Local and non-local shot noise in multiwalled carbon nanotubes

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Abstract - We have investigated shot noise in multiterminal, disordered multiwalled carbon nanotubes (MWNTs) at 4.2 K over the frequency \( f = 600–850 \text{MHz} \). Quantitative comparison of our data to semiclassical theory, based on non-equilibrium distribution functions, indicates that a major part of the noise is caused by a non-equilibrium state imposed by the contacts. Our data exhibits non-local shot noise across weakly transmitting contacts while a low-impedance contact eliminates such noise almost fully. Within our model we obtain \( F_{\text{tube}} < 0.1 \) for the intrinsic Fano factor of our MWNTs. This behavior cannot be explained by the presence of ballistic channels.

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Multiwalled carbon nanotubes (MWNTs) are miniscule systems, their diameter being only a few nanometers. Yet, in surprisingly many cases their transport properties can be described neglecting quantum coherence, interference effects showing up only through weak localization [1,2]. This is in contrast to single-walled tubes, where interference effects dominate, and give rise for example to Fabry-Perot resonances with distinctive features in conductance and current noise [3].

When interference effects are washed out, a semiclassical analysis based on non-equilibrium distribution functions is adequate, and the circuit theory of noise becomes a powerful tool in considering nanoscale objects [4,5]. This theory makes it straightforward to calculate the current noise of incoherent dots and wires, and to relate the current noise to the transmission properties of the corresponding section of the mesoscopic object. Semiclassical analysis of multiprobe samples provides a way to make a distinction between sample and contact effects — analogous to the analysis of the four-probe conductance — and thereby it allows one to investigate contact phenomena, of which only a little is known in carbon nanotube systems.

We have investigated the influence of contacts on the shot noise in multiterminal, disordered carbon nanotubes. We have made four-lead measurements on MWNTs in which two middle probes have been employed for noise measurements. We show that quantitative information can be obtained from such measurements using semiclassical circuit theory in the analysis. We find that probes with contact resistance \( R_{C} < 1 \text{kΩ} \) relax electrons strongly toward a local equilibrium, resulting in classical addition of noise of two adjacent sections, while “bad” contacts \( (R_{C} \sim 10 \text{kΩ}) \) act as weakly perturbing probes which need to be analysed on the same footing as the other parts of the sample. We also find that good contacts eliminate noise that couples to the probe from a non-neighboring voltage biased section. In addition, we find from our analysis that the tubes themselves are quite noise-free, with a Fano-factor \( F_{\text{tube}} < 0.1 \). As far as we know, our results are the first shot noise measurements addressing the contact issues in carbon nanotubes.

To clarify the results of our multi-probe noise measurements, let us consider the three-terminal structure depicted schematically in fig. 1. Assume that the average current \( \langle I \rangle \) flows between 1 and 2, and the average

Fig. 1: (a) 3-probe structure, describing two nanotube sections with resistances and Fano factors \( R_{1}, F_{1}, R_{2}, F_{2} \) and a contact \( (R_{3}, F_{3}) \). These are linked together in the circuit model by a node, shown as a circle. The noise power \( S_{i} \) at terminals \( i = 1, 2, 3 \) can be calculated for different current biases. (b) Extended contact model.
potentials of the terminal 3 adjusts to the potential of the node. In our work, terminal 3 is disconnected from the ground at low frequencies, but at the high frequencies of the noise measurement, the impedance to the ground is much lower than that of the contacts. As a result, the effect of voltage fluctuations in the third terminal on the overall noise can be neglected. We describe two kinds of noise measurements: “local”, where the noise is measured from one of the terminals 1 or 2, and “nonlocal”, where the noise is measured from terminal 3. The shot noise can thus be characterized by the local and non-local Fano factors, defined as $F_{il} = S_{il}/e(I)$, $i = 1, 2$, and $F_{nl} = S_{nl}/e(I)$. Here, $S_i = \int dt \langle \delta I_i(0) \delta I_i(t) \rangle$ is the low-frequency current noise measured in terminal $i$.

