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Citation: Appl. Phys. Lett. **91**, 133511 (2007); doi: 10.1063/1.2790839
View online: https://doi.org/10.1063/1.2790839
View Table of Contents: http://aip.scitation.org/toc/apl/91/13
Published by the American Institute of Physics

Effect of length and size of heterojunction on the transport properties of carbon-nanotube devices

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(Received 7 August 2007; accepted 10 September 2007; published online 27 September 2007)

By applying nonequilibrium Green's functions in combination with the density-functional theory, we investigate the electronic transport properties of molecular junctions constructed by the mirror symmetrical straight carbon-nanotube heterojunctions. The results show that the length and size of heterojunction play an important role in the electronic transport properties of these systems. The negative differential resistance behavior can be observed in such devices with certain length and size of heterojunction. A mechanism is suggested for the negative differential resistance behavior.
 [DOI: 10.1063/1.2790839]

In the past decade, molecular devices constructed by carbon nanotubes have attracted much attention. The electronic transport properties of carbon-nanotube devices have been studied experimentally[1,2] and theoretically.[3–7] It is known that an ideal single-walled carbon nanotube (SWNT) can be either a metal or a semiconductor, depending on its helicity and diameter. A joining of two or more distinct nanotubes with different helicities and diameters, by introducing pentagon-heptagon defect pairs into the perfect hexagonal network,[8] will produce a carbon-nanotube heterojunction. The electronic characteristics of semiconductor/metal and metal/metal heterojunctions have been investigated experimentally[9–11] and theoretically.[12,13] The carbon-nanotube *Y*-junctions[14] and *T*-junctions,[15] and superlattice nanotube structures[16] have also been reported. However, most of the these studies mainly focused on the electronic properties of carbon-nanotube heterojunctions (CNTHs), while the transport behavior of metal electrode/carbon-nanotube heterojunctions/metal electrode devices (MCNHM) were paid less attention.

In this present work, by applying nonequilibrium Green's functions and first-principles calculations, we investigate the electronic transport properties of MCNHM modulated with different CNTH configurations, as shown in Fig. 1. The structures have been optimized and the quantum transport calculations have been carried out by an *ab initio* code package, TRANSIESTA-C.[17] TRANSIESTA-C is based on the real-space, nonequilibrium Green's function formalism and the density-functional theory. The main feature of the computational package is to model a nanostructure coupled to external electrodes with different electrochemical potentials and to realize the transport simulation of the whole two-probe system. Our results show that electronic transport properties of the devices may be modulated by the size and length of the heterojunction. Moreover, a negative differential resistance (NDR) is observed and tuned by the length and size of the heterojunction. The NDR is a very useful property in electronic devices such as molecule switch. The NDR behavior has been found in mesoscopic systems such as double barrier quantum wells[18] and superlattices.[19,20] The resonant[18] and off-resonant[19] electronic tunneling mechanism have been proposed to explain the NDR behavior. NDR has also been found in a number of molecular devices. Several groups observed the NDR in a phenylene ethynylene trimer substituted by nitro and amino groups on the central ring.[21–24] A possible mechanism such as a two-step reduction process,[21] the change of the molecular conformation due to the change of the electronic charge state of the molecule under increasing bias,[22] a bias-induced alignment of the molecular orbitals,[23] and side groups effects[24] for the NDR in such a system have also been proposed by these groups. The NDR was also found in a single benzene ring with a NO_2 ligand.[25,26] D. Ventra *et al.* attributed the NDR to the rotation of the ligand activated by temperature,[25] while Luo and Fu suggested that a one-electron reduction is the mechanism behind the NDR.[26] Phonon-mediated NDR in suspended carbon-nanotube molecular wires has also been reported.[27,28] In analogy to a quantum double-dot structure, Liu *et al.* showed that molecular-level crossing in an organometallic molecular double-dot system can lead to NDR.[29] Recently,

