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Linear current fluctuations in the power-law region of metallic carbon nanotubes

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We study low-frequency noise in a non-Ohmic region of metallic single walled and multiwalled carbon nanotubes. The generalized relative noise appears to be independent of applied bias in the power-law regime of the tubes and in agreement with theoretical predictions. Beyond the power-law regime the suppression of conductance due to scattering with optical phonons is accompanied by a reduction of relative noise by an order of magnitude. Mobility fluctuations in the tubes due to optical phonon scattering cause the unexpected reduction in the relative noise magnitude which is modeled using a modified mobility fluctuation picture. The findings have important implications for metallic nanotubes being used as interconnects in nanoelectronic devices.

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I. INTRODUCTION

Low dimensional and disordered systems are easily driven into the nonlinear regime upon application of even very small bias voltages. In the linear regime, the resistance noise is analyzed within the realm of fluctuation-dissipation theorem.[1] Nanoscale devices and interconnects operate mainly in the nonlinear regime where the noise analysis through conventional means is arduous as the fluctuations become a function of applied bias. The dearth of literature dealing with noise in the nonlinear regime can also be partly attributed to it though it is an old but unresolved problem. Theoretical efforts in the past by Rammal *et al.*[2] first mooted the idea of analyzing noise in the nonlinear regime. Cohn's theorem[3,4] was used by the authors to derive exponent inequalities in charge density wave (CDW) systems and metal-insulator composites having I-V characteristics of the form of a power law. However, no experiment has explored the behavior of noise in this regime despite the fact that in the power-law regime noise analysis is simplified compared to other nonlinear regimes (e.g., where conductance is expanded in a Taylor series of dc bias voltage). Due to widespread nonlinear I-V properties, especially power-law I-V relations in nanoscale objects,[5–8] the measurement and interpretation of noise in this region have become increasingly significant.

For performing noise experiments in the nonlinear regime, metallic carbon nanotubes (CNTs) can be used as good test samples as they display power-law variation of conductance with bias voltage at low temperatures. The power-law dependence in metallic single walled nanotubes (SWNTs) is widely believed to be a signature of *Luttinger liquid* (LL) behavior[8,9] while in metallic multiwalled nanotubes (MWNTs) the origin has been ascribed to an *environment-quantum-fluctuation* process.[10,11] In this paper, we measure and analyze noise in the power-law regime and beyond in carbon nanotubes. The objective is to show that the generalized relative noise in a power-law regime remains independent of applied bias thereby making it possible for fluctuations in this regime to be analyzed in a manner similar to that in the Ohmic regime. To illustrate the usefulness of this analysis we show that by measuring noise beyond the power-law regime optical phonon (OP) scattering reduces the magnitude of relative noise from the constant value in the power-law regime. We propose a modified mobility fluctuation model which seems to explain the noise behavior beyond the power-law regime in SWNTs. Noise in the power-law regime is also measured and analyzed for MWNTs where a larger, cleaner power-law region exists with negligible OP scattering effects.

Thermal noise is usually dependent on sample temperature and is frequency independent. For shot-noise measurements the magnitude is dependent on the current flowing through the sample and is also white. The only frequency-dependent noise is the $1/f$ noise which is usually measured by passing a constant current/voltage through it and measuring voltage/current fluctuations across it. In the Ohmic regime the resistance noise (δR) manifests itself in the fluctuations of current/voltage ($\delta I/\delta V$) and the current/voltage is merely used to make the fluctuations "visible" while playing no role in the production of fluctuations.[1,12,13] The relative noise power (A_X) can be determined using a generalized version of Hooge's empirical formula:[14,15]

$$A_X = \frac{S_X}{X^2} = \frac{V^{\gamma_o}}{f^{\lambda}}\mathrm{Re}(R), \tag{1}$$

