

# Two-qubit gates between noninteracting qubits in endohedral-fullerene-based quantum computation

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We propose a scheme to implement the two-qubit gates between the nuclear spins of the encapsulated atoms in endohedral fullerenes  $^{15}\text{N} @ \text{C}_{60}$  or  $^{31}\text{P} @ \text{C}_{60}$ , within today's magnetic resonance techniques. Since there is no interaction between the nuclear spins, this scheme employs the electronic spins as medium and two swap operations are proposed to transfer the information between the nuclear spin  $\frac{1}{2}$  and the electronic spin  $\frac{3}{2}$ , and between two electronic spins  $\frac{3}{2}$ . These two-qubit gates, along with the single-qubit rotation gates, compose a universal set of quantum gates in fullerene-based quantum computation.

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## I. INTRODUCTION

Solid-state quantum computation has recently attracted much attention, for it is thought to hold the promise of building a large-scale quantum computer. This so-called scalability is an essential requirement [1] for building a practical quantum computer which can beat conventional computers on some specific problems [2–4]. Among those designs based on solid-state materials, Kane's proposal [5] appears very attractive if some technical challenges can be overcome. Specifically, it requires an accurate placement of dopant atoms inside a silicon crystal, which is rather difficult with today's technology.

This difficulty could be avoided in the proposal based on endohedral fullerenes [6–18], which encodes qubits in the electronic and nuclear spins of the encapsulated atoms N or P. The highly symmetric  $\text{C}_{60}$  molecule serves as both a sheltering environment for the inside spin system (the qubit) and a handle for the arrangement of the qubits on a surface (e.g., Si), which is realizable using state of the art scanning tunnelling microscopy (STM) techniques. Both are very important issues for solid-state quantum computation, for the former leads to long coherence times and the latter helps addressing the different qubits. These advantages make the endohedral fullerene scheme very promising. Recent experimental work by Mehring *et al.* [14] and Morton *et al.* [15,16] further demonstrates its feasibility.

In this system, the interqubit interaction required for two-qubit operations is provided by magnetic dipole-dipole coupling between nearest neighbors. Since this is an “always-on” interaction, it is necessary to design pulse sequences that refocus unwanted interactions. A theoretical proposal for such operations was given by Suter and Lim [8], using both the nuclear and electronic spin. A complication, which was avoided in their work, arises due to the different dimensions of the Hilbert spaces of nuclear and electronic spin: The electronic spin of the N and P ground state is  $3/2$ , while the nuclear spin is either  $1/2$  ( $^{15}\text{N}$ ,  $^{31}\text{P}$ ) or  $1$  ( $^{14}\text{N}$ ). To avoid this

difficulty, Ref. [8] assumed that electronic and nuclear spins were both  $1/2$ .

The purpose of this paper is to go beyond this approximation and demonstrate that analogous operations can also be designed for the full Hilbert space of the actual  $\text{N/P} @ \text{C}_{60}$ . As in Ref. [8], we encode the qubit in the nuclear spin of  $^{15}\text{N}$  or  $^{31}\text{P}$  (which is spin  $\frac{1}{2}$ ). Since there is no interaction between neighboring nuclear spins due to the shelter of  $\text{C}_{60}$  molecules [19], to perform the two-qubit gates we use the electronic spins as medium and introduce two SWAP operations to swap the information between the nuclear spin  $\frac{1}{2}$  and the electronic spin  $\frac{3}{2}$ , and between two electronic spins  $\frac{3}{2}$ . Based on them a scheme of implementing the nontrivial two-qubit quantum gates between the nuclear spins is proposed which works for arbitrary states of the electronic spins. Along with the single-qubit rotation gates which can be performed directly on the nuclear spins by radio frequency pulses, these two-qubit gates compose a universal set of quantum gates in fullerene-based quantum computation.

The paper is structured as follows: first we give a brief review of the endohedral fullerene system in Sec. II, then the proposed two-qubit gates scheme is presented in Sec. III. In Sec. IV we discuss some experimental issues in the implementation of the two-qubit gates, and Sec. V is the conclusion.

