Local charge carrier mobility in disordered organic field-effect transistors

C. Tanase a,*, E.J. Meijer b,c, P.W.M. Blom d, D.M. de Leeuw b

a Materials Science Centre and DPI, Physics of Organic Semiconductors, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands
b Philips Research Laboratories, 5656 AA Eindhoven, The Netherlands
c Department of Applied Physics and DIMES, Delft University of Technology, Lorentzweg 1 2628 CJ Delft, The Netherlands
d Materials Science Centre, Physics of Organic Semiconductors, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands

Received 23 September 2002; received in revised form 20 March 2003; accepted 1 April 2003

Abstract

In conventional field-effect transistors, the extracted mobility does not take into account the distribution of charge carriers. However, in disordered organic field-effect transistors, the local charge carrier mobility decreases from the semiconductor/insulator interface into the bulk, due to its dependence on the charge carrier density. It is demonstrated that the conventional field-effect mobility is a good approximation for the local mobility of the charge carriers at the interface.

© 2003 Elsevier Science B.V. All rights reserved.
PACS: 72.80.Le; 73.61.Ph; 85.60.Jb
Keywords: Poly (3-hexyl thiophene); Field-effect transistor; Charge carrier mobility

1. Introduction

Conjugated polymers have attracted attention as promising materials for applications in light-emitting diodes (PLEDs) [1,2] and field-effect transistors (FETs) [3–5]. An important point in understanding and developing the devices based on organic semiconductors is the mechanism of charge carrier transport. Most organic films have an amorphous structure and disorder dominates the charge transport. In the past years some models based on multiple trapping and release [6], variable range hopping [7] and grain boundary [8] have been used to explain the transport within organic semiconductors. For polymeric LEDs an important consideration for device modelling is the non-uniform charge distribution in the device. For polymeric field-effect transistors (FETs) charge carriers are induced by a gate electrode across an insulating layer. The standard metal-oxide-semiconductor FET equations for extracting the field-effect mobility take into account the total amount of induced charges \( C_i V_G \), but not their distribution in the channel [9]. In this paper, the extraction of the charge carrier mobility for disordered organic FETs with a non-uniform charge distribution is described.

*Corresponding author. Tel.: +31-50-363-8336.
E-mail address: c.tanase@phys.rug.nl (C. Tanase).
2. Results and discussion

A schematic structure of an organic p-type field-effect transistor is shown in the inset of Fig. 1. By applying a negative voltage at the gate electrode the top of the valence band bends upward closer to the Fermi level. This band bending gives rise to a positive accumulation layer of typically 1–2 nm into the semiconductor next to the interface [10]. Applying a voltage $V_D$ between the source and drain contact gives rise to a current in the channel:

$$I_{SD} = WC_i V_D \mu \frac{V_G}{L},$$  \hspace{1cm} (1)

with $W$ and $L$ the width and length of the channel, $C_i V_G$ the total amount of accumulated charge carriers, $V_D/L$ the electric field in the channel, and $\mu$ the charge carrier mobility. The gradual channel approximation is used, which means that the applied gate voltage is much larger than the source drain voltage. In this approximation variations along the source drain channel can be neglected. Furthermore, it should be noted that the use of the total amount of induced charge ($C_i V_G$) in Eq. (1) is only valid when all charge carriers have the same mobility.

In that case, the field-effect mobility is determined by using the equation [9]

$$\mu = \frac{\partial I_{SD}}{\partial V_G} \bigg|_{V_D=0} \frac{L}{WC_i V_D}. \hspace{1cm} (2)$$

In contrast to conventional monocrystalline silicon in disordered organic semiconductors the transport properties are dominated by localized states [7,10]. Due to an increase of the charge carrier density the lower states of the organic semiconductor are filled and any additional charges in the system will need less activation energy for the jumps to neighboring sites. As a result the mobility will be enhanced and thus depends on the charge carrier density. For the understanding of the transfer characteristics of organic semiconductors it is important to realize that in a FET the charge carrier density is not uniformly distributed in the accumulation channel, but depends on the distance from the interface. Therefore, the mobility which is charge carrier dependent is not uniformly distributed in the accumulation layer. The consequence is that for a given $V_G$ a distribution of charge carrier mobilities is present in the organic FET. In this paper, the distribution of the local mobility in the accumulation channel of disordered FETs and its relevance for the extracted field-effect mobility are described. Within 2 nm from the interface the mobility typically decreases by a factor of 2–4.

