Impurity effects on semiconductor quantum bits in coupled quantum dots

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We theoretically consider the effects of having unintentional charged impurities in laterally coupled two-dimensional double (GaAs) quantum dot systems, where each dot contains one or two electrons and a single charged impurity. Using molecular orbital and configuration interaction method, we calculate the effect of the impurity on the 2-electron energy spectrum of each individual dot as well as on the spectrum of the coupled-double-dot 2-electron system. We find that the singlet-triplet exchange splitting between the two lowest energy states, both for the individual dots and the coupled dot system, depends sensitively on the location of the impurity and its coupling strength (i.e. the effective charge). A strong electron-impurity coupling breaks down equality of the two doubly-occupied singlets in the left and the right dot leading to a mixing between different spin singlets. As a result, the maximally entangled qubit states are no longer fully obtained in zero magnetic field case. Moreover, a repulsive impurity results in a triplet-singlet transition as the impurity effective charge increases or/and the impurity position changes. We comment on the impurity effect in spin qubit operations in the double dot system based on our numerical results.

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I. INTRODUCTION

The goal of this work is the calculation of the low-lying energy spectra of 2-electron semiconductor quantum dots (QDs) (both single dots and coupled double-dots) in the presence of nearby static charged impurity centers within a minimal model. The purpose is to quantify the effects of quenched random charged impurities on the singlet-triplet splitting in QDs in order to assess the importance of background unintentional impurities, which are invariably present in the environment, in adversely affecting the operations of exchange-coupled dots as elementary spin qubits for solid state quantum computation. Since the positions and the strengths of the background unintentional impurities are unknown and random, we study the impurity effects as functions of impurity position and coupling strength (defined as the effective impurity charge $Z$) assuming the impurities to be Coulombic charge centers so that their effective potential falls off slowly as a $1/r$ potential away from the impurity location where ‘$r$’ is the distance from the impurity. Since the main background impurities in GaAs and Si, the two most relevant semiconductors of interest for solid state spin quantum computation, are random charge centers, our consideration of Coulombic impurities with a long-range impurity potential is reasonable. The theoretical results presented herein, while being completely microscopic and fully quantum mechanical, are phenomenological in nature since the impurity position and charge are treated as unknown parameters. While our results show clearly the strong effect of local background charged impurities on the low-lying QD spectra, its usefulness is limited in comparing with experiment since no direct information about impurity locations is currently available experimentally. On the other hand, our results establish the manifest importance of using the highest quality background materials for semiconductor spin qubit operations since the presence of even a single charged impurity in the vicinity of the QDs seems to completely ruin the operational logistics of QDs. The exchange coupling (or equivalently, the singlet-triplet splitting) depends strongly and sensitively on the location and the strength of the charged impurity, which means that (1) a single charged impurity located nearby could destroy the qubit, and (2) even a remote charged impurity could have a strong adverse effect inducing substantial qubit decoherence if the impurity is dynamic and has a fluctuation timescale comparable to gate operations timescales—in fact, the impurity fluctuation timescale will become a dominant decoherence time since the exchange energy will vary substantially over this timescale. The motivation of our work is a clear understanding of the energetics of QD systems in the few electron situation in the presence of charged impurities so that some rudimentary quantitative magnitudes of the impurity effects are available for qubit operation considerations.

Coupled QDs for quantum computation\cite{1,2,3,4,5} have been extensively studied due to the prospect of using QDs as ideal environment to confine and manipulate the QD electron spins. The quantum bit, or qubit, of information is encoded and stored in these localized single electron spins which exploit a spin relaxation time of the order of milliseconds\cite{2,4,5}, sufficiently long to allow the performance of coherent spin operations. The proposed quantum computer\cite{4} in solid states operates based on the exchange coupling $J$ between the two electron spin qubits manipulated by an external magnetic field. This exchange energy can be envisioned as the effective coupling between the two spins in the double dot via the Heisenberg spin Hamiltonian, $\hat{H} = J \mathbf{s}_1 \cdot \mathbf{s}_2$, which takes into account possible contributions from different hybridization between singlet and triplet states. Hence, $J$ is determined through the gate voltage control over...
the tunneling coupling between the coupled two QDs. A complete understanding of $J$ is important because it directly determines the $\sqrt{\text{SWAP}}$ operator which describes the exchange information between the two qubits in the double dots. The fast solid-state two-qubit operation\cite{26,27}, generated as a consequence of the electron spin exchange under electrical control, and the single-qubit operation\cite{28} suffice to assemble a standard quantum computing system. The number of electrons in such a QD system can be controlled precisely\cite{29,30} to 0, and the electron exchange interaction is tunable by the applied gate voltages. Thereby, the coupling between the dots can also be controlled.

Established quantum regimes such as quantum entanglement between individual electrons in one dot with the other electrons in the other dots and superposition of electron spin qubits are the major objects\cite{22,23} in such exchange coupled QD system. Prerequisite criteria for realization of a quantum computation system\cite{6} (including initialization, manipulation of spin qubits, and readout) have been demonstrated for single dot\cite{31} and coupled QD system\cite{32}, provided the long relaxation time\cite{33} of the spins, by using charge sensing and fast spin-to-charge conversion techniques. One of the perceived advantages of solid-state quantum computing is the scalability with existing semiconductors. In addition, integrating multi coupled dots into a quantum circuit is made possible by adding more suitable gate electrodes\cite{34,35}.

QDs in versatile GaAs semiconductors not only have been considered the most widely-studied objects in the QD science but also their well-understood physics are applicable to a variety of materials.\cite{2} Often, unintentional impurities found in the dot sample are used in the fabrication process to adjust the potential well height between different heterostructure semiconductors (such as Si in gated GaAs/AlGaAs QDs - see e.g. Ref.\cite{36}) allowing the charge flow of electrons. Statistically, such QD system containing impurities can be studied for one, two, ... impurities. On the other hand, when integrating multiple coupled 2-dot systems utilized as a multi-qubit gate, in the non-overlapping regime between different 2-dot systems, a single spin qubit in one coupled 2-dot system can be affected\cite{37} by the other coupled 2-dot systems. Each coupled 2-dot system acts as a source of electrostatic field to the others and can be treated as “charged impurities’’.

Theoretical studies of impurities in coupled QDs have been rarely found in the literature. In fact, the impurities are practically found randomly in/outside the dot sample and their positions cannot be specified precisely. In a coupled triple-dot system, a relatively large collection of impurities was considered\cite{38}. These statistical impurities were theoretically assumed to induce a weak perturbation to the coupled triple QDs. The authors\cite{39} found that the Coulomb exchange energy of the impurities with the QD electrons in this study resulted in decoherence of the coded qubit states. Thus, any information processing using electron charge degree of freedom needs to take into account the decoherence channel due to charge fluctuations.

In the present paper, we study the influence of charged impurities on the singlet-triplet splitting of the two lowest energy levels as well as the energy spectrum as a function of the impurity position, the impurity effective charge, and the number of impurities for a coupled two-dot system in zero magnetic field. We also examine the impurity effect on the coupled electron qubits by tuning the potential well height to different values and obtain different triplet-singlet transitions for the repulsive impurity case. To accomplish, we discuss the singlet-triplet splitting in the presence of two impurities with similar charge located in the two separate dots of the system.

The paper is organized as follows. In Sec. II we introduce the model and methodology. Impurity effect on energy spectrum of a two-electron single QD containing a single charged impurity is first discussed in detail in Sec. III. Sect. IV is spent to present our studies on a coupled 2-dot system in the presence of one or two charged impurities. We examine the impurity position and impurity effective charge dependence of the singlet-triplet spin splitting energy. Influence of the confining potential barrier height on the electron-impurity (e-I) and electron-electron (e-e) coupling is also explored. All the results presented in this paper are obtained at zero magnetic field, $B=0$. Summary of our results and conclusion are found in Sec. V.

II. THEORETICAL MODEL

Horizontally-coupled QDs are grown by the depletion of the two-dimensional electron gas (2DEG) using typically the gated mechanism\cite{40,41}. Such gated QDs have typical size of about few tens of nm. Consequently, the lowest excitation energy in such a QD system is found of the order of few meV. When the inter-dot coupling strength is substantial, the electrons in the coupled dots strongly quantum mechanically couple with each other. As long as the phase is coherent, the electrons can “tunnel” between the adjacent dots forming different entangled qubits. Using single-conduction-band effective-mass approximation which was justified in Ref.\cite{42}, the Hamiltonian describing a two-electron coupled double QD system containing unintentional charged impurities (charge size Ze) in zero magnetic field reads:

$$\hat{H} = \hat{h}_I + \hat{h}_C + \hat{V}_\text{e-Imp} + \hat{V}_\text{I-I}.$$\hspace{1cm} (1)

Here, the first two terms $\hat{h}_{i,j}$ are the single-particle Hamiltonian of the two quantum-dot electrons (coordinates $\mathbf{r}_i$)

$$\hat{h}_i = \frac{(i\hbar \nabla_i + e\mathbf{A}_i)^2}{2m^*} + V(\mathbf{r}_i)$$\hspace{1cm} (2)

confined by a potential well $V(\mathbf{r})$. In the present paper, this confining potential is constructed as a linear
combination of the three different Gaussians and can be separated into two parts:

\[
V(r) = V_0(e^{-(x-a_m)^2/2a_m^2} + e^{-(y-a_m)^2/2a_m^2}) + V_b e^{-\frac{r^2}{2a_m^2}}.
\]

The first part acts as a double-well confining potential for the coupled double dot system and the second part is used to control the electrostatic barrier height between the two dots independently. The set of varying parameters \(V_0, V_b, l_x, l_y, l_{bx}, l_{by}\) characterizes the potential well depth and barrier height. A nonvanishing overlap between the wave functions of the two quantum dots signifies the electron virtual tunneling between the two dots, i.e. the exchange energy is nonzero. Because this property can be tuned by the applied gate voltage this exchange energy thereby can be controlled. It is worth noting that \(V_b\) can independently modulate the barrier height resulting in concomitant change in the inter-dot separation without modifying the single-particle energy spectrum of the individual QDs in the system. The last three terms in Eq. (11) are the Coulomb interaction between, respectively, the two electrons

\[
\hat{V}_C = \frac{e^2}{4\pi\epsilon\varepsilon_0|R_{ij}|},
\]

the electron and the impurities

\[
V_{e-Imp} = \sum_{k=1}^{N_I} \sum_{l=1}^{N_e} \frac{Z_k e^2}{4\pi\epsilon\varepsilon_0|R_k - r_l|},
\]

where \(N_I\) and \(N_e\) denote the number of impurities and electrons, respectively, and the impurities

\[
V_{l-I} = \frac{Z_1 Z_2}{4\pi\epsilon\varepsilon_0|R_1 - R_2|}.
\]

The solution of a single particle confined by a parabolic potential is well known as the Fock-Darwin basis:

\[
\varphi_{nl}(r) = \frac{1}{l_0} \sqrt{\frac{n!}{\pi(n+|l|)!}} \left( \frac{r}{l_0} \right)^{|l|} e^{-\frac{r^2}{l_0^2}} L_n^{|l|} \left( \frac{r^2}{l_0^2} \right) (7)
\]

with corresponding energy

\[
E_{n,l} = \hbar \omega_0 (2n + |l| + 1)
\]

where \((n, l)\) stand for radial and azimuthal quantum number, respectively. \(l_0\) is the confinement length which is defined via the confinement frequency \(\omega_0\): \(l_0 = \sqrt{\frac{\hbar}{m^* \omega_0}}\). Using this basis as the radial part we can construct the many-body wave function for the considered system.

We introduce a dimensionless parameter,

\[
\lambda = \frac{l_0}{a_B}
\]

with \(a_B = \frac{4\pi\epsilon\varepsilon_0 \hbar^2}{m^* e^2}\) the effective Bohr radius, which is used to describe the relation between the effective Rydberg energy \(R_y^* = \frac{m^* e^4}{2\pi^2 \epsilon^2 \epsilon_0 l_0^2}\) and the confinement energy \(\hbar \omega_0\)

\[
R_y^* = \hbar \omega_0 \cdot \lambda^2 = 2.
\]

Both e-e and e-I Coulomb interaction are evaluated in terms of matrix elements as

\[
\langle \Psi | V_{e-e} | \Psi \rangle = V_0^C \langle \Psi | \frac{1}{\vec{r}_i - \vec{r}_j} | \Psi \rangle (11)
\]

\[
\langle \Psi | V_{e-Imp} | \Psi \rangle = V_0^C \langle \Psi | \frac{1}{\vec{R} - \vec{r}} | \Psi \rangle (11)
\]

where \(\Psi\) denotes the wave function of the system, \(\vec{r}_{i,j} = \vec{r}/l_0, \vec{R} = \vec{R}/l_0\), and \(V_0^C = e^2/4\pi\epsilon\varepsilon_0 l_0^2\) is the Coulomb energy unit. \(V_0^C\) relates to the confinement energy via the relation:

\[
V_0^C = \frac{e^2}{4\pi\epsilon\varepsilon_0 l_0^2} = \lambda \cdot \hbar \omega_0.
\]

The numerical results are implemented for e.g. a GaAs QD with \(m^* = 0.067m_0, \epsilon = 13.1, g_e = -0.44, R_y^* = 5.31\) meV (corresponding to \(a_B^* = 10.3\) nm). \(\lambda\) is changed by changing the confinement energy \(\hbar \omega_0\). \(\lambda = 1\) gives \(\hbar \omega_0 = 2R_y^* = 10.62\) meV and corresponding confinement length \(l_0 = a_B^* = 10.3\) nm.

