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The N@C$_{60}$ nuclear spin qubit: Bang-bang decoupling and ultrafast phase gates

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Nuclear spins have been proposed for the embodiment of quantum information and yielded the most complex demonstrations of quantum algorithms to date. However, the weak thermal polarisation of nuclear spins in experimentally accessible conditions has presented a barrier to further scaling. Here, we discuss the benefits of a coupled electron spin to the nuclear spin qubit and demonstrate the ideas using the ^{14}N nuclear spin in the N@C$_{60}$ molecule. In addition to providing a resource for nuclear spin polarisation and detection, the electron spin can be exploited to perform ultrafast nuclear spin phase gates, which can in turn be used to dynamically *bang-bang* decouple the nuclear spin from unwanted interactions.

1 Introduction

N@C$_{60}$ belongs to a unique class of hierarchical molecules where the trapped species (a nitrogen atom) retains much of its atomic character, benefitting from the isolation provided by the fullerene cage. Such properties have prompted proposals for the electron spin residing on the incarcerated nitrogen atom to be used to process quantum information [1, 2]. The electron spin of N@C$_{60}$ was preferred over the nuclear spin despite the fact that the most complex quantum information algorithms have been performed using nuclear spins [3]. While nuclear spins quantum bits (qubits) benefit from long relaxation times, they are plagued by the difficulty in obtaining a pure initial state, and single qubit operations are generally slow (on the 10–100 microsecond timescale). The Zeeman splitting of an electron spin is several orders of magnitude greater than that of the nuclear spin, permitting pure ground states in experimentally accessible conditions and much faster manipulation (on the tens of nanosecond timescale).

Here, we explore possibilities of using the ^{14}N nuclear spin of N@C$_{60}$ as a qubit. We note that the electron spin can act as a valuable resource for a nuclear spin qubit. First, the much greater spin polarisation of the electron can be transferred to the nuclear spin through the use of a selective microwave pulse. Second, the electron spin can be used to measure the state of the nuclear spin with greater sensitivity. Finally, the electron spin may be manipulated to perform phase gates on the nuclear spin which are orders of magnitude faster than conventional nuclear magnetic resonance (NMR) methods. Such ultrafast gates can be used to decouple the nuclear spin from unwanted interactions, further enhancing its robustness to relaxation. In this paper, we demonstrate these advantages using the coupled electron and nuclear spins in the N@C$_{60}$ molecule.

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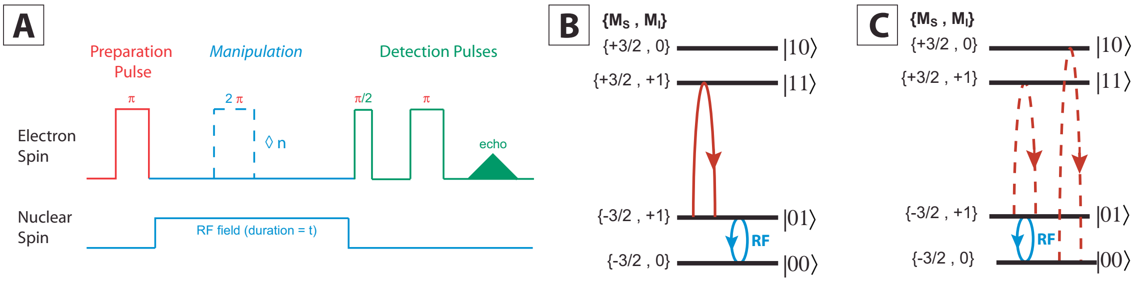

Fig. 1 (online colour at: www.pss-b.com) (A) General pulse sequence used: a microwave (preparation) pulse transfers electron spin polarisation to the nucleus, which is then manipulated using series of RF and microwave pulses. Finally, the electron spin is used to perform a measurement of the nuclear spin magnetisation. (B) Out of the 12 levels in the N@C_{60} spin system, four are chosen to represent the electron and nuclear spin states. For bang-bang decoupling, one electron spin transition is driven resonantly over a complete rotation. The $M_S = +3/2$ to $M_S = -3/2$ transition is driven via the intermediate levels $M_S = \pm 1/2$. No population remains in the intermediate levels after a complete π or 2π rotation. (C) To perform ultrafast nuclear phase gates, both electron spin transitions are driven simultaneously by a single, de-tuned microwave frequency.

2 Materials and methods

Our production and subsequent purification of N@C_{60} is described elsewhere [4]. High-purity N@C_{60} powder was dissolved in CS_2 to a final concentration of $10^{15}/\text{cm}^3$, freeze-pumped to remove oxygen, and finally sealed in a quartz tube. Samples were 0.7 cm long, and contained approximately 5×10^{13} N@C_{60} molecules. Pulsed electron paramagnetic resonance (EPR) measurements were made at 190 K using an X-band Bruker Elexsys580e spectrometer, equipped with a nitrogen-flow cryostat.

