Anomalous Coulomb diamonds and power-law behavior sensitive to back-gate voltages in carbon nanoscale peapod quantum dots

J. Mizubayashi,[1,2] J. Haruyama,[1,2] I. Takesue,[1,2] T. Okazaki,[3,2] H. Shinohara,[4,2] Y. Harada,[5,2] and Y. Awano[5,2]
[1]*Aoyama Gakuin University, 5-10-1 Fuchinobe, Sagamihara, Kanagawa 229-8558, Japan*
[2]*JST-CREST, 4-1-8 Hon-machi, Kawaguchi, Saitama 332-0012, Japan*
[3]*National Institute of Advanced Industrial Science and Technology, Tsukuba 305-8565, Japan*
[4]*Nagoya University, Furo-cho, Chigusa, Nagoya 464-8602, Japan*
[5]*Fujitsu Laboratory, 10-1 Wakamiya, Morinosato, Atsugi, Kanagawa 243-0197, Japan*
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We report anomalous charging effect of single electrons (Coulomb diamonds) observed in carbon nanoscale peapod quantum dots that encapsulate a series of C_{60} molecules. We find that behaviors of diamonds are anomalously sensitive to back-gate voltages (V_{bg}), exhibiting two evidently different V_{bg} regions and a large polarity on V_{bg}. In particular, we find only a sequence of one large diamond followed by three smaller ones existing around ground state. Magnetic-field dependence indicates the presence of shell filling by spin singlet to doubly degenerate electronic levels for these. The encapsulated-C_{60} molecules indirectly affect this shell filling at low V_{bg} possibly via nearly free electrons. In contrast, they act as individual quantum dots coupled in series in high V_{bg} region. It directly contributes to highly overlapped very large diamonds. Moreover, we report power-law behaviors on conductance versus energy relationships observed in the same carbon nanoscale peapods. We find that the values of powers are also highly sensitive to applied V_{bg} with three different regions and anomalously high at high source-drain voltages. Because the power laws are found at voltages, which are the nearest outside of the above-mentioned fourfold Coulomb diamonds, correlation of the anomalous powers with orbital-related Tomonaga-Luttinger liquid is discussed.

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I. INTRODUCTION

Carbon nanoscale 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) molecules in their inner space,[1,2] have recently attracted considerable attention. This is because their unique nanostructures are expected to yield exotic electronic states, quantum charge (spin) transports, and one-dimensional (1D) quantum phenomena. There are, however, still a few reliable reports that experimentally reported such electronic states and quantum phenomena.

From theoretical viewpoints, in $C_{60}@(n,n)$ peapods that are armchair-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, forming the so-called nearly free-electron (NFE) states.[3] Hybridization of these NFE states with the π and σ orbitals of C_{60} molecules introduced four asymmetric subbands including the approximately doubly degenerate ground states in the $C_{60}@(10,10)$ peapod in contradiction to 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] References 3–6 at least suggested the presence of charge transfer and orbital hybridization between the encapsulated fullerenes and SWNTs.

On the other hand, it is well known that SWNTs are within a 1D ballistic charge transport regime and exhibit a variety of quantum effects, such as quantized energy levels, Tomonaga-Luttinger liquids (TLLs), and shell (orbital) filling (atomiclike behaviors) as quantum dots.[7–10] For instance, when carbon nanotubes (CNs) act as quantum dots, electron can be placed on the quantized electronic levels (orbitals or shells) in the dots one by one via single-electron charging effect. This effect has caused shell filling in CN quantum dots,[7–10] such as even-odd effect, shell filling in two spin-degenerate electronic states, and Kondo effect. In particular, two different types of shell filling have been experimentally reported, that is, antiparallel spins (spin singlet) and parallel spins (spin triplet). Reference 22 proposed a theoretical model that explained the results by taking into consideration several families of single-electron states and Coulomb repulsion. Our present system is analogous to this model.

Moreover, the behavior of TLLs, 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 CNs.[12–15] The reported correlation exponent g, which denotes the strength of an electron-electron interaction, was as low as ~0.2. This implied the presence of a strong repulsive Coulomb interaction existing in CNs. How such phenomena are affected by encapsulating a series of fullerenes, however, has not yet been investigated in any carbon nanoscale peapods to date.

For present study, we report finding anomalous behaviors of Coulomb diamonds observed in carbon nanoscale peapods quantum dots that encapsulate a series of C_{60} molecules. First, we find that behaviors of diamonds are anomalously sensitive to back-gate voltages (V_{bg}), exhibiting two evi-

dently different V_{bg} regions [i.e., (1) small diamond region for $-1.7\ V < V_{bg} < +1.7\ V$, and (2) large diamond region for $V_{bg} < -1.7\ V$ and $+1.7\ V < V_{bg}$] and a large polarity on V_{bg}. In particular, we find only a sequence of one large diamond followed by three smaller ones existing only around ground state in $+V_{bg}$ region. Magnetic-field dependence indicates the presence of shell filling to doubly degenerate electronic levels by spin singlet for these. The size of diamond indicates that this is independent of the encapsulated-C_{60} molecules, basically in this low V_{bg} region. However, they might indirectly affect this shell filling via nearly free electrons, which accumulate on the space between C_{60}'s and SWNT by electron transfer from the SWNT. In contrast, we find that the encapsulated-C_{60} molecules directly contribute to overlapped very large diamonds by acting as individual quantum dots coupled in series in high V_{bg} region.

