Space-charge limited conduction with a field and temperature dependent mobility in Alq light-emitting devices

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Abstract

Electrical transport in organic light-emitting devices (OLEDs) based on tris(8-hydroxyquinolato)aluminium (Alq) is investigated as a function of temperature and organic layer thickness. It is shown that the thickness dependence of the current provides a unique criterion to discriminate between (1) injection limited behavior, (2) trap-charge limited conduction with an exponential trap distribution and a field independent mobility, and (3) trap-free space charge limited conduction with a field and temperature dependent mobility. The observed thickness and temperature dependent current–voltage characteristics are found to be in excellent agreement with trap-free SCLC with a hopping type charge carrier mobility.

Keywords: Organic light-emitting device; Charge carrier transport

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1. Introduction

Tris(8-hydroxyquinolato)aluminium (Alq) is one of the most widely used materials in organic light-emitting devices (OLEDs) based on molecular materials. The current–voltage (I–V) characteristics of Alq devices have been investigated by several groups before, however, their interpretation has been controversial ranging from injection limited to bulk limited conduction. Burrows et al. have reported the first extended studies of temperature and thickness dependent I–V characteristics in ITO/TPD/Alq/Mg:Ag devices [1]. From the experimentally observed power law dependence of the current $j \propto V^{l+1}$ with $l > 1$ they concluded that conduction is trap-charge limited with an exponential distribution of traps having a characteristic energy $E_t = 0.15$ eV. Based on these findings Shen et al. performed numerical studies of trap-charge limited currents (TCLC) in single and double carrier devices [2]. Stößel et al. also reported TCLC in their Mg/Alq/LiF/Al devices with $E_t = 0.11$ eV [3]. We have previously also used this model to derive a trap energy of 200 meV, however, it was noticed already that the observed thickness dependence of the current does not agree with the predictions of TCLC [4]. The TCLC model can be criticized as being not applicable to these materials since it was developed for band transport rather than hopping transport [5]. Furthermore, a constant charge carrier mobility is required, which is also not fulfilled here — on the contrary, Alq displays a pronounced field and temperature dependence of the mobility typical for the whole class of disordered molecular solids and polymers [6–8]. As pointed out by Ioannidis et al. [9] the obtained values of the trap energy in the range of 110–150 meV could evenly well be assigned to a distribution of transport states which is usually taken to be a Gaussian with typical width of 100 meV. Thus, they described the I–V characteristics of Al/Alq/LiF/Al devices simply by using trap-free SCLC with a field-dependent charge carrier mobility [9]. On the other hand, Barth et al. found that their I–V characteristics on Al/Alq/Mg:Ag devices are injection limited with an injection barrier of 0.5 eV [10]. Injection limitation was also reported by Campbell et al. on Ca/Alq/Ca devices with a barrier of 0.6 eV [11]. This brief and by far not complete overview of experimental investigations and description of electrical transport in Alq devices shows that it is obviously quite difficult to uniquely assign only one mechanism for the experimentally observed behavior. Such an assignment becomes in general more difficult if the range of experimental data is too limited or if important parameters like the thickness dependence [3,10], the temperature dependence [9,11], the field and temperature dependent mobility [1–4], the built-in voltage [1,9] or the influence of recombination in a bipolar device [1,4] are not considered.

2. Basic concepts

There are two limiting regimes of device operation, namely space-charge and injection limitation of the current.

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The occurrence of space-charge limited currents (SCLC) requires that at least one contact has good injecting properties to provide an inexhaustible carrier reservoir. Injection limitation by contrast occurs if the injection barrier is large such that the injection current from the contact into the organic is much less than the SCLC.

Carrier injection into a semiconductor is treated in terms of either Fowler–Nordheim tunneling or Richardson–Schottky (RS) thermionic emission [12]. Both concepts are appropriate in inorganic semiconductors with extended band states and large mean free path, yet one can not expect that they hold in organic semiconductors, where the average mean free path is of the order of the molecular distances. The process of injection into a disordered hopping system has been studied analytically [13] and by Monte Carlo simulations [14,15]. The simulations by Wolf et al. show that, although this injection mechanism resembles RS thermionic emission, yet quantitative differences exist concerning the field and temperature dependence as well as the absolute value of the current, which is found to be orders of magnitude lower than predicted by the Richardson constant [15].

