An improved mobility model with analytic current-voltage expression for disordered organic semiconductors

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The influence of temperature, electric field, and carrier density on hopping transport in disordered organic semiconductors is studied, and an improved mobility model that accounts for all those effects is derived. The model can accurately reproduce the experimental current-voltage characteristics of organic devices and can rather well fit numerical simulations from the master equation, and provides a clear picture of several physical effects by means of some mathematical expression. The space-charge limited current (SCLC) equations can be strictly solved with carrier density and electric field being replaced by their average value. An analytic current-voltage expression is derived, and is further validated with experimental data collected from different organic materials.

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1. Introduction

Disordered organic semiconductors have received widespread attention in the scientific community due to their easy manufacturing and possible applications in modern electronic devices such as organic light emitting diodes (OLEDs), organic field effect transistors (OFETs), and organic solar cells (OSCs) [1-8]. The efficiency of these electronic devices depends mainly on the transport properties of charge carriers in organic materials. Thus, an understanding of charge transport in disordered organic semiconductors is very important to design and synthesize better materials that can further improve the performance of organic electronic devices [9–11]. The central transport parameter is the charge carrier mobility μ which is much smaller than that of inorganic counterparts due to the spatial and energetic disorder. The mobility μ variation with the applied electric field E, temperature T, and charge carrier density p has been extensively studied both theoretically and experimentally [12–18]. Despite all the numerous consistent theoretical and experimental outputs, there is no consensus on a unified expression that accounts for charge transport in disordered organic semiconductors.

In disordered organic semiconductors, charge transport is based on thermally activated hopping [19-21]. The most commonly used method to estimate the mobility and the underlying hopping and disorder parameters is that of the space-charge limited current (SCLC) [22-24]. In a single-carrier device with an Ohmic contact, the current will be limited by the transport in the bulk of the semiconductors, commonly known as SCLC. The SCL current-voltage ($J-V$) characteristics is described by the solution of Eqs.(1-3):

$$J = p(x)e\mu(T, p(x), E(x))E(x) \tag{1}$$

$$\frac{dE}{dx} = \frac{e}{\varepsilon} p(x) \tag{2}$$

$$V = \int_0^L E(x)dx \tag{3}$$

where J is the current density, x is the distance from the injecting electrode and e is the elementary carrier charge; $\mu(T, p, E)$ is the carrier mobility as a function of temperature T, carrier density p and electric field E; $\varepsilon = \varepsilon_0\varepsilon_r$, ε_0 is the vacuum permeability, ε_r is the relative dielectric constant of the organic semiconductors; V is the potential along the device and L is the organic semiconductor layer thickness sandwiched between two electrodes.

It should be noted that there is no strict solution for the SCL Eqs. (1-3). In this work, we propose an improved expression of $\mu(T, p, E)$ by analyzing numerical results from the master equation. The improved expression has some merits as compared with the original one [12]. Firstly, the fitting precision to numerical results from the master equation is improved at high densities and high electric fields. Secondly, with p being assumed as constant in the modified $\mu(T, p, E)$, the corresponding SCL Eqs. (1-3)

can be strictly solved. Finally, by substituting p and E in $\mu(T, p, E)$ with their average values of $\overline{p}$ and $\overline{E}$, we can obtain an analytic $J-V$ expression that accounts for the most important physical quantities that influence charge transport in disordered organic semiconductors: temperature, electric field and carrier density.

2. Models and methods

Pasveer et al. have fitted the numerical results from the master equation by the following parameterization scheme [12]:

$$\mu(T, p, E) = \mu(T, p) f(T, E) \tag{4}$$

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

$$\mu(T, p) = \mu_0 b_1 \exp\left[-b_2\hat{\sigma}^2 + \frac{1}{2}(\hat{\sigma}^2 - \hat{\sigma})(2pa^3)^{\delta}\right] \tag{5}$$

$$\delta \equiv 2[\ln(\hat{\sigma}^2 - \hat{\sigma}) - \ln(\ln 4)]/\hat{\sigma}^2, \quad \mu_0 \equiv a^2\nu_0 e/\sigma \tag{6}$$

where $b_1 = 1.8\times10^{-9}$ and $b_2 = 0.42$, $\hat{\sigma} \equiv \sigma / k_B T$ is the reduced disorder parameter, σ is the width of Gaussian density of states (DOS), a is the lattice constant and ν_0 is the attempt-to-hop frequency. Eqs.(5) and (6) are obtained merely as the description of the numerical results from the master equation in a limited parameter range [12].

