Effect of nuclear quadrupole interaction on spin beats in photoluminescence polarization dynamics of charged excitons in InP/(In,Ga)P quantum dots

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The spin dynamics of positively (X^+) and negatively (X^-) charged excitons in InP/$In_{0.48}Ga_{0.52}P$ quantum dots subject to magnetic field is studied. We find that a characteristic feature of the system under study is the presence of nuclear quadrupole interaction, which leads to stabilization of the nuclear and electron spins in a quantum dot in zero external magnetic field. In detail, the nuclear quadrupole interaction leads to pinning of the Overhauser field along the quadrupole axis, which is close to the growth axis of the heterostructure. The nuclear effects are observed only when resident electrons are confined in the quantum dots, i.e., for X^- trion photoexcitation. The presence of X^- and X^+ trion contributions to the photoluminescence together with the quadrupole interaction significantly affects the dynamics of optical orientation in Voigt magnetic field. In the absence of dynamic nuclear spin polarization the time evolution of the photoluminescence polarization is fitted by a form which describes the electron spin relaxation in "frozen" nuclear field fluctuations. In relatively large external magnetic fields exceeding 60 mT good agreement between theory and experiment is achieved.

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I. INTRODUCTION

Quantum dots (QDs) are promising objects for use in spintronics devices [1,2] because the motion-related mechanisms of carrier spin relaxation that are important in bulk [3] are suppressed in QDs [4]. The practical requirement of long-lived carrier spin coherence calls for studying effects that stabilize spins in nanostructures. Here we investigate one such effect, the nuclear quadrupole interaction (QI) [5].

A nucleus with a spin exceeding 1/2 has a nonzero electric quadrupole moment [6]. The presence of an electric field gradient across the nucleus leads to the QI. For example, in bulk (Al,Ga)As semiconductors an electric field gradient appears when the gallium ions are replaced by aluminum ions [7]. In the case of InP/(In,Ga)P QDs, the field gradient is caused by a significant lattice deformation which is generated by the large mismatch of InP and (In,Ga)P lattice constants, reported to be 3.7% between InP and $In_{0.485}Ga_{0.515}P$ in Ref. [8]. The deformation occurs at the heterointerface, while the deformation axis is aligned with the QDs growth axis (in our case the [001] axis). The QI affects the spin state of the nucleus, but the projection of the nuclear spin on the main axis of the QI [001] is preserved. If the strength of the electron and nuclear spins hyperfine interaction is significant, the QI will also affect the electron spin. This effect can be probed by means of polarized photoluminescence (PL) as used in the present study.

The dynamic nuclear polarization (DNP) in the presence of strong QI was previously studied in bulk (Al,Ga)As [7]. Also, the effect of the QI on the spin systems of electrons and nuclei in InP/(In,Ga)P QDs was observed [5,9]. Steady-state magnetic field studies of the negatively charged exciton (X^- trion) PL polarization were carried out in Ref. [5], and the dynamics of nuclear spin polarization under circular polarized excitation in singly charged and neutral quantum dots in Faraday magnetic field were studied in Ref. [9].

It should be noted that unlike for positively charged excitons (X^+ trions) [10] and neutral excitons [11], the circular polarization dynamics of the negatively charged exciton PL does not exhibit oscillations in Voigt magnetic field as reported for InP/(In,Ga)P QDs in Refs. [11,12]. The absence of these oscillations is discussed in Sec. IV B. In the case of the neutral excitons, the anisotropic exchange interaction of the electron and hole spins [13] becomes relevant in the presence of anisotropy in the QD plane. As a result, the spin relaxation time of the carriers is significantly reduced [14]. Thus, studying the optical orientation dynamics in Voigt magnetic field is most informative for the X^+ trion PL.

In this paper we study the effect of nuclear quadrupole interaction on the spin dynamics of an ensemble of InP/(In,Ga)P QDs that are fractionally positively charged (X^+), negatively charged (X^-), and charge neutral. The PL circular polarization dynamics and the steady-state PL polarization subject to magnetic field applied in either Voigt or Faraday geometry are investigated. In the optical orientation dynamics in Voigt magnetic field pronounced spin beats are observed, which we study in the presence of DNP and QI. In particular, we find that the PL circular polarization oscillates not around the zero value, as typically observed [15,16], but around a polarization contribution that monotonically decays with time. We show that this behavior can be explained by the simultaneous contributions of X^- and X^+ trions to the PL.

