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Organic light-emitting diodes with 30% external quantum efficiency based on a horizontally oriented emitter

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((Optional Dedication))

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Keywords: Organic light-emitting diodes, Phosphorescent emitter, Emitter orientation, Optical simulation, Exciplex forming host, Ultra-high efficiency

High efficiency phosphorescent organic light emitting diodes (OLEDs) doped with $Ir(ppy)_2(acac)$ [bis(2-phenylpyridine)iridium(III)-acetylacetonate] in an exciplex forming co-host have been optically analyzed. This emitter has a preferred orientation with the horizontal to vertical dipole ratio of 0.77:0.23 as compared to 0.67:0.33 in the isotropic case. Theoretical analysis based on the orientation factor (Θ, the ratio of the horizontal dipoles to total dipoles) and the photoluminescence quantum yield (q_{PL}) of the emitter predicts that the maximum external quantum efficiency (EQE) of the OLEDs with this emitter is about 30% which matches very well with the experimental data, indicating that the electrical loss of the OLEDs is negligible and the device structure can be utilized as a platform to demonstrate the validity of optical modeling. Based on the results, we claim that the maximum EQE achievable with a certain emitting dye in a host can be predicted by just measuring q_{PL} and Θ in a neat film on

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glass without fabricating devices and offer a universal plot of the maximum EQE as functions of q_{PL} and Θ.

1. Introduction

Organic light emitting diodes (OLEDs) with horizontally aligned emitters have the potential to achieve high external quantum efficiency (EQE) without outcoupling enhancement layers. An emitter with a horizontal transition dipole moment results in much higher outcoupling efficiency than the vertically aligned dipole as demonstrated in polymers and vacuum evaporated organic molecules.[1-8] Recently, not only fluorescent molecules but also some phosphorescent dyes are reported to have preferred horizontal dipoles where high EQE over 30% is expected.[9-13] Unfortunately no experimental data have yet demonstrated the potential of horizontally oriented phosphorescent dyes to get high efficiencies over 30% to our best knowledge because of the lack of a device structure to get perfect electron and hole balance. Moreover, the most commonly used green phosphorescent dye, $Ir(ppy)_3$, doped in CBP was reported to have isotropic distribution of emitting dipoles.[13] Thus, it is important to develop or find a host/guest system that has preferentially horizontal dipoles as well as a device structure with nearly 100% charge balance, i.e. without electrical loss, to demonstrate the validity of the prediction of the optical model.

Recently our group reported a very high efficiency green phosphorescent OLED with the EQE of 29.1% and extremely low roll-off of efficiency at high current density.[14] In this device $Ir(ppy)_2(acac)$ [bis(2-phenylpyridine)iridium(III)-acetylacetonate] was doped in the exciplex forming co-host system of TCTA [4, 4′, 4 ″-Tri (N-carbazolyl) triphenylamine] and B3PYMPM [bis-4,6-(3,5-di-3-pyridylphenyl)-2-methylpyrimi-dine]. The high efficiency of the OLED with low roll-off of efficiency indicates that the electrical loss (including charge balance and exciton-polaron quenching) seems to be negligible in the device so that it can be used as a platform to analyze the outcoupling efficiency and the factors influencing the EQE

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and eventually validate the prediction of an optical model.

In this paper, we demonstrate by optical analysis that the phosphorescent emitter $Ir(ppy)_2(acac)$ in the device has a preferred non-isotropic orientation with horizontal to vertical dipole ratio of 0.77:0.23 ($\Theta = 0.77$), and the device has the maximum EQE of 30% based on the photoluminescence (PL) quantum yield (q_{PL}) of 0.94 in the thin film and 100% charge balance. The theoretical prediction agrees very well with the experimental data, validating the optical model used for the prediction of the EQE. Based on the validation, we offer a universal plot of maximum efficiency achievable with different values of q_{PL} and Θ in a dye doped EML without fabricating devices. The optical analysis indicates that OLEDs with EQE higher than 40% can be realized without any extra light extraction layers, if phosphorescent dyes with q_{PL} and Θ over 95% are used.

