# Precision tomography of a three-qubit electron-nuclear quantum processor in silicon

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Nuclear spins were among the first physical plat-1 forms to be considered for quantum information 2 processing[1, 2], because of their exceptional quan-3 tum coherence[3] and atomic-scale footprint. How-4 ever, their full potential for quantum computing 5 has not yet been realized, due to the lack of methods to link nuclear gubits within a scalable device combined with multi-qubit operations with sufficient fidelity to sustain fault-tolerant quanq tum computation. Here we demonstrate universal quantum logic operations using a pair of ion-11

implanted <sup>31</sup>P nuclei in a silicon nanoelectronic de-12 vice. A nuclear two-qubit controlled-Z gate is ob-13 tained by imparting a geometric phase to a shared 14 electron spin[4], and used to prepare entangled 15 Bell states with fidelities up to 94.2(2.7)%. The 16 quantum operations are precisely characterised us-17 ing gate set tomography (GST)[5], yielding one-18 qubit average gate fidelities up to 99.95(2)%, 19 two-qubit average gate fidelity of 99.37(11)% and 20 two-qubit preparation/measurement fidelities of 21 98.95(4)%. These three metrics indicate that nu-22 clear spins in silicon are approaching the perfor-23 mance demanded in fault-tolerant quantum pro-24 cessors [6]. We then demonstrate entanglement 25 between the two nuclei and the shared electron by 26 producing a Greenberger-Horne-Zeilinger three-27 qubit state with 92.5(1.0)% fidelity. Since electron 28 spin qubits in semiconductors can be further cou-29 pled to other electrons[7, 8, 9] or physically shut-30 tled across different locations[10, 11], these results 31

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<sup>32</sup> establish a viable route for scalable quantum in-

<sup>33</sup> formation processing using nuclear spins.

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Nuclear spins are the most coherent quantum systems 36 in the solid state [3, 12], owing to their extremely weak 37 coupling to the environment. In the context of quan-38 tum information processing, the long coherence is associ-39 ated with record single-qubit gate fidelities [13]. However, 40 the weak coupling poses a challenge for multi-qubit logic 41 operations. Using spin-carrying defects in diamond [14] 42 and silicon carbide [15], this problem can be addressed by 43 coupling multiple nuclei to a common electron spin, thus 44 creating quantum registers that can sustain small quan-45 tum logic operations and error correction [16]. Exciting 46 progress is being made on linking several such defects via 47 optical photons [17, 18]. 48

Still missing, however, is a pathway to exploit the 49 atomic-scale dimension of nuclear spin qubits to engineer 50 scalable quantum processors, where densely-packed qubits 51 are integrated and operated within a semiconductor chip 52 [19]. This requires entangling the nuclear qubits with elec-53 trons that can either be physically moved, or entangled 54 with other nearby electrons. It also requires interspersing 55 the electron-nuclear quantum processing units with spin 56 readout devices [20]. Here we show experimentally that 57 silicon - the material underpinning the whole of modern 58 digital information technology - is the natural system in 59 which to develop dense nuclear spin based quantum pro-60 cessors [1]. 61

#### 62 One electron – two nuclei quantum processor

The experiments are conducted on a system of two  $^{31}P$ 63 donor atoms, introduced in an isotopically purified <sup>28</sup>Si 64 substrate by ion implantation (see Methods). A three-65 qubit processor is formed by using an electron (e) with 66 spin S = 1/2 (basis states  $|\uparrow\rangle, |\downarrow\rangle$ ) and two nuclei (Q1, Q2) 67 with spin I = 1/2 (basis states  $|\uparrow\rangle, |\downarrow\rangle$ ). Metallic struc-68 tures on the surface of the chip provide electrostatic con-69 trol of the donors, create a single-electron transistor (SET) 70 charge sensor, and deliver microwave and radiofrequency 71 signals through a broadband antenna (Fig. 1a, Extended 72 Data Fig. 1). With this setup, we can perform single-shot 73 electron spin readout [20], and high fidelity ( $\approx 99.9\%$ ) 74 single-shot quantum nondemolition readout of the nuclear 75 spins [21], as well as nuclear magnetic resonance (NMR) 76 and electron spin resonance (ESR) [22] on all spins in-77 volved (see Methods). 78

The ESR spectra in Fig. 1c exhibit four resonances.
This means that the ESR frequency depends upon the

state of two nuclei, to which the electron is coupled by con-81 tact hyperfine interactions  $A_1 \approx 95$  MHz and  $A_2 \approx 9$  MHz. 82 We adopt labels where, for instance,  $\nu_{e|\downarrow\downarrow\downarrow}$  represents the 83 frequency at which the electron spin undergoes transitions 84 conditional on the two nuclear spin qubits being in the 85  $|Q_1Q_2\rangle = |\Downarrow\Downarrow\rangle$  state, and so on. The values of  $A_1, A_2$  can 86 be independently checked by measuring the frequencies 87  $\nu_{Q1|\downarrow}, \nu_{Q2|\downarrow}$  at which each nucleus responds while the elec-88 tron is in the  $|\downarrow\rangle$  state (Supplementary Information S1). 89 The hyperfine-coupled electron could either be the first 90 or the third electron bound to the donor cluster. Since 91 its spin relaxation time  $T_{1e}$  is three orders of magnitude 92 shorter than expected from a one-electron system (Ex-93 tended Data Fig. 3), we interpret the ESR spectrum in 94 Fig. 1c as describing the response of the third electron 95 bound to a 2P donor system. 96

An effective-mass calculation of the wavefunction of the 97 third electron in a 2P system (see Methods) reproduces the 98 observed values of  $A_1$  and  $A_2$  by assuming donors spaced 99 6.5 nm apart, and subjected to an electric field 2 mV/nm100 that pulls the electron wavefunction more strongly to-101 wards donor 1 (Fig. 1b). The <sup>31</sup>P nuclei in this 2P cluster 102 are spaced more widely than those produced by scanning 103 probe lithography [8, 23], where the sub-nanometre inter-104 donor spacing causes a strongly anisotropic hyperfine cou-105 pling, which randomizes the nuclear spin state each time 106 the electron is removed from the cluster for spin readout 107 [24]. Here, instead, the probability of flipping a nuclear 108 spin by electron ionisation is of order  $10^{-6}$  (Extended Data 109 Fig. 5), meaning that our nuclear readout is almost per-110 fectly quantum nondemolition. 111

### Nuclear two-qubit operations

We first consider the two <sup>31</sup>P nuclear spins as the 113 qubits of interest. One-qubit logic operations are triv-114 ially achieved by NMR pulses [21] (see Methods), where 115  $A_1 \neq A_2$  provides the spectral selectivity to address 116 each qubit individually (Fig. 1c). Two-qubit operations 117 are less trivial, since the nuclei are not directly coupled 118 to each other (Supplementary Information S1 and S9). 119 They are, however, hyperfine-coupled to the same elec-120 tron. This allows the implementation of a geometric two-121 qubit controlled-Z (CZ) gate [4, 16]. 122

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When a quantum two-level system is made to trace a closed trajectory on its Bloch sphere, its quantum state acquires a geometric phase equal to half the solid angle enclosed by the trajectory [25]. Fig. 1d illustrates how an electron  $2\pi$ -pulse at the frequency  $\nu_{e|\downarrow\downarrow\downarrow}$  (see Fig. 1d) constitutes a nuclear CZ 2-qubit gate. Starting from the state  $|\downarrow\rangle \otimes (|\downarrow\rangle + |\uparrow\rangle)/\sqrt{2} \equiv (|\downarrow\downarrow\rangle + |\downarrow\uparrow\rangle)/\sqrt{2}$ , the electron 129



Fig. 1 | Operation of a one-electron – two-nuclei quantum processor. a, Artist's impression of a pair of <sup>31</sup>P nuclei (red), asymmetrically coupled to the same electron (blue). The spins are controlled by oscillating magnetic fields (yellow) generated on-chip. b, Effective-mass calculation of the wavefunction  $\psi(y, z)$  of the third electron on the 2P cluster. The observed values of hyperfine coupling are well reproduced by assuming a 6.5 nm spacing between the donors. c, Experimental NMR spectrum of the <sup>31</sup>P nuclei (top) and ESR spectrum of the shared electron (bottom) at  $B_0 = 1.33$  T, along with energy level diagram (right) of the eight-dimensional Hilbert space (spacings not to scale). The spectra yield the hyperfine couplings  $A_1 \approx 95$  MHz and  $A_2 \approx 9$  MHz between the electron and the nuclear qubits Q1, Q2. d, Implementation of a geometric two-qubit CZ gate. A conditional  $\pi$  phase shift is acquired when a  $2\pi$  rotation is applied on the electron spin at frequency  $\nu_{e|\downarrow\downarrow\downarrow}$ , i.e. conditional on the nuclear spins being  $|\downarrow\downarrow\downarrow\rangle$ . This operation corresponds to the CZ gate on the nuclei when restricted to the electron  $|\downarrow\rangle$  subspace.

 $X_{2\pi}$  pulse at  $\nu_{e|\downarrow\downarrow\downarrow}$  introduces a phase factor  $e^{i\pi} = -1$ 130 to the  $|\Downarrow\Downarrow\rangle$  branch of the superposition, resulting in the 131 state  $(-|\Downarrow\Downarrow\rangle + |\Downarrow\uparrow\rangle)/\sqrt{2} \equiv |\Downarrow\rangle \otimes (-|\Downarrow\rangle + |\uparrow\rangle)/\sqrt{2}$ , i.e. a 132 rotation of Q2 by 180 degrees around the z-axis of its Bloch 133 sphere, which is the output of a CZ operation. Conversely, 134 if the initial state of Q1 were  $|\uparrow\rangle$ , the pulse at  $\nu_{e|\downarrow\downarrow\downarrow\downarrow}$  would 135 have no effect on the electron, leaving the nuclear qubits 136 unaffected. 137

A nuclear controlled-NOT (CNOT) gate is obtained by sandwiching the CZ gate between a nuclear  $-\pi/2$  and  $\pi/2$ pulse (Extended Data Fig. 6a). Applying an ESR  $X_{2\pi}$ pulse at  $\nu_{e|\Uparrow\Downarrow}$  transforms the sequence into a zero-CNOT gate, i.e. a gate that flips Q2 when Q1 is in the  $|0\rangle \equiv$  $|\Uparrow\rangle$  state (Extended Data Fig. 6b, and Supplementary Information S2).

