Exciton spin dynamics in quantum wells

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A theory of exciton spin dynamics is given in terms of the exchange spin-flip mechanism taking full account of the confinement of the quantum well. Exciton spin relaxation belongs to the motional narrowing class, with a characteristic inverse proportionality to the momentum scattering time of the exciton center of mass. Analysis of the time dependence of optical intensities of both circular polarizations, including competing relaxation mechanisms from exciton exchange and from single-particle spin flip into optically inactive states, leads to characteristic shapes reflecting their relative importance. The calculated well-width dependence from our theory of the exciton spin relaxation leads to polarization intensities in good agreement with measurement. Theoretical electric and magnetic field dependences have yet to be tested against experiment.

I. INTRODUCTION

Investigations of optical transitions in quantum wells have given us a good understanding of the exciton formation processes in these semiconductor heterostructures. Recent advances in ultrafast laser spectroscopy[1–5] have deduced from the time evolution of luminescence how excitons form and how their energy and momentum relax. Though this still is a subject of intense studies, the current picture[3] is that after the creation of a highly energetic electron-hole pair in an undoped quantum well by optical excitation, the relaxation process basically occurs in two steps. First, it takes a short period of time (of the order of tens of picoseconds) for this electron-hole pair to relax and form an exciton with a high center-of-mass momentum ($\mathbf{K}$). Then, a slow relaxation within the exciton branch (lasting hundreds of picoseconds) brings the exciton to rest (i.e., to the $\mathbf{K} \approx \mathbf{0}$ state), where it is possible for the exciton to recombine emitting a photon.

When luminescence or pump-and-probe experiments are carried out with polarized light, further insights into the relaxation processes may be obtained. One can investigate the spin-relaxation processes accompanying the energy-momentum relaxation.[6] For doped quantum wells it has been established[7] that the minority carrier spin population at the band edge determines the luminescence polarization when the exciting light intensity is weak. In undoped wells, the photoexcited populations of electrons and holes have the same density, and the observed luminescence polarization is influenced by relaxation processes occurring on both populations. There have been several theoretical explanations[8–11] of the experimental data for intrinsic samples. Emphasis is placed on the independent spin relaxation of the two distinct carrier populations although the spin relaxation of the exciton through exchange has been mentioned.[8,10] The observed decay in time of the luminescence polarization after *resonant* excitation of heavy-hole excitons in intrinsic quantum wells indicates clearly the importance of spin relaxation of the exciton and the necessity of a theory of exciton spin dynamics including exchange. The creation of heavy-hole excitons leaves little phase space for individual spin relaxation of the constituent carriers.

A number of basic mechanisms for spin relaxation have been studied in connection with the optical orientation in bulk semiconductors.[6] However, the confinement introduced by the quantum well barriers affects these mechanisms in quite a pronounced way. The distinction of the spin dynamics in quantum wells from the bulk arises from three factors due to the well structure: the subband electronic structures, the enhancement of the excitonic interaction, and the high mobility of the carriers. An example of the effect of the valence subband structure is the slowdown of the valence hole spin relaxation in the quantum well compared with the bulk, leading to a different and more satisfactory explanation of the cw luminescence polarization in quantum wells.[7,12] The enforced close proximity of the electron-hole pair leading to a strong exciton binding in a quantum well is also expected to lead to a strong spin relaxation due to exciton exchange.[8,10] Mechanisms which depend on motional narrowing,[13] such as the D'yakonov-Perel (DP) mechanism,[14] are sensitive to carrier momentum relaxation and should, therefore, have different effects for the high-mobility quantum well than for the dirty bulk semiconductor. Further experimental and theoretical studies are needed to assess the relative quantitative importance of these different spin-relaxation processes in confined systems.

A specific theory of spin relaxation including the excitonic effect is needed. This would be germane not only to the spin relaxation for resonantly created excitons in intrinsic quantum wells, but also to the highly excited electrons and holes in the same structures because, as described earlier, an electron-hole pair rapidly relaxes into an exciton with a large in-plane wave vector $\mathbf{K}$, whereupon the spin-relaxation processes are determined by the dynamics occurring within the exciton branch.

In this paper, we present a theory of spin relaxation of

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an exciton in a quantum well. The exchange interaction[15] between electron and hole is considered in conjunction with the center-of-mass motion to explain a spin-flip process for excitons. The exchange interaction consists of two parts: a long-range part of the Coulomb interaction and a short-range part of the Coulomb interaction plus umklapp terms. We analyze both contributions to spin relaxation. For the heavy-hole exciton, the long-range part is dominant, because the short-range exchange does not directly flip the heavy-hole spin and requires a second-order process involving spin flip from a heavy-hole spin state to a light-hole spin state and mixing in the valence band to go from the light-hole state to a heavy-hole state.

The relevance of the exchange interaction in luminescence polarization in quantum wells can be tested by the effect of a magnetic field or an electric field applied along the growth axis of the quantum well. The magnetic field changes the spin splitting and thus affects the spin flip. The electric field changes the distance between the electron and hole in the exciton along the growth axis and, hence, the exchange interaction. A theory is given of the effect of low magnetic fields. A calculation is given here of electric field dependence of the spin-relaxation time of the exciton. This is of particular interest because Shah[16] and collaborators have independently thought of using the electric field to change the spin-relaxation time and are carrying out luminescence experiments on quantum wells to test this idea.

The rest of this paper is organized as follows. In Sec. II, we survey the mechanisms for the electron, hole, and exciton spin relaxations and investigate their roles in determining the time dependence of the total intensity and polarization in emission or absorption. Characteristic time dependence of the total and polarization intensities leads us to infer the importance of the exciton exchange spin flip. In Sec. III, we set down the exchange interaction, including the long-range and short-range parts, appropriate for a quantum well. We also give the physical aspects of a theory of spin relaxation of an exciton driven by the long-range exchange and the short-range exchange. We relegate the details of the theory of the exciton spin dynamics in terms of the density-matrix representation to the Appendix, which also includes the spin effect of the magnetic field. As a specific example, we consider the heavy-hole exciton. The method is similar for the light-hole exciton. The results for the spin-relaxation times for the heavy-hole excitons in an infinite-barrier quantum well are presented in Sec. IV, including the electric field dependence. Section V contains a summary of the physical picture of the exciton spin relaxation and of the relevance of the theory to experimental observation.

II. TIME DEPENDENCE OF EXCITON SPIN POPULATIONS

The study of the exciton spin relaxation requires a clear representation of the spin states of the exciton. The spin state of an exciton is a direct product of the conduction electron and valence hole spin states. In a bulk III-V direct-gap semiconductor, the conduction-band edge has Γ_6 symmetry and the z component of the electron's angular momentum is represented by s with $s = \pm\frac{1}{2}$. The valence-band edge has symmetry Γ_8 and the index m_v which describes the valence electron spin states is isomorphous to the z component of the angular momentum of spin $\frac{3}{2}$. We shall represent the valence-hole spin states by the time-reversed states of the electronic Bloch states and associate the index m_h with the *hole* spins so that $m_h = -m_v$. A basis function for the exciton will be represented by the exciton wave function composed of an electron from a single conduction subband ν and a single valence-hole band μ from the diagonal part of the Luttinger Hamiltonian.[17] Subband mixing in the exciton state through the Coulomb attraction will be neglected in this paper. Hole subband mixing from the off-diagonal part of the Luttinger Hamitonian will be treated by perturbation theory because it is found to be small for the exciton states.[18]

The exciton basis state, thus, assumes the form

$$\psi_{s,m_h}(\mathbf{r_e},\mathbf{r_h}) = u_s(\mathbf{r_e})u_{m_h}(\mathbf{r_h})\xi_{\nu c}(z_e)\ \zeta_{\mu jh}(z_h) \times\phi_{n\ell(j)}(\boldsymbol{\rho})\ \frac{e^{i\mathbf{K}\cdot\mathbf{R}_{\|j}}}{\sqrt{A}}\ . \tag{2.1}$$

The Bloch wave parts are from the conduction electron $u_s(\mathbf{r_e})$ with spin s and from the valence hole $u_{m_h}(\mathbf{r_h})$ with spin m_h. The subband envelope functions along the growth axis are denoted by $\xi_{\nu c}(z_e)$ and $\zeta_{\mu jh}(z_h)$ for an electron in the νth and a hole in the μth subband, where $j = h$(heavy) or l(light), depending on whether the hole spin m_h is $\pm\frac{3}{2}$ or $\pm\frac{1}{2}$, respectively. The two-dimensional j (heavy- or light-hole) exciton's bound state with principal quantum number n and azimuthal quantum number ℓ is $\phi_{n\ell(j)}(\boldsymbol{\rho})$, with $\boldsymbol{\rho} = \boldsymbol{\rho}_e - \boldsymbol{\rho}_h$ being the relative coordinate in the quantum-well plane (the xy plane). The last factor represents the two-dimensional motion of the center-of-mass position $\mathbf{R}_{\|j}$, with wave vector $\mathbf{K}$ in the xy plane.

