Supporting Information

Qubit Crossover in the Endohedral Fullerene $Sc_3C_2@C_{80}$

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List of Supporting Information

SI1 Sample preparation and physical measurement information

$Sc_3C_2@C_{80}$ sample was solved in CS_2 (purchased from J&K Scientific, Super Dry Solvents) in a 3-mm quartz EPR tube, then froze and pumped for 5 times to exclude oxygen. The concentration of $Sc_3C_2@C_{80}$ is determined by Bruker® spin counting by calculating using the height and diameter of the sample and the cavity quality factor Q at room temperature.(Eaton, G. R.; Eaton, S. S.; Barr, D. P.; Weber, R. T., *Quantitative EPR*. Springer Vienna: 2010.)

Continuous wave (cw) EPR measurements were performed on a Bruker E580 spectrometer using ER 4122 SHQE high sensitive EPR cavity. Oxford ESR900 cryostat was used for temperature control.

Pulse EPR measurement were performed on a Bruker E580 spectrometer using ER 4118 X-MS3 cavity. Oxford CF935 cryostat was used for temperature control.

SI2 Continuous wave EPR measurements

Continuous wave EPR measurements were performed at various temperatures from 130 K to 150 K. The microwave frequency is at 9.367902 GHz. The spectra are shown in **Figure S2 (black)**. The best simulation for experiment data at 150 K are shown in **Figure S2 (red)**. The best parameters for simulation is g_{iso} = 2.00 , A_{iso} = 18.10 MHz at 150 K.

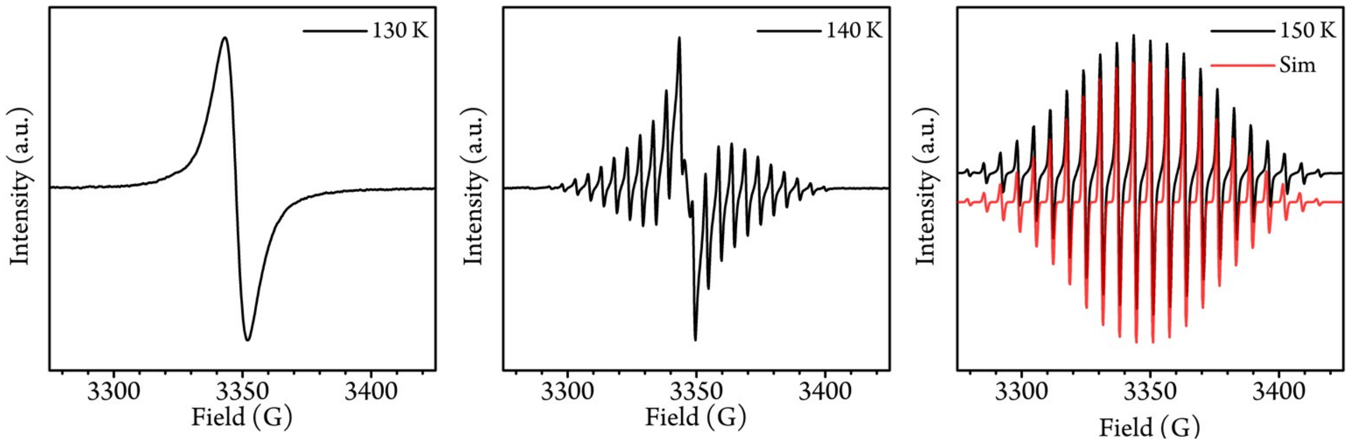

Figure S2. cw-EPR measurements on Bruker E580 at different temperature in CS_2 solution

SI3 Relaxation measurements at high temperature region

1. Spin-lattice relaxation (T_1) at different temperatures

   T_1 is measured using inverse-recovery method. The pulse sequence is π-T-π/2-τ-π-τ-echo with fixed τ and varied T. We use π/2 = 8 ns and π = 16 ns pulse for the measurement. τ = 100 ns is chosen for detection. 16-step phase cycling was used to eliminate the contribution of unwanted free-induction decay (FID) and echo signals. The experiment data are shown in **Table S3-1.**

Table S3-1. T_1 experiment data measured at different temperatures

Tempreature	T_1 value	Tempreature	T_1 value	Tempreature	T_1 value
150 K	177(1) ns	165 K	180(1) ns	180 K	180(1) ns
200 K	168(1) ns	240 K	165(1) ns	298 K	66(1) ns

2. Phase memory time (T_M) at different temperature

   T_M is measured using Hahn-echo decay method. The pulse sequence is π/2-τ-π-τ-echo with τ varies. We use π/2 = 8 ns and π = 16 ns pulse for the measurement. τ = 100 ns is chosen for initial interval. 16-step phase cycling was used to eliminate the contribution of unwanted free-induction decay (FID) and echo signals. The experiment data are shown in **Table S3-2.**

