Effect of ambient pressure on resistance and resistance fluctuations in single-wall carbon nanotube devices

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We report low-frequency resistance fluctuation (noise) measurements in semiconducting and metallic Ti/Au-contacted single-wall carbon nanotube devices. In both types of devices, the noise power spectra has a “1/f” dependence, and is proportional to the squared current. Semiconducting devices were found to have three orders of magnitude higher noise levels compared to the metallic ones. In vacuum, the resistance increases but noise decreases by over an order of magnitude for both metallic and semiconducting devices. The resistance and noise levels recover to their original values when the samples are brought back to atmospheric pressure. Both noise and resistance change simultaneously when the chamber is evacuated. However, when the chamber is brought back to atmospheric pressure, the noise level takes several tens of hours longer to recover. © 2006 American Institute of Physics. [DOI: 10.1063/1.2218265]

The electronic transport properties of single wall carbon nanotubes (SWNTs) have attracted tremendous attention in recent years, owing to their potential application as ballistic conductors, field effect transistors, nonvolatile memories, and chemical sensors. Nanotube based device properties have been shown to be very sensitive to the ambient conditions. Pressure, temperature, surface passivation, and annealing can significantly affect the charge injection into the nanotube conductance channel, and hence its conductivity. In any electronic device, a relevant parameter of extreme importance is the level of resistance fluctuations or noise, which can seriously affect the device performance and reliability. It is hence, important to characterize the noise in single nanotube devices, and understand the mechanism responsible. In particular, since the transport properties of typical nanotube devices are extremely sensitive to ambient conditions, it is of interest to investigate how the noise level responds to...
changes in ambient conditions. In this paper, we report the effect of the environment on the noise characteristics of two terminal devices constructed from individual SWNTs.

SWNTs were grown by thermal chemical vapor deposition (CVD) on oxidized Si substrates. The 100 nm oxide layer provided an insulating substrate, while the conducting Si acted as a back gate. High degree of control over the CVD conditions yielded a very low density of SWNTs that were on average about 2 μm long. The nanotubes were contacted by the Ti/Au metal bilayer electrodes having 1 μm separation, fabricated by electron-beam lithography, to yield devices containing one or a few individual SWNTs.6

Low-frequency noise was measured in the frequency range of 1 Hz < f < 3000 Hz. The voltage fluctuations ΔV(t) across a 10 kΩ resistor connected in series with the nanotube device was analyzed by a SR770 Network Analyzer. Figure 1(a) shows a schematic circuit configuration for current-voltage characteristics (I-V) and noise measurements, embedded with a tapping mode atomic force microscopy image of a typical single-SWNT device. A schematic view of a SWNT—metal contact area is shown in Fig. 1(b) (to be discussed later).

SWNTs can be either metallic (M-SWNT) or semiconducting (S-SWNT) depending on their chirality,7 despite their fundamentally similar structure based on a rolled-up graphite sheet. The randomly grown nanotubes gave a sizable number of each kind of devices. Metallic devices had linear and symmetrical current-voltage characteristics [see Fig. 2(a)], which were independent of the gate voltage V_G. The resistances of these devices range from 0.5 to 5 MΩ several orders of magnitude higher than the quantum limit h/4e^2 = 6.45 kΩ.8 Semiconducting nanotube devices demonstrated nonlinear, asymmetrical, and gate voltage dependent current voltage characteristics. Figure 2(b) shows an example of the current-voltage characteristic for the S-SWNT device in atmosphere and in vacuum. Asymmetry and nonlinearity of the S-SWNT current-voltage characteristics can be attributed to the Schottky contacts. Simulation studies have shown9 that the characteristic length of the Schottky barrier space charge region is 5–10 nm [see Fig. 1(b)]. The dependence of the current on the gate voltage at fixed lateral voltage V=5 mV is shown in the inset in Fig. 2(b). As seen, the current increases with an increase of the negative gate voltage, confirming the p-type conductivity of the S-SWNTs.

