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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.

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 ($\geq 99.8\%$) 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 ~ 1 μs, 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 fidelities with a characteristic time scale of $2\Gamma^{-1} \approx 1$ ns, where $\Gamma = 300$ 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 $\mathcal{H}_B^e = g_x^e \mu_B B_x s_x^e \equiv E_B^e s_x^e$, where g_x^e is the electronic g factor, μ_B is the Bohr magneton, B_x is the magnitude of the field, and $s_x^e = \pm 1/2$ corresponds to the
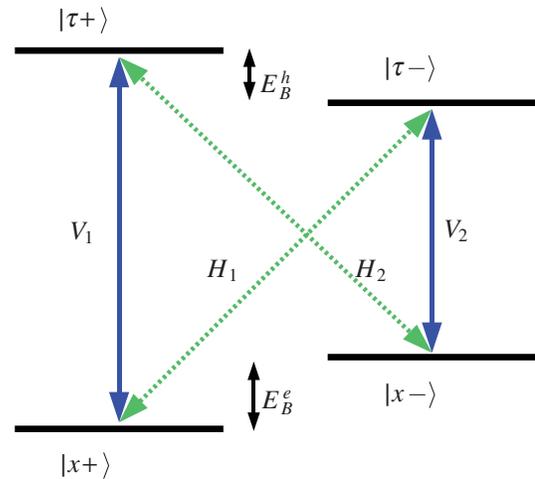
FIG. 1 (color online). The four levels of the electron-trion system in the Voigt basis consists of two Zeeman-split single-electron ground states $|x\pm\rangle$ with spins in the x direction, and two trion levels $|\tau\pm\rangle$ with heavy-hole spins also in the x direction. Arrows indicate allowed 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 Γ 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 $\Gamma = 1.2$ μeV and g factors $g_x^e = -0.46$ and $g_x^h = -0.29$. For a field of 1 T, the Zeeman splitting are then $E_B^e = -27$ μeV and $E_B^h = 17$ μeV.

electron spin. We have measured the magnitude of the
electron g factor to be $|g_x^e| = 0.46$ [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 $\mathcal{H}_B^h =
-g_x^h \mu_B B_x s_x^h \equiv E_B^h s_x^h$, where $s_x^h = \pm 1/2$ are the eigenval-
ues of a pseudospin, the components of which correspond
to heavy-hole states aligned in the x direction, and g_x^h is the
hole g factor, which we determine to have a magnitude of
$|g_x^h| = 0.29$. 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_x^h being nonzero.

The four levels of our model are then the two electron
ground states with spins in the x direction, $|x\pm\rangle \equiv
2^{-1/2}(|\downarrow\rangle \pm |\uparrow\rangle)$, where $|\downarrow\rangle$ and $|\uparrow\rangle$ represent electron spins
in the z direction, and the two trion levels, $|\tau\pm\rangle \equiv
2^{-1}(|\downarrow\uparrow\rangle - |\uparrow\downarrow\rangle)(|\Downarrow\rangle \pm |\Uparrow\rangle)$, where $|\Downarrow\rangle$ and $|\Uparrow\rangle$ 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 $\sigma_\pm$ cir-
cular polarizations as $V = 2^{-1/2}(\sigma_- + \sigma_+)$ and $H =
2^{-1/2}(\sigma_- - \sigma_+)$. 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 detun-
ing of V_2 with respect to driving transition V_1 is $\Sigma_B =
(g_x^e + g_x^h)\mu_B B_x = E_x^e - E_x^h$. The magnitude of this detun-
ing is greater than that of $\Delta_B = (g_x^e - g_x^h)\mu_B B_x = E_x^e +
E_x^h$, 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 sys-
tem with driven V_1 transition in the basis
$\{|x+\rangle, |x-\rangle, |\tau+\rangle, |\tau-\rangle\}$ is

$$
\mathcal{H} = \begin{pmatrix}
0 & 0 & \Omega & 0 \\
0 & 0 & 0 & \Omega e^{-i\Sigma_B t} \\
\Omega & 0 & 0 & 0 \\
0 & \Omega e^{i\Sigma_B t} & 0 & 0
\end{pmatrix}, \tag{1}
$$

