Selective pulse implementation of two-qubit gates for spin-3/2 based fullerene quantum information processing

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Abstract

We propose two potentially practical schemes to carry out two-qubit quantum gates on endohedral fullerenes $N@C_{60}$ or $P@C_{60}$. The qubits are stored in electronic spin degrees of freedom of the doped atom $N$ or $P$. By means of the magnetic dipolar coupling between two neighboring fullerenes, the two-qubit controlled-NOT gate and the two-qubit conditional phase gate are performed by selective microwave pulses assisted by refocusing technique. We will discuss the necessary additional steps for the universality of our proposal. We will also show that our proposal is useful for both quantum gating and the readout of quantum information from the spin-based qubit state.

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I. INTRODUCTION

Quantum information processing holds the promise to outperform the computational power of existing computers in the treatment of certain problems [1]. Although so far quantum gating has been successfully carried out in some systems, such as trapped ions and nuclear magnetic resonance [2], to have large-scale working quantum information processing a design based on solid-state materials may be very promising.

Since both electronic and nuclear spin states typically possess longer decoherence times than charge states, there have been a number of proposals for solid-state quantum information processing using spin-based qubits, for instance, [3, 4]. This work is focused on a spin-based quantum gating with endohedrally doped fullerenes, whose unusual properties have been mentioned in earlier publications [5, 6, 7]. These fullerenes are highly symmetric hollow molecules which behave as Faraday cages for the encapsulated atoms $N$ or $P$. The qubits can be encoded in the nuclear spins or the electronic spins of $N$ or $P$, and the quantum gating in this kind of system is done by using NMR (i.e. Nuclear Magnetic Resonance) or ESR (i.e. Electron Spin Resonance) pulse sequences. Since it is less sensitive than the electronic spin to decoherence due to environment, the nuclear spin is more suitable for hosting a qubit. However, due to larger energy spacing, quantum gating based on electronic spins can be carried out more quickly and readily than that based on nuclear spins. Therefore a promising design for quantum information processing has been proposed by encoding qubits into nuclear spins, but implementing quantum gates on electronic spins [6, 7]. The swap operation between nuclear and electronic spins via Hyperfine interaction to exchange quantum information is essential to these designs. The advantages of this type of quantum information processing device include the lengthy decoherence times of the qubit states due to the cage effects of $C_{60}$, and the potentially easier manipulation of the qubits than another proposal [3].

However, to carry out a non-trivial two-qubit gate, say, a controlled-NOT (CNOT) gate, we need at least four steps of manipulation with NMR or ESR hard pulse sequences [5, 6, 7]. Although these pulse sequences can be finished within the order of $\mu$s, since the coupling shifts the energy levels in the two coupled fullerenes with respect to the non-interacting case, the associated hard pulses must be of wide bandwidths, making their experimental realization challenging [6]. To overcome this difficulty, we can try to perform gating by
selective pulses as discussed in [6]. However, the dopant atom of $N@C_{60}$ or $P@C_{60}$ has three valence electrons, and possesses a quartet ground spin state of total spin $3/2$, instead of $1/2$ as simply treated in [6]. As a result, there are a number of degenerate transitions, which makes the problem complicated. To have a practical gating scheme, we have to first specifically study the true configuration of two coupled $S = 3/2$ spins. Based on the same system as in [5, 6, 7], we will try to propose two schemes to achieve non-trivial two-qubit quantum gates with selective pulse operations, which would be useful for both quantum gating and the qubit readout.

Our proposal is based on electronic spin degrees of freedom of the doped atom N or P, whose total electronic spin is $3/2$ with four Zeeman levels $|\pm 3/2\rangle$ and $|\pm 1/2\rangle$ in a magnetic field. Throughout the paper, we encode qubits into $|\pm 3/2\rangle$, i.e., $|3/2\rangle \equiv 1\rangle$ and $|1/2\rangle \equiv 0\rangle$. We will show that our schemes are also applicable to $|\pm 1/2\rangle$ states. Since the two neighboring fullerenes are only distant by the order of nm, to achieve a single-qubit operation by individually addressing the qubits, a magnetic field gradient has to be introduced. As shown in [6], for two nearest-neighbor fullerenes distant by 1.14 nm in the magnetic field gradient $dB/dx = 4 \times 10^5$ T/m, the difference between the ESR frequencies of $|\pm 3/2\rangle$ is $12.7 \times 3 \approx 38$ MHz, and thereby the single-qubit operation is possible using modern ESR spectrometers with narrow-band pulses.

