

Quantum-cellular-automata quantum computing with endohedral fullerenes

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We present a scheme to perform universal quantum computation using global addressing techniques as applied to a physical system of endohedrally doped fullerenes. The system consists of an $ABAB$ linear array of group-V endohedrally doped fullerenes. Each molecule spin site consists of a nuclear spin coupled via a hyperfine interaction to an electron spin. The electron spin of each molecule is in a quartet ground state $S = 3/2$. Neighboring molecular electron spins are coupled via a magnetic dipole interaction. We find that an all-electron construction of a quantum cellular automaton is frustrated due to the degeneracy of the electronic transitions. However, we can construct a quantum-cellular-automata quantum computing architecture using these molecules by encoding the quantum information on the nuclear spins while using the electron spins as a local bus. We deduce the NMR and ESR pulses required to execute the basic cellular automaton operation and obtain a rough figure of merit for the number of gate operations per decoherence time. We find that this figure of merit compares well with other physical quantum computer proposals. We argue that the proposed architecture meets well the first four DiVincenzo criteria and we outline various routes toward meeting the fifth criterion: qubit readout.

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I. INTRODUCTION

Quantum-information processing relies on the ability to perform a selection of unitary operations on a multipartite system. More precise criteria that must be met by any physical implementation of a quantum information processor have been proposed in [1]. In particular, in a spin-based quantum computer architecture the spin sites should (i) be easy to physically manipulate, (ii) be easy to replicate in numbers, (iii) possess some type of intersite interaction, and (iv) also be somewhat isolated from their environment. Meeting such ideals in practice is very demanding. It is believed by some that a solid-state implementation could prove highly advantageous in terms of the scaling up of the number of spin sites, and many solid-state proposals for a quantum-information processor are currently under study [2–4]. However, to achieve (iv) usually requires the spin sites to be located within a near-perfect crystal lattice and this leads to great difficulties in satisfying (i) and (ii) above. If the interaction between spin sites is mediated by a predominantly electronic interaction, as is the case in ion-trap quantum computers and [2], the quantum computer operation will be very sensitive to stray electric charges. Such systematic charge noise can be a source of heating and may be very difficult to eliminate. Additionally, the use of local qubit addressing is ubiquitous in very many quantum computer designs. However, the use of such local qubit addressing comes with a potentially very large overhead, such as the building of nanoscopic metal gates as in [2], or the greatly increased bandwidth required in NMR quantum computing in order to frequency address a large number of spins in a molecule [5]. For small numbers of qubits, this overhead may be acceptable, but the decoherence effects of imperfect local addressing in a device containing many tens of qubits may be much more serious. Fi-

nally, the use of nuclear spins alone in a spin-based quantum computer design leads to a serious difficulty in achieving a sufficiently high initial spin polarization with which to initialize the quantum computer. This is a primary obstacle in liquid-state NMR quantum computing, and the problem remains in solid-state designs even when the nuclei are cooled to millikelvin temperatures.

Faced with all these challenges, a number of us [6–8], following ideas put forward by the author in [6], have considered a molecular-based quantum computer which may offer a potentially more robust packaging of the qubit. The central idea is to encode a qubit into the spin system contained *within* an endohedrally doped fullerene.

In earlier publications [7], we outlined the unusual properties of group-V C_{60} endohedral fullerenes and indicated their potential use in a solid-state implementation. However, we did not there give a detailed description of a concrete quantum computing architecture for use with this material.

In [8], a locally addressed quantum computer design using endohedral fullerenes was proposed. Briefly, this design stored the qubit on the nuclear spin of the endohedral atom and used the endohedral electron spins as a local bus. The interaction between neighboring qubits is mediated by the magnetic dipole force between neighboring endohedral electrons. By constructing a linear chain of molecules and subjecting the chain to a large magnetic field gradient, one can frequency address the electronic spin of an individual molecule and through this perform universal quantum computation.

This type of design has a number of serious drawbacks. (a) To achieve fast local addressing the resonant frequencies of neighboring endohedral electrons must be well separated. Thus the microwave bandwidth required to frequency address many frequency separated spins grows with the number of qubits and may become technically quite challenging to achieve beyond a few hundred megahertz (which corresponds to a few tens of qubits). In addition, (b) to be scalable

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one must perform quantum error correction and to do so will require the simultaneous performance of various quantum gates within the device. Thus the number of independent spectrometer frequency channels required to produce such multifrequency microwave pulses will grow with the device. To engineer such a spectrometer may be not be ultimately scalable. Finally, (c) to achieve local addressing in [8] requires the generation of a magnetic field that is highly stable in time and possesses a very large, and preferably very homogeneous, spatial gradient. Currently, spectrometers are engineered to yield highly homogeneous, ultrastable constant magnetic fields (zero spatial gradients), and the engineering of the magnetic fields desired in [8], on a microscopic scale, although possible, will require further development.

The purpose of this work is to show that a quantum-cellular-automaton quantum computing architecture (QCAQCA), can successfully be applied to operate with group-V C_{60} endohedrals. Further, the resulting design does not exhibit the above mentioned drawbacks seen in [8]. The QCAQCA design requires only a homogeneous constant magnetic field. More importantly, in the QCAQCA, full scale universal quantum computation with quantum error correction requires frequency addressing at only a fixed, small number of frequencies, independent of the size of the device. These two advantages mean that a quantum-cellular-automaton-based device may be far easier to develop in hardware than any locally addressed architecture.

For the design developed below we exhibit the specific pulse primitives necessary for quantum-cellular-automaton quantum computation. We are also able to estimate rough figures of merit, or the number of quantum gates that can be executed in a decoherence time. The estimated figure of merit obtained below compares well with other proposed quantum computer designs.

In Sec. II below we review the properties of $\mathcal{X}@C_{60}$ ($\mathcal{X} = {}^{15}\text{N}, {}^{31}\text{P}$), pertinent for quantum-information processing. In Sec. III we briefly outline the essential ingredients of QCAQCA. In Sec. IV we develop a number of tools (pulse sequences) for manipulating the Hamiltonian of an alternating linear chain $ABABAB$ of these endohedrals where $\mathcal{A} = {}^{15}\text{N}@C_{60}$ and $\mathcal{B} = {}^{31}\text{P}@C_{60}$. In Sec. V we examine the possibility of an all-electronic QCAQCA and find that universal quantum computation seems not possible using the electrons alone. In Sec. VI we consider an architecture that also includes the nuclear spins and show, by developing new gate pulse sequences, that universal quantum computation can be achieved via a quantum-cellular-automata design. Finally, in Sec. VII, we propose a number of readout techniques that might be applicable to the proposed ensemble (a type-II quantum computer design [9]), and single-issue (or a type-I quantum computer design [9]) quantum computer designs.

II. GROUP-V ENDOHEDRALS

The endohedrals ${}^{15}\text{N}@C_{60}$, ${}^{31}\text{P}@C_{60}$, and $\text{N}@C_{70}$ all exhibit very sharp electron spin resonance (ESR) spectra [10,11]. This indicates the presence of free electrons within these molecules. It has been further shown, both theoretically

and experimentally [12–14], that the trapped atom (N or P), is at the geometric center of the fullerene cage and that the electrons are in an $S=3/2$ quartet ground state. The trapped atoms are extreme examples of compressed atoms [12,15], where the electronic wave function of the trapped atom is repulsed away from the encompassing carbon cage and suffers a spatial compression. This unique type of “bonding” leads to significantly higher nuclear-electron wave function overlap and thus larger hyperfine coupling constants than are found in “free” atoms. For the case of C_{60} group-V endohedrals, the large symmetry renders this hyperfine coupling highly isotropic. Another consequence of this compression is the virtual lack of any electronic interaction between the trapped atom and the carbon cage. The trapped atom is motionally confined by a harmoniclike potential to the center of the cage. In almost all respects, the trapped atom behaves as a “free” (unbonded) atom, although spatially restricted to be within the fullerene cage. All of the above findings imply that such endohedrals behave as *nanoscopic molecular neutral atom traps*.

The distributed π bonding electrons on the C_{60} also act as an almost perfect Faraday cage, strongly isolating the electrons of the trapped atom from external electric fields [16]. This observation implies that it would require the application of intense local electric field gradients ~ 1 V/nm in order to alter the hyperfine coupling constant of the trapped atom. Such an electrostatic addressing scheme has been proposed to address nuclear spin qubits in the phosphorus donors of the Kane design [2], but with far lower field gradients. Such a scheme would prove very difficult to execute here due to the requirement of such intense electric field gradients. One very significant advantage of containing the spin site inside a relatively large C_{60} molecule (diameter ~ 1 nm) is that such endohedrals can be nanopositioned using current scanning tunneling microscope techniques [17], and moreover neighboring C_{60} molecules can be nanopositioned on the silicon(100)- $2\times$ surface to be as close as ~ 1.1 nm. The C_{60} molecules, on the silicon(100)- $2\times$ surface, are fixed translationally at room temperature and also rotationally at lower temperatures. More speculatively, due to the electronic wave function spatial compression experienced by these molecules, the chemistry of the doped group-V fullerene material is practically identical to that of undoped fullerene. It may then be possible to self-assemble large organized spin structures using existing well-studied synthetic supramolecular chemistry techniques [7].

