

Observation of electron injection in an organic field-effect transistor with electroluminescence^{*}

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A tetracene field-effect transistor (FET) with Au source and drain electrodes showed p-channel type behaviour, where the injected holes were the main carriers. Application of an ac electric field to the source electrode, while drain and gate electrodes were grounded, induced electroluminescence (EL) around the source electrode that was caused by alternating electron and hole injection. This result indicated that electron injection into tetracene was possible from a metal with a high work function such as Au. The application of an ac voltage superposed on a dc voltage showed that electron injection was assisted by the space-charge field that originated from holes accumulated around the source electrode.

Key words: *electroluminescence; field-effect transistor; tetracene; space charge field*

1. Introduction

Organic materials have been used as electrical insulators because of their mechanical flexibility and high insulating performance which is why they are suitable for power cables, for example [1]. In order to use organic materials as insulators, we should always pay attention to dielectric breakdown phenomena, where charge injection from metal into insulator leads to electrical treeing and subsequently to dielectric breakdown [2]. Under an applied ac voltage, electrons and holes inject into an insulator alternately, and light emission is induced during their recombination process [3–5]. This light emission is known as electroluminescence (EL), and is utilized as one of the diagnostic methods for detecting dielectric pre-breakdown of an electrical insulator.

On the other hand, the recent discovery of organic semiconducting materials has opened a new way to use organic materials for electronic devices such as an organic

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field-effect transistor (OFET) and an organic light-emitting diode (OLED) [6]. In OLEDs operating under an applied dc voltage, electrons and holes are injected from cathode and anode, respectively, and they recombine, leading to the EL phenomenon [7, 8]. Here efficient charge injection is one of the key parameters for practical applications such as displays and light sources.

As mentioned above, organic EL is considered to be a phenomenon triggered by charge injection but it has two aspects; as a dielectric pre-breakdown phenomenon under ac electric field and as a light-emitting source of devices under a dc electric field. Therefore we consider two EL modes for discussing the EL mechanism, that is, an ac excited mode and a dc excited mode. For understanding the difference in the mechanisms of the two EL modes, we need to know the electric field distribution in organic devices because electrons and holes might be along the electric field, and carrier injection is ruled by the electric field formed at the electrode. However, injected charges are excess charges, and generate a space-charge electric field. This complicates the electric field distribution in organic devices and the carrier injection mechanism. Note that the determination of an electric field in OLED at the nanometer scale is generally difficult because active organic layers are installed as a metal–organic multilayer–metal systems.

OFETs have drawn our attention along with the discovery of high-mobility organic materials where injected carriers mainly contribute to a device performance [6], where carrier injection is a key parameter. Hepp et al. reported the enhancement of green light emitted from tetracene FET [9]. They argued that holes injected from the source electrode moved to the drain electrode, and then they recombined with electrons injected from the drain electrode via tunnelling. Interestingly, the use of three electrodes as in OFET provides a way to control hole and electron injection, because we can choose the drain and gate voltages independently. We can see such examples in recent experiments on organic EL using OFET structure [10, 11]. That is, light-emitting OFET can be used as one of the model systems to study physics of EL devices.

In our previous study, we observed EL from tetracene FET under an ac electric field, and concluded that the EL under an ac electric field is helpful for investigating the carrier injection process [12, 13]. In this study, we focused on the effect of space charge field caused by injected carriers under an ac electric field.

2. Experimental

Sample preparation. Tetracene was purchased from Tokyo Kasei Kogyo and was used without further purification. It is well known as an active material in OFET and OLED because of its high carrier mobility and intrinsic, visible luminescent emission [14, 15]. Therefore, tetracene is one of the most appropriate materials for observing the EL enhanced in an OFET structure. Tetracene thin films show absorption with the maximum at ca. 520 nm and photoluminescence (PL) at ca. 532 nm [16].

Figure 1 illustrates the structure of the tetracene FET employed. High-doped Si substrate, coated with a thermally grown SiO_2 layer (500 nm thick) was used for the OFET substrate. Interdigital Au electrodes were deposited on the SiO_2 as source and drain electrodes, using the photolithography technique. The channel length and width were 50 μm and 11 cm, respectively. High-doped Si was used as a gate electrode. The substrate was cleaned, in an ultrasonic cleaning system, with acetone, ethanol, and distilled water, and then it was subject to UV/ozone treatment for 30 min. After that, tetracene was deposited on the substrate by thermal evaporation in a vacuum at the pressure lower than 2×10^{-6} Torr. The deposition rate and the film thickness were adjusted to about 0.6–1.1 $\text{\AA}/\text{s}$ and 200 nm, respectively, using a quartz crystal microbalance.

