Effective bosonic hamiltonian for excitons: a too naive concept

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Abstract

Excitons, being made of two fermions, may appear from far as bosons. Their close-to-boson character is however quite tricky to handle properly. Using our commutation technique especially designed to deal with interacting close-to-boson particles, we here calculate the exact expansion in Coulomb interaction of the exciton-exciton correlations, and show that a naive effective bosonic hamiltonian for excitons cannot produce these X-X correlations correctly.

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Up to now, problems dealing with interacting particles have in common the fact that
the hamiltonian can be separated into \( H = H_0 + V \). The eigenstates of the non-interacting
part \( H_0 \) of this hamiltonian being usually known exactly, they can serve as an orthogonal
basis for the system, so that expansions in the interaction \( V \) are simply obtained by using
the identity

\[
\frac{1}{a - H} = \frac{1}{a - H_0} + \frac{1}{a - H} V \frac{1}{a - H_0} \\
= \frac{1}{a - H_0} + \frac{1}{a - H_0} V \frac{1}{a - H_0} + \frac{1}{a - H_0} V \frac{1}{a - H_0} V \frac{1}{a - H_0} + \ldots
\]

(with \( a \) being usually \((\omega + i\eta)\)), and by inserting the closure relation for \( H_0 \) eigenstates
on both sides of the \( V \) operators\(^{(1)}\).

The problem of interacting excitons is much more tricky because it is not at all of
that type. The excitons are indeed the exact one electron-hole pair eigenstates of the
semiconductor hamiltonian \( H \) and thus can serve as an orthogonal basis for any one-pair
state of the semiconductor. However when there is one pair only in the system, there is
no X-X interaction ! The problem starts with two pairs as there is no way\(^{(2)}\) to extract
from the semiconductor hamiltonian \( H \) an \( H_0 \) part for which the product of two excitons
would be the exact eigenstates and so could serve as an orthogonal basis for two-pair
states.

The fundamental difficulty with interacting excitons comes from the fact that they are
composite particles made of two fermions, so that they are not "clean" particles. There
are a priori two ways to couple two electrons and two holes to make two excitons. However
these two ways are hard to cope with the fact that electrons and holes are indiscernable
particles. Moreover, as electrons and holes are fermions, it is physically obvious that
two excitons must "feel" each other not only through Coulomb interaction between their
carriers but also through Pauli exclusion between their indiscernable components, even
in the absence of any Coulomb interaction : This Pauli exclusion is in fact the extremely
subtle part of the interacting exciton problem.

Attempts have been made, using various bosonisation procedures\(^{(3,4)}\), to replace the
exact semiconductor hamiltonian \( H \) by an effective hamiltonian\(^{(5)}\),
\[ H_{\text{eff}} = H_{\text{eff}}^0 + V_{\text{eff}} \]
\[ = \sum_i E_i \bar{B}_i^\dagger \bar{B}_i + \frac{1}{2} \sum_{mnij} E_{mnij} \bar{B}_m^\dagger \bar{B}_n^\dagger \bar{B}_i \bar{B}_j, \]

in which the excitons are assumed to be bosonic particles, \([\bar{B}_i, \bar{B}_j^\dagger] = \delta_{ij}\), provided that the underlying fermionic character of their components is included in an appropriate exciton interaction \(E_{mnij}\). We have recently shown \(^2\) that the proposed \(V_{\text{eff}}\) cannot be correct: First, it is not an hermitian operator as the given \(^6\) \(E_{mnij}\) is not equal to \(E_{ijmn}^*\). Even if this mistake is corrected, the bosonisation procedures actually dress the exact Coulomb interaction operator by exchange processes which originate from Pauli exclusion between carriers, so that by construction they miss purely Pauli terms, i.e. terms without \(e^2/r\) factors. We have shown \(^2\) that these Pauli terms are precisely those necessary to restore the hermiticity of the effective exciton Hamiltonian quoted up to now.

