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THE INVERTED DISTORTED PARABOLA-LIKE SHAPE OF THE BIAS-DEPENDENT ELECTRIC FIELD AT AN ELECTRON-INJECTING METAL/ORGANIC INTERFACE DEDUCED USING THE CURRENT-VOLTAGE METHOD

OBRNJENA, DEFORMIRANI PARABOLI PODOBNA ODVISNOST ELEKTRIČNEGA POLJA OD PRITISNJENE NAPETOSTI NA VMESNI PLOSKVI KOVINA/ORGANSKI POLPREVODNIK IZPELJANA Z UPORABO METODE TOKOVNE KARAKTERISTIKE

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Keywords: Metal-organic interface, Electric field, Bias-dependent interfacial field, Current density modelling, Effective electron mobility, Organic semiconductors

Abstract

Using the recently derived expression for the traditional Mott-Gurney charge-drift model extended by the non-zero electric field at the charge-injecting interface E_{int}, the published dependence of the

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current density on the applied electric field $j–E_a$ for two good-ohmic-contact, electron-only, metal/organic structures is analysed. It is argued that the Mott-Gurney law with the well-known empirical exponential bias-dependent mobility included, in spite of a very good fit to the $j–E_a$ measurements, represents an unsatisfactory method for data analyses. It is shown that the internal electric field at the electron-injecting interface is strongly bias dependent, and in such a way is coupled to the electron current within the organic bulk. The bias dependence of the interfacial field resembles an inverted, distorted, parabola-like-shaped curve, the maximum of which is organic-material dependent. Beyond the maximum, which occurs at high values of the externally applied electric field E_a, the interfacial electric field E_{int} exhibits a rapid decrease towards zero, and only at this limit can the traditional Mott-Gurney law be applied. In contrast to the present notion, it is found that the (large) electron effective mobility for the two samples investigated does not change with the bias, but it is the total effective mobility (its product with the specific non-linear algebraic function of E_a) that is bias dependent. The effective mobility may be uniquely determined, providing the applied electric field spans a sufficiently wide E_a interval. It is argued that an appropriate width of this interval may be tested by the judicious application of the derived expression in the limit $E_{int} \to 0$. The Alq_3 bias-dependent interfacial electric field at the electron injecting cathode/organic junction results in a non-linear response of the corresponding free electron density, $n_{free}(L=200\ nm)$, at this site. The possibility for an investigation of the electric field at the charge-injecting metal/organic interface using the $j–V$ method is therefore outlined.

Povzetek

V prispevku je analizirana odvisnost tokovne gostote od pritisnjenega električnega polja $j–E_a$ za primer dveh struktur kovina/organski polprevodnik, katerih značilnost so dobri ohmski kontakti in elektronsko prevajanje toka, pri čemer smo uporabili nedavno izpeljano enačbo za konvencionalni Mott-Gurney model dopolnjen z od nič različnim električnim poljem na vmesni ploskvi. Dokazano je, da Mott-Gurneyev zakon z vključeno empirično eksponentno odvisnostjo, kljub dobrem ujemanju z j-E_a meritvami, ne predstavlja zadovoljivo metodo za analizo podatkov. Pokazali smo, da je električno polje na vmesni ploskvi, na kateri poteka vbrizgavanje elektronov, močno odvisno od pritisnjene napetosti in tako povezano z elektronskim tokom skozi organski polprevodnik. Odvisnost električnega polja od pritisnjene napetosti je podobna obrnjeni, deformirani paraboli podobni krivulji, katere maksimum je odvisen od vrste organskega materiala. Konvencionalni Mott-Gurneyev zakon je možno uporabiti v limiti, ko električno polje na vmesni ploskvi, E_{int}, preseže maksimum in se potem hitro zmanjša do nule, kar se zgodi pri visokih vrednostih pritisnjene napetosti. Za razliko od trenutno veljavne razlage smo ugotovili, da se (velika) efektivna elektronska mobilnost v dveh raziskanih vzorcih ne spreminja s pritisnjeno napetostjo, pač pa je celotna efektivna mobilnost (produkt efektivne mobilnosti s specifično nelinearno algebraično funkcijo E_a) odvisna od pritisnjene napetosti. Efektivno mobilnost je možno določiti izključno pod pogojem, da se pritisnjeno električno polje spreminja v dovolj širokem intervalu. Dokazano je, da je primernost tega intervala možno preveriti s primerno uporabo izpeljanega izraza v limiti $E_{int} \to 0$. Električno polje na vmesni ploskvi katoda/organski polprevodnik Alq_3, na kateri poteka vbrizgavanje elektronov, ki je odvisno od pritisnjene napetosti, ima za posledico nelinearni odziv ustrezne gostote prostih elektronov, $n_{free}(L=200\ nm)$, na tem mestu. Podana je možnost za raziskave električnega polja na vmesni ploskvi kovina/organski polprevodnik, kjer poteka vbrizgavanje naboja, s pomočjo $j–V$ metode.

