Accumulation and Depletion Layer Thicknesses in Organic Field Effect Transistors

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We present a simple but powerful method to determine the thicknesses of the accumulation and depletion layers and the distribution curve of injected carriers in organic field effect transistors. The conductivity of organic semiconductors in thin film transistors was measured *in situ* and continuously with a bottom contact configuration, as a function of film thickness at various gate voltages. Using this method, the thicknesses of the accumulation and depletion layers of pentacene were determined to be $0.9 \, \text{nm}$ ($V_G = -15 \, \text{V}$) and $5 \, \text{nm}$ ($V_G = 15 \, \text{V}$), respectively. [DOI: 10.1143/JJAP.42.L1408]

KEYWORDS: field effect transistor, accumulation layer, depletion layer, organic film

Electric-field control of physical properties is highly desirable from fundamental and technological viewpoints, because it does not introduce any chemical or microscopic structural disorder in the pristine material. ^{1,2)} This is also a basis of field effect transistors (FET), in which accumulation, depletion, and inversion layers are formed at the interface. "Where is the region, in which carriers are practically injected or depleted by electric fields?" is, thus, an important problem relating closely with the operation principle of FET and the electric-field control of physical properties. Although it is quite a fundamental issue, there had been no investigation on evaluating experimentally the dimension of these regions.

We propose a simple but powerful method to observe directly the regions in which the carriers are exhausted or injected by electric fields. As shown in the Fig. 1, the source (S)-drain (D) current in thin film transistors (TFT) is measured as a function of the film thickness with a bottom-contact configuration. The gradual change in the conductivity is measured *in situ* under the same condition at different gate voltages (V_G) avoiding the problem of specimen dependence which is often encountered in the organic films due to the difference in grain size, crystallinity, interface state, etc. This *in situ* and continuous measurement would provide information on the accumulation and depletion

GATE
Insulator
S Semiconductor D
Pentacene
Vacuum Vessel

Fig. 1. Schematic experimental layout. The source (S)-drain (D) current was measured *in situ* at various gate voltages.

layers, which will be described later.

Despite the above mentioned advantage, the *in situ* and continuous measurement had not been examined except for a few cases. The major reason is that higher substrate temperature is required to obtain a flat film for inorganic semiconductors, and hence the measurement is difficult to be applied. In contrast, a flat organic film can be obtained even at low substrate temperature, which enables the *in situ* and continuous measurement during growth of organic films.

In the present study, we applied the measurement to pentacene, and evaluated the thickness of the depletion layer and distribution curve of injected carriers in accumulation layer.

The substrate is a highly doped silicon wafer, acting as a gate electrode. The gate dielectric layer is a 700 nm thermally grown silicon dioxide. On top of the surface, 30 nm thick gold S, D electrodes were deposited through a shadow mask. The channel length and the channel width were 100 μm and 5.4 mm. The organic materials used were pentacene (Aldrich), sexithiophene ($\alpha\text{-}6T$; synthesized by Syncom BV), and C_{60} (Materials and Electrochemical Research). Organic films were deposited by means of vacuum deposition. The substrate temperature was kept at 310 K during the growth. All the measurements were performed under high-vacuum condition.

Figure 2 shows S-D current (I_{SD}) versus S-D voltage (V_{SD}) for the 20 nm thick pentacene TFT. Typical p-type semi-

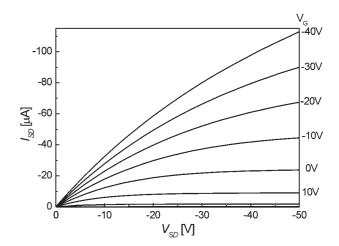


Fig. 2. Source (S)-drain (D) current ($I_{\rm SD}$) vs S-D voltage ($V_{\rm SD}$) characteristic of pentacene TFT at various gate voltages ($V_{\rm G}$).

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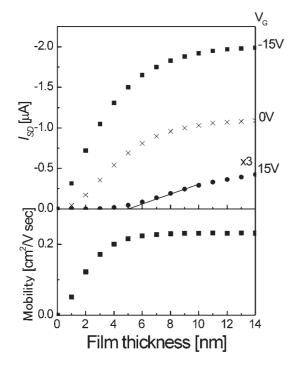


Fig. 3. a) Source-drain current ($I_{\rm SD}$) of pentacene measured as a function of film thickness at various gate voltages ($V_{\rm G}$); 15 (circle), 0 (cross), and -15 V (square). Source-drain voltage was kept at 1 V. b) Mobility as a function of film thickness.

