Commutation Technique
for interacting close-to-boson excitons

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Abstract

The correct treatment of the close-to-boson character of excitons is known to be a major problem. In a previous work, we have proposed a “commutation technique” to include this close-to-boson character in their interactions. We here extend this technique to excitons with spin degrees of freedom as they are of crucial importance for many physical effects. Although the exciton total angular momentum may appear rather appealing at first, we show that the electron and hole angular momenta are much more appropriate when dealing with scattering processes. As an application of this commutation technique to a specific problem, we reconsider a previous calculation of the exciton-exciton scattering rate and show that the proposed quantity is intrinsically incorrect for fundamental reasons linked to the fermionic nature of the excitons.

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Non-linear effects in the optical properties of semiconductors have received considerable interest, both experimentally and theoretically. In these non-linearities, the interactions between carriers play a crucial rôle. Up to now, two types of theoretical methods have been proposed to deal with these interactions.

One method starts with the exact semiconductor Hamiltonian written in terms of the free electron and free hole fermion operators $a_k$ and $b_k$, and ends with the so-called semiconductor Bloch equations (1), or better, with an elaborate hierarchy of $2n$-point density matrices (2−5) which are then dynamically truncated to a given order in the radiation field. These approaches basically lead to a set of coupled equations for the time evolution of some expectation values of these *free electron and free hole* fermion operators. As the Bloch equations originally rely on the Hartree Fock approximation, various extensions have been proposed to include some correlation effects. In particular, it is possible to recover the low excitation regime, in which the free electron-hole pairs give rise to exciton resonances, by dropping all non-linear terms denoted as exchange processes (1). However, being written in terms of free electron-hole pairs, these procedures are obviously appropriate to systems well represented by *free* pairs. Even if various correlation effects can be included, these methods are mainly suitable at high density, when the screening is such that the exact correlations making the excitons are not crucial. For dilute electron-hole plasma however, these approaches can appear as starting from the “wrong” side, in the sense that, the excitons being the proper states at low density, they not immediately appear as the zero order terms. It is a priori rather subtle to extend these procedures *with complete security* to the low density regime in which the excitons play the dominant rôle.

The other method seems much more appealing at first in the low density regime, since it relies on the fact that, in this limit, the electrons and the holes are bound into excitons; as these excitons are made of two fermions, they very much look like bosons due to the spin statistics theorem. This idea leads to the so-called bosonic method (6), in which the exact excitons are replaced by boson-excitons and the exact Hamiltonian by an effective Hamiltonian $H_{\text{eff}}$ in which appear boson-exciton operators. In addition to an obvious free exciton energy part, this effective Hamiltonian must contain the interactions between excitons which are dominant at low density. Besides a “direct” term, which corresponds to
Coulomb interactions between excitons made with the same couples of electrons and holes, \((e, h)\) and \((e', h')\), it has been shown\(^{(7)}\) that the interaction part of \(H_{\text{eff}}\) also contains an “exchange” Coulomb term which corresponds to Coulomb interactions between excitons made with different couples of electrons and holes, \((e, h)\) and \((e', h')\) on one side, while \((e, h')\) and \((e', h)\) on the other side. It is said that this exchange term has to be introduced in order to take into account the fermionic character of the excitons, i. e. the fact that the excitons are made of two fermions which can be coupled in different ways. We have been amazed to realize\(^{(8)}\) that this exchange term, quoted by everyone for 25 years, is incorrect since it induces a non-hermitian part in the effective exciton hamiltonian. Even if this dramatic error is fixed, which is always possible at least intuitively, the major trouble with this bosonic approach actually comes from the fact that it misses purely fermionic terms\(^{(9)}\): Indeed the fermionic character of the exciton does not appear through Coulomb exchange terms only.

This fermionic aspect is indeed very subtle, and it is necessary to use a “full-proof” procedure to derive all its consequences properly. The previous approaches, which all\(^{(10)}\) end with incorrect results, were too intuitive to pick up all the consequences of this fermionic aspect properly.

In short, we can say that :

(i) On one hand, the semiconductor Bloch equations and their extension through 2n-point density matrices use “clean” free electron and free hole fermion operators. They have however problems with Coulomb interaction and its resulting correlations, which suffer from truncation procedures (even if they are somehow “controlled”). Indeed this Coulomb interaction must be included exactly in order to properly handle the evolution of the exciton bound states, i. e. the poles in the response function, which dominate at small density.

(ii) On the other hand, the bosonic method introduces the exact exciton states a priori, but uses “too clean” boson operators to represent these excitons, so that it misses some important consequences of their underlying fermionic structure.

What we would really like to do is to work with exact excitons a priori, i. e. electron-hole pairs in which the Coulomb interaction responsible for the bound states is put \textit{exactly}, while we properly handle the fact that these excitons are not real bosons. This is this
approach we are now proposing. It has already been briefly reported in ref. (8), without
the spin degrees of freedom for simplicity.

The paper is organized as follows:

In part I, we recall some very basic results on excitons in semiconductors to settle the
notations.

In part II, we introduce the Coulomb creation operator $V_i^\dagger$ between the exciton $i$ and
the rest of the system and show how we can get out of it a direct Coulomb scattering
$\xi_{\text{dir}}^{lnij}$.

In part III, we introduce the boson-departure operator $D_{ij}$ and the bare exchange
coefficient $\lambda_{\text{lnij}}$ associated to it.

In part IV, we show how we can produce various Coulomb exchange scatterings out of
$\xi_{\text{dir}}^{lnij}$ and $\lambda_{\text{lnij}}$.

In part V, we use our commutation technique to calculate the matrix elements of the
exact semiconductor Hamiltonian between two-exciton states.

In part VI, we reconsider a previous approach (11) to the exciton-exciton scattering
rate in which enter these $H$ matrix elements and show why it cannot be correct.

1 Exact semiconductor hamiltonian and one-exciton states

The semiconductor conduction electrons correspond to $S$ states with a $s = \pm 1/2$ spin
along an arbitrary $z$ direction. Let us call $a_{k,s}^\dagger$ the creation operator for such a free
conduction electron with momentum $k$ and spin $s$. For the free valence electrons, the
situation is much more complex: According to the Kohn-Luttinger representation (12),
the valence energy matrix (13) is given by

$$H_k = Ak^2 + B(k.I_{3/2})^2,$$

if one neglects warping, $I_{3/2}$ being the $3/2$ angular momentum matrix. The $H_k$
diagonalization, which is done by taking the $z$ axis along the $k$ direction, generates the so-called
light and heavy valence electrons. If we now add Coulomb interaction, we have shown (14)
that this Coulomb interaction is not diagonal between heavy and light valence electrons,
a fact which seriously complicates the problem since it mixes the $m = \pm 3/2$ with the $m = \pm 1/2$ valence states. In quantum wells, one can forget about this complexity because the $\pm 3/2$ and $\pm 1/2$ valence states having different hole masses, they are shifted differently by the confinement, so that the heavy-light hole Coulomb coupling usually gives a negligible contribution due to the splitting energy induced by the confinement, which appears in denominators. In bulk material however, the only consistent way to forget about these heavy-light hole Coulomb couplings is to assume $B = 0$, which corresponds to take all the valence electrons with the same mass. In doing so, $H_k$ is diagonal for any $k$ direction, so that the valence electron states are simply characterized by a quantum number $m = \pm 3/2, \pm 1/2$, along a direction which is now arbitrary. Let us call $b_{k,m}^\dagger$ the creation operator for such a free hole with momentum $k$ and angular momentum $m$.

The exact electron-hole semiconductor hamiltonian then reads

$$H = H_0 + V_{\text{eh}} + V_{\text{ee}} + V_{\text{hh}},$$

where

$$H_0 = \sum_{k,s} (\Delta + \epsilon_k^e) a_{k,s}^\dagger a_{k,s} + \sum_{k,m} \epsilon_k^h b_{k,m}^\dagger b_{k,m}, \quad \epsilon_k^e = \frac{\hbar^2 k^2}{2m_e}, \quad \epsilon_k^h = \frac{\hbar^2 k^2}{2m_h},$$

$\Delta$ being the band gap. For quantum wells, $m = \pm 3/2$, while for bulk materials $m = \pm 3/2, \pm 1/2$, as a unique hole mass $m_h$ is then assumed for the two hole bands. Within this approximation, the Coulomb scattering of a hole is diagonal with respect to its “spin” $m$, so that a hole $(k,m)$ is scattered into $(k + q, m)$ with the same $m$. The hole-hole, electron-hole and electron-electron interactions thus read

$$V_{\text{hh}} = \frac{1}{2} \sum_{q \neq 0, k,k',m,m'} V_q b_{k+q,m}^\dagger b_{k'-q,m'}^\dagger b_{k',m'} b_{k,m},$$

$$V_{\text{eh}} = - \sum_{q \neq 0, k,k',s,m} V_q a_{k+q,s}^\dagger b_{k'-q,m}^\dagger b_{k',m} a_{k,s},$$

$$V_{\text{ee}} = \frac{1}{2} \sum_{q \neq 0, k,k',s,s'} V_q a_{k+q,s}^\dagger a_{k'-q,s'}^\dagger a_{k',s'} a_{k,s},$$

where $V_q = \frac{4\pi e^2}{Vq}$ in 3D and $V_q = \frac{2\pi e^2}{Sq}$ in 2D, with $V$ being the sample volume and $S$ the well area.

