Exciton spin manipulation in InAs/GaAs quantum dots: Exchange interaction and magnetic field effects

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We report quantum beat phenomena between different polarization states in InAs quantum dots, observed in time-resolved photoluminescence and transient dichroism measurements. The polarization of the emitted light can be changed from linear to circular by applying a small magnetic field. Following quasiresonant excitation the electron and hole spin states remain stable during the exciton lifetime, independent of the applied magnetic field. We show that the exciton spin coherence is partially preserved in the energy relaxation process. These experiments enable us to measure the anisotropic exchange energy ΔE_{XY} and the exciton g factor. We find $\Delta E_{XY} \simeq 30$ μeV and $g \simeq 2.5$ for the quantum dot excitons.

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Long optical coherence and spin relaxation times support the numerous proposals for future applications of quantum dots (QDs) in spin-dependent devices.[1–3] To manipulate the charge and spin of an exciton confined to a QD, it is necessary to understand the Coulomb interactions between electrons and holes. In particular, for applications of QDs as single-photon emitters, the anisotropic exchange interaction (AEI) between electrons and holes is crucial as it can determine both the polarization and entanglement of the emitted photons.[4] For perfectly symmetrical dots the heavy-hole exciton states $|+1\rangle$ and $|-1\rangle$ can be excited with circularly polarized light. However, the reduced symmetry of real quantum dots leads to a mixing of the exciton spin doublet via the AEI, resulting in two linearly polarized transitions which are aligned along the orthogonal in-plane crystallographic axes of the dot structure, separated by an energy ΔE_{XY} of up to 200 μeV, as measured in single-dot spectroscopy.[5] Measurements on dot ensembles have shown that under strictly resonant excitation these linearly polarized states are stable: no spin relaxation is observed during the exciton lifetime.[2]

Here we report spin quantum beat phenomena for InAs/GaAs QDs in time-resolved photoluminescence (PL) experiments and in transient dichroism measurements. Stable exciton spin states are created following resonant and quasiresonant excitation and the long-lasting coherence between the states, excited simultaneously by the same laser pulse, gives rise to spin quantum beats. For zero magnetic field we observe quantum beats in the circular polarization, for $B = 0.4$ T we observe beats of the linear polarization, and for an intermediate field of $B = 0.21$ T we observe beats in both circular and linear polarization. We explain these changes in quantum beat polarization and period in terms of an interplay between the AEI and the applied magnetic field. The beat period and decay time allow us to extract the energy differences ΔE_{XY} between different polarization eigenstates and to estimate the relaxation time of different spin states.

The investigated structure was grown by molecular beam epitaxy on a (001) GaAs substrate. It consists of 40 planes of InAs self-assembled QDs, separated by 15-nm-thick GaAs spacer layers. The average QD density is 1×10^{11} cm^{-2} with a typical dot height of 2 nm and a dot diameter of 15 nm. At $T = 10$ K, the ensemble PL transition energy is centered around 1.33 eV, with a full width at half maximum (FWHM) of 60 meV. The sample is excited by 1.5 ps pulses generated by a mode-locked Ti-doped sapphire laser. The time-resolved PL is then recorded by up-converting the luminescence signal in a $LiIO_3$ nonlinear crystal with the picosecond pulses generated by the Ti:sapphire laser. The time resolution is limited by the laser pulse width ($\approx$1.5 ps) and the spectral resolution is about 3 meV. Alternatively, the time-resolved PL is recorded with an S1 photocathode Hamamatsu streak camera system with an overall time resolution of 8 ps and a spectral resolution of 0.45 meV. All PL experiments were performed at 10 K.

