

Coupling a ^{73}Ge Nuclear Spin to an Electrostatically Defined Quantum Dot in Silicon

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Single nuclear spins in silicon are a promising resource for quantum technologies due to their long coherence times and excellent control fidelities. Qubits and qudits have been encoded on donor nuclei, with successful demonstrations of Bell states and quantum memories on the spin- $1/2$ ^{31}P and cat-qubits on the spin- $7/2$ ^{123}Sb nuclei. Isoelectronic nuclear spins coupled to gate-defined quantum dots, such as the naturally occurring ^{29}Si isotope, possess no additional charge and allow for the coupled electron to be shuttled without destroying the nuclear spin coherence. Here, we demonstrate the coupling of a spin- $9/2$ ^{73}Ge nuclear spin to a gate-defined quantum dot in SiMOS via Pauli spin blockade readout using rf reflectometry. We observe the hyperfine interaction (HFI) to the coupled quantum dot electron and are able to tune it from 180 to 350 kHz, through the voltages applied to the lateral gate electrodes. This smaller HFI combined with the faster readout enable easier quantum nondemolition readout of the nuclear spin state. Thus, this work lays the foundation for future spin control experiments on the spin- $9/2$ qudit as well as more advanced experiments such as entanglement distribution between distant nuclear spins or repeated weak measurements.

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Single nuclear spins in silicon are among the most coherent qubits, exhibiting exceptionally long coherence times [1] and high 1-qubit control and 2-qubit entangling fidelities [2–4]. As the magnetic moment of a single nucleus is very small, they are usually addressed through an accompanying, hyperfine-coupled electron [5]. Group-V dopant atoms or crystal defects in silicon experience a pronounced hyperfine coupling with the nuclear spins of their host atoms (~ 100 MHz) [5,6] and surrounding lattice nuclei (~ 1 –8 MHz) [7–9]. This arises from the highly confined electron wave function in such systems, which is bound by the Coulomb potential of the dopant's or defect's electric charge. In comparison, in gate-defined quantum dots, the electron wave function is typically less confined and overlaps with a much larger number of nuclear

spins [8,10–14]. This leads to a maximum hyperfine coupling of ~ 0.4 –1 MHz in silicon metal-oxide-semiconductor (SiMOS) quantum dots, where the combination of strong confinement at the Si/SiO₂ interface and the use of nanoscale gate electrodes confines the electron wave function to ≤ 10 nm in diameter in lateral direction and a couple of nanometers in vertical direction [14,15]. When using ^{28}Si -isotopically enriched silicon host material with good electron spin coherence times and narrow electron spin resonance linewidths [1,16,17], this hyperfine coupling is sufficient for addressing individual nuclear spins [14,15].

Most quantum error correction architectures require long-distance coupling of remote qubit sites [18–24]. The most promising approach in gate-defined quantum dots is shuttling [25–31], where electrons are physically moved along the interface while maintaining quantum information. In order to use shuttling in a quantum computing architecture with nuclear spins, the electron needs to be coupled and decoupled from the nuclear spin in a coherent manner. This requires deterministic movement of the coupled electron at a timescale that is much faster than the coupling, which is particularly challenging for strongly hyperfine-coupled donor electrons [32–34]. Isoelectronic group-IV atoms in quantum dots offer an

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elegant alternative. The hyperfine interaction (HFI) is weaker than for donors and, as the electrostatic potential is defined using tuneable metallic gates, the electron can be swiftly moved away [25–31].

