Spin Quantum Beats of 2D Excitons

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We report on spin quantum beats of excitonic kind in the time-resolved photoluminescence of quantum wells in a magnetic field. When this field is perpendicular to the growth direction, conditions for the manifestation of the electron or exciton spin precession in the circularly polarized components of the excitonic luminescence are obtained. These results lead to a direct measurement of the electron-hole exchange energy of the 2D exciton and give important insights into the exciton properties. [S0031-9007(97)02326-0]

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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). In semiconductors, QB from excitons have been seen in resonance fluorescence [1,2], optical absorption [3], degenerate-four-wave mixing [4], or linear birefringence [5] experiments.

Recently, QB have been reported in the time-resolved free exciton photoluminescence (PL) of type I quantum well (QW), when a magnetic field perpendicular to the growth axis is applied [6,7]. These QB, which are observed on a time scale of a few hundreds of picoseconds, are interpreted by Heberle *et al.* [6] in terms of Larmor precession of the electron spins around the axis of the magnetic field. The corresponding pulsation ω, which directly reveals the electron spin splitting $\hbar\omega = g_e \mu_B B$, allows the determination of the electron Landé g factor g_e.

However, at low temperature, when the laser excitation is resonant with the heavy-hole exciton (XH), the electron is bound into an exciton. This raises a fundamental question. On the ground of [8], in which all the spin relaxation processes were ignored, it is expected that QB in the excitonic photoluminescence should occur with a pulsation $\Omega = \hbar^{-1}\sqrt{\delta^2 + (\hbar\omega)^2}$, where δ is the electron-hole exchange energy which splits the XH $1s$ quadruplet into the radiative and the nonradiative pairs of states. As a matter of fact, the authors of Ref. [6] did not observe any change in the linear dependence of the oscillation frequency on the applied magnetic field for resonant or nonresonant excitation conditions, high or low excitation density, or higher temperatures (T up to 200 K) at which free electrons and holes certainly prevail. Moreover, this linear dependence was verified at the smallest field values ($B = 0.12$ T). This lack of excitonic manifestation is surprising.

We have performed time-resolved PL on type I GaAs and GaInAs undoped QW grown by molecular beam epitaxy along the z axis, in a transverse magnetic field (x axis). The samples, which are immersed in liquid helium at 1.7 K, are mounted in Voigt configuration. They are excited with 1.2 ps pulses from a mode-locked Ti:sapphire laser with a repetition rate of 82 MHz. The laser beam is circularly polarized (σ^+). The *exciton luminescence components* of opposed helicities I^+ and I^- are detected by the up-conversion technique with a time resolution limited by the laser pulse width. We report on resonant and nonresonant excitation experiments at photogenerated densities of about 10^9 cm^{-2}. The QB originating from Larmor precession of electron spins could be observed in all the samples. The new results relate to narrow $GaAs/Al_{0.33}Ga_{0.67}As$ QW when the excitation is resonant with XH, the situation which was not explored in [6,7]: Here, a specific excitonic behavior is observed. We report on two samples to illustrate this new and general effect. Sample I contains thirty-three 3 nm wells separated by 10 nm barriers; sample II contains sixty 4.8 nm wells separated by 15 nm barriers. The Stokes shift which separates the XH absorption and luminescence peaks is 7 and 6 meV, respectively; this allows the recording of the luminescence intensity dynamics in resonant excitation conditions.

In nonresonant excitation conditions, when the excitation energy is higher than the QW band gap ($E_1 - HH_1$) but lower than the light-hole exciton (XL) resonance, all the samples exhibit QB on I^+ and I^- when the magnetic field is applied, with a similar behavior as reported in [6,7]. As shown in Fig. 1(a) for the sample I, oscillations on I^+ and I^- are phase shifted by π; moreover, the oscillation frequency is proportional to the magnetic field intensity.