From the free electron and free hole operators $a_{k,s}^\dagger$ and $b_{k,m}^\dagger$, one can construct the exciton operators $B_i^\dagger$ as

$$B_i^\dagger \equiv B_{\nu_i,Q_i,s_i,m_i}^\dagger = \sum_{k_i} \langle k_i|x_{\nu_i}\rangle a_{k_i,s_i}^\dagger b_{k_i,m_i}^\dagger,$$
where we have set
\[ K^e_i = k_i + \alpha_e Q_i, \quad K^h_i = -k_i + \alpha_h Q_i, \quad \alpha_{e,h} = \frac{m_{e,h}}{m_e + m_h}. \] (8)

\( Q_i \) and \( \nu_i \) are the \( i \) exciton center of mass momentum and relative motion quantum parameter: In 3D, \( \nu_i \) corresponds to the quantum numbers \((n_i, l_i, m_i)\), while in 2D it corresponds to two quantum numbers only \((n_i, m_i)\). \( \langle k|\nu_i \rangle \) is the \( i \) exciton relative motion wave function in \( k \) space. In a similar way, it is possible to write the free electron-hole pair operator in terms of exciton operators:
\[ a^\dagger_{k, s_i} b^\dagger_{k', m_i} = \sum_{\nu_i} \langle x_{\nu_i}|\alpha_h k_e - \alpha_e k_h \rangle B^\dagger_{\nu_i, k_e + k_h, s_i, m_i}, \] (9)
as checked by inserting Eq (9) into Eq (7). Let us stress that this sum contains the bound states as well as the diffusive states.

The one-exciton state \( B^\dagger_i |v \rangle \), where \( |v \rangle \) is the electron-hole vacuum state, is eigenstate of \( H \). Using Eqs (2-9), and the following Schrödinger equation for the electron-hole relative motion in \( k \) space,
\[ (\epsilon^e_k + \epsilon^h_k)\langle k|x_{\nu_i} \rangle - \sum_{q \neq 0} V_q \langle k+q|x_{\nu_i} \rangle = \epsilon_{\nu_i} \langle k|x_{\nu_i} \rangle, \] (10)
where \( \epsilon_{\nu_i} \) is the energy of the relative motion state \( |x_{\nu_i} \rangle \), it is indeed easy to check that
\[ HB^\dagger_i |v \rangle = (H_0 + V_{eh})B^\dagger_i |v \rangle = E_i B^\dagger_i |v \rangle, \quad E_i = \Delta + \epsilon_{\nu_i} + \frac{\hbar^2 Q_i^2}{2(m_e + m_h)}. \] (11)

Let us again stress that in 3D this result strongly relies on the fact that the Coulomb interaction is diagonal between holes, i.e. it does not contain terms in \( b^\dagger_{k'-q,m'} b_{k',m} \) with \( m' \neq m \).

In many problems dealing with excitons, the photons play an important rôle. As a \( \sigma^+ \) photon with spin \((J = 1, M = \pm 1)\) creates an exciton with the same total angular momentum, it may appear appropriate to use, instead of the \((s_i, m_i)\) angular momentum variables of the separate electron and hole, the \((J_i, M_i)\) total angular momentum variables of the exciton, with \((J_i = 2, M_i = \pm 2, \pm 1, 0)\) or \((J_i = 1, M_i = \pm 1, 0)\). One can easily go from one set of operators to the other by
\[ B^\dagger_{\nu_i, Q_i, J_i, M_i} = \sum_{s_i, m_i} \langle s_i, m_i|J_i, M_i \rangle B^\dagger_{\nu_i, Q_i, s_i, m_i}, \] (12)
\[ B^\dagger_{\nu_i, Q_i, J_i, M_i} = \sum_{J_i, M_i} \langle J_i, M_i|s_i, m_i \rangle B^\dagger_{\nu_i, Q_i, J_i, M_i}. \] (13)
Only two of these $B_{\nu_i, \mathbf{Q}_i, J_i, M_i}^\dagger$ states are coupled to light, namely $(J_i = 1, M_i = \pm 1)$, while the six other exciton states correspond to the so-called “dark” excitons. However, even if these dark exciton states are not coupled to light, they are generated by Coulomb scatterings so that we must keep these 8 operators $B_{\nu_i, \mathbf{Q}_i, J_i, M_i}^\dagger$ anyway in order to possibly describe the exciton-exciton scatterings properly. Moreover, as the Coulomb interaction is diagonal within the $(s, m)$ quantum numbers, but not within the $(J, M)$’s, it is in fact far simpler to work with the 8 operators $B_{\nu, \mathbf{Q}, s, m}^\dagger$ for all processes dealing with Coulomb interactions, and just at the beginning and the end of the calculations, use Eqs (12-13) to transform these $B_{\nu, \mathbf{Q}, s, m}^\dagger$ operators into the $B_{\nu, \mathbf{Q}, J, M}^\dagger$ operators coupled to the light. This is why all over this work, we will use the $B_{\nu, \mathbf{Q}, s, m}^\dagger$ operators only.

2 Coulomb creation operator $V_{i}^\dagger$ and direct Coulomb scattering $\xi_{lnij}^{\text{dir}}$

In standard problems with interactions, one usually divides the system hamiltonian $H$ into $H_0 + V$, where $H_0$ is the so-called “non-interacting” part, i.e. the part of $H$ which can be diagonalized, while $V$ is the interaction part, i.e. the part of $H$ which cannot be handled exactly, but is hoped to be small enough to be treated as a perturbation. In addition, when $H$ can be written as $H_0 + V$, the $H_0$ eigenstates form an orthogonal basis which can be used to expand any state of the system.

For interacting excitons, even if we could guess that the energies $E_i$ of individual excitons should appear in the “non-interacting” part of an hypothetical exciton hamiltonian, one cannot divide the electron-hole Coulomb interaction given in Eq (5), into a part which would bind a specific electron to a specific hole to form the $i$ exciton, and a “rest” which would make this exciton to interact with other excitons: The electrons, as well as the holes, being indistinguishable particles, such a formal separation is indeed impossible.

A separation, similar in its spirit to the splitting $H = H_0 + V$, is nevertheless possible through our commutation technique. If $H$ is the exact semiconductor hamiltonian, given in Eqs (2-6), and if $B_i^\dagger$ is the exact exciton creation operator, given in Eq (7), we find that their commutator reads

$$\left[H, B_i^\dagger\right] = E_i B_i^\dagger + V_i^\dagger,$$

(14)
where $V_i^\dagger$ is given by

$$V_i^\dagger = \sum_{q \neq 0, \nu} V_q \gamma_{li}(q) B_{\nu_i, q, s_i, m_i}^\dagger W_{-q}.$$  
$$W_{-q} = \sum_{p, s} a_{p, q, s}^\dagger a_{p, s} - \sum_{p, m} b_{p, q, m}^\dagger b_{p, m}. \quad (15)$$

The coefficient $\gamma_{li}(q)$, characterizes the scattering of a $\nu_i$ exciton into a $\nu_l$ state under a $q$ Coulomb excitation. Using the following relation,

$$\sum_{k} \langle x_{\nu_i} | k + \alpha q | x_{\nu_i} \rangle = \langle x_{\nu_i} | e^{i\alpha q} r | x_{\nu_i} \rangle,$$  
$$\quad (16)$$

it is easy to show that it is given by

$$\gamma_{li}(q) = \langle x_{\nu_l} | e^{i\alpha q} r - e^{-i\alpha q} r | x_{\nu_i} \rangle.$$  
$$\quad (17)$$

The analogy with the usual separation $H = H_0 + V$ is quite transparent if we note that Eq (14) also reads

$$HB_i^\dagger = B_i^\dagger (H + E_i) + V_i^\dagger.$$  
$$\quad (18)$$

By considering the state $B_i^\dagger | \phi \rangle$, where $| \phi \rangle$ is any electron-hole state, Eq (18) leads to

$$HB_i^\dagger | \phi \rangle = E_i B_i^\dagger | \phi \rangle + B_i^\dagger H | \phi \rangle + V_i^\dagger | \phi \rangle.$$  

In the first term, the contribution of the $i$ exciton to the energy of the system is just $E_i$ as if this exciton were not interacting with the other electrons or holes of $| \phi \rangle$. The second term corresponds to $H$ acting on $| \phi \rangle$ independently of the presence of the $i$ exciton: The operator $(H + E_i)$, on the right hand side of $B_i^\dagger$, thus plays the rôle of the $H_0$ part of the hamiltonian for usual problems in which $H$ can be written as $H_0 + V$. The third term is there because the $i$ exciton does in fact interact with $| \phi \rangle$. This operator $V_i^\dagger$ thus describes all possible Coulomb interactions between the $i$ exciton and the rest of the system. It has to be seen as the formal equivalent of $V$ in $H = H_0 + V$. It is however important to stress that, while the usual $V$’s conserve particles, i.e. contain the same number of creation operators $a^\dagger$ and destruction operators $a$, the operator $V_i^\dagger$ is not a real potential in the sense that it contains one additional electron-hole pair creation operator $B^\dagger$ (see Eq (15)). This is why we call it Coulomb creation operator. In addition, usual two-body potentials have prefactors which depend on four indices which are the ones of two initial and two
final states. We here see that the prefactors appearing in $V_i^\dagger$ depend on two indices only, $l$ and $i$.