Next, we report finding the presence of power laws in conductance vs energy relationships, which is also highly sensitive to V_{bg} showing three V_{bg} regions in different power-law behaviors [i.e., (1) $V_{bg} < 0.8$ V, (2) $0.8\ V < V_{bg} < 1.7$ V, and (3) $1.7\ V < V_{bg}$]. Anomalously high values of powers (>8) are also found for V_{bg}'s >0.8 V in high V_{sd} regions. The power laws are found at voltages, which are the nearest outside of the above-mentioned fourfold Coulomb diamonds. Because these fourfold diamonds mean single-electron filling to doubly degenerate orbitals in the peapod quantum dot, correlation of the anomalous powers with orbital-related TLL states is discussed.

II. ANOMALOUS COULOMB DIAMONDS SENSITIVE TO BACK-GATE VOLTAGES AND SHELL-FILLING EFFECT

For the present study, field-effect transistors (FETs), using peapods encapsulating C_{60} molecules as the channel, were fabricated. A scanning electron microscopy (SEM) top view indicated that the FETs included two bundles of peapods[2] as the channels. The number of peapods included in one bundle was estimated to be approximately 20 from atomic force microscopy (AFM), and transmission electron microscopy (TEM) observations. Since change in the observed differential conductance was largely independent of the change in V_{bg}, metallic transport in the present peapod was confirmed.[11]

A. Anomalously V_{bg}-sensitive Coulomb diamonds

The measurement results by single-electron spectroscopy are shown in Fig. 1: (a) for $-4\ V < V_{bg} < +4$ V and (b) expansion of (a) for $0\ V < V_{bg} < +2$ V. Figure 1(a) indicates that (1) the sizes of Coulomb diamonds are highly sensitive to applied V_{bg} and (2) they have significant polarities on applied V_{bg} (i.e., asymmetric features between $+V_{bg}$ and $-V_{bg}$ ranges). From the first viewpoint, the sizes of diamonds can be evidently classified to the following two V_{bg} regions: (1) small diamond region for $-1.7\ V < V_{bg} < +1.7$ V, and (2) large diamond region for $V_{bg} < -1.7$ V and $+1.7\ V < V_{bg}$. In the first V_{bg} region, the sizes of diamonds are smaller than ~ 10 mV except for those appearing at $V_{bg} = +1$ V [noted as $n=4$ in Fig. 1(b)], whereas in the second V_{bg} region, they are approximately 20 mV except for a few diamonds at $V_{bg} < -1.7$ V and larger than 40 mV at $V_{bg} > +1.7$ V.

From the second viewpoint, the sizes of diamonds are very asymmetric as mentioned above. Moreover, periodicity of diamonds is quite different between $-V_{bg}$ and $+V_{bg}$ ranges in the first V_{bg} region. A sequence of closed one large diamond (noted as $n=4$) followed by three closed smaller ones [noted as $n=1-3$ in Fig. 1(b)] is observable for $0\ V < V_{bg} < +1.7$ V, whereas they cannot be observed for $-1.7\ V < V_{bg} < 0$ V. Although some similar-size diamonds are observable for $-0.5\ V < V_{bg} < 0$ V, they are unclosed and other diamonds for $-1.7\ V < V_{bg} < -0.5$ V become much smaller as V_{bg} value decreases. In the second V_{bg} region, the diamonds are highly overlapped and cannot be separated. In particular, the overlapping is very significant for $+1.7\ V < V_{bg}$.

It is a well-known fact that conventional CN quantum dots can place many electrons on those many quantized electronic levels one by one via single-electron tunneling and that shell-filling effects are mostly independent of V_{bg}, because the shape of dots is independent of V_{bg}. V_{bg} just changes the positions of chemical potentials in the conventional CN quantum dots, unlike most cases of semiconductor quantum dots. Only the case of impurity- or defect-induced many small quantum dots, which were connected in series, showed diamonds with inhomogeneous sizes, resulting in stochastic Coulomb diamonds. However, no such behaviors have been observed in our empty SWNT quantum dots (i.e., without C_{60}'s). Therefore, the above-mentioned anomalously high V_{bg}-sensitive Coulomb diamonds indicate strong association with contribution of the encapsulated-C_{60} molecules.