where Ω 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 $\pm\Sigma_B$. 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 ρ in the
Lindblad form

$$
\dot{\rho} = -i[\mathcal{H}, \rho] + \sum_i \mathcal{L}_i[\rho], \tag{2}
$$

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

$$
\mathcal{L}_i[\rho] = D_i \rho D_i^\dagger - \tfrac{1}{2} D_i^\dagger D_i \rho - \tfrac{1}{2} \rho D_i^\dagger D_i. \tag{3}
$$

Since the trion can relax through all four optical tran-
sitions shown in Fig. 1, we need to consider the four
independent jump operators: $D_1 = \sqrt{\Gamma}|x+\rangle\langle\tau+|$, $D_2 =
\sqrt{\Gamma}|x-\rangle\langle\tau+|$, $D_3 = \sqrt{\Gamma}|x+\rangle\langle\tau-|$, and $D_4 = \sqrt{\Gamma}|x-\rangle \times
\langle\tau-|$. 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 presen-
tation, that the rate Γ is the same for all channels. On the
time scales considered here, the hole-mixing spin-flip re-
laxation, central to the mechanism of Ref. [1], is negligible.
Furthermore, since we will work at significant magnetic
fields ($\gtrsim 1$ T), ground-state spin flips caused by the nu-
clear hyperfine interaction are suppressed, as demonstrated
by Atatüre *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 ρ 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 imposed by this nonideality.

With a trion relaxation rate of $\Gamma = 1.2$ μeV 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 Γ and
the Rabi energy Ω. In this limit we can assume that the
terms $\Omega e^{\pm i\Sigma_B t}$ 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 $\mathcal{H} =
\Omega|x+\rangle\langle\tau+| + \text{H.c.}$ 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 $\mathbf{v} = (\rho_{x+,x+}, \rho_{x-,x-}, \mathrm{Im}\rho_{x+,\tau+})$. We have utilized the normalization condition $1 = \mathrm{Tr}\rho$ to eliminate $\rho_{\tau+,\tau+}$.

The equation of motions for these components can then be rephrased in terms of this vector as

$$\dot{\mathbf{v}} = \mathbf{X} \cdot (\mathbf{v} - \mathbf{v}_\infty),\tag{4}$$

where

$$X = \begin{pmatrix} -\Gamma & -\Gamma & -2\Omega \\ -\Gamma & -\Gamma & 0 \\ 2\Omega & \Omega & -\Gamma \end{pmatrix},\tag{5}$$

and $\mathbf{v}_\infty = (0, 1, 0)$ is the stationary solution of this model, which represents the qubit population completely localized in state $|x-\rangle$ 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

$$\mathbf{v}(t) = \mathbf{v}_\infty + e^{\mathbf{X}t}(\mathbf{v}_0 - \mathbf{v}_\infty)\tag{6}$$

with initial vector $\mathbf{v}_0 = (\frac{1}{2}, \frac{1}{2}, 0)$. In the long time limit this can be approximated as

$$\mathbf{v} \sim \mathbf{v}_\infty + (\mathbf{v}_0 - \mathbf{v}_\infty)e^{-t/T_0}\tag{7}$$

with the characteristic time defined through $T_0^{-1} = \min\{|\mathrm{Re}(\lambda_i)|\}$, where $\{\lambda_i\}$ are the eigenvalues of matrix $\mathbf{X}$, all of which have negative real parts. This characteristic time is found to be

$$T_0 = \frac{3\lambda^{1/3}}{\Gamma}[3^{2/3}(1 - 4r^2) + 3^{1/3}\lambda^{2/3} - 3\lambda^{1/3}]^{-1}\tag{8}$$

with $\lambda = 9r^2 + \sqrt{192r^6 - 63r^4 + 36r^2 - 3}$ and $r = \Omega/\Gamma$. 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:

$$T_0 = \begin{cases} \Gamma/\Omega^2 & \text{if } \Omega \ll \Gamma, \\ 2/\Gamma & \text{if } \Omega \gg \Gamma. \end{cases}\tag{9}$$