When a photon is absorbed to resonantly excite an exciton with electron and hole spins denoted by (s, m_h), the polarization of light is governed by the selection rule[19] for the transition matrix element which takes the form of angular momentum conservation along the z axis,

$$s + m_h = m_p, \tag{2.2}$$

where m_p takes the value ±1 for the $\sigma\pm$ polarizations, i.e., light propagating along the z axis with circular polarization, and m_p takes on the value zero for the π polarization, i.e., light propagating normal to the z axis with the electric vector along the z direction. The $(s + m_h) = \pm2$ excitonic states are said to be optically inactive since they cannot be created by single-photon absorption although they can be excited by two-photon processes.

Consider, for example, the lowest energy heavy-hole exciton. The $m_h = \pm\frac{3}{2}$ components dominate the constituent valence hole of the exciton, so that the total exciton spin, from Eq. (2.2), is ±1 or ±2. From the selection rule described above, the spin ±1 excitons are

optically active and the spin ± 2 ones are inactive. The exciton spin can be changed by flipping the spin of either the electron or the hole or by simultaneously flipping the spin of both constituents. The electron can relax its spin by the D'yakonov-Perel[14] or Elliot-Yafet[20] mechanism. Both mechanisms require substantial phase space for momentum scatterings to be effective in changing the spin. The valence hole can relax its spin through energy relaxation which changes the heavy- and light-hole mixing of the valence state.[7] Since the ground-state heavy-hole exciton is made up of a linear combination of electron and hole states at an energy range from the band edges of the same order of magnitude as the exciton binding energy, there is unlikely to be sufficient phase space for the electron or hole for these mechanisms to be effective. In addition, the electron spin relaxation can be ruled out because its relaxation time is at least an order of magnitude longer than the exciton relaxation time.[8]

Consider an optical experiment with circularly polarized light incident or emitted along the growth axis of the quantum well. For an optically active heavy-hole exciton (with total spin ± 1), spin flip of either constituent would transform the exciton to spin ± 2, an optically inactive state. In that case, the decay of the absorption or emission of one polarization, say $\sigma+$, would not increase the other polarization $\sigma-$. We have investigated the results of the rate equations for N_{m_p}, the population of excitons of total spin m_p,

$$\frac{d}{dt}\begin{pmatrix} N_{+2} \\ N_{+1} \\ \\ N_{-1} \\ \\ N_{-2} \end{pmatrix} = \begin{pmatrix} -(W_e^+ + W_h^+) & W_e^- & W_h^- & 0 \\ W_e^+ & -(\frac{1}{\tau_R} + W_{\rm ex} + W_e^- + W_h^-) & W_{\rm ex} & W_h^+ \\ W_h^+ & W_{\rm ex} & -(\frac{1}{\tau_R} + W_{\rm ex} + W_e^- + W_h^-) & W_e^+ \\ 0 & W_h^- & W_e^- & -(W_h^+ + W_e^+) \end{pmatrix} \begin{pmatrix} N_{+2} \\ N_{+1} \\ \\ N_{-1} \\ \\ N_{-2} \end{pmatrix}, \tag{2.3}$$

where the transition rates are, for exchange,

$$W_{\rm ex} = 1/(2\tau_{\rm ex}) , \tag{2.4}$$

and for the electron spin flip,

$$W_e^{\pm} = \frac{1}{\tau_e(1 + e^{\pm \Delta_h / k_B T})} , \tag{2.5}$$

and a similar expression for the hole spin flip, Δ_h being the energy of the optically active states above the dark states [see Eq. (3.10)]. The various relaxation times are a recombination time τ_R and three spin relaxation times, τ_e for electron spin s flipping, τ_h for hole spin m_h flipping, and $\tau_{\rm ex}$ for exciton spin flipping between $m_p = \pm 1$. Since the exciton spin flip is driven by the exchange interaction it will only affect the optically active states [see Eqs. (3.4) and (3.10) below].

We illustrate the results for three cases, all with $T = 2K$, $\Delta_h = 0.1$ meV, and $\tau_R = 400$ ps: case I,

$$\tau_{\rm ex} = 50 \text{ ps}, \quad \tau_h = \infty, \quad \tau_e = \infty;$$

case II,

$$\tau_{\rm ex} = 50 \text{ ps}, \quad \tau_h = 100 \text{ ps}, \quad \tau_e = 200 \text{ ps};$$

case III,

$$\tau_{\rm ex} = \infty, \quad \tau_h = 100 \text{ ps}, \quad \tau_e = 200 \text{ ps}.$$

The second case represents a typical case with the times of the same order as measured[8] except that we have taken a somewhat longer hole spin-relaxation time on the ground that the phase space of the hole in an exciton is reduced compared with that of free holes. Cases I and III represent respectively the limiting cases of no single-particle spin relaxation and of no exciton spin relaxation.

Figure 1(a) shows the calculated time dependence of the spin populations of the optically active excitons, $N_{\pm 1}$, which are proportional to intensities of absorbed or emitted light of polarization $\sigma\pm$, given an initial spin population entirely polarized in the $+1$ spin state. In the top panel which corresponds to case I of only exciton spin flip, the growth of N_{-1} and the decay of N_{+1} towards the average population allowing for the depletion due to recombination are symmetrical. The bottom panel shows the other extreme of having only single-particle relaxations, dominated by the hole spin component. The initial $+1$ spin state is rapidly converted by the hole spin relaxation into dark states -2. The -1 spin population has a slow concave growth, in contrast to the fast initial convex growth in the top panel. The middle panel shows the results of the reasonably realistic model of coexisting exciton spin and individual-particle spin relaxations. The fast initial decay of N_{+1} is dominated by the hole spin relaxation. The fast initial rise of N_{-1} is characteristic of the exchange interaction but the maximum value it can achieve before recombination sets in is lower than that in the top panel.

The plot of polarization versus time involves the convolution of all four times, making its behavior difficult to interpret from the graph. Instead, we plot in Fig. 1(b) the logarithms of total intensity $I = N_{+1} + N_{-1}$ and the spin (or difference) intensity $S = N_{+1} - N_{-1}$. The top panel shows a single-time exponential decay for both quantities, given by the recombination time and the exciton spin-flip time. In the other two cases, each intensity shows two decay times. Analytical proof of this follows easily from Eq. (2.3), which yields two simultaneous equations for the total intensities $N_{+1} + N_{-1}$ and $N_{+2} + N_{-2}$ states decoupled from the two equations for

the spin intensities $N_{+1}-N_{-1}$ and $N_{+2}-N_{-2}$. Analysis of time-resolved luminescent polarization or pump-and-probe measurements in terms of I and S then offers a way to deduce the four times.

In the pump-and-probe experiment of Tackeushi *et al.*[21] at room temperature, the decrease of the absorption of the polarization $\sigma+$ parallel to the excitation polarization is accompanied by an almost exactly compensating increase of the opposite polarization $\sigma-$ on a time scale much shorter than the recombination time. This corresponds to case I, dominated by exchange spin flip. Roussignol *et al.*[11] also claimed to observe exciton polarization relaxation and not free-electron or hole relaxation.

Similar behavior to cases II and III shown in Fig. 1(b) was observed by Damen *et al.*[4] They have interpreted the fast initial drop on the intensity I as due to exciton thermalization to large $\mathbf{K}$ states. These states are not able to radiate and, in this sense, they are analogous to the dark states ± 2 considered here. The invocation of the thermalization processes was based on the temperature dependence exhibited by the fast initial decay of I. If the experiments were carried out at lower temperatures, where thermalization effects would not be important, an equally suitable explanation could be given considering only the single-particle spin relaxation. This was in fact done by Bar-Ad and Bar-Joseph[9] whose experimental data show the unremarked fact that the $\sigma+$ component drops faster then the rise of the $\sigma-$ component initially. Figure 2(a) shows our best fit of $\ln I$ and of $\ln S$ (solid curves) to the experimental data (dashed curves) for the asymmetric well of Ref. 9, using $T = 4.2$ K, $\Delta_h = 0.25$ meV, $\tau_R = 250$ ps, $\tau_{ex} = 65$ ps, $\tau_h = 20$ ps, and $\tau_e = 300$ ps. Figure 2(b), plotting the spin populations, N_+ and N_-, versus time, shows similar degree of good fit, validating the form of the multiple exponential decay. The quality of the fit is very sensitive to the spin splitting Δ_h between ± 1 and ± 2 states and sensitive to the hole spin relaxation, τ_h, but insensitive to 10% variation in the other relaxation times. Thus, we agree with the authors that the initial drop is a measure of the hole spin relaxation but we disagree with the interpretation of the longer time as due to the electron spin relaxation. Instead, we argue from above considerations that it is a qualitative measure of the exciton spin relaxation. In the following, we shall estimate the exciton spin-relaxation time from the exchange mechanism, which supports our interpretation.

