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Influence of interlayer on the performance of stacked white organic light-emitting devices

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Stacked white organic light-emitting devices (WOLEDs) comprising of blue fluorescent and orange phosphorescent emissive units employing tri(8-hydroxyquinoline) aluminum $(Alq_3):Mg/MoO_3$ as charge generation layer are fabricated. The working mechanism of $Alq_3:Mg/MoO_3$ is also discussed using a simple method. We demonstrate charge-carrier separation takes place only in MoO_3 layer. Stacked WOLED with better performance was obtained by adjusting the thickness of MoO_3. The stacked WOLED with efficiency of 39.2 cd/A has excellent color stability with the Commission Internationale de l'Eclairage coordinates only changing from (0.407, 0.405) to (0.398, 0.397) when luminance increases from 22 to 10 000 cd/m^2. © *2009 American Institute of Physics*. [doi:10.1063/1.3234379]

White organic light-emitting devices (WOLEDs) have aroused a great deal of attention in the past decade because of their potential use in full-color flat-panel displays or as low power consumption solid state lighting sources.[1–7] In order to improve the lifetime and reduce thermal degradation of organic light-emitting devices (OLEDs) due to the excessive current in practical use, it is necessary to obtain high brightness and high efficiency at lower current density. A promising approach is via a stacked OLEDs consisting of vertically stacked multiple emissive units by charge generating layer (CGL). Stacked OLEDs attract particular interest because the current efficiency and luminance can scale linearly with the number of emitting units.[8–17] The major challenge in stacked OLEDs is to prepare an effective CGL structure. Though several different CGL architectures and many concepts for the origin of the charge-carrier in CGL are proposed, the working mechanism of CGL is not clear so far. Kröger *et al.*[17] investigated an organic/organic CGL structure based on tetrafluorotetracyanoquinodimethane doped 4,4′,4″-tris(N-1-naphtyl-N-phenylamino)-triphenylamine (1-TNATA) and Li-doped 1,3,5-tri(phenyl-2-benzimidazole)-benzene (TPBi). By applying the Kelvin probe method and low temperature *I-V* measurements, they proposed a model of electrons tunneling through the depletion zone from the highest occupied molecular orbit of 1-TNATA to the lowest unoccupied molecular orbit of the TPBi.

In this letter, we study the working mechanism of the CGL of Alq_3:20 wt % Mg/MoO_3 using an ingenious method. Just from the difference of the spectra at the same current density, we can pinpoint the exact position where charge-carrier are generated and separated locating in the MoO_3 layer. More efficient and stable stacked WOLED can be obtained just by adjusting the thickness of MoO_3. The

maximum efficiency of the stacked WOLED with the MoO_3 thickness of 1.5 nm is 39.2 cd/A.

To illuminate the exact location where charge-carrier is generated and separated in $Alq_3:Mg/MoO_3$, devices with the following configurations were fabricated shown in Fig. 1: ITO/Blue fluorescent electroluminescent (EL-B) unit/$Alq_3:Mg$ (X nm)/MoO_3 (Y nm)/orange phosphorescent electroluminescent (EL-O) unit/LiF(0.8 nm)/Al. (For device A, X=0 nm, Y=0 nm; for device B, X=30 nm, Y=0; for device C, X=0 nm, Y=10 nm; for device D, X=30 nm, Y=10 nm), Where the EL-B unit is composed of 4,4′,4″-tris(3-methylphenylphenylamino)-triphenylamine (m-MTDATA: 30 nm)/*N*,*N*′-bis-(1-naphthyl)-*N*,*N*′-diphenyl-1,1′-biphenyl-4,4′-diamine (NPB: 20 nm)/4,4′-bis(2,28-diphenylvinyl)-1,1′-biphenyl (DPVBi: 30 nm)/4,7-diphenyl-1,10-phenanthroline (Bphen:20 nm)/Alq_3 (20 nm)

FIG. 1. (Color online) The structure of devices A–D. For device A, X=0 nm, Y=0 nm; for device B, X=30 nm, Y=0 nm; for device C, X=0 nm, Y=10 nm; for device D, X=30 nm, Y=10 nm.