where $X(=R,V,I)$ is the fluctuating quantity and $S_X = \langle \delta X^2 \rangle$ is the spectral density. R is the chordal resistance defined as V/I. $\gamma_o = 0$ for equilibrium resistance fluctuations and $\gamma_o \neq 0$ for a driven phenomena however in some cases it is also dependent on contact resistance. The function $\mathrm{Re}(R)$ depends upon the system under consideration. In the Ohmic regime, the relative current fluctuation is independent of bias i.e., $\delta I^2/I^2 = \mathrm{const}$ as resistance R is independent of the applied voltage. For the nonlinear regime resistance and hence fluctuations[16] become a function of the applied bias, i.e., $\delta R = \delta R(V)$ making the estimation of noise difficult. However, the situation is different in the case of I-V relations of the form[2] $I = gV^{\alpha}$, where $\alpha > 0$ and g is some generalized conductance defined as I/V^{α}. In such a case, if V is kept constant during an experiment, then current fluctuations in the sample will arise from fluctuations in g, i.e.,

$$\delta I = \delta g V^{\alpha} \tag{2}$$

The generalized relative noise A_I can then be written as

$$A_I = \frac{\delta I^2}{I^2} = \frac{\delta g^2}{g^2}. \tag{3}$$

Thus, in the power-law regime the relative noise, determined using suitable normalization and generalized Hooge's relation, is independent of bias. Any bias dependence of g can be taken into account by using the suitable normalization $\delta I^2/I^{2+\gamma_o}$. Thus we arrive at a nontrivial situation where noise in the power-law (nonlinear) regime can be determined in a manner similar to that in the linear regime. This will form the foundation using which we analyze our experimental results.

II. SAMPLE PREPARATION AND EXPERIMENTAL SETUP

The SWCNT and MWCNT devices investigated in this study were fabricated on top of a 300-nm SiO_2 layer thermally grown over highly doped Si wafers using standard electron-beam lithography. The semiconducting silicon was used as a back gate for the device. The suspension of commercial SWCNTs used were produced by NanoCyl S.A. (Sambreville, Belgium) and had an average diameter of 2 nm. MWNT samples were obtained from the Iijima group. The suspensions were then randomly deposited onto the chips. Subsequently CNTs were located using atomic force microscope (AFM) followed by metalization using a 25-nm-thick Pd layer followed by liftoff. Pd was chosen to improve the contact resistance. The transport measurements were done at several temperatures varying from room temperature down to liquid-helium temperature in a homemade dipstick. The voltage was supplied using a Yokogawa 7651 voltage source and current fluctuations were measured using a current preamplifier (Ithaco 1211) followed by a low-pass filter. The acquired time series was stored and further processed to find the spectral density using MATLAB. For all measurements the background noise was taken and subtracted from the main noise for each current value. All measurements were performed inside a rf-shielded room to get rid of electromagnetic interferences.

III. RESULTS AND DISCUSSION

A. SWNTs

To characterize the SWNTs, we first obtain the gate response as well as the I-V characteristics of the device. The AFM image of a typical CNT device is shown in Fig. 1(a). The metallic character of the nanotube is evident from the measured gate response curve in Fig. 1(b). In Fig. 1(c) the typical conductance (G) vs drain-source voltage (V_{ds}) curves for a metallic SWNT at different temperatures are shown. One can see that the linear region shrinks as the temperature decreases. At the lowest temperature 4.2 K, there is no visible linear regime within the measurement limit and a clear power-law dependence of conductance with V_{ds} is observed. For higher bias values, there is a tendency for the conductance to saturate which is present at all temperatures. The suppression of conductance is the result of electron backscattering due to emission of zone boundary or OPs.[17,18] We perform the noise measurements at 4.2 K in the the power-law regime for two metallic SWNTs, one with a high contact resistance (HR) and another with a low contact resistance (LR). This was done in order to understand any influence of contact resistance on the noise analysis for the tubes. The α values for the LR and HR SWNT are 0.9 ± 0.02 and 3.48 ± 0.06 respectively. The HR-SWNT has a diameter of 1.6 nm and resistance ~1 MΩ and the LR-SWNT has a diameter of 2.9 nm and resistance ~100 kΩ.

