

**Transition from bulk transport to surface transport in organic field effect transistors**

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We propose a theory of trap-filling transitions in organic thin films in a planar field effect transistor geometry containing an exponential trap distribution. We find that the thickness of the accumulation layer produced by the gate voltage in those devices depends strongly on the degree of energetical disorder in the active layer. As a consequence, the charge-carrier transport in systems with a high degree of energetical disorder can have two regimes: (i) a bulklike regime (BL), where the charge-carrier mobility decreases with the thickness of the semiconductor film and (ii) a surface transport (ST) regime, where the charge-carrier mobility saturates and does not depend on the thickness of the film. We derive approximate analytical expressions for the current-voltage characteristics, the saturation current as a function of the gate voltage (saturation transfer curve), and the field-effect mobility for each regime of charge-carrier transport. We show that the BL/ST transition is characterized by a variation of 2 in the power-law exponent followed by the mobility as a function of the gate voltage after a critical value. By means of our simple model, we discuss the conditions for the observation of the BL/ST transition in thin-film organic field effect transistors. We show that the mobility can have a maximum with increasing deposition of semiconductor material, depending on the nature of the percolative transport in the submonolayer and monolayer scales and on the degree of energetic disorder in the film. Finally we test our model using experimental data measured in  $\alpha$ ,  $\omega$ -dihexylquaterthiophene devices reported in the literature.

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**I. INTRODUCTION**

The application of the simplified theory of space charge limited conduction (SCLC) for bulk charge transport between two Ohmic electrodes is a powerful tool to study charge-carrier transport in insulators. By means of the SCLC theory, the magnitude of the current injected from Ohmic metallic contacts as well as the shape of the current-voltage characteristics ( $I$ - $V$ ) can give valuable information about localized traps in the forbidden gap.<sup>1,2</sup> For instance, in the presence of a single trap level, the  $I$ - $V$  characteristics follows a “modified” Mott-Gurney’s square law with  $I \propto V^2$ .<sup>1</sup> However, when traps are distributed exponentially within the forbidden energy gap, the SCLC theory predicts that the continuous filling of the states at the “tail” of the distribution leads to a power-law variation in the current with increasing voltage given by  $I \propto V^{\gamma+1}$ , where the exponent  $\gamma > 1$  and is related to the characteristic energetic depth of the distribution.<sup>1-3</sup>

Although the simple SCLC theory gives a very good description of bulk conduction between two Ohmic electrodes, the interaction of the injected charge carrier with traps (and its consequences for the device’s electronic properties) is poorly understood in injection-based surface field effect transistors (IFETs).<sup>4-8</sup> Such devices use a film of high-resistivity semiconductor between source and drain metal electrodes.<sup>4</sup> The IFET operation depends then on the injection of charge carriers induced by a voltage applied between the source and a gate electrode. Here we call semiconductor the film of low-conductivity solid in the device’s active layer and insulator the layer that separates the active layer from the gate electrode. Once injected in the semiconductor, the carriers are drifted toward the drain using a voltage applied between the source and drain electrodes. Hence the presence of trap-

ping states in the active layer of an IFETs can immobilize a considerable fraction of carriers injected by the gate voltage ( $V_g$ ), significantly influencing the magnitude and the shape of the current flowing between source and drain. For instance, the interaction of the injected carrier with traps can produce currents depending supralinearly on gate bias<sup>7,8</sup> so that the relation between the shape of those currents with characteristic parameters of the traps remains unclear.<sup>7</sup>

We recently proposed a simple theory of trap-filling effects in IFETs applying a generalization of the surface-charge formalism.<sup>8</sup> The main advantage of this theory is that every balance equation relating volumetric concentration of free and trapped carriers at each point of the channel can be easily written in terms of surface charges. The variation in the carrier populations along the thickness of the semiconductor is considered assuming a characteristic thickness of the accumulation layer produced by  $V_g$ . This thickness is derived from the solution of the trap-free drift-diffusion equation in the direction perpendicular to the semiconductor/insulator (I/S) interface. Using this procedure, the IFET’s characteristic curves can be easily calculated numerically and analytical approximations derived. One can then carry out a straightforward analogy between the simple SCLC theory for bulk conduction and the charge transport in an IFET structure.

In spite of this, the surface-charge theory in Ref. 8 was applied only to describe the IFET’s properties in the simple case of multiple discrete trap levels in the semiconductor. This is a good approximation for single-crystal materials of high structural order and chemical purity.<sup>1</sup> Yet this can be a poor description of IFETs formed by the evaporation of a thin semiconductor layer without a well-defined crystallinity. Due to differences in the environment from one trap site to another, the large amount of structural disorder may result in

a broad smearing out of the discrete energy levels and may also give rise to many different sets of traps.<sup>1,3</sup>

In this contribution we extend the theory proposed in Ref. 8 to the situation when there is an exponential distribution of traps in the semiconductor layer of the IFET. The exponential distribution is one simple and useful representation of many different sets of traps (all smeared out in energy) present in a disordered semiconductor. This is also a needful improvement of the IFET's theory since one possible application of the model is to describe the properties of field effect transistors based on organic wide-band-gap semiconductors (OFETs).<sup>9</sup> Usually the deposition of the organic semiconductor on the surface of the gate insulator create a polycrystalline thin film with a high degree of structural disorder so that the theory developed in Ref. 8 is not appropriate to fully model the carrier transport in those devices.

