Tunable Spin Qubit Coupling Mediated by a Multi-Electron Quantum Dot

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We present an approach for entangling electron spin qubits localized on spatially separated impurity atoms or quantum dots via a multi-electron, two-level quantum dot. The effective exchange interaction mediated by the dot can be understood as the simplest manifestation of Ruderman-Kittel-Kasuya-Yosida exchange, and can be manipulated through gate voltage control of level splittings and tunneling amplitudes within the system. This provides both a high degree of tuneability and a means for realizing high-fidelity two-qubit gates between spatially separated spins, yielding an experimentally accessible method of coupling donor electron spins in silicon via a hybrid impurity-dot system.

Single spins in solid-state systems represent versatile candidates for scalable quantum bits (qubits) in quantum information processing architectures [1–6]. In many proposals involving single-spin qubits localized on impurity atoms [2, 7] and within quantum dots [1, 8], two-qubit coupling schemes harness the advantages of tunneling-based nearest-neighbor exchange interactions: exchange gates are rapid, tunable, and protected against multiple types of noise [9–13]. These features have been demonstrated for electron spins in quantum dots [14–17], while a similar demonstration for spins localized on impurity atoms such as phosphorus donors in silicon remains an outstanding experimental challenge [6]. Although the exchange interaction originates from the long-range Coulomb interaction, its strength typically decays exponentially with distance [8, 18]. Long-range coupling via concatenation of multiple nearest-neighbor interactions is not ideal for coupling spatially separated electron spins, as it sets a low threshold error rate below which fault-tolerant quantum computing is feasible [19, 20]. A mechanism for long-range coupling that simultaneously enables scalability and robustness against errors is therefore key to realizing practical spin-based quantum information processing devices.

Approaches to implementing long-range interactions typically involve identifying a system that acts as a mediator of the interaction between the qubits, with proposed systems including optical cavities and microwave stripline resonators [21–26], floating metallic [27] and ferromagnetic [28] couplers, the collective modes of spin chains [29–31], superconducting systems [32, 33], and multi-electron molecular cores [34]. Recently, long-range coupling of electrons located in the two outer quantum dots of a linear triple dot system has been demonstrated [35, 36]. The effective exchange interaction in that system arises from electron cotunneling between the outer dots and exhibits the fourth-order dependence on tunneling amplitudes that is characteristic of superexchange [37], but suffers from a large virtual energy cost from the doubly occupied center dot states. In contrast, a many-electron quantum dot in the center can also couple distant spins via the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, with low-energy intermediate states [38], but perhaps at the cost of low fidelity as impurity-Fermi sea correlations become hard to disentangle.

Here, we show that a multi-level quantum dot containing two electrons can mediate a high-fidelity exchange interaction between two spatially separated single-electron spin qubits. We assume in what follows that the qubit electrons are localized on single-level impurity atoms, but our analysis also maps directly to the case of a triple quantum dot system with the same level structure and electron occupation. Our approach suggests an experimentally accessible method for achieving tunable coupling between donor electron spins in silicon [6, 39, 40].

Hubbard model description: The minimal model for our approach comprises a two-level quantum dot coupled to two impurities which are chosen to be near their ionization point by appropriate choice of gate voltages. This
reduces to a multi-orbital Hubbard model for a linear three-site system in the four-electron regime \cite{41,42}. We assume gate voltages can be applied to the system such that the total electron number can be set to be four, while the charge stability diagram prefers the initial configuration of (1,2,1). Here, \((n_L, n_M, n_R)\) represents the configuration with \(n_L, (n_R)\) electrons in impurity orbital \(L, (R)\) and \(n_M\) electrons in the mediator dot (Fig. 1). We work at a point in the charge stability diagram where transitions to charge configurations \((0,3,1)\) and \((1,3,0)\) are the closest available charge states, with detunings \(\Delta_J, \Delta_R\) [Fig. 1(a)].

