Hybrid Tamm plasmon-polariton/microcavity modes for white top-emitting organic light-emitting devices

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White top-emitting organic light-emitting devices (WTOLEDs), emitting white light through a transparent top metallic electrode, have emerged as promising candidates as energy-efficient solid-state lighting sources and full-color flat-panel displays. The microcavity effect due to usage of metallic electrodes results in emission enhancement solely at a particular color, and therefore sets an obstacle for WTOLEDs, where at least two colors with balanced intensity should be emitted. Current efforts solving the problem basically rely on the relaxation of the microcavity effect, resulting in sacrificed light outcoupling efficiency in the original resonance region. Here, we demonstrate that by integrating a photonic crystal structure upon the top metallic electrode, an additional emission enhancement peak other than the one determined by the microcavity resonance could be provided by the Tamm plasmon-polariton mode. Mode hybridization induced dual hybrid modes with comparable light outcoupling efficiency can then be excited, from which two colors with balanced intensity could be emitted. Both experimental and theoretical results demonstrate that the proposed mode hybridization strategy may pave the way for the realization of WTOLEDs towards high white color quality, improved viewing characteristics, and electroluminescence efficiency. © 2015 Optical Society of America

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1. INTRODUCTION

White organic light-emitting devices (WOLEDs) have great potential as energy-efficient solid-state lighting sources and full-color flat-panel displays [1–6]. Top-emitting OLEDs (TOLEDs), emitting light through a transparent top electrode, are attractive as they are independent from the substrate used [7–13], particularly when a driving circuit needs to be accommodated. However, realizing white emissions from TOLEDs has encountered challenges in high quality of the white color and high efficiency. Thin metallic films are generally chosen as the semi-transparent top cathode in white TOLEDs (WTOLEDs) [5], while the bottom and top metallic electrodes together with organic layers form a Fabry–Perot resonator. The resulting strong microcavity (MC) effect induces a narrow bandwidth of the electroluminescence (EL) spectrum as well as a blue shift of the emission color as the viewing angle increases [14]. These two features have been obstacles for the applications of WTOLEDs in both displays and lightings.

Current photon management designs to overcome these obstacles are mainly based on the idea of broadening the MC resonance, such as by introducing a capping layer on top of the thin metallic cathode [8,9], integrating a microstructured cavity with micrometer-scale periodicity [10], and using ultra-thin metallic electrodes [11]. In addition to the negative effect of narrowed bandwidth and angle-dependent viewing colors, however, the MC resonance has a positive effect on the light outcoupling efficiency in the resonance region [14]. All the aforementioned designs were taking effect through suppressing the MC effect, which may lead to a lowered light outcoupling efficiency. Designs based on introducing multiresonances were also proposed by increasing the cavity length [12], but the device may suffer degradation of the electrical performance due to the much increased thicknesses of the organic layers. So far, designing WTOLEDs with satisfied optical and electrical performance is still a challenge, and further efforts are required to realize both angle-independent broadband emissions and improved light outcouplings.

In this work, we demonstrate a simple and effective strategy for WTOLEDs with the improvement in both viewing characteristics and light outcouplings. Figure 1 shows a schematic of the
proposed strategy based on hybrid Tamm plasmon-polariton/microcavity (TPP-MC) modes. In addition to the emission enhancement peak determined by the MC resonance, we introduce a second emission enhancement peak via the excitation of a Tamm plasmon-polariton (TPP) mode [15–17], a surface state supported at the interface between the thin metallic cathode and an introduced one-dimensional (1D) photonic crystal (PC) structure. Two TPP-MC hybrid modes can then be excited through the mode hybridization between the intrinsic MC resonance and the introduced TPP mode. The two hybrid modes excited on resonance (that is, with zero detuning wavelength) are especially suitable for use in the WTOLEDs based on the two-complementary-color strategy, since they exhibit comparable light outcoupling efficiency, while the resonant wavelengths could be simply designed to well match the intrinsic emission peaks of the WTOLEDs. Therefore, no special structure adjustment has to be done to redress the white EL spectra. Simultaneous improvements of viewing characteristics and efficiency have been obtained for the WTOLEDs based on the mode hybridization strategy.

