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Observation of peak splitting in the Coulomb blockade regime of a carbon nanotube rope ☆

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Abstract

Electronic transport measurements have been carried out on a single-walled carbon nanotube (SWCN) rope contacted to a 4-probe Au/Pd electrode in the Coulomb blockade regime. With varying substrate backgate voltage, the observed Coulomb blockade peaks exhibit interesting three-way splitting. We find that this peak splitting can be attributed to a contribution from resonant tunnelling through discrete energy levels of a finite length metallic SWCN within the rope. We also consider the role that interactions between 'quantum dot' (Q-dot) regions within the rope can play in causing the peak splitting. © 2001 Elsevier Science B.V. All rights reserved.

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Carbon nanotubes [1–3] are well-defined mesoscopic systems making them ideal for comparison between the predictions of theoretical models and experiment. For example, band structure calculations performed for single-walled carbon nanotubes (SWCN) agree well with measurements of their DOS [4]. They are not prone to the effects of disorder which can obscure attempts to interpret electronic transport data for other systems such as the high-T_c cuprates or conducting polymers. Various facets of condensed matter theory can be explored; for example, their one-dimensionality lends then to the investigation of Luttinger liquid behaviour [5] while the ability to control the spin state of a carbon nanotube allows Kondo phenomena to be explored [6]. Previously, semiconductor quantum dots (Q-dots) have filled the role of 'toy model' for studying quantum mechanical behaviour in mesoscopic systems where, for example, artificial atoms and molecules can be created and their measured properties compared with theory. In this regard, carbon nanotubes share similarity with Q-dots [6,7] and, as such, various experiments which have been carried out on Q-dots lend themselves to also being carried out on carbon nanotubes.

In this paper, we focus on electronic transport properties of a metallic SWCN rope in the Coulomb blockade regime. To measure the electronic transport properties, the SWCN rope is typically contacted to a 4-probe electrode which has been fabricated lithographically onto an SiO_2 substrate. If the contact resistance between the electrodes and SWCN rope is high ($\gtrsim 1$ MΩ), the SWCN rope becomes isolated from the current leads and, due to the mesoscopic dimensions involved, the charging energy for a single electron to be transferred onto the rope inhibits the electronic transport through the rope at low temperatures and low bias voltages. With a gate voltage applied to the substrate a single electron transistor (SET) can be achieved [8] and, by varying the gate voltage, the ground state energy of the isolated SWCN rope can be tuned to allow current flow through the SET to be established [8]. Coulomb blockade peaks are then observed periodically in the transport current with varying gate voltage, with each peak corresponding to a single electron being added to the SET.

We report the results of Coulomb blockade behaviour for a SWCN rope deposited onto an Au/Pd 4-probe electrode. As seen by the previous workers, Coulomb blockade peaks are found in the SWCN rope

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current with varying gate voltage [9]. However, in our case, interesting three-way peak splitting is observed which has not been previously reported for an SWCN rope. On the other hand, such peak splitting behaviour has been reported for experiments involving two or three equal sized Q-dots in series [10,11]. The peak splitting arises from interaction between the individual Q-dots; be it the Hubbard interaction [11,12], quantum mechanical tunnelling [13,14] or classical capacitance between the dots [15]. Hence, if our SWCN rope has become fragmented into three separate metallic Q-dot regions along its length, then this could provide a source for the three-way peak splitting.

Interaction between adjacent parallel metallic SWCNs within the rope can also affect the rope's electronic transport properties. In this case the parallel interaction causes symmetry breaking [16,17] which induces a gap in the density of states at the Fermi level, causing the system to become semiconducting. If such symmetry breaking were induced locally at defects along the rope, then this could provide a mechanism for fragmenting the SWCN rope into separate metallic Q-dot regions [16] and so yield the peak splitting we observe.

