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Field dependence of the space-charge limited current in polycrystalline pentacene

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Abstract

Field dependence of the space-charge limited current (SCLC) in polycrystalline pentacene was studied with a Au / pentacene / Au diode. The data show the similarity between transport properties of polycrystalline pentacene and polymer semiconductors.

1. Introduction

Owing to a rapid improvement of organic semiconductor materials [1], the mobility of oligomer-based thin film transistors has increased drastically [2]. However, there is still a lack of understanding of fundamental transport properties of those polycrystalline oligomer materials. On the other hand, it is well-know that polymer-based diodes show field-dependent mobility and its current density can be expressed as following [3, 4]:

$$J = \frac{9}{8}\varepsilon\mu_0\frac{V^2}{L^3}\exp\left(0.891\gamma\sqrt{\frac{V}{L}}\right) \quad (1)$$

$$\mu_0 = \mu_0^*\exp\left(-\frac{E_A}{k_B T}\right) \quad (2)$$

$$\gamma = B\left(\frac{1}{k_B T}-\frac{1}{k_B T_0}\right), \quad (3)$$

where ε is the permittivity of the material, k_B is Boltzmann's constant, and L is the thickness of the diode. Zero-field mobility μ_0 is of thermally-activated type with a pre-exponential factor μ_0^* and an activation energy E_A. γ is called Poole-Frenkel factor in which B and T_0 are constant.

In this paper, we study the transport properties of poly crystalline pentacene sandwiched with two Au electrodes and analyze them by using the SCLC theory with a field-dependent mobility which was already developed for polymer diodes [3, 4].

2. Experimental procedure

Au / pentacene / Au diodes were fabricated on a sapphire substrate by successively depositing a bottom contact [Ti (10 nm) / Au (40 nm)], a semiconductor layer [pentacene (1030 nm)], and a top contact [Au (40 nm)] through a metal mask. Pentacene was deposited at 0.1 Å/s under a vacuum pressure of about 5 x 10^{-5} Pa. The device area (600 µm x 600 µm) is defined as the overlap between the top and bottom electrodes.

The *J-V* measurements were performed in vacuum in a temperature range of 40 – 300 K with a semiconductor parameter analyzer (Keysight 4156C). A bias voltage was applied to the bottom contact against the top contact. To prevent any joule heating [5] and possible electric breakdown [6] in the sample, the current was measured as soon as a target voltage was applied. Immediately after the measurement, the bias voltage was reduced to 0. The interval between the current measurements was 1 sec.

3. Results

Temperature dependence of the transport properties is shown in Fig. 1, where J/V^2 is plotted versus $\sqrt{V}$ and broken lines are the field-dependent SCLC according to Eq. (1). It is clearly seen that there is a voltage region where J/V^2 is proportional to $\sqrt{V}$ and its slope becomes larger with decreasing temperature. All J/V^2 curves, however, deviate from the corresponding broken line both at the lower and higher voltage regions.

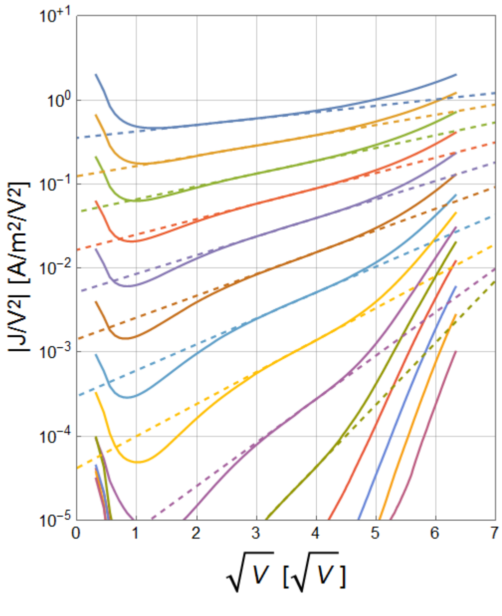

Fig. 1 J/V^2 versus $\sqrt{V}$ from 300 K (top-most line) to 40 K (bottom-most line) at the interval of 20 K. The broken lines are fits of Eq. (1) to experimental data.