1 INTRODUCTION

An understanding of the intrinsic charge-transport properties is vital to the optimum operation of any electronic device that is based on organic semiconductors. Such devices currently used in practical applications include flat-panel displays, organic solar cells, flexible electronics, solid-state lighting, etc. An important step towards the widespread additional application of organic electronic devices is the continuous striving for a fuller understanding of the numerous factors that are currently limiting their performance. Considerable research has been devoted to uncovering the exact operation mechanisms of such devices and to a precise determination of their electrical and optical properties. In this respect, two important issues that have received a great deal of attention recently are the effects of the organic electron and hole layers on the efficiencies of devices, [1], and the exact role of chemical impurities, [2], which may either hinder or enhance the performance of the device. Both issues are intimately related to an investigation of the intrinsic charge-carrier mobility, [3-5], which is often measured using the current-voltage method, admittance spectroscopy, the time-of-flight method and transient electroluminescence.

This work is focused on a determination of the electron mobility within a single organic layer using the current-voltage method. It represents the extension of a related investigation on hole charge carrying, [6], with the aim being to dispense with the charge-density singularity at the charge-injecting metal/organic interface that characterizes the well-known Mott-Gurney law. The Mott-Gurney expression, incorporated with the empirical exponential bias-dependent mobility, [7], is of paramount importance for the charge-carrier mobility determination using the current-voltage method.

It has been shown recently, [6], that the existence of a non-zero interfacial electric field at the charge-injecting metal/organic junction causes the extinction of the singularity of the free-charge density that the Mott-Gurney law predicts. The investigation of the published *j–V* data obtained on two distinct organic structures, characterized by a series of single organic layers that differ in thickness, has shown that the effective hole mobility (to be distinguished from the total hole mobility) is bias independent. It was shown in the literature that the well-known empirical exponential bias-dependent mobility, [7], is an artefact that should be replaced by a derived, non-linear algebraic expression that depends only on the ratio of the interfacial field to the externally applied electric field [6].

However, assuming a non-zero bias-independent interfacial electric field at the *electron-injecting* cathode/organic interface, E_{int}, it is not possible to describe the electron-only current-voltage, *j–V*, data of the good-ohmic-contact, single-layer organic structures of Yasuda et al., [8]. However, these data have been analysed by the authors in terms of the Mott-Gurney law that includes the empirical exponential bias-dependent mobility, and a very good fit was obtained; see Figs. 4 and 5 of Ref. [8].

In this work, it is shown that the electron-only current density within the two organic structures investigated in Ref. [8] is strongly coupled to the bias-dependent internal electric field existing at the electron-injecting interface. The bias dependence of the interfacial electric field, E_{int}, is explicitly revealed for the published *j–V* data [8] for two metal/organic structures. For small applied fields E_a, it is shown that the interfacial electric field of both structures coincides and exhibits a *linear increase* with an increasing external electric field E_a, a behaviour that is apparently independent of the organic composition. At a certain value of E_a, which is organic-material dependent, the straight line transforms into an inverted distorted parabola-like curve,

rapidly decreasing to a value close to zero. This small value of E_{int} apparently occurs when the last, the maximum, value of the current density in the *j–V* diagram is reached. If E_{int} turns out to be negligibly small at the maximum value E_a^{max} of the E_a interval, then the traditional Mott-Gurney limit is attained. The effective mobility within the electron-only, single-layer, metal/organic structure investigated in this work turns out to be bias independent. As shown earlier in [6], apart from the E_a^2 term, the *additional external bias dependence* of the current density is provided by the previously derived, non-linear algebraic function of the argument $\lambda(E_a) = \frac{E_{\text{int}}(E_a)}{E_a}$, which is implicitly and explicitly dependent on the external electric field. This fact offers an indication that the processes that determine the electron mobility differ from the ones that determine the hole mobility. In this work, it is once again confirmed that the empirical exponential bias-dependent function for the effective mobility is redundant, within the range of the *j–V* measurements.

