Time evolution of two ground-state excitons

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This work essentially deals with the time evolution of two ground-state excitons when these excitons have the same center-of-mass momentum. Since all coupled states have a higher energy, scatterings toward same energy states are formally impossible. As a result, previous works on the time evolution of two-exciton states should not apply. This led us to carefully reconsider the requirement of "energy conservation in the large time limit" through the Fermi golden rule, when dealing with composite excitons and to enlighten the key role of coherence time in the time evolution of excitons which is experimentally observed.

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I. INTRODUCTION

Extensive studies over the past decades of semiconductor excitations have led to the understanding of various sophisticated effects linked to exciton interactions. However, a few very fundamental questions still remain to be tackled properly. In this work, we address to one of them, the time evolution of exciton states when the Fermi golden rule does not *a priori* apply, as in the case of ground state excitons having *same* center-of-mass momentum, a quite common physical configuration.

The dynamical evolution of exciton states is an old topic. Excitons being made of two fermionic particles, a major difficulty is the proper handling of carrier exchanges, as induced by the Pauli exclusion principle. To overcome this problem, it was long ago proposed to "bosonize" the excitons, i.e., to replace these composite particles by elementary bosons interacting through effective scatterings, in which a certain amount of carrier exchange is included. The bosonization procedure commonly used for excitons follows Usui's work.[1] However, due to intrinsic inconsistency in the procedure, Haug and co-workers[2–4] end by constructing effective scatterings which produce a spurious non-Hermiticity in the resulting exciton-exciton Hamiltonian.[5]

Bosonization of excitons can appear as very appealing at first: its goal is to replace the electron-hole semiconductor Hamiltonian by a Hamiltonian between excitonic particles, which casts as H_X+V_{XX}. Interaction between excitons can then be handled through standard many-body procedures, these relying on the existence of a particle-particle interaction potential.

It is however clear that the concept of interactions "between" excitons is quite tricky.[6] Indeed, these interactions cannot be properly defined due to the exciton composite nature. Out of two indistinguishable electron-hole pairs $(e_1,e_2;h_1,h_2)$, we can construct excitons either on $(e_1,h_1)(e_2,h_2)$ or on $(e_1,h_2)(e_2,h_1)$. As a direct consequence, there is no clean way to decide, among the two sets of electron-hole interactions $(V_{e_1h_1}+V_{e_2h_2})$ and $(V_{e_1h_2}+V_{e_2h_1})$, the one which corresponds to interactions *between* two excitons.

In spite of this intrinsic difficulty and the fact that the exciton Hamiltonian commonly used in the literature is not Hermitian, this procedure has been very popular for quite a long time, various groups finding justifications, both theoretically[7–11] and even experimentally.[12–22] A few other groups tried full fermionic treatments of the problem. Those using semiconductor Bloch equations[23] are numerically quite demanding, mostly when elaborated truncated schemes are included.[24–26] Other mixed methods were also proposed,[27–32] in particular, to address to nonlinear optical susceptibilities.[27,28,33,34]

By contrast, we decided to try a totally different route which led us to produce a general many-body formalism for composite quantum particles,[35] in which the exciton is handled as an entity but, by contrast with bosonization, its composite boson nature is treated in an exact way. The key aspect of this theory is the existence of dimensionless "Pauli scatterings," which describe carrier exchanges between two excitons in the absence of carrier interaction. For readers not yet familiar with this formalism, we have in the Appendix given a brief survey of its key results, necessary to follow the present work.

This theory was shown to be quite successful in solving long standing theoretical problems such as the analytical resolution of the semiconductor Bloch equations[36] or the exact cancellation of volume linear terms in nonlinear susceptibility,[34] leading to results similar to the ones obtained by Sham and co-workers[27,28] or by Chernyak and Mukamel[33] through totally different approaches. However, the main advantage of this new theory relies on its diagrammatic representation through the so-called Shiva diagrams—due to their multiarm structure. These diagrams allow one to understand and, better, to predict physical effects like the ones induced by unabsorbed laser pulse: among them, we can cite spin precession,[37] Faraday rotation[38] and oscillation,[39] Bose-Einstein condensation of dark excitons,[40] and phase and Hadamard gates for trapped electron spin.[41]

Using this composite-boson many-body theory, we already considered the time evolution of exciton states.[42] This led us to discuss the various scatterings[43] which are, in a natural way, generated by this new theory and we related them to those appearing through the usual bosonization procedure.[2] This previous work on the exciton time evolution is briefly discussed in the last section of the present work. It uses a smart first line which allows us to generate energy conservation for processes at lowest order in Coulomb inter-

action, in a nicely direct way. However, this approach can hardly be extended to higher order terms. This forced us to work out a more general procedure to possibly generate these higher order terms. Indeed, these are *a priori* needed because all previous works on the time evolution of exciton states,[4,7,44] including ours,[42,43] end by considering the time evolution of two ground state excitons with same center-of-mass momentum. Since there is no other two-exciton state having same energy, transition toward same energy states, as imposed by lowest order Coulomb processes, are formally impossible. In spite of this difficulty, a $\delta(E_i+E_j-2E_0)$ factor appears in the transition rate of all these works, without any discussion.

This motivated us to reconsider the time evolution of two-exciton states through a procedure, which allows us to derive higher order contributions in Coulomb processes, this general procedure being of some possible use in other physical problems. The time evolution resulting from second-order processes actually has a rather unusual behavior. This pushed us to seriously reconsider the requirement of energy conservation in time evolution of exciton states and the validity of the Fermi golden rule in general for such a problem.

We already addressed in the past[45] to the question of possibly using the Fermi golden rule in the case of exciton formation through photon absorption. Textbooks tell us that the Fermi golden rule[46,47] applies to transitions toward a *continuum*. However, photons tuned on the ground state exciton level give rise to excitons, which have a momentum equal to the photon momentum so that the final exciton state in a photon absorption is definitely discrete. Other textbooks also tell us that exciton and photon form a mixed state called polariton.[48] In the polariton picture, which is said to be relevant for "strong coupling," these photons are not at all absorbed. Moreover, Rabi oscillations are known to exist in two-level atoms coupled to photons, these two-level atoms being often said to have analogies with excitons. We have been able to relate within a unique framework[49] these three rather different phenomena, namely, photon absorption with a transition rate given by the Fermi golden rule, photon-exciton coupled into polariton, and the Rabi oscillations of a two-level atom. We have shown that the Fermi golden rule is indeed valid for photon absorption provided that the exciton level has a broadening large enough compared to the exciton-photon coupling, this broadening playing the role of a continuum.

We wish to stress that, in the usual polariton picture, excitons are taken as noninteracting elementary bosons. This approximation is certainly incorrect for intense photon field. Interactions between polaritons have been first introduced through a bosonized procedure for the excitons,[50] with its usual problems. The clean treatment of polaritons while keeping the exciton composite nature is more difficult. We have recently succeeded to extend our composite-boson many-body formalism to polaritons;[51,52] however, more work is still needed, in particular, to properly control polarization effects[44] in the polariton framework.

We here study under which conditions the Fermi golden rule can be used to describe the time evolution of a two-exciton state. This leads us to highlight the key role played by a finite exciton coherence time in the time evolution which is experimentally observed, this coherence time controlling the effective quantization of the exciton center-of-mass momentum. To the best of our knowledge, we have not up to now seen such a discussion in the literature, although this discussion is crucial for possibly using the Fermi golden rule, as everybody does. Let us present the argument briefly: on the one hand, the uncertainty principle prevents energy conservation any better than $1/t$, where t is the experimental time. This experimental time cannot be any larger than the exciton coherence time τ_{coh} to possibly get meaningful data. On the other hand, the coherence time is related to the coherence length which physically scales the quantization of the exciton center-of-mass momentum so that the exciton kinetic energy is ultimately scaled by $1/\tau_{\mathrm{coh}}$. This proves that there always are many exciton states at $1/t$ from the initial state energy whatever this initial state since the condition t smaller than the coherence time prevents to take the $t\rightarrow\infty$ limit, in which a strict energy conservation would be required. Even if rather disappointing at first because nothing special happens when there is no other exciton state having exactly the energy of the two initial excitons, it nevertheless appears to us as important to establish using strong microscopical arguments, why what everyone commonly does is indeed correct.

The paper is organized as follows. In Sec. II, we point out a few difficulties encountered when tackling the time evolution of a two-exciton state, difficulties which have not been discussed in our previous works.[42,43] One of them is linked to the fact that the energy of a two-exciton state is not a well-defined quantity because there is no Hamiltonian for which this state is an exact eigenstate so that energy conservation cannot be a very precise concept. The other difficulty is that there is no other two-exciton state having exactly the energy of two ground state excitons when these excitons have same center-of-mass momentum. Directly linked to these difficulties is the problem of energy conservation at the $1/t$ scale and the proper way to take the large-time limit, and the large-sample limit because exciton momenta form a quasicontinuum at the L^{-1} scale where L *a priori* is the sample size.

Since a similar difficulty exists in the case of elementary particles, Sec. III is devoted to the time evolution of two-boson-exciton states. Its goal is to determine under which circumstances this time evolution is indeed controlled by the usual form of the Fermi golden rule, and what could be done instead when there are no other state having the energy of the initial state within a $1/t$ scale. In this section, we also question (i) the proper scale for the quantization of the exciton center-of-mass momentum, and (ii) the physical limitation on the time evolution of exciton states with respect to the exciton coherence time, this time being fundamentally linked to the quantization of the center-of-mass momentum.

