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Fullerene-based electron-spin quantum computer

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We propose an alternative concept for a scalable spin quantum computer that combines aspects of other proposals with the advantageous features of endohedral fullerenes. The key advantages are that electron spins instead of nuclear spins are used and that the manipulation of fullerene molecules is fairly easy. Qubits are set and read out via pulsed electron-spin resonance. Addressing is provided by local magnetic fields or field gradients (A gate). The qubit-qubit interaction is mediated by magnetic dipolar coupling and can be controlled via the direction of the magnetic field with respect to the distance vector of the qubits (J gate). Molecular as well as solid-state architectures are discussed.

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INTRODUCTION

The forebear of quantum-information processing, spin quantum computing, is presently at the cross roads from an experimental point of view: single-molecule NMR has proven to be very successful as a test bed for basic ideas [1] but is known to be ultimately limited in the number of possible qubits [2]. Proposals for scalable quantum computers based on nuclear spins embedded in a solid [3] are technically very challenging, and major realization problems have not been solved yet. Especially the interaction with spurious electronic charges in the solid will be a serious problem for the spin coherence.

Making use of a fullerene molecule as a container for the information-carrying spin system provides a bridge over the sketched technological gap: the fullerene is both a sheltering environment for the delicate spin system and a handle for the arrangement of spins, which is a key issue for solid-state spin quantum computing.

This work is part of the European network listed in Ref. [4]. Here, we explore the promise and limitations of using endohedral fullerenes N@C_{60} and P@C_{60} as quantum-information carriers.

A scheme of the proposed concept is shown in Fig. 1: The qubits are encoded in the electron spins of nitrogen or phosphorus atoms encapsulated in fullerenes. These recently discovered molecules [5,6] have exceptional properties for quantum computing, viz., long spin lifetimes and sharp resonances. Quantum operations and readouts are performed by electron-spin resonance (ESR). The coupling between qubits is mediated by the magnetic dipolar interaction of the encapsulated atoms. Addressing is performed by individual tuning of the qubit resonance frequency.

First, we briefly review the properties of endohedral group V fullerenes that are important for quantum computation. Second, we evaluate the general limits of ESR quantum computing with endohedral fullerenes including error sources. Third, we discuss the techniques to be developed for local spin addressing, coupling control, and readout for an ultimately scalable solid-state quantum computer. Finally, we describe a more short-term alternative: a purely molecular design for an ensemble-type quantum cellular automaton or ABC computer [7,8], i.e., without local control of the individual qubits.

A. Group-V endohedral fullerenes

Fullerenes are highly symmetric hollow molecules that can accommodate one or more atoms and even small molecules in their interior. For most of these endohedral fullerenes, a charge transfer of the enclosed atom to the fullerene cage occurs, resulting in a chemical bond and distorted overall structure (metallofullerenes [9]). Noble gases [10] and group-V atoms [5,6] (and recently [11], the molecule Sc_3N) have been found to reside in the center of the fullerene without any charge transfer to the cage. Among them, only the group-V atoms are paramagnetic due to their half-filled p orbitals ($^4S_0$ ground state) and are thus interesting for electron-spin quantum computation [12,13].

The paramagnetic atoms of group V can be implanted into fullerenes by simultaneous ion bombardment and fullerene evaporation onto a target. The resulting endohedrals are thermally [14] and chemically stable [15] at ambient conditions. In as-implanted samples only a small fraction of the fullerenes is actually filled, typically 1 in 10 000. It is possible to enrich and purify the fraction of filled molecules by high-pressure liquid chromatography (HPLC) [16].

