

Optimization of organic light emitting diode structures

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In this work we present detailed analysis of the emitted radiation spectrum from tris(8-hydroxyquinoline) aluminum (Alq₃) based OLEDs as a function of: the choice of cathode, the thickness of organic layers, and the position of the hole transport layer/Alq₃ interface. The calculations fully take into account dispersion in glass substrate, indium tin oxide anode, and in the organic layers, as well as the dispersion in the metal cathode. Influence of the incoherent transparent substrate (1 mm glass substrate) is also fully accounted for. Four cathode structures have been considered: Mg/Ag, Ca/Ag, LiF/Al, and Ag. For the hole transport layer, N,N'-diphenyl-N,N'-(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD) was considered. As expected, emitted radiation is strongly dependent on the position of the emissive layer inside the cavity and its distance from the metal cathode. Although our optical model for an OLED does not explicitly include exciton quenching in vicinity of the metal cathode, designs placing emissive layer near the cathode are excluded to avoid unrealistic results. Guidelines for designing devices with optimum emission efficiency are presented. Finally, the optimized devices were fabricated and characterized and experimental and calculated emission spectra were compared.

Keywords: organic light emitting diodes, device design

1. INTRODUCTION

Organic light emitting diodes (OLEDs) are attracting increasing attention for applications in display devices due to their high brightness and wide viewing angle.^{1,2} While there has been significant progress in this area over recent years, comprehensive optical studies device simulation and design have been scarce. The simulations of the charge transport and charge distribution in organic light emitting devices have been reported.^{3,4} There have been several works on calculating the emission pattern⁵ and external coupling in OLEDs.⁶ It has been shown that the emission from an OLED can be varied through interference effects by changing the thickness of the organic layer in an OLED with a dielectric layer located below the ITO anode⁷ or by changing the thickness of a ZnSe layer on top of the metal cathode.^{8,9} For an OLED with SiO₂ layer below the ITO, it was shown that the emission spectrum from an OLED is strongly dependent on the Alq₃ thickness, so that with the increase of Alq₃ thickness the emission peak shifts from ~525 nm to ~555nm, and with further increase of the thickness even splits into two peaks, one located ~495 nm and the other ~590-600 nm.⁷ It was also shown that, by changing the thickness of ZnSe layer deposited on top of the cathode, the efficiency of the OLED can be optimized so that for optimal ZnSe thickness efficiency as high as 64 cd/A can be achieved.^{8,9}

The spectral and angular distribution of the emission from an OLED has been studied in more details in organic microcavity devices^{10,11} than in non-cavity devices. However, if we consider a typical OLED structure consisting of a glass substrate, ITO anode, hole transport layer (HTL), emitting/electron transport layer (EML/ETL), and a metal cathode, as shown in Fig. 1, it is obvious that this device can be considered as a microcavity with very weakly reflecting bottom mirror. It was shown that weak microcavity effects can be observed in such simple bilayer OLEDs.¹² If we consider the refractive indices of the organic materials used,¹³ the ITO¹⁴ anode, and the glass substrate, the reflectance from the bottom side is just a few percent, but this is still sufficient to affect the emission pattern of an OLED and result

in the dependence of the emission wavelength and intensity on the organic layer thickness and possibly the cathode choice.

Therefore, in this work we investigated the influence of the cathode choice, organic layer thickness, and the position of the emission region to the output from an OLED consisting of N,N'-diphenyl-N,N'-(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD) as a hole transport layer (HTL) and tris(8-hydroxyquinoline) aluminum (Alq₃) as emitting/electron transport layer (EML/ETL). The cathodes considered are the structures commonly used in OLEDs: Mg/Ag, Ca/Ag, LiF/Al, and Ag. The model fully takes into account the dispersion in all the layers in the structure. The organic layer thickness values chosen are realistic, and the position of the emission region close to the metal cathode have not been considered in order to avoid unrealistic predictions since the model does not take into account exciton quenching due to defect states introduced during metal deposition. The calculations have been compared with the experimental data for ITO/TPD/Alq₃/LiF/Al devices. The paper is organized as follows. In the following section, the description of the model is given. In Section 3, experimental conditions are described. The results and discussion are presented in Section 4. Finally, conclusions are drawn.

