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Effects of the Heavy- and Light-Hole Mixing in the Exciton Dynamics in Semiconductor Quantum Wells

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We present the calculation of the low temperature exciton bimolecular formation rate in quantum wells through acoustic phonon scattering taking into account the heavy- and light-hole mixing in the hole and exciton states. We observe a decrease in the exciton formation rate compared to the parabolic approximation. For symmetric quantum wells, the hole-subband mixing allows a finite probability of scattering involving the change of parity. This opens a channel for spin-flip transitions during the exciton formation process.

Introduction In nonresonant photoluminescence experiments, optically created electron–hole pairs rapidly reach a quasi-thermal equilibrium distribution near the subband edges. They then decay into an exciton state. At low carrier densities this is dominated by phonon-assisted scattering and it is then a bimolecular exciton formation process. The excitons in the ground state are formed with a finite center-of-mass (CM) wave vector and have to relax to optically active states before emission takes place. The study of the exciton dynamics is particularly important in semiconductor quantum wells (QWs) where the exciton states dominate the optical emission.

Time-resolved optical spectroscopy has provided important information on the exciton formation process in semiconductor QWs. Kusano et al. [1] and Damen et al. [2] obtained the first results on the exciton formation time. More recently, Kumar et al. [3] measured a bimolecular exciton formation rate C equal to 0.5 cm^2/s for a 80 Å GaAs QW.

From the theoretical viewpoint, the exciton formation process has been studied following different approaches. Gulia et al. [4] solved a set of kinetic equations using an ensemble Monte Carlo technique. They obtained a time dependent exciton formation rate. Piermarocchi et al. [5] calculated the exciton dynamics in terms of rate equations for the carrier populations. In this description, the exciton formation rate is time independent. More recently, Kira et al. [6] studied the excitonic luminescence in the regime of very short time using the quantum theory of the interacting photon electron–hole system.

These works have considered a simple parabolic description for the hole and exciton states in the QW. The actual hole dispersion shows a complex pattern once the heavy- and light-hole coupling is taken into account [7]. Triques and Brum [8] showed that the exciton CM dispersion is also strongly non-parabolic as a consequence of this coupling. When we consider the complexity of the hole-subband mixing for GaAs QWs the spin is not a good quantum number. For symmetric QWs, the hole and the exciton Hamilto-

nians may be separated by a unitary transformation into two uncoupled blocks, which have degenerate eigenstates labeled by the quantum parity number Σ. The heavy- and light-hole mixing allows the formation of excitons with holes showing a different parity than that in the initial state (*parity-flip process*).

To account for these effects one has to consider the Luttinger Hamiltonian to describe the hole-subband states as well as the exciton states. The exciton–phonon interaction is then described by the Bir-Pikus Hamiltonian [9]. This considerably complicates the exciton dynamics which involves the participation of the excited exciton states. In this paper, we focus on the effects of heavy- and light-hole mixing in the formation of excitons in their ground state through longitudinal-acoustic (LA) phonon scattering.

Model We follow here the approach based on the rate equations for the carrier populations developed by Piermarocchi et al. [5]. In our case, the exciton formation process is characterized by two bimolecular formation rates, one for the parity-conserving process, $C_{\uparrow\uparrow}$, and one for the parity-flip process, $C_{\uparrow\downarrow}$, where $\uparrow$ and $\downarrow$ stand for the exciton and hole parities, Σ_{exc} and Σ_{h}. The rate equation for the electron density is

$$\frac{\mathrm{d}n_{\mathrm{e}}}{\mathrm{d}t} = -C_{\uparrow\uparrow} n_{\mathrm{e}} n_{\mathrm{h}} - C_{\uparrow\downarrow} n_{\mathrm{e}} n_{\mathrm{h}} \,, \tag{1}$$

where the exciton formation rates are given by

$$C_{\uparrow\uparrow(\downarrow)} = \int \mathrm{d}E_{\mathrm{f}}\, F_{\uparrow\uparrow(\downarrow)}(E_{\mathrm{f}})\, g(E_{\mathrm{f}}) \tag{2}$$

and

$$F_{\uparrow\uparrow(\downarrow)}(E_{\mathrm{f}}) = \frac{4}{n_{\mathrm{e}} n_{\mathrm{h}} S} \frac{2\pi}{\hbar} \sum_{\nu=+,-} \sum_{\mathbf{k}_{\mathrm{e}}, \mathbf{k}_{\mathrm{h}}, \mathbf{q}} |\langle n(\mathbf{q}) + \nu 1, \boldsymbol{\Psi}_{\mathrm{f}}^{\uparrow} \mid \mathbf{H}_{\mathrm{exc-ph}}^{\nu} \mid \boldsymbol{\Psi}_{\mathrm{i}}^{\uparrow(\downarrow)}, n(\mathbf{q}) \rangle|^2 \times f_{\mathrm{e}}(k_{\mathrm{e}})\, f_{\mathrm{h}}(k_{\mathrm{h}})\, \delta(E_{\mathrm{f}} - E_{\mathrm{i}} + \nu E_{\mathrm{ph}}) \,, \tag{3}$$

