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Recombination zone in white organic light emitting diodes with blue and orange emitting layers

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ABSTRACT

White fluorescent OLED devices with a 10 nm thick blue-emitting layer and a 31 nm thick orange-emitting layer have been fabricated, where the blue-emitting layer is stacked on a hole transport layer. An interlayer was inserted between the two emitting layers. The thickness of the interlayer was changed among 0.3, 0.4, and 1.0 nm. White emission with CIE coordinates close to (0.33, 0.33) was observed from all the OLEDs. OLED with 0.3 nm thick interlayer gives the highest maximum luminous efficiency (11 cd/A), power efficiency (9 lm/W), and external quantum efficiency (5.02%). The external quantum efficiency becomes low with increasing the interlayer thickness from 0 nm to 1.0 nm. When the location of the blue- and orange-emitting layers is reversed, white emission was not obtained because of too weak blue emission. It is suggested that the electron–hole recombination zone decreases nearly exponentially with a distance from the hole transport layer.

1. Introduction

White organic light-emitting diodes (OLEDs) have gained interest for the applications to flat panel displays and lighting sources. Three primary color emitting chromophores are doped in different conductive hosts in three emitting layers (EMLs) to achieve the white-light emission from the single OLED, or two complementary color emitting chromophores are used [1–3]. In the mass production of the devices, the OLEDs using the complementary color emitting dopants and a common host material are preferable because the fabrication cost is lowered due to the reduced number of the stacked EMLs.

Wide electron–hole recombination zone gives a longer OLED operational lifetime compared with narrow recombination zone, because narrow zone of high electron and hole densities induces heating. Therefore it is important to investigate the region of recombination zone although it is known that the recombination zone resides mostly at the boundary of hole transport layer (HTL) and EML. The characteristics of recombination zone, however, have not been fully clarified.

The present work was undertaken to investigate the location, width, and distribution of the recombination zone by inserting interlayer between blue- and orange-emitting layers in white OLED and by changing the thickness of EML adjacent to HTL.

Thin interlayer has been inserted between two emitting layers to avoid the energy transfer from the higher-energy emitter to the lower-energy emitter [4,5]. It has been suggested that the thickness of interlayer in the stacked emitting layer are critical to improve the light-emitting efficiency and color stability of the white OLEDs [6]. Here we also investigate whether the thin interlayer actually suppresses the energy transfer and how much influence the interlayer thickness gives to the efficiency and color stability.

2. Experimental procedure

We fabricate OLED with two emitting layers and its interlayer. The device structure is

ITO/HIL(70 nm)/HTL(20 nm)/BEML(10 nm)/ITL(x nm)/OEML(31 nm)/ETL(10 nm)/LiF(0.5 nm)/Al,

where HIL, BEML, ITL, OEML, and ETL mean hole injection layer, blue emitting layer, interlayer, orange emitting layer, and electron transport layer, respectively (Fig. 1). Fluorescent small-molecules are used for all the organic layers. The layer thickness of each layer is indicated in the parenthesis. The devices with the ITL of a different thickness (x = 0.3, 0.4, and 1.0 nm) were fabricated, which are called Device BR-3, BR-4, and BR-10, respectively. The emission area is 3×3 mm^2. The devices were fabricated using a thermally evaporation in vacuum chamber. BEML and OEML consist of the identical host material doped with 2% blue- and 4.5% orange-fluorescent

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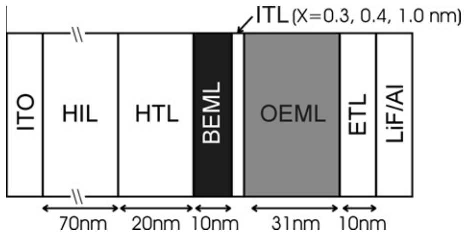

Fig. 1. Layer structure of OLED device.

materials, respectively. The material which is used as host of BEML and OEML was also used as ITL, while Alq_3 was used as ETL.

The current density–voltage–luminance (J–V–L_v (cd/m^2)) characteristics and electroluminescence (EL) spectra of the OLED devices were measured with a Konica-Minolta CS-1000 spectroradiometer in the normal direction of OLED surface, and with a computer controlled programmable Keithley 2400 dc Source Meter.

The external quantum efficiency (EQE) was measured using a Hamamatsu-Photonics C9920-12 EQE measurement system, where OLEDs were installed in an integrated sphere. Unlike the case of measurement with Konica-Minolta CS-1000 spectroradiometer, total photometric power emitted in all directions from a light source OLED was obtained from the EQE measurement system. The EL spectra, the current density–voltage–luminous flux (J–V–L (lumen)) characteristics, chromaticity, the luminous efficiency η_{lumi} (cd/A) and power efficiency η_{power} (lm/W) were also measured. The Keithley 2400 dc Source Meter was also used to control the applied voltage.

Two thin films of the material used in BEML were thermally evaporated on quartz plate. One film is doped with the same material used in BEML as emitter, the other is doped with the same material used in OEML as emitter. The doping level is 2% and 4.8% for the former and latter films, respectively. The photoluminescence (PL) spectra of these films were measured with a Spex FluoMax-P spectrophotometer at room temperature.

