



# Tunable full-color emission of two-unit stacked organic light emitting diodes with dual-metal intermediate electrode

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## ABSTRACT

The color-tunable stacked organic light-emitting devices (SOLEDs) with independently-addressable intermediate Al/Au electrode are examined. High brightness color-tunable SOLED can be achieved by properly optimizing the Al/Au electrode and using appropriate electron- and hole-injection layer. Full-color emission is achieved in a simple device structure with features of (1) only two-unit SOLED, and (2) the intermediate electrode is the Al/Au dual metal layers which are easy to process and have no damage to the organic layer.

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## 1. Introduction

Organic light-emitting device (OLED) is one of the promising technologies for lighting and display applications. In order to achieve full-color emission, complicated techniques have to be used to accurately fabricate three OLED structures for red, green, and blue emission in small areas. It will be highly desirable if a simple light-emitting device can emit light with a wide range of color, continuously tunable by changing the voltage. It may greatly simplify the fabrication techniques of multi- and full-color devices.

Voltage-controlled color tuning can be realized in a single unit OLED and has been reported in the devices using small molecules [1–3] and polymers [4–6]. We have reported a voltage-controlled color-tunable OLED based on a europium complex recently, in which the emitting color is tunable from red-to-blue or other designable color [7]. Another concept of voltage-controlled color tuning is the use of stacked structure. In this technique, the vertically stacked OLED (SOLED) is independently addressable and each emits different color to the others through the adjacent transparent organic layers, the transparent contacts and the glass substrate, allowing the entire device area to emit any mixture of the colors.

However, in the SOLED, the semitransparent intermediate electrodes are made from a thin metal layer and a transparent indium-tin oxide (ITO) thin film for electron and hole-injection, respectively [8–10]. ITO deposition usually requires magnetron sputtering process which is not compatible to the conventional thermal evaporation process of OLED devices. More importantly,

it may cause damage to the underlying organic thin films. In the full-color SOLED [10], the three OLED units require two intermediate transparent electrodes, thus the process of device fabrication is even more complicated. Another issue for the three-unit SOLED is the increased complexity [11,12] in driving circuit as compared with that of the two-unit one due to the fact that the three-unit series-connected OLEDs in the stack cannot be common-ground referenced in the driving circuit. Therefore, intermediate electrodes with simple fabricating process that can minimize the damage to the organic layers, and methods to achieve full-color emission in a simpler structure in term of processing and driving are worthy of exploring.

The electrical and optical properties of Al/Au thin film as the intermediate electrode in color-tunable SOLED are examined in this report. A bright red-green multicolor SOLED is demonstrated to convince its role of being an efficient intermediate electrode. Finally, we show that by combining the concepts of voltage-controlled color tuning in single unit OLED and stacked structure with Al/Au intermediate electrode, full-color emission is achieved in a two-unit stacked device.

## 2. Experiment

The molecular structures of the organics used in this study are presented in Fig. 1. The small molecules used in this study include a hole-transport and emitting material of *N,N'*-bis(2-naphthalenyl)-*N,N'*-bis(phenylbenzidine) (NPB), an electron-transport and emitting material of aluminum tris (8-hydroxyquinoline) (Alq<sub>3</sub>), a red triplet emitter Tris[1-phenylisoquinolino-C<sup>2</sup>,N]iridium(III) (Ir(piq)<sub>3</sub>), an europium complex of europium (dibenzoylmethanato)<sub>3</sub> (bathophenanthroline) (Eu(DBM)<sub>3</sub>bath), a electron-transport

