Electron Transport Through Metal–Multiwall Carbon Nanotube Interfaces

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Abstract—**In this paper, we examine mechanisms of electron transport across the metal–carbon nanotube (CNT) interface for two different types of multiwall carbon nanotube (MWNT) architectures, horizontal or side-contacted MWNTs and vertical or end-contacted MWNTs. Horizontally aligned nanotube growth and electrical characteristics are examined with respect to their potential applications in silicon-based technologies. Recent advances in the synthesis techniques of vertical MWNTs have also enhanced the possibility for a manufacturable solution incorporating this novel material as on-chip interconnects or vias as copper interconnect feature sizes are scaled into the sub-100-nm regime. A vertical MWNT architecture is presented that may be suitable for integration into silicon-based technologies. The growth method for this architecture and its effect on electrical characteristics are examined. Through simulations, dc measurements, and comparison of our results with previous studies, we explain why high contact resistance is observed in metal–CNT–metal systems.**

Index Terms—**Carbon nanotube (CNT), contact resistance, on-chip interconnect.**

I. INTRODUCTION

CARBON NANOTUBES (CNTs) exhibit unique electronic properties and extraordinary mechanical properties and have received much attention in the nanoelectronics community. Fabrication of field-effect transistors using single-walled carbon nanotubes (SWNTs) as conducting channels has been demonstrated [1], [2]. The very high current-carrying capacity of multiwall carbon nanotubes (MWNTs) has prompted investigations of their use as interconnects in silicon technology [3]. In all of these applications, the nanotube–metal contact resistance is an important parameter that needs to be minimized. Unlike the near-ohmic behavior of metal contacts and interconnects, contact resistance at the metal–CNT junction dominates the electrical performance of CNT wires. This interaction is largely dependent on physical design and process parameters such as electrical contact area [4], different electrode materials used [4]–[7], and the structure of the concentric shells within the MWNT. The ability to consistently fabricate high aspect-ratio structures such as vertical MWNTs makes them an excellent candidate to complement and improve upon copper-based interconnect technology. Successful integration of such architectures into current fabrication processes, however, requires that, among others, contact resistance be minimized so that the full benefit of ballistic transport in MWNTs can be exploited.

Growth processes can define both electrical and mechanical characteristics of a nanotube structure. The methods in which CNTs are fabricated have a direct impact on the contact characteristics between a metal electrode and multiwall nanotubes. The primary growth methods for MWNTs include arc discharge and chemical vapor deposition (CVD). In Section II, we explain the different types of MWNT growth methods and explore the structure–property relationship between the method of growth and electron transport across metal–CNT junctions. The various growth techniques and their effects on electrical characteristics of the MWNT are explored.

It is important to note that, for both side-contacted and end-contacted nanotubes (Figs. 1 and 2), we expect distinctly different mechanisms of electron transport across the metal–CNT interface. Side-contacted CNTs depend upon tunneling of electrons across a finite physical barrier created by van der Waals interaction at the metal–CNT interface [8]. The contact quality for end contacts depends on chemical (chemisorption) and/or physical (physisorption) processes at the nanotube tip. The implication of the two MWNT architectures on contact quality is discussed in Section III.

Measured resistance in metal–CNT–metal systems varies widely among experiments and is often much larger than its theoretical value [9]. Since high metal–CNT contact resistance limits the practical application of CNTs, it is important to be able to make reliable low-resistance contacts. Some novel ideas for optimizing measurement techniques and minimizing contact resistance have been presented in previous studies, such as electron bombardment used to enhance transmission of electrons between the metal and CNT [11], and using ropes of SWNTs to conduct four-point probe measurements in order to deembed probe resistance, subsequently reducing total resistance [12]. For the type of geometry presented in [11] and [12], tight-binding calculations [4], [5] have shown that contact area affects the transmission of electrons across the side-contacted metal–CNT junction, hence, directly impacting contact resistance. The configurations presented in [11] and [12] can be characterized as being side contacted, similar to state-of-the-art on-chip copper interconnects. Simulation results obtained by generalizing metal–insulator–metal (MIM) theory [13] for side-contacted metal–CNT systems are shown in Section IV.

