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Nanoscale fabrication of a single multiwalled carbon nanotube attached atomic force microscope tip using an electric field

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We report a simple, repeatable, reliable method and influential conditions for assembling a single multiwalled nanotube (MWNT) to the end of a metal coated atomic force microscope (AFM) tip. The influential conditions consist of the frequency and magnitude of the induced voltage, the concentration of carbon nanotube (CNT) solution and the shape of the tip's apex. The optimal experimental factors needed for a single MWNT deposition using the dielectrophoretic force were obtained through repeated experiments. Applying an electric field of 0.6 to 0.7 V_{pp}/μm at 5 MHz, dropping a droplet of the transparent MWNT solution dispersed in the ethanol in a range of 0.5 to 1 $\mu\ell$, we obtained a CNT AFM tip with just a single MWNT attached. Furthermore, we found that the curvature of the tip's apex is a great influential factor in a single MWNT-attached tip. We expect that the appropriate size of curvature can improve the yield of single MWNT attachment. The effectiveness of the MWNT-attached AFM tip is demonstrated by direct comparison with AFM images of a bare AFM tip for a standard sample. © *2005 American Institute of Physics.* [DOI: 10.1063/1.1891445]

Since their discovery in 1991,[1] Carbon Nanotubes (CNTs) have been used in many nanoscale applications due to their unique mechanical, electrical, and chemical properties. Distinctive mechanical properties of CNTs such as sharpness, high aspect ratio, high mechanical stiffness, high elasticity, and metallic or semiconducting characteristics have provided vast research and developmental opportunities in the application of CNT in the measurement of atomic force microscopy (AFM) images by the attachment of the CNT to AFM tips.[2,3] Previous research for CNT-attached AFM tips has focused on CNTs fabricated by chemical vapor deposition (CVD) method including a batch process for the fabrication of CNT tips.[4,5] This has been demonstrated to be a mass producible method, because multiple probes with single-walled nanotubes at the apex of the tips can be produced simultaneously on a large sized wafer. However, there are some limitations such as nonuniformity of growing conditions and length of the CNT at the tip. Recently, multiwalled nanotubes (MWNT)-attached AFM tips were obtained using a magnetophoretic method or manual attachment after CNT deposition on electrodes using an electric field.[6–8] A *W* tip with a long SWNT fibril was made by drawing mechanism under dielectrophoresis.[9,10] However, this process obtained a *W* tip not with a single CNT, but with a bundle of CNTs.

In order to obtain high resolution and chemically convenient modification, a thin and short nanotube tip with a single SWNT might be required. Further, a single MWNT AFM tip having an approximately 10–100 aspect ratio in the case of CNTs with 10 nm diameter is useful to measure deep trench structures in semiconductors, and bio and optic devices. For improving the resolution of surface measurements, the aligning and attaching technique of just a single CNT on the apex of an AFM tip is really needed.

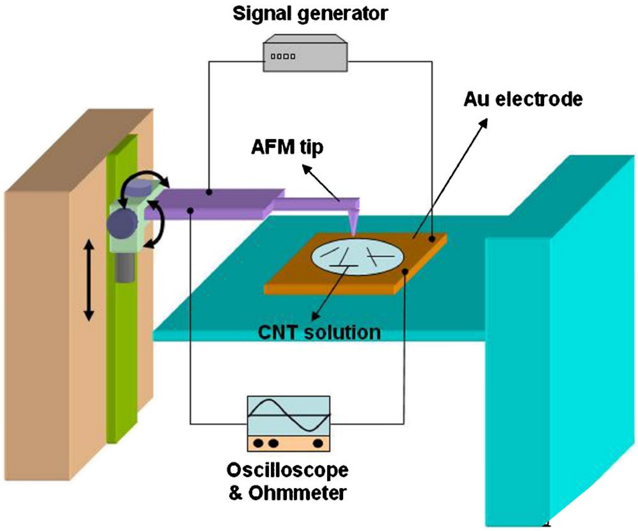

FIG. 1. (Color online) Dipole moment on CNTs and particles in nonuniform electric field. Dielectrophoresis, which is the translation motion of neutral matter caused by the polarization effect in a nonuniform electric field, should be carefully distinguished from electrophoresis, which is a motion caused by the response to a free charge on a particle in an electric field. A neutral particle polarized in an electric field such as MWNTs moves to the region of highest field intensity in a nonuniform electric field (Ref. 14).

