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High efficiency tandem organic light-emitting devices with Al/WO_3/Au interconnecting layer

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An interconnecting layer of Al (2 nm)/WO_3 (3 nm)/Au (16 nm) was studied for application in tandem organic light-emitting devices. It can be seen that the Al/WO_3/Au structure plays the role of an excellent interconnecting layer. The introduction of WO_3 in the connection unit significantly improves the device efficiency as compared to the case of Al/Au. Thus, the current efficiency of the two-unit tandem devices is enhanced by two factors with respect to the one-unit devices. The green two-unit tandem device of indium tin oxide/MoO_3/4,4′-*N*,*N*′-bis[*N*-(1-naphthyl)-*N*-phenylamino]biphenyl(NPB)/tris(8-hydroxylquinoline) aluminum (Alq_3):10-(2-benzothiazolyl)-1,1,7,7-tetramethyl-2,3,6,7-tetrahydro-1*H*,5*H*,11*H*-[1]benzopyrano[6,7,8-ij]quinolizin-11-one (C545T)/Alq_3/LiF/Al/WO_3/Au/MoO_3/NPB/Alq_3:C545T/Alq_3/LiF/Al showed a maximum current efficiency of 33.9 cd/A and a power efficiency of 12.0 lm/W. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2787877]

Rapid advances in organic light-emitting devices (OLEDs) and their applications in displays and lighting imposed substantial demands for OLED structures having improved both brightness and efficiency without increasing driving current.[1,2] Recently developed tandem structure OLEDs consisting of multiple electroluminescent units connected vertically in series have shown double enhancement in brightness and efficiency.[3–6] In tandem devices, the interconnecting layer between two emissive units plays an important role in device performance. Up to now, many interconnecting layers in tandem OLEDs have been tried, such as Mg:Ag/indium zinc oxide,[6] Cs:2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline (BCP)/indium tin oxide (ITO),[7] Cs:BCP/V_2O_5,[8] tris(8-hydroxyquinoline) aluminum (Alq_3):Li/$FeCl_3$:4,4′-*N*,*N*′-bis[*N*-(1-naphthyl)-*N*-phenylamino]biphenyl (NPB) and 1,3,5-tri(phenyl-2-benzimidazole) benzene:Li/$FeCl_3$:NPB,[9] Mg:Alq_3/WO_3,[10] Bphen:Li/MoO_3,[11] Cs:BPhen/tetrafluorotetracyanoquinodimethane (F_4-TCNQ):NPB,[12] Li_2O,[13] etc. However, it can be noted that the fabrication of these connecting units typically requires sputtering or coevaporation processes, and high temperature deposition and/or delicate handling care is necessary, somewhat inevitably complicating the fabrication process. Although recently undoped interconnecting layers such as Al/Au (Ca/Ag) (Ref. 14) and copper-hexadecafluoro-phthalocyanine/copper phthalocyanine,[15] which can be fabricated by simple thermal evaporation, were studied, the transmittance over a wide wavelength range perhaps remains a problem. Therefore, it is very necessary to develop interconnecting layers with high transmittance and simple processing.

In this letter, we report an interconnecting layer which consists of Al (2 nm)/WO_3 (3 nm)/Au (16 nm). High electroluminescence (EL) efficiency was obtained using the interconnecting layer in tandem OLEDs. It can be noted that the introduction of WO_3 between Al and Au significantly enhances the EL efficiency with respect to the use of Al/Au connecting layer. The current efficiency of two-unit tandem OLED is enhanced by two factors with respect to the one-unit device. As shown, the metal/metal oxide/metal structure can be easily fabricated by simple thermal evaporation and has excellent optical and electrical properties, and good ability to inject electrons and holes into the top and the bottom units, as well as high transmittance. Our results indicate that metal/metal oxide/metal should be a good structure for interconnecting layer in the fabrication of high efficiency tandem OLEDs.