The very sharp ESR spectra from these molecules indicate very long relaxation times, and more recent measurements have shown that the electronic relaxation times are $T_1 \sim 1$ s at $T \sim 7$ K, while $T_2 \sim 20 \mu\text{s} \forall T$ [18]. No nuclear relaxation times have yet been recorded but they are expected to be several orders of magnitude longer than the electronic relaxation times. The measured T_2 time contains contributions from several sources, e.g., unresolved dipolar couplings. The theoretical maximum of T_2 , in the complete absence of the unwanted interactions with other paramagnetic impurities, is the relaxation time T_1 . Indeed, for the case of phosphorus defects in isotopically ultrapure ${}^{28}\text{Si}$, the phase relaxation time of the loosely bound phosphorus electrons

TABLE I. Table of hyperfine coupling constants and interaction energies for interacting $^{15}\text{N}@C_{60}$ and $^{31}\text{P}@C_{60}$ molecules at $B_z=2$ T.

		$\mathcal{A}=^{15}\text{N}@C_{60}$	$\mathcal{B}=^{31}\text{P}@C_{60}$
Electronic Zeeman energy	$g_e\mu_e B_z$	56 GHz	56 GHz
Nuclear Zeeman energy	$g_N\mu_N B_z$	-6.1 MHz	34.5 MHz
Hyperfine coupling constant	A	21.2 MHz	138.4 MHz

can be as long as $T_2 > 0.1$ ms with $T_1 > 1$ h [19]. There T_2 is limited by hyperfine interactions with residual ^{29}Si nuclei [20]. The indicated increase of T_2 toward T_1 in the situation of vanishing spin density [21] is crucial for all current spin-based quantum computer proposals [3–8,22,23]. Achieving such limits will be very challenging. Making these assumptions for the endohedral electrons we expect $T_2 \sim T_1 \sim 1$ s, at $T \sim 7$ K, with perhaps longer times at lower temperatures. Such electronic T_1 relaxation times (~ 1 s) in such complicated molecules are highly unusual and may be unique in all noncrystal-hosted spin sites. Long relaxation times also add to the usefulness of such materials as hosts for storing and manipulating quantum information.

An essential ingredient for any quantum-information processing is an interqubit interaction which can generate entanglement. For group-V C_{60} endohedrals this cannot be a direct electronic exchange type interaction as the electronic wave functions are tightly compressed to be within the C_{60} cage. The C_{60} 's Faraday cage does not restrict magnetic interactions and neighboring endohedrals experience a significant magnetic dipole coupling $H_D \sim \tilde{J}(1 - 3\cos^2\theta)[3\hat{I}_z^A\hat{S}_z^B - \hat{\mathbf{I}} \cdot \hat{\mathbf{S}}] \sim J_D\hat{I}_z^A\hat{S}_z^B$ in the weak coupling limit. The strength of this dipolar coupling has been estimated to be $J_D \sim 50$ MHz $\times (1/r^3)$, where r is the separation between the neighboring trapped atoms in the endohedrals measured in nanometers [24] and θ is the angle between \vec{r} and the external \vec{B} field. The hyperfine coupling constants for $^{15}\text{N}@C_{60}$ and $^{31}\text{P}@C_{60}$ have been measured [25], and are given in Table I.

From the above, to a first approximation, the full spin Hamiltonian for the pair \mathcal{AB} ($\mathcal{A}=^{15}\text{N}@C_{60}$, $\mathcal{B}=^{31}\text{P}@C_{60}$) can be given as

$$\begin{aligned} \hat{H}/h = & g_e\mu_e B_z \hat{S}_z^A - g_N^A \mu_N B_z \hat{I}_z^A + A^A \hat{S}_z^A \hat{I}_z^A + g_e\mu_e B_z \hat{S}_z^B \\ & - g_N^B \mu_N B_z \hat{I}_z^B + A^B \hat{S}_z^B \hat{I}_z^B + J_D \hat{S}_z^A \hat{S}_z^B, \end{aligned} \quad (1)$$

where $I(S)$ labels nuclear (electronic) spin, and we have made the secular approximation, dropping terms that do not commute with the electronic Zeeman Hamiltonian since $\omega_{\text{Larmor}}^S \equiv g_e\mu_e B_z \gg (J_D, \omega_{\text{Larmor}}^I \equiv g_N\mu_N B_z, A)$. The individual ESR and NMR spectra of these molecules are simulated in Fig. 1 and have been measured in [11].

III. QUANTUM CELLULAR AUTOMATA

Typical quantum computer architectures assume the capability of locally addressing every qubit. Such a requirement is highly challenging to engineer, and the achievement of this level of individual quantum control is a major goal in almost

every current implementation. The use of such local control methods forces an interaction of the quantum information with very many ‘‘classical’’ control gates, each possibly providing a decohering effect on the quantum information. It is also important that the effects of such control gates on the computer’s Hamiltonian remain as ‘‘classical’’ as possible, in that they effect a change in the parameters appearing in the system Hamiltonian and do not themselves become entangled with the quantum computer. This criterion of classical gating becomes nontrivial as the length scale of the quantum computer architecture is reduced to nanometers.

An alternative architecture is to utilize a small number of identifiable spins, placed in a regular spatial pattern, and to manipulate the quantum information encoded in this spin chain via global addressing techniques. This type of globally

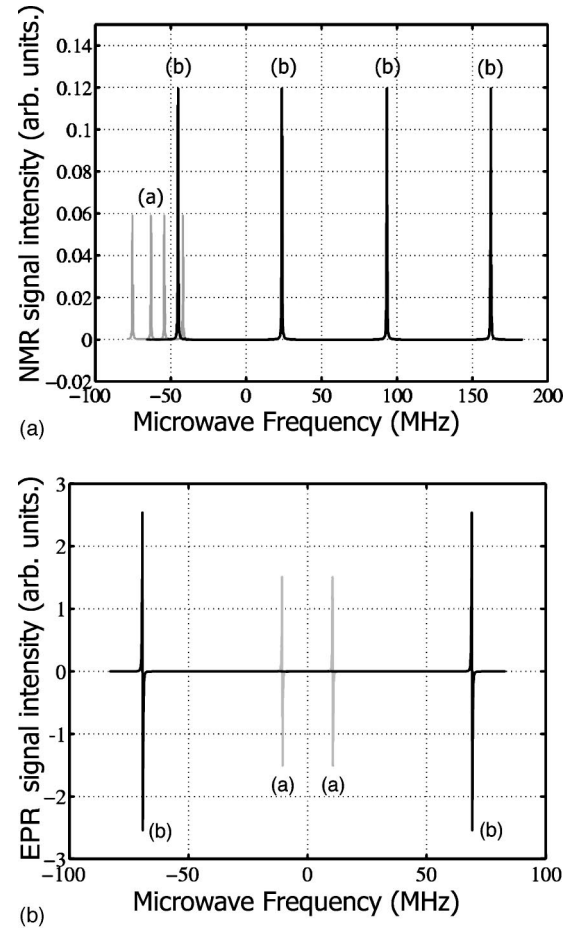


FIG. 1. Simulated NMR (a) and ESR (b) spectra of $\mathcal{A}=^{15}\text{N}@C_{60}$ and $\mathcal{B}=^{31}\text{P}@C_{60}$ with (a) and (b) labeling the spectral lines from \mathcal{A} and \mathcal{B} , respectively.

addressed architecture was invented by Lloyd in 1993 [26], and utilized three types of addressable spin arranged in the linear pattern $ABCABCABC$, where each spin site encodes one logical qubit. In fact, this cellular automata design was one of the very earliest quantum computer architectures proposed in the literature. Lloyd showed that such a quantum computer architecture was universal. Benjamin [27] found a similar architecture which used only two types of identifiable spin species arranged in the alternating linear pattern $ABABAB$. This reduction in spin resources came with an increase in logical encoding: a logical qubit is now encoded into four spin sites with a buffer space of at least four empty spin sites between each logical qubit.

The operation of Benjamin's architecture centers on the ability to implement the global unitary operator $\hat{\mathcal{A}}_f^U$. Denoting the spin up (down) state as 1 (0), $\hat{\mathcal{A}}_f^U$ is the conditional application of the unitary U to the \mathcal{A} spins in the alternating spin chain $ABABAB$, depending on the state of \mathcal{A} 's neighboring \mathcal{B} spins. In particular, letting f be the sum of the states of the neighboring \mathcal{B} spins, we have $f \in [0,1,2]$. Thus, for example, $\hat{\mathcal{A}}_1^U$ is the conditioned application of U to all \mathcal{A} spins in the alternating chain which have neighboring \mathcal{B} spins that are different from each other, i.e., $f=1$. One has a similar definition for $\hat{\mathcal{B}}_f^U$, for the application of U on all \mathcal{B} spins conditioned on their neighboring \mathcal{A} spins. The case when $U \sim G_{\text{NOT}}$ (a one-qubit NOT operator) in $\hat{\mathcal{A}}_f^U$ (or $\hat{\mathcal{B}}_f^U$), occurs quite frequently in Benjamin's architecture and is shortened to $\hat{\mathcal{A}}_f$ (or $\hat{\mathcal{B}}_f$).

