

Improvement of Emission Color and Efficiency in Blue Light Emitting Organic Devices Using External Microcavity

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ABSTRACT

An external microcavity structure was introduced to the stacked cathode in organic light emitting devices, and the influence on the luminescent characteristics were investigated. From the detail optical analysis, it was clarified that the surface plasmon loss in a metal cathode can be reduced to about one fifth by using a semi-transparent thin film cathode, and can be extracted outside by the microcavity effect after being converted to the thin film waveguide mode. More than 50% of optical power in dipole emission was successfully utilized as the external and substrate modes. The luminous efficiency was increased about 1.4 times, and the color purity of the blue emission was also improved. The relationship between the external microcavity effect and surface plasmon coupling will be discussed from a viewpoint of optical analysis.

KEYWORDS; Organic light-emitting diode, Light extraction, Microcavity, Surface plasmon

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I. INTRODUCTION

Organic light-emitting devices (OLEDs) are expected as a high performance flat panel display because of the excellent image quality and low power consumption. In addition, flexible displays have the potential to create new markets. The internal quantum efficiency of blue OLEDs has approaches to 100 % by the use of phosphorescence [1] or hyper fluorescence [2] of excited molecules. However, the external quantum efficiency (EQE) remains about 20% due to the poor light extraction efficiency [3]. In 2012, the international standard called BT.2020 was announced in an ultra-high definition 8K-TV, in which wide color gamut is recommended in order to achieve high saturation color reproducibility [4]. However, it is difficult with the current OLED technology to satisfy such high performance of emission color because of broad band emission in organic phosphors. In particular, development of a material that satisfies both luminous efficiency and color purity is difficult and significant in blue light emission. One of solution methods is an internal microcavity effect, which is often used for an enhancement of the forward directional emission intensity by the control of optical interference phenomenon in multi-stacked thin film layers [5, 6]. However the problem with this method is that it suffers from the limitations of electrical characteristics because the thickness of the organic layer should be as thin as possible for low voltage operation. We have already reported that the back-cavity structure by using a stacked cathode is useful for the improvement of the device performance in the green OLED [7].

In this paper, we apply such technology to a blue light emitting device and describe the possibility of a high efficiency light emitting device with excellent color purity. The relationship between the external microcavity and device performance will be theoretically explained from the experimental results and optical simulation analysis.

II. EXPERIMENTAL METHODS

Device Structure and Preparation

Figure 1(a) shows a normal device structure, which consists of an indium-tin-oxide (ITO) bottom electrode, a poly(3,4)-ethylenedioxythiophene-polystyrenesulfonate (PEDOT:PSS) hole-injection layer, bis[(1-naphthyl)-N-phenyl]benzidine (NPB) hole-transporting layer, a 4,4'-N,N'-dicarbazole-biphenyl (CBP) emissive layer (EML) doped with bis[2-(4,6-difluorophenyl)pyridinato-C2,N] (picolinato) iridium(III) (Flrpic) as a blue phosphorescent guest, a 2-(4-biphenyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole (Bu-PBD) electron transporting layer (ETL) and an aluminum (Al) cathode. Very thin lithium fluoride (LiF) of about 1 nm is inserted between Al and ETL to enhance electron injection. Figure 1(b) is a proposed device structure with the external microcavity, in which the cathode has multilayers consisting of semi-transparent MgAg thin film metal, an optical buffer layer (OBL) and high reflective silver film. In other words, we call it multi-cathode (MLC). The OBL uses ITO film prepared by rf/dc sputtering method, but other transparent conductive materials are

candidates. Each layer was sequentially deposited by a vacuum evaporation technique. Polymer layer of PEDOT:PSS was formed by spin-coating process.

