Supporting Information for: A Triple Quantum Dot defined in a Single-Wall Carbon Nanotube

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S1. Electrostatics of a serial triple quantum dot

The electrostatics of the triple quantum dot shown schematically in the Letter in Fig. 1(c) can be determined by calculating the electrostatic energy U_{el} for a given charge configuration (N_1, N_2, N_3) and voltages on the gates V_i , $i = s, Tg1, Tg2, Tg3, bg, d$ giving

$$U_{el} = U_{el}(N_1, N_2, N_3, V_s, V_{Tg1}, V_{Tg2}, V_{Tg3}, V_{bg}, V_d)$$

The electrostatic energy is obtained by setting up the matrix equation $\bar{Q} = \mathbf{C}\bar{V}$ between the charge $\bar{Q}$ and the voltage $\bar{V}$ with $\mathbf{C}$ being the capacitance matrix. The components of $\bar{Q}$ and $\bar{V}$ are the charges and voltages on the nodes and sources in the circuit^{1,2}. The nodes are the three quantum dots with charges and voltages Q_i and V_i , $i = 1, 2, 3$, respectively, while the source charges and voltages are Q_i and V_i , $i = s, Tg1, Tg2, Tg3, bg, d$. Only part of the capacitance matrix now written in more details is needed to obtain the electrostatic energy

$$\begin{bmatrix} \bar{Q}_{node} \\ \bar{Q}_{source} \end{bmatrix} = \begin{bmatrix} \mathbf{C}_{node,node} & \mathbf{C}_{node,source} \\ \mathbf{C}_{source,node} & \mathbf{C}_{source,source} \end{bmatrix} \begin{bmatrix} \bar{V}_{node} \\ \bar{V}_{source} \end{bmatrix} \quad (1)$$

The two matrices to be found are the matrix $\mathbf{C}_{node,node}$ connecting the node charge and the node voltages as well as the matrix $\mathbf{C}_{node,source}$ connecting the node charges with the source voltages¹.

The charge on a node Q_i can be evaluated by considering all capacitances connected to it using¹

$$Q_i = \sum_j c_{ij}(V_i - V_j) \quad (2)$$

where j runs over all the nodes and sources and c_{ij} is the connecting capacitance. For the charge Q_1 on QD1, we therefore obtain

$$Q_1 = C_s(V_1 - V_s) + C_{Tg1}(V_1 - V_{Tg1}) + C_{bg1}(V_1 - V_{bg}) + C_{m12}(V_1 - V_2) \quad (3)$$

$$= (C_s + C_{Tg1} + C_{bg1} + C_{m12})V_1 - C_{m12}V_2 - C_sV_s - C_{Tg1}V_{Tg1} - C_{bg}V_{bg} \quad (4)$$

Similarly, we find the charge on QD2

$$Q_2 = C_{m12}(V_2 - V_1) + C_{Tg2}(V_2 - V_{Tg2}) + C_{bg2}(V_2 - V_{bg}) + C_{m23}(V_2 - V_3) + C_{Tg1QD2}(V_2 - V_{Tg1}) \quad (5)$$

$$+ C_{Tg3QD2}(V_2 - V_{Tg3}) \quad (6)$$

$$= -C_{m12}V_1 + (C_{m12} + C_{Tg2} + C_{bg2} + C_{m23} + C_{Tg1QD2} + C_{Tg3QD2})V_2 - C_{m23}V_3 - C_{Tg1QD2}V_{Tg1} \quad (7)$$

$$- C_{Tg2}V_{Tg2} - C_{Tg3QD2}V_{Tg3} - C_{bg2}V_{bg} \quad (8)$$

and the charge on QD3

$$Q_3 = C_d(V_3 - V_d) + C_{Tg3}(V_3 - V_{Tg3}) + C_{bg3}(V_3 - V_{bg}) + C_{m23}(V_2 - V_3) \quad (9)$$

$$= -C_{m23}V_2 + (C_{Tg3} + C_{bg3} + C_{m23} + C_d)V_3 - C_{Tg3}V_{Tg3} - C_{bg3}V_{bg} - C_dV_d \quad (10)$$