Figures 3(a) and 3(b) show the relative spectral noise density S_n/Δf^2 of the short circuit current fluctuations measured at different voltages (currents) for M-SWNTs and S-SWNTs. Both metallic and semiconducting SWNTs exhibited 1/Δf^n noise with the exponent γ close to unity (γ=1.0 ±1.1). As seen from Figs. 3(a) and 3(b), the spectral noise densities S_n/Δf^2 measured at different currents coincide indicating that the absolute value of noise S_n scales as S_n ∝ Δf^2. This result was reported earlier for individual nanotubes and two-dimensional (2D) networks,10,11 with linear current-voltage characteristics. On the other hand, deviations from the S_n ∝ Δf^2 law were also reported for both single walled and multiwalled (iron filled) nanotubes with linear and nonlinear current-voltage characteristics.12–14 Those results indicate different possible origins of the 1/Δf noise in nanotubes grown under different conditions and fabricated with different contacts. The S-SWNTs under study demonstrated very

![Image](https://example.com/image.png)
strong nonlinearity [see Fig. 2(b)]. The dependence $S_f \propto I^2$ for such strongly nonlinear nanotube devices has not been reported before.

Provided the fluctuations of the contact resistance $R_C$ and fluctuations of the entire nanotube resistance $R_N$ are uncorrelated, the spectral noise density of the current fluctuations can be written in the form,

$$\frac{S_f}{I^2} = \frac{S_{R_C} + S_{R_N}}{(R_C + R_N)^2},$$

(1)

where $S_{R_C}$ and $S_{R_N}$ are the fluctuations of the resistances $R_C$ and $R_N$, respectively. As seen from the exponential dependence of the device current on the applied bias [Fig. 2(b)] in S-SWNTs, the total resistance of the S-SWNT device is dominated by the Schottky contacts ($R_C \gg R_N$). As mentioned before, simulations have shown that the potential distribution in a S-SWNT with a Schottky contact has shown that the length of the space charge region $L_{SCR}$ is about 5–10 nm. Therefore, the resistance $R_N$ and its fluctuations $S_{R_N}$ outside of this narrow space charge region are nearly constant (i.e., independent on current), since the length $L_{SCR}$ is much shorter than total SWNT length of about 1 μm. The assumption that $S_{R_C} \gg S_{R_N}$ is consistent with the observed independence of $S_f/I^2$ on current. The opposite assumption, $S_{R_C} \ll S_{R_N}$, would lead to an exponential dependence of noise on current in the S-SWNT devices [according to Eq. (1)], which was not observed. Therefore we conclude (in agreement with the previous studies) that the $1/f$ noise of S-SWNT originates from the contacts ($R_C \gg R_N$) and both $S_{R_C}$ and $R_C$ decrease with the current increase keeping the ratio $S_{R_C}/R_C^2$ constant. However, we note that the thickness of the Schottky barrier is finite and depends on the bias. Therefore, both nanotube/metal metallurgical interface and part of the nanotube adjacent to the metal should be included in the definition of contact and contact noise as shown in Fig. 1(b). It is known that oxygen atoms binding to the nanotube devices can $p$-type dope the nanotube and/or modify the metal-semiconductor barrier. Therefore, possible noise mechanisms could be fluctuations of the barrier height and/or fluctuations of the effective doping of the nanotube inside the depleted region.

The electrical properties of SWNTs have been reported to be sensitive to their environment, and various chemical sensor applications for SWNTs have been suggested. Among the most significant effects are those of oxygen and moisture, both of which are invariably adsorbed on the nanotubes and contacts to the nanotubes from exposure to atmosphere. We also found that when the nanotube devices were placed in vacuum, the resistance of the devices consistently increased with time for both metallic [Fig. 2(a)] and semiconducting [Fig. 2(b)] devices, consistent with previous reports.

Noise was measured in both metallic and semiconducting nanotube devices in atmospheric environment and under vacuum, repeated over several cycles. We found that the noise levels in both kinds of devices showed high sensitivity to the ambient pressure. Figure 4 shows the noise amplitude ($S_f/I^2 * f$) for the M-SWNT and S-SWNT devices. In both metallic [Fig. 4(a)] and semiconducting [Fig. 4(b)] devices, the noise amplitude decreased by about an order of magnitude in vacuum (similar result was obtained in Ref. 15). When the devices were restored to atmospheric pressure, the resistance and noise levels were restored to the original values, however, the noise level took significantly longer time to recover compared to the resistance. Even when the resistance was restored completely to its original value after several hours of the exposure to the atmosphere, the noise remained substantially lower than its initial value and required several tens of additional hours to get restored to the noise level in the atmosphere environment. As an example, Fig. 4(a), line 2 shows $S_f/I^2 * f$ for an M-SWNT device in vacuum. Then, the device was exposed to the atmosphere and kept there for ~24 h. As a result the initial value of resistance of 0.85 MΩ was completely restored. However, the noise level remained almost an order of magnitude smaller than its initial value at atmosphere [Fig. 4(a), line 1’]. This indicates a very high
sensitivity of noise (higher than that for the resistance) to the environment. Such dependence of noise might be used for potential chemical sensor applications. The similar noise behavior of M-SWNT and S-SWNT devices allows us to suppose the same origin of noise in these two types of nanotubes.

