Realizing singlet-triplet qubits in multivalley Si quantum dots

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There has been significant progress in the implementation and manipulation of singlet-triplet qubits in GaAs quantum dots. Given the considerably longer spin coherence times measured in Si, considerable interest has been generated recently in Si quantum dots. The physics of these systems is considerably more complex than the physics of GaAs quantum dots owing to the presence of the valley degree of freedom, which constitutes the focus of this work. In this paper we investigate the physics of Si quantum dots and focus on the feasibility of quantum coherent singlet-triplet qubit experiments analogous to those performed in GaAs. This additional degree of freedom greatly increases the complexity of the ground state and gives rise to highly nontrivial and interesting physics in the processes of qubit initialization, coherent manipulation and readout. We discuss the operational definition of a qubit in Si-based quantum dots. We find that in the presence of valley degeneracy a singlet-triplet qubit cannot be constructed, whereas for large valley splitting ($\gg k_B T$) the experiment is similar to GaAs. We show that experiments on singlet-triplet qubits analogous to those in GaAs would provide a method for estimating the valley coupling in Si. A Zeeman field distinguishes between different initialized states for any valley splitting and provides a tool to determine the size of this splitting.

I. INTRODUCTION

Spin-based qubits are seen as promising candidates for scalable quantum computation, with donor and quantum-dot spins at the focus of research. Electrical readout and control of single spins in quantum dots (QDs) have proven challenging, yet GaAs double quantum dots (DQDs), where spin blockade and charge sensors enable observation of single/two-spin dynamics, have seen impressive experimental progress. In this article we establish the precise criteria for realizing spin qubits in Si QDs, where the multivalley structure of the ground state introduces fundamental complications in distinguishing spin and orbital degrees of freedom.

The original proposal by Loss and DiVincenzo made use of a quantum dot array, in which one electron spin on each dot constitutes the qubit. More recently there has been significant progress in implementing an alternative scheme in which the singlet and triplet states of two electrons in a DQD make up the qubit. One particular successful experiment involves initialization, manipulation, and measurement of two-spin singlet and triplet states. Here a $(0,2)$ singlet state is initialized, where $(n,m)$ indicates the occupancy of the left and right dots. Since the $(0,2)$—single dot—singlet and triplet are separated by an $\text{meV}$ gap, initialization of the singlet is easy and reliable. Tuning the gate voltages then allows tunneling of one electron to the left dot to form a $(1,1)$ singlet. When the bias is pushed deep into the $(1,1)$ regime [where $(1,1)$ is by far the electrostatic ground state configuration], the singlet and triplet are essentially degenerate due to the small tunnel coupling between the dots, so that a small magnetic field inhomogeneity (e.g. due to the Overhauser field of the nuclei) between the dots can rotate the two-electron states between the singlet and the triplet. After some mixing time in the $(1,1)$ regime, tuning the bias returns the system to the $(0,2)$ configuration, where electrical readout is possible due to spin blockade. This experiment clearly illustrates the existence of quantum coherence in the DQD system, and the distinct possibility of using the two-electron singlet and unpolarized triplet as the two states of a logical qubit, with reliable initialization, single-qubit rotation, and measurement.

Silicon is often regarded as the best semiconducting host material for spin qubits because of its excellent spin coherence properties: spin-orbit coupling is very small, the hyperfine interaction can be reduced by isotopic purification, and the electron-phonon interaction is weak as well. Furthermore, the mature Si microfabrication technology will help attempts to scale up a Si-based quantum computer (QC) architecture. At present, Si/SiGe and Si/SiO$_2$ quantum dots, and Si:P are being actively investigated and progress has been made in spin blockade in Si quantum dots. The biggest obstacle to spin QC in Si is valley degeneracy: bulk Si has six degenerate conduction band minima. While this degeneracy can be reduced by strain or the presence of an interface, it complicates the orbital and spin state spectrum and leads to valley-interference effects for spin interaction. At the Si/SiO$_2$ interface only two valleys are relevant to the ground orbital state. Scattering at the interface further lifts the valley degeneracy by producing a valley-orbit coupling $\Delta$. The magnitude of $\Delta$ is generally not known a priori and is sample-dependent. Currently measurement of valley splitting $\Delta$ is generally done for 2D electron gases at high magnetic fields, and
the zero-field valley splitting is extrapolated\textsuperscript{22}. We note that the case of large valley splitting has been examined in a number of recent publications\textsuperscript{23,24}.

