Engineering long spin coherence times of spin–orbit qubits in silicon

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Electron-spin qubits have long coherence times suitable for quantum technologies. Spin–orbit coupling promises to greatly improve spin qubit scalability and functionality, allowing qubit coupling via photons, phonons or mutual capacitances, and enabling the realization of engineered hybrid and topological quantum systems. However, despite much recent interest, results to date have yielded short coherence times (from 0.1 to 1 μs). Here we demonstrate ultra-long coherence times of 10 ms for holes where spin–orbit coupling yields quantized total angular momentum. We focus on holes bound to boron acceptors in bulk silicon 28, whose wavefunction symmetry can be controlled through crystal strain, allowing direct control over the longitudinal electric dipole that causes decoherence. The results rival the best electron-spin qubits and are 10^4 to 10^6 longer than previous spin–orbit qubits. These results open a pathway to develop new artificial quantum systems and to improve the functionality and scalability of spin-based quantum technologies.

Spin–orbit coupling is an attractive ingredient in spin-based quantum information technologies1–4 because it allows long-range coupling of spin qubits via photon, phonon and mutual capacitance5–7 and would lead to engineering hybrid and topological quantum systems8–10. As such, spin–orbit qubits have attracted considerable attention11–16. Recently, hole spin qubits with spin–orbit coupling have been demonstrated in a silicon metal-oxide-semiconductor platform14 and in germanium15, but with relatively short coherence times (T2) reminiscent of previous studies11–13. This rapid progress further motivates the fundamental question of whether the T2 of the spin–orbit qubits can be engineered to rival spin qubits with weak spin–orbit coupling. Strong spin–orbit interaction mixes spin and orbital degrees of freedom, making it highly non-trivial to protect the coherence of spin–orbit qubits against charge noise. Here, we show that by engineering the quadrupole degree of freedom associated with the linear response of generalized spins to environmental electric fields using strain, total angular momentum J = 3/2 spin–orbit systems can be protected against decoherence and have a T2 that rivals the best results for spin S = 1/2 electrons in silicon1–3 and S = 1 nitrogen-vacancy centres in diamond14.

We study hole spins bound to group-III acceptors in silicon (Fig. 1a, inset at bottom left), which have strong spin–orbit coupling. Although very little is known about the spin coherence of group-III acceptors14, their properties are very favourable for the implementation of scalable qubits when tuned by strain, allowing for the formation of a generalized-spin qubit. Mechanical strain lifts the Γ8 symmetry of valence-band holes, inducing an energy gap Δ at zero magnetic field that qualitatively changes the two lowest-lying qubit states (see Fig. 1a,b and Methods for acceptor Hamiltonian): for relaxed silicon, |{3/2, +3/2}, {3/2, +1/2}⟩, and for strained silicon, |{3/2, +1/2}, {3/2, −1/2}⟩. Here, |{3/2, mJ}⟩ are Bloch states with total angular momentum J = 3/2, angular momentum L = 1 for the atomic orbitals |pJz⟩, spin S = 1/2 and the projection of total angular momentum mJ = ±1/2 (light holes) and mJ = ±3/2 (heavy holes). While the |{3/2, +1/2}, {3/2, +1/2}⟩ subsystem is charge-like, the |{3/2, +1/2}, {3/2, −1/2}⟩ subsystem is a ‘generalized spin’; the corresponding wavefunctions are a time-reversal symmetric combination of spin and orbitals (lower two illustrations, Fig. 1d), while the spin S is not a good quantum number, as seen in the expression of the Bloch states13:

\[
\begin{align*}
|{3/2, \pm 1/2}⟩ &= \frac{1}{\sqrt{6}} (|p_+⟩ \pm |p_-⟩) ⊗ |S_+⟩ = \pm 1/2⟩ \\
|{3/2, \pm 3/2}⟩ &= \frac{1}{\sqrt{2}} (|p_+⟩ ± |p_-⟩) ⊗ |S_±⟩ = \pm 1/2⟩
\end{align*}
\]

where |S±⟩ = ±1/2⟩ are the spin up/down of S = 1/2. Notably, the qubit subsystems are fundamentally distinguished from electron-spin qubits since J = 3/2, not L or S, is a good quantum number.

