Exchange in Silicon-Based Quantum Computer Architecture

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The silicon-based quantum computer proposal has been one of the actively pursued ideas during the past three years. Here we calculate the donor electron exchange in silicon and germanium, and demonstrate an atomic-scale challenge for quantum computing in Si (and Ge), as the six (four) conduction-band minima in Si (Ge) lead to intervalley electronic interference, generating strong oscillations in the exchange splitting of two-donor two-electron states. Donor positioning with atomic-scale precision within the unit cell thus becomes a decisive factor in determining the strength of the exchange coupling — a fundamental ingredient for two-qubit operations in a silicon-based quantum computer.

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Following the seminal proposal [1] by Kane, there has been a great deal of activity [2–4] in efforts to develop a silicon-based quantum computer (QC) architecture. The basic ideas of the Kane proposal are simple and attractive: to use donor nuclear spins as quantum bits (qubits), and to utilize the vast infrastructure and technology associated with the Si industry to fabricate precisely controlled Si nanostructures, where exchange effects between electrons and nuclei in neighboring donor impurities (e.g., ^{31}P in Si) could serve as the two-qubit gates, similar to the electron-spin-based QC proposal by Loss and DiVincenzo [5]. The motivation for a Si quantum computer is obvious: Once the basic one-qubit and two-qubit operations have been demonstrated using donor impurities in Si nanostructures, computer chip fabrication technology associated with the existing and dominant Si industry will easily enable the scale-up of information processing involving a large number of donor nuclear spin qubits. Indeed, one of the formidable stumbling blocks in developing working quantum computer hardware has been the scale-up problem, as the demonstrated qubits in trapped ion and liquid state NMR techniques are not readily scalable in any significant manner [6].

A great deal of experimental work is currently being aimed at developing suitable qubits in Si nanostructures with precisely introduced dopant impurities, using both a "top-down" approach with ion implantation, and a "bottom-up" approach with molecular beam epitaxy (MBE) growth and scanning tunneling microscopy [4]. In the Si QC model [1,2], donor electrons act as shuttles between different nuclear spins. For two-qubit operations, which are required for a universal QC, both electron-electron exchange and electron-nucleus hyperfine interaction need to be precisely controlled. These are unquestionably formidable experimental problems. In the original proposal, Kane used the Herring-Flicker exchange formula [7] for two hydrogenic centers to obtain an order of magnitude estimate of the electron exchange among donors in Si [1]. However, as he also pointed out, donor exchange in Si is not hydrogenic.

In this Letter, we show that exchange effects in proposed donor nuclei based Si QC architectures are actually very subtle due to quantum interference effects inherent in the complicated Si band structure. In particular, special care, going far beyond what is being currently attempted in the fabrication of Si QC, will be required in controlling the surface gates crucial to QC operations. We also provide results for Ge, which seems to have certain advantages over Si as a candidate for dopant exchange based quantum computation. Unfortunately, Ge is not such an attractive practical alternative since there is no well-established Ge technology to take advantage of.

The study of shallow impurities in Si and Ge is a quite mature field [8,9]. However, complications arising from the anisotropy of the electron effective mass and from interference among the Bloch wave functions at the degenerate conduction-band edges were never fully explored in detail. Both effects were discussed by Andres *et al.* [10] statistically, in a study of magnetic susceptibility of the Si:P system performed in the context of localization and magnetic phase transitions. The silicon conduction band has six minima located along the $\langle 100 \rangle$ directions, at about 85% between the center (Γ point) and the boundary (X points) of the Brillouin zone (BZ): $\mathbf{k}_{\pm z} = 0.85(0, 0, \pm 2\pi/a)$, and the equivalent x, y directions. The interference between these valleys causes fast oscillations (on the scale of the interatomic spacing, quite different from the slowly varying exchange interaction in the spin-based quantum dot QC model [5,11–13]) in the exchange interaction. Such oscillations cannot be accounted for by a simple calculation using hydrogenic centers. Here we perform a Heitler-London calculation of the exchange coupling [11] between two substitutional donors in bulk Si, incorporating the effects of both the anisotropic effective mass and the valley interference. Within the envelope function approach, the ground single valence donor electron state (with A_1

symmetry) is written as a symmetric superposition of the six conduction-band minima [8]:

