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Role of the exchange of carriers in elastic exciton-exciton scattering in quantum wells

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A study of the elastic exciton-exciton Coulomb scattering in a semiconductor quantum well is presented, including the interexciton exchange of carriers and the spin degrees of freedom. The theoretical results show that electron-electron and hole-hole exchanges are the dominant mechanisms of interaction, while the classical direct term is negligible. The density-dependent homogeneous linewidth is calculated within the Born approximation and good agreement with the existing experimental data is obtained. Owing to the interexciton exchange of carriers, collisions lead to spin relaxation as actually observed in recent time-resolved photoluminescence experiments. [S0163-1829(98)06736-8]

I. INTRODUCTION

The optical spectra of semiconductor quantum wells near the fundamental absorption edge at low temperatures and moderated excitation densities are dominated by excitons. An excitonic resonance is characterized by the energy of the peak, the oscillator strength, and the linewidth. These quantities are significantly influenced by the presence of a finite density of excitons and free carriers. For low temperatures and for optical excitation resonant with the exciton energy, the population of free carriers can be neglected and therefore the density-dependent features are due to exciton-exciton interaction. It is known that with increasing excitation density, the exciton energy blue-shifts and the oscillator strength saturates.[1] The exciton-exciton scattering also produces the so-called collisional broadening, that is a density-dependent homogeneous linewidth. This effect has been observed in the four wave-mixing experiments in pump and probe configuration by Honold *et al.*[2] and in the photoluminescence measurements under resonant excitation by Deveaud *et al.*[3]

The problem of exciton-exciton scattering in quantum wells has been considered theoretically within two-dimensional[4,5] (2D) and quasi-2D models.[6] These articles, however, do not provide a complete description of the interaction process. In the paper of Feng and Spector,[4] the classical electrostatic dipole-dipole interaction is the only considered scattering mechanism, while the contribution due to exchange effects is not taken into account. Moreover, the authors study the properties of the scattering matrix elements, but they do not calculate the collisional broadening, which is a very significant physical observable. In a recent paper,[6] the previous model is extended to the quasi-2D case, including also the symmetry effect when two excitons are identical (exciton-exciton exchange), but still neglecting the inter-exciton exchange of carriers (fermion-fermion exchange). On the other hand, in the article by Manzke *et al.*,[5] these effects are formally considered, but it is not shown whether they are significant when compared to the direct interaction. Furthermore, the dependence of these scattering matrix elements on exciton wave vectors is not studied and the results for the density-dependent homogeneous broadening are actually very small compared to the existing experimental data.[2,3] Finally, in all the above mentioned models, the spin degrees of freedom are not taken into account.

The role of the spin degrees of freedom in an interacting exciton gas has been actually investigated in the recent experiments by Amand *et al.*[7] In particular, they have studied the time-resolved photoluminescence on a GaAs quantum well for different degrees of elliptical polarization of the exciting beam. They have shown that the time-resolved signal is characterized by a very fast decay, followed by a much slower one. The initial fast component disappears in the limiting case of circular polarization or for low excitation density. This interesting feature is explained as a result of the transfer from optically active to optically forbidden exciton states, which correspond to different spin states. According to Ref. 7, the inter-exciton exchange of carriers is the mechanism responsible for this density-dependent spin-relaxation process. In order to fit the observed dynamics, they derive kinetic equations by employing an effective phenomenological spin-spin Hamiltonian.

In this paper, we present a theoretical study of the elastic exciton-exciton scattering due to Coulomb interaction in a two-dimensional system. In the model, the inter-exciton exchange of carriers and the spin degrees of freedom are included. The scattering matrix elements deriving from a four-carrier Hamiltonian are calculated within a two-band envelope function formalism. We show that electron-electron and hole-hole exchange are largely dominant with respect to the classical electrostatic dipole-dipole interaction and to exciton-exciton exchange. We study the behavior of the matrix elements in momentum space. Within the Born approximation, we calculate the collisional broadening, which is found in good agreement with the existing experimental results.[2,3] Owing to the inter-exciton exchange of carriers, Coulomb scattering can lead to spin relaxation. For example, two interacting excitons in the same elliptically polarized state can scatter into dark states. We also provide a complete description of the allowed spin channels for scattering.

The paper is organized as follows. In Sec. II, we describe the two-dimensional theoretical model. In Sec. III the problem of the collisional broadening is considered. Conclusions are drawn in Sec. IV, while in the Appendix we show a few technical details.

II. 2D-THEORETICAL MODEL

A. General remarks

A two-dimensional exciton in the $1s$ state whose center of mass has wave vector $\boldsymbol{Q}$ is described by an envelope function,[8,9] which is the product of terms relating to the center of mass motion and the internal relative one, respectively:

$$\Psi_{\boldsymbol{Q}}(\boldsymbol{r_e},\boldsymbol{r_h})=\frac{1}{\sqrt{A}}\exp[i\boldsymbol{Q}\cdot(\beta_e\boldsymbol{r_e}+\beta_h\boldsymbol{r_h})]\times\sqrt{\frac{2}{\pi}}\frac{1}{\lambda_{2\mathrm{D}}}\exp\left(-\frac{|\boldsymbol{r_e}-\boldsymbol{r_h}|}{\lambda_{2\mathrm{D}}}\right), \quad (1)$$

where $\boldsymbol{r_e}$ and $\boldsymbol{r_h}$ are the in-plane position vectors for the electron and the hole respectively, while A represents the normalization area. The coefficients β_e, β_h are defined as $\beta_{e(h)}=m_{e(h)}/M$, where $m_{e(h)}$ is the electron (hole) effective mass and $M=m_e+m_h$. Finally, $\lambda_{2\mathrm{D}}$ is the two-dimensional Bohr radius of the $1s$ state; $\lambda_{2\mathrm{D}}=\epsilon_0\hbar^2/(2e^2\mu)$, where μ is the reduced mass and ϵ_0 the static dielectric constant.

Let us take the growth direction $\hat{z}$ as the quantization axis for the angular momentum. The conduction band has a spherical symmetry and therefore the total angular-momentum projection is given just by the spin $s_e\in\{\pm 1/2\}$. Concerning the valence bands, we observe that in ordinary III-V quantum wells the heavy-hole/light-hole splitting is comparable or even larger than the exciton binding energy and therefore the valence-band mixing effects are negligible,[7] contrary to the bulk case where the splitting is absent. As if we focus our attention on heavy-hole excitons, we can limit ourselves to consider the heavy-hole subspace only. We remember that heavy-hole states have a total angular-momentum projection $j_h\in\{\pm 3/2\}$. Therefore, for heavy-hole excitons, we have the four independent states: the dipole-active states $|J_z=\pm 1\rangle=|s_e=\mp 1/2;\ j_h=\pm 3/2\rangle$ and the dipole-forbidden dark states $|J_z=\pm 2\rangle=|s_e=\pm 1/2;\ j_h=\pm 3/2\rangle$.

