Thin Solid Films 518 (2009) 485–488

Contents lists available at ScienceDirect

Thin Solid Films

journal homepage: www.elsevier.com/locate/tsf

Transient charge accumulation in pentacene field effect transistor with silver electrode

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ARTICLE INFO

Available online 5 August 2009

Keywords:
Optical second harmonic generation (SHG)
Organic field effect transistor
Electrode contact

ABSTRACT

Time-resolved microscopic optical second harmonic generation (TRM-SHG) imaging was employed to study a transient charge accumulation in top-contact pentacene field effect transistor (FET) with Ag electrodes. It was demonstrated that the SHG signal at the edge of the Ag electrode decayed but remained in a steady state depending on biasing condition. An electric field formed in pentacene layer below Ag electrode activates the SHG, indicating the insufficient accumulation of injected carriers in the FET channel. By using the TRM-SHG technique transient change of the carrier density in the OFET is obtained.

1. Introduction

Electrical properties of organic devices such as organic field effect transistor (OFET) and organic electroluminescent (OEL) devices are influenced strongly by the organic-metal contact because injected carriers from electrode dominate the device operation. For instance, ambipolar behavior in OFET was observed using appropriate electrode metals, *i.e.* electrons and holes are injected from small and large work function electrodes, respectively [1–3]. The operation of organic devices is rather different from that of inorganic devices. Carriers injected from the source electrode play a key role at the beginning of the device operation [4]. Nevertheless, fabrication process such as deposition of organic layer and metal electrode significantly affects the device properties [5,6], and the adequate control of injection and transport processes of carriers is insufficient. This implies that an individual evaluation of these processes is a significant subject for the development of organic electronics. Results obtained from these investigations will help to reveal the device mechanisms and to improve a device performance.

We have been developing a time-resolved optical second harmonic generation (TRM-SHG) measurement for visualizing a carrier motion in organic thin film transistors [7] on the basis of electric field induced SHG (EFISHG). For the EFISHG process, applied electric field induces the effective polarization in materials, and it generates second harmonic light of the fundamental light. The SHG intensity is proportional to the external field as,

$$I(2\omega) \propto |\chi^{(3)}(-2\omega; 0, \omega, \omega)E(0)E(\omega)E(\omega)|^2 \quad (1)$$

where $\chi^{(3)}(-2\omega; 0, \omega, \omega)$ represents a third order nonlinear optical (NLO) susceptibility for the EFISHG process, and $E(0)$ and $E(\omega)$ represent the static electric field and electric field of light, respectively. Therefore the information about electric field in materials was successfully obtained by the EFISHG measurement [8,9], and in combination with the time-resolved technique, transient electric field migration could be observed by the TRM-SHG measurement. The transient electric field migration corresponds to the carrier motion in the devices, because the inject carriers are the source of the electric field ruled by the Gauss law. Direct observation of carrier enables us to evaluate the carrier mobility in the devices. Thus, transport process is effectively explored using the TRM-SHG. In contrast, electrical contact between electrode and organic materials is also examined by monitoring the SHG signal near the electrode, because the SHG signal is enhanced in proportion to the local electric field at the interface. In this sense, TRM-SHG measurement allows us to evaluate injection and transport processes in organic devices, individually and simultaneously. In this paper, electrode contact in pentacene FET is discussed on the basis of TRM-SHG results. In a steady state, incomplete disappearance of the SHG signal at the edge of Ag electrode represents that the electric field remained at the electrode edge. This indicates the insufficient accumulation of carriers below the Ag electrode. These results support the presence of injection barrier at pentacene–Ag contact for hole injection, owing to rather small work function of Ag.