In order to quantitatively describe the PL polarization dynamics, it is necessary to determine whether the spin relaxation takes place in the limit of long [17] or short [3] correlation time: It is known that any mechanism of spin

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relaxation can be considered in terms of effective magnetic field fluctuations acting on the spin. An important characteristic of these fields is the correlation time τ_c, the period in time during which the field fluctuations remain unchanged. There are two extreme limits of spin relaxation. The limit of long correlation time ("frozen" fields) corresponds to the condition $\Omega_f \tau_c \gg 1$, where Ω_f is a spin precession frequency in a field fluctuation. The condition $\Omega_f \tau_c \ll 1$ corresponds to the limit of short correlation time. We show that in the studied QDs the electron spins relax in nuclear field fluctuations with a long correlation time. The time dependencies of the PL polarization (in the absence of DNP) for different magnetic field strengths are described within the approach developed in Ref. [17].

II. EXPERIMENTAL DETAILS

The single layer of lens-shaped, self-organized InP quantum dots embedded in a $In_{0.48}Ga_{0.52}P$ matrix was grown by metal-organic vapor-phase epitaxy on a (001) GaAs substrate. The QDs have a bimodal size distribution: The dots in one group have average sizes of about 100×5 nm^2 (diameter $\times$ height), and those in the second group have sizes of 133×20 nm^2. The QDs were covered with a 40-nm $In_{0.48}Ga_{0.52}P$ cap layer. No wetting layer is formed in these samples. A detailed description of the structure is given in [18], denoted there as sample (i).

Studies of the PL intensity and polarization were carried out in both the continuous-wave mode (cw) and pulsed regime (PR) with time resolution. The sample was placed in a cryostat with liquid helium at a temperature of 2 K (cw) or with helium vapor at a temperature of 6 K (PR). The external magnetic field B was generated by an electromagnet ($B = 0$–250 mT; cw) or superconducting coils ($B = 0$–400 mT; PR). The PL was excited by Ti:sapphire lasers (1.77-eV central photon energy) with a power density of about 75 W/cm^2 operated in continuous wave or pulsed mode. In the latter case, optical pulses with a duration of 150 fs were generated by a self-mode-locked oscillator at a repetition frequency of 75 MHz. The laser light was circularly polarized, and its direction approximately coincided with the sample growth axis. The PL was collected in "reflection" geometry, and the degree of its circular polarization, which is defined by

$$\rho_c = \frac{I^+ - I^-}{I^+ + I^-}, \tag{1}$$

was measured, where I^+ and I^- are the intensities of the PL components whose polarization coincides with and is opposite to the exciting light polarization, respectively. We note that when PL is excited with circularly polarized light, dynamic polarization of nuclear spins may occur through the hyperfine interaction with the spin-polarized electrons. When it was required to exclude DNP, a photoelastic modulator (cw) was placed in the excitation path, modulating the polarization of light between σ^+ and σ^- at a frequency of 26.61 kHz. In the PR studies an electro-optic modulator (EOM) was used (16 kHz). Due to the modulation the fast changes in the direction of the electron spin orientation prevent the buildup of DNP [7]. Finally, after passing through a double- (cw) or single-grating (PR) monochromator the PL was detected with an avalanche photodiode (cw) or with a streak camera (PR). In the latter case, the setup time resolution was about 30 ps.

In the cw regime, the PL intensities were measured using a two-channel photon counting unit. In the PR regime, the required σ^+ or σ^- polarization was selected manually in the detection path, and the corresponding transients $I^+(t)$ or $I^-(t)$ were accumulated. Here, the record time range corresponded to a window of about 2 ns, and accumulation was synchronized with the repetition frequency of the laser (75 MHz). When the excitation polarization was modulated, the EOM was synchronized with the streak camera using a blanking unit. The streak camera blanking unit allowed us to provide an additional time filtering of the recorded transients on a slow microsecond timescale. Here, the PL time dependencies in a 2-ns window were measured only for less than half of the EOM period (less than 20 μs), when the excitation polarization was constant (σ^+). Therefore, the PL polarization degree was analyzed only during the specified time interval (σ^+ excitation).