A deep understanding of the IFET's properties is of great interest because fundamental aspects of the charge-carrier transport in OFETs remain unclear. For example, the dependence of the OFET's performance on the thickness of the organic layer is still a matter of intense debate.<sup>10-17</sup> Although electrostatic models predict that the transport of free carriers occurs in the first few molecular layers (MLs) of the semiconductor adjacent to the gate insulator interface,<sup>8,18</sup> the field-effect mobility ( $\mu$ ) in some OFETs only saturates after the deposition of several MLs. Other devices using different organic semiconductors and different architectures (bottom-contact or top-contact devices) have a maximum  $\mu$  at a certain thickness of the active layer.<sup>11,15,19,20</sup> This behavior has been observed in some devices even after contact resistance correction<sup>11</sup> and the physical origin of this effect is unclear up to now. Using our IFET model, we provide here an explanation to the origin of the maximum  $\mu$  in some OFETs.

In the following we review the IFET formalism to the simple case of multiple discrete trap levels in the semiconductor. We then show how to extend this analysis to a continuous exponential distribution of traps in the semiconductor. From the solution of the drift-diffusion equation in the direction perpendicular to the I/S interface, we show that the thickness of the accumulation layer produced by the gate voltage increases with increasing characteristic energetic depth of the exponential distribution. As a result, the charge-carrier transport along the channel can have two different regimes: a "bulklike" regime, where the carriers transport near the source occurs along the whole thickness of the active layer, and a "surface-transport" regime, where the current flow is mainly confined within a thin layer near the I/S surface. In the last section, we discuss the thickness dependence of mobility in OFETs and the conditions for the observation of the bulklike regime in those devices. We find that the observation of the maximum mobility comes from the interplay between an improved connection among islands of semiconductor material formed at low coverages and a simultaneous reduction in the mobility within those islands due to a higher number of traps induced by increasing deposition of organic material. Finally, we compare the predictions of our model with experimental data measured in devices of  $\alpha$ ,  $\omega$ -dihexylquaterthiophene reported in the literature.

## II. THEORY

### A. Discrete trap levels

Before discussing the charge-carrier transport in an IFET with an exponential distribution of traps, we briefly review the main points of the surface-charge model for discrete trap levels developed in Ref. 8. Here we are going to highlight the main aspects of the theory relevant to the discussion in the next section. Details about the simplifications assumed in the model can be found in Refs. 6 and 8.

The structure of an IFET is formed by a thin film of a wide-band-gap semiconductor (the organic active layer in OFETs) that connects the source and drain contacts, and a high-resistivity gate insulator that separates the semiconductor from the gate electrode. Let us take the directions  $x$  and  $y$  as being, respectively, parallel and perpendicular to the gate insulator interface.

In IFETs with a semiconductor layer free of intrinsic carrier from doping, the charge-carrier conduction between source and drain is achieved by free carriers injected from the electrodes. Without loss of generality, we assume that only one type of charge carrier can be injected through Ohmic contacts at the source and drain and that those charge carriers are electrons. Using the surface-charge notation,<sup>6</sup> the current flow between source and drain is given by

$$\frac{I_{ds}}{W} = |Q_f(x)|\mu \frac{dV}{dx}, \quad (1)$$

where  $\mu$  the charge-carrier mobility,  $W$  the channel width,  $Q_f(x)$  the surface charge of free carriers injected from the electrodes, and  $V(x)$  the potential at position  $x$  from the source.

If  $L$  is the distance between source and drain, the integration of Eq. (1) from  $x=0$  to  $x=L$  gives

$$I_{ds} = \mu \frac{W}{L} \int_0^{V_{ds}} |Q_f(x)| dV, \quad (2)$$

where the limits of integration are  $V(0)=0$  at the source and  $V(L)=V_{ds}$  at the drain.

We assume the presence of multiple trapping states in the semiconductor. For each trap type  $j$ , the relation among the total available trap density  $N_{t,j}$ , the density of occupied traps  $n_{t,i}$ , and the free-carrier density  $n_f$  is given by the balance equation  $\partial n_{t,j} / \partial t = \lambda_j n_f [N_{t,j} - n_{t,j}] - \beta_j n_{t,j}$ , where  $\lambda_j$  ( $\beta_j$ ) is the capture (emission) rate at  $j$ th trap level. Assuming steady state between emission and capture at every coordinate  $x$ , we find

$$n_{t,j}(x) = \frac{N_{t,j}}{[1 + \delta_j/n_f(x)]}, \quad (3)$$

where  $\delta_j = \beta_j/\lambda_j$ .  $\delta_j$  has the units of a density and is characteristic of a particular trap level. It decreases with the thermal activation energy of the trap, i.e., its energetic "depth" in the band gap.

