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Study of Discharging Process of Double Layer Organic Diodes Using Displacement Current Measurement Coupled with Electric-Field-Induced Optical Second-Harmonic Generation Measurement

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Abstract

Displacement current measurement (DCM) is widely used to analyze I-V and C-V characteristics of organic devices, though this is an indirect method in the detection of carrier motions. Recently, electric-field-induced optical second-harmonic generation (EFISHG) measurement has been developed to directly observe *electric field* in organic devices. In this study, we employed EFISHG measurement in combination with the DCM to study discharging process of Au / pentacene / polyimide (PI) / indium-tin-oxide (ITO) double layer diodes, in terms of the I-V and C-V characteristics. EFISHG directly probed carrier behaviors in the pentacene layer, and it was shown that the EFISHG measurement coupled with the DCM is very helpful to study discharging process of organic diodes.

1. Introduction

Organic devices have been paid much attention in electronics, since the discovery of new organic semiconductor materials. As a result, a variety of devices such as organic FETs, OSCs, OLEDs have been developed. To evaluate organic device performance, many techniques have been proposed and employed. Among them is the DCM measurement, and it is widely used for analyzing carrier behaviors [1]. This method is very simple, but it is difficult to discuss the details of carrier behaviors, in terms of the I-V and C-V characteristics, because we cannot directly catch carrier behaviors in devices. Recently, it has been found that EFISHG measurement is available for directly analyzing carrier behaviors in organic devices [2][3]. In this work, we studied discharging process of Au/pentacene/polyimide (PI)/indium-tin-oxide (ITO) double layer organic devices by using EFISHG method in combination with the conventional DCM. Results evidently showed that this approach is very helpful for analyzing carrier behaviors in organic diodes.

2. Experiment

Figure 1 shows the double-layer diodes used here. This double-layer device was prepared as follows: after depositing a spin-coating polyimide precursor film with a thickness of 100 nm (=d_2) onto ITO substrate, it was baked and imidized in an oven at 260 ℃ for two hours, to produce a polyimide film. Subsequently, 100 nm (=d_1) thickness pentacene was thermally deposited on the substrate coated with a PI layer. Finally, Au was evaporated in a vacuum. The working electrode area was 3.1 mm^2. The capacitances of pentacene and PI layer measured by impedance spectroscopy were C_1= 1.1 nF and C_2= 0.63 nF, respectively.

In the DCM measurement, ITO electrode was grounded and ramp voltage V_{ex} was applied to Au electrode, in a manner as illustrated in Fig. 1. The displacement current flowing through the external circuit is given as

$$I = \frac{C_1 C_2}{C_1 + C_2}\frac{dV_{ex}}{dt} + \frac{C_2}{C_1 + C_2}\frac{dQ_s}{dt} \qquad (1)$$

Here, Q_s corresponds to the accumulated charge at the interface between pentacene and PI. Using eq. (1), we estimated ΔQ_s.

In the EFISHG measurement, we probed the transient of electric field formed in pentacene layer selectively, by using a 860 nm laser pulse [3]. Here, the square root of the SH intensity is proportional to the electric field E_1 in pentacene layer. The electric field in pentacene layer is given as

$$E_1 = \frac{1}{d_1}\frac{C_2}{C_1 + C_2}V_{ex} - \frac{1}{d_1}\frac{Q_s}{C_1 + C_2} \qquad (2)$$

Using eq. (2), we estimated Q_s.

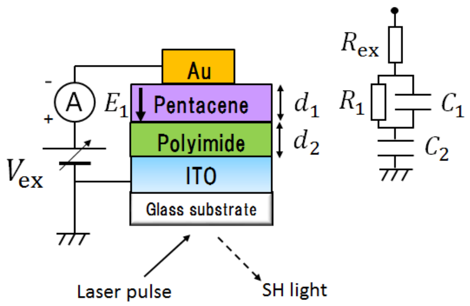

Fig. 1 Sample structure

3. Results and discussion

We applied an external ramp voltage at various sweep rates. Here, the initial voltage 10 V was applied for 100 ms to charge the diode and then dropped down to -20 V, -15 V, -10 V, -5 V, 0 V and 5 V, respectively. Figure 2 (a), (b) and (c) shows the external voltage applied to the sample, transient current flowing in the external circuit and square root of SH intensity, respectively. Results suggested that the property of pentacene layer changes gradually from conductive state to insulating state as external voltage drops. For analyzing the carrier behaviors when applying voltage 10 V to -20 V (brown lines and dots) to the diode, we divided the experimental result into four regions (i), (ii), (iii) and (iv), as shown in Fig. 2.

