Analysis of interfacial charging processes in pentacene/C$_{60}$/BCP triple-layer organic solar cells using a Maxwell-Wagner model

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1. Introduction

Among photovoltaic devices, organic solar cells (OSCs) are one of the candidates that can fulfill the demand in the market, because of simple fabrication onto various substrates, which includes flexible plastics and others [1]. Though it is well-known that carrier processes play a dominant role in OSCs devices, the detailed study related to the carrier behavior as well as the electrical phenomena induced by the introduction of a blocking layer is still not sufficient. In our previous study, we employed the Maxwell–Wagner (MW) model analysis for studying interfacial phenomena in double-layer organic devices e.g., double layer organic light emitting diode (OLED), and showed the potentiality of this analysis based on a dielectrics physics approach [2-3]. These results motivated us to employ the MW model analysis to multi-layer structure and analyze the photo-induced carrier behaviors in OSCs.

In this paper, we prepared IZO/pentacene/C$_{60}$/BCP/Al OSCs with a blocking BCP layer, and studied the charging process by using a Maxwell-Wagner model. Results evidently showed the presence of charge accumulation at the pentacene/C$_{60}$ and C$_{60}$/BCP interfaces, and the resulting two established electric fields by accumulated charges merge each other in the case of smooth carrier injection from electrodes. We also measured the variation of internal electric field of the OSCs which are connected to various external resistances. The results explained the relationship between interfacial charging and the fill-factor of the OSC devices.

2. Experiment

Figure 1 portrays OSCs with an indium zinc oxide IZO/pentacene/C$_{60}$/BCP/Al structure. These OSCs were prepared as follows: IZO-coated glass substrates were UV/ozone treated to be nearly free from organic residuals. The pentacene layer with a thickness of 40 nm (d_1) and the C$_{60}$ layer with a thickness of 50 nm (d_2) were successively deposited onto the UV/ozone treated IZO. After that, a BCP layer with a thickness of 10 nm was deposited. Finally, Al electrodes with a thickness of 100 nm were deposited onto the surface of C$_{60}$ layer, where the working area of the OSCs was A = 3.1 mm^2. In the measurement, a red light from a light-emitting diode was used as a light source to provide illumination pulse to the OSCs. Note that pentacene and C$_{60}$ layers absorb light at a wavelength of 630 nm [4], and generate excitons inside their layers.

Figure 1 also portrays the arrangement of the SHG meas-urement for probing the electric field in OSCs. A pulsed laser was used as a probing light, which was generated from an optical parametric oscillator pumped. 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 a photomultiplier tube, and its intensity was recorded with a digital multimeter. The generation of SHG is material dependent and it shows wavelength dependence of incident laser beam. Hence, we used a laser beam with a wavelength of λ_ω = 1000 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. Note that the square-root of the generated SHG intensity is in proportion to the electric field $E(0)$ in the C$_{60}$ layer [4]. $E(0)$ is given as $E_b + E_e + E_{s1} + E_{s2}$, where E_b is the background internal electric field established in the devices, E_e is the electric field originated from charges on electrodes, E_{s1} is the electric field originated from accumulated charges $Q_{s1}(t)$ at the pentacene/C$_{60}$ interface, and E_{s2} is the electric field originated from accumulated charges $Q_{s2}(t)$ at the C$_{60}$/BCP interface. Accordingly, we can discuss carrier behaviors in the OSCs by probing the transient EFISHG.
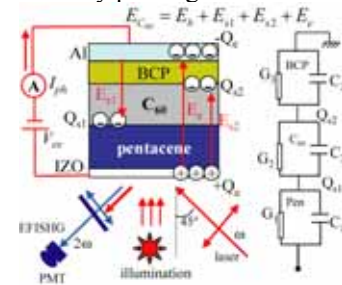

Fig. 1 Sample structure and equivelent circuit

3. Analysis

Using the equivalent circuit of Fig. 1, we get the relationship between the V_1, V_2 and V_3, under assumption that a constant D.C. current flows across the OSC. That is,

$$C_1 \frac{dV_1}{dt} + G_1 V_1 = C_2 \frac{dV_2}{dt} + G_2 V_2 \qquad C_1 \frac{dV_1}{dt} + G_1 V_1 = C_3 \frac{dV_3}{dt} + G_3 V_3$$

Here, $V_3 = V - V_1 - V_2$. After a length calculation, we get the V_2 as

$$V_2 = \frac{V}{C_1 C_2 + C_2 C_3 + C_3 C_1} \left(\frac{A}{s} + \frac{B}{s+\alpha} + \frac{C}{s+\beta} \right)$$

$$A = \frac{G_1 G_3}{G_1 G_2 + G_2 G_3 + G_3 G_1} (C_1 C_2 + C_2 C_3 + C_3 C_1)$$

$$B = \frac{-\alpha C_1 C_3 - A\beta + C_1 G_3 + C_3 G_1}{-\alpha + \beta}, \quad C = \frac{\beta C_1 C_3 + A\alpha - (C_1 G_3 + C_3 G_1)}{-\alpha + \beta}$$

$$\alpha = \frac{P + \sqrt{Q}}{2(C_1C_2 + C_2C_3 + C_3C_1)} \quad \text{and} \quad \beta = \frac{P - \sqrt{Q}}{2(C_1C_2 + C_2C_3 + C_3C_1)},$$

where $P = C_1(G_2 + G_3) + C_2(G_1 + G_3) + C_3(G_2 + G_3)$ and

$$Q = (C_2G_1 - C_1G_2)^2 + (C_3G_2 - C_2G_3)^2 + (C_1G_3 - C_3G_1)^2$$
$$-2(C_3G_1 - C_1G_3)(C_1G_2 - C_2G_1) - 2(C_3G_1 - C_1G_3)(C_2G_3 - C_3G_2)$$
$$-2(C_2G_3 - C_3G_2)(C_1G_2 - C_2G_1)$$

Here, the $1/\alpha$ and $1/\beta$ are the relaxation time of the two interfacial charging processes.

