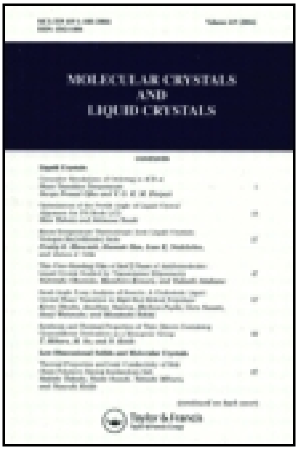

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Analysis of Organic Field Effect Transistors as a Maxwell-Wagner Effect Element: Measurement of Nano-Interfacial Polarization and Electric Field Distribution in Organic Film by Optical Second Harmonic Generation

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Analysis of Organic Field Effect Transistors as a Maxwell-Wagner Effect Element: Measurement of Nano-Interfacial Polarization and Electric Field Distribution in Organic Film by Optical Second Harmonic Generation

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A pentacene field-effect transistor (FET) is analyzed as a Maxwell-Wagner effect element, assuming that carriers injected from a source electrode are accumulated at the interface between the gate and pentacene, and they are then conveyed along the channel by the electric field formed between the Source and Drain electrodes. The optical second harmonic generation (SHG) from the channel well probes an electric field formed by injected carriers, and it is dependent on the off- and on-state of the FET channel. We conclude that the analysis of the pentacene organic FET as a Maxwell-Wagner effect element is acceptable.

Keywords: Maxwell-Wagner model; optical second harmonic generation; organic FET; pentacene

I. INTRODUCTION

Recent progress in the field of organic materials electronics motivates us to study organic devices for the ubiquitous use [1]. Among them, the discovery of high-mobility organic materials has called our attention [2]. Until now, many experimental and theoretical studies have devoted to the development of organic field effect transistors (OFETs), where pentacene is the organic material that is most often used. It has

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been revealed that the device performance of a pentacene FET is quite different from that of Si-FETs. For example, carriers injected from the source electrode dominate in the operation of OFET [3,4,5], a driving voltage is rather high in comparison with the voltage corresponding to the energy of the HOMO-LUMO gap of pentacene, ambipolar behavior is observed in pentacene FETs, and so on. It has been suggested that we must pay more attention to the nature of organic materials, when we discuss the device performance of organic FETs. The intrinsic carrier density of organic materials is generally low, and the establishment of thermodynamic equilibrium over the entire region of an organic FET is difficult, especially when high biasing voltages are applied. These facts suggest us that analyzing the organic device performance from the viewpoint of dielectric physics is very instructive. Carrier injection, carrier accumulation, and carrier transport are three basic processes to understand the organic FET device operation. In this article, we analyze the pentacene FET as a Maxwell-Wagner effect element [4,5] and show that the carrier injection from electrodes makes a significant contribution to the carrier transport. We also show experimentally that carriers injected from electrodes also make a significant contribution to the formation of a space charge field along the channel by means of the optical second harmonic generation (SHG).

II. ANALYSIS AND FET CHARACTERISTICS

A. Analysis as a Maxwell-Wagner Effect Element

According to the Maxwell's electromagnetic field theory, a charge q_s is accumulated at the interface between two dielectric materials with different relaxation times given by $\tau(=\varepsilon/\sigma$, ε: dielectric constant, σ: conductivity), when a current *I(t)* flows across the two-material interface. In more details, $\nabla\cdot\vec{j}=0$ with $\vec{j}=\varepsilon\vec{E}(\neq 0)$ is satisfied when the steady state current density $\vec{j}$ flows across the interface. Under this condition, the following relation concerning the electric flux density $\vec{D}$ is naturally derived:

$$\nabla\cdot\vec{D}=\nabla\cdot(\varepsilon\vec{E})=\nabla\cdot\frac{\varepsilon}{\sigma}\vec{j}=q_s(\neq 0). \tag{1}$$

