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Gate-controlled Tomonaga-Luttinger liquid and atomic-like behaviors in peapod quantum dots

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Abstract. Anomalously high values of power α ($1.6 < \alpha < 12$) are found in power laws in conductance versus energy relationships in carbon-nanotube peapod quantum dots, encapsulating a chain of C_{60} molecules. This power is controllable by the applied back gate voltage. Atomic-like behaviors with doubly degenerate ground states are also found by single electron spectroscopy. They reveal that a portion of power originates from the Tomonaga-Luttinger liquid via the occupied electronic levels, which originate from the subbands unique to the peapods. This observation also clarify that the encapsulated C_{60} molecules do not directly contribute to the above quantum phenomena in peapods.

Introduction

Nano-peapods, which are single-walled carbon nanotubes (SWNTs) encapsulating a series of fullerenes, such as C_{60}, C_{70}, and Gd@C_{82} (C_{82}encapsulating Gd) in their inner space (*1*)(*2*), have recently attracted considerable attention; this is because their remarkable nanostructures yield exotic electron states, charge transport, and one-dimensional (1D) quantum phenomena.

In C_{60}@(n,n) peapods, which are arm-chair type SWNTs encapsulating C_{60} molecules, it has been predicted that electrons that were transferred from the SWNT accumulated in the space between the C_{60} molecules and SWNTs, resulting in the so-called nearly free electrons (NFEs) (*3*). Hybridization of these NFEs with the π and σ orbitals of C_{60} introduced four asymmetric subbands including the approximately doubly degenerate ground states in the C_{60}@(10,10) peapod as compared with the two subbands in conventional SWNTs (*3, 4*).

Measurements of semiconductive peapods encapsulating a series of Gd@C_{82} by a scanning tunnel microscope revealed that a conduction band was periodically modulated around Gd@C_{82} in a real space due to the hybridization of orbitals between the SWNT and Gd@C_{82} (*5*). Moreover, electrical measurements of peapods encapsulating C_{60} and Gd@C_{82} indicated the possibility of the presence of variable range hopping (*6*). Refs. (*3*)–(*6*) at least suggested the presence of charge transfer and orbital hybridization between the encapsulated fullerenes and SWNTs.

On the other hand, SWNTs are within a 1D ballistic charge transport regime and have exhibited a variety of quantum effects, such as quantized energy levels, Tomonaga-Luttinger liquid (TLL) (*7* - *10*), and atomic-like behaviors as quantum dots (e.g., even-odd effect, shell-filling to two spin-degenerate electronic states, and Kondo effect) (*11* - *14*). In particular, the behavior of TLL, which is a collective phenomenon arising from electron-electron interaction in 1D conductors, has been identified by observing power laws in relationships of conductance vs. energy in carbon nanotubes (CNs) (*7* - *10*). The reported correlation exponent *g*, which denotes the

strength of an electron-electron interaction, was as low as ~ 0.2 and implied the presence of a strong repulsive Coulomb interaction in CNs. How such phenomena are affected by encapsulating a series of fullerenes, however, has not yet been investigated in any peapods.

For the present study, peapod field-effect transistors (FETs) were fabricated. An SEM top view revealed that the FETs included two bundles of peapods (*2*) as the channels. Since the observed differential conductance was largely independent of the change in back gate voltage (V_{bg}), metallic transport in the present peapod was suggested. In the relationship of zero-bias conductance (G_0) to the temperature for different values of V_{bg} on a doubly logarithmic scale, we confirmed that G_0 monotonically decreases as temperature decreases for any values of V_{bg}. A distinct linear relationship (i.e., power law) with the power α as high as ~2.2 was observable for the entire temperature region only at V_{bg}=+5V in the figure..

