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Analysis of anomalous discharging processes in pentacene/C_{60} double-layer organic solar cell

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1. Introduction

Among photovoltaic devices, organic solar cells (OSCs) are one of the candidates that can fulfill the demand, because of simple fabrication onto various substrates, which includes flexible plastics and others [1]. Though it is well-known that carrier processes, such as separation of excitons into electron and hole pairs, life-time of carriers, and so forth, play a dominant role in OSCs devices, the detailed understanding is somehow insufficient in terms of interfacial carrier behaviors [2, 3]. Therefore we need a technique that is capable of detecting carrier motion as well as electric field distribution in OSCs. Recently, we have shown that optical electric-field-induced second harmonic generation (EFISHG) measurement is available for probing the electrode charging and interface charging of pentacene/C_{60} OSCs under short circuit condition [4]. Along with these our recent experiments, it has been found that the charging and discharging processes on the interface is non-symmetrical, which means the charge density on the interface increases after stopping the illumination, though it should be decreased. Meanwhile, for the other kinds of double layer organic devices, such as OLEDs and OFETs [5, 6], such increase of charge density on the interface have not been observed, indicating that photovoltaic effect somehow dominated this anomalous discharging process. In the present study, in order to further clarify interfacial carrier behaviors in OSCs, we have carefully analyzed the discharging processes of the OSCs. Making use of EFISHG experiments on OSCs connected to various external resistances R_e, we have shown that the accumulated charge density at the interface increased, and the IS analysis well supported the results. Finally we concluded that the photovoltaic (PV) effect of the OSCs was responsible for the observed anomalous discharging.

2. Experiment

Double-layer OSCs with an indium zinc oxide (IZO) /pentacene/C60/Al structure were prepared as portrayed in Fig. 1. IZO-coated glass substrates were UV/ozone treated and nearly free from organic residuals. The pentacene layer with a thickness of 40 nm and C_{60} layer with a thickness of 100 nm were successively deposited onto the UV/ozone treated IZO surface. Finally, Al electrodes with a thickness of 100 nm were deposited onto the C_{60} surface. The working area of the OSCs was A = 3.1 mm^2. A red light from a light-emitting diode (wavelength 630 nm, intensity 1 mW/cm^2) was used as a light source. Note that pentacene and C_{60} layers absorb light at a wavelength of 630 nm, and generate excitons inside the layers.

The SHG measurements were carried out using an experimental arrangement portrayed in Fig. 1. A pulsed laser was used as a probing light (repetition rate 10 Hz, average power 1 mW, duration 4 ns), which was generated from an optical parametric oscillator pumped with the third-harmonic light of Q-switched Nd:YAG laser. A *p*-polarized pulsed laser beam was focused onto the sample surface at an incident angle of 45°. The SHG light generated from the sample was detected using the photomultiplier tube (PMT), and its intensity I (2ω) was recorded with a digital multimeter. In the present study, we used a laser beam with a wavelength of λ_ω=1,000 nm, and recorded the generated SHG signal at a wavelength of $\lambda_{2\omega}$= 500 nm to selectively measure the electric field in C_{60} layer [4].

In order to study the PV effect on carrier behavior in OSCs, we measured the EFISHG at various I-V conditions by connecting various external resistances R_e to the OSCs in series, where transient EFISHGs were recorded. Hence, the external resistances R_e were 10, 25, 47, 100 and 180 kΩ, and these resistances resulted in the external voltages of 0.010, 0.050, 0.075, 0.100 and 0.150 V, respectively, under illumination, which make the OSCs to exhibit the corresponding I-V characteristics.

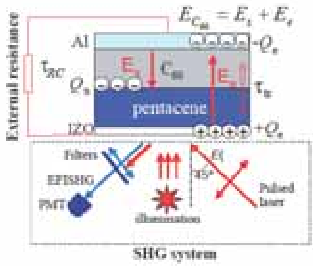


Fig. 1 Sample structure and experimental arrangement

3. Results and discussion

Figure 2 illustrates the dependence of electric field across the C_{60} layer in respect to different external resistance. The electric field is calculated from the SHG signal. The results were discussed based on the Maxwell-Wagner model, which include a paralleled circuit comprised of a constant conductance (G_1, G_2) and a capacitance (C_1, C_2). The accumulated charge Q_s at the interface is governed by the conduction current and relaxation time of two different layers. The charging process of the electrodes (E_e) was identified by the external voltage and was enhanced by increasing the external resistance. On the other hand, the charging of the interface was ruled by the current flowing through the circuit and was suppressed by the increasing of the external resistance. However, by comparing the SHG signals between charging and discharging processes, we found that the peak value of the SHG signal in discharging process was higher than the charging process, indicating that the charge density on the pentacene/C_{60} interface increased during the discharging process of the electrodes. Accordingly, we considered that there would be a re-charging process at the interface after stopping the illumination. For understanding of this situation, we should clarify two facts. First of all, the residual excitons remain in the organic layer after we stop the illumination, which still provide the conducting current for a very short time. As observed in the Fig. 2, the electrode discharging always has a delay time about 1×10^{-5} s, Secondly, the conductivity of the two layers would be decreased by stopping the illumination. This decrease is partially identified by the optical properties of the material and would happen immediately after stopping the illumination. With the decrease of the conductivity, the saturated value of the charge density on the interface would be increased. Hence, the re-charging process of the interface would happen again until the conductive current cease to flow.

