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In-situ study of *pn*-heterojunction interface states in organic thin film transistors

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ABSTRACT

In this paper, we have investigated the density of *pn*-heterojunction interface states by evaluating the threshold voltage shift with in-situ measurement of electrical characteristics of a sandwich fluorinated copper phthalocyanine/pentacene thin film transistor with various thicknesses of pentacene thin films. A threshold voltage (V_T) undergoes a significant shift from +20.6 to +0.53 V with increasing the thickness of pentacene. When the thickness of pentacene is more than a critical thickness of 15 nm, V_T undergoes hardly any shift. On the other hand, the value of mobility is lightly decreased with increasing the thickness of pentacene due to the effect of the bulk current. Thus the V_T shift is attributed to the increase of drain current in the sandwich device. In order to explain the V_T shift, a model was assumed in the linear region of thin film transistor operation and the V_T shift agrees with a $\tan^{-1}$ function of film thickness. The total charge density (Q_0) of 1.53×10^{-7} C/cm^2 (9.56×10^{11} electrons or holes/cm^2) was obtained. Furthermore, the V_T shift and Q_0 could be adjusted by selecting a *p*-type semiconductor.

1. Introduction

Organic thin films have been widely investigated as active layers in electronic and optoelectronic devices, such as light-emitting diodes (LEDs), thin film transistors (TFTs), and photovoltaic (PV) cells [1]. The use of more complex structures than just an organic thin film is needed in many organic devices. Several organic layers of different materials are used in organic LEDs, organic ambipolar TFTs or organic PV cells, where basically a *pn*-heterojunction allows the effective transport of electrons and holes towards or away from the organic–organic interface [2–7]. Similarly, additional organic layers as electron or hole blocking layers are often used to improve the performance of these devices. Especially, the study of the electronic structure of organic *pn*-heterojunction is both of technological and fundamental importance for understanding current and future electronic devices built from organic semiconducting molecules.

In the previous studies, we have reported a kind of organic heterojunctions, in which the high density of carriers formed a conduction channel at an interface [8–12]. Ambipolar transport is dependent on the first active layer thickness (F_{16}CuPc, fluorinated copper phthalocyanine) in a F_{16}CuPc/CuPc (copper phthalocyanine) heterojunction, and only *n*-channel operation is observed when F_{16}CuPc films are over a critical thickness of ~12 nm. Utilizing the heterojunction the *n*-channel device performance is significantly improved and threshold voltage is suppressed. Furthermore, only a threshold voltage shift is observed when F_{16}CuPc films are over a critical thickness of ~15 nm, where the heterojunction is away from the *n*-channel conduction modulated by gate voltage. In this study, we report on density of *pn*-heterojunction interface states by evaluating the threshold voltage shift with in-situ measurement of the electrical characteristics of a sandwich F_{16}CuPc/pentacene TFT with various thicknesses of pentacene thin films.

2. Experimental details

Fig. 1 shows a sandwich device configuration. First, a top-contact F_{16}CuPc TFT was fabricated. A heavily *n*-doped Si substrate acts as the gate electrode with a 300 nm thermally grown SiO_2 layer (C_i ~ 10 nF/cm^2) as the gate dielectric. F_{16}CuPc thin films of 20 nm were vacuum-deposited and the substrate temperature was set at 120 °C. Au source and drain electrodes of approximately 50 nm were vacuum-deposited through a shadow mask with a channel width of 5 mm and a length of 70 μm. Then, pentacene thin films were vacuum-deposited on the above-mentioned F_{16}CuPc device at room temperature. The characteristics of the device were in-situ measured with a two-channel voltage current source/monitor system (R6245, Advantest) at pentacene film thicknesses of 0, 1, 2, 3, 5, 7, 10, 15 and 20 nm controlled by a shutter. All films were deposited under a base pressure of less than 1×10^{-3} Pa, and thicknesses and growth rates were monitored by a thickness and rate monitor (CRTM-6000, ULVAC). F_{16}CuPc and pentacene were purchased from Aldrich and purified by gradient sublimation before being used.

3. Results and discussion

Fig. 2a shows the output characteristics of a sandwich device with 0 nm thick pentacene thin films (namely, the top-contact F_{16}CuPc

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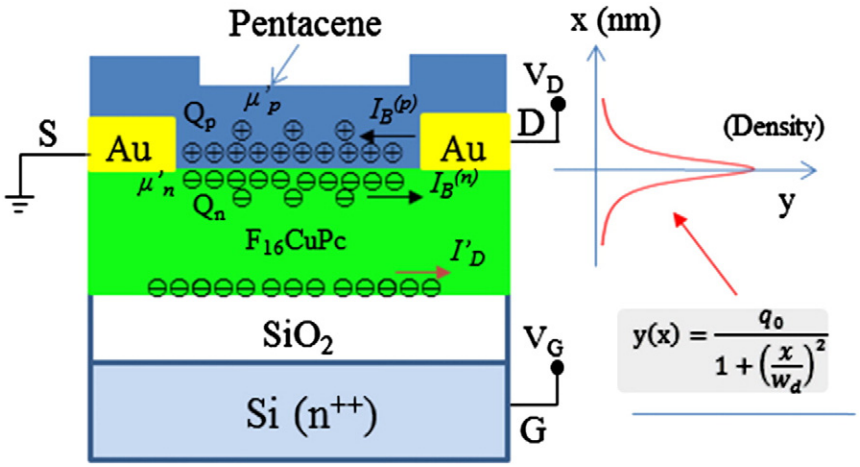