## II. THE SYSTEM

The system that we consider consists of a chain of endohedral fullerene molecules ( $^{15}\text{N} @ \text{C}_{60}$  or  $^{31}\text{P} @ \text{C}_{60}$ ), each containing a nuclear spin  $I$  and an electronic spin  $S$ . The Hamiltonian of a single molecule in an external magnetic field is in first order given by ( $\hbar=1$ )

$$H = g\mu_B BS_z - \gamma_n BI_z + AS_z I_z, \quad (1)$$

where the first two terms are the electronic and nuclear Zeeman interactions and the last term is the hyperfine interaction.  $g$  is the electronic  $g$  factor,  $\mu_B$  is the Bohr magneton and  $\gamma_n$  is gyromagnetic ratio of the nucleus. In an external field of  $B=1\text{T}$ , the electronic Larmor frequency is

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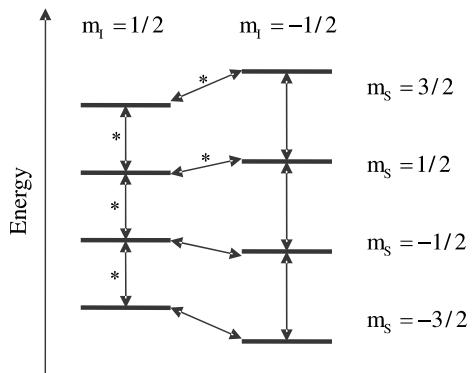


FIG. 1. The energy level diagram of endohedral fullerenes ( $^{15}\text{N} @ \text{C}_{60}$  or  $^{31}\text{P} @ \text{C}_{60}$ ). The vertical arrows indicate the allowed electronic transitions ( $\Delta m_S = \pm 1$ ) and horizontal arrows indicate the allowed nuclear transitions ( $\Delta m_l = \pm 1$ ). The starred transitions are used in the  $U_{\text{SWAP}}^{\text{SI}}$  operation (see Fig. 3 and the text).

$\nu_e \approx 28$  GHz and the nuclear Larmor frequency is  $\nu_n \approx 3.1$  MHz for  $^{15}\text{N}$  and  $\nu_n \approx 17.3$  MHz for  $^{31}\text{P}$ , while the hyperfine interaction is 21.2 MHz for  $^{15}\text{N}$  and 138.4 MHz for  $^{31}\text{P}$  [9]. Figure 1 shows the related energy level diagram with the arrows indicating the magnetic-dipole allowed transitions. The hyperfine interaction splits the ESR spectrum into two lines, each corresponding to three degenerate ESR transitions. The NMR spectrum consists of four nuclear spins transitions, corresponding to the four electronic spin states [14].

In the scalable quantum computer design of endohedral fullerenes [8], a chain of endohedral fullerenes is used. For two adjacent endohedral fullerenes (Fig. 2), the Hamiltonian can be written as [19]

$$H_{ij} = g\mu_B B_i S_z^i + g\mu_B B_j S_z^j - \gamma_n^i B_i I_z^i - \gamma_n^j B_j I_z^j + A_i S_z^i I_z^i + A_j S_z^j I_z^j + D_{ij} S_z^i S_z^j, \quad (2)$$

where  $i, j$  represent the different fullerenes, and  $B_{i(j)}$

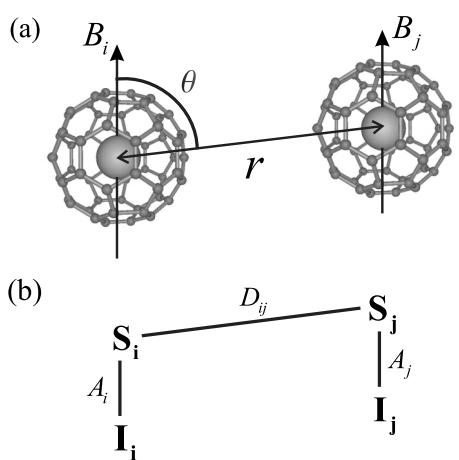


FIG. 2. Schematic illustration of two neighboring endohedral fullerenes (a) and the existing couplings (b).  $S_{i(j)}$  and  $I_{i(j)}$  represent the electronic and nuclear spins, respectively.