2. Simulation of external quantum efficiency

The EQE of an OLED has been expressed by the following equation: [15]

$$\eta_{EQE} = \gamma \times \eta_{S/T} \times q_{PL} \times \eta_{out} \quad (1)$$

where γ is the charge balance factor, $\eta_{S/T}$ is the singlet-triplet factor ($\eta_{S/T}$ =0.25 for fluorescent, $\eta_{S/T}$ =1 for phosphorescent emitter), q_{PL} is the PL quantum yield, and η_{out} is the outcoupling efficiency of the emitted light. However, the quantum efficiency of an emitter in a micro-cavity structure is influenced by the orientation of the emitter, the local electric field at the dipole position and the proximity to a metal layer. The η_{out} is influenced not only by the device structure but also by the orientation of emitting dipoles so that the EQE must be modified as follows:[13, 16]

$$\eta_{EQE} = \gamma \times \eta_{S/T} \times q_{eff}(q_{PL}, \Theta, \Gamma) \times \eta_{out}(\Theta, \Gamma) \quad (2)$$

where q_{eff} is the effective quantum yield that describes the probability of radiative exciton

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decay in an optical cavity structure (i.e. the Purcell effect [17]), which is generally depending on q_{PL}, Θ, and the geometric factor of the device (Γ) including the device structure and the location of the emission zone in the device. In a similar manner, η_{out} is influenced by Θ and Γ. The γ value is often assumed to be unity in state-of-the-art OLEDs, however, this value is not a constant but a fitting parameter in this study. With separately measured values of q_{PL} and Θ, and using known information of the device structure, we can now calculate q_{eff} and η_{out} from a classical dipole model and then fit to the experimentally obtained EQE to extract γ. Therefore, we can exactly analyze the effect of the emitter orientation, the electrical loss as well as the device structure on EQE by fitting theoretically predicted EQE to experimental data. Details of the method for calculating η_{out} were described elsewhere. [18-21] We assumed that the emission zone is located in the middle of the EML in OLEDs, which is a reasonable assumption for the device having a uniformly distributed profile of the emission zone such as the co-host system used here.[22]

A schematic diagram of the device structure and the chemical structure of the materials used in this study are shown in **Fig. 1a**. The refractive indices of the organic layers, except for B3PYMPM, were measured by a spectroscopic ellipsometer.[23] The refractive indices of B3PYMPM, glass substrate, ITO and metal are taken from literature.[7, 24, 25] The refractive indices of the undoped co-host layer (TCTA:B3PYMPM, 1:1 molar ratio) measured by a spectroscopic ellipsometer are displayed in **Fig. 1b**. We ignored the modification of the optical constant by the doping of the phosphorescent dye molecules due to the low doping concentration (< 10%). The optical constants of the co-host system show strong anisotropy with an ordinary refractive index (n_o) of 1.8342 and an extra-ordinary refractive index (n_e) of 1.6776 at the wavelength of 520 nm, respectively. The strong optical anisotropy indicates that the host materials are horizontally oriented.

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The dipole orientation of the emitting dyes in the host matrix was determined from the analysis of the angle dependent PL spectrum of the EML using the classical dipole model.[18, 19] The sample was prepared by co-depositing B3PYMPM, TCTA and $Ir(ppy)_2(acac)$ with the same molar ratio of the host materials and 8 wt% of the dopant on a pre-cleaned 1 mm-thick fused silica substrate.The thickness of the film was 30 nm, which corresponds to the thickness of the EML in the OLED. A He-Cd laser (325 nm) was used as the excitation source of the sample and the *p*-polarized emitted light at 520 nm, corresponding to the peak wavelength of the PL spectrum of the phosphorescent dye, was detected. Details of the experimental method and the analysis are described in literature. [8] In the simulation of the angle dependent PL spectrum, the optical anisotropy was accommodated in the model by using the effective refractive index as a function of the propagation direction of the *p*-polarized emitted light. The effective refractive index (n_{eff}) is defined as a function of the emission angle (θ) in the organic layer as follows:

$$\frac{1}{n_{eff}^2} = \frac{\cos^2\theta}{n_o^2} + \frac{\sin^2\theta}{n_e^2} \qquad (3)$$

