Highly efficient and wide-color-gamut organic light-emitting devices using external microcavity

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ABSTRACT
An external microcavity structure was introduced to the stacked cathode in organic light-emitting devices (OLEDs), and its influence on the luminescent characteristics was investigated. From the detailed optical analysis that was conducted, it was found 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 through the microcavity effect after being converted to the thin-film waveguide mode. No less than 50% of the optical power in dipole emission was successfully utilized as the external and substrate modes. The luminous efficiency increased about 1.4-fold, and the color purity of the luminance also improved. As a trial preparation of red, green, and blue OLEDs with the external microcavity structure, the color gamut improved greatly and approached the BT.2020 national standard. The effect of the external microcavity will be discussed from the viewpoint of multi-scale optical analysis.

1. Introduction
Organic light-emitting devices (OLEDs) are expected as a high-performance flat-panel displays because of their excellent image quality and low power consumption, including the possibility of coming up with flexible and transparent panels. The internal quantum efficiency of OLEDs approaches 100% through the use of the phosphorescence [1] or hyperfluorescence [2] of excited molecules. The external quantum efficiency (EQE), however, 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 a wide color gamut is recommended to realize higher color reproduction [4]. It is difficult, however, to satisfy such high performance of emission color with the current OLED technology. One of the solution methods is an internal microcavity effect, which is often used for the enhancement of the forward directional emission intensity by controlling the optical interference phenomenon in multi-stacked thin-film layers [5,6]. The problem with this method, however, is that it suffers from the limitations of the electrical characteristics because the organic layer should be as thin as possible for low-voltage operation. It has already been reported that the high-refractive-index glass and a weak microcavity effect are useful for the improvement of the out-coupling efficiency in OLEDs [7]. Furthermore, a back-cavity structure using a stacked cathode has been proposed, and an improved device performance in the green OLED has been demonstrated [8]. In this paper, the synergistic effect between the multi-cathode and an external microcavity on OLEDs with three primarily color emissions is systematically revealed. As a result, the color gamut in the blue, green, and red emissions improved 1.7-fold. In addition, the emission efficiency becomes higher without sacrificing the electrical characteristics. The relationship between the external microcavity and the device performance will be theoretically explained based on the results of the experiment and optical simulation analysis.

2. Experiment method
2.1. Device structure and sample preparation
Figure 1(a) shows a normal device structure consisting of an indium-tin-oxide (ITO) bottom electrode, a polyc(3,4)-ethylendioxy thiophene-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 a color-emitting guest, a 2-(4-biphenyl)-5-(4-tert-butylphenyl-1,3,4-oxadiazole) (Bu-PBD) electron-transporting layer (ETL), and an aluminum cathode. Phosphorescent materials like Flrpic, Ir(ppy)$_3$, and Ir(pic)$_2$acac are used as color dopants for blue, green, and red emission. Figure 1(b) is a proposed device structure with the external microcavity, in which the cathode has three layers consisting of a semi-transparent MgAg thin-film metal, an optical buffer (OB) layer, and a highly reflective silver film. As such, it is called ‘multi-cathode (MLC)’. The OB layer uses an ITO film prepared via the RF/DC sputtering method, but other transparent conductive materials are candidates. The thickness of the OB layer was changed to within the range of 90–140 nm depending on the emission color. Each layer was sequentially deposited through a vacuum evaporation technique. A polymer layer of PEDOT:PSS was formed via spin-coating. The normal device already has a weak microcavity between the metal cathode and the high-refractive-index ITO anode. It is called an ‘internal microcavity’. In addition, the proposed device has a cathode consisting of a transparent OB layer sandwiched by two kinds of metals. It is called an ‘external microcavity’.

2.2. Optical process and analysis of the surface plasmon

The optical phenomenon in an OLED is very complicated due to the stacked thin-film structure of the wavelength size. The radiation process in an OLED consists of the propagation and evanescent waves, which are generally divided into the external, substrate, waveguide, and surface plasmon (SP) modes [6]. The sum of the external and substrate modes, however, is only less than half even if the device shown in Figure 1(a) is carefully designed, due to the large losses induced by the waveguide and SP modes. For light extraction enhancement, it is necessary to suppress the SP loss and convert the waveguide mode to the substrate and external modes. Optical calculation was carried out using a software created by the author, which adopted a traditional approach for the dipole model developed for molecular fluorescence and energy transfer near the interface, based on near-field and wave optics [9].