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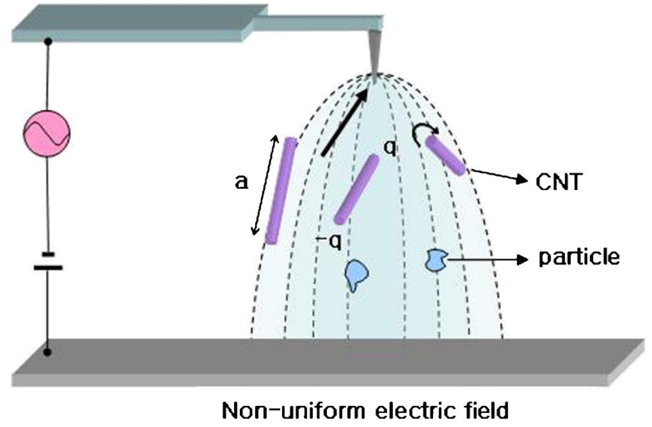

FIG. 2. (Color online) Schematic of the electrochemical assembly apparatus with ac electric source. The apparatus consists of two parts: a processing part and an inducing part. In the processing part, the holder of AFM tip was fixed to the *Z* axis translation stage for maintaining the distance from the counter electrode. This stage is able to be operated in rolling and pitching directions with a tilting angle of 0 to 90 deg. The inducing part comprised a signal generator and signal detector such as an oscilloscope and an ohmmeter.

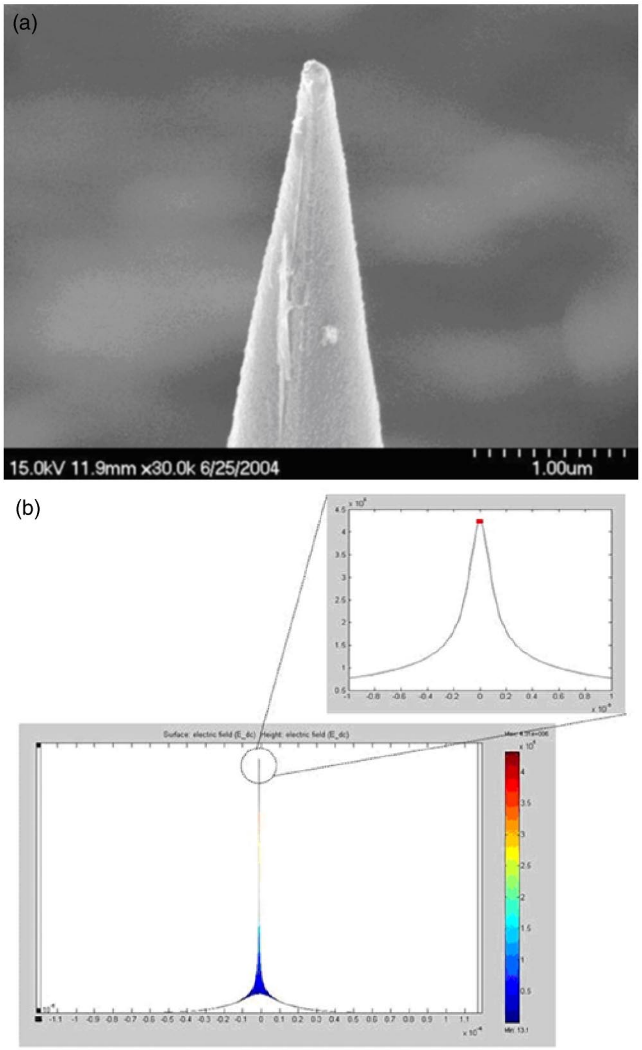

FIG. 3. (a) (Color online) FESEM images of AFM tip used in this research. The shape of the tip remained conical with a curvature of 100–150 nm. (b) The simulation result of the electric field. The area where the dielectrophoretic force is zero is the area where the gradient of the electric field is zero.