The accumulated charge density of the pentacene/C_{60} interface is given as $Q_1 = C_2V_2 - C_1V_1$. In the steady state, $Q_1(\infty) = I_c\Delta\tau$, where I_c is the conduction current and $\Delta\tau = (C_2/G_2 - C_1/G_1)$ is the relaxation time difference between the two organic layers.
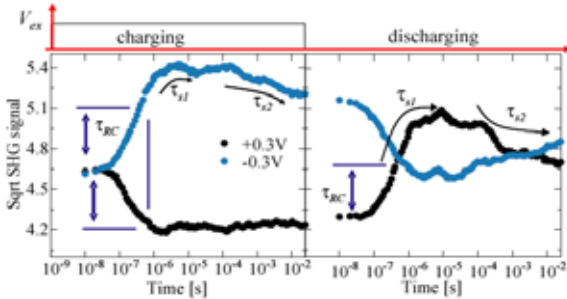
4. Results and discussion

Fig. 2 SHG measurements with applied voltage pulse.

Figure 2 shows the EFISHG generated from the IZO /pentacene/C_{60}/BCP/Al OSC in response to the AC square-wave voltage pulse (10 Hz), where the amplitude of the DC applied voltage V_{ex} was +0.3V or -0.3V. Three relaxation processes were identified during the EFISHG measurement. The first relaxation process was due to the electrode charging. Subsequently, two interfacial charging processes followed. The time constants of the charging processes were determined using a curve-fitting method based on a filtering technique. For V_{ex} = -0.3 V, the relaxation time of the electrode charging was $\tau_{RC} \approx 2.4 \times 10^{-7}$ s. Meanwhile, the relaxation time of the pentacene/C_{60} and C_{60}/BCP interface charging processes were $\tau_{s1} \approx 5 \times 10^{-6}$ s and $\tau_{s2} \approx 1 \times 10^{-3}$ s, respectively. On the other hand, for V_{ex} > 0, the application of positive voltage assisted holes to smoothly inject from the IZO to the pentacene layer and resulted in the acceleration of interfacial relaxation. Hence, as seen in Fig. 2, at V_{ex} = 0.3 V, the two interfacial charging processes were merged each other completely. As discussed in section 3, the time constant difference between the two interfacial charging processes is related to the value Q, which is regulated by the conductance difference of the three organic layers. Hence, this result also proved that conductance difference of the three organic layers became smaller with the smooth injection.

Figure 3 shows the results of the SHG measurements of a triple-layer OSC sample under the illumination. The sample was set up with various external resistances R_{ex}. The R_{ex} are 0, 1.8 kΩ, 4.7 kΩ and 100 kΩ, which make the external voltage V_{ex} to be 0V, 0.7 V, 0.15 V and 0.24 V, respectively. Three relaxation processes can be defined in Fig. 3. With the decrease of the external resistance, the flowing current increased gradually and thus relaxation time for the charging process of the E_{s1} became significantly faster. Meanwhile, with the external resistances changing from 0 to 100 kΩ, the electric field E_{s1} showed slightly variations, as can be observed from the SHG intensity changing in Fig. 3. Based on our previous analysis [5], we may conclude that the accumulated charges on the interfaces would suppress the extracted current of the OSCs. Hence, for the V_{ex} in the region from 0V to V_{oc}, the insignificant charging processes lead to the sufficient current extraction form the OSCs to the external circuit and this may account for the high fill-factor of the triple-layer OSCs sample.
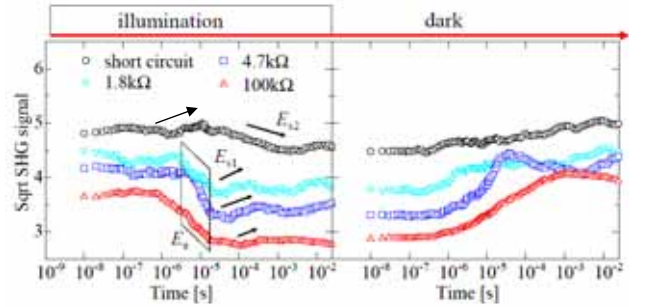

Fig.3 EFISHG measurement of the OSCs with different external resistances

5. Conclusions

We studied the interfacial charging of the triple layer OSCs, on bias of dielectric nature of the organic layer. Both experimental observation and theoretical analysis demonstrated that charge accumulation would occur on both pentacene/C_{60} and C_{60}/BCP interfaces. On the other hand, the changing of interfacial charging processes according to different the external resistances was also measured by the EFISHG system and results account for the contribution of the interfacial charging to the FF.

References

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