We apply this universal gate set (Fig. 2a) to produce 145 each of the four maximally-entangled Bell states of the 146 two nuclear spins,  $|\Phi^{\pm}\rangle = (|\Downarrow\Downarrow\rangle \pm |\Uparrow\Uparrow\rangle)/\sqrt{2}$  and  $|\Psi^{\pm}\rangle =$ 147  $(|\Downarrow \uparrow \rangle \pm |\uparrow \Downarrow \rangle)/\sqrt{2}$ . We reconstruct the full density matri-148 ces of the Bell states using maximum likelihood quantum 149 state tomography [26] (Supplementary Information S3). 150 The reconstructed states (Fig. 2f) have fidelities of up to 151 94.2(2.7)%, and concurrences as high as 0.93(4), proving 152 the creation of genuine two-qubit entanglement. Here and 153 elsewhere, error bars indicate  $1\sigma$  confidence intervals. Bell 154 fidelities and concurrences are calculated without remov-155 ing state preparation and measurement (SPAM) errors 156 (Extended Data Fig. 10). 157

#### 158 Gate set tomography

<sup>159</sup> We used a customized, efficient gate set tomography <sup>160</sup> (GST) [27, 28, 5] analysis (see Methods, and Supplementary Information S4, S5, S8) to investigate the quality of 161 six logic operations on two nuclear-spin qubits:  $X_{\pi/2}$  and 162  $Y_{\pi/2}$  rotations on Q1 and Q2, an additional  $Y_{-\pi/2}$  rota-163 tion on Q2, and the entangling CZ gate. No two single-164 qubit operations are ever performed in parallel. GST 165 probes these six logic operations and reconstructs a full 166 two-qubit model for their behavior. Earlier experiments 167 on electron spins in silicon used randomized benchmark-168 ing (RB) [29, 30] to extract a single number for the aver-169 age fidelity of all logic operations. Characterising specific 170 gates required "interleaved" RB, which can suffer system-171 atic errors [31, 32]. Most importantly, RB does not reveal 172 the cause or nature of the errors. Our GST method en-173 ables measuring each gate's fidelity to high precision, dis-174 tinguishing the contributions of stochastic and coherent 175 errors, and separating local errors (on the target qubit) 176 from crosstalk errors (on, or coupling to, the undriven 177 spectator qubit). 178

GST estimates a two-qubit process matrix for each logic 179 operation  $(G_i: i = 1...6)$  using maximum likelihood es-180 timation. We represent each  $G_i$  as the composition of 181 its ideal target unitary process  $(\mathbb{G}_i)$  with an error pro-182 cess written in terms of a Lindbladian generator  $(\mathbb{L}_i)$ : 183  $G_i = e^{\mathbb{L}_i} \mathbb{G}_i$ . Each gate's error generator (EG) can be 184 written as a linear combination of independent elemen-185 tary EGs that describe distinct kinds of error [33]. Each 186 elementary EG's coefficient in  $\mathbb{L}_i$  is the rate (per gate) at 187 which that error builds up. Any Markovian error process 188 can be described using just four kinds of elementary EGs: 189 Hamiltonian (H), indexed by a single two-qubit Pauli op-190 erator, cause coherent or unitary errors (e.g.,  $H_{ZZ}$  gen-191



Fig. 2 | Tomography of nuclear Bell states. a, Each of the four Bell states has been generated using the same quantum circuit, only varying the initial spin state. **b-e**, Quantum state tomography results for (b)  $\Phi^+$ ; (c)  $\Phi^-$ ; (d)  $\Psi^+$ ; (e)  $\Psi^-$  Bell state. No corrections have been applied to compensate readout errors. Hollow, black boxes indicate the outcome of an ideal measurement for each Bell state. **f**, Table of Bell state fidelities and concurrences. The error bars are estimated using Monte Carlo bootstrap re-sampling and represent  $1\sigma$  confidence level.

erates a coherent ZZ rotation); Pauli-stochastic (S), also 192 indexed by a single Pauli, cause probabilistic Pauli errors 193 (e.g.  $S_{IX}$  causes probabilistic X errors on Q2); Pauli-194 correlation (C), and active (A), indexed by two Paulis, 195 describe more exotic errors (see Methods) that were not 196 detected in this experiment. We found that each gate's 197 behavior could be described using just 13-14 elementary 198 EGs: 3 local S errors and 3 local H errors acting on each 199 of Q1 and Q2, and 1-2 entangling H errors (discussed in 200 detail below). Extended Data Figure 8 shows those errors' 201 rates, along with the process matrices and full EGs used to 202 derive them. To get a higher-level picture of gate quality, 203 we aggregate the rates of related errors (see Methods) to 204 report total rates of stochastic and coherent errors on each 205 qubit and on the entire 2-qubit system. We present two 206 overall figures of merit in Figure 3a,c: generator infidelity 207 and total error. Generator infidelity is closely related to 208 entanglement infidelity, which accurately predicts average 209 gate performance in realistic large-scale quantum proces-210 sors and can be compared to fault-tolerance thresholds 211 (see Methods and Supplementary Information S9). Total 212 error is related to diamond norm (see Supplementary In-213 formation S9) and estimates worst-case gate performance 214 in any circuit, including structured or periodic circuits. In 215 Fig. 3c, we additionally report each gate's average gate 216 fidelity on its target to ease comparison of these results 217 with those from the literature. 218

The process matrices estimated by GST are not unique. 219 An equivalent representation of the gate set can be con-220 structed by a gauge transformation [34, 5] in which all 221 process matrices are conjugated by some invertible matrix, 222  $G_i \to M G_i M^{-1}$ . Some gate errors, such as over/under-223 rotations or errors on idle spectator gubits, are nearly un-224 affected by choice of gauge; they are *intrinsic* to that gate. 225 But other errors, such as a tilted rotation axis, can be 226 shifted from one gate to another by changing gauge. These 227 *relational* errors cannot be objectively associated with any 228 particular gate. Recognizing this, we divide coherent er-229 rors into intrinsic and relational components (Fig. 3a,c). 230 Intrinsic errors perturb a gate's eigenvalues, whereas re-231 lational errors perturb its eigenvectors. In Fig. 3a,c we 232 follow standard GST practice by choosing a gauge that 233 makes the gates as close to their targets as possible. This 234 associates relational errors with individual gates, in a way 235 that depends critically on the choice of gauge. But the 236 magnitude of a given relational error between a set of 237 gates is gauge-invariant, and Fig. 3d illustrates the to-238 tal relational error between each pair of gates. In this 239 work, we found evidence only for pairwise relational er-240



Fig. 3 | Precise tomographic characterization of 1- and 2-qubit gate quality. Process matrices for all 6 gates (e.g., the CZ gate shown in b) were estimated using gate set tomography (GST) and represented as error generators with associated rates. **a**, Each gate's total error rate (columns) can be partitioned into coherent (blue) and stochastic (orange) components, then further into components acting on Q1 (left), Q2 (right), and on both at once (wide). Coherent errors are further partitioned into intrinsic (dark) and relational (light), which were assigned to specific gates by fixing a gauge. Each gate's generator infidelity (see Supplementary Information S9) is shown, on the whole 2-qubit system (hollow pins) and on its target qubit[s] only (black pins). The CZ gate's total infidelity is only 0.79(14)%. Single-qubit gates have on-target infidelities of 0.07(3)-0.79(6)%, but display significant crosstalk errors on the spectator qubit and unexpected entangling coherent (ZZ) errors. c, Error metrics for each gate are aggregated by type (stochastic/coherent) and support (Q1/Q2/total). In addition to generator infidelity, each gate's average gate fidelity on its target qubit[s] is shown, to facilitate comparison with literature. d, A gauge-invariant representation of relational errors between gates (e.g. misalignment of rotation axes) that were assigned to individual gates in **a**,**c** by fixing a gauge. Each gate is labeled with its intrinsic coherent (H) and stochastic (S) errors, while edges between two gates show the total amplitude of relational coherent error (misalignent) between them. Large gauge-invariant relational errors between single-qubit gates confirm that the entangling coherent errors observed in **a** are not an artifact of gauge-fixing.

rors, although more complex multi-gate relational errorsare possible.

All 6 gates achieved on-target fidelities > 99%, with in-243 fidelities as low as 0.07(3)% on Q1 and 0.68(7)% on Q2. 244 However, we observed significant crosstalk on the specta-245 tor qubit during 1-qubit gates, resulting in full logic opera-246 tions (1-qubit gate and spectator idle operation in parallel) 247 with higher infidelities of 0.68(6)% - 3.5(2)%. Remarkably, 248 the CZ gate's infidelity of 0.79(14)% is almost on par with 249 the single-qubit gates – a rare scenario in multi-qubit sys-250 tems (Fig. 3a,c). 251

SPAM errors were estimated by GST as 1.05(4)% on average, and as low as 0.25(3)% for the  $|\uparrow\uparrow\uparrow\rangle$  state (Extended Data Figure 10). This is a unique feature of nuclear spin qubits, afforded by the quantum nondemolition nature of the measurement process [21] (Methods and Extended Data Fig. 5).

GST provided unambiguous evidence for a surprising re-258 lational error: weight-2 (entangling)  $H_{ZZ}$  and/or  $H_{\mathbb{G}_i[ZZ]}$ 259 coherent errors on each 1-qubit gate  $G_i$ , with amplitudes 260 from 1.8 - 5.0% (Extended Data Figure 8). These er-261 rors are consistent with an intermittent ZZ Hamiltonian 262 during the gate pulses. After ruling out a wide range of 263 possible error channels, we propose that the observed  $H_{ZZ}$ 264 error arises from the spurious accumulation of geometric 265 phase by the electron spin, caused by off-resonance leak-266 age of microwave power near the ESR frequencies (Supple-267 mentary Information S9). This observation illustrates the 268 diagnostic power of GST, which revealed an error channel 269 we had not anticipated. It also shows GST's ability to 270 unveil correlated and entangling errors, whose detection 271 and prevention is of key importance for the realization of 272 fault-tolerant quantum computers [35]. 273

#### 274 Three-qubit entanglement

The nuclear logic gates shown above would not scale 275 beyond a single, highly localized cluster of donors. How-276 ever, adding the hyperfine-coupled electron qubit yields a 277 scalable heterogeneous architecture. Electron qubits de-278 cohere faster, but admit faster control. If high-fidelity 279 entanglement between electron and nuclear qubits can be 280 created, electron qubits can enable fast coherent commu-281 nication between distant nuclei (via electron-electron en-282 tanglement, or physical shuttling) or serve as high-fidelity 283 ancilla qubits for quantum error correction. To demon-284 strate this capability, we produce the maximally entan-285 gled three-qubit Greenberger-Horne-Zeilinger (GHZ) state 286  $|\psi_{\rm GHZ}\rangle = (|\uparrow\uparrow\uparrow\rangle + |\downarrow\downarrow\downarrow\downarrow\rangle)/\sqrt{2}$  using the pulse sequence 287 shown in Fig. 4a. Starting from  $|\Downarrow\Downarrow\downarrow\rangle\rangle$ , an NMR  $Y_{\pi/2}$  pulse 288 at  $\nu_{Q2|\downarrow}$  creates a coherent superposition state of nucleus 289

