Self-consistent model of unipolar transport in organic semiconductor diodes: accounting for a realistic density-of-states distribution

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A self-consistent, mean-field model of charge-carrier injection and unipolar transport in an organic semiconductor diode is developed utilizing the effective transport energy concept and taking into account a realistic density-of-states distribution as well as the presence of trap states in an organic material. The consequences resulting from the model are discussed exemplarily on the basis of an indium tin oxide/organic semiconductor/metallic conductor structure. A comparison of the theory to experimental data of a unipolar indium tin oxide/poly-3-hexyl-thiophene/Al device is presented.

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I. INTRODUCTION

During the last years the problem of an adequate description of charge injection and transport in organic semiconductors (OSCs) became of great importance in view of application of these materials as basic elements of electronic devices such as organic field-effect transistors, organic light-emitting diodes, or organic photovoltaic cells.

Organic systems are typically wide-bandgap disordered semiconductors possessing as a rule a relatively narrow intrinsic density-of-states (DOS) distribution without sharp band edges typically approximated by a Gaussian function. Transport in such systems is generally described by models based on hopping of charge carriers between localized states, which are disordered in space and energy. In addition to the intrinsic DOS distribution, disordered OSCs may exhibit deeper localized states originating from impurities, or from chemical and structural defects. Those states are normally located energetically well below the intrinsic DOS distribution and, therefore, they are referred to as deep traps. In general, the energy distributions of such trap states are often assumed to be also of Gaussian shape, however for some organic materials they are assumed to distribute exponentially.

To simplify the description of transport, a theoretical concept is often applied where charge transport is controlled by carrier jumps to the so-called effective transport energy level. This characteristic energy is localized within the intrinsic DOS-distribution and is defined as the energy of those sites a charge carrier visits with highest probability, independently of its energetic starting position. This concept allows one to reduce the description of hopping transport to a multiple trapping formalism. The transport energy plays then the role of the mobility edge in disordered materials and may be used to qualitatively separate mobile and immobile carriers in OSCs. In this concept, charge transport is provided by mobile carriers with a constant mobility where all temperature dependences in the transport description result from the Fermi distribution of carriers and from the temperature dependence of the transport energy. Another version of the transport energy approach utilizes the effective, average mobility of the whole ensemble of carriers. In this case, the mobility reveals, in general, dependences on the temperature and on the carrier density.

By now, the alternative concept of charge transport in OSCs has been also developed where all carriers in intrinsic DOS are considered mobile and their drift mobility is assumed to be dependent on the temperature, on the carrier density and, in general, on the electric field. Nevertheless, as regards the last dependence, it was noted in the literature that the field dependence of the carrier mobility is not strictly necessary, especially at high temperatures. The most advanced version of this approach, the so-called extended Gaussian disorder model (EGDM), has demonstrated good ability to simulate the current-voltage (I-V) characteristics of different OSC devices.

Another important process providing the functionality of organic devices is the injection of carriers from electrodes into the organic layer in view of a low intrinsic charge carrier density. However, the description of this process remains still controversial. For charge transport in bulk OSCs the electric field at the injecting interface is often taken equal to zero assuming space-charge limitation of the currents. Numerous papers assume finite values of the interfacial electric field and consider injection essentially as a single-particle process. Then injection is simulated as Fowler-Nordheim (FN) tunnelling through the surface energetic barrier or Richardson-Schottky (RS) thermionic emission over this barrier under an applied external field. In fact, the field and the charge carrier density at the interface are not known because these values depend on the height of the injection barrier which in turn is field-dependent. Thus, for a proper description of the charge carrier injection, a self-consistent determination of the field and the carrier density at the electrode/OSC interface is necessary which would contain both limits of weak (single-particle) and strong (many-particle) injection including the space-charge limited regime. An attempt of such a self-consistent approach was undertaken in Ref. where drift-diffusion and Poisson equations in the bulk were...
considered in conjunction with an injection described in the spirit of the RS model. However, application of the single-particle injection as a boundary condition for the many-particle equations makes this approach questionable. In view of disordered OSCs, sophisticated extensions of the RS model with account of possible injection into the tail states below the barrier were developed\textsuperscript{8,13,22,23,27,28,32} which, however, remain essentially single-particle injection models.