$$\psi(\mathbf{r}) = \frac{1}{\sqrt{6}} \sum_{\mu}^{6} F_{\mu}(\mathbf{r}) \phi_{\mu}(\mathbf{r}), \tag{1}$$

where $\phi_{\mu}(\mathbf{r}) = u_{\mu}(\mathbf{r}) e^{i\mathbf{k}_{\mu}\cdot\mathbf{r}}$ are Bloch wave functions. Here $\mathbf{k}_{\mu}$ refer to the six conduction-band minima, and $F_{\mu}(\mathbf{r})$ are the corresponding envelope functions. We use the Kohn-Luttinger variational form for these anisotropic envelopes [8,9], e.g.,

$$F_{\pm z}(\mathbf{r}) = \frac{1}{\sqrt{\pi a^2 b}} e^{-\sqrt{(x^2+y^2)/a^2 + z^2/b^2}}. \tag{2}$$

To determine the effective Bohr radii a and b, we have used the most recently measured Si and Ge parameters [14] (especially longitudinal and transverse effective masses) to perform a variational calculation [9]. The calculated wave function widths, as shown in Table I, are quite close to those obtained over 40 years ago [8]. The large difference in transverse mass has only a minimal effect on our results.

To calculate the exchange splitting between the ground singlet and triplet states for an impurity pair in silicon, we use an approximate form of the Heitler-London approach [10], which leads to

$$J(\mathbf{R}) = \int d^3\mathbf{r}_1\, d^3\mathbf{r}_2\, \psi^*(\mathbf{r}_1)\psi^*(\mathbf{r}_2 - \mathbf{R}) \times \frac{e^2}{\epsilon|\mathbf{r}_1 - \mathbf{r}_2|} \psi(\mathbf{r}_1 - \mathbf{R})\psi(\mathbf{r}_2) \tag{3}$$

$$= \frac{1}{36} \sum_{\mu,\nu} \left[\sum_{\mathbf{K},\mathbf{K}'} |c_{\mathbf{K}}^{\mu}|^2 |c_{\mathbf{K}'}^{\nu}|^2 e^{i(\mathbf{K}-\mathbf{K}')\cdot\mathbf{R}} \right] j_{\mu\nu}(\mathbf{R}) \times \cos(\mathbf{k}_{\mu}\cdot\mathbf{R}) \cos(\mathbf{k}_{\nu}\cdot\mathbf{R}), \tag{4}$$

where $\mathbf{R}$ is the relative position of the impurity nuclei pair, and

$$j_{\mu\nu}(\mathbf{R}) = \int d^3\mathbf{r}_1\, d^3\mathbf{r}_2\, F_{\mu}^*(\mathbf{r}_1) F_{\nu}^*(\mathbf{r}_2 - \mathbf{R}) \times \frac{e^2}{\epsilon|\mathbf{r}_1 - \mathbf{r}_2|} F_{\mu}(\mathbf{r}_1 - \mathbf{R}) F_{\nu}(\mathbf{r}_2). \tag{5}$$

The second summation in (4) comes from the reciprocal lattice expansion of the periodic part of the Bloch function, $u_{\mu}(\mathbf{r}) = \sum_{\mathbf{K}} c_{\mathbf{K}}^{\mu} e^{i\mathbf{K}\cdot\mathbf{r}}$, and is identically 1 when $\mathbf{R}$ is an fcc crystal lattice vector. Integrals in Eq. (3) with rapidly oscillating integrands are neglected in Eq. (4). For $\mu = \nu$, we calculate the integrals in (5) by replacing the denominator $|\mathbf{r}_1 - \mathbf{r}_2|$ by its value for the line along the impurities [10], and assume $j_{\mu\nu} = \sqrt{j_{\mu\mu} j_{\nu\nu}}$ when $\mu \neq \nu$. All are excellent approximations for large separations ($|\mathbf{R}| \gg a, b \gg$ a).

TABLE I. Experimental values for the lattice parameter, dielectric constant, and electron effective masses (in units of the free electron mass) for Si and Ge [14]. The last two columns give the calculated values of the effective Bohr radii.

	a (Å)	ϵ	$m_{\parallel}$	$m_{\perp}$	a (Å)	b (Å)
Si	5.43	12.1	0.916	0.191	25.09	14.43
Ge	5.657	16	1.58	0.082	64.21	22.83

The germanium conduction band has four minima along the $\langle 111 \rangle$ directions, at the zone boundary L points. Straightforward changes in (1) lead to expressions for the exchange coupling in Ge similar to (4), with μ and ν along the $\langle 111 \rangle$ directions. Because the conduction-band minima are located at the zone boundary, the oscillation in the exchange coupling should display a simpler behavior than in Si when both donors are on the same fcc sublattice, thus making Ge an intriguing candidate for the purpose of quantum computing.

