

## Coherent spin dynamics of excitons in quantum wells

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Coherent spin dynamics of excitons in quantum wells is investigated theoretically. Resonant and nonresonant excitation conditions are considered. The single-particle hole spin-flip mechanism within the exciton is always the dominant cause for breaking the exchange coupling. The two-dimensional confinement and low temperature are the most important factors which stabilize the hole spin orientation within the exciton. The experimental observations reported previously are interpreted. [S0163-1829(97)06235-8]

### I. INTRODUCTION

When two energetically closely spaced transitions are excited with a short optical pulse, the two induced polarizations in the medium oscillate with their slightly different frequencies. Their interference manifests in a modulation of the net polarization, the so-called quantum beats (QB's). This paper is devoted to the interpretation of QB's observed recently in the time-resolved free-exciton photoluminescence (PL) in type-I quantum wells<sup>1-3</sup> (QW's) on a time scale of a few hundreds of picoseconds, in a magnetic field perpendicular to the growth axis.

Modulation of the free-exciton luminescence components was discovered by Heberle *et al.*<sup>1</sup> The authors interpreted the QB in terms of the Larmor precession of electron spins in a magnetic field perpendicular to the growth axis. The corresponding pulsation  $\omega$  directly reveals the *electron spin splitting*  $\hbar\omega = g_e\mu_B B$ .

In a recent paper<sup>3</sup> we reported on investigations in relatively narrow QW's when the excitation is resonant with the heavy-hole exciton (XH). It was demonstrated that QB's then reflect the *exciton spin splitting* in the transverse magnetic field,  $\hbar\Omega = \sqrt{(\hbar\omega)^2 + \delta^2}$  where  $\delta$  is the electron-hole exchange energy which splits the XH-1s quadruplet into the radiative and nonradiative pair of states at  $B=0$ . It was shown moreover that QB's at the pulsation  $\omega$ , as Heberle observed, could be recorded from the same well with nonresonant excitation.

We suggested in Ref. 3 that the manifestation of QB's on the excitonic luminescence at the electronic or excitonic pulsation ( $\omega$  or  $\Omega$ , respectively) is related to the stability of the hole-spin orientation within the exciton. The argument was the following. Within the exciton, the correlation between electron and hole spins is held by the electron-hole exchange interaction. However, if this correlation is not strong enough to reduce the single-particle hole spin flip at a rate lower than  $\delta/\hbar$ , the exchange interaction splitting  $\delta$  no longer plays a role in the QB. Then the QB appears at the pulsation  $\omega$ . Finally an electron bound into an exciton precesses like a free electron in the transverse magnetic field provided that  $\tau \ll \hbar/\delta$  where  $\tau$  is the single-particle hole spin-flip time. This condition can be fulfilled in large and narrow QW's but for different reasons.

In large QW's (*a fortiori* in the bulk) such a hole spin flip occurs as a consequence of the mixing of states in the valence band; the observation of the electron precession in a QW of 25 nm well width *under the resonant or nonresonant excitation* conditions reported in Ref. 1 is understood on this ground.

In narrow QW's the hole spin flip, which results in the observation of QB's at the pulsation  $\omega$  *in nonresonant excitation*, is related to the formation-dissociation process of excitons and the related long cooling of the excited system.<sup>4</sup>

QB's of the excitonic kind have been observed only in narrow quantum wells ( $L_w \leq 10$  nm) *under resonant excitation*. This is the indication that the hole-spin orientation is stable in cold two-dimensional (2D) excitons ( $\tau > \hbar/\delta$ ). The experiments reported in Ref. 3 showed moreover that a shift of the excitation from the resonance position as small as +1 meV results in an intermediate situation, between pure excitonic QB (pulsation  $\Omega$ ) and pure electronic QB (pulsation  $\omega$ ) manifestations. It was interpreted as the indication that the stability of the hole-spin orientation within 2D excitons is strongly dependent on the exciton temperature, decreasing quickly when this temperature increases.

In this paper we propose a quantitative theory of the QB in exciton luminescence. Section II is devoted to the interpretation of experiments in which excitons are photogenerated directly by the laser beam tuned at the XH position. Electronic or excitonic QB, at pulsation  $\omega$  or  $\Omega$ , respectively, are indeed predicted depending whether  $\tau \ll \hbar/\delta$  or  $\tau \gg \hbar/\delta$ . The general features observed experimentally, including an acceleration of the luminescence depolarization in the conditions of observation of excitonic QB's, are qualitatively described by a simple theory in which the only relaxation mechanism considered is the hole-spin relaxation. However we cannot give a quantitative description without taking into account other relaxation processes. The most important of the processes left aside is certainly the *exciton spin-flip mechanism*, i.e., the spin flip of the exciton as a whole which corresponds to the simultaneous spin flip of the electron and the hole, a mechanism of the kind described in Refs. 5 and 6. This process is known to determine the depolarization at zero field and contribute to the damping of the exciton QB in the magnetic field. Unfortunately we do not actually know how to introduce the exciton spin-flip mechanism in the theory.

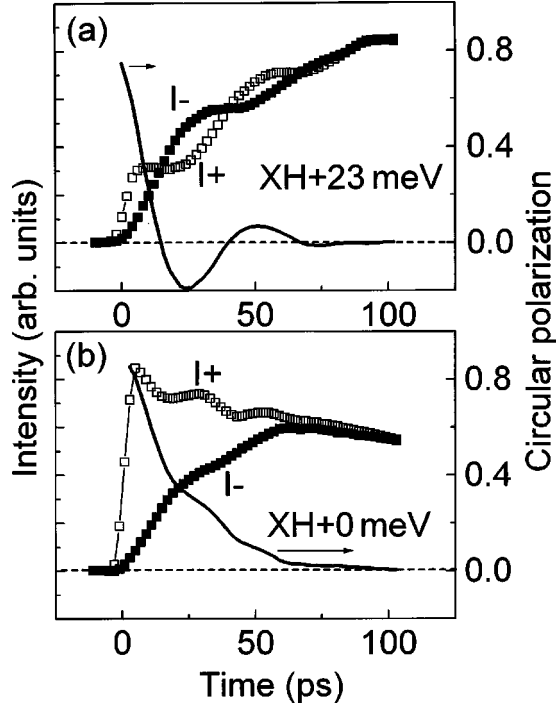


FIG. 1. Sample I: the luminescence intensities  $I^+$  and  $I^-$  and the circular polarization  $P_L$  after the  $\sigma^+$ -polarized excitation, at  $B=2.8$  T. (a) The excitation energy is nonresonant ( $E_1 - HH_1 < h\nu < XL$ , where  $E_1 - HH_1$  is the QW gap and  $XL$  the light hole spectral positions), (b) the excitation energy is resonant with XH.

Section III is devoted to the interpretation of nonresonant excitation experiments, when the laser beam generates free-electron-hole pairs (above bandgap excitation). In such conditions, excitons form by random binding of electrons and holes. We present the theory of the resulting excitonic luminescence in transverse magnetic field which displays QB's at the electron Larmor frequency  $\omega$ . The effect of the hole spin flip within the formed excitons is investigated. The additional implication of hot exciton redissociation during the decay is also examined. For realistic values of parameters we find that the formation and/or dissociation cannot explain alone the electronic nature of the QB. We conclude that the hole spin flip within the formed excitons is always the dominant reason for breaking the exchange coupling. The high hole spin-flip rate in nonresonant excitation experiments is related to the temperature of the hot exciton system which remains well above the lattice temperature during the whole depolarization phase.

It is useful first to recall briefly the important experimental features. Figure 1 illustrates the observations in a GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As QW of 3 nm well width (sample I). In nonresonant excitation conditions [Fig. 1(a)], oscillations on the two circularly polarized luminescence components  $I^+$  and  $I^-$  at the pulsation frequency  $\omega$  are phase shifted by  $\pi$  (electronic QB). Under resonant excitation [Fig. 1(b)], oscillations at the pulsation  $\Omega$  are visible on the  $I^+$  component only (excitonic QB). Similar effects have been observed on another GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As narrow QW sample, of 4.8 nm well width (sample II). Details can be found in Ref. 3. We mention two additional observations which shall be useful for the comparison between theory and experiment.

