Pulsed-gate measurements of the singlet-triplet relaxation time in a two-electron double quantum dot

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A pulsed-gate technique with charge sensing is used to measure the singlet-triplet relaxation time for nearly degenerate spin states in a two-electron double quantum dot. Transitions from the (1,1) charge occupancy state to the (0,2) state, measured as a function of pulse cycle duration and magnetic field, allow the (1,1) singlet-triplet relaxation time (≈70 μs) and the (0,2) singlet-triplet splitting to be measured. This technique can be readily applied to read out a spin-qubit operating in a singlet-triplet basis.

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Semiconductor quantum dots are promising systems for the manipulation of electron spin because of the relative ease of confining and measuring single electrons. A spin memory based on electron-spin orientation requires a long spin-relaxation time, $T_1$, for coherent manipulation of electronic-spin states, the spin dephasing time $T_2$ is the important figure of merit because it sets the time scale in which coherent operations must be performed. In order to make use of the spin degree of freedom as a holder of either classical or quantum information, it is first necessary to understand and characterize the mechanisms that lead to spin relaxation and decoherence.

Previous studies of spin relaxation in quantum dots have focused on systems with large spin splittings. Relaxation times for spin states separated by a singlet-triplet splitting $E_{ST} \approx 600 \mu$eV have been shown to approach 200 μs. Several groups have measured spin relaxation for Zeeman split spin states at high fields ($E_{Zeeman} \approx 200 \mu$eV), demonstrating long $T_1$ times. Since the readout techniques used in these experiments require coupling to the leads, the spin splitting must be larger than the thermal energy for accurate spin-state readout. At dilution refrigerator temperatures, this implies that $E_{Zeeman} > k_B T \sim 10 \mu$eV, or $B > 0.5 \text{T}$. For $B > 0.5 \text{T}$, $T_1$ shows a strong field dependence, as is expected theoretically for spin-orbit-mediated spin-relaxation processes.

Hyperfine interactions, coupled with phonon emission, can also lead to spin relaxation. The hyperfine interaction results in an effective magnetic field, $B_{\text{nucl}} \sim 4 T/\sqrt{N_k} \sim 2 \text{mT}$, where $N_k \sim 5 \times 10^6$ is the effective number of nuclei interacting with the electron spin for typical GaAs quantum dots.

When the splitting between spin states is on the order of, or less than, the hyperfine energy scale, the hyperfine interaction can result in electron-spin relaxation. Since $B_{\text{nucl}} \sim 2 \text{mT} \sim 0.05 \mu$eV $< k_B T$, a spin readout that can distinguish between nearly degenerate spin states is required to measure $T_1$ in this regime.

In this paper we describe an experimental technique that allows measurements of the singlet-triplet relaxation time for nearly degenerate two-electron spin states. This technique can be used to measure $T_1$ in regimes for which hyperfine-mediated relaxation processes are expected to be important. Spin relaxation between nearly degenerate electronic states is particularly relevant to the problem of controlled entanglement, as the singlet-triplet splitting goes to zero as the entangled spins become spatially separated. We also note that the technique presented in this paper can be easily extended to read out a spin qubit based on singlet-triplet states.

Measurements are performed using a few-electron double quantum dot fabricated from a GaAs/Al$_{0.33}$Ga$_{0.67}$As heterostructure grown by molecular-beam epitaxy. Electron-beam lithography and liftoff are used to create Ti/Au gates that deplete a 100-nm deep two-dimensional electron gas with electron density $2 \times 10^{11} \text{cm}^{-2}$ and mobility $2 \times 10^5 \text{cm}^2/\text{V}\cdot\text{s}$. Gates 2–6 and 12 form the double quantum dot [see Fig. 1(a)]. Gates 2 and 6 are connected via bias tees to dc voltage sources and to pulse generators through coaxial cables with $\sim 20 \text{dB}$ of inline attenuation. A quantum point contact (QPC) charge detector is created by depleting gate 1. Gates 7–11 are unused. The QPC conductance, $G_S$, is measured using standard lock-in amplifier techniques with a 1-nA current bias at 93 Hz. The electron temperature, $T_e \sim 135 \text{mK}$, was determined from Coulomb blockade peak widths.

