Electrical Applications for Novel Carbon Nanotube Morphologies: Does Function Follow Shape?

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Nanoscience promises a future of novel devices and technologies that harness the atomic or molecular structure of matter. An intriguing manifestation of this paradigm is in the electrical properties of nonlinear carbon nanotube (CNT) morphologies, such as nanocoils/helices and branched forms, which exploit novel functionality at the nanoscale. For example, in a Y-CNT, the junction region can possess a charge due to the necessary presence of topological defects and catalyst particles. The resultant induced charge and asymmetry is analogous to "gating" that could be harnessed for electrical modulation and switching. Helical morphologies, in addition to use as electrical inductors, could be pertinent for a new type of electronic architecture wherein a coil could correspond to a sequence of junctions with alternating metallic and semiconducting character. While these alternative device concepts represent a revolutionary departure from the conventional architectures, the exact underlying mechanism of self-gated switching behavior is yet to be clearly understood.

INTRODUCTION

Carbon nanotubes (CNTs) have been widely touted as the wonder materials of the new century, with all manner of superlatives ascribed to them, and are being considered for a range of applications[1] from structural motifs in automobiles to nanometer-scale electronics. This article will briefly review the electrical properties of nonlinear CNT-based morphologies and their possible use in entirely new multi-functional paradigms. The close relation of some of these naturally formed entities, such as Y-shaped and helical forms, to existing devices such as three terminal electronic transistors and electrical inductors respectively, excites the imagination of what else is possible at the nanoscale. See the sidebar for background on single- and multi-walled carbon nanotubes (MWNTs).

NOVEL ELECTRONIC FUNCTIONALITIES IN NONLINEAR CNT STRUCTURES

Carbon nanotubes have been widely tested for applications in advanced switching electronics. Two main device topologies are currently being worked upon. First is adaptation to the conventional silicon-based metal oxide semiconductor field effect transistor (MOSFET) architecture, where the CNT can serve as the channel. Second is novel morphologies, such as coils/helices, and Y- and T-junctions which possess intrinsic functionality (e.g., the gate could be fabricated as a part of the device).[16]

Excellent reviews of the applications of the former approach exist[17–20] and have merits, especially in terms of reducing the leakage current and minimizing the power—two issues of vital importance in the scaling of transistor devices, in accordance with International Technology Roadmap for Semiconductors recommendations.[21] While extremely important in elucidating fundamental properties, the experiments have used external electrodes (the well-known MOSFET architecture), made through conventional lithographic processes to contact the nanotubes (which serve as channels) and do not represent truly nanoelectronic circuits. In other demonstrations, cumbersome atomic-force microscope (AFM) manipulations[22] have been utilized to tune CNT properties.

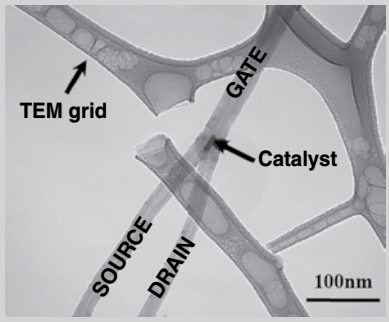


Figure 1. A carbon-nanotube (CNT) based Y-junction. The Fe-Ti catalyst particles along with the presence of topological defects at the junction region are thought to be responsible for novel electrical behaviors.

This article mainly focuses on the latter aspect of using novel CNT morphologies due to the possibility of obtaining unique functionality at the nanoscale. Such functionalities can be found, for example, in Y-junctions[16] (see Figure 1) or in more complex networks,[23] incorporating T- and X-junctions. (Note that these are naturally formed CNT forms and distinct from crossed nanotube junctions,[24] where nanotubes are individually assembled using an AFM.) Developing nonlinear CNT-based devices, besides miniaturization and lower power consumption, also allows the exploitation of the features of inherently quantum mechanical systems, such as ballistic transport and low switching voltages[25] ($\sim k_B T/e = 26$ mV) at room temperature.

Nonlinear CNT Morphologies

Branched nanotubes with T-,Y-, L-, and more complex junctions, were initially observed in arc-discharge produced nanotubes.[26] Early work on individual Y-junctions resulted in observation of nonlinear I-V characteristics[27] and opened up a vista of possibilities. While theoretical explanations of observed electrical behavior were based on SWNT Y-junctions due to computational ease, the experiments were performed on mul-

tiwalled carbon nanotubes (MWNTs), which are easier to manipulate.

