

# Exchange interaction between three and four coupled quantum dots: Theory and applications to quantum computing

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Several prominent proposals have suggested that spins of localized electrons could serve as quantum computer qubits. The exchange interaction has been invoked as a means of implementing two qubit gates. In this paper, we analyze the strength and form of the exchange interaction under relevant conditions. We find that, when several spins are engaged in mutual interactions, the quantitative strengths or even qualitative forms of the interactions can change. A variety of interaction forms can arise depending on the symmetry of the system. It is shown that the changes can be dramatic within a Heitler-London model. Hund-Mülliken calculations are also presented, and support the qualitative conclusions from the Heitler-London model. The effects need to be considered in spin-based quantum computer designs, either as a source of gate error to be overcome or a new interaction to be exploited.

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## I. INTRODUCTION

The exchange interaction between electrons has been studied since the early days of quantum mechanics<sup>1-3</sup> and has been reviewed in some classic references,<sup>4-6</sup> as well as textbooks.<sup>7</sup> Recently, a promising proposal<sup>8</sup> has emerged to use the exchange interaction as a tunable qubit-qubit interaction in a quantum computer, with the individual spins of electrons acting as qubits. To satisfy the conditions for constructing a universal quantum computer, the exchange interaction can either be supplemented with single-qubit operations,<sup>8</sup> or can be used by itself to construct a universal set of gates, in which case one encodes a logical qubit into the state of several spins.<sup>9-12</sup> (This alternative to the standard universality scheme has been termed “encoded universality.”<sup>13</sup>) Motivated by the proposal of Loss and DiVincenzo,<sup>8</sup> there have been a number of studies of the one- and two-particle behavior of electrons localized on quantum dots within a quantum computer.<sup>8,14-21</sup> Here, we expand on our work<sup>22</sup> considering the important situation of three or more coupled dots. We show how both quantitatively and qualitatively new interactions can appear, provide explicit formulas for the magnitudes of these interactions, and note consequences of breaking the symmetry of the system. These effects require consideration if one intends to make a quantum computer with more than two spins.

The exchange interaction between two localized electrons arises as a result of their spatial behavior, but it can be expressed as an effective spin-spin interaction. In conditions of rotation symmetry (i.e., neglecting external magnetic fields, spin-orbit coupling, etc.), a purely isotropic form of this interaction arises, which is known as Heisenberg exchange,

$$H_{\text{ex}} = JS_A \cdot S_B. \quad (1)$$

Here,  $\mathbf{S} = (S_x, S_y, S_z)$  is a vector of spin-half angular momentum operators, and  $A, B$  are indices referring to the location

of each electron. (We take spin operators to be dimensionless in this paper —  $\hbar$  is excluded from their definition.) This Hamiltonian has a spin-singlet eigenstate and degenerate spin-triplet eigenstates.<sup>1</sup> The quantity  $J$  is the exchange coupling constant, given by the energy splitting between the spin-singlet and spin-triplet states,<sup>1,14,16</sup>

$$J = \epsilon_t - \epsilon_s. \quad (2)$$

To date, studies of the exchange interaction in quantum computation have focused on the case of two quantum dots.<sup>8,14-21</sup> Starting from the simplest case of two electrons in singly occupied dots in the lowest orbital state, systematic generalizations have been introduced and their effect on the exchange interaction studied. In particular, researchers have analyzed the effect of double occupation,<sup>16,19,21</sup> higher orbital states,<sup>14,16</sup> and many-electron dots.<sup>17</sup> An accurate numerical study reporting singlet-triplet crossing via magnetic field manipulation in a lateral double quantum dot can be found in Ref. 23. Neglecting spin-orbit coupling, these studies have found increasingly accurate expressions for  $J$ , while focusing on the definition of Eq. (2). In the presence of spin-orbit coupling both rotation and inversion symmetry are broken, and anisotropic corrections to  $H_{\text{ex}}$  arise.<sup>20,24,25</sup>

In this work, we undertake a study of the case of three or four electrons, each in a quantum dot. Once the system involves more than two electrons, simultaneous multipartite exchanges can occur. For three coupled dots containing three electrons, processes in which all three electrons exchange contribute to a quantitative correction to the value of  $J$ . We show explicitly that, for three identical dots arranged on the corners of an equilateral triangle, the effective Hamiltonian can still be written using a Heisenberg exchange interaction

$$H_{\text{spin}} = K + J(\mathbf{S}_A \cdot \mathbf{S}_B + \mathbf{S}_B \cdot \mathbf{S}_C + \mathbf{S}_C \cdot \mathbf{S}_A),$$

but that  $J$  is changed and is found to be influenced by three-body exchange matrix elements. For four coupled dots containing four electrons, the actual form of the interaction Eq. (1) changes due to four-body effects. For identical dots arranged on the corners of a symmetric tetrahedron, the interaction takes the form

$$H_{\text{spin}} = K + J \sum_{A \leq i < j \leq D} \mathbf{S}_i \cdot \mathbf{S}_j + J' [(\mathbf{S}_A \cdot \mathbf{S}_B)(\mathbf{S}_C \cdot \mathbf{S}_D) + (\mathbf{S}_A \cdot \mathbf{S}_C)(\mathbf{S}_B \cdot \mathbf{S}_D) + (\mathbf{S}_A \cdot \mathbf{S}_D)(\mathbf{S}_B \cdot \mathbf{S}_C)],$$

where throughout this paper notation such as  $A \leq i \leq D$  means that  $i$  takes the letter values  $A$  to  $D$ . According to our Heitler-London (HL) calculations, the ratio  $|J'|/J|$  can reach 16 % in physically relevant parameter regimes.

Four-body exchange terms have been discussed in other contexts—for example, in a perturbative treatment of the two- and three-dimensional half-filled Hubbard models, four-body interactions were shown to suppress the Néel temperature and the temperature of the paraferromagnetic phase transition<sup>26,27</sup> (see Appendix A). Here, we present a nonperturbative derivation of these terms, starting from a finite-dimensional Hamiltonian and then highlighting their significance for quantum computation. Interaction Hamiltonian calculations such as ours are of significance in various quantum computation contexts, including (i) the encoded universality paradigm, where in the most efficient implementations, several exchange interactions are turned on simultaneously<sup>9–13</sup> (quantitative studies of parallel gate sequences<sup>11</sup> in particular may require revisiting in light of our results, as well as the “supercoherent qubits” method for reducing decoherence,<sup>28</sup> where four- and eight-spin interactions must be turned on simultaneously in order to enact quantum logic gates between encoded qubits); (ii) adiabatic quantum computing,<sup>29</sup> where the final Hamiltonian for any nontrivial calculation inevitably includes simultaneous interactions between multiple qubits; (iii) fault-tolerant quantum error correction, where a higher degree of parallelism translates into a lower threshold for fault-tolerant quantum computation operations;<sup>30–34</sup> (iv) the “one-way” quantum computer proposal,<sup>35</sup> where all nearest-neighbor interactions in a cluster of coupled spins are turned on simultaneously in order to prepare a many-spin entangled state; and (v) the search for physical systems with intrinsic, topological fault tolerance, where systems with four-body interactions have been identified as having the sought-after properties.<sup>36</sup>

We begin with a general description of a finite-dimensional effective spin Hamiltonian in Sec. II. (This is compared to the standard, perturbative derivation in Appendix A.) Section III shows how to compute the parameters in the effective spin Hamiltonian, with detailed consideration of the two-electron, three-electron, and four-electron cases. We introduce a specific model and calculate the parameters quantitatively, for three and four electrons, in Sec. IV. Appendixes B and C contain relevant technical details.

## II. ELECTRON-SPIN-OPERATOR HAMILTONIAN

In this section we present general arguments concerning the form of the effective spin Hamiltonian, as it arises from  $n$  localized electrons interacting via the Coulomb force. We start with the familiar electronic Hamiltonian

$$H = \sum_{i=1}^n \frac{1}{2m} \mathbf{p}_i^2 + V(\mathbf{r}_i) + \sum_{i < j} \frac{e^2}{\kappa |\mathbf{r}_i - \mathbf{r}_j|} \equiv \sum_{i=1}^n h(\mathbf{r}_i) + \sum_{i < j} w(\mathbf{r}_i, \mathbf{r}_j), \quad (3)$$

where the first term is the kinetic energy, the second is the confining potential, and the third is the Coulomb interaction. The confining potential  $V(\mathbf{r})$  contains  $n$  energy minima, which give rise to the  $n$  dots. To understand the dynamics of  $n$  electron-spin qubits in  $n$  quantum dots, it is desirable to eliminate the spatial degrees of freedom, leaving an effective Hamiltonian composed of electron-spin operators only.

The first step in changing Hamiltonian (3) to an electron-spin-operator Hamiltonian is to fix a basis. We first consider the case of two electrons in two dots, labeled  $A$  and  $B$ . We do not allow for double occupancy of a dot and consider only a single low-energy orbital per dot labeled as  $\phi_A(\mathbf{r}) \equiv \langle \mathbf{r} | A \rangle$  and  $\phi_B(\mathbf{r}) \equiv \langle \mathbf{r} | B \rangle$ . Electrons 1 and 2 occupy these low-energy orbitals. Each electron can have spin-up or spin-down, hence each electron can represent a qubit. A state with, for example, electron 1 in orbital  $B$  with spin-up and electron 2 in orbital  $A$  with spin down is represented as  $|BA\rangle |\uparrow \downarrow\rangle$ . Since electrons are fermions, this state needs to be antisymmetrized; the full state of the two electrons takes the form of a Slater determinant

$$|\Psi(\downarrow \uparrow)\rangle = (|AB\rangle |\downarrow \uparrow\rangle - |BA\rangle |\uparrow \downarrow\rangle) \propto a_{A\downarrow}^\dagger a_{B\uparrow}^\dagger |\text{vac}\rangle. \quad (4)$$

Note that the order of spins in the state label  $\Psi(\downarrow \uparrow)$  indicates that the electron in orbital  $A$  has spin down and the electron in orbital  $B$  has spin up. In Eq. (4) we introduced second-quantized notation (ignoring normalization), with  $a_{A\downarrow}^\dagger$  creating an electron with spin down in orbital  $A$  and  $a_{B\uparrow}^\dagger$  creating an electron with spin up in orbital  $B$ . The four states  $|\Psi(s_A, s_B)\rangle$  form the two-electron basis. The same procedure applies to three electrons in three dots. There is again a single low-energy orbital per dot, labeled as  $\phi_A(\mathbf{r}) \equiv \langle \mathbf{r} | A \rangle$ ,  $\phi_B(\mathbf{r}) \equiv \langle \mathbf{r} | B \rangle$ , and  $\phi_C(\mathbf{r}) \equiv \langle \mathbf{r} | C \rangle$  for dots  $A$ ,  $B$ , and  $C$ , respectively. Electrons 1, 2, and 3 occupy these low-energy orbitals. A state with, for example, electron 1 in orbital  $B$  with spin up, electron 2 in orbital  $A$  with spin down, and electron 3 in orbital  $C$  with spin up is represented as  $|BAC\rangle |\uparrow \downarrow \uparrow\rangle$ . This state is then antisymmetrized so that the full state of the three electrons is the Slater determinant

$$\begin{aligned} |\Psi(\downarrow \uparrow \uparrow)\rangle &\equiv (|ABC\rangle |\downarrow \uparrow \uparrow\rangle + |BCA\rangle |\uparrow \downarrow \uparrow\rangle + |CAB\rangle |\uparrow \downarrow \uparrow\rangle \\ &\quad - |BAC\rangle |\uparrow \downarrow \uparrow\rangle - |ACB\rangle |\downarrow \uparrow \uparrow\rangle - |CBA\rangle |\uparrow \downarrow \uparrow\rangle) \\ &\propto a_{A\downarrow}^\dagger a_{B\uparrow}^\dagger a_{C\uparrow}^\dagger |\text{vac}\rangle. \end{aligned} \quad (5)$$

The order of spins in the state label  $\Psi(\downarrow \uparrow \uparrow)$  indicates that the electron in orbital  $A$  has spin down, the electron in orbital  $B$  has spin up, and the electron in orbital  $C$  has spin up. The eight states  $|\Psi(s_A, s_B, s_C)\rangle$  form the three-electron basis.