The generalized spin has compelling properties for building spin–orbit qubits that couple to electric or elastic fields while maintaining long T2 (refs. 12,13,16). For holes, these couplings (blue arrows, Fig. 1c) take the form of quadrupolar tensor operators represented by quadratic forms of spin-3/2 matrices J±Jz (see Methods for acceptor Hamiltonian), having no analogue in the conduction band16. Combined with the J = 3/2 Zeeman interaction (orange arrows,
Fig. 1c), the quadrupolar couplings hybridize the generalized-spin states with different spin-orbital wavefunctions with higher energies (upper two illustrations, Fig. 1d) and endow the generalized spin with intrinsic spin–orbit coupling (red arrow, Fig. 1c). This scheme differs qualitatively from recent work on electron quantum dot systems in silicon where spin–orbit coupling is induced with extrinsic sources such as a charge qubit and micrometre-scale magnets. Recently it has been predicted that the quadrupoles allow longitudinal electric couplings responsible for qubit decoherence to be minimized while maintaining spin–orbit qubit functionality via large transverse electric coupling. Further advantages of acceptors in Si compared to conventional spin qubits include the removal of the nuclear spin bath by $^{28}$Si purification, single-atom addressability, the confinement of spin without gate electrodes and the reduced influence from charge traps at interfaces. Nevertheless, the coherence of acceptor-bound generalized spins has yet to be measured.

Here we experimentally demonstrate the long $T_1$ of the generalized spin defined by holes bound to boron acceptors in isotopically enriched silicon ($^{28}$Si:B, boron concentration is $\sim 10^{16}$ cm$^{-3}$). Without mechanical strain, this $^{28}$Si:B wafer shows a spin-echo signal at an effective g-factor $|g'|=1.17$ for $B_0 \parallel [110]$ (Fig. 2b), which is consistent with $|g'|=1.13$ in previous reports for the $\{3/2, +3/2\}$, $\{3/2, +1/2\}$ subsystem (black arrow, Fig. 1a). The $^{28}$Si:B sample is subjected to biaxial tensile strain (Fig. 2a) to obtain $\Delta$ exceeding the qubit energy splitting $\hbar \omega_0$, where $\hbar$ is the reduced Planck constant and $\omega_0/2\pi$ is the Larmor frequency (Methods for sample details). In this gapped configuration, we investigate the $\{3/2, +1/2\}$, $\{3/2, -1/2\}$ generalized-spin subsystem (Fig. 1c), which is predicted to have enhanced immunity to decoherence from electrical noise and is shown to have a longer longitudinal relaxation time $T_1$ (ref. 38). For comparison, a mechanically relaxed sample is also studied focusing on the $\{3/2, +3/2\}$, $\{3/2, +1/2\}$ subsystem (Fig. 1a). Coherence and longitudinal relaxation are studied by low temperature (~25 mK) pulsed electron paramagnetic resonance (EPR; Supplementary Information, section 1).

We experimentally confirm the formation of the generalized spin in the strain sample through its Hahn-echo spectrum. Figure 2c shows the spin-echo spectrum of the strained sample measured by a standard Hahn-echo sequence ($\pi/2-\pi-\pi/2$) with an interval $r$ (see Methods for spin-echo measurements). We observe a spin-echo signal over a broad range of $|g'|$ from 2.4 to 2.6 (Fig. 2c). No signal is found at $|g'|=1.17$ (not shown) in contrast with the relaxed sample (Fig. 2b), ensuring that the generalized spins are formed by properly applied strain. Another sharp signal at $|g'|=2.01$ is attributed to dangling-bond-related paramagnetic centres (P$_{\ell}$ centres) according to its g-factor of ~2.0 and short $T_1$ (not shown). We attribute the broad spin-echo signal in the strained sample to the $\{3/2, +1/2\}$, $\{3/2, -1/2\}$ generalized spin (black arrow, Fig. 1b), since a $|g'|$ in this range is expected for the configuration of strain and static magnetic field (Fig. 2a). The broadening and the detailed structure of the spin-echo spectrum is induced by strain inhomogeneity (Supplementary Information, sections 2 and 4).
rather than magnetic field inhomogeneity, since the P$_s$ centre signal is kept sharp. We provide a theoretical analysis about the spectral line shape in the Supplementary Information, section 4.

