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Letter

Trap origin of field-dependent mobility of the carrier transport in organic layers

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ABSTRACT

In the measurements of space-charge limited current (SCLC) transport in disordered organic semiconductors, it is often observed that carrier mobility depends on bias voltage. Two continuous models have been applied for the description of this dependence. One interpretation assumes the charge carrier mobility dependent on the local electrical field. In the other one, the mobility at the transport state is affected by the trapping–detrapping dynamics of an exponential distribution of localized states (traps) in the band-gap. Analysing the frequency dependent capacitance and conductance (corresponding to measurements of impedance spectroscopy, IS), we demonstrate that the apparent field-dependent mobility found in experiments can be interpreted in terms of the multiple trapping approach.

The development of organic optoelectronic devices (i.e., light-emitting diodes (OLEDs), and organic photovoltaic cells as well as transistors) has provided excellent performances in the past several years [1]. Particularly, white organic light-emitting diodes have recently reached the value of 90 lm W^{-1}, surpassing the benchmark of the fluorescent tube efficiency (60–70 lm W^{-1}) [2]. Moreover, large-area, full color and flat-panel displays may be achieved by the OLED technology [3]. Nevertheless, degradation still remains as a major issue in many organic-based devices. In order to improve stability and further optimization, the understanding of the physical processes occurring within organic semiconductor materials becomes of prime importance. Charge transport in organic semiconductors has been widely studied by using current density–potential (*J*–*V*) curves and time- or frequency-resolved measurements (e.g., time of flight, impedance spectroscopy (IS), etc.) [4]. In low-doped organic semiconductors, space-charge limited current (SCLC) regime usually governs the carrier transport in the bulk layer [5]. However, the well-known Mott–Gourney square law $J \propto V^2$, for trap-free materials with constant mobility μ, was not generally found in experiments whereas a stronger *J*–*V* dependence actually was [6,7].

Basically, two interpretations have been applied in the literature to understand this deviation and, thereby, to model the experimental data [8]. In the first approach, current–voltage characteristics behave as $J \propto V^2 \exp(0.89\gamma\sqrt{V/L})$ under the assumption of field-dependent mobility $\mu \propto \exp(\gamma\sqrt{F})$ that explains the extra-current required along the voltage range [9,10]. Experimental determination of mobility by different techniques such as IS supported this assumption [11]. However, the second interpretation is based on the framework of SCLC with constant mobility and a transport level, under the influence of an exponential distribution of traps in the band-gap that capture and release charge carriers, Fig. 1. Current–voltage characteristics display the law $J \propto V^m$ with $m > 2$ [12]. A similar behavior is explained by means of a carrier density-dependent mobility model that stems from hopping conductivity in an exponential density of states [13–15]. Following Tanase et al. [8], the carrier-density dependence of the mobility has been further developed [16–18] and these authors find out that the current–voltage characteristics of organic devices can be adequately modelled. However, in point of fact many groups modelling transport in organic layer continue to use the field-dependence of the mobility [19–23]. It seems therefore, very important to establish the connection between the two approaches and this is the topic of the present Letter. We show here that the apparent field-dependence of the mobility can be explained in terms of a multiple trapping scheme involving a broad distribution of localized states. This last model lies behind the carrier-dependence of the mobility [24].

In a previous publication, we implemented numerically the multiple-trapping model with only a single-trap [25]. We showed that the shape of capacitance spectra (obtained from IS) is critically modified by trapping properties (i.e., kinetic constants and energetic position). Particularly, fast trapping accounts for a transport

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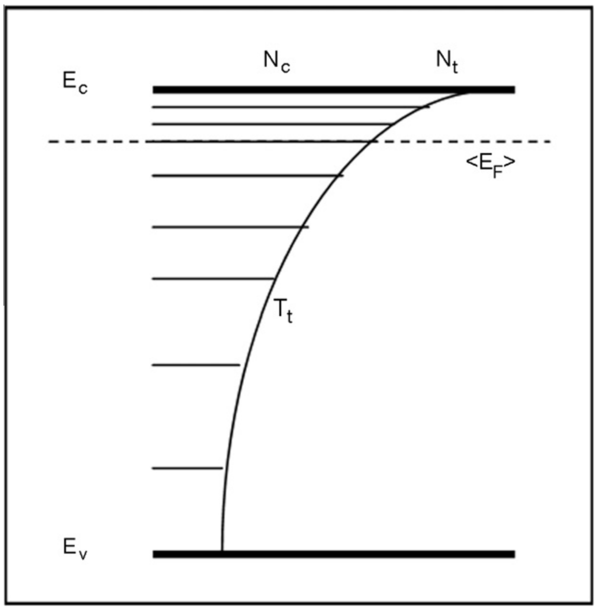

