

## Optical readout and initialization of an electron spin in a single quantum dot

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We describe theoretically the resonant optical excitation of a trion with circularly polarized light and discuss how this trion permits the readout of a single electron spin through a recycling transition. Optical pumping through a combination of circularly polarized optical  $\pi$  pulses with permanent or  $\pi$ -pulsed transverse magnetic fields suggests feasible protocols for spin initialization.

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An electron spin (ES) in a single quantum dot (QD) could be used as a solid-state qubit if certain requirements are met.<sup>1-3</sup> For example, it must be possible to coherently manipulate single and entangled spins in a time much less than the spin coherence time and to read out the state of the single ES during the spin-relaxation time. It has been suggested<sup>1</sup> that electronic gates could be used to accomplish these tasks in semiconductor materials, and great effort is being exerted currently to demonstrate the needed quantum tools in nanostructured systems. One of the most difficult of these requirements in a fully electronic implementation is the measurement of the state of a single spin.

An optical based computation scheme is of special interest because of the speed at which operations could be carried out using ultrafast laser technology. Similar techniques have achieved remarkable success in the measurement and control of the hyperfine states of individual ions in magnetic traps for quantum computing.<sup>4</sup> Over the last decade, materials development based on atomic layer growth, doping, and gating techniques have led to semiconductor QD systems in which a single electron can be injected into a QD, and most importantly, there have recently been a number of demonstrations in which a single QD with a single ES has been probed optically.<sup>5</sup> Furthermore, several groups have demonstrated optical Rabi oscillations in a single QD.<sup>6</sup> In this Rapid Communication we demonstrate theoretically that the resonant optical excitation of the spin to an intermediate trion state can lead both to readout and initialization capability in spite of random hyperfine fields of nuclei acting on the ES.

We consider singly charged QD's in which the lateral dimensions are much larger than the height—which serves as the quantization axis  $z$  for the electron and hole spin (HS) projections.<sup>7</sup> The ground electron and hole states in these QD's are degenerate with respect to their spin projection  $\pm 1/2$  (denoted as  $|\uparrow\rangle$  and  $|\downarrow\rangle$ ) and  $\pm 3/2$  (denoted as  $|\uparrow\rangle$  and  $|\downarrow\rangle$ ), respectively. The optical excitation of the charged dots leads to the formation of a trion that consists of two electrons sitting at the same quantum level and a hole (see Fig. 1). The  $|\uparrow\uparrow\downarrow\rangle$  ( $|\uparrow\downarrow\downarrow\rangle$ ) ground state of a trion created by  $\sigma^+$  ( $\sigma^-$ ) polarized light from the  $|\uparrow\rangle$  ( $|\downarrow\rangle$ ) electron state<sup>5</sup> can decay spontaneously only into the same initial electron state (see Fig. 1). This decay process is accompanied by emission of circularly polarized light in which  $\sigma^+$  or  $\sigma^-$  polarization is uniquely determined by the  $|\uparrow\rangle$  or  $|\downarrow\rangle$  ES projection, respec-

tively. Importantly, the selection rules allow the repetition of this cycle many times without losing the ES state.

As a result the polarization of emitted light allows us, in principle, to measure the ES projection through this recycling transition. However, the ES and the trion spin (TS) relaxation will limit the measurement time of the photoluminescence (PL) with the given polarization. It is obvious that one would want to collect as many photons as possible from a single QD before the ES relaxes. Even under resonant excitation condition, the rate of emitted photons cannot exceed  $\tau_r^{-1}$ , where  $\tau_r$  is the trion radiative decay time. At first glance this limits significantly the possibility of optical measurements of the spin state in a single QD because the spin-relaxation time does not exceed  $\sim 100\tau_r$  even in very optimistic estimates.