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 $|x-\rangle$ in time Γ^{-1}, whence $T_0 = 2\Gamma^{-1}$. With the value $\Gamma = 1.2\ \mu$eV we obtain $T_0 \approx 1.1$ ns, which is far shorter that the hole-
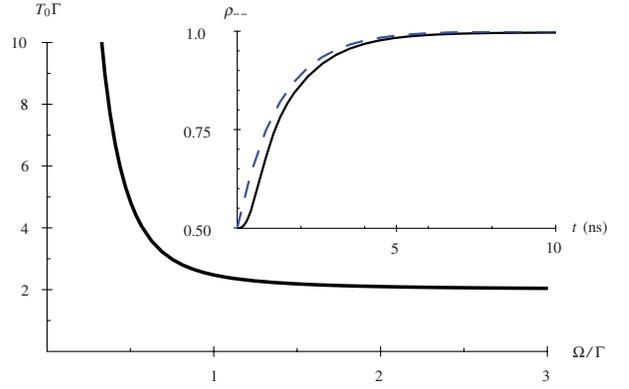
FIG. 2 (color online).　The characteristic time T_0 describing the approach of the system to its asymptotic limit as a function of the Rabi frequency Ω. When $\Omega/\Gamma \gtrsim 1$, $T_0 \approx 2/\Gamma$. Inset: The population of the state $|x-\rangle$ as a function of time under continuous illumination. The solid black line shows the result of numerical integration of the master equation, whereas the dashed blue line shows the analytic result of Eq. (7). Near-unity fidelity is approached with a characteristic time of $T_0 \approx 1$ ns. For both plots, the parameters are the same as in Fig. 1 and for the inset we have further set $\Omega = \Gamma$.

mixing spin-flip transition time of $\gamma^{-1} \approx 1.6\ \mu$s of Ref. [1]. Figure 2 also shows that this limit of $T_0 = 2\Gamma^{-1}$ is a good approximation for all $\Omega/\Gamma \gtrsim 1$.

We now consider the inclusion of the off-resonant transition, which acts to reduce the asymptotic value of ρ_{--} 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]:

$$\rho_{ij}(t \to \infty) = \rho_{ij}^{(0)} + \sum_{\pm}\rho_{ij}^{(\pm)}e^{\pm i\Sigma_B t},\tag{10}$$

where the coefficients $\rho_{ij}^{(0,\pm)}$ are stationary. We place this ansatz into Eq. (2) and neglect terms oscillating as frequencies faster than Σ_B. This results in a set of algebraic equation for the coefficients $\rho_{ij}^{(0,\pm)}$ which we simply solve. We obtain the following expressions for the steady-state coefficients:

$$\rho_{x+,x+}^{(0)} = \frac{\Gamma^2 + \Omega^2}{D};$$
$$\rho_{x-,x-}^{(0)} = 1 - \frac{\Gamma^2 + 3\Omega^2}{D}$$
$$\rho_{\tau-,\tau-}^{(0)} = \rho_{\tau+,\tau+}^{(0)} = \Omega^2/D;\tag{11}$$
$$\rho_{x+,\tau+}^{(0)} = i\Gamma\Omega/D;$$
$$\rho_{x-,\tau-}^{(-)} = (\rho_{\tau-,x-}^{(+)})^* = \Omega(i\Gamma - \Sigma_B)/D,$$