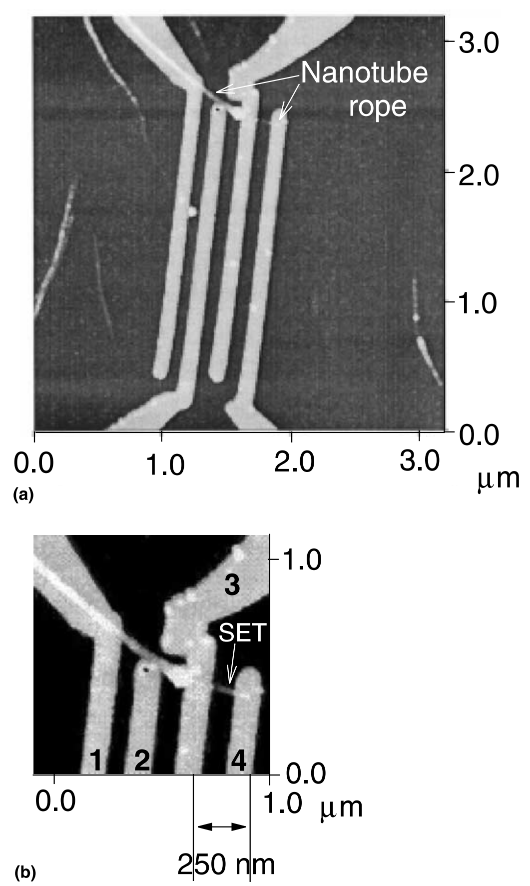

Fig. 1. (a) AFM image of an SWCN rope deposited onto an Au/Pd 4-probe electrode. (b) Emphasizing the section of SWCN rope comprising our SET.

Our method for synthesizing and preparing SWCN samples is well established [18,19]. SWCNs were produced using an arc discharge furnace with Ni/Y as the catalyst. Using transmission electron microscopy, it is found that the individual SWCNs produced typically have diameters of $\simeq$1.5 nm. This is significant since it is expected that SWCNs produced with a diameter in this vicinity will be of the metallic (10, 10) armchair variety [20]. Such a situation is required to achieve an SET comprising a metallic SWCN rope. Our sample with an individual SWCN rope deposited onto an Au/Pd 4-probe electrode is shown in Fig. 1. The SWCN rope is seen near the upper area of the electrodes. We also observe a bulge in the SWCN bundle between the middle two electrodes, presumably due to an Ni impurity from the catalyst material. In terms of measuring the electronic transport properties of our sample, the main characteristic is a contact resistance of the order of 1 MΩ which is applicable for the Coulomb blockade regime. In order to explore SET behaviour we also connect a gate electrode to the rear of the substrate. The labelling scheme for our electrodes is given in Fig. 1(b).

To explore SET behaviour we carry out a 2-probe measurement between electrode pair 3–4. These two electrodes are preferred since the carbon nanotube rope in this region is particularly narrow and without obvious defects. The current versus voltage (I–V) characteristics for the electrode pair 3–4 are given in Fig. 2. At $T = 4.3$ K we observe nonohmic, semiconductor-like behaviour with zero conductivity in the low voltage limit. This is a clear evidence for the Coulomb gap in the energy spectrum of the conduction electrons. At higher temperatures the I–V characteristics become linear, as for a metallic system, with the Coulomb gap becoming thermally obscured. As indicated in Fig. 2, during the measurement process a transient in the electrical circuit

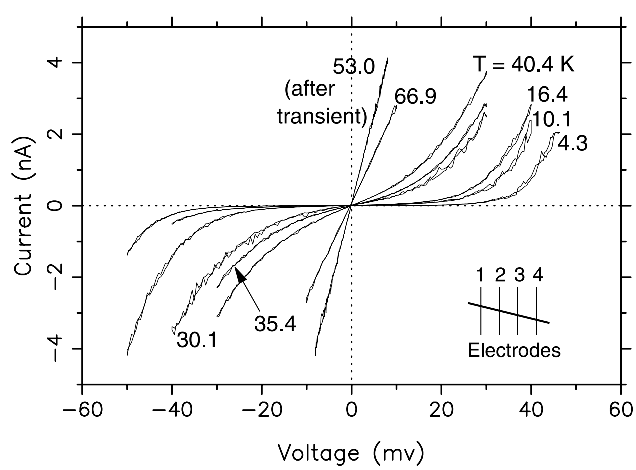

Fig. 2. Electrical characteristics for the SWCN rope of Fig. 1 between the right two electrodes. After the measurement at $T = 66.9$ K a transient occurred in the electrical circuit causing the sample to become slightly more metallic.

has slightly reduced one or both of the contact barriers, with an associated increase in the conductivity of the SET. At the temperature $T = 53$ K, the overall resistance of the SET is $\simeq 2$ MΩ.

The effect of varying the gate voltage, V_G, on the SET current is shown in Fig. 3 for a temperature of $T = 4.2$ K. To measure the current, a source–drain voltage of $V_{DS} = 20$ meV was applied. From the I–V characteristic in Fig. 2 we not that this is sufficiently low such that no significant current will flow when the Coulomb blockade peaks which allow interesting features to be observed within the peak width. With varying gate voltage, we observe clear Coulomb blockade peaks in Fig. 3 with interesting three-way splitting. The peak amplitude in Fig. 3 plotted in units of the quantum conductance, $G_0 = e^2/h$, highlights the low tunnelling probability for our contact barriers; consistent with the Coulomb blockade regime.