2 THEORETICAL OUTLINE

It can be easily verified that the drift-current density in a single-layer organic structure is, irrespective of the sign of the charge carriers, described by the expression, [6],

$$j = \frac{\varepsilon\varepsilon_0\mu_{eff}}{2L} E_a^2 \left[\frac{9}{8} - \frac{3}{2}\lambda^2 + \left(\frac{81}{64} - \frac{3}{4}\lambda^4 + 3\lambda^3 - \frac{27}{8}\lambda^2\right)^{\frac{1}{2}}\right] \tag{2.1}$$

where E_a is the externally applied electric field, defined as $E_a = V_a/L$, and V_a is the applied bias on the anode placed at the origin of the frame of reference (the cathode at $x = L$ is at zero potential), j is the current density through the metal/organic structure, μ_{eff} is the effective mobility, ε is the dielectric constant and ε_0 is the permittivity of free space, [6]. The current density, j, Eq. (2.1) replaces the well-known generalized Mott-Gurney model (in the sense that the empirical, [7], exponential bias-dependent charge mobility is included in the expression) of charged traps in the organic layer for completely empty (or equivalently, completely full) or the stated model, if the *non-zero interfacial electric field*, E_{int}, occurring at the charge-injecting metal/organic interface is taken into account. Here, the parameter λ denotes the ratio of the *non-zero* electric field at the charge-injecting interface, E_{int}, to the externally applied electric field, E_a,

$$\lambda = \frac{E_{\text{int}}}{E_a} \tag{2.2}$$

where E_{int} might be bias dependent The introduction of the interfacial field results in the disappearance of the free-charge-density singularity at the stated interface, a serious shortcoming of the above-mentioned, generalized Mott-Gurney model. Of equal importance is the fact that in Eq. (2.1) the effective mobility μ_{eff} is bias independent and the bias dependence is described in terms of the non-linear algebraic expression, which is a function of the parameter λ. Evidently, for $E_{int} = 0$, Eq. (2.1) reduces to the (original) Mott-Gurney model with a bias-independent effective mobility. In Ref. [6], Eq. (2.1) was tested on the published *j–V* data obtained on two different sets of single-layer, hole-only, metal/organic structures and good agreements with the measurements were obtained. For holes, it was shown that the interfacial electric field is bias independent and just slightly smaller than the initial value of the externally applied electric field. It was also explicitly shown that the effective mobility of the holes is thickness dependent,

but bias independent. Furthermore, it was shown that the empirical exponential bias-dependent effective hole mobility is redundant in *j–V* experiments since it represents merely an approximation of the derived algebraic expression represented by Eq. (2.1).

It can be easily shown that the expression for the current density *j*, i.e., Eq. (2.1), is invariant with respect to the sign of a drifting charge carrier within a single organic layer and is consequently also valid for electrons. Consequently, the spatial dependence of the internal electric field *E(x)* reads,

$$E(x) = \sqrt{E_{\text{int}}^{\,2} + \frac{2j(E_a)}{\varepsilon\varepsilon_0\mu_{eff}}(L-x)} \tag{2.3}$$

and the spatial dependence of the electric potential *V(x)*, taking into account that *V(L)* = 0, is then:

$$V(x) = \frac{\varepsilon\varepsilon_0\mu_{eff}}{3j(E_a)}\left\{\left[E_{\text{int}}^2 + \frac{2\varepsilon\varepsilon_0\mu_{eff}}{j(E_a)}(L-x)\right]^{\frac{3}{2}} - E_{\text{int}}^3\right\} \tag{2.4}$$

and the free-electron (number) density $n_f(x)$ reads:

$$n_f(x) = -\frac{\varepsilon\varepsilon_0\theta}{q}\frac{dE(x)}{dx} = \frac{j(E_a)\theta}{q\mu_{eff}}\frac{1}{\left[E_{\text{int}}^2 + \frac{2j(E_a)}{\varepsilon\varepsilon_0\mu_{eff}}(L-x)\right]^{1/2}} \tag{2.5}$$

In Eq. (2.5) ϑ denotes the ratio of the free to the total (free and the bound) charge densities [9], $\vartheta = \frac{n_f}{(n_f + n_b)}$ and the effective mobility [9] is defined as $\mu_{eff} = \mu\,\vartheta$.

Evidently, in the limit $E_{int} = 0$, Eq. (2.1) reduces to:

$$j(E_a) = \frac{9}{8}\frac{\varepsilon\varepsilon_0\mu_{eff}}{L}E_a^2 \tag{2.6}$$

the well-known Mott-Gurney expression, which in combination with the empirical exponential bias-dependent mobility of Gil, [7]

$$\mu_{eff} = \mu_0 \exp\left(\gamma\sqrt{E_a}\right) \tag{2.7}$$

has been in current-voltage, *j–V*, experiments exclusively used for the determination of the charge mobility. For $E_{int} \neq 0$, the curve evaluated by the combination of Eqs. (2.6) and (2.7) may, within the relevant interval of E_a, coincide with Eq. (2.1), see Ref. [6]. Here, the two parameters μ_0 and γ are determined from the fit to the experimental data, but have no clear physical meaning. However, as pointed out in Ref. [6], the combination of Eqs. (2.6) and (2.7) is incomplete, exhibiting a singularity of the free-charge density at the charge-injecting interface, see Eq. (2.5), and it should be substituted by the corresponding Eq. (2.1). This assertion was already empirically proved for hole charge carriers, [6], and so its extension to the electron current density is the subject of the work presented here.