$$f(T, E) = \exp\left\{0.44\left(\hat{\sigma}^{3/2} - 2.2\right)\left[\sqrt{1 + 0.8\left(\frac{eaE}{\sigma}\right)^2} - 1\right]\right\} \tag{7}$$

is a function of temperature and electric field. Eqs. (4-7) are sometimes considered universal and they are the basis for commercially available organic devices simulation software [14, 25], which is called the extended Gaussian disorder model (EGDM). However, the methodology followed to derive the above EGDM parametrizations has been heavily criticized for giving an inadequate description of especially the field dependence of the mobility [15, 18, 20, 21].

We carefully compare the mobility model in Eqs. (4-7) with numerical results from the master equation, and find that errors at high carrier density are evident, as shown in Figs. 1 and 3. This also resulted in large errors for the electric field dependence of Eq. (7) at high carrier density, as shown in Figs. 2 and 3. Thus, we propose an improved factorizing parameterization scheme adopting an approach similar to Pasveer et al. and Xue et al. [12, 26]:

$$\mu(T, p, E) = \mu_s(T, p)\left[1 + \gamma(eaE/\sigma)^2\right]^8 \tag{8}$$

$$\mu_s(T, p) = \mu_0(T)\exp\left[(0.45\hat{\sigma}^2)(pa^3)^{\delta}\right] \tag{9}$$

where $\mu_0(T)$, γ, and δ are temperature dependent coefficients.

$$\mu_0(T) = \mu_0 \exp(-20 - 0.44\hat{\sigma}^2) \tag{10}$$

$$\gamma = 0.01\hat{\sigma}^2 - 0.021\hat{\sigma} + 0.008 \tag{11}$$

$$\delta = (1.3\hat{\sigma} - 0.5)^{-1} \tag{12}$$

Obviously, the improved model of Eqs.(8-12) contains three independent parameters, μ_0, a, and σ, as also in the original model of Eqs. (4-7).

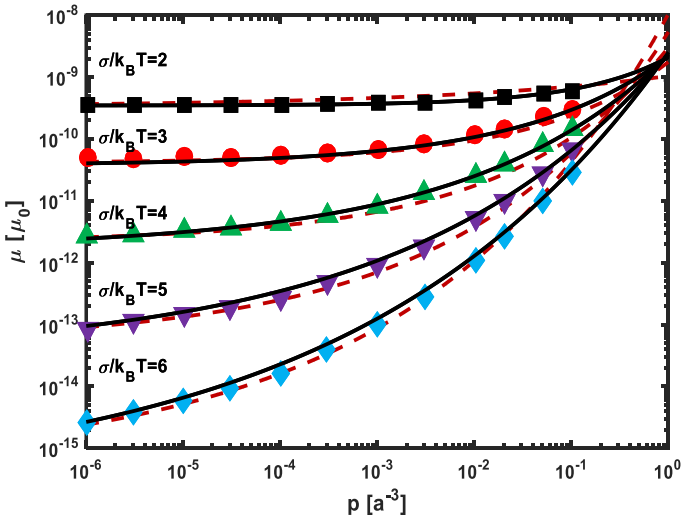

Fig. 1. Carrier density dependence of the mobility at various temperatures for a vanishing electric field. Symbols are numerical results from Ref. [12]. The dashed lines and solid lines represent theoretical fits using the parametrization scheme given in Eqs. (4-7) and Eqs.(8-12), respectively (color online)

