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Study of applicability of Boltzmann-statistics and two mobility models for organic semiconductors

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The organic semiconductors are treated as non-degenerate based on recent experiment on validity of classical Einstein relationship. The expression of density of holes is analytically derived by using the Boltzmann statistics. The mobility model of Pasveer et al. and the exponential model of Pai modified by Blom et al. are combined to solve drift-diffusion equations to extract information about the mobility and effective density of state. The results show that the mobility model of Pasveer et al. can not well fit the experimental $J-V$ data for a rubrene single crystal from Krellner et al. both at low and high voltages, and some of the parameters extracted show inconsistent temperature dependence which should be constants in the theoretical framework of Pasveer et al. Whereas the exponential model gives satisfactory fit for experimental $J-V$ data at all voltage ranges, and the extracted parameters show correct temperature dependence. The temperature dependence of parameters contained in the exponential model of mobility can be well fitted by using simple expressions proposed by Blom et al. And the temperature dependence of effective density of state derived from the non-degenerate Boltzmann statistics is verified. The distribution of potential, electric field and density of holes are calculated as analyzed.

1. Introduction

Organic semiconductors are the object of intense investigation because of the promise of low-cost and large-area electronic applications such as radio-frequency identification (RFID) tags or active matrix display backplanes [1–7]. Large efforts of the research community have led to a constant improvement of the carrier mobility over the years. Mobility as high as $1.4\,\mathrm{cm}^2/(\mathrm{V}\cdot\mathrm{s})$ for polymers [8], $3.4\,\mathrm{cm}^2/(\mathrm{V}\cdot\mathrm{s})$ for polycrystalline films [9], and up to $30\,\mathrm{cm}^2/(\mathrm{V}\cdot\mathrm{s})$ for single-crystal-like films of small molecule organic semiconductors [10], have been achieved in thin film transistors, exceeding that of hydrogenated amorphous silicon and approaching that of polycrystalline silicon.

A major bottleneck towards the design and development of new materials is the lack of fundamental understanding of what limits charge transport in organic semiconductors. In films of crystalline organic semiconductors, the effect of trap states located in the bandgap is commonly observed [11–16]. The energetic distribution of these localized gap states created by disorder, chemical or morphological defects, or impurities affects the performance of organic devices made with these materials. The accurate characterization of this trap distribution and its correlation to the nature of defects is crucial to fully understand the fundamental limits of these materials. In this regard, investigations of highly purified organic single crystals are fundamental, as these constitute model systems where the effect of microstructural features such as grain-boundaries is suppressed. Single-crystal studies might allow us to predict the upper performance bounds of organic semiconductors as well as help design materials with ever increasing carrier mobility. The observation of traps in organic single crystals is well documented [17–21]. The traps may be caused by residual impurities, lattice disorder, or defects such as dislocations. Because in high-quality crystals trap densities are extremely low, obtaining direct evidence of their existence is challenging. Indirect evidence can be obtained by measuring their effect on the $I-V$ characteristics of semiconductor devices where the level-Fermi in the semiconductor is made to sweep through trap distributions. Experimental techniques to estimate the trap distribution include the direct fitting of space charge limited current (SCLC) characteristics to transport models and assuming a particular energetic distribution of traps [11,22,23], but at present the Gaussian distribution for density of state (DOS) is most popular, and in some times the exponential is used to replace the Gaussian DOS for simplicity.

There are two types of methods to consider trap effects in literature. The first type treating all carriers in same way, the trap effects are considered by seeing the mobility of carriers as a function of electric field or density of carriers. Two representative methods are the exponential model of Pai [24] and unified model of Pasveer [25] et al. Pai has ever proposed that the mobility of amorphous semiconductor is an exponential function of root of electric field with two temperature dependent parameters determined by fitting experimental data. As organic semiconductors becoming hotspot, the exponential model was used to describe electric properties of organic semiconductors with various modifications. Dunlap et al. [26] analytically derived the expression of mobility, confirmed the exponential model of Pai [24], obtained mobility is in quantitative agreement with observation in experiment. Subsequently, Blom et al. [27] and Novikov et al. [28] proposed different temperature dependent expressions for parameters in the exponential model.

