Giant field enhancement at carbon nanotube tips induced by multistage effect

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Recently, we have reported an extremely strong field emission from carbon nanotubes grown on carbon cloth, with the field-enhancement factor of up to 18 800. In this paper, we study the origins of this effect, by investigating field emission from individual carbon nanotubes, in a transmission electron microscope equipped with a piezo manipulator. Microscopic analysis reveals a multistage structure of some of the nanotubes, characterized by an order of magnitude smaller nanotubes branching off the tips of bigger nanotubes (or carbon fibers). The multistage structure causes a macroscopic enhancement of the electric field, which can match that of a single macroscopically long nanotube with length equal to the combined length of all stages, and the tip radius equal to that of the thinnest nanotube in the structure. This not only explains the observed giant field enhancement, but also provides important clues for the design of nanotube emitters for electronic applications. © 2005 American Institute of Physics. [DOI: 10.1063/1.2008363]

One of the most promising applications employs carbon nanotubes as electron emitters in the field-emission devices, such as flat panel displays, x-ray tubes, and microwave power amplifiers.1–6 Recent experiments have shown that carbon nanotubes can function as excellent field emitters with high current density at low turn-on voltage.6,7 However, further improvements are necessary to decisively outperform the existing technologies. Most experiments were conducted with large area emitters, in which a variety of nanotubes participates in the emission. This randomness obscures the basic mechanisms of the field emission, understanding of which is crucial for applications.

We have recently reported a remarkable field-emission property from carbon nanotubes grown on carbon cloth,7,8 with the turn-on field of 0.17 V/μm, and the threshold field of 0.4 V/μm, to reach 1 mA/cm2. These are the lowest values achieved so far. The extremely large field-enhancement factor of about 1.88×104, extracted from the Fowler-Nordheim (FN) plot for the large area measurements,7 is responsible for low turn-on and threshold fields. However, mechanisms that led to this giant field enhancement remained unclear. The main goal of this letter is to explain this remarkable field enhancement.

We have performed a comprehensive study of the field emission from individual nanotubes, which were grown on a commercially available carbon cloth by thermal chemical vapor deposition (CVD) technique.2 Carbon cloth is two-dimensionally knitted carbon fibers with individual fiber diameters of about 12 μm.2 Transmission electron microscopy (TEM) inspection of the nanotubes shows that many of them have a multistage structure, as shown in Fig. 1. This structure is characterized by an order of magnitude smaller features (nanotubes or fibers), growing at the tips of larger features. The structure shown in Fig. 1 consists of three stages: a carbon fiber [Fig. 1(a)], a large nanotube [Fig. 1(b)], and a small nanotube [Fig. 1(c)]. It is this multistage structure that leads to the giant field enhancement. Our study was performed inside a JEOL 2010F TEM, equipped with a piezo manipulator. The current-voltage (I-V) curves were taken for a nanotube shown in Fig. 2(c), mounted on a gold rod shown in Figs. 2(a) and 2(b).

Our nanotubes obey the FN law, given by9,10

$$I = A \frac{1.5 \times 10^{-6}}{\phi} E^2 \exp \left( \frac{10.4}{\phi} \right) \exp \left( - \frac{6.44 \times 10^9 \phi^{1.5}}{E} \right),$$

(1)

where $I$ (in amperes) is the field emission current, $E$ (in V/m) is the local electric field at the tip, and $\phi$ (in eV) is the electron work function for the nanotube. Since $E$ is proportional to $V$, we can write

$$E = \frac{V}{d},$$

(2)

which defines $\gamma$. Here $d$ is the size of the gap between the anode and the tip of the nanotube. $\gamma$ can be determined from Eq. (1), provided that $\phi$ and $d$ are known.

So defined, $\gamma$ describes the field enhancement only due to the tip shape, and not due to the tip’s proximity to the anode. Figure 3 shows $\gamma$ vs $d$ plots for two nanotubes. Figure 3(a) shows data from two nanotubes (A and B) with different radii of 10 and 22 nm, respectively, in the small gap region, $d < 5 \mu$m. In this regime, $\gamma$ decreases linearly with $d$. Linear