In order to cope with these difficulties and generate a “scattering” which depends on four indices, we can push the commutation technique one step further and calculate $[V_i^\dagger, B_j^\dagger]$. Using Eqs (7-9) and (15), we find

$$[V_i^\dagger, B_j^\dagger] = \sum_{q \neq 0, \nu_i, \nu_n} V_q \gamma_{li}(q) \gamma_{nj}(-q) B_{\nu_i, Q_l+q, s_i, m_i}^\dagger B_{\nu_n, Q_j-q, s_j, m_j}^\dagger,$$

which formally reads

$$[V_i^\dagger, B_j^\dagger] = \sum_{l,n} \xi_{\text{dir}}^{\dagger} B_l^\dagger B_n^\dagger.$$

If we compare Eq (19) to Eq (20), and symmetrize the result with respect to $(l, n)$ (which will appear convenient afterwards), we find that the direct Coulomb scattering $\xi_{\text{dir}}^{\dagger}$ can be written as

$$\xi_{\text{dir}}^{\dagger} = \xi_{\text{dir}}^{\dagger} = \xi_{\text{dir}}^{\dagger} = \frac{1}{2} \left[ \Delta^{\text{dir}}(l\ i\ n\ j) \hat{\xi}^{\text{dir}}(l\ i\ n\ j) + (l \leftrightarrow n) \right].$$

It contains an angular momentum contribution and an orbital contribution which appear as two independent factors. For direct processes, this angular momentum contribution,

$$\Delta^{\text{dir}}(l\ i\ n\ j) = \delta_{s_i, s_j} \delta_{m_i, m_j} \delta_{s_n, s_j} \delta_{m_n, m_j},$$

just says that the $l$ exciton has the same electron and hole momenta as the $i$ exciton and similarly for the $n$ and $j$ excitons. Let us stress that this angular momentum part would be much more complicated if the $(J, M)$ variables for the exciton total angular momentum were used; this is why it is indeed appropriate to keep these $(s, m)$ variables as long as we deal with Coulomb scatterings even if they are not the good variables for semiconductor-photon interaction.

The orbital part of this direct Coulomb scattering is given by

$$\hat{\xi}^{\text{dir}}(l\ i\ n\ j) = \delta_{Q_l+Q_i, Q_l+Q_j} V_{Q_l-Q_i} \gamma_{li}(Q_l - Q_i) \gamma_{nj}(Q_n - Q_j).$$

It of course contains the fact that the center of mass momenta have to be conserved, $Q_i + Q_j = Q_l + Q_n$, in the scattering. It also contains the two factors which characterize the scatterings of one exciton from a $\nu_i$ state to a $\nu_l$ state and the other exciton from a $\nu_j$ state to a $\nu_n$ state, under the Coulomb excitation $Q_l - Q_i = -(Q_n - Q_j)$. 
The appendix A contains the explicit calculation of these $\gamma_{li}(q)$ factors for the lowest S states.

Even if the above expression of $\hat{\xi}_{\text{dir}}(l_i n_j)$ is quite transparent and extremely convenient for explicit calculations, it will appear useful, when we will generate exchange Coulomb processes, to note that this direct Coulomb scattering is also equal to

$$
\hat{\xi}_{\text{dir}}(l_i n_j) = \sum_{k_i, k_n, k_l, k_j} \langle x_{\nu_i} | k_l \rangle \langle x_{\nu_n} | k_n \rangle \langle k_l | x_{\nu_j} \rangle \langle k_j | x_{\nu_i} \rangle 
\times \sum_{q \neq 0} V_q \left[ \delta_{k_i, k_l} \delta_{k_n, K} - (e \leftrightarrow h) \right] \left[ \delta_{k_i, k_l} \delta_{K, k_n} - (e \leftrightarrow h) \right].
$$

In appendix B, we show that this $\hat{\xi}_{\text{dir}}(l_i n_j)$ is nothing but the direct part of the Coulomb interaction widely quoted (7,15) in the effective boson exciton hamiltonian, namely

$$
\hat{\xi}_{\text{dir}}(l_i n_j) = \int d\mathbf{r}_e d\mathbf{r}_h d\mathbf{r}_{e'} d\mathbf{r}_{h'} \phi_i^*(\mathbf{r}_e, \mathbf{r}_h) \phi_n^*(\mathbf{r}_{e'}, \mathbf{r}_{h'}) 
\times [V_{ee'} + V_{hh'} - V_{eh} - V_{e'h}] \phi_i(\mathbf{r}_e, \mathbf{r}_h) \phi_j(\mathbf{r}_{e'}, \mathbf{r}_{h'}) = \left[ \hat{\xi}_{\text{dir}}(l_i n_j) \right]^*,
$$

where $\phi_i(\mathbf{r}_e, \mathbf{r}_h)$ is the whole wave function of the $i = (\nu_i; Q_i)$ exciton,

$$
\langle \mathbf{r}_e, \mathbf{r}_h | B_i^\dagger | \nu \rangle = \phi_i(\mathbf{r}_e, \mathbf{r}_h) = \frac{1}{\sqrt{V}} e^{Q_i (\alpha_e \mathbf{r}_e + \alpha_h \mathbf{r}_h)} \langle \mathbf{r}_e - \mathbf{r}_h | x_{\nu_i} \rangle.
$$

By these two successive commutators $[H, B_i^\dagger]$ and $[V_i^\dagger, B_j^\dagger]$, we have found a formal way to generate a scattering of two excitons $(i, j)$ into two other excitons $(l, n)$. If we look at its expression given in Eq (25), we see that the coefficient $\hat{\xi}_{\text{dir}}(l_i n_j)$ obtained by this procedure turns out to have a very simple physical meaning: It corresponds to all electron-electron, hole-hole and electron-hole Coulomb interactions between $(i, j)$ and $(l, n)$, when the initial and final excitons are made with the same electron-hole pairs $(e, h)$ and $(e', h')$.

The scattering processes corresponding to $\hat{\xi}_{\text{dir}}(l_i n_j)$ are shown in Fig. (1): One of the electrons, or holes, suffers a $(Q_l - Q_i)$ Coulomb excitation, while another particle has a $-(Q_l - Q_i)$ excitation, the excitons before and after the scattering being made with the \emph{same} electron-hole pairs. This is why we call it a \textit{direct} Coulomb scattering, by contrast with the exchange Coulomb scatterings which will appear below, in which the excitons before and after scattering are made with different pairs.
3 Boson departure operator $D_{ij}$ and bare exchange coefficient $\lambda_{lnij}$

Even if the above procedure allows to formally extract from the whole electron-hole interaction the part which corresponds to a Coulomb interaction between excitons, such a formal extraction seeming not obvious at first, we are far from having picked all the physics which controls the interactions between excitons. Another rather subtle origin of these interactions comes from the fermionic character of the excitons. From handwaving arguments, we can say that two excitons feel each other even in the absence of any Coulomb interaction, because they are made of two fermions and the fermions of two excitons must be in different states. This condition in itself produces a “link” between excitons. There is no need of Coulomb forces. It is thus very likely that the exciton-exciton interaction has to contain a purely fermionic contribution, quite different from the Coulomb interaction dressed by exchange processes as thought by everyone up to now.

In this quite tricky determination of the correct exciton-exciton interaction, it is however highly necessary to formalize the above handwaving argument. We want to find a coefficient, which depends on four exciton indices ($lnij$), and which originates from the fermionic character of the excitons only. This coefficient has of course to be linked to the fact that the excitons are not real bosons. The most direct way to have this property appearing, is to start with the commutator of two (exact) exciton creation operators. It leads to the boson departure operator $D_{ij}$ defined as

$$D_{ij} = \delta_{ij} - \left[ B_i, B_j^\dagger \right]. \quad (27)$$

Using Eq (7) and the standard commutation rules for the fermion operators $a^\dagger$ and $b^\dagger$, we find that this operator reads

$$D_{ij} = \sum_{\kappa_i, \kappa_j} \langle x_{\nu_i} | k_i \rangle \langle k_j | x_{\nu_j} \rangle \left[ \delta_{m_i, m_j} \delta_{\kappa_i^h, \kappa_j^h} a_{\kappa_i^s, s_j}^\dagger a_{\kappa_j^s, s_i} + \delta_{s_i, s_j} \delta_{\kappa_i^h, \kappa_j^e} b_{\kappa_i^h, m_j}^\dagger b_{\kappa_j^h, m_i} \right]. \quad (28)$$

$D_{ij} \equiv 0$ if we have both $s_i \neq s_j$ and $m_i \neq m_j$.

