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Color-stable and efficient stacked white organic light-emitting devices comprising blue fluorescent and orange phosphorescent emissive units

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We have demonstrated two kinds of stacked white organic light-emitting diodes (WOLEDs) employing tri(8-hydroxyquinoline) aluminum:20 wt %Mg/MoO_3 as charge generation layer. White light emission can be obtained by mixing blue fluorescence and orange phosphorescence. Stacked WOLED with individual blue fluorescent and orange phosphorescent emissive units has better color stability and higher efficiency than that with double white emissive units, which is attributed to the avoidance of the movement of charges recombination zone and elimination of the Dexter energy transfer between blue and orange emission layers occurring in the latter. The efficiency of the stacked WOLED is 35.9 cd/A at 1000 cd/m^2.
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Organic light-emitting diodes (OLEDs), especially white OLEDs (WOLEDs), have attracted a great deal of attention in recent years for their potential use in full-color flat-panel displays with color filters or as a new generation of solid state lighting sources.[1–5] Efficient WOLEDs have been fabricated by adopting various configurations combined with efficient materials. Among these configurations, WOLEDs with multiple emission layers in which each layer emits a different color light are most common because of their facility in process and obtaining white light emission. However the undesirable change in emission color with the increasing voltage is often observed in WOLEDs with multiple emission layers. Some efforts have been made to improve the color stability of WOLEDs such as introducing interlayer at the interface of emitting layers to control the diffusion of excitons and charges[5] or by using a charge confining structure without interlayer.[6] Moreover, the peak efficiency is often achieved at low luminance due to the remarkable quenching effect at higher electric field.[7] For practical use in displays and lighting sources, high brightness, as well as the good color stability, is required. So it is necessary to obtain high brightness and high efficiency at low current density to reduce thermal degradation of the device due to excessive current.

Since the current efficiency and luminance can scale linearly with the number of emitting units, stacked OLEDs consisting of vertically stacked multiple emitting units in a device in series via charge generating layer (CGL) have attracted particular interest in recent years.[8–20] Moreover, stacked OLEDs can obtain high brightness and high efficiency at low current density, which is most important for practical use.

In a stacked OLED, CGL plays an important role in affecting the device performances. Up to now, various CGLs have been used. A Mg-doped organic layer can be easily deposited by thermal evaporation, and CGLs based on it such as tri(8-hydroxyquinoline) aluminum (Alq_3):Mg/WO_3 (Ref. 11) and Alq_3:Mg/tetrafluorotetracyanoquinodimethane (F_4-TCNQ):4,4′,4″-tris(3-methylphenylphenylamino)-triphenylamine (*m*-MTDATA) (Ref. 17) have demonstrated that Alq_3:Mg is a crucial component of CGL in improving the efficiency. MoO_3 (Refs. 14, 15, and 20) can be conveniently deposited by thermal evaporation. The film is homogeneous and more transparent over the wavelength range from 400 to 800 nm. So it is desirable to investigate the combination of MoO_3 with Alq_3:Mg as an alternative CGL.

In this letter, we demonstrate stacked orange OLED and WOLEDs based on the CGL of Alq_3:20 wt %Mg/MoO_3. Stacked WOLEDs are fabricated by connecting the blue fluorescent unit and the orange phosphorescent emission unit or by connecting double white light emission units in series. The former has much better color stability and higher efficiency than the latter. The spectra of the former remain almost the same over a large range of operation voltages.

m-MTDATA and *N*,*N*′-bis-(1-naphthyl)-*N*,*N*′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB) are used as hole-injection layer and hole-transportation layer, respectively. 4,4′_*N*,*N*′_ dicarbazole-biphenyl (CBP) doped with bis(2-(2-fluorphenyl)-1,3-benzothiozolato-N,C2′)iridium (acetylacetonate) [$(F-BT)_2$(acac)] is used as orange phosphorescent emission layer, and 4,4,8-bis(2,28-diphenylvinyl)-1,18-biphenyl (DPVBi) is the blue fluorescent emission material. 4,7-diphenyl-1,10-phenanthroline (Bphen) and Alq_3 act as hole-blocking layer and electron-transporting layer, respectively. Finally, 0.8 nm LiF covered by Al is used as cathode. Detailed processes of fabrication and measurement for OLEDs have been described in our previous paper.[22]

To illuminate the function of the CGL of Alq_3:20 wt %Mg/MoO_3, we fabricated a two-unit stacked orange device and a conventional orange one as the control device. The configurations of the two orange devices are as follows. For a conventional device it is ITO/*m*-MTDATA (30 nm)/NPB (20 nm)/CBP:8 wt % $(F\text{-}BT)_2$Ir(acac) (30 nm)/Bphen (20 nm)/Alq_3 (20 nm)/LiF (0.8 nm)/Al. For a