2. RESULTS

A. Hybrid Mode Formation by TPP-MC Coupling

We first investigate the mechanism of mode hybridizations. Figure 2(a) shows the configuration of a TPP-MC structure, in which a 1D PC structure consisting of double periods of ZnSe (24 nm) /CaF$_2$ (98 nm) is evaporated consecutively on top of a MC structure of Ag/tris-(8-hydroxyquinoline) aluminum (Alq$_3$)/Ag. The MC structure supports a bare MC resonance, which shifts to red as the cavity length increases, while the bare TPP mode can be supported at the Ag/PC interface [15–17] and independent of the cavity length (see Supplement 1). As a result, the excitation of both the MC resonance and the TPP mode could be expected in the investigated TPP-MC structure. Figure 2(b) shows the corresponding experimental reflectivity spectra in the normal direction, where two resonant dips emerge and shift to red as the cavity length increases. To explain the origin of the two resonances, we show in Fig. 2(c) the simulated TE-polarized reflectivity dispersion diagram obtained by the transfer matrix method [18]. The dispersions of the bare MC resonance and the bare TPP mode (see Supplement 1) are also shown by the dashed curves. We find an anticrossing behavior [19,20] between the two resonances supported in the TPP-MC structure, indicating that each resonance is a TPP-MC hybrid mode resulting from the mode hybridization between the MC resonance and the TPP mode.

The mode hybridizations in this work are categorized as the on-resonance condition and the off-resonance condition based on the detuning wavelength, that is, the difference of the resonant wavelength between the bare MC resonance and the bare TPP mode. More specifically, the term “on-resonance” is used for a condition in which the detuning wavelength is zero, such as in the case of a ~100-nm-thick cavity in Fig. 2(c). The corresponding profiles of the electric-field-intensity enhancement $|E_j/E_0|$ ($E_0$ corresponds to the intensity of incidence) are illustrated in Fig. 2(d) [18]. We find that the two TPP-MC hybrid modes located at ~450 and ~570 nm manifest themselves through similar mode characteristics, that is, with field enhancements both inside the cavity and at the metal/PC interface. In contrast, the term “off-resonance” is used for a condition with a relatively larger non-zero detuning wavelength. The characteristics of the induced two
TPP-MC hybrid modes can then be highly distinct, such as in the case of a ~140-nm-thick cavity in Fig. 2(c) [18]. We note the field profile of the longer resonance more resembles that of a MC resonance, because its dispersion curve is close to that of the bare MC resonance [Fig. 2(d)]. For the same reason, the shorter resonance behaves more like a TPP mode.

It is well known that light outcouplings could be enhanced in TOLEDs via the excitation of the MC resonance [14]. However, attempts were usually made to alleviate the MC resonance in WOLEDs, because broadband light outcouplings can hardly be accomplished by single resonant mode due to its relatively narrow bandwidth (see Supplement 1). The proposed mode hybridization strategy introduces two TPP-MC hybrid modes, and their resonant wavelengths could be tuned simply by varying the parameters of the device structure (see Supplement 1 for an analytical demonstration by a coupled oscillator model as well as the numerical results for the demonstration of the tunability of the hybrid TPP-MC mode). For the design in Fig. 2(c), the two hybrid modes excited on resonance are located at ~450 and ~570 nm, corresponding to the blue and orange emission regions, respectively. Moreover, we will show later that under such conditions the two TPP-MC hybrid modes could exhibit comparable light outcoupling efficiency. Therefore, both good white color quality and high outcoupling efficiency could be expected by employing the TPP-MC structure in two-complementary-color-strategy-based WOLEDs.