In the present work, we show that, in addition to the existence of these conceptually new purely Pauli terms, Pauli exclusion, when handled properly, dresses the Coulomb scatterings in such a subtle way that we do not see how a dressed exciton-exciton Coulomb interaction can produce the exchange Coulomb terms of the exciton-exciton correlations correctly. In other words, in the low density limit, it would be nice to replace the exact semiconductor Hamiltonian by an effective Hamiltonian for boson-excitons; but, unfortunately, the form quoted in Eq (2) is too naive \(^5\) to be satisfactory.

Our work relies on the fact that, although \(H\) cannot be written as \(H_0 + V\), with \(V\) describing interactions between excitons, it is nevertheless possible to generate an exact expansion in Coulomb interaction, in its spirit similar to Eq (1), owing to our commutation technique. This technique is briefly summarized in section 1 for excitons without spin degrees of freedom. While easy to include \(^7\), these spin parameters make the notations quite heavy so that they tend to hide the subtle physics induced by Pauli exclusion.

In section 2, we show how our commutation technique allows to calculate the X-X correlations exactly at any order in Coulomb interactions. From a trivial algebra, we see that exchange processes induced by Pauli exclusion enter the correlation terms in a far from obvious way. From the form of these correlation terms, we see no way to find an exciton-exciton scattering which corresponds to Coulomb interactions dressed by exchange processes and which can be valid beyond first order in Coulomb interaction!
1 Survey of the commutation technique

Our commutation technique \((^{2,7})\) allows to calculate any quantity dealing with interacting excitons in terms of two parameters \(\xi_{mnij}^{\text{dir}}\) and \(\lambda_{mnij}\).

The first one comes from Coulomb interaction. It appears through

\[
\left[ V_i^\dagger, B_j^\dagger \right] = \sum_{m,n} \xi_{mnij}^{\text{dir}} B_m^\dagger B_n^\dagger, \tag{3}
\]

where the operator \(V_i^\dagger\) describes \(^{(8)}\) the Coulomb interaction between the \(i\) exciton and the rest of the system. It is defined by

\[
\left[ H, B_i^\dagger \right] = E_i B_i^\dagger + V_i^\dagger, \tag{4}
\]

where \(H\) is the exact semiconductor hamiltonian, and \(B_i^\dagger\) the exact (one) exciton creation operator, \(HB_i^\dagger | v > = E_i B_i^\dagger | v >\), \(E_i\) being the \(i\) exciton energy, and \(| v >\) the vacuum state for electron-hole pairs.

The second parameter is linked to the close-to-boson character of the excitons induced by Pauli exclusion. It appears through

\[
\left[ D_{ni}, B_j^\dagger \right] = 2 \sum_m \lambda_{mnij} B_m^\dagger, \tag{5}
\]

where \(D_{ni}\) is the deviation-from-boson operator defined by \(\left[ B_n, B_i^\dagger \right] = \delta_{ni} - D_{ni}\). We note that, if the excitons were exact bosons, \(D_{ni} \equiv 0\), so that \(\lambda_{mnij}\) would be zero. This \(\lambda_{mnij}\) parameter also appears when one couples the electrons and holes of two excitons in a different way. We then get \(^{(2)}\)

\[
B_i^\dagger B_j^\dagger = - \sum_{m,n} \lambda_{mnij} B_m^\dagger B_n^\dagger. \tag{6}
\]

