Observation of Electron Injection from Au Electrode into Organic Field-Effect Transistor with Electroluminescence

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1. Introduction

Organic materials have been used as electrical insulators in electronics and electrical engineering, where the mechanical flexibility and excellent insulating performance are the most attractive functions. For organic insulating materials, charge injection from the metal contributes to the deterioration of the material leading to the dielectric breakdown. Especially under AC electric field, electrons and holes are injected from the metal alternately and then electroluminescence (EL) is observed due to the radiative recombination of them [1]. Accordingly, EL has been observed to detect the dielectric pre-breakdown phenomena in organic insulators. On the other hand, organic electronic devices, such as organic field-effect transistors (OFETs) and organic light-emitting diodes (OLEDs), have called our attention since the discovery of high-mobility organic materials. For organic semiconducting materials, charge injection makes a main contribution to the electric conduction because the carrier density in organic materials is determined by injected charges. For example, holes injected from cathode and electrons injected from anode make radiative recombination in OLEDs by DC electric field [2][3].

From these facts, it is considered that EL phenomena have two different aspects depending on injected carrier motions (for example, carrier motions under AC and DC electric fields). Therefore, the observation of carrier motion in organic materials concerning with EL phenomena is important to understand the EL physics.

Usually, metal-insulator-metal (MIM) structure is used to observe the EL. However, it is difficult to observe the carrier motion in MIM structure because the direction of carrier motion is perpendicular to the organic thin film. Therefore, we used the OFET structure where the direction of carrier motion is parallel to the organic thin film plane.

OFETs have been intensively studied as injection-type elements. Generally, most OFETs drive as a unipolar device since holes are dominantly injected from the Au source and drain electrodes. Recently, double-injection-type OFET was reported by Hepp et al. [4], where EL is observed because of the recombination of injected electrons with accumulated holes at the drain electrode. This fact motivates us to use the OFET structure for observation of carrier motion. Further, OFET has a three-electrode system, which is an advantage in the study of carrier motion because it can control the electric field distribution and consequently control the carrier distribution in organic thin film.

Our previous study reveals the alternate electron and hole injection from Au electrode into organic thin film in OFET structure with the observation of EL using AC electric field [5][6]. In this paper, we focus on the EL under AC electric field and investigate it in detail.

2. Experiment



Fig. 1: Experimental setup

We used tetracene as active layer of bottom-contact OFET, where Au was used as the source, drain, and gate electrodes. The source and drain were interdigital electrodes where the channel length and width were 50 μ m and 11 cm, respectively. SiO₂ was used as a gate insulator (500 nm thickness). Tetracene layer was deposited in a vacuum under 2×10⁻⁶ Torr, and the film thickness was adjusted to about 200 nm using a quartz crystal microbalance.

A square wave AC voltage was applied to the source electrode, where the drain and gate electrodes were connected to the ground (see Fig. 1). The measurement was carried out in a vacuum ambience under 10^{-7} Torr. Applied AC frequency was 500 kHz. Light emission from tetracene FET was detected using photomultiplier tube (PMT), charge-coupled device (CCD) image sensor, and photonic multichannel analyzer (PMA). The EL intensity was monitored as a PMT current, I_{photo}.

In our previous study, the EL under AC electric field was observed, but electron injection mechanism was not clear [5][6]. To investigate the mechanism of electron injection from Au electrode, we controlled the waveform of applied AC voltage. Only positive voltage was applied to the electrode (see Fig. 3). In this measurement, the positive voltage (V_p) dependence of EL intensity was evaluated.



Fig. 2: (a) EL intensity distribution in tetracene FET and (b) EL and PL spectra of tetracene thin film

3. Results and Discussion

Figure 2 (a) shows the EL intensity distribution under AC electric field. The EL was observed from the interface between Au source electrode and the tetracene thin film. This result indicates that electron-hole recombination zone was limited at the interface. Figure 2 (b) shows the spectra of photoluminescence (PL) and EL of tetracene thin film. EL spectrum corresponded well to PL spectrum around the wavelength of 540 nm, where the intrinsic luminescence spectrum peak of tetracene thin film appears [7]. On the other hand, EL spectrum had another peak around the wavelength of 625 nm, where no PL spectrum peak was observed. The origin of this spectrum peak was under investigation but we believe it was owing to the presence of the interfacial trapping state between metal electrode and tetracene thin film. Generally, there are lots of trapping states around the metal-organic interface [8]. Because of this, excitons formed around the metal-organic interface could recombine through these interfacial trapping states. Therefore, EL spectrum peak is considered to appear at the longer wavelength than intrinsic EL, where the peak shift corresponds to the average depth of the trapping states.



Fig. 3: Positive voltage dependence of EL intensity

Figure 3 shows the V_p dependence of EL intensity. EL intensity increased with the increase of V_p , indicating that electrons were injected from Au electrode when $V_{gs} \ge 0$ and they were then recombined with holes accumulated in the material. Further, it is considered that the injected electron density increased with V_p , which indicates injected hole density also increased. These results can be explained by assuming a presence of space charge (SC) electric field formed by accumulated holes.

Since the injected charges are the excess charges for organic materials, a SC field is formed. This electric field triggers electron injection from the Au metal electrode into organic material. In other words, SC field and external field are responsible for carrier injection.

4. Conclusion

We observed EL from tetracene FET under AC electric field caused by alternating electron and hole injection. The results suggest that not only external field but also SC field are responsible for charge injection.

References

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