ES relaxation at low temperatures is mainly caused by its interaction with nuclei. In a zero external magnetic field the ES precesses in the hyperfine field,  $\mathbf{B}_N$ , of the frozen configuration of nuclear spins interacting with the localized electron with frequency  $\Omega_N = \mu_B g_e B_N / \hbar$ ,<sup>8</sup> where  $\mu_B$  is the Bohr magneton, and  $g_e$  is the electron  $g$  factor.  $\mathbf{B}_N$  has an arbitrary direction and its component perpendicular to the  $z$  axis of the QD leads to ES relaxation.<sup>9</sup> Calculation shows that the ES relaxation time is on the order of  $\sim 1-10$  ns, depending on the QD size.<sup>8</sup> This is usually longer than the radiative decay time of the trion.<sup>10</sup> In contrast, the trion should not interact strongly with the nuclear spin because its electrons are in a singlet state and a hole has negligible hyperfine interaction. The HS relaxation in the trion state is a two-phonon assisted

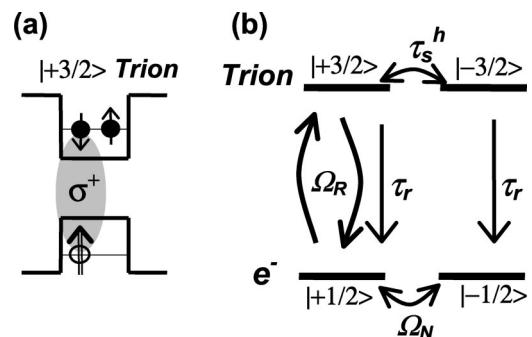


FIG. 1. Schematic presentation of (a) the trion created by  $\sigma^+$  polarized light in singly charged QD's; (b) coherent and spontaneous optical transitions and spin-relaxation processes that control the spin dynamics in the singly charged QD.

process.<sup>11</sup> The probability of such processes is proportional to the phonon concentration, which decreases exponentially with temperature. As a result the TS relaxation time  $\tau_s^h$  (as well as the ES time  $\tau_s^e$  connected with phonon assisted transitions) can be much longer than the ES relaxation time by nuclear interactions at low temperatures, and it is the latter that should limit our ability to measure the ES orientation. We will show, however, that ES relaxation in a nuclear field is suppressed by light in the strong coupling regime, and as a result the ES state in a single QD can be measured optically.

Intense resonant light drives the transitions between the electron and trion states. The frequency of these population oscillations is given by the Rabi frequency ( $\Omega_R$ ) at resonant excitation,  $\Omega_R = 2dE/\hbar$ , where  $d$  is the dipole moment of the optical transition and  $E$  is the electric-field amplitude. Under strong excitation conditions when  $\Omega_R \gg \Omega_N$  the ES cannot significantly change its direction because the spin precession is interrupted by the light-stimulated excitation of the electron into the trion state. The spin precession is interrupted periodically at the Rabi frequency, and at high excitation intensity the spin can change its projection by only a very small value  $(\Omega_N/\Omega_R)^2 \ll 1$ , a type of “line narrowing.”<sup>12</sup> As a result spin relaxation arising from the hyperfine field is suppressed.

The spontaneous decay of the trion is the incoherent process that destroys the synchronization of the ES motion with electromagnetic excitation and leads finally to ES relaxation in the strong excitation regime. A strong, polarized laser field splits the electron  $\pm 1/2$  spin states because of Rabi splitting. This reduces the admixture of the  $-1/2$  spin state to the  $+1/2$  spin state, which is proportional now to the small value  $\Omega_N/\Omega_R$ . As a result the rate of spontaneous trion decay into the electron  $-1/2$  spin state that describes the ES relaxation can be obtained in second-order perturbation theory:

$$\frac{1}{T_s^e} = \frac{1}{\tau_r} \left( \frac{\Omega_N}{\Omega_R} \right)^2. \quad (1)$$

The time of ES relaxation  $T_s^e$  under strong excitation,  $\Omega_N/\Omega_R \ll 1$ , is much longer than  $\tau_r$ . Thus, the optical recycling transition provides more PL photons before relaxation occurs.

We explore this problem using the density-matrix approach for the four-level system of Fig. 1(b). Polarized  $\sigma^+$  light with frequency  $\omega$  connects only the electron  $+1/2$  spin state with the trion  $+3/2$  spin state. ES relaxation arising from the hyperfine field leads to transitions between the ES states. We include now the HS relaxation, which leads to transitions between the TS states.

