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Interfacial carrier relaxation in the organic solar cell with inverted structure: the influence of conductivity degradation

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Abstract

The interfacial processes in pentacene/C60 solar cells and its inverted structure were studied using optical observation and Maxwell-Wagner model. The analysis revealed that the conductance degradation of C_{60} layer is the main origin of the interfacial charge accumulation.

1. Introduction

Recently, the power conversion efficiency of organic solar cells (OSCs) has been improved year by year. However, the further application of these devices in the field of electronics is still facing several critical problems, such as short life time, insufficient understanding of carrier behavior and so on. In OSCs, the actual carrier process, such as the carrier transport mechanism and driving force in actual PV cells, are still not so clear and the study of potential profiles as well as internal electric fields in PV devices will be helpful. In our previous researches[1, 2], we already proved that Maxwell-Wagner type interfacial charging happened on each interfaces of OSCs and these charging processes degrade the performance of OSCs. Hence, it is necessary to reduce these interfacial charging processes in order to maintain a high performance for OSCs. On the other hand, for the OSCs using C_{60} as the acceptor, the most possible reasons for the degradation of the I-V performance may relate to the decrease of C60 conductivity caused by oxygen absorption [3, 4]. By employing the tris-8-hydroxy-quinolinato aluminum (Alq3) or bathocuproine BCP to serve as the oxygen blocking layer, the lifetime of the OSCs can be significantly improved. Meanwhile, we also found that by inserting a blocking layer of BCP [5], the interfacial charging process happened on pentacene/C_{60} interface can be significantly suppressed, which suggested that the interfacial charging can be blocked by protecting the conductivity of C_{60}. These results motivated us to further study the detailed carrier behavior induced by the degradation of C_{60} for the further improvement of OSCs.

In the present study, we prepared OSCs with normal and inverted structure and studied the interfacial charging processes of the OSCs. In order to further clarify the contribution of the oxygen degradation to the interfacial carrier behavior, we carried out the transient electric-field-induced second-harmonics-generation (EFISHG) experiments and analyzed the results using a Maxwell-Wagner model. We concluded that the change in the conductance caused the converse charging behavior on the pentacene/C_{60} and C_{60}/pentacene interface.

2. Experiment

Figure 1 portrays OSCs with indium zinc oxide IZO/pentacene/C_{60}/Al and IZO /C_{60}/pentacene/Al inverted 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 50 nm (d_1) and the C60 layer with a thickness of 50 nm (d_2) were successively deposited onto the UV/ozone treated IZO. 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 (repetition rate 10 Hz, duration 50 ms, switching on and off time within 50 ns) 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.

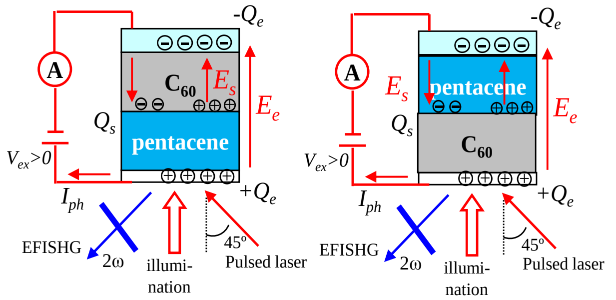

Fig. 1 Sample structure and experimental set up

Figure 1 also portrays the arrangement of the EIFSHG measurement 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 45º. 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 EFISHG 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 EFISHG 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 EFISHG intensity is in proportion to the electric field E (0) in the C_{60} layer [4]. E (0) is given as $E_b+E_e+E_s$ 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_s is the electric field originated from accumulated charges $Q_s(t)$ at the pentacene/C_{60} interface. Accordingly, we can discuss carrier behaviors in the OSCs by probing the transient EFISHG.

3. Results and discussion

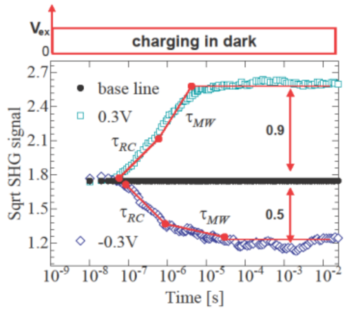

Fig. 2 SHG measurements with applied voltage pulse for the sample of normal pentacene/C_{60} OSCs.