For a generic exciton state $|S\rangle$ belonging to the above mentioned four-dimensional space, we define $\chi_S(s_e,j_h)=\langle s_e;j_h|S\rangle$. For example, $\chi_{J_z=1}=\delta_{s_e,-1/2}\delta_{j_h,3/2}$. In the case of excitation by elliptically polarized light, excitons are created in the elliptic state

$$|E_\alpha\rangle=\sin\,\alpha|+1\rangle+\cos\,\alpha|-1\rangle. \quad (2)$$

We notice that $|E_{\alpha+\pi/2}\rangle=\cos\,\alpha|+1\rangle-\sin\,\alpha|-1\rangle$. Therefore, $|E_\alpha\rangle$ and $|E_{\alpha+\pi/2}\rangle$ are two orthogonal elliptically polarized states. For particular values of α, we obtain the limiting cases of circular and linear polarization, namely $|E_0\rangle=|-1\rangle$, $|E_{\pi/2}\rangle=|+1\rangle$, $|E_{\pi/4}\rangle=|y\rangle$, $|E_{(3/4)\pi}\rangle=|x\rangle$.

In the present paper, we consider the elastic Coulomb scattering of $1s$ excitons by $1s$ excitons

$$(1s,\boldsymbol{Q},S)+(1s,\boldsymbol{Q}',S')\rightarrow(1s,\boldsymbol{Q}+\boldsymbol{q},S_f)+(1s,\boldsymbol{Q}'-\boldsymbol{q},S_f'). \quad (3)$$

As observed in the introduction, we are interested in the experiments of resonant optical excitation at low temperatures. In this paper, we limit ourselves to the channel described in Eq. (3). The justification lies in the assumption that, by resonant excitation, excitons are created in the $1s$ state with very small wave vector Q and thus with small kinetic energy. The energy of an exciton is given by the energy of the relative motion plus the kinetic energy of the center of mass. It is known that the $1s$ state is the ground exciton state of the electron-hole motion. In two dimensions, the other bound states $(2s,2p,\ldots)$ are very close to the continuum. In fact, the first excited state has a binding energy that is only 1/9 of the binding energy of the $1s$ state. This means that inelastic scattering channels towards different relative motion states are strongly suppressed, because of energy conservation. *A priori* biexciton bound state could contribute to the exciton collision broadening. In this paper, we do not consider the channels where biexciton are involved. By considering only the elastic $1s$ channel, we obtain good agreement with the experimental results. This, in the end, allows us to argue that the other channels play a marginal role.

Since electrons and holes are fermions, the wave function of a two-exciton state has to be antisymmetric under the exchange of the two electrons and, separately, under the exchange of the two holes. In the present model, we consider electrons and holes as different quasi-particles, not taking into account the so-called electron-hole exchange effects, as usually done in two-band many-body models.[10] The electron-hole exchange actually produces a very small splitting ($\approx$0.1 meV for a GaAs quantum well)[11] between the dipole-active and the dipole-forbidden states. As we are working in the framework of a two-band model within the effective mass approximation, we neglect also the effect of the valence-band mixing due to spin-orbit interaction. The electron-hole exchange and the spin-orbit interaction actually produce spin-relaxation mechanisms.[12–14] However, since the corresponding spin-flip times are very long (50–100 ps),[20] the above mentioned mechanisms can be reasonably neglected in the initial phase of the dynamics, which is dominated by the inter-exciton exchange of carriers.[7] Therefore, within the Hartree-Fock approximation, the two-exciton initial state in Eq. (3) is described by the following wave function:

$$\begin{aligned}\Phi^{SS'}_{\boldsymbol{QQ}'}(\boldsymbol{r_e},s_e,\boldsymbol{r_h},j_h,\boldsymbol{r_{e'}},s_{e'},\boldsymbol{r_{h'}},j_{h'})
&=\frac{1}{\sqrt{2}}\Bigg\{\frac{1}{\sqrt{2}}\Big[\Psi_{\boldsymbol{Q}}(\boldsymbol{r_e},\boldsymbol{r_h})\chi_S(s_e,j_h)\Psi_{\boldsymbol{Q}'}(\boldsymbol{r_{e'}},\boldsymbol{r_{h'}})\\
&\times\chi_{S'}(s_{e'},j_{h'})+\Psi_{\boldsymbol{Q}}(\boldsymbol{r_{e'}},\boldsymbol{r_{h'}})\chi_S(s_{e'},j_{h'})\Psi_{\boldsymbol{Q}'}(\boldsymbol{r_e},\boldsymbol{r_h})\\
&\times\chi_{S'}(s_e,j_h)\Big]-\frac{1}{\sqrt{2}}\Big[\Psi_{\boldsymbol{Q}}(\boldsymbol{r_{e'}},\boldsymbol{r_h})\chi_S(s_{e'},j_h)\\
&\times\Psi_{\boldsymbol{Q}'}(\boldsymbol{r_e},\boldsymbol{r_{h'}})\chi_{S'}(s_e,j_{h'})+\Psi_{\boldsymbol{Q}}(\boldsymbol{r_e},\boldsymbol{r_{h'}})\chi_S(s_e,j_{h'})\\
&\times\Psi_{\boldsymbol{Q}'}(\boldsymbol{r_{e'}},\boldsymbol{r_h})\chi_{S'}(s_{e'},j_h)\Big]\Bigg\}. \quad (4)\end{aligned}$$

We consider the following four-carrier Hamiltonian:

$$H=-\frac{\hbar^2}{2m_e}\nabla_e^2-\frac{\hbar^2}{2m_h}\nabla_h^2-\frac{\hbar^2}{2m_e}\nabla_{e'}^2$$
$$-\frac{\hbar^2}{2m_h}\nabla_{h'}^2-V(|\boldsymbol{r_e}-\boldsymbol{r_h}|)-V(|\boldsymbol{r_{e'}}-\boldsymbol{r_{h'}}|)+V(|\boldsymbol{r_e}-\boldsymbol{r_{e'}}|)$$
$$+V(|\boldsymbol{r_h}-\boldsymbol{r_{h'}}|)-V(|\boldsymbol{r_e}-\boldsymbol{r_{h'}}|)-V(|\boldsymbol{r_h}-\boldsymbol{r_{e'}}|), \tag{5}$$

where $V(r)=e^2/(\epsilon_0 r)$ is the Coulomb interaction energy, screened by the static dielectric constant ϵ_0. The scattering amplitude corresponding to the process in Eq. (3) is given by the matrix element

$$H_{SS'}^{S_fS_f'}(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})$$
$$=\int d^2\boldsymbol{r_e}\sum_{s_e}\int d^2\boldsymbol{r_h}\sum_{j_h}\int d^2\boldsymbol{r}_e'\sum_{s_{e'}}\int d^2\boldsymbol{r}_h'$$
$$\times\sum_{j_{h'}}\Phi_{\boldsymbol{QQ'}}^{SS'\star}(\boldsymbol{r_e},s_e,\boldsymbol{r_h},j_h,\boldsymbol{r_{e'}},s_{e'},\boldsymbol{r_{h'}},j_{h'})$$
$$\times H\Phi_{\boldsymbol{Q+qQ'-q}}^{S_fS_f'}(\boldsymbol{r_e},s_e,\boldsymbol{r_h},j_h,\boldsymbol{r_{e'}},s_{e'},\boldsymbol{r_{h'}},j_{h'}). \tag{6}$$