SCLC in a device can occur if at least one contact is able to inject more carriers than the material has in thermal equilibrium without carrier injection. The problem of SCLC in insulators has been extensively treated by Lampert and Mark [5]. In the case of a perfect insulator without intrinsic carriers and traps having a charge carrier mobility μ independent of the electric field and neglecting diffusion, the SCLC obeys the Mott–Gurney equation:

$$j_{\mathrm{SCLC}} = \frac{9}{8}\varepsilon\mu\frac{V^2}{d^3} \tag{1}$$

If the assumption of a field-independent charge carrier mobility is dropped, an approximate analytic solution in the absence of traps has been given for a Poole–Frenkel like field dependence of the mobility ($\mu(F) = \mu_0\exp(\beta\sqrt{F})$). The current density in this case is approximately the trap-free SCL current multiplied with the Poole–Frenkel mobility [16]:

$$j_{\mathrm{SCLC}}^{\mathrm{PF}} = \frac{9}{8}\varepsilon\mu_0\frac{V^2}{d^3}\exp\ \ 0.89\beta\sqrt{\frac{V}{d}}\Bigg) \tag{2}$$

In the presence of traps the current is in general lower and the quadratic field dependence is retained in the case of a discrete trap level only (or when all traps are filled). If traps are distributed in energy they will be gradually filled with increasing electric field and the current will increase faster than quadratic until all traps are filled. The problem has been solved analytically for an exponential trap distribution $\mathcal{N}_{\mathrm{t}}(E) = (N_{\mathrm{t}}/k_{\mathrm{B}}T_{\mathrm{t}})\exp[(E - E_{\mathrm{C}}/k_{\mathrm{B}}T_{\mathrm{t}})]$ with a constant charge carrier mobility. In this case the so-called trap-charge limited current (TCLC) is given by

$$j_{\mathrm{TCLC}} = N_{\mathrm{C}}\mu q\ \ \frac{\varepsilon l}{N_{\mathrm{t}}q(l+1)}\ ^{l}\ \ \frac{2l+1}{l+1}\ ^{l+1}\ \frac{V^{l+1}}{d^{2l+1}} \tag{3}$$

with the parameter $l = T_{\mathrm{t}}/T$ derived from the trap distribution. If both the presence of traps and a field dependent mobility are included, in general, only numerical solutions of the problem are possible.

A central question in this paper is whether the current in a device is injection or space-charge limited, and if the latter is the case, whether TCLC with an exponential trap distribution or trap-free SCLC with a field and temperature dependent charge carrier mobility play the dominant role. We will show that apart from the dependence of the current on voltage and temperature, which have already been investigated in these devices, the dependence on the thickness of the organic layer provides a unique criterion to distinguish between these cases. For this it is important to consider the thickness dependence at constant applied field for these three situations:

1. For purely injection limited behavior (regardless what the actual mechanism is in detail) the current has no explicit thickness dependence: $j = j(F) \neq j(d)$.
2. For trap-free space-charge limited conduction with (or without) a field dependent mobility the current at constant field scales with d^{-1}: $j = j(F)/d$.
3. For space-charge limited conduction with an exponential trap distribution and a constant mobility the current at constant field scales with d^{-l} with $l > 1$: $j = j(F)/d^{l}$.

3. Experimental results and discussion

From the above said it is obvious that experiments have to cover a large range of organic layer thickness and temperature and probably different electrodes in order to be able to distinguish between different models. In the following we will show experimental data for Al/Alq/Ca single layer electron-only devices as a function of organic layer thickness and temperature and will discuss these data in the framework of the above mentioned competing models. We will demonstrate that especially the thickness dependence provides a unique criterion to decide which mechanism prevails. We will also present simulations of I–V characteristics based on numerical solutions of the transport equations.