III. EXPERIMENTAL RESULTS

The excitation photon energy (1.77 eV) is smaller than the band gap of the $In_{0.48}Ga_{0.52}P$ barrier (1.96 eV). As a result, the carriers are generated in excited QDs states. The two PL spectral bands in Fig. 1(a) correspond to the two characteristic QD sizes [18]. The relatively small dots give rise to the band with a maximum at 1.75 eV, while the large dots give rise to the band at 1.63 eV. In this work, we focus on the small QDs. All measurements [except the spectrum in Fig. 1(a)] were performed at a detection photon energy of 1.75 eV. The PL of the small QDs has a significant degree of circular polarization (40%–50%) in zero magnetic field [Figs. 1(a) and 1(b)], which is a signature for X^+ or X^- trion PL [19]. We note that for these quantum dots optical orientation of the neutral exciton PL is almost absent [14].

Let us compare the magnetic field dependencies of the steady-state PL polarization in the case of excitation with constant and modulated polarization [Fig. 1(b)]. Without modulation the half width at half maximum (HWHM) of the Hanle curve is three times larger (50 vs 16 mT) compared to the case with modulation; the polarization in zero magnetic field is also larger (48% vs 41%), and the curve of polarization recovery in Faraday field is asymmetric with respect to an inversion of the field sign. Here, the Hanle curves were fitted by single Lorenz curves for HWHM determination. Such a drastic difference between the magnetic field dependences of the polarization under excitation with constant and modulated polarization provides clear evidence for the presence of DNP in the case of constant excitation and for the absence of DNP in the case of modulation. We recall that the buildup time of DNP is significantly longer than the modulation period of about 50 μs, and therefore, the DNP is absent [7]. The scenario observed in Voigt field deviates from the typical situation where the Overhauser field narrows the Hanle curve, enhancing the effect of the external field and thus increasing the depolarization rate [7]. To describe the observed effects, it is straightforward to assume that the DNP takes place also in zero external magnetic field. As a result, the effective nuclear field stabilizes the electron spin. We note that the spin

