

Supplementary Information – Spin-Orbit Coupling in Silicon for Electrons Bound to Donors

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I. SINGLE-SHOT SPIN READ-OUT

Electron spin relaxation rates were obtained by single shot spin readout [1–4], using spin selective tunneling of donor-bound electrons to the nearby island of a single electron transistor (SET). The drain current of the SET as a function of V_{G1} and V_{G2} ($V_D = 750 \mu\text{V}$, $V_{SET} = V_T = 1 \text{ V}$) is plotted in Fig. S1a. Here, diagonal lines are Coulomb blockade (CB) peaks of the SET with two parallel lines of charge offsets (dashed white lines) indicating electron transfers between donor and SET island whenever their electrochemical potentials align (compare Fig. S1c) [2]. The presence of precisely two charge-offset lines indicate charge transitions $D^+ \leftrightarrow D^0$ and $D^0 \leftrightarrow D^-$ between the donor's three possible charge states, D^+ (0 electrons), D^0 (1 electron), D^- (2 electrons). The separation of the charge offset lines in gate voltage allows to extract an electron addition energy, $E_{add} = (50 \pm 7) \text{ meV}$, consistent with previous measurements in STM-patterned devices [5, 6]. No further charge transitions on the donor were observed within the accessible gate voltage range $\simeq -500 \text{ mV}$ to $\simeq 1000 \text{ mV}$, applied to all gates [4].

In Fig. S1b, we show a close-up where the electrochemical potential of the SET aligns with that of the donor D^0 charge state, allowing $D^+ \leftrightarrow D^0$ charge transitions [2]. An external magnetic field B_{ext} splits the spin-degenerate D^0 $1s(A_1)$ state into $|\uparrow\rangle$ and $|\downarrow\rangle$, separated by the Zeeman energy $\Delta E_Z = g\mu_B|B_{\text{ext}}|$, such that $\Delta E_Z \gg k_B T_{el}$ (low temperature limit [2]).

Electron spin read-out was facilitated by a three-level pulse sequence (Fig. S1d) [2–4] on gates G_1 and G_2 , upon monitoring the time dependent response $I_{\text{SET}}(t)$ (Fig. S1e) of the SET drain current (bandwidth of 3 kHz). In the first phase of the pulse, the spin-split electrochemical potentials are lowered beneath that of the SET to initialize a spin of arbitrary orientation, $|\uparrow\rangle$ or $|\downarrow\rangle$. During the following read-out phase, the donor electrochemical potentials are raised as shown in Fig. S1c, such that $|\uparrow\rangle$ above and $|\downarrow\rangle$ below the electrochemical potential of the SET. This now only allows spin-up electrons to tunnel

to the SET, followed by a spin-down electron tunneling back to the donor. As result, a single rectangular current pulse is observed in the SET current response $I_{\text{SET}}(t)$ at the start of the read-out phase (Fig. S1e, red trace). If a $|\downarrow\rangle$ was initialized, the current response of the SET stays flat (Fig. S1e, black trace). In the final phase of the pulse, the spin-split electrochemical potentials are raised above that of the SET to vacate the donor to reset the gate pulse sequence. As previously shown, we achieve a spin detection fidelity as large as 99.6% in this device [4], due to the large "on" state currents of the SET and optimized electron tunnel rates.

The probability $P_{\uparrow}$ to find an electron in the $|\uparrow\rangle$ state as a function of the wait time t after initializing is plotted in Fig. S1f. Each data point is based on 100–250 (depending on wait time) repetitions of the same pulse cycle. We show data from three different magnetic field orientations at $B_{\text{ext}} = 3.5 \text{ T}$. Spin lifetimes T_1 were extracted from these data by exponential fits $P_{\uparrow}(t) = P_{\uparrow}(0) \propto \exp(-t/T_1)$ (black lines) varying between $T_1 \simeq 25 \text{ ms}$ for $(B||[001])$ and $T_1 \simeq 150 \text{ ms}$ for $(B||[110])$.

II. ELECTRON TEMPERATURE / GATE LEVER

The electron temperature of the SET can be extracted [2] by scanning the electrochemical potential of the donor with respect to that of the SET island around the $D^+ \leftrightarrow D^0$ transition (see blue arrow in Fig. S2a, inset). Examples of the averaged and normalized SET current at two mixing chamber temperatures ($T_{M/C} = 21 \text{ mK}$ and $T_{M/C} = 675 \text{ mK}$) are shown in Fig. S2a. Here, each trace has been averaged over 100 repetitions of a slow ($f = 0.5 \text{ Hz}$) gate voltage ramp (amplitude 4–16 mV, depending on M/C temperature). These data can be fit with a Fermi-function,

$$\langle I_{\text{SET}} \rangle = \frac{1}{1 + \exp[\alpha(V_g - V_0)/k_B T_{el}]} \quad (1)$$

where gate voltage difference ($V_g - V_0$) and energy are related by the effective gate lever α . From the fit, we can