The Hahn-echo decay (Fig. 3a) shows the enhancement of $T_1$ in the strained sample (red) in comparison with the relaxed sample (black). We obtain $T_2$ measured by the Hahn-echo decay, $T_{2\text{rel}}$, by fitting to a compressed exponential $A \exp(-2t/T_{2\text{rel}}^2)$, where $A$ is a fitting parameter and $\beta$ reveals the temporal noise characteristics. For the strained sample, we observe a $T_{2\text{rel}}$ of 0.92 ± 0.01 ms with $\beta = 2.45$ at $B_0 = 175.7$ mT (green curve), which is much longer than $T_{2\text{rel}} = 23 ± 1$ ms with $\beta = 1.05$ for the relaxed sample (blue curve). We observe an improvement in $T_{2\text{rel}}$ over the full range of $|g'| = 2.4-2.6$: $T_{2\text{rel}} = 0.93 ± 0.02$, $0.97 ± 0.02$ and $0.78 ± 0.03$ ms and $\beta = 3.5, 3.0$ and 2.9, for $B_0 = 190.5, 182.0$ and 171.7 mT, respectively (Supplementary Information, section 5). This improvement compared to the relaxed sample is attributed to the strain-engineered quadrupole coupling as discussed later. We also find a noticeable component of fast decay ($2\tau < ~300$ µs, black dashed line) for the strained sample. A spin ensemble showing slow and fast decay components indicates that it is split to spatially separated ensembles experiencing different noise, with the generalized-spin subset less affected by noise responsible for the slow decay with $T_{2\text{rel}}$ of 0.92 ± 0.01 ms. Theoretical analysis shows that the fast decay can be explained by surface defects and the dipole–dipole interaction between the generalized spins (Supplementary Information, section 6).

Figure 3b shows $T_1$ measurements by an inversion-recovery pulse sequence $(\pi/2)\rightarrow(t)\rightarrow-(\pi/2)\rightarrow-(t)$, with an interval $t'$ (top left; see also Supplementary Information, section 3). Inversion recovery signals are well fitted by an exponential function $A - B \exp(-t'/T_1)$ where $B$ is a fitting parameter in both samples (solid curves in Fig. 3b); we obtain $T_1$ values of $5 ± 1$ ms and $85 ± 9$ µs for the strained and relaxed samples, respectively. The $T_1$ improvement in strained Si:B is consistent with a previous experimental report. We attribute this to the strain-induced gap, which suppresses spin–photon coupling, and note that increasing the strain or reducing the magnetic field should enhance $T_1$, more if desired. We also note that the $T_{2\text{rel}}$ and $T_1$ of the relaxed sample are substantially longer than those observed in boron acceptors in natural silicon (~2 µs and 4 µs, respectively)\(^{1,31}\), which could be explained by the lower temperature than in the previous report, while the reduced $^{28}$Si concentration of our sample could also play a role.

Before making a broad comparison of our results, we remark on how they compare to electronic Hahn-echo coherence times in isotope-purified silicon. Remarkably, $T_{2\text{rel}}$ of the generalized spin $[3/2, ±1/2]_g$ in the strained sample is comparable to the $T_{2\text{rel}}$ of ~3 ms obtained for P donors with $10^{15}$ cm$^{-3}$ concentration in $^{28}$Si (ref. 1). The spin-echo decay curves of the strained sample have $\beta = 2.5-3.5$ such that decoherence is induced by slow fluctuations (spectral diffusion)\(^{12-13}\). By contrast, the $^{28}$Si:P ensemble has $\beta \approx 1$ due to decoherence from instantaneous diffusion caused by EPR-driven flips of neighbouring electron spins\(^{12-13}\). This process can be partially suppressed to yield $T_{2\text{rel}} \approx 100$ ms for $^{28}$Si:P, while it is suppressed by the inhomogeneous broadening in $^{28}$Si:B, which is still comparable to our result. Importantly, $T_{2\text{rel}}$ is also comparable to measured values for state-of-the-art electron-spin qubits defined by $^{28}$Si quantum dots with very weak spin–orbit coupling\(^{12-13}\), and exceeds measured values for electron-spin qubits with extrinsic spin–orbit coupling induced by integrated micromagnets\(^{12-13}\).

