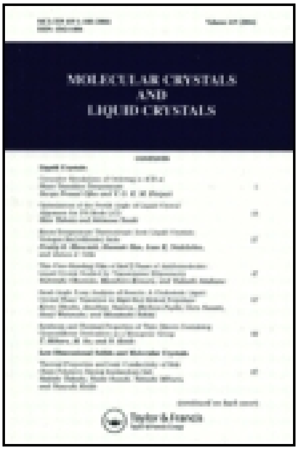

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Thickness Dependent Characteristics of a Copper Phthalocyanine Thin-Film Transistor Investigated by in situ FET Measurement System

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The influence of active layer thickness on transport characteristics of a copper phthalocyanine thin-film transistor was investigated by using the in situ FET measurement system with the film deposition continued up to several hundred nm in thickness. The drain current and mobility showed maximum values in the early stage of film growth and then decreased with the increasing film thickness. This result suggests that the over-grown layer affects the transport characteristics of the conductive accumulation layer, for example, owing to change of the electric field in the device.

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Keywords: copper phthalocyanine; *in situ* FET measurement; organic thin-film transistor; thickness dependence

INTRODUCTION

Organic thin-film transistors (OTFTs) are now attracting much attention due to their cost reduction, flexibility, large-area electronic applications, and less load on the environment [1–3]. With the aim of improving the TFT characteristics, many studies have been performed to understand their working mechanism, in particular, carrier injection by field effect. We made an *in situ* FET measurement system and revealed the thickness of accumulation layer and depletion layer in OTFTs [4,5]. There are some other reports of *in situ* conductivity measurements of OTFTs [6,7]. Including our researches, these studies have focused on the transport characteristics in the thin region of the organic active layer because it is considered that the region of a few or several nm just above the gate insulator is the most important for the conductivity. In the present study, we applied our *in situ* measurement system to investigate the thickness dependence of OTFTs up to much thicker regions beyond accumulation thickness. Hoshino *et al.* [8] prepared bottom-contact type copper(II)-phthalocyanine (CuPc) TFT specimens of various CuPc thicknesses. They showed that the mobility decreased with the increasing CuPc thickness after the thickness exceeded 80 nm. Using the different specimens for each thickness, the individual difference of the conductivity is feared, especially in OTFTs of which characteristics largely depend on the preparation process and measurement environment. Thus we applied our *in situ* measurement system to verify such thickness dependence of bottom-contact CuPc-TFTs up to several hundred nm using a single sample.

EXPERIMENTAL

Substrate Preparation

On a highly-doped n-type silicon substrate (0.01 Ωcm) with thermally oxidized surface (300 nm in thickness), source and drain electrodes of Cr (20 nm)/Au (30 nm) were prepared by photolithography. The electrodes had simple straight shape and channel length and width were 2 μm and 3 mm, respectively. The aim of such short channel is for the cross-sectional potential imaging which will be performed in the near future.

In situ FET Measurement

In situ FET measurement system [4,5] was used for investigating the thickness dependent transport characteristics. In vacuum chamber under a base pressure of 3×10^{-7} Pa, CuPc (purchased from Wako Pure Chemical Industries, Ltd. and purified two times by the vacuum sublimation technique) was evaporated from a Knudsen cell and deposited on the substrate with growth rate of 2 nm/min. The film thickness was monitored by the quartz crystal microbalance (QCM). Gate voltage (V_G) was changed repeatedly ($0 \rightarrow -25 \rightarrow -50 \rightarrow -75 \rightarrow -100 \rightarrow 0$ V) and drain current (I_D) was measured during the deposition of the CuPc film. Drain voltage (V_D) was kept constant (-10 V) throughout the experiment. Acquisition of I_D values was carried out 20s after the change of V_G.

RESULTS AND DISCUSSION

Figure 1 shows the variation of I_D during the deposition of the CuPc film. In the initial stage of the film growth, I_D sharply increased toward the film thickness of about 20 nm. The tendency of I_D variation after such thickness was different depending on V_G. At low V_G (-25 V), I_D gradually increased during deposition even in the thicker region. This suggests that conductivity is almost saturated when the transport path just above the CuPc/SiO_2 interface (accumulation layer) is

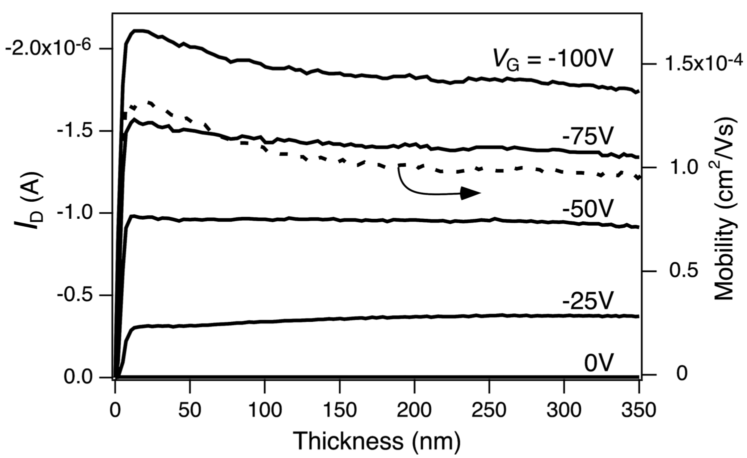

FIGURE 1 Variation of I_D during deposition of CuPc (solid lines). V_D was kept constant (-10 V) while V_G was repeatedly changed. The broken line shows variation of mobility estimated using the I_D data at $V_G \geq -50$ V.

completed, and small current through the upper (bulk) region is added to the major current with the increasing film thickness. At high V_G (-75 V and -100 V), on the other hand, I_D evidently deceased with the increasing film thickness.

