

Color-tunable triple-layer electroluminescent devices based on organic salt

Xueyuan Feng (冯雪元), Jiayu Zhang (张家雨), Chunxiang Xu (徐春祥), Chang Liu (刘畅),
Guangcai Lü (吕广才), Zhenning Tang (唐振宁), and Yiping Cui (崔一平)

Advanced Photonics Center, Southeast University, Nanjing 210096

A triple-layer light-emitting diode based on an organic salt ASPT (trans-4-[p-[N-methyl-N-(hydroxyethyl)amino]styryl]-N-methylpyridinium tetraphenylborate), in which TPD and Alq₃ were employed as hole and electron transporting materials respectively, exhibits variable electroluminescence (EL) spectra under different applied voltage. At lower voltage, the EL spectrum peaks at 560 nm, which emanates from the TPD/ASPT interface; when the voltage is further increased, the peak at 610 nm, which is originated from ASPT, increases; at higher applied voltage, the device yields green light with a peak at 530 nm and a shoulder at 610 nm. The stronger emission peaking at 530 nm stems from the Alq₃. It reveals the hole-electron recombination zone depends on the applied voltage, so the color-variable EL can be observed by adjusting the applied voltage.

OCIS codes: 250.0250, 310.3840, 310.6870.

Organic electroluminescence (EL) has been paid considerable attention due to its many advantages such as high luminescence, wide ranging angle, high contrast, quick response, and low drive voltage, and the attractive application in flat-panel displays. For most organic devices, the color of the emitted light is fixed once the device is fabricated. In recent years, devices that can control their emission color by applied voltage have been studied^[1-13]. The voltage controlled monolayer structure is the most common approach to color-variable devices. A blend of organic materials, either polymers or small molecules or both, is often employed as the emitting layer^[1-5]. Due to the phase separation or energy transfer phenomena in the devices, different components in the blend exhibit different colors of light simultaneously with the intensity of each component varying with the applied voltage. Similar voltage-dependent EL has been observed in the devices using conducting polymer-doping fluorescent dye as the emitting layer. On the other hand, the change of the EL spectra with applied voltage has also been observed in multi-layer structures devices in separate layer configurations^[6-13]. The phenomena were due to the change of hole-electron recombination zone when the applied voltage. Here we report the fabrication and study of triple-layer organic light-emitting device (LED), composed of small-molecule materials. Voltage-dependent multicolor emission was observed.

In this study, TPD, Alq₃, and an organic dye salt ASPT, whose molecular structures are presented in Fig. 1, are used as hole transport, electron transport, and emitting material, respectively, to form a stable organic EL device with the structure: indium-tin-oxide (ITO)/TPD(40 nm)/ASPT(50 nm)/Alq₃(30 nm)/Al. The organic layers were sequentially deposited by the conventional vacuum evaporation method onto an ITO-coated glass substrate at room temperature. Finally, aluminum (Al) as cathode was evaporated on the top of the organic materials and the active area of the device was about 5 mm². The EL spectra were taken using an ARC SpectraPro 300i monochromator/spectrograph. Meanwhile, the current density was recorded with a digital multimeter. All the device-testing procedures were performed at room temperature.

The EL peak of the single-layer device (ITO/ASPT/Al) was centered at $\lambda = 610$ nm, whose color of the emitting-light is orange-red with the full-width at half-maximum (FWHM) of about 160 nm, as shown in the inset of Fig. 2. Threshold voltage for the detection of light emission was 5 V. The device showed high thermal stability because of the ionic interaction within the organic salt molecules and high quantum efficiency owing to the formation of dipole moments in the ASPT layer. We have assumed a physical model to interpret the results previously^[14,15]. The ions re-arranged under the electric field and caused the dipoles whose moments aligned along the electric field and were reverse to the external electric field. Therefore, space charges appear near the interfaces between the organic salt layer and the electrodes and thus cause the bend of the energy band. It is assumed that the injection barriers of hole from anode and electron from cathode were reduced because of existence of space charge.

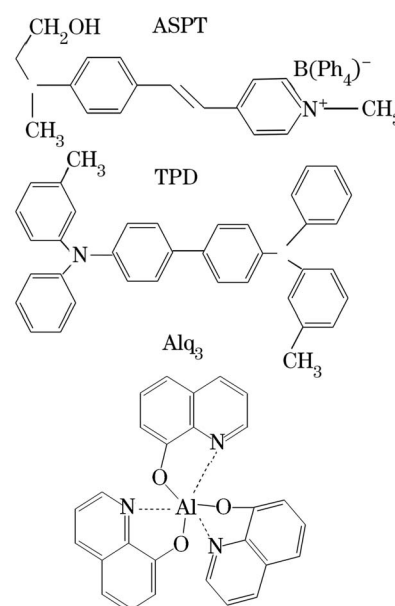


Fig. 1. The molecular structures of ASPT, TPD, and Alq₃.

Figure 2 shows the EL spectra of the three-layer device with different applied voltage. It should be noted that yellow emission centered at 560 nm instead of organic-red light is observed only when the applied voltage is relatively low. The new component (560 nm) does not belong to any of the individual material, and this suggests that the emission comes from the interface between TPD and ASPT. The shape of the EL spectrum of three-layer device is significantly different from that of monolayer device, indicating the difference between emission from TPD/ASPT interface and that from bulk ASPT. With increasing the applied voltage, the emission color tends to become orange-red to the eye, and the EL peak at 610 nm, which originated from ASPT layer, becomes dominant over that at 560 nm. When the voltage is high enough, the EL peak at 530 nm, which stems from Alq₃, becomes dominant over those at 560 and 610 nm, and then the light is of green color. As shown in Fig. 2, the three-layer EL device does not show any emission in the blue characteristic of TPD.

