Letter

Long-lived photon echo induced by nuclear spin fluctuations in charged InGaAs quantum dots

A. V. Trifonov [1,2,*] I. A. Yugova,[2] A. N. Kosarev,[1] Ya. A. Babenko [2] A. Ludwig,[3] A. D. Wieck [3]
D. R. Yakovlev [1,4] M. Bayer,[1] and I. A. Akimov [1]
[1]*Experimentelle Physik 2, Technische Universität Dortmund, 44221 Dortmund, Germany*
[2]*Spin Optics Laboratory, St. Petersburg State University, Peterhof, 198504 St. Petersburg, Russia*
[3]*Angewandte Festkörperphysik, Ruhr-Universität Bochum, 44780 Bochum, Germany*
[4]*Ioffe Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia*

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We study the role of the hyperfine interaction between nuclei and resident electrons in the formation of a three-pulse spin-dependent photon echo from self-assembled (In,Ga)As semiconductor quantum dots. In zero magnetic field we observe an oscillatory coherent optical response, which is detected in a time window of 10 ns, which is significantly longer than the radiative lifetime of trions of 0.26 ns. The oscillations occur due to spin precession of the resident electron spins in the effective fluctuating nuclear field, whose direction and magnitude are different in each quantum dot. We evaluate the mean field of nuclear spin fluctuations $\sqrt{\langle B_{\rm N}^2\rangle} = 6.4$ mT. Owing to the specific mechanism of spin initialization, which depends on the direction and magnitude of the effective nuclear field, the spin-dependent photon echo extends the possibilities for exploring the hyperfine interaction in semiconductor quantum dots.

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The spins of a resident electron and nuclei in semiconductor nanostructures are considered as promising candidates for applications in quantum information and communication [1–3]. In particular, self-assembled semiconductor quantum dots (QDs) are appealing for photonics applications due to the fast optical initialization on a sub-ps timescale and relatively long optical coherence time of elementary optical excitations (excitons), which is limited by a radiative lifetime of about 1 ns [4,5]. Charged quantum dots are especially interesting for spin-photon entanglement [6–8], the generation of polarization-encoded photonic cluster states [9,10], as well as possible implementation in quantum memories [11–13]. All of these tasks require long spin relaxation times for resident electrons. In QDs, due to the strong localization of carriers in all three dimensions, spin-orbit relaxation mechanisms are suppressed, while the hyperfine interaction of electrons with nuclei becomes very important and determines the spin dynamics of the coupled electron-nuclear system [14]. It results in spin dephasing and relaxation of the electron spin [15–19]. However, it can be also exploited for the long-lived storage of information, which is particularly attractive for light-matter interfaces [2,20].

Optical studies of electron spin dynamics in QDs are usually based on optical orientation where the out-of-plane electron spin component parallel to the direction of incident light [axis z in Fig. 1(a)] is initialized by means of pumping with circularly polarized light [17–19,21]. The readout can be subsequently accomplished by measuring the polarization state of the emitted photons from a QD [22] or via Faraday/Kerr rotation of the polarization plane for a transmitted/reflected laser beam in pump-probe [23] and spin noise [24,25] experiments.

In this Letter, we employ a sequence of two linearly polarized picosecond optical pulses [Fig. 1(a)] tuned in resonance with the trion optical transition in QD to initialize a spin grating in an ensemble of singly charged QDs. Subsequent excitation with a third optical pulse results in a coherent optical response represented by a three-pulse photon echo (PE) which can be used to study the spin dynamics of resident electron spins [26]. Previous studies were limited to the observation of long-lived photon echoes from resident electron spins in the presence of an in-plane external magnetic field only [13,27,28]. Here, we demonstrate experimentally and theoretically that even in zero magnetic field the temporal decay of photon echo in (In,Ga)As QDs possesses a long-lived oscillatory component induced by effective fluctuating nuclear fields with $\delta_{\rm N} = \sqrt{\langle B_{\rm N}^2\rangle} = 6.4$ mT. In addition, a weak nonoscillating long-lived echo signal is detected due to spin relaxation of the hole in the trion state with a relaxation time $T_{\rm sh} \gtrsim 5$ ns. To describe the experiment theoretically, we extend the model for oblique magnetic fields and include the hyperfine interaction between electrons and nuclei.

