Supplementary Information for
"Storage of Multiple Coherent Microwave Excitations in an Electron Spin Ensemble"

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1 ^{14}N@C$_{60}$

In our experiments, one of the electron spin systems we used is based on the N@C$_{60}$ molecule, consisting of an nitrogen atom enclosed within a C$_{60}$ fullerene. The electron spin arises from the $S = 3/2$ atomic nitrogen, which although normally highly reactive is stable because of the protection afforded by the carbon cage. The sample we used was in liquid carbon disulfide (CS$_2$) solution at room temperature. The EPR spectrum of ^{14}N@C$_{60}$ has three lines due to the nuclear spin $I = 1$ of the ^{14}N atom. We centered on the central line ($m_I = 0$) for our pulsed experiments. The electron spin relaxation time T_1 and decoherence time T_2 at room temperature are $T_1 = 120\mu$s and $T_2 = 80\mu$s [1, 2].

For further information: the synthesis and early EPR studies of N@C$_{60}$ can be found in Ref. [3, 4, 5]; the electron spin relaxation times and mechanisms of N@C$_{60}$ are reported in Ref. [1, 2]; the applications of N@C$_{60}$ in quantum information processing are discussed in Ref. [6, 7].

2 Microwave pulses and cavities

All microwave pulses were in the range 9.7 – 9.9 GHz, supplied from a Bruker Elexsys 680 pulsed ESR spectrometer.

For experiments on N@C$_{60}$, microwave pulses were on resonance with the electron spins in the $m_I = 0$ hyperfine line in the EPR spectrum. The strength of the pulses was such that a π rotation of the electron spin state is achieved in 112 ns (which corresponds to a microwave intensity of 1.5 Gauss). The microwave cavity used was a cylindrical dielectric ring resonator (Bruker, ER 4118X-MD5).

For experiments on Si:P, the strength of the microwave pulses was such that a π rotation of the electron spin state is achieved in 200 ns. In the experiment of storing multiple weak excitations (shown in Fig. 3), the length of each small tipping pulse was 4 ns, which corresponds to a flip angle of $\sim 0.01\pi$. The microwave cavity used was a cylindrical dielectric ring resonator (Bruker, EN 4118X-MD4).

3 Derivation of the echo intensity as a function of wave number k

The ^{14}N@C$_{60}$ sample used in our experiment was in solution in a cylindrical tube. The distribution of spins along z direction is $n(z) \propto \sqrt{r_0^2 - z^2}$, with r_0 the radius of the tube. Thus, the orthogonality relation given in the first page of the main text becomes

$$\frac{1}{N} \int_{-r_0}^{r_0} \sqrt{r_0^2 - z^2} \cdot e^{-ik \cdot z} \mathrm{d}z \propto \frac{J_1(kr_0)}{kr_0} = 0, \tag{1}$$

where $k = k_i - k_j$, k_i, k_j are two different modes in the k-space, J_1 is the first order Bessel function.

For the P doped Si sample which has a uniform a cross-section, the orthogonality relation becomes

$$\frac{1}{N} \int_{-r_0}^{r_0} e^{-ik \cdot z} \mathrm{d}z \propto \mathrm{sinc}(kr_0) = 0. \tag{2}$$

In this case, since $\mathrm{sinc}(x)$ has equidistant zero points, the modes $k, 2k, \ldots, nk$ are orthogonal to each other.

