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1/f noise and mechanisms of the conductivity in carbon nanotube bundles

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Abstract: Experimental results are reported of the investigation of conductivity mechanisms in metallic single-wall carbon nanotube (SWCNT) bundles in a wide temperature range from 4.2 K to 300 K. The temperature dependence of the resistance and noise parameters – the logarithmic slope of the current dependence of noise as well as the normalized current noise – are compared. Remarkable changes in noise characteristics are registered at temperatures typical of the transition from hopping conductivity to Luttinger liquid conductivity and the transition from Luttinger liquid conductivity to diffusion conductivity. In the first transition region, the slope of the normalized noise level of the current changes significantly as a function of temperature. In the region of diffusion conductivity, a stronger variation of the normalized noise level is revealed. These changes in noise properties are correlated with changes in the transport characteristics of SWCNT bundles that allow us to adequately explain the mechanisms of conductivity in the system.

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1. Introduction

Recently, carbon nanotubes (CNTs) have attracted considerable attention due to their remarkable properties, which are very promising for the development of a new generation of molecular nanoelectronics. Depending on the type of conductivity, carbon nanotubes can be divided into semiconductor or metallic. CNTs with a nanoscale diameter and a few micrometers in length demonstrate unique physical phenomena related to the ideal one-dimensional (1D) systems. Among these phenomena, the most outstanding effect is massless charge transport phenomena in metallic nanotubes. Such phenomena were predicted theoretically more than 50 years ago by Tomanaga [1] and Lüttinger [2]. It was shown the bosonization of carrier excitation spectra in the vicinity of the Fermi level is responsible for charge transport by waves. This behavior contradicts the well-known charge transport by particles, electrons or holes in systems of larger dimensions. The Tomanaga–Lüttinger concept has been justified by numerous studies and remains a hot topic for theoretical and experimental study. Metallic single-wall carbon nanotubes (SWCNTs) demonstrate ballistic charge transport even at room temperature on the scale of a few micrometers. These properties of CNTs are attracting considerable attention from the point of view of their applications as ideal molecular conductors in nanoelectronic devices. In this respect, the study of current noise in carbon nanotubes is extremely important. In a large number of studies, the $1/f$ noise level in molecular transistors based on semiconducting CNTs was found to be high. At the same time, in nanoobjects even properties of contact systems can differ considerably from those in bulk materials. Therefore studies of noise properties may give useful insights into nanodevice characteristics. Additionally, they can be used to study transport phenomena and to reveal features that are not accessible using conventional methods. It should be noted that mesoscopic systems often demonstrate unusual properties, which differ considerably from the microscopic and the macroscopic systems. On the one hand, the crystal structure is different compared with the bulk crystal, because often the structure has an extensive surface, as in the case of 0D nanoparticle or one-dimensional systems: graphene, nanotubes, fullerenes. On the other hand, the sizes of these structures are so small that they have a collective quantum level. The electronic properties of the structures differ from those of bulk materials, and from the properties of individual atoms. Nanotubes are one of the most outstanding and most intensively studied objects of mesoscopic systems, especially carbon single- and multiwall nanotubes [3].

The mechanical, the chemical, and, more recently, the electrical properties of carbon nanotubes are being actively studied. The charge carriers in nanotubes demonstrate uniquely high mobility which is promising for the development of novel field-effect transistors. A single CNT or CNTs grown in the structure of arrays of parallel nanotubes can be used in order to fabricate these devices. Studies of the electrical and noise characteristics also focus on the systems. Such interest in carbon nanotubes is mainly due to the fact that the CNTs have almost ideal structures of surface atoms with the one-dimensional character of the conductivity.

Ultra-high values of mobility of nanotubes are determined by the specific character of the electron gas formation. In nanotubes with metallic conductivity, due to the closed surface, standing electron waves are formed in the section of the nanotube between defects, thus violating the ideality of the graphene lattice. Conductivity in this case is determined by the resistance (equal to the quantum resistance) of these regions and the tunneling of electrons between adjacent regions. Tunneling is possible between adjacent segments of one nanotube as well as between adjacent nanotubes. Such conductivity was theoretically justified by Lüttinger, therefore the electron gas is called the “Lüttinger liquid” [4–7].