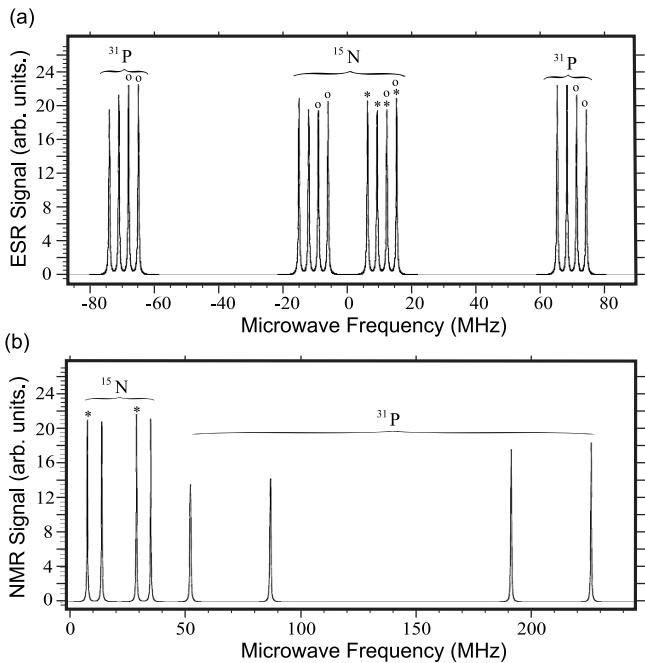


FIG. 3. Simulated ESR (a) and NMR (b) spectra of two neighboring endohedral fullerenes  $^{15}\text{N} @ \text{C}_{60}$ – $^{31}\text{P} @ \text{C}_{60}$ . The transitions labelled with stars are used in the  $U_{\text{SWAP}}^{\text{SI}}$  operation (just for  $^{15}\text{N} @ \text{C}_{60}$ ), and those labelled with circles are used in the  $U_{\text{SWAP}}^{\text{SS}}$  operation. The dipolar coupling strength chosen in the simulation is  $D=3$  MHz, which corresponds to the separation of the two fullerenes  $r \approx 2.027$  nm and  $\theta=90^\circ$ . The endohedral fullerenes are placed in an external magnetic field of 1 T. No additional addressing part is needed for this heteronuclear system, but as the tradeoff the experimental preparation of this sample is more difficult than the homonuclear sample.

represents the external magnetic field located at the site of fullerene  $i(j)$ . Here the different nuclear gyromagnetic ratios  $\gamma_n^{i(j)}$  and hyperfine interaction constants  $A_{i(j)}$  are used to describe the general situation where the spin chain consists of different types of endohedral fullerenes ( $^{15}\text{N} @ \text{C}_{60}$  and  $^{31}\text{P} @ \text{C}_{60}$ ).  $D_{ij} S_z^i S_z^j$  is the secular part of the dipolar interaction between adjacent electronic spins, where  $D \propto (1-3 \cos^2 \theta)/r^3$ ,  $r$  is the distance between the electronic spins and  $\theta$  is the angle between  $r$  and  $B$  (see Fig. 2). The coupling strength reaches a value of  $D \sim 50$  MHz for  $r=1$  nm and  $\theta=0^\circ$  [7].

To simplify the theoretical treatment, we will introduce the two-qubit gates scheme based on the heteronuclear system of two neighboring  $^{15}\text{N} @ \text{C}_{60}$  and  $^{31}\text{P} @ \text{C}_{60}$ , in which the transition frequencies of the spins naturally differ (see Fig. 3 for the simulated spectra). This avoids the additional addressing of the nuclear spins [i.e.,  $B_i=B_j$  in Eq. (2)], which is rather difficult with today's addressing technology (but is necessary for the homonuclear system). The issues of implementing the two-qubit gates in a homonuclear system will be discussed in Sec. IV B. In fact, if the transition frequencies of the nuclear spins can be separated efficiently, the procedure would be the same as in the heteronuclear system.

