin{pmatrix} r'_2 & r_2 \\ r'_1 & r_1\end{pmatrix} B^\dagger_{r'_1}B^\dagger_{r'_2}|v\rangle.\end{aligned} \tag{3.11}$$

If we insert this result into Eq. (3.10), relabel bold indices, and remove projector $|v\rangle\langle v|$, we end with

$$H|\psi_2\rangle = \left(\frac{1}{2}\sum_{r_1,r_2} E_{r_1}B^\dagger_{r_1}B^\dagger_{r_2}B_{r_2}B_{r_1} + \frac{1}{4}\sum_{r_1,r_2,r'_1,r'_2} \xi\begin{pmatrix} r'_2 & r_2 \\ r'_1 & r_1\end{pmatrix} B^\dagger_{r'_1}B^\dagger_{r'_2}B_{r_2}B_{r_1}\right)|\psi_2\rangle. \tag{3.12}$$

Let us now turn to $H^{(1)}$ acting on $|\psi_1\rangle$. We first note that $B_{r_1}|\psi_2\rangle$ has one pair so that closure relation for one-pair subspace leads to

$$H^{(1)}|\psi_1\rangle = \sum_{r_1,r_2} E_{r_1}B^\dagger_{r_1}B^\dagger_{r_2}|v\rangle\langle v|B_{r_2}B_{r_1}|\psi_1\rangle, \tag{3.13}$$

in which we can remove projector $|v\rangle\langle v|$ since $B_{r_2}B_{r_1}|\psi_1\rangle$ has zero pair. This readily shows that $H^{(2)}$, such that $H\psi_2\rangle = (H^{(1)}+H^{(2)})|\psi_2\rangle$, can be identified with

$$H^{(2)} = -\frac{1}{2}\sum_{r_1,r_2} E_{r_1}B^\dagger_{r_1}B^\dagger_{r_2}B_{r_2}B_{r_1} + \frac{1}{4}\sum_{r'_1,r'_2,r_1,r_2} \xi\begin{pmatrix} r'_2 & r_2 \\ r'_1 & r_1\end{pmatrix} B^\dagger_{r'_1}B^\dagger_{r'_2}B_{r_2}B_{r_1}. \tag{3.14}$$

C. Derivation of $H^{(3)}$

To grasp how series H is constructed, let us calculate one more $H^{(n)}$ explicitly. From closure relation for three-pair states, we find

$$H|\psi_3\rangle = \left(\frac{1}{3!}\right)^2 \sum_{\{r\}} HB^\dagger_{r_1}B^\dagger_{r_2}B^\dagger_{r_3}|v\rangle\langle v|B_{r_3}B_{r_2}B_{r_1}|\psi_3\rangle. \tag{3.15}$$

We then use Eqs. (1.10) and (1.11) to find

$$\begin{aligned}HB^\dagger_{r_1}B^\dagger_{r_2}B^\dagger_{r_3}|v\rangle &= (E_{r_1}+E_{r_2}+E_{r_3})B^\dagger_{r_1}B^\dagger_{r_2}B^\dagger_{r_3}|v\rangle \\ &+ \sum_{s,t} B^\dagger_s B^\dagger_t\left[\xi\begin{pmatrix} t & r_2 \\ s & r_1\end{pmatrix}B^\dagger_{r_3} + \xi\begin{pmatrix} t & r_3 \\ s & r_2\end{pmatrix}B^\dagger_{r_1} \right.\\ &\left.+ \xi\begin{pmatrix} t & r_1 \\ s & r_3\end{pmatrix}B^\dagger_{r_2}\right],\end{aligned} \tag{3.16}$$

so that, if we insert this result into Eq. (3.15), relabel bold indices, and remove projector $|v\rangle\langle v|$, we end with

$$H|\psi_3\rangle = \left[\frac{3}{(3!)^2}\sum_{\{r\}} E_{r_1}B^\dagger_{r_1}B^\dagger_{r_2}B^\dagger_{r_3}B_{r_3}B_{r_2}B_{r_1} + \frac{C_3^2}{(3!)^2}\sum_{r'_1,r'_2,\{r\}} \xi\begin{pmatrix} r'_2 & r_2 \\ r'_1 & r_1\end{pmatrix} B^\dagger_{r'_1}B^\dagger_{r'_2}B^\dagger_{r_3}B_{r_3}B_{r_2}B_{r_1}\right]|\psi_3\rangle, \tag{3.17}$$

where $C_N^2 = N(N-1)/2$ is the number of ways we can choose two excitons among N. This makes $C_3^2=3$.

