Injection currents in thin disordered organic films

V R Nikitenko, N A Sannikova, M N Strikhanov
National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), 115409, Moscow, Russia

E-mail: vladronik@yandex.ru

Abstract. Analytic model of barrier-limited injection of charge carriers from metal electrodes into organic film, which was introduced by Arkhipov and co-workers, is modified, considering effects of multiple image charges and injection from both electrodes. Limits of applicability of Arkhipov’s model are discussed. Variations from Arkhipov’s model are important, if film thickness is comparable with Onsager length.

1. Introduction
Injection and transport of charge carriers in organics have important peculiarities, caused by hopping nature of these processes [1]. Correct description of charge injection from metal electrodes is an important part of any predictive model of operation of organic light-emitting diodes (OLEDs) and other elements of organic electronics. Although real OLEDs include numerous layers of different organic materials, single-layer structures are useful for experimental characterization of individual layers. The simple analytic model [2] describes correctly field and temperature dependence of barrier-limited injection from electrode to organic material, accounting for energetic disorder and image-force potential. However, this model considers organic material as a semi-infinite media, while thickness of an active layer of OLEDs, \( L \), is typically less than 100 nm, hence it is comparable with Onsager length, \( r_c = \frac{e^2}{4\pi\varepsilon_0 k T} \), where \( e \) is elementary charge, \( \varepsilon_0 \) is electric constant, \( \varepsilon \) is relative permittivity, \( k \) is Boltzmann constant and \( T \) is absolute temperature. Indeed, \( r_c = 18.9 \) nm and 55.7 nm, providing that \( \varepsilon = 3 \) and \( T = 295 \) K and 100 K, respectively. Consequently, one can expect deviations from results of Arkhipov’s model in thin films and under low temperatures, i.e. under the condition \( L \leq r_c \). Indeed, effects of charge redistribution in both electrodes, caused by injected charge, may be important. In addition, contribution of backward current, caused by diffusion of charge carriers from opposite electrode, is not negligible at low voltages \( V, eV < kT \). In the present work the model of ref. [2] is modified, considering these effects.

2. The model
We consider an organic layer, being placed between identical contacts (electron-only or hole-only device), providing considerable energy barriers for injection. The model of ref. [2] considers injection of charge carriers as a two-step process. First step is phonon-assisted tunnelling jump (the rate is described by Miller-Abraham’s form) from the Fermi level of electrode to a localized state in organic material, in proximity to the surface. Second step is hopping diffusion in disordered system, biased by...
electric field, which results from Coulomb image-charge and applied voltage, $V$. Eventually, a carrier has to overcome a potential barrier, see the dashed line in the inset of the Figure 1. Interaction of an injected carrier with multiple image charges (except of the nearest one) results in additional term in a potential energy, which can be expressed in terms of polygamma function of zero order,

$$\psi(z) = -\gamma + \sum_{n=0}^{\infty} \left( \frac{1}{n+1} - \frac{1}{n+z} \right),$$

$$\delta U(y) = \left( e^2 / 16\pi \varepsilon_0 \varepsilon_L \right) \left[ 2\gamma + \psi(1+y) + \psi(1-y) \right],$$

where $y = x/L$ is a dimensionless coordinate ($y = 0$ at injecting electrode), and $\gamma \approx 0.57721$ is Euler–Mascheroni constant. The mean potential energy of the charge carrier (excluding energy disorder), $U_s(y)$, is a superposition of charge energy in an external electric field $F_0$, the Coulomb interaction energy with the nearest image charge and energy of interaction with the rest of image charges $\delta U(y)$, see eq. (1):

$$U_s(y) = \frac{e^2}{16\pi \varepsilon_0 \varepsilon_L} \frac{1}{y} + \delta U(y) + \Delta \mp eF_0 y,$$

where field $F_0$ is assumed to be homogeneous, $\Delta$ is barrier height at zero field. The signs "+" and "-" correspond to the field direction from the electrode and to the electrode, respectively.

The model takes into account the currents injected from both electrodes:

$$J_{\text{inf}} = j_+ - j_-,$$

where $j_+$ and $j_-$ are injection currents from the left (in the direction of the field, $j_+$) and the right (the opposite direction to the field, $j_-$) electrodes, respectively. A carrier can pass the layer in both directions due to diffusion, providing low applied voltage ($eV < kT$), hence it is necessary to consider both terms.

The rate of the first jumps, i.e. jumps from the Fermi level of the injecting electrode to a state with energy $E$, which is located at a distance $x_0$ from the electrode, is calculated according to the Miller-Abrahams model [3], $v = \nu_0 \exp(-2x_0/a_L) Bol(E)$, where $Bol(E) = \exp\left[-(E + |E|)/2kT\right]$, $\nu_0$ is attempt-to-hopping frequency, which is determined by the properties of the organic material and the injection electrode, $a_L$ is localization radius of wave functions. Fermi level of the injecting electrode is taken as zero point of energy.

