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Planar Field Emission Current from Individual Carbon Nanotubes

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Carbon nanotube (CNT) planar field emitters were fabricated on a SiO_2/Si substrate. The anode, cathode and CNT all lay on the same substrate for the promising advantage of intergratibility with planar technology. The emission current was acquired in a scanning electron microscope (SEM). Despite the asymmetry (tip-electrode) of our field emission sample, a symmetrical *I–V* curve consistent with the Fowler-Nordheim theory was acquired. Using Zener theory on quantum tunneling in insulators, the observed phenomenon was explained to be a possible leakage current through the insulating SiO_2 instead of real field emission. Moreover, the simulated local electric fields at the emitter apex exclude the possibility of an accountable emission current. Our results are of great importance in studying planar field emission since it draws attention to avoid mistaking leakage current for the actual field emission current in planar field emission devices.

Keywords: Planar Field Emission, Carbon Nanotube, Leakage Current, Zener Tunneling, Finite Element Method.

1. INTRODUCTION

The field induced emission of electrons from cold cathodes is a well understood phenomenon and is, at present, still an active research area for many one-dimensional nanowires and nanotubes.[1–8] Two important parameters determining the performance of a field emitter: the radius of curvature of the emitter tip, and the aspect ratio of the emitter: generally, the sharper the emitter, the better the field emission properties. Carbon nanotubes (CNTs) are obviously the most prospective candidates as field emitters with the very small radius curvature of the tip on nanoscale and very large aspect ratio. Acknowledging this, CNT field emitters have drawn much attention lately, and have been demonstrated to have outstanding field emission properties.[9–15] They are shown capable of delivering 1 μA per single CNT and high current density in excess of 1 A/cm^2.

Most of the pioneer works on the CNT field electron emitters were designed within the framework of the traditional vacuum tubes. The three-dimensional tip-to-electrode setup was adopted; the anode and the cathode are separated by a high vacuum empty space (Fig. 1(a)). Planar field emission in general, is the field emission achieved on and/or across a substrate; it is characterized by the fact that the cathode, the anode, and the emitters lay on an insulating substrate.[16] So the electron emission is reduced from three-dimensional to two-dimensional. Figures 1(b) and (c) illustrates these planar devices: (b) is the so called tip–tip (or bilateral) configuration and (c) is the tip-electrode (or unilateral) configuration. The benefits of such a design include usage of thinner CNT emitters, integratability with planar technology, stable construction, and etc. The concept is ideal for the incorporation of emitter devices into integrated circuits. However, it is only until recently—with the development of nanotechnology and the advent of sophisticated instruments and fabrication techniques—that devices based on the idea were realized.[17]

Many recent reports on planar field emission have chosen to assemble the emitter and electrodes on doped Si wafers with a SiO_2 insulating layer (typically a few hundred nanometers in thickness).[16, 18–22] Employing such constructions, a gate voltage can be applied, which can increase the functional options of the field emitter device. The inherent problem with such a construction is that leakage current through the insulator is very easily neglected. In this report, a significant leakage was found in the insulator layer in our planar field emission device based on individual CNT. Furthermore, the *I–V* relation closely resembles that of field emission with similar Fowler-Nordheim (FN) plots. Thus, one can easily mistake the obtained current for actual field emission phenomenon. Possible reasons were carefully analyzed for the observed results. And suggestions were given in order to confirm the experimental data to be real field emission.

