Electron and hole spins in InP/(Ga,In)P self-assembled quantum dots

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The properties of electron and hole spins in InP/(Ga,In)P self-assembled quantum dots are studied through their coherent dynamics using time-resolved Kerr rotation. From these studies information about the g factor and dephasing of the spin excitations is extracted. The electron spin shows a behavior similar to that of electron spins in quantum dots of different material: The g factor is isotropic in the dot plane and with increasing applied magnetic field the spin dephasing accelerates due to variations in the dot ensemble. On the other hand, the hole spin demonstrates a behavior different from other dot systems. Namely, the signal decay on a time scale of about 100 ps does not depend on magnetic field, and the g factor is isotropic in the dot plane. These findings underline previous suggestions that only the electrons are well localized in the InP/(Ga,In)P quantum dots, while the holes are weakly confined and carry bulklike character.

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I. INTRODUCTION

During recent years the coherent dynamics of carrier spins in quantum dots (QDs) have attracted considerable interest due to their long-lived coherence.[1,2] The dynamics are largely determined by the g factors of the confined carriers, which determine the spin precession frequency about a magnetic field. Despite the interest, the understanding of spin excitations in self-assembled QDs is still rather fragmentary, in contrast to the knowledge about the orbital excitations in these systems. Best studied are (In,Ga)As/GaAs quantum dots, for which the electron spins are only weakly affected by the anisotropic confinement, so that in lowest approximation the electron g factor is isotropic, in particular in the QD plane. This behavior is intimately related to the s-type character of the electron Bloch wave function. On the other hand, the hole with a p-type Bloch function has to be treated as a Luttinger spinor that is strongly affected by the anisotropic confinement: Not only is there a strong difference between the g factors along and normal to the growth direction (that occurs similarly in quantum wells grown on [001] substrates), but also in the plane there occurs a strong asymmetry.[3,4] Studies of II-VI self-assembled QDs based on ZnSe or CdTe indicate the same behavior.[5–7]

In the QD structures mentioned so far both electrons and holes are strongly confined. This situation appears to be distinctly different in self-assembled InP/(Ga,In)P QDs. Previous studies indicate that the electrons are strongly localized while the holes are weakly confined or even unconfined, so that they become bound to the dots by the electron Coulomb attraction only.[8–12]

In this paper we study how the confinement conditions influence the electron and hole spins in InP/(Ga,In)P QDs. For that purpose we apply time-resolved pump-probe spectroscopy to measure the Kerr rotation (KR) due to spin precession about a perpendicular magnetic field. From the recorded traces we extract information about the carrier g factors and the spin dynamics in the QD ensemble. For the electrons we find a behavior observed similarly for other dot systems. In contrast, the holes show a surprisingly different behavior. The hole g factor does not exhibit an in-plane anisotropy, and the decay time of the spin coherent signal does not vary with magnetic field. This indicates that hole g-factor variations in the QD ensemble are not the dominant factor responsible for the signal decay, but fast relaxation rather masks the impact of these variations.

II. EXPERIMENT

The sample was grown by metal-organic vapor phase epitaxy on a (100) oriented GaAs substrate tilted by 6° toward the [111]A direction. A 100-nm-thick GaAs buffer layer was deposited onto the substrate at 750 °C and covered by a 100-nm-thick $Ga_{0.51}In_{0.49}P$ layer, grown at 720 °C. Then five stacks of QD layers were deposited being separated by a 5-nm-thick $Ga_{0.51}In_{0.49}P$ spacer from each other. The QDs were formed after deposition of 2.1 monolayers (5 nm) of InP at 710 °C. Finally, the structure was capped by a 30-nm-thick $Ga_{0.51}In_{0.49}P$ layer. The QDs studied here are nominally undoped, however, a small n-type background doping is present due to donor impurities in the band gap of $Ga_{0.51}In_{0.49}P$.[13] The residual electron doping concentration is of the order of 10^{15} cm^{-3}.

