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Investigation of PEDOT: PSS Insertion Effect on Carrier Transport in PCPDTBT: $PC_{71}BM$ Bulk Heterojunction Organic Solar Cell by EFISHG

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1. Abstract
Carrier transport processes in the bulk heterojunction (BHJ) organic solar cells (OSCs) are investigated and analyzed using optical electric-field-induced second-harmonic generation measurement (EFISHG). Then, we study carrier transport processes by adding PEDOT: PSS layer. The results show improvement on the transit time of the electrons and holes in BHJ organic solar cells (OSCs).

2. Introduction
Most of the OSC devices are based on the bulk heterojunction (BHJ) structures, a blend of an electron donating polymer (donor (D) with an electron accepting molecule acceptor (A)) [1]. BHJ fits low-cost solution processing because donor-acceptor phase separation is spontaneously formed by simple spin-coating method from the binary solution. It also delivers effective structure to split the excitons into free carriers [2]. Relationship between nanoscale morphology and solar cell performance have been investigated by using scanning probe microscope, transmission electron microscope, and other techniques. However, understanding of the carrier transport process in the bulk-heterojunction layer is not sufficient for example, we cannot answer how carrier transport process in the bulk-hetero structured layer changes if, the interface layer such as poly(3,4-ethylenedioxythiophene)-polystyrene sulfonate (PEDOT: PSS) is induced. This is due to indefinite carrier motion caused by the complex donor-acceptor phase structure in BHJ OSC. In this paper, we focus on the carrier transport in poly [2,6-(4,4-bis-(2-ethylhexyl)-4H-cyclopenta[2,1-b;3,4-b0] dithiophene)-alt-4,7-(2,1,3-benzothiadiazole)] (PCPDTBT) and [6,6 -phenyl C71 butyric acid methyl ester ($PC_{71}BM$), bulk heterojunction organic solar cells studied by the electric field induced optical second harmonic generation (EFISHG) measurement. Nonetheless, using buffer layers such as PEDOT: PSS is known to significantly improves the OSC performances [3,4]. For this reason, the effect of the PEDOT: PSS layer on the carrier transport process is also discussed in terms of the transient change in the internal electric field.

3. Experiment
A. Sample preparation
Figure 1 shows structure of the PCPDTBT: $PC_{71}BM$ based bulk heterojunction solar cells used in this experiment. For the fabrication of the OSC devices, the PEDOT: PSS layer with a thickness of 40 nm was firstly placed on the ITO surface by spin coating process from binary solution of 25 mg/ml solution of PCPDTBT and $PC_{71}BM$ in chloroform. Volume ratio of the solution was 1:1.5 (PCPDTBT: $PC_{71}BM$) (Please check!). About 100 nm thick PCPDTBT: $PC_{71}BM$ BHJ films were prepared at the top of PEDOT: PSS layers. Then, the top aluminum (Al) electrode (thickness: 100 nm) was placed on the BHJ films by vacuum deposition.

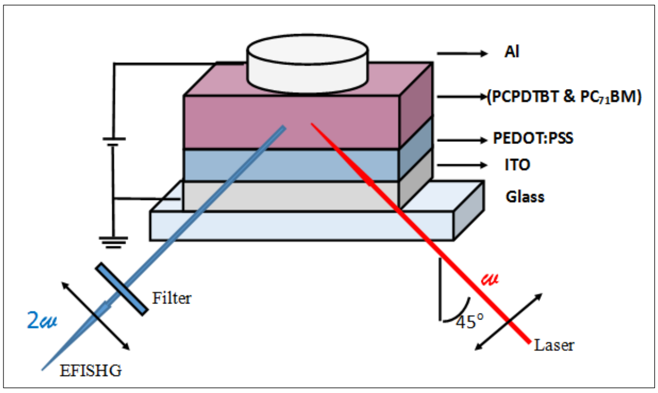

Fig. 1 Schematic structure of BHJ solar cells

B. EFISHG measurement
A pulsed laser was utilized as an fundamental light (repetition rate, 10 Hz; average power, 1mW; duration, 4 ns) for the SHG measurements, which is produced from an optical parametric oscillator pumped with the third-harmonic light of a Q-switched neodymium-doped yttrium aluminum garnet (Nd-YAG) laser. A p-polarized laser light was impinged into the surface of the sample at an incident angle of 45°. [5,6] The SHG light, which is generated from the sample, was observed using a photomultiplier tube, and its intensity was measured with a digital multimeter. The nonlinear optical susceptibility strongly depends on the material, and it shows angular frequency dependence of the incident light. Accordingly, the $PC_{71}BM$ layer delivers a robust SHG signal with a wavelength of $\lambda_{2\omega}$=540 nm, whereas PCPDTBT shows no SHG response at this wavelength. Hence, we used a laser light with a wavelength of λ_{ω}= 1080 nm and record the SHG signal at a wavelength of $\lambda_{2\omega}$= 540 nm to measure the electric

field in the $PC_{71}BM$ layer selectively. The EFISHG signal is generated owing to the coupling of electrons in $PC_{71}BM$ molecules and incident laser light $E(\omega)$ in the presence of a local electrostatic field $E(0)$ in the PCPDTBT layer, and thus the square root of the SHG signal is in proportion to the electric field $E(0)$.

