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Organic Single Crystal Transistors and Interface Control

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1. Introduction

Organic electronics has been a subject of active research for over a decade now, with applications emerging in light-emitting displays and printable electronic circuits. Particularly, organic field effect transistors (FETs) exhibit great promise as fundamental element devices for low cost and flexible electronics such as displays, radio-frequency identification tags, and mechanical or chemical sensors. [1] Despite a substantial body of work aimed at understanding charge transport in these materials, a well-developed, microscopic description is still lacking. This type of fundamental understanding is crucial for further improvement of device performance and search for novel functionalities of the OFET devices. For this purpose and to show the physical limit of organic materials, single crystal FETs have played major roles in these several years, since they are free from grain boundaries which are dominant limiting factors for practically useful polycrystalline thin film FET devices.

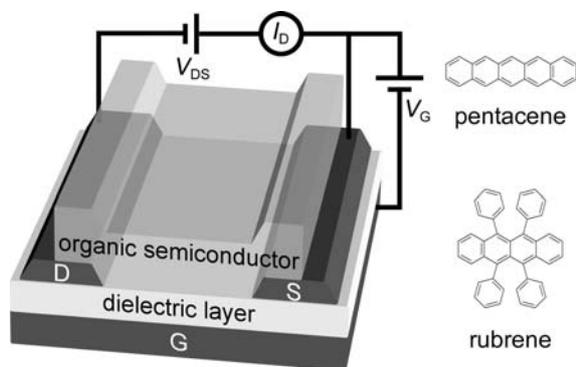


Figure 1 Schematic structure of an organic FET device with a bottom gate configuration, and organic molecules for semiconductors.

Figure 1 displays a schematic view of organic FETs. Organic molecules used for single crystal FET studies are also shown in Fig. 1. Pentacene is well known as a molecule which enables one to fabricate the most reliable thin film transistors with highest mobility, and thus is the most promising material for practical applications [2]. Rubrene is used mainly for basic studies in the single crystal form and displays the highest mobility above $20 \text{ cm}^2/\text{Vs}$ among organic materials [3].

An important difference between organic FETs and the conventional metal-oxide-semiconductor FETs of Si single crystals is that the organic semiconductors are used without intentional doping. In sharp contrast, the state-of-the-art

doping technique controls the device performance, not only for Si but also other inorganic semiconductors, such as GaAs. Because of the absence of such detailed doping technique in organic semiconductors, a possible way of controlling device performance is the interface modification.

In this paper we review the current status of the study on organic single crystal FETs and basic understandings on the mechanism of charge injection and accumulation in organic FETs, by demonstrating an ambipolar operation of organic transistors.

2. Experimental results and discussions

Single crystals of polyacene organic semiconductors such as pentacene and rubrene were grown by a physical vapor deposition method [4]. Configurations of organic single crystal transistors are basically classified into two; one is Insulator on Organics and the other is Organics on Insulator (Fig. 2). In the former (left of Fig. 2) case, developed by Pozorov et al., an organic gate insulator (parlylene in most cases) is grown on top of an organic single crystal after evaporating source and drain electrodes [5]. Finally, a gate electrode is also thermally evaporated on the parlylene insulator. In the latter configuration, first reported independently by de Boer et al., and Takeya et al., thin crystallites are laminated on the pre-patterned substrates (right of Fig. 2), to which crystallites adheres by electrostatic force [6,7]. A notable advantage of the latter is that the device fabrication process such as patterning can be made without organics, providing a large amount of degrees of freedom in the device structure, including the compatibility with thin film transistors. In fact, the highest mobility among organic FETs has been realized in the air-gap devices based on the Organics on Insulator configuration [3]. In this study, we also adopted the Organics on Insulator configuration.

Transfer and output characteristics of a rubrene OFET with gold source and drain electrodes are shown in Fig. 3,

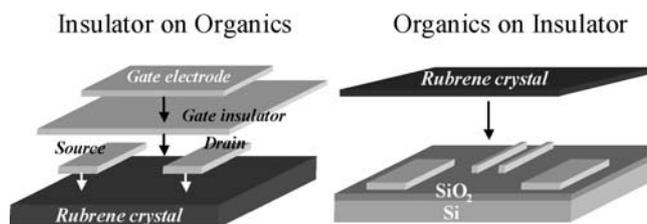


Figure 2 Fabrication processes for two types of organic single crystal FETs.

displaying typical p-type behaviors. The mobility derived from the slope of transfer curve (left of Fig. 3) was about $2 \text{ cm}^2/\text{Vs}$. The values larger than $1 \text{ cm}^2/\text{Vs}$ were routinely observed in rubrene single crystal devices.

