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Influence of source/drain electrodes on external quantum efficiency of ambipolar organic light-emitting transistors

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ABSTRACT

The influence of source/drain (S/D) electrodes on the external quantum efficiency (EQE) of ambipolar organic light-emitting transistors (OLETs) based on fluorene-type polymer films is investigated. The electrical properties and the maximum EQE value of the device with indium tin oxide (ITO) S/D electrodes are almost the same as those of the device with Ag S/D electrodes. A relatively high EQE of 1% is achieved regardless of the emission site for the OLET with ITO. In contrast, the EQE of the OLET with Ag is low when the emission occurs close to the S/D electrodes. The maximum EQE of the device with Ag is obtained when the emission is observed in the middle of the channel. It is found that the exciton quenching by Ag electrodes significantly influences the low EQE of the OLET with Ag electrodes. The achievement of high EQE regardless of the emission site is attributable to both better carrier injection and lower exciton quenching at the interface of S/D electrodes for the OLET with ITO.

1. Introduction

Organic light-emitting transistors (OLETs) have attracted considerable attention because of their potential application to new optoelectronics [1–5]. OLETs are multifunctional devices that combine the light emission property of an organic light-emitting diode (OLED) with the switching property of a field-effect transistor in a single device architecture. The degree of integration can be increased by using OLETs, and optoelectronic devices can be fabricated by simple and low-cost processes owing to the reduction of the number of parts.

OLETs with various device structures have been reported to show unipolar [1,3] and ambipolar characteristics [5–18]. In unipolar OLETs, light emission is observed at the edge of the electrode and is generally inefficient because of carrier imbalance and the quenching of excitons by the electrode. Ambipolar device operation has been demonstrated using a variety of organic materials, including small molecules and conjugated polymers [19–21]. When appropriate bias voltages are applied to ambipolar OLETs, both electrons and holes are injected into an organic semiconductor from source or drain electrodes. Efficient charge recombination and light emission can be obtained at the interface between an organic layer and a gate dielectric. The emission site can be moved by varying the applied voltages and be observed directly. This allows the investigation of carrier behavior or charge recombination.

In organic semiconductors, π-conjugated polymers, being quasi-one-dimensional macromolecular electronic systems, offer a number of unique properties. In particular, poly(alkylfluorene), a liquid–crystalline semiconducting polymer, exhibits blue emission and various morphological behaviors [22–25] and has emerged as an important class of conducting polymers owing to its efficient emission, relatively high mobility, and high stability. Top-gate-type OFETs based on fluorene polymers exhibit ambipolar and light-emitting properties [7,15–18].

Some electrodes have been used for the source/drain (S/D) electrodes of ambipolar OLETs. For example, highly

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efficient OLETs with asymmetric gold (Au) and zinc oxide S/D electrodes have been achieved [26], and the difference in carrier injection between Au and silver (Ag) electrodes has been investigated [27]. However, the difference in electrical properties and external quantum efficiency (EQE) between OLETs with metal electrodes and those with oxidized electrodes has not been sufficiently investigated. In particular, Ag and indium tin oxide (ITO) are useful for the following reasons. Ag electrodes can be fabricated using Ag nano-ink, which enables the realization of all-solution-processed devices [18,28]. ITO is suitable for the electrode of light-emitting devices and is often used as the anode of OLEDs because of its optical transparency. Thus, investigating the differences in optoelectrical properties between OLETs with Ag and ITO electrodes should help to expand device applications. Furthermore, we will investigate the influence of S/D electrodes on the EQE of top-gate-type polymer light-emitting transistors.