The general case of  $n$  electrons gives  $2^n$  fully antisymmetrized basis vectors of the form

$$|\Psi(s_A, s_B, \dots, s_Z)\rangle = \sum_P \delta_P P[|AB\dots\rangle]_{s_A s_B \dots}, \quad (6)$$

where the sum runs over all permutations  $P$  of both orbitals and spins, and  $\delta_P = 1(-1)$  if the permutation is even (odd). In this basis, the Hamiltonian (3) takes the form of a  $2^n \times 2^n$  Hermitian matrix. Like any  $2^n \times 2^n$  Hermitian matrix, the Hamiltonian can be written as a sum

$$H_{\text{spin}} = \sum_{i,j,\dots=0}^3 l_{i,j,\dots} \sigma_i(A) \otimes \sigma_j(B) \otimes \dots \quad (7)$$

of Hermitian spin matrices of the form  $\sigma_i(A) \otimes \sigma_j(B) \otimes \dots$ , each multiplied by a real coefficient  $l_{i,j,\dots}$ . Here,  $\sigma_i(p)$  denotes the Pauli matrix  $\sigma_i$  acting on the electron in dot  $p$ , with  $i = 0, 1, 2, 3$  and with  $\sigma_0$  equal to the identity matrix. There are  $n$  factors in the tensor product  $\sigma_i(A) \otimes \sigma_j(B) \otimes \dots$ , so that it can be written as a  $2^n \times 2^n$  matrix, and there are similarly  $n$  subscripts on the coefficient  $l_{i,j,\dots}$ . This decomposition (7) into spin matrices produces an effective electron-spin Hamiltonian that conveniently describes the dynamics of  $n$  qubits.

The procedure we have just described is framed within the Heiter-London approximation.<sup>37</sup> The approximation consists of neglecting excited states and has been criticized on the grounds that it does not produce the correct asymptotic behavior in the limit of very large distances.<sup>4</sup> However, in the context of our system of interest, this asymptotic limit is not a concern, and moreover, recent studies have verified the utility of the approximation in the case of large (but not infinite) interdot separation.<sup>16</sup> We will thus proceed with the HL approximation, which has the advantage of conceptual simplicity and physical clarity. In the three-electron case, we show that Hund-Mülliken (HM) calculations, in which double occupation is permitted, support the conclusions of our HL results.

Symmetry considerations fundamentally constrain the form of the electron-spin Hamiltonian. The coordinate system used to define  $\uparrow$  and  $\downarrow$  is arbitrary if there is no spin-orbit coupling and no external magnetic field. In this case, the effective spin-operator Hamiltonian has rotation, inversion, and exchange symmetry. The coefficients  $l_{i,j,\dots}$  in (7) are strongly constrained by this symmetry. The Hamiltonian can only be a function of the total spin squared  $\mathbf{S}_T^2 = (\mathbf{S}_A + \mathbf{S}_B + \dots)^2$ , where

$\mathbf{S}_A \equiv \frac{1}{2}[\sigma_1(A)\hat{x} + \sigma_2(A)\hat{y} + \sigma_3(A)\hat{z}] \otimes \sigma_0(B) \otimes \sigma_0(C) \otimes \dots$ ,  
 $\mathbf{S}_B \equiv \sigma_0(A) \otimes \frac{1}{2}[\sigma_1(B)\hat{x} + \sigma_2(B)\hat{y} + \sigma_3(B)\hat{z}] \otimes \sigma_0(C) \otimes \dots$ , etc.  
 A pseudoscalar, such as  $\mathbf{S}_A \cdot (\mathbf{S}_B \times \mathbf{S}_C)$ , cannot appear in the Hamiltonian because of inversion symmetry. We must have

$$H_{\text{spin}} = L_0 + L_1 \mathbf{S}_T^2 + L_2 (\mathbf{S}_T^2)^2 + \dots, \quad (8)$$

where  $L_0, L_1, L_2, \dots$  are real constants with dimensions of energy. The constant  $L_0$  is an energy shift. The term proportional to  $L_1$  gives rise to the familiar Heisenberg interaction. Here we see that, in principle, higher-order interactions may be present in the spin Hamiltonian, starting with a fourth-order term proportional to  $L_2$ . In this highly symmetric situ-

ation, the eigenstates of the spin Hamiltonian are clearly just eigenstates of  $\mathbf{S}_T$ .

### III. COMPUTATION OF THE SPIN HAMILTONIAN PARAMETERS

To compute the values of  $L_0, L_1, L_2, \dots$  we consider an eigenstate  $|\Psi\rangle$  of  $\mathbf{S}_T^2$ , with known eigenvalue  $S_T(S_T+1)$ . If there are  $n$  electrons in the system, we write  $|\Psi\rangle = |\Psi_{S_T}^n\rangle$ . To proceed, one (i) computes the expectation value of the effective spin Hamiltonian (8) in this state, (ii) computes the expectation value of the spatial Hamiltonian (3) in this state, and then (iii) equates the two expectation values

$$\langle \Psi | H_{\text{spin}} | \Psi \rangle = \langle \Psi | H | \Psi \rangle. \quad (9)$$

This procedure is repeated for all eigenvalues of  $\mathbf{S}_T^2$ , thus generating a set of linear equations for the parameters  $L_0, L_1, L_2, \dots$ , in terms of matrix elements of  $H$  between different orbital states. For  $n$  electrons the number of distinct eigenvalues of  $\mathbf{S}_T^2$  is  $[n/2]+1$  (where  $[n/2]$  denotes the greatest integer less than  $n/2$ ), so this is the maximum number of distinct energy eigenvalues of the Hamiltonian (8). Thus, the coefficients  $L_m$  for  $0 \leq m < [n/2]+1$  have enough degrees of freedom to completely and uniquely specify the matrix (8); without loss of generality, we can set  $L_m = 0$  for  $m \geq [n/2]+1$ . We are led to  $[n/2]+1$  coupled linear equations for the nonzero  $L_m$  parameters. In the case that  $n$  is even,  $S_T$  takes on the integer values  $0, 1, \dots, n/2$ . In the case that  $n$  is odd,  $S_T$  takes on the half-integer values  $1/2, 3/2, \dots, n/2$ . We have

$$\langle \Psi_{S_T}^n | H_{\text{spin}} | \Psi_{S_T}^n \rangle = \sum_{m=0}^{[n/2]} L_m [S_T(S_T+1)]^m. \quad (10)$$

Having completed step (i) of our program, we now turn to step (ii), the calculation of  $\langle \Psi_{S_T}^n | H | \Psi_{S_T}^n \rangle$ . We make this calculation separately for the cases of two, three, and four electrons.

#### A. Two-electron case

As a simple illustration of our procedure we rederive the well-known result for two electrons: the exchange constant equals the difference between the (degenerate) triplet states and the singlet state. The spin singlet ( $S_T=0$ ) and spin triplet ( $S_T=1$ ) states have eigenvalues of  $\mathbf{S}_T^2$  equal to 0 and 2, respectively. Thus, the Hamiltonian (8) can only have two distinct eigenvalues, and we need to solve  $[2/2]+1=2$  equations for  $L_0$  and  $L_1$ . A convenient  $S_T=1$  eigenstate is the normalized state  $|\Psi_{S_T=1}^{n=2}\rangle \equiv \mathcal{N}(|\Psi(\uparrow\uparrow)\rangle) = \mathcal{N}(|AB\rangle|\uparrow\uparrow\rangle - |BA\rangle|\uparrow\uparrow\rangle) = \mathcal{N}(|AB\rangle - |BA\rangle)|\uparrow\uparrow\rangle$ . The normalization constant  $\mathcal{N}$  has the value

$$\mathcal{N} = (\langle AB|AB\rangle + \langle BA|BA\rangle - \langle AB|BA\rangle - \langle BA|AB\rangle)^{-1/2}.$$

Inserting this state into Eq. (9) yields

$$\langle \Psi_{S_T=1}^{n=2} | H_{\text{spin}} | \Psi_{S_T=1}^{n=2} \rangle = \langle \Psi_{S_T=1}^{n=2} | H | \Psi_{S_T=1}^{n=2} \rangle.$$

The spin Hamiltonian's expectation value is immediately found to be  $L_0 + 2L_1$ , as can be seen from Eq. (10). Expand-

ing out the spatial Hamiltonian's expectation value gives

$$L_0 + 2L_1 = \frac{\langle AB|H|AB\rangle + \langle BA|H|BA\rangle - \langle AB|H|BA\rangle - \langle BA|H|AB\rangle}{\langle AB|AB\rangle + \langle BA|BA\rangle - \langle AB|BA\rangle - \langle BA|AB\rangle}, \quad (11)$$

which can be evaluated once a choice of orbital states is specified; we do this in Sec. IV. To compare with Eq. (2), we note that this equation specifies the triplet energy  $\epsilon_t \equiv L_0 + 2L_1$ . A second equation is found from the  $S_T=0$  state  $|\Psi_{S_T=0}^n\rangle = \mathcal{N}[|\Psi(\uparrow\downarrow)\rangle - |\Psi(\downarrow\uparrow)\rangle] = \mathcal{N}(|AB\rangle + |BA\rangle)(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$ , which leads to

$$L_0 = \frac{\langle AB|H|AB\rangle + \langle BA|H|BA\rangle + \langle AB|H|BA\rangle + \langle BA|H|AB\rangle}{\langle AB|AB\rangle + \langle BA|BA\rangle + \langle AB|BA\rangle + \langle BA|AB\rangle}, \quad (12)$$

giving the singlet energy  $\epsilon_s = L_0$ . To exhibit the exchange coupling explicitly, we rewrite the Hamiltonian as

$$H_{\text{spin}} = L_0 + L_1 \mathbf{S}_A^2 + L_1 \mathbf{S}_B^2 + 2L_1 \mathbf{S}_A \cdot \mathbf{S}_B \equiv K + J \mathbf{S}_A \cdot \mathbf{S}_B, \quad (13)$$

where  $K = L_0 + (3/2)L_1$  and  $J = 2L_1$ . Expression (2) follows when we note that  $L_1 = (\epsilon_t - \epsilon_s)/2$ .