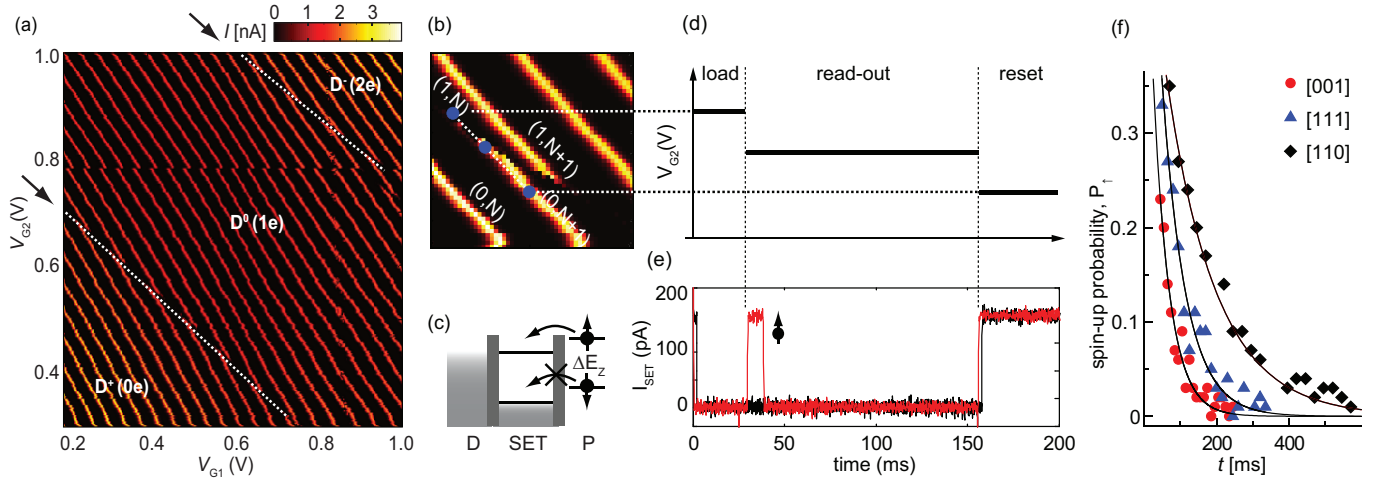


Figure S1. **Single-shot spin readout of a single P donor qubit.** **a**, Charge stability diagram, recorded at $V_D = 750 \mu\text{V}$ and $V_{SET} = V_T = 1 \text{ V}$. Two parallel lines of charge offset breaks (black dashed black lines and arrows) correspond to the single donor $D^+ - D^0$ and $D^0 - D^-$ transitions (The horizontal offset line at $\approx 800 \text{ meV}$ indicates is due to a spontaneous charge rearrangement in the device during measurement). Spin read-out was performed at a region as shown in **b**, where the electrochemical potential of the donor and the SET island align to allow $D^+ - D^0$ charge transitions (**c**). **d**, Gate-pulse sequence and **e**, time-dependent SET current response. **f**, Spin-up probability $P_\uparrow$ as a function of wait time t after initialization for three orientations of the magnetic field with $B \parallel [001]$, $B \parallel [111]$, and $B \parallel [110]$. Black lines show fits to the data to extract T_1 .

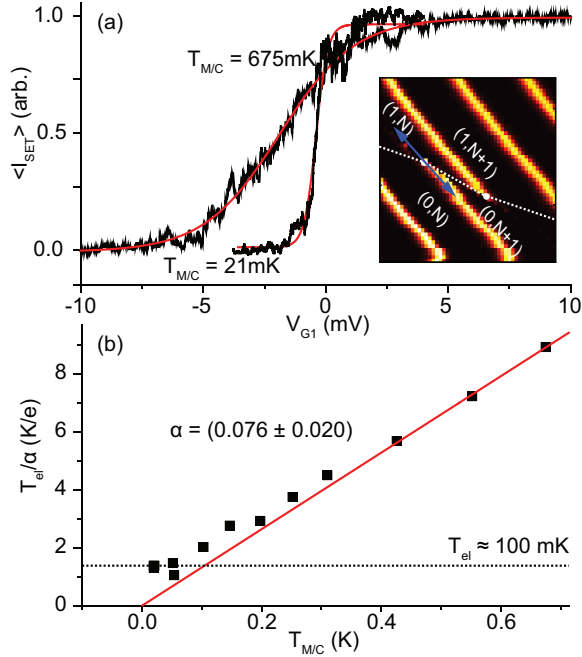


Figure S2. **Electron temperature and gate lever α .** **a**, Time-averaged current response $\langle I_{SET} \rangle$ across 100 repetitions of slow gate ramp and for two temperatures of the dilution fridge mixing chamber ($T_{M/C}$). The direction of the gate ramp is indicated by a blue arrow in the inset. The ratio of electron temperature and lever arm (T_{el}/α) is extracted by fitting to a Fermi distribution (eq. (1)). **b**, The ratio of electron temperature and lever arm (T_{el}/α) is plotted versus mixing chamber temperature. A linear fit allows extraction of the gate lever arm $\alpha = (0.076 \pm 0.02)$ and electron temperature $T_{el} \approx 100 \text{ mK}$.

extract the temperature-broadening parameter $\alpha/k_B T_{el}$, with Boltzman constant k_B and electron temperature T_{el} . T_{el}/α is shown as a function of the mixing chamber temperature $T_{M/C}$ in Fig. S2b. From a linear fit to the data (red line), we extract the an effective gate lever $\alpha = (0.076 \pm 0.02)$, and an approximate electron temperature $T_{el} \approx 100 \text{ mK}$ where T_{el}/α saturates (black dashed line).

III. SPIN-RELAXATION THEORY

Historical background: Roth and Hasegawa's mechanism in bulk donors

Spin-lattice relaxation of electron spins in bulk donors has been investigated since the 1960s [7–9]. It is well established that the electron spin relaxes through interacting with acoustic phonons in the presence of spin-orbit coupling (SOC). The magnitude of $1/T_1$ with respect to the alignment of an external magnetic field B_{ext} depends on how B_{ext} aligns with the effective magnetic field produced by the crystal's spin-orbit coupling. As a result, spin relaxation rates have been shown both theoretically and experimentally to be anisotropic with respect to the crystal lattice as shown in Fig. S3a [10].

Two mechanisms have been shown to cause spin relaxation, namely the *valley repopulation* mechanism and the *single valley* effect. First described by Hasegawa [7], the valley repopulation relaxes the electron spin by mixing the donor ground state with states of different valley compositions and along different crystalline directions

through diagonal phonon strain. In contrast, the single valley effect was described by Roth [8] and refers to relaxation within each valley, where the bottom of the conduction band mixes with the next higher band through off-diagonal phonon strain. The measured anisotropic T_1 behavior of a bulk P donor in Si is described in Ref. [10], and also reproduced here in Fig. S3a with black dotted data points. Note that these experimental data points of Wilson and Feher [10] were explained by the sum of both the relaxation rates due to valley repopulation and the single valley effect.

Theoretically $1/T_1$ due to valley repopulation alone is shown separately in Fig. S3b. Here, a maximum $1/T_1$ occurs when the external magnetic field B_{ext} is aligned along crystalline directions $\langle 111 \rangle$, and a minimum whenever B_{ext} points along $\langle 100 \rangle$. The $1/T_1$ anisotropy can be described by a sinusoidal anisotropy function $f(\theta, \phi)$ with a periodicity of π [7, 10], with the following form (Fig. S3b):

$$f(\theta, \phi) = \sin^2 \theta (4 \cos^2 \theta + \sin^2 \theta \sin^2 2\phi) \quad (2)$$

For the case of the single valley effect, independently shown in Fig. S3c, the opposite holds true: A maximum $1/T_1$ occurs when $B_{\text{ext}} \parallel \langle 100 \rangle$ and a minimum when $B_{\text{ext}} \parallel \langle 111 \rangle$. The $1/T_1$ anisotropy function due to the single valley effect [7, 10] is of the form (Fig. S3c):

$$f(\theta, \phi) = \cos^4 \theta + \frac{1}{2} \sin^4 \theta (1 + \cos^2 2\phi) \quad (3)$$

Summed together these two effects can account for the experimentally observed anisotropy (black line in Fig. S3a) in bulk donors [10].