The thickness dependence of the I–V characteristics has been investigated in a series of nine devices with thickness ranging from about 100 to 350 nm all fabricated in one vacuum cycle, especially the electrodes were evaporated simultaneously. Commercially available Alq was purified by sublimation and deposited on patterned Al electrodes in a high vacuum system (10^{-6} mbar) at a deposition rate of about 1 Å/s. Fig. 1 shows the I–V characteristics of three selected thicknesses in different representations. For purely injection limited behavior the current plotted versus the electric field should be identical for different organic layer thickness and should according to the simulations by Wolf et al. follow a Richardson–Schottky-like linear dependence

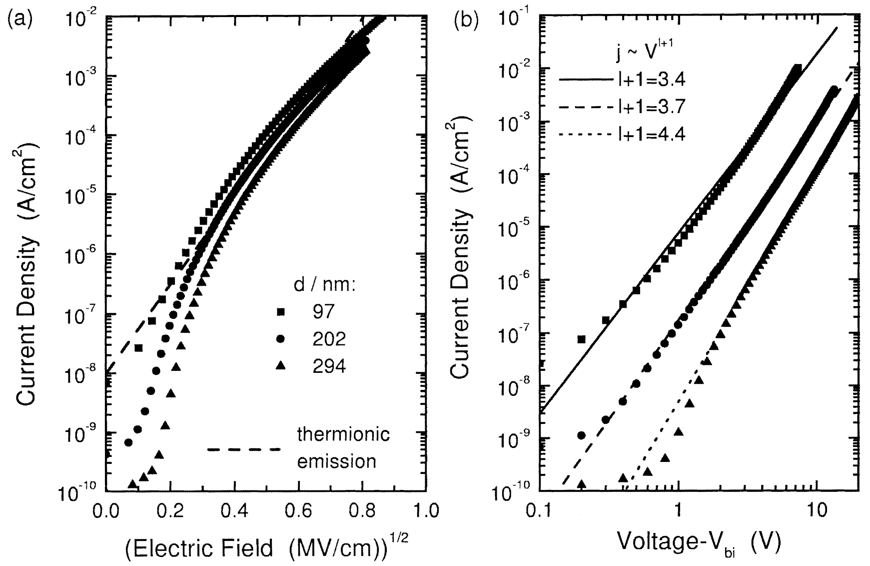

Fig. 1. Current–voltage characteristics of Al/Alq/Ca electron-only devices with different Alq layer thickness: (a) Richardson–Schottky plot; (b) double logarithmic representation with power laws (a built-in voltage $V_{\mathrm{bi}} = 0.8$ V was taken into account in all analyses).

of log(j) on the square root of the electric field. As is clearly seen in Fig. 1(a) the current does not obey such a linear dependence on $\sqrt{F}$ and the curves are not on top of each other but differ by a factor of 3 between 100 and 300 nm. From the double-logarithmic plot (Fig. 1(b)) one can see that the current approximately obeys a power law $j \propto V^{l+1}$ with $l+1$ between 3.4 and 4.4 for different thickness. This behavior could be taken as an indication for trap-charge limited conduction, which would require a correspondingly strong thickness dependence of the current.

In order to investigate the thickness dependence in more detail we have plotted in Fig. 2(a) the current at a constant field of 0.5 MV/cm versus the Alq thickness. If the current were purely injection limited, it could be expressed as a function of the field alone without an explicit thickness dependence. On the other hand, if it were space-charge limited with an exponential trap distribution, it should obey a power law of the form $j \propto V^{l+1}/d^{2l+1} = F^{l+1}/d^{l}$. With $l+1$ between 3.4 and 4.4 from the voltage dependence j should vary as $d^{-2.4}$ to $d^{-3.4}$. Thus, by plotting the current at constant electric field versus the thickness should allow to discriminate between different situations. It is clear from Fig. 2(a) that none of the two above mentioned models can account for the observed thickness dependence. It seems that