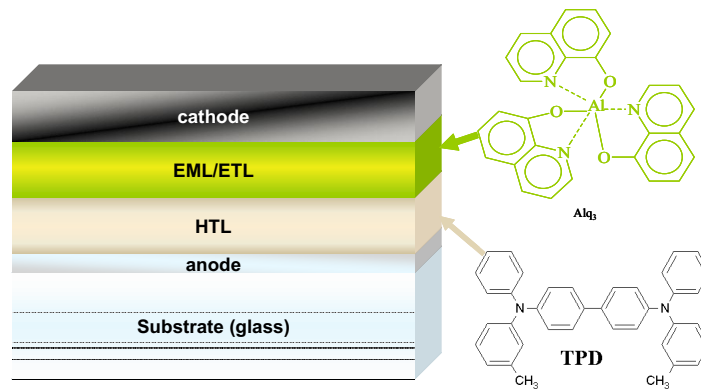


Figure 1 The typical OLED device structure.

2. DESCRIPTION OF THE MODEL

This paper investigates the effect of cathode metal and the position of the emission region on the spectral and luminous characteristics of the OLED. We analyzed conventional OLEDs fabricated on ITO glass substrates as microcavity devices with ITO/glass considered as a bottom mirror. Due to the small index offset between the ITO and the adjacent HTL the cavity effects are weak. Resonant modes of the microcavity have to satisfy the condition that the phase change during one round trip is a multiple of 2π . In other words, for normal incidence, the following equation is valid:

$$\sum_i \frac{4\pi}{\lambda} d_i n_i(\lambda) - \varphi_{top}(0, \lambda) - \varphi_{bot}(0, \lambda) = 2m\pi, \quad (1)$$

where λ is the emission wavelength, $\varphi_{top}(\lambda)$, $\varphi_{bot}(\lambda)$ are the wavelength dependent phase changes upon reflection from top and bottom mirrors, respectively, m is an integer which defines the mode number, and the summation in is performed over all the layers inside the cavity with thicknesses d_i and refractive indices $n_i(\lambda)$. The phase shift upon reflection from the mirrors is calculated using matrix method. The wavelength dependence of the index of refraction for aluminum (Al) and silver (Ag) was modeled using Brendel-Bormann's model, with model parameters given in Ref.¹⁵ The refractive index of magnesium was modeled by oscillator (Lorentz-Drude) model with parameters given in Ref.,¹⁶ while optical constants of calcium films were obtained from the recent work of Ramsdale and Greenham.¹⁷ The refractive index of Alq₃ and TPD was modeled using Cauchy equation $n(\lambda)=A+B/\lambda^2+C/\lambda^4$, where the coefficients A , B , and C were determined by fitting the data in Ref.¹⁵ The refractive index of ITO was modeled using the model given in Ref.¹⁴ As all devices investigated here are bottom emitted, the reflection from the back face of the glass substrate needs to be considered as well. Reflection coefficient was determined by applying matrix formalism with incoherent substrate correction using the

modified matrix approach of Katsidis and Siapkas.^{18,19} The refractive index of glass was described with Cauchy equation. The external emission spectrum of the OLED is given by:^{10,11}

$$I_{cav}(\lambda) = \frac{1 - R_{bot}}{i} \sum_i \left[\frac{1 + R_{top} + 2\sqrt{R_{top}} \cos\left(\frac{4\pi z_i}{\lambda} - \varphi_{top}\right)}{1 + R_{bot}R_{top} - 2\sqrt{R_{bot}R_{top}} \cos\left(\frac{4\pi L}{\lambda} - \varphi_{top} - \varphi_{bot}\right)} \right] I_{nc}(\lambda), \quad (2)$$

where $L = \sum_j n_j d_j$ is the optical thickness of the cavity, z_i is the optical distance of the emitting dipoles to the metal mirror (cathode) with reflectivity R_{top} , R_{bot} is the reflectivity of bottom mirror (ITO/glass), $I_{nc}(\lambda)$ is the free space emission of the emitting material, and the summation over i is performed assuming that the dipoles are located within 10 to 20 nm from the HTL/emitting layer interface.¹⁰ The actual thickness of the emitting layer depends on the exciton diffusion length. Therefore, it can be affected by the quality of organic layers (deposition rate, purity of materials, vacuum level, interface surface roughness), as well as the driving current and environmental factors since organic materials are very sensitive to temperature and atmosphere exposure.

