Secondary electron emission from magnesium oxide on multiwalled carbon nanotubes

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We have investigated effects of electric fields on the yield of secondary electron emission (SEE) from the primary electron bombardment on magnesium oxide (MgO) covering vertically grown multiwalled carbon nanotubes (MWCNTs). We observe that the yield of SEE increases up to at least 22,000 at a special condition. The strong local field generated by the sharp tip of vertically grown MWCNTs accelerates secondary electrons generated by primary electrons. This eventually gives rise to so called Townsend avalanche effect, generating huge number of secondary electrons in a MgO film. Emission mechanism for such a high SEE will be further discussed with energy spectrum analysis. © 2002 American Institute of Physics. [DOI: 10.1063/1.1498492]

Secondary electron emission (SEE) with bombardment of primary electrons plays an important role in vacuum devices. For instance, electron multiplier, microchannel plate, and electron gun require the SEE materials with a high amplification yield. In general, insulators are good candidates for high SEE. The secondary electrons generated by the primary electrons in a magnesium oxide (MgO) film move to the surface with relatively weak electron-electron scatterings due to the absence of free electrons in insulator, and finally escape from the surface if they have enough energy to overcome the work function of the materials. Single crystal MgO, for instance, has a SEE yield of about 25 at best. On the other hand, the porous MgO produces high SEE yield of about 1000 under the high electric field. However, for a given yield, it is always desirable to look for the condition in which the lowest field is used. Recently a MgO film was deposited on randomly oriented carbon nanotube (CNT) powder, where relatively large SEE of maximum 15,000 at a backbias of 1400 V was obtained. Yet the SEE obtained was strongly dependent on the sample positions and not reproducible either. In this report, we introduce a systematic approach to reproduce high SEE using MgO on vertically grown multiwalled carbon nanotubes (MWCNTs). We observed an unusually high SEE yield of greater than 22,000 (beyond the limit of a detector) at a backbias of 850 V, which was strongly related to the MgO film thickness.

MWCNTs were grown by thermal chemical vapor deposition on the Ni-coated Si substrate using a C$_2$H$_2$ gas at 650 °C. The average diameter and length of the grown CNTs were 300 Å and 20 μm, respectively. A MgO thin film was deposited on the vertically grown MWCNTs using electron beam deposition. Figure 1(a) shows the scanning electron microscope (SEM) image of MgO-coated MWCNTs, where CNTs are encapsulated by MgO only on the top area. The inset shows a typical transmission electron microscope (TEM) image, where the thickness of MgO film at the sidewall is about 50–300 Å and the thickness at top of the CNT tip about 500–2000 Å, although the nominal thickness of MgO film from an electron-beam evaporator was 3000 Å. This suggests that relatively small amount of MgO was deposited on CNTs compared to the thin film on a flat surface.

Figure 1(b) shows a schematic diagram of our apparatus setup to measure the SEE. The MgO-coated CNT films were bombarded by the primary electrons ($I_p$) which generates the secondary electrons ($I_s$) in the MgO film. The $I_p$ was measured by applying a positive bias of 200 V to the sample holder in order to avoid the leakage current to the chamber wall. The $I_p$ was fixed at 233 nA during the experiment, while its kinetic energy was varied. The yield of secondary electron emission ($\delta$) is then defined as $I_s/I_p=(I_p+I_t)/I_p = 1+I_t/I_p$, where $I_s$ can be measured by the Faraday cup. In practice, $I_s$ can be easily obtained by measuring $I_p$ and $I_t$ (the current supplied from the substrate), instead. The negative backbias to the sample decelerates the incident primary electrons, while accelerating the secondary electrons to escape from the surface. In case of no primary electron, no secondary electrons are generated. Note that the electric field generated by the negative backbias is small such that no field emission currents are observed. The charge replenishment is achieved by the current through the sample from the $I_t$.

Figure 2(a) shows the yield as a function of net primary electron energy ($\Delta E$), which is defined by the energy difference $E_p-eV_t$, where $E_p$ and $V_t$ are primary electron energy

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and the negative bias applied to the substrate, respectively. The nominal MgO thickness was 3000 Å. The yield is relatively small for low $E_p$ over the whole range of $\Delta E$ up to 900 eV, whereas this value shows an abrupt increase near $\Delta E$ of 250 eV for the $E_p$ of 1000 eV. The existence of the threshold voltage suggests that the SEE mechanism may be related to an avalanche effect. With further increase of the $E_p$ the yield was too large to measure by our multimeter whose maximum current is limited to 5 mA. The yield does not increase with low $\Delta E$ and the maximum peak is observed near 200 eV. We measured the SEE by varying the thickness of the MgO film. Figure 2(b) shows the yield as a function of time with different nominal MgO film thicknesses at fixed $E_p$ of 1100 eV. The yield becomes stabilized after long measurement time. The very thin film of 1000 Å gives a low yield. The MgO film with a nominal thickness of 3000 Å gives rise to a maximum yield of 22,000, which is beyond the detection limit of our multimeter. The SEE yield drops at larger MgO thickness. This may indicate the existence of an optimum penetration depth. The TEM image, as shown in the inset of Fig. 1(a), shows the actual thickness of the MgO film of about 500–2000 Å on the top of the CNT. In general, the maximum yield is obtained when the penetration depth is about five times the escape depth in insulator. The escape depth of MgO is known as 60–200 Å, which implies the penetration depth of the MgO film to be 300–1000 Å. This is in good agreement with our observations of the TEM image. We emphasize that the MgO film without CNTs gives the yield of up to 800 at best at very high $E_p$ of 1500 eV and the CNTs without a MgO film gives a yield of less than one. Figure 2(c) shows the yield as a function of the backbias for fixed net $E_p$. The yield increases abruptly with increasing the backbias, indicating again an avalanche effect. The values of on-set backbias are large at low and large net $E_p$. The minimum on-set backbias exists at net $E_p$ of 250 eV.

