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Direct observation of carrier behavior leading to electroluminescence in tetracene field-effect transistor

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1. Introduction

Organic materials, such as polyethylene (PE) and polyimide (PI), have been used as electrical insulators because of their high insulating performance and mechanical flexibility [1]. As a dielectric pre-breakdown phenomenon, electroluminescence (EL) is enhanced under AC electric field. Hence organic EL (OEL) has been used as a diagnostic method for dielectric breakdown, caused by alternately carrier injection [2]. On the other hand, OEL has been intensively studied for use as a light source, e.g., organic light-emitting diode (OLED) [3]. In OLED devices, EL is generated from the materials subjected to DC electric field, where carriers injected from two-facing electrodes recombine to radiate light. These results suggested that there are two modes in OEL, and this motivated us to study EL enhanced from OLED under AC electric field, in terms of carrier behavior leading to the degradation.

In our previous study, we showed that alternating carrier injection from the source and drain electrodes caused OEL from organic field-effect transistors (OFET) with an active layer of tetracene [4]. Results suggested that space charge field formed by accumulated carriers around the electrodes also made a significant contribution to carrier injection, leading to the OEL. However we could not evidently show the details. In this study, carrier behavior leading to this OEL in tetracene field-effect transistor (FET) was directly probed by using electric field induced optical second harmonic generation (EFISHG) technique.

2. Experiment

EFISHG was employed for probing carrier behaviors leading to OEL from tetracene-FET. Injected carriers are excess charges, and a space charge field could be formed in an active layer of tetracene. As a result, non-linear polarization $P_i(2\omega)$ is induced in tetracene when a laser with an angular frequency ω impinges on the tetracene, and it is given by

$$P_i(2\omega) = \varepsilon_0 \chi^{(3)}{}_{ijkl} E_j(0) E_k(\omega) E_k(\omega)\,. \qquad (1)$$

Here ε_0 is the dielectric permittivity of vacuum, $\chi^{(3)}{}_{ijkl}$ is third order nonlinear susceptibility, $E_i(0)$ is static electric field, which contains space charge field and external field,

$$E_i(0) = E_{sc} + E_{ext}\,, \qquad (2)$$

and E_i (ω) is electric field of impinging laser. EFISHG is generated by $P_i(2\omega)$ and it has double angular frequency2ω. Therefore, we could measure static electric field, $E_i(0)$, by probing the EFISHG. The experimental set up was shown in Fig.1(a). Enhanced EFISHG was recorded using a CCD imaging sensor with band-pass filter. Transient EFISHG was monitored, by choosing the timing of incident pulsed-laser light and applied pulsed-voltage (t_d) (see Fig.2 (c)). EL measurement was also carried out using a photomultiplier tube with band-pass filter, where transient EL was monitored. Images of EL enhanced from tetracene-FET were recorded using a CCD imaging sensor. In EL and EFISHG experiments for tetracene FET, we applied pulse voltage with amplitude of 120V.

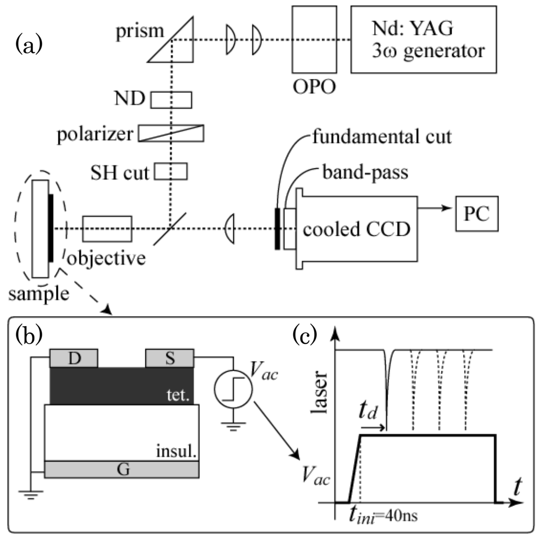

Fig. 1: experimental setup for SHG measurement

We prepared OFETs with an active layer of 200 nm-thick tetracene, in the way same as in our previous study [4], using Si-substrate, where the source and drain electrodes were Au. The thickness of gate insulator, SiO_2, was 500 nm, and the channel length and width were 30 µm and 3 mm, respectively. We also prepared the single layer OLED with a structure of ITO/tetracene/Al, where the thickness of tetracene was 400 nm.

