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Effect of top metal contact on electrical transport through individual multiwalled carbon nanotubes

Neha Kulshrestha,[1,a)] Abhishek Misra,[2] Senthil Srinivasan,[2] Kiran Shankar Hazra,[1] Reeti Bajpai,[1] Soumyendu Roy,[1] Gayatri Vaidya,[2] and D. S. Misra[1]
[1]*Department of Physics, Indian Institute of Technology, Bombay, Mumbai 400076, India*
[2]*Department of Electrical Engineering, Centre of Excellence in Nanoelectronics, Indian Institute of Technology, Bombay, Mumbai 400076, India*

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The effect of position of top metal contact on the electrical transport through individual multiwalled carbon nanotubes (MWNTs) has been investigated using gas injection system *in situ* in scanning electron microscope to deposit the top platinum metal contacts at different desired sites on the side contacted MWNTs in bridging structure. Current-voltage measurements reveal a significant improvement in electrical properties of the tubes after the top contact is made. This improvement has been found to be independent of position of top contact, i.e., whether the top contact is made on the ends or at any other site of the tube. © *2010 American Institute of Physics*.
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The inefficient contact at the metal-nanotube interface is a great obstruction to reap the benefits of the true electrical characteristics of carbon nanotubes (CNTs).[1,2] The theoretical studies done for single wall nanotubes deal with the two so called side contact and end contact approaches for metal-nanotube interface and conclude that for buried or embedded end contact the contact resistance is comparably less than the side contacted tubes as the open ends of the tube lead to the strong interaction between carbon and metal atoms.[3–5] However the end contact approach needs chemical modification at the tip to open the cap otherwise high resistivity perpendicular to the tube axis may lead to high resistance values.[6] In case of multiwalled carbon nanotubes (MWNTs), however the effect of contact is more critical as coupling of the multiple shells of different electrical nature with the contact metal atoms is one major factor to determine the amount of carrier conduction. The work by de Pablo *et al.* shows that the open ends of the MWNT buried in metal account for the conduction through about one-third of the total number of shells in the tube.[5] In the same study (Ref. 5), the difference in current for some other devices (nanoampere instead of milliampere) has been accounted by internal breaks in the tube itself. The comparison of the two approaches (side and end) is a rigorous task in the wake of repeatability of the experiments on the same tube, however one relevant study has shown the improvement in conductance due to wetting effect by forming liquid Gallium metal contacts and this approach is again a buried end approach.[7]

In this letter, we demonstrate a systematic approach to study the effect of top metal contact and find that the position of top metal contact, i.e., whether the top metal has been deposited at the ends of the nanotube or somewhere else at the tube excluding its ends, does not alter the conduction through nanotube significantly. Hence the chemical modification at the tip or opening the cap is not necessary to draw the electrical properties fully and thick metal deposition at any site may lead to the multiple shell conduction.

The gold microelectrodes (squares of dimensions of 100 μm and separation of 3 μm) were fabricated by direct writing using electron beam lithography on the sputtered deposited chrome-gold layer. The resulting electrode pads are shown in Fig. 1. The MWNTs detached from the pillars synthesized by thermal chemical vapor deposition method were dispersed on the pads after 30 minutes of ultasonication in isopropyl alcohol. The countable number of rows and columns written on the wafer makes it a reliable technique to determine the exact position of MWNT after dispersion. Electrical measurements were done on the tubes bridging the gap between two pads using a Keithley 4200 sourcemeter (produced from Keithley instruments Inc., Cleveland, Ohio, USA).

The gas injection system in Raith 150-TWO direct write setup (produced from RAITH GmbH, Dortmund, Germany) has been used to deposit the platinum (Pt) metal contacts at the two ends of the tube. The metal organic precursor $(Me)_3MeCpPt$ (trimethyl-methylcyclopentadienyl-platinum) in combination with the focused electron beam at the desired area results in the local deposition of Pt metal. Using this technique we are able to deposit Pt metal contact pads of desired dimensions at the desired sites with nanometer precision. One such device, before and after the Pt top contact

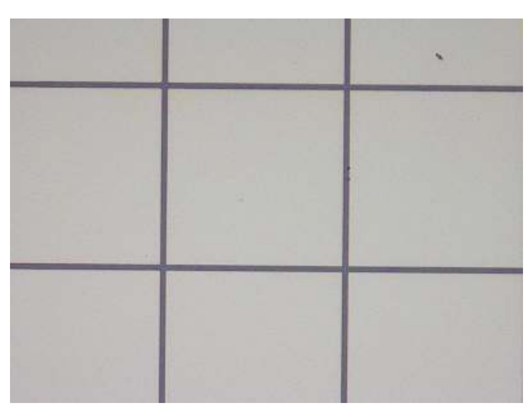

FIG. 1. (Color online) Optical image of gold microelectrodes, separated by dielectric SiO_2 lines.

a)Electronic mail: nehaphysics@gmail.com.

