Singlet-Triplet Relaxation in Two-electron Silicon Quantum Dots

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We investigate the singlet-triplet relaxation process of a two electron silicon quantum dot. In the absence of a perpendicular magnetic field, we find that spin-orbit coupling is not the main source of singlet-triplet relaxation. Relaxation in this regime occurs mainly via virtual states and is due to nuclear hyperfine coupling. In the presence of an external magnetic field perpendicular to the plane of the dot, the spin-orbit coupling is important and virtual states are not required. We find that there can be strong anisotropy for different field directions: parallel magnetic field can increase substantially the relaxation time due to Zeeman splitting, but when the magnetic field is applied perpendicular to the plane, the enhancement of the spin-orbit effect shortens the relaxation time. We find the relaxation to be orders of magnitude longer than for GaAs quantum dots, due to weaker hyperfine and spin-orbit effects.

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I. INTRODUCTION

A promising technology for the implementation of quantum computation (QC) involves the storage of quantum information in the spin of electrons in quantum dots (QDs). The key requirement is that the lifetime of the spins is long compared with the time required for the operation of logic gates. This has motivated the development of dots in silicon, where spin-orbit coupling is weak and isotopic enrichment can eliminate hyperfine coupling (HC). Indeed, recent experiments demonstrate the presence of long-lived spin states in silicon QDs. Understanding the processes that relax spins can point to strategies for minimizing relaxation and coherence times, thereby improving coherent control of quantum systems. In the case of electron spins embedded in semiconductor nanostructures, the relaxation properties are strongly affected by the regime of operation. Thus it is important to identify the dominant sources of fluctuations in these systems, the mechanisms by which they couple to the spins, and to analyze the non-equilibrium decay laws in different regimes of external fields. Considerable theoretical work has been performed on lifetimes for single-electron spin flip $T_1$ and dephasing $T_2$ for GaAs and for Si. In GaAs these times have been measured. Single-spin values for $T_1$ of about 0.5 ms at a field of 10 T up to 170 ms at 1.75 T were obtained, while for $T_2$ one finds a value of about 1 $\mu$s. A qubit consisting of the singlet and triplet states of a two-electron system is also a proposal for QC. The singlet-triplet lifetime has been studied in GaAs. In particular, Hanson et al. found $T_1$ for the singlet-triplet transition in a two-electron GaAs dot to be 2.6 ms at B=0.02 T. We shall call this $T_{ST}$. Extensive theoretical work has been done for $T_{ST}$ in GaAs and for Si, and our methods are similar to those found in these references.

In this paper we study the relaxation process for a doubly-occupied Si QD in an excited (triplet) state to the ground (singlet) state, focusing on the computation of $T_{ST}$. Our main motivation is to understand transport through double quantum dots. Thus we are concerned with lateral dots defined by gates in strained silicon quantum wells. Such dots have a two-fold valley degeneracy as well as spin degeneracy, but we will deal here with dots where the valley splitting is large compared with the first orbital excitation energy. We will focus on natural Si with a 4% concentration of $^{29}$Si nuclei, since this is the material on which experiments have been performed, but we comment on isotopically enriched Si below.

We assume the levels to be ordered as shown in Fig. 1. The relevant energy scales are then the exchange, $J = E_J - E_T$, and the difference between the ground singlet and the first triplet, $\varepsilon_{ST} = E_T - E_S$, where the triplet is formed with a higher energy orbital, as depicted in Fig. 1(a). The dominant mechanism available in the absence of an external magnetic field is the hyperfine coupling with nuclei via a virtual state (left arrows of Fig. 1(b)). HC cannot cause a direct $T \rightarrow S$ transition because the nuclei cannot absorb the energy. So the change in energy of the electron spin must be accompanied by the emission of a phonon. The electron-phonon interaction mixes thus different orbital states via a deformation potential in this process, while the spin-flip is provided by the HC. This is the dominant process at zero applied magnetic field. A second relaxation channel is through spin-orbit (SO) coupling. SO coupling mixes different spin states through the Rashba SO coupling. This leads to a non-vanishing matrix element for the phonon-assisted transition between a singlet and a triplet state in the absence of time-reversal symmetry (right of Fig. 1) leading to an increase in the relaxation rate, $\Gamma_{ST}$ as the field is increased. Our aim here is to compute the singlet-triplet relaxation rates due to these two mechanisms as a function of external field.

We outline the method in the following section, and justify the approximations that are made. We then present results and discussion.

