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Electroluminescence Spectra in Microcavity Organic Light-emitting Devices

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Abstract Microcavity structure consisting of distributed Bragg reflector and metal silver mirror is designed. The structure is glass/DBR/ITO/TPD/Alq$_3$/Ag. The tris(8-hydroxyquinoline) aluminum (Alq$_3$) is the electron transport layer and the emissive layer; and the N, N′-diphenyl-N, N′-bis(3-methyl-phenyl)-1, 1′biphenyl-4, 4′diamine(TPD) is the hole-transport layer. Compared to the electroluminescence(EL) spectra of non-cavity OLEDs, the linewidth of the MOLEDs is narrower; and the emission peak is enhanced. So the microcavity effect is very observable. In this work, the matrix method was adopted. The dependence of the electroluminescence(EL) spectra on the cavity length, the emitting layer thickness, the position of the interface between EML and HTL and the position of the emission region was analysed detailedly. In all calculation, the thickness and refraction of ITO and the thickness of the metal silver were kept constant. The results show: 1. with increasing the thickness of cavity, the normalized electroluminescence(EL) intensity decreased continually; 2. with increasing the thickness of the emitting layer, the normalized EL intensity discontinuously changed; 3. because the electron mobility in Alq$_3$ is different from the hole mobility in TPD, the emitted radiation was strongly dependent on the position of the emissive layer inside the cavity. Finally, the emission region should be narrow at the center of the electric field in the resonant cavity to optimize MOLED.

Key words: microcavity organic light-emitting device; distributed Bragg reflector; electroluminescence(EL)
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1 Introduction

Since Tang & VanSlyke introduced the first ultra-thin and low voltage organic light-emitting device(OLED)[1], much development has been made to improve this device for applications in flat panel displays. Research on how to improve the device performances continues to be a major focus[2,3].

Recently, microcavity effects which offer the possibility to control the spectral properties of emission have attracted a great deal of attention[4~11]. Because the microcavity enhances the emission rate at the resonance wavelengths and suppresses the emission rate of other wavelengths, it can modify the spontaneous emission rate and the emission spectrum of an optical emitter. So a microcavity structure leads to spectrum narrowing of the emission band, emission peak enhancement, and directional modified output. Several research groups reported the microcavity effects occurred in the microcavity organic light-emitting diodes (MOLEDs)[12~17]. But most of them were researched from the experiments. Some of these papers also discussed the influence of the cavity length on the property of the diodes. But the

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papers which deeply researched the influence of the structure of every layer on the property of the diodes are very few.

In this article, we designed a microcavity organic light-emitting diode. The microcavity structure is glass/DBR/ITO/TPB/Alq$_3$/Ag. We detailedly discussed the dependence of the electroluminescence (EL) spectra on cavity length, the emitting layer thickness, the position of the interface between EML and HTL and the position of the emission region. Results show that the device performances are influenced by not only the cavity thickness, but also the position of the interface between EML and HTL. This result is helpful to the selection of the organic materials and their arrangement in the optimal design of MOLEDs.

2 Model

The schematic structure of the microcavity OLED is shown in Fig. 1. The bottom mirror is composed of a dielectric distributed Bragg reflector (DBR). The DBR consists of three periods of quarter wavelength stack of titanium dioxide and silicon dioxide. The stop band width of DBR is approximately $\lambda_0 \Delta n / n$, where λ_0 is the center of the stop band, Δn is the refractive index difference between the layers of DBR and n is the average refractive index. The refractive index of titanium dioxide and silicon dioxide are 2.4 and 1.46, respectively. A high refractive index difference results in a broad stop band. The top mirror is the metal Ag film. The total optical thickness of the cavity $L(\lambda)$, is given by[15]

$$L(\lambda) = \frac{\lambda}{2}\left(\frac{n}{\Delta n}\right) + \sum_i n_i d_i + \frac{|\Phi_m|}{4\pi}\lambda \quad (1)$$