3 BIAS DEPENDENCE OF THE INTERFACIAL ELECTRIC FIELD

Yasuda et al. [8] used the current-voltage method to investigate the electron mobility of six different electron-only, single-layer organic structures characterized by a quasi-ohmic contact. It should be emphasized that all the data were analysed in terms of Eqs. (2.6) and (2.7), since an excellent agreement between the calculated curves and the measurements was obtained, see Figs. 4 and 5 of Ref. [8]. For the purposes of this work, the interval of the externally applied electric field constitutes indispensable information for the application of Eq. (2.1). For two of the six organic structures, i.e., tris-(8-hydroxyquinoline), denoted as Alq_3, and 2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline, known as the BCP organic material, this information is clearly revealed, see Figure 2 of Ref. [8]. As a result of the excellent agreement between the data and the calculated fit shown in Ref. [8], in this work the data will be represented in terms of Eqs. (2.6), and (2.7) for the following values of the parameters, [8]: $\mu_0 = 4.7 \times 10^{-13}$ m^2/Vs and $\gamma = 6.9 \times 10^{-4}$ (m/V)$^{1/2}$ valid within the interval of the bias 1 V $\leq V_a \leq$ 20 V for the Alq_3 organic, and $\mu_0 = 2.3 \times 10^{-12}$ m^2/Vs and $\gamma = 11.0 \times 10^{-4}$ (m/V)$^{1/2}$ valid within the interval of the bias 3 V $\leq V_a \leq$ 15 V for the BCP structure. In both cases, an average value [8] L = 200 nm was taken as the layer thickness in the calculations.

It is clear that because of the considerable disagreement between the two curves shown in Figure 1, the interfacial electric field E_{int} may not be constant in the experiment of Yasuda et al. [8]. In order to test the hypothesis, the current density of Eq. (2.1) at a given E_a should equal the corresponding data (i.e., using Eqs. (2.6), and (2.7) for convenience) and the parameter λ is then deduced as a real root of the expression,

$$\mu_{eff}\left[\frac{9}{8}-\frac{3}{2}\lambda^2+\left(\frac{81}{64}-\frac{3}{4}\lambda^4+3\lambda^3-\frac{27}{8}\lambda^2\right)^{\frac{1}{2}}\right] = \frac{9}{4}\,\mu_0 \exp\left(\gamma\sqrt{E_a}\right) \tag{3.1}$$

The obtained value of λ and the associated value of the interfacial electric field at the electron-injecting interface E_{int} are presented in Table 1. Using these values in Eq. (2.1), the two curves presented in Figure 1 then coincide to an excellent degree. The deduced bias dependence of E_{int} for the Alq_3 organic (circles) is presented in Figure 2.

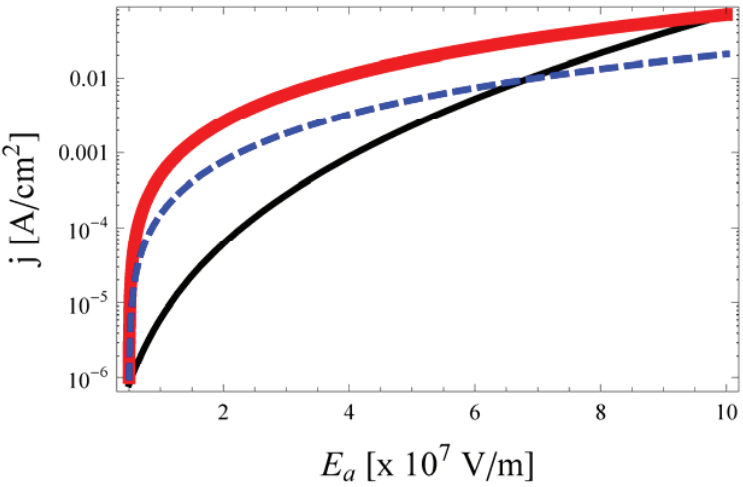

Figure 1: *The room-temperature j–E_a data of the electron-only, good-ohmic-contact, Al/Alq_3(200 nm) structure are within the complete interval 5 MV/m ≤ E_a ≤ 100 MV/m presented in the analytical form (thin line) of Ref. [8]. For fixed, i.e., bias-independent, parameters μ_{eff} = 4.7 × 10^{-10} m²/Vs and E_{int} = 4.986 MV/m, the current density calculated from Eq. (2.1) matches the data only at the initial and at the end value of the E_a interval (thick line). Assuming the interfacial electric field at the electron-ejecting contact E_{int} to be E_a dependent, then a practically exact match with the corresponding experimental current-density data is obtained. The dashed curve presents the results of Eq. (2.1) within the shortened E_a interval, i.e., 5 MV/m ≤ E_a ≤ 60 MV/m. The curve calculated for the fixed parameters μ_{eff} = 1.4 × 10^{-10} m²/Vs and E_{int} = 4.956 MV/m, which also intersects the data at the initial value of the current density, illustrates the dependence of the deduced effective mobility on the width of the E_a interval considered.*