Group-V endohedral fullerenes exhibit very sharp ESR lines even in the solid state (Fig. 2) where usually broad powder spectra occur. The absence of broadening is due to

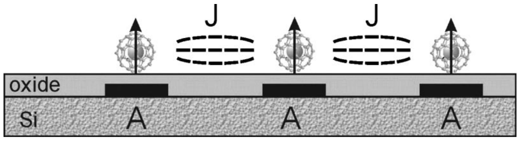

FIG. 1. Conceptual figure for a solid-state quantum computer using the electron spins (arrows) of endohedral fullerenes as qubits. Qubits are set and read out via ESR pulses. Addressing is provided by local magnetic fields or field gradients (A gate). The qubit-qubit interaction is mediated by magnetic dipolar coupling and can be controlled via the direction of the magnetic field with respect to the distance vector of the qubits (J gate).

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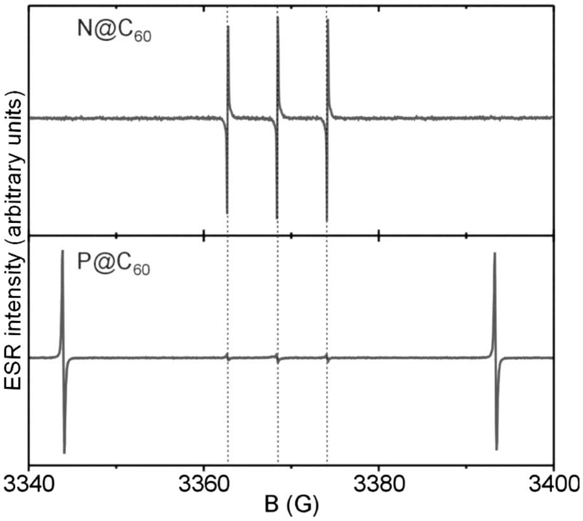

FIG. 2. Room-temperature ESR spectra of $^{14}N@C_{60}$ and $^{31}P@C_{60}$ embedded in a polycrystalline matrix of empty C_{60}, based on Ref. [17]. The group-V endohedral fullerenes show a hyperfine splitting according to the nuclear-spin multiplicity of the paramagnetic atom: ^{31}P has nuclear-spin $I=1/2$ leading to two hyperfine lines, the three lines of $^{14}N@C_{60}$ are due to $I=1$. In the $P@C_{60}$ spectrum, a small contamination of $N@C_{60}$ is visible.

the high symmetry of the C_{60} molecules, which eliminates all contributions of anisotropic interactions. So far, nitrogen and phosphorus in C_{60}, and nitrogen in C_{70} have been realized [17]; their properties are summarized in Table I. All ESR spectra show the typical hyperfine splitting according to the nuclear-spin multiplicity of the paramagnetic atom (two lines for ^{15}N and ^{31}P with nuclear spin $I=1/2$, and three lines for $^{14}N@C_{60}$ with $I=1$).

The electron spin of endohedral fullerenes has a remarkably long lifetime that makes it a useful embodiment of a qubit for quantum computation. The temperature-dependent spin-lattice relaxation time T_1 and spin-spin relaxation time T_2 of the electron spin have been measured [18–20] for magnetically dilute powder samples of $N@C_{60}$ and $P@C_{60}$. T_1 increases with decreasing temperature from about 100 μs at $T=300$ K to several seconds below $T=5$ K. T_2 is practically independent of temperature and about 20 μs in the present case. T_2 is probably foremost due to dipolar interaction with electron spins of other, randomly distributed, endohedral fullerenes. In a careful quantum computer design this interaction should be controlled and thus the effective coherence time could be much longer than 20 μs.

Experimental studies on the concentration-dependent line broadening [21] as well as theoretical calculations [22] show that the exchange interaction between two endohedrals, either directly by contact or mediated by a substrate is vanishingly small, thus leaving the magnetic-dipole interaction as the only strong coupling between two endohedrals fullerenes. For two adjacent endohedrals at a center-to-center distance of 1 nm (touching fullerene shells) the magnetic-dipole interaction can be as high as 50 MHz, being comparable to the hyperfine splitting of individual molecules.