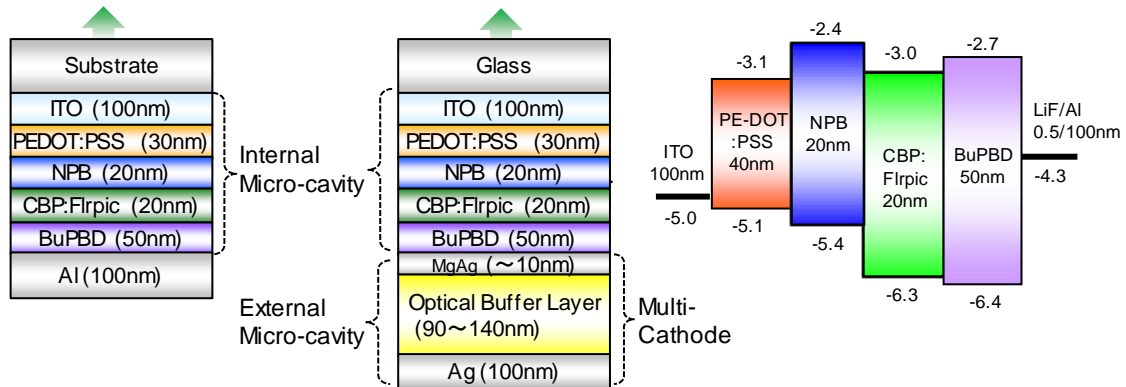


Fig. 1 Device structures of blue light emitting OLEDs with (a) multi-cathode and (b) normal cathode. Energy potential diagram of the normal device was shown in (c).

Optical Analysis

Optical phenomena in an OLED is very complicate because the device consists of nanometer-sized stacked thin film layers. As shown in figure 2, radiation process in an OLED consists of propagation and evanescent modes. Propagation mode is further divided into external, substrate and waveguide modes on the basis of total internal reflection in high refractive index thin film layers. When the dipole is close to the metal cathode, evanescent mode such as SP should be further considered to evaluate exactly the power dissipation in the device [6, 7]. For the evaluation of the optical modes, it is convenient to introduce in-plane wavenumber vector k_h which is defined as a horizontal component of actual wave-vector k_l . If we use an in-plane wave vector, these optical modes can be arranged one-dimensionally as shown in the figure. The external mode will be treated by classical ray-optics. As for the substrate mode, wave-optics of incoherent light will be useful. Waveguide mode is calculated by electro-magnetic optics of coherent light. The SP is related to the near-field optics. Wide range of optics are deeply involved. The optical calculation was carried out by using a software created by the author, which adopted a traditional approach for the dipole model developed to molecular fluorescent and energy transfer near interface on the basis of near-field optics and wave optics [8].

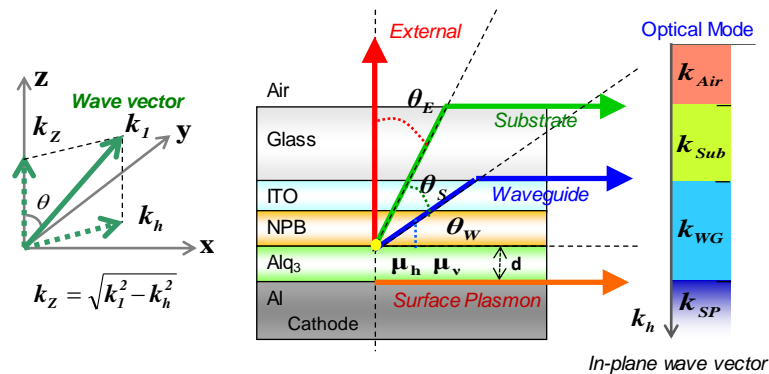


Fig. 2 Relationship between various optical modes in the device and wave vector. Optical phenomena in OLED are composed of various optical processes including a wide wave vector.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Optical Power Distribution

Figure 3(a) and 3(b) show optical power spectra of OLEDs with the normal and MLC structures, respectively, which indicate the optical energy density as a function of in-plane wave-vector k_h . Since k_h is normalized by the wave-vector in the air, the horizontal axis actually corresponds to the refractive index of each material used in the device. Red and blue lines are horizontal and vertical dipoles. In the normal device, the SP peak appears at 1.92 in k_h . It should be noted that the SP peak of the vertical dipole moment is much larger than the horizontal one. In addition, two kinds of sharp peaks signed TE and TM can be observed in the waveguide mode region. Broad substrate and external modes were observed in the horizontal dipole. In contrast, when we

use the MLC structure, the SP mode almost disappears and strong waveguide TM mode and broad substrate modes becomes dominant in the vertical dipole as shown in Figure 3(b).

