Fast Initialization of the Spin State of an Electron in a Quantum Dot in the Voigt Configuration

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We consider the initialization of the spin state of a single electron trapped in a self-assembled quantum dot via optical pumping of a trion level. We show that with a magnetic field applied perpendicular to the growth direction of the dot, a near-unity fidelity can be obtained in a time equal to a few times the inverse of the spin-conserving trion relaxation rate. This method is several orders of magnitude faster than with the field aligned parallel, since this configuration must rely on a slow hole spin-flip mechanism. This increase in speed does result in a limit on the maximum obtainable fidelity, but we show that for InAs dots, the error is very small.

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In a recent experiment [1], Atatüre and co-workers demonstrated high fidelity spin-state preparation of an electron in a self-assembled InAs/GaAs quantum dot (QD). Their purifying mechanism coupled the resonant laser excitation of the QD with heavy-light hole mixing to induce a small, but finite, degree of spin-flip Raman scattering. These experiments were performed with the magnetic field aligned in the growth direction of the QD (the Faraday configuration), and this was shown to be effective in suppressing deleterious spin flips caused by the nuclear hyperfine field.

Although fidelities very close to unity (≈ 99.8%) were obtained through this mechanism, for quantum information processing purposes one would also like state preparation to be fast. The speed of the scheme in Ref. [1] is limited by the rate of hole-mixing spin-flip trion relaxation, which was determined to have a characteristic time of \( \sim 1 \mu s \), corresponding to the measured rate of 100 kHz. This is slow compared with the picosecond time scale on which it is hoped that quantum operations will be performed in such dots [2–6].

It is the purpose of this Letter to show that a magnetic field aligned perpendicular, rather than parallel, to the growth axis allows the purification of the spin to near-unity fidelity with a characteristic time scale of \( 2\Gamma^{-1} = 1 \text{ ns} \), where \( \Gamma = 300 \text{ MHz} \) is the trion relaxation rate without spin flip as measured by Atatüre. This Voigt configuration is therefore some 3 orders-of-magnitude quicker than the Faraday configuration of Ref. [1].

The price paid for this dramatic speedup is that now both ground states are optically coupled to the trion. This inevitably leads to a reduction in the maximum obtainable fidelity, as it provides a path back for the population localized in the desired level. However, as we will show, this effect decreases with increasing field strength such that, for a typical InAs QD, the maximum obtainable fidelity typically differs from unity by only 0.3% at a field of 1 T and 0.005% at 8 T.

We consider a singly charged self-assembled InAs QD with growth direction \( z \). Figure 1 shows our four-level model that describes the pertinent features of the system. With \( B \) field aligned in the \( x \) direction, the Zeeman energy of a QD electron is \( \mathcal{H}_B = g_s^e \mu_B B_x s^e_x = E^e_B s^e_x \), where \( g_s^e \) is the electronic \( g \) factor, \( \mu_B \) is the Bohr magneton, \( B_x \) is the magnitude of the field, and \( s^e_x = \pm \frac{1}{2} \) corresponds to the electron ground states.

![FIG. 1 (color online). The four levels of the electron-trion system in the Voigt basis consists of two Zeeman-split single-electron ground states \( |x\pm\rangle \) with spins in the \( x \) direction, and two trion levels \( |\tau\pm\rangle \) with heavy-hole spins also in the \( x \) direction. Arrows indicate allowed optical transitions with \( H, V \) denoting two orthogonal linear polarizations. State preparation is achieved by resonantly pumping the \( V_1 \) transition. This populates the trion level \( |\tau_+\rangle \), which subsequently relaxes with rate \( \Gamma \) back to both ground states, resulting in a partial transfer of population from \( |x+\rangle \) to \( |x-\rangle \). This simple picture is complicated by the fact that the same laser also drives the \( V_2 \) transition, albeit off resonantly. This results in a small pumping of population in the opposite direction and hence a slight decrease in the maximum obtainable purity. We take the relaxation rate \( \Gamma = 1.2 \text{ \mu eV} \) and \( g \) factors \( g^e_s = -0.46 \) and \( g^h_s = -0.29 \). For a field of 1 T, the Zeeman splitting are then \( E^e_B = -27 \text{ \mu eV} \) and \( E^h_B = 17 \text{ \mu eV} \).](0031-9007/07/98(4)/047401(4))
electron spin. We have measured the magnitude of the electron \( g \) factor to be \( |g_{\text{e}}^0| = 0.46 \) [7], which is similar to values in the literature [1,8].