The photoinduced dichroism measurements were performed at 2 K, using the apparatus described in Ref. 6. The pulsed beam of a picosecond Ti:sapphire laser is split into pump and probe beams. The probe beam is focused onto the sample in a spot of 50 μm in diameter, and the pump beam is slightly defocused at the same point, in order to have a pump spot larger than the probe one. The photon energy is tuned to 1.35 eV. In the measurements presented here, the pump beam is circularly polarized. The probe beam has a linear polarization, aligned with a crystallographic axis of the QDs. After transmission through the sample, a balanced optical bridge analyzes two linear components of the transmitted beam lying at 45° to the original linear direction of the probe beam. The difference in the intensity of these components is proportional to the Faraday rotation of the principal polarization direction undergone by the probe beam. The

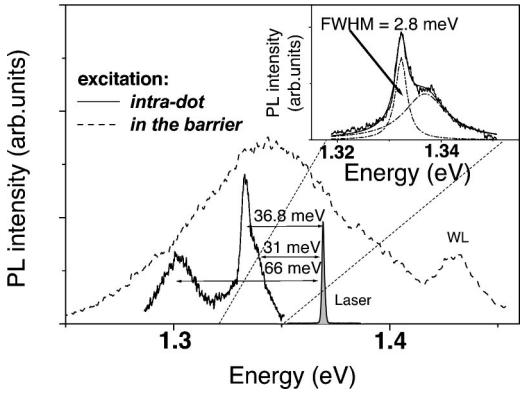

FIG. 1. Time-integrated photoluminescence of the dot ensemble for two different laser photon energies: The laser pulse for intradot excitation is sketched schematically (laser) and the resulting spectrum shown as a solid line. Excitation in the barrier results in a broad emission line (FWHM=60 meV) shown as a dashed line. The emission of the wetting layer (WL) is marked. Inset: The narrow emission following intradot excitation can be fitted with a Lorentzian line shape (dash-dotted lines).

pump and probe beams are chopped at different frequencies, and the heterodyne signal is measured at the difference frequency with a lock-in amplifier. Time-resolved behavior is obtained by recording the rotation angle versus the pump-probe delay. The time and spectral resolutions of the experiment are estimated to be 3 ps and 1 meV, respectively.

We denote linearly and circularly polarized photons as Π^X, Π^Y and σ^-, σ^+, respectively. The linear and the circular polarization degrees of the luminescence are defined as

$$P_{lin} = (I^X - I^Y)/(I^X + I^Y) \tag{1}$$

and

$$P_C = (I^+ - I^-)/(I^+ + I^-), \tag{2}$$

respectively. Here I^X (I^Y) and I^+ (I^-) denote, respectively, the X (Y) linearly polarized and the right (left) circularly polarized luminescence components, where X and Y are chosen to be parallel to the [110] and $[1\bar{1}0]$ crystallographic directions.

For a resonantly photogenerated electron-hole pair in the QD ground state, neither the electron nor the hole spin relaxes during the radiative lifetime of the exciton,[2] allowing in principle the observation of spin quantum beats. In practice, the PL signal is initially obscured by backscattered laser light following resonant excitation. This can be avoided by exciting quasiresonantly:[7] The excitation laser energy is tuned to the energy of one GaAs longitudinal optical (LO) phonon ($\approx$36 meV) above the ground-state energy, at which we detect the PL signal.

The time-integrated PL spectrum is presented in Fig. 1 for two different laser excitation energies. When carriers are created in the GaAs layer surrounding the dots (i.e., above 1.56 eV), a broad PL spectrum centered around 1.33 eV is observed (dashed line in Fig. 1). As the excitation energy is reduced below 1.37 eV, corresponding to a photogeneration of electron-hole pairs directly into the QDs (intradot excitation), the broad spectrum is replaced by a narrower emission consisting of three distinct transitions. As shown in Fig. 1, the first peak is separated from the excitation laser energy by 31 meV, the second peak by 36.8 meV, and the third by 66 meV. We attribute the first two peaks to the emission of the QDs with ground-state energies of one InAs LO phonon and one GaAs LO phonon below the laser excitation energy. The third peak may be attributed either to the emission of QDs with a ground-state energy of (one InAs plus one GaAs LO phonon) below the laser excitation energy, or to the ground-state transition (s_v-s_c) after photogeneration of carriers in the (p_v-p_c) excited state, where s_v (s_c) and p_v (p_c) represent the hole (electron) ground and excited states, respectively. In the following we focus our attention on the quantum dot transitions giving rise to the two peaks at higher energy (shown in the inset of Fig. 1). Similar LO-phonon-associated features have already been observed for InAs dots in ensemble and single-dot measurements; see, for example, Refs. 8 and 9. In our case the separation between the three different peaks is independent of the laser energy, which is characteristic for an LO-phonon-related emission process.

