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Lateral Carrier Transport in Pentacene Polycrystalline Films –Hole Transport Barrier and Effective Mass in Crystals–

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1. Introduction

Field-effect mobility is one of the most important parameters in organic thin-film-transistors (OTFTs). In most cases, mobility is estimated from transistor characteristics of OTFTs according to the standard metal-insulator-semiconductor field-effect transistor model [1]. However, this method does not always provide us an intrinsic mobility of organic thin films because the characteristics of OTFTs are influenced by various effects originated in the electrode: for example, contact resistance at electrode/semiconductor interface, gate voltage dependence of carrier injection barrier, degradation of the films by electrode deposition in the case of the top-contact configuration [2], and degradation of crystallinity in the case of the bottom-contact configuration [3]. To exclude these problems, we developed a four-point-probe field-effect-transistor (FPP-FET) measurement system which can eliminate such effects and enables us to estimate an intrinsic carrier mobility of materials [4].

In this study, we have measured intrinsic mobility of various pentacene thin films by the FPP-FET measurements. From temperature and grain-size dependences of the mobility, we have calculated the hole transport barrier and the hole effective mass, which are practically important in disordered polycrystalline films.

2. Experimental

Figure 1 shows circuit diagram of FPP-FET measurement system [5]. The four-point probe is provided from *Kiyota Manufactuaring co*. The dependence of I_{14}/V_{23} ($V_{23}=V_{24}-V_{34}$) on V_G in linear region is described as,

$$\frac{I_{14}}{V_{23}} = \frac{\mu\pi C_i}{\ln 2}\left(V_G - \frac{V_{24}+V_{34}}{2} - V_T\right), \qquad (1)$$

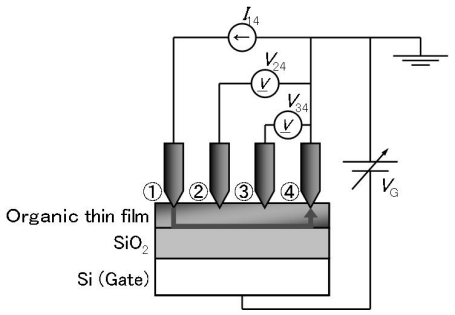

Fig. 1 Schematic of FPP-FET measurement system.

where μ is carrier mobility, C_i is insulator capacitance per unit area, and V_T is gate threshold voltage. According to eq.(1), we can estimate carrier mobility from the linear slope of the I_{14}/V_{23} vs V_G-(V_{24}+V_{34})/2 plot.

Thin-film transistors were fabricated on highly doped n-type Si wafers on which 300 nm silicon oxide was thermally grown. The highly-doped silicon acts as a gate electrode, and silicon oxide as gate insulator. Pentacene was deposited on the oxide by molecular beam deposition (MBD) under a vacuum of 2×10^{-8} Pa. We fixed growth rate at 0.3 nm/min, and varied substrate temperature in the range of 20–60ºC. As a result, we could prepare pentacene thin films with the grain size of 600 nm to 1.7 μm.

Measurements were carried out under a vacuum of 10^{-2} Pa and in dark. First of all, we measured the mobility at room temperature, and then measured temperature dependence in the range of 5–25ºC.

3. Results and Discussion

Figure 2 shows the grain size dependence of the mobility. Here, we define half of the apparent grain size observed by AFM as 'domain size', because, from a result of potential mapping within the grain measured by atomic-force-microscope potentiometory (AFMP), we found potential barriers which divide the apparent grain into four parts [3]. From this graph, the mobility is judged to be proportional to the domain size both at 23°C and 10°C.

Figures 3 (a) and (b) show a typical Arrhenius plot of the mobility and domain-size dependence of the activation energy, respectively. We can find that the Arrhenius plot shown in Fig.3(a) is almost linear. This means that the car-

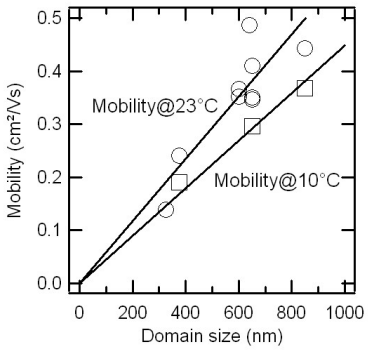

Fig. 2 Domain size dependence of mobility at 23ºC and 10ºC.