Advanced initialization and control of high-dimensional nuclear spin systems have already been established for the ^{123}Sb donor in silicon [35–38]. Here, the bound electron has a larger HFI on the order of $A = 100\text{--}200$ MHz [21], which enables high-fidelity mapping of the nuclear spin state to the electron spin state typically on the order of $1\text{ }\mu\text{s}$. However, readout out of the electron spin state via Elzerman typically takes >1 ms. In this work, we demonstrate the coupling of a single spin- $9/2$ ^{73}Ge nuclear spin to the electron spin of a SiMOS gate-defined quantum dot read out via Pauli spin blockade (PSB) using fast rf reflectometry ($t_{\text{int}} = 100\text{ }\mu\text{s}$). We can tune the HFI between $180\text{--}350$ kHz via the gate electrodes that change the electron wave function confinement and location, which nicely matches simulations. The smaller HFI of group-IV nuclei constrains the linewidth of the ESR signal such

that the mapping takes around $7\text{ }\mu\text{s}$. However, the critical advantage is that it increases the preservation of spin coherence during electron movement [14,39]. Furthermore, readout via PSB and the high-bandwidth reflectometry charge measurements have the potential to unlock intriguing physics. Future directions are repeated nondemolition measurements of a high-dimensional spin or distributed entanglement of nuclear spins via a shuttled electron spin intermediary. Thus, our work lays the foundation for future spin control experiments on the spin- $9/2$ qudit, pointing toward the broader potential of isoelectronic species for scalable nuclear-spin-based quantum computing hardware.

Device fabrication and operation—The quantum dots are electrostatically confined by a multilayer aluminum gate-stack [40] fabricated on top of an isotopically enriched ^{28}Si substrate with 800 ppm residual ^{29}Si [16] and the equivalent of 800 ppm implanted ^{73}Ge in the active region [see Figs. 1(a) and 1(b)]. We chose the implantation dose of 800 ppm ^{73}Ge because in previous work Ref. [14] on a

