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Direct Observation of Field Emission Sites in a Single Multiwalled Carbon Nanotube by Lorenz Microscopy

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Emission sites were observed as bright spots near the tip end of a multiwalled carbon nanotube (MWNT) by means of Lorenz microscopy. The bright spots appeared above electric fields as electrons were emitted. A marked fluctuation was observed in the emission current above 20–30 μA, which was closely related to structural changes at the tip of the MWNT. The layers of the MWNT were peeled off during field emission and they functioned as the second emission sites for the concentration of electric field. [DOI: 10.1143/JJAP.44.1661]

KEYWORDS: multiwalled carbon nanotube, field emission, transmission electron microscopy, Lorenz microscopy

1. Introduction

Carbon nanotube (CNT) electron emitters are expected to have a range of applications as electron sources in high-resolution electron beam instruments.1,2 Recent experiments have confirmed that CNT electron sources have extremely high brightness.1,3 A high-brightness electron source is an important prerequisite for achieving high resolution in an electron microscope.

The stability of the emission current is important for realizing these electron beam instruments. However, it was reported by Bonard et al. that the current became highly unstable above 10 μA.4 Wang et al. reported that structural damage of the CNT occurred during field emission when the electric field was higher than 45 V/μm.5 The electric field is defined classically as \( E = V/d \), where \( V \) is the applied voltage and \( d \) is the distance from the tip of the field emitter to the surface of the counter electrode. With respect to the stability of the emission current, structural change at the tip of the CNT is an important subject in understanding the mechanism of the current fluctuation if the current exceeds 20–30 μA. Though it is expected that the emission sites are closely related to the tip form of the CNT, the relation between the emission sites and the tip form is not clear. The emission sites of the CNT are observed using field emission microscopy (FEM) and field ion microscopy (FIM).5,7 We can observe the pole figure of the electrons’ emitting direction and the surface atomic structures of the tip end of the CNT using FEM and FIM, respectively.

In this study, we have used a Lorenz microscopy method in transmission electron microscopy (TEM) operational mode to observe emission sites near the tip end of a single CNT while measuring the field emission currents. In this Lorenz microscopy method, we applied the defocus method in order to observe the electron emitting sites as bright spots. On the basis of the in situ observation of the emission sites and the tip form of a multiwalled carbon nanotube (MWNT), we clarified the mechanism of the instability of the emission current that appeared above 20–30 μA.

2. Experiments

The TEM specimen holder was designed as shown in Fig. 1(a) to apply voltages up to 1.0 kV across the cathode and the anode electrodes. The distance between these electrodes can be controlled from 0 to 5.0 mm via the mechanical movement of a micrometer. The distance was normally fixed at 500 μm, which was sufficiently large to...
avoid electric discharge during field emission. The TEM specimen holder was inserted into the field emission-TEM (Hitachi, HF-2000) operating at 200 kV, where no electron irradiation damages occurred in MWNT during TEM observation. The pressure in the specimen chamber was approximately 10^{-6} Pa during measurement. Arc-discharge grown MWNTs with diameters in the range from 10 to 30 nm were used in this study. A single MWNT was mounted on the tip of the tungsten needle using a piezoceramic manipulator and then the contact area between the MWNT and the needle was strengthened by metal deposition using the chemical vapor deposition (CVD) method in the scanning electron microscope (SEM) as shown in Fig. 1(b).\textsuperscript{8,9} The electrical contact resistance between the scanning electron microscope (SEM) as shown in Fig. 1(b).\textsuperscript{8,9} The electrical contact resistance between the MWNT and the needle was a few hundreds of kΩ, and the incident electron beam passing through the MWNT and its counterelectrode in the SEM after the metal deposition.

### 3. Results and Discussion

The emission sites near the tip end of the MWNT were observed in the underfocused condition, which is realized by focusing an objective lens below an in-focus imaging plane. Figure 2 shows a series of underfocused images obtained under increasing applied voltage and current. The defocus distance (ΔZ) is the distance from the in-focus position, which was set at 136 μm in the particular case of Fig. 2. A bright spot appeared at an electric field of 0.68 V/μm, where electrons were emitted. As the applied voltage increased, the bright spot appeared at the end of the MWNT tip with an increase in the emission current, while the spot was confirmed by applying a voltage across the MWNT and its counterelectrode in the SEM after the metal deposition.

