

# Analysis of Sensing Mechanisms in a Gold-Decorated SWNT Network DNA Biosensor

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**Abstract**—We show that carbon nanotube sensors with gold particles on the single-walled carbon nanotube (SWNT) network operate as Schottky barrier transistors, in which transistor action occurs primarily by varying the resistance of Au-SWNT junction rather than the channel conductance modulation. Transistor characteristics are calculated for the statistically simplified geometries, and the sensing mechanisms are analyzed by comparing the simulation results of the MOSFET model and Schottky junction model with the experimental data. We demonstrated that the semiconductor MOSFET effect cannot explain the experimental phenomena such as the very low limit of detection (LOD) and the logarithmic dependence of sensitivity to the DNA concentration. By building an asymmetric concentric-electrode model which consists of serially-connected segments of CNTFETs and Schottky diodes, we found that for a proper explanation of the experimental data, the work function shifts should be  $\sim 0.1$  eV for 100  $\mu\text{M}$  DNA concentration and  $\sim 0.4$  eV for 100  $\mu\text{M}$ .

**Index Terms**—Carbon nanotube, DNA sensor, Schottky barrier, work function, CNT network

## I. INTRODUCTION

Nano-biosensor technology, a convergence of biosensor and nano technologies has been emerged to be one of the most promising candidates to overcome the barriers of current biomedical issues such as single-molecule analysis, real-time detection, low-power consumption and miniaturization, to be used as in vivo applications [1, 2]. Among a number of approaches using various nanomaterials, single-walled carbon nanotubes (SWNTs) are the ultimate biosensor in this class for a couple of reasons: SWNTs have the smallest diameter ( $\sim 1$  nm), directly comparable to the size of single biomolecules. Furthermore, the one-dimensional (1-D) structure of carbon nanotubes (CNTs) allows signals to be propagated in a confined 1-D space, making them extremely sensitive to electrical and chemical changes in their immediate environments.

A drawback of sensors using a single SWNT is that its electrical properties are hard to control. For example, the energy gap of a nanotube is dependent on the tube diameter and the chiral vector, which implies the very high variability of electrical properties. In addition, the lack of reliable solutions to incorporate the biomaterials to the SWNT surface poses another challenge. A variety of biological molecules have been reported that can be bonded to CNTs [3, 4]. However, this often alters the intrinsic structure and properties of CNTs as well as those of attached biomolecules.

Recently, a new CNT-based electrical DNA biosensor system was reported by our group, which consists of gold (Au) nanoparticle-decorated SWNT network on top of concentric Au electrodes [5]. The Carbon Nanotube

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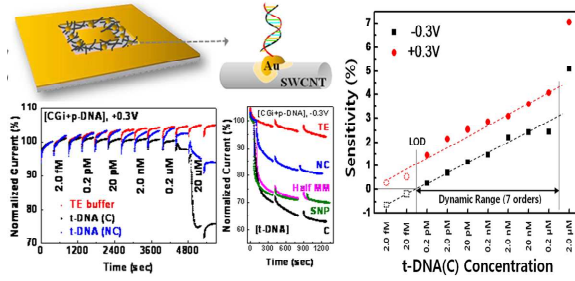
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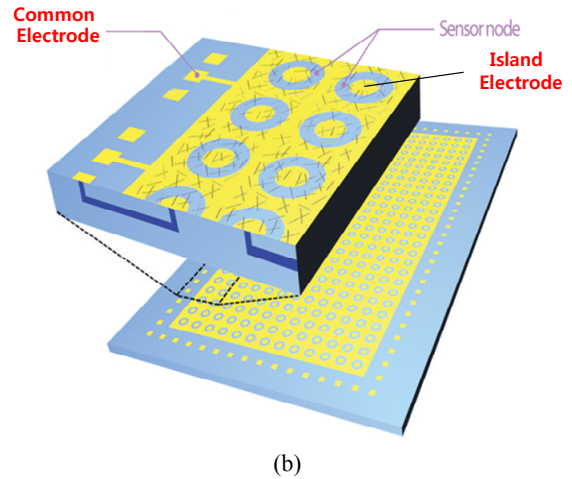
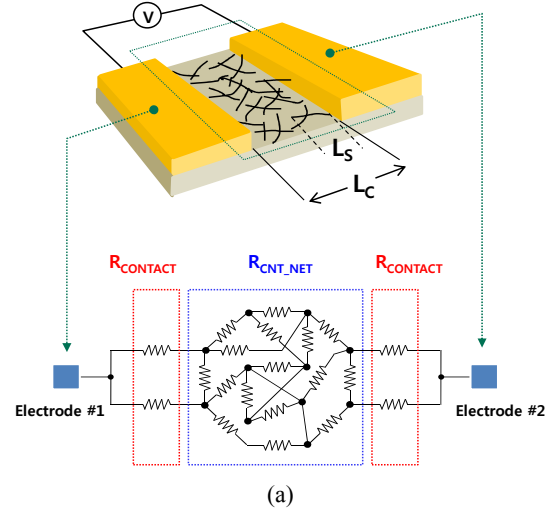
**Fig. 1.** DNA real-time measurement results and sensitivity curve using a gold-decorated SWNT network DNA Sensor [5].

