Optical Control of Spin Coherence in Singly Charged (In, Ga)As/GaAs Quantum Dots

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Electron spin coherence has been generated optically in n-type modulation doped (In, Ga)As/GaAs quantum dots (QDs) which contain on average a single electron per dot. The coherence arises from resonant excitation of the QDs by circularly polarized laser pulses, creating a coherent superposition of an electron and a trion. Time dependent Faraday rotation is used to probe the spin precession of the optically oriented electrons about a transverse magnetic field. The coherence generation can be controlled by pulse intensity, being most efficient for $(2n + 1)\pi$ pulses.

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An electron spin in a single quantum dot (QD) represents a qubit candidate that is very attractive for solid state quantum information processing [1–3], as suggested by long electron spin coherence times, T_2, measured in bulk semiconductors [4]. These long times are required for performing a sufficient number of quantum manipulations during which coherence needs to be retained. Recent QD studies have demonstrated long electron spin relaxation lifetimes, T_1, in the millisecond range [5]. This has raised hopes that T_2, which may last as long as $2T_1$ [6], could be similarly long, with encouraging indications to that effect found lately [7]. For fast spin manipulation, rotations by Raman processes are envisaged whose cross sections can be enhanced by resonant excitation of a charged exciton [8]. In a first step, however, electron spin coherence must be established, which was recently addressed in GaAs/AlGaAs interface QDs [9]. However, only rather low excitation powers were used in those experiments, so that coherent control of electron spin polarization in the form of Rabi oscillations did not occur.

Here we demonstrate by pump-probe Faraday rotation (FR) that electron spin coherence can be generated by circularly polarized optical excitation of singly charged QDs. Resonant excitation creates an intermediate superposition of a singlet trion and an electron, which, after trion radiative decay, leaves behind a long-lived electron spin coherence. Coherence is controlled by the pump pulse area, $\propto \int E(t)dt$, where $E(t)$ is the electric field amplitude. It reaches maximum for $(2n + 1)\pi$ pulses [10], in accordance with our theory, which also shows that $2n\pi$ pulses can be used for refocusing the precessing spins.

The experiments were performed on self-assembled (In, Ga)As/GaAs QDs. To obtain strong enough light-matter interaction, the sample contained 20 QD layers separated by 60 nm wide barriers, fabricated by molecular beam epitaxy on a (001)-oriented GaAs substrate. The layer dot density is about 10^{10} cm^{-2}. For an average occupation by a single electron per dot, the structures were n-modulation doped 20 nm below each layer with a Si-dopant density roughly equal to the dot density. The sample was thermally annealed so that its emission occurs around 1.396 eV, as seen from the luminescence spectrum in Fig. 1(a), inset. The full width at half maximum of the

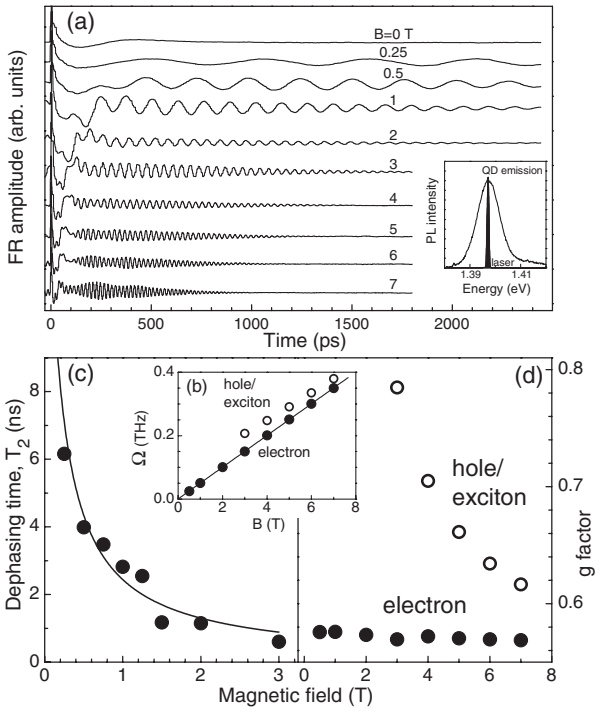

FIG. 1. (a) FR traces of n-doped (In, Ga)As/GaAs QDs vs delay between pump and probe at different B. Pump power was ~10 mW. Inset: Photoluminescence of QDs and laser spectrum in FR studies. (b) and (d) Field dependencies of electron and hole or exciton precession frequencies and g factors. (c) Spin dephasing time T_2^* vs B. Line is a $1/B$ fit to data.

emission is 10 meV. Further optical properties of these dots can be found in Refs. [11,12].

