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Different interfacial carrier behaviors between α-NPD and pentacene double-layer device with a polyimide blocking-layer by time-resolved optical second harmonic generation

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1. Introduction

Much attention has been paid to organic devices, e.g. organic field effect transistors (OFETs), organic solar cells, and organic light-emitting diodes (OLEDs) [1]. For typical organic devices such as OFET and OLED, the main carriers responsible for device performance are injected from electrodes. For OFETs, such as pentacene OFET, usually holes accumulate at the interface between active organic layer and gate insulator. They are conveyed along the channel. On the other hand, for OLEDs with a double-layer structure, electrons and holes injected from two facing electrodes, respectively, meet and recombine at the organic/organic interface for electroluminescence. However, a detailed understanding of carrier behaviours in these organic devices is not so straightforward owing to either the ambiguous properties of organic material itself or the multilayer structure and multi-species transportation of organic devices. Hence, to clarify the carrier behaviours in organic devices, experimental techniques that can probe the carrier motion are thus anticipated. Recently, we have developed a time-resolved optical second harmonic generation method (TRM-SHG) that can visualize carrier motion in organic devices [2]. With this technique, we successfully showed the diffusion-like carrier transport process in pentacene OFET [3] and the charging/ discharging process in α-NPD/Alq3 double-layer OLED [4, 5]. In addition, to avoid difficulties encountered in multi-species carrier transportation, using a double-layer structure with one polyimide blocking layer is of help. This structure makes it possible to analyze single carrier behaviour at interface. Results obtained by using this device structure will be helpful for analysing OLED as well as OFET.

In the paper, we prepared two kinds of sample devices with a structure of ITO/polyimide(PI)/α-NPD(or pentacene) /Au. Polyimide serves as a blocking layer for carrier injection from ITO electrode so that the carrier behaviors in active layer (α-NPD or pentacene) and interface can be investigated exclusively. Time-resolved electric field induced second harmonic generation (EFISHG) measurements were performed on the devices. By choosing the wavelength of fundamental laser, the voltage changes in active layers can be detected. In addition, with different bias condition, the carrier behaviors of different species can be studied as well.

2. Experimental

Polyimide layer was spin-coated onto ITO surface with pre-determined spin-coat conditions to make the thickness as 100nm. Then, active layer (α-NPD or pentacene) was evaporated onto polyimide surface in a vacuum with a pressure less than 10^{-5} torr. The thickness of active layer is 100nm. Last, Au electrode was evaporated onto active layer surface under vacuum. The time-resolved EFISHG measurement set-up is similar to our previous [4, 5]. The wavelength of impinging fundamental laser light was set at 860nm for pentacene and 820 nm for α-NPD, and second-harmonic light intensity $I(2\omega)$ generated at 430nm and 410 nm were probed for determining the electric-field change in active layer (pentacene or α-NPD) only. Furthermore, the incident angle between *p*-polarized fundamental laser beam and ITO surface was set at 45° to probe the electric field in active layer along the film thickness direction

3. Results and Discussion

Fig. 1 a and b show the SHG response for charging/ discharging processes in α-NPD/PI double layer, when α-NPD layer is forward and reverse biased, respectively. In forward bias (Fig. 1a), holes are injected into α-NPD layer from Au electrode while electron injection from ITO electrode is prohibited by polyimide blocking layer. During charging process, SH intensity from α-NPD layer starts from the 'zero level' (SH^0) and then increases to saturated value (SH^∞). After that it remains constant. On the other hand, during discharging process, SH intensity decreases from SH^∞ to SH^0 at around 10^{-7}s and remains unchanged thereafter.

Fig. 2 a and b show the SHG response for charging/ discharging processes in pentacene/PI double layer device when pentacene layer is forward and reverse biased, respectively. Though both α-NPD and pentacene are well-known hole transport materials, they show quite different carrier behaviours, implying different electrical properties. In forward bias (Fig. 2a) during charging process, SH intensity firstly decreases across the zero level, reaches minimum value and then returns to zero level at around 10^{-3}s. Correspondingly, during discharging process, the SH intensity decreases quickly to zero level after discharging of charges on electrodes. On the other hand, in reverse bias (Fig. 2b), the EFISHG responses for charging and discharging are quite similar to the case when pentacene was forward biased (Fig. 2a).

