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Citation: Applied Physics Letters **87**, 253501 (2005); doi: 10.1063/1.2147730
View online: http://dx.doi.org/10.1063/1.2147730
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/87/25?ver=pdfcov
Published by the AIP Publishing

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APPLIED PHYSICS LETTERS **87**, 253501 (2005)

# Highly efficient white organic electroluminescent devices based on tandem architecture

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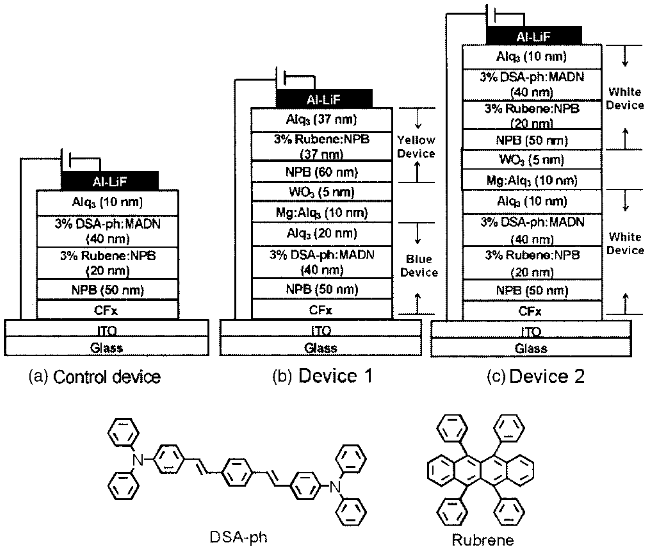
(Received 6 July 2005; accepted 9 November 2005; published online 12 December 2005)

Two types of tandem organic light-emitting diodes (OLEDs) with white-light emission have been developed by using $Mg:Alq_3/WO_3$ as the interconnecting layer. While the Commission Internationale d'Eclairage (CIE) coordinates of the tandem device with individual blue- and yellow-emitting OLEDs was sensitive to the viewing angle and the operating time, the tandem device connecting two white-emitting OLEDs was considerably less. At an optimal $WO_3$ thickness of 5 nm, the tandem two-unit device produced three higher luminance efficiency than that expected of a single-unit device. A maximum efficiency of 22 cd/A was achieved by the tandem device comprised of two white-fluorescent OLEDs, and the projected half-life under the initial luminance of 100 cd/m$^2$ was over 80 000 h. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2147730]

Organic light-emitting devices have attracted a great deal of attention as they possess attributes that are superior to many of today's mainstream displays and illumination sources. They can emit various colors by using a wide selection of organic fluorescent or phosphorescent dyes. Various green, blue, and red organic light-emitting diodes (OLEDs) with high luminance efficiency, low-power consumption, and long operational life have been demonstrated, and their commercialization is well underway. Recently, white OLEDs (WOLEDs) have also attracted considerable attention due to their applications in the "maskless" fabrication process of large-area full color displays by coupling with color filters, or simply by using them as backlight for liquid crystal displays and paper-thin light sources. For large-scale applications, WOLEDs are a high-quality, low-cost, thin-film, flexible option, superior to inorganic light-emitting diodes (LEDs) or fluorescent lamps.

White-light emission requires the mixing of two complementary colors or three primary colors. Various device structures and research for generating white light have been reported.[1–3] Doping the light-emitting layers with various fluorescent or phosphorescent dyes has been widely used since the first demonstration by Tang and co-workers at Kodak. Careful control of the location of exciton-recombination zone and/or the energy-transfer between the host and dopant molecules is needed to obtain a balanced white emission.[4,5] Although at the present, the phosphorescent OLEDs have been demonstrated to achieve white emission with high efficiency, the operating reliability still requires further research.[6] Recently, another elegant solution was provided by Matsumoto *et al.*[7] who, in IDMC'03, reported a stacked OLED by connecting individual blue and red devices in series to achieve pink emission with CIE$_{x,y}$ of 0.36, 0.23. It is expected that various hues can be obtained by this kind of tandem device without worrying about the potential shift of the recombination zone, which leads to undesired emission color. Furthermore, Kido found that the luminance efficiency of the tandem device with $N$ units is usually $N$ times as high as that of the single-unit device. This stratagem seems to be attractive for getting highly efficient WOLEDs. However, few articles discuss the influence of design principle on the devices' performance and stability.