Figure 2 shows the I_D-V_G relation at the thicknesses of 20 nm and 350 nm. In the thin stage of film growth (20 nm) I_D of low V_G region gradually increases and becomes almost linear at high V_G region. This trend is almost consistent with those of ordinary OTFTs. On the other hand, the slope ($\partial I_D/\partial V_G$) in the thicker stage (350 nm) clearly becomes smaller, in particular, at high V_G. This suggests the decrease of mobility with the increase of film thickness. The mobility at each thickness was roughly estimated using the I_D values at $V_G = -50$ V, -75 V and -100 V under the assumption of the linear regime [1,2], and the result was drawn in Figure 1. The mobility also decreases with the increasing film thickness. This tendency is similar to that reported in the previous study [8].

The most important problem is why mobility decreases with the increasing film thickness. If the quality of the film was damaged during measurement, I_D at low V_G (-25 V) should also decrease with the film thickness. In another work, we carried out the cross-sectional potential imaging of a working CuPc-TFT by Kelvin probe force

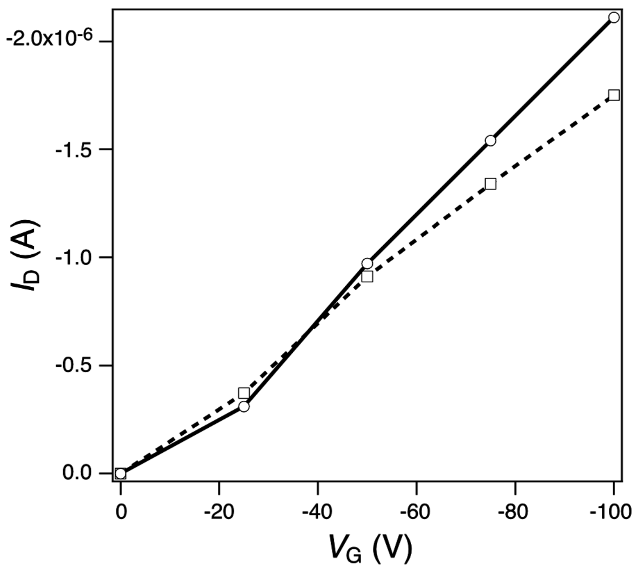

FIGURE 2 $I_D - V_G$ relation at the thicknesses of 20 nm (the solid line with open circles) and 350 nm (the broken line with open squares) when V_D was -10 V.

microscopy (KFM) and the results suggested that unexpected charge is induced also in grain boundaries in the CuPc film and such charge affects the conductivity at the CuPc/SiO_2 interface, when the CuPc film is thick [9]. The result of the present study suggests that the influence of such film thickness increases with the increasing V_G. Thus, there is possibility that the amount of the unexpectedly induced charge in the grain boundaries increases with the increasing V_G, and consequently changed electric field in the device reduces the conductivity.

In summary, the thickness dependent transport characteristics of CuPc-TFT, that is, the decrease of the carrier mobility, was verified by our *in situ* FET measurement system. I_D at high V_G decreased with the increasing film thickness though I_D at low V_G kept increasing gradually. This effect decreases the slope of $I_D - V_G$ relation ($\partial I_D/\partial V_G$) and the carrier mobility, particularly in high V_G region. The major factor that is behind this phenomenon is probably the injection of unexpected charge to the grain boundaries in the active layer and consequent change of the electric field in the device. The detailed mechanism of this effect and its universality are the subjects for future studies.

REFERENCES

[1] Dimitrakopoulos, C. D. & Mascaro, D. J. (2001). *IBM J. Res. Dev.*, *45*, 11.
[2] Dimitrakopoulos, C. D. & Malenfant, P. R. L. (2002). *Adv. Mater.*, *14*, 99.
[3] Newman, C. R., Frisbie, C. D., da Silva Filho, D. A., Jean-Luc Brédas, J.-L., Ewbank, P. C., & Mann, K. R. (2004). *Chem. Mater.*, *16*, 4436.
[4] Kiguchi, K., Nakayama, M., Fujiwara, K., Ueno, K., Shimada, T., & Saiki, K. (2003). *Jpn. J. Appl. Phys.*, *42*, L1408.
[5] Kiguchi, M., Nakayama, M., Shimada, T., & Saiki, K. (2005). *Phys. Rev. B*, *71*, 035332.
[6] Haddon, R. C., Perel, A. S., Morris, R. C., Palstra, T. T. M., & Hebard, A. F. (1995). *Appl. Phys. Lett.*, *67*, 121.
[7] Muck, T., Fritz, J., & Wagner, V. (2005). *Appl. Phys. Lett.*, *86*, 232101.
[8] Hoshino, S., Kamata, T., & Yase, K. (2002). *J. Appl. Phys.*, *92*, 6028.
[9] Ikeda, S., Shimada, T., Kiguchi, M., & Saiki, K. In preparation.