2. EXPERIMENTAL DETAILS

The CNTs in this investigation were grown via a chemical vapor deposition (CVD) method[23] on a pre-marked, highly doped,

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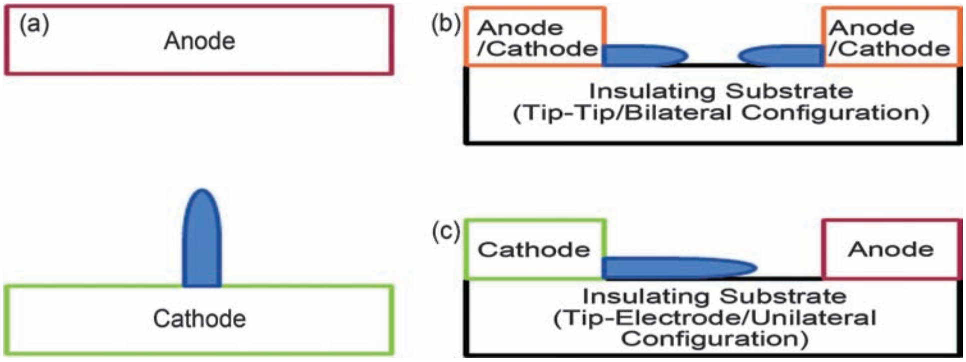

Fig. 1. An illustration of a traditional vacuum field emission setup characterized by two topologically separated electrodes. (b) and (c) are two possible configurations of a planar field emission device: (b) with two opposing emitter is the tip–tip (or bilateral) configuration. And (c) with only one emitter is the tip-electrode (or unilateral) configuration; the planar devices are characterized by the fact that the electrodes share a common substrate.

Si substrate covered with a 500 nm SiO_2 insulating layer. This method produces roughly straight CNTs lying along the same direction, convenient for producing planar field emitters. Figure 2(a) is the SEM micrograph of the as-grown CNT on the premarked substrate.

For the measurement of field emission current, Ti (20 nm)/Au (30 nm) electrodes were deposited on the individual as-grown CNTs using standard electron beam lithography (EBL) and lift-off process. Similar to the work previously reported,[16] sharply tipped glass needles were employed in cutting the CNT under an optical microscope (Fig. 2(b)). Our procedures produced a tip-electrode planar CNT emitter like those illustrated in Figure 1(c); the surface morphology of the prepared CNT emitter was characterized with an atomic force microscope (SEIKO SPI3800N) under contact mode (Fig. 2(b)). The CNT emitter apparently folded back upon itself at the tip; the surface analysis of an uncoupled part (blue arrow) and the apex (red arrow) of the emitter reveals a diameter of ~3.6 nm (blue box) and ~3.9 nm (red box) respectively. Hence, considering uncertainties and the coupling, the apparent apex diameter is estimated to be ~9 nm. The length of the emitter is approximately 2.6 μm; therefore, giving an aspect ratio of about 289.

I–V measurements of the planar field emission current on the individual CNT were conducted in a SEM chamber under a vacuum of 2×10^{-6} Torr. A Keithley-6430 Sub-Femtoamp Remote SourceMeter together with its pre-amplifier was used to apply