Figure 2 shows an optical process including the SP mode in OLEDs. It is known that dipole emission has three recombination paths consisting of radiative recombination, non-radiative recombination, and the excitation of SP on the metal cathode. As phosphorescent materials have a nearly 100% quantum yield, the non-radiative recombination path is negligible. The SP mode will generally result in an ohmic damping in a short lifetime. Re-emission from the SP is also possible, however, when the horizontal wave vector of the SP mode matches that of the waveguide mode. Thus there are two approaches for reducing the SP loss. One is to suppress the generation of the plasmon itself, and the other is to combine it with the propagating wave and return it to the radiation. The former method involves the use of a thick electron-transporting layer to separate the dipole and the metal, but this is not desirable because it involves an increase in drive voltage. In this work, re-emission of the SP mode was used, and subsequently coupling with the microcavity effect.

3. Experiment results and discussion

3.1. Optical power distribution

Figure 3(a) and (b) shows the 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 $k_h$. As $k_h$ is normalized by the wave vector in the air, the horizontal axis actually corresponds to the refractive index of each of the materials used in the device. The red and blue lines are horizontal and vertical dipoles. In the normal device, the SP peak appears at 1.82 in $k$-space. It should be noted that the SP peak of the vertical dipole moment is much larger than that of the horizontal one. In addition, two sharp peaks can be observed in the waveguide mode region. The broad substrate and external modes can also be observed...
in the horizontal dipole. In contrast, the SP mode almost disappears, and instead, the waveguide TM and broad-substrate modes become dominant in the MLC device, as shown in Figure 3(b). This means that the evanescent wave is successfully converted to the propagation mode by employing the multi-stacked cathode.

The dashed and solid lines in Figure 4 show a variation of optical mode ratio with distance $D_{dc}$ between the dipole and metal cathode in the devices with the normal cathode and MLC structure, respectively. As the dipole is assumed to be located at the interface of EML and ETL, distance $D_{dc}$ was adjusted based on the thickness of ETL. The thicknesses of the semi-transparent MgAg and OB layers are 10 and 120 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 60 nm in $D_{dc}$. The ratios of the optical mode intensities were 22.7%, 22.0%, 6.4%, and 48.9% for the external, substrate, waveguide, and SP modes, respectively. This means that the SP loss is dominant and that only less than half of the total energy is available for out-coupled emission as a propagation light, even if the device structure is carefully designed. As shown by the solid lines in Figure 4, the intensity of the SP mode decays exponentially with the increase of $D_{dc}$, and finally decreases to less than 10% at above 60 nm in $D_{dc}$. Consequently, the ratios of the external, substrate, and waveguide modes increase to 28.1%, 32.0%, and 30.8%, respectively, which means that the SP loss is successfully converted to propagation wave and approximately 90% of the total radiation energy is available for the emission. The mode ratio and its difference in both devices are summarized in Table 1. The sum of the external and substrate modes reaches 60% in the MLC device. Especially, the waveguide mode increases from 6.4% to 30.8% by recovering the SP loss to propagation light. It is expected that the increased waveguide mode will be directly coupled with the external microcavity structure. In the detailed optical analysis that was conducted, it was found that a significant reduction of the SP loss is caused by the interaction between two kinds of SP coupling on both sides of the MgAg layer as the MgAg film becomes so thin that the evanescent wave of dipole emission reaches the opposite side of the MgAg layer. As a result, long- and short-range SPs are induced, and later disappear by cancelling each other [10].

Figure 5 shows images of the optical power density distribution in optimized devices with the normal and MLC structures, which were calculated through the finite-difference time domain (FDTD) method. The enhanced waveguide mode is caused by the conversion from plasmon by the MLC structure. Furthermore, a strong forward-directional emission is caused by the interference effect of the guided light in the external microcavity structure.

### 3.2. Emission efficiency and spectrum

The influence of the external microcavity effect on the emission characteristics was experimentally investigated by preparing phosphorescent green OLEDs. Figure 6 shows the dependence of the EQE, power efficiency,
Figure 5. Optical power density distribution in OLEDs with (a) a normal cathode and (b) a multi-cathode. The finite-difference time domain (FDTD) method was used for optical calculation.

Figure 6. EQE, power efficiency, luminance, and applied voltage as function of the current density in devices with normal and MLC structures: (a) normal device, (b) MLC device, (c) MLC device with a half-sphere lens on the glass.

Table 2. External quantum efficiency (EQE) and power efficiency at current density of 0.01 mA/cm² in the devices with normal and MLC cathodes.

| Device structure | EQE (%) | Power efficiency (lm/W) |
|------------------|---------|-------------------------|
| Normal device    | 21.9    | 84.9                    |
| MLC device       | 30.0    | 116.6                   |
| MLC device with half-lens | 56.9 | 200.2                   |

When a half-sphere lens is used in the MLC device, the maximum EQE increases up to 56.9% because the optical power in the substrate can also be extracted and released in the air. According to the calculation results shown in Table 1, the sum of the external and substrate modes in the MLC device is 60.1%. There is almost an agreement between the experiment and simulation results.