The effective refractive index of the organic layer for the *p*-polarized light changes from n_o to n_e as the angle of emission varies from 0° to 90°. The experimental data and the simulation results of the angle dependent PL spectrum of the 30 nm thick $Ir(ppy)_2(acac)$ doped TCTA:B3PYMPM film are shown in **Fig. 1c**. The experimental data are well fitted with the horizontal to vertical dipole ratio of 0.77:0.23, indicating that the emitter in the EML has more horizontally oriented dipoles than vertically oriented ones compared to the isotropic orientation and the result is consistent with the previous report. [12] It is interesting to note that the Θ value is the same as that of $Ir(MDQ)_2(acac)$. [9-11]

3. Results and discussion

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The device under investigation has a simple structure of glass/ITO (70 nm)/TAPC (x nm)/TCTA (10 nm)/TCTA:B3PYMPM:Ir(ppy)$_2$(acac) (1:1 of molar ratio and 8 wt.%) (30 nm)/B3PYMPM (40 nm)/Al (100 nm), where TAPC represents 1,1-bis(di-4-tolylaminophenyl)cyclohexane. The thickness of the TAPC layer was set as a parameter and varied from 40 to 100 nm.

The current density−voltage−luminance ($J-V-L$) characteristics of the devices with different thicknesses of the TAPC layer are shown in **Fig. 2a**. The $J-V$ characteristics of all the devices are not significantly different from each other, especially in the low current region, due to a high hole mobility of TAPC (~10^{-2} cm^2/Vs).[26] Thus, the difference in the efficiency of the OLEDs with different thicknesses of the TAPC layer originates mostly from an optical effect. The turn-on voltages of all the devices are identical at 2.4 V and the driving voltages of the devices are less than 3.9 V and 6.3 V for 1000 and 10000 cd/m^2, respectively. Maximum current efficiency and power efficiency are 106 cd/A (60 nm-thick TAPC) and 127.3 lm/W (80 nm-thick TAPC), respectively (not shown). The EQE of the devices corrected with their emission patterns (**inset of Fig. 2b**) are displayed in **Fig. 2b**. The maximum EQE of 30.2% was obtained with the 80 nm-thick TAPC layer. The EQE value is the highest one in bottom emitting green phosphorescent OLEDs based on an ITO electrode to our best knowledge. There is a paper reporting the EQE of 29.2% and 93 cd/A using the same emitter. [27] Unfortunately, the authors of that work did not take into account emitter orientation, which is obviously the key to achieve the high EQE. The measured EQE values of the devices are plotted in **Fig. 2c** against the thickness of the TAPC layer.

Optical simulation of the EQE of the devices was performed using the experimentally obtained values of q_{PL}=0.94 and Θ=0.77±0.02. The PL quantum yield was measured using an integrating sphere [20, 28] and the sample consisted of the 50 nm thick emitting layer (TCTA:B3PYMPM:Ir(ppy)$_2$(acac), 1:1 of molar ratio and 8 wt.%) on a quartz substrate. The

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experimental results are very well described by the simulated results as shown in **Fig. 2c** under the condition of γ=1, indicating that the electrical loss is indeed negligible. In other words, the injected electrons and holes into the EML of the OLEDs efficiently recombine to form excitons. The excellent match between the experimental and the simulation results clearly indicates that the optical simulation describes the maximum achievable EQE under the known values of q_{PL} and Θ when the device structure is optimized electrically and optically. This fact implies that we can predict the maximum EQE achievable with a certain emitting dye in a host by just measuring q_{PL} and Θ on a neat film of the EML on glass without the need for fabrication of full OLED devices.