4 Crosstalk between two different modes

In Fig. 1 we show the crosstalk between different modes k as a function of the intensity of the stored microwave excitations. We plot both experimental data and theoretical calculations. The curves for the theoretical results are $D_1 = (1 - \cos\theta_2)/2$, $D_2 = 1 - \cos\theta_1$, with θ_1 and θ_2 the flip angles of P$_1$ and P$_2$. The deviation between the experimental data and the theoretical curves is less than 0.5%.
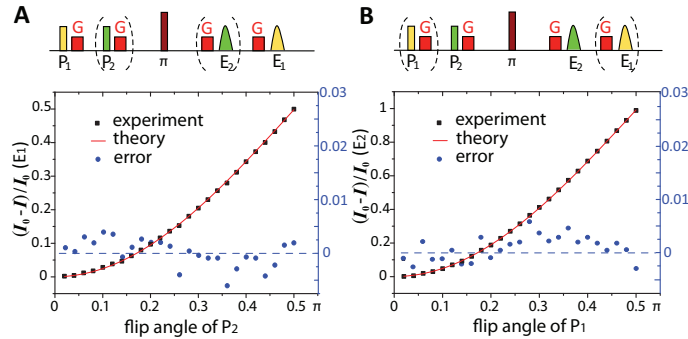

Figure 1: (color online). Difference in the recovered echo intensity (D) of one microwave excitation caused by another excitation. (A): D for the echo of the first microwave pulse (P$_1$) E$_1$ as a function of the flip angle of the second microwave pulse P$_2$. (B): D for E$_2$ as a function of the flip angle of P$_1$. In both cases the durations of P$_1$ and P$_2$ are swept from less than 0.02π to 0.6π, however D was independent of the intensity of the microwave pulse whose echo intensity was measured.

5 Coherence transfer between electron and nuclear spins

The pulse sequence for the coherence transfer from electron spins to nuclear spins is composed of a radio frequency π pulse on resonance with the transition between nuclear spin levels $|m_S = \frac{1}{2}, m_I = -\frac{1}{2}\rangle \rightarrow |m_S = \frac{1}{2}, m_I = \frac{1}{2}\rangle$ followed by a microwave π pulse on resonance with the transition between electron spin levels $|m_S = \frac{1}{2}, m_I = -\frac{1}{2}\rangle \rightarrow |m_S = -\frac{1}{2}, m_I = -\frac{1}{2}\rangle$. Each of the selective π pulses can be considered as a C-NOT

operation, therefore the pair of π-mw, π-rf pulse constitute a SWAP operation between the electron spin and nuclear spin.

Supposing an arbitrary superposition state $\alpha|0\rangle + \beta|1\rangle$ is initially stored in the electron spin coherence, and the nuclear spin is polarized in the $m_I = -1/2$, i.e. $|0_n\rangle$ state. The initial two qubit state can be written as $(\alpha|0_e\rangle + \beta|1_e\rangle)|0_n\rangle$, where $|0_e\rangle$ and $|1_e\rangle$ denote the $m_S = 1/2$ and $m_S = -1/2$ states, respectively. After the first rf π pulse, which will only flip the nuclear spins with the electron spin in the $|0_e\rangle$ state, the two qubit state becomes $\alpha|0_e1_n\rangle + \beta|1_e0_n\rangle$. In a similar way, the second mw π pulse will only flip the electron spins with the nuclear spin in the $|0_n\rangle$ state, therefore the final state will be $\alpha|0_e1_n\rangle + \beta|0_e0_n\rangle = |0_e\rangle(\alpha|0_n\rangle + \beta|1_n\rangle)$. The states of the electron spin and nuclear spin are swapped, and the superposition state is now stored in the nuclear spin coherence.

The inverse pulse sequence (a mw π pulse followed by an rf π pulse) transfers the spin coherence from nuclear spins back to electron spins. The refocusing is achieved by an rf π-pulse applied at time $\tau_n/2$, where τ_n is the time during which the coherence is stored in the nuclear spins (τ_n must be shorter than the nuclear decoherence time T_{2n}). If the delay between the input of the microwave excitations and the start of the coherence transfer sequence is τ_d, the spin echo signals of the microwave excitations will be observed at τ_d after the inverse transfer sequence.

More detailed description of the coherence transfer procedure can be found in Ref. [8].

In our experiment, the frequency of the rf pulses is $\nu_{rf} = 65$ MHz, and the length of an rf π pulse is 25 μs. Since the mw π pulse (112 ns) is much shorter than rf pulses, the length of the coherence transfer sequence is decided by the rf π pulse.

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