Figure 1 shows the schematic diagrams of the tandem OLEDs used in this study. The one-unit OLED is also given in Fig. 1. In this tandem device, the two emissive units used here are identical and consist of MoO_3/NPB/Alq_3:10-(2-benzothiazolyl)-1,1,7,7-tetramethyl-2,3,6,7-tetrahydro-1H,5H,11H-[1] benzopyrano [6,7,8-ij] quinolizin-11-one (C545T)/Alq_3. All devices were fabricated on ITO coated glass with a sheet resistance of 10 Ω/□, and a thermally deposited LiF/Al was used as the cathode. ITO substrate was cleaned and treated by O_2 plasma. The deposition was carried out at a pressure less than 3×10^{-4} Pa without vacuum breaks. The organics and metal oxide were evaporated at the rate in a range of 0.2–0.3 nm/s, and the metals were evaporated at the rate of 0.8–1 nm/s. The devices have an emissive area of 16 mm^2. In order to measure the transmittance, Al (2 nm)/WO_3 (3 nm)/Au (16 nm)/MoO_3 (8 nm) and Al (2 nm)/Au (16 nm)/MoO_3 (8 nm) films were evaporated on a glass substrate. The current-voltage-luminance characteristics were recorded using a computer controlled sourcemeter

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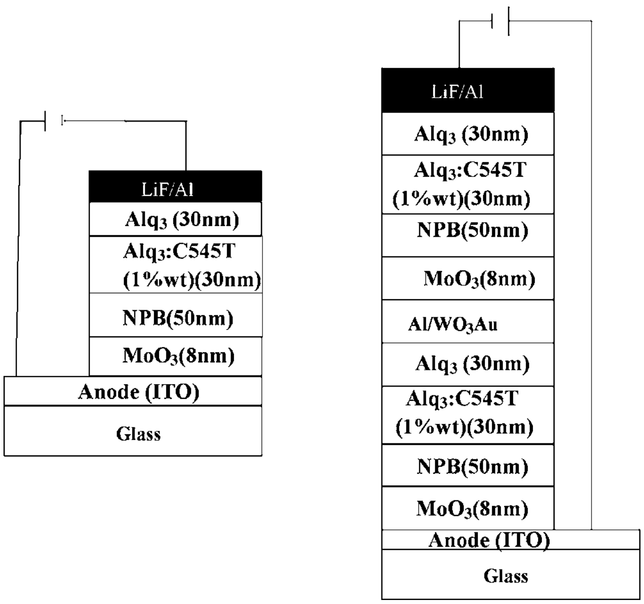

FIG. 1. Schematic diagram of one-unit and tandem devices.

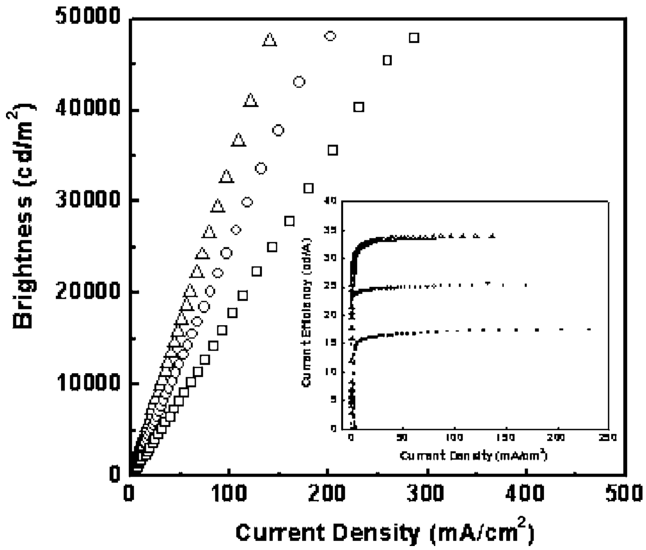

FIG. 3. Current density–luminance characteristics of Al/WO_3/Au tandem device (up triangle), Al/Au tandem device (circle), and one-unit device (square). The inset gives the current density–current efficiency characteristics of Al/WO_3/Au tandem device (up triangle), Al/Au tandem device (circle), and one-unit device (square).

(Keithley 2400) and multimeter (Keithley 2000) with a calibrated silicon photodiode. Here, the emission pattern was assumed to be Lambertian. The EL spectra were measured by JY SPEX CCD3000 spectrometer. The transmittance was measured by Shimadzu UV-3600. All the measurements were carried out in ambient atmosphere at room temperature.

Figure 2 shows the current density–voltage–luminance (*J*-*V*-*L*) characteristics of two-unit tandem devices with Al/WO_3/Au and Al/Au interconnecting layers and one-unit control device. As expected, the driving voltage of tandem devices is about twice that of the control device at the same current or luminance. For example, at 800 cd/m^2, the driving voltages of Al/WO_3/Au and Al/Au tandem devices are about 10.2 and 9.4 V, respectively, whereas that of the control device is 5.4 V. Furthermore, it is found that the driving voltage is reduced in our tandem devices as compared to the previously reported similar tandem OLED.[14] This reduction should be related to the utilization of MoO_3 buffer layer on ITO and Au top. In tandem devices with Al/WO_3/Au interconnecting layer, the turn-on voltage is 3.5 V (defined at 1 cd/m^2) and the maximum luminance reaches 48000 cd/m^2 at 19.5 V bias.

