Low noise and stable emission from carbon nanotube electron sources

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Remarkably stable and low noise field-emission electron sources have been obtained using individual carbon nanotubes. The maximum current fluctuation observed over 1 h was merely 0.5% and the emission frequency spectrum exhibited $1/f$ behavior over a bandwidth of 0.1–25 Hz, above which the shot-noise limit was reached. The average noise percentage was determined to be 0.08% (i.e., a signal-to-noise ratio of 1250) for seven CNT emitters operated at several current levels. The influence of the vacuum level was also investigated. © *2005 American Institute of Physics*.
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Carbon nanotubes (CNTs) are being investigated as next generation high-quality electron point sources.[1] Several types of devices are under active consideration; for example, field-emission displays,[2] x-ray sources,[3] and electron sources for electron microscopes.[4] It is expected that CNTs have several advantages[1] over field-emission sources made from sharp metal tips (typically tungsten), which have been used for many decades.[5] The drawback of a cold field emitter of metal is that the current fluctuations are too large; these are caused by surface migration of atoms under the high field and also by back ion sputtering. The CNT is not a metal, but a highly ordered crystalline structure built by covalent bonds. Each carbon atom is bound to three other carbon atoms by a covalent $sp^2(\sigma)$ bond. The threshold for the removal of one atom is 17 eV (Ref. 6), much higher than the activation energy for surface migration of a tungsten atom (3.2 eV) (Ref. 5). Thus, the CNT should be much less sensitive to surface migration of the carbon atoms. In addition, CNTs have an extremely large Young's modulus and a high tensile strength. With no (or few) dangling bonds, CNTs are also chemically inert and react only under extreme conditions or at high temperature with oxygen or hydrogen.[7] Moreover, carbon has one of the lowest sputter coefficients.[8] The current emitted from a CNT is, therefore, expected to be highly stable in comparison with metal emitters and the CNT itself is expected to be a robust emitter, even at high temperatures.[9] In this work, we quantitatively measure the stability and noise of the CNT emitter.

Field emission is described by the Fowler-Nordheim (FN) model,[10] in which the emitted current I is an exponential function of the electric field F at the emitting tip with a radius of curvature R and work function ϕ (Ref. 11),

$$I \cong c_1 \frac{(RF)^2}{\phi} \exp\left\{-c_2 \frac{\phi^{3/2}}{F}\right\}, \qquad (1)$$

where c_1 and c_2 are constants and $F = U\beta$, with the extraction voltage U and the field enhancement factor β. The current fluctuations have two major causes, variations of F and variations of ϕ, for which I is extremely sensitive due to the exponential nature of Eq. (1). A 1% change in F can readily lead to a 15% change in I. Local changes of F are likely to occur due to geometric changes of the tip caused by surface migration in the extremely large electric field at the tip and from ion bombardment.[5] On the other hand, ϕ is sensitive to adsorbates on the surface and can change by 25%, which may lead to a two orders of magnitude change in I. Even when operating in ultrahigh vacuum and with a fully baked field emitter, a layer of adsorbates forms after a few tens of minutes followed by the unavoidable dynamic process of adsorbation, vaporizing, and molecular flow which occurs at room temperature. To obtain the best performance, field emitters usually have to be flashed (i.e., quickly heated) regularly and they are often operated under extremely good vacuum ($<10^{-10}$ torr) with special electronic feedback loops to stabilize the current.[5] The CNT, being a highly crystalline covalent structure,[6] does not suffer from surface migration and hence can be operated at a few hundred degrees Celsius to continuously clean the emitter through desorption. As a result, the vacuum requirements for the CNT emitter could be relaxed in comparison to that required for the metal emitters.

Individual multiwalled CNTs of both arc-discharge[12] and chemical vapor deposition (CVD) (Ref. 13) type were mounted on tungsten support tips using a previously reported procedure.[14] Small diameter (2–5 nm) CNTs with a closed cap at the emitting end were used. The CNT electron source was then transferred into an ultrahigh vacuum system (10^{-10} torr) for characterization of its electron emission properties. The nanotube was heated first to the carbonization temperature[7] of 700 °C for 10 min to remove volatile species from the tube. The nanotube was then operated at a temperature of 500 °C during the emission experiments to continuously keep the emitter clean. The emission pattern of CNT 1 (CVD grown) is shown in Fig. 1(a) and is typical for a CNT with a closed cap.[14]

The current-voltage characteristic of CNT 1 was measured and fitted to the FN model. The full experimental method is described elsewhere.[15] The FN plot, as shown in Fig. 1(b), is a linear curve. The value of β was determined to

