Scattering amplitudes for dark and bright excitons

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Abstract – Using the composite boson many-body formalism that takes single-exciton states rather than free carrier states as a basis, we derive the integral equation fulfilled by the exciton-exciton effective scattering from which the role of fermion exchanges can be unraveled. For excitons made of (±1/2)-spin electrons and (±3/2)-spin holes, as in GaAs heterostructures, one major result is that most spin configurations lead to brightness-conserving scatterings with equal amplitude ∆, despite differences in the carrier exchanges involved. A brightness-changing channel also exists when two opposite-spin excitons scatter: dark excitons (2, −2) can end either in the same dark states with an amplitude ∆e, or in opposite-spin bright states (1, −1), with a different amplitude ∆o, the number of carrier exchanges involved in these scatterings being even or odd, respectively. Another major result is that these amplitudes are linked by a striking relation, ∆e + ∆o = ∆, which has decisive consequence on exciton Bose-Einstein condensation. By using Born values, we show that the exciton condensate can be optically observed through a bright part when excitons have large dipole only, that is, when the electrons and holes are in two well-separated layers, as in current experiments.

In contrast to the structureless 4He, a number of bosonic condensates have more than one component inherited from the internal spin and orbital degrees of freedom of their constituents, the superfluid then being multi-component. Dipolar Bose gases [1] and superfluid phases of 3He [2] are prime examples. This also occurs to excitons, which are composite bosons (cobosons for short) made of one conduction electron and one valence hole. Since electrons and holes carry spins, so do excitons, their condensate depending on these internal degrees of freedom. Recently, it has been shown that signatures of exciton condensates were long held back by the missed fact that the lowest-energy states are dark [3,4], that is, not coupled to light. This precludes a direct photoluminescence observation of exciton condensate in the very dilute regime. Optical evidences for Bose-Einstein condensation are bound to a density regime where dark and bright components coexist coherently [5]. The darkening of the exciton gas upon cooling has been recently seen [6–10], and the macroscopic spatial coherence of the bright component has just been revealed [7,10], which provides unambiguous evidence for the coexistence of dark and bright exciton condensates.

As a rule, the energy of a condensate depends weakly on its internal degrees of freedom, all possible “spin” phases being essentially degenerate. This degeneracy is lifted by interactions. One then has to determine which combination of the competing phases produces the lowest energy that rules the condensate properties.

In this letter, we first derive the brightness-conserving and brightness-changing scattering amplitudes that are crucial to characterize the exciton Bose-Einstein condensate, brightness-changing scattering being the unique channel that introduces a bright component into the otherwise dark condensate. We end with a discussion of the condensate brightness and polarization in the light of these scatterings.
Having in mind GaAs heterostructures with spin (±1/2) electrons and spin (±3/2) holes, we first show that for all spin configurations in which brightness is conserved, the scattering amplitudes have the same value, Δ, despite differences in the carrier exchanges they contain. By contrast, when two opposite-spin dark excitons (+2, −2) scatter, they can either keep their darkness or become bright with spins (+1, −1), through different scattering amplitudes, Δₐ or Δₒ, the number of carrier exchanges involved being even or odd, respectively. We find that these scattering amplitudes are linked by Δₐ + Δₒ = Δ, which is reasonable because Δ contains even and odd numbers of carrier exchanges.

A salient consequence of this study is that, for exciton scatterings taken in the Born approximation, the lowest-energy state is optically dark in 2D: the exciton condensate must then be fully dark, ruling out any signature of macroscopic spatial coherence through photoluminescence experiments. Such a signature can however be obtained in bilayer heterostructures, with electrons and holes confined in two adjacent layers that are sufficiently far apart [5].

Starting from the 1970’s, there has been continuous study of scattering length for semiconductor excitons [11–15]. However, the long-range character of Coulomb potential, and mostly the lack of appropriate procedure to handle carrier exchanges occurring along with repeated fermion-fermion interactions, have impeded significant progress. Two decades ago, the realization of Bose-Einstein condensates in ultracold atomic gases brought a new impetus, prompting scattering length studies for fermionic-atom dimers [16–20], and more recently for positronium atoms [21–28]. These studies, which commonly use fermions as elementary quantum objects, include the Pauli exclusion principle between the particle elementary constituents in the most natural way. However, in doing so, one loses the fact that the two-pair scattered states stay very close to two non-interacting paired states. To take advantage of this physical fact, one has to represent four fermions as two interacting paired states. To take advantage of this physical fact, one has to represent four fermions as two interacting paired states.