Conductivity determined by the “Lüttinger liquid” can be found in a limited temperature range. From the low temperatures, this range is limited by the transition point from the degenerate state of the electron gas (metallic conductivity) to a non-degenerate state with the hopping conductivity. The temperature of this transition, estimated theoretically, is around $T = 10\text{--}20\text{ K}$. At high temperatures (at $T > 200\text{ K}$), according to the theoretical predictions, the quantized electronic states are destroyed, resulting in a classical behavior of electron gas and the conductivity is of a normal diffusive nature [8].

Determination of the conductivity type in different temperature ranges is important not only from a practical point of view, but also in order to validate the theoretical predictions and assumptions. In addition, establishment of the characteristic temperatures of the transition between different types of conductivity in the samples after their modification or functionalization may facilitate an understanding of the impact of such effects on properties of a single nanotube or CNT arrays.

In this paper, the spectra of low-frequency noise measured for the region corresponding to the initial quasi-linear current–voltage characteristics (i.e. in equilibrium state) of the SWCNT bundles are studied. Specific features of the noise characteristics in different temperature ranges are revealed. The results analyzed together with transport properties of the CNT samples. Theoretical consideration of the different types of conductivity, typical for different temperature ranges, allows us to elucidate the transport mechanisms in the system.

2. Experimental details

We studied the phenomena of charge transport in structures consisting of bundles of metallic carbon nanotubes prepared by applying uniaxial pressure. The source powder was compressed at room temperature and a pressure of 1 GPa. The initial powder contained 90% single-walled carbon nanotubes. According to the results of Ref. [9], the structures formed at a pressure represented by bundles, mainly oriented in the plane perpendicular to the axis of the applied pressure. Nanotubes bind together by weak van der Waals forces. The lengths of the bundles of nanotubes were in the range of 5–30 μm . After pressing the powder, the structure of the sample represents a system of multiple twisted bundles, which form a set of overlapping contacts, as can be seen from a fragment of the structure shown in Fig. 1a. Images were obtained using high-resolution electron microscopy. The material structure of the sample displays the twisted and overlapped strands of carbon nanotubes. Nanotubes in the individual bundles are parallel to each other.

A Raman spectrum (Fig. 1b) shows the main peak at 1602 cm^{-1} due to the covalent bonds between carbon atoms, and two peaks at 137 cm^{-1} and 235 cm^{-1} , which correspond to the breathing modes of single nanotubes with a diameter of 1.02–1.05 nm and 1.68–1.73 nm, respectively. The data demonstrate that the bundles mainly consist of single-walled nanotubes of two diameters. As will be shown below, the conductivity of the structures was of a metallic character with characteristic features of a one-dimensional metal system.

Low-frequency noise and transport properties of the structures were studied for samples in the two-terminal configuration. Samples were prepared in the form of dumbbells, containing a constriction with a characteristic size of $100\cdot 100\text{ }\mu\text{m}^2$ in a cross-sectional area and a length of about 200 μm . The configuration of the sample is similar to one described in Ref. [8].

Electrical contacts were prepared using a conductive paste of silver powder. Such contacts were ohmic over the entire temperature range of 4.2–300 K and remained stable during repeated processes of cooling and heating of the samples under study [8,10].

In a wide temperature range, the conductivity of the structures was investigated by applying a constant voltage. The resistance of the samples changes over the whole temperature range from 7 Ω to 100 Ω . These resistance values allow us to measure the current–voltage and noise characteristics with high accuracy. Electrical measurements were performed at voltages not exceeding 100 mV at a power dissipation of less than $3 \cdot 10^{-5}$ W over the entire temperature range. This allowed us to avoid the influence of self-heating effects on the results obtained. In the voltage range investigated, the deviation of current–voltage characteristics from linear behavior was negligibly small. The noise spectra were measured using a home-made Fourier spectrum analyzer in the frequency range 1 Hz–100 kHz and a home-made low-noise preamplifier. Measurements were performed in the constant current regime, which was realized by choosing the load resistance value at least 10 times higher than the resistance of the sample. Relatively low resistance of the sample required the use of a low-noise preamplifier with its own equivalent noise resistance, which is in the same order of magnitude as the resistance of the sample. In this case, it is possible to measure very low-noise characteristics, comparable to the thermal noise of the sample. We used the amplifier with the equivalent noise resistance at a frequency $f = 1$ kHz of $R_{\text{noise}} = S_{V0}/4kT = 50 \Omega$, where S_{V0} is the spectral density of the noise voltage amplifier at the resistance of the signal source equal to zero, k is the Boltzmann constant and T is the temperature.