When checking the points for universality of our proposal, however, we will have to introduce the nuclear spins of the dopant atoms for Hadamard operation. Before going to that point, we will only focus on the treatment of electronic spin degrees of freedom.

Our proposal includes two schemes for a CNOT gate and a conditional phase (CPHASE) gate respectively. Both CNOT and CPHASE are non-trivial two-qubit gates, which are the essential parts of universal quantum information processing. We will first study the configuration of the system of two coupled endohedral fullerenes. Then the main steps of our schemes will be presented. We will investigate how to eliminate unwanted relative phases. The usefulness and universality of our proposal will be also discussed.
II. CONFIGURATION OF TWO COUPLED FULLERENES

Consider a system with two $C_{60}$ fullerenes $A$ and $B$, and by neglecting the small terms associated with nuclear spins, we have in units of $\hbar = 1$,

$$H = g\mu_B B_A S^A_z + g\mu_B B_B S^B_z + JS^A_z S^B_z$$  \hspace{1cm} (1)

where $g$ is the electron $g$-factor, $\mu_B$ is the Bohr magneton, and $B_k$ ($k = A$ and $B$) is the magnetic field strength sensed by fullerene $k$. $J$ is the magnetic dipolar coupling strength between neighboring fullerenes. As in [6], we suppose $J \sim 50 \text{ MHz}$.

$$S^A_z = \frac{1}{2} \begin{pmatrix} 3 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -3 \end{pmatrix} \otimes I_B$$

and

$$S^B_z = I_A \otimes \frac{1}{2} \begin{pmatrix} 3 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -3 \end{pmatrix}$$.

Direct calculation shows that in the space spanned by

$$|\frac{3}{2}, \frac{3}{2}\rangle, \quad |\frac{3}{2}, \frac{1}{2}\rangle, \quad |\frac{3}{2}, -\frac{1}{2}\rangle, \quad |\frac{3}{2}, -\frac{3}{2}\rangle, \quad |\frac{1}{2}, \frac{3}{2}\rangle, \quad |\frac{1}{2}, \frac{1}{2}\rangle, \quad |\frac{1}{2}, -\frac{1}{2}\rangle, \quad |\frac{1}{2}, -\frac{3}{2}\rangle, \quad |\frac{3}{2}, \frac{3}{2}\rangle, \quad |\frac{3}{2}, \frac{1}{2}\rangle, \quad |\frac{3}{2}, -\frac{1}{2}\rangle, \quad |\frac{3}{2}, -\frac{3}{2}\rangle,$$

the eigenenergies are respectively,

$$3\omega_1 + 3\omega_2 + 9J/4, \quad 3\omega_1 + \omega_2 + 3J/4, \quad 3\omega_1 - \omega_2 - 3J/4, \quad 3\omega_1 - 3\omega_2 - 9J/4,$$
$$\omega_1 + 3\omega_2 + 3J/4, \quad \omega_1 + \omega_2 + J/4, \quad \omega_1 - \omega_2 - J/4, \quad \omega_1 - 3\omega_2 - 3J/4,$$
$$-\omega_1 + 3\omega_2 - 3J/4, \quad -\omega_1 + \omega_2 - J/4, \quad -\omega_1 - \omega_2 + J/4, \quad -\omega_1 - 3\omega_2 + 3J/4,$$
$$-3\omega_1 + 3\omega_2 - 9J/4, \quad -3\omega_1 + \omega_2 - 3J/4, \quad -3\omega_1 - \omega_2 + 3J/4, \quad -3\omega_1 - 3\omega_2 + 9J/4,$$

where $\omega_1 = g\mu_B B_A/2$, and $\omega_2 = g\mu_B B_B/2$. It is obvious that each level has shifted from its original position due to the magnetic dipolar coupling, and there are numerous degenerate transitions in this two spin-3/2 system, as shown in Fig. 1. This is an important difference between systems comprising of two spin-1/2 particles and two spin-3/2 particles.
III. TWO-QUBIT QUANTUM GATING