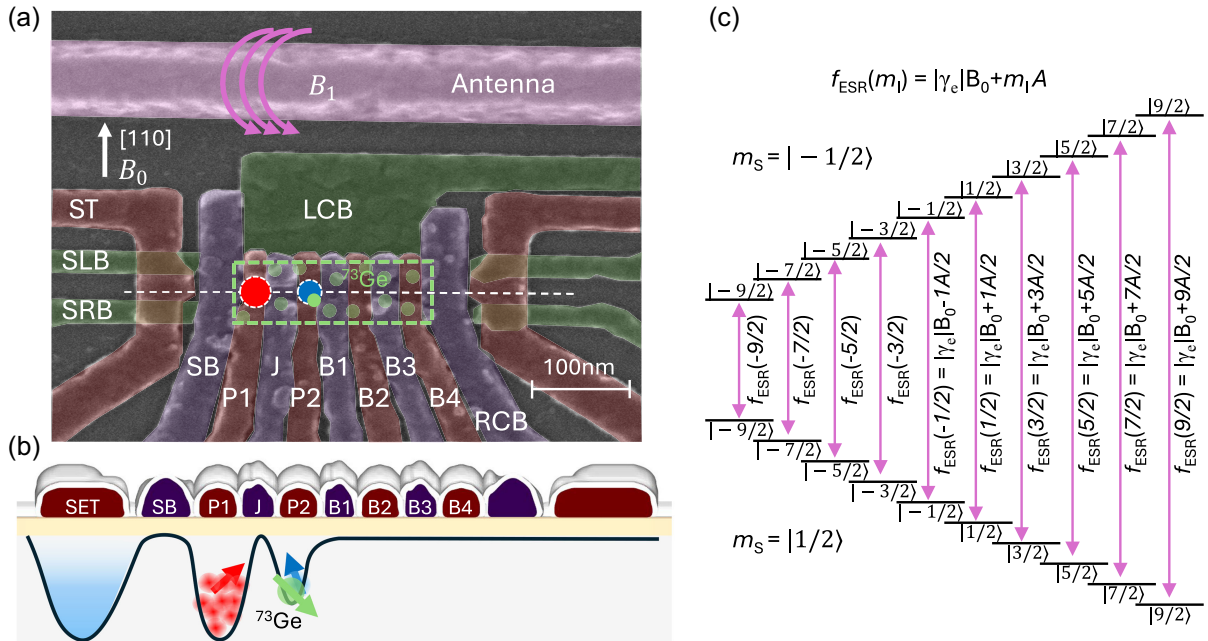


FIG. 1. Device and electron-nucleus coupled system. (a) Scanning electron micrograph of a nominally identical device. The four gate deposition layers are highlighted in green, red, purple, and pink, respectively. Arrows indicate the directions of the external applied dc magnetic field B_0 and the ac magnetic field B_1 the antenna generates. The formed quantum dots under the plunger gates P1 and P2 are indicated by the blue and red circle, respectively. The green circles indicate a possible isotopically selected ^{73}Ge nuclei distribution in the implantation window depicted by the green dashed box. The device is operated at the dilution refrigerator base temperature of $T = 50$ mK. (b) Schematic of the device's cross section in the active region, marked as the dashed white line in (a). We form the two-dimensional electron gas at the Si/SiO₂ interface. The single-electron transistor is confined with the SET left barrier (SLB), SET right barrier (SRB), and SET barrier (SB). The quantum dots to form qubits are laterally confined with the left confinement barrier (LCB) and right confinement barrier (RCB). The single-electron dot under the plunger gate P2 reveals a strongly coupled $I = 9/2$ nuclear spin. (c) Energy level diagram of an isoelectronic ^{73}Ge nucleus ($I = 9/2$) coupled to an electron spin ($S = 1/2$) of a quantum dot. The Zeeman interaction splits the states into an electron spin-up ($m_S = 1/2$) and electron spin down ($m_S = -1/2$) manifold, which is then further split by the hyperfine interaction based on the nuclear spin quantum number $m_I \in \{-9/2, -7/2, -5/2, -3/2, -1/2, 1/2, 3/2, 5/2, 7/2, 9/2\}$ and the hyperfine interaction strength A . In this coupled system, the electron spin resonance frequency f_{ESR} depends on the nuclear spin orientation. All $2I + 1 = 10$ different ESR transitions are equally spaced by A .

similar device with residual 800 ppm ^{29}Si , two $I = 1/2$ nuclear spins were hyperfine-coupled to the electron spin of a quantum dot. We biased the device to form two quantum dots separated by approximately 60 nm occupying around 80 nm^2 underneath the plunger gates (P1, P2) at the Si/SiO₂ interface. In the experiments, we operated the device in a configuration, where a double quantum dot with a charge configuration of (P1, P2) = (9, 1), was kept isolated from the charge reservoir. Under the influence of an external dc magnetic field $B_0 = 0.3 \text{ T}$, the unpaired single electron spin in each quantum dot forms an effective two-level system utilized as a qubit [41,42]. We chose the (9,1) charge configuration because nine electrons in P1 enhance the exchange coupling between the two electron spins, while a single electron in P2 maximizes the hyperfine coupling between the electron and the nuclear spin.

For single-shot charge readout, we used a radiofrequency single-electron transistor (RF-SET) [43] and integrated the signal at 205 MHz for $t_{\text{RO}} = 100 \mu\text{s}$. We delivered the ac magnetic field B_1 that drives the electron spin state transitions via an on-chip antenna.

We initialized an odd electron spin state $|\downarrow\uparrow\rangle$ with a $t_{\text{init}} = 10 \mu\text{s}$ ramp at $V_J = 3.2 \text{ V}$ across the (8,2) to (9,1) interdot charge transition. We read out the spin state parity based on Pauli spin blockade [44]. Charge movement near the interdot charge transition from (9,1) to (8,2) is blockaded when both unpaired spins are parallel.