2, followed by a nuclear zCNOT gate (as in Fig. 2a) to 290 produce a nuclear  $|\Phi^+\rangle$  state, and an ESR  $X_{\pi}$  pulse at 291  $\nu_{e|\downarrow\downarrow\downarrow}$  to arrive at  $|\psi_{GHZ}\rangle$ . Since the ESR frequency di-292 rectly depends on the state of both nuclei, the latter pulse 293 constitutes a natural 3-qubit Toffoli gate, making the cre-294 ation of 3-qubit entanglement particularly simple, as in 295 nitrogen-vacancy centres in diamond [36]. Executing Tof-296 foli gates on electrons in quantum dots [37] requires more 297 complex protocols, but can be simplified by a combination 298 of exchange and microwave pulses [38]. 299

Measuring the populations of the eight electron-nuclear 300 states (Supplementary Information S7) after each step 301 confirms the expected evolution from  $|\downarrow\downarrow\downarrow\downarrow\rangle$  to  $|\psi_{\rm GHZ}\rangle$ 302 (Fig. 4b). The evolution can be undone by applying the 303 sequence in reverse, yielding a return probability to  $|\Downarrow\Downarrow\downarrow\rangle$ 304 of 89.6(9)%, including SPAM errors. As in the two-qubit 305 case, measuring the populations is a useful sanity check 306 but does not prove multipartite entanglement, which re-307 quires knowing the off-diagonal terms of the density ma-308 trix  $\rho_{\rm GHZ} = |\psi_{\rm GHZ}\rangle\langle\psi_{\rm GHZ}|.$ 309

Standard tomography methods require measuring the 310 target state in different bases, obtained by rotating the 311 qubits prior to measurement. However, the superposition 312 of  $\left|\Downarrow\Downarrow\downarrow\downarrow\downarrow\rangle\right\rangle$  and  $\left|\Uparrow\uparrow\uparrow\rangle\right\rangle$  dephases at a rate dominated by the 313 electron dephasing time  $T_{2e}^* \approx 100 \ \mu s$  (Extended Data 314 Fig. 3), which is only marginally longer than the nuclear 315 spin operation time  $\approx 10-20 \,\mu s$ . Therefore, the GHZ state 316 will have significantly dephased by the time it is projected 317 onto each measurement basis. 318

We circumvent this problem by adopting a tomography 319 method that minimises the time spent in the GHZ state. 320 An extension of a method first introduced for the measure-321 ment of electron-nuclear entanglement in spin ensembles 322 [39], it is related to the parity scan commonly used in 323 trapped ions [40] and superconducting circuits [41]. We 324 repeat the reversal of the GHZ state (Fig. 4b) N = 100325 times, each time introducing phase shifts  $\theta_{1,2,3}$  to the ro-326 tation axes of the three reversal pulses, with  $\theta_1 = 3\theta_2 =$ 327  $9\theta_3 = 9N/125$ . The return probability to  $|\Downarrow\Downarrow\downarrow\downarrow\downarrow\rangle$  oscillates 328 with N; the amplitude and phase of the oscillations yield 329 the off-diagonal matrix element  $\langle \Downarrow \Downarrow \downarrow \mid \rho_{\text{GHZ}} \mid \uparrow \uparrow \uparrow \rangle = \rho_{18}$ . 330

Since the ideal  $\rho_{GHZ}$  has nonzero elements only on 331 its four corners, the populations  $\rho_{11}, \rho_{88}$  and the coher-332 ence  $\rho_{18}$  are sufficient to determine the GHZ state fidelity 333  $\mathcal{F}_{GHZ} = 92.5(1.0)\%$ . Also here, SPAM errors remain in-334 cluded in total infidelity. By comparison, an 88% GHZ 335 state fidelity has been reported in a triple quantum dot 336 after removing SPAM errors, whereas the uncorrected fi-337 delity is 45.8% [37]. This highlights the drastic effect of 338



Fig. 4 | Creation and tomography of an electron-nuclear three-qubit GHZ state. a, Starting from  $|\Downarrow\Downarrow\downarrow\downarrow\downarrow\rangle$ , the first three gates generate an entangled three-qubit GHZ state. All eight state populations are read out (b) at each circuit step (red dashed lines), and estimated without correcting for SPAM errors (Supplementary Information S7). The final three gates  $R(\theta_i)_{\phi}$  reverse the operations of the first three if the rotation angles are  $\theta_1 = \theta_2 = \theta_3 = 0$ , returning to the initial state in the absence of errors. The two gates that are conditional on Q<sub>2</sub> are composed of multiple pulses (Supplementary Information S6). c, The coherence between the GHZ components  $|\Downarrow\Downarrow\downarrow\downarrow\rangle$  and  $|\uparrow\uparrow\uparrow\uparrow\rangle$  is probed by incrementing the phases  $\theta_i$  of the reversal pulses. This induces oscillations at frequency  $f = 2\pi/(\theta_1 + \theta_2 + \theta_3)$  whose amplitude and phase correspond to the purity and phase relation between  $|\Downarrow\downarrow\downarrow\downarrow\rangle$  and  $|\uparrow\uparrow\uparrow\rangle$ . d, Density-matrix extrema of the GHZ state. The state populations of the GHZ components  $|\Downarrow\downarrow\downarrow\downarrow\rangle$  and  $|\uparrow\uparrow\uparrow\uparrow\rangle$ . d, Density-matrix the diagonal entries, while the oscillation amplitude and phase (c) provide the off-diagonal entries. From these values, the fidelity to the nearest GHZ state is estimated as 92.5(1.0)%, including SPAM.

<sup>339</sup> SPAM of multi-qubit entanglement, and the robustness <sup>340</sup> of our system against such errors. The different coher-<sup>341</sup> ence and operation timescales for electron and nuclei need <sup>342</sup> not be an obstacle for the use of such entangled states in <sup>343</sup> scaled-up architectures, because all further entangling or <sup>344</sup> shuttling operations between electrons will occur on  $\simeq 1 \ \mu s$ <sup>345</sup> time scales.

#### 346 Outlook

The demonstration of 1-qubit, 2-qubit and SPAM er-347 rors at or below the 1% level highlight the potential of 348 nuclear spins in silicon as a credible platform for fault-349 tolerant quantum computing. An often-quoted example, 350 based on surface code quantum error correction, sets a 351 fault-tolerance threshold of 0.56% for the entanglement 352 infidelity of 1- and 2-qubit gates and the SPAM errors [6]. 353 Several avenues are available to harness the high-fidelity 354 operations demonstrated here. Replacing the <sup>31</sup>P donors 355 with the higher-spin group-V analogues such as <sup>123</sup>Sb 356 (I = 7/2) or <sup>209</sup>Bi (I = 9/2) would provide access to a 357

much larger Hilbert space in which to encode quantum information. For example, a cluster of two<sup>123</sup>Sb donors<sup>359</sup> contains the equivalent of six qubits in the nuclear spins,<sup>360</sup> plus an electron qubit. An error-correcting code can be<sup>361</sup> efficiently implemented in high-spin nuclei [42], where our<sup>362</sup> method would provide a pathway for universal operations<sup>363</sup> between the logical qubits encoded in each nucleus.<sup>364</sup>

Moving to heavier group-V donors also allows the electrical control of the nuclear spins [43]. Combined with the electrical drive of the electron-nuclear 'flip-flop' transition [44], this implies the ability to control electron and nuclei by purely electrical means. In a two-donor system as shown here, the entangling CZ gate could similarly be obtained by an electrical  $2\pi$ -pulse on a flip-flop transition. 371

The electron-nuclear entanglement we have demonstrated can be harnessed to scale up beyond a pair of nuclei coupled to the same electron. Neighbouring donor electrons can be entangled via exchange interaction by performing controlled-rotation resonant gates [9] or  $\sqrt{SWAP}$ 

gates [8]. Wider distances could be afforded by physi-377 cally shuttling the electron across lithographic quantum 378 dots [45, 46], while preserving the quantum information 379 encoded in it [11]. Our methods would apply equally 380 to isoelectronic nuclear spin centres like <sup>73</sup>Ge and <sup>29</sup>Si, 381 where it has been shown that the nuclear qubit coherence 382 is preserved while shuttling the electron across neighbour-383 ing dots [10]. Furthermore, electron spins can mediate the 384 coherent interaction between nuclear spin qubits and mi-385 crowave photons [47, 48]. Recent experiments on electron 386 spin qubits in silicon report 1- and 2-qubit gate fidelities 387 above 99% [49, 50]. Therefore, the fidelity of electron qubit 388 operations will not constitute a bottleneck for the perfor-389 mance of electron-nuclear quantum processors. These ex-390 amples illustrate the significance of universal high-fidelity 391 two-qubit operations with nuclear spins in a platform like 392 silicon, which can simultaneously host nuclear and elec-393 tron spin qubits, lithographic quantum dots, and dense 394 readout and control devices [19]. 395

# 396 Methods

### <sup>397</sup> Device fabrication

The quantum processor is fabricated using methods com-398 patible with standard silicon MOS processes. We start 399 from a high quality silicon substrate (p-type (100); 10-20 400  $\Omega$ cm), on top of which a 900 nm thick epilayer of iso-401 topically enriched <sup>28</sup>Si has been grown using low-pressure 402 chemical vapour deposition (LPCVD). The residual <sup>29</sup>Si 403 concentration is 730 ppm. Heavily-doped  $n^+$  regions for 404 Ohmic contacts and lightly-doped p regions for leakage 405 prevention are defined by thermal diffusion of phosphorus 406 and boron, respectively. A 200 nm thick SiO<sub>2</sub> field oxide 407 is grown in a wet oxidation furnace. In the centre of the 408 device, an opening of 20  $\mu m \times 40 \mu m$  is etched in the field 409 oxide using HF acid. Immediately after, a 8 nm thick, 410 high quality dry SiO<sub>2</sub> gate oxide is grown in this opening. 411 In preparation for ion implantation, a 90 nm  $\times$  100 nm 412 aperture is opened in a PMMA mask using electron-beam-413 lithography (EBL). The samples are implanted with  $P^+$ 414 ions at an acceleration voltage of 10 keV per ion. Dur-415 ing implantation the samples were tilted by 8 degrees and 416 the fluence was set at  $1.4 \times 10^{12}$ /cm<sup>2</sup>. Donor activation 417 and implantation damage repair is achieved through the 418 process of a rapid thermal annealing (5 seconds at 1000 419 °C). The gate layout is patterned around the implantation 420 region in three EBL steps, each followed by aluminium 421 thermal deposition (25 nm thickness for layer 1; 50 nm for 422

layer 2; 100 nm for layer 3). Immediately after each metal deposition, the sample is exposed to a pure, low pressure (100 mTorr) oxygen atmosphere to form an Al<sub>2</sub>O<sub>3</sub> layer, which electrically insulated the overlapping metal gates. At the last step, samples are annealed in a forming gas (400 °C, 15 min, 95% N<sub>2</sub> / 5% H<sub>2</sub>) aimed at passivating the interface traps.

