Single-shot readout and relaxation of singlet/triplet states in exchange-coupled $^{31}\text{P}$ electron spins in silicon

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We present the experimental observation of a large exchange coupling $J \sim 300 \mu eV$ between two $^{31}\text{P}$ electron spin qubits in silicon. The singlet and triplet states of the coupled spins are monitored in real time by a Single-Electron Transistor, which detects ionization from both energy- and tunnel-rate-dependent processes in the coupled spin system, yielding single-shot readout fidelities above 95%. The triplet to singlet relaxation time $T_1 \sim 1$ ms at zero magnetic field agrees with the theoretical prediction for $J$-coupled $^{31}\text{P}$ dimers in silicon. The ability to follow the time evolution of the 2-spin state populations at different magnetic fields gives further insight into the dynamics of the coupled donors system, and the role of hyperfine interactions. These results pave the way to the realization of 2-qubit quantum logic gates with spins in silicon, and highlight the necessity to adopt gating schemes compatible with weak $J$-coupling strengths.

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Entangling two-qubit operations, together with single-qubit rotations, form a universal set of quantum logic gates for circuit-based quantum computing [1]. These have been demonstrated in several physical qubit platforms [2], including spins in semiconductors [3–5]. The best qubit coherence times in the solid state have been obtained with spins in isotopically purified group-IV materials such as silicon [6, 7] and carbon [8, 9]. Silicon offers the additional advantage of being the material that underpins all of modern computer hardware, which makes it a very appealing candidate for spin-based quantum technologies [10–12]. The coherent operation of spin-based qubits in Si has been demonstrated in single $^{31}\text{P}$ donor atoms [13, 14] and double quantum dots [15, 16]. Conversely, an entangling quantum logic gate for a pair of spin qubits in silicon is still awaiting experimental demonstration. Several coupling mechanisms can be used for this purpose, including magnets [17] and microwave photons [18], but the simplest coupling for a pair of spins is the exchange interaction $J$, arising from the overlap of electron wavefunctions [19, 20].

Exchange interaction between pairs of donors in silicon has been observed in bulk spin resonance experiments [21] and, very recently, by electron transport experiments through a donor molecule [22]. However its application to quantum information processing requires the ability to dynamically control it, and to measure the instantaneous quantum state of the qubits. Here we report the time-resolved observation of large exchange coupling $J \sim 300 \mu eV$ between the electrons of a $^{31}\text{P}$ donor pair. The $^{31}\text{P}$ pair is integrated within a top-gated silicon Single-Electron Transistor (SET) [23] that we employ to perform single-shot readout of the spin singlet ($|S\rangle = (|↑↓⟩ - |↓↑⟩) / \sqrt{2}$) and triplet ($|T_−⟩ = |↓↓⟩$, $|T_0⟩ =$ ($|↑↓⟩ + |↓↑⟩) / \sqrt{2}$, $|T_+⟩ = |↑↑⟩$) states of the two-electron system. Operating the device at low electron temperatures ($T_{el} = 125 \pm 25 \text{ mK}$) allows us to perform energy-selective readout (E-RO) [24, 25]. In addition we exploit the significant difference in the size of the orbital wavefunctions for $|S⟩$ and $|T⟩$ states to demonstrate tunnel-rate-selective readout (TR-RO) [26]. We apply these techniques to measure the $|T⟩ \rightarrow |S⟩$ relaxation time $T_1$, and its dependence on the external magnetic field $B$.