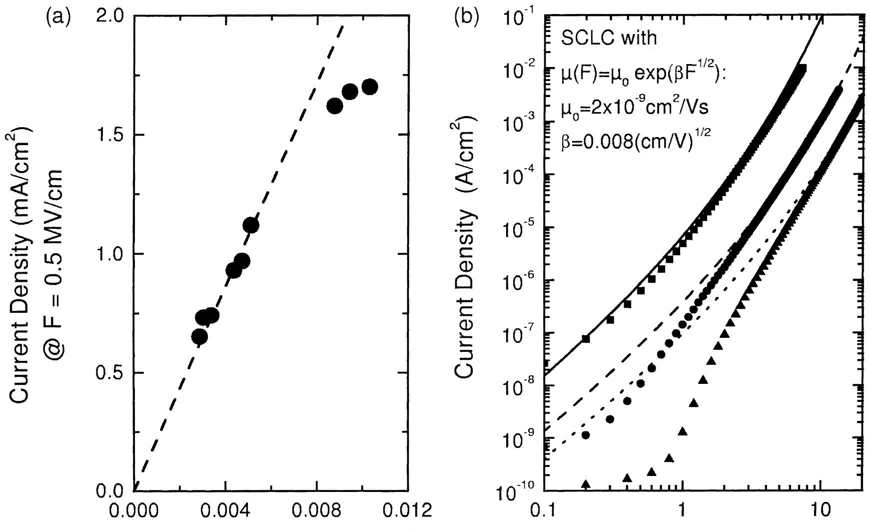

Fig. 2. (a) Thickness dependence of the current at a constant electric field of 0.5 MV/cm for Al/Alq/Ca electron-only devices with nine different values of the Alq layer thickness. (b) Current–voltage characteristics of the same devices as in Fig. 1 with simulated currents for SCLC with a field dependent mobility.

the current at constant field rather is proportional to d^{-1}. As discussed before this is exactly the behavior which is expected for trap-free SCLC with a field dependent carrier mobility. Thus, it should be possible to describe the I–V characteristics for different organic layer thickness using the same mobility parameters. This is shown in Fig. 2(b), where the experimental data for $d \approx 100$, 200 and 300 nm are compared with numerical simulations of the SCLC including a Poole–Frenkel type mobility $\mu(F) = \mu_0 \exp(\beta\sqrt{F})$ with $\mu_0 = 2 \times 10^{-9}\ \text{cm}^2/\text{V s}$ and $\beta = 0.008\ (\text{cm/V})^{1/2}$. While for the thinnest device the agreement is fairly good in the whole voltage range, there are noticeable deviations in the voltage range below 3 V for $d = 200$ nm which become more pronounced for the 300 nm device. However, this behavior is not unexpected and can be explained by trap filling. It is known that for a given trap density the crossover voltage V_{TFL} between the trap-dominated regime and the trap-filled SCL regime scales with the square of the device thickness [5]. Thus, one can expect to see an increasing influence of traps for thicker organic films. On the other hand, reducing the thickness by a factor of 3 will decrease V_{TFL} by almost an order of magnitude, which means that for a device with 100 nm thickness virtually all traps could be filled at a relatively small voltage above the built-in voltage.

The second important parameter for analyzing current–voltage characteristics is their temperature dependence. Fig. 3(a) shows a set of data for the 300 nm thick device between room temperature and 100 K in a double logarithmic plot. In this representation the curves seem to nicely follow power laws $j \propto V^{l+1}$ with increasing exponent l for decreasing temperature. This behavior is predicted in the TCLC model with an exponential trap distribution and has been used to derive the characteristic energy $E_t = k_B T_t$ from $l = T_t/T$. According to this relation a plot of the temperature dependent exponent from the power laws in the I–V characteristics versus the reciprocal temperature should yield a straight line through the origin. The corresponding plot (Fig. 3(b)) shows indeed a linear dependence with a slope yielding a trap energy of about 0.16 eV very similar to values reported earlier by other groups [1,3], however, it does not go through the origin. More severe is the fact that this model requires a mobility independent of the electric field to arrive at the used equations. As it is known from mobility measurements this is not fulfilled for Alq [6–8]. Moreover, the field dependence of the mobility becomes increasingly more pronounced for lower temperatures [8] so that there is also a strong effect of the mobility on $j(T)$. So we have analyzed the temperature dependent I–V characteristics in the framework of trap-free SCLC including a field dependent mobility of the Poole–Frenkel type with parameters μ_0 and β to be determined for each temperature. In order to minimize the influence of trap filling the analysis has been performed on the 100 nm thick device. Fig. 4(a) shows the experimental data together with numerically calculated I–V curves using adjusted μ_0 and β values to obtain the best agreement between experiment and simulation. It is seen that reasonable fits can be obtained for all temperatures, especially the characteristic upward curvature of the I–V curves is reproduced well by the simulations. There is a strong temperature dependence of μ_0 typical for disordered molecular solids [17]. The analysis of such data can be performed using the modified Poole–Frenkel model. Then the temperature and field dependent mobility is given by [18]