The experiment was performed on a sample, which comprises four layers of n-doped (In,Ga)As QDs in a GaAs matrix, embedded in the antinodes of the standing wave of the microcavity with $Q \sim 1000$ [Fig. 1(a)]. Optical pulses with a photon energy of 1.434 eV, duration of 2.5 ps, and repetition frequency of 75.75 MHz are tuned in resonance with the spectrally broader photonic mode of the microcavity [Fig. 1(b)]. As shown in Ref. [13], the coherent optical response is represented by a three-pulse PE and the main contribution in the studied sample is attributed to trions. The signal is measured

*Corresponding author: artur.trifonov@tu-dortmund.de

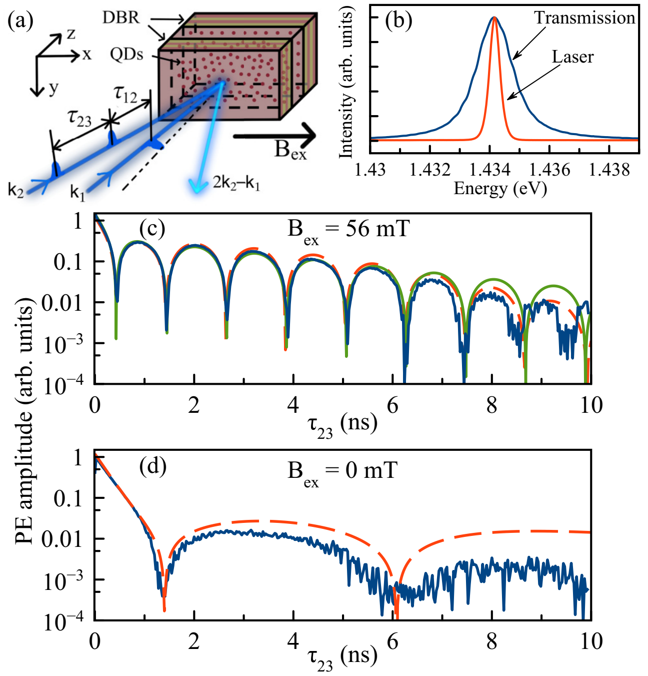

FIG. 1. (a) Schematic presentation of the experiment, including the coordinate system and studied structure which comprises the QD layers embedded between the distributed Bragg reflectors. Three pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ are incident on the sample, separated in time by delays τ_{12} and τ_{23}, respectively. The direction of the detected signal is $2\mathbf{k}_2 - \mathbf{k}_1$. (b) Sample transmission spectrum (blue) and laser spectrum (red). Dependence of three-pulse photon echo on τ_{23} in a transverse magnetic field $B_{\mathrm{ex}} = 56$ mT (c) and in zero magnetic field (d) in the polarization configuration HVVH. Blue lines in (c) and (d) show the experiment and red lines are calculations in the nuclear spin fluctuation model. The green line in (c) is the fit by Eq. (1). $\tau_{12} = 33.3$ ps. $T = 1.5$ K.

in the most informative HVVH polarization configuration, where the first pulse and detection polarization are linearly and horizontally polarized along the external magnetic field, while the second and third pulses are vertically polarized. The signal is measured in reflection geometry using heterodyne detection. All measurements are performed at a temperature of 1.5 K. A weak in-plane external magnetic field is applied by means of a resistive electromagnet. In our studies, the sample was oriented with the [110] crystallographic axis parallel to the external magnetic field, ensuring that the effective magnetic field acting on the heavy hole is parallel to the actual external magnetic field [12]. Further details of the sample growth and transient four-wave mixing setup are described in Refs. [12,29].