The sample was immersed in liquid helium at temperature $T = 2$ K. The magnetic field $B \leq 7$ T was aligned perpendicular to the structure's growth direction z. For FR [3], a Ti-sapphire laser emitting pulses with a duration of $\sim$1 ps ($\sim$2 meV full width at half maximum) at 75.6 MHz repetition rate was used, hitting the sample along z. The laser was tuned to the QD ground state transition [see inset of Fig. 1(a)]. For detecting the rotation angle of the linearly polarized probe pulse, a homodyne technique based on phase-sensitive balanced detection was used.

Figure 1(a) shows the FR signal of the QDs vs delay between pump and probe for different magnetic fields. Pronounced electron spin quantum beats are observed with some additional modulation at high B. The oscillations at low B last much longer (for example, about 4 ns at 0.5 T) than the radiative trion lifetime of $\tau_r = 400$ ps, as measured by time-resolved photoluminescence and therefore are due to long-lived residual electrons. Three features are to be noted:

(1) The oscillation frequency increases with magnetic field as expected from the electron spin splitting: $\hbar\Omega_e = g_e\mu_B B$ with the Bohr magneton μ_B. g_e is the electron g factor. We have analyzed the FR dynamics by an oscillatory function with exponentially damped amplitude, $\propto \exp(-t/T_2^*)\cos(\Omega_e t)$. The resulting B dependence of the electron precession frequency is shown in Fig. 1(b). From a B-linear fit we obtain $|g_e| = 0.57$.

(2) The spin beats become increasingly damped with increasing magnetic field, corresponding to a reduction of the ensemble spin dephasing time T_2^*, plotted in Fig. 1(c). The damping arises from variations of the electron g factor within the QD ensemble, causing an enhanced spread of Ω_e with increasing B, whose impact on dephasing can be described by $[T_2^*(B)]^{-1} = [T_2^*(0)]^{-1} + \Delta g_e \mu_B B/\sqrt{2}\hbar$. The solid line in Fig. 1(c) shows a $1/B$ fit to the T_2^* data, by which a g factor variation $\Delta g_e = 0.004$ is extracted. From the data one can also conclude that $T_2^*(0)$ exceeds 6 ns. The zero-field dephasing is mainly caused by electron spin precession about the frozen magnetic field of the nuclei [13]. The net nuclear orientation varies from dot to dot, and it is these variations that lead to ensemble spin dephasing.

(3) The additional modulation at high fields with frequency Ω_X is observable only during the trion lifetime. Therefore, it can be assigned either to photoexcited holes having a spin splitting $\hbar\Omega_h = g_h\mu_B B$ with the hole g factor g_h, or to excitons in neutral QDs in the ensemble. At $B = 5$ T, the decay time of the modulation is 170 ps, significantly shorter than the 400 ps dephasing of the electron spin precession, which indicates either strong dispersion of Ω_X, or fast hole or exciton spin dephasing. The B dependence of Ω_X is shown in Fig. 1(b) and of the effective g factor, $\hbar\Omega_X/(\mu_B B)$, in Fig. 1(d). In the range from 3 to 7 T this g factor decreases from 0.78 to 0.62 while g_e is constant. If Ω_X orignates from the hole, this dependence suggests significant mixing of light-hole and heavy-hole states in the studied QDs [14]. For a neutral QD contribution, it arises from the bright exciton fine structure splitting.

Figure 2(a) shows FR signals at $B = 1$ T for different pump powers. The corresponding FR amplitudes are plotted in Fig. 2(b) vs pulse area, defined as $\Theta = 2\int[\boldsymbol{dE}(t)]dt/\hbar$ in dimensionless units with the dipole transition matrix element $\boldsymbol{d}$. For pulses of constant duration, but varying power, as used here, Θ is proportional to the square root of excitation power, and it is given in arbitrary units in Fig. 2(b). The amplitude shows a nonmonotonic behavior with increasing pulse area. It rises first to reach a maximum, then drops to about 60%. Thereafter it shows another strongly damped oscillation. This behavior is similar to the one known from Rabi oscillations of the Bloch vector [15]. The FR amplitude reaches its maximum when applying a π pulse as pump, for which the z component of the Bloch vector is fully inverted. It goes to its minimum for a 2π pulse, for which the Bloch vector is turned by 360°, and so on. The damping of Rabi oscillations is most likely due to ensemble inhomogeneities of QD properties such as the dipole moment $\boldsymbol{d}$ [16]. The results in Figs. 1 and 2 are an important input for identifying the origin of spin coherence.

