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Real-Time Color-Tunable Electroluminescence From Stacked Organic LEDs Using Independently Addressable Middle Electrode

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Abstract—Independently controllable stacked organic light emitting devices (OLEDs) are fabricated by using interconnecting electrode of Al(2 nm)/WO₃(3 nm)/Au(16 nm) for connecting two primary color OLED units of blue and red. The middle electrode simultaneously functions as the cathode and anode for the bottom and top units respectively with a feature of over 60% optical transmission in a wide wavelength range from 500 to 700 nm such that the color can be tuned in real time from red to blue by changing the bias voltage to the two units. The undistorted primary colors and high efficiency have been obtained through optimizing structure and properly arranging the ordering of the blue and red subpixels.

Index Terms—High optical transmission electrode, intermediate electrode, real-time color tuning, stacked organic light-emitting devices (SOLEDs).

I. INTRODUCTION

ORGANIC light-emitting devices (OLEDs) have gained interest for their promising applications in full-color flat panel display [1]–[3]. For making such full-color displays, schemes such as multiple-emitting layers [4], microcavity structure [5], [6], voltage-controlled dye doping [7], and inserting carrier blocking layers [8] for achieving color tunability have been proposed. Among the methods, vertically stacked red, green, and blue-emitting OLEDs have led to a threefold increase in resolution and display fill factor as compared to the other traditional side-by-side subpixel arrangement [4]. The emission from a subpixel transmits through the vertically adjacent units in the independently addressable stacked OLEDs (SOLEDs), thus the high transparency of the interconnecting electrodes with appropriate carrier injection properties are necessary. Some attempts have been made on exploring high-performance interconnecting electrodes. For the independently controllable SOLEDs, the interconnecting layer should simultaneously function as an anode for one unit and as a cathode for another unit besides the necessity of good transmission. To realize

the functions, at present a semitransparent metal layer or a transparent indium-tin oxide (ITO), such as, Mg:Ag–indium tin oxide (ITO) [9], CuPc–ITO [10], and LiF–Ca–Ag [11], are generally used. However, there may be potential thin-film damage by using sputtering of ITO. Meanwhile, the typically low transmittance of the metal electrodes may limit the improvement of the device performance. Therefore, it is desirable to develop the intermediate controllable metal electrodes with the targets of not only improving the transmission but also enhancing the efficiency of the SOLEDs; however, the studies on the high transmission metal electrodes are limited.

In this letter, we shall use controllable middle electrodes of Al(2 nm)/WO₃(3 nm)/Au(16 nm) to realize color-tunable independently voltage-controlling SOLEDs. Besides the good transmission of the middle electrodes of above 60% over a wide wavelength range from 500 to 713 nm with a peak value of 62.5%, the metal Au layer on top of WO₃ layer serves as the hole-injection contact to the top unit and the thin metal Al serves as the electron injecting contact for the bottom OLED. The MoO₃ treatment of the Au layer surface can provide a good hole injection. As a consequence, each emitting unit in the SOLED can be independently addressable such that the color of pixel output can be continuously tuned from red through blue in real time. The undistorted primary color and high efficiency have been obtained through optimizing the structure and properly arranging the ordering of the blue and red subpixels. The efficiency of each emitting unit is close to the measured efficiency of the corresponding single-unit red and blue OLEDs.

II. EXPERIMENT

Fig. 1(a) shows the schematic diagrams of SOLEDs used in this letter. In the SOLEDs, the two emissive units consist of ITO/MoO₃(8 nm)/NPB(100 nm)/DSA-Ph:MADN(40 nm)/Alq₃(10 nm)/LiF(1 nm)/Al(2 nm)/WO₃(3 nm)/Au(16 nm)/MoO₃(5 nm)/NPB(60 nm)/Alq₃:DCJT(30 nm)/Alq₃(30 nm)/LiF(1 nm)/Al(150 nm). Apart from sequencing red top and blue bottom units as the SOLED, the red and blue units were also exchanged to study the effect of device sequence. All devices were fabricated on indium tin oxide (ITO) coated glass with a sheet resistance of 10 Ω and thermally deposited LiF–Al was used as a cathode. ITO substrate was cleaned and treated by O₂ plasma. The deposition was carried out at 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

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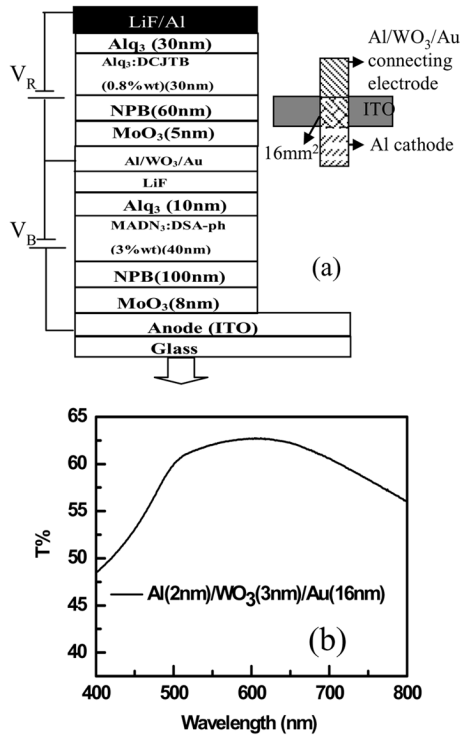