Benjamin shows in [27] that through a judicious sequence of applications of $\hat{\mathcal{A}}_f^U$ and $\hat{\mathcal{B}}_f^U$ one can implement single-qubit operations and two-qubit (controlled-NOT) (CNOT) operations, and thus the architecture is universal. In particular, to move qubits through the spin chain one applies an alternating pulse sequence of $\hat{\mathcal{A}}_1$ followed by $\hat{\mathcal{B}}_1$, while the generation of a control U between two neighboring logical qubits requires ~ 30 global pulses [28]. One can translate all the standard circuit-based quantum algorithms to run with this quantum-cellular-automata architecture [28]. This programming architecture, although somewhat expensive in terms of spatial and temporal resources, is ideal for use in systems where the individual control of qubits is difficult, and it could provide an interim test bed for various implementations while the more long term goal of local gating is developed.

Since Lloyd's and Benjamin's pioneering work little has appeared in the literature with regard to quantum-cellular-automata designs for quantum computers [29]. In this paper we will consider a quantum-cellular-automata architecture consisting of an alternating linear array of group-V endohedrals, $ABABAB$, with the AB Hamiltonian given by Eq. (1) (schematically depicted in Fig. 2). Essentially this task reduces to developing a "quantum algorithm" for generating Benjamin's global operation $\hat{\mathcal{A}}_f^U$ (similarly for $\hat{\mathcal{B}}_f^U$). We first investigate the possibility of implementing $\hat{\mathcal{A}}_f^U$ using the S (electron) spins alone.

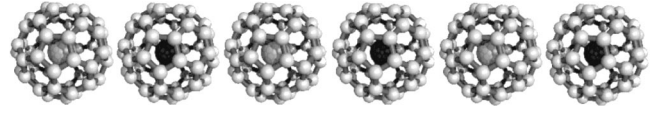


FIG. 2. Schematic depiction of the $ABABAB$ quantum-cellular-automata chain using group-V endohedral fullerenes.

IV. THE GLOBAL OPERATION

In this section we consider an alternating spin 1/2 chain of electronic spins $S^A S^B S^A S^B$. We begin by considering the $S^A S^B$ unit alone where \mathcal{A} spin has only one neighboring \mathcal{B} spin. We assume that we know how to perform the controlled operation of applying U to S^A , with S^B controlling, an operation we denote by $C(\mathcal{B}, \mathcal{A}; U)$. We shall describe below the details regarding the sequence of pulses required to execute $C(\mathcal{B}, \mathcal{A}; U)$. We next consider the case when \mathcal{A} has two neighbors, e.g., $S^B S^A S^B$. Remembering that one cannot separately address an individual neighboring \mathcal{B} spin, one can show that instead of performing $C(\mathcal{B}, \mathcal{A}; U)$ the above mentioned sequence of pulses now performs

$$\Xi(\mathcal{B}, \mathcal{A}; U) \equiv C(\mathcal{B}_{\text{on the left}}, \mathcal{A}; U) C(\mathcal{B}_{\text{on the right}}, \mathcal{A}; U), \quad (2)$$

where Eq. (2) defines the function Ξ . One can observe that the case when the desired global operation is $\hat{\mathcal{A}}_1 \equiv \hat{\mathcal{A}}_1^{\text{NOT}}$, we have $\hat{\mathcal{A}}_1 = \Xi(\mathcal{B}, \mathcal{A}; G_{\text{NOT}})$.

We now indicate the procedure for implementing $\hat{\mathcal{A}}_2^U$, when the desired $U \sim \exp(i\theta S_x^A)$. For ease of notation we define the standard NMR and ESR symbol for $\pi/2$ pulses [30–32]

$$[+S_y^B] \equiv \exp\left(i\frac{\pi}{2} S_y^B\right) \quad (3)$$

and the more general rotation through the angle γ about the z axis, for example, by the symbol

$$\{Z_\gamma^A\} \equiv \exp(i\gamma S_z^A). \quad (4)$$

We find that we can execute $\hat{\mathcal{A}}_2^U$ when $U \sim \exp(i\theta S_x^A)$ through the following pulse sequence (read right to left):

$$\begin{aligned} \hat{\mathcal{A}}_2^U = & \Xi(\mathcal{B}, \mathcal{A}; \{X_\delta^A\}) \{X_{\pi/2-\delta}^A\} \{Z_{-\pi/2}^A\} \\ & \times \Xi(\mathcal{B}, \mathcal{A}; \{X_\pi^A\}) \{Z_{-\pi/2}^A\} \{X_{\pi/2-\delta}^A\} \Xi(\mathcal{B}, \mathcal{A}; \{X_\delta^A\}), \end{aligned} \quad (5)$$

where $2\delta = \theta$. To implement $\hat{\mathcal{A}}_1^U$ one sets $\delta = \theta$ and replaces the leftmost term in Eq. (5), $\Xi(\mathcal{B}, \mathcal{A}; \{X_\delta^A\})$, by $\{X_\pi^A\}$, while to implement $\hat{\mathcal{A}}_0^U$ one applies $\{X_\pi^B\}$ to all the \mathcal{B} spins, then $\hat{\mathcal{A}}_2^U$, and finally flips the \mathcal{B} spins back with $\{X_{-\pi}^B\}$. The above pulse sequence (5) can be easily modified to suit other forms of U in addition to $\exp(i\theta S_x^A)$; however, we show below that the most useful case is when $\theta = \pi$ and $U = \exp(i\pi S_x^A) \sim G_{\text{NOT}}$ on the \mathcal{A} spin.

V. ALL ELECTRONIC QUANTUM-CELLULAR-AUTOMATA QUANTUM COMPUTER

From Fig. 1, we can separately address the electronic spins S^A and S^B with a multifrequency selective soft pulse of length greater than 20 ns. If we first consider both S^A and S^B to be spin 1/2, then one can use standard pulse encodings of the CNOT [31,32],

$$C(\mathcal{A}, \mathcal{B}, G_{\text{NOT}}) = [-S_y^B][\mp S_z^A \mp S_z^B][\pm 2S_z^A S_z^B][+S_y^B]e^{\pm i\pi/4}, \quad (6)$$

$$C(\mathcal{B}, \mathcal{A}; G_{\text{NOT}}) = [-S_y^A][\mp S_z^A \mp S_z^B][\pm 2S_z^A S_z^B] \times [+S_y^A]e^{\pm i\pi/4}, \quad (7)$$

to generate the operations \hat{A}_1 and \hat{B}_1 of the previous section. To perform universal quantum computation one must be at least capable of implementing a two-qubit CNOT and a minimal set of one-qubit operations such as the phase gate, the $\pi/8$ gate, and the Hadamard gate. In the quantum-cellular-automata architecture of Benjamin this means that one must at least be able to implement the global operation \hat{A}_f^U , where U is taken from this minimal set of one-qubit gates, and the case where $U \sim G_{\text{NOT}}$. NMR and ESR pulses effectively generate terms in the Hamiltonian that are linear in the spin operators $I_{x,y,z}$ and $S_{x,y,z}$. In particular, the spin operators when commuted close to form the Pauli algebra. In the case of spin 1/2, the Pauli algebra generates the group SU(2), or all possible spin-1/2 unitaries. Thus one can generate any desired spin-1/2 unitary through a sequence of pulses when acting on an isolated spin. Furthermore, in the case of the alternating chain $\mathcal{A}\mathcal{B}\mathcal{A}\mathcal{B}\mathcal{A}\mathcal{B}$, one can find suitable alterations of Eq. (7), when used in Eqs. (2) and (5), to generate any given unitary U in \hat{A}_f^U , and similarly for \hat{B}_f^U . Thus an all-electronic quantum-cellular-automata quantum computer architecture using a spin-1/2 $\mathcal{A}\mathcal{B}\mathcal{A}\mathcal{B}\mathcal{A}\mathcal{B}$ chain is possible.

The endohedrals $^{15}\text{N}@C_{60}$ and $^{31}\text{P}@C_{60}$, however, possess electron spin $S=3/2$ and the application of Benjamin's QCAQCA is not as straightforward. One possibility is to consider the four electronic levels of the electron spin with $S=3/2$ to constitute a qudit with $d=4$. Multilevel quantum computation has been proposed elsewhere [33], but to extend Benjamin's formulation of the spin-1/2 QCAQCA to operate with qudits is far beyond the scope of this article. Indeed, as shown below, there are reasons to believe that such an extension may not be possible within the confines of NMR and ESR.

Remaining with qubits, we instead propose to encode a qubit into the four levels of a spin 3/2 in two ways: an "inner qubit" $\{|m_s = \pm 1/2\rangle\}$ and an "outer qubit" $\{|m_s = \pm 3/2\rangle\}$. By a small alteration of Eq. (7), one can obtain

$$C_{\text{inner}}(S^A, S^B; G_{\text{NOT}}) = C_{\text{outer}}(\overline{S^A}, S^B; G_{\text{NOT}}) = [S_y^B][-S_z^A - S_z^B][+2S_z^A S_z^B] \times [-S_y^B]e^{+i\pi/4}, \quad (8)$$

$$C_{\text{inner}}(S^B, S^A; G_{\text{NOT}}) = C_{\text{outer}}(\overline{S^B}, S^A; G_{\text{NOT}}) = [S_y^A][-S_z^A - S_z^B][+2S_z^A S_z^B] \times [-S_y^A]e^{\pm i\pi/4}, \quad (9)$$

where C_{inner} is a CNOT on the Hilbert space spanned by the inner qubits, and similarly for C_{outer} , $\overline{S^A} = G_{\text{NOT}}(S^A)$, and we have used the spin-3/2 representations of the SU(2) group. Armed with this one can easily construct the Benjamin global operations \hat{A}_1 and \hat{B}_1 on the inner qubit subspace. One can further construct (see Sec. VI C) the global operations \hat{A}_f^{NOT} (and similarly \hat{B}_f^{NOT}) for $f=0,1$ and $f=2$, the operations most frequently used in quantum-cellular-automata quantum computation.

