Spin-based quantum computation in multielectron quantum dots

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In a quantum computer the hardware and software are intrinsically connected because the quantum Hamiltonian (or more precisely its time development) is the code that runs the computer. We demonstrate this subtle and crucial relationship by considering the example of electron-spin-based solid-state quantum computer in semiconductor quantum dots. We show that multielectron quantum dots with one valence electron in the outermost shell do not behave simply as an effective single-spin system unless special conditions are satisfied. Our work compellingly demonstrates that a delicate synergy between theory and experiment (between software and hardware) is essential for constructing a quantum computer.

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Ever since the pioneering work on quantum computation and quantum error correction [1–4], there have been many-proposed quantum computer (QC) hardware architectures based on different quantum systems [5], such as trapped ions [6], cavity QED [7], liquid-state nuclear magnetic resonance (NMR) [8], nuclear spins in solids [9], electron spins [10–12], superconducting Josephson junctions [13], and electrons on He surface [14], etc. Currently, experimental progress has mostly occurred in proposals based on atomic, optical, and NMR physics. Many solid-state proposals have remained in the model stage because of the immense experimental difficulties. To help overcome these difficulties, more theoretical work is needed to explore the optimal operating regimes, figure out the operational constraints and tolerances, and discover potential sources of errors, just to name a few directions [15–18]. While the optical and atomic physics based architectures have been crucial in demonstrating the proof of principle for quantum computation, it is generally believed that solid-state QC architectures, with their obvious advantage of controllable scale-up possibilities, offer the most promising potential for realistic large-scale QC hardwares. The fundamental problem plaguing the solid-state QC architectures has been the fact that the basic quantum bit (qubit), the QC building block, has not been compellingly demonstrated in any solid-state QC architectures, although there is no reason to doubt that they exist in nature. Thus, the construction of successful QC hardwares has faced the somewhat embarrassing dichotomy: the architectures (ion traps, etc.) demonstrating the existence of quantum bits cannot be easily scaled up, while the architectures (solid-state QCs) that may be easily scaled up have not yet experimentally demonstrated quantum bits.

Quantum computation with fermionic spins is considered to be a potentially promising prospect for solid-state quantum computers [9–12,19]. Among the many proposed solid-state QC architectures the spin quantum computer has several intrinsic advantages: (1) A fermionic spin, being a quantum two-level system, is a natural qubit with its spin-up and spin-down states; (2) it is fairly straightforward to carry out single-qubit operations on spin-up and spin-down levels by applying suitable magnetic fields (or through a purely exchange-based scheme [15]); (3) two-qubit operations can, in principle, be carried out rather easily (in theory, at least) by using the exchange interaction between two neighboring spins; and (4) quantum spin is fairly robust and does not decohere easily (typical electron spin relaxation times in solids are many orders of magnitude longer [20] than the momentum relaxation time)—in particular, electron spin relaxation times could be microseconds in semiconductors [21].

Our work presented in this paper deals with a crucial aspect of solid-state spin qubits that has so far been neglected in the literature. The intrinsic advantages of spin-based solid-state quantum computation have led to several concrete proposals for using electron spins [in semiconductor quantum dots (QD) or in donor impurity atoms] as qubits in semiconductor based solid-state QC architectures [9–12]. One exciting proposal [10] deals with one electron spin per quantum dot working as a qubit, with two coupled spins on two neighboring dots (forming a QD molecule [22–24]) providing two-qubit operations through the interdot electronic exchange coupling. The electron spin on shallow donor states in semiconductors, while differing in some details with the QD spin-qubit architecture, still exploits the idea of only one effective spin-1/2 fermion (i.e., one electron) per donor state participating in the quantum computation [12]. At first sight this idea of a single electron in a dot may seem far fetched because an array of semiconductor QDs, even under the most advanced growth and nanofabrication constraints, is likely to have more than a single electron on each dot [25]. However, the idea of one effective electron spin per quantum dot working as a qubit is not as crazy as it may seem at first sight. In particular, QD electronic states are, similar to real atomic electronic states, naturally divided into quantized shells (i.e., *S*, *P*, *D*, *F*, etc.) corresponding to the quantization of the orbital motion [25]. Furthermore, the orbital excitation energy in a small QD is much higher than the spin-flip energy for realistic fields. The single electron spin per quantum dot idea is therefore based on the closed-shell principle, where what is required is just one ''valence'' electron per quantum dot in the outermost ''open'' shell. The underlying idea here is that the closed-shell electrons (equivalent to the core electrons in atoms) are ''inert'' and could be ignored as far as qubit dynamics goes, and the unoccupied states are energetically too unfavorable to be involved as well. This principle in a different context has, in fact, worked for trapped-ion quantum computation [2] where

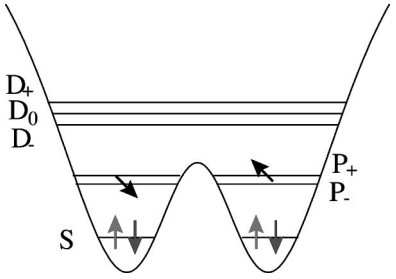

FIG. 1. Here we show a schematic of six electrons in a double dot. Four of the electrons will occupy the four lowest spin orbitals (two *S* orbitals with two spin orientations), and thus fill up the *S*-shell states. The other two electrons can be in any one of the remaining ten *P* and *D* spin orbitals in our calculations.

