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Keywords: Organic transistor; Organic single-crystal transistor; Carrier injection

On July 30th 2015, the Editor-in-Chief sent a letter to the corresponding author of the above-mentioned manuscript [1] about the result of survey by the Editorial Committee of e-J. Surf. Sci. Nanotech. According to the letter, the Committee concluded that the above-mentioned manuscript resembles the paper published in another journal [2] too much.

As a result of this report, all of the authors of the above-mentioned manuscript have agreed to a complete retraction of the paper.

Organic Single-Crystal Transistors with Secondary Gates on Source and Drain Electrodes

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Rubrene and tetracyanoquinodimethane single-crystal transistors are fabricated incorporating secondary gates (split gates) on source and drain electrodes to reduce the interfacial barriers at the metal/semiconductor contacts. Separating the effect of the injection barriers, the intrinsic carrier transport in the semiconductor channels is extracted for the p-type rubrene crystal transistors and the n-type tetracyanoquinodimethane crystal transistors. The transconductance of the tetracyanoquinodimethane devices is drastically improved by activating the split-gate electrodes, indicating significant injection barriers in the n-type transistors. The result demonstrates that the technique is useful to improve transistor performance when it is restricted by the injection barriers.

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I. INTRODUCTION

To achieve the maximum device performance of organic field-effect transistors (OFETs), a still challenging subject is to realize efficient carrier injection at the electrodes [1]. The contact performance is seriously concerned for such devices as short-channel FETs with the channel length typically less than sub-micrometers, though they are highly attractive because of their high-frequency response and capability of high-density integration. The problem of the contact barriers appear to be serious also for most of n-type organic transistors when injecting electrons at the Fermi level of the electrode up to the lowest unoccupied molecular orbital (LUMO) of the semiconductor molecules. In order to study the effects of the interfacial contacts systematically in such devices of particular technological importance, the method of controlling injection rate in the same devices is desired. In this work, we develop a device structure with “split gates” on the source and drain electrodes buried in the gate-insulating layers, so that the carrier density in the organic semiconductors can be varied separately in the vicinity of the electrodes independently of the primary gate electric fields to modulate the central semiconductor channels.

II. EXPERIMENTAL

A. Device preparation

Figure 1(a) shows the whole design of the split-gate single-crystal. We first deposit the split-gate electrodes on 500-nm thick SiO$_2$/doped silicon substrates; 5-nm thick chromium and 15-nm thick gold are deposited by the vacuum evaporation technique and are patterned by photolithography. Cross-linked polyvinylphenol (PVP) dielectric is prepared to the thickness of 2-3 µm for the second gate dielectric layer, and source and drain electrodes are formed to the size slightly smaller than the split-gate electrodes by photolithography, so that the split gates extend to the semiconductor channel in the vicinity of the edges of the source and drain electrodes. Finally, a thin platelet of organic crystal is laminated by natural electrostatic force, as already described elsewhere [2–6]. The organic crystals of rubrene and tetracyanoquinodimethane (TCNQ) were independently grown by physical vapor transport [7]. Figures 1(b) and (c) show the cross section and a top-view picture of a product device. Typically the width $W$ and length $L$ of the channel are 100 µm and 30 µm, respectively. The distance between the electrodes $L_{\text{single}}$ is set as $\sim 35$ µm. The permittivity of the cross-linked PVP is defined to be $\sim 4.0$ as the result of our AC impedance measurement [8].

B. Measurement method

The transistor performance is measured with three source measure units (SMUs) equipped in a Keithley 4200 semiconductor parameter analyzer: one applies the source-drain voltage $V_D$ and measured the drain current $I_D$, another applies the primary gate voltage $V_G$ to the doped Si, and the other applies the secondary gate voltage $V_G^{\text{split}}$ to the split gates buried in the insulating layer. To detect the influence of the split gates, we compare results of the following two operation modes.

1. Operation of the split-gate FET mode

We measure the transfer characteristics sweeping the primary gate voltage $V_G$ applied to the doped silicon layer, with fixed $V_G^{\text{split}}$. For convenience, we denote “conductivity” $\sigma^{\text{split}}$ and mobility $\mu^{\text{split}}$ of this mode as

$$\sigma^{\text{split}} = \frac{L_{\text{split}}}{W} \frac{I_D}{V_D}, \quad \mu^{\text{split}} = \frac{1}{C_i} \frac{\partial \sigma^{\text{split}}}{\partial V_G},$$

using the combined capacitance $C_i$ of the PVP and SiO$_2$ layers per area.
2. Operation of the single-gate FET mode