From these three equations, the matrix equation $\bar{Q}_{node} = \mathbf{C}_{node,node}\bar{V}_{node}$ and thus $\mathbf{C}_{node,node}$ are easily found

$$\begin{bmatrix} Q_1 \\ Q_2 \\ Q_3 \end{bmatrix} = \begin{bmatrix} C_1 & -C_{m12} & 0 \\ -C_{m12} & C_2 & -C_{m23} \\ 0 & -C_{m23} & C_3 \end{bmatrix} \begin{bmatrix} V_1 \\ V_2 \\ V_3 \end{bmatrix} \quad (11)$$

with the diagonal elements given as stated in the Letter

$$C_1 = C_s + C_{Tg1} + C_{bg1} + C_{m12} \quad (12)$$

$$C_2 = C_{m12} + C_{Tg2} + C_{bg2} + C_{m23} + C_{Tg1QD2} + C_{Tg3QD2} \quad (13)$$

$$C_3 = C_{Tg3} + C_{bg3} + C_{m23} + C_d \quad (14)$$

The matrix $\mathbf{C}_{node,source}$ is also identified by $\bar{Q}_{node} = \mathbf{C}_{node,source} \bar{V}_{source}$

$$\begin{bmatrix} Q_1 \\ Q_2 \\ Q_3 \end{bmatrix} = \begin{bmatrix} -C_s & -C_{Tg1} & 0 & 0 & -C_{bg1} & 0 \\ 0 & -C_{Tg1QD2} & -C_{Tg2} & -C_{Tg3QD2} & -C_{bg2} & 0 \\ 0 & 0 & 0 & -C_{Tg3} & -C_{bg3} & -C_d \end{bmatrix} \begin{bmatrix} V_s \\ V_{Tg1} \\ V_{Tg2} \\ V_{Tg3} \\ V_{bg} \\ V_d \end{bmatrix} \quad (15)$$

The voltages on the three nodes $\bar{V}_{node}$ (QD1-3) can be found from Eq. (1) to be

$$\bar{V}_{node} = \mathbf{C}_{node,node}^{-1} (\bar{Q}_{node} - \mathbf{C}_{node,source} \bar{V}_{source}) \quad (16)$$

Finally, the electrostatic energy of each charge state as a function of the source voltages is given by

$$U_{el}(N_1, N_2, N_3, V_s, V_{Tg1}, V_{Tg2}, V_{Tg3}, V_{bg}, V_d) = \frac{1}{2} \bar{V}_{node} \mathbf{C}_{node,node} \bar{V}_{node} \quad (17)$$

where $Q_i = -eN_i$, $e = 1.6 \times 10^{-19}$ C being the electronic charge. In the calculation of the stability diagrams in the Letter we set $V_d = V_s = 0$ V. Furthermore, the backgate voltage is kept constant at $V_{bg}^{theory} = 0$ V. This is not the same value as in the experiment $V_{bg}^{exp} = 4$ V, but it yields the same stability diagram (except shifting) and is chosen so only states with few electrons are needed in the calculation. The stability diagrams are calculated numerically by finding the electrostatic energy for each charge configuration with a certain set of gate voltages. A point in the stability is obtained when the two lowest charge states are degenerate. In gate voltage ranges with only one charge ground-state (non-degenerate), a stable charge configuration exists indicated in some regions on the stability diagrams by the electron filling $N_1 N_2 N_3$.

Important energies and voltages

We also like to deduce characteristic energies like the charging energy of each QD U_{ci} , $i = 1, 2, 3$ and the electrostatic coupling energies U_{cm12} and U_{cm23} between QD1 and QD2, and QD2 and QD3, respectively. Furthermore, the change in gate voltages needed to add an electron in the different QD is also derived. These formulas are used to determine the 12 capacitance used in this model.

The electrochemical potentials for adding the N_i electron to QD i are defined as follows with $i = 1, 2, 3$ given in the corresponding line

$$\mu_1(N_1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) = U(N_1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) - U(N_1 - 1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) \quad (18)$$

$$\mu_2(N_1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) = U(N_1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) - U(N_1, N_2 - 1, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) \quad (19)$$

$$\mu_3(N_1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) = U(N_1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) - U(N_1, N_2, N_3 - 1, V_{Tg1}, V_{Tg2}, V_{Tg3}) \quad (20)$$