$$\mu = \mu_{\text{PF}} \exp\ -\frac{\Delta E - \beta_{\text{PF}}\sqrt{F}}{k_B T_{\text{eff}}}\ , \quad \frac{1}{T_{\text{eff}}} = \frac{1}{T} - \frac{1}{T_0} \tag{4}$$

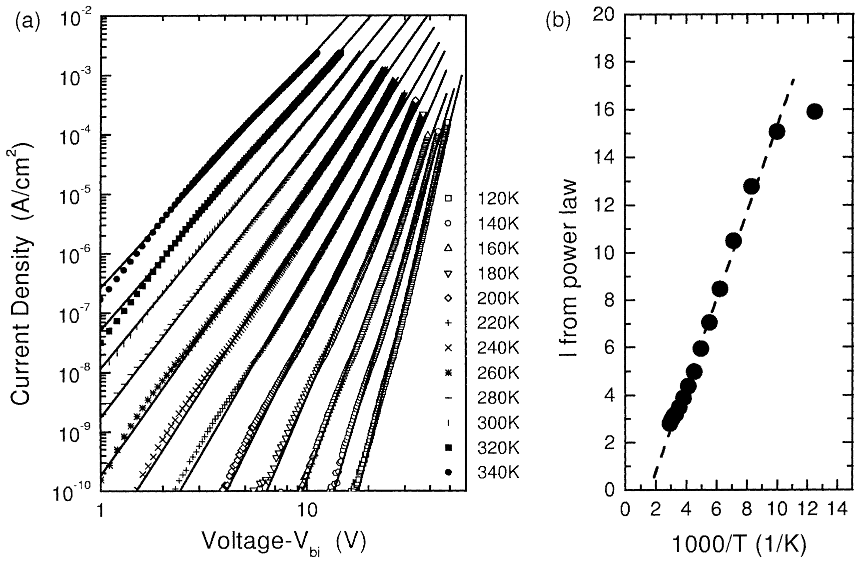

Fig. 3. (a) Temperature dependence of current–voltage characteristics of an Al/Alq/Ca electron-only device with an Alq layer thickness of 300 nm in double logarithmic representation with fits to power laws $j \propto V^{l+1}$. (b) Temperature dependence of the power law exponents l.