From this boson departure operator, we can construct a four index coefficient by taking the commutator of this $D_{ij}$ operator with an exciton operator. Using Eqs (7,9,28), we
where the bare exchange coefficient \( \lambda_{n_{l_{i}j}} \) can here again be split into an angular momentum part and an orbital part:

\[
\lambda_{n_{l_{i}j}} = \lambda_{n_{l_{i}j}} = \frac{1}{2} \left[ \Delta^{\text{exch}} \left( \frac{i}{n} \right) \hat{\lambda} \left( \frac{i}{n} \right) + (l \leftrightarrow n) \right].
\] (30)

The angular momentum part,

\[
\Delta^{\text{exch}} \left( \frac{i}{n} \right) = \delta_{s_{l_{i}},s_{l}} \delta_{m_{i},m_{j}} \delta_{s_{n_{l}},s_{n}} \delta_{m_{n},m_{i}},
\] (31)

just says that the spins of the \( l \) and \( i \) electrons are the same while the angular momentum of the \( l \) hole is not the one of the \( i \) hole, as in \( \Delta^{\text{dir}} \left( \frac{i}{n} \right) \), but the one of the \( j \) hole. The orbital part appears as

\[
\hat{\lambda} \left( \frac{i}{n} \right) = \sum_{k_{l_{i}},k_{n_{i}}} \langle x_{\nu_{l_{i}}} | k_{l_{i}} \rangle \langle k_{l_{i}} | x_{\nu_{i}} \rangle \langle k_{l_{i}} | x_{\nu} \rangle \delta_{K_{l_{i}},K_{l}} \delta_{K_{n_{i}},K_{n}} \delta_{K_{l},K_{h_{l}}} \delta_{K_{n},K_{h_{n}}}. \] (32)

In appendix B, we show that this orbital part \( \hat{\lambda} \left( \frac{i}{n} \right) \) can be rewritten in \( r \) space as

\[
\hat{\lambda} \left( \frac{i}{n} \right) = \int dr_{e} dr_{h} dr_{e'} dr_{h'} \phi_{i}^{*}(r_{e},r_{h}) \phi_{n}^{*}(r_{e'},r_{h'}) \phi_{i}(r_{e},r_{h'}) \phi_{j}(r_{e'},r_{h}) = \left[ \hat{\lambda} \left( \frac{i}{n} \right) \right]^{*}. \] (33)

From the above expression, we see that, in this orbital part, the electrons and the holes forming the excitons are exchanged, the \( l \) and \( i \) excitons being made with the same electron but different holes. Note that the \( (l \leftrightarrow n) \) term of Eq (30) restores the \((e,h)\) symmetry in \( \lambda_{n_{l_{i}j}} \), which is broken in \( \hat{\lambda} \left( \frac{i}{n} \right) \).

Eq (33) shows in a transparent way that this \( \hat{\lambda} \left( \frac{i}{n} \right) \) exchange coefficient can exist because of the composite nature of the excitons which can be formed in different ways, the electrons and the holes being indistinguishable fermions. An equally transparent link between this coefficient and the composite nature of the excitons can be obtained by considering the two-exciton operator \( B_{i}^{\dagger} B_{j}^{\dagger} \), each \( B^{\dagger} \) being given by Eq (7), and by binding the electrons and holes of these two \( B^{\dagger} \)'s in a different way through Eq (9). This gives

\[
B_{i}^{\dagger} B_{j}^{\dagger} = - \sum_{l,n} \lambda_{n_{l_{i}j}} B_{l}^{\dagger} B_{n}^{\dagger}, \] (34)

with the same prefactor \( \lambda_{n_{l_{i}j}} \) as the one of Eq (29).
The electron-hole exchange appearing in the first term of Eq (30) is shown in Fig. (2). It corresponds to cross the holes when forming the \((i, j)\) or \((l, n)\) excitons: It is indeed a bare exchange process in the sense that it does not contain any Coulomb interaction, by contrast with the Coulomb exchange processes which will appear later on.

The four \(\delta\)’s appearing in \(\hat{\lambda}(l_{i,n})\) given in Eq (32) impose the expected exciton total momentum conservation \(Q_i + Q_j = Q_l + Q_n\) as well as \(k_i + k_j = k_l + k_n\). By expliciting these four \(\delta\)’s, we can rewrite \(\hat{\lambda}(l_{i,n})\) in a compact form,

\[
\hat{\lambda}(l_{i,n}) = \delta_{Q_l + Q_n, Q_i + Q_j} F_{\text{lnij}}(\alpha_e(Q_l - Q_i), \alpha_h(Q_n - Q_l)) ,
\]

(35)

where \(F_{\text{lnij}}(p, p')\) is a sum over one momentum only,

\[
F_{\text{lnij}}(p, p') = \sum_k \langle x_{\nu_l} | k - \frac{p + p'}{2} \rangle \langle x_{\nu_n} | k + \frac{p + p'}{2} \rangle \langle k + \frac{p - p'}{2} | x_{\nu_i} \rangle \langle k - \frac{p - p'}{2} | x_{\nu_j} \rangle .
\]

(36)

Although less transparent for physical understanding, the above expression of \(\hat{\lambda}(l_{i,n})\) is more convenient for explicit calculations. When the four excitons are in S states, the function \(F_{\text{lnij}}(p, p')\) a priori depends on three independent parameters, \(p, p'\) and the angle \(\theta\) between \(p\) and \(p'\). Some interesting values of these parameters are:

(i) \(p = 0\) or \(p' = 0\), which corresponds to \(Q_i = Q_i\) or \(Q_n = Q_i\): The final excitons have the same momenta as the initial excitons;

(ii) \(\theta = 0\) or \(\theta = \pi\), which corresponds to \(p\) parallel to \(p'\). \(Q_l - Q_i\) is parallel to \(Q_n - Q_i\) \(\equiv Q_j - Q_i\) in the particular case of \(Q_i = Q_j\) or \(Q_l = Q_n\), i.e. when the two initial or the two final excitons have the same momentum. In this case, the two independent parameters can be taken either as \(\alpha_e|Q_l - Q_i|\) and \(\alpha_h|Q_n - Q_i|\), or better as the momentum transfer \(|Q_l - Q_i|\) and the mass ratio \(m_e/m_h\).

In appendix C, we have calculated \(F_{\text{lnij}}(p, p')\) when all the \(\nu\)'s are equal to the 1S ground state and \(Q_i = Q_j = Q_l = Q_n\). We get

\[
F_{1s1s1s1s}(0, 0) = \frac{(33\pi/2)a_x^3}{\mathcal{V}} \quad \text{in 3D}
\]

\[
= \frac{(4\pi/5)a_x^2}{S} \quad \text{in 2D} ,
\]

(37)

\(a_x\) being the 3D exciton Bohr radius.

We have also numerically computed \(\hat{\lambda}(l_{i,n})\) in the 2D case, when \(\nu_l = \nu_n = \nu_i = \nu_j = 1S\), \(Q_i = Q_j = 0\), \(Q_l = -Q_n = p\). Fig. (3) shows

\[
\Lambda(p) = \frac{S}{\lambda_{2D}^2} \hat{\lambda}(\frac{p}{p}, 0) ,
\]

(38)
for three values of \(m_e/m_h\), as a function of \(P = p\lambda_{2D}\), with \(\lambda_{2D} = a_x/2\) being the 2D Bohr radius. We see that \(\Lambda(p)\) weakly depends on the mass ratio, and that it is significant for values of \(p\) such that \(p\lambda_{2D} < 4\) only.

4 Coulomb exchange scatterings

From Eq (33), we see that the \(\lambda_{\text{lnij}}\) coefficient is dimensionless. So that it has to be “cooked” with quantities homogeneous to an energy in order to possibly appear in an exciton-exciton scattering. From the two energy-like quantities we have yet found in this problem, namely the bare energies of the excitons \(E_i\) and the direct Coulomb scattering \(\xi_{\text{lnij}}^{\text{dir}}\), there are of course various ways to construct a \(\lambda_{\text{lnij}}\) dependent scattering. In the next section, we will show that two specific combinations of the bare exchange coefficient \(\lambda_{\text{lnij}}\) and the direct Coulomb scattering \(\xi_{\text{lnij}}^{\text{dir}}\) appear in a natural way, namely

\[
\xi_{\text{lnij}}^{\text{right}} = \sum_{p,r} \xi_{\text{lnipr}}^{\text{dir}} \lambda_{\text{prij}} ,
\]

\[
\xi_{\text{lnij}}^{\text{left}} = \sum_{p,r} \lambda_{\text{lnipr}} \xi_{\text{prij}}^{\text{dir}} .
\]

They are shown in Fig. (4).

