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 state. Time dependent Faraday rotation is used to probe the spin precession of the optically oriented electrons about a transverse magnetic field. Spin coherence generation can be controlled by pulse intensity, being most efficient for (2n + 1)\pi-pulses.

An electron spin in a single QD represents a qubit candidate that is very attractive for solid state quantum information processing as has been suggested by long electron spin coherence times, T2, measured in bulk semiconductors. 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, T1, in the millisecond-range at cryogenic temperatures. This has raised hopes that T2, which may last as long as 2T1, could be similarly long, with encouraging indications to that effect found lately. For fast spin manipulation, rotations by Raman processes are envisaged whose cross sections can be enhanced by resonant excitation of a charged exciton. In a first step, however, electron spin coherence must be established, which recently was addressed in charged GaAs/AlGaAs interface QDs. However, only rather low excitation powers were used in those experiments, so that coherent control of electron spin polarization in form of Rabi oscillations did not occur.

In this Letter 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 is converted into a long lived electron spin coherence. The 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, in good accord with our theoretical model. Theory also shows that \( 2\pi t \)-pulses can be used for refo- cussing of 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 layers of QDs separated by 60 nm wide barriers. It was fabricated by molecular beam epitaxy on a (001)-oriented GaAs substrate. The layer dot density is about \( 10^{10} \text{ cm}^{-2} \). 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 the inset of Fig. 1(a). The full width at half maximum of the emission line is 10 meV, demonstrating good ensemble homogeneity. Further optical properties of these dots and undoped reference sample can be found in Ref. 11.

The sample was immersed in liquid helium at temperature \( T = 2 \text{K} \). The magnetic field \( B \leq 7 \text{T} \) was aligned perpendicular to the structure growth direction \( z \). In the FR pump-probe studies a Ti-sapphire laser emitting pulses with a duration of \( \sim 1 \text{ps} \) (full width at half maximum of \( \sim 2 \text{meV} \)) at 75.6 MHz repetition rate was used, hitting the sample along \( z \). The laser was tuned to the QD ground state transition energy (see inset of Fig. 1(a)). The circular polarization of the pump beam was modulated at a frequency of 50 kHz, to avoid nuclear polarization effects. For detecting the rotation angle of the linearly polarized probe pulses, 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 \( B \). With increasing delay time the beats become damped. The oscillations at low \( B \) last much longer (for example, about 4 ns at 0.5 T) than the radiative trion lifetime of \( \tau_r = 400 \text{ps} \), 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 spin-splitting of electron states: \( h\Omega_e = g_e\mu_B B \), where \( g_e \) is the electron \( g \)-factor and \( \mu_B \) is the Bohr magneton. We have analyzed the FR dynamics in Fig. 1(a) by an oscillatory function with exponentially damped amplitude, \( \propto \exp(-t/T_2^*) \cos(\Omega_e t) \). The resulting \( B \)-field 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 of the spin precession arises from variations of the electron $g$-factor within the QD ensemble, causing an enhanced spread of $\Omega_e$ with increasing $B$, whose impact on the depolarization time 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 experimental data for $T_2^*$, from which a $g$-factor variation of $\Delta g_e = 0.004$ can be extracted. From the data one can also conclude that $T_2^*(0)$ is longer than 6 ns. The zero-field precession is mainly caused by electron spin precession about the frozen magnetic field of the dot nuclei in a QD. The net orientation of nuclei varies from dot to dot, and it is these variations that lead to ensemble spin dephasing.

3) The additional modulation of the FR traces at high fields is observable only during the trion lifetime and therefore can be naturally assigned to the photoexcited holes with a spin-splitting $\hbar \Omega_h = g_h \mu_B B$, where $g_h$ is the hole $g$-factor. The analysis gives $|g_h| = 0.66$ at $B = 5$ T. The decay time of this mode is 170 ps which is significantly shorter than the 400 ps decay of the electron spin precession for this field strength, pointing either at strong variations of $g_h$ or at fast hole spin relaxation.