Our measurements also indicate that the behavior of the heavy-hole component of the trion in this field can be described with a Zeeman Hamiltonian \( \mathcal{H}^g_B = -g_B^h \mu_B B_x s_x^h = \mathcal{E}^h_B s_x^h \), where \( s_x^h = \pm 1/2 \) are the eigenvalues of a pseudospin, the components of which correspond to heavy-hole states aligned in the \( z \) direction, and \( g_B^h \) is the hole \( g \) factor, which we determine to have a magnitude of \( |g_B^h| = 0.29 \). Our measurements do not give us access to the signs of these two \( g \) factors, but here we take both to be negative as suggested by some recent results [1,9]. Our scheme relies neither on this assumption about the signs, nor indeed even on \( g_B^h \) being nonzero.

The four levels of our model are then the two electron ground states with spins in the \( x \) direction, \( \{|\pm\rangle\} \equiv 2^{-1/2}(\{|0\rangle \pm \{|1\rangle\}) \), where \( \{|0\rangle \} \) and \( \{|1\rangle\} \) represent electron spins in the \( z \) direction, and the two trion levels, \( \{\tau\pm\rangle\} \equiv 2^{-1/2}(\{|0\rangle \pm \{|1\rangle\}) (\{|\pm\rangle\} \pm \{|\mp\rangle\}) \), where \( \{|\pm\rangle\} \) and \( \{|\mp\rangle\} \) denote heavy-hole states also aligned in the \( z \) direction. Figure 1 shows the allowed optical transitions between these levels. These transitions are linearly polarized and we have defined the polarization vectors in terms of \( \sigma_x \) circular polarizations as \( V = 2^{-1/2}(\sigma_x + \sigma_y) \) and \( H = 2^{-1/2}(\sigma_x - \sigma_y) \). Our experiments [7] confirm that the level diagram of Fig. 1 provides an accurate description of the system up to a field strength of a few Tesla, and we expect this model to hold for even higher fields [10].

We drive the system with a \( V \)-polarized laser tuned on resonance with the transition from \( |x\rangle \) to \( |\tau\rangle \), which is denoted \( V_1 \) in Fig. 1. This illumination will also drive the \( V_2 \) transition and this off-resonant driving is the main source of nonideality considered in our model. We elect to drive the \( V_1 \) transition because, since we take the sign of both electron and hole \( g \) factors to be the same, the detuning of \( V_2 \) with respect to driving transition \( V_1 \) is \( \Delta_B = (g_B^e + g_B^h) \mu_B B_x = \mathcal{E}_x^e - \mathcal{E}_x^h \). The magnitude of this detuning is greater than that of \( \Delta_B = (g_B^e - g_B^h) \mu_B B_x = \mathcal{E}_x^e + \mathcal{E}_x^h \), which is the detuning of transition \( H_2 \) with respect to transition \( H_1 \). As we will show, the larger this detuning, the smaller the deleterious effects of the off-resonant transition [11].

In the rotating frame then, the Hamiltonian of our system with driven \( V_1 \) transition in the basis \( \{x\}, \{|\tau\rangle\}, \{|\sigma\rangle\}, \{|\sigma\rangle\} \) is

\[
\mathcal{H} = \begin{pmatrix}
0 & 0 & \Omega & 0 \\
0 & 0 & 0 & \Omega e^{-i\omega_1 t} \\
\Omega & 0 & 0 & 0 \\
0 & \Omega e^{i\omega_1 t} & 0 & 0
\end{pmatrix},
\]

where \( \Omega \) is the Rabi energy of the \( V_1 \) transition, which we assume to be independent of magnetic field strength and identical with that of the \( V_2 \) transition. We have set \( \hbar = 1 \). As this Hamiltonian shows, the laser drives not only the transition with which it is resonant, but also the unintended transition with terms oscillating with frequency \( \pm \Omega \). In writing this Hamiltonian, we have neglected hole mixing since it is both expected to be small [1], and can in any case be incorporated into the current scheme without significant change [12].