For Si/SiGe (\(\epsilon = 13\)) and Si/SiO\(_2\) (\(\epsilon = 6.8\)) quantum dots with a heavier effective mass \(m^* = 0.19m_0\), effective
Bohr radius and effective Rydberg energy are, respectively, \( a_b^\pi = 4 \text{ nm} \) and \( R_b^\pi = 15 \text{ meV} \), and \( a_b^\ell = 2.11 \text{ nm} \) and \( R_b^\ell = 44.76 \text{ meV} \) (see e.g. Ref.\(^{20}\)). \( \lambda = 1 \) gives \( h\omega_0 = 2R_b^\ell = 30 \text{ meV} \) \((l_0 = 4 \text{ nm})\) for a Si/SiGe QD and \( h\omega_0 = 2R_b^\ell \approx 90 \text{ meV} \) \((l_0 = 2.11 \text{ nm})\) for a Si/SiO\(_2\) QD system. Even though our numerical results are applicable to GaAs QD systems, singlet-triplet splitting energies for Si QD systems are also provided in Appendix D for comparison purposes.

Reducing \( \lambda \) means that the effective length, \( l_0 \), of the QD decreases while the energy spacing, \( h\omega_0 \), between the 2D shells, i.e. the \( s-, p-, d-\) levels, will increase. In the small \( \lambda \) limit, the problem at hand converts to the problem of independent particles. In the opposite case, very large \( \lambda \), the problem approaches the classical situation.

In the coupled double QD system, the single-particle solutions in each dot are obtained approximately based on an assumption that around the center of the each dot \((\pm a_m, 0)\) the single-electron problem can be treated as a 2D harmonic oscillator. This means that the confining potential well \( V(r) \) performs a quadratic form \( V(r) = V_0 \approx \frac{\hbar^2}{2m}(x^2 + y^2) \), with \( E_m \) the bottom energy of the potential well, around \((\pm a_m, 0)\). Changes in \( E_m \) when \( V_b \) varies can be obtained as the open circles in Fig.\(^1\) The single-particle wave functions now are identical to the Fock-Darwin levels centered at \((\pm a_m, 0)\) and the single-particle energy spectrum is shifted by an amount of \( E_m \).

\[
\varphi_{L(R)}(r) = \frac{1}{l_0^{n!}} \sqrt{\frac{n!}{\pi (n+|l|)!}} \left( \frac{r_{L(R)}}{l_0} \right)^{|l|} e^{-l \theta} \quad (13)
\]

where \( r_{L,R} = (x \pm a_m, y) \) and \( \omega_0 = \sqrt{\frac{2V_0}{m\cdot l_0^2}} \) is the quadratic confining frequency which defines a new length \( l_0^2 = \frac{\hbar}{m\omega_0} \) called the confinement length. The single-particle ground-state wave function is

\[
\varphi_{L,R} = \frac{1}{\sqrt{\pi l_0}} e^{-\frac{[(x \pm a_m)^2 + y^2]}{2l_0^2}}. \quad (14)
\]

To quantitatively evaluate the advantage of using the above confining potential present in Eq. (3), we plot in Fig.\(^1\) the confining potential well where its barrier height and the QD centers are modified by changing \( V_b \). Here, \( V_b \) is reduced from 35 meV to 30, 25, 20, 15, and 11.4 meV. As a result, the barrier height will decrease making the electron exchange energy increased. For example, the system with \( V_b = 35 \text{ meV} \) has the corresponding \( a_m \approx 17.52 \text{ nm} \) \((\approx 1.75l_0)\), and barrier height \( \Delta V_b = 13.26 \text{ meV} \). For \( V_b = 30 \text{ meV} \), these parameters are \( a_m \approx 16.41 \text{ nm} \) \((\approx 1.64l_0)\) and \( \Delta V_b = 9.65 \text{ meV} \). Decreasing \( V_b \) leads to a shortened inter-dot separation 2\( a_m \) and a smaller \( \Delta V_b \). Details can be obtained in Fig.\(^1\)

In our numerical results, we use \( V_b = 30 \text{ meV} \) for most of our calculations except when we examine the barrier height dependence of the exchange energy \( J \) where \( V_b \) can vary. The center region of each QD, however, has unchanged effective length, namely \( l_0 \), regardless to the change in the barrier height. The other parameters taken after Refs.\(^2\)\(^{22}\)\(^{23}\) are \( l_x = l_y = 30 \text{ nm} \), \( a = l_b = l_x/2 = 15 \text{ nm} \), \( b_y = 80 \text{ nm} \).

We assume that the impurities are located arbitrarily in or outside the coupled QDs. Their coordinates are \( x_{k}(y, z) \), \( k=1,2 \). Theoretically, the effective coupling between electron and the localized impurities as well as the coupling between the impurities with each other can be tuned by engineering the impurity charge \( Z \).

Configuration interaction (CI) and molecular orbital (MO) method are used to numerically solve the Hamiltonian Eq. (1). Both construct the total wave function of the system as a superposition of different possible quantum configurations (CI) or molecule states (MO) extended in the basis of single-particle wave functions:

\[
\Psi(r_1, r_2) = \sum_{i} \psi_i(r_1, r_2) \quad (15)
\]

where \( \psi_i(r_1, r_2) \) represents one many-electron configuration as a Slater determinant. Each term of this Slater determinant is a single-electron wave function consisted of the radial part, the Fock-Darwin state \( \varphi(r_1, r_2) \) defined in Eq. (13), and the electron spin part (detailed justifications are referred to Ref.\(^{22}\)).

The singlet-triplet spin splitting energy of the electrons \( J \) in such a coupled 2-dot system is defined as the energy difference between the two lowest singlet (\( \Psi^S \)) and triplet (\( \Psi^T \)) states:

\[
J = \langle \Psi^T | \hat{H} | \Psi^S \rangle - \langle \Psi^S | \hat{H} | \Psi^T \rangle. \quad (16)
\]

### A. Hund-Mulliken

In Hund-Mulliken model, the energy spectrum consists of four levels which are four possible superpositions of the four basis (three singlets and one triplet) wave functions

\[
\psi^S(r_1, r_2) = \frac{1}{\sqrt{2}}[\varphi_L(r_1)\varphi_R(r_2) + \varphi_L(r_2)\varphi_R(r_1)], \quad (17)
\]

\[
\psi^S(r_1, r_2) = \varphi_L(r_1)\varphi_R(r_2)
\]

\[
\psi^S(r_1, r_2) = \varphi_R(r_1)\varphi_L(r_2)
\]

\[
\psi^T(r_1, r_2) = \frac{1}{\sqrt{2}}[\varphi_L(r_1)\varphi_R(r_2) - \varphi_L(r_2)\varphi_R(r_1)].
\]

In the coupled QD system without an impurity, these three singlets do not couple with the maximally entangled triplet state \( \psi^T \) therefore they can be treated separately. The entire Hamiltonian matrix represents these singlets and triplets as independent blocks.
B. Configuration Interaction

The most difficult task in finding the eigenvalues of a coupled 2-dot system lies in the basis choice among which Fock-Darwin basis and Gaussian basis are the most widely used. However, in both cases a closed analytical form for the Hamiltonian matrix elements, essentially the e-e Coulomb matrix elements, has not yet been obtained. The reason is that the single-particle solutions of different dots i) have distinct zero-points shifted to the two bottoms of the confining potential well and ii) thus are not orthogonal with each other. Consequently, the number of distinguishable single-particle quantum states for the double-dot system will be doubled. Effectively, the size of the entire Hamiltonian increases in comparison to the single dot case. Quantitatively, for $S_z = 0$ subspace, such a number is 4 times larger than that of the single dot problem ($2N \times 2N$). Specifically, if only the $s$-waves are taken into account, the number of configurations in the subspace $S_z = 0$ is 1 for single QD and 4 for coupled 2-dot system and if the $s$- and $p$-waves are included those numbers are 9 and 36, respectively.

The above fact poses much difficulty to solving the eigenvalues of the coupled QD Hamiltonian. The most time consuming part is spent to calculate the Coulomb matrix elements. In the problem at hand, the number of Coulomb elements increases due to the e-I exchange interaction. However, based on the following facts one can take out nonphysical excited single-particle quantum states: i) the form of the confining potential, which is tuned electrostatically by metal top gates, is not exactly known and ii) the barrier potential height is finite. The latter factor validates the harmonic approximation for the confinement potential double well resulting in only a limited number of Fock-Darwin states involved.

III. IMPURITY EFFECTS IN SINGLE QUANTUM DOTS

In the single QD case, we assume that there is only one impurity and the impurity is located along $z$-axis, i.e. $\vec{R} = (0, 0, d)$. The addition Coulomb interaction of the electrons with the impurity is obtained analytically (see Appendix A):

$$V_{n_1l_1}^{n_2l_2}(\vec{R}) = \delta_{l_1, l_2} \frac{2}{\sqrt{n_1!n_2!}} \frac{\Gamma(n_1 + l_1 + 1) \Gamma(n_2 + l_2 + 1)}{\Gamma(n_1 + l_1 + 1)! \Gamma(n_2 + l_2 + 1)!} \times$$

$$\sum_{j=0}^{n_1} \sum_{k=0}^{n_2} (-1)^j \Gamma(l_1 + j + 1) \Gamma(l_2 + k + 1) \times$$

$$\text{erfc}(\frac{n_1 - l_1 + 1}{2}) I_{n_1 - l_1}(\vec{R})$$

$$+ \sum_{j=0}^{n_1} \sum_{k=0}^{n_2} (-1)^k \Gamma(l_1 + j + 1) \Gamma(l_2 + k + 1) \times$$

$$\text{erfc}(\frac{l_2 + 1}{2}) I_{l_2 + 1}(\vec{R})$$

$$+ \sum_{j=0}^{n_1} \sum_{k=0}^{n_2} (-1)^{j+k} \Gamma(l_1 + j + 1) \Gamma(l_2 + k + 1) \times$$

$$\text{erfc}(\frac{n_1 + l_1 + 1}{2}) I_{n_1 + l_1}(\vec{R}) + \text{erfc}(\frac{l_2 + 1}{2}) I_{l_2 + 1}(\vec{R})$$

where $l^+ = l_1 = l_2$ and

$$I_m(\vec{R}) = \int_0^\infty \frac{e^{-\vec{R}^2 u^2}}{1 + u^2} du$$

Integral (19) can be obtained through the recurrence:

$$I_0(\vec{R}) = \frac{\pi}{2} e^{\vec{R}^2} \text{erfc}(\vec{R})$$

$$I_1(\vec{R}) = \frac{1}{2} [1 - 2 \text{erfc}(\vec{R})] I_0(\vec{R}) + \sqrt{\pi} \vec{R}$$

$$I_m(\vec{R}) = \frac{-\vec{R}^2}{m} \frac{1}{m} I_{m-1}(\vec{R}) + \frac{\vec{R}^2}{m} I_{m-2}(\vec{R})$$

where erfc($x$) is the complementary error function,

$$\text{erfc}(x) = \int_x^\infty \frac{e^{-t^2}}{1 + t^2} dt$$

which rapidly decreases in $x$. $(x)_n = x(x + 1)(x + 2)...(x + n - 1)$ is the Pochhammer function. Several values of the complementary error function which can be obtained in any numerical library are: erfc(0) = 1, erfc(0.01) ≈ 0.998872, erfc(0.1) ≈ 0.88754, erfc(0.5) ≈ 0.47950, erfc(1)≈ 0.1573, erfc(2)≈ 0.00468, etc.