However, one finds that one cannot generate all the one-qubit gates required for universal quantum computation. As above, ESR pulses essentially generate terms in the Hamiltonian that are linear in the spin operators and again these terms commute to form a Pauli algebra. Evolution under such terms leads to dynamics that resides in the group generated by this Pauli algebra [or SU(2)]. However, since now we have $S=3/2$, this SU(2) is merely a subgroup of the full group of unitaries that can act on a spin 3/2, but as long as the system Hamiltonian is linear in the spin operators we need only consider the dynamics within the SU(2) subgroup. Normally, a unitary operation $U \in \text{SU}(2) \subset \text{SU}(4)$ will not factor into unitary operations on the inner and outer qubit subspaces. However, if U is diagonal in the $|m_s\rangle$ basis it can so factor, and with this one can find pulse sequences to implement \hat{A}_f^U where U is a phase gate and the $\pi/8$ gate, but crucially not the Hadamard gate. Thus an all-electronic QCAQCA with a system Hamiltonian that is linear in the spin operators and where $S>1/2$ seems not possible.

One way of implementing SU(2) operations on the qubit subspaces and one which is well known in NMR and ESR is to introduce terms in the system Hamiltonian that are not linear in the spin operators. Such terms lie in the full spin-3/2 algebra and the generated unitaries no longer lie in the SU(2) subgroup but in the much larger SU(4) group. One effect of such nonlinear terms, for example zero-field-splitting terms $H_{\text{ZFS}} \propto S_z^2$, can be to lift the degeneracy of the microwave transition frequencies between $|m_s\rangle \leftrightarrow |m_s+1\rangle$. This allows one to frequency address the transition $|m_s = -1/2\rangle \leftrightarrow |m_s = +1/2\rangle$, or the inner qubit subspace, and directly implement SU(2) operations on this subspace. This allows the generation of arbitrary one-qubit unitaries U [34]. However, this solution frustrates the coherence transfer portion $[+2S_z^A S_z^B]$ of the pulse sequences (8) and (9), and thus the construction of the global QCAQCA operator \hat{A}_f^U . This can be seen by realizing that to obtain a $[+2S_z^A S_z^B]$ pulse one must effectively cancel out all the terms in the system Hamiltonian but the term $S_z^A S_z^B$. As is standard in NMR and ESR, one does this cancellation through "average Hamiltonian" theory [35], via the application of various SU(2) unitaries by rf or microwave (MW) pulses. As long as the system evolution remains within the SU(2) subspace such an averaging out can be achieved. However, when terms non-

linear in the spin operators are introduced into the system Hamiltonian, no application of SU(2) unitaries can, in general, average out the resulting “nonlinear” evolution (i.e., in the average Hamiltonian theory nonlinear terms in the Hamiltonian will almost always average to other nonlinear terms), and thus the generation of $[+2S_z^A S_z^B]$, and from this \hat{A}_f^U , is not possible.

Thus we have found that when operating on an $ABABAB$, spin-3/2 chain with ESR pulses one cannot generate all of the necessary global operations required for universal quantum computation. We next find that the combination of nuclear and electronic spins within the chain can improve matters.

VI. NUCLEAR-ELECTRON QUANTUM CELLULAR AUTOMATA

$^{15}\text{N}@C_{60}$ and $^{31}\text{P}@C_{60}$ both have nuclear spin 1/2. In this section we assume that the quantum information is stored in the nuclear spins of the dopant atoms in the $ABABAB$ chain. This has the significant advantage that the nuclear spin relaxation times are typically longer than the electronic spins by several orders of magnitude. As the nuclei are coupled only via the hyperfine interaction to the electrons, one can, via a suitable rf and MW pulse sequence, cancel out this hyperfine interaction with high precision. This allows the nuclei to act as a quantum memory and store the quantum information for very long periods of time in between processing.

To execute the Benjamin global operation \hat{A}_f^U , we essentially use the inner electronic qubits as a local “bus” for the quantum information stored in the nuclei. The procedure can be thought of as a quantum algorithm for the generation of the global operation and consists of several steps which are pictorially represented in Fig. 3.

The algorithm starts with the inner electronic qubit bus in the ground state ($|m_s\rangle = |-1/2\rangle$ for all molecules), and this bus is returned to the ground state after implementing the global operation \hat{A}_f^U . (a) We first assume that we have an arbitrary pattern of quantum information written onto the nuclear spins of the A and B molecules with the electrons set as above. (b) We then swap the quantum information of all the B molecules from their nuclei to their inner electronic qubits. (c) We then tag those A molecules that will receive the unitary operation U in \hat{A}_f^U by flipping the state of the electronic qubit of each A molecule conditioned on the state of its neighboring B electronic qubits. (d) We then undo (b) by swapping the quantum information back into the B nuclei from their electrons.

At this point all the inner electronic qubits are back in their ground states except for the “tagged” A molecules. (e) We then perform a controlled- U operation on the nuclear qubits of *all the molecules* using their inner electronic qubits as the control. (f) Finally we undo the operations (d), (c), and (b). The system is then ready for the execution of the next global operation.

One restriction we make in the following is that we rule out the use of selective pulses on the nuclear spins of the

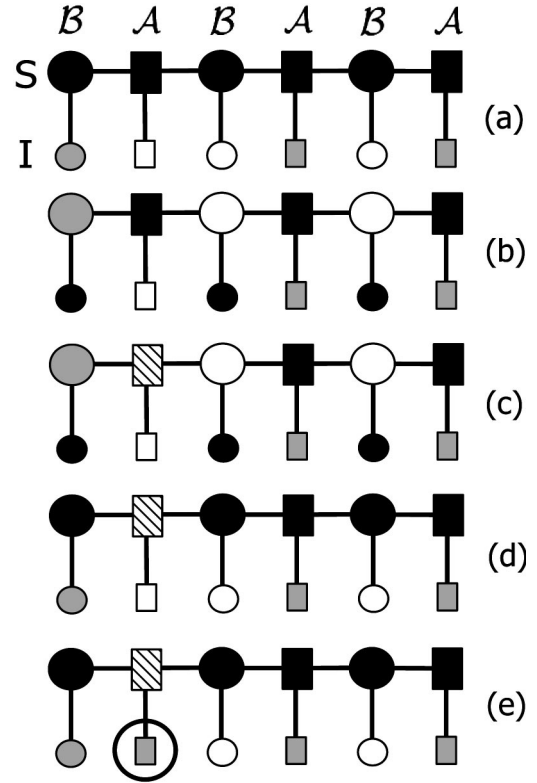


FIG. 3. Schematic representation of the steps (a)–(e) described in Sec. VI to execute the global operation \hat{A}_f^U . The alternating molecular chain $BABABAB$ is depicted with the inner electronic or S qubits (larger shapes) connected together via the magnetic dipole interaction, while the nuclear or I qubits (smaller shapes) are connected to the electronic qubits via the hyperfine interaction. (a) The S qubits in their ground states with some pattern of I qubits; (b) a hyperfine SWAP of the quantum information in the B molecules; (c) conditional flipping of the A electronic qubit depending on the neighboring B electronic qubits; (d) hyperfine SWAP of the B qubits back into the nuclei; (e) controlled U applied to the nuclei of all molecules conditioned by the state of their electronic qubits. Here the effect of \hat{A}_f^U is to flip the state of the leftmost A nuclear qubit.

nitrogen and phosphorus. From Fig. 1, to execute such selective pulses would require rf pulses of very long duration to frequency differentiate between the NMR transitions of the nitrogen and phosphorus. Such slow nuclear pulse sequences would lead to very long gate times. We will therefore not allow ourselves to use such selective nuclear pulse sequences and instead make use of fast “hard” nuclear pulse sequences [36]. We do allow selective addressing of the electronic transitions of the nitrogen and phosphorus as these pulse sequences are much shorter in duration. With this restriction, however, we are still able to design all the pulse sequences that are required to carry out the steps (b)–(e) above. The resulting combined total pulse sequence is somewhat lengthy but could be substantially shortened by applying the principles of optimal control theory [37].

We now expand on the above steps (a)–(e), in detail. In Sec. VIA below we describe the initialization of the spin chain. In Sec. VIB we design the SWAP pulse sequence required for step (b). In Sec. VIC we design the electronic

pulse sequence to implement the conditional operation required in step (c), and finally in Sec. VID we outline the execution of the controlled U needed in step (d). In Sec. VIE we discuss the undoing of the previous steps (b)–(d), while in Sec. VIF we provide some estimates of the pulse durations and logical QCA gate times.