Assuming  $L \gg D$ , where  $D$  is the thickness of the semiconductor, the density of charge carriers injected by the field  $E_x(n_{ds})$  can be neglected compared to the density of charge carriers injected by the field  $E_y(n_g)$ .  $n_g$  is then distributed between the free and the trapped carriers, or

$$n_g(x) = n_f(x) + \sum_j n_{t,j}(x), \quad (4)$$

where the summation over  $j$  in Eq. (4) accounts for the presence in the semiconductor of traps with different trapping properties.

The model in Ref. 8 is based on the assumption that the charge-carrier density along the  $y$  direction is uniform and represented by a characteristic thickness  $\ell$ . Using this approximation, we substitute Eq. (3) in Eq. (4) and rewrite the resulting equation in terms of surface-charge densities, where the surface charges are related to the respective volumetric densities by  $Q_i = en_i \ell$ . Once the variation in  $\ell$  with  $V$  is determined, a polynomial can be solved to find  $Q_f(V)$ . Inserting this  $Q_f(V)$  in Eq. (1) gives then the characteristic curves of the device.<sup>8</sup>

The variation in  $\ell$  with  $V(x)$  and  $V_g$  is approximated using the trap-free equilibrium density of charge carriers resulting from the balance between drift and diffusion currents in the  $y$  direction. The current density along this direction is then

$$j = \mu \rho E_y - \mathcal{D} \frac{d\rho}{dy} = 0, \quad (5)$$

where  $\mathcal{D}$  is the diffusion coefficient which is related to the mobility of the charge carriers by the Einstein's relation  $\mathcal{D} = \mu kT/e$  and  $\rho$  is the charge density of free carriers accumulated at the I/S interface. Neglecting the trapped charge, the Poisson equation in the  $y$  direction is  $dE_y/dy = -(1/\epsilon)\rho$ . Introducing the Poisson equation in Eq. (5) and using the Einstein relation, after some calculations one finds

$$\left(\frac{kT}{e}\right) \left(\frac{d^2 E_y}{dy^2}\right) - E_y \frac{dE_y}{dy} = 0. \quad (6)$$

The solution of Eq. (6) is  $E_y(x) = 2kT/e[y + y_0(x)]$ ,<sup>4</sup> where  $y_0$  represents a characteristic distance from the I/S interface given by  $y_0 = 2ekT/eC_i[V_g - V(x)]$ .<sup>8</sup> The physical meaning of  $y_0$  is that half of the charge induced by the field  $E_y$  is within a distance  $y_0$  from the I/S interface. Hence one can assume that  $\ell \sim 2y_0$ . Since  $y_0$  increases fast when  $V(x) \rightarrow V_g$ , we can also define a voltage  $V'$  so that  $\ell = D$  when  $V(x) \geq V'$ . The variation in the thickness of the accumulation layer with  $V(x)$  is then

$$\ell = \begin{cases} 4ekT/eC_i[V_g - V(x)] & \text{for } V(x) < V' \\ D & \text{for } V(x) \geq V', \end{cases} \quad (7)$$

where  $V' = V_g - 4ekT/eC_i D$ . Following Eq. (7),  $\ell$  is thinner at the source and is given by  $\ell_0 = 4ekT/eC_i V_g$ . This result is useful to derive analytical expressions for  $I_{ds}$  in the simple case of a single trap level in the semiconductor.<sup>8</sup>

### B. Exponential trap distribution

In amorphous semiconductors, the large amount of disorder can create a broad smearing out of the trap's energy

levels. To describe IFETs based on this kind of semiconductor, we consider a distribution of traps with a continuous variation in thermal excitation energy. In this case the quantities in Eq. (3) must be replaced with corresponding quantities belonging to this distribution.<sup>21</sup> For instance,  $N_{t,j}$  is replaced by  $g(\mathcal{E})$ , the density of localized states per unit of energy, and  $\delta_j$  is replaced by  $\delta(\mathcal{E}) = \mathcal{N} \exp(-\mathcal{E}/kT)$ , where  $\mathcal{E} = 0$  corresponds to the energy of the lowest unoccupied conducting state and  $\mathcal{N}$  is the density of the unoccupied conducting states. The total density of trapped electrons  $n_t(x) = \sum_j n_{t,j}$  in Eq. (4) is then calculated by an integration

$$n_t(x) = \int_0 \frac{d\mathcal{E} g(\mathcal{E})}{1 + \exp[-(\mathcal{E} - \mathcal{E}_F)/kT]}, \quad (8)$$

where  $\mathcal{E}_F(x)$  is determined by the density of free electrons or  $\mathcal{E}_F(x) = kT \ln[\mathcal{N}/n_f(x)]$ .<sup>21</sup> A useful approximation for the distribution of trap levels in disordered semiconductors<sup>1,2,22</sup> is to use a trap density that decreases exponentially for decreasing thermal-excitation energies or,  $g(\mathcal{E}) = (N_t/kT_c) \exp(-\mathcal{E}/kT_c)$ , where  $T_c$  is a temperature that characterizes the effective energetic depth of the trap distribution. Introducing this expression in Eq. (8) and performing the integration gives<sup>1</sup>

$$n_t(x) = N_t \left[ \frac{n_f(x)}{\mathcal{N}} \right]^{1/\gamma}, \quad (9)$$

where  $\gamma = T_c/T$ . In writing Eq. (9) we approximate the Fermi-Dirac occupation function in Eq. (8) by a step function of value 0 for  $\mathcal{E} < \mathcal{E}_F$  and 1 for  $\mathcal{E} > \mathcal{E}_F$ , which is a good approximation when  $\gamma > 1$ .<sup>1</sup>