In yet another manifestation of nonlinear CNT forms, the synthesis of helical morphologies of nanotubes (NTs) and nanofibers (NFs), through chemical vapor deposition (CVD) techniques, has been widely reported[28–30] (see Figure 2) and can be made practical for a wide variety of applications (e.g., nanoscale mechanical springs and electrical inductors).[28] Additionally, the observed nano-coils/-helices (NC/NHs) could be pertinent for a new type of electronic architecture. It was suggested that a coil could correspond to a sequence of junctions with alternating metallic and semiconducting character,[31] adding to the possibilities of novel behavior in nonlinear nanostructures. Nano-coils/helix formation, mostly observed in carbon nanotube/-fiber[32] (CNT/CNF) syntheses, is also scientifically interesting in that helices abound in nature (e.g., DNA, proteins etc.) and a connection is being made at the nanoscale between carbon-based inorganic and organic structures. In our preliminary studies, we have seen[30] that the observed coiling is catalyzed by indium addition: if the In/Fe ratio ($R_{In/Fe}$) in the feedstock mixture was large, ≥3, helical carbon NFs were formed in the majority, while a decreased $R_{In/Fe}$, say ≤2, forms helical carbon NTs (see Figure 2c) with a larger pitch. (Nanotubes are distinguished from NFs in that the former comprise parallel walls with a clearly distinguishable core while the latter is typically a solid filament of amorphous carbon.)

A study of the inductive electrical characteristics of the helical morphologies is under progress by the authors. It is to be noted that in these CNT forms, quantum mechanical considerations dealing with the restricted space of electron movement leads to a kinetic inductance (L_K) of ~10 nH/μm which could be four orders of magnitude larger[33] than the classical electrostatic inductance (L_{ES} ~1 pH/μm). Preliminary experiments have also been conducted aimed at investigating the electrical characteristics of the Y- and T-shaped forms, some of which will be reported here.

THE BASICS: SWNTs AND MWNTs

A single-walled carbon nanotube (SWNT) is formed by wrapping a single sheet of graphite (graphene), seamlessly, into a tubular form. It is interesting to note that graphene by itself, can be characterized as either a zero-energy band gap semiconductor or a semimetal, and imparts such flexibility to a nanotube. It is well known[2,3] that the fundamental electrical conducting properties of a nanotube depend on the nature of wrapping (chirality) and the diameter (typically, SWNTs have diameters of 0.4–2 nm). On the other hand, multi-walled carbon nanotubes (MWNTs) are composed of coaxial nanotube cylinders, of different helicities, with a typical spacing of ~0.34 nm, which corresponds closely to the inter-layer distance in graphite of 0.335 nm. These adjacent layers are generally non-commensurate (different chiralities) with a negligible inter-layer electronic coupling and could alternate randomly[4,5] between metallic and semiconducting varieties. The layers, constituting the individual cylinders, are found to close in pairs at the very tip of a MWNT, and their detailed structure plays an important role, say in electronic and field emission[6] properties of CNTs.

Single-walled carbon nanotubes and MWNTs have been synthesized by various methods (e.g., by arc discharge and laser ablation methods) which seem to have a higher degree of structural perfection due to the high temperatures (>3,000°C) involved in the synthesis.[7] Nanotubes are also grown through chemical vapor deposition (CVD) at a lower temperature (<1,000°C) with a higher defect density, which in turn adversely affects the electrical and structural characteristics. Typical MWNT diameters grown by the arc-discharge method are ~20 nm, while CVD-grown nanotubes can have much larger diameters of up to 100 nm. Larger-diameter tubes are generally found to have a greater density of defects (i.e., vacancies or interstitials). Most of the nonlinear CNT morphologies that have been studied are of the MWNT variety and their practical use is predicated on nanoscale defect engineering.