### B. WOLEDs based on the Mode Hybridization Strategy with Maintained White Color Quality

On the basis of the mode hybridization strategy described above, we design a two-complementary-color-based WOLED employing the TPP-MC structure that is shown as device TPP-MC in Fig. 3. The white emission in this WOLED could be realized by doping one phosphorescent orange-light-emitting complex into a suitable deep-blue-emitting fluorescent complex as the emissive layer [21]. For reference, we fabricate a bottom-emitting WOLED (device R) with indium-tin-oxide (ITO) as the anode, in order to verify the white color quality from the emissive layer. In addition, a conventional microcavity-based device MC and a capping layer strategy [8–9] based device C-MC will be investigated. We will also consider a virtual device TPP (not shown in Fig. 3) in the following simulations, whose structure is the same as that of device TPP-MC but without the anode. The bare TPP mode can be supported in the virtual device TPP.

We first investigate the EL spectra of the above devices. The total cavity length (~100 nm) and PC thicknesses are chosen as that in Fig. 3 so that the on-resonance condition can occur in the device TPP-MC, which can be confirmed by the experimental reflectivity spectra in Fig. 4(a) as well as the calculated dispersion diagrams in Fig. 5(a) [18]. We note that the two TPP-MC hybrid modes match the blue and orange emission regions fairly well. More importantly, we demonstrate that the two hybrid modes excited on resonance exhibit comparable light outcoupling efficiency, which could be evaluated by the calculated spectral radiant intensity in Fig. 5(b) [22]. As a result, the device TPP-MC shows consistence in the EL spectrum, especially the intensity ratio between the two peaks, with the device R [Fig. 5(c)], since the EL spectrum is determined by both the intrinsic white emission from the emissive layer (which can be considered as purely white for all the devices due to the appropriate doping rate) and the outcoupling efficiency (or spectral radiant intensity). For comparison, the device TPP-MC under the off-resonance condition is also investigated by varying the thicknesses of the organic and PC layers (NPB, 40 nm; Bepp2, 45 nm; ZnSe, 19 nm; CaF2, 79 nm). Such increment of the cavity length (~110 nm) together with the decrement of the PC thicknesses can turn the detuning wavelength from zero to nonzero, while at the same time maintaining the two resonant wavelengths, which still well match the blue and orange emission regions [Figs. 4(b) and 5(d)]. In this case, however, the two TPP-MC hybrid modes exhibit distinct light outcoupling efficiency [Fig. 5(e)] [22]. Therefore, the EL spectrum of the device TPP-MC is no longer consistent with that of the device R [Fig. 5(f)], showing yellow–white emission only. We also note that for the device MC without the broadband light outcoupling strategy, the EL spectra are dominated by the MC resonance [Figs. 5(c) and 5(f)], resulting in greenish emissions.

The white color quality of these devices can be evaluated by the Commission International de L’Eclairage (CIE) coordinates (Fig. 6). We see that the CIE coordinates of the device TPP-MC are (0.35, 0.35) under the on-resonance condition, indicating a pure white emission. However, they are only (0.42, 0.42) under the off-resonance condition. The corresponding photographs of the operating devices are also shown in the inset of Fig. 6. The distinctions between the on-resonance and off-resonance conditions could be interpreted from the viewpoint of mode characteristics. The outcoupling efficiency in WOLEDs via the bare MC resonance and the bare TPP mode should be distinct because of their different intrinsic mode characteristics.
This is applicable to the off-resonance condition, where the TPP-MC hybrid mode located at the longer and shorter wavelength behaves more like the bare MC resonance and the bare TPP mode [see Figs. 2(c) and 5(d)], respectively. The outcoupling efficiency via the bare MC resonance is higher than that via the bare TPP mode, which results in the higher intensity of the longer wavelength observed in both the experimentally measured EL spectrum [Fig. 5(f)] and the calculated spectral radiant intensity [Fig. 5(e)] for the device TPP-MC. Such distinctions can be compensated by introducing the on-resonance condition, which turns the bare modes (MC resonance and TPP mode) into dual TPP-MC hybrid modes with comparable light outcoupling efficiency [Fig. 5(b)] since they are both half-MC and half-TPP (see Supplement 1 for a demonstration of the hybrid mode characteristics using a Hopfield model). The maintained white EL spectrum means that no special structure adjustment has to be done to redress the spectrum, which is important for the real application of the mode hybridization strategy. In the following, we study the device TPP-MC under the on-resonance condition only.