Network Field-Effect Transistor (CNNFET) is made by using the SWNT network deposited on the concentric electrodes and in-between, to form an electrical channel of circular shape between the island electrode and the common enclosing one (see Figs. 1 and 2). Decorated Au nanoparticles on the SWNT network can be used as a platform or a linker for biomolecular sensor application by attaching probe molecules with thiol functional groups. They also provide better adhesion between the SWNT network and the chip substrate. Impressive features of this sensor include a very good sensitivity showing the limit of detection (LOD) of  $\sim 100$  fM and a very wide dynamic range as large as seven orders of magnitude of target molecule concentration as shown in Fig. 1.

In Sec. II, an electrical model of the CNNFET is presented. Specifically, a serially-connected segment model for an asymmetric electrode structure is introduced. Transistor's current-voltage ( $I$ - $V$ ) characteristics are calculated for statistically-simplified geometries and compared with the measurement results. In Sec. III, to match the experimental and simulation results, a metal-Schottky junction model is incorporated into the serially-connected segment model. With this model, we show that carbon nanotube sensors with gold particles on the SWNT network operate as Schottky barrier transistors, in which a transistor action occurs primarily by varying the contact resistance of Au-SWNT junction rather than the channel conductance modulation as in a conventional MOSFET. Experimental and simulation results are discussed in Sec. IV and conclusions follow in Sec. V.

## II. ELECTRICAL MODEL OF CNNFET

Changes in the conductivity of CNNFETs are mostly



**Fig. 2.** Schematic of randomly-networked CNTs and the geometries of electrode formations (a) The networked CNT resistors with bar-type electrode configuration, (b) The networked CNT resistors with concentric-type electrode configuration.

due to the gate-coupling effect and the Schottky-barrier effect. To model the CNNFETs, we need to simplify the structures from both a physical and a statistical point of view. The basic structure of DNA sensor is the percolating network of randomly-oriented carbon nanotubes, as shown in Fig. 2. In the “short-channel” limit of  $L_C < L_S$  (where  $L_C$  is the channel length and  $L_S$  the CNT stick length) and at low CNT density  $\rho_S$ , the CNTs behave as individual transistors connected in parallel bridging the source and drain electrodes. Therefore, the ratio of the transistor drain current  $I_D$  for any two bias points is independent of the geometry of the CNT networks. This implies that the scaling relationship can be written as,

$$I_{DS} = \frac{A}{L_S} \xi \left( \frac{L_S}{L_C}, \rho_S L_S^2 \right) f(V_{GS}, V_{DS}) \quad (1)$$

where the proportionality constant  $A$  depends on the gate capacitance  $C_{CNT}$  and the tube diameter  $d$ .  $\xi$  and  $f$  are functions of geometrical parameters  $L_C$ ,  $L_S$ , and  $\rho_S$  and bias conditions (drain-to-source voltage  $V_{DS}$  and gate-to-source voltage  $V_{GS}$ ), respectively [6, 7]. For long-channel CNTs with  $L_C > L_S$  (see Fig. 2(a)), the individual CNTs cannot bridge the channel by themselves and the CNT-to-CNT interaction ( $C_{ij}$  between the  $i$ -th and  $j$ -th CNT) becomes important.