Table 1: *From Eq. (3.1), the calculated dependence of the internal electric field, E_{int}, on the externally applied electric field, E_a, at the electron-ejecting good-ohmic-contact Al-Liac/Alq_3(200 nm) interface based on the room-temperature j-V data of Ref. [8] is presented. For comparison, taking the initial, bias independent value of the interfacial electric field $E_{int}^{ini} = 4.986$ MV/m, with a bias-independent effective mobility within the Alq_3 organic layer equal to $\mu_{eff} = 4.7 \times 10^{-10}$ m²/Vs, then the resulting curve, Eq. (2.1) (thick line), intersects the measurements, [8], only at two points, the data endpoints, Figure 1. Consequently, the deduced E_a dependence of E_{int} is presented in Figure 2 as the distorted inverted-parabola-like curve and the resulting current density (last column) as a function of E_a then practically exactly reproduces the analytical data description of Ref. [8] in terms of Eqs. (2.6) and (2.7).*

E_a [× 10^7 V/m]	λ	E_{int} [× 10^7 V/m]	j [A/cm²]
	Alq_3 L = 200 nm thick layer	**μ_{eff} = 4.7 × 10^{-10} m²/Vs**	
0.5	0.9974	0.4986	8.224×10^{-7}
1.0	0.9950	0.9950	6.225×10^{-6}
2.0	0.9876	1.9753	6.135×10^{-5}
3.0	0.9752	2.9255	2.764×10^{-4}
4.0	0.9551	3.8205	8.819×10^{-4}
5.0	0.9241	4.6203	2.306×10^{-3}
6.0	0.8769	5.2614	5.291×10^{-3}
7.0	0.8058	5.6408	1.105×10^{-2}
8.0	0.6972	5.5773	2.150×10^{-2}
9.0	0.5204	4.6839	3.957×10^{-2}
10.0	0.07736	0.7737	6.961×10^{-2}

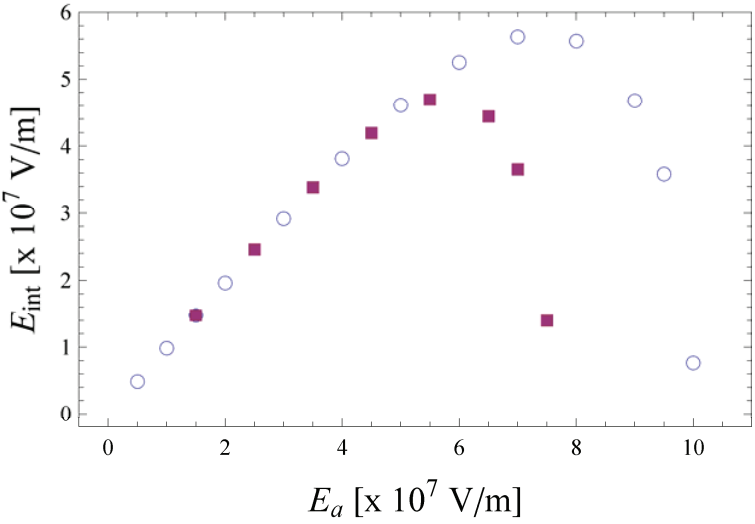

Figure 2: *The evaluated interfacial electric field E_{int}, see Eq. (3.1), at the electron-injecting metal/organic interface as a function of the externally applied electric field E_a is presented. The circles refer to the 200 nm-thick Alq_3 layer, and the filled boxes refer to the 200 nm-thick BCP layer. The calculated results are characterized by the inverted distorted-parabola-like curve, which, within the range of small values of the externally applied electric field, coincides and exhibits an identical linear E_a dependence.*

Figure 3 shows the calculated spatial dependence of the internal electric field, *E(x)*, for a 200 nm-thick Alq_3 organic layer for externally applied electric fields of E_a = 60 MV/m (dashed line) and 100 MV/m (dash-dot curve). The calculated spatial dependence of the corresponding free electron (number) density, $n_{free}(x)$, is shown in Figure 4. The calculated bias dependence of the free-electron density, $n_{free}(L=200\ nm)$, at the electron-injecting cathode/organic junction for the Alq_3 organic structure, taking into account the bias-dependent interfacial electric field of Figure 2, is presented in Figure 5.