In this work, two types of tandem WOLEDs containing an interconnecting layer of $Mg:Alq_3/WO_3$, have been fabricated and investigated in detail. $Mg:Alq_3/WO_3$ thin film which has low absorption in the visible region.[8] It is suitable to be the interconnecting layer for WOLEDs. The architectures of the two kinds of tandem WOLEDs are shown in Fig. 1 in which the white emission was obtained by mixing complementary blue and yellow colors. Device 1 was obtained by connecting blue and yellow devices in series, while Device 2 stacked two white-emitting devices with the same blue and yellow dopants as used in Device 1. The objective of this research is to obtain a tandem WOLED with high



(a) Control device (b) Device 1 (c) Device 2

FIG. 1. Schematic architectures of the devices and the structures of the dopants.

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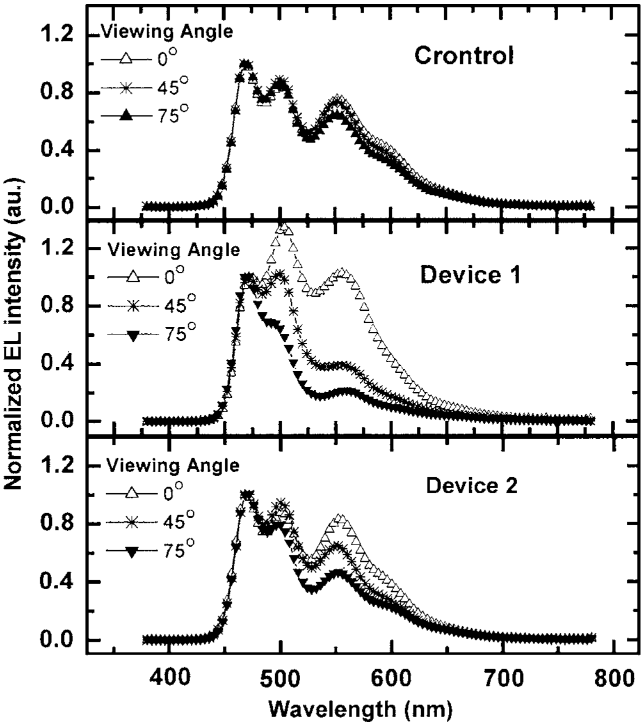


FIG. 2. Normalized EL spectra of the control and tandem devices at various viewing angles.

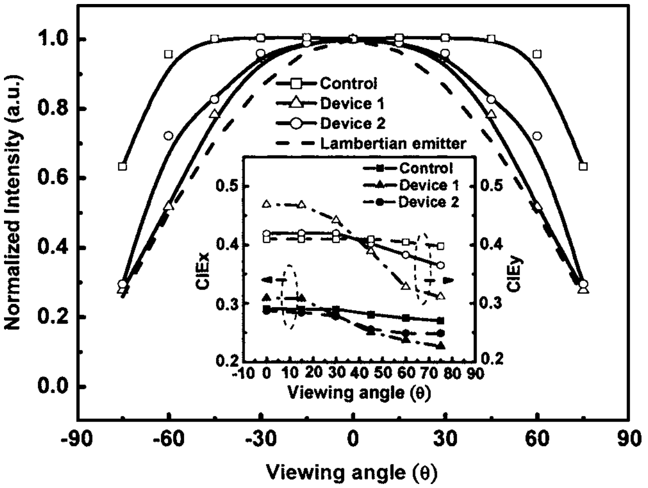


FIG. 3. Normalized EL intensity vs viewing angle. Inset: 1931 $\mathrm{CIE}_{x,y}$ coordinates vs viewing angle. All data were measured at 20 $\mathrm{mA/cm^2}$.

efficiency and broadband emission, and to gain some insight in understanding the relationship between the device structure and characteristics, which is expected to aid the future design and fabrication of tandem WOLEDs.

