Quantum Beats of Electron Larmor Precession in GaAs Wells

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We observe quantum beats in the time-resolved photoluminescence of GaAs quantum wells in a magnetic field perpendicular to the growth direction. These beats originate from Larmor precession of electron spins and show a coherence time of 500 ps, which is much longer than the optical dephasing time of 7 ps. The long coherence time allows us to determine the electron Landé g factors with an accuracy better than 1%.

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Coherent phenomena in solids have been of great interest since the development of ultrafast optical techniques capable of resolving a pico- or femtosecond phase relaxation time T_2. The optical phase relaxation time T_2^o is the time in which the electronic excitation loses coherence with the exciting optical field. In GaAs and type I $GaAs/Al_xGa_{1-x}As$ quantum wells (QWs), T_2^o is typically shorter than 10 ps. A striking feature sometimes observed after pulsed excitation is a temporal modulation in the light signal which arises from the fact that a short light pulse can prepare a coherent superposition of excited states with slightly different energy. The coherent evolution of these states then causes an oscillation with a frequency corresponding to the energy splitting of the excited states. This interference between quantum states, called quantum beats, allows splittings to be determined with high resolution. [1] In solids, quantum beats from excitons have been seen with resonance fluorescence [2,3], optical absorption [4], degenerate four wave mixing (DFWM) [5–7], and Faraday rotation [8]. These quantum beats were observed on a time scale comparable to T_2^o.

In this paper, we report on quantum beats in the time-resolved free exciton photoluminescence of GaAs QWs on a time scale which is 2 orders of magnitude longer than the optical phase relaxation time T_2^o. The quantum beats originate from Larmor precession of electron spins in a magnetic field. The phase relaxation time of these spin oscillations, the transverse spin relaxation time, is $T_2^s = 500$ ps compared to the value $T_2^o = 7$ ps obtained from time-resolved DFWM. Our experiments reveal a coherence time scale which exceeds the optical phase relaxation time by orders of magnitude. This long time scale cannot be observed with conventional coherent techniques such as DFWM.

The oscillation frequency of the spin quantum beats directly reveals the electron spin splitting ΔE with a resolution determined by the homogeneous linewidth $\Gamma_h^s = 2\hbar/T_2^s$ of the spin system which is much smaller than the homogeneous exciton linewidth. We are therefore able to measure the electron Landé g factor with high accuracy, and, in particular, at low carrier densities. The g factors in quantum wells were not accessible at low carrier densities using electron spin resonance (ESR) [9] or the Hanle effect [10].

Our sample contains ten 25 nm GaAs QWs separated by barriers which consist of 0.85 nm AlAs, 30 nm $Al_{0.3}Ga_{0.7}As$, and 0.85 nm AlAs. The AlAs layers were introduced to reduce the linewidth. The splitting between the lowest heavy and light hole levels is smaller than the exciton binding energy, which enables us to excite the excitonic and continuum levels separately. We have selected this specific sample for the demonstration of our experiments because, at low excitation density, the exciton linewidth of 300 μeV is close to the homogeneous linewidth of 180 μeV and because of the long electron spin relaxation time. However, similar results were observed in other QWs with different widths as well as in bulk GaAs.

The sample is mounted in Voigt configuration in a He gas flow cryostat in a superconducting magnet. The magnetic field is in the x direction; observation and growth are in the z direction. The sample temperature is 10 K. We excite with ps pulses from a mode-locked Ti:sapphire laser with a repetition rate of 80 MHz. The photoluminescence is dispersed in a 0.32 m spectrometer and detected with a spectral and temporal resolution of 0.5 nm and 10 ps, respectively, by a streak camera with two dimensional readout.

Figure 1(a) shows the polarized (I_+) and depolarized (I_-) luminescence decay of the heavy hole (hh) exciton for the case of circularly polarized resonant excitation of the light hole (lh) exciton. Polarized, I_+ luminescence, means that both luminescence and reflected laser beam have the same circular polarizations, I_- means that they have opposite handedness. The luminescence shows circular polarization with an exponential decay of the degree of polarization $P = (I_+ - I_-)/(I_+ + I_-)$ of 500 ps. The depolarized intensity I_- strongly exceeds I_+ directly after the excitation. This reversal of circular polarization is explained by the level scheme in the inset of Fig. 1(a), indicating the optical transitions which are allowed due to conservation of angular momentum in the z direction. Excitation of lh excitons with σ^+ polarized light creates electrons with spin orientation $s_z = +1/2$. The holes lose their angular momentum rapidly because of the strong