The approach we have followed to calculate the scattering amplitude has been suggested in an old paper by Haug.[15] After some algebra, we find that this matrix element is the sum of four contributions

$$H_{SS'}^{S_fS_f'}(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})=\langle S|S_f\rangle\langle S'|S_f'\rangle H_{\rm dir}(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})$$
$$+\langle S|S_f'\rangle\langle S'|S_f\rangle H_{\rm exch}^X(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})$$
$$+\mathcal{S}_{\rm exch}^e(S,S',S_f,S_f')H_{\rm exch}^e(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})$$
$$+\mathcal{S}_{\rm exch}^h(S,S',S_f,S_f')H_{\rm exch}^h(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q}). \tag{7}$$

$H_{\rm dir}$ is the direct Coulomb term which corresponds to the classical electrostatic interaction between the two excitons. On the other hand, $H_{\rm exch}^X$ is the term due to the exciton-exciton exchange (simultaneous exchange of the two identical electrons and the two identical holes). The third term $H_{\rm exch}^e$ is the term due to the electron-electron exchange, while $H_{\rm exch}^h$ is the analogous contribution arising from the hole-hole exchange. The factors $\mathcal{S}_{\rm exch}^e$ and $\mathcal{S}_{\rm exch}^h$ are given by the following spin-exchange sums

$$\mathcal{S}_{\rm exch}^e(S,S',S_f,S_f')=\sum_{s_e}\sum_{j_h}\sum_{s_{e'}}\sum_{j_{h'}}\chi_S^\star(s_e,j_h)$$
$$\times\chi_{S'}^\star(s_{e'},j_{h'})\chi_{S_f}(s_{e'},j_h)\chi_{S_f'}(s_e,j_{h'}), \tag{8}$$

$$\mathcal{S}_{\rm exch}^h(S,S',S_f,S_f')=\sum_{s_e}\sum_{j_h}\sum_{s_{e'}}\sum_{j_{h'}}\chi_S^\star(s_e,j_h)$$
$$\times\chi_{S'}^\star(s_{e'},j_{h'})\chi_{S_f}(s_e,j_{h'})\chi_{S_f'}(s_{e'},j_h). \tag{9}$$

The spin-exchange sums $\mathcal{S}_{\rm exch}^e$ and $\mathcal{S}_{\rm exch}^h$ satisfy the equalities

$$\mathcal{S}_{\rm exch}^h(S,S',S_f,S_f')=\mathcal{S}_{\rm exch}^e(S,S',S_f',S_f), \tag{10}$$

$$\mathcal{S}_{\rm exch}^{e(h)}(S,S',S_f,S_f')=\mathcal{S}_{\rm exch}^{e(h)\star}(S_f,S_f',S,S'). \tag{11}$$

If excitons are created by elliptically polarized light, then the spin states $|E_\alpha\rangle$, $|E_{\alpha+\pi/2}\rangle$, $|+2\rangle$ and $|-2\rangle$ are the proper basis to consider. It is crucial to remark that, due to electron-electron and hole-hole exchange, two interacting excitons in the same elliptic state E_α can scatter to different spin states. In particular, $\mathcal{S}_{\rm exch}^{e(h)}(E_\alpha,E_\alpha,\pm2,\mp2)\neq0$. On the other hand, the scattering of two excitons in the same circular state cannot change their spin polarization. A list of the allowed spin channels and of their corresponding spin-exchange factors $\mathcal{S}_{\rm exch}^e$ and $\mathcal{S}_{\rm exch}^h$ is shown in Table I.

The expression for the direct Coulomb integral is

$$H_{\rm dir}(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})=\int d^2\boldsymbol{r_e}\int d^2\boldsymbol{r_h}\int d^2\boldsymbol{r_{e'}}\int d^2\boldsymbol{r_{h'}}$$
$$\times\Psi_{\boldsymbol{Q}}^\star(\boldsymbol{r_e},\boldsymbol{r_h})\Psi_{\boldsymbol{Q'}}^\star(\boldsymbol{r_{e'}},\boldsymbol{r_{h'}})V_I(\boldsymbol{r_e},\boldsymbol{r_h},\boldsymbol{r_{e'}},\boldsymbol{r_{h'}})$$
$$\times\Psi_{\boldsymbol{Q+q}}(\boldsymbol{r_e},\boldsymbol{r_h})\Psi_{\boldsymbol{Q'-q}}(\boldsymbol{r_{e'}},\boldsymbol{r_{h'}}), \tag{12}$$

with

$$V_I(\boldsymbol{r_e},\boldsymbol{r_h},\boldsymbol{r_{e'}},\boldsymbol{r_{h'}})=V(|\boldsymbol{r_e}-\boldsymbol{r_{e'}}|)+V(|\boldsymbol{r_h}-\boldsymbol{r_{h'}}|)$$
$$-V(|\boldsymbol{r_e}-\boldsymbol{r_{h'}}|)-V(|\boldsymbol{r_h}-\boldsymbol{r_{e'}}|). \tag{13}$$

The term corresponding to the exciton-exciton exchange is given by

$$H_{\rm exch}^X(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})=\int d^2\boldsymbol{r_e}\int d^2\boldsymbol{r_h}\int d^2\boldsymbol{r_{e'}}\int d^2\boldsymbol{r_{h'}}\Psi_{\boldsymbol{Q}}^\star(\boldsymbol{r_e},\boldsymbol{r_h})$$
$$\times\Psi_{\boldsymbol{Q'}}^\star(\boldsymbol{r_{e'}},\boldsymbol{r_{h'}})V_I(\boldsymbol{r_e},\boldsymbol{r_h},\boldsymbol{r_{e'}},\boldsymbol{r_{h'}})$$
$$\times\Psi_{\boldsymbol{Q+q}}(\boldsymbol{r_{e'}},\boldsymbol{r_{h'}})\Psi_{\boldsymbol{Q'-q}}(\boldsymbol{r_e},\boldsymbol{r_h}). \tag{14}$$

$H_{\rm exch}^X$ differs from $H_{\rm dir}$ because of the simultaneous exchange $\boldsymbol{r_e}\leftrightarrow\boldsymbol{r_{e'}}$ and $\boldsymbol{r_h}\leftrightarrow\boldsymbol{r_{h'}}$ in the final state. $H_{\rm exch}^X$ is related to $H_{\rm dir}$ by the equality

$$H_{\rm exch}^X(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})=H_{\rm dir}(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{Q}'-\boldsymbol{Q}-\boldsymbol{q}). \tag{15}$$