3. EXPERIMENTAL DETAILS

The materials used (Alq3 from Syntech GmbH, and TPD from H. W. Sands Corp.) were purified by vacuum sublimation before fabrication. ITO substrates with nominal sheet resistance of 40 ohm/square were supplied by Varitronix Limited, Hong Kong. All the ITO substrates were cleaned before fabrication with the following cleaning procedure: the substrates were first rinsed by ethanol, acetone, toluene, then acetone, ethanol, and deionized (DI) water. After that, the substrates were cleaned in ultrasonic bath for 10 minutes in acetone, ethanol, and DI water respectively, and dried in an oven. We have performed optimal surface treatment of the cleaned ITO according to the previously reported results²⁰ by dipping the ITO into 12% aqueous solution of HCl for 15 minutes, followed by 15 minutes UV-ozone treatment.

Devices were fabricated by vacuum deposition. The pressure during evaporation was of the order 10^{-4} Pa. The evaporation rate was kept at $1\text{\AA}/\text{s}$ - $2\text{\AA}/\text{s}$. The distance from source to film was about 23 cm to ensure uniformity of film thickness, and the substrate holder was rotating. The thickness of the films was controlled by a quartz thickness monitor. After evaporation, film thickness was verified using a Dektak 3 step profiler. The electrode consisted of 0.5 nm LiF and 100 nm Al. Eight cells were fabricated on each ITO substrate in one fabrication, and fabrications were repeated several times to verify reproducibility of the obtained results. The electroluminescence (EL) was measured using luminance-current-voltage (L-I-V) characteristics were measured using Keithley 2400 sourcemeter and Oriel MS 260I monochromator with integrating sphere and CCD detector in ambient environment. Photoluminescence (PL) spectra were recorded using Perkin Elmer Instruments LS50B Luminescence Spectrometer for excitation wavelengths 350 nm.

4. RESULTS AND DISCUSSION

As we have mentioned before, the cavity effects in structures used in this study (shown in Fig. 1) are relatively weak. The substrate used is 1mm thick glass where the anode is a 50 nm thick ITO layer. We investigated four different metallic cathode structures, namely: LiF 0.5nm|Al 100nm, Mg 30 nm|Ag 70 nm, Ag 80 nm, and Ca 30 nm| Ag 70 nm. Figure 2 shows the effect of the cavity length on electroluminescent spectrum of devices with these four different top metallic mirrors.

