Localized-State Distributions in Polymer Transistors Studied by a Field-Effect Method

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1. Introduction

Organic field-effect transistors (OFETs) have attracted growing attention in recent years because of their possible application in flat-panel displays, electronic papers and RFID tags. It is well known that the electrical properties of OFETs depend crucially on processing conditions and that disorder in the channel influences strongly the OFET performances. It has been found that the thermal treatment and the modification of substrate surfaces using self-assembled monolayers (SAMs) lead to the enhancement of carrier mobility [1]. However, the influences of such treatments on localized states in OFETs has not been fully investigated. For the determination of distributed localized states in disordered semiconductors, a number of methods have been proposed so far. Among these methods, field-effect (FE) methods such as the Spear technique [2] and the Suzuki technique [3] are very useful to investigate the localized states in OFETs because the localized-state distributions can be determined from transfer characteristics of FET devices.

In this paper, we report on the determination of localized-state distribution in OFETs by the FE method. We examined the validity of the methods using transfer characteristics computed for model localized-state distributions. By the FE methods, we investigate the effect of surface treatments with SAMs on localized-state distributions in OFETs fabricated using a poly(3-hexylthophene) (P3HT) as the semiconductor.

2. Theory

In the Suzuki technique [3], the localized-state distributions are determined by analyzing the increase of conductance when the voltage is applied to the gate electrode of metal-insulator-semiconductor structures. The band bending u(x) from a flat-band condition is solved using the Poisson's equation. Using a zero-temperature approximation, the density of localized state N(E) can be given by

$$N(E_F + u_s) = \frac{\varepsilon_s}{2} \frac{\partial^2}{\partial (eu_s)^2} \left[\frac{du}{dx} \Big|_{x=0} \right]^2 \quad (1)$$

$$\frac{du}{dx} \Big|_{x=0} = -\frac{\varepsilon_{ox}}{\varepsilon_s} \frac{V_G - V_{FB}}{d_{ox}} \quad (2)$$

where x is the distance from the insulator surface, E_F is the Fermi level, ε_{ox} is the dielectric constant of an insulator, d_{ox} is the thickness of an insulator and V_{FB} is the flat-band

voltage, and u_s and ε_s are the surface potential and dielectric constant of a semiconductor, respectively. The relation between the increase of conductance δG and band bending δu_s is expressed by

$$\delta u_s = \frac{\delta G}{G_0} \frac{d_s}{d_{ox}} \frac{\varepsilon_{ox}}{\varepsilon_s} \frac{V_G - V_{FB}}{\exp[u_s / kT] - 1}$$
(3)

where d_s is the thickness of a semiconductor. There, the localized-state density N(E) can be calculated using Eqs. (1) - (3) from the conductance by FE measurements.

3. Numerical Simulations and Experiments

In order to examine the validity of the FE method for determining localized-state distributions, we applied the FE methods to transfer characteristics calculated using a device simulator ATLAS (Silvaco). For model localized-state distributions, exponential distributions with different characteristic temperatures T_0 were used because they were often found in disordered organic semiconductors. The localized-state distributions were determined by the Suzuki technique from simulated transfer characteristics and then were compared with the input localized-state distributions.

In the experiment, top-contact FET devices were fabricated on a heavily doped silicon wafer with a thermally grown 365-nm thick SiO₂ layer, whose surfaces were chemically modified by SAMs. For SAMs, we used alkyltrichrolosilanes with different chain lengths such as ethltrichrolosilane (ETS), hexyltrichrolosilane (HTS), dodecyltrichrolosilane (DTS) and octadecyltrichlorosilane (OTS). The SAM treatments were preformed by immersing substrates in a 10 mM solution of anhydrous toluene at room temperature for 24 h. As the semiconductor layer, regioregular P3HT was spin-coated on the SAM surfaces from 1 wt% toluene solution. For comparison, the P3HT FETs with the bare SiO₂ surfaces were also fabricated.

4. Results and Discussion

Numerical simulation of the FE method

Figure 1 shows the localized-state distributions determined by the Suzuki technique from transfer characteristics calculated for exponential distributions with different T_0 . It can be seen that although the determined distributions deviates from the input distribution in the vicinity of the Fermi level, localized-state distributions are well reconstructed in the shallower energy side. It is found that localized-state distributions can be determined correctly except for deeper energy regions.



Fig. 1. Localized-state distributions determined by the Suzuki technique from simulated transfer characteristics. The solid lines represent input distributions.



Fig. 2. Transfer characteristics of P3HT FETs with the SAM-treated SiO_2 surfaces and with the bear SiO_2 surface.

Localized-state distributions of P3HT FETs

Figure 2 shows the transfer characteristics of the P3HT FETs with and without the SAM treatments. The drain current in saturation regime and mobility increase with length of the alkyl chain and show a tendency to be saturated for the longer-chain SAMs. The water contact angle measurements reveal that a longer alkyl chain length yields a higher contact angle. It has been thought that the mobility enhancement by the surface treatment is attributed to improvement of structural ordering of P3HT molecules on the hydrophobic surfaces.

The localized-state distributions of the P3HT FETs determined by the Suzuki technique from the transfer characteristics in Fig. 2 are shown in Fig. 3. We find that the localized-state distributions of all devices are well characterized by an exponential distribution with $T_0 \sim 500$ K and the surface treatment affect the density of localized states. The exponential localized-state distribution determined by the FE method is consistent with that deduced from time-of-flight (TOF) measurements [4].

We show in Fig. 4 the plots of the mobility and the density of localized states at 0.3 eV below the E_F versus the number of alkyl units. It can be seen that there is a clear correlation between mobility and localized-state density: the mobility increases with decreasing density of localized states. This result indicates that mobility improvement by the surface treatment using SAMs is attributable to the decrease of carrier trapping in the localized states. We note that the T_0 , *i.e.*, the energy width of the localized-state dis-

tribution, is not influenced by the SAM treatments. These results suggest that the SAM treatment of P3HT FETs increases the size of P3HT crystalline domains without changing structural ordering of P3HT in the domains but does not affect energetic disorder at the grain boundaries.



Fig. 3. Localized-state distributions of P3HT FETs with different dielectric surfaces determined by the Suzuki technique from transfer characteristics in Fig. 2.



Fig. 4. Mobility and localized-state density versus the number of alkyl units.

5. Conclusions

We have investigated that the localized-state distributions of P3HT FETs by the field-effect method. We find that the method gives accurate localized-state distributions in the energy regime close to the mobility edge. We also find that P3HT have exponential localized-state distributions with T_0 =500 K. The surface treatment with the SAMs affects the density of localized states, not characteristic temperature of the exponential density of states. The mobility improvement by the surface treatment is attributable to the decrease of charge trapping in localized states.

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