The blue lines in Fig. 1(c) show a typical dependence of three-pulse PE amplitude as a function of delay between the second and third pulses, τ_{23}, measured for an external magnetic field $B_{\mathrm{ex}} = 56$ mT. Note that heterodyne detection allows us to measure only the absolute value of the PE signal. The signal is given by an initial decay due to the radiative recombination of trions with $\tau_r = 250$ ps. At longer delays oscillations are observed due to the spin precession of resident electrons in charged QDs in an external magnetic field. This

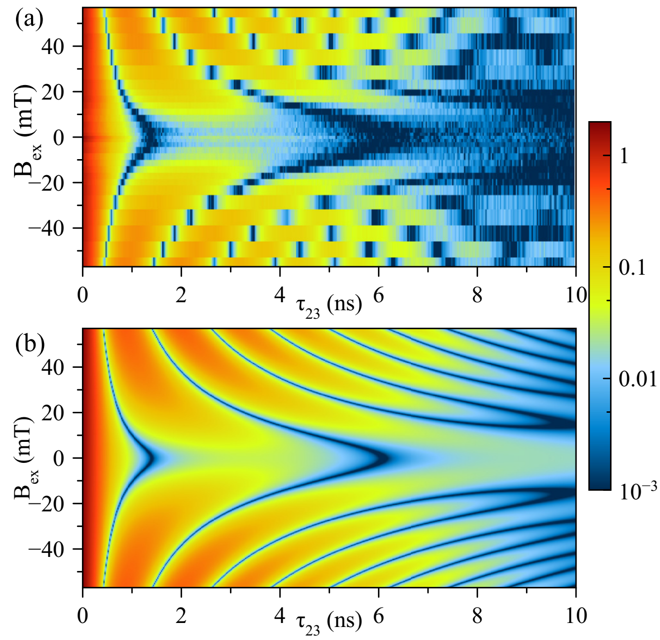

FIG. 2. Three-pulse photon echo as a function of τ_{23} and B_{ex} in the HVVH polarization configuration. $\tau_{12} = 33.3$ ps. (a) Experiment. (b) Calculation in the nuclear spin fluctuation model.

PE signal is described by the theory developed in Ref. [13], where spin dephasing is described phenomenologically by

$$P_{\mathrm{HVVH}} \propto \left[e^{-\frac{\tau_{23}}{\tau_r}} r_{\mathrm{h}} \cos\left[\omega_{\mathrm{h}}\tau_{23} - (\omega_{\mathrm{e}} - \omega_{\mathrm{h}})\tau_{12} - \phi_{\mathrm{h}}\right] + e^{-\frac{\tau_{23}}{T^*_{2,\mathrm{e}}}} r_{\mathrm{e}} \cos\left[\omega_{\mathrm{e}}\tau_{23} + (\omega_{\mathrm{e}} - \omega_{\mathrm{h}})\tau_{12} - \phi_{\mathrm{e}}\right]\right] e^{-\frac{2\tau_{12}}{T_2}}. \tag{1}$$

The green solid line in Fig. 1(c) is the fit using Eq. (1) with $T^*_{2,\mathrm{e}} = 3.3$ ns. Other parameters r_{e}, r_{h}, etc., are discussed in Ref. [13].

Figure 1(d) shows the dependence of the three-pulse PE amplitude as a function of τ_{23} measured at zero external magnetic field (blue line). The signal again consists of short-lived and long-lived components. For $\tau_{23} < 1$ ns, the signal is determined by trion recombination and decays exponentially with $\tau_r = 250$ ps. Surprisingly, the long-lived signal experiences unexpected oscillations. The possible residual magnetization in the used cryostat is less than 1 mT, which is much less than the value of a 13 mT magnetic field required to reproduce the experiment with sufficient accuracy by Eq. (1). In order to verify the absence of residual magnetic fields that can cause the long-lived PE signal, a series of measurements was carried out at various magnetic field values of both signs. Figure 2(a) presents these data by a color map, which is symmetric with respect to the magnetic field and clearly demonstrates the presence of an oscillating signal for $B_{\mathrm{ex}} = 0$. Note that the buildup of dynamic nuclear polarization does not occur due to the modulation technique of the experiment, as discussed below.