N@C_{60} has electron spin $S = 3/2$ coupled to the ^{14}N nuclear spin $I = 1$. The EPR spectrum consists of three lines centered at electron g-factor $g = 2.003$ and split by a ^{14}N isotropic hyperfine interaction $a = 0.56$ mT in CS_2 [5]. The electron nuclear double resonance (ENDOR) spectrum consists of four principle lines associated with the four M_S states, each of which is further split into two by the second-order hyperfine interaction [6], enabling selective excitation of (for example) the $M_I = 0$ to $M_I = +1$ transition. In these experiments we applied two simultaneous RF fields to coherently drive both the $|00\rangle - |01\rangle$ and $|10\rangle - |11\rangle$ nuclear transitions (see Fig. 2C). This is done solely to improve sensitivity, and as all microwave pulses do not exchange populations between these two subspaces, they can be treated independently. An alternative method to improve sensitivity by applying a second RF pulse after the electron spin echo detection is described elsewhere [7].

3 Nuclear spin Rabi oscillations

The observation of nuclear Rabi oscillations in N@C_{60} cannot practically be performed using NMR as the sensitivity is orders of magnitude too low. Instead we use a technique similar to the Davies ENDOR, and vary the duration of the RF pulse (as shown in Fig. 1A). To increase sensitivity, both the $M_S = \pm 3/2$ hyperfine lines are driven, as shown in Fig. 2. The powers of the two frequencies applied must be carefully tuned to drive both nuclear transitions coherently (cf. Figs. 2A and B).

4 A fast nuclear spin phase gate

It is possible to apply a phase gate to a qubit by rotating one of the basis states around a complete cycle through an auxiliary level. This phase has a geometric interpretation and is proportional to the area swept out by the path of this auxiliary cycle [8]. Crucially, all populations must remain unchanged after this operation.

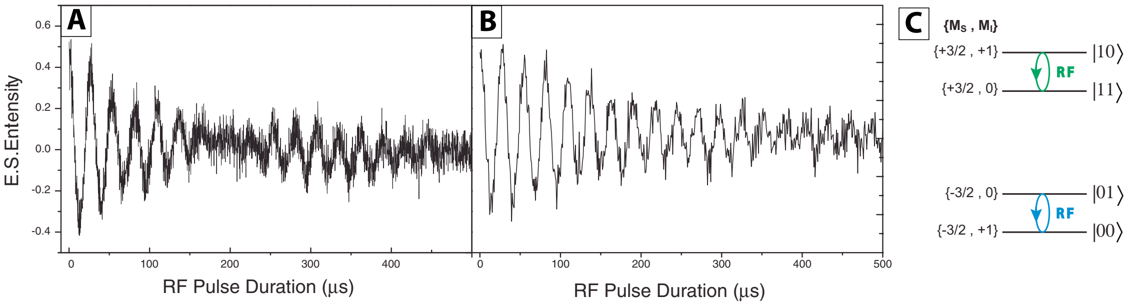

Fig. 2 (online colour at: www.pss-b.com) (A) Nuclear spin Rabi oscillations obtained when both the $M_S = \pm 3/2$ nuclear spin transitions (22.597 and 24.781 MHz) are driven simultaneously as in (C). The beating is due to a difference in the two Rabi frequencies. (B) The two concurrent Rabi oscillations can be brought into phase by adjusting the relative power in the two RF frequencies applied.

Such a phase gate can be implemented in the coupled electron/nuclear spin system of N@C$_{60}$ [9]. A phase shift is applied to the nuclear spin qubit by applying a microwave pulse to selectively rotate one of the transitions of a coupled electron spin (see Fig. 1B). The duration is chosen to leave all populations unchanged (i.e. a 2π rotation), depositing a global phase on the levels associated with the excited electron spin transition. When this microwave pulse is applied on resonance with the electron spin transition, a phase gate of π results on the nuclear spin. When the electron spin is excited by an off-resonant pulse, the axis of precession around the Bloch sphere becomes tilted and the area enclosed by the evolution of the magnetisation falls (see Fig. 3A). This allows the application of phase shifts less than π.