A. Initialization

The QCA architecture may be implemented on an ensemble (type-II) or single-issue (type-I) quantum computer. The achievement of high nuclear polarizations has proved to be a major obstacle in NMR quantum computation. It remains a source of difficulty here as well. Polarizing the electrons is far easier, and one can achieve a difference in the ground to excited electronic populations of $\epsilon \sim 0.999$, at a temperature of 1 K and $B \sim 10$ T. Even when multiple spins are tensored to produce a pseudopure state, with such high individual polarizations one can still achieve a pseudopure state with a purity on the order of $\epsilon \sim 0.998$, with 1000 spins at these temperatures and magnetic fields. This polarization can be transferred to the nuclei via an INEPT pulse sequence [38]. To repump the electronic polarization and achieve a subset of completely spin (nuclear and electron) polarized molecules one could consider two chains of equal length, an $ABAB \dots$ chain and a $CD CD \dots$ chain, where all A , B , C , and D are globally addressable. Since at 1 K and $B = 10$ T, half of the spins (the electrons), are completely polarized, one can consider an $ABAB \dots ABAB \mathcal{X} CD CD \dots CD CD$ superchain where \mathcal{X} has a different resonant frequency yet again. One can then use a spin cooling quantum algorithm [39] to efficiently spin cool *all* the nuclear and electronic spins on one side of \mathcal{X} (i.e., \mathcal{X} acts as a spin gate). One can then switch off the interaction between the two subchains at \mathcal{X} and use the spin polarized half chain to perform the quantum computation. Such a design would be suitable for a type-II (ensemble) quantum computer. Alternatively, in the case of a type-I quantum computer one can replace the $CD CD$ chain with an $ABAB$ chain and place the readouts on the polarized half chain. More practically, any type-I quantum computer will require a single-spin readout of the electronic spins of the endohedral atoms. Such a readout may also serve to initialize the electronic qubits. We discuss possibilities for a single-spin readout further below.

B. Nuclear-electronic SWAP

A crucial ingredient in the above mentioned quantum algorithm to generate $\hat{\mathcal{A}}_f^U$ is the hyperfine SWAP. This operation performs a logical swap between a qubit stored in the nuclear spin and a qubit stored in the electron spin. Below we find two types of hyperfine SWAP, one that performs the swap using the inner electronic qubit and one that uses the outer electronic qubit. For the most part we use the former type of swap but we find later that the latter swap, which incorporates the outer electronic qubit, may be of significant use in the problem of qubit readout, and we discuss this below.

To develop the pulse sequence for the hyperfine SWAP it is useful to break down the SWAP operation into three CNOT operations. With the notation $G_{\text{SWAP}}(I:S)$, we have

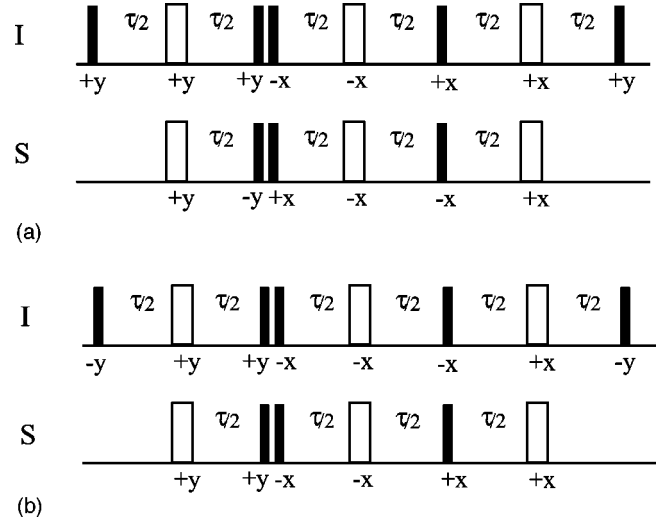


FIG. 4. Pulse sequence for a logical SWAP between spin $I = 1/2$ and the inner (a) and outer (b) qubits of spin $S = 3/2$ as described in Eqs. (12) and (13). The solid black bars are $\pi/2$ pulses while the white bars are refocusing π pulses.

$G_{\text{SWAP}}(I:S) = G_{\text{CNOT}}(I:S)G_{\text{CNOT}}(S:I)G_{\text{CNOT}}(I:S)$, where $G_{\text{CNOT}}(I:S)$ is the controlled NOT for I controlling S . In our case $I = 1/2$ while $S = 3/2$. Substituting the spin-3/2 representations for S into the standard spin-1/2 CNOT pulse sequence (7) fails to yield a proper CNOT operation. One must make reference instead to the two cases of a CNOT with respect to the inner and outer qubits of S . With this one can find

$$G_{\text{CNOT}_{\text{outer}}}(I:S) = [S_y][\mp S_z \mp I_z][\pm 2S_z I_z][S_y]e^{\pm i\pi/4},$$

$$G_{\text{CNOT}_{\text{outer}}}(S:I) = [I_y][\pm S_z \pm I_z][\pm 2S_z I_z][I_y]e^{\pm i\pi/4},$$
(10)

while

$$G_{\text{CNOT}_{\text{inner}}}(I:S) = [S_y][\mp S_z \mp I_z][\pm 2S_z I_z][S_y]e^{\pm i\pi/4},$$

$$G_{\text{CNOT}_{\text{inner}}}(S:I) = [-I_y][\mp S_z \mp I_z][\pm 2S_z I_z][I_y]e^{\pm i\pi/4}.$$
(11)

With these one can derive

$$G_{\text{SWAP}_{\text{inner}}}(I:S) = [I_y][+2I_z S_z][S_y - I_y][S_x - I_x][+2I_z S_z]$$

$$\times [I_x - S_x][+2I_z S_z][I_y]e^{-i\pi/4},$$
(12)

$$G_{\text{SWAP}_{\text{outer}}}(I:S) = [-I_y][+2I_z S_z][I_y - S_y][I_x - S_x]$$

$$\times [+2I_z S_z][S_x + I_x]$$

$$\times [+2I_z S_z][I_y]e^{+3i\pi/4},$$
(13)

and the related pulse sequences are graphically shown in Fig. 4. We note for later the curious case of Eq. (13), where we have in some sense “amplified” the magnetic signature of the qubit from a $\Delta m_I = \pm 1$ transition to a $\Delta m_S = \pm 3$ transition. In the following we will restrict ourselves to the inner electronic qubit subspace and thus Eq. (12).

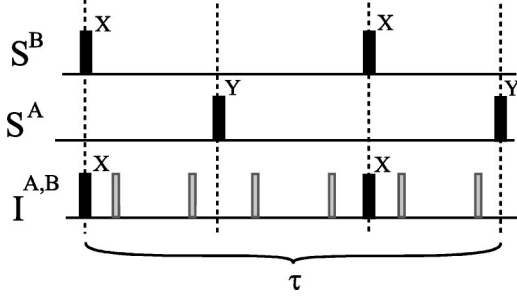


FIG. 5. Pulse sequence to selectively couple I^B with S^B while decoupling S^A from S^B , and simultaneously decoupling S^A from I^A . The black pulses are π pulses and the duration of the entire sequence is matched to yield $[+2I_z^B S_z^B]$ in Eq. (12). The gray pulses are fast hard X_π pulses, which are required to extend the duration of $[2I_z S_z]$ for the case when swapping occurs between the nuclear and electronic spins in $^{31}\text{P}@C_{60}$. By placing these pulses at the points $(m-1)/(4m)$ and $(3m+1)/(4m)$ in each third section as shown, we can extend the pulse sequence by a factor of m . Setting $m=6$ brings the $^{31}\text{P}@C_{60}$ SWAP to ~ 20 MHz, and thus long enough for the electronic spin selective pulses to operate on S^A and S^B .

Having found a SWAP operation between the nuclear and inner electronic qubits of either molecule, we now must show how one can adapt the sequence (12) to swap out only the B qubits while leaving the A nuclear qubits alone. We now consider the case of two coupled AB molecules with the Hamiltonian (1). The desired selective swap action can be achieved by replacing the $[+2I_z S_z]$ terms in Eq. (12) with the pulse sequence shown in Fig. 5. This new pulse sequence effectively averages out the hyperfine coupling interaction in the A molecules, thus turning off the action of the hyperfine CNOT and SWAP gates for these molecules. The hyperfine interaction for the B molecules is left intact by the pulse sequence but is reduced in magnitude, thus lengthening their hyperfine gate durations.

This particular pulse sequence has a number of advantages, the most important of which is that we do not selectively address the nuclear spins and thus can apply fast hard pulses to I^A and I^B simultaneously. We take advantage only of the selective addressing of the electronic spins of A and B as outlined at the beginning of Sec. V. Alternatively, one can use soft multifrequency nuclear pulses in the following to approximate the hard nuclear pulse. In more detail, from [40,41], the action of this pulse sequence is to remove to all orders (within the secular approximation made in Sec. II) all interactions between $S^A \leftrightarrow S^B$ and $S^A \leftrightarrow I^A$. This sequence also removes all the Zeeman terms in the AB Hamiltonian and one is thus left with $\hat{H}/h \sim A^B \hat{S}_z^B \hat{I}_z^B$. When inserted into Eq. (12), the effect is to swap the qubit that was stored in I^B into the inner electronic qubit of S^B . As mentioned above, the quantum information stored in I^A is not swapped into S^A but these spins do receive the local unitary transformation $[S_y^A][I_y^A]$. The local unitary $[S_y^A]$ is removed with a spin selective electronic pulse $[-S_y^A]$; however, we cannot remove the $[I_y^A]$ without performing a spin selective nuclear pulse, an operation we wish to avoid. We will see, however, that this extraneous local operation on I^A is not important for

the tagging of the A molecules, and furthermore the operation will be “undone” in step (d) (cf. Fig. 3).