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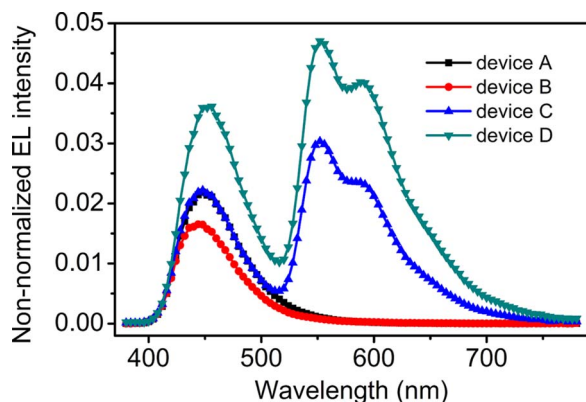b)Electronic mail: syliu@jlu.edu.cn.

FIG. 2. (Color online) Non-normalized EL spectra of the stacked devices A–D at the current density of 10 mA/cm^2.

FIG. 3. (Color online) Non-normalized EL spectra of the stacked devices E-H at the current density of 10 mA/cm^2.

and the EL-O unit is m-MTDATA (30 nm)/NPB(30 nm)/ CBP: 8 wt % (F-BT)$_2$Ir(acac) (30 nm)/Bphen (20 nm)/Alq$_3$ (20 nm), respectively. CBP and (F-BT)$_2$Ir(acac) stand for 4,4′ N,N′ dicarbazole-biphenyl and *bis* [2-(2-fluorphenyl)-1,3-benzothiozolato-N,C$^{2'}$] iridium (acetylacetonate). m-MTDATA, NPB, Bphen, and Alq$_3$ were used as hole-injection layer, hole-transporting layer, electron-transporting layer and electron-injection layer, respectively. DPVBi and CBP doped with (F-BT)$_2$Ir(acac) were selected as blue fluorescent and orange phosphorescent emission layer, respectively. LiF/Al was used as the cathode. Detailed fabrication and measurement processes of devices have been described in our previous paper.[18]

Figure 2 shows the non-normalized electroluminescent (EL) spectra of stacked devices A-D at the same current density of 10 mA/cm^2. The spectra of the devices A and B both show only one emission peak originating from the EL-B unit while that of device C and D contain two primary peaks originating from the EL-B unit and the EL-O unit, respectively. For devices A and B, the excitons recombination zone only lies in the EL-B unit and there are no excitons generated in the EL-O unit due to the fact that the holes aren't injected into the EL-O unit. The emission intensity from the EL-B unit of device B decreases relative to device A. It is because the number of electrons diffusion to EL-B unit becomes less due to the introduction of 30 nm Alq$_3$:Mg, which causes charge more imbalance in emission layer. But for device C, the strong orange emission from the EL-O unit is observed. We think charge-carrier is generated and separated in MoO$_3$ layer, and the generated holes can be easily injected into the EL-O unit, then the injected holes recombine with electrons injected from the cathode. Some generated electrons are injected into EL-B unit which reduces the charge imbalance, so the intensity of blue emission increases a little. When we introduce a whole CGL of Alq$_3$:Mg/MoO$_3$ in device D, the intensity of the emission peaks both sharply increase relative to that of device C, which demonstrates that the performances of the stacked device are much better when Alq$_3$:Mg and MoO$_3$ are combined. Because when Alq$_3$:Mg is adjacent to MoO$_3$, the electrons injection into EL-B unit become much easier, and more holes also can be injected into EL-O unit without trapped by electrons in device C, which causes both the intensity of the emission from DPVBi and (F-BT)$_2$Ir(acac) increase.
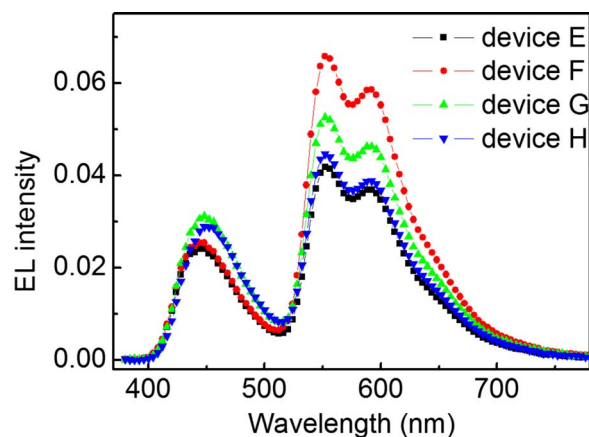
As can be seen, for the stacked devices based the CGL of Alq$_3$:Mg/MoO$_3$, MoO$_3$ has more important effect on the performances especially for the upper EL-O unit near the cathode. In the following, we demonstrate stable and more efficient stacked WOLEDs by changing the thickness of MoO$_3$. The architectures of the stacked WOLEDs are: ITO/ EL-B unit/Alq$_3$:Mg (30 nm)/MoO$_3$ (Z nm)/EL-O unit/ LiF(0.8 nm)/Al, Z=0.5,1.5,4,8 nm for device E, F, G, and H, respectively.