We next measured kinetic energies of the emitted secondary electrons by an electron energy analyzer (VG Science, Clam IV). Figure 3(a) shows the distribution of the kinetic energy of the secondary electrons in terms of three different backbiases at fixed $E_p$ of 1000 eV. The small peak at 1000 eV, independent of the backbias, indicated by $K_1$, results from the elastic backscatterings of the primary electron.
The electrons generated at the MgO surface \((K_2)\) at a backbias of \(-800\) V are negligible within kinetic energies of \(800–1000\) eV. The secondary electrons start emitting from below \(800\) eV, where the sharp peak \((K_3)\) at \(800\) eV is attributed to the field emission. The broad peak \((K_4)\) from \(320\) to \(800\) eV is considered to originate from the SEE that are generated through the whole MgO film layer. This trend of broad energy distribution is very similar for other backbiases generated through the whole MgO film layer. This trend of broad energy distribution is very similar for other backbiases generated through the whole MgO film layer. The voltage drop across the MgO film is below \(800\) V, where the sharp peak \((K_3)\) at around \(320\) eV. The voltage drop across the MgO film is shown in the diagram of Fig. 3(b). Although the backbias of, for instance, \(-800\) V is applied between the chamber wall and the substrate, it is only \(-480\) V that is applied across the MgO film with a voltage drop of \(320\) V between the chamber wall and the MgO surface. The position of the on-set kinetic energy is dependent on the geometry, i.e., the separation distance between the MgO surface and the chamber wall.

In general, the electric field within MgO is considered to be small, since the dielectric constant of MgO \((\varepsilon = 9.8\varepsilon_0)\) is about ten times larger than that of vacuum. However, the electric field of MgO in this experiment may be enhanced under the bombardment of primary electrons due to the escape of secondary electrons of the surface from MgO. We now estimate the strength of the electric field across the MgO film. For instance, at a backbias of \(-800\) V, large secondary electrons are obtained as shown in Fig. 2(a). The potential drop across MgO film is considered to be about \(500\) V, i.e., corresponding to the voltage difference between \(K_3\) and \(K_5\) in Fig. 3(a). The average field strength is \(500\) V/d (\(500–2000\) Å) = \((2.5–10) \times 10^7\) V/cm, where the thickness (d) of the MgO film on top of the CNT was measured from the TEM image. This value is the same order of magnitude of the field strength corresponding to the on-set field of avalanche effect in typical metal oxides. This field may launch the cascade emission of secondary electrons, but still cannot explain such a high amplification factor. Thus, a higher back-bias is required to ensure the avalanche effect in an MgO insulator. We note that without CNTs such a high amplification factor could not be observed. This proves that the small diameter of CNTs enhances the local field strength near the tip of nanotubes, since the typical diameter of MWCNTs is about \(100–300\) Å. The field enhancement factor from a CNT film is usually an order of \(10^3\) and the field enhancement factor of a MgO coated CNT film was found to be \(700–1200\), from the \(I–V\) curve of the field emission experiment. Therefore, the local field near the tip can be enhanced by a factor of \(10^3\). This high local field will accelerate the primary electrons arrived near the tip and generate a cascade emission of secondary electrons. This process results in the charge depletion on the MgO surface and induces an addition local field across the MgO film. The charge depletion was replenished by the \(I–V\) curve. In order to have avalanche phenomenon to occur, the primary electrons should reach the MgO film near the nanotube tip and therefore optimum net energy for the primary electrons is expected, as observed from our results. Our data were fully reproducible with repetition of the measurements. The variance of the SEE yield over the entire sample was within \(10\%\).

In summary, we have investigated the yield of SEE from MgO/MWNTs. The SEE yield was measured for various MgO film thicknesses, primary electron energies, and backbiases. The SEE yield was achieved to be much greater than \(22,000\). The electron energy analysis revealed that this high yield originates from an avalanche phenomenon due to the strong local field generated by the sharp CNT tip. We emphasize that this high SEE is obtainable over the sample with good reproducibility. We expect our approach to be applied for various vacuum electronic devices.

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