Following the e-I interaction whose matrix elements are expressed in Eq. (18), there is mixture between different states with different total radial quantum numbers. Consequently, the ground-state wave function e.g. for $N_e = 1$ electron is no longer the only one $s$- $(n=0,l=0)$ quantum state. Instead, it consists of several quantum states whose contributions $C_i$ are different

$$\Psi = \sum_{i=1}^{N_e} C_i \psi_i$$

For $N_e = 2$ electrons without impurity, by increasing $\lambda$ results in the ground state mixing of different configurations. The two electrons will occupy higher Fock-Darwin states with decreasing energy spacing to lower their Coulomb repulsion. The easiest way to check the accuracy of the numerical results when the impurity is present is to consider the limitation when weak perturbation works perfectly. The first-order perturbation ap-
proximation:

\[ E = E_0 + Z \sum_{i=1}^{N_e} \langle \Psi_0 | V(|r_i - R|) | \Psi_0 \rangle + O(Z^2) \]  

(23)

where \( E_0 \) is the GS energy of the QD with the ground-state wave function \( \Psi_0 \) in case without impurity is a good one when \( Z < 1 \). For the sake of simplicity in evaluating the perturbative part in Eq. (23) for both \( N_e = 1 \) and 2 electrons, we assume that \( \Psi_0 \) in case \( Z = 0 \) is a single term: the \( s \)-state. For \( N_e = 1 \) electron, this is always satisfied and its radial part is

\[ \Psi_0(r) = \frac{1}{l_0 \sqrt{\pi}} e^{-\frac{r^2}{4l_0^2}}. \]  

(24)

The situation changes for \( N_e = 2 \). The above condition for \( \Psi_0 \) to be a single term is only satisfied when \( \lambda < 1 \). If so, the two electrons obeying Pauli exclusion principle with spins antiparallel will mainly stay in the \( s \)-orbital in case no impurity is present because their Coulomb repulsion is small as compared to the confining energy. The ground-state wave function is

\[ |\Psi_0 \rangle = c_{s \uparrow}^\dagger c_{s \downarrow}^\dagger |0\rangle. \]  

(25)

Using this assumption, the total energy is estimated theoretically for the single-electron QD:

\[ E_{(N_e=1)} = E_{0(N_e=1)} + 2\sqrt{\pi}Z\lambda \hbar \omega_0 e^{\tilde{R}^2} \text{erfc} \left( \frac{\tilde{R}}{\sqrt{2}} \right) + O(Z^2) \]  

(26)

and the two-electron QD:

\[ E_{(N_e=2)} = E_{0(N_e=2)} + 4\sqrt{\pi}Z\lambda \hbar \omega_0 e^{\tilde{R}^2} \text{erfc} \left( \frac{\tilde{R}}{\sqrt{2}} \right) + O(Z^2). \]  

(27)

We plotted in Fig. 2 the energy shift due to the presence of the impurity as a function of \( d \) using the above theoretical estimations (red dash-dotted) and the numerical results (black solid) for \( Z = 0.1 \) and \( \lambda = 0.1 \) (\( l_0 = 1.03 \text{nm} \) and \( \hbar \omega_0 = 200\tilde{R}_0^2 = 1.062 \text{eV} \)) for \( N_e = 1 \) (upper curves) and 2 (lower curves) electrons. Both theoretical and numerical results are in good agreement.

A small comment is made in case \( d \) is large. As seen from Fig. 2 the e-I interaction goes to zero slowly when \( d \) increases to a relatively large value, say \( d > a_B^* \). The answer lies in the product \( e^{\tilde{R}^2} \cdot \text{erfc} \left( \frac{\tilde{R}}{\sqrt{2}} \right) \) where the exponential function increases in \( d \) competitively with the complementary error function which decreases in \( d \).

The first-order perturbation theory works very well as long as \( Z < 1 \) and \( \lambda \) is small enough. Because if \( \lambda \) increases different e-I couplings of different configurations will occur which result in the presence of a substantial number of nonzero off-diagonal terms. This fact leads to an increasing difference between the first-order calculations and the numerical results. For example, in the two-electron QD as plotted in the inset of Fig. 2, the e-I interaction goes to zero slowly when \( d \) increases to a relatively large value, say \( d > a_B^* \). The answer lies in the product \( e^{\tilde{R}^2} \cdot \text{erfc} \left( \frac{\tilde{R}}{\sqrt{2}} \right) \) where the exponential function increases in \( d \) competitively with the complementary error function which decreases in \( d \).

The Coulomb interaction unit (\( V_0^C = 5.31 \text{ meV} \)) in such a QD system is (2 times) larger than the confining energy \( \hbar \omega_0 \).

**B. Singlet-triplet splitting energy**

The Coulomb interaction between the QD electrons strongly competes with the confining energy and with the e-I interaction as \( Z \) and/or \( \lambda \) increase. Increasing \( \lambda \) means that the confining energy becomes smaller with respect to the Coulomb interaction. As a result, electrons start to occupy higher Fock-Darwin levels. At zero B-field and in the absence of impurity, the ground state consists of several Fock-Darwin states where the dominant component is the \( s \)-wave term. In the presence of a charged impurity, those electron configurations that
fulfill $L=\text{const}$ form different $L$-subgroups with nonzero contributions to the total wave function of the system. Increasing the effective charge $Z$ results in a strong mixing between those subgroups. Consequently, there are more than 1 state which play as dominant components to the total wave function. We plot in Figs. 3(a) and (b), respectively, the splitting energy between the ground state (singlet) and the first excited state (triplet) as a function of $\lambda$ and $Z$ in case the impurity is located at the center of the single QD. As $\lambda$ increases [see Fig. 3(a)], the splitting becomes smaller for both with and without impurity cases. It is because the singlet and the triplet states have many similar nonzero configurations. Details are found below from the discussion for particular values of $Z$. From Fig. 3(a) we also notice that the energy splitting shift due to the presence of the impurity remains almost unchanged by changing $\lambda$ for both cases $Z = -0.1$ (blue dashed dot) and $Z = 0.1$ (red dash-dotted-dot). It is because the impurity location is examined at the center of the QD $d=0$. Such splitting shift will change if the impurity is displaced to any other off-center position $d \neq 0$.

The $Z$-dependence of the splitting energy in Fig. 3(b) for $\lambda = 2$ shows a continuous decrease as $Z$ changes its sign from negative (-1.0) to positive (0.1). The decrease is significant around $Z=-1$ which coincides with the physics discussed above for the negative $Z=-0.1$ and -1 cases.

It is found that for $Z = -0.1$ and -1 the ground state and the first-excited state in the two-electron QD containing a single charged impurity are the singlet and the triplet, respectively. Their major components are, respectively, the $s-s$ and the $s-p$ configuration, i.e. one electron is in the s- and the other in the s-(p-)orbital. Note that the $s-s$ overlap has only one maximum at the origin. In opposite, the $s-p$ overlap exhibits a minimum at the origin.

With changing the position of the impurity, the energy spin splitting of the singlet-triplet states is obtained in Fig. 4(a) for $Z=0.1$ and Fig. 4(b) for $Z=-1$. The presence of the impurity results in an increase in the splitting which is largest when the impurity is located at the center of the dot [as illustrated in Fig. 3(a) and Fig. 3(b) for $Z = 0$ (dot lines) and for $Z < 0$ (solid lines)].

The magnitude of the splitting over an impurity charge unit $\left[ (E_1^L - E_0^L) \equiv \text{splitting} / Z, \text{with } E_{0,1}^L \text{ the energy of the ground state and the first excited state in the doped QD} \right]$ is found smaller for the larger $Z$ case [see Figs. 3(a) and (b)] for a certain $d$ value. This means that the e-I attraction becomes dominant over the e-e Coulomb interaction. As a result, each state consists of different configurations with compatible contributions to the total wave function. For example, by increasing the e-I effective strength from $Z = -0.1$ [Fig. 3(a)] to $Z = -1$ [Fig. 3(b)] the contribution of the singlet $s$-configuration decreases from $C_0^a \approx 0.83$ to respectively $C_0^a \approx 0.64$. The compensatory parts come from other configurations which also have $L = 0$ such as $(n_1 = 0, l_1 = 0; n_2 = 1, l_2 = 0), (0,0;2,0), (0,0;3,0), \text{etc.}$ Those states stay closer in energy with increasing $\lambda (=2$ in this case).

Above we discussed the impurity effect for an attractive impurity (positively charged $Z < 0$). A negatively charged impurity which induces a repulsive coupling with electron is examined in Figs. 4(c) and (d) for two effective charges $Z=0.1$ and $Z=1$. The splitting energy between the ground state singlet and the first excited triplet is plotted as a function of the impurity position $d$. Apparently, when the impurity is found at the origin $d=0$, it repels the two electrons most. The probability of the electrons to be found at the origin (s-orbital) reduces significantly. In this case, the electrons can be found
at other higher Fock-Darwin states. This means that the ground state energy becomes closer in energy to the first excited state. That explains a smaller splitting energy we obtain for \( Z=0.1 \) [see Fig. 4(a)] and \( Z=1 \) [see Fig. 4(d)] as compared to, respectively, the cases \( Z=-0.1 \) [see Fig. 4(a)] and \( Z=-1 \) [see Fig. 4(b)]. Moreover, we see from Figs. 4(c) and (d) that when the impurity is moved away from the center of the QD, the splitting energy starts to increase. In other words, the probability of finding the electrons in the s-orbital increases.

C. Impurity effect on the energy spectrum

Technically, the presence of a charged impurity leads to an increase in the number of nonzero off-diagonal elements of the Hamiltonian matrix. We present such examination on the energy spectrum of the two-electron single QD as a function of the impurity position. The competition between the two types of Coulomb interaction results in different relative orders of the energy levels depending on both the impurity charge \( Z \) and the impurity position \( d \).

For the weak perturbation case, \( Z=-0.1 \) [Fig. 4(a)], the e-I interaction strength is much smaller (10 times) than the e-e interaction. The ground-state energy (level’s degeneracy \( g=1 \)) is the singlet state where the configuration with the two electrons in the s-orbital is highly dominant. The first- and second-excited states have degeneracies \( g=2 \) due to the symmetry of \( L = \pm 1 \) states. Note that at \( B=0 \) the results are independent of spins. We present the results in the \( S_z=0 \) subspace. We discuss the \( L = 1 \) case. In the first-excited state, the most dominant configuration is \((0,0;0,1)\), the second configuration is \((0,-1;0,2)\), and the last configuration is \((0,1;1,1)\). These three configurations and their exchange states have coefficients with opposite signs in the wave function (\( C_{0,1,0,0}) = -C_{(1,0,0,0)} \approx 0.7 \), \( C_{(-1,0,2)} = -C_{(2,0,-1)} \approx 0.07 \), and the other \( C_{(1,1,0,0)} = -C_{(1,1,0,0)} \approx 0.06 \) leading to the triplet first-excited state. The second-excited state has an opposite manner leading to the total spin \( S=0 \). Plus, the second dominant configuration and the third dominant configuration in the second-excited state switch their relative orders (sorted in probability) in the total wave function as compared to their orders in the first-excited state. The highest energies in Fig. 5 are the \((L=\pm 2,S=0)\) states with the largest configuration \((0, \pm 1; 0, \pm 1)\). Other configurations are \( (0,0;0, \pm 2) \) and \( (0, \mp 1; 0, \mp 3) \).

In the two energy spectra presented in Fig. 5, the ground-state and the first-excited state remain as singlet and triplet states. The fourth, fifth, and the sixth levels, which are the \((L=0,S=1)\) and \((L=\pm 1,S=0)\) states, exhibit a crossing at \( d = 0.0008a_B^* \). For \( d < 0.0008a_B^* \), the \((L=0,S=1)\) state has a lower energy than the 2-fold degenerate \((L=\pm 1,S=0)\) state. The major contribution to the wave function of the lower energy are, sorted in probability, \((0,0;1,0), (0,0;2,0)\), and \((0,0;3,0)\). Increasing \( d \geq 0.0008a_B^* \) leads to the exchange between the \((L=0,S=1)\) and \((L=\pm 1,S=0)\) states where the 2-fold degenerate \((L=\pm 1,S=0)\) state has lower energy.

The above physics is resulted from the dominant e-I interaction when \( Z=-1 \). The \((L=0,S=1)\) and \((L=1,S=0)\) states now have smaller energies than the \((L=2,S=0)\) state. Besides, it is found that the 2-fold degenerate states \((0, \pm 1,0, \mp 1)\), which dominate in the third-excited energy level \((L=0,S=1)\) for \( Z=0 \), have higher energies when the impurity is present.

The positive-\( Z \) case does not affect the energy spectrum as strongly as does for the negative-\( Z \) case. The reason is that the impurity in the positive-\( Z \) case induces the same type of Coulomb interaction with the QD e-e interaction. However, such an exchange in the relative order of the energy levels e.g., between the \((L=0,S=1)\) and \((2,0)\) states (corresponding to levels 4 – 6) can still be observed [see the crossing at \( d = 0.44a_B^* \) in Fig. 4]. The point \( d = 1.52a_B^* \) appears as a crossing-like point but it is only an almost-zero energy gap between the \((1,0)\) and \((0,1)\) states. Similar to the attractive impurity case, a larger e-I interaction also results in a smaller en-
energy gap between the first-excited state and the second-excited state.

