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Line shapes in the current–voltage characteristics of single layer metal/organic semiconductor structures as response to the electric field at the charge injecting interface

B. Cvikl *

Faculty of Energy Technology and Faculty of Civil Engineering, University of Maribor, 2000 Maribor, Slovenia
“Jožef Stefan” Institute, 1000 Ljubljana, Slovenia

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ABSTRACT

By incorporating a non-zero electric field at the charge injecting metal/organic interface, E_{int}, into the Mark–Helfrich model of charge traps exponentially distributed in energy, a general expression for the current density–voltage dependence of single layer metal/organic structures is presented. It is expressed in terms of the solutions of the non-linear algebraic equation relating E_{int} to the externally applied electric field, E_a. As a result, the finite and continuous spatial distribution of the free and trap charge density at the boundaries and within the organic layer is obtained. Arguments are presented that for an interfacial field directly proportional to the applied electric field, $E_{int} = \lambda E_a$, with λ bounded between $0.1 \le \lambda \le 0.99$, the λ dependent current density follows an identical bias and organic thickness prediction as the original Mark–Helfrich model, but with a substantially different factor of proportionality. The resulting current density is characterized by a negligible space charge limited current (SCLC) effect. The bias independent E_{int} at the charge-injecting interface leads to a concave line shape (when viewed through the current axis) in the current–voltage diagram. The intense (but always) finite peak of the free charge density at this interface occurs only for $E_{int} \ll E_a$, thus leading to the strong SCLC effect. These predictions are tested on some published experimental data and good agreement is observed. The combination of an interfacial electric field linearly dependent upon the applied bias up to a given value and constant thereafter results in at least two distinct line slopes in the $\log j - \log E_a$ plane.

1. Introduction

Investigations of organic thin films are currently particularly intensive due to their significant potential for application in the area of electronic devices. In this respect the central issue which is presently attracting interest is a detailed understanding of charge transport within organic thin film structures as a precondition for optimization of the electrical characteristics of such devices. In numerous studies it has been pointed out that the transport of charge carriers within organic films is greatly influenced (i.e. reduced) by the trapping of free charges. These traps are spatially distributed within the organic medium and are attributed to doping, and chemical and structural effects occurring at the metal/organic interfaces, as well as within the bulk. The experimental method that is suitable for the analysis of charge transport in such organic structures is current–voltage (j–V) measurement. The strong power law dependence of the electrical current as a function of the applied voltage that such data often display is attributed to the effect of charge traps and is rather commonly (but not exclusively) interpreted in terms of the Mark–Helfrich (MH) model of charge traps exponentially distributed in energy within the transport gap [1]. Likewise, as shown recently [2], the strong hysteresis effects occasionally exhibited in the j–V measurements of various metal/organic devices are particular features assigned to the existence of deep traps.

The MH model, although widely used nowadays for the interpretation of current–voltage measurements of organic semiconductor structures [2–10], inorganic semiconductor structures [11,12], and organic nanostructures [13–15], is incomplete [16]. Namely, since the electric field at the charge-injecting interface, E_{int}, is commonly assumed to be zero [1,3] the free charge density, n_{free}, and the density of charge traps, n, diverge at this position. However, an elementary proof that the stated boundary condition $E_{int} = 0$ is an improper one has been given recently. The arguments were tested [16] on published room temperature electron-only j–V data of a metal/organic single layer structure characterized by a broad interval of external bias [9].

Considering a single layer organic semiconductor structure and unipolar charge carriers, it is the purpose of this work to present an

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* Faculty of Energy Technology and Faculty of Civil Engineering, University of Maribor, 2000 Maribor, Slovenia. Tel.: +386 22294362.
E-mail address: bruno.cvikl@ijs.si.

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expression for the bias dependent current density such that the singularities of all physical quantities are removed.

It was already shown previously [16] that the values of the material parameters of an organic semiconductor are constrained by a rather simple nonlinear algebraic equation that depends on both the electric field at the interface, E_{int}, as well as on the externally applied electric field, E_a. Since E_{int} is non-zero the internal electric field, E(x), is always finite and non-zero, resulting in a finite spatial distribution of the free (and also trap) charge density throughout the organic, including the interfaces.

The first case investigated assumes E_{int} to be directly proportional to E_a, i.e. $E_{int} = \lambda E_a$, where $0.1 \le \lambda \le 0.99$. It is determined that the spatial distribution of the (λ dependent) internal electric field, E(x), and the related free charge density, $n_{free}(x)$, is nearly constant throughout the organic and at both boundaries. Consequently, a negligible trap charge limited current effect is found to appear under this linear dependence of $E_{int,}$ irrespective of the magnitude of λ. This finding evidently differs from the original MH prediction, but the bias and thickness dependence of the current density in single layer organics remain unchanged. An expression for the current density, which depends upon λ is derived and proposed as a substitute for the original MH expression.

On the other hand, the conjecture of constant electric field at the charge-injecting interface, E_{int}, leads to a current density that extends beyond the original MH predictions. It is found that the resulting current–voltage line shape turns out to be concave when viewed through the current axis. It is shown that in the limit when $E_{int} \ll E_a^{max}$, the induced free charge density at the charge injecting interface may become very large, yet at some 10 nm within the organic it already rapidly decays to its bulk value.