Nuclear spin signature—We describe the joint electron-nuclear spin system under an external magnetic field B_0 along the z direction by the simplified Hamiltonian

$$H = H_{Z,e} + H_{Z,n} + H_{\text{HF}} \quad (1)$$

$$= B_0(\gamma_e S_z + \gamma_{\text{Ge}} I_z) + A(\mathbf{S} \cdot \mathbf{I}), \quad (2)$$

with the electron spin operator $\mathbf{S}$ and nuclear spin operator $\mathbf{I}$. In Eq. (2), we assume that the general hyperfine tensor $\mathbf{A}$ is dominated by the isotropic component

$$A = \frac{2}{3} \hbar^2 \mu_0 \gamma_e \gamma_{\text{Ge}} |\Psi(0)|^2, \quad (3)$$

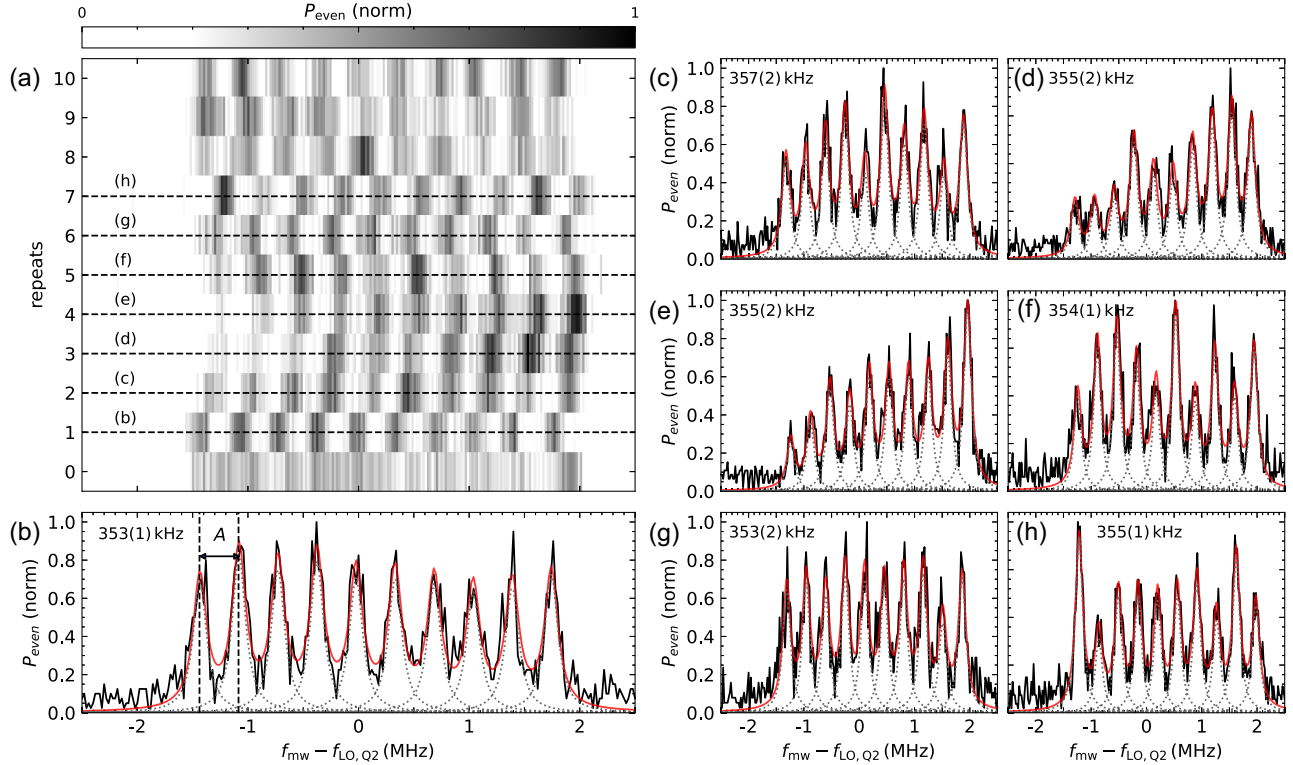


FIG. 2. Nuclear spin signature in ESR. (a) Normalized even electron spin probability P_{even} for repeated electron spin resonance probing with $t_{\text{mw}}(\pi) = 7 \mu\text{s}$ as a function of the applied microwave signal frequency f_{mw} . The local oscillator frequency of qubit 2 is $f_{\text{LO},Q2} = 8.401 \text{ GHz}$. The frequency range of 5 MHz is swept with a resolution of 251 points, while each frequency step is averaged over 500 shots. (b) Line cut of the normalized electron spin-up probability P_{even} at repetition No. 1 at the dashed black line in (a). The sum of 10 Lorentzian peaks (red line) fitted with equal spacing A . The peak spacing is the hyperfine interaction strength A between the electron and nuclear spin. The dotted lines are the individual Lorentzian peak fits. (c)–(h) Line cut of the normalized electron spin-up probability P_{even} at repetitions #2–7 at the dashed black lines in (a). Hyperfine interaction ranges from $A = 353(2)$ – $357(2) \text{ kHz}$ across all repeats. Error bars represent a 95% confidence interval determined from the estimated covariance matrix.

referred to as the Fermi-contact interaction, due to the quadratic dependence of the electron wave function overlap with the nucleus $|\Psi(0)|$. $\hbar$ is the reduced Planck's constant, μ_0 the magnetic permeability in vacuum, $\gamma_e = -28.02$ GHz/T the gyromagnetic ratio of the electron, and $\gamma_{\text{Ge}} = 1.49$ MHz/T the gyromagnetic ratio of the ^{73}Ge nuclear