430

### Experimental setup

The device was wire-bonded to a gold-plated printed cir-431 cuit board and placed in a copper enclosure. The enclosure 432 was placed in a permanent magnet array [51], producing a 433 static magnetic field of 1.33 T at the device (see Extended 434 Data Fig. 1 for field orientation). The board was mounted 435 on a Bluefors BF-LD400 cryogen-free dilution refrigerator, 436 reaching a base temperature of 14 mK, while the effective 437 electron temperature was  $\approx 150$  mK. 438

DC bias voltages were applied to all gates using Stan-439 ford Research Systems (SRS) SIM928 voltage sources. A 440 room-temperature resistive combiner was used for the fast 441 donor gates (Extended Data Fig. 1) to add DC voltages 442 to AC signals produced by the LeCroy Arbstudio 1104, 443 which then passed through an 80 MHz low-pass filter; all 444 other gates passed through a 20 Hz low-pass filter. All 445 filtering takes place at the mixing chamber plate. The 446 wiring includes graphite-coated flexible coaxial cables to 447 reduce triboelectric noise [52]. 448

Microwave pulses to induce ESR transitions were ap-449 plied to an on-chip broadband antenna [53] using a Rohde 450 & Schwarz SGS100A vector microwave source combined 451 with an SGU100A upconverter. The microwave carrier 452 frequency remained fixed at 37.1004125 GHz, while the 453 output frequency was varied within a pulse sequence by 454 mixing it with a radiofrequency (RF) signal using double-455 sideband modulation, i.e. by applying RF pulses to the 456 in-phase port of the SGS100A IQ mixer (the quadrature 457 port was terminated by a 50  $\Omega$  load). The carrier fre-458 quency was chosen such that whenever one sideband tone 459 was resonant with an ESR pulse, the second sideband was 460 off-resonant with all other ESR frequencies. To suppress 461 microwave signals when not needed, 0 V was applied to 462 the in-phase port of the IQ mixer. Under these circum-463 stances, the carrier frequency is expected to be suppressed 464 by 35 dB, according to the source data sheet. The RF 465 pulses used for double-sideband modulation were gener-466 ated by one of the two channels of the Agilent 81180A 467 arbitrary waveform generator; the second channel deliv-468 ered RF pulses to the microwave antenna to drive NMR 469 transitions. The microwave signal for ESR and RF signal 470 471 for NMR were combined in a Marki Microwave DPX-1721
472 diplexer.

The SET current passed through a Femto DLPCA-200 473 transimpedance amplifier  $(10^7 \text{ V/A gain}, 50 \text{ kHz band})$ 474 width), followed by an SRS SIM910 JFET post-amplifier 475  $(10^2 \text{ V/V gain})$ , SRS SIM965 analog filter (50 kHz cut-476 off low-pass Bessel filter), and acquired via an AlazarTech 477 ATS9440 PCI digitizer card. The instruments were trig-478 gered by a SpinCore PulseBlasterESR-PRO. The measure-479 ments instruments were controlled by Python code us-480 ing the quantum measurement software packages QCoDeS 481 and SilQ. 482

## 483 System Hamiltonian

The static Hamiltonian of our combined electron-nuclei system is

$$H_{\rm s} = -\gamma_{\rm e} B_0 \hat{S}_z - \gamma_{\rm n} B_0 (\hat{I}_{1,z} + \hat{I}_{2,z}) + A_1 \vec{S} \cdot \vec{I}_1 + A_2 \vec{S} \cdot \vec{I}_2, \quad (1)$$

where  $\gamma_{\rm e} \approx -27.97 \text{ GHz T}^{-1}$  is the electron gyromagnetic ratio [54],  $\gamma_{\rm n} \approx 17.23 \text{ MHz T}^{-1}$  is the nuclear gyromagnetic ratio [55],  $\vec{S} = [\hat{S}_x, \hat{S}_y, \hat{S}_z]$  are the electron spin operators, and  $\vec{I}_i = [\hat{I}_{i,x}, \hat{I}_{i,y}, \hat{I}_{i,z}]$  are the nuclear spin operators for nucleus  $i \in 1, 2$ . The static magnetic field  $B_0 = 1.33$  T is aligned along  $\hat{z}$ , and  $A_1 \approx 95$  MHz,  $(A_2 \approx 9 \text{ MHz})$  is the hyperfine interaction strength between the electron and nucleus 1 (2).

An AC drive applied to the microwave line is used to induce transitions between nuclear spin states and between electron spin states. The drive predominantly modulates the transverse magnetic field as

$$H_{\rm rf}(t) = -\gamma_{\rm e}\vec{B}_1 \cdot \vec{S}\sin\omega t - \gamma_{\rm n}\vec{B}_1 \cdot (\hat{I}_1 + \hat{I}_2)\sin\omega t, \quad (2)$$

where  $\vec{B}_1$  is the oscillating magnetic field strength, primarily aligned along  $\hat{y}$ .

#### <sup>494</sup> Electron spin readout

An electron spin readout is realized through the spin to 495 charge conversion [56, 57]. This method utilizes a single 496 electron transistor (SET) as both a charge sensor and an 497 electron reservoir. The electron spin  $|\downarrow\rangle$  and  $|\uparrow\rangle$  states 498 are separated by the Zeeman energy, which scales linearly 499 with the external magnetic field. Thermal broadening of 500 the SET at 100 mK is much smaller than the Zeeman 501 splitting of two electron spin states. This means that, 502 at the read position, the donor electron spin down state 503 faces only occupied levels in the SET island (tunneling 504 is prohibited) and the spin up state faces only unoccupied 505

states and can freely tunnel out the SET island. This event 506 will shift the energy ladder in the SET island, bringing it 507 out of the Coulomb blockade, thus causing a burst in the 508 current. This burst will last until  $|\downarrow\rangle$  electron tunnels to 509 the donor. If the electron has been projected to the  $|\downarrow\rangle$ 510 state then no change in the SET current will be recorded, 511 as the electron cannot tunnel to the SET island. At the 512 end of each read phase the electron spin is reinitialized in 513  $|\downarrow\rangle$  for the next single shot cycle. The fidelity of single-shot 514 electron readout and  $|\downarrow\rangle$  initialisation by spin-dependent 515 tunnelling is  $\approx 80\%$  in this device. However, we further 516 increase the initialisation fidelity by letting the electron 517 thermalise to the lattice temperature for a time  $\gg T_{1e}$ 518 (Fig. 3b) before triggering further operations. 519

#### Nuclear spin readout and initialisation

520

The readout of the two nuclear spin qubits is an extension of the well-known method developed for a single donor [21], based on the excitation of the electron bound to the nuclei, conditional on a particular nuclear state, followed by electron spin readout [20]. The same method is used to initialise the nuclei in a known state. 526

In the present system, consisting of an electron cou-527 pled to two <sup>31</sup>P donors with different hyperfine couplings 528  $A_1 \gg A_2$ , we find four well-separated electron spin res-529 onance (ESR) frequencies (Fig. 1c), conditional on the 530  $|\Downarrow\Downarrow\rangle$ ,  $|\Downarrow\uparrow\rangle$ ,  $|\uparrow\uparrow\rangle\rangle$ ,  $|\uparrow\uparrow\rangle\rangle$  nuclear states. An electron in the 531  $|\downarrow\rangle$  state is initially drawn from a cold charge reservoir 532 onto the donor cluster (independently of nuclear states). 533 We then apply a microwave  $\pi$ -pulse at a particular ESR 534 frequency, for instance  $\nu_{e|\downarrow\downarrow\downarrow}$  corresponding to the  $|\downarrow\downarrow\downarrow\rangle$  nu-535 clear spin state, and then measure the electron spin. If it is 536 found in the  $|\uparrow\rangle$  state, then the nuclear spins are projected 537 to the  $|\Downarrow\Downarrow\rangle$  state. If the electron is  $|\downarrow\rangle$  (i.e. the pulse at 538  $\nu_{\text{ell}}$  failed to flip it to  $|\uparrow\rangle$ , the nuclear spins are projected 539 to the subspace orthogonal to the  $|\Downarrow\Downarrow\rangle$  state. This con-540 stitutes a nuclear spins single-shot readout, with a fidelity 541 given by the product of the electron single-shot readout 542 fidelity (typically  $\approx 80\%$ ) and the electron  $\pi$ -pulse fidelity 543  $(\gg 99\%).$ 544

This nuclear readout is a projective, approximately 545 quantum non demolition (QND) process [21]. The ideal 546 QND measurement relies on the observable  $I_z$  to com-547 mute with the Hamiltonian  $H_{\rm int}$  describing an interaction 548 between the observable and the measurement apparatus 549  $[I_z, H_{int}] = 0$  [58]. In our case the hyperfine terms  $A_1 S_z I_{z1}$ 550 and  $A_2 S_z I_{z2}$  constitute  $H_{\text{int}}$ . The observation of nuclear 551 spin quantum jumps originating from the electron mea-552 surement by spin-dependent tunnelling (ionization shock) 553

hints at a deviation from QND nature of the readout process [21]. It implies the presence of terms of the form  $A_{||}/2(S_{+}I_{-} + S_{-}I_{+})$  in the hyperfine coupling, and possibly additional anisotropic terms, which do not commute with  $I_z$ . In our experiment, the deviation from the ideal QND measurement is extremely small, of order  $10^{-6}$ , as shown in Extended Data Figure 5.

We exploit the near-perfect QND nature of the nuclear spin readout by repeating the cycle [load  $|\downarrow\rangle - \text{ESR} \pi$ pulse – electron readout] between 7 and 40 times, to substantially increase the nuclear single-shot readout fidelity. This is the fundamental reason why our average SPAM errors are  $\approx 1\%$  (Extended Data Fig. 10), and we have thus reported Bell and GHZ state fidelities without removing SPAM errors from the estimate.

### 569 ESR and NMR calibration

#### 570 Gate calibration

Both the 1-qubit NMR gates and the 2-qubit ESR gate 571 were iteratively calibrated using a combination of GST 572 573 and other tuning methods. Rabi flops were first used to obtain roughly calibrated 1-qubit NMR gates. Next, 1-574 gubit GST was repeatedly employed to identify and cor-575 rect error contributions such as over-/under-rotations and 576 detunings. Other routines such as the repeated application 577 of gates were performed in between GST measurements to 578 independently verify the improvements to 1-qubit gate fi-579 delities of GST. The calibrated NMR  $\pi/2$  pulse duration 580 of Q1 (Q2) is 12.0  $\mu$ s (25.3  $\mu$ s). The discrepancy between 581 the two durations is largely due to the hyperfine interac-582 tion enhancing the Rabi frequency of Q1 and reducing the 583 Rabi frequency of Q2, combined with line reflections and 584 filtering. 585

For the geometric 2-qubit gate based upon an electron 2 $\pi$  pulse, we found that a trivial calibration using Rabi flops already gave a near-optimal result. GST was then used for fine-tuning and for the detection of small error contributions such as a minor frequency shift. The calibrated ESR 2 $\pi$  pulse duration of the CZ gate is 1.89  $\mu$ s at an output power of 20 dBm.