The effect of the polarity of the applied voltage on the defocused image is shown in Fig. 3. Figures 3(a) and 3(b) are underfocused and overfocused images of the tip of the MWNT, respectively, taken while applying negative voltage to the MWNT tip. Figure 3(c) shows the bright spot appeared at the end of the MWNT tip at the underfocused condition as shown in Fig. 3(a). Figures 3(c) and 3(d) are underfocused and overfocused images of the end of the MWNT tip, respectively, taken while applying a positive voltage to the MWNT tip. In this case, the bright spot appeared in the overfocused condition as shown in Fig. 3(d). This result indicates that the incident electrons in the TEM passing through the MWNT are deflected in a direction that is dependant on the polarity of the applied voltage; the incident electrons are deflected in the outward or inward direction due to electrostatic repulsion or attraction. It is assumed that the bright spot appeared as a result of interference between the incident electron beam in the TEM deflected by applying voltages to the MWNT and the incident electron beam passing straight that remained unaffected by the electric field.

The relation between the defocus distance (ΔZ) and the radius of the bright spot at the tip end under a fixed condition of field emission was examined. A series of underfocused images taken during field emission with increasing ΔZ is shown in Figs. 4(a)–4(d). No changes were recognized while applying negative voltage in focus (ΔZ = 0), as is apparent in Fig. 4(a). The radius of the MWNT tip was measured to be 6.3 nm in Fig. 4(a). The Fresnel fringe contrasts along the longitudinal direction of the MWNT. Figure 4(e) shows the relation between ΔZ and the radius of the bright spot. The half values of the spot width in the lateral direction of the MWNT were plotted as a function of ΔZ in order to avoid overestimation due to the convolution of the bright spot and the Fresnel fringes. Defocusing alters the magnification of the image because of the change in the objective lens current. Revision of the magnification by means of defocusing was investigated by measuring Au particles of nanometer size. A linear relationship between ΔZ and the radius of the bright spot was obtained as shown in Fig. 4(e). The radius of the bright spot at ΔZ = 0 is measured to be 2.8 nm by extrapolation.

Figure 5 shows the relation between the voltage applied to the tip and the radius of the bright spot at ΔZ = 0 in another MWNT, whose tip radius was 7.0 nm. The radius of the
bright spot increased linearly as the applied voltage increased. This means that the size of the bright spot relates to the strength of the electric field at the end of the MWNT tip or the area where the electric field is concentrated.

We examined the effect of structural changes near the end of the MWNT tip on the stability of the emission current above a few tens of µA. Figures 6(a) and 6(b) show the changes in the in-focus images obtained during field emission. The emission current was rapidly increased from 15 to 30 µA when the layers of the MWNT were peeled off, as shown in Fig. 6(a). This structural change is similar to the damage process of a MWNT during field emission at electric fields higher than 45 V/µm as reported by Wang et al. 4) When the peel-off layers were removed as shown in Fig. 6(b), the emission current decreased suddenly from 30 to 18 µA. Figures 6(c) and 6(d) show the changes in the underfocused images obtained during field emissions above a few tens of µA. The emission current increased suddenly from 20 to 35 µA when a new bright spot appeared near the main bright spot, as shown in Fig. 6(c). When this new bright spot disappeared as shown in Fig. 6(d), the emission current decreased suddenly from 35 to 23 µA. This indicates that the peel-off layers shown in Fig. 6(a) may function as the second emission site for the concentration of the electric field. However, Wang et al. reported that a fluctuation in the emission current was due to the variation in the distance between the MWNT tip and the anode owing to a head-shaking effect of the MWNT during field emission. 5) A fluctuation in the emission current was reported to be very sensitive to the change of the gap between the MWNT and the anode because the gap was only 2 µm in their emission experiment, which is significantly smaller than the gap (500 µm) in our experiment. Moreover, we did not observe the head-shaking of MWNT in our TEM observation during field emission. We conclude that the marked fluctuation in emission current during field emission above 20–30 µA can be ascribed to structural changes in the tip of the MWNT.
4. Conclusions

A bright spot appeared in underfocused images of the tip of MWNT at electric field of 0.68 V/\(\mu\)m where electrons were emitted. The bright spot is assumed to be related to the emission site on the MWNT. The radius of the bright spot became larger linearly as the applied voltage is increased. This means that the spot size is related to the strength of the electric field at the end of the MWNT tip or the area in which the electric field is concentrated.

A marked fluctuation in the emission current above 20–30\(\mu\)A was observed, and is ascribed to structural changes of the tip of the MWNT. The layers of the MWNT were peeled off during field emission and they functioned as the second emission site for the concentration of the electric field.