When the excitation is resonant with XH, we observe a completely different behavior. Now, the QB are visible in samples I and II, only at the highest field values that

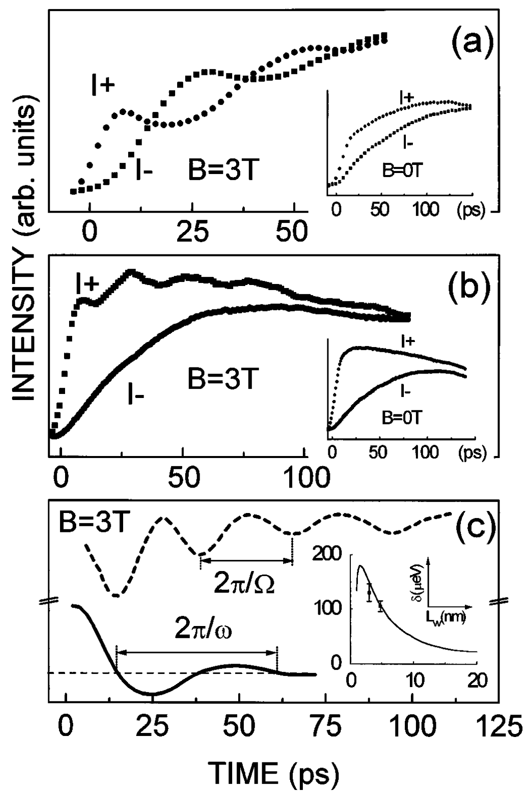

FIG. 1. Sample I: Luminescence intensity dynamics after σ^+-polarized excitation. (a) The excitation energy is nonresonant ($E_1 - HH_1 < h\nu < XL$) and $B = 3$ T (inset, $B = 0$ T). (b) The excitation energy is resonant with XH and $B = 3$ T (inset, $B = 0$ T). (c) The oscillations of the luminescence intensity component I^+ in resonant excitation (dashed line) and of the luminescence polarization P_L in nonresonant excitation $E_1 - HH_1 < h\nu < XL$ (full line), under the same magnetic field $B = 3$ T. For the sake of clarity, the monotonous component has been subtracted from I^+. Inset: the well-width dependence of the exciton exchange energy δ, from our experiment (dots with error bars) and theory after Ref. [9] (full line).

could be produced by our coil, i.e., between 2.5 and 3.5 T. As shown in Fig. 1(b) for sample I, they appear then as a weak amplitude modulation on the I^+ component, but they are not really observable on I^-. Moreover, as the comparison in Fig. 1(c) shows, the modulation pulsation Ω is higher than in nonresonant excitation conditions at the same field value.

The modulations observed in nonresonant and resonant excitation conditions, Figs. 1(a) and 1(b), respectively, are attributed to the Larmor precession of the free electron (ω) and the heavy-hole exciton (Ω), respectively, referred to as electron QB and exciton QB in the following. The resulting $g_e = \hbar\omega/\mu_B B$ values ($g_e = 0.50 \pm 0.01$ and $g_e = 0.24 \pm 0.01$ in samples I and II, respectively) are in excellent agreement with the g_e factor measured in Ref. [6,7]. Also the resulting excitonic exchange energy $\delta = \hbar\sqrt{\Omega^2 - \omega^2}$ values ($\delta = 130 \pm 15$ and $\delta = 105 \pm 10$ μeV in sample I and II, respectively) are in agreement with the theoretical prediction for free excitons, after Ref. [9], as shown in the inset of Fig. 1(c). Nevertheless in the present experiment the Stokes shift means that the excitons are bound, not free. We conclude that the spin dynamics of a free versus bound exciton are similar. Moreover, we show below that the amplitude of the excitonic oscillations should be reduced by the factor $(\omega/\Omega)^2$. This explains why these oscillations are detected only at the highest field values.

