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Nonlinear characteristics of the Fowler–Nordheim plots of carbon nanotube field emission

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Abstract

The barrier between substrate and field emitters can dominate the overall process of field emission. Carbon nanotube (CNT) films were synthesized by thermal chemical vapor deposition (CVD) on silicon substrate covered with a very thin SiO_2 layer as the interface contact barrier. The current versus applied voltage curve shows a high turn-on voltage, and the Fowler–Nordheim (FN) plot exhibits nonlinearity characteristics and departures from the original line and exhibits current saturation in the high-voltage region. However, the FN plot of CNTs grown on Si substrates without SiO_2 layers had no obvious critical voltage and approximately followed the FN law in our experimental voltage region. The reasons for the nonlinearity of FN plots of CNTs grown on a SiO_2 layer were discussed in terms of circuit theory.

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1. Introduction

Electron field emission from carbon nanotubes (CNTs) has been studied extensively since their discovery in 1991 [1–3]. It is believed that CNTs are ideally suited for vacuum microelectronic devices, such as large-area field emission flat panel displays (FED) [4], vacuum microwave tubes [5], x-ray sources [6], etc. CNTs are often supported on substrate, which will result in heterojunctions, Schottky barriers or contact barriers between CNTs and substrates. Therefore, the electrical contact between CNTs and substrates becomes one of the most important issues in nanoelectronics, and this topic has been studied theoretically [7, 8] and experimentally [9–11]. In general, the reported electrical contact resistances between CNTs and substrates are very large [12–15]. The electron field emission current of CNTs deposited on substrates will be greatly tailored by the contact effect [16]. Such high resistance severely prevents CNT-based devices from reaching the intrinsic electronic properties of CNTs. The stability, reliability and repeatability contact between CNTs and substrates is an important factor for the CNT field emission property. So it is important to understand the role of the interface contact on the CNT field emission property. In this paper, CNTs were deposited on Si substrate covered with a thin SiO_2 layer that acts as a certain interface resistance, and the field emission property was investigated. The reason for nonlinearity of the FN plot was discussed in terms of circuit theory [17].

2. Experiment details

The substrates used in this study are pristine *n*-type Si (001) wafer. Firstly, the wafer was cut into two pieces of 5 mm × 5 mm size. One of them was treated for 5 min in 1% HF solution to remove native silicon dioxide. Another one was covered with a SiO_2 dielectric layer as the interface

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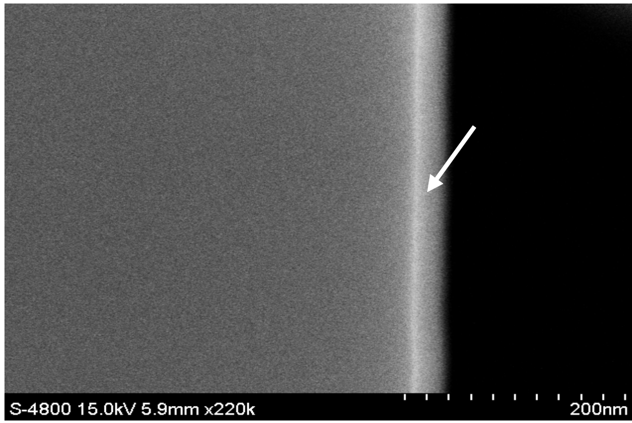

Figure 1. Thickness of the SiO_2-covered Si substrate in SEM. The white arrow indicates SiO_2 layers.

barrier using wet thermal oxidation at 1100 °C for 3 min. The thickness of the SiO_2 layer was checked by scanning electron microscopy (SEM) and was determined to be about 30 nm, as shown in figure 1. Then, substrates of pure Si and Si covered with a SiO_2 layer were supersonically cleaned in acetone, alcohol and deionized water baths and subsequently dried. Thirdly, Fe catalyst particles were deposited on these substrates by ion beam sputtering at room temperature in the same conditions. After deposition of the catalysts, the substrates were annealed at 850 °C in a hydrogen atmosphere for 1.5 h in a quartz-tube furnace to promote the formation of catalyst particles. Lastly, CNTs were grown at 1000 °C in a gas mixture of methane (400 SCCM) and hydrogen (30 SCCM) for 15 min.