The electrical properties of both SWNTs and MWNTs have been relatively well explored. While SWNTs can be described as quantum wires due to the ballistic nature of electron transport, the transport in MWNTs is found to be diffusive/quasi-ballistic.[8,9] In MWNTs, it was determined[10] that current flow only occurs through the outermost nanotube and many features peculiar to reduced dimensionality are manifest. Additionally, the mutual interaction between the adjacent coaxial cylinders makes for a richer band structure[11] than SWNTs and greater tunability. Quantum dots can also be formed in both SWNTs[12] and MWNTs[8] and the Coulomb blockade and the quantization of the electron states are exploited for single-electron transistors.[13]

New physical principles are often found to be necessary to explain phenomena associated with CNTs. For example, the traditional Fermi liquid theory, used to describe electronic excitations and properties in the non-interacting electron limit, is often inadequate.[14] Electron correlations have to be considered and the Luttinger liquid theory, which does take into account electron interactions, is more appropriate and manifests novel physics[15] such as spin-charge separation and interaction-dependent power laws in the electrical conduction. In most experimental studies, however, the details of such behavior are masked by imperfect contacts and contact resistances.

Electron Momentum Engineering in CNT Y-Junctions

The basic functionality is derived from an electron-wave Y-branch switch (YBS)[34] where a refractive index change of either branch through electric field modulation can affect switching. This device, demonstrated in the GaAs/AlGaAs[35] based two-dimensional electron gas system using conventional lithographic schemes, relies on ballistic transport and is useful for low power, ultra-fast (THz) signal processing. It was derived theoretically[25] and proven experimentally[36] that the ballistic nature of the electron transport makes possible nonlinear (diode-like) I-V characteristics.

It was also shown that defect topology can affect the electron momentum and guide the current to a pre-determined spatial location independent of input current direction. This type of current tuning/rectification involves a new principle of electron momentum engineering in contrast to currently used band engineering, and CNTs provide a more natural avenue to explore such behavior. It was theoretically postulated[37] that switching and rectification could be observed in symmetric Y-junction NTs,[38] where both the junction geometry and the presence of catalyst particles at the junction, introduced during synthesis, are influential.

New Physical Principles and Potential Applications

As examples of novel functionality at the nanoscale, the Y-junctions represent one of the first attempts in realizing a nanoelectronic device from CNTs alone, through the use of new physical principles peculiar to nonlinear structures. At the nanometer scale the dimensions of the device are comparable to electron wavelength (λ_F) and electron travel/current must be considered in terms of wave propagation, analogous to the propagation of light down an optical fiber. Wave phenomena, such as interference and phase shifting, can be used to construct new kinds of devices. A sampling of the possibilities is shown in Figure 3.

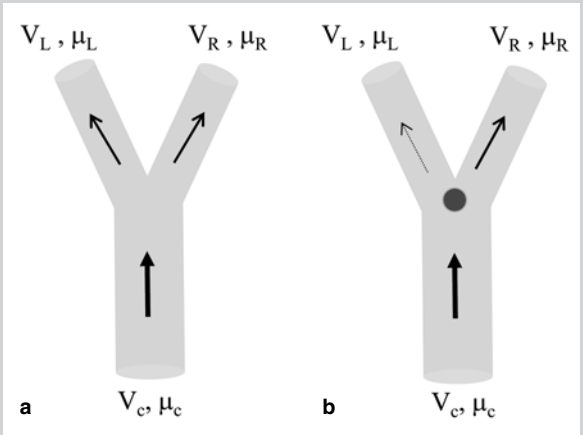


Figure 3. A schematic representation of a Y-CNT that can be used as a prototypical nanoelectronic element for a variety of functions such as switching or as a quantum dot (say, for photodetector applications), depending on the transmission characteristics of the constituent branches. (a) A Y-branch switch, L = R ≠ C; (b) quantum dot, L ≠ R ≠ C. L, R, and C refer to the left, right, and central/stem branches of the Y-junction.

Switching and Transistor Applications

In a basic Y-junction switch, an electric field (corresponding to an applied voltage of a few mV) can direct electrons into either of two branches, while the other branch is cut off.[25] Such a switch can operate over a wide range of electron velocities and energies, as the carriers are not stopped by a barrier but are only deflected. An operational advantage over a conventional field effect transistor is that the current is only redirected between two outputs rather than completely turned on/off,[39] leading to faster modulation/switching. An electrical asymmetry can also be induced through structural or chemical means across the two branches in a nanostructured junction. The Y-junction region, for instance, can possess a positive charge[37] due to the presence of topological defects,[40] viz., by the formation of non-hexagonal polygons (e.g., pentagon-heptagon pair defects) to satisfy the local bond order,[41] where delocalization of the electrons over an extended area is equivalent to a net positive charge, and also due to catalyst particles formed during synthesis.[42,43] This charge and the induced asymmetry is analogous to a "gating" action that could be responsible for electrical modulation and switching.