Another approach to charge carrier injection including Schottky barrier lowering and space-charge effects was recently developed in Ref\textsuperscript{12} where three-dimensional (3D) hopping of charge carriers on sites of a cubic lattice with randomly distributed energy levels was considered as well as a sophisticated one-dimensional (1D) continuous model. The site occupancies and the electric field were calculated self-consistently by solving the three-dimensional master equation and the Poisson equation in successive iterations with account of the field-dependent injection barriers. Yet, the effect of the individual image potential was thereby overestimated because of duplication with the mean field deep in the sample.

Recently, a self-consistent continuous description of injection in insulating media in terms of carrier densities and mean fields was developed\textsuperscript{14–37} based on the matching of the electric displacement and the electrochemical potential at the interface while still accounting for discreteness of injected charge carriers. The latter aspect is important for a wide range of values of injection barriers and applied voltages where the individual image force dominates the injection process. This 1D model exhibits a plausible crossover from the barrier-dominated behaviour at low voltages to the space-charge-dominated behaviour at high voltages and reveals a field-induced reduction of the injection barrier as well. The barrier lowering relates both to the Schottky effect and the voltage drop in the electrodes initiated by substantial interfacial charge carrier transfer. The model applies directly to inorganic crystal insulators and wide-bandgap non-degenerate semiconductors as well as to very narrow-band insulators and semiconductors as was indicated in Ref\textsuperscript{27}. It applies also to wide-bandgap OSCs if the narrow band approximation assuming a negligible width of the Gauss DOS is valid. The latter restriction, however, seems to fail in many organic semiconductors\textsuperscript{2,3,26} and excludes the possibility of injection into the tail states which proved to be essential for disordered semiconductors\textsuperscript{8,11,27,28,32}.

In the present paper the self-consistent approach of Refs\textsuperscript{24–35,37} is extended to account for a realistic DOS shape of the organic material. The charge transport in semiconductor/OSC/conductor diode structure is modeled using the transport energy concept in the spirit of Refs\textsuperscript{11,13} and focusing on the influence of the injection barrier heights and the DOS parameters on the \textit{I-V} characteristics. As an example, a system is studied where only holes are injected from an indium tin oxide (ITO) electrode into the OSC layer, whereas the possible injection of electrons from the metallic conductor electrode into the OSC as well as subsequent recombination effects are excluded at this stage, for simplicity. The obtained model will be compared to experiments of Ref\textsuperscript{14} where \textit{I-V} characteristics of poly-3-hexylthiophene (P3HT) based unipolar diodes were analyzed assuming space charge limited current boundary conditions at the electrodes.

\section{Theoretical Model}

Let us consider an OSC layer of thickness \( L \) sandwiched in between a heavily doped semiconductor and a metallic conductor electrode. The organic layer is supposed to be extended over the space with \(-L/2 < x < L/2\), whereas the semiconductor and conductor electrodes are extended over the half-spaces with \( x < -L/2 \) and \( x > L/2 \), respectively. ITO, being an electron-conducting semiconductor with a deep lying conduction band, is considered as the hole-injecting electrode\textsuperscript{38–39} while Al is considered as collecting electrode. The band structure of the system under consideration is shown schematically in Fig. 1.

