Passive electrical properties of multi-walled carbon nanotubes up to 0.1 THz

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Abstract. Metallic carbon nanotubes (CNTs) are promising as transmission lines or interconnects in radio-frequency nanoelectric circuits. This paper presents passive network properties of individual multi-walled carbon nanotubes (MWNTs) up to 110 GHz measured at room temperature. From the S-parameter data, frequency-dependent electric properties of the MWNT were extracted using an equivalent R-L-C circuit model. The ac impedance of the MWNT decreases significantly with increasing frequency, as predicted by earlier theoretical work. In particular, the equivalent resistance decreases a few hundred times. Our findings show that MWNTs can carry high-frequency currents much better than dc.
1. Introduction

Metallic carbon nanotubes (CNTs) possess great potential as transmission lines, interconnects or passive components in nanoelectronic circuits, because their current density, thermal stability and mechanical properties greatly surpass those of conventional metal wires. Whereas it is difficult to control chirality and thus the metallic properties of single-walled carbon nanotubes (SWNTs) during production processes, multi-walled carbon nanotubes (MWNTs) are considered to be metallic. With the rigidity enhanced by multi-layered tubes and current density exceeding $10^{10}$ A cm$^{-2}$ [1], there is much optimism that MWNTs could be a solution to overcome issues such as reliability, scattering and signal loss in metallic nanoscale wires. In particular, as research activities to utilize millimetre waves grow, understanding properties of MWNTs in the microwave range is valuable.

Compared with the active CNT properties, their passive properties in the microwave range have been less investigated. Theoretical work predicted significant frequency dependence in the quantum point contact [2], and the dynamical impedance of both MWNTs [3] and SWNTs [4]. However, in an experiment using a single SWNT up to 10 GHz, significant frequency dependency was not observed [5]. On the other hand, an experiment using a bundle of SWNTs up to 20 GHz reported frequency-dependent equivalent-resistance of the SWNT bundle [6]. This discrepancy may have come from various factors, such as different sample structures and fabrication processes leading to different scattering and contact resistance, or effects of applied voltage on semiconducting SWNTs included in the bundle.

Due to the complexity of theoretical modeling and unconventional metallic states of MWNTs, electronic characteristics of MWNTs are not well-understood, and most of what has been found is on their dc properties. A recent experiment on the frequency-dependent MWNT impedance was limited to 200 kHz [7]. Also, from the $I-V$ data measured with very narrow electrode gaps of 0.05–1 µm, it is impossible to distinguish parasitic effects due to electrodes and contacts and the intrinsic MWNT properties.

In this work, we successfully measured the two-port network properties of individual MWNTs at room temperature, up to 110 GHz. Furthermore, from the measured S-parameter data, individual electric properties (rather than just the overall impedance) of MWNTs are extracted using equivalent R-L-C circuit modeling. Our results support previous theoretical predictions that the dynamic impedance of MWNTs significantly depends on the frequency. Especially, the effective resistance of an MWNT decreases hundreds of times as frequency increases from 0.5 to 110 GHz.
Figure 1. A sample prepared for two-port S-parameter measurements (a) TEM image of a selected CNT, (b) SEM image of the CNT part of the sample, which shows an MWNT connecting the IN/OUT electrodes.

2. Experiment

MWNTs were grown by a plasma-enhanced chemical-vapor depositions (PECVD) technique. They were estimated to be about 8 $\mu$m long, with about 21–25 shells, inner diameter of 2 nm, and outer diameter of about 18–20 nm, as can be seen from TEM images (figure 1(a)). The probing pads consist of two ground electrodes IN and OUT, compatible with ground–signal–ground probing. The design and fabrication of probing pads is very important to reduce the effects from contact resistance and measurement noise. Especially to reduce the background noise, a 7000 Å-thick oxide layer was deposited on a silicon wafer. To remove any impurities from the surface, the Si wafer was cleaned in $\text{H}_2\text{SO}_4/\text{H}_2\text{O}_2$ (3:1) solution. In order to improve contact with the MWNT, the configuration of the IN/OUT electrodes was specially designed to be a gradually narrowed shape toward the MWNT. The electrodes were made of Cr/Au (5/150 nm) bilayer at a low deposition rate (0.01 nm s$^{-1}$), in order to accommodate the full wrapping of the MWNT by Au. Figure 1(b) shows the connection of both the IN and OUT electrodes by an MWNT. The gap between the electrodes is 4 $\mu$m, and the MWNT extends into the electrode about 2 $\mu$m on either side. For comparison and deembedding of the parasitic effects from the measured data, the identical pad structures without an MWNT (called hereafter open), and with an Au wire of 2 $\mu$m in diameter (called short), respectively, were also fabricated. S-parameter data of samples with the MWNT, open and short samples were measured from 0.5 to 110 GHz, using the network analyzer HP8510XF at $-20$ dBm, which corresponds to 10 $\mu$W. Figure 2(a) shows the measurement setup. The results were independent of the power at this level. S-parameter $S_{ij}$ is found by applying an incident wave of voltage to port $j$ and measuring the reflected wave amplitude coming out of port $i$, while setting incident voltage to all the other ports except port $j$ to zero.