The fact that \(B_i^\dagger B_j^\dagger | v >\) is not a well defined state as it contains a piece of any other two-exciton state, makes the exciton problem quite tricky. In particular, these \(B_i^\dagger B_j^\dagger | v >\) states do not form an orthogonal basis as

\[
\frac{1}{2!} < v | B_m B_n B_i^\dagger B_j^\dagger | v > = \delta_{mnij} - \lambda_{mnij}, \tag{7}
\]

which is easy to deduce from Eq (5). This matrix element thus differs from zero even if \(\delta_{mnij} = (\delta_{mi} \delta_{nj} + \delta_{mj} \delta_{ni})/2\) is zero, i.e. even if \((m, n) \neq (i, j)\).
A clear physical understanding of these two crucial parameters for interacting excitons, $\xi_{mnij}^{\text{dir}}$ and $\lambda_{mnij}$, is easy to get from their expressions in $r$ space. The (direct) Coulomb parameter is equal to (2)

$$
\xi_{mnij}^{\text{dir}} = \frac{1}{2} \int de_1 \, de_2 \, dh_1 \, dh_2 \, \phi_m^*(e_1, h_1) \, \phi_n^*(e_2, h_2) \left[ V_{e_1e_2} + V_{h_1h_2} - V_{e_1h_2} - V_{e_2h_1} \right] \times \phi_i(e_1, h_1) \, \phi_j(e_2, h_2) + (m \leftrightarrow n),
$$

where $V_{eh} = e^2 / |r_e - r_h|$ while $\phi_i(e, h)$ is the total wave function of the $i$ exciton with its electron in $r_e$ and its hole in $r_h$. $\xi_{mnij}^{\text{dir}}$ thus corresponds to all Coulomb processes between $(m,n)$ and $(i,j)$ excitons when these excitons are built on the same pairs $(e_1, h_1)$ and $(e_2, h_2)$. The $\lambda_{mnij}$ parameter reads in $r$ space,

$$
\lambda_{mnij} = \frac{1}{2} \int de_1 \, de_2 \, dh_1 \, dh_2 \, \phi_m^*(e_1, h_1) \phi_n^*(e_2, h_2) \phi_i(e_1, h_2) \phi_j(e_2, h_1) + (m \leftrightarrow n),
$$

so that it simply describes the possible exchange of the two electrons (or holes) when building the $(m,n)$ and $(i,j)$ excitons.

In many physical quantities also appear the combinations,

$$
\xi_{mnij}^{\text{right}} = \sum_{r,s} \xi_{mnrs}^{\text{dir}} \lambda_{rsij}, \quad \xi_{mnij}^{\text{left}} = \sum_{r,s} \lambda_{mnrs} \xi_{rsij}^{\text{dir}},
$$

which are linked by (2)

$$
(E_m + E_n - E_i - E_j) \lambda_{mnij} = \xi_{mnij}^{\text{left}} - \xi_{mnij}^{\text{right}}.
$$

Their half sum reads in $r$ space,

$$
\xi_{mnij}^{\text{exch}} = \frac{1}{2} \left( \xi_{mnij}^{\text{right}} + \xi_{mnij}^{\text{left}} \right) = \frac{1}{2} \int de_1 \, de_2 \, dh_1 \, dh_2 \, \phi_m^*(e_1, h_1) \phi_n^*(e_2, h_2) \
\times \left[ V_{e_1e_2} + V_{h_1h_2} - (V_{e_1h_1} + V_{e_2h_2} + V_{e_1h_2} + V_{e_2h_1})/2 \right] \phi_i(e_1, h_2) \phi_j(e_2, h_1) + (m \leftrightarrow n),
$$

so that $\xi_{mnij}^{\text{exch}}$ contains all possible Coulomb processes between the two electrons and the two holes of the two excitons, when these $(m,n)$ and $(i,j)$ excitons are built with different electron-hole pairs. $\xi_{mnij}^{\text{right}}$ reads as $\xi_{mnij}^{\text{exch}}$ with the e-h contribution being $(V_{e_1h_2} + V_{e_2h_1})$, while for $\xi_{mnij}^{\text{left}}$ this e-h contribution is $(V_{e_1h_1} + V_{e_2h_2})$.