Pasveer et al. [25] further deduce the dependence of mobility $\mu(T, p, E)$ on temperature T, density of charge carriers p , and electric field E, through numerically solving the master equation to describe hopping transport of charge

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carriers on a lattice of sites based on the uncorrelated Gaussian disorder model (GDM). Both exponential model and the model of Pasveer et al. have been widely applied to study electric properties of organic semiconductors [25–31]. The second type of method separating all carriers into free and trapped, the representative is the mobility edge (ME) model [32–43]. The ME model divides the DOS into mobile states and trap states. Mobile states, with mobility μ_0, are located above (below) the mobility edge in the case of an *n*-type (*p*-type) semiconductor. Quasi-equilibrium is assumed everywhere such that the density of mobile holes (for a *p*-type semiconductor) can be calculated using Fermi–Dirac statistics. Dacuna and Salleo [32] have used the mobility edge (ME) model [35] to simulate the experiment data of Krellner [36]. Yampolskii et al. [34] also proposed similar model without application to practical materials or devices.

It is obvious that above two types of methods are different from each other. The first type of method is simpler and more similar to traditional theory for inorganic semiconductors. Since there is not determinative judgment between two types of methods, it is meaningful to apply both to same systems to explore which one is more reasonable. In this paper, we apply the first method to single-crystal rubrene organic semiconductor as studied by Dacuna and Salleo [32] using the second type of ME model. We adopt both exponential mobility model [27] and $\mu(T, p, E)$ mobility model of Pasveer et al. [25] to solve the drift-diffusion equations to research current–voltage $(J{-}V)$ single-crystal rubrene diode and extract parameters in Gaussian DOS.

2. Method and models

It has been demonstrated that the hole current in organic diodes is space-charge limited (SCL) [12–28]. The SCL current–voltage $(J{-}V)$ characteristic usually is described by the solution of Poisson equation

$$\frac{d^2\varphi}{dx^2} = -\frac{q}{\varepsilon_r\varepsilon_0}\,p(x) \tag{1}$$

for electric potential φ or field, and drift equation for current J [37]. Where x is the coordination, q is the elementary charge, p is density of hole; ε_0 is the free-space permittivity, ε_r is the dielectric constant of the semiconductor. A general simplification used in the derivation of analytical models is to neglect the diffusion current as long as the applied voltage is larger than few kT/q. But it has been shown not necessarily true [38,39]. So we adopt the drift-diffusion equation for current:

$$J(x) = -q\mu(x)p(x)\,\frac{\partial\varphi}{\partial x} - kT_\mu\,\frac{\partial p(x)}{\partial x}, \tag{2}$$

where μ is the mobility of holes. The voltage V is the drop of electric potential

$$V = \varphi(0) - \varphi(L) \tag{3}$$

and L is the separation between the contacts, or thickness of the semiconductor layer Although the mechanism of electric conduction is not thorough determined, the Gaussian model [30] for DOS has been recognized:

$$D(E) = \left(N_0/\sigma\sqrt{2\pi}\right)\exp\left\lfloor -(E - E_v)^2/2\sigma^2\right\rfloor, \tag{4}$$

where N_0, E_v, and σ are the total number of states, the center energy and the standard deviation of the Gaussian distribution, respectively. If organic semiconductors are degenerate, the Fermi–Dirac statistics must be adopted, the density of hole is expressed as

$$p = \int_{-\infty}^{\infty} D(E)\,\frac{1}{1 + \exp\left[\left(E_{\mathrm{F}} - E + q\varphi(x)\right)/kT\right]}\,dE. \tag{5}$$