4. Results and Discussion

Figure 2 shows transient response of the EFISHG signal from ITO/BHJ/Al device in the dark, under the application of the AC square-wave voltage pulse (10 Hz). The amplitude of the AC voltage Vex was +1V, 0V or -1V, and it was applied to the Al electrode with respect to the ITO electrode grounded. Two carrier relaxation processes denoted by the electrode charging (Ee) and the interfacial charging (Es) are identified through the transient SHG signals, as represented in Figure 2. The transit time of carriers is evaluated directly from the SHG results, by assuming that the SH intensity begins to increase at the time corresponding to? the transit time. Interestingly, SHG increases after voltage application of +1 V and decrease after applying -1 V, and these SHG response was in a symmetric shape about the baseline at 0 V.

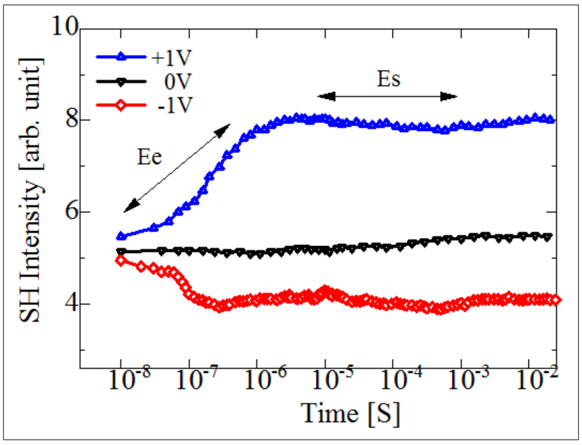

Fig. 2 SHG 500 nm BHJ without PEDOT: PSS

The transient EFISHG for the devices with the PEDOT: PSS layer (ITO/ PEDOT: PSS/BHJ/Al) are presented in figure3. In this figure, SH intensity decreased when negative voltage was applied at the SH wavelength of 500 nm. On the contrary, the SH intensity increased when positive voltage was applied. Consequently, the electric field is increased in PCPDTBT when positive voltage is applied, while the electric field is decreased when negative voltage is applied. In addition, the SHG intensity at 0 V increases in solar cell with PEDOT: PSS layer. This means that the internal field in OSCs is increased by the presence of PEDOT: PSS layer as indicated by EFISHG measurement (see Figure 2 and Figure 3). Thus, the internal field pointing from Al and ITO contributes the transfer of free holes to the ITO through PCPDTBT, while free electrons are transferred to Al electrode through PC71BM. For this reason, the flow of short-circuit current efficiently respects the proper direction thru the OSCs with the PEDOT: PSS layer in our results. The efficiency of OCSs is improved in our experimental results because the PEDOT: PSS contributes better connection between polymer and ITO electrodes.

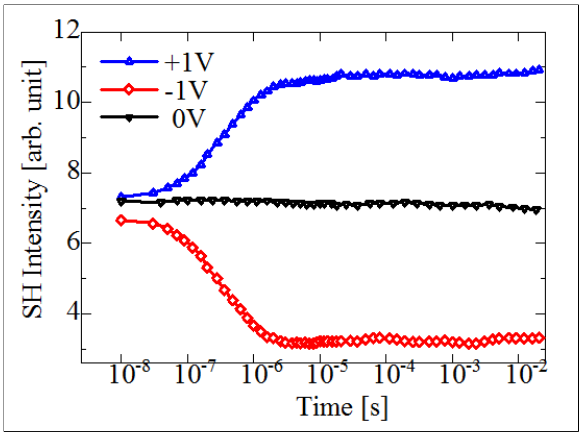

Fig. 3 SHG 500 nm BHJ with PEDOT: PSS

5. Conclusions

In this paper, the EFISHG techniques have been utilized in order to investigate the BHJ OSCs. We conclude that the selectively probed SHG measurement is beneficial to examine electron and hole behaviors in BHJ OSCs and delivers a direct way to study PEDOT: PSS layer effect on the transit time of the electrons and holes in BHJ OSCs.

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