In classical circuit theory, the only essential components in the analysis are the fluctuating potentials in the nodes. This theory is valid in the case of strong inelastic scattering inside the node. In this case, the expressions for the Fano factors are

$$F_{11} = \frac{(G_2 + G_3)^2}{G_1^2} F_1 + \frac{G_3^2}{G_1^2} F_2, \quad F_{nl} = \frac{G_3^2}{G_1^2} (F_1 + F_2), \quad (1)$$

where $G_i = G_1 + G_2 + G_3$. If the nonlocal terminal 3 is well connected to the node, $G_3 \gg G_1, G_2$, the local noise measurements find only the local Fano factor, $F_{11} = F_1$ and $F_{22} = F_2$, whereas the nonlocal noise is the sum of the terminal 3, $F_{nl} = F_1 + F_2$. This is because in this limit the terminal 3 suppresses the voltage fluctuations from the node, and the resulting noise is only due to the contacts. In the opposite limit $G_3 \rightarrow 0$, the nonlocal noise vanishes, and the local noise is given by the classical addition of voltage fluctuations, $F_{11} = F_{22} = (G_2 F_1 + G_3 F_2)/G_1^2$.

At low temperatures the inelastic scattering inside the nanotubes is suppressed. In this case, assuming that the momentum of the electrons inside the node is isotropized [5,6], the noise can be calculated with the semiclassical Langevin approach [4,6]. It considers, instead of the node potential, the full electron energy distribution function inside each node as a fluctuating quantity $f(E) = \langle f(E) \rangle + \delta f(E)$. Inside the terminals, the distribution function is assumed not to fluctuate, because of relaxation and strong coupling to the ground. The fluctuations $\delta f(E)$ are induced by intrinsic fluctuations of currents between the nodes $I_{ij}(E) = G_{ij} [f_i(E) - f_j(E)] + \delta I_{ij}(E)$. Properties of $\delta I_{ij}$ are known from scattering theory, which allows calculating all noise correlators in the circuit [6].

The fluctuation-averaged energy distribution $f_{\text{av}}(E)$ at the node is given by a weighted average of (Fermi) distributions at the terminals, $f_{\text{av}}(E) = \sum G_j f_j(E)/G_i$. In the general case, the resulting expressions for Fano factors $F_{11}, F_{22}, F_{nl}$ are lengthy, but in the case of the strongly coupled terminal 3, $G_3 \gg G_1, G_2$, one obtains the same expressions as in the classical case. This is because in this limit the distribution function of the node is given by the Fermi function of terminal 3. In the opposite limit $G_3 \ll G_1, G_2$ the nonlocal noise vanishes and the local noise is given by the semiclassical sum rule,

$$F_{11} = F_{22} = \frac{G_1^2 F_1 + G_2^2 F_2 + G_1 G_2 (G_1 + G_2)}{(G_1 + G_2)^2}. \quad (2)$$

This sum rule applies for any pair of neighboring nodes of an arbitrary one-dimensional chain of junctions, assuming that inelastic scattering can be neglected. Apart from the different power of the conductances compared to eq. (1), it contains an additional term, independent of the Fano factors of the contacts. This term is induced by the nonequilibrium electron distribution in the node. In the limit of a long chain with many nodes, applying the rule (2) repeatedly makes the Fano factor approach the universal value $F = 1/3$ characteristic for a diffusive wire [7].

When comparing the semiclassical model to the experiments, we ignore the resistance of the wires under the contacts and use a model of localized contacts. In practice, the large width and low interfacial resistance of the contacts makes the current flow through them non-uniform. Including this fact in the theoretical model by describing the tube using a large number of nodes on top of the contacts (see fig. 1(b)) did not improve the fit to the experimental noise data, whereas some improvement was obtained in the fit to the resistance data. This means that a situation between the localized and distributed contacts is realized — however including this fact in the model would increase the number of fitting parameters.

