Study of carrier transport in flexible organic field-effect transistors: Analysis of bending effect and microscopic observation using electric-field-induced optical second-harmonic generation

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Abstract

We studied the effect of bending on the carrier transport properties of flexible 6,13-Bis(triisopropylsilylethynyl)-pentacene (TIPS-pentacene) field-effect transistors. Results showed that the effective carrier mobility increased ~30 % with a 1.5 % mechanical strain (compressive stress), whereas it decreased ~15 % with a -1.5 % strain (tensile stress). Theoretical analysis based on the Maxwell-Wagner model was carried out, and suggested that both carrier mobility and carrier density in the organic field-effect transistor (OFET) channel were modulated due to the mechanical strains. The microscopic electric-field-induced second harmonic generation (EFISHG) images showed that carrier transport was governed by the presence of grains of TIPS-pentacene. The EFISHG observation is a powerful tool to investigate carrier transport in flexible OFETs which are being subjected to mechanical strains.

Keywords: flexible organic field-effect transistor, Maxwell-Wagner effect, electric-field-induced optical second-harmonic generation, carrier transport

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1. Introduction

Organic field effect transistors (OFETs) have attracted considerable attention since the discovery of conducting organic semiconductors [1]. As a result, development of advanced organic semiconductor materials, improvement of device fabrication process and so forth have provided OFETs with a carrier mobility >1 cm^2/Vs [2,3]. Organic semiconductors have a lot of advantages. Among them is so-called mechanical flexibility, which opens the way to electronics applications such as flexible displays and so forth. Sekitani et al. [4] and Chen et al. [5] experimentally studied the effects of mechanical strains (~1.5 %) on the electrical properties of pentacene FETs prepared on flexible substrates, e.g., polyethylene naphthalate (PEN) and metal foils. These studies suggested that the gate capacitance of OFETs decreases almost linearly with increasing compressive strains, whereas the field effect mobility increases linearly with the increasing compressive strains. The changes of the capacitance were on the order of a few %; the same order of the magnitudes of the applied strains. However, they have not derived theoretical formula that is available for the evaluation of their experimental data. The author's group has been analyzing the effect of applied strains on the charge accumulations and transports in OFETs, and derived the formula that express effective mobility of OFETs in terms of applied strains [6]. On the other hand, we have been developing an experimental tool based on the optical second harmonic generation measurement [7-10]. The measurement is capable of directly probing carrier motions in the OFET channels. Therefore, by employing the electric-field-induced second harmonic generation (EFISHG) measurement together with the analysis based on the derived formula, we can fully understand the bending effect on the carrier transport in the flexible OFETs. In this paper, on the basis of our theoretical background, we studied the bending effect which appears in the carrier transport in flexible 6,13-Bis(triisopropylsilylethynyl)-pentacene (TIPS-pentacene) OFETs, fabricated on PEN substrate by means of the time-resolved EFISHG measurement.

2. Experiment

Top contact TIPS-pentacene FETs were fabricated on the PEN substrate with a thickness of 300 μm, which can be bended by applying mechanical stress. The device structure was as portrayed in Fig. 1. As a gate insulator, a 600 nm-thick polyimide was spin coated on the gate electrode, onto which TIPS-pentacene was deposited as an active layer. Here the TIPS-pentacene was deposited by the direct-write process using a hollow pen [11], which resulted in an active TIPS-pentacene layer with relatively huge grain size. The mechanical strains applied to the TIPS-pentacene FET were 1 %, 1.5 % (compressive strain) and -1 %, and -1.5 % (tensile strains) (see Fig. 2(b)) by placing the device on half-cylindrical blocks with a curvature of ±10 mm, and ±15 mm. For measuring the mechanical strain effect on the transfer characteristics, the prepared TIPS-pentacene FETs were placed on these blocks alternating (see Figs. 2(a) and (b)).

The TIPS-pentacene OFETs were characterized using the transfer curves. Here gate voltage (V_g) was swept from 30 V to -30 V with reference to the source electrode, on keeping drain voltage V_{ds} = -30 V. The TIPS-pentacene FETs showed p-type transfer characteristics, from which the effective mobility was calculated. In order to study the effect of bending on the carrier transport, mechanical strains were applied to the OFETs in a way as mentioned earlier. The effective carrier mobility of the OFETs with or without bending was estimated. After that, results were analyzed on the basis of the Maxwell-Wagner effect model [6].