The addition energy (charging energy, we here ignore shell structure for simplicity) for QD i is given by

$$U_{c1} = \mu_1(N_1 + 1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) - \mu_1(N_1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3})$$

and similarly for QD2 and QD3 giving

$$U_{c1} = e^2 \gamma (C_2 C_3 - C_{m23}^2) \quad (21)$$

$$U_{c2} = e^2 \gamma C_1 C_3 \quad (22)$$

$$U_{c3} = e^2 \gamma (C_1 C_2 - C_{m12}^2) \quad (23)$$

$$\gamma = 1/(C_1 C_2 C_3 - C_3 C_{m12}^2 - C_1 C_{m23}^2) \quad (24)$$

For $C_{m12}, C_{m23} \ll C_{1,2,3}$, which is the case in this system, the expressions can be simplified to

$$U_{c1} \simeq \frac{e^2}{C_1} \left(1 - \frac{C_{m23}^2}{C_2 C_3}\right) \quad (25)$$

$$U_{c2} \simeq \frac{e^2}{C_2} \quad (26)$$

$$U_{c3} \simeq \frac{e^2}{C_3} \left(1 - \frac{C_{m12}^2}{C_1 C_2}\right) \quad (27)$$

i	$ \Delta V_i \text{ [mV]} $	i	$U_{ci} \text{ [meV]}$	i	$t_i \text{ [meV]}$
Tg1	14	1	6.0	-	-
Tg2	3019	2	3.5	-	-
Tg3	22	3	12.0	-	-
Tg1QD2	15	m12	0.26	12	0.3
Tg3QD2	26	m23	≤ 0.05	23	≤ 0.03

TABLE I: Table showing the top-gate voltage change to add one electron in the different QDs, charging and coupling energies, and the tunnel couplings (see section S2) used in the model.

as given in the Letter. We also like to find the electrostatic coupling energy between QD1-2 (and QD2-3), which states how much the electrochemical potential in QD1 (QD2) changes when adding an electron to QD2 (QD3) or viceversa

$$U_{cm12} = \mu_1(N_1, N_2 + 1, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) - \mu_1(N_1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) = \gamma C_3 C_{m12}$$

$$U_{cm23} = \mu_3(N_1, N_2 + 1, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) - \mu_3(N_1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) = \gamma C_1 C_{m23}$$

For general formulas including direct capacitance between QD1 and QD3 (in our model this capacitance is zero) and more compact notation, see Ref.² (no backgate capacitances).

The change in gate voltage $\Delta V_{j(i)}$ on a gate top-gate j corresponding to the addition of an electron to QD i with $j = Tg1, Tg2, Tg3$ and $i = 1, 2, 3$ is found below. Here the subscript i is only written if the quantum dot is different from the top-gate j . The voltage change on Tg1 needed to add an electron in QD1 is found by solving the following equation for ΔV_{Tg1}

$$\mu_1(N_1 + 1, N_2, N_3, V_{Tg1} + \Delta V_{Tg1}, V_{Tg2}, V_{Tg3}) = \mu_1(N_1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) \quad (28)$$

From this we obtain ΔV_{Tg1} and in similar fashion for QD2 and QD3 with respect to Tg2 and Tg3

$$\Delta V_{Tg1} = \frac{e(C_2 C_3 - C_{m23}^2)}{C_{Tg1}(C_2 C_3 - C_{m23}^2) + C_3 C_{Tg1QD2} C_{m12}} \quad (29)$$

$$\Delta V_{Tg2} = \frac{e}{C_{Tg2}} \quad (30)$$

$$\Delta V_{Tg3} = \frac{e(C_1 C_2 - C_{m12}^2)}{C_{Tg3}(C_1 C_2 - C_{m12}^2) + C_1 C_{Tg3QD2} C_{m23}} \quad (31)$$

For no cross capacitances all three expressions reduce to a simple form. Because of the cross capacitances between Tg1/3 and QD2, it is also possible to add an electron in QD2 by tuning either Tg1 or Tg3. The change in gate voltage needed to add an electron to QD2 with Tg1 and Tg3 are found by

$$\mu_2(N_1, N_2 + 1, N_3, V_{Tg1} + \Delta V_{Tg1QD2}, V_{Tg2}, V_{Tg3}) = \mu_2(N_1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) \quad (32)$$

$$\mu_2(N_1, N_2 + 1, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3} + \Delta V_{Tg3QD2}) = \mu_2(N_1, N_2, N_3, V_{Tg1}, V_{Tg2}, V_{Tg3}) \quad (33)$$

giving

$$\Delta V_{Tg1QD2} = \frac{eC_1}{C_1 C_{Tg1QD2} + C_{Tg1} C_{m12}} \quad (34)$$

$$\Delta V_{Tg3QD2} = \frac{eC_3}{C_3 C_{Tg3QD2} + C_{Tg3} C_{m23}} \quad (35)$$

The values obtained in the model is given in Table I by using the capacitances stated in the Letter.