## B. Three-electron case

### 1. Heitler-London model

In the three-electron case, the possible values that the total spin can take are  $S_T=1/2$  (with two, two-dimensional eigenspaces) or  $S_T=3/2$  (with a four-dimensional eigenspace). We, therefore, again need to solve  $[3/2]+1=2$  equations, and it is sufficient to keep only two constants  $L_0$  and  $L_1$  in  $H_{\text{spin}}$ , setting  $L_2$  and the rest to zero. As a convenient state with known  $S_T=3/2$  we take the normalized state  $|\Psi_{3/2}^3\rangle \propto |\Psi(\uparrow\uparrow\uparrow)\rangle$ , so that the energy is  $E_{3/2} \equiv \langle \Psi_{3/2}^3 | H_{\text{spin}} | \Psi_{3/2}^3 \rangle = L_0 + L_1(3/2)(5/2)$ . We use  $|\Psi_{1/2}^3\rangle \propto [|\Psi(\uparrow\downarrow\uparrow)\rangle - |\Psi(\downarrow\uparrow\uparrow)\rangle]/\sqrt{2}$  as a normalized state with known  $S_T=1/2$ , for which the energy is  $E_{1/2} \equiv \langle \Psi_{1/2}^3 | H_{\text{spin}} | \Psi_{1/2}^3 \rangle = L_0 + L_1(1/2)(3/2)$ . Then equating expectation values of Hamiltonian (3) and Hamiltonian (8), i.e., requiring  $\langle \Psi_{S_T}^3 | H_{\text{spin}} | \Psi_{S_T}^3 \rangle = \langle \Psi_{S_T}^3 | H | \Psi_{S_T}^3 \rangle$  for each of our states  $S_T=1/2$  and  $S_T=3/2$  as in Eq. (9), we can solve for  $L_0$  and  $L_1$ . To do so we need to obtain more explicit expressions for  $\langle \Psi_{S_T}^3 | H | \Psi_{S_T}^3 \rangle$ . We assume that  $\phi_A(\mathbf{r})$ ,  $\phi_B(\mathbf{r})$ , and  $\phi_C(\mathbf{r})$  are real and satisfy  $\langle A|A\rangle = \langle B|B\rangle = \langle C|C\rangle$  and  $\langle A|B\rangle = \langle A|C\rangle = \langle B|C\rangle$  (this is consistent with our original assumption of rotational invariance, inversion invariance, and equilateral triangle geometry). First, let us normalize  $|\Psi_{3/2}^3\rangle$

$$|\Psi_{3/2}^3\rangle = \mathcal{N}[|ABC\rangle + |CAB\rangle + |BCA\rangle - |BAC\rangle - |CBA\rangle - |ACB\rangle]|\uparrow\uparrow\uparrow\rangle,$$

where the normalization constant  $\mathcal{N}$  is given by

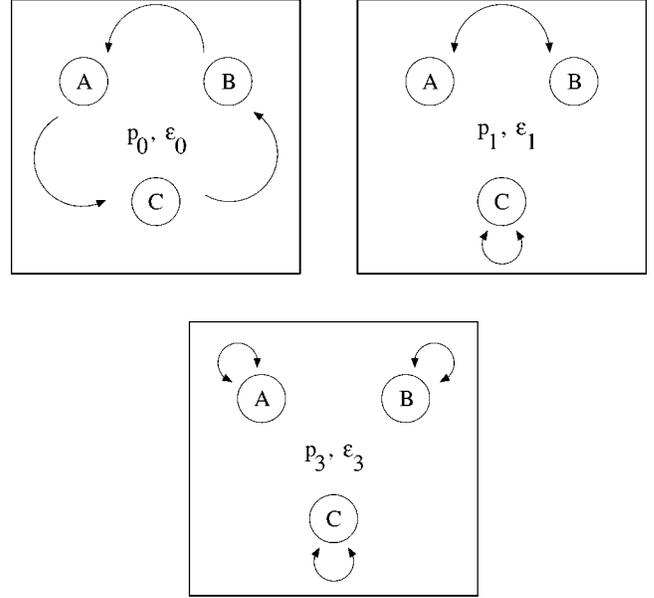


FIG. 1. Matrix elements relevant to three-electron case. Arrows indicate transition from localized state on initial dot to localized state on final dot.

$$\mathcal{N} = \frac{1}{\sqrt{6(p_3 + 2p_0 - 3p_1)}}.$$

The quantities  $p_3$ ,  $p_1$ , and  $p_0$  are given by

$$p_3 = \langle ABC|ABC\rangle,$$

which is an overlap integral when all three electrons retain the same state in the bra and ket,

$$p_1 = \langle BAC|ABC\rangle = \langle CBA|ABC\rangle = \langle ACB|ABC\rangle,$$

which is an overlap integral when one electron has the same state in the bra and ket, and

$$p_0 = \langle CAB|ABC\rangle = \langle BCA|ABC\rangle,$$

which is an overlap integral when zero electrons have the same state in the bra and ket — all three electrons change their states. In evaluating the matrix element  $\langle \Psi_{3/2}^3 | H | \Psi_{3/2}^3 \rangle$  we use the notation

$$\epsilon_0 = \langle CAB|H|ABC\rangle = \langle BCA|H|ABC\rangle,$$

$$\epsilon_1 = \langle BAC|H|ABC\rangle = \langle CBA|H|ABC\rangle = \langle ACB|H|ABC\rangle,$$

$$\epsilon_3 = \langle ABC|H|ABC\rangle,$$

where the physical interpretation is that  $\epsilon_k$  involves  $3-k$  electrons exchanging orbitals (Fig. 1).

Computing the expectation value of  $H$  in the state  $|\Psi_{3/2}^3\rangle$  then leads to the result

$$E_{3/2} = L_0 + \frac{15}{4}L_1 = \frac{\epsilon_3 + 2\epsilon_0 - 3\epsilon_1}{p_3 + 2p_0 - 3p_1}. \quad (14)$$

For the case  $S_T=1/2$ , using  $|\Psi_{1/2}^3\rangle$  an analogous calculation yields

$$E_{1/2} = L_0 + \frac{3}{4}L_1 = \frac{\epsilon_3 - \epsilon_0}{p_3 - p_0}. \quad (15)$$

These equations give  $L_0$  and  $L_1$  in terms of the  $p_i$  and  $\epsilon_i$ .

To compute the usual exchange coupling, it is useful to rewrite  $H_{\text{spin}}$  as

$$\begin{aligned} H_{\text{spin}} &= \left( L_0 + L_1 \sum_{A \leq i \leq C} \mathbf{S}_i^2 \right) + 2L_1 \sum_{A \leq i < j \leq C} \mathbf{S}_i \cdot \mathbf{S}_j \\ &\equiv K + J(\mathbf{S}_A \cdot \mathbf{S}_B + \mathbf{S}_A \cdot \mathbf{S}_C + \mathbf{S}_B \cdot \mathbf{S}_C), \end{aligned} \quad (16)$$

where

$$K = L_0 + \frac{3}{4}L_1, \quad (17)$$

$$J = 2L_1. \quad (18)$$

Solving for the exchange constant  $J=2L_1$  we find, finally,

$$J = \frac{2}{3}(E_{3/2} - E_{1/2}).$$

A couple of comments are in order concerning this result. First, the energies  $E_{3/2}, E_{1/2}$  can be calculated once the orbitals are specified, as we do in Sec. IV below. We see that, similar to the two-electron case, the physical interpretation of the exchange constant is that (up to a multiplicative factor) it is given by the energy difference between the  $S_T=3/2$  and  $S_T=1/2$  states. Second, note from Eqs. (14) and (15) that the value of the exchange constant  $J$  is determined in part by the ‘‘three-electron-exchange’’ terms of the form  $p_0 = \langle CAB|ABC \rangle$  and  $\epsilon_0 = \langle CAB|H|ABC \rangle$ . It is apparent that such terms involve a cooperative effect between all three electrons and hence cannot be seen in two-electron calculations. It follows that *the presence of the third electron quantitatively changes the exchange coupling between the other two electrons.*

## 2. Hund-Mülliken model

We have been working within the HL approximation in which there is one orbital per quantum dot occupied by a single electron. To check its physical validity, we make three-electron computations within the HM approximation as well, in which double occupation of quantum dots is permitted. This leads to a total of  $8+12=20$  basis states in the three-spin case ( $2^3=8$  from the HL basis and  $3 \times 2 \times 2=12$  double-occupation states). In the HL approximation, the eight states divide into a degenerate four-dimensional  $S=3/2$  subspace with energy  $E_{3/2}$  and a degenerate four-dimensional  $S=1/2$  subspace with energy  $E_{1/2}$ . In the HM case, the degenerate four-dimensional  $S=3/2$  subspace is unaffected by the double-occupation states, which must all have  $S=1/2$ ; the energy of these four  $S=3/2$  states remains  $E_{3/2}$ . (The  $S=3/2, S_z=3/2$  state has three spin-up electrons and so the Hamiltonian cannot mix it with any other state. Since the other  $S=3/2$  states are related by a rotation generated by the total spin operator, which commutes with the Hamiltonian, they must be eigenstates of the Hamiltonian with the same energy.) The 12 double-occupation states enlarge the  $S=1/2$  subspace, which becomes 16-dimensional and has a nontrivial spectrum.

In the HM case, the decomposition (7) is no longer meaningful because the basis states do not necessarily have one spin per quantum dot. This complicates the computation of the eigenspectrum of this 16-dimensional space. First, we note that the projection  $S_z$  (the number of spin-up electrons) is still a good quantum number because the Hamiltonian (3) cannot mix two states with different numbers of spin-up electrons. The 16-dimensional subspace, therefore, splits into two degenerate eight-dimensional  $S_z = \pm 1/2$  subspaces. The  $S_z=1/2$  subspace consists of two HL states and six double-occupation states analogous to (5)

$$|\Psi_{1/2}^3\rangle \propto \frac{1}{\sqrt{2}}(|\Psi(\uparrow\downarrow\uparrow)\rangle - |\Psi(\downarrow\uparrow\uparrow)\rangle),$$

$$|\Phi_{1/2}^3\rangle \propto \frac{2}{\sqrt{6}}|\Psi(\uparrow\uparrow\downarrow)\rangle - \frac{1}{\sqrt{6}}(|\Psi(\uparrow\downarrow\uparrow)\rangle + |\Psi(\downarrow\uparrow\uparrow)\rangle),$$

$$|\Psi_{AAB}(\uparrow\downarrow\uparrow)\rangle \propto a_{A1}^\dagger a_{A1}^\dagger a_{B1}^\dagger |\text{vac}\rangle, |\Psi_{AAC}(\uparrow\downarrow\uparrow)\rangle, |\Psi_{BBA}(\uparrow\downarrow\uparrow)\rangle,$$

$$|\Psi_{BBC}(\uparrow\downarrow\uparrow)\rangle, |\Psi_{CCA}(\uparrow\downarrow\uparrow)\rangle, |\Psi_{CCB}(\uparrow\downarrow\uparrow)\rangle. \quad (19)$$