In this article, we report a simple instrument and attachment process of manufacturing of AFM tips with a single MWNT attached by utilizing an electrochemical method. This method is a simple, inexpensive, and highly reproducible approach to obtaining a single CNT-attached AFM tips.

The most plausible explanation for the MWNT tip assembling process is dielectrophoresis. This induced dipole, or polarization, can make a particle move, translate, and rotate along the gradient of the electric field. Figure 1 shows the dipole moment on a CNT and a particle in a nonuniform electric field.

When an ac electric field is applied to the electrodes, polarizability is induced on the carbon nanotubes, and a CNT is affected by a dipole moment due to the nonuniform electric field. The dielectrophoretic force that arises due to the nonuniform electric field can be expressed by the following equation:[11–15]

$$F_{\mathrm{DEP}} = 2\pi a^3 \varepsilon_m \mathrm{Re}\left[\frac{\varepsilon_p^* - \varepsilon_m^*}{\varepsilon_p^* + 2\varepsilon_m^*}\right] \nabla |E|^2, \qquad (1)$$

where a is the longest dimension of the particle, ε_m is the dielectric constant of the medium, ε_p is the dielectric constant of the particle, and E is the electric field. This force is affected by the applied frequency due to the complex permittivity expression, $\varepsilon^* = \varepsilon - i\sigma/\omega$, where σ is conductivity and ω is the frequency of the ac electric field.

In this assembling process, particles with dipole moment move toward the high density of the electric field gradient in a dielectric medium such as ethanol. When we dropped the solution with nanotubes onto the gap between the AFM tip and the electrode plate, particles with longer dimension and high dielectric constant were attracted first into the highest electric field area; in this case, the end of the tip. Small and low dielectric particles slowly approach the highest field.

Consequently, the positive dielectrophoresis is used for attracting the nanotubes. Conversely, particles with lower dielectric constant than the medium move in the opposite direction indicating negative dielectrophoresis. According to this theory, in the case of an ac electric field, nanoparticles, such as carbon nanotubes, with the longest dimension have larger dipole moment. These are attracted and aligned to the tip before smaller particles such as carbon debris or other impurities. If long and thin nanotubes attach onto the apex of the tip, smaller particles that arrive at the tip later cannot completely cover the nanotube. If the assembling time is properly controlled, we can expect a protruding nanotube tip.

An apparatus for aligning a MWNT on the apex of an AFM tip is shown in Fig. 2. The apparatus consists of two parts, a processing part and an inducing part. The processing part consists of an Al electrode as a counter electrode and an AFM tip, which is fixed to the z axis translation stage for maintaining the distance from the counter electrode. This stage can be tilted at 0 to 90 ° in rolling and pitching direction to establish perpendicularity between the sidewall of an AFM tip's apex and the electrode. The coarse control of approaching the tip to the counter electrode was conducted through the magnifier. Moreover, by dropping a droplet of the MWNT solution at the desired spot and using the mag-