We will determine the properties of this system through the master equation for the density matrix \( \rho \) in the Lindblad form

\[
\dot{\rho} = -i[\mathcal{H}, \rho] + \sum_i \mathcal{L}_i[\rho],
\]

where the sum is over all trion relaxation channels, each of which is described by a Lindblad superoperator

\[
\mathcal{L}_i[\rho] = D_i \rho D_i^\dagger - \frac{1}{2} D_i^\dagger D_i \rho - \frac{1}{2} \rho D_i^\dagger D_i.
\]

Since the trion can relax through all four optical transitions shown in Fig. 1, we need to consider the four independent jump operators: \( D_1 = \sqrt{\Gamma}(|\tau\rangle\langle x| + |x\rangle\langle \tau|) \), \( D_2 = \sqrt{\Gamma}(|\tau\rangle\langle x| + |x\rangle\langle \tau|) \), \( D_3 = \sqrt{\Gamma}(|\tau\rangle\langle x| + |x\rangle\langle \tau|) \), \( D_2 = \sqrt{\Gamma}(|\tau\rangle\langle x| + |x\rangle\langle \tau|) \). In writing these operators, we have assumed that the relaxation channels proceed incoherently. This is justified since we will work in a regime where the Zeeman splittings are large enough that \( |\Delta_B|, |\Sigma_B| > \Omega \) and the degree of spontaneously generated coherence [13] is negligible. We also assume, for the sake of simplicity and ease of presentation, that the rate \( \Gamma \) is the same for all channels. On the time scales considered here, the hole-mixing spin-flip relaxation, central to the mechanism of Ref. [1], is negligible. Furthermore, since we will work at significant magnetic fields ( \( > 1 \) T), ground-state spin flips caused by the nuclear hyperfine interaction are suppressed, as demonstrated by Atatür et al. [1], and further effects of the nuclear spins are negligible, since they are operative over time scales far longer than our initialization time [14]. Finally, we assume that the initial state of the spin is unpolarized with \( \rho_{++} = \rho_{--} = 1/2 \) and all other elements of \( \rho \) zero.

Our elucidation of the properties of this system consists of two parts. First, we derive the time taken for the system to reach its asymptotic limit. This we do by neglecting the effects of the off-resonant transition. Second, we include the off-resonant effects and derive a limit on the maximum fidelity obtainable by this nonideality.

With a trion relaxation rate of \( \Gamma = 1.2 \mu \text{eV} \) and \( g \) factors as stated, then even with a small applied magnetic field, we work in a regime in which the detuning of the off-resonant transition \( |\Sigma_B| \) is much greater than both \( \Gamma \) and the Rabi energy \( \Omega \). In this limit we can assume that the terms \( \Omega e^{\pm i\omega_1 t} \) in the Hamiltonian of Eq. (1) oscillate sufficiently rapidly that they approximate as self-averaging to zero. In this case, the state \(|\tau\rangle\) decouples from the rest of the system and the Hamiltonian reduces to \( \mathcal{H} = \Omega(|x\rangle\langle \tau| + |\tau\rangle\langle x|) \). Physically this means that the off-resonant transition is so far off resonance that the laser
induces no transitions from it. We will derive a correction to this behavior later.

With this simplified Hamiltonian, there are only three independent nonzero density matrix elements to consider and these we organize into the vector \( \mathbf{v} = (\rho_{x^+,x^+}, \rho_{x^-,x^-}, \text{Im}\rho_{x^+,x^+}) \). We have utilized the normalization condition \( 1 = \text{Tr} \rho \) to eliminate \( \rho_{x^+,x^+} \).

