



An endohedral fullerene-based nuclear spin quantum computer

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ABSTRACT

We propose a new scalable quantum computer architecture based on endohedral fullerene molecules. Qubits are encoded in the nuclear spins of the endohedral atoms, which possess even longer coherence times than the electron spins which are used as the qubits in previous proposals. To address the individual qubits, we use the hyperfine interaction, which distinguishes two modes (active and passive) of the nuclear spin. Two-qubit quantum gates are effectively implemented by employing the electronic dipolar interaction between adjacent molecules. The electron spins also assist in the qubit initialization and readout. Our architecture should be significantly easier to implement than earlier proposals for spin-based quantum computers, such as the concept of Kane [B.E. Kane, Nature 393 (1998) 133].

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1. Introduction

Solid-state quantum computation is thought to hold the promise of building a large-scale quantum computer, which would be qualitatively more powerful than conventional computers for some specific problems [1–3]. Among the designs using solid-state materials, Kane's proposal [4], which uses donor nuclear spins in silicon, appears very attractive if the technical challenges can be overcome. Specifically, it requires donor atoms placed with nanometer precision inside a silicon crystal, which is rather difficult with today's technology.

This difficulty is avoided in the proposals based on endohedral fullerenes [5–9], in which the qubits are placed on the surface rather than inside the crystal. The C₆₀ molecule can serve as a handle for the arrangement of the qubits on the surface, which is possible with state-of-the-art scanning tunneling microscopy (STM) techniques [10,11]. Its highly symmetric structure also provides a sheltering environment for the enclosed spin. The nitrogen and phosphorus atoms inside these cages are virtually free atoms, with very long relaxation times [5,7]. These properties make the endohedral fullerenes very promising for storing quantum information.

While the previous endohedral fullerene proposals mainly use the electron spins as qubits, here we propose a scalable quantum computer scheme based on the nuclear spins. The main advantage of using the nuclear spins is their even longer spin coherence

times, compared to the electron spins. The electron spins are used here as auxiliaries which help for addressing individual nuclear spin qubits, providing effective couplings between them, as well as for initialization and readout of the nuclear spin qubits. In this respect, our approach is similar to that of Kane, but it may incur fewer technical challenges.

2. Qubit implementation

We consider a chain of endohedral fullerene molecules (¹⁵N@C₆₀ or ³¹P@C₆₀) placed in an external magnetic field oriented along the z-direction. Each molecule contains a nuclear spin $I = \frac{1}{2}$ and an electron spin $S = \frac{3}{2}$, whose Hamiltonian is in first order given by ($\hbar = 1$)

$$H_0 = g\mu_B B_0 S_z - \gamma_n B_0 I_z + A S_z I_z, \quad (1)$$

where the first two terms are the electron and nuclear Zeeman interactions and the last term is the hyperfine interaction. g is the electron g -factor, μ_B is the Bohr magneton and γ_n the gyromagnetic ratio of the nucleus. In an external field of $B_0 = 1$ T, the electron Larmor frequency is $\nu_e \approx 28$ GHz and the nuclear Larmor frequency is $\nu_n \approx 3.1$ MHz for ¹⁵N and $\nu_n \approx 17.3$ MHz for ³¹P, while the hyperfine interaction is 21.2 MHz for ¹⁵N and 138.4 MHz for ³¹P [7]. Fig. 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 (Fig. 2), each corresponding to three degenerate ESR transitions. The NMR spectrum consists of four nuclear spin transitions, corresponding to the four electron spin states [12]. Both spins can be manipulated by resonant radio-frequency (rf) and microwave pulses respectively.

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The Hamiltonian of the linear chain of N endohedral fullerenes (with the same molecules) is

$$H_{\text{chain}} = \sum_{i=1}^N (\Omega_S^i S_z^i - \Omega_I^i I_z^i + A S_z^i I_z^i) + \sum_{i<j} D_{ij} S_z^i S_z^j, \quad (2)$$

where the first term is the Hamiltonian for each molecule, and the second term contains the truncated dipolar interactions between the electron spins. $\Omega_S^i = g\mu_B B^i$ is the Larmor frequency of the i th electron spin, which experiences the field $B^i = B_0 + B_G^i$, where B_G^i describes the position-dependent component used to address the electron spins. Similarly, $\Omega_I^i = \gamma_n B^i$ is the nuclear Larmor frequency of the same molecule. The magnetic dipole–dipole coupling between the electron spins is the only strong coupling between the molecules [13], with the coupling strength $D_{ij} \propto (1 - 3\cos^2\theta)/r_{ij}^3$, where $r_{ij} = |\vec{r}_{ij}|$ is the distance between the electron spins and θ is the angle between \vec{r}_{ij} and \vec{B} . To have specific numbers, we will in the following assume that $r = 3.5$ nm is the spacing between nearest neighbors and $\theta = 90^\circ$. This corresponds to a coupling strength of $D_{ij} = D_0/(j-i)^3$ between the j th and i th electron spins, with $D_0 \approx 0.92$ MHz. As discussed below, under the conditions assumed here, the corresponding frequency difference between the j th and i th electron spins is $\approx 39(j-i)$ MHz $\gg D_{ij}$. The truncation of the dipole–dipole coupling is therefore very well justified.