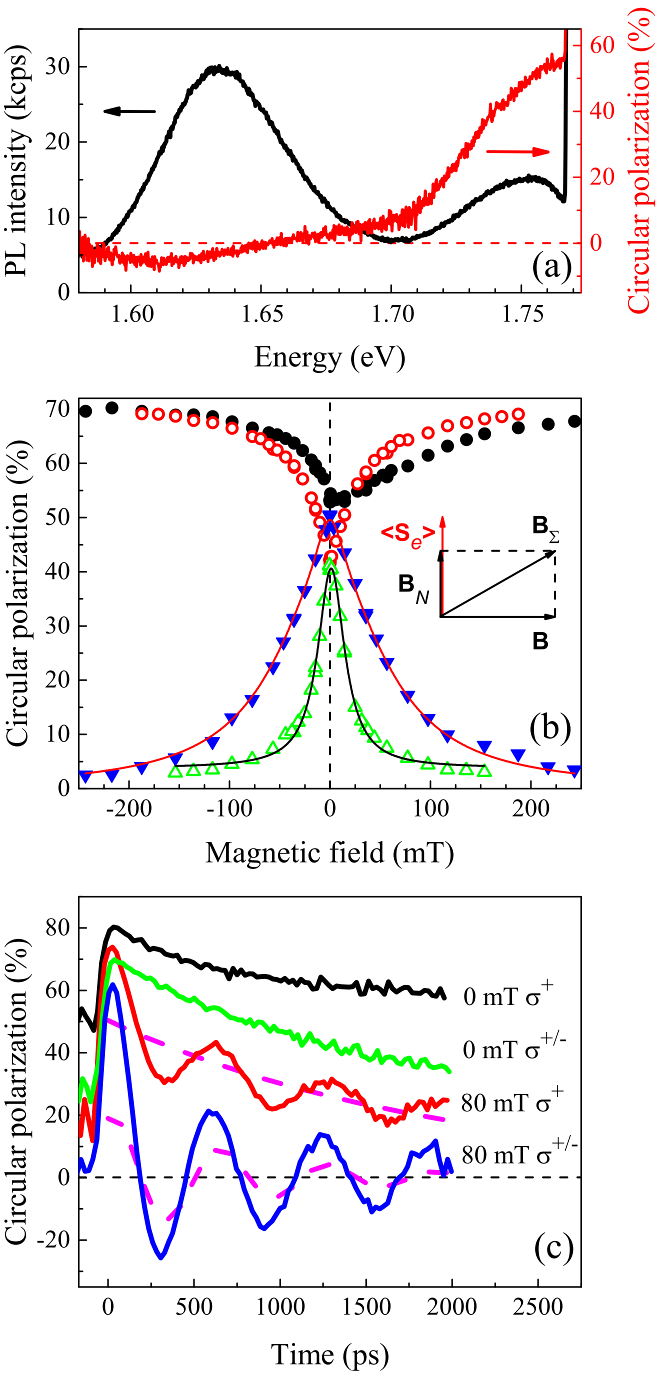

FIG. 1. (a) Spectra of the PL intensity (black line) and circular polarization (red line). (b) Dependence of the PL circular polarization degree on magnetic field in Voigt (triangles) and Faraday (circles) geometries. The open (solid) symbols give the results obtained in the absence (presence) of DNP. The black (red) line gives the fitting by one (two) Lorentz curves in the case of the absence (presence) of DNP. The inset sketches the orientation of the mean electron spin $\langle \mathbf{S}_e \rangle$ and the magnetic fields relative to each other. (c) Dynamics of the PL circular polarization. Solid lines represent measured dependencies in zero magnetic field and in a Voigt field of 80 mT. For each field value the curves with and without DNP are presented. Dashed lines show curves obtained by fitting of the experimental data, the sum of which describes well the 80-mT curve obtained in the presence of DNP.

lifetime of the resident electrons in the sample is much shorter ($\leqslant$ 100 ns) than the modulation period of excitation polarization (about 50 μs) [12]. Thus, the modulation will not reduce the efficiency of the resident electron spin pumping if the nuclear effects are not considered. In addition, it can be assumed that even in the presence of the Voigt field, the Overhauser field is oriented along the growth axis of the heterostructure. In this case, the Voigt field acting on the electron spins has to overcome the nuclear field in order to depolarize the PL. The asymmetry of the dependence in Faraday field arises from the fact that for one sign of the external field the nuclear field enhances it, while for the other sign the nuclear field reduces it. The reason for stabilization of the nuclear spins along the growth axis of the QDs is the nuclear quadrupole interaction caused by the lattice deformation, as discussed in Sec. IV A.

Let us compare the time dependences of the PL circular polarization in the presence and absence of DNP [Fig. 1(c)]. For the latter case, we recall that the polarization of the exciting laser light was modulated in order to exclude DNP. At zero magnetic field, in the presence of DNP the PL polarization is larger than in the absence of DNP (maximum is 80% vs 70%). As mentioned above, even in zero field the electron spins are stabilized by the nuclear field. When the external magnetic field is switched on in Voigt geometry, pronounced oscillations in the dynamics of the PL polarization are observed. The oscillations correspond to the Larmor precession of the electron spin contributing to the X^+ trion. In the presence of dynamic polarization of the nuclear spins, the PL polarization oscillates not around the zero value, but around some finite polarization contribution which monotonically decays with time. In the polarization dynamics we therefore distinguish between the "monotonically decaying" and "oscillating" (around zero polarization) contributions, which together form the experimentally measured curve [dashed lines in Fig. 1(c)]. The existence of the monotonically decaying contribution to the PL polarization can be interpreted by assuming that the Overhauser field is pinned along the QD growth axis. In this case, the electron spin is affected by the total magnetic field with oblique orientation $\mathbf{B}_\Sigma$, given by the sum of the nuclear ($\mathbf{B}_N$) and external ($\mathbf{B}$) fields [see the inset in Fig. 1(b)]. Thus, there is a component of the mean electron spin normal to $\mathbf{B}_\Sigma$ (and precessing about it) and a spin component parallel to $\mathbf{B}_\Sigma$ (no precession occurs). As a result, there are oscillating and monotonically decaying contributions to the PL polarization.

Figure 2 shows a series of PL polarization time dependencies in different magnetic fields measured in the presence [Fig. 2(a)] and in the absence [Fig. 2(b)] of DNP. Let us consider the results shown in Fig. 2(a). We assume that the electron spin in the X^+ trion is affected by the nuclear Overhauser field pinned along the QD growth axis. The angle between the mean electron spin $\langle \mathbf{S}_e \rangle$ and the total field $\mathbf{B}_\Sigma$ increases with increasing external field $\mathbf{B}$. As a result, the value of the spin projection on the $\mathbf{B}_\Sigma$ direction decreases, while the value of the spin projection on the axis normal to $\mathbf{B}_\Sigma$ increases. Thus, the amplitude of the monotonically decaying polarization contribution decreases with external magnetic field, as observed in Fig. 2(a). The strength of the field at which the amplitude of the monotonically decaying contribution equals the amplitude of the oscillating contribution is