The magnetic field $\boldsymbol{B} \parallel \boldsymbol{e}_x$ leads to a splitting of the electron spin into eigenstates with spin parallel $|+x\rangle$ or antiparallel $|-x\rangle$ to the field. Disregarding the hole for simplicity, the optical pulses create in *neutral* QDs electrons with spin states $|\uparrow\rangle$ or $|\downarrow\rangle$ along the z direction of light propagation, $S_z = \pm 1/2$, which can be expressed as coherent superpositions of the two eigenstates $|\pm x\rangle$. Therefore spin quantum beats occur, which in a classical picture correspond to electron spin precession about the magnetic field. In any case, the precession cannot last longer than the exciton lifetime.

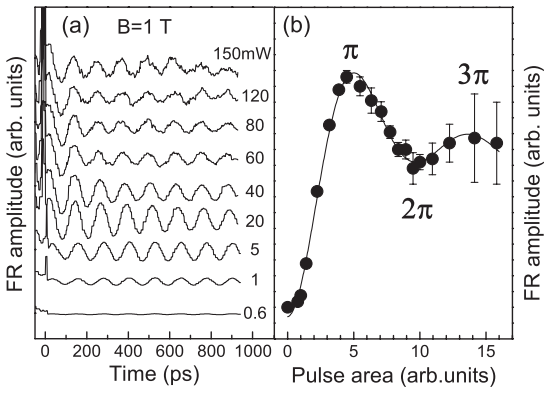

FIG. 2. (a) Short delay closeup of FR signal at $B = 1$ T for different pump powers. (b) FR amplitude vs laser pulse area. The line is a guide to the eye.

In *charged* QDs, resonant optical excitation leads to formation of trions $|\uparrow\downarrow\Uparrow\rangle$ or $|\uparrow\downarrow\Downarrow\rangle$, consisting of two electrons forming a spin singlet and a hole in one of the states $|\Uparrow\rangle$ or $|\Downarrow\rangle$ with spin projection $J_{h,z} = \pm 3/2$ for σ^+ or σ^- polarized light [17]. Initially, the resident electron may have arbitrary spin orientation $|\psi\rangle = \alpha|\uparrow\rangle + \beta|\downarrow\rangle$, where $|\alpha|^2 + |\beta|^2 = 1$, if the trion state is not populated, and $|\alpha|^2 + |\beta|^2 < 1$, otherwise. The electron spin polarization is described by the spin vector $\boldsymbol{S} = (S_x, S_y, S_z)$ defined by $S_x = \mathrm{Re}(\alpha\beta^*)$, $S_y = -\mathrm{Im}(\alpha\beta^*)$, $S_z = (1/2) \times (|\alpha|^2 - |\beta|^2)$. Similarly, one can introduce the spin vector, $\boldsymbol{J} = (J_x, J_y, J_z)$, that represents the polarization of the trion, $|\bar{\psi}\rangle = \bar{\alpha}|\uparrow\downarrow\Uparrow\rangle + \bar{\beta}|\uparrow\downarrow\Downarrow\rangle$.

A *short* pulse of circularly polarized light is a remarkable tool for coherently controlling the electron spin in a transverse magnetic field. If the pulse length is much shorter than the radiative decay and the carrier spin relaxation times, it mixes the electron and trion spin states into a coherent superposition, which is not affected by decoherence during generation. For controlled resonant pumping as in the experiments, the superposition is uniquely determined by the pulse area Θ. By variation of this area not only are the electron and trion state populations changed periodically with period $\Theta = 2\pi$, but also the orientation of electron and trion spins $\boldsymbol{S}$ and $\boldsymbol{J}$ are controlled.