3. Results and discussion

The temperature dependence of the sample resistance is shown in Fig. 2. The data demonstrate three regions with different temperature behavior. The experimental data (Fig. 2a, open circles) are compared with those characteristic for Luttinger liquid, a temperature dependence of resistance $R \propto T^\alpha$ with exponent $\alpha = 0.4$. In the temperature range from 20 K to 200 K the measured dependence corresponds to the conductivity of a Luttinger liquid and is in good agreement with literature results [8,10]. Such behavior, characteristic for Luttinger liquids, was previously also reported for bulk-contacted CNTs [6]. At temperatures higher than 200 K, the measured dependence deviates from this simple function. At lower

temperatures, $T \approx 4.2\text{--}20$ K, the resistance follows an exponential function $R \propto \exp T_0/T^{0.25}$ (Fig. 2b). An exponential dependence of the resistance versus the sample temperature with an exponent of 0.25 indicates the hopping mechanism of conductivity with variable hopping length.

The noise spectra of the sample have a $1/f^\gamma$ dependence with $\gamma \approx 1$ (Fig. 3), which is a characteristic function of flicker noise. Such kind of noise is typical for the case of conductivity in disordered structures. Pressed nanotubes constituting the sample studied are just one example of such a structure. Moreover, the deviations from the $1/f$ spectrum in disordered structures have to be regarded as an anomaly and are characterized from the point of view of the sample-specific features in conductivity under certain measurement conditions.

The shape of the noise spectra shown in Fig. 3 does not change with variation of temperature over a wide range (4.2–300 K). Therefore, the noise analysis can be further performed at a certain fixed frequency. The most convenient value of this frequency, for technical reasons, is the frequency value $f = 10$ Hz. Below we analyze how this noise varies with temperature and applied voltage or current. Fig. 4 shows the spectral density of current noise as a function of the current flowing through the sample at different temperatures.

At temperatures of 30 K and 100 K, the dependencies have a nearly quadratic function. Usually, this behavior corresponds to the bulk noise in the conducting structures with a diffusion conductivity, which can be described by the formula

$$S_I = \alpha_H I^2 / fN, \quad (1)$$

where α_H is the dimensionless Hooge parameter, N is the total number of carries, I is the current and f is the frequency.

At the same time, the current dependence of noise at a temperature of 5.6 K is nearly linear. Such a dependence is registered in the temperature range corresponding to hopping conductivity ($T = 4.2\text{--}20$ K, Fig. 2b) in the current range of $I \approx 10^{-5}\text{--}10^{-4}$ A. Noise models of hopping conductivity in semiconductors predict a quadratic dependence of the noise as a function of the current through the sample ($\beta = 2$). The experimental results confirm these

findings [11–15]. Since our results contradict these data, we consider the origin of such a disagreement in more detail based on the analysis of experimentally obtained results.

The conductivity of the sample is determined primarily by metal nanotubes. At the same time, up to now the noise models of hopping conductivity have been developed for the case of semiconductor materials. It is logical to assume that the mechanism of hopping conductivity in metallic carbon nanotubes may differ from well-known mechanisms in semiconductors. For example, it is known [16] that in high-temperature superconductors in the region of hopping conductivity values of $\beta = 1\text{--}2$ are recorded. The authors relate the hopping conductivity in these materials to the hopping of carriers between the weakly localized states. In our case, the analog of these states can be a grid of bundles of carbon nanotubes (Fig. 1). In this system, the mechanism of charge transfer at low temperatures is assumed as a hopping conductivity [8,16,17]. The quadratic dependence (1) describes the current fluctuations that occur due to changes of the linear resistance. In the case of $\beta < 2$ the situation is different because of two reasons. Firstly, such relations are observed in nonlinear systems. For example, the shot noise inherent in the current passing through a potential barrier (non-linear system) is equal to

$$S_I = 2eI, \quad (2)$$

where e is the electron charge. Of course, the shape of the noise spectrum for the investigated nanotubes is far from the noise spectrum of shot noise, since the latter does not depend on frequency in a wide range of frequencies (“white” noise). However, linear noise dependence on current, as well as the assumption of hopping conductivity generating current, allows us to suggest that the noise at low temperatures can be considered as quasi-shot noise.