This is the Maxwell-Wagner effect, indicating that q_s is accumulated at the interface, when the adjacent two materials have the different relaxation times. This theory is simple but introduces us the possibility that the excess charge Q_s is accumulated at the pentacene film-SiO_2 interface, when a charge is injected from the source electrode. This accumulated charge Q_s will be conveyed along the channel when an effective electric field V_{ds}/L is formed between the source and drain electrodes with a spacing of channel length L by the application of a biasing voltage V_{ds} between the source and drain electrodes. Therefore, we expect that a current I_{ds} given by

$$I_{ds} = \frac{Q_s}{L}\mu\frac{V_{ds}}{L} \tag{2}$$

flows across the channel when the amount of charge Q_s is continuously supplied to the interface without being trapped, while the charge Q_s is conveyed along the channel. The accumulated charge Q_s is an excess charge and approximately given by

$$Q_s = -\left(\frac{\tau_1}{\tau_2} - 1\right)C\,LW\left(V_{gs} - \frac{1}{2}V_{ds}\right) \tag{3}$$

with $C_g = \varepsilon_0\varepsilon_2/d_2$, because the voltage difference across the gate insulator is V_g at $x = 0$ (at the source electrode) and is $V_g - V_{ds}$ at $x = L$ (at the drain electrode). Here, τ_1 and τ_2 are the relaxation times of pentacene and gate insulator, and ε_2 and d_2 are the dielectric constant of the gate insulator and the thickness, respectively; L and W are the channel length and channel width, respectively. The derived relations (2) and (3) are very similar to the relation for I_{ds} that can be derived by basing on a gradual channel model under the assumption that the organic film is functioning as a semiconductor [6]. For semiconductor FETs, carriers accumulated at the interface are supplied from the bulk, not from the source electrode, and their statistical distribution in the state of thermodynamic equilibrium is expressed by the Fermi-Dirac distribution function. On the other hand, for organic films we discuss here, carriers are supplied from the source electrode, and they do not need to be in the thermodynamic equilibrium state, though the current flows in a steady state. As such, it should be noted that the derived relation (2) with (3) is valid even when V_{ds} is very large. Of course, this relation represents the most ideal steady state case, i.e., the case without carrier trapping, etc. On the other hand, in actual FETs, carrier traps exist in organic materials, in gate insulators, and at the organic material-gate insulator interface. In the case of the pentacene FETs we discuss here, carriers injected from the source electrode are accumulated at the interface, and they are then conveyed along the channel. Therefore, we need to consider that some of the carriers will be trapped in organic materials and especially at the interface. When the carrier trapping at the interface makes a significant contribution, this leads to the establishment of a threshold voltage V_{th}, as well as to a decrease of the amount of mobile carriers. Consequently, relations (2) and (3) can be rewritten as

$$I_d = \theta\left(1 - \frac{\tau_1}{\tau_2}\right)C_g\left[V_{gs} - V_{th} - \frac{1}{2}V_{ds}\right]\mu\frac{V_{ds}}{L}W, \tag{4}$$

where θ is the ratio of the accumulated charge Q_s and mobile charges Q_m. As usual, $0 \le \theta \le 1$, when a quasi-thermodynamic equilibrium is established between mobile carriers and trapped carriers [7]. On the other hand, V_{th} is given by

$$V_{th} = \frac{Q_t}{C_g}, \tag{5}$$

where Q_t is the trapped charge given by $Q_t = Q_s - Q_m$. It follows from Eq. (4) that the effective mobility μ_{eff} given by $\mu_{\text{eff}} = \theta(1 - \tau_1/\tau_2)\mu$ is dependent on the carrier trapping at the interface. On the other hand, when the electric field is high, injected carriers are swept out from the channel and transported in the direction from the source to drain electrodes, as soon as they reach the pentacene-gate insulator interface. Therefore, I_{ds} is limited by the transit time of carriers along the channel, $\tau_r = L/(\mu E) = L^2/(\mu V_{ds})$. In more details, when $\tau_r < \tau_{inj}$ (the time required for the carrier accumulation at the interface), the total charge that can be conveyed along the channel is limited to $Q_m = -\theta(\tau_1/\tau_2 - 1)C_g LW(V_g - V_{th} - 1/2V_{ds})$ (=const, $V_{sat} = E_c L$) at $V_{ds} = V_{sat}$ in the presence of trapping. In the case without trapping, $\theta = 1$ and $V_{th} = 0$. The limitation of the accumulated charge leads to the saturation of the current at the high electric field region $E \le V_{sat}/L$. Interestingly, the current increases proportionally to the gate voltage V_g in this region see Eq. (4). This type of the current-flow is seen for polydiacethylene FETs [8]. On the other hand, for the pentacene FET, we conclude that this accumulation does not happen because of $\tau_r > \tau_{inj}$ [5]. However, the saturation happens when $V_{ds} > V_g - V_{th}$. In this case, at $x = x_{\max} \le L$, $V_{xs} - V_{th} = V_g$ is satisfied. This means that the accumulation of charge at the interface is confined in the region $0 \le x \le x_{\max}$. This boundary condition eventually limits the amount of the accumulated charge and then finally leads to the saturation of the current to