The $B$-dependence of $\Omega_h$ is shown in Fig. 1(b), and the hole $g$-factor vs $B$ is given in Fig. 1(d). $g_h$ varies with $B$ over the studied field range, while $g_e$ is constant. In the range from 3 to 7 T, where the beat modulation is detectable, $g_h$ decreases from 0.78 to 0.62. This dependence suggests that mixing of light-hole and heavy-hole states is significant for the studied QDs.

Figure 2(a) shows FR signals at $B = 1$ T for different pump powers. The corresponding FR amplitude is plotted in Fig. 2(b) as function of the pulse area, which is defined as $\Theta = 2 \int |dE(t)| dt / \hbar$ in dimensionless units, where $d$ is the dipole transition matrix element. For pulses of constant duration, but varying power, as used here, the pulse area is proportional to the square root of excitation power, and it is given in arbitrary units in Fig. 2(b). The amplitude shows a non-monotonic 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 very similar to the one known from Rabi-oscillations of the Bloch vector for varying excitation power. The FR amplitude becomes maximum when applying a $\pi$-pulse as pump, for which the $z$-component of the Bloch vector is fully inverted. It becomes minimum for a $2\pi$-pulse, for which the Bloch vector is turned by $360^\circ$, and so on. The damping of the Rabi oscillations most likely is due to ensemble inhomogeneities of QD properties such as the transition dipole moment. These observations are important input for identifying the origin of spin coherence.

This origin will be discussed in the following: The magnetic field $B \parallel e_x$ leads to a splitting of the electron spin into eigenstates with spin parallel ($+x$) or antiparallel ($-x$) to the field. Disregarding the hole for simplicity, for neutral QDs the optical pulses create 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 spin eigenstates $|\pm x\rangle$. Therefore spin quantum beats occur, which in...
The three dimensional spin vectors $S$ and $J$ represent six of the sixteen components of the four level density matrix and their dynamics can be described by density matrix equations of motion. The evolution of the electron spin vector as function of $\Theta$ is shown in Fig. 3 for two initial spin directions: one is parallel to the magnetic field and the other exemplifies an arbitrary direction. A short $\sigma^+$ polarized pulse resonantly excites the initial electron spin state, $|\psi_i\rangle = |\alpha\rangle |\uparrow\rangle + \beta|\downarrow\rangle$, into the coherent superposition of the electron and trion states $|\psi_{\text{ET}}\rangle = \alpha_0^* \cos(\Theta/2)|\uparrow\rangle + \beta_0^* |\downarrow\rangle - i \alpha_0 \sin(\Theta/2) |\uparrow\downarrow\rangle$. The light induced deviation of the $S_z$ component, $|S_z - S_z^0| = |\alpha_0|^2 \sin^2(\Theta/2)$ changes with the $|\uparrow\rangle$ state population, and independently of the initial conditions it reaches a maximum for $(2n + 1)\pi$-pulses with $\Theta = (2n + 1)\pi$, 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. [18]. It is interesting to note that, unlike the $S_z$ component, the electron spin, swings between the initial direction $S_z^0$ and the direction $(-S_x^0, S_y^0)$ with a period $4\pi$. This is because the $S_{x,y} \sim \cos(\Theta/2)$ components describe the coherence of the electron spin state and both components change with the phase of the spin wave function, $|\psi_{\text{ET}}\rangle$.