Figure 2(b) shows an equivalent circuit model for the samples with an MWNT. Both $R_p$, $C_p$ denote the resistance and capacitance of the probing pads. $R_c$, $C_c$ denote the contact resistance and capacitance due to the electron transport barriers between the MWNT and Au,

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5 On-wafer vector network analyzer calibration and measurements, application note, Cascade Microtech.
Figure 2. Measurements and modelling (a) schematic diagram of the Cr/Au bilayer electrodes fabricated for ground–signal–ground probing, (b) equivalent circuit model for a sample with an MWNT. The dotted circle represents the MWNT part. $C_p$ and $R_p$ denote the capacitance and resistance of the probing pads, $C_{ps}$ denote the parasitic capacitance of the gap, $R_c$ and $C_c$ denote the contact resistance and capacitance between the MWNT and the electrodes, and $R, L, C$ are the resistance, inductance, capacitance of the MWNT.

and $R, L, C$ are the resistance, inductance, capacitance of the MWNT, respectively. A circuit model without $C_c$ has been suggested in the experiment involving a bundle of SWNTs [6]. Whereas the previous work provided only the overall two-terminal equivalent resistance of the bundled SWNTs, we use this model to derive the individual circuit elements of $R_c, C_c, R, L, C$. Unlike the transmission line model, figure 2(b) allows one to separately parameterize the contact resistance and the inherent CNT resistance. The inductive contribution of the electrodes is expected to be negligible ($\sim 50$ pH) compared with the MWNT.

3. Results and discussion

Figure 3 shows the S-parameter magnitude data of four samples with an MWNT, two open samples and two short samples. Our samples are symmetric two-port networks, (i.e. $S_{11} = S_{22}, S_{12} = S_{21}$), so $S_{11}$ and $S_{21}$ measurements provide the complete transmission characteristics of each sample at specific frequency. The graph $S_{21}$ suggests that the resistance of the MWNT dominates in the lower frequency region, whereas open samples mainly behave like a capacitor. Although, the noise and parasitic effects are more noticeable as the frequency increases and the differences between the data sets become smaller, the $S_{21}$ value of the samples with MWNT is always larger than the open samples. The values of the samples with an MWNT are about 19 dB smaller than the short samples. Considering that the cross-sectional area ratio of the MWNT and Au wire in the short sample is of the order of $10^{-4}$, this hints at the potential of MWNTs. We found that the responses of all the samples with NT were almost identical, which suggests our MWNT samples are structured similarly.

We note the apparent difference of our data from the SWNT experiment mentioned previously [5], which did not find the significant frequency dependence. In addition to the most important difference of SWNT versus MWNT, the one-port experiment [5] used the $S_{11}$
Figure 3. S-parameter data of samples with an MWNT, open samples (without MWNT), and short samples. (a) $S_{11}$ magnitude (dB) and phase (deg), (b) $S_{21}$ magnitude (dB) and phase (deg). They all show different trends, except that $S_{11}$ data are similar for samples with and without MWNT.

data from a SWNT connecting a pair of asymmetric pads up to 10 GHz, unlike our two-port experiment. Whereas their $S_{11} 0 \pm 0.02$ dB data was dB in magnitude with $\text{Im}(S_{11}) = 0 \pm 0.002$ up to 10 GHz, our magnitude is around $-0.3$ dB, with $\text{Im}(S_{11}) = 0.0365$ at 10 GHz. In the previous experiment, the small imaginary part compared to the real part is taken as a sign that the nanotube impedance is mostly real and does not depend on the frequency. The magnitude ratios between the real and imaginary parts of these two experiments seem similar (10:1 versus 9:1) in this relatively lower frequency region. However, the magnitude of our $S_{11}$ phase data increases with the frequency (figure 3(a inset)). Also, our $S_{11}$ data of samples with and without MWNT are very similar (figure 3(a)), but the $S_{21}$ data are distinct (figure 3(b)). In our two-port measurements, it seems that the $S_{11}$ data are affected more by the pads or parasitic effects, rather than by the MWNT, whereas the $S_{21}$ data contain rich information.