Fig. 1. Schematic energy diagram of the multiple-trapping transport picture. The horizontal scale indicates the density of traps in the band-gap. Representative model quantities are indicated: effective density of transport states N_c, band-gap energy E_c–E_v, effective density of trap states N_t, characteristic trap temperature T_t (determining the shape of the exponential tail) and the average Fermi level $\langle E_F \rangle$.

limitation since carriers may be harshly hindered. The case of a slow-shallow trap provides a low-frequency capacitance increase, whereas for a fast-shallow trap, the step-up of the capacitance exhibits a deviation that directly affects the determination of mobility. In addition, the density of states in organic semiconductors has been conventionally treated as a Gaussian function [26,27], however, an exponential distribution was found to be a good approximation of the effective distribution of the tail states [8]. For low drive voltages, the distribution of occupied trapping states is fairly similar either for a Gaussian or an exponential density, whereas at high voltages, mainly above 10 V, the approximation fails (see Fig. 2). Here we extend the previous analysis, from a single-trap to an exponential density, in order to discuss the measurements of mobility by means of IS at different voltages. The results of our calculation indicate that field-dependent mobility, commonly found in experimental measurements, may be understood in terms of the multiple-trapping picture.

The IS technique is based on the measurement of impedance or equivalently, admittance, $Y(Y(\omega) = G(\omega) + iB(\omega))$, obtained from the application of a small voltage harmonic modulation of angular frequency ω. $G(\omega)$ and $B(\omega)$ (=$\omega C(\omega)$) are the conductance and susceptance, respectively, as a function of the radian frequency. Capacitance spectra can be extracted as $C(\omega) = \mathrm{Im}(Y/\omega)$ [4]. Model representations have been carried out by an extension of the model of [25] to an exponential distribution of traps. Fig. 3a shows capacitance spectra at different steady state voltages applied on a thin film at room temperature. Input parameters concerning the device geometry, charge transport and carrier trapping values are displayed in Table 1. In contrast to the single-trap model, two different behaviors are obtained in the frequency ranges of low and intermediate frequencies. This is due to the fact that the exponential distribution comprises a wide span of localized states according to energetic position and trap dynamics. First, at low frequencies, capacitance undergoes an increase which is more appreciable the less voltage is applied due to the lower occupation of the exponential density of traps. Second, by lowering the Fermi level, more slow-shallow traps within the distribution are emptied causing the low-frequency contribution to capacitance.