The coboson many-body formalism introduces:

i) The energy-like direct-Coulomb scattering, ξ_i^q_j(p_i), in which exciton in state i scatters to state p while keeping its two fermions (see fig. 1(a)). Index i denotes the exciton center-of-mass and relative-motion degrees of freedom. ξ_i^q_j(p_i) contains all possible fermion-fermion interactions between two excitons.

ii) The hole-exchange scattering, λ_h_i^q_j(p_i), in which the excitons (i, j) exchange their holes, excitons p and i having the same electron (see fig. 1(c)). No fermion-fermion interaction occurs in this scattering; so, λ_h_i^q_j(p_i) is dimensionless. This hole-exchange scattering is topologically equivalent to an electron-exchange scattering within a (i, j) permutation, λ_h_i^q_j(p_i) = λ_e_i^q_j(p_i), as seen from their Shiva diagrams in figs. 1(c), (d).

iii) The “in” exchange-Coulomb scattering, defined as $\xi_i^u_v(u_i, v_i) = \sum_{u,v} \lambda_h_i^q_j(u_i, v_i)\xi_i^u_v(u_i, v_i)$, in which Coulomb interaction between the “in” states (i, j) is followed by a hole exchange (see fig. 1(b)). A similar “out” exchange-Coulomb scattering (not shown) has its Coulomb interaction between the “out” states (p, q).

This formalism makes it easy to trace exciton spins through Shiva diagrams, each carrier keeping its spin in the scattering. Excitons made of s-spin electron and m-spin hole have a total spin S = s + m. For s = ±1/2 and m = ±3/2, S can be ±1 (excitons are bright) or ±2 (excitons are dark, i.e., not coupled to light). Three configurations can occur:

Case (1): When two excitons have opposite-spin electrons and same-spin holes, (1/2, −1/2; m, m), they form a dark and a bright exciton, (S = 2, S’ = 1) or (S = −2, S’ = −1), depending on m. The spins, or brightness, of these (S, S’) excitons are conserved through direct-Coulomb scatterings and hole-exchange scatterings. A similar conclusion holds for (s, s; 3/2, −3/2) (see fig. 2(a)).

Case (2): When two excitons have same-spin electrons and same-spin holes, (s, s; m, m), they form two same-spin excitons, either dark (S = ±2), or bright (S = ±1), depending on (s, m). The spin, or brightness, of these (S, S) excitons is conserved through direct-Coulomb scatterings, electron-exchange and hole-exchange scatterings (see fig. 2(a)).
Case (3): When two excitons have opposite-spin electrons and opposite-spin holes, $(1/2, -1/2; 2/3, -2/3)$, they can form two opposite-spin dark excitons, $(2, -2)$, or two opposite-spin bright excitons, $(1, -1)$. Excitons $(2, -2)$ can keep their spins, or darkness; they can also change into bright excitons through an odd number of electron or hole exchanges (see fig. 2(b)); carrier exchange opens a brightness-changing channel that splits the dark-bright exciton quasi-degenerate subspace. Different from a standard scattering problem, its resolution requires a novel approach compared to those in which brightness is conserved.

**Brightness-conserving scattering.** – The energy difference, $\Delta$, between the two-correlated-exciton ground state, $E_2$, and two single-exciton ground states, $2E_1$, scales as the inverse of the sample volume. In 3D, it is commonly written in terms of the scattering length $a_s$. Dimensional arguments give it as $\Delta \propto a_s/M L^3$ (for $h = 1$), with a $4\pi$ prefactor [31].