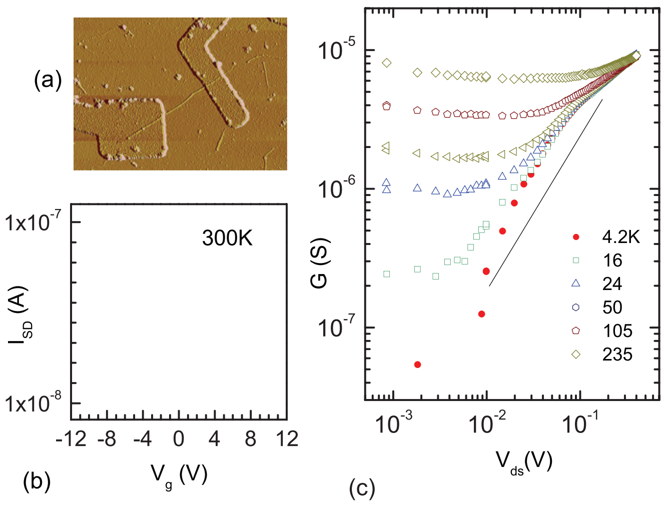

FIG. 1. (Color online) (a) The tapping mode AFM images of a metallic SWNT of channel length 700 nm and diameter 2.9 nm with top contacted Pd electrodes. (b) The source drain current I_{ds} as a function of gate voltage V_g. The metallic character of the tube is evident from the gate response at $V_{sd} = 10$ mV. (c) The conductance G vs drain source bias V_{ds} at different temperatures. At 4.2 K the conductance is a true power law within the measurement range.

In Fig. 2 we show the behavior of relative noise (A_I) as a function of V_{ds} for both the SWNTs in the power-law regime and beyond at 4.2 K. The relative noise power ($S_I/I^{2+\gamma_o}$) is extracted using the generalized Hooge's relation for both SWNTs in the frequency interval 20–40 Hz. The γ_o values for the LR and HR-SWNT are 0 and −1 respectively. γ_o, used to parametrize the noise and remove any bias dependence, might be affected by the quality of the contacts also. However this has no effect our analysis in the present case. From the plots it is clear that the relative noise power remains constant (shown by the yellow band) up to ~170 mV, i.e., up to the power-law region for both tubes. Recall that the relative noise is also expected to be a constant in the power-law regime according to Eq. (3). The situation is similar to the linear

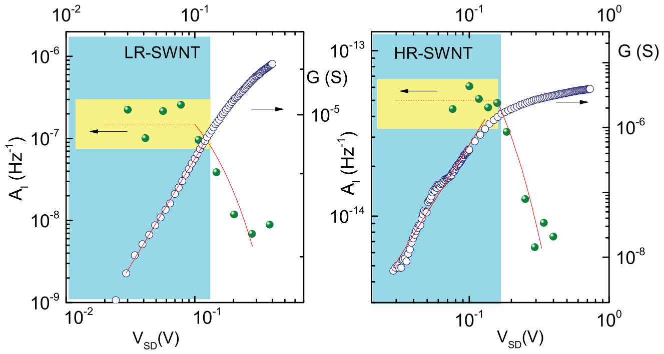

FIG. 2. (Color online) Relative noise (A_I) and conductance (G) vs V_{ds} for (a) LR-SWNT and (b) HR-SWNT at 4.2 K. The relative noise is a constant in the power-law region for both the tubes (shown by the yellow band) and is represented by the dotted line as a guide to the eye. Bold line: the fit to the noise using the expression given in the text.