Based on this idea, we extend the simulation to calculate the maximum achievable EQEs as functions of q_{PL} and Θ. The corresponding simulation results are shown in **Fig. 3** as a contour plot. We used the structure shown in Fig.1a with the optimized thickness of the TAPC layer (75 nm) for the simulation. The maximum efficiency increases as q_{PL} and Θ approach 1 as expected. Surprisingly, a maximum EQE of 46% can be achieved in normal ITO based bottom emitting OLEDs without any extra outcoupling layers using a phosphorescent dye with q_{PL}=1 and Θ = 1. Practically over 40% EQE is possible with q_{PL} = 0.95 and Θ = 0.95. In contrast, the maximum EQE of the OLED with isotropically oriented phosphorescent dyes is much lower (~ 25%).

4. Conclusions

In summary, optical analysis of the high efficiency OLED showed that the phosphorescent dye $Ir(ppy)_2(acac)$ in the EML has a preferred horizontal dipole orientation (parallel to the substrate plane) to result in a maximum EQE of 30% under the condition of negligible electrical losses in the device. The prediction matches very well with the experimental value,

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suggesting that firstly the device has almost perfect electron-hole balance and the classical dipole model used for the calculation of q_{eff} and η_{out} are valid for the analysis of the performance of OLEDs. The analysis indicates that an EQE of 40% is possible in ITO based bottom emitting OLEDs without any extra light extraction layers.

5. Experimental

Device fabrication: The OLEDs were fabricated by thermal evaporation onto cleaned glass substrates pre-coated with ITO. Prior to the deposition of the organic layers, the ITO substrates were exposed to UV-ozone flux for 10 min following degreasing in acetone and isopropyl alcohol. All the layers were grown by thermal evaporation at a base pressure of < 5×10^{-7} Torr without breaking the vacuum.

Measurement of angle dependent PL intensity: The experimental setup was composed with a motorized rotation stage, a fused silica based half cylindrical lens with a holder of a sample, a dichroic mirror to filter the excitation beam, a polarizer to classify the polarity of emitted light, fiber guided detector combined with a monochromator and a photomultiplier tube. The continuous wave He-Cd laser (325 nm, Mellisgriot Co.) was used as the excitation source of the sample and their incident angle was 45 degree. The *p*-polarized emitted light at 520 nm, corresponding to the peak wavelength of the PL spectrum of the phosphorescent dye, was detected.

Characterization of the OLEDs: The current density, the luminance, and the EL spectra were measured using a Keithley 2400 source meter and a SpectraScan PR650 (Photo Research). The angular distribution of the EL intensity was measured using the Keithley 2400 source meter, a rotation stage and an Ocean Optics S2000 fiber optic spectrometer. The EQE and the power efficiency of the OLEDs were calculated from the current density, the luminance, the EL spectra, and the angular distribution of the EL intensity.

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Measurement of PL quantum yield of the emitter: The PL efficiency of the emitter was measured using integrating sphere (Labsphere Co., 6 inches of diameter). He-Cd laser (325 nm) was used as the excitation source, which is the same as the one used in the measurement of angle dependent PL intensity. A monochromator (Acton Research Co.) attached with a photomultiplier tube (Hamamatsu Photonics K.K.) was used as the optical detector system. To avoid the degradation of the samples by laser excitation, the samples were kept in inert environment by blowing nitrogen gas into the integrating sphere.