The growth of CNTs by the CVD method depends on the substrates under the same experimental conditions. In this experiment, many repeated experiments were carried out and a large number of samples were obtained. Samples of CNTs with similar morphology and density were chosen. Hence the screening effect on CNT field emission should be the same in the range of experimental error and could be neglected in our contrasted experiments. The surface topography of CNT films was illustrated by SEM, as shown in figures 2(a) and (b). The CNTs on the two substrates are randomly oriented and entangled with each other. There was no obvious difference in the morphologies of CNTs grown on Si substrate covered with a SiO_2 layer and on the Si substrate without oxidation. The differences of emission behavior should be primarily attributed to the interfaces effect.

In order to study the field emission properties of CNTs, they were placed in a vacuum chamber with the base pressure lower than 9×10^{-5} Pa. The samples were used as the cathode and the indium tin oxide (ITO) glass was used as the anode. The current–voltage (I–V) curves were taken by a Keithley 237 source measure unit to carry out field emission measurements. Figure 3(a) shows the field emission I–V curves of CNT film grown on Si substrate covered with a SiO_2 layer and Si substrate without thermal oxidation. The Flower–Nordheim (FN) plots of $\mathrm{Ln}(I/V^2)$ versus $1/V$ are shown in figure 3(b). We repeated the field emission test of each sample three times for CNTs grown on bare Si substrate and for CNTs grown on SiO_2-covered Si substrate. Although some points of the I–V curves or the FN plots had larger departures, the FN plot shapes were almost identical in the three cases. So we chose the fine I–V curve and the fine FN plot from them. Therefore, we could find that figure 3(a) and (b) are not consistent.

3. Result and discussion

As shown in figure 3, the I–V curve of CNTs grown on Si substrate with an interface SiO_2 layer showed a high turn-on electric field. FN plots of CNTs grown on SiO_2-covered Si substrates show nonlinearity characteristics. But the FN plots of CNTs grown on pure Si substrates without SiO_2 layers had no obvious critical field and approximately followed the FN law in our experimental voltage region.

For the CNT field emission process, firstly, electrons should transport from the substrate to the CNT and overcome the contact resistance between the substrate and the CNT (figure 4). Secondly, electrons pass through the CNT, and finally emit into the vacuum. Therefore, contact resistance can play an important role in the field emission process. If we disregard the intrinsic resistance of the CNT, then taking the resistance of the back contact as R_B, the elementary FN equation can be modified by using

$$I = A\beta^2(V - IR_B)^2\phi^{-1}\exp[-B\phi^{3/2}/\beta(V - IR_B)] \equiv (V - IR_B)/R_F, \quad (1)$$

where I is the current [19], A and B are constants, ϕ is the work function of the emitter material, V is the total voltage applied between the anode and the conducting Si substrate, and β is the conversion factor that relates the field in the tunneling barrier to the voltage difference between the anode and a point just inside the end of the emitting CNT. The parameter R_F, called the 'front resistance', is defined by equation (1). As recently pointed out by Forbes [20], the elementary FN equation is not a quantitatively accurate emission equation, particularly for CNTs. However, it does get trends in emission behavior qualitatively correct, and this is sufficient for the arguments here.

In this paper, we consider that changes in the relative values of R_B and R_F are the reason for the nonlinear characteristics of the FN plots of CNT field emission. Both resistances R_B and R_F are functions of current, and we have the implicit equation

$$I = V/[R_F(I) + R_B(I)]. \quad (2)$$

The current flow can be simply illustrated by an equivalent circuit as shown in figure 5. $R_F(I)$ varies significantly with current, getting smaller as I increases. In our experiments, we assume that the variability of the effective resistance R_B for the SiO_2 layer is less than the variability in R_F.