regime where the relative noise (S_I/I^2) is a constant which is a consequence of Ohm's law. This is the main result of our work where we show that even in this highly nonlinear regime the relative noise behavior is the same as in the Ohmic regime. The analysis can be extended well above the power-law regime also. Beyond the power-law region (∼170 mV), the relative noise power seems to decrease with increasing V_{ds}. It is well known from various transport and optical measurements[19,20] that backscattering by OP is responsible for saturation of conductance in metallic SWNTs at phonon energies of $\hbar\Omega = 0.16$–0.17 V. This energy value coincides with the energy where the conductance starts to saturate and the relative noise (A_I) magnitude begins to reduce. This suggests that OP scattering causes the reduction in relative noise beyond the power-law regime. The detailed mechanism for the reduction in noise magnitude is explained later. Although contacts play an important role for producing noise in CNT devices, identical qualitative behavior of relative noise for both tubes proves that the origin of noise is indeed intrinsic. Furthermore, in the high bias regime we can effectively neglect the contribution of contacts[18,21] to the overall noise magnitude.

In the case of SWNTs, two-level fluctuations [random telegraph noise (RTN)] have been observed at 4.2 K in previous studies[22] which arise due to carrier trapping and detrapping from individual defects located in the oxide. In this case also the time series of fluctuations for LR-SWNTs show distinct switching between two voltage levels. The distinction between the two levels in the time series fades gradually with increase in bias and at the highest V_{ds} it completely vanishes as shown in Fig. 3. The current power spectral density (PSD) can be fitted by the sum of two Lorentzians up to $V_{ds} = 210$ mV after which the spectra has $1/f$ character. Looking at the evolution of time series, the noise in LR-SWNTs can be thought to arise from a combination of RTN from trap fluctuations and OP scattering. However, the blurring of two distinct levels in the time series as well as distribution of fluctuations anticipate the onset of OP scattering (0.17 V) effectively ruling out any correlation between the two noise sources. For the HR-SWNTs, we do not observe any RTN and the noise remains $1/f$ throughout the measurement range. This is due to the reason that in HR-SWNTs noise rises above the background only after a relatively high bias value (∼80 mV) making observation of RTN difficult which is usually observable at low bias values.[23]

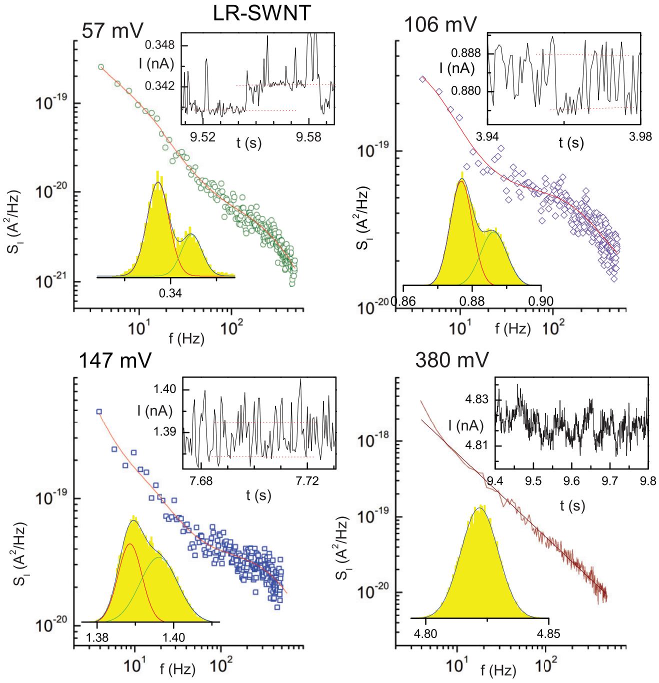

FIG. 3. (Color online) Change in nature, distribution, and PSD of noise with increase in V_{sd}. (a) Time series (top inset), distribution of the fluctuations (bottom inset), and the PSD of the fluctuations for a bias of (a) 57 mV, (b) 106 mV, (c) 147 mV, and (d) 380 mV. The two levels in the time series are indicated by red dashed lines. The PSD of the fluctuations can be fitted by Lorentzian for lower bias values. At high bias values PSD is characteristic of $1/f$ noise.

