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

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This document provides supplementary information to: “Hybrid Tamm plasmon-polariton/microcavity modes for white top-emitting organic light-emitting devices,” <http://dx.doi.org/10.1364/optica.2.000579>. Experimental dispersions of the bare MC resonance and the bare TPP mode, TPP mode enhanced transmissions and light outcouplings, dependence of the mode hybridization on the structure parameters, angular EL spectra as well as an analytical model for the derivation of the splitting frequency are reported here. © 2015 Optical Society of America
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S. 1 Dispersions of the bare MC resonance and the bare TPP mode

The bare MC resonance can be supported in a MC structure as shown schematically in Fig. S1(a). The corresponding experimental reflectivity spectra in the normal direction are shown in Fig. S1(b) with the cavity length varying from 80 nm to 140 nm. Each reflection dip indicates the excitation of a MC resonance, which exhibits a red shift as the cavity length increases. Figure S1(c) illustrates the simulated TE-polarized reflectivity dispersion diagram of the MC structure. The white circles represent the experimental results extracted from Fig. S1(b), in good agreement with the simulations. From the viewpoint of field distributions, the excitation of the MC resonance can be characterized by an enhancement of the field intensity inside the cavity as shown in Fig. S1(d).

The bare TPP mode can be supported in a TPP structure as shown schematically in Fig. S2(a). In such metal/PC structure with the first PC layer exhibiting a higher refractive index [1], the TPP mode at the metal/PC interface can be excited. The experimental reflection dips in Fig. S2(b) confirm the excitation of the TPP mode, of which the resonant wavelength only depends on the characteristics of the metal and PC instead of the cavity length. Therefore, a nearly flat dispersion can be established, as simulated in Fig. S2(c). With only double periods of the PC, the TPP mode considered here exhibits a much broader bandwidth than that of

the MC resonance. From the viewpoint of field distributions, the excitation of the TPP mode can be characterized by an enhancement of the field intensity at the metal/PC interface as shown in Fig. S2(d).

S. 2 TPP mode induced light transmissions and outcouplings

It is shown that the existence of the TPP-MC hybrid mode is a result of the couplings between the MC resonance and the TPP mode. It is well known that light can be enhanced outcoupled in OLEDs via the excitation of the MC resonance [2]. Therefore, we here demonstrate that light outcouplings in OLEDs can also be enhanced via the excitation of the TPP mode.

We first consider the transmission characteristics of a 20 nm-thick Ag layer as shown by structure A in Fig. S3(a). In contrast, double bilayers of ZnSe/CaF₂ are added on the Ag layer, forming a structure B, where d_1 and d_2 represent the thicknesses of ZnSe and CaF₂, respectively. The experimental transmittivity spectra of structure A and B in the normal direction are shown in Fig. S3(b). We note that the transmittivity of structure B is greater than that of structure A in a quite broadband region, which shifts to red as the bilayer thicknesses increases. This tendency indicates the excitation of the TPP mode, of which the resonant wavelength will move to red with the increase of the bilayer thicknesses. The photos of the fabricated samples are also shown in the inset of Fig. S3(b), from which the transmission enhancement can be

intuitional seen. Figure S3(c) shows the simulated transmittivity spectra, which match the experimental results very well.

Fig. S1. MC resonance in the MC structure. (a) Schematic of the Ag/Alq₃/Ag MC structure. (b) Experimental reflectivity spectra in the normal direction with various cavity lengths. (c) Simulated TE-polarized reflectivity dispersion diagram of the MC structure in the normal direction. (d) Simulated profiles of the electric-field-intensity enhancement with the cavity length of 100 nm.

Fig. S2. TPP mode in the TPP structure. (a) Schematic of the TPP structure. (b) Experimental reflectivity spectra in the normal direction with various cavity lengths. (c) Simulated TE-polarized reflectivity dispersion diagram of the TPP structure in the normal direction. (d) Simulated profiles of the electric-field-intensity enhancement with the cavity length of 100 nm.

We then introduce the TPP mode to OLEDs for light outcouplings. Figure S3(d) shows the schematic of an ITO based

WOLED named device A and another one deposited with double periods of PC structure named device B. The emissions in these two devices can be outcoupled from both the bottom substrate and top thin metallic cathode side. In spite of this, we only pay attention to the top side in order to study the outcoupling enhancement via the TPP mode. Figure S3(e) shows the EL spectra for the device A and B from the top thin metallic cathode side in the normal direction with the same current density ($\sim 350 \text{ mA cm}^{-2}$). It is noted that good qualities of the white color can be realized in the device A with the CIE coordinate of (0.37, 0.35). For device B, the light outcouplings from the top metallic cathode side can be considerably enhanced in a specific wavelength region, which depends on the PC thicknesses. Such light outcoupling enhancement should be attributed to the excitation of the TPP mode. Although the EL intensity can be enhanced via the TPP mode, the good quality of the white color is deteriorated due to the relatively narrow bandwidth of the TPP mode. For instance, the CIE coordinate even shifts to (0.49, 0.44) for the device B with the ZnSe/CaF₂ thicknesses of 30 nm/119 nm. This situation is the same as that for WOLEDs with the excitation of the single MC resonance. Therefore, in order to enhance the light outcouplings while at the same time maintain the good quality of the white color, we introduce the dual TPP-MC hybrid modes for WOLEDs in the main text.

