



In-plane electroluminescence from microcavity organic light-emitting transistors



Yongsheng Hu, Li Song, Dongwei Li, Jie Lin, Xingyuan Liu*

State Key Laboratory of Luminescence and Applications, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun 130033, China

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ABSTRACT

We report the observation of in-plane emission beneath the drain electrode in multilayer heterostructure organic light-emitting transistors (OLETs). A novel modification method for the interface between the hole transport layer and the emission layer has been proposed, which brought a great enhancement for the light power and external quantum efficiency. Further, distributed Bragg reflector was incorporated to the in-plane-emitted OLETs, which combined with the top thin layer of Au, forming a vertical microcavity. The electroluminescence spectra were significantly altered by the microcavity and much narrower linewidth was obtained. The results will help to develop high color purity and white OLETs with high performance, which would be useful for multifunctional displays.

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1. Introduction

Organic light-emitting transistors (OLETs) are a novel kind of multifunctional optoelectronic devices which combined the electroluminescence (EL) characteristic of organic light-emitting diodes (OLEDs) and the switching characteristic of organic field-effect transistors [1,2]. The multifunctional property ensures them wide applications such as flat panel displays and optical communication systems, since much simpler process and higher degree of integration could be achieved [3,4].

In an OLET, the location of the emission zone is greatly determined by the device structure and the relative transportation ability between holes and electrons. Generally, the emission zone lies within the channel and presents as a line-shape with a width between 2 and 10 μm [5,6]. The location and width of the emission zone is usually gate voltage (V_{GS}) depended for devices with ambipolar characteristic. For OLETs with unipolar characteristic, the emission zone is likely to be restricted to the vicinity of the electrode where the minority carriers are injected [4,7]. Recently, Toffanin et al. observed in-plane emission both in the channel and beneath the electrode in trilayer heterostructure OLETs, which overcomes the long-existed drawback of line-shaped emission which restricts the aperture ratio for OLETs-based displays [6,8]. Ullah et al. achieved an intense in-plane emission beneath the

electrode in double layer heterostructure OLETs by utilizing a non-planar asymmetrical electrodes technology [3]. The device also presented impressive performance which manifests the advantages of heterostructure OLETs for the use of displays.

Nevertheless, when incorporating carrier transport layer with high carrier mobility and emission layer (EML) with high photoluminescence efficiency simultaneously, there would probably be energy-misalignment and thus great carrier injection barriers are formed between the two layers, which would result in low efficiency for exciton formation and finally restrict the further promotion for the performance of the devices [9–11]. Interfacial modification is an effective way to minimize the injection barriers. However, large amount of studies have been focused on the interfaces of the electrode/organic layer and the insulator/organic layer [10,12–14]. To the best of our knowledge, there is little report on the interface modification between organic/organic layers in OLETs. Transition metal oxide, i.e. MoO_x , is a common kind of modification layer, which is used between the electrode and organic layer to promote the hole injection [7,15]. In our work, we tried to incorporate MoO_x between the organic layers, and found that it brought a great enhancement for the emissive characteristic of the devices.

Microcavity formed by distributed Bragg reflector (DBR)/DBR or DBR/metal reflectors is able to manipulate the distribution of optical field in the cavity. It has played an important role in the application of adjusting the EL properties of OLEDs and providing an optical resonator for organic lasers [16–19]. Namdas et al. has

* Corresponding author.

E-mail address: liuxy@ciomp.ac.cn (X. Liu).

observed enhanced performance of OLETs by using DBR as the bottom reflector, which was composed of 3 pairs of $\text{SiN}_x/\text{SiO}_2$ [20]. However, as the insulator layer simultaneously, the DBR used had only a limited pairs and a low reflectance. In addition, one DBR could not make an effective microcavity structure that ideal cavity effect required.

