

Ambipolar light-emitting organic field-effect transistor using a wide-band-gap blue-emitting molecule

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Abstract

We prepared ambipolar organic field-effect transistors and observed blue emission when both hole and electron accumulation layers were in the channel. We found that the reduction of carrier traps and controlling devices' preparation and measurement conditions were crucial for ambipolar operation.

1. Introduction

Integration of organic light-emitting diodes (OLEDs) and organic field-effect transistors (OFETs) have been aggressively studied to construct active-matrix displays. There have already been some reports on the fabrication of organic semiconductor based integrated devices. For example, Sirringhaus et al. and Dodabalapur et al. have reported integrated optoelectronic devices which combine organic-FETs and organic-LEDs^{1,2}. Since the most of integrated devices consist of two parts of FET and LED, rather complicated construction processes are required.

On the contrast, since light-emitting organic field-effect transistors (LEOFETs) have two functions of current switching and light emission in a single device with a simple structure, they have a great advantage for application to use in active-matrix displays. Such LEOFET was first reported in 2003 by using a tetracene thin film as an active layer and the green light emission was observed from the channel region of the FET³. However, since their device was operated in p-channel unipolar mode, the number of electrons injected was thought to be quite low compared to that of holes. There have been several reports to improve electron injection by tuning the organic/metal interface⁴⁻⁶. As a result, their luminance intensities were considerably improved and became sufficient for use in active-matrix displays. However, achieving

high efficiency in unipolar LEOFETs was rather difficult because only one type of charge carriers is accumulated in the channel. On the other hand, balance of holes and electrons is perfectly equal in ambipolar LEOFETs because accumulation layers of holes and electrons can coexist in the channel. Rost et al. reported such ambipolar LEOFETs by using a bilayer of p-type and n-type molecules⁷. However, strong exciton quenching is usually occurred at the interface of p-type and n-type organic semiconductors. Thus, ambipolar devices with a single organic semiconducting layer are expected to obtain high efficiency LEOFETs. Here, we prepared ambipolar LEOFET using a blue-emitting organic semiconducting layer and observed electroluminescence (EL)⁸. Recently, two groups reported ambipolar LEOFETs by using a single polymer active layer^{9,10}. These LEOFETs are also expected to be a novel light source for optoelectronic devices such as organic laser diodes.

2. Experimental

We used an organic semiconductor of 4,4'-bis(styryl) biphenyl (BSBP, Fig.1(a)) to prepare ambipolar LEOFETs. BSBP was train-sublimated 5 times before use. BSBP has a wide-band-gap of 2.9 eV and showed blue emission with a peak wavelength of 442 nm⁸. The absolute PL efficiency of the film was $\eta_{\text{PL}} = 20\%$. Figure 1 (a) shows a schematic view of the LEOFETs prepared in this study. A 100 nm thick hydroxyl-free poly(methyl-methacrylate) (PMMA) layer was dip-coated onto a SiO₂(300 nm)/n⁺⁺-Si substrate to eliminate electron traps on the SiO₂ surface. The substrates were baked at 120 °C for 1 hour to remove the residual solvent under the nitrogen atmosphere. After this procedure, we

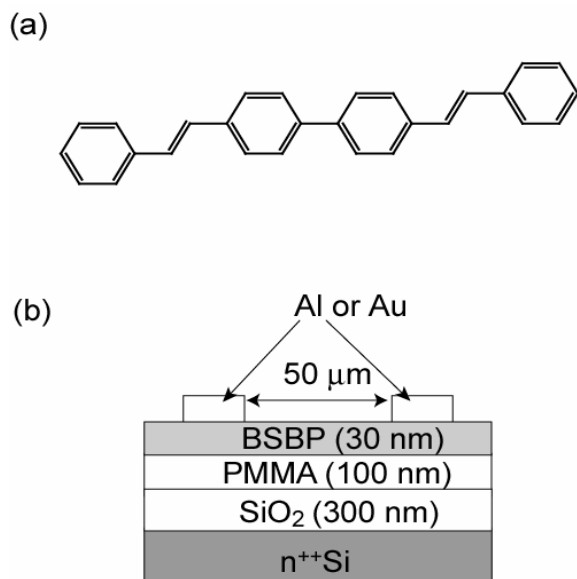


Fig. 1. Molecular structure of BSBP (a) and device structure of LEOFETs (b).

