Study of Electron Injection of Pentacene Field Effect Transistor with Au Electrodes by C-V and SHG Measurements

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Using pentacene field effect transistors (FETs) with Au source and drain electrodes, electron injection from the Au electrodes into the pentacene was investigated. The capacitance-voltage (C-V) and optical second harmonic generation (SHG) measurements were employed. Electron injection from the Au electrodes was suggested by the hysteresis behavior with the C-V characteristics and slowly decaying SHG signal under DC biasing. A mechanism of hole-injection assisted by trapped electrons is proposed. To confirm electron injection process, light-emitting behavior under the application of AC applied voltage was observed.

Keywords : Pentacene FET, Maxwell-Wagner model, SHG, Trapped charge

1. INTRODUCTION

In order to drive the field effect transistor (FET), we need to apply a drain-source voltage V_{ds} and a gate bias voltage V_{gs} on the order of several 10 volts[1]. As such, charge carriers forming the conducting channel of pentacene FETs with Au source and drain electrodes are mainly holes injected from the Au source electrode[2,3]. However, the application of such high positive voltage suggests us the possibility of electron injection into pentacene, even though high work function Au metal electrode is used as source and gate electrodes. Recent intensive study on organic FETs (OFETs) has revealed the presence of ambipolar behavior in many organic materials used for OFETs[4,5]. Beyond that, ambipolar behavior has been found out in many dielectric materials, including insulating polymers, etc[6]. Up to now, without using semiconductor device physics, we have shown that the pentacene FET is analyzed as a Maxwell-Wagner effect element[7-9]. This means that pentacene can be treated as dielectric materials rather than as semiconductor materials in the analysis of the performance of the OFET. These facts motivated us to study electron injection into pentacene from metal electrodes with high work function. In this study, using pentacene FETs with Au source and drain electrodes, electron injection from the electrodes into the pentacene FET was examined by capacitance-voltage (C-V) and optical second harmonic generation (SHG) measurements[10,11]. Note that electric field induced SHG (EFISHG) is effectively used in the SHG measurement[12,13]. Results revealed that electrons are injected from the Au source and drain electrodes and they are subsequently trapped in the pentacene. Based on these results, a mechanism of hole-injection assisted by trapped electrons in the pentacene is discussed in terms of the hysteresis behavior with C-V curves. In this study, electron injection from the Au electrodes into pentacene was confirmed from the light-emitting behavior from the pentacene layer of the OFET under AC biasing.

2. EXPERIMENT

Figure 1 shows the device configuration of top-contact (TC) pentacene FET used here. Before deposition of the pentacene active layer, the SiO_2 gate insulator was subjected to the UV/ozone and octadecyltrichlorosilane ($C_{18}H_{37}SiCl_3$) (OTS) chemical treatments. The UV/ozone and OTS treatments were performed in the same manner as that done in our previous study[10]. Finally, we deposited Au electrodes (= 80 nm) as source and drain electrodes. Their channel length L and width W were, respectively, 50 μm and 1.25 mm. We masked the FET sample, except the channel and electrode regions during the deposition to avoid unnecessarily carrier diffusion. Using a source meter (type-2400; Keithley Instruments, Inc.) and an LCR meter (type-3522-50; Hioki E.E. Corp.), C-V measurements were carried out in laboratory ambient conditions where source and drain electrodes were

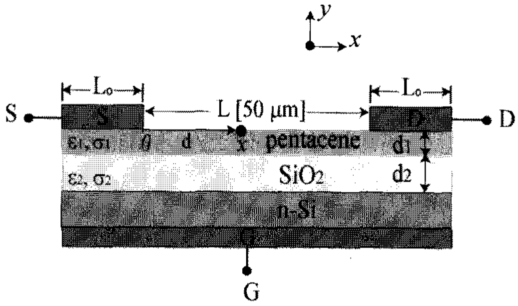

Fig. 1. Schematic cross-sectional view of TC pentacene FET.

electrically shorted (V_{ds} = 0). For SHG measurements, the experimental system was used as in ref.[12]. Briefly, using reflection optical geometry, fundamental light was focused on the channel region of the OFET using a microscopic objective lens (50 ×, NA=0.42, WD=20.5 mm, M Plan Apo SL; Mitsutoyo Co. Ltd.). The spot size was approximately 20 μm for microscopic SHG measurement. The light source was an optical parametric oscillator (OPO, Surelite OPO; Continuum Inc.) pumped by the third-harmonic light of a Q-switched Nd-YAG laser (Surelite II-10; Continuum Inc.). The wavelength was fixed at 1120 nm to probe the resonance enhancement of the EFISHG process from the pentacene, eliminating the generation of SH from gate-SiO_2 layer. SHG measurement is a spectroscopic method, and it can selectively probe the SHG signal enhanced from pentacene subjected to a local electric field. The modulation of the electric field along the FET channel by injected carriers was examined using the microscopic SHG measurements. The SH light from the sample was detected using a photomultiplier tube (PMT, R3896; Hamamatsu Photonics KK). In this configuration, the polarization direction was chosen as that along the channel between the source and drain electrodes, i.e., in the *x*-direction (see Fig. 1). All SHG measurements were performed in the laboratory's ambient atmosphere. The EL intensity from the OFET was also monitored with PMT under the application of AC voltage to confirm electron injection.