2. Experimental procedure

ITO and Ag were used for the S/D electrodes. ITO electrodes were patterned by photolithography on a substrate. Ag electrodes were vacuum-deposited onto a substrate at a background pressure of about 10^{-4} Pa. The channel length and width were 0.1 and 2 mm, respectively. A polyfluorene-based block copolymer, poly[(9,9-dioctylfluorene)-co-(benzothiadiazole)] (F8BT), was used as the semiconducting layer. The F8BT layer was formed by spin-coating from a xylene solution onto each substrate and baked at 200 °C in a dry nitrogen glove box. The typical thickness of the semiconducting layer was approximately 60 nm. Poly(methyl methacrylate) (PMMA), which does not contain electron trapping groups, such as OH, was used as the gate insulator. Using anhydrous *n*-butyl acetate as a solvent, a PMMA solution was spun onto an active layer and annealed at 150 °C. The PMMA gate insulator was formed on the active layer without intermixing. The typical thickness of the gate insulator was approximately 550 nm. A 50-nm-thick gate electrode of Ag was vacuum-deposited onto the polymer gate insulating layer which was formed on the semiconducting layer at a background pressure of about 10^{-4} Pa. We fabricated an OLET with ITO S/D electrodes (device A), an OLET with Ag electrodes (device B) and an OLET with asymmetric S/D electrodes, i.e., both ITO and Ag electrodes (device C). The device structure and the energy diagram of the OLETs are illustrated in Fig. 1.

All measurements of the characteristics of the OLETs were carried out in a vacuum chamber at a background pressure of about 10^{-4} Pa. The current–voltage characteristics were obtained using 2400 and 6517 A source meters (Keithley). The electroluminescence (EL) output and EL spectra were measured using a S1337 or S2281 silicon photodiode (Hamamatsu Photonics) and a photonic multichannel spectral analyzer (Hamamatsu Photonics PMA-11), respectively. The photoluminescence (PL) spectra, and absolute PL quantum yields (PLQYs) were measured using an absolute PL quantum yield measurement system (Hamamatsu Photonics Quantaurus-QY). The values of the work functions of ITO and Ag were measured by a photoelectron spectrometer (Riken Keiki, AC-2).

3. Results and discussion

We investigated the influence of exciton quenching near the electrodes on the basis of the PL characteristics of F8BT films without electrodes and with ITO and Ag electrodes. We prepared F8BT films with a thickness of 15, 35, 60 and 90 nm. PLQYs of F8BT films without electrodes were estimated to be approximately 47% (15 nm), 63% (35 nm), 67% (60 nm) and 69% (90 nm). PLQYs of F8BT films on ITO electrodes were estimated to be approximately 25% (15 nm), 40% (35 nm), 43% (60 nm) and 45% (90 nm). PLQYs of F8BT films on Ag electrodes were estimated to be approximately 8.7% (15 nm), 26% (35 nm), 38% (60 nm) and 38% (90 nm). Fig. 2 shows the F8BT film thickness dependence of the quenching efficiency. The quenching efficiency, Q, is given by Eq. (1):

$$Q = \frac{I_A - I_B}{I_A} \tag{1}$$

where I_A and I_B are the PLQY of a neat F8BT film without electrodes and the PLQY of a F8BT film on an electrode, respectively. The quenching efficiencies of F8BT films on Ag electrodes were higher than those of F8BT films on ITO electrodes. While the quenching efficiency of the 15-nm F8BT film on the ITO electrode was approximately 0.5, the corresponding value for the Ag electrode was approximately 1. This indicates that the emission close to the Ag electrode is less efficient owing to the quenching of excitons by the electrode. We will discuss the

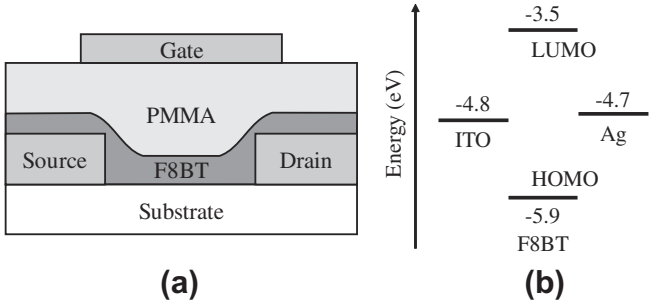

Fig. 1. (a) Device structure and (b) schematic energy diagram of a top-gate-type OLET.