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著者	畠山 力三
journal or publication title	IEEE Conference on Nanotechnology, 2007. IEEE-NANO 2007. 7th
volume	2007
page range	175-179
year	2007
URL	http://hdl.handle.net/10097/46677

doi: 10.1109/NANO.2007.4601165

Proceedings of the 7th IEEE
International Conference on Nanotechnology
August 2 - 5, 2007, Hong Kong

Resonance Tunneling Transistors Based on C_{60} Encapsulated Double-Walled Carbon Nanotubes

Y.F. Li*, T. Kaneko, and R. Hatakeyama,

Department of Electronic Engineering, Tohoku University, Sendai 980-8579, Japan

Abstract—**We report electrical transport properties of resonance tunneling transistors fabricated using C_{60}-filled metallic double-walled carbon nanotubes. All the examined devices exhibit a strong negative differential resistance (NDR) behavior, and the high peak-to-valley current ratios more than 10^3 are observed at room temperature. More importantly, the observed NDR characteristics remain stable under forward and backward measurements. In addition, it is found that the applied gate voltages exercise a great influence on the peak voltage of NDR.**

Keywords — carbon nanotubes, encapsulation, fullerene, negative differential resistance

I. INTRODUCTION

Over the past ten years, great interest in nanostructured carbons, such as fullerene C_{60}, single-walled carbon nanotubes (SWNTs), and double-walled carbon nanotubes (DWNTs) has been aroused steadily due to many of their unique properties in nanoscale and potential applications in lots of fields. Specially, DWNTs consisting two concentric cylindrical graphene layers have attracted increasing interests owing to their advantages over other types of carbon nanotubes in size, electrical and mechanical properties [1]. Recently, with their unique combination of physical properties, fullerenes and carbon nanotubes are currently considered to be likely candidates for functional building blocks of nanoscale devices, and electronic transports in these structures prove to be strongly affected by the single-electron charging and quantization of energy levels. Up to now, various nanodevices with unusual transport properties, such as single C_{60} transistor, carbon nanotube-field effect transistors (FETs), single-electron transistors, and rectifying diodes have been reported. However, the experimental work on making resonant tunneling diodes (RTDs) and resonant tunneling transistors (RTTs) constructed with carbon nanotubes is only beginning to emerge, and also some theoretical predications [2] remain to be tested.

Our recent experiments indicate that RTTs with NDR behavior can possibly be fabricated by encapsulating the fullerene C_{60} into DWNTs with large inner and outer diameters. In our case, numerous fullerenes encapsulated in one-dimensional (1D) DWNTs may provide an ideal superlattice heterostructure for testing quantum mechanism effects. Here we have examined transport properties of metallic DWNTs in which C_{60} molecules are encapsulated, and find that they can exhibit a distinct room temperature NDR behavior compared with the case of empty DWNTs. High peak to valley current ratios over 10^3 are significantly observed in most of examined devices.

This work was supported by Tohoku University 21st Century Center of Excellence (COE) Program, JSPS-CAS Core-University Program on Plasma and Nuclear Fusion, and Research Fellowship of JSPS.
**Contact author: yfli@plasma.ecei.tohoku.ac.jp*

II. EXPERIMENTAL

The synthesis of C_{60} molecules encapsulated DWNTs can be realized by either a vapor reaction method or a plasma ion irradiation method [3]. Pristine DWNTs are prepared by an arc discharge method with Fe as catalyst. Transmission electron microscopy (TEM) observations and Raman spectra have confirmed that most pristine DWNTs have uniform inner diameter of 4.0 nm, outer diameter of 4.8 nm, and higher purity than 99%. For the plasma irradiation method, DWNTs were dispersed by brief supersonic treatment in ethanol. Then, droplets of this suspension were dripped and dried on the stainless steel substrates (15 mm × 15 mm). This DWNTs substrate is installed in a magnetized alkali-fullerene plasma column (B = 0.2 T). The experiment on fullerene encapsulation inside carbon nanotubes is carried out using the plasma ion irradiation method. The details on the production of alkali-fullerene plasmas used here are described elsewhere [4]. As a result, we use throughout in this study a low-temperature ion plasma of $n_p \sim 1 \times 10^{10}$ cm^{-3} (n_p: plasma density measured by a Langmuir probe). When the positive dc bias (ϕ_{ap} = 20 V) is applied to the substrate with respect to a grounded plasma-source electrode, negative fullerene ions are substantially accelerated by a plasma sheath in front of the substrate and finally bombard the carbon nanotubes. Such a plasma irradiation process is performed for 1 hour.