Using the boson many-body formalism, we find this energy difference as [15] (see supplementary material Supplementarymaterial.pdf (SM))

$$\Delta = E_2 - 2E_1 = \tilde{\zeta}(p_{00}/p_{00}),$$

with $\tilde{\zeta}(q_{ij}/p_{ij})$ solution of the integral equation (see fig. 3),

$$\tilde{\zeta}(q_{ij}/p_{ij}) = \zeta(q_{ij}/p_{ij}) + \sum_{ij \neq 00} \frac{1}{E_{ji} - E_{ij}} \tilde{\zeta}(q_{ij}/p_{ij}),$$

where $E_{ij} = E_i + E_j$ with $E_i$ being the i exciton energy. The kernel scattering $\zeta(q_{ij}/p_{ij})$ has a direct part and an exchange part (see fig. 4)

$$\zeta(q_{ij}/p_{ij}) = \xi(q_{ij}/p_{ij}) - \xi\text{exch}(q_{ij}/p_{ij}).$$

The exchange part, given by (SM)

$$\xi\text{exch}(q_{ij}/p_{ij}) = \xi^{\text{in}}(q_{ij}/p_{ij}) + \lambda(q_{ij}/p_{ij}) (E_{ij} - E_{00})$$

$$= \xi^{\text{out}}(q_{ij}/p_{ij}) + (E_{pq} - E_{00}) \lambda(q_{ij}/p_{ij}),$$

contains the physically expected exchange-Coulomb scattering, $\xi^{\text{in}}(q_{ij}/p_{ij})$ or $\xi^{\text{out}}(q_{ij}/p_{ij})$, and a less obvious part constructed on the dimensionless carrier-exchange scattering, $\lambda(q_{ij}/p_{ij})$, multiplied by an energy difference that makes it energy-like and band-gap free, as physically required.

This kernel scattering is symmetrical with respect to “in” and “out” states, as seen after summing the two expressions of eq. (4). Its $\lambda(q_{ij}/p_{ij})$ part is necessary to preserve time-reversal symmetry, $\zeta(q_{ij}/p_{ij}) = \zeta(q_{ij}/p_{ij})^\ast$: without it, the kernel scattering would lead to a non-Hermitian effective Hamiltonian [32].

When the two excitons have same hole spin and opposite electron spins, $(1/2, -1/2; m, m)$, the exchange scattering that appears, $\xi\text{exch}(q_{ij}/p_{ij})$, is constructed on $\lambda_h$, the two excitons exchanging their same-spin holes. And, similarly for $(s, s; m, m)$, as for two same-dark or bright excitons, $\xi\text{exch}(q_{ij}/p_{ij})$ is equal to $(\xi\text{exch}(q_{ij}/p_{ij}) + \xi\text{exch}(q_{ij}/p_{ij})/2$, electron exchange and hole exchange being both possible.

The first term in eq. (2) corresponds to the Born approximation, $\zeta(q_{ij}/p_{ij}) \approx \zeta(q_{ij}/p_{ij})$. Since $\lambda_h(q_{ij}/p_{ij})$ and $\lambda_e(q_{ij}/p_{ij})$ are equal for $i = j$, as seen from the Shiva diagrams of figs. 1(c), (d), we do have $\zeta_e(q_{ij}/p_{ij}) = \zeta_h(q_{ij}/p_{ij}) = \zeta_{ve}(q_{ij}/p_{ij})$. So, exchanging a hole, an electron or both produces the same scattering amplitude at the Born level.

This result remains true at all orders in interaction. It follows from $\xi\text{exch}(q_{ij}/p_{ij}) = \xi(q_{ij}/p_{ij})$ and

$$\xi\text{exch}(q_{ij}/p_{ij}) = \xi\text{exch}(q_{ij}/p_{ij}) = \xi\text{exch}(q_{ij}/p_{ij}) = \xi\text{exch}(q_{ij}/p_{ij}),$$

which come from the topological equivalence of their Shiva diagrams. These relations lead to $\xi\text{exch}(q_{ij}/p_{ij}) = \xi\text{exch}(q_{ij}/p_{ij}) = \xi\text{exch}(q_{ij}/p_{ij})$; so, $\zeta_h(q_{ij}/p_{ij}) = \zeta_e(q_{ij}/p_{ij}) = \zeta_{ve}(q_{ij}/p_{ij})$. Turning to the second-order term in eq. (2), it reads for excitons having same hole spins as

$$\sum_{ij \neq 00} \xi_h(q_{ij}/p_{ij}) \frac{1}{E_{00} - E_{ij}} \zeta_e(q_{ij}/p_{ij}) =$$

$$\sum_{ij \neq 00} \xi_h(q_{ij}/p_{ij}) \frac{1}{E_{00} - E_{ij}} \zeta_e(q_{ij}/p_{ij})$$