As a first step the charge carrier distribution in the active channel of an organic FET is calculated. An unintentionally doped system is considered. In the gradual channel approximation the distribution of charge carriers has to be described only in the direction perpendicular to the semiconductor/insulator interface ($x$). Using Poisson’s equation and the relation between the electric field in the $x$ direction (see inset of Fig. 1) and the potential in the channel for zero source-to-drain voltage [11], the distribution of the electric field in the accumulation layer is given by

$$F_x = \left[ \frac{2}{\varepsilon_0 \varepsilon_r} \right] \left[ \int_0^{V'} \rho(V') \, dV' \right]^{1/2}, \hspace{1cm} (3)$$

where $V'$ is the local potential, which varies from zero far away in the semiconductor bulk to $V$ in the accumulation channel, $\varepsilon_r$ is the relative dielectric constant of the semiconductor, and $\rho(x)$ is
density of charge carriers. We assume that $\varepsilon_r$ at the interface region has the same value as in the bulk. The potential distribution as a function of $x$, follows from the relation

$$x = \int_0^{V_0} \frac{dV'}{F_z(V')},$$

(4)

where $V_0$ is the surface potential of the semiconductor/insulator (S/I) interface. From the variation of the gate-induced potential $V(x)$ as a function of distance $x$ the density of holes $\rho(x)$ can be calculated. The induced charge per unit area $Q_{\text{ind}}$ is related to the gate voltage as follows:

$$V_G = \frac{Q_{\text{ind}}}{C_i} + V_{\text{fb}} = \frac{\varepsilon_0 \varepsilon_r F_z(0)}{C_i} + V_{\text{fb}},$$

(5)

where $C_i$ is the insulator capacitance per unit area, $F_z(0) = F_z(V = V_0)$ is the electrical field at the S/I interface and $V_{\text{fb}}$ is the flat-band voltage. By increasing the gate voltage the surface potential increases resulting in an increase of charge carrier density. Assuming $V_{\text{fb}} = 0$, $\rho(x)$ is calculated for an undoped semiconductor with $C_i = 15.5$ nF/cm$^2$ and $\varepsilon_r = 2.9$. In Fig. 1 the concentration of charge carriers as a function of distance $x$ is shown for gate voltages of $V_G = -19$ V and $V_G = -10$ V at room temperature. It appears that at $V_G = -19$ V the charge carrier density decreases from $3.5 \times 10^{19}$ cm$^{-3}$ at the semiconductor/insulator interface ($x = 0$) to $1.3 \times 10^{18}$ cm$^{-3}$ at a distance of 2 nm from the interface. For $V_G = -10$ V the total induced charge is about half that of $V_G = -19$ V. It should be noted that the calculated charge distribution is not specific for organic semiconductors but generally applicable to field-effect devices of undoped semiconductors in the linear operating regime, since it only depends on $C_i$ and $\varepsilon_r$.

In order to take into account the charge carrier density-dependent mobility in disordered organic FET a model recently developed by Vissenberg and Matters [7] is used. In their model the charge transport is governed by hopping between localized states. For such a system with low doping level the Fermi level is placed in the tail states of the Gaussian distribution and the density of states (DOS) is approximated by an exponential. Using percolation theory they found an expression for the conductivity of the system. The obtained conductivity is used to find an expression for the local charge carriers mobility $\mu_l$ as a function of density of charge carriers:

$$\mu_l = \frac{\sigma(\rho, T)}{\rho} = \frac{\sigma_0}{e} \left( \frac{\pi(T_0/T)^3}{(2\pi)^{3/2} \Gamma(1 - (T/T_0)) \Gamma(1 + (T/T_0))} \right)^{T_0/T},$$

(6)

where $\sigma_0$ is the prefactor for the conductivity, $x^{-1}$ is the effective overlap parameter between localized states, $T_0$ gives the width of the exponential, $B_c$ is the critical number for the onset of percolation and $\Gamma(a) = \int_0^\infty dx \exp(-x)x^{a-1}$. For three-dimensional amorphous systems $B_c \approx 2.8$ [12]. This equation predicts a power law dependence of $\mu_l$ with $\rho$. It should be noted that in disordered semiconductors the field- and temperature dependence of the mobility are consistently described by models based on hopping in a Gaussian DOS [13,14]. These models are developed specifically for low charge carrier densities. For high carrier densities, as in the accumulation channel of a FET, a model describing the dependence of the mobility on charge carrier density in a Gaussian DOS is not available. Now taking into account the spatial dependence of the charge carrier density and mobility, the field-effect current is calculated over the accumulation channel:

$$I_{\text{SD}} = W \frac{V_D}{L} \int_0^{x_{\text{max}}} \sigma(\rho(x), T) dx = W \frac{V_D}{L} \int_0^{x_{\text{max}}} e\mu_l(\rho(x), T)\rho(x) dx,$$

(7)

where $x_{\text{max}}$ represents the thickness of the accumulation layer which is assumed sufficiently large such that $V(x_{\text{max}}) = 0$. Applying Eqs. (3)–(7) to the transfer characteristics of disordered FETs then provide information on the distribution of the charge carrier density and mobility in the active channel of the FET.

