Fast Spin State Initialization in a Singly Charged InAs-GaAs Quantum Dot by Optical Cooling

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(Received 1 February 2007; published 28 August 2007)

Quantum computation requires a continuous supply of rapidly initialized qubits for quantum error correction. Here, we demonstrate fast spin state initialization with near unity efficiency in a singly charged quantum dot by optically cooling an electron spin. The electron spin is successfully cooled from 5 to 0.06 K at a magnetic field of 0.88 T applied in Voigt geometry. The spin cooling rate is of order 10^9 s^{-1}, which is set by the spontaneous decay rate of the excited state.

DOI: 10.1103/PhysRevLett.99.097401 PACS numbers: 78.67.Hc, 33.80.Ps, 42.50.Hz

The use of electron spins in semiconductor quantum dots (QDs) as quantum bits (qubits) is being widely explored for quantum information and quantum computation (QIQC) [1–6]. A key element for QIQC is the initial quantum state preparation. QIQC requires not only qubits initialized in a known state for computation and gate operations, but also a continuous supply of low-entropy ancillary qubits for quantum error correction (QEC) [7,8]. A fault-tolerant quantum computation requires about 10^4 quantum operations before the qubits lose their coherence [7,8]. This requirement demands that the state initialization speed must be much faster than the quantum state decoherence rate.

The spin relaxation time ($T1$) of an electron spin trapped in a self-assembled In(Ga)As QD has been measured to be on the order of 20 ms [3], which sets an upper limit for the spin decoherence time in the absence of other interactions (e.g., phonon scattering, hyperfine interaction). The initialization of an electron spin state has been demonstrated recently in a singly charged QD by applying magnetic fields in the Faraday geometry [2]. Although near unity fidelity is successfully achieved, the initialization rate is about 3×10^5 s^{-1} [9], making it challenging for this scheme to satisfy the QEC requirement. A fast state initialization method with a high efficiency is essential for practical QEC processes.

In this Letter, we demonstrate fast spin state initialization (optical pumping or laser cooling of an electron spin [2]) in a singly charged InAs/GaAs self-assembled QD (SAQD) in the presence of a magnetic field in the Voigt geometry. The fast spin cooling with near unity efficiency is achieved at the spin cooling rate of about 10^9 s^{-1} via the resonant excitation of a trion state. The spin state is cooled from 5 to 0.06 K at a magnetic field of 0.88 T at a rate considerably faster than the spin decoherence rate.

The sample under study contains InAs SAQDs embedded in a Schottky diode structure. An aluminum mask with 1 μm apertures on the surface provides the spatial resolution to study a single QD. The number of electrons in a QD can be controlled by varying the bias voltages across the sample. The experiment is performed at 5 K. Figure 1(a) displays a bias dependent photoluminescence (PL) intensity map of a single QD showing the charging effects. The state of interest is a single negatively charged exciton (X^-), which is called a trion and about 5.5 meV lower in energy than the neutral exciton state. Details regarding the physics of the bias dependent PL and discussion of the transition assignments are found in Ref [10–12].

The projection of the hole angular momentum on the sample growth direction determines the angular momentum of the trion state to be $|\pm\frac{3}{2}\rangle$. At zero magnetic field, the electron spin ground states $|\pm\frac{1}{2}\rangle$ are twofold degenerate. The only dipole allowed transitions are from the spin states $|\pm\frac{1}{2}\rangle$ to the trion states $|\pm\frac{3}{2}\rangle$ with σ^+ and σ^- excitations, respectively. Figure 1(b) shows the spin flip

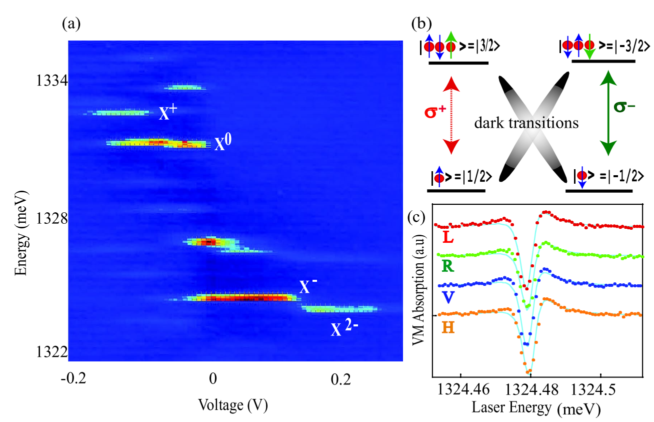

FIG. 1 (color online). (a) Bias dependent PL intensity map of a single InAs SAQD, where $X^{n\pm}$ denotes the charged exciton with n excess electrons ($-$) or holes ($+$). (b) Trion energy level diagram at zero magnetic field. (c) Polarization dependent VM spectra of a singly charged QD at zero magnetic field. L, R, V, and H denote left circular, right circular, vertical, and horizontal polarization, respectively. Curves are the fittings of the data.