### III. TWO-QUBIT GATES

#### A. Two SWAP operations

The central part of our two-qubit gates proposal consists of two SWAP operations, which are used to transfer the information between the nuclear spin  $\frac{1}{2}$  and the electronic spin  $\frac{3}{2}$  in the single endohedral fullerene system, and between the two electronic spins  $\frac{3}{2}$  of the adjacent endohedral fullerenes. While a literal SWAP between the nuclear spin  $\frac{1}{2}$  and the electronic spin  $\frac{3}{2}$  is not possible due to the different spin size, we can exchange the information in the spin  $\frac{1}{2}$  system with a subspace of the spin  $\frac{3}{2}$  system.

For this purpose, we choose the pulse sequence

$$U_{\text{SWAP}}^S = [S_y^{1/2}] [-I_x^{1/2}] [-I_x^{3/2}] [-S_y^{1/2}] [S_z I_z]. \quad (3)$$

The sequence is written in time-reversed order, i.e., it starts with the delay  $[S_z I_z] = e^{-i\pi S_z I_z}$ . Here the notation  $[A] = e^{-i\pi A}$  is used. The term  $[S_y^{1/2}]$  represents a selective ( $\pi$ )<sub>y</sub> pulse on the electronic spin corresponding to the nuclear spin state  $|\frac{1}{2}\rangle$ , and  $[-I_x^{3/2}]$  represents a selective ( $\pi$ )<sub>x</sub> pulse on the nuclear spin corresponding to the electronic spin state  $|\frac{3}{2}\rangle$ . The whole sequence yields the unitary operator

$$U_{\text{SWAP}}^S = \begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & i & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & i & 0 & 0 & 0 \\ 0 & 0 & 0 & i & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & i & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 \end{pmatrix}, \quad (4)$$

which is written in the basis  $\{|m_S\rangle \otimes |m_I\rangle\}$ .

Similarly we need to exchange the information between two electronic spins  $\frac{3}{2}$ . Again, we do not require a complete exchange of their Hilbert spaces, but we only have to transfer the information from the specific subsystem that was exchanged with the nuclear spin qubit. A suitable SWAP operation is

$$U_{\text{SWAP}}^{SS} = [S_{1y}^{1/2}] [S_{1y}^{3/2}] [S_{2x}^{1/2}] [S_{2x}^{3/2}] \times [-S_{1y}^{1/2}] [-S_{1y}^{3/2}] [-S_{1z} S_{2z}], \quad (5)$$

where  $[S_{1y}^{1/2}]$  represents a selective ( $\pi$ )<sub>y</sub> pulse on the first electronic spin ( $S_1$ ) corresponding to the second electronic spin ( $S_2$ ) state  $|\frac{1}{2}\rangle$ . The corresponding propagator is a  $16 \times 16$  unitary matrix.

Though it is also not a perfect state swap operation, it does exchange information between the two electronic spins and helps to achieve the two-qubit gates between the nuclear spins of adjacent endohedral fullerenes.

#### B. Implementing the two-qubit gates

In the following text, we take the controlled-NOT (CNOT) gate as an example to show how to implement nontrivial

two-qubit gates on the nuclear spins of adjacent endohedral fullerenes. We denote the electronic and nuclear spins in the first endohedral fullerene as  $S_1$  and  $I_1$ , and the spins in the second endohedral fullerene as  $S_2$  and  $I_2$ . A CNOT gate on the two nuclear spins corresponds to the following operation: if  $I_1$  is in state  $|\frac{1}{2}\rangle$  then  $I_2$  undergoes a NOT gate and if  $I_1$  is in state  $|\frac{1}{2}\rangle$  then nothing happens. The procedure to realize this gate consists of the following four steps.

(1) Transfer the information encoded on  $I_1$  to  $S_2$  by the operation

$$U_1 = U_{\text{SWAP}}^{S_1 I_1} U_{\text{SWAP}}^{S_1 I_1}, \quad (6)$$

where the  $U_{\text{SWAP}}^{S_1 I_1}$  comes first.