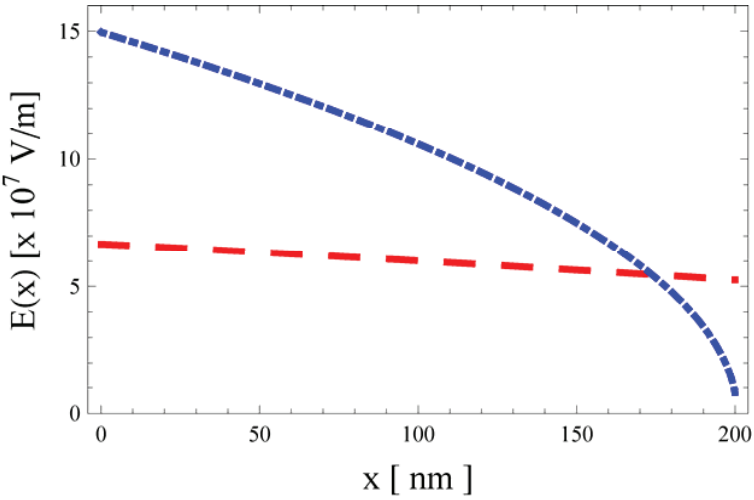

Figure 3: *The calculated spatial dependence of the internal electric field, E(x) for L = 200 nm-thick Alq_3 organic layer for two values of the externally applied electric field E_a is shown. (a) E_a = 60 MV/m (dashed line), and (b) E_a = 100 MV/m (dash-dot line). The corresponding value of the non-zero interface electric field is given in Table 1. The anode is placed at the origin, and the cathode is located at x = 200 nm.*

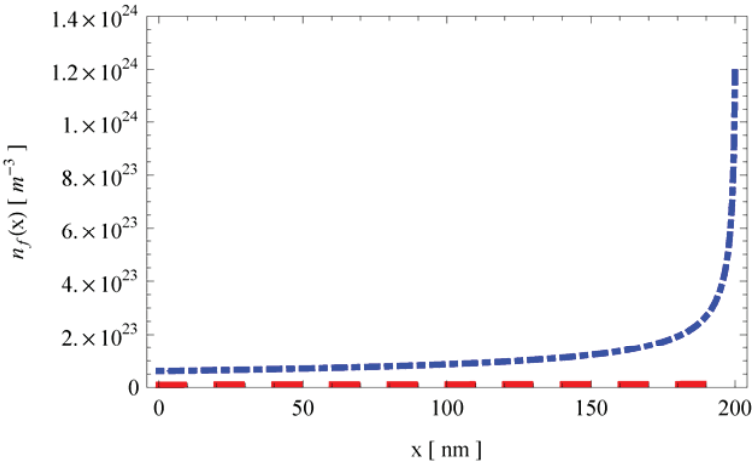

Figure 4: *The detail of the calculated spatial dependence of the free-electron (number) density $n_{free}(x)$ for the L = 200 nm-thick Alq_3 organic layer for the two values of the externally applied electric field E_a is shown. (a) E_a = 60 MV/m (dashed line), and (b) E_a = 100 MV/m (dash-dot line). The finite values of the corresponding electron charge density at both interfaces are: (a) $n_{free}(x=0) = 1.05 \times 10^{22}$ m^{-3}, and $n_{free}(x=200\ nm) = 1.34 \times 10^{22}$ m^{-3}, and (b) $n_{free}(0) = 6.19 \times 10^{22}$ m^{-3}, and $n_{free}(x=200\ nm) = 1.20 \times 10^{24}$ m^{-3}. No evidence of the space charge limited current for E_a < 60 MV/m is exhibited. The anode is placed at the origin, and the cathode is located at x = 200 nm.*

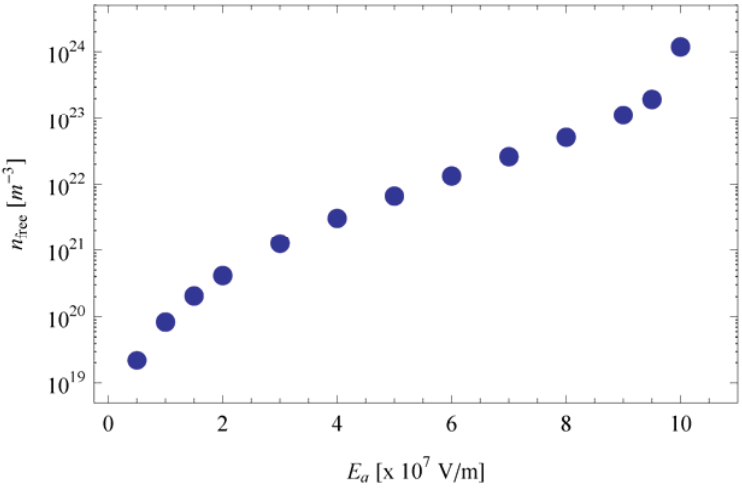

Figure 5: *The calculated bias dependence of the free-electron density, n_{free}(L=200 nm), at the electron-injecting cathode/organic junction for the Alq_3 organic structure, taking into account the bias-dependent interfacial electric field of Figure 2 is shown.*

