Giant anisotropy of spin relaxation and spin-valley mixing in a silicon quantum dot

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Abstract

It is well known that for Si quantum dots (QDs), at a certain magnetic field that is commonly referred to as the “hot spot”, the electron spin relaxation rate ($T_1^{-1}$) can be drastically enhanced due to strong spin-valley mixing. Here, we experimentally find that with a valley splitting of ~78 µeV, this “hot spot” spin relaxation can be suppressed by more than 2 orders of magnitude when the in-plane magnetic field is oriented at an optimal angle, about 9° from the [100] sample plane. This directional anisotropy exhibits a sinusoidal modulation with a 180° periodicity. We explain the magnitude and phase of this modulation using a model that accounts for both spin-valley mixing and intravalley spin-orbit mixing. The generality of this phenomenon is also confirmed by tuning the electric field and the valley splitting up to ~268 µeV.
Main text

Single-spin qubits in Si quantum dots (QDs) are considered one of the most promising contenders for large scale quantum computation [1-3]. In silicon, the relatively weak spin-orbit interaction (SOI) and the existence of an abundant spin-zero isotope allow the electron spin to preserve its quantum state for exceptionally long times, leading to a spin relaxation time ($T_1$) over hundreds of milliseconds [4-6] and a spin coherence time ($T_2$) over tens of microseconds [7,8]. However, adverse effects from an imperfect substrate may weaken some of these advantages [2]. For spin relaxation, the relatively small energy splitting of the two lowest orbital states (namely, the valleys) caused by interface disorder [9-12] introduces a spin relaxation “hot spot” when its magnitude $E_{VS}$ matches the Zeeman energy $E_Z$ [13]. As a result, spin relaxation rate can be enhanced to $10^3$ to $10^6$ s$^{-1}$ [6,14-16], depending on the noise environment in contact. To mitigate such effects, it is crucial to better understand and control the interactions between the spin and valley degrees of freedom in silicon.

Over the past decade, spin relaxation in Si QDs has been investigated both experimentally [4-6,14-17] and theoretically [13,18,19]. It was found that electrical noise via SOI plays an important role in determining spin relaxation in silicon. For magnetic fields near the spin relaxation “hot spot”, the relaxation process is dominated by the SOI with valley states (spin-valley mixing), while for magnetic fields away from the “hot spot”, especially higher fields, $T_1$ is dominated by the intravalley SOI with higher orbital states (intravalley spin-orbit mixing). The effect of SOI on spin relaxation can be viewed as a result of an effective spin-orbit magnetic field $B_{SO}$. A finite angle between $B_{SO}$ and the external magnetic field $B_{ext}$ leads to mixing of spin eigenstates [20,21], allowing electrical noises to induce spin transitions between the excited and ground states. Within this physical picture, spin mixing would vary as the angle between $B_{SO}$ and $B_{ext}$ is changed. Therefore, $T_1^{-1}$ should be anisotropic with respect to the external magnetic field direction.

Previous studies have revealed an anisotropic $T_1^{-1}$ in GaAs QDs [22,23] and a
tunable SOI in silicon using the magnetic field direction [24,25], but so far, an anisotropic $T_{1}^{-1}$ in Si QDs has not been investigated. Indeed, $T_{1}^{-1}$ anisotropy could help suppress the fast relaxation rate from interface disorder and environment noise. Furthermore, it is also a probe into the anisotropy of both spin-valley mixing and intravalley spin-orbit mixing.

Here, we investigate extensively the spin relaxation anisotropy near the “hot spot” in a Si metal-oxide semiconductor (MOS) QD. We find that the variation in $T_{1}^{-1}$ can be as large as 2 orders of magnitude for a certain range of magnetic fields, but is significantly suppressed for other magnetic fields. Based on a model of multiple relaxation channels and a modified picture of the effective spin-orbit magnetic field, we explain our observations by identifying the limiting mechanisms of spin-valley mixing and intravalley spin-orbit mixing. We have also tuned the gate voltage to examine the effect of interface electric field on the anisotropy and find that this phenomenon is quite general, with only somewhat changed parameters. Overall, our results should provide useful guidance for future research on spin-valley mixing and spin control experiments.