B. Model

Recall that the unusual reduction in the magnitude of relative noise with bias shown in Fig. 2 has been ascribed to OP scattering. In semiconducting carbon nanotubes the origin of low-frequency noise has been attributed to number fluctuations modulated by the gate bias.[24–26] Since the tubes studied in this work are metallic in nature with little or no gate modulation, the origin of low-frequency noise in this case can be assumed to be caused by mobility fluctuations. We now develop a model based on the Hooge's mobility fluctuation approach to gain a more quantitative understanding of the origin of noise in these systems. One can use Boltzmann transport theory[17] to calculate the current in metallic SWNTs at high bias. At high bias, the current can be approximated as $I = Bl_{\text{eff}}/L$, where B is a constant, L is the length of the CNT, and l_{eff} is the effective mean free path. It is well known that low-frequency noise in SWNTs originate from mobility fluctuations.[22,24] In this case, the modulation of the effective mean free path (mfp) due to OP scattering translates into mobility fluctuations of the device. By Mathissen's rule, the effective mfp is given by $l_{\text{eff}}^{-1} = l_e^{-1} + l_{\text{hp}}^{-1}$, where l_e is the elastic scattering mfp and l_{hp} is the mfp of backscattering phonons. Therefore, the only possibility is that the noise at high bias for SWNTs originates due to slow fluctuations in l_{hp}, caused by scattering with OPs. This modulation of path lengths in turn translates into mobility fluctuations of the device. The fluctuations in current I is then given by $\delta I = B\delta l_{\text{eff}}/L$. At a given instant if the SWNT has N electrons with mean free path lengths $l_i (i = 1,\ldots,N)$ then an effective mfp is given by $l_{\text{eff}} = (\sum l_i)/N$. These slow fluctuations in the path lengths δl_i would naturally be related to the scattering rate of the electrons with the OPs. The relative noise power spectra A_I is then given by

$$S_I = B^2 \frac{\left(\sum S_{l_i}\right)}{L^2 N^2}, \quad A_I = \left[\frac{S_{l_{\text{hp}}}}{l_{hp}^{2+\gamma_o}}\right] \frac{1}{N} \tag{4}$$

as $(\sum S_{l_i})/N = S_{l_{\text{hp}}}$ and $S_I \propto I^{2+\gamma_o}$.

Due to the presence of a component of l_{hp} term in each l_i, the low-frequency fluctuations will have a signature from OP scattering which would be proportional to the scattering rate of the electrons with the OPs even though OP scattering takes place in the time scale of picoseconds. Using Fermi's "golden rule" and considering only emission processes, the decay time for an electron in state $\mathbf{k}$, band l, and energy $\epsilon_{\mathbf{k}l}$ to another

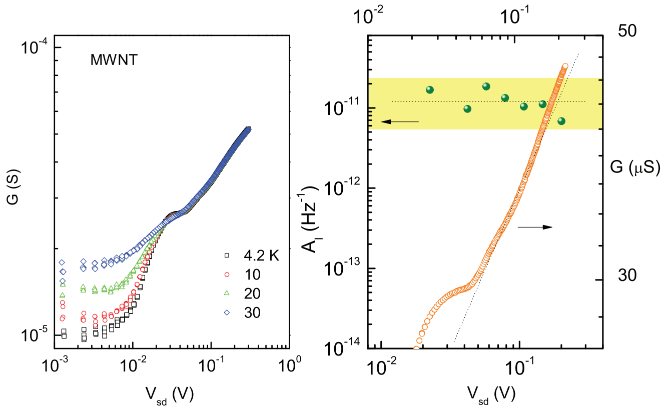

FIG. 4. (Color online) (a) Conductance G of a MWNT as a function of source-drain voltage V_{sd} at different temperatures. A power law above $V_{ds} = 10$ mV is evident at 4.2 K. (b) Relative noise A_I vs source-drain bias V_{sd} at 4.2 K. The bias independence of the relative noise can be observed in the power-law regime which is indicated by the yellow band.