We now turn to $(H^{(1)}+H^{(2)})|\psi_3\rangle$ that we calculate by injecting closure relations for two-pair states in front of $B_{r_1}|\psi_3\rangle$ and for one-pair states in front of $B_{r_1}B_{r_2}|\psi_3\rangle$. By collecting all these results, we find that $H^{(3)}$ such that $H|\psi_3\rangle = (H^{(1)}+H^{(2)}+H^{(3)})|\psi_3\rangle$ can be identified with

$$H^{(3)} = \frac{1}{3}\sum_{\{r\}} E_{r_1}B^\dagger_{r_1}B^\dagger_{r_2}B^\dagger_{r_3}B_{r_3}B_{r_2}B_{r_1} - \frac{1}{6}\sum_{r'_1,r'_2,\{r\}} \xi\begin{pmatrix} r'_2 & r_2 \\ r'_1 & r_1\end{pmatrix} B^\dagger_{r'_1}B^\dagger_{r'_2}B^\dagger_{r_3}B_{r_3}B_{r_2}B_{r_1}. \tag{3.18}$$

D. Derivation of $H^{(n)}$

The above results lead us to think that operator $H^{(n)}$ can be written as

$$H^{(n)} = \alpha_n \sum_{\{r\}} E_{r_1} B^\dagger_{r_1} \cdots B^\dagger_{r_n} B_{r_n} \cdots B_{r_1} + \beta_n \sum_{r'_1, r'_2, \{r\}} \xi\begin{pmatrix} r'_2 & r_2 \\ r'_1 & r_1 \end{pmatrix} B^\dagger_{r'_1} B^\dagger_{r'_2} B^\dagger_{r_3} \cdots B^\dagger_{r_n} B_{r_n} \cdots B_{r_1}, \tag{3.19}$$

with $\alpha_n = -2\beta_n = \gamma_n$ for $n > 1$, with γ_n being the prefactor appearing in D_{mi} series [see Eq. (2.23)], while $(\alpha_1 = 1,\ \beta_1 = 0)$ for $n = 1$. In order to show this nicely simple result, we are going to determine the recursion relations which exist between α_n's and between β_n's. For that, we follow the procedure we have previously used, namely, we first insert closure relation for the N-pair states in front of $|\psi_N\rangle$. This leads to

$$H|\psi_N\rangle = \frac{1}{(N!)^2} \sum_{\{r\}} H B^\dagger_{r_1} \cdots B^\dagger_{r_N} |v\rangle\langle v| B_{r_N} \cdots B_{r_1} |\psi_N\rangle. \tag{3.20}$$

We then calculate H acting on N excitons through Eq. (1.10). This leads to

$$H B^\dagger_{r_1} \cdots B^\dagger_{r_N} |v\rangle = (B^\dagger_{r_1} H + E_{r_1} B^\dagger_{r_1} + V^\dagger_{r_1}) B^\dagger_{r_2} \cdots B^\dagger_{r_N} |v\rangle. \tag{3.21}$$

By using Eq. (1.11), we find

$$\begin{aligned} V^\dagger_{r_1} B^\dagger_{r_2} \cdots B^\dagger_{r_N} |v\rangle &= ([V^\dagger_{r_1}, B^\dagger_{r_2}] + B^\dagger_{r_2} V^\dagger_{r_1}) B^\dagger_{r_3} \cdots B^\dagger_{r_N} |v\rangle \\ &= \sum_{r'_1, r'_2} \xi\begin{pmatrix} r'_2 & r_2 \\ r'_1 & r_1 \end{pmatrix} B^\dagger_{r'_1} B^\dagger_{r'_2} B^\dagger_{r_3} \cdots B^\dagger_{r_N} |v\rangle \\ &\quad + B^\dagger_{r_2} V^\dagger_{r_1} B^\dagger_{r_3} \cdots B^\dagger_{r_N} |v\rangle. \end{aligned} \tag{3.22}$$

We iterate the procedure to end with

$$\begin{aligned} H B^\dagger_{r_1} \cdots B^\dagger_{r_N} |v\rangle &= (E_{r_1} + \cdots + E_{r_N}) B^\dagger_{r_1} \cdots B^\dagger_{r_N} |v\rangle \\ &\quad + \left\{ \sum_{r'_1, r'_2} \xi\begin{pmatrix} r'_2 & r_2 \\ r'_1 & r_1 \end{pmatrix} B^\dagger_{r'_1} B^\dagger_{r'_2} B^\dagger_{r_3} \cdots B^\dagger_{r_N} |v\rangle + \text{permutations} \right\}, \end{aligned} \tag{3.23}$$

the total number of these ξ terms being the number of ways C_N^2 we can choose among N, the two excitons having direct Coulomb process.