Raman scattering transitions are ideally dark [13]. The imperfect selection rules will lead to finite spontaneous emission rates for the dark transitions [2], which ultimately limit the spin cooling speed under these conditions [14].

Figure 1(c) shows a polarization dependent study on a trion by voltage modulation (VM) spectroscopy [15] with a modulation amplitude about a quarter of the trion linewidth. No fine structure splitting was observed [16]. Analysis of the line shape shows an asymmetry that could arise from some kind of interference. Independent experiments [17] have suggested it arises from a coupling of the hole to the cap layer when the thickness of the cap layer exceeds a critical value. The effect, still under investigation, complicates the exact determination of the linewidth, but does not interfere with the qualitative features described below. Fitting yields a trion linewidth of 1.5 GHz.

In order to implement a fast spin state initialization, the dark transitions have to become bright, since the optical pumping rate depends on the spin flip Raman scattering process. This can be realized by applying a magnetic field perpendicular to the sample growth direction [001]. Figure 2(a) shows the VM absorption map [18,19] as a function of the applied bias at a magnetic field of 0.88 T along the [110] axis. The laser field is linearly polarized and 45° to the polarization axis ($\vec{\rho}$) of the quantum dot. In bias region II, the optical pumping rate is larger than the spin relaxation rate. Fast spin cooling is demonstrated, where the absorption of the laser beam is strongly suppressed by optical pumping. In region I, cotunneling (the tunneling of the electron between the quantum dot and the Fermi sea [2,20]) induced spin relaxation rate is comparable or larger than the optical pumping rate, so the depletion of the spin ground states is not achieved. Thus, the strong suppression of the absorption disappears in region I and a quartet transition pattern appears. The physics of the two bias regions is discussed below along with the bias dependent g factor associated with transition $H1$ and $H2$.

We first start with the bias region I of Fig. 2(a). As a magnetic field is applied along the [110] axis, it induces off diagonal terms in the Hamiltonian that couple both electron and hole states. The coupling leads to linearly polarized transitions from the spin ground states to the trion states. Figure 2(b) shows the corresponding four level trion model, where $|x\pm\rangle$ ($|t\pm\rangle$) are the spin (trion) eigenstates in the magnetic field. The Zeeman splitting of the electron spin (trion) states is $|g_{e\perp}\mu_B B_x|$ ($|g_{h\perp}\mu_B B_x|$), where $g_{e\perp}$ ($g_{h\perp}$) is the electron (hole) spin in-plane g factor, μ_B is Bohr magneton, and B_x is the applied magnetic field. The four linearly polarized transitions are labeled as $V1$, $H1$, $H2$, and $V2$, where $\overrightarrow{V1}\|\overrightarrow{V2} \perp \vec{\rho}$ and $\overrightarrow{H1}\|\overrightarrow{H2}\|\vec{\rho}$.

Our measurements confirm that the dark transitions become bright and all four transitions are linearly polarized. However, the polarization axis ($\vec{\rho}$) of the QD is not parallel or perpendicular to the applied magnetic field direction, rather it is 45° to $\vec{B}$ in the X-Y plane for this