A. CNOT Gate

CNOT is the most commonly used two-qubit quantum gate. Our implementation is based on a characteristic of the configuration shown in Fig. 1, that is, the degenerate transition frequency of a spin state is heavily dependent on the coupled (or neighboring) spin state. Due to degeneracy and the magnetic field gradient, the radiation of a ESR pulse on a single qubit yields following Hamiltonian in units of $\hbar = 1$,

$$H = \omega_0 S_z + \Omega \left( e^{-i\omega_L t} S_+ + e^{i\omega_L t} S_- \right)$$  \hspace{1cm} (2)

where $\omega_0$, different from the non-interacting counterpart, is one of the degenerate transition frequencies labeled in Fig. 1. $\omega_L$ is the frequency of the microwave pulse. $S_k \ (k = z, +, \text{or } -)$ is a $4 \times 4$ Pauli operator. $\Omega$ is the Rabi frequency. When $\omega_L = \omega_0$, in the interaction Hamiltonian we have $H_I = \Omega S_x$ with

$$S_x = \frac{1}{2} \begin{pmatrix} 0 & \sqrt{3} & 0 & 0 \\ \sqrt{3} & 0 & 2 & 0 \\ 0 & 2 & 0 & \sqrt{3} \\ 0 & 0 & \sqrt{3} & 0 \end{pmatrix}.$$

For a $\pi$ pulse radiation of ESR, i.e. $\Omega t = \pi$, $H_I$ yields operator

$$\hat{P} = i \begin{pmatrix} 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{pmatrix},$$

which works independently in the subspace spanned by $| \pm 3/2 \rangle$ or the one spanned by $| \pm 1/2 \rangle$.

Therefore, with the operator $\hat{P}$, we can flip states $| \pm 3/2 \rangle$ of a single qubit with a ESR pulse whose frequency is determined by the neighboring spin state. This is actually a CNOT operation. Since with the magnetic field gradient we are able to flip different spin states with different ESR frequencies, we can perform different CNOT gates. For example, with the frequency $2\omega + \Delta - 3J/2$, we have a CNOT$_{AB}$, i.e., the target spin state in fullerene B flipped only in the case when the control qubit is $| -3/2 \rangle_A$. While to have a CNOT$_{BA}$, we use the ESR pulse with the frequency $2\omega - 3J/2$.
Given a perfect experimental implementation, the fidelity of our scheme only depends on the exact knowledge of the configuration of the considering system. The implementation time is determined by the Rabi frequency $\Omega$.

B. CPHASE Gate

CPHASE gate is another useful two-qubit gate, which plays an important role in Grover search to flip the phase of the labeled state $|\tilde{1}\rangle$. CPHASE gate is always considered to be equivalent to CNOT if single qubit rotation is easily performed. But in the system under consideration, single qubit gating is not an easy job (see Sec IV). So it is interesting to have a straightforwardly produced CPHASE gate. The key idea of our scheme is to radiate the two $C_{60}$ fullerenes by using two detuned microwaves, which is similar to what is done in ion trap quantum computing proposals [9]. However, the fullerene problem under consideration is different from atomic problems in [9]. First, there is no vibrational degrees of freedom attached to the qubit states. Secondly, we are considering a spin-$3/2$ system, which is more complicated. Thirdly, the two fullerenes under the magnetic field gradient possess different transition resonance frequencies for the two qubit states.