III. SPIN RELAXATION VIA THE EXCHANGE INTERACTION

The theory of spin relaxation of an exciton via exchange has two components. One is the spin flip via the exchange interaction and the other is the motional

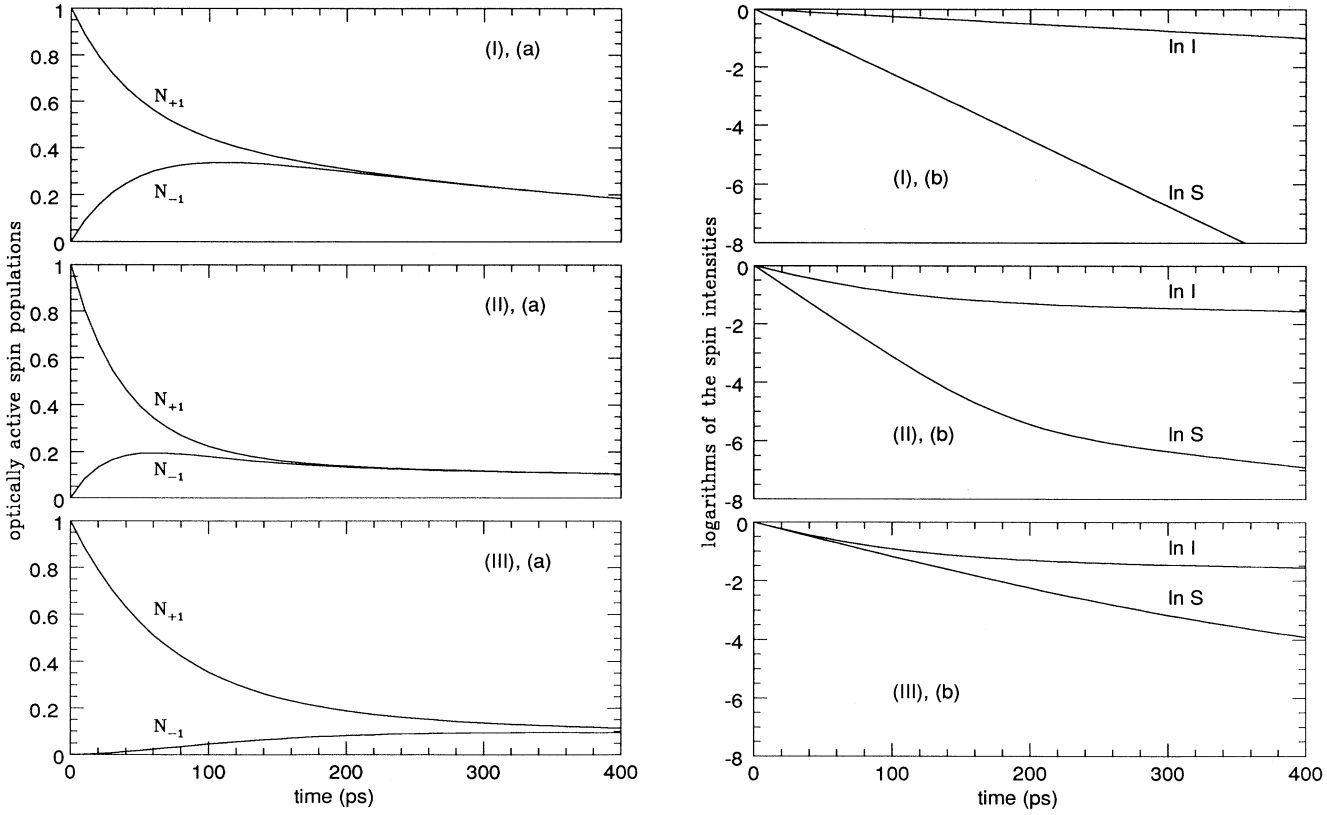

FIG. 1. (a) Time dependence of spin populations of heavy-hole excitons. (b) Time dependence of logarithms of the total and spin intensities (in arb. units). See text for the designation of the different panels.

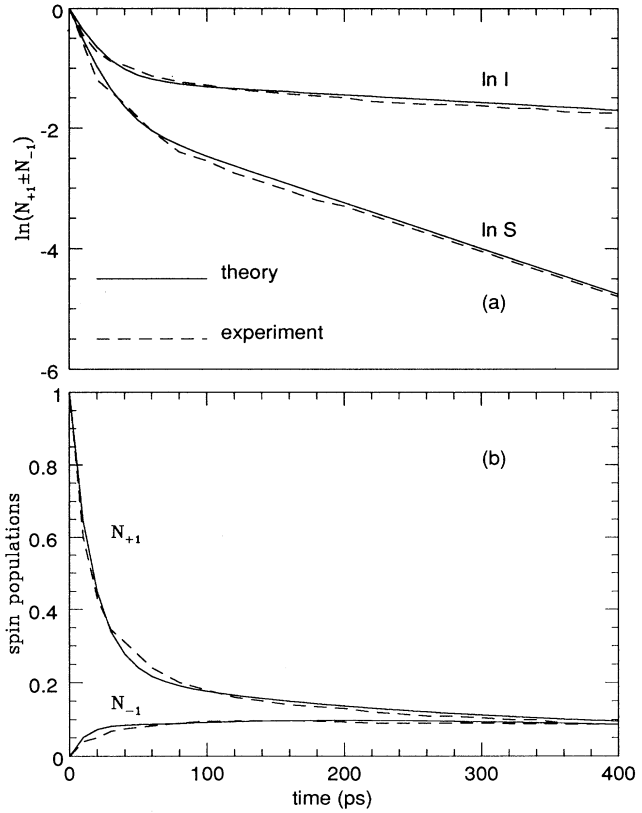

FIG. 2. (a) Theoretical fit of time dependence of the total and difference intensities (in arb. units) to the experiment of Ref. 9. See text for the parameters used. (b) Comparison of the theoretical and experimental time dependence of the exciton individual spin populations.

narrowing[13] through the scattering of the center of mass of the exciton. In addition to the direct Coulomb attraction between the electron-hole pairs, as illustrated by the Feynman diagram in Fig. 3(a), which binds them into an exciton, there are corrections to the effective-mass equation which are the exchange counterpart of the direct processes,[22] as illustrated in the lowest two orders of perturbation theory by the diagrams in Figs. 3(b) and 3(c). Figure 3(b) contains the long-range Coulomb interaction, and for excitons confined in a two-dimensional quantum well is the dipolar interaction dependent on the momentum transfer of the Coulomb line and, therefore, the center-of-mass momentum of the exciton in the interface plane of the quantum well. The same diagram also

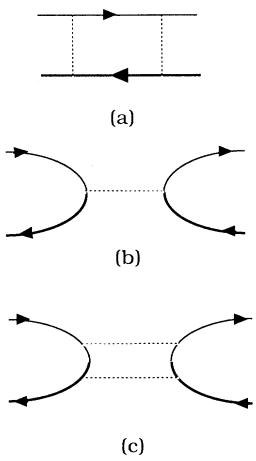

FIG. 3. Feynman diagrams for electron-hole interaction: (a) direct second-order process; (b) lowest-order exchange; (c) second-order exchange.

contains umklapp terms of the Coulomb line, i.e., the center-of-mass momentum plus a reciprocal-lattice vector times $\hbar$, which contribute to the short-range part of the exchange. Figure 3(c) gives an example of the higher order processes which contribute only to the short-range part of the exchange. Details of the long-range part and the short-range part of the exchange and their role in the exciton spin relaxation are given in two following subsections.

