Tricolor microcavity OLEDs based on P-nc-Si:H films as the complex anodes*

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Abstract: A P$^+$-nc-Si:H film (boron-doped nc-Si:H thin film) was used as a complex anode of an OLED. As an ideal candidate for the composite anode, the P$^+$-nc-Si:H thin film has a good conductivity with a high work function (~ 5.7 eV) and outstanding optical properties of high reflectivity, transmission, and a very low absorption. As a result, the combination of the relatively high reflectivity of a P$^+$-nc-Si:H film/ITO complex anode with the very high reflectivity of an Al cathode could form a micro-cavity structure with a certain Q to improve the efficiency of the OLED fabricated on it. An RGB pixel generated by microcavity OLEDs is beneficial for both the reduction of the light loss and the improvement of the color purity and the efficiency. The small molecule Alq would be useful for the emitting light layer (EML) of the MOLED, and the P$^+$-nc-Si film would be used as a complex anode of the MOLED, whose configuration can be constructed as Glass/LTO/P$^+$-nc-Si:H/ITO/MoO$_3$/NPB/Alq/LiF/Al. By adjusting the thickness of the organic layer NPB/Alq, the optical length of the microcavity and the REB colors of the device can be obtained. The peak wavelengths of an OLED are located at 486, 550, and 608 nm, respectively. The CIE coordinates are (0.21, 0.45), (0.33, 0.63), and (0.54, 0.54), and the full widths at half maximum (FWHM) are 35, 32, and 39 nm for red, green, and blue, respectively.

Key words: P$^+$-nc-Si:H thin film; organic light emitting display; microcavity; tricolor
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1. Introduction

Recently, the world flat-panel display (FPD) industry has experienced a rapid progress of fabrication technologies. For example, the liquid crystal display (LCD) and the plasma display panel (PDP)[1] have become major display panels instead of the traditional cathode ray tubes (CRT). However, due to their advantages of initiative irradiance, instantaneous response, and low power, self-illuminating organic LEDs (OLED) are the most potential display panel type of the next generation[2]. An OLED has the properties of a flexible substrate, a high brightness, and a high resolution[3]. The realization of an explicit coloration should be the most important goal for the OLED exploitation, and it determines the competitive power of OLEDs on the flat panel display market. According to the classification of panel types, the technical realization of explicit coloration of OLEDs has following several ways: a light-color conversion; the usage of a color filter membrane; R, G, and B tricolor independent luminescence; and a microcavity color tuning method.

Light-color conversion can be carried out by combining the blue light OLED and a light-color converting membrane and by utilizing the energy transfer between the materials[4]. Based on the blue light OLED device, the red and green lights can be aroused under the excitation of blue light. The disadvantage of such kind of conversion is that, the blue light will be absorbed by the surroundings of the material, and this lowers the picture quality.

In the color membrane sieve method[5], a white-light OLED device was first fabricated, and the tricolor can be obtained by color membrane sieves. Finally, the three base colors can be combined to form a color display. The key technique is the realization of a high efficiency and a high-purity, white light. However, this leads to enormous light loss, when the white light passes through the membrane.

In the independent tricolor method, the red, green, and blue tricolors were first generated separately as the basic color centres of luminescence. Afterwards, they were mixed according to certain ratios to produce a real nature color. Each independent light unit represents a picture element. The key point of the technique is to enhance the purity and luminosity of the device. At present, Alq$_3$ is a very good small molecule material with green light, and its color purity, luminosity, and stability are very good. But the red and blue light small molecule material has comparatively weak luminosity and a short lifetime. Recently, by adding the dopant to the main body of the luminescent material, the color purity, the luminosity, and the stability have been clearly improved. However, to successfully fabricate red, green, and blue basic luminescence centers separately needs more complex manufacturing techniques.

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The microcavity color tuning method is a special method, in which the tricolor is emitted. The luminescent material in an OLED can be white light or a single base color (as green) material. For example, the green light Alq$_3$, by tuning the length of the microcavity, makes the wavelength of the light from the microcavity resonant with the wavelength of red, green, and blue tricolor accordingly, to realize the tricolor display. The advantage of this method is that it can improve the intensity of luminescence and enhance the color purity effectively with a comparatively simple technique. By taking advantage of the white light and microcavity effect, Sony Company[6] have manufactured a 12.5 inch organic full color display monitor.