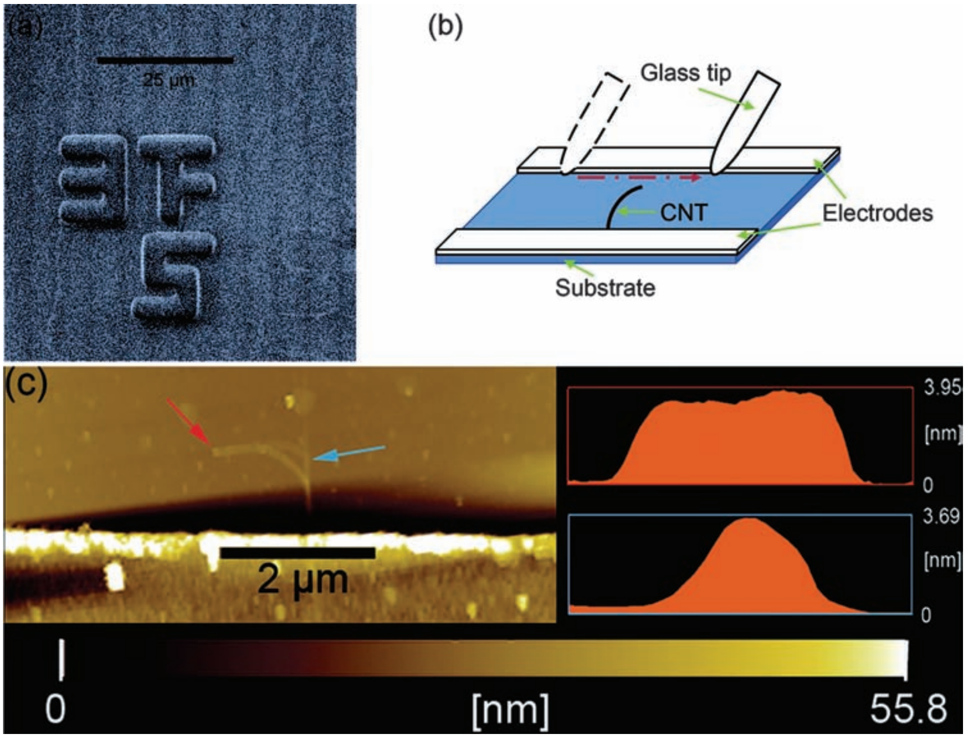

Fig. 2. The SEM micrograph of the as-grown CNTs, laying alone the same direction on the SiO_2/Si substrate. (b) An illustrated demonstration showing CNT emitters readily fabricated with the use of glass needle tips under an optical microscope. (c) AFM micrograph of our prepared CNT emitter: the color boxes are surface analysis, they correspond to the site of the color arrows on the image. The CNT diameter is ~3.6 nm (blue box), however, due to a folding back of the CNT at the tip, the apparent diameter is over twice the actual CNT diameter (red box).

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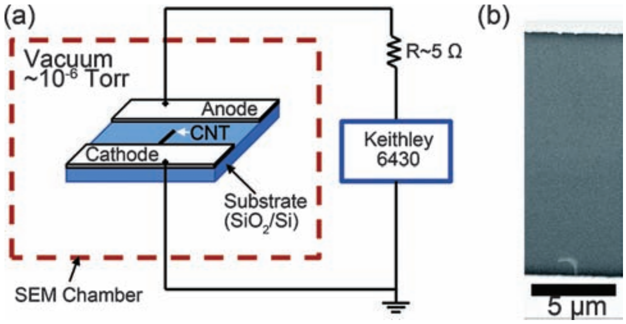

Fig. 3. An illustration of the measurment setup in a SEM with a vacuum environment of 10^{-6} Torr; a keithley-6430 Sub-Femtoamp Remote SourceMeter was used to source the voltage and measure the current. (b) The SEM micrograph of the tested planar CNT emitter before current acquisition.

the voltage and measure the emission current; the voltage was ramped in steps of 1 V. The peripheral circuit resistance of our setup was less than 5 Ohm. The electron beam was initially used to determine the tip-electrode distance ($\sim$14 μm) then subsequently turned off for the emission current acquisition. Figure 3(a) is a schematic illustration of the current measurement setup, and the SEM micrograph of the tested planar CNT emitter is shown in Figure 3(b).

3. RESULTS AND DISCUSSION

Figure 4(a) shows the measured *I–V* curve: the forward curve is the obtained current when a negative bias is applied on the emitter part and the reverse curve the vice versa. The onset voltage at 1 nA was around 10 V. Comparing the results to the 115 V onset voltage of a similarly thin traditional single CNT emitter, with a tip-electrode distance of a mere 1 μm, reported by Bonard et al.,[11] the apparent improvement of field emission properties is quite significant. The onset voltage of our planar CNT emitters is much smaller than most of the reported CNT emitters in traditional setups.[11, 24–26] The obtained *I–V* implies a bilateral emission in nature; it bears resemblance to that previously obtained from a tip–tip configuration in planar field emission devices by Song[16] and Wang.[27] Moreover, complying with the Fowler-Nordheim theory,[28, 29] the FN plot (inset of Fig. 4(a)) of the curve does reveal a linear trend at emission field strengths. So if it is real field emission, the improvement is significant. However, considering the fact that an asymmetric tip-electrode configuration was deliberately prepared (Fig. 1(c)), the result seems absurd, since it should give current only when negative bias was applied on the CNT, and no emission current when positive voltage was applied. The symmetric shape of the curve makes us assume a leakage current might exist between the electrodes through the insulating SiO_2 layer, for leakage current is independent of the direction of bias voltage. Nevertheless, this alone cannot account for the consistency of the *I–V* with field emission theory.

