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# Three-layered multicolor organic electroluminescent device

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We have realized a three-layered multicolor organic electroluminescent device utilizing poly (9,9-dihexylfluorene) and perylene derivatives as emission layers, with diamine derivatives as an electron blocking layer. Emission colors could be selected either blue or red by changing the polarity of the applied voltage. By driving with an ac biased electric field, both emission colors were observed and the intensity ratio of each emission color was modulated by the frequency of the applied ac field. © 1996 American Institute of Physics. [S0003-6951(96)00432-9]

Recently, organic electroluminescent (EL) devices<sup>1-9</sup> have attracted great interest because of their great advantage for visible emission and potential for large-area flat panel display devices driven at low voltage. Since Tang and VanSlyke have demonstrated a high efficient EL diode consisting of light-emitting layers and carrier transport layers,<sup>1</sup> many kinds of conducting polymers and fluorescent dyes have been developed in order to obtain high efficiency, long lifetime, and white-color emission.<sup>10</sup> We have already reported color-variable EL diodes,<sup>11</sup> in which the emission color has changed with increasing applied dc field.

In this letter, we report a three-layered multicolor EL device utilizing poly (9,9-dihexylfluorene) (PDHF), N, N'-Bis (2,5-d-tert-butylphenyl)-3, 4, 9, 10-perylenedicarboximide (BPPC) and N,N'-diphenyl-N, N'-(3-methylphenyl)-1, 1'-biphenyl-4, 4'-diamine (TPD). In the device, we have realized switching of the emission color by the polarity of the applied dc field and modulation of the emission color by the frequency of the applied ac field.<sup>12</sup> Mechanisms of color change in the EL device are discussed in view of carrier transport properties.

Figure 1 shows a schematic description of the multicolor EL device and molecular structures of organic materials used in this study. We have already reported results from a vapor-deposited PDHF double-heterostructured EL diode, which exhibited blue-green EL emission.<sup>13</sup> TPD is mainly used as a hole transport and electron blocking material, and BPPC is one of the materials for red EL emission. The device consists of an indium-tin-oxide (ITO)-coated glass substrate/BPPC emission layer (30 nm)/TPD electron blocking layer (40 nm)/PDHF emission layer (30 nm)/an aluminum (Al) electrode. Organic thin films were fabricated by organic molecular beam deposition onto ITO-coated glass substrates. The deposition of the organic material was done at a background pressure of  $10^{-4}$ – $10^{-5}$  Pa in the deposition chamber. Organic materials were loaded into separate Knudsen cells and subsequently heated up to sublimation temperature, and then deposited onto the substrate. The thickness of the deposited film was determined by an oscillating quartz thickness monitor. Finally, aluminum was vapor deposited at  $10^{-4}$  Pa onto the film. Total thickness of the organic film was 100 nm and the active device area was 0.04 cm<sup>2</sup>. All measurements were carried out at 77 K.

Current (*I*)-voltage (*V*) and EL intensity (*L*)-*V* characteristics are shown in Fig. 2. Positive biased condition is

defined as the case in which the ITO electrode is biased positively against the Al electrode. In both positive and negative biased conditions, the current increased superlinearly with increasing applied voltage, and light emission was observed at about 17 V in the positive biased condition. It should be emphasized here that light output was also observed over -16 V in the negative biased condition. *I*-*V* and *L*-*V* curves exhibited nearly symmetrical behavior in this EL device.

EL spectra of this EL device driven in positive and negative biased condition are shown in the inset of Fig. 3. In the case of positive biased condition, blue light originating from PDHF was obtained from the device, with a maximum peak at 470 nm. On the other hand, in the negative biased condition, red light from the BPPC layer, whose peak was existed at 640 nm, was observed. That is, the device could emit either blue or red light by the selection of the polarity of the applied field. In the positive biased condition, the dips of emission at 500 and 530 nm correspond to the absorption by BPPC. This result indicates that the blue emission occurred at the interface of PDHF layer and Al electrode, and passed through the BPPC layers.

The energy band diagram of the device is schematically shown in Fig. 4. In the positive biased condition, holes are injected from ITO into the highest occupied molecular orbital (HOMO) state of BPPC over the injection barrier (1.3 eV) at the ITO/BPPC interface and electrons are injected from the Al electrode into the lowest unoccupied molecular orbital (LUMO) state of PDHF over the injection barrier (1.7

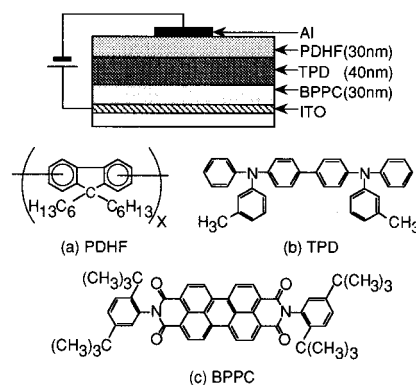


FIG. 1. Schematic description of three-layered EL device and molecular structures of organic materials used in each layer.