Here, the value of $U_c = e^2/2C_{eff}$ (the single-electron charging energy of the system; C_{eff} is the effective capacitance for single charging effect[21]) should be approximately 6–10 meV in the case of empty and high-quality SWNTs quantum dots (i.e., without C_{60}'s, defects, and impurities), if one follows the expectations based on only the length of present carbon peapod of 500 nm following a previous study of SWNT bundles (e.g., a U_c of ~ 25 meV for a tube length of 100–200 nm.[7] The values of $U_c (< 10$ meV) in the first V_{bg} region mentioned above are in approximately good agreement with this estimation of $U_c = 6-10$ meV. This strongly indicates no contribution of the encapsulated-C_{60} molecules at least to C_{eff} of U_c and only the SWNT acts as a quantum dot. Hence, single electrons flow only through the SWNT in this low V_{bg} range around 0 V and cannot flow into the C_{60} molecules.

B. Shell filling to doubly degenerated electronic levels at low $+V_{bg}$

In particular, the period of Coulomb diamonds observed for $0\ V < V_{bg} < +1.7$ V, as shown in Fig. 1(b), can be interpreted as the four diamonds, a sequence of one large diamond (noted as $n=4$) followed by three smaller ones (noted as $n=1-3$), as mentioned above. This sequence indicates the possible presence of shell-filling effect with the doubly degenerate electronic levels only at ground states, based on

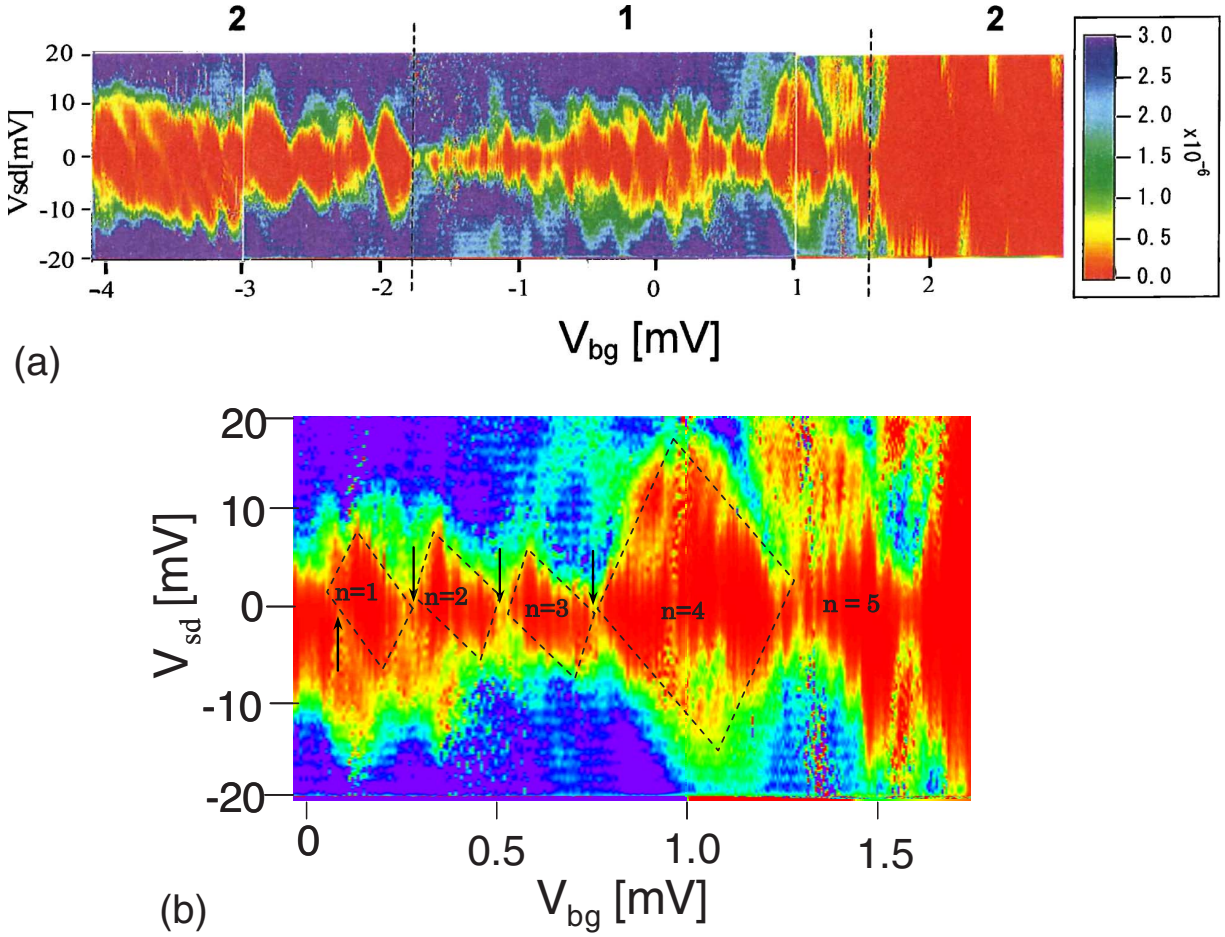

FIG. 1. (Color online) (a) Coulomb diamonds observed in a carbon nanoscale peapod quantum dot at $T=1.5$ K. The z axis is the differential conductance with the magnitudes of which are indicated on the right side. Dotted lines at $V_{bg}=\pm 1.7$ V indicate boundaries for two different V_{bg} regions for small and large diamonds. (b) Expansion of Coulomb diamonds (red regions surrounded by the dotted lines) for $0\ \mathrm{V}<V_{bg}<+1.7$ V in (a). n indicates the number of electrons confined in peapod quantum dots for each diamond. The dotted diamonds are guides to the eye. Arrows mean the V_{bg} points for Fig. 2.