conducted the following device preparation and measurements under a vacuum below 1×10^{-5} Pa. The substrates were introduced into a vacuum chamber and baked at 80°C for 1 hour to remove adsorbed water. BSBP was vacuum deposited under a vacuum of 2×10^{-6} Pa. Their thickness and deposition rate were controlled to be 30 nm and 0.05 nm/s, respectively. The substrates were held at room temperature during deposition. We then transferred the specimens into another vacuum chamber to deposit Al or Au contacts through a shadow mask. The channel length and width were $50\ \mu\text{m}$ and 6 mm, respectively. The completed devices were transferred into a measurement chamber and electrical and optical properties were obtained under the vacuum of 1×10^{-5} Pa. We measured the devices' electrical characteristics with a semiconductor parameter analyzer (Agilent 4156C), and detected their light intensity through a quartz window of the vacuum chamber by using a calibrated Si photodiode (Newport 818UV) placed just above the devices.

3. Results and discussion

We first prepared a conventional bottom-contact type BSBP-based OFET having photolithographically patterned Au/Cr contacts to clarify typical semiconducting behavior of a BSBP film. Figure 2 shows transfer characteristics of the BSBP-based

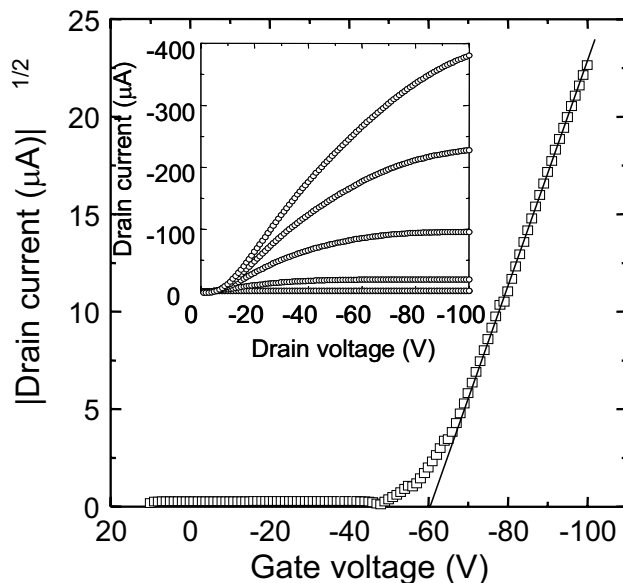


Fig. 2. Transfer characteristics of BSBP based OFET with bottom-contact type electrode of Au/Cr. Inset shows output characteristics. Gate voltages were set from 0 V to -100 V with step of -20 V.

OFET. The channel length and width were $25\ \mu\text{m}$ and 76 mm. The drain current increased with increasing gate voltage negatively, indicating that the device showed p-type semiconducting behavior. The field-effect hole mobility calculated from saturation regime was $0.01\ \text{cm}^2/\text{Vs}$. We confirm that the device did not show any indication of n-type semiconducting behavior when the gate and drain voltages were set to positive bias.

We prepared an ambipolar LEOFET having Al top contact electrodes and PMMA as an insulating layer. Since a BSBP layer has a wide-band-gap of 2.9 eV, we chose Al for both injections of holes and electrons. The PMMA layer was used to eliminate electron traps on a SiO_2 surface¹¹. Figures 3 (a) and (b) show transfer characteristics of the BSBP-based OFET operated in p-channel and n-channel modes, respectively. In both operation modes, the drain current took the minimum value and increased with increasing gate voltage, indicating clear ambipolar operation. The field-effect mobilities of electron and hole calculated from the saturation regimes were $1.5 \times 10^{-3}\ \text{cm}^2/\text{Vs}$ and $2.5 \times 10^{-5}\ \text{cm}^2/\text{Vs}$, respectively.