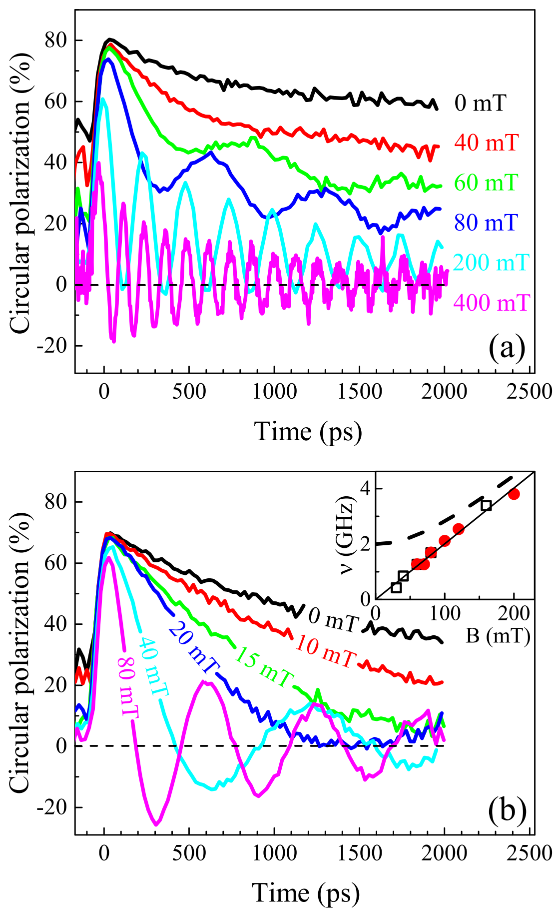

FIG. 2. Dynamics of the PL circular polarization in the (a) presence and (b) absence of DNP in different Voigt magnetic fields. The inset in (b) shows the dependence of the polarization oscillation frequency on magnetic field strength in the presence (solid circles) and in the absence (empty squares) of DNP. The calculated dependence is shown by the dashed line.

approximately 100 mT. This value determines the strength of the effective field of the dynamically polarized nuclear spins $\mathbf{B}_N$. In the absence of DNP [Fig. 2(b)] no noticeable monotonically decaying contribution is observed. Indeed, the electron spin projection on the direction of the external field is close to zero.

To determine the frequency of the electron spin Larmor precession, the signals shown in Figs. 2(a) and 2(b) were Fourier transformed. The oscillation frequency ν increases linearly with external magnetic field both in the presence and in the absence of DNP [inset in Fig. 2(b)]. The electron g factor value, $|g_e| = 1.43$, is determined from the slope of this linear dependence using $h\nu = \mu_B|g_e|B$. It agrees with the value obtained in [11], while it differs from the value of 1.6 obtained in other studies [20,21]. However, if the nuclear field is pinned along the growth axis, the oscillation frequency will be proportional to $\sqrt{B^2 + B_N^2}$ [see the dashed line in the inset of Fig. 2(b)]. The model predicts a significant deviation from the straight line in fields smaller than or equal to the nuclear field, $B_N = 100\,\text{mT}$, which we do not observe [see the inset of Fig. 2(b)]. Thus, in the range of small fields there is a contradiction between model and experiment. On the one hand, the magnetic field dependencies of the PL circular polarization indicate the presence of the nuclear field. On the other hand, the Larmor precession of the electron spin occurs as if the nuclear field is absent.

In order to resolve this contradiction, we propose that the PL consists of two independent contributions. The oscillating contribution corresponds to the X^+ trion, and the monotonically decaying contribution corresponds to the X^- trion. Therefore, in the ensemble of nominally negatively charged QDs there is a subensemble of positively charged dots generated by photodoping. A quantum well recharging under excitation below the barriers was reported for GaAs/(Al,Ga)As semiconductors [22]. The possibility of the simultaneous X^+ and X^- contributions to a single InAs/GaAs QD spectrum was demonstrated in [23]. In addition, to describe the experimental results it is necessary to assume that in the presence of resident electrons (leading to X^- trions) dynamic polarization of the nuclear spins takes place, while in the absence of resident electrons (leading to X^+ trions) the DNP can be neglected. This point can be explained by the different lifetimes of resident and photoexcited carriers. In the latter case, there is most likely not sufficient time for significant interaction between the photoexcited and subsequently radiatively decaying electrons and the nuclei. Moreover, it is still assumed that the Overhauser field of the dynamically polarized nuclei (the X^- case) is pinned along the QDs growth axis.