In Fig. 2 we compare the time-resolved PL following strictly resonant ($E_{laser}=E_{detection}$ in the left column) and quasiresonant excitation ($E_{laser}=E_{detection}+1$ LO in the right column). After strictly resonant excitation of the sample with Π^X linearly polarized light at $E_{laser}=1.33$ eV [see Fig. 2(a)], we observe for times t longer than 300 ps (for earlier times the signal is obscured by backscattered laser light) a degree of linear polarization $P_{lin}\simeq 70\%$ that remains constant during the exciton lifetime [see Fig. 2(b)]. This confirms that the eigenstates of the quantum dots at zero magnetic field are linearly polarized. Also in the case of quasiresonant excitation ($E_{laser}=1.366$ eV, 36 meV higher in energy than strictly resonant excitation) we observe a constant P_{lin} for $t>300$ ps, and therefore no spin relaxation during the remaining radiative lifetime, although the absolute value of P_{lin} dropped down to 40% [see Fig. 2(e)]. This is comparable to results obtained in CdSe/ZnSe quantum dots for which quasiresonant excitation (one LO phonon above the ground state) also resulted in emission with high $P_{lin}\simeq 65\%$ that remained constant during the radiative lifetime of the exciton.[10] In the remainder of this paper we present the PL measurements following quasiresonant excitation always using the same laser energy of $E_{laser}=1.366$ eV.

Following σ^+ excitation, the two linearly polarized eigenstates $|X\rangle$ and $|Y\rangle$ of the exciton are populated as the laser linewidth is larger than the AEI splitting, resulting in an oscillating PL signal during the first few hundred picoseconds ($t<300$ ps) shown in Fig. 3(a), with a period reflecting the energy splitting between $|X\rangle$ and $|Y\rangle$.[11]

The energy difference between $|X\rangle$ and $|Y\rangle$ varies from dot to dot and due to this inhomogeneous distribution the observed oscillations are damped, as we average the signal over many dots.[5] As the oscillations become damped we find a remaining circular polarization rate of $P_C\simeq 10\%$ for $t>300$ ps [see Fig. 2(f)]. The light emitted from a trion in its ground state is circularly polarized and $P_C\simeq 10\%$ reflects the presence of unintentionally doped QDs in our sample.[12,13] With the trions present we can therefore not reach $P_{lin}=100\%$ as reflected by the highest experimental value of $P_{lin}\simeq 70\%$ [Fig. 2(b)]. Finally the PL signal decays on a time