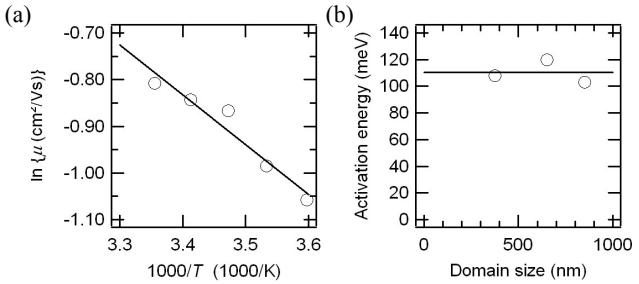

Fig. 3 (a) Arrhenius plot of the mobility in pentacene TFT (domain size: 850 nm) and (b) Domain size dependence of the activation energy.

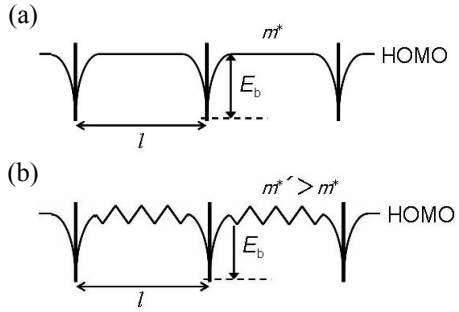

Fig. 4 (a) Band diagram of polycrystalline model and (b) new model including HOMO band fluctuation.

rier transport is thermally activated. We can also find that the activation energy shown in Fig.3(b) is independent of domain size. The average value of activation energy was estimated to be 110 meV.

The results above are well explained by 'polycrystalline model' which is proposed by Horowitz et al. for the field-effect mobility of oligothiophene [6]. In this model, only back-to-back Schottky barriers come up at grain boundaries limit carrier transport {Fig.4(a)}. And the carriers pass over these barriers by thermionic emission. Moreover, the transport in the grain is assumed to be equal to the one in an ideal single crystal. If the grain size is larger than double of the Debye length, carrier transport is proportional to the grain size. All the samples in this work satisfy this requirement. In such a case, carrier mobility is described as,

$$\mu = \frac{q\langle v\rangle l}{8kT}\exp\left(-\frac{E_b}{kT}\right), \qquad (2)$$

where l is the domain (grain) size, k is the Boltzmann constant, E_b is the height of back-to-back Schottky barrier, and $\langle v\rangle$ is carrier mean velocity. $\langle v\rangle$ is then described as,

$$\langle v\rangle = \sqrt{8kT/\pi m^*}\ , \qquad (3)$$

where m^* is the effective mass in the crystal.

Using eqs.(2) and (3), the hole effective mass in our pentacene films is calculated to be $1.55m_0$ (m_0 is mass of free electron). Here, we have to note that this is a mean value in randomly oriented crystals since pentacene is known to have anisotropic m^* even in the *ab*-plane, i.e. within the thin film. According to other reports of band calculation and experiment, the hole effective mass of pentacene film is estimated to be less than $1.26m_0$ [7,8]. So we conclude that this value of $1.55m_0$ is reasonable but slightly heavier than that in ideal crystals. This suggests that there is something which retards carrier transport even in the crystalline domain.

From recent AFMP experiment, it has been found that small potential fluctuation exists even in a crystalline domain [9]. We concluded that it is caused by deviation of carrier density originated from fluctuation of HOMO band. Increase of the effective mass is possibly due to this fluctuation. Based on the above discussion, we propose a new model which we should consider in pentacene polycrystalline films in Fig.4(b).

4. Conclusion

We carried out FPP-FET measurements to investigate carrier transport in pentacene polycrystalline films. According to the temperature and domain size dependences, the carrier transport in the pentacene films was well explained by the polycrystalline model. Then, we demonstrated that hole effective mass in crystalline domains can be estimated with electrical measurements. Obtained effective mass, $1.55m_0$, is reasonable but slightly heavier than that in an ideal pentacene crystal.

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