$$\begin{aligned} I_d &= \theta\left(1 - \frac{\tau_1}{\tau_2}\right)C_g\left[V_g - V_{th} - \frac{1}{2}V_p\right]\mu\frac{V_{ds}}{L}W \\ &= \theta\left(1 - \frac{\tau_1}{\tau_2}\right)C_g\mu\frac{(V_g - V_{th})^2}{2}\frac{W}{L}. \end{aligned} \tag{6}$$

Here, in the case without carrier trapping, $\theta = 1$ and $V_{th} = 0$.

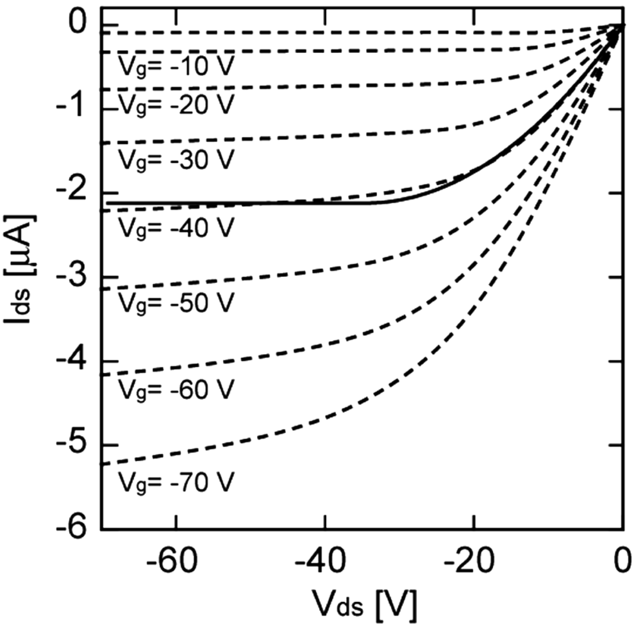

FIGURE 1 Pentacene FET characteristics. Dashed and solid lines represent the experimental and fitting results.

B. FET Characteristics

Figure 1 shows a typical example of the FET characteristics of our pentacene FET. In the figure, the sample structure is illustrated. The gate insulator is SiO_2, and Au is used as the source and drain electrodes. The pentacene thickness is 200 nm, channel length L is 100 μm, and channel width W is 3 mm. The FET characteristics are typical and well reproduced by using Eqs. (4) and (6) under the assumption of V_{th} and effective mobility μ_{eff}, e.g., $V_{th} = -0.27\,\text{V}$ and $\mu_{\text{eff}} = 1.3 \times 10^{-2}\,\text{cm}^2/\text{Vs}$ at $V_g = -40\,\text{V}$. It should be noted here that a voltage higher than 10 V is required for the drive of FETs. Interestingly, this FET characteristic is well explained by assuming that carriers are accumulated at the interface due to the Maxwell-Wagner effect, and then they are transported.

III. SHG GENERATION

The successful explanation of our FET characteristics as a Maxwell-Wagner effect element forces us to consider a space charge field formation along the FET channel. This means that the average electric field $\overline{E}$ along the FET channel is given by V_{ds}/L, but the actual electric field along the channel is dependent on the position. As we described in our previous article [11], the basic idea adopted in the derivation of the current I_{ds} given by Eq. (4) is the same with the idea adopted in the derivation of SCLC flowing across an organic film [7]. Taking into account this similarity, we consider that the electric field around the injection electrode (the source electrode) is relaxed in proportion to the accumulated charge along the channel. Therefore, it is expected that the electric field across the channel is decreased due to the carrier injection, especially the electric field at the injection electrode is relaxed. In other words, this occurs in the on-state of the FET due to the carrier injection from the source electrode followed by the charge accumulation at the interface between pentacene and the gate insulator, rather than in the off-state of the FET, because there is no carrier injection and only the Laplace electric field is formed along the FET channel depending on the electrode arrangement. Taking this into account, we carried out the macroscopic and microscopic SHG experiments to examine a deformation of the space charge field along the OFET channel [9].