The electron-electron exchange is given by

$$H_{\rm exch}^e(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})=-\int d^2\boldsymbol{r_e}\int d^2\boldsymbol{r_h}\int d^2\boldsymbol{r_{e'}}\int d^2\boldsymbol{r_{h'}}$$
$$\times\Psi_{\boldsymbol{Q}}^\star(\boldsymbol{r_e},\boldsymbol{r_h})\Psi_{\boldsymbol{Q'}}^\star(\boldsymbol{r_{e'}},\boldsymbol{r_{h'}})$$
$$\times V_I(\boldsymbol{r_e},\boldsymbol{r_h},\boldsymbol{r_{e'}},\boldsymbol{r_{h'}})\Psi_{\boldsymbol{Q+q}}(\boldsymbol{r_{e'}},\boldsymbol{r_h})$$
$$\times\Psi_{\boldsymbol{Q'-q}}(\boldsymbol{r_e},\boldsymbol{r_{h'}}). \tag{16}$$

The expression for $H_{\rm exch}^e$ differs from $H_{\rm dir}$ because of the exchange $\boldsymbol{r_e}\leftrightarrow\boldsymbol{r_{e'}}$ in the final state and a minus sign. In an analogous way, $H_{\rm exch}^h$ is obtained from $H_{\rm dir}$, by exchanging $\boldsymbol{r_h}\leftrightarrow\boldsymbol{r_{h'}}$ and by changing the sign. We observe that all the considered Coulomb integrals $H_{\rm dir}$, $H_{\rm exch}^X$, $H_{\rm exch}^e$, $H_{\rm exch}^h$ are real, because the complex conjugation is equivalent to the transformation $\boldsymbol{Q}\to-\boldsymbol{Q}$, $\boldsymbol{Q}'\to-\boldsymbol{Q}'$, $\boldsymbol{q}\to-\boldsymbol{q}$, which does

TABLE I. Allowed scattering spin channels and their corresponding spin-exchange factors $\mathcal{S}^e_{\rm exch}$ and $\mathcal{S}^h_{\rm exch}$, corresponding to electron-electron and hole-hole exchange, respectively. $|S\rangle$ and $|S'\rangle$ are the initial spin states, which scatter in the final ones $|S_f\rangle$, $|S'_f\rangle$. The considered basis is $\{|E_\alpha\rangle,|E_{\alpha+\pi/2}\rangle,|+2\rangle,|-2\rangle\}$, where $|E_\alpha\rangle$, $|E_{\alpha+\pi/2}\rangle$ are two generic elliptically polarized states, which are orthogonal; $|\pm2\rangle$ are dipole-inactive dark states. The allowed channels, which are not explicitly indicated in this table, can be obtained by using Eqs. (10) and (11).

S	S'	S_f	S'_f	$\mathcal{S}^e_{\rm exch}(S,S',S_f,S'_f)$	$\mathcal{S}^h_{\rm exch}(S,S',S_f,S'_f)$
E_α	E_α	E_α	E_α	$\sin^4\alpha+\cos^4\alpha$	$\sin^4\alpha+\cos^4\alpha$
E_α	E_α	$E_{\alpha+\pi/2}$	$E_{\alpha+\pi/2}$	$\frac{1}{2}\sin^2 2\alpha$	$\frac{1}{2}\sin^2 2\alpha$
E_α	E_α	E_α	$E_{\alpha+\pi/2}$	$-\frac{1}{4}\sin 4\alpha$	$-\frac{1}{4}\sin 4\alpha$
E_α	E_α	$+2$	-2	$\frac{1}{2}\sin 2\alpha$	$\frac{1}{2}\sin 2\alpha$
E_α	$E_{\alpha+\pi/2}$	E_α	$E_{\alpha+\pi/2}$	$\frac{1}{2}\sin^2 2\alpha$	$\frac{1}{2}\sin^2 2\alpha$
E_α	$E_{\alpha+\pi/2}$	$E_{\alpha+\pi/2}$	$E_{\alpha+\pi/2}$	$\frac{1}{4}\sin 4\alpha$	$\frac{1}{4}\sin 4\alpha$
E_α	$E_{\alpha+\pi/2}$	$+2$	-2	$-\sin^2\alpha$	$\cos^2\alpha$
E_α	$+2$	E_α	$+2$	$\cos^2\alpha$	$\sin^2\alpha$
E_α	$+2$	$E_{\alpha+\pi/2}$	$+2$	$-\frac{1}{2}\sin 2\alpha$	$\frac{1}{2}\sin 2\alpha$
E_α	-2	E_α	-2	$\sin^2\alpha$	$\cos^2\alpha$
E_α	-2	$E_{\alpha+\pi/2}$	-2	$\frac{1}{2}\sin 2\alpha$	$-\frac{1}{2}\sin 2\alpha$
±2	±2	±2	±2	1	1

not change the result of the integration. In fact, the terms that are momentum independent (i.e., the relative motion wave functions and the Coulomb potentials) are even under inversion of all spatial coordinates.

We remark that the expression for the fermion-fermion exchange terms are the same, which have been obtained in Refs. 5 and 16. In this paper, we study in detail the dependence of such terms as a function of the exchanged momentum. Furthermore, we compare them to the direct term and calculate the collisional broadening. As shown in Appendix A, we have

$$H_{\rm dir}(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})=H_{\rm dir}(q). \tag{17}$$

Furthermore, owing to symmetry $H_{\rm dir}|_{q=0}=0$. Moreover, when $m_e=m_h$, $H_{\rm dir}(q)=0$ for every value of q. The fact that the direct term vanishes in the limit $q\to0$ is of crucial importance, because excitons created by optical excitation have small wave vectors. For the fermion-fermion exchange term, there are rather different properties. We have

$$H^e_{\rm exch}(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})=H^e_{\rm exch}(\Delta Q,q,\theta), \tag{18}$$

where $\Delta Q=|\boldsymbol{Q}'-\boldsymbol{Q}|$ and θ is the angle between $\boldsymbol{Q}'-\boldsymbol{Q}$ and $\boldsymbol{q}$. As explained in Appendix A, $H^e_{\rm exch}$ does not vanish neither for $q=0$ nor for $m_e=m_h$. The same properties are of course valid for $H^h_{\rm exch}$.

B. Calculation of matrix elements

The calculation of the direct contribution $H_{\rm dir}(q)$ can be performed analytically, by changing from variables $\boldsymbol{r_e},\boldsymbol{r_h}$ to the center of mass and relative coordinates $\boldsymbol{R}=\beta_e\boldsymbol{r_e}+\beta_h\boldsymbol{r_h}$, $\boldsymbol{\rho}=\boldsymbol{r_e}-\boldsymbol{r_h}$. In this way, the multiple integrals in Eq. (12) can be factorized and the problem is reduced to the calculation of simple two-dimensional Fourier transforms. The final result is

$$H_{\rm dir}(q)=\frac{1}{A}\frac{e^2}{\epsilon_0}\lambda_{2D}\left(\frac{2}{\pi}\right)^2 I_{\rm dir}(q), \tag{19}$$

where

$$I_{\rm dir}(q)=\frac{\pi^3}{2q\lambda_{2D}}\left\{\left[1+\left(\frac{1}{2}\beta_e q\lambda_{2D}\right)^2\right]^{-3/2}-\left[1+\left(\frac{1}{2}\beta_h q\lambda_{2D}\right)^2\right]^{-3/2}\right\}^2. \tag{20}$$

