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HIGH CONTRAST TANDEM ORGANIC LIGHT EMITTING DEVICES EMPLOYING SEMITRANSSPARENT INTERMEDIATE NANO METAL LAYERS AND A PHASE SHIFTING LAYER

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ABSTRACT

A high contrast-ratio organic light emitting device (OLED) is proposed and experimentally demonstrated. The OLED is implemented by stacking two organic phase tuning layers between composite metal layers and optimizing their thicknesses. Such a tandem device can increase the current efficiency by 120%, and reduce the operating voltage by 1.1 V, in comparison to conventional high contrast OLEDs. Measured reflection spectra validate the high-contrast capability of the OLED, and demonstrate experimentally an average reflectivity of 6% under ambient light illumination. This is the lowest reflectivity reported to date for OLEDs employing organic phase tuning layers.

INTRODUCTION

Organic semiconductors offer several advantages, namely, variety of materials, simple fabrication processes, cost-effectiveness and transparency, which make organic optoelectronic devices attractive for many applications.[1–5] Organic light emitting diodes (OLEDs) have particularly been used in flat panel displays (FPDs) due to their wide viewing angle, ultra thin thickness requirements, low power consumption and ability to emit light without the need for external backlight sources, in addition to the possibility of growing them on flexible substrates.

In a conventional single-cell OLED, the reflective metal layer of the cathode enables the back emission from organic layer to be reflected forward resulting in a high light coupling

efficiency. However, such OLEDs have the drawback of low contrast ratio due to the reflection of ambient light by the highly reflective cathode, which degrades the performance of OLEDs especially in outdoor applications where strong ambient light might be present.[2] Recently, a black composite layer has been introduced to OLED structures in order to increase the contrast ratio. This composite layer consists of a thin semi-transparent metal layer, a phase-tuning (PT) layer made of organic materials and a thick reflective metal layer,[6–8] and its low reflection is produced by the cancellation (destructive interference) of the two light waves reflected off the front thin metal layer and the rear thick metal layer which induces a π phase difference with respect to the front layer.[8] Due to their simple thermal evaporation, organic materials are considered as the best candidates for the realization of PT layers, such as tris(8-hydroxyquinoline) aluminum (Alq_3) (Refs. 8,9) and copper phthalocyanine (Ref. 10). To obtain a π phase difference, the thickness of the organic PT layer must be around 80 nm, and this is the same order of the emissive layer thickness. However, with a high carrier injection barrier between the PT layer and the intermediate metal layer, the operating voltage more than doubles, whereas, the current efficiency is reduced by 50%, because the black cathode absorbs half of the generated light from the emissive layer. The twofold increase in the operating voltage and the 50% reduction in the current efficiency reduce the overall power efficiency by 75%, thus making them impractical for emerging applications.

In this paper, we propose the use of an alternative black-layer cathode employing a composite semitransparent LiF/Al/Au in conjunction with stacked PT NPB/ Alq_3 layers to achieve much better current efficiency and even lower reflectivity of ambient light in comparison with conventional OLEDs employing LiF/Al as intermediate layers and Alq_3 as PT layers.[8–10]

SYSTEM DESCRIPTION AND EXPERIMENTAL RESULTS

Figure 1(a) shows the structures of a conventional single-cell OLED (Device 1), conventional high-contrast OLED (Device 2) and the proposed high contrast tandem OLED (Device 3), which were developed using thermal sublimation of organic materials in an ultra-high vacuum environment onto a transparent glass substrate coated with indium tin oxide (ITO), similar to the process used in Refs.[5–11] Prior to the gas treatment, the routine procedure of cleaning the substrate included ultrasonication in detergent for 30 minutes, spraying with de-ionized water for 2 minutes, ultrasonication in de ionized water for 20 minutes and drying by rotating at the spinning speed of 2000 rpm in a spin coater for 40 seconds. UV-ozone treatment was then made in a chamber with a high-purity oxygen flow. Immediately after the treatment, the samples were transferred into a growth chamber with a base pressure of 5×10^{-6} Pa for subsequent depositions of various layers. Alq_3 and NPB were chosen, respectively, as the electron transporting and emitting layer and the hole transporting layer, and Al on the top was chosen as the cathode. To increase the electron injection efficiency, a thin layer of LiF buffer was inserted between the Al and Alq_3 layers, as usually done for conventional OLEDs. The top Al layer was also in the shape of narrow strips, crossing the bottom ITO strips to form active device area of approximately $4 \times 4 \text{ mm}^2$.