In this work, we report the observation of in-plane EL emission beneath the drain electrode in multilayer heterostructure OLETs. Ultra-thin MoO_x layer was used to modify the interface between the hole transport layer (HTL) and the EML, which brought a great enhancement for the light power and external quantum efficiency (EQE). By incorporating high reflective bottom DBR, we constructed microcavity OLETs with in-plane narrow band light emission due to microcavity effect. The results are beneficial to the development of high color purity and white OLETs with high performance for multifunctional displays.

2. Experimental details

2.1. Materials and device preparation

Pentacene, CBP, $\text{Ir}(\text{piq})_2\text{acac}$ and TPBI were purchased from Lumtech. PTCDI-C13, polystyrene (PS), Poly-4-vinylpenol (PVP) and MoO_x were purchased from Sigma-Aldrich. 66 nm TiO_2 , 106 nm SiO_2 and 78 nm ITO were deposited onto the glass substrate by e-beam evaporation under a base pressure of 1.5×10^{-4} Pa, evaporation rate of 2 Å/s, and substrate temperature of 300 °C. An end-Hall ion source was used to assist the deposition. PVP and PS were spun coated in the way reported elsewhere [21]. Pentacene, MoO_x , $\text{CBP}:\text{Ir}(\text{piq})_2\text{acac}$ (~6 Wt%), TPBI and PTCDI-C13 were successively thermal evaporated with the rate of 0.2, 0.05, 2, 0.2 and 0.5 Å/s, respectively. Au was evaporated on top of PTCDI-C13 through a shadow mask with channel length and width of 60 μm and 3000 μm , respectively, which acted as the source and drain electrodes. The substrate was kept at room temperature during the deposition process. The devices were encapsulated with UV glue in the glovebox (H_2O , $\text{O}_2 < 0.1$ ppm) before testing.

2.2. Characterization

The electrical characteristics were performed by Keithley 4200 SCS at room temperature under air ambient. The photocurrent was recorded by HAMAMATSU S1336 photodiode. The channel images were captured by Olympus BX51TRF CCD microscope with a 20 \times objective lens. The EL spectra were recorded by AvaSpec-ULS2048L fiber spectrometer. The absorption spectra were recorded by Shimadzu UV-3101PC UV-vis-NIR spectrophotometer. The carrier mobilities were calculated by the formula for the saturation regime: $I_{\text{DS}} = \mu C_i (W/2L)(V_{\text{GS}} - V_T)^2$, (where μ is the field-effect mobility, C_i is the gate dielectric capacitance density, V_T is the threshold voltage, V_{GS} is the gate-source voltage, and W and L are the channel width and length, respectively). The EQE was calculated as follows:

$$\text{EQE} = \frac{P_{\text{tot}}/E_{\text{ph}}}{I/q} = \frac{P_{\text{tot}}(W)}{I(A)E_{\text{ph}}(\text{eV})} = \frac{I_{\text{ph}}/K(A/W)}{I_{\text{DS}}E_{\text{ph}}(\text{eV})} \approx 1.5 \frac{I_{\text{ph}}}{I_{\text{DS}}} \quad (1)$$

where P_{tot} , K , E_{ph} , I_{ph} , I_{DS} and q are the total emitted light power, photo sensitivity of the detector (0.3 A/W), average photon energy, photocurrent, drain current and electron charge, respectively.

3. Results and discussion

Fig. 1 shows the schematic of the device, the energy levels of the materials and the molecular structures. Organic semiconductors with high carrier mobility rather than a matched energy level are

our priorities to choose materials. Pentacene and PTCDI-C13 were therefore used as the HTL and the electron transport layer (ETL), respectively, since they have shown good ambipolar transportation characteristic [22]. Highly efficient phosphorescent donor-acceptor system $\text{CBP}:\text{Ir}(\text{piq})_2\text{acac}$ was used as the EML. TPBI is a widely used ETL and hole block layer in OLEDs [23,24]. Since the LUMO level of TPBI lies between that of CBP and PTCDI-C13, it can lower the electron injection barrier between ETL and EML as a secondary ETL. According to Fig. 1(b), there is large hole injection barrier between pentacene and CBP, which will result in low efficiency for hole injection and exciton formation. Considering that MoO_x is able to function as a charge generation layer between the two subunits of a tandem OLED [25], we attempted to introduce it as a modification layer at the interface of HTL/EML, expecting for an enhancement for the performance of the devices.