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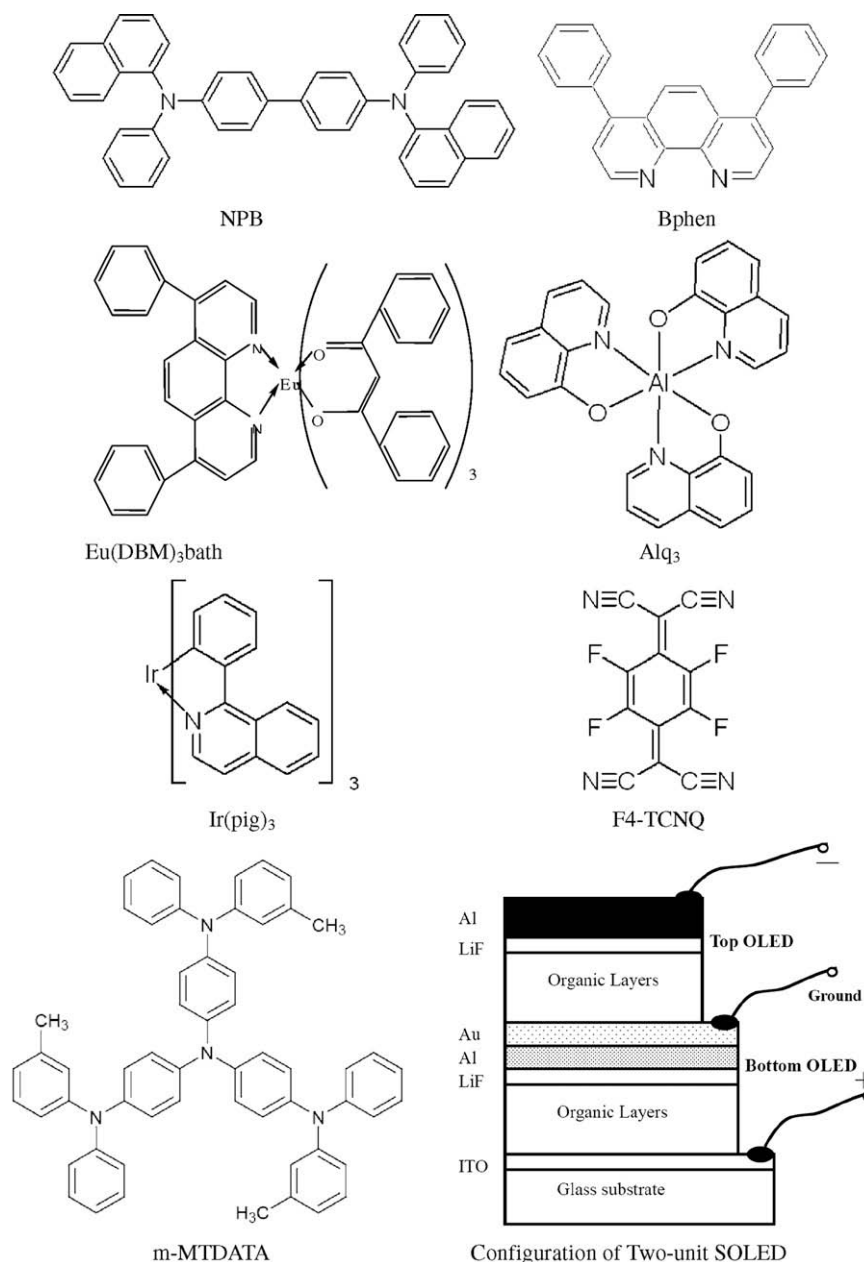


Fig. 1. The molecular structure of the organics used in this study and the device configuration of the two-unit SOLED.

material of 4,7-diphenyl-1,10-phenanthroline (BPhen) [13], a hole-injection and transport material of 4,4',4''-tris(3-methylphenyl)phenylamino triphenylamine (m-MTDATA), and an electron acceptor of 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F4-TCNQ). Fig. 1 also shows the device configuration of the two-unit SOLED. In this stacked device, the top and bottom OLED emit different colors and can be electrically controlled independently with the intermediate Al/Au layer acting as a common electrode. The ITO glass substrates with a sheet resistivity of  $20 \Omega/\square$  were cleansed as discussed in [14]. The organic layers of the bottom OLED, the intermediate electrode, the organic layers for the top OLED and the cathode metal for the top OLED were successively deposited by thermal vacuum evaporation at a pressure of  $10^{-4}$  Pa, with carefully designed shadow masks to confine the emitting area. The current–voltage characteristics were measured with a Keithley 2400 SourceMeter. The emitting spectra, brightness and color coordinates of the devices were measured with a PR650 SpectraScan.