The simplest way to calculate these sums is to use Eqs (30-32) for \(\lambda_{\text{lnij}}\) and Eqs (21,22,24) for \(\xi_{\text{lnij}}^{\text{dir}}\). We find

\[
\xi_{\text{lnij}}^{\text{right}} = \frac{1}{2} \left( \Delta_{\text{exch}}^{\text{right}} \left( \frac{l_i}{n_j} \right) \right) \hat{\xi}_{\text{lnij}}^{\text{right}} \left( \frac{l_i}{n_j} \right) ,
\]

where the angular momentum conservation part \(\Delta_{\text{exch}}^{\text{right}} \left( \frac{l_i}{n_j} \right)\) is just the one appearing in the bare exchange coefficient \(\lambda_{\text{lnij}}\) (Eq (31)), while the orbital part of this right exchange Coulomb term is given by

\[
\hat{\xi}_{\text{lnij}}^{\text{right}} \left( \frac{l_i}{n_j} \right) = \sum_{p,r} \hat{\xi}_{\text{lnipr}}^{\text{dir}} \lambda_{\text{prij}} \left( \frac{l_i}{n_j} \right) ,
\]

\[
\hat{\xi}_{\text{lnij}}^{\text{right}} \left( \frac{l_i}{n_j} \right) = \sum_{p,r} \lambda_{\text{lnipr}} \xi_{\text{prij}}^{\text{dir}},
\]

Note that the second term of each bracket does not correspond to \((e \leftrightarrow h)\) as for \(\hat{\xi}_{\text{lnij}}^{\text{dir}} \left( \frac{l_i}{n_j} \right)\) given in Eq (24).
The set of four $\delta$’s appearing in $\hat{\xi}^{\text{right}} \left( \frac{l}{n} \right)$ again impose the expected momentum conservation $Q_i + Q_j = Q_l + Q_n$. By expliciting these four $\delta$ functions, it is possible to rewrite $\hat{\xi}^{\text{right}} \left( \frac{l}{n} \right)$ in a more compact form in terms of sums over $q$ and one $k$ only. In addition to the function $F_{lnij}(p, p')$ appearing already in the bare exchange coefficient (Eq (36)), two other functions enter $\hat{\xi}^{\text{right}} \left( \frac{l}{n} \right)$. They are defined by

$$G^{(i)}_{lnij}(p, p'; q) = \sum_k \langle x_{\nu_l} | k - \frac{p + p'}{2} \rangle \langle x_{\nu_n} | k + \frac{p + p'}{2} \rangle \langle k + \frac{p - p'}{2} + q | x_{\nu_i} \rangle \langle k - \frac{p - p'}{2} + q | x_{\nu_j} \rangle ,$$

(43)

$$G^{(j)}_{lnij}(p, p'; q) = \sum_k \langle x_{\nu_l} | k - \frac{p + p'}{2} \rangle \langle x_{\nu_n} | k + \frac{p + p'}{2} \rangle \langle k + \frac{p - p'}{2} + q | x_{\nu_i} \rangle \langle k - \frac{p - p'}{2} + q | x_{\nu_j} \rangle .$$

(44)

(We can see that $F_{lnij}(p, p')$ is also either $G^{(i)}_{lnij}(p, p'; 0)$ or $G^{(j)}_{lnij}(p, p'; 0)$). In terms of these three functions, the right exchange Coulomb term reads

$$\hat{\xi}^{\text{right}} \left( \frac{l}{n} \right) = \delta_{Q_i + Q_n, Q_j + Q_l} \sum_{q \neq 0} V_q \times [F_{lnij} (\alpha_e(Q_l - Q_i) - q, \alpha_h(Q_n - Q_j)) + F_{lnij} (\alpha_e(Q_l - Q_j), \alpha_h(Q_n - Q_i) + q) - G^{(i)}_{lnij} (\alpha_e(Q_l - Q_i), \alpha_h(Q_n - Q_j); -q) - G^{(j)}_{lnij} (\alpha_e(Q_l - Q_j), \alpha_h(Q_n - Q_i); q)].$$

(45)

This $\hat{\xi}^{\text{right}} \left( \frac{l}{n} \right)$ coefficient can be rewritten in a form much more transparent for the physical understanding, although much less convenient for calculations. In appendix B, we do show that

$$\hat{\xi}^{\text{right}} \left( \frac{l}{n} \right) = \int dr_e dr_h dr_{e'} dr_{h'} \phi^*_l(r_e, r_h) \phi^*_n(r_{e'}, r_{h'}) \times [V_{ee'} + V_{hh'} - V_{eh'} - V_{e'h}] \phi_i(r_e, r_{h'}) \phi_j(r_{e'}, r_h) .$$

(46)

The physical meaning of this $\hat{\xi}^{\text{right}} \left( \frac{l}{n} \right)$ becomes now clear : It contains the electron-electron and hole-hole interactions between two excitons when these excitons are made with their electrons and holes coupled in a different way. With respect to the electron-hole interactions, the situation is however more subtle as these interactions are between the excitons on one side, but inside the excitons of the other side, due to the exchange of the electrons or holes making the excitons of the two sides. This $\hat{\xi}^{\text{right}} \left( \frac{l}{n} \right)$ turns out to be nothing but the exchange Coulomb interaction which appears in the exciton-exciton scattering coefficient of the effective boson exciton hamiltonian widely quoted (7).

The coefficient $\xi^{\text{left}}_{lnij}$ reads as $\xi^{\text{right}}_{lnij}$ except that $V_{eh} + V_{e'h}$ is replaced by $V_{eh} + V_{e'h}$ in Eq (46), so that the electron-hole Coulomb interactions are now between the $(i, j)$ excitons...
and inside the \((l, n)\) excitons. As a consequence, the two coefficients \(\xi^\text{left}\) and \(\xi^\text{right}\) verify

\[
\xi^\text{left}_{\text{lnij}} = \left[\xi^\text{right}_{\text{ijnl}}\right]^* ,
\]

with a similar relation between the orbital parts \(\hat{\xi}^\text{left}\) and \(\hat{\xi}^\text{right}\). This can also be seen from Eqs (25,33,39,40).

From \(\xi^\text{right}_{\text{lnij}}\) and \(\xi^\text{left}_{\text{lnij}}\), it is possible to construct a quantity symmetrical with respect to the electron-hole interactions, by taking

\[
\xi^\text{exch}_{\text{lnij}} = \frac{1}{2} (\xi^\text{right}_{\text{lnij}} + \xi^\text{left}_{\text{lnij}}) .
\]

Using Eq (46), the corresponding orbital part of this Coulomb exchange coefficient reads

\[
\hat{\xi}^\text{exch}_{\text{lnij}} = \int \, d\quad \phi^*_i(r_e, r_h) \phi^*_n(r_{e'}, r_{h'})
\]

\[
\times \left[ V_{ee'} + V_{hh'} - \frac{1}{2} (V_{eh} + V_{e'h'} + V_{eh'} + V_{e'h}) \phi_i(r_e, r_h') \phi_j(r_{e'}, r_h) \right].
\]

It contains the four possible electron-hole Coulomb interactions between two electrons and two holes. This \(\xi^\text{exch}_{\text{lnij}}\) could be a reasonable scattering for the interaction part of an hypothetical effective exciton hamiltonian: As \(\xi^\text{exch}_{\text{lnij}} = \left(\xi^\text{exch}_{\text{ijnl}}\right)^*\), its contribution would be hermitian, which is not the case neither for \(\xi^\text{right}_{\text{lnij}}\) nor for \(\xi^\text{left}_{\text{lnij}}\) alone.

## 5 Matrix element of \(H\) between two-exciton states

In some physical problems such as the one considered in section VI of this paper, we are led to consider the matrix elements of the exact hamiltonian \(H\) between two-exciton states, namely \(\langle v | B_l B_n H B_i^\dagger B_j^\dagger | v \rangle\). Using Eqs (14,20) and \(V_i^\dagger | v \rangle = 0\), as obvious from Eq (15), we find

\[
H B_i^\dagger B_j^\dagger | v \rangle = (E_i + E_j) B_i^\dagger B_j^\dagger | v \rangle + \sum_{p,r} \xi^\text{dir}_{\text{prij}} B_p^\dagger B_r^\dagger | v \rangle .
\]

So that this matrix element reads

\[
\langle v | B_l B_n H B_i^\dagger B_j^\dagger | v \rangle = (E_i + E_j) \langle v | B_l B_n B_i^\dagger B_j^\dagger | v \rangle + \sum_{p,r} \xi^\text{dir}_{\text{prij}} \langle v | B_l B_n B_p^\dagger B_r^\dagger | v \rangle .
\]

To go further, we must calculate \(\langle v | B_l B_n B_i^\dagger B_j^\dagger | v \rangle\). This is easily done using Eqs (27) and (29). Since \(D_{ij} | v \rangle = 0\), as obvious from Eq (28), we get

\[
\langle v | B_l B_n B_i^\dagger B_j^\dagger | v \rangle = \delta_{li} \delta_{nj} + \delta_{ni} \delta_{lj} - 2\lambda_{\text{lnij}} .
\]
The two first terms are naïve: They differ from zero for \((l, n) = (i, j)\), i.e. when the excitons on both sides are identical. The last term is more subtle. It has a fermionic origin: In addition to the fact that the coefficient \(\lambda_{lnij}\) is directly related to the boson deviation operator \(D_{li}\) through Eq (29), its fermionic origin can also be traced back to Eq (34) which says that a product of two exciton operators writes as a sum of products of any two other \(B_l^\dagger\)'s so that \(B_l^\dagger B_j^\dagger\) in fact contains a “piece” of any \(B_i^\dagger B_i^\dagger\), even if \((l, n) \neq (i, j)\). In other words, the \(B_l^\dagger B_j^\dagger |v\rangle\) states do not form an orthogonal basis for two-pair states.