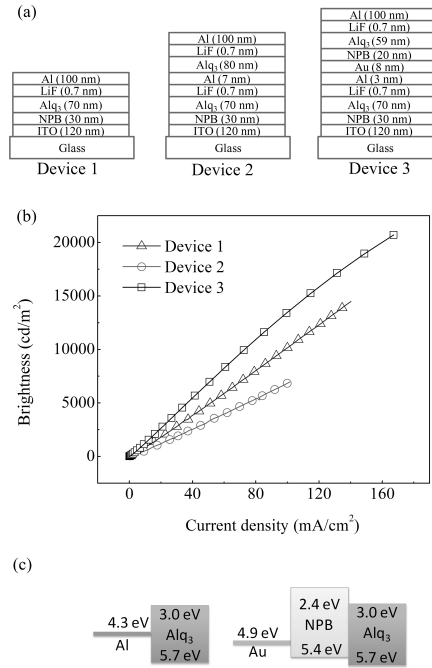


Fig. 2: (a) Architectures and (b) B-J Characteristics of Three Devices, Namely Device 1: Single-cell OLED, Device 2: High Contrast OLED with the PT Layer and Device 3: Proposed High Contrast Tandem OLED

RESULTS AND DISCUSSION

Figure 1(b) shows the measured brightness-current density characteristics for the three devices. It is obvious from Figure 1(b) that the brightness of Device 2 is approximately half of that of Device 1 at a given current density. For instance, at 40 mA/cm², the electroluminescence (EL) of Device 2 is 2500 cd/m², compared with 4050 cd/m² luminescence for Device 1 at the same current density. Such reduction in luminance for Device 2 is due to the additional metal-organic-metal (MOM) black cathode introduced on top of the bottom emissive cell.[8] Almost half of the photons emitted by the bottom emissive layer are reflected by the bottom and top surfaces of the black cathode. Therefore, even if the PT layer thickness is optimized to suppress the reflected light through destructive optical interference, the current efficiency of this OLED structure is limited since half of the generated light is lost by the black cathode. Therefore, the theoretical limit of current efficiency in such black-cathode-based OLEDs is only half of that of conventional counterparts. However, for the proposed Device 3, the EL at the same current density dramatically increases to 5500 cd/m², and in comparison with Device 2, the brightness increases by 120%. Interestingly, the EL of Device 3 is even higher than that of Device 1. Our measured results show that besides the bottom cell, the top MOM structure of Device 3 contributes to photon emission as well. In the MOM structures, the interfaces between intermediate layers and the PT

layers in Device 2 and Device 3 are Al/Alq₃ and Au/NPB respectively. Generally, the hole-injection barrier ϕ_B in this interface is the energy difference between the work function of the composite intermediate layer and the highest occupied molecular orbit (HOMO), which is analogous to the top of the valence band in an inorganic semiconductor.

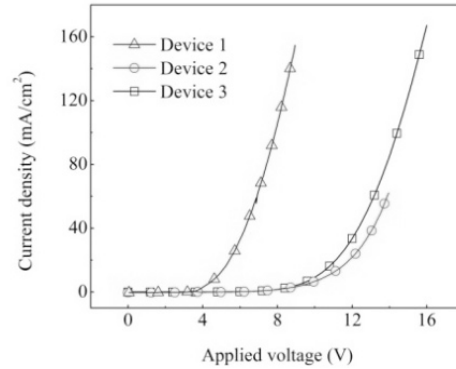


Fig. 3: *J-V* Characteristics of Devices 1, 2 and 3

Figure 1(c) illustrates the energy level diagrams of the interfaces between the composite intermediate layer and the PT layer for Device 2 and Device 3. The work function of Al and the HOMO of Alq₃ are respectively 4.3 eV and 5.7 eV,[12] the ϕ_B is 1.4 eV, which is high enough to prevent hole injection from the composite intermediate layer to the PT layer, restricting the exciton formation and photon emission in the PT layer. As for Device 3, the work function of Au and HOMO of NPB are 4.9 eV and 5.4 eV, respectively.[12,13] Thus ϕ_B is reduced from 1.4 eV for Device 2 to 0.5 eV for Device 3, and this barrier is low enough for hole injection from Au to NPB. The injected holes pass through the hole-transporting materials of NPB and are subsequently captured by the electrons in the Alq₃ layer to form excitons, resulting in the light emission from the PT layer. Thus, Device 3 has two emissive cells in tandem, namely, the lower cell and the upper MOM cell. We term Device 3 a “high contrast tandem OLED”. Noticeably, the current efficiency of Devices 3 is even higher than that of Device 1. This implies that the current efficiency of the upper MOM cell is higher than that of the bottom cell. This originates from much more efficient hole injection from Au to NPB in the upper cell than that from ITO to NPB in the lower cell due to the lower ϕ_B at the interface of Au/NPB.[12]

