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Fabrication and electrical properties of a carbon nanotube quantum dot*

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Single-walled carbon nanotubes (SWNTs) were synthesized by pyrolyzing methane (CH_4) at a temperature of 900°C on SiO_2 substrates pre-coated with iron nano-particles. Electrical contacts were fabricated onto one of the SWNTs by using an electron beam lithography process. Coulomb blockade and single-electron tunnelling characters were found at low temperatures, indicating that the SWNT in-between the electrodes forms a quantum dot. It is found that the Coulomb gap of the quantum dot is about 8.57 meV, and the factor α, which converts the gate voltage to the true electrostatic potential shift, is around 200 for this device.

Keywords: quantum dot, carbon nanotube
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1. Introduction

Carbon nanotubes have been widely studied due to their outstanding physical properties and important potential applications. One of the aimed applications is to fabricate single-electron devices. Generally, carbon nanotubes can be synthesized by arc discharge,[1] laser ablation,[2] and CVD process.[3−7] In some of the methods, in order to obtain purified individual nanotubes, one has to treat the product afterwards with highly oxidative chemicals that are known to create defects and alter their electronic properties, which would prevent carbon nanotubes from many applications. Efforts have been made to avoid the post purification and dispersion processes, such as to grow carbon nanotubes directly onto Si substrates.[3,4,6,7] In this paper we report our investigation on this kind of approach, and show that individual carbon nanotubes with no amorphous carbon on the wall can be obtained. Then, we fabricate an electronic device using one carbon nanotube.

2. Fabrication of a quantum dot

In our experiment, the carbon nanotubes were grown by chemical vapour deposition, using CH_4 as the feeding gas and iron nanoparticles as the catalyst spread on Si substrates through a wet chemical route.[4] The density of the iron nanoparticles was carefully controlled in order to realize a sparse growth of nanotubes on the substrates. The catalyst was prepared as follows. One milligram of $Fe_2(SO_4)_3$ powder was added into 40 ml of isoprophylalcohol (IPA) solvent and was ultrasonicated for several minutes to ensure that the powder is fully dissolved in the solvent. Silicon substrates with a 500 nm thick silicon dioxide layer were dripped into the solution for a few seconds, and then taken out. After the solution was dried, the SiO_2/Si substrates were placed in a quartz boat and put into the centre of a quartz tube of 3.7 cm in diameter and 120 cm in length located in the middle of a high-temperature cylindrical tube furnace. The furnace was heated up in an argon gas flow at a rate of 750 ml/min. The $Fe_2(SO_4)_3$ nanoparticles on the substrates decomposed into Fe_2O_3 when the temperature reached about 750°C. Then, hydrogen gas was introduced into the quartz tube at a flow rate of 200 ml/min for 15 min. During this treatment the Fe_2O_3 reacted with H_2 to produce Fe nanoparticles. Carbon nan-

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otubes were then grown on Fe-coated substrates via pyrolysis of CH_4 gas flowing at a rate of 1.5 l/min and 900°C. The growth process lasted for 10μ,. Then the tube furnace was cooled down slowly in an argon gas ambient.

Figures 1(a) and 1(b) are the scanning electron microscopy (SEM) images of the carbon nanotubes grown on Si substrates. The typical length of the nanotubes is about ten microns. Figure 1(a) shows the case of high density growth, in which the carbon nanotubes form a network. Figure 1(b) gives an example of low density growth using low concentration solution. The density of Fe-catalyst nanoparticles on Si/SiO_2 substrates is of critical importance for determining the population of carbon nanotubes on the substrates. In our experiment the density of Fe-catalyst particles was controlled by using different concentration of $Fe_2(SO_4)_3$ /IPA solution.

Figure 1(c) shows a HRTEM image of a SWNT whose diameter is about 2 nm. It is an individual tube, with no glassy carbon covering its wall. In our experiment we also see SWNT bundles and even multiwalled carbon nanotubes of several nanometres in diameter.