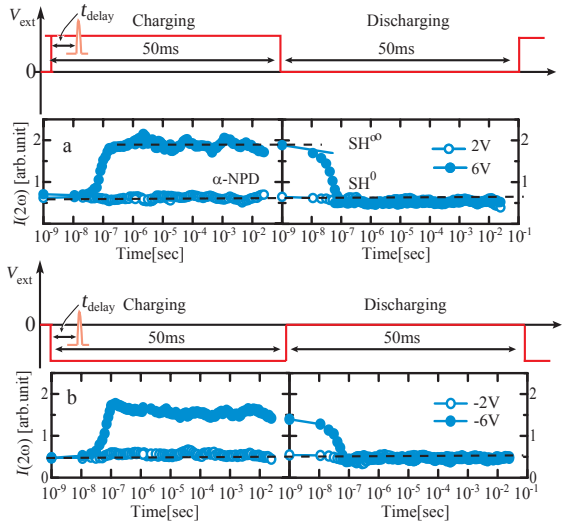

Fig. 1 SH intensity $I(2\omega)$ for charging and discharging processes of ITO/PI/α-NPD/Au double layer under a, forward and b, reserves bias

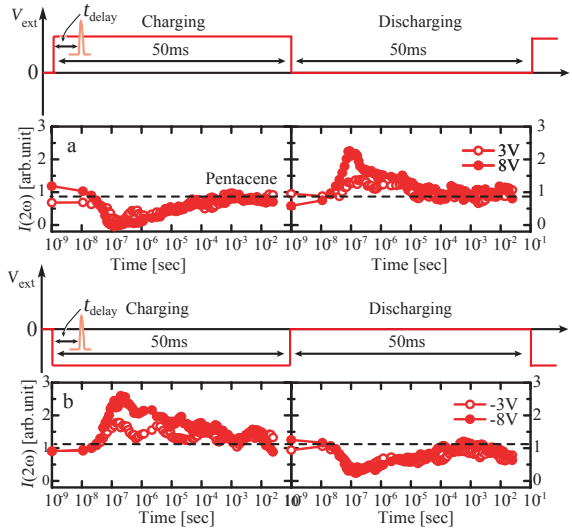

Fig. 2 SH intensity $I(2\omega)$ for charging and discharging processes of ITO/PI/pentacene/Au double layer under a, forward and b, reserves bias

It is found that the SHG responses of pentacene device between forward bias charging (FC) and reverse bias discharging (RD) are very similar to each other. On the basis of MW model and the intrinsic hole-transport properties of pentacene, it can be explained. In FC, holes are injected into pentacene from Au electrode and then transport through the pentacene layer. While in RD, although in RC process electrons accumulated at interface, during discharging holes are proposed to be injected from Au rather than drift back of accumulated electrons. In other words, it is holes that transport in pentacene layer during RD. Thus, in both FC and RD, holes are the mobile carriers, which should be responsible for similar SHG responses. Experiments and analysis accounting for this model is being carried out, e,g. experiments by changing duration of charging-and discharging time and voltage and charging time dependence on response time.

4. Conclusions

The carrier behaviours of pentacene and α-NPD thin film in charging and discharging process with different bias conditions were studied in a double-layer structure with polyimide as a blocking layer. Time-resolved EFISHG method well revealed the different carrier behaviours in α-NPD/PI and pentacene/PI double-layer device. For α-NPD/PI device, only charging/ discharging on electrodes were observed. However, for pentacene/PI device, carrier accumulation at interface due to MW effect was clearly observed. On the basis of MW effect, the charging and discharging processes between different bias conditions were discussed. It is suggested that different carrier species should be responsible for the observed SHG response in pentacene/PI double layer with different bias condition.

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