### 3. Results and discussion

#### 3.1. Optimization of Al/Au dual films as an intermediate electrode

Al and Au are the mostly used metals in OLED that show no obvious damage to the organic layer. Al is favorable for electron-injection and Au is favorable for hole-injection. Thermal evaporated Al/Au dual metal layers were shown to be an effective intermediate layer for highly efficient SOLED [15]. This implies that Al/Au thin film could be a candidate for intermediate electrode in color-tunable SOLED. It should be noted that the Al/Au dual layer in Ref. [15] is an intermediate layer but not an intermediate electrode because in the SOLED there is no need to control the device through the intermediate layer. However, in color-tunable SOLED, the intermediate electrode is electrically controlled independently. The electrical and optical property of the intermediate electrode, such as electron-injection ability, hole-injection ability, surface conductivity and film transparency, should be considered as a

whole. The feasibility of Al/Au films to be an intermediate electrode is still need to be carefully examined. Here we use simple OLED structures to examine and optimize Al/Au dual films as an intermediate electrode.

The electron-injection ability of Al/Au film is found to be sensitive to the thickness of Al layer as investigated in the device of ITO/NPB(50 nm)/Alq<sub>3</sub>(55 nm)/LiF(0.5 nm)/Al/Au(10 nm). The dependence of current–voltage characteristic and current efficiency on Al thickness is shown in Fig. 2. At Al thickness of 70 nm, the device shows current efficiency of 3.0 cd/A, which is the typical result of NPB/Alq<sub>3</sub> OLED, however the Al/Au film is not transparent because of the thick Al layer. The transparency of Al/Au film increase to above 50% across all the visible range as the Al thickness is reduced to 8 nm, but the efficiency decrease to 1.0 cd/A and the driving voltage dramatically increase indicating that electron-injection of the Al/Au electrode become inefficient in this case. At the Al thickness of 16 nm, the transparency is around 30% across visible range, the efficiency is kept at an acceptable value of 2.2 cd/A, and the driving voltage is much lower than that of 8 nm Al. Thus the thickness of Al layer is kept at 16 nm for all of the following experiments.

The hole-injection ability of the Al/Au electrode is optimized in the device of Al(16 nm)/Au (or Au/hole-injection buffer layer)/NPB(55 nm)/Alq<sub>3</sub>(55 nm)/LiF(0.5 nm)/Al(70 nm). A comparison of the *I*–*V* characteristics and efficiencies at different Au thickness and with/without the hole-injection buffer layer are shown in Fig. 3. As shown in the figure, the device with 7 nm Au layer shows low efficiency of 0.1 cd/A and driving voltage of higher than 18 V. The efficiency is increased to 0.5 cd/A with the 10 nm Au layer and the driving voltage is reduced apparently. Further increasing the Au thickness to too thick will increase the possibility of device failure due to short circuit. However, a further increased efficiency and dramatically reduced driving voltage are achieved by inserting a thin layer of hole-injection buffer layer m-MTDATA or a layer of F4-TCNQ doped m-MTDATA [16] between Al/Au and NPB layer. Especially for the device having the F4-TCNQ doped m-MTDATA layer the efficiency is 2.3 cd/A, which is highly acceptable considering that the transparency of the metal anode is only about 30%, and the driving voltage is low and comparable to the device using ITO as the anode. In Fig. 3 the continuous left-shift of the *I*–*V* curve with consistent increasing efficiencies indicate improved hole-injection and thus more balanced electrons and holes in the optimization efforts.

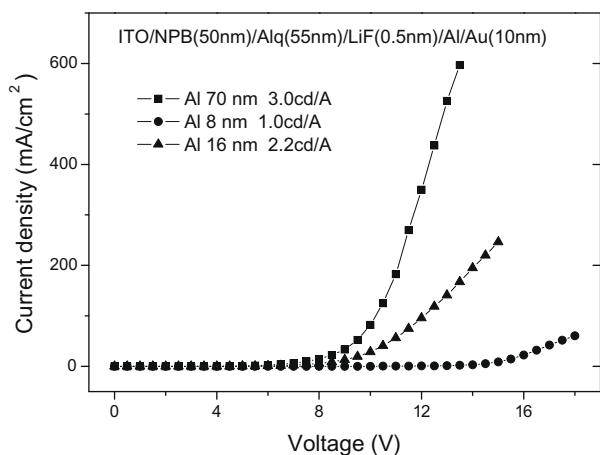


Fig. 2. The current–voltage characteristics and current efficiencies of the OLED with the structure of ITO/NPB(50 nm)/Alq<sub>3</sub>(55 nm)/LiF(0.5 nm)/Al/Au(10 nm) at different Al thickness.