We employ spin states $|-1/2\rangle_A|-3/2\rangle_B$ as auxiliary states, and couple the states $|-3/2\rangle$ and $|-1/2\rangle$ of the two endohedrals by two detuned microwaves. Keep in mind that although the computational subspace is spanned by $|\frac{1}{2}\rangle_A|\frac{3}{2}\rangle_B$, the subspace under consideration is spanned by $|\frac{1}{2}\rangle_A|\frac{3}{2}\rangle_B$ and $|\frac{3}{2}\rangle_A|\frac{1}{2}\rangle_B$. From the above eigenenergies, we know that $\omega_{-1/2,-1/2} - \omega_{-3/2,-3/2} = 2\omega_1 + 2\omega_2 - 2J$, $\omega_A = \omega_{-1/2,-3/2} - \omega_{-3/2,-3/2} = 2\omega_1 - 3J/2$ and $\omega_B = \omega_{-3/2,-1/2} - \omega_{-3/2,-3/2} = 2\omega_2 - 3J/2$. So as shown in Fig. 2, an additional shift of $J$ is suffered by $|\frac{1}{2}\rangle, |\frac{3}{2}\rangle$. Since the inter-fullerene spacing is only 1.14 nm, we consider two in-plane directed microwaves, which can be described as $\cos(\omega_d t + \phi_1)\hat{e}_x + \sin(\omega_d t + \phi_1)\hat{e}_y$ with $\omega_d$ the frequency of the microwave pulse, $\phi_1$ the phase of the microwave and $l = 1, 2$. These radiate the two fullerenes simultaneously. The Hamiltonian for such a system can be written in units of $\hbar = 1$ as $H = H_0 + H_1$ where

$$H_0 = J \langle \frac{1}{2}, \frac{1}{2}\rangle \langle \frac{1}{2}, \frac{1}{2}| \frac{1}{2}, \frac{1}{2} | - \frac{3}{2}, \frac{3}{2} | \frac{1}{2}, \frac{1}{2} \rangle_{A} \otimes I_B + \frac{\omega_B}{2} I_A \otimes \left( \frac{1}{2}, \frac{1}{2} | \frac{3}{2}, \frac{3}{2} \right)_{B}$$

and

$$H_1 = \frac{\Omega_A}{2} \left[ e^{i(\omega_d t + \phi_1)} + e^{i(\omega_d t + \phi_2)} \right] | -\frac{3}{2}\rangle_A \otimes I_B + \frac{\Omega_B}{2} \left[ e^{i(\omega_d t + \phi_1)} + e^{i(\omega_d t + \phi_2)} \right] I_A \otimes | -\frac{3}{2}\rangle_B + h.c.$$

(3)
where $I_k = (|\frac{-1}{2}\rangle\langle\frac{-1}{2}| + |\frac{-3}{2}\rangle\langle\frac{-3}{2}|)_k$. For simplicity, we have supposed that $\phi_t$ is the same for two fullerenes experiencing the same microwave, and the coupling strength $\Omega_k$ is identical for each fullerene irradiated by the separate microwave sources. In the rotating frame with respect to $H_0$, we have

$$H_R = \frac{\Omega_A}{2} [e^{i(\omega_{s1} t + \phi_1)} + e^{i(\omega_{s2} t + \phi_2)}] e^{-i\omega_A t} \frac{3}{2} \langle \frac{-1}{2} | A (\frac{3}{2} \langle \frac{1}{2} | e^{-iJt} + \frac{3}{2} \langle \frac{-3}{2} | B) + \frac{\Omega_B}{2} [e^{i(\omega_{s1} t + \phi_1)} + e^{i(\omega_{s2} t + \phi_2)}] e^{-i\omega_B t} \left( \frac{-1}{2} \langle \frac{1}{2} | e^{-iJt} + \frac{3}{2} \langle \frac{-3}{2} | A \right) \otimes \frac{3}{2} \langle \frac{-1}{2} | B + h.c. \tag{5}$$

Since the two qubits are radiated simultaneously by two detuned microwave pulses, there should be four different detunings, say, $\delta_1, \delta_2, \delta_3$ and $\delta_4$, with $\delta_1 + \delta_3 = \delta_2 + \delta_4 = J$ to achieve resonance. So we consider this scheme to be also a selective pulse method. If we can make max $\{\Omega_A, \Omega_B\} \ll 2 \min \{|\delta_1|, |\delta_2|, |\delta_3|, |\delta_4|\}$, then we will force effective transitions between $|\frac{-1}{2}, \frac{-1}{2}\rangle$ and $|\frac{3}{2}, \frac{-3}{2}\rangle$, via two virtually occupied intermediate states $|\frac{-1}{2}, \frac{-3}{2}\rangle$ and $|\frac{-3}{2}, \frac{-1}{2}\rangle$ by second-order perturbative expansion \[9, 10\]. The effective Hamiltonian is thus