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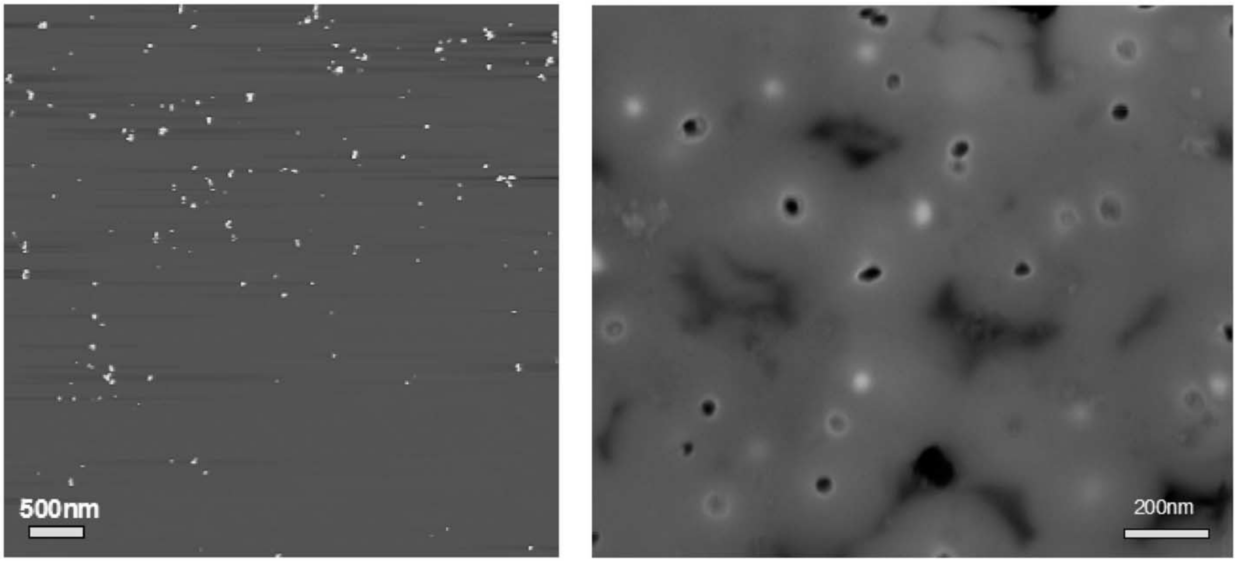

Fig. 1. (a) Current-sensing AFM image of end-contacted MWNT configuration. (b) Scanning electron microscope (SEM) top view. In (a), the lighter areas represent MWNTs embedded in an SiO_2 matrix (represented by the dark background). In (b), the circular structures represent top views of vertical MWNTs.

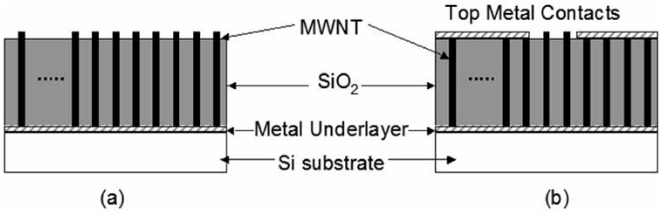

Fig. 2. Schematic cross section of: (a) pre-metal deposition and (b) post-metal deposition vertical MWNT.

Analyzing metal–CNT contact characteristics of vertical nanotube structures requires a different fundamental approach to understand electron transport at the interface. Vertically aligned nanotubes [14], shown in Fig. 2(a), can be characterized as having end-contacted geometry since electrically connecting these structures requires a conductive material contacting the end of the MWNT. Following the deposition of metal electrodes over a finite area of vertical MWNTs, as shown in Fig. 2(b), we demonstrate through resistance characterization the possibility of using vertically aligned CNTs for on-chip interconnect applications. The details and results of the experiment to obtain dc measurements are discussed in Section V. In Section VI, we compile some previous studies of resistance in MWNT systems and analyze these results with respect to the theory and experiment presented in Sections II–V.