The equation of motions for these components can then be rephrased in terms of this vector as
\[
\dot{\mathbf{v}} = \mathbf{X} \cdot (\mathbf{v} - \mathbf{v}_\infty),
\]
where
\[
\mathbf{X} = \begin{pmatrix} -\Gamma & -\Gamma & -2\Omega \\ -\Gamma & -\Gamma & 0 \\ 2\Omega & \Omega & -\Gamma \end{pmatrix},
\]
and \( \mathbf{v}_\infty = (0, 1, 0) \) is the stationary solution of this model, which represents the qubit population completely localized in state \( \ket{x^-} \) and hence 100% purified.

The time taken to reach this limit can be derived in the following way [15]. The solution of Eq. (4) is
\[
\mathbf{v}(t) = \mathbf{v}_\infty + e^{\mathbf{X}t}(\mathbf{v}_0 - \mathbf{v}_\infty)
\]
with initial vector \( \mathbf{v}_0 = (\frac{1}{2}, \frac{1}{2}, 0) \). In the long time limit this can be approximated as
\[
\mathbf{v} \sim \mathbf{v}_\infty + (\mathbf{v}_0 - \mathbf{v}_\infty)e^{-t/T_0}
\]
with the characteristic time defined through \( T_0^{-1} = \min(|\text{Re}(\lambda_i)|) \), where \( \lambda_i \) are the eigenvalues of matrix \( \mathbf{X} \), all of which have negative real parts. This characteristic time is found to be
\[
T_0 = \frac{3\lambda^{1/3}}{\Gamma} \left[ 3^{2/3}(1 - 4\epsilon^2) + 3^{1/3}\lambda^{2/3} - 3\lambda^{1/3} \right]^{-1}
\]
with \( \lambda = 9r^2 + \sqrt{192}r^6 - 63r^4 + 36r^2 - 3 \) and \( \epsilon = \Omega/\Gamma \). In Fig. 2, we plot this characteristic time as a function of the laser Rabi frequency in units of linewidth and the inset shows a typical evolution of the system and shows how well the behavior of the full system is approximated by Eq. (7) with \( T_0 \) as above.

The characteristic time \( T_0 \) has the following simple limits:
\[
T_0 = \begin{cases} \Gamma/\Omega^2 & \text{if } \Omega \ll \Gamma, \\ 2/\Gamma & \text{if } \Omega \gg \Gamma. \end{cases}
\]

If the driving is weak \( \Omega \ll \Gamma \) then the time to reach the asymptotic population is slow. However, for laser amplitudes greater than the relaxation rate, the characteristic time saturates at a value twice that of the trion lifetime. This makes sense since, in this limit, the speed of the system is limited by trion relaxation, in which case, one half of the spin population is transferred to \( \ket{x^-} \) in time \( \Gamma^{-1} \), whence \( T_0 = 2\Gamma^{-1} \). With the value \( \Gamma = 1.2 \mu eV \) we obtain \( T_0 = 1.1 \text{ ns} \), which is far shorter that the hole-mixing spin-flip transition time of \( \gamma^{-1} = 1.6 \mu s \) of Ref. [1]. Figure 2 also shows that this limit of \( T_0 = 2\Gamma^{-1} \) is a good approximation for all \( \Omega/\Gamma \approx 1 \).