We can write the Hamiltonian as \(H_{\text{hubb}} = H_n + H_t\), where

\[
H_n = \sum_i \epsilon_i n_i + \frac{U_i}{2} n_i(n_i-1) + \sum_{i \neq j} \frac{K_{ij}}{2} n_i n_j + J_{12} \sum_{\sigma, \sigma'} c_{i, \sigma}^\dagger c_{j, \sigma'} c_{2, \sigma'} c_{1, \sigma},
\]

\[
H_t = - \sum_{i=1,2} \sum_{\sigma} \left( t_{Li} c_{i, \sigma}^\dagger c_{L\sigma} + t_{Ri} c_{i, \sigma}^\dagger c_{R\sigma} + h.c. \right),
\]

with \(i, j = L, R, 1, 2\) denoting the left \((L)\) and right \((R)\) impurity orbital levels and the lower-energy \((1)\) and higher-energy \((2)\) orbitals of the center quantum dot [Fig. 1(b)]. \(H_n\) is diagonal with respect to the charge occupation defined by the set of eigenvalues of the electron number operators \(n_i = \sum_\sigma n_{i, \sigma} = \sum_\sigma c_{i, \sigma}^\dagger c_{i, \sigma}\), where \(c_{i, \sigma}\) creates an electron in orbital \(i\) with spin \(\sigma\). The quantity \(\epsilon_i\) denotes the on-site energy of orbital \(i\). \(U_i\) and \(K_{ij}\) are the Coulomb repulsion energies for two electrons in the same orbital \(i\) and in different orbitals \(i\) and \(j\), respectively, and \(J_{12}\) is the exchange energy for electrons in orbitals 1 and 2 of the dot with spins \(\sigma, \sigma' = \uparrow, \downarrow\).

Since we assume at most single occupancy of the impurity levels \(L\) and \(R\) and a linear geometry for the three sites, we implicitly have set \(U_L, U_R \to \infty\) and have neglected \(K_{LR}\) in Eq. (1). We also neglect the term involving \(U_2\), as configurations such as \((0,4,0)\) which involve double occupancy of orbital 2 are both high in energy and not well justified within a two-orbital picture for the dot in the presence of the Coulomb interaction. Additionally, we assume symmetric Coulomb repulsion energies between the impurities and the dot and set \(K_{Li} = K_{Ri} \equiv K_i\) for \(i = 1, 2\), while we take exchange terms \(J_{Ri} = J_{Li} = 0\), appropriate for weak tunneling. The tunneling term \(H_t\) couples subspaces of fixed charge occupation and is expressed in terms of the complex tunneling amplitudes \(t_{Li, Ri}\) between orbitals \(L, R\) and orbital \(i\) of the dot. Note that we define \(t_{Li, Ri}\) as the amplitudes for tunneling from the outer sites into the center dot and \(t_{Li, Ri}^*\) as the amplitudes for tunneling in the opposite direction [see Fig. 1(b)].

In the present work, we are interested in a system where we can effectively turn on and off the induced exchange, either by gate voltage (varying the energy difference between different charge sectors) or by tunneling. We consider our low-energy manifold to be the \((1,2,1)\) charge configuration with the center dot spins in the lowest-energy singlet. This set of states is gapped (as shown below) from other configurations by an energy large compared to typical dilution refrigerator temperatures and provides the starting point for our perturbation theory, in which we take \(H_t\) as a perturbation to \(H_n\). To further simplify the calculation, we note that the Hubbard Hamiltonian \(H_{\text{hubb}}\) conserves both the total spin \(S_{\text{tot}}\) and the total \(z\) component of spin \(S_z\) for the four-electron system. Thus, we can independently consider the two subspaces \((S_{\text{tot}} = 0, S_z = 0)\) and \((S_{\text{tot}} = 1, S_z = 0)\). Since \(H_t\) is independent of spin, the set of charge configurations generated by applying \(H_t\) to \((1,2,1)\) is identical for these two spin subspaces. Neglecting configurations which involve double occupancy of orbitals \(L, R, 2\), the intermediate charge configurations generated by \(H_t\) are \((0,3,1), (1,3,0), (1,2^*, 1)\), where \(n_M = 2^*\) denotes an excited two-electron state of the dot in which one electron is in orbital 1 and the second electron is in orbital 2 (see Fig. 2). According to Eq. (1), the energies of the \((1,2,1)\) states are identical in the absence of tunneling via the center dot and are equal to \(E_0 = \epsilon_L + \epsilon_R + 2t_1 + U_1 + 4K_1\). Choosing \(E_0\) as the energy origin, we can determine the zeroth-order energies of the intermediate states from Eq. (1). The energies of the \((0,3,1) [(1,3,0)]\) states are

\[
\Delta_{L(R)} = \epsilon_2 - \epsilon_L(R) + W,
\]

where \(W \equiv -2K_1 + K_2 + 2K_{12} - J_{12}\). The energy of each \((1,2^*, 1)\) state has one of two values, depending on the two-spin state of the center dot electrons: for the triplet and singlet states, the energies [Fig. 1(c)] are, respectively,

\[
\Delta_M = \epsilon_2 - \epsilon_1 + W - U_1 + K_2 - K_{12},
\]

\[
\Delta_J = \Delta_M + 2J_{12}.
\]