where n_i and d_i are the refractive index and thickness of indium-tin oxide (ITO) and the organic films between the two mirrors, Φ_m is the phase shift at the metal mirror, and λ is the wavelength in the microcavity. The first term of Eq. (1) is the penetration depth of the electromagnetic field into DBR, the second term is the sum of optical thickness of the layers between the two mirrors, and the last term is the effective penetration depth into the top metal mirror. The phase shift upon reflection from the mirrors is calculated using the matrix method[12]

$$\Phi_m = \arctan\left[\frac{2n_s k_m}{n_m^2 - n_s^2 + k_m^2}\right] \quad (2)$$

where n_m and k_m are the real and imaginary parts of the refractive index of the metal, and n_s is the refractive index of the material in contact with the metal. The values of these refractive indexes are the functions of the wavelength. The resonance modes are determined by the relation $m\lambda = 2L(\lambda)$, where m is the mode index and it is an integer. By modify the optical length, the mode positions and mode spacing can be varied. The theoretical spectrum for emission normal to the plane of the devices layers was calculated following the approach of Deppe *et al.*[16]. The calculated spectrum compared with that of non-cavity structure is[13]:

$$|E_m(\lambda)|^2 = \frac{\frac{1-R_{DBR}}{j}\sum_j\left[1 + R_m + 2\sqrt{R_{DBR}}\cos\left(\frac{4\pi x_j}{\lambda}\right)\right]}{1 + R_m R_{DBR} - 2\sqrt{R_m R_{DBR}}\cos(4\pi L/\lambda)} \cdot |E_{nc}(\lambda)|^2 \quad (3)$$

where R_m and R_{DBR} are the reflectivity of the metal and DBR mirror, respectively. L is the total optical thickness of the cavity given in Eq. (1), $|E_{nc}(\lambda)|$ is the free space emission intensity, and x_j is the effective distance of the emitting dipoles from the metal mirror.

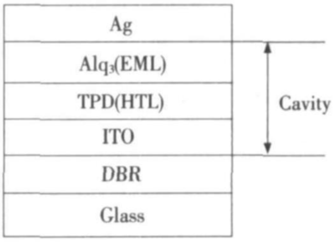

Fig. 1 Structure diagram of the microcavity organic light-emitting diode

In order to get the dependence of the peak intensity of the EL spectra on the organic layer thickness, the thickness variation of the organic films is used to control the effective cavity length. In calculation, the thickness and refraction of ITO is 220 nm and 2.0 respectively, and the metal silver thickness is 100 nm.

3 Results and Discussion

The cavity length plays an important role in the design of the microcavity OLED. In this paper, the thickness variation of the organic films is used to control the effective cavity length. Fig. 2 shows the EL spectra of the MOLEDs with different organic layer thickness. It can be seen that the microcavity effect is very notable. The spectral narrowing at the resonance wavelength is observed. Varying the thickness of the organic films, the resonance wavelength can be selectively scanned over a very wide range of wavelengths that cover almost 140 nm. And with the increasing of thickness of the organic film from 60 nm to 90 nm, the normalized EL intensity decreased continually. Based on the resonance condition $m\lambda = 2L(\lambda)$, we can get that when the thickness is 130 nm, two resonance peaks were appeared at visible light region which locate at 425 nm and 655 nm, respectively. The peak located at 650 nm is stronger than another. In all calculations we assumed physical thickness of the HTL (TPD) and the ETL (Alq_3) layers equal to a half of the cavity length. According to the Schubert-Hunt's theory[18], minimization of the cavity length would maximize the integrated intensity. Thus, the shortest possible cavity length could lead to a maximized resonant light intensity. It can be understood easily. The enhancement factor of emission peak value is given by[19]:

$$F = \frac{\pi\sqrt[4]{R_{DBR}R_m}}{1-\sqrt{R_{DBR}R_m}} \tag{4}$$

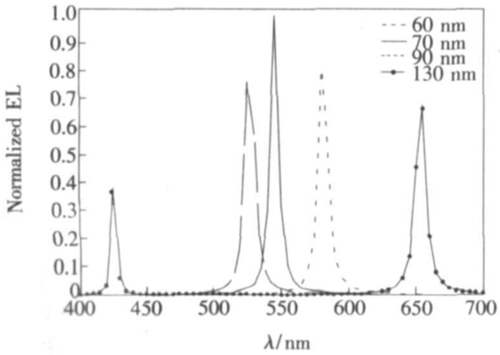

Fig 2 The normalized EL spectra with different organic layer thicknesses.