previous reports of the fourfold diamonds in SWNT (Ref. 9) and multiwalled CN (MWNT) quantum dots.[10] However, the observed sequence of these diamonds is only one set for $0\ \mathrm{V}<V_{bg}<+1.7$ V, because appearance of much larger diamonds obstructs the observation of further sets of such sequence. This result is very anomalous as compared with those periodically observed over wide ranges of V_{bg} in Refs. 9 and 10 In conventional CN quantum dots, such one-set degenerate levels cannot be found, because individual nondegenerate electronic level is formed only from quantization of two subbands existing in bulk of a SWNT, while only in some cases, all levels are doubly degenerate, such as the fourfold diamonds, as observed in Refs. 9 and 10.

Hence, in order to confirm the presence of shell-filling effects and doubly degenerate levels for Fig. 1(b), we have investigated the V_{bg} shift of the linear-response conductance peaks [i.e., shown by arrows in Fig. 1(b)] as a function of magnetic field B perpendicular to the tube axis. The result, Fig. 2(a), reveals that adjacent peaks shift in opposite directions. This is a behavior of spin singlet state whose spins alternate as $S=0\rightarrow 1/2\rightarrow 0\ldots$ and exist on the same orbital state, unlike a spin triplet state formed by Hund's rule. In Fig. 2(b), we plot additional energy, which was deduced from the separation of adjacent peaks involving electrons on the same orbital in Fig. 2(a) (i.e., peaks 1 and 2, and peaks 3 and 4), as a function of magnetic field B. A dashed line shows the result of the best fit of the data to $U_c+g_L\mu_B B$,

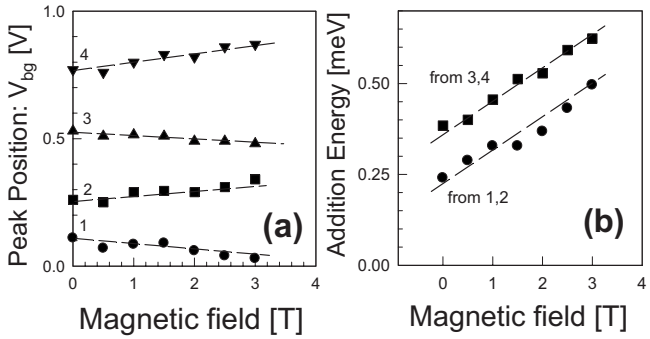

FIG. 2. (a) V_{bg} shift of conductance peak positions [at $V_{bg}=0.11$, 0.26, 0.53, and 0.77 around $V_{sd}=0$ V shown by arrows in Fig. 1(b)] in Fig. 1(b) as a function of magnetic field B. (b) Additional energy obtained from each peak pair in Fig. 2(a) versus B.

where μ_B is the Bohr magneton and g_L is the Lande factor, and gives $g_L=1.96$. This value of g_L is approximately consistent with those mentioned in Ref. 10. Therefore, we conclude that Fig. 1(b) indicates the presence of doubly degenerate electronic levels existing only at ground states and the presence of a shell filling to such levels by spin singlet.

Because of the absence of interaction between the C_{60} molecules and the SWNT as mentioned above, these doubly degenerate electronic levels are not directly associated with the encapsulated-C_{60} molecules. Therefore, this does not correspond to the model predicted in Ref. 22. These, however, may be indirectly associated with the C_{60} molecules via NFEs and doubly degenerate subbands due to the NFE states as follows.

Reference 3 predicted that the doubly degenerate subbands, which exist only at the ground states, originated from the hybridization of orbitals of the encapsulated-C_{60} molecules and NFE states in bulk of C_{60}@(10,10) peapod as mentioned in the Introduction. In contrast, in the case of conventional quantum dot structure, this degeneration is solved and the subbands are quantized to many electronic levels (shells). However, because the channel length of present peapod FET is as long as 500 nm, the level spacing $\Delta E=h\nu_F/2L$ (L and ν_F are the tube length and Fermi velocity, respectively) should be as small as ~3 meV even in nondegenerate levels. In such a case, the doubly degenerate subbands around the ground state may still remain, resulting in formation of approximately double-degenerate electronic levels around $V_{bg}=0$ V, which was observed in Fig. 1(b), even in the quantum dot.

In contrast, this shell-filling effect was not observable for $-1.7\ \text{V}<V_{bg}<0$ V as mentioned above. Moreover, the sizes of unclosed diamonds are inhomogeneous without specified periodicities in this V_{bg} range. These behaviors are analogous to stochastic Coulomb diamonds, which have been reported in impurity- or defect-induced small quantum dots connected in series, although the number of quantum dots is very small in the present case because the size of diamonds is as small as less than 10 mV.