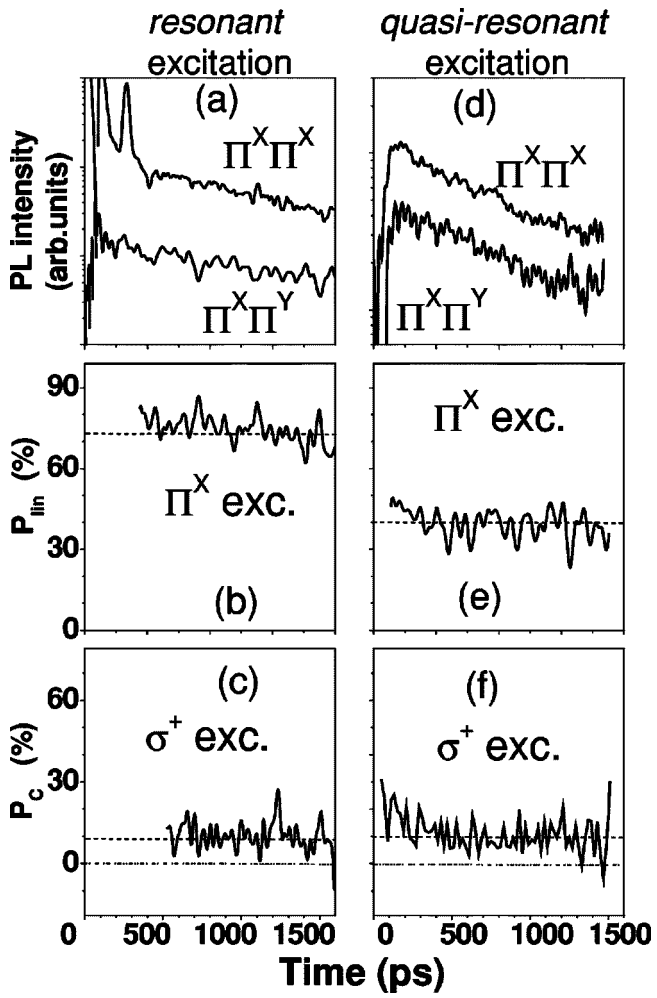

FIG. 2. (a)–(c) Time-resolved photoluminescence following *resonant excitation* ($E_{laser}=1.33$ eV$=E_{detection}$). (a) Photoluminescence versus time following Π^X excitation detected in co- and counterpolarized geometry ($\Pi^X\Pi^X$ and $\Pi^X\Pi^Y$). The signal at early times is obscured by backscattered laser light for the $\Pi^X\Pi^X$ configuration. (b) The degree of linear polarization P_{lin} remains constant during the exciton lifetime at a value of $\approx 70\%$. (c) Following circularly polarized excitation, a polarization degree $P_C \simeq 10\%$ is measured. (d)–(f) *Quasiresonant*, GaAs LO-phonon-assisted excitation ($E_{laser}=1.366$ eV$=E_{detection}+$one GaAs LO phonon). (d) Photoluminescence versus time following Π^X excitation detected in co- and counterpolarized geometry ($\Pi^X\Pi^X$ and $\Pi^X\Pi^Y$). (e) For $t>300$ ps the degree of linear polarization P_{lin} remains constant during the exciton lifetime at a value of 40%. (f) Following circularly polarized excitation, a polarization degree $P_C \simeq 10\%$ is measured.

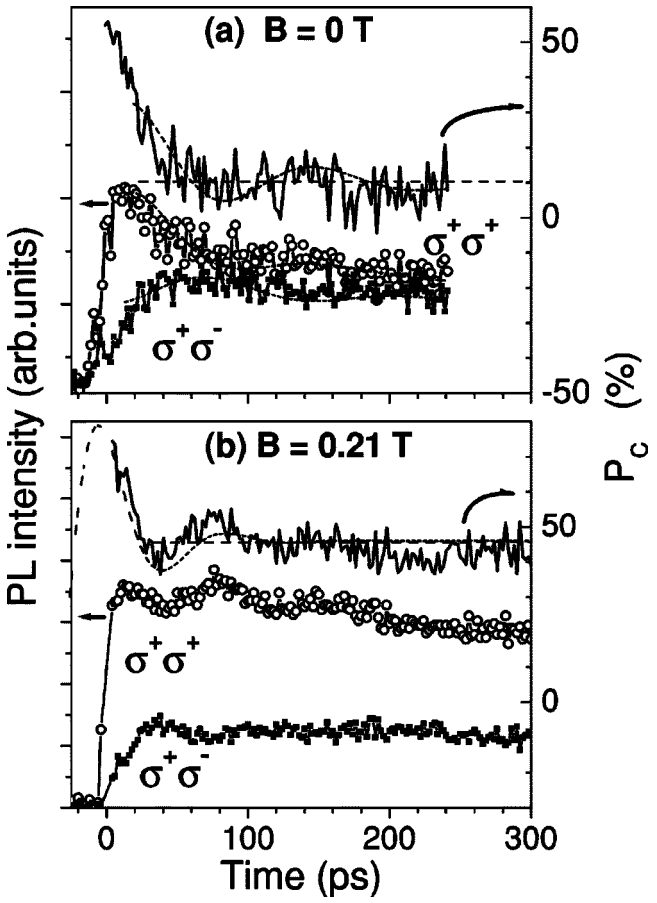

FIG. 3. Time-resolved photoluminescence, co- (open circles) and counterpolarized (solid squares) after quasiresonant σ^+ excitation. The dashed line is a fit of the oscillations using Eq. (3). (a) For $B=0$ oscillations in the circular polarization degree P_C with a period of $T=135$ ps are observed. (b) For $B=0.21$ T the period shortens to $T=90$ ps.

scale of about 1 ns, which is due to the radiative recombination of the exciton.