Table S3-2. T_M experiment data measured at different temperatures

Tempreature	T_M value	Tempreature	T_M value	Tempreature	T_M value
150 K	98(1) ns	165 K	128(1) ns	180 K	139(1) ns
200 K	139(1) ns	240 K	111(1) ns	298 K	75(2) ns

3. Phase memory time (T_M) at different magnetic field

   T_M is measured using Hahn-echo decay method. The pulse sequence is π/2-τ-π-τ-echo with τ varies. We use π/2 = 8 ns and π = 16 ns pulse for the measurement. τ = 100 ns is chosen for initial interval. 16-step phase cycling was used to eliminate the contribution of unwanted free-induction decay (FID) and echo signals. The experiment data are shown in **Table S3-3.** The first and the last transitions are hardly observable due to the weak signal.

Table S3-3. T_M experiment data measured at magnetic field

Magnetic Field	TM value	Magnetic Field	TM value	Magnetic Field	TM value
3257.6 G	119(1) ns	3264.1 G	126(1) ns	3270.6 G	130(1) ns
3277.6 G	137(1) ns	3283.6 G	138(1) ns	3289.6 G	137(1) ns
3296.2 G	138(1) ns	3302.7 G	136(1) ns	3309.2 G	137(1) ns
3315.7 G	138(1) ns	3322.3 G	137(1) ns	3328.8 G	136(1) ns
3334.8 G	137(1) ns	3341.8 G	137(1) ns	3348.4 G	139(1) ns
3354.4 G	138(1) ns	3360.9 G	135(1) ns	3367.4 G	132(1) ns
3373.9 G	128(1) ns	3380.5 G	114(1) ns		

SI4 Relaxation measurements at low temperature region

4. Spin-lattice relaxation (T_1) at different temperatures

   T_1 is measured using inverse-recovery method. The pulse sequence is π-T-π/2-τ-π-τ-echo with fixed τ and varied T. We use π/2 = 8 ns and π = 16 ns pulse for the measurement. τ = 150 ns is chosen for detection. 16-step phase cycling was used to eliminate the contribution of unwanted free-induction decay (FID) and echo signals. The experiment data are shown in **Table S4-1.**

Table S4-1. T_1 experiment data measured at different temperatures

Tempreature	T_1 value	Tempreature	T_1 value	Tempreature	T_1 value
5 K	96(11) ms	8 K	84(11) ms	10 K	21(1) ms
15 K	13(3) ms	20 K	0.9(1) ms	25 K	0.27(3) ms
40 K	30(2) μs	60 K	10(1) μs	80 K	6.9(3) μs

5. Phase memory time (T_M) at different temperature

   T_M is measured using Hahn-echo decay method. The pulse sequence is π/2-τ-π-τ-echo with τ varies. We use π/2 = 8 ns and π = 16 ns pulse for the measurement. τ = 150 ns is chosen for initial interval. 16-step phase cycling was used to eliminate the contribution of unwanted free-induction decay (FID) and echo signals. The experiment data are shown in **Table S4-2.**

Table S4-2. T_M experiment data measured at different temperatures

Tempreature	T_M value	Tempreature	T_M value	Tempreature	T_M value
5 K	13.6(4) μs	8 K	16.4(7) μs	10 K	17.2(7) μs
15 K	14.3(7) μs	20 K	10.6(5) μs	25 K	8.4(2) μs
40 K	6.4(2) μs	60 K	1.6(1) μs	80 K	1.2(1) μs

6. Phase memory time (T_M) using dynamic decoupling

   T_M is measured using Hahn-echo decay method. The pulse sequence is $\pi/2\text{-}\tau\text{-}(\pi\text{-}2\tau)_{n-1}\text{-}\pi\text{-}\tau$-echo with τ variation. We use π/2 = 8 ns and π = 16 ns pulse for the measurement. τ = 150 ns is chosen for initial interval. Phase cycling was used to eliminate the contribution of unwanted free-induction decay (FID) and echo signals. The experiment data are shown in **Table S4-3.**

Table S4-3 T_M experiment data measured at different temperatures using dynamic decoupling

CPMG-n	T_M value	CPMG-n	T_M value	CPMG-n	T_M value
CPMG-1	13.6(4) μs	CPMG-2	22.1(1) μs	CPMG-4	29.2(1) μs
CPMG-8	38.5(1) μs	CPMG-16	61(1) μs	CPMG-32	68.0(4) μs

SI5 ESEEM measurements at low temperature region

3-pulse ESEEM is measured using the sequence: π/2-τ-π/2-T-π/2-τ-echo, which measures the intensity of the echo as function of delay time T. We use π/2 = 8 ns and π = 16 ns pulse for the measurement. τ = 146 ns and initial T = 400 ns are chosen for interval. The 3-pulse ESEEM effect strongly depends on the magnetic field, so the 3-pulse ESEEM vs B_0 2D experiment was performed. The deepest field position is at the field of 3290 G as shown below in **Figure S5-1**. The 3-pulse ESEEM measurement with good signal to noise is shown in **Figure S5-2**.