Fig. S3. TPP mode enhanced light transmissions and outcouplings. (a) Schematic of structure A and B. (b) Experimental and (c) Simulated transmittivity spectra in the normal direction of the two structures. (d) Schematic of an ITO based WOLED named device A and another one deposited with double periods of PC structure named device B. (e) EL spectra for the device A and B from the top thin metallic cathode side in the normal direction with the same current density.

S. 3 Dependence of the mode hybridization on structure parameters

In this section, we theoretically investigate the dependence of the characteristics of the mode hybridization on the structure parameters. Here, we mainly concern the splitting wavelength, that is, the difference of the resonant wavelength between the two hybrid modes. Note that the splitting wavelength here is different from the detuning wavelength in the main text, which is defined as the wavelength difference between the two bare modes. The studied structure parameters include the cavity length, top metal thickness, and optical parameters of the PC including the period number, thickness and refractive index.

In Fig. S4, we study the dependence of the mode hybridization on the period number of the PC structure. We note that with only one period of PC, the TPP mode cannot be supported in the TPP structure (Fig. S4(a)), which results in the vanishing of the mode hybridization behavior in the TPP-MC structure (Fig. S4(b)). With four periods of PC, the bandwidth of the TPP mode is significantly narrowed down associated with the emersion of a new PC mode in the TPP structure in the wavelength region of interest (Fig. S4(c)). Both of these two modes can couple with the MC resonance, resulting in a more complex dispersion diagram in the TPP-MC structure (Fig. S4(d)). However, the coupling induced resonance exhibits a relatively narrow bandwidth. Considering the requirement of broadband light outcouplings in WOLEDs, we use double periods in this work.

Fig. S4. **Dependence of the mode hybridization on the period number of the PC structure.** (a) Simulated reflectivity dispersion diagrams in the normal direction of the TPP structure and (b) TPP-MC structure with one period of PC. (c-d) The items shown are as those in a-b but with four periods of PC. Other structure parameters are kept the same as those in Figs. 2(a) and S2(a).

Fig. S5. **Dependence of the mode hybridization on mode order of the MC resonance.** (a-c) Simulated reflectivity dispersion diagrams in the normal direction of the MC structure, TPP structure and TPP-MC structure, respectively, with the cavity length varying from 70 nm to 550 nm.

In Fig. S5, we study the dependence of the mode hybridization on mode order of the MC resonance. We give the simulated reflectivity dispersion diagrams with the cavity length varying from 70 nm to 550 nm. We note that the TPP mode exhibits a slightly oscillating but nearly flat dispersion relation with the cavity length varying (Fig. S5(b)). Besides the fundamental MC resonance, high-order MC resonances can also be supported in the MC structure (Fig. S5(a)). All of these MC resonances can couple with the TPP mode, resulting in a dispersion diagram with

multiple anti-crossings in the TPP-MC structure (Fig. S5(c)). Considering the conventional total organic layer thicknesses (~ 100 nm) in WOLEDs, the mode hybridization between the TPP mode and the first-order MC resonance are applied in this work. Moreover, the mode hybridization between the TPP mode and the high-order MC resonance may find applications in other optoelectronic devices, where the cavity is required to be several hundred nanometers thick.

Fig. S6. **Dependence of the mode hybridization on top metallic layer thickness.** (a) Simulated reflectivity spectra in the normal direction of the MC structure, TPP structure and TPP-MC structure with various top Ag layer thicknesses. (b) Extracted splitting wavelength associated with the splitting energy as a function of the top Ag layer thickness.

In Fig. S6, we study the dependence of the mode hybridization on the top metallic layer thickness. Here the splitting wavelength/energy is defined as the wavelength/energy difference between the two TPP-MC hybrid modes. For the calculation of Fig. S6(a), since the MC resonant wavelength as well as the TPP wavelength would be affected by changing the top Ag layer thickness, the cavity length and the PC thicknesses are slightly tuned in each choice of the top Ag layer thickness, in order to make both the MC resonant wavelength and TPP wavelength locate at the same wavelength (~ 500 nm) to achieve an on-resonance condition. We find that the splitting wavelength increases dramatically with the top Ag layer thickness decreasing. This tendency can be understood by an enhancement of the mode hybridization effect between the MC resonance and TPP mode. Therefore, changing the top metallic layer thickness can be considered as the most convenient way to tune the splitting wavelength associated with the bandwidth of the two resonances in the proposed mode hybridization strategy.