When calculated using  $\sim 200$  statistical samples, the scaling formula of Eq. (1) still holds for arbitrary geometrical and biasing conditions even in the ‘‘long-channel limit’’ of  $C_{ij} \neq 0$ . Moreover, it was shown that the bias-dependent scaling function

$$f(V_{GS}, V_{DS}) = [(V_{GS} - V_{TH})V_{DS} - \beta V_{DS}^2] \quad (2)$$

is independent of geometrical parameters ( $\beta \sim 0.5$ ), again satisfying Eq. (1) [7]. This means that the voltage scaling function  $f(V_{GS}, V_{DS})$  would follow the classical square-law formula at very low densities ( $\rho \ll \rho_{th}$  and  $L_C < L_S$ ), and at very high densities ( $\rho \gg \rho_{th}$ ) where  $\rho_{th}$  is the percolation threshold. After all, for  $\rho \ll \rho_{th}$  and  $L_C < L_S$ , the CNT-to-CNT interaction is negligible, and the CNTs bridge the source and drain (S/D) directly, so that the system behaves as an independent collection of 1-D conductors. At  $\rho \gg \rho_{th}$ , the percolating network approximates a classical 2-D homogeneous thin film, and once again, the classical MOSFET formula should hold. These results imply that for any networked CNT sensor, only one CNNFET can model the current ratio of all the system.

A simplified diagram of the CNNFET is shown in Fig. 3. It consists of a networked CNT film as a semiconductor on top of an oxide as an insulator. Two metal contacts with the CNT film act as source and drain terminals. On top of the CNT film, there is a DNA membrane. For the biosensor application, the gate consists of a reference electrode and an electrolyte.

To solve the CNNFET’s current equation, the Poisson-Boltzmann (PB) equations involving the charge and potential drop in the DNA membrane and bulk electrolyte are employed as follows.

The PB equations for the electrolyte region and DNA

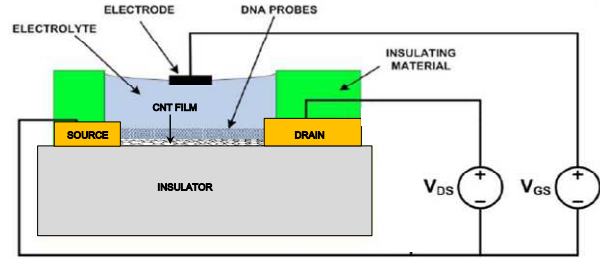


Fig. 3. Simplified diagram of the CNNFET for DNA sensor.

membrane region are given by [8, 9]

$$\frac{\partial^2 \psi}{\partial x^2} = \frac{2qz n_0}{\epsilon_{sol}} \sinh(\beta \psi(x)) \quad (3a)$$

$$\frac{\partial^2 \psi}{\partial x^2} = \frac{2qz n_0}{\epsilon_m} \sinh(\beta \psi(x)) - \frac{q \nu N_m}{\epsilon_m} \quad (3b)$$

where  $\beta = q/kT$ ,  $\epsilon_{sol}$  and  $\epsilon_m$  are the electrical permittivity of the electrolyte and DNA membrane, where the electrolyte contains anions and cations with concentration  $n_0$  and equal valence  $z$  (a  $z:z$  electrolyte). The membrane contains uniformly distributed fixed charges which have a valence  $\nu$  and a concentration  $N_m$ . These equations can be solved to give the surface potential of the electrolyte, denoted by  $\psi_0$ .