Secondly, in the case of $\beta < 2$ the voltage applied to the sample affects the development of fluctuations. It reduces or increases the potential barrier height of a p–n junction or a Schottky junction and reduces the potential barrier height for isolated charges (Poole–Frenkel effect). These, in fact, lead to nonlinear current–voltage characteristics of such objects. Flicker noise in these systems for $\beta = 1$ is described by the model of Hooge–Kleinpenning for current flowing through the p–n junction:

$$S_I = \alpha_H (eI/\tau f) M, \quad (3)$$

where τ is the minority carrier lifetime and M is a dimensionless coefficient, which has a value in the range from 0.25 to 2.5. This expression is obtained using Eq. (2) for the shot noise [18–20]. To test the applicability of this model of flicker noise to the particular situation it is usually necessary to calculate the value of α_H and compare it with the most commonly obtained value of $2 \cdot 10^{-3}$.

In the paper [21], a new approach is proposed for estimating the value of flicker noise using the coefficient α_H/τ , which has a narrow range of value dispersion of $10^2 - 5 \cdot 10^3 \text{ s}^{-1}$. This range has much smaller range of dispersion in comparison with $\alpha_H = 10^{-2} - 10^{-9}$. Based on the data shown in Fig. 4 (curve 1), the estimated value of $\alpha_H/\tau = fS_I/eIM$ obtained using Eq. (3) is found to be equal to $\alpha_H/\tau = 1250/M \text{ s}^{-1}$. This estimated value falls within the above-mentioned range for α_H/τ .

This estimation allows us to suggest that the mechanism of current flow through carbon nanotubes in the regime of hopping conductivity is similar to the mechanism of current flow in the system with potential barriers. The linear dependence of current noise on the current of CNT bundles in a temperature range with characteristic hopping conductivity can be described by the model of Hooge and Kleinpenning [18–20].

To analyze the nature of the noise current in the nanotube bundles, we measured the spectral density of current noise, S_I , as a function of the current at different temperatures in the range from 4.2 K to 300 K. Typical dependencies are shown in Fig. 4. Based on these results, we found the dependence of the coefficient β on the temperature (Fig. 5). Up to a temperature of $T = 8 \text{ K}$ the parameter is constant: $\beta = 1$. Above this temperature a transition region was registered, where the parameter increases up to a value of 1.9 ($S_I \propto I^{1.9}$). In the high temperature range at $T > 200 \text{ K}$, the parameter value is equal to 2.

In the temperature range relevant to $\beta \approx 2$, we determined the dependence of the normalized level of current noise on the temperature (Fig. 6). A characteristic feature of the dependence is a weak increase in noise ($S_I/I^2 \propto T^{0.16}$) in the temperature range 20–215 K with a further transition to a stronger dependence ($S_I/I^2 \propto T^{3.28}$) at temperature higher than 215 K. As can be

found from the data (Fig. 6), the ratio of noise levels at 300 K and 100 K is about 3. It is important that in the temperature range corresponding to the conductivity of Luttinger liquid [8] the temperature dependence of the noise is weak. Above the temperatures corresponding to Luttinger liquid conductivity, a sharp increase in the normalized current noise is registered.

The measurement results presented above allow us to analyze the mechanisms of conductivity in the carbon nanotube bundles and their correlation with the noise characteristics. There are three mechanisms of conductivity with characteristic behavior in different temperature ranges, as described above. Experimental temperature dependence of the resistance (Fig. 2) shows a transition from hopping conductivity to Luttinger liquid conductivity at temperatures of about 20 K. In the dependence of the noise characteristic parameter on temperature (Fig. 5), there is also a transition from the value equal to 1 to a value equal to 2 of the logarithmic slope in current noise dependence on current. A transition region is recorded in the temperature range $T = 8\text{--}25$ K, which includes the transition temperature from hopping conductivity to Luttinger liquid conductivity.