Figure 2 shows the EFISHG response of the double-layer IZO/pentacene/C60/Al device in dark, in response to the AC square-wave voltage pulse (10 Hz). Here the biasing d.c. voltage V_{ex} was +0.3V or -0.3V, and it was applied on the IZO electrode with reference to the Al electrode. By using a curve-fitting method based on a filtering technique, two carrier relaxation processes (denoted by τ_{RC}, τ_{MW}) were identified from the transient SHG signal, as depicted in Fig. 2. The is the response time for electrode charging through the external circuit, and is the response time for the Maxwell-Wagner type interfacial charging and with V_{ex}=0.3 V, while with V_{ex}= -0.3 V. The magnitude of applied external field E_{ex} was the same for both positive and negative biasing conditions, i.e., at Vex=±0.3 V, but the MW interface charge field Es was more significant for the positive biasing voltage, Vex=+0.3 V. This results proved that $\tau_{pentecene} < \tau_{C60}$.

Figure 3 (a) shows the EFISHG generated from the IZO/C_{60}/pentacene/Al inverted OSCs in response to the AC square-wave voltage pulse (10 Hz) in dark, where the amplitude of the DC applied voltage V_{ex} was +0.3V or -0.3V. Two relaxation processes were identified during the EFISHG measurement, corresponding to the electrode charging (E_e) and the interfacial charging (E_s). The first relaxation process was observed just after applying AC square-wave voltage pulse, due to the electrode charging, which resulted in the E_e across the whole sample. For V_{ex} = 0.3 V, the SHG signal was decreased, suggesting that the C_{60} layer was relaxed and holes accumulated at the C_{60}/pentacene interfaces. Accordingly the two electric fields, E_e and E_s, were successively formed in the C_{60} layer along with electrode charging and interfacial charging and their electric field directions inside the C_{60} layer were opposite with each other. Similarly, for V_{ex} = -0.3 V, EFISHG signal decreased firstly and then increased, indicating that electrons accumulated at the C_{60}/pentacene interfacesFigure 3 (b) also shows the EFISHG generated from the inverted OSCs in response to the AC pulse (10 Hz) under illumination. The relaxation of the C_{60} layer was further enhanced and the interfacial charging processes is more significant than dark condition. Both experiments proved that C_{60} layer relaxed faster than pentacene layer and thus $\tau_{pentecene} > \tau_{C60}$. These results can be explained by considering the effect of the degradation caused by the open air. For the inverted sample, C_{60} layer is covered by the pentacene and thus the oxygen absorption can be blocked by the pentacene. Hence, the conductivity of the C_{60} is enhanced by this blocking effect, which leads to the opposite interfacial charging behavior on the interface.

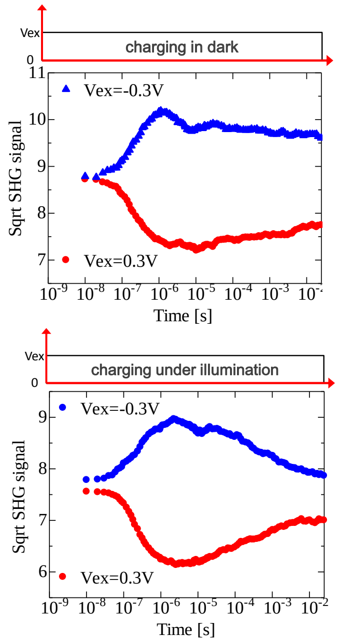

Fig.3 EFISHG measurement of the OSCs with voltage pulse for the sample of inverted C_{60} /pentacene OSCs.

5. Conclusions

By using the electric-field-induced optical second-harmonic generation measurement, we studied interfacial charging process on the interface of IZO/pentacene/C_{60}/Al and IZO/C_{60}/pentacene/Al OSCs. Results showed that under the same external voltage degradation effect happened with C_{60} layer can switch the polarity of the accumulated charges on the donor-acceptor interface. The Maxwell–Wagner model analysis can well explain all the results.

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