Fig. S7. **Dependence of the mode hybridization on the thickness and refractive index of the PC structure.** Simulated reflectivity spectra in the normal direction of the TPP-MC structure and TPP structure with various (a) thicknesses and (b) refractive index of the PC structure.

In Fig. S7, we study the dependence of the mode hybridization on the thickness and refractive index of the PC structure. The simulated reflectivity spectra in the normal direction of the TPP-

MC structure and TPP structure with various thicknesses of the PC structure are shown in Fig. S7(a), where the cavity length is fixed at ~ 100 nm with the bare MC resonance located at ~ 500 nm. In each case, the PC thicknesses are chosen optimally to tune the TPP wavelength at ~ 500 nm for realization of the on-resonance condition. We find that the splitting wavelength would not be affected much by changing the PC thicknesses, which offers us the flexibility for thickness design. Figure S7(b) shows the simulated reflectivity spectra for the same structures in Fig. S7(a) but with various refractive index of the PC structure. We note that by applying PC layers with a higher refractive index contrast, the bandwidth of the two resonances can be significantly narrowed down, which can be considered as an effective way for tuning the bandwidth.

As a brief summary of this section, we show that the splitting wavelength associated with the bandwidth of the two hybrid modes could be tuned most conveniently by changing the thickness of the top metallic layer, while they would also be affected by the PC parameters. Besides, we also give the reason for the choice of double periods of PC in the design. These results indicate that the excited hybrid mode exhibits highly tunable and controllable characteristics, which permit an easy design of the WTOLEDs structure based on the mode hybridization strategy.

S. 4 Angular EL spectra

Fig. S8. **Angular EL spectra.** (a) EL spectra of the device R and (b) device MC with the viewing angle varying from 0° to 60° .

Figure S8 shows the EL spectra of the device R and MC with the viewing angle varying from 0° to 60° . The normalized spectra are also shown in the inset of each figure. We note that the ITO based device R exhibits much stable angular EL spectra. From 0° to 60° , the corresponding CIE coordinate shifts only from (0.36, 0.37) to (0.35, 0.33). In device MC, the MC resonance with a narrow bandwidth shifts to blue with the increase of the viewing angle, resulting in a dramatical increment of the blue emission proportion. The corresponding CIE coordinate then shifts from (0.28, 0.44) to (0.23, 0.23), indicating an emission color variation from green to blue. The photographs of the operating devices can be found in the inset of Fig. 7(c) in the main text.

Fig. S9. Angle dependent intensity distributions of the device MC, TPP-MC and C-MC in the orange emission region (~ 570 nm), in comparison with the Lambertian emitter (black line).

The angle dependent intensity distributions of the device MC, TPP-MC and C-MC in the orange emission region (~ 570 nm) are shown in Fig. S9. We find the roll-off of device TPP-MC is comparable to that of device C-MC, and slower than that of device MC, indicating a better angular performance. In addition, the roll-off is faster than the Lambertian emitter (black line), which is due to the microcavity nature of the TPP-MC hybrid modes.

S. 5 Derivation of the splitting frequency

To gain a deeper physical insight into the mode hybridization process, we apply a coupled oscillator model [3] to derive the splitting frequency between the two TPP-MC hybrid modes in the normal direction. The eigenfrequencies of the coupled system with two oscillators can be solved through the equation

Fig. S10. Structure model for the derivation of the splitting frequency.

$$\begin{vmatrix} \omega - \omega_{MC} & \Omega_{TPP-MC} \\ \Omega_{TPP-MC} & \omega - \omega_{TPP} \end{vmatrix} = 0 \quad (S1)$$

where ω_{MC} and ω_{TPP} are the eigenfrequencies of the MC resonance and TPP mode, respectively, while Ω_{TPP-MC} represents the coupling strength between the MC resonance and TPP mode. To start with, we introduce a structure model shown in Fig. S10, where φ_C and φ_S are the phase accumulation through the cavity and a defined spacing layer, respectively, r_{Ag} , r_{PC} and r are the amplitude reflection coefficient from the bottom thick Ag layer, PC and the top thin Ag layer, respectively, and t is the amplitude transmission coefficient from one side of the top thin Ag layer to the other. By applying the transfer matrix technique and simply setting $\varphi_S = 0$, we have

$$A \begin{pmatrix} 1 \\ r_{PC} \end{pmatrix} = \begin{pmatrix} t^2 - r^2 & r \\ -r & 1 \end{pmatrix} \begin{pmatrix} \exp(i\varphi_C) & 0 \\ 0 & \exp(-i\varphi_C) \end{pmatrix} \begin{pmatrix} r_{Ag} \\ 1 \end{pmatrix} \quad (S2)$$

where A is a constant. Eliminating A from Eq. (S2) reads

$$\left(1 - \frac{1}{rr_{Ag} \exp(2i\varphi_C)}\right) \left(1 - \frac{1}{rr_{PC}}\right) = \frac{t^2}{r^2} \quad (S3)$$