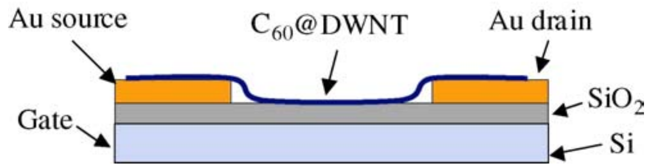

Fig.1 A schematic diagram for a C_{60}-filled DWNT-based nanodevice

Raw C_{60}-filled DWNT samples are obtained by the above plasma irradiation process, and then sonicated in toluene in order to remove the excess exterior C_{60} powder attached to the outer side of DWNTs. The purified samples are examined in detail by TEM (Hitachi HF-2000) operated at 200 kV and Raman spectroscopy (Jovin Yvon T-64000) with Ar laser at

488 nm. The resonance tunneling device fabrication using C_{60}-filled DWNTs undergoes a typical process of making nanotube FETs, as described in our previous work [5-7]. Source and drain electrodes used are made of Au placed on a SiO_2 insulating layer (thickness of 500 nm) and have a channel length of 500 nm. A heavily doped Si substrate is used as a back gate, as schematically shown in Fig. 1. The purified C_{60}-filled DWNTs are firstly dispersed by sonication in *N, N*-dimethylformamide (DMF) solvent and then spincoated on a substrate where 63 pairs of Au electrodes are pre-prepared. After that, a baking process at 400 K is finally carried out in atmosphere for 30 min to remove DMF remnants. Electrical transport properties of the devices are measured using a semiconductor parameter analyzer (Agilent 4155C) in vacuum at room temperature.

III. Results and Discussions

A TEM image for an empty DWNT with an outer diameter of about 4.8 nm and inner diameter about 4 nm is shown in Fig. 2(a). In contrast, Fig. 2(b) presents a typical TEM image for a DWNT, which is filled with numerous C_{60} molecules. Owing to the large inner diameter of DWNT, only an amorphous morphology of C_{60} molecules inside the DWNT is observed.

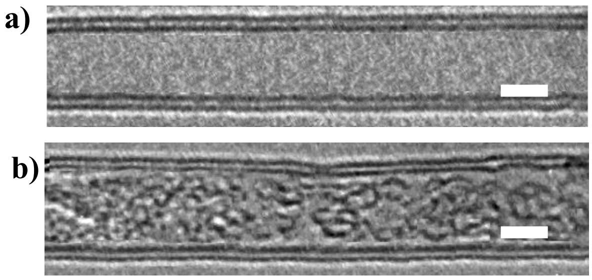

Fig. 2 a) TEM image of an empty DWNT with inner diameter of 4.0 nm and outer diameter of 4.8 nm; b) that of C_{60}-filled DWNT in which amorphous C_{60} molecules are found (Scale bar: 2 nm).

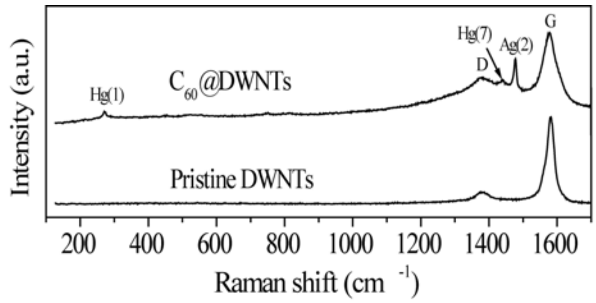

Fig. 3 Raman spectra for pristine DWNTs and C_{60}-encapsulated DWNTs.

In addition, the measured Raman spectra, as shown in Fig. 3, have further confirmed that C_{60} molecules have been filled inside the DWNTs. Due to their larger diameter, the radial breathing mode (RBM) band is not detected for both the pristine and C_{60}-filled DWNTs. However, great signal changes can be recognized on the sample of C_{60}-filled DWNTs, and two distinct peaks between D-band (1378 cm^{-1}) and G-band (1584 cm^{-1}) are clearly observed. A strong peak at 1476 cm^{-1}, corresponding to intermolecular Raman active frequency (tangential mode) Ag (2) of C_{60} molecules, is clearly observed due to the C_{60} encapsulation. While the weak peak at 1437 cm^{-1} near D-band can be attributed to the Hg (7) mode of C_{60} molecules. Moreover, another weak peak at a low frequency of 271 cm^{-1}, corresponding to the Hg (1) mode of C_{60} molecule, is also observed, which may suggest the formation of dimers or clusters since a large amount of C_{60} molecules can be filled inside DWNTs having large diameter, which is also in agreement with the TEM observations.