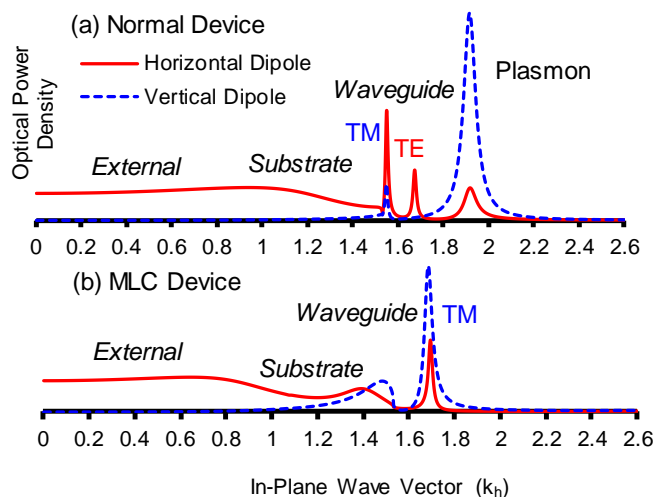


Fig. 3 Optical power spectrum of the dipole emission as a function of in-plane wave vector. (a) Normal device and (b) Multi-cathode device.

Figure 4 shows a variation of optical mode distribution with the film thickness d_{OBL} of OBL in the MLC structure. The thickness of the OBL was changed in the range of 0~200 nm. When the film thickness of OBL is zero, it is a normal structure using an MgAg / Ag cathode. The intensity of external mode greatly increases when the film thickness exceeds 80 nm, and takes a maximum of 24.7% at 100 nm. The SP loss gradually decreases and is about 10% at a film thickness of around 100 nm. It should be noted that the waveguide mode is increasing as the SP loss decreases. This means that the evanescent wave is successfully converted to the propagation mode by using the multi-stacked cathode.

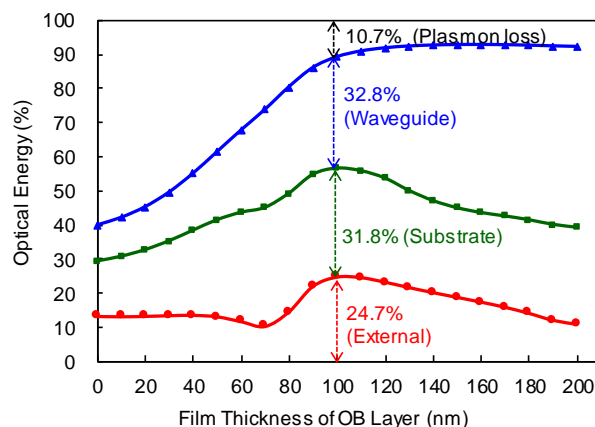


Fig. 4 Variation of optical energy distribution with the thickness of OB layer in the MLC device. As the OB layer gets thicker, the waveguide mode increases because of the decrease of surface plasmon loss.

Dashed and solid lines in Figure 5 show a variation of optical mode ratio with the distance D_{dc} between dipole and metal cathode in the devices with the normal cathode and MLC structure, respectively. Since the dipole is assumed to be located at the interface of EML and ETL, the distance D_{dc} was adjusted by the thickness of ETL. The thicknesses of semi-transparent MgAg and OBL are 10 nm and 100 nm, respectively. The external and substrate modes change periodically with D_{dc} . In the case of the normal device, the external mode takes the first maximum at about 50 nm in D_{dc} . The ratio of optical mode intensities were 18.3%, 19.0%, 9.0% and 53.7% for external, substrate, waveguide and SP modes, respectively. This means that only half of total energy is available for out-coupled emission as a propagation light, even if we carefully design the device structure. The SP loss is responsible for the poor external efficiency. Therefore it is necessary to suppress the SP loss and convert the waveguide mode into the substrate and external modes for the enhancement of light extraction. There are two approaches for reducing the SP loss. One way is to suppress the generation of the plasmon itself,