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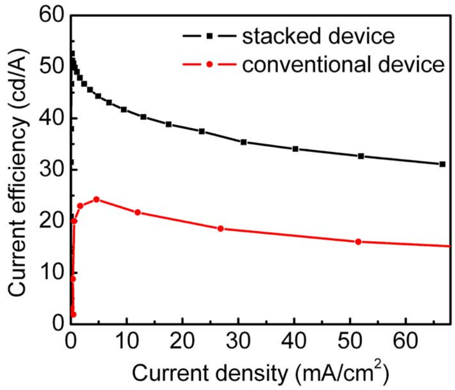

FIG. 1. (Color online) Current efficiency vs current density characteristics of the conventional device and the two-units stacked device.

two-unit stacked device it is ITO/*m*-MTDATA (30 nm)/NPB (20 nm)/CBP:8 wt % $(F-BT)_2Ir(acac)$ (30 nm)/Bphen (20 nm)/Alq_3 (20 nm)/Alq_3:20 wt %Mg (30 nm)/MoO_3 (3 nm)/*m*-MTDATA (30 nm)/NPB (30 nm)/CBP:8 wt % $(F-BT)_2Ir(acac)$ (30 nm)/Bphen (20 nm)/Alq_3 (20 nm)/LiF (0.8 nm)/Al.

Figure 1 shows the current efficiency versus current density characteristics of the two orange devices. As can be seen, the current efficiency of the two-unit stacked device is about two times as high as that of the conventional device.

In the followig, we fabricated efficient stacked WOLEDs based on two complement colors of blue fluorescent and orange phosphorescent dyes. Figure 2 shows the electronic level configurations of the two stacked WOLEDs, and both the devices are based on the CGL of Alq_3:20 wt %Mg (30 nm)/MoO_3 (3 nm). Device A is comprised of blue fluorescent and orange phosphorescent emissive units vertically stacked via the CGL, while device B contains double white emissive units with the same blue and orange emissive materials.

Figure 3 shows Electroluminescence (EL) spectra normalized at the maximum value of [Fig. 3(a)] device A and [Fig. 3(b)] device B at different luminance; the inset shows the Commission International de L'Eclairage (CIE) coordinates and the color rendering index (CRI) at the corresponding luminance. As can be seen, the spectra of device A remain almost the same over a wide range of luminance; the slight decrease in the intensity of the orange emission is mainly due to the stronger dissociation and annihilation of excitons on charge carriers at higher current density, which was examined by Kalinowski *et al.*[7] However for device B, the intensity of the blue emission sharply increases relative to the orange emission with increasing luminance. The change in the spectra in device B is mainly due to the movement of charges recombination zone, which is commonly observed in WOLEDs with multiple emission layers. However for device A, the blue fluorescent emissive unit and the orange phosphorescent emissive one are separated by the CGL, so the blue charges recombination zone is practically independent of the orange one that avoids the movement of the charges recombination zone. Device A with CIE coordinates of (0.363, 0.361) has a CRI of 73, which may be lower than that for many types of WOLEDs reported in literature[21] because it only utilizes two complementary colors to obtain white light emission.

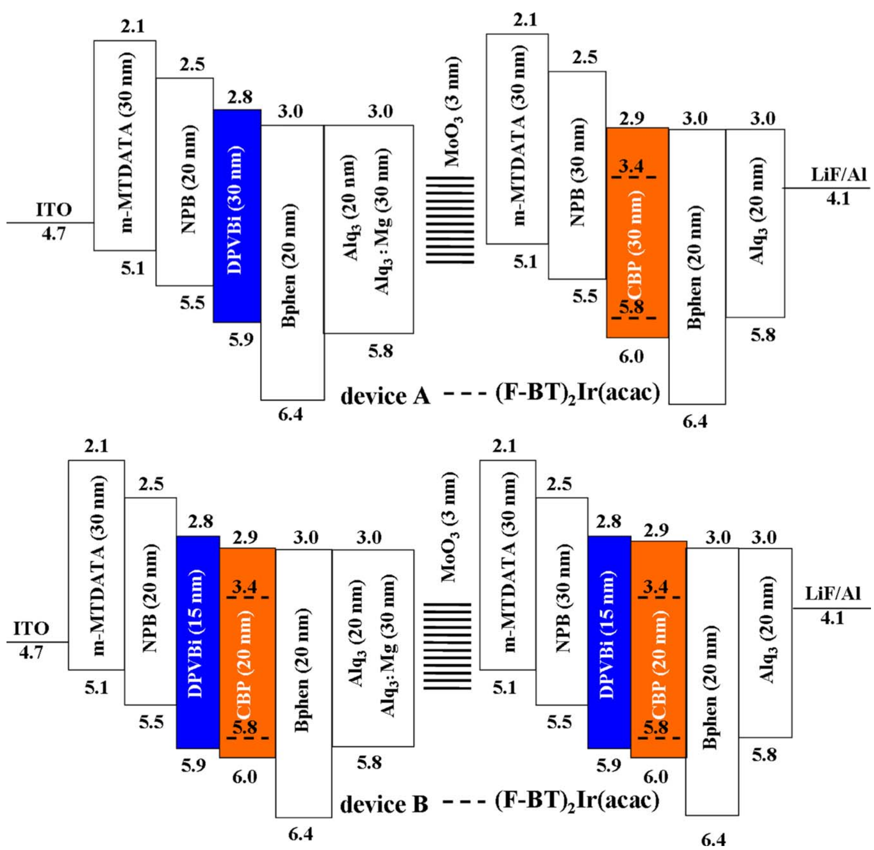

FIG. 2. (Color online) The electronic level configurations of both the stacked WOLEDs. Numbers indicate the respective highest occupied and lowest unoccupied molecular orbital energies of all organic materials relative to vacuum.