II. MWNT GROWTH

Creating multiwall nanotubes using the arc-discharge method [16] produces high-quality samples of 2–200 nm in diameter and 1–100 μm in length. The parallel nature of the nanotube walls demonstrated by Iijima [16] is ideal for facilitating ballistic conduction within the CNT. MWNTs grown using the arc-discharge method suffer from two drawbacks when applied to interconnect applications. Arc-discharge-grown nanotubes are produced in soot form, mixed with various other forms of carbon materials, making it necessary for additional processing steps such as purification, sonication, and dispersion onto a substrate to be performed. In addition, nanotubes grown using this method are primarily laid horizontally on a flat substrate in a slow and random process, restricting the nanotubes to side-contacted electron transport across a Schottky barrier formed at the metal–CNT junction. To circumvent such issues in ultra-large scale integration (ULSI), a more robust and consistent architecture for nanotube growth must be used.

CVD growth of nanotubes [3], [14] has been shown to yield promising electrical characteristics for integration into on-chip interconnect systems. The ability to grow well-aligned nanotube arrays using standard microelectronic fabrication techniques [17]–[19], [21] opens up many possibilities for applications to silicon technology. Within the broad category of CVD growth of MWNTs, there are two variations that have been discussed in the literature: plasma-enhanced chemical vapor deposition (PECVD), and thermal CVD. The growth of CNTs using thermal CVD normally results in “spaghetti-like” CNTs, as shown in [3]. In the PECVD process, an additional measure of nanotube orientation control is introduced by inducing the electric field normal to the substrate surface such that the direction of the nanotube can be uniformly aligned along the direction of the electric field [19]. We must note, however, there are some properties of CNTs grown by the CVD process that are not desirable for electronic applications. Undesired structural properties resulting from the PECVD process can be seen at the nanotube tip using transmission electron microscope (TEM) analysis of the nanotube. The pear-shaped catalyst particle reveals that the concentric shells of a CNT grown using the PECVD process form a bamboo-like structure, which is observed throughout the entire length of the nanotube [20], [21]. This type of structure obtained from plasma-enhanced growth was examined in [21] by increasing capacitive bias on the substrate, which resulted in an increase in atomic hydrogen intensity. The unique physical property of the PECVD-grown nanotube inhibits ballistic conduction through the MWNT by forcing electrons to “hop” across multiple graphitic layers to facilitate electrical conduction along the nanotube axis.

III. IMPLICATIONS OF CONTACT GEOMETRIES ON THE RESISTANCE OF MULTIWALL NANOTUBES

A. *Side-Contacted Metal–CNT Junction*

Contact resistance in traditional CMOS on-chip interconnect technology has long been modeled by studying well-known interfaces between different types of bulk materials. The merits of such models can be validated through experimental techniques such as kelvin structures for resistivity measurements [22]. In deep submicrometer circuits, current crowding is a phenomenon seen at the narrowing or bend of a copper interconnect line or via, causing carriers to flow predominantly near the "corners" of such a structure. Since CNTs are treated as one-dimensional systems, the current-crowding mechanism is less likely to be observed in metal–CNT systems.

The transport mechanism across a side-contacted metal–CNT junction is explained in [6] as tunneling across an energy barrier created by the finite separation between the gold electrode and MWNT. Combining this theory with observations that the wave function orthogonal to the nanotube axis decays rapidly away from the CNT surface [5], we conclude that electron injection into the nanotube is accomplished by quantum-mechanical tunneling through this physically induced energy barrier at the metal–CNT junction. Since the physical separation between metal and CNT is almost the same as the bond length between carbon and the metal atom, tunneling depends on the chemical composition and configuration of electronic states at the surface. The wave-function overlap is reduced by this gap-induced energy barrier, and transmission is decreased for side-contacted CNTs with small contact area. For large contact area, electronic structure calculations suggest better electronic coupling between the electrode material and MWNT [4], [5].

