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Three-Colour Single-Mode Electroluminescence from Alq_3 Tuned by Microcavities *

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Organic metal microcavities were fabricated by using full-reflectivity aluminium film and semi-transparent silver film as cavity mirrors. Unlike conventional organic microcavities, such as the typical structure of glass/DBR/ITO/-organic layers/metal mirror, a microcavity with a shorter cavity length was obtained by using two metal mirrors, where DBR is the distributed Bragg reflector consisting of alternate quarter-wave layers of high and low refractive index materials. It is realized that red, green and blue single-mode electroluminescence (EL) from the microcavities with the structure, glass/Ag/TPD/Alq_3/Al, are electrically-driven when the thickness of the Alq_3 layer changes. Compared to a non-cavity reference sample whose EL spectrum peak is located at 520 nm with a full width at half maximum (FWHM) of 93 nm, the microcavity devices show apparent cavity effects. The EL spectra of red, green and blue microcavities are peaked at 604 nm, 540 nm and 491 nm, with FWHM of 43 nm, 38 nm and 47 nm, respectively.

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In recent years, the study of microcavity effects of luminescent materials, such as spectrum narrowing, intensity enhancement and sharply directed emission,[1−4] etc, has attracted much interests. A variety of cavity structures, including micro-rings,[5] micro-discs,[6] and Fabry–Perot planar cavities,[1−4] have been employed to build microcavities. Among these, the planar microcavity is mostly studied due to its simple structure, and it has already involved the applications of highly efficient semiconductor light-emitting diodes (LEDs)[1−4] and lasers.[7]

The planar microcavity has also been used in organic LEDs to realize an emission with a narrowed spectrum linewidth[1,3,4] and sharp direction as well as red, green and blue full-colour emission.[2] The typical structure of an organic planar microcavity is glass/DBR/organic layers/metal mirror for photoluminescence and glass/DBR/ITO/organic layers/metal mirror for electroluminescence, where DBR is the distributed Bragg reflector consisting of alternate quarter-wave layers of high and low refractive index materials. The total effective optical length (TEOL) L of a conventional organic microcavity is given by

$$L = \frac{\lambda}{2}\left(\frac{n_{\text{eff}}}{\Delta n}\right) + \sum_i n_i L_i + \left[\frac{\phi_m}{4\pi}\lambda\right], \qquad (1)$$

where λ is the free space light wavelength, n_i and L_i are the refractive index and the thickness of the organic layer, respectively, n_{eff} is the effective refractive index of the DBR, Δn is the difference of refractive indices between the materials that constitute the DBR, and ϕ_m is the phase shift at the metal reflector.[8] The first term in Eq. (1), i.e. the penetration depth in the DBR, is about λ magnitude or larger. Considering the second term, i.e. the optical length of the total organic layers, we obtain that the TEOL of an organic microcavity should be at least $3\lambda/2$. In Ref. [8], the authors have proposed an attractive idea to obtain red, green and blue emissions from a single material by combining microcavity effects and the broad luminescence spectrum of organic materials, since patterning three different materials on the scale of a typical pixel dimension (100 μm) is extremely difficult. However, the blue emission spectrum they demonstrated is not pure due to the existence of another resonant peak at 650 nm. It is believed that the existence of two peaks is attributed to the larger TEOL of the microcavity.

For a microcavity device, the resonant wavelength λ reads

$$2L\cos\theta = m\lambda \quad (m = 1, 2, 3\cdots), \qquad (2)$$

where θ is the inner off-axis angle of the resonant light wave, and m is the mode order. When θ is zero in the normal direction, Eq. (2) can be rewritten as

$$2L = m\lambda, \qquad (3)$$

then the space $\Delta\lambda$ of the cavity modes is

$$\Delta\lambda = \frac{\lambda^2}{2L}, \qquad (4)$$

where λ is the average value of the multi-resonance wavelengths. According to the formula it can be obtained that $\Delta\lambda$ is $\lambda/3$ for the microcavity with a

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length of $3\lambda/2$. If λ is set in the visible spectrum region, for example, 550 nm, the space of adjacent cavity modes will be about 180 nm. Thus, the existence of two peaks in the emission spectrum of microcavity is inevitable for $a > 180$ nm broad spectrum material. Based on this analysis, it can be found that a microcavity whose TEOL is $\lambda/2$ or λ will be optimal for realizing red, green and blue single-mode emission from a single broad spectrum material. Because the penetration depth in metal films is only about tens of nanometres[12] in the visible range, we have successfully fabricated a $\lambda/2$-length planar microcavity using two separate metal mirrors, as reported in our previous paper.[10] Therefore, it would be convenient to realize single-mode emission in a microcavity using a semi-transparent metal mirror as a substitute for the dielectric mirror.