For the pump-probe KR experiments an optical parametric oscillator pumped by a mode-locked Ti:sapphire laser was used as excitation source. The laser system generates pulses with a duration of 1.6 ps at a repetition rate of 75.6 MHz, corresponding to 13.2 ns pulse separation. The photon energy is tunable in the range 1.8–1.9 eV. The sample was held in a helium bath cryostat equipped with a superconducting split-coil magnet. Magnetic fields **B** up to 7 T were applied perpendicular to the structure growth direction which we take as z axis, coinciding with the optical axis (Voigt geometry, $\mathbf{B}\perp\mathbf{z}$).

In the KR experiment the laser pulse train was split into a circularly polarized pump beam and a linearly polarized probe beam. Both beams were focused onto the same spot of the sample with a diameter of ∼150 μm. To avoid buildup of dynamic nuclear polarization, the helicity of the pump beam was modulated at 50 kHz frequency using a photoelastic modulator. The pump pulse excites carriers with spins polarized along the *z* axis. The subsequent coherent spin dynamics in form of Larmor precession about the magnetic field **B** is measured by the Kerr rotation angle Θ_{KR} of the polarization plane of the probe beam reflected from the sample. To detect Θ_{KR} a homodyne technique based on phase-sensitive balanced detection was used.[14]

Time-integrated and time-resolved photoluminescence (PL and TRPL, respectively) were measured with standard setups. For that purpose the sample was mounted in a helium flow cryostat at a temperature $T = 5$ K. As an excitation source the frequency-doubled output of the mode-locked Ti:sapphire laser was used providing optical pulses with a photon energy of 3.3 eV. The photoluminescence (PL) emission was dispersed by a 0.3 m monochromator and detected either by a silicon charge-coupled-device camera or a synchroscan streak camera with a S20 photocathode. The overall time resolution of the TRPL setup was 15 ps.

III. RESULTS AND DISCUSSION

A. Quantum dot photoluminescence

The low temperature PL spectrum of the studied InP/$Ga_{0.51}In_{0.49}P$ QDs is shown in Fig. 1(a). The high intensity peak centered at 1.84 eV corresponds to radiative recombination of neutral or charged exciton (trion) complexes confined in the dots. The inhomogeneity of the QD ensemble results in a substantial broadening of the PL emission band of ∼80 meV. The additional peak in the PL spectrum at ∼1.96 eV energy is related to exciton recombination in the $Ga_{0.51}In_{0.49}P$ barriers.

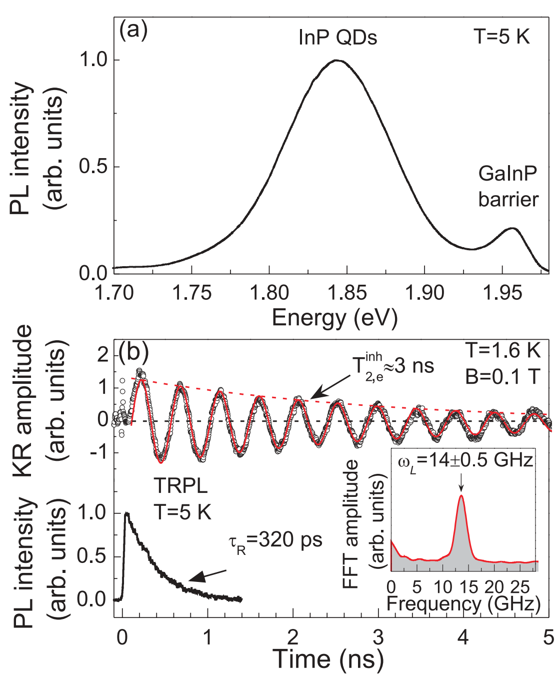

FIG. 1. (Color online) (a) Photoluminescence spectrum of the InP/$Ga_{0.51}In_{0.49}P$ QDs measured for nonresonant pulsed excitation at 3.3 eV photon energy. (b) Kerr rotation trace for the InP/$Ga_{0.51}In_{0.49}P$ QDs recorded at $B = 0.1$ T together with the corresponding fast Fourier transform (inset). Laser photon energy 1.86 eV with pump power 85 W/cm^2 and probe power 3 W/cm^2. Bottom: Example of the TRPL intensity profile measured at 1.86 eV for 3.3 eV photon energy of the excitation laser with power density 0.07 W/cm^2.