#### 593 Periodic frequency recalibration

To keep the system tuned throughout the measurements, the NMR frequencies  $\nu_{Q1|\downarrow}$  and  $\nu_{Q2|\downarrow}$  and ESR frequency  $\nu_{e|\downarrow\downarrow}$  were calibrated every ten circuits. The ESR frequency was calibrated by measuring the ESR spectrum and selecting the frequency of the ESR peak. The NMR frequencies were measured by a variant of the Ramsey se-599 quence, consisting of an  $X_{\pi/2}$  and  $Y_{\pi/2}$  separated by a 600 wait time  $\tau$ . An off-resonant RF pulse was applied dur-601 ing the wait time to mitigate any frequency shift caused 602 by the absence of an RF drive. Since nuclear readout has 603 a near-unity fidelity, this measurement should result in 604 a nuclear flipping probability  $P_{\rm flip} = 0.5$  if the RF fre-605 quency  $f_{\rm RF}$  matches the average NMR frequency  $f_{\rm NMR}$ 606 throughout the measurement. Therefore, any deviation of 607  $P_{\rm flip}$  from 0.5 provides a direct estimate of the frequency 608 mismatch  $\Delta f = f_{\text{NMR}} - f_{\text{RF}} = \arcsin\left(2P_{\text{flip}} - 1\right)\right)/(2\pi\tau),$ 609 provided that  $|\Delta f/\tau| < 0.25$ . A higher  $\tau$  more accu-610 rate estimates  $\delta f$ , while a lower  $\tau$  results in the condition 611  $|\Delta f/\tau| < 0.25$  being valid for a broader range of  $\Delta f$ . The 612 NMR recalibration sequence iteratively increased the wait 613 time  $\tau = 40 \ \mu s \rightarrow 100 \ \mu s \rightarrow 160 \ \mu s$  to ensure that the con-614 dition  $|\Delta f/\tau| < 0.25$  remains satisfied while increasing the 615 accuracy at which the NMR frequency is estimated. For 616 each  $\tau$ , the NMR frequency was estimated by repeating 617 this sequence and updating the RF frequency until  $P_{\text{flip}}$ 618 fell within the range [0.4, 0.6]. 619

### Measurement overhead

Instrument setups and calibration routines add a signif-621 icant overhead to the GST measurements. An estimate 622 of this overhead can be obtained by comparing the total 623 measurement duration to the duration of a single pulse 624 sequence. The 2Q GST measurement shown in Fig. 3 was 625 acquired over 61 hours, during which 300-503 shots were 626 acquired for each of the 1593 circuits. This results in an 627 average duration of 340 ms per GST pulse sequence itera-628 tion. Compared to the average pulse sequence duration of 629 around 121 ms, this corresponds to an overhead of 185%. 630

620

# Effective mass theory simulations of the hyperfine interaction 632

To simulate the wave function of the third electron in the 2P system, the effective mass theory (EMT) model of the neutral 2P system in Ref. [59] is extended in a mean-field approach. 636

For short donor separations, the two inner electrons are tightly bound in a magnetically inactive singlet orbital. The third electron then only interacts with the inner ones to the extent that it experiences the Coulomb repulsion of their fixed charge distribution

$$V(\vec{r}) = \frac{e^2}{4\pi\epsilon_{\rm Si}} \int \frac{\rho_{\rm S}(\vec{r}')}{|\vec{r}' - \vec{r}|} d^3\vec{r}'.$$
 (3)

Here, e is the electron charge,  $\epsilon_{\rm Si}$  the dielectric constant in silicon and  $\rho_{\rm S}(\vec{r}')$  is the charge density of the tightly bound electrons found in Ref. [59]. The third electron is then effectively described by the sum of the 2P EMT Hamiltonian in an electric field [59] and the corresponding mean-field potential in Eq. (3).

Here, only 2P configurations along the [100] crystal axis with distances  $d \le 7$  nm and realistic fields  $E \le 2$  mV/nm are considered. In this regime the inter-donor exchange dominates the on-site exchange and the mean-field approach is justified.

The chosen basis is a combination of two STO-3G [59] orbitals, one variationally optimized at d=0.5 nm and the other at d=7 nm.

To compute the hyperfine interaction strength, the electron density at the nucleus is rescaled by a bunching factor of 440 [60]. The experimentally found hyperfine configuration is found for donors spaced 6.5 nm apart, and subjected to an electric field 2 mV/nm.

#### <sup>656</sup> Gate set tomography experiments

We designed a customized GST experiment for a set of 657 6 logic gates:  $X_{\pi/2}$  and  $Y_{\pi/2}$  rotations on each qubit, 658 an additional  $Y_{-\pi/2}$  rotation on Q2, and the symmetric 659 CZ gate between them. A basic 2-qubit GST experiment 660 for this gate set comprises a list of quantum circuits de-661 fined by: (1) choosing a set of 75 short "germ" circuits 662 that, when repeated, collectively amplify every error rate; 663 (2) repeating each germ several times to times to form 664 "germ power" circuits whose lengths are approximately 665  $L = 1, 2, 4, \dots L_{\text{max}}$ ; and (3) prefacing and appending each 666 germ power with each of 16 "preparation fiducial" circuits 667 and each of 11 "measurement fiducial" circuits. We used 668  $L_{\rm max} = 8$ , yielding a set of 20606 circuits (this is not 669 a simple multiplication because germ circuits with depth 670 > 1 do not appear at shorter L). We eliminated 92%671 of these circuits using two techniques from [5]. First, we 672 identified a subset of 18 germs that amplify any dominant 673 errors in each gate (if  $L_{\max}$  was very large, subdominant 674 errors would get echoed away by dominant errors). This 675 yielded a total of 50 germ powers. Second, for the L > 1676 germ powers, we identified and eliminated pairs of fidu-677 cial circuits that provided redundant information. This 678 trimmed the circuits per germ power from 176 to as few 679 as 16, and the total number of circuits from 8800 to just 680 1592. Each of those circuits was repeated 300-500 times to 681 gather statistics. We used maximum likelihood estimation 682 (MLE) implemented in the pyGSTi software [61, 62] to es-683 timate  $16 \times 16$  2-qubit process matrices  $\{G_i : i = 1 \dots 6\}$ 684

for all six operations.

### Constructing and selecting reduced models 686

Process matrices are a comprehensive, but not especially 687 transparent, representation of gate errors. So we used each 688 gate's ideal target (unitary) operation  $\mathbb{G}_i$  to construct an 689 error generator [33]  $\mathbb{L}_i = \log(G_i \mathbb{G}_i^{-1})$  that presents the 690 same information more usefully. Representing noisy gates 691 this way enables us to split each gate's total error into 692 parts that act on Q1 only, Q2 only, or both qubits together 693 - and then further into coherent and stochastic errors -694 to reveal those errors' sources and consequences. It also 695 enables the construction of simple, efficient "reduced mod-696 els" for gate errors, by identifying swaths of elementary er-697 ror generators whose rates are indistinguishable from zero. 698

Pinning the coefficients of k elementary error generators 699 to zero yields a reduced model with k fewer parameters, 700 whose likelihood  $(\mathcal{L})$  can be found by MLE. We evaluate 701 the statistical significance of error rates that were pinned 702 by seeing how much  $\mathcal{L}$  declines. If a given error's true rate 703 is zero, then pinning it to zero in the model reduces  $2\log \mathcal{L}$ , 704 on average, by 1 [63]. So when we pin k rates, we com-705 pute the "evidence ratio"  $r = 2\Delta \log \mathcal{L}/k$ , where  $\Delta \log \mathcal{L}$ 706 is the difference between the two models' likelihood [64]. 707 If r < 1, the pinned rates are strictly negligible; if r < 2, 708 then the smaller model is preferred by Akaike's informa-709 tion criterion (AIC) [65]; other criteria (e.g. the Bayesian 710 BIC) impose higher thresholds. We used a slightly higher 711 threshold and chose the smaller model whenever r < 5. 712 Using this methodology, we constructed a model that de-713 scribes the data well, in which just 83 (out of 1440) ele-714 mentary errors' rates are significantly different from zero. 715

The rates of all the un-pinned elementary errors form a 716 vector describing the noisy model. In general, un-physical 717 gauge degrees of freedom [5] will give rise to a foliation of 718 the model space into gauge manifolds on which the loglike-719 lihood is constant. In our analysis, we work in the limit 720 of small errors and gauge transformations where the space 721 is approximately linear, and identify the subspace that 722 is gauge invariant. We are able to construct a basis for 723 the gauge-invariant subspace whose elements correspond 724 to relational or intrinsic errors and have a definite type 725 (H, S, or A), allowing us to decompose the model's total 726 error as shown in Figure 3. 727

Extended Data Figure 8 presents each gate's 13-14 728 nonzero elementary error rates after projecting the error vector onto the gauge-invariant subspace (column 3), 730 along with the process matrices (column 1) and error generators (column 2) from which they are derived. Here and 732 elsewhere, error bars are  $1\sigma$  confidence intervals computed using the Hessian of the loglikelihood function.

## 735 Aggregated error rates and metrics

Our GST analysis aims to identify specific gate errors and 736 understand how these errors affect the overall performance 737 of our system. It begins with the raw output of GST 738 rates of elementary errors on gates. We aggregate these 739 error rates in different ways, yielding each gate's total er-740 ror and infidelity, and partitioning those metrics into their 741 components on Q1 or Q2 or both qubits together, in or-742 der to summarize different aspects of system performance. 743 We additionally report average gate fidelities to facilitate 744 comparison with the literature. 745

Gate errors by definition cause unintended changes in 746 the state of the system. S error generators produce 747 stochastic errors that transfer *probability* to erroneous 748 states; H generators produce coherent errors that trans-749 fer *amplitude* to erroneous states. We can interpret the 750 rate of an error generator, to first order, as the amount 751 of erroneous probability (denoted  $\epsilon$  for S generators) or 752 amplitude (denoted  $\theta$  for H generators) transferred by a 753 single use of the gate when acting on one half of a maxi-754 mally entangled state. 755

It is useful to group similar errors together and aggregate their rates. We classify and combine error generators
according to:

• Their type (H or S),

• Their support (Q1, Q2, or joint),

Whether they are intrinsic to a single gate, or relational between gates (H errors only; relational S errors were negligible).