3. Experimental results and discussion

Fig. 2 shows the normal-direction EL spectra of Devices BR-3, BR-4, and BR-10 at 8 V. The blue emission has a peak at 472 nm and a shoulder at 498 nm which are attributed to the 0–0 and 0–1 vibrational transitions, respectively. The orange emission has a peak at 588 nm with a vibronic structure at about 632 nm. It is found that the intensity ratio depends on the thickness of ITL. Similar results are obtained for the all-direction EL spectra as shown in Fig. 3. Several different results, however, are found between the normal- and all-direction EL spectra. An example is the intensity ratio of the 0–0 vibrational component to the 0–1 component. The peak-height ratio is 1.10 and 1.54 for the normal- and all-direction blue emission spectra in BR-4, respectively, and 1.56 and 2.08 for the orange emission spectra.

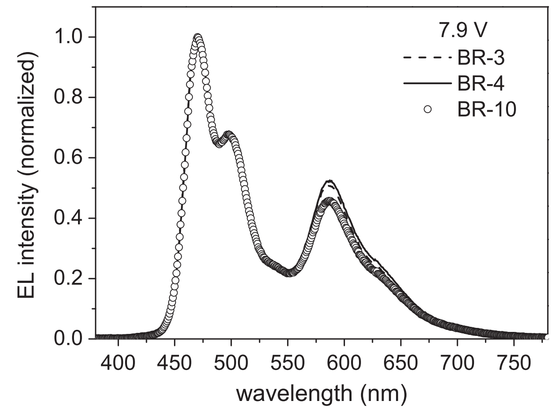

Fig. 3. EL spectra of Devices BR-3, BR-4, and BR-10 at 7.9 V measured in all directions from the device surface.

Such a difference is understood by the microcavity effects due to variation of layer thickness in the OLED devices. The microcavity effects are caused by internal reflection and refraction at the boundary in optical cavity, giving rise to influence to propagation of light generated in EML in multi-layer OLED cavity. One of the effects leads to variation in the emission spectrum from OLED surface with viewing angle [7–9]. The EL spectrum shifts toward short wavelength with increasing the viewing angle changes from 0° to 60° [10,11], indicating that the EL intensity at the shorter wavelength side in an emission band is more reduced in the normal direction observation than the intensity at the longer wavelength side. This is confirmed from Figs. 2 and 3 where the peak height ratio of the 0–0 EL intensity at 472 nm to the 0–1 EL intensity at 498 nm is more reduced in the normal-direction EL (Fig. 2) than in the all-direction EL (Fig. 3). The all-direction EL spectra are quite similar to the PL spectra from the evaporated thin film on quartz plate as shown in Fig. 4. From this figure, it is confirmed that the blue EL spectra measured in the normal direction are strongly deformed by the microcavity effect.

The EL appears at applied voltages above 3 V. The EL spectra at various voltages are shown for BR-3 in Fig. 5, which were measured in the integrated sphere. Both the blue and orange EL intensities increase with increasing the bias voltage. The two intensities, how-

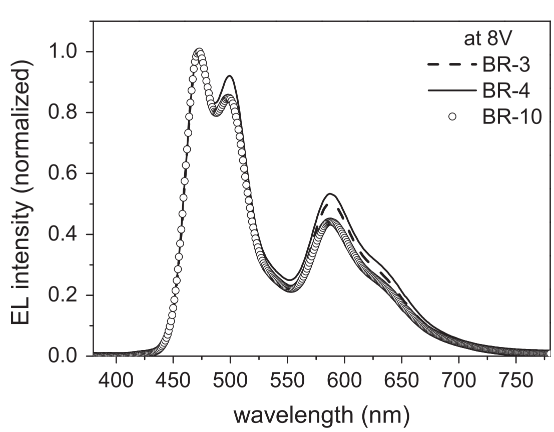

Fig. 2. EL spectra of Devices BR-3, BR-4, and BR-10 at 8 V measured in a normal direction of the device surface.

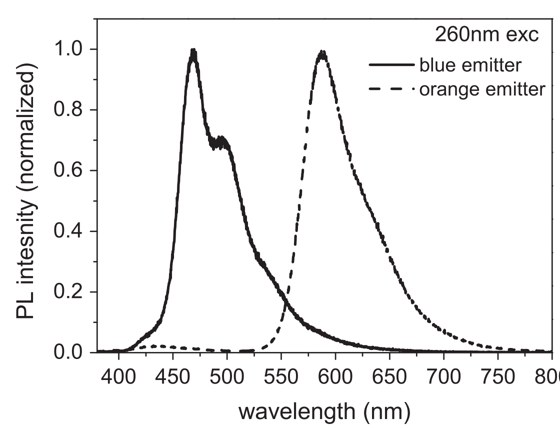

Fig. 4. PL spectra of blue-emitting and orange emitting materials used in OLED devices, which were obtained from films evaporated on quartz. Optical excitation was made at 260 nm.