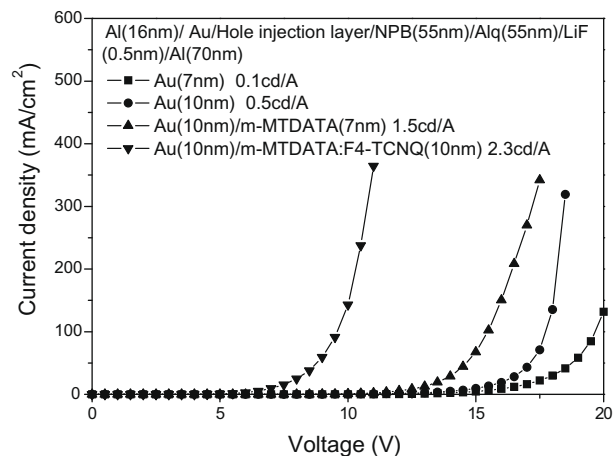


Fig. 3. The current–voltage characteristics and current efficiencies of the OLED with the structure of Al(16 nm)/Au (or Au/hole-injection buffer layer)/NPB(55 nm)/Alq<sub>3</sub>(55 nm)/LiF(0.5 nm)/Al(70 nm) at different Au thickness and using different hole-injection buffer layer.

Based on the above results, we conclude that the bilayer of Al/Au metal films with proper thickness can act well as both cathode and anode with LiF as the electron-injection buffer layer and F4-TCNQ doped m-MTDATA as the hole-injection buffer layer. The transparency is around 30% across the visible wavelength range. Four-point probe measurement shows that the sheet resistivity of the dual metal film is 15 Ω/□, which is low enough to be an electrode layer in OLEDs.

### 3.2. Bright multicolor-tunable SOLED using Al/Au films as an intermediate electrode

The performance of Al/Au as the intermediate electrode in the color-tunable SOLED is demonstrated in a multicolor device (SOLED1): ITO/NPB(40 nm)/NPB:Ir(piq)<sub>3</sub>(30 nm, weight ratio 10:1)/Bphen(20 nm)/Alq<sub>3</sub>(40 nm)/LiF(0.5 nm)/Al(16 nm)/Au(10 nm)/m-MTDATA:F4-TCNQ(7 nm, weight ratio 50:1)/NPB(50 nm)/Alq<sub>3</sub>(60 nm)/LiF(0.5 nm)/Al(70 nm), in which the bottom is an OLED emitting red color from the triplet emitter of Ir(piq)<sub>3</sub> and the top is an OLED emitting green color from Alq<sub>3</sub> layer. The current–voltage and luminance–voltage behaviors of the bottom and top OLED are shown in Fig. 4. The efficiencies are 5.5 cd/A and 2.3 cd/A at the current density of 20 mA/cm<sup>2</sup> for the bottom and top OLED, respectively. The brightness of higher than 10000 cd/m<sup>2</sup> is achieved in

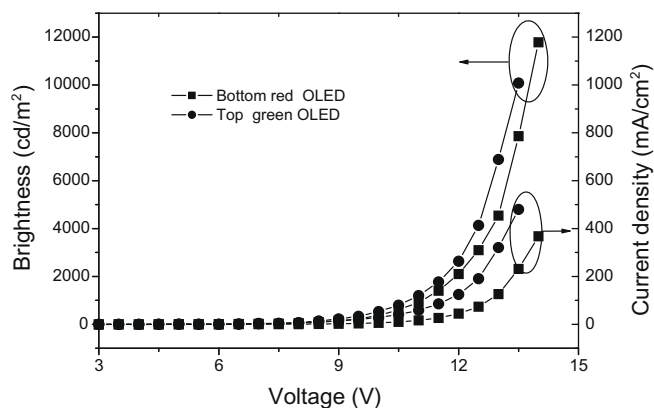
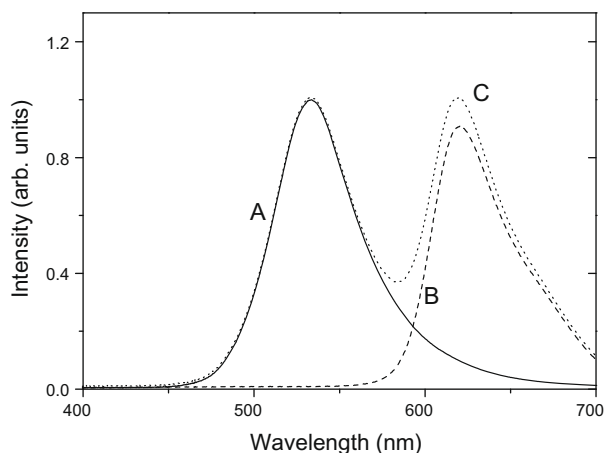