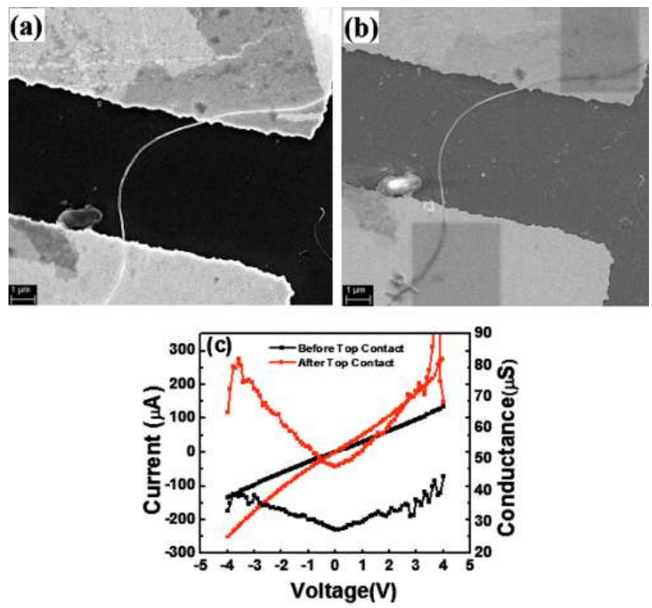

FIG. 2. (Color online) MWNT between the gold pads, (a) before top metal deposition, and (b) after top metal deposition. (c) I-V and G-V curves of the tube are shown.

deposition is shown in Figs. 2(a) and 2(b), respectively. To study the effect of position of top contact on its electrical properties, Pt contact pads have been deposited first at the tube excluding its ends followed by current-voltage (I-V) measurements and then Pt is deposited at the ends of the same tube and again I-V measurements were done. Similar devices have been prepared altering the sequence of Pt metal deposition with I-V measurements at every step. Different permutations of the positions of top contact deposition have also been studied.

The I-V curves and corresponding conductance curves for the tube (Fig. 2), before and after Pt top contact deposition, are shown in Fig. 2(c). Since the study of same tube before and after top contact has been aimed, the joule heating induced breakdown is avoided deliberately before top contact deposition by not exceeding the voltage beyond 4 V.

The two terminal electrical conductance for this tube in the voltage range of -4 to 4 V, before top contacts deposition has been found to be 33.6 μS (0.86 e^2/h) which corresponds to a resistance of 29.8 kΩ. However after top contact deposition the conductance improves significantly to a value of 65.7 μS (1.68 e^2/h, $R=15.2$ kΩ). The current densities for this tube of diameter 110 nm compared at 4 V are 1.4×10^{10} A/m^2 and 2.9×10^{10} A/m^2 before and after top contact, respectively. Same measurements done on other tubes have shown prominent improvement in the current at all biases.

One may suspect that current induced annealing, reported in some previous studies,[8,9] may lead to the improvement in current. To separate out contribution of this current induced annealing in the improvement, the current was measured at 0.5 V after applying every higher biases, which means that first it was measured at 0.5 V, then after 1 V it was again measured at 0.5 V and so on up to 4 V. For the tube shown in Fig. 2, the current at 0.5 V is 13.63 μA and has increased to 14.08 μA when measured after 4 V, hence only 0.03% improvement in current is due to current induced annealing while for the same tube the percent improvement

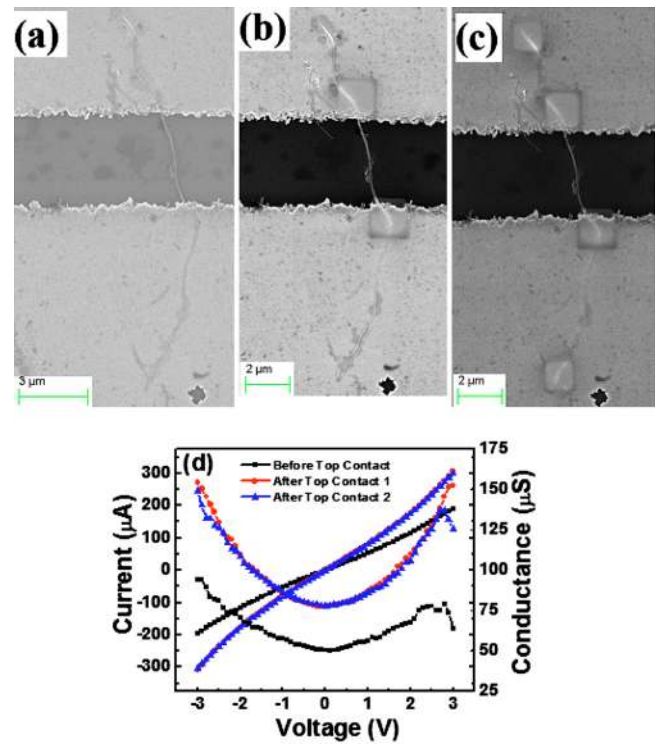

FIG. 3. (Color online) SEM images of the tube (a) before top contact deposition, (b) after top contact deposition at the middle, and (c) after top contact deposition including the ends. (d) I-V and G-V curves of the tube are shown.

in current due to top contact is 0.75% at 0.5 V. In fact the amount of improvement in current due to annealing may be different for different tubes but it is always less when compared to the improvement due to top contact deposition. This is because the current annealing leads to the better contact of only the outer shell with the electrode metal hence reduces the contact resistance but the deposition of top metal leads to the multiple shells to participate in conduction.