D. Summary for the impurity effect on the energy spectrum of 2-electron single QDs

The cases of negative and positive \( Z \) affect the energy spectrum in different manners. Apparently, the impurity effect for the \( Z < 0 \) case is expected to be stronger than the \( Z > 0 \) case. When \( Z = -1 \) the e-I interaction strongly competes and dominates over the e-e interaction. Let us take an example to illustrate the point. In the weak perturbation regime, i.e. \( |Z| = 0.1 \), the e-e interaction dominates over the e-I interaction. The order of the low-level energy spectrum, then, exhibits no difference between those two cases \( Z = \pm 0.1 \). Now, \( |Z| \) is increased to 1. However, the system with negative \( Z \) exhibits a stronger e-I interaction. Such a strong effect for the attractive case \( Z = -1 \) is observed in the exchange of the singlet and triplet states with \( L=1 \) and \( L=0 \) as seen in the inset of Fig. 5(b). For the repulsive case \( Z = 1 \), this exchange is no longer observed. Only the exchange between the triplet \( L=0 \) and singlet \( L=2 \) is found. The repulsive e-I interaction in case \( Z = 1 \) is manifested in the presence of the \( (L=2,S=0) \) state which was not seen in the lowest energy levels shown in Fig. 5(b) for \( Z = -1 \).

IV. COUPLED QUANTUM DOTS

Numerical results for the singlet-dot case were discussed in detail mostly for \( \lambda = 2 \) corresponding to \( l_0 = 2a_B^d = 20.6 \text{ nm} \). This value of \( \lambda \) was studies based on the realistic sizes of GaAs single QDs. However, we also theoretically discussed the results for a typical range of \( \lambda = (1,2) \) as seen in Fig. 3(a). For the coupled QD problem, we used the optimized parameter set after Ref. 22 where \( l_0 = 10.01 \text{ nm} \) (\( \hbar \omega_0 = 11.24 \text{ meV} \)). Mapped on the single-dot case, the coupled 2-dot system will have \( \lambda \approx 1 \). The model of coupled dot system in the presence of a charged impurity can be schematically described in Fig. 7. We plotted the impurity potential in the top and the charged impurity potential (top) in a coupled 2-dot system. We assume the impurity is negatively charged with \( Z_1 = 1 \) and is located along the line connecting the two double-well minima (\( \pm a_m \)) where \( \pm \), respectively, indicates the left (L)- and the right (R)-dot. The two solid lines on top depict the impurity potential on the two individual electrons in the two separate QDs. The upper solid line is the potential of the electron in the right- and the lower solid line of the electron on the left-dot, respectively. Dotted lines on top depict the other case when the impurity is located on the other half of the \( x \)-axis (\( x_1 < 0 \)), i.e. it can be found inside the left-dot. \( V_b \) is taken to be 30 meV. The inset is the Coulombic potential of the impurity, located along the growth direction, which equally repels the two separate quantum dot electrons.

FIG. 6: Low-level energy spectrum of two-electron single QD containing a repulsive impurity (a) \( Z = 0.1 \) and (b) \( Z = 2 \). Inset in (a) is the magnification of the main plot into the region \( d = 0.1a_B^d \) to signify the crossing at \( d = 0.44a_B^d \) between the states \( (L=0,S=1) \) and \( (L=2,S=0) \).

FIG. 7: Schematic plot of the confining potential (bottom) and the charged impurity potential (top) in a coupled 2-dot system. We assume the impurity is negatively charged with \( Z_1 = 1 \) and is located along the line connecting the two double-well minima (\( \pm a_m \)) where \( \pm \), respectively, indicates the left (L)- and the right (R)-dot. The two solid lines on top depict the impurity potential on the two individual electrons in the two separate QDs. The upper solid line is the potential of the electron in the right- and the lower solid line of the electron on the left-dot, respectively. Dotted lines on top depict the other case when the impurity is located on the other half of the \( x \)-axis (\( x_1 < 0 \)), i.e. it can be found inside the left-dot. \( V_b \) is taken to be 30 meV. The inset is the Coulombic potential of the impurity, located along the growth direction, which equally repels the two separate quantum dot electrons.

TABLE 1

| Confinement (meV) | Impurity potential (meV) |
|-------------------|--------------------------|
| 0                 | 0                        |
| 20                | 20                       |
| 40                | 40                       |
| 60                | 60                       |
| 80                | 80                       |
| 100               | 100                      |

FIG. 8: Energy levels for a typical range of \( \lambda = (1,2) \) as seen in Fig. 3(a). For the coupled QD problem, we used the optimized parameter set after Ref. 22 where \( l_0 = 10.01 \text{ nm} \) (\( \hbar \omega_0 = 11.24 \text{ meV} \)). Mapped on the single-dot case, the coupled 2-dot system will have \( \lambda \approx 1 \). The model of coupled dot system in the presence of a charged impurity can be schematically described in Fig. 7. We plotted the impurity potential in the top panel for the case the impurity is located along the line connecting the two minima of the double well and in the inset for the case along the growth direction. It is clear that the impurity effect is largest when the impurity is at either of the two minima of the confining potential well in the former case. Whereas the latter case has the...
largest impurity effect when the impurity is found at center of the system, i.e. $R=0$. We label the two impurity coordinates as $\mathbf{R}_{1(2)}=(x_{1(2)}, y_{1(2)}, z_{1(2)})$.

A. Singlet-triplet splitting

1. Impurity position dependence

In the coupled QD system without impurity and without magnetic field ($B=0$), the two lowest energy levels are the maximally entangled exchange spin states, respectively, the singlet $\Psi^0$ and the triplet $\Psi^T$. The next higher excited states are the linear combination of the two doubly-occupied singlets which result in “bonding” ($\psi^S + \psi^S$) and “anti-bonding” ($\psi^S - \psi^S$) states.

Let us first consider the simplest case when the system contains only a single charged impurity $\mathbf{R}_{1}=(x_1, y_1, z_1)$ (effective charge $Z_1$) and the impurity plays only as a weak perturbation to the coupled dot system. However, there will be no restriction to the impurity location in or outside the system. Such a system allows us to provide a direct comparison to the single-dot case discussed earlier.

In case the e-I interaction is attractive coupling, i.e. $Z_1 < 0$, the singlet-triplet exchange energy $J$ is shown in Fig. $\mathbf{8a}$. When the impurity is located along the $z$-axis (see the black solid curve in Fig. $\mathbf{8a}$), the presence of the impurity increases the singlet-triplet spin splitting $J$ between the two electrons as compared to the case when no impurity is present [see horizontal grey dot curve in Fig. $\mathbf{8a}$]. This is understood as both electrons are attracted toward the impurity. Because the impurity equally couples to the electrons, the system favors the antiparallel electron spin state. This type of e-I coupling reduces the total energy of the system [negative binding energy presented as black solid in Fig. $\mathbf{8c}$]. As the impurity is moved away from the origin ($z_1 \neq 0$ - still the black solid curve in Fig. $\mathbf{8a}$), $J$ will decrease. Such a decrease can be evaluated via Eq. (A1) (see Appendix A) as the product of an exponential and complementary error function.

When the impurity is located along the ‘$z$’-direction of either the two separate dots, e.g. of the right-dot i.e. $\mathbf{R}_{1}=(a_m, 0, z_{1})$ [see red dash curve in Fig. $\mathbf{8a}$], $J$ remains larger than the $J$ for the case without impurity $Z_1 = 0$. However, $J$ behaves very differently from the above two cases $x_1 = 0$ and $x_1 = l_0$. The impurity not only no longer attracts equally the two QD electrons (similar to the $x_1 = l_0$ case) but also affects the doubly-occupied states most. $J$ in this case intersects the splitting energy of the $Z_1 = 0$ case at $z_1 \approx l_0$. Close to the QD sample, i.e. $z_1 << l_0$, $J$ is always smaller than the splitting energy for the without impurity case, about 10%. We reserve the detailed physical discussion around $(\pm a_m, 0)$ for a later discussion when we examine the case the impurity is located along the $x$-axis.

We expect that a repulsive impurity induces an opposite spin order of the two electron spin orientations: the parallel spin state. This is illustrated in Fig. $\mathbf{8b}$. A weakly repulsive impurity ($Z_1 = 0.1$), located along the $z$-axis (black solid curve), has the smallest exchange energy ($\approx 0.17 \text{ meV}$) when it is, apparently, at the origin: $z_1 = 0$. The reason is the impurity now repels both electrons. The e-I addition energy lifts up, $\approx$ few meV, the total energy of the coupled 2-dot system [see the positive binding energy presented in Fig. $\mathbf{8d}$]. $J$ rapidly increases and reaches the value of the non-impurity case as the impurity is engineered relatively far from the origin, say $3l_0$.

The ground state consists of all three Hund-Mulliken singlets, i.e. the total wave function $\Psi_{GS} = \{\psi^S_1, \psi^S_2, \psi^S_3\}$. However, two doubly occupied states play a small part, which are $\approx 2\%$, to the total wave function.

The binding energy of the impurity, which is defined as the energy difference of the system with and without a charged impurity, shown in Figs. $\mathbf{8c}$ and (d) was partially discussed above. In both cases $Z_1 = \mp0.1$, around $z_1 = 0$, the absolute value of the binding energy is found largest in case the impurity is placed closer to either of the two minima of the well ($x_1 = a_m$): 2.2 meV as compared to 2 meV and 1.53 meV of the $x_1 = l_0$ and $x_1 = 0$ case, respectively. Beyond a critical $z_1$, say $z_1 > 2l_0$ the most dominant binding energy case ($x_1 = a_m$ - black solid curve) becomes less dominant and compatible to the other cases $z_1 = 0$ and $l_0$. All three curves in Figs. $\mathbf{8c}$ and (d) convert to the situation without impurity at the large $z_1$ limit.

Now, we consider the case when the impurity is found inside the QD. In particular, we discuss the impurity effect when the impurity is found on the $x$- or $y$-axis, along which the confining potential is constructed.

Because the $Z_1 = \mp0.1$ case was found to weakly affect the quantum dot qubits and that the doubly-occupied states have very small contributions to the singlet-triplet splitting, we can now use Heitler-London model to analytically check our numerical results. We found qualitative agreement between the results presented in e.g. Fig. $\mathbf{9}$ and the analytical results shown in Eq. (B1), in particular the minimum (maximum) in $J$ for $Z=-0.1$ ($0.1$). Details are collected in Appendix B.

As signified in the top plot in Fig. $\mathbf{8}$ the absolute value of the effective e-I coupling exhibits a maximum at the either of the two well minima and decreases rapidly as the impurity position is out of the minima [analytical matrix elements are presented in Eq. (A5) in Appendix A]. However, the overlap between the two coupled QDs has a maximum at the origin (see e.g. Ref. [24]). In the weak impurity perturbation, i.e. $Z_1 << 1$, these two terms compete with each other. As a result, the singlet-triplet spin splitting $J$ for $Z_2 = -0.1$ has the impurity position dependence as shown in Fig. $\mathbf{9}$. $J$ has a maximum at the origin where the overlap between the two coupled dots is largest and the binding energy exhibits a maximum around $x_1 = 1.6l_0$ which is identical to the quasi-bottom positions of the left and the right dots $\pm a$. As
FIG. 8: (Color online) Singlet-triplet splitting energy calculated as a function of \( Z \) and (b) \( x \) (red dash), and minimum is obtained for an approximation (see Appendix B). We note that such an analytical minimum obtained using Heitler-London approximation (solid) to \( x \) the point \( z \) compared to the case the impurity is located along the \( x \)-direction, the considered case has a minimum around \( z_1 = 0 \) for \( V_0 = 30 \) meV. For comparison purpose, we recall the value of \( J \approx 0.204 \) meV as horizontal dashed-dot line in case no impurity is present for \( B = 0 \). The insets (c) and (d) show the corresponding total binding energy of the three cases plotted in each main plot (a) and (b).

FIG. 9: (Color online) Singlet-triplet splitting energy as a function of the \( x \)-coordinate of the impurity position for (a) \( Z_1 = -0.1 \) and (b) \( Z_1 = 0.1 \) for \( V_0 = 30 \) meV. The \( y \)- and \( z \)-coordinates of the impurity position, \( R_y \) and \( R_z \), are varied from the origin (black solid) to \( R_y = 0.2l_0 \) (green dotted), \( R_y = 0.5l_0 \) (violet short-dashed), \( R_y = 0.2l_0 \) (blue dash-dotted), \( R_y = 0.2l_0 \) (red dashed), and \( R_z = 0.5l_0 \) (dash-dot-dotted). The omitted coordinates are implied to be 0. We cover many different possible positions of the impurity such that: it can be along the \( x \)-axis or along the \( y \)-axis, or around the either the two minima with both \( x_1 \) and \( y_1 \) coordinates slightly changed while \( z_1 = 0 \), etc. Notice the case where the impurity is shifted along the \( y \)-axis. This shift does not add much physics to the system. As for example, see the insubstantial difference between the exchange energies in the black solid (\( y_1 = x_1 = 0 \)) and green dot (\( y_1 = 0.2l_0 \)) curves, or between the blue dashed-dot (\( z_1 = 0.2l_0 \)) and red dash (\( y_1 = z_1 = 0.2l_0 \)) curves, etc, presented in both (a) and (b).

compared to the case the impurity is located along the \( z \)-direction, the considered case has a minimum around the point \( x_1 \approx 2l_0 \) (see Fig. 9) which is identical to the analytical minimum obtained using Heitler-London approximation (see Appendix B). We note that such a minimum is obtained for an attractive impurity which has a positive charge \( Z_1 < 0 \) and \( |Z_1| \ll 1 \). Moving the impurity out of the dot system (blue dashed-dot, red dash, and cyan dash-dotted dot in Fig. 9) will lead to a decrease in \( J \). This situation can be considered as the coupled QDs interacting with charged impurities found close to the surface during the growth process. In particular, the case that the impurity \( y \)-coordinate is displaced to \( 0 < y_0/l_0 \ll 1 \) has only small difference in \( J \) from the
case the impurity is positioned exactly along the z-axis, \( y_0 = 0 \) (see the green short-dot curves and solid black curves in Fig. 9).