Within our toy model, the effective exchange coupling is given by the energy splitting between the states \(|(1,2,1); S_{LR}, S_{11}\rangle\) and \(|(1,2,1); T_{LR}^{(0)}, S_{11}\rangle\) in the presence of the tunneling term \(H_t\). Here, \(|S_{ij}\rangle\) and \(|T_{ij}^{(m)}\rangle\) represent two-electron singlet and triplet spin states of the electrons in orbitals \(i, j\) and \(m = 0, \pm\) indicates the spin magnetic quantum number of the triplet state. Since there is no magnetic field term explicitly present in our model, the three states \(|T_{ij}^{(0,\pm)}\rangle\) are degenerate in energy [see Fig. 1(c)] and we may choose a representative triplet state to calculate the singlet-triplet energy splitting. Extensions to large parallel magnetic field cause no changes for homogeneous \(g\) factors throughout the impurity-dot system; inhomogeneous corrections are considered at the end of this work.
We now calculate the energy shifts of the \((1,2,1)\) states due to \(H_\ell\). For \(S_{\text{tot}} = 0\), the matrix representation of \(H_{\text{hubb}}\) in the basis \(\{(1,2,1); S_{LR}, S_{11}\}, \{(0,3,1); S_{R1}, S_{11}\}, \{(1,3,0); S_{LR}, S_{11}\}, \{(1,2^*,1); T_{LR}, T_{12}, +\}\), where \(T_{LR}, T_{12}, + \equiv \left(\frac{T_{LR}^{(0)}-T_{LR}^{(+)}}{\sqrt{3}} - \frac{T_{LR}^{(-)}+T_{LR}^{(1)}}{\sqrt{3}}\right)\) is symmetric with respect to exchange of the electron spin pairs \(LR\) and \(12\), is given by

\[
H_S \equiv \begin{pmatrix}
0 & -t_{L2}^* & -t_{R2}^* \\
-t_{L2} & \Delta_L & \frac{t_{L2}}{\sqrt{2}} - \frac{\sqrt{3}t_{L1}}{2} \\
-t_{R2} & \Delta_R & \frac{t_{R2}}{\sqrt{2}} + \frac{\sqrt{3}t_{R1}}{2} \\
\end{pmatrix} \cdot (6)
\]

For the \(S_{\text{tot}} = 1\) subspace in the basis \(\{(1,2,1); T_{LR}^{(0)}, S_{11}\}, \{(0,3,1); T_{R2}^{(0)}, S_{11}\}, \{(1,3,0); T_{LR}^{(0)}, S_{11}\}, \{(1,2^*,1); T_{LR}^{(0)}, S_{12}\}, \{(1,2^*,1); S_{LR}, T_{12}^{(0)}\}, \{(1,2^*,1); T_{LR}, T_{12}, -\}\), where \(T_{LR}, T_{12}, - \equiv \left(\frac{T_{LR}^{(0)}+T_{LR}^{(-)}}{\sqrt{2}} - \frac{T_{LR}^{(+)}+T_{LR}^{(1)}}{\sqrt{2}}\right)\) is antisymmetric with respect to exchange of the electron spin pairs \(LR\) and \(12\), \(H_{\text{hubb}}\) takes the form

\[
H_T \equiv \begin{pmatrix}
0 & t_{L2} & -t_{R2}^* \\
t_{L2} & \Delta_L & \frac{-t_{L2}}{\sqrt{2}} - \frac{t_{L1}}{\sqrt{3}} \\
-t_{R2} & \Delta_R & \frac{t_{R1}}{\sqrt{2}} + \frac{t_{R2}}{\sqrt{3}} \\
\end{pmatrix} \cdot (7)
\]