The OSC is characterized by a DOS represented here as a superposition of two Gaussian DOSs (see, for example, Refs\textsuperscript{2,3,12}). The first, intrinsic DOS represents the highest occupied molecular level (HOMO band) and the second one describes the spatially and energetically distributed trap states (see Fig. 1). The total DOS distribution then can be written as:

\[ g(E) = \frac{P_c}{2\sigma_c^2} \exp\left(-\frac{E^2}{2\sigma_c^2}\right) + \frac{P_t}{\sqrt{2\pi\sigma_t^2}} \exp\left(-\frac{(E - E_t)^2}{2\sigma_t^2}\right), \]

where \( E_t \) denotes the average trap energy, \( \sigma_c \) and \( \sigma_t \) the widths of the intrinsic and trap DOS parts, \( P_c \) and \( P_t \) the

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig1.jpg}
\caption{Schematic band diagram of the considered semiconductor/OSC/conductor structure and the density-of-states distribution in its insulator constituent. The assumed hole transport under voltage application is also depicted.}
\end{figure}
numbers of intrinsic states and traps, respectively (notice that the level \( E = 0 \) coincides with the DOS maximum of the HOMO band).

Spatial distributions of the charge-carrier density \( p \) and of the electric field \( F_i \) in the OSC are described by the following system of equations:

\[
\begin{align*}
  kT \mu_i \frac{dp_i (x)}{dx} - e \mu_i p_i (x) F_i (x) &= -j, \\
  \frac{dF_i (x)}{dx} &= \frac{e}{\epsilon_i \epsilon_0} p (x),
\end{align*}
\]

where \( j \) is the position-independent steady-state current density, \( \mu_i \) denotes the hole mobility in the conductive states of the OSC, \( \epsilon_i \) its static relative permittivity, \( k \) the Boltzmann constant, \( e \) the positive elementary charge, \( \epsilon_0 \) the dielectric permittivity of vacuum, and \( T \) the absolute temperature. The total density of charge carriers \( p (x) = p_c (x) + p_t (x) \) is the sum of mobile and immobile carrier densities, which are defined as

\[
p_c (x) = \int_{-E_c}^{E_c} g (E) f_p (E) dE
\]

and

\[
p_t (x) = \int_{E_c}^{\infty} g (E) f_p (E) dE,
\]

with \( f_p (E) \) the Fermi-Dirac distribution of holes,

\[
f_p (E) = \left\{ \exp \left[ \frac{\kappa_i (x) + e \phi_i (x) - E}{kT} \right] + 1 \right\}^{-1}.
\]

Here, \( E_c \) denotes the effective energy of the transport level, \( \kappa_i \) the electrochemical potential for holes and \( \phi_i \) the electrostatic potential in the organic layer.

Assuming \( \kappa_i (x) + e \phi_i (x) - E_c \gg kT \), Boltzmann statistics for the mobile carriers can be applied and one obtains

\[
p_c (x) = N_c \exp \left[ -\kappa_i (x) - e \phi_i (x) \right],
\]

where

\[
N_c = \int_{-\infty}^{E_c} g (E) \exp \left( \frac{E}{kT} \right) dE
\]

\[
= \frac{P_c}{2} \exp \left[ \frac{\sigma^2_c}{2 (kT)^2} \right] \text{erfc} \left( \frac{E_c}{\sqrt{2} \sigma_c \sqrt{kT}} \right)
\]

\[
+ \frac{P_t}{2} \exp \left[ \frac{E_t}{kT} + \frac{\sigma^2_t}{2 (kT)^2} \right] \text{erfc} \left( \frac{E_t - E_c}{\sqrt{2} \sigma_t \sqrt{kT}} \right),
\]

Using Eq. (7) in combination with Eqs. (6) and (5), we can express the density \( p_t (x) \) through \( p_c (x) \) as follows

\[
p_t (x) = p_c (x) \int_{E_c}^{\infty} \frac{g (E) dE}{N_c \exp (-E/kT) + p_c (x)},
\]

and, correspondingly, the total carrier density \( p (x) \) as

\[
p (x) = p_c (x) + p_t (x) \int_{E_c}^{\infty} \frac{g (E) dE}{N_c \exp (-E/kT) + p_c (x)}.
\]

The latter equation together with Eqs. (2) and (3) presents a system of equations describing the stationary charge transport in the OSC where the appropriate boundary conditions have to be applied.