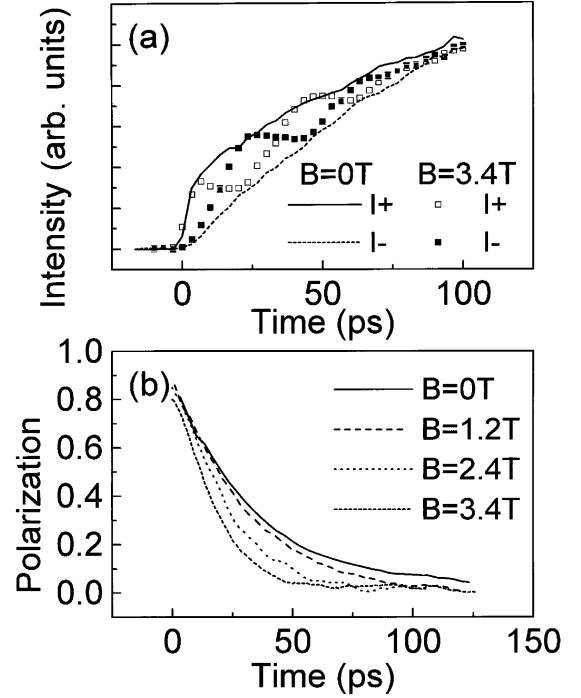


FIG. 2. The excitation is  $\sigma^+$  polarized. (a) Sample I. The excitation is nonresonant ( $E_1 - HH_1 < h\nu < XL$ ). The luminescence intensities  $I^+$  and  $I^-$  at  $B=3.4$  T (dots) and  $B=0$  T (lines). Note that the amplitude of the oscillations in the presence of the magnetic field is equal to  $I^+ - I^-$  at zero field. (b) Sample II. The excitation is resonant with XH. Illustration of the acceleration of the depolarization by the transverse magnetic field.

(1) Under nonresonant excitation, the amplitude of the oscillations of  $I^+$  and  $I^-$  in the magnetic field is equal to  $I^+ - I^-$  at zero field: this observation, which has been previously reported by Heberle *et al.*,<sup>1</sup> is illustrated in Fig. 2(a).

(2) Under resonant excitation, the magnetic field induces an acceleration of the depolarization: this effect is shown in Fig. 2(b).

## II. RESONANT PHOTOGENERATION OF EXCITONS

### A. The spin precession of coherently photogenerated excitons: Theory of the luminescence signal

For a (001)-grown QW, the conduction band is  $S$ -like with two spin states  $S_z = \pm 1/2$ . The valence band is split into a heavy-hole band with the total angular-momentum projection  $J_{h,z} = \pm 3/2$  and a light-hole band with  $J_{l,z} = \pm 1/2$ . The heavy-hole exciton (XH) states are described using the basis set  $\{|J_z + S_z\rangle\}$  where  $J_z \equiv J_{h,z}$ . In the conditions of 2D confinement, the transverse  $g$  factor of the hole is zero. The spin Hamiltonian of the XH exciton in the transverse magnetic field ( $\mathbf{B} \parallel \mathbf{Ox}$ ) is

$$\mathcal{H} = \hbar \omega \cdot \mathbf{S} - \frac{2\delta}{3} J_z S_z, \quad (1)$$

where  $\hbar \omega = g \mu_B \mathbf{B}$  and  $\delta$  is the zero-field exchange splitting between the optically active doublet  $|\pm 1\rangle$  and the two close-lying singlets of the  $\{|\pm 2\rangle\}$  subspace (the much smaller splitting between the singlets  $|\pm 2\rangle$  is neglected<sup>7</sup>). The equation of motion for any time independent observable  $\mathcal{O}$  is

$$\frac{d\mathcal{O}}{dt} = \frac{1}{i\hbar} [\mathcal{O}, \mathcal{H}].$$

Applying it to  $\mathbf{S}$ ,  $J_z\mathbf{S}$ , and  $J_z$  successively, we obtain

$$\frac{d\mathbf{S}}{dt} = \boldsymbol{\omega} \wedge \mathbf{S} - \frac{2}{3} \tilde{\delta} \mathbf{n} \wedge J_z \mathbf{S}, \quad (2a)$$

$$\frac{d(J_z \mathbf{S})}{dt} = \boldsymbol{\omega} \wedge (J_z \mathbf{S}) - \frac{3}{2} \tilde{\delta} \mathbf{n} \wedge \mathbf{S}, \quad (2b)$$

$$\frac{dJ_z}{dt} = 0, \quad (2c)$$

where  $\tilde{\delta} = \delta/\hbar$  and  $\mathbf{n} = (0, 0, 1)$  is a unit vector normal to the 2D plane of the QW. In the following, we denote  $\mathbf{Q} = (2/3)J_z\mathbf{S}$  and take the quantum-mechanical average of Eqs. (2). From now on we will write  $\mathbf{S}$  for  $\langle \mathbf{S} \rangle$ ,  $\mathbf{Q}$  for  $\langle \mathbf{Q} \rangle$ , and  $J_z$  for  $\langle J_z \rangle$ . Note that  $\mathbf{Q}$  describes the correlation between  $J_z$  and  $\mathbf{S}$  within the exciton. Equations (2) take the form

$$\frac{d\mathbf{S}}{dt} = \boldsymbol{\omega} \wedge \mathbf{S} - \tilde{\delta} \mathbf{n} \wedge \mathbf{Q}, \quad (3a)$$

$$\frac{d\mathbf{Q}}{dt} = \boldsymbol{\omega} \wedge \mathbf{Q} - \tilde{\delta} \mathbf{n} \wedge \mathbf{S} - \frac{\mathbf{Q}}{\tau}, \quad (3b)$$

$$\frac{dJ_z}{dt} = -\frac{J_z}{\tau}. \quad (3c)$$

Extra terms  $-\mathbf{Q}/\tau$  and  $-J_z/\tau$  have been added in the equations ‘‘by hand’’ in order to take into account any specific hole spin-relaxation mechanism, represented by a phenomenological relaxation time  $\tau$ . Equations (3) govern the evolution of the mean values of the observables in the excitonic population (statistical average). The present formulation is, of course, in agreement with the standard density-matrix formalism (see the Appendix).

Projection of Eqs. (3) on the Cartesian axes leads to the three independent sets of equations:

$$\begin{aligned} (a) \quad & \begin{cases} \dot{S}_z = \omega S_y, \\ \dot{Q}_x = \tilde{\delta} S_y - \frac{Q_x}{\tau}, \\ \dot{S}_y = -\tilde{\delta} Q_x - \omega S_z, \end{cases} \\ (b) \quad & \begin{cases} \dot{Q}_z = \omega Q_y - \frac{Q_z}{\tau}, \\ \dot{S}_x = \tilde{\delta} Q_y, \\ \dot{Q}_y = -\tilde{\delta} S_x - \omega Q_z - \frac{Q_y}{\tau}, \end{cases} \\ (c) \quad & \dot{J}_z = -\frac{J_z}{\tau}, \end{aligned} \quad (4)$$

where  $\dot{S}_z \equiv dS_z/dt$ , etc.

The luminescence intensity components of right and left helicity correspond to the mean value of the operator  $|\pm 1\rangle\langle \pm 1|$ , respectively. They take the form

$$I^\pm = \frac{2}{9} \left\langle \left( \frac{3}{4} - J_z S_z \right) \left( \frac{3}{2} \pm J_z \right) \right\rangle. \quad (5)$$

The interpretation is straightforward:  $2/9$  is the normalization factor;  $3/4 - J_z S_z$  is the factor which ensures that  $I^\pm = 0$  when nonoptically active states are concerned, i.e.,  $(S_z, J_z) = (\pm 1/2, \pm 3/2)$ ;  $3/2 \pm J_z$  is the factor which ensures that  $I^\pm = 0$  when  $(S_z, J_z) = (\pm 1/2, \mp 3/2)$ . Equations (5) may be written in the form

$$I^\pm = \frac{1}{2} \left( \frac{1}{2} - Q_z \right) \pm \frac{1}{2} \left( \frac{1}{3} J_z - S_z \right). \quad (6)$$

The total luminescence intensity ( $I$ ) and polarization ( $P$ ) are

$$I = I^+ + I^- = \frac{1}{2} - Q_z, \quad (7a)$$

$$P = \frac{I^+ - I^-}{I^+ + I^-} = \frac{J_z/3 - S_z}{1/2 - Q_z}. \quad (7b)$$

Initial conditions depend on the photogeneration conditions. With a  $\sigma^+$ -polarized short pulse at  $t=0$  they are

$$\begin{aligned} S_x(0) &= 0, & S_y(0) &= 0, & S_z(0) &= -1/2, \\ Q_x(0) &= 0, & Q_y(0) &= 0, & Q_z(0) &= -1/2, \\ J_z(0) &= 3/2. \end{aligned} \quad (8)$$

Note that  $I^+(0) = 1$  and  $I^-(0) = 0$ .