''valence''-type ionic orbital states are manipulated as qubits, and the filled inner-shell states are inert and are ignored.

The all-important question for quantum dot electron spin quantum computation is therefore the extent to which this same scenario applies, i.e., both the *inert* filled core of a quantum dot and the outer-shell unoccupied orbital states can be ignored for quantum computation because they do not affect the qubit dynamics either for single qubit or for exchange-mediated two-qubit operations. The answer to this question is nontrivial and non-obvious because the confinement potential in quantum dots is very different from and much softer than that for real atoms. In addition, the gated circular QDs are essentially two dimensional and the Fock-Darwin states (two-dimensional electron eigenstates in a magnetic field and a harmonic confinement) are isotropic, unlike the three-dimensional anisotropic atomic states. We address this crucial issue of electron-spin-based QD quantum computation by accurately calculating the energy levels and exchange couplings in multielectron QD molecules where two semiconductor (GaAs) quantum dots, each with three electrons, are used as the fundamental building block of the quantum computer architecture (Fig. 1). We perform a configuration-interaction (CI) calculation with a Hartree-Fock basis. Specifically, we expand the single-electron states in a basis including all 12 *S*, *P*, and *D* Fock-Darwin states located at the two potential minima. This leads to $12\times 2 = 24$ Hartree-Fock spin orbitals (each spatial orbital has two-spin orientation). We include both singly and doubly excited six-electron states in the CI basis, and solve the Schrödinger equation by expanding on the six-electron Slater basis (Zeeman coupling has been neglected in this calculation):

$$H(1,\ldots,6)\sum_i c_i\Psi_i(1,\ldots,6)=E\sum_i c_i\Psi_i(1,\ldots,6)\,, \tag{1}$$

where H is the six-electron Hamiltonian including kinetic and potential energy and electron Coulomb interaction. As our theory is based on a sophisticated quantum-chemistry approach [26], our results should have general qualitative and semiquantitative validity. There have been several recent theoretical calculations of the ground-state spin polarization properties of multielectron quantum dot systems using the density-functional theory [27–29]. For the purpose of quantum computation of interest to us in this paper, however, the knowledge of the excited states is crucial in determining whether a particular number of electrons can serve as an effective qubit—in particular, we need an accurate evaluation of the singlet-triplet energy splitting in the system. Such excited-state information is beyond the scope of density-functional theories that are restricted to ground states only. The quantum-chemical CI calculations we present in this paper are particularly well suited in dealing with the low-lying excited states and in providing information about the exchange splitting in the system (in contrast to ground-state density-functional theories).

Our findings, shown in Fig. 2(a), are rather striking: we find that the six-electron Hilbert space (i.e., energy-level spectra of the two-dot system) is qualitatively different from the two-electron double quantum dot case [17] shown in Fig. 2(b) (included here for comparison), and the multielectron system (with one electron in the outermost open ''valence'' shell) does not necessarily behave as a simple one-effective spin per dot model. We find that quantum computation using quantum-dot spin qubits and exchange gates will most probably require the application of an external magnetic field or other means to ensure a well-defined sub-Hilbert space, which is an essential QC requirement [19], in the exchange-based two-qubit operations.

In the representative energy spectra presented in Fig. 2(a) (with parameters in the figure caption), the *S* electrons are tightly confined to the individual QDs. We include (for comparison) results of our restricted Hartree-Fock ground-state energies as dotted thick black lines. Notice that the CI calculation produces a 3–4 meV improvement in the ground-state energy, mostly by introducing electron correlations to minimize Coulomb repulsion. The lowest lines at each field in Fig. 2(a) actually consist of two lines corresponding to the lowest singlet and triplet states. As their energy splitting is in the range 0.01–0.05 meV, the difference is too small to show up in this figure. Since this energy difference (singlet-triplet or exchange splitting) is crucial in two-qubit operations, we plot the magnetic-field dependence of the singlet-triplet splitting in the insets of Fig. 2. Notice that the high magnetic-field part of the inset of Fig. 2(a) is quite similar to that for the two-electron double-dot case shown in the inset of Fig. 2(b). Here both triplet and singlet states consist mainly of the lower-energy *P* orbitals ψ_{LP-} and ψ_{RP-}; the first subscript refers to the left or right QD, the second refers to the orbital quantum number (*S*, *P*, or *D*) of the Fock-Darwin state sequence, and the third is the orbital magnetic quantum number. Strong magnetic fields tightly squeeze the radii of these *P* states so that their overlap originates entirely from their exponentially vanishing tails, leading to the similar high field behavior in Figs. 2(a) and 2(b). At low fields the exchange splitting in the multielectron system has a much more complicated behavior than its single-electron counterpart. At zero field the splitting is close to zero, implying a delicate balance between electron kinetic energy and Coulomb interaction. The splitting quickly increases for lower central barriers as the outer-shell Hartree-Fock states change quickly from an even superposition of ψ_{P-} and ψ_{P+}