The experiment is carried out in a Si-MOS QD device [Fig. 1(a)] fabricated from an 8-inch natural silicon wafer grown by the float zone (FZ) method. Four layers of overlapping aluminum gates with insulating oxide in between are employed to laterally confine the QDs [26] (see Supplementary Material, Sec. 1). During the experiment, gates T, SB1 and SB2, are used to define a single electron transistor (SET) to monitor the charge state of the QD. By differentiating the SET current $I_S$ with respect to gate voltages $V_P$ and $V_{B1}$, a charge stability diagram can be obtained [Fig. 1(b)]. Here we use (NL, NR) to refer to the number of electrons in the dot under gates P and B1, respectively, and we perform the spin relaxation measurements near the (0, 0)-(1, 0) charge transition far detuned from the interdot transition (0, 1)-(1, 0), which allows us to treat it as an isolated QD. The orientation of the QD gate pattern with respect to the main crystallographic directions is also shown in Fig. 1(a) and we apply an in-plane magnetic field at an angle $\phi$ from [100] direction. For the convenience of discussion, we also define [110] and [$\bar{1}$10] to be the x and y axes,
respectively.

To measure the spin relaxation time $T_1$, we apply to gate P the well-known three-step pulse sequence first implemented by Elzerman et al. [27], as shown by points E (empty), R (read) and W (wait) in Fig. 1 (b): first, at point W an electron is injected into the QD with a random spin state and after a time $t_{\text{wait}}$, the spin state is read out via spin-to-charge conversion by pulsing to point R, finally, the QD is emptied at point E. By measuring the spin-up probability as a function of $t_{\text{wait}}$ and fitting the data with an exponential decay, we can extract the value of $T_1$. Some examples of the exponential decays of the normalized spin-up probability $p^\uparrow$ from the experiments can be seen in Fig. 1(c), showing a striking variation in $T_1$ upon rotating the magnetic field orientation. The experimental details of the $T_1$ measurements and device parameter extraction are described in Supplementary Material, Sec. 2 and 3.

The measured $T_1^{-1}$ as a function of the magnetic field oriented along the $y$ axis ($\phi = 117^\circ$) is presented in Fig. 2(a), showing a typical spin relaxation “hot spot” with $E_{\text{VS}} = 78.2 \pm 0.6 \mu$eV. By rotating the in-plane magnetic field orientation over the whole $360^\circ$ range with a constant strength of 0.8 T and 1.5 T, we observe a sinusoidal modulation of the spin relaxation rate with a $180^\circ$ periodicity. Interestingly, as shown in Fig. 2(b), while the data for the two different magnetic field strengths show a nearly common minima angle of $9^\circ$ with respect to the $[100]$ plane, the variation ranges are significantly different: for 0.8 T, $T_1^{-1}$ varies by more than 2 orders of magnitude, which is approximately 1 order of magnitude larger than that in GaAs QDs, while for 1.5 T, the variation range decreases to only six times.

To understand these distinctive behaviors of the $T_1^{-1}$ anisotropy, we first identify different origins of spin relaxation in silicon [13,14]. The expression for $T_1^{-1}$ can be written in the form

$$T_1^{-1} = \Gamma_{J,SV} + \Gamma_{\text{ph},SV} + \Gamma_{J,SO} + \Gamma_{\text{ph},SO} + \Gamma_{\text{const}},$$