Because R_{DBR} and R_m are the same in the all calculations, the F is a constant. But with the cavity length increasing, the mode index increased, *i.e.* the enhancement factor of every singer mode decreased. So the result we got agreed with the theory.

According to Ref. [12], it is also important that an Alq_3 thickness of 30~100 nm is optimal from the electrical standpoint, larger thicknesses will lead to significant increases in the forward voltage drop, while smaller thicknesses will result in a substantial lowering of EL efficiency because of exciton quenching at the Alq_3/metal interface. Fig. 3 shows the normalized EL intensity of the MOLEDs with different ETL layer thickness. With the thickness of Alq_3 increasing from 30 nm to 65 nm, the EL intensity is an oscillatory curve. And with the thickness increasing, the peak value decreased. It can be explained by the experiment result from the Ref. [20]. In the Ref. [20], it has been given that with the cavity length increasing, spontaneous emission rate of an electric dipole in a planar microcavity changed discontinuously in the planar semiconductor microcavity. When $L < \lambda/2$, the spontaneous emission rate of the dipole was suppressed partly. And there are discontinuous points at the certain positions. So in the Fig. 3, the peaks located at the certain wavelengths which were the discontinuous points to the spontaneous emission rate. In the all calculations, we assumed the free space emission intensity $|E_{nc}(\lambda)|^2$ is equal to 1.

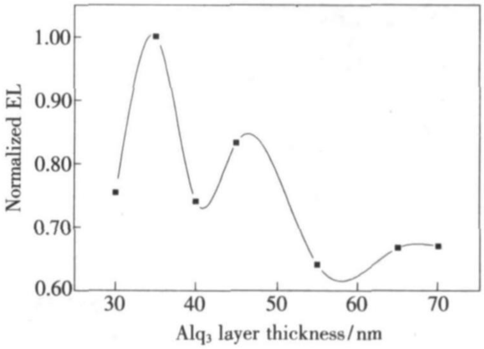

Fig 3 The normalized EL intensity with different Alq_3 layers thicknesses

Fig. 4 shows the normalized EL spectra with different interface position of EML/HTL. We changed the thickness of the ETL (Alq_3) to investigate the effect of the position of the light emitting layer on the EL spectrum. Because of the difference between the refractive indices of TPD and Alq_3, the total physical cavity length is almost dependent on the position of the TPD/Alq_3 interface. Keeping the other parameters of the microcavity constants, this enabled us to single out and investigate the effect of the position of the emitting layer on the EL spectrum. However, one has to bear in mind that changing the relative thicknesses of TPD and Alq_3 electron transport will be significantly affected due to the different carrier mobility in the two layers. The mobility of electrons in Alq_3 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. From Fig. 4 with the interface varying from 25 nm to 45 nm, the EL intensity decreased. To the structure of 25/45, the result is not reasonable, if we considered the exciton quenching at the Alq_3/metal interface. The result of the structures 35/35 and 45/25 agreed with Fig. 3, because the interface of the structure 35/35 is nearer the antinode of the resonant cavity than the interface of the structure 45/25. So the stronger EL enhancement is obtained when the emission layer is aligned with the position of the antinode of the resonant cavity of the ETL layer affected strongly the EL intensity.

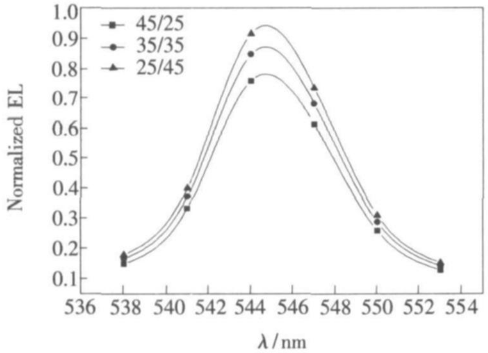

Fig. 4 Calculated normalized EL spectra versus different interface position of EML/HTL.