In Sec. IV, we study somewhat in details the consequences of the overcompleteness of the two-exciton state basis on the various possible scatterings between two composite excitons. In addition to the standard scatterings described in details in our previous works,[43] we pay particular attention to scatterings associated to transitions toward the subspace perpendicular to the initial state, because states belonging to this subspace are the only ones which can be cleanly defined as "different" from the initial state.

In Sec. V, we show how to perform a Coulomb expansion of the time evolution of a two-exciton state when the system Hamiltonian does not split as H_X+V_{XX}. We explicitly calculate the probability to reach another two-exciton state, as well as the probability to stay in the initial state, at second order in Coulomb interaction. Particular attention is paid to initial state made of two ground-state excitons having same center-of-mass momentum.

In Sec. VI, we discuss the link between the present work and our previous works on the time evolution of exciton states. In Sec. VII, we conclude.

II. INTRINSIC DIFFICULTIES WITH TIME EVOLUTION OF EXCITON STATES

Let us consider an initial state

$$|\phi_{ij}\rangle = B_i^\dagger B_j^\dagger |v\rangle, \tag{2.1}$$

made of two excitons in states i and j. The index i stands for $(\nu_i, \mathbf{Q}_i)$ with $\mathbf{Q}_i$ being the exciton center-of-mass momentum and ν_i the relative motion index. $|v\rangle$ is the vacuum state and $B_i^\dagger$ is the creation operator of exciton i. These two excitons scatter with time as they are not eigenstates of the semiconductor Hamiltonian H due to Coulomb interaction *between* excitons—although, as explained above, this interaction cannot be properly defined due to the exciton composite nature. This means that

$$|\phi_{ij}(t)\rangle = e^{-iHt}|\phi_{ij}\rangle \tag{2.2}$$

is not barely $|\phi_{ij}\rangle$ within a phase factor but also contains contributions on other two-exciton states $|\phi_{mn}\rangle$, provided that $\mathbf{Q}_m+\mathbf{Q}_n=\mathbf{Q}_i+\mathbf{Q}_j$ since both Coulomb interaction and carrier exchange conserve momentum.

(i) A major problem immediately arises: due to the exciton composite nature, two-exciton states form an overcomplete set for two electron-hole pairs. This is evidenced by the closure relation in the two-electron-hole-pair subspace,[35,53] which, for $\langle v|B_m B_i^\dagger|v\rangle = \delta_{m,i}$, reads in terms of excitons as

$$I = \left(\frac{1}{2!}\right)^2 \sum_{i,j} |\phi_{ij}\rangle\langle\phi_{ij}|. \tag{2.3}$$

The prefactor $(\frac{1}{2!})^2$ would be $(\frac{1}{2!})$ only if excitons were taken as elementary bosons.

Another signature of the overcompleteness of the two-exciton basis is the identity[35]

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

which follows from exchanging the carriers of the two excitons i and j. This overcompleteness leads to a scalar product of two-exciton states given by (see Appendix)

$$\begin{aligned}\langle\phi_{mn}|\phi_{ij}\rangle &= \delta_{m,i}\delta_{n,j} - \lambda\begin{pmatrix} n & j \\ m & i \end{pmatrix} + (m \leftrightarrow n) \\ &= \delta_{m,i}\delta_{n,j} + \delta_{m,j}\delta_{n,i} - 2\lambda_{mnij}.\end{aligned} \tag{2.5}$$

It differs from zero for $(m,n) \neq (i,j)$, provided that the Pauli scattering for fermion exchanges $\lambda(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix})$ differs from zero. This is always so for $\mathbf{Q}_m+\mathbf{Q}_n=\mathbf{Q}_i+\mathbf{Q}_j$ since exchanges conserve momentum.

The above equation shows that, when thinking of the time evolution of a two-exciton state toward "another" two-exciton state, we certainly have in mind something more elaborate than just $|\langle\phi_{mn}|\phi_{ij}(t)\rangle|^2$ because, when $\mathbf{Q}_m+\mathbf{Q}_n=\mathbf{Q}_i+\mathbf{Q}_j$, this quantity differs from zero for $t=0$ already, i.e., before any time evolution. The clean elimination of the part of $|\phi_{mn}\rangle$ already included in the initial state $|\phi_{ij}\rangle$ is in itself a major problem. When considering time evolution toward "another" state, we expect a quantity which cancels with t. However, as shown in more details below, this cancellation is not enough to univocally define the relevant quantity, in particular, for problems in which we must go beyond first order in Coulomb interaction.

(ii) Even if we forget the difficulty linked to overcompleteness, the time evolution of two excitons raises another problem. We expect the states reached through the time evolution of $|\phi_{ij}\rangle$ to have the same energy in the large time limit. The problem we here address however is rather different from a standard time evolution. In the standard case, we start with an initial state $|\varphi_i\rangle$, eigenstate of $\mathcal{H}_0$, and we look for its time evolution under a perturbation W which is suddenly turned on, bringing the system Hamiltonian to $\mathcal{H}=\mathcal{H}_0+W$. This perturbation is later on turned off. The energy of the initial state is then well defined through $(\mathcal{H}_0-\mathcal{E}_i)|\varphi_i\rangle=0$, as well as the energy of the final state $|\varphi_f\rangle$, also taken as $\mathcal{H}_0$ eigenstate. The system evolves from $|\varphi_i\rangle$ to $|\varphi_f\rangle$, under the Hamiltonian $\mathcal{H}$ different from $\mathcal{H}_0$. In the large time limit, the states $|\varphi_f\rangle$ possibly reached through the time evolution of $|\varphi_i\rangle$ have the same energy as $|\varphi_i\rangle$, at the scale $1/t$ (for $\hbar=1$). This scale is just the signature of the uncertainty principle which, for large t, leads to the energy conservation of the Fermi golden rule,[45,46] this conservation being not strict, but at the $1/t$ scale only.

The problem we here address is conceptually quite different. The system Hamiltonian H, made of a free part H_0 and a Coulomb part between electrons and holes $V=V_{ee}+V_{hh}+V_{eh}$, stays the same all over the time. The initial state made of two independent excitons, changes with time because it is not eigenstate of the Hamiltonian. The difficulty is that, for the same reason as $|\phi_{ij}\rangle$ is not H eigenstate, its energy is not properly defined, nor the energy of the final state $|\phi_{mn}\rangle$. While for one exciton $(H-E_i)B_i^\dagger|v\rangle=0$, for two excitons $HB_i^\dagger B_j^\dagger|v\rangle$ is close but close only to $(E_i+E_j)B_i^\dagger B_j^\dagger|v\rangle$. The sum of the two free-exciton energies $E_i+E_j=E_{ij}$ could be seen as the "unperturbed energy." The problem, however, is that there is no "unperturbed Hamiltonian" for which this E_{ij} energy would be the exact eigenvalue. This "unperturbed Hamiltonian" must differ from the free part H_0 of H since E_i and E_j already contain electron-hole contributions, as necessary to get the exciton bound state. It must also differ from H_0+V_{eh} because the interaction between two excitons, which makes $(H-E_i-E_j)B_i^\dagger B_j^\dagger|v\rangle$ different from zero, contains contributions coming from interaction between the electron of one exciton and the hole of the other exciton, so that V_{eh} must enter interactions between two excitons. Consequently, even if we guess that E_{ij} essentially is the initial state unperturbed energy, this cannot be as clean as in the standard

Fermi golden rule. As a result, if an energy conservation similar to $\delta_t(E_{mn}-E_{ij})$ has to exist, it must be proved from *ab initio* calculations.

(iii) A last problem arises when the two excitons are in the ground state, these two excitons moreover having the same center-of-mass momentum—a highly relevant physical configuration. If we accept that $|\phi_{ii}\rangle$ can only evolve toward two-exciton states $|\phi_{mn}\rangle$ having same energy as $|\phi_{ii}\rangle$, we are led to conclude that $|\phi_{ii}\rangle$ cannot evolve with time because there is no such state. Indeed, momentum conservation through Coulomb and exchange processes imposes $\mathbf{Q}_m+\mathbf{Q}_n=\mathbf{Q}_i+\mathbf{Q}_j=2\mathbf{K}$. This leads us to set $\mathbf{Q}_i=\mathbf{K}+\mathbf{q}$, $\mathbf{Q}_j=\mathbf{K}-\mathbf{q}$ and $\mathbf{Q}_m=\mathbf{K}+\mathbf{q}'$, $\mathbf{Q}_n=\mathbf{K}-\mathbf{q}'$. If we now impose a strict energy conservation for process staying in the ν_0 relative motion ground state, we must have $\mathbf{Q}_m^2+\mathbf{Q}_n^2=\mathbf{Q}_i^2+\mathbf{Q}_j^2$. This leads to $|\mathbf{q}|=|\mathbf{q}'|$. The momenta $(\mathbf{Q}_m,\mathbf{Q}_n)$ of the exciton pair having same energy and momentum as $(\mathbf{Q}_i,\mathbf{Q}_j)$, evolve on a sphere of radius q, its center being at $\mathbf{K}$ from the momentum origin, $\mathbf{K}$ being possibly set equal to zero due to referential invariance.