$$n_{\mathrm{e}} = \frac{1}{S} \sum_{\mathbf{k}_{\mathrm{e}}, \sigma_{\mathrm{e}}} f_{\mathrm{e}}(k_{\mathrm{e}}) \,, \qquad n_{\mathrm{h}} = \frac{1}{S} \sum_{\mathbf{k}_{\mathrm{h}}, \Sigma_{\mathrm{h}}} f_{\mathrm{h}}(k_{\mathrm{h}}) \,, \tag{4}$$

$\mathbf{k}_{\mathrm{e}}$ and $\mathbf{k}_{\mathrm{h}}$ are the electron and hole wave vectors, respectively, σ_{e} is the electron spin and S is the sample area. $f_{\mathrm{e}}(k_{\mathrm{e}})$ and $f_{\mathrm{h}}(k_{\mathrm{h}})$ are, respectively, the electron and hole distributions in the quasi-thermal-equilibrium situation characterized by a common carrier temperature T_{c}. $\mathbf{H}_{\mathrm{exc-ph}}^{\nu}$ is the exciton–phonon interaction Hamiltonian, where $\nu = +(-)$ refers to the emission (absorption) process. $\boldsymbol{\Psi}_{\mathrm{i}}^{\Sigma_{\mathrm{h}}}$ and E_{i} are the wave function and the energy of the initial state, respectively, which is formed by an electron–hole pair, where the Coulomb interaction is neglected. $\boldsymbol{\Psi}_{\mathrm{f}}^{\Sigma_{\mathrm{exc}}}$ and E_{f} are the wave function and the energy of the final state, respectively. $g(E_{\mathrm{f}})$ is the density of states of the exciton ground state. $n(\mathbf{q})$ is the Bose-Einstein distribution of phonons with wave vector $\mathbf{q}$ and energy E_{ph} at the lattice temperature T_{L}.

The hole and the exciton CM states are calculated in the frame of the 4×4 Luttinger Hamiltonian [7, 8]. The LA-phonons are described by their bulk dispersion in the Debye approximation. The hole part of the exciton–LA-phonon interaction Hamiltonian is given by the Bir-Pikus Hamiltonian [9]. We are interested here in the effects of the hole-subband complexity in the electronic states and we therefore simplify the Bir-Pikus Hamiltonian by considering only its diagonal part. We assume that the initial electron and hole densities are low and $f_{\mathrm{e}}(k_{\mathrm{e}})$ and $f_{\mathrm{h}}(k_{\mathrm{h}})$ are Boltzmann distributions. The multiple integrals in Eqs. (2) and (3) are solved using the Monte Carlo method.

The parity-conserving channel involves both the electron and the hole parts of $\mathbf{H}^{\nu}_{\text{exc-ph}}$ while only the hole part of $\mathbf{H}^{\nu}_{\text{exc-ph}}$ is present in the parity-flip channel. In the parabolic approximation the spin is conserved and $C_{\uparrow\downarrow}$ is zero.

The modifications introduced in the exciton formation rate by taking into account the heavy- and light-hole mixing are twofold: (i) The hole and the exciton CM in-plane dispersions are non-parabolic. Their average masses are heavier than in the parabolic approximation and strongly dependent on the QW width. (ii) There is another channel for exciton formation in which the parity is not conserved.

Results and Discussions We consider a GaAs–$Ga_{0.7}Al_{0.3}As$ QW at the lattice temperature $T_L = 10$ K and carrier quasi-temperature $T_c = 30$ K. Figures 1a and b show F as a function of the exciton CM kinetic energy, $E_k = E_f - E_f|_{k_{CM}=0}$, in the parabolic (dashed lines) and non-parabolic (full lines) dispersions for QW widths of 100 and 150 Å, respectively. The contributions for the two channels, $F_{\uparrow\uparrow}$ (dash-dotted lines) and $F_{\uparrow\downarrow}$ (dotted lines), in the non-parabolic dispersion are also shown. At the lattice temperature we considered, the LA-phonon emission largely dominates over the LA-phonon absorption (not shown here). The results show that most of the excitons are formed with an excess of kinetic energy of the order of the exciton ground state bind-