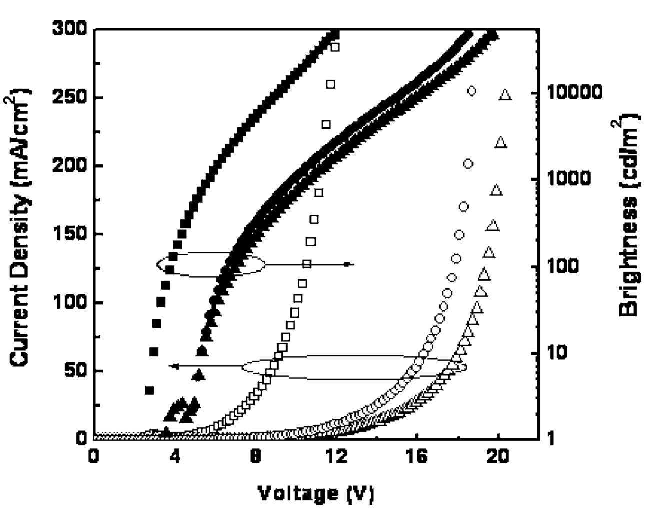

FIG. 2. Current density-luminance-voltage characteristics of Al/WO_3/Au tandem device (uptriangle), Al/Au tandem device (circle), and one-unit device (square).

As shown in Fig. 3, at any current density, the luminance of the Al/WO_3/Au tandem device is about twice that of the one-unit device and higher than that of the Al/Au tandem device. At current density of 20 mA/cm^2, the luminance arrives at 6700 cd/m^2 for the Al/WO_3/Au tandem device, which is higher than 5000 cd/m^2 for Al/Au tandem device, and much higher than 3200 cd/m^2 for one-unit device. This means that the Al/WO_3/Au tandem device emits higher luminescence efficiency. The inset of Fig. 3 shows the current efficiency versus current density curves of the tandem devices with Al/WO_3/Au, Al/Au, and one-unit devices. It can be clearly seen that the current efficiency of the Al/WO_3/Au tandem device is twice that of the one-unit device and higher than that of the Al/Au tandem device. At 20 mA/cm^2, the current efficiency of the Al/WO_3/Au tandem device reaches 33.02 cd/A, whereas the current efficiencies, respectively, arrive at 24.4 and 16.05 cd/A for the Al/Au tandem device and the one-unit device. It can be seen that although higher driving voltage is needed in tandem devices, the power efficiency is still improved with respect to one-unit devices. The power efficiency of tandem devices reaches about 12 lm/W, which is higher than 8.9 lm/W of one-unit devices. These results indicate that Al/Au and Al/WO_3/Au are excellent interconnecting layers in tandem devices and the introduction of WO_3 results in the interconnecting layer more effectively.

Figure 4 shows the EL spectra of the Al/WO_3/Au tandem device and the one-unit device in normal direction. Al/Au tandem device also shows similar EL spectrum. All devices emit green light at about 525 nm, but tandem devices with Al/WO_3/Au and Al/Au connecting layers show narrower spectrum with a full width at half maximum (FWHM) of 42 nm, while the one-unit control device shows a FWHM of 69 nm. The narrowing phenomenon is obviously due to the microcavity effect. However, it can be clearly seen from the inset of Fig. 4 that there is hardly any spectral deviation with viewing angles in the Al/WO_3/Au tandem device with