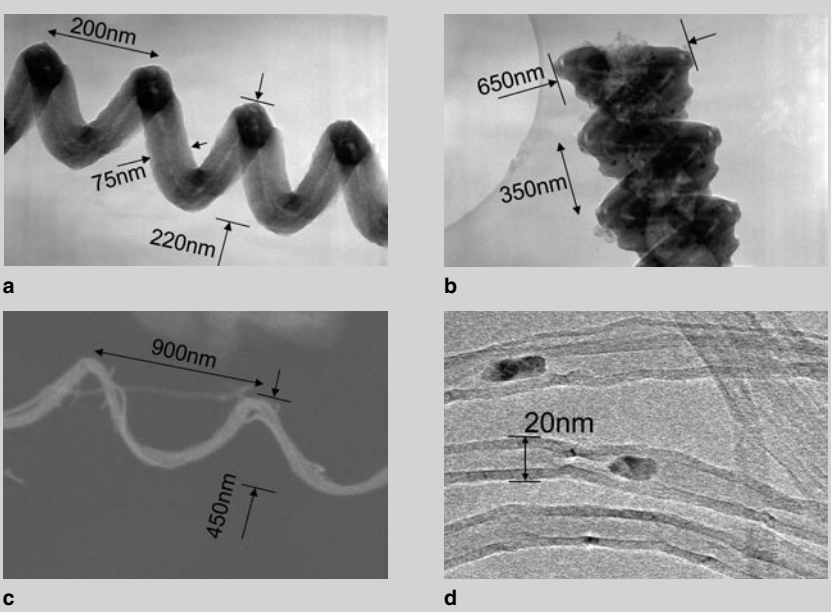


Figure 2. Transmission-electron microscope (TEM) micrographs of (a) singly wound or (b) doubly wound, coiled carbon nanofibers synthesized through thermal CVD at high In concentration (In/Fe ratio >3). (c) Scanning-electron microscope micrograph of nanotube bundles obtained at lower indium concentrations (In/Fe ratio <2). (d) High-resolution TEM of the constituent tubes in (c) shows the random location of iron-based catalyst particles throughout the nanotubes.

Rectification and Logic Function

It is possible to design logic circuitry, based on electron waveguiding, to perform operations similar to and exceeding the performance of conventional electronic devices.[39] When finite voltages are applied to the left and the right branches of a Y-junction, in a push-pull fashion (i.e., $V_L = -V_R$ or vice versa), the voltage output at the stem would have the same sign as the terminal with the lower voltage.

This dependence follows from the principle of continuity of electro-chemical potential ($\mu = -eV$) in electron transport through a Y-junction and forms the basis for the realization of an AND logic gate (i.e., when either of the branch voltages is negative, say, corresponding to a logic state of 0), the voltage at the stem is negative and positive voltage (logic state of 1) at the stem is obtained only when both the branches are at positive biases. The change of μ is also not completely balanced out due to the scattering at the junction and results in nonlinear interaction of the currents from the left and the right sides.[36] To compensate, the resultant center branch voltage (V_c) is always negative and varies parabolically (as V^2) with the applied voltage.

Harmonic Generation/Frequency Mixing

The nonlinear interaction of the currents at the junction region also suggests

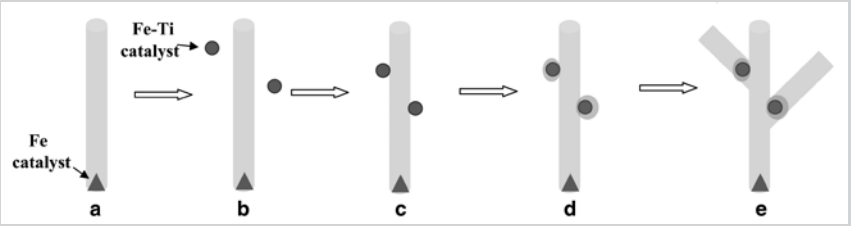


Figure 4. The postulated growth sequence of a Y-junction nanotube.[43] (a) Initial seeding of a straight nanotube through conventional catalytic synthesis, (b) titanium-doped iron catalyst particles (obtained from decomposition of a ferrocene and $C_{10}H_{10}N_4Ti$ mixture) attach (c) to the sidewalls and (d,e) nucleate the side branches. The high heat of formation of TiC is a possible reason for the attachment of titanium-doped iron particles.

the possibility of higher frequency/harmonic generation. When an alternating current (AC) signal of frequency ω, V_{L-R} = A cos [ωt], is applied between the left (L) and right (R) branches of the Y-junction, the output signal from the stem (V_c) would be of the form:

$$V_c = \mathbf{a} + \mathbf{b} \cos [2\omega t] + \mathbf{c} \cos [4\omega t],$$

where **a**, **b**, and **c** are constants.