In this study, an organic microcavity, defined between a silver mirror and an aluminium mirror, was fabricated. We used 8-hydroxyquinoline aluminium (Alq_3) as the electron-transporting and luminescence layers, and N, N'-diphenyl-N, N-bis(3-methylphenyl)1, 1'-diphenyl-4,4' diamine (TPD) as the hole-transporting layer. The silver and aluminium mirors were simultaneously used as the anode and cathode, respectively. The schematic structure of the devices is shown in Fig. 1. Three microcavity devices with the same thickness for the Ag (35 nm), TPD (50 nm) and Al (150 nm) layers were prepared in our experiment. The only difference among the different devices is the thickness of the Alq_3 layer, which was 35 nm, 60 nm and 85 nm for the red, green and blue devices, respectively.

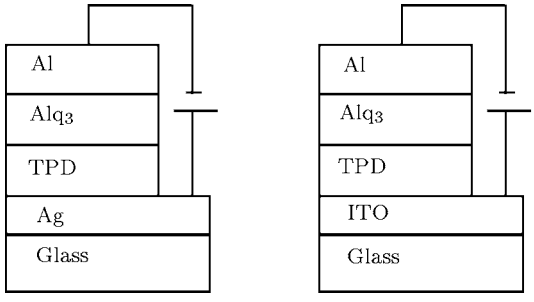

Fig. 1. Configuration of the microcavity and non-cavity devices.

The microcavity devices were first fabricated by depositing a 30 nm thick silver film on to cleaned glass substrates, followed by deposition of variously thick Alq_3 films for different devices, and then a TPD layer with the same thickness. All the devices were finished with the deposition of an aluminium film. A reference sample was simultaneously prepared, whose anode was not the silver layer but a transparent ITO one. The two metal mirrors and the organic films were deposited by the thermal evaporation method with the deposition rates of 2 nm/s for the silver and aluminium layers, and 0.1 nm/s for the organic layers, respectively. The thickness of the deposited films and the deposition rate were monitored with a quartz crystal thickness monitor placed near the substrate. All the electroluminescence (EL) spectra were measured in the normal direction of the devices with a HITACHI F-4500 fluorescence spectrophotometer

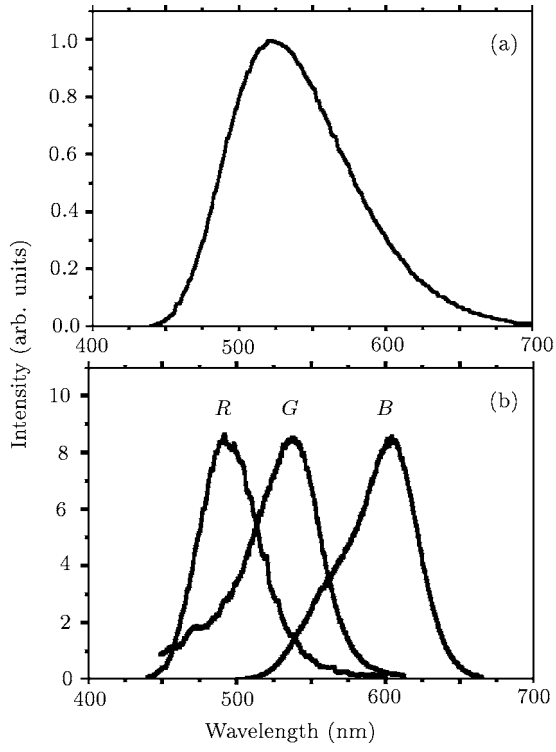

Fig. 2. EL spectra of (a) the non-cavity sample and (b) the red, green and blue microcavities.