A similar analysis can be made for the BCP structure within the interval of 15 MV/m $\leq E_a \leq$ 75 MV/m. Analogous to the above-described steps, the parameters of the curve evaluated by Eq. (2.1) through both endpoints of the complete E_a interval were found to be: $\mu_{eff} = 3.3 \times 10^{-8}$ m^2/Vs, and (the initial) E_{int} = 14.958 MV/m. In Table 2, the roots of λ and the associated E_{int} deduced from Eq. (3.1) together with the corresponding current density, Eq. (2.1), are presented, and the bias variation of the BCP interfacial electric field is depicted in Figure 2. As is clear from Figure 2, the interfacial electric field E_{int} for both organic structures exhibits, within the linear increase, practically identical behaviour, i.e., both calculated curves coincide and are linearly increasing functions of E_a. For a given value of E_a, which is organic-material dependent, the linear function transcends into an inverted-parabola-like behaviour, characterized by a rapid decrease in the interfacial field E_{int} towards zero. The maximum of the inverted-parabola-like curve occurs at E_a, which as is clear from Figure 2, is organic-material dependent. By using such deduced values of λ for each of the described two cases in Eq. (2.1), the experimental data are fully recovered. The bias-independent value of the deduced effective mobility, μ_{eff}, is clearly noted as well as the incompatibility with the corresponding result obtained on the basis of Eqs. (2.6) and (2.7).

Table 2: *From Eq. (3.1), the calculated dependence of the internal electric field, E_{int}, on the externally applied electric field, E_a, at the electron-ejecting good-ohmic-contact Al-Liac/BCP(200 nm) interface based on the room-temperature j–V data of Ref. [8] is presented. The calculated E_a dependence of E_{int} is presented in Fig. 2 as the distorted inverted-parabola-like curve and the resulting current density (last column) as a function of E_a then to an excellent approximation reproduces the analytical data description [8] in terms of Eqs. (2.6) and (2.7).*

E_a [× 10^7 V/m]	λ	E_{int} [× 10^7 V/m]	j [A/cm²]
	BCP L = 200 nm thick layer	**μ_{eff} = 3.3 × 10^{-8} m²/Vs**	
1.5	0.9972	1.4958	5.512×10^{-4}
2.5	0.9904	2.4759	5.259×10^{-3}
3.5	0.9735	3.4072	2.819×10^{-2}
4.5	0.9358	4.2111	0.112
5.5	0.8559	4.7075	0.362
6.5	0.6852	4.4539	1.030
7.5	0.1887	1.4149	2.648

On the basis of the above-presented results, we are led to the conclusion that the electron-only current density for both good-ohmic-contact structures of Ref. [8], i.e., the Al-Liac/Alq_3(200 nm) and the Al-Liac/BCP (200 nm), is strongly coupled by the non-zero bias-dependent electric field existing at the cathode/organic interface. Here, Al-Liac denotes the 40 nm-thick layer of Al with a 2 nm-thick buffer layer of lithium acetylacetonate serving as the cathode in contact with the organic layer of interest, [8].

Regarding the magnitude of the bias-independent effective electron mobility in Alq_3 and BCP, we observe that it is roughly of the same order of magnitude as the (thickness-dependent but bias-independent) hole mobility. At this point, the apparent contradiction between the stated claim and the literature reports, according to which the bias-dependent electron mobility is measured in general, deserves a comment. From examining Eq. (2.1), we see that the *total mobility* is a product of the bias-independent effective mobility, as referred to in this work, and a non-linear algebraic function of the applied field E_a.

$$M_{tot} = \mu_{eff}\left[\frac{9}{8}-\frac{3}{2}\lambda^2+\left(\frac{81}{64}-\frac{3}{4}\lambda^4+3\lambda^3-\frac{27}{8}\lambda^2\right)^{\frac{1}{2}}\right] \tag{3.2}$$

Evidently, the total mobility M_{tot} is, therefore, E_a dependent and apparently it is this bias dependence that is measured in *j–V* and similar experiments, although this fact is not recognized in the literature.