A. The long-range part

We express the long-range part of the exciton exchange in terms of the basis of *unmixed* exciton ground states for the quantum well, Eq. (2.1). By using the long-range part (also known as the nonanalytical part) of the exchange for the bulk semiconductor already obtained,[15] we take the matrix elements between two exciton ground states with fixed subband indices and in-plane center-of-mass wave vectors $(\nu, \mu, \mathbf{K})$ and $(\nu', \mu', \mathbf{K}')$ for various spin states (s, m_h) and (s', m_h'), which may be written in matrix form, with (s, m_h) and (s', m_h') in the order,

$$(\tfrac{1}{2}, \tfrac{3}{2}), (-\tfrac{1}{2}, \tfrac{3}{2}), (\tfrac{1}{2}, \tfrac{1}{2}), (-\tfrac{1}{2}, \tfrac{1}{2}), (\tfrac{1}{2}, -\tfrac{1}{2}), (-\tfrac{1}{2}, -\tfrac{1}{2}),$$

$$(\tfrac{1}{2}, -\tfrac{3}{2}), (-\tfrac{1}{2}, -\tfrac{3}{2}),$$

as

$$H^{\mathrm{LR}} = \frac{3}{8}\,\delta_{\mathbf{K},\mathbf{K}'}\frac{\Delta E_{\mathrm{LT}}}{|\phi_{3\mathrm{D}}(0)|^2}\begin{bmatrix}
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & F^0_{hh}K^2 & -\frac{F^0_{hl}}{\sqrt{3}}K^2 & -\frac{2F^1_{hl}}{\sqrt{3}}K_- & \frac{2F^1_{hl}}{\sqrt{3}}K_- & -\frac{F^0_{hl}}{\sqrt{3}}K_-^2 & F^0_{hh}K_-^2 & 0 \\
0 & -\frac{F^0_{lh}}{\sqrt{3}}K^2 & \frac{F^0_{ll}}{3}K^2 & \frac{2F^1_{ll}}{3}K_- & -\frac{2F^1_{ll}}{3}K_- & \frac{F^0_{ll}}{3}K_-^2 & -\frac{F^0_{lh}}{\sqrt{3}}K_-^2 & 0 \\
0 & -\frac{2F^1_{lh}}{\sqrt{3}}K_+ & \frac{2F^1_{ll}}{3}K_+ & \frac{4F^2_{ll}}{3} & -\frac{4F^2_{ll}}{3} & \frac{2F^1_{ll}}{3}K_- & -\frac{2F^1_{lh}}{\sqrt{3}}K_- & 0 \\
0 & \frac{2F^1_{lh}}{\sqrt{3}}K_+ & -\frac{2F^1_{ll}}{3}K_+ & -\frac{4F^2_{ll}}{3} & \frac{4F^2_{ll}}{3} & -\frac{2F^1_{ll}}{3}K_- & \frac{2F^1_{lh}}{\sqrt{3}}K_- & 0 \\
0 & -\frac{F^0_{lh}}{\sqrt{3}}K_+^2 & \frac{F^0_{ll}}{3}K_+^2 & \frac{2F^1_{ll}}{3}K_+ & -\frac{2F^1_{ll}}{3}K_+ & \frac{F^0_{ll}}{3}K^2 & -\frac{F^0_{lh}}{\sqrt{3}}K^2 & 0 \\
0 & F^0_{hh}K_+^2 & -\frac{F^0_{hl}}{\sqrt{3}}K_+^2 & -\frac{2F^1_{hl}}{\sqrt{3}}K_+ & \frac{2F^1_{hl}}{\sqrt{3}}K_+ & -\frac{F^0_{hl}}{\sqrt{3}}K^2 & F^0_{hh}K^2 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{bmatrix}. \tag{3.1}$$

We have defined $\Delta E_{\rm LT}$ to be the longitudinal-transverse splitting in bulk semiconductor and used the three-dimensional hydrogenic exciton wave function at zero relative distance,[23] $\phi_{\rm 3D}(0)$. We also have defined the center-of-mass momentum $\mathbf{K}=(K_x,K_y)$ and $K_\pm=(K_x\pm iK_y)$. The form factors are written as, for integer p,

$$F^p_{j',j}(K)=\phi_{1s(j')}(0)\;\phi_{1s(j)}(0) \times\int\frac{dQ_z}{2\pi}\;\frac{Q_z^p}{K^2+Q_z^2}\;f_{j'j}(Q_z)\;, \tag{3.2}$$

with

$$f_{j'j}(Q_z)=\int dz\int dz'\;\xi_{\nu' c}(z')\;\zeta_{\mu' j' h}(z') \times e^{iQ_z(z-z')}\;\xi_{\nu c}(z)\zeta_{\mu j h}(z)\;. \tag{3.3}$$

The confinement splits the valence-band edge into heavy- and light-hole subbands with energy differences of the order of 10 meV, for a typical quantum well around 100 Å wide. Since $\Delta E_{\rm LT}$ is considerably smaller than this,[23] we do not expected a strong heavy-hole–light-hole coupling to take place via the long-range exchange interaction (3.1). Thus, we may restrict our attention to the heavy-hole exciton (*hhe*) subspace where we can write down the matrix elements of the long-range interaction acting only on the spins of the ground state *hhe* (i.e., $\nu,\mu,\nu',\mu'=1$ and $j,j'=h$) in the matrix form in the order $(\frac{1}{2},\frac{3}{2}),(-\frac{1}{2},\frac{3}{2}),(\frac{1}{2},-\frac{3}{2}),(-\frac{1}{2},-\frac{3}{2})$,

$$H^{\rm LR}_{(hhe)}=\delta_{\mathbf{K},\mathbf{K}'}\;\frac{3}{16}\;\frac{|\phi_{1s(h)}(0)|^2}{|\phi_{\rm 3D}(0)|^2}\;\Delta E_{\rm LT}\frac{f(K)}{K} \times\begin{bmatrix}0&0&0&0\\0&K^2&K_-^2&0\\0&K_+^2&K^2&0\\0&0&0&0\end{bmatrix}\;. \tag{3.4}$$

The form factor $f(K)$ in the above equation is given by

$$f(K)=\int dz\int dz'\;\xi_{1c}(z)\zeta_{1hh}(z)\;e^{-K|z-z'|} \times\xi_{1c}(z')\zeta_{1hh}(z')\;. \tag{3.5}$$

The long-range interaction for *hhe*'s in a quantum well vanishes linearly with K when $\mathbf{K}$ goes to zero.[24] Therefore, due to momentum conservation, no longitudinal-transverse splitting is expected for excitations with light propagating along the growth axis of the well.

B. The short-range part

The other exchange term which should be included in the exciton's effective-mass equation is due to the short-range part [also called analytical part; see Fig. 3(c)] of the Coulomb interaction between the electron and hole in the same site.[15] Using the exciton basis function (2.1) for two-dimensional excitons with different $\mathbf{K}$'s, the spin Hamiltonian for the short-range interaction[25] has the form

$$H^{\rm SR}=\frac{3}{4}\;\delta_{\mathbf{K},\mathbf{K}'}\;\frac{\Delta E_{\rm SR}}{|\phi_{\rm 3D}(0)|^2}\begin{bmatrix}0&0&0&0&0&0&0&0\\0&I_{hh}&\frac{-I_{hl}}{\sqrt{3}}&0&0&0&0&0\\0&\frac{-I_{lh}}{\sqrt{3}}&\frac{I_{ll}}{3}&0&0&0&0&0\\0&0&0&\frac{2I_{ll}}{3}&\frac{-2I_{ll}}{3}&0&0&0\\0&0&0&\frac{-2I_{ll}}{3}&\frac{2I_{ll}}{3}&0&0&0\\0&0&0&0&0&\frac{I_{ll}}{3}&\frac{-I_{lh}}{\sqrt{3}}&0\\0&0&0&0&0&\frac{-I_{hl}}{\sqrt{3}}&I_{hh}&0\\0&0&0&0&0&0&0&0\end{bmatrix}\;, \tag{3.6}$$

with

$$I_{j'j}=\phi_{1s(j')}(0)\;\phi_{1s(j)}(0)\;\int dz\;|\xi_{1c}(z)|^2\zeta_{1j'h}(z)\zeta_{1jh}(z)\;, \tag{3.7}$$

if we only consider the excitons associated with the two topmost hole subbands, one heavy and one light. This interaction splits the eightfold degenerate exciton in bulk into two levels, with total angular momentum $J=1$ lying above $J=2$ in energy by $\Delta E_{\rm SR}$ in the bulk.[23]

It is important to note that the short-range exchange, which takes the form $\boldsymbol{\sigma_e}\cdot\mathbf{j}_h$, conserves total spin and only couples a *hhe* state to a *lhe* state. Thus, on the one hand, the confinement may enhance the short-range interaction between electrons and holes and, on the other hand, it also lifts the degeneracy between *hhe*'s and *lhe*'s which weakens the effect of the exchange in the spin relaxation process. In particular, the conservation of the total spin forbids the direct transitions between the optically active *hhe* spin states $(-\frac{1}{2},+\frac{3}{2})$ and $(+\frac{1}{2},-\frac{3}{2})$. An additional spin nonconserving mechanism is necessary to accomplish that.