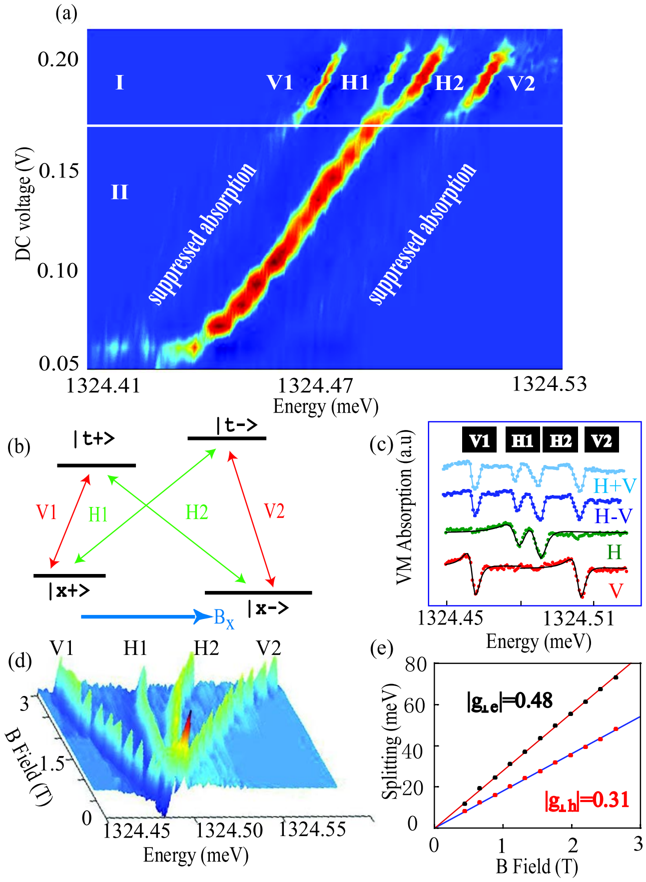

FIG. 2 (color online). (a) Bias dependent VM absorption map of a singly charged SAQD at a magnetic field of 0.88 T. The laser is 45° polarized. Voltage region I shows all four trion transitions. Spin state preparation is achieved in Voltage range II. (b) Energy level diagram of a trion. The gate voltage is set at 0.19 V for plots (c), (d), and (e). (c) Polarization dependent VM spectra of a singly charged QD at magnetic fields 0.88 T. The black curves are the fittings. (d) 3d plot of the trion evolution with various magnetic fields. The laser is 45° polarized. The data are inverted for clarity. (e) The electron (black dots) and hole (orange dots) Zeeman splitting as a function of the magnetic fields.

particular QD ($\vec{\rho}$ may vary from dot to dot). This observation indicates the existence of heavy and light hole mixing in our QDs caused by the QD in-plane anisotropy. The mixing effects observed here agree with the previous reports on the CdSe/ZnSe SAQDs [21]. The hole mixing only affects the intermediate trion states, but not the spin ground states. The only change introduced by the mixing effect is that $\vec{\rho}$ rotates away from $\vec{B}$, which is not essential to our optical pumping scheme. For simplicity, the light polarized along (perpendicular to) $\vec{\rho}$ is referred to as horizontally (vertically) polarized.

Figure 2(c) shows the polarization study of the trion state at gate voltage 0.19 V. A quartet transition pattern is excited with a 45° linearly polarized light. When the light is vertically (horizontally) polarized, the optical field only excites the outer (inner) two transitions of the quartet.

Thus, the inner and outer transitions are strictly linearly polarized and orthogonal to each other, which inhibits spontaneously generated coherence (SGC) [13]. The observation of the quartet demonstrates that under a transverse magnetic field, all four trion transitions are optically allowed and obey well defined polarization selection rules.

The evolution of the trion states as a function of the magnetic field is illustrated with the fan diagram in Fig. 2(d). The trion states start with a single peak at zero magnetic field and split into four lines at finite magnetic fields. The energy difference between transitions $V1$ and $H1$ ($H2$) corresponds to the hole (electron) Zeeman splitting. The electron and hole Zeeman splittings are plotted in Fig. 2(e) as a function of the applied magnetic fields. The linear fittings yield $|g_{e\perp}|$ and $|g_{h\perp}|$ of 0.48 and 0.31, respectively. Although the absolute signs of $|g_{e\perp}|$ and $|g_{h\perp}|$ are not identified experimentally, we are able to tell that $|g_{e\perp}|$ and $|g_{h\perp}|$ have the same sign.

In region II of Fig. 2(a), spin relaxation is inhibited. When the laser beam is resonant with transition $V1$, as shown in Fig. 3(a), the electron spin in $|x+\rangle$ state will be excited to the trion state $|t+\rangle$ and then relax equally to the two spin ground states as suggested by the comparable absorption strengths and linewidths. That is to say, because the spin flip resonant Raman scattering process is now allowed in the Voigt profile, the optical induced spin flip process is dramatically "sped up", ensuring fast spin cooling. Since the electron spin in the ground state has a much slower relaxation rate than the trion spontaneous decay rate, the electron spin will be optically pumped into the $|x-\rangle$ spin state within a few radiative cycles. The signature of optical pumping is that transition $V1$ becomes transparent to the laser beam. The preparation of $|x+\rangle$ works in a similar way. This is clearly demonstrated in bias range II of Fig. 2(a). When the laser is on resonance with $V1$ ($V2$), the absorption is strongly suppressed and the transition becomes transparent. Thus, the polarized spin states can be selectively prepared in either the $|x-\rangle$ or $|x+\rangle$ spin state.