B. *End-Contacted Metal–CNT Junction*

Similar to the side-contacted geometry, the chemistry at the end-contacted metal–CNT junction must be well understood. The TEM analysis of the MWNT tip structure allows us to understand how the physical properties of the nanotube affect electron movement through metal–CNT–metal systems. The presence of dangling bonds at the end of a multiwall nanotube after oxide planarization provides an opportunity for chemisorption of the CNT by the electrode material to occur. The end-contacted structure shown in Fig. 2 represents a possible architecture for using MWNTs as on-chip interconnects. The size and vertical nature of the aligned array make them good candidates for use as vias, bypassing processing problems for high aspect-ratio structures with current silicon-based technology, while retaining desirable electronic properties. In addition, this bottom-up approach for synthesizing CNTs allows for silicon-compatible processing, ideal for integration into current wafer manufacturing processes. As MWNT growth processes mature, the initial step for implementing them into silicon-based systems would be to replace copper via technology with a similar CNT-based via structure [3].

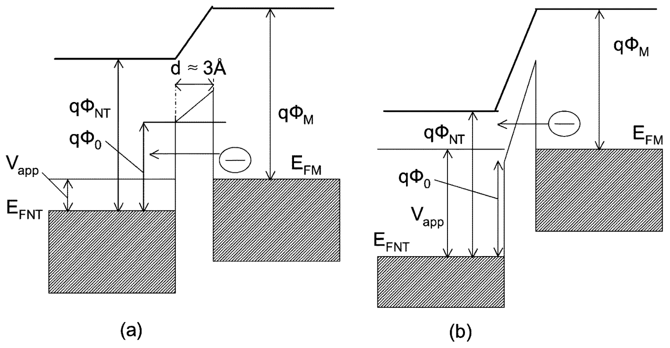

Fig. 3. Energy-band diagram for metal–CNT interface under different bias conditions. The main parameters for modeling this junction as a MIM junction are: finite separation distance (d), the energy barrier height (Φ_0), and the electrode work function ($E_{\rm FM}$). (a) $0 < V_{\rm app} < \Phi_0$. (b) $V_{\rm app} > \Phi_0$.

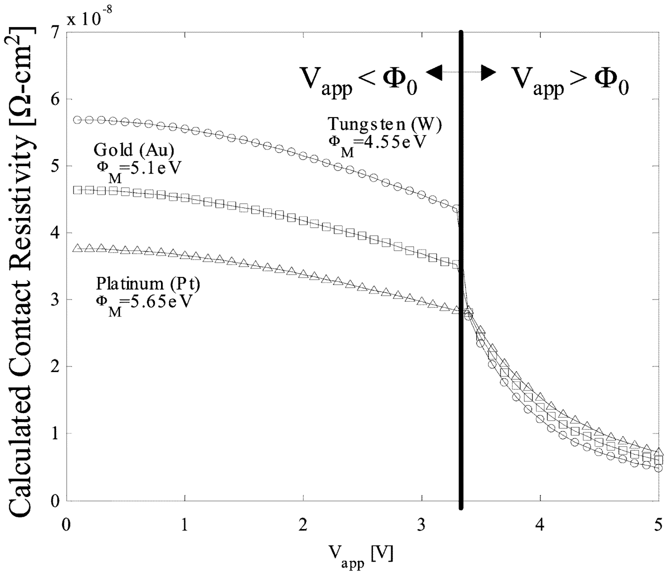

Fig. 4. Calculated contact resistivity across the metal–CNT interface using MIM tunneling formulation. When higher voltage is applied ($V_{\rm app} > \Phi_0$), the effects of FN tunneling will be observed.

IV. SIMULATION OF CONTACT RESISTIVITY VERSUS APPLIED VOLTAGE FOR SIDE-CONTACTED GEOMETRIES

Fig. 3 shows an energy-band diagram illustrating how electron transport across the metal–CNT junction is achieved. To predict the behavior of this effect on contact resistance, we modify the approach presented in [13] for a MIM system. We know that bulk properties will not always be observed in thin-film systems such as the metal–CNT interface under consideration. We can, however, gain some insight by finding a correlation between different tunneling mechanisms at the interface and how they affect contact resistivity. Contact resistivity is presented in the context of applied voltage across the nanotube. The band diagram for the contact–CNT interface in Fig. 3 is for two different applied bias conditions. The finite separation was approximated [8] by a van der Waals interaction at the metal–CNT interface. For our analysis, we neglect hot-electron effects such as thermionic emission that is more prominent in transistors. The following closed-form expression used to describe this junction was derived from the tunneling