![FIG. 1. (Color online) a. Scanning electron microscope image of a device similar to the one used in the experiments. The gates TG, LB, RB along with the S, D diffusion regions make up the Single Electron Transistor. Inset shows a sketch of our system of two electron spins with overlapping wavefunctions. b. Charge stability diagram, showing two closely spaced charge transitions (dashed lines).](image)

The device described in this work was obtained from the same batch of devices as the one described in detail by Pla et al. [13]. A near-intrinsic [001] natural silicon substrate was implanted with phosphorus ions, energy 14 keV [27], using a surface mask to obtain $\sim 3$ ions (Poison statistics) in a 30 nm $\times$ 30 nm window. The donor electrons are tunnel-coupled to the island of a SET, induced under a SiO$_2$ layer by a stack of Al gates consisting...
of a top gate (TG) and two (left and right) barriers gates (LB and RB) [23], see Figure 1a. The electrochemical potential $\mu_D$ of the donor electrons can be varied using a donor gate (DG) above the implant window. This architecture [28] (Figure 1a) allows for charge detection with very high signal-to-noise ratio [25].

By scanning the TG and DG voltages ($V_{TG}$ and $V_{DG}$) while measuring the SET current $I_{SET}$, we obtain the charge stability diagram shown in Figure 1b, where a characteristic pattern of conductance peaks is observed. Ionization of a nearby donor causes the potential of the SET island to shift, generating a discontinuity in the pattern of $I_{SET}$ peaks [25, 28]. The ionization takes place when $\mu_D$ is raised above the Fermi energy $E_F$ of the SET island. A first hint of the presence of two closely spaced donors is the presence of two nearby transitions in the charge stability diagram in Figure 1b, indicating that two donors change their charge state at similar values of electrostatic potentials.

With $V_{DG}$ set near a donor charge transition, the device is tuned so the SET will switch between $I_{SET} = 0$ (Coulomb-blockade) and $I_{SET} \neq 0$, when the system is neutral or ionized, respectively. In the measurement shown in Figure 2a, we use a 3-level single-shot spin readout sequence [24] consisting of load, read and empty phases. During the read-phase, we measure $I_{SET}$ while varying $V_{DG}$ such that the $\mu_D$ goes from higher to lower than $E_F$. As seen in Figure 2a, there is a well-defined “tail” where excess current occurs at the start of the read-phase. This indicates the presence of an energy-split pair of electron states. The high-energy electron tunnels out of the donor ($I_{SET} \neq 0$) shortly after the start of the read-phase, and is replaced by one in the low energy state ($I_{SET} = 0$) thereafter. For a single spin in the presence of a large magnetic field $B$, the pair of states involved in the process is $|\downarrow\rangle, |\uparrow\rangle$ and the length of the readout “tail” is proportional to their Zeeman splitting $E_Z = \mu_T - \mu_S = g \mu_B B [25]$, where $g \approx 2$ is the Landé $g$-factor and $\mu_B$ is the Bohr magneton. However, the data in Figure 2 was taken at $B = 0$. Therefore we must attribute the observed energy splitting to a different physical mechanism, which acts in the absence of magnetic field.

We postulate that the measurement in Figure 2a constitutes the observation of the $|S\rangle$ and $|T\rangle$ states of a pair of $^{31}$P donors, split by an exchange interaction $J = \mu_T - \mu_S$, where $\mu_T$ and $\mu_S$ are the $|T\rangle$ and $|S\rangle$ electrochemical potentials at $B = 0$. To extract the value of $J$ we first convert $V_{DG}$ to a shift in $\mu$, by fitting a Fermi distribution function to the shape of $I_{SET}(V_{DG})$ for $0.25 < V_{DG} < 0.35$ V in the read-phase after the decay of the “tail”, and using the value $T_{el} = 125 \pm 25$ mK (measured separately) to calibrate the energy scale. Then, the length of the readout “tail” $\Delta V_{DG} = 0.6 \pm 0.1$ V can be converted into the value of $J = 345 \pm 100$ $\mu$eV. This value of $J$ is expected to correspond to donors $\lesssim 8$ nm apart [19, 20, 22].