Specific equations for  $S_z, Q_z, J_z$  which determine  $I$  and  $P$  are derived from Eq. (4):

$$\tau \dot{S}_z + \ddot{S}_z + \tau \Omega^2 \dot{S}_z + \omega^2 S_z = 0, \quad (9a)$$

$$\tau^2 \ddot{Q}_z + 2\tau \ddot{Q}_z + (\Omega^2 \tau^2 + 1) \dot{Q}_z + \tau \tilde{\delta}^2 Q_z = 0, \quad (9b)$$

$$\tau \dot{J}_z + J_z = 0, \quad (9c)$$

where  $\Omega = \sqrt{\omega^2 + \tilde{\delta}^2}$ .

The following approximate solutions of the third-order differential equations (9a) and (9b) can be found easily for low-, intermediate-, and high-hole spin-flip rates (we introduce  $\tau^* = \tau \tilde{\delta}^2 / \omega^2$  and note that  $\tau^* \gg \tau$ ):

$$\begin{aligned} 1/\tau \ll \tilde{\delta} \quad & \begin{cases} S_z(t) = -\frac{1}{2} \left[ 1 - \frac{\omega^2}{\Omega^2} (1 - \cos \Omega t) \right], \\ Q_z(t) = -\frac{1}{2} \left[ 1 - \frac{\omega^2}{\Omega^2} (1 - \cos \Omega t) \right], \end{cases} \\ \tilde{\delta} \ll 1/\tau \ll \tilde{\delta}^2/\omega \quad & \begin{cases} S_z(t) = -\frac{1}{2} e^{-t/\tau^*}, \\ Q_z(t) = 0, \end{cases} \\ \text{and } t \gg \tau \quad & \\ \omega, \tilde{\delta}, \tilde{\delta}^2/\omega \ll 1/\tau \quad & \begin{cases} S_z(t) = -\frac{1}{2} \cos \omega t, \\ Q_z(t) = 0. \end{cases} \end{aligned} \quad (10)$$

When  $1/\tau \ll \tilde{\delta}$ , Eqs. (9a) and (9b) reduce to  $\dot{S}_z + \Omega^2 S_z = 0$  and  $\ddot{Q}_z + \Omega^2 \dot{Q}_z = 0$ , respectively. When  $\tilde{\delta} \ll 1/\tau \ll \tilde{\delta}^2/\omega$ , Eq. (9a) reduces to  $\tau \Omega^2 \dot{S}_z + \omega^2 S_z = 0$  or  $\tau \tilde{\delta}^2 \dot{S}_z + \omega^2 S_z = 0$ . When  $\omega, \tilde{\delta} \ll 1/\tau$ , Eq. (9a) reduces to  $\ddot{S}_z + \omega^2 S_z = 0$ .

The corresponding luminescence components  $I^\pm$  are then derived from Eq. (7):

$$\begin{aligned}
& \text{(a)} \quad 1/\tau \ll \tilde{\delta} \quad \begin{cases} I^+(t) = 1 - \frac{\omega^2}{\Omega^2} \frac{1 - \cos \Omega t}{2}, \\ I^-(t) = 0, \end{cases} \quad \begin{array}{l} \text{excitonic} \\ \text{oscillations} \end{array} \\
& \text{(b)} \quad \dots \quad \dots, \\
& \text{(c)} \quad \begin{array}{l} \tilde{\delta} \ll 1/\tau \ll \tilde{\delta}^2/\omega \\ \text{and } t \gg \tau \end{array} \quad \begin{cases} I^+(t) = \frac{1 + \exp(-t/\tau^*)}{4}, \\ I^-(t) = \frac{1 - \exp(-t/\tau^*)}{4}, \end{cases} \quad \begin{array}{l} \text{damped regime} \\ [\tau^* = \tau \delta^2 / (\hbar \omega)^2] \end{array} \\
& \text{(d)} \quad \dots \quad \dots, \\
& \text{(e)} \quad \begin{array}{l} \omega, \tilde{\delta}, \tilde{\delta}^2/\omega \ll 1/\tau \\ \text{and } t \gg \tau \end{array} \quad \begin{cases} I^+(t) = \frac{1 + \cos \omega t}{4}, \\ I^-(t) = \frac{1 - \cos \omega t}{4}, \end{cases} \quad \begin{array}{l} \text{electronic} \\ \text{oscillations.} \end{array} \quad (11)
\end{aligned}$$

Note that, in regime (c), the depolarization rate described by the effective relaxation time  $\tau^*$  increases with the magnetic field as  $B^2$ .

When the condition  $1/\tau \ll \tilde{\delta}$  is fulfilled, Eqs. (11) predict the appearance of excitonic QB's on the  $I^+$  component of the luminescence, as the magnetic field increases. When the opposite condition is fulfilled ( $\tilde{\delta} \ll 1/\tau$ ), Eqs. (11) predict the appearance of electronic QB on the two luminescence components, with a phase shift of  $\pi$  between them, as the magnetic field increases.

More generally, the luminescence components are obtained from the numerical solution of Eqs. (4) and (6). Figures 3(a)–3(e) illustrate the results for the specific ranges of parameters distinguished above [(a)–(e), Eq. (11)]. In the excitonic QB regime [Figs. 3(a) and 3(b)], modulations at the frequency  $\Omega$  are visible on the component  $I^+$  only. In the electronic QB regime [Figs. 3(d) and 3(e)] the two luminescence components oscillate with a phase shift of  $\pi$  between them. These conclusions are in agreement with the experiments.

### B. The interpretation of resonant excitation experiments

The present theory, which puts forward the role of the hole spin-flip process in the manifestation of electronic or excitonic spin QB, supports the interpretation of *resonant excitation experiments* given previously<sup>3</sup> and summarized in the Introduction. In QW's, the electron-hole exchange interaction within the exciton increases rapidly with the confinement, and simultaneously the mixing between the light- and heavy-hole states in the valence band decreases. This results in the decrease of the single-particle hole-spin-relaxation rate. As a consequence, exciton QB's are visible in narrow enough QW's, where the exchange splitting becomes larger than the lifetime broadening of the exciton spin state due to the single-particle hole spin-flip process ( $1/\tau \ll \delta/\hbar$ ). In large quantum wells ( $\geq 10$  nm) the electron-hole exchange is not strong enough to counterbalance the effect of spin mixing in the valence band. As a consequence, the condition for the manifestation of the electronic QB's is fulfilled ( $1/\tau \gg \delta/\hbar$ ).

Figures 3(a')–3(e') show the decisive role of the hole spin flip on the luminescence depolarization in the presence of a transverse magnetic field. In the intermediate range [Figs. 3(b')–3(d')], the hole spin flip is the cause of an acceleration of the depolarization by the field [this effect is explicit in Eq. (11c)]. This acceleration is observed experimentally [Fig. 2(b)]. On the other hand, the simulation confirms the theoretical prediction that the magnetic field has no damping effect in the extreme cases,  $1/\tau \ll \delta/\hbar$  or  $1/\tau \gg \delta/\hbar$ . We will comment on that point in Sec. III.

The *excitonic spin-flip mechanism*, which corresponds to the simultaneous spin flip of the electron and the hole within the exciton, is ignored in the present theory. This mechanism however has been recognized as the relevant depolarizing mechanism of 2D excitons when  $B=0$ .<sup>5,7,8</sup> It has been shown that it affects free excitons<sup>5</sup> and bound excitons as well.<sup>9</sup> Nevertheless, the free-exciton spin-flip theory has not been currently extended to the case of excitons in a transverse magnetic field. Due to this lack of a relevant theory, we cannot propose a full fit of the experimental data. It is indeed possible to fit the luminescence depolarization curves if we arbitrarily damp the calculated luminescence polarization by the factor  $\exp(-t/\tau_x)$  which is needed to fit the  $B=0$  data ( $\tau_x$  is the exciton *spin-flip time*). Good fits of the polarization dynamics including the acceleration of the depolarization by the magnetic field are obtained with  $\tau=14$  ps, a value which satisfies the condition  $1/\tau < \delta/\hbar$ . However, this relatively fast single-particle hole spin-flip time involves a fast initial decay of the total luminescence due to the transfer of the exciton to the optically nonactive states ( $J_z=2$ ). This is never observed in the experiments. This is the proof that the hole spin-flip time is really longer and probably indicates that the excitonic spin-flip mechanism has to be introduced rigorously in the theory, a task beyond the scope of this paper.