Here, let us assume the CNT film as the conventional semiconductor bulk. The electrostatic potential drop within the semiconductor CNT is related to the total charge density through Poisson’s equation, which, in 1-D, is given by

$$\frac{\partial^2 \psi}{\partial x^2} = -\frac{\rho(x)}{\epsilon_c} \quad (4)$$

where  $\epsilon_c$  is the permittivity of the CNT film. Furthermore, the charge density of the semiconductor is composed of three parts: fixed ionized-impurity charges, minority-carrier charges, and majority-carrier charges. Thus, we can write:

$$\rho(\psi) = q(p(\psi) - n(\psi) - N_A) \quad (5)$$

The density of mobile carriers can be related to the electrostatic potential (band bending) by Boltzmann’s statistics which applies in the non-degenerate case [10]:

$$p(\psi) = p_0 e^{-\beta \psi}, \quad n(\psi) = n_0 e^{\beta \psi} = p_0 e^{\beta(\psi - 2\phi_F)} \quad (6)$$

where  $\phi_F (= kT/q \log(N_A/n_i))$  is the Fermi potential of the semiconductor and  $p_{c0}$  and  $n_{c0}$  are the hole and electron concentration in the CNT bulk. Combining these three equations, we arrive at the Poisson–Boltzmann equation for the semiconductor’s potential:

$$\frac{\partial^2 \psi}{\partial x^2} = -\frac{q}{\epsilon_c} (p_{c0} e^{-\beta \psi} - n_{c0} e^{\beta \psi} - N_A) \quad (7)$$

From this differential equation, the total charge density per unit area is given by [10]:

$$\begin{aligned} \sigma_s(\psi_s) &= -\text{sgn}(\psi_s) \sqrt{2\epsilon_c N_A kT} \\ &\times \sqrt{e^{-\beta \psi_s} + \beta \psi_s - 1 + e^{-2\beta \phi_F} (e^{\beta \psi_s} - \beta \psi_s - 1)} \end{aligned} \quad (8)$$

To complete the electrolyte-semiconductor system, it is required to satisfy the Kirchhoff’s voltage law and the Gauss law given by

$$V_{GC} = E^M + \frac{1}{q} \mu_e^C - \psi_0 + \chi^{\text{Sol}} - \chi^C + \psi_s \quad (9)$$

$$\epsilon_c \frac{\partial \psi_s}{\partial x} = -\epsilon_m \frac{\partial \psi_0}{\partial x} \quad (10)$$

where  $V_{GC}$  is the potential applied between the reference electrode and the CNT,  $E^M = -\mu_e^M / q + (\phi^M - \phi^{\text{sol}})$ , the reference electrode potential,  $\chi^{\text{Sol}}$  and  $\chi^C$  the surface dipole potential of the electrolyte solution and CNT, respectively and  $\mu_e^C$  is the electron’s chemical potential energy of the CNT.

To determine the  $C_g$  which acts as the gate capacitance of the CNNFET, we need to review the electrical double-layer (EDL) theory as follows.

Fig. 4 shows a charge distribution of CNT–electrolyte system under an applied external bias. This distribution can be modeled using the known Poisson–Boltzmann equation. The diffuse layer is known as the Gouy–Chapman layer. However, it was noticed that the original theory of Gouy–Chapman overestimates the interface charge, and therefore, the capacitance of high-concentration electrolytes. This was remedied by Stern, who realized that ions cannot approach the surface closer than their ionic radii. The distance of the closest approach is called the outer Helmholtz plane (OHP) as

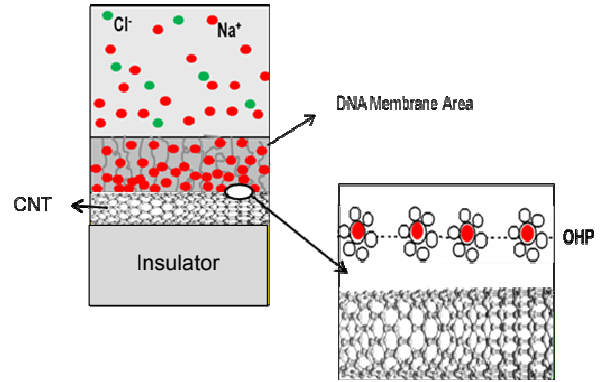


Fig. 4. Charge distribution of CNT–electrolyte system.

shown in Fig. 4.