Notice that $H_{\rm dir}$ actually vanishes when $q=0$ or when $m_e=m_h$, as previously discussed. The integral $I_{\rm dir}$ depends critically on the mass ratio m_e/m_h, as depicted in Fig. 1. For the exciton-exciton exchange contribution, we have

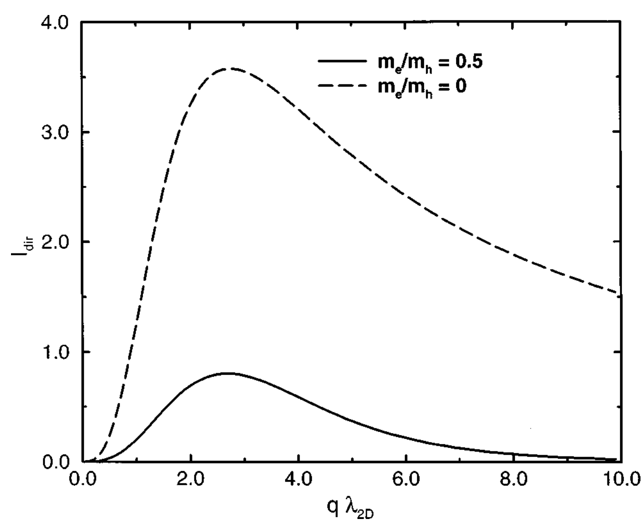

FIG. 1. The dimensionless direct integral $I_{\rm dir}$ as a function of the transferred wave vector q. Solid line: $m_e/m_h=0.5$. Dashed line: $m_e/m_h=0$. In the case $m_e/m_h=1$, $I_{\rm dir}\equiv0$.

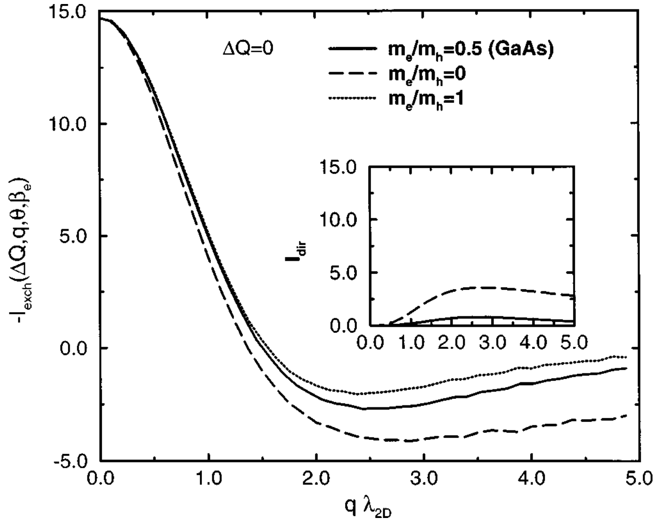

FIG. 2. The dimensionless electron-electron exchange integral $-I_{\rm exch}(\Delta Q=0,q,\theta,\beta_e)$ as a function of q for three different values of m_e/m_h. Solid line: $m_e/m_h=0.5$, corresponding to a GaAs quantum well. Long-dashed line: $m_e/m_h=0$. Dotted line: $m_e/m_h=1$. Inset: the dimensionless direct integral $I_{\rm dir}$ is shown for comparison.

$$H^X_{\rm exch}(\Delta Q,q,\theta)=\frac{1}{A}\frac{e^2}{\epsilon_0}\lambda_{\rm 2D}\left(\frac{2}{\pi}\right)^2 \times I_{\rm dir}[\sqrt{(\Delta Q)^2+q^2-2\Delta Q q\cos\theta}]. \quad (21)$$

For the fermion-fermion exchange terms $H^e_{\rm exch}$ and $H^h_{\rm exch}$ the involved multiple integrals can not be computed analytically. They can be rewritten in the form

$$H^e_{\rm exch}(\Delta Q,q,\theta)=-\frac{1}{A}\frac{e^2}{\epsilon_0}\lambda_{\rm 2D}\left(\frac{2}{\pi}\right)^2 I_{\rm exch}(\Delta Q,q,\theta,\beta_e), \quad (22)$$

$$H^h_{\rm exch}(\Delta Q,q,\theta)=-\frac{1}{A}\frac{e^2}{\epsilon_0}\lambda_{\rm 2D}\left(\frac{2}{\pi}\right)^2 I_{\rm exch}(\Delta Q,q,\theta,\beta_h). \quad (23)$$

The quantity $I_{\rm exch}$ is a real number, whose explicit expression is given in Appendix B. We have computed $I_{\rm exch}$ by a standard Monte Carlo integration. In Fig. 2, we show a plot of $-I_{\rm exch}(\Delta Q,q,\theta,\beta_e)$ in the case $\Delta Q=0$. As it can be seen in the expression given in Appendix B, for $\Delta Q=0$, the hole-hole term is equal to the electron-electron one. Furthermore, in this case there is no dependence on the angle θ. The modulus of the fermion-fermion exchange integral has its maximum value at $q=0$, while the direct term vanishes. The dependence on the mass ratio m_e/m_h for $I_{\rm exch}$ is not as critical as for $I_{\rm dir}$. In fact, in the region of small q, which is the most relevant for optical excitation, the matrix elements are weakly influenced by m_e/m_h. The fermion-fermion exchange contributions are largely dominant with respect to the direct interaction.

III. COLLISIONAL BROADENING

As far as optical experiments are concerned, the exciton-exciton scattering produces the so-called collisional broadening, that is a density-dependent homogeneous line width. Starting from a nonequilibrium formalism within the Born approximation,[10] it is possible to show that the line width of an exciton with wave vector $\boldsymbol{Q}$ and in the spin state $|S\rangle$ obeys the implicit equation

$$\Gamma^S_{\boldsymbol{Q}}=2\pi\sum_{\boldsymbol{Q}'}\sum_{S',S_f,S'_f}N_{S'}(\boldsymbol{Q}')\sum_{\boldsymbol{q}\neq\boldsymbol{0}}|H^{S_fS'_f}_{SS'}(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})|^2 \times\mathcal{L}(E_{\boldsymbol{Q}}+E_{\boldsymbol{Q}'}-E_{\boldsymbol{Q}+\boldsymbol{q}}-E_{\boldsymbol{Q}'-\boldsymbol{q}},\Gamma^S_{\boldsymbol{Q}}+\Gamma^{S'}_{\boldsymbol{Q}'} +\Gamma^{S_f}_{\boldsymbol{Q}+\boldsymbol{q}}+\Gamma^{S'_f}_{\boldsymbol{Q}'-\boldsymbol{q}}), \quad (24)$$

where $E_{\boldsymbol{Q}}=\hbar^2Q^2/2M$ is the exciton kinetic energy and $N_{S'}(\boldsymbol{Q}')$ is the number of excitons in the state $(1s,\boldsymbol{Q}',S')$, with $S'\in\{E_\alpha,E_{\alpha+\pi/2},+2,-2\}$. The Lorentzian distribution $\mathcal{L}$ is defined as

$$\mathcal{L}(E,\gamma)=\frac{1}{\pi}\frac{\gamma/2}{E^2+(\gamma/2)^2}. \quad (25)$$

