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# Thin Solid Films



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# Space charge field effect on light emitting from tetracene field-effect transistor under AC electric field

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# article info abstract

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By applying square wave AC voltage to the Au source electrode of tetracene based field-effect transistor (FET), electroluminescence (EL) was obtained. The results suggest that electrons and holes were injected alternately from the source electrode and recombined each other, and lead to the EL. This type of EL was localized at the interface between the source electrode and tetracene, and enhanced periodically with two relaxation times in accordance with the applied AC voltage cycle. We modeled the carrier behavior in the FET and explained the decay of EL, taking into account the space charge field contribution. Finally, using an AC voltage superposed on DC bias voltage, it was shown that electron injection was prompted only by space charge field.

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# 1. Introduction

Organic polymer materials such as polyethylene are well known as a high-performance insulator, which are used in power cables, and so on [\[1\].](#page--1-0) For sustainable power supply, diagnostic method to detect dielectric deterioration and breakdown phenomena is very important in electrical insulating engineering. Charge injection from metals into organics causes dielectric breakdown. Consequently, electroluminescence (EL) is enhanced as a premonitory phenomena of dielectric breakdown under AC electric field, where electrons and holes are injected alternately into organics and recombine [2–[4\].](#page--1-0) Hence EL under AC electric field is available for detecting the deterioration of materials.

On the other hand, since the discovery of high efficiency and long lifetime EL phenomena, OEL has been investigated in electronics, especially as organic light-emitting diodes (OLEDs) [\[5,6\]](#page--1-0). In OLEDs, electrons and holes are injected from the two different facing electrodes into materials under DC electric field, and they recombine during the light-emitting process. Therefore, OEL under DC electric field has been intensively investigated for the device application, where the lifetime of light-emission is one of the most important issues for the practical use. Recently, OEL from organic field-effect transistors (OFETs) has been investigated as a new structural device for displays and so on [\[7,8\].](#page--1-0)

In this study, making use of the experiments on tetracene fieldeffect transistor (FET) with AC applied voltages, we observed EL and examined the carrier behaviors in organic devices, especially charge injection and space charge field effect. In our previous study, we could show the enhancement of EL from tetracene FET under AC electric field [\[9,10\].](#page--1-0) Other groups also showed the EL enhancement from OFETs using conducting polymers [\[11\]](#page--1-0). However, the details are still not clear. In this study, we focused on the relaxation process of the EL intensity under AC electric field and analyzed the details, using a simple model. The importance of space charge field contribution was suggested, and we confirmed it by making the experiments with applying AC voltage superposed on DC voltage.

# 2. Experimental

#### 2.1. Sample preparation

Tetracene purchased from Tokyo Kasei Kogyo was used as an active layer of the OFET without any purification. Tetracene thin film was prepared by evaporation in a vacuum at a pressure of less than  $2\times10^{-6}$  Torr. The evaporation rate was 0.6–2.0 Å/s, and the film thickness was adjusted to about 73 nm using a quartz crystal microbalance (QCM). The substrate was high-doped-Si coated with 500-nm-thickness  $SiO<sub>2</sub>$ . Au source and drain electrodes were deposited on the  $SiO<sub>2</sub>$  surface, where the channel length and width were  $50 \mu m$  and 11 cm, respectively. The electrode structure was interdigital. The substrate was cleaned in an ultrasonic cleaning system with acetone, ethanol, and distilled water. Before the deposition of tetracene, the substrate was subjected to UV/ozone treatment for 30 min. After that, the substrate was transferred from the cleaning chamber to the evaporation chamber, and the film was deposited. After the deposition, the tetracene FET was transferred to the measurement chamber immediately, and then we started the experiment.

### 2.2. Measurement procedure

The current–voltage and EL characteristics were measured in the vacuum chamber at a pressure of less than  $5 \times 10^{-7}$  Torr. In the current–

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Fig. 1. Applied voltage waveforms and the OFET situations. (a) Experiment 1 represents alternating electron and hole injection, and (b) experiment 2 represents electron injection by space charge field.

voltage measurements, the  $I_{ds}-V_{ds}$  and  $I_{ds}-V_{gs}$  characteristics were measured using a digital source meter (Keithley 2400). In the  $I_{ds}-V_{gs}$ measurement,  $V_{ds}$  was  $-100$  V.

EL from tetracene FET was monitored using a photo-multiplier tube (PMT, Hamamatsu R3896) with digital oscilloscope (Tektronix TDS220). In EL measurement, a square wave voltage was applied to the source electrode, where the drain and gate electrodes were connected to the ground. The applied gate-source voltage provoked the hole and electron injection alternately from the source electrode, while the drain-source voltage provoked the carrier transport. The schematic of this experiment is portrayed in Fig. 1(a), where the alternating voltage is applied to inject holes and electrons alternately. Hence EL is assumed to be enhanced due to the recombination of injected holes and electrons. The frequencies were 100 Hz, 300 Hz, and 500 Hz, and the applied voltage amplitude was changed from 30 V to 50 V. The experiments using AC applied voltage superposed on DC voltage was also carried out. By choosing a DC bias voltage appropriately, we could make an experimental condition, where only hole injection is allowed, as illustrated in Fig. 1(b). The frequency used was 500 Hz, and applied voltage was chosen from 50 V to 70 V. In all of the above experiments, 128-time-mean-value was used to obtain the EL data.

# 3. Results and discussion

# 3.1. Current–voltage characteristics

Fig. 2(a) and (b) show the  $I_{ds}-V_{ds}$  and  $I_{ds}-V_{gs}$  characteristics of tetracene FET, respectively. The tetracene FET showed a typical p-type FET characteristic, and we could not see the current caused by electron transport. The hole mobility and threshold voltage were about  $5 \times 10^{-4}$  cm<sup>2</sup>V<sup>-1</sup> s<sup>-1</sup> and -18 V.

# 3.2. Observation of EL

[Fig. 3](#page--1-0) shows an example of OEL enhanced from the tetracene FET. [Fig. 3\(](#page--1-0)a) represents the spatial image of EL enhancement. Lightemission was localized at the edge of the source electrode, indicating that injected electrons and holes recombined around the interface between source electrode and tetracene. [Fig. 3](#page--1-0)(b) shows the EL spectrum with the photoluminescence (PL) spectrum. Two peaks were observed in the EL spectrum. One is the intrinsic EL of tetracene film (540 nm), which appears at a wavelength corresponding to the PL peak, and the other is trap-induced extrinsic EL (625 nm) [\[10\].](#page--1-0)

# 3.3. Waveforms of the EL intensity

[Fig. 4](#page--1-0) shows the EL waveforms monitored using a PMT (experiment 1). The EL enhanced periodically, in accordance with the applied alternating voltage. In that figure, only one cycle of the recorded EL



Fig. 2. (a) The  $I_{ds}$ – $V_{ds}$  and (b)  $I_{ds}$ – $V_{gs}$  characteristics of tetracene FET.  $V_{ds}$  was  $-100$  V in  $I_{ds}$ – $V_{gs}$  measurement.

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