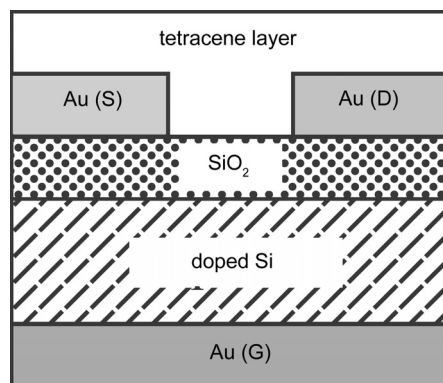


Fig. 1. Device structure of tetracene FET

Measurement. Figure 2 shows the experimental setup for EL measurement. A square wave voltage was applied between the source (S) and gate (G) electrodes

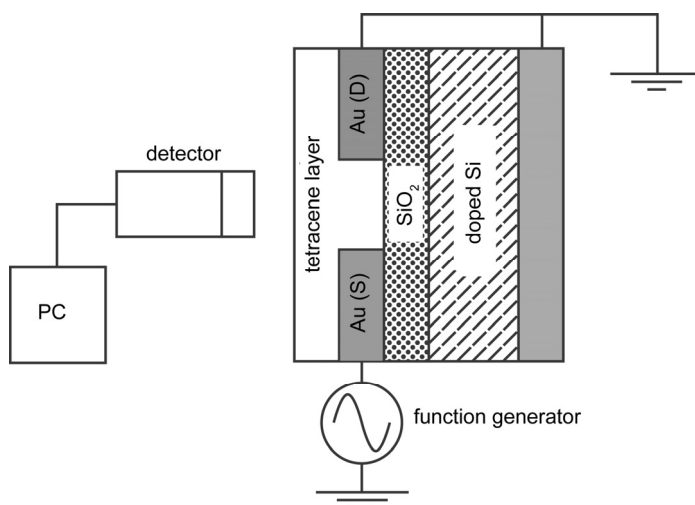


Fig. 2. Experimental setup for the observation of EL under ac electric field. This setup was located in the chamber and the measurement was carried out in vacuum or in Ar gas atmosphere

using a function generator (NF WF1944) and a high-speed amplifier (NF 4005), while the drain (D) and gate electrodes were grounded. Hereafter, we refer to this square wave voltage as “ac voltage”. By applying the ac voltage, electron and hole injection take place alternately from the source electrode. The spatial distribution of the EL intensity in tetracene FET was monitored using a charge-coupled device (CCD) imaging sensor (Andor DU420-BV), where a 40 V ac voltage at 300 kHz was applied in an Ar gas atmosphere. The EL spectrum from tetracene FET was obtained by using a multi-channel spectrometer (Andor SR163 with Newton 920-BV) in a vacuum of less than 10^{-6} Torr. Here, the EL was measured by applying a 50 V ac voltage at 500 kHz. Photoluminescence (PL) spectrum was obtained by using the spectrometer (BWtek BTC112E) where the ultraviolet light emitting diode (LED) at the wavelength of 400 nm was used for the excitation of tetracene thin film. In addition, EL intensity was measured in a vacuum using a photomultiplier tube (PMT; Hamamatsu R3869) by changing the ac voltage amplitude from 10 V to 60 V. EL intensity was monitored as a PMT current, I_{photo} , by using a digital picoammeter (Keithley 6487). In this measurement, the applied voltage was always positive, in order to examine the effect of space charge field.

3. Results and discussion

3.1. Current-voltage characteristics

The output and transfer characteristics of tetracene FET are shown in Fig. 3a, b, respectively. Tetracene FET operated as a p-channel type OFET, where holes injected from an Au electrode made the main contribution to the drain current. The hole mobility, calculated from the slope of the transfer characteristics, was about $5 \times 10^{-4} \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$.

3.2. Observation of EL

Figure 4 shows the spatial distribution of EL intensity in the channel region of tetracene FET. The enhancement of EL was observed around the edge of the source electrode, because holes and electrons injected into the tetracene alternately, and recombined in the same region. Note that the light emitted in this process could be seen in the dark with the naked eye.

The result suggested that EL was enhanced only around the edge of the source electrode, not in the channel region of the FET. The following reasons were considered:

1. ac frequency was too high and injected carriers did not spread over the channel.
2. Nearly all injected electrons were trapped at the interface between the tetracene and the source electrode, and they did not enter into the channel region.
3. Carrier injection was only possible from the source electrode, and injected electrons and holes recombined only around the edge of the electrode.