By applying the current and voltage law to the circuit model in figure 2(b), the relationship between the current and the voltage drop between the points 1 and 2 can be computed. The frequency-dependent matrix from those points $V_1$, $V_2$ to the currents $I_1$, $I_2$ (so-called admittance matrix) can be obtained as below:

$$
\begin{bmatrix}
I_1 \\
I_2
\end{bmatrix} =
\begin{bmatrix}
\hat{Y}_{11} & \hat{Y}_{12} \\
\hat{Y}_{21} & \hat{Y}_{22}
\end{bmatrix}
\begin{bmatrix}
V_1 \\
V_2
\end{bmatrix},
$$

$$
\hat{Y}_{12}(j\omega) = \hat{Y}_{21}(j\omega) = -\left(\frac{1}{Z(j\omega)} + j\omega C_p\right),
$$

$$
\hat{Y}_{11}(j\omega) = \hat{Y}_{22}(j\omega) = -\hat{Y}_{12}(j\omega) + \frac{j\omega C_p}{1 + j\omega R_p C_p},
$$

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Figure 4. Real and imaginary parts of the characteristic impedance of an MWNT, and its magnitude (inset). The impedance decreases significantly from 15 kΩ at 0.5 GHz to 0.2 kΩ at 110 GHz.

where

\[
Z(j\omega) = -\left(\frac{2R_c}{1 + j\omega R_c C_c} + \frac{R + j\omega L}{(j\omega)^2 LC + j\omega RC + 1}\right),
\]

represents the impedance of the CNT part, corresponding to the dotted circle in figure 2(b).

On the other hand, the measured S-parameter data \([S_{11}, S_{12}, S_{21}, S_{22}]\) can be converted to the \(Y\)-matrix data \([Y_{11}, Y_{12}, Y_{21}, Y_{22}]\) using the standard two-port network parameter relationships \([8]\). We fit the plot of \(Y_{21}\) with \(\hat{Y}_{21}\) using (1) and (2), i.e. we numerically obtained the set of \(R_c, C_c, R, L, C\) values that minimizes the error between the data and the circuit model over the frequency range.

The magnitude of the characteristic impedance \(Z(j\omega)\) is shown in figure 4 and figure 5 shows the \(\hat{Z}(j\omega)\) from the measurement and from the fitted model. The extracted circuit elements are shown in figure 6.

The open samples are considered to be the same circuit without \(R, L, C\) components. From the \(S_{11}\) magnitude/phase plots of the open sample, \(R_p = 7\) kΩ, \(C_p = 0.3\) pF, \(C_{ps} = 3\) fF were estimated. With these values, figure 5 compares the \(\hat{Z}(j\omega)\) from the measurement and from the fitted model using the relationship of (1). The magnitude data were fit very closely.

The monotone decrease in \(Z(j\omega)\) in figure 4 is somewhat compatible with the theoretical prediction for the LC-transmission line model or the overdamped SWNT dynamical impedance \([4]\), in that it decreases with respect to the frequency and the slope slows down around \(\omega = 2\pi \times 10^{10}\) rad s\(^{-1}\). The relationship \(\omega = \frac{R_{eff}}{L_{eff}}\) is satisfied with the effective resistance \(R_{eff}\) in the order of hundreds of \(\Omega\) and inductance \(L_{eff}\) in the order of nH, which is compatible with the high-frequency values of \(R\) and \(L\) given in figures 6(c) and (d). The real part dominates the overall characteristic impedance up to 10 GHz, as predicted for the SWNT \([4]\), but after that, the magnitude of real and imaginary parts are comparable.
For MWNTs, kinetic inductance of $0.1 - 4.2 \text{nH} \mu \text{m}^{-1}$ and capacitance of $31 - 111 \text{aF} \mu \text{m}^{-1}$ had been estimated [3]. In figure 6(d), the inductance of our MWNT samples were computed to be less than 1 nH when the frequency is higher than 30 GHz, much reduced from up to 15 nH at the lower frequency. In figure 6(e), the capacitance was reduced to around the order of 100 aF as frequency increases. Our results of $L \sim 1 \text{nH}$, given the effective length of 4 $\mu \text{m}$, are compatible with the predictions. If we quote the theoretical prediction $L \approx L_{\text{kin}} = R_q/(8N\nu_F)$ [9], with the quantum resistance $R_q = h/e^2$ and the Fermi velocity $\nu_F = 8 \times 10^5 \text{m s}^{-1}$, we can conclude that the number of conducting shells in our MWNT is $N = 16$, which corresponds to about two-thirds of the total number of shells. Our estimate of $C$ in the order of 0.1 to a few fF is slightly larger, and contact capacitance $C_c$ shown in figure 6(b) increases at very high frequencies, which may be due to further capacitive coupling with the SiO$_2$ substrate.