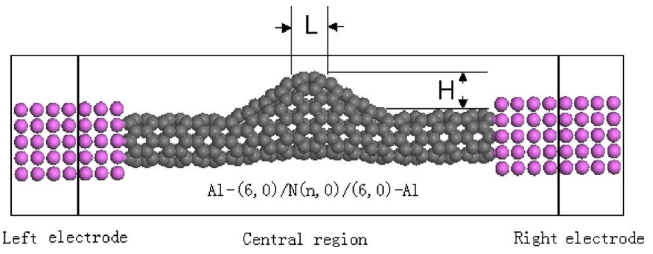

FIG. 1. (Color online) Structures of the extended molecules: A mirror symmetrical straight SWNT heterojunction is coupled to two semi-infinite Al electrodes, and the extended molecule consists of 5×5 (100) four layers of Al slab with 50 atoms per unit cell in the right lead, three layers of Al slab with 37 atoms in the left lead, and the mirror symmetrical SWNT heterojunction indicated as $M(6,0)/N(n,0)/M(6,0)$, where M and N describe the number of cells of zigzag carbon nanotubes. Here, we choose the values of n to be 6, 7, and 8 and those of N to be 1, 2, 3, and 5. The number M of cells of the (6, 0) tube is fixed at 3. The distance between the SWNT end and the metal surface is chosen to be 1.6 Å, which is a typical electrode-tube distance.

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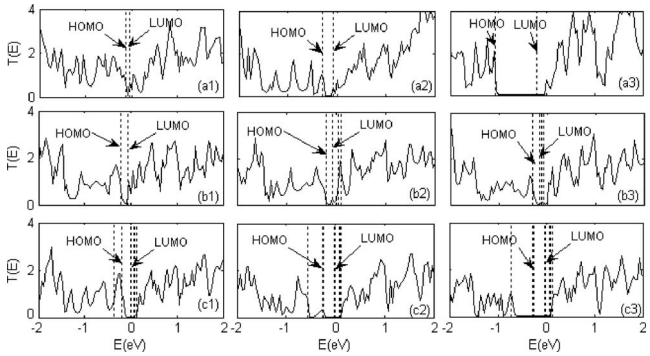

FIG. 2. Transmission coefficient $T(E,V_b)$ for the two-probe systems shown in Fig. 1 as a function of injected electron energy at zero bias (V_b=0): (a1), (a2), and (a3) correspond to (6, 0), (7, 0), and (8, 0) tubes without heterojunction; (b1), (b2), and (b3) correspond to the numbers of cells N=1,3,5 in (6,0)/N(7,0)/(6,0) systems; (c1), (c2), and (c3) correspond to N=1,3,5 in (6,0)/N(8,0)/(6,0) systems, respectively. Some frontier molecular orbitals have been denoted by dashed lines, and HOMO and LUMO have also been labeled. Here, the energy origin is set to be the Fermi level of the system.

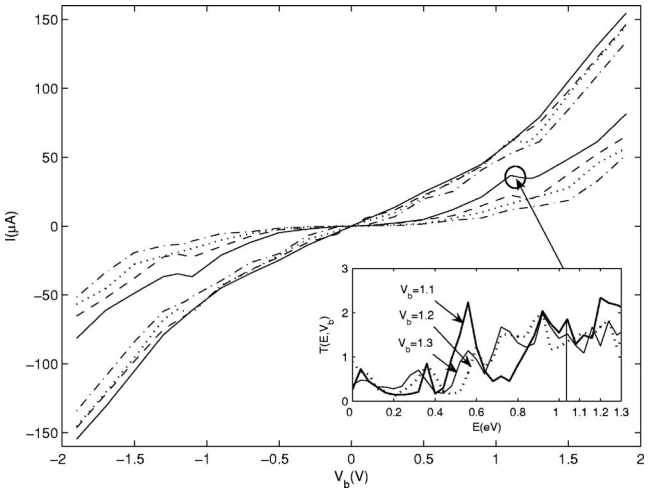

FIG. 3. Calculated current as a function of the applied bias for carbon-nanotube heterojunction devices: The curves are divided into two bundles. the above and below bundles (for positive bias) correspond to (6,0)/N(7,0)/(6,0) and (6,0)/N(8,0)/(6,0) carbon-nanotube heterojunctions, respectively. The solid, dashed, dotted, and dot-dashed curves correspond to N=1, 2, 3, and 5, respectively. The insert gives $T(E,V_b)$ of the (6,0)/1(8,0)/(6,0) system under biases V_b=1.1, 1.2, and 1.3 V.

Bandyopadhyay and Wakayama found that NDR could be generalted and tuned in the junctions of a Rose Bengal molecule.[30] In spite of a number of theoretical and experimental studies about NDR in various kinds of molecular devices, the origin for NDR is still under intense debate. Our results show that the conduction channel being suppressed at a certain bias is responsible for the NDR behavior in the MCNHM systems considered here.