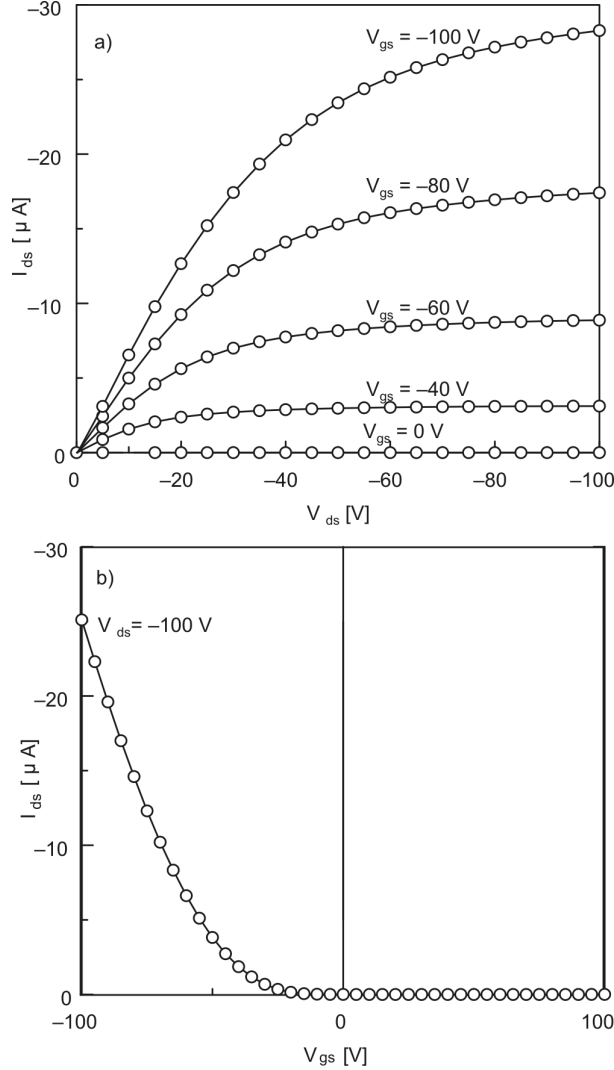


Fig. 3. Output characteristic (a), and transfer characteristics (b) of tetracene FET. Digital source meter (Keithley 2400) was used for voltage source and measurement of the drain current. V_{ds} was set at -100 V in transfer characteristics

Here it is instructive to calculate the hole transport distance, L_{drift} , within a half cycle of ac voltage. L_{drift} is given by

$$L_{\text{drift}} = \mu_h E_{\text{ext}} \frac{1}{2f} \quad (1)$$

where μ_h is the hole mobility, E_{ext} is the external electric field from the source to drain direction, f is the ac frequency. From Eq. (1) with $\mu_h = 5 \times 10^{-4} \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$, $E = 8 \times 10^3 \text{ V} \cdot \text{cm}^{-1}$, and $f = 300 \text{ kHz}$, L_{drift} is calculated to be about 67 nm from the source elec-

trode. Therefore, the first explanation (i.e. reason 1 above) accounts for the local light-emission around the source electrode. However, we cannot exclude the other reasons (2 and 3) from the experimental results obtained here, and further investigation is needed.

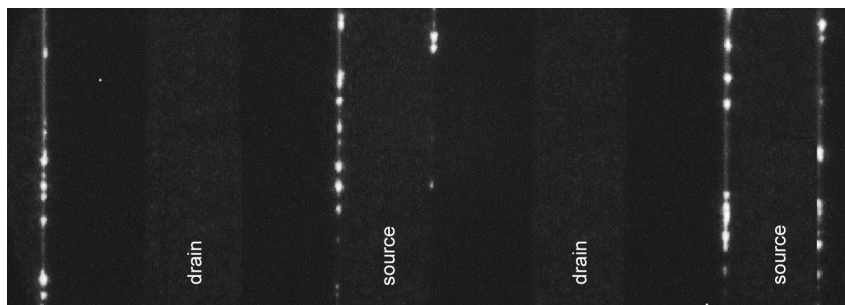


Fig. 4. Spatial distribution of EL intensity in tetracene FET

In summary, electron injection was detected using the EL under an ac electric field, even when this electron injection was undetectable from the FET characteristics.