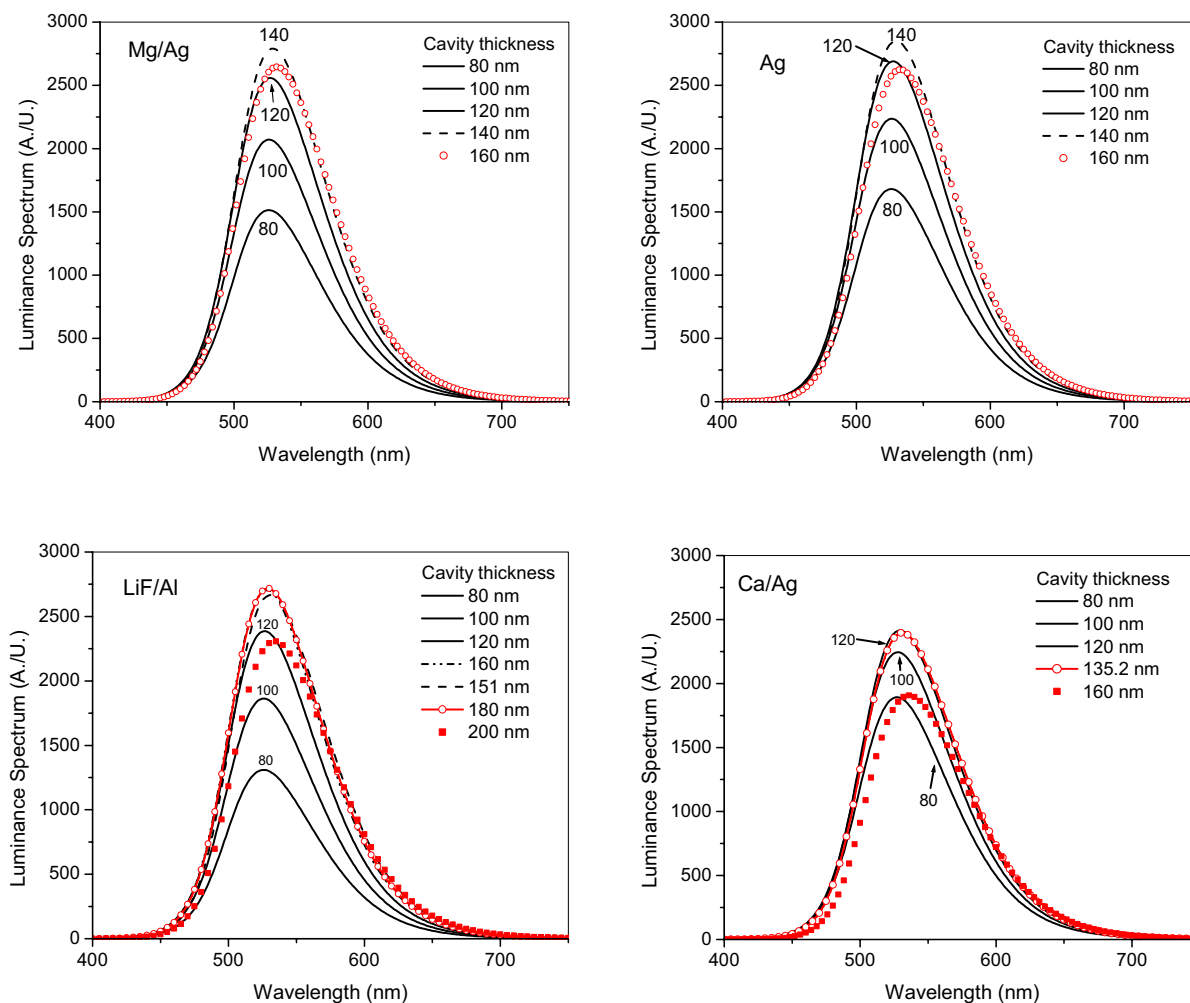
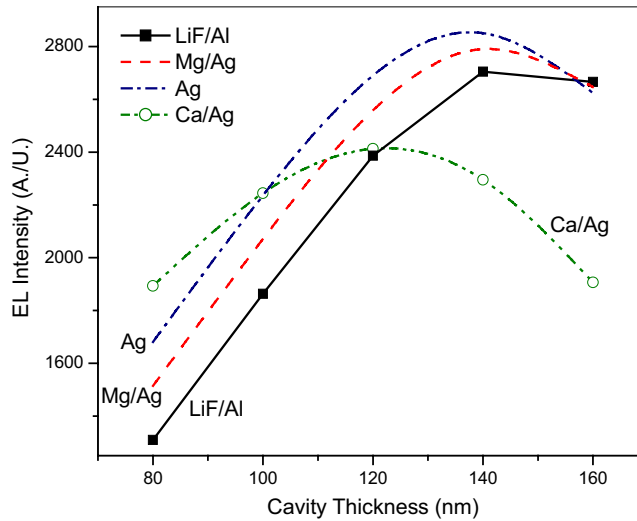


Figure 2: Effect of the cavity length on electroluminescent spectrum of devices with different top metallic mirrors.

All four structures show the similar pattern of dependence on the cavity thickness: monotonous increase in extracted EL intensity with increasing cavity thickness [solid lines in Figs. 2(a) to (d)] up to the value corresponding to the first longitudinal mode of the cavity. Beyond the resonant thickness the intensity is decreasing (lines with markers). Structures with LiF/Al, Mg/Ag and Ag mirrors have similar optical behavior due to the strong free-electron-like behaviour of these metals. The device with Ca/Ag mirror has significantly reduced EL intensity due to the lower reflectivity of calcium in comparison to other used cathodes. In all cases strong enhancement in extracted EL intensity was obtained for cavity lengths corresponding to the resonance of the first longitudinal mode of the cavity given by Eq. (1) notwithstanding the fact that the bottom mirror is virtually non-existing. In all calculations we assumed physical thicknesses of the HTL (TPD) and the ETL (Alq₃) layers equal to 0.5 of the cavity length. Figure 3(a) shows the peak EL intensity as a function of the cavity thickness. First three devices have the optimal thickness close to 140 nm, while the Ca/Ag cathode structure allows for a reduced cavity thickness. Figure 3(b) shows the magnitude of the EL peak as a function of Alq₃ layer thickness. It should be noted that our calculations deal with optical factors only. Actual EL intensity will also depend on the charge transport properties in the organic layers and the electron injection efficiency of the cathode. In this work, we mainly study the optical effects of changing the layer thickness, interface position, and the cathode material. In future work, the optical design considerations will be combined with the electronic design issues to present comprehensive guidelines for efficient OLED fabrication.