spin. In the experiments, we tuned the device electrically such that the HFI is maximized, which results in the anisotropic terms becoming negligible [11,15,45]. The ^{73}Ge isotope has a nuclear spin number of $I = 9/2$, which gives rise to $2I + 1 = 10$ possible nuclear spin states. The ten energy levels are separated by the nuclear Zeeman interaction and shifted by the HFI depending on the nuclear spin orientation [compare Fig. 1(c)].

In the experiments, we resonantly drove the electron spin conditional on the nuclear spin state at a frequency of $f_{\text{ESR}} = |\gamma_e|B_0 + m_1A$, with $m_1 = -9/2, -7/2, \dots, 7/2, 9/2$. Figure 2(a) shows the Larmor frequency as a function of measurement time or repeats, as we changed the applied microwave signal frequency f_{mw} . In Fig. 2(b), we fitted 10 Lorentzian peaks to repetition No. 1 in Fig. 2(a) and identified 10 peaks with equal spacing of the hyperfine constant $A = 353(1)$ kHz, in agreement with the model introduced in Eqs. (1)–(3).

Furthermore, Fig. 2(a) shows a small overall background frequency shift without a significant change of the HFI as a function of time or repeats, as evidenced by Figs. 2(c)–2(h). This drift might originate from charge noise shifting the Larmor frequency via spin-orbit interaction, an Overhauser field based on the surrounding ^{29}Si and ^{73}Ge nuclear spins, and/or a drift in the external dc magnetic field amplitude B_0 . Again, we fitted 10 Lorentzian peaks to repetitions 2–7 in Fig. 2(a) and extracted $\text{HFI} = 352(2)–357(2)$ kHz.

The hyperfine interaction is also tunable with the applied gate voltages. We increased the confinement voltage from $V_{\text{CB}} = V_{\text{RCB}} - V_{\text{LCB}} = -0.1$ V to $V_{\text{CB}} = 0.32$ V, which resulted in an estimated lateral shift of ~ 7 nm and stronger confinement of the electron wave function close to the ^{73}Ge nucleus [compare Fig. 3(a)]. Figure 3(b) shows the simulated shift of the electron density center and the thereby achieved tunability of the HFI ranging from $A = 179(1)$ kHz to the maximum value of $A = 357(2)$ kHz, roughly doubling the coupling.