Measured anisotropy in a nanoelectronic device

Fig. S3d shows the measured $1/T_1$ anisotropy found in this work for a donor within a nanoelectronic device. Comparison between the experimental curves in panels a) and d) of Fig. S3 highlights the completely different anisotropy observed from Feher's bulk measurements. The anisotropy observed in our experiment neither agrees with panel b) nor with c), and notably has a much higher value of $1/T_1$. In addition, no combination of the valley repopulation and single valley effect can reproduce this anisotropy. This motivates us to consider what environmental factors are present in a nanoelectronic device that were not taken into account in the bulk relaxation theories.

Donors in a nanoelectronic device: Role of strain, electric fields, and Johnson noise

In contrast to the results for bulk donors in silicon, recent experiments in the context of donor based quantum computing have enabled the realization of spins on

single donors fabricated within a nanoelectronic device [5]. In this paper we have shown experimentally that the anisotropy of the spin relaxation rate of a donor in a nanoelectronic device is significantly different from that in bulk. Whereas the anisotropy observed in T_1 in bulk donors can be explained by a combination of the valley repopulation and the single valley effect, in a nanoelectronic device we also need to consider the effects such as strain due to surrounding device contacts, electric fields due to applied gate voltages, and Johnson noise on the device electrodes. In addition to the valley repopulation and single valley effect, a donor in a nanoelectronic device may be susceptible to additional relaxation mechanisms with their own specific anisotropy functions. Therefore, by measuring the anisotropy function of $1/T_1$, we can determine if the silicon lattice is strained or if electric fields or noise sources are present. Strain and electric fields can modify the relaxation mechanisms in two aspects. First, both strain and electric field can cause mixing between the donor valley states and repopulate the valleys [10, 11]. Second, having electric fields in the crystal introduces additional SOC. Considering these two effects could greatly modify the valley repopulation mechanisms. The single valley effect, however, is minimally affected by uniaxial or biaxial strain, which is the type of strain most likely to be in the device as a result of the device geometry, and electric fields. This is because uniaxial or biaxial strain and electric fields cannot cause mixing between the bottom two conduction bands, important in the single valley effect [9–11]. Noise, on the other hand, can couple the ground spin states to each other directly and cause relaxation without the mixing between valley states, as in valley repopulation, or the mixing between conduction bands, as in the single valley effect. Below we analyze each of the above-mentioned relaxation mechanisms and determine whether their anisotropy functions can explain the measured data.

A. Strain: modified valley repopulation

In addition to the time-dependent lattice strain which causes the phonon relaxation, the surrounding contacts made of delta-doped layers might produce static strain in the device. If the lattice is under static uniaxial strain, valleys repopulate according to the strain direction. This leads to a different anisotropy which depends on the strain direction. For example compressive strain along $[001]$ yields a maximum at $\theta = 45^\circ$ (between $[111]$ and $[001]$) and a minimum at $[100]$, as shown in Fig. S3e. The anisotropy function due to valley repopulation under compressive uniaxial strain along $[001]$ [7] is given below:

$$f(\theta, \phi) = \sin^2 \theta (\cos^2 \theta \cos^2 2\phi + 4 \sin^2 \phi \cos^2 \phi) \quad (4)$$

We have considered all uniaxial strain directions and

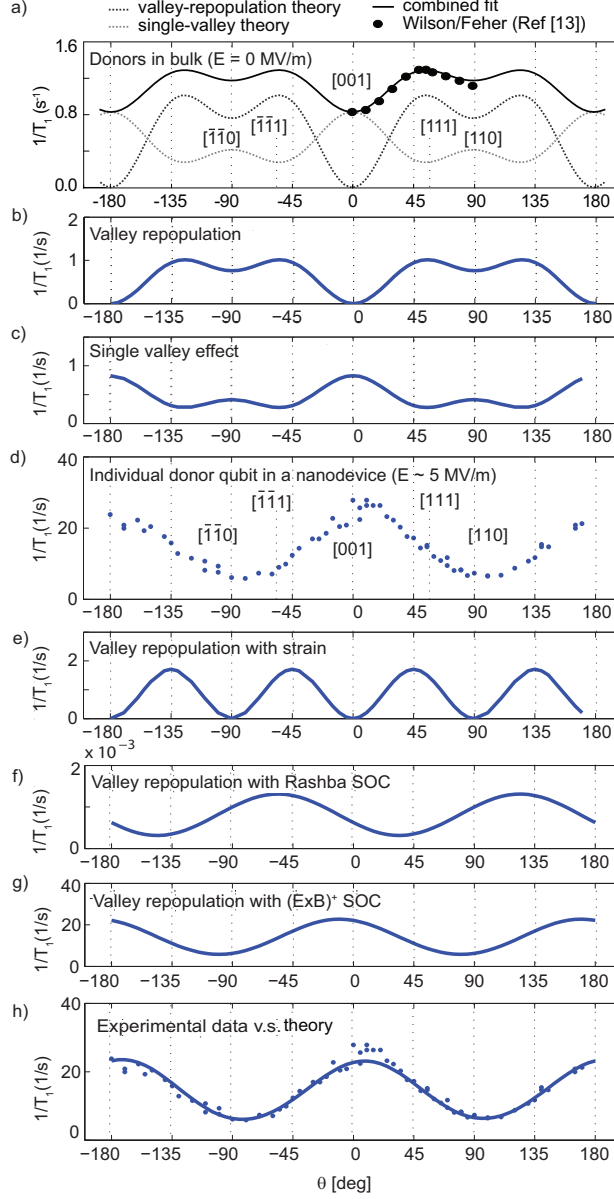


Figure S3. **Comparison of spin relaxation anisotropy measurements with various spin relaxation mechanisms.** **a**, Spin relaxation for bulk donors. **b**, Valley repopulation with bulk spin-orbit. **c**, Single valley effect with bulk spin-orbit. **d**, Experimental data ($\phi = 45^\circ$). **e**, Valley repopulation under compressive uniaxial strain along z with bulk spin-orbit. **f**, Valley repopulation under dominating electric field along y with Rashba spin-orbit. **g**, Valley repopulation under dominating electric field along y with H_{EB} spin-orbit. **h**, Comparison of the experimental $1/T_1$ with the combined theory curve, which includes the effect of b), c), f) and g).

found that no matter how the valley repopulates, strain alone cannot reproduce the experimentally observed anisotropy.