the other way is to combine it with the propagating wave and return it to radiation. As shown by the blue solid line in Figure 4, the intensity of the SP mode decays exponentially with the increase of D_{cd} and finally decreased to about 10% above 50 nm in D_{cd} . Consequently, the ratio of external, substrate and waveguide modes increases to 24.7%, 31.8% and 32.8%, respectively, which means that the SP loss is successfully converted to propagation wave and approximately 90% of total radiation energy is available for the propagation light. The mode ratio and its difference in both devices are summarized in Table (b) of Fig. 5. The sum of external and substrate modes reaches to about 56% in the MLC device. Especially, it is worth the waveguide mode increases from 9.0 % to 32.8 % by recovering the SP loss to propagation light. It will be expected that the increased waveguide mode is directly coupled with the external microcavity structure. According to the detail optical analysis, it was found that significant reduction of SP loss is caused by the interaction between two kinds of SP coupling on both sides of the MgAg layer. As the film thickness of MgAg becomes thin so that an evanescent wave of dipole emission penetrate in the opposite side of MgAg layer. As a result, long-range and short-range SPs are induced by the coupling of two SP resonance. , and disappear by cancelling each other [9]. The long-range SP can coupled with waveguide mode, and the short-range SP disappears because the wave-vector exceeds the limit of resonance in the material.

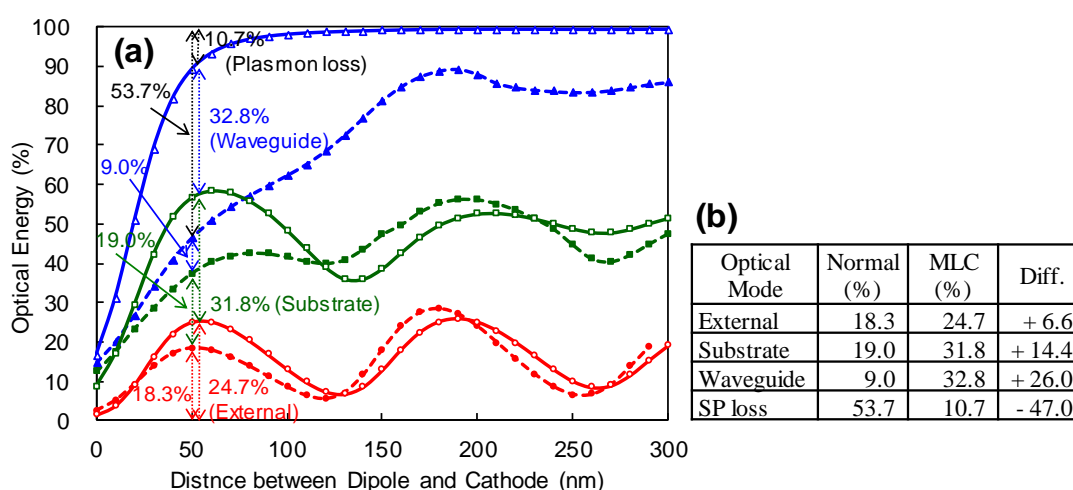


Fig.5 Variation of optical energy consisting of external emission, substrate propagation, thin film waveguide and surface plasmon modes with the distance D_{cd} between dipole and cathode in the devices with normal and multi-cathodes. Optical power ratio of both devices at 50 nm in D_{cd} are shown in the table (b).

Figure 6 shows color images of optical power density distribution in the optimized devices with the normal and MLC structure, which were calculated by finite-difference time-domain method (FDTD) method. Random dipole emission is excited here at the interface between EML and ETL. There is no optical power in the waveguide mode, and the emission distribution in the glass is weak and isotropic in the normal device. In contrast, the waveguide mode is enhanced by the reduction of SP loss in the MLC structure. As a result, a strong forward directional emission is caused by the interference effect of the guided light in the external microcavity structure.

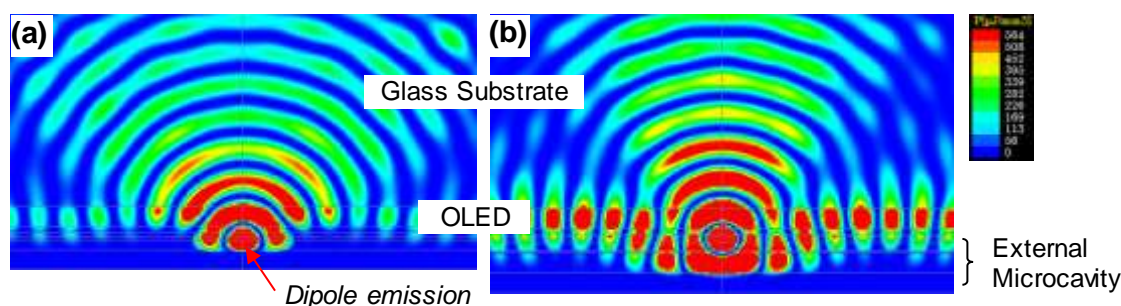


Fig. 6 Color images of optical power density distribution in OLEDs, in which FDTD analysis was used for optical simulation. (a) Normal Structure (b) Multi-Cathode Structure