(6)
with $\xi_{ex, ch}^{(q, j)}$ possibly replaced by $\xi_{ex, ch}^{(q, j)}$. Interchanging the dummy indices $(i, j)$ in this exchange part gives the above right-hand-side as

$$
\sum_{ij \neq 00} \xi_e \frac{1}{E_{00} - E_{ij}} \xi_e (0 0) ,
$$

with $\xi_e$ possibly replaced by $\xi_{eh}$ through a similar procedure. By iterating the argument, we end with $\xi_e (0 0) = \xi_{eh}(0 0) = \xi_{eh}(0 0)$. The key for this equality is that the initial scattered states $(0, 0)$ are the same.

**Brightness-changing scatterings.** In Case (3), the brightness-changing channel couples the two-dark-exciton state $(2, -2)$ to the two-bright-exciton state $(1, -1)$. This leads us to look for the two-exciton eigenstates as

$$
|\Psi_2\rangle = \sum_{ij} b_{ij} B_{ij}^1 B_{ij}^{-1} |v\rangle + \sum_{ij} d_{ij} B_{ij}^1 B_{ij}^{-1} |v\rangle,
$$

where $|v\rangle$ denotes the vacuum state. In calculating the scatterings, one can neglect interband-Coulomb processes that produce a very small dark-bright energy splitting, and take the exciton states as degenerate. This degeneracy is lifted by interactions.

The Schrödinger equation for $|\Psi_2\rangle$ projected over $\langle v|B_{q,-1}B_{p,1}$ gives

$$
0 = (E_{pq} - E_2) b_{pq} + \sum_{ij} \left( \xi \left( \frac{q}{p} \right) \right) b_{ij} - \xi_e \left( \frac{q}{p} \right) d_{ij},
$$

with a similar equation when projected over $\langle v|B_{q,-3}B_{p,2}$. By adding and subtracting these two equations and by writing them for $(p, q) = (0, 0)$ and $(p, q) \neq (0, 0)$, we find (see SM)

$$
0 = \left( d_{00} + \xi_{eh} \right) \left( \xi_{eh} (0 0) - \Delta_{\pm} \right),
$$

where $\xi_{eh}$ follow from eq. (2) with $\xi$ replaced by $\xi_{eh} = \xi + \xi_{eh}$, the eigen-energy differences being $\Delta_{\pm}$. The above equation gives $E_2 = 2E_1 + \xi_{eh}(0 0)$ and $d_{00} = 0$, or $E_2 = 2E_1 + \xi_{eh}(0 0)$ and $d_{00} = -b_{00}$.

From this solution, we deduce the effective scatterings between two dark excitons ($\Delta_{dd}$), two bright excitons ($\Delta_{bb}$), and a dark and a bright exciton, ($\Delta_{db}$, $\Delta_{bd}$), through

$$
\begin{bmatrix}
\Delta_{bb} & \Delta_{ch} & \Delta_{bd} \\
\Delta_{db} & \Delta_{dd} & \Delta_{ch} \\
\Delta_{ch} & \Delta_{ch} & \Delta_{ch}
\end{bmatrix}
\begin{bmatrix}
b_{00} \\
b_{00} \\
\pm b_{00}
\end{bmatrix} = 0.
$$

This yields

$$
\begin{align*}
\frac{1}{2} \left( \xi_{eh} (0 0) + \xi_{eh} (0 0) \right) & = \Delta_{dd} = \Delta_{bb} = \Delta_e, \\
\frac{1}{2} \left( \xi_{eh} (0 0) - \xi_{eh} (0 0) \right) & = \Delta_{db} = \Delta_{bd} = \Delta_o.
\end{align*}
$$