B. Quantum computing with endohedral fullerenes

Due to the very special properties of nuclear spins, NMR quantum computation has proven very successful so far. Let us compare now some important issues arising when using electron spins instead of nuclear spins.

First, electron spins are sensitive to electromagnetic stray fields in their vicinity, leading, in general, to relaxation times much shorter than those of nuclear spins. On the other hand, using ESR instead of NMR leads to a higher calculation speed compensating to some extent the disadvantage of shorter lifetimes. But nevertheless decoherence is the major problem and in this respect the relatively long lifetimes of endohedral fullerenes are a major advantage of the present system.

TABLE I. Properties of group-V endohedral fullerenes [17–20] for quantum computation.

Endohedral fullerene	Thermal stability	Hyperfine constant	Relaxation in solution	Relaxation in solid C_{60}	Relaxation in solid C_{60}[a]
	T_S(K)	a_{iso} (G)	T_1 (μs) T_2 (μs)	T_1 (μs) T_2 (μs)	T_1 (μs) T_2 (μs)
$^{14}N@C_{60}$	500	5.665	120 50	120 20	$\gtrsim 10^6$ 20
$^{15}N@C_{60}$	500	7.944	Not measured	≈ 45 11	$\gtrsim 10^6$ 14
$^{31}P@C_{60}$	400	49.4	5 3	2.7 1.3	$\gtrsim 10^6$ 14
$^{14}N@C_{70}$	450	5.395	133 40	Not measured	Not measured
$^{15}N@C_{70}$	450	7.565	Not measured	Not measured	Not measured

[a] At $T=5$ K. All other values are given for room temperature.

Second, electron spins of different molecules often have a similar resonance frequency and thus show “spectral crowding” since the chemical shifts are small compared to the base frequency. Making use of the hyperfine splitting in combined electron-nuclear spin systems as those provided by endohedral fullerenes avoids this problem. Indeed, using only the C_{60} endohedrals, three very distinct electron-spin systems ($^{14}N@C_{60}$, $^{15}N@C_{60}$, and $^{31}P@C_{60}$) can be supplied.

Using electron spins for quantum computation instead of nuclear spins has two additional advantages: (i) the sensitivity of ESR is much higher than that of NMR due to the larger gyromagnetic ratio of electrons and (ii) thermal spin polarization is much larger for electrons. Both aspects combined give a very large improvement of the signal-to-noise ratio, an essential requirement for efficient quantum computation. In a solid-state implementation without single-spin readout capacity, one could use a sufficient number of computer copies for the readout.

A requirement for quantum computation using error-correction schemes is that “cold” qubits (i.e., qubits in a well-defined state) have to be supplied continuously while the computation takes place. With the present $S=3/2$ spin system, a recently proposed coherent repopulation scheme [23] could provide large spin polarization even at room temperature. In this scheme, two phase-shifted ESR pulses are applied simultaneously, which manipulate the spins in the sublevels differently. It is interesting that this method is applicable only for systems with a spin of at least $S=1$ and therefore can be used here, but not with the usual $S=1/2$ systems.

We now discuss quantum operations performed on a linear arrangement of endohedral fullerenes coupled by the dipolar spin-spin interaction. For clarity, we consider two spins only (which could be realized using dimers of endohedral fullerenes, see below) and assume that a large number of them is present in order to allow ESR detection. Single spin operations on electron spins are very straightforward to implement using standard ESR pulses. The pulse length is of the order of 10 ns so that many operations can be performed before decoherence sets in even if T_2 is only 20 μs (we expect T_2 to be longer in a carefully designed arrangement).

For high external fields B and dipolar interactions J small compared to the hyperfine interaction A ($B \gg A \gg J$), the effective additional Hamiltonian due to the coupling of two electron spins is

$$H_{eff}=J(r,\vartheta)S_{1z}S_{2z}, \tag{1}$$

where

$$J(r,\vartheta)=\frac{\hbar\gamma^2}{r^3}(1-3\cos^2\vartheta)=J_0(1-3\cos^2\vartheta) \tag{2}$$

giving $J\sim 50$ MHz for $r=1$ nm and $\vartheta=0°$. Here γ is the gyromagnetic ratio of the electron, r is the distance vector between the two electron spins, and ϑ is the angle between r and B.