C. WTOLEDs based on the Mode Hybridization Strategy toward Improved Viewing Characteristics

As one of the key issues for WTOLEDs, we examine the viewing characteristics of the proposed device TPP-MC. For comparison, the device C-MC based on the conventional capping layer strategy is also studied, as it is the most commonly used configuration for WTOLEDs to improve the viewing characteristics. Their angular EL spectra are shown in Figs. 7(a) and 7(b). We note that the two emission peaks in the device TPP-MC both exhibit a slight blue shift as the viewing angle increases, but their intensity ratio is almost invariant. This feature results in a small variation of the CIE coordinates, that is, from (0.35, 0.35) at 0° to (0.31, 0.34) at 60°. Such CIE variation is almost comparable to that of the device C-MC [Fig. 7(c)], in which as the viewing angle increases, there is an obvious change of the intensity ratio between the two emission peaks [Fig. 7(b)]. The mechanism difference between the mode hybridization strategy and the capping layer strategy can be clearly seen from the calculated spectral radiant intensity for the two devices at different viewing angles in Fig. 7(d) [22]. The introduction of a capping layer in the device C-MC results in a broadened but weakened MC resonance, which still exhibits the angular blue shift, inducing a growing proportion of the blue emission [Fig. 7(b)]. In contrast, the CIE coordinates would be less affected by the slight blue shift of the two resonances in the device TPP-MC, owing to the nearly unchanged ratio between the blue and orange peak intensities [Fig. 7(a)]. Note that the shift range of the peaks of the EL spectrum with the increase of the viewing angle in OLEDs is always smaller than that of the calculated outcoupling efficiency (or spectral radiant intensity) spectrum due to the
invariant intrinsic emission spectrum from the emissive layer. Here the simultaneous blue shift of the two emission peaks in the device TPP-MC further confirms the formation of the two TPP-MC hybrid modes. The angular emission characteristics of the device R and MC are also investigated, with the EL spectra and corresponding CIE coordinates shown in Supplement 1 and Fig. 7(c), respectively. From this comparison it can be concluded that the proposed WTOLED with the mode hybridization strategy shows comparable viewing characteristics to those of the ITO-based WOLED.

D. WTOLEDs based on the Mode Hybridization Strategy toward Improved EL Efficiency

High EL efficiency could be expected in the WTOLED based on the mode hybridization strategy, considering the simultaneous outcoupling enhancement in the blue and orange emission regions. The current density-luminance and luminance-power efficiency characteristics of the device TPP-MC and the reference ones are shown in Figs. 8(a) and 8(b), respectively. As expected, the device TPP-MC exhibits the highest luminance and power efficiency. The maximum luminance is \(\sim 43,450 \text{ cd m}^{-2}\) at the current density of \(\sim 760 \text{ mA cm}^{-2}\), while the highest power efficiency is \(\sim 16.01 \text{ lm W}^{-1}\), which corresponds to enhancement of 11.6%, 114.9%, and 46.1%, respectively, compared to that of the device R \(\sim 14.35 \text{ lm W}^{-1}\), the device MC \(\sim 7.45 \text{ lm W}^{-1}\), and the device C-MC \(\sim 10.96 \text{ lm W}^{-1}\). The external quantum efficiency (EQE) of the device TPP-MC is also higher than that of the device C-MC [inset of Fig. 8(b)]. The two hybrid resonances in the mode hybridization strategy show superiorities in matching the light outcoupling enhancement peaks to the intrinsic white emissions, and therefore the device TPP-MC exhibits the highest efficiency as a result of efficient light outcouplings.