These significantly different behaviors of diamonds for $\pm V_{bg}$ range evidently support the correlation of the doubly degenerate levels and shell filling with NFE states mentioned above. This is because NEF states can be formed by electron transfer from SWNT to the space between the encapsulated C_{60}'s and the SWNT as mentioned in the Introduction[3] and applying $+V_{bg}$ induces this electron transfer as well as the single-electron injection from source electrode. In contrast, applying $-V_{bg}$ obstructs these electron transfers, resulting in the above-mentioned stochastic diamonds in the low $-V_{bg}$ range. This also implies a possibility of the presence of some defect-induced small quantum dots in the SWNT for yielding these stochastic diamonds, because no electron transfer from the SWNT exists and, thus, such electrons in the SWNT may be injected into the small dots.

C. Interplay between encapsulated-C_{60} molecules and SWNTs at high V_{bg}

On the other hand, the sizes of diamonds in the second V_{bg} region are much larger than those in the first V_{bg} region showing heavy overlapping and, thus, this means the values of C_{eff} are smaller than those in the first V_{bg} region. This implies disappearance of the shell filling and the connection of a few dots mentioned above in the first $\pm V_{bg}$ regions. In conventional Coulomb diamonds, such large and unclosed diamonds have been interpreted by a series connection of defect- or impurity-induced many small dots. However, because such large diamonds have not been found in our empty SWNT quantum dots as mentioned earlier, they do not correspond to the present case. This indicates a possibility that individual encapsulated-C_{60} molecules behave as individual small quantum dots and they are electrostatically coupled to each other, because applying high $+V_{bg}$ strongly induces the above-mentioned electron transfer from SWNT to the space between C_{60}'s and SWNT for yielding NFEs. The induced transfer results in excess accumulation of the NEFs and the excess NFEs can make single-electron tunneling into individual C_{60} molecules. In such a case, electron will flow only through the encapsulated-C_{60} molecules connected in series under applied V_{sd}, because C_{60} molecules are fully encapsulated into the inner space of the SWNT and neighboring C_{60}'s face each other via tunnel junction in most parts of the present peapods.

If there is no such a coupling and encapsulated-C_{60} molecules act as independent quantum dots, our system corresponds to the model proposed in Ref. 22. This will lead to shell filling by parallel spins (spin triplet), because of the presence of Tomonaga-Luttinger liquid (Coulomb repulsion) as mentioned in the next section.

The values of U_c's of ~20 meV for $V_{bg}<-1.7$ V and ~40 meV for $+1.7\ \text{V}<V_{bg}$ in the second V_{bg} region are at least two and four times larger than those for the first V_{bg} region (<10 meV). Hence, the values of the total capacitance of C_{60} molecules, which are coupled with SWNTs, can be estimated to be the value of 1/3 capacitance of SWNTs (C_{SW}) for $+1.7\ \text{V}<V_{bg}$. In contrast, it can be estimated to be the same value as (C_{SW}) for $V_{bg}<-1.7$ V. This indicates the presence of different origins to yield effective capacitance C_{eff} for U_c for $\pm V_{bg}$. As mentioned above, electron transfer from SWNT to the C_{60}'s could not occur in the $-V_{bg}$ range. Hence, this large value of C_{eff} may be attributed to single charging effect of defect or impurity-induced small quantum dots as well as that for $-1.7\ \text{V}<V_{bg}<0$ V, although large $-V_{bg}$ value induces charging effect, resulting in the larger diamonds.

In conclusion, our argument implies that the anomalously high V_{bg}-sensitive Coulomb diamonds are attributed to the following: (1) shell filling to doubly degenerate electronic levels associated with NFE states in low $+V_{bg}$ region with no direct interaction between the encapsulated-C_{60} molecules and the SWNT and (2) single-electron injection from the SWNT into the C_{60} molecules, which act as individual quantum dots coupled in series, in high $+V_{bg}$ range. Moreover, we argued that diamonds in $-V_{bg}$ region originate from defect- or impurity-induced small quantum dots. In order to further clarify these arguments, more investigation is expected, such as by changing the number of encapsulated-C_{60} molecules.

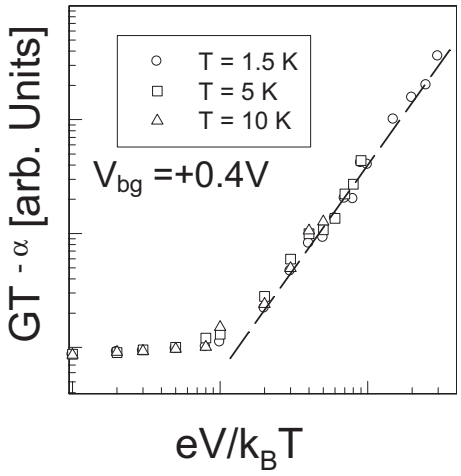

FIG. 3. The double-logarithmic plot of differential conductance ($G=dI_{sd}/dV_{sd}$) divided by T^{α} as a function of eV/k_BT measured at $V_{bg}=+0.4$ V for three different temperatures.

