Effects of an Interface Dipole Monolayer on Pentacene Organic Field-Effect Transistors

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1. Introduction

Recently, there has been a remarkable interest in organic field-effect transistors (OFETs) because of their unique advantages [1]. For practical application in electronics, it is required to intentionally control all electrical properties, including mobility, threshold voltage, on/off current ratio, etc. [2]. Unlike the successfully developed doping method for silicon metal-oxide-semiconductor FETs, for OFETs, similar doping techniques have not yet been developed due to the ambiguities of energetics and dielectric nature of organic active layer. Thus another approach has been proposed to control carrier density or channel conductance using chemically modified self assembled monolayers (SAMs) [3]. Afterwards, an extensive study of the effect of SAM on threshold voltage was carried out [4-5]. Although these investigations greatly expanded the understanding of effect interface dipole monolayer, detailed study such as quantitative analysis is still difficult mainly due to the problem of precisely measuring dipole moment of the monolayer on insulator surface.

Hence, we proposed an alternative approach based on the use of Langmuir monolayer to overcome the difficulty. By recording molecular orientation change with the Maxwell displacement current (MDC) technique, the transient dipole moment change of monolayers under compression on water surface can be detected [6]. With the method, recently we found that adding divalent cations into water subphase can regulate dipole moment in normal of the amphiphilic molecules [7] and further investigation has shown electric field of deposited dipole monolayer greatly influences threshold voltage of pentacene OFETs [8]. Thus, it is expected to be a useful technique to control carrier behavior by tuning the built-in electric field of the dipole monolayer.

In this study, to investigate the electric field effect of interface dipole monolayer on OFET performance, aligned dipole monolayers with opposite orientation direction were introduced into pentacene OFET structure as an interlayer between pentacene and gate insulator using the Langmuir-Blodgett (LB) technique. It was found the dipole monolayer greatly shifted threshold voltage, mobility and on/off current ratio and these changes directly depended on the orientation of the monolayer.

2. Experiment

To investigate the effect of an interface dipole monolayer on OFET performance, we fabricated a top-contact structure with a dipole monolayer having different molecular orientation between organic active layer of pentacene (PEN) and gate insulator of SiO₂ or polyimide (PI), as shown in Fig. 1(a) and (b). For SiO₂ based OFET, we used a highly doped Si wafer with 100 nm thermally grown silicon dioxide (SiO₂) insulating layer as substrate, while for polyimide based OFET, we took a spin-coated polyimide layer on glass substrate having a thermal evaporated Au/Cr layer (the gate electrode). The thickness of polyimide gate insulator layer was around 500 nm measured by a profilometer (Dektak 3ST). For the dipole monolayer deposition, Langmuir film of dipalmitoylphosphatidylcholine (DPPC) was first prepared on an aquatic subphase in a manner as in ref. [7]. Then using the LB technique, the Langmuir film was transferred to the clean substrate at a surface pressure of 20 mN/m at room temperature. Note that the deposited monolayer on SiO₂ and PI substrate has different molecular orientation, as shown in Fig. 1(a) and (b), due to the different surface properties of the two kinds of substrates. After that, pentacene (from Tokyo Kasei Kogyou Co. Japan) film was evaporated onto the deposited DPPC layer in a high vacuum around 2×10⁻⁴ Pa. The layer thickness was regulated using a quartz crystal microbalance (QCM) to 100 nm with a deposition speed of 0.5 Å/s. The source and drain electrodes with a thickness of 50 nm were formed by thermal deposition of Au onto the pentacene layer at a pressure of about 5×10^{-4} Pa. The channel length and width were 80 µm and 3 mm, respectively. At the same time a reference sample without dipole monolayer was also prepared. The transfer characteristics were investigated by employing traditional current - voltage (I-V) measurement with two source meters (Keithley 2400). All measurements were carried out in ambient atmosphere within a dark box.

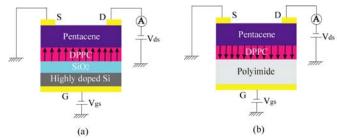


Fig. 1 Top-contact structure of pentacene FET with a dipole monolayer interlayer having opposite orientation direction between organic active layer pentacene and gate insulator of silicon oxide (a) and polyimide (b), respectively.

3. Results and discussion

Figure 2 shows a typical transfer characteristics of the OFET with and without the dipole monolayer. For monolayer orientated up in the case of SiO₂ OFET (see Fig. 1(a)), we observed a large threshold voltage shift to negative direction together with great decrease of effective carrier mobility (μ_{eff}) in presence of the dipole monolayer, as shown in Fig. 2(a). Note that the μ_{eff} evaluated from the transfer characteristics includes both effects of carrier injection and transport [9]. The large negative shift of threshold votage can be explained by our previously proposed model as shown in the following equation [8]

$$\Delta V_{th} = \frac{-(C_s + C_g)t_s}{\varepsilon_0 \varepsilon_d C_g} P,$$
(1)

where C_s and C_g are organic semiconductor (pentacene) and gate insulator capacitance, respectively, t_s is thickness of the semiconductor, ε_0 and ε_d are vacuum permittivity and relative permittivity of the dipole monolayer, respectively, and P is spontanteous polarization of the dipole monolayer whose value can be determined by the MDC technqiue. The impressively depressed μ_{eff} directly means the amount of mobile carriers are greatly suppressed since the estimated trapped charge density is nearly same. One reason for the observation is due to enhanced carrier injection barrier in the presence of electric field, which was confirmed by the increased contact resistance evaluated from the transmission line method (TLM). If so, we can expect increasing tendency of μ_{eff} in the presence of dipole monolayer with opposite (down) orientation direction. This expectation is confirmed by experimental results of polyimide OFET with downward orientation of a dipole monolayer (see Fig. 1(b)), as shown in Fig. 2(b). Except the prominent increse of $\mu_{e\!f\!f}$, a remakable positive threshold voltage shift is observed, which is opposite to the previous case (Fig. 2(a)). This positive shift due to the downward orentation of dipole monolayer can be also expected from Eq. (1). Additionly, the on/off current ratio is two orders less $(10^7 \text{ to } 10^5 \text{ for without and with the dipole monolayer})$ for SiO₂ based pentacene OFETs and one order higher for polyimide based pentacene OFETs $(10^3 \text{ to } 10^4)$, respectively. Note that these opposite changing tendency of OFET properties are directly related to the orientation of interface dipole monolayer.

4. Conclusions

In order to understand the effect of interface monolayer on performance of organic devices, we studied pentacene OFET with a dipole monolayer having opposite molecular oreientation between pentacene and gate insulator of SiO_2 or polyimide. It showed all main parameters, threshold voltage, effective mobility and on/off ratio, were greatly influenced by the introduced dipole monolayer and the change tendency was directly related to the molecular orientation. These observations depending on molecular orientation of the interface dipole monolayer reveal the possibility of control carrier behavior in organic devices.

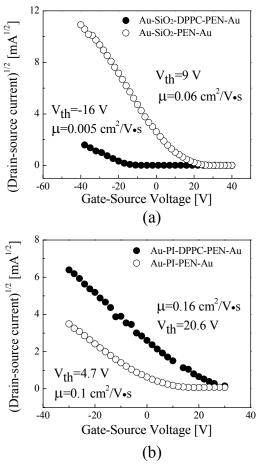


Fig. 2 Transfer characteristics of pentacene OFET with and without an interface DPPC dipole monolayer between organic active layer pentacene and gate insulator of silicon oxide (a) and polyimide (b), respectively.

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