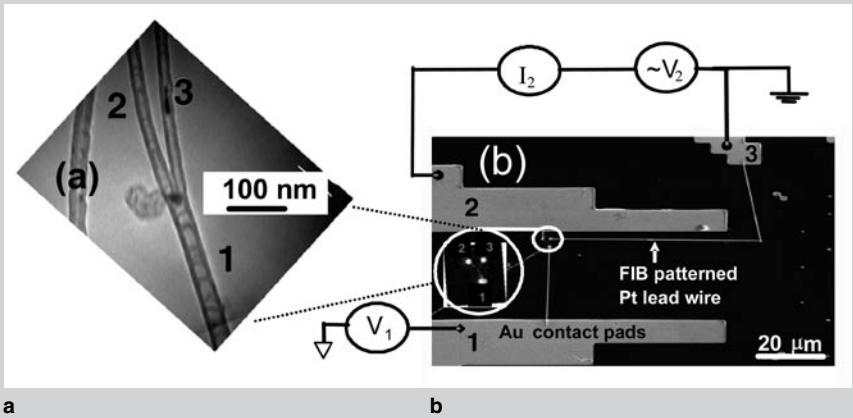
The Y-junction can then be used for second and higher harmonic generation or for frequency mixing. The second harmonic (2ω) output is orthogonal to the input voltage and can be easily separated out. These devices can also be used as ultra-sensitive power meters, as the output is linearly proportional to V^2 to very small values of V. A planar CNT Y-junction, with contacts present only at the terminals, suffers from less parasitic effects than a vertical transistor structure and high frequency operation, up to 50 GHz at room temperature,[44] is possible.

Experimental Measurements on Y-Junctions

The Y-junction nanotubes were grown[43] on bare quartz or SiO_2/Si substrates through thermal CVD (Figure 4). The ratio of the titanium-precursor gas and the feedstock gases can be adjusted to determine the growth of the side-branches at specific positions. The branch formation has also been found to be sensitive to temperature, time of growth, and catalyst concentration.

The details of sample preparation and arrangement for electrical characterization are briefly illustrated in Figure 5 and the details have been reported elsewhere.[16,43] The circuit arrangement for probing the Y-junctions is shown in Figure 5b and the nonlinear characteristics are shown in Figure 5c. The possibility of using the CNT Y-junctions for switching applications as an electrical inverter analogous to earlier[34,36,45] Y-switch studies was explored. In this measurement, a direct current (d.c.) voltage was applied on one branch of the Y-junction while the current through the other two branches was probed under a small AC bias voltage of 0.1 V. As the d.c. bias voltage is increased, at a certain point the Y-junction goes from nominally conducting to a "pinched-off" state. This switching behavior was observed for all the three branches of the Y-junction, at different d.c. bias voltages. The absolute value of the voltage at which the channel is pinched off is similar for two branches (2 and 3) (~2 V, as seen in Figure 6c and d), and is different for the third stem branch (1) (~4.6 V, as in Figure 6a and b). The switching behavior was seen over a wide range of frequencies, the upper limit of 42 kHz being set by the capacitive response of the Y-junction. A more in-depth analysis of the contributing factors to the electrical conductance of a Y-junction is also possible (e.g., an increased resistance in a branch could be correlated with the presence of a catalyst particle).