Let us end this survey by noting that, as obvious from Eqs (8,9,10,12), all the $\xi$ parameters are homogeneous to an energy, while the $\lambda$ parameter is dimensionless.
2 Exciton-exciton correlations

In plenty of problems, the hamiltonian $H$ appears through matrix elements of $1/(a-H)$ where $a$ is usually $(\omega + i\eta)$. As the excitons are the exact one-pair eigenstates of $H$, the matrix elements of $1/(a-H)$ between one-exciton states are simply

$$<v|\frac{1}{a-H}B_i^\dagger B_j^\dagger|v>=\frac{1}{a-E_i}\delta_{ij}. \quad (13)$$

The difficulties arrive with two-pair states because $HB_i^\dagger B_j^\dagger|v>\neq (E_i+E_j)B_i^\dagger B_j^\dagger|v>$, so that

$$G_{mnij}(a) = \frac{1}{2!} <v|B_mB_n\frac{1}{a-H}B_i^\dagger B_j^\dagger|v> \quad (14)$$

is not equal to

$$G^0_{mnij}(a) = \frac{\delta_{mnij}}{a-E_i-E_j} = \delta_{mnij}g_{ij}(a) \quad (15)$$

$$= \frac{1}{2!} <v|\tilde{B}_m\tilde{B}_n\frac{1}{a-H^0_{\text{eff}}}\tilde{B}_i^\dagger \tilde{B}_j^\dagger|v>, \quad (16)$$

as it would be for excitons interacting neither by Coulomb interaction nor by Pauli exclusion. The matrix elements of $1/(a-H)$ between two-exciton states have additional terms we are now going to calculate.

A clean way to have the Coulomb interactions between excitons appearing is via the operators $V_i^\dagger$ defined in Eq (4). From this equation, we get $B_i^\dagger(a-H-E_i) = (a-H)B_i^\dagger + V_i^\dagger$, so that we do have

$$\frac{1}{a-H}B_i^\dagger = B_i^\dagger\frac{1}{a-H-E_i} + \frac{1}{a-H}V_i^\dagger\frac{1}{a-H-E_i}. \quad (17)$$

Eq (17) is, in its spirit, very similar to Eq (1) which allows to derive correlation effects for usual problems dealing with interactions. This Eq (17) is in fact the key equation for problems dealing with correlation effects between excitons: For two excitons, it leads to

$$\frac{1}{a-H}B_i^\dagger B_j^\dagger|v> = \frac{1}{a-E_i-E_j}\left(B_i^\dagger + \frac{1}{a-H}V_i^\dagger\right)B_j^\dagger|v>, \quad (18)$$

with a similar equation for $<v|B_mB_n(1/(a-H))$. From them and Eqs (3,7), we can obtain the integral equations verified by the matrix elements of $1/(a-H)$ in the two-exciton subspace. They read

$$G_{mnij}(a) = G^0_{mnij}(a) - \sum_{pqrst}G^0_{mnpq}(a)X^\text{right}_{pqrs}(a)G^0_{rsij}(a) + \sum_{pqrst}G^0_{mnpq}(a)\zeta^\text{dir}_{pqrs}G^0_{rsij}(a) \quad (19)$$

$$= G^0_{mnij}(a) - \sum_{pqrst}G^0_{mnpq}(a)X^\text{left}_{pqrs}(a)G^0_{rsij}(a) + \sum_{pqrst}G^0_{mnpq}(a)\zeta^\text{dir}_{pqrs}G^0_{rsij}(a), \quad (20)$$
where we have set

\[
\begin{align*}
X_{mnij}^{\text{right}}(a) &= (a - E_m - E_n) \lambda_{mnij}, \\
X_{mnij}^{\text{left}}(a) &= \lambda_{mnij} (a - E_i - E_j).
\end{align*}
\] (21)

The X's appear as the vertices one has to associate to Pauli exclusion between two excitons. Due to Eq (11), the X's and the \(\xi\)'s are linked by

\[
X_{mnij}^{\text{left}}(a) - X_{mnij}^{\text{right}}(a) = \xi_{mnij}^{\text{left}} - \xi_{mnij}^{\text{right}}. \tag{22}
\]