An OLED microcavity is composed of two parallel plane reflectors, namely the Fabry–Perot (F–P) resonance cavity[7], which is used to determine the required wavelength of the light. The reflecting mirror is made of aluminum acting as a cathode, and the half reflecting surface is made of half reflecting or half penetrating materials. The hydrogenized nano crystal silicon (P$^+$-nc-Si:H) thin films have a small absorptivity and half reflection and half penetration of optics in the visible light range. Thus, by using it as the anode of an OLED, it can be fabricated into a microcavity OLED (MOLED).

In this paper, by using a VHF-PECVD technique and doping with boron, the P$^+$-nc-Si:H thin film was fabricated successfully, and its optical and electrical properties are investigated. Based on a P$^+$-nc-Si:H thin film as an anode, the MOLED is also fabricated, and its luminescent properties are studied. By regulating the thickness of an organic layer, red, green, and blue base-color devices are obtained. Experimental results indicate that our MOLED devices have a very good microcavity effect with a narrow spectrum width and an enhanced EL intensity. It would be fruitful to research tricolor OLEDs by the color-tuning microcavity method further.

2. Fabrication of a P$^+$-nc-Si:H thin film and its electrical and optical properties

P$^+$-nc-Si:H thin film materials were fabricated by a PECVD cluster thin film depositing system in the optical-electric lab of the Nankai University. Cleaned EAGEL2000 optical glass, used as the substrate, was put into the upper electrode of this system chamber with a vacuum of less than 5.0×10^{-6} Pa. The substrate was heated in the range of 170–250 °C. The distance between the two electrodes can be changed from 20 to 50 mm. The reacting gases of SiH$_4$, H$_2$, and B$_2$H$_6$ (Hydrogen gas 0.1%) can be controlled and checked by a flowmeter (Brooks 8085E). The gas pressure was varied within 10–150 Pa. The applied 60 MHz VHF power loaded from the bottom electrode was from 6 to 30 W.

The optimized deposition condition is chosen as follows: SC = 4%, bottom temperature $T_s = 250$ °C, reacting pressure $P_s = 1.7$ Torr, power span from 12 to 50 W, and the doped concentration of boron is 0.9%. In order to obtain different thicknesses of the nano crystal silicon films, the deposition times are chosen as 8, 9, 10, 11, 12, and 13 min, respectively, which corresponds to a thickness ranging from 30 to 50 nm .

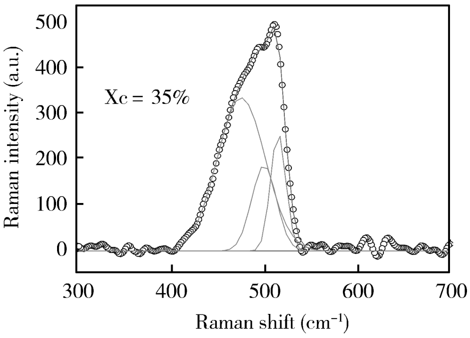

Fig. 1. Raman spectra of the sample and three-peak Gaussian fitting diagrams.

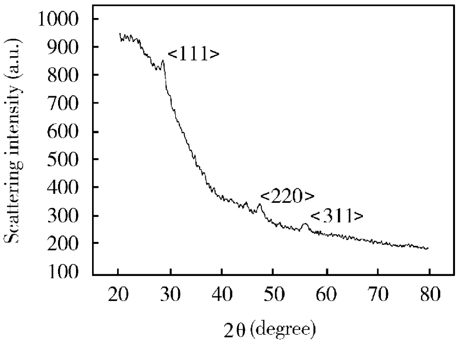

Fig. 2. XRD pattern of the sample with a thickness of 40 nm.

Figures 1 and 2 are the Raman spectra and the XRD pattern of the sample with a film thickness of 40 nm. From Fig. 1, we see that the sharp Raman peak (the doping concentration of boron has a mole ratio of 0.9%) is located at 516 cm^{-1}, demonstrating that the sizes of the crystal grains are small. By simulating the experimental curve of the Raman spectra with the three-peak Gauss fitting method of the Origin software, we obtain the formula of the crystallization coefficient X_c to be[8]:

$$X_c = (I_{500} + I_{520})/(I_{500} + I_{520} + I_{480}), \tag{1}$$

And, thus, the X_c of P$^+$-nc-Si: H film is about 35%.