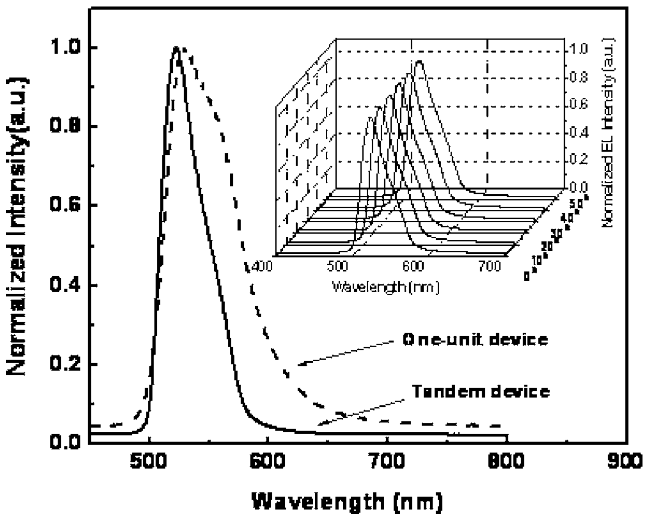

FIG. 4. EL spectra of Al/WO_3/Au tandem device and one-unit device. The inset is the EL spectra of Al/WO_3/Au tandem device at different viewing angles. The Commission Internationale de l'Eclairage chromaticity coordinates are (0.238, 0.672), (0.236, 0.675), (0.238, 0.673), (0.231, 0.669), (0.230, 0.668), and (0.229, 0.673) at viewing angles of 0°, 10°, 20°, 30°, 40°, and 50°, respectively.

respect to the Al/Au tandem device, where the small deviation between spectra at different viewing angles is observed. Obviously, the WO_3 layer also plays an important role in reducing the angle dependence of spectrum caused by microcavity effect.

We attributed the enhancement in EL performance of Al/WO_3/Au tandem devices to the improvement of transmittance of Al/WO_3/Au with respect to Al/Au. Figure 5 shows the transmittance spectra of Al/WO_3/Au/MoO_3 and Al/Au/MoO_3. It can be seen that the Al/WO_3/Au interconnecting layer exhibits better transmittance at wide visible wavelength ranges compared to the Al/Au interconnecting layer. This should be important in the improvement of device performance. Furthermore, Al/WO_3/Au may be considered as a capacitor, and generated charges should lead to more

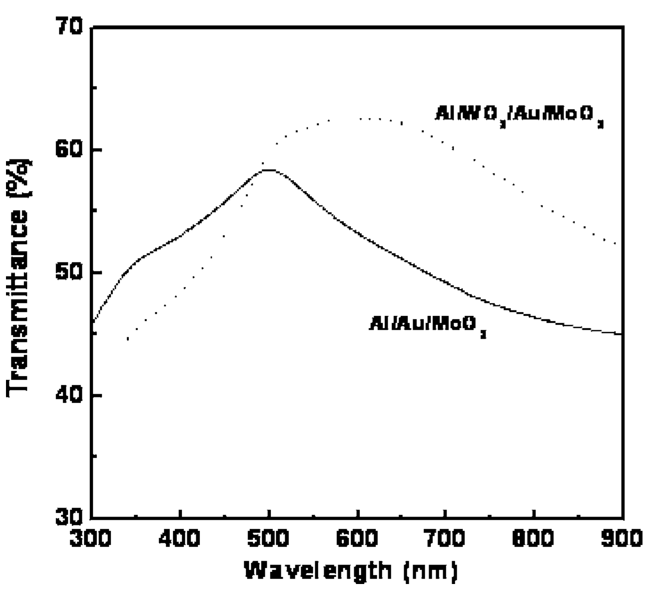

FIG. 5. Transmittance spectra of Al/WO_3/Au/MoO_3 and Al/Au/MoO_3.

holes and electrons to inject into top and down unit devices at bias condition of the electric field assisted bipolar charge separation. Therefore, the efficiency is further enhanced due to the formation of more excitions in Al/WO_3/Au tandem device. The detailed improvement mechanism investigation is underway.

In summary, an effective interconnecting structure for high efficiency tandem OLEDs is reported. The connecting structure is composed of a thin metal oxide sandwiched between Al and Au metals. It is demonstrated that such a connecting structure allows more electrons and holes to be effectively injected into two adjacent emitting units, and the utilization of metal oxide between Al and Au significantly improves the transmittance of the interconnecting layer. Thus, the device efficiency is significantly enhanced and the angular dependence of spectrum caused by the microcavity effect is also greatly eliminated. Furthermore, such a connecting structure can be easily fabricated by simple thermal evaporation, greatly simplifying device processing. It is also easily used to fabricate color-tunable OLEDs by independently changing the external bias, thus realizing high-resolution, independently addressable, stacked red-green-blue pixels for the application of color displays.

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Dr. Hongmei Zhang was listed twice in the byline. The second byline for Dr. Zhang should have been as a footnote stating that she was visiting The University of Hong Kong during the work. Dr. Wallace C. H. Choy was inadvertently omitted from the original byline. Also, a footnote with an e-mail for Dr. Choy was omitted. The correct listing for the authors is shown above.

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