One can construct the  $8 \times 8$  Hamiltonian in this subspace and diagonalize it. The eigenstates exhibit degeneracies arising from the symmetry of the Hamiltonian under the exchange of a pair of dots. Assuming that our dots are all equivalent, there are three dot-pair exchange operators that commute with the Hamiltonian:  $E_{A,B}$  that exchanges dots  $A, B$ ;  $E_{B,C}$  that exchanges dots  $B, C$ ; and  $E_{C,A}$  that exchanges dots  $C, A$ . For instance,  $E_{A,B}|\Psi_{AAC}(\uparrow\downarrow\uparrow)\rangle = |\Psi_{BBA}(\uparrow\downarrow\uparrow)\rangle$ . We can require that the eigenstates of the Hamiltonian also be eigenstates of  $E_{A,B}$  or  $E_{B,C}$  or  $E_{C,A}$ . Using our eight states (19), it is possible to construct two linearly independent states that are simultaneous eigenstates of all three exchange operators. The two (unnormalized) eigenstates are

$$\begin{aligned} &(|\Psi_{AAB}(\uparrow\downarrow\uparrow)\rangle + |\Psi_{AAC}(\uparrow\downarrow\uparrow)\rangle + |\Psi_{BBA}(\uparrow\downarrow\uparrow)\rangle + |\Psi_{BBC}(\uparrow\downarrow\uparrow)\rangle \\ &+ |\Psi_{CCA}(\uparrow\downarrow\uparrow)\rangle + |\Psi_{CCB}(\uparrow\downarrow\uparrow)\rangle) \end{aligned}$$

and

$$\begin{aligned} &(|\Psi_{AAB}(\uparrow\downarrow\uparrow)\rangle - |\Psi_{AAC}(\uparrow\downarrow\uparrow)\rangle - |\Psi_{BBA}(\uparrow\downarrow\uparrow)\rangle + |\Psi_{BBC}(\uparrow\downarrow\uparrow)\rangle \\ &+ |\Psi_{CCA}(\uparrow\downarrow\uparrow)\rangle - |\Psi_{CCB}(\uparrow\downarrow\uparrow)\rangle), \end{aligned}$$

with eigenvalue  $+1$  and  $-1$ , respectively. Each such state turns out to be an eigenstate of the Hamiltonian with its own nondegenerate energy. The remaining six members of the eight-dimensional subspace are not simultaneous eigenstates of all three exchange operators:  $E_{A,B}$  and  $E_{B,C}$  and  $E_{C,A}$ . To ensure that we can nevertheless choose the eigenstates of the Hamiltonian to be simultaneous eigenstates of  $E_{A,B}$  or  $E_{B,C}$  or  $E_{C,A}$ , the energy eigenstates occur in degenerate pairs that can be superposed as desired to form eigenstates of the exchange operators. When the parameters of the spatial Hamiltonian (3) make double occupation energetically expensive, one of the degenerate pairs will be low in energy and will consist mainly of the HL states  $|\Psi_{1/2}^3\rangle$  and  $|\Phi_{1/2}^3\rangle$ . In this way, the HM calculation reduces to the HL result plus high-energy double-occupation states, and (16) still describes the low-energy spin dynamics.

### 3. Unequal Coupling

We emphasize that Eq. (16) was derived assuming rotation, inversion, and exchange symmetry. Exchange symmetry, in particular, is broken whenever there is unequal coupling between dots, and then the Hamiltonian can involve more constants. This situation is realized when the dots are not all equidistant or when they have been shifted electrically, as in the case of dots defined by electrodes creating confinement potentials,<sup>38</sup> or when there are unequal tunneling barriers between different dots.<sup>14</sup> For instance, in the case of three unequally coupled dots, the Hamiltonian will have the form

$$H_{\text{spin}} = K + J_{AB}\mathbf{S}_A \cdot \mathbf{S}_B + J_{BC}\mathbf{S}_B \cdot \mathbf{S}_C + J_{AC}\mathbf{S}_A \cdot \mathbf{S}_C$$

if we still assume rotation and inversion invariance. (An external magnetic field, which has been shown to be instrumental in changing the sign of  $J$  in the case of two dots,<sup>14,16</sup> could lead to a very different Hamiltonian. It would break rotation symmetry, introducing operators into  $H_{\text{spin}}$ , such as  $S_T^z$ .) This three electron Hamiltonian commutes with the  $z$ -component of the total spin operator  $S_T$ , so they can be simultaneously diagonalized. The state  $|\Psi(\downarrow\downarrow\downarrow)\rangle$  has  $S_T^z = -\frac{3}{2}$  and energy eigenvalue  $K + (J_{AB} + J_{BC} + J_{AC})/4$ . It is found to be degenerate with  $(|\Psi(\downarrow\downarrow\uparrow)\rangle + |\Psi(\downarrow\uparrow\downarrow)\rangle + |\Psi(\uparrow\downarrow\downarrow)\rangle)/\sqrt{3}$ , a state with  $S_T^z = -\frac{1}{2}$ . There are two remaining  $S_T^z = -\frac{1}{2}$  eigenvectors, which have the (unnormalized) forms

$$\left( \frac{J_{BC} - J_{AB} + \tilde{J}}{J_{AB} - J_{AC}} |\Psi(\downarrow\downarrow\uparrow)\rangle + \frac{J_{BC} - J_{AC} + \tilde{J}}{J_{AB} - J_{AC}} |\Psi(\downarrow\uparrow\downarrow)\rangle + |\Psi(\uparrow\downarrow\downarrow)\rangle \right),$$

$$\left( \frac{J_{AB} - J_{BC} + \tilde{J}}{J_{AC} - J_{AB}} |\Psi(\downarrow\downarrow\uparrow)\rangle + \frac{J_{AC} - J_{BC} + \tilde{J}}{J_{AB} - J_{AC}} |\Psi(\downarrow\uparrow\downarrow)\rangle + |\Psi(\uparrow\downarrow\downarrow)\rangle \right)$$

and have energies  $a + \tilde{J}, a - \tilde{J}$  respectively, where  $a \equiv -3(J_{AB} + J_{BC} + J_{AC})/2$  and  $\tilde{J} \equiv (J_{AB}^2 + J_{BC}^2 + J_{AC}^2 - J_{AC}J_{BC} - J_{AB}J_{AC} - J_{AB}J_{BC})^{1/2}$ . The remaining four energy eigenvectors, with  $S_T^z = \frac{3}{2}$  and  $S_T^z = \frac{1}{2}$ , can be obtained from these four by inversion. From these results it is possible to derive equations analogous to (14)–(18) in the case when  $J_{AB}$ ,  $J_{BC}$ , and  $J_{AC}$  are not equal.

#### C. Four-electron case

In the case of four electrons, the effective Hamiltonian again takes the form (8). Since four electrons can have  $S_T = 0$ ,  $S_T = 1$ , or  $S_T = 2$ , we must keep three constants  $L_0$ ,  $L_1$ , and  $L_2$  in  $H_{\text{spin}}$ . It follows immediately that  $H_{\text{spin}}$  includes terms of the form  $L_2(\mathbf{S}_A \cdot \mathbf{S}_B)(\mathbf{S}_C \cdot \mathbf{S}_D)$  and permutations. Unless  $L_2$  happens to vanish, the presence of a fourth electron introduces a qualitatively different four-body interaction as well

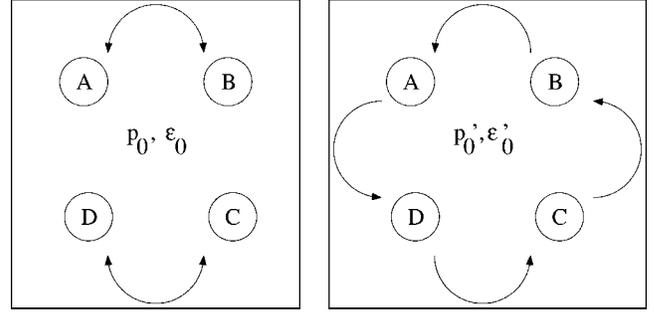


FIG. 2. Selected matrix elements relevant to four-electron case. Arrows indicate transition from localized state on initial dot to localized state on final dot.

as a quantitative change in the exchange coupling between the other electrons.

We now calculate  $L_0$ ,  $L_1$ , and  $L_2$  just as we calculated  $L_0$  and  $L_1$  for three particles. Let us define

$$p_0 = \langle BADC | ABCD \rangle, \quad \epsilon_0 = \langle BADC | H | ABCD \rangle,$$

$$p'_0 = \langle DABC | ABCD \rangle, \quad \epsilon'_0 = \langle DABC | H | ABCD \rangle,$$

$$p_1 = \langle ADBC | ABCD \rangle, \quad \epsilon_1 = \langle ADBC | H | ABCD \rangle,$$

$$p_2 = \langle BACD | ABCD \rangle, \quad \epsilon_2 = \langle BACD | H | ABCD \rangle,$$

$$p_4 = \langle ABCD | ABCD \rangle, \quad \epsilon_4 = \langle ABCD | H | ABCD \rangle,$$

where the subscript indicates how many electrons retain the same state in the bra and the ket, just as in the three-electron case. The terms  $\epsilon_0$  and  $\epsilon'_0$  involve four-body effects:  $\epsilon_0$  involves two pairs of electrons exchanging orbitals and  $\epsilon'_0$  involves all four electrons exchanging orbitals cyclically (Fig. 2).

A convenient state to use for  $S_T = 0$  is  $|\Psi_0^4\rangle = \mathcal{N}(|\Psi(\uparrow\downarrow\uparrow\downarrow)\rangle - |\Psi(\uparrow\downarrow\downarrow\uparrow)\rangle - |\Psi(\uparrow\uparrow\downarrow\downarrow)\rangle + |\Psi(\downarrow\uparrow\uparrow\downarrow)\rangle)$ , keeping in mind the definition (6). After normalization, this state yields the singlet energy

$$E_0 = L_0 = \frac{\epsilon_4 - 4\epsilon_1 + 3\epsilon_0}{p_4 - 4p_1 + 3p_0}. \quad (20)$$

A convenient state to use for  $S_T = 1$  is  $|\Psi_1^4\rangle = \mathcal{N}(|\Psi(\uparrow\downarrow\uparrow\downarrow)\rangle + |\Psi(\uparrow\downarrow\downarrow\uparrow)\rangle - |\Psi(\downarrow\uparrow\uparrow\downarrow)\rangle - |\Psi(\downarrow\uparrow\downarrow\uparrow)\rangle)$ . This state, after normalization, yields the triplet energy

$$E_1 = L_0 + 2L_1 + 4L_2 = \frac{\epsilon_4 - 2\epsilon_2 - \epsilon_0 + 2\epsilon'_0}{p_4 - 2p_2 - p_0 + 2p'_0}. \quad (21)$$

Finally, a convenient state to use for  $S_T = 2$  is  $|\Psi(\uparrow\uparrow\uparrow\uparrow)\rangle$ . We find for the quintet energy

$$E_2 = L_0 + 6L_1 + 36L_2 = \frac{\epsilon_4 - 6\epsilon_2 + 8\epsilon_1 + 3\epsilon_0 - 6\epsilon'_0}{p_4 - 6p_2 + 8p_1 + 3p_0 - 6p'_0}. \quad (22)$$