Substituting Eq. (9) in Eq. (4) gives the free charge density as a function of the injected and trapped charges. We have then to rewrite the resulting equation in terms of the surface-charge notation. Following the procedure described in Ref. 8 this is straightforward once the characteristic thickness of the accumulation layer ( $\ell$ ) is determined. Using Eq. (14) and assuming  $n_t(x) \gg n_f(x)$ , Eq. (4) can be written

$$Q_f(x) \approx Q_N \left\{ \frac{C_i[V_g - V(x)]}{Q_T} \right\}^\gamma, \quad (10)$$

where  $Q_N = e\mathcal{N}\ell$ ,  $Q_T = eN_t\ell$ , and we use  $Q_g = en_g\ell = C_i[V_g - V(x)]$ . Equation (10) gives the variation in  $Q_f(x)$  as a function of  $V(x)$ . Performing the numerical integration of Eq. (2) using this function gives the  $I_{ds} \times V_{ds}$  curve. The variation in the saturation current  $I_{sat}$  with  $V_g$  is calculated using the same procedure but taking  $V_{ds} = V_g$  as the upper limit of integration in Eq. (2).

We proceed now to calculate  $\ell$  and its dependence on  $V_g$  and  $V(x)$ . We assume again that the current flowing along the  $y$  direction is zero so that Eq. (5) is valid. Considering again  $n_t(x) \gg n_f(x)$  and using Eq. (9), the Poisson equation in the  $y$  direction is

$$\frac{dE_y}{dy} = -\frac{e}{\epsilon} n_t(x) = -a\rho^{1/\gamma}, \quad (11)$$

where  $a = (1/\epsilon)(N_t/\mathcal{N}^{1/\gamma})$ . Substituting  $\rho$  from Eq. (11) into Eq. (5) and using the Einstein's relation, we find

$$\frac{kT}{e} \frac{d}{dy} \left[ \left( \frac{dE_y}{dy} \right)^\gamma \right] - \left( \frac{dE_y}{dy} \right)^\gamma E_y = 0. \quad (12)$$

The derivation of the first term on the right-hand side of Eq. (12) gives

$$\left( \frac{kT_c}{e} \right) \frac{d^2 E_y}{dy^2} - E_y \frac{dE_y}{dy} = 0. \quad (13)$$

Comparing Eq. (13) with Eq. (6), one can see that the two equations are identical but the temperature  $T$  in Eq. (6) is replaced by the characteristic temperature  $T_c$  in Eq. (13). Therefore, the solution of Eq. (13) is easily obtained from the well-known solution of the trap free Eq. (6) by simply substituting  $T$  for  $T_c$ . Hence, following Eq. (7), the variation in  $\ell$  with  $V(x)$  in the presence of an exponential trap distribution can be approximate by

$$\ell = \begin{cases} 4\epsilon kT_c / eC_i [V_g - V(x)] & \text{for } V(x) < V' \\ D & \text{for } V(x) \geq V', \end{cases} \quad (14)$$

where  $V'$  is now  $V' = V_g - 4\epsilon kT_c / eC_i D$ . Since  $T_c > T$ , the  $\ell$  at a fixed  $V_g$  obtained from Eq. (14) is always thicker compared to the value obtained from Eq. (7). Hence, there is a considerable range of gate voltages where the thickness of the accumulation layer is approximately the thickness of the active layer  $D$ . This range increases with increasing energetic disorder represented by the parameter  $T_c$ . Thus, if the gate voltage is lower than a characteristic voltage given by  $V_{tr} = 4\epsilon kT_c / eC_i D$ , the accumulation layer spreads along the whole thickness of the semiconductor and the charge transport is similar to the bulk conduction between two Ohmic electrodes described by the SCLC theory. We call this transport regime “bulklike” (BL) regime. On the other hand, in the range  $V_g > V_{tr}$ ,  $\ell < D$  in the vicinities of the source and the flow of carrier is constrained in the  $y$  direction by the field  $E_y$ . In this region the transport occurs mainly near the  $I/S$  interface. We call this regime a “surface transport” (ST) regime. Compared to the BL transport, the variation in the accumulation layer along the channel introduces further dependences of the current on  $V_g$  in the ST regime. Finally, the thickness of the accumulation layer at the source in the ST regime is  $\ell_c = 4\epsilon kT_c / eC_i V_g = \gamma \ell_0$ .