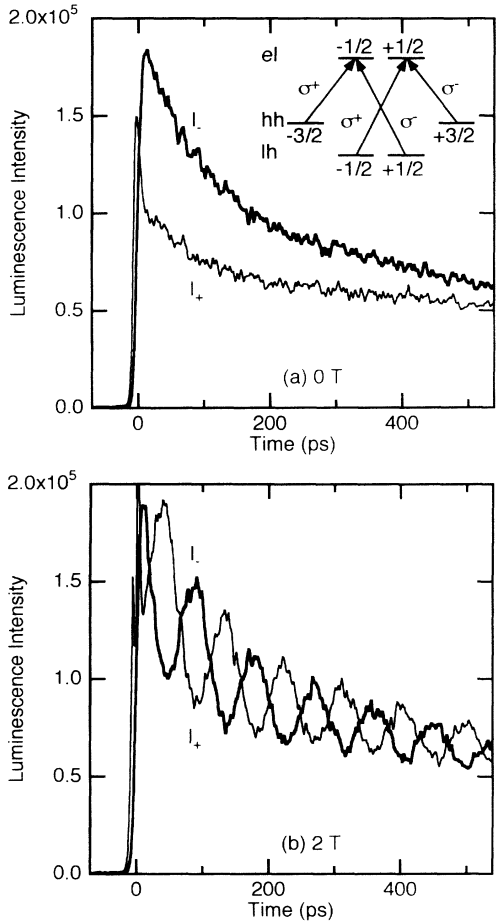

FIG. 1. Luminescence decay at (a) 0 T and (b) 2 T after circularly polarized excitation of the lh exciton. The label I_+ (I_-) marks the intensity of the luminescence having the same (opposite) circular polarization as the reflected excitation laser beam. Scattered laser light causes peaks at time $t = 0$ in I_+. Inset: Level scheme.

spin-orbit coupling in GaAs, whereas the electrons have a significantly longer spin relaxation time [11]. These electrons emit σ^- polarized light when they recombine with heavy holes. The ratio of I_-/I_+ is therefore equal to the ratio of $s_z = +1/2$ to $s_z = -1/2$ electrons.

A magnetic field $B = B_x \perp z$ strongly modifies the luminescence decay, as shown in Fig. 1(b): pronounced oscillations appear. The amplitude of the oscillations is equal to $I_- - I_+$ observed for $B = 0$. The oscillations of I_- have a maximum at time $t = 0$. A phase shift of π is observed for I_+. Such oscillations appear in all investigated bulk and quantum well samples, which preserve spin polarization for a sufficiently long time. The oscillations originate in a Larmor precession of the electron spins around the axis of the magnetic field: this Larmor precession causes oscillations in the average spin polarization in the z direction $\langle s_z \rangle$, which modulates the circular polarization of the luminescence. The resulting

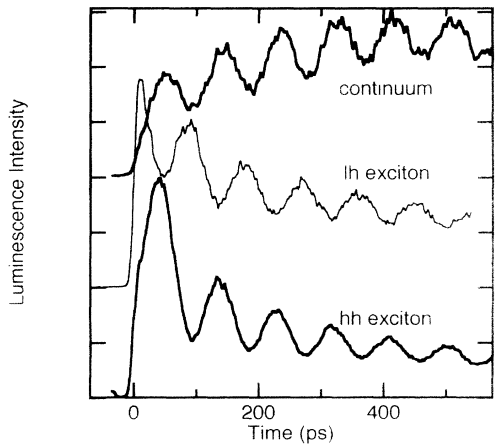

FIG. 2. Dependence of spin oscillations in I_- on laser photon energy.

Larmor angular frequency

$$\omega_L = g_e \mu_B B/\hbar \tag{1}$$

allows us to determine the electron Landé g factor g_e.

Quantum mechanically, the oscillations must be interpreted as quantum beats between spin split ($\Delta E = \hbar\omega_L$) electron levels $\chi_x^{\pm}$ with the spin quantization axis parallel to the magnetic field (x direction). Excitation of an electron with $s_z = +1/2$, however, generates a spin state χ_z^+ with angular momentum in the z direction which is a coherent superposition of the $\chi_x^{\pm}$ states as long as the laser pulse is short enough to excite both states: $\chi_z^+ = (\chi_x^- + \chi_x^+)/\sqrt{2}$. The time evolution of this state is

$$\chi_z(t) = (\chi_x^- e^{+i(\omega_L/2)t} + \chi_x^+ e^{-i(\omega_L/2)t})/\sqrt{2} \tag{2}$$

$$= \chi_z^+ \cos(\omega_L t/2) + i\chi_z^- \sin(\omega_L t/2). \tag{3}$$

Equation (3) shows that the spin in the z direction oscillates between $s_z = +1/2$ (χ_z^+) and $s_z = -1/2$ (χ_z^-) as in the semiclassical picture above.