The substrates used in the present experiments were indium tin oxide (ITO)-coated glass, and the thickness and sheet resistance of the ITO layer were 75 nm and 35 $\Omega/\mathrm{sq}$, respectively. Prior to deposition of organic compounds, the substrates were treated with oxygen plasma of 200 W for 30 s. Then organic materials were deposited by thermal evaporation in an ULVAC SOLCIET coater at a base vacuum of $10^{-7}$ Torr. The OLED architectures of the control and two tandem devices are shown schematically in Fig. 1 for comparison, in which the fluorescent dopants for blue and yellow are *p*-bis(*p*-*N*,*N*-di-phenyl-aminostyryl)benzene and 5,6,11,12-tetraphenylnaphthacene (rubrene), respectively. The Mg:$\mathrm{Alq_3}$ layer was co-evaporated from separate heating sources at the same rate of 1 Å/s. All devices were hermetically sealed under nitrogen prior to testing in ambient. The active area of the electroluminescence (EL) device, defined by the overlap of the ITO and the cathode electrodes, was $3\times3$ $\mathrm{mm^2}$. The current density ($J$)-voltage ($V$)-luminance characteristics of the devices were measured with a Photo Research PR650 spectrophotometer and a computer-controlled programmable dc source (Keithley 2400). The stability tests of these devices were carried out under a nitrogen atmosphere.

White emission is observed from these devices when operated in a continuous dc bias with ITO at positive polarity. Figure 2 shows the EL spectra of these three devices at various viewing angles. The EL spectrum of the control WOLED, which covers a wide range of the visible region with three peaks at 470, 500, and 550 nm corresponding to the emissions from DSA-Ph and rubrene, respectively,[9] stays essentially the same at different viewing angles. On the contrary, the relative intensities of peaks at 500 and 550 nm in tandem devices decreased by increasing the viewing angle. This phenomenon is not expected to result from shifting of the recombination zone because these devices were all driven at a fixed current. We attribute the spectrum change to the optical interference or weak microcavity effect in multilayer devices, which has been reported in stacked OLEDs.[10] Furthermore, the EL spectrum of Device 1 revealed a more acute change with the viewing angle than that of Device 2. This may be due to the fact that the blue and yellow units in Device 1 differed considerably in optical length and emissive position.[11] Emission with long wavelength is easy to escape from the normal direction. We conclude that by stacking two white-emitting OLEDs, it is possible to obtain a high-efficiency tandem white device with acceptable angular dependency characteristics.

Figure 3 shows the normalized EL intensity versus the viewing angle characteristics of the control and tandem devices. It is apparent that the emissive intensity of the tandem devices is more sensitive to the viewing angle than that of the control device. The tandem devices revealed the emissive profiles are approximately the same as the Lambertian emitter. The inset of Fig. 3 shows the relationship between the

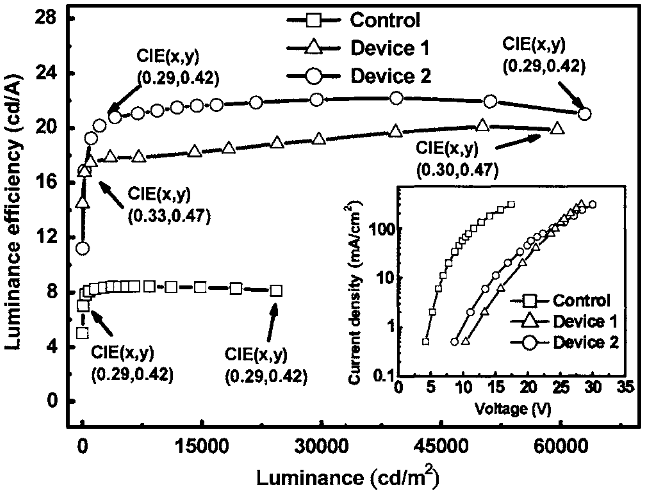
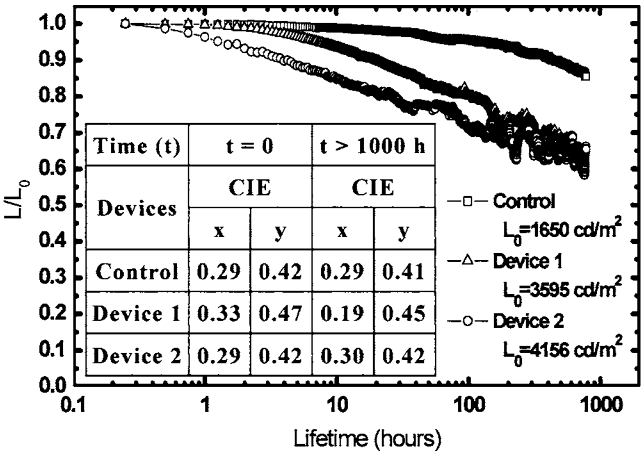


FIG. 4. The luminance efficiency vs current density characteristics of the control and tandem devices made in the same lot. Inset: $J$-$V$ diagrams of the control and tandem devices.



| Time (t) | t = 0 | | t > 1000 h | |
|---|---|---|---|---|
| Devices | CIE | | CIE | |
| | x | y | x | y |
| Control | 0.29 | 0.42 | 0.29 | 0.41 |
| Device 1 | 0.33 | 0.47 | 0.19 | 0.45 |
| Device 2 | 0.29 | 0.42 | 0.30 | 0.42 |

FIG. 5. Comparison of the operational stability of the control and tandem devices at 20 mA/cm$^2$. Inset table: The CIE$_{x,y}$ coordinates of the control and tandem devices at initial and after testing for 1000 h.