In practice, it may not be possible to excite one electron spin transition without also partially driving the other (especially if this is being done with a detuned pulse). A simple solution is to drive both the electron spin transitions ($|00\rangle - |10\rangle$ and $|01\rangle - |11\rangle$), with equal and opposite detuning (i.e. at a frequency half-way between their resonances). Thus, both transitions are driven through complete cycles, whilst the phase acquired by each is opposite. The relative phase accumulated (which defines the phase gate applied) is determined only by the microwave pulse power, allowing an arbitrary phase gate to be

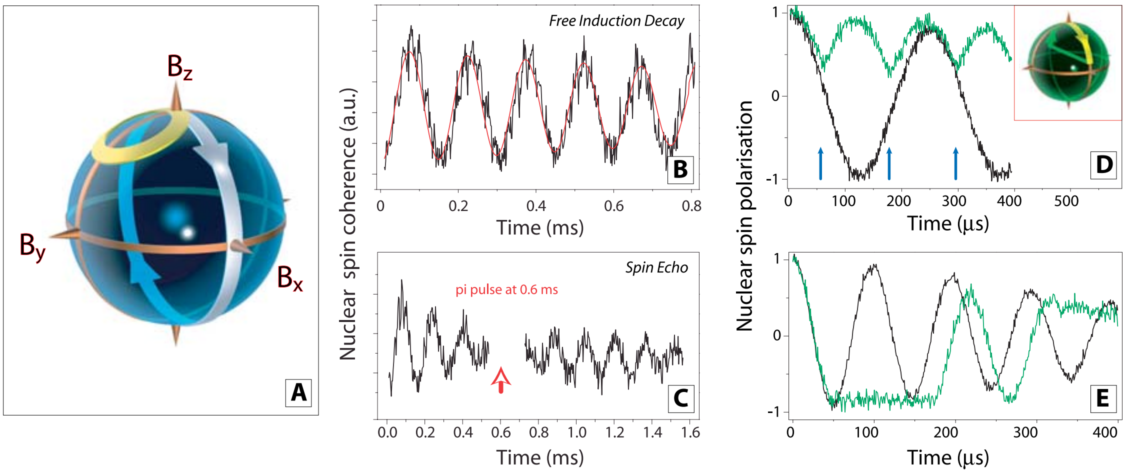

Fig. 3 (online colour at: www.pss-b.com) (A) Effect of applying on-resonance (white) and detuned (yellow) microwave pulses on the electron spin magnetisation. (B) The nuclear spin free induction decay and (C) nuclear spin echo indicate that the nuclear coherence time $T_{2,n}$ is limited only by the electron spin $T_{1,e}$. In the echo experiment, the FID is artificially shortened by deteriorating the homogeneity of the magnetic field, in order to yield a visible echo. (D & E) Black curves show unperturbed Rabi oscillations between two nuclear spin states of the nitrogen atom, while green curves show nuclear spin evolution under the influence of *bang-bang* decoupling pulses.

applied on the nuclear spin (see also [11]). This phase gate, whilst applied to the nuclear spin, is on the time scale of the electron spin rotation.

5 *Bang-bang* decoupling and nuclear spin relaxation

The relaxation properties of the ^{14}N nucleus in N@C$_{60}$ were determined using techniques related to Davies ENDOR which have been proposed for measuring T_1 [7] and T_2 [10]. The nuclear spin T_1 was measured to be 44 ms at 170 K, while the nuclear free induction decay (FID) and spin echo experiments shown in Fig. 3 indicate that (at 170 K) the nuclear spin T_2 is limited only by longitudinal electron spin relaxation, $T_{1,e} = 0.4$ ms.

Given these long relaxation times, and in order to demonstrate the effectiveness of the bang-bang decoupling scheme in N@C$_{60}$, we simulated a strong environmental interaction by applying a strong RF field to drive Rabi oscillations in the nuclear spin; this strong RF field was then decoupled by applying fast phase 'kicks' using the π phase gate described above. The general pulse sequence used is essentially the same as used previously (see Fig. 1A), with additional microwave pulses applied at certain points during the RF pulse in order to suppress the nuclear spin evolution.

Figure 3D shows the effect of applying a π phase shift during the nuclear Rabi oscillations. The evolution is reversed each time a microwave pulse is applied, as illustrated on the nuclear Bloch sphere in the inset. By increasing the repetition rate of the phase shift pulses, the nuclear spin evolution can be locked in one particular state and released as desired (see Fig. 3E).

6 Conclusions

We have demonstrated how the electron spin in N@C$_{60}$ can be used as a resource for the nuclear spin qubit: polarisation can be transferred from the electron spin to the nucleus, and the nuclear spin state can be measured via the electron. The electron spin can be used to apply a phase gate on the nucleus which is orders of magnitude faster than typical NMR phase gates, and about 10^5 times faster than existing geometric phase gates in NMR [12]. Finally, this phase gate can be used to turn off unwanted nuclear spin interactions – this is of particular importance in a molecular system such as N@C$_{60}$ where other techniques (such as electrode gating) may not be possible.

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