In this article we study the physics of Si-based quantum dots and the feasibility of experiments analogous to Ref. \textsuperscript{4} in a Si/SiO\textsubscript{2} (or Si/SiGe)\textsuperscript{15} DQD, focusing on the effects of the valley degree of freedom on qubit initialization, operation, and spin blockade within the effective mass approximation. We identify the conditions required for an operational singlet-triplet qubit in Si. We further demonstrate that a quantum coherent experiment analogous to Ref. \textsuperscript{4} may provide a direct way to estimate the valley splitting $\Delta$. While our discussion focuses on Si/SiO\textsubscript{2} and is directly relevant to experiments on Si/SiO\textsubscript{2} quantum dots\textsuperscript{22}, the findings are generally applicable to Si quantum dots. In addition, we expect our findings to be at least qualitatively relevant to other systems in which the valley degree of freedom plays an important role, such as carbon, in which significant progress has been made lately\textsuperscript{25,26}.

The outline of this paper is as follows. We introduce the model of the DQD in Section II. We proceed to study the initialization process in singlet-triplet qubits in Section III followed by manipulation of the qubit in Section IV. In Section V we demonstrate that a quantum coherent experiment on singlet-triplet qubits can be used to estimate the valley splitting. Section VI is devoted to issues specific to silicon, such as interface roughness and the need for an external inhomogeneous magnetic field. Finally, Section VII contains a summary of our findings.

\section{Double Quantum Dot}

We choose nominally $\hat{z}$ as the growth direction for the Si/SiO\textsubscript{2} heterostructure we consider. The two dots are located at $R_{R,L}=(\pm X_0,0,0)$, where $R$ and $L$ stand for right and left respectively. The Hamiltonian is $H=H_0+H_v$, with $H_0=(\sum_{i=x,y,z} T^{(i)}+V_Q^{(i)})+V_{cc}$, where $T$ is the kinetic energy operator and $V_Q$ the confinement potential

$$V_Q = (1/2) m_1\omega_0^2 \min[(x-X_0)^2, (x+X_0)^2] - eE x$$

$$+ (1/2) m_1\omega_0^2 y^2 + (1/2) m_2\omega_2^2 z^2, \quad (1)$$

with $m_1$ and $m_2$ respectively the in- and out-of-plane Si effective masses. The Coulomb interaction between electrons at $r_1$ and $r_2$ is $V_{cc} = e^2/(e r_1 - r_2)$, where $e = (\epsilon_{Si}+\epsilon_{SiO2})/2$ includes the image charge in the SiO\textsubscript{2} layer. $H_v$ is a single-particle phenomenological coupling between the valleys discussed below. The electric field $E$ raises the energy of the left dot with respect to the right dot. The confinement potential and ground state for $E=0$ are identical in each dot, with the single-dot potentials $V_{R,L}(x) = (1/2) m_1\omega_0^2 (x\mp X_0)^2$. At the Si/SiO\textsubscript{2} interface the lowest valleys are at $\pm(k_z)$, with $k_z = 0.85(2\pi/a_{Si})$, and the lattice constant $a_{Si} = 5.43\AA$. The ground-state single-electron wave functions $R_{z,\bar{z}}$ and $L_{z,\bar{z}}$ represent the degenerate $\pm(k_z)$ valleys on the right and left dots respectively. In the right dot $(T+V_R)R_{z,\bar{z}} = \epsilon_0 R_{z,\bar{z}}$, with $R_{z,\bar{z}} = F_R(r-R_R)e^{\pm ik_z(r-R_R)}u_{z,\bar{z}}(r-R_R)$, and on the left $R \rightarrow L$. The envelope functions are