Using Eqs. (6) and (7), we calculate the energy shifts of \((1,2,1); S_{LR}, S_{11}\) and \((1,2,1); T_{LR}^{(0)}, S_{11}\) up to fourth order in \(H_\ell\). We find that the first-order and third-order corrections to the energy vanish, while the second-order shifts are identical for both states. The fourth-order shifts \(\delta E_S^{(4)}\) and \(\delta E_T^{(4)}\) are therefore the lowest-order corrections that give rise to an energy splitting. The difference \(\delta E_T^{(4)} - \delta E_S^{(4)}\) is the Heisenberg exchange coupling \(J\), which we find to be given by

\[
J = -2 \left(\frac{t_{L2} t_{R1} t_{L1} t_{L2}}{\Delta_R \Delta_M \Delta_L} + \text{c.c.}\right) \cdot (8)
\]

This is the central result of our paper: using an initial singlet configuration yields an RKKY-like interaction \([43]\), including both small-energy intermediate states (\(\Delta_M\) being ‘small’ compared to the dot charging energy) and non-trivial interference terms \((J\) depends on the phases of the tunneling terms in the presence of the magnetic fields typically present in experiments).

Examining Eq. (8), we first remark that \(\Delta_J\), which differs from \(\Delta_M\) by the intradot exchange splitting \(2J_{12}\), does not appear in this expression. From the dependence of Eq. (8) on \(\Delta_L, \Delta_R, \text{and } \Delta_M\), we see that \(J\) is inversely proportional to the energy detunings \(\epsilon_2 - \epsilon_L\) and \(\epsilon_2 - \epsilon_R\) between orbital 2 of the quantum dot and the impurity orbitals as well as to the on-site energy difference \(\epsilon_2 - \epsilon_1\) between the two levels of the quantum dot. As the detunings can be controlled via the voltages applied to the dot and have a lower limit set only by the tunnel coupling and magnetic field magnitudes, the strength of the exchange coupling mediated by the two-level dot is highly tunable and may be made large. This is in contrast to keeping a large detuning to suppress sequential tunneling \([35, 36]\), which limits the maximum achievable coupling strength.

We now turn to the phase dependence in Eq. (8). The terms correspond to two alternative pathways for the electrons which give rise to the effective coupling \(J\) (Fig. 2); thus, the interaction can have interference between these pathways, and their non-trivial relative phase for finite magnetic fields leads to an interaction strength that depends on the tunneling phase factors \([43]\). This provides a glimpse of the beginning of the expected sign fluctuations in exchange for a true RKKY interaction, where the finite Fermi wave vector \(k_F\) of the two-electron Fermi ‘sea’ matters. We note that for phosphorus donor electrons in silicon, the tunneling amplitudes also oscillate rapidly with the donor positions due to interference between electronic states associated with different degenerate minima, or valleys, existing in the conduction band \([44, 45]\). This can be seen by taking \(t_{ij} \propto \langle \psi_i | \psi_j \rangle\) for \(i = L, R\) and \(j = 1, 2\), where \(\psi_{1,2}\) are superpositions of orbital wave functions associated with each valley. The resulting sinusoidal dependence of the tunneling ampli-
tudes on the positions of the donors relative to the dot center leads to a strong dependence of the terms in Eq. (8) on these relative positions.

**Charge noise and exchange gate fidelity:** Fluctuating electric fields introduce variations in the parameters determining the effective exchange \( J \) in Eq. (8) and consequently affect the operation of exchange-based gates [1, 46, 47]. Here, we consider the effects of classical charge noise on the detuning parameters \( \Delta_n \) for \( \alpha = L, M, R \) and calculate the fidelity of the exchange gate \( \hat{U}(\tau) = \exp(-i\hat{H}_\text{exch}\tau) \), where \( \hat{H}_\text{exch} = -J\langle S_{LR, S_{11}} \rangle \langle S_{LR, S_{11}} \rangle \) and \( \langle S_{LR, S_{11}} \rangle \) is the corrected state after elimination of states outside the \((1, 2, 1)\) subspace (note that we suppress the charge state in this notation, since the effective Hamiltonian acts only in this subspace). Letting \( \Delta_n \rightarrow \Delta_n + \delta_n \), where \( \delta_n \) represents small fluctuations about the average detuning \( \Delta_n \), and expanding to first order in \( \delta_n \) gives \( J \rightarrow J' = J(1 - \sum_n \delta_n / \Delta_n) \). We assume that the fluctuations \( \delta_n \) are independent and described by Gaussian distributions \( \rho_\delta(\delta_n) = e^{-\delta_n^2/2\sigma_n^2} / \sqrt{2\pi}\sigma_n \) with charge noise standard deviations \( \sigma_n \). The average of the exchange gate over these fluctuations is then given by \( \langle e^{iJ'\tau} \rangle = 1 + \left(e^{iJ\tau} - 1\right) \langle S_{LR, S_{11}} \rangle \langle S_{LR, S_{11}} \rangle \), where \( \langle \psi \rangle \) is the corrected state after elimination of charge noise due to screening of the Coulomb interaction by the paired “core” electrons already present in the dot. Varying the number of electrons in the dot changes the spacing between the outermost levels [3] and consequently \( \Delta_M \), so that \( J \) may be tuned in discrete steps. Provided this discrete level description remains valid, the larger sizes associated with multi-electron dots may also enable longer-range coupling.