Figure 2 shows the response of the SHG with applying voltages at a wavelength of 1120 nm. The applied voltage sequence is as follows: Region 1 ($V_d = 0\,\text{V}$, $V_g = 0\,\text{V}$) → Region 2 ($V_d = -90\,\text{V}$, $V_g = 0\,\text{V}$) → Region 3 ($V_d = -90\,\text{V}$, $V_g = -90\,\text{V}$) → Region 4 ($V_d = -90\,\text{V}$, $V_g = 0\,\text{V}$) → Region 5 ($V_d = 0\,\text{V}$, $V_g = 0\,\text{V}$), where V_d and V_g represent the applied voltages between the source and drain electrodes and the source and gate electrodes, respectively, with respect to the source electrode ($V_s = 0\,\text{V}$. Region 1 is the initial state, Region 2 and 4 correspond to the off-state, and Region 3 and 5 correspond to the on-state. As shown in the figure, the small SHG signal in region 1 was enhanced by applying the source-drain voltage in region 2. Since the channel conductance is generally low at zero gate bias (off-state), the SHG process is induced by the electric field applied to the insulating pentacene layer, i.e., typical EFISHG. The centrosymmetry of the molecule is broken by the external field, i.e., extensively delocalized π-electrons are disordered and effective dipoles are induced. According to the SHG spectrum of the vacuum-deposited pentacene film and the results of molecular orbital calculations, the allowed excited states, which lead the absorption peak and a resonance enhancement of the EFISHG process, are located around 1120 and 1320 nm. In region 3, the enhanced SHG signal drastically decreases with applying the gate voltage, and then the SHG intensity completely recovered after the gate voltage turned off. Interestingly, this remarkable change coincides well with

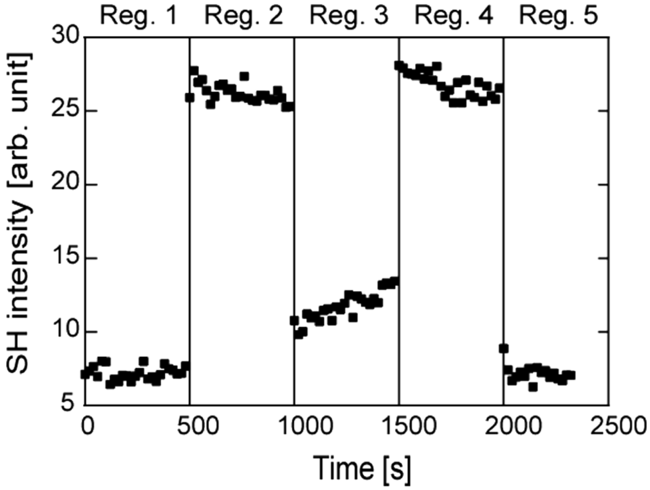

FIGURE 2 Typical response of the SHG with applying voltages [9]. Region 1 ($V_d = 0\,V, V_g = 0\,V$) → Region 2 ($V_d = -90\,V, V_g = 0\,V$) → Region 3 ($V_d = -90\,V$, V, $V_g = -90\,V$) → Region 4 ($V_d = -90\,V, V_g = 0\,V$) → Region 5 ($V_d = 0\,V, V_g = 0\,V$).

the FET characteristics. That is, a decrease of the SHG intensity corresponds to an increase of the drain current due to the FET operation (the on-state).

Generally, the carrier concentration of an organic semiconductor is considered to be intrinsically low, and the carriers that form the channel are supplied from the source or drain electrode in organic materials. In the OFET, the switching-on and -off of the devices correspond to the transition between the conducting and insulating states of the material at the channel region. For the SHG process [10], the second-order nonlinear polarization, $P_i = \chi_{ijkl}\, E_j(0)\, E_k(\omega)\, E_l(\omega)$, becomes the source of a SHG signal. Here $E_i(0)$ and $E_{i,j}(\omega)$ are the static electric field due to the source-drain voltage and the electric field of the fundamental light, respectively, and χ_{ijkl} represents the nonlinear susceptibility of the molecules. In the off-state, the SHG process is active, but in the on-state, injected carriers contribute to the space charge field formation, and this leads to a decrease of the nonlinear polarization induced in the off-state. This is a possible process to change the SHG intensity during the FET operation. In the on-state of the FET, an increase of the channel conductance reduced the polarization that contributes to the SHG. In other words, the SHG measurement also supports the suggested operation mechanisms of the OFET, i.e., the channel formation due to the injected carriers. This means