$$F_{R,L}(r-R_{R,L}) = \frac{1}{\pi^{3/4}(a b)^{1/2}} e^{-\frac{(x+x_0)^2}{a^2}} e^{-\frac{y^2}{b^2}} e^{-\frac{z^2}{2\omega_2^2}}, \quad (2)$$

where $a = \sqrt{\frac{\hbar}{m_1\omega}}$ and $b = \sqrt{\frac{\hbar}{m_2\omega_2}}$ are the in-plane (Fock-Darwin radius) and growth-direction confinement length. The lattice-periodic Bloch function $u_{z,\bar{z}}(r) = \sum K \sqrt{K^2} e^{iK \cdot r}$ with $K$ reciprocal lattice vectors. The overlap $\langle L_{z,\bar{z}} | R_{z,\bar{z}} \rangle = e^{-d^2}$ where $d = X_0/a$. Overlaps such as $\langle L_{z} | L_{\bar{z}} \rangle$, $\langle L_{\bar{z}} | L_{z} \rangle$ are suppressed by an exponential of the form $e^{-\frac{z^2}{2\omega_2^2}}$, where $Q_z = \frac{2\pi a_{Si}}{a_{Si}} - 2(k_z)$, with $n_z$ an integer. Such an exponential appears in all but one of the matrix elements of $H_0$ involving functions from different valleys. All such intervalley terms can be neglected except one, discussed below. The only nonzero matrix elements of $H_v$ are $\langle L_{z,\bar{z}} | H_v | L_{z,\bar{z}} \rangle = \langle R_{z,\bar{z}} | H_v | R_{z,\bar{z}} \rangle = \Delta$, with $\Delta > 0$ and assuming $\Delta$ has the same form on each dot. We define also $\varepsilon_R = \langle R_{z,\bar{z}} | (T+V_Q)R_{z,\bar{z}} \rangle$, $\varepsilon_L = \langle L_{z,\bar{z}} | (T+V_Q)L_{z,\bar{z}} \rangle$, and the dimensionless detuning as $\varepsilon_{R,L} = (\varepsilon_R - \varepsilon_L)/2\omega_0$. Diagonalizing the single-particle Hamiltonian with the valley coupling we obtain the valley eigenstates $R_{\pm} = (1/\sqrt{2}) (R_{z} \pm R_{\bar{z}})$ and $L_{\pm} = (1/\sqrt{2}) (L_{z} \pm L_{\bar{z}})$ with corresponding energies $\varepsilon_{R,L} \pm \Delta$. We orthogonalize these following Ref. \textsuperscript{27}, with $L_{\pm} = (L_{\pm} - gR_{\pm})/\sqrt{1-2g^2}$, with $g = (1 - \sqrt{1-T^2})/t$; for $R_{\pm}$ one swaps $L \leftrightarrow R$. These are the states that will be used henceforth.

Figure 1: (a) Single dot energy levels for finite $\Delta$ and $B$ such that $2\Delta > E_{\bar{z}}$. The lowest energy state is the singlet $S_1$, followed by the triplet $T_+$, the degenerate singlet $S_{z,\bar{z}}/triplet$ $T_0$ and triplet $T_-$, and the singlet $S_3$. Spin orientations of electrons in triplet states are indicated by arrows. (b) Schematic of the biased double dot. During initialization the detuning is large, and the $(0,2)$ states are lowest in energy. After loading the detuning is lowered and the $(1,1)$ states are at the same energy as the $(0,2)$ states – the charge transition regime. An inhomogeneous magnetic field mixes the singlets and triplets.
III. INITIALIZATION