$$\tilde{H} = \frac{\tilde{\Omega}}{2} \left( \frac{-3}{2}, \frac{-3}{2} \right) \langle \frac{-1}{2}, \frac{-1}{2} e^{i(\phi_1 + \phi_2)} + h.c. \right) \tag{6}$$

where

$$\frac{\tilde{\Omega}}{2} = 4 \Omega_A \Omega_B \left( \frac{1}{\delta_1} + \frac{1}{\delta_2} + \frac{1}{\delta_3} + \frac{1}{\delta_4} \right), \tag{7}$$

$\delta_1 = \omega_{s1} - \omega_A, \delta_2 = \omega_{s1} - \omega_B, \delta_3 = \omega_{s2} - \omega_B, \text{ and } \delta_4 = \omega_{s2} - \omega_A$. Returning to the Schrödinger representation, the time evolutions based on Eq. (6) are

$$|\frac{-3}{2}, \frac{-3}{2}\rangle \rightarrow \cos(\tilde{\Omega}t/2)|\frac{-3}{2}, \frac{-3}{2}\rangle - i e^{-i(\omega_A + \omega_B + J)t} e^{-i(\phi_1 + \phi_2)} \sin(\tilde{\Omega}t/2)|\frac{-1}{2}, \frac{-1}{2}\rangle, \tag{8}$$

$$|\frac{-1}{2}, \frac{-1}{2}\rangle \rightarrow e^{-i(\omega_A + \omega_B + J)t} \cos(\tilde{\Omega}t/2)|\frac{-1}{2}, \frac{-1}{2}\rangle - i e^{i(\phi_1 + \phi_2)} \sin(\tilde{\Omega}t/2)|\frac{-3}{2}, \frac{-3}{2}\rangle. \tag{9}$$

Our two-qubit CPHASE gate is carried out by Eq. (8). Since $|\frac{-1}{2}, \frac{-1}{2}\rangle$ is out of our computing subspace, and is not populated initially, if we implement a $2\pi$-pulse of microwave radiation, i.e. $\tilde{\Omega}t = 2\pi$, we will have $|\frac{-3}{2}, \frac{-3}{2}\rangle \rightarrow -|\frac{-3}{2}, \frac{-3}{2}\rangle$, but no change for other qubit states in the computational subspace. This is a typical CPHASE gate with the form $|\alpha, \beta\rangle \rightarrow e^{i\alpha\beta \pi}|\alpha, \beta\rangle$, where $\alpha$ and $\beta$ are the logic state 0 or 1 respectively. To achieve this CPHASE gate, we should make sure that the implementation time is shorter than the decoherence time of the electronic spin. Since $\delta_1 + \delta_3 = \delta_2 + \delta_4 = 50 \text{ MHz}$ and $\omega_B - \omega_A = 12.7 \text{ MHz}$, Eq. (6) can be rewritten as

$$\frac{\tilde{\Omega}}{2} = 4 \Omega_A \Omega_B \left( \frac{1}{\delta_1} + \frac{1}{\delta_1 - 12.7} + \frac{1}{50 - \delta_1} + \frac{1}{62.7 - \delta_1} \right). \tag{10}$$

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To have a two-qubit CPHASE gate with high fidelity, we introduce $P = \Omega_0/2\delta_{\text{min}}$ with $\Omega_0 = \max \{\Omega_A, \Omega_B\}$ and $\delta_{\text{min}} = \min \{\delta_1, |\delta_1 - 12.7|, |50 - \delta_1|, |62.7 - \delta_1|\}$. The excitation probability of the intermediate states is defined as $\tilde{n} = 2P^2$. The smaller the value of $\tilde{n}$, the higher the fidelity of our gate, but the longer the implementation time, as shown in Fig. 3. From Fig. 3, we also know that the shortest implementation time occurs in detuning $\delta_1 = 31.35$ MHz.