The QD recombination dynamics was measured under nonresonant pulsed excitation using an excitation power density of 0.07 W/cm^2 in order to provide on average much less than one photogenerated electron-hole pair per dot. The TRPL intensity traces for a given photon emission energy show a resolution-limited rise time and a monoexponential intensity decay, see Fig. 1(b) bottom.

The PL decay time τ_R varies only weakly with transition energy around 320 ± 15 ps in the range 1.82–1.9 eV. This value is slightly less than the reported exciton radiative lifetime of 350–450 ps measured on a single InP/$Ga_{0.51}In_{0.49}P$ QD,[15] and agrees well with the 280–330 ps exciton recombination lifetime deduced from TRPL experiments performed on an ensemble of InP/$Ga_{0.5}In_{0.5}P$ QDs.[16,17] This also shows that electronic coupling between the QDs in the five-layer stack, which may be expected for only 5-nm-thick $Ga_{0.51}In_{0.49}P$ spacers between the dot layers, is not relevant. Most likely such coupling effects are negligible because of off-resonance between the confined carrier levels in the different layers at least for the vast majority of dots in the ensemble.

B. Time-resolved Kerr rotation experiment at low magnetic field

Let us turn to the results of the KR studies on the InP/$Ga_{0.51}In_{0.49}P$ QDs. At the moment of excitation the circularly polarized pump pulse generates electrons and holes in the QDs with spins oriented along the optical axis.[18] The background doping results in a fraction of QDs charged by electrons, while another fraction in the ensemble is uncharged. For the charge neutral QDs the spin coherence is carried by the photoexcited exciton, and therefore cannot persist longer than the exciton lifetime. Among the charged quantum dots the singly charged ones dominate due to the low background doping level. In these dots the pump excitation leads to a long-lived spin polarization beyond the lifetime of the charged exciton.[18]

This time evolution determines the appearance of the KR traces, where after initialization the carrier spins precess about the magnetic field as time elapses resulting in coherent oscillations of the Kerr signal. Figure 1(b) (circles) presents an example of a KR trace recorded at $B = 0.1$ T. The photon energy of pump and probe beams are set to 1.86 eV, resonant with the QD ground state transition. The KR amplitude reflects the coherent evolution of injected or resident carrier spins in the optically excited subset of InP/$Ga_{0.51}In_{0.49}P$ QDs. KR signal can be observed over delay times up to 5 ns relative to the pump pulse arrival. This clearly indicates that dominant long-lived spin coherence due to resident electrons is generated. Photoexcited carriers have disappeared 1 ns after the pump pulse due to radiative recombination occurring on characteristic time scale of 320 ps.

With increasing delay the KR amplitude, however, decreases significantly which does not correspond to a loss of coherence, but is a result of ensemble dephasing due to inhomogeneities. At low magnetic fields the inhomogeneity might be either related to g-factor variations in the different quantum dots or variations of the effective magnetic field from the nuclear spin fluctuations in the QDs.

To reassess the supposed origin of the observed KR signal at $B = 0.1$ T we extract the Larmor spin precession frequency ω_L from the data. Because of the long-lived signal ω_L can be reliably evaluated through the fast Fourier transform (FFT) of the KR trace. The result is presented in Fig. 1(b) (inset), from which we obtain $\omega_L = 14 \pm 0.5$ GHz. The Larmor precession frequency is related to the Zeeman splitting of the spin states through $\hbar\omega_L = g\mu_B B$, where $\hbar$ is the Planck's constant and μ_B is the Bohr magneton. From this we evaluate a Lande g factor $g \approx 1.61$, which is in the range of expected electron g factors in InP QDs (see also below).