Preliminary evidence of AND logic gate behavior was also observed.[16] The logic characteristics are not perfect, however, due possibly to imperfections[46] of the nanostructure and need to be investigated. The detailed nature of the electrical switching behavior is also not understood at present. The presence of catalyst nanoparticles (Figures 1 and 5a) in the conduction paths could blockade



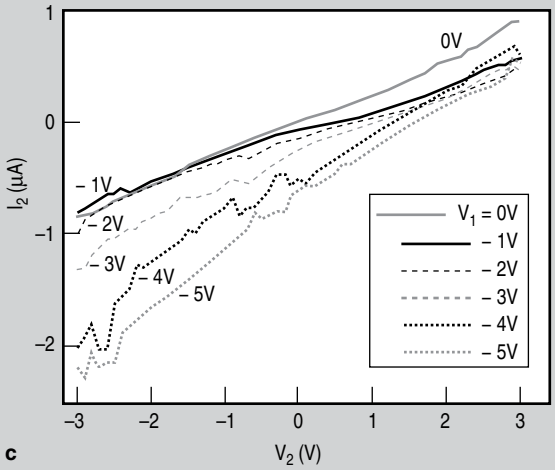


Figure 5. (a) A TEM image of a MWNT Y-junction, and (b) the electrical measurement configuration as imaged in the SEM. (c) Current (I)-voltage (V) characteristics of a Y-junction. A constant d.c. voltage is applied on the stem (1 in Figure 5a), while the current in the branch 2 is monitored as a function of applied voltage (from top to bottom: 0 V, –1 V, –2 V, –3 V, –4 V, and –5 V). The gating action of the applied stem branch voltage and the asymmetric response is to be noted.

current flow, and their charging could account for the abrupt drop-off of the current. The exact magnitude of the switching voltage would then be related to the exact size of the nanoparticle, which suggests the possibility of nano-engineering the Y-junction to get a variety of switching behaviors. An alternative possibility is that there is intermixing of the currents in the Y-junction, where the electron transmission is abruptly cut off due to the compensation of currents. For example, the current through branches 2 and 3 is cancelled by current leakage through the central stem (1 in Figures 3a and 5a). The geometry and defects could also be responsible. Further research is needed to clarify the exact mechanisms in these interesting phenomena.

It is to be noted that in all of the described applications multi-mode elements, such as MWNTs, are essential. Multi-walled carbon nanotubes also have the advantage of being less susceptible than SWNTs to potential fluctuations along the length of the nanotube, which can adversely affect device characteristics.

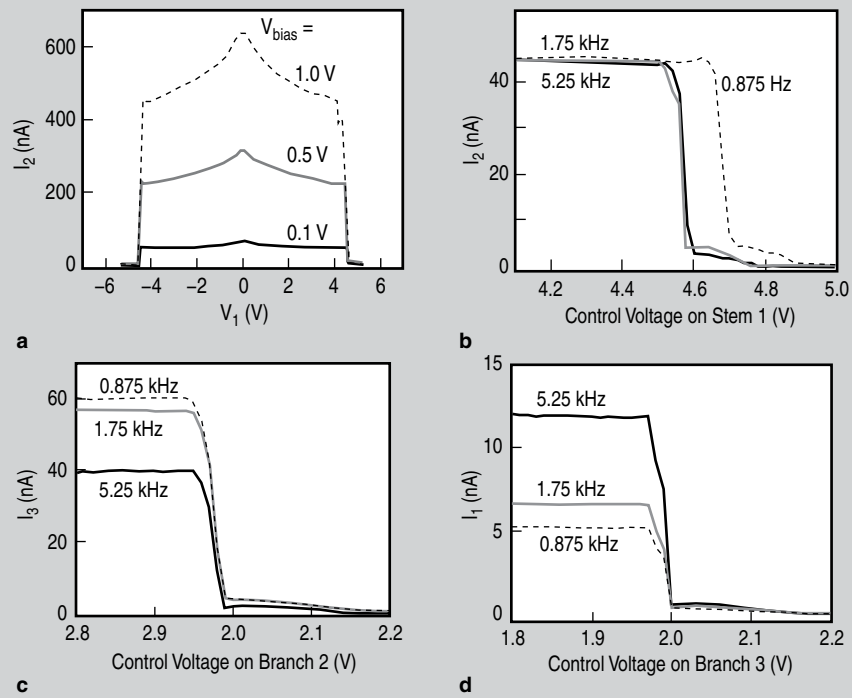


Figure 6. An observation of near-perfect electrical switching in Y-junctions. (a) An abrupt modulation of the electrical current through two branches of the Y-junction is seen on varying the voltage on the third branch. The voltage at which the switching action occurs on the stem (a and b) is ~4.6 V, while the turn-off voltages on the two branches are ~2 V (c and d). The frequency response of the Y-CNTs is also indicated. In (b) the control voltage is applied on the stem (1) of the Y-junction while the current, under an AC bias, is monitored across the other two terminals of the Y-junction. In (c) and (d), a control d.c. voltage is applied on branches 2 and 3.