FIG. 10: (Color online) (a) Singlet-triplet spin splitting \( J \) as a function of the impurity effective charge \( Z_1 \) within the range \( Z_1 = (-1, 1) \) for three different positions of the impurity along the \( z \)-axis and (b) the singlet-triplet state-diagram as a function of the impurity charge \( Z_1 \) and its position along the \( z \)-axis. \( \psi_0 = 30 \) meV. S and T refer, respectively, to the singlet and triplet state. When the impurity is positioned close to the origin \( z_1 = 0 \) (black solid) and \( z_1 = 0.25a_0 \) (red dash) in (a), the system exhibits a triplet-singlet transition at \( Z_1 = 0.54 \) and 0.8, respectively. Green dot curve is added for reference purpose which shows that as the impurity is found at (or close) to either left- or right-minimum of the well the system is always the singlet.

2. Impurity charge dependence

The above critical point \( x_1 \approx 2l_0 \) at which the singlet-triplet splitting exhibits a maximum/minimum (see Fig. 9) depends on the effective impurity charge \( Z_1 \) and the well barrier height \( V_b \). In this subsection, we examine the \( Z_1 \) dependence of the singlet-triplet spin splitting \( J \).

Besides, we notice the physics sampled around the origin in (Fig. 9(d) and 11(b)) where the presence of a repulsive charged impurity significantly lowers the singlet-triplet spin splitting \( J \). It is expected that by further increasing the impurity charge \( Z_1 \), the system can be visited in the triplet state, i.e., \( J < 0 \).

Such triplet-singlet transition occurs as \( Z_1 \) is larger than \( \approx 0.55 \) in both cases the impurity is located along the \( z \)-axis and along the \( x \)-axis as shown, respectively, in Figs. 10 and 11. Our calculations for the attractive coupling case \( Z_1 < 0 \) show that there is no triplet-singlet transition (see Figs. 11 and 11) due to the fact that a positively charged impurity attracts both electrons therefore the favored spin state is always the singlet.

The triplet-singlet transition is further explored for different negative charges \( Z_1 > 0 \) as seen in both Figs. 10(a) and 11(a) for different impurity locations: at the center [black solid in Fig. 10(a)] and full rectangles with black solid in [11(a)] and off-center - slightly away from the origin. We only examine the triplet-singlet transition for the systems with the e-I coupling smaller or compatible to the e-e interaction. In fact, the limit of \( Z_1 = \pm 1 \) does not bear much physical meaning. However, we theoretically examine that limit to support a complete understanding of the effective e-I strength on the exchange electron qubits in zero \( B \)-field.

It is undoubted that when the impurity equally interacts with the two quantum-dot electrons and the e-I interaction strength is considerable the triplet ground state occurs at a smaller \( Z_1 \), as compared to the unequal electron(s)-impurity interaction case. This argument is clarified in e.g. Fig. 11(a) where the singlet state occurs by increasing \( Z_1 \) to \( \approx 0.54 \) for \( R_1 = 0 \) and further up to 0.7 for \( R_1 = a_m/4 \) (\( \approx 0.43a_0 \)). On the other hand, at a certain strong enough \( Z_1 \), the triplet state in case the impurity is located along the \( z \)-axis remains longer than in case the impurity is located along the \( z \)-axis. For example, for \( Z_1 = 1 \), we obtain that the triplet state stays up to \( x_1 \approx 0.6l_0 \) while it is found only up to \( z_1 \approx 0.46l_0 \). It is worth noting that at a higher \( V_b = 35 \) meV, due to the influence of the impurity potential, the triplet state remains up to a larger \( x_1 \) in comparison to the 30 meV case. For example, such a triplet-singlet transition is obtained at \( x_1 = 0.42l_0 \) and 0.72\( l_0 \) for \( Z_1 = 0.6 \) and 1, respectively.

When the impurity is located at the right well bottom for \( Z_1 = \mp 1 \), the ground-state is always a singlet however the maximally entangled component \( \psi_S^1 \) is replaced by one of the two double-occupied components \( \{ \psi_2^S, \psi_3^S \} \) as the major contribution to the total wave function.

We summarize in Figs. 10(b) and 11(b) the occurrence of the triplet or singlet as the ground state when changing the impurity position \( R_1 \) and its effective charge \( Z_1 \) for \( V_b = 30 \) meV. The triplet state starts to occur when \( Z_1 \) is increased to \( \approx 0.54 \) at \( R_1 = 0 \). The largest triplet region \( [R_1 = (0, 0.38a_0) \text{ in Fig. 10(b)} \) and \( (0, 0.6a_0) \text{ in Fig. } 11(b)] \) is, apparently, seen for the largest considered \( Z_1 \) (=1). In particular, close to the origin, the triplet region is rapidly shortened. For example, for \( Z_1 = 0.6 \) the triplet region is in \( R_1 = (0, 0.3) \) while such a region squeezes to \( (0, 0.07) \) as \( Z_1 \) decreases to 0.54. Note that we show the singlet-triplet state-diagram on the right half of the \( x \)- (\( z \))-axis. As the impurity is positioned along the other half of the \( x \)- (\( z \))-axis, the state-diagram is found similar.

3. Inter-dot separation dependence

In this subsection, we study the inter-dot dependence of the singlet-triplet splitting \( J \). First, we place the impurity at the center of the dot system, i.e., \( R_1 = (0, 0, 0) \), and tune the barrier height from \( V_b = 35 \) meV down to 13 meV. In the meantime, the inter-dot separation \( (2a_m) \) will decrease from \( 2a_m = 35 \) nm \( (\approx 3.5a_0) \) to 17 nm \( (\approx 1.6a_0) \). It is worth noting that the case that the impurity is placed exactly at either of the two well bot-
Figs. 11 and 12: (Color online) (a) Singlet-triplet splitting of a coupled 2-dot system as a function of the effective charge $Z_1$ within the range $-1 < Z_1 < 1$ for $x_1 = 0$ (full squares with black solid) and $x_1 = a_m/4$ (full circles with red solid). (b) Singlet-triplet state diagram plotted in the impurity-position–effective-charge $R_1 - Z_1$ plane. $V_b$ is taken to be 30 meV. Horizontal dot line in (a) is used to clarify the triplet-singlet transition. In the region close to the center of the system, at $\approx 0.07l_0$ (about 0.04$a_m$), a relatively large repulsive impurity potential, say $Z_1 = 0.54$, is enough to induce a triplet-singlet transition [see the state-diagram (b)]. The stronger the effective charge, the broader the triplet region. Consequently, when moving the impurity close to the right bottom it requires a larger $Z_1$ to observe the triplet-singlet transition occur. S and T stands, respectively, for the singlet and triplet state.

Figs. 13: (Color online) The same plot as Fig. 12 for $Z_1 = 0.1$. As the impurity position is engineered further away from the origin, the double-dot system reveals crossings between different $J$ for different $R_1$. The impurity position should be fixed thus its position only varies relatively with respect to the double-well minima of different-$V_b$ systems. Consequently, different-$V_b$ 2-dot systems exhibit various impurity effect on $J$. We obtain the inter-dot separation dependence of the singlet-triplet spin splitting $J$ for $Z_1 = -0.1$ in Fig. 12 and $Z_1 = 0.1$ in Fig. 13.

For $Z_1 = -0.1$, the splitting energy $J$ becomes smaller as the inter-dot separation increases (see Fig. 12). However, $J$ is found larger than that of the case without charged impurity as illustrated in e.g. the full rectan-
gles with black solid line ($R_1 = 0$) and open rectangles with magenta short dash (no impurity) in Fig. 12. In this case, this means that the potential well height is “weakened” by the attractive impurity which attracts the electrons toward and therefore supports the anti-parallel spin interaction of the two electrons. Displacing the impurity away from the center of the double-dot system (full rectangles) along the $x$-axis results in changes as seen in a series of $J$ presented as full circles with red solid (for $R_1 = 0.5 l_0$), full up-triangles with green dot (for $R_1 = 0.7 l_0$), full down-triangles with blue solid (for $R_1 = l_0$), full rhombuses with dashed-dot (for $R_1 = 1.5 l_0$), full right-triangles with yellow short-dot (for $R_1 = 2 l_0$), full circles with dash-dotted-dot (for $R_1 = 2.5 l_0$), and full stars with solid (for $R_1 = 3 l_0$).

First, we analyze the data for the lowest case of $V_b = 13$ meV (corresponding to $2 a_m \approx 1.65 l_0$). The minimum in $J$ for this case is found at $x_1 \approx 1.3 l_0$, while the maximal value in $J$ is always found at the origin $R_1 = 0$. Moreover, the system tends to convert to the no-impurity situation when the charged impurity is engineered far enough from the center of the dot system. Therefore, from the $R = 0$-case to the $R = 0.7 l_0$-case we obtained such a decrease in $J$ from $2.12$ meV to $1.87$ meV but for $R = l_0$ we obtained $J$ almost identical to the $J$ of a similar system but without impurity. Note that this impurity position $l_0$ is considered still close to the right-bottom of the well barrier. This feature is similar to the physics around the impurity position to the minimal point of $V_b$. Further increasing $x_1$ to $1.5 l_0$ and $2 l_0$ (as seen in the full rhombuses with cyan dashed-dot and full right-triangles with short-dot, respectively) we obtain the increase back in the latter case. This is found due to the fact that both $1.5 l_0$ and $2 l_0$ are located on the right-hand-side of the minimal point in $J$. The latter case exhibits an increase because the system to this extent tends to convert to the non-impurity case. In the furthermore case $R_1 = 3 l_0$ (full stars with solid line) $J$ is found closer to that of the non-impurity case which has higher energy than the cases $R_1 = 1.5 l_0, 2 l_0,$ and $2.5 l_0$.

Such a shift in the maximum of $J$ by changing $V_b$ can be seen in Fig. 12 and its inset for the high-$V_b$ limit where the furthermore impurity positions $R_1 = 1.5 l_0, 2 l_0, 2.5 l_0,$ and $3 l_0$ stay closer to the non-impurity case in comparison to the small-$V_b$ cases. This is related to the fact, which was mentioned above, that the relative distance of the impurity position to the minimal point of $J$ varies where the impurity can be found either in the left- or the right-hand-side of the minimal $J$ as $V_b$ changes. For $V_b = 30$ meV (corresponding to $2 a_m \approx 3.3 l_0$), from the center-$R_1 = 0$- to the $l_0$-case the singlet-triplet spin splitting stays higher in energy while the others ($R_1 = 1.5 l_0, 2 l_0, 2.5 l_0,$ and $3 l_0$) have lower $J$ than the non-impurity case. The minimal point for this system is $\approx 2 l_0$. Therefore, the $R_1 = 1.5 l_0$-case has a higher $J$ than the $R = 2 l_0$-case which was opposite in the previous case $V_b = 13$ meV. As a consequence, there appear crossings in $J$ for different $R_1$. Obvious crossings are seen for the cases $R_1 = 1.5 l_0, 2 l_0, 2.5 l_0,$ and $3 l_0$. For example, $J$ for the $R_1 = 0.5 l_0$-case (full rhombuses with cyan dashed-dot) stays lower in energy however it changes as $V_b$ varies and is found higher (or equal) in energy than the $J$ in the others (see the inset as a magnification for the high-$V_b$-limit).

Apparently, when placing the impurity close to the origin we always obtain a larger singlet-triplet spin splitting as compared to the non-impurity case. The largest case has $\Delta J \approx 0.6$ meV for $R_1 = 0$ and $V_b = 13$ meV. It is worth noting that a larger $V_b$ results in a smaller difference in $J$ which can be understood as a lower “tunneling” rate of the two electrons in the two separate QDs. Quantitatively, such a decrease can be roughly estimated by Eq. 11.