Solving, we have

$$L_0 = E_0,$$

$$L_1 = -\frac{1}{12}(E_2 - 9E_1 + 8E_0),$$

$$L_2 = \frac{1}{24}(E_2 - 3E_1 + 2E_0).$$

We would like to exhibit interaction constants explicitly in the spin Hamiltonian. We have  $S_T = \sum_{i=A}^D \mathbf{S}_i$ , so that

$$S_T^2 = 3I + 2 \sum_{i<j} \mathbf{S}_i \cdot \mathbf{S}_j,$$

while

$$\begin{aligned} (S_T^2)^2 &= \left( 3I + \sum_{A \leq i \neq j \leq D} \mathbf{S}_i \cdot \mathbf{S}_j \right)^2 \\ &= 9I + 6 \sum_{i \neq j} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i \neq j} \mathbf{S}_i \cdot \mathbf{S}_j \sum_{k \neq l} \mathbf{S}_k \cdot \mathbf{S}_l, \end{aligned}$$

and it can be shown that

$$\begin{aligned} \sum_{i \neq j} \mathbf{S}_i \cdot \mathbf{S}_j \sum_{k \neq l} \mathbf{S}_k \cdot \mathbf{S}_l &= \sum_{i \neq j \neq k \neq l} (\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l) \\ &\quad + 4 \sum_{i \neq j \neq l} (\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_j \cdot \mathbf{S}_l) \\ &\quad + 2 \sum_{i \neq j} (\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_i \cdot \mathbf{S}_j) \\ &= \sum_{i \neq j \neq k \neq l} (\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l) + 4 \left( \frac{1}{2} \sum_{i \neq j} \mathbf{S}_i \cdot \mathbf{S}_j \right) \\ &\quad + 2 \left( \frac{9}{4} - \frac{1}{2} \sum_{i \neq j} \mathbf{S}_i \cdot \mathbf{S}_j \right). \end{aligned}$$

We are led to

$$(S_T^2)^2 = \frac{27}{2} + 7 \sum_{i \neq j} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i \neq j \neq k \neq l} (\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l).$$

The spin Hamiltonian can now be written as follows:

$$\begin{aligned} H_{\text{spin}} &= K + J \sum_{i<j} \mathbf{S}_i \cdot \mathbf{S}_j + J' [(\mathbf{S}_A \cdot \mathbf{S}_B)(\mathbf{S}_C \cdot \mathbf{S}_D) \\ &\quad + (\mathbf{S}_A \cdot \mathbf{S}_C)(\mathbf{S}_B \cdot \mathbf{S}_D) + (\mathbf{S}_A \cdot \mathbf{S}_D)(\mathbf{S}_B \cdot \mathbf{S}_C)], \end{aligned}$$

where

$$K = L_0 + 3L_1 + \frac{27L_2}{2} = \frac{-2E_0 + 9E_1 + 5E_2}{16}, \quad (23)$$

$$J = 2L_1 + 14L_2 = \frac{-2E_0 - 3E_1 + 10E_2}{12}, \quad (24)$$

$$J' = 8L_2 = \frac{-2E_0 - 3E_1 + E_2}{3}. \quad (25)$$

Generically,  $J'$  does not vanish, and four-body interactions arise. The physical interpretation of the exchange constants as simple energy differences between different spin multiplets is now lost; we find energy differences with numerical coefficients that are not intuitively obvious.

Of central physical importance to us is the relative sizes of the coefficients  $J$  and  $J'$ . This is studied in Sec. IV, where a HL calculation suggests that  $J'$  is substantial in comparison to  $J$  in physically important regions of parameter space. We also find that both coefficients are affected by three-body ( $p_1, \epsilon_1$ ) and four-body exchanges ( $p_0, p'_0, \epsilon_0, \epsilon'_0$ ).

In the general case of  $2n$  electrons, two-body, four-body, ...,  $2n$ -body interaction terms appear in the Hamiltonian. Computing the strengths of the interactions for larger  $n$  is a topic of interest, but we do not address it here. One expects the strengths of the terms to decrease with the number of bodies involved.

#### IV. MODEL POTENTIAL CALCULATIONS

To compute the values of the  $L_i$ , we select the following specific form for the one-body potential in (3):

$$V(\mathbf{r}) = \frac{1}{2(2l)^6} m\omega_o^2 |\mathbf{r} - \mathbf{A}|^2 |\mathbf{r} - \mathbf{B}|^2 |\mathbf{r} - \mathbf{C}|^2 |\mathbf{r} - \mathbf{D}|^2. \quad (26)$$

This potential has a quadratic minimum at each of the vertices of an equilateral tetrahedron  $\mathbf{A} = (0, 0, 0)$ ,  $\mathbf{B} = (2l\sqrt{1/3}, 0, -2l\sqrt{2/3})$ ,  $\mathbf{C} = (-l\sqrt{1/3}, l, -2l\sqrt{2/3})$ , and  $\mathbf{D} = (-l\sqrt{1/3}, -l, -2l\sqrt{2/3})$ . The distance between vertices is  $2l$ . We select a potential with four minima so that it can be used in the four-electron case without modification. This facilitates comparison between the two-, three-, and four-electron cases, and the extra minima do not influence the two- and three-electron cases in any significant way.

At vertex  $\mathbf{A}$ , we define the localized Gaussian state as follows:

$$\phi_A(\mathbf{r}) \equiv \langle \mathbf{r} | A \rangle \equiv \left( \frac{m\omega_o}{\pi \hbar} \right)^{3/4} \exp \left( -\frac{m\omega_o}{2\hbar} |\mathbf{r} - \mathbf{A}|^2 \right),$$

which is the ground state of the quadratic minimum at that vertex. We define localized states similarly for the other vertices.

The following one-body Hamiltonian matrix elements are needed to evaluate the coupling constants in  $H_{\text{spin}}$ :

$$\langle A | A \rangle = 1,$$

$$\begin{aligned} \langle A | h | A \rangle &= \langle A | \frac{\mathbf{p}^2}{2m} + \frac{1}{2} m\omega_o^2 |\mathbf{r} - \mathbf{A}|^2 | A \rangle \\ &\quad + \langle A | V(\mathbf{r}) - \frac{1}{2} m\omega_o^2 |\mathbf{r} - \mathbf{A}|^2 | A \rangle \\ &= \hbar\omega_o \left[ \frac{3}{2} + \frac{15}{2048} (63x_b^{-3} + 280x_b^{-2} + 320x_b^{-1}) \right], \end{aligned}$$

$$\langle A | B \rangle = e^{-x_b},$$

$$\begin{aligned} \langle A | h | B \rangle &= \hbar\omega_o \left[ \frac{3}{2} e^{-x_b} + \frac{1}{2048} (945x_b^{-3} + 1680x_b^{-2} + 936x_b^{-1} \right. \\ &\quad \left. - 1216 - 880x_b) e^{-x_b} \right]. \quad (27) \end{aligned}$$

In these equations, we have added and subtracted a harmonic-oscillator potential from the one-body Hamiltonian  $h$  for ease of calculation. The dimensionless tunneling parameter  $x_b$  is the square of the ratio of the interdot distance  $2l$  to the characteristic harmonic-oscillator width  $2\sqrt{\hbar/(m\omega_0)}$

$$x_b \equiv m\omega_0 l^2/\hbar,$$

and which is also the ratio of the tunneling energy barrier  $m\omega_0^2 l^2/2$  to the harmonic-oscillator ground-state energy  $\hbar\omega_0/2$ .

Matrix elements of the Coulomb interaction are given by

$$\begin{aligned} \langle FG|w|UV\rangle &= \hbar\omega_0 \left[ 2x_c \sqrt{x_b} \frac{2\sqrt{2}}{|\mathbf{f} + \mathbf{u} - \mathbf{g} - \mathbf{v}|} e^{-(1/4)(|\mathbf{f} - \mathbf{u}|^2 + |\mathbf{g} - \mathbf{v}|^2)} \right. \\ &\quad \left. \times \operatorname{erf}\left(\frac{|\mathbf{f} + \mathbf{u} - \mathbf{g} - \mathbf{v}|}{2\sqrt{2}}\right) \right], \\ &|\mathbf{f} + \mathbf{u} - \mathbf{g} - \mathbf{v}| \neq 0 \end{aligned} \quad (28)$$

$$\begin{aligned} \langle FG|w|UV\rangle &= \hbar\omega_0 \left[ \frac{4}{\sqrt{\pi}} x_c \sqrt{x_b} e^{-(1/4)(|\mathbf{f} - \mathbf{u}|^2 + |\mathbf{g} - \mathbf{v}|^2)} \right], \\ &|\mathbf{f} + \mathbf{u} - \mathbf{g} - \mathbf{v}| = 0. \end{aligned} \quad (29)$$

In these equations, the dimensionless parameter  $x_c$  is the ratio of the Coulomb energy  $e^2/(\kappa 2l)$  to the harmonic oscillator ground state energy  $\hbar\omega_0/2$

$$x_c \equiv e^2/(\kappa \hbar\omega_0).$$

The symbols  $F, G, U$ , and  $V$  take values from the set  $\{A, B, C, D\}$ . The lowercase vectors are defined by  $\mathbf{f} \equiv \sqrt{m\omega_0/\hbar} \mathbf{F} = \sqrt{x_b}/l \mathbf{F}$ , etc. The symbol  $\operatorname{erf}(x) \equiv (2/\sqrt{\pi}) \int_0^x e^{-s^2} ds$  denotes the error function.

### A. Two electrons

In the case of two electrons, we assume that two of the potential minima of (26) are occupied; there is an electron at  $\mathbf{A} = (0, 0, 0)$  and an electron at  $\mathbf{B} = (2l\sqrt{1/3}, 0, -2l\sqrt{2/3})$ . In order to compute  $L_0$  and  $L_1$  from Eqs. (11) and (12), we require only the matrix elements  $\langle AB|H|AB\rangle = 2\langle A|h|A\rangle + \langle AB|w|AB\rangle$ ,  $\langle AB|H|BA\rangle = 2\langle A|h|B\rangle\langle A|B\rangle + \langle AB|w|BA\rangle$ ,  $\langle AB|AB\rangle = 1$ , and  $\langle AB|BA\rangle = \langle A|B\rangle^2$ . (We have simplified using the fact that  $\langle A|h|A\rangle = \langle B|h|B\rangle$  and using the fact that the wave functions are real.) Once  $L_0$  and  $L_1$  have been computed, it is straightforward to obtain  $K = L_0 + (3/2)L_1$  and  $J = 2L_1$ .