Figures 3(a): Peak EL intensity as a function of the cavity thickness for different metal cathodes

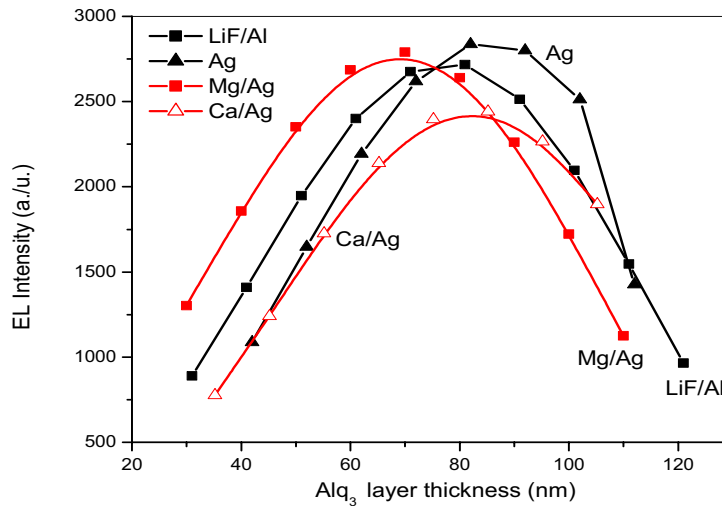


Figure 3(b) Peak EL intensity as a function of the Alq₃ layer thickness for different metal cathodes

Figure 4 shows the dependence of the extracted EL spectrum on the position of the light emitting layer. For all four structures, we used the total cavity length corresponding to the first longitudinal mode ($m = 1$); we changed the thickness of the ETL (Alq₃) to investigate the effect of the position of the light emitting layer on the EL spectrum. Thin, electrically unsuitable layers (thinner than 30 nm) have been avoided in calculations. The difference between the refractive indices of TPD and Alq₃ is less than 1% in the vicinity of 530 nm (position of the EL peak of the noncavity device). Therefore, the total physical cavity length is almost independent on the position of the TPD/Alq₃ interface. This enabled us to single out and investigate the effect of the position of the emitting layer on the EL spectrum, without altering the other parameters of the optical design of the cavity. However, one has to bear in mind that by changing the relative thicknesses of HTL and ELT (TPD and Alq₃ respectively) electron transport is significantly affected due to different carrier mobility in the two layers. The mobility of electrons in Alq₃ is about two orders of magnitude lower than the hole mobility in TPD.²¹ Therefore, changing the thickness of these two layers will significantly affect the balance between electrons and holes reaching the interface, and thus affect the emission intensity.