The FN field emission theory is basically the rationalization of the quantum tunneling phenomenon.[28] The theory has been proved useful in describing the relationship between the field emission current I and the local field F at the emitter surface.

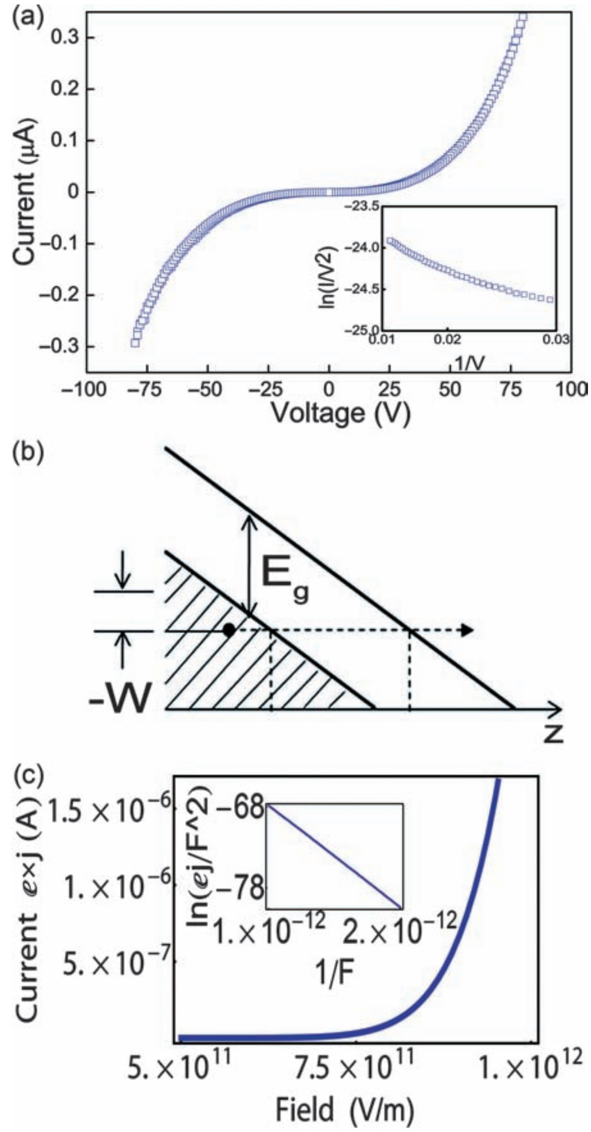

Fig. 4. *I–V* curve acquired from the planar CNT. The forward curve is in the sense that a negative bias is applied on the emitter. The inset shows the corresponding FN plot of the emission data; the linearity complies with the FN theory. (b) is an illustration of an electron with energy $-W$ tunneling through the field tilted bands; E_g is the band gap (here, analogous to the work function ϕ) (c) is the current (*ej*), calculated from Zener's expression, tunneling into the conduction band versus the applied field; the inset shows the corresponding FN plot.