3. RESULTS AND DISCUSSION

In Fig. 2, the C-V characteristics show a hysteresis behavior that depends on gate-source (drain) stress biasing, V_{gs} (V_{gd})[11], where source and drain electrodes are electrically shorted. Results suggest that the hysteresis behavior is attributable to injected carrier trapping in the FET channel from the electrodes, because the possibility of carrier injection from gate-SiO_2 to pentacene can be tentatively discarded in our pentacene FETs. As is well known, the presence of trapped carriers in OFETs gives rise to a threshold voltage V_{th} shift in its C-V characteristics. In more detail, Fig. 2 shows that hole trapping is initiated by stress biasing, V_{gs} (V_{gd}) = -40 V for 30 min, and this leads to a shift of C-V characteristics with respect to V_{gs}. The C-V of the backward direction shifted to a negative V_{gs} in comparison with that of the forward direction. This result indicates that hole injection is suppressed after $V_{gs}<0$ stress biasing due to hole-trapping. Note that the increase of capacitance with respect to V_{gs} appears in the region $V_{gs}<0$, suggesting that hole trapping is main origin. On the other hand, interestingly, capacitance increases during the forward sweep in the region $V_{gs}>0$. That is, hole injection is assisted even in the region $V_{gs}>0$. This result suggests that electron injection and subsequently trapping must occur after stress biasing $V_{gs} = +40$ V for 30 min. From these, we argue that electron injection is a possible carrier injection process from the Au source (drain) electrode into pentacene by stress biasing. To confirm the electron injection and trapping into pentacene by $V_{gs}>0$ stress biasing, SHG measurement was carried out. Injected carriers are excess charge Q_s that form a space charge field along the FET channel. To confirm the presence of injected electrons into pentacene, we examined the SH enhancement from the pentacene FET channel carefully under electrically shorted biasing condition $V_{gs}=V_{gd}= +100$ V (electron injection condition judged by the C-V measurement).

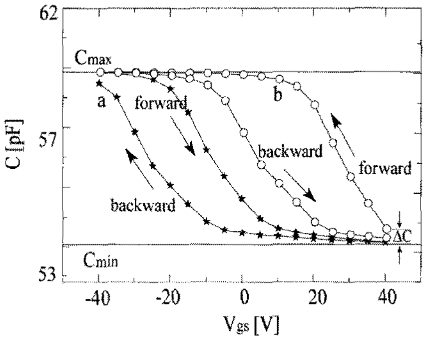

Fig. 2. C-V characteristics in the region between V_{gs}=-40 V and +40 V. Curves a (full stars) and b (open circles) were taken before applied V_{gs}=-40 V and V_{gs}=+40 V for 30 min. The scan direction is indicated by arrows for the forward and backward sweeping directions.

On fixing the SHG probe above the petnacene FET, we examined the SH enhancement with elapsed time. Curve 2 in Fig. 3 shows the SHG enhancement with time, which was monitored on fixing a incidence laser beam position at a position from 10 μm from drain electrode (see inset Fig. 3). As carrier injection relaxes an electric field formed in FET channel before carrier injection, SHG intensity decay suggests electron injection. The SHG enhancement gradually decays with a response time of c.a. 1000 sec. As the dielectric relaxation time τ representing spreading time of charges in dielectric materials is defined by $\tau=\varepsilon/\sigma$, such slow decay of SHG does not indicate the increase of sufficient number of electrons in the FET channel that contribute electronic conduction; that is, electron spreading over the FET channel is difficult. Here, ε is dielectric constant of pentacene and σ is conductivity. We argue that the response time of 1000 sec reflects the electron trapping process, as has been confirmed by the hysteresis behavior in the C-V characteristics for $V_{gs}(V_{gd})>0$ stress biasing. As the reference, response of SH with $V_{gs} = V_{gd} = -100$ V (hole injection condition judged by the C-V measurement) was shown as curve 1. The response time is very short and SHG signal is not observed to nearly zero, that is, injected holes spread over entire region of the FET channel more quickly comparison with electron injection process.

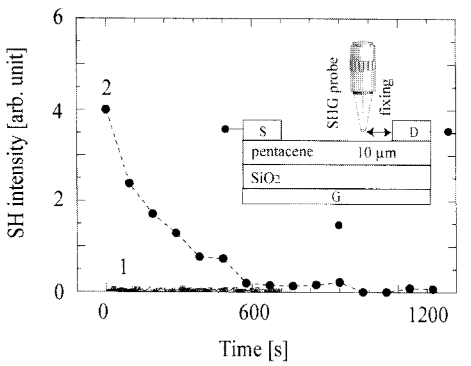

Fig. 3. Results of microscopic SHG measurement vs. biasing time. Curve 1 shows the on-state with $V_{gs} = V_{gd} = -100$ V and curve 2 is the off-state with $V_{gs} = V_{gd} = +100$ V. Insert figure represents observation schematically for the pentacene FET with the SHG measurement.