Finally, numerical solutions of Eqs. (4) and (6) lead us to the conclusion that the spin oscillations in a transverse magnetic field are a general phenomenon observable in the excitonic luminescence as long as the field intensity is high enough. The luminescence oscillations are of excitonic or electronic kind for  $1/\tau < \delta/\hbar$  or  $1/\tau > \delta/\hbar$ , respectively.

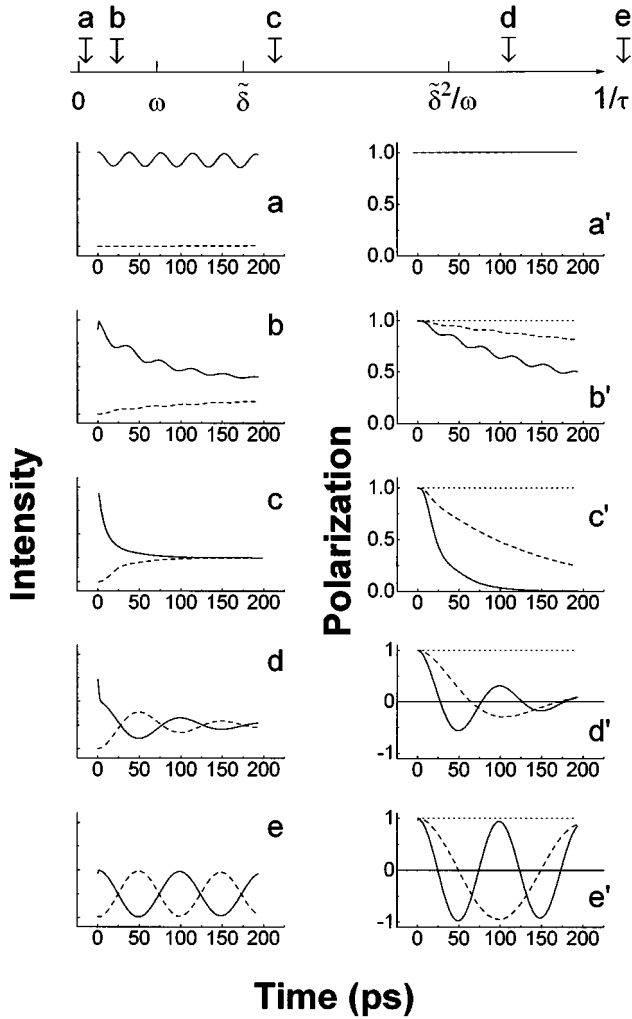


FIG. 3. Simulation of resonant excitation experiments with the sample II parameters:  $g_e = 0.24$ ,  $\delta = 0.10$  meV. (a)–(e) The luminescence intensity dynamics  $I^+$  (solid line) and  $I^-$  (dashed line) in the transverse magnetic field, from the numerical solution of Eqs. (4) and (6). The different ranges of the hole spin-flip rate  $1/\tau$  listed in Eqs. (11) are illustrated at  $B = 3$  T. (a')–(e') The polarization decay for the same ranges of  $1/\tau$ , from the numerical solution of Eqs. (4) and (7b) at  $B = 0$  (dots),  $B = 1.5$  (dashed line),  $B = 3$  T (solid line).  $\tau = 2000$  ps in (a) and (a'),  $\tau = 50$  ps in (b) and (b'),  $\tau = 6$  ps in (c) and (c'),  $\tau = 1$  ps in (d) and (d'),  $\tau = 0.05$  ps in (e) and (e').

### III. BIMOLECULAR FORMATION OF EXCITONS

The observation of electronic QB's is the general rule for the experiments performed with excitation energies above the QW gap. But the most intriguing question is the following: why does the narrow QW, which exhibits excitonic QB's when the excitation energy is resonant with XH, show electronic QB's when the excitation energy is tuned above the band gap?

When the laser excitation energy is above the band gap ( $HH_1 - E_1$ ), excitons are not directly formed by the laser pulse. It has been demonstrated previously that, in such photogeneration conditions, (i) the holes are immediately depolarized (from Ref. 10,  $\tau < 4$  ps at low sample temperature) and (ii) the excitons form by random binding of the generated free electrons and holes.<sup>11</sup> The exciton formation time

has never been measured directly but was deduced indirectly by various ways. Damen *et al.* conclude from their analysis of the rapid decay of the homogeneous exciton linewidth that the exciton formation time is less than 20 ps for an excitation density of  $2 \times 10^{10}$  cm<sup>-2</sup>.<sup>12</sup> The result we obtained in Ref. 4 is not very different although our analysis was based on a modified interpretation of the decay of the homogeneous linewidth: we found less than 10 ps for the density range  $10^9 - 10^{10}$  cm<sup>-2</sup>. Much longer formation times are reported by Deveaud *et al.*<sup>13</sup> ( $\sim 200$  ps for densities of the order of  $10^{11}$  cm<sup>-2</sup>) and more recently by Kumar *et al.*<sup>14</sup> (50 ps at the density of  $4 \times 10^{10}$  cm<sup>-2</sup>).

Another important question concerns the stability of the exciton formed by the binding of an electron-hole pair. We observed previously that the rapid initial decay of the exciton homogeneous luminescence linewidth is consistent with the assumption of a dynamical thermal equilibrium between dissociated  $e-h$  pairs and the excitons.<sup>4</sup> This implies that excitons form and ionize at a very high rate, the time dependence of the different populations arising mainly due to the slow cooling of the electronic system ( $> 100$  ps) by acoustic phonon emission. On the contrary, Kumar *et al.*<sup>14</sup> considered that excitons formed by the random binding process at low crystal temperature are definitively stable, i.e., cannot ionize again.

The question of hole-spin stability within the excitons formed by the bimolecular process is fundamental in the prospect of the interpretation of electronic QB's always observed in nonresonant excitation conditions. Moreover, the two frames of formation just recalled, i.e. (i) binding without redissociation, or (ii) multiple binding and/or dissociation during the decay, have to be considered separately. This will be done below. We present in Sec. III A the general theory of spin precession in a population of excitons formed by random binding of electron-hole pairs, after the photogeneration of a free-electron-hole gas. We shall consider the specific frames (i) and (ii) in Secs. III B and III C, respectively.

#### A. The spin precession in a population of excitons formed by random binding of $e-h$ pairs: Theory of the luminescence signal

The laser pulse creates  $n_0$  electron-hole pairs at  $t = 0$ . We denote by  $n(t)$  the density of free  $e-h$  pairs and by  $N(t)$  the exciton density at time  $t > 0$ . It is assumed that the photogenerated free holes are depolarized quasi-instantaneously. Excitons form by random binding of  $e-h$  pairs at a rate  $\gamma n^2(t)$  and redissociate at a rate  $\alpha N(t)$  where  $\gamma$  and  $\alpha$  are the bimolecular formation and dissociation coefficients, respectively. We introduce the following notations:  $\mathbf{s}$  for the average spin of free electrons and  $\boldsymbol{\sigma} = n(t)\mathbf{s}$  for the electron spin density within the free-electron-hole system;  $\mathbf{S}$  for the average spin of electrons bounded into excitons and  $\boldsymbol{\Sigma} = N(t)\mathbf{S}$  for the electron spin density within the exciton system;  $\mathbf{Q}$  for the average correlation between electron and hole spin orientations within excitons and  $\boldsymbol{\mathcal{Q}} = N(t)\mathbf{Q}$  for the "correlation density" within the exciton system;  $\mathbf{J}$  for the average spin of holes bounded into excitons and  $\boldsymbol{\mathcal{J}} = N(t)\mathbf{J}$  for the hole spin density within the exciton system. The rates of change of  $\boldsymbol{\sigma}$  and  $\boldsymbol{\Sigma}$  due to exciton formation and dissociation express as

$$\left. \frac{\partial \boldsymbol{\sigma}}{\partial t} \right|_{\text{form.}} = - \left. \frac{\partial \boldsymbol{\Sigma}}{\partial t} \right|_{\text{form.}} = - \gamma n \boldsymbol{\sigma} + \alpha \boldsymbol{\Sigma}. \quad (12a)$$

This equation expresses the conservation of the average electron spin in the formation and dissociation processes. Since free holes are depolarized, the rate of change of  $\mathcal{J}_z$  and  $\mathcal{Q}$  comes entirely from dissociations:

$$\left. \frac{\partial \mathcal{Q}}{\partial t} \right|_{\text{form.}} = - \alpha \mathcal{Q}, \quad \left. \frac{\partial \mathcal{J}_z}{\partial t} \right|_{\text{form.}} = - \alpha \mathcal{J}_z. \quad (12b)$$