Fig. 4. The current–voltage and luminance–voltage behaviors of the bottom and top unit in SOLED1 with the structure of ITO/NPB/NPB:Ir(piq)<sub>3</sub>/Bphen/Alq<sub>3</sub>/LiF/Al/Au/m-MTDATA:F4-TCNQ/NPB/Alq<sub>3</sub>/LiF/Al.

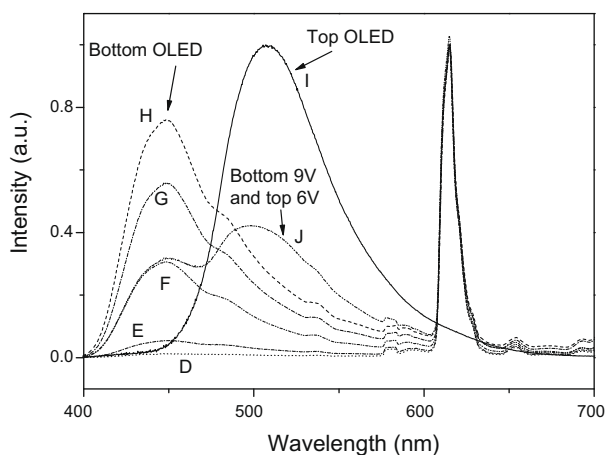


**Fig. 5.** The emitting spectra of SOLED1. (A) (solid line), the EL spectrum of the top OLED; (B) (dash line), the spectrum of the bottom OLED; (C) (dot line), the spectrum when both top and bottom OLED are biased at 5 V.

both bottom and top OLEDs at driving voltage of lower than 14 V. The emitting spectra of SOLED1 are shown in Fig. 5. The electroluminescent (EL) spectrum of the top OLED peaks at 533 nm and shows a full width at half maximum of 55 nm, which is narrower than that of Alq<sub>3</sub> emission spectrum in the conventional OLED due to the microcavity effects [17] in the SOLED structure. The bottom OLED emitting red color from the triplet emitter of Ir(piq)<sub>3</sub>. A superimposed EL spectrum is obtained when the two units of SOLED1 are biased at the same time. With independent driving the SOLED can exhibit any mixed color of the bottom and top OLEDs.

### 3.3. Full-color tunable emission in a two-unit SOLED

In a two-unit SOLED, it is possible to realize full-color tuning by combining one voltage-controlled OLED emitting any of two colors in the RGB primary colors with another OLED unit emitting the other primary color. We demonstrate the full-color tunable OLED using structure SOLED2: ITO/NPB(40 nm)/Eu(DBM)<sub>3</sub>Bath(20 nm)/Alq<sub>3</sub>(30 nm)/LiF(0.5 nm)/Al(16 nm)/Au(10 nm)/m-MTDATA:F4-TCNQ(7 nm)/NPB(40 nm)/Alq<sub>3</sub>(55 nm)/LiF(0.5 nm)/Al(70 nm). The EL spectra of the device are shown in Fig. 6. The bottom unit is a