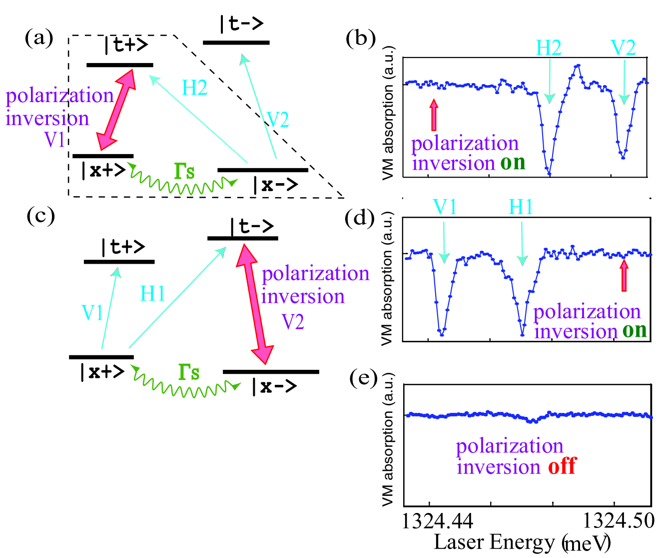

FIG. 3 (color online). Demonstration of the spin state preparation in $|x+>$ ($|x->$) state at magnetic field 1.32 T and gate voltage 0.12 V. The scanning laser is 45° polarized and the PIB is vertically polarized. (a) and (b) [(c) and (d)]. The PIB is resonant with the transition $V1$ ($V2$) while probing the transitions $H2$ and $V2$ ($H1$ and $V1$). (e) One beam absorption spectrum of the trion state in the absence of the PIB. The absorption of transitions $V1$ and $V2$ are strongly suppressed due to the optical pumping effect. Since the degeneracy of transitions $H1$ and $H2$ are lifted by increasing the magnetic field to 1.32 T, the central peak is also suppressed.

The mechanism of the spin state preparation can also be explained in terms of saturation spectroscopy. A simple rate equation calculation for a three level lambda system gives the saturation intensity for a trion transition as $I_{SAT} \simeq I_{SAT_0}\frac{\Gamma_s}{\Gamma}$, where I_{SAT_0} is the trion saturation intensity at zero magnetic field, Γ_s is the spin relaxation rate, and Γ is the trion spontaneous decay rate. The spin relaxation time $\frac{1}{\Gamma_s}$ has been reported to be on the order of tens of milliseconds [3]. It is much longer than the trion decay time $\frac{1}{\Gamma}$, which is about a few hundred picoseconds [20]. Therefore, in the presence of the transverse magnetic fields, the saturation intensity I_{SAT} of the trion system could be about 6 to 7 orders of magnitude weaker than I_{SAT_0}. Thus, the trion transition is easily saturated and becomes transparent to the optical beam.

The data in the transition region from I to II in Fig. 2(a) show the signature of a bias dependent electron g factor, which leads to transitions $H1$ and $H2$ evolving from two well-resolved lines in region I into a central absorption peak in region II. Since transitions $H1$ and $H2$ are nearly degenerate in region II, when the laser is on resonance with transition $H1$, it is also nearly resonant with $H2$. Therefore, the optical pumping effect is partially canceled by the bidirectional pumping induced by the same optical field. Hence, the optical pumping effect is suppressed and results in the central absorption peak. The origin of this behavior remains under investigation, but it is likely that the strong bias dependence is more complex than the bias dependent g factors reported earlier in quantum wells [22] and for hole in QDs [23]. Fortunately, the behavior does not impact the main qualitative conclusion of the work.