Figure 2 shows the time dependence of the diagonal component of the density matrix  $P_{3/2}$  describing the population of the  $+3/2$  trion state.  $P_{3/2}$  characterizes the intensity of the  $\sigma^+$  polarized PL because the rate of the  $\sigma^+$  polarized photon emission  $\sim P_{3/2}/\tau_r$ . The calculations were done for the two initial ES orientations ( $s_z = \pm 1/2$ ) for zero detuning,  $\varepsilon_{3/2} - \varepsilon_{1/2} - \hbar\omega = 0$ . One can see that at low excitation intensity [see Fig. 2(a)] the population of the trion state  $P_{3/2}$  excited by  $\sigma^+$  polarized light oscillates with the frequency of the ES precession in the hyperfine field of the nuclei. The oscillations

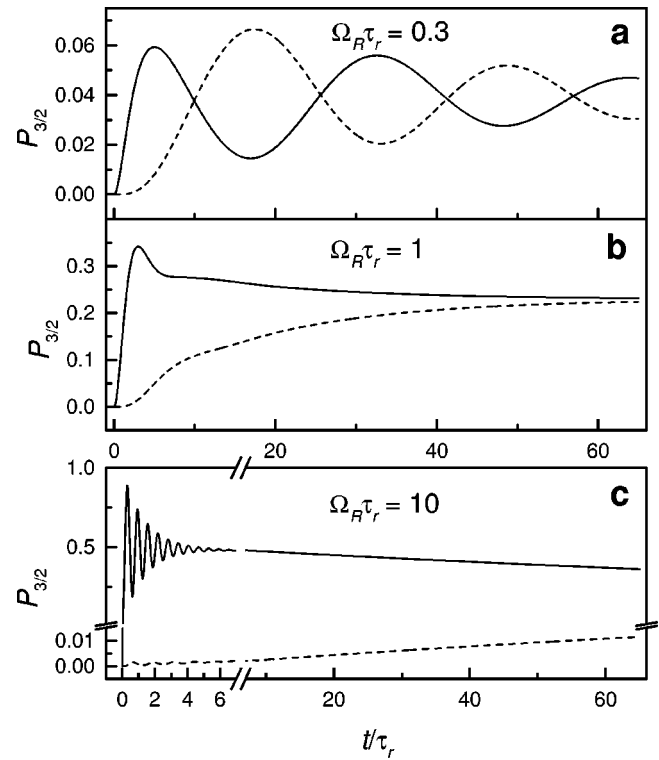


FIG. 2. Time dependence of the  $|\uparrow\downarrow\uparrow\rangle$  trion state population,  $P_{3/2}$ , for three excitation intensities for  $\Omega_N\tau_r=0.1$ . The solid and dashed lines are calculated for the  $|\uparrow\rangle$  and  $|\downarrow\rangle$  initial ES states, respectively.

have opposite phase for the two opposite initial conditions of the ES. At intermediate excitation intensity  $\Omega_R\tau_r = 1$  [see Fig. 2(b)] the precession oscillations are suppressed and the  $\sigma^+$  polarized PL from the QD when the electron initially has  $+1/2$  spin is more intense than if the electron has  $-1/2$  spin. When the optical excitation is further increased [Fig. 2(c)], Rabi oscillations appear at short times and the difference in PL intensity for the two cases remains much longer. The difference in the PL intensity for the two cases allows us to measure the ES polarization with high accuracy in the case of strong optical excitation [see Fig. 2(c)]. The technically challenging problem to distinguish emission from the scattering light in this scheme can be resolved, for example, by using their difference in spectral and angular distributions.

It is important to note that eventually the density matrix relaxes to the stationary state, which does not depend on the initial condition. That is why only the transient part of the PL polarization, measured at  $t < T_s^e$ , provides information on the ES polarization. At longer times *optical pumping* of the ES becomes important. This suggests using intense circularly polarized light for optical *initialization* of the ES's in the QD's.

The standard optical pumping of ES's demonstrated in atoms almost 50 years ago<sup>13</sup> is based on the depletion of one of the ground-state sublevels and accumulation in the other spin sublevel. Unfortunately, this optical pumping scheme does not normally work in our QD's<sup>7</sup> because the excited trion decays only into the same initial state [see Fig. 1(a)].