Some works pointed out the Fermi–Dirac statistics used in Eq. (5) would lead to following generalized Einstein relation (GER) instead of the traditional one [37–40]

$$\frac{D_p}{\mu} = -p\left[\frac{qdp}{dE_{\mathrm{F}}}\right]. \tag{6}$$

Where D_p is the diffusion coefficient of holes. However, Neumann et al. [41] oppose the GER through theoretical analysis. Recently, Wetzelaer et al. [42] also confirmed the validity of the Einstein relation in organic semiconductors by studying the diffusion-driven currents of single-carrier diodes. In contrast to earlier reports, a temperature independent ideality factor was found for both *n*- and *p*-type conduction, which is in contradiction with the generalized Einstein relation derived for a Gaussian DOS.

In terms of Refs. [41,42], we think that the organic semiconductors should be nondegenerate, $(E_{\mathrm{F}}{-}E_v) \gg kT$, the Fermi–Dirac statistics in Eq. (5) can be replaced by the Boltzmann statistics, Eq. (5) is changed as

$$p = \int_{-\infty}^{\infty} D(E)\exp\left[\left(E - E_{\mathrm{F}} - q\varphi(x)\right)/kT\right]dE. \tag{7}$$

Substituting Eq. (4) into Eq. (7), introducing dimensionless variable, $y = (E - E_v)/\sigma\sqrt{2}$, we can evaluate the integral

$$\int_{-\infty}^{\infty}\exp\left[-(E - E_v)^2/2\sigma^2\right]\exp\left[\frac{(E - E_v)}{kT}\right]dE = \sigma\sqrt{2\pi}\exp\left[\frac{\sigma^2}{(\sqrt{2}kT)^2}\right].$$

And the density of hole can be expressed as

$$p = N'_v\exp[-q\varphi(x)/kT]. \tag{8}$$

Here the coefficient N'_v is defined as

$$N'_v = N_0\exp\left[(E_v - E_{\mathrm{F}})/kT\right]\exp\left[\sigma^2/2(kT)^2\right] \equiv N_v\exp\left[(E_v - E_{\mathrm{F}})/kT\right]. \tag{9}$$

And $N_v = N_0 \exp[\sigma^2/2(kT)^2]$, is the effective DOS. It is obvious that this DOS is very very different from that of inorganic semiconductors:

$$N_v = \frac{2}{h^3}\,(2\pi mkT)^{3/2}. \tag{10}$$

The effective DOS Eq. (10) of inorganic semiconductors is a slowly increasing function of temperature; whereas the effective DOS of organic semiconductors is a dramatically decreasing function of temperatur. Eq. (9) can be reformulated as

$$\ln N'_v = \ln N_0 + \frac{E_v - E_\mathrm{F}}{kT} + \frac{\sigma^2}{2(kT)^2}. \tag{11}$$

It is shown that logarithm of effective DOS is a quadratic function of inverse temperature. The coefficients of Eq. (10) can directly determine Fermi energy and the standard deviation of the Gaussian distribution.

In order to solve drift-diffusion equation (2), we need the models of mobility. The first model we would consider is the model of Pasveer [25] et al. Pasveer et al. have fitted the numerical results of the master equation by using factorizing parametrization scheme:

$$\mu(T, p, E) \approx \mu(T, p) f(T, E), \tag{12}$$

$\mu(T, p)$ is a function of temperature and charge-carrier density, and

$$\mu(T, p) = \mu_0 b_1 \exp\left[-b_2\sigma^2 + (1/2)(\sigma^2 - \sigma)(2pa^3)^\delta\right], \tag{13}$$

$$\delta = 2\sigma^{-2}\left[\ln(\sigma^2 - \sigma) - \ln(\ln 4)\right], \quad \mu_0 = ev_0a^2/\sigma \tag{14}$$

with $b_1 = 1.8 \cdot 10^{-9}$, $b_2 = 0.42$, and

$$f(T, E) = \exp\left\{0.44(\sigma^{3/2} - 2.2) \times \left[-1 + \sqrt{1 + 0.8(eaE/\sigma)^2}\right]\right\} \tag{15}$$