conductor features are exhibited. We observed ohmic behavior of the S, D contacts to the pentacene active layer. The carrier mobility, estimated in the linear transport regime, was $0.23\,\mathrm{cm^2\cdot V^{-1}\cdot s^{-1}}$, which is comparable to the world's highest TFT mobility value. ⁶⁾

We then discuss the depletion layer thickness from the in situ and continuous measurement. V_{SD} was kept at 1 V and $V_{\rm G}$ was set at three different voltages; +15, 0, -15 V. Electric fields were not applied during the growth in order to exclude the influence of the electric current on the film growth. Figure 3 shows I_{SD} as a function of pentacene film thickness at different V_G . TFT characteristics were measured at each film thickness to obtain the mobility, and the thickness dependence of the mobility was included in the bottom part of Fig. 3. Here we find the presence of threshold thickness (d_{th}) at which electric current begins to flow. There are two points to note about d_{th} . First, d_{th} is 0.6 nm at $V_{\rm G}$ =0 V, and electric current is observed even for the 1.0 nm thick film. Considering the thickness (1.5 nm) of 1 monolayer (ML) pentacene, the present result indicates that the S, D electrodes (width 100 μm) can be electrically connected by only 1 ML pentacene. Second, $d_{\rm th}$ shows clear $V_{\rm G}$ dependence, and it shifts up to 5.0 nm at $V_G=15$ V. When positive V_G is applied to p-type semiconductors, the depletion layer with a low conductance is formed at the semiconductor/insulator interface. Therefore, on the thin limit, the whole film can be depleted, and I_{SD} does not flow at positive V_G . Consequently, d_{th} (5.0 nm) corresponds to the thickness of the depletion layer at $V_G=15$ V. By the in situ and continuous measurement, the thickness of the depletion layer can be observed directly as a shift of d_{th} .

In order to discuss the meaning of the depletion thickness, we will try to apply the conventional model which has been

already established for inorganic semiconductors. In the depletion approximation,^{3,8)} the carrier density is given by $N = Q/eT_{\rm S}$ where $Q = \varepsilon_{\rm ox}V_{\rm G}/T_{\rm Ox}$, $T_{\rm s}$: thickness of the depletion layer, ε_{ox} : dielectric constant of SiO₂, T_{ox} : thickness of SiO₂. Using T_S =5.0 nm at V_G =15 V, the carrier density amounts to $N=9.3 \times 10^{17} \,\mathrm{cm}^{-3}$. Considering the mobility (μ =0.21 cm²·V⁻¹·s⁻¹) at T_S =5.0 nm, the conductivity of pentacene is $3.1 \times 10^{-2} \,\Omega^{-1} \cdot \text{cm}^{-1}$ for the 5.0 nm thick film. This value is obtained on the basis of FET characteristics. On the other hand, the conductivity can be calculated from I_{SD} (6.9 × 10⁻⁷ A) for the 5.0 nm thick film at $V_G=0$ V, taking account of the channel width and the channel length. This yields the conductivity of $2.6 \times$ $10^{-2} \Omega^{-1} \cdot \text{cm}^{-1}$. The fair agreement between these two values indicates the validity of the above estimation, showing that the depletion layer of the organic TFT can be explained in terms of the conventional model applicable for inorganic semiconductors. Clear $V_{\rm G}$ dependence of $d_{\rm th}$ was observed also for other organic films, 6T and C₆₀. In case of 6T, d_{th} is 5.0 nm at $V_{\text{G}}=15\,\text{V}$, while the onset was not observed even for the 100 nm thick C_{60} film at $V_G = -15$ V. The thickness of the depletion layer is inversely proportional to the major carrier concentration of the semiconductors. Therefore, the onset was not observed for C_{60} with a low carrier concentration. Here, we comment the carrier concentration of the present specimens. We used as-received specimens without extra purification, and thus the impurity (carrier) concentration is rather high, resulting in a small on/ off ratio of Fig. 2. However, the high carrier concentration never affects the present estimation of depletion layer, instead helps observe the depletion layer as a delay of the onset at moderate condition (5 nm at $V_G=15 \text{ V}$), and assure the uniformity of electric field in the thin film thickness

