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Direct monitoring of recombination zone shift during lifetime measurement of phosphorescent organic light-emitting diodes

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ABSTRACT

Recombination zone of phosphorescent organic light-emitting diodes during lifetime measurement was directly monitored using a red sensing layer inserted at different positions of blue phosphorescent emitting layer. The shift of recombination zone could be identified by the change of red intensity of the red sensing layer because the red intensity reflects the exciton density around the red sensing layer. Gradual shift of the recombination zone from hole transport layer side to electron transport layer side could be directly tracked according to driving time of the device.

Introduction

Exciton formation in the light-emitting layer of phosphorescent organic light-emitting diodes (PHOLEDs) has great impact on the quantum efficiency and lifetime because of large exciton diffusion length of triplet excitons generated in the phosphorescent emitting layer. Exciton diffusion length of triplet emitter can be as high as 100 nm due to long excited state lifetime of a few μs of common triplet emitters [1]. Therefore, the management and monitoring of the triplet exciton formation is critical to the device performances of PHOLEDs.

There are several methods to examine the recombination zone of PHOLEDs. One method is to introduce a thin layer of red emitter as a sensing layer in the emitting layer [2–5]. The light emission intensity of the red sensing layer reflects the exciton density near the sensing layer and recombination zone of the device can be directly monitored by the red intensity of the sensing layer. The use of the red sensing layer was effective to investigate the recombination zone of the green and blue PHOLEDs. Another method is to use an emitting layer consisting of triplet emitter doped layer and non-doped layer [6,7]. Relative thickness of the doped and non-doped layers was changed, and the quantum efficiency and electroluminescence (EL) spectra of the device gave hints about the recombination zone of the device. However, this method is an indirect method because the exact location of the recombination zone cannot be extracted. Additionally, the non-doped layer affects the charge transport of the devices and can provide incorrect information about the exciton formation zone of the device. Other than these, optical simulation method can also be utilized to study the recombination zone [8] and a lot of researches about the recombination zone of the PHOLEDs have been carried out [9–20] because of the importance of the recombination zone management for lifetime. It has been known that the lifetime of the PHOLEDs is improved when the recombination zone of the emitting layer is widespread due to suppressed triplet-polaron and triplet–triplet exciton quenching [19–21]. However, most studies about the lifetime of PHOLEDs were focussed on the recombination zone before driving the device and the recombination zone change during driving was not considered.

There have been several papers reporting indirect evidence of recombination zone shift during lifetime measurement by increased resistance at the electron transport layer side [22–25]. Current density recovery after fresh cathode deposition and current increase of electron only devices were proposed as the supporting data to suggest recombination zone shift from hole transport layer side to electron transport layer side. However, these methods could not show the real recombination zone shift before and after lifetime measurement. Therefore, it is very important to develop an analytical method to monitor the recombination zone change during lifetime measurement and this information can be used to improve the lifetime of blue PHOLEDs.

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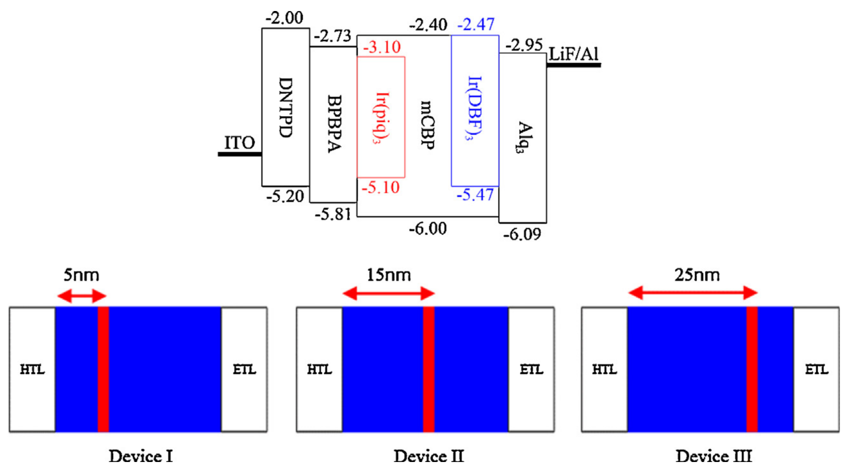

Fig. 1. Energy level diagram and device structure of blue PHOLEDs with a red sensing layer.

In this work, direct monitoring of the recombination zone before and after lifetime measurement was carried out using a red sensing layer inserted at different positions of blue phosphorescent emitting layer. It was demonstrated that the recombination shift during lifetime measurement can be directly traced using a red sensing layer and the recombination zone of the blue PHOLEDs was shifted from hole transport layer side to electron transport layer side during driving of the device.