The components  $\mathcal{I}^\pm = N(t)I^\pm$  of the excitonic luminescence may be expressed by equations derived from Eq. (7):

$$\mathcal{I}^\pm(t) = \frac{1}{2} \left[ \left( \frac{N}{2} - \mathcal{Q}_z \right) \pm \left( \frac{\mathcal{J}_z}{3} - \Sigma_z \right) \right]. \quad (13)$$

The kinetic equations for  $n(t)$  and  $N(t)$  are

$$\frac{dn}{dt} = - \gamma n^2 + \alpha N, \quad (14a)$$

$$\begin{aligned} \frac{dN}{dt} &= \gamma n^2 - \alpha N - \frac{I^+ + I^-}{\tau_{\text{rad}}}, \\ &= \gamma n^2 - \alpha N - \frac{N/2 - \mathcal{Q}_z}{\tau_{\text{rad}}}, \end{aligned} \quad (14b)$$

where  $\tau_{\text{rad}}$  is the radiative lifetime of an optically active exciton. Neglecting direct recombination of free-electron-hole pairs, the rates of change of  $\boldsymbol{\sigma}$ ,  $\boldsymbol{\Sigma}$ , and  $\mathcal{Q}$  due to radiative recombination are given by [the Appendix, Eq. (A7)]

$$\left. \frac{\partial \boldsymbol{\sigma}}{\partial t} \right|_{\text{rad}} = 0, \quad (15a)$$

$$\left. \frac{\partial \boldsymbol{\Sigma}}{\partial t} \right|_{\text{rad}} = - \frac{\boldsymbol{\Sigma} - \mathcal{J}_z \mathbf{n}/3}{2 \tau_{\text{rad}}}, \quad (15b)$$

$$\left. \frac{\partial \mathcal{Q}}{\partial t} \right|_{\text{rad}} = - \frac{\mathcal{Q} - N \mathbf{n}/2}{2 \tau_{\text{rad}}}, \quad (15c)$$

$$\left. \frac{\partial \mathcal{J}_z}{\partial t} \right|_{\text{rad}} = - \frac{\mathcal{J}_z - 3 \Sigma_z}{2 \tau_{\text{rad}}}. \quad (15d)$$

The evolution of the coupled system of free-electron-hole pairs and excitons is now described by the following set of equations [extension of Eq. (3)]:

$$\begin{aligned} \frac{d\boldsymbol{\sigma}}{dt} &= \boldsymbol{\omega} \wedge \boldsymbol{\sigma} + \left. \frac{\partial \boldsymbol{\sigma}}{\partial t} \right|_{\text{form.}} + \left. \frac{\partial \boldsymbol{\sigma}}{\partial t} \right|_{\text{rad}}, \\ \frac{d\boldsymbol{\Sigma}}{dt} &= \boldsymbol{\omega} \wedge \boldsymbol{\Sigma} - \tilde{\delta} \mathbf{n} \wedge \mathcal{Q} + \left. \frac{\partial \boldsymbol{\Sigma}}{\partial t} \right|_{\text{form.}} + \left. \frac{\partial \boldsymbol{\Sigma}}{\partial t} \right|_{\text{rad}}, \\ \frac{d\mathcal{Q}}{dt} &= \boldsymbol{\omega} \wedge \mathcal{Q} - \tilde{\delta} \mathbf{n} \wedge \boldsymbol{\Sigma} - \frac{\mathcal{Q}}{\tau} + \left. \frac{\partial \mathcal{Q}}{\partial t} \right|_{\text{form.}} + \left. \frac{\partial \mathcal{Q}}{\partial t} \right|_{\text{rad}}, \\ \frac{d\mathcal{J}_z}{dt} &= - \frac{\mathcal{J}_z}{\tau} + \left. \frac{\partial \mathcal{J}_z}{\partial t} \right|_{\text{form.}} + \left. \frac{\partial \mathcal{J}_z}{\partial t} \right|_{\text{rad}}. \end{aligned} \quad (16)$$

These equations are projected on the three Cartesian axes:

$$\dot{\sigma}_z = \omega \sigma_y - \gamma n \sigma_z + \alpha \Sigma_z,$$

$$\dot{\sigma}_y = - \omega \sigma_z - \gamma n \sigma_y + \alpha \Sigma_y,$$

$$\dot{\sigma}_x = - \gamma n \sigma_x + \alpha \Sigma_x, \quad (17a)$$

$$\dot{\Sigma}_z = \omega \Sigma_y + \gamma n \sigma_z - \alpha \Sigma_z - \frac{\Sigma_z - \mathcal{J}_z/3}{2 \tau_{\text{rad}}},$$

$$\dot{\mathcal{Q}}_x = \tilde{\delta} \Sigma_y - \frac{\mathcal{Q}_x}{\tau} - \alpha \mathcal{Q}_x - \frac{\mathcal{Q}_x}{2 \tau_{\text{rad}}},$$

$$\dot{\Sigma}_y = - \tilde{\delta} \mathcal{Q}_x - \omega \Sigma_z + \gamma n \sigma_y - \alpha \Sigma_y - \frac{\Sigma_y}{2 \tau_{\text{rad}}},$$

$$\dot{\mathcal{Q}}_z = \omega \mathcal{Q}_y - \frac{\mathcal{Q}_z}{\tau} - \alpha \mathcal{Q}_z - \frac{\mathcal{Q}_z - N/2}{2 \tau_{\text{rad}}},$$

$$\dot{\Sigma}_x = \tilde{\delta} \mathcal{Q}_y + \gamma n \sigma_x - \alpha \Sigma_x - \frac{\Sigma_x}{2 \tau_{\text{rad}}},$$

$$\dot{\mathcal{Q}}_y = - \tilde{\delta} \Sigma_x - \omega \mathcal{Q}_z - \frac{\mathcal{Q}_y}{\tau} - \alpha \mathcal{Q}_y - \frac{\mathcal{Q}_y}{2 \tau_{\text{rad}}},$$

$$\dot{\mathcal{J}}_z = - \frac{\mathcal{J}_z}{\tau} - \alpha \mathcal{J}_z - \frac{\mathcal{J}_z - 3 \Sigma_z}{2 \tau_{\text{rad}}}. \quad (17b)$$

Equations (14a), (14b), and (17) are integrated numerically and the luminescence components are derived, according to Eq. (13). In Sec. III B exciton redissociation is not considered ( $\alpha=0$ ). Exciton redissociation will be considered in the frame of the dynamical equilibrium concept in Sec. III C.

## B. The exciton redissociation is neglected ( $\alpha=0$ )

The two cases where the hole-spin orientation is stable (i) or unstable (ii) within the formed excitons are examined separately.

### 1. The hole-spin orientation is locked at the moment of exciton formation

Equations (17) are solved taking  $\alpha=0$  and  $1/\tau=0$ . The result depends on the initial value of the exciton formation time  $1/\gamma n_0$ . Four representative values are considered in Fig. 4. The temporal evolution of the luminescence is dominated by the competition between exciton formation and exciton recombination. The formation rate is higher than the recombination rate during the intensity rise and lower during the decay. As a consequence, the increase of the photogenerated density (or a higher formation coefficient  $\gamma$ ) results in the decrease of the rise time.

The simulation illustrates the two consequences of the coupling of the electron spin with the transverse magnetic fields.

(1) The small oscillations of the components  $I^+$  and  $I^-$  (Fig. 4): with a phase shift of  $\pi$  between them, these oscillations are superimposed on the luminescence evolution just described. The general trend is that their pulsation is close to  $\omega$  when the rise time of the luminescence is long (long exciton formation time  $1/\gamma n_0 > 1/\omega$ ) and close to  $\Omega$  when the

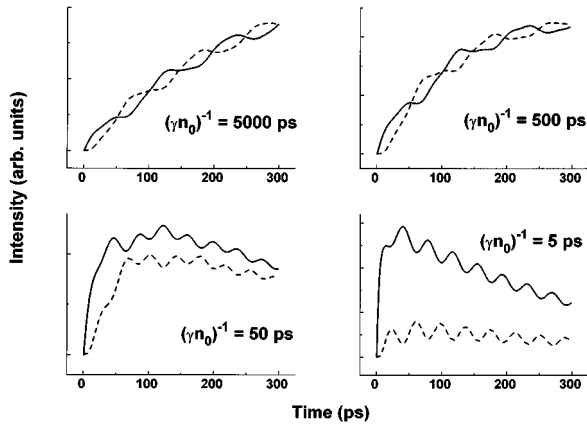


FIG. 4. Simulation of nonresonant excitation experiments with the parameters of sample II ( $g_e = 0.24$ ,  $\delta = 0.10$  meV). The excitons form by the bimolecular process. The excitonic luminescence components  $I^+$  (solid line) and  $I^-$  (dashed line) are obtained from the numerical solution of Eqs. (13), (14), and (17) at the transverse magnetic field  $B = 3$  T. The redissociation of the formed exciton is not considered ( $\alpha = 0$ ). The hole-spin orientation in the formed excitons is blocked ( $1/\tau = 0$ ). Four representative values of the formation time are considered.

rise time of the luminescence is short (small exciton formation time  $1/\gamma n_0 < 1/\Omega$ ).