4 UNIQUENESS OF THE DEDUCED μ_{EFF}

The above-presented method for the experimental determination of the effective mobility strongly depends on the width of the E_a interval, resulting in a given set of current-voltage data. This means that one has to know, to a good approximation, the maximum value of E_a at which Eqs. (3.1) and (2.7) are no longer able to fit the data or, alternatively, the current density can no longer be measured.

Let us assume that an incomplete E_a is taken for the data analysis, as shown in Figure 1 by the dashed curve for the Alq_3 organic. The initial data point coincides with the previous one, but the end point is placed at E_a^{max} = 60 MV/m, for which the experimental current density reads j_{max} = 5.291 × 10^{-3} A/cm^2 (Table 1), while the rest of the interval is ignored. The parameters of Eq. (2.1) for which the dashed curve is passing through the stated two points are: μ_{eff} = 1.4 × 10^{-10} m^2/Vs and E_{int} = 4.956 MV/m. The new value of the effective mobility differs from the previously determined one μ_{eff} = 4.7 × 10^{-10} m^2/Vs, but E_{int} remains nearly unchanged. As seen in Figure 2 (see also Table 1), the former two values are deduced close to the upper boundary of the E_a interval, characterized by the rapid decrease towards zero of the interfacial field E_{int}. In the limit $E_{int} \rightarrow 0$, Eq. (2.1) reduces to Eq. (2.6), and we can assume that this occurs at the maximum value of the applied field $E_a = E_a^{max}$. Consequently one has:

$$\mu_{eff} = \frac{8}{9} \frac{j_{\max} L}{\varepsilon\varepsilon_0} \frac{1}{\left(E_a^{\max}\right)^2} \tag{4.1}$$

Inserting the above two values, i.e., μ_{eff} = 1.4 × 10^{-10} m²/Vs and E_a^{max} = 60 MV/m, into Eq. (4.1) means the result obtained is equal to μ_{eff} = 9.85 × 10^{-11} m²/Vs, which is in clear contradiction to the input value of μ_{eff} = 1.4 × 10^{-10} m²/Vs. The "self-consistency" test, Eq. (4.1), applied to the curve passing through the "true" end points of the E_a interval (dashed line in Figure 1) then results in μ_{eff} = 4.66 × 10^{-10} m²/Vs, which is in good agreement with the input of μ_{eff} = 4.7 × 10^{-10} m²/Vs, in spite of the fact that at 100 MV/m the corresponding E_{int} is small, but still non-zero, see Figure 2 and Table 1.

Since the E_a intervals of the four additional organic structures investigated in Ref. [8] are unfortunately not explicitly stated, this omission precludes any similar analyses.

The presented arguments then emphasize the importance that for a determination of the total effective mobility of the electron-only, good-ohmic-contact, single-layer organic structure, a set of data measured over a wide interval of bias is obtained, providing that it has a good fit with Eqs. (2.6) and (2.7) to the data. These two expressions serve as a simple approximation function for the measurement, ensuring that Eq. (2.1) may be applied with credibility. Only under such conditions is Eq. (2.1) expected to provide reliable, physically meaningful information, i.e., the magnitude of the (bias-independent) effective mobility μ_{eff} and the magnitude and bias dependence of the internal electric field at the electron-injecting cathode/organic interface, E_{int}.

5 CONCLUSIONS

Using the recently upgraded Mott-Gurney charge-drift model, as extended by the non-zero electric field at the charge-injecting interface, the experimental dependence of the current density versus the applied electric field for two good-ohmic-contact, electron-only, 200 nm-thick, Alq_3, and BCP organic layers, as published in the literature, is analysed.

It is shown that the (traditional) well-known Mott-Gurney law in combination with the empirical exponential bias-dependent electron mobility, which in fact describes the experimental j–E_a line shape to a very good approximation, represents an unsuitable method for data analysis. In the presented work, it is shown that the internal electric field at the electron-injecting interface is

strongly bias dependent, and in such a way it is coupled to the unipolar electron current within the organic layer. The bias dependence of the interfacial field resembles an inverted, distorted-parabola-like curve, the maximum of which is organic-material dependent. Beyond the maximum that occurs at high values of the externally applied electric field, the interfacial electric field rapidly decreases towards zero and only in this limit can the traditional Mott-Gurney model be applied.

It is shown that the effective electron mobility for the two samples investigated is bias independent, but its exact value may be uniquely determined, providing that the experimental electric field spans a wide enough E_a interval. As shown, the appropriate width of this interval may be tested by the judicious application of the derived expression for the drift current density.

The calculated bias dependence of the free electron density, $n_{free}(L=200\ nm)$, at the electron injecting cathode/organic junction for an Alq_3 organic structure, taking into account the bias dependent interfacial electric field, is a non-linear function of the applied electric field.

The possibility for the investigation of the electric field at the charge-injected metal/organic interface using the *j–V* method is thus outlined.

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