F is conventionally written as: $F = \gamma V/d$: V is the applied voltage, γ is the geometric enhancement factor, and d is the tip-electrode distance. The law relating I and F is thus written as[29]

$$I = A\frac{1.5\times10^{-6}}{\phi}\left(\frac{V}{d}\right)\gamma^2 \exp\left(\frac{10.4}{\sqrt{\phi}}\right)\times\exp\left(-\frac{6.44\times10^9\phi^{1.5d}}{\gamma V}\right) \tag{1}$$

where A has the dimension of an area m^2 and ϕ is the work function in eV of the emitting material. From Eq. (1) it can be observed that if $\ln(I/V^2)$ is plotted against I/V, then, at emission field range, one will arrive at a linear function with a slope $-6.44\times10^2\phi^{1.5}d/\gamma$; this is the so called FN plot. By fitting the experimental data in a FN plot, either ϕ or the field enhancement factor γ can be determined. Usually it is the enhancement factor that is calculated for, since it is a measure of emitter performance. The FN plot has also been used over the past as a supporting evidence for field emission. Despite this, the following paragraph will prove it to be only a necessary condition for field emission.

Returning to the question at hand, with the FN law in mind, our observed current should also be expressible in a relation similar to Eq. (1) to show such analogous behavior as field emission. In fact, the phenomenon is known as internal field emission of insulators; it was first treated by Zener in 1935.[30] The idea is basically shown in Figure 4(b), an energy level versus position illustration for insulators or semiconductors with a band gap E_g between the valence and conduction band. Upon application of an electric field, the band gap edges become tilted in space. An electron with energy $-W$ with respect to the valance band edge can make transitions to the conduction band not only vertically (requiring an energy $> E_g + W$), but also horizontally, owing to the applied field. Namely, the valence electrons can tunnel into a current carrying band state. The expression derived limiting the rate of transition is, as expected, an exponential. It is written as

$$j = \frac{eFa}{h}\exp\left(-\frac{\pi^2}{h^2}\frac{ma\epsilon^2}{|eF|}\right) \tag{2}$$

where a is the spatial periodicity of electron potential energy, m is the electron mass, E_g is the energy band gap, and F is the applied local field. Intuitively, the electric field F here can also be related to the applied voltage through an analogous enhancement factor. Figure 4(c) is a plot of current I (electronic charge $e \times$ Eq. (2)) versus electric field considering reasonable values for the parameters in our experimental setup: $a = 50 \times 10^{-9}$m (presuming thinner amorphous SiO_2 at leakage sites) and $E_g = 9.1$ eV for SiO_2; the calculation is a periodic boundary estimation using the thickness of the amorphous SiO_2 as a large unit cell. The curve thus obtained share similarities with that of vacuum field emission. To show this curve will derive a similar linearity in a FN plot, Eq. (2) is rewritten as

$$\ln\left(\frac{ej}{F^2}\right) = \ln\left(\frac{e^2a}{Fh}\right) - \frac{\pi^2}{h^2}\frac{ma\epsilon^2}{|eF|} \tag{3}$$

Contrary to FN law, the leading part of the right hand side of Eq. (3) ($\ln(e^2a/Fh)$) is here also a function of local field; one would expect to see a non-linear relation. Nonetheless, the plot of Eq. (3) (Fig. 4(c) inset) does still reveal a linear relation. The reason for this is that the variation of $\ln(e^2a/Fh)$ is small within the field range of observable Zener tunneling. Within this field range, $\ln(e^2a/Fh)$ varied $\Delta_1 \sim 0.7$ whereas the second term ($\pi^2ma\epsilon^2/h^2|eF|$) varied $\Delta_2 \sim 13.3$; the former is only about 5.2% of the latter. So when we plot $\ln(ej/F^2)$ against $-1/F$ as shown in inset of Figure 4(c), a linear relationship similar as FN plot was obtained. This is the main reason for mistaking the obtained linear "FN plot" for real vacuum field emission tunneling. In fact, it is the leakage current through SiO_2 insulating layer explained in the framework of Zener Theory. This result supports our initial assumptions and speculations.