We now consider the inclusion of the off-resonant transition, which acts to reduce the asymptotic value of \( \rho_{x^-} \) away from unity. The full Hamiltonian of Eq. (1) depends on time, and therefore we cannot simply set \( \dot{\rho} = 0 \) to find the asymptotic solutions. Rather, we proceed by making the following ansatz for the asymptotic density matrix elements [16]:
\[
\rho_{ij}(t \to \infty) = \rho^{(0)}_{ij} + \sum \rho^{(z)}_{ij} e^{z \Sigma_B t},
\]
where the coefficients \( \rho^{(0, z)}_{ij} \) are stationary. We place this ansatz into Eq. (2) and neglect terms oscillating as frequencies faster than \( \Sigma_B \). This results in a set of algebraic equation for the coefficients \( \rho^{(0, z)}_{ij} \) which we simply solve. We obtain the following expressions for the steady-state coefficients:
\[
\rho^{(0)}_{x^+,x^+} = \frac{\Gamma^2 + \Omega^2}{D};
\]
\[
\rho^{(0)}_{x^-,x^-} = 1 - \frac{\Gamma^2 + 3\Omega^2}{D};
\]
\[
\rho^{(0)}_{x^+,x^-} = \frac{\Omega^2}{D};
\]
\[
\rho^{(0)}_{x^-,x^+} = i\Gamma \Omega/D;
\]
\[
\rho^{(z)}_{x^-,x^-} = (\rho^{(0)}_{x^-,x^+})^* = \Omega (i\Gamma - \Sigma_B)/D,
\]
where the denominator \( D = \Sigma_B^2 + 2\Gamma^2 + 4\Omega^2 \), and all the other coefficients are zero.
Let us define the fidelity of the state preparation as $F = \langle \Psi | \rho | \Psi \rangle$, where $| \Psi \rangle$ is the desired target state with population localized in the state $| x- \rangle$ and $\rho$ is the actual density matrix of the final states. This evaluates simply as $F = \rho_{x-,x-}^{(0)} = \rho_{x-,x-}^{(0)}$ and starts with a value of $\frac{1}{2}$ in the initial unpolarized state, and is unity for 100% purification \[17\]. Let us define as $\epsilon$ the amount by which $F$ differs from unity: $\epsilon = 1 - F$. From Eqs. (11), we therefore find that the state-preparation error is

$$\epsilon = \frac{\Gamma^2 + 3\Omega^2}{\Sigma_B^2 + 2\Gamma^2 + 4\Omega^2} = \frac{\Gamma^2 + 3\Omega^2}{\Sigma_B^2},$$

where we have made use of $| \Sigma_B | \gg \Omega, \Gamma$.

In Fig. 3 we plot the full result for $\epsilon$ as a function of the Rabi frequency. The most salient point is that for a field of the order of 1 T, and with $\Omega/\Gamma = 1$, the error $\epsilon$ is of the order of $3 \times 10^{-3}$, which is very small, and of the order of the measurement threshold described in Ref. [1]. Increasing the field, decreases the error and at a high laboratory field such as 8 T the error is reduced to $\epsilon = 5 \times 10^{-5}$. These estimates agree very well with the results of numerical integration of the equations of motion. It should be noted that these values apply while the CW illumination is still in effect. Turning off the laser allows population trapped in the trion states to relax back to the ground-state sector with rate $\Gamma$. Half of this population ends up in the required state $| x- \rangle$, reducing the error by a factor of 2/3.

In summary then, we have considered the advantages of using the Voigt configuration for the preparation of the spin state of an electron in a self-assembled QD. Provided that the Rabi frequency of the laser is greater than the trion relaxation rate, the state preparation is fast, proceeding with a time scale of $2\Gamma^{-1} \approx 1$ ns, which is orders of magnitude faster than in the Faraday configuration. Use of the Voigt configuration does, however, impose an upper limit on the maximum obtainable fidelity, but this is small, with the deviation from unity being $\epsilon = (T^2 + 3\Omega^2)/\Sigma_B^2 \approx 10^{-3}$ at 1 T. This approach therefore represents a fast way of initializing an electron spin to high fidelities for quantum information processing.

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[11] If the $g$ factors had opposite signs, we would have $| \Delta_B | > | \Sigma_B |$ and therefore driving one of the $H$ transitions instead would be the best strategy to minimize initialization error.
[12] The dominant hole mixing here is between heavy hole $| 1; \pm \frac{1}{2} \rangle$ and light hole $| 1; \pm \frac{3}{2} \rangle$. By adjusting the relative phase between circular-polarized components of the applied laser, the effect of this mixing can be reduced to only altering the Hamiltonian of Eq. (1) through a rescaling of the Rabi frequency.
[17] In the limit that $1 - \rho_{x-,x-} \ll 1$, this fidelity is approximately the same as the definition $F = 1 - \rho_{x+,x+}/\rho_{x-,x-}$ used in Ref. [1] [M. Atatüre (private communication)].