Various mechanisms could be used to complete the channel between the two optically active *hhe* states, for instance, scattering-related processes which flip the spin of one particle (or even of both particles). As already argued in the previous section, this is rather unlikely to happen when excitons are being considered. We have chosen to investigate here the spin-orbit coupling in the valence band. This is a very strong effect in the bulk and has been commonly used to support the assumption of a rapidly depolarized spin population of holes.[6]

In quantum wells, the hole relaxation is slowed down by the subband formation[7] as well as by the phase space limitation in an exciton as explained above.

The spin-orbit coupling in the valence band is introduced via the $\mathbf{k}\cdot\mathbf{p}$ terms in the Luttinger Hamiltonian, H_{kp}, which has off-diagonal terms like[26]

$$S = \frac{\hbar^2}{m_0}\sqrt{3}\gamma_3 k_z k_-,\ R = \frac{\hbar^2}{m_0}\frac{\sqrt{3}}{2}(-\bar{\gamma}k_-^2 + \mu k_+^2)\ , \quad (3.8)$$

where

$$\bar{\gamma} = \tfrac{1}{2}(\gamma_2 + \gamma_3),\ \mu = \tfrac{1}{2}(\gamma_3 - \gamma_2),$$

with $\mathbf{k}$ being the momentum operator acting only on the hole coordinates and γ's the Luttinger parameters for the host semiconductor. We treat the off-diagonal part of H_{kp} by perturbation theory to take into account the heavy- and light-hole mixing. Thus, a *hhe* ground state is given by

$$\psi^{\rm mix}_{s,m_h} = \psi_{s,m_h} + \sum_{m'_h=\pm\frac{1}{2}} \frac{\langle\psi_{s,m'_h}|H_{kp}|\psi_{s,m_h}\rangle}{E_{hhe} - E_{lhe}}\ \psi_{s,m'_h}\ , \quad (3.9)$$

where ψ_{s,m'_h} are all possible light-hole exciton states with energies E_{lhe} of the diagonal part of the Luttinger Hamiltonian. The ψ_{s,m_h} are similarly the unperturbed [i.e., unmixed, cf. Eq. (2.1)] *hhe* ground states with spins $(s, m_h = \pm\frac{3}{2})$ and energy E_{hhe}.

For practical computations, we shall consider only the ground states for the *hhe* and *lhe* associated with the topmost heavy- and light-hole subbands. This can be justified if we recall that the well confinement introduces energy splittings between subbands of order of 10 meV for a typical quantum well with width around 100 Å, whereas the correction due to mixing is small, about 1 meV. Therefore, the topmost light-hole subband gives the major contribution to Eq. (3.9).

Instead of calculating the matrix elements of the short-range exchange interaction with the simple *hhe* ground states in Eq. (2.1), as done before in Eq. (3.4) for the long-range case, we now use the *hhe* ground states which include mixing, i.e., Eq. (3.9). We obtain the result

$$H^{\rm SR}_{(hhe)} = \delta_{\mathbf{K},\mathbf{K}'}\begin{bmatrix} E_h & 0 & 0 & 0 \\ 0 & E_h+\Delta_h & W(K) & 0 \\ 0 & W^{\dagger}(K) & E_h+\Delta_h & 0 \\ 0 & 0 & 0 & E_h \end{bmatrix}\ , \quad (3.10)$$

where the diagonal part is the eigenvalue solution neglecting the hole spin mixing, E_h being the *hhe* energy and Δ_h an exchange induced splitting between the dark and optically active states given by

$$\Delta_h = \frac{3}{4}\,\Delta E_{\rm SR}\,\frac{I_{hh}}{|\phi_{\rm 3D}(0)|^2}\ ,$$

with I_{hh} from Eq. (3.7). The off-diagonal terms arise from contributions of both the valence-band mixing via the R terms in the Luttinger Hamiltonian, Eq. (3.8), and the exchange matrix elements between heavy- and light-hole states:

$$W(K) = -H^{\rm SR}_{hl}\frac{1}{E_0}R_{lh} - R_{hl}\frac{1}{E_0}H^{\rm SR}_{lh}, \quad (3.11)$$

where $E_0 = (E_{lhe} - E_{hhe})$ is the energy splitting between the two exciton ground states associated with the two topmost valence subbands and the spin states in the matrix elements of $H^{\rm SR}$ and of R are abbreviated to h and l. The matrix elements are given by

$$R_{lh} = 4\ p\ L_{lh}\ Q_{lh}\ \left(\tilde{m}^2 + \frac{B(\beta)}{a_{0h}a_{0l}K^2}\right)\ \frac{K_-^2}{(1+\beta^2)^{3/2}} \quad (3.12)$$

and

$$H^{\rm SR}_{hl} = -\frac{3}{4}\ \frac{\Delta E_{\rm SR}}{|\phi_{\rm 3D}(0)|^2}\ \frac{I_{hl}}{\sqrt{3}}\ , \quad (3.13)$$

in which we have used the two-dimensional exciton wave function in the form

$$\phi_{1s(j)}(\rho) = \sqrt{8/\pi a_{0j}^2}\ e^{(-2\rho/a_{0j})}\ , \quad (3.14)$$

and the quantities in Eq. (3.12) defined by

$$p = -\frac{\sqrt{3}}{2}\frac{(\gamma_2+\gamma_3)\hbar^2}{2m_0}\ , \quad (3.15)$$

$$L_{lh} = \int dz\ \zeta_{1hh}(z)\ \zeta_{1lh}(z)\ , \quad (3.16)$$

$$Q_{lh} = \int d^2\rho\ \phi_{1s(h)}(\rho)\ \phi_{1s(l)}(\rho) = \frac{4\ a_{0h}\ a_{0l}}{(a_{0h}+a_{0l})^2}\ , \quad (3.17)$$

$$\tilde{m} = \frac{m_{h\|}\ m_{l\|}}{m_{l\|}(m_e+m_{h\|}) + m_{h\|}(m_e+m_{l\|})}\ , \quad (3.18)$$

$$\beta = -\frac{K}{2}\ \frac{m_e(m_{h\|}-m_{l\|})}{(m_e+m_{h\|})(m_e+m_{l\|})}\ \frac{1}{(1/a_{0h}+1/a_{0l})}\ , \quad (3.19)$$

and

$$B(\beta) = \frac{(\beta^2+1)}{\beta^2}\ [\sqrt{\beta^2+1}-1]^2\ . \quad (3.20)$$

We have used the axial approximation[26] to obtain Eq. (3.12), neglecting the anisotropy of the hole energy in the k space, i.e., we have set $\mu = \frac{1}{2}(\gamma_3 - \gamma_2) = 0$.

Similar to the spin flip of a *hhe* by a combination of band-mixing and short-range exchange via the light-hole states, the same can be accomplished by the long-range exchange. Replacing $H^{\rm SR}$ by $H^{\rm LR}$ in Eq. (3.11) leads to

$$W_{\text{LR-mix}} = -H_{hl}^{\text{LR}} \frac{1}{E_0} R_{lh} - R_{hl} \frac{1}{E_0} H_{lh}^{\text{LR}} , \tag{3.21}$$

with

$$H_{hl}^{\text{LR}} = -\frac{3}{8} \frac{\Delta E_{\text{LT}}}{|\phi_{\text{3D}}(0)|^2} \frac{F_{hl}^0}{\sqrt{3}} K^2 , \tag{3.22}$$

where F_{hl}^0 was defined in Eq. (3.2).

C. Exciton spin relaxation

Equations (3.4), (3.10), and (3.21) yield the exchange-driven spin flip among the heavy-hole exciton states, either directly, or through a combination of an exchange process to a light-hole exciton and then back to a heavy-hole exciton state by means of the valence-band mixing. As an example, consider the case of resonant excitation of heavy-hole excitons with spins $(s, m_h) = (-\frac{1}{2}, +\frac{3}{2})$ by a positive-circularly polarized light normally incident on a quantum well. Spin-relaxation produces spin states $(+\frac{1}{2}, -\frac{3}{2})$ which, on recombination, emit negative-circularly polarized light. The off-diagonal terms of Eqs. (3.4), (3.10), and (3.21) (all added together to include interference effects) are responsible for the coupling between these two exciton spin states with *finite* center-of-mass momentum. Neglecting the dark states, the dynamics is that of a pseudo-spin-$\frac{1}{2}$ system. The off-diagonal terms of the effective spin Hamiltonian represent an effective magnetic field in the xy plane. If such a field is fixed (due to fixed center-of-mass wave vector, $\mathbf{K}$) the pseudospin (i.e., the polarization) will precess about this field. Now, if this is a random-oscillating field (i.e., $\mathbf{K}$ is being changed by scattering) which changes its direction faster than the pseudospin precession, then a spin-relaxation process ensues. The *spin dynamics* which we have just described is, of course, analogous to the one observed in the motional narrowing effect in nuclear spin relaxation in metal[13] and also in the DP mechanism[14] for conduction electron spin relaxation in semiconductors. In such motional narrowing,[13] the inverse spin-relaxation time is proportional to the square of the precession frequency about the effective field $\Omega_{\parallel}$ times the momentum relaxation time τ^* [see Eq. (A12)],

$$\frac{1}{T_{s1}} = \Omega_{\parallel}^2 \tau^* . \tag{3.23}$$

Thus, the faster the momentum changes, the longer the spin relaxation is. More details on the treatment by the density-matrix formalism of the dynamics of the spin relaxation given in the Appendix, including the effect of an applied magnetic field which splits the two optically active heavy-hole exciton states.

