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Extended Abstracts of the 2012 International Conference on Solid State Devices and Materials, Kyoto, 2012, pp1283-1284

Mobility Limiting Factors in Pentacene Thin-Film Transistors: Influence of the Film Growth Rate

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1. Introduction

In the past decade, to elucidate limiting factors of the field-effect mobility in organic thin-film transistors (OTFTs) has been one of big issue to realize higher mobility. Against this issue, we have studied the limiting factors quantitatively using our original techniques, Atomic-Force-Microscope Potentiometry (AFMP) [1], Four-Point-Probe Field-Effect measurement [2], and Grazing Incidence X-ray Diffraction using synchrotron radiation source [3]. As a result, we clarified that apparent mobility in pentacene polycrystalline film is described as follows [1],

$$\mu = \frac{ql_{\mathrm{D}}}{2k_{\mathrm{B}}T}\mu_{\mathrm{h}}\sqrt{\frac{qN_{\mathrm{A}}\phi_{\mathrm{b}}}{2\varepsilon_{\mathrm{S}}}}\exp\left(-\frac{q\phi_{\mathrm{b}}}{k_{\mathrm{B}}T}\right) \qquad (1)$$

where q is the elementally charge, l_{D} crystalline domain size, k_{B} the Boltzmann constant, T absolute temperature, μ_{h} mobility in crystalline domain, N_{A} acceptor density, ε_{S} dielectric constant of semiconductor, and $q\phi_{\mathrm{b}}$ height of carrier transport barrier at domain boundaries. Moreover, we also clarified that in-domain mobility μ_{h} is not equal to field-effect mobility of transistors using single-crystal active layer but decreased by HOMO-band-edge fluctuations that appear at crystallite boundaries within a crystalline domain [3]. Therefore, to enhance the mobility, we need to improve three factors by increasing crystalline domain size l_{D}, increasing in-domain mobility μ_{h}, and decreasing barrier height at domain boundaries $q\phi_{\mathrm{b}}$.

As a method to increase the mobility, chemical modification with HMDS or using polymer gate insulator have been most widely used technique. However, recently, we have reported that these techniques improve only crystalline domain size l_{D} [4] but do not vary the in-domain mobility and the carrier transport barrier. On the other hand, in the case of varying the pentacene film growth rate, a mobility difference is not always explained by the only domain size. In this work, we estimated the mobility limiting factors quantitatively when the film growth rate is varied, and discussed the influence of the film growth rate on the limiting factors.

2. Experimental

Top contact thin-film transistors were fabricated on highly doped n-type Si wafers on which 300 nm silicon oxides were thermally grown. The doped silicon acts as a gate electrode and the silicon oxide as a gate insulator. 30 nm-thick pentacene films were deposited on the substrate by vacuum evaporation after substrate cleaning. Substrate temperature during evaporation was kept at 20°C, and growth rate was changed for each sample (0.1 Å/sec, 0.5 Å/sec, and 1.0 Å/sec). 20-nm-thick source and drain gold electrodes were fabricated on the films by vacuum evaporation using metal shadow mask (channel length and width are 20 μm and 5 mm respectively).

AFMP measurements were performed using JSPM-5200 (JEOL). Measurement details are described in our previous report [5]. Gate-Source voltage (V_{GS}) and Drain-Source voltage (V_{DS}) during measurement were fixed at V_{GS}= -40 V and V_{DS}= -8 V, respectively. All AFMP measurements were carried out in Nitrogen atmosphere.

3. Results and Discussion

Figure 1 show AFM height images and transfer characteristics of each sample. Crystalline domain size

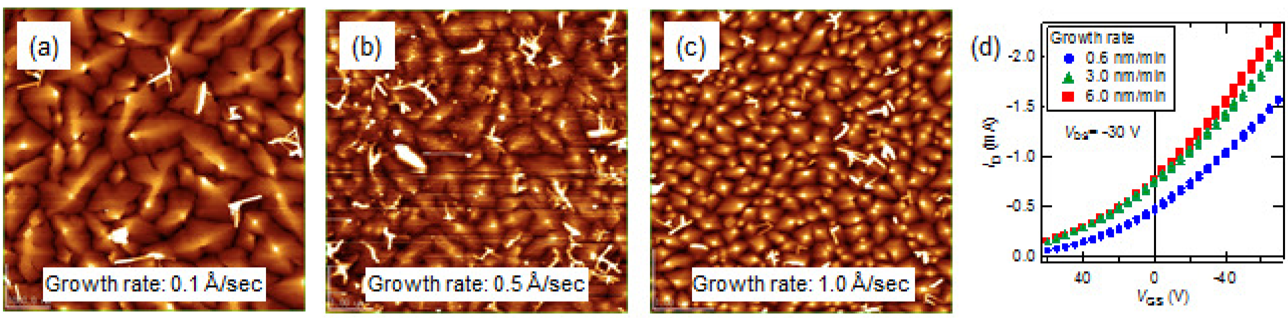

Fig. 1 (a)-(c) AFM height images and (d) transfer characteristics of each samples. In the AFM images, growth rate is (a) 0.1 Å/sec, (b) 0.5 Å/sec, and (c) 1.0 Å/sec, respectively.