Our individual nanotube samples S1 and S2 were made out of a plasma-enhanced CVD MWNTs [8] with length $L = 2.6$ and $5 \mu m$ and diameter $d = 8.9$ nm and 4.0 nm, respectively. The main parameters of the samples are given in table 1. The contacts on the PECVD tubes were made using standard e-beam overlay lithography. In these contacts, 2 nm of Ti was employed as an adhesive layer before depositing 30 nm of gold. The widths of the four contacts were $L_{1C} = 400$ nm and $L_{2C} = 550$ nm for samples 1 and 2, respectively. The strongly doped Si substrate was employed as the back gate ($C_v \sim 5 aF$), separated from the sample by 150 nm of SiO$_2$. The charge neutrality point for similarly prepared semiconducting tubes is usually at $V_g = -2$ V. Using the diameter dependence of the transverse mode energy and the gate capacitance, we may estimate that, at $V_g = 0$, the elastic mean free path $\ell_m = 30–60$ nm and $\ell_d = 50–110$ nm for S1 and S2, respectively.

Our measurement setup is illustrated in fig. 2. Bias-tees are used to separate dc bias and the bias-dependent noise signal at microwave frequencies. We use a LHe-cooled, low-noise amplifier [9] with an operating frequency range of $f = 600–950$ MHz. A microwave switch and a high-impedance tunnel junction were used to calibrate the gain. Our setup measures voltage fluctuations with respect to ground at the node next to the contact terminal; these are converted to current fluctuations by the contact resistance of the measuring terminal. The sample was biased using
Table 1: Main parameters of our samples S1 (upper part) and S2 (lower part). The diameter is given by \( \phi \) and the lengths of the tube sections are denoted \( L_{ij} \) (excluding the range of contacts), where the indices \( ij \) correspond to pairs of terminals (see fig. 2) 12, 23, and 34. Resistances of the individual sections \( R_{ij} \) at \( V = 0 \) were measured using bias \( V = 0.1-0.2 \, \text{V} \) in order to avoid zero-bias anomalies; \( R_{C2} \) indicates the 4-wire resistance \( R_{4p} \). Contact resistances \( R_{Ck} \), weakly dependent on \( V \) and bias voltage polarity, are given at \( V_{g} = 0 \) and \( V > 0 \); index \( k \) identifies the contact. All the resistance data refer to \( T = 4.2 \, \text{K} \). The lower row values correspond to resistance values obtained in fitting of the semiclassical model. For details, see text.

| \( \phi \) (nm) | \( L_{12} \) (nm) | \( L_{23} \) (nm) | \( L_{34} \) (nm) | \( R_{12} \) (kΩ) | \( R_{23} \) (kΩ) | \( R_{34} \) (kΩ) | \( R_{67} \) (kΩ) | \( R_{C1} \) (kΩ) | \( R_{C2} \) (kΩ) | \( R_{C3} \) (kΩ) | \( R_{C4} \) (kΩ) |
|----------------|-----------------|-----------------|-----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|----------------|
| S1 exp.        | 8.9             | 430             | 300             | 540            | 35             | 30             | 34             | 17.5           | -              | 0.5            | 12             | -              |
| S1 fit         |                 |                 |                 |                | 27             | 30             | 41             | 17             | 2.4            | 0.1            | 9.7            | 0              |
| S2 exp.        | 4.0             | 940             | 440             | 1110           | 21             | 25             | 28             | 16.5           | 5              | 1.7            | -              | 0              |
| S2 fit         |                 |                 |                 |                | 27             | 16             | 31             | 12             | 0              | 1.5            | 1.3            | 0.8            |

Fig. 2: Schematics of our high-frequency setup. Indices 5–8 refer to nodes with different distribution functions on the nanotube. Contacts are drawn as tunnel junctions with resistances \( R_{ij} \); numbers 1–4 represent the measurement terminals. A sum of lead and bonding pad capacitance is given by \( C_p \sim 1 \, \text{pF} \) while the inductors represent bond wires of \( L_s \sim 10 \, \text{nH} \). TJ denotes a tunnel junction for noise calibration.