We calculate the exchange energy for a pair of donors at $\mathbf{R}_A$ and $\mathbf{R}_B$ as a function of their relative position $\mathbf{R} = \mathbf{R}_A - \mathbf{R}_B$ from Eq. (4), taking the expression in the square bracket equal to unity. This means that the results are appropriate for any lattice vector $\mathbf{R}$, while for general interdonor distances our values should be taken as an estimate. For definiteness, we consider $\mathbf{R}$ along high-symmetry directions in the crystal lattice. Figure 1 shows the calculated values of $J(\mathbf{R})$ for donors in Si and Ge with $\mathbf{R}$ along the [100], [110], and [111] directions [frames (a), (b), and (c), respectively]. The solid lines give the results for Si, and exhibit the expected decay of J with increasing $|\mathbf{R}|$ due to the decrease of the donor wave function overlap. Other general features of Si and Ge exchange are clearly illustrated in this figure, namely, the oscillatory behavior of J superimposed on its overall decay with distance, and the strong anisotropy of $J(\mathbf{R})$ which is apparent by comparing different frames. Both features are consequences of the host material band structure and have not been considered in detail in previous studies [15], either for simplicity [1,3] or because such effects are averaged out for a random donor distribution [10]. The filled circles in Fig. 1 indicate the accessible values of $J(\mathbf{R})$ when the impurity pair in Si is located at lattice sites along the considered directions. Since for Si the conduction-band minima correspond to points inside the BZ, the period of oscillation in J and the lattice periodicity are not commensurate. We have also investigated the effect of small perturbations in the atomic positions: The open squares give the resulting exchange values when one of the impurities is slightly displaced to off-lattice positions. The set of squares around each circle in Fig. 1 corresponds to displacements along different directions, with the distance from the original lattice position arbitrarily taken as $\delta = 0.235$ Å (i.e., 10% of the nearest-neighbor distance in Si). The squares follow to a very good approximation the behavior of the calculated $J(\mathbf{R})$, with $\mathbf{R}$ along the unperturbed crystal direction, regardless of the direction of δ. The upper bound of $\Delta J/J$ calculated from the small-displacement data is about 2.5% to 5% along the [100] direction, which means that the requisite control over donor positioning should be much better

than 10% of the nearest-neighbor distance, i.e., better than 0.235 Å, a rather difficult task.

The corresponding results for donor pairs in Ge are given by the dashed lines, diamonds, and crosses in Fig. 1. In these calculations we have also assumed nearly free electrons, which means $c_0^\mu \gg c_{\mathbf{K}\neq 0}^\mu$. The main qualitative difference between the calculated exchange coupling for donors in Ge and Si arises from the different locations of the conduction-band minima in the BZ. Since for Ge the minima occur at the zone boundary L points, the oscillations in $J(\mathbf{R})$ are commensurate with the lattice periodicity. For donors at lattice sites along the [100] direction, the accessible values of $J(\mathbf{R})$ correspond exactly to successive local maxima of Eq. (3). As a consequence, off-lattice displacements from the original lattice sites by 10% of the nearest-neighbor distance in germanium ($\delta = 0.245$ Å) have a negligible effect on the exchange coupling for donors along this direction. Quantitatively, the longer range for the interactions in Ge as compared to Si is due to the larger values of the effective Bohr radii for Ge (see Table I).

Our results indicate that a ^{31}P donor array along the [100] direction in either of the host materials meets the requirements for quantum computer implementation. The exchange energy for two ^{31}P substitutional impurities 100 to 200 Å apart along this direction ranges between a few tenths of one meV to one μeV, corresponding to less than 100 ps to 100 ns gate operation time. These values are robust with respect to small off-lattice displacements.