The most straightforward way to implement a controlled-NOT (CNOT) operation would be to execute a pulse sequence such as those known from nuclear magnetic resonance (NMR) quantum computation [24], see Fig. 3. Known error sources for this scheme are (i) mismatched gating times, (ii) varying coupling strengths, and (iii) spin leakage to neighboring dimers.

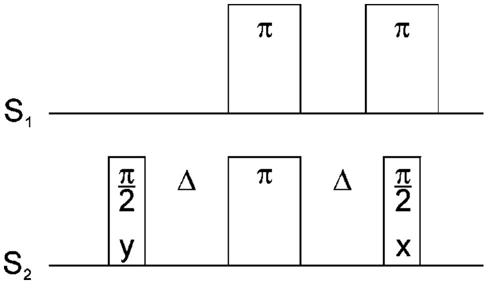

FIG. 3. ESR pulse sequence of a controlled-NOT operation [24] for two distinguishable spins S_1 and S_2. For $N@C_{60}$, the length of a selective π pulse is $t_\pi \sim 10$–100 ns. The time delay between two pulses is $\Delta=(2J)^{-1}\sim 10$ ns. Thus, a CNOT takes about 0.05 μs and about 400 of them can be performed before decoherence sets in for $T_2=20$ μs. If ultimately T_1 limits the coherence, 10^7 operations can be performed at low temperatures.

(i) The gating-time resolution in ESR is about 2 ns and limits the useful pulses on the short side to some 10 ns. Furthermore, tight field-strength control has to be exerted for the pulses.

(ii) The chemical realization of the spin separation leads to a negligible error, as opposed to mechanically arranged spins or qubits. A distribution $\delta\vartheta$ of dipolar angles, e.g., due to insufficient alignment in a liquid-crystal matrix (see below), leads to a variation in $\delta J/J_0 = 3\sin\vartheta\cos\vartheta\,\delta\vartheta$, which is vanishingly small for the “ON” state ($J=0$ or $\pi/2$) and larger for the “OFF” state ($\vartheta=54.7°\pm 2°, \delta J=\pm 0.05 J_0$). It is thus important not to make J_0 too large by adjusting the distance of the molecules.

(iii) Leakage to neighboring dimers scales with the inverse of the third power of the distance between them and can be made arbitrarily small.

C. Solid-state approach to quantum computing

Kane [3] proposed to use as qubits the well-isolated nuclear spins of phosphorus embedded in a silicon matrix, and the electron-mediated exchange coupling as the qubit-qubit interaction. The main difficulty lies in producing the well-defined geometrical arrangements of the embedded phosphorus atoms and in eliminating decoherence due to stray charges and spins. Therefore it would be tempting to use fullerene-shielded phosphorus atoms for this purpose. A straightforward replacement of the phosphorus atoms of the Kane concept by the present endohedral fullerenes is, however, not possible since the spin coupling has a different origin in the two cases, as described in Sec. A.

A solid-state architecture for an ESR quantum computer based on endohedral fullerenes therefore has to follow a different route (Fig. 1). The main difference from the Kane concept [3] lies in the addressing and readout of individual spins and in the coupling mechanism. Let us first examine the latter. Dipolar coupling between next neighbors can be used in ordered chains of fullerenes placed on a surface. We now discuss how such arrangements can be realized.

Fullerenes can be individually positioned on Si surfaces with a scanning-tunnel-microscope tip [25]. The problem for the group-V endohedrals is to get the fullerenes on the surface in a rational manner. The standard technique of evaporation at $T = 500–600\,°\mathrm{C}$ is not feasible due to insufficient thermal stability of the endohedral atom (see Table I). Another issue to be resolved for this direct nanomanipulation is how to identify individual endohedral fullerenes if more complex sequences involving different endohedral species are envisaged.