![Fig. 1](image-url) FIG. 1. Multistage growth geometry for nanotubes grown on a carbon fiber. (a), (b), and (c) are subsequent magnifications of the stages.
shown in Fig. 3. For mental data in the entire range of field. This formula is in excellent agreement with the experimental results shown in Fig. 4(c). For \( l_2 \ll l_1 \) and \( l_2 \ll r_1 \), we get, \( \gamma = (l_2/r_2)(l_1/r_1) \) in agreement with Eq. (6). However, for \( l_2 \gg r_1 \), we get \( \gamma_m = (l_1 + l_2)/r_2 \), which is identical with the enhancement produced by a single nanotube with the combined length of the two stages, and the radius of the smaller nanotube. The same holds for a multistage system with an arbitrary relation between \( l_{i+1} \) and \( r_i \), in which \( l_{i+1} \ll l_i, r_i \ll l_i \). In particular, Eq. (6) is still approximately valid, but with

\[
\gamma_i = l_i/r_i = l_i/(l_i + l_{i+1})
\]

As in the two-stage system, for \( l_{i+1} \ll r_i \), Eq. (7) yields \( \gamma_i = l_i/r_i \), i.e., we recover from Eq. (6) the original aspect ratio product formula. For \( l_{i+1} \gg r_i \) we get from Eq. (7), \( \gamma_i = l_i/l_{i+1} \), and from Eq. (6), \( \gamma_m = l_i/l_{m} = l_i/or \), where \( L \) is the total length of all stages. This field enhancement approaches that of a single nanotube with the length \( L \), and the tip radius \( r_m \) (radius of the smallest nanotube). The result highlights that a multistage system can perform as well as a macroscopically long single nanotube of the same length. Obviously, such a macroscopically long nanotube of a very small radius is much more difficult to fabricate than a multistage emitter with a very fine, final stage nanotube.

Importance of the multistage enhancement is illustrated in Fig. 3(b). The aspect ratio of nanotube A is \( l/r = 300 \), but Fig. 3(b) clearly shows that the \( \gamma \) for large \( d \), \( d > 4 \), exceeds 4000. The reason for that is the multistage enhancement. This nanotube is attached to a conical rod [Figs. 2(a) and 2(b)], which enhances the field also. According to Eq. (6)

\[
\gamma_{tot} = \gamma_{rad} \gamma_{rod} = \frac{l}{l + d} \left( 1 + \frac{d}{r} \right) \frac{L'}{L' + d} \left( 1 + \frac{d}{R'} \right).
\]

\( L' \) and \( R' \) are the corrected effective length and radius of the conical rod. Choosing \( L' = 85 \mu m, R' = R + l = 3.5 \mu m \), we obtain an excellent fit to the experimental data for nanotube

The proportionality of \( E \) to \( E_0 \) [Eq. (3)] implies that for a multistage structure, with each stage much smaller than the previous one \( (l_{i+1} \ll l_i, r_i \ll l_i, \text{and} \ l_{i+1} \ll r_i) \), the total enhancement factor at the tip of the smallest stage, \( \gamma_{tot} \), is a product of the enhancement factors for individual stages. Thus,

\[
\gamma_{tot} = \prod_{i=1}^{m} \gamma_i,
\]

where \( i \) is the stage index, \( m \) is the total number of stages, and \( \gamma_i = l_i/r_i \).

The electrostatics,11 with the assumption of \( d \gg R \) and \( l \gg r \), yields the following simple formula for the local electric field at the top of the small sphere (nanotube tip):

\[
E = E_0 \times \gamma = E_0 \times \frac{l}{D} \left( 1 + \frac{d}{r} \right),
\]

with \( \gamma = l/D[1+(d/r)] \), \( E_0 = V/d \) being the reference electric field. This formula is in excellent agreement with the experimental data in the entire range of \( d \) values measured, as shown in Fig. 3. For \( d \ll D \) (small gap regime), Eq. (3) reduces to

\[
\gamma = 1 + d/lr,
\]

which agrees excellently with the experimental results [Fig. 3(a)]. Microscopic measurements of the nanotube diameters agree very well with those obtained from the slopes of Fig. 3(a). For \( d = D \) (large gap regime) Eq. (3) yields the well-known approximate result

\[
\gamma = l/r.
\]
of at least 1 000 000. Such a giant field enhancement could revolutionize the field emission applications of nanotubes.

In conclusion, we find that the multistage emitter geometry, such as that of a large nanotube grown on a tip of a carbon fiber, followed by a small nanotube on the tip of the large nanotube, etc., can lead to a very large field enhancement at the tip of the smallest nanotube in the chain, and consequently to a very strong field emission. We show that the field-enhancement factor of emitters with a multistage structure is a product of the individual field-enhancement factors of individual stages, and that this not only explains the giant field enhancement that we have reported earlier in a large area measurement of carbon nanotubes grown on carbon cloth, but also provide important implications for the design of nanotube emitters for electronic devices.

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