TABLE I
SINGLE MWNT RESISTANCE IN RECENT STUDIES

Electrode Configuration (metal-CNT configuration)	Electrode Material	Overall Resistance	Reference
Metal-on-tube (side-contact)	W	1.7kΩ‡	[10]
Metal-on-tube (side-contact)	Ti	28kΩ	[29]
Metal-on-tube (side-contact)	Pt/Au	13kΩ	[29]
Metal-on-tube (side-contact)	Pt*	50kΩ	[26]
Point contact (end contact)	Pt†	50kΩ-300kΩ	[14,15]
Buried nanotube (side + end contact)	Ti/Au	~500Ω	[27]
Deposited metal contact (end-contact)	Cr	11kΩ	This work

*Actual contact is made to platinum via pre-deposited aluminum pads

†Electrode is a platinum covered, current-sensing atomic force microscope (CSAFM) tip, second contact is chromium

‡ Four-terminal measurement

current density equation presented in [24], originally used to analyze metal–semiconductor heterojunctions:

$$J = \frac{q}{4\pi^2\hbar} \iint T_t \big[F_1(E) - F_2(E)\big]\, dk^2\, dE. \qquad (1)$$

Here, T_t is the tunneling probability and F_1 and F_2 are the Fermi distribution functions in the conducting regions (MWNT and metal contact). We integrate over the wave vector and energy domains where tunneling occurs.

Three electrode materials, gold, platinum, and tungsten are studied using this method. The work-function differences in these materials show a distinct effect on contact resistance. Fig. 4 presents results obtained based on (1). Most dc current–voltage (*I–V*) measurements in the low-bias range (<1 V) have shown that contact resistance values remain relatively constant with respect to applied voltage. According to our bias-dependent contact resistivity predictions, this is expected behavior at low bias. At high bias, electron transport resembles Fowler–Nordheim (FN) [25] tunneling, as depicted in Fig. 3(b). Reference [10] reports data for resistance in the high-bias range (25 V). At this applied voltage, our tunneling model would predict a marked decrease in contact resistivity, consistent with the high-bias result observed [10]. Whether CNT wires will be biased at such high voltages for interconnect applications is yet unknown. However, to realize the full potential range of applications for MWNTs, it is imperative to study the dependence of R_C over a wide range of applied voltages.

V. COMPARISON AND ANALYSIS OF RESISTANCE IN MWNT SYSTEMS

The work presented in [7] reflects a systematic approach for analyzing the dependence of CNT resistance on a variety of different factors including electrode metal, bonding configuration, and the type of nanotube (SWNT or MWNT). Our research summarizes more recent studies, as well as the work presented here, that focus on MWNT systems and discusses the manner in which the electrode was configured with respect to the CNT. Recognizing that some of the experiments summarized in Table I were not specifically targeted for contact resistance measurement, we will qualify our discussion accordingly.

Table I reveals the many variations seen for the metal-on-tube (MOT) configuration. In all of the MOT studies, the nanotube was placed on a silicon substrate, and metallic contacts were deposited using either focused ion-beam lithography [10], [26], or e-beam lithography [27]. The high resistance observed for titanium contact [29] is a result of the coulomb blockade (CB) effect and the material properties of titanium, a metal susceptible to oxidation when exposed to air. For realistic integrated circuit (IC) applications, however, CNTs should be studied under ambient conditions. Considering only direct and FN tunneling, materials with higher work functions contributes to more efficient electron injection, thereby exhibiting more desirable resistance properties for side-contacted geometry. Fig. 5 shows an energy-band diagram depicting the work-function dependence of electron injection into the CNT. Low work-function materials, such as tungsten, suffer from higher resistance across the interface as compared to high work-function materials because higher voltages need to be applied to inject electrons into the nanotube. To induce an electric field that facilitates tunneling into the nanotube, as in Fig. 5(b), a sufficient voltage $(V_{\mathrm{app}} > \Phi_{\mathrm{NT}} - \Phi_W)$ needs to be applied to the tungsten electrode.