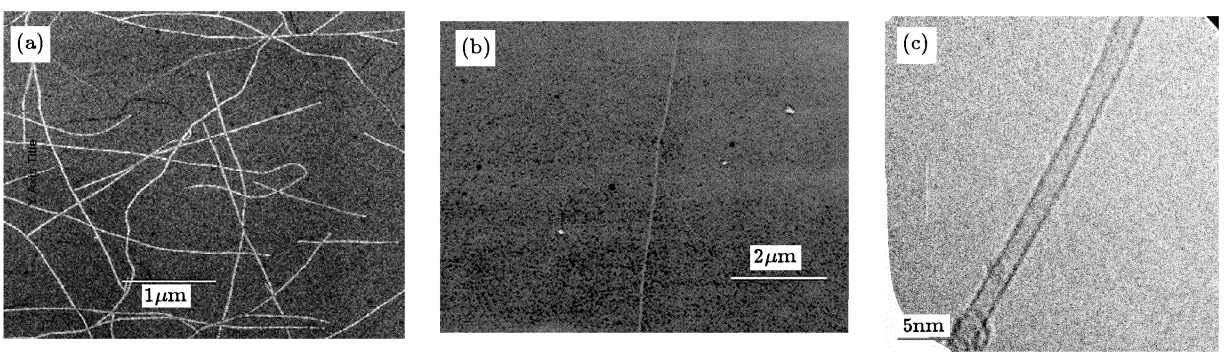


Fig.1. (a) and (b): SEM images of carbon nanotubes grown on Si/SiO_2 substrates. The population of carbon nanotubes on the substrates can be controlled by the density of catalyst particles, or, by the concentration of $Fe_2(SO_4)_3$ in IPA solution. (a)The ratio of $Fe_2(SO_4)_3$ /IPA is 5 mg/50ml; (b) the ratio of $Fe_2(SO_4)_3$ /IPA is 1 mg/50ml. (c) High resolution TEM (HRTEM) image of a SWNT.

Using a mixture of powders of $Fe_2(SO_4)_3/Al_2O_3$ would increase the yield of SWNTs,[4] which is also true in our case. However, it is difficult to control the Al_2O_3 particle density on SiO_2 surface, since Al_2O_3 is insoluble in IPA. Adding Al_2O_3 powder may also dirty the surface and afterwards affect the process of electrode fabrication.

We then fabricated electrodes onto one of the carbon nanotubes that were sparsely grown on Si/SiO_2 substrates, by using electronic beam lithography technique. The electrodes contain 10 nm of Ti and 50 nm of Au layers. The Ti layer provides good adhesion on the SiO_2 surface. It also helps to obtain ohmic contact with carbon nanotube.[6] The degenerate doped silicon substrate under the SiO_2 surface layer (500 nm in thickness) was used as a backgate. The device, in which the carbon nanotube in-between the electrodes can presumably be regarded as a quantum dot, is shown in Fig.2(a).

3. The transport properties of the quantum dot

Figure 2(b) shows the typical bias voltage dependence of the differential conductance $\mathrm{d}I/\mathrm{d}V$ at several different temperatures. The $\mathrm{d}I/\mathrm{d}V$ curves near zero bias voltage are suppressed at low temperatures, indicating the formation of a Coulomb gap. When the contact resistance of the electrodes is larger than the quantum unit (R_Q=26.5kΩ), the carbon nanotubes in-between the two electrodes can be regarded as a quantum dot with a very small electrical capacitance C, which yields a relatively large Coulomb charging energy $e^2/2C$. Therefore, the electron conduction within the gap will be blocked when the thermal fluctuation is small: i.e., $k_BT << e^2/2C$. In this case, the current dependence of bias voltage can be expressed as

$$V = R_T I + \frac{e}{2C}, \qquad (1)$$

where V is the voltage, I is the current, R_T is resistance; and the existence of an offset voltage $\frac{e}{2C}$ is the fingerprint of the Coulomb gap.[8] Figure 2 (c) shows the $I-V$ curves of our device measured at different temperatures. At high temperatures, the curves are almost straight lines passing through the (0, 0) point. At low temperatures, however, the $I-V$ curves at

high bias voltages seem to be shifted horizontally because of the existence of the Coulomb gap. For the curve taken at 1.9 K, as shown in the inset of Fig.2(c), the shift is 8.57 mV.