III. ANOMALOUS POWER-LAW BEHAVIORS AND ORBITAL-RELATED TOMONAGA-LUTTINGER LIQUID

A. Anomalous power-law behaviors

Figure 3 shows the double-logarithmic plot of differential conductance divided by T^{α} as a function of eV/k_BT measured at $V_{bg}=+0.4$ V for three different temperatures. All data collapse on a single universal value showing saturation at $eV/k_BT<hv_F/L$. These results are qualitatively consistent with those in previous reports of TLL states in CNs.[13]

Figure 4 shows the relationships of differential conductance (dI_{sd}/dV_{sd}) to V_{sd} on doubly logarithmic scales for one $-V_{bg}$ and three $+V_{bg}$ regions. In the $-V_{bg}$ region, any differential conductance did not follow a linear relationship, as shown in Fig. 4(a). On the contrary, saliently linear relationships with different α values are observable in the $+V_{bg}$ region, although the V_{sd} regions with exhibiting power laws are narrow at V_{sd}'s >10 mV in Figs. 4(c) and 4(d) (e.g., half decade). The behaviors are classified into the following three V_{bg} regions; (1) $V_{bg}<0.8$ V, the linearities with $1.6<\alpha<2$ are observable only at $V_{sd}<0.01$ V [Fig. 4(b)]; (2) 0.8 V $<V_{bg}<1.7$ V; two linear relationships with different α ranges (i.e., $\alpha=2-3$ and $\alpha=8-10$ for $V_{sd}<0.01$ V and $V_{sd}>0.02$ V, respectively) are observable [Fig. 4(c)], and 3. 1.7 V $<V_{bg}$, the linearities with $\alpha=10-12$ are observable only at $V_{sd}>0.01$ V [Fig. 4(d)].

The summary of values of α observed in all the V_{bg} region included in Figs. 4(b)–4(d) is shown in Fig. 5. The differences in tendencies of α among the three regions are apparent in this figure. Moreover, the values of α observed in empty SWNTs are also shown in this figure. All the values are less than 1, which is consistent with previous reports of TLLs in SWNTs. This implies that Fig. 4 is unique to peapods.

B. Correlation with possibly orbital-related Tomonaga-Luttinger liquid

The presence of power laws has been discussed as evidence for TLLs in CNs,[12–15] as mentioned in the Introduc-

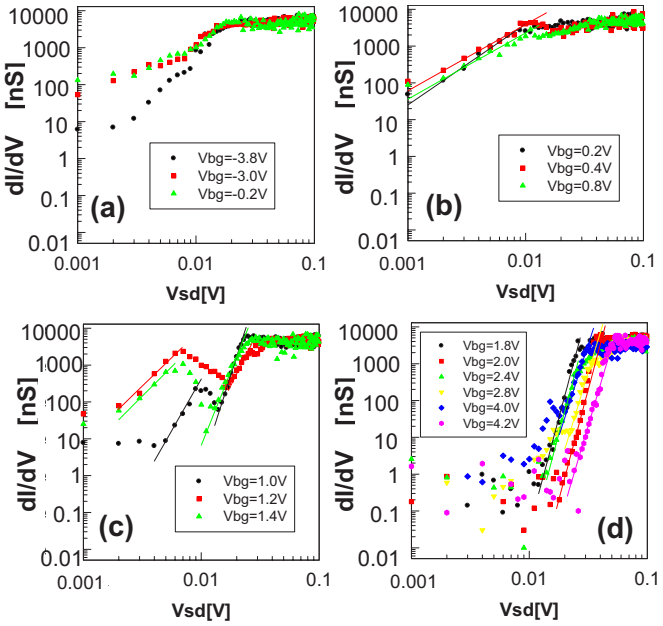

FIG. 4. (Color online) Relationships of dI_{sd}/dV_{sd} to source-drain voltage ($eV_{sd}/k \gg T=1.5$ K measured) on doubly logarithmic scales for four different V_{bg} regions; (a) $V_{bg}<0$ V, (b) 0 V $<V_{bg}<0.8$ V, (c) 0.8 V $<V_{bg}<1.7$ V, and (d) 1.7 V $<V_{bg}$. These power laws primarily appear just at the nearest-outside regions of fourfold Coulomb diamonds in Fig. 1(b). Only the power law in the low V_{sd} region at $V_{bg}=1$ V appears inside of the large Coulomb diamond in fourfold diamonds. The liner lines were obtained from accurate data fitting including measurement points as many as possible and values of power α were exactly estimated.