C. Electronic tagging of f

Summarizing the previous steps: we have now swapped out the nuclear qubits of the B molecules in the $ABABABAB$ chain into their inner electronic qubits. Additionally, all the inner electronic qubits of the A molecules are in their ground states $|m_s\rangle = |-1/2\rangle$, i.e., we are at step (b) in Fig. 3. We will now electronically “tag” those A molecules targeted by the global operation \hat{A}_f^U . More precisely, we wish to flip the state of the A inner electronic qubit subject to the function f , with the control being A ’s nearest neighbor B ’s inner electronic qubits. We noted at the end of Sec. V that many intermolecular electronic unitaries cannot be achieved with $SU(2)$ pulses. However, the case of $f=1$ is simply the operation $\Xi(B,A;G_{\text{NOT}})$ (see Sec. IV), where one uses Eq. (8) for the CNOT in the inner electronic qubit subspace. The case $f=2$ is more difficult and one must make full use of the construction given in Eq. (5) to obtain the following pulse sequence:

$$\begin{aligned} & \Xi(B,A;\sqrt{G_{\text{NOT}}})\{Z_{-\pi/2}^A\}\Xi(B,A;\sqrt{G_{\text{NOT}}})\{Z_{-\pi/2}^A\} \\ & \times \Xi(B,A;\sqrt{G_{\text{NOT}}})e^{3i\pi/4}, \end{aligned} \quad (14)$$

where $\Xi(B,A;\sqrt{G_{\text{NOT}}}) \equiv C(S^B,S^A;\Sigma)^2$ [see Eq. (2)], and

$$\begin{aligned} & C(S^B,S^A;\Sigma) \\ & \equiv e^{i(1/2)\pi S_Y^A} e^{-i(1/4)\pi(S_Z^A+S_Z^B-2S_Z^A S_Z^B)} e^{-i(1/2)\pi S_Y^A} e^{i\pi/4}. \end{aligned} \quad (15)$$

With this pulse sequence one achieves the required spin flip of the A molecule’s inner electronic qubit up to a phase. This global phase is unimportant as it will be removed in step (e), when we undo the unitaries. Finally, the case $f=0$ is dealt with by first flipping all B inner electronic qubits, then using the $f=2$ operation above, and then flipping back. In the case of Fig. 3, we have chosen to implement the tag $f=1$ operation in step (c). Once we have set the electronic tag on the appropriate A molecules we “undo” the SWAP, returning B ’s quantum information back into its nuclei and B ’s inner electronic qubits to the ground state, arriving at step (d) in Fig. 3.

D. Implementing U

We can now implement *any* desired unitary on the flagged A qubits. The construction of this operation is well known [42], and consists of applying hard pulses [36] to *all* of the A and B and nuclei interspersed by CNOT gates between the inner electronic qubits and nuclear qubits of all the molecules. More precisely, the controlled execution of any given unitary operation on the nuclear spin I controlled by the inner electronic qubit S on all the molecules may be written as

$$C(S,I;U) = DC(I,S;X)EC(I,S;X)F, \quad (16)$$

$$U \equiv e^{i\alpha} R_z(\beta) R_y(\gamma) R_z(\delta) = e^{i\alpha} D X E X F. \quad (17)$$

In Eq. (16), β , γ , and δ are the Euler angles of the desired nuclear SU(2) operation, $C(I,S;X)$ is the hyperfine CNOT given in Eq. (11), the nuclear rotation operators are given by $R_\delta(\theta) \equiv \exp(-i\theta\delta/2)$, where $\delta=x,y,z$, and standard pulse sequences for these spin rotations can be found in [38]. Further, $D \equiv R_z(\beta)R_y(\gamma/2)$, $E = R_y(-\gamma/2)R_z[-(\delta+\beta)/2]$, $F = R_z[(\delta-\beta)/2]$, $X \sim G_{\text{NOT}}$, and α is a phase. Crucially, one can show that $DEF = I$. From this one can see that the action of the sequence (16) on the nuclear spin is unity when the inner electronic qubit is not set. When the inner electronic qubit is set, however, the action of Eq. (16) is to execute U from Eq. (17) on the nuclear spin up to the phase α . One then finally applies the local unitary $\exp[i\alpha(S_z^A + S_z^B)]$ to all molecules to yield the final phase factor α in Eq. (17). With this one has the very powerful capability of applying *any* desired single-qubit unitary to the nuclear qubits of the tagged \mathcal{A} molecules. However, the typical operation in the QCAQCA is $\hat{\mathcal{A}}_f$, and this can be executed more simply by a hyperfine CNOT between the electronic and nuclear qubits of all the molecules. In Fig. 3(e), we illustrate the application of $\hat{\mathcal{A}}_1$.

E. Undoing the unitaries

Following the arguments made above, we execute step (e), that is, once we have implemented the desired unitary on the \mathcal{A} nuclear qubits we “undo” steps (b), (c), and (d). We SWAP the nuclear qubit into the electrons on all \mathcal{B} molecules, undo the tagging of the \mathcal{A} molecules, and then SWAP the quantum information back into the nuclei of the \mathcal{B} molecules. The system is left with all the inner electronic qubits in their ground states while the nuclear qubits have received the global operation $\hat{\mathcal{A}}_f^U$.

F. Gate duration

As we noted above, the resulting pulse sequence is quite lengthy. However, it may be possible to compress many of the above operations. Also, all nuclear pulses are fast hard pulses of little duration [43], while the primary slowdown arises from the nuclear-electronic SWAP whose duration is limited by the value of the hyperfine coupling constant of $^{15}\text{N}@C_{60}$ (~ 20 MHz), and the separation of the spectral lines of the \mathcal{A} and \mathcal{B} molecules in the ESR spectrum of Fig. 1 (~ 50 MHz). The typical QCAQCA global operation $\hat{\mathcal{A}}_f$ entails a pulse sequence in which the term $[+a_z b_z]$, occurs 15 times, each with an average duration of 50 ns, thus roughly bringing the cycle time of the global operation down to 1 μs . As noted above, the simplest quantum logic gate in the QCAQCA scheme requires ~ 30 global operations. This finally brings the cycle time for logical gates in the resulting QCAQCA to $\sim 30 \mu\text{s}$.

We noted in Sec. II that one has $T_1 \sim 1$ s at $T = 7$ K, and $T_2 \sim 20 \mu\text{s}$ for concentrated samples of $^{15}\text{N}@C_{60}$ group-V endohedral material. As mentioned above, recent experiments have indicated a dependence on concentration for T_2 , and it is commonly expected in all current spin-based quantum computer implementations that T_2 will rise toward the value of T_1 in the limit of zero spin concentration. If T_2

~ 1 s then one can expect on average 10 000–30 000 logical operations within this dephasing time (figure of merit). Indeed, at temperatures lower than 7 K one might be able to achieve far greater figures of merit as the decoherence and dephasing times increase. A figure of merit of 10^4 compares very favorably with most alternative proposed implementations for quantum-information processors.

VII. READOUT

One of the most challenging aspects of any solid-state implementation is that of readout. We first discuss the possibilities for an ensemble readout for the QCAQCA and later a single-spin readout for a type-I implementation.

In the original scheme of Lloyd [26], the only spin sites that could be individually addressed were those at either end of the chain as these spins were frequency differentiated by having a single neighbor. This is also the situation here. However, if these two sites were the only readout sites on the chain then the architecture would not be scalable in the presence of noise. One ensemble readout possibility would be to attach an electrically isolated paramagnetic adduct to every \mathcal{B} molecule ($\text{Gd}@C_{82}$ may be a possibility). Such readout sites would typically possess short relaxation times and one would have to isolate such sites from the operation of the primary processor using ESR pulses. It should be possible within the cellular automata architecture to selectively read out the state of a single qubit, that is, to transfer the state of a given logical qubit into the readout site. This can be done by adapting the single-qubit unitary operator U pulse scheme in [27]. This pulse sequence is arranged so that a logical qubit (which is usually encoded in four spins $ABAB$) gets driven into the state of a single spin which is then subject to a unitary operation U through the application of the global operation (say $\hat{\mathcal{B}}_f^U$, when the qubit has been forced into the spin state of a \mathcal{B} molecule). The spin neighborhood of this target \mathcal{B} spin is arranged so that this global operation affects only the state of that spin alone with an appropriate choice for f . For readout one could instead apply an operation $\hat{\mathcal{B}}_f^{\text{SWAP}}$ which swaps out the state of the spin into the readout site. This would be read and then SWAPed back.

For a type-I quantum-information processor based on the $ABAB$ chain of $^{15}\text{N}@C_{60}$ and $^{31}\text{P}@C_{60}$, one must be capable of performing a single-electron-spin readout. We suggest a number of possible technologies that may be capable of performing such a readout below. Before mentioning these we return to Sec. VI B and Eq. (13). There we noticed that this pulse sequence was able to SWAP the qubit between the highest and lowest weight subspaces of the nuclear and electronic spins, $I = 1/2$ and $S = 3/2$, and in the process increase the detectable magnetic signature by a factor of ~ 1000 .