If we now relabel bold indices and remove projector $|v\rangle\langle v|$, we end with

$$\begin{aligned} H|\psi_N\rangle &= \frac{N}{(N!)^2} \sum_{\{r\}} E_{r_1} B^\dagger_{r_1} \cdots B^\dagger_{r_N} B_{r_N} \cdots B_{r_1} |\psi_N\rangle \\ &\quad + \frac{C_N^2}{(N!)^2} \sum_{r'_1, r'_2, \{r\}} \xi\begin{pmatrix} r'_2 & r_2 \\ r'_1 & r_1 \end{pmatrix} \\ &\quad \times B^\dagger_{r'_1} B^\dagger_{r'_2} B^\dagger_{r_3} \cdots B^\dagger_{r_N} B_{r_N} \cdots B_{r_1} |\psi_N\rangle. \end{aligned} \tag{3.24}$$

We now turn to $H^{(n)}$ acting on $|\psi_N\rangle$ and assume that its general form is indeed given by Eq. (3.19). Since state $B_{r_n} \cdots B_{r_1} |\psi_N\rangle$ has $(N-n)$ pairs, we get, by injecting closure relation for $(N-n)$-pair subspace,

$$\begin{aligned} H^{(n)}|\psi_N\rangle &= \frac{1}{[(N-n)!]^2} \sum_{\{r\}} \Bigg[\alpha_n E_{r_1} B^\dagger_{r_1} B^\dagger_{r_2} \\ &\quad + \beta_n \sum_{r'_1, r'_2} \xi\begin{pmatrix} r'_2 & r_2 \\ r'_1 & r_1 \end{pmatrix} B^\dagger_{r'_1} B^\dagger_{r'_2} \Bigg] \\ &\quad \times B^\dagger_{r_3} \cdots B^\dagger_{r_n} B^\dagger_{r_{n+1}} \cdots B^\dagger_{r_N} |v\rangle\langle v| B_{r_N} \cdots B_{r_1} |\psi_N\rangle. \end{aligned} \tag{3.25}$$

We then remove projector $|v\rangle\langle v|$ as usual. By collecting all these results, we find that operator $H^{(n)}$ defined through Eq. (3.6) has form (3.19) provided that α_n's and β_n's are linked by

$$\alpha_N = \frac{N}{(N!)^2} - \sum_{n=1}^{N-1} \frac{\alpha_n}{[(N-n)!]^2}, \tag{3.26}$$

$$\beta_N = \frac{C_N^2}{(N!)^2} - \sum_{n=2}^{N-1} \frac{\beta_n}{[(N-n)!]^2}, \tag{3.27}$$

with $\alpha_1 = 1$ and $\beta_1 = 0$ due to Eq. (3.9), while $\beta_2 = 1/4$ due to Eq. (3.14). By comparing Eqs. (2.23) and (3.26), we readily see that $\alpha_N = \gamma_N$. In order to determine β_N, we first note that the recursion relation for α_N also reads

$$\begin{aligned} \alpha_N &= \frac{N}{(N!)^2} - \frac{1}{[(N-1)!]^2} - \sum_{n=2}^{N-1} \frac{\alpha_n}{[(N-n)!]^2} \\ &= -\frac{N(N-1)}{(N!)^2} - \sum_{n=2}^{N-1} \frac{\alpha_n}{[(N-n)!]^2}. \end{aligned} \tag{3.28}$$

Since $C_N^2 = N(N-1)/2$, this equation is nothing but the recursion relation for β_N provided that we replace α_N by $(-2\beta_N)$ for any $N \geq 2$. Consequently, we end with

$$\gamma_N = \alpha_N = -2\beta_N \quad \text{for } N \geq 2, \tag{3.29}$$

while $\alpha_1 = \gamma_1 = 1$ and $\beta_1 = 0$, in agreement with our original guess.