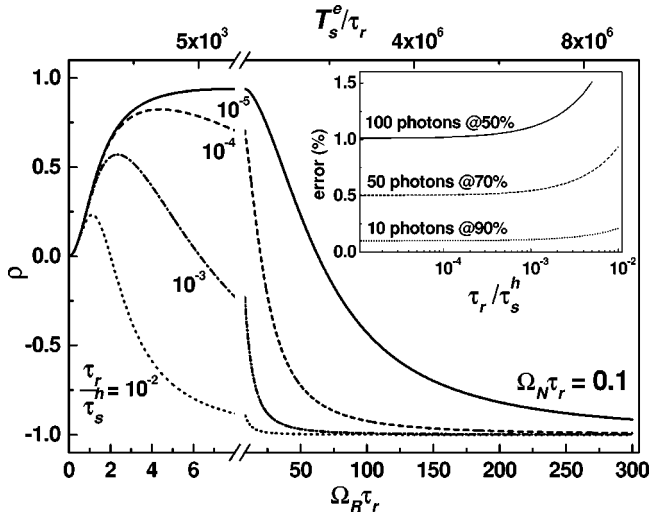


FIG. 3. Dependence of the steady-state degree of ES polarization on  $\Omega_R$  (proportional to the laser field amplitude) for different HS relaxation rates,  $1/\tau_s^h$ . Inset, dependence of the single spin measurement error on the HS relaxation rate for  $\Omega_R \tau_r = 10$ .

ES and HS relaxations allow us to overcome this restriction. Figure 3 shows the degree of ES polarization  $\rho = (P_{1/2} - P_{-1/2}) / (P_{1/2} + P_{-1/2})$  calculated as a function of excitation intensity in steady state ( $t \rightarrow \infty$ ), where  $P_{\pm 1/2}$  are the populations of the electron states. At times  $t > T_s^e$  the intense  $\sigma^+$  polarized light keeps the electron in one of its  $\pm 1/2$  spin states independent of the initial ES orientation. In the case when  $T_s^e \ll \tau_s^h$  one can completely neglect the HS relaxation processes. The steady-state solutions of the density matrix equations show that under strong excitation conditions,  $\Omega_R \gg \tau_r^{-1} \gg \Omega_N$ , an electron is practically always in the  $+1/2$  spin state and the probability to find it in the  $-1/2$  spin state is proportional to  $(\tau_r \Omega_R)^{-2} \ll 1$ . In the opposite limiting case,  $T_s^e \gg \tau_s^h$ , as a result of the HS flip processes, the electron is finally transferred to the optically passive  $-1/2$  state similar to that in the pumping experiment of Ref. 13. The probability to find an electron in the  $+1/2$  spin state is  $\sim \tau_s^h / T_s^e \ll 1$ .

As one can see in Fig. 3, a high degree of ES polarization can be achieved using a very intense polarized light source, such that  $\Omega_R \tau_r \gg 1$ . However, optical pumping of ES can also be achieved using a remarkable property of our QD's: the trion state is not affected by a magnetic field lying in the quantum wall (QW) plane in a first approximation. This allows one to use a combination of  $\pi$ -optical pulses and a transverse magnetic field for spin initialization in QD's.

The transverse magnetic field does not affect the singlet trion, consisting of two electrons in a singlet state and a heavy hole, because the  $|\uparrow\rangle$  and  $|\downarrow\rangle$  heavy-hole states have zero  $g$  factor in the plane of the quantum well.<sup>14</sup> A simple protocol leading to ES orientation is a sequential action of optical and magnetic  $\pi$  pulses. The first  $\sigma^+$  ( $\sigma^-$ ) polarized optical  $\pi$  pulse creates the  $|\uparrow\downarrow\uparrow\rangle$  ( $|\uparrow\downarrow\downarrow\rangle$ ) singlet trion state if the QD contains an electron in the  $|\uparrow\rangle$  ( $|\downarrow\rangle$ ) spin state. According to the selection rules, this optical pulse does not affect the QD if the electron is in the opposite  $|\downarrow\rangle$  ( $|\uparrow\rangle$ ) spin

state. The electron in this QD can be transferred to the  $|\uparrow\rangle$  ( $|\downarrow\rangle$ ) spin state by the short magnetic  $\pi$  pulse which at the same time would not affect the trion state, if it was formed. As a result 100% polarized electrons can be prepared in the  $|\uparrow\rangle$  ( $|\downarrow\rangle$ ) spin state after the trion relaxation to the ground state of the QD. For realization of this protocol the duration of the magnetic  $\pi$  pulse should be shorter than the lifetime of the trion.<sup>10</sup> Creation of short magnetic pulses is a difficult technical problem that perhaps can be solved, for example, using electric-field variation of the electron  $g$  factor in a permanent magnetic field.<sup>15,16</sup>