IV. EXCITON SPIN RELAXATION TIME IN AN INFINITE-BARRIER QUANTUM WELL

A. Results in zero electric field

Calculated exciton spin relaxation times driven by exchange for resonantly excited excitons are presented here for an infinite-barrier quantum well of width L. The wave functions along the z direction are given by sines and cosines within the well, vanishing in the barriers. The wave functions are the same for electrons, heavy or light holes. The parameters describing the particles in the well are given by those from the GaAs: $\gamma_1 = 6.85$, $\gamma_2 = 2.1$, $\gamma_3 = 2.9$, $\epsilon_0 = 12.5$, $m_e = 0.067 m_0$, $\Delta E_{\text{LT}} = 0.08$ meV, $\Delta E_{\text{SR}} = 0.02$ meV.[23] The simple form of the wave functions makes it easier to evaluate the form factors appearing in Eqs. (3.4), (3.10), and (3.21), as well as facilitating the variational solution for the bound state of the exciton in the plane of the well necessary to calculate the exciton radii a_{0j} in Eq. (3.14). We also notice that in this case the inclusion of only two subbands in treating the valence-band mixing is exact because terms like L_{lh} in Eq. (3.16) will all vanish for different subbands.

In order to estimate the off-diagonal terms of the spin Hamiltonians for the short-range and long-range interactions (i.e., the magnitude of the effective magnetic field), we need to know the value of $\mathbf{K}$. Since we are only considering elastic scatterings (see the Appendix), the knowledge of the center-of-mass kinetic energy E_K for the exciton suffices. When light propagating along the growth axis is used to resonantly excite excitons, the *hhe*'s created have negligible $\mathbf{K}$'s. At low temperatures and carrier densities, the interactions of excitons with acoustic phonons, impurities, and well interfaces are responsible for a finite homogeneous linewidth Γ_h around the $\mathbf{K} = \mathbf{0}$ states.[27,28] All these finite-$\mathbf{K}$ states are able to radiate and since $\mathbf{K} \neq \mathbf{0}$ they can have their spins relaxed via the exchange interaction. Thus, one may estimate K by setting $E_K \approx \Gamma_h$. A better approximation would be obtained by averaging over the momentum distribution around $\mathbf{K} = \mathbf{0}$, especially if thermalization to large $\mathbf{K}$ states is present. However, if the temperature is kept low enough for the thermalization effects to be negligible, we expect the simple approach to give a reasonable value for the average $\mathbf{K}$ from Γ_h defined as "full width at half maximum."

Also necessary for the calculation of the spin-relaxation time T_{s1} is the value of the center-of-mass scattering time τ^* [see Eqs. (A11) and (A12) in the Appendix]. Since there is at present no direct measurement of τ^* or the related transport time for the same samples on which the time-resolved optical experiments are carried out, we extract it from the study of exciton dephasing mechanisms for which the linewidth is related to the dephasing time[29] T_2 as $\Gamma = 2\hbar/T_2$ for *homogeneous* broadening and $\Gamma = 4\hbar/T_2$ for *inhomogeneous* broadening, and by setting[30] $\tau^* \approx T_2$.

In Fig. 4 we show the dependence of the off-diagonal terms of the short-range [Fig. 4(a)] and long-range [Fig. 4(b)] interactions on the well width L for various values of the longitudinal electric field F. The dependence on F will be discussed in the next subsection, and we focus our attention here on the $F = 0$ curves. Notice that the short-range contribution is considerably weaker than that from the long-range interaction. In addition, they show different dependence on L: the long-range term increases with decreasing L and the short-range term does the opposite. Such behavior can be argued as

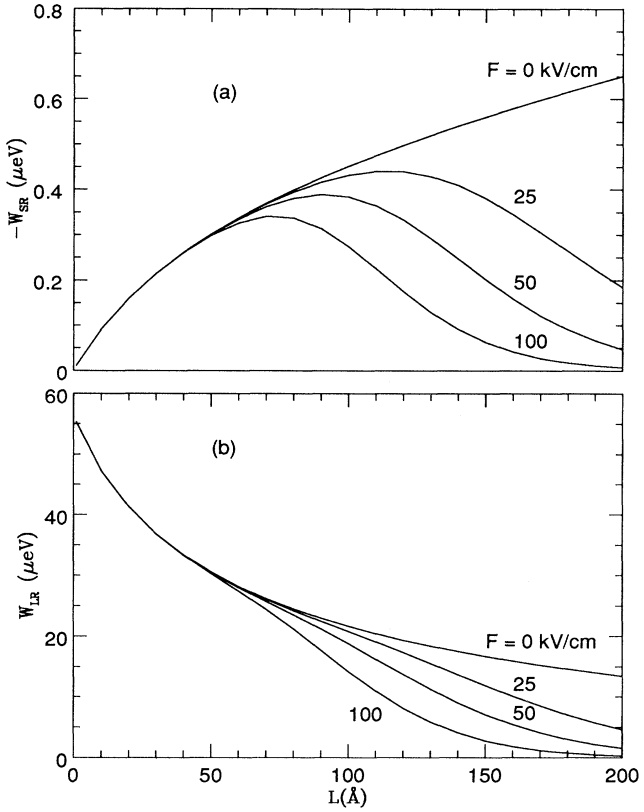

FIG. 4. Dependence of (a) short-range and (b) long-range exchange spin-flip energy on quantum well width and on longitudinal electric field strength.

a consequence of the enhancement of the exchange interaction (both the short-range and long-range parts) due to confinement, but the reduction of the valence-band mixing due to subband formation has the net effect of reducing the short-range contribution to the spin flip. In these figures we have used a value for K corresponding to a kinetic energy $E_K = 0.3$ meV, which is a reasonable value from the measured Γ_h in the actual samples. From Fig. 4 we conclude that the long-range interaction is the dominant factor in relaxing exciton spins in quantum wells and, in what follows, it is used to explain the major features observed.

We estimate the momentum scattering time by $\tau^* \approx T_2$, the exciton dephasing time which, from the homogeneous broadening relation with $\Gamma_h = 0.3$ meV, is approximately 4.4 ps. For samples in which the observed 1-meV linewidth is associated with *inhomogeneous* broadening, we estimate $T_2 \approx 2.6$ ps. In Fig. 5, the full lines are the spin-relaxation times obtained for a *hhe* with $E_K = 0.3$ meV and momentum scattering time $\tau^* = 2$, 4, and 8 ps as a function of the well width L. Also in this figure are the few experimental results (triangles[11] and square[4]) available in the literature for resonantly excited *hhe*'s. We note that our results are very close to the experimental data for $\tau^* = 4$ ps obtained from homogeneous broadening. While these results indicate that the theory is on the right track, further experiments and theoretical

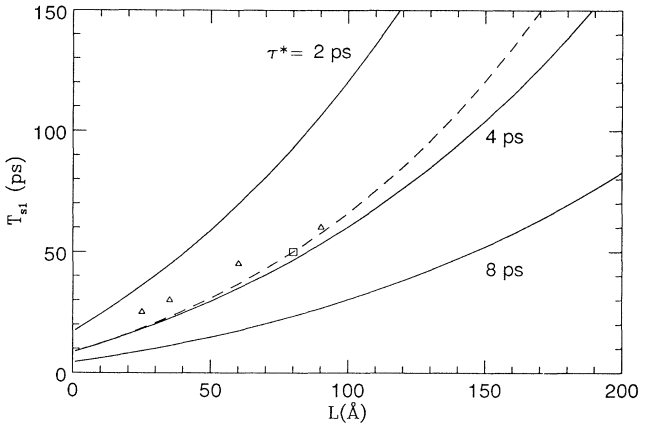

FIG. 5. Exciton spin-relaxation time vs well width for different values of the momentum scattering time, τ^*. Experimental points are from Ref. 11 (triangles) and Ref. 4 (square).

analysis are necessary to make the results quantitative.