It was made clear in the presence of an attractive charged impurity the e-I coupling increases the “tunneling” rate between the two dots. Positioning the impurity at different locations inside the coupled 2-dot system, in particular around the $(x_1 \approx 2 l_0, 0)$ point, gives rise to different $V_b$-dependent singlet-triplet splitting which has crossings between different $R_1$-curves. As illustrated now in Fig. 13 for $Z_1 = 0.1$ case, $J$ in the $R_1 = 0$ case (full squares with black solid) stays lowest in energy and the $J$-difference as compared to the non-impurity case is found largest. For example, $\Delta J \approx 5.5$ meV for $V_b = 13$ meV. $J$ in the furthermore case $R_1 = 3 l_0$ (full stars with navy solid) stays slightly above the non-impurity curve (open squares with magenta short-dot). This implies that such an impurity position is on the right-hand-side of the maximal point in $J$ for all the considered $V_b \in (13, 35)$ meV in Fig. 13. It is found opposite to the $Z_1 = -0.1$ case where the $R_1 = 3 l_0$-$J$-curve stays slightly lower than the non-impurity one. Crossings between different $R_1$-dependent $J$-curves are still obtained in the current case $Z_1 = 0.1$, e.g. the one around $2 a_m = 2.1 l_0$ (corresponding to $V_b = 15.5$ meV) between the $l_0$- (full down-triangles with blue solid) and the $3 l_0$-$J$-curves, or the one at $2 a_m = 2.61 l_0$ ($V_b = 20$ meV) between the $1.5 l_0$- (full rhombuses with cyan dashed-dot) and the $2 l_0$-$J$-curve (see also the inset of Fig. 13), etc.

It is important to note that the $|Z_1| = 0.1$ case studied so far has not exhibited any crossing between the singlet and triplet states, i.e. the triplet state always stays higher in energy than the singlet state. The presence of an attractive impurity increases the singlet-triplet splitting opposed to the effect seen for a repulsive impurity. The extreme behavior of $J$ is obtained at either of the two minima of the potential well. An attractive impurity seems to increase the “tunnel” rate of the coupled double-dot system.

### B. Energy Spectrum

In the single QDs as discussed in Sec. III, impurity effect on the energy spectrum of the system is substantial
as e-I interaction is competitive to the e-e interaction. We obtained different crossings between low-lying excited states and anti-crossings with varying energy gaps. If such a similar effect is found in the coupled double QD system, the question “whether quantum operations in the coupled dot system are affected” attracts our attention.

In order to examine the validity of using coupled dots containing charged impurities as the basis of quantum logic gates in quantum computation, we mainly look into the low-level energy spectrum.

The energy spectrum of double QDs containing a single impurity located at \( \mathbf{R}_1 = (0,0,z_1) \) and \((0.5l_0,0,z_1)\), respectively, is shown as solid and dash-dotted curves in Fig. 14(a). The latter case (dash-dotted) has a larger impurity effect on the energy gap between the first two lowest levels and the two highest energy levels which can be understood in an earlier discussion. The energy levels bend down to the \( z_1 = 0 \) as its \( x \)-coordinate \( x_1 \) is different as compared to the case \( x_1 = 0 \) because the impurity has a stronger interaction to the electrons when it is closer to the well bottoms.

The \( x_1 \)-dependence of the energy spectrum is studied in Fig. 14(b) for two different \( z_1 = 0 \) and \( z_1 = l_0/2 \). The minimum, discussed earlier in Fig. 9 related to the physics of having an impurity around the well bottoms, is seen in the energy levels of Figs. 14(b) for \( z_1 = 0 \). The spin splitting between the two fully-filled states is most affected also when the impurity is at the well minima. The probability of finding the two electrons in the right dot (containing the impurity) increases. The absolute value of the e-I coupling for the right (in the present case) doubly-occupied state is maximal at \((a_m, 0)\).

For \( Z_1 = 0.1 \) [see Fig. 14(b)], it is found opposite to the attractive \( Z_1 = -0.1 \) case. The impurity (in the right-dot) now tends to repel both electrons which leads to a higher probability of finding the two electrons in the left-dot. Consequently, the doubly-occupied state of the right-dot gains energy via the e-I interaction and stays higher in energy (blue dashed-dot) than the singlet state (green dot) of the left-dot. Such a maximum around the bottom \((a_m, 0)\) in the blue dashed-dot curve in Fig. 14(b)
has similar physics discussed earlier for \( Z_1 = -0.1 \). Remember that in the attractive case, the singlet-triplet spin splitting has a minimum around \( 2l_0 \) (see Fig. 9) which is resulted due to i) the competition between the wave function overlap of the coupled 2-dots and the impurity potential on the right-dot electron and ii) the doubly-occupied states have relatively small contribution to the total singlet state (the ground state). Now, the effective charge changes its sign (0.1) and we obtain a maximum [see the inset of Fig. 15(b)]. The other left-dot fully occupied state [green dot in Fig. 15(b)] only shows a decrease in \( x_1 \).

Even though the presence of the impurity changes the singlet-triplet splitting as discussed above, the energy gap \( \Delta \) between the highest spin state that stores information and the lowest unwanted state, in this case the first and the second excited states, remains much larger than the singlet-triplet splitting energy \( J \). In case \( V_b = 30 \) meV, \( J/\Delta \) is typically \( \leq 0.07 \). This means that adiabatic condition is satisfied so that higher excited states are not involved when the system is evolved to its desired state.

C. Strong electron-impurity coupling destroys coupled qubits

1. Strong perturbation

In the following, we discuss in detail the influence of a strongly perturbative impurity on the energy spectrum for \( Z_1 = \mp 1 \). Results for other intermediate \( Z_1 \) (e.g. \( \mp 0.6, \mp 0.8 \)) are collected in Appendix C.

We plot in Fig. 16 the impurity-position dependence of the singlet-triplet spin splitting \( J \) when the impurity is located along the \( z \)-axis for (a) \( Z_1 = -1 \) and (b) \( Z_1 = 1 \). The former case has a similarly qualitative curve to the case \( Z_1 = 0.1 \) with a much larger exchange energy \( J \). The latter case has a triplet-singlet transition which occurs at \( z_1 = 0.38l_0 \) as obtained already in the singlet-triplet state-diagram Fig. 15(b). This property is resulted from the strong e-I interaction as \( Z_1 \) is increased to 1.

For the case the impurity is located along the \( x \)-axis and \( Z_1 = 1 \), we obtain in Fig. 17 and its inset the entire triplet state for the case the impurity is engineered at the origin or very close to the origin, say \( R_1 < 0.6l_0 \) (see the inset). The \( R_1 = 0 \)-case has the impurity that equally repels the two electrons in the two individual QDs and the impurity is kept distant from the two well minima. As a result, the favored state becomes the triplet with two electron spins aligned parallel to each other. Moving the impurity off-center means that there appears a bias in the impurity-electron coupling with the two electrons. Such a triplet state becomes weakened and disappeared when the bias increases to its maximum for the case the impurity is found around the bottoms of the confining potential.

![FIG. 16: (Color online) Singlet-triplet spin splitting \( J \) calculated as a function of the impurity position along the \( z \)-axis for \( Z_1=-1 \) (a) and 1 (b). \( V_b = 30 \) meV. S and T stand for, respectively, singlet and triplet. The horizontal dotted line in (b) is used to clarify the triplet and singlet states. Such a triplet-singlet transition is obtained at \( z_1 = 0.3l_0 \) for \( Z_1=1 \).](image1)

![FIG. 17: (Color online) Singlet-triplet spin splitting \( J \) of a singly-doped charged impurity coupled double-dot system as a function of the impurity position when it is located along the \( x \)-axis for \( Z_1 = 1 \). \( V_b = 30 \) meV. Inset is the magnification of the circled region in the main plot which highlights the triplet-singlet transition in the small \( x_1 \) region.](image2)
and 0.1(b) and strong \[ (Z_1=-1) \text{ and } (b) \text{ negatively } (Z_1=1) \text{ charged impurity. The inset in each layer is the corresponding singlet-triplet spin splitting } J \text{ where both exhibit a maximum around the right-bottom of the well. Different from the attractive case } Z_1 = -1 \text{ in (a), the repulsive case } Z_1 = 1 \text{ in (b) overcomes a triplet-singlet transition at } \approx 0.6l_0 \text{ [see the inset of (b)]. The green dot curve in (a) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet (see discussion in text) while the green dot in (b) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet. Red dash in both (a) and (b) is the triplet state. Circles that sample the anti-crossings indicate the transition of } \psi_1^S \text{ as the ground-state to the first excited state and the transition back to the ground state as } x_1 \text{ increases. Anti-crossings and almost zero energy gap between the first and second excited states in both (a) and (b) are the evidences to claim that the coupled qubits are "destroyed" and the adiabatical condition is violated.}

\[ \begin{align*}
\psi_1^S = 1 & \quad \text{in (b) overcomes a } \psi_2^S \text{ triplet-singlet transition at } \approx 0.6l_0 \\
& \quad \text{[see the inset of (b)]. The green dot curve in (a) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet (see discussion in text) while the green dot in (b) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet. Red dash in both (a) and (b) is the triplet state. Circles that sample the anti-crossings indicate the transition of } \psi_1^S \text{ as the ground-state to the first excited state and the transition back to the ground state as } x_1 \text{ increases. Anti-crossings and almost zero energy gap between the first and second excited states in both (a) and (b) are the evidences to claim that the coupled qubits are "destroyed" and the adiabatical condition is violated.}
\end{align*} \]

\[ \begin{align*}
\psi_2^S = & \quad \text{in (b) overcomes a } \psi_2^S \text{ triplet-singlet transition at } \approx 0.6l_0 \\
& \quad \text{[see the inset of (b)]. The green dot curve in (a) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet (see discussion in text) while the green dot in (b) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet. Red dash in both (a) and (b) is the triplet state. Circles that sample the anti-crossings indicate the transition of } \psi_1^S \text{ as the ground-state to the first excited state and the transition back to the ground state as } x_1 \text{ increases. Anti-crossings and almost zero energy gap between the first and second excited states in both (a) and (b) are the evidences to claim that the coupled qubits are "destroyed" and the adiabatical condition is violated.}
\end{align*} \]

\[ \begin{align*}
\psi_1^S & \quad \text{in (b) overcomes a } \psi_2^S \text{ triplet-singlet transition at } \approx 0.6l_0 \\
& \quad \text{[see the inset of (b)]. The green dot curve in (a) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet (see discussion in text) while the green dot in (b) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet. Red dash in both (a) and (b) is the triplet state. Circles that sample the anti-crossings indicate the transition of } \psi_1^S \text{ as the ground-state to the first excited state and the transition back to the ground state as } x_1 \text{ increases. Anti-crossings and almost zero energy gap between the first and second excited states in both (a) and (b) are the evidences to claim that the coupled qubits are "destroyed" and the adiabatical condition is violated.}
\end{align*} \]

\[ \begin{align*}
\psi_2^S & \quad \text{in (b) overcomes a } \psi_2^S \text{ triplet-singlet transition at } \approx 0.6l_0 \\
& \quad \text{[see the inset of (b)]. The green dot curve in (a) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet (see discussion in text) while the green dot in (b) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet. Red dash in both (a) and (b) is the triplet state. Circles that sample the anti-crossings indicate the transition of } \psi_1^S \text{ as the ground-state to the first excited state and the transition back to the ground state as } x_1 \text{ increases. Anti-crossings and almost zero energy gap between the first and second excited states in both (a) and (b) are the evidences to claim that the coupled qubits are "destroyed" and the adiabatical condition is violated.}
\end{align*} \]

\[ \begin{align*}
\psi_1^S & \quad \text{in (b) overcomes a } \psi_2^S \text{ triplet-singlet transition at } \approx 0.6l_0 \\
& \quad \text{[see the inset of (b)]. The green dot curve in (a) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet (see discussion in text) while the green dot in (b) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet. Red dash in both (a) and (b) is the triplet state. Circles that sample the anti-crossings indicate the transition of } \psi_1^S \text{ as the ground-state to the first excited state and the transition back to the ground state as } x_1 \text{ increases. Anti-crossings and almost zero energy gap between the first and second excited states in both (a) and (b) are the evidences to claim that the coupled qubits are "destroyed" and the adiabatical condition is violated.}
\end{align*} \]

\[ \begin{align*}
\psi_2^S & \quad \text{in (b) overcomes a } \psi_2^S \text{ triplet-singlet transition at } \approx 0.6l_0 \\
& \quad \text{[see the inset of (b)]. The green dot curve in (a) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet (see discussion in text) while the green dot in (b) represents either the } \psi_2^S \text{ singlet or the } \psi_1^S \text{ singlet. Red dash in both (a) and (b) is the triplet state. Circles that sample the anti-crossings indicate the transition of } \psi_1^S \text{ as the ground-state to the first excited state and the transition back to the ground state as } x_1 \text{ increases. Anti-crossings and almost zero energy gap between the first and second excited states in both (a) and (b) are the evidences to claim that the coupled qubits are "destroyed" and the adiabatical condition is violated.}
\end{align*} \]
dot the $\psi^S_1$ (major) state in the region $R_1 < 0.8l_0$ and $R_1 > 2.7l_0$. The region in between $0.8l_0 < R_1 < 2.7l_0$ has the ground state as the doubly occupied singlet $\psi^S_2$ in Fig. 18(a) and $\psi^S_3$ in Fig. 18(b). The minor components in the first excited state in both cases, respectively, $\psi^S_1$ plus $\psi^S_3$ and $\psi^S_1$ plus $\psi^S_2$ fluctuates around 7% and 3% as the impurity is located close to the center of the system. In Fig. 18(b) we see a switch between the left- and right-doubly-occupied singlets where the singlet of the right-dot (with impurity - blue dashed-dot) stays higher in energy than the one of the left-dot (green dot).