We perform E-RO in the region indicated by a dashed line in Figure 2a, where $\mu_S < E_F < \mu_T$. In analogy

FIG. 2. (Color online) a. Three-phase pulse sequence and averaged SET current ($I_{SET}$), used to identify the DG voltage range where single-shot spin readout can be performed. The dashed and dash-dotted lines identify the appropriate readout-phase voltages to perform energy-selective (E-RO) and tunnel-rate selective (TR-RO) readout, respectively. b. E-RO and c. TR-RO: Diagrams of the electrochemical potentials $\mu_S, \mu_T$ relative to the SET Fermi energy $E_F$; data plots are examples of readout traces identifying each of the states. d. E-RO fidelity measurement: histogram of maximum $I_{SET}$ per shot, with optimal discrimination threshold (vertical dashed line). e. TR-RO fidelity measurement: histograms of the detection times of a pulse in $I_{SET}$ during the read-phase. The two histograms are obtained from the same data set, with different bin resolution to distinguish two separate tunneling processes. All histograms are obtained from 1000 single-shot traces, at $B = 0$, with 1 ms load time.
with the single-spin case [24, 25], the excited state \(|T\rangle\) is identified by a pulse of current at the beginning of the read-phase, while \(|S\rangle\) keeps \(I_{SET} = 0\) (see Figure 2b for sample real-time readout traces). We estimate the E-RO fidelities using the histogram of maximum current per shot on Figure 2d, with the method described in [25], from which we extract readout fidelities above 95%, with an optimal current threshold \(I_T = 0.07\) nA.

Tuning the device to the region indicated by the dash-dot line in Figure 2a, the single-shot readout traces reveal two distinct tunnel-out processes (shown in Figure 2e): A slow process with a tunnel time \(\tau_w = 0.9\) ms, and a faster process for which the tunnel time is shorter than the rise-time \(\approx 35\) \(\mu\)s of the amplifier chain (see Figure 2e for sample traces). The observation of two very distinct tunnel rates reinforces the interpretation that we are observing the spin states of a \(J\)-coupled donor pair. The \(|T\rangle\) state must correspond to an excited 2-electron orbital, with a more extended wavefunction [29] that results in stronger tunnel coupling to the nearby SET island. This tunnel rate asymmetry allows us to perform single-shot TR-RO [26] by setting a time threshold \((t_T)\) and declaring that each readout event with tunnel-out time \(t_T < t_T\) is a \(|T\rangle\) state, and a \(|S\rangle\) otherwise (Figure 2e). A statistical analysis of this measurement method reveals a TR-RO readout fidelity \(\approx 97\%\), for an optimal \(t_T = 44\) \(\mu\)s.

We measure the \(|T\rangle\) to \(|S\rangle\) relaxation time, \(T_1\), by taking repeated E-RO traces as a function of the duration \(\tau_w\) of the load phase, and calculating the probability \(P_T(\tau_w)\) of measuring a \(|T\rangle\) state (see Figure 3). By fitting the data in Figure 3b with the function \(P_T(\tau_w) = P_T(0) \exp(-\tau_w/T_1)\) we extract \(T_1 = 1.7 \pm 0.1\) ms. This value agrees well with the \(|T\rangle\rightarrow|S\rangle\) relaxation times predicted by Borhani and Hu [30] specifically for \(^{31}\)P donor pairs in Si in the presence of an exchange interaction \(J \approx 300\) \(\mu\)eV, providing yet another argument to claim that we observed a \(J\)-coupled donor pair. The electron-nuclear hyperfine coupling \(A\) (assumed \(\ll J\)) mixes the \(J\)-split \(|S\rangle,|T\rangle\) states and provides a new channel for spin-lattice relaxation which is \(\approx 3\) orders of magnitude faster than a single-spin flip at an equivalent value of the Zeeman splitting \(E_Z \approx 300\) \(\mu\)eV corresponds to \(B \approx 2.5\) T on a single spin, where \(T_1 \approx 1\) s [25]). The \(|T\rangle\rightarrow|S\rangle\) relaxation is predicted to slow down at lower \(J\), giving \(T_1 \gg 1\) s for \(J \sim 1\) \(\mu\)eV. For \(J < A = 117\) MHz \(\approx 0.5\) \(\mu\)eV this relaxation channel becomes suppressed. Therefore our measurements clearly indicate that 2-qubit coupling schemes which do not require large values of \(J\) [31, 32] will have the additional benefit of preserving the long spin lifetime of the individual qubits.