is a function of temperature and field. Here $\sigma \equiv \sigma/kT$ is a reduced variable, and σ is the width of the Gaussian. We have three parameters, μ_0, σ and a in the model of Pasveer et al. The second model we would consider is the exponential model of Pai [24]

$$\mu_p(E) = \mu_p(0) \exp(\gamma\sqrt{E}), \tag{16}$$

where denotes the mobility at zero field. Blom et al. [27] observed a thermally activated behavior, according to

$$\mu_p(0) = \mu_0 \exp(-\Delta/kT). \tag{17}$$

They also demonstrated the linear dependence between γ and $1/T$, as following

$$\gamma = B\left(\frac{1}{kT} - \frac{1}{kT_0}\right). \tag{18}$$

This empirical dependence of γ on T has originally been proposed by Gill from TOF experiments on molecularly doped PVK [43]. We have four parameters, μ_0, Δ, B, T_0, in the exponential model.

3. Results and discussion

As pointed out by Dacuña and Salleo [32], the data set measured by Krellner et al. [36] has high quality, which span a large temperature interval (110 to 200 K), with curves that exhibit smooth variations at all temperatures and very low currents measured at low bias. As a result, this data set enables us to test our model over orders of magnitude in current density and allows us to determine the validity of the physical models we used. Dacuña and Salleo also pointed out that due to the intrinsic difference in the fabrication of the top and bottom contacts, an asymmetric behavior of the contacts was reported by Krellner et al. One side can be seen as Ohmic with low potential barrier W_left. And another side can be seen as Schottky contact with high potential barrier W_right. The difference between right and left potential barriers, $W = W_\mathrm{right} - W_\mathrm{left}$, just is the built-in potential in the device. Thus in our calculations, we need to determine three parameters N'_v, W_left and W_right at every temperatures except other parameters contained in the mobility models. And thickness of rubrene single crystal is $L = 600$ nm, the relative dielectric constant is taken as $\varepsilon_r = 3.5$.

As we use the mobility model of Pasveer et al. [25] to fit these $J-V$ data, we have six parameters N'_v, W_left, W_right, μ_0, σ and a. In our fitting procedure, it is found that $W_\mathrm{left} = 0.06$ eV, $\sigma = 0.09$ eV and $a = 2.8$ nm can be taken as constants for different temperatures, and other three parameters N'_v, W_right, μ_0 should be taken as variable for different temperatures. The coefficient N'_v can be well described by using Eq. (11), which has been derived based on the Gaussian DOS for carriers and the non-degenerate Boltzmann statistics. The determined $N_0 = 1.367 \cdot 10^{25}\,\mathrm{m}^{-3}$, $(E_v - E_\mathrm{F}) = -0.0588$ eV, $\sigma = 0.051$ eV. In terms of theoretical framework of Pasveer et al., the built-in potential W and μ_0 should be constants independent to temperature, whereas our fitting results show that they are temperature dependent, $W = (0.5488 - 0.0013q/kT)$ eV, $\mu_0 = 1.227 \cdot 10^5 \exp(0.0363q/kT)$, $(\mathrm{m^2V^{-1}s^{-1}})$.

Considering that the temperature has been included in the model of Pasveer et al., μ_0 should be seen as a constant independent to temperature, the variation of μ_0 versus temperature may exhibit some inherent inconsistence of model of Pasveer et al. Even so, we found that it is impossible to arrive at good fitting quality, even we adjust six parameters N'_v, W_right, and μ_0 at same time for every temperatures. The fitting quality for five temperatures is exhibited in Fig. 1. It can be seen from this figure that the calculated $J-V$ curves are too small as voltage higher than 2 V, and the increasing slopes are too low. At middle voltage range (0.8–1.5 V), the curves are qualitatively in agreement with data points. But at low voltage range (< 0.8 V), the curves deviate from the data points evidently, even the shapes are not correct. Therefore the results do not support the mobility model of Pasveer et al. We then apply the exponential mobility model to fit the experimental data. We have seven parameters N'_v, W_left, W_right, μ_0, Δ, B, and T_0 need be determined. It should be pointed out

