Geometrical enhancement of field emission of individual nanotubes studied by in situ transmission electron microscopy

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Field emission of an individual multiwalled carbon nanotube, driven by a customer-built piezomanipulator, was measured in a transmission electron microscope. The measurement geometry and the nanotube structure were imaged in situ. A linear dependence of field enhancement factor $\beta$ on the distance $d$ between the nanotube tip and its counteranode is found. The enhanced field emission mechanism is studied by a tip-flat emission model. The results indicate that the radius of emission apex $r$ is an important factor in field emission with a relationship of $\beta \approx r^{-1/2}$, while the tube length has little influence on $\beta$. © 2006 American Institute of Physics.

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It has been well known that the local electrical field at the tip of the needle-shaped emitter is much higher than the mean electrical field, which is due to a field enhancement effect. The one-dimensional nanomaterials, such as nanotubes, nanowires, and polymerized nanobells have a high aspect ratio of length to radius, which are promising to be served as electron emission sources. A high field enhancement factor $\beta$ makes a low turn-on voltage and high emission current, which is desired for emitter applications. To study the field emission mechanism, several models of a “floating sphere at emitter-plane potential” and a “hemisphere on a post” are often used theoretically to understand the field enhancement effect. So far, various field emission enhancement effects induced by the geometrical parameters, including the measurement environment and geometrical size of the emitter, have been reported. Bonard et al. studied the field emission of an individual nanotube in a scanning electron microscopy. Their experimental result is consistent with the prediction of the Edgcombe and Valdrè model. But the other experiments deviate from these models. Usually, the measurement setup is more complicated, which results in the difference one from the others.

In this Letter, we will present an in situ transmission electron microscopy (TEM) measurement, which quantifies the dependence of the field emission factor on the distance between the nanotube tip and its counterelectrode. By using the measurement geometrical environment as a boundary condition, the applied electric field was mapped theoretically based on a tip-flat field emission model. The calculated results of the field enhancement factor are in agreement with the experimental data. The effects of the nanotube radius and length on the field enhancement are also discussed.

For an in situ TEM measurement, a special TEM specimen holder is designed and built up in a JEOL 2010 FEG TEM operated under the vacuum of $10^{-7}$ Torr and at room temperature. By using this setup, the field emission properties of carbon nanotubes with well-defined structures were studied previously. The schematic frame is shown in Fig. 1(a). An electrochemically etched tungsten needle serves as the movable cathode, and its opposite is a melted gold wire with a ball shape at the end as the anode. The distance between the two electrodes can be precisely controlled by the piezomanipulator. Figure 1(b) shows an individual carbon nanotube fixed on the tungsten tip. The TEM beam is blanked off during field emission measurement and the nanotube is discharged in advance.

To study the dependence of the field enhancement factor on the distance between the nanotube tip and its counterelectrode, as denoted by $d$ in Fig. 1(b), the field emission measurement was performed as a function of the distance $d$. The corresponding $I$-$V$ and $F$-$N$ curves are plotted in Fig. 2(a) and Fig. 2(b), respectively. The $F$-$N$ curves are basically fitted to straight lines, indicating that the field emission of an individual nanotube still follows the Fowler-Nordheim law. The turn-on voltage decreased with decreasing the distance $d$. From the slope of the $F$-$N$ plots, the enhancement factors $\beta$ are measured to be several tens to more than one thousand. Basically, it is found that $\beta$ increases linearly with the increasing of distance $d$ from 1.2 to 46.8 $\mu$m, as shown in Fig. 2(c).