From Fig. 2, we know that in the case of a 0.4% boron concentration, there are three crystal orientations in the silicon film namely <111>, <220>, and <311>. Based on the Debye–Scherer formula[9],

$$d = \frac{K\lambda}{B \sin\theta}, \tag{2}$$

the average crystal grain sizes of the film along the three directions <111>, <220>, and <311> are given as 15, 17, and 21 nm, respectively. The measured conductivity of the sample is $\sigma = 5.86$ S/cm. Thus, a good electrical performance of the P$^+$-nc-Si:H film was obtained.

The optical energy gap of the film was measured by using OMA, giving a value of more than 2.0 eV. The transmission, absorption, and reflection coefficients of the P$^+$-nc-Si:H thin film were measured in the visible light range, as shown in Fig. 3.

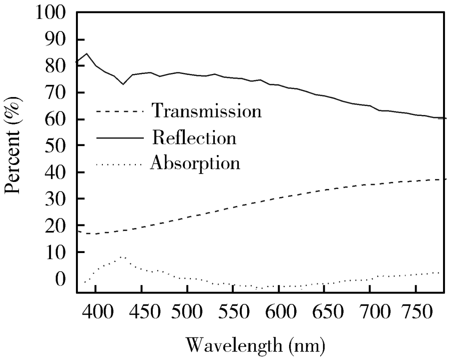

Fig. 3. Optical transmission, reflection, and absorption of glass/LTO/P-nc-Si:H.

From Fig. 3, we can see that the 40 nm P$^+$-nc-Si:H thin film has a reflectivity and transmission in the visible light range. With the increase of the wavelength, the reflectivity tends to decrease, and the transmittance tends to increase. But the absorbance of light is less than 5%. Figure 3 suggests that the 40 nm P$^+$-nc-Si:H thin film can be used as an anode of a MOLED in the range of visible light, just as we expected, and a microcavity with superior properties has been made.

3. Tricolor MOLED based on P$^+$-nc-Si:H compounds

We first fabricate an OLED by using the small molecular Alq as the luminescent material and ITO and nano silicon compound thin films (nc-Si/ITO) as the two anodes of the two OLEDs in the following structure:

Glass/ITO(80 nm)/MoO$_3$(0.5 nm)/NPB(108 nm)/Alq(65 nm)/LiF(1 nm)/Al(100 nm), and Glass/P$^+$-nc-Si:H(40 nm)/ITO(80nm)/MoO$_3$(0.5 nm)/NPB(108 nm)/Alq(65 nm)/LiF (1 nm)/Al(100 nm).

Here, MoO$_3$ is acting as a hole injection layer (0.5 nm)[10], NPB (108 nm) is a layer for hole transportation, Alq (65 nm) is used as a luminescent layer, and LiF (1 nm)/Al (100 nm) is the cathode. Two anodes were fabricated by ITO (80 nm) and P$^+$-nc-Si:H (80 nm)/ITO (80 nm).

Figure 4 is the comparison of EL spectra of MOLED with P$^+$-nc-Si:H/ ITO and ITO anodes. The triangle-line represents the spectrum of the anode with P$^+$-nc-Si:H/ITO, and the square-line represents that of the normal anode made by ITO. The left chart is the normalized spectrum, and the right picture represents the non-normalized one. From Fig. 4(a), we can see that the spectrum with P$^+$ -nc-Si:H/ITO as its anode evidently narrows compared to that of the anode made by ITO. The spectrum changes from 126 to 33 nm. Both of the central wavelengths of the two devices are at 550 nm, which is typical of green light. From Fig. 4(b), the intensity of the emitted light is greatly enhanced. The light intensity of the device made by P$^+$-nc-Si:H/ITO is about 25 times higher than that of ITO.

From Table 1 we notice that, the the microcavity OLED with a P$^+$-nc-Si:H/ITO anode can improve the EL color center efficiently, and the CIE value approaches the value of a standard CIE. The purity of the color greatly improves, which is good for composing color images. This illustrates that the color purity of our device gains a desirable improvement, useful in the synthesis of standard tricolor OLED devices.