B. Electric fields: Rashba spin-orbit coupling

Applied electric fields within a nanoelectronic device gives rise to the Rashba SOC [12]. The Rashba Hamil-

tonian, described in Eqn. (2) of the main text, couples the electron spin with the electric field and the electron's momentum.

$$H_{Rashba} = (\mathbf{k} \times \mathbf{E}) \cdot \overleftrightarrow{R} \cdot \boldsymbol{\sigma} \quad (5)$$

The valley repopulation mechanism in the presence of a Rashba coupling can yield an anisotropy function that is dependent on the electric field distribution of the device. From detailed electrostatic modelling using COM-

SOL (described in the main text), we expect a dominating electric field pointing in the y direction (i.e. $[010]$) due to the device geometry and applied biases. With this field distribution, $1/T_1$ due to Rashba SOC is shown in Fig. S3f. The analytically derived anisotropy function for valley repopulation under an electric field along y (i.e. $[010]$) with Rashba SOC [13, 14] is given as:

$$f(\theta, \phi) = \frac{1}{2}(1 - \sin 2\theta \cos \phi + \sin^2 \theta \sin^2 \phi) \quad (6)$$

This curve has a minimum between $\theta = 0^\circ$ and $\theta = 45^\circ$, and a maximum between $\theta = 90^\circ$ and $\theta = 135^\circ$, almost exactly opposite to the experimental curve. The magnitude is also found to be almost three orders lower than the experimental data. The presence of Rashba SOC alone therefore cannot reproduce the experimental data.

C. Electric fields: H_{EB} spin-orbit coupling

In the presence of electric and magnetic fields, while Rashba SOC couples the electron spin with the electron's momentum, the higher order spin-orbit coupling also allows coupling of the electron spin with external magnetic field and electric fields. Such a SOC has previously been considered to describe hole spins [15]. A more recent derivation, based on $\mathbf{k} \cdot \mathbf{p}$, was given by Winkler [12]. The Hamiltonian, shown in Eqn. (1) of the main text is

$$H_{EB} = \begin{pmatrix} E_y B_z + E_z B_y \\ E_z B_x + E_x B_z \\ E_x B_y + E_y B_x \end{pmatrix}^T \cdot \mathbf{C} \cdot \boldsymbol{\sigma} \quad (7)$$

For example, H_{EB} SOC from an electric field along y couples spins pointing along x and z , but not spins pointing along y , giving rise to a highly anisotropic $1/T_1$. The relaxation rate due to valley repopulation in the presence of this H_{EB} SOC under the electric field in the device is shown in Fig. S3g. Remarkably, this curve shows a maximum at $\theta \sim 0^\circ$ and a minimum $\theta \sim 90^\circ$, in very close agreement with the experimental data. The magnitudes are also close to the experimental curve, as shown in Fig. S3g. The analytically derived anisotropy function of valley repopulation with H_{EB} SOC under electric field in y (i.e. $[010]$) is given below:

$$f(\theta, \phi) = \cos^2 \theta \sin^2 \phi + \cos^2 \phi \quad (8)$$

The relaxation mechanism of H_{EB} captures both the experimentally observed anisotropy and the magnitude of $1/T_1$.

By now considering all the effects of valley repopulation with Rashba SOC and H_{EB} SOC, as well as the single valley effect, with the exact electric fields in the device, we can determine the final anisotropy shown as the solid line in Fig. S3h. As we can see this provides a very good fit to the experimental data.

D. Evanescent wave Johnson noise

In this section we consider the effect of Evanescent wave Johnson noise (EWJN). Previous discussions with the different spin relaxation mechanisms are all based on the interaction of the electron spin with phonons. While phonon fields can couple the donor ground state to the excited valley states, and affect T_1 through valley repopulation, EWJN can directly couple the ground donor spin states to each other, leading to spin relaxation.

The relaxation rate can be written as [16]

$$\frac{1}{T_1} = f(\theta, \phi) S(\omega_Z) \quad (9)$$

where $S(\omega)$ is the magnetic field spectrum, ω_Z is the angular frequency of the Zeeman splitting of the donor ground spin states.

As shown in Fig. 1a in the main text, both the gates and donor are in-plane. Since the noise that can reach the donor must propagate through the plane, and the electric field can only fluctuate in-plane, the magnetic field fluctuates in the z direction. With this fluctuating magnetic field the resulting anisotropy function is

$$f(\theta, \phi) = \sin^2 \theta \quad (10)$$

The fluctuating electric field can couple the spins by producing an effective magnetic field through Rashba SOC. The wavefunction is asymmetric in the y direction with the strong static electric field along y . The matrix element of the operator $\mathbf{k}$ in the Rashba SOC therefore points in the y direction. Since both the matrix element of the operator $\mathbf{k}$ and the electric field from the noise are in-plane, the effective magnetic field ($B_{eff} = \mathbf{k} \times \mathbf{E}$) points in the z direction. The resulting anisotropy function from fluctuating electric field is the same as Eqn. 10.

The spectrum $S(\omega)$ of the effective magnetic field ($B_{eff} = \mathbf{k} \times \mathbf{E}$) has a linear B to B^3 dependency, since it depends on the electric field spectrum $S_E(\omega)$ of EWJN, which is proportional to B [16], and the operator $\mathbf{k} = -i\nabla + \frac{e}{\hbar}\mathbf{A}$, which has a constant to linear B dependency as the magnitude of the vector potential $\mathbf{A}$ is proportional to the external magnetic field B .

If we now compare these results to the experiment observation, in Fig. 1f in the main text we see that $1/T_1$ has a B^5 dependency, in contrast to the linear B to B^3 dependency expected with EWJN. Furthermore the $1/T_1$ caused by EWJN has its minimum at $\theta = 0^\circ$ (Eqn. 10), different from the experiment measurements (As shown in Fig. S3d, which has minimum at $\theta = 90^\circ$). We therefore conclude that spin-relaxation caused by EWJN cannot explain our measurement results.

Dominant relaxation mechanism in the experiment

From the above comparison of the measured anisotropy data with the anisotropy functions of the various mechanisms, we can make two important conclusions. First, H_{EB} -dominated spin relaxation is the only spin-relaxation mechanism which correctly describes the measured anisotropy and magnitude of the relaxation rates. Here the anisotropy shows a 180° phase with a maximum at [001] and minimum at [110] for out-of-plane rotation. Second, its spin-relaxation rates are one order of magnitude larger than any other spin-relaxation mechanisms which makes H_{EB} -driven spin-relaxation the only mechanism, able to describe our data.

IV. QUANTITATIVE ANALYSIS OF THE EXPERIMENTAL DATA

Having derived the $1/T_1$ anisotropy of each of these mechanisms, and found that an electric field induced higher order spin-orbit coupling of the form H_{EB} has the same anisotropy as the measured data, we now describe how we obtained the quantitative description of the anisotropy data.