Figure 3 (a) also plots light intensity as a function of the gate voltages in p-channel mode. Although a current flow was observed when the gate voltage was

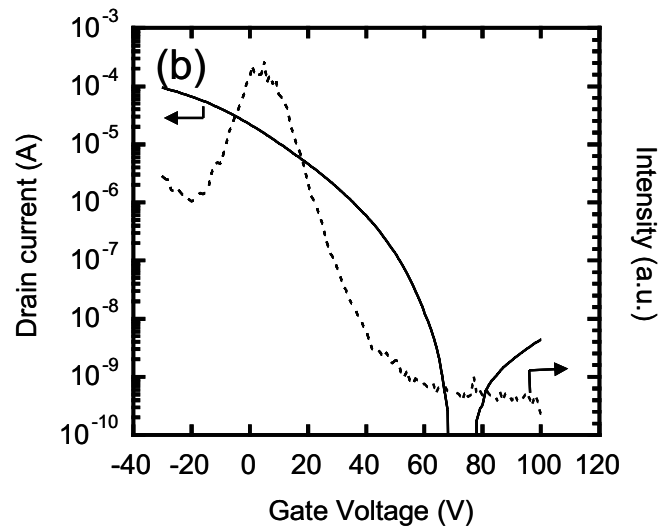
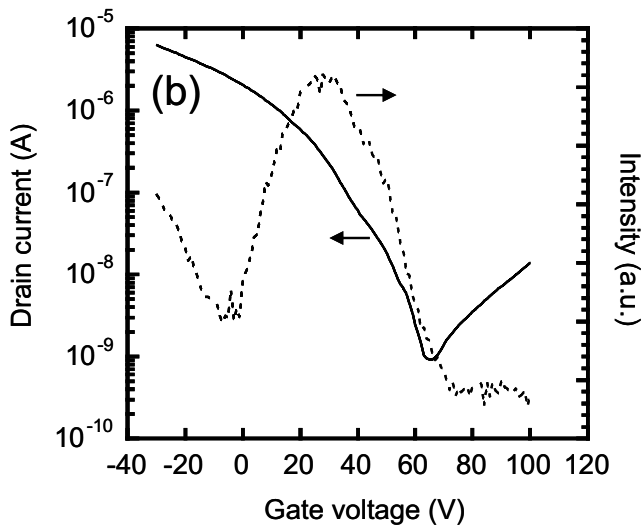
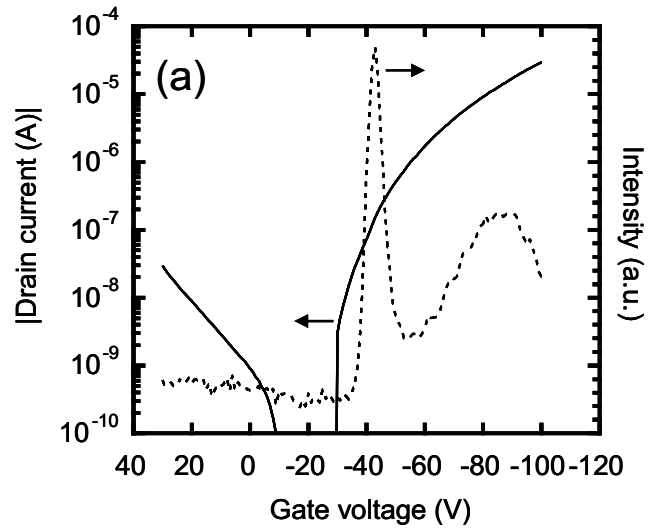
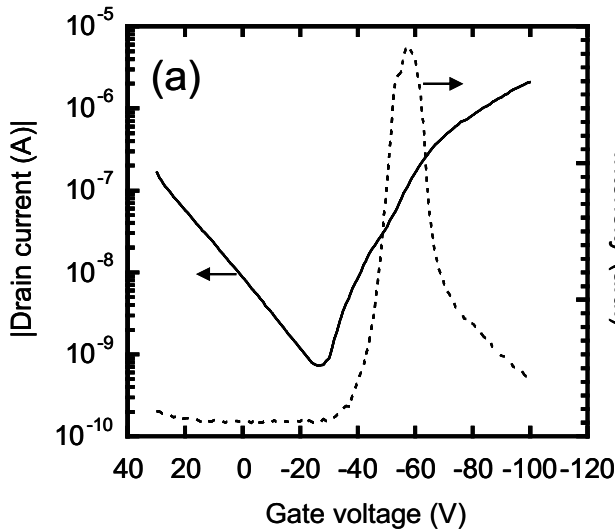