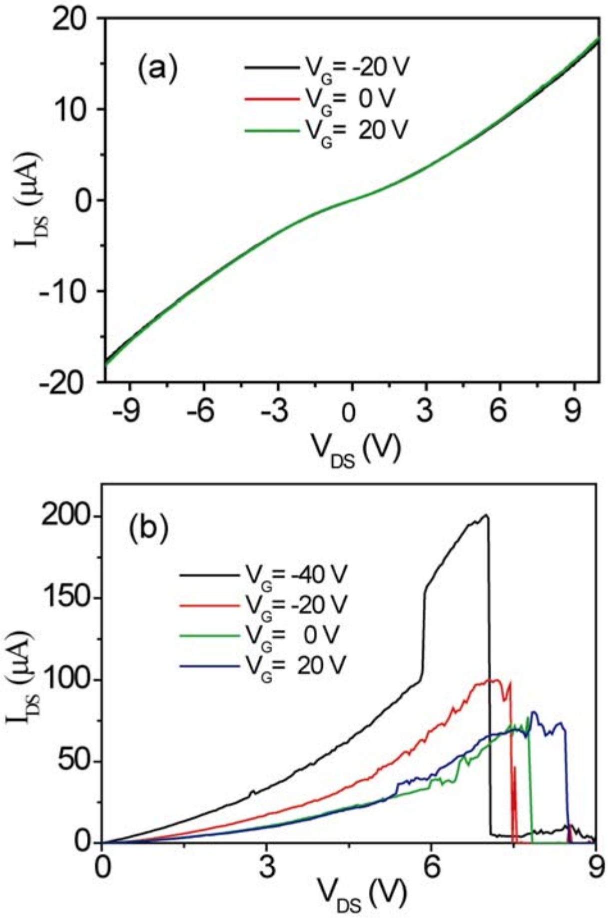

Fig. 4 Characteristics of I_{DS}-V_{DS} curves measured for devices fabricated with a pristine DWNT (a) and C_{60}-encapsulated DWNT (b).

Figure 4(a) shows the current versus source-drain voltage (I_{DS}-V_{DS}) characteristic measured at various gate voltages (V_G = -20, 0 and 20 V) for a device fabricated using a pristine DWNT. The measured current gives rise to a linear increase under different gate voltages, which presents a typical metallic behavior. By comparison, for a C_{60}-filled DWNT device the unique NDR characteristic is significantly observed in the I_{DS}-

V_{DS} curves, as shown in Fig. 4(b). Namely, an initial rise of the current is followed by a sharp decrease instead of the linear increase expected from Ohm's law when the voltage is progressively increased. For instance, at V_G = -40 V the I_{DS}-V_{DS} curve exhibits a NDR characteristic at about V_{DS} = 7 V. Moreover, it is found that the peak voltage of NDR is greatly affected by the gate voltages, and a positive shift of peak voltage from ~7.0 V to ~ 8.5 V is found when V_G is increased from -40 V to 20 V. A high peak-to-valley current ratio (PVCR) over 10^3 is surprisingly observed for this device, which is much higher than that reported previously for NDR devices fabricated using other nanomaterials [8-10].