Using these hypotheses, we can consistently describe all experimental results. Let us consider now the oscillation frequency dependence on the magnetic field [inset in Fig. 2(b)]. As noted in the Introduction, the dynamics of the optical orientation of X^- trion PL does not exhibit oscillations in Voigt magnetic field (see also Sec. IV B). Thus, the dependence is completely determined by the Larmor precession of the electron spin in the X^+ trion. In the case of the X^+ trion, the absence of the DNP is proposed, so that the observed linear dependence of the frequency on magnetic field as well as the coincidence of the results in the absence and presence of excitation modulation is expected. We recall that modulation of the excitation polarization at a sufficiently high frequency prevents DNP.

Let us consider the time dependencies of the PL polarization in the presence of DNP [Fig. 2(a)]. The polarization of the X^- trion monotonously vanishes in the Voigt magnetic field without observing oscillations. If one subtracts this contribution from the experimental signal, a polarization oscillating around the zero value attributed to the X^+ trion will remain [the dashed line in Fig. 1(c)]. However, in the absence of DNP [Fig. 2(b)] the monotonically decaying contribution (related to the X^- trion) is vanishing already in smaller fields (about 16 mT). In this case, the electron spin is influenced by only the Voigt external magnetic field, while in the presence of DNP [Fig. 2(a)] the Voigt external field has to be larger than the nuclear field, whose direction coincides with the QD growth axis, in order to depolarize the X^- trion PL.

For the steady-state magnetic field dependencies of the polarization [Fig. 1(b)] the arguments proposed earlier regarding the nuclear field that is pinned along the growth axis remain valid. At the same time, we assume that the Hanle curve, measured in the absence of DNP, is the sum of two Lorentz curves, reflecting the contributions of the X^+ and X^- trions,

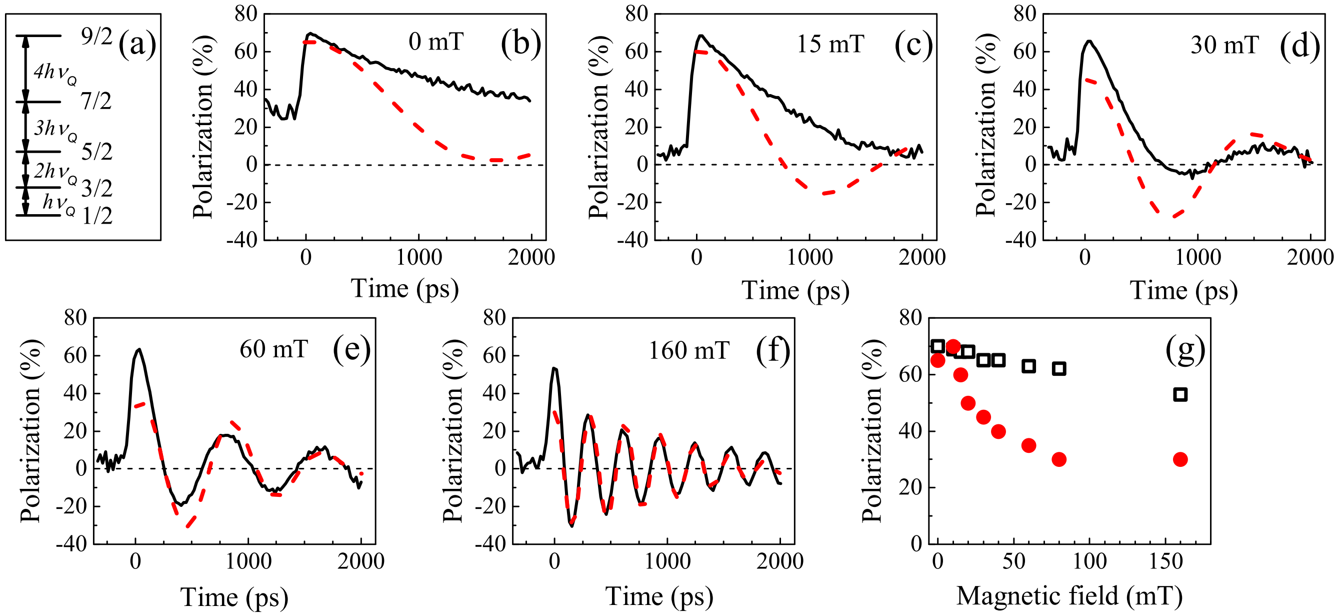

FIG. 3. (a) Energy states corresponding to the modulus of the indium nuclei spin projection. (b)–(f) Time dependencies of the PL circular polarization in various Voigt magnetic fields. The experiment is shown by solid lines; the modeling is shown by dashed lines. (g) Dependence of the experimental data (empty squares) and the calculated values (solid circles) of the polarization maximum on the external magnetic field.