The energetic differences between the equilibrium values of chemical potential in the electrodes far away from the electrode/OSC interfaces and the maximum of the HOMO band in the organic layer are defined as the injection barriers \( \Delta^\pm \) for charge carriers (from now on, the minus and plus superscripts denote the quantities at the interfaces \( x = -L/2 \) and \( x = L/2 \), respectively). These barriers relate to the difference between the electrode work functions \( E^\pm_A \) (see Fig. 11):

\[
E^+_A + \Delta^- = E^-_A + \Delta^+.
\]

Assuming neither surface charge nor dipole layers at the electrode/OSC interfaces one can require continuity of the electrical displacement and of the electrochemical potential across the entire device. This continuity results in the following nonlinear boundary conditions:

\[
p_c \left( \pm \frac{L}{2} \right) = N_c \exp \left\{ -\frac{\Delta^\pm}{kT} + \frac{e l^\pm_{TF} \gamma_e}{kT} \left[ \frac{\epsilon_i}{\epsilon_e} F_1 \left( \pm \frac{L}{2} \right) - \frac{j}{\sqrt{\gamma_e}} \right] \right\}
\]

\[
+ \frac{e}{kT} \delta \phi^\pm_{Sch} \theta \left( 0.2 r^\pm_s - x^\pm_m \right),
\]

where \( \gamma^\pm_e \) denote the specific conductivities of the electrodes, \( \theta (z) \) the Heaviside unit step function, \( l^\pm_{TF} = \left[ p_e (\pm L/2) \right]^{-1/2} \) the characteristic distance between charge carriers near the respective electrode, and \( x^\pm_m = \{ e/16 \pi \epsilon_0 \epsilon_i \left[ \mp F_1 (\pm L/2) \right] \}^{1/2} \) the distance from the respective electrode to the maximum of the single-particle Schottky potential barrier. The Thomas-Fermi screening lengths in the electrodes are introduced as

\[
l^\pm_{TF} = \sqrt{\frac{2 \epsilon_0 \epsilon_i \epsilon_e \kappa^\pm_{TF}}{3 e^2 p^2_{\infty}}}.
\]

with \( \epsilon^\pm_e \) being the static relative permittivities of the electrodes, \( p^2_{\infty} \), and \( \kappa^\pm_{TF} \) the equilibrium values of the carrier density and of chemical potential in the electrodes far away from the electrode/OSC interfaces, the latter ones calculated with respect to the bottom of the respective electrode conduction band (Fig. 11). The last term in the exponent of Eq. (12) accounts for the discreteness of charge carriers and thus determines the range of the
injection barriers and field values where the individual image forces dominate the injection process resulting in the so-called Schottky lowering of injection barriers\(^{41}\),

\[
e\delta \phi_{S\text{ch}}^\pm = \sqrt{\frac{\varepsilon^2}{4\pi\varepsilon_0\varepsilon_i}} \left[ \mp F_i \left( \pm \frac{L}{2} \right) \right].
\] (14)

Details about single particle consideration in the mean field description provided here can be found in Ref.\(^{37}\). Notice that the contribution of the current \(j\) in Eq. (12) can often be neglected since it is very small in all practical cases concerning organic semiconductors\(^{34,35}\).

The nonlinear differential equations (2) and (3) with \(\rho(x)\) from Eq. (11) and with the boundary conditions (12) have to be solved numerically. Knowledge about the spatial distribution of the electric field gives access to the voltage drop \(V\) across the system for a given current density \(j\), which follows by direct integration of the field over the device thickness\(^{34,36}\) and reads:

\[
V = \int_{T_F}^{t_F} \left[ \frac{\varepsilon_i}{\varepsilon_c} F_i \left( \frac{L}{2} \right) - \frac{j}{\gamma_e} \right] \, dt + \int_{-L/2}^{L/2} F_i(x) \, dx - V_{bi},
\] (15)

where \(-V_{bi}\) is the voltage drop in the case of \(j = 0\), i.e., the built-in potential, given by the difference of the electrode’s work functions,

\[
e V_{bi} = E_A^- - E_A^+ = \Delta^- - \Delta^+.
\] (16)

Now, \(I-V\) characteristics of the structure under consideration can be calculated.