(2) The strong depolarization of the luminescence (Fig. 5): we emphasize that no spin-relaxation mechanism other than the instantaneous and full hole depolarization in the free state, before the exciton formation, has been introduced in the theory which, as a consequence, predicts no depolarization at  $B = 0$  T. The strong depolarization predicted by the present theory is the consequence of the random phase of electron spin at the moment of the electron-hole binding.

These theoretical expectations must be compared with the experimental observations. We consider the narrow QW of Ref. 3 (samples I and II of 3.0 and 4.8 nm well width, respectively) which show clear excitonic QB's when the excitation is resonant. Under nonresonant excitation, both samples show pronounced oscillations of  $I^+$  and  $I^-$  at the

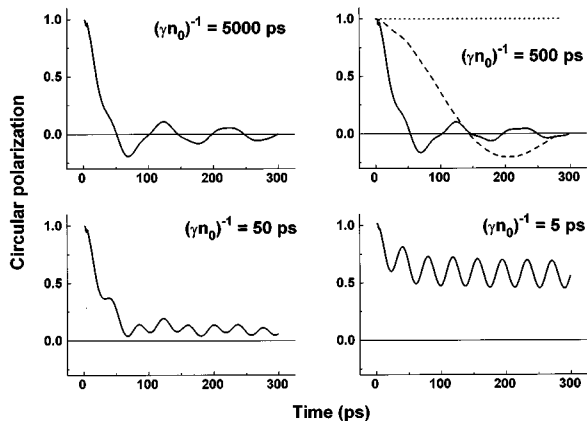


FIG. 5. Simulation of nonresonant excitation experiments. The polarization decays correspond to the luminescence intensity dynamics of Fig. 4. The effect of the magnetic field is illustrated for  $(\gamma n_0)^{-1} = 500$  ps (dotted line  $B = 0$ , dashed line  $B = 1$  T, solid line  $B = 3$  T).

electron Larmor pulsation  $\omega$ , with a phase shift of  $\pi$  between them [Fig. 1(a)]. The oscillations super-impose on the long rise of the exciton PL mainly related to the cooling of the electronic system, an usual effect after above band-gap excitation:<sup>11</sup> in the experiment, the role of the bimolecular formation of excitons on the luminescence rise is hidden by the cooling effect.

When the luminescence components recorded at  $B = 0$  T are reported on the same figure [Fig. 2(a)], it appears clearly that the amplitude of the oscillations in the magnetic field is equal to  $I^+ - I^-$  at zero field. This proves that the coupling of electron spins with the transverse magnetic field does not really result in an additional depolarization when the excitation is nonresonant. This is in contradiction with the above theoretical expectation that a strong depolarization contribution arises from the dephasing of the electron spins when  $1/\tau = 0$  in the excitonic system, related to the random binding. As a consequence, the hypothesis that the random binding of free electrons and holes generates excitons in which the hole-spin orientation is blocked, does not hold.

## 2. The hole-spin orientation is not stable in the hot excitons formed by random binding

It was shown in Sec. II that the amplitude of the PL oscillations is directly related to the hole spin-flip rate within the excitons. The conclusion was that the application of the transverse magnetic field does not involve an additional damping of the electronic QB provided that the condition  $\tau \ll \hbar/\delta$  is fulfilled. This suggests that the hole-spin orientation is really unstable in the hot exciton formed by random binding of  $e-h$  pairs. The effect of this instability is investigated now.

Two cooling regimes have been observed in the thermalization of the excited electronic system (electrons, holes, excitons), after the strong initial LO-phonon emission which has reduced the electronic temperature to about 80–60 K within the excitation pulse.<sup>4,15</sup>

The fast cooling regime, in which the temperature drops from 80–60 K to 30–20 K in about 20 ps: this regime corresponds to the end of the LO-phonon emission (the emission of LO phonons is possible even if the carriers are excited with an excess energy lower than the LO-phonon energy since a fraction of the carriers, after the internal thermalization process is achieved, has kinetic energy greater than the LO-phonon energy).

The slow-cooling regime which occurs at a much longer time scale of several hundreds of picoseconds: this regime corresponds to acoustic phonon emission.

Theory predicts that the stability of the hole-spin orientation within the exciton decreases when the temperature increases.<sup>16</sup> We checked this point previously.<sup>10</sup> As a consequence, a fast increase of the hole spin-flip time  $\tau$  is expected during the fast-cooling regime and a slow increase is expected during the slow-cooling regime. We consider here the crude model of variation according to which  $\tau$  increases exponentially in time from an initial value  $\tau_0$  (less than  $\sim 1$  ps) to a quasistabilized value denoted  $\tau_\infty$  reached at the end of the fast cooling regime, i.e., in a characteristic time of let us say 20 ps. The numerical solution of Eqs. (13), (14), and (17) is performed taking  $\tau_0 = 1$  ps (shorter initial spin-flip times do not change the results) and  $\hbar/\delta = 6.6$  ps, the exchange

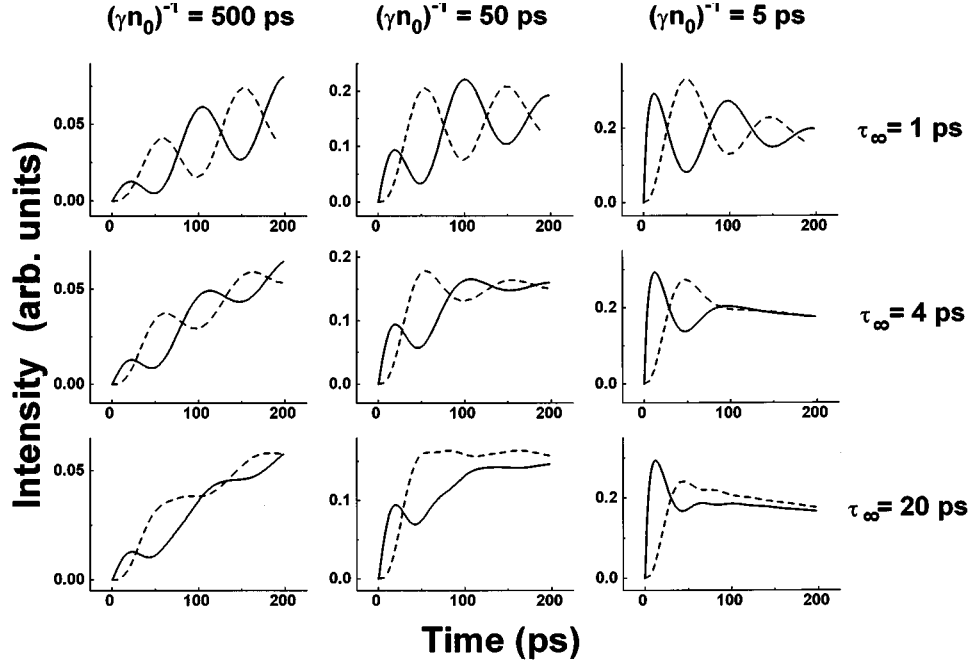


FIG. 6. Simulation of nonresonant excitation experiments. The excitonic luminescence components  $I^+$  and  $I^-$  are obtained from the numerical solution of Eqs. (13), (14), and (17). The redissociation of the formed exciton is not considered ( $\alpha=0$ ). The hole-spin relaxation within the formed excitons is considered for three typical values of the hole spin-flip rate at long delay ( $1/\tau_\infty$ ). The consequence for the luminescence quantum beats is shown for different exciton formation times.

splitting measured in sample II. The resulting luminescence dynamics  $I^\pm$  are displayed in Fig. 6 for representative values of the exciton formation time at  $t=0$ . For formation times longer than 500 ps, we do not see significant changes of the dynamics in the time range of interest. The corresponding luminescence polarization dynamics is shown in Fig. 7. Three different values of the quasistationary hole spin-flip time into the formed excitons ( $\tau_\infty$ ) are considered. Two of them ( $\tau_\infty=1$  and 4 ps) satisfy the condition  $\tau_\infty < \hbar/\delta$  while the third one ( $\tau_\infty=20$  ps) satisfies the opposite condition  $\tau_\infty > \hbar/\delta$ . It appears clearly that the simulation of electronic QB's of large amplitude, the requirement for the interpretation of the experiments, is obtained only for values of  $\tau_\infty$  for which the condition  $\tau_\infty < \hbar/\delta$  is fulfilled. One can conclude

that the observation of electronic QB in above bandgap excitation conditions implies that the condition  $1/\tau > \delta/\hbar$  is fulfilled during all the time range of the depolarization ( $\sim 100$  ps).