We need to obtain the strengths of each of the relaxation mechanisms described above. The magnitude of the valley repopulation and the single valley effect are known from the experiments of Wilson and Feher [10]. For the remaining mechanisms of Rashba and H_{EB} , we determined their strengths given by the coefficients R and C of Eqn. 5 and Eqn. 7, respectively. Furthermore, since these mechanisms are dependent on the electric field, we obtained the electric field components (in particular, the ratio of E_x , E_y , and E_z) in the device. We treat R , C , and the ratio of the E -field components as fitting parameters to the experimental $1/T_1$ data points to reproduce the experimental curves of Fig. 2 of the main text.

Functional form of the $1/T_1$ anisotropy due to all mechanisms combined

In order to obtain the contributions of Rashba and H_{EB} SOC driven spin relaxation, in the main text we fitted the measured spin relaxation rates with an expression which adds all the valley repopulation mechanisms, Rashba SOC and H_{EB} SOC and the single valley effect, together.

$$\begin{aligned} \frac{1}{T_1} = & C_{\text{valleyrep.}} |(g_l - g_t) \mu_B \mathbf{B} \cdot \overleftrightarrow{D} \cdot \sigma^{\uparrow\downarrow} \\ & + (R_l - R_t) (\mathbf{k}_{0r} \times \mathbf{E}) \cdot \overleftrightarrow{D} \cdot \sigma^{\uparrow\downarrow} \\ & + (C_l - C_t) (\mathbf{E} \times \mathbf{B})^+ \cdot \overleftrightarrow{D} \cdot \sigma^{\uparrow\downarrow}|^2 \\ & + C_{\text{singlevalley}} |(B_x \sigma_y^{\uparrow\downarrow} + B_y \sigma_x^{\uparrow\downarrow}) + \text{cycl.perm.}|^2 \end{aligned} \quad (11)$$

where R_l and R_t are the longitudinal and transverse components, respectively, in the tensor $\overleftrightarrow{R}$. C_l and C_t are the longitudinal and transverse components, respectively, in the tensor $\overleftrightarrow{C}$. *cycl.perm.* represents terms having the same form of $B_i \sigma_j^{\uparrow\downarrow} + B_j \sigma_i^{\uparrow\downarrow}$, with the index i, j being cyclic permutations of x, y and z .

Further substituting the magnetic field

$$\mathbf{B} = |B|(\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta) \quad (12)$$

and the Pauli matrix elements $\sigma_x^{\uparrow\downarrow} = \cos \theta \cos \phi + i \sin \phi$, $\sigma_y^{\uparrow\downarrow} = \cos \theta \sin \phi - i \cos \phi$, and $\sigma_z^{\uparrow\downarrow} = -\sin \theta$, as well as

$$\overleftrightarrow{D} = \begin{pmatrix} \alpha_0^X \alpha_r^X & 0 & 0 \\ 0 & \alpha_0^Y \alpha_r^Y & 0 \\ 0 & 0 & \alpha_0^Z \alpha_r^Z \end{pmatrix} \quad (13)$$

allows us to represent the $1/T_1$ anisotropy with the solid lines of Fig. 1e and Fig. 2a-b of the main text. For these fits, we have assumed the strength of valley repopulation and single valley effect ($C_{\text{valleyrep}} = 18.507 \text{ 1/s}(\mu\text{eV})^2$, $C_{\text{singlevalley}} = 0.0338 \text{ 1/sT}^2$), as well as $g_l - g_t = 0.001$, as previously measured [10]. We also assumed bulk-donor valley weights $\alpha_0 = (1, 1, 1)$ for the $1s(A_1)$ state and $\alpha_r = (1, 0, 1)$ for the $1s(E)$ state [17], and $k_{0r} = 10^3 \text{ 1/m}$, the matrix element of $\mathbf{k} = -i\nabla + \frac{e}{\hbar} \mathbf{A}$ with respect to the $1s(A_1)$ and $1s(E)$ states. The latter was obtained from NEMO tight-binding calculations [18].

This only leaves the relative electric field contributions ($E_x : E_y : E_z$) and the coupling constants of Rashba ($R = R_l - R_t$) and H_{EB} ($C = C_l - C_t$) as fitting parameters. We note that we can only fit the ratio and not the absolute values of the electric field due to the interdependency of R and C with the electric field strength. A least absolute error fit to the experimental data points gives the coefficients $R = 9.05 \times 10^{-19} e \text{ m}^2$ and $C = 5.86 \times 10^{-14} e \text{ m/T}$ for the out-of-plane rotation, as stated in the manuscript, and a ratio of $(E_x : E_y : E_z) = (-0.26 : 1 : -0.20)$, showing the dominating y component of the electric field. Another data set of the relaxation rate was given by the in-plane rotation measurements, where the gate applied bias is different from the out-of-plane measurements (see main text for details). Following the same procedure, the fitting results show $R = 9.53 \times 10^{-19} e \text{ m}^2$ and $C = 6.01 \times 10^{-14} e \text{ m/T}$. If we compare the fitting results of R and C from the out-of-plane measurements to the in-plane measurements, we see that the values are consistent with each other.

V. INDEPENDENT CHECK OF THE FITTED PARAMETER VALUES

An independent check of the parameters fitted to the experimental $1/T_1$ data points, electrostatic and atomistic modeling were performed. Detailed COMSOL modeling showed a dominant E_y field (see the method section of the main text), justifying the electric field ratio fitted. To confirm the strength of the Rashba coupling, atomistic tight-binding calculations of the donor g -factor was performed as a function of B -field angle. By fitting to the g -factor tight-binding calculation results with an effective mass expression based on valley repopulation and Rashba effect, the Rashba strength (R) was extracted for the device electric field as shown in the next section. The difference between the Rashba strength obtained from these tight-binding calculations ($R = 1.47 \times 10^{-19} \text{ e m}^2$) and the value obtained from the $1/T_1$ fit ($R = 9.05 \times 10^{-19} \text{ e m}^2$) was found to be within one order of magnitude. Lastly, to obtain the strength of the H_{EB} SOC (C), the ratio of the Rashba strength and the H_{EB} strength ($C/R = 5.7 \times 10^4 \text{ m/Vs}$) was derived using the $k \cdot p$ theory in the subsequent section and found a good correspondence with the values obtained from the fit ($C/R = 6.5 \times 10^4 \text{ m/Vs}$). Therefore, an independent justification of the T_1 anisotropy fitting results was provided. Finally, since this is the first time the H_{EB} SOC has been reported in donor systems, we checked the effect of H_{EB} SOC on the donor g -factor and found the g -factor is increased by only 0.0001% including the H_{EB} SOC (with $E = 3 \text{ MV/m}$ and $B = 2 \text{ T}$), explaining why it might not have been observed in the Stark shift experiments of donor spin resonance [19].