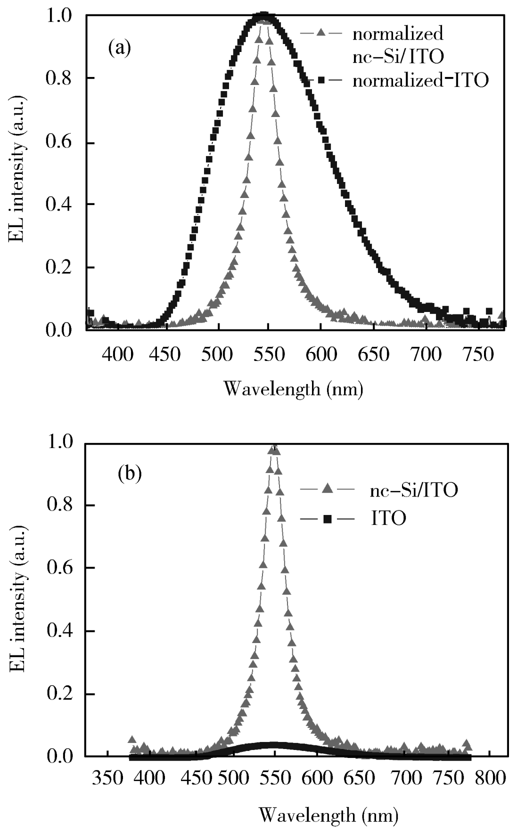

Fig. 4. Comparison of the EL spectra for P-nc-Si: H/ITO and ITO anodes.

Table 1. CIE of two anode materials.

Anode	x	y
ITO	0.39	0.53
P-nc-Si:H/ITO	0.33	0.63
CIE	0.21	0.71

In order to understand how the anode affects the optical properties of an MOLED, the current density–voltage relation, luminescence–voltage relation, and luminescent efficiency–current density relation are measured, as shown in Figs. 5, 6, and 7, respectively.

Figure 5 shows the relationship between the current density and the voltage of two different anodes. We find that two devices have nearly the same I–V relation, demonstrating the good current–voltage properties of the device.

The work function of the P$^+$-nc-Si:H/ITO can be considered as follows: normally, the electron affinity of the silicon thin film is 4 eV; but for a nanocrystalline silicon (nc-Si: H) thin film, whose energy band gap is between 1.2 and 1.5 eV, the electron affinity depends on the crystallization ratio. The energy band gap of our sample (P$^+$-nc-Si:H thin film) is wider than 2.0 eV. According to the doping efficiency, the active energy is 0.1–0.3 eV over the top of the Valence Band[11]. The Fermi energy level of the P$^+$-nc-Si:H thin film, whose energy band gap is over 2.0 eV, is 1.7 or 1.9 eV below the bottom of the conduction band. The electron affinity of the silicon thin film is 4 eV. As a result, the work function of the P$^+$-nc-Si:H

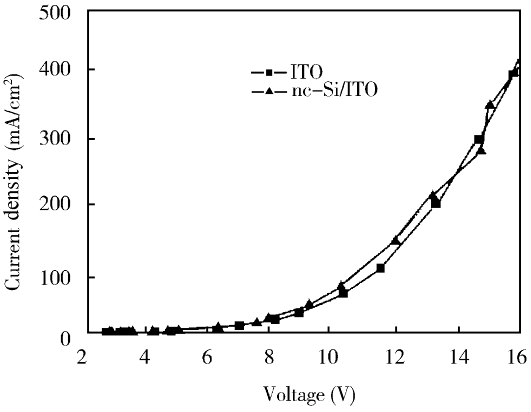

Fig. 5. Current density–voltage characteristics of the EL devices with the P-nc-Si:H/ITO and the ITO anode.

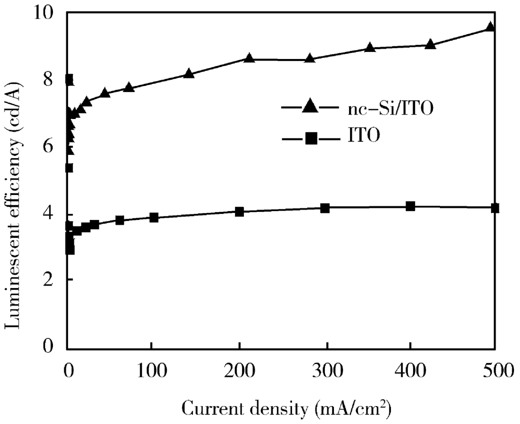

Fig. 7. Luminescent efficiency and current density characteristics of devices with P-nc-Si:H/ITO.