By inserting Eq (52) into Eq (51), we get
\[
\langle v | B_l B_n H B_i^\dagger B_j^\dagger |v\rangle = (E_i + E_j)(\delta_{li} \delta_{nj} + \delta_{lj} \delta_{ni} - 2\lambda_{lnij}) + 2(\xi_{lnij}^{\text{dir}} - \xi_{lnij}^{\text{left}}) .
\] (53)

This calculation thus produces the Coulomb exchange term \(\xi_{lnij}^{\text{left}}\). A similar calculation done with \(H\) acting on the left gives
\[
\langle v | B_l B_n H B_i^\dagger B_j^\dagger |v\rangle = (E_l + E_n)(\delta_{li} \delta_{nj} + \delta_{lj} \delta_{ni} - 2\lambda_{lnij}) + 2(\xi_{lnij}^{\text{dir}} - \xi_{lnij}^{\text{right}}) .
\] (54)

Here appears \(\xi_{lnij}^{\text{right}}\). From these two expressions of the \(H\) matrix element, we deduce that \(\xi_{lnij}^{\text{right}}\) and \(\xi_{lnij}^{\text{left}}\) are linked by
\[
\xi_{lnij}^{\text{left}} - \xi_{lnij}^{\text{right}} = (E_l + E_n - E_i - E_j)\lambda_{lnij} ,
\] (55)
so that they are equal for \(E_l + E_n = E_i + E_j\) only.

It is possible to check that Eq (55) remains valid when \(\xi\) and \(\lambda\) are replaced by their orbital parts \(\hat{\xi}\) and \(\hat{\lambda}\). Using this equation, we can easily calculate the difference \(\hat{\xi}_{lnij}^{\text{left}} - \hat{\xi}_{lnij}^{\text{right}}\) and compare it to \(\hat{\xi}_{lnij}^{\text{left}}\) (or \(\hat{\xi}_{lnij}^{\text{right}}\)). Ciuti et al \(^{(11)}\) and Rochat et al \(^{(9)}\) have calculated \(\hat{\xi}_{lnij}^{\text{left}}(i, j)\) for 2D scatterings between \(\nu_i = \nu_j = 1S\), \(Q_i = Q_j = 0\), and \(\nu_l = \nu_n = 1S\), \(Q_l = -Q_n = p\). Fig. (4) of reference \(^{(9)}\) precisely shows
\[
g_{\text{exch}}^{\text{exch}}(p) = -\frac{\pi^2 S}{4e^2 \lambda_{2D}^2} \hat{\xi}^{\text{left}}_{lnij} \left( \begin{array}{c} p \\ 0 \end{array} \right) ,
\] (56)
as a function of \(P = p\lambda_{2D}\), with \(\lambda_{2D}\) being the 2D Bohr radius. On our Fig. (5) is shown
\[
\Delta g(p) = \frac{\pi^2 S}{4e^2 \lambda_{2D}^2} \left[ \hat{\xi}^{\text{left}}_{lnij} \left( \begin{array}{c} p \\ 0 \end{array} \right) - \hat{\xi}^{\text{right}}_{lnij} \left( \begin{array}{c} p \\ 0 \end{array} \right) \right] = \frac{\pi^2}{2} \alpha_e \alpha_h p^2 \lambda_{2D}^2 \Lambda(p) ,
\] (57)
with \(\Lambda(p)\) given by Eq (38). We see that, except for very small \(p\lambda_{2D}\), this difference is of the order of \(g_{\text{exch}}^{\text{exch}}(p)\), unless \(m_e << m_h\).
We can rewrite the $H$ matrix element between two-exciton states in a more symmetrical form with respect to $(l,n)$ and $(i,j)$ by taking half the sum of Eqs (53) and (54). If in addition, we consider this $H$ matrix element between two-exciton states *normalized*, but still not orthogonal, namely

$$|\psi_{ji}\rangle = |\psi_{ij}\rangle = B_i^\dagger B_j^\dagger |v\rangle = \frac{1}{N_{ij}} B_i^\dagger B_j^\dagger |v\rangle,$$  

(58)

where $N_{ij} = (1 + \delta_{ij} - 2\lambda_{ijij})^{1/2}$ due to Eq (52), we find, using Eqs (53-54),

$$\langle \psi_{ln}|H|\psi_{ij}\rangle = \frac{1}{2}(E_l + E_n + E_i + E_j)\delta_{lnij} + \frac{2}{N_{ln}N_{ij}} \left[ \xi_{lnij}^{\text{dir}} - \xi_{lnij}^{\text{exch}} - (1 - \delta_{lnij})\eta_{lnij} \right].$$  

(59)

$\delta_{lnij}$ is equal to 1 when the excitons on both sides are identical i.e. $(l,n) = (i,j)$, and 0 otherwise, while $\eta_{lnij}$ is given by

$$\eta_{lnij} = \frac{1}{2}(E_l + E_n + E_i + E_j)\lambda_{lnij}.$$

(60)

The first term of this $H$ matrix element just corresponds to the energies of the non-interacting $(i,j)$ or $(l,n)$ excitons if, in addition, the two-exciton states $|\psi_{ij}\rangle$ are assumed to be orthogonal. The last terms of this $H$ matrix element come from interactions. They have three origins. The first term, $\xi_{lnij}^{\text{dir}}$, is a direct Coulomb scattering between excitons made on both sides with the same electrons and holes. The second term, $\xi_{lnij}^{\text{exch}}$, is an exchange Coulomb scattering in which the holes (or electrons) making the excitons are exchanged. The last term, $\eta_{lnij}$, has a purely fermionic origin: It is directly linked to the fact that the $|\psi_{ij}\rangle$ and $|\psi_{ln}\rangle$ states are not orthogonal for $(l,n) \neq (i,j)$. It does not exist in diagonal scatterings. Let us however stress that, as $\eta_{lnij}$ depends on the sum of the four exciton energies, it is band gap dependent; so that it is very unlikely that such a $\eta_{lnij}$ term appears in a physical scattering. We are going to come back to this problem in the next paragraph.

It might be useful to mention that the diagonal matrix element of $H$ reduces to

$$\langle \psi_{ij}|H|\psi_{ij}\rangle = E_i + E_j + (\xi_{ijij}^{\text{dir}} - \xi_{ijij}^{\text{exch}})/(1 + \delta_{ij} - 2\lambda_{ijij}),$$

(61)

with $\xi_{ijij}^{\text{exch}}$ possibly replaced by $\xi_{ijij}^{\text{right}}$ or $\xi_{ijij}^{\text{left}}$ as these three scatterings are equal for diagonal processes. $(E_i + E_j)$ is just what we would expect for the expectation value of the hamiltonian between two non-interacting boson-excitons $i$ and $j$. The other terms come from scattering processes between the $i$ and $j$ excitons resulting from Coulomb interaction and Pauli exclusion.
6 Exciton-exciton scattering rate

Let us end this work by considering a physical quantity in which these \( H \) matrix elements between two-exciton states may appear in a direct way. Ciut et al (CSPQS) have proposed \(^{(11)}\) to calculate the elastic Coulomb scattering of 1s excitons by 1s excitons, namely

\[
(1s, Q, S) + (1s, Q', S') \rightarrow (1s, Q + q, S_f) + (1s, Q' - q, S'_f) ,
\]

(62)

(see their Eq (3)), from the matrix element of the exact hamiltonian \( H \) between these two-exciton states. Let us rewrite this Eq (62) as \((i) + (j) \rightarrow (l) + (n)\). In the following we will drop the angular momentum parts for simplicity, as they are unimportant for the problem we raise.

The one-exciton wave function \( \phi_Q(r_e, r_h) \) given in their Eq (1) is just our \( \phi_i(r_e, r_h) \) (see Eq (26)), while the two-exciton wave function \( \phi_{QQ}^{S'S} \) given in their Eq (4) reads, within our notations,

\[
\phi_{ij}(r_e, r_h, r_{e'}, r_{h'}) = \frac{1}{2} [\phi_i(r_e, r_h) \phi_j(r_{e'}, r_{h'}) + (r_e \leftrightarrow r_{e'}, r_h \leftrightarrow r_{h'}) - (r_e \leftrightarrow r_{e'}) - (r_h \leftrightarrow r_{h'})] ,
\]

(63)

so that it is just the wave function of the state \( B^\dagger_i B^\dagger_j |v\rangle \).

It is possible to show directly from Eq (63), i.e. their Eq (4), that

\[
\langle \phi_{ln} | \phi_{ij} \rangle = \delta_{li} \delta_{nj} + \delta_{lj} \delta_{ni} - 2\lambda_{lnij} ,
\]

(64)

with

\[
\lambda_{lnij} = \frac{1}{2} \int dr_e dr_h dr_{e'} dr_{h'} [\phi_i^*(r_e, r_h) \phi_j^*(r_{e'}, r_{h'}) + (l \leftrightarrow n)] \phi_i(r_e, r_h) \phi_j(r_{e'}, r_{h'}) ,
\]

(65)

which is nothing but our Eqs (30,33) and (52). As a consequence, the \( |\phi_{ln}\rangle \) states are not normalized. Eq (64) also shows that the initial and final states of the exciton-exciton scattering considered in Eq (62) are not orthogonal for \( q \neq 0 \) or \( q \neq Q' - Q \). So that, even if for such \( q \), the \( \delta \) terms of Eq (64) give zero, this matrix element is not zero due to the \( \lambda_{lnij} \) term.