where the denominator $D = \Sigma_B^2 + 2\Gamma^2 + 4\Omega^2$, and all the other coefficients are zero.
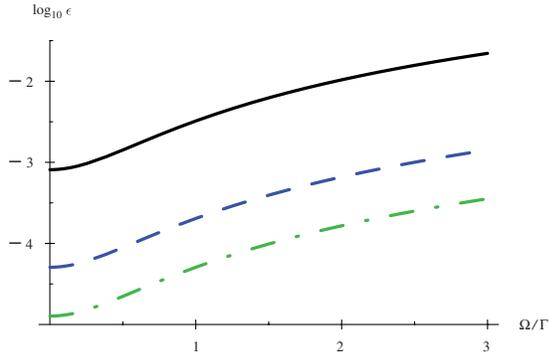
FIG. 3 (color online). The logarithm of the error $\epsilon = 1 - F$, with F the fidelity, as a function of the Rabi energy Ω. Results are shown for three different fields: 1 T (black, solid), 4 T (blue, dash), and 8 T (green, dash-dot). Other parameters as Fig. 1.

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 ρ is the actual density matrix of the final states. This evaluates simply as $F = \rho_{x-,x-} = \rho_{x-,x-}^{(0)}$ 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 ϵ the amount by which F differs from unity: $\epsilon = 1 - F$. From Eqs. (11), we therefore find that the state-preparation error is

$$\epsilon = \frac{\Gamma^2 + 3\Omega^2}{\Sigma_B^2 + 2\Gamma^2 + 4\Omega^2} \approx \frac{\Gamma^2 + 3\Omega^2}{\Sigma_B^2}, \qquad (12)$$

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

In Fig. 3 we plot the full result for ϵ 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 ϵ is of the order of 3×10^{-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 Γ. 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 $\epsilon \approx (\Gamma^2 + 3\Omega^2)/\Sigma_B^2 \approx 10^{-3}$ at 1 T. This approach therefore represents a fast way of initializing an electron spin to high fidelities for quantum information processing.

[1] M. Atatüre, J. Dreiser, A. Badolato, A. Högele, K. Karrai, and A. Imamoglu, Science **312**, 551 (2006).

[2] C. Piermarocchi, Pochung Chen, L. J. Sham, and D. G. Steel, Phys. Rev. Lett. **89**, 167402 (2002).

[3] T. Calarco, A. Datta, P. Fedichev, E. Pazy, and P. Zoller, Phys. Rev. A **68**, 012310 (2003).

[4] B. W. Lovett, A. Nazir, E. Pazy, S. D. Barrett, T. P. Spiller, and G. A. D. Briggs, Phys. Rev. B **72**, 115324 (2005).

[5] P. Chen, C. Piermarocchi, L. J. Sham, D. Gammon, and D. G. Steel, Phys. Rev. B **69**, 075320 (2004).

[6] C. Emary and L. J. Sham, cond-mat/0608518; cond-mat/0609094.

[7] Xiaodong Xu *et al.* (unpublished).

[8] I. Hapke-Wurst, U. Zeitler, R. J. Haug, and K. Pierz, Physica (Amsterdam) **12E**, 802 (2002).

[9] M. Bayer, O. Stern, A. Kuther, and A. Forchel, Phys. Rev. B **61**, 7273 (2000).

[10] M. V. Gurudev Dutt, Jun Cheng, Bo Li, Xiaodong Xu, Xiaoqin Li, P. R. Berman, D. G. Steel, A. S. Bracker, D. Gammon, S. E. Economou, Ren-Bao Liu, and L. J. Sham, Phys. Rev. Lett. **94**, 227403 (2005).

[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 $|\frac{3}{2}; \pm \frac{3}{2}\rangle$ and light hole $|\frac{3}{2}; \mp \frac{1}{2}\rangle$. 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.

[13] S. E. Economou, R.-B. Liu, L. J. Sham, and D. G. Steel, Phys. Rev. B **71**, 195327 (2005).

[14] A. Greilich, D. R. Yakovlev, A. Shabaev, Al. L. Efros, I. A. Yugova, R. Oulton, V. Stavarache, D. Reuter, A. Wieck, and M. Bayer, Science **313**, 341 (2006).

[15] B. Michaelis, C. Emary, and C. W. J. Beenakker, Europhys. Lett. **73**, 677 (2006).

[16] T. H. Stoof and Yu. V. Nazarov, Phys. Rev. B **53**, 1050 (1996).

[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)].