We begin by studying the initialization process, which involves loading two electrons onto the right dot. For this purpose it is imperative to analyze first the spectrum of the doubly-occupied right dot, that is the configuration $(0,2)$. The lowest-energy two-particle spatial wave functions are $\phi_{S1,S3} = \hat{R}^{(1)}_+ \hat{R}^{(2)}_+$ and

$$\phi_{S2} = \frac{1}{\sqrt{2}} (\hat{R}^{(1)}_+ \hat{R}^{(2)}_- + \hat{R}^{(2)}_+ \hat{R}^{(1)}_-)$$

$$\phi_T = \frac{1}{\sqrt{2}} (\hat{R}^{(1)}_+ \hat{R}^{(2)}_+ - \hat{R}^{(2)}_+ \hat{R}^{(1)}_-),$$

where $(i)$ denotes the $i$th electron. In the basis $\{\phi_{S1}, \phi_{S2}, \phi_T, \phi_{S3}\}$ the matrix elements of the Hamiltonian are $2\varepsilon R + u + \text{diag}(-2\Delta, 0, 0, 2\Delta)$, where $u = \int d^3r_1 f d^3r_2 R_{z,1}^{(1)} R_{z,2}^{(2)} V_{ee} R_{z,1}^{(1)} R_{z,2}^{(2)}$. The valley-exchange Coulomb integral $\int d^3r_1 f d^3r_2 R_{z,1}^{(1)} R_{z,2}^{(2)} V_{ee} R_{z,1}^{(1)} R_{z,2}^{(2)}$ is not suppressed by an exponential, however we find its value to be $\ll 1\mu$eV and it will therefore be assumed of no consequence henceforth. For $\Delta = 0$ and no external magnetic field $B$, all levels are degenerate so that it is impossible to load any particular two-electron state. The spectrum for finite $\Delta$ and $B$, yielding a Zeeman energy $E_Z$ with $2\Delta > E_Z$, is shown in Fig. 1. The triplet states thus split into $T_+, T_0$, and $T_-$, separated in energy by $E_Z$.

The loading process makes use of an outside reservoir with Fermi energy $E_F$ which is thermally broadened by $\sim k_B T$. The reservoir is tuned to be on resonance with the lowest-energy singlet state. Here we neglect the the differences in tunnel couplings between the various states and the reservoir. Consequently the probability of loading any of the states is proportional to the Fermi distribution at its energy. If $\Delta \gg k_B T$, the lowest-energy singlet state can be loaded exclusively. Numerically $\Delta \approx 0.1\mu$eV is sufficient to fulfill this condition at dilution refrigerator temperatures of $T=100$mK, where $k_B T \approx 0.01\mu$eV. In this regime the two-electron initialization process in a Si DQD is identical to the GaAs DQD in Ref. [4].

IV. QUBIT MANIPULATION

Manipulation of the singlet-triplet qubit involves switching to the configuration $(1,1)$, where singlet-triplet mixing is achieved through the us of an inhomogeneous $B$. We study the Hilbert space of two electrons in a Si DQD. We do not include the high-energy $(2,0)$ states. The seven space-symmetric Hund-Mulliken (HM) wave functions of the lowest-energy singlets are $\phi_{S1}, \phi_{S2}, \phi_{S3}$ and the functions

$$\phi^+_S = \frac{1}{\sqrt{2}} (\hat{L}^{(1)}_+ \hat{R}^{(2)}_+ + \hat{L}^{(2)}_+ \hat{R}^{(1)}_+)$$

$$\phi^m_S = \frac{1}{\sqrt{2}} (\hat{L}^{(1)}_+ \hat{R}^{(2)}_+ + \hat{L}^{(2)}_+ \hat{R}^{(1)}_+),$$

These singlet states split into three uncoupled subspaces. The $\{\phi^+_S, \phi_{S1}\}$ and $\{\phi^-_S, \phi_{S1}\}$ subspaces are composed of HM wave functions where the two electrons are in the same valley eigenstate, while the subspace $\{\phi^m_S, \phi^m_{S2}\}$ consists of wave functions where electrons are in different valley eigenstates. The five antisymmetric counterparts of the states in Eq. (4), denoted by $\phi^n_T, \phi^{m^n}_T$ and $\phi^n_{T2}$, are evident (clearly $\phi_{S1}$ and $\phi_{S3}$ do not have antisymmetric counterparts.) These triplets in turn split into three subspaces, with $\{\phi^+_T\}$ and $\{\phi^-_T\}$ single-valley HM triplets, and $\{\phi^{m^+_T}, \phi^{m^-_T}, \phi^{m^n}_T\}$ mixed-valley triplet states. Since the overlap between states from different valleys is negligible, matrix elements of the form $\langle \phi^m_S | H_0 | \phi^+_S \rangle$ and $\langle \phi^{m^n}_T | H_0 | \phi^{m^+_T} \rangle$ are equal. As a result of this the mixed singlet and triplet subspaces always yield the same energy eigenvalues.

