Gate-Voltage Dependence of Zero-Bias Anomalies in Multiwall Carbon Nanotubes

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Temperature dependence of zero-bias conductance of the vanadium (V)/multiwall carbon nanotube (MWNT)/V structure is studied. As temperature is reduced, the conductance decreases with a functional form consistent with a power law. For the first time, we find that the exponent depends significantly on gate voltage. This exponent dependence cannot be explained by Luttinger-liquid theory for ballistic MWNTs. We interpret the obtained results within the framework of the nonconventional Coulomb blockade theory for strongly disordered MWNTs.

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Carbon nanotubes [1] have attracted much attention due to their potential application to nanoelectronic devices. Two types of carbon nanotubes, single-walled carbon nanotubes (SWNTs) and multiwall carbon nanotubes (MWNTs), are reported to display different electronic properties: metallic SWNTs have two one-dimensional (1D) conduction modes and μm-long mean-free path, even when they are weakly disordered [2,3]. Their transport displays zero-bias anomalies (ZBA) [4–8], whose functional form is characteristic of the Luttinger-liquid (LL) state in 1D ballistic systems [9]. On the other hand, electron transport in MWNTs is controversial; a variety of transport mechanisms have been observed from ballistic transport (conductance quantization [10,11], LL-like ZBA [12–16]) to diffusive transport [17,18] and also strong localization [19,20]. Theories show that even a mismatch of helicity between adjacent shells causes a short mean-free path [21,22]. In addition, all kinds of likely defects, such as vacancies, contamination, contact to the substrate, and adsorbed molecules may result in nonballistic transport. From this viewpoint, interpretation of the ballisticlike transport in MWNTs seems to need special attention. Here, we focus on the LL-like ZBA.

The LL state is characteristic of 1D clean (ballistic) systems of interacting electrons, in which low-energy excitation is achieved by exciting an infinite number of plasmons, making the transport intrinsically different from that of a Fermi liquid. One of the main features of the LL state is power-law dependence of physical quantities, such as tunneling density of states (DOS), as a function of energy or temperature; for tunneling conductance, dI/dV, Kane and Fisher [9] obtained the following power-law dependence:

$$dI/dV \propto T^{\alpha} \quad \text{when } eV \ll k_B T, \tag{1}$$

and

$$dI/dV \propto V^{\alpha} \quad \text{when } eV \gg k_B T. \tag{2}$$

Here, T is temperature, V is bias voltage, and k_B is Boltzmann's constant. The exponent α depends not only on material but also on tunneling geometry (i.e., whether the electrons are injected into the end or the bulk). This LL-like ZBA is the most useful criterion for determining the existence of the LL state in electron-transport experiments. The LL-like ZBA has been observed in SWNTs with the exponent α consistent with the theoretical predictions [4–8], whereas in MWNTs the observed values of α are rather scattered between 0.04 and 0.3, and the interpretation is not straightforward; Bachtold *et al.* [13] observed the ZBA both in NT-gold and NT-NT junctions and explained it with large-mode LL theory, and Graugnard *et al.* [14] pointed out that the LL interpretation taking into account the intrinsic resistance of the NT itself is the most accurate. On the other hand, from the high-bias I-V characteristics, Tarkiainen *et al.* [15] concluded that the environmental-quantum-fluctuation theory associated with the Coulomb blockade successfully explains the ZBA better than the LL theory does, and Liu *et al.* [16] observed the ZBA in strongly disordered boron-doped MWNTs, attributing it to the localization effect.

Here, we report on our systematic study of the gate-voltage dependence of the LL-like ZBA in MWNTs. From gate-voltage dependence of the exponent α, we directly show that MWNTs are not in the LL state even if the LL-like ZBA is seen.

MWNTs used in this experiment were produced by the arc-discharge method and used without further treatment. The outermost diameter is about 10 nm and the length is several μm. The samples have a vanadium (V)-MWNT-V structure, where the V electrodes (source and drain) are placed upon a MWNT, as shown in Fig. 1(a). This structure was fabricated with e-beam lithography followed by helicon-sputtering deposition of V on a Si/SiO_2 substrate. Details of the process are described elsewhere [23]. The V films are 60 nm thick. Electron transport was measured in a cryostat, in which all measurement leads