Experimental

The red sensing layer was inserted in the emitting layer of blue PHOLEDs with a device structure of indium tin oxide (ITO, 120 nm)/N,N′-diphenyl-N,N′-bis-[4-(phenyl-m-tolyl-amino)-phenyl]-biphenyl-4,4′-diamine (DNTPD, 60 nm)/N,N,N′N′-tetra[(1,1′-biphenyl)-4-yl]-(1,1′-biphenyl)-4,4′-diamine (BPBPA, 30 nm)/3,3′-di(carbazol-9-yl)biphenyl (mCBP):tris[2-[1-[1,3-bis(1-methylethyl)2-dibenzofuranyl]-1H-imidazol-2-yl-N]phenyl-C^2] iridium(III) ($Ir(DBF)_3$) (25 nm, 10% doping)/tris(8-hydroxyquinolinato)aluminum (Alq_3, 35 nm)/LiF (1 nm)/Al (200 nm). Sensing layer was made up of mCBP doped with tris[1-phenylisoquinolinato-N,C^2] iridium(III) ($Ir(piq)_3$) red triplet emitter (5% doping). The thickness of the sensing layer was 2 nm and it was inserted at different locations of the blue emitting layer. The distances of the sensing layer from the hole transport layer were 5 nm (device I), 15 nm (device II) and 25 nm (device III). Vacuum evaporation was the basic device fabrication method. The emitting layer with the red sensing layer was constructed by stepwise deposition of the blue emitting layer, red sensing layer, and blue emitting layer. Device structures are shown in Fig. 1. EL spectra were gathered using CS2000 spectroradiometer. Electrical measurements and luminance measurements were performed by combining the CS2000 spectroradiometer and Keithley 2400 source measurement unit. Quantum efficiency was calculated from electrical and optical measurements results.

Results and discussion

It is important to check the emission zone of the PHOLEDs to get deep insight about light emission of PHOLEDs and tracing method using a sensing layer was popular. In order to trace the recombination zone shift of the blue PHOLEDs using a red sensing layer, the red sensing layer should be as stable as the blue emitting layer within the time range for the lifetime measurement of the blue PHOLEDs because discrepancy of the blue and red lifetime may distort the emission spectrum of the device. Similar degradation of the red and blue emitting layer may allow the interpretation of the emission spectra from the view point of recombination zone because different red intensity according to driving time would reflect the recombination zone change. In this work, mCBP:$Ir(piq)_3$ was a red emitting layer and mCBP:$Ir(DBF)_3$ was a blue emitting layer.

The compatibility of the mCBP:$Ir(piq)_3$ emitting layer with mCBP:$Ir(DBF)_3$ as a sensing layer was confirmed by fabricating PHOLEDs with both mCBP:$Ir(DBF)_3$ and mCBP:$Ir(piq)_3$ emitting layers. Device structure was ITO (120 nm)/DNTPD (60 nm)/BPBPA (30 nm)/mCBP:$Ir(DBF)_3$ (5 nm, 10%)/mCBP:$Ir(piq)_3$ (25 nm, 5%)/Alq_3 (35 nm)/LiF (1 nm)/Al (100 nm). The lifetime of the device was measured at an initial luminance of 1000 cd/m^2 up to 500 cd/m^2 and the EL spectra according to the luminance decrease were compared (Fig. 2). The white PHOLEDs showed the same EL spectra before and after lifetime test except for the suppressed emission of hole transport layer. This result indicates that the luminance decrease of the blue and red emitting layers is similar. Therefore, the use of the mCBP:$Ir(piq)_3$ emitting layer as a sensing layer to track the recombination zone shift during driving can be considered as a proper approach.

Based on the EL spectra change of the white PHOLEDs, the recombination zone of the blue PHOLEDs was checked by the red sensing layer. The mCBP:$Ir(piq)_3$ sensing layer was inserted at different locations of mCBP:$Ir(DBF)_3$ emitting layer as shown in Fig. 1. The distances of the red sensing layer from the hole transport layer were 5 (device I), 15 (device II) and 25 nm (device III). Basic device characteristics of the device I, II and III were studied by measuring current density and luminance according to driving

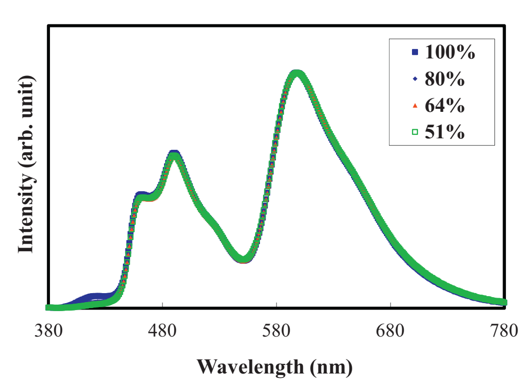

Fig. 2. EL spectra of the white PHOLEDs with a stacked mCBP:$Ir(DBF)_3$/mCBP:$Ir(piq)_3$ emitting layer according to the relative luminance during lifetime test.