IV. DISCUSSION

A. Summary of the results

The above results lead us to write deviation-from-boson operator D_{mi} of two composite excitons defined as

$$[B_m, B_i^\dagger] = \delta_{m,i} - D_{mi}, \tag{4.1}$$

through an infinite series of exciton-operator products, according to

$$D_{mi} = \sum_{r',r}\left[\lambda\begin{pmatrix} r' & r \\ m & i\end{pmatrix} + \lambda\begin{pmatrix} m & r \\ r' & i\end{pmatrix}\right] B_{r'}^\dagger\left(1 + \sum_{n=2}^{\infty}\gamma_n P_n\right)B_r, \tag{4.2}$$

$$P_n = \sum_{\{j\}} B_{j_1}^\dagger \cdots B_{j_{n-1}}^\dagger B_{j_{n-1}} \cdots B_{j_1}. \tag{4.3}$$

$\lambda(\begin{smallmatrix} r' & r \\ m & i\end{smallmatrix})$ is the Pauli scattering for carrier exchanges between in excitons (i,r) leading to out excitons (m,r'), with excitons (m,i) having same electron. Electron-hole symmetry is restored through the fact that, in the second term of Eq. (4.2), namely, $\lambda(\begin{smallmatrix} m & r \\ r' & i\end{smallmatrix})$, excitons (m,i) have same hole (see Fig. 1). γ_n's are numerical prefactors which obey the recursion relation

$$\gamma_N = \frac{N}{(N!)^2} - \sum_{n=1}^{N-1}\frac{\gamma_n}{[(N-n)!]^2}, \tag{4.4}$$

with $\gamma_1=1$ so that $\gamma_2=-1/2$, $\gamma_3=1/3$, and so on, with γ_N going to zero for increasing N.

In the same way, the system Hamiltonian, when acting on electron-hole-pair states, can be written as an infinite series of exciton-operator products, according to

$$H = \sum_r E_r B_r^\dagger\left(1 + \sum_{n=2}^{\infty}\gamma_n P_n\right)B_r + \frac{1}{2}\sum_{r_1,r_2,r_1',r_2'}\xi\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1\end{pmatrix}B_{r_1'}^\dagger B_{r_2'}^\dagger\left(\frac{1}{2} - \sum_{n=3}^{\infty}\gamma_n P_{n-1}\right)B_{r_2}B_{r_1}. \tag{4.5}$$

Let us again stress that, since there are two ways to form two excitons out of the two electron-hole pairs, any product $B^\dagger B^\dagger$ can be written as a sum of $B^\dagger B^\dagger$ according to Eq. (2.4). Consequently, it is always possible to rewrite sums appearing in D_{mi} and H in various different ways, Eqs. (4.2)–(4.5) being the simplest ones.

B. Comparison with the results of Mukamel and co-workers (Refs. 10–12)

In their works, Mukamel and co-workers[10–12] used free-pair operators $\hat{B}_m^\dagger = c_{m_1}^\dagger d_{m_2}^\dagger$, where $c_{m_1}^\dagger$ creates electron on site m_1 while $d_{m_2}^\dagger$ creates hole on site m_2. The fact that they use sites while we here use momenta [see Eq. (3.2)] is basically unimportant. They however keep the possibility for electrons and holes of these free pairs to differ from free Hamiltonian eigenstates. This is why they have nondiagonal contributions in the one-body part of their Hamiltonian,

$$H_0 = \sum_{m_1,n_1} t_{m_1n_1}^{(1)} c_{m_1}^\dagger c_{n_1} + \sum_{m_2,n_2} t_{m_2n_2}^{(2)} d_{m_2}^\dagger d_{n_2}. \tag{4.6}$$

As these free-pair states are not physically relevant states to describe a set of N interacting pairs, Mukamel and co-workers[10–12] only reach the two first terms of H series, namely, $H^{(1)}$ and $H^{(2)}$, and the first term of D_{mi} series, their results reading already as rather complicated [see Eq. (18) of Ref. 10]. To make precise link with their work, we are going to recover their results from our compact forms.