In region (i), current I negatively increases as the external voltage decreases. At the end of this region, discharging current I reaches the $C_2 dV/dt$ with a response time of $\tau_{RC} = RC_2$ = 0.72 μs. Note that τ_{RC} was estimated by tangent line. Therefore, a series resistance R of external circuit (R_{ex} = 680 Ω) and pentacene layer (R_1) can be obtained as 1.1 kΩ. In region (ii), the current I saturates and the displacement current $C_2 dV/dt$ = 0.95 mA continuously flows, where C_2 is the capacitance of polyimide single layer. In this situation, pentacene layer serves like a conductor. Accordingly, the constant electric field E_1 was observed during the EFISHG measurement, and the electric field was not zero. Results indicate that the pentacene has non-zero electrical resistance. In region (iii), the current begins to decrease and the electric field increases because carriers Q_s accumulated on the interface between pentacene and PI layer return to the gold electrode. This region corresponds

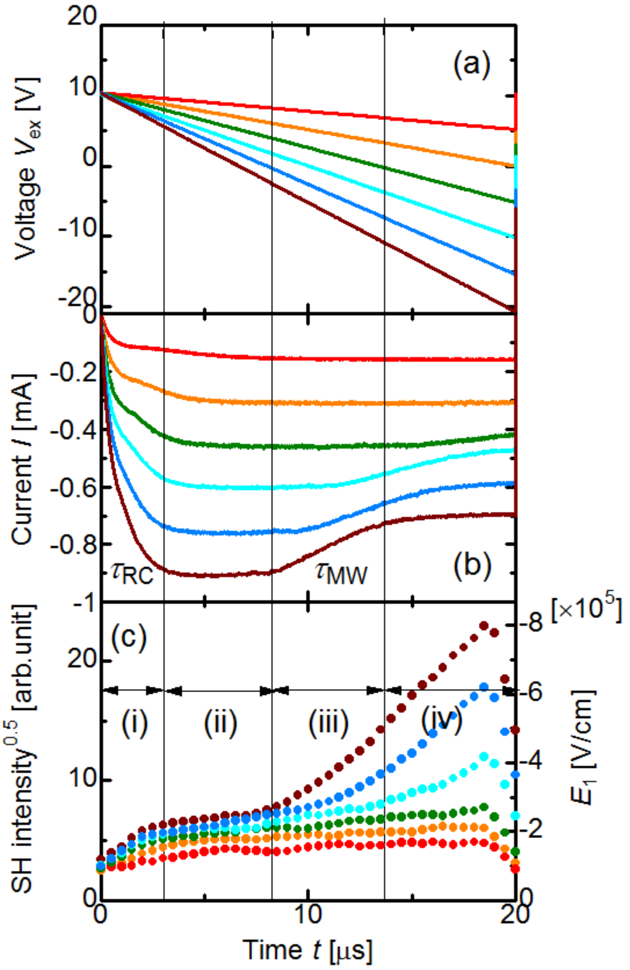

Fig. 2 Transient of (a) External voltage, (b) current and (c) square root of SH intensity.

to the transient state in Maxwell-Wagner model, where a response time of τ_{MW} = 4.8 μs was obtained by using equations in the other paper [4]. This value is well consistent with that estimated by Fig. 2 (b). Finally, in region (iv), the current flows constantly. The current level is given by CdV/dt = 0.6 mA, with $C=C_1//C_2$=0.40 nF in eq. (1). This current level suggests that carriers accumulated on the interface are completely escaped from the interface to the gold electrode.
That is, in this region, this device is serving as simple double layer insulator.

Similar experimental results were seen at other sweep rates. Noteworthy, regions (i) to (iv) appeared without depending on the sweep rate.

Finally, we calculated the charge ΔQ_s at the interface between pentacene and PI and then plotted them in Figure 3. Here, solid lines and dots were obtained using eq. (1) in the DCM measurement and eq. (2) in the EFISHG measurement, respectively. It was found that ΔQ_s estimated by both methods are good agreement with each other.

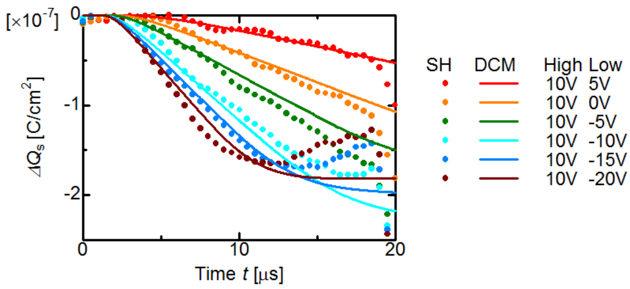

Fig. 3 Transient of ΔQ_s calculated by DCM and EFISHG measurements.

4. Conclusions

It was found that carrier behaviors in double layer organic devices can be analyzed precisely by EFISHG measurement coupled with the DCM measurement. We showed that an experimental approach based on dielectric physics is very useful. DCM and EFISHG results will give us more detail information on carrier behaviors for understanding discharging of organic diodes.

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