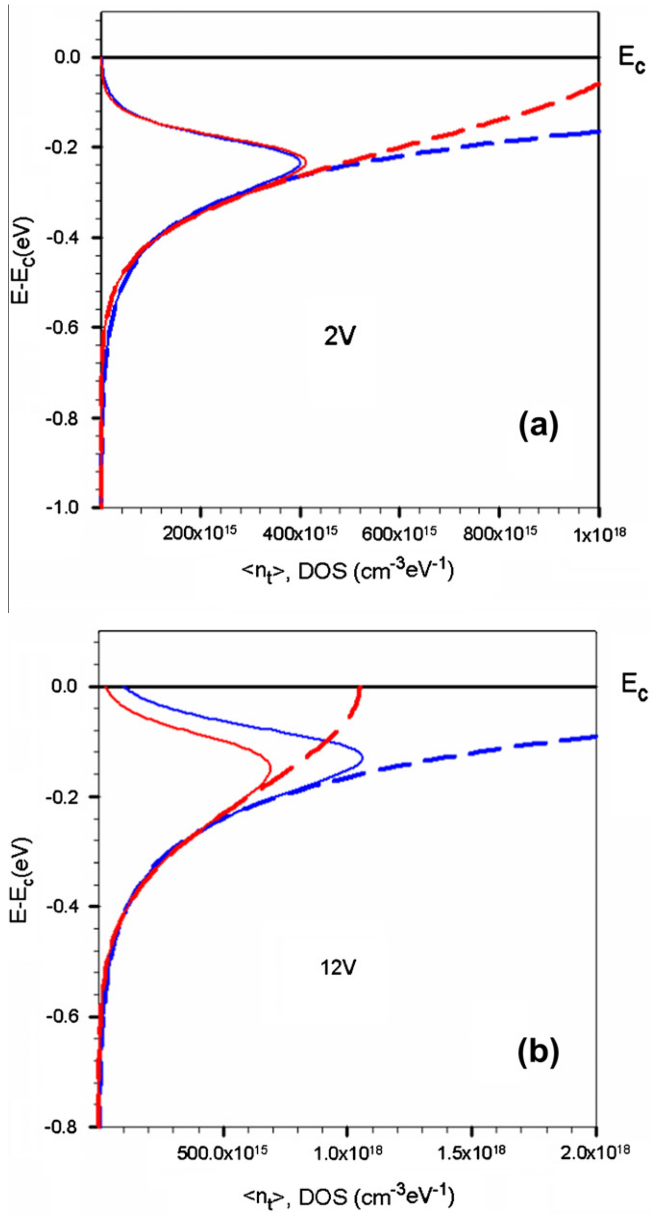

Fig. 2. Comparison of the average of trapped charge occupying the localized states in a Gaussian (red) and exponential (blue) distribution at different voltages: (a) at 2 V and (b) at 10 V. The DOS is shown in dashed line and the occupancy in continuous line. For the Gaussian one, σ = 0.19 eV and for the exponential one T_t = 1250 K in addition to data of Table 1. (For interpretation of the references to colours in this figure legend, the reader is referred to the web version of this paper.)

To determine the mobility by means of the IS technique, we use the representation of the negative differential susceptance $(-\Delta B(\omega) = -\omega(C(\omega) - C_g))$, where C_g is the geometrical capacitance. Fig. 3b displays peaks at intermediate frequencies f_{max} (arrows) that provide the mobility by the expression [28]:

$$\mu = \frac{4}{3}\frac{L^2 f_{\max}}{0.72 \times (V_{bias} - V_{bi})} \tag{1}$$

where $(V_{bias} - V_{bi})$ is the voltage drop in the bulk layer in SCLC.

Calculations of mobility are carried out for different carrier capture coefficients as a function of the square root of the electric field $F^{1/2}$, which is approximated by $(V/L)^{1/2}$, see Fig. 4. Remarkably, the fitting to the field-dependent expression

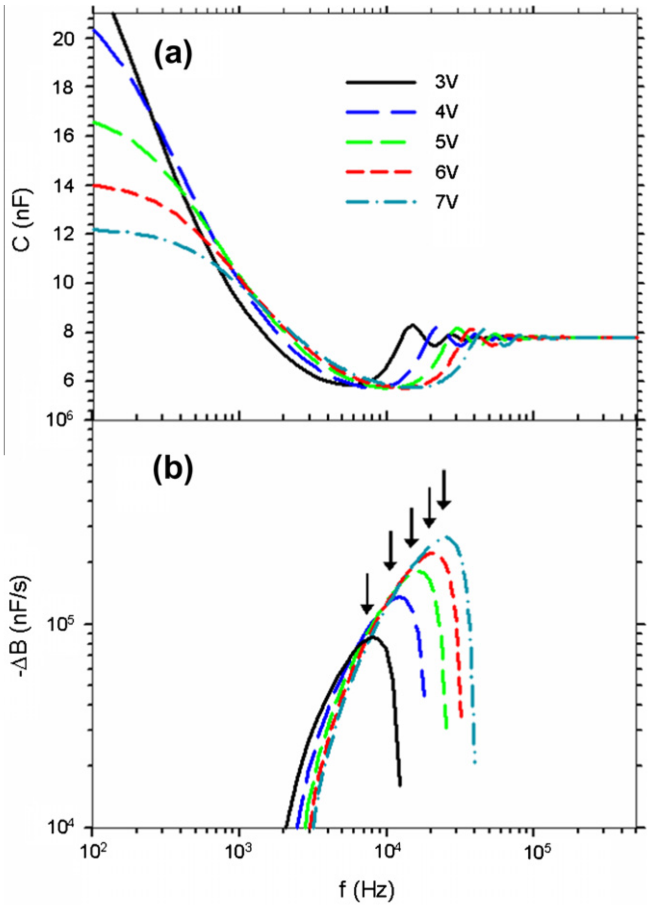

Fig. 3. Model representations at voltages ranging from 3 V to 7 V. (a) capacitance spectra and (b) negative differential susceptance extracted from the upper panel.