As electron-hole states used by Mukamel and co-workers[10–12] form a complete set, we can expand exciton operators $B_r^\dagger$ in terms of electron-hole operators $\hat{B}_n^\dagger$, according to

$$B_r^\dagger = \sum_n \hat{B}_n^\dagger \langle \hat{n}|r\rangle, \tag{4.7}$$

where $|r\rangle = B_r^\dagger|v\rangle$, while $|\hat{n}\rangle = \hat{B}_n^\dagger|v\rangle = c_{n_1}^\dagger d_{n_2}^\dagger|v\rangle$. Using this expansion (4.7), we see that the first term of the H series we have obtained also reads

$$H^{(1)} = \sum_{r_1} E_{r_1} B_{r_1}^\dagger B_{r_1} = \sum_{m,n} h_{mn}\hat{B}_m^\dagger \hat{B}_n, \tag{4.8}$$

where prefactor h_{mn} is nothing but

$$h_{mn} = \sum_{r_1} E_{r_1}\langle \hat{m}|r_1\rangle\langle r_1|\hat{n}\rangle = \langle \hat{m}|H|\hat{n}\rangle, \tag{4.9}$$

since $H|r_1\rangle = E_{r_1}|r_1\rangle$. If we now introduce the two-body part of the Hamiltonian as written in Eq. (16) of Ref. 10, namely,

$$H_c = \frac{1}{2}\sum V_{m_1n_1j_1k_1}^{(1)} c_{m_1}^\dagger c_{n_1}^\dagger c_{j_1} c_{k_1} + \frac{1}{2}\sum V_{m_2n_2j_2k_2}^{(2)} d_{m_2}^\dagger d_{n_2}^\dagger d_{j_2} d_{k_2} - \sum W_{m_1n_2k_1j_2} c_{m_1}^\dagger d_{n_2}^\dagger d_{j_2} c_{k_1}, \tag{4.10}$$

we see that, for $H=H_0+H_c$ with H_0 given in Eq. (4.6), prefactor h_{mn} defined in Eq. (4.9) splits as

$$h_{mn} = h_{mn}^{(0)} - W_{m_1m_2n_1n_2},$$

$$h_{mn}^{(0)} = t_{m_1n_1}^{(1)}\delta_{m_2,n_2} + t_{m_2n_2}^{(2)}\delta_{m_1,n_1}, \tag{4.11}$$

in agreement with the result obtained by Mukamel and co-workers[10] for the first term of H expansion.

We now turn to $H^{(2)}$. In view of Eq. (3.14), $H^{(2)}$ splits as $H^{(2)} = H_E^{(2)} + H_\xi^{(2)}$, where $H_E^{(2)}$ depends on exciton energy E_r while $H_\xi^{(2)}$ depends on exciton scattering ξ. Before going further, let us note that, since $H_E^{(2)}$ contains exciton energy E_r, this term, by construction, contains a part of electron-hole Coulomb interaction, namely, the one acting *inside one exciton*. On the other hand, as $H_\xi^{(2)}$ reads in terms of direct Coulomb scattering $\xi(\begin{smallmatrix} r_2' & r_2 \\ r_1' & r_1\end{smallmatrix})$ *between the two excitons* (r_1,r_2), it contains Coulomb scattering resulting from electron-electron and hole-hole interactions, as well as from electron-hole interaction *between* excitons r_1 and r_2 [see Fig. 3(a)]. Since electron-hole interaction already appears in the first-order term $H^{(1)}$ through $W_{m_1m_2n_1n_2}$ in h_{mn} [see Eq. (4.11)], these two electron-hole contributions of $H^{(2)}$ must somehow cancel, as we now show.

If we symmetrize $H_E^{(2)}$ and write exciton operators in terms of free pairs according to Eq. (4.7), we find

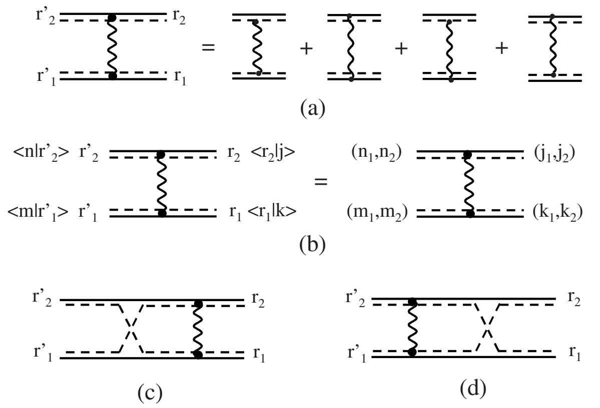

FIG. 3. (a) Direct Coulomb scattering $\xi\binom{r_2'\ r_2}{r_1'\ r_1}$ between the in excitons (r_1,r_2) leading to the out excitons (r_1',r_2'). This scattering is "direct" in the sense that the electron-hole pairs are coupled similarly in the in and out states. (b) Diagrammatic representation of the sum over $\{r\}$ appearing in Eq. (4.14) and leading to Eq. (4.15). (c) Shiva diagram representation for the in exchange Coulomb scattering $\xi^{\text{in}}\binom{r_2'\ r_2}{r_1'\ r_1}$ defined in Eq. (4.22). In this scattering, the Coulomb interactions are between the in excitons. (d) Shiva diagram representation of the out exchange Coulomb scattering $\xi^{\text{out}}\binom{r_2'\ r_2}{r_1'\ r_1}$ defined in Eq. (4.23). In this scattering, the Coulomb interactions are between the out excitons.