For the CNT sample grown on Si substrate covered with a SiO_2 layer, we assume that the resistance of the back contact (including the SiO_2 layer) is 'nearly constant'. As the applied voltage V (and hence the current) is increased from a low value, R_F decreases. For the regime where $R_F \gg R_B$, the current is dominated by the CNT-emitting surface taken in isolation, and the emission follows the FN law (which determines the slope of the FN plot).

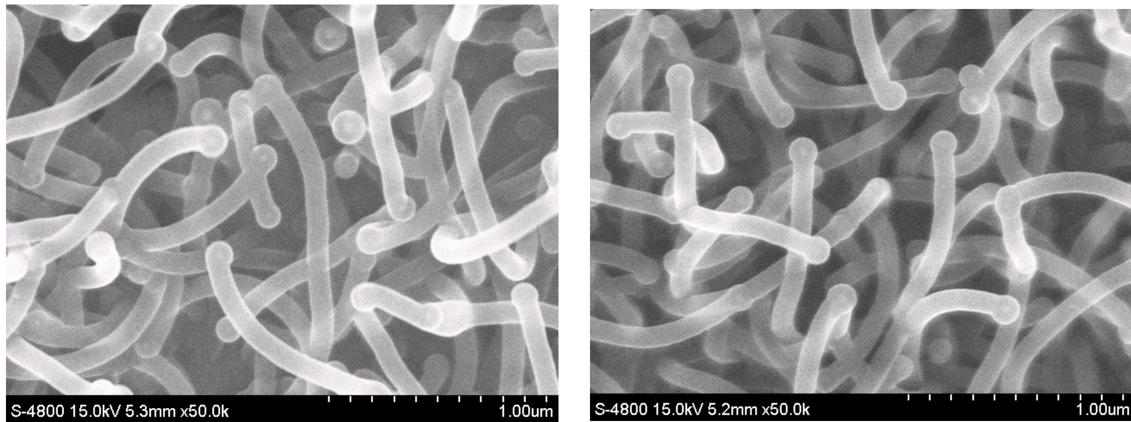

Figure 2. Typical surface topography of CNT films on Si substrate covered with a SiO_2 layer and without thermal oxidation in SEM.

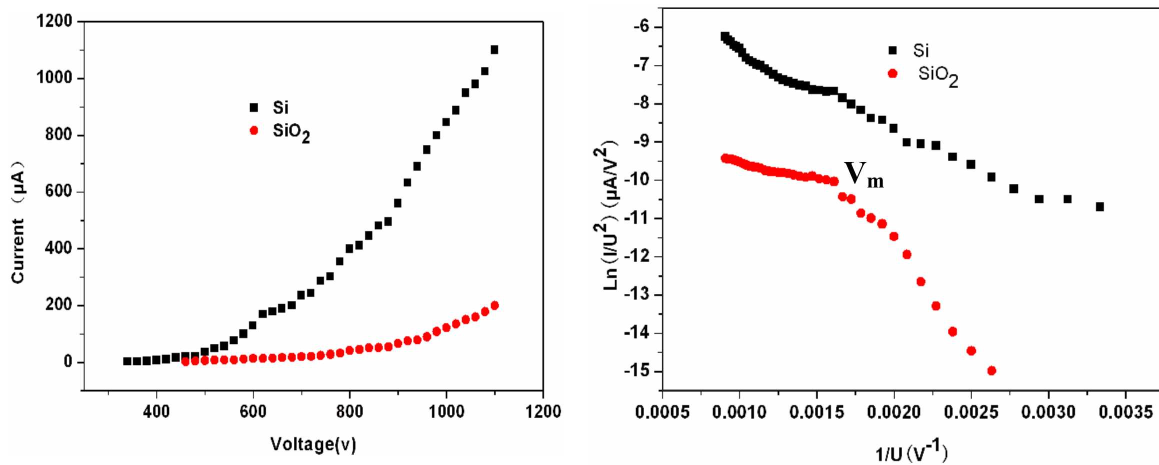

Figure 3. *I–V* and FN plots of CNTs grown on Si substrates covered with SiO_2 and without thermal oxidation.