QPC charge sensing is used to determine the absolute number of electrons in the double dot. Figure 1(b) shows a large-scale charge stability diagram for the double dot. As electrons enter or leave the double dot, or transfer from one dot to the other, $G_S$ changes, resulting in sharp features in $dG_S/dV_e$ (numerically differentiated). In the lower left corner of Fig. 1(b), the double dot is completely empty. As the gate voltages are made more positive, electrons are added to the double dot. We will focus on the two-electron regime near the (1,1) to (0,2) charge transition (integer pairs specify the equilibrium charge occupancy on the left and right dot) [Fig. 1(e)].

In the two-electron regime, charge transport in a double dot shows a striking asymmetry in bias voltage due to spin-selection rules (Pauli blocking). This asymmetry is due to the different singlet-triplet splittings in the (1,1) and (0,2)
charge states. Spin states of the weakly coupled (1,1) charge configuration are nearly degenerate, while the (0,2) spin states are separated by a ~400 μeV singlet-triplet splitting, J. At forward bias, transitions from the (0,2) singlet state, (0,2)S, to the (1,1) singlet state, (1,1)S, are allowed. However, for reverse bias, (1,1) to (0,2) transitions can be blocked if the (1,1) state forms a triplet (1,1)T because the (0,2)T state resides outside the transport window due to the large singlet-triplet splitting in (0,2). This asymmetry results in current rectification, which will be used in the present pulsed-gate experiment for spin to charge conversion.

Charge transitions are driven by applying pulses to gates 2 and 6. Pulse heights are calibrated by applying pulses to a single gate and measuring the charge stability diagram. Figure 1(d) shows a charge stability diagram acquired with square pulses applied to gate 2 (Vp2 = 25 mV, 50% duty cycle, period τ = 10 μs). This results in two copies of the charge stability diagram, the right-most (left-most) charge stability diagram reflects the ground-state charge configuration during the low (high) stage of the pulse sequence. The gate-voltage offset between the charge stability diagrams, ΔV, is used to calibrate pulse amplitudes. Due to attenuation in the coax cables, ΔV is less than the pulse amplitude at the signal generator, Vp2. Additional calibrations are performed for gate 6, which primarily shifts the honeycomb in the vertical direction (not shown). The charge stability diagram can be shifted in any direction in gate space by simultaneously applying calibrated pulses to gates 2 and 6.

In a double dot, charge can be pumped by pulsing gates around a triple point, e.g. (0,1)\rightarrow(1,1)\rightarrow(0,2)\rightarrow(0,1). Our spin-relaxation measurement technique relies on the fact that (1,1)T to (0,2)S transitions are spin blocked. Measuring this charge transition probability as a function of time using charge sensing allows a T1 measurement. Two control experiments, discussed below, demonstrate that the observed time dependence of the charge sensing signal is due to spin-blocked transitions.

\begin{align*}
T_1 & \text{ is measured using a forward pulse sequence (0,1)\rightarrow(1,1)\rightarrow(0,2)\rightarrow(0,1) as shown in [Fig. 2(a)]. The sequence begins with the gates at point E for 10% of the period, emptying the second electron from the double dot, leaving the (0,1) charge state. The gates then shift to the reset point R for the next 10% of the period, which initializes the system into the (1,1) configuration. The interdot tunnel coupling, t, is tuned with } t \approx k_BT \text{ so that the (1,1) singlet-triplet splitting } J \sim 4k_BT/U, \text{ where } U \text{ is the single dot charging energy. Due to this degeneracy, and at low fields such that }
\end{align*}
where only the singlet state is accessible, and the transitions defined by the charge states, respectively. The pulse data differ from transitions on a time scale given by the interdot tunneling rate, \( \Gamma(t) \) [we estimate the slowest \( \Gamma(e) \) (1 \( \mu s \)) from finite bias data\(^8\)]. If the \( m_s=1 \) (1,1) triplet state (1,1)\(_T\) is loaded, it dephases into (1,1)\(_S\) on a time scale of \( T_2 \) (expected to be \( \approx 100 \) ns). (Refs. 10, 17, and 18) followed by a direct transition to (0,2)\(_S\). About half the time the R step will load the \( m_s=1 \) (1,1) triplet state (1,1)\(_T\) or the \( m_s=-1 \) (1,1) triplet state (1,1)\(_T\). At low \( B_z \), (0,2)\(_T\) is inaccessible, and a transition from (1,1)\(_S\) or (1,1)\(_T\) to (0,2) requires a spin flip and will be blocked for times shorter than the singlet-triplet relaxation time \( \tau_{ST} \).