II. METHOD AND RESULTS

We consider first the case of low fields. For reasons to be discussed below, we may neglect the spin-orbit coupling and the Hamiltonian is written as $H_0 + \delta H$, where $H_0$ contains the confinement potential of the QD and $\delta H = H_{hc} + H_{ph}$. Here $H_{ph}$ corresponds to the electron-phonon coupling (which...
states for the lateral dimensions orbital and spin degrees of freedom factorize:

\[ |S_0 \rangle, |T \rangle \]

Thus, the interaction is included phenomenologically through a parameter \( J \), the singlet-triplet splitting. Instead, \( \Delta \) is given by deformation potentials so the Hamiltonian reads:

\[ H_{\text{ph}} = \sum_{i} \left[ a_{i} \epsilon e^{-i q \cdot r} + a_{i} \epsilon e^{i q \cdot r} \right] q(\Xi_{i} \delta \hat{e}_{i} \hat{q}_{x} + \Xi_{i} \delta \hat{e}_{i} \hat{q}_{y} + (\Xi_{i} \delta + \Xi_{i}) \delta \hat{q}_{z}), \]

where \( \langle n_{q} | a_{q} | n_{q} \rangle = \sqrt{(h \gamma_{q} / 2M_{c} \omega_{q})} \), \( M_{c} \) is the mass of the unit cell, \( n_{q} \) is the phonon occupation number and \( h \omega_{q} \) is the phonon energy. Here \( \gamma_{q} \) describes the polarisation of the phonon (two transverse and one longitudinal), \( \epsilon \) is the wavevector, and \( \Xi_{i} \) and \( \Xi_{i} \) are the electron-phonon coupling parameters. This is slightly simpler than the corresponding Hamiltonian in GaAs because of the absence of the piezoelectric coupling in (centrosymmetric) Si.

Next, we evaluate the spin-flip matrix element given by \( \langle \Psi_{s}'|H_{\text{ph}}|\Psi_{T} \rangle \), which is provided, at low magnetic fields, by the bath of nuclear spins of the \( ^{29}\text{Si} \) isotope. Accordingly, we consider a contact Hamiltonian,

\[ \hat{H}_{\text{ph}} = \sum_{i,j} \frac{A_{i,ij}}{M_{c}} \epsilon \delta_{s,i} \delta_{T,j} (r_{i} - R_{j}) = A \sum_{i,j} S_{i} I_{j} \delta_{s,i} \delta_{T,j} (r_{i} - R_{j}), \]

where \( S_{i} (I_{j}) \) and \( r_{i} (R_{j}) \) denote the spin and position of the \( i \)th electron (\( j \)th nuclei), and \( \eta \) and \( A \) are hyperfine coupling constants. Inserting eq. (3) and (4) into (2), we get an expression for the singlet-triplet rate:

\[ \Gamma_{ST} = \Gamma_{ph} \times \left( \frac{A}{2J} \right)^{2} \sum_{i,j} \left[ (\Xi_{i} \delta \hat{e}_{i} \hat{r}_{i} - R_{j}) \right] \right] \right)^{2}, \]

The transition rate from \( |\Psi_{T} \rangle \) to \( |\Psi_{S_{0}} \rangle \) is then given by Fermi’s golden rule:

\[ \Gamma_{ST} = \frac{2\pi}{\hbar} \langle \Psi_{S_{0}} | H_{\text{ph}} | \Psi_{T} \rangle \left| \epsilon \right|^{2} \delta(E_{T} - E_{S_{0}}). \]

In this notation, \( |\Psi_{T} \rangle \) denotes the initial state of electron, nuclei and phonons, \( |\Psi_{T} \rangle \equiv |S_{0} \rangle \otimes |n_{i} \rangle \otimes |i_{ph} \rangle \), likewise \( |\Psi_{S_{0}} \rangle \equiv |S_{0} \rangle \otimes |f_{i} \rangle \otimes |f_{ph} \rangle \). The \( av \) subscript indicates that the initial states of the nuclear and phonon systems are averaged over thermal ensembles, and that the final states of these systems are summed over. In this paper we take the temperature to be 100 mK, as this is roughly the temperature at which experiments are done. The chief approximations involved in the calculation are the use of second-order perturbation theory and the truncation of the Hilbert space to just two singlet states and one triplet state. The first approximation is excellent - the rates turn out to be on the order of seconds; at those time scales the Born-Markov approximation implicit in Golden-Rule calculations is surely valid - the time scales in the bath are probably of the order of a time for a phonon to traverse the dot. The validity of the second approximation is less clear - in high-symmetry dots such as we are considering here the phonons do not couple to highly excited states in the dipole approximation, but real dots may be more disordered.