Alternatively, the combined effect of  $\pi$ -optical pulses and permanent transverse magnetic field allows one to initialize an ES into a state with a well-defined phase. The permanent magnetic field  $B$  lying in the QW plane leads to the precession of the ES with Larmor frequency  $\Omega_e = \mu_B g_e B / \hbar$ . However, the phase of its precession is not defined. The phase of the ES precession can be fixed by applying synchronized periodic optical  $\pi$  pulses with period  $T_p = 2\pi n / \Omega_e$ , where  $n = 1, 2, \dots$ . Such an experimental setup was used already for nonresonant excitation of QW's.<sup>17</sup>

To describe this process quantitatively, let us consider the dynamics of the electron and TS polarization during the time  $t$  between two optical circularly polarized  $\pi$  pulses:  $t_{N+1} > t > t_N$ , where  $t_N = N T_p$  ( $N = 0, 1, \dots$ ). In vector representation, the evolution of the ES polarization  $\mathbf{S}$  is described by

$$\frac{d\mathbf{S}}{dt} = [\mathbf{\Omega}_e \times \mathbf{S}] - \frac{\mathbf{S}}{\tau_s^e} + \frac{J(t)}{\tau_r} \mathbf{e}_z, \quad (2)$$

where  $\mathbf{e}_z$  is the unit vector along the  $z$  axis,  $\mathbf{\Omega}_e \perp \mathbf{e}_z$ ,  $\tau_s^e$  is the ES relaxation time, which is much longer than  $2\pi / \Omega_N$  in an external magnetic field  $B \gg B_N$ .<sup>12</sup>  $J(t) = J(t_N) \exp(-\gamma \Delta t)$  is the time dependence of  $1/3$  of the average spin projection of the trion hole on the  $z$  axis after the optical  $\pi$  pulse, where  $\Delta t = t - t_N$  and  $\gamma = 1/\tau_r + 1/\tau_s^h$ . Equation (2) can be solved for the complex spin components  $S_{\pm} = S_y \pm i S_z$  representing rotation of the spin around the magnetic field, which is taken along the  $x$  axis.

The important characteristic of the ES precession that describes ES polarization along the  $z$  axis is  $S_z(t) = \text{Im}[S_+(t)]$ . At the time when the trion recombines to the ground one-electron state ( $\Delta t \gg \tau_r$ ),  $S_z(t) = 0.5[P_{1/2}(t) - P_{-1/2}(t)]$ , and can be written

$$S_z(t) = \text{Re} \left[ \left( S_z(t_N + \delta) + \frac{J(t_N + \delta)}{\tau_r(\gamma + i\omega)} \right) e^{i\omega \Delta t} \right], \quad (3)$$

where  $\omega = \Omega_e + i/\tau_s^e$  and  $\delta \rightarrow 0$ . The initial values of the ES and TS polarizations in Eq. (3) after the  $\sigma^+$  polarized  $\pi$  pulse are connected with the density-matrix elements before this pulse:  $S_z(t_N + \delta) = -0.5P_{-1/2}(t_N - \delta)$  and  $J(t_N + \delta) = 0.5P_{1/2}(t_N - \delta)$ . These values are expressed through the ES polarization  $P_{1/2}(t_N - \delta) = 0.5[2S_z(t_N - \delta) + 1]$  and  $P_{-1/2}(t_N - \delta) = 0.5[1 - 2S_z(t_N - \delta)]$ . Substituting  $S_z(t_N + \delta)$  and  $J(t_N + \delta)$  into Eq. (3) one can obtain the recursive relation for  $S_z(t_{N+1} - \delta)$  and  $S_z(t_N - \delta)$ :

$$S_z(t_{N+1}) = \text{Re} \left\{ \left[ \frac{2S_z(t_N) - 1}{4} + \frac{2S_z(t_N) + 1}{4\tau_r(\gamma + i\omega)} \right] e^{i\omega T_p} \right\}.$$