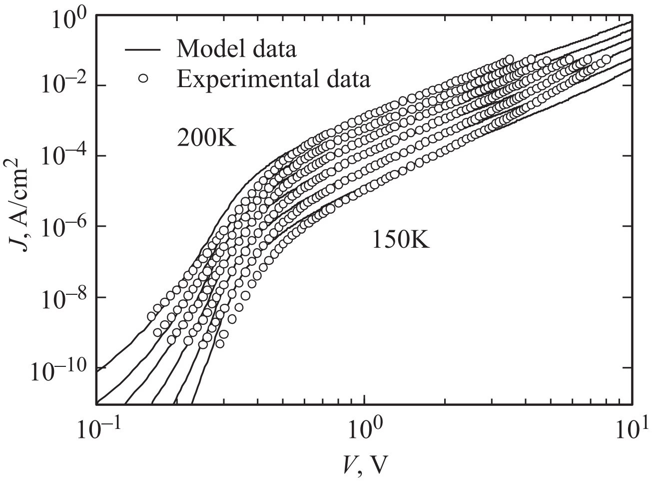

Figure 1. $J-V$ relationship from 150 to 200 K (with varying step 10 K) for a rubrene single crystal diode ($L = 600$ nm). Symbols: Experimental data from [23]; lines: theoretical results using the mobility model of Pasveer.

that the number of parameters in the exponential model seemingly is more than that of the model of Pasveer et al., considering that W and μ_0 in the model of Pasveer et al. should vary with temperature, and other two parameters have been introduced to describe such variation, so we think that the number of independent parameters in the model of Pasveer et al. even more than that in the exponential model.

Our fitting procedure shows that the $W_{\text{left}} = 0.06$ eV and $W_{\text{right}} = 0.5$ eV can be taken as constants independent with temperature; and such asymmetric values are very important for good fitting of $J-V$ data at low voltages, as having been shown by Dacuña and Salleo [32]. We then need determine three parameters N'_v, $\mu_p(0)$ and γ in Eq. (16) for every temperatures. The optimized values are plotted in Figs. 2–4 with smoothed curves fitted by using Eqs. (11,17,18), respectively. Fig. 2 shows that the coefficient N'_v also can be well described by using Eq. (11). The determined $N_0 = 4.866 \cdot 10^{24}\,\text{m}^{-3}$, $(E_v - E_F) = -0.0492$ eV, $\sigma = 0.0382$ eV. The equivalent temperature 492 K of $(E_v - E_F)$ shows that the non-degenerate approximation used to derive Eq. (11) is reasonable, and the classical Einstein relation used in the drift-diffusion equation (2) also is tenable. Figs. 3 and 4 show that $\mu_p(0)$ and γ in Eq. (16) can be well fitted by using Eqs (17,18). $\mu_p(0)$ indeed holds the thermally activated behavior versus temperature T, as having pointed out by Blom [27]. And the linear relationship of γ versus $1/T$ also is well satisfied. The determined coefficients are $\mu_0 = 0.1532\,\text{m}^2\text{V}^{-1}\text{s}^{-1}$, $\Delta = 0.2662$ eV, $B = 3.713 \cdot 10^{-5}\,\text{eV}\,(\text{m/V})^{1/2}$, and $T_0 = 394$ K, respectively.

Fig. 5 compares the experimental data to the numerical results of the exponential model. The figure shows that agreement of the numerical results of the exponential model is fairly satisfactory at low, middle and high voltage ranges, and is superior to that of the model of Pasveer et al.

