Single-Shot Measurement of Triplet-Singlet Relaxation in a Si/SiGe Double Quantum Dot


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We investigate the lifetime of two-electron spin states in a few-electron Si/SiGe double dot. At the transition between the (1,1) and (0,2) charge occupations, Pauli spin blockade provides a readout mechanism for the spin state. We use the statistics of repeated single-shot measurements to extract the lifetimes of multiple states simultaneously. When the magnetic field is zero, we find that all three triplet states have equal lifetimes, as expected, and this time is $\sim 10$ ms. When the field is nonzero, the $T_\text{0}$ lifetime is unchanged, whereas the $T_-$ lifetime increases monotonically with the field, reaching 3 sec at 1 T.

The device is fabricated on a phosphorus-doped Si/Si$_{0.97}$Ge$_{0.03}$ heterostructure with a strained Si quantum well approximately 75 nm below the surface. Palladium surface gates labeled 1–9 in Fig. 1(a) are used to form the double quantum dot at $B_{\parallel} = 1$ T. These long times are achieved up until now.

FIG. 1 (color online). (a) Scanning electron microscopy (SEM) image of a device identical to the one used. Quantum dots are formed at the approximate locations of the two circles. Charge sensing is performed by monitoring the current $I_{\text{QPC}}$ through a nearby point contact. (b) Charge stability diagram of the double dot showing the detuning voltage $V_\epsilon$. (c) Energies of two-electron states as a function of detuning energy $\epsilon$. $T_+$, $T_0$, and $T_-$ are the (1,1) triplets; the (0,2) triplets are higher in energy. The (1,1) and (0,2) singlets $S_{11}$ and $S_{02}$ are coupled by spin-preserving, interdot tunneling. A magnetic field separates the triplet energies by $E_z = g \mu_B B$. (d) Time-averaged occupation of the (0,2) charge state $P_{02}$ at $B_{\parallel} = 0$ with 5 kHz square pulses of peak-to-peak amplitude $\Delta V_\epsilon$ applied along $V_\epsilon$. The pulses drive (1,1)-(0,2) transitions within the dotted triangle. The suppression of $P_{02}$ above the dashed line shows where (1,1) to (0,2) tunneling is suppressed by spin blockade.
The ground state of the system is blocked into a spin blockaded configuration [13–15], where the (1,1)-(0,2) transition as a function of detuning energy $\epsilon$, where the transition is at $\epsilon = 0$ [12]. The detuning energy is controlled by varying the voltages on gates 2 and 4 along $V_{\epsilon}$, shown in Fig. 1(b). The interdot tunnel coupling $t_c$ was measured by determining where the $S_{11}$ and $T_-$ states cross at finite $B_{\parallel}$. This is shown as $\epsilon_{\text{mix}}$ in Fig. 1(c), and depends on both $B_{\parallel}$ and the curvature of the avoided singlet crossing. Using this approach [6], we find $t_c = 2.8 \pm 0.3$ $\mu$eV (677 $\pm$ 73 MHz).

To measure the spin of a (1,1) state we pulse the system into a spin blocked configuration [13–15], where the ground state of the system is $S_{02}$ and the (0,2) triplet states are higher in energy than all of the (1,1) triplets: $T_-, T_0, T_+$. We characterize the parameters needed to reach this configuration by detecting spin blockade in the time-averaged measurement shown in Fig. 1(d). Square pulses at 5 kHz are applied along $V_{\epsilon}$. The color scale in Fig. 1(d) shows the time-averaged probability $P_{02}$ of finding the system in (0,2) as a function of pulse amplitude and offset along $V_{\epsilon}$. When the pulse crosses the (1,1)-(0,2) transition, tunneling between charge states occurs in results in $0 < P_{02} < 1$. The region where this occurs is bounded by the dotted triangle in Fig. 1(d). Spin blockade occurs in the part of the pulse triangle that is above the dashed white line in Fig. 1(d). Here we see $0 < P_{02} < 0.5$, because the system is residing in (1,1) the majority of the time.