Fig. 3: (Color online) a) Current noise power (arbitrary units) measured from lead 2 in sample 2 at \( V_g = 0 \) as a function of bias current, (see text for labeling). The circles are the measured data while the solid lines are theoretical fits to direction-averaged data over \( 0.1 < |I| < 2 \, \mu\text{A} \). The theoretical line \( S_{1,13} \) corresponds to \( F = 0.30 \). b) The noise measured from terminal 3 — presentation details as above. \( S_{2,24} \) line corresponds to \( F = 0.29 \). The insets illustrate the differential contact resistances determined as \( R_{C2} = (R_{12} + R_{23} - R_{13})/2 \) and \( R_{C3} = (R_{23} + R_{34} - R_{24})/2 \).

One voltage source, one lead connected to the (virtual) ground of the input of the DL1211 current preamplifier and with the remaining two terminals floating.

From four-point measurements at 0.1–0.2 V, we get \( R_{4p} = 17.5 \, \text{kΩ} \) and \( R_{4p} = 16.5 \, \text{kΩ} \) (section 6–7 in fig. 2) for samples S1 and S2, respectively. Assuming constant conductivity, this yields for the resistance per unit length \( r_l = 37–58 \, \text{kΩ}/\mu\text{m} \), which amounts to \( 20 \, \text{kΩ} \) over the length of a contact. The contact resistances for contacts 2 and 3 of S1 and S2 were determined as averages from a set of two-lead measurements: \( R_{C2} = (R_{12} + R_{23} - R_{13})/2 \) and \( R_{C3} = (R_{23} + R_{34} - R_{24})/2 \); this scheme was adopted as there is a non-local contribution in voltage [10]. \( R_{C2} \) and \( R_{C3} \) were found to be nearly constant except for a small region near zero bias. Variation of \( V_g = -4 \ldots +4 \, \text{V} \) changes \( R_{C2} \) from \( 4 \) to \( 6 \, \text{kΩ} \) and \( R_{C3} \) from \( 1 \) to \( 3 \, \text{kΩ} \) on S2 on average. In both cases, \( R_{C2} \) increases when going from \( V < 0 \) to \( V > 0 \) (see table 3). We cannot determine the contact resistance of contacts 1 and 4 from the resistance data, only the sum of the contact and the nanotube section. For sample 1, we find \( R_{C2} = 0.1-1 \, \text{kΩ} \), indicating an excellent contact, while \( R_{C3} = 12 \, \text{kΩ} \) points to a weak, tunneling contact.

Figure 3 shows the measured noise as a function of current for sample S2. We measured the current noise
Table 2: Fano factors measured at $0.1 < |I| < 2 \mu A$ for sample S1 at terminals $k = 2$ and 3, for the two different directions of current. The values in column “Fit” have been calculated using semiclassical circuit theory (see text), and the values in column “Dev” show the deviation between the semiclassical theory and averaged experimental Fano factor. “Fit (cl.)” shows the best fit to classical circuit theory, which is achieved at a high contact resistance $R_{C1} \sim 22 k\Omega$ and low contact and high tube Fano factors $F \sim 0$, $F_{\text{tube}} \sim 1$.

| Terminal 2 | Terminal 3 |
|------------|------------|
| $I < 0$ | $I > 0$ | $I < 0$ | $I > 0$ | $V_g = +4V$ | $V_g = 0$ | $V_g = -4V$ |
| $S_{k,12}$ | 0.11 | 0.080 | 0.11 | 0.14 | 0.005 | 0.005 | 0.006 | 0.002 |
| $S_{k,13}$ | 0.48 | 0.46 | 0.39 | 0.46 | 0.41 | 0.39 | 0.38 | 0.5 | 0.23 |
| $S_{k,14}$ | 0.51 | 0.50 | 0.46 | 0.52 | 0.50 | 0.46 | 0.45 | 0.7 | 0.57 |
| $S_{k,23}$ | 0.41 | 0.40 | 0.29 | 0.32 | 0.42 | 0.38 | 0.38 | 0.6 | 0.23 |
| $S_{k,24}$ | 0.43 | 0.42 | 0.35 | 0.37 | 0.54 | 0.47 | 0.45 | 0.12 | 0.56 |
| $S_{k,34}$ | 0.24 | 0.24 | 0.30 | 0.26 | 0.46 | 0.50 | 0.40 | 0.18 | 0.34 |