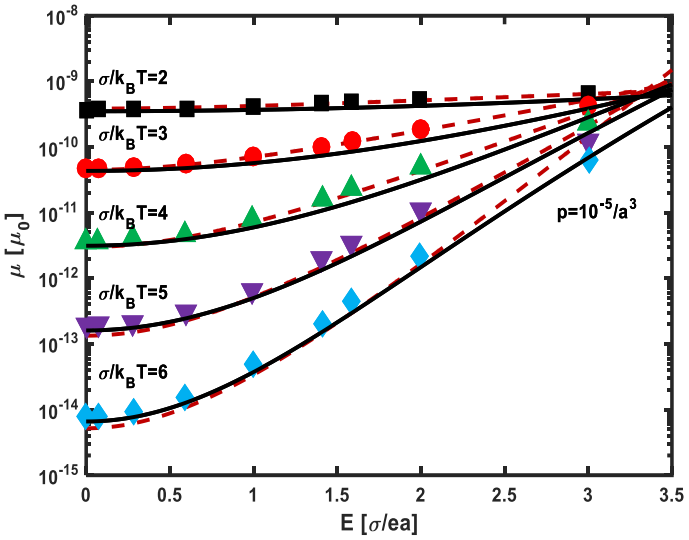

Fig. 2. Electric field dependence of the mobility at various temperatures for low densities in LEDs. Symbols are numerical results from Ref. [12]. The dashed lines and solid lines represent theoretical fits using the parametrization scheme given in Eqs. (4-7) and Eqs.(8-12), respectively (color online)

It should be noted that the SCL Eqs.(1-3) cannot be analytically solved by using the mobility model of Eqs. (4-7), and the analytic $J-V$ expression is inaccessible based on the EGDM. However, the SCL Eqs. (1-3) can be analytically solved based on the improved model of Eqs. (8-12) using $\mu(T, \bar{p}, \bar{E})$ by substituting p and E with their average values of $\bar{p}$ and $\bar{E}$, and then an analytic $J-V$ expression can be derived. The $\mu(T, \bar{p}, \bar{E})$ can be obtained as follows. Substituting Eq. (2) into Eq. (1), we obtain

$$J = \varepsilon\mu(T, p(x), E(x))E(x)\frac{dE}{dx} \tag{13}$$

Applying the boundary condition $E(0) = 0$ when integrating Eq. (13), we obtain

$$E(x) = \left(\frac{2Jx}{\varepsilon\mu}\right)^{\frac{1}{2}} \tag{14}$$

Then, the mean values of E and p can be calculated as

$$\bar{E} = L^{-1}\int_0^L E(x)dx = \frac{V}{L} \tag{15}$$

$$\bar{p} = L^{-1}\int_0^L p(x)dx = \frac{\varepsilon}{eL}E(L) = \frac{3\varepsilon V}{2eL^2} \tag{16}$$

Substituting Eq. (16) into Eq. (9), we can obtain an improved temperature and carrier density dependence.

$$\mu_s(T, \bar{p}) = \mu_0(T)\exp\left[(0.45\hat{\sigma}^2)\left(\frac{3\varepsilon Va^3}{eL^2}\right)^{\delta}\right] \tag{17}$$

Substituting Eqs. (15) and (17) into Eq. (8), we can obtain an improved temperature, electric field and carrier density dependence.

$$\mu(T, \bar{p}, \bar{E}) = \mu_s(T, \bar{p})\left[1 + \gamma(ea\bar{E}/\sigma)^2\right]^8 \tag{18}$$