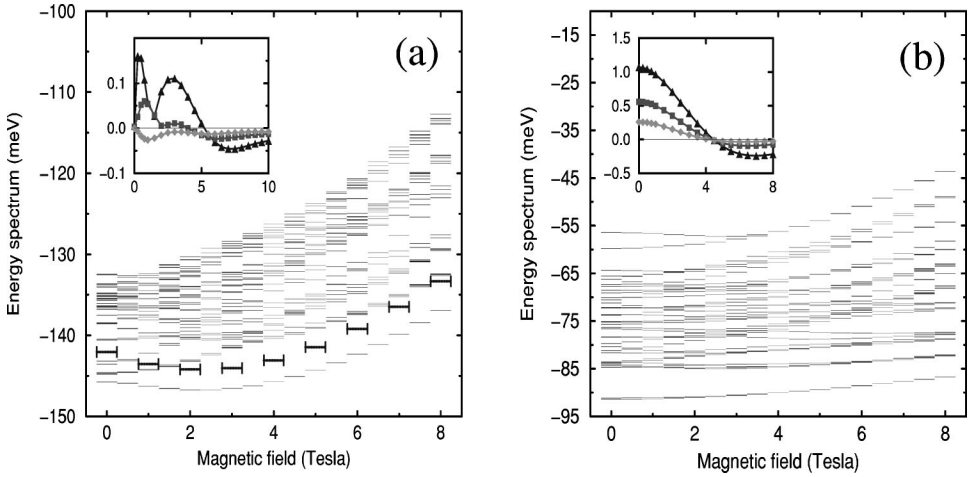

FIG. 2. In (a) we plot the energy spectra (lowest 40 states) of a particular six-electron horizontal double dot as a function of an applied magnetic field along the *z* direction. The quantum dot widths (Gaussian confinement widths) are 30 nm in radius. The distance between the two confinement potential minima is 40 nm. The central barrier height is 30 meV (with an effective height of 19.28 meV). For a more detailed description of the Gaussian confinement and barrier we use here, and a description of the horizontal quantum dots we study, see [17]. The thick dotted black lines with risers are the ground-state energies of our restricted Hartree-Fock self-consistent states, plotted here as a starting point to compare our CI results with. The inset shows how the splitting of the lowest singlet and triplet states varies with the external magnetic field (at three different effective barrier heights of 15.10, 19.28, and 23.65 meV). For the purpose of comparison, in (b) we plot the energy spectra (lowest 36 states) of a particular two-electron horizontal double dot as a function of an applied magnetic field along the *z* direction. Here the interdot distance is 30 nm, the dot Gaussian confinement radius is 30 nm, and the effective central barrier is 9.61 meV. Again, the inset shows the magnetic-field dependence of the splitting between the lowest singlet and triplet states at three effective barrier heights of 3.38, 6.28, and 9.61 meV.

states to mainly ψ_{P-} states. For large barriers the triplet state is the ground state even at zero field, in analogy to, for example, the oxygen molecule.

A crucial feature of our results, not obviously apparent from Fig. 2, is the constituency of the lowest two states. At zero magnetic field, the ground singlet is an equal superposition of the singlet states formed from the core and four different pairs of Fock-Darwin states: ψ_{PL-} and ψ_{PR-}, ψ_{PL-} and ψ_{PR+}, ψ_{PL+} and ψ_{PR-}, and ψ_{PL+} and ψ_{PR+}. Therefore, if initially in the single QD the outermost electron is in an arbitrary superposition of orbital *P* states, then as the barrier between the two neighboring QDs is lowered, several low-lying excited states will inevitably get involved as we project the initial state into a superposition of all the double-QD eigenstates. Indeed, using the four *P* states listed above one can form four singlet and four triplet states, so that there are seven energy parameters (neglecting the splitting of any triplet state due to external fields). Therefore, in the most general case one has to manipulate seven different phases to produce a swap—a formidable (if not completely intractable) task. As the magnitude of the external magnetic field increases, the lowest singlet and triplet states become simpler, consisting mainly the ψ_{PL-} and ψ_{PR-} states. Thus if the initial single-QD outer-shell electron state is purely ψ_{P-}, then only the lowest two states get involved as the interdot barrier is lowered, and the electron dynamics is directly analogous to the original proposal of a single electron confined in each QD. In other words, the orbital degrees of freedom for the outermost electrons are essentially frozen, so that the electron dynamics can be described by a simple spin Hamiltonian—the Heisenberg Hamiltonian, and important two-qubit operations such as swap can be easily realized.