For conductance peaks due to pure Coulomb blockade, the peak spacing is periodic in V_G with peak spacing given by the condition [21]

$$Q_0 = C_G V_G = e(n + 1/2), \tag{1}$$

where n is an integer, Q_0 is the charge on the isolated metallic region and C_G is the gate capacitance. The peaks in Fig. 3 display some aspects which diverge from this condition. The peaks are not equally spaced but increase their spacing with increasing gate voltage. This can occur if resonant tunnelling through discrete energy levels of the SET also contributes to the peaks [8]. There is a peak occurring at $V_G = 0$ which is where the Coulomb blockade condition should apply. This is most likely due to an experimental artefact causing an offset in V_G.

The overall width of the Coulomb blockade peaks in Fig. 3 is rather wide compared to the previous works [9,22] to the extent that the peaks encompass most of the available 'V_G-space'. This is partly due to the temperature of $T = 4.2$ K being somewhat high for this type of measurement [9,22]. We also note that our bias voltage of $V_{DS} = 20$ meV, while allowing for negligible current to flow when the Coulomb blockade condition holds, is still rather high; to the extent that it is approaching the apex of the diamond shape which defines a boundary for allowed current flow in (V_G, V_{DS})-space for the Coulomb blockade regime. This implies that the trajectory traced through (V_G, V_{DS})-space with varying V_G will be dominated by regions where the Coulomb blockade condition is lifted, thus yielding the wide Coulomb blockade peaks we observe.

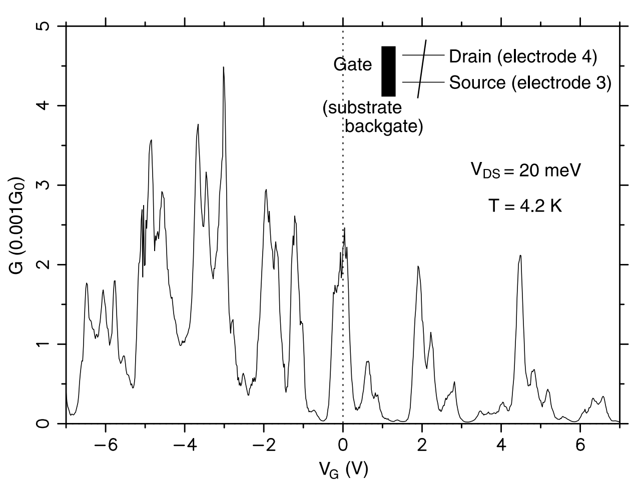

Fig. 3. Variation of the SET current with substrate back-gate voltage.

Regarding the origin of the three-way peak splitting in Fig. 3, one possibility is the effect of fractional charge due to Luttinger liquid behaviour in a metallic SWCN, [23] though, more evidence would be required, for example, from the quantum shot noise measurements [23,24]. Another possibility which can be understood more straightforwardly is that the rope between electrodes 3 and 4 has become fragmented along its length into three metallic regions separated by insulating barriers. The analogous situation of three Q-dots in series readily reveals Coulomb blockade peaks exhibiting three-way splitting [10,11]. The nanotube rope could become fragmented into separate metallic Q-dot regions due to defects or inter-tube contacts within the rope which introduce barriers to metallic conduction since in one dimension such defects cannot be circumvented. Even coupling to the substrate could provide a source of disorder and defects [25].

The degree of peak splitting depends on the amount of interaction between the Q-dots [13], which in turn depends on the size of the intervening barriers. With no inter-dot interaction there is no peak splitting. For the special case of three equal sized Q-dots in series, as the interaction is increased, the degree of three-way peak splitting increases until, with the barriers completely removed, the three sub-peaks become maximally spaced to the extent that they effectively become single Coulomb blockade peaks corresponding to a single Q-dot with size three times the previous individual Q-dots. The associated Coulomb blockade peaks then have a period one-third of the previous.

Given the large degree of peak splitting seen in Fig. 3, we follow Waugh et al. [10] and take the interaction to be due to inter-dot tunnelling. For significant peak splitting, Waugh et al. [10] have established a direct relationship between the fractional peak splitting and the measured barrier tunnelling conductance between two Q-dots in series. From the fractional peak splitting in Fig. 3, we endeavour to estimate the associated barrier size as follows. The relationship between the fractional peak splitting and barrier tunnelling conductance has been estimated theoretically for the case of two Q-dots in series [14] but not the three-dot case. Nonetheless, we surmise that the case of three Q-dots will follow similar behaviour to the two Q-dot case and utilize the results from [26]. From Fig. 3 we estimate an average peak splitting fraction of $\simeq$0.6. From [26], this corresponds to a barrier tunnelling probability of $\simeq$0.8 which, given the exponential dependence of tunnelling probability with barrier size, implies a considerably small barrier. For example, the mechanism suggested by Delaney et al. [16] for creating barriers local to defects due to a symmetry breaking effect would involve a barrier height of $\sim$0.1 eV. In terms of a one-dimensional rectangular barrier, this would give a barrier width of less than 1 Å. With such tiny barriers there is a large degree of coupling between the three supposed Q-dot regions to the extent that we could consider that there is effectively just a single metallic region. Hence, an alternative explanation for the observed peak splitting is next considered.