The elementary error generators described in the main 764 text have definite type and support. For example, the 765  $H_{XI}$  generator has type H and support on Q1. Any er-766 ror generator on a given gate is intrinsic to that gate if 767 it commutes with the gate, and relational otherwise. For 768 example, if single-qubit  $X_{\pi/2}$  and  $Y_{\pi/2}$  gates produce ro-769 tations around axes that are separated by only 89° instead 770 of  $90^{\circ}$ , then either gate can be considered perfect at the 771 cost of assigning a 1° tilt error to the other gate. This 772 error can be moved between the two gates by a gauge 773 transformation M that rotates both gates by  $1^{\circ}$  around 774 the Z-axis. This error is purely relational; it cannot be 775 assigned definitively to one gate or the other, but can be 776 unambiguously observed in circuits containing both gates. 777

To divide each gate's errors into intrinsic and relational 778 components, we represent the gate's error generator as a 779 vector in a space spanned by the H and S elementary error 780 generators. Error generators that commute with the tar-781 get gate form a subspace that is invariant under gauge 782 transformations. The error generator's projection onto 783 this space is its intrinsic component. Error generators in 784 the complement of the intrinsic subspace are relational – 785 they can be changed or eliminated by gauge transforma-786 tions – and the projection of the gate's error generator 787 onto this complement is its relational component. 788

To construct aggregated error metrics, we start by ag-789 gregating H and S rates separately. They add in differ-790 ent ways, because H error rates correspond to amplitudes 791 while S error rates correspond to probabilities. Rates of 792 S generators add directly  $(\epsilon_{agg} = \sum_i \epsilon_i)$ , while rates of H generators add in quadrature  $(\theta_{agg} = (\sum_i \theta_i^2)^{1/2})$ . Com-793 794 bining H and S error rates into a single metric is trickier 795 - there is no unique way to do so because the impact of 796 coherent errors depends on how they interfere over the 797 course of a circuit. We therefore consider two quanti-798 ties: total error  $\epsilon_{tot} = \epsilon_{agg} + \theta_{agg}$  and generator infidelity 799  $\hat{\epsilon} = \epsilon_{\text{agg}} + \theta_{\text{agg}}^2$ . Total error approximates the maximal 800 rate at which gate errors could add up in any circuit, while 801 infidelity quantifies the same errors' average impact in a 802 random circuit. 803

Both of these metrics appear in Fig. 3, where in panels a, c, and d we report aggregated error rates that partition the overall error in various ways (see the discussion in S10 of the Supplement). We report a third metric, the *average gate fidelity* (AGF) on each gate's target qubit[s], in Fig. 3c and in the abstract to aid comparison with other published results. The on-target AGF provides an overall (and gauge-dependent) measure of the average performance of a gate when acting only on the target qubit(s). For a gate targeting Q1, it is defined as:

$$\bar{\epsilon}^{(Q1)} = 1 - \frac{1}{2} \int d\psi \, \langle \psi | \operatorname{tr}_{Q2} \left[ e^{\mathbb{L}} \left( |\psi\rangle \langle \psi | \otimes \mathbb{I} \right) \right] |\psi\rangle \quad (4)$$

For a two-qubit gate, the on-target AGF is simply the AGF of the two-qubit operation:

$$\bar{\epsilon} = 1 - \int d\psi \, \langle \psi | e^{\mathbb{L}}(|\psi\rangle \langle \psi|) | \psi \rangle, \tag{5}$$

In both cases,  $d\psi$  is the Haar measure (over 1-qubit states in Eq. 4 and over 2-qubit states in Eq. 5) and L is the error generator of the gate. Although AGF is provided for comparison to the literature, it is not a good predictor of performance in general circuits (see Supplemental <sup>809</sup> Information S9), and when we use the unqualified term <sup>810</sup> "fidelity", it always denotes generator fidelity,  $\hat{\epsilon}$ . Section <sup>811</sup> S9 of the Supplement includes an extensive discussion of <sup>812</sup> overall gate error metrics and their relationships.

# **Data availability**

The experimental data that support the findings of this study are available in Figshare with the identifier doi.org/10.6084/m9.figshare.c.5471706.

# <sup>817</sup> Code availability

Multivalley effective mass theory calculations, some of the 818 results of which are illustrated in Fig. 1b, were performed 819 using a fork of the code first developed in the production 820 of Ref. [60] that was extended to include multielectron in-821 teractions as reported in Ref. [59]. Requests for a license 822 for and copy of this code will be directed to points of con-823 tact at Sandia National Laboratories and the University 824 of New South Wales, through the corresponding author. 825

# **References**

- [1] Kane, B. E. A silicon-based nuclear spin quantum computer. *Nature* **393**, 133 (1998).
- [2] Vandersypen, L. M. & Chuang, I. L. NMR techniques
   for quantum control and computation. *Reviews of Modern Physics* 76, 1037 (2005).
- [3] Saeedi, K. *et al.* Room-temperature quantum bit
  storage exceeding 39 minutes using ionized donors in
  silicon-28. *Science* 342, 830 (2013).
- [4] Filidou, V. *et al.* Ultrafast entangling gates between nuclear spins using photoexcited triplet states. *Nature Physics* 8, 596–600 (2012).
- [5] Nielsen, E. *et al.* Gate set tomography. *Quantum* 5, 557 (2021).
- Fowler, A. G., Mariantoni, M., Martinis, J. M. & Cleland, A. N. Surface codes: Towards practical large-scale quantum computation. *Physical Review A* 86, 032324 (2012).
- [7] Harvey-Collard, P. et al. Coherent coupling between
  a quantum dot and a donor in silicon. Nature Communications 8, 1–6 (2017).

- [8] He, Y. *et al.* A two-qubit gate between phosphorus donor electrons in silicon. *Nature* **571**, 371–375 (2019).
- [9] Mądzik, M. T. *et al.* Conditional quantum operation of two exchange-coupled single-donor spin qubits in a MOS-compatible silicon device. *Nature Communications* 12, 181 (2021).
- [10] Hensen, B. et al. A silicon quantum-dot-coupled nuclear spin qubit. Nature Nanotechnology 15, 13–17 (2020).
- [11] Yoneda, J. et al. Coherent spin qubit transport in silicon. Nature Communications 12, 4114 (2021).
- [12] Zhong, M. et al. Optically addressable nuclear spins in a solid with a six-hour coherence time. Nature 517, 177–180 (2015).
- [13] Muhonen, J. T. et al. Quantifying the quantum gate fidelity of single-atom spin qubits in silicon by randomized benchmarking. Journal of Physics: Condensed Matter 27, 154205 (2015).
- Bradley, C. *et al.* A ten-qubit solid-state spin register with quantum memory up to one minute. *Physical* 867 *Review X* 9, 031045 (2019).
- Bourassa, A. *et al.* Entanglement and control of single nuclear spins in isotopically engineered silicon carbide. *Nature Materials* 19, 1319–1325 (2020).
- [16] Waldherr, G. et al. Quantum error correction in a solid-state hybrid spin register. Nature 506, 204 (2014).
- Bhaskar, M. K. *et al.* Experimental demonstration of memory-enhanced quantum communication. *Nature* **580**, 60–64 (2020).
- [18] Pompili, M. et al. Realization of a multinode quantum network of remote solid-state qubits. Science 379 372, 259–264 (2021).
- [19] Vandersypen, L. *et al.* Interfacing spin qubits in quantum dots and donors—hot, dense, and coherent. *npj Quantum Information* **3**, 1–10 (2017).
- [20] Morello, A. *et al.* Single-shot readout of an electron spin in silicon. *Nature* **467**, 687–691 (2010).
- [21] Pla, J. J. et al. High-fidelity readout and control of a nuclear spin qubit in silicon. Nature 496, 334–338 (2013).

- <sup>889</sup> [22] Pla, J. J. *et al.* A single-atom electron spin qubit in
   <sup>890</sup> silicon. *Nature* 489, 541–545 (2012).
- [23] Ivie, J. A. et al. The impact of stochastic incorporation on atomic-precision Si:P arrays. arXiv preprint arXiv:2105.12074 (2021).
- <sup>894</sup> [24] Hile, S. J. *et al.* Addressable electron spin resonance
  <sup>895</sup> using donors and donor molecules in silicon. *Science*<sup>896</sup> Advances 4, eaaq1459 (2018).
- <sup>897</sup> [25] Anandan, J. The geometric phase. Nature 360, 307–
   <sup>898</sup> 313 (1992).
- <sup>899</sup> [26] James, D. F. V., Kwiat, P. G., Munro, W. J. & White,
   A. G. Measurement of qubits. *Physical Review A* 64,
   <sup>901</sup> 052312 (2001).
- <sup>902</sup> [27] Dehollain, J. P. et al. Optimization of a solid-state
  <sup>903</sup> electron spin qubit using gate set tomography. New
  <sup>904</sup> Journal of Physics 18, 103018 (2016).
- <sup>905</sup> [28] Blume-Kohout, R. *et al.* Demonstration of qubit op<sup>906</sup> erations below a rigorous fault tolerance threshold
  <sup>907</sup> with gate set tomography. *Nature Communications*<sup>908</sup> 8, 14485 (2017).
- <sup>909</sup> [29] Huang, W. *et al.* Fidelity benchmarks for two-qubit
   <sup>910</sup> gates in silicon. *Nature* 569, 532–536 (2019).
- <sup>911</sup> [30] Xue, X. *et al.* Benchmarking gate fidelities in a
  <sup>912</sup> Si/SiGe two-qubit device. *Physical Review X* 9,
  <sup>913</sup> 021011 (2019).
- <sup>914</sup> [31] Kimmel, S., da Silva, M. P., Ryan, C. A., Johnson,
  <sup>915</sup> B. R. & Ohki, T. Robust extraction of tomographic <sup>916</sup> information via randomized benchmarking. *Physical* <sup>917</sup> *Review X* 4, 011050 (2014).
- <sup>918</sup> [32] Carignan-Dugas, A., Wallman, J. J. & Emerson, J.
  <sup>919</sup> Bounding the average gate fidelity of composite chan<sup>920</sup> nels using the unitarity. New Journal of Physics 21,
  <sup>921</sup> 053016 (2019).
- [33] Blume-Kohout, R. et al. A taxonomy of small markovian errors. arXiv preprint arXiv:2103.01928 (2021).
- <sup>924</sup> [34] Proctor, T., Rudinger, K., Young, K., Sarovar, M.
  <sup>925</sup> & Blume-Kohout, R. What randomized benchmark<sup>926</sup> ing actually measures. *Physical Review Letters* **119**,
  <sup>927</sup> 130502 (2017).
- [35] Novais, E. & Mucciolo, E. R. Surface code threshold
  in the presence of correlated errors. *Physical Review Letters* 110, 010502 (2013).