In the case that the DNA layer is treated as an ion-permeable membrane, the Donnan potential which is solely determined by the DNA concentration and the ion concentration of the electrolyte is formed. It is known that the Donnan potential is expressed as [8, 9]

$$\psi_{DP} = \frac{kT}{zq} \sinh^{-1} \left( \frac{\nu N_m}{2z n_0} \right) \quad (11)$$

Because the total net charge  $\sigma_{md}$  in the membrane and electrolyte is

$$\sigma_{md} = -C_{DL}(\psi_0 - \psi_{DP}) \quad (12)$$

we can deduce the DNA-dependent double layer capacitance from a following equation:

$$\frac{1}{C_{DL}} = \frac{1}{C_{\text{Stern}}} + \frac{\lambda_m}{\epsilon_m \sqrt{\cosh \left( \frac{zq}{kT} \right) \psi_{DP}}} \quad (13)$$

where  $\lambda_m = \sqrt{\frac{\epsilon_m kT}{2n_0 z^2 q^2}}$  is the Debye length in the

membrane. The gate capacitance  $C_g$  is modeled to be the double layer capacitance  $C_{DL}$ .

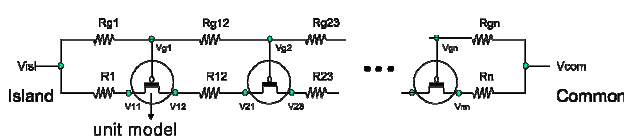
If we disregard the surface charge  $\sigma_f$  under an assumption that the CNT surface charge is small and does not change in time which is usually the case considering the inert surface characteristics of CNT to the electrolyte, we can solve all the equations in a self-consistent manner (using iterations), and the drain current can be calculated.

For the CNNFET with concentric electrodes which consists of the small island electrode and the common

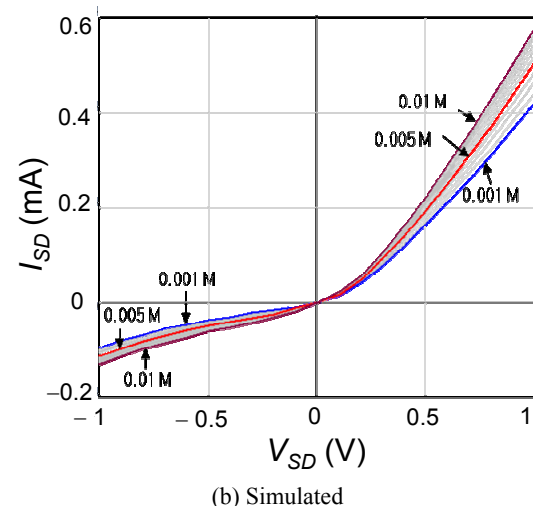
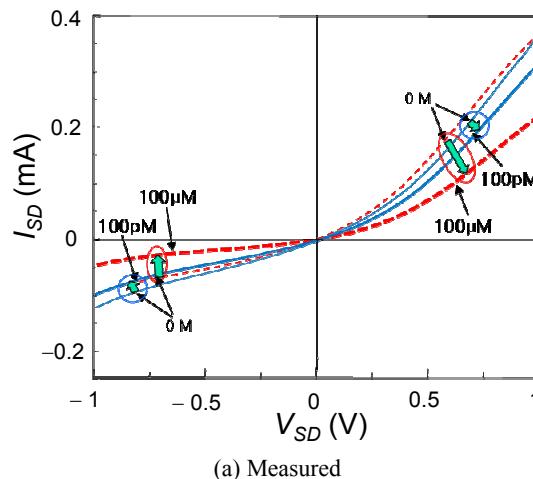
enclosing electrode as shown in Fig. 2(b), there is another issue to be considered due to the asymmetrical nature of the biasing system. When this device is placed in an aqueous solution without the reference electrode for sensor applications, the electric potential of the liquid tends to follow the electric potential of the enclosing electrode due to the smaller redox resistance and larger electrical double layer capacitance between the liquid and the enclosing electrode. This different gate action to the carbon nanotube network channel results in an asymmetrical current profile depending on the voltage polarity.