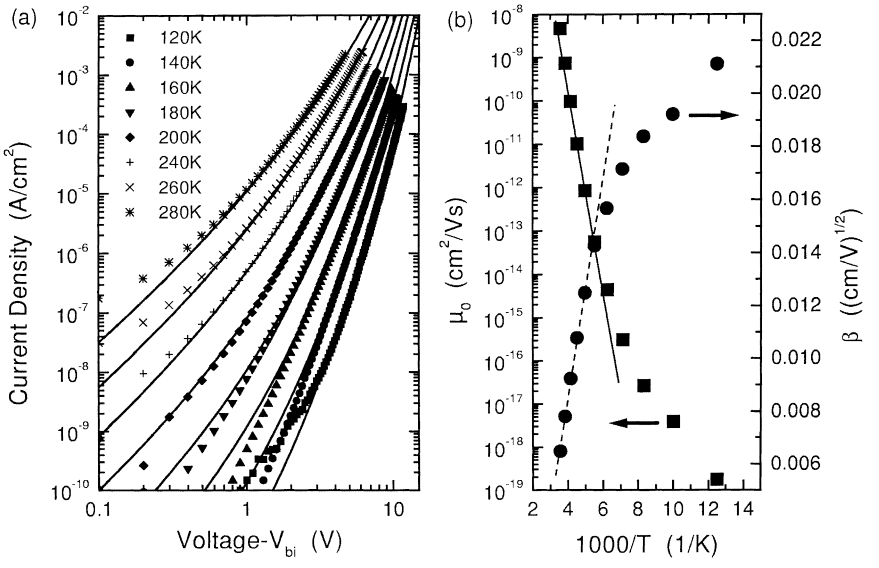

Fig. 4. (a) Temperature dependence of current–voltage characteristics of an Al/Alq/Ca electron-only device with an Alq layer thickness of 100 nm in double logarithmic representation with simulated currents for SCLC with a field and temperature dependent mobility. (b) Temperature dependence of the mobility parameters.

where ΔE is the activation energy at zero electric field, β_{PF} the so-called Poole–Frenkel factor, T_0 an empirical parameter and μ_{PF} the mobility at $T = T_0$. From a plot of $\log(\mu_0)$ and β versus $1/T$ one should obtain two linear dependencies from which it is possible to determine the parameters in the Poole–Frenkel equation. The corresponding graph is shown in Fig. 4(b). From the linear behavior of $\log(\mu_0)$ and β observed between room temperature and about 200 K the following parameters have been obtained: $\mu_{PF} = 5.4 \times 10^{-5}$ cm^2/V s, $T_0 = 500$ K, $\Delta E = 0.52$ eV and $\beta_{PF} = 5.8 \times 10^{-23}$ J(cm/V)$^{1/2}$. The deviations of $\log(\mu_0)$ and β from the linear dependence on T^{-1} below 200 K indicate that at low temperature other transport mechanisms like, e.g. tunneling may come into play. These values are in a typical range for a whole class of disordered molecular solids. Blom et al. have previously derived very similar parameters for hole transport in a soluble PPV derivative [19]. In [8] we have analyzed results on the temperature and field dependent mobility in Alq obtained by transient electroluminescence within this model and obtained comparable parameters. Thus, we can say that the dominant factor that determines the observed behavior of the current in Alq single layer devices is a hopping-type field and temperature dependent charge carrier mobility.

Of course this does not rule out an influence of traps on the device current. On the contrary, as we have seen from the thickness dependence the current can be lowered significantly by trapping at low voltage as long as the trap-filled limit is not reached. This effect is more pronounced for thick organic layers (300 nm or more) and for low temperature. To include trapping in numerical calculations information on the underlying trap distribution (concentration and energy) is needed. This issue has been addressed by measurements of thermally stimulated luminescence and currents on Alq [20,21]. Although, traps were detected in a wide energy range up to 0.5 eV, it was not possible to reconstruct the trap distribution from these data. In spite of the advantage that calculations become much easier with an exponential distribution, the huge amount of work on molecular crystals has revealed that a Gaussian distribution of traps centered around some maximum value lying in the energy gap is physically more realistic than an exponential distribution falling off from the band edge [22,23]. Thus, as long as there is no other independent information about the energetic distribution of traps in a material, the usage of an exponential trap distribution seems rather arbitrary. Certainly, more work is needed to determine trap distributions in molecular organic films.

Another aspect is also important to mention, namely the influence of charge carrier injection. The fact that the I–V characteristics can be sufficiently well described by SCLC with a field and temperature dependent mobility does not mean that the barrier for charge carrier injection is completely negligible. Looking at the thickness dependence of the current at constant field (Fig. 2(a)) one can see that for thin layers (around 100 nm) the dependence becomes weaker than $j \propto d^{-1}$, indicating an increasing influence of the injection barrier for thin layers. This means that the electric field at the injecting contact may no longer vanish as required for SCLC, nevertheless, it is still different from $F = V/d$ as it were for pure injection limitation. As has been demonstrated by Wolf et al. there is an interdependence of the tolerable injection barrier to achieve SCLC and the mobility of the organic material. Thus, the differences in device characteristics reported in the literature may be partly due to different material and preparation conditions leading to variations in the mobility of Alq and different boundary conditions for SCLC.