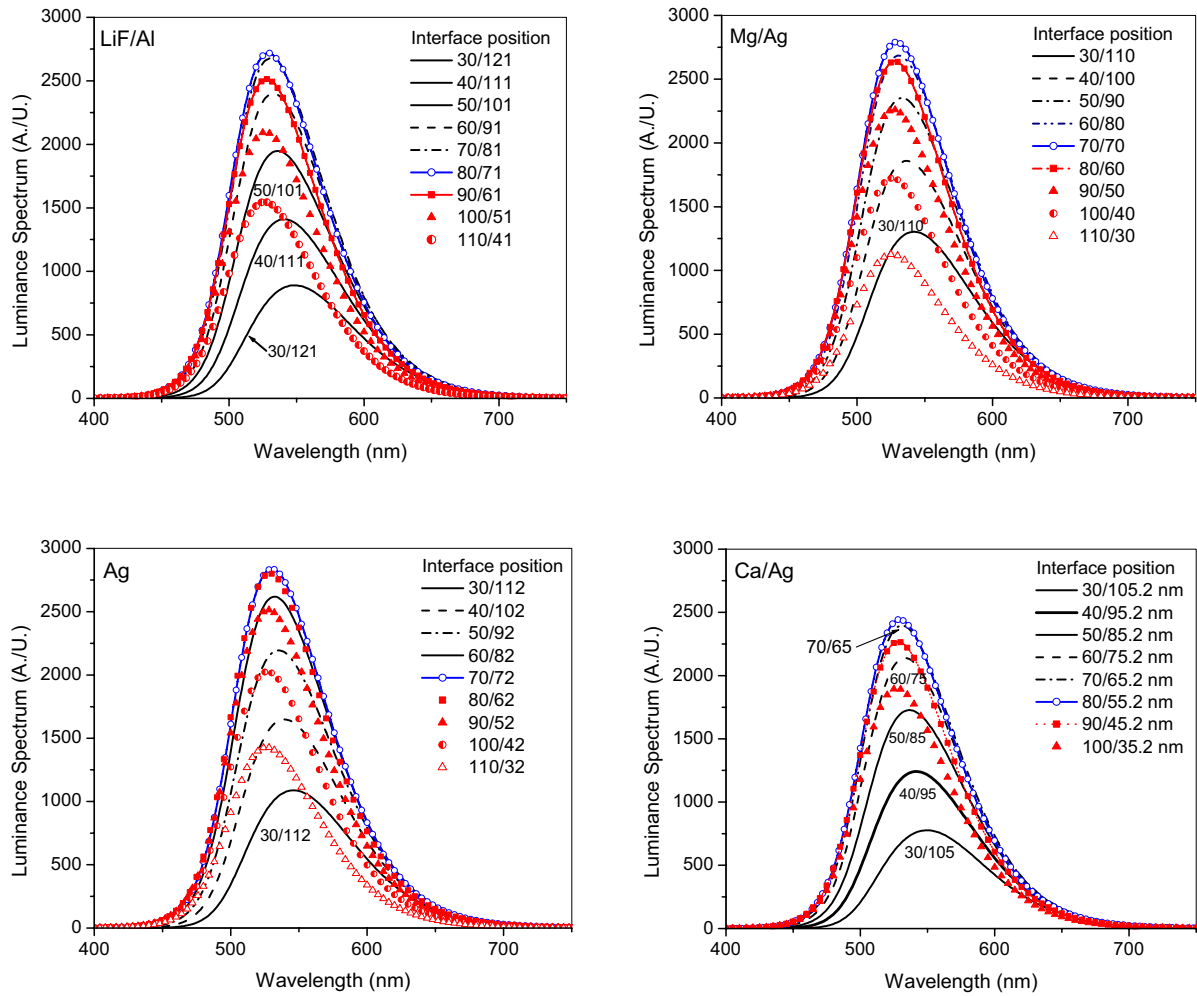


Figure 4(a) to 4(d): Dependence of the extracted EL spectrum on the position of the light emitting layer for different metal cathodes.

We will now have a closer look at the LiF/Al mirror device, since LiF/Al is the most commonly used cathode in OLEDs. The thickness of the Alq3 layer was changed from 30 to 110 nm. The total thickness of the cavity spacer was kept at 151 nm, corresponding to the wavelength of the $m = 1$ resonant mode of the cavity. Effect of the position of the TPD/Alq3 interface on the electroluminescence spectrum in an OLED with LiF/Al mirror is shown in Fig 4(a). Clearly the strongest EL enhancement is obtained when the emission layer is aligned with the position of the antinode of the resonant cavity.²² Fig 5 depicts the electric field intensity in the optimally designed cavity with the HTL/ETL interface (emission layer) located at the antinode of the electric field. The phase shift on reflection from the metal mirror plays the critical role in determining the position of the antinode of the electric field. It is well known that the optical constants of metallic films depend strongly on the evaporating conditions and evaporation rate.¹⁵ Optical constants of aluminum in particular may vary significantly due to the variable surface roughness of the film and due to the rapid volume oxidation of the aluminum film, unless the evaporation was performed in ultrahigh vacuum. It is usually assumed that the light emission occurs from the 20 nm thick layer of Alq3 adjacent to the TPD/Alq3 interface.¹⁰ Figure 6 shows the calculated dependence of the EL intensity and EL peak wavelength as a function of the emission layers thickness.

