# Hall effect of polycrystalline pentacene field-effect transistors on plastic films

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# 1. Introduction

We have successfully performed the systematic Hall effect measurements on polycrystalline pentacene field-effect transistors (FETs) manufactured on a plastic base film with polyimide gate dielectric layers. This is the first demonstration of the Hall effect measurements on polycrystalline organic FETs on plastic films.

There were a few reports on the Hall effect of OFETs using single crystal rubrene [1, 2], which pointed out that its transport mechanism was band-like. It is very important to research the single-crystalline OFETs to understand the solid-state property in pentacene grains. On the other hand, although mechanical flexibility is noted as one of the most important features in practical applications [3], the charge transport of polycrystalline organic semiconductor has not yet fully understood.

In this work, we performed precisely Hall effect measurements on the high-quality polycrystalline pentacene **FETs** manufactured on plastic films. Measurement temperature and gate voltage  $(V_G)$ dependence of the sheet carrier density (N<sub>S</sub>) and hall mobility  $(\mu_H)$  were systematically investigated. It was found that  $N_S$  was evaluated to be  $4\pm0.7{\times}10^{12}~\text{cm}^{-2}$  and  $\mu_H$ to be 0.4 cm<sup>2</sup>/Vs at V<sub>G</sub> = - 40 V. A typical Hall effect measurement has not yet been achieved in polycrystalline organic FETs on plastic films thus far. This is mainly due to the fact that the application of continuous DC voltage bias degrades the transistor performance. Recently, we succeeded in suppressing the DC bias stress degradations of organic FETs using annealing process, an excellent stability under DC bias stress open up to perform high-precision Hall effect measurements under high magnetic fields.

# 2. Experiment and discussion

High performance pentacene FETs for the Hall effect measurement (using the Van der Pauw method) have been fabricated by a vacuum evaporation process. First, the gate electrode was formed by thermal evaporation of 50-nm-thick Au through a shadow mask on a 75-µm-thick polyimide-sheet plastic film. Then, a 500-nm-thick polyimide gate dielectric layer was prepared by spin coating and a 50-nm-thick pentacene was deposited to form the 600-µm-square through a thermal evaporation. Finally



Fig.1 (a) The cross-sectional illustration of the organic transistor. (b) The photograph of organic transistor for the Van der Pauw method in overhead view.

the 50-nm-thick Au was evaporated to form four electrodes of 50  $\times$  50  $\mu m^2$  at the corner using a shadow mask. For reference, pentacene FETs with the channel length of 100  $\mu m$  and width of 1 mm were also fabricated. The evaluated field-effect mobility ( $\mu_{FET}$ ) was 0.4 cm²/Vs in the linear regime and 1.0 cm²/Vs in the saturation regime, indicating high mobility even on plastic films. And an on/off current ration is above  $10^6.$ 

The transistor properties of the FETs were measured using a precision semiconductor-parameter analyzer (Agilent Technologies 4156c). The Hall effect measurement were performed using a lock-in amplifier system 7265, while electric current were applied by Keithley 6221. The sample was set in a physical property measurement system (PPMS; Quantum Design) which could apply magnetic fields up to 9 T and shield from light in helium gas environment. These system and sample enable us to perform typical Hall effect measurement.

There have been difficulties to measure the Hall effect of OFETs. This is because the polycrystalline OFETs have quite high impedance of the order of  $M\Omega$ , resulting in small Hall voltages of the order of  $\mu V$ . Furthermore, transistor performance was changed with continuous voltage applications to gate, source, and drain electrodes, which was so-called DC bias stress, resulting in low reliability of observed Hall voltages. We fabricated high-mobility OFETs (typically > 1 cm<sup>2</sup>/Vs) and suppress the DC bias stress degradations, which solved these problems.

We confirmed the magnetic field dependence of Hall voltage with applications of  $V_G = -40$  V and current of 0.5



Fig.2 The magnetic field dependence of Hall Voltage as a function of time for a pentacene OFETs at fixed  $V_G$ =-40 V and T=300 K, with ±9, ±6, and ±3 T.