A plot of the energy shift  $K$  as a function of  $x_b$  (the tunneling energy) and  $x_c$  (the Coulomb energy) is shown in Fig. 3 in units of  $\hbar\omega_0$ . Following Ref. 8, we estimate realistic values for  $x_b$  and  $x_c$  by considering the case of GaAs heterostructure single dots. An estimated value for  $x_b$  is  $x_b \equiv m\omega_0^2 l^2/(\hbar\omega_0) \approx 1$ , since the harmonic oscillator width  $2\sqrt{\hbar/(m\omega_0)}$  should be approximately equal to the distance between dots  $2l$  in a quantum computer. The parameter  $x_c \equiv e^2/(\kappa \hbar\omega_0) \approx 1.5$  taking  $\kappa = 13.1$ ,  $\hbar\omega_0 = 3$  meV and  $x_b$

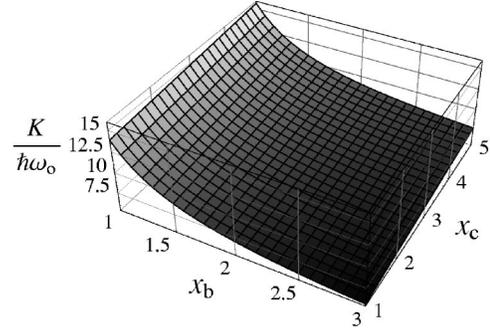


FIG. 3. Plot of  $K$  as a function of dimensionless tunneling barrier  $x_b$  and Coulomb energy  $x_c$  in the case of two interacting electrons.

$\approx 1$ . Note that the energy  $K$  increases when the one-electron tunneling barrier energy decreases and the Coulomb-interaction energy increases (i.e., for small  $x_b$  and large  $x_c$ ).

In Fig. 4, we plot the exchange-interaction constant  $J$  as a function of  $x_b$  and  $x_c$ . The plot generally indicates that  $J$  increases as the tunneling barrier decreases ( $x_b$  smaller)—an intuitively reasonable result. Although it is outside the regime of physical interest depicted in the plot, when the Coulomb interaction is extraordinarily strong,  $x_c \rightarrow \sim 15$ ,  $J$  develops a negative minimum at  $x_b \sim 1.5$ . The reason is that the  $S_T = 3/2$  state has a totally antisymmetric spatial wave function, while the  $S_T = 1/2$  state does not. The antisymmetry tends to reduce the Coulomb-repulsion energy between electrons while increasing the one-electron tunneling energy. When parameters are tuned to make the Coulomb repulsion important, the energy of the  $S_T = 3/2$  state dips down, eventually decreasing below the energy of the  $S_T = 1/2$  state. This leads to  $J < 0$ . The negative value of  $J$  signals the breakdown of the HL approximation in this region. The exact two-electron ground state is known<sup>39</sup> to have  $S_T = 1/2$ , while  $J < 0$  would imply an  $S_T = 3/2$  ground state. The HL representation of the  $S_T = 1/2$  state is simply too rigid to represent the exact ground state when interactions are extremely strong. The inflexibility of the HL wave functions should be kept in mind when there are extremely strong interactions in the three-electron and four-electron cases, as well. Fortunately, in our region of physical interest,  $x_b \approx 1$ ,  $x_c \approx 1.5$ , HL results should be meaningful. Even then, however, it should be kept in mind that the barrier between minima of the potential (26) is

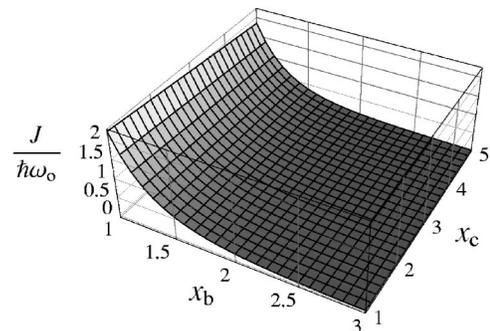


FIG. 4. Plot of  $J$  as a function of  $x_b$  and  $x_c$  in the case of two interacting electrons.

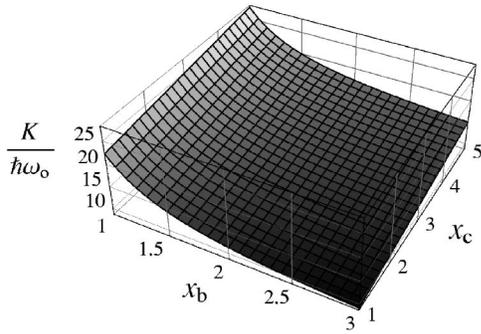


FIG. 5. Plot of  $K$  as a function of  $x_b$  and  $x_c$  in the case of three mutually interacting electrons.

shallow, and so the calculation will become increasingly inaccurate as the minima get close together. Alternative numerical methods can be found, e.g., in Refs. 14–17.

### B. Three electrons

In the case of three electrons we assume three of the potential minima in Eq. (26) are occupied at, say,  $\mathbf{A} = (0, 0, 0)$ ,  $\mathbf{B} = (2l\sqrt{1/3}, 0, -2l\sqrt{2/3})$ , and  $\mathbf{C} = (-l\sqrt{1/3}, l, -2l\sqrt{2/3})$ . The electrons are therefore arranged at the corners of an equilateral triangle, and the minimum at  $\mathbf{D}$  is unoccupied. To solve Eqs. (14) and (15), we need to evaluate three-body matrix elements. Details are given in Appendix B.

A plot of the energy shift  $K$  as a function of  $x_b$  (the tunneling energy) and  $x_c$  (the Coulomb energy) is shown in Fig. 5 in units of  $\hbar\omega_0$ . The plot's shape is quite similar to that of Fig. 3. Figure 6 displays the change  $\Delta K$  given by subtracting from  $K$  the value that  $K$  would take if the three-electron swap matrix elements  $\epsilon_0 = \langle CAB|H|ABC \rangle$  and  $p_0 = \langle CAB|ABC \rangle$  were zero. The axis directions are reversed in this plot to make its shape easier to inspect. The figure shows that  $\Delta K$  is most important when the one-electron tunneling barrier energy and the Coulomb-interaction energy are small in magnitude (small  $x_b$  and small  $x_c$ ).

In Fig. 7, we plot the exchange-interaction constant  $J$  as a function of  $x_b$  and  $x_c$ . A similar figure appeared previously<sup>22</sup> with an erroneous scale on the  $x_c$  axis. (On the four plots

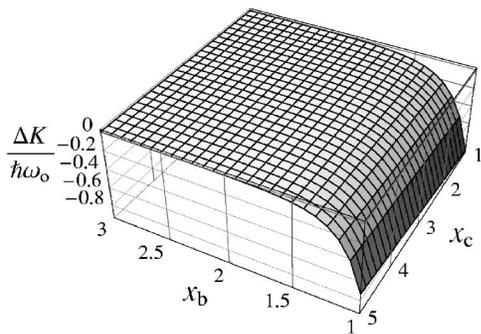


FIG. 6. Plot of  $\Delta K$  as a function of  $x_b$  and  $x_c$  in the case of three mutually interacting electrons. Axis directions are reversed from the preceding figure.

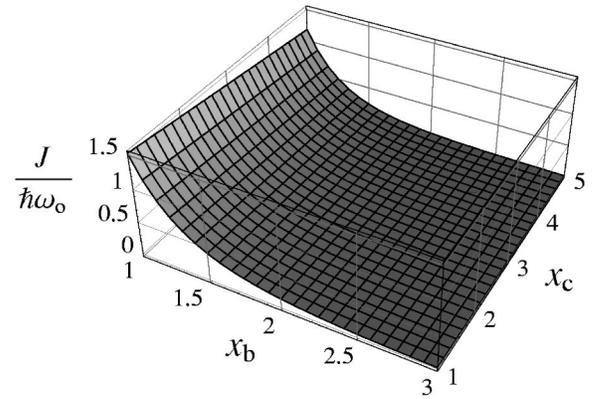


FIG. 7. Plot of  $J$  as a function of  $x_b$  and  $x_c$  in the case of three mutually interacting electrons.

appearing in Ref. 22, the ticks on the  $x_c$  axis ran from 1 to 5, but the plots actually depicted the range  $2\sqrt{2} < x_c < 10\sqrt{2}$ ; the correct figures appearing here do not change the conclusions of that work.) Figure 7 shows a physically reasonable parameter range in which the qualitative appearance of  $J$  is similar to that of the two-electron case, Fig. 4.

Figure 8 shows the change  $\Delta J$  given by subtracting from  $J$  the value that  $J$  would take if the three-electron swap matrix elements  $\epsilon_0$  and  $p_0$  were zero (note that the axis directions are flipped to make the plot clearer). Comparing the scales of Figs. 7 and 8, one finds that the three-electron swap matrix elements can have a powerful influence on  $J$ .

To complement our HL results, we have computed the HM spectrum. For reasonable parameter values ( $x_b = 1.0, x_c = 1.5$ ), we have found that the lowest four states of the 16-dimensional  $S=1/2$  subspace are degenerate and have an energy (that we call  $E_{1/2, \text{HM}}$ ) that is well separated from that of the remaining 12 states with  $S=1/2$ . These four states are similar in composition to the four members of the HL  $S=1/2$  subspace. The remaining 12 states of the HM  $S=1/2$  subspace consist mainly of states with two electrons on a single dot. The four  $S=3/2$  states have an energy that is *in between*  $E_{1/2, \text{HM}}$  and the energy of the higher-lying  $S=1/2$  states. We thus have a situation that is analogous to the one we encountered in the HL case. It is reasonable to project out

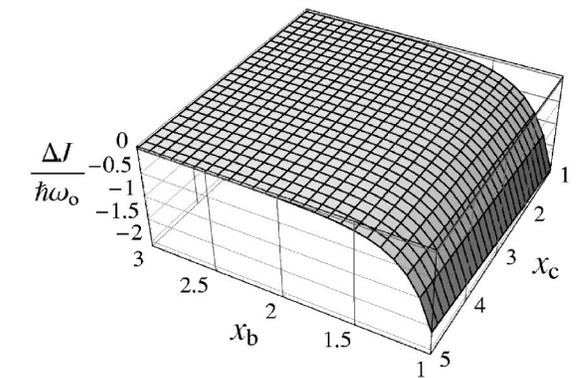


FIG. 8. Plot of  $\Delta J$  as a function of  $x_b$  and  $x_c$  in the case of three mutually interacting electrons. Axis directions are reversed from the preceding figure.

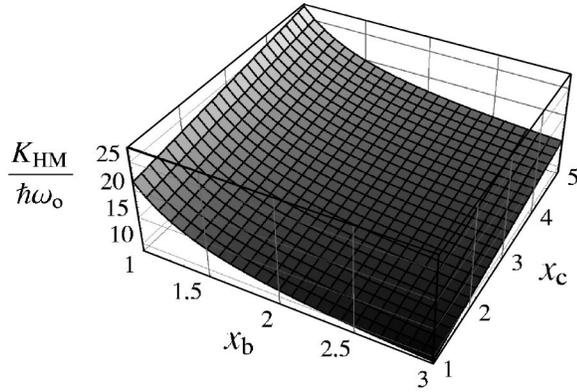


FIG. 9. Plot of  $K$  as a function of  $x_b$  and  $x_c$  in the case of three mutually interacting electrons, computed within the HM approximation.

the eight low-energy states of the HM calculation and compare with the HL calculation. Figures 9 and 10 show the values of  $K$  and  $J$  for an effective Hamiltonian of the form (16) that gives this eight-dimensional low-energy subspace's spectrum. These figures should be compared to Figs. 5 and 7.