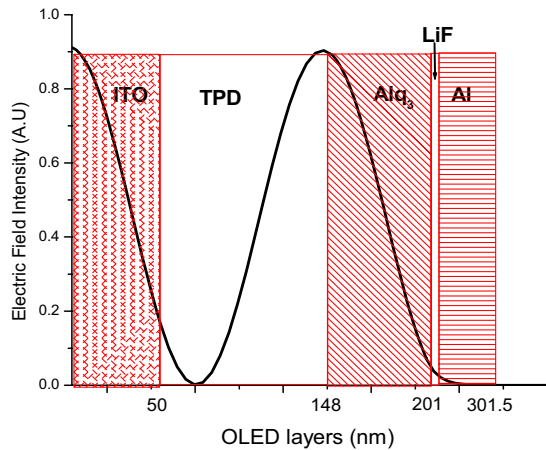


Figure 5: Electric field intensity of optimally designed cavity.

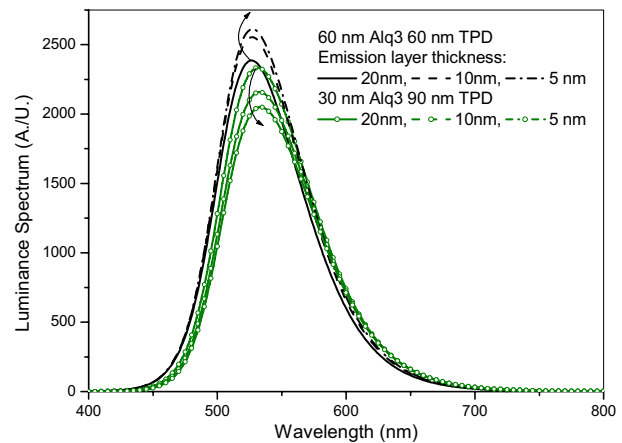


Figure 6: Dependence of the EL intensity and EL peak wavelength as a function of the emission layers thickness.

We have intentionally selected two interface positions (emitting layer positions) with emitting layer on the opposite sides of the standing wave maximum. By reducing the thickness of the emission layer the 60 nm TPD/ 60 nm Alq3 structure is effectively better aligned with the antinode position of the field, while the other structure (90 nm TPD/30 nm Alq3) is effectively misaligned by the reduction on the emission layer thickness. As pointed out before, emission layer thickness is determined by the exciton diffusion length. The exciton diffusion length can be affected by the experimental parameters, such as the purity of material used, vacuum level during deposition, deposition rate etc., as well as experimental conditions during measurement, such as bias, temperature, and measurement environment (with or without encapsulation, in atmosphere or in inert gas). Since some of the experimental factors may be difficult to control in a reliable and reproducible manner, it is difficult to ensure that all the fabricated devices would have the same emission region width. However, it is possible to artificially confine the excitons by insertion of an additional wide band gap organic layer such as bathocuproine, which is typically used in organic light emitting diodes and organic solar cells as hole blocking or exciton blocking layer.^{23,24} In such a manner, the width of the emission region can be sufficiently narrow and very reproducible. The width of the emission region can also be reduced by doping (with low dopant concentrations).²⁵

Figure 7 compares the calculated and experimental EL spectra for two devices fabricated with LiF/Al mirror and with two different thicknesses of the Alq3 layer: 90 nm and 60 nm. Photoluminescence spectra for the same structures are shown in Fig 8. The photoluminescent spectra reveal the same shifts in peak positions as our optical model predicts. EL spectra suggest an increased peak wavelength shift with the position of the emitting layer in comparison to PL and model. In organic materials, the existence of a small emission peak shift between photoluminescence and electroluminescence is not uncommon.^{26,27} The EL spectra can be red shifted²⁶ or blue shifted²⁷ in comparison to the PL spectra, depending on the material. One of the possible reasons for the shift between EL and PL spectra is the formation of an exciplex complex.²⁶ However, the formation of the exciplex complex is not likely in Alq3/TPD based devices. An earlier study indicates that the shift between EL and PL spectra in Alq3 in poly(N-vinylcarbazole) is relatively small (a 2 nm blue shift).²⁷ The magnitude of the shift depends on the material and the device structure. Possibility that a slightly different excited state was produced by photo-excitation and electron injection was proposed as an explanation for the small blue shift between PL and EL.²⁷ It is also possible that the difference is caused by the different emission region thickness. Since the device is very thin, the large electrical field generated when the device is forward biased could affect the width of the emitting region. In addition, some other complex experimental factors, such as enhanced joule heating around pinhole or defect areas, emissive layer degradation due to formation of unstable Alq3 cation species or reaction with oxygen and water in the atmosphere etc., may affect the obtained results. In order to obtain better agreement with the EL experimental data, it would be necessary to fully take into account the charge injection, charge