On the ground of these new results, we now understand why the QB reported in Ref. [6,7] in resonant and nonresonant excitation conditions both originate from Larmor precession of electrons. 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 splitting δ no longer plays a role in the QB pulsation which then reflects simply the electron Larmor precession. The argument follows. For the electron into an exciton, the exchange interaction with the hole in a defined spin state is equivalent to an external magnetic field of intensity $B_{\mathrm{ex}} = \delta/g_e\mu_B$, orientated along the z axis. Then the hole spin instability is equivalent to the instability of $\vec{B}_{\mathrm{ex}}$, orientated along either oz^+ or oz^-. If the rate of change of this orientation is higher than $\delta/\hbar$, this magnetic field B_{ex} has no effect on the electron spin precession. Hence QB are observed at the pulsation ω provided that many hole spin-flip events occur during one period of precession. Finally an electron bound into an exciton precesses like a free electron in the transverse magnetic field provided that $\mathcal{T}_h \ll \hbar/\delta$, $1/\omega$ where $\mathcal{T}_h$ is the single particle hole spin-flip time [10]. This conclusion is supported by the quantitative approach summarized hereafter. These two conditions may be fulfilled in large and narrow QW but for different reasons.

In large QW (*a fortiori* in the bulk) such a hole spin instability occurs as a consequence of the mixing of states in the valence band. The observation of the electron Larmor precession in a QW of 25 nm well width under resonant XH excitation condition reported in [6] has to be understood on this ground. But the hole spin instability is not specific to large QW. When the excitation energy is nonresonant, higher than the QW band gap, the hole spin-flip time is very short: typically $\mathcal{T}_h \lesssim 4$ ps in intrinsic QW of 5 nm at 1.7 K for all the excitation energies above the band gap, according to Ref. [11]. The condition $\mathcal{T}_h \ll \hbar/\delta$ for the observation of QB at the pulsation ω is thus fulfilled in nonresonant excitation, regardless of the well-width value. The origin of the instability of the hole spin orientation is related here to the high temperature of the electronic system after a nonresonant excitation. This point shall be analyzed in a separate publication.

On the other hand, the observation of QB of the excitonic kind in narrow enough QW under resonant excitation conditions proves that the hole spin is much more stable in cold 2D excitons ($\mathcal{T}_h \gg \hbar/\delta$), in agreement with other recent indications [10,12]. However, if there is no hole spin scattering during the QB observation time, σ^- luminescence is, at first sight, not expected. In fact, Fig. 1(b) clearly shows such luminescence. This luminescence is not attributed to scattered holes, but to the simultaneous spin flip of the electron and the hole, i.e., the spin flip of the exciton as a whole, a process similar to the one described in Ref. [12].

Figure 2 displays the progressive change of I^+, I^-, and P_L dynamics when the excitation energy is tuned from a position above the band gap [Fig. 2(a)] to below the band gap [Figs. 2(b) and 2(c)], down to the XH resonance [Fig. 2(d)], at $B = 2.8$ T. The conclusion is that a very accurate resonant excitation (± 1 meV) is required for a clear observation of exciton QB. Figures 2(b) and 2(c) correspond to intermediate situations where the hole spin in the photogenerated excitons is not stable enough for the observation of pure exciton QB but not unstable enough for the observation of the pure and strong electronic oscillations which change the sign of the polarization, as reported in Fig. 1(a).

The quantitative approach is based on the density matrix formalism. 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 XH states are described using the basis set $|J_z\rangle = |s_z + j_{h,z}\rangle$. The exciton density matrix satisfies the kinetic equation

$$\frac{d\rho}{dt} = \frac{1}{i\hbar}[\mathcal{H}, \rho] + \left(\frac{\partial \rho}{\partial t}\right)_{\text{s.r.}}. \tag{1}$$

The exciton spin Hamiltonian in the transverse magnetic field $\vec{B} \parallel \overrightarrow{ox}$, including the exchange term, writes $\mathcal{H} = \hbar\omega\hat{s}_x - (2/3)\delta\hat{j}_z\hat{s}_z$ (in QW, the transverse g factor of a $j_{h,z} = \pm 3/2$ hole is zero). In order to interpret the impact of the hole spin-flip mechanism on the QB oscillations, the relaxation term in (1) is restricted to the hole spin relaxation contribution (s.r.). With hole spins orientated either along oz^+ or oz^-, the nonzero components in the basis $\{|1\rangle, |2\rangle, |\bar{1}\rangle, |\bar{2}\rangle\}$ take the form

$$\left(\frac{\partial \rho_{s+j,s'+j}}{\partial t}\right)_{\text{s.r.}} = -\frac{\rho_{s+j,s'+j} - \rho_{s-j,s'-j}}{2\mathcal{T}_h}. \tag{2}$$