The “virtual” coupling between the three singlets and the triplet addressed above is now examined in both cases $Z_1 = \mp1$. At the well right-bottom $R_1 = (a_m,0)$, the triplet (red dash) “couples” with the first excited singlet as green dot curve [\psi^S_3 in Fig. 18(a)] and [\psi^S_2 in Fig. 18(b)]. The energy gap between the triplet and the first excited state is found close to zero. This virtual coupling can be understood by the two anti-crossings between the $\psi^S_1$ and $\psi^S_3$ in Fig. 18(a) and between $\psi^S_1$ and $\psi^S_2$ in Fig. 18(b).

To better understand the physics manifested in the mixing of different singlet states as well as the “coupling” between the triplet and the first excited singlet as shown in Figs. 18(a) and (b) and the triplet-singlet transition in case $Z_1 = 1$ we calculate the probability of finding the three singlet states as a function of the impurity position inside the double-dot system for weak and strong e-I coupling, respectively, in Figs. 19(a) and (b) and Figs. 19(c) and (d) for the ground state. We first explain for the triplet-singlet transition in case $Z_1 = 1$ [see Fig. 17(a)] and the “coupling” of the first excited singlet and the triplet in case $Z_1 = \mp 1$ [see Figs. 18(a) and (b)]. The triplet-singlet transition can be understood from Fig. 19(d) where the region $x_1/l_0 \leq 0.6$ has zero probability of finding the $\psi^S_1$ state in the ground state. It is because the triplet state becomes the ground state and the singlet $\psi^S_1$ is the major component of the first excited state wave function in this considered region. The dominant of the doubly occupied singlet $\psi^S_3$ in the ground state for $Z_1 = -1$ and of the $\psi^S_2$ for $Z_1 = 1$ is now specified as the region $0.8l_0 < x_1 < 2.7l_0$ in Figs. 19(c) and (d). This region is identical to the maximal behavior of $J$ as seen in Figs. 18(a) and (b) and their two insets. For weak impurity potential cases $Z_1 = \mp 0.1$ there are no anti-crossings in the energy spectrum and the doubly-occupied singlets remain always as the excited singlets with small probability [see Figs. 19(a) and (b)].

**D. Coupled qubits perturbated by two impurities**

For simplicity, we first assume that two impurities have identical charges $Z_1 = Z_2$ and one impurity is kept at one of the two minima $R_2 = (-a_m,0,0)$. The other impurity can be arbitrarily located along the other side of the $x$-axis, $R_1 = (x_1 > 0,0,0)$. Under these circumstances, the two impurities generally induce different e-I couplings and hence affect the qubits asymmetrically except when $x_1 = -x_2 = a_m$. In this case, both doubly-occupied singlets are found compatible which have similar contribution to the two excited singlet states. Therefore, we no longer see a minimum or maximum, depending on the sign of $Z_1$, in the second or third excited state as obtained in case $Z_2 = 0$. Such a pronounced minimum is now “eliminated” due to the presence of the second impurity, located at the other bottom (left) of the 2-dot system, which induces a competing e-I coupling. The singlet-triplet spin splitting $J$ has maximal behavior when the impurity is found close and at the bottom of the right-dot.

Another way to study the asymmetric impurity effect in the considered double-dot system with two impurities is to adjust the effective impurity charges with respect to each other while their positions are kept symmetrical to the origin, i.e. $Z_1/Z_2 \neq 1$ while $R_1 = -R_2$. We compute the singlet-triplet spin splitting for three different values of $Z_2/Z_1 = 1, 1.1$, and $1.2$ while $R_{1,2}$ changes for both positively and negatively charged cases in Fig. 21(a) and (b), respectively. The results shown in Fig. 21 are obtained for $Z_2/Z_1$ slightly different than unit to examine the asymmetry arisen due to the dissimilarity in impurity charges as compared to the symmetry due to the impurity positions discussed earlier in Fig. 20 for $Z_1 = Z_2$.

Apparently, if one replaces one of the two identical impurities by an opposite charge to the other one can revisit the “asymmetric” phenomenon between the two doubly
Our further calculations show that the occupied singlets in the two dots which now can be enhanced as a double. Our further calculations show that they are far enough from the target (A) coupled 2-dot sample and only induces electrostatic interaction to the two electrons of the target system. We examine the case that the two impurities have varied positions but their center-of-mass coordinates are kept unchanged, in this case $R_{12} = (R_1 + R_2)/2 = (5a_m, 0, 0)$. This center-of-mass location can be imagined as another “center” of another coupled 2-dot system (B) where the two impurities are mostly found at the well bottoms of the system B. That the relative positions of individual impurities vary with respect to their center-of-mass position can be imagined as the well barrier height is tuned resulting in varying inter-dot separation in system B. A and B only interact via the e-I Coulomb interaction. Now, we study the influence of the presence of system B on the qubits of the system A. We plot in Fig. 22 the singlet-triplet spin splitting as a function of one impurity position for system A. We find that $J$ decreases as the impurity which stays closer to the system A is positioned further from the system A. This effect is expected to be significantly enhanced if one applies an external field to engineer the impurities in and out the active region of the target system A. As a result, one can see such anti-crossings as seen earlier for $Z = \mp 1$ where the triplet state stays in-between the two lowest singlets.

![FIG. 21: (Color online) Singlet-triplet spin splitting $J$ as a function of the impurity position in case the two impurities are symmetrically located, $R_1 = -R_2$, but they are charged slightly differently (a) both impurities are positively charged and (b) both impurities are negatively charged with $Z_1 = \pm 0.1$ and $Z_2 = Z_1$ (black solid), 1.1$Z_1$ (red dash), and 1.2$Z_1$ (blue dash-dotted). $V_b$ is taken to be 30 meV. $V_b$ is fixed at 30 meV which corresponds to $a_m \approx 1.64a_0$. Note that the system converts to the situation without impurity at a much larger $x_1$ than the singly-doped impurity case (previously seen around 3.5$b_0$). In this plot, at $x_1 = 4b_0$ the system does not convert to the case without impurity (e.g. $\Delta J \approx 0.01$ meV) and the total binding energy $\Delta E \approx 0.8$ meV for $Z_1 = -0.1$ [black solid in (a)].](image)

![FIG. 22: (Color online) Singlet-triplet spin splitting $J$ as a function of the impurity position in case the two impurities (blue circles) are located relatively far from the two potential minima, $R_1 + R_2 = 5a_m$ and the closest impurity to the active coupled 2-dot system (the grey box) starts at $R_1 = 3.5a_m$. By fixing the center-of-mass coordinate of the two impurities at $R_c = 5a_m$, we imply such two impurities can be considered as another coupled 2-dot system which has no tunneling coupling with the considered 2-dot system in the grey box. That the relative positions of each impurity $R_{1,2}$ vary with respect to the origin $(0,0)$ of the active system can be mapped to the problem of other 2-dot system (in open dotted box) with varying potential barrier height. $V_b$ of the active system is taken to be 30 meV. $J$ is slightly different from that of the system without impurity.](image)
V. DISCUSSION AND CONCLUSION

The coupling of two qubits is sensitively affected by the presence of a charged impurity. A charged impurity which is found inside the right- or the left-dot “breaks down” the equality between the right and the left doubly occupied singlet states. In case the impurity weakly couples with the electrons, the maximally doubly-occupied singlet of the dot containing the impurity has a smaller energy due to the e-I coupling. This can be considered as a tool to distinguish the doped and un-doped QD components in the coupled QD system.

A relatively strongly repulsive impurity which almost equally couples to the two individual QD electrons will result in an entire triplet state at B=0 due to the small competition between the two e-I couplings. This can be observed by positioning the impurity around the origin of the double-dot system. When the impurity is found close to the bottoms of the confining potential well the system will stay in the singlet state where the two electrons perform the favored singlet state with spins anti-parallel to each other. Due to this fact, the influence of the inter-dot separation in the presence of impurity can be examined by modulating the well barrier $V_b$ without changing the single-particle properties in individual QDs. The reason is that the relative position of the impurity with respect to the minima of the 2-dot system is effectively controlled by changing $V_b$. We found that an attractive impurity serves to increase the “tunnel” rate between the two coupled dots.

In general, charged impurities “destroy” the maximally entangled singlet state by mixing the different singlets having varying contributions to the total wave function. A strongly perturbative impurity really “messes up” the quantum information obtained from this coupled QDs because the triplet state “couples” to the second excited state by the presence of anti-crossings with a slightly larger than zero energy gap. The ground state now can favor a doubly occupied state. Because the triplet is always obtained with probability 1, it occurs in between the two lowest singlets: the exchange singlet $\psi_1^S$ and the doubly-occupied $\psi_2^S$ or $\psi_3^S$ depending on the sign of the effective charge $Z_1$. The energy spectrum appears with anti-crossings as seen in our results.

In the presence of two identical charged impurities, the breakdown between the two doubly-occupied singlets of the left and the right dots is “mended” if the impurities are located exactly symmetrically along the line connecting the two confining potential minima, i.e. the x-axis. As a result, one no longer obtains such a bias between the two doubly-occupied singlets in the left- or the right-dot as seen in the energy spectrum.

The mixing of different singlet states and anti-crossing coupling of the singlet and triplet states due to the impurity presence results in significant affect on the qubit operations, e.g. exchange and C-NOT, in the considered coupled 2-quantum-dot system.

Finally, we discuss the implications of our results for the operation of semiconductor spin qubits. Obviously, without some quantitative knowledge of impurity locations near the qubits, no real comparison between our results and experiments would be possible. But we can make some general remarks based on statistical considerations.

First, semiconductor qubits are typically fabricated by lithographic techniques creating QDs from parent 2DEG systems. The low-temperature mobility of the 2DEG is controlled entirely by the background charged impurity density both in Si and in GaAs systems. Theoretical calculations can provide quantitatively accurate information about the ensemble averaged impurity density in the 2DEG from the mobility measurements. This information about the background impurity density can be converted to a statistical probability of finding impurities located in specific QD structures since the effective active areas of the dots would be known from the lithographic structures. For example, typical GaAs 2DEG would have $10^{10}$ to $10^{11}$ charged impurities per sq. cm. whereas Si systems are typically dirtier with $10^{11}$ to $10^{12}$ charged impurities per sq. cm. This crudely translates to one charged impurity every 10 to 100 nm in linear distance statistically. Typical lithographic qubits are 20-100 nm squares, indicating the presence of 1-5 charged impurities per qubit statistically with GaAs (Si) being near the lower (higher number) number. Of course, the details of impurity locations matter very much, and the statistical considerations could be improved and combined with our exchange splitting calculations to obtain the effective sample “yield” for a given 2DEG mobility, i.e. estimate the fraction of samples that would statistically have the impurities far enough for spin qubit operations to work. But even these elementary considerations show that the effective yield is likely to be very low with probably only about 1-5% of QD systems being “lucky enough” to have the impurities located far enough from the dots for them to work as spin qubits. The rest, even before taking into account extraneous experimental factors (e.g. leakage, noise), simply would not work because there is no effective singlet-triplet coupling providing the necessary entanglement in the system. A possible way of circumventing this problem may perhaps be having several electrons per dot so that the external charged impurity potential is effectively screened by the QD electrons, but this raises the problem of having rather weak exchange coupling in multielectron dots. The complex interplay of multielectron dots in the presence of random charged impurities in the background is left as an important future open problem in this subject, which may very well be the next important step in this direction.

Second, an important effect of background charged impurities is their fluctuations which produce various charge noise signals in semiconductor devices. This charge noise may very well be the limiting decoherence mechanism in currently existing semiconductor QD spin qubits. Normally, of course, charge noise would not adversely affect spin configurations, but semiconductor spin qubits de-
pend on the electrostatic exchange coupling which is affected by external charged impurities as discussed extensively in this paper. Therefore, any impurity fluctuations would lead to spin qubit decoherence through the charge noise mechanism. Our current work provides quantitative estimate of the strength of such charge noise if the fluctuation spectrum (or the fluctuation timescale) is known. Deriving microscopic information about charge noise from our results could be another future interesting direction of research.

Third, the inevitable presence of static charged impurities in the background makes every semiconductor dot spin qubit, whether in GaAs or in Si (or some other material), unique since the microscopic electrostatic potential environment for each qubit will necessarily differ in a random manner from qubit to qubit due to background impurities as shown in this paper. In particular, the singlet-triplet energy separation and the consequent exchange coupling will thus be somewhat random in a collection of many qubits. This would in turn necessitate characterization of each qubit in the eventual quantum computer rather precisely since the gate operations depend very strongly on the knowledge of the exchange coupling between the dots. Such a characterization will adversely affect the scalability of semiconductor spin quantum computer architectures. Our work shows the importance of having ultraclean impurity-free environment even for solid state quantum computation, similar to the requirement for topological quantum computation using non-Abelian fractional quantum Hall states, not because spin quantum computation needs ultra-high mobilities, but because it requires stable values of exchange coupling without large qubit-to-qubit variations. Our current work indicates that an order of magnitude reduction in the background impurity concentration in GaAs, bringing it down to around 10^{12} per cubic cm, which is also the goal for topological quantum computation, should lead to the production of 100-1000 spin qubits with impurity-free environment rather easily. Unfortunately, for Si-SiO_{2} systems this is a very stringent condition because of the invariable presence of large oxide charges near the interface, but for GaAs and Si-Ge based quantum dot systems, low-impurity materials may become available for quantum computer architectures in the near future.