Limitations in the experiment were a weakly effective J-gate likely due to a fabrication misalignment, which lead to a lack of tunnel coupling and exchange interaction tunability. This reduced the observed electron spin readout visibility and inhibited deterministic electron spin initialization, which is otherwise routinely done in SiMOS quantum dot experiments [46–48]. Thus, we were not able to initialize the spin system by electron-nuclear-double resonance (ENDOR) [33,35,36,49]. Additionally, this limited the displacement of the wave function mostly to modulation of the confinement barriers. With increased electrostatic control we expect to achieve even larger HFI.

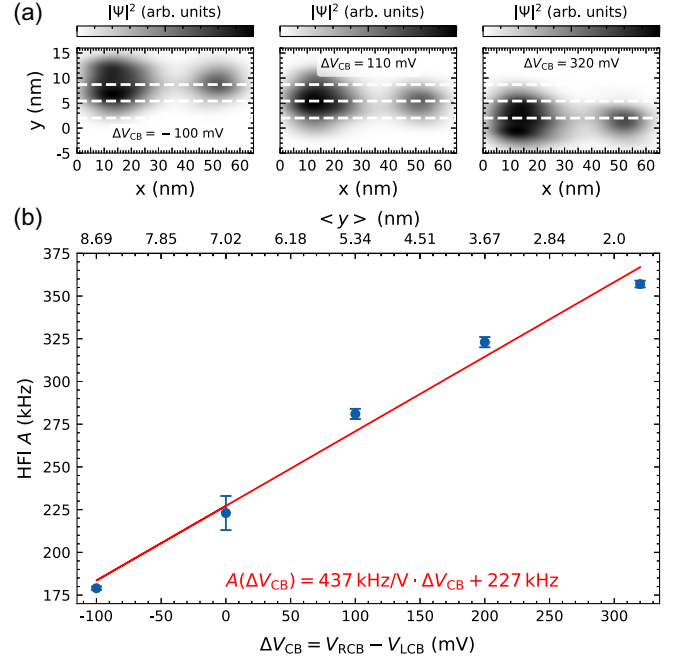


FIG. 3. Hyperfine interaction tuning. (a) Simulated electron wave function of the double-dot system for three different confinement voltages $\Delta V_{\text{CB}} = -100, 110, 320$ mV. The white dashed lines indicate the respective wave function center. (b) Hyperfine interaction strength A of the quantum-dot-coupled ^{73}Ge nuclear spin system as a function of confinement voltage V_{CB} (lower x-axis) and simulated mean of the electron density $\langle y \rangle$ (upper x-axis). The confinement voltage V_{CB} is the difference of the right and left confinement barrier voltage. Qualitatively, this corresponds to moving the center of the electron wave function closer to the ^{73}Ge nuclear spin, leading to a larger Fermi-contact hyperfine interaction component. The red line is a linear fit with slope $m = 473$ kHz/V and vertical intercept of $A = 227$ kHz. Error bars represent a 95% confidence interval when determining the hyperfine constant from Larmor frequency time traces.

Conclusion—In conclusion, we measured the coupling of a single isoelectronic ^{73}Ge isotope nuclear spin to the electron spin of an electrostatically defined quantum dot in silicon. Using the surrounding metal gates, we changed the electrostatic environment of the electron wave function to achieve tunability of the interaction strength by a factor of two. This opens a promising alternative approach to donors as qubits in silicon, as it has the potential to significantly improve the phase coherence during spin readout. Having demonstrated access to the 10-dimensional nuclear spin Hilbert space in the ^{73}Ge isotope lays the foundation for more sophisticated experiments in the future. Deterministic nuclear spin initialization, coherent nuclear control, and readout will enable a thorough characterization of the spin properties. The improved phase coherence of the spin and readout via PSB enables repeated nondemolition measurements of a high-dimensional spin that can be used for real-time tracking of spin dynamics and the observation of quantum chaos. In combination with the ability to

coherently shuttle the electron to neighboring dots, the integration of nuclear spins into a hybrid quantum computing architecture will allow for the electron spins to be used as data qubits and nuclear spins as memory qubits. Electrons can be coherently coupled and decoupled from the nuclear spin during computation, which also allows for the creation of distributed entanglement of nuclear spins via a shuttled electron spin intermediary. Ultimately, a single ^{73}Ge physical qubit can be envisioned as a logical qubit utilizing a quantum error-correction encoding like moment angular system [50,51].