4. Conclusion

It has been demonstrated that the thickness dependence of the current provides a unique criterion to distinguish between different conduction mechanisms in OLEDs. The thickness and temperature dependent *I–V* characteristics of Alq electron-only single layer devices can be well described by trap-free SCLC with a hopping-type field and temperature dependent charge carrier mobility. To include the influence of trapping further independent information about trap distributions in the organic materials is required.

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References

[1] P.E. Burrows, Z. Shen, V. Bulovic, D.M. McCarty, S.R. Forrest, J.A. Cronin, M.E. Thompson, J. Appl. Phys. 79 (1996) 7991.

[2] J. Shen, J. Yang, J. Appl. Phys. 83 (1998) 7706.

[3] M. Stößel, J. Staudigel, F. Steuber, J. Blässing, J. Simmerer, A. Winnacker, Appl. Phys. Lett. 76 (2000) 115.

[4] S. Berleb, A.G. Mückl, W. Brütting, M. Schwoerer, Synth. Met. 111–112 (2000) 341.

[5] M.A. Lampert, P. Mark, Current Injection in Solids, Academic Press, New York, 1970.

[6] R.G. Kepler, P.M. Beeson, S.J. Jakobs, R.A. Anderson, M.B. Sinclair, V.S. Valencia, P.A. Cahill, Appl. Phys. Lett. 66 (1995) 3618.

[7] B.J. Chen, W.Y. Lai, Z.Q. Gao, C.S. Lee, S.T. Lee, W.A. Gambling, Appl. Phys. Lett. 75 (1999) 4010.

[8] A.G. Mückl, S. Berleb, W. Brütting, M. Schwoerer, Synth. Met. 111–112 (2000) 91.

[9] A. Ioannidis, E. Forsythe, Y. Gao, M.W. Wu, E.M. Conwell, Appl. Phys. Lett. 72 (1998) 3038.

[10] S. Barth, U. Wolf, H. Bässler, P. Müller, H. Riel, H. Vestweber, P.F. Seidler, W. Rieß, Phys. Rev. B 60 (1999) 8791.

[11] I.H. Campbell, D.L. Smith, Appl. Phys. Lett. 74 (1999) 561.

[12] S.M. Sze, Physics of Semiconductor Devices, Wiley, New York, 1981.

[13] V.I. Arkhipov, E.V. Emelianova, Y.H. Tak, H. Bässler, J. Appl. Phys. 84 (1998) 848.

[14] Y.N. Gartstein, E.M. Conwell, Chem. Phys. Lett. 255 (1996) 93.

[15] U. Wolf, V.I. Arkhipov, H. Bässler, Phys. Rev. B 59 (1999) 7507.

[16] P.N. Murgatroyd, J. Phys. D: Appl. Phys. 3 (1970) 151.

[17] P.M. Borsenberger, D.S. Weiss, Organic Photoreceptors for Imaging Systems, Marcel Dekker, New York, 1993.

[18] W.D. Gill, J. Appl. Phys. 55 (12) (1972) 5033.

[19] P.W.M. Blom, M.C.J.M. Vissenberg, Phys. Rev. Lett. 80 (1998) 3819.

[20] E.W. Forsythe, D.C. Morton, C.W. Tang, Y. Gao, Appl. Phys. Lett. 73 (1998) 1457.

[21] S. Karg, J. Steiger, H. von Seggern, Synth. Met. 111–112 (2000) 277.

[22] M. Pope, C.E. Swenberg, Electronic Processes in Organic Crystals, Clarendon Press, Oxford, 1982.

[23] E.A. Silinsh, Organic Molecular Crystals, Springer, Berlin, 1980.