Emission Color Properties

The normal device already has a weak microcavity between the metal cathode and high refractive-index ITO anode. This is because periodic fluctuations can be observed in the film thickness dependence of the external mode shown in Figure 5. We call it an internal microcavity. The MLC device has the cathode consisting of the transparent OBL sandwiched by two kinds of metals. This configuration can also be expected to produce a microcavity effect. We call it an external microcavity.

Figure 7(a) shows a variation of normalized emission spectra of the MLC device with the film thickness d_{OBL} of OBL in the range of 80~140 nm. The organic phosphor used has a blue light emission having an emission peak near a wavelength of 460 nm and a shoulder band on the long wavelength side. The relative intensity of the shoulder band and the half width of the spectrum strongly depend on the film thickness. As a result, the chromaticity coordinates (u' , v') of the light emission change as shown in Figure 7(b) and the color purity of the blue emission can be greatly improved by adjusting d_{OBL} . This result means that the MLC structure also acts as an external cavity structure, and the increase of the waveguide mode is effectively utilized.

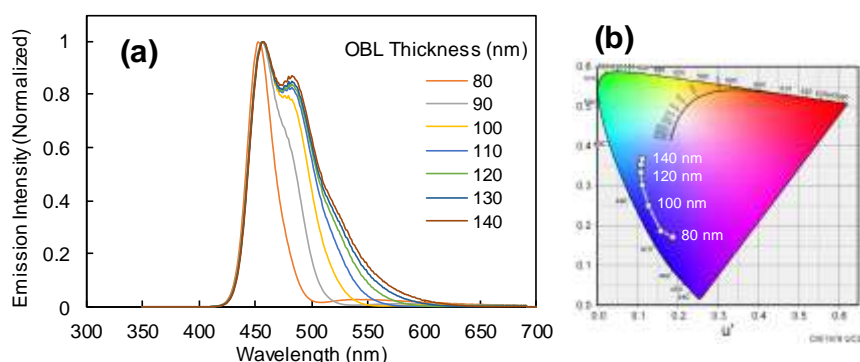


Fig. 7 Film thickness dependence of the emission spectra in the devices with multi-cathode structure. (a) Variation of the emission spectra with the film thickness of OBL (b) Color coordinates in the CIE 1976 UCS color diagram (u' - v').

Figure 8(a) and 8(b) show dependence of electroluminescent spectra in the blue light emitting device with and without the MLC structure, respectively. The normal device exhibits the blue emission with a peak wavelength of 460. The emission color is light blue because of the slight shoulder band around 490 nm. Angular dependence of the emission intensity shown by the semicircle in the figure is nearly Lambertian. In contrast, the half width of the emission spectrum is much narrower in the MLC device because the strength of the shoulder band is rather suppressed. As a result, the emission color is changed to a pure blue. Figure 8(c) shows viewing angle dependence of the chromaticity coordinates (u' , v') in both devices. In addition, forward directional emission is another feature in the MLC structure which suggests that the external microcavity effect works well.

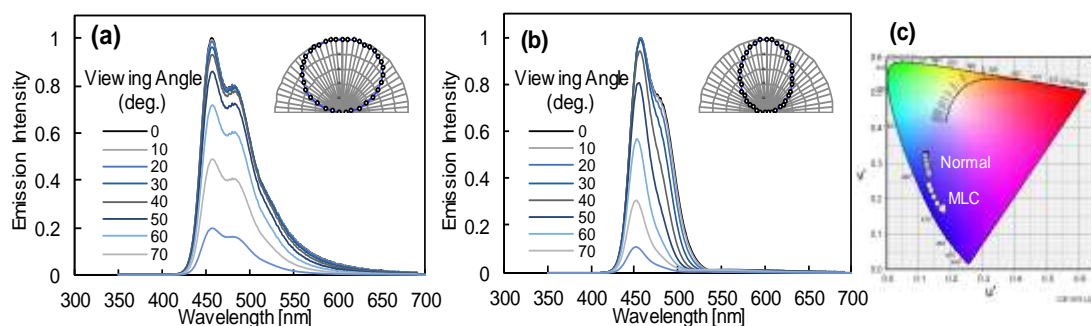


Fig. 8 Viewing angle dependence of electroluminescent spectrum and emission intensity in the green OLEDs with normal and MLC structures. (a) Normal Device (b) Multi-Cathode Device (c) Color coordinates in the CIE 1976 UCS color diagram (u' - v')