Strength of the Rashba SOC

We can obtain an independent estimate for the Rashba SOC strength R of donors in Si by calculating the g -factor anisotropy using the nanoelectronic modeling tool NEMO. NEMO uses a 20 band spin-resolved tight binding method with $\text{sp}^3\text{d}^5\text{s}^*$ atomic orbital model, including nearest-neighbor and spin-orbit interactions to represent the Hamiltonian of a nanoelectronic device, such as a donor in silicon [20, 21]. To calculate the electronic states in donor-based systems, a Coulomb potential of a positive charge screened by the dielectric constant of Si is added to the Hamiltonian. At the donor site, a cut-off potential is adjusted to yield the measured binding energy of a single donor [18]. The model reproduces known experiments [5, 11, 18, 22–25].

While $1/T_1$ depends on the off-diagonal coupling in the spin Hamiltonian between ground and excited states, the g -factor comes from the diagonal term [7]

$$\langle \psi | \mu_B \mathbf{B} \cdot \overleftrightarrow{g} \cdot \sigma | \psi \rangle = \mu_B \mathbf{B} \cdot (g_t \overleftrightarrow{I} + \frac{1}{3}(g_l - g_t) \overleftrightarrow{D}) \cdot \sigma, \quad (14)$$

$$\overleftrightarrow{D} = 3 \begin{pmatrix} (\alpha^X)^2 & 0 & 0 \\ 0 & (\alpha^Y)^2 & 0 \\ 0 & 0 & (\alpha^Z)^2 \end{pmatrix} \quad (15)$$

Here, $\overleftrightarrow{I}$ is the identity matrix and $\overleftrightarrow{D}$ contains the valley weights α^i with valley index $i = X, Y, Z$. For the A_1 donor ground state, the valley weights are equal in all six valleys, hence $\overleftrightarrow{D}$ is simply the identity matrix. The g -factor of the A_1 ground state is therefore $g = \frac{2}{3}g_t + \frac{1}{3}g_l$, independent of the alignment of the external magnetic field. However, for higher valley states (see inset of Fig. S4a), $\overleftrightarrow{D}$ is different. As a result different magnetic field directions can result in different Zeeman energies. For example, for an electric field pointing along y , the degeneracy of the E state manifold is broken, whereby the lowest state of this manifold has valley weights $\alpha^X = 1, \alpha^Y = 0, \alpha^Z = -1$. Consequently, for this state,

$$\overleftrightarrow{D} = 3 \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (16)$$

and

$$\langle \psi | \mu_B \mathbf{B} \cdot \overleftrightarrow{g} \cdot \sigma | \psi \rangle = g_t \mu_B \mathbf{B} \cdot \sigma + \frac{1}{3}(g_l - g_t) \mu_B (B_x \sigma_x + B_z \sigma_z), \quad (17)$$

The second term on the right hand side of Eqn. 17 adds an anisotropic term to the Zeeman energy. This term is larger if B_{ext} is aligned along $[001]$ ($\theta = 0^\circ$) compared to $B_{\text{ext}} \parallel [110]$ ($\theta = 90^\circ$), since the y component does not contribute. At $\theta = 0^\circ$ the total g -factor is therefore larger than at $\theta = 90^\circ$, as shown by the line marked as “E-1” in Fig. S4a. In general, the g -factor anisotropy is therefore different for different excited states, depending on the valley weights. To demonstrate this, Fig. S4a shows the A_1 , T_2 and E states’ g -factor anisotropy calculated from NEMO under an electric field $E_y = 3 \text{ MV/m}$.

The spin Hamiltonian in the presence of an electric field is given by

$$H = \mu_B \mathbf{B} \cdot \overleftrightarrow{g} \cdot \sigma + (\mathbf{k} \times \mathbf{E}) \cdot \overleftrightarrow{R} \cdot \sigma \quad (18)$$

where the first term is the Zeeman term considering the bulk spin-orbit coupling and the second term is the Rashba SOC. The corresponding Zeeman energy of a donor state ψ_r is given by

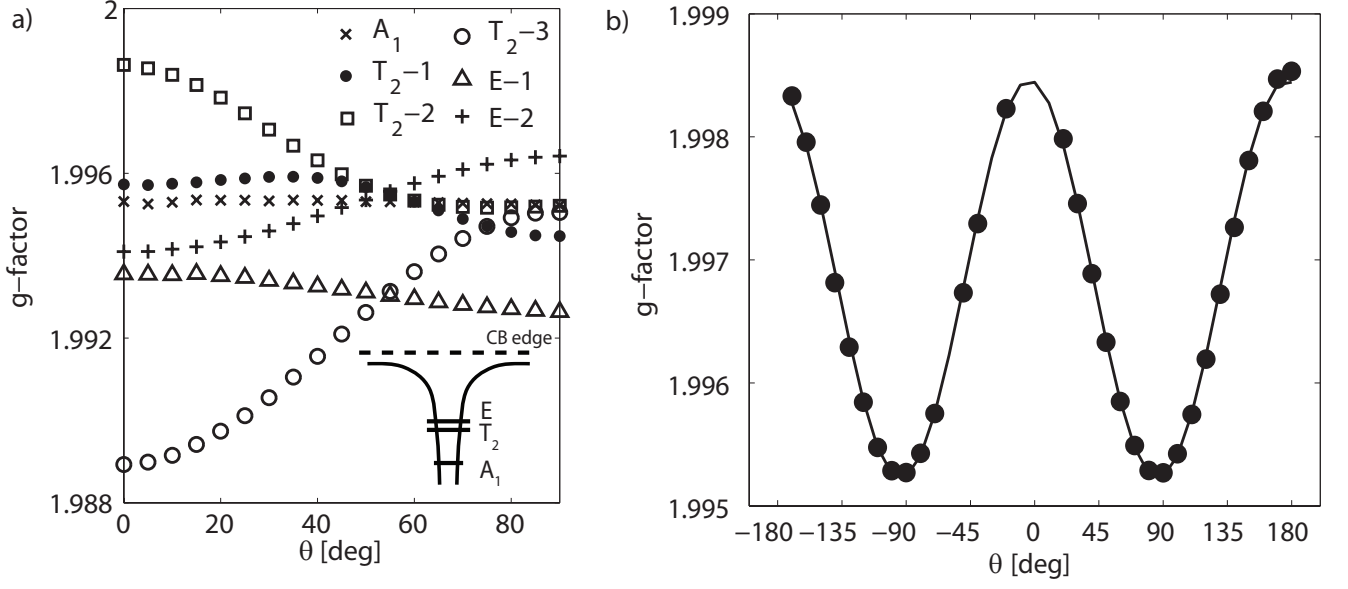


Figure S4. **Extraction of the magnitude of the Rashba coefficient by calculating the g -factor anisotropy of the valley-orbit states from NEMO.** **a**, The g -factor anisotropy of the donor valley-orbit states from NEMO with $E_y = 3\text{ MV/m}$. The inset shows a schematic of the donor valley-orbit states. **b**, The g -factor anisotropy of the low-lying donor E state with the electrostatic potential profile obtained from COMSOL. The dots represent the simulation results from NEMO, and the line represents theoretical fits from Eqn. 19.