As $\Delta = \Delta_e + \Delta_o$, these scattering amplitudes are linked by

$$
\Delta = \Delta_e + \Delta_o.
$$

At the Born level [30,33], the brightness-conserving scattering $\Delta_e$ reduces to the direct-Coulomb scattering $\xi(0 0)$, equal to zero since $\xi(0 1) = 0$ [29]. The brightness-changing scattering reduces to $\Delta_o \approx -\xi(0 0) = \xi(p/L) R_p$ where $a_p$ is the 3D exciton Bohr radius, $R_p$ the 3D exciton Rydberg and $D$ the space dimension, with $\xi_p = 26\pi/3$ as first obtained by Keldysh-Kozlov [11], and $\xi_2 = 8\pi - 315\pi^2/512$ for electrons and holes in the same quantum well [34], this value turning negative for carriers in two distant planes [35].

The integral equation (2) allows going beyond the Born approximation. From eq. (12), we see that $\Delta_o$ contains an odd number of fermion exchanges while in $\Delta_e$, this number is even, as physically expected because dark and bright states are coupled by carrier exchange while two exchanges reduce to an identity. The calculation of $\xi(0 0)$ requires the knowledge of the kernel scattering $\xi(0 0)$ which reads in terms of $\lambda, \xi, \xi^{in}$, all of which depend on single-exciton wave functions [29,36]. The numerical resolution of the integral equations for effective scatterings in the relevant experimental configurations will be addressed in a near future.

**Condensate brightness and polarization.** The scattering amplitudes between dark $S = \pm 2$ and bright $S = \pm 1$ electrons lead to the effective Hamiltonian

$$
H_{eff} = \xi_{bd} \sum_{S=\pm 1} B_S^1 B_S + \frac{\Delta}{2} \sum_{S=(\pm 2, \pm 1)} B_S^1 B_S^\dagger B_S B_S
$$

$$
+ \Delta \sum_{S=\pm 2} B_S^1 B_S^\dagger B_S^\dagger B_S B_S + \Delta_o \sum_{S=(\pm 2, \pm 1)} B_S^1 B_S^\dagger B_S B_S + \Delta_o \left( B_1^1 B_1^\dagger B_{-1} + \text{h.c.} \right),
$$

where $B_S^1$ is the $S$-spin exciton creation operator and $\xi_{bd}$ the bright-dark splitting, the dark exciton energy being taken as zero. Relation (13) between scattering amplitudes gives $H_{eff} = H'_{eff} + N_1(N-1)\Delta/2$ where $N_1 = \sum_S B_S^1 B_S$ is the total exciton-number operator and

$$
H'_{eff} = \xi_{bd} \sum_{S=\pm 1} B_S^1 B_S
$$

$$
- \Delta_o \left( B_1^1 B_1^\dagger B_{-1} - B_{-1}^1 B_1 \right) B_S B_S - B_S B_S B_S, \Delta_1 B_1^1 B_1^\dagger B_S B_S - B_S B_S B_S.
$$

Standard mean-field substitution, with $B_S^1$ replaced by $e^{i\pi S / N_1}$, gives the energy of $N = N + N_{-2} + N_1 + N_{-1}$ excitons as $E_N = N(N-1)\Delta/2$ with, after minimization with respect to the phase $\Theta = \pi S - \varphi_S - \varphi - \varphi_0 - \varphi_{-1}$,

$$
E_N = \xi_{bd}(N_1 + N_{-1}) - \Delta_o \left( \sqrt{N_2 N_{-2}} - \frac{\Delta_1}{\Delta_o} \sqrt{N_1 N_{-1}} \right)^2.
$$