The oscillation period T and decay time τ were deduced from a simple equation which reproduced the measured oscillations accurately:

$$P_{c/lin} = P_{c/lin}(0)e^{-t/\tau}\cos(2\pi t/T). \quad (3)$$

We obtain an oscillation period of 135 ± 10 ps, corresponding to an average energy splitting $\Delta E_{XY}=30\pm3$ μeV, and a decay time of 30 ps, implying a dispersion of the AEI splitting in the order of 40 μeV. The measured ΔE_{XY} is in agreement with values reported by other groups on similar samples, from single-dot PL and transmission,[5,14,15] differential transmission experiments,[11] and transient four-wave-mixing experiments on ensembles.[16,17] The creation of a coherent superposition of states after strictly resonant excitation results in quantum beats. In our experiment the excitation is quasiresonant and LO-phonon-assisted and the creation of a coherent superposition of states is more complex. We explain the origin of the observed beats as a quantum interference originating from dots in a state $|X\rangle+i|Y\rangle$. Another possible origin of the beats could be the optical interference of photons emitted from dots that are in either state $|X\rangle$ or $i|Y\rangle$. In the following we will argue that quantum interference is the most likely reason for the observed oscillations. We consider two different processes that lead to the quantum dot ground-state PL in our experiment following quasiresonant excitation.

(i) First we consider a second-order absorption process of a photon, assisted by the emission of a LO phonon. In this case each dot will be in either state $|X\rangle$ or $i|Y\rangle$ after excitation, resulting in the emission of photons linearly polarized

as either Π^X or Π^Y. But since the spontaneous emission of a phonon (typically within a time interval[18] of 140 fs) will introduce a random phase difference between the photon emission of the states from different dots, no oscillations of the polarization will be observable in this case. As a result of the second-order absorption process the dots are in either state $|X\rangle$ or $i|Y\rangle$, without any well-defined phase relationship between states in different dots, which would be necessary to create a coherent superposition.[19] This process could be responsible for the drop from $P_{lin} \simeq 70\%$ to 40% linear polarization as observed in the PL experiment when going from strictly to quasiresonant excitation.

(ii) Second, we consider the direct, resonant absorption of a photon by a polaron state of a dot. Using the $|X\rangle$ and $i|Y\rangle$ exciton states, two polaron states can be built, involving one LO phonon and one excited $(p_v\text{-}s_c)$ exciton state. These polaron states are lifetime broadened due to the short lifetime of the LO phonon[20] ($\approx$3 ps) corresponding to an energy uncertainty of 220 μeV. This is larger than the anisotropic exchange energy of $\Delta E_{XY} \simeq 30$ μeV measured; hence the laser excites the two polaron components simultaneously, with a well-defined phase relationship between the $|X\rangle$ and the $i|Y\rangle$ components. As the polaron state disintegrates through the annihilation of its phonon component into two acoustic phonons,[21] the phase between $|X\rangle$ and $i|Y\rangle$ is preserved. Since the phonon annihilation occurs on a time scale much shorter than the beat period $h/\Delta E_{XY}$ the coherence between $|X\rangle$ and $i|Y\rangle$ is maintained. We have therefore created a coherent superposition of states through quasiresonant excitation via a polaron state. We believe this process to be responsible for the observed 40% of linear polarization remaining after quasiresonant excitation. For a more detailed discussion of optical versus quantum coherence see Ref. 19.