This shows that, for two ground state excitons having same momentum, i.e., for $\mathbf{q}=0$, the final state exactly having the same energy corresponds to $\mathbf{q}'=0$: it is identical to the initial state so that this initial state does not evolve with time. It is however clear that, since $|\phi_{ii}\rangle$ is *not* H eigenstate, it has to change with time. Evolution toward final states with different energy must exist. The question now is what are the states, which control the time evolution of two ground-state excitons when these excitons have the same center-of-mass momentum?

Actually, this difficulty can be somehow separated from the ones linked to the exciton composite nature. This is why we are going to tackle it first by considering "boson-excitons," i.e., excitons taken as elementary bosons. We will show that, since the Fermi golden rule in its standard form imposes energy conservation at the $1/t$ scale only, while the exciton center-of-mass momenta form a quasicontinuum at the L^{-1} scale, we can still have many states within $1/t$ from the initial state energy provided that $1/t$ is not too small. This will lead us to discuss the problem of sample volume possibly replaced by coherence volume as well as the problem of "large-time limit" with respect to the exciton coherence time.

III. TIME EVOLUTION OF TWO-BOSON-EXCITON STATE

The effective Hamiltonian of excitons taken as elementary bosons splits as $H=H_0+V$, where H_0 is the free part H_X of this exciton Hamiltonian, while V is the effective potential V_{XX} between two boson excitons. We consider an initial state $|o\rangle$ made of two boson excitons, eigenstate of H_0. Under V_{XX}, it becomes $|o_t\rangle$. We first look for the V expansion of $|o_t\rangle$—a problem we studied some years ago.[45]

A. Interaction expansion of the $|o_t\rangle$ state

To calculate the time evolution of the $|o\rangle$ state, we are going to use a procedure which can be extended to composite bosons. The integral representation of the exponential[47] allows us to write this time evolution as

$$|o_t\rangle = e^{-iHt}|o\rangle = \int_{-\infty}^{+\infty} \frac{dx}{(-2i\pi)} \frac{e^{-i(x+i\eta_+)t}}{x+i\eta_+-H}|o\rangle, \tag{3.1}$$

where η_+ is an arbitrary positive constant, not necessarily small. Contributions induced by the interaction potential V follow from the standard key identity for many-body effects when $H=H_0+V$, namely,

$$\frac{1}{a-H} = \frac{1}{a-H_0} + \frac{1}{a-H} V \frac{1}{a-H_0}. \tag{3.2}$$

We iterate this equation and, in front of each $(a-H_0)^{-1}$ factor, we insert the closure relation $\Sigma_n|n\rangle\langle n|=1$ constructed upon the H_0 eigenstates, $(H_0-\epsilon_n)|n\rangle=0$. This gives

$$|o_t\rangle = \int_{-\infty}^{+\infty} \frac{dx}{(-2i\pi)} \frac{e^{-i(x+i\eta_+)t}}{x+i\eta_+-\epsilon_o}|o_x\rangle, \tag{3.3}$$

where, up to second order in V,

$$|o_x\rangle = |o\rangle + \sum_m |m\rangle \left[\frac{V_{mo}}{x+i\eta_+-\epsilon_m} + \sum_k \frac{V_{mk}V_{ko}}{(x+i\eta_+-\epsilon_m)(x+i\eta_+-\epsilon_k)} + \cdots \right], \tag{3.4}$$

with $V_{mk}=\langle m|V|k\rangle$, the states $m=o$ and $k=o$ being included in the sum. Integration using the residue theorem readily gives[45]

$$|o_t\rangle = e^{-i\epsilon_o t}|o\rangle + \sum_m |m\rangle[\mathcal{V}_{mo}^{(1)}(t) + \mathcal{V}_{mo}^{(2)}(t) + \cdots], \tag{3.5}$$

where the first-order term in the interaction reads

$$\mathcal{V}_{mo}^{(1)}(t) = V_{mo} \frac{e^{-i\epsilon_o t} - e^{-i\epsilon_m t}}{\epsilon_o - \epsilon_m}, \tag{3.6}$$

while the second-order term is given by

$$\mathcal{V}_{mo}^{(2)}(t) = \sum_k V_{mk}V_{ko} \left[\frac{e^{-i\epsilon_o t}}{(\epsilon_o-\epsilon_m)(\epsilon_o-\epsilon_k)} + \frac{e^{-i\epsilon_k t}}{(\epsilon_k-\epsilon_o)(\epsilon_k-\epsilon_m)} + \frac{e^{-i\epsilon_m t}}{(\epsilon_m-\epsilon_o)(\epsilon_m-\epsilon_k)} \right], \tag{3.7}$$

the values for $\epsilon_m=\epsilon_o$ or $\epsilon_k=\epsilon_o$ being simply the limit of the corresponding ratios when ϵ_m or ϵ_k go to ϵ_o.

The first-order term in the interaction brings a function $\Delta_t(\epsilon)$ which, for $\epsilon\neq 0$, is defined as

$$\Delta_t(\epsilon) = \frac{e^{i\epsilon t}-1}{\epsilon} = 2i\pi e^{i\epsilon t/2}\delta_t(\epsilon), \tag{3.8}$$

while, for $\epsilon=0$, it reduces to its $\epsilon\to 0$ limit, namely, $\Delta_t(0)=it$. This function cancels for $t=0$ whatever ϵ, while for large t it differs from zero for ϵ small compared to $1/t$ only. Indeed $\delta_t(\epsilon)$, equal to $(\pi\epsilon)^{-1}\sin(\epsilon t/2)$, is one of the various forms of delta function, with width $(2/t)$. As a result of the uncertainty principle, $\Delta_t(\epsilon)$ insures energy conservation at the $(1/t)$ scale.

Since $(\epsilon_m-\epsilon_o)^{-1}(\epsilon_m-\epsilon_k)^{-1}$ also reads $[(\epsilon_m-\epsilon_o)^{-1}-(\epsilon_m-\epsilon_k)^{-1}](\epsilon_o-\epsilon_k)^{-1}$, the second-order term, given by Eq. (3.7), brings a function $\Delta_t(\epsilon,\epsilon')$ which can be written in three different ways in terms of $\Delta_t(\epsilon)$, namely,

$$\Delta_t(\epsilon,\epsilon') = \frac{\Delta_t(\epsilon-\epsilon') - \Delta_t(\epsilon)}{\epsilon'}, \tag{3.9}$$

$$= \frac{\Delta_t(\epsilon-\epsilon') - e^{i\epsilon t}\Delta_t(-\epsilon')}{\epsilon}, \tag{3.10}$$

$$= \frac{\Delta_t(\epsilon) - e^{i(\epsilon-\epsilon')t}\Delta_t(\epsilon')}{\epsilon-\epsilon'}. \tag{3.11}$$

As $\Delta_t(\epsilon)$ stays finite when ϵ goes to zero, $\Delta_t(\epsilon,\epsilon')$ also stays finite when ϵ or ϵ' goes to zero, its value when they are both equal to zero reducing to $\Delta_t(0,0)=t^2/(2!)$.

This leads us to rewrite the time evolution of state $|o\rangle$ given in Eq. (3.5) as[45]

$$|o_t\rangle = e^{-i\epsilon_o t}\Bigg\{|o\rangle - \sum_m |m\rangle\Bigg[V_{mo}\Delta_t(\epsilon_o-\epsilon_m) + \sum_k V_{mk}V_{ko}\Delta_t(\epsilon_o-\epsilon_m,\epsilon_k-\epsilon_m)+\cdots\Bigg]\Bigg\}. \tag{3.12}$$

It can be of interest to note that, in the large t limit, i.e., for $\Delta_t(\epsilon_o-\epsilon_k)$ small for any $k\neq o$, the $|o\rangle$ prefactor in Eq. (3.12) reduces to

$$e^{-i\epsilon_o t}\left[1 - itV_{oo} + \frac{1}{2}(-itV_{oo})^2 - it\sum_{k\neq o}\frac{|V_{ko}|^2}{\epsilon_o-\epsilon_k}+\cdots\right]. \tag{3.13}$$

This expansion casts as $\exp(-i\tilde{\epsilon}_o t)$, where $\tilde{\epsilon}_o$ is the V expansion of the $|o\rangle$ state energy for the Hamiltonian H_0+V, namely,[46]

$$\tilde{\epsilon}_o = \epsilon_o + V_{oo} + \sum_{k\neq o}\frac{|V_{ko}|^2}{\epsilon_o-\epsilon_k}+\cdots. \tag{3.14}$$

B. Probability to reach another state

The probability for the $|o\rangle$ state to reach a state $|m\rangle$ different from $|o\rangle$ in its time evolution under the Hamiltonian H, is equal to $|\langle m|o_t\rangle|^2$. The $|m\rangle$ states within the energy scale $1/t$ from the initial state $|o\rangle$ control the time evolution of $|o\rangle$ in the large t limit since they are those for which $\Delta_t(\epsilon_m-\epsilon_o)$ does not reduce to zero for large t. As $\Delta_t(\epsilon\to 0)\simeq it$, the probability to reach one of these states, at the lowest order in the interaction, is given by

$$|\langle m|o_t\rangle|^2 \simeq |V_{mo}\Delta_t(\epsilon_o-\epsilon_m)|^2 \simeq 2\pi t|V_{mo}|^2\delta_t(\epsilon_m-\epsilon_o). \tag{3.15}$$

