Gold nanoparticle single-electron transistor with carbon nanotube leads

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(Received 5 February 2001; accepted for publication 31 July 2001)

We have used carbon nanotubes to electrically contact a 7 nm gold particle by scanning-probe manipulation. The result was a single-electron transistor showing a periodic modulation of the current as a function of gate voltage for temperatures up to ~200 K, with the particle responsible for the main features of the electron transport. This interpretation could be verified when the particle was removed and the two nanotubes were moved into electrical contact. © 2001 American Institute of Physics. [DOI: 10.1063/1.1405154]

Carbon nanotubes are exceptional molecules, having outstanding electrical and mechanical properties.1 Recent experiments have probed the electrical transport through nanotubes, and it has been shown that metallic nanotubes can function as nanoscale wires.2,3 While the transport properties of nanotubes themselves have been studied in detail, no experiments have so far investigated the transport in structures where nanotubes are actually used as passive leads to contact other nanosized objects. In the present letter, we address this important aspect experimentally.

The mechanical strength and flexibility of nanotubes allow manipulation of single tubes using scanning-probe techniques.4–7 We have utilized this to form a structure where two nanotubes, with lengths greater than 1 μm, were used as leads to contact a single 7 nm gold particle. The transport properties of this combined structure were investigated at various temperatures. In particular, we found single-electron transistor (SET) behavior up to 200 K. The large difference between the length of the tubes and the diameter of the particle means that the transport was mainly governed by the latter. Thus, Coulomb blockade in the tubes, which is only effective below a few K, can be disregarded and their main function was to provide contact between the macroscopic electrodes and the nanoparticle.

Carbon nanotube devices were fabricated by chemical-vapor deposition (CVD) following the method developed by Soh et al.8 Areas with Fe- and Mo-containing catalyst material were patterned by electron-beam lithography on a silicon substrate with a 355 nm thermal oxide top layer.9 In the following CVD step, a methane flow rate of 5200 bar cm³/min was maintained for 10 min at 900 °C. The result is most often individual tubes, with diameters around 1–3 nm and lengths up to several micrometers, spreading out from the catalyst areas. Most tubes appear to be single walled as observed by transmission electron microscopy. The catalyst areas were then covered with an evaporated metal layer consisting of Cr (6 nm) and Au (15 nm). This layer extended 3 μm beyond the catalyst areas, resulting in electrodes separated by 1 μm. The doped silicon substrate was used as a third, back-gate electrode.

Figure 1(a) shows a part of the 3-nm-thick, individual carbon nanotube that was used in the experiment. The initial resistance of the tube—including contact resistance—was 21 kΩ and showed no gate dependence at room temperature, i.e., the tube was metallic. When the tube was strained with the tip of an atomic-force microscope (AFM), it began to slide under one of the electrodes, until approximately 1 μm of the tube had been pulled out. (This is often observed for different kinds of tubes and electrode metals. For details of AFM manipulation, see Ref. 10.) In order to cut the tube, and to prevent further sliding, a certain strain in combination with a sharp angle was required. These conditions could be realized by first shaping the tube into a loop [Fig. 1(b)] and then positioning the AFM tip inside this loop. A very sharp angle was created as the tip strained the loop and the tube was finally cut [Fig. 1(c)]. When the two parts were put back into contact, resistances down to 500 kΩ were observed. The resistance seemed to depend on the contact area between the tubes, on the force that was used to press the tubes together, and also on what parts of the tube that were connected. The
last may have been due to local contamination of the tubes by residues from the processing. Finally, the area around the tubes was thoroughly mapped with the AFM. The sample was then cooled to 4.2 K. Figure 2(a) is a differential conductance plot (dI/dV_{SD}) as a function of both bias voltage (V_{SD}) and gate voltage (V_G) for the system. The plot shows a regular pattern of diamond-shaped areas with zero conductance. These are regions of Coulomb blockade corresponding to a charging energy \( E_C = 60 \text{ meV} \) of 60 \( \pm 5 \) meV. The plot suggests that one small island controlled the transport in the system. This island coupled rather weakly to the gate, as only six blockade regions show up for a span of 12 V on the gate. However, there is also a fine structure present that has a much stronger coupling to the gate [Fig. 2(b)]. This structure has a smaller energy scale and may be interpreted as charging effects in larger islands. The diamond patterns give directly the gate coupling, \( \alpha = C_G / C_S \), where \( C_G \) is the capacitance between the island and the gate and \( C_S \) is the total capacitance of the island. With 2\( \Delta V_{SD} \) as the vertical diagonal in a diamond and \( \Delta V_G \) being the horizontal diagonal, we have \( \alpha = \Delta V_{SD} / \Delta V_G \). This results in \( \alpha \approx 0.03 \) for the large diamonds [Fig. 2(a)], while the smaller structure [Fig. 2(b)] corresponds to \( \alpha \approx 0.3 \), i.e., a ten times stronger coupling. Based on these observations, we attribute the larger blockade structure to the particle, and the complicated fine structure to single-electron charging effects in the nanotube leads. This would be consistent with the difference in gate coupling, as \( C_G / C_S \) should be larger for a one-dimensional tube section compared to a zero-dimensional particle with the current relative dimensions.

A charging energy of \( E_C = 60 \text{ meV} \) for the larger blockade areas is in good agreement with an estimation of \( E_C = e^2 / 2C_S = 68 \text{ meV} \), resulting from the self-capacitance, \( C_S = 2 \pi \varepsilon_0 \varepsilon_r D \), of a sphere with a diameter \( D = 7 \text{ nm} \) (estimated dielectric constant \( \varepsilon_r \approx 3 \)). The bright diagonal stripes in Fig. 2(a) surround areas with constant charge on the smallest island in the system, and are indications of a strong asymmetry in the resistance of the two tunnel junctions that connect this island. Since the sum of the two tunnel resistances connecting the particle was 50 M\Omega, while the upper limit for the total resistance of the nanotube leads was only 500 k\Omega, this observation supports the suggestion that the gold particle, rather than an intratube section, was the smallest island.

The sample was then slowly warmed to room temperature. Figure 2(c) shows that oscillations in the current as a function of gate voltage persisted for temperatures up to around 200 K. The sample was transferred back to the AFM to resume the manipulation. First, the particle was carefully pushed aside, and then the two nanotube parts were moved into contact at the point where the particle had been pushed aside, and then the two nanotube parts were moved into contact at the point where the particle had been [Fig. 1(f)]. At a resistance of 50 M\Omega, the sample was once more removed from the AFM and cooled to 4.2 K. A dI/dV_{SD} plot, shown in Fig. 3(a), was recorded, but no periodic features can be observed on the scale of the blockade areas in Fig. 2(a). Instead, the plot reveals many areas with negative diff.
The gold particle can be replaced with a magnetic, a semiconducting, or a superconducting particle, or perhaps a molecule. However, interpretation of data obtained from low-temperature studies of such systems will require a more-detailed understanding of the transport in the nanotube leads.

This work has been supported by grants from the Natural Science and the Engineering Science Research Councils in Sweden, the Swedish Foundation for Strategic Research, and the Danish Natural Science Foundation.