Fig. 1. Schematic cross sections of a sandwich device (the dimensions are not scaled). A model is given and assumes a parallel conduction mechanism consisting of: 1) a conduction channel in which the carrier density in the conduction accumulation layer modulated by V_G is equivalent to $I'_D = \frac{W}{L}\mu_n C_i(V_G - V_T)V_D$, and 2) a '*pn*-heterojunction interface' layer away from the above-mentioned conduction channel, whose mobility carrier density is not modulated by V_G: $I_B^{(p)} = \frac{W}{L}\mu'_p V_D Q_p$; $I_B^{(n)} = \frac{W}{L}\mu'_n V_D Q_n$. The charge distribution in the *pn*-heterojunction interface layer can be indicated by a Lorentz function: $y(x) = q_0 / \left[1 + \left(\frac{x}{w_d}\right)^2\right]$, where q_0 and w_d are the *pn* interface charge density and the full width at half maximum, respectively.

TFT), which typically works in an *n*-channel operational mode. The linear and saturation regions can be observed with the increases of drain voltage (V_D) and gate voltage (V_G). Fig. 2b shows the same for the sandwich device when 20 nm thick pentacene thin films were deposited. We observe a dramatic difference in the shape and magnitude of the drain current (I_D). In the sandwich device I_D is up to 15.8 μA at V_G and V_D of 60 V and does not saturate. Furthermore, a large bulk current (I_B) in the sandwich device at a V_G of 0 V could also be observed, which originates from the charge carriers at the interface of F_{16}CuPc/pentacene [6–12].

Fig. 3 shows the transfer characteristics of the sandwich TFT with various thicknesses of pentacene thin films at $V_D = 10$ V. With increasing the thickness of pentacene, I_D is remarkably increased. μ_n and V_T were extracted in the linear region as follows [13]:

$$I_D = \frac{W}{L} C_i \mu_n (V_G - V_T) V_D \ (V_G \gg V_D), \tag{1}$$

where W, L, and C_i are the channel width, channel length, and gate dielectric capacitance per unit area, respectively. The dependence of μ and V_T on the thickness of pentacene is shown in Fig. 4. Obviously, V_T undergoes a significant shift from +20.6 to +0.53 V with increasing the thickness of pentacene. When the thickness of pentacene is more than a critical thickness of 15 nm V_T undergoes hardly any shift. On the other hand, the values of μ_n is lightly decreased with increasing the thickness of pentacene due to the effect of the I_B. Thus the V_T shift (ΔV_T) is attributed to the increase of I_D in the sandwich device.

It has been known that I_B results from the organic *pn*-heterojunction interface dipolar, and the electrons and holes are free charges, which are significantly distinguished from ionized impurities, localized in the space charge region for conventional inorganic *pn*-junctions, as simply drawn in Fig. 1 [14,15]. In the context of the following model, the *pn*-heterojunction interface charge density can explain the ΔV_T in the linear region of TFT operation. The model assumes a parallel conduction mechanism consisting of: 1) a conduction channel in which the carrier density in the conduction accumulation layer is modulated by V_G, equivalent to Eq. (1), and 2) a '*pn*-junction interface' layer away from the above-mentioned conduction channel, whose mobility carrier density is not modulated by V_G. Thus I_D can be expressed as follows:

$$\begin{aligned} I_D &= I'_D + I_B^{(n)} + I_B^{(p)} \\ &= \frac{W}{L}\mu_n C_i V_D (V_G - V_T) + \frac{W}{L}\mu'_n V_D Q_n + \frac{W}{L}\mu'_p V_D Q_p, \end{aligned} \tag{2}$$

where μ'_n = field-effect mobility of F_{16}CuPc away from the conduction channel, μ'_p = field-effect mobility of pentacene, Q_n = charges

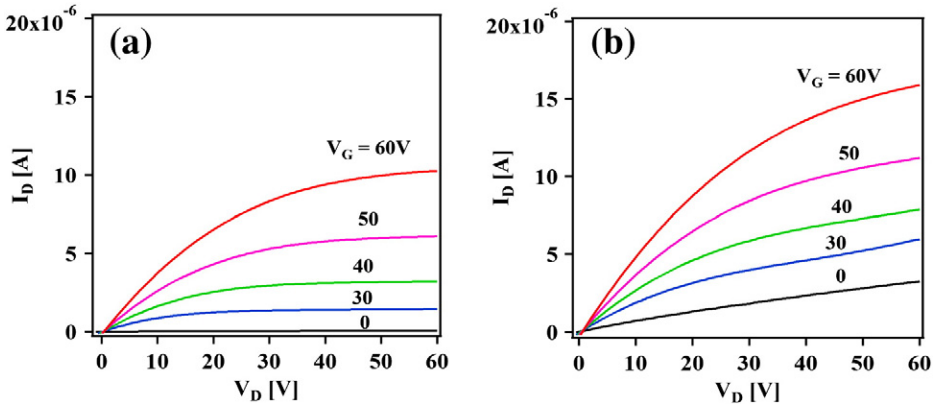

Fig. 2. Output characteristics of a sandwich F_{16}CuPc/pentacene TFT with pentacene thin film thicknesses of (a) 0 nm and (b) 20 nm, respectively.