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V. M. and A. L. devised the project. W. H. L., K. W. C., and F. E. H. designed and fabricated the device. S. Q. L., J. C. M., and B. C. J. prepared and performed the isotope-selective ion implantation. P. S. and G. G. conducted the experiments with A. L.'s supervision and input from T. T., T. B., R. Y. S., A. S., A. M., C. H. Y., C. C. E., and A. S. D., S. S., and T. B. assisted with the experimental setup. G. G. performed initial cryogenic device screening characterization with W. H. L.'s supervision. J. D. C. performed the electron wavefunction simulations. P. S., G. G., and A. L. wrote the manuscript, with input from all authors. A. S. D. is supported by Diraq Pty Ltd. T. T., W. H. L., S. S., T. B., J. D. C., F. E. H., K. W. C., C. C. E., A. S., C. H. Y., A. S. D., and A. L. declare equity interest in Diraq. Other authors declare no competing interest.

Data availability—The data that support the findings of this article are openly available [52], embargo periods may apply.

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End Matter

Experimental device—The device studied in this work was fabricated using multi-layer aluminum (Al) gate-stack silicon MOS technology on a isotopically enriched silicon-28 substrate with 800 ppm residual ^{29}Si . We implanted isoelectronic ^{73}Ge in the active dot region with an energy of 12 keV and a fluence of $4 \times 10^{13}/\text{cm}^2$, aiming for around 800 ppm at the interface. An 8 nm high-quality SiO_2 was thermally grown on the silicon substrate. Al gates were fabricated using an electron-beam lithography, thermal deposition of Al and lift-off process. Each Al electrode is electrically isolated by a layer of aluminum oxide of 4 nm formed via thermal oxidation on a hotplate at 150°C. The devices were designed with a plunger gate width of 35 nm and a gate pitch as small as 55 nm. This allows a 20 nm gap between the plunger gates for the J gates.

Measurement setup The device was measured in a Bluefors LD400 dilution refrigerator and mounted on the cold finger. An external dc magnetic field pointing in the [110] direction of the Si lattice was supplied by a superconducting split coil magnet and an American Magnetics AMI430 controller. We used Stanford SIM928 rechargeable isolated voltage sources to supply dc voltages through filtered lines with a bandwidth from 0 to 20 Hz. Dynamic voltage pulses were generated with a field-programmable gate array (FPGA). In this experiment, the Quantum Machines (QM) Operator-X (OPX) was combined with dc biases using custom linear bias combiners at room temperature. The OPX has a sampling time of 1 ns. The dynamic pulse lines in the dilution refrigerator have a bandwidth of 0 to 50 MHz, which translates to a minimum rise time of 20 ns. Microwave pulses were generated by a Keysight PSG8267D Vector Signal Generator, with

in-phase and quadrature (I/Q) and pulse modulation waveforms generated by the QM OPX. The modulated signal spans from 250 kHz to 44 GHz, but is bandwidth-limited by the cryostat line and a dc block.

The charge sensor comprises a single-island SET connected to a tank circuit for reflectometry measurement. The return signal was amplified by a Cosmic Microwave

Technology CITFL1 LNA at the 4 K stage followed by one ZX60-P103LN+ and one Mini-circuits ZFL-1000LN + LNAs at room temperature. All three amplifiers were powered by a Keysight E36312A Triple Output Programmable dc Power Supply. The Quantum Machines OPX generated the tones for the rf-SET, and digitized as well as demodulated the signals after the amplification.