- [36] Neumann, P. et al. Multipartite entanglement among
   single spins in diamond. Science 320, 1326–1329
   (2008).
- [37] Takeda, K. et al. Quantum tomography of an entangled three-qubit state in silicon. Nature Nanotechnology 16, 965–969 (2021).
- [38] Gullans, M. & Petta, J. Protocol for a resonantly driven three-qubit toffoli gate with silicon spin qubits. *Physical Review B* 100, 085419 (2019).
- [39] Mehring, M., Mende, J. & Scherer, W. Entanglement
   between an electron and a nuclear spin 1/2. *Physical Review Letters* 90, 153001 (2003).
- [40] Sackett, C. A. *et al.* Experimental entanglement of four particles. *Nature* **404**, 256–259 (2000).
- [41] Wei, K. X. et al. Verifying multipartite entangled greenberger-horne-zeilinger states via multiple quantum coherences. *Physical Review A* 101, 032343 947 (2020).
- [42] Gross, J. A., Godfrin, C., Blais, A. & Dupont-Ferrier, E. Hardware-efficient error-correcting codes for large nuclear spins. arXiv preprint arXiv:2103.08548 951 (2021). 952
- [43] Asaad, S. et al. Coherent electrical control of a single high-spin nucleus in silicon. Nature 579, 205–209 (2020).
- [44] Tosi, G. et al. Silicon quantum processor with robust long-distance qubit couplings. Nature Communications 8, 450 (2017).
- [45] Pica, G., Lovett, B. W., Bhatt, R. N., Schenkel, T. & 959
   Lyon, S. A. Surface code architecture for donors and dots in silicon with imprecise and nonuniform qubit couplings. *Physical Review B* 93, 035306 (2016). 962
- [46] Buonacorsi, B. et al. Network architecture for a topological quantum computer in silicon. Quantum Science and Technology 4, 025003 (2019).
- [47] Tosi, G., Mohiyaddin, F. A., Tenberg, S., Laucht,
  A. & Morello, A. Robust electric dipole transition at microwave frequencies for nuclear spin qubits in silicon. *Physical Review B* 98, 075313 (2018).
- [48] Mielke, J., Petta, J. R. & Burkard, G. Nuclear
   spin readout in a cavity-coupled hybrid quantum dotdonor system. *PRX Quantum* 2, 020347 (2021).

- 973 [49] Xue, X. *et al.* Computing with spin qubits at 974 the surface code error threshold. *arXiv preprint* 975 *arXiv:2107.00628* (2021).
- <sup>976</sup> [50] Noiri, A. *et al.* Fast universal quantum control above
  <sup>977</sup> the fault-tolerance threshold in silicon. *arXiv preprint*<sup>978</sup> *arXiv:2108.02626* (2021).
- 979 [51] Adambukulam, C. et al. An ultra-stable 1.5
  980 tesla permanent magnet assembly for qubit exper981 iments at cryogenic temperatures. arXiv preprint
  982 arXiv:2010.02455 (2020).
- <sup>983</sup> [52] Kalra, R. *et al.* Vibration-induced electrical noise in a cryogen-free dilution refrigerator: Characterization, mitigation, and impact on qubit coherence. *Review* of Scientific Instruments 87, 073905 (2016).
- <sup>987</sup> [53] Dehollain, J. *et al.* Nanoscale broadband transmission lines for spin qubit control. *Nanotechnology* 24, 015202 (2012).
- <sup>990</sup> [54] Feher, G. Electron spin resonance experiments on
  <sup>991</sup> donors in silicon. i. electronic structure of donors
  <sup>992</sup> by the electron nuclear double resonance technique.
  <sup>993</sup> Physical Review 114, 1219 (1959).
- [55] Steger, M. et al. Optically-detected NMR of optically hyperpolarized <sup>31</sup>P neutral donors in <sup>28</sup>Si. Journal of
   Applied Physics **109**, 102411 (2011).
- <sup>997</sup> [56] Elzerman, J. M. *et al.* Single-shot read-out of an
  <sup>998</sup> individual electron spin in a quantum dot. *Nature*<sup>999</sup> **430**, 431 (2004).
- <sup>1000</sup> [57] Morello, A. *et al.* Architecture for high-sensitivity single-shot readout and control of the electron spin of individual donors in silicon. *Physical Review B* 80, 081307 (2009).
- <sup>1004</sup> [58] Braginsky, V. B. & Khalili, F. Y. Quantum nonde <sup>1005</sup> molition measurements: the route from toys to tools.
   <sup>1006</sup> Reviews of Modern Physics 68, 1 (1996).
- [59] Joecker, B. *et al.* Full configuration interaction simulations of exchange-coupled donors in silicon using
  multi-valley effective mass theory. *New Journal of Physics* (2021).
- [60] Gamble, J. K. *et al.* Multivalley effective mass theory
  simulation of donors in silicon. *Physical Review B* **91**, 235318 (2015).

- [61] Nielsen, E. et al. Python GST implementation 1014 (PyGSTi) v. 0.9. Tech. Rep., Sandia National 1015 Lab.(SNL-NM), Albuquerque, NM (United States) 1016 (2019).
- [62] Nielsen, E. et al. Probing quantum processor performance with pyGSTi. Quantum Science and Technology 5, 044002 (2020).
- [63] Wilks, S. S. The large-sample distribution of the likelihood ratio for testing composite hypotheses. The 1022 Annals of Mathematical Statistics **9**, 60 – 62 (1938). 1023
- [64] Nielsen, E., Rudinger, K., Proctor, T., Young, K. 1024
   & Blume-Kohout, R. Efficient flexible characterization of quantum processors with nested error models. 1026
   arXiv preprint arXiv:2103.02188 (2021). 1027
- [65] Akaike, H. Information theory and an extension of the maximum likelihood principle. In *Selected papers* 1029 of *Hirotugu Akaike*, 199–213 (Springer, 1998).
- [66] Tenberg, S. B. *et al.* Electron spin relaxation of single phosphorus donors in metal-oxide-semiconductor nanoscale devices. *Physical Review B* **99**, 205306 (2019).
- [67] Hsueh, Y.-L. et al. Spin-lattice relaxation times of 1035 single donors and donor clusters in silicon. Physical 1036 review letters 113, 246406 (2014).

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# **Author contributions**

M.T.M., V.S. and F.E.H. fabricated the device, with 1066 A.M.'s and A.S.D.'s supervision, on an isotopically-1067 enriched <sup>28</sup>Si wafer supplied by K.M.I. A.M.J., B.C.J. 1068 and D.N.J. designed and performed the ion implantation. 1069 M.T.M. and S.A. performed the experiments and anal-1070 ysed the data, with A.L. and A.M.'s supervision. B.J. 1071 and A.D.B. developed and applied computational tools 1072 to calculate the electron wavefunction and the Hamilto-1073 nian evolution. A.Y. designed the initial GST sequences, 1074 with C.F.'s supervision. K.M.R., E.N., K.C.Y., T.J.P. 1075 and R.B.-K. developed the and applied the GST method. 1076 A.M., R.B-K., M.T.M. and S.A. wrote the manuscript, 1077 with input from all coauthors. 1078

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1084 **Competing interest** The authors declare no compet-1085 ing interests.

# Extended data figures and tables



**Extended Data Fig. 1** | **Device layout.** Scanning electron micrograph of a device identical to the one used in this experiment. <sup>31</sup>P donor atoms are implanted in the region marked by the orange rectangle, using a fluence of  $1.4 \times 10^{12}$ /cm<sup>2</sup> which results in a most probably inter-donor spacing of approximately 8 nm. Four metallic gates are fabricated around the implantation region, and used to modify the electrochemical potential of the donors. A nearby SET, formed using the SET top gate and barrier gates, enables charge sensing of a single donor atom, as well as its electron spin through spin-to-charge conversion (Methods). The tunnel coupling between the donors and SET is tuned by the rate gate situated between the SET and donor implant region. A nearby microwave (MW) antenna is used for ESR and NMR of the donor electron and nuclear spins, respectively.



Extended Data Fig. 2 | Electrical tunability of the hyperfine interaction and the electron gyromagnetic ratio. a, Map of the SET current as a function of SET gate and fast donor gates (pulsed jointly). The white dashed line indicates the location in gate space where the 2P donor cluster changes its charge state. The third, hyperfinecoupled electron is present on the cluster in the region to the right of the line. Electron spin readout is performed at the location indicated by the pink star. b, ESR spectrum of the electron bound to the 2P cluster, acquired while the system was tuned within the blue dashed rectangle in panel **a**. The hyperfine couplings  $A_1, A_2$  are extracted from ESR frequencies as shown, namely  $A_1 = (\nu_{e|\uparrow\downarrow\downarrow} + \nu_{e|\uparrow\uparrow\uparrow})/2 - (\nu_{e|\downarrow\downarrow\downarrow} + \nu_{e|\downarrow\uparrow\uparrow})/2$ ;  $A_2 = \nu_{e|\uparrow\uparrow\uparrow} - \nu_{e|\uparrow\downarrow\downarrow}$ . **c-d**, Extracted hyperfine couplings within the marked area. The data shows that  $A_1$  decreases and  $A_2$  increases upon moving the operation point towards higher gate voltages and away from the donor readout position. **e**, A small change is also observed in the sum of the two hyperfine interactions  $A_t = A_1 + A_2$ . **f**, Electrical modulation (Stark shift) of the electron gyromagnetic ratio  $\gamma_e$ , extracted from the shift of the average of the hyperfine-split electron resonances. The ESR frequencies can be tuned with fast donor gates at the rate of  $\Delta \nu_{e|\uparrow\uparrow\uparrow} = 0.3$  MHz/V;  $\Delta \nu_{e|\uparrow\downarrow\downarrow} = 5.2$  MHz/V;  $\Delta \nu_{e|\downarrow\downarrow\uparrow} = 7.6$  MHz/V;  $\Delta \nu_{e|\downarrow\downarrow\downarrow} = 2.4$  MHz/V.



Extended Data Fig. 3 | Coherence metrics of the electron spin qubit. The columns correspond to the nuclear configurations  $|\downarrow\downarrow\downarrow\rangle$ ,  $|\downarrow\uparrow\uparrow\rangle$ ,  $|\uparrow\uparrow\downarrow\rangle$ ,  $|\uparrow\uparrow\downarrow\rangle$ , respectively. All measurements start with the electron spin initialized in the  $|\downarrow\rangle$  state. Error bars are 1 $\sigma$  confidence intervals. **a**, Electron Rabi oscillations. The measurements were performed by applying a resonant ESR pulse of increasing duration. The different Rabi frequencies  $f_{\text{Rabi}}$  on each resonance are likely due to a frequency-dependent response of the on-chip antenna and the cable connected to it. b, Electron spin-lattice relaxation times  $T_{1e}$ . Measurements were obtained by first adiabatically inverting the electron spin to  $|\uparrow\rangle$ , followed by a varying wait time au before electron readout. The observed relaxation times are nearly three orders of magnitude shorter than typically observed in single-electron, single-donor devices [66], and even shorter compared to 1e-2P clusters. This strongly suggests that the measured electron is the third one, on top of two more tightly-bound electrons which form a singlet spin state [67]. We also observe a strong dependence of  $T_{1e}$  on nuclear spin configuration. c, Electron dephasing times  $T_{2e}^*$ . The measurements were conducted by performing a Ramsey experiment, i.e. by applying two  $\pi/2$  pulses separated by a varying wait time  $\tau$ , followed by electron readout. The Ramsey fringes are fitted to a function of the form  $P_{\uparrow}(\tau) = C_0 + C_1 \cos(\Delta \omega \cdot \tau + \Delta \phi) \exp[-(\tau/T_{2e}^*)^2]$ , where  $\Delta \omega$  is the frequency detuning and  $\Delta \phi$  is a phase offset. The observed  $T_{2e}^*$  times are comparable to previous values for electrons coupled to a single <sup>31</sup>P nucleus. **d**, Electron Hahn-echo coherence times  $T_{2e}^{\rm H}$ , obtained by adding a  $\pi$  refocusing pulse to the Ramsey sequence. We also varied the phase of the final  $\pi/2$  pulse at a rate of one period per  $\tau = (5 \text{ kHz})^{-1}$ , to introduce oscillations in the spin-up fraction which help improve the fitting. The curves are fitted to the same function used to fit the Ramsey fringes, with fixed  $\Delta \omega = 5$  kHz. The measured  $T_{2e}^{\rm H}$  times are similar to previous observations for electrons coupled to a single <sup>31</sup>P nucleus.



