Selecting single quantum dots from a bundle of single-wall carbon nanotubes using the large current flow process

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Abstract

We demonstrate selecting a Coulomb peak which originates from a single quantum dot, from a bundle of many single-wall carbon nanotubes. The method uses the previously reported current flowing process, by which the number of nanotubes can be reduced for the transport. By adjusting the gate voltage in an appropriate range, the single peak belonging to the single quantum dot has been selected. The effect of the high frequency application on the peak has been investigated, and it is shown that the basic response can be explained by the adiabatic response of the single dot to the high frequency signal.

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Carbon nanotubes may be used as a building block of extremely small quantum-dot (QD-) based nanodevices which could work at high temperature [1,2]. Because of the small dot size, the corresponding energy scales associated with the dot, such as the charging energy \( E_c = e^2/C_S \) and the level spacing of the zero-dimensional confinement states (0-D states) \( \Delta E \), can be larger by an order of magnitude or even more, compared with those for lithographically made semiconductor quantum dots [3]. The QDs in carbon nanotubes are usually formed by just depositing the metallic contacts on single-walled carbon nanotubes (SWNTs) [4,5], however, the process is not sufficiently reliable and controllable in order to realize more complicated nanodevices. One of the reasons for this could be that the contacted nanotubes usually form a bundle of many indvidual nanotubes which could be either semiconducting or metallic. Furthermore, the deposited metal is not necessarily contacted to all of the individual nanotubes which form the bundle. In the previous report [6], we presented a large current flow process to modify transport properties of quantum dots in the bundle of SWNTs at 1.5 K. After the process was carried out a few times and the resistance \([h/e^2]\) became high, the gate voltage (\( V_g \)) dependence of the current (\( I_d \)) showed irregular Coulomb blockade oscillations, and the number of peaks decreased as the process was repeated further more. The observation could be explained by assuming that each current flowing process broke some of nanotubes in the bundle. In this report, we apply this technique to select the peak belonging to the single quantum dot in a bundle of SWNTs, and show that the high frequency response of the selected peak is consistent with the behavior of the single quantum dots.

The scanning electron microscope (SEM) image of the sample is shown in Fig. 1(a). SWNTs were dispersed on the Si substrate with a thermal oxide on its surface. The Au/Ag alloy for the source-drain contacts was deposited on top of SWNTs which were very likely to be a bundle of many SWNTs [7,8]. All the measurements and the large current flowing process were carried out at 1.5 K in vacuum. Just after the metal deposition, the current–voltage (\( I_d - V_{sd} \)) curve showed a linear behavior as shown in Fig. 1(b), giving...
the two-terminal linear resistance of about 30 kΩ at 1.5 K for this specific sample. After the current flowing process was repeated several times, the $I_d - V_{sd}$ curves became to show the non-linear characteristic due to the Coulomb blockade, as described below.

Fig. 1(c) shows the several consecutive $I_d - V_{sd}$ curves which correspond to the current flowing process. The general observation of the figure is that the $I_d - V_{sd}$ curves appear to be linear at the low bias voltages and start to bend downwards at higher voltages. The bending may be associated with the heating of SWNTs due to the large current. It is also seen that the small bias resistance increases as the process is repeated. Fig. 1(d) shows the magnified graph of Fig. 1(c) near the zero-bias region. It is clearly seen that the curves begin to exhibit the non-linearity even at the small bias region after several processes. In the 13th process (13), the current is suppressed in the bias voltage range of about $|V_{sd}| < 20$ mV, which turns out to be due to the Coulomb blockade effect. Fig. 2(a)–(d) show the Coulomb oscillations measured at $V_{sd} = 1$ mV after each current flowing process. It is also observed that the number of current peaks appeared in the fixed $V_g$ range ($-5 \text{V} < V_g < +5 \text{V}$) seems to decrease as the current flowing process is repeated. However, when the flat regions in Fig. 2(c) are zoomed up, the small current peaks can be found, as shown in the inset. These peaks could be originated from a single dot, and the periodic peaks appeared while other dots happened to be in the Coulomb blockade condition. The basic explanation of what happens by the current flowing process is as follows. The bundle of many nanotubes is considered to be many nanotubes connected in parallel between source and drain contacts. As the current flowing process is repeated, nanotubes with a small contact resistance would be broken because the current preferably flows through them. After some nanotubes were burned down by the process, leaving a few numbers of nanotubes with a large contact resistance, the Coulomb peaks belonging to the alive nanotubes start to be sufficiently separated in the gate voltage range. In some appropriate range, shown in the inset of Fig. 2(c), the observed peaks are possibly originate from the single quantum dot. The Coulomb gaps are from 10 to 20 meV. However, they are periodic in some region, and the charging energy could be estimated about $E_c \approx 10$ meV which is consistent with the previous measurement of the similar devices [5].

To confirm whether the Coulomb peak could be originated from the single dot, the peak shape fitting has been performed for the peak after the #13 process with the following expression [9].

$$G = G_{\text{max}} \frac{\delta k_B T}{\sinh(\delta k_B T)}$$

Here, $G_{\text{max}}$ is the peak maximum and is independent of temperature in this regime. $\delta$ measures the distance to the center of the peak in units of energy, which is related to the gate voltage in terms of $\delta = e(C_g/C_2)(V_{g\text{res}} - V_g)$, with
the gate voltage at resonance ($V_{g,\text{res}}$). The formula assumes the Coulomb peak in the classical regime where $\Delta E$ is smaller than the temperature. The peak width is basically determined by the thermal smearing of the Fermi level in the source and drain contacts. The agreement between the theoretical expression and the experimental peak that is shown in Fig. 3(a) is fairly good with an obtained fitting parameter of the electron temperature $T_e = 1.5$ K.

The peak shape of the Coulomb oscillation is strongly affected by the microwave irradiation, as seen in Fig. 3(b). As the microwave power of $f = 1.5$ GHz is increased, the peak shape starts to change and show the S-like shape. The behavior is similar to the rectification effect, first observed in the semiconductor single quantum dot with the lower frequency application [10]. In that case, electrons followed the time varying field (adiabatic regime) and these experimental observations could be explained by the classical square-law type response of the single dot. The estimated escape rate from the current peak for the single electron transport ($I = I/e$, $I \approx 200$ pA) is about $1 \times 10^{-9}$, comparable to the applied frequency. Although the rough estimate of the time scale does not sufficiently support the adiabatic transport regime, the basic behavior of the experimental results appears to support it. For the qualitative understanding of the Coulomb peak under the adiabatic time varying field, we could reasonably assume that the microwave radiation couple to the source drain contacts with a macroscopic scale (a few hundreds of micrometer).

The electrons in the nanotube is very likely to be shielded by the electrical leads and contacts which exist very close to the nanotube (within less than 1 $\mu$m) in a scale of the wave length of the irradiated microwave ($\sim 5$ cm). When the oscillating field is coupled to the source-drain contacts with some difference of the phase, it has been shown in Ref. [10] that the rectification effect occurs and the peak shape is modulated as seen in Fig. 3(b). These facts strongly suggest that the selected peak originates from a single quantum dot.

In conclusion, we have fabricated metallic contacts on a bundle of SWNTs. It is demonstrated that the current flowing process reduces the number of nanotubes which contribute the transport. When the number of nanotubes is sufficiently small, the Coulomb peaks from the different dots are distinguished, and the peak belonging to some specific single quantum dot is shown to be selected. The peak shape fitting and the microwave response of the peak have confirmed the validity of the present method. The present results indicate that the bundle of nanotubes could be used as a single quantum dot in the limited gate voltage range, to find some useful applications of single quantum dots.

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