Figure 3 shows the non-normalized EL spectra of the stacked devices E-H. For device E, because MoO$_3$ layer is too thin, fewer charge-carriers are generated in MoO$_3$ layer and the injection of holes into the EL-O unit is inefficient, which causes weaker emission of (F-BT)$_2$Ir(acac). The intensity of the orange emission from (F-BT)$_2$Ir(acac) sharply increases when the thickness of MoO$_3$ changes from 0.5 to 1.5 nm, but when Z increases from 1.5 to 4 nm, the intensity of the emission from (F-BT)$_2$Ir(acac) decreases. We think the optimized thickness of the MoO$_3$ to generate charge-carrier and improve the holes injection into the EL-O unit is 1.5 nm. Note that when Z increases from 4 to 8 nm, the spectra show only a minor change. It may be because the injection of the charge-carriers into the two emission units reaches equilibrium.

Figure 4 shows the normalized EL spectra of the stacked device F at different luminance. As can be seen, the spectra
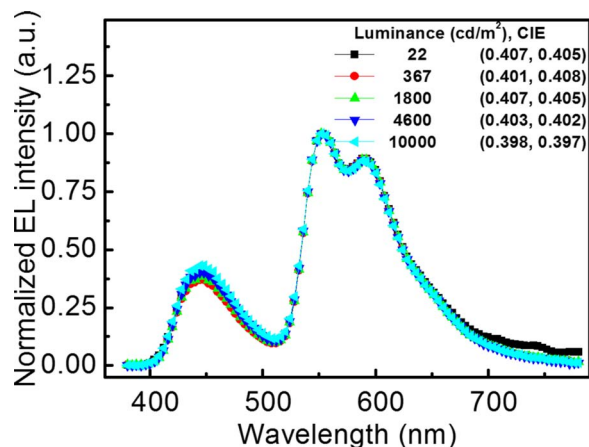

FIG. 4. (Color online) Normalized EL spectra of the stacked device F at different luminance.
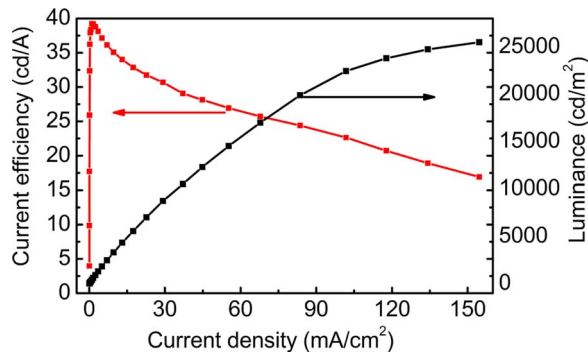
FIG. 5. (Color online) Current efficiency-current density-luminance characteristics of the stacked device F.

remain almost the same over a wide range of luminance. The Commission Internationale de l'Eclairage (CIE) coordinates only change from (0.407, 0.405) to (0.398, 0.397) with luminance increasing from 22 to 10 000 cd/m^2. The excellent color stability of the stacked WOLED is attributed to the avoidance of the movement of exciton recombination zone usually occurring in WOLEDs with multiple emission layers.[18]

Current efficiency-current density-luminance characteristics of the stacked device F are shown in Fig. 5. Device F has a maximum current efficiency of 39.2 cd/A. The device can obtain high luminance and efficiency at very low current density, which can reduce thermal degradation due to the excessive current and increase the lifetime. For example, at the luminance of 500 and 10 510 cd/m^2 corresponding to the current density of as low as 1.30 and 37.1 mA/cm^2, device F has current efficiencies of 39.2 and 29 cd/A, respectively.

In summary, we have demonstrated stable and efficient stacked WOLEDs based on the CGL of Alq_3:Mg/MoO_3. We also discuss the working mechanism of the stacked devices with different interlayer. One stacked WOLED with stable spectra has a maximum current efficiency of 39.2 cd/A.

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