Let us consider the spin dynamics of the initially unpolarized ES after action of the  $\sigma^+$  polarized  $\pi$  pulse. In this case the initial condition  $S_z(0 - \delta) = 0$ . The recursive relation has a clear physical interpretation in two limiting cases. First, when  $\Omega_e \tau_r \ll 1$  the denominator  $\tau_r(\gamma + i\omega) \approx 1$  because  $\tau_s^h, \tau_s^e \gg \tau_r$ . The ES polarization is conserved during trion relaxation; the trion contribution to the ES polarization,  $1/4$ , completely compensates the electron contribution,  $-1/4$ , and the electron remains unpolarized. In the opposite limit of strong magnetic fields,  $\Omega_e \tau_r \gg 1$ , the electron left after slow recombination of the trion is out of phase and this compensation does not happen. As a result, a significant ES polarization is created in the QD after trion recombination.

Repeating the optical  $\pi$  pulses allows us to pump the ES of a certain phase to high polarization if the ES relaxation time is long enough:  $T_p \ll \tau_s^e$ . Figure 4 shows the degree of ES polarization,  $\rho(t_N) = 2S_z(t_N)$ , as a function of the  $\pi$ -pulse number. The polarization degree reaches 99% after  $N = 8$  pulses.

This last *initialization* protocol can be matched with the *readout* scheme suggested above. Intense resonant steady-state excitation of the trion by circularly polarized light turned on at  $t = NT_p$  would allow one to measure the ES polarization if  $\Omega_R \gg \Omega_e$ .

The above consideration assumed ideal selection rules which hold for QD's with cylindrical shape or at least  $C_{2v}$  symmetry. Only significant deviations from this shape can change them in epitaxial QD's due to weak admixture of light-and heavy-hole subbands. To check the feasibility of our measurement scheme we include weakly allowed forbidden transitions in our four-level model as well as additional dephasing which may be connected with fluctuations of the trion-electron transition energy in a random electric field. These deviations from the ideal model increase the spin-relaxation rate. They do not affect significantly, however, our ability to determine an ES by measuring the burst of the photons or its absence. The effect can be quantified by the

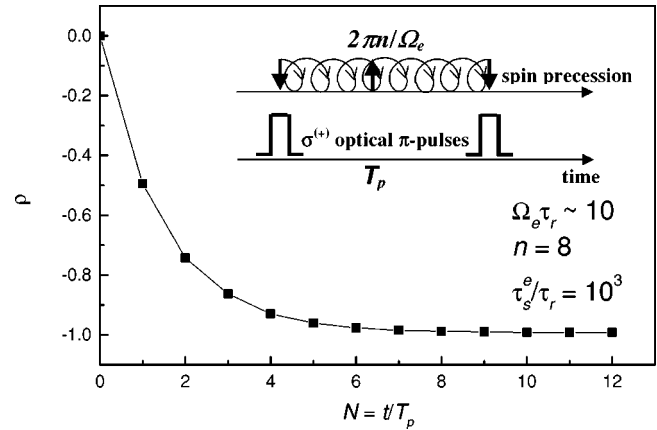


FIG. 4. The degree of ES polarization as a function of the  $\sigma^+$  polarized optical  $\pi$ -pulse number ( $N$ ).

measurement error introduced in Ref. 18. The inset in Fig. 3 shows the dependence of the error on the total HS relaxation rate where both additional mechanisms are included into  $1/\tau_s^h$  and  $1/T_p^e$ , calculated for various photon bursts and registration efficiencies 50%, 70%, and 90%. An additional dephasing rate smaller than  $1/\tau_r$  and probability of the forbidden transition smaller than  $0.001/\tau_r$  does not practically increase the error of our measurements.

In conclusion, our analysis shows that polarization of a single ES in a QD can be measured optically using an intense resonant light source that suppresses ES relaxation. We also propose using a combination of  $\pi$  pulses and transverse magnetic field for optical pumping of the ES's in QD's. The simplest protocol leading to ES orientation is a sequential combination of optical and magnetic  $\pi$  pulses. A high degree of ES polarization can also be achieved with a permanent transverse magnetic field by applying optical  $\pi$  pulses with repetition rate equal to an integer multiple of the magnetic precession frequency.

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