$$\langle \psi_r | H | \psi_r \rangle = \frac{1}{2} \mu_B \mathbf{B} \cdot (g_t \vec{I} + \frac{1}{3} (g_l - g_t) \vec{D}) \cdot \sigma + (\mathbf{k}_{rr} \times \mathbf{E}) \cdot (R_t \vec{I} + \frac{1}{3} (R_l - R_t) \vec{D}) \cdot \sigma \quad (19)$$

The total g -factor is defined as the Zeeman energy divided by $\mu_B B$.

To extract the strength of Rashba SOC ($R = R_l - R_t$) from the spin relaxation rate measurements, the electrostatic potential profile of the device was obtained from finite-element modeling of the Poisson equation in COMSOL, and was used as an input to NEMO. The spin resolved tight-binding Hamiltonian of the donor in the presence of this electrostatic potential is then solved in NEMO to obtain the donor eigen-energies and the corresponding g -factors.

Fig. S4b shows the g -factor anisotropy of the low-lying E state under the COMSOL electrostatic potential. With this, we fit Eqn. 19 to our NEMO data and obtain the magnitude of each term in Eqn. 18. The electric fields at the donor site obtained from the COMSOL simulations are $E_x = 1.25\text{ MV/m}$, $E_y = 5.33\text{ MV/m}$, and $E_z = 0.26\text{ MV/m}$. Substituting $\mathbf{k}_{rr} = -i \langle \psi_r | \nabla | \psi_r \rangle + \frac{e}{\hbar} \langle \psi_r | \mathbf{A} | \psi_r \rangle$, the second term can be evaluated using the symmetric gauge ($A_x, A_y, A_z = \frac{1}{2}(B_y z - B_z y, B_z x - B_x z, B_x y - B_y x)$). Here, we approximate the dipole moments $\langle \psi_r | x | \psi_r \rangle \sim \langle \psi_r | y | \psi_r \rangle \sim \langle \psi_r | z | \psi_r \rangle \sim 10^{-10}\text{ m}$ under the electric field distribution [18]. Finally, from the fitting results we obtain

$$R = 1.47 \times 10^{-19} e m^2 \quad (20)$$

This number is only a factor of six different from the value obtained by fitting the $1/T_1$ anisotropy data from Fig. 2 in the main text and Eqn. 9 ($R = 9.05 \times 10^{-19} e m^2$). Compared to other systems such as quantum dots [26], where the Rashba parameter is known to be around $\alpha = RE \sim 10^{-15} \text{ eV m}$ for an electric field of a few MV/m, we have $\alpha \sim 10^{-13} \text{ eV m}$ in donors under similar magnitude of the electric field. The large difference of two orders of magnitude in the Rashba parameter is mainly due to the strong confinement of the donor wavefunction compared to the weaker confinement in quantum dot systems which the electron wavefunction spreads out more [26]. In general, the magnitude of the Rashba SOC is determined by the Rashba parameter α multiplied by the matrix element of the operator k . The Rashba parameter in quantum dots, as shown above, is two-orders of magnitude smaller than in donors. However, the matrix element of the operator k for quantum dots, with respect to the ground states, for example, is usually two to three orders of magnitude larger than in donors (consider dots having radius of 10-100nm). As a result, the Rashba SOC contribution in Zeeman energy is usually larger in quantum dots than in donors.

In Eqn. 10, H_{EB} is neglected because it always produces a small effective magnetic field which is perpendic-

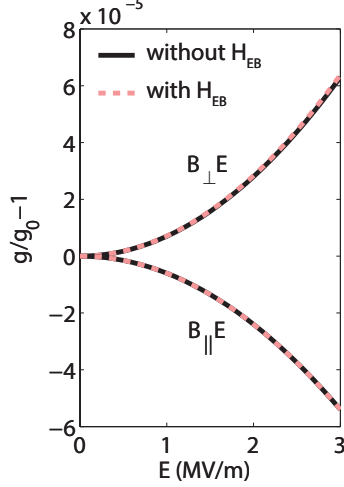


Figure S5. **Comparison of the g -factor Stark effect with and without H_{EB} .** The data without the H_{EB} are taken from the calculations of Ref. [11]. The magnitude of the magnetic field is 2 T. When E points along y and E and B are parallel, H_{EB} has no effect on the donor g -factor. When E and B are perpendicular, the g -factor increases slightly (0.0001% at $E = 3\text{MV/m}$) due to H_{EB} .

ular to the external magnetic field, and results in negligible corrections to the total g -factor (see Fig. S5).

Strength of the H_{EB} SOC

Having obtained the Rashba strength (R), the remaining parameter is the H_{EB} strength (C). Below we derive the relationship of R and C and compare the ratio C/R with the fitting results as an independent check. From perturbation theory,

$$H_{Rashba} = (\mathbf{k} \times \mathbf{E}) \cdot \overleftrightarrow{R} \cdot \sigma \quad (21)$$

where

$$\overleftrightarrow{R} = \frac{e\hbar^2}{2im_0^2} \sum_{n,\mu \neq \alpha} \frac{(\mathbf{p}_{\alpha\mu} \times \mathbf{p}_{\mu n}) \otimes \mathbf{h}_{n\alpha}}{(E_\alpha^0 - E_{\mu,n}^0)^3} \quad (22)$$

h is defined by

$$H_{so} = \frac{\hbar}{2m_0^2 c^2} (\nabla V \times \mathbf{p}) \cdot \sigma \equiv \mathbf{h} \cdot \sigma. \quad (23)$$

and α represents the Si conduction band, n, m sums over all other bands. Similarly

$$H_{EB} = \begin{pmatrix} E_y B_z + E_z B_y \\ E_z B_x + E_x B_z \\ E_x B_y + E_y B_x \end{pmatrix}^T \cdot \mathbf{C} \cdot \sigma \quad (24)$$