For silicon under compressive stress along [001], the electron interacts with a phonon of momentum \( q \) via deformation potentials so the Hamiltonian reads:

\[ H_{\text{ph}} = \sum_{i} \left[ a_{i} \epsilon e^{-i q \cdot r} + a_{i} \epsilon e^{i q \cdot r} \right] q(\Xi_{i} \delta \hat{e}_{i} \hat{q}_{x} + \Xi_{i} \delta \hat{e}_{i} \hat{q}_{y} + (\Xi_{i} \delta + \Xi_{i}) \delta \hat{q}_{z}), \]

where \( \langle n_{q} | a_{q} | n_{q} \rangle = \sqrt{(h \gamma_{q} / 2M_{c} \omega_{q})} \), \( M_{c} \) is the mass of the unit cell, \( n_{q} \) is the phonon occupation number and \( h \omega_{q} \) is the phonon energy. Here \( \gamma_{q} \) describes the polarisation of the phonon (two transverse and one longitudinal), \( \epsilon \) is the wavevector, and \( \Xi_{i} \) and \( \Xi_{i} \) are the electron-phonon coupling parameters. This is slightly simpler than the corresponding Hamiltonian in GaAs because of the absence of the piezoelectric coupling in (centrosymmetric) Si.

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where \( S_{i} (I_{j}) \) and \( r_{i} (R_{j}) \) denote the spin and position of the \( i \)th electron (\( j \)th nuclei), and \( \eta \) and \( A \) are hyperfine coupling constants. Inserting eq. (3) and (4) into (2), we get an expression for the singlet-triplet rate:

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To evaluate the integral over momenta of (6), we assume an isotropic phonon spectrum, $E_{ph} = \hbar \omega_{ph}$ and a linear dispersion relation, $\omega_{ph} = v_s q$, $v_s$ being the sound velocity of the mode $s$.

The sum over $j$ of (5) can be transformed to an integral by introducing $C_n$ as the volume density of $^{28}$Si nuclei, resulting in a compact expression for triplet-singlet relaxation:

$$\Gamma_{ST} \approx \left( \frac{A}{27} \right)^2 C_n \left( \int d^3 R_j \left[ |\phi(0,0)(R_j)|^2 - |\phi(0,\pm 1)(R_j)|^2 \right]^2 \right) \times \frac{1}{2\hbar S_{\Delta}^2} \left( \frac{J + \varepsilon_{ST}}{h} \right)^5 \sum_{s,s'} \frac{Y_{st}^2}{v_s^2},$$

(7)

where $Y_{st}$ contains the result of the angular integral which depends on the mode $s$ along the coordinate $i$: $Y_{si} = Y_{ti} = 4\pi (\Sigma_t^2/3 + 2 \Sigma_r \Sigma_a/15 + \Sigma_0^2/35)$, $Y_{tl} = 4\pi (\Sigma_r^2/3 + \Sigma_r \Sigma_a/5 + \Sigma_0^2/7)$, $Y_{tv} = Y_{tv} = 4\pi \Sigma_a^2/35$, and $Y_{t2} = 4\pi \Sigma_a^2/70$.

It is important to note that $\Gamma_{ST}$ is proportional to $C_n$, i.e., to the total number of nuclei $N_n$ with which the electrons interact. This is consistent with the simple picture that the relaxation rate is proportional to the mean-square fluctuations in the random hyperfine field. Formulas for spin relaxation rates due to hyperfine coupling that give an apparent proportionality to $N_n^{-1/2}$ are common in the literature, and have given rise to the incorrect notion that some sort of motional narrowing is at work. This is not possible, since the fluctuations in the nuclear spin system are slow. In any case the rate must vanish as $N_n \to 0$. These formulas are correct, but they generally involve other parameters that actually vary with $N_n$.

Fig. 2 represents $T_{ST} = \Gamma_{ST}^{-1}$ obtained as described in Eq. (7) as a function of the level separation, $J = E_{\uparrow} - E_{\downarrow}$ for a given $\varepsilon_{ST}$. In case of Si, $\eta = 186\text{ meV}$ yielding $A \approx 2 \times 10^{-7}\text{ eV nm}^3$. Only about 4% of the nuclei have spin, so $C_n \approx 0.04 \times 8/0.00 \approx 2$ nm$^{-3}$ ($v_0 \approx 0.17$ nm). Other parameters used are: $\Sigma_0 = 9.29$ eV, $\Sigma_1 = -10.7$ eV, $\rho_1 = 2330$ kgm$^{-3}$, $v_1 = 9330$ ms$^{-1}$, $v_2 = 5420$ ms$^{-1}$, $\varepsilon_{ST} = 200$ eV. At small $J$, $T_{ST}$ increases as a function of $J$: the triplet $|T_{\uparrow}\uparrow\downarrow\rangle$ and singlet $|S\rangle$ levels are strongly mixed by the hyperfine interaction and the phonon density of states increases as a function of level separations. In the limit $J \to 0$, the rate is given by phonon emission, which we found to be of the order of $10^{12}$ s$^{-1}$. Thus $T_{ST}$ appears very small as $J \to 0$ in Fig. 2, but it is not zero. At large $J$, spin mixing is lessened because there is an energy denominator and phonon relaxation is then suppressed by spin conservation.