These Pauli vertices are however rather peculiar since, depending on \(a\), they depend on the particular problem of interest. We can note that the Coulomb interactions dressed by exchange processes, \(\xi_{mnij}^{\text{right}}\) and \(\xi_{mnij}^{\text{left}}\) introduced previously, are just a sequence of a direct Coulomb scattering and a right or left Pauli scattering before or after it:

\[
\begin{align*}
\xi_{mnij}^{\text{right}} &= \sum_{pqrs} \xi_{mnpq}^{\text{dir}} G_{pqrs}^{0}(a) X_{rsij}^{\text{right}}(a), \\
\xi_{mnij}^{\text{left}} &= \sum_{pqrs} X_{mnpq}^{\text{left}}(a) G_{pqrs}^{0}(a) \xi_{rsij}^{\text{dir}}. \tag{23}
\end{align*}
\]

Let us however stress that, while the X's depend on \(a\), neither \(\xi_{mnij}^{\text{left}}\) nor \(\xi_{mnij}^{\text{right}}\) depend on it.

Before going further, let us note that the integral equations (18-19) for \(G_{mnij}(a)\) are indeed rather nice as they look like Dyson equations \(^{(9)}\), \(G = G^{0} + G^{0} V G\), except for the additional Pauli term which is \(G^{0} X G^{0}\) and not \(G^{0} X G\). This ”little” change makes all the subtle effects induced by Pauli exclusion between excitons.

As for \(\xi_{mnij}^{\text{exch}}\), it will be convenient to introduce the Pauli scattering

\[
X_{mnij}(a) = \frac{1}{2} \left[ X_{mnij}^{\text{right}}(a) + X_{mnij}^{\text{left}}(a) \right] = \left( a - \frac{E_i + E_j + E_m + E_n}{2} \right) \lambda_{mnij}. \tag{24}
\]

By inserting Eq (19) into Eq (20) and vice versa, and by using Eqs (12,23,24), the integral equations verified by \(G_{mnij}(a)\) take a form more symmetrical with respect to the left and right exchange processes:

\[
G_{mnij}(a) = G_{mnij}^{0}(a) + g_{mn}(a) \left[ \xi_{mnij}^{\text{dir}} - \xi_{mnij}^{\text{exch}} - X_{mnij}(a) \right] g_{ij}(a)
\]

\[
+ g_{mn}(a) \left( \sum_{pqrs} \xi_{mnpq}^{\text{dir}} G_{pqrs}^{0}(a) \xi_{rsij}^{\text{dir}} \right) g_{ij}(a), \tag{25}
\]

with \(g_{ij}(a)\) defined in Eq (15). This leads to

\[
G_{mnij}(a) = G_{mnij}^{0}(a) + g_{mn}(a) \left[ \xi_{mnij}^{\text{dir}} - \xi_{mnij}^{\text{exch}} + \xi_{mnij}^{\text{corr}}(a) - X_{mnij}(a) \right] g_{ij}(a), \tag{26}
\]
where $\xi_{mnij}^{\text{corr}}(a)$ corresponds to all direct and exchange correlation processes of the $(i, j)$ excitons into the $(m, n)$ states, with two or more Coulomb interactions. From Eqs (23-25), we find that $\xi_{mnij}^{\text{corr}}(a)$ verifies the integral equation

$$\xi_{mnij}^{\text{corr}}(a) = \Xi_{mnij}^{(2)}(a) + \Xi_{mnij}^{(3)}(a) + \sum_{pqrst} \xi_{mnpq}^{\text{dir}}(a) g_{pq}(a) \xi_{pqrs}^{\text{corr}}(a) g_{rs}(a) \xi_{rsij}^{\text{dir}}, \quad (27)$$

where $\Xi_{mnij}^{(2)}(a)$ contains all direct and exchange second order contributions in $\xi$, namely,