Once the Larmor precession frequency is known, it is possible to determine accurately the decay time of the spin coherent signal by fitting the KR trace with an exponentially damped harmonic function of time, assuming the following form:

$$\Theta_{\mathrm{KR}}(t) = \Theta_{\mathrm{KR}}(0) \exp\left(-\frac{t}{T_{2,e}^{\mathrm{inh}}}\right) \cos(\omega_L t), \qquad (1)$$

where $\Theta_{\mathrm{KR}}(0)$ is the KR amplitude at the moment of pump pulse arrival, and $T_{2,e}^{\mathrm{inh}}$ is the decay time associated with electron spin dephasing (see below). A fit to the experimental KR trace is shown by the red solid line in Fig. 1(b). The obtained $T_{2,e}^{\mathrm{inh}} \approx 3$ ns at $B = 0.1$ T is comparable to the scaled dephasing time of $g_e T_{2,e}^{\mathrm{inh}} = 1.7$ ns measured in a Hanle experiment for $\mathrm{InP/Ga_{0.5}In_{0.5}P}$ QDs.[19] It is also similar to the $T_{2,e}^{\mathrm{inh}}$ values reported for other QD systems: $T_{2,e}^{\mathrm{inh}} = 2$ ns at $B = 0.25$ T was measured for (In,Ga)As/GaAs QDs by Faraday rotation,[20] $T_{2,e}^{\mathrm{inh}} = 5.6$ ns at 0.25 T was reported for CdSe/(Zn,S)Se self-assembled QDs measured by Kerr rotation.[21]

C. Time-resolved Kerr rotation experiment vs magnetic field

In the following we consider the carrier spin dynamics at different magnetic field strengths. Figure 2(a) shows KR traces in the time delay range up to 1 ns for magnetic fields up to 7 T. At $B = 0$ the light induced spin polarization decays

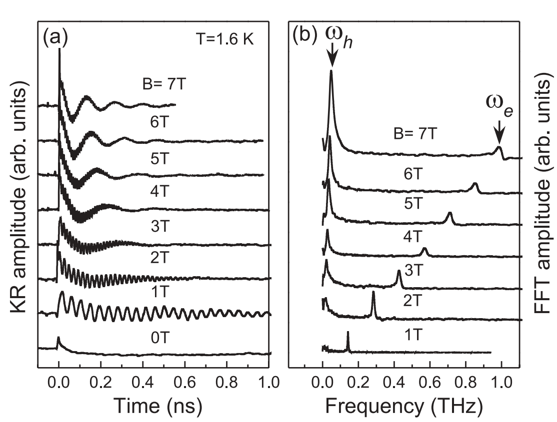

FIG. 2. (a) KR signals measured at different magnetic fields in the $\mathrm{InP/Ga_{0.51}In_{0.49}P}$ QDs. Pump power 85 W/cm^2, probe power 3 W/cm^2, laser photon energy 1.86 eV. (b) FFT analysis of the KR traces from (a).

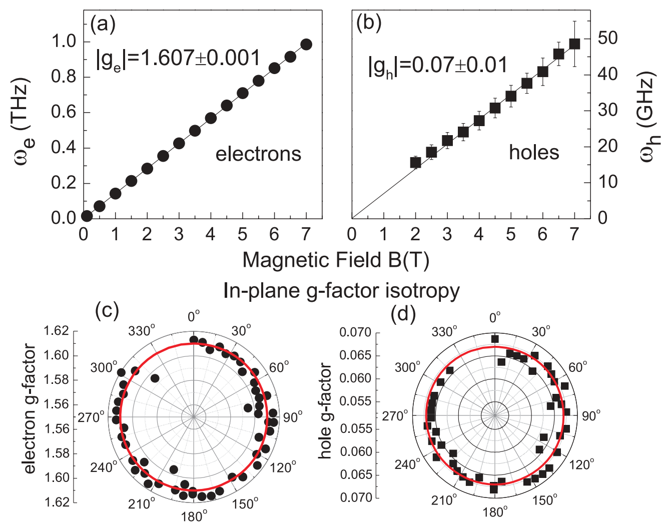

FIG. 3. (Color online) (a) Electron and (b) hole Larmor precession frequencies vs magnetic field, from which their g factors in the $\mathrm{InP/Ga_{0.51}In_{0.49}P}$ QDs are obtained. Lower panels give the dependence of the (c) electron and (d) hole g factors in the QD plane.