Qubits are encoded in the nuclear spins. To address a specific qubit, we need a way to distinguish between them. In Kane's proposal [4], electrodes that access individual molecules (the A -gates) are used to control the resonance frequencies of the donor ^{31}P nuclear spins. However, this method still remains technically very challenging and has not been implemented so far. Magnetic field gradients were proposed as an alternative addressing mechanism [6]. If currents of $I = 1$ A flow through 2 parallel wires at

a distance of 1 μm , they generate a magnetic field gradient of ≈ 4 G/nm. This yields a frequency difference between the electron spins of adjacent molecules (which are 3.5 nm apart) of ≈ 39 MHz.

The corresponding frequency differences for adjacent nuclear spins are much smaller: 4.3 kHz for ^{15}N and 24.2 kHz for ^{31}P . Applying selective pulses at such small frequency differences would result in very slow gate operations lasting several hundred microseconds.

Here we propose a method to effectively address the nuclear spins with the assistance of the electron spins. Due to the electron–nuclear hyperfine interaction (Eq. (1)), the different electron spin states would yield different local magnetic fields on its corresponding nuclear spin, resulting in different transition frequencies. For example, the frequencies of the ^{15}N nuclear spin corresponding to the two electron spin states $|-\frac{3}{2}\rangle$ and $|\frac{3}{2}\rangle$ differ by $\approx 2\nu_n \approx 6.2$ MHz when $B_0 = 1$ T. For ^{31}P it is even larger. In our scheme we utilize this effect to achieve the nuclear spin addressing. Specifically, we distinguish two modes of an individual nuclear spin: the *passive mode*, which corresponds to the electron spin state $|-\frac{3}{2}\rangle$ and the *active mode*, which corresponds to the electron spin state $|\frac{3}{2}\rangle$. These two modes of the nuclear spin have very distinct resonance frequencies (see Fig. 2) and we can switch between them by flipping the electron spin with selective microwave π pulses. When the quantum computer is idle all the electron spins stay in its ground state $|-\frac{3}{2}\rangle$, and hence all the nuclear spins are in their passive modes. If one needs to operate on a specific nuclear spin, it is first switched to the active mode by flipping its corresponding electron spin to the state $|\frac{3}{2}\rangle$. The nuclear spin in the active state can be individually manipulated by resonant rf pulses (with a speed ~ 1 μs), without affecting the passive qubits. Afterwards the nuclear spin is switched back to its passive mode by flipping the corresponding electron spin back to the state $|-\frac{3}{2}\rangle$. The above procedure achieves the addressing of the nuclear spins.

The nuclear spin addressing method presented here requires no additional apparatus as long as the electron spins are addressed. As mentioned above the latter may be realized by the magnetic field gradients method with existing technology. The requirement that the electron spins are fully polarized to its ground state is a natural consequence of the initialization step of the quantum computer and can be achieved within reasonable experimental conditions, as discussed below. This initialization ability is a basic requirement for all physical implementations of quantum computation [14]. Besides, since the electron spin states $|-\frac{3}{2}\rangle$ and $|\frac{3}{2}\rangle$ are the eigenstates of the static Hamiltonian (Eq. (2)), and the electron spins in fullerenes possess a very long spin–lattice relaxation time (≥ 1 s at a temperature of 5 K [5] and much larger in the mK region), the two electron spin states can be well preserved during the computation.

The magnetic field gradient used for addressing the electron spins also results in a frequency distribution of the nuclear spins.

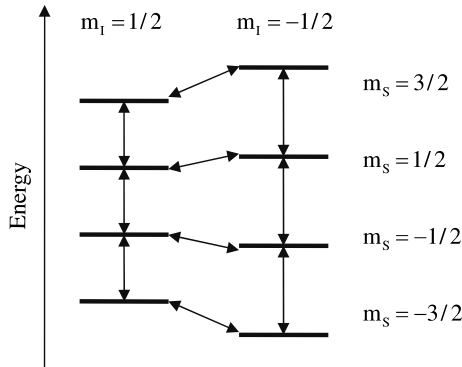


Fig. 1. The energy level diagram of endohedral fullerene ($^{15}\text{N}@C_{60}$ or $^{31}\text{P}@C_{60}$). The vertical arrows indicate the allowed electronic transitions ($\Delta m_s = \pm 1$) and horizontal arrows indicate the allowed nuclear transitions ($\Delta m_I = \pm 1$).

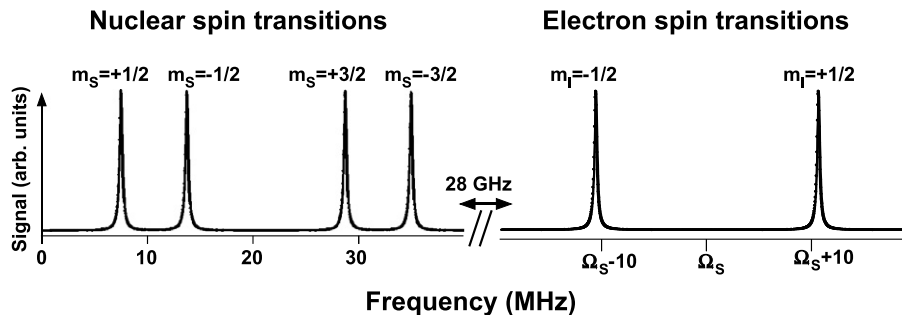


Fig. 2. Allowed magnetic resonance transitions of $^{15}\text{N}@C_{60}$ ($B_0 = 1$ T). The transition frequencies of the nuclear spin (low-frequency range) are labeled with the corresponding states of the electron spin and vice versa. The amplitudes of the electron- and nuclear transitions correspond to the high-temperature limit and the nuclear spin transitions are in reality much weaker than the electron spin transitions.

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