It might be expected that deposition of top metal contact at the ends of the tube will lead to the more improvement in the current rather than depositing them at any other site (excluding its ends) as in the former case multiple shells must come in contact with the metal. To investigate the effect of position of the top contact on the electrical properties of the tubes, first the current on the MWNT with only bottom electrodes has been measured [Fig. 3(a)], then I-V measurements were performed after depositing Pt on the tube excluding its ends as shown in Fig. 3(b) and then again Pt had been deposited including the ends of the tube as shown in Fig. 3(c) followed by I-V measurements. The I-V curves after every top metal deposition are shown in Fig. 3(d). Here we assume that as the tubes were detached from the very long (50–80 μm) CNT pillars and further ultrasonicated for separation purpose, hence they are open ended. The length of the tubes after ultasonication becomes less (4–20 μm) which also supports this fact.

A significant improvement in the current has been observed after first metal deposition (when the ends are not buried in Pt) and there is no improvement in the current when the ends of the tube are also buried by top metal, as shown in Fig. 3(d). Similar study is carried out for other tubes in which the sequence of metal deposition has been reversed, i.e., first the metal is deposited at the ends followed by I-V measurements and then again I-V measurements are

TABLE I. Effect of position of top contact (TC) on conductance (G) and current density (J) of different devices.

Position of TC	Diameter (nm)	G before TC (μS)	G after TC1 (μS)	G after TC2 (μS)	J before TC (A/m^2)	J after TC1 (A/m^2)	J after TC2 (A/m^2)
TC1 at middle then TC2 at ends	60	64.73	103.42	101.18	7.0×10^{10}	1.1×10^{11}	1.0×10^{11}
TC1 at middle then TC2 at ends	52	44.63	72.25	71.76	6.4×10^{10}	1.0×10^{11}	1.0×10^{11}
TC1 at end then TC2 at middle	60	39.90	44.40	45.50	4.4×10^{10}	4.7×10^{10}	4.8×10^{10}
TC1, one pad at end one at middle, TC2, vice versa	40	27.98	39.36	47.01	6.6×10^{10}	9.4×10^{10}	1.1×10^{11}
TC1, at middle, TC2, one pad again at middle	73	36.64	82.90	84.03	2.6×10^{10}	5.8×10^{10}	5.8×10^{10}

done after depositing the metal at the inner site of the tube. Different permutations of the positions of top contact have also been studied and for each device improvement in the current is observed only after the first top metal deposition and the sequence of metal deposition or the position (whether the ends of the tube are buried in the metal or not) does not alter the current values much. The results for some devices with different permutations of top contact deposition are tabulated in Table I for the voltage range of −3 to 3 V.

This study suggests that it is not necessary to open and bury the ends of the tube in metal to improve its electrical conduction and deposition of metal at any site may lead to multiple shell conduction. Simple geometrical argument suggests the maximum possible diameter of a sphere inside a regular hexagon of length L is $\sqrt{3}L$. As the carbon-carbon bond length a_{cc} is 1.42 **Å** thus the Pt atom (diameter 2.45 **Å**) may diffuse in the hexagonal networks of carbon atoms, leading to the increased interaction of inner shells of the tube. This type of diffusion is important because as the metal is deposited from the top, the conduction through nanotube will increase even if the outermost shell is semiconducting. Moreover the stress on MWNT due to the atomic weight of deposited metal may also lead to the increased intershell coupling. The same results are expected with other metals like gold (Au) or silver (Ag), commonly used for electrical contacts, however the amount of ultimate improvement in current after top metal deposition will depend on factors such as diffusion coefficient and wettability of the metal. Pertinent studies reviewed by Florian Banhart provides the insights of possible metal-nanotube interactions and diffusion phenomenon for Au and Pt atoms in carbon nanotubes.[10] It is worth pointing out here that after the metal deposition, since multiple shells with different electrical nature couples with the electrodes, the overall improvement in conductance includes conduction through semiconducting shells with high resistance (compared to the metallic ones), which are present in multishell structure of the tube. The increased metal (beneath)-metal (top) interaction via nanotube-metal (top) interaction leads to the more density of states available for the conduction. Thus increased carrier injection may contribute in the improvement in current. In fact, platinum deposited by the metal organic precursor may have carbon contamination hence we predict that the current density may be improved at least by ten times if the pure metal with appropriate thickness (having the bulk Pt metal properties) is used. Once the multiple shells start participating in conduction the position of top metal contact does not alter the electrical properties of the tube. However the quality and thickness of deposited metal pads which are not the subject of this study surely affect the electrical properties. These findings are very important for the on-chip interconnect applications of CNTs in very large scale integration.

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