At higher temperatures, the logarithmic slope reaches a value of about 1.9, while above $T = 200$ K the slope is equal to 2. In the temperature dependence of the resistance (Fig. 2), this transition is very weakly resolved. At the same time, more pronounced changes in the behavior of the noise at $T > 200$ K are visible in the temperature dependence of the normalized current noise (Fig. 6). In this dependence, a slow increase in the level of noise as a function of temperature is registered in the temperature range from $T = 20$ K to $T = 215$ K. At higher temperatures, the noise increases sharply, in the range from $T = 215$ K to 300 K, the noise level increases to a value about 3 times higher than in the previous case. Based on the results of Ref. [8], the following conclusion can be drawn. The temperature of the sharp bend in the temperature dependence of the noise corresponds to the transition temperature from Luttinger liquid conductivity to diffusion conductivity.

Our experimental results show that the conductivity of a Luttinger liquid is a less “noisy” process than diffusion conductivity. A sharp increase of the noise in the case of diffusion conductivity can be explained by increased scattering processes of the carriers by phonons. At the same time, the conductivity of Luttinger liquid combines the conductivities determined by the quantum of resistances, which by their definition do not depend on temperature.

4. Conclusion

The results obtained allow us to analyze the mechanisms of conductivity in the system with carbon nanotube bundles and find their correlation with noise characteristics. Three different mechanisms of conductivity are considered depending on temperature. Experimental data show that the transition temperature from hopping conductivity to Luttinger liquid conductivity is about 20 K. It should be emphasized that also in this case the noise characteristics display a transition from the value of the logarithmic slope of the current dependence of noise equal to 1 to a value equal to 1.95. The transition region corresponds to the range of temperatures $T = 8\text{--}25$ K, which includes the transition temperature from hopping conductivity to Luttinger liquid conductivity.

At higher temperatures, the logarithmic slope increases to a value of about 1.95, while above $T = 200$ K, the slope is equal to 2. In contrast to relatively weak resistance dependence on temperature, more pronounced changes in the behavior of the transport at $T > 200$ K are registered in the temperature dependence of the normalized current noise. The latter demonstrates a slow increase in the noise level as the temperature increases from $T = 20$ K to $T = 215$ K. At higher temperatures, the noise increases sharply. There is a roughly threefold increase in the range $T = 215\text{--}300$ K. The temperature of the sharp bend in the temperature dependence of the noise corresponds to the upper temperature of the existence of Luttinger liquid in bundles of carbon nanotubes.

Our experiment showed that the conductivity of a Luttinger liquid is less “noisy” than the conductivity of the Fermi liquid. A sharp increase of noise in this latter region can be explained by increased phonon scattering and, as a result, decreasing of mean free path of carriers. At the same time, the conductivity of the Luttinger liquid up to the temperature $T \approx 200$ K is of a ballistic nature and is characterized by a low noise level, weakly dependent on temperature.

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Figure Captions:

Fig. 1 – (a) TEM micrograph of carbon nanotubes; (b) Raman spectrum, measured using an argon laser.

Fig. 2 – The resistance of SWCNT bundles as a function of temperature: (a) plotted in the double logarithmic scale, symbols are experimental data, solid line is a power law function $R \propto T^{-\alpha}$ with $\alpha = 0.4$; (b) plotted on a semi-logarithmic scale with dependence of $1/T^{0.25}$ in the temperature interval 5–25 K. The solid line is an exponential function $R \propto \exp(T_0/T)^{0.25}$ with $T_0 = 800$ K.

Fig. 3 – Typical voltage noise spectral density of metallic carbon nanotubes bundles, measured at applied voltage $V = 50$ mV and different ambient temperatures T : 1–5.6 K; 2–30 K; 3–100 K.

Fig. 4 – Current noise spectral density as a function of current, measured at frequency $f = 10$ Hz and different temperatures: 1–5.6 K; 2–30 K; 3–100 K. Solid lines show approximation by power law function: $S_I \propto I^\beta$ with $\beta = 1$ for line 1 and $\beta = 1.95$ for lines 2, 3.

Fig. 5 – The current noise exponent, β of power function $S_I \propto I^\beta$ determined from measured noise spectra as a function of temperature. The lines are guides for the eye.

Fig. 6 – Temperature dependence of the normalized current noise on the temperature at $f = 10$ Hz. Straight lines correspond to the slope of 0.16 (1) and 3.28 (2).

FIGURE 1

FIGURE 2

FIGURE 3

FIGURE 4

FIGURE 5

FIGURE 6