$$H_E^{(2)} = -\frac{1}{4}\sum_{r_1,r_2}(E_{r_1}+E_{r_2})B_{r_1}^\dagger B_{r_2}^\dagger B_{r_2}B_{r_1} = -\frac{1}{4}\sum_{m,n,j,k}\hat{B}_m^\dagger\hat{B}_n^\dagger\hat{B}_j\hat{B}_k \times\left[\sum_{r_1}\langle\hat{m}|H|r_1\rangle\langle r_1|\hat{k}\rangle\sum_{r_2}\langle\hat{n}|r_2\rangle\langle r_2|\hat{j}\rangle+(1\leftrightarrow 2)\right]. \tag{4.12}$$

Using Eq. (4.9) and orthogonality of free-pair states, we readily find

$$H_E^{(2)} = -\frac{1}{4}\sum_{m,n,j,k}\hat{B}_m^\dagger\hat{B}_n^\dagger\hat{B}_j\hat{B}_k(h_{mk}\delta_{n,j}+h_{nj}\delta_{m,k}), \tag{4.13}$$

with h_{mn} given in Eq. (4.11).

If we now consider the part of $H_\xi^{(2)}$ coming from Coulomb scattering ξ between excitons, we can rewrite it, using again Eq. (4.7), as

$$H_\xi^{(2)} = \frac{1}{4}\sum_{r_1,r_2,r_1',r_2'}\xi\begin{pmatrix} r_2' & r_2\\ r_1' & r_1\end{pmatrix}B_{r_1'}^\dagger B_{r_2'}^\dagger B_{r_2}B_{r_1} = \frac{1}{4}\sum_{m,n,j,k}\hat{B}_m^\dagger\hat{B}_n^\dagger\hat{B}_j\hat{B}_k\sum_{r_1,r_2,r_1',r_2'}\xi\begin{pmatrix} r_2' & r_2\\ r_1' & r_1\end{pmatrix}\langle\hat{m}|r_1'\rangle\langle\hat{n}|r_2'\rangle\langle r_2|\hat{j}\rangle \times\langle r_1|\hat{k}\rangle. \tag{4.14}$$

The sum over r's is readily obtained from diagrams of Fig. 3(b) in terms of interactions $V^{(1)}$ between electrons, $V^{(2)}$ between holes, and W between electrons and holes. It reduces to

$$\{V^{(1)}_{m_1n_1j_1k_1}\delta_{m_2,k_2}\delta_{n_2,j_2} - W_{m_1n_2k_1j_2}\delta_{m_2,k_2}\delta_{n_1,j_1}\} + \{1\leftrightarrow 2\}. \tag{4.15}$$

If we now collect the two parts of $H^{(2)}$, we can rewrite it as

$$H^{(2)} = \sum_{m,n,j,k}U_{mnjk}\hat{B}_m^\dagger\hat{B}_n^\dagger\hat{B}_j\hat{B}_k + Z, \tag{4.16}$$

where U_{mnjk} is just the prefactor obtained by Mukamel *et al.* in Eq. (18) of Ref. 10. Operator Z contains all electron-hole contributions. Its precise value reads

$$Z = \sum_{m,n,j,k}\hat{B}_m^\dagger\hat{B}_n^\dagger\hat{B}_j\hat{B}_k\left[\frac{1}{4}(W_{m_1m_2k_1k_2}\delta_{n,j}+W_{n_1n_2j_1j_2}\delta_{m,k}) - \frac{1}{4}(W_{m_1n_2k_1j_2}\delta_{m_2,k_2}\delta_{n_1,j_1}+W_{n_1m_2j_1k_2}\delta_{m_1,k_1}\delta_{n_2,j_2})\right]. \tag{4.17}$$