Another important feature of the spectra in Fig. 2(a) is the splitting between the lowest two states and the higher excited states (defining the sharpness of the exchange sub-Hilbert space). This splitting is relatively small at zero and low fields, increases with the field for a few Tesla, then gradually decreases again at higher fields. This means that there exists an optimal intermediate magnetic-field regime where the adiabatic condition necessary for quantum computation can be most easily satisfied. Indeed, this optimal field regime is defined close to the $P+$ and $D-$ crossing of the Fock-Darwin state sequence.

Our results show that certain multielectron cases, such as the situation of three electrons in each quantum dot in a two-QD system, can be mapped on to the effective single-electron picture only at intermediate external magnetic fields. Essentially, the field lifts the *P*-state degeneracy so that a sufficiently large energy gap opens up between the states involved in the exchange process and higher excited states, and the six-electron ground state is formed from the single-dot three-electron ground states. The energy gap is $\hbar\omega_C$ where ω_C is the cyclotron frequency and is linearly proportional to the magnetic field. Thus, a 1-T field will lead to a 1.5-meV splitting, a large energy considering that the exchange constant *J* is typically of the order of 0.1 meV or smaller. On the other hand, at low (or zero) fields, there exists a multitude of low-lying excited states due to the *P*-state degeneracy, and the ground-state electronic wave functions are quite complicated. At high fields, there are again relatively low energy excited states coming from the lower energy *D* states. Thus the adiabatic condition dictates that

intermediate external fields near the *P*-*D* crossing provide the optimal operating condition for a multielectron quantum computer.

There are other means (not involving the application of an external magnetic field) one can employ to break the degeneracy in the *P* and higher excited states. For example, deformation of a circular quantum dot can lift the degeneracy in the *P* (and presumably all the higher excited) states, thus facilitating a more reliable and accurate two-qubit operation. If a circular parabolic well is slightly deformed into an elliptical well, the energy splitting between the two new *P* levels is $\frac{1}{2}\hbar\omega_0 e$ where e is the ellipticity. Alternatively, spin-orbit coupling can lift the orbital degeneracy, although it would be quite small in our system, as we have only a few conduction electrons at the bottom of the GaAs conduction band [16,17], and it mixes the orbital and spin states, which is what we try to avoid.

If we examine the physical picture underlying the effective qubit behavior of the multielectron scenario closely, it is clear that a crucial point is that the extra unpaired electron should not have access to low-energy excited orbital states. Thus, in general, multielectron coupled quantum-dot systems do not reduce to a simple Heisenberg exchange Hamiltonian in zero magnetic fields. One might speculate that a multielectron case may be analogous to the single-electron case when the number of electrons in a single dot is a full shell minus one: $1,5,11,\ldots,\ n(n+1)-1$, etc. However, particle-hole symmetry determines that in these cases pair breaking excitations in the outer shell will affect the low energy dynamics so that these multielectron systems would actually be similar to the three-electron (in a single dot) case we study here (and not to single electron systems). We therefore conclude that multielectron circularly symmetric quantum dot systems in zero external magnetic fields may not be suitable as solid-state spin qubits. Thus one should either use single-electron quantum dots as in the original Loss-DiVincenzo proposal (which may be a difficult task in practice) or apply external magnetic fields (or break the circular symmetry using controlled deformation) as we show in this paper. The understanding of multielectron systems as carried out in this paper may be an important step in the realistic fabrication of spin-based QD-QC architecture.

We conclude by emphasizing a general principle that is explicitly demonstrated by the theoretical results presented in this paper. Quantum computation, in contrast to regular digital Boolean classical computation, is analog, and the algorithm is defined by the system Hamiltonian. One must know the quantum Hamiltonian (e.g., the exchange Hamiltonian in our QD-QC example) controlling the qubit dynamics in the system accurately in order to carry out meaningful quantum computation. Our multielectron QD calculations compellingly demonstrate the potential problems that may arise—the effective single-electron Heisenberg exchange Hamiltonian seems an eminently reasonable choice for QD-QC until one looks carefully at the multielectron situation as we do here, finding important qualitative differences with the effective single-electron approach that can only be remedied through detailed theoretical calculations. We believe that the important lesson presented in our example in this paper is quite generic: Know your Hamiltonian well before you build your quantum computer.

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