The spin vectors $\boldsymbol{S}$ and $\boldsymbol{J}$ represent 6 of the 16 components of the four level density matrix, and their dynamics is given by density matrix equations of motion [18]. The electron spin vector evolution as a function of Θ is shown in Fig. 3 for two initial orientations: one is parallel to the magnetic field and the other exemplifies an arbitrary direction. A short σ^+ polarized pulse excites the initial electron spin state, $|\psi_0\rangle = \alpha_0|\uparrow\rangle + \beta_0|\downarrow\rangle$, into an electron-trion superposition state $|\psi_{\mathrm{ET}}\rangle = \alpha^0 \cos(\Theta/2)|\uparrow\rangle + \beta^0|\downarrow\rangle - i\alpha^0 \sin(\Theta/2)|\uparrow\downarrow\Uparrow\rangle$. The light induced change of the S_z component, $|S_z - S_z^0| = |\alpha_0|^2 \sin^2(\Theta/2)$ varies with the $|\uparrow\rangle$ state population, and independently of the initial conditions it reaches its maximum for $\Theta = (2n+1)\pi$ pulses, for which the S_x and S_y components vanish. In particular, $S_z([2n+1]\pi) = -0.25$ for $S_z^0 = 0$, in agreement with Ref. [19]. Note also that, unlike the S_z component, the electron spin swings between its initial direction (S_x^0, S_y^0, S_z^0) and the direction $(-S_x^0, -S_y^0, S_z^0)$ with period 4π. This is because the $S_{x,y} \sim \cos(\Theta/2)$ components describe the coherence of the electron spin state and both vary with the spin wave function phase, $|\psi_{\mathrm{ET}}\rangle$.

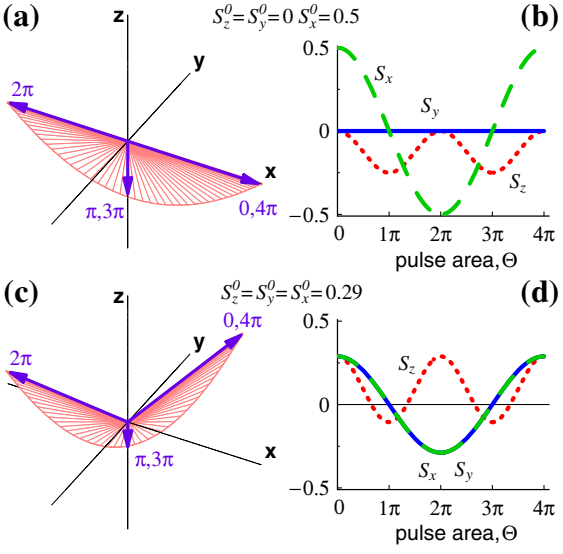

FIG. 3 (color online). (a) and (c) Reorientation of electron spin polarization by application of a resonant optical pulse of varying area as denoted. Calculations have been done for two different initial values of spin polarization, S_x^0 and S_z^0. (b) and (d) Electron spin polarization components vs pulse area Θ.

The control of spin dynamics by an optical pulse allows for a fast spin alignment. In a QD ensemble, a small area pulse, $\Theta \ll 1$, induces a coherent spin polarization proportional to Θ [9]. With increasing Θ, the total spin polarization oscillates with a period 2π as does the S_z component of each individual spin in the ensemble, explaining the FR amplitude oscillations in Fig. 2. The long trion lifetimes in our QDs could enable realization of a regime in which a pulse of rather low power but long duration can be used to reach a large pulse area without decoherence due to radiative decay. Further, the S_x and S_y components change sign with period 2π. This implies that $2n\pi$ pulses can be used for refocusing the precessing spins, similar to spin-echo techniques [20].

Let us turn now to the spin dynamics after initialization by a short pulse. Then the off-diagonal component of the density matrix, describing electron-trion coherence, is decoupled from the electron and trion spin vectors, which are governed independently by two coarse-grained vector equations:

$$\frac{d\boldsymbol{J}}{dt} = [\boldsymbol{\Omega}_h \times \boldsymbol{J}] - \frac{\boldsymbol{J}}{\tau_s^h} - \frac{\boldsymbol{J}}{\tau_r},$$
$$\frac{d\boldsymbol{S}}{dt} = [(\boldsymbol{\Omega}_e + \boldsymbol{\Omega}_N) \times \boldsymbol{S}] + \frac{(\hat{\boldsymbol{J}}\,\hat{z})\hat{z}}{\tau_r}, \quad (1)$$

where $\boldsymbol{\Omega}_{e,h} \parallel \boldsymbol{e}_x$ and $\boldsymbol{\Omega}_N = g_e \mu_B \boldsymbol{B}_N/\hbar$ is the electron precession frequency in an effective nuclear magnetic field, $\boldsymbol{B}_N$. In the second equation we do not include the electron spin relaxation time, τ_s^e, explicitly. At low temperatures, τ_s^e is at least of the order of μs and is mainly determined by fluctuations of the nuclear field $\boldsymbol{\Omega}_N$ in a single QD [5,12,13,21]. This time scale is irrelevant to our problem. The spin relaxation of the hole in the trion, τ_s^h, is caused by phonon assisted processes and at low temperatures may be as long as τ_s^e [22,23].