According to CSPQS, the scattering amplitude corresponding to the process of Eq (62) should be equal to \( H_{SS'}^{S'S'}(Q, Q', q) = \langle \phi_{ij} | H | \phi_{ln} \rangle \) (see their Eq (6)), where \( H \) is the exact two-electron and two-hole semiconductor hamiltonian. Their Eq (6) is nothing but
\langle v|B_i B_j H B_i^\dagger B_j^\dagger|v\rangle \text{ written in } r \text{ space. Our equations } (53,54) \text{ immediately give}

\begin{align*}
\langle v|B_i B_j H B_i^\dagger B_j^\dagger|v\rangle &= 2 \left[ \xi_{ijln}^{\text{dir}} - \xi_{ijln}^{\text{left}} - (E_l + E_n)\lambda_{ijln} \right] \\
&= 2 \left[ \xi_{ijln}^{\text{dir}} - \xi_{ijln}^{\text{right}} - (E_i + E_j)\lambda_{ijln} \right],
\end{align*}

(66)

(67)
as the \( \delta \) terms give zero for \( \mathbf{q} \neq \mathbf{0} \) and \( \mathbf{q} \neq \mathbf{Q'} - \mathbf{Q} \). If we consider the results given by CSPQS in their Eq (7), we see that the first two terms given by their Eqs (12-14), exactly correspond to the two terms of \( (2\xi_{ijln}^{\text{dir}}) \) (see our Eqs (21,25)), while the last two terms (their Eq (16) and its “hole” equivalent) correspond to the two terms of \( (-2\xi_{ijln}^{\text{right}}) \) (see our Eqs (41,46)). Consequently they should have found the result given in Eq (67).

Even if calculations in \( r \) space are quite cumbersome, it is in fact possible to check the existence of the missing term \( (E_i + E_j)\lambda_{ijln} \) directly from a (tedious) calculation of the right hand side of their Eq (6). The origin of their mistake probably comes from the fact that they have considered the two states \( |\phi_{ln}\rangle \) and \( |\phi_{ij}\rangle \) as orthogonal, which is not true (see Eq (64)).

The correct value of the \( \langle \phi_{ij}|H|\phi_{ln}\rangle \) matrix element given in Eqs (66-67) leads us to question the validity of the whole procedure to determine the exciton-exciton scattering rate.

(i) First the \( |\phi_{ij}\rangle \) wave functions are not normalized, so that it would be reasonable to use normalized states and relate the exciton-exciton scattering not to \( \langle \phi_{ij}|H|\phi_{ln}\rangle \) but to \( \langle \phi_{ij}|H|\phi_{tn}\rangle/\langle \phi_{ij}|\phi_{ij}\rangle^{1/2}\langle \phi_{tn}|\phi_{tn}\rangle^{1/2} \). This would add a prefactor to all scatterings (see Eqs (58-59)).

(ii) There is however a much more dramatic problem with CSPQS procedure. The existence of the last terms of Eqs (66-67) is highly non physical for an exciton-exciton scattering : As it contains the exciton energy \( E_l + E_n \) (or \( E_i + E_j \)), which is essentially equal to twice the band gap, such a scattering would be band gap dependent. We could of course get rid of this problem by deciding to use for \( H \) an hamiltonian without the band gap i.e. by dropping \( \Delta \) in Eq (3).

Actually the exciton-exciton scattering rate cannot be related to this matrix element for more fundamental reasons. In usual problems dealing with interactions, the hamiltonian can be written as \( H = H_0 + V \). The Fermi golden rule then says that the transition rate between two different \( H_0 \) eigenstates \( |i\rangle \) and \( |f\rangle \) results from the possible interaction between these two states through \( |\langle f|V|i\rangle|^2 \). When these states are eigenstates of \( H_0 \),
they form an orthogonal basis, so that this transition rate is also equal to $|\langle f | H | i \rangle|^2$, since $\langle f | H_0 | i \rangle = 0$ for $|f\rangle \neq |i\rangle$. All the difficulties here come from the fact that $H$ cannot be written as $H_0 + V$: There is no hamiltonian for which the two-exciton states $|\phi_{ln}\rangle$ and $|\phi_{ij}\rangle$ are the (exact) eigenstates, so that (i) these two-exciton states are not orthogonal for $(l,n) \neq (i,j)$; (ii) the matrix elements of $H$ between these states have no reason to be equal to the matrix element of “the” interacting potential $V$—which, anyway, cannot be formally extracted from $H$. Consequently there is no reason to believe that the exciton-exciton scattering rate is given by $|\langle \phi_{ln} | H | \phi_{ij} \rangle|^2$.

7 Conclusion

We have reconsidered our commutation technique designed to deal with interacting close-to-boson particles and extended it to excitons with spin degrees of freedom. Although more cumbersome, the expressions of the two important parameters of this commutation technique, namely $\xi_{lnij}^{\text{dir}}$ and $\lambda_{lnij}$, are still rather transparent. When the “spins” of the electron and the hole making the exciton are used—instead of the total kinetic momentum of the exciton—, these two parameters appear as a product of a spin part and an orbital part which have both a very simple physical meaning (see Eqs (22) and (25) for $\xi_{lnij}^{\text{dir}}$ and Eqs (31) and (33) for $\lambda_{lnij}$). The first one $\xi_{lnij}^{\text{dir}}$ corresponds to all Coulomb interactions between the $(i,j)$ or $(l,n)$ excitons when, on both sides, these excitons are made with the same electron-hole pairs $(e,h)$ and $(e',h')$. The second one $\lambda_{lnij}$ has a purely fermionic origin and is simply related to the fact that the $(i,j)$ and $(l,n)$ excitons can be made with different pairs, $(e,h)$ $(e',h')$ and $(e,h')$ $(e',h)$. This commutation technique allows to easily calculate any matrix elements between exciton states in an exact way. In this paper we have explicitly calculated the matrix elements of $H$ between two-exciton states and we have shown that they cannot be related to the scattering rate of two excitons as previously proposed.
APPENDIX A : Calculation of the $\gamma_{li}(q)$

Using Eq (17), $\gamma_{li}(q)$ reads

$$\gamma_{li}(q) = \beta_{\nu\nu'}(\alpha_h a_x q) - \beta_{\nu\nu'}(-\alpha_e a_x q), \quad (68)$$

with $\beta_{\nu\nu'}(u)$ defined by

$$\beta_{\nu\nu'}(u) = \langle x_\nu | e^{iu r/a_x} | x_{\nu'} \rangle = \beta_{\nu\nu'}^*(-u), \quad (69)$$

and $a_x$ chosen to be the 3D Bohr radius.

From Eq (69), we see that $\beta_{\nu\nu'}(0) = \delta_{\nu\nu'}$, the value $u = 0$ being obtained either for $q = 0$ or for $\alpha_{e,h} = 0$, i.e. $m_h$ (or $m_e$) infinite.

We can note that, if the $\nu$ and $\nu'$ states have the same parity, $\beta_{\nu\nu'}(u) = \beta_{\nu\nu'}(-u)$, so that $\gamma_{li}(q) = 0$ for $m_e = m_h$ : There is no direct scattering towards a same parity state if the electron and hole masses are equal.

We can also note that the scattering $\gamma_{li}(q)$ depends on $q = |q|$ only if the $(l, i)$ states are S states.

Let us now calculate some values of these $\gamma_{li}(q)$, for 3D and 2D systems, when the $(l, i)$ states are S states.

1) 3D case

In 3D, the exciton relative motion wave function reads

$$\langle r | x_\nu \rangle = a_x^{-3/2} \varphi_{\nu}^{(3D)}(\rho, \theta, \varphi), \quad (70)$$

where $\rho = r/a_x$. The wave functions of the lowest energy S states are given by

$$\varphi_{1s}^{(3D)} = e^{-\rho/\sqrt{\pi}}, \quad \varphi_{2s}^{(3D)} = (2 - \rho)e^{-\rho/2\sqrt{32\pi}}. \quad (71)$$

In 3D, Eq (69) reads for S states,

$$\beta_{\nu\nu'}^{(3D)}(u) = \int_0^{+\infty} \rho^2 d\rho \int_0^{2\pi} d\varphi \int_0^\pi \sin \theta d\theta e^{iu \rho \cos \theta} \varphi^{(3D)*}_\nu (\rho, \theta, \varphi) \varphi^{(3D)}_{\nu'} (\rho, \theta, \varphi). \quad (72)$$

By inserting Eq (71) into Eq (72), we find

$$\beta_{1s,1s}^{(3D)}(u) = \frac{1}{(1 + u^2/4)^2}; \quad (73)$$

$$\beta_{2s,2s}^{(3D)}(u) = \frac{(1 - u^2)(1 - 2u^2)}{(1 + u^2)^4}; \quad (74)$$

$$\beta_{1s,2s}^{(3D)}(u) = \frac{2^{17/2} u^2}{3^6 (1 + 4u^2/9)^3}. \quad (75)$$
From these $\beta_{\nu\nu'}^{(3D)}(u)$, we can easily deduce the $\gamma_{lii}^{(3D)}(q)$ according to Eq (68), and the direct scattering $\xi_{lnij}^{\text{dir}}$ according to Eq (23).