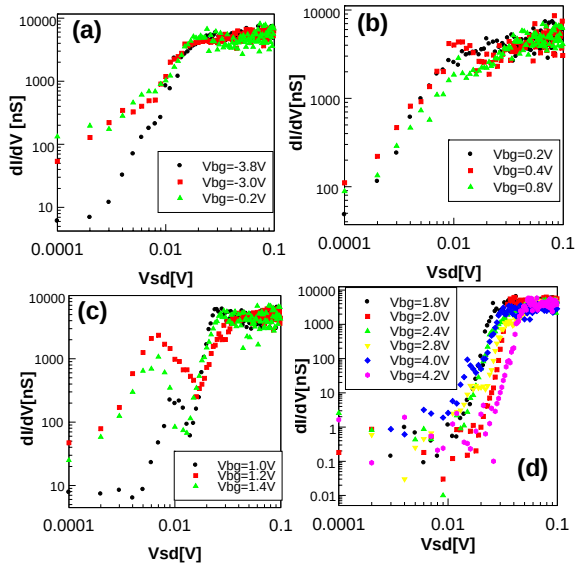

Fig.1

Fig.1: Relationships of differential conductance (dI_{sd}/dV_{sd}) to source-drain voltage (V_{sd}) on doubly logarithmic scales for four different V_{bg} regions at T = 1.5 K. **(a):** at $-V_{bg}$, no dI/dV follows a linear relationship. **(b)-(d):** $+V_{bg}$ regions. **(b):** The linearities with $1.6 < \alpha < 2$ are observable only at $V_{sd} < 0.01$ V. **(c):** Two linear relationships with different α ranges (i.e., α = 2–3 and α = 8–10 for $V_{sd} < 0.01$ V and $V_{sd} > 0.02$ V, respectively) are observable. **(d):** The linearities with α = 10–12 are observable only at $V_{sd} > 0.01$ V.

Fig.2: Dependence of α on different V_{bg} values, estimated from Fig.1. Three V_{bg} regions (**(I):** $V_{bg} <$ 0.8 V, **(II):** 0.8 V < V_{bg} < 1.8 V, and **(III):** 1.8 V < V_{bg} corresponding to Figs.1(b), (c), and (d), respectively) clearly exist.

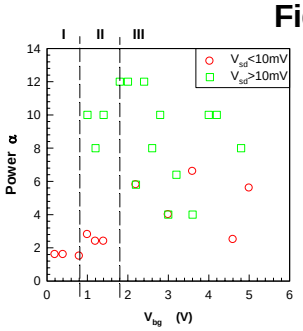

Fig.2

Figure 1 shows the relationships of differential conductance (dI_{sd}/dV_{sd}) to source-drain voltage (V_{sd}) on doubly logarithmic scales for one negative and three positive regions of V_{bg}. In the $-V_{bg}$ region, any dI/dV did not follow a linear relationship (a). On the contrary, saliently linear relationships with different α values are observable in the $+V_{bg}$ region. The behaviors are classified into three regions (b) - (d) as mentioned in the figure captions, showing an anomalously large α ($1.6 < \alpha < 12$). The values of α observed in all the V_{bg} values are shown in Fig.2. The differences in α among the three regions are apparent in this figure.

The presence of power laws was discussed as evidence for TLLs in CNs (*7* - *10*), as mentioned in the introduction. The values of α were very sensitive to the boundary conditions between the metal electrodes and CNs (*8*), namely the tunneling density of state; such as $\alpha^{bulk} = \sim 0.3$ and $\alpha^{end} = 2\,\alpha^{bulk}$ for the tunneling from an Au electrode to the bulk and to the end of CNs within the large-channel number TLL states, respectively (*8*). The formulas of α for each tunneling were also given by $\alpha^{bulk} = (g^{-1} + g - 2)/8$ and $\alpha^{end} = (g^{-1} - 1)/4$. However, the maximum value of α reported in CNs to date is approximately 1.25, except for refs.(*9*,*16*). Therefore, the α values of 1.6~12 observed in Figs.1 and 2 are anomalously large in comparison with the α values reported thus far in conventional TLLs (*9*). The junction structures in this study, in which the ends of the peapod bundles were placed under an Au electrode, should have shown a maximum α^{end} of only ~0.6. In fact, the empty peapods (i.e., SWNTs), which have the same structures as those used for the present peapods, have exhibited α ~0.8 even at the maximum case.