where subscripts “SV” and “SO” denote spin-valley mixing and intravalley spin-orbit mixing, while subscript “J” or “ph” indicates that the type of electrical noise
facilitating spin relaxation is Johnson noise or phonon noise, which leads to a different Zeeman energy dependence for the corresponding relaxation rates. Finally, $\Gamma_{\text{const}}$ describes a relaxation channel that is independent of (or at least insensitive to) the external magnetic field. By including all the major contributions to spin relaxation, we can fit the experimental data really well, and can identify the dominant relaxation channel at different field ranges, as illustrated in Fig. 2(a). In general, spin-valley mixing and intravalley spin-orbit mixing dominate spin relaxation for $B_{\text{ext}} < 1.5$ T and $B_{\text{ext}} > 1.5$ T, respectively, and $\Gamma_{\text{const}}$ is negligibly small for most external fields ($B_{\text{ext}} > 0.4$ T). More specifically, for $1.5$ T $< B_{\text{ext}} < 3$ T, $\Gamma_{J,SO}$ is much greater than $\Gamma_{\text{ph,SO}}$. Therefore, the giant $T_1^{-1}$ anisotropy at $B_{\text{ext}} = 0.8$ T is probably due to anisotropic spin-valley mixing, which is largely suppressed by the fast increase in $\Gamma_{J,SO}$ at $B_{\text{ext}} = 1.5$ T. Regarding the anisotropy of $\Gamma_{J,SO}$, the nearly unchanged modulation phase from 0.8 T to 1.5 T suggests that either it has the same modulation phase as $\Gamma_{J,SV}$ and $\Gamma_{\text{ph,SV}}$ or that it is negligible.

With the anisotropy of spin-valley mixing the probable cause for spin relaxation anisotropy at 0.8 T, we now examine this mechanism in more detail. It is useful to reconsider the intuitive picture of the interplay between $B_{SO}$ and $B_{\text{ext}}$ [20,21]. As shown in Fig. 3(a), the presence of $B_{SO}$ causes the spin to precess around an axis different from that of $B_{\text{ext}}$, creating a channel for the spin to relax. If $B_{SO}$ is a real magnetic field, this spin-mixing effect would be maximum when $B_{SO} \perp B_{\text{ext}}$ and is zero when $B_{SO} \parallel B_{\text{ext}}$. As a result, the extrema position should be determined by the direction of $B_{SO}$ and there are two opportunities in the whole rotation range for $B_{\text{ext}}$ to be parallel or perpendicular to $B_{SO}$, leading to a modulation cycle of 180°, which is consistent with the experimental results. However, within this simple geometric picture spin relaxation should be completely suppressed when the two fields are in parallel, leading to an infinite degree of anisotropy in $T_1^{-1}$, which is obviously not what we observed. To address this issue, we revisit the inter-valley spin-orbit Hamiltonian, from which $B_{SO}$ for spin-valley mixing can be expressed as (see also Supplementary Material, Sec. 6) [6,13]
Here, \( \gamma \) is the gyromagnetic ratio, \( \alpha_\pm = \beta \pm \alpha \) are the SOI constants from the Dresselhaus SOI (\( \beta \)) and Rashba SOI (\( \alpha \)), and \( r_y^{-}\)(\( r_x^{-}\)) represents the intervalley transition element along the \( y \)(\( x \)) axis. Since \( r_y^{-}\) and \( r_x^{-}\) are naturally complex numbers [6,10,13], the effective spin-orbit magnetic fields are also complex. To enhance the effective field picture above, the complex nature of \( B_{sO} \) should be included. Here we assume that \( B_{sO,x} \) is fully real and \( B_{sO,y} \) is complex with a phase \( e^{i\theta} = \frac{r_y^{-}/r_x^{-}}{|r_y^{-}|/|r_x^{-}|} \), the total spin-orbit field \( B_{sO} \) can then be represented by a vector in three-dimensional space with an extra axis referring to the imaginary part of \( B_{sO,y} \) [see Fig. 3(b)]. Notice the angle between \( B_{sO,y} \) and the \( y \) axis just corresponds to the phase \( \theta \). If \( \theta \) is finite, \( B_{\text{ext}} \) in the two-dimensional plane would never be parallel to \( B_{sO} \), resulting in a residual SOI induced \( T_1^{-1} \) when \( B_{\text{ext}} \) is along the minimum angle. Conversely, if the angle \( \theta \) can be tuned, it would enable control of the magnitude of the spin mixing and relaxation anisotropy. Based on the parameters extracted from Fig. 2(a), a best fit for the data in Fig. 2(b) yields \( \theta = 0.14 \) and \( R = \alpha_- |r_y^{-}|/\alpha_+ |r_x^{-}| = -1.35 \). Here, \( R \) is chosen to be consistent with the extrema position (see also Supplementary Material, Sec. 7). The nonzero \( \theta \) leads to a reduced anisotropy of spin-valley mixing and causes a nonvanishing “hot spot” when rotating the magnetic field orientation. This is proven by the calculated “hot spot” over the whole 360° range in the inset of Fig. 2(c) and further confirmed by the experimental results reported in Fig. 4(b).