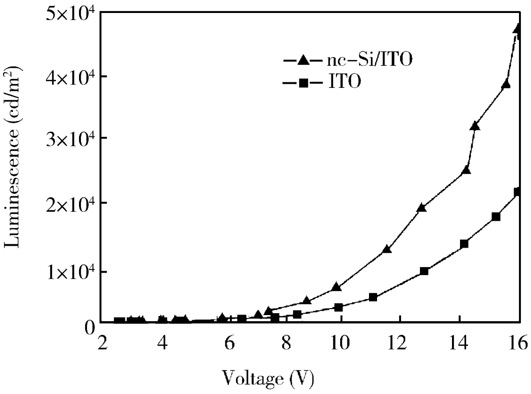

Fig. 6. Luminescence–voltage characteristics of devices with a P-nc-Si:H/ITO and an ITO anode.

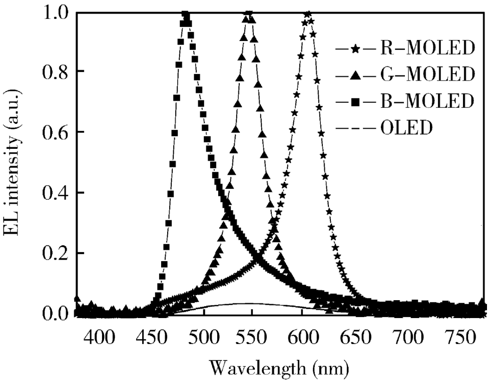

Fig. 8. Spectra comparison of the red, green, and blue EL devices with the P-nc-Si: H/ITO anode.

thin film is 5.7 or 5.9 eV. The current in the device can be improved because the work function of the P^+-nc-Si:H thin film is high and, thus, the cavity injection capability is high. The sizes of the grains in the thin film also have a certain influence on the conductivity and the work function. For a certain amount of grains, the crystallization ratio is decided by the size of the grain. So, the energy band gap width and work function of the thin film are different. The conductivity can also be changed according to the crystallization ratio.

Figures 6 and 7 show luminescence versus voltage and luminescent efficiency versus the current density of two kinds of anode devices. From the figures we can see that the luminescence of an MOLED with a P^+-nc-Si:H/ITO anode is much larger than that of an ITO anode when applied voltage exceeds 12 V. In addition, the luminescent efficiency as a function of the current density in the two cases has a distinct performance. When the current density is 493 mA/cm^2, the luminescent efficiency can reach the highest value of 9.5 cd/A. This is higher than that of the ITO anode devices, corresponding to an enhanced efficiency of 127%. For the luminescence, as shown in Fig. 6, when the applied voltage is 16.5 V, the luminescence of the P^+-nc-Si:H/ITO MOLED can reach a value as high as 47130 cd/cm^2, which is much larger than that of ITO anode.

If we still use Alq as the light emitting material, we can get light with different wavelengths by adjusting the length of the microcavity. Using Alq as the luminescent material and changing the effective length of the microcavity, we can obtain several characteristic spectra with different wavelengths. Since the emitted light comes from the resonant microcavity, it makes the light brighter, the spectrum narrower, and the desirable color saturation can be obtained. The key technology we used here is that the different color irradiance are coming from the same layer, in which the difficulty of the adjustment of the cover membrane board in the vacuum chamber for the full color synthesis can be avoided. That is the reason why it attracts great attention of many researchers[12–16].

In this way, we can obtain different characteristic colors by changing the thickness of the organic layer instead of changing the luminescent materials. The fabrication structures of the RGB devices are given as follows.

For red (A) color:

Glass/P^+-nc-Si:H/ITO(80 nm)/MoO_3(0.5 nm)/NPB(140 nm)/ Alq(70.6nm)/LiF(1nm)/Al(100nm).

For green (B) color:

Glass/P^+-nc-Si:H/ITO(80 nm)/MoO_3(0.5 nm)/NPB(108 nm)/ Alq(65 nm)/LiF(1 nm)/Al(100 nm).

For blue (C) color:

Glass/P^+-nc-Si:H/ITO(80 nm)/MoO_3(0.5 nm)/NPB(82 nm)/ Alq(75 nm)/LiF(1 nm)/Al(100 nm).