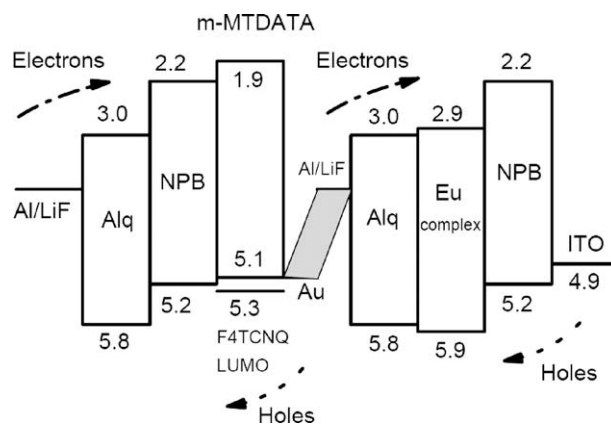


**Fig. 6.** The emitting spectra of SOLED2 with the structure of ITO/NPB/Eu(DBM)<sub>3</sub>Bath/Alq<sub>3</sub>/LiF/Al/Au/m-MTDATA:F4-TCNQ/NPB/Alq<sub>3</sub>/LiF/Al. (D, E, F, G, and H) are the spectra of the bottom OLED when biased at 5 V, 7 V, 9 V, 11 V and 13 V, respectively; (I) is the emitting spectrum of the top OLED; (J) is the spectrum of SOLED2 when the bottom OLED are biased at 9 V and the top OLED are biased at 6 V.

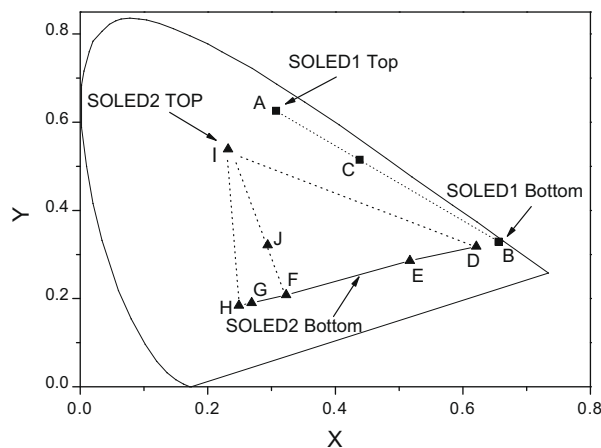
voltage-controlled color-tunable OLED [7]. At 5 V it shows merely the shape red emission from the Eu complex with good color purity, and the blue emission from NPB appears and dominates the EL spectra with increasing voltage. Thus it shows red-to-blue color tuning with the driving voltage. Meanwhile, the top unit shows green emission from Alq<sub>3</sub> layer under bias. As a result, SOLED2 realizes the superimposed EL spectrum of the bottom and top OLEDs for full-color emission when the two-stacked OLEDs units are biased simultaneously. An energy level diagram showing the HOMO and LUMO levels and carrier flow of SOLED2 is shown in Fig. 7.

### 3.4. Color tuning of the multicolor and full-color SOLEDs on the CIE diagram

The color tuning of SOLED1 and SOLED2 are shown in CIE diagram of Fig. 8. In the figure, points A and B are the color coordinates of the top and bottom OLED of SOLED1, and C represents the emitting color of SOLED1 when both units are driven at 5 V. Thus SOLED1 is a multi-color device that can exhibit any color on the straight line linked between A and B by selectively driving



**Fig. 7.** The energy level diagram showing the HOMO, LUMO levels and carrier flow in SOLED2.



**Fig. 8.** The color tuning of SOLED1 and SOLED2 on CIE coordinates. The CIE coordinates on this figure are corresponding to the spectra in Figs. 5 and 6 with the same identifier. (A) and (B) are the coordinates of the top and bottom OLEDs in SOLED1, respectively; (C) is the color coordinate when both top and bottom OLEDs in SOLED1 are biased at 5 V; (D, E, F, G, and H) are the color coordinates of the bottom OLED in SOLED2 when biased at 5 V, 7 V, 9 V, 11 V and 13 V, respectively; (I) is the color coordinate of the top OLED in SOLED2; (J) is the CIE coordinate of SOLED2 when the bottom OLED are biased at 9 V and the top OLED are biased at 6 V.