Solving Eq. (1) we obtain the time evolution of the spin vectors $\boldsymbol{S}$ and $\boldsymbol{J}$. After trion recombination ($t \gg \tau_r$), the amplitude of the long-lived electron spin polarization ex-

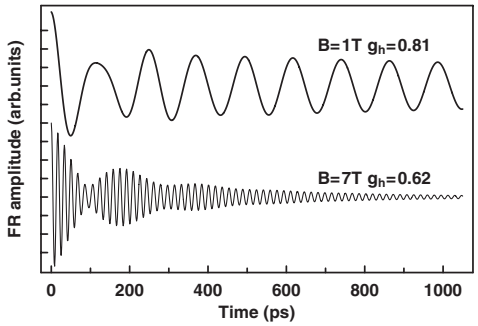

FIG. 4. Calculated time dependence of pump-probe FR signal of n-doped QDs excited by a σ^+ polarized pulse. $\tau_r = 400$ ps, $\tau_s^h = 170$ ps, $|g_e| = 0.57$, and $\Delta g_e = 0.004$.

cited by a $(2n+1)\pi$ pulse is given by

$$S_z(t) = \mathrm{Re}\left\{\left(S_z(0) + \frac{0.5J_z(0)/\tau_r}{\gamma_T + i(\omega + \Omega_h)} + \frac{0.5J_z(0)/\tau_r}{\gamma_T + i(\omega - \Omega_h)}\right)\exp(i\omega t)\right\}, \quad (2)$$

where $S_z(0)$ and $J_z(0)$ are the electron and trion spin polarizations created by the pulse. $\omega = \Omega_e + \Omega_{N,x}$. $\gamma_T = 1/\tau_r + 1/\tau_s^h$ is the total trion decoherence rate. If the radiative relaxation is fast, $\tau_r \ll \tau_s^h$, $\Omega_{e,h}^{-1}$, on average the induced spin polarization $S_z(t)$ is nullified by trion relaxation, as $S_z(0) = -J_z(0)$. In contrast, if the spin precession is fast, $\Omega_{e,h} \gg \tau_r^{-1}$, the electron spin polarization is maintained after trion decay [19,24], as observed in our case.

For an ensemble of QDs, the electron spin polarization is obtained by averaging Eq. (1) over the distribution of g factors and nuclear configurations. At low B, the random nuclear magnetic field becomes more important for the electron spin dephasing than g-factor dispersion, leading to dephasing during several nanoseconds [13]. The rotation of the linear probe polarization is due to the difference in scattering of its σ^+ and σ^- polarized components by one of the transitions $|\uparrow\rangle \rightarrow |\uparrow\downarrow\Uparrow\rangle$ and $|\downarrow\rangle \rightarrow |\uparrow\downarrow\Downarrow\rangle$. The scattering efficiency is proportional to the population difference of the states involved in these transitions $\Delta n_+ = n_\uparrow - n_\Uparrow$ or $\Delta n_- = n_\downarrow - n_\Downarrow$. The FR angle is $\phi(t) \sim (\Delta n_+ - \Delta n_-)/2 = S_z(t) - J_z(t)$.

Figure 4 shows the FR signal after a σ^+-polarized excitation pulse, calculated with input parameters from experiment. At $B = 7$ T, the FR shows modulated beats resulting from interference of the electron and trion precessions during the 400 ps trion lifetime. At $B = 1$ T the beats are less pronounced due to the larger difference between electron and hole g factors. These results are in good agreement with the experiment. The modeling, however, does not allow us to determine the additional modulation origin uniquely. Joint contribution of charged and neutral QDs would also lead to pump-probe FR traces similar to those in Fig. 4 if we assume that the 170 ps is the exciton dephasing time and trion hole dephasing occurs on a scale of tens of ps. Further experimental investigations of this question are required.

In conclusion, we have demonstrated that pulses of circularly polarized light allow for coherent phase control of an electron spin in a QD. The coherent control results in FR amplitude oscillations with varying laser pulse area.

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