electronic band l' with energy $\epsilon_{(\mathbf{k}+\mathbf{q})l'}$ is given by[27,28]

$$\frac{1}{\tau} = \sum_{\eta} \frac{\pi}{M\omega_{\mathbf{q}\eta}} |D_{(\mathbf{k}+\mathbf{q})l',\mathbf{k}l}|^2 \rho[\epsilon_{(\mathbf{k}+\mathbf{q})l'}](n_{-\mathbf{q}\eta}+1). \qquad (5)$$

Here $\mathbf{q}$ is the phonon wave vector in branch η with energy $\hbar\omega_{\mathbf{q}\eta}$, $|D|$ is the electron-phonon-coupling (EPC) strength, ρ is the density of states, and n is the phonon occupation factor given by $n_{\mathbf{q}\eta} = [\exp(\hbar\omega_{\mathbf{q}\eta}/k_B T) - 1]^{-1}$. For $eV_{ds} \geqslant \hbar\omega_q$, where ω_q is the optical phonon frequency, a key role is played by phonon occupation number in limiting the high-field transport as well as low-frequency noise in nanotubes. In this limit, assuming the energies are shifted such as $E \to eV_{ds} + E_F$ by applied bias V_{ds}, the scattering rate becomes[21] $\tau(V_{ds})^{-1} \propto \exp{-[(eV_{ds} + E_F) - \hbar\omega_q]/k_b T}$. We find that the noise can be fit remarkably well by the simple exponential function of V_{ds} beyond the power-law region as shown in Fig. 2. Physically, the low-frequency noise in metallic CNTs caused by mobility fluctuations is related to the OP scattering rate through the phonon occupation number.

C. MWNTs

As the clean power-law regime is not sufficiently extended in SWNTs due to the intervention of optical phonons we perform further noise measurements in MWNTs which have a cleaner and much larger power-law regime than SWNTs. In MWNTs the power-law regime extends up to a much higher source drain bias V_{ds} due to their ability to carry much higher currents. In Fig. 4(a) the power-law dependence of conductance on drain-source voltage V_{ds} at different temperatures in an MWNT device is shown. Note that the power law spans the entire measurement range (more than a decade) and there is no conduction saturation due to optical-phonon scattering as the threshold for OP scattering is at a much higher bias in MWNTs compared to SWNTs.[29–31] The origin of the unusual kink in conductance seen in the figure is also not clear and might be caused by some disorder in the tube. The relative noise (A_I) in the power-law region for the MWNT is shown in Fig. 4(b). In agreement with Eq. (3) we observe that the relative noise (A_I) remains constant with voltage (V_{ds}) in the power-law regime. The obtained γ_0 for the MWNT is 0.56 in this case. Another interesting point is that for MWNTs the PSD of noise also shows a $1/f$ character in the entire power-law regime with not a single Lorentzian spectra. Measurements in MWNTs also once again clearly show that noise in a power-law regime behaves similarly to that in linear regime.

IV. CONCLUSIONS

We have presented a framework for studying current fluctuations in the non-Ohmic regime. This study was demonstrated by measuring current noise in the power-law region and beyond in SWCNTs and MWCNTs. It is shown that the generalized relative noise in this highly nonlinear regime remains independent of bias similar to the linear regime. As a utility of this study we show that, by measuring noise beyond the power-law regime, mobility fluctuations of charge carriers in the tube caused by scattering with OPs is the dominant source of noise in CNTs in this regime. We propose a mobility fluctuation model to relate mobility fluctuations to the phonon occupation number. Our experimental results have widespread ramifications on noise characterization for high bias interconnect applications of nanoelectronic devices which operate mostly in the nonlinear regime.

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