Few Electron Physics in a 4-electron Shell of a Nanotube Double Quantum Dot

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We will here model the nanotube double quantum dot with a Hamiltonian and derive the following expressions used in the Letter: (i) Chemical potential for the singlet bonding and triplet bonding (with $Z_z = -1$), (ii) an expression for the exchange energy (singlet-triplet splitting) at large detuning, and (iii) a formula for the separation between the two wings at the anticrossing between region (1,1) and (0,2) for extraction of the coupling parameters (t and $U_{\rm Cm}$).

THE MODEL HAMILTONIAN

We model the nanotube double quantum dot by two identical quantum dots having charging energy ($U_{\rm C}$), mutual charging energy ($U_{\rm Cm}$), and inter-dot tunnel coupling (t). A 4-electron shell is considered, where each dot has spin degenerate orbitals separated by level spacings. We analyze the anticrossing region between charge state (1,1) and (0,2) and therefore include one spin-degenerate orbital in dot 1, and two spin-degenerate orbitals in dot 2 separated by a level spacing ΔE_2. Two orbitals in dot 2 are included in order to include the local triplet states ($\rm T_-(02)$, $\rm T_0(02)$, and $\rm T_+(02)$) (see below).
The Hamiltonian therefore has the following three parts, an electrostatic part, a tunnel coupling or hybridization part, and a magnetic field part:

$$H_{\rm el} = U_{\rm Cm} n_1 n_2 + \frac{1}{2} U_{\rm C}\Big(n_1(n_1-1) + n_2(n_2-1)\Big) + \Delta E_2(n_{2^*\uparrow} + n_{2^*\downarrow}) - n_1 E_1 - n_2 E_2 \tag{1}$$

Where $n_1 = n_{1\uparrow} + n_{1\downarrow}$, and $n_2 = n_{2\uparrow} + n_{2\downarrow} + n_{2^*\uparrow} + n_{2^*\downarrow}$ is the total shell occupation number of electrons in dot 1 and dot 2, respectively. The subscript number "1" refers to orbital 1 in dot 1, "2" orbital 1 in dot 2, and "2*" orbital 2 in dot 2. The electrostatic potentials in each dot, E_1 and E_2, are tuned by gates.
The tunnel part has to include the two orbitals in dot 2

$$H_{\rm t} = -t \sum_{\sigma} \Big(c^\dagger_{1\sigma}(c_{2\sigma} + c_{2^*\sigma}) + (c^\dagger_{2\sigma} + c^\dagger_{2^*\sigma}) c_{1\sigma} \Big) \tag{2}$$

where $c^\dagger$ and c is creation and annihilation operators, and $\sigma = \uparrow, \downarrow$ is the electron spin.
The magnetic field contribution to the Hamiltonian is

$$H_{\rm B} = b\,(n_{1\uparrow} - n_{1\downarrow} + n_{2\uparrow} - n_{2\downarrow} + n_{2^*\uparrow} - n_{2^*\downarrow})\ , \quad b = \frac{1}{2} g \mu_{\rm B} B \tag{3}$$

where $g = 2$ for nanotubes, $\mu_{\rm B}$ is Bohr's magneton, and B is the magnetic field.

TWO-ELECTRON SPIN STATES: SINGLET AND TRIPLETS

There are singlet and triplet states in region (2,0), (1,1), and (0,2), but since we here analyze the anticrossing between region (1,1) and (0,2) where the states in region (2,0) has a much higher energy we neglect the (2,0) spin states. The low energy singlet and triplets in region (1,1) and (0,2) are given by:

$$\begin{array}{lcllcl} \rm S(11) & = & \frac{1}{\sqrt{2}}(|\uparrow_1\downarrow_2\rangle - |\downarrow_1\uparrow_2\rangle) & \rm S(02) & = & |\uparrow_2\downarrow_2\rangle \\ \rm T_+(11) & = & |\uparrow_1\uparrow_2\rangle & \rm T_+(02) & = & |\uparrow_2\uparrow_{2^*}\rangle \\ \rm T_0(11) & = & \frac{1}{\sqrt{2}}(|\uparrow_1\downarrow_2\rangle + |\downarrow_1\uparrow_2\rangle) & \rm T_0(02) & = & \frac{1}{\sqrt{2}}(|\uparrow_2\downarrow_{2^*}\rangle + |\downarrow_2\uparrow_{2^*}\rangle) \\ \rm T_-(11) & = & |\downarrow_1\downarrow_2\rangle & \rm T_-(02) & = & |\downarrow_2\downarrow_{2^*}\rangle \end{array} \tag{4}$$