with approximately equal HWHM (16 mT) [Fig. 1(b), the black line]. In the presence of DNP, the Hanle curve can be fitted by the sum of two Lorentz curves with HWHMs of 16 and 70 mT [Fig. 1(b), the red line]. Thus, the half width of the X^+ trion depolarization curve remains the same, while the X^- trion depolarization curve is noticeably broadened due to the presence of the nuclear field. The strength of the nuclear field of 70 mT can be estimated from the HWHM of the Lorentz curve corresponding to the X^- trion PL depolarization.

IV. DISCUSSION

A. Nuclear quadrupole interaction

The pinning of the nuclear spins along the growth axis of the QDs is caused by the quadrupole interaction of the indium nuclei (95.5% ^{115}In and 4.5% ^{113}In, both with spin $I = 9/2$ [24]). For the nuclei of phosphorus (with spin 1/2) the QI is absent. In the case of uniaxial strain along the QDs growth axis (z) the QI Hamiltonian is given by [6]

$$h\nu_Q\left[\hat{I}_z^2 - I(I+1)/3\right]/2, \qquad (2)$$

where h is the Planck constant. The constant ν_Q is defined by

$$\nu_Q = \frac{3eV_{zz}Q}{2I(2I-1)h}, \qquad (3)$$

where e is the electron charge, V_{zz} is the electric field gradient, and Q is the quadrupole moment of the nuclei. The QI lifts the degeneracy of eightfold nuclear spin states already in zero magnetic field, and the nuclear energy structure comprises four doubly degenerate Kramers doublets, as shown in Fig. 3(a).

The QI plays a significant role in external magnetic field when the Zeeman splitting of nuclear spins is smaller than the quadrupole splitting. Estimates show that for the ^{115}In nuclei and uniaxial strain of 2% directed along the QD growth axis the quadrupole splitting dominates over the Zeeman splitting up to 100 mT external magnetic field [5]. In this field range the quadrupole effects are observed in our case. Fields below 100 mT and oriented perpendicular to the quadrupole axis do not split doubly degenerate states, which are associated with a fixed modulus of the nuclear spin projection (greater than 1/2) on the QD growth axis [Fig. 3(a)]. As a result, the nuclear dipole-dipole interaction does not destroy the orientation of the dynamically polarized nuclear spins even in the absence of an external magnetic field. It should be noted that the effect of QI on the system of electron and nuclear spins should manifest in different types of self-organized quantum dots with nuclear spins greater than 1/2.

B. Quantitative description of the PL polarization dynamics in the absence of DNP

As can be seen in Fig. 1(b), the widths of the Hanle curve and the polarization restoration curve in Faraday magnetic field are equal in the absence of DNP. This is a characteristic feature of electron spin relaxation in nuclear field fluctuations with a long correlation time. In this case, there is a well-proven theory for describing the time dependencies of polarization [17]. The theory describes the QD ensemble-averaged electron spin relaxation in frozen nuclear field fluctuations subject to external magnetic field. No recombination or other spin relaxation mechanisms are taken into account. The results of the polarization dynamics fitted by the corresponding functions are presented in Figs. 3(b)–3(f). The parameters of the model are the characteristic value of nuclear field fluctuations Δ_b, the modulus of the electron g factor $|g_e|$, and the characteristic value of the electron spin dephasing time T_Δ. Moreover, it is sufficient to know any two of these parameters to determine the third one. As noted above, $|g_e| = 1.43$ was obtained from the experiment. Δ_b was chosen as a fitting parameter. In addition, the functions were multiplied by the factor A normalizing the amplitude. The best match is obtained for $\Delta_b = 12$ mT, which is close to the HWHM of the Hanle curve (16 mT). It should be noted that in the limit of long correlation time of the field fluctuations the HWHM of the Hanle curve is determined by the strength of the nuclear field fluctuations. Based on the known dimensions of the QD (100×5 nm^2) and the electron g factor, one obtains a theoretical estimate of the value of the nuclear field fluctuations Δ_b of 20 mT [17], which agrees well with the value experimentally obtained in this work (16 mT).