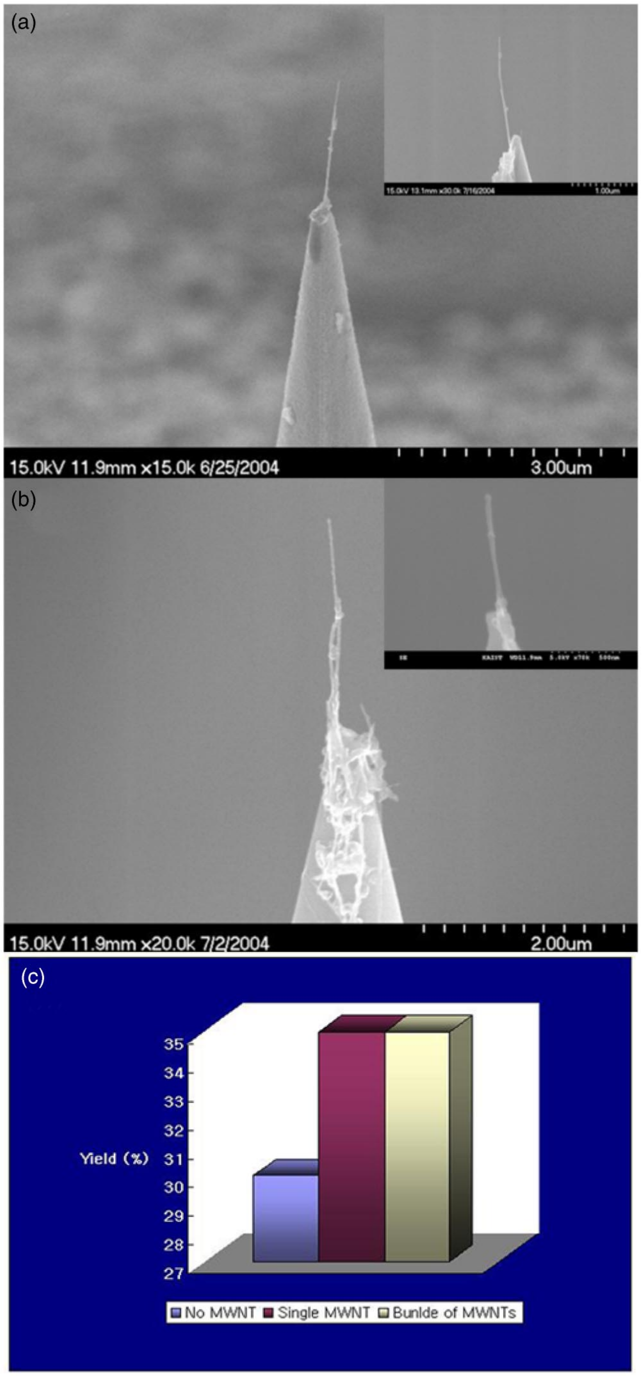

FIG. 4. (a) (Color online) FESEM images of CNT AFM tips with a single MWNT attached. These tips were fabricated under an ac electric field with 0.61 V_{pp}/μm at 5 MHz and, in the case of inner figure, 0.7 V_{pp}/μm at 5 MHz, 10 μm gap distance. (b) Some of tips have the bundle CNTs on the end of the tips. The contact area between the MWNTs and the tip was covered by impurities. It caused a strong bonding between the MWNT and the tip. (c) The distribution of the yield in this experiment is shown. CNT AFM tip with a single MWNT attached has a 35% yield.

nifier for our observations, we can control the angle between the sidewall of the tip and the electrode. The gap between the end of the tip and the surface of the Al electrode is controlled by a translation stage with micrometer resolution.

The inducing part of the apparatus is composed of a signal generator. The electric field can be generated by the ac component. Through the oscilloscope, the gap between the end of the tip and the surface of the Al electrode can be