Figure 7(a) and (b) shows the viewing angular dependence of the electroluminescent spectra in the green-light-emitting device with the normal and MLC cathodes, respectively. The normal device exhibits the green emission with a peak at the 515 nm wavelength. The emission color is light green due to the slight shoulder band around 550 nm. The angular dependence of the emission intensity shown by the semicircle in the figure is nearly Lambertian. In contrast, the MLC device exhibits a single sharp emission band with a peak at 515 nm. Forward-directional emission is another feature, suggesting that the external microcavity effect works well. The emission color is changed to pure green. Similar effects have been observed for blue and red OLEDs, and will be published in detail in a separate paper.

In addition to the green OLED, the effect of the external microcavity in blue- and red-emitting devices was also examined. Organic materials of Flrpic and Ir(pic)2acac were used as color dopants, respectively. The film thickness of the OB layer in the cathode was optically adjusted according to the emission wavelength to optimize the interference effect. Figure 8(a) shows the electroluminescent spectra of three primary colors, including the green emission, as shown in Figure 7. The solid and dashed lines represent the MLC and normal...
Figure 7. Viewing angle dependence of the electroluminescent spectrum and emission intensity in green OLEDs with normal and MLC structures.

Figure 8. Change of the emission spectra and chromaticity coordinates of red, green, and blue OLEDs with and without external microcavity: (a) electroluminescent spectra of three primary-color OLEDs with normal and MLC structures and (b) color gamut in the CIE chromaticity diagram, including the BT.2020 standard values.

Table 3. Chromaticity coordinates of red, green, and blue light-emitting devices with MLC and normal cathodes. BT.2020 international standard values are shown in comparison.

| Color | MLC       | Normal  | BT.2020 |
|-------|-----------|---------|---------|
| Red   | 0.657, 0.307 | 0.617, 0.379 | 0.708, 0.292 |
| Green | 0.161, 0.685 | 0.326, 0.621 | 0.170, 0.797 |
| Blue  | 0.136, 0.118 | 0.175, 0.266 | 0.131, 0.046 |

The color gamut improved about 1.7-fold through the use of the external microcavity, and approached the BT.2020 national standard. These results can be successfully explained as follows. The MLC structure makes it possible to reduce the huge SP loss in the cathode, and the evanescent mode is directly converted to the propagation wave. As a result, the external microcavity acts effectively for light extraction and the sharp emission band.

4. Conclusion

The effect of the external microcavity on the emission properties was investigated in the color OLEDs. The SP loss is successfully suppressed and converted to the waveguide mode by using the MLC structure. Approximately 60% of the optical power in the dipole emission can be converted to the glass and air modes. It was clarified that the MLC structure increases the power ratio of the waveguide mode, resulting in an enhanced microcavity effect on the luminescent properties. The emission spectra become sharp, and the color purity greatly improves. As a result, the color gamut of the three primary emissions expands and approaches the BT.2020 standard. In addition, the EQE is increased by...
a factor of 1.4. This optical technique is useful for the improvement of the emission color and efficiency without sacrificing an electrical property.

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**Notes on contributor**

Akiyoshi Mikami received his Doctorate in Electrical Engineering in 1985 from Osaka City University. Subsequently, as a researcher at Sharp Corporation, he worked in Central Research Labs and Liquid Crystal Labs for 14 years. In 1999, he became a professor at Kanazawa Institute of Technology, where he engaged in electronic and optical engineering research. His recent research field is the optical design theory and technology for improving the performance of organic light-emitting devices.

**References**

[1] M.A. Baldo, S. Lamansky, P.E. Burrow, M.E. Thompson, S.R. Forrest, Appl. Phys. Let 75, 4–6 (1999).
[2] H. Uoyama, K. Goushi, K. Shizu, H. Nomura, C. Adachi, Nature 11687 (2012).
[3] D. Tanaka, H. Sasabe, Y.J. Li, S.J. Su, T. Takeda, J. Kido, Jpn. J. Appl. Phys. 46 (1), L10–L12 (2007).
[4] Rec. ITU-R BT.2020, Parameter values for ultra-high definition television system for production and international programme exchange. Aug. (2012).
[5] V. Bulovic, V.B. Khalffin, G. Gu, P.E. Burrowst, Phys. Rev. B 58 (7), 3730–3740 (1998).
[6] K.A. Netz, J. Opt. Soc. America A 15 (4), 962–971 (1998).
[7] A. Mikami, Phys. Status Solidi C8 (9), 2899–2902 (2013).
[8] A. Mikami, S. Doi, Soc. for Inform. Disp., International Symposium, Digest of Tech. Papers, San Jose, P171, 1695-1698, (2015).
[9] R.R. Chance, A. Prock, R. Sibey, Adv. Chem. Phys. 37, 3 (1978).
[10] A. Mikami, Opt. Photonics J. 6, 226–232 (2016).