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Study of TIPS-pentacene diode using electrical and electric field induced optical second harmonic generation measurement coupled with I–V and C–V measurements

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A B S T R A C T

By using the time-resolved electric field induced optical second harmonic generation (TR-EFISHG) measurement system, we studied the current-voltage (I–V) and capacitance-voltage (C–V) characteristics of metal-insulator-organic semiconductor diodes with a structure of indium tin oxide/Polyimide(PI)/6,13-Bis(triisopropylsilylethynyl)-pentacene(TIPS-pentacene)/Au. The TR- EFISHG directly probed the electric field across the TIPS-pentacene layer, and showed evidence of the charge accumulation at the TIPS-pentacene/PI interface region. Results of TR-EFISHG coupled with the I–V and C–V electrical measurements clearly demonstrated the rectifying properties and the threshold voltage shift of the diodes, before and after stress biasing. The results showed that the TR-EFISHG measurement coupled with the I–V and C–V measurements is a very useful way to analyze carrier behaviors and trapping charges of organic semiconductor diodes, even when the electrical property of the organic semiconductor layer is unknown.

1. Introduction

Organic semiconductor devices have received considerable attention in electronics, including organic light emitting diodes, organic field effect transistors, organic memory devices, organic photovoltaics, etc. [1–4]. In these devices, the many attractive potentialities of organic materials are well utilized, such as mechanical flexibility, large area, low power consumption etc. [2–5]. However, the operation of actual organic semiconductor devices is not as simple [6–8], and understanding of the carrier behavior is no longer sufficient, owing to the dielectric nature of active organic layers, the complexity of interfaces, the trapping effect due to stress biasing, chemical doping by oxidation, etc. [9–12]. Most of organic semiconductor materials such as 6,13-Bis(triisopropylsilylethynyl)-pentacene (TIPS-pentacene) show dielectric nature when they are used as an active layer in organic semiconductor devices because they usually have large energy gaps and their carrier densities are quite low. As a result, device operation of most of organic semiconductor devices is governed by carrier injection process from electrode [13,14]. TIPS-pentacene organic double-layer diode is also working as injection type device and Maxwell-Wagner effect model is very helpful for the understanding the driving mechanism of organic devices [15]. Generally, for the study of diodes, current-voltage (I–V) and capacitance-voltage (C–V) measurements are used to analyze the carrier injection, accumulation, carrier trapping etc., under the assumption that the active organic layer works as a semiconductor. As a result, experiments and analyses have been carried out on the basis of semiconductor device physics. However, it has yet to be determined whether the active organic semiconductor layer works in the same way as predicted by the general Si-based typical semiconductor physics, where chemical impurity doping is well conducted to control the n- and p-type behaviors of semiconductors. Many organic semiconductor materials have been discovered and synthesized over the past few decades. Among them are conducting polyacethelene, TIPS-pentacene, Tris(8-hydroxyquinolinato)aluminium (Alq_3), 4,4-bis[*N*-(1-naphthyl)-*N*-phenyl-amino]biphenyl (α - NPD), Copper(II) phthalocyanine (CuPc) etc. [14]. However, the semiconductor properties of these materials in organic semiconductor devices differ somewhat and, of course, differ from typical semiconductor materials. For example, it

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is known that the conductivity of TIPS-pentacene increases by carrier doping due to the carrier injection from the electrode whereas, similar to the insulator, it is quite low before carrier doping [16–18]. It is worth noting that this type of carrier behavior of TIPS-pentacene differs from that of Alq_3, α-NPD, CuPc, and others [19,20]. Therefore, illustrating the potential distribution profile of organic devices using TIPS-pentacene, on the basis of the general semiconductor device physics can only be very speculative, mainly because we do not know the density of carriers doped by carrier injection. Accordingly, it is very helpful to carry out experiments that can directly measure the electric field distribution in devices. Time-resolved electric field induced optical second harmonic generation (TR-EFISHG) is capable of probing the electric field distribution on the TIPS pentacene organic semiconductor layer.

The I–V and C–V measurements coupled with the TR-EFISHG observation will be effective for analyzing organic semiconductor devices. In this study, we examine organic double-layer ITO/Polyimide(PI)/TIPS-pentacene/Au diodes, by using TR-EFISHG coupled with I–V and C–V electrical measurements. We then show that the rectification and threshold-voltage shift AV_{th} of these organic diodes under trapping conditions can be well analyzed without the knowledge of the typical semiconductor properties of TIPS-pentacene.

2. Experiment

Organic double-layer diodes with a structure of ITO/PI (100 nm thickness)/TIPS- pentacene (10–20 nm thickness)/Au (100 nm thickness) were prepared in the same way as in our previous study [15,21]. PI is an insulator polymer and is lightweight, mechanically flexible, and resistant to heat and chemistry. Accordingly, we use it in organic semiconductor devices as an insulating layer and its dielectric constant is 3.4. PI and TIPS-pentacene layers were deposited on the etched ITO substrate by spin-coating method, followed by successive heat treatment at a temperature of 327 K in the air.