The singular delta behavior in fact disappears from the transition rate per unit time when many states have an energy within $1/t$ from ϵ_o so that sum over m must be taken in the above equation. Equation (3.15) thus leads to the well-known transition rate to leave the initial state $|o\rangle$,

$$\frac{1}{T_o} = \frac{1}{t}\sum_m |\langle m|o_t\rangle|^2 = 2\pi\sum_m |V_{mo}|^2\delta_t(\epsilon_m-\epsilon_o), \tag{3.16}$$

as given by the Fermi golden rule.[45–47]

By contrast, if all $|m\rangle$ states coupled to $|o\rangle$ by one interaction are far away in energy from $|o\rangle$ at the $1/t$ scale, i.e., if all $\Delta_t(\epsilon_m-\epsilon_o)$ for $m\neq o$ are essentially equal to zero when $t\to+\infty$, we must turn to the second-order term in Eq. (3.12) to possibly get a nonzero probability to leave the initial state $|o\rangle$. Equations (3.11) and (3.12) then give

$$|\langle m|o_t\rangle|^2 \simeq \left|\sum_k \frac{V_{mk}V_{ko}}{\epsilon_k-\epsilon_o}e^{-i\epsilon_k t}\Delta_t(\epsilon_k-\epsilon_m)\right|^2. \tag{3.17}$$

Due to the delta function included in $\Delta_t(\epsilon)$, the above sum is *de facto* restricted to states $|k\rangle$ close in energy to the final state $|m\rangle$. Note that, in this case, the denominator $(\epsilon_k-\epsilon_o)$ in Eq. (3.17) is never small at the $1/t$ scale because this result by construction holds when there is no state close in energy to $|o\rangle$.

This shows that the time evolution of an initial state $|o\rangle$ is *a priori* controlled by transitions toward states $|m\rangle$ which are coupled to $|o\rangle$ by one interaction and which, in addition, are close in energy to $|o\rangle$ at the $1/t$ scale. If such states do not exist, transitions to states $|m\rangle$ different from $|o\rangle$ are nevertheless possible by using a second interaction. Intermediate states $|k\rangle$ in this two-interaction process are then required to have an energy in the $1/t$ neighborhood of the final state $|m\rangle$ of interest. If such $|k\rangle$ states do not exist, a third interaction is needed, and so on.

Before going further, let us make a few comments.

(i) As physically required, the probability $|\langle m|o_t\rangle|^2$ to go from state $|o\rangle$ to a state $|m\rangle$ different from $|o\rangle$ goes to zero at all orders in V, when t goes to zero, the function $\Delta_t(\epsilon)$ reducing to zero for $t=0$. As shown below, this contrasts with composite excitons, due to the overcompleteness of the exciton basis.

(ii) For large t, the probability $|\langle m|o_t\rangle|^2$ to reach a state $|m\rangle$ has a linear dependence in t when this probability is controlled by linear terms in the interaction. This happens when states with energy within $1/t$ from the initial state $|o\rangle$ are coupled to it by one V only. Transitions from state $|o\rangle$ can then be considered as "easy." This shows up through a transition rate behavior. By contrast, when states with energy within $1/t$ from the initial state do not exist, the probability to reach a given final state $|m\rangle$ can nevertheless differ from zero through second order processes, if this final state lies within a quasicontinuum at the $1/t$ scale. However, the probability to reach this final state then is not proportional to t anymore as in the "easy" situation when the initial state lies in a quasicontinuum, but reaches a finite value.

(iii) When the state characteristics contain a center-of-mass momentum, their energies *de facto* form a quasicontinuum since this momentum scales as L^{-1} where L is the sample size. The number of states, within a $1/t$ energy scale, possibly entering first order processes then seems to depend upon whether the large time limit is taken before or after the large sample limit. Physically, the sample size is large but given, while t depends on the experimental set up. With in-

creasing t, we should thus always end with a time evolution controlled by second-order processes in the interaction. We must then note that, due to additional inelastic processes, the size which really controls the quantization of the exciton center-of-mass momentum is not the sample size but the exciton coherence length $L_{\rm coh}$. The $|m\rangle$ state energy then scales as $(2\pi/L_{\rm coh})^2/2M_X=1/\tau_{\rm coh}$. In order to get a time evolution controlled by second-order processes, we should not have exciton state lying within $1/t$ from the initial state. This imposes $1/t \ll 1/\tau_{\rm coh}$, i.e., t much larger than $\tau_{\rm coh}$. Since experiments on excitons performed at a time scale much larger than their coherence time are physically meaningless, the relevant time evolution of exciton states is thus going to be controlled by first-order processes, even in the case of two ground-state excitons having same center-of-mass momentum.

C. Lifetime of state $|o\rangle$

In addition to the probability to reach a state $|m\rangle$ different from the initial state, another quantity of physical interest is the lifetime of state $|o\rangle$. It is obtained from the probability $|\langle o|o_t\rangle|^2=\langle o_t|o\rangle\langle o|o_t\rangle$ to stay in $|o\rangle$. By using the closure relation for $|m\rangle$ states, the norm $\langle o_t|o_t\rangle$ staying constant with time, we find that this probability also reads

$$|\langle o|o_t\rangle|^2 = \langle o_t|\left[1 - \sum_{m\neq o} |m\rangle\langle m|\right]|o_t\rangle = 1 - \sum_{m\neq o} |\langle m|o_t\rangle|^2. \tag{3.18}$$

By writing it as $e^{-t/\tau_o} \simeq 1-t/\tau_o$, we end, using Eq. (3.15), with the standard expression for the state $|o\rangle$ lifetime, namely,

$$\frac{1}{\tau_o} = 2\pi \sum_{m\neq o} |V_{mo}|^2 \delta_t(\epsilon_m - \epsilon_o). \tag{3.19}$$

As physically expected, this lifetime is directly related to the transition rate to leave the $|o\rangle$ state.

The above result is *a priori* valid for $|o\rangle$ lying in a $1/t$ continuum from ϵ_o, this continuum being coupled to $|o\rangle$ by one interaction scattering only. If not, second-order processes would be necessary. Equation (3.17) should then be used for $|\langle m|o_t\rangle|^2$, and $|\langle o|o_t\rangle|^2$ would not cast as $(1-t/\tau_0)$ anymore. However, as discussed above, experiments done on a t scale small compared to the coherence time avoid such difficulty. The question now is to reach conceptually similar conclusions for composite excitons. Since the above results crucially depend on the existence of a potential V through its matrix elements V_{ij}, a new procedure which does not require to split the Hamiltonian H as H_X+V_{XX} is necessary.

IV. COULOMB SCATTERINGS BETWEEN TWO-EXCITON STATES

The above discussion evidences that states coupled to the initial state by one interaction play a key role in the time evolution of this initial state. A simple extension of this idea to composite excitons however is far from trivial already, due to difficulty in properly defining "state coupled to a given two-exciton state." This difficulty comes from the exciton composite nature which makes the N-exciton states, an overcomplete set of N electron-hole pairs when N is larger than 1. In this section, we consider the various possible states scattered from a two-exciton state and carefully analyze their physical meaning.

A. "Naïve" scattered state

While $(H-E_i)B_i^\dagger|v\rangle=0$, so that the one-exciton states $B_i^\dagger|v\rangle$ are H eigenstates, for two excitons we do have [see Eq. (A5) in the Appendix]

$$HB_i^\dagger B_j^\dagger|v\rangle = (B_i^\dagger H + E_i B_i^\dagger + V_i^\dagger)B_j^\dagger|v\rangle = E_{ij}B_i^\dagger B_j^\dagger|v\rangle + V_i^\dagger B_j^\dagger|v\rangle, \tag{4.1}$$

with the indices i and j possibly exchanged in the last term. This leads us to introduce the "naïve scattered state" $|C_{ij}\rangle$ defined as

$$|C_{ij}\rangle = (H-E_{ij})|\phi_{ij}\rangle \tag{4.2}$$

$$= V_i^\dagger B_j^\dagger|v\rangle = V_j^\dagger B_i^\dagger|v\rangle. \tag{4.3}$$

$|C_{ij}\rangle$ can be seen as the state coupled to $|\phi_{ij}\rangle$ by *one* Coulomb process. However, note that, unlike in usual cases, it is not possible to cancel this scattered state by canceling the interaction because Coulomb interaction, through its electron-hole part, also enters the exciton energy E_i: if we drop the Coulomb interaction in $V_i^\dagger$, we also destroy the exciton.

This scattered state can be written in terms of the interaction scatterings of the coboson theory[35] (see Appendix) as

$$|C_{ij}\rangle = \sum_{p,q} \xi\begin{pmatrix} q & j \\ p & i \end{pmatrix}|\phi_{pq}\rangle. \tag{4.4}$$

It *a priori* contains contributions from *all* two-exciton states including $(p,q)=(i,j)$.

By using the scalar product of two-exciton states given in Eq. (2.5), the $|\phi_{mn}\rangle$ part of this scattered state follows from

$$\langle\phi_{mn}|C_{ij}\rangle = \hat{\xi}\begin{pmatrix} n & j \\ m & i \end{pmatrix} + (i\leftrightarrow j) = 2\hat{\xi}_{mnij}, \tag{4.5}$$

$$\hat{\xi}\begin{pmatrix} n & j \\ m & i \end{pmatrix} = \xi\begin{pmatrix} n & j \\ m & i \end{pmatrix} - \xi^{\rm in}\begin{pmatrix} n & j \\ m & i \end{pmatrix}, \tag{4.6}$$

with $\hat{\xi}_{mnij}$ defined in terms of $\hat{\xi}\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right)$ as in Eq. (2.5).