3. CONCLUSIONS

To conclude, we have proposed a novel light outcoupling strategy based on mode hybridization induced TPP-MC hybrid modes, which shows benefits to both the electrical and optical performance of the WTOLEDs. From the viewpoint of electrical performance, the strategy is performed by introducing a PC structure outside the device structure, which would not affect the intrinsic electrical performance. Moreover, the planar PC structure can be fabricated simply via thermal evaporations. This is superior to the light outcoupling strategies performed by adjusting the internal structure of the OLEDs, such as those by employing periodic gratings and nanoparticles, where the electrical performance may be deteriorated. From the viewpoint of optical performance, the tunability of the TPP-MC hybrid mode permits an easy and flexible design of the WTOLED structure. The on-resonance condition can be easily satisfied, and under such a condition the two TPP-MC hybrid modes could exhibit comparable light outcoupling efficiency. Moreover, the simultaneous outcoupling enhancement in the blue and orange emission regions results in similar EL spectrum and CIE coordinates to those of the ITO-based device, but a higher EL efficiency. Therefore, the proposed mode hybridization strategy can be regarded as a simple and effective light outcoupling strategy for WTOLEDs towards maintained high quality of white color, improved viewing characteristics, and EL efficiency.

4. METHODS

A. Device Fabrication and Evaluation

Glass substrates were prepared in advance and placed into a thermal evaporation chamber. For the fabrication of the device
TPC-MC, an Ag anode (80 nm), a MoO$_3$ anodic modification layer (4 nm), a hole-transporting layer of N,N’-diphenyl-N,N’-bis(1,1’-biphenyl)-4,4’-diamine (NPB, 35 nm), an emitting layer of 2 wt% bis-(7,8-benzoquinolinate)iridium(III) (N,N’-dipropyl-benzamidine) ((bpy)$_2$Ir(dpbpa)) doped into bis-(2-(hydroxyphenyl)-pyridine)beryllium (Bepp$_2$, 20 nm), an electron-transporting layer of Bepp$_2$ (40 nm), a cathode of LiF (1 nm)/Al (1 nm)/Ag (20 nm), and double periods of ZnSe (24 nm)/CaF$_2$ (98 nm) were evaporated sequentially at a base pressure of 5 x 10$^{-4}$ Pa. The fabrication processes for the device R, MC, C-MC, and the TPP-MC were the same, except that ITO-coated glass substrates were used for the device R. The active area of the WOLED was 2 mm x 2 mm. The reflectivity and transmittivity spectra were measured by using a UV–Vis spectrophotometer (UV-2550, SHIMADZU). The angular EL spectra were measured by means of a fiber-optic spectrometer. The corresponding current density–voltage–luminance (J–V–L) characteristics were measured by using a Keithley 2400 programmable voltage-current source and a Photo Research PR-655 spectrophotometer. All the measurements were performed in air at room temperature.

**B. Numerical Simulations**

The transfer matrix method was applied to simulate the reflectivity and transmittivity spectra associated with the profiles of the electric-field-intensity enhancement. In the simulation, the permittivity of Ag was chosen from Ref. [28], while the refractive indices of the other organic and inorganic materials (e.g., Alq$_3$) were measured by ellipsometry experimentally. Specifically, for the simulation of the dispersion diagrams in Figs. 5(a) and 5(d), it was difficult to define the variation of the total cavity length, as the cavity consisted of multiple organic layers. Therefore, we simply assumed the cavity to be one uniform organic layer with an averaged refractive index that could best fit the experimental results. For the simulation of the spectral radiant intensity, a numerical model [22] was applied, where a classical dipole emission was modeled in the center of the emissive layer, assuming an isotropic orientation and spectrally equal energy radiation from the dipole.

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See Supplement 1 for supporting content.

**REFERENCES**