Electric field driven donor-based charge qubits in semiconductors

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We investigate theoretically donor-based charge qubit operation driven by external electric fields. The basic physics of the problem is presented by considering a single electron bound to a shallow-donor pair in GaAs. This system is closely related to the homopolar molecular ion $H^+$. In the case of Si, heteropolar configurations such as $PSb^+$ pairs are also considered. For both homopolar and heteropolar pairs, the multivalley conduction band structure of Si leads to short-period oscillations of the tunnel-coupling strength as a function of the inter-donor relative position. However, for any fixed donor configuration, the response of the bound electron to a uniform electric field in Si is qualitatively very similar to the GaAs case, with no valley quantum interference-related effects, leading to the conclusion that electric field driven coherent manipulation of donor-based charge qubits is feasible in semiconductors.

In this Communication we investigate the electric field manipulation of donor-based charge qubits in Si. For a better physical insight, it is instructive to consider initially donor-pair molecular ions in GaAs. Although shallow donor pairs in GaAs have simple and easy-to-control properties, sample preparation in GaAs poses an obvious difficulty that substitutional impurities at Ga or As sites will have entirely different behaviors (e.g., group-IV elements act as donors when replacing Ga, but acceptors when replacing As). Therefore the GaAs results presented here should be taken as a benchmark over which Si band effects are more clearly assessed. Furthermore, shallow donor binding energies in Si present a much wider distribution of values compared to GaAs, ranging from 43 meV for Sb to 71 meV for Bi. Thus the optimal coupling of different donor species in Si (forming an heteropolar $AB^+$ molecular ion, which is feasible experimentally) would require the assistance of an external electric field, which again poses the interesting question on whether valley interference in these double donor systems might lead to any difficulty in the charge manipulation by the external field. Our goal here is to assess the prospect for the coherent manipulation of donor-based Si charge qubits, similar to that achieved in GaAs, and Si quantum dot based systems, in light of the known problem associated with the quantum interference among the valleys in the Si donor-based systems.

We model shallow donors in semiconductors within the effective mass approximation (EMA) where the electronic properties of the host material are described by a few parameters, usually taken to be the static dielectric constant $\varepsilon$ and the effective masses at the conduction-band edge. The case of GaAs is particularly simple theoretically, since the conduction band minimum at $k = 0$ is non-degenerate and isotropic, characterized by an effective mass $m^* = 0.067m_0$, where $m_0$ is the free electron mass, and $\varepsilon = 12.56$. We consider group-IV (VI) impurities replacing the group-III (V) element Ga (As), leading to a donor state, i.e., an additional electron in the system...
bound to a hydrogenic potential $U_i(r) = -e^2/(\varepsilon |r - \mathbf{R}_i|)$ centered at the impurity site $\mathbf{R}_i$. The one-electron Hamiltonian is written as $H = H_{GaAs} + U_i(r)$, where the first term describes the bulk semiconductor material (kinetic energy and periodic potential). Within EMA the electron bound to the donor is assumed to be highly delocalized in real space (with respect to the lattice parameter), thus strongly localized in reciprocal space. Additional approximations allow the electron wave function to be written as $\psi_i = F_i(\mathbf{r})\phi_k(\mathbf{r})$, the product of a slowly varying envelope function $F$ by the band-edge Bloch state $\phi_k$ which, in the case of GaAs, reduces to the rapidly varying periodic part of the Bloch function $u_0$. The envelope function is the solution of the EMA Hamiltonian $H_i = -\hbar^2 \nabla^2/(2m^*) + U_i(r)$, leading to $F_i(\mathbf{r}) = [1/\sqrt{\pi(m^*)}] \exp(-r - R_i)/a^* )$ with effective Bohr radius $a^* = \varepsilon(m_0/m^*)a_0 \approx 10$ nm and the eigen-energy for the donor state with respect to the bottom of the conduction band $E_D = -[m^*/(\varepsilon^2 m_0)]Ry \approx -6$ meV, in excellent agreement with shallow donor binding energies in GaAs (see Table I).

The Hamiltonian for a single electron bound to an $A - B$ donor pair at sites $\mathbf{R}_A$ and $\mathbf{R}_B$ in GaAs is

$$H_{A-B}(\mathbf{E}) = H_{GaAs} + U_A + U_B + |e|\mathbf{E} \hat{s} \cdot \mathbf{r},$$

(1)

where the last term accounts for an external axial field $\mathbf{E} = \hat{s} \mathbf{E}$, with $\hat{s} = \mathbf{R}/R$ and $\mathbf{R} = \mathbf{R}_B - \mathbf{R}_A$. For large enough $R (R > 4a^*)$ this problem may be solved in analogy with the LCAO solution for the $H^+_2$ molecular ion.