Note the difference in state S(11) and S(02), which is due to the two electrons being in the same orbital in S(02) and in different orbitals in S(11). The singlet ground state in the region between (1,1) and (0,2) is a molecular singlet bonding state, which can be written as

$$\begin{array}{rcl} \rm S_B & = & \sin(\theta/2)\rm S(11) + \cos(\theta/2)S(02) \\ & = & \frac{1}{\sqrt{2}}\Big[\sin(\theta/2)(|\uparrow_1\downarrow_2\rangle - |\downarrow_1\uparrow_2\rangle) + \sqrt{2}\cos(\theta/2)\,|\uparrow_2\downarrow_2\rangle\Big] \end{array} \tag{5}$$

with

$$\tan(\theta) = \frac{2\sqrt{2}t}{\varepsilon'} \tag{6}$$

Where we have defined a (shifted) detuning parameter, $\varepsilon' = \varepsilon + U_{\mathrm{Cm}} - U_{\mathrm{C}}$ with $\varepsilon = E_2 - E_1$, and energy $E = E_2 + E_1$ The eigenenergy is:

$$E_{\mathrm{S_B}}(E, \varepsilon') = U_{\mathrm{Cm}} - E - \frac{1}{2}\Big(\sqrt{(2\sqrt{2}t)^2 + \varepsilon'^2} + \varepsilon'\Big) \tag{7}$$

So, this is the E and ε dependent eigenenergy of the singlet bonding state between region (1,1) and (0,2).
The triplet ground state at finite magnetic field is written as

$$\begin{aligned} \mathrm{T_{B-}} &= \sin(\theta/2) T_-(11) + \cos(\theta/2) T_-(02) \\ &= \sin(\theta/2)\,|{\downarrow_1}{\downarrow_2}\rangle + \cos(\theta/2)\,|{\downarrow_2}{\downarrow_2}\rangle \end{aligned} \tag{8}$$

where

$$\tan(\theta) = \frac{2t}{\varepsilon' - \Delta E_2} \tag{9}$$

and the eigenenergy is:

$$E_{\mathrm{T_{B-}}}(E, \varepsilon') = U_{\mathrm{Cm}} - E - \frac{1}{2}\Big(\sqrt{(2t)^2 + (\varepsilon' - \Delta E_2)^2} + \varepsilon' - \Delta E_2\Big) - 2b \tag{10}$$

The eigenenergy for $\mathrm{T_{B0}}$ and $\mathrm{T_{B+}}$ are found in the same way, and they only differ by the magnetic field contribution.

The exchange energy (J) is defined as the energy splitting between $\mathrm{S_B}$ and $\mathrm{T_{B0}}$, which for $-\varepsilon', \Delta E_2 \gg t$ is given by:

$$\begin{aligned} J(\varepsilon') &= E_{\mathrm{T_{B0}}}(E, \varepsilon') - E_{\mathrm{S_B}}(E, \varepsilon') \\ &\simeq \frac{(2\sqrt{2}t)^2}{|\varepsilon'|} \quad \text{, for} \quad -\varepsilon', \Delta E_2 \gg t \end{aligned} \tag{11}$$

We use this expression in the Letter for large negative detuning $\varepsilon' \sim -U_{\mathrm{C}}$.

The exchange energy at zero detuning ($\varepsilon' = 0$), and for $\Delta E_2 \gg t$, is given by:

$$J(\varepsilon' = 0) \simeq t\sqrt{2} \tag{12}$$

CHEMICAL POTENTIAL FOR THE SINGLET AND TRIPLETS

The 1-particle ground state at finite magnetic field in region (0,1) is $|{\downarrow_2}\rangle$ with eigenenergy:

$$E_{01}(\varepsilon) = -E_2 - b = -\frac{1}{2}(E + \varepsilon) - b \tag{13}$$

The chemical potential for adding an electron to the singlet bonding state ($\mu_{\mathrm{S_B}\leftrightarrow 01}$), and to the triplet bonding state with $S_z = -1$ ($\mu_{\mathrm{T_{B-}}\leftrightarrow 01}$), given one electron in state $|{\downarrow_2}\rangle$, are given by

$$\begin{aligned} \mu_{\mathrm{S_B}\leftrightarrow 01}(E, \varepsilon') &= E_{\mathrm{S_B}} - E_{01} \\ &= \tfrac{1}{2}U_{\mathrm{C}} + \tfrac{1}{2}U_{\mathrm{Cm}} - \tfrac{1}{2}E - \tfrac{1}{2}\sqrt{(2\sqrt{2}t)^2 + \varepsilon'^2} + b \end{aligned} \tag{14}$$