We also carefully examined the decay of SH signal profile along the FET channel using different biasing time with particular emphasis on the trapped electron situation. Our microscopic SHG experiments on moving SHG probe revealed the decay of SHG with position-dependence (not shown here). At the initial state, two peaks depending on the Laplace field appeared along the FET channel in a manner as we described in our previous paper[10], but it gradually decays with biasing time. In greater detail, the I_{SHG} near the Au drain electrode is higher than that near the source electrode, depending on the scanning direction from the drain to the source electrode. The SHG decrease occurs because of the formation of an electric field that arises from electrons trapped near the Au source and drain electrodes and it is reasonably interpreted as the increase of trapped electrons with biasing time. In this way, at $V_{gs}= V_{gd}= +100$ V stress biasing, trapped electrons are generated around the source and drain electrodes. Therefore, we might argue that the hole injection from Au source and drain electrodes into pentacene is assisted by the electric field formed by already trapped electrons around the Au source and drain electrodes during the sweep of V_{gs} in the forward direction from $V_{gs}=+40$ V to -40 V in the C-V curves in Fig. 2. That is, the situation of electrons trapped near the Au source and drain electrodes enhances the hole injection from the electrodes during the forward sweep of V_{gs}. Consequently, holes are more smoothly injected from Au source and drain electrodes and they are accumulated at the pentacene FET channel, even though $V_{gs}(V_{gd})>0$. The capacitance increase was observed in the forward direction for curve b in Fig. 2 and it is explainable based on this model. On the basis of our discussion mentioned above, we further confirm electron injection from the Au electrodes by the light emitting behavior from the pentacene FETs. Electron-hole recombination leading to electroluminescence (EL) is expected to occur from pentacene FETs with Au source

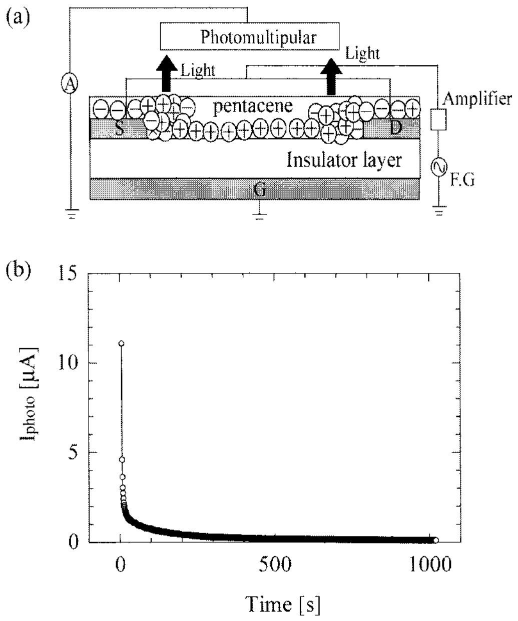

Fig. 4. (a) Set up for the electro-luminescence measurement with pentacene FET and (b) output electroluminescence characteristics.

and drain electrodes in the presence of hole and electron at the same time, in a manner as that seen in tetracene FET by Hepp et. al[14]. For this purpose, we apply AC voltage so that we can inject electrons and holes alternately into pentacene FETs. The time- dependence of the EL intensity was monitored with PMT under the application of AC bias with a frequency of 500 kHz. The measurement was performed in a laboratory ambient atmosphere. Figure 4 shows the preliminary experimental result of time-dependence of pentacene EL intensity under AC electric field. The EL intensity decreased monotonously as time elapsed and it increased in proportion to AC frequency up to 500 kHz. This result indicates that electrons and holes are alternately injected into the pentacene and generates the EL caused by their recombination, where electrons and holes injected from the Au electrodes into materials are main origins. There are two possible mechanisms of EL, i.e., one is an intrinsic process in which electrons and holes recombine directly, and the other is an extrinsic process in which both carriers recombine via trapping states[15]. For Fig. 4, we are still not clear, but we may conclude electron injection is a possible process of pentacene FET with Au source and drain electrode. However, further investigation is needed to clarify the details. Note that we recently developed an advanced SHG measurement. That is, the time-resolved SHG measurement (TRM-SHG) that enables us to directly probe and visualize carrier motion in organic materials[16]. Our preliminary TRM-SHG experiment supported our result shown in Fig. 3, and illustrated smoothly hole injection from Au electrodes into pentacene on the order of nano-second scale, whereas it illustrated very difficult situation for electron injection. This time-Resolved SHG experiment will be of help for further understanding of carrier injection process of organic thin film transistors.

4. CONCLUSION

On the basis of C-V and modulated SHG signal results, we discussed electron injection process into pentacene from the Au TC-electrodes. Hysteresis behavior caused by injected carrier trapping suggested a possibility of electron trapping by $V_{gs}(V_{gd})>0$ biasing. Based on this result, a mechanism of hole injection process assisted by trapped electrons was proposed. Moreover, C-V and SHG measurements revealed electron injection that followed by trapping. Finally, preliminary EL luminescent behavior from the pentacene FET under AC biasing showed the clear evidence of electron injection from the Au electrode.

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