At low voltages, J is linearly proportional to V (Ohmi's-Law Regime), which leads to a decrease in J/V^2

against $\sqrt{V}$ in the low voltage region. There are two possibilities which give rise to the ohmic conduction. One is that the current of the thermally generated holes dominates that of the holes injected from the contact at low voltages [7]. It is, however, unlikely the case because the pentacene film used in this study is not doped intentionally. The other is that the diffusion current plays an important role in the transport at low voltages [4, 8]. The transport properties at low voltages are affected substantially by the existence of charge traps, the injection efficiency and the built-in potential corresponding to the difference between the work functions of the anode and cathode [9]. Therefore it is still difficult to analysis the data quantitatively.

With increasing voltage, J/V^2 at high temperatures (> 260 K) follow field-dependent SCLC. One of the important requirements for SCLC is that the current is not limited by the injection rate but the bulk transport. Therefore the observation of SCLC means that the Au-pentacene contact can be considered as ohmic at high temperatures. On the other hand, J/V^2 curves at low temperature go below the broken lines, change to a rapid increase, and finally merge with field-dependent SCLC, as predicted with numerical device simulation [4]. At low temperatures, the number of holes which overcomes the injection barrier is quite low. Therefore the current at ~1 V is limited by the injection. However, this number increases with increasing voltage further due to the Schottky effect and eventually the current is limited by the bulk transport again.

To characterize the current properties in the field-dependent SCLC regime, both the zero-field mobility μ_0 and Poole-Frenkel factor γ were estimated by fitting the data with Eq. (1). As seen in Fig. 2, μ_0 at high temperatures (>200 K) gives a straight line in an Arrhenius plot, which means the transport is dominated by a thermally-activated process. The activation energy and pre-exponential factor are estimated to be 0.3 eV and 12 cm^2/Vs, respectively. On the other hand, γ is inversely proportional to temperature according to Eq. (3) with B = 2.2 x 10^{-5} eV$(m/V)^{1/2}$ and T_0 = 435 K. These values are very close to those obtained by Blom for poly(paraphenylene vinylene) (PPV) [3]: B = 2.9 x 10^{-5} eV$(m/V)^{1/2}$ and T_0 = 600 K. There are two possibilities which give rise to $\exp\left(\gamma\sqrt{V/L}\right)$ -dependence. One is the enhancement of the thermal emission from traps to the valence band is enhanced by applying an electric field (the Poole-Frenkel effect). It is convincing because the emission rate determines the overall transport on the basis of multiple trapping and releasing model. In this case, however, B should be $(q^3/\pi\varepsilon_0\varepsilon_r L)^{1/2}$=4.2 x 10^{-2} eV$(m/V)^{1/2}$ and there is no straightforward explanation for the offset which is often observed in amorphous organic semiconductors. The other is hopping between localized sites that are subject to both positional and energetic disorder. As far as we know, no consensus has been obtained yet on this matter.

At high voltages (>25 V), J/V^2 deviates from the $\exp\left(\gamma\sqrt{V/L}\right)$–dependence and approaches to $\exp(-kL/V)$ with decreasing temperature where k is a constant. This field-dependence has been often attributed to Fowler-Nordheim tunneling through the injection barrier [10, 11]. As stated above, however, the bulk transport is dominant at high temperatures in our device, and it should not be ignored even at low temperatures because the mobility at low temperatures is supposed to be extremely low. The detail study of transport properties at this voltage region is still under survey.

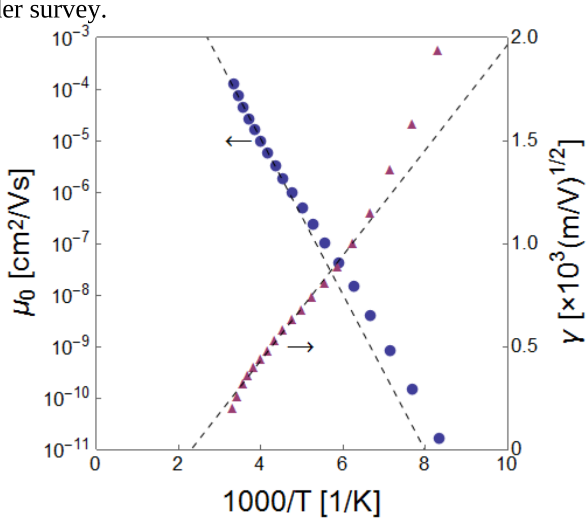

Fig. 2 Zero-field mobility μ_0 marked with circles and Poole-Frenkel factor γ marked with triangles. Dotted lines are fits of Eqs. (2) and (3).

4. Conclusions

We studied the transport properties of polycrystalline pentacene by using the theory of field-dependent SCLC and estimated the zero-field mobility and Poole-Frenkel factor.

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