The implicit expression for the line width in Eq. (24) reduces to the usual Born approximation[17] in the limit of very small damping. In fact, for $\gamma\to0$, $\mathcal{L}(E,\gamma)\to\delta(E)$. The physical meaning of Eq. (24) is that, in presence of Coulomb interaction, excitons are damped quasi-particles and therefore the energy conservation in the scattering processes is softened.[18] In a typical experiment under pulsed excitation, the value of the collisional broadening actually depends on the distribution of excitons both in momentum and spin space [i.e., $N_S(Q)$]. This distribution can be obtained only by considering the dynamics of the exciton gas, which is not the purpose of this paper. At this point we remark that, because of the softened energy conservation, a single exciton-exciton scattering event may take place between initial and final states whose energy separation is of the order of the collisional broadening. Taking into account the exciton dispersion and assuming a collisional broadening of the order of 1 meV (as we will actually find), then the corresponding in-plane momentum space spanned by the scattering events is such that $N_S(Q)$ is significant only well within $\lambda^{-1}_{\rm 2D}$. In this range the value of the fermion-fermion exchange matrix elements is approximately constant. We thus make the approximation of assuming an exciton population in the states with $Q=0$ only. We consider the two particular cases of excitation by linearly and by circularly polarized light. Let us consider the case of exactly resonant excitation by a linearly polarized laser pulse, which propagates along the growth direction. Initially, excitons are all in the same linear spin state. As we can see from Table I in the case $\alpha=\pi/4$, two interacting excitons in the same linear polarization can scatter to the orthogonal linear polarization and also to the dark states. Considering all the allowed scattering channels and their relative weight, we realize that in the steady-state case, collisions lead to an equal population of these four spin states (i.e., $|x\rangle$, $|y\rangle$, $|+2\rangle$, $|-2\rangle$). This equal redistribution takes place on a time scale given by the inverse of the broadening. Therefore, we assume that $N_x(Q)=N_y(Q)=N_{+2}(Q)=N_{-2}(Q)=\frac{1}{4}N\delta_{Q,0}$, where N is the total number of excitons. For simplicity, we neglect any Q dependence of Γ, consistently with previous arguments. Eq. (24) becomes

$$\Gamma=\left(\frac{2}{\pi}\right)^4\left(\frac{\hbar^2}{2\mu}\right)^2\sum_{S'}\ \frac{1}{4}n\int_0^\infty dq\,qF_{xS'}(q)\times\frac{1}{\pi}\frac{2\Gamma}{\left(\frac{\hbar^2q^2}{M}\right)^2+(2\Gamma)^2},\qquad(26)$$

where $n=N/A$ is the total density of excitons. The function F is defined as

$$F_{SS'}(q)=\sum_{S_f,S_f'}|I_{SS'}^{S_fS_f'}(\Delta Q=0,q,\theta)|^2,\qquad(27)$$

where

$$\begin{aligned}I_{SS'}^{S_fS_f'}(\Delta Q,q,\theta)=&\langle S|S_f\rangle\langle S'|S_f'\rangle I_{\rm dir}(q)\\&+\langle S|S_f'\rangle\langle S'|S_f\rangle\\&\times I_{\rm dir}[\sqrt{(\Delta Q)^2+q^2-2\Delta Qq\cos\theta}]\\&-\mathcal{S}^e_{\rm exch}(S,S',S_f,S_f')I^e_{\rm exch}(\Delta Q,q,\theta,\beta_e)\\&-\mathcal{S}^h_{\rm exch}(S,S',S_f,S_f')I^h_{\rm exch}(\Delta Q,q,\theta,\beta_h).\end{aligned}\qquad(28)$$

In the case of excitation by a circularly polarized pulse, the situation is different. As we can deduce from Table I in the case $\alpha=0$, collisions between excitons in the same circular polarization do not lead to spin relaxation, because only the channel $(\pm1,\pm1)\rightarrow(\pm1,\pm1)$ is allowed. Thus, excitons remain all in the same circularly polarized states as long as other spin-relaxation mechanisms are negligible. Therefore, if we take $N_{+1}(Q)=N\delta_{Q,0}$, we finally obtain

$$\Gamma=\left(\frac{2}{\pi}\right)^4\left(\frac{\hbar^2}{2\mu}\right)^2 n\int_0^\infty dq\,qF_{11}(q)\ \frac{1}{\pi}\frac{2\Gamma}{\left(\frac{\hbar^2q^2}{M}\right)^2+(2\Gamma)^2}.\qquad(29)$$

In Fig. 3, we show a plot of the dimensionless function $F_{SS'}(q)$, defined in Eq. (27). As shown in Table I, in the case of scattering of two excitons in the same linearly polarized state (namely x), the four allowed channels $(x,x)\rightarrow\{(x,x);(y,y);(2,-2);(-2,2)\}$ have the spin factors such that $\mathcal{S}^e_{\rm exch}=\mathcal{S}^h_{\rm exch}$. In particular, $|\mathcal{S}^e_{\rm exch}|^2=|\mathcal{S}^h_{\rm exch}|^2=\frac{1}{4}$. On the other hand, when the two interacting excitons are in the same circular spin state (namely $+1$) only the channel $(+1,+1)\rightarrow(+1,+1)$ is allowed, but with spin factors $\mathcal{S}^e_{\rm exch}=\mathcal{S}^h_{\rm exch}=1$. Thus, as far as fermion-fermion exchange is concerned, F_{xx} and F_{11} are equal. As depicted in Fig. 3, F_{xx} and F_{11} coincide except in the region of large q where $I_{\rm dir}$ contributes. In this region the curves do not coincide because of a different interference between the fermion-fermion exchange terms and $I_{\rm dir}$. In fact, with respect to F_{xx}, $I_{\rm dir}$ appears only in the channel $(x,x)\rightarrow(x,x)$, where the spin-exchange factors are $\mathcal{S}^e_{\rm exch}=\mathcal{S}^h_{\rm exch}=\frac{1}{2}$. On the other hand, in the channel $(+1,+1)\rightarrow(+1,+1)$ these spin-exchange factors are both equal to 1. Concerning F_{xy}, we have the four channels $(x,y)\rightarrow\{(x,y);(y,x);(2,-2);(-2,2)\}$. In the first two channels $\mathcal{S}^e_{\rm exch}=\mathcal{S}^h_{\rm exch}=\frac{1}{2}$, while in the last two $\mathcal{S}^e_{\rm exch}=-\mathcal{S}^h_{\rm exch}$. Thus, in these two channels there is destructive interference between the electron-electron and the hole-hole contribution. Therefore, as far as fermion-fermion exchange is concerned, F_{xy} is one half of F_{xx}. The same result holds for $F_{x\pm2}$. In Fig. 3, we actually see that, because fermion-fermion exchange contributions dominate, $F_{xy}\approx\frac{1}{2}F_{xx}$.