Displacements of the ^{31}P atoms into neighboring lattice sites, which are bound to occur in the fabrication process due to either uncontrolled implantation or surface diffusion during MBE growth, also deserve careful theoretical investigation. In Fig. 2 we present the calculated $J(\mathbf{R})$ for a specific relative position $\mathbf{R}_0$ along the [100] direction (black symbols), as well as for values of $\mathbf{R} = \mathbf{R}_0 + \mathbf{\Delta}$, with $\mathbf{\Delta}$ ranging over the 4 nearest neighbors, 12 second nearest neighbors, and 6 third nearest neighbors in the diamond structure (see inset of Fig. 2). These displacements cause a relatively small change in the interdonor distance (see horizontal scale in the figure). For third neighbors (blue symbols) the trends in Fig. 1 are reasonably preserved. Surprisingly, most first and second neighbor displacements essentially destroy the exchange coupling. This effect is entirely due to the $\cos(\mathbf{k}_\mu \cdot \mathbf{R})$ factors in Eq. (4), coming from the plane-wave part of the Bloch functions, and is not directly related to the numerical values or approximations involved in obtaining the $j_{\mu\nu}$ coefficients. Therefore we do not attempt to refine the envelope function calculation at this stage, e.g., by including central cell corrections [8] and intervalley coupling [16]. The results obtained here are

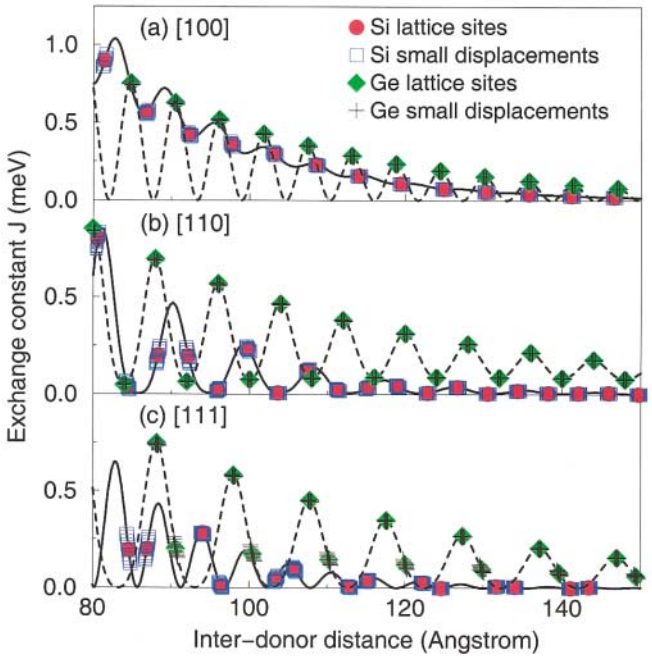

FIG. 1 (color). Calculated exchange coupling between two phosphorus donors in Si (solid lines) and Ge (dashed lines) along high-symmetry directions for the diamond structure (see inset of Fig. 2). Values appropriate for impurities at substitutional sites are given by the circles (Si) and diamonds (Ge). Off-lattice displacements by 10% of the nearest-neighbor distance lead to the perturbed values indicated by the squares (Si) and crosses (Ge).

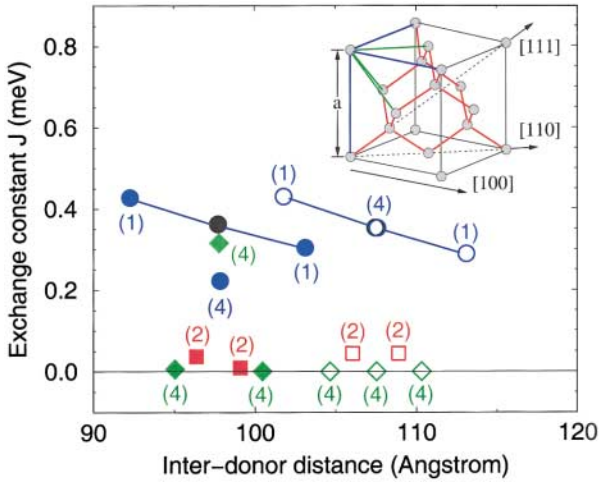

FIG. 2 (color). Variations in the exchange coupling between two phosphorus donors in Si (solid symbols) and Ge (open symbols). Lattice displacements into one of the first (red), second (green), or third (blue) neighboring positions follow the color code represented in the inset, which depicts a diamond structure with colored bonds. The numbers in the parentheses next to the data points are their degeneracies, respectively. For clarity, not all bonds connecting neighbors are shown in the inset. Black circles refer to the reference positions $\mathbf{R}_0$, with the two donors exactly along the 100 direction. The red squares give the exchange constant when one of the donors is displaced into one of its nearest-neighbor sites, while green diamonds represent the second nearest-neighbor values and blue circles the third nearest-neighbor values. The points connected by a line refer to pairs along the [100] direction, displaced by $\pm a$ with respect to the reference position, and may be easily identified in Fig. 1(a).