The Hamiltonian is written in the non-orthogonal basis set $\{\psi_A, \psi_B\}$, which amounts to writing an EMA Hamiltonian in the $\{F_A, F_B\}$ basis set, since the periodic part of the Bloch function may be ignored in practice. All matrix elements are integrated analytically by separation of variables using spheroidal coordinates $\lambda = (r_A + r_B)/R$ and $\mu = (r_A - r_B)/R$, with $r_i = |\mathbf{r} - \mathbf{R}_i|$, $i = A, B$. The electric field related term, not considered in Ref. 19, is transformed here through the relation $\hat{s} \cdot \mathbf{r} = \lambda \mu R/2$.

Figure 1 summarizes our results for donor charge qubits in GaAs. The inset gives the characteristic level anticrossing of the qubit states, driven here by the axial electric field, with the minimum gap $\Delta_{min}$ occurring at the anticrossing field $\mathbf{E}_c = 0$. Far from the anticrossing region, the eigenstates correspond to the electron localized around one of the individual donors, which we represent by $|A\rangle$ and $|B\rangle$ states, while for $\mathbf{E} = \mathbf{E}_c$ the ground state is the symmetric superposition $|+\rangle = (|A\rangle + |B\rangle)/\sqrt{2}$ and the first excited state is the anti-symmetric superposition $|-\rangle = (|A\rangle - |B\rangle)/\sqrt{2}$. As the interdonor distance $R$ increases, $\Delta_{min}$ decreases smoothly, ranging typically from one to two orders of magnitude smaller than the single donor binding energy as $R$ varies from $4a^*$ to $8a^*$.

The theoretical description of shallow donors in Si is more complex than in GaAs due to the six-fold degeneracy of the conduction band minimum of Si, corresponding to non-zero values of $k_u$ with $\mu = 1, \ldots, 6$. Minima are located in reciprocal space 0.85 of the way along the Γ-X line. The energy dispersion near each of the band minima is anisotropic, leading to different effective masses for the longitudinal and transverse directions along each of the Γ-X lines. The tunnel coupling of $P_2^+$ homopolar charge qubits in Si was investigated in detail in Ref. 13 with the single donor wavefunctions taken in the Kohm-Luttinger (KL) EMA form, i.e., for a donor at $\mathbf{R}_A$,

$$\psi_A(\mathbf{r}) = \frac{1}{\sqrt{6}} \sum_{\mu=1}^{6} F_\mu(\mathbf{r} - \mathbf{R}_A)u_\mu(\mathbf{r})e^{ik_u \cdot (\mathbf{r} - \mathbf{R}_A)},$$

(2)

where the envelope functions $F_\mu(\mathbf{r})$ are deformed 1S hy-
driodogenic orbitals. With respect to the GaAs results in Fig. 1, the most striking difference is the oscillatory and anisotropic behavior of $\Delta_{\text{min}}$ vs $R$ for $P^+_2$ in Si. It is well established\cite{15,21,22} that this behavior arises from interference between the plane-wave parts of the Bloch functions, which are pinned to the respective donor sites.

For the heteropolar donor pairs, the multivalley character of the donor electron wavefunction is accounted for in the form of Eq. (2). We adopt an additional simplification assuming isotropic envelope functions: $F_{\mu}(r) = [1/\sqrt{\pi}(a_D^*)^3]\exp(-|r - R_\mu|/a_D^*)$, where $D$ labels the donor species. We also assume that, as for the case of GaAs, the experimental donor binding energy satisfies $E_D = \langle \psi_D | H_D | \psi_D \rangle$, where $H_D$ is the single-donor multivalley Hamiltonian. Our treatment of the donor pair under an electric field follows closely the procedure outlined above for GaAs: The donor pair Hamiltonian in the presence of an axial electric field, $H_{A-B}(E)$, is written in the $\{\psi_A, \psi_B\}$ representation, in which one and two-center integrals are readily calculated in spheroidal coordinates. We adopt here the values of the effective Bohr radii calculated by Ning and Sah\cite{21} for the $1S(A_1)$ state of the different donors within the multivalley effective mass spherical-band approximation. This approach involves a parametrization of the donor potential to fit the experimental values of $E_D$. In Table I we present the values of $a_D^*$ and $E_D$ adopted here. Comparison with the KL effective Bohr radii (2.5 nm and 1.4 nm) shows that $a_D^*$ is probably underestimated. We expect our model to be more reliable regarding the energy estimates, since all donor binding energies are taken from experiment.