2) 2D case

In 2D, the exciton relative motion wave function reads

$$\langle \mathbf{r} | x_{\nu} \rangle = a_{x}^{-1} \varphi_{\nu}^{(2D)}(\rho, \varphi).$$

The lowest energy S state wave functions are given by

$$\varphi_{1s}^{(2D)} = \left[ \frac{2^{3/2}}{\sqrt{\pi}} \right] e^{-2\rho}, \quad \varphi_{2s}^{(2D)} = \left[ \frac{(2/3)^{3/2}}{\sqrt{\pi}} \right] \left( 1 - \frac{4\rho}{3} \right) e^{-2\rho/3}.$$ (77)

For 2D S states, Eq (69) reads

$$\beta_{\nu\nu'}^{(2D)}(u) = \int_{0}^{+\infty} \rho d\rho \int_{0}^{2\pi} d\varphi \ e^{iu\rho \cos \varphi} \varphi_{\nu}^{(2D)}(\rho, \varphi) \varphi_{\nu'}^{(2D)}(\rho, \varphi).$$ (78)

Inserting Eq (77) into Eq (78), we find

$$\beta_{1s,1s}^{(2D)}(u) = \frac{1}{(1 + u^2/16)^{3/2}},$$
$$\beta_{2s,2s}^{(2D)}(u) = \frac{1 - (27u^2/16) + (81u^4/256)}{(1 + 9u^2/16)^{7/2}},$$
$$\beta_{1s,2s}^{(2D)}(u) = \frac{3^{7/2}u^2}{2^9(1 + 9u^2/64)^{5/2}}.$$ (80)

From these $\beta_{\nu\nu'}^{(2D)}(u)$, we can deduce the $\gamma_{lii}^{(2D)}(q)$ according to Eq (68) and the direct scattering $\xi_{lnij}^{\text{dir}}$ according to Eq (23). For direct scatterings within the 1S states only, we recover the result given in ref. (11).
APPENDIX B : Expressions of $\hat{\xi}_{\text{dir}}$, $\hat{\lambda}_{\text{lnij}}$ and $\hat{\xi}_{\text{right}}$ in $\mathbf{r}$ space

In order to show the equivalence between Eqs (24) and (25), it is simpler to start from Eq (25). Let us consider the $V_{ee'}$ term. By taking the Fourier transform of $V_{ee'}$, namely

$$V_{ee'} = \sum_{q \neq 0} V_q e^{i\mathbf{q} \cdot (\mathbf{r}_e - \mathbf{r}_{e'})} ,$$

and the Fourier transform of the relative motion part of the wave function given in Eq (26), namely

$$\phi_i(\mathbf{r}_e, \mathbf{r}_h) = \frac{1}{V} \sum_{k_i} \langle x_{\nu_i} | k_i \rangle e^{i\mathbf{k}_i \cdot \mathbf{r}_e} e^{i\mathbf{k}_i \cdot \mathbf{r}_h} ,$$

with $\mathbf{k}_i = \mathbf{k}_i + \alpha_e \mathbf{Q}_i$ and $\mathbf{k}_h = -\mathbf{k}_i + \alpha_h \mathbf{Q}_i$, we can rewrite the $V_{ee'}$ term of Eq (25) as

$$\hat{\xi}_{ee'}(\mathbf{lnij}) = \sum_{k_i, k_n, k_j} \langle x_{\nu_i} | k_i \rangle \langle x_{\nu_n} | k_n \rangle \langle x_{\nu_j} | k_j \rangle \sum_{q \neq 0} V_q \frac{1}{V^3} \int d\mathbf{r}_e d\mathbf{r}_h d\mathbf{r}_e' d\mathbf{r}_h' \times e^{i[(-\mathbf{k}_i + \mathbf{q} \cdot \mathbf{r}_e + (-\mathbf{k}_h + \mathbf{q} \cdot \mathbf{r}_h) + (-\mathbf{k}_n + \mathbf{k}_i - \mathbf{q} \cdot \mathbf{r}_e + (-\mathbf{k}_h + \mathbf{k}_j) \cdot \mathbf{r}_h'] .}$$

The integral being equal to $\delta_{\mathbf{k}_i - \mathbf{k}_e + \mathbf{q}, \mathbf{k}_h} \delta_{\mathbf{k}_h, \mathbf{k}_i - \mathbf{q}} \delta_{\mathbf{k}_n, \mathbf{k}_j}$, we immediately recover the first term of Eq (24).

By transforming in the same way the other terms in $V_{hh'}, V_{eh'}, V_{e'h}$, it is easy to derive Eq (24) from Eq (25).

In order to show the equivalence of Eqs (32) and (33), we can proceed similarly : By inserting Eq (83) into Eq (33) and by performing the integral, we immediately obtain Eq (32).

We do the same to show the equivalence of Eqs (42) and (46) : We insert Eqs (82,83) into Eq (46) and perform the integral. This immediately gives the first term of Eq (42). The same transformation of the terms in $V_{hh'}, V_{eh'}, V_{e'h}$ leads to the three other terms of Eq (42).
APPENDIX C : Explicit calculation of $F_{1s1s1s}(0,0)$

From Eq (36), we get

$$F_{1s1s1s}(0,0) = \sum_{k} |\langle k|x_{1s}\rangle|^4.$$  \hspace{1cm} (85)

The Fourier transforms of the relative motion wave functions $\langle k|x_{1s}\rangle$ are respectively given, in 3D and 2D, by

$$\langle k|x_{1s}\rangle^{(3D)} = \frac{8\sqrt{\pi} a_x^{3/2}}{\sqrt{V}(1 + a_x^2 k^2)^2},$$ \hspace{1cm} (86)

$$\langle k|x_{1s}\rangle^{(2D)} = \frac{\sqrt{2\pi} a_x}{\sqrt{S} \left(1 + \frac{a_x^2 k^2}{4}\right)^{3/2}},$$ \hspace{1cm} (87)

$a_x$ being the 3D exciton Bohr radius.

In 3D we thus obtain

$$F_{1s1s1s}^{(3D)}(0,0) = \frac{2^{11} a_x^3}{V} \int_{0}^{+\infty} \frac{x^2 dx}{(1 + x^2)^8} = \frac{33\pi a_x^3}{2V}.$$ \hspace{1cm} (88)

In 2D we get

$$F_{1s1s1s}^{(2D)}(0,0) = \frac{2\pi a_x^2}{S} \int_{0}^{+\infty} \frac{xdx}{(1 + x^2)^6} = \frac{4\pi a_x^2}{5S}.$$ \hspace{1cm} (89)
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they give in their Eqs (19-20).
FIGURE CAPTIONS

Fig. (1)

Direct Coulomb scattering of an $i$ exciton into an $l$ exciton, while a $j$ exciton is scattered into an $n$ exciton. As Coulomb interaction conserves the angular momenta of electrons and holes, we must have $s_l = s_i$, $s_n = s_j$, $m_l = m_i$, $m_n = m_j$. As it also conserves momenta, we must have $Q_i + Q_j = Q_l + Q_n$, between the centers of mass of the “in” and “out” excitons. Finally this direct Coulomb scattering contains the quantities $\gamma_{li}(q)$ and $\gamma_{nj}(-q)$, with $q = Q_l - Q_i$, which characterize the scattering of a $\nu_i$ exciton into a $\nu_l$ state and a $\nu_j$ exciton into a $\nu_n$ state, under a $q$ excitation (see Eq (17)).

Fig. (2)

Bare exchange coefficient : The $i$ and $l$ excitons are made with the same electron but different holes ; we do have $s_l = s_i$ but $m_l = m_j$.

Fig. (3)

The dimensionless exchange coefficient $\Lambda(p)$, given by Eq (38), as a function of $P = p\lambda_{2D}$, for 2D excitons and three different values of $m_e/m_h$. Thick solid line : $m_e/m_h = 0$. Thin solid line : $m_e/m_h = 0.5$. Dashed line : $m_e/m_h = 1$.

Fig. (4)

Right exchange Coulomb scattering (a), and left exchange Coulomb scattering (b). In the right exchange scattering, the exchange of holes making the $(i,j)$ excitons is made before the direct Coulomb interaction which then scatters the $(p,r)$ excitons into $(l,n)$ states. Note that the electron-hole interactions are between the $(l,n)$ excitons but inside the $(i,j)$ excitons. Exchange and Coulomb processes conserving spins, we do have $s_l = s_i$, $s_n = s_j$ along with $m_l = m_j$, $m_n = m_i$, the holes being crossed in the process. Note that the $(l \leftrightarrow n)$ change appearing in the definition of $\xi^{\text{dir}}_{lmij}$ and $\lambda_{lmij}$ generates a similar term in which the electrons are crossed instead of the holes. This restores the electron-hole symmetry.

Fig. (5)

The dimensionless difference between left and right exchange scatterings $\Delta g(p)$, given by Eq (57), as a function of $P = p\lambda_{2D}$, for 2D excitons and three different values of $m_e/m_h$. Thick solid line : $m_e/m_h = 0$. Thin solid line : $m_e/m_h = 0.5$. Dashed line : $m_e/m_h = 1$. 

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