When an electrolyte resistance is small, the use of the reference electrode using easily polarizable material, e.g. Ag/AgCl, can be used to remove this asymmetry. However, when the electrolyte resistance is high or real-time sensing is performed, the time-dependent current drift which occurs during the electrical double layer charge redistribution is inevitable. Therefore, we need to include this asymmetry in the modeling assuming the measurement is done in the steady-state after long wait time. Fig. 5 shows a schematic of a CNNFET with concentric electrodes which consists of  $n$  serially-connected transistor segments. Because the gate potential of each segment is determined by electrolyte resistances which can be calculated from the concentric disk problem, the previous single CNTFET equation can be applied to these small CNTFETs individually and the current flowing in the whole system can be obtained when the source and drain voltages of each small CNTFETs are determined by another iterations based on electrical circuit laws, e.g. Kirchhoff's laws.

Fig. 6 shows the comparison between actually measured and simulated DNA sensing results. The asymmetrical profile dependence on  $V_{DS}$  appears to be similar. However, several significant differences are found between the measured and simulated DNA sensing results. First of all, the current responses to DNA concentration are opposite. That is, as the concentration



**Fig. 5.** Schematic of a CNNFET with concentric electrodes which consists of  $n$  serially-connected transistor segments.



**Fig. 6.** Comparison between (a) measured, (b) simulated DNA sensing results.

of DNA increases, the measured current decreases as shown in Fig. 6(a), while the simulated current increases as in Fig. 6(b). The reason that the simulated current increases as the DNA concentration does can be explained as follows: a DNA sequence has negative charges because of the phosphate ions in its chemical backbone. In Eq. (3b), the negative charge of DNA is modeled by  $\nu q N_m$  where  $\nu$  is equal to 1 and  $N_m$  is the density of charges in the membrane region and is a negative value for negative charge case.  $N_m$  is proportional to the DNA concentration ( $C_{DNA}$ ) and can be expressed as  $N_m = -K_a \cdot (N_{DNA} \cdot C_{DNA})$  where  $K_a$  is the surface adsorption factor which represents the ratio of surface DNA concentration to its bulk concentration.  $N_{DNA}$  is the number of negative charges contained in the one DNA molecule. In our CNNFET model, as the

magnitude  $|N_m|$  of the negative charge density increases, the magnitude  $|\psi_s|$  of the semiconductor surface potential increases. We modeled the change of CNNFET current using the strong inversion and charge-sheet models.

Secondly, the sensitivity to the DNA concentration is much better in the measured results than in the simulated ones. For example, the LOD of the measured results is less than 100 pM, while the LOD of the simulated ones is about 1 mM. Thirdly, the measured current shows the logarithmic response dependence on the DNA concentration, while the simulated current shows a linear response. These results clearly show that there is the other governing mechanism of CNT current flow which is different from the field-effect mechanism of CNTFET.

### III. ELECTRICAL MODEL OF AU-CNT SCHOTTKY JUNCTION

To explain the Au-decorated CNNFET current phenomena which are different from the field-effect semiconductor theory, there have been a number of papers regarding metal-CNT interface effect, which explain the phenomena by a change of energy barrier height due to the shift of the energy alignment between the metal contact and CNT [11]. According to these reports, we can postulate that the modulation of the Schottky barrier height (SBH) at the metal-CNT contact by the efficient hybridization of DNA on metal electrodes is the dominant sensing mechanism. For the cause of the SBH change from the adjacent molecules in the electrolyte, it has been naturally assumed that this effect arises from the charge doping of the CNT surface [12, 13]. However, in the recent experiments for CNTs, it has been reported that the metal work function change is more plausible to the SBH change than the charge-doping effect is [14, 15].