where

$$\overleftrightarrow{C} = \frac{e^2 \hbar^2}{2im_0^3} \sum_{n,\mu,\nu \neq \alpha} \frac{((\mathbf{p}_{\alpha\mu} \times \mathbf{p}_{\mu\nu}) \times \mathbf{p}_{\nu n}) \otimes \mathbf{h}_{n\alpha}}{(E_\alpha^0 - E_{n,\mu,\nu}^0)^4} \quad (25)$$

therefore

$$\frac{C}{R} = \frac{e}{m_0} \frac{p_{\mu\nu}}{E_{\alpha\mu}} \quad (26)$$

$\frac{\hbar}{m_0} p_{\mu\nu} = 1.5 \times 10^{-10}$ eV m is the momentum matrix element between the silicon valence bands μ and ν , evaluated at the conduction band minimum k_0 . $E_{\alpha\mu} \sim 4$ eV is the energy difference between the conduction band minimum and the valence band at k_0 [8]. Substituting these values to Eqn. 26, the resulting C/R is 5.7×10^4 m/Vs. This matches well with the ratio obtained from the fit ($C/R = 6.5 \times 10^4$ m/Vs.). The observed relationship between the strength of Rashba and H_{EB} SOC is therefore well-explained by the $k \cdot p$ theory.

Effect of H_{EB} SOC on the donor g -factor

As we have shown, valley repopulation with H_{EB} SOC taken into account dominates spin-relaxation in the presence of electric fields. Finally for completeness we consider how H_{EB} affects the electron's g -factor.

Fig. S5 compares the donor ground state g -factor under electric fields with and without the H_{EB} , where data without H_{EB} was taken from Ref. [11]. We see that H_{EB} has a negligible effect on the g -factor under electric fields below 3MV/m ($< 0.0001\%$ change) which explains why it may not have been observed in previous measurements of the donor g -factor [11, 19].

VI. CONCLUSION

To conclude, the experimentally observed T_1 anisotropy can be fully explained by the H_{EB} theory. With atomistic calculation, electrostatic modelling and the $k \cdot p$ theory, we give a comprehensive quantitative description of the observed data.

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- [1] Elzerman, J. M. *et al.* Single-shot read-out of an individual electron spin in a quantum dot. *Nature* **430**, 431–435 (2004).
- [2] Morello, A. *et al.* Single-shot readout of an electron spin in silicon. *Nature* **467**, 687–691 (2010).
- [3] Büch, H., Mahapatra, S., Rahman, R., Morello, A. & Simmons, M. Y. Spin readout and addressability of phosphorus-donor clusters in silicon. *Nat Comm* **4**, 2017 (2013).
- [4] Watson, T. F., Weber, B., House, M. G., Büch, H. & Simmons, M. Y. High-fidelity rapid initialization and read-out of an electron spin via the single donor D^- charge state. *Physical Review Letters* **115**, 166806 (2015).

- [5] Fuechsle, M. *et al.* A single-atom transistor. *Nature Nanotechnology* **7**, 242–246 (2012).
- [6] Watson, T. F. *et al.* Transport in asymmetrically coupled donor-based silicon triple quantum dots. *Nano Letters* **14**, 1830–1835 (2014).
- [7] Hasegawa, H. Spin-lattice relaxation of shallow donor states in ge and si through a direct phonon process. *Physical Review* **118**, 1523–1534 (1960).
- [8] Roth, L. M. g factor and donor spin-lattice relaxation for electrons in germanium and silicon. *Physical Review* **118**, 1534–1540 (1960).
- [9] Roth, L. *Massachusetts Institute of Technology Lincoln Laboratory Reports* (1960).
- [10] Wilson, D. K. & Feher, G. Electron spin resonance experiments on donors in silicon. iii. investigation of excited states by the application of uniaxial stress and their importance in relaxation processes. *Physical Review* **124**, 1068–1083 (1961).
- [11] Rahman, R. *et al.* Gate-induced g -factor control and dimensional transition for donors in multivalley semiconductors. *Phys. Rev. B* **80**, 155301 (2009).
- [12] Winkler, R. *Spin-Orbit Coupling Effects in Two-Dimensional Electron and Hole Systems* (Springer Verlag, 2003).
- [13] Tahan, C. & Joynt, R. Relaxation of excited spin, orbital, and valley qubit states in ideal silicon quantum dots. *Phys. Rev. B* **89**, 075302 (2014).
- [14] Tahan, C., Friesen, M. & Joynt, R. Decoherence of electron spin qubits in si-based quantum computers. *Physical Review B* **66**, 035314 (2002).
- [15] Bir, G., Butkov, E. & Pikus, G. Spin and combined resonance on acceptor centres in ge and si type crystals—i: Paramagnetic resonance in strained and unstrained crystals. *Journal of Physics and Chemistry of Solids* **24**, 1467 – 1474 (1963).
- [16] Huang, P. & Hu, X. Electron spin relaxation due to charge noise. *Physical Review B* **89**, 195302 (2014).
- [17] Kohn, W. & Luttinger, J. M. Theory of donor states in silicon. *Physical Review* **98**, 915 (1955).
- [18] Rahman, R. *et al.* High precision quantum control of single donor spins in silicon. *Physical Review Letters* **99**, 036403 (2007).
- [19] Sigillito, A. J., Tyryshkin, A. M. & Lyon, S. A. Anisotropic stark effect and electric-field noise suppression for phosphorus donor qubits in silicon. *Phys. Rev. Lett.* **114**, 217601 (2015).
- [20] Klimeck, G. *et al.* Atomistic simulation of realistically sized nanodevices using NEMO 3-D – Part I: Models and benchmarks. *IEEE Transactions on Electron Devices* **54**, 2079–2089 (2007).
- [21] Slater, J. C. & Koster, G. F. Simplified lcao method for the periodic potential problem. *Phys. Rev.* **94**, 1498–1524 (1954).
- [22] Rahman, R. *et al.* Orbital stark effect and quantum confinement transition of donors in silicon. *Physical Review B* **80**, 165314 (2009).
- [23] Weber, B. *et al.* Ohm’s law survives to the atomic scale. *Science* **335**, 64–67 (2012).
- [24] Weber, B. *et al.* Spin blockade and exchange in coulomb-confined silicon double quantum dots. *Nature Nanotechnology* **9**, 430–435 (2014).
- [25] Salfi, J. *et al.* Spatially resolving valley quantum interference of a donor in silicon. *Nat Mater* **13**, 605–610 (2014).
- [26] Nestoklon, M. O., Ivchenko, E. L., Jancu, J.-M. & Voisin, P. Electric field effect on electron spin splitting in SiGe/Si quantum wells. *Phys. Rev. B* **77**, 155328 (2008).