Spin blockade does not occur below the white dashed line in Fig. 1(d), resulting in $P_{02} = 0.5$. In this region the pulse amplitude exceeds the (0,2) singlet-triplet splitting energy $E_{\text{ST}}$, and the pulse offset is such that the (0,2) triplet states have lower energy than the (1,1) triplets. From the size of the blockaded region, and the conversion from detuning voltage $V_{\epsilon}$ to detuning energy $\epsilon$ ($\Delta \epsilon = \Delta V_{\epsilon} \times 0.0676$ eV/V, see [11] for additional details), we find $E_{\text{ST}} = 124 \pm 4$ $\mu$eV.

Figures 2(a) and 2(b) show single-shot initialization and readout of (1,1) singlet and triplet states using real-time measurement of the charge state while pulsing across the (1,1)-(0,2) transition. The system is initialized by starting from the ground state $S_{02}$ at $0 < \epsilon < E_{\text{ST}}$. The occupation of $S_{02}$ is verified by measuring the charge state: $S_{02}$ is the only (0,2) state accessible at this detuning. We then pulse to $\epsilon < 0$ to transfer the prepared $S_{02}$ to the (1,1) singlet $S_{11}$. To measure the (1,1) spin state at some later time, we pulse back to $0 < \epsilon < E_{\text{ST}}$ where a singlet can tunnel quickly to (0,2) but the triplets cannot. The measurements are performed using detuning pulses with two levels that are at the positions of the filled triangle and circle in Fig. 2(c), which correspond to detuning energies of $\epsilon = -160$ $\mu$eV and 60 $\mu$eV, respectively, at $B_{\parallel} = 0$.

We measure the lifetimes of the (1,1) singlet and triplet states by detecting the spin state as we repeatedly pulse back and forth across the (1,1)-(0,2) transition at a frequency of 300 Hz. Figures 2(d)–2(f) show real-time measurements of the charge state as the pulses are applied. In
this regime spin blockade is active and the system switches randomly between free shuttling of a singlet state and blockade of a (1,1) triplet state. The typical length of time spent in a blockaded triplet increases dramatically as $B_{||}$ increases. Figure 2(g) is a control, demonstrating that charge shuttles freely in both directions when the pulse is offset to reach outside the spin-blockade regime.

To determine the lifetimes of the states at $B_{||} = 0$ we plot in Figs. 3(a) and 3(b) the number of times that blockaded periods of duration $t_b$, and unblockaded periods of duration $t_u$ are observed in 6.4 minutes of data (115 200 pulse periods). The histograms are very well fit by exponential decays, and fits to the two distributions give characteristic times of $\tau_u = 9.6 \pm 0.2$ ms for the blockaded configuration and of $\tau_u = 23 \pm 3$ ms for the unblockaded configuration. From these times we find that the lifetimes of the spin states are $\sim 10$ ms, using a rate-equation model that we describe below.

The $B_{||} = 0$ lifetimes are 2 orders of magnitude longer than have been seen in comparable low-field measurements of GaAs quantum dots [8,9]. We suggest that this is due to the small hyperfine coupling in natural silicon, arising from the high abundance of zero-spin nuclei. At $B_{||} = 0$, the (1,1) triplets are degenerate and separated from the singlet-triplet mixing times may be different in the two configurations of the system. The characteristic time of blockade is the mixing time as arising from the contact-hyperfine interaction with nuclear spins [16–18]. Predictions for the hyperfine coupling of (1,1) spin states are $h \sim 3$ neV in silicon [18], compared to measured values of $h \sim 50$ neV in GaAs [8,19]. The expected coupling is small enough that, in our measurements, it would be exceeded by the exchange splitting $J$. Given $t_c$ and the pulse amplitude, hyperfine induced singlet-triplet mixing should be suppressed by a factor of $[1 + (J/h)^2] \sim 500$, compared to the maximum mixing rate when $J \ll h$.