The nano silicon compound structure of the composite anodes of the above three devices are taken to be the same as P^+-nc-Si:H(40 nm)/ITO(80 nm). By regulating the thickness of the hole transport layer NPB and the luminescent layer Alq, we can construct three microcavities, which generate the three

Table 2. Characteristic parameters of the RGB MOLED.

Structure	Thickness (nm)	Efficiency (cd/A)	Peak wave-length (nm)	FWHM (nm)
Red MOLED	210.6	2.8	608	39
Green MOLED	173	9.5	550	32
Blue MOLED	157	1.9	486	35

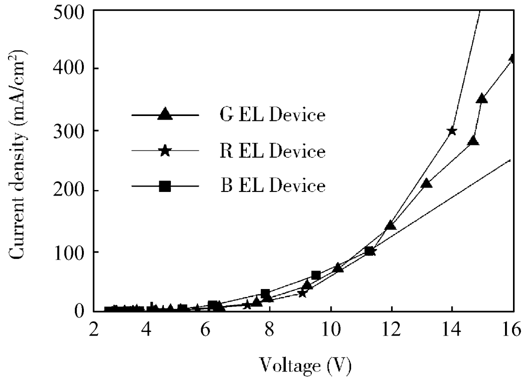

Fig. 9. Current density–voltage characteristics of the red, green, and blue EL devices with the P-nc-Si:H/ITO anode.

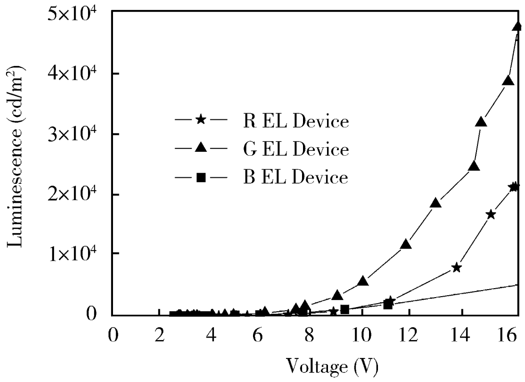

Fig. 10. Luminescent–voltage characteristics of the red, green, and blue EL devices with the P-nc-Si: H/ITO.

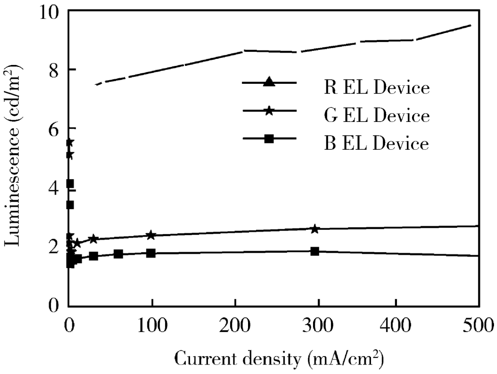

Fig. 11. Luminance efficiency–current density characteristics of the red, green, and blue EL devices with the P-nc-Si:H/ITO anode.

base colors. Figure 8 is the EL tricolor spectra obtained by fabricating the nano silicon compound anode on the device. By changing the thickness of the organic layer, the red, green, and blue LEDs, corresponding to microcavity lengths of 210.6, 173, and 157 nm, respectively, can emit light at wavelengths of 608, 550, and 486 nm with a spectrum half width of 39, 32 and 35 nm. The black line in Fig. 8 is the emission spectrum of a traditional Alq based LED, where the half width is about 126 nm.

The characteristic parameters of red, green, and blue naked MOLEDs are presented in Table 2. From Table 2, we find that with the regulation of the thickness of an organic layer, the current efficiency, and the spectra peak values are changing accordingly. This generates the red, green, and blue emissions, respectively. We note that, because the luminescence centre of Alq is located at 550 nm, the current efficiency of the spectrum in which the spectrum peak is close to 550 nm is comparatively higher than others in which the spectrum peak is far away from 550 nm. In order to understand the properties of the nano crystal silicon anode MOLED, the current density–voltage, the luninescent–voltage, and the luminescent efficiency–current density relations of our red, green, and blue OLED are studied, as shown in Figs. 9, 10, and 11, respectively.

From Figs. 9 and 10, we know that the open voltage of red, green, and blue light of a MOLED is the same at a value of 2.5 V. At the same voltage value, green light shows the strongest luminescent, but the current density is between that of the red and blue light diodes. The blue light shows the lowest current density and luminance.