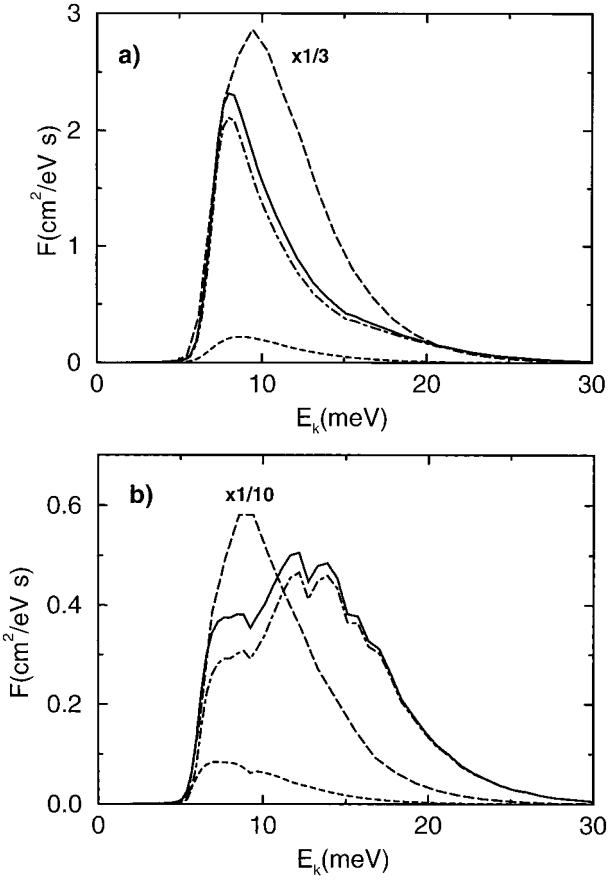

Fig. 1. F as a function of E_k for a GaAs/$Ga_{0.3}Al_{0.7}As$ QW at $T_c = 30$ K and $T_L = 10$ K in the non-parabolic case (solid lines) and the parabolic approximation (dashed lines) for a) $L = 100$ Å and b) $L = 150$ Å. The parity-conserving, $F_{\uparrow\uparrow}$ (dash-dotted lines), and the parity-flip, $F_{\uparrow\downarrow}$ (dotted lines), contributions are also shown

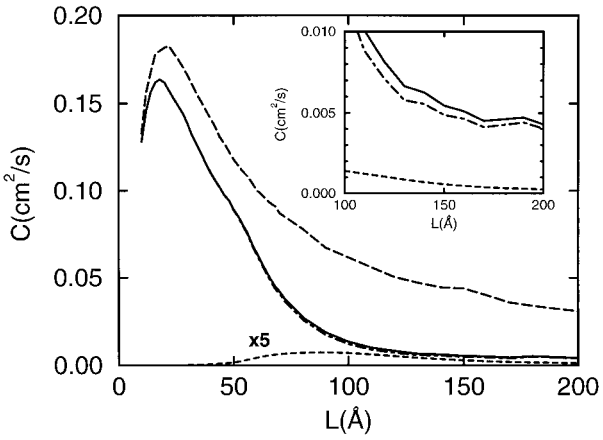

Fig. 2. C as a function of the QW width L for the same parameters as in Fig. 1 in the non-parabolic case (solid line) and the parabolic approximation (dashed line). $C_{\uparrow\uparrow}$ (dash-dotted line) and $C_{\uparrow\downarrow}$ (dotted line) contributions are also shown

ing energy. This is a consequence of the low energy phonons that effectively participate in the process. The high energy tail of F is due to the finite carrier temperature. For the QW width $L = 150$ Å the formation of excitons at higher energy than the exciton binding energy is particularly favored. For this value of L, the holes thermally occupy the anticrossing region of the dispersion which has a higher density of states, favoring the formation of more energetic excitons.

Figure 2 shows the total bimolecular exciton formation rate, $C = C_{\uparrow\uparrow} + C_{\uparrow\downarrow}$, as a function of QW width in the non-parabolic case (full line) and in the parabolic approximation (dashed line) for the same parameters as in Fig. 1. The two contributions, $C_{\uparrow\uparrow}$ (dash-dotted line) and $C_{\uparrow\downarrow}$ (dotted line), are also shown. The inset gives the details of the non-parabolic case at wide QWs. The exciton formation rate increases with the degree of confinement of the carrier wave function in the QW, reaching its maximum for $L \approx 20$ to 30 Å in both cases. As a consequence of the hole–subband coupling, C decreases to lower values as compared to the parabolic approximation. This effect is present already at narrow QWs becoming important as the QW width increases. Most of the hole states which are thermally occupied present a predominantly heavy-hole character. The excitons, however, are formed with a finite CM wave vector and always present a mixed character. The result is a decrease in the scattering matrix element which becomes more significant as the coupling increases.

The contribution of the parity-flip channel, $C_{\uparrow\downarrow}$, is weak. It increases with the coupling at large QWs. Since only the hole flips its spin, this channel plays a role for the formation of dark excitons.

In conclusion, we calculated the bimolecular exciton formation rate assisted by LA-phonon scattering in GaAs QWs including the hole–subband mixing and exciton CM dispersions. The main results are the reduction of the formation rate and the opening of a parity-flip channel for the exciton formation. Our results suggest that these effects have to be considered for a complete understanding of the exciton dynamics.

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