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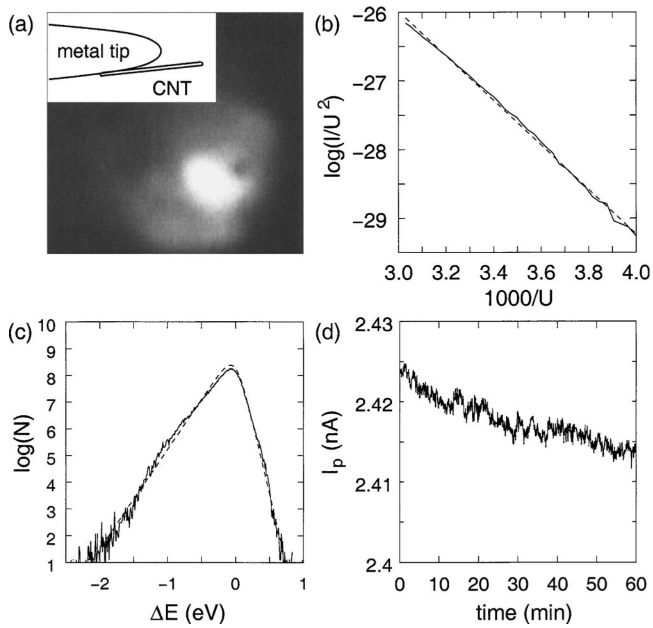

FIG. 1. Emission measurements on electron source CNT 1 for $T=500$ °C and a vacuum of 2×10^{-10} torr. (a) Field-emission pattern recorded with a microchannel plate and a phosphor screen. Inset: schematic drawing of a CNT on a support tip. (b) FN plot and a linear fit with a slope of −3244 (dashed). (c) Energy spectrum recorded at $U=290$ V, $I=130$ nA, and a fit with the FN theory (dashed). (d) I_p measured as function of time at $U=270$ V and $I=190$ nA.

amount to 2.3×10^{7} m^{-1} and R was 1.0 nm. These are values typical of our CNTs (Ref. 15). The energy spectrum of the emitted electron beam is plotted in Fig. 1(c) and fits well with the expected energy distribution $I(E)\propto(\exp E/d)/(1+\exp E/k_bT)$ (Ref. 11), with the tunneling parameter d, temperature T, and the constant of Boltzmann k_b. The values extracted from Fig. 1(c) are $d=0.27$ eV and $T=500\pm50$ °C. The data of the FN plot and the energy spectrum can be combined to obtain ϕ, using $\phi=-1.64bd/U$ (Refs. 15 and 16), with the slope of the FN plot b and the extraction voltage U at which d was measured. For CNT 1 the resulting value $\phi=5.0$ eV, as expected.[15] Data of several other CNTs showed the same consistency with the FN model.[15]

The stability of the emitted current of CNT 1 was determined by running the source at a constant voltage provided by a high-stability power supply [in-house design, with less than 50 parts per million (ppm) ripple and 5 ppm drift]. The probe current I_p was collected in a Faraday cup with an opening diameter of 1 mm, connected to a preamplifier (Perkin Elmer). Figure 1(d) shows that the emitted current has a maximal drift of 0.5% over 1 h. The drift is probably caused by a small drift of the temperature. The short-term peak-peak fluctuations were smaller than 0.2%. The different $I-U$ behavior with respect to Fig. 1(c) is attributed to the different extractor geometries used in both measurements and a difference in temperature. The normalized angular current density of the probe beam was 4.9 nAsr^{-1} V^{-1}, representative for a high-brightness electron beam.[1,4]

The frequency characteristic of the emission process was investigated by recording the fast Fourier transformation (FFT) of I_p with a spectrum analyzer (Hewlett Packard). The related root-mean-square (rms) voltage $U_{\rm rms}$ is plotted in Fig. 2(a). The background voltage $U_{\rm bkg}$ (i.e., the noise floor of the measuring system) was also recorded, with the CNT electron source switched off and fitted to a constant. The

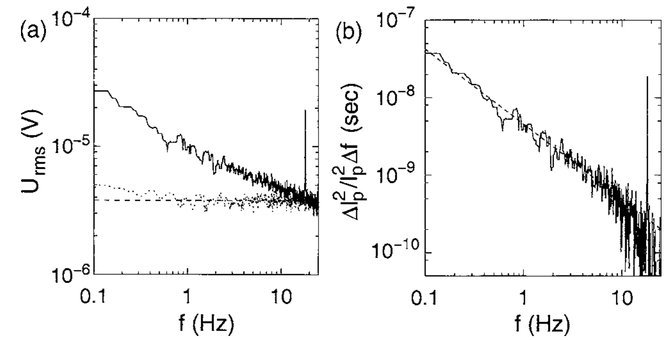

FIG. 2. Spectral analysis of the probe current of CNT 1. (a) $U_{\rm rms}$ as function of f. The data presents the average of 10 measurements recorded in a period of 10 min. The noise of the measuring system (dotted) and a constant fitted to it (dashed) are also plotted. (b) The normalized spectral density of CNT 1 and a fit with the function $y=a/x^b$ (dashed).