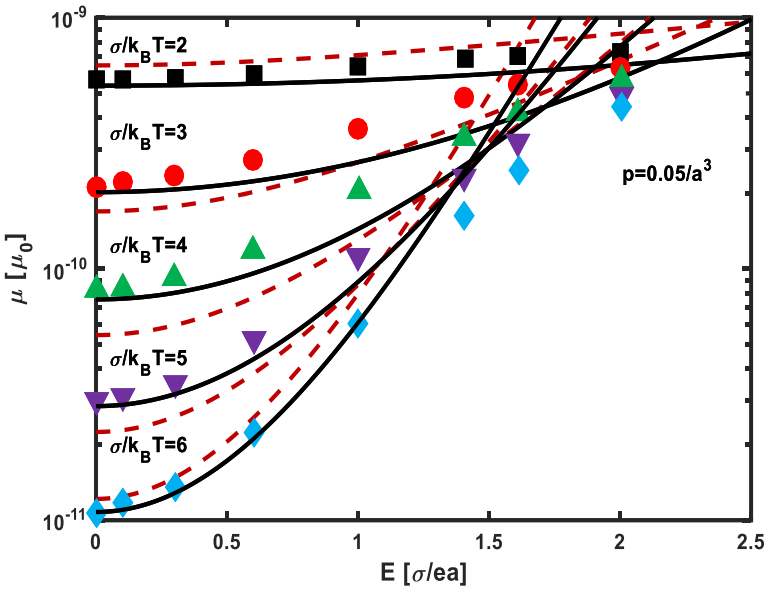

Fig. 3. Electric field dependence of the mobility at various temperatures for high densities in FETs. Symbols are numerical results from Ref. [12]. The dashed lines and solid lines represent theoretical fits using the parametrization scheme given in Eqs. (4-7) and Eqs.(8-12), respectively (color online)

3. Results and discussion

In this section, we will firstly compare the improved mobility model in Eqs. (8-12) with the EGDM, and then apply our improved model and numerical calculation method as described in section 2 to various disordered organic semiconductors. In Fig. 1, we display the carrier density p dependence of the mobility μ for different temperatures. It can be found that our description is fairly good agreement with the numerical results from the master equation. In particular, it is clear that Eqs. (8-12) give an improved description for numerical results at high carrier density compared with Eqs. (4-7).

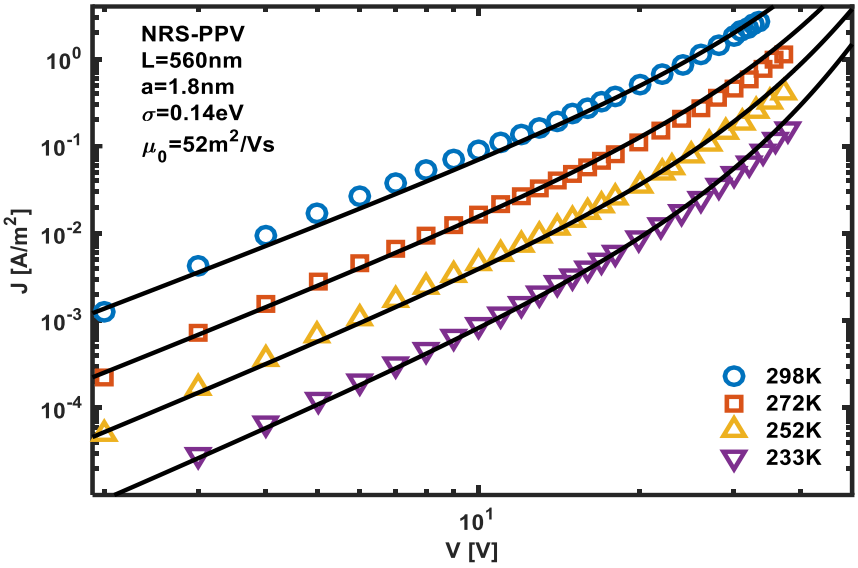

Fig. 4. Temperature dependent J-V characteristics of NRS-PPV hole-only device. Symbols are experimental data from Ref. [12]. Lines are the numerically calculated results from the improved mobility model (color online)