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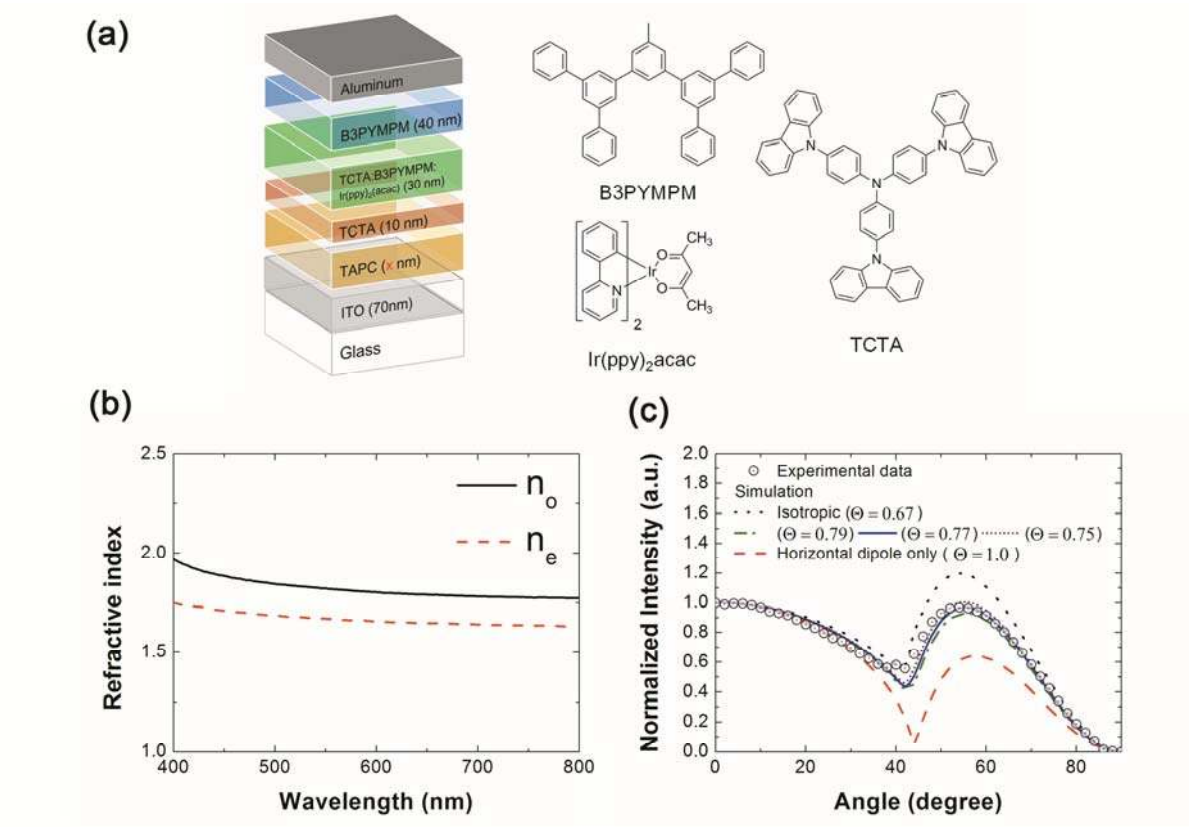

Figure 1. (a) Schematic diagram of the device and molecular structures of co-host and a phosphorescent dye used in this study. The thickness of the TAPC layer was varied from 40 to 100 nm. (b) Refractive indices of undoped co-host layer (TCTA:B3PYMPM, 1:1 of molar ratio) measured by a variable angle spectroscopic ellipsometer. (c) Experimentally obtained angle dependent PL spectrum of EML on fused silica substrate (circle) is compared with the simulated ones (lines) with different portion of the horizontal dipoles (Θ) (red dashed line for Θ=1 (fully horizontal), green dash-dot line for Θ=0.79, blue thick-solid line for Θ=0.77, purple short-dot line for Θ=0.75, and black dotted line for Θ=0.67 (isotropic)).