Let us turn to the data in zero magnetic field [Fig. 3(b)]. After 100 ps from the moment of excitation the circular polarization degree reaches its maximum, which is equal to 70% (but not 100%). This indicates the presence of a fast spin relaxation mechanism (during the thermalization of carriers to the trion ground state). The polarization magnitude then decays in a few nanoseconds by a factor of 2.8 from 70% to 25%, where it remains constant until the next laser pulse comes in [in Fig. 3(b) one can see the constant level at "negative delays"; the pulse repetition period is 13 ns]. This situation is characteristic of spin relaxation in frozen fields. However, in magnetic fields smaller than 60 mT there is a significant mismatch between theory and experiment. Good agreement is achieved only in relatively large magnetic fields exceeding 60 mT [Figs. 3(e) and 3(f)].

The presence of two contributions to the PL (X^+ and X^-) could be the reason for the mismatch between theory and experiment in small fields. In the singlet state of the X^+ trion the spins of the two holes compensate each other, so that the electron spin in the QD determines the trion spin during its lifetime. In this case, the model [17] can be used. In the case of the X^- trion, the nuclear field acts on the resident electron spin only until the electron pair singlet state with zero total spin is formed by photoexcitation. Then the spin dynamics is determined by the spin of the heavy hole in the trion. Thus, the case of the X^- trion is beyond the scope of the theory, and as a result, the description of the experimental data in low fields faces a problem. In large fields (exceeding 40 mT) the monotonically decaying PL contribution of the X^- trion vanishes, making the theory application possible.

Let us consider the influence of a magnetic field in Voigt geometry on the spin dynamics of the X^- trion. The transverse g factor of the heavy hole is small ($g_{hh}^{\perp} \ll 1$) [25]. As a result, a Voigt magnetic field of about 100 mT does not affect the hole spin dynamics. In other words, the precession period of the hole spin and, consequently, of the polarization is longer than the hole lifetime. We recall that the orientation of the hole spin determines the PL polarization after formation of the trion singlet state. Prior to that, the electron spins precess in magnetic field, and their orientation at the moment of singlet state formation affects the subsequent hole spin orientation. Now if there is a dispersion (over the QD ensemble) of the time of electron energy relaxation into the singlet state, the precession of the electron spins in different QDs will end at different times, so they will be differently oriented at the moment of thermalization. As a result, the average hole spin at the moment of singlet state formation will decrease with increasing field. The latter circumstance leads to the decrease (from 70% to 50%) of the polarization maximum (obtained from the dynamic curves), which is reflected in the decrease (from 70% to 30%) of the fit parameter A [Fig. 3(g)] that is introduced for normalization of the fit functions amplitude. The difference between the measured and calculated values of the polarization maximum [Fig. 3(g)] in strong fields is related to the presence of the short-in-time polarization peak right after the laser pulse. Such behavior can be attributed to contributions of short-lived PL from the trion excited states and requires further investigation.

In the field range of 60–320 mT the polarization dynamics are determined by the electron spin in the X^+ trion. As a result, good agreement between theory and experiment is achieved everywhere except the short time interval right after the laser pulse. In this case of strong fields, the depolarization time T_Δ does not depend on magnetic field, indicating the absence of a noticeable dispersion of the electron g factor.

V. CONCLUSION

We have shown that in the presence of nuclear quadrupole interaction in InP/(In,Ga)P QDs the dynamically polarized spins of the nuclei and the spins of the electrons are stabilized when there are resident electrons in the QDs. Moreover, the stabilization takes place even in zero external magnetic field. In addition, the QI leads to pinning of the Overhauser field along the quadrupole axis, close to the QD growth axis. The latter circumstance, as well as the presence of two contributions to the PL, one which corresponds to the PL of the X^- trion and one that corresponds to the X^+ trion, significantly affects the dynamics of circular polarization in the Voigt magnetic field.

We have also shown that relaxation of the electron spins in the frozen fluctuations of the nuclear field takes place. The experimentally measured time dependencies of the PL polarization were modeled using the theory presented in Ref. [17]. In the range of relatively large magnetic fields (60–320 mT), good agreement between theory and experiment was achieved. The mismatch between theory and experiment in the field range up to 60 mT may be caused by the presence of the X^- contribution to the PL polarization, which is outside of the scope of the model.

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