In contrast, the Coulomb diamonds that are the results of the measurements with single electron spectroscopy in the low V_{bg} region (Fig. 3) explains that the TLL is the origin of a portion of α ($1.6 < \alpha < 3$). These four diamonds as shown by n = 1 – 4 indicate a possibility of the presence of atomic-like behaviors with the doubly

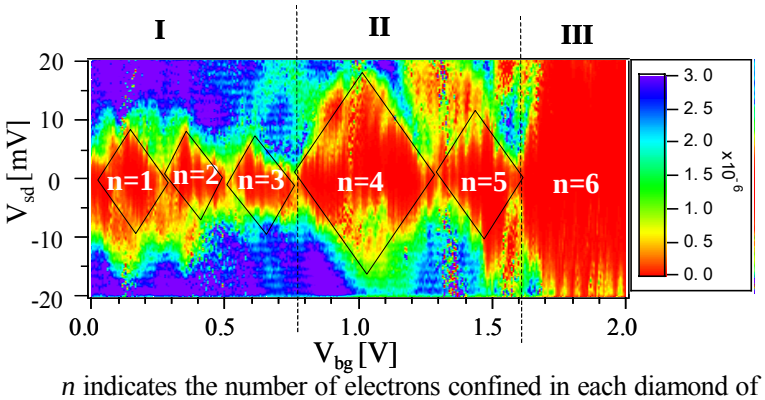

Fig.3: Coulomb diamonds (red regions surrounded by the solid lines) that show a sequence of one large diamond (n = 4) followed by three smaller ones (n = 1–3) of approximately equal size, observed at T = 1.5 K. The z-axis is the differential conductance with the magnitudes of which are indicated on the right side.

n indicates the number of electrons confined in each diamond of the peapod quantum dot. Regions I, II, and III well correspond to those in Fig.2. The power laws observed in Figs.1 and 2 primarily appear in the green areas. Only the power law in the low V_{sd} region at V_{bg} = 1 V appears in the n = 4 diamond.

degenerate ground states, which are similar to the diamonds observed in SWNT (*13*) and multi-walled CN (MWNT) quantum dots (*14*). It should be noted that the power laws shown in Fig.1 are observable in the green areas, which are outside of the diamonds. In contrast, the empty peapod (i.e., SWNTs encapsulating no C_{60}) quantum dots with exactly the same structures as those in the present peapod quantum dots exhibited no such a degenerate sate in contradiction to ref. (*13*) and showed just even-odd effect reported in conventional SWNT quantum dots. No atomic-like behavior was also observable in the $-V_{bg}$ region.

The value of U_c (the single-electron charging energy of the system) is approximately 6 ~ 10 meV in the small diamonds in Fig.3. This value for the present peapod with a length of 500 nm is approximately consistent with expectations based on a previous study of SWNT bundles (e.g. a U_c ~25 meV for a tube length of 100–200 nm) (*11*). This result emphasizes that the encapsulated C_{60} molecules do not contribute to the effective electrostatic capacitance for the single charging effect in this low $+V_{bg}$ region. Due to the high value of the applied V_{bg}, the outermost shell was depleted and the next outermost shell acted as a quantum dot in the MWNT (*14*). In contrast, this result implies that the SWNT is not depleted in this low $+V_{bg}$ region. Therefore, the observed atomic-like behaviors do not originate directly from the encapsulated C_{60} molecules.

On the other hand, ΔE (the energy spacing in the dot), which was estimated from the difference in the size of the small and large diamonds shown in Fig.3, is ~13 meV. This value is the energy spacing between the doubly degenerate electronic-states and the upper state. However, it is much greater than expectations from the previous studies in SWNTs (e.g., a ΔE of ~5 meV for a tube length of 100–200 nm) and the relationship $\Delta E = h v_F / 2L$ (L and v_F are the tube length and Fermi velocity, respectively). In addition, the observed larger diamonds in the $V_{bg} > 1.6$ V region in Fig.3 indicate the presence of an electronic state located at $\Delta E >$ ~25 meV. In the regions where V_{bg} is even higher, more diamonds and electronic states exist due to the quantization of the excited subbands.