### C. Analytical approximation for traps exponentially distributed in energy

Approximate analytical expressions for  $I_{ds}$  in the case of an exponential distribution of traps can be derived from Eq. (10) by considering a constant thickness of the accumulation layer given by the value of  $\ell$  at the source. The surface-charge densities in Eq. (10) are then given by multiplying the respective volumetric densities to the factor  $eD$  if  $V_g < V_{tr}$  or to the factor  $e\ell_c$  if  $V_g > V_{tr}$ . Inserting Eq. (10) in Eq. (2) and performing the integration yields

$$I_{ds} = \begin{cases} \frac{W}{L} \mu \frac{\mathcal{N}}{N_t} f(V_{ds}, V_g) \left[ \frac{C_i V_g}{eDN_t} \right]^{\gamma-1} & \text{for } V_g < V_{tr} \\ \frac{W}{L} \mu \frac{\mathcal{N}}{N_t} f(V_{ds}, V_g) \left[ \frac{(C_i V_g)^2}{(4\epsilon kT_c N_t)} \right]^{\gamma-1} & \text{for } V_g > V_{tr}, \end{cases} \quad (15)$$

where  $f(V_{ds}, V_g) \equiv C_i V_g^2 (\gamma+1)^{-1} (1 - [1 - (V_{ds}/V_g)]^{\gamma+1})$ .

In the limit  $V_{ds} \ll V_g$ , Eq. (15) yields a linear dependence of the source-drain current on  $V_{ds}$  or

$$I_{ds} = \begin{cases} \frac{W}{L} \mu \frac{\mathcal{N}}{N_t} C_i V_g \left[ \frac{C_i V_g}{eDN_t} \right]^{\gamma-1} V_{ds} & \text{for } V_g < V_{tr} \\ \frac{W}{L} \mu \frac{\mathcal{N}}{N_t} C_i V_g \left[ \frac{(C_i V_g)^2}{(4\epsilon kT_c N_t)} \right]^{\gamma-1} V_{ds} & \text{for } V_g > V_{tr}, \end{cases} \quad (16)$$

where we apply the relation  $(1+x)^\gamma \approx 1 + \gamma x$  for  $x \ll 1$ .

Finally, the saturation current is obtained doing  $V_{ds} = V_g$  in Eq. (15)

$$I_{sat} = \begin{cases} \frac{W}{L} \frac{\mu}{(\gamma+1)} \frac{\mathcal{N}}{N_t} C_i V_g^2 \left[ \frac{C_i V_g}{eDN_t} \right]^{\gamma-1} & \text{for } V_g < V_{tr} \\ \frac{W}{L} \frac{\mu}{(\gamma+1)} \frac{\mathcal{N}}{N_t} C_i V_g^2 \left[ \frac{(C_i V_g)^2}{(4\epsilon kT_c N_t)} \right]^{\gamma-1} & \text{for } V_g > V_{tr}. \end{cases} \quad (17)$$

A valuable quantity to characterize charge transport is the charge-carrier mobility. In the linear region of the  $I$ - $V$  characteristics, the effective-field-effect mobility can be obtained from the IFET's transconductance

$$\mu_{eff} = \frac{L}{C_i W V_{ds}} \frac{\partial I_{ds}}{\partial V_g}. \quad (18)$$

Using Eqs. (16) and (18), the field-effect mobility in the BL regime ( $V_g < V_{tr}$ ) is

$$\mu_{eff} = \gamma \mu \frac{\mathcal{N}}{N_t} \left[ \frac{C_i V_g}{eDN_t} \right]^{\gamma-1}. \quad (19)$$

Similarly, using again Eqs. (16) and (18), the field-effect mobility in the ST regime ( $V_g > V_{tr}$ ) is

$$\mu_{eff} = (2\gamma - 1) \mu \frac{\mathcal{N}}{N_t} \left[ \frac{(C_i V_g)^2}{4\epsilon kT_c N_t} \right]^{\gamma-1}. \quad (20)$$

From Eqs. (19) and (20) one sees that it is expected a factor-of-2 variation in the exponent of the power law followed by  $\mu_{eff}$  with  $V_g$  when the gate voltage rises above  $V_{tr}$  (BL/ST transition). It is also important to point out that Eq. (19) is very similar to the expression for the field-effect mobility in OFETs derived using the multiple trapping and release (MTR) model.<sup>18</sup> In both pictures (our model and the MTR model), the increase in the field-effect mobility with increasing gate voltage is due to the filling of the low-lying trapping states of the exponential distribution. On the other hand, the  $\mu_{eff}$  in Eq. (20) has the same dependence on the gate voltage obtained in Ref. 23 using a variable-range hopping (VRH) transport between energy levels from an expo-

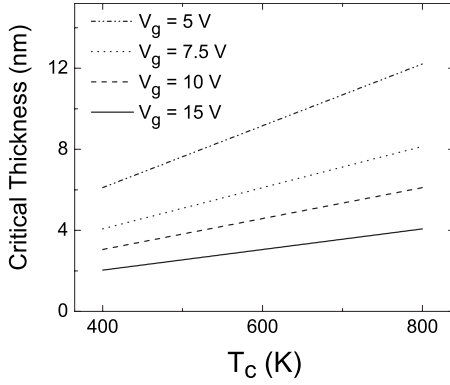


FIG. 1. Variation in the critical thickness ( $\ell_c$ ) as a function of  $T_c$  for different gate voltages. The simulation parameters are  $C_i=1.2 \times 10^{-4}$  F/m<sup>2</sup>,  $\epsilon=3\epsilon_0$ ,  $L=25$   $\mu$ m,  $W=1.5 \times 10^{-3}$  m,  $\mu=10^{-4}$  m<sup>2</sup>/V.s,  $D=20$  nm,  $N_c=10^{27}$  m<sup>-3</sup>, and  $N_i=10^{26}$  m<sup>-3</sup>.