In the absence of gate voltage, the hole carrier injection through the Schottky barrier formed at the metallic electrode/SWNT interface is dominated, at room temperature, by thermal emission. According to the theoretical descriptions of metal-semiconductor contacts by Schottky and Mott, the SBHs for electrons and hole are

$$\phi_{SB_e} = \phi_m - \chi \quad (14a)$$

$$\phi_{SB_h} = \chi + E_g - \phi_m \quad (14b)$$

where  $\phi_m$  is the work function of the metal which is the energy needed to remove an electron from the Fermi level to the vacuum level and  $\chi$  the electron affinity of the semiconductor which is the energy needed to remove an electron from the bottom of the conduction band to the vacuum level, and  $E_g$  is the band gap.

The current due to thermionic emission in a SB between two bulk materials is given by the ideal diode equation:

$$I = AA'T^2 e^{-\frac{\phi_{SB}}{kT}} e^{\frac{qV}{n kT}} \left(1 - e^{-\frac{qV}{kT}}\right) \quad (15)$$

where  $A$  is the contact area,  $A' = 4\pi m^* q k^2 / h^3$  the effective Richardson constant where  $m^*$  is the effective mass of the carrier,  $V$  the voltage applied to the semiconductor,  $T$  the temperature,  $n$  the ideality factor and  $I_{sat} = AA'T^2 \exp(-\phi_{SB}/kT)$  the saturation current [16]. If  $n > 1$ , the current in the reverse direction increases exponentially with bias while the current for a forward bias is lowered compared to the ideal case. In this paper, however, we would like to focus the discussion only to the ideal case with  $n = 1$  because we are interested in the change of work function itself, not the cause of work function change. If  $n = 1$ , there are no mechanisms that lowers the barrier and the current saturates at  $I_{sat}$  for a reverse bias larger than a few times  $kT/q$  since the reverse thermionic emission current injected into the metal from the semiconductor (first term in the parenthesis in Eq. (15)) vanishes. In the back-to-back Schottky diode model, the width of the depletion region is assumed so large that the tunneling current is negligible compared to the thermionic emission current. Further works may deal with non-ideal cases where the barrier height changes as the bias applies such as the image-force lowering of the SBHs under the reverse bias. If  $n = 1$ , Eq. (15) can be solved to extract the voltage drop existing in the Schottky junction, and the  $I_{SD}$  vs.  $V_{SD}$  characteristics of CNNFET can be described by the following analytical equation:

$$V_{SD} = R_f(V_{GS})I_{SD} + \frac{kT}{q} \log\left(\frac{I_{sat} + I_{SD}}{I_{sat} - I_{SD}}\right) \quad (16)$$

where  $R_f(V_{GS})$  is the resistance of CNNFET which is a function of  $V_{GS}$ . The first term on the RHS of Eq. (16) describes the linear part of the  $I_{SD}$  vs.  $V_{SD}$  curve due to



the CNNFETs, while the second one characterises the nonlinear part attributed to SBH.

**IV. RESULTS AND DISCUSSION**

To address the metal-CNT Schottky junction, back-to-back Schottky barrier diodes are employed. Because the gold particle size is larger than the CNT diameter, the interface between the CNT and the Au particle can be modeled as back-to-back Schottky diodes [17].

The gold nanoparticles are assumed to change the work function when the DNA hybridization occurs, causing the change of the Schottky barrier height (SBH) between the gold and the CNT. We disregarded the Schottky junction between the gold electrodes and the CNTs which are located on the Au electrodes because they are not the bottlenecks of the electrical current. Here it is noted that when the Schottky junction is formed at the interface between the Au electrode and the CNN channel, it can be a bottleneck as well as those between the gold nanoparticles and the CNTs in the CNN channel.