Figure 2 shows the spin oscillations in I_- at a magnetic field of 2 T for different laser photon energies. A phase shift of π is observed between resonant excitation of the hh exciton and resonant excitation of the lh exciton, because electrons are excited with $s_z = +1/2$ in the former case, and with $s_z = -1/2$ in the latter case. Oscillations are still seen even when states 25 meV into the hh continuum are excited, clearly demonstrating that electrons still preserve spin phase memory after many scattering events during their relaxation into exciton states at lower energy [12]. We are able to observe spin oscillations in all investigated bulk and QW samples, and even at much higher excitation densities of 10^{12} cm^{-2} and in the free electron-hole plasma recombination where optical dephasing is below 100 fs [13,14]. Oscillations remain detectable up to sample temperatures of 200 K. The spin oscillations are only observed if excitation and detection

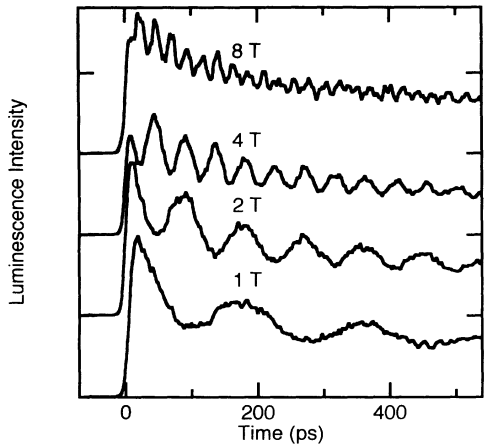

FIG. 3. Luminescence decay I_- for different applied magnetic fields.

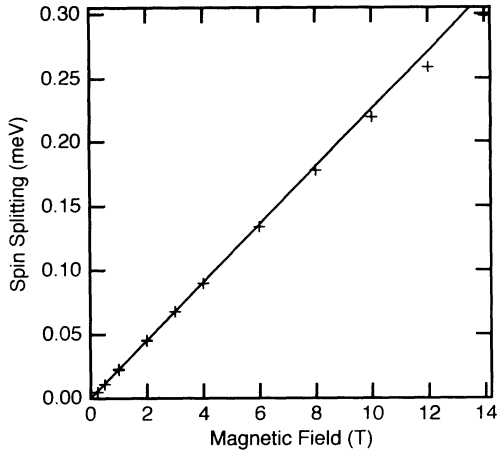

FIG. 4. Dependence of the electron spin splitting on the applied magnetic field (crosses), and spin splitting expected for a constant g factor of -0.390 (line).

are circularly polarized. They vanish for linearly polarized excitation or detection, or if $B \| z$.

Figure 3 shows the oscillations in I_- for different magnetic fields. A doubling of the oscillation frequency is obtained when the magnetic field strength is doubled. Figure 4 depicts the spin splitting as a function of the applied magnetic field (crosses). A weak deviation from a linear behavior (line) is observed for $B > 8$ T revealing a field dependent g factor. We determine an electron g factor of -0.390 from the linear field dependence of the spin splitting below 3 T [15]. For bulk GaAs we obtain a value of -0.447 in good agreement with the published value of -0.44 in literature [16]. This difference of the g factor between QWs and bulk is partly caused by nonparabolicity of the conduction band [17]. This nonparabolicity also causes the field dependence of the g factor.

The g factor determined at low excitation density is the value for an electron bound in an exciton. We did not observe a different g factor for higher densities or temperatures at which free electrons and holes prevail. The nearly constant g factor over the magnetic field range used here leads to the conclusion that the coupling between between electron spin and applied magnetic field is always larger than the coupling between electron and hole spin. Otherwise, a transition between the different coupling regimes should be observable.

The spin oscillations are damped with a spin phase relaxation time T_2^s. A value of $T_2^s = 500$ ps is obtained from the decay of P between $B_\perp = 1/8$ T and $B_\perp = 14$ T and is independent of magnetic field. This time for loss of coherence, the transverse spin relaxation time T_2^s, is equal to the longitudinal spin relaxation time T_1^s measured for $B = 0$ [compare Figs. 1(a) and 1(b)]. Note that the beats are of quantum mechanical nature and do not originate from a polarization interference because they are observed on a much longer time scale than T_2^o. The observation of the Hanle effect in QWs at low magnetic fields is already an indication of extremely long spin phase relaxation times.

Coherent techniques such as quantum beat spectroscopy [4,8] and hole burning [18] were used before to determine g factors in quantum wells. These experiments were performed in Faraday configuration (magnetic field in growth and observation direction) with linear polarized light and revealed the exciton spin splitting from the energy difference of the σ^+ and the σ^- transition. The homogeneous exciton linewidth determined therefore the energy resolution. The choice of Voigt geometry and circular polarization in our experiment selects the electron spin splitting, where the much smaller linewidth of the electron spin system is limiting, when detection insensitive to optical coherence is used.

In conclusion, we observed spin quantum beats in photoluminescence of a GaAs quantum well. The selection of the Voigt geometry allowed us to investigate selectively the electron spins. The long phase relaxation time of the quantum beats of 500 ps results in an energy resolution of the spin splitting of 2.6 μeV. Our method therefore allows a precise determination of Landé g factors (accuracy of 1%).

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