with two components. The fast one of about 50 ps decay time, shorter than the exciton lifetime, can be related to spin relaxation processes within the exciton or trion complexes. The decay time of the second component of ~3 ns is similar to the dephasing time at $B = 0.1$ T. At higher magnetic fields a complex spin beating pattern appears. The KR signal consists of two oscillating components with very different periods as confirmed by the FFT analysis presented in Fig. 2(b). The FFT spectra show two well separated peaks. The high frequency peak ω_e and the resulting g factor correspond to the resident electron contribution discussed above. For the hole a significantly smaller in-plane hole g factor is expected than for the electron. Therefore the peak at smaller Larmor frequency in the FFT spectrum, denoted by ω_h, is tentatively attributed to hole spin precession.

Figure 3 shows the linear variation of the electron [Fig. 3(a)] and hole [Fig. 3(b)] spin precession frequencies ω_e and ω_h as functions of magnetic field. The linearity, in particular of the hole g factor, indicates that valence band mixing does not change notably in the applied magnetic field range. From the slopes the g factors of electron and hole along the applied field direction can be determined with high accuracy: $|g_e| = 1.607 \pm 0.001$ and $|g_h| = 0.07 \pm 0.01$.

In order to compare the measured electron g factor with those reported in literature, we choose a presentation in which g_e is plotted vs the energy of the optical transition. According to the Roth-Lax-Zwerdling relation[22] the electron g factor is mostly determined by the energy gap. This relation, that was originally derived for bulk systems, can be successfully extended to quantum well and quantum dot heterostructures, where the quantum confinement energies increase the band gap energy (see, e.g., Refs. 23 and 24). In Fig. 4 the value for the studied $\mathrm{InP/Ga_{0.51}In_{0.49}P}$ QDs is shown by the open star, whereas the literature data (Refs. 29–35) are given by other symbols denoted in the figure legend.

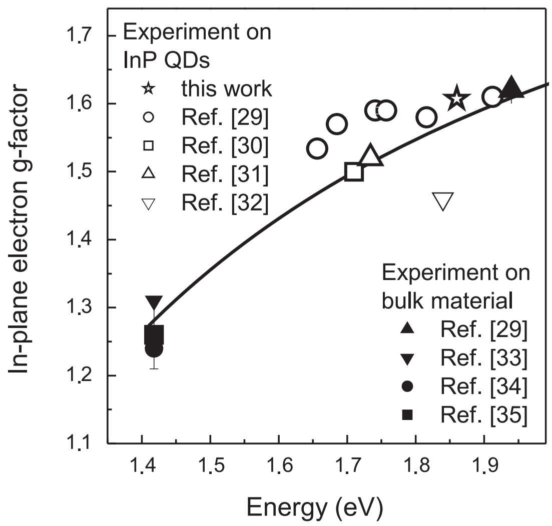

FIG. 4. Electron g factor as function of optical transition energy. Open symbols indicate values measured for InP/(Ga,In)P QDs. Values for InP and $Ga_{0.52}In_{0.48}P$ bulk material are represented by solid symbols. The g-factor dispersion for (Ga,In)P calculated after Ref. 22 is shown by the solid line.

The curve shown by the solid line in Fig. 4 was calculated after the Roth-Lax-Zwerdling relation[22]:

$$g_e = g_0\left[1 - \frac{P^2\Delta_0}{3E_g(E_g+\Delta_0)}\right]. \quad (2)$$

Here g_0 is the free-electron Lande factor, P^2 is the squared interband matrix element, Δ_0 is the valence-band spin-orbit splitting, and E_g is the band-gap energy. The parameters used for the calculations are $P^2 = 21.6$ eV, and $\Delta_0 = 0.108$ eV, taken after Ref. 25. The g_e value measured in this work agrees well with the predicted trend and agrees with other experimental data reported in the literature.