nential density of localized states (DOS). The fact that our model reproduces the same  $\mu_{eff}$  dependence on  $V_g$  derived from the VRH theory<sup>23</sup> is a consequence of the great population of available states for hopping conduction at high energies in an exponential DOS. Those levels can have the role of a transport level whereas the low population of available states for hopping at low energies can have the role of traps. In this sense the model for the charge transport proposed here is equivalent to the hopping model in an exponential DOS since the latter can be effectively described in terms of thermal activation from the Fermi energy to a specific transport energy.<sup>24</sup> Moreover, from the definition of  $\mathcal{E}_F$  above and using Eq. (10), one can write  $\mathcal{E}_F=kT_c \ln[eN_i D / (C_i V_g)]$  (BL regime) or  $\mathcal{E}_F=kT_c \ln[4ekT_c N_i / (C_i V_g)^2]$  (ST regime). Introducing those equations in Eqs. (19) and (20), respectively, yields  $\mu_{eff} \propto \exp[-(\mathcal{E}_F/k)(T^{-1}-T_c^{-1})]$ . Therefore, since  $\mathcal{E}=0$  corresponds to the energy of the lowest conducting state, one can see that field-effect mobilities in Eqs. (19) and (20) follow a simple Arrhenius behavior on temperature and also obey the Meyer-Neldel rule (MNR) for both regimes of charge-carrier conduction. The simple Arrhenius behavior of the field-effect mobility and the MNR for  $\mu_{eff}$  has been observed in OFETs using a variety of organic semiconductor.<sup>7,23,25</sup> However, the activation energies and their dependences on  $V_g$  for the BL and ST regimes are different: at a constant gate voltage in the interval  $V_g > V_{tr}$ , the activation energy for the BL conduction is a factor  $D/\ell_c$  higher than the activation energy in ST conduction.

Another important point is the dependence of the field-effect mobilities in Eqs. (19) and (20) with the thickness of the active layer  $D$ . At constant  $V_g$ ,  $\mu_{eff}$  decreases with increasing film thickness (BL transport) following a power law described in Eq. (19) till a critical thickness ( $\ell_c$ ) is reached. For thicker films in the range  $D > \ell_c$ ,  $\mu_{eff}$  does not depend on  $D$  (ST regime) and the mobility saturates in values given by Eq. (20) for measurements in the linear region. Figure 1 shows the variation in the critical thickness ( $\ell_c$ ) as a function of the characteristic temperature  $T_c$ . The parameters used are characteristic of OFETs. One can see that the energetic disorder strongly influences the transition between the BL and the ST conduction regimes. At high  $T_c$  and low  $V_g$  the field-

effect mobility may not depend on  $D$  only for films thicker than  $\sim 10$  nm.

### III. DISCUSSION: THICKNESS-DEPENDENT MOBILITY IN OFETS

The main purpose of the IFET model is to provide a simple (yet realist) description of the charge-carrier transport in OFETs. In the equations above,  $\mu_{eff}$  decreases with increasing film thickness and then saturates after a critical value  $\ell_c$  is reached. However, the field-effect mobility measured in different OFETs shows a step increase in  $\mu_{eff}$  with increasing  $D$  in the submonolayer and single-monolayer regimes. This behavior is attributed to a percolation transition<sup>10,16</sup> where island of the semiconductor material come into contact with increasing coverage of the gate insulator area. At coverages just above the percolation threshold, the number of semiconductor's islands clusters increases and eventually a two-dimensional (2D) semiconductor layer is formed.<sup>16</sup> The improved connections between the semiconductor islands explain the increase in the mobility with  $D$ . When a second molecular layer is beginning to grow on top of the first layer, the field-effect mobility can still increase since islands of the second layer can provide additional conducting paths bridging isolated islands of the first layer.<sup>10,14,16</sup> We will call those transport regimes at low coverages as percolativelike (PL) transport regimes.

The description of the mobility in the PL transport regimes is challenging since the formation of three-dimensional (3D) islands at low coverages complicates the geometric description of the percolation transition.<sup>16</sup> One recent example of complex behavior derived from charge-carrier transport in the PL regime is observed in the field-effect mobility of transistors made from self-assembled monolayers of liquid-crystal molecules.<sup>26</sup> Due to charge percolation in two dimensions,  $\mu$  in those devices depends on channel length only when the monolayer coverage is incomplete.<sup>26</sup> Moreover, the growth of the second layer on the top of the first layer can induce traps in the semiconductor film which adds further complexity to the problem.<sup>15</sup> Hence, we are not going to discuss PL transport regimes here. This would require a microscopic theory linking aspect of film morphology to transport properties that goes far beyond the simple continuous electrostatic formalism developed in the last section. Nevertheless, some insights on the mobility variation (and charge-carrier-transport transitions) with increasing thickness can be obtained from our model.