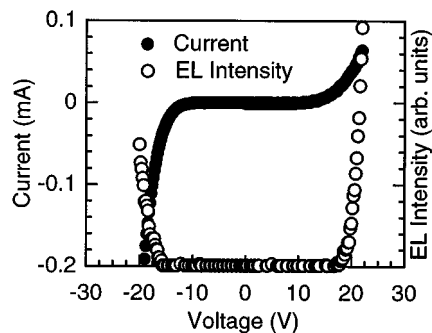


FIG. 2. Current-voltage and EL intensity-voltage characteristics of the three-layered EL device.

eV) at the Al/PDHF interface. Injected holes migrate in the BPPC and TPD layers until reaching the PDHF layer. Most electrons are localized in the PDHF layer and could not move into the BPPC layer through the TPD layer, because both PDHF and TPD are hole transport materials and a substantial barrier (0.4 eV) for electrons exists at the PDHF/TPD interface. Accordingly, formation and radiative recombination of excitons occur in the PDHF layer, which is a reason why the blue light is observed in the positive biased condition.

In the case of the negative biased condition, holes are injected from Al to the HOMO state of PDHF and electrons are injected from ITO to the LUMO state of BPPC over the injection barriers of 1.4 eV and Al/PDHF interface and 0.7 eV at the ITO/BPPC interface, respectively. The barrier height for electron injection in the negative biased condition is lower than that in the positive biased condition. Therefore, the current in negative biased case is larger at the same applied voltage than that in the positive biased case. For example, the current at  $-20$  V is seven times as large as that at  $20$  V. Holes injected into the HOMO level of PDHF migrate in the PDHF and TPD layers and reach the BPPC layer. Electrons injected from the ITO into the LUMO state of BPPC are confined in the BPPC layer, since the barrier at the BPPC/TPD interface (1.7 eV) is too high to move into the TPD layer. Consequently, formed excitons are confined in the BPPC layer and EL emission takes place in the BPPC layer, which is a reason why red light is observed in the negative biased condition. It is an important conclusion that the emission occurred from the layer near the electron inject-

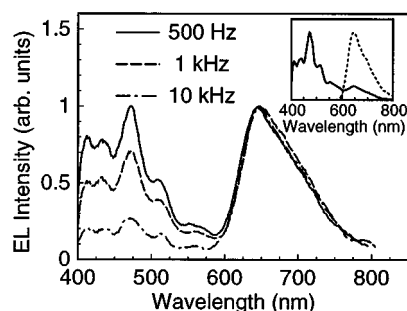


FIG. 3. EL spectra as a function of frequency of applied ac field. These spectra are normalized to unity at 640 nm. Inset: EL spectra of three-layered EL device in positive bias condition ( $+21$  V) (solid line), and in the negative one ( $-18$  V) (dashed line).

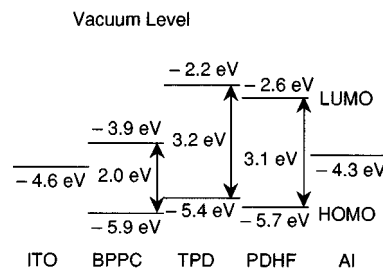


FIG. 4. Schematic energy band diagram of the three-layered EL device.

ing electrode. The TPD layer plays a role of an electron blocking layer in the three-layered multicolor EL device. The introduction of the electron blocking layer between the two emission layers makes it possible to change the emission region to the other side of the electron blocking layer when the polarity of applied voltage is reversed. Consequently, the alternative of two emission colors may be determined by the polarity of the applied field.

Figure 3 shows the EL spectra under ac biased condition as a function of the frequency of the applied ac field. These spectra are normalized by the BPPC peak intensity at 640 nm. The applied field waveform is an ac rectangular-wave (voltage pulses of  $+21$  and  $-18$  V in height and 50% duty cycle). Under an ac biased condition of 500 Hz, both blue and red emission were observed. Upon applying ac fields of higher frequency, however, the blue light originating from PDHF was suppressed compared with the red emission from BPPC. That is, the emission color can be gradually modulated by sweeping only the frequency of the applied ac field, while maintaining a constant pulse height.

Figure 5 shows the pulse response waveform of the EL emission, when driven with an ac rectangular-wave field at 1 and 10 kHz. As is evident in Fig. 5, when driven with ac field at 10 kHz, the blue light intensity in the positive biased condition was suppressed strongly in comparison with the red light in the negative one. The response time in the positive biased condition was estimated to be  $150 \mu\text{s}$  and that in the negative one, on the contrary, was within  $20 \mu\text{s}$ . The pulse response of the blue emission was so slow that the blue emission could not follow the high frequency of the ac field. This is why, when driven with a high-frequency ac field, the blue light is quenched strongly. As a result, the EL spectrum can be changed by varying the frequency of the applied ac field.

In conclusion, a three-layered multicolor EL device has

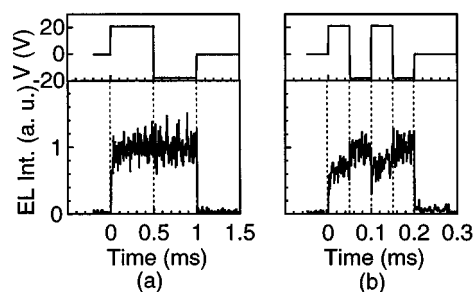


FIG. 5. Pulse response waveforms of EL emissions driven with ac biased field of (a) 1 kHz and (b) 10 kHz.

been successfully realized utilizing PDHF, TPD, and BPPC, in which the TPD layer functions as an electron blocking layer between separate PDHF and BPPC emission layers. The emission color can be either blue or red depending on the polarity of the applied field. Blue light is obtained with positive bias, and red light with negative bias. The device, furthermore, can be driven with an ac field, and the emission color can be gradually modulated by changing the frequency of applied AC field.

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