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Potential Fluctuation within a Crystalline Domain in Pentacene Thin Film Transistors and its Origin

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1. Introduction

Organic semiconducting materials are attractive in electronics applications because of their possibility of flexibility, low-cost and large-area fabrication. To utilize them as active materials of transistors, carrier mobility is an important factor because it limits operation frequency and output conductance.

Horowitz and coworkers reported, for first time, that the field-effect mobility in organic thin-film transistors (TFT) with polycrystalline sexithiophene increases in proportion to grain size [1]. Polycrystalline pentacene TFTs also exhibited the same tendency [2, 3]. Therefore, it is considered that mobility in a polycrystalline TFT is mainly limited by grain boundaries.

This phenomenon is explained by so called 'polycrystalline model' [1]. In this model, carrier transport rate is determined by thermionic current at grain boundaries, and overall mobility is described as follows:

$$\mu_{\text{FET}} = \frac{ql}{8kT} \cdot \sqrt{\frac{8kT}{\pi m^*}} \exp\left(-\frac{E_b}{kT}\right), \quad (1)$$

where q is the elementary charge, l is grain size, k is the Boltzmann constant, T is absolute temperature, m^* is effective mass within the grain, and E_b is barrier height at grain boundary. m^* is ideally considered to have a value in the single crystal. However, effective mass of $1.55 m_0$ (m_0 is the mass of free electron) has been estimated with polycrystalline pentacene TFTs by four-point-probe measurement [3], which is larger than that with a single crystal [4]. This implies that the effective mass within the grain is enlarged for some reason, and hence the field-effect mobility is also restrained by a certain in-grain property.

In this paper, we report potential and topographic profiles measured by atomic-force-microscope potentiometry (AFMP) [5] in the channel of a working polycrystalline pentacene TFT to examine the limiting factor of effective mass within a grain. Potential fluctuation observed within a grain is then analyzed to reveal its origin.

2. Experimental Details

A top-contact TFT was fabricated as follows. Heavily doped, thermally oxidized (300 nm in thickness) n-type Si was used as a gate-substrate. The substrate was exposed to UV/O₃. Further surface treatment was not used in this work. A pentacene film was then deposited by molecular

beam deposition (substrate temperature was kept at 67°C and deposition rate at 0.3 nm/min). Total thickness of the pentacene film was about 30 nm. Source and drain gold electrodes were finally deposited on the pentacene film.

Topographic and potential profiles in the channel region of TFT were obtained simultaneously by AFMP. Measurement details of AFMP are described elsewhere [5]. Gate-source voltage (V_{GS}) and drain-source voltage (V_{DS}) during the measurement were fixed at $V_{\text{GS}} = -40$ V and $V_{\text{DS}} = -10$ V, respectively. All measurements were performed in nitrogen atmosphere.

3. Results and Discussions

Figure 1 shows topographic and potential profiles in the working TFT. We measured these profiles to include a flat molecular terrace of a grain (within monolayer height, 1.5 nm, in roughness) to avoid measuring surface-structure-induced potential fluctuation which appears independently of the channel potential. In the distance range from 300 to 1600, we can see a molecular terrace. On the potential profile, small fluctuation is frequently seen even in the molecular-terrace region. The amplitude of fluctuation is around 19 mV_{RMS}, which is higher than noise level of 0.4 mV_{RMS}. We concluded that the potential fluctuation on the surface is correctly tracing the channel potential because there is no structural feature in this region and, in such a case, surface electric potential is mainly determined by local channel conductance where source-drain current concentrates. On the right side of point (a), the potential profile seems to be affected by the potential barrier of grain boundary. Therefore, we did not use this region for further analysis.

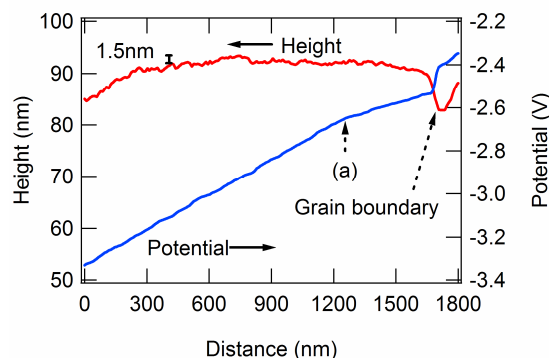


Fig. 1 Topographic and potential profiles taken to include a molecular terrace in an operating pentacene TFT.