3.3. EL spectrum

EL and PL spectra are shown in Fig. 5. There is an EL spectrum peak at the wavelength of ca. 540 nm, corresponding to the position of the PL spectrum peak. This

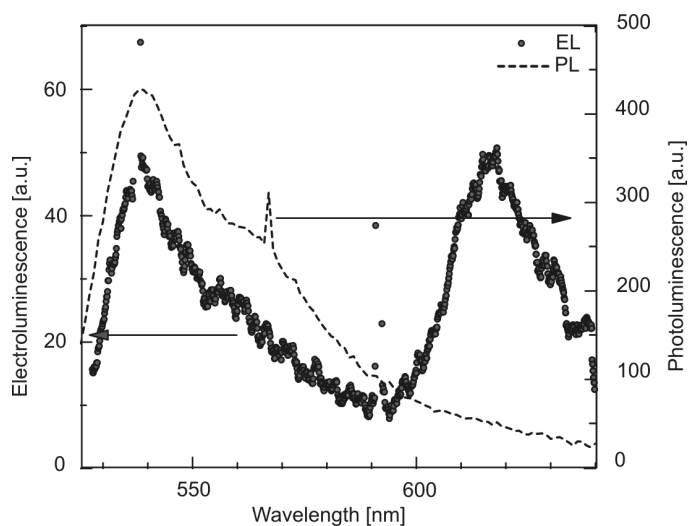


Fig. 5. EL and PL spectra of tetracene FET

peak indicates that this emission originates from the intrinsic luminescence of tetracene thin film [9, 13]. On the other hand, another EL peak appeared at a longer wavelength of ca. 620 nm, possibly due to the presence of an interfacial trapping state be-

tween the metal electrode and the tetracene thin film [17]. The average depth of the trapping state, calculated from the peak separation, was estimated to be about 0.29 eV from the highest occupied molecular orbital (HOMO) state, for a hole, or -0.29 eV from the lowest unoccupied molecular orbital (LUMO) state, for an electron. However, we should consider a possible presence of electron traps, because holes inject and transport very smoothly in comparison with electrons (see Fig. 3).

3.4. Electron injection process from Au electrode

From the current–voltage characteristics, we could not detect the presence of electron injection. However, the EL measurement under an ac electric field suggested that alternating carrier injection from the electrodes did indeed occur. Two reasons were considered; one is that injected electrons do not move in the material; the other is that electron injection is assisted by the space-charge field formed by accumulated holes in the FET channel region. To clarify the electron injection process assisted by the space charge field, a square-wave voltage, having the form illustrated in Fig. 6, was applied to the source electrode. Note that the voltage applied to the source electrode changed in a positive region. That is, electrons could not inject into tetracene by an external electric field. Voltage amplitude was defined V_p , as given in Fig. 6, and the V_p dependence of EL intensity was measured.

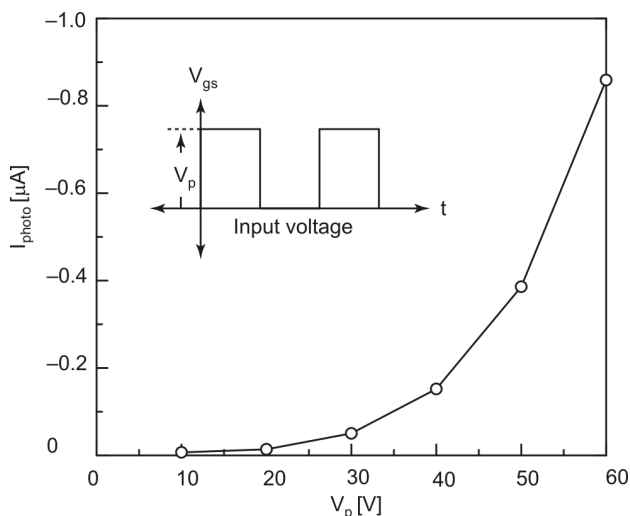


Fig. 6. V_p dependence of EL intensity. The voltage waveform illustrated in the graph was applied to the source electrode in this measurement

Figure 6 shows that EL intensity increased as the V_p increased, though the applied voltage was always in a positive region. This indicates that electrons were injected not only by an external electric field but also by the space charge field. Injected electrons recombined with holes which were injected and accumulated in tetracene by an exter-

nal electric field V_p . That is, the accumulated holes form a space charge field that assists electron injection. As mentioned above, the space charge field plays a significant and important role in the charge injection process under an ac electric field.

Furthermore, the light emission initiated at the voltage V_p of about 20 V, where the electric field is about $4 \times 10^5 \text{ V} \cdot \text{cm}^{-1}$. This field reasonably accounts for charge injection from metal into organic materials.

4. Conclusion

We investigated the charge injection from an Au electrode into tetracene in an OFET structure using EL under an ac electric field. We detected the existence of EL under an ac electric field using a CCD imaging sensor and a multichannel spectrometer. We observed EL from the edge of the source electrode of tetracene FET by applying an ac voltage to the source electrode. This EL had a spectrum peak at a wavelength corresponding to the PL spectrum peak. We confirmed the electron injection without the external electric field by using the EL. The effect of a space charge field formed by accumulated holes is important in the charge injection process under an ac electric field.

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