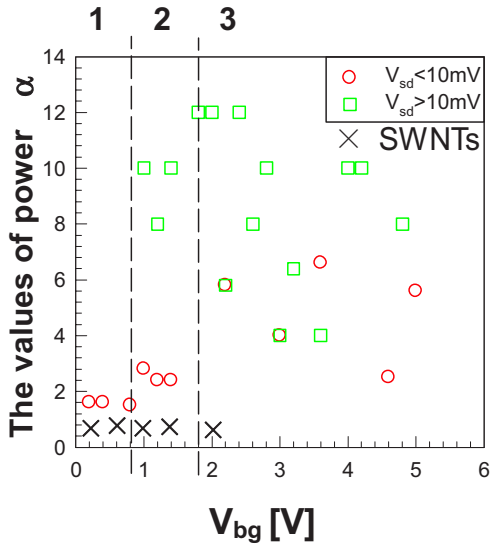

FIG. 5. (Color online) Dependence of power α on different V_{bg} values, estimated from Figs. 4(b)–4(d), in both the present peapod and the empty SWNT quantum dots. Three different V_{bg} regions [(1) $V_{bg}<0.8$ V, (2) 0.8 V $<V_{bg}<1.7$ V, and (3) 1.7 V $<V_{bg}$ corresponding to Figs. 4(b)–4(d), respectively] separated by dotted lines are evident. Several values of α were added in addition to Fig. 4(d) only in region 3.

tion. The values of α were very sensitive to the boundary conditions between the metal electrodes and CNs,[13] namely, the tunneling density of state, such as $\alpha^{\text{bulk}}=\sim 0.3$ and $\alpha^{\text{end}}=2\alpha^{\text{bulk}}$ for the tunneling from a Au electrode to the bulk and to the end of CNs within the large-channel number TLL states, respectively.[13] The formulas of α for each tunneling were also given by $\alpha^{\text{bulk}}=(g^{-1}+g-2)/8$ and $\alpha^{\text{end}}=(g^{-1}-1)/4$. However, it should be noted that even the maximum value of α reported in previous CNs to date is approximately 1.25, except for Refs. 14 and 17. Therefore, we imply that the α values of 1.6–12 observed in Figs. 3 and 4 are anomalously large in comparison with the α values reported thus far in conventional TLLs.[14] The junction structures in this study, in which the ends of the peapod bundles were placed under a Au electrode, should have shown a maximum α^{end} of only ~ 0.6. In fact, the empty SWNTs have exhibited $\alpha=\sim 0.8$ even at the maximum case, as explained for Fig. 5 above.

Here, it should be noticed that the power laws, as shown in Figs. 4(b)–4(d), were found at the V_{bg}'s in the nearest-outside voltage regions of the Coulomb diamonds, as shown in Fig. 1 of Sec. II. Region 1 ($V_{\text{bg}}<0.8$ V) for Fig. 4(b) mostly agrees with the V_{bg} region including three small diamonds [$n=1,2,3$ in Fig. 1(b)], whereas region 2 ($0.8\text{ V}<V_{\text{bg}}<1.7$ V) for Fig. 4(c) agrees with the V_{bg} region including one large diamond and one small diamond [$n=4$, 5 in Fig. 1(b)]. Region 3 ($V_{\text{bg}}<1.7$ V) for Fig. 4(d) agrees with the V_{bg} region, showing very large diamonds without shell filling. Moreover, no shell-filling effect was observed in $-V_{\text{bg}}$ region in Fig. 1(a). This agrees with the presence of no power laws in Fig. 4(a).

Each Coulomb diamond region for this shell (orbital)-filling region meant the presence of electrons, which were placed one by one on electron orbitals via single-electron charging effect, in the peapod quantum dot. Hence, the observed power laws sensitive to V_{bg} are strongly associated with the number of such electrons (n) in the peapod quantum dot and the number of (partially) occupied electronic levels (N); i.e., $n=1,2,3$ ($N=1,2$) for $V_{\text{bg}}<0.8$ V and $n=4,5$ ($N=2,3$) for $0.8\text{ V}<V_{\text{bg}}<1.7$ V.

The correlation of power laws, the values of α, and TLL states with the electronic-level filling effect (i.e., orbital-filling effect) in CNs have not yet been reported in previous studies. Only a single study,[13] however, 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 bandwidth, 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 results by replacing N to the number of (partially) occupied electronic levels (orbitals) and using the same value of v_q. The value of $g=0.135$ is obtained from $\alpha^{\text{end}}=(g^{-1}-1)/4$ (Ref. 13) using $\alpha=1.6$ that was observed in region 1 ($N=1,2$) in Figs. 4(b) and 5. The value of v_q can be estimated by substituting these values of $g=0.135$ and $N=2$ in $g=(1+2(N/2)v_q/\pi\hbar v_F)^{-1/2}$.[18] Then, $g=0.11$ and $g=0.099$ are, respectively, obtained for $N=3$ and $N=4$ by substituting this estimated value of v_q in $g=(1+2(N/2)v_q/\pi\hbar v_F)^{-1/2}$. The value of $g=0.11$ obtained for $N=3$ is in approximately good agreement with $g=0.082$ estimated from $\alpha^{\text{end}}=(g^{-1}-1)/4$ by using $\alpha=2.8$ that was observed in the portion of region 2 with low V_{sd} values in Figs. 4(c) and 5.