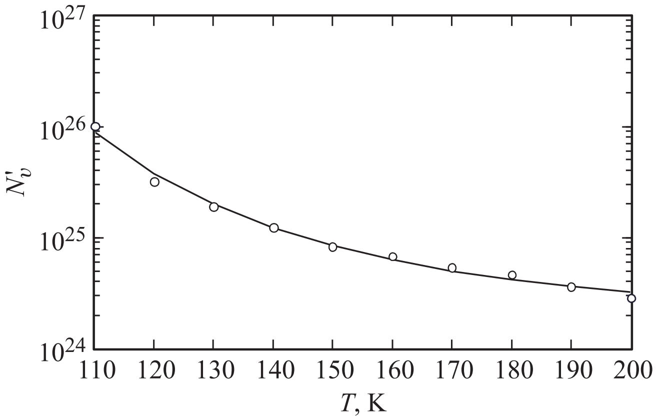

Figure 2. Varition of the coefficient N'_v versus temperature T (110–200 K). Symblos: optimized values using the exponential mobility model; line: soothed curve using Eq. (9).

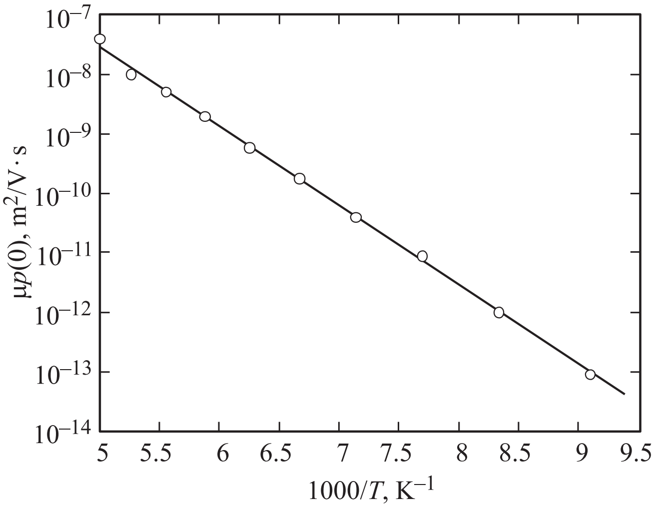

Figure 3. Varition of coefficient $\mu_p(0)$ in Eq. (16) versus temperature T (110–200 K). Symblos: optimized values using the exponential mobility model; line: soothed curve using Eq. (17).

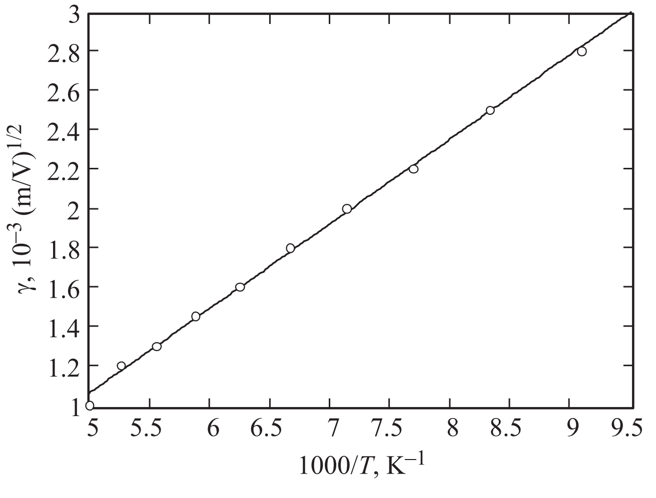

Figure 4. Varition of parametre γ in Eq. (16) versus temperature T (110–200 K). Symblos: optimized values using the exponential mobility model; line: soothed curve using Eq. (18).