inconsistent with the commonly accepted idea that, when the Bohr radii of the envelope functions are much bigger than the lattice parameter, the substitutional positioning of the impurity atoms is not of much importance in determining the exchange coupling.

In a Si QC architecture, there are surface gates (so-called A and J gates) that are used to control single- and two-qubit operations [1]. By adjusting the gate potential(s), one can shift the center of a donor electron wave function, thus quickly traversing the fast oscillating terrain in the exchange coupling. The results obtained in this paper also indicate that donor electron exchange depends very sensitively on the applied gate potential(s). Furthermore, whether the electrons are trapped by donor ionic potentials or by the applied gate potentials, the fast oscillation in exchange will persist as long as the electrons are in the bulk of a silicon crystal. For a two-qubit gate of a QC, it is the time integral of the exchange constant J that determines the gating time [5]. The implication of the oscillatory exchange is that the A and J gate voltages corresponding to the peak exchange coupling have to be well controlled, optimally close to a local maximum where the exchange is least sensitive to the gate voltage. Since the oscillatory exchange period is close to lattice spacing, the positioning of the donor electrons by the A and J surface gates must be controlled at least to that precision. Furthermore, if one intends to achieve adiabaticity by increasing the switch-on time of the gate, so that $\int J\,dt = (2n + 1)\pi\hbar$ with $n \gg 1$ [11], the error in J due to gate inaccuracies would accumulate in the integral. It is thus crucial to precisely control the surface gates in order to minimize the possible errors.

Placing the donors and therefore their electrons in the middle of a symmetric Si quantum well will help reduce the fast phase oscillations because only two of the six bulk Si conduction band valleys would contribute to the donor electron ground state [2], suppressing the magnitude of the interference effect. In a symmetric quantum well, the splitting between the ground state and the next excited state (in analogy to the A_1 and T donor electron states in bulk) is much smaller than in the bulk (crudely estimated to be about 3 meV from symmetry arguments, in contrast to nearly 12 meV in bulk Si; the smaller splitting implies that the adiabatic condition is somewhat harder to satisfy in the quantum well). Strains, electric fields, and/or asymmetry in the quantum well might help eliminate the remaining valley degeneracy [3] so that all the fast oscillating factors in the exchange constant J may be removed, thus leading to a slowly varying J and a much easier control of the exchange gate. However, strains themselves are hardly controllable, while asymmetry in a quantum well may introduce additional complications that lead to decoherence. Further investigations of these factors are still ongoing and the results will be reported elsewhere [17].

As we have demonstrated above, moving one of the donors to its second nearest-neighbor position causes strong suppression in exchange coupling between the two donor electrons. This cancellation of exchange due to valley interference might be useful for isolating the neighboring qubits. In a Si QC, for most of the time the qubits should not "talk" to each other. Unwanted interaction would lead to leak of information and decoherence. Therefore, the positions with vanishing exchange interaction might provide a quieter environment compared to an arbitrary pair of positions for qubits.

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[15] Oscillatory exchange is not uncommon in solids. For example, the RKKY mechanism produces oscillatory exchange between core spins mediated by conduction electrons due to the sharpness of the Fermi surface. Our results are completely different from RKKY-type Fermi surface related mechanisms as we are dealing with direct exchange between donor electrons. The oscillatory behavior here is due to intervalley quantum interference arising from the particular indirect band structure of Si and Ge. Indeed, the oscillation in donor exchange might provide a means to measure the locations of the conduction-band minima in Si. Since the fast oscillation in exchange is incommensurate with the lattice constant, one might be able to obtain information about $\mathbf{k}_\mu$ by examining the slow oscillation of exchange when both donors sit on the same cubic lattice, assuming such precise placement of the donors can be done.
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