These results of the EL spectra change from the multi-layer structure can be explained using the energy-band diagram of the layered materials as shown in Fig. 3. It is well known that the generation of light is a consequence of the recombination of holes and electrons injected from the electrodes, respectively. The dependence of display color on the applied voltage as shown in Fig. 2 results from the variation of the location and width of the recombination zone. For the lower applied voltage, the recombination zone mainly takes place in the TPD/ASPT interface. With the increase of applied voltage, it can be seen that emission from the ASPT layer is gradually enhanced, and thus it is proposed that the recombination zone in the triple-layer device is extended towards the emitting layer and the amount of hole-electron pairs recombined inside the ASPT layer is increased. The extension of the recombination zone results in the red-shift in the right half (longer wavelength) of the EL spectrum. At a bias of 8 V, as shown in Fig. 2, the EL spectrum is dominated by ASPT layer. The main EL peak shifts to 610 nm, though the intensity at 560 nm is still strong. At higher applied bias, the recombination region extends further and moves into Alq₃ layer. So there is a clear blue-shift in the left half (shorter wavelength) of the spectrum. At a bias of 10 V, the EL spectrum is dominated by Alq₃ layer. The detail

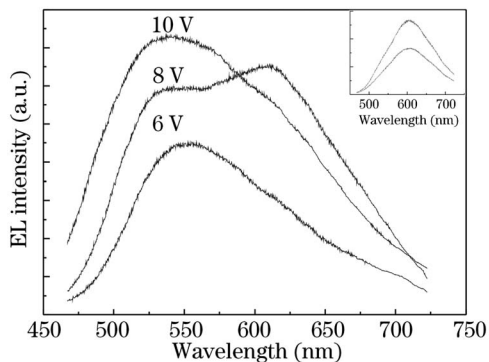


Fig. 2. Applied voltage-dependent EL spectra of the triple-layer EL device. The inset shows the EL spectra of the single-layer device (ITO/ASPT(50 nm)/Al) at 6 and 8 V.

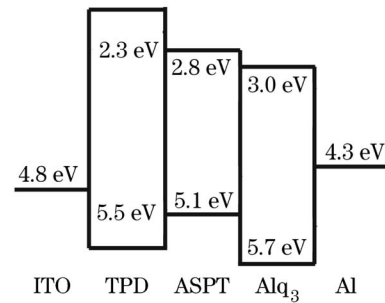


Fig. 3. Schematic energy-band diagram of the triple-layer EL device.

mechanism is under further study. We believe that the different field-dependent mobility of the materials makes the emission zone be voltage-dependent^[6].

To summarize, a triple-layer EL device of ITO/TPD(40 nm)/ASPT(50 nm)/Alq₃(30 nm)/Al has been fabricated by the conventional vacuum evaporation method, and its display color varies with the applied voltage. The EL spectra were measured to investigate the emission characteristics of the device. The phenomena are attributed to the variation of the location and width of the recombination zone. This technique provides a simple way to fabricate color-tunable EL displays.

This work was supported by the Jiangsu High-Technology Program (No. BG2003032) and National Natural Science Foundation of China (No. 10474010 and 60125513). Y. Cui is the author to whom the correspondence should be addressed (e-mail: cyp@sue.edu.cn).

References

1. E. J. W. List, S. Tasch, C. Hochfilzer, G. Leising, P. Schlichting, U. Rohr, Y. Geerts, U. Scherf, and K. Mullen, *Opt. Mater.* **9**, 183 (1998).
2. C. X. Xu, Y. P. Cui, Y. Z. Shen, H. W. Gu, Y. Pan, and Y. K. Li, *Appl. Phys. Lett.* **75**, 1927 (1999).
3. J. Kim, B. Seo, and H. Gu, *Synth. Met.* **132**, 285 (2003).
4. M. Uchida, Y. Ohmori, T. Noguchi, T. Ohnishi, and K. Yoshino, *Jpn. J. Appl. Phys.* **32**, L921 (1993).
5. A. Bolognesi, C. Botta, L. Cecchinato, V. Fattori, and M. Cocchi, *Synth. Met.* **106**, 183 (1999).
6. T. J. Chow, S. Tsai, C. Chiu, and T. Yeh, *Synth. Met.* **149**, 59 (2005).
7. J. Kalinowski, P. Di Marco, V. Fattori, L. Giulietti, and M. Cocchi, *J. Appl. Phys.* **83**, 4242 (1998).
8. L. Ding and F. E. Karasz, *J. Appl. Phys.* **96**, 2272 (2004).
9. J. Kalinowski, P. Di Marco, M. Cocchi, V. Fattori, N. Camaioni, and J. Duff, *Appl. Phys. Lett.* **68**, 2317 (1996).
10. C. Qiu, H. Chen, M. Wong, and H. S. Kwok, *Synth. Met.* **140**, 101 (2004).
11. Y. Z. Wang, R. G. Sun, D. K. Wang, T. M. Swager, and A. J. Epstein, *Appl. Phys. Lett.* **74**, 2593 (1999).
12. Y. Z. Wang, R. G. Sun, F. Meghdadi, G. Leising, and A. J. Epstein, *Appl. Phys. Lett.* **74**, 3613 (1999).
13. J. Lee, S. Kim, S. Ju, W. Lee, J. Choi, Y. Kim, and W. Y. Kim, *Synth. Met.* **111–112**, 63 (2000).
14. H. G. Xu, R. P. Meng, C. X. Xu, J. X. Zhang, G. H. He, and Y. P. Cui, *Appl. Phys. Lett.* **83**, 1020 (2003).
15. R. P. Meng, H. G. Xu, C. X. Xu, J. X. Zhang, G. H. He, and Y. P. Cui, *Chin. Phys. Lett.* **20**, 935 (2003).