Statistical Characterization of Dispersed Single-Wall Carbon Nanotube Quantum Dots

M. Shimizu$^{1,2}$, S. Moriyama$^1$, M. Suzuki$^{1,3}$, T. Fuse$^{1,4}$, Y. Homma$^{2,3}$, K. Ishibashi$^{4,3}$

1 Advanced Device Laboratory, The Institute of Physical and Chemical Research (RIKEN), 2-1, Hirosawa, Wako, Saitama 351-0198, Japan

2 Department of Physics, Tokyo University of Science, 1-3, Kagurazaka, Shinjuku-ku, Tokyo, 162-8601, Japan

3 CREST, Japan Science and Technology (JST), Kawaguchi, Saitama, 332-0012, Japan

4 Department of Information Processing, Tokyo Institute of Technology, 4259, Nagatsuta-cho, Midori-ku, Yokohama 226-8503, Japan.

e-mail: kishiba@riken.jp

Abstract. Quantum dots have been fabricated in single-wall carbon nanotubes (SWCNTs) simply by depositing metallic contacts on top of them. The fabricated quantum dots show different characteristics from sample to sample, which are even different in samples fabricated in the same chip. In this report, we study the statistical variations of the quantum dots fabricated with our method, and suggest their possible origin.

1. Introduction
Recently, transport properties of an individual carbon nanotube (CNT) have been measured, and interesting physics has been revealed [1-3]. Besides, the CNT is attractive for building blocks of extremely small nanodevices [4-6] that are not possible to fabricate with conventional lithography techniques.

However, the device process to fabricate electrical contacts to an individual CNT is still in a preliminary stage. In most cases, CNTs are grown on a substrate by the Chemical Vapor Deposition (CVD) with catalysts, or CNTs are dispersed on a substrate from the liquid solution that contains CNTs. Then, the electrical contacts are fabricated by using standard semiconductor processing techniques. Despite the big potential of the CNT as a subject for physics studies and as a building block of the new devices, the reproducibility of the device process is usually low. The aim of this study is to characterize the variation of the quantum-dot transport of the samples fabricated by the dispersion process. It appears that the two-terminal resistance is an important measure to determine the quantum dot behavior at low temperatures.

2. Sample fabrication and experimental procedure
Single-wall carbon nanotubes (SWCNTs) are used in this study. Samples are fabricated as follows. First, the large pads for the electrical wiring and the marks for the electron beam lithography were fabricated. Then, the SWCNTs that were contained in a solution were dispersed on the Si substrate, the surface of which was thermally oxidized. The metallic contacts, which were Ti in this case, were fabricated on the specific SWCNT by electron beam lithography. To do this, the position of the SWCNT with respect to the alignment marks was measured with an atomic force microscope (AFM). The contacted SWCNTs are considered to be individual, judged from their height of ~1nm. We should note that before evaporating metal, the substrate with SWCNTs on it were rinsed in water to hopefully remove chemicals that might be attached on the SWCNTs. The distance between two contacts were set at L~200nm for all samples (See inset of Fig.1(a))

The samples were set in a ⁴He cryostat, and the two terminal current measurements were carried out. The metallic SWCNTs were selected for the study. After the resistance was measured at room temperature (RT), the sample was cooled down to ~3K, and the single electron transport measurements were carried out, which was the current measurement as a function of the gate voltage (V_g) and the source-drain voltage (V_sd). The heavily doped substrate was used for the back gate.

3. Results and discussions

Figure 1(a) shows the number of counts as a function of the RT resistance, and information on whether the sample was a single dot or multi-dots at low temperature was included. The resistance varies very much from sample to sample, but there is a clear tendency that the samples with a large resistance tend to contain multi-dots, while those with a relatively low resistance tend to be single dot nature. Here, we define the type of dots (single or multi), depending on whether the Coulomb oscillations have a single period or are irregular. The examples of the Coulomb oscillations are shown in Fig.2, where (a)-(c) are those of the single dot, while (d) are those of the multi-dot. In Fig.2 (d), a current peak rarely appears, but the number of peaks increased as V_sd was increased, a typical behavior of multi-dots. From the histogram (a), it can be seen that the resistance in the multi-dots is much larger than that of single dot. This means that the resistance of the multi-dots which originates from the scattering centers in or near the dots dominates the total resistance, and is much larger than the contact resistance between the SWCNT and Ti.

Figure 1(b) categorizes various types of the single quantum dots, assigning (a) the close-dot, (b) the open/close dot and (c) the open dot. Typical Coulomb oscillations of these single dots are shown in Fig.2 (a)-(c). The close-dot is defined as a dot whose Coulomb gaps go to zero, as shown in Fig.2 (a). The open-dot is defined as a dot whose Coulomb gaps do not go to zero and have a finite current, as shown in Fig.2 (b). The open/close dot is an intermediate case where Coulomb gaps go to zero in some gate voltage ranges, but have a finite current in other gate voltage ranges, as shown in Fig.2 (c). It is reasonable in the histogram that the open-dot has a smaller resistance and the close-dot has a larger

![Figure 1](image)

**Figure 1** (a) Number of counts as a function of the RT resistance. This also includes the low temperature behaviour of the sample. (b) The variation of the degree of confinement as a function of the room temperature resistance
resistance. The Coulomb oscillations of the open/close dot, shown in Fig.2 (b), are not fully understood, in that the current at the Coulomb gap varies very much in different gate voltage ranges. In some gate voltage ranges, the device behaves like a close-dot, while in other gate voltage ranges, it behaves like an open-dot sample. This observation may suggest that the tunnel barrier depends on the gate voltage.