Work function is by no means the exclusive factor in determining differences in contact resistance. The quality and method in which the contacts were fabricated must also be analyzed to interpret the results of these experiments. Perhaps the most striking study was [27] due to the small value obtained for CNT resistance. The authors introduced a method for "burying" the MWNT under a layered contact constructed using titanium and gold. The resulting low resistance indicates that multiple walls, i.e., concentric nanotubes, are being contacted in parallel. To enable us to bring the resistance down to a manageable level for practical use in on-chip interconnect

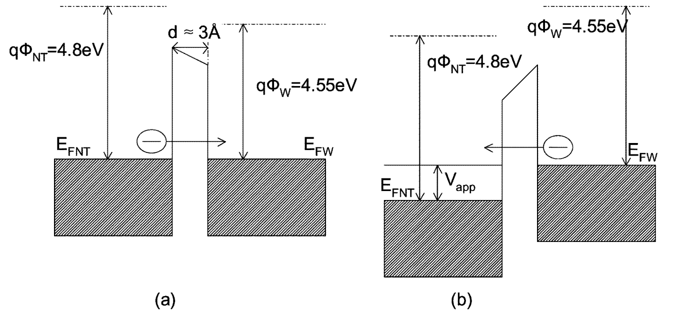

Fig. 5. Energy-band diagram for CNT–tungsten interface under different bias conditions. (a) Effect of the work-function difference between tungsten ($\Phi_W = 4.55$ eV) and CNT ($\Phi_{\mathrm{CNT}} = 4.8$ eV [28]). (b) Bias condition sufficient for facilitating tunneling into the CNT. (a) $V_{\mathrm{app}} = 0V$. (b) $V_{\mathrm{app}} > (\Phi_{\mathrm{NT}} - \Phi_W)$.

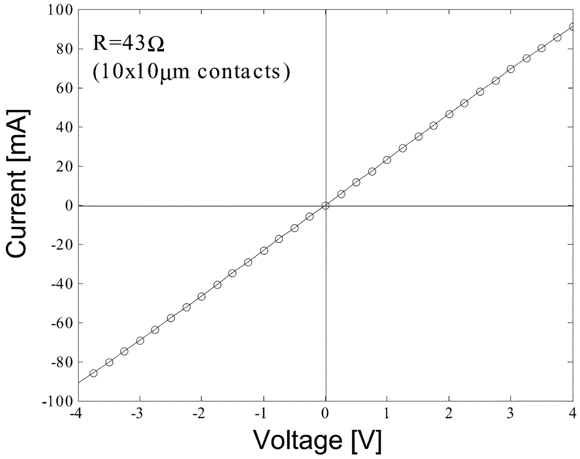

Fig. 6. Typical *I–V* curve for parallel MWNTs connected in series through two 10×10 μm chromium contact pads.

systems, methods need to be developed to optimize the number of walls that couple to the metal contact.

VI. CHARACTERIZATION OF PECVD-GROWN VERTICAL MWNTS FOR INTERCONNECT APPLICATIONS

To demonstrate the viability of implementing CNTs grown using the PECVD process in interconnect systems, we present results of electrical characterization of multiple MWNTs connected in parallel. For our study, we use 2000 Å of chromium as the metal on which the catalyst film is patterned. Nanotubes are then grown using the PECVD process described in Section II. For consistency, we also use chromium as the top metal layer. The completed structure looks similar to the schematic representation shown in Fig. 2. The top deposited metal layer was patterned using e-beam lithography to obtain 10×10 μm^2 chromium squares with a thickness of 2000 Å that make contact to the tips of the MWNTs. Fig. 6 shows a typical *I–V* curve of nanotubes contacted through two contact pads. The near-ohmic behavior of the curve suggests that parallel MWNTs may provide performance comparable to copper vias for on-chip interconnect architectures without the processing difficulties presented by current state-of-the-art copper deposition technology. The sheet resistance of the chromium underlayer was also considered in our study. An area of bare chromium, subjected to the same growth conditions as the CNTs, was characterized using two- and four-terminal measurements. The resulting chromium resistance for the two-terminal measurement was 10–15 Ω, while the four-terminal measurement yielded 4–6 Ω, most of which can be attributed to the contact–Cr interface. While these resistance values seem small, if we compare the two-terminal *I–V* measurement of the nanotubes to the contact–Cr resistance, we can see that the proportion of contact resistance to overall resistance is quite high.