Cold thin-film-deposition techniques like Langmuir-Blodgett (LB) have been developed [26], usually requiring the use of chemically modified fullerenes for true monolayer formation. Making use of the chemical stability of the endohedral complexes, one can indeed provide such modified endohedral fullerenes. The basic LB technique can be combined with more advanced nanopatterning techniques like selective adhesion on hydrophilic and hydrophobic patches on a surface. Even scanning-probe-microscopy-based techniques like the atomic force microscope nanoplotter [27] could be employed.

For a true solid-state quantum computer, one would like to have local control of individual qubits (addressing or A gate) and their mutual interaction (coupling or J gate). A possible addressing gate could be built, in particular, by a variable local magnetic field or field gradient. Changing the local magnetic field of a qubit leads to a shift in resonance frequency that can then be selectively excited by the ESR microwave. Modern magnet technology provides large magnetic-field gradients useful for spin-frequency control. For a dysprosium micromagnet, the calculated realizable field gradient is $dB_z/dz \approx 14$ G/nm at $dz = 2$ μm from the sample [28]. Similarly, currents through nanometer-sized wires could provide the required local fields. The field gradient needed to distinguish two adjacent fullerene molecules (1 nm apart) would be about 6 G/nm for $^{15}\mathrm{N@C_{60}}$, corresponding to the size of the hyperfine splitting (see Table I).

Using the dipolar interaction offers obvious approaches for controlling the coupling strength (J gate). First of all, a spin chain can be oriented at a chosen angle to the external magnetic field (see Fig. 4) providing for a simple means to adjust the overall coupling strength. Micromechanical engineering could be used to control the dipole angle ϑ for individual qubits or frequency-labeled groups. However, decoherence induced by the controls might be a serious limitation of the application of nanomechanical actuators. Alternatively, one could reorient the magnetic field with custom pulse sequences. This "continuous-recoupling" scheme is well known from NMR quantum computation [2]. Combining such pulses with the local addressing could also provide a local control of the interaction.

D. Molecular approach to quantum computing

Solid-state quantum computing is still some way down the technologically feasible route, the most obvious obstacle being the absence of methods to read out a single spin. Until such methods are developed, much can be learned form taking measurements on large molecular ensembles.

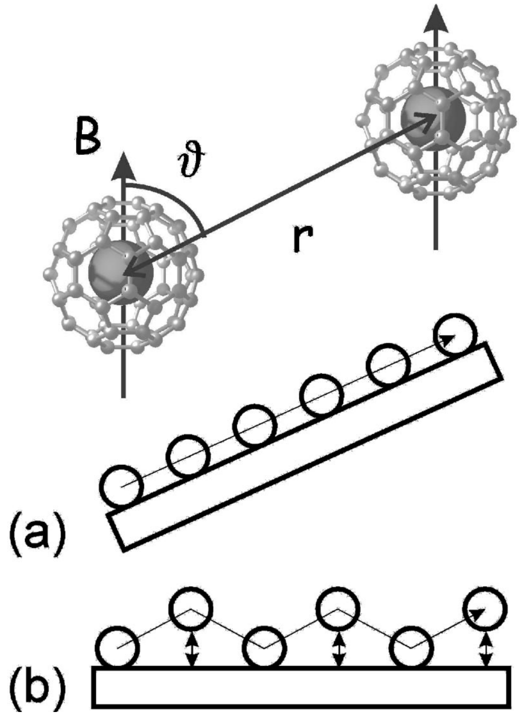

FIG. 4. Methods to control the dipolar coupling $J(\vartheta)$, see Eq. (2). (a) Global control of the dipolar angle ϑ by a tilt of the sample with respect to the magnetic field or vice versa. (b) Local control by a displacement of individual fullerenes along the magnetic-field direction.