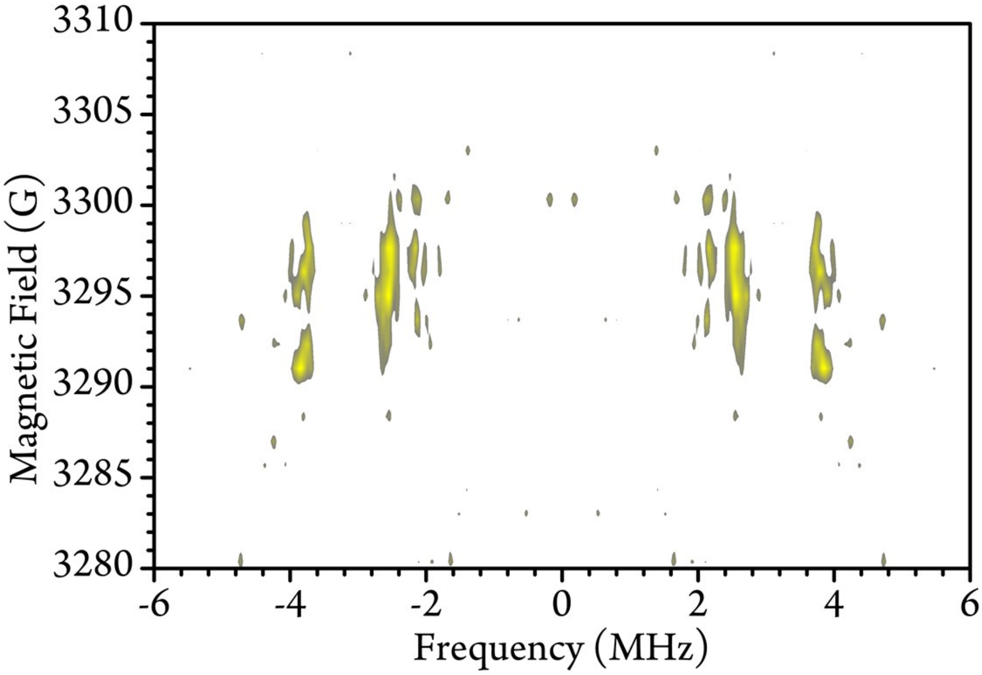

Figure S5-1. The 3pESEEM measurements are performed at different fields. B_0 = 3296 G was selected to excite both allowed and forbidden transitions, therefore it is able to generate the deepest modulation.

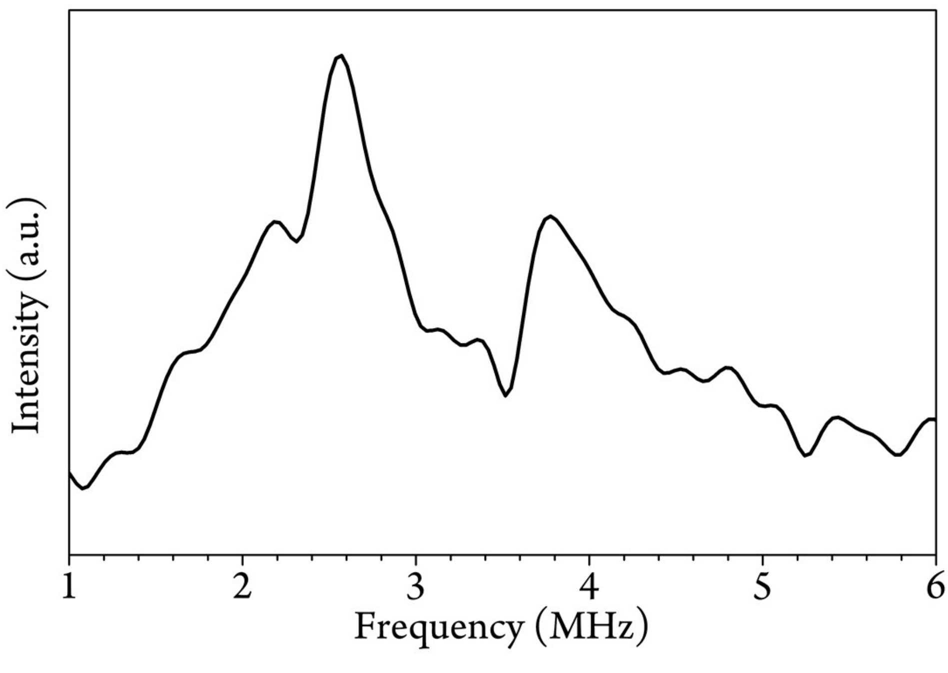

Figure S5-2. The 3pESEEM measurements performed at 3296 G.