(2) Perform a CNOT gate between  $S_2$  and  $I_2$  by selective rf pulses on  $I_2$ , which realize the operation

$$U_2 = [-I_{2x}^{1/2}] [-I_{2x}^{3/2}]. \quad (7)$$

(3) Transfer the information encoded on  $S_2$  back to  $I_1$  by  $U_1^{-1}$ . After this step, an operator

$$U = U_1^{-1} U_2 U_1 = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & i \\ 0 & 0 & i & 0 \end{pmatrix} \otimes \text{Id}_4 \otimes \text{Id}_4 \quad (8)$$

is realized. The two  $4 \times 4$  identity matrices  $\text{Id}_4$  are performed on the electronic spins, indicating the states of the electronic spins do not change after the above operations. This CNOT operation works independently of the initial state of the electronic spins.

(4) If needed, the additional phase  $i$  in Eq. (8) can be corrected by a phase gate  $e^{i(\pi/2)I_{1z}}$  performed on  $I_1$ . Then a standard CNOT gate  $|0\rangle\langle 0| \otimes \text{Id}_2 + |1\rangle\langle 1| \otimes \sigma_x$  is realized, here  $\sigma_x$  is the Pauli operator.

Other nontrivial two-qubit gates, like controlled-rotation (CROT) gate and controlled-phase (CPHASE) gate, can be implemented in a similar way by changing the  $S_2$  controlled-NOT gate in step (2) into an  $S_2$  controlled rotation or phase gate. In those cases, a different phase gate in step (4) is needed to correct the additional phase.

There is another way to implement the CNOT gate between the nuclear spins: first we transfer the information encoded on  $I_1$  and  $I_2$  to  $S_1$  and  $S_2$  by the swap operations  $U_{\text{SWAP}}^{S_1 I_1}$  and  $U_{\text{SWAP}}^{S_2 I_2}$ , respectively. Then we can realize a CNOT gate between the two electronic spins by the selective microwave pulses  $[S_{2x}^{1/2}] [S_{2x}^{3/2}]$  applied to  $S_2$ . Finally, the information encoded on  $S_1$  and  $S_2$  is switched back to  $I_1$  and  $I_2$  by  $(U_{\text{SWAP}}^{S_1 I_1})^{-1}$  and  $(U_{\text{SWAP}}^{S_2 I_2})^{-1}$ , respectively. If required, the phases then can be corrected by the phase gate  $e^{i(\pi/2)I_{1z}}$  on  $I_1$ .

## IV. DISCUSSION

#### A. Some experiment related issues

These two-qubit gates can be implemented with existing magnetic resonance technology. All the pulses that are used to construct the swap operations and the two-qubit gates, can be realized on a state of the art pulsed electron spin reso-

nance spectrometer equipped with a pulsed ENDOR (electron-nuclear double resonance) part, as demonstrated in Refs. [14,16]. For the realistic implementation of these two-qubit gates, a set of the system parameters, e.g., the separation of the endohedral fullerenes, should be carefully selected to achieve a good fidelity and acceptable gating times. Figure 3 just shows an example of the feasible experimental systems. A detailed research on this topic will be reported elsewhere.