It is usually assumed that the light emission occurs from the 20 nm thick layer of Alq_3 adjacent to the TPD/Alq_3 interface[12]. Fig. 5 shows the calculated dependence of the EL intensity as a function of the emission region thickness. The thickness of the emission region within the Alq_3 film was varied between 5 and 20 nm. By reducing the thickness of the position of the emission region of the 35 nm Alq_3/35 nm TPD structure, the position of the emission region is better aligned with the position of the antinode of the resonant cavity. In Fig. 5, the normalized EL spectra increased with the emission region thickness increasing. It is reasonable, because with the recombination region becoming wider, the number of the carrier recombination became more. Therefore, to maximize the emitted power, the antinode of the optical field should be aligned with the middle of the emission region. So the thickness of the Alq_3 in the optimally designed device is determined by the thickness of the light emission region.

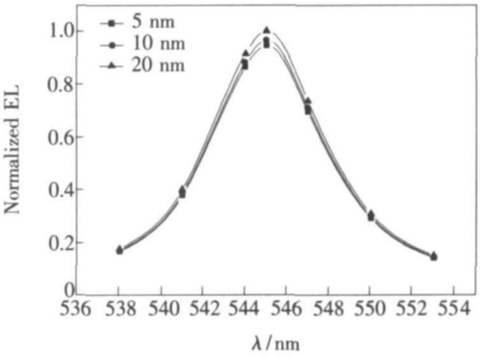

Fig. 5 The normalized EL spectra with different emission region thickness

4 Conclusion

We researched the dependence of the cavity structure on the EL intensity deeply. We detailedly discussed the normalized EL spectra from the microcavity as the function of the thickness of the organic layers, and the position of TPD/Alq_3 interface. It was found that the emission spectrum is strongly dependent on the layer thickness and the position of TPD/Alq_3 interface, as well as the thickness of the emission region. For the optimal efficiency, the emission region should be narrow and its center aligned with the antinode of the electric field in the resonant cavity.

References

[1] Tang C W, VanSlyke S A. Organic electroluminescent diodes [J]. *Appl. Phys. Lett.*, 1987, **51**(12): 913-915.

[2] Xu Xurong, Xu Zheng, Qu Chong, *et al*. Dispute between molecular and band theory in organic electroluminescence [J]. *Chin. J. Lumin.* (*发光学报*), 2005, **26**(1): 1-7 (in Chinese).

[3] Liu Hongwei, Laskar I R, Huang Chiping, *et al*. Synthesis and applications of luminescent CdSe quantum dots for OLED [J]. *Chin. J. Lumin.* (*发光学报*), 2005, **26**(3): 321-326 (in English).

[4] Jordan R H, Rothberg L J, Dodabalapur A, *et al*. Efficiency enhancement of microcavity organic light emitting diodes [J]. *Appl. Phys. Lett.*, 1996, **69**(14): 1997-1999.

[5] Lu M H, Weaver M S, Zhou T X, *et al*. High-efficiency top-emitting organic light-emitting devices [J]. *Appl. Phys. Lett.*, 2002, **81**(21): 3921-3923.

[6] Riel H, Karg S, Beierlein T, *et al*. Tuning the emission characteristics of top-emitting organic light-emitting devices by means of a dielectric capping layer: an experimental and theoretical study [J]. *J. Appl. Phys.*, 2003, **94**(8): 5290-5296.

[7] Chen C W, Hsieh P Y, Chiang H H, *et al*. Top-emitting organic light-emitting devices using surface-modified Ag anode [J]. *Appl. Phys. Lett.*, 2003, **83**(25): 5127-5129.

[8] Zhang Hongmei, You Han, Wang Wei, *et al*. Organic white-light-emitting devices based on a multimode resonant microcavity [J]. *Semiconductor Science and Technology*, 2006, **21**(8): 1094-1097.