CIE$_{x,y}$ coordinates of the emission of these WOLEDs and the viewing angle. The shifts in CIE $x$ and $y$ coordinates of the control device from the viewing angle of 0° to 75° are 0.021 and 0.012, respectively. Compared to the control device, the CIE$_{x,y}$ coordinate shifts of Device 2 increase only slightly when the viewing angle is larger than 30°. However, it is apparent that the color of Device 1 with shifts of CIE $x$ and $y$ coordinates of 0.082 and 0.158, is more sensitive to the viewing angle than that of Device 2. According to our observation of the angular dependence of intensity and color, the microcavity effect is present in these tandem devices.

Figure 4 shows the luminance efficiency versus luminance characteristics of these WOLEDs, which were measured normal to the device surface. The maximum luminance of tandem devices reaches beyond 60 000 cd/m$^2$. The $J$-$V$ characteristics of the control and the tandem devices are shown in the inset of Fig. 4. As expected, the driving voltage increases with the increasing number of active units. The driving voltages (at 20 mA/cm$^2$) of the control, and Devices 1 and 2 are 7.8 V, 19.1 V, and 16.9 V, respectively. Device 2 exhibited the highest efficiency of 22 cd/A, which was almost three times that of the control device. Under different levels of brightness, i.e., various current densities, no current induced quench and CIE coordinate shift were observed in the control device and Device 2. This signifies that the recombination zone was near the interface of the blue- and yellow-emitting layer, and there was no significant shift of the emissive zone in the devices.[12] Therefore, Device 2 appeared to display the nature of its constitutor, i.e., the control device. An interesting amplification effect was also observed in Device 2, in which three times the efficiency of the control WOLED could be achieved by just connecting two devices. This presumably is due to the microcavity effect which enhances the amount of light emitted in the forward direction as shown in Fig. 3. In our previous study, a two-unit green device had been observed to produce four times the amplification.[13] Therefore, it is important to have a good optical design for producing better light extraction from the device.

The operational stability of these devices, tested under the condition of constant current density of 20 mA/cm$^2$, is shown in Fig. 5. Different decay trends were observed in the three devices. Device 2 was the least stable, while the control device showed the longest half-life ($t_{1/2}$). This may be due to the fact that Device 2 suffered more driving power ($J \times V = 0.382$ W/cm$^2$) than the control (0.156 W/cm$^2$) and Device 1 (0.338 W/cm$^2$). Thermal breakdown processes may be present in these tandem devices due to the nonohmic contact of the interconnecting layer.[14] However, assuming the scalable law of Coulombic degradation for driving at $L_0$ of 100 cd/m$^2$, the half-life ($t_{1/2}$) of Device 2 is projected to be greater than 80 000 h. The inset of Fig. 5 also shows the CIE coordinates of the control device versus Devices 1 and 2. It is evident that the color of Device 2 almost remained the same after 1000 h while that of Device 1 due to the different decay rate of blue and yellow devices changed dramatically.

In summary, two tandem organic white-emitting diodes have been developed by using Mg:Al$q_3$/WO$_3$ as the interconnecting layer. In these tandem devices, the emissive intensity and color are dependent on the viewing angle. The microcavity effect may be one of the dominant reasons for the angular dependence effect. So, it is important to have a good optical design for the tandem device. However, the tandem device made by stacking two white-emitting OLEDs could produce devices with high efficiency, good color reliability, and sufficient operating stability. The stratagem seems favorable for obtaining a white tandem device with an efficiency as high as 22 cd/A and acceptable angular dependency characteristics.

This work was supported by the MOE Program for Promoting Academic Excellence of Universities under Grant No. 91-E-FA04-2-4-B. The research grant of Industry/Academia Cooperation Project provided by Chunghwa Picture Tubes Ltd. is gratefully acknowledged.

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