Equations (4.2) and (4.5) lead us to understand $\hat{\xi}_{mnij}$ as the effective scattering for the $|\phi_{ij}\rangle$ to $|\phi_{mn}\rangle$ transition. As seen from Eq. (4.6), it contains direct as well as exchange processes. Note that the carrier-exchange part $\xi^{\rm in}\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right)$ of this scattering is not symmetrical since it takes place *after* Coulomb interaction [see Eq. (A7)]. The "in" and "out" exchange scatterings for carrier exchanges taking place after and before interaction are equal for energy conserving processes only [see Eq. (A8)].

Equation (4.5) moreover shows that, since $\hat{\xi}\left(\begin{smallmatrix} j & j \\ i & i \end{smallmatrix}\right)$ is not equal to zero, $|C_{ij}\rangle$ contains a nonzero contribution on $|\phi_{ij}\rangle$. This contribution must be somehow removed when thinking of "other" two-exciton states coupled to the initial state $|\phi_{ij}\rangle$

by one Coulomb process. Let us now show that this extraction can *a priori* be done in various ways.

B. Scattered state "different" from the initial state

Since the indices (p,q) in the sum of Eq. (4.4) can be taken equal to (i,j), a simple idea to get rid of the $|\phi_{ij}\rangle$ part in the scattered state $|C_{ij}\rangle$ is to extract the $|\phi_{ij}\rangle$ term from the sum. This leads us to remove two terms when $(i\neq j)$,

$$|C_{i\neq j}^{\neq}\rangle = \left[H - E_{ij} - \xi\begin{pmatrix} j & j \\ i & i \end{pmatrix} - \xi\begin{pmatrix} i & j \\ j & i \end{pmatrix}\right]|\phi_{ij}\rangle$$

$$= \sum_{(p,q)\neq(i,j)} \xi\begin{pmatrix} q & j \\ p & i \end{pmatrix}|\phi_{pq}\rangle, \tag{4.7}$$

while for $i=j$, we only have to remove $\xi\left(\begin{smallmatrix} i & i \\ i & i \end{smallmatrix}\right)|\phi_{ii}\rangle$. The scalar product with a two-exciton state $|\phi_{mn}\rangle$ can however be written in the same compact form for $(i\neq j)$ or $(i=j)$,

$$\langle\phi_{mn}|C_{ij}^{\neq}\rangle = 2\xi_{mnij}^{\neq},$$

$$\xi_{mnij}^{\neq} = \hat{\xi}_{mnij} - (1-\delta_{i,j}/2)\xi_{ijij}(\delta_{m,i}\delta_{n,j} + \delta_{m,j}\delta_{n,i} - 2\lambda_{mnij}). \tag{4.8}$$

This new scattering also splits into direct and exchange contributions according to

$$\xi_{mnij}^{\neq} = [\xi_{mnij} - (1-\delta_{i,j}/2)(\delta_{m,i}\delta_{n,j} + \delta_{m,j}\delta_{n,i})\xi_{ijij}]$$
$$- [\xi_{mnij}^{\text{in}} - (2-\delta_{i,j})\lambda_{mnij}\xi_{ijij}], \tag{4.9}$$

where ξ_{mnij} and λ_{mnij} are defined in terms of $\xi\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right)$ and $\lambda\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right)$ in the same way as for $\hat{\xi}$ in Eq. (4.5). The effective scattering $\xi_{mnij}^{\neq}$ obtained through this procedure differs from $\hat{\xi}_{mnij}$ obtained through $\langle\phi_{mn}|C_{ij}\rangle$. Since we have in mind transition toward two-exciton states "different" from $|\phi_{ij}\rangle$, the scattered state $|C_{ij}^{\neq}\rangle$, with $(p,q)=(i,j)$ excluded from the sum, can appear at first as more relevant than $|C_{ij}\rangle$.

Actually, to exclude $(p,q)=(i,j)$ from the $|C_{ij}\rangle$ sum is not yet enough due to the nonorthogonality of N-exciton states. Indeed, for $(m,n)=(i,j)$, the first bracket in Eq. (4.9) cancels, but not the second one so that $\langle\phi_{ij}|C_{ij}^{\neq}\rangle\neq 0$: the scattered state $|C_{ij}^{\neq}\rangle$, with $(p,q)=(i,j)$ excluded, still contains $|\phi_{ij}\rangle$ contributions.

C. Scattered state perpendicular to the initial state

Due to the overcompleteness of the two-exciton-state basis, there is one way only to get rid of the $|\phi_{ij}\rangle$ part in the scattered state $|C_{ij}\rangle$. This is through the projector over the subspace perpendicular to $|\phi_{ij}\rangle$,

$$P_{\perp}^{(ij)} = I - \frac{|\phi_{ij}\rangle\langle\phi_{ij}|}{\langle\phi_{ij}|\phi_{ij}\rangle}, \tag{4.10}$$

as easy to check from $P_{\perp}^{(ij)}|\phi_{ij}\rangle=0$. In the usual case, i.e., for $H=H_0+V$ and $(H_0-\epsilon_n)|n\rangle=0$, the projector over the subspace perpendicular to $|o\rangle$ reduces to $P_{\perp}^{(o)}=\Sigma_{n\neq o}|n\rangle\langle n|$. To get an explicit form of $P_{\perp}^{(ij)}$ in the case of composite boson excitons, we can replace I in Eq. (4.10) by the closure relation for two-pair states given in Eq. (2.3). We then see that $P_{\perp}^{(ij)}$ still contains a $|\phi_{ij}\rangle\langle\phi_{ij}|$ contribution:

(i) For $i\neq j$, we can either have $(p=i,\ q=j)$ or $(p=j,\ q=i)$ in the sum of Eq. (2.3) so that, due to Eq. (2.5), the $|\phi_{ij}\rangle\langle\phi_{ij}|$ prefactor in $P_{\perp}^{(ij)}$ reduces to

$$\left(\frac{1}{2!}\right)^2 2 - \frac{1}{\langle\phi_{ij}|\phi_{ij}\rangle} = \frac{1}{2} - \frac{1}{1-2\lambda_{ijij}} \simeq -\frac{1}{2}, \tag{4.11}$$

since λ_{ijij} is negligible in front of 1 in the large sample limit, Pauli scatterings between bound state excitons being of the order of $(a_X/L)^D$, where a_X is the Bohr radius and L is the sample size.

(ii) For $i=j$, we can only have $p=q=i$ so that the prefactor of $|\phi_{ii}\rangle\langle\phi_{ii}|$ is equal to

$$\left(\frac{1}{2!}\right)^2 - \frac{1}{2-2\lambda_{iiii}} \simeq -\frac{1}{4}, \tag{4.12}$$

which roughly is twice smaller.

This shows that the projector $P_{\perp}^{(ij)}$ over the subspace perpendicular to $|\phi_{ij}\rangle$ always has a nonzero $|\phi_{ij}\rangle\langle\phi_{ij}|$ part. Such a contribution, which can appear strange at first, is directly linked to the overcompleteness of exciton states and their resulting nonorthogonality. The $|\phi_{ij}\rangle\langle\phi_{ij}|$ part of $P_{\perp}^{(ij)}$ is in fact necessary to remove the various $|\phi_{ij}\rangle$ parts included in all other $|\phi_{pq}\rangle$'s. This shows that a full-proof elimination of a given $|\phi_{ij}\rangle$ state from an expansion is nontrivial at all. There is one way only to do it: the projection over the subspace perpendicular to $|\phi_{ij}\rangle$ through $P_{\perp}^{(ij)}$ defined in Eq. (4.10).

This understanding leads us to introduce the scattered state in the subspace perpendicular to the initial state. Due to Eqs. (4.2), (4.4), and (4.10), it reads

$$|C_{ij}^{\perp}\rangle = P_{\perp}^{(ij)}|C_{ij}\rangle = (H - \langle H\rangle_{ij})|\phi_{ij}\rangle, \tag{4.13}$$

where $\langle H\rangle_{ij}=\langle\phi_{ij}|H|\phi_{ij}\rangle/\langle\phi_{ij}|\phi_{ij}\rangle$ is the Hamiltonian mean value in the initial state. $\langle H\rangle_{ij}$ can be seen as the energy of the $|\phi_{ij}\rangle$ state at first order in the interaction. It precisely reads

$$\langle H\rangle_{ij} = E_{ij} - \frac{2\hat{\xi}_{ijij}}{1+\delta_{i,j}-2\lambda_{ijij}}. \tag{4.14}$$

This $|C_{ij}^{\perp}\rangle$ state, which, due to Eq. (4.4), also reads

$$|C_{ij}^{\perp}\rangle = P_{\perp}^{(ij)}\sum_{p,q}\xi\begin{pmatrix} q & j \\ p & i \end{pmatrix}|\phi_{pq}\rangle, \tag{4.15}$$

is the state in the subspace *perpendicular* to $|\phi_{ij}\rangle$, which can be reached by one Coulomb process from $|\phi_{ij}\rangle$. This state has no component on $|\phi_{ij}\rangle$ by construction. Due to Eqs. (4.5) and (4.10), its components on other two-exciton states are given by

$$\langle\phi_{mn}|C_{ij}^{\perp}\rangle = \langle\phi_{mn}|P_{\perp}^{(ij)}|C_{ij}\rangle = 2\xi_{mnij}^{\perp}, \tag{4.16}$$

$$\xi_{mnij}^{\perp} = \hat{\xi}_{mnij} - \frac{\langle\phi_{mn}|\phi_{ij}\rangle}{\langle\phi_{ij}|\phi_{ij}\rangle}\hat{\xi}_{ijij}. \tag{4.17}$$

The second term in $\xi_{mnij}^{\perp}$ allows the exact cancellation for $(m,n)=(i,j)$, as required; it is then as large as the first term.