Similar to Arkhipov's model, injection current from each electrode is an average of a rate of first jump, multiplied by the probability to overcome the potential barrier (see the solid line in the inset; this probability is calculated in the framework of one-dimensional version of Onsager’s model, as in the ref. [2], but with the term $\delta U$ in the energy $U_s$), over energies of hopping states and hopping distances $x$:

$$j_s = j_0 \int_{y_0}^{\infty} dy \exp\left(-2L/a_L y_0\right) \int_{y_0}^{\infty} dE Bol(E) g(E-U_s(y)) \int_{a/L}^{1-a/L} dy \exp\left[U_s(y)/kT\right]$$

$$\int_{a/L}^{1-a/L} dy \exp\left[U_s(y)/kT\right],$$

where $j_0 = eN_i \nu_0 L$, $N_i$ - concentration of localized states, $a$ - distance from the electrode to the nearest state hopping, $y = x/L$, $y_0 = x_0/L$, and energy distribution of localized states $g(E)$ is a Gaussian function:
\[ g(E) = \frac{1}{\sqrt{2\pi\sigma}} \exp\left( -\frac{E^2}{2\sigma^2} \right), \]  

(6)

At variance with original Arkhipov’s model, the present model accounts for the finite size of a layer, see limits of integration in eq. (5), and contribution of injection from opposite electrode, see eq. (4), which occurs against applied electric force, due to diffusion. Since the field is considered to be homogeneous, the voltage is connected with field strength as follows: \( V = F_0 L \).

3. Results and conclusions

Figure 1 shows I-V characteristics of thin organic layers, calculated at the following parameters: injection barrier is \( \Delta = 0.5 \text{ eV} \), variance of Gaussian distribution of localized states is \( \sigma = 0.1 \text{ eV} \), concentration of localized states \( N = 10^{27} \text{ m}^{-3} \), hopping frequency factor \( \nu_0 = 1.4 \times 10^{12} \text{ s}^{-1} \), length of a first jump \( a = 0.5 \times 10^{-9} \text{ m} \), \( \varepsilon = 3 \), \( T = 100 \text{ K} \).

![Figure 1. I-V characteristics, parametric in thickness of the layer L, see the figure. Solid lines shows results of the modified model, dashed lines – Arkhipov’s model (\( \delta U = 0 \)) with account of backward current and dash-dotted lines – original Arkhipov’s model. Inset shows the total potential energy (solid line), and the same in Arkhipov’s model (\( \delta U = 0 \), dashed line), the voltage is 0.01 V.](image)

Figure 2 shows I-V characteristics for the case of different temperatures, calculated at the following values of parameters: \( \Delta = 0.6 \text{ eV} \), \( \sigma = 0.1 \text{ eV} \) and \( L = 20 \text{ nm} \).

The results shown in Figures 1 and 2 are in good agreement with the assumption that the effect of backward (diffusion) current becomes significant under the condition \( eV < kT \) (see dashed and dot-dash curves, considering the criterion of difference of 2 or more times). Dashed lines in Figures 1 and 2 shows results of eqs. (4) and (5), neglecting contribution of multiple image charges (\( \delta U = 0 \)). Original model of the ref. [2] results from eqs. (4) and (5), if, in addition, \( L \to \infty \), hence \( j \to 0 \) (dash-dotted lines).

In thin films and at low voltages and temperatures effect of multiple image charges yields large increase of current at a given voltage relative to predictions of Arkhipov’s model [2], due to reduction
of a potential barrier, see inset in the Figure 1, in spite of negative contribution of backward current, except of high temperatures, see Figure 2. Deviation from predictions of the ref. [2] can exceed the factor 2, if $L < \frac{r_c}{2}$.

![Figure 2. I-V characteristics, values of temperature, K: 80, 100, 200, 300, increases in the direction of arrow. Solid lines shows results of the modified model, dashed lines – Arkhipov’s model ($\delta U = 0$) with account of backward current and dash-dotted lines – original Arkhipov’s model.](image)

The proposed analytic model, being relatively simple, is useful for testing and correct choice of parameters of numerical models in predictive modelling of OLEDs, photovoltaic elements and other devices of organic electronics.

This work was supported by Russian Ministry of Education and Science in the frames of Competitiveness Growth Program of National Research Nuclear University MEPhI, Agreement 02.A03.21.0005

**References**

[1] Bässler H 1993 *Phys. Stat. Sol. (b)*. 175 15

[2] Arkhipov V I, Emelianova E V, Tak Y-H, Bässler H 1998 *J. Appl. Phys.*. 84 848-856

[3] Miller A, Abrahams E 1960 *Phys. Rev.*. 120 745