The in-plane hole g factor cannot be easily compared to the different literature values. g_h is expected to be not only dominantly affected by the QD band-gap energy, but also depends strongly on several other parameters including dot shape, strain, and valence band confinement.[26–28] Only limited information on these parameters is available for the QDs studied here.

Additional insight into the g-factor tensors can be obtained by studying the g factor in-plane anisotropy for electrons and holes. Due to the s-type atomic orbitals g_e is expected to be almost isotropic. For the holes, on the other hand, with their p-type orbitals a strong anisotropy is expected, as demonstrated recently for (In,Ga)As/GaAs self-assembled QDs.[3,4] For these systems the electron g factor is isotropic in the QD plane, however, the hole g factor shows variations up to ∼50% in this plane.

Data on the in-plane g-factor anisotropy for the studied $InP/Ga_{0.51}In_{0.49}P$ QDs are collected in Fig. 3, for which the sample was rotated relative to the magnetic field. In Fig. 3(c) the dependence for the electrons is shown: As expected we find an in-plane isotropy within the experimental accuracy. However, for the hole a behavior very different from the one in (In,Ga)As/GaAs QDs is found, as seen in Fig. 3(d). Here also the hole shows symmetry in the dot plane. This unexpected hole g-factor behavior indicates that the confinement of the holes is different from those in "standard" self-assembled dot structures.

The confinement conditions are determined by the valence band offsets and the strain. Neglecting strain, the valence band offset between InP and $Ga_{0.51}In_{0.49}P$ is negative (i.e., the hole has lower energy in $Ga_{0.51}In_{0.49}P$) and can reach −45 meV,[8] much less than the valence band offset in (In,Ga)As/GaAs QDs. Strain typically increases the band gap of the QD materials and shifts it towards the band gap of the barriers. In conjunction with the quantization energy of the hole levels this suggests that the confinement of the hole levels is weak, if there is a confinement at all. In case of an exciton complex the hole confinement will be significantly contributed by the Coulomb attraction of the electrons trapped in the dots. This weak confinement apparently is reflected by the hole g factor, which is not influenced notably by confinement potential asymmetries, resulting in its isotropic in-plane behavior.

D. Decay of the electron and hole spin coherent signal

If the hole spin is weakly confined which could be caused by the electron Coulomb attraction only, this should also be reflected by the spin dynamics. Only when well confined three dimensionally a spin is effectively protected from relaxation mechanisms involving, for example, spin-orbit coupling. For low magnetic fields below 2 T only long-lived electron spin precession is seen in the KR traces. In the traces for magnetic fields exceeding 2 T electron and hole contributions are involved (see Fig. 2) so that it is possible to get information about both the electron and hole spin decay. To that end, the KR traces from Fig. 2(a) are fitted by a superposition of two damped harmonics:

$$\Theta_{KR}(t) = \sum_{i=e,h} \Theta_{KR,i}(0)\exp\left(-\frac{t}{T_i}\right)\cos(\omega_i t). \quad (3)$$

Here $i = e,h$ is the electron or hole component, $\Theta_{KR,i}(0)$ is the corresponding KR amplitude, ω_i is the Larmor precession frequency taken from FFT analysis, and T_i is the decay time of the electron or hole signal, respectively.