Table 3: Summary of Fano factors measured at $0.1 < |I| < 2 \mu A$ for S2 at three different gate voltage values as well as the measured contact resistances for sample 2 at terminals 2 (top) and 3 (bottom). Columns “Fit” and “Dev” refer to theoretical fit using semiclassical analysis and its deviation from the experimental data as in table 2.

| $V_g = +4V$ | $V_g = 0$ | $V_g = -4V$ |
|--------------|--------------|--------------|
| $R_{C2}$ ($k\Omega$) | 5.8 | 6.4 | 5.3 | 4.9 | 1.5 | 70 | 3.9 | 5.1 |
| $S_{2,12}$ | 0.22 | 0.26 | 0.20 | 0.22 | 0.15 | 29 | 0.21 | 0.20 |
| $S_{2,13}$ | 0.31 | 0.30 | 0.21 | 0.22 | 0.30 | 39 | 0.34 | 0.31 |
| $S_{2,14}$ | 0.35 | 0.35 | 0.42 | 0.38 | 0.35 | 11 | 0.34 | 0.31 |
| $S_{2,23}$ | 0.23 | 0.23 | 0.22 | 0.21 | 0.20 | 9 | 0.25 | 0.26 |
| $S_{2,24}$ | 0.31 | 0.28 | 0.26 | 0.27 | 0.25 | 5 | 0.26 | 0.27 |
| $S_{2,34}$ | 0.080 | 0.11 | 0.040 | 0.075 | 0.09 | 57 | 0.055 | 0.045 |

$S_{2}$ using both DC current, $S_{2} = S(I) - S(0)$, and AC modulation on top of DC bias: $S_{2} = \int_{0}^{I}(dQ/dI) dI$, where $S$ represents the noise power integrated over the 250 MHz bandwidth (divided by 50 $\Omega$) and $(dQ/dI)$ denotes the differentially measured noise. As seen in fig. 3, the measured noise for each section of the tube is quite well linear with current up to 1–2 $\mu A$, while at larger currents the Fano factor decreases gradually with $I$. We determine the Fano factor using linear fits to $S_{2}$ in the range 0.1–2 $\mu A$: the results vary over 0.1–0.5 as seen in table 2 and table 3 for S1 and S2, respectively.

The basic finding of our measurement is that the noise of the sections adjacent to probes 2 and 3 may behave quite differently, depending on how strong the contact is between the gold lead and the nanotube. For terminal 2 of S1, the noise adds in a classical fashion as expected for a good contact, i.e., $S_{2,13} \approx S_{2,12} + S_{2,23}$ (see table 2). Here, the first index indicates that noise is measured from contact 2 while the bias is applied to the terminal specified by the middle index and the last index tells the grounded contact. For the terminal 3 of S1, on the other hand, we find that $S_{3,24} \approx S_{3,23} \approx S_{3,34}$. In sample S2, the results are intermediate to the above extreme cases (for example, $S_{3,23}, S_{3,34} < S_{3,24}$ and $S_{3,23} + S_{3,34} > S_{3,24}$). All the above basic relations are in accordance with semiclassical analysis, whereas only the results related to good contacts can be accounted for by purely classical circuit theory (cf. eq. (1)).