In our samples, a whole SWCNT between source-drain contacts form a single quantum dot. This is confirmed by the zero-dimensional level spacing ($\Delta E$) which is usually determined by the distance between the contacts (L), as discussed later. This means that the SWCNT underneath the contacts appears to be insulating, and the tunnel barrier appears to be formed at the edge of the contact metal, as indicated in the inset of Fig.1 (a). The reason for the formation of the insulating layer is not clear, but the electron beam lithography with an acceleration voltage of 50kV is a possible origin for a damage layer formation. In fact, we have observed that the electron beam irradiation to the SWCNT through the electron beam resist increases the two-terminal resistance, suggesting that the e-beam irradiation produces some damages to the SWCNT.

Having understood that the tunnel junction is formed in a tiny area of the order of ~1x10nm$^2$ between the SWCNT with a diameter of ~1nm and the edge of the metallic contacts, it may be reasonable that the tunnel resistance shows the observed fluctuations. The origin of the tunnel barrier may be Schottky barrier or a kind of insulating layer which is possibly a residue of organic molecules that come from the solution of SWCNTs. The gate voltage dependent tunnel barrier observed in the open/close samples may possibly account for the assumed origin of the tiny tunnel barrier. Since the back gate is used, the gate field not only affects the dot itself, but also affects the tunnel barrier that may have energy dependence. This could lead to the gate-dependent tunnel barrier.

Figure 3 shows the Coulomb diamonds and corresponding Coulomb oscillations of single dots in different regimes. Fig.3 (a) is for the open dot, (b) is an open regime of the open/close dot, and (c) is for the closed region of the open/close-dot. Fig.3 (d) corresponds to the close-dot. From Fig.3 (d), the charging energy for a single electron ($E_c=e^2/C_6$) and $\Delta E$ are roughly estimated as ~23meV and ~3.7meV, respectively. The experimentally estimated $\Delta E$~3.7meV is in good agreement with the theoretical estimation of 4.0meV that is obtained from the expression, $h\nu_F/4L$, where $v_F=8.1x10^5$m/s and L=200nm is the length between the contacts. This supports the previous assumption that the dot is a whole SWCNT between the source-drain contacts. There are some interesting observations in the

![Figure 2 Coulomb oscillations of (a) the close-single dot, (b) open/close-single dot, (c) open single-dot, and (d) multi-dots](image-url)
Figure 3 Coulomb diamonds and oscillations of (a) open-dot, (b) the open region of the open/close-dot, (c) the closed region of the open/close-dot, and (d) the close-dot diamonds. First, the effect of $\Delta E$ is not observed when the dot is not completely closed ((a) and (b)), possibly because the lifetime broadening may be larger than $\Delta E$. Second, $E_c$, which is the maximum value of the Coulomb gap in the diamond, is smaller for the open dot, compared with that of the closed dot (See Coulomb diamonds of (a)-(d)). Even in the same sample, such as (a) and (b), $E_c$ appears to be larger as the current at the Coulomb gap becomes small. These observations suggest that the quantum fluctuation due to weak confinement effectively reduces the charging energy [17]. Third, the observed $\Delta E \sim 3.8\text{meV}$ in the closed region of the open/close-dot (Fig.3(c)) is almost equal to that of the close-dot, even though $E_c$ is different for the two samples. This indicates that the degree of the confinement is effective on $E_c$, but not on $\Delta E$.

4. Conclusion
In conclusion, we have fabricated electrical contacts on an individual SWCNT that is dispersed on a thermally oxidized Si wafer. The fabricated samples showed large variations in the room temperature resistance that is related to the characteristics of the quantum dot transport at low temperatures. The variation of the samples may come from the microscopic details of the tiny tunnel barriers formed at the SWCNT and the contact-edge junction. The charging energy appears to depend on the degree of the confinement, but the level spacing does not. For the reliable and reproducible fabrication of CNT based nanodevices, the fabrication process that is controlled in nanoscale dimensions is necessary.

References
[1] S. Moriyama, T. Fuse, M. Suzuki, Y. Aoyagi, K. Ishibashi, Phys. Rev. Lett., 94, 186806 (2005)
[2] M. Bockrath, D. H. Cobden, J. Lu, A. G. Rinzler, R. E. Smalley, L. Balents, P. E. McEuen, Science, 397, 598 (1999)
[3] J. Nygrad, D. Cobden, P. E. Lindelof, Nature, 408, 342 (2000)
[4] S. J. Wind, J. Appenzeller, R. Martel, V. Derycke, Ph. Avouris, Appl. Phys. Lett. 80, 3817 (2002)
[5] K. Ishibashi, D. Tsuya, M. Suzuki, and Y. Aoyagi, Appl. Phys. Lett., 82, 3307 (2003)
[6] N. Mason, M. J. Biercuk, C. M. Marcus, Science 303, 655 (2004)
[7] L. W. Molenkamp, K. Flemmsberg, M. Kemerink, Phys. Rev. Lett., 75, 4282 (1995)