Here, $s \equiv s_z$ and $j \equiv j_{h,z}$. The resolution shall be developed in a forthcoming publication. For the two extreme situations of actual interest, i.e., short and long

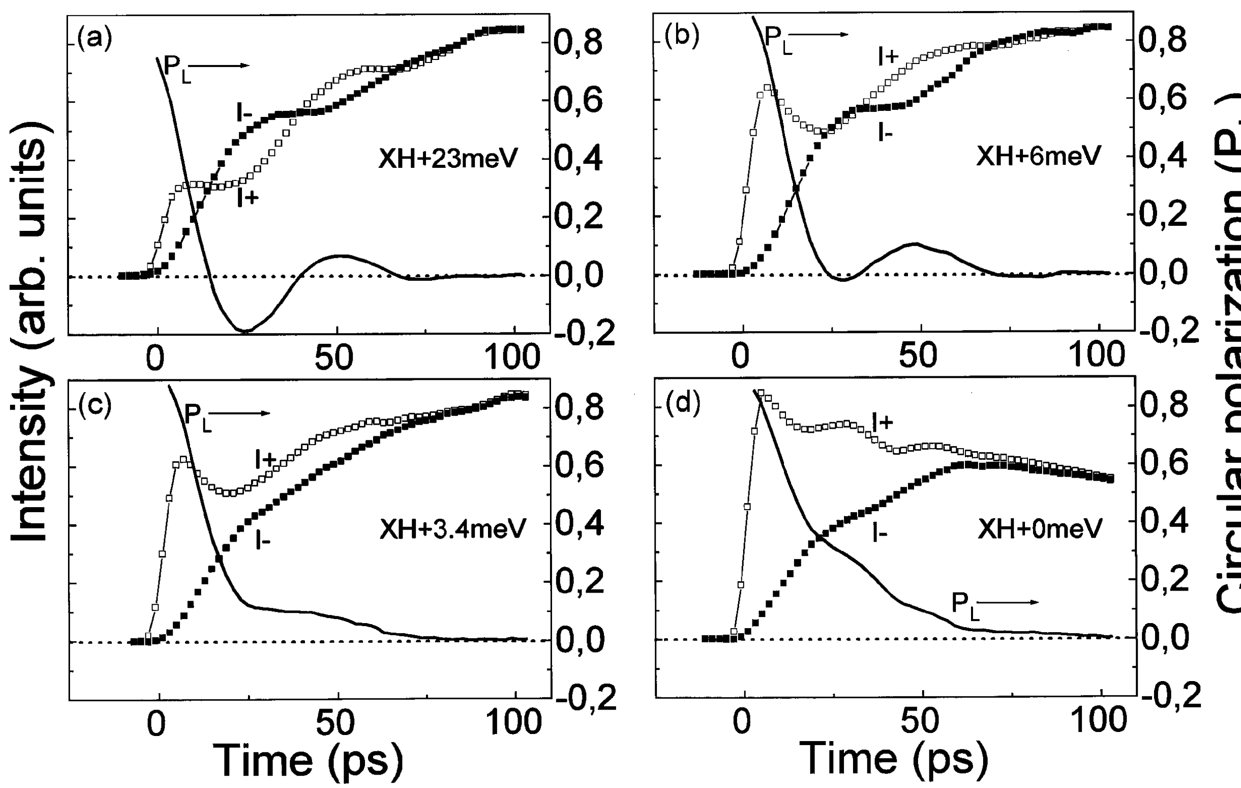

FIG. 2. Sample I: Transverse magnetic field $B = 2.8$ T. Luminescence intensity dynamics I^+, I^- and luminescence polarization P_L after σ^+-polarized laser excitation. (a) The excitation energy is above the QW band gap $HH_1 - E_1$ but below the XL resonance. (b), (c) The excitation energy is between the XH resonance and the QW band gap $HH_1 - E_1$. (d) The excitation energy is resonant with XH.