 $\mu$ A. Hall voltages were linearly decreased from 98.8 mV to 98.2 mV with increasing magnetic fields from 0 T to 9 T. Figure 2 shows the change in Hall voltage with applying magnetic fields of ±3, ±6, and ±9 T, which had linear response to magnetic field. There is a monotonic drift of the offset Hall voltage. This may be caused by the asymmetry of electrodes upon the organic semiconductor. In this work the device is so small that it is very hard to make precise electrodes. The error from electrode displacement was about 15%.

Second, the gate-voltage bias dependence of Hall voltage was investigated at 300 K. When gate-voltage bias was changed from - 20 to - 60 V with applying magnetic field of 9 T, Hall voltage was decreased, where  $N_S$  is linearly increased from  $3.9 \times 10^{12}$  cm<sup>-2</sup> to  $1.1 \times 10^{13}$  cm<sup>-2</sup>.

This result is suitable to the fact that carriers are induced by the gate-voltage bias. In contrast,  $\mu_H$  decreased from 0.5 to 0.3 cm<sup>2</sup>/Vs as gate-voltage bias was changed from - 20 to - 60 V. The value of  $\mu_H$  was comparable to that of  $\mu_{FET}$ . This result indicates the validity of the  $\mu_{FET}$ , which is derived from the assumption that all the carriers induced by  $V_G$  can contribute to transport. It explains that



Fig.3 Inverse Hall coefficient (a) and Hall mobility (b) of pentacene OFETs as function of  $V_G$  at 300 K. (a) Black circles with dotted line are the inverse Hall coefficient, gray squares are longitudinal surface conductivity, and black broken line are electric field-induced charge assumed in a standard model of FET. (b) The change of Hall mobility. The dotted horizontal line shows the  $\mu_{FET}$  of reference device.

the majority of carriers in polycrystalline organic semiconductor by gate-voltage bias are delocalized.

In the Hall effect measurements using single-crystalline rubrene FETs, the electric charge  $eN_S(=1/R_H)$  calculated from Hall voltage was almost comparable to that calculated from gate-voltage bias (Q=CV<sub>G</sub>), indicating band-like transport [1].

On the other hand, in poly-crystalline pentacene FETs, the electric charge  $eN_s(=1/R_H)$  calculated from Hall voltage are two or three times larger than that calculated from gate-voltage bias where the gate-capacitance (C) is 6 nF/cm<sup>2</sup>. This result is reasonable as compared to the amorphous Si [1,4]. Considering the pentacene mean free path to be 0.08 ~ 0.3 nm and intermolecular distance to be 0.3 ~ 0.6 nm, the transport is dominated by hopping transport between grain boundaries rather than the free electron transport as the band conduction.

Finally, we investigated the temperature dependence of  $N_S$  and  $\mu_H$ . Measurements were performed at 270 K, 300 K, and 330 K. There is no significant change in  $N_S$ , while  $\mu_H$  increasing from 0.2 up to 0.3 cm<sup>2</sup>/Vs as temperature changes from 270 to 330 K. It is reasonable that  $\mu_H$  increases with temperature increasing where the carrier transport is ruled by hopping conduction. Still, further experiment is needed in the wide range of temperature change to understand transport mechanism in details.

#### 3. Conclusion

We have observed the Hall effect in the polycrystalline pentacene FETs manufactured on a plastic base film with polyimide gate dielectric layers. The electric charges evaluated from the Hall voltages were two or three times larger than that evaluated from  $V_G$ . This result clearly indicates transport mechanism of high-mobility pentacene FETs is dominated by carrier hopping rather than band-like transport. This experiment enabled us to understand the relation between  $\mu_H$  and  $\mu_{FET}$ , and to shed light on transport mechanism of polycrystalline organic semiconductor.

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