At low coverages, let us assume that the mobility scales such as the conductivity versus coverage in a percolative problem<sup>10</sup> and that  $D$  is proportional to the coverage.  $\mu_{eff}$  follows then a power law of the kind  $\mu_{eff} \propto (D-D_c)^\alpha$  (Ref. 27) in the range  $D \geq D_c$ , where  $D_c$  is average thickness of the film at the percolation threshold. The critical exponent is  $\alpha=1-1.4$  for percolation in 2D and  $\alpha=1.5-2$  for percolation in 3D.<sup>10</sup> By the other hand, following the equations for the BL regime derived above, the mobility in the islands decreases with increasing film thickness following also a power law of the kind  $\mu_{eff} \propto D^{-(\gamma-1)}$ . Hence, the mobility rise due to improved island contacts with increasing  $D$  can be compen-

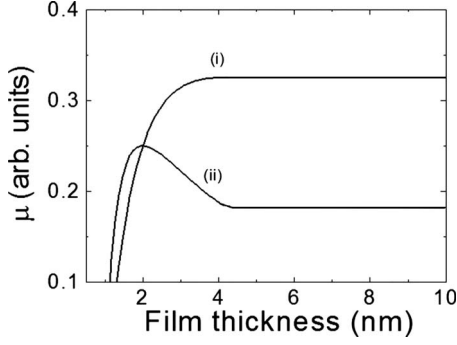


FIG. 2. Schematic behavior of the field-effect mobility as a function of the film thickness after the percolation threshold when  $\gamma > \alpha + 1$ . The nonconstant ranges of the curves are calculated from  $\mu \propto (D - D_c)^\alpha D^{-(\gamma-1)}$  with  $D_c = 1$  nm,  $\gamma = 3$ ,  $\alpha = 1.5$  [curve (i)], and  $\alpha = 1$  [curve (ii)]. We assume  $\ell_c = 4$  nm.

sated by poor conduction within the islands produced by a higher number of traps. The interplay between those two effects can give rise to a maximum effective mobility at a thickness

$$D_0 \sim \frac{\gamma - 1}{(\gamma - 1) - \alpha} D_c \quad (21)$$

if  $\gamma > \alpha + 1$ . The relation between the exponents  $\alpha$  and  $\gamma$  determines then different transitions among the PL, BL, and ST regimes in an IFET as a function of the semiconductor thickness. For values of  $\alpha$  and  $\gamma$  satisfying  $\gamma > \alpha + 1$ , the mobility can have two different behaviors that are schematically illustrated in Fig. 2: (i) if  $D_0 \geq \ell_c$ ,  $\mu_{eff}$  raises and then saturates at  $D = \ell_c$  with increasing  $D$  [curve (i)]. Hence there is a direct transition from the PL transport regime to the ST regime or (ii) if  $D_0 < \ell_c$ , the mobility rises till a maximum value is reached and then starts to decrease at  $D_0$  (transition from the PL to the BL transport regimes). The mobility keeps decreasing in the range  $D_0 < D < \ell_c$  and then saturates at  $D \sim \ell_c = \gamma \ell_0$  (transition from the BL regime to the ST regime) [curve (ii)]. Finally, for values of  $\gamma$  and  $\alpha$  in the range  $\gamma \leq \alpha + 1$ , the BL transport regime is never present and  $\mu_{eff}$  rises till the critical thickness  $D \sim \ell_c = \gamma \ell_0$ . Thereafter the mobility becomes independent on  $D$  (ST regime).

It is clear from the discussion above that the mobility variation with the film thickness can have different profiles after the percolation threshold depending on the nature of the percolative transport in the PL regime (scaling in 2D or 3D) and on the degree of energetic disorder within the semiconductor. This can explain the variety of results measured in devices using different combinations of organic semiconductors films and gate insulator surfaces.<sup>10,11,14,15,17</sup> Moreover, if the PL regime scales such as the percolation in 2D,<sup>10</sup> the necessary (but not sufficient) condition for the observation of BL/ST transition is  $\gamma > 2$ . Again this means that the BL/ST transition may be observed only in semiconductor films with a high degree of energetical disorder or when the variation in the mobility on  $D$  is measured at temperatures lower than  $T < T_c/2$  (assuming that  $\alpha$  does not depend on  $T$ ). Since the range of values of  $T_c$  is usually around 400 K,<sup>22,23</sup> the BL/ST transition in the  $\mu_{eff} \times D$  curve would be observed when  $T$

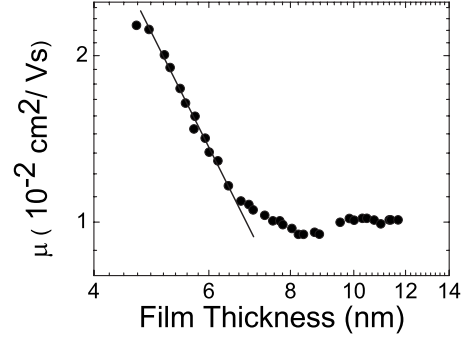


FIG. 3. Variation in the field-effect mobility with the thickness of the active layer measured in DH4T devices in the range  $D > 4.5$  nm (Ref. 15) (circles). The straight line is a power-law fit of the experimental data with exponent  $-2.36$ .