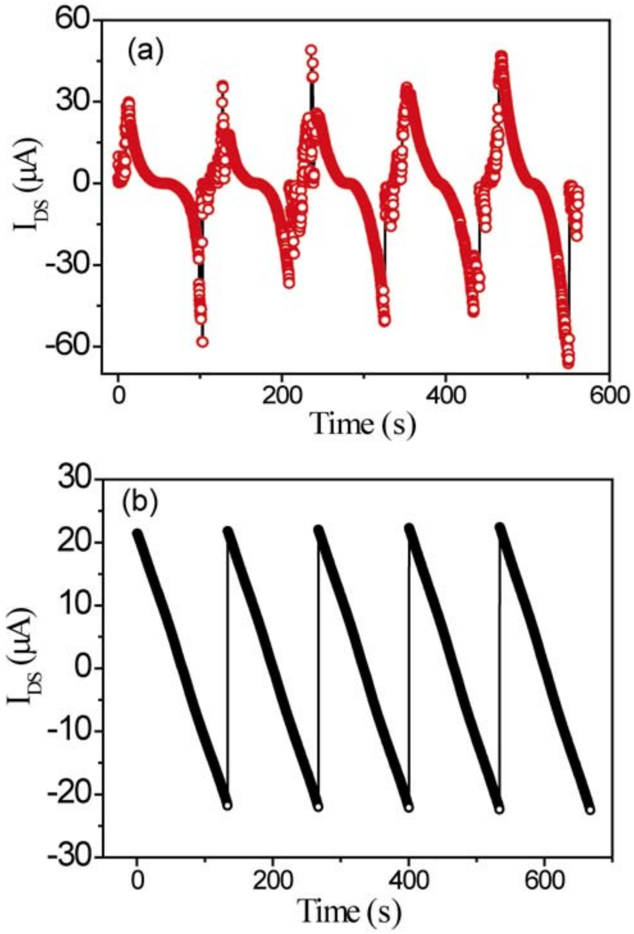

Fig. 5 Characteristics of I_{DS}-V_{DS} curves measured with V_{DS} in the range of -6 ~6 V as a function of time for nanodevices fabricated with C_{60}-encapsulated DWNT (a) and pristine DWNT (b).

In addition, we have carried out the I_{DS}-V_{DS} measurements as a function of time, as shown in Fig. 5. For a C_{60}-filled DWNT based device, the NDR behavior is repeatedly observed with increasing time when the current is measured with V_{DS} from -6 V to 6 V at V_G = 0 V, as shown in Fig. 5(a), which demonstrates evidently that the fabricated device exhibits good reproducibility. In contrast, only linear I_{DS}-V_{DS} curves are observed in the device fabricated with a pristine DWNT, and this result has further confirmed that the encapsulated C_{60} molecules must play a critical role for the observed NDR phenomenon in DWNTs.

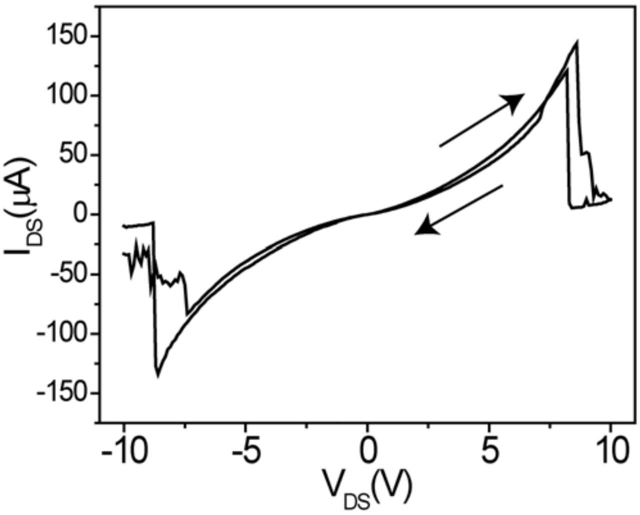

Fig. 6 Characteristics of I_{DS}-V_{DS} curves measured for a C_{60}-encapsulated DWNT device under forward and backward bias sweeps.

In our case, I_{DS}-V_{DS} curves with NDR characteristics are fully reversible upon change of the source-drain bias sweep direction, as shown in Fig. 6 where the I_{DS}-V_{DS} curves are recorded at forward and backward biases with V_{DS} in the range of -10 ~ 10 V. Only small shifts in the peak voltage and peak current are observed with consecutive positive and negative sweeps. Obviously, the performance exceeds that observed in typical solid-state quantum well resonant tunneling heterostructures [11, 12].

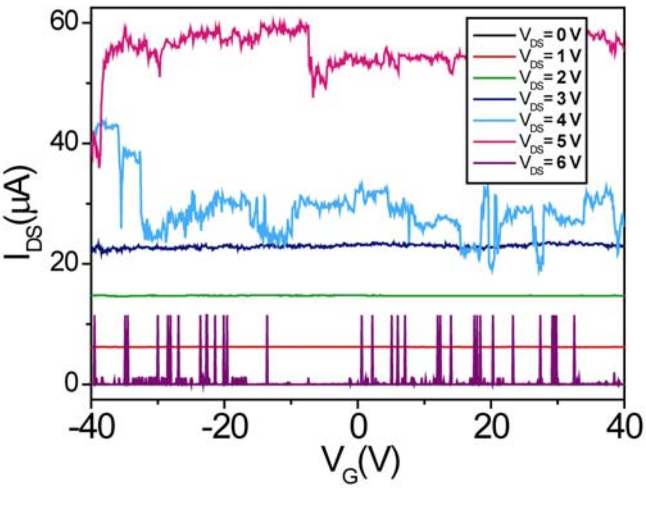

Fig. 7 I_{DS}-V_G characteristics measured at V_{DS} in the range of 0 ~ 6 V ($V_{DS} \leq 3$ V: initial increase; 3 V < V_{DS} < 6 V: unstable region; V_{DS} = 6 V: occurrence of Coulomb blockade behavior).

