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AC-driven, color- and brightness-tunable organic light-emitting diodes constructed from an electron only device



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ABSTRACT

In this paper, a color- and brightness-tunable organic light-emitting diode (OLED) is reported. This OLED was realized by inserting a charge generation layer into an electron only device to form an n-i-p-i-n structure. It is shown that, by changing the polarity of applied voltage, only the p-i-n junction operated under positive bias can emit light and, by applying an AC voltage, emission from both junctions was realized. It is also shown that, by using a combination of blue- and red-emiting layers in two p-i-n junctions, both the color and brightness of the resulting white OLED can be tuned independently by changing the positive and negative amplitudes of the AC voltage.

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1. Introduction

Fundamentally, light emission from an organic lightemitting diode (OLED) [1] results from the electron/hole recombination in the emissive layer (EML), which requires simultaneous hole and electron injection from the anode and the cathode, and then transport through the hole transporting layer (HTL) and the electron transporting layer (ETL), respectively, as shown in Fig. 1a. However, in a single carrier device [2], as only one charge carrier is injected into the device, light emission is impossible. For example, Fig. 1b shows an electron only device (EOD), which can be considered as an n-i-n structure. The low injection barrier for electrons and high injection barrier for holes allow only the electrons to be injected from the electrode with a negative bias across the device while the holes are blocked at the other electrode. Therefore, the electron/hole recombination is internally not possible. Single carrier devices as such were usually used to determine electrical transporting parameters [2–9], such as mobility, carrier density and trap levels. It would be very interesting if one can convert an EOD to a light-emitting device. One



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Fig. 1. Schematic energy level diagrams of a typical OLED (a), electron only device (b) and electron only device with a hole source (c).

possible way is to incorporate a hole source at one side of the EML, as shown in Fig. 1c.

Fortunately, a charge generation layer (CGL), which is generally used in tandem OLEDs [10-30], can be used to create extra charge carriers. To date, many CGL structures have been proposed, such as n-doped ETL/p-doped HTL (e.g., Alq₃:Li/NPB:FeCl₃) [10], organic p/n junction (e.g., Pentacene/C60) [23,24,26,27] and n-doped ETL/electron acceptor/electron donor structure (e.g., BCP:Li/MoO₃/NPB [21], Bphen:Li/HAT-CN/NPB [18]). It was previously shown that the high work function (WF) of transition metal oxides (TMOs) [31-40], such as WO₃ (WF = 6.78 eV) and MoO₃ (WF = 6.83 eV), plays an important role in the charge generation process. Many electronic structure [32-34,37,38,40] and device physics studies suggest that charge generation originates from the electron transfer from the hole transporting material to TMOs (e.g., in Bphen:Cs/MoO₃/NPB CGL, electron transfers from NPB to MoO₃), which is the core of CGL. It is thus anticipated that the incorporation of CGL in a single carrier device could enable the dual carrier injection. Qi et al. showed that a BCP:Li/MoO₃ CGL is capable of injecting both holes (device structure: Al/BCP:Li/MoO₃/NPD/MoO₃/Al) and electrons (device structure: Al/BCP/BCP:Li/MoO₃/Al) by means of charge generation [41]. Lee et al. also employed a p-doped HTL/n-doped ETL type CGL (CuPc:ReO₃/Bphen:Rb₂CO₃) as an electron injection layer in an inverted OLED structure for a p-n-i-p structure [42], which can inject electrons well and is independent of the choice of the electrode.

In this paper, we investigated the effect of a TMO (MoO₃)-based charge generation layer in an EOD and successfully converted the EOD to a light-emitting device (from n-i-n structure to n-i-p-i-n structure). Due to the symmetrical energy band diagram design, our device can be operated to emit light by both positive and negative bias. By employing a blue dye in one n-i-p junction and a red dye in another n-i-p junction, the device can emit either blue or red color, depending on the polarity of the bias. We also found that, by applying an alternating current

(AC) signal, alternating blue and red emission was observed. By changing the relative positive and negative amplitudes of the AC voltage, various white colors with tunable brightness were realized, which is impossible for a conventional OLED.

2. Experimental

2.1. Device fabrication

All devices were fabricated on commercial ITO-coated glass substrates. The ITO substrates were treated in order by ultrasonic bath sonication of detergent, de-ionized water, isopropanol and acetone, each with a 20 min interval. Then the ITO substrates were dried with nitrogen gas and baked in an oven at 80 °C for 30 min. Subsequently, the substrates were transferred into a thermal evaporator. where the organic, inorganic and metal functional layers were grown layer by layer at a base pressure better than 4×10^{-4} Pa. The evaporation rates were monitored with several guartz crystal microbalances located above the crucibles and thermal boats. For organic semiconductors, metal oxides and calcium, the typical evaporation rates were about 0.1 nm/s and for aluminum, the evaporation rate was about 1–5 nm/s. For the dyes, typical evaporation rates were 0.01 nm/s, as reported in the literature [43,44]. The intersection of Al and ITO formed a $3 \text{ mm} \times 3 \text{ mm}$ active device area.

2.2. Device characterization

Current density-voltage (J-V) and luminance-voltage (L-V) data were collected with a source meter (Yokogawa GS610) and a luminance meter (Konica Minolta LS-110) with a customized Labview program. Electroluminance (EL) spectra were recorded with a spectrometer (Photo Research PR705). The AC voltage used to drive the device for EL spectrum measurement was generated by the same sourcemeter (Yokogawa GS610) operating in the pulse mode (rectangular shape, 50% duty cycle, 50 Hz). For EL transient study, a function generator (Tabor Electronics



Fig. 2. Device structures for EOD w/o (left) and w/ (right) MoO₃ layer.

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