Based on these results, we noted the number of electrons (*n*), which are confined in the peapod quantum dot, on each diamond (Fig.3). It should be noted that these electronic states near the ground states—observed at $V_{bg} <$ +2 V—are qualitatively in good agreement with the subband structures predicted in the C_{60}@(10,10) peapod (*3*); this is particularly evident in the presence of approximately doubly degenerate ground states, nevertheless the influence of V_{bg} and dot structure were not taken into consideration in the previous study (*3*). In contradiction to ref.(*13*), it is remarkable that the doubly degenerate subbands in the bulk of the peapod predicted in ref.(*3*) are still conserved even in the quantum dot. Since the empty peapods did not exhibit such behaviors as mentioned above, this is due to the subbands formed via NFEs and, hence, to a unique feature of the peapod quantum dot. The significantly low value of the applied V_{bg} shown in Fig.3 enabled us to observe the electronic states near the ground states unlike that reported in ref.(*14*).

Since the predicted subband structures originate only from the hybridization of orbitals in C_{60} and NFEs (*3*), the agreement with ref.(*3*) strongly indicates that the applied low $+V_{bg}$ plays a major role in the modulation of neither NFE nor C_{60}, and shifts only the position of the Fermi level in the SWNT. This is consistent with the absence of both depletion of C_{60} molecules by V_{bg} and contribution of C_{60} molecules to U_c, as explained above. Consequently, the encapsulated C_{60} molecules contribute only to the production of the four subbands via the NFEs and the electronic levels in the peapod quantum dots. The abovementioned two excited electronic states with large ΔE occur due to the quantization of the two excited subbands in the peapod.

Here, in order to interpret the power laws shown in Figs.1 and 2, it is crucial to note that the three V_{bg} regions observed in Figs.1 and 2 are in good agreement with the three V_{bg} regions classified by using the diamonds shown in Fig.3, and that the power laws shown in Figs.1 and 2 are basically observed in the green areas in Fig.3. These results clearly indicate that the power law behaviors with the large values of α are strongly associated with the number of (partially) occupied electronic states, *N*, in each diamond

The correlation of α (TLLs) with the orbital filling effect in CNs has not yet been reported in previous studies. Only ref. (17) predicted that a small *g* and large α could be obtained from the large *N* in peapods. The theory predicted $g = (1+2Nv_q/\pi\hbar v_F)^{-1/2}$ for armchair CNs, where *N* and v_q are the number of (partially) occupied symmetric subbands with degenerate Fermi vector waves and the same band width, and the electron-electron interaction matrix element, respectively. If the subbands are asymmetric and each of them crosses the Fermi level only once, *N* can be replaced by *N*/2. This holds true for the subbands of the C_{60}@(10,10) peapod in this study.

We quantitatively examine the validity of this theory for the present measurement by replacing *N* to the number of electronic states. The value of g = 0.135 is obtained from $\alpha^{end} = (g^{-1} - 1)/4$ (*8*) using $\alpha = 1.6$ that is observed in region I (N=2). The value of v_q can be estimated by substituting these values of *g* and N in $g = (1 + 2(N/2)v_q/\pi\hbar v_F)^{-1/2}$ (*17*). Then, g = 0.11 and 0.099 are respectively obtained for N = 3 and N = 4 by substituting the estimated value of v_q in $g = (1 + 2(N/2)v_q/\pi\hbar v_F)^{-1/2}$. The value of g = 0.11 is in approximately good agreement with g = 0.082 estimated from $\alpha^{end} = (g^{-1} - 1)/4$ by using $\alpha = 2.8$ that is observed in the portion of region II with low V_{sd} values. Consequently, the theory (17) is quantitatively relevant only when N = 2 and 3 under the same value of v_q. Therefore, at least, the power laws with $\alpha = 1.6 - 3$ can be attributed to the TLL via the occupied electronic levels with N = 2 and 3, which are located near the ground states unique to the peapod quantum dots. Further investigation is, however, required to clarify the origin of the extremely large $\alpha = 8 - 12$ observed in regions II and III as well as the presence of the two power laws for a single value of V_{bg}.

The observations in this study strongly suggest that the 1D quantum phenomena observable in peapods are very exotic, associated with the unique electronic states (formed via NFEs) of the peapod with gate-controllable fashion. Further investigation is required in order to develop a comprehensive understanding of these phenomena. .

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