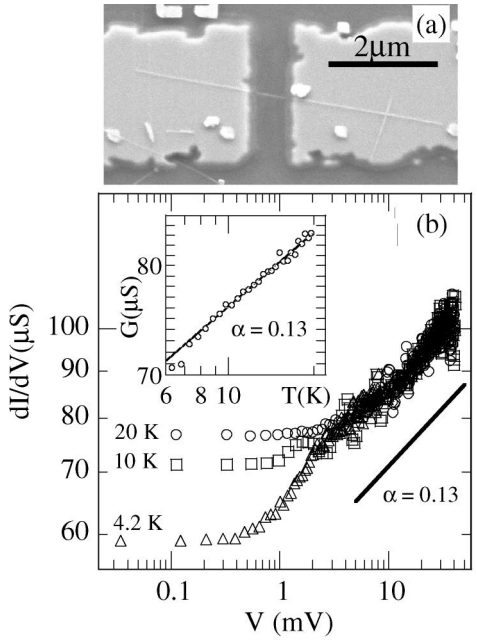

FIG. 1. (a) A scanning electron microscopy image of sample A. The right and left bright regions are V electrodes connected by a MWNT (white line). The MWNT is placed under the V electrodes (i.e., metal-on-tube configuration). The entire length of the MWNT and the length of its portion between electrodes are 5.7 μm (4.7 μm) and 0.7 μm (1.0 μm) for sample A (B), respectively. The diameter is about 10 nm. The gate voltage is applied through a back gate (under the substrate). (b) Inset: The temperature dependence of zero-bias conductance of sample A, which shows the power-law dependence with an exponent $\alpha = 0.13$. Main figure: The differential conductance dI/dV at three temperatures 4.2, 10, and 20 K, plotted as a function of applied voltage V. At high bias voltages ($V > 10$ mV), the data follow the power-law behavior with an exponent that is consistent with the $G - T$ characteristics.

were properly filtered for low-noise measurement. Possibly due to poor sputtering conditions without substrate annealing, we found no indication of superconductivity in the V film in our measurements down to 4 K. The V films were selected since their peak of sample-resistance distribution shifts to lower values in comparison with Ti or Pt/Au electrodes [24]. Zero-bias conductance was measured using a lock-in method with an excitation frequency of 27 Hz, while differential conductance was obtained by numerically differentiating I-V characteristics measured using low-noise current and voltage amplifiers. Gate-voltage (V_g) was applied through a back gate.

Although more than ten samples were examined, we focus on two samples (A and B) that showed a clear LL-like power-law dependence of zero-bias conductance at low temperatures. The high-temperature resistances are around 10 kΩ [8.5 to 10 kΩ (sample A) and 12 to 14 kΩ (sample B) at 200 K, depending on V_g]. The other samples showed zero-bias conductance decreasing faster than the power-law behavior with lowering temperature. Figure 1 shows the temperature dependence of the zero-bias conductance (G) and the I-V characteristics of sample A in a log-log plot at $V_g = 0$. $G - T$ characteristics are on a straight line between 6 and 20 K with an exponent $\alpha = 0.13$, and $dI/dV - V$ characteristics show power-law behavior at high-bias voltages, also with $\alpha = 0.13$. These features are consistent with the LL-like ZBA. To investigate them in more detail, we extended the temperature range to 200 K and applied gate voltage. For higher-temperature measurement, however, we gave up measuring I-V characteristics, because the expected voltage range for the LL-like behavior [Eq. (2)] was so high ($V > 7k_BT/e$) that the whole device might have been broken. Thus, only the $G - T$ characteristics are shown below.

Figure 2 is $G - T$ characteristics for the extended temperature range at different gate voltages. In Fig. 2(a), the LL-like power-law behavior obviously holds up to

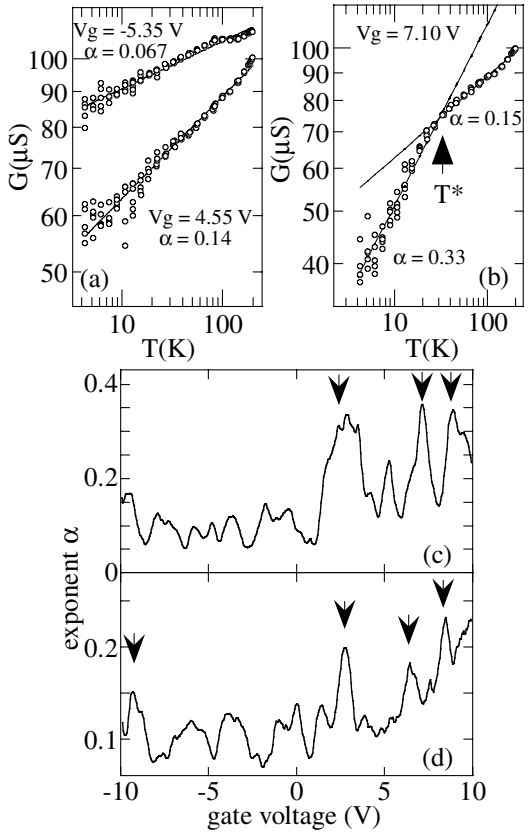

FIG. 2. Temperature dependence of the zero-bias conductance for different gate voltages in an extended temperature range between 4.2 and 200 K. (a) Both at $V_g = -5.35$ and 4.55 V, the data (white circles) follow the LL-like power-law behavior (solid lines), but the exponent takes a different value ($\alpha = 0.067$ and 0.14, respectively). (b) At $V_g = 7.10$ V, the exponent α changes from 0.33 to 0.15 at an inflection temperature T^*. (c) A figure summarizing the gate-voltage dependence of α of sample A, calculated from the data below 30 K. The inflection temperature T^* is observed around three V_g values (indicated by arrows), where α takes a peak value. (d) A similar $\alpha - V_g$ plot for sample B.