The I–V and C–V measurements were carried out using a source meter (Keithley 2400) and an impedance analyzer (Solartron SI 1260). For the C–V measurements, the amplitude and frequency of the input AC signals were set to 0.1 V and 100 Hz, respectively, at room temperature in air atmosphere. The TR-EFISHG measurement was performed with a fundamental light source (average power, 1 mW/cm^2; duration, 4 ns; repetition rate, 10 Hz) and an optical parametric oscillator (OPO) pumped by the third-harmonic light of a Q-switched Nd:YAG laser. The p-polarized fundamental light was focused on the sample by using a convex lens (f = 150 mm) with an incident angle of 45°, after passing through an SH-cut filter. The p-polarized SH light was reflected from the sample from the ITO substrate side of the diode; it was filtered using a fundamental light cut filter and was detected using a photomultiplier tube after passing through a monochromator. For the TR-EFISHG measurement, an AC voltage with square wave from (10 Hz with duty ratio of 50%) was applied to the device. The voltage range was form −20 V to+20 V under room temperature in a laboratory atmosphere.

3. Results and discussion

3.1. I — V and C — V characteristics

Fig. 1 shows a typical example of the I–V characteristics of the PI/TIPS-pentacene diode before (closed circle) and after stress biasing (open triangle) of −20 V for 0.5 h. The current showed a typical diode property, where the current level was high when the ITO electrode was negatively biased. The results show a p-type

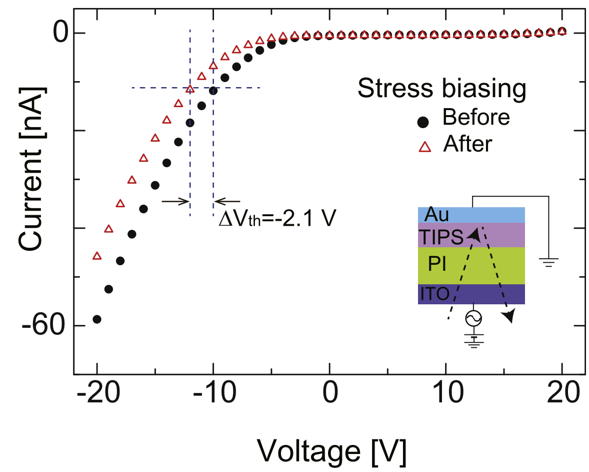

Fig. 1. I–V characteristics of ITO/PI/TIPS-pentacene/Au diode for measuring trapping carrier mechanism under stress biasing condition: before stress (closed circles) and after biasing stress (open triangle). The inset illustrates the applied organic device structure.

diode rectifying property, possibly due to the hole carrier injection into the TIPS-pentacene from the Au electrode under negative DC voltage application, and the ΔV_{th} shift of −2.1 V to negative voltage direction was observed, possibly due to the hole trapping effect [22–24]. The current flowing across the diode decreased after it was stressed by biasing. It is important to note that the Maxwell-Wagner effect model accounts for the hole accumulation at the PI/TIPS-pentacene interface, and this charge accumulation leads to the decrease of carrier transport [25,26]. Fig. 2 shows the C–V characteristics of the PI/TIPS-pentacene diode with (open triangle) and without (closed circle) stress biasing. A diode property is seen due to carrier injection, carrier accumulation, and carrier trapping. The C–V characteristics corresponded well with the I–V characteristics of Fig. 1, indicating the ΔV_{th} of −1.6 V, which is slightly small compared with −2.1 V. It is suggested that the holes smoothly inject into the TIPS-pentacene layer from the Au electrode and accumulate at the PI/TIPS-pentacene interface due to the Maxwell-Wagner effect. It is significant that, while the injected holes are trapped at the interface by stress biasing, some of them appear to move further into the insulator layer to be trapped [26]. However, this discussion is merely speculation, and we cannot conclude the details based only on the I–V and C–V characteristics, because we do not have a solid physics picture of the TIPS-pentacene layer on

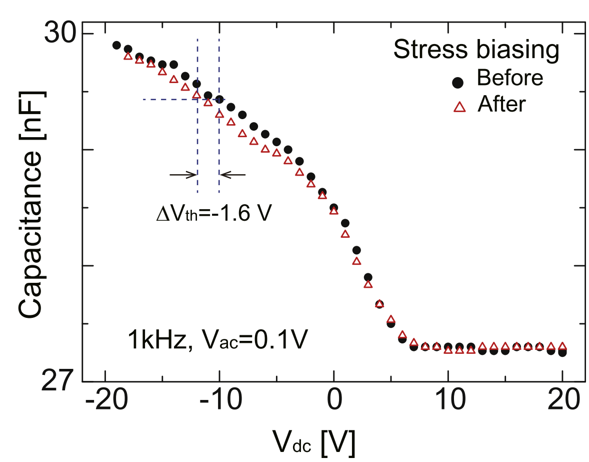

Fig. 2. C–V characteristics of ITO/PI/TIPS-pentacene/Au diode of stress biasing effect.