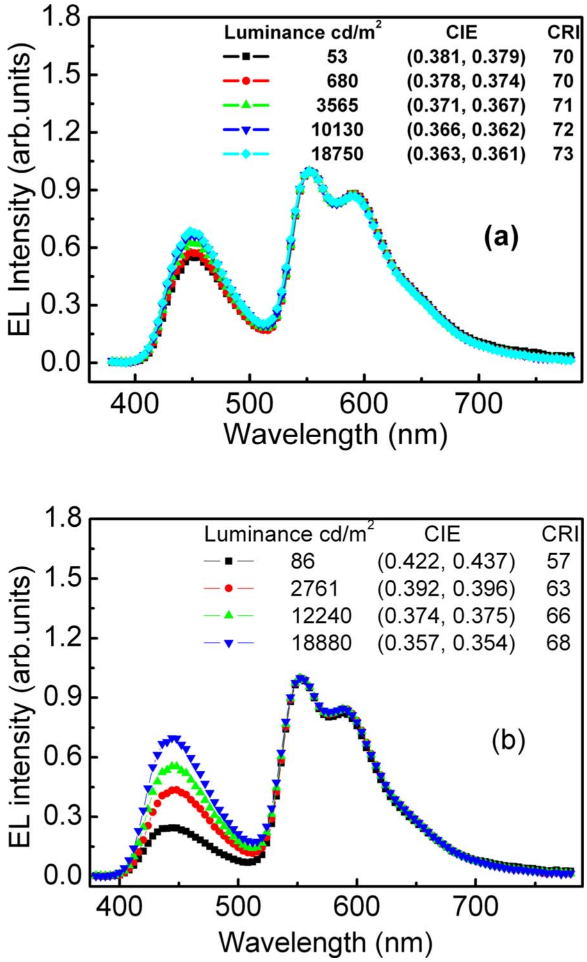

FIG. 3. (Color online) EL spectra of (a) device A and (b) device B at different luminance normalized at the maximum value. The inset shows the CIE coordinates and the CRI values at the corresponding luminance.

The current density versus voltage characteristics of the two devices are shown in Fig. 4. It can be seen that the current density for device B is slightly lower than that for device A at the same voltage. The lower current density for device B may be the result from the multiple emission layers in each unit that affects the charge transport. The inset of Fig. 4 shows the current efficiency-current density-luminance characteristics of the two devices. Device A has higher efficiency than device B, and the maximum efficiencies of devices A and B are 36.3 cd/A and 27.8 cd/A, respectively. The following reason may be accounted for the lower efficiency of device B. In the white emissive unit of device B, the blue fluorescent layer is adjacent to the orange phosphorescent emission one, so the Dexter energy transfer from the orange phosphor triplet state to the lower nonradioactive blue triplet state can occur,[22] which quenches the emission of $(F\text{-}BT)_2Ir(acac)$ and reduces the performance of device B. Moreover, device A can obtain high efficiency at high luminance. For example, current efficiencies at the luminance of 1000 and 10 000 cd/m^2 are 35.9 and 27.8 cd/A, respectively.

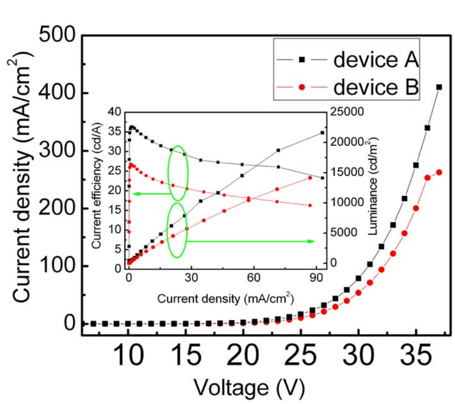

FIG. 4. (Color online) Current density vs voltage characteristics of the two stacked WOLEDs. The inset shows current efficiency-current density-luminance characteristics of the two stacked WOLEDs.

In summary, we have demonstrated stacked orange and WOLEDs based on the CGL of Alq_3:20 wt %Mg/MoO_3. The current efficiency of the stacked orange OLEDs with two orange emitting units is about two times as high as that of the single-unit device. Stacked WOLED consisting of vertically stacked blue fluorescent and orange phosphorescent units has excellent color stability and higher current efficiency. The white device has a current efficiency of 35.9 cd/A at the luminance of 1000 cd/m^2.

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