It is noted that the relationship between $\beta$ and $d$ is not consistent with the Edgcombe and Valdrè model. Therein $\beta = \beta_0 \times [1 + A \times (d/(d+l))^3 - B \times (d/(d+l))]$, while $l$ is the length of a tube, and $A$ and $B$ are constants. In order to understand the present experiments, further theoretical calcu-
lations are carried out by a tip-flat field emission model. Here, the finite element method is used to solve the Laplace equation \( \nabla^2 \varphi = 0 \). To get the real distribution of the applied electric field, it is important to set up an appropriate boundary condition. On the basis of the actual experimental setup, the boundary condition was defined as in Fig. 3(a). By comparing with the nanotube radius, the gold ball can be reasonably considered as a flat. Unlike a normal model with two flat electrodes, we use a flat electrode for the gold ball and a needle electrode for the nanotube in the present experiments. A tungsten tip is also presented in the boundary condition. The electric potential at an emission electrode is set to \(-100\) V and the counterplane electrode is 0 V. The potential far away from the two electrodes is also set to 0 V. The effective electric field \( E_{\text{eff}} \) is defined as the maximum electric field at the emitting tip. The mean electric field \( E_{\text{mean}} \) is defined as the electric field near to the anode. Thus, the field enhancement factor \( \beta = E_{\text{eff}} / E_{\text{mean}} \). In the study, the radius curvature of the apex is set to 50 nm. The \( \beta \)'s were calculated when the \( d \) value varies from 0.8 to 12 \( \mu \)m. Figure 3(b) shows the distribution mapping of the applied electric field. Figure 3(c) shows a linear relationship between \( \beta \) and \( d \), which is in agreement with the experimental results.

Furthermore, we have studied the effect of the tube length \( l \) on \( \beta \). In this case, \( r \) is kept as a constant. The calculation shows that \( \beta \) is not sensitive to \( l \) while \( d \) is on the order of several micrometers. As shown in Fig. 4(a), there is no a clear relationship between them for larger \( d \). We have also studied the dependence of \( \beta \) on the nanotube radius \( r \) while \( l \) is kept as a constant. It indicates that \( \beta \) increases very quickly with the decreasing of \( r \), as shown in Fig. 4(b). It is clear that the tube radius plays a key role in the field enhancement. The relationship between \( \beta \) and \( r \) can be approximately fitted to \( \beta \sim r^{-1/2} \), which is consistent with the results in Ref. 16.

Herein, the dependence of the field enhancement factor on geometry parameters, including the distance \( d \) between the tube tip and its counteranode, the tube length \( l \), and the apex radius \( r \), have been studied systematically. The field emission mechanism of an individual carbon nanotube is revealed by the experimental measurements together with theoretical calculations. Because of the complication of the measurement environments and the sample conditions, many proposed models vary one from the other in previous studies. For example, the shape of the two electrodes is important to determine the field emission property. In the present study, our anode is a flat electrode, while the cathode is a long needle. Therefore, a tip-plane model is proposed based on the present experimental configuration. A linear relationship of \( \beta \) vs \( d \) is valid for a large range of \( d \). But in a flat-flat model, \( \beta \) approaches to a constant when the distance of the two flats is about 100 times larger than the nanotube radius.14 In that case, when the anode flat is sufficiently far away from the emitter tip, the \( \beta \) is mainly determined by the emitter

![FIG. 2.](image1.png) (a) I-V curves of the field emission for different distance \( d \). (b) Corresponding F-N plots. (c) Dependence of \( \beta \) on \( d \).

![FIG. 3.](image2.png) (Color) (a) Scheme of boundary condition in numeric simulation for electric field. \( E_{\text{eff}} \) is defined as the maximum electric field at the emitting tip. \( E_{\text{mean}} \) is defined as the electric field at the plane electrode opposite to the emitting tip. (b) Calculated electric field mapping for two different \( d \). (c) Calculated relationship between \( \beta \) and \( d \).
geometry. In a tip-tip emission setup,\textsuperscript{10,13} this asymptotic line is also observed. In our study, however, no asymptotic behavior has been found even at a $d$ about 800 times larger than the nanotube radius. In addition, the tip-flat model shows that nanotube length has little influence on the field enhancement factor $\beta$, different from other models. Our results indicate that electrode geometry plays an important role for the field emission property of an individual nanotube.

In summary, the field electron emission property of an individual carbon nanotube has been studied by \textit{in situ} TEM measurements and theoretical calculations. The field enhancement factor $\beta$ increases linearly with the increasing of the distance $d$ between the nanotube tip and its counter anode. A tip-flat field emission model is proposed based on the experimental details. Further theoretical calculations show that the tube length have little influence on $\beta$, while the radius of emission apex $r$ is an important factor in field emission with a relationship of $\beta \propto r^{-1/2}$. This result will be useful to understand the field emission mechanism of an individual carbon nanotube.

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