To complete our analysis, the electric field at the CNT tip is calculated using Comsol Multiphysics, a commercial program based on finite element method, is employed. For a quantitative comparison, a traditional configuration was considered along with the configuration in this study. The dimension in the traditional case closely follows those reported by Bonard et al.[11] on

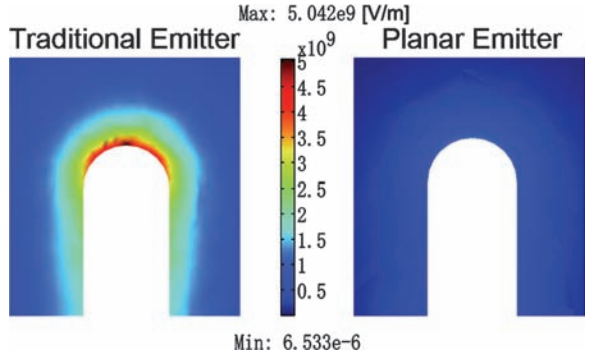

Fig. 5. The slice plot of calculated local fields for both configurations. As it is shown, for the traditional configuration the tip field is around 5 times larger than the planar setup.

a single standing CNT: 1.45 μm in length, a 7.5 nm radius, and an approximately 1 μm tip-electrode distance. Meanwhile, the model bent CNT for planar configuration in our case, lying on a 500 nm thick SiO_2, is 5 nm in radius and roughly 2 μm long with a tip-electrode distance of about 14 μm. A hemispherical cap was drawn to model the tip head for both cases. The voltage applied on the emitter is chosen as 115 V so as to model the onset voltage (the voltage required to extract a current of 1nA) reported by Bonard et al.[11] The calculated field at the apex of the free standing model and planar model was $\sim 5 \times 10^9$ and $\sim 1 \times 10^9$ V/m respectively (Fig. 5). The electric field at the apex of the CNT in planar configuration is only fifth of that in free standing configuration. In other words, the onset field for vacuum field emission in our case was not achieved, so the obtained *I–V* curve could not be vacuum field emission, in accordance with our previous explanation.

Although we have proved the current to be of leakage through the insulating layer, the origin of such a large leakage is difficult to identify. Repeated experiments with or without CNT do not always reveal significant leakage current within the voltage source range (200 V): there are cases where a relative flat curve (no current) is obtained and sometimes the phenomenon is triggered by a breakdown first. The evidences suggest that leakage may be due to defects in the insulating layer, local sites where the SiO_2 is thinner. The defects may be originally present from fabrication process or introduced by material breakdown from local static charge accumulation. It is also reasonable to assume breakdown from the applied field through a field enhancement mechanism analogous to the vacuum field emission situation.

Our results and analysis suggest, where the FN theory is applicable, the linearity of FN plot is only a necessary condition (at least in planar emission setups) for real field emission; other tests must be performed in order to confirm the results. For unilateral assemblies, a reproducible asymmetric *I–V* curve, from negative to positive bias should be sufficient. Meanwhile, for symmetric bilateral assemblies, leakage between electrode and Si wafer must be re-examined after observing a supposed vacuum field emission.

4. CONCLUSIONS

Carefully designed CNT planar system for field emission was fabricated. Utilizing SEM to provide the vacuum and precise measurements, a planar field emission current from the planar

CNT was observed. The obtained I–V was symmetric for forward and reverse bias and the FN plot complied with the FN theory. But our tip to electrode experimental setup should give asymmetric I–V curve instead of a symmetric one. Finite element calculations of our planar device revealed the fact that the onset field strength for vacuum field emission was not achieved. Thus the current cannot be originated from the CNT. Meticulous analysis based on Zener theory reveals the current to be originated from leakage through the insulating SiO_2 layer instead of real vacuum field emission phenomenon. The large leakage was attributed to possible defects in the SiO_2 layer.

One of the aims in studying planar field emission is its prospects in integrated circuits. Our work is of great importance in studying field emission from planar field emission devices since it is easy to mistakenly identify the obtained current to be originated from vacuum field emission instead of other possible cases such as leakage from insulating layer.

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