As well as the quantized momentum in the lateral direction, with finite length, the energy spectrum of a SWCN will also become discretized in the longitudinal direction, as for a one-dimensional square well. For gate voltages such that the Coulomb blockade condition is lifted, as prescribed by Eq. (1), resonant tunnelling will occur through discrete energy states which happen to coincide with the Fermi level. With varying gate voltage, the discrete energy levels of the SET are swept passed the Fermi level resulting in a series of peaks superimposed on the Coulomb blockade peaks. If the resonant tunnelling peak spacing is less than the Coulomb blockade peak width, then this may yield peak splitting as seen in Fig. 3.

To confirm this we first estimate the relationship between varying V_G and shifting the energy levels of the isolated metallic region for the SET. From the electrode characteristics in Fig. 2 we can estimate the energy gap associated with the single electron charging energy, $U = e^2/2C$, plus the quantized level spacing, ΔE, using [9] $V_{th} = U + \Delta E$, where V_{th} is the threshold voltage for current flow. From the I–V curve at $T = 4.3$ K in Fig. 2, the threshold voltage is estimated as $U + \Delta E \simeq 35$ meV. The spacing in the Coulomb peaks is given by [9]

$$\Delta V_G = \frac{U + \Delta E}{e\alpha}, \tag{2}$$

where $\alpha = C_G/C$ with C_G the capacitance between the gate and the isolated metallic region and C the total capacitance between the metallic region and its environment. From Fig. 3 we estimate an average spacing between the major Coulomb blockade peaks of $\Delta V_G \simeq 1.8$ V. Using Eq. (2) and $U + \Delta E \simeq 35$ meV we determine the ratio $\alpha = 0.019$. From Fig. 3 we estimate the average spacing between the supposed resonant tunnelling peaks as $\simeq$0.36 V. The energy level spacing between discrete states is then determined from 0.36 V $= \Delta E/e\alpha$ as $\Delta E = 6.8$ meV. We note that our ratio of U to ΔE is similar to that reported by the previous workers [9,22]. Following Tans et al. [22] ΔE is related to the length of the tube using $\Delta E = hv_F/2L$, where the Fermi velocity is estimated as $v_F = 8.1 \times 10^5$ m/s. We determine a length of $L \simeq 250$ nm. Interestingly, this length agrees well with the length of the particularly narrow section of SWCN rope seen in Fig. 1 which comprises our SET. This length is measured from the bulge which lies partially over electrode 3 to the right-most end of the rope which is situated on electrode 4. This agreement lends support to resonant tunnelling as the cause of the peak splitting in Fig. 3. This also highlights that ballistic transport is occurring within the SWCN rope over this distance. Fig. 4 shows the affect of increasing temperature on the peaks in Fig. 3. As expected, the peaks become broadened as the Coulomb blockade gap becomes thermally obscured and the overall conductance increases with temperature due to the increased number of contributing discrete energy levels becoming encompassed within the Fermi window.

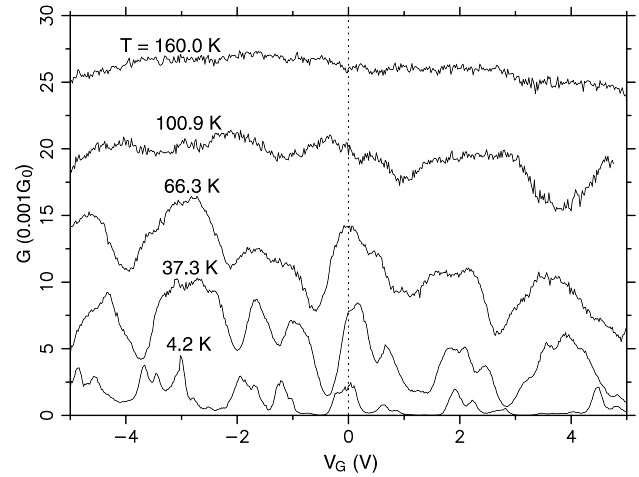

Fig. 4. Demonstrating the effects of temperature on the Coulomb blockade peaks of Fig. 3.

Finally, we re-iterate the observation made by the previous workers [9]. Most likely the entire SWCN rope is not metallic but just a single SWCN within the rope is metallic and having sufficient contact to the electrodes to play an active role in the SET.

Acknowledgements

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