Extended Data Fig. 4 | Nuclear spin coherence times. Panels in column 1 (2) correspond to nucleus Q1 (Q2). Error bars are  $1\sigma$  confidence intervals. **a**, Nuclear dephasing times  $T_{2n}^*$ , obtained from a Ramsey experiment. Results are fitted with a decaying sinusoid with fixed exponent factor 2 (see Extended Data Fig. 3). **b**, Nuclear Hahn-echo coherence times  $T_{2n}^{\rm H}$ . To improve fitting, oscillations are induced by incrementing the phase of the final  $\pi/2$  pulse with  $\tau$  at a rate of one period per (3.5 kHz)<sup>-1</sup>. Results are fitted with a decaying sinusoid with fixed exponent factor 2 (see Extended Data Fig. 3). **c**, Dependence of  $T_{2n}^{\rm H}$  on the amplitude of an off-resonance pulse. We perform this experiment to study whether a qubit, nominally left idle (or, in quantum information terms, subjected to an identity gate) is affected by the application of an RF pulse to the other qubit, at a vastly different frequency. Here, during the idle times between NMR pulses, an RF pulse is applied at a fixed frequency 20 MHz – far off-resonance from both qubits' transitions – with varying amplitude  $V_{\rm RF}$ . The red dashed line indicates the applied RF amplitude for NMR pulses throughout the experiment. We observe a slow decrease of  $T_{2n}^{\rm H}$  with increasing  $V_{\rm RF}$ . This is qualitatively consistent with the observation of large stochastic errors on the idle qubit, as extracted by the GST analysis in Fig. 3.



Extended Data Fig. 5 | Nuclear spin quantum jumps caused by ionization shock. The electron and nuclear spin readout relies upon spin-dependent charge tunnelling between the donors and the SET island. If the electron tunnels out of the two-donor system, the hyperfine interactions  $A_1, A_2$  suddenly drop to zero. If  $A_1$  and  $A_2$  include an anisotropic component (e.g. due to the non-spherical shape of the electron wavefunction which results in nonzero dipolar fields at the nuclei), the ionisation is accompanied by a sudden change in the nuclear spin quantisation axes ("ionisation shock"), and can result in a flip of the nuclear spin state. We measure the nuclear spin flips caused by ionisation shock by forcibly loading and unloading an electron from the 2P cluster every 0.8 ms. **a**, For qubit 1 with  $A_1 = 95$  MHz, the flip rate is  $\Gamma_1 = 2.8 \times 10^{-6} \frac{\text{N}_{\text{flip}}}{\text{N}_{\text{ion}}}$ . **b**, For qubit 2 with  $A_2 = 9$  MHz, the flip rate is  $\Gamma_2 = 4.0 \times 10^{-7} \frac{\text{N}_{\text{flip}}}{\text{N}_{\text{ion}}}$ . This means that the nuclear spin readout via the electron ancilla is almost exactly quantum non-demolition. From this data, we also extract an average time between random nuclear spin flips of 283 seconds for qubit 1, and 2000 seconds for qubit 2. The extremely low values of  $\Gamma$  – comparable to those observed in single-donor systems – are the reason why we can reliably operate the two  $^{31}$ P nuclei as high-fidelity qubits.



Extended Data Fig. 6 | CNOT and zero-CNOT nuclear two-qubit gates. We perform Rabi oscillation on the control qubit followed by the application of **a**, zCNOT or **b**, CNOT gates. The two qubits are initialized in the  $|\Downarrow\Downarrow\rangle \equiv |11\rangle$  state. We observe the Rabi oscillations of both qubits in phase for zCNOT and out of phase for CNOT. At every odd multiple of  $\pi/2$  rotation of the control qubit the Bell states are created.



**Extended Data Fig. 7** | **Two-qubit gate set tomography. a**, Measurement circuit for the two-qubit gate set tomography. A modified version of this circuit has been used for Bell state tomography. The green box prepares the qubit 2 in the  $|\uparrow\rangle$  state, then the orange box prepares the qubit 1 in the  $|\uparrow\rangle$  state. The readout step in the blue box (see Methods) determines whether the  $|\uparrow\uparrow\rangle$  state initialization was successful. Only then the record will be saved. The electron spin is prepared in  $|\downarrow\rangle$  during the nuclear spin readout process. Subsequently, the GST sequence is executed. The red box indicates the Q1,Q2 readout step. The total duration of the pulse sequence is 120 ms, of which nuclear spin initialization is 8.6 ms (green and yellow), initial nuclear spin readout is 26.5 ms (blue), 3 ms delay is added for electron initialization (between blue and purple), GST circuit is 10  $\mu$ s - 300  $\mu$ s (purple), and nuclear readout is 80 ms (orange). **b**, Measurement results for individual two-qubit gate set tomography circuit. The first 145 circuits estimate the preparation and measurement fiducials, and the subsequent circuits are ordered by increasing circuit depth. At the end of a circuit, there are three situations for the target state populations: 1) the population is entirely in one state, while all others are zero; 2) the population is equally spread over two states, while the other two are zero; 3) the population is equally spread over all four states. The measured state populations for the different circuits therefore congregate around the four bands 0, 0.25, 0.5, and 1, as indicated by black dashed lines.



Extended Data Fig. 8 | Estimated gate set, from process matrices to error rates. Experimental GST data were analyzed using pyGSTi to obtain self-consistent maximum likelihood estimates of 2-qubit process matrices for all 6 elementary gates. These are represented ("Process Matrix" column) in a gauge that minimizes their average total error, as superoperators in the 2-qubit Pauli basis. Green columns indicate positive matrix elements, orange ones are negative. Wireframe sections indicate differences between estimated and ideal (target) process matrices. Those process matrices can be transformed to error generators ("Error Generator" column) that isolate those differences, and are zero if the estimated gate equals its target. Each gate's error generator was decomposed into a sparse sum of Hamiltonian and stochastic elementary error generators [33]. Those rates are depicted ("All Error Rates" column) as contributions to the gate's total error, with one-sigma uncertainties indicated in parentheses. Each non-vanishing elementary error rate (error generators are denoted "H" or "S" followed by a Pauli operator) is listed, and identified with its role in the total error budget (reproduced from Figure 3). Orange bars indicate stochastic errors, dark blue indicate coherent errors that are intrinsic to the gate, and light blue indicate relational coherent errors that were assigned to this gate. Total height of the blue region indicates the total coherent error, but because coherent error amplitudes add in quadrature, individual components' heights are proportional to their quadrature.



Extended Data Fig. 9 | Simulation of standard and interleaved randomized benchmarking (RB). All simulated RB experiments used 2-qubit Clifford subroutines compiled from the 6 native gates, requiring (on average) 14.58 individual gate operations per 2-qubit Clifford. **a**, Standard randomized benchmarking, simulated using the GST-estimated gate set, yields a "reference" decay rate of  $r_r = 22.2(2)\%$ , suggesting an average per-gate error rate of  $r_r/14.58 \approx 1.5\%$ . One-sigma confidence intervals are indicated in parentheses. **b-f**, Simulated interleaved randomized benchmarking for the CZ gate, and 1-qubit  $X_{\pi/2}$  and  $Y_{\pi/2}$  gates on each qubit, yielded interleaved decay rates  $r_r + r_i$ . For each experiment, 1000 random Clifford sequences were generated, at each of 15 circuit depths m, and simulated using the GST process matrices. Exact probabilities (effectively infinitely many shots of each sequence) were recorded. Inset histograms show the distribution over 1000 random circuits at m=4. Observed decays are consistent with each gate's GST-estimated infidelities – e.g. 1 - F = 0.79% for the C-Z gate (b). Performing these exact RB experiments in the lab would have required running 90000 circuits to estimate a single parameter ( $r_i$ ) for each gate to the given precision of  $\pm 0.25\%$ . Using fewer (< 1000) random circuits at each m would yield lower precision. GST required only 1500 circuits to estimate *all* error rates to the same precision.

	介介>	$ \uparrow\downarrow\rangle$	↓↑	$ \Downarrow\Downarrow\rangle$
$\Pr( \uparrow\uparrow\rangle)$	99.75(3)%	0.53(7)%	0.53(6)%	0.52(4)%
$\Pr( \Uparrow\Downarrow\rangle)$	0.04(1)%	99.09(8)%	0.02(1)%	0.06(2)%
$\Pr( \Downarrow \uparrow \rangle)$	0.20(3)%	0.18(3)%	97.73(10)%	0.20(5)%
$\Pr( \Downarrow\Downarrow\rangle)$	0.02(1)%	0.20(3)%	1.72(8)%	99.22(6)%

Extended Data Fig. 10 | Estimated state preparation and measurement (SPAM) error rates. In the GST analysis, the system's initial state was represented by a 4 × 4 density matrix  $\rho$ , and the final measurement/readout by a 4-element 4 × 4 POVM (positive operator-valued measure)  $\{E_{\uparrow\uparrow\uparrow}, E_{\uparrow\downarrow\downarrow}, E_{\downarrow\downarrow\uparrow}, E_{\downarrow\downarrow\downarrow}\}$  with  $E_j \ge 0$  and  $\sum_j E_j = I$ . We quantified the overall quality of the SPAM operations by using the GST estimate to compute the table of conditional probabilities shown here. Each cell shows the estimated probability of a particular readout (e.g.  $\uparrow\uparrow\uparrow$ ) given (imperfect) initialization in a particular state (e.g.  $|\downarrow\downarrow\downarrow\rangle$ ). The  $|\uparrow\uparrow\uparrow\rangle$  column can be read out directly from the estimate, since the experiment initialized into  $|\uparrow\uparrow\uparrow\rangle$ . Other states must be prepared by applying  $X_{\pi/2}$  or  $Y_{\pi/2}$  pulses. These add additional error, which should not be attributed to SPAM operations. To correct for this, we simulated ideal unitary rotation of the real  $|\uparrow\uparrow\uparrow\rangle$  state into each of the other 3 states by (1) taking the GST-estimated  $X_{\pi/2}$  gates on each qubit and removing all intrinsic errors from them, and (2) simulating a circuit comprising initialization in  $\rho$ , an appropriate sequence of those idealized gates, and readout according to  $\{E_j\}$ . The resulting analysis shows probabilities of all but one readout error to be below 1%, which is unprecedented in semiconductor spin qubit systems, and competitive with the state of the art in other physical platforms.