IV. DISCUSSION

With current experimental techniques, Rabi frequencies of $20 \sim 30$ MHz are available [11], which fully meets the requirements of our schemes. However, compared to our CNOT gate scheme, our CPHASE gate works somewhat slowly due to the detuning and weak interaction. It takes time of the order of $\mu$s, which is only slightly shorter than $T_2 (\sim 20\mu$s) of the electronic spin at low temperature ($\sim 7^\circ$ Kelvin, high spin concentration) [12]. So, in order to carry out this scheme, we need an increased $T_2$. With decoupling, and decreasing spin density, $T_2$ will be increased to $T_1 (\sim 1$ sec) [7]. The expectation of $T_2$ prolonged toward $T_1$ is essentially not only to our proposal, but also to all the other spin-based quantum information processing schemes. If $T_2$ can be as long as 1 sec, millions of our CPHASE gates will be available within the decoherence time of the electronic spin.

The numerical simulation for the CPHASE gate also shows that the smaller the difference between $\omega_1$ and $\omega_2$, the shorter the implementation time of our gating. The shortest implementation time is $1.54 \mu$s in the case of $\omega_1 = \omega_2$ and $\delta_1 = \delta_2 = \delta_3 = \delta_4 = J/2$, which corresponds to the situation of $dB/dx = 0$. This can be understood in that the optimal operation of our gate corresponds to the same detunings of the two microwaves for two exactly identical fullerenes. However, in the architecture proposed in [5, 6, 7], a magnetic field gradient is necessary for single qubit operation. What is the optimal field gradient depends on the fluctuations of the magnetic field, our capability to distinguish different fullerenes, and $T_2$ of the electronic spins.
A. Single Qubit Operation

So far, we have only discussed the non-trivial portion of the two-qubit operation. Before discussing the rephasing of the trivial system dynamics, it is essential to note that to carry out universal quantum information processing we need single qubit rotation besides the above mentioned two-qubit gates. Due to the magnetic dipolar coupling, as mentioned above, the transition frequencies between the two qubit states are dependent on the relative spin states. This enables CNOT gates but complicates the implementation of single qubit rotations. Fortunately, the operator \( \hat{P} \) can be carried out independently in \(| \pm 3/2 \rangle \) or \(| \pm 1/2 \rangle \). So in the presence of the magnetic field gradient we can perform the single qubit operation by simultaneously irradiating with two selective ESR pulses. For example, to flip a single spin state in fullerene B, we use ESR pulses with frequencies \( 2\omega + \Delta - 3J/2 \) and \( 2\omega + \Delta + 3J/2 \). If the Rabi frequency is 25 MHz, the implementation time would be 0.126 \( \mu s \).

However, we have no way to execute a Hadamard gate on the \(| \pm 3/2 \rangle \) subspace within our model. To this end, we have to introduce nuclear spins, as done in Ref. In most doped fullerene proposals for quantum information processing the qubits are encoded in nuclear spins and the electronic spins are only employed for quantum gating, as mentioned in Sec I above. Therefore, with fullerenes containing both nuclear and electronic spins, our schemes incorporating selective pulse operations for two-qubit gates on electronic spins will suffice to yield universal quantum information processing.

B. Removal of relative phases

What we have proposed in section III above however, is the nontrivial portion of the specified two-qubit gates with one-step manipulation. If we consider a general case, i.e. the initial two-qubit state to be \( a|3/2, 3/2 \rangle + b|3/2, -3/2 \rangle + c| -3/2, 3/2 \rangle + d| -3/2, -3/2 \rangle \), our schemes could not be carried out so simply because additional relative phases will appear in the superposition during the gating due to free evolution. To get rid of these trivial phases, we have to use a refocusing pulse sequence of \( \pi/2 \) hard ESR pulses \( \square \).