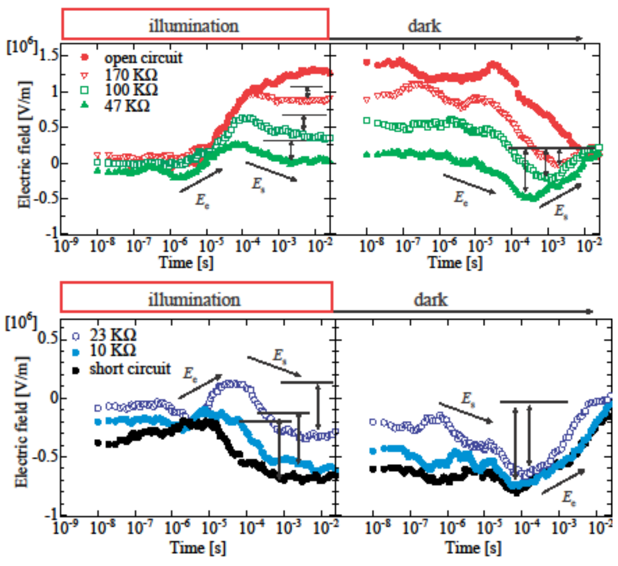
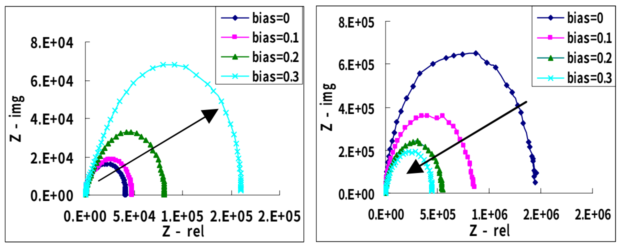


Fig. 2 EFISHG measurements with different external resistance.

Figure 3 shows the results from impedance spectroscopy (IS) measurements. In dark condition, the increase of DC bias led to the narrowing of the Nyquist circle, indicating that the conductivity of the organic layer increased due to the injection of the carriers. However, under the illumination, the Nyquist circle of the impedance is getting larger with increase of DC bias, indicating the conductivity of the organic layer decreased due to the applied external voltage. Due to the PV effect, the illumination induce the photo current across the OSCs device and this account for the different bias dependence of IS measurement under illuminated and dark conditions. Therefore, the illumination enhances the conductivity of the OSCs in two aspects. Firstly, the charges generated by the PV effect increase the conductivity of the OSCs devices. Secondly, the illumination enhances the conductivity due to photoconductive effect. When we apply a DC bias corresponding to V_{oc}=0.22V, the contribution of the PV effect to the conductivity should be negligible. However, from Fig. 3 we found that the conductance of the illuminated device at the DC bias 0.2V ($\approx V_{oc}$) is still twenty times smaller than that in dark condition. Hence, the IS results confirmed that the conductivity of the OSCs can be changed by photoconductive effect. This result supports our speculation about the reason for the re-charging process in the dark condition.



(a) under illumination (b) in dark condition

Fig.3 IS measurement of the OSCs in the illuminated and dark condition

4. Conclusions

We observed nonsymmetrical charging and discharging process in OSCs, which suggests that the accumulated charges on the interface increase after stopping the illumination. The IS results proved that the illumination changes the conductivity of the organic layer without the contribution of photo current. This change and the lingering behavior of the PV effect account for the re-charging of the interface after terminating the illumination.

References

[1] P. Aliberti, Y. Feng, Y.Takeda, K. Shrestha, M. A. Green, and G. Conibeer, J. Appl. Phys. 108, 094507 (2010).

[2] T. Manaka, F. Liu, M. Weis, and M. Iwamoto, J. Phys. Chem. C 113, 10279 (2009).

[3] L. Zhang, D. Taguchi, J. Li, T. Manaka, and M. Iwamoto, J. Appl. Phys. 108, 093707 (2010).

[4] D. Taguchi, T. Shino, X. Chen, L. Zhang, J. Li, M. Weis, T. Manaka, and M. Iwamoto, Appl. Phys. Lett. 98, 133507 (2011).

[5] D. Taguchi, L. Zhang, J. Li, M. Weis, T. Manaka, and M. Iwamoto, Jpn. J. Appl. Phys. 50, 04DK08 (2011).

[6] M. Weis, T. Manaka, M. Iwamoto, J. Appl. Phys. 105, 024505 (2009).