In Fig. 2(a) the average charge sensor signal, \( G_S \), is measured as a function of the dc gate voltages \( V_2 \) and \( V_6 \), while a pulse sequence is repeated. This has the effect of translating the points \( E, R, \) and \( M \) throughout the charge stability diagram, keeping their relative positions constant. Because most of the time is spent at point \( M \), the grayscale data primarily map out the ground-state population for this point, with plateaus at \( G_S \approx 0.0, 6.0, 16, \) and \( 23 \times 10^{-3} \) \( e^2/h \) indicating full population of the (1,2), (0,2), (1,1), and (0,1) charge states, respectively. The pulse data differ from ground-state data only when point \( M \) resides in the triangle defined by the (1,1) to (0,2) ground-state transition and the extensions of the (1,1) to (0,1) and (1,1) to (1,2) ground-state transitions [bounded by the red marks in Fig. 2(a)]. Within this “pulse triangle” transitions from (1,1) to (0,2) may be blocked as described above, and the charge sensor registers a conductance intermediate between the (1,1) and (0,2) plateaus. If \( M \) moves above the pulse triangle [red dot in Fig. 2(a)], the (1,1) to (0,2) transition can occur sequentially via (1,2) with no interdot tunneling: a new electron enters the right dot, then the electron in the left dot leaves. Likewise, if \( M \) moves below the pulse triangle [orange dot in Fig. 2(a)] the transition can occur via (0,1): the left-dot electron leaves, then a new electron enters the right dot. By similar logic, point \( R \) must be to the left of the (0,1) to (0,2) transition extension [dotted line in Fig. 2(a)] to avoid resetting through (0,2) and preferentially loading (1,1)\(_S\). Figure 2(a) shows a signal of \( 11 \times 10^{-3} \) \( e^2/h \) in the pulse triangle for \( \tau=10 \) \( \mu s \), which indicates that approximately 50% of the time the dots remain in (1,1) even though (0,2) is the ground state. This is direct evidence of spin-blocked (1,1) to (0,2) transitions.

As a control, we compare the forward \( T_1 \) pulse sequence with a reverse pulse sequence that does not involve spin-selective transitions (0,1) \( \rightarrow \) (0,2) \( \rightarrow \) (1,1) \( \rightarrow \) (0,1). With the pulse sequence reversed the reset position \( R \) occurs in (0,2) where only the singlet state is accessible, and \( M \) occurs in (1,1). Now tunneling from \( R \) to \( M \) should always proceed on a time scale set by the interdot tunneling coupling, since the (0,2)\(_S\) to (1,1)\(_S\) transition is not spin blocked. As anticipated, no signal is seen in the pulse triangle for this reversed “control” sequence [Fig. 2(c)].

Spin selectivity of the forward pulse sequence in Fig. 3 can be used to measure \( J \) as a function of \( B_\perp \) (Ref. 19). This also confirms that the charge sensing signal in the pulse triangle is due to spin-blocked interdot charge transitions. Figure 3(a) shows \( G_S \) as a function of \( V_2 \) and \( V_6 \) while applying the forward pulse sequence with \( B_\perp =1.2 \) T and \( \tau=10 \) \( \mu s \). For these data, (0,2)\(_T\) resides outside of the pulse triangle \( (J>E_M, \) the mutual charging energy) and the (1,1)\(_T\) to (0,2) transitions are spin blocked. For \( B_\perp =1.4 \) T [Fig. 3(b)] the (0,2)\(_T\) state is low enough in energy that the (1,1)\(_T\) states can directly tunnel to the (0,2)\(_T\) manifold at high detunings. Now (1,1) to (0,2) tunneling can proceed, and there is no longer a (1,1) charge signal in the (0,2) region of the pulse triangle at high detuning. This cuts off the tip of the pulse triangle. The spin-blocked region continues to shrink as \( B_\perp \) is increased. From these data, we find \( J \approx 340, 280, \) and \( 180 \) \( \mu eV \) for \( B_\perp =1.4, 1.6, \) and 1.8 T, respectively\(^20\).