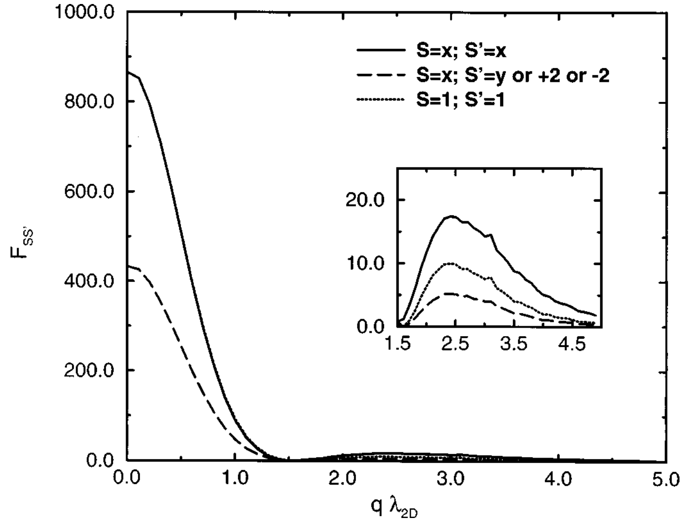

FIG. 3. The dimensionless function $F_{SS'}$ as a function of q for $m_e/m_h=0.5$ (GaAs). Solid line: S and S' are the same linearly polarized spin state. Dashed line: S is a linearly polarized state, while S' is orthogonal. Dotted line: S and S' are the same circular spin state. Inset: the region of large q is enlarged.

We have solved numerically Eqs. (26) and (29) by a standard iteration technique. In Fig. 4, the results for the self-consistent density-dependent homogeneous line width are shown for the two above mentioned situations. The self consistency lead to a sublinear behavior of the line width as a function of the total density of excitations. In the case n_x

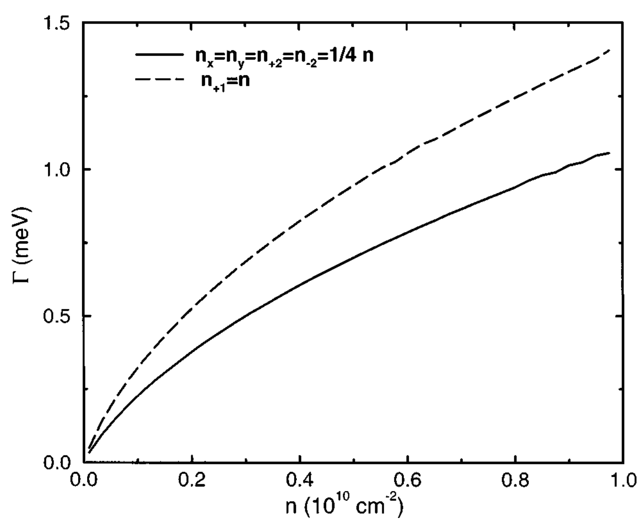

FIG. 4. The self-consistent collisional broadening of an exciton with $Q=0$ as a function of the total exciton density (per unit area) for two particular situations. Solid line: the $|x\rangle$, $|y\rangle$, $|+2\rangle$, $|-2\rangle$ states are equally populated. Dashed line: only a circular polarization is populated.

$=n_y=n_{\pm 2}=\frac{1}{4}n$, the collisional broadening is smaller than in the case $n_1=n$, because $F_{xx}\approx F_{11}$, while $F_{xS'}\approx\frac{1}{2}F_{11}$. To the authors' knowledge, the difference in the value of the collisional broadening in the two above mentioned situations has never been specifically investigated in experiments.

IV. DISCUSSION AND CONCLUSIONS

In this paper, we have presented a theoretical model of the elastic exciton-exciton scattering in a semiconductor quantum well. Within a two-band envelope function formalism, we have calculated the scattering matrix elements, including also the exchange terms and the spin degrees of freedom. Our results show that the inter-exciton exchange of carriers is the dominant interaction mechanism. In fact, the direct term and the exciton-exciton exchange are found to be negligible compared to the electron-electron and hole-hole exchange. Within the Born approximation, we have calculated the density-dependent homogeneous broadening.

Comparing our results with the existing experimental data, we find satisfactory agreement. In a photoluminescence experiment under resonant excitation, Deveaud *et al.*[3] measured a collisional broadening of nearly 1 meV at the estimated density of 10^{10} cm^{-2}. The experiment was performed on a 45 Å GaAs quantum well, by employing linearly polarized light. As we can see in Fig. 4, this is in good agreement with our results. On the other hand, the four-wave-mixing experiment in pump and probe configuration by Honold *et al.*[2] gave smaller values for the density-dependent homogeneous broadening ($0.1\div 0.2$ meV at an estimated density of 10^{10} cm^{-2}). The measurement was still performed with linearly polarized light, but on a much thicker GaAs quantum well (120 Å), where the two-dimensional character is weaker. Our calculation is for a purely two-dimensional quantum-well system and therefore form factors for realistic quasi-2D structures are not taken into account. We remind that for thicker wells the efficiency of Coulomb scattering is expected to be weaker. Furthermore, in the experiment by Honold *et al.*, the probe beam arrived on the sample with a delay of 20 ps after the pump, in order to avoid coherent effects. Theoretical calculations[19] and more recent experimental studies[3,20] have shown that in high-quality quantum wells, excitons have a radiative lifetime of the order of 10 $\div$ 20 ps. This should lead in Ref. 2 to an overestimate of the actual density of excitons when the sample is probed and consequently to an underestimate of the broadening.

Our model takes into account also the spin degrees of freedom. Owing to fermion-fermion exchange, collisions can lead to spin relaxation. We have derived the selection rules for the allowed scattering spin channels. As already observed in the introduction, the problem of the spin relaxation of an interacting exciton gas has been very recently investigated by Amand *et al.*[7] They have performed time-resolved photoluminescence experiments, under resonant excitation by elliptically polarized light. Their measurements show that a density-dependent spin relaxation takes place. In particular, they observe a fast decay of the luminescence followed by a much slower one. The fast component disappears in the low-excitation regime and in the limiting case of circular polarization. According to Amand *et al.*, the fast decay is a consequence of the transfer from the optically active states to the dipole-forbidden dark ones. They attribute to the inter-exciton exchange of carriers the characteristic features of the observed photoluminescence signal. In order to fit their time-resolved signal, they consider an effective phenomenological spin-spin Hamiltonian. Our model naturally shows how this spin-relaxation mechanism originates from Coulomb interaction, when the antisymmetry under carrier exchange in the two-exciton wave function is correctly included. In particular, two interacting excitons in the same elliptically polarized state can scatter to the dipole-forbidden dark states. On the other hand, two excitons in the same circularly polarized state cannot. This spin relaxation mechanism holds because of the dominant role of the inter-exciton exchange of carriers in the exciton-exciton Coulomb interaction.