For reasonable parameter values (again,  $x_b=1.0, x_c=1.5$ ), we find in the HL approximation that  $J=2.2$  for two particles (in units of  $\hbar\omega_0$ ),  $J=1.5$  for three particles, indicating a change of  $-32\%$  (or an absolute change of  $-0.7$ ). In the HM approximation,  $J=3.2$  for two particles and  $J=2.4$  for three particles, indicating a change of  $-25\%$  (or an absolute change of  $-0.8$ ). Thus, the same effect is seen. The absolute value of  $J$  is larger in the HM case (this is expected since the basis has increased, leading to a decrease in the ground state energy  $E_{1/2}$  while  $E_{3/2}$  stays constant), but the qualitative HL conclusions are well substantiated.

**C. Four electrons**

The actual calculation for the four-electron case is more involved than that of the three-electron case but identical in procedure. Details are given in Appendix C. The resulting quantities  $K$  and  $\Delta K$  appear as functions of  $x_b$  and  $x_c$  in Figs. 11 and 12, respectively. Here,  $\Delta K$  is the value of  $K$  minus the value of  $K$  obtained by setting to zero both three-body ( $p_1, \epsilon_1$ ) and also four-body ( $p_0, p'_0, \epsilon_0, \epsilon'_0$ ) matrix elements.

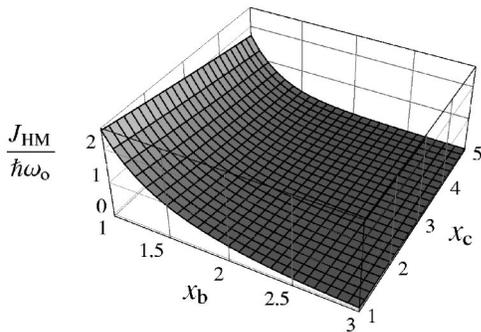


FIG. 10. Plot of  $J$  as a function of  $x_b$  and  $x_c$  in the case of three mutually interacting electrons, computed within the HM approximation.

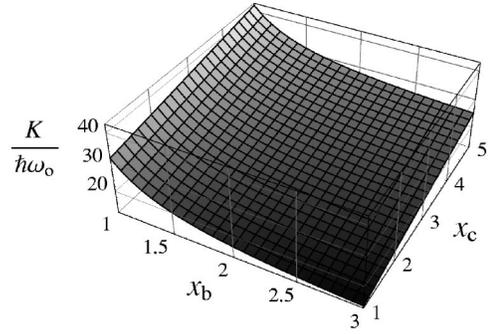


FIG. 11. Plot of  $K$  as a function of  $x_b$  and  $x_c$  in the case of four mutually interacting electrons.

The behavior of the exchange-interaction constant  $J$  as a function of  $x_b$  and  $x_c$  (Fig. 13) is similar to that of the three-electron case (Fig. 7). The appearance of  $\Delta J$  (Fig. 14, given by subtracting from  $J$  the value that  $J$  would take if the three-body and four-body matrix elements were zero) is also reminiscent of  $\Delta J$  in the three-electron case (Fig. 8). On the other hand,  $J'$  (Fig. 15) exhibits different behavior while  $\Delta J'$  (Fig. 16) is qualitatively similar in form to  $\Delta J$  from the three-electron case.

The interaction constant  $J'$  can be quite significant compared to  $J$ , which is remarkable and requires attention in quantum computer design. In fact, at the point  $x_b=1, x_c=1.5$ , our calculation yields  $J=0.93$  and  $J'=-0.15$  so  $J'/J=-16\%$ , implying substantial four-body interactions. We caution, though, that these values were obtained within a HL approximation that will become inaccurate as  $x_b$  decreases and the minima of (26) get closer together. Our intention is to highlight the possible significance of the four-body terms. Such terms have been observed experimentally in  $^3\text{He}$  (Ref. 40), and  $\text{Cu}_4\text{O}_4$  square plaquettes in  $\text{La}_2\text{CuO}_4$  (Ref. 41), where  $J'/J$  was found to be  $\sim 27\%$ .

**V. CONCLUSIONS**

The exchange interaction between localized electrons is a basic phenomenon of condensed-matter physics, with a history that dates back to Heisenberg's pioneering work.<sup>1</sup> The details of its behavior are of great significance to quantum

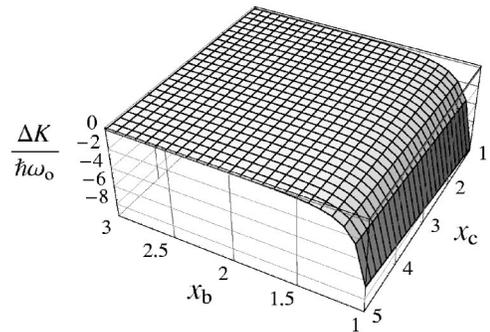


FIG. 12. Plot of  $\Delta K$  as a function of  $x_b$  and  $x_c$  in the case of four mutually interacting electrons. Axis directions are reversed from the preceding figure.

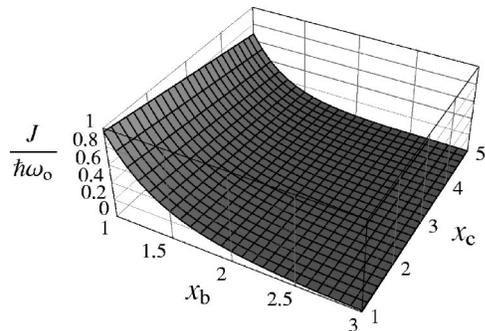


FIG. 13. Plot of  $J$  as a function of  $x_b$  and  $x_c$  in the case of four mutually interacting electrons.

information processing using quantum dots. Here we have considered the effects that arise when three or more electrons, each localized in a low-energy orbital on a quantum dot, are simultaneously coupled. We have shown that both quantitative and qualitative effects arise, due to many-body terms, that modify the standard form of the Heisenberg exchange interaction. Most significantly, in the case of four coupled electrons, there is a four-body interaction that is added to the Heisenberg exchange interaction, and our HL calculations suggest that it could be strong in physically relevant parameter regimes. This possibility needs to be considered in electron-spin-based quantum computer design because, on the one hand, of the problems it could produce when its presence is unwelcome and, on the other hand, because of its potential uses in novel designs. In other designs as well, the possibility should be considered that many-qubit terms could arise in the effective qubit Hamiltonian.

*Note added in proof:* A recent paper<sup>42</sup> has quantitatively verified an effect alluded to above in Eq. (8). This paper demonstrates that the application of a magnetic field breaks the inversion symmetry of the system, allowing chiral terms of the form  $\mathbf{S}_A \cdot (\mathbf{S}_B \times \mathbf{S}_C)$  to arise in the Hamiltonian.

#### ACKNOWLEDGMENTS

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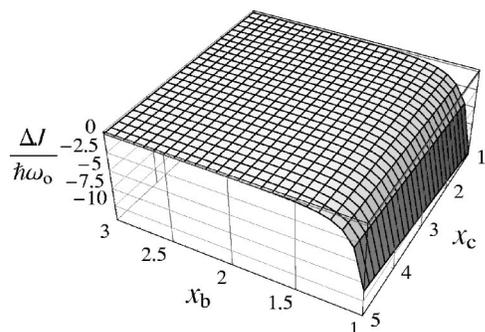


FIG. 14. Plot of  $\Delta J$  as a function of  $x_b$  and  $x_c$  in the case of four mutually interacting electrons. Axis directions are reversed from the preceding figure.

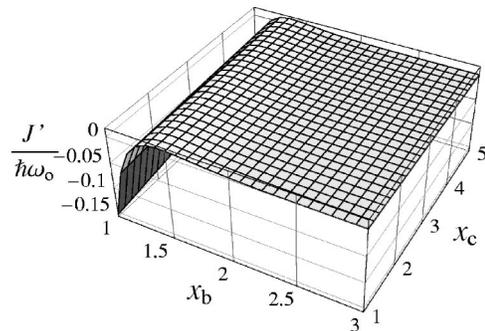


FIG. 15. Plot of  $J'$  as a function of  $x_b$  and  $x_c$  in the case of four mutually interacting electrons.

by AFOSR under Contract No. F49620-01-1-0468), PREA and the Connaught Fund. We thank Professor T.A. Kaplan for useful correspondence.

#### APPENDIX A: PERTURBATIVE APPROACH TO HEISENBERG EXCHANGE

Here we summarize the perturbative approach to deriving corrections to the Heisenberg exchange interaction. See, e.g., Refs. 7, 26, and 27 for more details.

After second quantization of the Coulomb interaction Hamiltonian (3), one arrives at the result

$$H = \sum_{i,s} \varepsilon_i n_{i,s} + \sum_{i<j} \sum_s t_{ij} a_{i,s}^\dagger a_{j,s} + U \sum_i n_{i,s} n_{i,-s} - \sum_{i<j} \sum_{s,s'} J_{ij} a_{i,s}^\dagger a_{i,s'}^\dagger a_{j,s'} a_{j,s},$$

where  $a_{i,s}^\dagger$  creates an electron with spin  $s$  in the  $i$ th Wannier orbital  $\phi(\mathbf{r}-\mathbf{r}_i)$ ,  $n_{i,s} = a_{i,s}^\dagger a_{i,s}$  is the number operator,

$$t_{ij} = \int \phi^*(\mathbf{r}-\mathbf{r}_i) h(\mathbf{r}) \phi(\mathbf{r}-\mathbf{r}_j) d\mathbf{r}$$

is the hopping energy for  $i \neq j$ ,  $\varepsilon_i \equiv t_{ii}$  is the energy of the electron in the  $i$ th orbital,

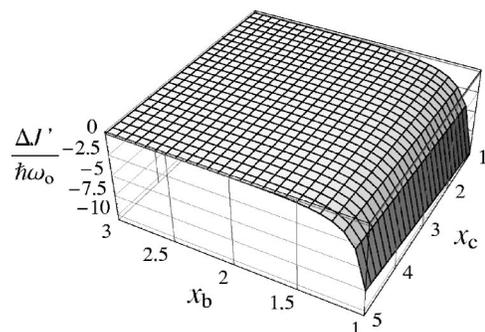


FIG. 16. Plot of  $\Delta J'$  as a function of  $x_b$  and  $x_c$  in the case of four mutually interacting electrons. Axis directions are reversed from the preceding figure.

$$U = \frac{1}{2} e^2 \int \frac{|\phi(\mathbf{r})|^2 |\phi(\mathbf{r}')|^2}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r} d\mathbf{r}'$$

is the on-site interaction energy, and

$$J_{ij} = e^2 \int \frac{\phi^*(\mathbf{r} - \mathbf{r}_i) \phi(\mathbf{r} - \mathbf{r}_j) \phi^*(\mathbf{r}' - \mathbf{r}_j) \phi(\mathbf{r}' - \mathbf{r}_i)}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r} d\mathbf{r}' \geq 0$$

is the customary direct exchange integral (ferromagnetic).