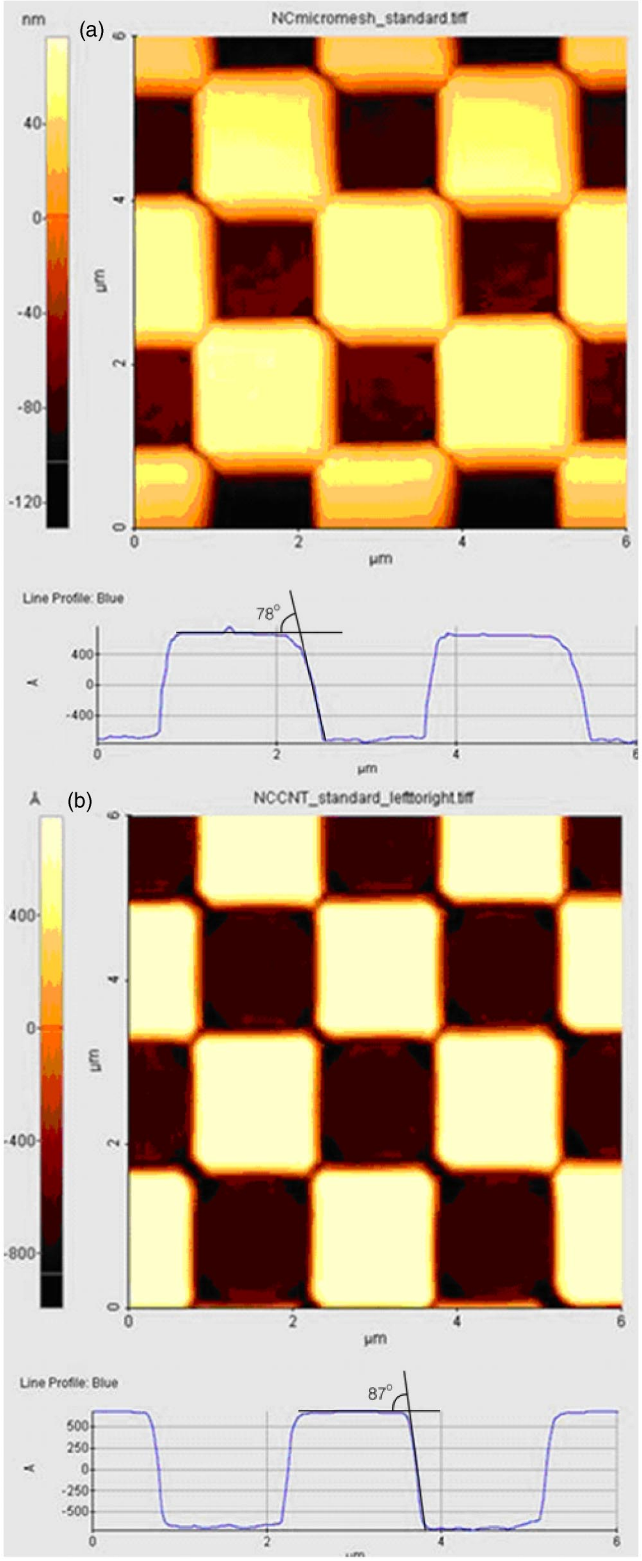

FIG. 5. (Color online) Noncontact mode AFM images and line profiles obtained with (a) bare tip (1.85 um width) and (b) CNT-attached tip (1.5 um width). As shown in the AFM images, the cross-sectional line profile measured with the bare tip deviates significantly from the real profile, whereas the profile measured with the CNT AFM tip with a single MWNT attached resembles that of the standard sample. By verifying the existence of the MWNT after scanning, we have demonstrated that the MWNT tip made by this method is able to be used as a measurement tip.

closely controlled. It is very important to determine the contact position between the AFM tip and the electrode, because we can measure the gap distance by setting the contact position as a reference point. If the distance is too close, numerous CNTs are likely to be attached on the end of the AFM tip. On the contrary, if the gap is too wide, the attraction force for the nanotubes will not be sufficient. From experiments it was found that the gap distance, which was determined based on an analysis of the electric field, should be controlled within 10 μm.

We experimentally investigated the tendency of the MWNT attachment according to ac or dc voltage only. When only dc voltage was applied, impurities were overwhelmingly deposited on the tip relative to the amount of deposited nanotube. Because impurities occupied almost the entire surface of the end of the tip, there is likely to be little or no space for the deposition of nanotubes. Moreover, the attached CNTs were not aligned parallel to the tip axis. Hence, we could not obtain a well-aligned nanotube tip. In contrast, when only an ac electric field was applied, even though impurities were deposited on the tip, MWNTs were attached on the end of the tip with a protruding shape.

Several factors for assembling the nanotube were considered in this experiment. We determined the assembling factors to be the gap distance, applied voltage, and frequencies. A protruding nanotube could form successfully at the tip end only when the applied ac voltage was larger than 5 V at a 10 μm gap and 5 MHz. When the gap distance was over 10 μm, the yield of the assembly was remarkably reduced. This indicates that the gap distance and the applied voltage are critical to attract the nanotubes. In terms of theoretical approach, these factors must be related to the magnitude of the dielectrophoretic force. When the frequency was varied from 100 kHz to 10 MHz, there was little difference in the yield. The experimental conditions are closely related with each other. The concentration of the MWNT solution affect on the MWNT's deposition. It is difficult to make a single CNT tip with the dense or sparse solution. As CNTs are aligned and attached so fast at the area where the electric field is induced on,[16] the quantity of a droplet of CNT solution does not greatly affect the making of a single MWNT attachment. From this fact, we can see that the CNT's alignment and attachment occurs before the CNT solution is dried out.