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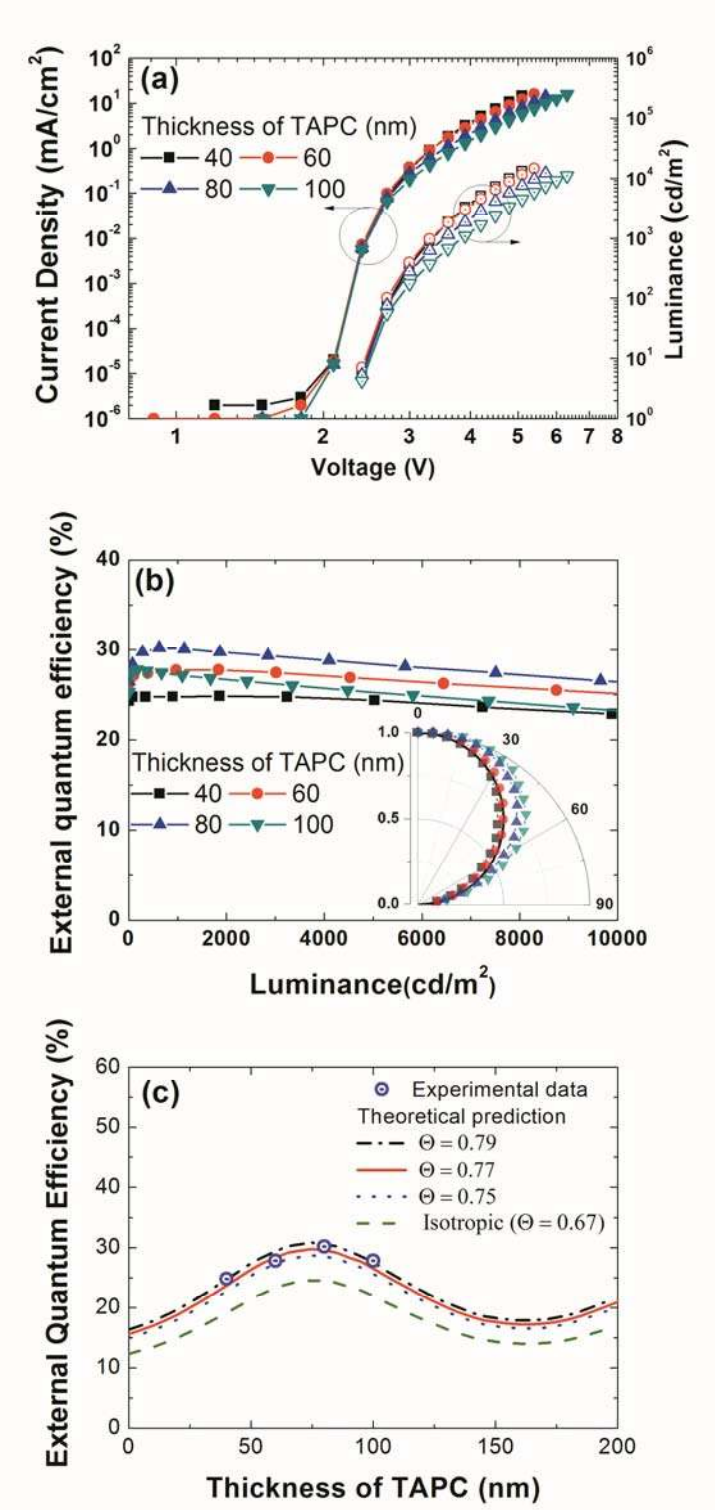

Figure 2. The experimental data of the OLEDs with different thicknesses of TAPC layer (40, 60, 80 and 100 nm, respectively): (a) Current density−voltage−luminance curves (b) EQE vs. Luminance. (Inset: angle dependent emission patterns of the OLEDs. Black solid line indicates the Lambertian distribution). (c) Experimental EQEs (circles) are compared with simulated EQEs with different orientation factors of dipoles; black dash-dot line for Θ=0.79, red solid line for Θ=0.77, green dotted line for Θ=0.75, and green dashed line for Θ=0.67 (isotropic). q_{PL} of 0.94 and γ of 1.0 were used for the simulation.