We fit the semiclassical theory to our data with six fitting parameters: the tube resistance per unit length $r_1$, assumed uniform, the interfacial resistances $R_{Ck}$ in the contact regions, and the Fano factor $F_{\text{tube}}$ of the tube sections. For contacts we use $F = 1$ as it makes only a small contribution to noise in good-quality contacts. The model thus contains 6 adjustable quantities, and can be used to predict the measured six resistances and the 12 noise correlators. These are fit through a least-square minimization procedure.
Table 2 and table 3 display the calculated results for the noise correlators, and the corresponding resistances are shown in table 1. The overall agreement of calculated noise with the measurement is 10–35%, excluding the smallest non-local noise value. Within the error bars for the measured data (uncertainty is mainly due to the difficulties in determining the exact linear-response resistance values, see below), we obtain an upper limit $F_{\text{tube}} \leq 0.1$. For values larger than this, $S_{2,12}$, $S_{2,13}$, and $S_{2,14}$, which are most sensitive to it, are larger than measured by more than 15%, and also the overall quality of the fit decreases. The smallness of $F_{\text{tube}}$ means that most of the noise comes from the non-equilibrium state generated by the current, as in the last term in eq. (2), not from the transport in the tube itself 1.

For comparison, the classical circuit theory is less successful in explaining the results (see table 2), as the noise sum rule restricts $S_{3,23}$, $S_{3,34}$, and $S_{3,24}$ so that at least one of them is $\sim 30\%$ away from the measured value, and the same applies to $S_{5,13}$, $S_{5,34}$, and $S_{5,14}$. Moreover, the complete relaxation assumed in the model significantly reduces the predicted non-local noises $S_{2,34}$ and $S_{3,12}$. The Fano factors associated with the good contact $C2$, however, agree better with the data as expected.

Within our model, there are two possible ways to explain the low tube Fano factor: a) transport is dominated by close-to-ballistic channels or b) there is an extra energy-absorbing inelastic-scattering mechanism. In our samples, the measured resistances typically exceed the value $h/(2e^2) = 12.9$ k$\Omega$ that would be obtained if the tube contained only a single open spin-degenerate channel with unit transmission. Moreover, the resistances only weakly depend on the bias current above the Coulomb blockade threshold, even though one would expect opening of new channels at a high bias. These properties indicate a diffusive rather than ballistic behavior, and hence we can dismiss the mechanism a). A similar low Fano factor was measured in SWNT bundles [11], but there the result could be explained by the presence of ballistic channels. The case b) would require an inelastic-scattering mechanism with a voltage- and temperature-independent scattering length $\ell_{\text{in}}$ that is of the order of the tube length. However, the simplest approach of including such a mechanism in our semiclassical circuit model in a relaxation time approximation and assuming $F_{\text{tube}} = 1/3$ resulted in poorer fits than those described above. We think such inelastic scattering cannot be due to the electron-phonon interaction, which should be relatively weak in nanotubes [12]. An alternative relaxation process, consistent with energy-independent $\ell_{\text{in}}$, could be due to the tube radiating energy into the surroundings [13]. We aim to investigate this mechanism in more detail in the future.

The fitted resistances are slightly off from the measured values. This is partly because in nanotubes it is difficult to avoid uncertainties in four-probe measurements (current goes in part through the voltage probes), and partly because of the presence of non-local voltages. In addition, $\ell_d$ in our MWN Ts may be so long that our analysis is not strictly valid any more. For example, in section 1–2 of S1, the IV curve displays a power law which clearly differs from the rest of the sample. The fitted contact resistances range over $R_C = 0.1–10$ k$\Omega$, which is in agreement with typical measured values [1,14].

In summary, we have investigated experimentally shot noise of multiterminal MWNTs under several biasing conditions. The noise was found to depend strongly on the contact resistance. At small interfacial resistance, our 0.4–0.5 micron contacts acted as inelastic probes and the classical noise analysis was found sufficient. Weaker contacts could be accounted for by using semiclassical theory. The latter allows to comprehend the observed, broad spectrum of Fano factors, but it leads to the conclusion that the intrinsic noise of MWNTs is nearly zero, $F_{\text{tube}} < 0.1$. Most of the observed noise is generated by metal-nanotube contacts, which govern the non-equilibrium distributions of charge carriers on the tube. The low $F_{\text{tube}}$ may reflect the presence of an inelastic-scattering mechanism with a low scattering length, or may arise from interaction effects not present in our model.

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