2. Experiments

Samples used in the experiments were top-contact pentacene FET. Heavily-doped Si wafers were used as the base substrate, and they were covered with a 500 nm thick silicon dioxide (SiO_2) insulating layer. The material for the organic semiconductor layer was pentacene ($C_{22}H_{14}$) purchased from Tokyo Chemical Industry Co., Ltd., and was

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0040-6090/$ – see front matter
doi:10.1016/j.tsf.2009.07.028

used as received. Before the pentacene deposition, 100 nm thick of poly (methyl-methacrylate) layer was spin-coated, then the pentacene layer, approximately 100 nm thick, was deposited. The process pressure during deposition of pentacene was kept at less than 1×10^{-4} Pa, and the deposition rate was controlled at approximately 3 nm/min. After the deposition of pentacene, top-Ag electrodes (source and drain electrodes) with a thickness of 100 nm were deposited on the pentacene surface. The channel length (L) and width (W) were 25 μm and 3 mm, respectively. Field effect mobility, on/off ratio, and subthreshold swing of the device under investigation were 0.014 cm^2/V s, 10^4 and 13.7 V/decade, respectively. The FET was operated with the application of pulse voltages using a function generator (NF Corp.: WF1947) and a high speed bipolar amplifier (NF Corp.: HSA4101) during the FET operation.

Optical setup for the SHG measurement is shown in Fig. 1. The light source for the TRM-SHG measurement was an optical parametric oscillator (OPO:Continuum Surelite OPO) pumped by the third-harmonic light of a Q-switched Nd-YAG laser (Continuum:SureliteII-10). External Q-switch signal of YAG laser was also supplied from a function generator to synchronize between voltage pulse applied to OFET and laser pulse. Timing chart of voltage pulse applied to OFET and laser pulse is shown in Ref [7]. Fundamental light (wavelength was 1120 mn [10]) was focused onto the channel region of the OFET with normal incidence using an long working distance objective lens (Mitutoyo: M Plan Apo SL20×, NA = 0.28, W.D. = 30.5 mm). SHG signal generated from the FET was filtered by a fundamental-cut filter and an interference filter to remove fundamental and other unnecessary light. Finally, SHG signal was detected by a cooled CCD camera (Andor technology: DV420-BV). In this configuration, the polarization direction of the light was chosen in the direction corresponding to the channel direction (source–drain direction).

3. Results and discussion

Fig. 2 shows the change in the SHG intensity distribution along the FET channel with different delay times. An actual SHG image from the OFET channel is shown in the inset of Fig. 2 as an example. To take these images, positive pulse (100 V) was applied to the Ag source electrode, where the source electrode was defined as an electrode from which carriers are injected by the applied voltage pulse (see also Fig. 1b). The SHG intensity distribution was obtained from the SHG image by taking a line scan of the SHG image [9]. As shown in the figure, SHG peak shifted gradually from the source electrode to the drain, indicating that holes were injected from the source electrode, and then spread into the FET channel with an increase of delay time. That is, positive voltage applied to the Ag source electrode injects holes into pentacene. As discussed in our previous paper [11], the motion of the SHG peak followed a diffusion-like behavior, namely, the square of the SHG peak position was proportional to time. This result indicates that the driving force of the carrier motion is the space charge field by carriers injected from source electrode.