### C. The exciton redissociation is considered in the frame of a dynamical equilibrium between excitons and $e-h$ pairs

In a previous paper<sup>4</sup> we found that a similar time dependence observed for the exciton homogeneous linewidth and the carrier temperature in GaAs QW is consistent with the assumption of thermodynamical equilibrium for the free carriers and the exciton assembly. This would imply that the number of  $e-h$  pairs forming excitons per unit time is equal

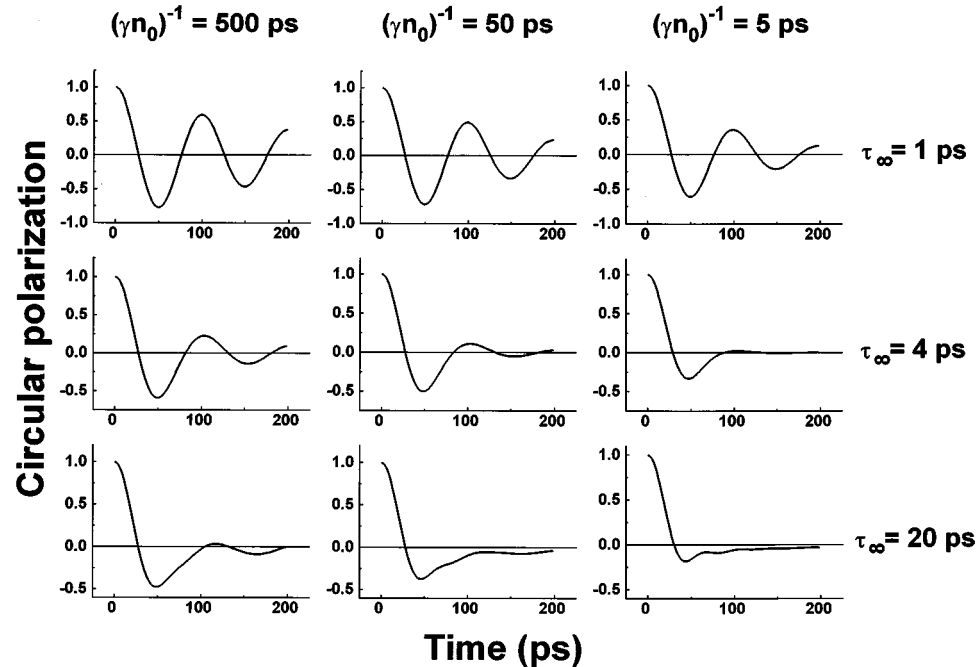


FIG. 7. Simulation of the polarization dynamics corresponding to the luminescence intensity dynamics of Fig. 6.



to the number of excitons ionizing per unit time at any time; the time dependence of the corresponding populations is then governed by the cooling of the electronic system. In this case, the dissociation time  $1/\alpha$  is related to the formation coefficient  $\gamma$ . From the detailed balance principle,

$$K(T) = \frac{\alpha}{\gamma} = \frac{\mu_x}{2\pi\hbar^2} k_B T e^{-E_x/k_B T}, \quad (18)$$

$\mu_x = (m_e^{-1} + m_h^{-1})^{-1}$  is the reduced effective mass;  $E_x$  is the exciton binding energy;  $T$  is the temperature of the excited system. We examine here if the instability of the exciton state related to the concept of dynamical equilibrium can be the basis of an alternative interpretation of electronic QB's observed in above bandgap excitation experiments.

In the frame of the dynamical equilibrium, the dissociation time of excitons  $T_x = 1/\alpha$ , is the *stability time* of the exciton state. Then the condition for the observation of electronic QB's in the excitonic luminescence is  $T_x < \hbar/\delta$  during all the depolarization period ( $\sim 100$  ps). Taking Eq. (18) into account and considering the long cooling of the electronic system described above, this condition writes

$$\gamma(T) > \frac{\delta}{\hbar K(T)} \quad \text{for } T \geq 20 \text{ K}. \quad (19)$$

The function  $\delta/\hbar K(T)$  is displayed in Fig. 8 for the parameter values ( $\delta$  and  $E_x$ ) of samples I and II. It is not expected that condition (19) is fulfilled: from Fig. 8, this should require very high values of the formation coefficient ( $\gamma \geq 1000 \text{ cm}^2/\text{s}$ ) which have never been reported. We have checked these qualitative conclusions by simulations using Eqs. (13) and (14), in which  $\alpha$  was replaced by  $\gamma K(T)$  and  $1/\tau$  taken equal to zero.

One can conclude that the instability of the exciton state, related to a possible dynamical equilibrium between excitons and dissociated  $e$ - $h$  pairs, is not strong enough to explain alone the observation of electronic QB's. This means that if the concept of dynamical equilibrium is correct during the first hundred of picoseconds after the above bandgap laser excitation, the instability of the hole-spin orientation in the hot excitons, related to the long cooling of the electronic system, is here also a necessary additional hypothesis.

#### IV. CONCLUSION

We have developed a theory of the QB in the excitonic PL accounting for the hole-spin instability within the exciton. This theory provides theoretical support to the intuitive interpretation given previously.<sup>3</sup>

When excitons are generated resonantly by the laser pulse the luminescence oscillations are of excitonic or electronic kind according to whether  $1/\tau < \delta/\hbar$  or  $1/\tau > \delta/\hbar$ , respectively.

When the excitation is nonresonant, above the QW bandgap, QB's of electronic kind are always observed, regardless of the well-width value. The theoretical study led us to the conclusion that the random binding of  $e$ - $h$  pairs gives hot excitons in which the hole-spin orientation is not stable. The progressive stabilization of the orientation in excitons is due to the effect of the cooling *via* the emission of acoustic phonons. But the stabilization is far from being achieved at

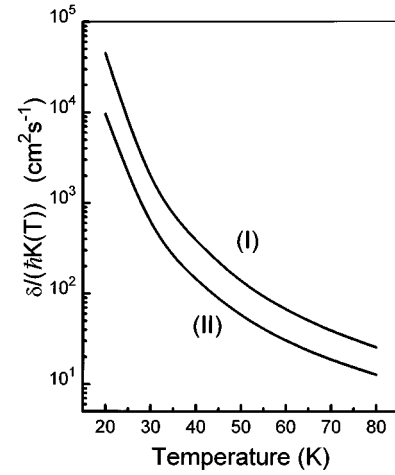


FIG. 8. The minimum value of the formation coefficient [ $\delta/\hbar K(T)$ ] vs temperature for the observation of electronic QB's (hypothesis of a dynamical equilibrium between free electrons holes and excitons). (I) Sample I parameters:  $E_x = 14$  meV,  $\delta = 0.13$  meV. (II) Sample II parameters:  $E_x = 12$  meV,  $\delta = 0.10$  meV.

the end of the depolarization period which lasts typically about 100 ps. We show, moreover, that the additional implication of exciton redissociation during the decay, considered in the frame of the dynamical equilibrium between excitons and dissociated pairs, does not change this conclusion: the rate of the alternative exciton formation and dissociation is not high enough, in principle, to explain alone the manifestation of the strong electronic QB's observed.

Finally the main factors which contribute to the stabilization of the hole spin orientation within an exciton are the following: the increase of the electron-hole spin correlation *via* the increase of the exchange interaction related with the confinement and the corresponding decrease of the valence band mixing; the decrease of the exciton temperature.

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#### APPENDIX

Optical orientation and alignment of excitons are usually obtained by solving the equation of evolution of the density operator whose components are denoted by  $\rho_{mn}(t)$  in the excitonic basis  $\{|m = J_z + S_z\rangle\}$  where  $m = 1, 2, 1, 2$ :

$$\frac{d\rho}{dt} = \frac{1}{i\hbar} [\mathcal{H}, \rho] + \left. \frac{\partial \rho}{\partial t} \right|_{\text{relax}}. \quad (A1)$$

$\left. \partial \rho / \partial t \right|_{\text{relax}}$  is the relaxation term which generally includes spin relaxation and recombination.