Figure 2 shows the *J-V* characteristics for the three devices. At a current density of 40 mA/cm², the operating voltage of Device 2 is 13.2 V, almost 7 V higher than that of Device 1. This indicates that the extra 80 nm thick Alq₃ higher than that of Device 1, and that the extra 80 nm thick Alq₃ layer introduces a considerably high resistance, resulting in a remarkably higher operating voltage. This is consistent with previous reported results.[14] For Device 3, the operating voltage is 1.1 V lower than that of Device 2. Since ϕ_B in the MOM structures for Device 2 and Device 3 is higher than 0.3 eV, the current flow through the MOM is dominated by the injection limited current (J_{ILC}).[15] From the J_{ILC} expression of Eq. (1) in Ref.,[15] at a given

current density, the operating voltage increases monotonically with the ϕ_B . Therefore, the lower ϕ_B at the Au/NPB interface leads to the lower operating voltage in Device 3.

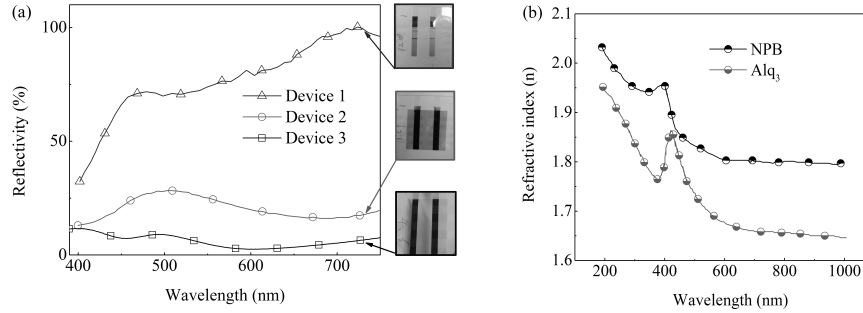


Fig. 4: (a) The Reflectance Spectra of Three devices as Schematically Shown in Figure 1a (b) The Refractive Indices of NPB and Alq₃.

Figure 3(a) shows the optical reflectance spectra for Devices 1–3 measured at 5° off the surface normal. The average reflectance of the OLED is 80% for Device 1, 20% for Device 2, mainly due to the addition of the MOM structure. To attain maximum destructive interference with the stacked NPB/Alq₃ PT layers in Device 3, the phase difference between the two light waves reflected off the upper and lower cells should be π . This corresponds to half of the design wavelength ($\frac{1}{2} \lambda$). If the refractive indices of NPB and Alq₃ are respectively $n_1(\lambda)$ and $n_2(\lambda)$, the optimized thicknesses, t_1 and t_2 , of NPB and Alq₃, respectively, for obtaining destructive interference should satisfy:

$$2[n_1(\lambda) \cdot t_1 + n_2(\lambda) \cdot t_2] = \frac{\lambda}{2} \quad \dots (2)$$

The spectral range of the ambient visible light extends from 400 nm to 750 nm, however, the elimination of the light around 550 nm is the main concern since 550 nm is the most sensitive wavelength to the human eyes. As shown in Figure 3(b), at 550 nm, $n_1(\lambda)$ and $n_2(\lambda)$ are 1.7 and 1.8, respectively. With the NPB thickness being fixed to 20 nm, the optimal Alq₃ thickness is 59 nm, according to Eq. (2). The measured reflectance from the proposed Device 3 was only 6.0% over the range of 400 to 750 nm. As we know, this is the lowest reflectance among all high contrast OLEDs employing an organic PT layer. While in the human-eye-sensitive range of 500–600 nm, the reflection value is further reduced to 4%, which nearly approaches the minimum limitation value since the reflectance of air/glass is around 4%.

CONCLUSIONS

We have proposed and demonstrated the concept of a high contrast-ratio organic light emitting device (OLED) realized through the stacking of two organic phase tuning layers between composite metal layers. Experimental results have demonstrated an increase in current efficiency

by 120%, a reduction in operating voltage by 1.1V and an ambient reflection as low as 6%, which are attributed to the photon emission from the PT layer, the reduced carrier injection barrier, between the intermediate layer and the PT layer, and a proper optical design.

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