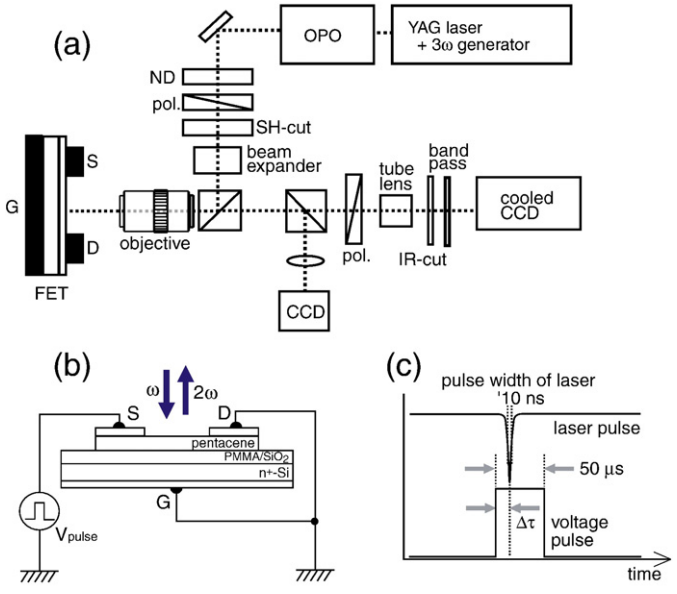

Fig. 1. Optical setup for the microscopic SHG measurement. The fundamental light is focused on the device using a microscopic objective lens. A cooled CCD camera was used to take SHG images. (b) Schematic image for the electrical connection and the sample structure. The gate and drain electrodes were connected to ground, and negative or positive voltage was applied to the source electrode. (c) Timing chart of voltage pulse applied to the OFET and laser pulse for the TRM-SHG measurement. Delay time is represented as $\Delta\tau$.

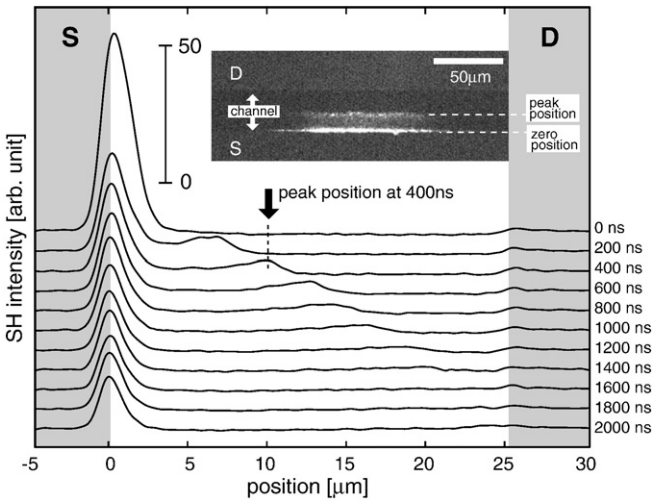

Fig. 2. Change in the SHG intensity distribution under a positive pulse application to the Ag source electrode. The inset is an actual SHG image from the OFET channel. For the SHG intensity distribution, the origin of the x-axis is set to the edge of source electrode ($x = 0$). Peak position of the SHG distribution at 400 ns is also indicated by black arrow as an example.

SHG intensity at electrode edge gradually decreases, and seems to be saturated at 2000 ns, hence, the SHG signal from electrode edge remains after reaching a steady state. Thus, the electric field below the source electrode decreases, but it remains even in a steady state. The carriers are accumulated at the pentacene/PMMA interface under the source electrode, and such carrier compensates the external electric field originating from the source voltage. This result indicates that accumulation of injected charges is not sufficient, and relaxation of electric field at the edge of electrode is not complete.

Above-mentioned insufficient accumulation of holes in the channel of the OFET with Ag electrodes was clearly confirmed by another transient experiment. Fig. 3 represents the change in the SHG intensity distribution under a positive pulse application to both the Ag source and drain electrodes. To take this image, the source and drain electrodes were connected with a wire and positive pulses were applied to the both electrodes simultaneously.

Therefore there was no potential difference between the source and drain electrodes. Interestingly, the SHG intensities at both the electrode edges gradually decreased and finally disappeared completely. For such an electrode connection, holes were injected from the source and drain electrodes, but they were not drained from the FET channel. Thus, the injected charges were gradually accumulated, and the electric field at the two electrodes edges decreased in accordance with the accumulated charges [8] at the interface.

Notes that by contrast, for the OFET with Au electrode, the SHG intensity at the electrode edge immediately disappears (less than 100 ns) after the voltage application. This indicates a rapidly decay of the electric field around the source electrode. Positive voltage applied to the Au source electrode injects holes easily into the pentacene