and

$$\begin{aligned} \mu_{\mathrm{T_{B-}}\leftrightarrow 01}(E, \varepsilon') &= E_{\mathrm{T_{B-}}} - E_{01} \\ &= \tfrac{1}{2}U_{\mathrm{C}} + \tfrac{1}{2}\Delta E_2 + \tfrac{1}{2}U_{\mathrm{Cm}} - \tfrac{1}{2}E - \tfrac{1}{2}\sqrt{(2t)^2 + (\varepsilon' - \Delta E_2)^2} - b \end{aligned} \tag{15}$$

Equation (14) and (15) are the same as Eq. (3) and (4) in the Letter with $E = 0$, i.e., along the black dashed line in Fig. 4(a). We have in the Letter also changed the energy zero-point of Eq. (14) and (15) for simplicity by subtracting the term $\frac{1}{2}U_{\mathrm{C}} + \frac{1}{2}U_{\mathrm{Cm}}$.

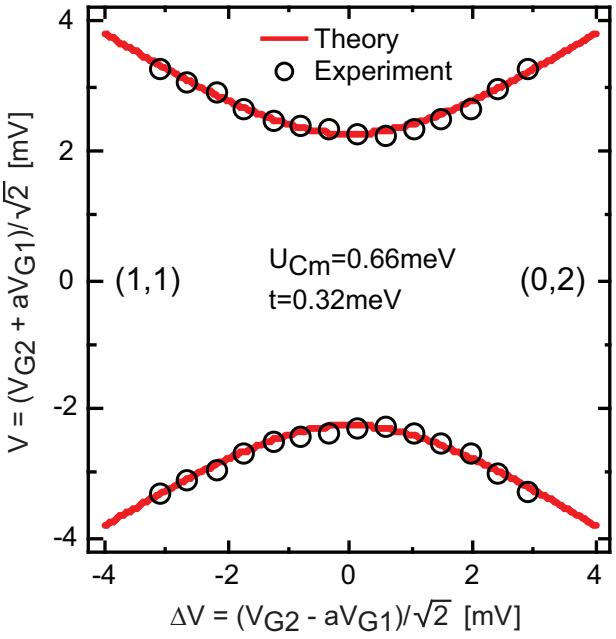

Figure S1: **Bend-fit for extraction of tunnel coupling (t) and mutual charging energy (U_{Cm}).** Black circles are the position of the upper and lower wing between region (1,1) and (0,2) extracted from Fig. 4(a) in the Letter. The separation between the two red lines are Eq. 21 with $U_{\mathrm{C}} = 3.25\,\mathrm{meV}$ and $\Delta V_{\mathrm{G}} = 8\,\mathrm{mV}$. Least square fitting yield $t = 0.32\mathrm{meV}$, and $U_{\mathrm{Cm}} = 0.66\,\mathrm{meV}$. The factor a$= 7 \cdot 10^{-3}$ on the x and y axis compensates for the asymmetric gate coupling of G1 and G2.

BEND-FIT FOR EXTRACTION OF TUNNEL COUPLING AND MUTUAL CHARGING ENERGY

We will in this section find analytical expressions for the shape of the two wings at the anti-crossing point between hexagon (1,1) and (0,2) in gate-voltage space, i.e., in V_{G1}, V_{G2} space. We use the expression to extract the two coupling parameters; mutual charging energy (U_{Cm}) and tunnel coupling (t) by fitting to the measured data [1].

The condition for transport through the lower wing is that the chemical potential for adding an electron to the singlet bonding state ($\mu_{\mathrm{S_B}\leftrightarrow 01}$) has to be between the chemical potential of source (μ_{S}) and drain (μ_{D}) (the bias window) which we approximate to be zero ($\mu \equiv \mu_{\mathrm{S}} = \mu_{\mathrm{D}} = 0$), i.e., zero bias voltage. The condition for transport through the lower wing is therefore $\mu_{\mathrm{S_B}\leftrightarrow 01} = 0$, which yields (total energy E versus detuning ε'):

$$E_{\mathrm{LW1102}}(\varepsilon') = -\sqrt{(2\sqrt{2}t)^2 + \varepsilon'^2} + U_{\mathrm{C}} + U_{\mathrm{Cm}} \tag{16}$$