Fourth, most of the spin qubit manipulation experiments in semiconductor quantum dots are carried out under transport situations using an external dc voltage bias. Such external voltage affects the two dots differentially somewhat similar to the impurity effects discussed in the current work since the confinement potentials for the two dots are affected differently by the external voltage. Our technique can be used to study this effect. Another possible future direction of study could be impurity effects on the full coupled quantum dot energy spectra (i.e. in addition to just the double-dot exchange energy mainly considered in the current work) since in some experiments higher energy levels come into play.

It will, in fact, be interesting also to consider multielectron quantum dot systems with more than one electron per dot and ask how impurity disorder affects the energy spectra. This would necessitate a calculation of the quantum dot electronic structure using many orbitals per dot (e.g. s, p, d, f, ...), which is beyond the scope of the current work.

VI. ACKNOWLEDGMENTS

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Appendix A: Coulomb matrix elements

1. Single QDs

To evaluate the e-I Coulomb matrix element, we calculate the integral:

\[ V_{n_{1}l_{1}n_{2}l_{2}}^{m_{1}m_{2}} (\vec{R}) = \int \varphi_{n_{1}l_{1}}^{*} (\vec{R}) \frac{1}{|\vec{R} - \vec{r}|} \varphi_{n_{2}l_{2}} (\vec{R}) \, d\vec{R} \quad (A1) \]

where \( \varphi_{n,l}(\vec{R}) \) is defined in Eq. (7). The denominator in the right hand side of Eq. (A1) is eliminated using the Gaussian identity:

\[ \frac{1}{r} = 2 \pi \int_{0}^{\infty} e^{-u^{2}r^{2}} \, du. \quad (A2) \]

Integrating integral (A1) over \( \theta \) and set \( \vec{r}^{2} = t \), we arrive at:

\[ V_{n_{1}l_{1}n_{2}l_{2}}^{m_{1}m_{2}} (\vec{R}) = \delta_{l_{1},l_{2}} \left( \frac{n_{1}!n_{2}!}{(n_{1} + l^{+}!)!(n_{2} + l^{+}!)} \right) \]

\[ \times \int_{0}^{\infty} t^{l^{+}} e^{-t(1+u^{2})} L_{n_{1}}^{l^{+}} (t) L_{n_{2}}^{l^{+}} (t) e^{-u^{2}R^{2}} \, dtdu. \]

Next, we use one of the properties of the Laguerre polynomials:

\[ \int_{0}^{\infty} t^{\alpha-1} e^{-pt} L_{m}^{\lambda} (at) L_{n}^{\beta} (bt) \, dt = \frac{\Gamma (\alpha + 1)^{m}(\beta + 1)^{n} p^{-\alpha}}{m!n!} \sum_{j=0}^{m} \frac{(-m)_{j}(\alpha)_{j}}{(\lambda + 1)_{j}j!} \left( \frac{a}{p} \right)^{j} \]

\[ \times \sum_{k=0}^{n} \frac{(-n)_{k}(j + \alpha)_{k}}{(\beta + 1)_{k}k!} \left( \frac{b}{p} \right)^{k} \]

and the gamma functions for integers to calculate the integral in the right hand side of (A3). Finally, we arrive at the formula presented in Eq. (13).

2. Coupled QDs

In general, the Coulomb matrix elements between the electrons and impurity are obtained numerically. Except
for few cases with applied constraint conditions to the impurity position, it is impossible to obtain a closed algebraic form for this type of Coulomb interaction. Using a similar method as before to calculate the e-I coupling elements, i.e. \( \langle \Psi | \mathcal{V}_{\text{e-Imp}} | \Psi \rangle \), we obtain:

\[
\langle \varphi_L(R) | \mathcal{V}_{\text{e-Imp}} | \varphi_L(R) \rangle = \frac{ZV_0^C}{\sqrt{\pi}} \int_0^\infty \frac{x e^{-x^2}}{v(x+1)} dx \quad \text{(A4)}
\]

\[
\langle \varphi_L(R) | \mathcal{V}_{\text{e-Imp}} | \varphi_R(L) \rangle = ZV_0^C \sqrt{\pi e^{z_0^2 - a_0^2} \text{erfc}(z_0)} \quad \text{for } R = (0, 0, z)
\]

\[
\langle \varphi_L(R) | \mathcal{V}_{\text{e-Imp}} | \varphi_R(L) \rangle = ZV_0^C \sqrt{\pi e^{-(z_0^2 + a_0^2)/2}} \quad \text{I}_0([x_0 \pm a_0]^2) \quad \text{for } R = (x_0, y_0, 0)
\]

\[
|\langle \varphi_L(R) | \mathcal{V}_{\text{e-Imp}} | \varphi_R(L) \rangle|^2 = ZV_0^C \sqrt{\pi e^{-(a_0^2 + z_0^2)/2} \text{I}_0([x_0^2 + y_0^2]/2)} \quad \text{for } R = (x_0, y_0, 0)
\]

\[
J_{\text{HL}} = -\frac{e^{a_0^2}}{1 - e^{4a_0^2}} \left\{ 2h\omega_0 a_0^2 e^{a_0^2} + \frac{4V_0(e^{\frac{a_0^2}{1 + a_0^2}} - e^{\frac{a_0^2}{2}})}{\sqrt{(1 + a_0^2)(1 + a_0^2)}} \right. \right.
\]

\[
+ \frac{4V_0(e^{\frac{a_0^2}{1 + a_0^2}} - e^{\frac{a_0^2}{2}})}{\sqrt{(1 + a_0^2)(1 + a_0^2)}} + \sqrt{\pi} V_0^C \left[ -2e^{a_0^2} + \sqrt{2} \text{I}_0(a_0^2) - 4Ze^{-\frac{a_0^2 + x_0^2 + y_0^2}{2}} \text{I}_0(\frac{x_0^2 + y_0^2}{2}) \right]
\]

\[
+ 2e^{\frac{a_0^2 + x_0^2 + y_0^2}{2}} \text{I}_0(\frac{a_0^2 - 2a_0x_0 + x_0^2 + y_0^2}{2}) + 2e^{\frac{a_0^2 - x_0^2 + y_0^2}{2}} \text{I}_0(\frac{a_0^2 + 2a_0x_0 + x_0^2 + y_0^2}{2}) \right\}.
\]

Here, \( a_k = a/l_0, a_{0x} = l_0/l_x, a_{0y} = l_0/l_y, a_{0xk} = l_0/l_{xk}, \) and \( a_{0y} = l_0/l_{y} \) are dimensionless parameters associated with a series of lengths such as \( a, l_x, l_y, l_{xk}, l_{y} \). These parameters together with depths \( V_0 \) and \( V_6 \) define the size and the shape of the confinement double well.

Heitler-London approximation does not allow one to examine the impurity effect on the coupled qubit operations manifested in the mixing of the different singlets and “coupling” between the singlet-triplet states as already observed and discussed in e.g. Fig. 18(b). The impurity-position dependence of \( J_{\text{HL}} \), as obtained analytically in the last three terms in Eq. (B1), now can be studied in Fig. 23 as a function of \( x_0 \). We see that the presence of the charged impurity only plays a role as a weak perturbative interaction to the total energy even in case the effective e-I coupling is found relatively large (the case \( Z_1 = -1 \)). Such a property like the triplet-singlet transition for weak repulsive e-I exchange interaction is not found in this case.

It is of interest to consider the special situation when the impurity is located along the line connecting the two potential well minima, i.e. along the \( x \)-axis in this case. In the above sections, we discussed this case in depth. The Coulomb matrix elements for the left and the right QD electron are as follows:

\[
\langle \varphi_L(R) | \mathcal{V}_{\text{e-Imp}} | \varphi_L(R) \rangle = ZV_0^C \sqrt{\pi e^{-(a_0^2 + z_0^2)/2}} \quad \text{I}_0([x_0 \pm a_0]^2) \quad \text{(A6)}
\]

\[
\langle \varphi_L(R) | \mathcal{V}_{\text{e-Imp}} | \varphi_R(L) \rangle = ZV_0^C \sqrt{\pi e^{-(a_0^2 + z_0^2)/2}} \quad \text{I}_0([x_0^2 + y_0^2]/2).
\]

**Appendix B: Heitler-London approximation**

The closed analytical form of the exchange energy between the two coupled QDs for a single charged impurity located arbitrarily in the \( xy \)-plane \( [R = (x_0, y_0, 0)] \) is obtained by solving a basic two-level problem:

\[
J_{\text{HL}} = \frac{e^{a_0^2}}{1 - e^{4a_0^2}} \left\{ \right. \right.
\]

\[
\left. \right\}.
\]

**Appendix C: Singlet-triplet splitting \( J \) for intermediate impurity effective charges**

We show the results obtained for several different intermediate \( Z_1 \) for both the negatively and positively charged impurity cases in Figs. 24, 25, and 26. These plots provide a clearer and deeper look into the triplet-singlet transition (Figs. 25 and 26) for a strongly repulsive impurity as well as the physics of anti-crossings found in the energy spectrum of the coupled 2-dot system. For the \( Z_1 < 0 \) case, Fig. 24 serves to explain in detail the transition from having a minimum to a maximum in the singlet-triplet splitting \( J \) between the two lowest energy levels.
FIG. 23: (Color online) Singlet-triplet spin splitting $J$ as a function of the impurity position along the $x$-axis for (a) $Z_1=-0.1$ and (b) 0.1. Black solid curves are extracted from Fig. 9 and blue dashed curves are obtained from Eq. (B1). We receive qualitative agreement between our numerical calculations and analytical results where the singlet-triplet splitting energy calculated using these two methods exhibits a (a) minimum for $Z_1=-0.1$ and (b) maximum for $Z_1 = 0.1$ around $x_1 \approx 2l_0$.

FIG. 24: (Color online) Singlet-triplet spin splitting $J$ as a function of the impurity position along the $x$-axis for different $Z_1 < 0$. In this plot one can see the gradual change in $J$ from having a minimum around $x_1 \approx 2l_0$ to having a maximum at different $x_1$ depending on $Z_1$ as $Z_1$ is increases from -0.1 (black solid) to -0.8 (cyan dash-dotted-dot).

FIG. 25: (Color online) Singlet-triplet spin splitting $J$ as a function of the impurity position along the $x$-axis for different $Z_1 > 0$. The maximum in $J$ is found at different $x_1$ as $Z_1$ changes.

FIG. 26: (Color online) Singlet-triplet spin splitting $J$ as a function of the impurity position along the $x$-axis for a specific case of $Z_1 > 0$: 0.6. Inset is the low-level energy spectrum of the system. As compared to the pronounced anti-crossing seen in Fig. 18(b), the $Z_1 = 0.6$ case has the spectrum which is close to having pronounced anti-crossings as obtained for $Z_1 = 1$ in Fig. 18(b).

Appendix D: Spin splitting energy of Si/SiGe coupled quantum dots

Si QDs have a relatively large effective mass $m^*_{Si} \approx 2.8m_{GaAs}$. Plus, the effective Rydberg energy in Si QDs, as discussed in Sec. II, is higher than the effective Rydberg energy of GaAs QDs. These bring into the fact that the Si QDs have a smaller electron kinetic energy, which means that the electrons tend to be more localized as compared to the case of GaAs. Thus the tunnel rate is expected to be smaller. Our calculations show that for the same model of a coupled 2-dot system in which the shape and the size and the barrier height are kept unchanged: e.g. $V_0 = -50$ meV and $V_b = 30$ meV (correspond-
ing to $\Delta V_b = 9.65$ meV), Si/SiGe double-dot system has $\hbar \omega_b \approx 6.67$ meV ($l_0 \approx 8.48$ nm). The spin singlet-triplet splitting $J$ between the two lowest energy levels in Si/SiGe double dots is found $J \approx 0.003$ meV, much smaller than the $J \approx 0.204$ meV of the GaAs/AlGaAs double dots. In experiments, the electron tunneling rate in Si/SiGe coupled double QDs is indeed found much lower than the tunneling rate in GaAs/AlGaAs coupled QDs (see e.g. Ref. 81). In the presence of a charged impurity, e.g. $Z_1 = -0.1$ located at the origin, we obtained $J$ increased to $J \approx 0.008$ meV for Si/SiGe double QDs as compared to $J \approx 0.26$ meV for GaAs/AlGaAs double QDs.

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