In addition to the PL results, quantum beats have been observed in resonant transient dichroism measurements using a pump-probe arrangement. The pump polarization was either σ^+ or σ^- and the probe beam is Π^X linearly polarized. After transmission through the sample, its Π^{X+Y} and Π^{X-Y} components are analyzed. The difference between the intensities of these two components reveals a transient dichroism sensitive to the coherent superposition of the $|X\rangle$ and $i|Y\rangle$ states, initiated by the circular pump pulse. We observe oscillations of the dichroism with a period $T = 130 \pm 10$ ps and a decay time $\tau = 26 \pm 3$ ps (see Fig. 4). This result, obtained in a resonant configuration at zero magnetic field, confirms the values obtained in the time- and polarization-resolved PL measurements after quasiresonant excitation. Obtaining the same period *and* decay time in both experiments is a strong indication that the quantum beats observed are due to the creation of a coherent superposition $|X\rangle + i|Y\rangle$.[22]

A magnetic field applied along the growth axis of the sample (Faraday configuration) was used to investigate the behavior of the excitonic fine structure states split by the magnetic field. At zero magnetic field the optically active eigenstates for neutral dots are $|X\rangle$ and $i|Y\rangle$, as confirmed by our PL measurements of a constant $P_{lin} = 40\%$ after quasiresonant excitation. By increasing the magnetic field, the Zeeman splitting between the $|+1\rangle$ and $|-1\rangle$ states starts to dominate the AEI splitting, and $|+1\rangle$ and $|-1\rangle$ become the optically active eigenstates.[5,23] We have verified this experimentally by measuring P_{lin} and P_C for $t > 300$ ps: for a field $B = 0.4$ T we find $P_{lin} \simeq 10\%$ and $P_C = 60\%$. The conversion from linearly to circularly polarized dot eigenstates is shown in Fig. 5.

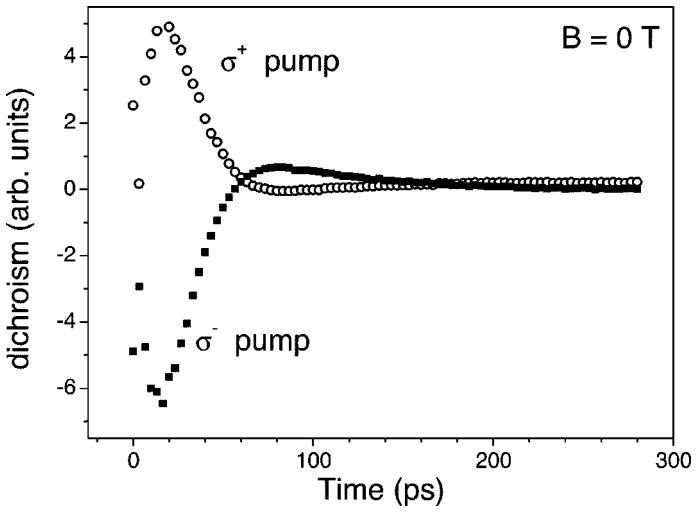

FIG. 4. Resonant ($E_{laser} = E_{detection}$) transient dichroism from pump-probe measurements, for σ^+ (open circles) and σ^- (solid squares) pump polarization.

The circular polarization $P_C \simeq 60\%$ measured at $B = 0.4$ T includes contributions from neutral as well as charged dots as the PL polarization of trions remains circular under an applied magnetic field.[24] By applying a magnetic field we have changed the PL polarization of photons from charge neutral dots from linear to circular and for both orientations the degree of polarization remains strictly constant during the exciton lifetime. In analogy to the quantum beats observed at $B = 0$ for $t < 300$ ps of the circular polarization, after σ^+ excitation we expect quantum beats for $B = 0.4$ T in linear polarization after Π^X excitation and indeed we do observe this type of oscillation, as shown in the time-resolved copolarized and counterpolarized PL in Fig. 6(b) with a period of 65 ps.