Assuming the light frequency is closer to the Bragg frequency ω_0 , but much lower than the Ag plasma frequency ω_p , the expressions for r_{PC} , r_{Ag} and r can be approximated as [1]

$$r_{PC} \approx -\exp\left(i\beta \frac{\omega - \omega_0}{\omega_0}\right) \quad (S4)$$

$$r_{Ag} \approx r \approx \exp\left[i\left(\pi + \frac{2n_1\omega}{\sqrt{\epsilon_b}\omega_p}\right)\right] \quad (S5)$$

where ϵ_b is the infinite frequency permittivity of Ag, n_1 and n_2 are the refractive indices of the PC layers ($n_1 > n_2$), and $\beta = \frac{\pi n_1}{n_1 - n_2}$. By

substituting Eqs. (S4) and (S5) into (S3), and transforming the obtained one into the form of Eq. (S1), we get

$$\Omega_{TPP-MC}^2 = -\frac{t^2}{r^2} \frac{\epsilon_b \omega_p^2}{4n_1^2} \frac{1}{\frac{\pi \sqrt{\epsilon_b} \omega_p}{(n_1 - n_2) \omega_0} + 2} \quad (S6)$$

Under the on-resonance condition ($\omega_{MC} = \omega_{TPP}$), the splitting frequency Δ between the two TPP-MC hybrid modes can be obtained as

$$\Delta = 2|\Omega_{TPP-MC}| = |t| \frac{\sqrt{\epsilon_b} \omega_p}{n_1 \sqrt{\frac{\pi \sqrt{\epsilon_b} \omega_p}{(n_1 - n_2) \omega_0} + 2}} \quad (S7)$$

From Eq. (S7), the results in the Figs. S4-S7 can be explained more straightforwardly. More specifically, the splitting frequency can easily be tuned by changing the top metallic layer thickness, which directly affects the value of $|t|$. The optical parameters of the PC, especially the refractive index, can also act as a factor to tune the splitting frequency.

S. 6 Fitting the hybrid modes with Hopfield model

In this section, we fit the TPP-MC hybrid modes in Fig. 2(c) using a conventional two-level oscillator model-Hopfield model [4]. In this model, the energy for the TPP-MC hybrid mode E_{TPP-MC} can be calculated by

Fig. S11 (a) Fitting the hybrid modes with the Hopfield model and (b) the corresponding Hopfield coefficients.

$$E_{TPP-MC} = \frac{1}{2} \left[E_{MC} + E_{TPP} \pm \sqrt{4g_0^2 + (E_{MC} - E_{TPP})^2} \right] \quad (S8)$$

where E_{MC} and E_{TPP} denote the energy of the bare MC resonance and the bare TPP mode, respectively, and can be obtained from the transfer matrix simulation (see the dashed lines in Fig. 2(c)). The term g_0 represents the interaction strength between the two bare modes, and is considered here as a fitting term. We then do a fitting, aiming to find a g_0 , with which the calculated E_{TPP-MC} could best match with those from the transfer matrix simulation (see the dispersion in Fig. 2(c)). The fitting results are shown in Fig. S11(a) by the dashed lines, and in this fitting we obtain a Rabi-splitting energy ($= 2g_0$) of ~ 0.53 eV. Then the Hopfield coefficients (defined as $|H_{MC}|^2$ and $|H_{TPP}|^2$) for the two modes can be calculated by,

$$\begin{aligned} |H_{MC}|^2 &= \frac{1}{2} \left[1 - \frac{(E_{MC} - E_{TPP})}{\sqrt{(E_{MC} - E_{TPP})^2 + 4g_0^2}} \right] \\ |H_{TPP}|^2 &= \frac{1}{2} \left[1 + \frac{(E_{MC} - E_{TPP})}{\sqrt{(E_{MC} - E_{TPP})^2 + 4g_0^2}} \right] \end{aligned} \quad (S9)$$

and we also have $|H_{MC}|^2 + |H_{TPP}|^2 = 1$. The corresponding results are shown in Fig. S11(b), and we see that under the on-resonance condition, both hybrid modes have a Hopfield coefficient of 0.5, corresponding to a mode half MC-like and half TPP-like. Then if we increase the cavity length, the hybrid mode with longer resonant wavelength becomes more MC-like, and vice versa for the other hybrid mode. All these results again support the conclusion in the main text.

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