Singlet-Triplet Physics and Shell Filling in Carbon Nanotube Double Quantum Dots

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(Dated: February 5, 2008)

PACS numbers: 73.63.-b, 03.67.-Lx, 73.63.Fg, 73.63.Kv, 05.60.Gg

An artificial two-atomic molecule, also called a double quantum dot (DQD), is an ideal system for exploring few electron physics. Spin-entanglement between just two electrons can be explored in such systems where singlet and triplet states are accessible. These two spin-states can be regarded as the two states in a quantum two-state system, a so-called singlet-triplet qubit. A very attractive material for realizing spin based qubits is the carbon nanotube (CNT). Because it is expected to have a very long spin coherence time, it has clear shell structures of both four and eight electrons, with the singlet-triplet qubit present in the four-electron shells. We furthermore observe inelastic cotunneling via the singlet and triplet states, which we use to probe the splitting between singlet and triplet, in good agreement with theory.

Creating a qubit in a solid-state system demands control of the number of interacting electrons. This control has to date been obtained using semiconducting materials operated close to the band-gap edge. We show in this Letter that shell structures in CNT DQDs, owing to the 1-dimensional nature of the CNT, can be used to obtain the same kind of control. Both 4-electron and 8-electron (DQD)-shells are observed. We use one of the 4-electron shells to entangle the spin of two electrons and show that by separating the two electrons into separate QDs they form a spin triplet state, and by collecting them into the same QD they form a spin singlet state, i.e., a gate-tunable singlet-triplet qubit.

The device analyzed in this Letter, schematically shown in Fig. 1(a), is comprised of a CNT contacted by titanium electrodes, and gated by three top-gate electrodes, G1, CG (center-gate), and G2, made of aluminum oxide and titanium. The device has two strongly coupled quantum dots in series as confirmed by the observation of the so-called honeycomb pattern in current (Isd) versus voltage applied to G1 (V_G1) and G2 (V_G2) (Fig. 1(b) and Fig. 2(a)). The tunneling barrier between the two dots is due to a defect in the CNT (similar to Ref. 17). The resulting two dots have roughly equal charging energies and level spacings (see below), from which we infer that the defect is located under or close to the center gate. The number of electrons in dot 1 and dot 2 can be controlled by tuning V_G1 and V_G2.

In the middle of each hexagon (white areas in Fig. 1(b) and Fig. 2(a)) a fixed number of electrons are localized in each dot, and electron transport is suppressed by Coulomb blockade. Along the entire edge of the hexagons (blue lines), single electron transport is allowed through molecular states formed in the DQD, indicating a strong coupling between the two dots. The height (width) of the hexagons corresponds to the energy required to add an extra electron in dot 1 (dot 2). In Fig. 1(b) the width and height of the hexagons alternate in size in a regular pat-
tern. The four hexagons marked with red numbers are distinctively larger than the other hexagons with three smaller hexagons in between, indicating that each dot has four-fold degenerate levels due to spin and orbital degeneracy. An 8-electron shell structure of the DQD can therefore be identified in this plot. Shell occupation numbers (N,M), where N (M) is the level occupation of voltage applied to G1 (G2), can be extracted from bias spectroscopy plots (not shown), and \( \Delta E_1 \) to only spin degeneracy of the energy levels in each hexagon. The pattern in the sizes of the hexagons is observed. The dots are roughly equal in size. We have observed both 4-electron and 8-electron shell structures in two different devices.

We will in the rest of the Letter focus on a 4-electron shell with level spacings and charging energies similar the the 4-electron shell shown in Fig. 2(a), except \( \Delta E_1 \sim 1.9 \text{ meV} \). The singlet ground state between region (1,1) and (0,2) is in general a bonding state of the local singlet (S(11), one electron in each dot):\[
S_B = \alpha S(11) + \beta S(02) \tag{1}
\]
The detuning (\( \varepsilon = E_2 - E_1 \)) dependent parameters \( \alpha \) and \( \beta \) determine the weight of each state, and \( E_1 \) and \( E_2 \) are the electrostatic potentials in dot 1 and dot 2, respectively. Similarly for the triplets:

\[
T_{B-} = \alpha T_-(11) + \beta T_-(02) \tag{2}
\]
\[
T_{B0} = \alpha T_0(11) + \beta T_0(02) \tag{2}
\]
\[
T_{B+} = \alpha T_+(11) + \beta T_+(02) \tag{2}
\]
FIG. 4: Singlet-triplet splitting probed by inelastic cotunneling. (a) Small section of the honeycomb diagram analyzed in Fig. 3 with $V_{sd} = 50 \mu V$ at $B = 0$ T (left) and $B = 6.5$ T (right). The numbers $(N,M)$ indicate electron occupation of the 4-electron shell. (b) Chemical potentials for the singlet bonding ($\mu_{S_{B\rightarrow 00}}$) and triplet bonding ($\mu_{T_{B\rightarrow 00}}$) with $B = 0$ T (solid green and blue lines) and with $B = 6$ T (dashed green and blue lines) calculated using Eq. (3) and (4). Onset of inelastic cotunneling which excites electrons from singlet to triplet (black arrow marked B) and from triplet to singlet (black arrow marked C) occurs when the separation between $\mu_{S_{B\rightarrow 00}}$ and $\mu_{T_{B\rightarrow 00}}$ is equal to $eV_{sd}$. (c) Schematic transport diagrams for elastic cotunneling (A) and inelastic cotunneling (B and C). (d) Surface plot of current ($I_{sd}$) at $V_{sd} = 0.2 \mu V$ versus magnetic field ($B$), and detuning ($c$) along the black dashed line in (a). Onset of inelastic cotunneling occurs along the white lines marked B and C, calculated using Eq. (5) with $t = 0.32$ meV, $\Delta E_2 = 1.5$ meV, $V_{sd} = 0.2$ meV, and $g = 2$. Dashed grey lines indicate where the two inelastic cotunneling processes shown in (b) occurs.

where $-, 0, +$ denotes the spin magnetic moment in the z-direction, $S_z = -1, 0, +1$. We will in the following show the existence of the singlet and triplet states, i.e., the singlet-triplet qubit, on the basis of a magnetic field spectroscopy on the 4-electron shell.

In Fig. 3(a), we analyze the magnetic field dependence of the width of hexagon $(0,1)$, which involves 0, 1, and 2 electrons in dot 2. The chemical potential for these two Coulomb peaks is given by $\mu_{S_{B\rightarrow 00}} \propto -\frac{1}{2} g\mu_B B$ and $\mu_{S_{B\rightarrow 00}} \propto \frac{1}{2} g\mu_B B$, where $\mu_{01-00}$ is the chemical potential for adding an electron to charge state (01) given no electron in the DQD-shell, and $\mu_{S_{B\rightarrow 00}}$ is the chemical potential for adding an electron in state $S_B$ given one electron charge state (01). These two Coulomb peaks are therefore expected to separate by $g\mu_B B$ as shown in Fig. 3(a). The height of hexagon $(0,1)$ is analogously expected to separate by $g\mu_B B$. The measurements in Fig. 3(a) and (c) are in good quantitative agreement with the theory in Fig. 3(a). The measured separation at 7 T is 0.95 meV and 0.8 meV in (c) and (e), respectively, where theory predicts $g\mu_B 7 T = 0.81$ meV with $g = 2$ for nanotubes.

We now analyze the size of hexagon (1,1), which involves 1, 2, and 3 electrons in the DQD-shell. We show that by applying a magnetic field the 2-electron ground state can be changed from $S_B$ to $T_{B-}$, which is used to estimate the exchange energy ($J$) (energy separation between $S_B$ and $T_{B-}$). Transport at the first Coulomb peak in Fig. 3(d) is through different chemical potentials at low and high magnetic field, given by $\mu_{S_{B\rightarrow 10}} \propto +\frac{1}{2} g\mu_B B$ at the low magnetic field ($g\mu_B B < J$), and $\mu_{T_{B\rightarrow 10}} \propto -\frac{1}{2} g\mu_B B$ at high magnetic field ($g\mu_B B > J$). Similarly, transport at the second Coulomb peak in Fig. 3(d) is through $\mu_{12\rightarrow S_B} \propto -\frac{1}{2} g\mu_B B$ at low magnetic field ($g\mu_B B < J$) and through $\mu_{12\rightarrow T_{B-}} \propto +\frac{1}{2} g\mu_B B$ at high magnetic field ($g\mu_B B > J$). The same magnetic field dependence is expected for the height of hexagon (1,1) (see Fig. 3(f)). Therefore, for increasing magnetic field, hexagon (1,1) decreases in size when $S_B$ is ground state, and increases in size when $T_{B-}$ is ground state, schematically shown in Fig. 3(b). The measurements in Fig. 3(d) and (f) are in good agreement with the theory in Fig. 3(b) with the bend (shift of ground state from singlet to triplet) occurring at $B \approx 2 - 3$ T, corresponding to an exchange energy of $J \approx 0.23 - 0.35$ meV.