For $N$ excitons created by photon absorption in bright ($\pm 1$) states, $N = N_1(0) + N_{-1}(0)$, the number of created electrons and holes with up or down spins are $N_{1/2} = N_{1/2}(0) = N_{1/2}(0) = N_{1/2}(0)$. When dark states exist, these carriers split
between bright and dark excitons as \( N_{\pm 1/2} = N_{\pm 1} \). Carrier spins being long-lived compared to the condensate lifetime, the carrier numbers of each species do not change when condensation occurs, i.e., \( N_\sigma = N_\sigma^{(0)} \) for \( \sigma = (\pm 1/2, \pm 3/2) \). Moreover, \( N_2 = N_{-2} \equiv N_d/2 \leq N^{(0)}_d \), since a \( S = 2 \) exciton is created along with a \( S = -2 \) exciton by carrier exchange between \( (1, -1) \) excitons. Consequently, the exciton condensate always has a bright part when \( N_1^{(0)} \neq 0 \), and no dark part when \( N_1^{(0)} = 0 \) or \( N_{-1}^{(0)} \) equals zero. Besides, since \( N_2 = N_{-2} \), the dark part is unpolarized — which cannot be distinguished from linearly polarized within the mean-field approximation.

Due to spin relaxation during the building-up of the excitonic system, \( N_1^{(0)} \) is equal to \( N_{-1}^{(0)} \); so, \( N_1 = N_{-1} \equiv N_b/2 \). The \( E'_N \) energy then reads in terms of dark and bright exciton numbers as

\[
E'_N = \varepsilon_{bd} N_b - \frac{\Delta_o}{4} \left( N_d + \frac{\Delta_o}{\Delta_e} N_b \right)^2.
\]

For positive \( \Delta_o \), as in the Born approximation for electrons and holes in a narrow quantum well, \( E'_N \), then equal to \( \varepsilon_{bd} N_b - \Delta_o N^2/4 \), is minimum for \( N_b = 0 \) whatever \( N \): the condensate is fully dark whatever the exciton density. By contrast, for negative \( \Delta_o \), as for electrons and holes in two widely separated 2D layers [7,10], \( E'_N \), then equal to \( \varepsilon_{bd} N_b + |\Delta_o| (N - 2 N_b)^2/4 \), is minimum for \( N_b = (N - N_{th})/2 \). Since \( N_b \) must be positive, the condensate turns “gray” with a bright and a dark component, when the exciton number \( N \) gets larger than \( N_{th} = \varepsilon_{bd}/|\Delta_o| \).

Note that for very short spin relaxation time, the constraint \( N_2 = N_{-2} \) is released. The lowest-energy state for \( \Delta_o \) negative would then be a fully polarized dark condensate, \( N_2 = 0 \) or \( N_{-2} = 0 \), whatever \( N \).

To conclude. – We have tackled the gray character [5] of exciton condensate in its full generality, without knowing relation (13) between scattering amplitudes. Comparing the effective Hamiltonian (14) with eq. (6) in ref. [5], we are led to set \( \Delta = v_{dd} = 2v_{dd} = 2v_{bd} \) and \( \Delta_o = 2 \varepsilon_{bd} \), with \( \Delta_o \) taken as negligible due to its zero Born value. For long carrier-spin lifetime, a gray condensate was found to appear under a density increase, whatever \( \Delta_o \) sign. By taking eq. (13) into account, the threshold for a bright component to appear becomes infinite for \( \Delta_o > 0 \), due to a hard-to-guess prefactor cancellation, thereby making the existence of a gray condensate very borderline for such \( \Delta_o \). Spin-orbit coupling [37,38] and experimental imperfections affecting the exciton energy degeneracy, like crystal strains, could also produce a gray condensate whatever the sample topology. Still, the fundamental relation (13) leads us to conclude that using dipolar excitons was indeed an excellent idea to evidence exciton condensation by optical means [7,10].

We have used the coboson many-body formalism to obtain the ground-state energy of two excitons whatever their carrier spins and, from it, to determine the various scattering amplitudes between dark and bright excitons. This formalism, which can also be used to study other multi-component condensates, allows us i) to understand the effects of fermion exchanges, ii) to obtain the integral equations fulfilled by the scattering amplitudes of brightness-conserving and brightness-changing channels, iii) to derive a general relation between them, remarkable for its simplicity, \( \Delta = \Delta_e + \Delta_o \).

The present work leads us to predict a darkening of the gray condensate when the distance between electron and hole layers decreases, excitons with large dipolar momentum being necessary for the condensate to have a bright component. We hope that this prediction will stimulate more challenging experiments on exciton condensation which, below a critical electron-hole separation, should occur in a dark state, hence impossible to “see” through standard photoluminescence experiments.

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