We will now further enquire whether there exist similar pulse sequences that could further SWAP out the quantum information now stored in the outer electronic qubit into a separate coupled electronic system of larger spin. This could be very advantageous, for example, for the purposes of coupling into the spin chain a nanomolecular magnet with a spin of ~ 10 – 30 as a potential readout site [44]. As the SWAP gate is built from CNOT gates, we can focus on the latter. Further,

the pulse sequences must consist only of higher spin representations of the Pauli group. There may be many such pulse sequences, and we here present one set of sequences. The ones given below perform a CNOT between the highest and lowest weight subspaces of spins I and S . We set $I = a/2$ and $S = b/2$, where $b \geq a$ and both a and b are odd integers. Letting $\alpha = [a/2]$ and $\beta = [b/2]$, where $[]$ is the integer part of the fraction, we can find CNOT pulses for the following four cases:

α even, β even:

$$C(I, S; G_{\text{NOT}}) = [-S_y][-S_z - I_z][+2S_z I_z][S_y]e^{+i\pi/4}, \quad (18)$$

$$C(S, I; G_{\text{NOT}}) = [-I_y][-S_z - I_z][+2S_z I_z][I_y]e^{+i\pi/4}, \quad (19)$$

α odd, β odd:

$$C(I, S; G_{\text{NOT}}) = [-S_y][+S_z + I_z][+2S_z I_z][S_y]e^{+i\pi/4}, \quad (20)$$

$$C(S, I; G_{\text{NOT}}) = [-I_y][+S_z + I_z][+2S_z I_z][+I_y]e^{+i\pi/4}, \quad (21)$$

α odd, β even:

$$C(I, S; G_{\text{NOT}}) = [S_y][+S_z + I_z][+2S_z I_z][-S_y]e^{+i\pi/4}, \quad (22)$$

$$C(S, I; G_{\text{NOT}}) = [I_y][-S_z - I_z][+2S_z I_z][-I_y]e^{+i\pi/4}, \quad (23)$$

α even, β odd:

$$C(I, S; G_{\text{NOT}}) = [S_y][-S_z - I_z][+2S_z I_z][-S_y]e^{+i\pi/4}, \quad (24)$$

$$C(S, I; G_{\text{NOT}}) = [I_y][+S_z + I_z][+2S_z I_z][-I_y]e^{+i\pi/4}. \quad (25)$$

A type-I quantum computer design requires the capability of single-qubit readout and this translates in our case to the capability of reading out the spin state of the endohedral electrons. One can classify the various readout methods into the general categories (i) force, (ii) electric, and (iii) optical measurement techniques. As was mentioned before, since the electronic wave function of the trapped atom is totally confined within the C_{60} cage, spin measurement techniques that involve the physical transport of this spin outside the molecule are not possible here. Some techniques, such as ODMR (optically detected magnetic resonance), have so far not proved possible with $^{15}\text{N}@C_{60}$ and $^{31}\text{P}@C_{60}$, and thus the addition of separate readout sites that can be dynamically coupled and decoupled from the primary processor via NMR or ESR pulses is warranted.

Possible techniques in the above mentioned categories include the following. (i) Magnetic resonance force microscopy (MRFM) [45]. (ii) Micro-SQUID (superconducting quantum interference device), a technology that is already capable of discriminating $\Delta m_S = 30$ [46]. (iib) Scanning-tunneling-microscope ESR, a technique that, though not well understood, has yielded single-molecule ESR spectroscopy

of iron impurities in silicon [47], and in single BDPA molecules on silicon [48]. (iic) A single-molecule endohedral fullerene single-electron transistor [49]. This is a very challenging technology which has yielded a single-electron tunneling current through a C_{60} molecule with an electromechanical coupling to the quantized motion of the entire molecule. Performing such an experiment with $^{15}\text{N}@C_{60}$ may yield endohedral spin information. (iiia) Magnetic coupling of the endohedral electronic spin to a solid-state paramagnetic optical dipole such as a nanocrystalline N-V center in diamond. The optical paramagnetic center can then be probed via optical shelving techniques [50]. (iiib) ODMR via a magnetic coupling to a paramagnetic endohedral adduct which is chemically bonded to the $^{15}\text{N}@C_{60}$ molecule. For use with ODMR the adduct endohedral should ideally possess an optical transition in the visible spectrum, such as $\text{Er}_x\text{Sc}_{3-x}\text{N}@C_{80}$ [51].

The techniques suggested above are, in some cases, already at the single-electron detection level (though not capable of detecting the spin orientation), while the uncoupling of the quantum information into a large-spin system may allow the techniques of micro-SQUIDs and MRFM as they presently stand to act as readouts of a single endohedral electronic spin.

VIII. CONCLUSION

The philosophy taken in this work with regard to the construction of a quantum-information processor is to follow as closely as possible the system Hamiltonian that nature provides. The globally addressed approach has very significant advantages over the local gating approach. Global addressing avoids the buildup of numerous decoherence pathways associated with the effects of the local gates on the processor. Global addressing also avoids the very significant problem of the scaling up of the external resources required to execute local gating, e.g., numerous metallic contacts or multifrequency MW and rf generators with increasing bandwidths. Although it may be true that for local gating the external resources required increase polynomially with the number of qubits in the processor, such an overhead may not be experimentally feasible as the size of the processor grows large.

In some sense the technical complexities involved in the construction and operation of local gating are transferred into the quantum-cellular-automata “software.” As shown above, the detailed operation of a quantum-cellular-automata quantum computer architecture does not follow the standard quantum circuit model. This should not be seen as a disadvantage, as all quantum circuit algorithms can be “compiled” to run on a QCAQCA with a polynomial overhead [27]. Indeed, the QCAQCA may be able to run programs that do not follow the quantum circuit model (see [52] for an example of a noncircuit quantum program in another architecture). Furthermore, as the fundamental quantum operations are implemented via the very well developed methods of NMR and ESR, the fidelity of software execution is heightened. The QCAQCA also has the very important property that it can execute quantum computations in parallel, a

characteristic necessary for the ultimate scalability of the architecture.

In this work we considered the endohedral fullerene material $^{15}\text{N}@C_{60}$ and $^{31}\text{P}@C_{60}$ and argued that the system Hamiltonians and interactions present are sufficient to implement universal quantum computation via a quantum-cellular-automata architecture. Following the DiVincenzo criteria, we argued as follows. (i) The group-V endohedral materials behave as essentially electrostatically isolated nanoscopic molecular neutral atom traps that can be physically manipulated with relative ease using STM techniques and perhaps may be self-assembled using fullerene supra-chemistry. (ii) The electronic decoherence times are perhaps the longest seen in any molecular system, of the order of seconds at temperatures of ~ 7 K, which is also an upper limit for the electronic dephasing time. The nuclear decoherence times have not yet been measured but they are expected to be several orders of magnitude longer than the electronic decoherence times. (iii) The complete polarization of both the nuclear and electronic computational spins is feasible as half of the spins (electrons) within the entire system are completely polarized at moderate conditions of temperature and magnetic field strength. There are efficient cooling schemes to shuffle the unpolarized spins away from the computational spins. (iv) Entanglement can be generated via the intermolecule magnetic dipole interaction, the strength of which has been measured to be ~ 50 MHz. Armed with the two species $^{15}\text{N}@C_{60}$ and $^{31}\text{P}@C_{60}$, we showed that one has sufficient control to implement a two-component quantum-cellular-automata

quantum computing architecture, storing the quantum information in the nuclear spins while using the electrons as a local bus. We further found that the expected “figure of merit” compares very well with other proposed solid-state quantum computer designs. We also discovered pulse sequences that SWAPed the quantum information between the highest and lowest weight spaces of two spins of different size, e.g., $S=1/2 \leftrightarrow S=11/2$. These sequences could prove useful for out-coupling the quantum information into a spin readout system with large spin.

DiVincenzo’s final criterion (v), an efficient single qubit readout, is perhaps the most difficult challenge for any solid-state-based quantum computer design. We have proposed various possible ensemble and single-issue (type-I), readout technologies, many of which are themselves the subjects of intense study. The ideas presented here combine the tremendous power of NMR and ESR science together with the very “clean,” and almost atomic, systems presented by the endohedrals $^{15}\text{N}@C_{60}$ and $^{31}\text{P}@C_{60}$. These, combined with supramolecular chemistry, may provide a very real possibility for physical implementation of a quantum-information processor.