3. Results and Discussions

We observed the EL spectra of ITO/tetracene/Al under DC and AC electric fields (see Fig. 2). One luminescent peak was observed at a wavelength of 540 nm, and it was assigned to the intrinsic luminescence of tetracene thin film [5]. On the other hand, one small EL spectrum peak was also observed at a wavelength of 620 nm, but it was obtained only when the AC electric field was applied, suggesting that alternating electron and hole injection was

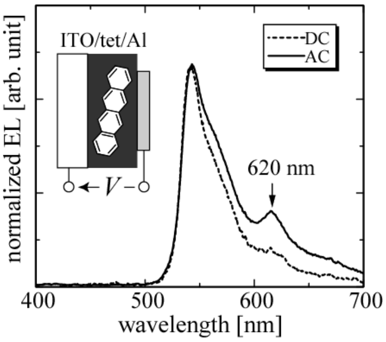

Fig. 2: EL spectra from tetracene

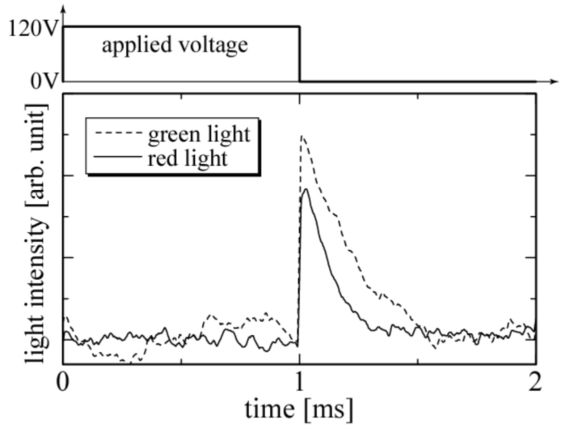

Fig. 3: time dependence of EL intensity

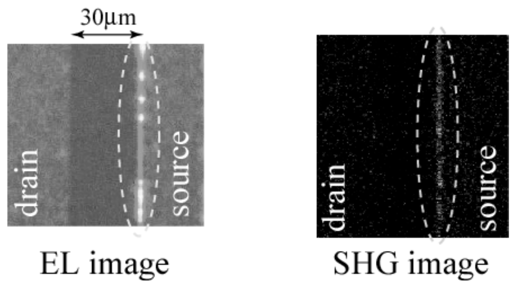

Fig. 4: spatial distribution of EL and SH intensity

responsible for this EL enhancement. Results evidently showed that there are two kinds of EL from the tetracene. The EL enhanced around 620nm was our main interest here.

Fig.3 shows the transient EL from the tetracene-FET. Green (540 nm) and Red (620 nm) emitting lights were observed when an applied voltage was switched-off. Note that this EL was a transient one and never enhanced continuously. Results suggested that this EL was generated only when holes were accumulated at the tetracene/SiO_2 interface in the absence of external voltage.

Fig.4 shows the spatial distribution of enhanced EL and EFISHG from the tetracene FET. Interestingly, these enhancements were observed only around the tetracene/electrode interface. Results of EFISHG evidently showed the presence of the electric field formed by accumulated charges (holes) around the tetracene/electrode interface (see Eq.(2)). This suggested that the space charge field assisted charges with opposite polarity (electrons) to go into the tetracene layer, resulting in the hole-electron recombination at the tetracene/electrode interface leading to

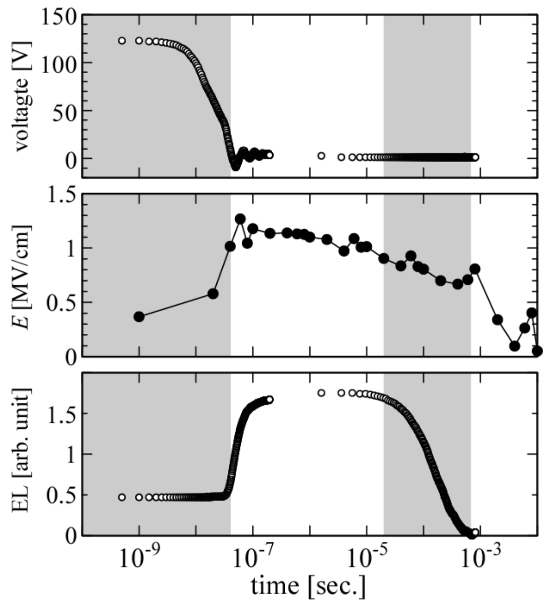

Fig. 5: time dependence of EL and SH intensity

the enhancement of transient EL.

Fig.5 shows the waveform of applied voltage, enhanced EL, and electric field at the tetracene/electrode interface, *E*, with time. The EL and electric field immediately rose after the applied voltage was switched-off. After that, the electric field gradually decayed, while the EL immediately extinguished around 10^{-6} s. Results suggested that electron-injection for the EL enhancement ceased, though a space charge field was non-zero. It indicates that there is high injection barrier for electron-injection, and electrons cannot enter into tetracene below the electric field of about 10^6 V/cm. The accumulated holes eventually disappeared from tetracene-FET, in a manner similar to the general discharging [1].

4. Conclusion

We observed the transient EL and EFISHG simultaneously, and evaluated the carrier behavior in the OEL under AC electric field. Results evidently showed that a space charge field formed by accumulated holes assisted electron injection, and resulting in electron-hole recombination followed by OEL.

Acknowledgements

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