In what follows, we demonstrate that the long-lived PE signal in zero magnetic field is attributed to the influence of nuclear spin fluctuations. These fluctuations create a local effective magnetic field $\mathbf{B}_{\mathrm{N}}$, which is random for each QD.

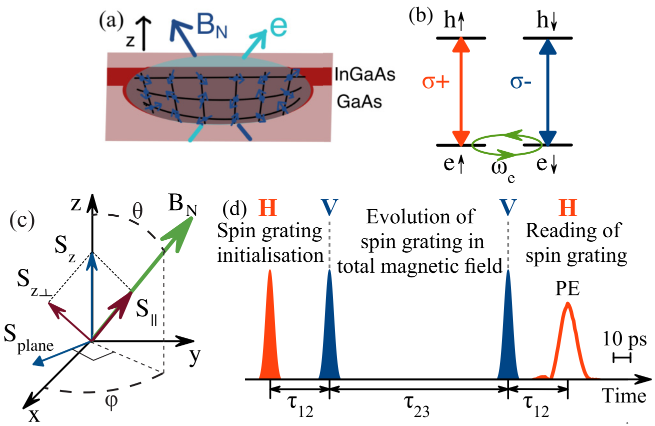

FIG. 3. (a) Schematic diagram of an (In,Ga)As QD within a GaAs matrix. Small blue arrows at the nodes of the lattice represent random orientations of nuclear spins within the quantum dot. The large arrow "$\mathbf{B}_N$" corresponds to the effective magnetic field of nuclear fluctuations acting on the resident electron spin "e." (b) Energy level structure and optical transitions in negatively charged QD. (c) Initialization of the electron spin by the sequence of two linearly cross-polarized pulses. The angles θ and φ define the orientation of the magnetic field acting on the electron. $\mathbf{S}_z$ and $\mathbf{S}_{\text{plane}} \sin\theta = \mathbf{S}_x + \mathbf{S}_y$ are electron spin components lying orthogonal and in the sample's plane, respectively. $\mathbf{S}_\parallel$ and $\mathbf{S}_{z\perp}$ are components of $\mathbf{S}_z$ directed along and perpendicular to the magnetic field. (d) Temporal sequence of optical pulses applied to the sample, showing the delays between the pulses, the moment of occurrence of the spin-dependent PE pulse, and the main stages of the spin-dependent photon echo protocol. Polarization of the pulses and detection of PE are indicated with H and V corresponding to linear horizontal and vertical polarizations.

This phenomenon, along with its impact on electron spin relaxation, is elaborated upon in greater detail in Refs. [15,16]. Despite the extensive studies of nuclear spin fluctuations in semiconductors, their manifestation in optical coherent spectroscopy has remained unexplored.

We have developed a theoretical description of the spin-dependent three-pulse photon echo for trions in QDs, which are subject to an external magnetic field in the Voigt geometry and nuclear spin fluctuations. For clarity we present the case of zero external magnetic field, because this problem has an analytical solution. The more general case is presented in the Supplemental Material (SM) [30]. We consider the spin of a single resident electron localized in a QD which experiences a hyperfine interaction with the spins of the nuclei in the electron localization area as it is shown in Fig. 3(a). In thermodynamic equilibrium, the average value of the nuclear spin at zero external magnetic field is equal to zero. However, the action of fluctuations is equivalent to an effective magnetic field $\mathbf{B}_N$ around which the electron spin experiences Larmor precession. This leads to damping of the electron spin [14,16]. The hyperfine interaction of the hole spin with nuclear spins is weaker by about an order of magnitude [19]. Therefore we assume that the nuclei affect only the electron spin dynamics, while the hole spin undergoes relaxation at a rate of $1/T_{sh}$ which is introduced phenomenologically. Finally, since the change in $\mathbf{B}_N$ occurs on a microsecond timescale, we assume that during its evolution, the electron spin is subject to constant $\mathbf{B}_N$, i.e., a frozen fluctuation model is used.