Red, green and blue single-mode EL were obtained from the three microcavity devices. The red, green and blue emission peaks were located at 604 nm, 540 nm and 491 nm, whose full widths at half maximum (FWHM) were 43 nm, 38 nm, and 47 nm, respectively. The EL spectra of three microcavities and the reference sample are shown in Fig. 2, where it can be found that the microcavity shows apparent cavity effects. The EL characteristics of microcavity and non-cavity reference samples are given in Table 1, from which we can find that the red and blue emissions have a larger FWHM than the green emission. We attribute the broader blue emission to the lower reflectivity of the 30 nm silver film in the blue region. Figure 3 shows the calculated reflectivity of a 30 nm thick silver mirror. For the thicker aluminium mirror, the reflectivity is almost equal in the visible range.[13] It is shown in Fig. 2 that the broader red emission is due to some short wavelength light signals. This can be realized by the fact that, from Eq. (2), the resonant mode has a blueshift with the increasing angle θ deviating from the normal direction. Thus, the resonant mode of a microcavity device will have larger

angular dependence of the emission if it lies in the long-wavelength region of a natural emission spectrum. That is, the resonant mode will not exist without a larger angular dependence. Additionally, the off-axis resonant mode intensity may be larger than the normal one when the mode is positioned at the long-wavelength range of the Alq_3 natural spectrum.[11] In this case, the emission area of the microcavities is ($4 \times 4\,mm^2$). The detected distance between the device and the receiving slit was about 40 mm. No diaphragm was used. Therefore, some emission light resonating at larger off-axes angle may be collected by the detector. Compared to some reported results,[2,9] all the microcavity emission spectra are broader. This is attributed to the lower reflectivity of metal cavity mirrors.

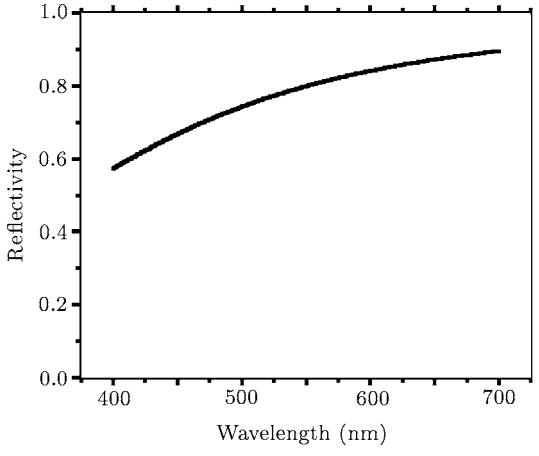

Fig. 3. Calculated reflectivity of the 30 nm silver film in the visible light range.

Table 1. Comparison of the EL spectral characteristics of microcavity and non-cavity devices.

	Peak (nm)	FWHM (nm)
Non-cavity	520	93
R microcavity	491	43
G microcavity	540	38
B microcavity	604	47

From the application point of view, the metal microcavity is not very suitable due to limited reflectivity and the intense absorption of the metal film. If the refractive index difference (Δn) of the two materials constituting the DBR is large enough for $n_{\rm eff}/\Delta n < 2$, according to Eq. (1), the penetration depth in the DBR will be small enough to make a λ-length cavity. Such a DBR will enable us not only to realize the purpose of this Letter but also to optimize for applications.

The red, green and blue emission wavelengths used for display are normally set near 460 nm, 540 nm, and 630 nm, respectively.[8] Compared with these values, in our experiment the red emission wavelength is shorter and the blue emission is longer. The FWHM of the Alq_3 emission spectra reported in the literature[8,9] is broader than that of Alq_3 used in our experiment. This type of Alq_3 or any other material whose emission spectrum is broader will be preferable for this use.

In conclusion, the three-colour single-mode EL from one broad spectrum material, Alq_3, has been obtained by using a microcavity defined between two metal mirrors. Compared with a non-cavity reference sample the microcavity devices show apparent cavity effects. The EL spectra of red, green and blue microcavities are peaked at 604 nm, 540 nm, and 491 nm, with FWHM of 43 nm, 38 nm and 47 nm, respectively.

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