Fig. 3. Transfer characteristics and corresponding emission intensity of BSBP-based LEOFET with Al contacts operated in p-channel (a) ($V_d = -100$ V) and n-channel (b) modes ($V_d = 100$ V).

under -30 V, no light emission was detected because the charge carrier through the device was dominated by the electron flow from the drain to the source contacts. When the gate voltage was more than -30 V, light emission was observed and maximum intensity was obtained around -50 V. In this bias region, both electron and hole accumulation layers coexisted in the channel, leading to the light emission. This emission was observed with the naked eye in a darkened room, and we confirmed blue EL. We roughly estimated the external quantum efficiency of EL at the maximum intensity to be 0.5% . When the gate voltage was higher than -60 V, the emission intensity decreased

Fig. 4. Transfer characteristics and corresponding emission intensity of BSBP-based LEOFET with Au contacts operated in p-channel (a) ($V_d = -100$ V) and n-channel (b) modes ($V_d = 100$ V).

with increasing the gate voltage. This is due to the decrease of electron accumulation layer, leading to dominant flow of holes from the source to the drain contacts. Furthermore, when the device was operated in n-channel mode, light emission was also observed in the bias region where the electron and hole accumulation layers were coexisted in the channel (Fig. 3 (b)). These results indicate that light emission truly originated from the ambipolar operation. However, the maximum light emission was observed at the bias region where the drain current was unbalanced. This is probably due to the electron mobility was two order of magnitude lower than that

of the holes.

We also used Au for source and drain electrodes to clarify effect of the work function of the metal contacts. Figure 4 shows transfer characteristics and corresponding light intensity of the BSBP device with Au contacts operated in p-channel (a) and n-channel mode (b). Since the LUMO level of the BSBP was 2.7 eV and work function of Au was around 5.1 eV, it was thought to be difficult to inject electrons. However, n-type semiconducting behavior with light emission was also observed as in the case of the device with Al contacts. The field-effect mobilities of the hole and electron were $0.03 \text{ cm}^2/\text{Vs}$ and $8 \times 10^{-6} \text{ cm}^2/\text{Vs}$, respectively. The hole mobility was higher than that of the device with Al contacts because the energy gap between HOMO and the work function of Au is smaller than that of Al. On the other hand, electron mobility was lower compared to the device with Al contacts. These results indicate that control of the organic/metal interface is crucial to obtain high mobility, although it is not the requirement for ambipolar operation.

Furthermore, we should mention that the purity of the evaporation source is important. When we used the BSBP powder after one time purification by sublimation for evaporation source, we observed no n-type semiconducting behavior even though p-type was clearly observed. These results indicate that both controlling the organic/insulator and organic/metal interface and preparing high purity of the evaporation source are significant for obtaining ambipolar operation.

4. Summary

In summary, we demonstrated fabrication and successive measurement of ambipolar OFETs based on BSBP under high-vacuum without breaking vacuum. The application of an Al contact and a PMMA insulating layer allowed ambipolar operation, and light emission was observed when both hole and electron accumulation layers coexisted in the channel region. We found that low electron traps can be

achieved by controlling the organic/insulator interfaces, device preparation and measurement conditions. In addition, preparing high purity of the evaporation source is crucial for obtaining ambipolar operation. By optimizing the organic/metal interface, and enhancing both holes and electron mobilities, we can expect much stronger light emission.

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