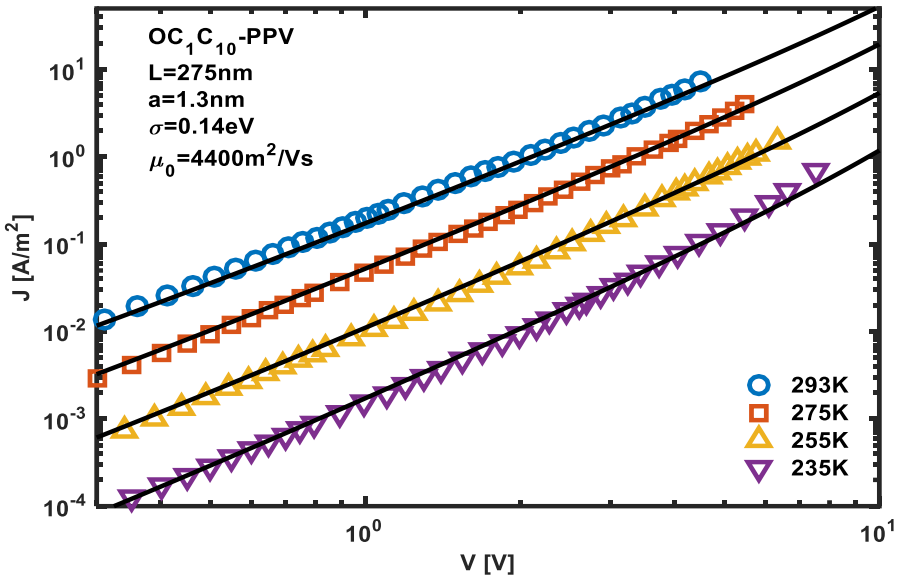

Fig. 5. Temperature dependent J-V characteristics of OC_1C_{10}-PPV hole-only device. Symbols are experimental data from Ref. [12]. Lines are the numerically calculated results from the improved mobility model (color online)

In Fig. 2 and Fig. 3, the mobility μ as a function of the electric field E is plotted at a low carrier density, $p = 10^{-5}/a^3$, a typical value for the operation regime of organic light emitting diodes, and a high carrier density, $p = 0.05/a^3$, a typical value for the operation regime of organic field effect transistors. It can be found that the agreement of electric field dependence given by both Eqs. (8-12) and Eqs. (4-7) with numerical results is satisfactory at low carrier density. However, the improved model gives slightly better description at high carrier density and high electric field than the EGDM. We think that the reason can be found in Fig. 1, both the improved model and EGDM exhibit equivalently good agreement with numerical results at low carrier density, but the improved model give slightly better results than the EGDM at high carrier density.

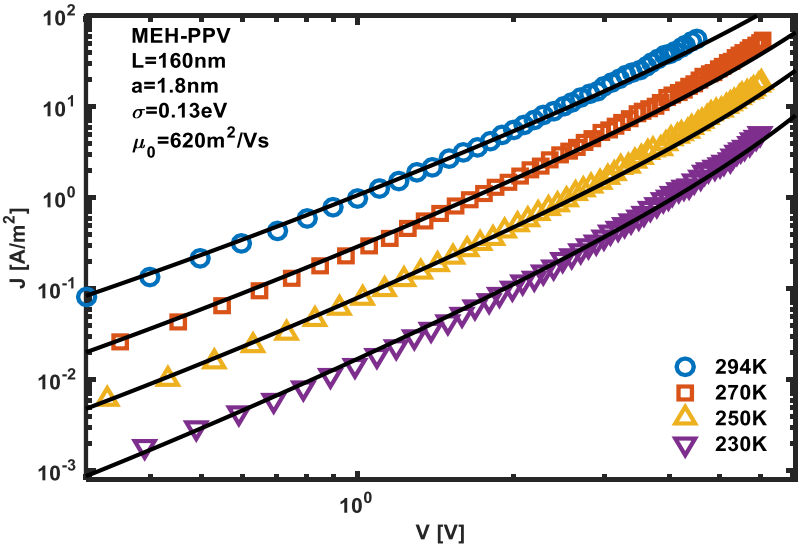

Fig. 6. Temperature dependent J-V characteristics of MEH-PPV hole-only device. Symbols are experimental data from Ref. [27]. Lines are the numerically calculated results from the improved mobility model (color online)