It was reported that the DC resistance of MWNT decreases when exposed to micro-waves [10]. It was also expected that the two-terminal equivalent resistance of CNTs will decrease with increasing frequency as all metallic layers take part in the conduction at very high frequencies. According to our results shown in figures 6(a) and (c), the values of $R_c$ and $R$ decrease with the frequency, compatible with the previous predictions. Since multiple pairs of $R_c$, $C_c$, $R$, $L$, $C$ values could exist that fit the measurement data equally well, and noise exists in the measurements, it is not possible to say whether the small jumps in figure 6 are due to resonant behavior or just noise.

Often the resistance of conventional metallic wires increases with respect to the frequency. When AC is applied to a metallic wire, there is a delay in the generated magnetic field to the current change and the current tends to be pushed towards the surface of the conductor. The current density decreases exponentially with depth from the conductor surface, and at frequency $\omega$, the depth $d$ at which the current density decays to 1/e is $d = \sqrt{\rho/\mu\omega}$, where $\rho$ is resistivity and $\mu$ is magnetic permeability [11]. This nonuniform current density causes an increase in the wire resistance. This effect increases as the frequency increases until at very high frequencies the entire current flows in a very thin skin on the wire. MWNTs will not be prone to this skin effect.
Whereas the intrinsic four-point resistance for a SWNT generated by electronic backscattering is $h/4e^2 = 6.5 \text{k}\Omega$, conductance of a MWNT depends on various factors such as the mean free path, number of shells that carry current, number of conducting modes per shell [12]. All these factors depend on the bias voltage, temperature and preparation of samples [13]. We conjecture that the effects of high frequency may be similar to the high bias voltage, in a sense that application of high-frequency currents to MWNTs promotes photon-assisted transport. Even the electrons with energy outside the gap region can contribute to the transport by absorbing or emitting the energy of photons, as the effect of microwaves increases. Only the outer shell is in direct contact with the electrodes initially, and the inner shells that are in indirect contact become activated as the input frequency increases. However, this experiment was done with MWNTs at room temperature, so it is difficult to think that the energy of 100 GHz currents alone caused such a drastic change in the CNT conductivity. Another possibility is local
heating at the contact points, which increases the temperature and excites the electrons to higher subbands, therefore increasing the carrier density.

Interestingly, $R_c$ and $R$ decrease with the frequency but this trend slows down around 30 GHz. This might be the regime above which the inner tubes are saturated and the effective number of conducting shells remains constant. To confirm this conjecture, further understanding of how the contact resistance at very high frequency can differ from the DC contact resistance will be essential.

Clearly, there are multiple ways to model MWNTs, and each can lead to different values of $R_c$, $C_c$, $R$, $L$, $C$. Also, their computation is sensitive to the measurement noise, which may worsen the error caused by the small magnitude of S-parameter data. In order to confirm whether the $R$, $L$, $C$ values computed here are indeed quantitatively exact characters of general MWNTs, it would be helpful to investigate samples made of various configurations and fabrication processes. Although further theoretical investigation on the interaction of shells and mean free path in the GHz/THz range is required. This study provides insight into analyzing the passive electric parameters of MWNTs.

4. Conclusions

We have presented the radio-frequency measurements of MWNTs up to 110 GHz in two-port experiments at room temperature. We applied an equivalent R-L-C circuit model to the measured S-parameter data in order to derive the dynamic impedance and passive electric properties (contact $R_c$, $C_c$ and inherent $R$, $L$, $C$ values) over the frequency range. The magnitude of the extracted $L$ and $C$ values are compatible with previous predictions for CNTs. We find that the dynamic impedance and the computed $R_c$ and $R$ values significantly decrease with increasing frequency, as predicted by earlier theoretical work. This clearly shows that MWNTs are promising as a transmission or interconnect line in the GHz and THz regime.

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