In the previous section, we have discussed the thickness of the depletion layer. We now proceed to the accumulation layer, relating closely with the electric-field control of physical properties through charge injection. Here, we assume electric fields at the interface are determined only by $V_{\rm G}$, independently of film thickness, and hence the differentiation $(dI_{SD}(x)/dx)$ corresponds to the local conductance at the distance x from the interface. The carrier density (n(x)) can be approximated by dividing the local conductance by the mobility $\mu(x)$ (see Fig. 3) under the condition that the mobility is constant in the whole film at each film thickness. The carrier density thus obtained is shown in Fig. 4 as a function of the distance x from the interface. The large carrier density at $V_G=0$ V in small xregion is due to the charge transfer from the Au electrodes to the pentacene molecules. The density of the carriers injected by electric fields $n_i(x)$ is, thus, the difference between the carrier density $n_{-15}(x)$ at $V_G = -15 \text{ V}$ and $n_0(x)$ at $V_G = 0 \text{ V}$. As seen in the figure, $n_i(x)$ decays steeply with increasing x, meaning that the injected carriers are localized at the interface. The thickness of the accumulation layer is much smaller than that of the depletion layer, which is estimated to be 5.0 nm in the previous section. For quantitative estimation on the thickness of the accumulation layer, $n_i(x)$ is fitted with an exponential function $(a \times \exp(-x/b))$. The fitted value of b=0.91 nm can be considered as an effective thickness of the accumulation layer, which is just thinner than 1 ML. In other

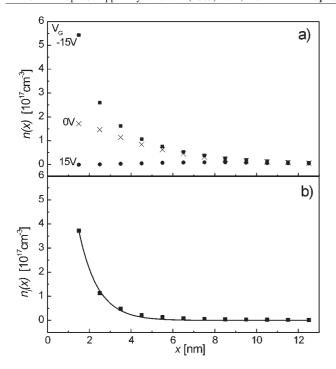


Fig. 4. a) Carrier density (n(x)) as a function of the distance (x) from the interface at various gate voltages (V_G) ; 15 (circle), 0 (cross), and $-15\,\mathrm{V}$ (square). b) The difference $(n_i(x))$ between charge carrier density at $V_G = -15\,\mathrm{V}$ and that at $V_G = 0\,\mathrm{V}$.

words, most of carriers are localized in the first ML next to the interface.

The obtained results on the accumulation and depletion layers can be a guide to the operation principle of organic TFT and electric-field control of physical properties by field induced charge injection. Since charge carries reside in the first one ML from the interface, the physical properties only at the interface could be controlled by the charge injection. This implies the importance of a well-ordered interface through which charges are efficiently injected. Furthermore, we propose that the semiconductor-metal or -superconductor

transition is a promising target to study. When the semi-conductor-metal transition occurs by charge injection, metallic and semiconducting regions sit side by side with only an atomic distance apart in the organic film. Under this situation, free carries can interact with exciton, associated with the semiconductor, at the interface, thus leading a possible ground for superconductivity by exciton mechanism. ^{9,10)}

In conclusion, we present the *in situ* and continuous measurement of the conductivity of growing organic films, as a simple but powerful method to determine the distribution curve of injected carriers and the dimension of the accumulation and depletion layers.

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- H. Ohno, D. Chiba, F. Matsukura, T. Omiya, E. Abe, T. Dietl, Y. Ohno and K. Ohtani: Nature 408 (2000) 944.
- C. H. Ahn, S. Gariglio, P. Paruch, T. Tybell, L. Antognazza and J.-M. Triscone: Science 284 (1999) 1152.
- S. M. Sze: Physics of Semiconductor Devices (John Wiley & Sons, New York, 1985).
- 4) The thickness of the depletion layer can be estimated indirectly by the capacitance measurement assuming the dielectric constant of semiconductors, but the thickness of the accumulation layer is hard to be determined.
- R. C. Haddon, A. S. Perel, R. C. Morris, T. T. M. Palstra, A. F. Hebard and R. M. Fleming: Appl. Phys. Lett. 67 (1995) 121.
- H. Klauk, D. J. Gundlach, J. A. Nichols and T. N. Jackson: IEEE Trans. Electron Devices 46 (1999) 1258.
- T. Komoda, Y. Endo, K. Kyuno and A. Toriumi: Jpn. J. Appl. Phys. 41 (2002) 2767.
- E. H. Rhoderick: Monographs in Electrical and Electronic Engineering, Metal-Semiconductor Contacts (Clarendon Press, Oxford, UK, 1978).
- 9) V. L. Ginzburg and D.A. Kirzhnits: *High-Temperature Superconductivity* (Consultants Bureau, New York, 1982).
- M. Kiguchi, R. Arita, G. Yoshikawa, Y. Tanida, M. Katayama, K. Saiki, A. Koma and H. Aoki: Phys. Rev. Lett. 90 (2003) 196803.