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 $\Theta$, as observed in shallow GaAs/AlGaAs interface QDs Ref. [9]. With increasing $\Theta$, the total spin polarization oscillates as does the $S_z$ component of each individual spin in the ensemble all of which oscillate with the same period, $2\pi$. The long trion lifetimes in our QDs could enable realization of regime in which pulse of rather low power, but long duration can be used to reach these large pulse area without decoherence due to radiative decay. This explains the FR amplitude oscillations with pulse area in Fig. 2. Further, the $S_x$ and $S_y$ components change sign with period $2\pi$. This implies that $2n\pi$-pulses can be used for refocusing of precessing spins, very similar to spin-echo techniques. [19]

Let us turn now to the spin dynamics after its initialization by the short pulse: The trion component of the electron-trion coherent superposition decays spontaneously leading to the observed long-lived spin precession of the resident electron. After the pulse, the off-diagonal component of the density matrix, describing electron-trion coherence becomes decoupled from the electron and trion spin vectors, $S$ and $J$, which are governed independently by two coarse-grained vector equations:

$$\frac{dJ}{dt} = [\Omega_{x,h} \times J] - \frac{J}{\tau_s} - \frac{J}{\tau_r},$$

$$\frac{dS}{dt} = [(\Omega_e + \Omega_N) \times S] + \frac{(Jz) \hat{z}}{\tau_e},$$

where $\Omega_{x,h} \parallel e_x$ and $\Omega_N = g_e \mu_B B / h$ is the precession frequency of the electron in an effective nuclear magnetic field, $B_N$. We do not include in the second

\[ J_\text{int} = J_s - J_\text{bulk}, \]

\[ S_\text{int} = S_s - S_\text{bulk}. \]
The equation the spin relaxation time, \( \tau^e_s \), in an explicit form. At low temperatures, \( \tau^e_s \) is on the order of \( \mu \)s and it is mainly determined by fluctuations of the nuclear field \( \Omega_N \) in a single QD. This time scale is irrelevant to our present problem. The spin relaxation of the hole in the trion, \( \tau^h_r \), is caused by phonon assisted processes and at low temperatures it may be as long as \( \tau^e_s \) in QDs.

Solving Eq.(1) we obtain the time evolution of the spin vectors \( S \) and \( J \). After trion recombination \( (t \gg \tau_r) \), the amplitude of the long-lived electron spin polarization excited by a \((2n+1)\pi\)-pulse is given by

\[
S_z(t) = \text{Re} \left\{ \left( S_z(0) + \frac{0.5J_z(0)}{\tau_T + i(\omega + \Omega_h)} \right) \exp(i\omega t) \right\},
\]

where \( S_z(0) \) and \( J_z(0) \) are the electron and trion spin polarizations created by the pulse. \( \omega = \Omega_e + \Omega_N, \gamma_T = 1/\tau_T + 1/\tau^h_r \) is the total trion decoherence rate. On average, the induced spin polarization \( S_z(t) \) is nullified by trion relaxation, as \( S_z(t) = -J_z(t) \), if the radiative relaxation is fast \( \tau_r \approx \tau^e_s, \Omega^e_s^{-1} \). In contrast, if the spin precession is fast, \( \Omega^e,h \gg \tau^{-1}_r \), the electron spin polarization is maintained after trion decay, 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 magnetic fields, the random magnetic field of nuclei becomes more important for the electron spin dephasing than \( g \)-factor dispersion, leading to dephasing during several nanoseconds. The rotation of the linear probe polarization is due to the difference in scattering of the \( \sigma^+ \) and \( \sigma^- \) polarized components by one of the allowed transitions \( |\uparrow\rangle \rightarrow |\uparrow\downarrow\rangle \) and \( |\downarrow\rangle \rightarrow |\uparrow\downarrow\rangle \).

The scattering efficiency is proportional to the population difference of the states involved in the transition \( \Delta n_+ = n_+ - n_- \) or \( \Delta n_- = n_+ - n_- \), and 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 \( \sigma^+ \)-polarized excitation pulse, calculated with input parameters from experiment. At \( B = 7 \text{T} \), the FR shows modulated beats resulting from interference of the electron and trion precessions during the 400 ps trion lifetime. At \( B = 1 \text{T} \) the beats are less pronounced because of the larger difference between electron and hole \( g \)-factors. These results are in good agreement with experiment.

In conclusion, we have shown experimentally and theoretically that pulses of circularly polarized light allow for a 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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