For instance, within \( \tau \), we have finished a CPHASE gate with our scheme. In contrast to the desired state \( \Psi_1 = a|3/2, 3/2 \rangle + b|3/2, -3/2 \rangle + c| -3/2, 3/2 \rangle - d| -3/2, -3/2 \rangle \), we
actually have \( \Psi_2 = ae^{-i\theta_1}|3/2, 3/2\rangle + be^{-i\theta_2}|3/2, -3/2\rangle + ce^{-i\theta_3}|-3/2, 3/2\rangle - d|-3/2, -3/2\rangle \), where \( \theta_1 = (12\omega + 3\Delta)\tau \), \( \theta_2 = (6\omega + 3\Delta - 9J/2)\tau \), \( \theta_3 = (6\omega - 9J/2)\tau \), and we omitted the global phase. By making use of \( e^{i\pi S_z} S_x e^{-i\pi S_z} = -S_z \), we can remove the relative phases by sequentially sending \( \pi/2 \) hard ESR pulses to specific electronic spins of the fullerenes and then waiting for \( \tau \). As shown in Table I, the undesired phases are eliminated by continuously carrying out the three steps. The removal of relative phases for our CNOT gating can be done analogously.

Although our scheme is more complicated with the inclusion of this rephasing pulse sequence, our schemes could perform quantum computing correctly, and the implementation is not more difficult than in \[6,7\]. Moreover, the refocusing is a mature technique, which has been widely applied in various experiments of NMR \[1\]. Furthermore, we will show below that our schemes would be also very useful for the last stage of quantum computing, i.e. the qubit readout.

C. Readout

The readout problem in spin-based solid state quantum computing is still an open question \[14\]. For fullerene-based quantum information processing, possible methods to read out the states of the qubit may be to use Magnetic Resonance Force Microscopy \[15\] or spin-dependent Single Molecule Transistor \[16\]. Achieving single spin sensitivities in such methods will require significant developments. Another promising technology for spin state detection is Micro-SQUID (superconducting quantum interference device). It had been reported that this device is able to detect the spin state in systems with \( S \geq 10 \) \[17,18\]. So by coupling a nanomolecular magnet \( Fe_8 \) or \( Mn_{12} \) with total spin \( S = 10 \) \[18\] to an adjacent fullerene in a magnetic field, we can attempt a SWAP or CNOT between \( |\pm 3/2\rangle \) states of our fullerene and \( |\pm 10\rangle \) states of the nanomolecular magnet by our selective pulse scheme, along with quantum tunneling of magnetization \[19\]. Since the spin states should be well polarized in the readout stage of quantum information processing, no relative phase would appear during the gating and thereby no repair step should be taken. So the CNOTs could be carried out by one-step operations. After the information is converted from \( |\pm 3/2\rangle \) to \( |\pm 10\rangle \) \[20\], our readout can be achieved by detecting \( |\pm 10\rangle \) with an array of Micro-SQUIDs.
V. CONCLUSION

To achieve our schemes, we need stable and homogeneous magnetic field gradients as well as reliable radiation sources. As stated in [6], the desired magnetic field gradient can be provided by currents through nanometer-sized wires. Experimentally, high quality ESR pulses have been widely applied, and utilizing two detuned radiation sources is already a sophisticated technique [10]. With current $T_2$ of the electronic spins, our proposed CNOT can be carried out with fidelity of 100% by more than 100 times, and our CPHASE with the fidelity of 95% can be achieved coherently approximately eight times.

Although our discussion has been focused on qubit states $|±3/2\rangle$, our proposal is easily applicable to $|±1/2\rangle$ encoding qubits. In this case, for example, the CPHASE gate works with $|3/2\rangle$ to be the auxiliary state. Moreover, as discussed in [7], with a nonlinear term in the spin Hamiltonian, for example, a Zero-Field-Splitting term $\sim S_z^2$, one can lift the degeneracy and can execute a Hadamard gate on qubit states $|±1/2\rangle$ using selective excitations. Furthermore the rephasing steps are identical to the above case of $|±3/2\rangle$. So universal quantum information processing is available in principle in this case too.