Fig. 1. (a) Schematic diagram of independently voltage controllable SOLEDs. (b) Transmittance spectrum of Al(2 nm)/WO₃(3 nm)/Au(16 nm).

an emissive area of 16 mm². The current-voltage-brightness characteristics were recorded using a computer-controlled sourcemeter (Keithley 2400) and multimeter (Keithley 2000) with a calibrated silicon photodiode. The EL spectra were measured by JY SPEX CCD3000 spectrometer. All the measurements were carried out in ambient atmosphere at room temperature.

III. RESULTS AND DISCUSSION

The SOLED is operated by common grounding the middle Al-WO₃-Au electrode as shown in Fig. 1(a). The top Al electrode and bottom ITO electrode are biased negatively and positively (V_R and V_B), respectively, with respect to the Al-WO₃-Au electrode. The output spectra from the SOLED at various drive voltages applied to the red and blue units are measured through the glass substrate and shown in Fig. 2(a). The top unit (red light), bottom unit (blue light), and corresponding (red and blue) single-unit control devices were biased at 11, 7, 10, and 6 V, respectively, the peak wavelength of the red (621 nm) and blue (461 nm) units match their corresponding single-unit control OLEDs as shown in Fig. 2(b). The device emission spectrum can be varied by a linear combination of the red and blue colors through independently varying the driving voltage of each unit at the ratio of the drive voltage (V_R/V_B) without the formation of exciplex [3]. Fig. 2(a) clearly shows that the emission spectrum from each unit in the SOLED can be tuned independently. For the blue and red emission as shown in Fig. 2(b), the full-width at half-maximum (FWHM) of the red and blue emission spectra from SOLED is 65 nm (red) and 58 nm (blue) with respect to the corresponding single-unit control device of 70 nm (red) and 52 nm (blue) without any

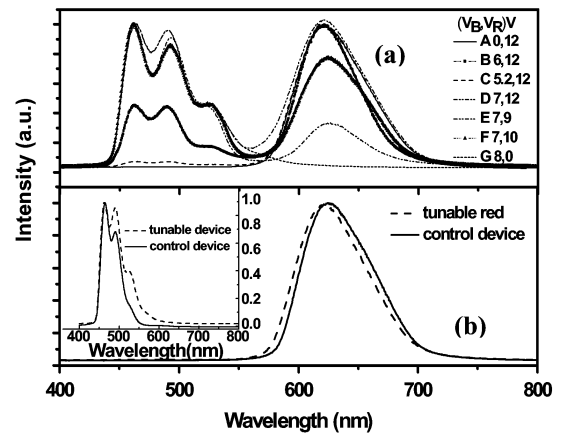


Fig. 2. (a) Emission intensity versus wavelength under various operating voltages at normal direction. (b) The emission spectra of the red light unit in the SOLEDs and the corresponding control devices biased at 11 and 10 V, respectively. The inset is the emission spectra of the blue light unit of the SOLEDs and the corresponding control devices biased at 7 and 6 V, respectively.

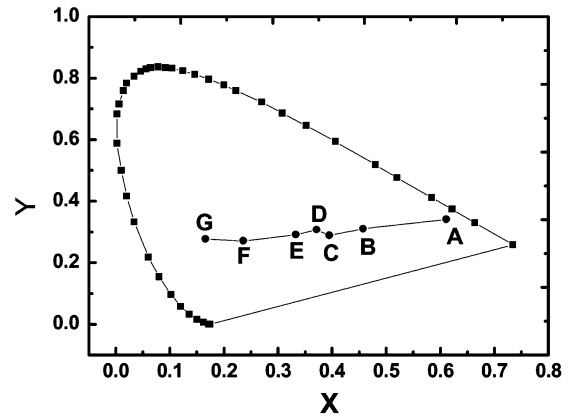


Fig. 3. The CIE color coordinates of the spectra in Fig. 2(a) with conditions A–G stated in Fig. 2(a).

significant change or obvious microcavity effect [12]. Fig. 3 shows the Commission Internationale de l’Eclairage (CIE) diagram of color tunable SOLEDs which indicates the continuous color tuning between the colors offered by the individual units in real time.