In Fig. 4, the experimental results have demonstrated that the gate voltages exhibit a great effect on the peak position of NDR characteristic, and so there is a natural question how the current should behave with the gate voltage at a fixed bias. In

Fig. 7, a series of I_{DS}-V_G curves are recorded when V_{DS} is increased from 0 to 6 V [13]. Interestingly, the obtained I_{DS}-V_G curves indicate a multiple feature, and the linear current shows an initial increase when V_{DS} is less than 3 V. However, the current becomes gradually unstable with V_{DS} larger than 3 V. At V_{DS} = 6 V, the current is encountered with an unexpected decrease together with Coulomb oscillations, and this result is well consistent with characteristics of I_{DS}-V_{DS} shown in Fig. 4.

At this moment, although the reason involved in the observed NDR phenomenon in C_{60}-encapsulated DWNTs is still not clear, one candidate mechanism for NDR is a superlattice structure formed in DWNTs after the C_{60} encapsulation. In one pioneering work the NDR characteristic has ever been observed in superlattice structure of tellurium-filled zeolite [10]. In our case, by means of C_{60} encapsulation, one can easily conceive the construction of tunnel barriers and quantum dots inside DWNTs. Since the number of encapsulated C_{60} molecules with diameter of 0.71 nm in a 500-nm-long DWNT is roughly estimated to be more than 6×10^3. With these in mind, ideal superlattice structure is possibly formed in such one-dimensional space of DWNTs. When electrons tunnel through the potential barriers in series separated by C_{60} molecules, the coherent nature of ballistic electron propagation can generate constructive interference, resulting in the occurrence of resonance tunneling.

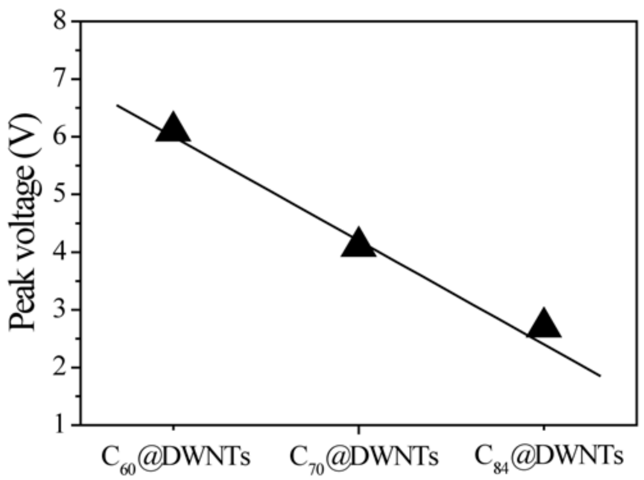

Fig. 8 Peak voltages of NDR for C_{60}@DWNT, C_{70}@DWNT, and C_{84}@DWNT devices, showing a linear decrease with increasing the diameter of fullerene.

Moreover, it is noted that the observed peak voltage and PVCR in our case are much larger than those reported previously [8, 9]. A plausible reason can be explained in terms of the unique structure of C_{60}, which is smaller than the usual semiconductor quantum dots investigated so far. Also, it is found that the NDR characteristics have been observed in devices fabricated with C_{70}, and C_{84} fullerenes encapsulated DWNTs. It is interesting to mention that the peak positions of the NDR move towards lower voltages with increasing diameter of the encapsulated fullerenes, as indicated in Fig. 8, in which peak voltages for the C_{60}, C_{70} and C_{84} encapsulated DWNT devices are estimated on average to be 6.1 V, 4.1 V and 2.7 V, respectively, based on our numerous measurements. Further studies are needed to understand the origin of NDR characteristic and their difference observed in various fullerenes encapsulated DWNTs.

IV. Conclusions

In summary, we have measured the electrical transport properties of metallic DWNTs filled with C_{60} molecules at room temperature. In contrast to pristine DWNTs, the unique NDR behavior with high PVCR more than 10^3 is significantly observed in C_{60}-filled DWNTs due to the formation of superlattice structure. The observed NDR behavior is fully reversible upon change of the source-drain bias sweep direction. Our results reveal evidently that C_{60}-filled DWNTs hold a great potential in the design of resonance tunneling transistors.

Acknowledgment

The authors would like to thank Prof. K. Tohji and Mr. K. Motomiya for their help in the TEM observation

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