From Fig. 11, we can see that the luminescent efficiency of the green light MOLED is higher than for the other two. It reaches a maximum value of 9.54 cd/A at a current density of 493 mA/cm^2. In comparison, the red light and the blue light MOLED show a stable small value and a slight increase with increasing current density. The red light luminescent efficiency is in the range of 2–3 cd/A, and the blue one is 1–2 cd/A. From the above discussion, we know that the fabrication of the RGB MOLED is successful and controllable by simply regulating the thickness of the organic layer (the length of the microcavity) in the devices.

4. Conclusion

The anode, which is made by a P$^+$-nc-Si:H thin film or by ITO, and the metal Al cathode can form the microcavity structure in an OLED because of the properties of high reflection, transmission, and small absorption of the P$^+$-nc-Si:H thin film in the visible light range. The microcavity structure was successfully fabricated by combining a complex anode and a high reflection aluminum cathode. The intensity of the emitted light of the microcavity device are enhanced and the spectrum width is narrowed. The luminescent center of the emitting spectrum is located at 550 nm, and it can reach a maximum value of the luminance of 47130 cd/m^2 and a luminance efficiency of 9.5 cd/A. Compared with tradition ITO devices, it has a value enhanced by about 127%.

In terms of the luminescent material Alq and by regulating the effective length of the microcavity, an RGB tricolor

MOLED can be obtained with center wavelengths of 486, 550, and 608 nm, color coordinates of (0.21, 0.46), (0.33, 0.63), and (0.54, 0.54), and FWHM of 35, 32, and 39 nm, respectively. Experimental results indicate that the microcavity color tuning method and more deep research on the color display are practical and reliable.

References

[1] Xue J Z. Future slab shows of compete—OLED to LCD's impact with countermeasure brief analysis. New Material Industry, 2008, Issue 1: 20 (in Chinese)

[2] Xie Y Q, Guo J H, Peng J B, et al. High-efficiency red-phosphorescent 6CPt electroluminescence. Acta Phys Sin, 2005, 54(8): 3424 (in Chinese)

[3] 3GPP TS 25.346: introduction of the multimedia broadcast multicast service (MBMS) in the radio access network (RAN); stage 2 [S]. 2006

[4] Wang J, Miao H, Yang A L, et al. Study of luminescence ability of organic white light emitting diode by light conversion. Spectroscopy and Spectral Analysis, 2005, 25(5): 672

[5] Meng Z, Kwok H, Wu C, et al. Fabrication of a 125 mm poly-Si TFT active-matrix driving color AMOLED. Chinese Journal of Semiconductors, 2006, 27(8): 1514 (in Chinese)

[6] Kashiwabara M, Hanawa K, Asaki R, et al. Advanced AM-OLED display based on white emitter with microcavity structure. SID Digest, 2004: 1017

[7] Fisher T A, Lidzey D G, Pate M A, et al. Electroluminescence from a conjugated polymer microcavity structure. Appl Phys Lett, 1995, 67(10): 1355

[8] Droz C, Vallat-Sauvain E, Bailat J, et al. Application of Raman spectroscopy for the microstructure characterization in microcrystalline silicon solar cells. 17th European Photovoltaic Solar Energy Conference, Munich, Germany, 2001: 2917

[9] Cullity B D. Elements of X-ray diffraction. Addison-Wesley Publishing Company, Inc, 1978: 283

[10] Cao J, Jiang X Y, Zhang Z L. MoO_x modified Ag anode for top-emitting organic light-emitting devices. Appl Phys Lett, 2006, 89: 252108

[11] Zhu F, Chao G, Wei C, et al. P-nc-Si thin film materials and its application on slight crystal silicon pellicle solar battery. Photoelectron·Laser, 2004, 15(4): 381 (in Chinese)

[12] Lu M H, Weaver M S, Zhou T X, et al. High-efficiency top-emitting organic light-emitting devices. Appl Phys Lett, 2002, 81: 3921

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

[14] Dai G Z, Li H J, Pan Y Z, et al. Energy transfer probability in organic electrophosphorescence device with dopant. Chin Phys, 2005, 14(12): 2590

[15] Dodabalapur A, Rothberg L J, Miller TM, et al. Microcavity effects in organic semiconductors. Appl Phys Lett, 1994, 64: 2486

[16] Kido J, Kimura M, Nagai K. Multilayer, white light-emitting organic electroluminescent device. Science, 1995, 267: 1332