Decoherence due to spectral diffusion can be ameliorated by dynamical decoupling. Figure 4 shows the refocussed echo intensity in the strained sample as a function of time after the first ($\pi/2$) pulse, measured by the Carr–Purcell–Meiboom–Gill (CPMG) pulse sequence (top right), $(\pi/2)\rightarrow(-\pi)\rightarrow-2(-\pi)\rightarrow-(\pi/2)$ with $t = 10$ ms, which suppresses decoherence by filtering out noise over the whole frequency range except for $1/4\tau = 25$ kHz (ref. 4). We find a $T_{2\text{CPMG}}$ of 9.2 ± 0.1 ms, ten times (400 times) longer than $T_{2\text{rel}}$ for the strained (relaxed) sample, and over four orders of magnitude longer than reports of intrinsic spin–orbit-coupled solid-state systems\(^{11-18}\). Our 10 ms coherence time should not be regarded as a limit and can be improved by strain engineering and proper choice of
The observed improvement of $T_\text{rel}$ in the strained sample ($\sim 10^2$ for a typical electric-field noise of $\sim 10^4$ in Si quantum dots). The ratio $\Omega_2/\delta_\omega \approx 5.000$ indicates many quantum gate operations can be performed before the qubit is dephased, implying that the longitudinal and transverse dips can be controlled favourably. This ratio will be further improved by aligning the magnetic field to the [010] direction of silicon. Another figure of merit of a qubit is the ratio between $1/T_1$ and $\Omega_2/2\pi$, characterizing the number of quantum gate operations that can be executed before the qubit relaxes. This ratio can be improved by larger strain because $1/T_1$ is suppressed by $\delta_\omega/2\Delta$. Notably, it has recently been shown that monolithically fabricated silicon field-effect transistors can present a very small strain environment, and thus the engineering of intentional strain should enable the implementation of acceptor-based generalized-spin qubits with long $T_1$ values and spin–orbit functionality in field-effect transistors.

Together with long $T_1$ values, several mechanisms could be employed to realize hybrid spin–photon systems or long-range spin–spin interactions via photons in superconducting microwave cavities. Despite the longitudinal and transverse dipoles having the same dependence on strain, we expect that the transverse coupling can be engineered without increasing the longitudinal coupling by combining strain and electric field. This is in line with the key strategy to engineer the spin–photon interaction to realize the strong coupling regime without increasing decoherence. This could be achieved by modulating periodically in time the transverse or longitudinal couplings to enhance the spin–photon interaction. Indeed, an oscillating control electric field $E_\text{c}(t)$ can be used to periodically modulate $v_\text{c}(E(t))$ and $\chi(E(t))$ via the second-order effect of the electric field attributed to the quadrupolar coupling of hole generalized spins (Supplementary Information, section 7). Alternatively, the transverse coupling to x- and y-oriented electric fields could be statically enhanced using a z-oriented electric field and interface via a Rashba-like quadrupolar interaction at a sweet spot with long $T_1$ values (ref. 23). This idea is based on a remarkable difference in the electric-field dependence of $v_\text{c}(E)$ and $\chi(E)$: $\chi(E)$ is insensitive to $E$, while $v_\text{c}(E)$ is sensitive (Supplementary Information, section 7). Owing to this difference, $E$ along the z axis enhances $v_\text{c}(E)$ without changing the $\chi(E)$ up to higher-order terms for generalized spins. This means that the generalized-spin qubit allows control of the transverse coupling without increasing the longitudinal coupling by $E$ along $z$. Phonon coupling stands out as an alternative qubit-coupling mechanism, and the coherence properties we have verified make Si:B hole generalized spins interesting candidates for phonon-coupled hybrid systems using silicon mechanical
resonators. Systems where holes are allowed to tunnel between neighbouring quantum dots or Si:B sites will experience spin–orbit coupling, which could be useful to realize exotic spin–orbit-coupled states. It should be possible to combine these spin–orbit functionalities with long $T_\text{s}$ values because the transverse coupling could be engineered without increasing the decoherence induced by the longitudinal coupling to electric fields.