signal of the CNT was corrected for the background using: sqrt$(U_{\rm rms}^2-U_{\rm bkg}^2)$. Finally, the signal was normalized on the average probe current to obtain the normalized spectral density $S_n(f)=\Delta I_p^2/I_p^2\Delta f$, with the measuring bandwidth Δf. $S_n(f)$ gives the spectral noise power of the emitter, as shown in Fig. 2(b). Note that this spectrum appears to increase in variation at the higher frequencies; this is actually an artifact of the measurement and arises because the signal becomes increasingly buried in the noise floor of the measuring system at higher frequencies. A peak is also clearly visible at 18 Hz. This peak corresponds with typical mechanical vibrations in the measuring system and is not related to the actual noise characteristic, and hence is ignored in the following analysis.

Defining $S_n(f)=a/f^b$ and fitting this to our results yields $a=(4.5\pm0.1)\times10^{-9}$ sec and $b=0.97\pm0.01$. Thus, the emission frequency characteristics of the CNT electron source exhibits $1/f$ behavior. At 25 Hz in Fig. 2 $S_n(f)$ is almost equal to the shot-noise limit $S_{n,\rm shot}(f)=2e/I_p=1.3\times10^{-10}$ Hz^{-1} for our $I_p=2.4$ nA, which is in general the lowest possible noise level. Measurements at frequencies up to 3 kHz showed that $S_n(f)$ was below the noise floor of the measuring system and did not unexpectedly increase above this level. We expect, therefore, that $S_n(f)$ of the whole frequency range can be described by

$$S_n(f)=\frac{a}{f}+\frac{2e}{I_p}. \tag{2}$$

The left-hand term is the fit of the observed noise from the CNT and the right-hand term accounts for shot noise.

Integrating the measured $S_n(f)$,

$$\int_{f_1}^{f_2} S_n(f)df=\frac{\langle\Delta I_p^2\rangle}{I_p^2}\equiv np^2, \tag{3}$$

leads to a noise percentage np of 0.02% as measure for the emission stability in the frequency range 0.1–25 Hz. Inverting this value gives us the signal-to-noise ratio of the CNT emitter to be 5000. Six additional CNT electron sources were characterized: three deposited by arc discharge and three from chemical vapor deposition. Measurements were also recorded at different total currents and different vacuum levels. As can be seen in Fig. 3(a) the CNT emitter can be operated at currents up to a few microamperes without losing its stability. The average value of $np=0.08\%$ with a standard

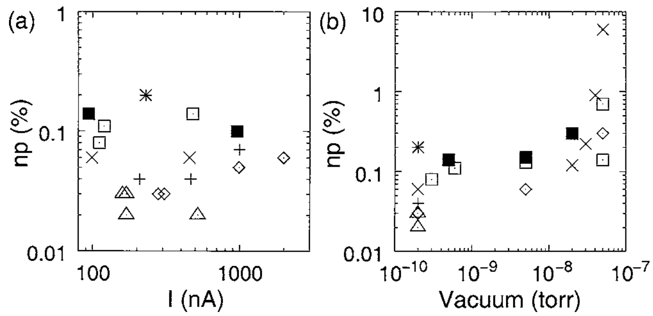

FIG. 3. Noise percentages of seven CNTs measured in the frequency range 0.1–25 Hz with an integration time of 10 min. The probe current was typically 2–10 nA, depending on the total current level and the distance from the CNT to the Faraday cup. The temperature was between 400 and 600 °C. (a) Noise percentage as function of the total current for a vacuum level of 10^{-10} torr. (b) Noise percentage as function of the vacuum level for total current levels of 90–500 nA.

deviation of 0.06% for all seven CNTs. The worst *np* measured was 0.2%, corresponding with signal-to-noise ratio of 500. Small differences between the structures of the CNTs possibly explain the differences in the values of *np*.

The vacuum level is an important factor that influences the noise observed in the emission current. At lower vacuum levels, the rate of molecules adsorbing and subsequently desorbing (because the CNT is at 500 °C) becomes higher, thus affecting the emitter stability.[5] We measured *np* as a function of vacuum level as shown in Fig. 3(b). Stable operation was maintained up to a vacuum level of 5×10^{-9} torr. At higher levels, the *np* increased and occasionally large (>10%) current fluctuations occurred.

The current fluctuation and noise percentages of the CNTs measured here are extremely low. Metal cold field emitters typically exhibit 3–5% fluctuations of the probe current under similar measurement conditions.[5,17] The value $np=0.08\%$ is comparable with that obtained from the state-of-the-art Schottky emitter of approximately 0.1% in the frequency range 1–25 Hz (Refs. 17 and 18) (N.B. the Schottky emitter is often chosen in the electron microscopy industry over metal field emitters because of its stability). Thus, the CNT is a stable and low noise electron point source.

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