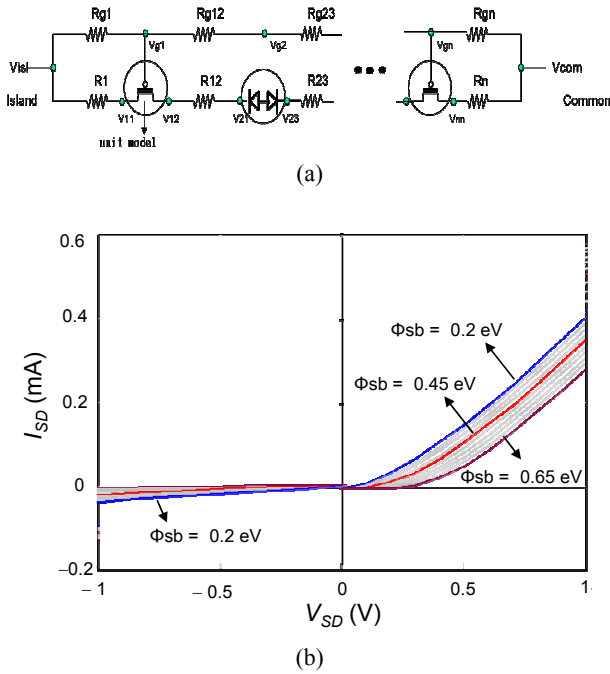
Fig. 7(a) shows a modified schematic of Fig. 5 which

include back-to-back Schottky barrier diodes.

The device parameters used for this simulation are listed in Table 1. The simulated current profile shows that the current varies almost linearly as the Schottky barrier height changes, and the opposite response to the DNA concentration with the field effect transistor (CNTFET) model (Fig. 6(b)), can be resolved. In addition, we can calculate the work function change of DNA hybridization by comparing the experimental data and simulation results. By comparing Figs. 6(a) and 7(b), we can see that 100 pM DNA concentration gives about 0.1 eV work function change and 100 μM, about 0.4 eV work function change. From the measurement data and modeling, the work function shift is about 50 meV/decade of DNA concentration. The work function shifts show the logarithmic dependence of DNA concentration, and are in reasonable range when compared to the measurement data in the literature [18-20]. However, there are differences in Figs. 6(a) and 7(b) at the small voltage region and negative voltage region. In these regions, the current levels of Fig. 7(b) are very small and do not vary linearly according to the applied voltage bias, while Fig. 6(a) shows linear-like curves.

In the reverse voltage region, the field effect model looks better to fit the measured data when there are no DNA molecules. In the back-to-back Schottky model, in the reverse voltage region, it will be better for us to include the reverse current increase due to the Schottky barrier lowering by the image charge effect. But, in this paper, to show the main points and to reduce the complexity of the model, we did not include the reverse current increase effect.

Because a Schottky diode junction area is dependent on the CNT and gold particle densities, we should consider all possible current paths to be added for this parameter. In our simulation, the Schottky diode junction area is chosen to be 1 μm<sup>2</sup> as a fitting parameter.



**Fig. 7.** Electrical model of Au-CNT and the simulation results (a) The schematic of a CNNFET with concentric electrodes where one of the segments is replaced by a back-to-back Schottky diode pair, (b) The simulated CNNFET current profile depending on the Schottky barrier height which comes from the work function modulation.

**Table 1.** Device Parameters

Device Parameters	Simulation Values
Hole mobility of CNTFET	1000 cm <sup>2</sup> /(V sec)
CNTFET channel length	10 μm
CNTFET channel width	10 μm
Schottky diode junction area	1 μm <sup>2</sup>
Work function difference between Au and CNT	0.2 eV

## V. CONCLUSIONS

By comparing the simulation results with the experimental data for the biosensor consisting of the SWNT FET network and the gold nanoparticles decorated on top of the CNT network, we suggest that the main origin of sensitivity is the work function shift of the gold, which changes the Schottky barrier height at the gold-CNT contact depending on the attached biomolecules. We demonstrated that the semiconductor MOSFET effect cannot explain the experimental phenomena such as the superb LOD and the logarithmic dependence of sensitivity to the DNA concentration. By building the asymmetric concentric-electrode model which consists of serially-connected segments of CNTFETs and back-to-back Schottky diodes, we found that the work function shifts are  $\sim 0.1\text{eV}$  for 100 pM DNA concentration and  $\sim 0.4\text{eV}$  for 100  $\mu\text{M}$  DNA concentration.

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