Let us examine a concrete Si DQD with $a=8.2$nm, $b=3$nm, $d=2.45$ and $\Delta=0.1\mu$eV and zero magnetic field. The top and bottom anticrossings each consist of two singlets (solid lines) and one triplet (dotted line). In the middle anticrossing each of the three dashed lines represents a degenerate singlet/triplet level.

Figure 2: DQD spectrum for $a=8.2$nm, $b=3$nm, $d=2.45$ and $\Delta=0.1\mu$eV and zero magnetic field. The top and bottom anticrossings each consist of two singlets (solid lines) and one triplet (dotted line). In the middle anticrossing each of the three dashed lines represents a degenerate singlet/triplet level.
VI. ISSUES SPECIFIC TO SILICON

In GaAs the inhomogeneous magnetic field is produced by the hyperfine interaction. In Si the hyperfine inter-
action is smaller and singlet-triplet mixing will be about two orders of magnitude slower than in GaAs. Using a nanomagnet one can design a particular field magnitude and direction, enabling better control of the spin qubit. For example an inhomogeneous magnetic field along the $\hat{z}$-direction mixes only the singlet and $T_0^+$, whereas a field along the $\hat{x}$-direction mixes only the singlet and $T_\pm^L$. Evidently the issues discussed in this work are insensitive to the origin of the inhomogeneous magnetic field as long as this field is present in the system.

The length scale of surface roughness, which determines the spatial variation of $\Delta$, as compared with the dot size and location is not precisely known. The proposed experiments will work as long as $\Delta$ varies over a length scale larger than the DQD size, or as long as the change in $\Delta$ does not lead to change in the compositions of the valley eigenstates. We have assumed the same valley-orbit coupling in both QDs, thus the same valley splitting and eigenstates. A change in the valley composition of the eigenstates could lead to intervalley scatterings in the (0,2) to (1,1) transition, so that control of electron orbital states may become intractable. At the same time large variation of $\Delta$ across the DQD will hinder the effectiveness of the experiment. It is imperative for experimental setups to ensure firstly that the interface is as smooth as possible and secondly that the DQD spans an area over which the interface roughness profile varies as little as possible.

VII. SUMMARY

We have studied the feasibility of initialization and coherent manipulation of singlet-triplet qubits in multivalley Si DQDs, demonstrating that the valley degree of freedom makes the physics of Si quantum dots considerably different from that of dots made out of single-valley systems such as GaAs. Various experimental outcomes are possible depending on the value of the valley splitting $\Delta$. For large $\Delta$ (i.e. $\Delta \gg k_B T$) a quantum coherent experiment identical to Ref. [4] is feasible. For small $\Delta$ a number of different states may be initialized, leading to different experimental outcomes. One interesting highlight of our work is that, although several singlet/triplet states may be initialized, in general each state can mix with one other state, and no more. Therefore, in principle, once a state is loaded, operations on it can proceed in a similar way to the scheme implemented in GaAs dots. For any $\Delta$, sweeping a uniform magnetic field provides a useful method for estimating $\Delta$. In fact, one very important consequence of our work is the proposed new method for estimating the valley splitting $\Delta$ in Si quantum dots, particularly when $\Delta \lesssim k_B T$. Considering the difficulties inherent in proving that a certain state belongs to a particular valley, and thus in identifying a particular energy splitting with the valley splitting, it will be important to have as many different methods as possible to measure/estimate the size of the valley splitting.

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