In order to prove that the laser beam leads to nearly complete spin polarization and prepares the spin state as $|x-\rangle$ ($|x+\rangle$) by pumping transition $V1$ ($V2$), a polarization inversion beam (PIB) is tuned to be on resonance with transition $V2$ ($V1$). As shown in Fig. 3(a), while the PIB is tuned to be on resonance with transition $V1$, it repolarizes the spin ground states prepared by pumping transition $V2$ (i.e., redistributes the population between the spin ground states). This leads to the recovery of the absorption peaks at transitions $V2$ and $H2$. Figure 3(c) and 3(d) show that transitions $V1$ and $H1$ can also be recovered by tuning the PIB to be on resonance with transition $V2$. Considering that the spin cooling process prepares a low-entropy polarized spin state, depending on the intensity of the PIB, the effect of the PIB is to increase the entropy of the system by generating a mixed spin ground state at low intensity, or reversing the spin polarization at high intensity.

The excited state decay rate is 2π times the absorption linewidth in the absence of pure dephasing or spectral wandering. Nearly degenerate differential transmission (NDT) is particularly sensitive to these latter two effects [24,25]. We have performed NDT on the trion state (data not shown), and while the data are complicated by the effect of the Fano interference, they show no evidence of either significant pure dephasing or spectral wandering.

The optical pumping rate is analyzed by the method developed in Ref. [14]. When the laser is on resonance with one trion transition, the four level model can be reduced to a three level lambda system, as shown in the dashed box in Fig. 3(a). To keep the discussion general, the trion decay rates to the two spin ground states $|x+\rangle$ and $|x-\rangle$ are Γ_{t+x+} and Γ_{t+x-}, respectively. The optical pumping rate is calculated to be

$$\lambda = \frac{-\Gamma_{t+x-}}{6b\kappa^{1/3}}(3^{2/3} + 2\times 3^{2/3}b + 3^{2/3}b^2 - 16\times 3^{2/3}r^2 - 3\kappa^{1/3} - 3b\kappa^{1/3} + 3^{1/3}\kappa^{2/3}), \quad (1)$$

$\kappa = 72r^2 + \frac{1}{3}[46\,656r^4 - 27(1+2b+b^2-16r^2)^3]^{1/2}$, where $r = \frac{\Omega}{\Gamma_{t+x+}}$, Ω is the Rabi frequency, the branching ratio b is $\frac{\Gamma_{t+x-}}{\Gamma_{t+x+}}$. If we take $b = 1$ and $r \gg 1$, the optical pumping rate is $\Gamma_{t+x-}/2$, as shown in Ref. [14].

Using the line shape data in Fig. 3, a linewidth of order 1.2 GHz (1.5 GHz), in the absence of pure dephasing or spectral wandering, corresponds to Γ_{t+x+} (Γ_{t+x-}) of $7.5\times 10^9\ \mathrm{s}^{-1}$ ($9.4\times 10^9\ \mathrm{s}^{-1}$). In the spin state preparation experiment, the Rabi frequency of the pump beam approximately equals Γ_{t+x+}. Thus, by inserting $b = 1.25$ and $r = 1$ into Eq. (1), we infer an optical pumping rate of order $4\times 10^9\ \mathrm{s}^{-1}$. The optical pumping rate inferred from the measured linewidth may be an upper limit if the linewidth is broadened by the spectral diffusion process [16]. The hole coupling to the continuum states might also result uncertainty in the radiative lifetime (at most a factor of 3) but not affect the qualitative speed up by a few orders of magnitude due to the magnetic field induced state mixing. Using a more conservative trion relaxation rate of $1\times 10^9\ \mathrm{s}^{-1}$ reported by Ref [26], our scheme infers an spin state initialization rate of order $5\times 10^8\ \mathrm{s}^{-1}$.

The Voigt profile introduces a limitation to the optical pumping efficiency [14]. This arises from the fact that transitions $V1$ and $V2$ have the same polarization selection rules. When the laser is resonant with transition $V1$ ($V2$), transition $V2$ ($V1$) will be off-resonantly coupled, which causes a small amount of the spin population to be pumped back. As mentioned above, this type of off-resonant coupling is responsible for the central absorption peak in Fig. 2(a). At a magnetic field of 0.88 T, a spin state preparation efficiency of $(98.9 \pm 0.4)\%$ (the error comes from the measurement noise) is achieved experimentally, which corresponds to a spin temperature of 0.06 K [2,27,28]. This demonstrates laser cooling of an electron spin from 5 (the experimental temperature) to 0.06 K in a singly charged QD. To reach the same efficiency at 5 K by thermal equilibration, the applied magnetic field would need to be 69 T at a much slower initialization speed as well.

The authors would like to thank Dr. Luming Duan for helpful discussions. This work is supported by the U. S. ARO, AFOSR, ONR, NSA/LPS, and FOCUS-NSF.

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