In order for Eq. (4.16) to agree with the expression of $H^{(2)}$ obtained by Mukamel and co-workers,[10] operator Z must reduce to zero. This is actually true, as shown by noting that

$$\hat{B}_m^\dagger\hat{B}_n^\dagger\hat{B}_j\hat{B}_k = c_{m_1}^\dagger d_{m_2}^\dagger c_{n_1}^\dagger d_{n_2}^\dagger d_{j_2}c_{j_1}d_{k_2}c_{k_1} = c_{m_1}^\dagger(-d_{n_2}^\dagger c_{n_1}^\dagger d_{m_2}^\dagger)(-d_{j_2}c_{k_1}d_{k_2})c_{j_1} \tag{4.18}$$

and by exchanging bold indices $(m_2\leftrightarrow n_2)$ and $(j_2\leftrightarrow k_2)$ in the sums appearing in Z. This explicitly shows that electron-hole interaction does not appear in $H^{(2)}$ as reasonable since, due to Eq. (3.3), V_{eh} can be exactly written in terms of $B_i^\dagger B_j$ or $\hat{B}_m^\dagger\hat{B}_n$.

We thus conclude that expressions of $H^{(1)}$ and $H^{(2)}$ given by Mukamel and co-workers[10] agree with our compact form of $H^{(n)}$. As the exciton operators we here use are physically relevant operators for interacting electron-hole pairs, we have been able to write the whole infinite series for H in a compact form in terms of these operators. Let us however stress that, even with this infinite series now known, it is far simpler to calculate $H|\psi_N\rangle$ through the commutators $[H,B_i^{\dagger N}]$ and $[V_i^\dagger,B_j^{\dagger N}]$, given in Eqs. (1.6) and (1.7), than through this $H^{(n)}$ series, mostly when the state $|\psi_N\rangle$ of interest has many identical excitons, as in usual physically relevant situations.

C. Possible use of series expansion for *H*

The procedure proposed by Mukamel and co-workers[10–12] is definitely not a bosonization procedure, since *exact* deviation-from-boson operators are *a priori* kept through their expansion as a series of pair operators. We can however be tempted by comparing prefactors obtained in this expansion of the system Hamiltonian H in terms of exciton operators, with effective scatterings produced by bosonization.

When truncated to its one- and two-body terms, the effective Hamiltonian for bosonized excitons reads as

$$\bar{H} = \sum_i E_i \bar{B}_i^\dagger \bar{B}_i + \frac{1}{2} \sum_{mnij} \bar{\xi}\begin{pmatrix} n & j \\ m & i \end{pmatrix} \bar{B}_m^\dagger \bar{B}_n^\dagger \bar{B}_i \bar{B}_j, \quad (4.19)$$

with $[\bar{B}_m, \bar{B}_i^\dagger] = \delta_{m,i}$ for elementary bosons. We see that the prefactor of the first term of $\bar{H}$ is nothing but the one of $H^{(1)}$. If we now consider $H^{(2)}$ given in Eq. (3.14), we can rewrite it as

$$H^{(2)} = \frac{1}{4} \sum_{r_1,r_2,r_1',r_2'} \left[\xi\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} - (E_{r_1} + E_{r_2})(\delta_{r_1',r_1} + \delta_{r_2',r_2}) \right] \times B_{r_1'}^\dagger B_{r_2'}^\dagger B_{r_2} B_{r_1}. \quad (4.20)$$

Due to the two ways to form two excitons out of two electron-hole pairs which lead to Eq. (2.4), we get from this equation used for $B^\dagger B^\dagger$ or BB,

$$\sum_{r_1,r_2,r_1',r_2'} \xi\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} B_{r_1'}^\dagger B_{r_2'}^\dagger B_{r_2} B_{r_1} = - \sum_{r_1,r_2,r_1',r_2'} \xi^{\text{in}}\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} B_{r_1'}^\dagger B_{r_2'}^\dagger B_{r_2} B_{r_1} = - \sum_{r_1,r_2,r_1',r_2'} \xi^{\text{out}}\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} B_{r_1'}^\dagger B_{r_2'}^\dagger B_{r_2} B_{r_1}, \quad (4.21)$$

where ξ^{in} and ξ^{out}, shown in Figs. 3(c) and 3(d), are defined as

$$\xi^{\text{in}}\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} = \sum_{p_1,p_2} \lambda\begin{pmatrix} r_2' & p_2 \\ r_1' & p_1 \end{pmatrix} \xi\begin{pmatrix} p_2 & r_2 \\ p_1 & r_1 \end{pmatrix}, \quad (4.22)$$

$$\xi^{\text{out}}\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} = \sum_{p_1,p_2} \xi\begin{pmatrix} r_2' & p_2 \\ r_1' & p_1 \end{pmatrix} \lambda\begin{pmatrix} p_2 & r_2 \\ p_1 & r_1 \end{pmatrix}. \quad (4.23)$$

This shows that in Eq. (4.20), ξ can be replaced by $(-\xi^{\text{in}})$ or $(-\xi^{\text{out}})$ or even by $(a\xi - b\xi^{\text{in}} - c\xi^{\text{out}})$ with $a+b+c=1$.