$$\Xi_{mnij}^{(2)}(a) = \sum_{pq} \left[ \xi_{mnpq}^{\text{dir}}(a) \xi_{pqqij}^{\text{dir}} - \frac{1}{2} \xi_{mnpq}^{\text{dir}}(a) \xi_{pqqij}^{\text{dir}} - \frac{1}{2} \xi_{mnpq}^{\text{dir}}(a) \xi_{pqij}^{\text{dir}} \right], \quad (28)$$

while $\Xi_{mnij}^{(3)}(a)$ contains all third order contributions,

$$\Xi_{mnij}^{(3)}(a) = \sum_{pqrs} \xi_{mnpq}^{\text{dir}}(a) \left[ \xi_{pqrs}^{\text{dir}} - \xi_{pqrs}^{\text{exch}} \right] g_{rs}(a) \xi_{rsij}^{\text{dir}}. \quad (29)$$

Let us examine the equations (26-29) giving the matrix elements of $1/(a - H)$ in the two-exciton subspace. From the first order contribution in Coulomb interaction, i.e. in $\xi$, we could think that Pauli exclusion dresses the bare Coulomb interaction by transforming $\xi^{\text{dir}}$ into $\xi^{\text{dir}} - \xi^{\text{exch}}$. A rapid glance at the higher order processes immediately shows that the way the exchange processes enter the correlation terms is much more subtle, and that the linear combination $\xi^{\text{dir}} - \xi^{\text{exch}}$ appearing at first order in $\xi$, does not remain unchanged at higher order: While odd terms in Coulomb interaction do depend on $\xi^{\text{exch}}$, this symmetrical combination of $\xi^{\text{right}}$ and $\xi^{\text{left}}$ does not appear in the even terms, as they contain the exchange Coulomb processes in different positions in the sequence of direct Coulomb interactions. Moreover, we see that the exchange contributions appear once only, in the "middle" of a set of $\xi^{\text{dir}}$.

As a major consequence, this shows that there is little hope to identify an effective boson-exciton interaction resulting from direct and exchange Coulomb processes which could produce the expansion of the $1/(a - H)$ matrix elements exactly: Indeed if an effective hamiltonian like the one of Eq (2) were to exist, the expansion would be:
\[ G_{mnij}^{\text{eff}} = \frac{1}{2!} \langle v | \bar{B}_m B_n \frac{1}{a - H_{\text{eff}}} \bar{B}_i \bar{B}_j | v \rangle \]
\[ = G_{mnij}^0(a) + g_{mn}(a) \mathcal{E}_{mnij} g_{ij}(a) \]
\[ + \sum_{pq} g_{mn}(a) \mathcal{E}_{mnpq} g_{pq}(a) \mathcal{E}_{pqij} g_{ij}(a) + \cdots \]  

(30)

with the same scattering \( \mathcal{E} \) between the \( g \) factors.

Let us end by stressing that the scattering of two excitons from \((i, j)\) to \((m, n)\) states also contains a purely Pauli contribution \( X_{mnij}(a) \), i.e. a contribution which does not contain any \( \xi \) Coulomb interaction at all (see Eqs (24-26)). This is conceptually quite new and very specific to close-to-boson particles, which feel each other through Pauli exclusion between their components, quite independently from any Coulomb scattering.

### 3 Conclusion

By using our commutation technique, we have derived the exact Coulomb expansion of the \( 1/(a - H) \) matrix elements in the two-exciton subspace. From the obtained expression of the exciton-exciton correlations, we show that the exchange processes appear in such a subtle way in the sequence of direct Coulomb scatterings, that there is no hope for a naive effective Coulomb interaction between boson-excitons to take into account their composite nature properly.
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(5) Higher order terms like $BBBB^\dagger B^\dagger B^\dagger$ are sometimes mentioned in this effective hamiltonian. It is however clear that they cannot help for the point raised here, as we consider matrix elements in the two-exciton subspace, so that all these higher order terms would give zero in this subspace.

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