transport, and exciton diffusion in modeling the emitted spectra. Work is in progress to develop a comprehensive model taking into account both electronic and optical properties of all the layers in the device.

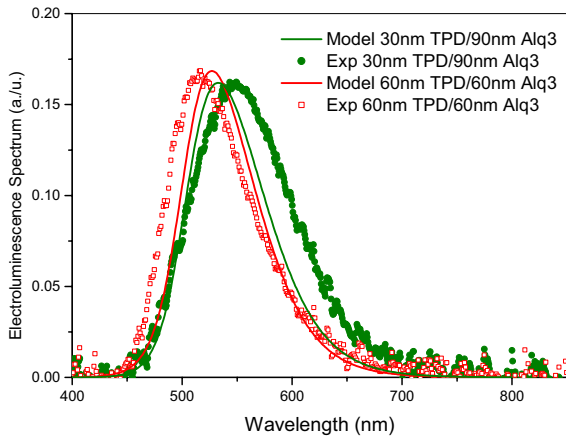


Figure 7: Comparison between experimental results and calculations different for different emission region positions.

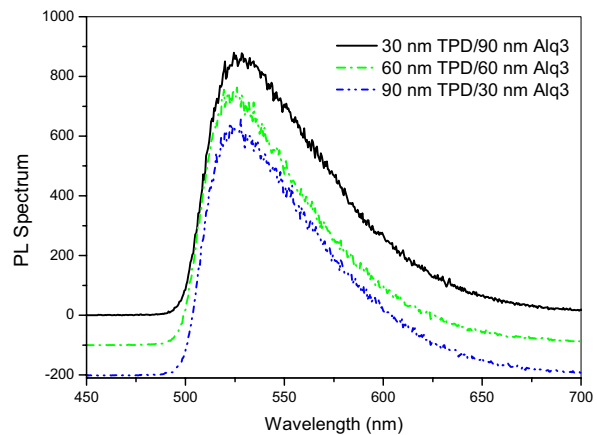


Figure 8: Experimental photoluminescence curves for Emission region positions.

5. CONCLUSION

In conclusion, we have presented calculation of the emitted radiation from organic OLEDs as a function of the choice of cathode, the thickness of organic layers, and the position of hole transport layer/Alq₃ interface. Four cathode structures have been considered: Mg/Ag, Ca/Ag, LiF/Al, and Ag. For the hole transport layer, N,N'-diphenyl-N,N'-(3-methylphenyl)-1,1'-biphenyl-4,4'-diamine (TPD) was used, while tris(8-hydroxyquinoline) aluminum (Alq₃) was the emitting layer. It was found that the emission spectrum is strongly dependent on the layer thickness, interface position, as well as thickness of the emission region. For the optimal efficiency, the emission region should be narrow and aligned with the antinode of the electric field in the resonant cavity. Since the emission region width depends on the exciton diffusion length, in order to confine the excitons to a narrow region insertion of an additional exciton blocking layer or doping of the emitting layer may be advisable. Our simulation results showed very good agreement with experimental photoluminescence results, but the electroluminescence spectra exhibited larger emission wavelength shift with layer thickness change than predicted by our model. The objective of our work was to analyze an OLED structure from an optical point of view, and the electronic considerations (electron injection and charge transport) were not taken into account. In order to obtain better agreement with electroluminescence experimental data, it will be necessary to integrate electrical and optical models. Inclusion of electron injection efficiency for different cathodes and the charge transport in the organic layers will also ensure better prediction of electroluminescence intensity for devices with different cathodes.

ACKNOWLEDGEMENTS

This work has been partially supported by the RGC earmarked grant of Hong Kong (Project HKU 7056/02E) and the University of Hong Kong University Research Committee seed funding grant.

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