In Fig. 2, we show the transmission coefficient $T(E,V_b)$ at zero bias (V_b=0) for the two-probe systems shown in Fig. 1. From Figs. 2(a1)–2(a3), as expected, it is found that no transmission gap (namely, zero transport flat) exists in the (6, 0) tube system, while a narrow and a wider transmission gap exist in the transmission spectra of (7, 0) and (8, 0) tube systems, respectively. This means that the (6, 0) tube system is of a metallic behavior, while (7, 0) and (8, 0) tube systems display a semiconducting behavior. When introducing a (7, 0) or (8, 0) tube into a (6, 0) tube, a metal/semiconductor/meter carbon-nanotube heterojunction is formed. For such a system, the transmission behavior is different from that of a single (6, 0), (7, 0), or (8, 0) tube system. From Figs. 2(c1)–2(c3), it can be found that transmission gap appears, and it becomes wider with an increase of the number N of cells of the (8, 0) tube. It is known that an interface can form in the matching region between (6, 0) and (7, 0) or (6, 0) and (8, 0) tubes. The electron transport through heterojunction is dependent on the resonant coupling between different tubes. For a single (6, 0) tube system, we find by calculation that its transport channels [lowest unoccupied molecular orbital (LUMO), highest occupied molecular orbital (HOMO), and HOMO-1 to HOMO-5] lie within the energy gap of a single (8, 0) tube. For (6,0)/N(8,0)/(6,0) systems, when the system contains only one cell of the (8, 0) tube, it is found by calculation that only LUMO channels will be suppressed, so a narrow transmission gap appears. When more cells of the (8, 0) tube is contained in CNTH, more channels will be closed, and the transmission gap becomes wider. From Figs. 2(b1)–2(b3), we find that for (6,0)/N(7,0)/(6,0) systems, the transmission coefficient is decreased with the number N of cells of the (7, 0) tube, and a small transmission gap appears when N is 5. This results from the fact that the energy gap of the (7, 0) tube is narrow and the geometric distortion is weak in the matching region between (6, 0) and (7, 0) tubes.

Figure 3 describes the currents as a function of the applied bias for MCNHM systems. From the figure, we can see clearly that when the bias is larger than a certain threshold bias, the current can flow apparently in (6,0)/N(8,0)/(6,0) systems, and the value of the threshold bias becomes bigger when changing N from 1 to 5, which indicates that these systems are of semiconducting characteristics and they can be modulated by the length of the junctions. However, for (6,0)/N(7,0)/(6,0) systems, there is a small zero-current flat (about ±0.25 V) only when N=5. From the figure, we find an interesting feature: The current decreases obviously when the bias is between $|V|$=1.1 V and $|V|$=1.3 V for both (6,0)/3(7,0)/(6,0) and (6,0)/1(8,0)/(6,0) systems, which means NDR appears in the bias range in the these systems. However, for other systems, we do not find the NDR behavior. To understand the NDR behavior in our systems, we calculated the transmission coefficient $T(E,V_b)$ of the (6,0)/1(8,0)/(6,0) system under biases V_b=1.1, 1.2, and 1.3 V, as shown in the inset in Fig. 3. It is clearly seen that for V_b=1.1 V and V_b=1.3 V, there are three peaks in the region of energy E between 1.0 and 1.3 eV. However, only two peaks appear in the transmission spectra in the region of energy E between 1.0 and 1.3 eV for V_b=1.2, which means that the conduction channel at E=1.04 eV is suppressed.

To understand the origin of the peaks being suppressed in transmission curves, in Fig. 4, we give the molecularly projected self-consistent Hamiltonian (MPSH) at the eigenvalue E=1.04 eV of the (6,0)/1(8,0)/(6,0) system for the biases V_b=1.1, 1.2, and 1.3 eV, respectively. Considering the fact that the Fermi level (energy zero) is varied with the bias, the levels of the molecular orbitals are also varied with the bias. By our calculation, at the energy E=1.04 eV, the molecular orbitals correspond to LUMO+19, LUMO+19, and LUMO+20 for the biases V_b=1.1, 1.2, and 1.3 V, respectively. From Fig. 4(a), it can be found that when the bias is 1.1 V, the MPSH of LUMO+19 is delocalized, which shows that the conduction channel can make a larger contribution to