The main stages of a three-pulse PE are schematically shown in Fig. 3(d). First, resonant excitation of a trion in a QD with a sequence of two linearly cross-polarized pulses [e.g., the first pulse is horizontally polarized (H), and the second one is vertically polarized (V)] leads to the orientation of a resident electron [ground state $e\uparrow, e\downarrow$ in Fig. 3(b)] and a trion [excited states $h\uparrow, h\downarrow$ in Fig. 3(b)] spin,

$$S_z = -J_z = \text{Re}(A)\cos\left[\frac{\omega_e\tau_{12}}{2}\right],$$
$$S_x = J_x = \text{Im}(A)\sin\theta\sin\varphi\sin\left[\frac{\omega_e\tau_{12}}{2}\right],$$
$$S_y = J_y = -\text{Im}(A)\sin\theta\cos\varphi\sin\left[\frac{\omega_e\tau_{12}}{2}\right], \quad (2)$$

where θ is the angle between $\mathbf{B}_N$ and normal to the sample plane (z axis), φ is the direction of $\mathbf{B}_N$ projection in the sample plane [see Fig. 3(c)], $\omega_e = \mu_B g_e B_N/\hbar$ is the electron Larmor precession frequency, μ_B is the Bohr magneton, and g_e is the g factor of the electron [see Eq. (S16) in SM [30]]. The amplitude $A \propto e^{-\tau_{12}/T_2} e^{i[\omega_0\tau_{12}+(\mathbf{k}_1-\mathbf{k}_2)\mathbf{r}]} e^{i\Delta\varphi}$ with trion optical coherence time T_2 and an optical phase factor which is determined by the frequency of the trion resonance ω_0, $(\mathbf{k}_1 - \mathbf{k}_2)\mathbf{r}$ with $\mathbf{r}$ being the coordinate of the particular QD, and relative optical phase $\Delta\varphi$ between pulses [31].

As follows from Eq. (2), the S_z component is created even in the absence of an effective magnetic field, when $\omega_e = 0$, and is independent of the direction of the magnetic field. Its appearance is related to direct optical orientation, when the phase between the two linearly cross-polarized pulses (HV) is equal to $\pm\pi/2$, which is equivalent to excitation with $\sigma^\pm$ circularly polarized light. However, the spin components in the plane of the sample are formed only in the presence of the magnetic field with a nonzero in-plane component. Their appearance is related to the coherent evolution of the electron-trion system during the time interval τ_{12}, under conditions where the magnetic field leads to the mixing of electron $e\uparrow, e\downarrow$ states and the optical phase corresponds to $0, \pi$. Interestingly, it turns out that the spin initialized in the plane of the sample, $\mathbf{S}_{\text{plane}} \sin\theta = \mathbf{S}_x + \mathbf{S}_y$, is orthogonal to $\mathbf{B}_N$, where amplitude $|S_{\text{plane}}|$ corresponds to the in-plane spin component in the case of an in-plane field. Figure 3(c) shows the directions of the created spin components.

The experiment is performed on a QD ensemble with a strong inhomogeneous broadening of optical transitions. For simplicity, we assume that all of the QDs experience the same nuclear magnetic field $\mathbf{B}_N$. Averaging over fluctuating fields can be performed at the last stage. In this case, according to Eq. (2), the sequence of two pulses leads to the formation of spectral spin grating with a period of $1/\tau_{12}$ [factors Re(A) and Im(A)]. After that, the spin dynamics is determined by the spin precession of electron spin gratings in the effective magnetic field as well as the population decay of trions with recombination time τ_r. The third V-polarized optical pulse with wave vector $\mathbf{k}_3 = \mathbf{k}_2$ reads out the spin grating by transferring it back into optical electron-trion coherence and after the rephasing time τ_{12} the spin-dependent echo is formed. The expression for the long-lived part of the signal ($\tau_{23} \gg \tau_r$) is