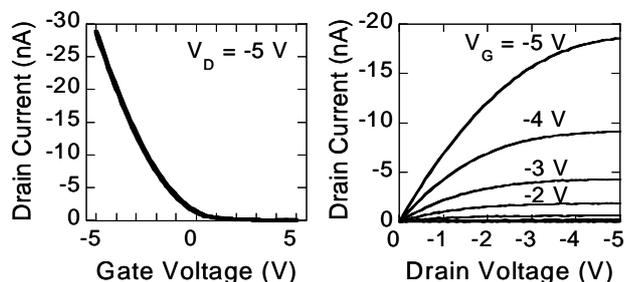


Figure 3 Transfer (left) and output (right) characteristics of rubrene single crystal transistor fabricated on SiO_2 gate dielectrics and gold source-drain electrodes.

Since this rubrene crystal is not intentionally doped, the mechanism of p-type operation or a mechanism of carrier sign determination should be a very fundamental issue to be understood. For the carrier conduction in the FET channel, the carriers should be injected and the channel should be trap-free. Considering the work function of gold and rubrene, hole injection is much easier than electron injection, supporting the p-type operation in Fig. 3. However, by employing an interface modification technique, we succeeded in making an ambipolar transistor with rubrene single crystals [8].

So far, no electron current has been observed in rubrene or pentacene, when SiO_2 dielectrics are used as gate-insulators, suggesting the trap-state is formed at the interface between organic semiconductors and SiO_2 . A possible origin of the trap-states is the hydroxyl group that terminates the SiO_2 surface, acting as electron traps [9]. To avoid the trap-states, we inserted a PMMA (polymethylmethacrylate) buffer layer between the SiO_2 surface and the rubrene crystal. The transfer characteristics of a rubrene single crystal FET with a PMMA buffer gate dielectric, as

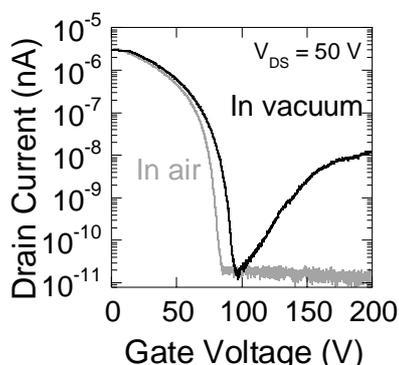


Figure 4 Transfer characteristics of rubrene single crystal FETs with PMMA coated gate and silver paste electrodes. Electron current is observed in vacuum (black line), whereas only the hole enhancement is seen in air (gray line).

shown in Fig. 4, displayed both hole- and electron-enhancement modes. Hole and electron mobilities were derived from the saturated current as $1.8 \text{ cm}^2/\text{Vs}$ and $1.1 \times 10^{-2} \text{ cm}^2/\text{Vs}$, respectively. The occurrence of ambipolar operation is possibly understood in terms of removal of electron traps by the organic polymer coating. In addition, an air-instability of electron carriers was observed in this device (Fig. 4). Such an extreme sensitivity to air exposure is a generic feature of n-type organic FETs.

Another key issue to achieve the electron current is to apply high source-drain voltages. Since the output characteristics for the electron injection is highly nonlinear in contrast to the linear behavior for the hole-injection, the ambipolar characteristic in Fig. 4 was observed only when the high source-drain voltage is applied (In the particular case of Fig. 4, $V_{DS} = 50 \text{ V}$ was applied). All these data related to the ambipolar rubrene FETs provide various clues on the carrier injection/accumulation mechanisms, in other words, transistor operation mechanisms.

3. Conclusions

An ambipolar FET operation was demonstrated in rubrene single crystal FET, which has been known as a p-type semiconductor, using a PMMA buffer layer as a gate insulator. This provides novel opportunity to understand the origin and mechanisms for electron traps in organic semiconductors. Also, an injection mechanism of electrons from gold electrode with a large work function should be urgently understood. These interface controls in organics FETs are crucial for further improvement of device characteristics as well as for search for novel properties of devices, such as light emitting transistors.

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