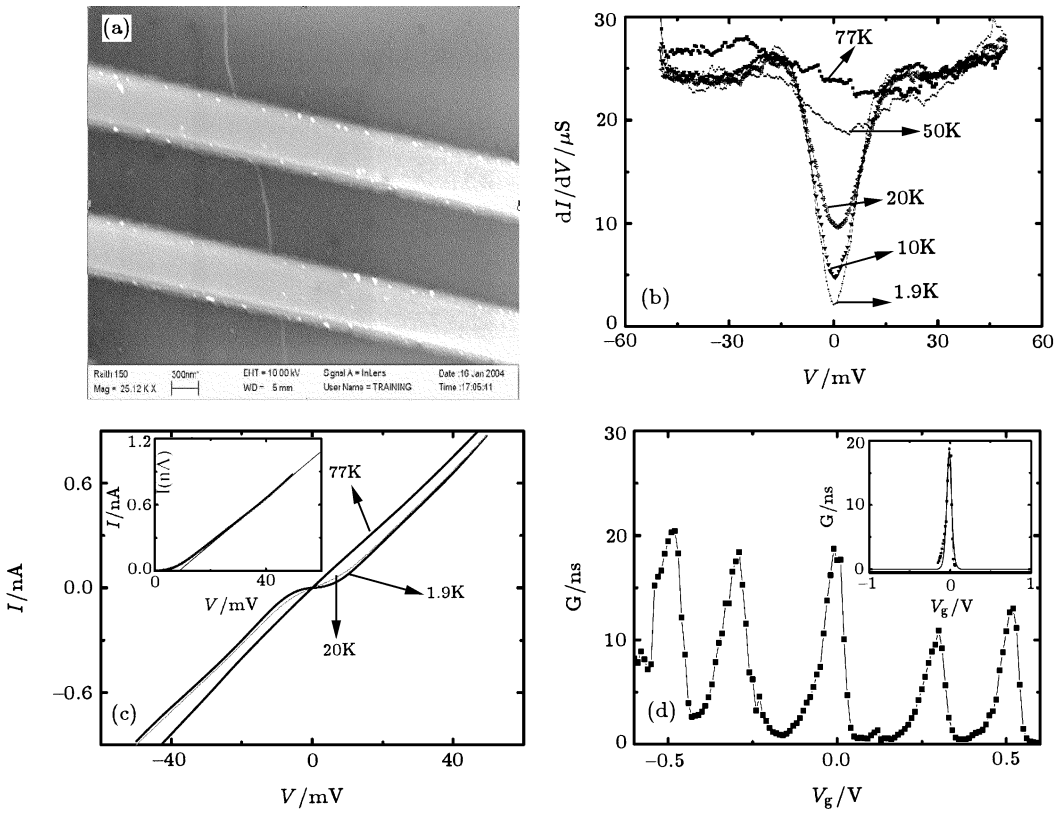


Fig.2. (a) SEM image of a nanotube on Si/SiO_2 substrate with two electrodes attached. (b) The dI/dV curves measured at several different temperatures. A zero-bias suppression in the conductance curves can be seen at low temperatures, indicating the formation of a Coulomb gap. (c) $I-V$ curves measured accordingly at different temperatures. The Coulomb gap causes a horizontal offset to the data points at high bias voltages, by fitting which one can obtain the value of the gap (the inset). (d) Conductance oscillation as a function of the gate voltage. Inset: the conductance peak near zero bias is fitted to Eq.(2).

In Fig.2(d) the conductance is plotted as a function of the backgate voltage applied to the degenerate doped Si substrate. Conductance oscillation can be observed while sweeping the backgate voltage. It is known that the energy band in the carbon nanotube quantum dot is split to discrete levels. Between the highest occupied level and the lowest unoccupied level there is a Coulomb gap, which blocks the electron conduction if the bias window of the two electrodes is located within this gap. The backgate voltage can shift the discrete energy levels in the nanotube quantum dot upwards or downwards. When one of the discrete levels is shifted into the bias window, resonant tunnelling occurs, leading to a conductance peak. Further varying the backgate voltage will shift the discrete energy level out of the bias window, therefore the electron conduction will be blocked again. That is why the conductance oscillates with the gate voltage, as shown in Fig.2(d).

Assuming that a single energy level is weakly coupled to the two electrodes, we can fit the conductance peak near the zero backgate voltage by using the following formula:[9]

$$G \propto \cosh^{-2}(e\Delta V_g/\alpha 2k_B T), \qquad (2)$$

where G is the differential conductance, ΔV_g denotes the gate voltage shift, and α is a factor describing the efficiency of how ΔV_g is converted to the true electrostatic potential shift of the electrons in the carbon nanotube quantum dot.[10] The fitting, as shown in the inset of Fig.2(d), yields an α of about 200. A smaller α can be obtained by reducing the thickness of the SiO_2 layer.

4. Conclusion

In summary, SWNTs have been synthesized by a chemical vapour deposition process using iron nanoparticles as the catalyst. HRTEM observation

reveals that the diameter of SWNTs is about 2 nm and there is no glassy carbon covering the wall of the tube. The nanotube population on Si substrates can be controlled by adjusting the $Fe_2(SO_4)_3$ powder concentration in the catalyst solution. We have also fabricated a nanodevice using the carbon nanotube thus synthesized. The carbon nanotube in the device behaves like a quantum dot, with a Coulomb charging energy of 8.57 meV and a factor α of about 200, which converts ΔV_g to the true electrostatic potential shift.

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