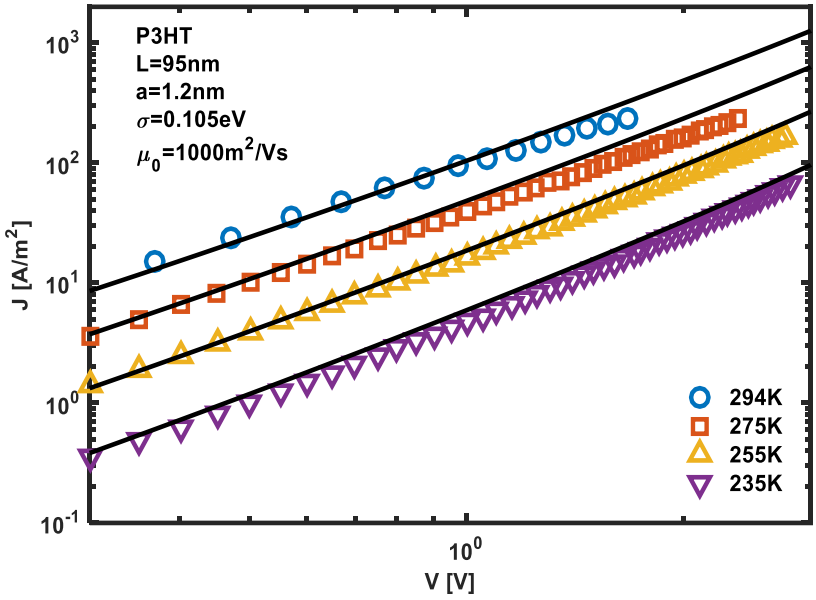

Fig. 7. Temperature dependent J-V characteristics of P3HT hole-only device. Symbols are experimental data from Ref. [28]. Lines are the numerically calculated results from the improved mobility model (color online)

In Figs. 4-6, we display the solution of Eqs. (1-3) with the T, p, and E dependence on μ from the improved model and the experimental $J-V$ measurements of NRS-PPV, OC_1C_{10}-PPV and MEH-PPV hole-only devices. It is clear that the improved model can excellently describe the temperature dependent $J-V$ curves of hole-only devices based on these PPV polymers using only three parameters, viz. σ, a and μ_0, each with a clear physical meaning. The parameters σ, a and μ_0 are determined by using a way that an optimal overall fit is obtained. Here, σ mainly controls the temperature and carrier density dependence of the mobility, whereas a predominantly affects the field dependence of the mobility and μ_0 only influences the magnitude of the mobility. Moreover, we note that the improved model provides a much better description to the $J-V$ curves for these PPV polymers than the EGDM [12], especially at low temperatures and low voltages. This also further suggests that our description is a useful approach to study the $J-V$ relationship of disordered organic semiconductors.

In order to investigate the applicability of our improved model, we further compare it to experimental measurements on P3HT. Fig. 7 shows the numerically calculated $J-V$ characteristics of P3HT hole-only device using the improved model, as well as the experimental data from Ref. [28]. It can be seen from the figure that our calculated results are in fairly good agreement with the original experimental data. The excellent description of $J-V$ curves strongly suggests that the underlying dependence of the mobility on temperature, carrier density, and electric field in these materials are correctly described by the improved model. Therefore, we conclude that the improved model captures the physics of charge transport in disordered organic semiconductors. It is also confirmed that the improved model is more applicable for a large variety of disordered organic semiconductors than the EGDM.

4. Summary and conclusions

In summary, an improved expression of $\mu(T, p, E)$ is developed by analyzing numerical results from the master equation. The precision at high carrier density and high electric field is improved by using our description. The SCLC equations can be strictly solved with carrier density and electric field being replaced by their average value. Then, an analytic $J-V$ expression that accounts for the most important physical quantities that influence charge transport in disordered organic semiconductors is derived. The excellent fits we obtain with experimental current-voltage data of four semiconducting polymers strongly suggest that the underlying dependencies of the mobility on temperature, carrier density, and electric field in these materials are correctly described by the present work.

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