In conclusion, this work shows that spin–orbit coupled $J=3/2$ systems can have 10 ms coherence times rivalling the best $S=1/2$ and $S=1$ systems in silicon and diamond, respectively. This is highly non-trivial because total angular momentum, rather than spin or charge, is quantized. This opens a pathway to build engineered quantum systems and improve the scalability of spin-based qubits for quantum information science and technologies.

Online content
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Fig. 4 | Dynamical decoupling. Decay of spin echo refocused by the CPMG pulse sequence (top right diagram) with $\tau = 10$ μs in the strained sample as a function of elapsed time after the first $(\pi/2)_Y$ pulse. Based on exponential fit to the data (solid line), we obtain $T_{\text{CPMG}} = 9.2 \pm 0.1$ ms.
Methods

Accepter Hamiltonian. Si:B holes couple to uniform magnetic, electric and elastic fields as described in the literature. The coupling Hamiltonian to the fields is expressed as follows:

$$\hat{H}' = \hat{H}'_A + \hat{H}'_{\text{elec}} + \hat{H}'_{\text{el}} + \hat{H}'_{\text{mag}}$$

where the $x$, $y$ and $z$ axes correspond to the [100], [010] and [001] axes of the silicon crystal, respectively, $\mu_B$ is the Bohr magneton, $\mathbf{E}$ are electric fields and $\mathbf{e}_a$ are normal (shear) strains for $i=j$ ($i \neq j$). Quadrupole operators $Q_i$ ($i=x, y, z$) for $I=3/2$ are expressed by angular momentum operators $\hat{J}$, as $Q_i = \{\hat{J}_i, \hat{J}_j\}/2 - S_{ij}S_{ij}/4$, where $\{\hat{J}_i, \hat{J}_j\} = \hat{J}_i \hat{J}_j + \hat{J}_j \hat{J}_i$, $S_{ij}$ is Kronecker's delta, and $\hat{J}$ is the identity operator. Notice that all interactions with electrical and elastic degrees of freedom are proportional to $Q_i$ (refs. 36, 37) and as such these interactions couple the light and heavy holes. While $\hat{H}'_A$ generally includes a hydrostatic term such as $\mathbf{d} \cdot \mathbf{E}$, it does not cause any relative energy change of the $J=3/2$ hole states and thus is dropped here.

For the coefficients, we use $g$-factors $g_E = -1.07$ and $g_B = -0.03$ from ref. 36, linear electric-field coupling coefficient $p = 0.26$ D from ref. 37 and deformation potentials $\mathbf{b}' = -1.42$ eV and $\mathbf{d}' = -3.7$ eV from ref. 38. $b$ and $d$ are cubic electric-field coupling coefficients. While there is no experimental information on the values of $b$ and $d$, we estimate them at approximately $-3$ D MV$^{-1}$ m and approximately $-5$ D MV$^{-1}$ m, respectively, based on the effective mass approach in ref. 38.

By numerically diagonalizing $\hat{H}$ in equation (1) with certain sets of $\mathbf{E}$ and $\mathbf{e}_a$ as a function of $\mathbf{B}_0$, we obtain eigenenergy spectra as shown in Fig. 1a,b. In Fig. 1c, $\mathbf{e}_p = (1 \neq \hat{J})$ and $\mathbf{B}_0 || [110]$ for Fig. 1c, and $\mathbf{e}_p = \mathbf{e}_z = 0.02\%$, $\mathbf{e}_y = -0.0156\%$ and $\mathbf{B}_0 || [110]$ for Fig. 1c. ($\mathbf{E}$ and $\mathbf{e}_y$ (1 $\neq \hat{J}$) are kept zero.)