hole spin relaxation time, the solution reduces to

$$\begin{aligned} &\mathcal{T}_h \ll \hbar/\delta, 1/\omega \atop \text{electron QB} \Longrightarrow I^{\pm}(t) \propto \rho_{\pm1,\pm1}(t) = \frac{1 \pm \cos\omega t}{4}, \\ &\mathcal{T}_h \gg \hbar/\delta \atop \text{exciton QB} \Longrightarrow \begin{cases} I^{+}(t) \propto \rho_{1,1}(t) = 1 - \dfrac{\omega^2}{\Omega^2}\dfrac{1-\cos\Omega t}{2} \\ I^{-}(t) \propto \rho_{\bar{1},\bar{1}}(t) = 0 \end{cases}, \end{aligned} \tag{3}$$

which describes the electron and exciton spin precessions at pulsation ω and Ω, respectively. In the case of exciton QB the copolarized luminescence component modulated at the frequency Ω has an amplitude reduced by the factor $(\omega/\Omega)^2$, while the counterpolarized component is unmodulated. This supports clearly the interpretation of the experimental observations given above.

Bar-Ad *et al.* [3] have also reported QB in pump-probe measurements with linear polarization in multiple quantum well (MQW) GaAs/AlGaAs samples under *a longitudinal magnetic field* ($\vec{B} \parallel \overrightarrow{oz}$). These QB between the Zeeman-split levels ($J_z = \pm 1$), at the pulsation $\Omega_{\parallel} = \hbar^{-1}(g_{e,z} + g_{h,z})\mu_B B$, were observed in a stepped MQW in which the exciton wave function is confined mainly in the 3 nm GaAs layer while no trace of modulation could be detected in a square MQW of 8 nm well width. More recently, QB of the same origin were observed by transient linear birefrigence experiments in 2.7 nm well-width MQW [5]. Figure 3 displays the PL linear polarization dynamics for sample II after a linearly polarized excitation, the magnetic field being applied along the z axis. The recording shows that this kind of exciton spin QB at the pulsation $\Omega_{\parallel}$, which requires again the correlation between the electron and the hole spin orientations, is observable also in luminescence. We did not succeed in observing them in QW of 12 nm or wider: This is because this correlation is not strong enough to block the single particule hole spin-flip mechanism.

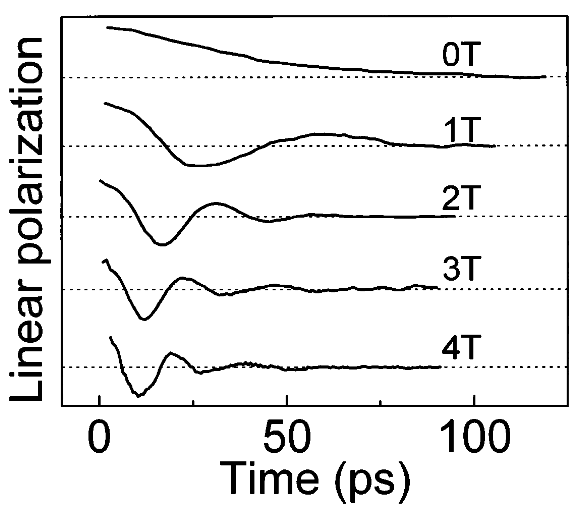

FIG. 3. Sample II: Longitudinal magnetic field, from 0 to 4 T. The luminescence linear polarization dynamics $(I^x - I^y)/(I^x + I^y)$ after a linearly x-polarized excitation.

The present interpretation does not conflict with the measurement of the exciton exchange splitting by Blackwood *et al.* [9] in a cw experiment where the linearly polarized laser excitation was tuned above the QW band gap under a longitudinal magnetic field. In this experiment, the exciton temperature is much lower than it is during the time range of QB observation in the nonresonant transient PL. Within *each cold exciton*, the hole spin orientation (either oz^+ or oz^-) is thus stabilized: This is why the exchange splitting can be measured in the cw experiment.

This Letter reports on the observation of excitonic spin QB in relatively narrow QW, when the excitation is resonant with XH. We demonstrate that the blocking of the single particle hole spin-flip mechanism is the condition for their manifestation. The role played by the electron-hole exchange interaction is enlightened and the exchange energy measured.

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