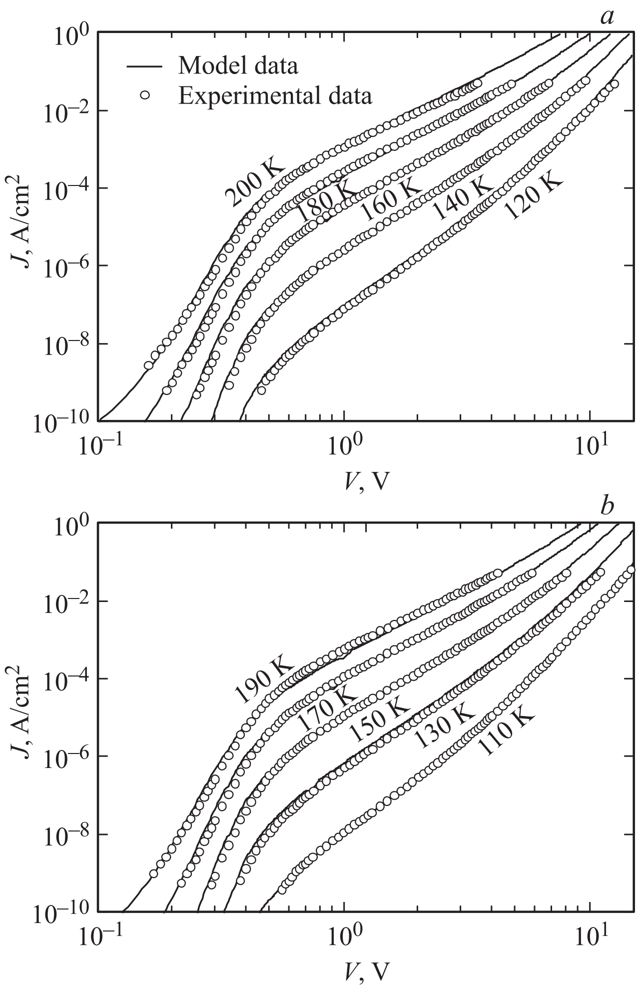

Figure 5. *a* — $J-V$ relationship from 120 to 200 K (with varying step 20 K) for a rubrene single crystal diode ($L = 600$ nm). Symbols: experimental data from [23]; lines: theoretical results using the exponential mobility model. *b* — $J-V$ relationship from 110 K to 190 K (with varying step 20 K) for a rubrene single crystal diode ($L = 600$ nm). Symbols: experimental data from [23]; lines: theoretical results using the exponential mobility model.

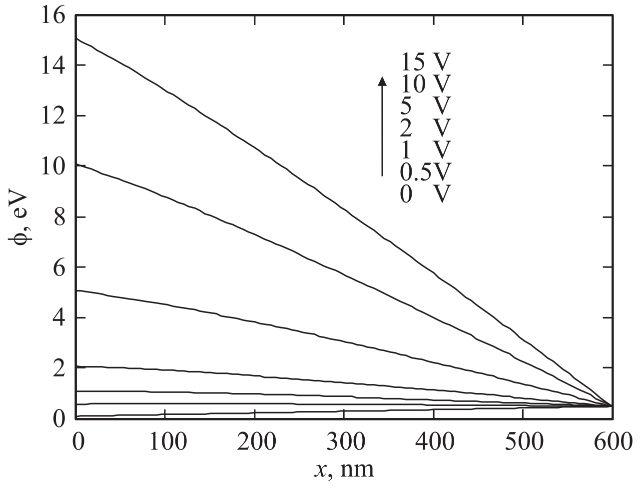

Figure 6. Variation of electric potential versus position x at 110 K and different applied biases V, V: 0.5, 1, 2, 5, 10, 15.