We now discuss the possibility of designing molecular arrangements of well-isolated electron spins using endohedral fullerenes. Modern fullerene chemistry [29] allows a large variety of functionalized fullerenes. Recently, dimers of $\mathrm{C_{60}}$ with one filled moiety, $\mathrm{N@(C_{60})_2}$, have been produced demonstrating that the endohedral fullerenes are stable enough for this type of reaction [30]. Doubly filled dimer molecules and longer all-endohedral fullerene chains are obtainable using pure endohedral starting material. In such chains, all endohedral fullerenes will be of the same type.

There exists, however, a possibility to create chains of ordered sequences of differently filled fullerenes by using the key-keyhole principle. A possible route is depicted in Fig. 5. This example shows a minichain $\mathrm{N@C_{60}}$-XY-$\mathrm{P@C_{60}}$, where X and Y are specially designed molecular bridge groups [29]. One can first produce the adducts, $\mathrm{N@C_{60}}$-X and $\mathrm{P@C_{60}}$-Y, that cannot combine with themselves (taking, e.g., X: R-$\mathrm{NH_2}$, Y: R'-COOH). Then, one simply mixes the two substances to arrive at the chain molecule. Continuing this procedure, a quantum cellular automaton of the ABC type proposed by Lloyd [7] and Benjamin [8] can be realized. In this approach, no local control of the qubit-qubit interaction strength is needed. If it turns out that the flexibility of the molecules shown in Fig. 5 is a serious hindrance for the interaction control, stiffer bridge groups could be designed.

A very powerful tool to align these molecules with respect to one another and with respect to an external magnetic field

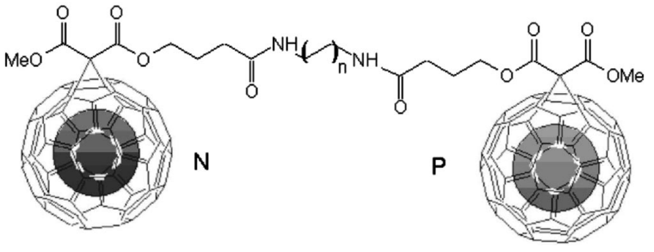

FIG. 5. Construction of a bimolecule from two different endohedrals by the key-keyhole principle. A realistic heteroendohedral dimer N@C_{60}-XY-P@C_{60} based on realizable addends [29] is shown. The addends X and Y can be produced using reactions for which the endohedral fullerenes are known to be stable [15]. Before the combining reaction, the X addend (R-NH_2) and the Y addend (R'-COOH) cannot combine with themselves.

is to embed them in a liquid crystal. Earlier experiments by us have shown that the fullerenes are soluble in liquid crystals and that they can be aligned by this procedure [31]. Thus, the realization of an electron-spin quantum computer with a small number of qubits is feasible with this concept.

SUMMARY

The well-isolated electron spins of group-V atoms encapsulated in fullerenes offer a very flexible possible implementation of quantum-information bits. Spin manipulation and readout can be done via standard ESR pulses, using the dipolar spin-spin interaction as a coupling mechanism. Both solid-state and molecular quantum-computer architectures using these building blocks have been presented.

Hundreds of operations should be possible using endohedral dimer molecules oriented in a liquid crystal as an elementary building block. More sophisticated molecular arrangements can be built using state-of-the-art fullerene chemistry. When spreading the molecules on a functional surface to produce a solid-state computer, all knowledge gained on these macromolecules can be directly transferred.

A preliminary technological road map has been sketched for a truly scalable solid-state quantum computer with local control of qubit states (addressing and readout) and qubit interaction. The most severe drawback at present is the lack of single-spin-detection methods, which are nevertheless being developed worldwide. Once developed, single-spin-readout procedures could be implemented in the present concept.

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