The calculations are obviously consistent with the observed lower bound of $T_{ST} > 15\mu s$ given in Ref. 2. It is expected that more stringent bounds, hopefully also upper bounds, will be available soon.

We now move to the case of finite applied magnetic field $B$. We first take the the field along the growth direction (perpendicular to the 2-DEG): $\vec{B} = \vec{B}_z$. This allows a direct $T \to S$ transition to occur in the presence of a Rashba SO coupling. The Rashba field is a consequence of structural inversion asymmetry in the heterostructure. Note that no bulk inversion asymmetry needs to be considered in a centrosymmetric crystal like Si. The SO Hamiltonian due to the full confining potential for the device considered here where

$$l_{\alpha}/r_{QD} \gg 1, \quad l_{\alpha} \text{ being the Data-Das device length},$$

is then given by

$$\vec{H}^{SO} = \alpha (\vec{\sigma} \times \vec{J}) \zeta.$$  (8)

$H^{SO}$ mixes the spin states $|S\rangle$ and $|T_{\pm}\rangle$, and the orbital wavefunctions as well, so virtual transition to a higher state $|s\rangle$ is no longer needed and we have:

$$\langle S_i^z | \delta H^{SO} | T_{\pm} \rangle \approx \frac{i\langle \phi | H_{ph} | \phi \rangle}{\varepsilon_{ST} + \Delta^z}, \quad \Delta^\pm = \pm g_{\mu_B} B.$$  (9)

Here, the phonon just ensures energy conservation. Notice that $\langle S_i^z | \delta H^{SO} | T \rangle$ is zero if $B = 0$. This ‘Van Vleck cancellation’ has been known for decades, but has been clarified in recent years particularly by performing a spin-dependent unitary transformation in which the first-order term in $\alpha$ is eliminated. In explicit calculations in the original basis the cancellation occurs due to the fact that the admixture of $T^+$ and $T^-$ is equivalent in magnitude but opposite in sign. The key point is that spin-orbit-induced transition rates are always proportional to $B^2$ (or higher powers of $B$ in the case of spin 1/2 dots).

Proceeding as in Eqs. (1) to (5), we find the rate for the direct transition:

$$\Gamma_{ST}^{D} = \Gamma_{ph}^{D} \times \left| \sum_{i=\pm} \langle S_i^z | H^{SO} | T_{\pm} \rangle \right|^2 \langle \varepsilon_{ST} + \Delta^z \rangle.$$  (10)

Note that for $B = 0$ $\Gamma_{ST}^{D}$ vanishes. Since the phonon does not mix different orbitals in zeroth order in the multipole expansion to zeroth order (i.e., $e^{\pm q r} \approx 1$), $\Gamma_{ph}^{D}$ reads now:

$$\Gamma_{ph}^{D} \approx \frac{(n_q + 1)}{2\pi q s (2\pi)^2} \sum_{s} \int d\Omega \int_{0}^{\infty} \frac{dqq^{4}}{\omega_{q}} |\mathcal{E}_d \hat{e}_{x} \hat{q}_x + \mathcal{E}_d \hat{e}_{y} \hat{q}_y + (\mathcal{E}_d + \mathcal{E}_a) \hat{e}_{z} \hat{q}_z|^{2} \delta(\hbar \omega - \varepsilon_{ST})$$  (11)