### III. COMPARISON WITH EXPERIMENT AND DISCUSSION

To test the presented model, it is applied to experimental \(I-V\) characteristics measured on a unipolar device consisting of a single P3HT layer of thickness \(L = 125\ \text{nm}\) sandwiched between ITO and Al electrodes. It was deduced from different experiments\(^{12,14,42}\) that the total DOS in P3HT consists of the superposition of two Gaussian peaks, similarly to Eq. (1).

Figure 2 depicts \(I-V\) characteristics of the ITO/P3HT/Al structure from Ref.\(^{12}\) measured at different temperatures as well as best fits calculated with the OSC model parameters indicated. All parameters of the electrodes as well as the typical value of the OSC relative permittivity, \(\varepsilon_i = 3\), are taken over from Ref.\(^{37}\). At first, fitting the \(T = 300\ \text{K}\) curve, we establish the best values for DOS parameters and carrier mobility. Next, by fitting of the curves for the other temperatures, these model parameters remain unchanged and only the barrier heights \(\Delta^\pm\) and the transport energy \(E_c\) are allowed to vary with temperature. One can see that the calculations reproduce satisfactorily the magnitude and the general form of the \(I-V\) characteristics in the wide range of applied voltages and at temperatures between 160 and 300 K with the fitting parameters indicated in the figure. Remarkable deviation of the theory from the experiment in the low-voltage parts of \(I-V\) characteristics (below \(-V_{bi}\)) is explained by a parallel leakage current which is not taken into account in the presented model.

The obtained fitting values of the transport energy are in a qualitative agreement with the temperature dependence of this value predicted in the literature (see, for example, Refs.\(^{41,42}\)). However, this energy is not the only parameter needed to be varied with temperature. The best fitting can be achieved only if we assume the barriers \(\Delta^\pm\) to change with temperature too. There are at least two reasons which could make such changes likely. One can see that the ITO/OSC barrier increases from 0.1 to 0.3 eV with decrease of the temperature from 300 to 160 K. Such a change corresponds qualitatively to the temperature dependence of the bandgap energy well-known in classical semiconductors\(^{41}\) and recently established in charge transfer complexes based on OSC thin films\(^{43,44}\) (it should be noted, however, that such an effect is not yet known in P3HT). On the other hand, the direction of the barrier change for the collecting electrode is different from that for the injecting one. It first slightly decreases and afterwards does not change with the temperature decrease, contrary to the increase of the injecting barrier. It may be supposed that the barrier heights are additionally affected by dipole layers which may emerge at the electrode/OSC interfaces, typically for these systems\(^{45,46}\). They can have an intrinsic, individual temperature behavior and thus may contribute to the temperature dependence of injection barriers.

It should be noted that the obtained fitting clearly emphasizes the essential role of the self-consistent boundary conditions in the proposed theoretical model. In
Ref. 22, experimental $I$-$V$ curves have been simulated within the basically similar mobility edge concept but using the temperature-independent transport energy $E_c = 0$ and the space-charge limited boundary conditions of $F_l (± L/2) = 0$. There, the simulation has revealed less satisfactory agreement between the calculated and experimental curves and unreasonably low number of intrinsic states of the order of $10^{18}$ cm$^{-3}$ in contrast to the number of $10^{21}$ cm$^{-3}$ obtained in our simulations.

In conclusion, a one-dimensional mean-field model has been presented which describes self-consistently the charge-carrier transport across a semiconductor/OSC/conductor structure accounting for effects of discreteness of injected carriers and a realistic DOS distribution in the organic layer. The energy distribution of DOS was modelled by the superposition of two Gaussian peaks for the HOMO-level and deep traps. In the framework of the transport energy concept, assuming temperature-dependent injection barriers, good qualitative and quantitative agreement between the experimental and simulated $I$-$V$ characteristics has been obtained in a wide temperature range. Compared with the popular EGDM model our approach has a comparable number of fitting parameters and gives similar fitting accuracy (cf., for example, with Refs. 5, 20, 23).

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