The time dependence of the charge sensing signal can be investigated by varying \( \tau \), the overall period of the cycle. Figure 4(a) shows \( G_S \) as a function of \( V_2 \) and \( V_6 \) acquired using the forward pulse sequence with \( \tau=8 \) \( \mu s \) at \( B_\perp =100 \) mT. A clear pulse signal is observed in the pulse triangle. As \( \tau \) is increased, the pulse signal decreases as shown in (b) and (c). \( G_S \) is measured inside the pulse triangle \( (V_2, V_6 \) held fixed at \( -403, -523.8 \) mV, respectively) and is plotted as a function of \( \tau \) in Fig. 4(d). In (1,1), \( G_S \approx 20 \times 10^{-3} \) \( e^2/h \), whereas outside the pulse triangle in (0,2), \( G_S \approx 10 \times 10^{-3} \) \( e^2/h \). For small \( \tau \), \( G_S \approx 15 \times 10^{-3} \) \( e^2/h \) in the pulse triangle. At long \( \tau \), \( G_S \) approaches \( 10 \times 10^{-3} \) \( e^2/h \) in the pulse triangle, which indicates complete transfer from the (1,1) to (0,2) charge state.

These data are consistent with spin-blocked transitions preventing the (1,1) to (0,2) charge transition. Approximately 50% of the time, the (1,1) \( R \) pulse loads into either (1,1)\(_T\) or (1,1)\(_S\). These states may relax into the (1,1)\(_S\) state and then tunnel to (0,2)\(_S\) on a time scale set by \( \tau_{ST} \). For \( \tau\ll\tau_{ST} \), we...
time, resulting in a pulse signal of 15/200849 pulse periods, the data in the triangle. The and the expected as a function of gate voltage position indicated by the black triangle in =100 mT. Color online to the time-averaged occupation of the left dot. Modeling an exponential decay of the sensing signal weighted over 80% of the cycle corresponding to the (0,2) measurement gives \[ G_S(\tau) = A + B(\tau_{ST}\tau)(1 - e^{-0.8\pi\tau_{ST}}), \] where \( A \) is the conductance asymptote at long times [full occupation of the (0,2) state] and \( B \) is the additional conductance in a short pulse due to the blocked states [approximately 50% of the (0,2) to (1,1) step height]. The best fit to the data in Fig. 4(d) gives \( A = 0.009 e^2/h \) and \( B = 0.007 e^2/h \), consistent with these expectations. In the center of the pulse triangle the best-fit \( \tau_{ST} \) reaches a maximum of 70±10 \( \mu \)s. Near the (1,1) to (0,2) transition, \( \tau_{ST} \) decreases to 20±5 \( \mu \)s at \( V_2 = -403.8 \text{ mV} \) and \( V_b = -523.0 \text{ mV} \). Closer to the tip of the pulse triangle, \( \tau_{ST} \) decreases due to thermally activated exchange with the leads (see Fig. 2, red and orange diagrams), thus the 70-\( \mu \)s relaxation time represents a lower bound on the spin-relaxation time within the (1,1) manifold. This technique can be used to explore the full dependence of the spin-relaxation time on detuning and magnetic field.

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12. We use Anritsu K251 bias tees. Tektronix AWG520 pulse generators are used for high speed manipulation of the gate voltages.
20. The conversion from gate voltage to energy is determined by measuring finite bias triangles.
21. \( \Gamma(e) \) is set to be much faster than the characteristic relaxation times that we measure in this experiment. \( \Gamma(e) \) varies strongly with \( e \) (see Ref. 13). The fact that \( \tau_{ST} \) only varies by roughly a factor of 3 in the pulse triangle also indicates that the (1,1)–(0,2) transitions are spin blocked and not limited by a slow \( \Gamma(e) \).