In quantum computation, one needs to perform as many gates as possible during the qubit coherence time [1]. Thus a short gating time and a relatively long coherence time are required. The duration of the pulse time will depend on the fidelity that we need in the experiments, but they are roughly of the order of the inverse coupling strength. For the two neighboring endohedral fullerenes system whose spectra are simulated in Fig. 3, one can estimate that the duration of a selective ESR pulse is of the order of  $0.33 \mu\text{s}$ , and the duration of a selective rf pulse is of the order of  $1 \mu\text{s}$ . Thus the durations of  $U_{\text{SWAP}}^{SI}$  and  $U_{\text{SWAP}}^{SS}$  operations are estimated to be  $2.31 \mu\text{s}$  and  $2.05 \mu\text{s}$ , respectively, and our two-qubit gates require an operation time of the order of  $11 \mu\text{s}$ . (Note that in some operations we use semiselective pulses to hit two or more neighboring resonance lines, this needs a shorter time than the line-selective pulse.) The implementation time of the two-qubit gates is just a little shorter than the  $T_2$  ( $\sim 14 \mu\text{s}$ ) of the electronic spin, which is measured for the magnetically dilute  $^{15}\text{N}@\text{C}_{60}$  powder [7]. The spin-spin relaxation time  $T_2$  of the electronic spin is the main restriction of the coherence time in the implementation of the two-qubit gates (since the electronic spins are used as medium), and it is foremost due to the dipolar interaction with electronic spins of other endohedral fullerenes which are randomly distributed in the sample [7]. In a carefully designed scalable quantum computer scheme, these interactions should be well controlled and  $T_2$  of the electronic spin is expected to be as the same order as its spin-lattice relaxation time  $T_1$  ( $\sim 1 \text{s}$ ) [9]. Then within the coherence time of the electronic spin  $10^5$  two-qubit gates will be available.

### B. Extend to a chain of endohedral fullerenes

So far the discussion was based on the two-neighbor situation. In the scalable quantum computer design a chain of endohedral fullerenes is used. Here we discuss some issues about the implementation of the two-qubit gates in this extended system.

To carry out the quantum computation in the spin chain an appropriate addressing method is needed to address the qubits. In Ref. [8] Suter and Lim suggested using a magnetic field gradient to address the electronic qubits in the chain. This method well separates the transition frequencies of the electronic spins, but not the nuclear spins of the same type of endohedral fullerenes. For a current of  $1 \text{ A}$  (which is used to generate the magnetic field gradient), the frequency difference of the nuclear spins in two  $^{15}\text{N}@\text{C}_{60}$  molecules which are  $4 \text{ nm}$  away is just about  $5 \text{ kHz}$ . Thus in order not to affect the irrelative nuclear spins while implementing the two-qubit gates, the duration of a selective rf pulse should

have an order of  $0.2 \text{ ms}$ . It is too long compared to the coherence time of the electronic spins. So an improved addressing method is needed to implement the two-qubit gates, e.g., a way to individually tune the resonance frequencies of the nuclear and electronic spins. This would necessitate further development of addressing technologies.

The methods described in this paper offer an alternative way to implement the CNOT gate which does not require the individual addressing of the nuclear spins, i.e., to use the CNOT gate proposal described in the last paragraph of Sec. III B. In this proposal the nuclear spins just undergo two operations  $U_{\text{SWAP}}^{SI}$  and  $(U_{\text{SWAP}}^{SI})^{-1}$ . Since  $(U_{\text{SWAP}}^{SI})^{-1} \cdot U_{\text{SWAP}}^{SI} = 1$ , there is no need to selectively excite the target nuclear spins and the effective operations on the irrelative nuclear spins at the end of the CNOT gate would be the identity operations. Hence the CNOT gate still has a short operation time of the order of  $11 \mu\text{s}$ . This idea has been discussed in Ref. [8] for the situation of electronic spins  $\frac{1}{2}$ . Here we showed it also works for electronic spins  $\frac{3}{2}$ .

Another issue is the decoupling of the irrelative qubits during the gate implementation. It is easy to verify that the common decoupling technique (invert the spins at some times) can also be implemented on the electronic spin  $\frac{3}{2}$  by the inversion operation

$$[S_x] = i \begin{pmatrix} 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{pmatrix}. \quad (9)$$

Hence the decoupling methods described in Ref. [8] can also be used here.

## V. CONCLUSION

In summary, we proposed a scheme to implement two-qubit gates between the nuclear spins of the encapsulated atoms in endohedral fullerenes, taking into account the full Hilbert space of two nuclear spins  $\frac{1}{2}$  and two electronic spins  $\frac{3}{2}$ . The scheme requires selective excitation of electronic and nuclear spin transitions in addition to local addressing capabilities by a suitable addressing method. Along with the single-qubit rotation gates which can be performed directly on the nuclear spins by radio frequency pulses, these two-qubit gates compose a universal set of quantum gates for a quantum computer using the spins of endohedral fullerenes as qubits.

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