Inserting (9) into (2) and evaluating (10), we get the spin-orbit-induced relaxation rate, which is quadratic in $B$ and $\alpha$, in agreement with earlier treatments:

$$\Gamma_{ST}^{D} \approx \left( \frac{4m\alpha}{\hbar^2} \right)^2 \langle r^2 \rangle \sum_{i} \frac{Y}{v_i^2}.$$  (11)
with \( \gamma_1 = 4\pi(\tilde{\Xi}_d^2 + \tilde{\Xi}_d^2/2 + \tilde{\Xi}_u^2/5) \) and \( \gamma_2 = 4\pi(\tilde{\Xi}_d^2/5 + 4\tilde{\Xi}_d^2/15 + 2\tilde{\Xi}_u^2/15) \). Here we use \( \alpha \approx 50 \text{ m/s} \), following Ref. 4 and adapting the result to an electric field of \( 10^{-7} \text{V/m} \), an estimate for \( E_z \) for a QD with a 2DEG density of \( \sim 4 \times 10^{11} \text{cm}^{-3} \). (One should note, however, that this value of \( \alpha \) is very uncertain.) Fig 3 (a) contains in black the relaxation in the absence of the Rashba coupling (\( \alpha = 0 \)) and in red the relaxation with the additional \( T^D \) (\( \Gamma^ST + \Gamma^D \)). We plot the results for two values of \( J \). For \( \alpha = 0 \), \( \Gamma^ST \) is a relatively weak function of \( J \), since the hyperfine interaction is not very sensitive to field. At finite \( \alpha \), \( B_\perp \) activates the mixing and \( \Gamma^ST \) decreases rapidly.

If the magnetic field \( B_\parallel \) is parallel to the 2DEG, the only effect is that the spin splitting increases, and larger relaxation times are obtained (Fig. 3b), as these increases, on average, the energy separation \( E_{T^+} - E_{T^\ominus} \) (see diagram). In contrast, the perpendicular magnetic field \( B_\perp \) decreases the relaxation time, as we have seen. This anisotropy in applied field of \( \Gamma^ST \) would be a critical signature of the spin-orbit effect. We compare our result for a \( B_\parallel = 0.02 \text{ T} \) (\( \Gamma^ST \approx 500 \text{ ms} \)) to the experimental value obtained for GaAs (2.6 ms, 10) and find that Si has a singlet-triplet relaxation time more than two orders of magnitude larger than GaAs.

![FIG. 3: (Color online) \( \Gamma^ST \) as a function of \( B \) for two values of \( J \): The solid lines correspond to a plausible value of \( J = 0.02 \text{ meV} \) and the broken lines to \( J = \varepsilon_{ST} = 0.2 \text{ meV} \) for comparison, (a) as a function of \( B_\parallel \) and (b) as a function of \( B_\perp \), including Rashba (red) and without Rashba (black). \( \Gamma^ST \) increases with \( B_\parallel \) and decreases with \( B_\perp \).](image)

Note that the behaviour of \( \Gamma^ST \) here is different to the much studied GaAs-based device because of the nature of spin-orbit coupling and electron-phonon coupling in non-centrosymmetric materials: The BIA is absent and there are no piezo-phonons, also avoided crossings of the singlet and triplet energy levels does not occur for the magnetic fields considered here, giving a monotinous behaviour.

### III. DISCUSSION

There are several ways to measure \( \Gamma^ST \). In single-dot systems, this can be realized using a single pulse. Alternatively, one may use the following sequence: in the first phase, the state can be prepared so that only one electron is present in the QD, and in the next phase, the triplet would be available for conductance, unless it relaxes to the singlet. Measurement of the current for different values of the pulse duration then gives a direct method to determine \( \Gamma^ST \). The latter experiment has been performed in GaAs. In double-dot experiments, \( \Gamma^ST \) is one of the parameters in the rate equations that determine the measured current, so these experiments also provide an avenue for the determination of the singlet-triplet lifetime.

One should note immediately that \( \Gamma^ST \) is considerably longer in natural Si than in GaAs, generally by orders of magnitude. This is expected in a system with weaker spin-orbit coupling and fewer spinful nuclei. The times we find are of the order of seconds for the most part. It is possible to reduce the time by applying a perpendicular field, which can serve as a very useful diagnostic. It is also possible to lengthen \( \Gamma^ST \) by the use of isotopically enriched Si, i.e., pure \( ^{28} \text{Si} \). This would eliminate the hyperfine mechanism but it would not get rid of spin relaxation entirely, as higher-order effects of SOC are still present even at \( B = 0 \). However, these effects are quite small in Si. It seems likely that other effects such as flux noise will be the limiting factor in isotopically enriched Si.

In summary, we have calculated the dominant rates for phonon-assisted triplet-singlet relaxation of a silicon quantum dot. \( \Gamma^ST \) is found to be of the order of hundreds of ms, very sensitive to the exchange energy \( J \), and even longer in the presence of a \( B_\parallel \), to seconds. In the presence of a \( B_\perp \), a direct transition becomes possible, increasing (decreasing) \( \Gamma^ST \). Due to weak spin-orbit and hyperfine coupling, silicon offers very long coherence times, which are required for solid state qubits.

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