The subscript "LW1102" refers to the Lower Wing in the region between (1,1) and (0,2). Similarly we get for the upper wing ($\mu_{12\leftrightarrow \mathrm{S_B}} = E_{12} - E_{\mathrm{S_B}} = 0$):

$$E_{\mathrm{UW1102}}(\varepsilon') = \sqrt{(2\sqrt{2}t)^2 + \varepsilon'^2} + U_{\mathrm{C}} + 3U_{\mathrm{Cm}} \tag{17}$$

Where $E_{12} = 2U_{\mathrm{Cm}} + U_{\mathrm{C}} - \frac{3}{2}E - \frac{1}{2}\varepsilon$ is the eigenenergy of the three particle state in region (12). The subscript "UW1102" refers to the Upper Wing in the region between (1,1) and (0,2).
The separation between the upper and lower wing is:

$$E_{\Delta 1102}(\varepsilon') = 2\sqrt{(2\sqrt{2}t)^2 + \varepsilon'^2} + 2U_{\mathrm{Cm}} \tag{18}$$

To be able to fit Eq. 18 to measured data and thereby extract t and U_{Cm} we need to transform it to be given in terms of the gate voltages, V_{G1} and V_{G2}. We do this by using the following relations,

$$\begin{aligned} E_1 &= \alpha_{11}V_{\mathrm{G1}} + \alpha_{21}V_{\mathrm{G2}} \\ E_2 &= \alpha_{12}V_{\mathrm{G1}} + \alpha_{22}V_{\mathrm{G2}} \end{aligned} \tag{19}$$

where the four α_{ij} factors are gate couplings. We assume no direct cross capacitance, only effective cross capacitance due to the inter-dot capacitance. For identical dots we have $\alpha_{11} = \alpha_{22}$ and $\alpha_{12} = \alpha_{21}$. We also define a new coordinate

system (shifted, and 45^o rotated), $V = (V_{G2} + V_{G1})/\sqrt{2}$, and $\Delta V = (V_{G2} - V_{G1} - \Delta V_G)/\sqrt{2}$, where ΔV_G is the width or height of hexagon (1,1) (since we assume identical dots the height and width are the same). Eq. (18) can now be written as:

$$V_{\Delta 1102}(\Delta V) = 2\sqrt{\left(\frac{2\sqrt{2}t}{\sqrt{2}(\alpha_{11}+\alpha_{12})}\right)^2 + \left(\frac{\alpha_{11}-\alpha_{12}}{\alpha_{11}+\alpha_{12}}\,\Delta V\right)^2} + \frac{2U_{Cm}}{\sqrt{2}(\alpha_{11}+\alpha_{12})} \tag{20}$$

By using the following relations[2]:

$$\alpha_{11} = \frac{U_C}{\Delta V_G} \quad , \quad \alpha_{12} = \frac{U_C \Delta V_G^m}{\Delta V_G^2} \quad , \quad \frac{U_{Cm}}{U_C} = \frac{\Delta V_G^m}{\Delta V_G}$$

it can be shown that

$$\frac{\alpha_{11}-\alpha_{12}}{\alpha_{11}+\alpha_{12}} = \frac{U_C - U_{Cm}}{U_C + U_{Cm}} \quad , \quad \sqrt{2}(\alpha_{11}+\alpha_{12}) = \sqrt{2}(U_C + U_{Cm})/\Delta V_G \equiv \alpha$$

The parameter α can be interpreted as a gate coupling factor. We can now rewrite Eq. (20) as:

$$V_{\Delta 1102}(\Delta V) = 2\sqrt{\left(\frac{2\sqrt{2}t}{\alpha}\right)^2 + \left(\frac{U_C - U_{Cm}}{U_C + U_{Cm}}\,\Delta V\right)^2} + \frac{2U_{Cm}}{\alpha} \tag{21}$$

We have extracted the position of the two wings at the anticrossing between region (1,1) and (0,2) from Fig.4(a) in the Letter and plotted them as black circles in Fig. S1. The separation between the two red lines in Fig. S1 is a least square fit of Eq. 21 to the data, yielding $t = 0.32\,\mathrm{meV}$, and $U_{Cm} = 0.66\,\mathrm{meV}$.

[1] M. R. Gräber, W. A. Coish, C. Hoffmann, M. Weiss, J. Furer, S. Oberholzer, D. Loss, C. schönenberger , Phys. Rev. B **74**, 075427 (2006).

[2] W. G. van der Wiel, S. de Franceschi, J. M. Elzerman, T. Fujisawa, S. Tarucha, L. P. Kouwenhoven, Rev. Mod. Phys **75**, 1-22 (2002).