Sample details. Both of the mechanically relaxed and strained samples are prepared from pieces of the same uniformly boron-doped $^{30}$Si wafer with dimensions of $4.0 \text{ mm} \times 3.5 \text{ mm}$ in area and $500 \mu\text{m}$ in thickness. The purity of $99.99\%$ and boron concentration $n_0$ of $1.0 - 1.5 \times 10^{16} \text{ cm}^{-3}$. A diced piece of this crystal is used for the relaxed sample. To prepare the strained sample, another piece is thinned down to $50 \mu\text{m}$ and glued to a fused-silica chip (5.0 mm $\times$ 3.5 mm in area and $500 \times 500 \mu\text{m}$ in area and $1 \text{ mm}$ in thickness) using a two-component epoxy adhesive (Fig. 2a). While both the $^{30}$Si and fused-silica chips in this stack are mechanically relaxed at room temperature, biaxial tensile strain is applied to the $^{30}$Si chip at low temperature owing to the difference in the thermal expansion coefficient of the two materials. The magnitude of the strain applied to the strained sample is discussed in the Supplementary Information, section 2.

To perform EPR spectroscopy at milli-Kelvin temperatures, we use a superconducting coplanar waveguide cavity with a small mode volume. The cavity is fabricated from 100-nm-thick niobium film and consists of a 20-μm-wide centre conductor and 12-μm-wide separations between the centre conductor and ground plates, so that the characteristic impedance is 50 Ω on a 400-μm-thick highly resistive silicon substrate. The centre conductor length is 28 mm, designed to obtain the fundamental resonance mode of ~2.1 GHz. Each sample is mounted to the cavity in an independent experimental run to avoid overlapping signals from different samples. To couple to the cavity modes, the samples are directly placed on the cavity surface and closely fitted so that the polished silicon surface faces the cavity structure, and then fixed by GE Varnish. The samples cover at least one antinode and one node of the microwave magnetic field.

Spin-echo measurements. The Hahn-echo spectra in Fig. 2b,c are measured by a Hahn-echo sequence $(\pi/2)_H \rightarrow \mathbf{−}(n) \rightarrow (\pi/2)_{H}$, with $(\pi/2)_{H}$ and $(\pi)$ pulses separated by a time interval $\tau$, where the $X$ and $Y$ subscripts indicate the rotation axes in a qubit subsystem. The effective $g$-factor $g_E$ is obtained by equating the microwave angular frequency $\omega_{\text{microwave}}$ with $\omega_B = |I| \mu_B B_0/\hbar$ where $\mu_B$ is the Bohr magneton. For the relaxed (strained) sample, $\omega_{\text{microwave}}/2\pi$ of 6.255 GHz (6.331 GHz) is used. More details are presented in the Supplementary Information, section 3.

Data availability

The data represented in Figs. 1–4 are provided with the paper as source data. All other data that support results in this Article are available from the corresponding author upon reasonable request. Source data are provided with this paper.

Code availability

The custom codes that were used for drawing energy level diagrams and fitting are available from the corresponding author upon reasonable request.

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Author contributions

T.K., J.S. and S.R. designed the experiment. T.K. carried out the experiments (except for X-ray diffraction) and analysed the data, with input from J.S., J.v.d.H., C.C., B.C.J., J.C.M. and S.R.; T.K. and J.S. and D.C. carried out the theory calculations. T.K. and J.S. performed the numerical simulation of the strain distribution. W.D.H. carried out the X-ray diffraction analyses. H.R., N.A., P.B. and H.-J.P. supplied the boron-doped $^{30}$Si crystal. All authors discussed the results. T.K. wrote the manuscript with contributions from all authors.

Competing interests

The authors declare no competing interests.

Additional information

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