The above findings are tested on a number of published data. The examples presented emphasize the fact that the electric field at the charge injecting interface influences the bias dependence and the line shape of the measured current density, j. Thus it is explicitly shown that the combination of an interfacial electric field linearly dependent upon the applied bias up to a given value of the external electric field and constant thereafter, results in a continuous curve (proportional to the current density) which exhibits at least two distinct line slopes in the $\log j - \log E_a$ plane.

2. The extended MH model

It was shown recently [16] that the general solution of the extended MH model of traps exponentially distributed in energy within the organic transport gap may be expressed in terms of the quantity u. At a given applied electric field, E_a, the quantity u depends on the bias dependent non-zero electric field at the charge-injecting interface, E_{int}, according to the expression,

$$\mathrm{u} = \left(\frac{m}{2m+1}\right)\frac{1}{E_\mathrm{a}}\left\{\left[\frac{m+1}{m}\ u\ +\ E_\mathrm{int}^{\frac{m+1}{m}}\right]^{\frac{2m+1}{m+1}}\ -\ E_\mathrm{int}^{\frac{2m+1}{m}}\right\}\ . \tag{1}$$

The quantity u is defined as

$$\mathrm{u} = \frac{qN_tL}{\varepsilon\varepsilon_0}\ \left[\frac{j(E_\mathrm{a})_\mathrm{exp}}{\mu qN_C}\right]^{\frac{1}{m}}. \tag{2}$$

In the expressions above $j(E_a)_{exp}$ is the electron current density at the applied electric field E_a. It is defined as $E_a = V_a/L$, where V_a is the applied bias and L is the organic thickness. N_t denotes the total concentration of traps exponentially distributed within the organic band gap, and N_C is the effective density of states in the charge carrier transport band [1]. The charge carrier mobility is μ, and q is the magnitude of the elementary charge. The relative permittivity of the organic is ε, and the exponent $m = T/T_C > 1$, where T_C is the characteristic temperature of the trap distribution function [1] at the temperature T.

The spatial distribution of the internal electric field within the organic, E(x), measured from the origin set at the charge injecting interface at the position x = 0 is then [16],

$$\mathrm{E(x)} = \left[\frac{m+1}{m}\ \left(\frac{x}{L}\right)u + E_\mathrm{int}^{\frac{m+1}{m}}\right]^{\frac{m}{m+1}}. \tag{3}$$

The spatial dependence of the free carrier density, $n(x)_{free}$, within the organic reads

$$\mathrm{n(x)_{free}} = \frac{j(V_a)_\mathrm{exp}}{q\mu\,E(x)} \tag{4}$$

and since $E_{int} \neq 0$ it is a finite and continuous quantity throughout the organic.

It should be noted that it has been established that the trap-limited current occurs at higher electric field across the organic, say, $E_a \approx 10^7$ V/m and above.

The general expression for the current density, $j(E_a)_{exp}$, as a function of the applied electric field $E_a = V_a/L$ of the modified MH model description of the trap-limited current, Eq. (2), therefore reads,

$$\mathrm{j(E_a)_{exp}} = \mu qN_\mathrm{C}\left[\frac{\varepsilon\varepsilon_0}{qN_tL}\ \ u(E_a)\right]^m \tag{5}$$

where $u(E_a)$ represents the root of Eq. (1). Note that according to Eq. (1) the quantity u for the given exponent m and the applied field, $E_{a,}$ is uniquely determined by the electric field at the charge-injecting interface, E_{int}. The expression above is valid even for material parameters dependent on bias, provided that the quantity u satisfies Eq. (1).

In the limit of $E_{int} = 0$ (note that such a step, according to [16], and also on account of the resulting singularity of n_{free} at the interface, is not allowed) Eq. (1) reduces to the well known MH equation,

$$\mathrm{j(V_a)_{exp}} = \mu qN_\mathrm{C}\left(\frac{\varepsilon\varepsilon_0}{qN_\mathrm{t}}\right)^m\left(\frac{2m+1}{m+1}\right)^{m+1}\ \left(\frac{m}{m+1}\right)^m\ \frac{V_a^{m+1}}{L^{2m+1}} \tag{6}$$

an expression which is commonly used in the analysis of j–V data. Namely, whenever the measured current density, $j(V_a)_{exp}$, turns out to be described by the expression

$$\mathrm{j(V_a)_{exp}} = \mathrm{K_s}V_\mathrm{a}^\mathrm{s} \tag{7}$$

where s is larger than 2, and K_s is a bias independent constant, then the exponent s is directly related to the parameter m by the relation

$$\mathrm{s = m + 1.} \tag{8}$$

Eq. (6) is very often used in the literature for the interpretation of the trap-limited current (TLC) within the given organic structure whenever the current density is found to be proportional to the power law of the applied bias with an exponent s > 2.

3. Results and discussion

The algebraic non-linear Eq. (1) relates the known parameter E_a, the external electric field over the organic, to the two unknown parameters, the quantity u, and the interfacial field E_{int}. Thus if the bias dependence of E_{int} is known, then Eq. (1) uniquely determines (for the model under consideration) the bias dependence of the quantity u, Eq. (2), i.e. the variation of the current density and organic material properties with bias. In this work two alternative examples of the bias dependence of the electric field at the charge injecting interface placed at the coordinate x = 0, are postulated, and their influence on the solution u is investigated. In this respect the power law expression Eq. (6) according