the bottom and top units. For SOLED2, point I is the color coordinate of the top green OLED unit, and the line linked between D and H represents the red-to-blue tunable color of the bottom unit. As shown in the figure, SOLED2 emit color at F when only the bottom OLED is driven at 9 V, and emit at J when the top unit is driven at 6 V and the bottom is at 9 V. Consequently, SOLED2 is a full-color device that can present any color inside the DHI triangle when the driving voltages of the bottom and top units are properly selected. Thus we show that the full-color emission can be achieved in a simple structure of two-unit SOLED. As compared with the three-unit SOLED [10,12], the two-unit SOLED is much simpler in processing. Meanwhile, the two-unit SOLED only needs a simple driving circuit because the two OLEDs can be biased independently without the common-ground referencing difficulty in three-unit SOLED.

#### 4. Conclusion

In conclusion, our results demonstrate that thermal evaporated Al/Au metal layer can act as an efficient intermediate electrode in color-tunable SOLED device. High brightness color-tunable OLEDs can be achieved by properly designing the Al/Au electrode thickness and using appropriate electron-injection and hole-injection buffer layer. Full-color emission is achieved in a simple device structure featured with (1) only two-unit SOLED, in which one unit is a voltage-controlled color-tunable OLED; (2) commonly used dual metals (Al/Au) as the intermediate electrode which can be easily process and have no damage to the organic layer. Based on the Al/Au intermediate electrode, efficient SOLED with wide color-tunable range can be realizable by using efficient emitting materials taking into account the microcavity effect in the stacked structure.

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#### References

- [1] M. Yoshida, A. Fujii, Y. Ohmori, K. Yoshino, *Appl. Phys. Lett.* 69 (1996) 734.
- [2] J. Kalinowski, P. Di Marco, M. Cocchi, N. Camaioni, J. Duff, *Appl. Phys. Lett.* 68 (1996) 2317.
- [3] R. Reyes, M. Cremona, E.E.S. Teotonio, H.F. Brito, O.L. Malta, *Chem. Phys. Lett.* 396 (2004) 54.
- [4] M. Granstrom, O. Inganas, *Appl. Phys. Lett.* 68 (1996) 147.
- [5] Y.Z. Wang, R.G. Sun, F. Meghdadi, G. Leising, A.J. Epstein, *Appl. Phys. Lett.* 74 (1999) 3613.
- [6] C.C. Huang, H.F. Meng, G.K. Ho, C.H. Chen, C.S. Hsu, J.H. Huang, S.F. Horng, B.X. Chen, L.C. Chen, *Appl. Phys. Lett.* 84 (2004) 1195.
- [7] C.J. Liang, W.C.H. Choy, *Appl. Phys. Lett.* 89 (2006) 251108.
- [8] P.E. Burrows, S.R. Forrest, S.P. Sibley, M.E. Thompson, *Appl. Phys. Lett.* 69 (1996) 2959.
- [9] G. Gu, V. Khalfin, S.R. Forrest, *Appl. Phys. Lett.* 73 (1998) 2399.
- [10] Z. Shen, P.E. Burrows, V. Bulovic, S.R. Forrest, M.E. Thompson, *Science* 276 (1997) 2009.
- [11] G. Gu, S.R. Forrest, *IEEE J. Sel. Top. Quantum Electron.* 4 (1998) 83.
- [12] G. Parthasarathy, G. Gu, S.R. Forrest, *Adv. Mater.* 11 (1999) 907.
- [13] H.H. Fong, W.C.H. Choy, K.N. Hui, Y.J. Liang, *Appl. Phys. Lett.* 88 (2006) 113510.
- [14] See for example: W.C.H. Choy, K.N. Hui, H.H. Fong, Y.J. Liang, P.C. Chui, *Thin Solid Films* 193 (2006) 509.
- [15] J.X. Sun, X.L. Zhu, H.J. Peng, M. Wong, H.S. Kwok, *Appl. Phys. Lett.* 87 (2005) 093504.
- [16] Norbert Koch, Steffen Duhm, Jurgen P. Rabe, Antje Vollmer, Robert L. Johnson, *Phys. Rev. Lett.* 95 (2005) 237601.
- [17] P.E. Burrows, V. Khalfin, G. Gu, S.R. Forrest, *Appl. Phys. Lett.* 73 (1998) 435.