In order to directly observe the carrier transport in the TIPS-pentacene FETs, the time-resolved EFISHG measurement was carried out [7-10]. The EFISHG process is the third-order nonlinear optical process, and the intensity of the SH signals is proportional to the square of the nonlinear optical polarization $P(2\omega)$ induced in the TIPS-pentacene layer. The polarization is given in a form as

$$P(2\omega) = \chi^{(3)} \vdots E(0)E(\omega)E(\omega)\,. \qquad (1)$$

Here $\chi^{(3)}$ is the third-order nonlinear optical susceptibility, and $E(0)$ and $E(\omega)$ are the static electric field and the electric field of laser light, respectively. That is, the SHG intensity is in proportion to the square of this static electric field, $I_{2\omega} \propto |P_{2\omega}|^2 \propto |E(0)|^2$. In OFETs, by applying a voltage to the gate electrode in reference to the source electrode, carriers are injected, accumulated, and then transported along the FET channel at the semiconductor/insulator interface. These carriers are the source of electric fields, and induce SH signals. Noteworthy that the transient carrier transport is visualized by the time-resolved microscopic SHG (TRM-SHG) measurement [7-10]. In the measurement, pulsed laser beam supplied from an optical parametric oscillator coupled with Nd:YAG laser (average power 0.5 mW, repetition rate 10 Hz, duration 4 ns), was irradiated through an objective lens (×20), and focused onto the FET channel region. The irradiated laser beam was polarized in the FET channel direction. Accordingly, the laser beam couples with the electrostatic field in the channel direction formed by injected charges, and thus radiates SH signal. The SH signals are collected using an objective lens, and SH images were recorded on a cooled CCD camera, whereas the reflected pulsed laser beam from the OFET was blocked by using optical filters equipped with the CCD camera. Figure 3a portrays optical arrangement for the TRM-SHG measurement. A pulsed voltage is applied to the source electrode in reference to the gate and drain electrodes. The laser pulses were irradiated on the channel region after applying the pulse voltage with delay time t_d (see Fig. 3(b)). Further it should be noted that $\chi^{(3)}$ is a material dependent parameter and is a function of ω. Consequently, by choosing the wavelength of incident laser beam appropriately, we can selectively probe carrier dynamics in the active layer of TIPS-pentacene FETs. Here we chose incident laser beam at a wavelength of 1180 nm. In this study, positive voltage pulses of 60 V were applied to the source electrode for a period of 200 μs. Accordingly, holes are allowed to be injected from the source electrode and the injected holes are transported along the channel from the source to the drain electrode.

3. Results and discussion

Figure 4 shows the changes in the field effect mobility of TIPS-pentacene FET under the alternating application of tensile and compressive bending, which were calculated from the transfer characteristics. The mobility increased and decreased alternately, by alternating compressive bending and tensile bending, though the mobility at without bending fluctuated; the effective mobility increased about 30 % by a 1.5 % compressive strain, whereas it decreased about 15 % by a 1.5 % tensile strain. The observed mobility change was comparable with the bending effect of pentacene FETs reported in Ref. [4]. As shown in Fig. 4, the mobility partially recovered after removing the applied strains. However, the mobility change from the value measured before applying the strains was reproducible.

To analyze the observed mobility changes, we here introduce a phenomenological theory [6], by taking into account the effect of the applied strains on the charge accumulation and transport in the OFET. Briefly, by applying bending strain, the device parameters change as [6],

$$L' = L(1 - dH) \tag{2}$$

$$h' = h\left(1 + 2dH\frac{\sigma}{1-\sigma}\right) \tag{3}$$

$$\varepsilon' = \varepsilon(1 + 2\alpha dH) \tag{4}$$

with

$$\alpha = \frac{\varepsilon_1}{\varepsilon_{ins}}\frac{1+\sigma}{1-\sigma} - \frac{\varepsilon_2}{\varepsilon_{ins}}\frac{1-2\sigma}{1-\sigma}.$$

Here L, h and ε are the channel length, thickness of the gate insulator, and the dielectric constant of the gate insulator, respectively. As a result, carrier density is given as follows:

$$q' = \frac{\varepsilon' S'}{h'}V_{ch} = \frac{\varepsilon S}{h}V_{ch}\left(1 - dH - 2dH\frac{\sigma}{1-\sigma} + 2\alpha dH\right), \tag{5}$$

where d, H and σ are the half of the PEN thickness, curvature, and Poisson ratio, respectively. Here the dH represents a bending strain. The parameter α is the effect of strains on the dielectric constant of the gate insulator. The correction terms result from change in the dielectric constant of the gate insulator induced by the mechanical strain. Figure 5 plots the carrier mobility μ of the OFETs with strains, normalized with the mobility μ_0 of the OFETs without strains, i.e., μ/μ_0. As shown in the figure, observed changes in the mobility are much larger than those of the theoretical prediction.

This result implies that other factors which are not considered in the derivation of eq. (5) make a significant effect on the change in the mobility with bending strain. One of the possible factors would be the presence of grain boundaries created in the TIPS-pentacene. The grain boundary governs the carrier transport process, very sensitive to the bending, because the strains are possibly concentrated at the boundary. To verify our hypothesis, the EFISHG measurement, which enables us to directly visualize the carrier transport, was carried out. Figure 6 shows the polarizing microscope image of the channel in the TIPS-pentacene FET. Images of distinguished grains across the channel are clearly observed. Figure 7 shows the transient change in the EFISHG image of the channel with different elapsed times (see Fig. 3(b)), where the region indicated by the dotted circle in Fig. 6 was observed.