By contrast, for (m,n) different from (i,j), the ratio is essentially of the order of the Pauli scattering λ_{mnij} so that this second term, in $(a_X/L)^D$, is negligible in front of $\hat{\xi}_{mnij}$.

To conclude this section, we have identified three quantities that we can associate to the scattering of excitons (i,j) into states (m,n), namely, $\langle\phi_{mn}|C_{ij}\rangle$, $\langle\phi_{mn}|C_{ij}^{\neq}\rangle$, and $\langle\phi_{mn}|C_{ij}^{\perp}\rangle$. The second one is definitely irrelevant because, although $(p,q)=(i,j)$ terms are removed from the sum, $|\phi_{ij}\rangle$ contributions still are in the scattered state. We are thus left with two effective Coulomb couplings, $\hat{\xi}_{mnij}$ and $\xi_{mnij}^{\perp}$. The first one corresponds to coupling toward the full scattered state $|C_{ij}\rangle$, while $\xi_{mnij}^{\perp}$ corresponds to coupling toward the part of this scattered state *restricted* to the orthogonal subspace.

The question now is what is the appropriate scattering for the time evolution of a two-exciton state? We are going to show that the answer is somewhat more elaborate than just choosing between these two relevant scatterings.

V. TIME EVOLUTION OF TWO-EXCITON STATE

Following the procedure used for elementary bosons, the time evolution of the two-exciton state $|\phi_{ij}\rangle=B_i^\dagger B_j^\dagger|v\rangle$ is given by

$$|\phi_{ij}(t)\rangle = e^{-iHt}|\phi_{ij}\rangle = \int \frac{dx}{(-2i\pi)}\frac{e^{-i(x+i\eta_+)t}}{x+i\eta_+-H}B_i^\dagger B_j^\dagger|v\rangle. \tag{5.1}$$

A. Coulomb expansion of the state $|\phi_{ij}(t)\rangle$

While Eq. (3.2) cannot be used to expand $|\phi_{ij}(t)\rangle$ in the interaction since the electron-hole Hamiltonian H does not cast as H_X+V_{XX}, we can instead use its equivalent for composite bosons given by Eq. (A9). Equation (3.3) then transforms into

$$|\phi_{ij}(t)\rangle = \int \frac{dx}{(-2i\pi)}e^{-i(x+i\eta_+)t}\left(B_i^\dagger+\frac{1}{x+i\eta_+-H}V_i^\dagger\right) \times\frac{1}{x+i\eta_+-H-E_i}B_j^\dagger|v\rangle. \tag{5.2}$$

As the last H can be replaced by E_j, integration using the residue theorem readily gives

$$|\phi_{ij}(t)\rangle = e^{-iE_{ij}t}|\phi_{ij}\rangle+\left(\frac{e^{-iE_{ij}t}}{E_{ij}-H}+\frac{e^{-iHt}}{H-E_{ij}}\right)V_i^\dagger B_j^\dagger|v\rangle. \tag{5.3}$$

This time evolution casts as

$$|\phi_{ij}(t)\rangle = e^{-iE_{ij}t}[|\phi_{ij}\rangle-\Delta_t(E_{ij}-H)V_i^\dagger B_j^\dagger|v\rangle], \tag{5.4}$$

where $\Delta_t(E_{ij}-H)$ formally is the function $\Delta_t(\epsilon)$ introduced in Eq. (3.8), with ϵ replaced by the operator $E_{ij}-H$.

The second term of $|\phi_{ij}(t)\rangle$ is first order at least in Coulomb process. Indeed Eqs. (A5) and (A6) give

$$V_i^\dagger B_j^\dagger|v\rangle = \sum_{mn}\xi\begin{pmatrix} n & j\\ m & i\end{pmatrix}|\phi_{mn}\rangle. \tag{5.5}$$

Note that this second term also has higher order contributions hidden in H. To get them, we again use Eq. (A9). This gives

$$\Delta_t(a-H)|\phi_{mn}\rangle = \frac{e^{i(a-H)t}-1}{a-H}|\phi_{mn}\rangle = (e^{i(a-H)t}-1)\left[B_m^\dagger+\frac{1}{a-H}V_m^\dagger\right] \times\frac{1}{a-H-E_m}B_n^\dagger|v\rangle. \tag{5.6}$$

The last H can be replaced by E_n, so that the term with $V_m^\dagger$ simply reads $(a-E_{mn})^{-1}\Delta_t(a-H)V_m^\dagger B_n^\dagger|v\rangle$. By using Eq. (5.4) for $e^{-iHt}|\phi_{mn}\rangle$ in the term with $B_m^\dagger$, we get

$$\Delta_t(a-H)|\phi_{mn}\rangle = \Delta_t(a-E_{mn})|\phi_{mn}\rangle + \frac{\Delta_t(a-H)-e^{i(a-E_{mn})t}\Delta_t(E_{mn}-H)}{a-E_{mn}} \times V_m^\dagger B_n^\dagger|v\rangle. \tag{5.7}$$

Using Eq. (3.10), we can rewrite it in a more compact form as

$$\Delta_t(a-H)|\phi_{mn}\rangle = \Delta_t(a-E_{mn})|\phi_{mn}\rangle + \Delta_t(a-E_{mn},H-E_{mn})V_m^\dagger B_n^\dagger|v\rangle. \tag{5.8}$$

If we now insert Eqs. (5.5) and (5.7) into Eq. (5.4), the time evolution of $|\phi_{ij}\rangle$ splits as

$$|\phi_{ij}(t)\rangle = e^{-iE_{ij}t}(|\phi_{ij}\rangle+|\phi'_{ij}\rangle+|\phi''_{ij}\rangle). \tag{5.9}$$

The first-order term in Coulomb scattering reduces to

$$|\phi'_{ij}\rangle = -\sum_{m,n}\xi\begin{pmatrix} n & j\\ m & i\end{pmatrix}\Delta_t(E_{ij}-E_{mn})|\phi_{mn}\rangle, \tag{5.10}$$

while all higher order terms read as

$$|\phi''_{ij}\rangle = -\sum_{m,n,p,q}\xi\begin{pmatrix} n & q\\ m & p\end{pmatrix}\xi\begin{pmatrix} q & j\\ p & i\end{pmatrix} \times\frac{\Delta_t(E_{ij}-H)-e^{i(E_{ij}-E_{pq})t}\Delta_t(E_{pq}-H)}{E_{ij}-E_{pq}}|\phi_{mn}\rangle. \tag{5.11}$$

The Coulomb scattering expansion of $|\phi_{ij}(t)\rangle$ follows from the iteration of the above equation using Eq. (5.8). The ξ^2 term is simply obtained by replacing H by E_{mn} in Eq. (5.11), while, to get the ξ^3 term, we also have to keep the second term of Eq. (5.8), and so on. By using Eq. (3.11), we find that the two first terms of the ξ expansion of $|\phi_{ij}(t)\rangle$ end by reading as

$$e^{iE_{ij}t}|\phi_{ij}(t)\rangle = |\phi_{ij}\rangle - \sum_{m,n} |\phi_{mn}\rangle \Bigg[\xi\begin{pmatrix} n & j \\ m & i \end{pmatrix} \Delta_t(E_{ij} - E_{mn}) + \sum_{p,q} \xi\begin{pmatrix} n & q \\ m & p \end{pmatrix} \xi\begin{pmatrix} q & j \\ p & i \end{pmatrix} \times \Delta_t(E_{ij} - E_{mn}, E_{pq} - E_{mn}) + \cdots \Bigg]. \quad (5.12)$$

We see that the structure of the $|\phi_{ij}(t)\rangle$ expansion in the interaction is very similar to the one found for $|o_t\rangle$ in Sec. III [see Eq. (3.12)], the *direct* Coulomb scattering ξ playing the role of the potential matrix element. Note that, at this stage, exchange processes do not appear yet.

B. Probability to reach another state

We now look for the $|\phi_{mn}\rangle$ component of $|\phi_{ij}(t)\rangle$. To get it in a compact form, it is actually simpler not to use the above expression of $|\phi_{ij}(t)\rangle$ but to come back to Eq. (5.4) and make $\Delta_t(E_{ij}-H)$ act on the left, through Eq. (5.8). This leads to

$$e^{iE_{ij}t}\langle\phi_{mn}|\phi_{ij}(t)\rangle = \langle\phi_{mn}|\phi_{ij}\rangle + \mathcal{V}_{mnij}(t), \quad (5.13)$$

where the interaction part is given by

$$\mathcal{V}_{mnij}(t) = -\Delta_t(E_{ij} - E_{mn})\langle\phi_{mn}|V_i^\dagger B_j^\dagger|v\rangle - \langle v|B_n V_m \Delta_t(E_{ij} - E_{mn}, H - E_{mn}) V_i^\dagger B_j^\dagger|v\rangle. \quad (5.14)$$

$\mathcal{V}_{mnij}(t)$ cancels for $t=0$ since all the Δ_t's cancel. However, since the first term of Eq. (5.13) does not cancel for $(m,n) \neq (i,j)$ due to carrier exchanges [see Eq. (2.5)], the scalar product $\langle\phi_{mn}|\phi_{ij}(t)\rangle$ differs from zero for $t=0$ so that this quantity cannot be used to get the probability to reach a state $|\phi_{mn}\rangle$ "different" from $|\phi_{ij}\rangle$ during the time evolution of this initial state.