Emission Efficiency

The influence of the MLC structure and external microcavity effect on the emission efficiencies was investigated experimentally by preparing the phosphorescent blue light emitting OLEDs. Figure 9 shows the dependence of EQE, power efficiency and luminance on current density in the devices with different structures. One is a reference device with the normal cathode (device (A)) and the others are MLC devices with and without half-sphere lens (devices (B) and (C)). The film thickness of each layer was adjusted to optimum values

obtained from optical analysis described in the previous section. A half-sphere lens with the same refractive index as the glass was mounted via index-matching oil on the substrate to measure the total emission of both the external and substrate mode in the device (C). The MLC device exhibits much higher efficiency and luminance compared to the normal device. Table 2 shows the comparison of EQE and power efficiency in three devices. Maximum values of EQE in low current region are 18.2% and 26.1% in the devices (A) and (B), respectively. The maximum EQE becomes higher by a factor of 1.4 by introducing external microcavity in the MLC structure. When we use a half-sphere lens in the MLC device, a maximum EQE increases up to 54.2% because an optical power in the substrate can be also extracted out to the air. According to the calculation results shown in Table 1, the sum of external and substrate modes is 56.5 % in the MLC device. There is almost agreement between the experimental and simulated results.

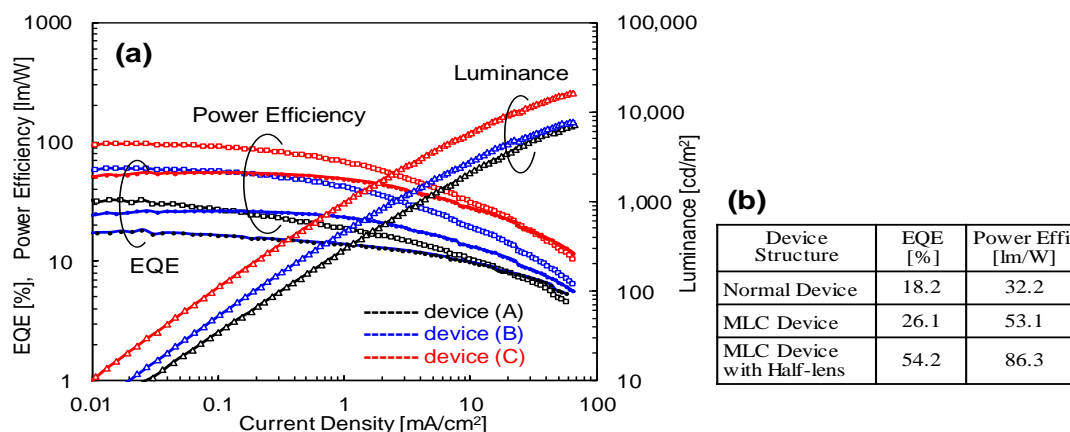


Fig. 9 External quantum efficiency (EQE), power efficiency and luminance as a function of current density in the devices with normal and MLC structures. (A) Normal device (B) MLC device (C) MLC device with a half sphere lens on the glass. Maximum EQE and power efficiency in these devices are compared in Table (b).

IV. CONCLUSION

The effect of external microcavity on the emission properties has been investigated in the blue light emitting OLED. It was clarified that the MLC structure increases the power ratio of the waveguide mode resulting in the enhancement of microcavity effect on the luminescent properties. The emission spectra becomes sharp so the color purity of the blue emission is greatly improved. In addition, the external quantum efficiency is increased by a factor of 1.4, which originates in the reduction of SP loss accompanied with increased propagation mode. This optical technique is useful for the improvement of the emission color and efficiency without sacrificing an electrical property.

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