These electric and experimental conditions exceedingly influence the CNT's alignment and attachment. However, through the experiments, we found that the curvature size of the tip's apex bring out a single MWNT-attached tip. Figure 3(a) shows the AFM tip used in this experiment. It was coated with Al 40 nm in thickness such that the apex of the tip became slightly blunter than a bare tip. The shape of the tip remained conical with a curvature of 100–150 nm. MWNTs less than 10 μm length synthesized by an arc discharge were used in this study. The outer diameter of the MWNTs is in a range between 15 and 20 nm.

In Eq. (1), we can see that the dielectrophoretic force is proportional to the gradient of the square of the electric field, which means that CNTs are aligned and attracted to the area of the highest field intensity in proportion to the gradient of the electric field. The CNTs then stay in the area where the dielectrophoretic force is nearly zero, and the area where the dielectrophoretic force is zero is the area where the gradient of the electric field is zero. In the case of the AFM tip, this area is the apex of the tip. We expect the probability of a single CNT deposition to change through the size of the tip's curvature according to the CNT's diameter. Figure 3(b) presents the simulation result of the electric field. As shown in Fig. 3(b), the area where the dielectrophoretic force is zero exists at the end of the tip and this area is narrower than the real area. Because the area where the dielectrophoretic force is zero was so narrow in the very sharp tip, we expected there to be no room for a CNT to stay before depositing.

As shown in Fig. 4, MWNTs deposited at the end of the AFM tip was observed by field emission scanning electron microscopy (FESEM) and the contact area between the MWNT and the tip was covered by impurities. Figure 4(a) shows a FESEM image of MWNT-attached AFM tip in which MWNTs were connected to each other and the single of MWNTs were deposited on the apex of the tip. Figure 4(a) is under an ac electric field with 0.61 V_{pp}/μm at 5 MHz, and the inner figure shows the result from 0.7 V_{pp}/μm at 5 MHz. Figure 4(b) shows the CNT AFM tips with a bundle of CNTs. We obtained a 35% yield in a single CNT attachment. The distribution of the yield in this experiment was shown in Fig. 4(c).

In these experiments, we observed several phenomena. First, the orientation of the nanotubes was generally in parallel alignment to the tip axis, because the attached parts of the nanotube onto the end of the tip were guided according to the angle of the tip. We found that the angles of the obtained samples were in the range of $\pm 20°$ to the tip axis. Next, when a current of 10 to 20 μA was run through the nanotube (a tip would often be connected to an electrode due to the nanotubes), the nanotube was burned and shortened. With regard to the interaction between the nanotubes and the tip, van der Waals force might be the major cause of CNT deposition, as shown in other studies,[6,7] and additionally the impurities on the tip could help to firm attachment of the nanotube. Further, the maximum length of a protruding nanotube would always be less than the gap distance, because the part of the nanotube that was longer than the gap would be used to attach MWNTs on the tip area.

In this article, we found that the probability of single MWNT attachment on the AFM tip is improved by not only the appropriate electric conditions but also the curvature size of the tip's apex.

In order to demonstrate the effectiveness of the CNT-attached AFM tip in terms of topographic measurement, we obtained atomic force microscopy (model: XE-100, PSIA) images of a standard sample by utilizing two AFM tips; one is bare and the other is a CNT-attached AFM tip. The standard sample used in this measurement is a patterned silicon grating sample with rectangular lines of 142 nm height, 1.5 um width, and 1.5 um of interline spacing.

The measured AFM images of the sample are shown in Figs. 5(a) and 5(b), respectively. The AFM images were obtained by noncontact mode with a scan rate of 0.2 lines per second. As depicted in the AFM images, the cross-sectional line profile measured with the bare tip deviates significantly from the real profile, whereas the profile measured with the

CNT-attached tip resembles that of the standard sample. The lateral angle by CNT tip presents 87 deg, but normal tip shows 78 deg. Furthermore, the measured dimensions of the height and width of the rectangular line with the two different tips are significantly different. This indicates that the lateral resolution was markedly improved by utilizing the CNT-attached tip. The different scale value shows that the image by CNT tip is exceedingly clearer than that by normal tip. Moreover, by verifying the existence of the MWNT after scanning, we have demonstrated that the MWNT tip made by this method is able to be used as a measurement tip.

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