As explained in the preceding section, the probability to reach a state different from $|\phi_{ij}\rangle$ must be associated to the $|\phi_{mn}\rangle$ component of the $|\phi_{ij}(t)\rangle$ state in the subspace perpendicular to $|\phi_{ij}\rangle$. Using Eqs. (5.4), (4.10), and (5.8), we then find

$$e^{iE_{ij}t}\langle\phi_{mn}|P_\perp|\phi_{ij}(t)\rangle = \mathcal{V}_{mnij}(t) - \frac{\langle\phi_{mn}|\phi_{ij}\rangle}{\langle\phi_{ij}|\phi_{ij}\rangle}\mathcal{V}_{ijij}(t). \quad (5.15)$$

The effect of $P_\perp$ in this matrix element is twofold: it of course removes the first term of Eq. (5.13); it *also* adds a term to $\mathcal{V}_{mnij}(t)$, as necessary for $\langle\phi_{mn}|P_\perp|\phi_{ij}(t)\rangle$ to exactly cancel when $(m,n)=(i,j)$.

Let us concentrate on the first-order term in Coulomb scattering of Eq. (5.15) right-hand side, as obtained by only keeping the first term of Eq. (5.14). For $(m,n)\neq(i,j)$, the scalar product $\langle\phi_{mn}|\phi_{ij}\rangle$ reduces to $(-2\lambda_{mnij})$ so that this first-order term reads $[-2\hat{\xi}_{mnij}\Delta_t(E_{ij}-E_{mn}) - 4it\lambda_{mnij}\hat{\xi}_{ijij}/(1+\delta_{i,j}-2\lambda_{ijij})]$. The second term contains λ_{mnij} which goes to zero with sample size as $(a_X/L)^D$ but it contains a t prefactor which goes to infinity in the large time limit so that to possibly drop this second term seems to depend on the way the $t\to\infty$ and $L\to\infty$ limits are taken. If we now turn to the first term, we note that the $(E_{mn}-E_{ij})$ energy difference scales as $(2\pi/L)^2/2M_X$, where the sample size L must be physically replaced by the exciton coherence length L_{coh}. This energy difference thus scales as $1/\tau_{\mathrm{coh}}$, where τ_{coh} is the coherence time. For t smaller than this coherence time, as experimentally required, there are many (m,n) states for which the energy difference $(E_{mn}-E_{ij})$ is essentially zero at the $1/t$ scale; consequently, $\Delta_t(E_{ij}-E_{mn})$ is of the order of it not only for (m,n) exactly equal to (i,j) but also for a large amount of other (m,n) states. For these states, the second term of Eq. (5.15) is then negligible because it is essentially $(a_X/L)^D$ smaller than the first one due to the additional λ_{mnij} Pauli scattering. This shows that we end with

$$|\langle\phi_{mn}|P_\perp|\phi_{ij}(t)\rangle|^2 \simeq |2\hat{\xi}_{mnij}\Delta_t(E_{ij}-E_{mn})|^2 \quad (5.16)$$

The above expression is similar to the one we have obtained for boson excitons in Eq. (3.15), provided that we take as effective scattering, the quantity $2\hat{\xi}_{mnij}$ defined in Eqs. (4.5) and (4.6), namely, $\langle\phi_{mn}|C_{ij}\rangle = \langle\phi_{mn}|V_i^\dagger B_j^\dagger|v\rangle$. This effective scattering contains direct and exchange contributions due to the exciton composite nature. Let us once more stress that the exchange part of this scattering is not symmetrical with respect to carrier exchange and interaction. Indeed, $[\hat{\xi}\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right)]^*$ differs from $\hat{\xi}\left(\begin{smallmatrix} j & n \\ i & m \end{smallmatrix}\right)$ as $[\xi^{\mathrm{in}}\left(\begin{smallmatrix} n & j \\ m & i \end{smallmatrix}\right)]^* = \xi^{\mathrm{out}}\left(\begin{smallmatrix} j & n \\ i & m \end{smallmatrix}\right)$. Consequently, this effective scattering can by no way be used in an effective exciton-exciton potential because it would produce a spurious non-Hermiticity in the resulting effective Hamiltonian.

This first-order term in Coulomb scattering is of physical relevance for $|\phi_{mn}\rangle$ state having an energy at $1/t$ from the $|\phi_{ij}\rangle$ energy. If this were not so, we would *a priori* have to turn to the second term of $\mathcal{V}_{mnij}(t)$ in Eq. (5.14). To get it, we first replace $\langle v|B_n V_m$ by $\Sigma_{p,q}\langle\phi_{pq}|\xi\left(\begin{smallmatrix} n & q \\ m & p \end{smallmatrix}\right)$, using Eq. (5.5), and then make $\Delta_t(E_{ij}-E_{mn}, H-E_{mn})$ act on the left so that, to lowest order in ξ, the corresponding H can be replaced by E_{pq}. Since $\langle\phi_{pq}|V_i^\dagger B_j^\dagger\rangle$ is just $2\hat{\xi}_{pqij}$, due to Eq. (4.5), we then end with

$$e^{iE_{ij}t}\langle\phi_{mn}|P_\perp|\phi_{ij}(t)\rangle \simeq -\sum_{p,q} 2\xi\begin{pmatrix} n & q \\ m & p \end{pmatrix}\hat{\xi}_{pqij} \times \Delta_t(E_{ij}-E_{mn}, E_{pq}-E_{mn}). \quad (5.17)$$

As, by construction, we now deal with $|\phi_{mn}\rangle$ state such that $(E_{ij}-E_{mn})$ is large at the $1/t$ scale, we find, using Eq. (3.11), that $\langle\phi_{mn}|P_\perp|\phi_{ij}(t)\rangle$ reduces to

$$|\langle\phi_{mn}|P_\perp|\phi_{ij}(t)\rangle|^2 \simeq \left| \sum_{p,q} 2\frac{\xi\begin{pmatrix} n & q \\ m & p \end{pmatrix}\hat{\xi}_{pqij}}{E_{pq}-E_{ij}} e^{-iE_{pq}t}\Delta_t(E_{pq}-E_{mn}) \right|^2. \quad (5.18)$$

We see that this second-order term again has the same structure as the one for elementary bosons [see Eq. (3.17)]. However, the Coulomb scatterings which enter this second order

term are far more elaborate: they are not just the product of the two effective scatterings appearing at first order, one of the two scatterings is dressed by carrier exchanges, not the other. A way to physically accept this result is to note that two exchanges reduce to an identity so that it can appear as natural to have carrier exchange appearing once only in a set of interaction processes.

C. Lifetime of state $|\phi_{ij}\rangle$

We now consider the lifetime of the $|\phi_{ij}\rangle$ state. It is related to the probability to stay in $|\phi_{ij}\rangle$. By using the definition of $P_\perp$ given in Eq. (4.10), we can rewrite this probability as

$$|\langle\phi_{ij}|\phi_{ij}(t)\rangle|^2 = \langle\phi_{ij}|e^{iHt}|\phi_{ij}\rangle\langle\phi_{ij}|e^{-iHt}|\phi_{ij}\rangle = \langle\phi_{ij}|\phi_{ij}\rangle\langle\phi_{ij}|e^{iHt}(1-P_\perp)e^{-iHt}|\phi_{ij}\rangle, \tag{5.19}$$

so that the probability to stay in the initial state $|\phi_{ij}\rangle$ also reads

$$\frac{|\langle\phi_{ij}|\phi_{ij}(t)\rangle|^2}{|\langle\phi_{ij}|\phi_{ij}\rangle|^2} = 1 - \frac{\langle\phi_{ij}|e^{iHt}P_\perp e^{-iHt}|\phi_{ij}\rangle}{\langle\phi_{ij}|\phi_{ij}\rangle}, \tag{5.20}$$

the second term canceling for $t=0$, as required.

The simplest way to calculate this second term is to note that $P_\perp = P_\perp^2$ and to insert the closure relation for two-exciton states given in Eq. (2.3) between the two $P_\perp$'s. This gives

$$\langle\phi_{ij}|e^{iHt}P_\perp^2 e^{-iHt}|\phi_{ij}\rangle = \frac{1}{(2!)^2}\sum_{m,n}|\langle\phi_{mn}|P_\perp|\phi_{ij}(t)\rangle|^2. \tag{5.21}$$

It is of importance to see that the $(1/2!)^2$ prefactor in place of $(\frac{1}{2!})$ in the closure relation of composite bosons brings an extra factor $\frac{1}{2}$ in the usual link between lifetime and sum of probabilities to reach two-exciton states different from the initial state.[42,43,53]

The above equation shows that, as for elementary bosons, a lifetime behavior for the probability to stay in the initial state $|\phi_{ij}\rangle$, i.e., a linear t dependence in Eq. (5.21), imposes a time evolution controlled by first-order processes in the interaction. This requires to have a set of states within $1/t$ in energy from the initial state. As in the case of elementary bosons, this always is so for experiments done on a time scale small compared to the exciton coherence time, which are the ones of physical relevance.