We attempted to extract the conductivity in the channel using an equation:

$$\sigma(x) = J \cdot \frac{1}{E(x)} = J \cdot \frac{1}{dV_{DS}/dx}, \quad (2)$$

where J is current density in the channel and E is electric field. As an indicator of $\sigma(x)$, J is set to a constant value of one on the whole region. Figure 2 shows the indicator of conductivity profile in the distance range of 100-1200 nm. Gray band in Fig. 2 (a) stands for $\pm\Sigma$ (Σ is the standard deviation) obtained from a histogram in Fig. 2 (b). Most part of the conductivity indicator except for some abruptly higher points is included in the $\pm\Sigma$. Therefore, the histogram has a shoulder on the higher side of the peak. This sort of profile is considered to be originated in energy fluctuation of valence-band edge rather than energetic barriers caused by defects in the grain.

A drift current in the channel region with band edge fluctuation is described as follows:

$$J = \sigma(x)E(x) = q\mu N_v \exp\left(-\frac{E_f - E_v(x)}{kT}\right)E(x), \quad (3)$$

where N_v is effective density of state at valence band, E_f is the Fermi level, and E_v is energy level at valence-band edge. By rearranging eqs. (2) and (3), E_v is extracted as follows:

$$\ln(1/E(x)) \cdot kT = \left(\ln \frac{q\mu N_v}{J} \cdot kT - E_f \right) + E_v(x). \quad (4)$$

Figure 3 shows the extracted band fluctuation in the grain. In this figure, average energy level $\langle \ln(1/E(x)) \cdot kT \rangle$ was subtracted as a background. The band profile has become symmetrical about the energy axis, and follows a normal distribution. A histogram of random events usually follows a normal distribution. Therefore, this result suggests that the origin of potential fluctuation is the fluctuation of valence-band edge. Maximum amplitude of

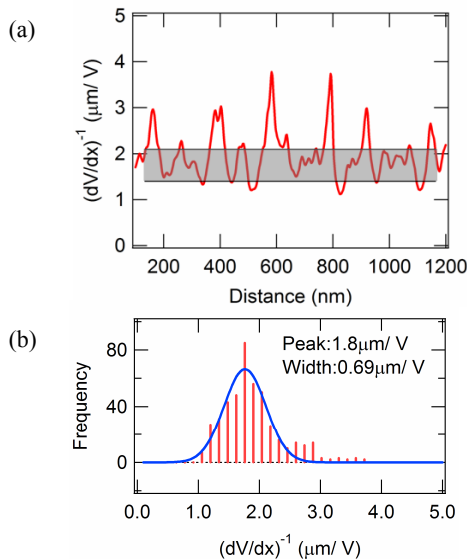


Fig. 2 Indicator of conductivity in a molecular terrace: (a) a profile and (b) a histogram.

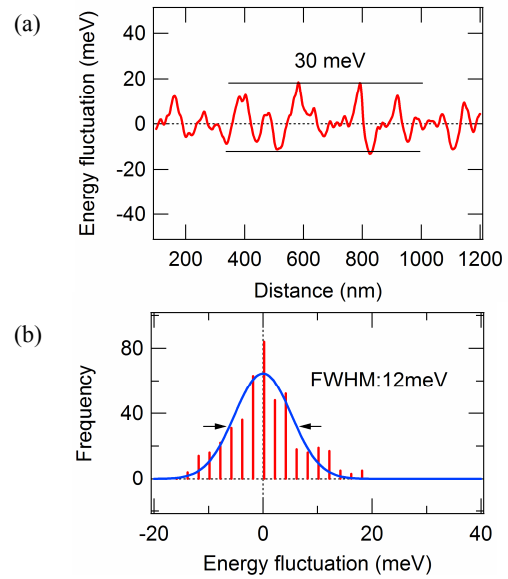


Fig. 3 Fluctuation of valence-band edge in a molecular terrace: (a) a profile and (b) a histogram.

30 meV and full width at half maximum (FWHM) of 12 meV was obtained. Here, maximum amplitude within the grain has to be much smaller than the barrier height of 110 meV at grain boundary [3] because the mobility of pentacene TFT shows thermally active behavior with a 110 meV barrier. The obtained result is consistent with this requirement.

If the band fluctuation is worked as shallow traps, carrier transport within the grain follows multiple-trapping and release mechanism [6]. As a result, m^* in eq. (1) would become larger than that in an ideal single crystal.

4. Conclusion

Potential fluctuation within a crystal domain was found for the first time by AFMP measurements on a flat molecular terrace of pentacene. Its origin was concluded to be the fluctuation of valence-band edge, of which FWHM is 12 meV. Overall mobility of pentacene TFT could be further improved by reducing this fluctuation.

Acknowledgement

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