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# Device Physics of Organic Transistors

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## 1. Introduction

Here we present our recent studies of organic field-effect transistors, especially focusing on the understanding of microscopic charge transport through channel organic semiconductor layers and also on the essence of the use of organic semiconductors. Firstly we discuss the interface charge transport as investigated by field-induced electron spin resonance spectroscopy (FI-ESR) [1,2]. In organic transistors, the carrier concentration is quite low of less than one carrier per one hundred molecules at maximum. In such a case, the charge transport is considerably affected by imperfections or impurities existed in ordered crystals or films with regularly aligned organic molecules. We recently demonstrated by the FI-ESR spectroscopy that the charge transport in high mobility organic transistors (> 1cm$^2$/Vs) is understood in terms of the multiple trap-and-release (MTR) transport through the shallow trap states. It is concluded that the nearly perfect alignment of molecules is more important for obtaining the high device performance than the size of intermolecular interactions. Next, we present a novel inkjet printing technique for manufacturing highly-uniform organic semiconductor thin films [3]. It is shown that single-crystal films with nearly perfect molecular alignments can be manufactured by a combination of two kinds of inks, *i.e.* semiconductor solution ink and antisolvent ink. We also discuss the importance of self-organizing nature of the organic molecules for the higher uniformity of the semiconductor layers.

## 2. Microscopic Charge Transport in Organic Transistors[1,2]

In organic transistors, it is possible to observe the FI-ESR signal from to the tiny amount of carriers that are accumulated only at the semiconductor/insulator interfaces by the gate voltage. This feature originates in the fairly weak spin-orbit interactions in organic material, resulting in a considerably sharp FI-ESR spectrum. In particular, the FI-ESR spectra allow us to probe the microscopic carrier motion without the current measurements. When we measure the temperature dependence of the FI-ESR spectra at a fixed gate voltage (*i.e.* fixed carrier concentration), we can clearly observe the narrowing of FI-ESR linewidth due to the carrier motions by increasing the temperature, which is in sharp contrast to the conventional temperature dependence of linewidth due to the spin relaxation processes. The analysis of temperature-dependent linewidth enables us to evaluate the average residence time of carriers at respective trap sites. By the comparison of the average residence time with carrier velocity as estimated by the mobility, we successfully demonstrated the trap-and-release transport of carriers where the carriers repeat the long-time trapping at respective trap sites and the thermally-activated releasing from the trap sites to move rapidly until the subsequent trapping.

In the lower temperature range below 50 K, all the carrier motions are frozen at the trap sites where the temperature variation of linewidth becomes unobserved. The devices cannot also be operated in the low temperature range. Detailed analyses of high-precision low-temperature FI-ESR spectra allow us to obtain the information how the carrier trap states extend spatially over several molecules. In the analyses, we made use of the relationship between extension of trap states and ESR linewidth through the hyperfine mechanism. By the analyses, it is shown that the trap states in pentacene thin-film transistors can be classified into the following three groups: deep trap states with spatial extension of ca. 1.5 molecules, relatively shallow trap states which extend over ca. 5 molecules, and shallower trap states which extends over 6 - 20 molecules.

## 3. Inkjet Printing of Single-Crystal Organic Transistors[3]

The relatively weak intermolecular interaction (ca. 100 meV) enables production of thin films of organic semiconductors at ambient pressure and at room temperature. The organic semiconductors can be also controlled in its solubility in organic solvents by various chemical modifications of the organic molecules. These features make the organic semiconductors most promising in realizing the "printable electronics" in which the printing technologies are applied into the device processing. On the other hand, it is quite important to eliminate the disorders or imperfections in channel organic semiconductors, since they become the origin of carrier traps and thereby deteriorate the device performance, as is discussed in Sec. I. So our main subject in the “printable electronics” is to manufacture highly-ordered organic semiconductor films by solution-mediated printing processes. Recently, we succeeded in fabricating organic semiconductor films with extraordinary high uniformity by introducing the renowned concept of “antisolvent crystallization” into the microliquid inkjet printing process.

In the newly developed “double-shot inkjet printing (DS-IJP)” process, we individually deposit two kinds of inks, *i.e.* semiconductor ink which dissolves an organic semiconductor in an organic solvent and crystallization ink of the antisolvent which does not dissolve the organic semiconductor. The saturated state for the organic semiconductor is formed in the mixed deposited droplet on top of the substrate. By the method, solidification of semiconductor layers and evaporation of the organic solvents can be temporally discriminated, which results in the growth of uniform semiconductor layers. In particular, we found that the adequate selection of organic solvents and printing conditions for organic semiconductor of $C_8$-BTBT enables fabrication of single-domain crystalline thin films over a wide region. The field-effect transistors composed of the films exhibit high mobility of 16.4 $cm^2/Vs$ in average in the saturated regime, and high on/off ratio of 5 – 7 orders of magnitude.

**4. Summary**

Based on the microscopic studies with the FI-ESR spectroscopy, we have shown that the charge transport in organic transistors is dominated by charge traps which are associated with the disorder or imperfections in channel organic semiconductor layers. It is concluded from this that it is quite important to manufacture organic thin films with nearly perfect molecular alignments for obtaining the high device performance. We have also presented the novel technique for the print production of organic semiconductor layers with extraordinary high uniformity and high molecular order by the novel inkjet printing technique with use of two kinds of inks. Based on the technique, we successfully realized the highest performance organic thin-film transistors.

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