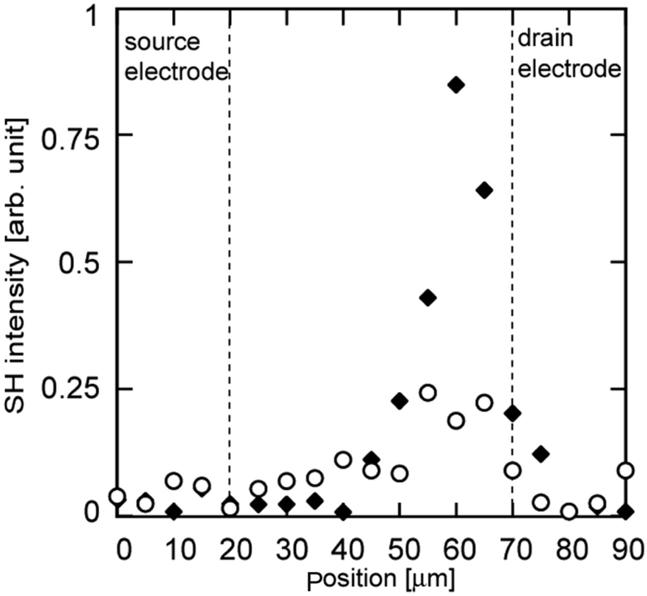

FIGURE 3 Response of the microscopic SHG along the FET channel with applying voltages. Filled squares and open circles represent the SHG intensity from each spot position in the off- and on-states of the FET.

that our pentacene FET is functioning as an injection-type FET, and the analysis developed in Section 2 is helpful within the constraints of our experiments. As the SHG can probe the space charge formation in the FET channel, we can determine the electric field distribution of the on- and off-states of the FET channel. Figure 3 shows the result of microscopic SHG measurements using a laser beam with a spot size of 10 µm [12]. Finally, we should note that our pentacene FET shows the hysteresis behavior in its FET characteristics during the measurement, depending on the time of applying the gate voltage, etc. This would be related to the trapping and detrapping behaviors of carriers injected from the source electrode. The SHG measurements can also probe these phenomena.

IV. CONCLUSION

The pentacene FET was analyzed as a Maxwell-Wagner effect element, by assuming that carriers accumulated at the pentacene-gate insulator are excess carriers injected from the source electrode. The relation for the current was derived, by taking into account the carrier injection, carrier accumulation, and carrier transport. It was shown that the derived relation is valid even when a high voltage is applied to the FET, and that the space charge field is formed along the channel in the on-state due to the carrier injection from the source electrode. The optical second harmonic generation from the channel was dependent on the off- and on-state of the FET channel, by supporting the operation mechanism of the pentacene FET as a Maxwell-Wagner effect element.

REFERENCES

[1] Rogers, J. A. & Bao, Z. (2002). *J. Poly. Sci. Part A*, *40*, 3327.
[2] Katz, H. E. (2004). *Chem. Mater.*, *16*, 4748.
[3] Ogawa, S., Naijo, T., Kimura, Y., Ishii, H., & Niwano, M. (2005). *Synth. Metals*, *153*, 253.
[4] Manaka, T., Lim, E., Tamura, R., & Iwamoto, M. (2006). *Thin Solid Films*, *499*, 386.
[5] Lim, E., Manaka, T., Tamura, R., & Iwamoto, M. (2006). *Jpn. J. Appl. Phys.*, *45*, 3712.
[6] Sze, S. M. (1981). *Physics of Semiconductor Devices*, Wiley: New York.
[7] Lampert, M. A. & Mark, P. (1970). *Current Injection in Solids*, Academic Press: New York.
[8] Zou, G., Lim, E., Tamura, R., Kajimoto, N., Manaka, T., & Iwamoto, M. (2006). *Jpn. J. Appl. Phys.*, *45*, 6436.
[9] Manaka, T., Lim, E., Tamura, R., & Iwamoto, M. (2005). *Appl. Phys. Lett.*, *87*, 222107.
[10] Bloembergen, N. (1996). *Nonlinear Optics*, World Scientific: Singapore, Chap. 3.
[11] Tamura, R., Lim, E., Manaka, T., & Iwamoto, M. (2006). *J. Appl. Phys.*, *100*, 114515.
[12] Manaka, T., Lim, E., Tamura, R., & Iwamoto, M. (2006). *Appl. Phys. Lett.*, *89*, 072113.