VI. LINK WITH OUR PREVIOUS WORKS

Let us end by making the link with our previous works on the time evolution of exciton states.[42,43] In these previous works, we wrote the time evolution of $|\phi_{ij}\rangle$ as

$$e^{-iHt}|\phi_{ij}\rangle = e^{-i\langle H\rangle_{ij}t}(|\phi_{ij}\rangle + |\tilde{\phi}_{ij}(t)\rangle). \tag{6.1}$$

Equation (4.14) shows that $\langle H\rangle_{ij}$ reduces to E_{ij} at lowest order in Coulomb scattering. It however is convenient to also keep the Coulomb part of $\langle H\rangle_{ij}$ because the state change $|\tilde{\phi}_{ij}(t)\rangle$ can then be written as

$$|\tilde{\phi}_{ij}(t)\rangle = [e^{i(\langle H\rangle_{ij}-H)t}-1]|\phi_{ij}\rangle = \frac{e^{i(\langle H\rangle_{ij}-H)t}-1}{\langle H\rangle_{ij}-H}[\langle H\rangle_{ij}-H]|\phi_{ij}\rangle. \tag{6.2}$$

By noting that $[\langle H\rangle_{ij}-H]|\phi_{ij}\rangle$ also reads $P_\perp H|\phi_{ij}\rangle$, we ended with a state change given by

$$|\tilde{\phi}_{ij}(t)\rangle = -\Delta_t(\langle H\rangle_{ij}-H)P_\perp H|\phi_{ij}\rangle, \tag{6.3}$$

where the function Δ_t is the one defined in Eq. (3.8). This brings energy conservation readily into the time evolution. Since $P_\perp H\phi_{ij}\rangle$ is first-order in Coulomb scattering already, the $|\tilde{\phi}_{ij}(t)\rangle$ first-order term in Coulomb interaction is then obtained by replacing $\langle H\rangle_{ij}$ by E_{ij} and H by E_{mn} when calculating $\langle\phi_{mn}|\tilde{\phi}_{ij}(t)\rangle$. The obtained result however holds for E_{mn} lying within $1/t$ from E_{ij} only.

Actually, in these previous works,[42,43] we did not question the existence of such states, even if in the very end, the initial state (0,0) was taken as two ground state excitons with same center-of-mass momentum. In particular, we did not discuss the proper way to take the large time limit in finite size sample nor the key role played by the coherence length in the physical evolution of exciton states.

We also wish to note that the introduction of a phase shift $\langle H\rangle_{ij}$, as done in Eq. (6.1), is quite convenient when the time evolution is ruled by first-order Coulomb processes between excitons because it readily brings energy conservation. By contrast, the extraction of $e^{-i\langle H\rangle_{ij}t}$ is not of particular interest when second-order terms in Coulomb scattering are needed because, to get these Coulomb second-order terms, we must rewrite $\Delta_t(\langle H\rangle_{ij}-H)$ in terms of e^{-iHt} and then use an equation similar to Eq. (5.4). This is why, to derive these second-order terms, we have had to work out a completely different procedure, which does not start with Eq. (6.1).

The main result of our previous works on the time evolution of exciton states actually was to point out for the first time, through a rather simple procedure, that, for time evolution controlled by first-order Coulomb processes, the link between lifetime and scattering rates is not the same for elementary and composite excitons. This difference was later shown[53] to come from difference in closure relations between elementary and composite particles—as also evidenced here through Eq. (5.21).

VII. CONCLUSION

In this work, we reconsider the time evolution of two-exciton states. We show that two regimes have *a priori* to be distinguished.

(i) If the initial state is unambiguously coupled by first-order Coulomb processes to states having the same energy, as in the case of two ground-state excitons having *different* center-of-mass momenta, the time evolution is controlled by these states. Using a new formalism, easy to extend to higher order Coulomb processes, we have rederived the lifetime of such two-exciton state as well as the transition rate toward states belonging to this quasicontinuum. This leads us to identify the effective Coulomb scattering appropriate to time evolution. It contains a mixture of direct and exchange Cou-

lomb processes. This mixture however is of no use in the construction of an effective potential for exciton-exciton interaction because it would bring a spurious non-Hermiticity in the resulting exciton effective Hamiltonian.

(ii) If there are no states with same energy as the initial state and coupled to it by first-order Coulomb process, second-order Coulomb processes are *a priori* required. We show that the mixture of direct and exchange Coulomb processes appearing in this second-order term is far more elaborate than the one appearing at first order. We also show that, within this second order regime, the exciton time evolution does not have the usual lifetime behavior.

However, in order to experience this second-order unusual regime, the time t of the experiment must be large compared to the exciton coherence time. Indeed, on the one hand, energy conservation resulting from time evolution at first order in the interaction, scales as $1/t$ due to the uncertainty principle for experiments performed over t. On the other hand, the scale for the quantification of the exciton center-of-mass momentum is more likely to be the exciton coherence length than the sample size, so that the exciton energy scale is more likely to be the inverse coherence time. Since experiments performed over t larger than the coherence time are physically meaningless, the time evolution in relevant experimental conditions must end by being controlled by first-order Coulomb processes only, *even* when the two ground state excitons have the same center-of-mass momentum, a configuration in which all the other two-exciton states formally have a higher energy.

APPENDIX: KEYS FROM COBOSON MANY-BODY THEORY

The many-body theory for composite bosons ("cobosons") made of two fermions that we have constructed,[35] is based on four commutators. Two commutators control fermion exchanges in the absence of fermion interaction. They read

$$[B_m, B_i^\dagger] = \delta_{m,i} - D_{mi}, \tag{A1}$$

$$[D_{mi}, B_j^\dagger] = \sum_n \left[\lambda \begin{pmatrix} n & j \\ m & i \end{pmatrix} + (i \leftrightarrow j) \right] B_n^\dagger |v\rangle, \tag{A2}$$

with $B_i^\dagger$ being the creation operator of one coboson in state i. From them, we can show that the scalar product of two-coboson states reads in terms of Pauli scattering $\lambda\binom{n\ j}{m\ i}$ as

$$\langle v|B_n B_m B_i^\dagger B_j^\dagger|v\rangle = \langle v|B_n(B_i^\dagger B_m + \delta_{m,i} - D_{mi})B_j^\dagger|v\rangle$$
$$= \delta_{m,i}\delta_{n,j} - \lambda \begin{pmatrix} n & j \\ m & i \end{pmatrix} + (i \leftrightarrow j), \tag{A3}$$

the $(i \leftrightarrow j)$ exchange being possibly replaced by $(m \leftrightarrow n)$, since $\lambda\binom{n\ j}{m\ i} = \lambda\binom{m\ i}{n\ j}$.

These Pauli scatterings also appear in

$$B_i^\dagger B_j^\dagger = -\sum_{m,n} \lambda \begin{pmatrix} n & j \\ m & i \end{pmatrix} B_m^\dagger B_n^\dagger, \tag{A4}$$

which results from the two ways to construct two cobosons out of two fermion pairs. Equation (A4) is one of the signatures of the overcompleteness of the coboson basis induced by their composite nature.

Two other commutators control fermion interactions in the absence of fermion exchanges. For coboson i being eigenstate of the system Hamiltonian, $(H-E_i)B_i^\dagger|v\rangle = 0$, they read

$$[H, B_i^\dagger] = E_i B_i^\dagger + V_i^\dagger, \tag{A5}$$

$$[V_i^\dagger, B_j^\dagger] = \sum_{m,n} \xi \begin{pmatrix} n & j \\ m & i \end{pmatrix} B_m^\dagger B_n^\dagger. \tag{A6}$$

By calculating the H matrix element in the two-coboson subspace, we generate "in" and "out" exchange interaction scatterings defined as

$$\xi^{\text{in}} \begin{pmatrix} n & j \\ m & i \end{pmatrix} = \sum_{p,q} \lambda \begin{pmatrix} n & q \\ m & p \end{pmatrix} \xi \begin{pmatrix} q & j \\ p & i \end{pmatrix} = \left[\xi^{\text{out}} \begin{pmatrix} j & n \\ i & m \end{pmatrix} \right]^*. \tag{A7}$$

They are linked to Pauli scattering through

$$\xi^{\text{in}} \begin{pmatrix} n & j \\ m & i \end{pmatrix} - \xi^{\text{out}} \begin{pmatrix} n & j \\ m & i \end{pmatrix} = (E_m + E_n - E_i - E_j) \lambda \begin{pmatrix} n & j \\ m & i \end{pmatrix}. \tag{A8}$$

This shows that these two exchange interaction scatterings are equal for energy-conserving processes.

A last equation of interest for correlations between cobosons is

$$\frac{1}{a-H} B_i^\dagger = B_i^\dagger \frac{1}{a-H-E_i} + \frac{1}{a-H} V_i^\dagger \frac{1}{a-H-E_i}, \tag{A9}$$

which follows from Eq. (A5). This equation allows us to make interaction expansion when the Hamiltonian does not split as H_0+V, as in the case of composite quantum particles.

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