Table 1
Parameters used in the numerical simulation.

Parameter	Value
Thickness L	80 nm
Device active area A	0.235 cm^2
Transport effective density of states N_c	10^{19} cm^{-3}
Relative dielectric constant ε_r	3
Trap-free mobility μ_{0n}	5×10^{-7} $cm^2/(Vs)$
Temperature T	300 K
Band-gap E_c–E_v	3 eV
Trap effective density of states N_t	5×10^{17} cm^{-3}
Characteristic trap temperature T_t	1500 K
Trapping capture coefficient c	7×10^{-14} cm^3/s

$$\mu = \mu_0 \exp(\gamma\sqrt{F}) \quad (2)$$

provides common mobility values obtained in organic materials. Mobility parameters μ_0 and γ are the zero-field mobility and the field activation factor, respectively. While the values of μ_0 are expected (the trap-free mobility was appropriately selected for simulation), the most interesting feature corresponds to the exponential factor γ governing the voltage-variation of the mobility. The values $1.1 \times 10^{-3}(\text{cm/V})^{1/2} < \gamma < 2.2 \times 10^{-3}(\text{cm/V})^{1/2}$ derived from the simulation are in the suitable order of magnitude for organic layers as reported in the literature [29,30]. According to the multiple-trapping picture, trap-limitation of mobility stems from the fast-shallow traps within the exponential distribution of localized states. By the application of more voltage, Fermi level covers more trapping states resulting in a less trap-limited transport that enhances the device performance.

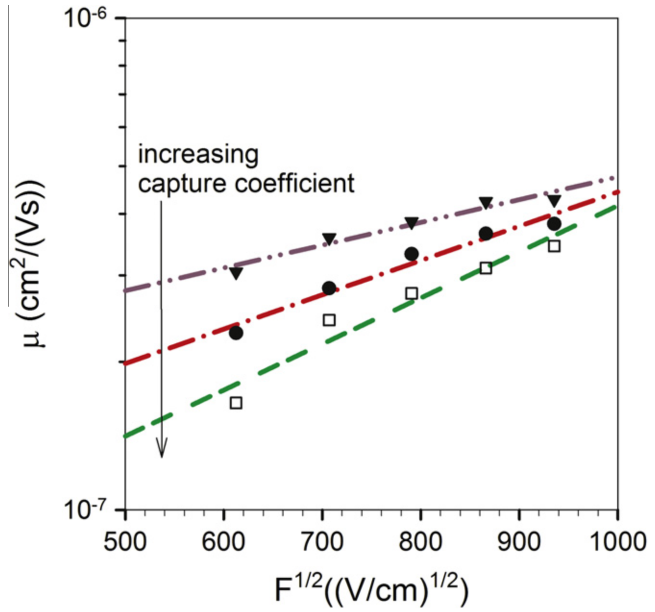

Fig. 4. Model representation of mobility versus $F^{1/2}$ by the IS technique for different capture coefficients $c = 7 \times 10^{-13}$, 4×10^{-12}, 9×10^{-12} cm^3/s, from top to bottom. Mobility fittings are ranged as: 1.6×10^{-7} $cm^2/(Vs) > \mu_0 > 4.8 \times 10^{-8}$ $cm^2/(Vs)$ and 1.1×10^{-3} $(cm/V)^{1/2} < \gamma < 2.2 \times 10^{-3}$ $(cm/V)^{1/2}$.

In summary, we have demonstrated that the apparent mobility dependence on the electric field $\mu \propto \exp(\gamma\sqrt{F})$, usually found by experimental techniques such as IS, may be explained in terms of a multiple-trapping picture. Computational results of our model (SCLC with constant mobility and a transport level under the trapping–detrapping dynamics of an exponential density of traps) yielded a mobility enhancement with the electric field in IS simulations. The main reason is that the trap-limitation of mobility (due to the exponential distribution of localized states) is reduced as more voltage is applied.

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