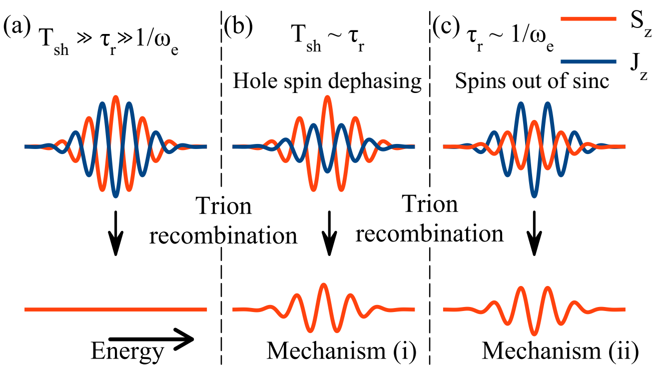

FIG. 4. (a) Complete compensation of electron spin grating S_z upon trion recombination for the case when the hole spin relaxation and electron spin precession are negligible. (b) Hole spin relaxation during recombination results in a decrease in the amplitude of the z projection of the hole spin grating, leading to the emergence of a long-lived electron spin grating during recombination. (c) Precession of electron spins around the magnetic field (out of synchronization with a hole), leading to the emergence of a long-lived electron spin grating during recombination.

given by [see Eq. (S42) in SM [30]]

$$\begin{aligned} P^{\text{LSPE}}_{\text{HVVH}} \propto\ & S_z \frac{\tau_r}{T_{\text{sh}}+\tau_r} \cos\left[\frac{\omega_e \tau_{12}}{2}\right]\cos^2\theta \\ & - S_z \frac{(\omega_e\tau_r)^2}{1+(\omega_e\tau_r)^2}\cos\left[\frac{\omega_e\tau_{12}}{2}+\omega_e\tau_{23}\right]\sin^2\theta \\ & + S_z \frac{\omega_e\tau_r}{1+(\omega_e\tau_r)^2}\sin\left[\frac{\omega_e\tau_{12}}{2}+\omega_e\tau_{23}\right]\sin^2\theta \\ & + S_{\text{plane}}\sin\left[\frac{\omega_e\tau_{12}}{2}+\omega_e\tau_{23}\right]\sin^2\theta. \end{aligned} \tag{3}$$

The signal is determined by the spin dynamics during the time interval τ_{23}. The first term on the right-hand side of the equation represents the component $\mathbf{S}_{\parallel}$ that is directed along the magnetic field. This component is subjected only to longitudinal spin relaxation and does not oscillate as a function of τ_{23}. The factor involving T_{sh} and τ_r indicates that this component of the signal arises due to spin relaxation of the hole during the trion radiative recombination, such that not all of the electron spin polarization disappears upon recombination. The second and third terms correspond to the evolution of the transverse component of $\mathbf{S}_{z,\perp}$ with respect to the magnetic field, which precesses about the magnetic field. The factors containing the product $\omega_e\tau_r$ indicate that these terms correspond to the spin grating components that appeared due to the different precession frequencies of electron and hole spins, so that after radiative recombination their z-spin components do not completely cancel each other (out of synchronization) [32,33]. Figures 4(a)–4(c) schematically illustrate the first three terms in Eq. (3), which are analogous to spin dynamics in optical orientation experiments. The last term in Eq. (3) is determined solely by the spin grating components in the plane of the sample created directly by the sequence of first two pulses.

Thus, the long-lived part of the spin-dependent PE signal is determined by three mechanisms: (i) hole spin relaxation in the trion state [first term in Eq. (3)], (ii) out of synchronization between the hole and electron spins due to spin precession [second and third terms in Eq. (3)], and (iii) direct generation of the electron spin in the sample plane and orthogonal to the magnetic field [fourth term in Eq. (3)]. Mechanisms (ii) and (iii) lead to the oscillating signal with the frequency ω_e as a function of delay time τ_{23}. In the fluctuation regime $\omega_e\tau_{12} \lesssim \omega_e\tau_r \ll 1$. In this case, the main contribution is given by mechanism (ii) [third term in Eq. (3)]. Its result is equivalent to the oscillating signal in optical orientation experiments [17,19]. Moreover, it turns out that the third and fourth terms in Eq. (3) have the same functional dependence. The fourth term with amplitude $S_{\text{plane}} \propto \omega_e\tau_{12}$ can give a reasonable contribution when $\tau_{12} \sim \tau_r$.