The near-unity value of \(P_T(0) = 0.91 \pm 0.03\) in Figure 3b indicates that the system is preferentially initialized in \(|T\rangle\), as expected from the faster tunnel rate into that state. By performing relaxation measurements in the TR-RO regime, we can follow in real time the evolution of the state populations after the loading of the \(|T\rangle\) state. For these measurements we consider three detection thresholds: fast tunneling events \((t < 44\) \(\mu\)s\), slow tunneling events \((44\) \(\mu\)s < \(t < 2\) ms\) and no tunneling events within the readout phase \((> 2\) ms\). An intriguing observation is that, at \(B = 0\), the sum of the \(|S\rangle\) and \(|T\rangle\) detection probabilities as a function of load time \(\tau_w\) is not constant, but exhibits a dip for \(\tau_w \sim 1 - 10\) ms (Figure 4a). This indicates that there are intermediate states with extremely low tunnel probability. We suggest that such “shelving states” are the \(|T_+\rangle\) and \(|T_+\rangle\) states, which at \(B = 0\) can be mixed with \(|T_0\rangle\) by the Overhauser field from the surrounding nuclear spin bath (in this case, the 4.7\% natural abundance of \(^{31}\)Si).

To simulate this effect in our relaxation measurements, we designed the following model of rate equations:

\[
\begin{align*}
\frac{dT_+}{d\tau_w} &= \Gamma_{Tmix}T_0 - (\Gamma_{Tmix} + \Gamma_{T\rightarrow S})T_+
\frac{dT_0}{d\tau_w} &= \Gamma_{Tmix}(T_+ + T_0) - (2\Gamma_{Tmix} + \Gamma_{T\rightarrow S})T_0
\frac{dT_-}{d\tau_w} &= \Gamma_{Tmix}T_0 - (\Gamma_{Tmix} + \Gamma_{T\rightarrow S})T_-
\frac{dS}{d\tau_w} &= \Gamma_{T\rightarrow S}(T_+ + T_0 + T_-)
\end{align*}
\]

Here \(T_0, T_-, T_+, S\) are the populations of the corresponding states, \(\tau_w\) is the wait time at the load-phase, \(\Gamma_{Tmix}\) is the rate of mixing of the \(|T\rangle\) states, and \(\Gamma_{T\rightarrow S} = 1/T_1\) is the \(|T\rangle\) to \(|S\rangle\) relaxation rate. We further assume that the tunnel rate of \(|T_-\rangle\) and \(|T_+\rangle\) is \(0\), i.e., no tunnel-out signal is obtained if the system is found in those states at the start of the read-phase. We include the parameters \(\alpha_T \equiv T_0|_{\tau_w=0} \) and \(\alpha_S \equiv S|_{\tau_w=\infty} \in [0, 1]\) that multiply the corresponding populations to account for initialization and measurement imperfections. A least-squares numerical fit to the data yields \(\Gamma_{Tmix}^{-1} = 6.7 \pm 1.8\) ms, \(\Gamma_{T\rightarrow S}^{-1} = 8.7 \pm 1.0\) ms, \(\alpha_T = 0.94 \pm 0.05\), and \(\alpha_S = 0.95 \pm 0.05\). The calculated population curves are plotted in Figure 4a, and show good agreement with the data. We attribute the difference between \(\Gamma_{T\rightarrow S}^{-1}\) extracted here and \(T_1\) from Figure 3 (both at \(B = 0\)) to a change in \(J\) caused by a different
At fields where $E_Z > J$, we use a combination of E-RO and TR-RO to map out the population evolution of $|T_0\rangle$, $|S\rangle$ and $|T_-\rangle$. Figure 4b shows a measurement at $B = 2.5$ T. As $\tau_w$ increases, the relaxation proceeds sequentially from $|T_0\rangle$ to $|S\rangle$ to $|T_-\rangle$ (we neglect the single-spin $|T_0\rangle \rightarrow |T_-\rangle$ relaxation channel, for which $\Gamma^{-1} \approx 1 \text{ s}$ at 2.5 T [25], and we assume that $|T_-\rangle$ is never loaded). In the $B \neq 0$ regime the mixing between triplets is suppressed and we can modify the rate equation model as:

$$dT_0/d\tau_w = -\Gamma T_0 \rightarrow S T_0$$
$$dS/d\tau_w = \Gamma T_0 \rightarrow S T_0 - \Gamma S \rightarrow T_- S$$
$$dT_-/d\tau_w = \Gamma S \rightarrow T_- S$$

This model fits the data in Figure 4b with $\Gamma T_0 \rightarrow S = 5.4 \pm 0.4 \text{ ms}$, $\Gamma S \rightarrow T_- = 130 \pm 70 \text{ ms}$, $\alpha T_0 = 0.69 \pm 0.29$, and $\alpha S = 0.30 \pm 0.12$. Here the rates $\Gamma S \rightarrow T_- S$ and $\Gamma S \rightarrow T_- S$ are easily resolved because they differ by over an order of magnitude, resulting in a $|S\rangle$ population that first increases ($(T_0 \rightarrow S)$) then decreases ($(S \rightarrow T_-)$).

When $B \gtrsim 4$ T, $\Gamma S \rightarrow T_- S$ becomes the fastest rate, and at $B = 5.5$ T (Figure 4d) only $\Gamma S \rightarrow T_- S = 1.16 \pm 0.06 \text{ ms}$, with $\alpha T_0 = 0.88 \pm 0.03$, can be reliably extracted from the data. Interestingly, we observe a constant population of $|S\rangle$ for $\tau_w \gtrsim 1 \text{ ms}$. This is again consistent with the spin relaxation mechanism described by the theory of Borhani and Hu [30], where the hyperfine interaction $A$ mixes states having the same total value of the electron ($m_e$) and nuclear ($m_N$) spin quantum number. The transition $|S\rangle \rightarrow |T_-\rangle$ yields $\Delta m_e = -1$, thus requires $\Delta m_N = +1$. Therefore the transition becomes forbidden if the $^{31}$P nuclei are in the state $|\psi_N\rangle = |\uparrow\uparrow\rangle$, resulting in a long-time plateau of $S$ with height depending on the probability that $|\psi_N\rangle = |\uparrow\uparrow\rangle$. $|\psi_N\rangle$ is unknown and uncontrolled in this experiment, but we may assume that the nuclei randomly populate all possible states over the
time necessary to acquire a set of data as in Figure 4. At 
$B < 2.5 \text{ T}$ this selection rule does not affect the experi-
ment, because $|S\rangle$ is the ground state and mixes with 
$|T_0\rangle$ for any $|\psi_N\rangle$.

The time-resolved observation of singlet and triplet 
states of an exchange-coupled $^{31}\text{P}$ donor pair reported 
here provides a physical basis for the construction of 
large-scale donor-based quantum computer architectures 
[11]. The short $|T\rangle \leftrightarrow |S\rangle$ relaxation times $T_1 \sim 1 \text{ ms}$ 
in this experiment arise from the interplay of a large ex-
change coupling $J \approx 300 \mu\text{eV}$ with the hyperfine inter-
action $A = 117 \text{ MHz} \approx 0.5 \mu\text{eV}$. Therefore, our results 
indicate that the best regime to operate $J$-mediated 2-
qubit logic gates is where $J \lesssim A$, as described in recent 
proposals [31, 32].

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