The exchange energy can, for large negative detuning (center of hexagon (1,1)), also be estimated from the tunnel coupling strength ($t$) using $J \approx 4(t\sqrt{2})^2/U_{C1}$ (see supplement material) [19][30]. We estimate $t \approx 0.32$ meV from the curvature of the hexagons at the anticrossings (see supplement material) [20]. This estimate of $t$ yields a consistent estimate of the exchange energy.
$J \simeq 4(\sqrt{2})^2/U_{C1} \sim 0.27$ meV.

The anticrossing between (1,1) and (0,2) (red area in fig. 2(b)) is analyzed in Fig. 3. We find that transport is governed by elastic and inelastic cotunneling via $S_B$ and $T_{B-}$. The chemical potential for adding an electron to $S_B$ and $T_{B-}$ occurs at $E_1 + E_2 = 0$, i.e., along the black dashed line in Fig. 3(a) is given by (see supplement):

$$\mu_{S_B \rightarrow 01}(\epsilon, B) = -\frac{1}{2} \sqrt{2(\sqrt{2})^2 + \epsilon^2 + \frac{1}{2}g\mu_B B}$$

(3)

$$\mu_{T_{B-} \rightarrow 01}(\epsilon, B) = -\frac{1}{2} \sqrt{2(\sqrt{2})^2 + (\epsilon - \Delta E_2)^2 - \Delta E_2}$$

$$-\frac{1}{2}g\mu_B B$$

(4)

We plot Eq. (3) and (4) in Fig. 4(b) with $B = 0$ T (solid green and blue lines), and with $B = 6$ T (dashed green and blue lines). We see that $S_B$ is in the ground state for $B = 0$, and that the two chemical potentials cross at $\epsilon = 0$. At high magnetic field the elastic cotunneling via $S_B$ becomes suppressed because the ground state at $\epsilon = 0$ changes from $S_B$ to $T_{B-}$, Fig. 4(d) shows a surface plot of $L_d$ versus $\epsilon$ and $B$ along the black dashed line in Fig. 3(a). The white vertical line marked A is the expected position of the elastic cotunneling.

At high magnetic field we observe two narrow peaks, marked B and C in Fig. 4(a). These two narrow peaks are due to the onset of inelastic cotunneling via $S_B$ and $T_{B-}$, schematically shown in Fig. 4(c) (mark A). Since elastic cotunneling via $S_B$ involves both $S(11)$ and $S(02)$, which have equal weight at $\epsilon = 0$, the elastic cotunneling peak is centered around $\epsilon = 0$. At high magnetic field the elastic cotunneling via $S_B$ becomes suppressed because the ground state at $\epsilon = 0$ changes from $S_B$ to $T_{B-}$. Fig. 4(d) shows a surface plot of $L_d$ versus $\epsilon$ and $B$ along the black dashed line in Fig. 3(a). The white vertical line marked A is the expected position of the elastic cotunneling.

We have from these two conditions calculated the onset of inelastic cotunneling in $(\epsilon, B)$-space and plotted it as white lines marked B and C in Fig. 4(d). Note that no fitting parameters are used in Fig. 4(d), the parameters used, $t = 0.32$ meV, $\Delta E_2 = 1.5$ meV were found in the analysis above.

METHODS

Fabrication and measurement setup

The devices are made on a highly doped silicon substrate with a top layer of silicon dioxide. The CNTs are grown by chemical vapor deposition from islands of catalyst material and subsequently contacted by 50 nm Titanium source and drain electrodes. Next, three narrow top-gate electrodes are fabricated between the source and drain electrodes, consisting of aluminum oxide and titanium [10]. A schematic figure of the device together with the measurement setup is shown in Fig. 1(a). Source-drain voltage ($V_{sd}$) is applied to the source electrode and the drain electrode is connected through a current-to-voltage amplifier to ground. The three top-gate electrodes are named G1, CG (center gate), and G2 starting from the source electrode. For the device reported on in this Letter we saw that G1 had a much lower gate-coupling than G2 and CG (see Fig. 1(b) and Fig. 2(a)) which we attribute to the G1-electrode being damaged somewhere, weakening its gate-coupling. The gate coupling of G1 to dot 1 is $\alpha_{G1} = 2.9$ meV/V, and gate coupling of G2 to dot 2 is $\alpha_{G2} = 400$ meV/V. The center gate is kept at $V_{CG} = 0$ V for the measurements shown in this Letter. All data presented in this Letter are measured in a sorption pumped $^3$He cryostat at 350 mK.

ACKNOWLEDGEMENTS

We wish to acknowledge the support of the EU-STREP ULTRA-1D program and the EU FP6 CANAPE project.

COMPETING FINANCIAL INTERESTS

The authors declare no competing financial interests.

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