Although different samples may have different values of τ^* and E_K, we see in Fig. 5 that the experimental points for various values of L fall quite close to a calculated curve for a constant τ^*. It may be due to a combination of circumstances: (1) the connection between the linewidth and the dephasing time, i.e., $\Gamma_h \propto \hbar/T_2$; (2) $T_2 \approx \tau^*$; and (3) $\Gamma_h \approx E_K$. The sample dependence through the variation of E_K and of τ^* is thus roughly constant in our calculation because $1/T_{s1}$ is approximately proportional to $E_K\tau^*$. This is illustrated by the proximity between the $\tau^* = 4$ ps curve and the dashed curve in Fig. 5 in which we have doubled E_K to 0.6 meV and reduced τ^* to 2 ps. For narrow wells, the change in the spin-relaxation time is slight.

B. Effects of magnetic and longitudinal electric fields

A way to test our theory of the spin-relaxation time is to make controlled changes by external means, such as stress, magnetic, or electric field along the growth axis. The effect of weak magnetic fields (i.e., without the well separated Landau levels) is formulated in the Appendix. Except for a brief comment below, further study of the magnetic field effect will be given in the future. In this subsection, we concentrate mostly on the effects of the electric field on the *hhe* spin relaxation.

An electric field applied along the z axis (the growth direction) will polarize the exciton (increasing the separation between the electron and hole). This strongly affects the long-range part of the exchange since it depend strongly on the overlap between the electron and hole wave functions. Other possible effects caused by the introduction of the electric field, such as the increase in the impurity- and interface-related scattering processes, will not be considered here.

The quantum well problem with electric field applied along its growth direction is solved using the variational approach.[31] Due to the asymmetry of the ground states in the well the terms S and $S^\dagger$ in the Luttinger Hamiltonian

will contribute. These terms give off-diagonal elements in the *hhe* spin Hamiltonian that connect the optically active states with the dark states $(s + m_h) = \pm 2$. We have done a detailed investigation of this contribution, but we found it to be very insignificant in relaxing the exciton spin to dark spin states and we do not intent to consider it here.

As expected, we see in Fig. 4 that the electric field strongly reduces the strength of the exchange interaction. This effect is more noticeable for the wider wells ($L \gtrsim 60$ Å) since those are the ones in which the excitons are more readily polarized by the field. If we neglect the change in momentum scattering processes due to the change in the field and in the well width L, i.e., fixing $E_K = 0.3$ meV and $\tau^* = 4$ ps, Fig. 6 shows how the spin-relaxation time T_{s1} varies with well width and with field strength. The spin relaxation process is strongly suppressed by the field for the wider wells. We expect this effect to be even more evident in finite-barrier quantum wells which should show a more accentuated field polarization.

Future spin-relaxation measurements in electric fields may not directly yield theses results. Analysis of the *measured* spin-relaxation time along the lines in Sec. II has to include the single-particle spin-flip processes. In the high-electric-field regime, where the exchange interaction is weakened, the single-particle spin-flip processes will dominate over exchange in spin relaxation. Therefore, a characteristic transition from case II to case III in Figs. 1(a) and 1(b) should be observed. The exciton spin relaxation in finite electric fields, extracted by the process described in Sec. II, can then be tested against the calculated field dependence given here.

Moderate magnetic field normal to the quantum well plane also inhibits the spin-relaxation process. For field strength for which the orbital effects are negligible, the Zeeman splitting of the two optically active degenerate *hhe* spin states is given by $\hbar\Omega_0$. [See the Appendix: the correct dependence of T_{s1} on the field Ω_0 is given in Eq. (A12).] This dependence is in qualitative agreement with the experimental results obtained by Ref. 9 in the moderate field regime.

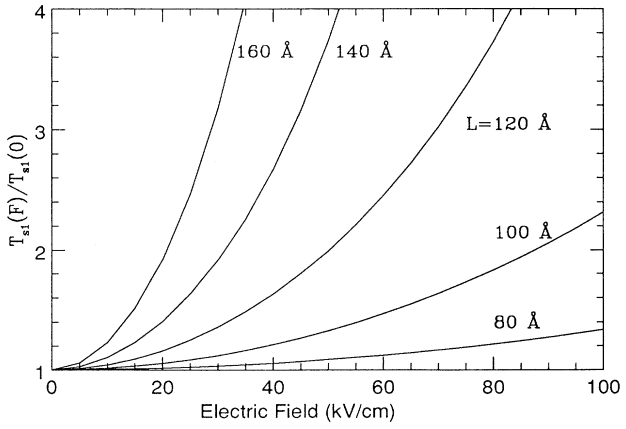

FIG. 6. Electric field dependence of exciton spin-relaxation time for various well widths.

The Appendix shows that the exciton spin-flip term can be represented by an effective depolarizing magnetic field in the well plane normal to the applied magnetic field $\hbar\Omega_0$. A distinction between *longitudinal* and *transverse* spin-relaxation times (T_{s1} and T_{s2}, respectively) can be made, following the relation $T_{s2} = 2T_{s1}$, provided that $\Omega_{\|}\tau^* \ll 1$; otherwise, the full expression, Eq. (A14), with interesting magnetic field dependence must be used. The formal derivation is given in the Appendix, but this factor of 2 is easily understood by using the fact that the in-plane polarization, say S_x, is only affected by the y component of the depolarizing field, whereas the longitudinal polarization S_z is relaxed by both x and y components. Therefore, the theory shows that luminescence experiments with linearly polarized light should exhibit a longer spin-relaxation rate driven by exchange than those performed with circularly polarized light.

The experiment done by Tackeushi *et al.*[21] showed instantaneous spin relaxation for linear excitations, probably because it was performed at room temperature where nonelastic dephasing mechanisms, such as optical-phonon scattering, may strongly enhance the spin relaxation process. On the other hand, Stolz *et al.*[30] reported a spin-relaxation time of approximately 25 ps for linear polarization, but, unfortunately, no circularly polarized light was studied at the same time to provide a test of our theoretical prediction. A very large broadening of the excitonic lines (≈ 10 meV) was observed in this experiment, indicating that extrinsic effects were certainly present and making it difficult for further comparison with our results.

V. SUMMARY

We have proposed a theory of spin relaxation of heavy-hole excitons in a quantum well. The spin-flip process occurs via the exchange Coulomb interaction between the electron and hole. The process may be viewed as due to an effective magnetic field in the well interface plane. The magnitude and direction of this field depends on the center-of-mass momentum $\mathbf{K}$, vanishing for $\mathbf{K} = \mathbf{0}$ states. The scattering of the center-of-mass momentum creates a random effective magnetic field, responsible for the exciton spin relaxation, in the same manner as any other motional narrowing spin-flip processes, with the characteristic dependence of the spin-relaxation time on the inverse momentum scattering time.

The quantum-well confinement enhances the exchange interaction over its value in bulk. The long-range exchange interaction is found to be the dominant contribution to the spin-relaxation process, whereas the short-range contribution is rendered less important by the need of the assistance of the heavy- and light-hole coupling in the valence band that is reduced by the subband formation in the lower-dimensional system.

Exciton spin relaxation leads to an observable effect as luminescence depolarization. The theory is directly applicable to resonantly excited heavy-hole excitons in undoped quantum wells. For highly excited electron-hole

pairs and doped samples, the exciton spin dynamics is important in the final stages before recombination. In the time dependence of the optical intensities of two opposing circular polarizations after resonant excitation, the exciton spin relaxation through exchange competes with other time-dependent processes, namely the individual-particle spin relaxation and the electron-hole recombination. The sum and difference of the two polarization intensities have each two relaxation times. Because the two shorter times are the exciton exchange spin relaxation and the hole relaxation, qualitatively they determine the characteristic shape of initial time dependence. Analysis of data from time-resolved polarized luminescence measurements leads to exciton spin-relaxation time and electron and hole spin-relaxation times in order-of-magnitude agreement with theoretical estimates.

Crucial tests of the theory revolve around its ability to predict changes in behavior due to changes in system and in external parameters. The exciton spin-relaxation time is inversely proportional to both the strength of the exchange interaction and the momentum scattering time. The strength of the exchange interaction depends on the well width and on the electric field applied along the growth direction of the quantum well. The theoretical and observed well-width dependence are in good accord. For finite electric fields, our calculations show that the exchange interactions are strongly inhibited in relatively wide quantum wells, lengthening their spin-relaxation times. There is yet no experiment with electric field. The dependence on momentum scatterings means that exciton spin relaxation is sample dependent and characterization by exciton linewidth measurements and by transport measurements will be helpful. In the moderate magnetic field regime, theory shows increase in the exciton spin-relaxation time but there is yet no experiment. In the high magnetic field regime, where Landau levels are important, there are experiments[32,33] but yet no theory.

