Supporting Information

Title A nanoporous polymer film as a diffuser as well as a light extraction component for top emitting organic light emitting diodes with a strong microcavity structure

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A TEOLED which has a planar microcavity structure forms a Fabry-P$\acute{e}$rot resonator because the anode and the cathode are parallel mirror. The general equation for satisfying such Fabry-P$\acute{e}$rot resonator could be expressed as follows:

$$-\varphi_t(\lambda) - \varphi_b(\lambda) + \frac{4\pi}{\lambda}\sum_i n_i(\lambda) d_i = 2m\pi \quad \text{(S1)}$$

where λ is the resonant wavelength or the peak wavelength of emission, $\varphi_t(\theta_t,\lambda)$ and $\varphi_b(\theta_b,\lambda)$ are phase changes during reflection at the top and the bottom mirrors which is affected by the incident angle (θ_t and θ_b) and wavelength (λ) of the emission, respectively. We could ignore the incident angles (θ_t and θ_b), if we consider only the light travelling through a normal direction. Meanwhile, $n_i(\lambda)$ and d_i are the refractive index and thickness of the i-th layers in between two mirrors, respectively. In addition, m is the mode number.

When we fabricate microcavity structure, two types of interferences (e.g. a multiple-beam

interference and a two-beam interference) should be considered and they are significant factors to affect the extracted light intensity. A multiple-beam interference (Fabry-P$\acute{e}$rot interference) normally occurs between two electrodes and is caused by the infinite reflection between a semi-transparent electrode (top) and a reflective electrode (bottom). Meanwhile, two-beam interference occurs between directly emitted light from the emitting dipole and the reflected light at the bottom mirror or reflective electrode. It is significantly affected by the distance (z_0) between the displacement of the emitting dipole and that of the bottom electrode. From the consideration of all of those interferences, the theoretical spectral intensity $I_{ext}(\theta,\lambda,z_0)$ from the device with microcavity in free space could be represented as follows [33, 35, 42, 44]:

$$I_{ext}(\theta,\lambda,z_0) = \frac{T_t}{1 + R_t R_b - 2\sqrt{R_t R_b}cos(\Delta\phi_{FP})} \times (1 + R_b + 2\sqrt{R_b}\cos(\Delta\phi_{TI})) \times I_{int}(\theta,\lambda) \quad \text{(S2)}$$

$$\Delta\phi_{FP} = -\varphi_t - \varphi_b + \sum_i \frac{4\pi n_i d_i cos\theta_{org,i}}{\lambda} \quad \text{(S3)}$$

$$\Delta\phi_{TI} = -\varphi_b + \frac{4\pi n_{org,EML} z_0 cos\theta_{org,EML}}{\lambda} \quad \text{(S4)}$$

where $\Delta\phi_{FP}$ and $\Delta\phi_{TI}$ are the phase terms for the Fabry-P$\acute{e}$rot and two-beam interference, respectively. R_b and R_t refer to the reflectivity at the anode-organic and cathode-organic

interfaces, respectively. φ_b and φ_t are the phase changes upon reflections at the anode-organic interface and cathode-organic interface, respectively. T_t is the transmittance of the top electrode and $\theta_{org,i}$ is the internal observation angle from the surface normal of the microcavity inside the i-th organic layer. In the same fashion, $\theta_{org,EML}$ is the angle of the light propagation in the emitting layer. n_i and d_i are the refractive index and thickness of the i-th organic layer, respectively. Especially, those terms could be denoted as a total optical thickness $L = \sum_i n_i d_i$ between two metallic electrodes (e.g. the anode and cathode). $I_{int}(\theta,\lambda)$ is the intensity of initial emission spectrum in the free space at θ, λ. Especially, the term $\frac{T_t}{1 + R_t R_b - 2\sqrt{R_t R_b}cos(\Delta\phi_{FP})}$ corresponds Fabry-P$\acute{e}$rot interference; meanwhile, the term $1 + R_b + 2\sqrt{R_b}\cos(\Delta\phi_{TI})$ describes the two-beam interference as shown in **Eq. S2 (Equation S2)**.

However, **Eq. (2) – (4)** describes about TEOLEDs having organic layers sandwiched between two electrodes. Thus, we should modify such relationship because we additionally used a capping layer (CPL, NPB) as well as a passivation layer formed by ALD (Al_2O_3) above the cathode. Thus, we additionally consider phase change at the interface of Ag / CPL (NPB), CPL / Al_2O_3 layer, and Al_2O_3 layer / air. Those phase change upon reflection or refraction when the light travel through multiple interfaces should be in-phase or out-of-phase to enhance the light power which can be escaped from the passivation layer. To maximize EQE, the light transmitted from inside layers to the outside layers should interfere as a constructive interference while the light reflected at the interfaces aforementioned should interfere as a destructive interference. The elucidation of CPL effect was reported by J. W. Huh *et al*. [38, 39]. Meanwhile, we also

introduce the resonant emission enhancement factor (G_{cav}) at a peak wavelength λ for consideration of Purcell effect [25-28, 35, 36] term as follows:

$$G_{cav}(\lambda) = \frac{\xi I_{ext}(\lambda)}{2 I_{int}(\lambda)} \times \frac{\tau_{cav}}{\tau} = \frac{\xi}{2} \frac{T_t\left[(1+\sqrt{R_b})^2 - 4\sqrt{R_b}\, cos^2\left(\frac{\Delta\phi_{TI}}{2}\right)\right]}{\left(1-\sqrt{R_t R_b}\right)^2 + 4\sqrt{R_t R_b}\, sin^2\left(\frac{\Delta\phi_{FP}}{2}\right)} \frac{\tau_{cav}}{\tau} \quad \text{(S5)}$$

where τ_{cav} and τ are the exciton lifetime in the microcavity and in an infinite medium (e.g. the free space), respectively as shown in **Eq. S5**. The antinode enhancement factor ξ has a value of 2 when the exciton recombination zone is placed at the antinode and ξ has a value of 0 if that active region is located at optical node of the standing wave. Then the emission spectrum can be significantly suppressed because the value of G_{cav} is theoretically 0. In other words, G_{cav} could only be improved at resonant wavelength for its microcavity. Thus, in this study, we could deal with this factor as a $\xi = 2$ for applying the value of G_{cav}.

In conclusion, the spectral emission shift with viewing angles from the strong microcavity devices could be explained as follows:

$$-\varphi_t(\theta_t,\lambda) - \varphi_b(\theta_b,\lambda) + \frac{4\pi}{\lambda}\sum_i n_i(\lambda) d_i = 2m\pi \quad \text{(S6)}$$

$$\sum_i \frac{4\pi\, n_i(\lambda) d_i cos\theta_i}{\lambda} = 2m\pi + \varphi_t(\theta_t,\lambda) + \varphi_b(\theta_b,\lambda) \quad \text{(S7)}$$

Eq. S6 is the general equation modified from **Eq. S1** and we could obtain **Eq. S7** for oblique incidence of light against each surface normal of electrode at peak wavelength or resonant wavelength (λ) in Fabry-P$\acute{e}$rot resonator.

To consider the peak wavelength shift ($\Delta\lambda$), we can convert λ into $\lambda + \Delta\lambda$ in **Eq. S7**. Then, we could express the resonant wavelength for solutions as **Eq. S8.**

$$\sum_i \frac{4\pi n_i(\lambda+\Delta\lambda)d_i cos\theta_i}{\lambda+\Delta\lambda} = 2m\pi + \varphi_t(\theta_t,\lambda+\Delta\lambda) + \varphi_b(\theta_b,\lambda+\Delta\lambda) = \Delta\Phi \quad \text{(S8)}$$

If we combine **Eq. S7** and **S8**, we could get **Eq. S9** which explains the peak emission wavelength shift $\Delta\lambda$;

$$\Delta\lambda = \frac{1}{\Delta\Phi}\sum_i 4\pi n_i(\lambda+\Delta\lambda)d_i cos\theta_i - \frac{1}{\Delta\Phi}[\sum_i 4\pi n_i(\lambda)d_i + \lambda(\Delta\phi_t+\Delta\phi_b)] = \frac{1}{\Delta\Phi}[\sum_i 4\pi d_i\{n_i(\lambda+\Delta\lambda)cos\theta_i - n_i(\lambda)\} - \lambda(\Delta\phi_t+\Delta\phi_b)] \quad \text{(S9)}$$

We could estimate that the extent of the peak wavelength shift upon variation of viewing angle could be inversely proportional to the mode number m (see the relationship between $\Delta\lambda$ and $\frac{1}{\Delta\Phi}$) as shown in **Eq. S9**. However, it's very difficult to reduce the $\Delta\lambda$ although m value increases because meaningful shift of λ ($\Delta\lambda$) by increasing m is only possible when the organic layer is fairly thick. Meanwhile, d_i is directly proportional to the $\Delta\lambda$ from **Eq. S9**. According to A. B. *Djurišić et al.* [40, 41], the increase of mode number m could not seriously affect peak

wavelength shift on light emission spectrum (e.g. the difference of $\Delta\lambda$ is only less than 1 nm between each neighboring mode number m; m=1, 2 or m=2, 3) and m is in the inversely proportional relation with the maximum out-coupling efficiency. Thus, the 1st order or 2nd order will be generally used to obtain optimized microcavity conditions. For example, those mode number conditions (e.g. m=1 or m=2) could be realized by using thick HTL or thin ETL. In fact, the device efficiency could be similar if we can control the particle issues or leakage current.

The spectral emission shift $\Delta\lambda_{\theta_V}$ of the peak emission wavelength with the viewing angle could be approximately estimated as follows [36, 40, 41]

$$\Delta\lambda_{\theta_V} \sim \sum_{\theta_V}\left(\sum_i \frac{4\pi d_i}{\lambda} n_i[cos\theta_i - 1] + \Delta\phi_t + \Delta\phi_b\right) \quad \text{(S10)}$$

where θ_V is the viewing angle.

$$\Delta\phi_t = \varphi_t(\theta_t, \lambda + \Delta\lambda) - \varphi_t(0, \lambda) \quad \text{(S11)}$$

$$\Delta\phi_b = \varphi_b(\theta_b, \lambda + \Delta\lambda) - \varphi_b(0, \lambda) \quad \text{(S12)}$$

According to **Eq. S10**, the spectral emission shift $\Delta\lambda_{\theta_V}$ with the viewing angle will be effectively suppressed by reducing phase changes ($\Delta\phi_t$, $\Delta\phi_b$) which are significantly dependent on reflection at each contact for all angles (θ_t, θ_b).