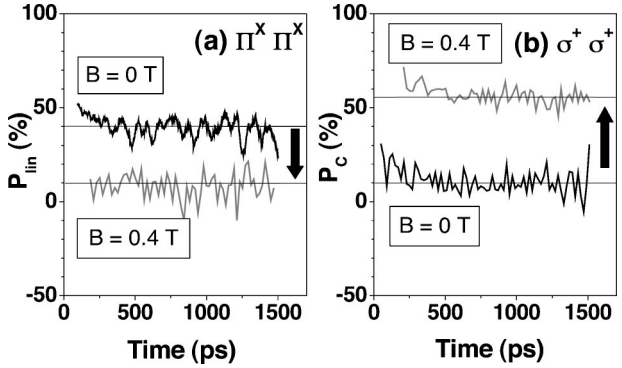

FIG. 5. (a) Π^X excitation and Π^X detection: For $t > 300$ ps the degree of linear polarization P_{lin} remains constant during the exciton lifetime at a value of 40% at $B = 0$ T (black curve). At $B = 0.4$ T P_{lin} is about 10% (gray curve). (b) σ^+ excitation and σ^+ detection: At $B = 0.4$ T for $t > 300$ ps the degree of circular polarization P_C remains constant during the exciton lifetime at a value of 60% (gray curve). For $B = 0$ T P_C is about 10% (black curve).

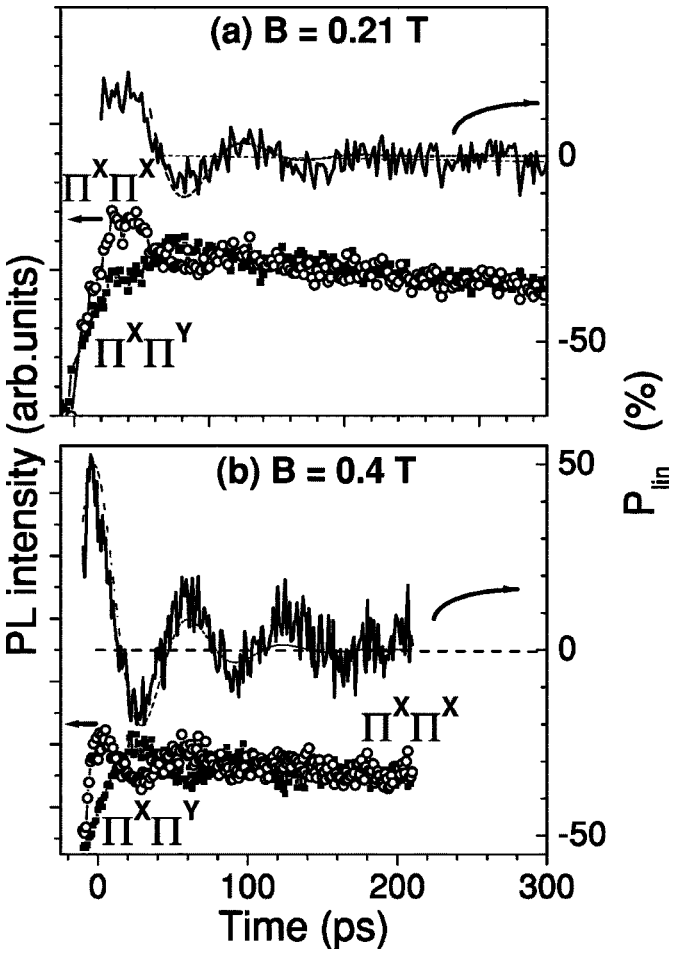

FIG. 6. Time-resolved photoluminescence, co- (open circles) and counterpolarized (solid squares) after quasiresonant Π^X excitation. The dashed line is a fit of the oscillations using Eq. (3). (a) For $B=0.21$ T we observe oscillations in the linear polarization degree P_{lin} with a period of $T=90$ ps. (b) For $B=0.4$ T the period shortens to $T=65$ ps.

For an intermediate field of $B=0.21$ T we observe a beating of the *circular* polarization following *circularly* polarized excitation, as at $B=0$, but with a shorter period of 90 ps [see Fig. 3(b)]. In addition, we observe oscillations of the *linear* polarization following *linearly* polarized excitation with the same period of 90 ps [see Fig. 6(a)].