From the list of topics yet to be investigated either in theory or in experiment, we see that the field of time-dependent optical polarization studies in confined systems is young. We hope that our theory provides somewhat of a framework for the direction of further studies as well as a degree of understanding of current observations.

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APPENDIX: SPIN DYNAMICS BY THE DENSITY-MATRIX METHOD

The density-matrix formalism is used here to describe the exciton spin dynamics in quantum wells. The most important spin states are the optically active states $(-\frac{1}{2},+\frac{3}{2})$ and $(+\frac{1}{2},-\frac{3}{2})$. In the following, as an illustration of how a spin-flip mechanism leads to spin relaxation, we reduce the spin Hamiltonian including connection to all other spin states to that of a pseudo-1/2-spin system. The reduced spin Hamiltonian has the form

$$H_{\rm red} = H_0 + H_1(\mathbf{K}) = \begin{bmatrix} E_+ & W(\mathbf{K}) \\ W^\dagger(\mathbf{K}) & E_- \end{bmatrix}, \tag{A1}$$

where H_0 is the diagonal matrix including an applied static magnetic field along the z axis causing an energy splitting $\hbar\Omega_0 = (E_+ - E_-)$. The off-diagonal matrix $H_1(\mathbf{K})$ is due to a $\mathbf{K}$-dependent exchange-driven spin-flip which leads to an effective magnetic field $\mathbf{\Omega}_\parallel$ in the xy plane. From Eqs. (3.4), (3.10), and (3.21), H_1 has the following form:

$$H_1(\mathbf{K}) = \frac{\hbar\,\Omega_\parallel(K)}{2} \begin{bmatrix} 0 & e^{-i2\phi} \\ e^{i2\phi} & 0 \end{bmatrix}, \tag{A2}$$

where $\Omega_\parallel(K)$ is a function of the magnitude of $\mathbf{K}$ only and ϕ is the angle between $\mathbf{K}$ and the x axis. The ϕ dependence comes from the factor $K_\pm^2$ [cf. Eqs. (3.4), (3.10), and (3.21)]. The two pseudo-1/2-spin states have angular momentum components along the z axis differing by $2\hbar$ and the total momentum conservation is maintained by the orbital angular momentum in the 2ϕ term in Eq. (A2).

We now introduce the density matrix $\rho(\mathbf{K})$ to describe the optically active excitonic spin states (therefore, a 2×2 matrix) and the exciton center-of-mass momentum. Its equation of motion is given by[34]

$$\frac{d\rho(\mathbf{K})}{dt} = \frac{i}{\hbar}\,[\rho(\mathbf{K}),\ H_0 + H_1(\mathbf{K})] + \sum_{\mathbf{K}'} W_{\mathbf{K}\mathbf{K}'}\,(\rho(\mathbf{K}') - \rho(\mathbf{K})) - \frac{1}{\tau}\rho(\mathbf{K}) + G\,, \tag{A3}$$

where $W_{\mathbf{K}\mathbf{K}'}$ represents the momentum scattering rate, G represents the generating rate of optically active excitons, and $\frac{1}{\tau}$ represents symbollically the recombination rate of those excitons which are able to recombine, that is, those with kinetic energy within the homogeneous linewidth Γ_h of $\mathbf{K} = \mathbf{0}$. By writing the density matrix in terms of its trace N and spin trace $\mathbf{S}$,

$$\rho(\mathbf{K}) = \frac{N}{2} + \mathbf{S}\cdot\boldsymbol{\sigma}, \tag{A4}$$

in terms of the Pauli spin matrices $\boldsymbol{\sigma}$, and the generating matrix G in a similar expression, we can express the equation of motion in the physically revealing form

$$\frac{d\mathbf{S}(\mathbf{K})}{dt} = \mathbf{\Omega}(\mathbf{K})\times\mathbf{S}(\mathbf{K}) + \sum_{\mathbf{K}'} W_{\mathbf{K}\mathbf{K}'}\,(\mathbf{S}(\mathbf{K}') - \mathbf{S}(\mathbf{K})) - \frac{1}{\tau}\mathbf{S}(\mathbf{K}) + \mathbf{G}\,, \tag{A5}$$

where $\mathbf{\Omega} = [\Omega_\parallel\cos(2\phi), \Omega_\parallel\sin(2\phi), \Omega_0]$ are the precession

frequencies due to the combined applied magnetic field along the z axis and the effective magnetic field due to the exchange spin flip in the interface plane.

We shall consider here the elastic-scattering problem in which the scattering rate W at a given momentum magnitude K depends only the angle between the initial and final momentum. The final answer has to be suitably averaged over K within the values given by the homogeneous linewidth. This simplifies considerably the solution of Eq. (A5) and still retains the essential physical picture. Then, the spin flip is due to an effective magnetic field with fixed amplitude but with a randomly varying direction in the xy plane.

Equation (A5) can then be solved exactly at each constant K by using the expansions

$$S_\alpha(\mathbf{K}) = \sum_n S_\alpha^n \, e^{in\phi} \tag{A6}$$

and

$$W(\phi - \phi') = \sum_n W_n \, e^{in(\phi-\phi')} \, . \tag{A7}$$

This yields an infinite set of first-order differential equations which couple $\mathbf{S}^m$ to $\mathbf{S}^{m\pm2}$.

The total longitudinal spin, S_z^0, at momentum K is governed by the equation

$$\frac{dS_z^0}{dt} = \frac{i\Omega_\parallel}{2} \, (S_-^{-2} - S_+^2) - \frac{S_z^0}{\tau} + G_z^0 \, , \tag{A8}$$

where $S_\pm = S_x \pm iS_y$. In turn,

$$\frac{dS_\pm^{\pm2}}{dt} = \pm \, i\Omega_0 \, S_\pm^{\pm2} \mp i\Omega_\parallel \, S_z^0 - \frac{S_\pm^{\pm2}}{\tau_2^*} \, , \tag{A9}$$

where we have defined

$$\frac{1}{\tau_m^*} = \frac{1}{\tau_m} + \frac{1}{\tau} \, , \tag{A10}$$

with the introduction of the momentum scattering times

$$\frac{1}{\tau_m} = (W_0 - W_m) = \int \frac{d\phi}{2\pi} \, W(\phi) \, [1 - \cos(m\phi)] \, . \tag{A11}$$

Equations (A8) and (A9) form a closed set of coupled equations which can be easily solved to yield the *longitudinal* spin-relaxation time T_{s1}:

$$\frac{1}{T_{s1}} = \Omega_\parallel^2 \, \frac{\tau_2^*}{1 + (\Omega_0 \tau_2^*)^2} \, . \tag{A12}$$

Since the depolarizing field $\Omega_\parallel$ is in the xy plane we are able to make a distintion between *longitudinal* and *transverse* spin-relaxation processes even at $\Omega_0 = 0$. To obtain the *transverse* spin-relaxation time T_{s2} we have to solve Eq. (A5) for $S_\pm^0$:

$$\frac{dS_+^0}{dt} = i\Omega_0 S_+^0 - i\Omega_\parallel \, S_z^{-2} - \frac{S_+^0}{\tau} + G_+^0 \, . \tag{A13}$$

The equation is coupled to the one for S_z^{-2}, which is in turn coupled to one for S_-^{-4}. These three equations formed a closed set. Solution for a single-frequency driving term leads to the transverse spin-relaxation time T_{s2} given by

$$\frac{1}{T_{s2}} = \mathrm{Re} \left[\frac{\Omega_\parallel^2/2}{\frac{1}{\tau_2^*} + \frac{\Omega_\parallel^2/2}{1/\tau_4^* + i\Omega_0}} \right] \, , \tag{A14}$$

where Re stands for the real part of the right-hand expression. The imaginary part gives the shift in precession frequency.

In the limiting case where $\Omega_\parallel^2 \ll (\tau_2^* \tau_4^*)^{-1}$, which is the case of interest for motional narrowing effects, the expression simplifies to

$$\frac{1}{T_{s2}} = \frac{1}{2} \, \Omega_\parallel^2 \, \tau_2^* \, . \tag{A15}$$

This is the usual relation $T_{s2} = 2T_{s1}$, when $\Omega_0 = 0$, already mentioned in Sec. IV B.

We finally note that results similar to Eqs. (A12) and (A15) could also be obtained via an explicit time-dependent perturbation theory,[13] in which τ^* is interpreted as the correlation time associated to the random field $\Omega_\parallel(t)$. The approach in this appendix (that is similar to the one in Ref. 34) gives a very precise definition of the momentum scattering time τ_2^*, Eq. (A11), which differs from the usual definition of momentum scattering time $\tau_\mathbf{K}$ for transport which has $\cos(\phi)$ term instead of the $\cos(2\phi)$ term in τ_2^*.

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