To obtain the final expression describing the spin-dependent PE signal from an ensemble of charged QDs taking into account fluctuating effective magnetic fields, it is necessary to average Eq. (3) over the directions and magnitude of the magnetic field. The distribution function over directions corresponds to a random uniform distribution, and the magnitude distribution is a Gaussian function with dispersion $\delta_N = \sqrt{\langle B_N^2\rangle}$. If the external magnetic field is zero and only the field of nuclear fluctuations is acting, it is possible to obtain an analytical expression, which is presented by Eq. (S53) in SM [30]. In the case of the simultaneous existence of an external magnetic field acting both on electron and hole spins and a nuclear fluctuation field, averaging can be performed only numerically.

The red dashed line in Fig. 1(d) is the theoretical curve obtained in the discussed above model [Eq. (S53) in SM [30]]. For the modeling we used $g_e = -0.52$ and $\tau_r = 250$ ps as evaluated in our previous studies [12,13]. The last two fitting parameters are found as $T_{\text{sh}} \gtrsim 5$ ns and $\delta_N = \sqrt{\langle B_N^2\rangle} = 6.4$ mT, which are in agreement with the literature data [17,19,25]. The red dashed line in Fig. 1(c) shows numerically obtained dependences for $B_{\text{ex}} = 56$ mT, which reproduce the decay of the experimental signal with high accuracy. Here, we use $g_h = 0.18$. The signal decay is related to the spin dephasing of resident electrons in a fluctuating nuclear spin environment. Other mechanisms of the electron spin relaxation are not considered. Here, the phenomenological parameter $T^*_{2,e} = 3.3$ ns [see Eq. (1) and the green line] can also be estimated as $T^*_{2,e}(B=0) = 3\hbar/(\pi\mu_B g_e \delta_N)$ for the obtained value of $\delta_N = 6.4$ mT. We find good agreement between experiment and theory by taking into account the nuclear spin fluctuations for all measured values of the external magnetic fields. Figures 2(a) and 2(b) present these data by a color map.

Spin-dependent PE can be also observed in the HHHH polarization configuration when all optical pulses and the detected signal are linearly copolarized. As shown in Sec. 3.3 of SM [30], the long-lived signal due to nuclei spin fluctuations is very low, being much smaller than the offset due to other contributions as discussed in Ref. [13].

In conclusion, we demonstrate that a hyperfine interaction between nuclear spins and resident electrons in self-assembled (In,Ga)As QDs leads to the formation of a long-lived coherent optical response at the trion transition in the absence of an external magnetic field. We expanded the theory of the spin-dependent photon echo for the case of arbitrarily oriented magnetic fields and show that nuclear spin fluctuations directed in the sample plane lead to oscillatory behavior, while out-of-plane fluctuations do not contribute to a three-pulse PE. We evaluate the magnitude of $\sqrt{\langle B_N^2 \rangle} = 6.4$ mT. In addition, a weak nonoscillating signal, which appears due to hole spin relaxation on times $T_{sh} \gtrsim 5$ ns, is detected. For small nuclear fields, the result is similar to the oscillatory signal in optical orientation experiments. However, a three-pulse echo excitation protocol does not continuously pump the spin along the z axis in all QDs. Moreover, it employs the specific spin initialization with a sequence of two cross-polarized pulses, where electron spins are generated perpendicularly to the direction of the nuclear field, which is individual for each QD and may have a different feedback on the nuclear system in a particular QD. This opens different possibilities for exploring the hyperfine interaction in semiconductor quantum structures.

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