We measure the transfer characteristics sweeping both $V_G$ and $V_{\text{split}}$ concomitantly so that the resultant gate field is identical in the whole region of the length $L_{\text{single}}$ between the source and drain electrodes [see Fig. 1(b)], i.e.

$$V_GC_I = V_{\text{split}}^C_{\text{PVP}},$$

(2)

where $C_{\text{PVP}}$ is the capacitance of PVP per area. To compare with the result of the above split-gate operation mode, we denote “conductivity” $\sigma_{\text{single}}$ and mobility $\mu_{\text{single}}$ of this mode as

$$\sigma_{\text{single}} = \frac{L_{\text{single}} I_D}{W V_D}, \quad \mu_{\text{single}} = \frac{1}{C_I} \frac{\partial \sigma_{\text{single}}}{\partial V_G}.$$  

(3)

III. RESULTS AND DISCUSSIONS

Plotted in Fig. 2 are transfer characteristics of two rubrene single-crystal transistors comparing the results of the single-gate FET mode and the split-gate FET mode. $V_{\text{split}}^C$ is fixed to $-100$ V for the latter. Note that the values of mobility differ even among similarly prepared crystals depending on difference in surface quality of the PVP gate insulators. For the higher-mobility sample, only threshold voltage $V_{\text{th}}$ is slightly shifted so that the steepest slope is preserved in the transfer characteristics as the results of single-gate and split-gate modes are compared in Fig. 2(a). The result of the minor effects of the split gates suggests that good ohmic contacts are formed at the rubrene/gold interfaces with sufficient carriers accumulated in the channel. It is suggested that the hole injection is relatively easy because the highest occupied molecular orbital (HOMO) level of rubrene 5.2 eV is lower than the work function of Cr/Au which is $\sim 5.1$ eV [9]. The observation also means that the experiment is performed properly as we intended.

Figure 3 shows the result of the same experiment for the TCNQ device. Electrons are accumulated with the application of positive gate voltage this time. $V_{\text{split}}^C$ is fixed to $+100$ V for the split-gate FET mode. Obviously, the slopes of the two curves differ with each other, resulting in different values of mobility as evaluated by the standard formula of Eqs. (1) and (3). The curve of the singlet-gate FET shows subthreshold region in relatively broad range of $V_G$ up to $\sim 30$ V, where the enhancement rate of the channel conductivity gradually changes with the gate voltage. On the other hand, the slope of the transfer curve reaches the maximum (corresponding to $\mu \sim 0.1$ cm$^2$/Vs) in the same range of $V_G$ for the split-gate FET. Since the middle of the channel is identical for the two measurements, the subthreshold in the singlet-gate FET is not merely caused by interface states in the channel. Therefore, it turned out that the injection barriers provoke broadening of the subthreshold regime and significantly reduce the transistor mobility in the present TCNQ device. It is suggested that the electron injection may not be easy because the lowest unoccupied molecular orbital (LUMO) level of TCNQ 4.8 eV [10] is lower than the work function of Cr/Au.

The application of the split-gate voltage fills the interface states so that the injection barrier is reduced. We note that the split-gated region with the overdoped electrons conceptually resembles $n^{++}$ contact region which facilitates electron injection in Si-MOSFETs. It is also suggested that mobility values can be more properly eval-
FIG. 2: Transfer characteristics of two rubrene single-crystal split-gate transistors with different mobility values. Blue and red curves show the results of split-gate and single-gate operation modes, respectively. See text for the details.

FIG. 3: Transfer characteristics of a TCNQ single-crystal split-gate transistor. Blue and red curves show the results of split-gate and single-gate operation modes, respectively. See text for the details.

uated with the use of the split-gate structure for organic semiconductor materials whose mobility is underestimated because of the problem of the injection barriers. Though the reason of the slight shift of $V_{th}$ was not clear in Fig. 2(a) for the rubrene transistor, it is qualitatively understandable assuming the same mechanism as we considered for the TCNQ device; the subthreshold regime due to the interface states at gold/rubrene contacts is reduced with the application of the split-gate voltages.

IV. CONCLUSIONS

We have shown the effect of the secondary gates on source and drain electrodes to reduce carrier injection barrier and to improve the device performances. As $p$-type rubrene crystal transistors and $n$-type TCNQ crystal transistors are compared, the effect is more pronounced in the latter device. It turned out that the interfacial states at the TCNQ/gold contact are effectively filled with the application of the split-gate voltage, reducing height of the injection barrier. The experiment demonstrated that the split-gate structure is useful to extract more intrinsic carrier mobility of the semiconductor channels and to improve the device performances for such devices.

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