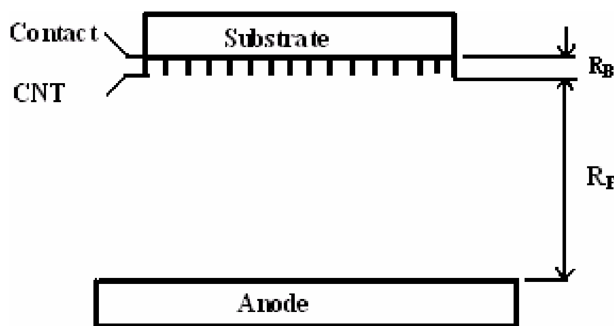

Figure 4. Schematic diagram of resistance distribution between substrate and anode.

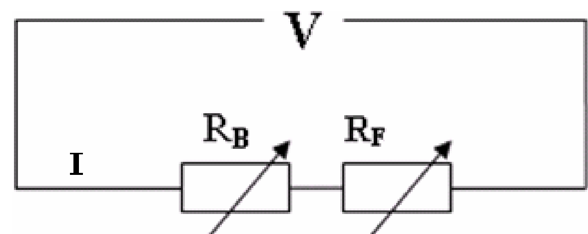

Figure 5. Equivalent circuit diagram for the field emission of CNTs grown on SiO_2/Si substrate.

When (with increasing applied voltage) R_F becomes smaller than R_B, the emission current is restricted by the electron conduction through the series combination of the contact resistance and the intrinsic resistance of the CNT emitters (but we assume here that the CNT intrinsic resistance can be neglected). In this regime, the differential characteristics of the *I–V* behavior are related to the *I–V* characteristics of the back contact taken in isolation. Therefore, as current increases, and the emission moves into this regime (where $R_B \gg R_F$), the FN plot deviates from its original straight line and exhibits nonlinearity, as shown in figure 3(b). In this process of regime change, the voltage V_M, defined as the voltage difference across the back contact, moves (as *I* increases) from being 'close to zero' to a higher value determined by the *I–V* characteristics of the back contact taken in isolation.

For the sample of CNTs grown on Si substrate, as shown in figure 3(b), the FN plot has no distinct break of slope in our experimental voltage region. We assume that this is because R_B is always relatively small, so R_F is greater than R_B for all currents of interest. In this case, in the whole field emission range, the emission current is dominated by the CNT front surface properties taken in isolation; thus, field emission follows the FN law and the FN plot exhibits good linearity. Because R_B is small, and there is little voltage drop across

the back contact, the interface between the substrate and the CNT can supply enough electrons for CNT electron field emission [21].

Nonlinearity of the FN plots in field emission has been studied by many researchers in recent years [22–25]. Two slope of the FN plot were found when GaN nanorod films [26] were deposited on Si substrates with a native silicon oxide layer because of the existence of parasitic resistance in the $GaN/SiO_2/Si$ sample. CNTs were grown on different substrates (SiO_2, Al, Cu, Ti), and it was found that there were explicit knees in the FN plots [27, 28]. Our paper describes the effect of interface contact resistance on field emission, and these findings could provide a deeper insight into further understanding the field emission of CNTs.

4. Conclusion

The field emission property of CNTs deposited on silicon substrates covered with SiO_2 as an interface layer has been qualitatively discussed in terms of circuit theory. The FN plot of CNT grown on silicon substrate covered with SiO_2 exhibits nonlinearity characteristics in the high electric field. The two variable resistances of the interface layer (R_B) and the surface (R_F) are the reason for the nonlinearity of the FN plots. When $R_B < R_F$, emission current is dominated by the CNT surface taken in isolation and field emission follows the FN law. When $R_B > R_F$, the emission current is restricted by the electron conduction in contact resistance and the intrinsic resistance of CNT emitters, and the FN plot deviates from the original line and exhibits nonlinearity. It was proved that the limitation process of the current flow under high electric field was connected with difference resistances of contact and surface. The above discussion seems to be helping us in further understanding the reason for the nonlinearity of FN plots in a high electric field.

Acknowledgments

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