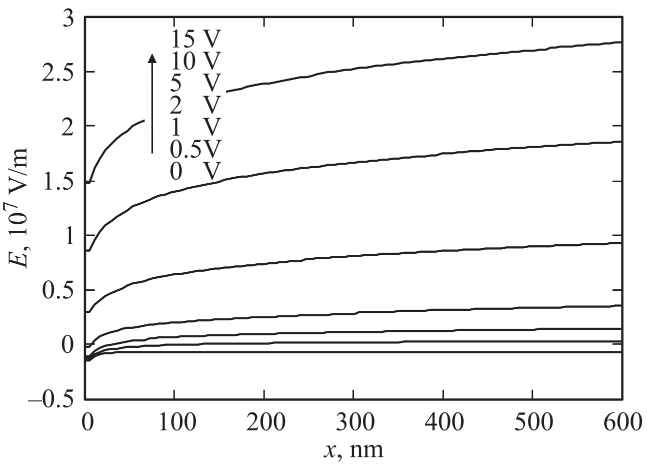

Figure 7. The electric field versus position x at 110 K and different applied biases V, V: 0.5, 1,2, 5, 10, 15.

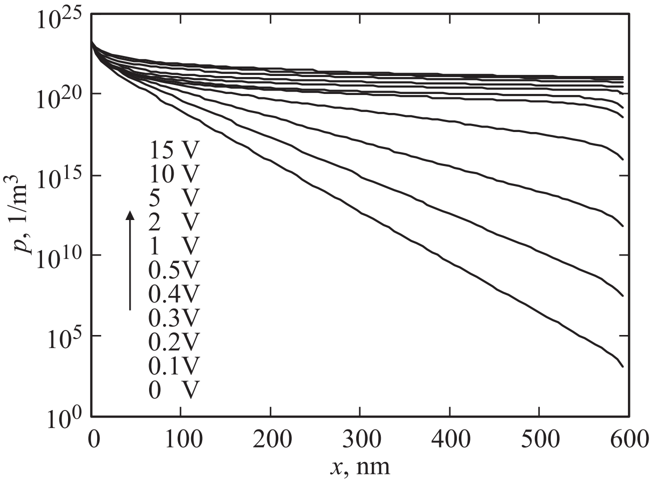

Figure 8. The hole densities versus position x at 110 K and different applied biases V, V: 0, 0.1, 0.2, 0.3, 0.4, 0.5, 1, 2, 5, 10, 15.

shown in Fig. 1. In Figs 6, 7 and 8, we plot variation of electric potential, field and density holes versus position, respectively. It can be seen from Fig. 6 that electric potential is increasing function of position at zero bias voltage for built-in potential, and becomes decreasing of position as bias voltage increasing. Fig. 7 shows that electric field always is increasing function of position, but its direction would turn from zero to high voltages. Fig. 8 shows that density of holes always is decreasing function of position, but the slope would decrease as the voltage increasing. The density of holes always is increasing function of voltage, but the increase would slow down at high voltage.

4. Conclusion

In this paper, the organic semiconductors are treated as non-degenerate based on recent experiment on validity of classical Einstein relationship. The expression of density of holes is analytically derived by using the non-degenerate Boltzmann statistics. The effective DOS shows variation versus temperature different from the traditional inorganic semiconductors, and is verified by analysis of experimental data for single-crystal organic semiconductor conducted by Krellner et al.

We then combine the mobility model of Pasveer et al. [25] and the exponential model of Pai modified [24] by Pasveer et al. to solve drift-diffusion equations to extract information about the mobility and effective DOS. The numbers of parameters needed to fit experimental $J-V$ data for a single-crystal organic diode are the same. The results show that the mobility model of Pasveer et al. can not well fit the experimental $J-V$ data both at low and high voltages, and some of the parameters extracted show inconsistent temperature dependence which should be constants in the theoretical framework of Pasveer et al.

Whereas the exponential model gives satisfactory fit for experimental $J-V$ data at all voltage ranges, and the extracted parameters show correct temperature dependence. The temperature dependence of effective DOS derived from the non-degenerate Boltzmann statistics is verified. The temperature dependence of parameters contained in the exponential model of mobility can be well fitted by using simple expressions proposed by Blom et al. Then the distribution of potential, electric field and density of holes are calculated as analyzed. These results are useful for determination of the mechanism electric conductance in organic semiconductors.

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