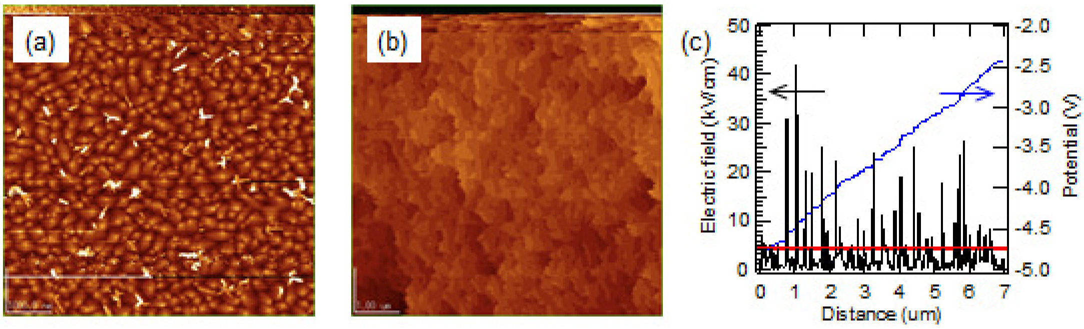

Fig. 2 Results of AFM potentiometry (growth rate is 1.0 Å/sec). (a) A height image, (b) a potential image, and (c) electric field and potential profile. The red line in (c) shows the average of electric field.

decreases with increasing growth rate. However, although the crystalline domain size decreases in almost half with increasing the growth rate, mobility does not decrease, but rather increase (0.20 cm^2/Vs for 0.1 Å/sec, 0.22 cm^2/Vs for 0.5 Å/sec, and 0.26 cm^2/Vs for 1.0 Å/sec). According to Eq. (1), this result means that in-domain mobility μ_h increased or carrier transport barrier $q\phi_b$ decreased. To distinguish

these effects, we defined relative conductivity that is ratio of conductivity at domain boundary (σ_{db}) to that within crystalline domain (σ_{di}), and compared the relative conductivity of each film.

Figure 2 shows a typical result of AFMP measurement. Fig. 2(a) shows a height image and Fig. 2(b) a potential image (average potential gradient is subtracted) at the same area with the height image, and Fig. 2(c) potential and electric field profile between source and drain electrode. Since it is difficult to recognize all crystalline domains from this scale image, we regarded an area where electric field is higher than average as a boundary region, and estimated relative conductivity using following formula,

$$\frac{\sigma_{db}}{\sigma_{di}} = \frac{E_{di}}{E_{db}} = \frac{\sum v_{di} l_{db}}{\sum v_{db}(l_D - l_{db})} \qquad (2)$$

where l_{db} is length of boundary region (=debye length, 160 nm in this work), v_{di} total potential drop of in-domain region between source and drain, and v_{db} that of the boundary region.

Table 1 shows a summary of obtained results in this work. By increasing the film growth rate, the crystalline domain size decreased, in contrast, relative conductivity increased. These facts mean that σ_{db} was increasing or σ_{di} was decreasing with increasing growth rate. However, since the apparent mobility increased with increasing the growth rate, it is reasonable to conclude that σ_{db} was increasing, that is, the height of carrier transport barrier at domain

Table 1 Summary of the results obtained in this work.

Growth rate (nm/min)	Domain size (nm)	Apparent mobility (cm^2/Vs)	Relative conductivity σ_{db}/σ_{di}
0.6	943	0.20	0.31
3.0	543	0.22	0.47
6.0	456	0.26	0.71

boundaries $q\phi_b$ can be decreased. Detail mechanisms have not been clarified yet, but we have found that film thickness around domain boundaries of fast grown film become thicker than that of slowly grown film. It can be considered that inter-domain connection is more conductive in the fast grown films that that in the slowly grown films.

4. Conclusions

We studied influence of film growth rate on the mobility in pentacene thin film using AFMP. Although crystalline domain size decreased, the mobility increased by increasing the growth rate. Considering the relative conductivity of in-domain and boundary region, we concluded that carrier transport barrier at domain boundaries can be decreased by increasing the film growth rate.

Acknowledgements

This work was partly supported by a Global COE program in Chiba University "Advanced School for Organic Electronics" and the Green Photonics Project in Nara Institute of Science and Technology.

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