OTHER APPLICATIONS

The notion of nonlinearity in a structure brings forth a dazzling array of potentialities. Some of these set the stage for a multifunctional use of CNTs. For example, the curvature[47] of CNTs could lead to the creation of new electron-lattice vibration scattering channels and consequent attractive electron-phonon interactions[48] (according to the Bardeen–Cooper–Schrieffer theory) can induce superconductivity. It was also proposed that the superconducting transition temperature (T_c) can be enhanced by chemical doping of the nanotubes. However, the reported T_cs are quite low (~15 K).[49]

The corresponding author has recently formulated that quantum dots placed in the channel of an MWNT Y-junction offer a natural template for exploring the effects of reduced dimensionality toward fabricating the best thermoelectric[50] material. Higher thermopower (S) values can be obtained[51] on MWNTs containing catalyst particles, through the Kondo resonance effect. The interaction of the iron, cobalt, and nickel nanoparticles, inevitably present due to catalytic synthesis, with the electron spins in the nanotubes could enhance S and thermoelectric efficiency.

The chemical inertness of SWNTs makes covalent attachment difficult. In MWNTs the presence of defects and catalyst particles can be used for chemical sensing, by appropriate functionalization to achieve specificity. As one example, a helical configuration of the streptavidin protein, useful in many biochemical assays, could be crystallized[52] on a helically shaped MWNT surface through both steric and hydrophobic effects.[53] It is also possible to attach carboxylic acid groups resulting in a hydrophilic nanotube surface preventing protein attachment.[54] While the current detection level is around ten streptavidin molecules, higher sensitivity can be achieved through a better control of the surface charges.

OUTLOOK FOR THE FUTURE

Richard Smalley, one of the pioneers of nano-carbon related research, was said to have remarked, "These nanotubes are so beautiful that they must be useful for something." This article has attempted to illustrate some of these possibilities in branched and helical CNT structures. One can also envision a more ambitious scheme and circuit topology—where both interconnect and circuit elements are based on nanotube structures, realizing true nanoelectronics.

Carbon-based electronic materials promise greater flexibility compared to conventional inorganic electronics—as exemplified by the large variety of structures in organic chemistry. However, issues remain in their widespread application, necessitating effort from both a fundamental and technological perspective.

One issue is the controlled doping of nanotubes. It is generally found that when connected to external contacts, in the ambient, CNTs are measured to be p-type. This characteristic could be induced by exposure to oxygen and/or the higher work function of the contact[13] whereby holes could be generated in the nanotube due to electron transfer from the NT to the contact. Annealing in vacuum[55]

through oxygen removal converts p-type devices to n-type. Considering that the widespread utility of semiconductors in modern devices arises from their ability to be controllably doped, the underlying mechanisms still need to be investigated. These issues are especially pertinent in that charged defects, either from the substrate or adsorbates from the ambient, are also known to modify electronic properties.

An issue of outstanding importance for the practical application of nanotubes, indeed in nanotechnology itself, is that of aligning and fabricating large-scale arrays of nanostructured elements. While no satisfactory methods exist, some promising ones include the use of electric fields[56] and microfluidic techniques.[57] Large-scale assembly of CNTs, at a level that would be impressive to a systems designer,[58] is far from feasible.

The issue of power generation and transfer at the nanoscale,[59] using nanowires and nanotubes, makes for some of the most exciting research at the frontiers incorporating high frequency (THz scale) electronics.[33]

Silicon technology, over the years, has proven to be very robust and adaptable and CNTs could be embraced into the fold as just another nanostructured material, say for interconnects. In order for CNTs to stand on their own in nanoelectronics, it is necessary to demonstrate novel functionality as can be obtained in the nonlinear CNT forms. In this connection, a fundamental analysis of what new technologies/devices are possible in reduced dimensions, using nanotubes, is needed. A few such examples are the recent analyses of the high frequency properties[33] and array-based architectures[58] for CNT electronics. Forays in such directions are necessary and would greatly contribute to the vitality of CNT electronics.

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