As already discussed, SHG images reflect the in-plane electric field distribution formed in the TIPS-pentacene layer. In the channel region, an origin of the electric field is the injected carriers from the source electrode. Note that a space charge field E_s originates from charges and it follows the Gauss law. That is, E_s is formed to satisfy its divergence to be in proportion to the space charge density distribution along the channel. Hence, the EFISHG image represents the transient carrier distribution at an elapsed time after the carrier injection starts from the source electrode [12]. At the elapsed time of 0 ns, carrier injection starts, and the strong SHG signal is observed near the edge of the source electrode. This indicates that the injected carriers are still accumulated near the electrode. The electric field at the source electrode edge is estimated as $E(0)$ = 1.2 MV/cm, which induces SH signal at the source electrode edge at the time t_d = 0 ns in Fig. 7 [13]. As elapsed time increases, carriers transport along the channel from the source to the drain electrode (20~60 ns). That is, carriers (holes) transport from the source to the drain electrode at t_d = 60 ns. As shown in Fig. 6, grains with rectangular shape are formed across the channel, and the EFISHG images (Fig. 7) shows that SH signals propagates inside the same rectangular-shaped grains. This result indicates that carrier transport in TIPS-pentacene is governed by the grains. Interestingly, the SHG signal is still observed in the channel as small spots after reaching to the drain electrode of the carrier sheet at the elapsed time of 80 ns. Such SHG signal is originated from the trapped carriers at the grain boundary. Hence, the grain boundaries work as trap sites, and the presence of the grain boundary restricts the carrier transport. As the results, there is a strong possibility that the grains and grain boundaries affect the carrier transport with bending strain. According to the TRM-SHG measurement, carriers prefer to transport in the grain, and the grain boundaries work as the barrier. It is reasonable to consider that the carrier behavior at the grain boundary is strongly affected by the bending strain. Therefore, this is an additional factor to enhance the mobility change with a bending stress, as shown in Fig. 5.

4. Conclusions

We studied the bending effect on the electrical property of TIPS-pentacene OFETs. The I-V characteristics were measured with or without bending strain. Results showed that the effective carrier mobility increased with compressive strain, whereas it decreased with tensile strain. The mobility changes were much larger than that obtained from the theoretical analysis. To reveal the origin of such difference, TRM-SHG measurement was carried out. The TRM-SHG observation indicated that the presence of grain boundary is responsible for the carrier transport. It is likely that carrier transport with bending strain is more affected by the strain of grains.

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Figure captions

FIGURE 1: The structure of organic field effect transistor used here. 6,13-Bis(triisopropylsilylethynyl)-pentacene (TIPS) and polyethylene-naphthalate (PEN) were used as an organic active layer and flexible substrate, respectively. S, D, G denotes source, drain, and gate electrodes.

FIGURE 2: A model of flexible OFETs under (a) compressive, and (b) tensile strains. The dotted line in the substrate represents the neutral plane.

FIGURE 3: (a) Optical arrangement for the EFISHG measurement. The probing layer beam (angular frequency ω) is polarized in the FET channel direction, and electric fields from the charge carriers in the OFETS were directly visualized by the induced second-harmonic light image (2ω). (b) Timing chart of the EFISHG measurement. The pulsed YAG laser (width 4 ns) was incident onto the FET channel with delay time t_d = 0~80 ns after the rise edge of the pulsed voltage (width of 200 μs).

FIGURE 4: The carrier mobility calculated from the I-V characteristics. The tensile (-1 %, -1.5 %) and compressive (1%, 1.5 %) strains were applied sequentially from the left to right. The mobility without bending (0 %) was measured after removing strains.

FIGURE 5: Theoretical (solid line) and experimental (filled dots) carrier mobility of OFETs with bending strains. Dotted line is a linear fit of the experimental data. μ/μ_0 in the vertical axis is the mobility μ normalized by the value μ_0 measured before applying each strain.

FIGURE 6: Polarizing microscope images of the OFET channel. The channel region between the drain (D) and source (S) electrodes, showed that organic layer (TIPS) have grains (width ~ 10 μm) bridging D and S electrodes. Dotted circle indicates the area where the carrier motion is visualized in the EFISHG measurement.

FIGURE 7: EFISHG images captured at the delay time t_d=0-80 ns (see the timing chart in Fig. 3(b)). The generated SHG was recorded on the CCD camera and traced charges propagating in the channel region. The frontier of the SHG patters was indicated by arrows. From the top to the bottom, The SHG penetrates from the source (S) electrode toward the drain (D) electrodes, as indicated by the frontier of the SHG pattern shifted from the S to D electrodes with increasing the delay time (t_d=0~40 ns). Afterwards, SHG patterns decayed (t_d=60~80 ns), indicating that the carriers filled the channel region.