high temperatures at both $V_g = -5.35$ and 4.55 V but the exponent is different, while in Fig. 2(b) the exponent changes at an inflection temperature $T^* \approx 30$ K. In the measurement between $V_g = -10$ and 10 V with 0.05 V steps, such change in α was observed in three V_g ranges, i.e., around 2.4, 7.1, and 8.7 V, with width of about 1 V. In all cases, $T^* = 30$–50 K and α for higher temperatures is smaller. In the other gate-voltage ranges, the $G - T$ data follow the power-law dependence with a single exponent.

Figure 2(c) shows gate-voltage dependence of the exponent calculated from the data below 30 K. This is the main result we report in this Letter. From this figure, one notices that α depends significantly on gate voltage and ranges from 0.05 to 0.35, which is almost identical to the values of α reported thus far [12–16]. The characteristic V_g scale for α variation, ΔV_g, is around 1 V. $G - T$ characteristics with inflection [Fig. 2(b)] are observed around the peaks of α, as indicated by arrows. These characteristics were also observed in sample B, as shown in Fig. 2(d).

We now discuss the causes underlying these observations. Two theories have been suggested for the power-law behavior in MWNTs: one supports the existence of the LL state in MWNTs [25] and the other does not [26,27]. Reference [25] considers an MWNT composed of N ballistic metallic shells with long-range Coulomb interaction, internal screening effects, and single-electron hopping between shells. Power-law behavior corresponding to the LL state is derived with an exponent for bulk contact, $\alpha_b = (g^{-1} + g - 2)/8N$, where g is the N-dependent LL parameter $g = (1 + NU)^{-1/2}$ (U: electrostatic coupling constant between metallic shells). Substitution of proper parameter values gives an exponent that is consistent with experiments; e.g., $N = 10$ and $U = 16$ result in $\alpha_b = 0.13$ [25]. It should be noted that this theory does not take into account the effect of doping. It is generally accepted that an MWNT is hole doped in air with $E_F = 0.3$ to 0.5 eV, probably due to charge transfer from adsorbed oxygen or from the attached electrodes [28]. Furthermore, the Fermi level changes with gate voltage. The resulting exponent obtained under the doping effect would depend not only on N and U but also on the number of 1D conducting modes, M, which is modulated by doping level. The expected $e\Delta V_g$ is the product of the transverse quantization energy of the nanotube, $\Delta E_\perp = \hbar v_F/R$ ($v_F = 8 \times 10^5$ m/s is the Fermi velocity, $R \approx 5$ nm) and the coupling constant between the gate electrode and the MWNT, which is approximately C/C_0 (C: capacitance between the metal electrodes and the MWNT; C_0: capacitance between the gate electrode and the MWNT). The transverse quantization energy is about 0.1 eV, and previous data [24] show that $C \approx 1$ fF and $C_0 \approx 1$ aF in our sample layout; typical ΔV_g for the change in M is around 10^2 V. This value is much larger than the observed value of about 1 V, and, moreover, this picture predicts a stepwise change in α as a function of V_g, which is not consistent with the oscillatory behavior shown in Figs. 2(c) and 2(d). Thus, the LL picture fails to explain the observation.

A second possible interpretation of the power-law behavior would be that for a MWNT as a diffusive conductor. Egger and Gogolin [26,27] showed that doped MWNTs that are highly disordered (mean-free path $l \lesssim 2\pi R$) and that have electron-electron interaction display an effective and nonconventional Coulomb blockade arising from tunneling into a strongly interacting disordered metal, leading to the LL-like ZBA with an exponent $\alpha = (R/2\pi\hbar D\nu_0)\log(1 + \nu_0 U_0)$, where D is the diffusion constant, $\nu_0 = M/2\pi\hbar v_F$ is the noninteracting density of states, and U_0 is an effectively short-ranged 1D interaction. Here, the second factor is close to unity for weak interactions. By using $D \approx v_F l$, one obtains $\alpha \approx R/Ml$. It is not possible to estimate M and l for a particular MWNT, but substitution of reported values of $M \approx 10$ and $l = 5$–60 nm yields values of α that are near the experimentally observed values. To obtain oscillatory behavior in α, the mean-free path should fluctuate with the gate voltage. The question is whether this is possible or not.