[9] Cheng C H, Djuri》i A B, Kwong C Y, *et al*. Reduced angular dependence of the emission from tris(8-hydroxyquinoline) aluminum based microcavity [J]. *Opt. Commun.*, 2005, **248**(1-3): 287-293.

[10] Ma Fengying, Jin Changqing, Liu Yun, *et al*. Three color emission from organic light emitting diode in ultra-short metal microcavity [J]. *Chin. J. Lumin.* (*发光学报*), 2003, **24**(2): 144-146 (in Chinese).

[11] Sun Xiaoyan, Li Wenlian, Hong Ziruo, *et al*. Improved performance of Eu complex OLEDs with microcavity structure [J]. *Chin. J. Lumin.* (*发光学报*), 2005, **26**(2): 262-264 (in Chinese).

[12] Dodabalapur A, Rothberg L J, Jordan R H, *et al*. Physics and applications of organic microcavity light emitting diodes [J]. *J. Appl. Phys.*, 1996, **80**(12): 6954-6964.

[13] Jung B Y, Kim N Y, Lee C H, *et al*. Optical properties of microcavity structures using the organic light emitting materials [J]. *Mat. Res. Soc. Symp. Proc.*, 2000, **621**: Q3.5.1-Q3.5.6.

[14] Juang F S, Laih L H, Lin C J, *et al*. Angular dependence of the sharply directed emission in organic light emitting diodes with a microcavity structure [J]. *Jpn. J. Appl. Phys.*, 2002, **41**(4B): 2787-2789.

[15] Li Yongjun, Shi Jiawei, Liu Mingda, *et al*. Linewidth narrowing and intensity enhancement of wavelength tunable MOLEDs [J]. *Semiconductor Photonics and Technology*, 2001, **7**(1): 13-16.

[16] Peng H J, Wong M, Kwok H S. Design and characterization of organic light emitting diodes with microcavity structure [J]. *SID Int. Symp. Digest Tech. Papers*, 2003, **32**: 516-519.

[17] Hou Lintao, Hou Qiong, Mo Yueqi, *et al*. All-organic flexible polymer microcavity light-emitting diodes using 3M reflective multilayer polymer mirrors [J]. *Appl. Phys. Lett.*, 2005, **87**(24): 243504-1-3.

[18] Han Sijin, Huang Changjun, Lu Zhenghong. Color tunable metal-cavity organic light-emitting diodes with fullerene layer [J]. *J. Appl. Phys.*, 2005, **97**(9): 091302-1-5.

[19] Liu Xingyuan, Zhao Jiamin, Liu Yun, *et al*. Photoluminescent properties of organic film in flat optical microcavity [J]. *Spectroscopy and Spectral Analysis* (*光谱学与光谱分析*), 2000, **20**(2): 212-215 (in Chinese).

[20] Liu Wenkai, Lin Shining, Qu Yanfeng, *et al*. Spontaneous emission of electrical dipoles in a semiconductor microcavity [J]. *J. Optoelectronics•Laser* (*光电子•激光*), 2002, **13**(1): 12-15 (in Chinese).

[21] Naka S, Okada H, Onnagawa H, *et al*. Carrier transport properties of organic materials for EL device operation [J]. *Synth. Met.*, 2000, **111-112**: 331-333.

微腔有机发光器件中的电致发光光谱

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摘要: 设计了结构为 Glass/DBR/ITO/TPD/Alq$_3$/Ag的微腔有机发光器件。从理论上详细地研究了腔内各层结构对器件电致发光谱性能的影响。结果表明:随着腔长厚度的增加,器件的归一化电致发光谱强度不断减小;在可见光区,器件的 EL谱随发光层厚度的增加出现振荡变化。空穴传输层和发光层的界面位置对器件电致发光谱的影响也很大。最后得到,在设计微腔时发光层厚度要尽量窄,并且中心发光区域应位于谐振腔中电场的峰值位置。

关　键　词: 微腔有机发光器件; 分布布喇格反射镜; 电致发光(EL)

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