In Fig. 2 the transfer characteristic for organic transistors based on poly(2,5-thienylene vinylen) (PTV) and poly(3-hexyl thiophene) (P3HT) at room temperature is shown. On top of a heavily
doped $n^{++}$-Si used as a gate contact an insulating layer of SiO$_2$ used as a dielectric was thermally grown. Gold contacts for the source and the drain contacts were evaporated onto SiO$_2$. The substrate was then treated with hexa-methyldisilizane (HMDS) to make the surface hydrophobic. Finally the polymer has been spin-coated on top. The devices measured have a channel of 10 $\mu$m in length and 2 cm in width for PTV and 2.5 mm in width for P3HT and the film thickness is 200 nm. PTV has been prepared via a precursor route [10] with a molecular weight around 100 000 g/mol and is an homogeneous amorphous semiconductor. The P3HT has been prepared via the first McCullough synthesis, has a molecular weight of 40 000 g/mol and a very low level of impurities. The microstructure of solution processed P3HT consists of nanocrystalline domains embedded into an amorphous matrix [15]. Due to this, the thin film P3HT can also be regarded as a disordered system with localized states originating in structural defects. From temperature dependent measurements on PTV- and P3HT-FETs $\sigma_0 = 1.6 \times 10^6$ S/m, $\alpha^{-1} = 1.6 \times 10^{-10}$ m and $T_0 = 425$ K have been obtained for P3HT and $\sigma_0 = 5.6 \times 10^6$ S/m, $\alpha^{-1} = 1.5 \times 10^{-10}$ m and $T_0 = 382$ K for PTV, respectively [16]. Using Eq. (7) in Fig. 2 the calculated transfer characteristics for the PTV- and P3HT-FETs are shown (solid lines). In Fig. 2 it can be seen that for $V_G = 0$ V the transistors are “off” indicating that there is no significant unintentional doping. The non-linear increase of the channel current is described by the density-dependent mobility. Another possible mechanism as injection-limited currents can be excluded from non-contact potentiometry measurements on P3HT, which also exhibit strong non-linear behavior [17].

Knowing these parameters the mobility can be calculated from Eq. (6). In Fig. 3 the corresponding local mobility is plotted for $V_G = -19$ V at room temperature for PTV and P3HT. It is demonstrated that for PTV the local mobility $\mu_l(x)$ varies from $2.1 \times 10^{-3}$ cm$^2$/V s at the interface to $1 \times 10^{-3}$ cm$^2$/V s at a distance of 2.7 nm from the
interface at room temperature (Fig. 3(a)). For P3HT $\mu_l(x)$ varies from $6.6 \times 10^{-4}$ cm$^2$/Vs at the interface to $1.7 \times 10^{-4}$ cm$^2$/Vs at a distance of 2 nm from the interface for $V_G = -19$ V (Fig. 3(b)). Thus, due to the inhomogeneous charge carrier density in a disordered FET the local mobility demonstrates a strong variation in the active channel.

For the interpretation of the charge transport in disordered FETs it is crucial to understand how such a mobility distribution compares to the extracted conventional field-effect mobility. In Fig. 3 the local (Eq. (6)) and field-effect mobility (Eq. (2)) are compared. It appears that the local mobility of the charge carriers at the semiconductor/insulator interface at $V_G = -19$ V is 15% and 9% larger than the extracted field-effect mobility for PTV (see Fig. 3(a)) and P3HT (see Fig. 3(b)), respectively. The reason for this relatively small difference is that (as shown in Figs. 1 and 3) not only a major part of the charge carriers is located close to the interface, but also that these charge carriers have the highest mobility. As a result the field-effect current is mainly determined by the charge carriers at the interface. Consequently, the error due the approximation used in Eq. (1), namely that all charge carriers have the same mobility, is relatively small.

3. Conclusions

In conclusion, in disordered organic transistors the dependence of the mobility with gate voltage is determined by the charge carrier dependence of the local mobility. Taking into account the distribution of the charge carrier density in the active channel perpendicular to the insulator the local mobility has been calculated as a function of position in the accumulation layer. It is demonstrated that for disordered organic FETs, in spite of the strong variations in the local mobility in the active channel, the conventional field-effect mobility is a relatively good estimate for the local mobility of the charge carriers at the interface.

References