The results of this analysis for the T_i of electron and hole are presented in Fig. 5 as functions of magnetic field. For the electron a strong reduction in T_e is observed starting from 3 ns at 0.1 T down to 110 ps at 7 T. If these times are converted into rates ($1/T_e$) we find a linear dependence of this rate on magnetic field [see the inset of Fig. 5(a)]. In general the decay time $T_i, i = e,h$ is contributed by the coherence time of individual spins $T_{2,i}$, the inhomogeneous dephasing time from inhomogeneity of the spin ensemble $T_{2,i}^{\mathrm{inh}}$, and the lifetime of the spin excitation τ_R[14]:

$$\frac{1}{T_i} = \frac{1}{T_{2,i}} + \frac{1}{T_{2,i}^{\mathrm{inh}}} + \frac{1}{\tau_R}. \quad (4)$$

For resident electrons the last term on the right-hand side vanishes. The characteristic $1/B$ dependence of T_e clearly indicates that the inhomogeneity of the electron g factor in the ensemble plays the dominant role. A variation of the electron g factor Δg_e is translated into a variation of the precession

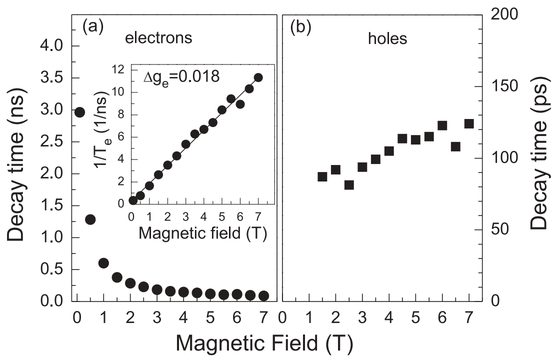

FIG. 5. Decay times of the (a) electron and (b) hole spin coherent signal vs magnetic field. Inset in (a): Corresponding magnetic field dependence of electron spin dephasing rate. Pump power 85 W/cm^2, probe power 3 W/cm^2, laser photon energy 1.86 eV.

frequency $\Delta\omega_e = \Delta g_e \mu_B B/\hbar$, and therefore into a B-linear dependence of the decay rate, or a $1/B$ dependence of the decay time. From a linear fit to $1/T_e$ we obtain a Δg_e of 0.018 for the optically excited spin ensemble, which is about 1% of the average g factor.

The hole spin decay time vs magnetic field is presented in Fig. 5(b). Obviously it shows a radically different behavior from the electron spin in that it does not decrease, but is mostly constant or even increases slightly. From this finding strong ensemble inhomogeneities of the g factor which we would expect for confined spin excitations can be excluded. The observed decay time around 100 ps is also considerably shorter than the exciton lifetime, which is equal to the lifetime of the photoexcited hole, so that also the third term in Eq. (4) cannot be the dominant contribution. Instead, true spin relaxation by scattering has to occur beforehand, reassuring that the hole spin is not well protected from spin relaxation. As the results discussed so far hint at quite weakly confined holes, also spin-orbit related relaxation might be relevant for holes. Indeed hole spin decay times around 100 ps appear realistic for only weakly confined or quasi-free hole spins: No data on hole spin relaxation were reported for (Ga,In)P crystals. For bulk GaAs and InP relaxation times of 110 and 100–200 fs were measured.[36,37]

Localization generally tends to increase carrier spin relaxation times. Information for InP/(Ga,In)P QWs has not been established. In GaAs/(Al,Ga)As quantum wells, however, the hole spin relaxation ranges from 4 ps[38] to hundreds of ps[39] and further up to 70 ns,[40] increasing steadily with degree of localization and exceeding by far the bulk values. The times measured by us fall into the range of these times.

IV. CONCLUSIONS

We have studied the electron and hole spins in InP/(Ga,In)P quantum dots, for which a peculiar confinement situation was observed, that is distinct from the strong electron and hole confinement in other QD systems such as (In,Ga)As/GaAs. While the electrons are well confined, the confinement for the holes in the valence band is rather weak. This weak confinement of the holes leads to considerably different hole spin properties, namely a g-factor isotropy in the QD plane and a spin relaxation time in the order of 100 ps, significantly shorter than the recombination time of the excitons of which the holes are part of. This shows that the weak confinement does not protect the hole spins from efficient scattering during their lifetime.

ACKNOWLEDGMENTS

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