Fig. 4 shows the current efficiency versus current density curves of the red and blue emission from SOLEDs, where each unit was biased individually in the measurement. The performance of the bottom red unit and top red unit of the two sequence of SOLEDs and the control device with the same structure as the top red unit have been studied. The inset of Fig. 4 shows the blue devices. It is clearly seen that the current efficiency of each unit from SOLEDs [7.2 cd/A (red) and 3.5 cd/A (blue)] is close to that of the corresponding single-units red (7 cd/A) and blue (4 cd/A) OLED, when the red OLED is arranged as the top unit of the SOLED. In fact, the blue OLED should be arranged as the bottom unit because the blue OLED as the bottom unit has higher efficiency, as shown in the inset of Fig. 4, and the current efficiency of the red-emitting device has a minor change. Regarding the sequence of the blue and red units in SOLEDs, the transmission properties of the Al-WO₃-Au has to be investigated, as shown in Fig. 1(b). It is clearly shown that

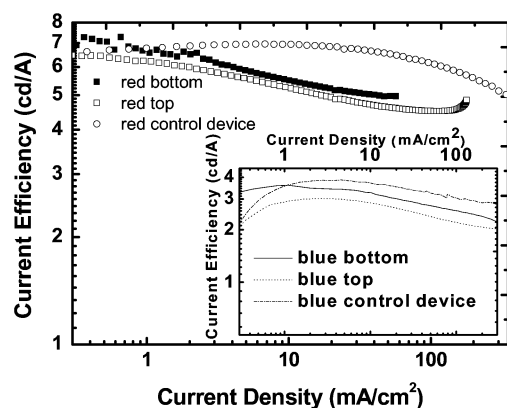


Fig. 4. The current efficiency versus current density characteristics of red light unit (top and bottom) in the two sequences of SOLEDs with the structure: NPB(60 nm)/Alq₃:DCJTB(30 nm)/Alq₃ (30 nm) (top) and NPB(100 nm)/Alq₃:DCJTB(30 nm)/Alq₃(30 nm) (bottom); The inset is that of the blue light unit (top and bottom) in the two sequences of SOLEDs with the same structure: NPB (60 nm)/MADN: DSA-ph(3%) (40 nm)/Alq₃ (10 nm).

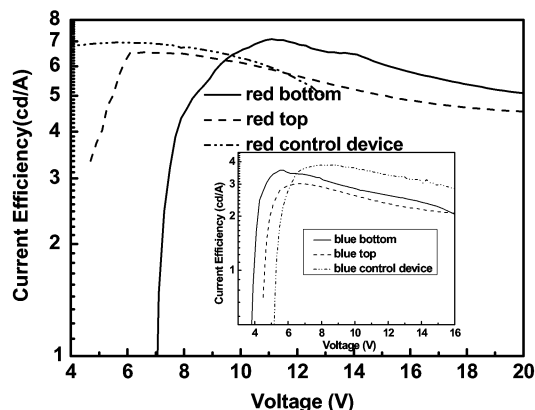


Fig. 5. The current efficiency versus applied voltage characteristics of red light unit (top and bottom) with the same structures as in Fig. 4. The inset is that of the blue light unit (top and bottom) with the same structures as in Fig. 4.

the Al-WO₃-Au structure exhibits a good transmittance in the green-red region of 500 to 700 nm of over 60%, favoring the red OLED as the top unit of the SOLED with lower absorption loss. The low transmittance of $\sim 52\%$ at 450 nm makes the efficiency of blue OLED as the top unit of SOLED reduces (see the inset of Fig. 4), apart from the possible reabsorption of blue light in the red bottom unit in the blue-top and red-bottom SOLED. Furthermore, in many SOLEDs, the interconnecting layer is used to generate electrons and holes at an electric field. It is typically considered that the electrons are injected into bottom device and the holes are injected into top device, thus playing the role of charge generation layer. Here, Al-WO₃-Au may be considered as a capacitor, since a layer of WO₃ between Al and Au layers serves as a dielectric layer. There is no direct contact between the Au and Al; thus the positive charges exist at the Au anode and the negative charges exist at the Al cathode for assisting the injection of the carriers. As a result, the efficiency is further enhanced due to the formation of more excitons in Al-WO₃-Au SOLEDs.

Fig. 5 and the inset figure show the current efficiency versus driving voltage curves of the red and blue emission from SOLED and the control device with the same arrangement as in Fig. 4. It can be seen that although the applied voltage is higher, the current efficiency of bottom red emission unit is slight higher than that of the upper unit and the control device. Besides, as shown in the inset of Fig. 5, the driving voltage of the blue bottom unit is lower than that of the blue top unit and the control device. As a result, putting the blue OLED as the bottom unit is reasonable for providing better performance.

IV. CONCLUSION

In summary, an effective interconnecting structure for high efficiency SOLEDs is reported. The connecting structure is composed of a thin metal oxide sandwiched between Al and Au metals. It is clearly seen that such a connecting structure permits effective electrons and holes injection into the two adjacent emitting units. The utilization of metal oxide between Al and Au provides the interconnecting layer an over 60% transmittance in a wide wavelength range from 500 to 700 nm, for a better transmission of red light from the blue-bottom and red-top SOLED. Furthermore, such a connecting structure can be easily fabricated by thermal evaporation for achieving real-time color-tunable SOLEDs through independently changing the biases to the two units, thus realizing high-resolution, independently addressable, stacked red-green-blue pixels for application of color displays.

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