To fit the dependence of the energy splittings deduced from the oscillation periods as a function of magnetic field, we use the function

$$\Omega = \sqrt{\omega_{exch}^2 + (g_{ex}\mu_B B/\hbar)^2}, \tag{4}$$

which takes the exchange energy $\Delta E_{XY}=\hbar\omega_{exch}$ and the Zeeman splitting $\hbar\Omega_z=g_{ex}\mu_B B$ into account.[5] This is in contrast with recent measurements performed on InP quantum dots in a magnetic field where no influence of the AEI on the Zeeman fine structure could be detected.[25,26] We deduce an effective exciton g factor of $g_{ex}\simeq 2.5$ for the dots investigated here (see Fig. 7). Experimentally, the amplitude decay of the oscillations is the same for zero magnetic field as for

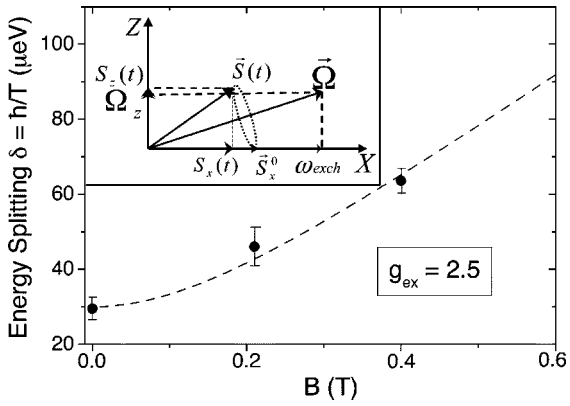

FIG. 7. Dependence of the energy level splitting corresponding to the quantum beat period versus magnetic field. An exciton g factor of 2.5 can be deduced. Inset: Pseudospin formalism.

$B=0.4$ T. This means that no significant additional dispersion is introduced under magnetic field, i.e., the dispersion on the neutral exciton longitudinal g factor from dot to dot is small compared to the dispersion on the anisotropic exchange splitting. The small dispersion of the g factor for neutral excitons has been established previously experimentally by Bayer *et al.*[27] and by Chen *et al.*[28]

The switching from linear to circular exciton polarization through the application of an external magnetic field can be modeled in the framework of an effective pseudospin with $S=\frac{1}{2}$. In this formalism, the exciton states $|1\rangle$ and $|-1\rangle$ and the exciton states $|X\rangle$ and $|Y\rangle$ are equivalent to pseudospins parallel to the Oz and Ox axes, respectively, represented in the diagram in Fig. 7 (inset). As a result, the PL polarization can be written as $P_C=2S_Z$ and $P_{lin}=2S_X$. A circularly polarized excitation is represented by an initial pseudospin $\vec{S}(t=0)$ parallel to the Oz axis. The projection of $\vec{S}(t)$ on the Oz axis yields the circular polarization, which oscillates as a function of time, as shown in Figs. 3(a) and 3(b). Following linearly polarized excitation, the initial pseudospin $\vec{S}(t=0)$ is parallel to the Ox axis and the projection of $\vec{S}(t)$ on the Ox axis yields the linear polarization, which oscillates as a function of time, as can be seen in Figs. 6(a) and 6(b). Using this formalism it becomes clear that for intermediate fields, for comparable AEI and Zeeman splittings, both linear and circular polarizations oscillate with the same period, $T=90$ s for $B=0.21$ T [see Figs. 3(b) and 6(a)]. For $B=0.4$ T the pseudospin is almost parallel to Oz and we do not resolve quantum beats after a circularly polarized σ^+ excitation. The initial modulation amplitude of P_C of $(\omega_{exch}/\Omega)^2\approx 21\%$ (as compared to 49% for $B=0.21$ T) is damped too quickly for the beats to be detectable.

In summary, we have observed spin quantum beats for an ensemble of InAs quantum dots using time-resolved PL experiments after quasiresonant, LO-phonon-assisted excitation and resonant transient dichroism measurements. The polarization and the period of the spin quantum beats can be changed by applying a small external magnetic field. The conversion from linearly to circularly polarized eigenstates can be explained by an interplay between the AEI and the Zeeman splitting. The spin states created at different mag-

netic fields have all been found to be stable during the exciton lifetime, an encouraging result for future attempts of coherent control of these spin states in multiple-pulse experiments.

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