Typical electric-field driven level anticrossing diagrams for donor molecular ions $AB^+$ are given in Fig. 2, where one of the donors ($A$) is always chosen to be $P$. As compared to the $P^+_2$ case, the eigen-energies for $PSh^+$ are higher while for $PAs^+$ and $PBi^+$ are lower, reflecting respectively the shallower or deeper nature of donor species $B$ with respect to $P$. Since shallower donors have larger effective Bohr radii, yielding larger wavefunction overlap, the gap at anti-crossing $\Delta_{\text{min}}$ also results to be larger for shallower donors, following the eigen-energies trend. We note that, as compared to the GaAs case given in the inset of Fig. 1, no important qualitative difference emerges for the electric field dependence of the anti-crossing gap. Of course, for the heteropolar cases, level anticrossing...
occurs at a non-zero bias field $E_c$. The energy scales for GaAs are typically one order of magnitude smaller than for Si, reflecting the difference in donor binding energies among these materials.

Figure 4 gives the gap at anticrossing versus interdonor separation along three high-symmetry crystal directions in Si, as well as the corresponding bias field $E_c$. A major difference between these results and the GaAs case is the oscillatory and anisotropic behavior of $\Delta_{\text{min}}$ vs $R$. Results for the $\text{P}^+_2$ molecular ion [solid lines in frames (a), (b) and (c)] are in excellent qualitative agreement with those reported in Ref. 12 (see Fig. 1 there). It is interesting to note that, even considering isotropic envelope functions, anisotropic behavior is obtained due to interference among the plane-wave parts (pinned at sites $\mathbf{R}_A$ and $\mathbf{R}_B$) of the six Bloch functions, corresponding to the wave vectors $\mathbf{k}_n$ at the Si conduction-band minima. The results for heteropolar donor pairs, given by the dashed and dotted lines, follow the same overall behavior as the $\text{P}^+_2$ case, showing that the oscillatory tunnel-coupling strength is inherent to the Si host, and persists in asymmetric molecular configurations. The same conclusions applicable to the $\text{P}^+_2$ case, in particular the tendency towards gap closing, remain valid here. Comparison of the solid line with the dashed and dotted lines in Fig. 4(a), (b) and (c) show that the coupling strength increases (decreases) when shallower (deeper) donors form the pair, as already noted in Fig. 2. The bias field at anticrossing is plotted in Fig. 4(d), where positive bias is arbitrarily chosen to lower the electronic potential at $\mathbf{R}_B$. Note that, for all heteropolar cases, $|E_c|$ decreases monotonically as $R$ increases, which may be understood from the resonance condition $\langle \psi_A | H_{A-B} | E_c | \psi_A \rangle = \langle \psi_B | H_{A-B} | E_c | \psi_B \rangle$: Each matrix element is taken between donor wavefunctions pinned to a single site, thus no quantum interference effects play a role, leading to the isotropic non-oscillatory behavior of $E_c(R)$ in Fig. 4(d). The resonance condition in the lowest order approximation leads to $|E_c| = |E_A - E_B|/R$, which fits the data presented in Fig. 4(d) quite well.

In this study we have shown that heteropolar donor pairs, like homopolar pairs, suffer from sensitive dependence of anticrossing gap on interdonor distance and orientation, resulting in a general narrowing of this gap, due to the valley interference in the Si conduction band. In the mean time, as summarized in Figs. 2 and 4(d), we have also shown that single electron response in a double donor system to an external electric field is not affected by the valley interference in the Si conduction band. In other words, applying an external field will not cause the tunnel coupling to exhibit oscillatory behavior as it does when inter-donor distance is varied. This conclusion is also applicable to Si double dot charge qubit, no matter whether its state structure is similar to that of a double donor in Si or double dot in GaAs. Given a fixed double-donor or double-dot geometry that has a large enough tunnel coupling, coherent charge manipulation in Si with electric field should be quite similar to that in GaAs, as recently demonstrated in Ref. 12 via electrostatic pulse sequences in a Si double quantum dot. We conclude that external electric field induced coherent manipulation of donor-based charge qubits should be experimentally possible in spite of the inter-valley quantum interference problem\textsuperscript{13,21} in silicon.

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22 This separation of variables also allows solving the problem for a pair of different donors, which is of no practical merit here since the binding energy of shallow donors in GaAs is essentially independent of the donor element.