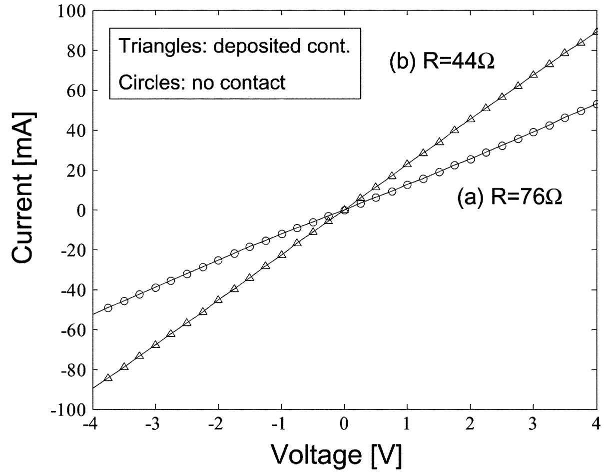

Fig. 7. *I–V* curve comparing parallel MWNTs: (a) contacted directly with a 25-μm-diameter probe and (b) with deposited 10×10 μm contact pads.

The importance of depositing a quality contact becomes apparent when we compare directly contacting parallel MWNTs with a 25-μm-diameter tungsten probe tip to contacting nanotubes through the patterned 10×10 μm^2 contacts. Fig. 7 shows the stark contrast in resistance values when considering the deposition of metal contacts over multiple nanotubes. This result suggests that there is a distinct chemical and physical interaction between the deposited top metal layer and the end of the MWNT that creates a less resistive metal–CNT junction. Dangling bonds at the nanotube tip after chemical mechanical polishing (CMP) enhance the possibility of creating a quality metal–CNT contact using metal deposition. A similar study using thermal CVD to grow nanotubes has been conducted in [3], however, the structural [23] and electrical integrity [14] of such devices under standard microelectronic fabrication techniques such as CMP must also be considered.

An inherent obstacle for characterizing single nanotubes is the limitation of currently available instruments, specifically the atomic force microscope (AFM). Typical AFM probe tips to perform *I–V* measurements are ~10 nm in diameter. The MWNTs under study here are 50–100 nm in diameter, as can be seen in Fig. 1(b). The mismatch of the probe tip and MWNT diameter alters the direct measurement of electrical characteristics (i.e., AFM probe to the end of a MWNT). Resistances of 50–300 kΩ [14] have been reported using this methodology. In this study, we use a statistical approach by measuring the *I–V* curve of many parallel nanotubes. Using current-sensing atomic force microscope (CSAFM) imaging, we can approximate the number of nanotubes per unit area. From our analysis, there are 5–6 MWNTs per 1 μm^2, corresponding to a resistance of 11 kΩ–13.2 kΩ per single nanotube since we are measuring two sets of MWNTs in series. This type of analysis for single and multiple MWNTs has direct relevance to application of nanotubes to interconnect systems since the structure used in this study is similar to copper via technology.

VII. CONCLUSION

We have presented an analysis of transport mechanisms related to contact resistance in metal–CNT systems. Our study has compared and contrasted the different mechanisms of electron transport across the contact interface. Based on our analysis, a MIM model traditionally used for two conductive materials separated by an ultra-thin barrier layer has been proposed. Simulation and experimental data have been presented to demonstrate the utility of MWNTs for on-chip interconnect systems.

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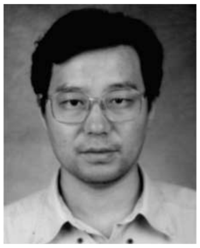

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