If we now turn to the E part of $H^{(2)}$, the same Eq. (2.4) used for $B^\dagger B^\dagger$ or BB leads to

$$\sum_{r_1,r_2} (E_{r_1} + E_{r_2}) B_{r_1}^\dagger B_{r_2}^\dagger B_{r_2} B_{r_1} = - \sum_{r_1,r_2,r_1',r_2'} (E_{r_1} + E_{r_2}) \lambda\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} B_{r_1'}^\dagger B_{r_2'}^\dagger B_{r_2} B_{r_1} = - \sum_{r_1,r_2,r_1',r_2'} (E_{r_1'} + E_{r_2'}) \lambda\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} B_{r_1'}^\dagger B_{r_2'}^\dagger B_{r_2} B_{r_1}, \quad (4.24)$$

so that E prefactor in $H^{(2)}$ gives rise to two-body scattering between excitons in $E\lambda$. This shows that the second term of H expansion can also be written as

$$H^{(2)} = \frac{1}{2} \sum_{r_1,r_2,r_1',r_2'} S\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} B_{r_1'}^\dagger B_{r_2'}^\dagger B_{r_2} B_{r_1}, \quad (4.25)$$

$$S\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} = \frac{1}{2}\left[a\xi\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} - b\xi^{\text{in}}\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} - c\xi^{\text{out}}\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} - (E_{r_1'} + E_{r_2'})\lambda\begin{pmatrix} r_2' & r_2 \\ r_1' & r_1 \end{pmatrix} \right], \quad (4.26)$$

with $a+b+c=1$. It is, however, clear that such a $S\left(\begin{smallmatrix} r_2' & r_2 \\ r_1' & r_1 \end{smallmatrix}\right)$ cannot be used as an effective scattering between two excitons. Indeed, S depends on energy origin through exciton energy E_i which includes the band gap in the case of excitons, while it is physically irrelevant to have the band gap entering exciton scattering. Even if we drop these spurious $E\lambda$ terms, this $S\left(\begin{smallmatrix} r_2' & r_2 \\ r_1' & r_1 \end{smallmatrix}\right)$ has a problem since its ξ part differs from the effective scattering between bosonized excitons mostly used in the literature, namely, $\bar{\xi}\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right) = \xi\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right) - \xi^{\text{out}}\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right)$, by at least a factor of 1/2, in addition to the fact that effective Hamiltonians with such a $\bar{\xi}$ are not Hermitian. Indeed, in order for $\bar{H}$ to be Hermitian, we must have $\bar{\xi}\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right) = \bar{a}\xi\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right) - \bar{b}\xi^{\text{in}}\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right) - \bar{c}\xi^{\text{out}}\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right)$, with $\bar{a} = \bar{a}^*$ and $\bar{b} = \bar{c}^*$. With respect to Hermiticity, let us recall that Eqs. (4.25) and (4.26) are written with composite-boson operators, not elementary-boson operators $\bar{B}_r$; this makes Eq. (4.21) correct, i.e., $H^{(2)}$ Hermitian, even for $a \neq a^*$ and $b \neq c^*$.

V. CONCLUSION

In this paper, we revisit the procedure proposed by Mukamel and co-workers[10–12] to approach interactions between excitons while keeping their composite nature exactly, through infinite series of composite-boson operators for both the system Hamiltonian and the deviation-from-boson operator of these composite bosons. While Mukamel and co-workers[10–12] used free-electron-hole-pair operators, we here use exciton operators which are physically relevant operators for problems dealing with excitons. This allows us to write all terms of these two infinite series explicitly. They read in terms of exciton energies as well as Pauli and interaction scatterings that appear in the composite-boson many-body theory we have recently constructed. We show that the first-order terms found by Mukamel and co-workers[10–12] agree with our results. However, the necessary handling of these two infinite series for calculations dealing with N excitons makes this approach far more complicated than the ones based on the many-body theory for composite bosons we have proposed and which only relies on four nicely simple commutators.

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