The average value of any single-particle observable  $\mathcal{O}$  can be expressed in terms of the density-matrix components from  $\langle \mathcal{O} \rangle = \text{Tr}\{\rho(t)\mathcal{O}\}$ . Thus, for  $S_x$ ,  $Q_x$  and  $J_z$  we find

$$\begin{aligned}
\langle S_x \rangle &= \frac{1}{2} (\rho_{12} + \rho_{-1-2} + \rho_{21} + \rho_{-2-1}), \\
\langle S_y \rangle &= \frac{i}{2} (-\rho_{12} + \rho_{-1-2} + \rho_{21} - \rho_{-2-1}), \quad (\text{A2a}) \\
\langle S_z \rangle &= \frac{1}{2} (-\rho_{11} + \rho_{-1-1} + \rho_{22} - \rho_{-2-2}), \\
\langle Q_x \rangle &= \frac{1}{2} (\rho_{12} - \rho_{-1-2} + \rho_{21} - \rho_{-2-1}), \\
\langle Q_y \rangle &= \frac{i}{2} (-\rho_{12} - \rho_{-1-2} + \rho_{21} + \rho_{-2-1}), \quad (\text{A2b}) \\
\langle Q_z \rangle &= \frac{1}{2} (-\rho_{11} - \rho_{-1-1} + \rho_{22} + \rho_{-2-2}), \\
\langle J_z \rangle &= \frac{3}{2} (\rho_{11} - \rho_{-1-1} + \rho_{22} - \rho_{-2-2}). \quad (\text{A2c})
\end{aligned}$$

The time evolution of the average value of any time-independent observable  $\mathcal{O}$  is derived from Eq. (A1). Using the cyclic property of the Trace operator we obtain

$$\begin{aligned}
\frac{d\langle \mathcal{O} \rangle}{dt} &= \text{Tr} \left[ \frac{d\rho}{dt} \mathcal{O} \right], \\
\frac{d\langle \mathcal{O} \rangle}{dt} &= \frac{1}{i\hbar} \langle [\mathcal{O}, \mathcal{H}] \rangle + \left. \frac{\partial \langle \mathcal{O} \rangle}{\partial t} \right|_{\text{relax}}, \quad (\text{A3a})
\end{aligned}$$

with the expression of the relaxation term in Eq. (A3a):

$$\left. \frac{\partial \langle \mathcal{O} \rangle}{\partial t} \right|_{\text{relax}} = \text{Tr} \left[ \left. \frac{\partial \rho}{\partial t} \right|_{\text{relax}} \mathcal{O} \right]. \quad (\text{A3b})$$

In order to interpret the role of the hole spin-flip process on the time evolution of the excitonic luminescence in the presence of a transverse magnetic field (Sec. II), we take the Hamiltonian in Eq. (A1) in the form given by Eq. (1) and restrict the relaxation term to the single-particle hole-spin relaxation. With holes oriented either along  $Oz^+$  or  $Oz^-$ , the nonzero components in the excitonic basis  $\{|m\rangle\}$  take the form

$$\left. \frac{\partial \rho_{S+J, S'+J'}}{\partial t} \right|_{\text{sr}} = - \frac{\rho_{S+J, S'+J'} - \rho_{S-J, S'-J'}}{2\tau}. \quad (\text{A4})$$

In this formula,  $S \equiv S_z$  and  $J \equiv J_{h,z}$ . The relaxation terms in the time evolution of  $\langle \mathbf{S} \rangle$ ,  $\langle \mathbf{Q} \rangle$ , and  $\langle J_z \rangle$  are derived taking Eq. (A4) into Eq. (A3b). Taking account of Eq. (A2) the result may be written in the form

$$\left. \frac{\partial \langle \mathbf{S} \rangle}{\partial t} \right|_{\text{sr}} = 0, \quad (\text{A5a})$$

$$\left. \frac{\partial \langle \mathbf{Q} \rangle}{\partial t} \right|_{\text{sr}} = - \frac{\langle \mathbf{Q} \rangle}{\tau}, \quad (\text{A5b})$$

$$\left. \frac{\partial \langle J_z \rangle}{\partial t} \right|_{\text{sr}} = - \frac{\langle J_z \rangle}{\tau}. \quad (\text{A5c})$$

Equations (4) result from Eqs. (A3a) and (A5) with  $\mathcal{O} \equiv S_\lambda$ , and  $Q_\lambda$  and  $J_z$  successively.

The radiative recombination of excitons could be included similarly in Eq. (4). The recombination contribution to the relaxation term in Eq. (A1) has the standard form:<sup>17</sup>

$$\left. \frac{\partial \rho_{m,m'}}{\partial t} \right|_{\text{rec}} = - \frac{1}{2\tau_{\text{rad}}} (\delta_{|m|,1} + \delta_{|m'|,1}) \rho_{m,m'}. \quad (\text{A6})$$

Taking this matrix form of  $\partial\rho/\partial t|_{\text{relax}}$  in Eq. (A3b) and taking account of Eq. (A2), the contributions take the form

$$\left. \frac{\partial \langle \mathbf{S} \rangle}{\partial t} \right|_{\text{rad}} = - \frac{\langle \mathbf{S} \rangle - \langle J_z \rangle \mathbf{n}/3}{2\tau_{\text{rad}}}, \quad (\text{A7a})$$

$$\left. \frac{\partial \langle \mathbf{Q} \rangle}{\partial t} \right|_{\text{rad}} = - \frac{\langle \mathbf{Q} \rangle - \mathbf{n}/2}{2\tau_{\text{rad}}}, \quad (\text{A7b})$$

$$\left. \frac{\partial \langle J_z \rangle}{\partial t} \right|_{\text{rad}} = - \frac{\langle J_z \rangle - 3\langle S_z \rangle}{2\tau_{\text{rad}}}. \quad (\text{A7c})$$

In Sec. III, we consider the macroscopic observables of the excitonic system  $\Sigma$ ,  $\mathcal{Q}$ , or  $\mathcal{J}$  and of the electronic system  $\sigma$ . Their time evolution is also described by equation of the form (A3).

For the excitonic system, the Hamiltonian is still expressed by Eq. (1). The single-particle hole spin flip and the radiative recombination contributions to the relaxation term  $\partial\rho/\partial t$  keep the form [Eqs. (A5) and (A7)] but with  $\Sigma$ ,  $\mathcal{Q}$ , and  $\mathcal{J}$  replacing  $\mathbf{S}$ ,  $\mathbf{Q}$ , and  $\mathbf{J}$ . The relaxation term includes in addition the contributions related to the exciton formation and exciton dissociation, the form of which is represented by Eqs. (12). Equations (17a) and (17b) are obtained by this way.

For the electronic system, the spin Hamiltonian reduces to the Zeeman term  $\hbar\boldsymbol{\omega}\cdot\mathbf{s}$  and the relaxation term reduces to the contributions of the exciton formation and dissociation processes [Eq. (12a)]. Equations (17a) are obtained in this way.

The contribution of radiative recombination to the time evolution of the exciton density is derived from Eq. (A3b) taking  $\mathcal{O} \equiv N$ . We obtain

$$\begin{aligned}
\left. \frac{\partial N}{\partial t} \right|_{\text{rad}} &= \text{Tr} \left[ \left. \frac{\partial \rho}{\partial t} \right|_{\text{rad}} N \right] = N \text{Tr} \left[ \left. \frac{\partial \rho}{\partial t} \right|_{\text{rad}} \right] \\
&= N \left( \left. \frac{\partial \rho_{11}}{\partial t} \right|_{\text{rad}} + \left. \frac{\partial \rho_{22}}{\partial t} \right|_{\text{rad}} + \left. \frac{\partial \rho_{\bar{1}\bar{1}}}{\partial t} \right|_{\text{rad}} + \left. \frac{\partial \rho_{\bar{2}\bar{2}}}{\partial t} \right|_{\text{rad}} \right). \quad (\text{A8})
\end{aligned}$$

From Eq. (A6) the result is

$$\left. \frac{\partial N}{\partial t} \right|_{\text{rad}} = - \frac{1}{\tau_{\text{rad}}} \left( \frac{N}{2} - \langle Q_z \rangle \right). \quad (\text{A9})$$

This contribution is added to the contributions coming from the exciton formation and the exciton dissociation processes in Eq. (14b).

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