$\leq 200$  K, provided that the condition  $D_0 < \ell_c$  is simultaneously satisfied. From Eq. (21) and the definition of  $\ell_c$ , this happens for every  $T$  if  $\ell_c > D_c$  or, equivalently,  $V_g < 4ekT_c/eC_iD_c$ . Hence, measurements of  $\mu_{eff}$  as a function of temperature in the range  $V_g < 4ekT_c/eC_iD_c$  may show a variation by a factor 2 in the power law followed by the mobility as a function of  $V_g$  [see Eqs. (19) and (20)] when the temperature rises above  $T \sim 200$  K.

A strong experimental evidence of the BL/ST transition can be found in OFETs with  $\alpha$ ,  $\omega$ -dihexylquaterthiophene (DH4T) as active layer.<sup>15</sup> In Ref. 15,  $\mu_{eff}$  was obtained in the linear regime as a function of the thickness of the active layer. In the submonolayer region of the DH4T OFET, a step increase in the mobility is observed after  $D \sim 1.5$  nm, following approximately a quadratic power law. This quadratically increasing mobility continues until  $D = 2.15$  nm (0.77 ML). With further increasing coverage, a first peak of mobility is reached at  $D = 2.5$  nm (0.9 ML). After the beginning of the second ML, a reduction in  $\mu_{eff}$  is observed due to disturbing effects on the morphology of the first ML created by the growth of the second monolayer.<sup>15</sup> Thereafter, a second lower peak on  $\mu_{eff}$  is observed at  $D \sim 4.5$  nm, corresponding to the completion of the second monolayer. For higher coverages the mobility decreases and saturates for  $D$  beyond 7 nm. Figure 3 shows the variation in  $\mu_{eff}$  measured at  $|V_{ds}| = 0.2$  V in the DH4T devices of Ref. 15 for  $D > 4.5$  nm. One can see that the field-effect mobility decreases with increasing thickness following a power law with exponent  $-2.36$  until  $D \sim 6.5$  nm. This behavior corresponds to  $\gamma = 3.36$  in Eq. (19) and  $|V_{tr}| \sim 5$  V for a  $C_i$  corresponding to a 100 nm silicon oxide layer as gate insulator.<sup>15</sup> Since  $\mu_{eff}$  observes a quadratically power-law rise in the submonolayer region,<sup>15</sup> the geometry-corrected percolation exponent is  $\alpha = 1$ ,<sup>10</sup> consistent with a percolation scaling in 2D. Thus the values of  $\alpha$  and  $\gamma$  above satisfy the necessary condition for the observation of a maximum  $\mu$ ,  $\gamma > \alpha + 1$ . Moreover, using again those values of  $\alpha$  and  $\gamma$  in Eq. (21) and taking  $D_c = 1.5$  nm, one gets  $D_0 = 2.6$  nm for the thickness of the maximum mobility. This result is in excellent agreement with the experimental maximum at 2.5 nm.

#### IV. CONCLUSION

We develop a model for charge-carrier transport in IFETs with an exponential distribution of traps in the active layer.

The major implication of the model is that the energetical disorder greatly influences the thickness of the accumulation layer created by the gate voltage in the vicinities of the gate insulator interface. This accumulation layer gets thicker with increasing  $T_c$ , the parameter that determines the energetic “depth” of the exponential distribution. As a consequence, the charge-carrier transport in IFETs can have two regimes: (i) when the accumulation layer spreads along the whole  $D$  near the source, a bulklike transport between source and drain is established. The BL transport in IFETs is similar to the bulk transport in an insulator with an exponential distribution of traps and sandwiched between two Ohmic contacts. In this regime the mobility decreases with increasing film thickness due to the higher number of traps within the active layer and (ii) when the thickness of the accumulation layer is thinner compared to the thickness of the semiconductor, the charge-carrier conduction between source and drain mainly occurs in a thin layer near the I/S interface. This is the ST regime, characterized by a field-effect mobility that does not depend on  $D$ . We show that the BL/ST transition produces a variation of 2 in the power-law exponent followed by the mobility as a function of  $V_g$  after a critical gate voltage ( $V_{tr}$ ).

Assuming that the mobility scales such as the conductivity in a percolation problem for submonolayer and monolayer coverages of the gate insulator area, we apply our theory to discuss the thickness-dependent mobility in OFETs. We derive the conditions for the observation of the BL and ST conduction regimes in those devices. We find that the necessary conditions for the observation of a maximum mobility in OFETs are related to the exponents  $\alpha$  and  $\gamma$ , where  $\gamma$  is the parameter that quantifies the energetic disorder in the semiconductor and  $\alpha$  the critical exponent that characterizes the dimension of the percolative transport at low coverages. Finally, we test our model using experimental data measured in DH4T OFETs. We find good agreement between the thickness of the maximum mobility measured in those devices and the thickness of the maximum  $\mu$  calculated from our analysis. The observation of the maximum mobility and BL/ST transition in those devices is a consequence of a percolation transport scaling in 2D in the submonolayer region together with a high degree of energetical disorder of the DH4T films in the OFETs under consideration.<sup>15</sup>

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