Electroluminescence in diamond-like carbon films

S. B. Kim and J. F. Wager
Department of Electrical and Computer Engineering, Center for Advanced Materials Research, Oregon State University, Corvallis, Oregon 97331

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White electroluminescence (EL) was observed for the first time from diamond-like carbon (DLC) films at room temperature. Ac voltages in excess of 200 V were applied to a metal-insulator-semiconductor (i.e., DLC)-insulator-metal device structure to observe EL. At an applied voltage of 235 V, the brightness and efficiency were 0.5 fl and 0.002 lm/W, respectively.

Diamond-like carbon (DLC) films have been investigated for a variety of applications such as optical, wear resistant, and corrosion-resistant coatings. We are interested in employing wide optical band gap, amorphous DLC films as an active material for visible electroluminescent (EL) devices which operate at room temperature. We contend that DLC is a promising EL material for the following reasons:

1. DLC films can be deposited with optical band gaps wide enough to be potentially useful for visible applications ($E_{\text{opt}} > 2$ eV).

2. Luminescence in DLC is potentially efficient because of the absence of the $k$-conservation selection rule in an amorphous material.

3. Visible photoluminescence (PL) has been observed in wide optical band gap DLC films.

4. EL has been observed in a similar material: amorphous, hydrogenated silicon carbide, $\alpha$-Si$_x$C$_{1-x}$:H.  

5. Unlike ZnS, which is hygroscopic, DLC is relatively insensitive to moisture.

The purpose of this letter is to report the first observation of EL from a DLC active layer in the metal-insulator-semiconductor (i.e., DLC)-insulator-metal (MISIM) structure shown in Fig. 1. A glass slide substrate coated with indium tin oxide (ITO) and silicon oxynitride (the bottom insulator) was supplied by Planar Systems of Beaverton, Oregon. A 2000-Å-thick DLC active layer was deposited by plasma-enhanced chemical vapor deposition using a SemiGroup System 1000CC. Methane diluted in helium (10% CH$_4$ in He) was used as a source gas. The deposition rate was 17 Å/min using the parameters shown in Table I. The top insulator was Al$_2$O$_3$ which was deposited at room temperature by secondary-ion beam sputtering using a Veeco 3" Microtech System. The top electrode was evaporated Al.

White EL was observed at room temperature from this device when an ac voltage of $\approx 200$ V was applied between the ITO and Al electrodes. The ac drive voltage (3.5 kHz, 18% duty cycle) and light output voltage waveforms are shown in Fig. 2. The polarity dependence of the light output waveform is attributed to the asymmetry of the insulators in the MISIM structure. In contrast to the $\alpha$-Si$_x$C$_{1-x}$:H ac EL device which emits light only during voltage polarity changes, visible light is emitted from the DLC device during virtually the entire duty cycle. The DLC device is similar to the $\alpha$-Si$_x$C$_{1-x}$:H ac EL device, however, in that the decay time of the emission, measured at the trailing edge of the voltage pulse, is very fast ($\approx 10 \mu$s) in contrast to that of ZnS: Mn ($\approx 1$ ms).

The brightness-voltage ($B$-$V$) and efficiency versus voltage characteristics of the device are shown in Fig. 3 for a 6 kHz ac drive voltage with a 35% duty cycle. The white EL emission was dim but observable with the naked eye in a dark room when the drive voltage amplitude exceeded a voltage of $\approx 200$ V, the threshold voltage obtained from an extrapolation of Fig. 3. Using a dielectric constant value of 4 for DLC, 10 for Si$_3$O$_7$N$_4$, and 14 for Al$_2$O$_3$, the effective field in the DLC is estimated to be $6 \times 10^6$ MV/cm at threshold. DLC films deposited in the same manner as for this device exhibited breakdown fields of 1.2 MV/cm (defining breakdown as the field at which a current of 1 $\mu$A is measured using an electrode area of $4 \times 10^{-3}$ cm$^2$, i.e., $J = 0.25$ mA/cm$^2$). Thus, this device clearly operates in a breakdown mode. The $B$-$V$ curve exhibits a small amount of counterclockwise hysteresis. For voltages greater than approximately 210 V the EL efficiency and brightness increase linearly with applied voltage until catastrophic breakdown at approximately 245 V. The brightness and EL efficiency are 0.5 fl and 0.002 lm/W, respectively, at an applied voltage of 235 V. These values are very low.

The EL and PL spectra are compared in Fig. 4. The EL peak is located at an energy about 0.4 eV lower than the PL peak. This shift in the luminescent spectrum is typical of amorphous materials. The abrupt cutoff of the PL spectrum is a consequence of using an Ar laser, with an energy of 2.54 eV, for photoexcitation.

![FIG. 1. Structure of the DLC ac EL device.](image)

<table>
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<th>TABLE I. Summary of the parameters used for the DLC deposition. Structures were placed on a powered electrode. The diameter of the powered electrode was 12.7 cm.</th>
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<tr>
<td>Power density: 175 mW/cm$^2$</td>
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<td>Substrate temperature: $\sim$ 250$^\circ$ C</td>
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<tr>
<td>Chamber pressure: 800 mTorr</td>
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<td>Electrode spacing: 34 mm</td>
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<tr>
<td>Frequency: 13.56 MHz</td>
</tr>
</tbody>
</table>

The brightness-frequency (B-F) characteristic is illustrated in Fig. 5 for a fixed voltage of 240 V. The brightness increases in almost a linear fashion over the entire range of frequency investigated (up to 6 kHz). To accomplish this measurement, the pulse width was held constant at 30 µs such that an increase in frequency corresponds to an increase in the duty cycle. Preliminary analysis suggests that it is the duty cycle rather than the frequency which leads to the observed increase in brightness. A similar increase in brightness with frequency has been observed in ZnS:Mn devices but the issue of the duty cycle was not discussed.

In summary, the potential of DLC as an EL material has been experimentally demonstrated by the fabrication of a DLC EL device which emits broadband, visible EL. The luminescence intensity and efficiency of this device are presently very low. Further work is required to improve the luminescent quality of the DLC films.

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