**Effects of inhomogeneous g factors:** In the presence of an external magnetic field, a difference in the g factors of the impurities and the quantum dot couples the \( S_{tot} = 0 \) and \( S_{tot} = 1 \) subspaces. To investigate the form of this coupling, we assume an applied magnetic field \( \mathbf{B} = B_z \hat{z} \) and add a magnetic gradient term of the form

\[
H_Z = \frac{\Omega_z}{2} \sum_{i=1,2} (n_{i\uparrow} - n_{i\downarrow})
\]

to the Hubbard Hamiltonian [Eqs. (1) and (2)], where \( \Omega_z \equiv \Delta_g e \mu_B B_z \) is the magnetic field splitting due to a g-factor gradient \( \Delta g_z \) parallel to the external field [see Fig. 1(c)]. The full Hamiltonian is then given by \( \hat{H} = \hat{H}_\text{Hub} + H_Z = \hat{H}_n + \hat{H}_f + H_Z \) and acts in the combined space consisting of both the \( S_{tot} = 0 \) and \( S_{tot} = 1 \) subspaces. We transform to a basis which diagonalizes \( \hat{H}_0 \equiv \hat{H}_n + H_Z \) and treat \( \hat{H}_f \) as...
a perturbation to \( H_0 \). Keeping terms up to second order in the tunneling amplitudes and up to linear order in \( \Omega \), we find that the correction to the effective exchange Hamiltonian \( H_{exch} \) is given by

\[
H_g = f_g \left( \langle T_{LR}^{(0)} | S_{11} \rangle \langle S_{LR}, S_{11} | + \langle S_{LR}, S_{11} \rangle \langle T_{LR}^{(0)} | S_{11} \rangle \right),
\]

where

\[
f_g = \frac{\Omega}{2} \left( \frac{|t_{L2}|^2}{\Delta_L^2} - \frac{|t_{R2}|^2}{\Delta_R^2} \right).
\]

(11)

From this expression, we see that the effects of the \( g \) factor inhomogeneity described by Eq. (10) can be eliminated up to first order in \( \Omega \) and second order in the tunneling amplitudes by choosing \( t_{L2}, t_{R2}, \Delta_L \) and \( \Delta_R \) such that the constraint \( \Delta_L^2/\Delta_R^2 = |t_{L2}|^2/|t_{R2}|^2 \) is satisfied. Note that the preceding analysis assumes \( \Omega < \Delta_{M,L,R} \), which sets an upper bound on \( J \) [see Eq. (8)]. For impurity atoms with nonzero nuclear spin, hyperfine coupling represents an additional source of magnetic gradients between the impurity and dot electrons that may prove useful for alternative coupling schemes. Indeed, for direct exchange coupling between two donor electron spins in silicon, recent work [54] shows that a difference in the effective coupling between the donors enables two distinct methods for realizing high-fidelity two-qubit gates.

The validity of the toy model for the effective exchange coupling considered in the present work is limited by the validity of the two-level approximation for the mediator quantum dot in the presence of the Coulomb interaction among the four electrons. Future work should consider a detailed calculation of the effective exchange interaction mediated by the two-level quantum dot in terms of the general form of the pairwise Coulomb interaction and explore how this analysis may be extended to gain insight into the form of the coupling mediated by a quantum dot with more than two levels.

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