To identify the limiting mechanisms at different magnetic fields for the spin relaxation anisotropy, we numerically calculate the anisotropy magnitude \( T_{1,\text{max}}^{-1}(\phi)/T_{1,\text{min}}^{-1}(\phi) \). As shown in Fig. 2(c), the variation range is mostly limited by \( \theta \) from spin-valley mixing for \( B_{\text{ext}} < 0.85 \) T, and by the residual relaxation rate \( \Gamma_{JSO} \) for \( B_{\text{ext}} > 0.85 \) T. These conclusions are also illustrated in the inset of Fig. 2(b), if \( \theta \) is set to zero, \( T_{1,\text{min}}^{-1}(\phi) \) is further reduced for \( B_{\text{ext}} = 0.8 \) T, but remains nearly the
same for $B_{\text{ext}} = 1.5$ T. Notice in Fig. 2(c), the limiting mechanism of $\Gamma_{\text{ph,SO}}$ is not considered since its magnitude is much smaller than that of $\Gamma_{\text{SO}}$ for the range of magnetic field.

It is well known that the valley splitting and the valley-dependent SOI constants are dependent on the applied electric field in Si MOS QDs [6,28]. Here we examine how the interface electric field affects $T_{1}^{-1}$ anisotropy via spin-valley mixing. First, as shown in Fig. 4(a), we confirm that the valley splitting in our device increases almost linearly with $V_{p}$ (For the measurement of the valley splitting, see Supplementary Material, Sec. 4). Then we investigate the behavior of $T_{1}^{-1}$ anisotropy with $E_{\text{VS}}$ increased up to $\sim 268 \mu$eV. The measured $T_{1}^{-1}(B_{\text{ext}})$ along the direction of $\phi = 117^\circ$ and $\phi = -178^\circ$ (near the minimum $T_{1}^{-1}$ direction. See Supplementary Material, Sec 5) and the calculated “hot spot” variation by rotating the magnetic field are shown in Fig. 4(b). While the “hot spot” anisotropy magnitude is similar to that in Fig. 2(c), the extrema position is shifted by $\sim 7^\circ$. By choosing $\theta = 0.22$ and $R = -1.1$, which are somewhat increased compared to the fit in Fig. 2(b), the data along the two different directions can be well fitted simultaneously. The rise in $\theta$ can be attributed to the increased anisotropy of the intervalley transition elements that arises from the stronger interface electric field and possible variation in the QD shape [29]. If we suppose that $|r_{y}^{++}|/|r_{x}^{++}|$ remains nearly unchanged with a varying electric field, the slightly larger $R$ in the higher electric field may be an indication of an increased Rashba SOI [28,30]. Further insights into the electrical field effect can be obtained by independently verifying the variation of valley-dependent SOI and inter-valley transition elements. Overall, the increased electric field leads to moderate changes in both the magnitude and the orientation of $T_{1}^{-1}$ anisotropy, but the basic features of the giant $T_{1}^{-1}$ anisotropy remain even though the valley splitting is increased by over 2 times.

In the discussion above, the relative phase of $r_{y}^{++}$ and $r_{x}^{++}$ can be obtained directly by $\theta$, although the absolute value of the SOI strength $\alpha_{-}/\alpha_{+}$ and the intervalley transition matrix elements $r_{y}^{++}/r_{x}^{++}$ cannot be distinguished. To extract
their exact values, more information is needed, such as the physical mechanism of the intervalley transition elements and their dependence on the electric and magnetic fields [10,12,28,31,32]. Nonetheless, the modified picture of a complex $B_{SO}$ mixing the spin eigenstates of $B_{ext}$ helps us determine both the magnitude and orientation of the anisotropic spin-valley mixing, which is a clear indication that $T_1^{-1}$ anisotropy is an effective approach for characterizing spin-valley mixing in silicon.