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APPENDIX A: SYMMETRY PROPERTIES OF MATRIX ELEMENTS

Here we show some peculiarities of the direct integral $H_{\rm dir}$ and of the fermion-fermion exchange terms $H^e_{\rm exch}$ and $H^h_{\rm exch}$. As far as $H_{\rm dir}$ is concerned, there are significant symmetry properties. First of all, in $H_{\rm dir}$, the product

$$\Psi^\star_{\boldsymbol{Q}}(\boldsymbol{r_e},\boldsymbol{r_h})\Psi^\star_{\boldsymbol{Q'}}(\boldsymbol{r_{e'}},\boldsymbol{r_{h'}})\Psi_{\boldsymbol{Q+q}}(\boldsymbol{r_e},\boldsymbol{r_h})\Psi_{\boldsymbol{Q'-q}}(\boldsymbol{r_{e'}},\boldsymbol{r_{h'}}) \tag{A1}$$

does not depend on $\boldsymbol{Q},\boldsymbol{Q'}$, as it is clearly obtained from Eq. (1). Therefore we have

$$H_{\rm dir}(\boldsymbol{Q},\boldsymbol{Q'},\boldsymbol{q})=H_{\rm dir}(q). \tag{A2}$$

The dependence on $\boldsymbol{q}$ of the product (A1) appears through the factor

$$\exp\{i\boldsymbol{q}\cdot[\beta_e(\boldsymbol{r_e}-\boldsymbol{r_{e'}})+\beta_h(\boldsymbol{r_h}-\boldsymbol{r_{h'}})]\}. \tag{A3}$$

$H_{\rm dir}(q=0)$ does not depend on the carrier masses ($m_e\neq m_h$). In particular, if we perform in Eq. (12) the change of variables $\boldsymbol{r_{e''}}=\boldsymbol{r_{h'}}$, $\boldsymbol{r_{h''}}=\boldsymbol{r_{e'}}$, we find that

$$H_{\rm dir}(q=0)=-H_{\rm dir}(q=0)\Rightarrow H_{\rm dir}(q=0)=0. \tag{A4}$$

Furthermore, if $m_e=m_h$, $H_{\rm dir}(q)=0$ for every value of q.

The fermion-fermion exchange integrals $H^e_{\rm exch}$ and $H^h_{\rm exch}$ have very different properties. If we consider, for instance, $H^e_{\rm exch}$ given in Eq. (16), we have that

$$\begin{aligned}&\Psi^\star_{\boldsymbol{Q}}(\boldsymbol{r_e},\boldsymbol{r_h})\Psi^\star_{\boldsymbol{Q'}}(\boldsymbol{r_{e'}},\boldsymbol{r_{h'}})\Psi_{\boldsymbol{Q+q}}(\boldsymbol{r_{e'}},\boldsymbol{r_h})\Psi_{\boldsymbol{Q'-q}}(\boldsymbol{r_e},\boldsymbol{r_{h'}})\\&\quad\propto\exp\{i(\boldsymbol{Q'}-\boldsymbol{Q})\cdot[\beta_e(\boldsymbol{r_e}-\boldsymbol{r_{e'}})]\}\\&\quad\times\exp\{i\boldsymbol{q}\cdot[\beta_e(\boldsymbol{r_{e'}}-\boldsymbol{r_e})+\beta_h(\boldsymbol{r_h}-\boldsymbol{r_{h'}})]\}.\end{aligned} \tag{A5}$$

This means that

$$H^{e}_{\rm exch}(\boldsymbol{Q},\boldsymbol{Q}',\boldsymbol{q})=H^{e}_{\rm exch}(\Delta Q,q,\theta), \tag{A6}$$

where $\Delta Q=|\boldsymbol{Q}'-\boldsymbol{Q}|$ and θ is the angle between $\boldsymbol{Q}'-\boldsymbol{Q}$ and $\boldsymbol{q}$. The exchange of electron coordinates in the final state breaks the symmetry between electron and hole, which is instead present in $H_{\rm dir}$ in the case $q=0$ and, more generally, in the case $m_e=m_h$. Thus, we have $H^{e}_{\rm exch}(\Delta Q=0,q=0,\theta)\neq 0$ and $H^{e}_{\rm exch}(\Delta Q,q,\theta)\neq 0$ also in the case $m_e=m_h$. The same properties are of course valid for $H^{h}_{\rm exch}$.

APPENDIX B: FERMION-FERMION EXCHANGE INTEGRAL

In this appendix, we give the simplified expression for $H^{e}_{\rm exch}$ [see Eq. (22)] after shortly explaining how it is obtained starting from Eq. (16). Let us define $\boldsymbol{R}=\beta_e\boldsymbol{r_e}+\beta_h\boldsymbol{r_h}$ and $\boldsymbol{\rho}=\boldsymbol{r_e}-\boldsymbol{r_h}$. Changing the variables $\boldsymbol{r_e}$, $\boldsymbol{r_{e'}}$, $\boldsymbol{r_h}$, $\boldsymbol{r_{h'}}$ to $\boldsymbol{\xi}=\boldsymbol{R}-\boldsymbol{R}'$, $\boldsymbol{\sigma}=\{\boldsymbol{R}+\boldsymbol{R}'\}/2$, $\boldsymbol{\rho}$, $\boldsymbol{\rho}'$, it is possible to reduce the dimension of the integration from eight to six; in fact the integrand does not depend on $\boldsymbol{\sigma}$ and therefore the corresponding integration is trivial, giving the factor A, that is the normalization area. If we again change the variables $\boldsymbol{\xi}$, $\boldsymbol{\rho}$, $\boldsymbol{\rho}'$ into the new ones $\boldsymbol{y_1}=\{\boldsymbol{\xi}-\beta_e\boldsymbol{\rho}-\beta_h\boldsymbol{\rho}'\}/\lambda_{2\rm D}$, $\boldsymbol{y_2}=\{\boldsymbol{\xi}+\beta_h\boldsymbol{\rho}+\beta_e\boldsymbol{\rho}'\}/\lambda_{2\rm D}$, $\boldsymbol{x}=\boldsymbol{\rho}/\lambda_{2\rm D}$, we finally obtain Eq. (22), where

$$\begin{aligned}I_{\rm exch}(\Delta Q,q,\theta,\beta_e)=&\int_0^\infty dx\int_0^{2\pi}d\theta_x\int_0^\infty dy_1\int_0^{2\pi}d\theta_1\int_0^\infty dy_2\int_0^{2\pi}d\theta_2\, x\, y_1 y_2 \cos\{\Delta Q\lambda_{2\rm D}[\beta_e x\cos(\theta-\theta_x)\\&+\beta_e y_1\cos(\theta-\theta_1)]+q\lambda_{2\rm D}[-x\cos\theta_x-\beta_e y_1\cos\theta_1+(1-\beta_e)y_2\cos\theta_2]\}\\&\times\exp(-[(y_2\cos\theta_2-y_1\cos\theta_1-x\cos\theta_x)^2+(y_2\sin\theta_2-y_1\sin\theta_1-x\sin\theta_x)^2]^{1/2})\exp(-x)\\&\times\exp(-y_1)\exp(-y_2)\left\{\frac{1}{\sqrt{y_1^2+x^2+2y_1x\cos(\theta_1-\theta_x)}}+\frac{1}{\sqrt{y_2^2+x^2-2y_2x\cos(\theta_2-\theta_x)}}-\frac{1}{y_1}-\frac{1}{y_2}\right\}.\end{aligned} \tag{B1}$$

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