The noise amplitude $A = S_f/I^2 \cdot f$ is often empirically related to the resistance of the device according to the relation $A = 10^{-11}R$. Although different versions of this relationship have been proposed,$^{10}$ $A/R$ has been found to be close to a constant. We found that in our experiments the $A/R$ ratio varied from $10^{-9}$ to $10^{-11}$ for both M-SWNT and S-SWNT devices. However, as was discussed before, the change in noise and resistance in vacuum is in different directions: the resistance increases but the noise decreases when the devices are placed in vacuum. Therefore, the $A/R$ ratio depends on the environment and this ratio does not reflect the noise nature and can be used only for qualitative characterization of noise level.

The situation when the resistance of the electronic device increases and noise decreases at the same time is not typical. For example, the noise from channel of the field effect transistor usually increases when the channel is getting pinched off. In the forward biased $p-n$ diode, the relative spectral noise density $S_f/I^2$ decreases with increase of the forward current and decrease of the differential resistance.

Since the resistance of the S-SWNT devices is limited by the Schottky contact, we deduce that the device resistance rises in vacuum because the desorption of naturally adsorbed gas molecules (in ambient conditions) on the nanotube and on the metal electrodes changes the Schottky barrier height.$^{16-18}$ It is found that the M-SWNT device resistance also increases in vacuum. In both cases, it is consistent with the picture that the presence of adsorbed species can modify the lineup of the CNT bands at the metal-nanotube interface relative to the metal Fermi level.$^{18}$ The adsorption/desorption of gas molecules in this interface region, denoted by a circle in Fig. 1(b), plays a significant role in determining the observed behavior of resistance and noise. By removing the adsorbed species in vacuum we change the resistance of the device. At the same time, decreasing the concentration of the adsorbed species also diminishes the fluctuations in their concentration, therefore reducing the noise.

Another way to characterize the noise level is calculating the Hooge parameter, $\alpha_{H}=(S_{f}/P_{0})n$, where $N$ is the total number of carriers in the sample.$^{20}$ This parameter $\alpha_{H}$ has been previously used to characterize noise in carbon nanotubes and was shown to be $\alpha_{H}=4 \times 10^{-3}$.$^{11}$ Taking for the upper bound estimate the number of carriers equal to the number of atoms (as was done in Ref. 14) we obtained $\alpha_{H} \sim 1 - 10$, for M-SWNTs. Note that, the Hooge parameter has some physical meaning only for the bulk origin of noise. Since we concluded that contacts or the parts of the carbon nanotubes adjacent to the contacts are the prime source of noise in S-SWNTs, the Hooge parameter might find only limited meaning for the characterization of the noise level in carbon nanotube devices.

In conclusion, our studies of low-frequency (1/f) noise show that the noise level scales with $I^2$ in both S-SWNT and M-SWNT based devices. Both metallic and semiconducting devices show one order of magnitude or more decrease of the noise level in vacuum compared to the ambient level, although the resistance increases with lowering the pressure. After return to atmospheric pressure, the noise level was found to take much longer time to recover to its atmospheric pressure value than the resistance. In nanotube devices, contacts play an important role, and extend into the part of the nanotube geometrically outside the topological metal contacts. We argue that the measured fluctuations result from the changes of the electrical properties of this extended contact region, and show that it is sensitive to small variations in the environment pressure. The diminishing of noise under vacuum is an extremely favorable step towards the usage of nanotubes in future nanoelectronics, and is an important result in understanding the nature of electron transport in low-dimensional systems. In addition, the high sensitivity of noise to the environment shows a possible potential for chemical sensor applications based on carbon nanotube noise characteristics.

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