S2. Tunnel coupling and electrostatics

The tunnel coupling between the three quantum dots are included in the model by first order tunneling processes. This means that the charge states calculated above are no longer good quantum numbers (close to crossings), and

electron are tunneling to states which are superpositions of being in the three quantum dots. The hamiltonian for the system in the simplest approximation (disregarding shell structure, spin and tunneling to the leads) can be written as

$$H = H_{el} + H_t \quad (36)$$

The term H_{el} is the electrostatic part calculated above with all matrix elements zero except the diagonal elements with respect to the basis set $\{|N_1 N_2 N_3 \rangle\}$ given by

$$\langle N_1 N_2 N_3 | H_{el} | N_1 N_2 N_3 \rangle = U_{N_1 N_2 N_3}$$

where $U_{N_1 N_2 N_3}$ is the electrostatic energy written in short (see Eq. 17). The second term H_t includes first order tunneling processes between charge states in the triple QD

$$H_t = t_{12}(c_1^\dagger c_2 + c_1 c_2^\dagger) + t_{23}(c_2^\dagger c_3 + c_2 c_3^\dagger)$$

where c_i and $c_i^\dagger$ are the annihilation and creation operators for QD i and we assume t_i , $i = 12, 23$ to be real. A matrix element between two charge states is therefore $t_{12}(t_{23})$, if the first charge state can be obtained from the second by one tunnel event from QD1(QD2) to QD2(QD3) or viceversa, e.g., $(100) \leftrightarrow (010)$ giving $\langle 100 | H_t | 010 \rangle = t_{12}$ or $(010) \leftrightarrow (001)$ giving $\langle 010 | H_t | 001 \rangle = t_{23}$.

The hamiltonian is now written in matrix representation in the basis of the charge states $|N_1 N_2 N_3 \rangle$ in increasing order of total $N = N_1 + N_2 + N_3$, i.e., $\underbrace{\{|000 \rangle\}}_{N=0}, \underbrace{\{|100 \rangle, |010 \rangle, |001 \rangle\}}_{N=1}, \underbrace{\{|200 \rangle, \dots, |002 \rangle, |003 \rangle, \dots\}}_{N=2}$. The matrix

therefore becomes

$$\begin{pmatrix} U_{000} & 0 & & & 0 & & \dots \\ & U_{100} & t_{12} & 0 & & & \dots \\ 0 & t_{12} & U_{010} & t_{23} & & & \dots \\ & 0 & t_{23} & U_{001} & & & \dots \\ & & & & U_{200} & t_{12} & 0 & 0 & 0 & 0 \\ & & & & t_{12} & U_{110} & t_{12} & t_{23} & 0 & 0 \\ & & & & 0 & t_{12} & U_{020} & 0 & t_{23} & 0 \\ 0 & & 0 & & 0 & t_{23} & 0 & U_{101} & t_{12} & 0 & \dots \\ & & & & 0 & 0 & t_{23} & t_{12} & U_{011} & t_{23} \\ & & & & 0 & 0 & 0 & 0 & t_{23} & U_{002} \\ \dots & & \dots & & & & \dots & & & \dots \end{pmatrix} \quad (37)$$

The eigenvalues $\epsilon_i = \epsilon_i(N_1, N_2, N_3, V_s, V_{g1}, V_{g2}, V_{g3}, V_{bg}, V_d)$ of this matrix yield the new ground and excited state energies of the many-particle system including electrostatics and first order tunneling within the triple dot as a function of electron number and gate voltages. Each block corresponds to a total filling N and has $(N+1)(N+2)/2$ charge states. Therefore the same number of eigenvalues are obtained, one of them being the new ground-state energy. The stability diagram is calculated numerically in the same way as described in the previous section. When the ground-state energy for $N+1$ electrons in the triple quantum dot is equal to the ground-state energy for N electrons, transport can occur illustrated by a point in the stability diagrams at the corresponding gate voltages. The stability diagram therefore only takes into account transport involving transitions between ground-states and not transitions involving excited states, a topic, which would be very interesting to study in triple quantum dots. Furthermore, the current through the triple quantum dot^{3,4} is not predicted by this simple model. We also note, that the stability diagrams found by this method for simplicity only considers spinless particles and no shell structure justified by the relative good agreement with the data.

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