In conclusion, two potentially practical two-qubit gates have been proposed for spin-based quantum information processing with endohedral fullerenes. Since we considered a true system with coupling spin-3/2 components, the situation is more complicated than in [6]. By using selective pulses and excluding hard, wideband pulses, we can reduce the susceptibility to decoherence. We have discussed how to efficiently implement the selective pulse schemes and shown that our schemes are alternative ways to two-qubit gating in fullerene based systems. Furthermore, it can be easily checked that our proposal, based on the pairwise coupling, i.e. the magnetic dipolar coupling, can be generalized to the multi-fullerene case, where our proposed quantum gates can be implemented in parallel. Based on the above discussion, we argue that our proposal is not only useful for quantum gating, but also advantageous in the achievement of single spin detection.

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[20] Here we only briefly describe the transfer of spin message between two well polarized states in the readout stage of quantum information processing. The detailed discussion for our readout scheme based on a large spin nanomolecular magnet Fe$_8$ or Mn$_{12}$ will be given in [19].
Table I. The refocusing steps for removing relative phases, where $\theta_1$, $\theta_2$ and $\theta_3$ are the undesired phases related to $|3/2, 3/2\rangle$, $|3/2, -3/2\rangle$ and $|-3/2, 3/2\rangle$, respectively. $[S_k]$ means $\exp(-i\pi S^k_x)$ with $k = A, B$. $H$ is given in Eq. (1).

| Step | Refocusing | Relative phases |
|------|------------|-----------------|
| 0    |            | $\theta_1 \theta_2 \theta_3$ |
| 1    | $W_1 = [-S^A_x] \exp(-iH\tau)[S^A_x]$ | 0 0 2$\theta_3$ |
| 2    | $W_2 = [-S^B_x] \exp(-iH\tau)[S^B_x]$ | $-\theta_1 \theta_2 \theta_3$ |
| 3    | $W_3 = [-S^A_x][-S^B_x] \exp(-iH\tau)[S^B_x][S^A_x]$ | 0 0 0 |
\[
\begin{align*}
\text{(a)} \\
|3/2,-3/2> & \quad |3/2,-1/2> & \quad |3/2,1/2> & \quad |3/2,3/2> \\
0 & \quad 2\omega & \quad 4\omega & \quad 6\omega \\
-2\omega & \quad 0 & \quad 2\omega & \quad 4\omega \\
-4\omega & \quad -2\omega & \quad 0 & \quad 2\omega \\
-6\omega & \quad -4\omega & \quad -2\omega & \quad 0 \\
(2\omega-3J/2) & \quad (2\omega-J/2) & \quad (2\omega+J/2) & \quad (2\omega+3J/2) \\
(2\omega+\Delta-3J/2) & \quad (2\omega+\Delta-J/2) & \quad (2\omega+\Delta+J/2) & \quad (2\omega+\Delta+3J/2)
\end{align*}
\]
FIG. 1: (a) The spectrum of the eigenenergies of the two fullerene system coupled by the magnetic dipole-dipole interaction, where $\omega = \omega_1$ and $\Delta = 2\omega_2 - 2\omega_1 = 12.7\text{ MHz}$. (⋯) represents the degenerate frequency difference between the nearest-neighbor levels in a row or in a column. (b) The spectrum of transition frequency corresponding to (a), where $a = -3/2, -1/2$ and $1/2$. The dashed lines correspond to the shift of the solid counterparts by $\Delta$. 
FIG. 2: Two neighboring fullerenes $A$ and $B$, radiated simultaneously by two in-plane directed microwaves, coupling spin states $|-1/2\rangle$ and $|-3/2\rangle$. The two-photon process is for resonant transition between $|-1/2,-1/2\rangle$ and $|-3/2,-3/2\rangle$, where $|-1/2,-3/2\rangle$ and $|-3/2,-1/2\rangle$ are non-populated intermediate states due to large detuning and weak radiation coupling.
FIG. 3: The implementation time $T = 2\pi/\bar{\Omega}$ with respect to $\delta_1$ in the implementation of our proposed CPHASE gate, based on Eq. (7). The dotted, dashed and solid curves correspond to gate fidelity 98\% (i.e., $\Omega_A/2 = \Omega_B/2 = \delta_{\text{min}}/10$), 95\% (i.e., $\Omega_A/2 = \Omega_B/2 = \delta_{\text{min}}/7$), and 92\% (i.e., $\Omega_A/2 = \Omega_B/2 = \delta_{\text{min}}/5$), respectively.