The values $\tau_u$ and $\tau_b$ are determined by the rate of singlet-triplet mixing, but they do not directly correspond to mixing times in any static configuration of the system. This is because the pulses continuously switch between two configurations, one at $\epsilon < 0$ and one at $\epsilon > 0$. The singlet-triplet mixing times may be different in the two configurations, and at $\epsilon > 0$ there are also fast, one-way transitions from $S_{11}$ to $S_{02}$. We relate the measured values of $\tau_u$ and $\tau_b$ to singlet-triplet mixing times in the two configurations of the system by using rate equations to model state occupations during a single pulse cycle. The inputs to the model are 2 times; one time $\tau_+$ is the mixing time when the ground state is $S_{11}$ during the $\epsilon < 0$ half of the pulse, and the other time $\tau_-$ is the mixing time when the ground state is $S_{02}$ during the $\epsilon > 0$ half of the pulse. Tunneling between $S_{11}$ and $S_{02}$ is assumed to be instantaneous. Mixing during the pulse transitions is ignored because the period of the pulse is $10^3$ times larger than the pulse rise time. We solve for $\tau_+$ and $\tau_-$ by numerical optimization of the model to match the measured values of $\tau_u$ and $\tau_b$ (see [11] for additional details). We find $\tau_- = 24.5 \pm 3$ ms and $\tau_+ = 5.8 \pm 0.3$ ms. We attribute the difference between $\tau_+$ and $\tau_-$ to a difference in $t_c$ between the two halves of each pulse cycle.

As $B_{||}$ increases from 0 T, we observe a qualitative change in the spin dynamics: the statistics of the blockaded durations show two separate characteristic times. As shown
in Figs. 3(c) and 3(e), there are short blockaded periods with a characteristic time $\tau_p$, that is field independent, and there are longer blockaded periods whose characteristic time $\tau_e$ increases with field. The 2 times arise because the system can be blockaded if it is in either a $T_0$ or a $T_-$ state, and the $T_+$ has a field dependent energy, whereas the $T_0$ does not. The $T_+$ state does not play a role at $B_\parallel > 0$ because its higher energy means that it is rarely populated. Combined with statistics of unblockaded durations, as in Fig. 3(d), each measurement at $B_\parallel > 0$ can contain simultaneously information about the lifetimes of three states: $S_{1,1}, T_-, T_0$.

Figure 3(f) shows the $T_-$ lifetime $\tau_T$ and $S_{1,1}$-$T_0$ mixing times $\tau_+ \tau_-$ calculated from the data in Fig. 3(e). We find $\tau_+ \tau_-$ from $\tau_+$ and $\tau_0$, using a rate-equation model similar to the zero field case, but with no transitions to $T_+$ and $T_-$ included. This is because mixing from the $S_{1,1}$ or $T_0$ to the $T_-$ and $T_+$ will be suppressed due to their separation in energy. At $B_\parallel \approx 0.5$ T, the system spends so much time in the $T_-$ state that it is impractical to collect enough statistics to accurately determine $\tau_T$. Within the range of $B_\parallel$ where $\tau_0$ can be measured, the $S_{1,1}-T_0$ mixing rates are largely independent of field and similar to the rates seen at $B_\parallel = 0$.

The time $\tau_T$ is the lifetime of the $T_-$ during the $\epsilon > 0$ half of the pulse and is well approximated as $\tau_T = 2\tau_0 / 1$. At high magnetic fields. During the $\epsilon < 0$ half of the pulse, $T_-$ is the ground state and it will remain populated with high probability when $g\mu B_\parallel > k_BT$. In the $\epsilon > 0$ half of the pulse the $T_-$ is the first excited state and can decay to the $S_{0,2}$ ground state at a rate of $\tau_T^{-1}$. Such transitions could be induced by phonons and a spin nonconserving process such as hyperfine coupling [8,16,17] or spin-orbit coupling [20–23]. We find that the $T_-$ lifetime $\tau_T$ increases strongly with field, rising to 3 sec by $B_\parallel = 1$ T. This is consistent with single-spin lifetimes measured at similar magnetic fields [1–4].

In summary, we have shown that we can initialize the singlet-triplet qubit state into a singlet and subsequently measure, in single-shot mode, transitions to the (1,1) triplet states. Using this initialization and real-time measurement, we have measured the lifetime of singlet and triplet states versus magnetic field. When the magnetic field is zero, the lifetime for the singlet and all three triplets is $\sim 10$ ms. When the magnetic field is nonzero, the $T_0$ and $S_{1,1}$ lifetimes are almost unchanged, whereas the $T_-$ lifetime grows significantly, reaching 3 sec at 1 T.

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