On the other hand, this g value is irrelevant to $\alpha=8-10$ that is observed in the portion of region 2 with high V_{sd} values in Figs. 4(c) and 5. Moreover, the g value of 0.099 leads to $\alpha=2.96$ for $N=4$, which is significantly less than the values of $\alpha>10$ observed in region 3 at high V_{sd} values in Figs. 4(d) and 5. These indicate that different values of v_q should be used for the case of higher V_{sd}. Because strength of electron-electron interaction varies from low to high V_{sd}'s, this is reasonable. When different values of v_q are used for large N, $\alpha=10$ (for $N=3$) and 12 (for $N=4$) could be obtained from the values of $2\,v_q/\pi\hbar v_F=1160$ and 1250, respectively, $g=(1+2(N/2)v_q/\pi\hbar v_F)^{-1/2}$, and $\alpha^{\text{end}}=(g^{-1}-1)/4$.

Consequently, the theory[18] is quantitatively relevant when $N=2$ and 3 (at lower V_{sd}) under the same value of v_q and $N=3$ (at higher V_{sd}) and 4 under the larger values of V_q. This indicates that the presence of two power laws observed in Fig. 2(c) is attributed to change in v_q due to increase in V_{sd}. Therefore, we conclude that the power laws with large values of $\alpha(1.6<\alpha<12)$ can be attributed to the TLL via the occupied doubly degenerated electronic levels, which are located near the ground states unique to the peapod quantum dots.

However, the high V_{sd} region for power laws in region 2 does not locate at the nearest outside of Coulomb diamonds. In addition, power laws observed in region 3 are not consistent with no shell-filling effect reported in. (Ref. 16) These may indicate that such power laws are not associated with orbital-related TLLs. Therefore, further investigation is required to reconfirm relevance of the values of v_q and comprehensive understanding of these power-law behaviors.

Hence, other interpretation should be discussed described as follows. If the capacitance of peapods is ~ 600 times smaller than those in MWNTs due to the presence of C_{60} molecules electrostatically coupled with the SWNT in series and the value of N is as large as 10–20 like those in MWNTs, the large-channel TLL model coupled with external electromagnetic environment shown for MWNTs (Refs. 13, 19, and 20) may explain the $\alpha=8-12$, because α in conventional MWNTs is given by $2R/R_Q=2(L/C)^{1/2}/R_Q\approx 0.44$, where L is the kinetic inductance given by $R_Q/2Nv_F$ (≈ 1 nH/μm), C is the external electrostatic capacitance (≈ 30 aF/μm), and R_Q ($=h/e^2$) is the quantum resistance. This may correspond to the high values of α in region 3, because we reported that the encapsulated-C_{60} molecules were not electrostatically coupled with the SWNT at $V_{\text{bg}}<+1.7$ V (i.e., in regions 1 and 2), while the coupling occurred at $V_{\text{bg}}>+1.7$ V (region 3).

IV. CONCLUSION

In conclusion, we reported anomalous behaviors of Coulomb diamonds observed in carbon nanoscale peapod quan-

tum dots that encapsulated a series of C_{60} molecules. We found that behaviors of diamonds were anomalously sensitive to V_{bg}, exhibiting two evidently different V_{bg} regions [i.e., (1) small diamond region for $-1.7\ \mathrm{V} < V_{bg} < +1.7\ \mathrm{V}$, and (2) large diamond region for $V_{bg} < -1.7\ \mathrm{V}$ and $+1.7\ \mathrm{V} < V_{bg}$] and a large polarity on V_{bg}. In particular, we found only a sequence of one large diamond followed by three smaller ones existing around ground state. Magnetic-field dependence indicated the presence of shell filling to doubly degenerate electronic levels by spin singlet state for these. The size of diamond indicated that this was independent of the encapsulated-C_{60} molecules. However, they might indirectly affect this shell filling via NFEs, which accumulate on the space between C_{60}'s and SWNT by electron transfer from the SWNT. In contrast, we found that the encapsulated-C_{60} molecules directly contributed to overlapped very large diamonds by acting as individual quantum dots coupled in series in high V_{bg} region.

Moreover, we reported the power-law behaviors on conductance vs energy relationships observed in the same carbon nanoscale peapods. We found that the values of powers α were highly sensitive to V_{bg} also showing three different regions [(1) $V_{bg} < -1.7\ \mathrm{V}$, (2) $-1.7\ \mathrm{V} < V_{bg} < +1.7\ \mathrm{V}$, (3) $+1.7\ \mathrm{V} < V_{bg}$] and anomalously high ($\alpha > 8$) at high V_{sd} voltages. Because the power laws were found at the nearest-outside voltages of the above-mentioned fourfold Coulomb diamonds, the correlation of the anomalous powers with orbital-related TLL states was discussed. Further investigation is required in order to develop a comprehensive understanding of these phenomena (e.g., changing the number of encapsulated-C_{60} molecules).

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