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- [1] D.P. DiVincenzo, *Fortschr. Phys.* **48**, 771 (2000).
 [2] B.E. Kane, *Nature (London)* **393**, 133 (1998).
 [3] G.P. Berman, G.D. Doolen, and V.I. Tsifrinovich, *Superlattices Microstruct.* **27**, 89 (2000); X. Hu, R. de Souse, and S. Das, e-print cond-mat/0108339.
 [4] J. Tejada, E.M. Chudnovsky, E. del Barco, J.M. Hernandez, and T.P. Spiller, *Nanotechnology* **12**, 181 (2001).
 [5] J.A. Jones, *PhysChemComm* **11**, 11 (2001).
 [6] European 5th Framework IST FET Project Report, coordinated by J. Twamley, <http://planck.thphys.may.ie/QIPDDF>
 [7] W. Harneit, M. Waiblinger, K. Lips, C. Meyer, A. Weidinger, and J. Twamley, in *Proceedings of the First Conference on the Experimental Implementation of Quantum Computation (IQC'01)*, edited by R. Clarke (Rinton Press, Princeton, 2002); W. Harneit, *Phys. Rev. A* **65**, 032322 (2002).
 [8] D. Suter and K. Lim, *Phys. Rev. A* **65**, 052309 (2002).
 [9] J. Yezpez, *Int. J. Mod. Phys. C* **12**, 1273 (2001).
 [10] A. Weidinger, M. Waiblinger, B. Pietzak, and T.A. Murphy, *Appl. Phys. A: Mater. Sci. Process.* **66**, 287 (1998).
 [11] C. Knapp, N. Weiden, K. Kass, K.P. Dinse, B. Pietzak, M. Waiblinger, and A. Weidinger, *Mol. Phys.* **95**, 999 (1998).
 [12] J.C. Greer, *Chem. Phys. Lett.* **326**, 567 (2000).
 [13] N. Weiden, B. Goedde, H. Kass, K.P. Dinse, and M. Rohrer, *Phys. Rev. Lett.* **85**, 1544 (2000); K.P. Dinse, H. Kass, C. Knapp, and N. Weiden, *Carbon* **38**, 1635 (2000).
 [14] P.D. Godwin, S.D. Kenny, R. Smith, and J. Belbruno, *Surf. Sci.* **490**, 409 (2001).
 [15] A.L. Buchachenko, *J. Phys. Chem. B* **105**, 5839 (2001).
 [16] B. Pietzak, M. Waiblinger, T.A. Murphy, A. Weidinger, M. Hohne, E. Dietel, and A. Hirsch, *Chem. Phys. Lett.* **279**, 259 (1997); K. Lips, M. Waiblinger, B. Pietzak, and A. Weidinger, *Phys. Status Solidi A* **177**, 81 (2000).
 [17] P.H. Beton (private communication).
 [18] S. Knorr, A. Grupp, M. Mehring, M. Waiblinger, and A. Weidinger, in *Electronic Properties of Novel Materials—Molecular Nanostructures*, edited by H. Kuzmany, J. Fink, M. Mehring, and S. Roth, AIP Conf. Proc. No. 544 (AIP, Melville, NY, 2000), p. 191.
 [19] G. Feher, *Phys. Rev.* **114**, 1219 (1959); G. Feher and E.A. Gere, *ibid.* **114**, 1245 (1959); J.P. Gordon and K.D. Bowers, *Phys. Rev. Lett.* **1**, 368 (1958); M. Chiba and A. Hirai, *J. Phys. Soc. Jpn.* **33**, 730 (1972).
 [20] B.E. Kane, *Fortschr. Phys.* **48**, 1023 (2000).
 [21] To effectively achieve “vanishing spin density” one must remove, to some precision, all spin interactions involving the computational spin sites that are extraneous to the quantum computer architecture. The primary sources of such interactions are electron-electron dipole-dipole interactions and, to a much lesser extent (10^{-3}), electron-nuclear spin interactions.

- To achieve the fault tolerant limit it is not necessary to achieve perfect spin isolation, but rather one must eliminate these extraneous interactions to some high precision. To reduce the dominant effects of electron-electron dipole-dipole interactions, one must reduce the electronic paramagnetic spin density, while to reduce the lesser effects of electron-nuclear interactions one must utilize isotopically purified ^{12}C in the fullerene generation.
- [22] D. Loss and D.P. DiVincenzo, Phys. Rev. A **57**, 120 (1998); V. Privman, I.D. Vagner, and G. Kventsel, Phys. Lett. A **239**, 141 (1998); C.H.W. Barnes, J.M. Shilton, and A.M. Robinson, Phys. Rev. B **62**, 8410 (2000); J. Levy, Phys. Rev. A **64**, 052306 (2001); T.D. Ladd, J.R. Goldman, F. Yamaguchi, Y. Yamamoto, E. Abe, and K.M. Itoh, Phys. Rev. Lett. **89**, 017901 (2002); G. Burkard, D. Loss, and D.P. DiVincenzo, Phys. Rev. B **59**, 2070 (1999); X. Hu and S. Das Sarma, Phys. Rev. A **61**, 062301 (2000); **64**, 042312 (2001); J. Schliemann, D. Loss, and A.H. MacDonald, Phys. Rev. B **63**, 085311 (2001); M. Friesen, P. Rugheimer, D.E. Savage, M.G. Lagally, D.W. van der Weide, R. Joynt, and M.A. Eriksson, e-print cond-mat/0204035; e-print cond-mat/0208021.
- [23] T.D. Ladd *et al.*, Phys. Rev. Lett. **89**, 017901 (2002); F. Yamaguchi and Y. Yamamoto, Appl. Phys. A: Mater. Sci. Process. **68**, 1 (1999).
- [24] M. Waiblinger, B. Goedde, K. Lips, W. Harneit, P. Jakes, A. Weidinger, and K.-P. Dinse, in *Electronic Properties of Novel Materials—Molecular Nanostructures* [18], p. 195.
- [25] M. Waiblinger, Ph.D. thesis, Universität Konstanz, 2001.
- [26] S. Lloyd, Science (Washington, DC, U.S.) **261**, 1569 (1993).
- [27] S.C. Benjamin, Phys. Rev. A **61**, 020301(R) (2000).
- [28] T. Hovland, M.Sc. thesis, National University of Ireland, Maynooth, 2000, <http://planck.thphys.may.ie/jtwamley/jtwamley.htm>
- [29] S.C. Benjamin, Phys. Rev. Lett. **88**, 017904 (2002).
- [30] Note: in NMR and ESR, the conventional matrix representations for the Pauli matrices for a spin-1/2 particle are $S_z = \text{diag}(1, -1)/2$, etc. See A. Schweiger and G. Jesche, *Principles of Pulse Electron Paramagnetic Resonance* (Oxford University Press, Oxford, 2001), Appendix B.2.
- [31] N. Linden, H. Barjat, E. Kupče, and R. Freeman, Chem. Phys. Lett. **307**, 198 (1999).
- [32] T. Schulte-Herbruggen and O.W. Sorensen, Concepts Magn. Reson. **12**, 389 (2000).
- [33] J.L. Brylinski and R. Brylinski, e-print quant-ph/0108062.
- [34] A. Wokaun and R. Ernst, Chem. Phys. **67**, 389 (1977).
- [35] R.R. Ernst, G. Bodenhausen, and A. Wokaun, *Principles of Nuclear Magnetic Resonance in One and Two Dimensions* (Clarendon Press, Oxford, 1990).
- [36] Pulse sequences are said to be “hard” when the rf power is large enough to allow the desired pulses to be of very short duration. Such hard pulses are broadband and excite a range of frequencies [38].
- [37] N. Khaneja, S.J. Glaser, and R. Brockett, Phys. Rev. A **65**, 032301 (2002).
- [38] M.H. Levitt, *Spin Dynamics: Basics of Nuclear Magnetic Resonance* (Wiley, New York, 2001).
- [39] L.J. Schulman and U. Vazirani, in *Proceedings of 31st ACM Symposium on Theory of Computing* (Association for Computer Machinery, New York, 2001), p. 322; P.O. Boykin, T. Mor, V. Roychowdhury, F. Vatan, and R. Vrijen, e-print quant-ph/0106093.
- [40] M. Stollsteimer and G. Mahler, Phys. Rev. A **64**, 052301 (2001).
- [41] P. Wocjan, M. Röetteler, D. Janzing, and T. Beth, e-print quant-ph/0109088.
- [42] M.A. Nielsen and I.L. Chuang, *Quantum Computation and Quantum Information* (Cambridge University Press, London, 2001), Sec. 4.2.
- [43] It may be, however, that the attainment of hard nuclear pulses on the scale required is beyond current hardware capabilities. An alternative is to substitute a multifrequency soft pulse for the hard nuclear pulse. This will take slightly longer to implement depending on how “soft” the pulse is, but this may be improved with pulse shaping.
- [44] W. Wernsdorfer and R. Sessoli, Science (Washington, DC, U.S.) **284**, 133 (1999); A. Caneschi, D. Gatteschi, C. Sangregorio, R. Sessoli, L. Sorace, A. Cornia, M.A. Novak, C. Paulsen, and W. Wernsdorfer, J. Magn. Magn. Mater. **200**, 182 (1999).
- [45] G.P. Berman, G.D. Doolen, P.C. Hammel, and V.I. Tsifrinovich, Phys. Rev. B **61**, 14 694 (2000).
- [46] C.I. Pakes, P.W. Josephs-Franks, R.P. Reed, S.G. Corner, and M.S. Colclough, IEEE Trans. Instrum. Meas. **50**, 310 (2001).
- [47] Y. Manassen, I. Mukhopadhyay, and N.R. Rao, Phys. Rev. B **61**, 16 223 (2000); Y. Manassen, E. Terovanesyan, D. Shachal, and S. Richter, *ibid.* **48**, 4887 (1993).
- [48] C. Durkan and M.E. Welland, Appl. Phys. Lett. **80**, 458 (2002).
- [49] H. Park, J. Park, A.K.L. Lim, E.H. Anderson, A.P. Alivisatos, and P.L. McEuen, Nature (London) **407**, 57 (2000).
- [50] X.F. He, N.B. Manson, and P.T.H. Fisk, Phys. Rev. B **47**, 8816 (1993); F.T. Charnock and T.A. Kennedy, *ibid.* **64**, 041201 (2001); J. Wrachtrup, S.Y. Kilin, and A.P. Nizovtsev, Opt. Spectrosc. **91**, 429 (2001).
- [51] R.M. Macfarlane, G. Wittmann, P.H.M. van Loosdrecht, M. deVries, D.S. Bethune, S. Stevenson, and H.C. Dorn, Phys. Rev. Lett. **79**, 1397 (1997); M. Macfarlane, D.S. Dethune, S. Stevenson, and H.C. Dorn, Chem. Phys. Lett. **343**, 229 (2001).
- [52] R. Raussendorf, D.E. Browne, and H.J. Briegel, e-print quant-ph/0108118.