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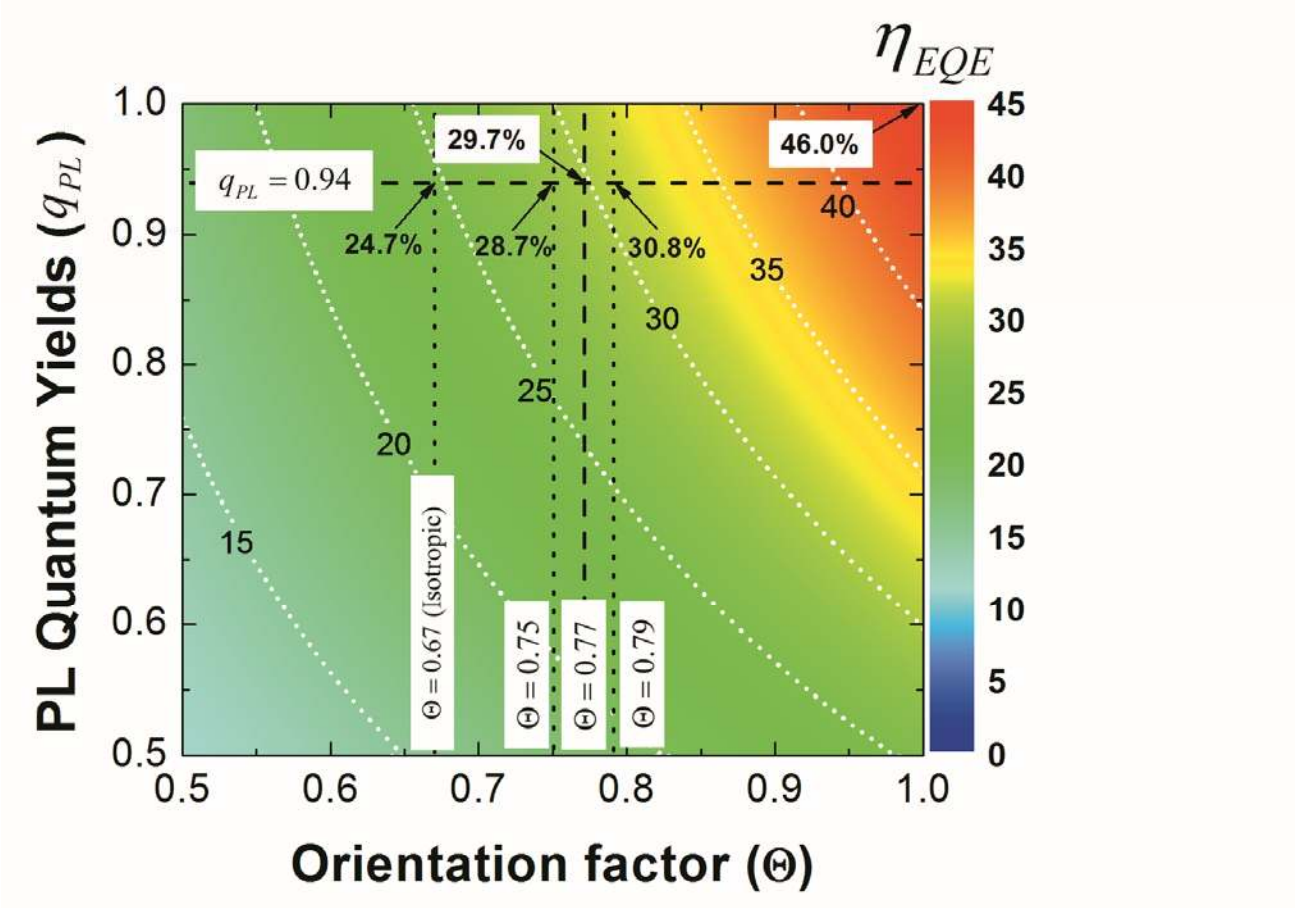

Figure 3. Contour plot of the simulation result of EQE as functions of q_{PL} and Θ. The device structure shown in Fig. 1 with TAPC thickness of 75 nm was used for the simulation. The dashed lines indicated the locus of EQE when Θ and q_{PL} are 0.77 and 0.94, respectively. In the same manner, the dotted lines indicate the EQE for Θ= 0.67 (isotropic), 0.75 and 0.79, respectively.

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Keyword: Organic light-emitting diodes, Phosphorescent emitter, Emitter orientation, Optical simulation, Ultra-high efficiency

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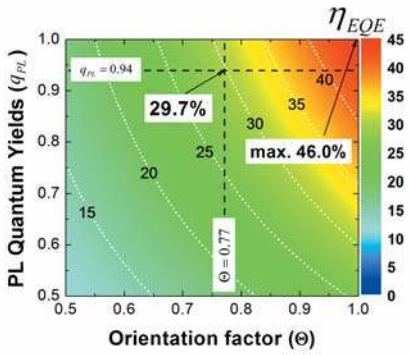
Title: **Optical analysis of organic light-emitting diodes with 30% external quantum efficiency**



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