To answer this, we refer to a theoretical work by Choi *et al.* [29]. They found that a single defect in a metallic carbon nanotube produces a quasibound state which causes a dip in conductance at a particular energy away from E_F via resonant backscattering. In experimental situations, this means that a defect affects conductance only in a limited range of V_g. This effect was experimentally verified by scanned gate microscopy [30]. Assuming that this is the case in our MWNTs, the nanotube conductance would vary in a complicated manner as a function of V_g due to the likely presence of quite a few defects. This would lead to oscillatory variation of l, because the conductance of a nanotube per unit length, G', is directly related to the electron mean-free path as $1/G' = R_K/Ml$ ($R_K = e^2/h$: the quantum resistance). From the almost perfect power-law behavior we observed in our analysis, we believe that the contact resistance between V and MWNT is dominant over the MWNT intrinsic resistance. To further study the intrinsic resistance of MWNTs, four-terminal measurement is essential.

A possible cause of the inflection temperature T^* is associated with the energy range of the power-law behavior in the nonconventional Coulomb blockade theory [27]. The upper limit shown in Ref. [27] as an example is around $(0.01–0.1)\Delta E_\perp$, depending on parameters such as l. In our case, this would correspond to a temperature of 10–100 K, which is in agreement with the observed values. The theory does not predict power-law behavior above T^*, as seen in Fig. 2(b); detailed theoretical and experimental investigations would be required for quantitative consideration.

Mishchenko *et al.* [31] developed a theory for disordered MWNTs that is valid for any relation between l

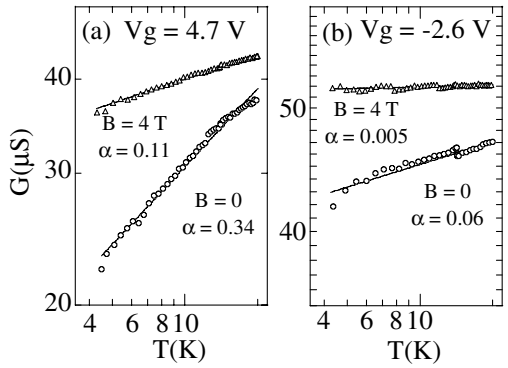

FIG. 3. Perpendicular magnetic field dependence of the $G - T$ data of sample B for two gate voltages $V_g = 4.7$ V (a), and -2.6 V (b), which correspond to a peak and a dip in $G - V_g$ characteristics (not shown), respectively. In both cases, the data follow the power-law behavior, and the exponent depends on the magnetic field.

and R. They showed that the power-law behavior of ZBA is absent in the case $l \gtrsim 2\pi R$. This suggests that our MWNTs are extremely disordered.

We also examined the dependence of α on perpendicular magnetic fields in sample B. Figures 3(a) and 3(b) show $G - T$ data for $B = 0$ and 4 T for two gate voltages, $V_g = 4.7$ and -2.6 V, which correspond to a peak and a dip in V_g dependence of conductance, respectively. In both cases, the $G - T$ data show the power-law dependence. The value of α depends not only on gate voltage but on magnetic field and, in most cases, α is smaller for higher magnetic fields [32].

Theoretically, it is predicted that a perpendicular magnetic field modifies the DOS of a nanotube [33], leading to the Landau level formation. This effect was observed in a MWNT single-electron transistor [34]. In addition, for disordered MWNTs, Roche *et al.* [35] showed that the diffusion constant depends both on the magnetic field and on the chemical potential. Assuming that the LL-like ZBA stems from the nonconventional Coulomb blockade effect, this modulation of the diffusion constant by magnetic fields also leads to the change in α, successfully explaining the results in Fig. 3.

In conclusion, we have studied the power-law dependence of conductance of MWNTs on temperature, which is sometimes regarded as evidence of the LL state. For the first time, we found that the exponent α depends significantly on the gate voltage. Neither the characteristic V_g scale for α variation nor the oscillatory behavior of α can be explained by the LL theory. We interpret the results within the framework of the nonconventional Coulomb blockade theory for a strongly interacting disordered metal. In some V_g ranges with large α, the exponent changes at an inflection temperature $T^* = 30$–50 K, with smaller α for higher temperatures. The detailed cause of this is not clear, but it might be related to the energy range of the power-law behavior. These results suggest that an observation of power-law behavior is not enough to prove the existence of the LL state. We also found that the exponent depends on a perpendicular magnetic field. This is attributed to the modulation of diffusion constant by magnetic fields, as suggested in Ref. [35].

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