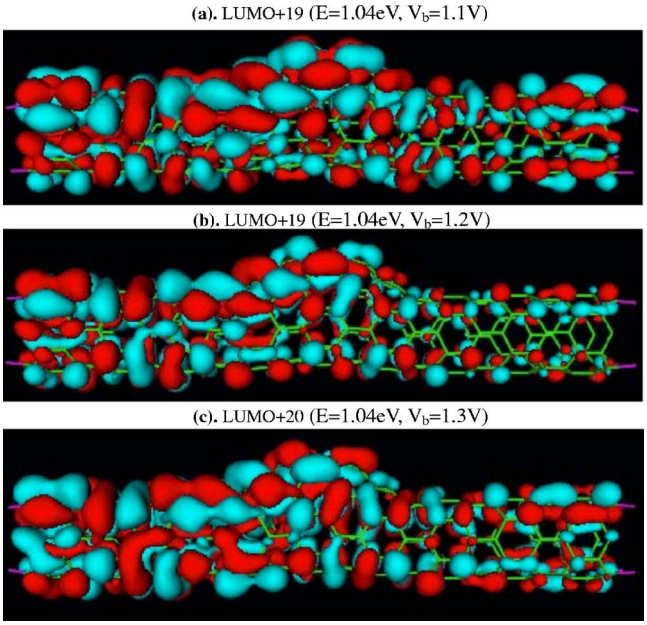

FIG. 4. (Color online) The molecularly projected self-consistent Hamiltonian (MPSH) at the eigenvalue $E=1.04$ eV of the $(6,0)/1(8,0)/(6,0)$ system under the biases $V_b=1.1$ V(LUMO+19), 1.2 V(LUMO+19), and 1.3 V(LUMO+20).

the current. As a result, a transport peak appears at the $E=1.04$ eV for the bias of 1.1 V, as shown in the inset in Fig. 3. From Fig. 4(b), we can find that when the bias is 1.2 V, the MPSH of the channel LUMO+19 localizes on the left part of the structure, which means that the channel is suppressed. Consequently, the current at the bias of 1.2 V is smaller than that at the bias of 1.1 V. As the bias is further increased, the electronic current remains low due to LUMO+19 being suppressed. When the bias is increased to 1.3 V, channel LUMO+20 is opened, so a peak appears at the $E=1.04$ eV again. This leads to the current being rapidly increased. So, whether the channel is opened or suppressed is responsible for the decrease of the current of the $(6,0)/1(8,0)/(6,0)$ system at the bias being about 1.1 V in Fig. 3. In fact, this is the reason for NDR. A similar explanation is for the NDR behavior in the $(6,0)/3(7,0)/(6,0)$ system at the bias of 1.2 V. It should be noted that the as long as not too many molecular orbitals contribute to the electron transport, the role of suppressed channel is obvious to the electron current, and so the NDR can be observed clearly. This is similar to the negative differential capacitance in the quantum dot.[31] From our calculations, it is known that the NDR behavior can be observed only in the $(6,0)/1(8,0)/(6,0)$ and $(6,0)/3(7,0)/(6,0)$ systems, while in $(6,0)/N(8,0)/(6,0)$ ($N=2$, 3, and 5) or $(6,0)/N(7,0)/(6,0)$ ($N=1$, 2, and 5) systems, the NDR does not occur for the explored bias range. These results show that the NDR behavior originates from the conduction channel being suppressed at a certain bias. Whether the channel is opened or suppressed depends on the length and size of the semiconductor tube in the molecular junction.

In summary, using first-principles quantum transport calculations, we have investigated the transmission properties of electrons in molecular junctions constructed by the mirror symmetrical straight carbon-nanotube heterojunctions with different sizes and lengths. The calculated results show that the size and length of the heterojunction play a significant role on the current-voltage characteristics. The NDR behavior can be observed in such devices and depends on the size and length of the heterojunction. It is suggested that the conduction channel being suppressed at a certain bias voltage is the origin of the NDR behavior.

This work was supported by the National Natural Science Foundation of China (Nos. 90403026, 10674044, and 90606001) and by the Hunan Provincial Natural Science Foundation of China (No. 06JJ20004).

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