We first investigated the influence of the thickness of MoO_x . Devices with different thicknesses of MoO_x were fabricated: Glass/ITO/PVP(420 nm)/PS(30 nm)/pentacene(48 nm)/ MoO_x (0 nm, 0.15 nm, 0.25 nm, 0.5 nm)/ $\text{CBP}:\text{Ir}(\text{piq})_2\text{acac}$ (24 nm)/TPBI(14 nm)/PTCDI-C13(30 nm)/Au(15 nm). We observed the light emission from the top of the devices.

Fig. 2(a) shows the transfer characteristics ($V_{\text{DS}} = -100$ V) for different devices. For device without MoO_x , ambipolar characteristic is observed, which is resulted from the carrier transport layer material system we chose. The hole and electron mobilities are $0.04 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $0.0014 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively. Fig. 2(b) and (c) show the corresponding photocurrent (I_{ph}) and EQE. The I_{ph} gets a maximum around $V_{\text{GS}} = -40$ V. The EQE also reaches to the maximum of 0.007% at this regime, where the device is under ambipolar operation. The well-balanced transportation of carriers at this regime is attributed to the maximum EQE. For device with 0.15 nm MoO_x , it can be seen from Fig. 2(a), the drain current (I_{DS}) increases significantly at the hole accumulation regime ($V_{\text{GS}} = -80 \text{ V} \sim -120 \text{ V}$), which indicates that the hole concentration is greatly increased. On the other hand, the I_{DS} is partly restricted at the electron accumulation regime ($V_{\text{GS}} = 0 \text{ V} \sim 20 \text{ V}$). The I_{ph} generally increases as the V_{GS} increases and reaches to 20 nA at $V_{\text{GS}} = -120 \text{ V}$, which is approx. 50 times higher than that without MoO_x . The EQE reaches to 0.01%, which is also promoted compared to that without MoO_x . As MoO_x grows to 0.25 nm, the I_{DS} continues to increase, which indicates the continuous increase of hole concentration. At the regime of $V_{\text{GS}} = 0 \text{ V} \sim 20 \text{ V}$, the I_{DS} is around 40 μA , which is approx. one order of magnitude higher than that without MoO_x . This implies that the I_{DS} at this regime is probably resulted from the carriers generated by MoO_x . We will stress this point later. As for the I_{ph} and EQE, there is further improvement compared to that with 0.15 nm MoO_x . The maximum I_{ph} and EQE reach to 87 nA and 0.03%, respectively, and the EQE maintains at 0.015% even for high V_{GS} (which means high current density). Although the EQE is lower than state-of-the-art for heterojunction OLETs [3,6], there is several orders of magnitude enhancement for devices based on pentacene and PTCDI-C13 [26,27]. As MoO_x increases to 0.5 nm, the I_{DS} still continues to increase, while the on/off ratio decreases greatly. This implies that MoO_x would generate more carriers, which beyond the modulation by the potential difference between the gate electrode and an opposite electrode [28]. However, the increasing of I_{ph} is slowing down, and even lower than that with 0.25 nm MoO_x at high V_{GS} . The EQE is also smaller than that with 0.25 nm MoO_x , which is probably related to the unbalanced carrier transportation and charge induced quenching of excitons due to the excess holes. Fig. 2(d) presents the output characteristic of device with 0.25 nm MoO_x , which owns the best EQE of the four devices. It is shown that the device exhibits unipolar operation characteristic.

We also observed the characteristic of emission zones for the four devices under different V_{GS} . Fig. 3 presents the images of the

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