One now assumes  $U \gg \varepsilon_i, t_{ij}, J_{ij}$  to ensure that all orbitals are singly occupied. One then evaluates  $H$  in the HL basis

$$|s_1, s_2, \dots, s_n\rangle = a_{1s_1}^\dagger a_{2s_2}^\dagger \cdots a_{ns_n}^\dagger |\text{vac}\rangle$$

in which states differ one from the other only in the distribution of spins  $s_i$  in the orbitals  $i$ . The evaluation is facilitated by noting that the operators

$$S_i^\alpha \equiv \frac{1}{2} \sum_{s, s'} a_{is}^\dagger \sigma_{s, s'}^\alpha a_{is'}$$

are the component of spin-half operators, where  $\sigma_{s, s'}^\alpha$  are the matrix elements of the Pauli matrices ( $\alpha = x, y, z$ ). This allows one to rewrite the exchange term as

$$\sum_{s, s'} J_{ij} a_{is}^\dagger a_{is'}^\dagger a_{js'} a_{js} = 2J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \text{const},$$

which is the familiar Heisenberg exchange Hamiltonian.

The contribution of the hopping term to  $H$  can be neglected in the limit  $t_{ij}/U \rightarrow 0$ . However, when  $t_{ij}/U \ll 1$  but nonvanishing, it can be shown,<sup>7</sup> using standard perturbation theory in powers of  $1/U$ , that the effective hopping Hamiltonian in the HL basis takes the form

$$H_{\text{eff}} = -\frac{H_h^2}{U} + \frac{H_h^3}{U^2} + \frac{H_h^4}{U^3} + \dots,$$

where  $H_h = \sum_{i < j} \sum_s t_{ij} a_{is}^\dagger a_{js}$  is the original hopping Hamiltonian, which vanishes in the HL basis. The first-order correction  $-H_h^2/U$  gives rise to a term of the form  $(1/U) \sum_{i < j} |t_{ij}|^2 \mathbf{S}_i \cdot \mathbf{S}_j + \text{const}$ , which quantitatively modifies (with opposite sign, i.e., antiferromagnetically) the Heisenberg Hamiltonian.<sup>6</sup> However, it is clear that higher-order terms can contribute multispin terms of the form we have considered in this paper. It can be shown<sup>26</sup> that all odd orders vanish, in agreement with our general symmetry argument of Sec. II. The term  $H_h^4/U^3$  then gives rise to four-spin interactions of the form  $(\mathbf{S}_i \cdot \mathbf{S}_j)(\mathbf{S}_k \cdot \mathbf{S}_l)$ , proportional to  $t_{ij} t_{jk} t_{kl} t_{li}/U^3$ , with  $i < j < l$ ,  $i < k$ ,  $k \neq j, l$ .<sup>26</sup> This can be interpreted diagrammatically as a cycle in which the electrons interchange dots in the order  $i \rightarrow l \rightarrow k \rightarrow j \rightarrow i$ . Thus, perturbation theory shows that when  $t^4/U^3$  is significant, the four-spin interaction cannot be neglected.

### APPENDIX B: DETAILS OF CALCULATIONS FOR THREE ELECTRONS

The Hamiltonian (3) contains three one-body terms  $h$  and three Coulomb-interaction terms  $w$ , and the contribution of each term is given in Eqs. (27)–(29). These contributions

determine the parameters  $p_3, p_1, p_0, \varepsilon_3, \varepsilon_1$ , and  $\varepsilon_0$  that appear in Eqs. (14) and (15)

$$\varepsilon_3 = \langle A|h|A\rangle + \langle B|h|B\rangle + \langle C|h|C\rangle + \langle AB|w|AB\rangle + \langle AC|w|AC\rangle + \langle BC|w|BC\rangle,$$

$$\varepsilon_1 = \langle B|h|A\rangle \langle A|B\rangle + \langle A|h|B\rangle \langle B|A\rangle + \langle C|h|C\rangle + \langle BA|w|AB\rangle + \langle BC|w|AC\rangle \langle A|B\rangle + \langle AC|w|BC\rangle \langle B|A\rangle,$$

$$\varepsilon_0 = \langle C|h|A\rangle \langle A|B\rangle \langle B|C\rangle + \langle A|h|B\rangle \langle C|A\rangle \langle B|C\rangle + \langle B|h|C\rangle \langle C|A\rangle \times \langle A|B\rangle + \langle CA|w|AB\rangle \langle B|C\rangle + \langle CB|w|AC\rangle \langle A|B\rangle + \langle AB|w|BC\rangle \langle C|A\rangle,$$

and

$$p_3 = 1,$$

$$p_1 = \langle A|B\rangle \langle B|A\rangle,$$

$$p_0 = \langle C|A\rangle \langle A|B\rangle \langle B|C\rangle.$$

We apply the symmetries of an equilateral triangle,  $\langle B|A\rangle = \langle C|A\rangle = \langle B|C\rangle$ ,  $\langle B|h|A\rangle = \langle C|h|A\rangle = \langle B|h|C\rangle$ ,  $\langle A|h|A\rangle = \langle B|h|B\rangle = \langle C|h|C\rangle$  to get the matrix elements not explicitly listed in Eqs. (27)–(29). All matrix elements are functions of  $\hbar\omega_o, x_b$ , and  $x_c$ , so  $L_0$  and  $L_1$  obtained from Eqs. (14) and (15), and  $K$  and  $J$  obtained from Eqs. (17) and (18), are functions of  $\hbar\omega_o, x_b$ , and  $x_c$  as well.

### APPENDIX C: DETAILS OF CALCULATIONS FOR FOUR ELECTRONS

Here, there is an electron in a Gaussian orbital at each of the four potential minima of (26). We use the analytical expressions (27)–(29) to evaluate the many-body matrix elements that appear in Eqs. (20)–(22). The Hamiltonian (3) contains four one-body terms  $h$  and six Coulomb-interaction terms  $w$ . Taking them all into account, we have

$$\varepsilon_4 = \langle A|h|A\rangle + \langle B|h|B\rangle + \langle C|h|C\rangle + \langle D|h|D\rangle + \langle AB|w|AB\rangle + \langle AC|w|AC\rangle + \langle AD|w|AD\rangle + \langle BC|w|BC\rangle + \langle BD|w|BD\rangle + \langle CD|w|CD\rangle,$$

$$\varepsilon_2 = \langle B|h|A\rangle \langle A|B\rangle + \langle A|h|B\rangle \langle B|A\rangle + \langle C|h|C\rangle \langle B|A\rangle \langle A|B\rangle + \langle D|h|D\rangle \langle B|A\rangle \langle A|B\rangle + \langle BA|w|AB\rangle + \langle BC|w|AC\rangle \langle A|B\rangle + \langle BD|w|AD\rangle \langle A|B\rangle + \langle AC|w|BC\rangle \langle B|A\rangle + \langle AD|w|BD\rangle \times \langle B|A\rangle + \langle CD|w|CD\rangle \langle B|A\rangle \langle A|B\rangle,$$

$$\varepsilon_1 = \langle A|h|A\rangle \langle D|B\rangle \langle B|C\rangle \langle C|D\rangle + \langle D|h|B\rangle \langle B|C\rangle \langle C|D\rangle + \langle B|h|C\rangle \times \langle D|B\rangle \langle C|D\rangle + \langle C|h|D\rangle \langle D|B\rangle \langle B|C\rangle + \langle AD|w|AB\rangle \langle B|C\rangle \times \langle C|D\rangle + \langle AB|w|AC\rangle \langle D|B\rangle \langle C|D\rangle + \langle AC|w|AD\rangle \langle D|B\rangle \times \langle B|C\rangle + \langle DB|w|BC\rangle \langle C|D\rangle + \langle DC|w|BD\rangle \langle B|C\rangle + \langle BC|w|CD\rangle \langle D|B\rangle,$$

$$\begin{aligned}\epsilon_0 = & \langle B|h|A\rangle\langle A|B\rangle\langle D|C\rangle\langle C|D\rangle + \langle A|h|B\rangle\langle B|A\rangle\langle D|C\rangle\langle C|D\rangle \\ & + \langle D|h|C\rangle\langle B|A\rangle\langle A|B\rangle\langle C|D\rangle + \langle C|h|D\rangle\langle B|A\rangle\langle A|B\rangle\langle D|C\rangle \\ & + \langle BA|w|AB\rangle\langle D|C\rangle\langle C|D\rangle + \langle BD|w|AC\rangle\langle A|B\rangle\langle C|D\rangle \\ & + \langle BC|w|AD\rangle\langle A|B\rangle\langle D|C\rangle + \langle AD|w|BC\rangle\langle B|A\rangle\langle C|D\rangle \\ & + \langle AC|w|BD\rangle\langle B|A\rangle\langle D|C\rangle + \langle DC|w|CD\rangle\langle B|A\rangle\langle A|B\rangle,\end{aligned}$$

$$\begin{aligned}\epsilon'_0 = & \langle D|h|A\rangle\langle A|B\rangle\langle B|C\rangle\langle C|D\rangle + \langle A|h|B\rangle\langle D|A\rangle\langle B|C\rangle\langle C|D\rangle \\ & + \langle B|h|C\rangle\langle D|A\rangle\langle A|B\rangle\langle C|D\rangle + \langle C|h|D\rangle\langle D|A\rangle\langle A|B\rangle\langle B|C\rangle \\ & + \langle DA|w|AB\rangle\langle B|C\rangle\langle C|D\rangle + \langle DB|w|AC\rangle\langle A|B\rangle\langle C|D\rangle \\ & + \langle DC|w|AD\rangle\langle A|B\rangle\langle B|C\rangle + \langle AB|w|BC\rangle\langle D|A\rangle\langle C|D\rangle \\ & + \langle AC|w|BD\rangle\langle D|A\rangle\langle B|C\rangle + \langle BC|w|CD\rangle\langle D|A\rangle\langle A|B\rangle.\end{aligned}$$

The overlap matrix elements are simpler

$$p_4 = 1,$$

$$p_2 = \langle B|A\rangle\langle A|B\rangle,$$

$$p_1 = \langle D|B\rangle\langle B|C\rangle\langle C|D\rangle,$$

$$p_0 = \langle B|A\rangle\langle A|B\rangle\langle D|C\rangle\langle C|D\rangle,$$

$$p'_0 = \langle D|A\rangle\langle A|B\rangle\langle B|C\rangle\langle C|D\rangle.$$

Analytical forms are then available for all of the matrix elements of  $H$  and all the overlap matrix elements using expressions (27)–(29) and using the tetrahedron symmetries

$$\langle B|A\rangle = \langle C|A\rangle = \langle D|A\rangle = \langle B|C\rangle = \langle B|D\rangle = \langle C|D\rangle,$$

$$\langle B|h|A\rangle = \langle C|h|A\rangle = \langle D|h|A\rangle = \langle B|h|C\rangle = \langle B|h|D\rangle = \langle C|h|D\rangle,$$

$$\langle A|h|A\rangle = \langle B|h|B\rangle = \langle C|h|C\rangle = \langle D|h|D\rangle.$$

With all of the matrix elements of  $H$  and the overlap matrix elements in hand, we evaluate  $K$ ,  $J$ , and  $J'$  by solving Eqs. (20)–(25).

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