The large anisotropy of the spin relaxation “hot spot” observed in this work not only helps clarify basic features of spin-valley mixing, but also provides a new method to suppress $T_1^{-1}$, which would in turn allow a larger magnetic field range for high fidelity readout and control of qubits. Such an increased workable field range may specifically inspire experiments in Si/SiGe heterostructure QDs where the valley splitting may be less controllable [14,15]. Additionally, the great modulation of spin-valley mixing creates a new way to optimize qubit performance, especially for qubits driven by spin-orbit coupling [33,34].

In conclusion, we have studied how spin relaxation in silicon depends sensitively on the external field orientation. By rotating an in-plane magnetic field, we find that the spin relaxation rate near the spin-valley “hot spot” can be reduced by more than 2 orders of magnitude. The range of this large variation is found to be controlled both by spin-valley mixing and intravalley spin-orbit mixing. We have also shown that this great anisotropy holds in a larger electric field with slightly varied parameters of spin-valley mixing compared to the significant increase of valley splitting. For future work, the anisotropy of intravalley spin-orbit mixing at much larger magnetic fields could be investigated, which should offer a deeper understanding of the mechanism for SOI with valley and orbital states in silicon.

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**Figure Captions**

**FIG. 1.** (a) Scanning electron microscope image of a device identical to the one measured. The dashed orange circle denotes the position of the quantum dot for the $T_1$ measurements. Inset: the crystallographic directions with respect to the sample. (b) Charge stability diagram of the QDs under gates B1 and P. The pulse sequence for measuring $T_1$ is overlaid on the data. (c) Normalized spin-up fraction as a function of the waiting time $t_{\text{wait}}$ for different angles $\phi$ of the 0.8 T in-plane magnetic field with $E_{VS} = 78.2 \pm 0.6 \mu$eV. The solid lines are exponential fits to the data with the values of $T_1$ (ms) indicated aside.
FIG. 2. (a) Relaxation rates as a function of the magnetic field strength with an in-plane angle of $\phi = 117^\circ$. The fittings include contributions from different relaxation channels obtained through the model discussed in the main text. (b) Angle dependence of the relaxation rate measured with different magnetic field strengths. The red and blue solid lines are fits to the spin relaxation model, while the corresponding shaded areas indicate a 95% confidence interval with a sine fit. Inset: $T_{1,\min}^{-1}(\phi)$ as a function of the fitting parameter $\theta$ for $B_{\text{ext}} = 0.8$ T (red) and $B_{\text{ext}} = 1.5$ T (blue). (c) Anisotropy magnitude as a function of the magnetic field strength under real experimental conditions or with certain assumptions. The shaded areas indicate the amount of anisotropy suppressed by corresponding mechanism. Inset: numerical simulation of the spin relaxation “hot spot” as a function of the external magnetic field angle.
FIG. 3. (a) Illustration of the intuitive classical picture of the interaction between the effective spin-orbit magnetic field $B_{SO}$ and the external magnetic field $B_{ext}$. The dashed circle shows the rotation of $B_{ext}$. (b) Modified intuitive picture of the interaction between $B_{SO}$ and $B_{ext}$.

FIG. 4. (a) Valley splitting $E_{VS}$ as a function of the gate voltage $V_p$. A linear fit shows a tunability of 0.667 meV V$^{-1}$. The deviation from the linear fit at small $V_p$ perhaps results from an interface localized interaction [28]. (b) Relaxation rates as a function of the external magnetic field along different directions and the calculated anisotropy magnitude with experimental parameters. The solid lines are simultaneously fitted to the data using the model explained in the main text with $E_{VS} \sim 268 \mu$eV. Inset: numerical simulation of the spin relaxation “hot spot” as a function of the orientation of the external magnetic field.