Monte-Carlo modeling of drift mobility in thin organic layers: lattice type influence

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Abstract. Hopping transport of charge carriers in thin (up to 100 nm) organic layers, which are suitable for organic light-emitting diodes and solar cells, is modeled in the framework of Gaussian disorder model. Drift mobility dependence within ranges of layer thickness, energy disorder and field strength is considered. Monte-Carlo simulations for low concentration case show weak dependence on type of lattice – simple cubic, body-centered and face-centered lattice. Results of this work are compared with the results obtained recently by Cottaar et al., done within master equation approach. The comparison shows the deviation at low disorder. Additionally we take into account influence of barrier-limited injection in case of cubic lattice.

1. Introduction
Organic electronics devices consist of thin active layers. Predictive modeling of operation of such devices unavoidably needs correct characterization of charge transport in thin organic layers. Charge transport, being of hopping nature, can be described in the framework of Gaussian disorder model [1]. Values of mobility, which have been obtained from experiments for thicker layers (> 1 micrometer), are often used for modeling the light-emitting diodes. The recent review [2] combined all the results that had been published in this scientific area for the last few decades. As it was shown, lattice type of an organic volume does not have big influence on dynamic characteristics of a particle. It can be explained in terms of percolation theory [3]: correlation radius $R$ is much bigger than lattice constant $a$ of a cell, so there is no difference what type of a lattice is. However, all main computing results, dedicated to dependence on lattice type, were obtained within master equation approach to electron transport with periodic boundary conditions [4]. So it would be of interest to check numerically, whether mobility in thin films depend on lattice type, or not, using complete Monte-Carlo (MC) modeling, which is superior respective to master equation approach.

2. Monte-Carlo procedure and scheme of modeling
In our previous work [5] we took into account the scenario of MC simulations without the influence of the electrode (dummy injecting electrode), in order to model properties of a thin organic layer itself, irrespective to conditions of injection. Here we use a scenario, where an event starts with an electron being born in the first layer and this scenario allows the particle to be reflected from left boundary, in order to compare results with more realistic case of energetic barrier-limited injection, with barrier being reduced by image-charge Coulomb potential (thermionic injection). A carrier, being born in the 1-th layer, walks through the whole volume until it reaches the last layer. Influence of image charge was obtained for simple cubic (SC) lattice only, but it is enough to estimate main dependences of the phenomenon. We consider not only SC, but two more options: body-centered (BC) and face-centered (FC) lattice (see Fig. 1, a-c).

Injection of a charge carrier from the electrode into the layer of organic material should be presented as a drift and diffusion via manifold of localized states, [6] and the potential energy of a
carrier has to be given in the form: 
\[ U(x) = H - e^2/16\pi\varepsilon_0 x - eF_0 x, \]
where \( H \) is the height of the barrier (see Fig.2), which is equal to 0.7 eV (this barrier is reduced by one-dimensional Coulomb interaction with image charge), and \( \varepsilon \) in our calculations is equal to 3 [6].

![Fig.1](image1.png)

**Fig.1** Lattice types involved in MC modeling. Fig. 1, a-c, represents cubic, face-centered and body-centered lattice types, respectively. Fig. 1, d shows a feature of MC modeling for being able to model different lattice types: each standard cubic cell is subdivided into eight elementary cells with a lattice constant \( a' \).

![Fig.2](image2.png)

**Fig.2** Scheme of electron injection in the presence of an energy barrier of height \( H \).

A conventional model is used, where hopping centers are located at sites of a lattice. Number of sites along any transversal direction exceeds 10 times a number of sites along the electric field direction. Energies of sites are distributed randomly according to Gaussian distribution
\[ g(E) = \left( \frac{1}{\sqrt{2\pi\sigma^2}} \right) \exp \left[ -E^2/2\sigma^2 \right] \]
with the variance \( \sigma \). If an electron appears in the nearest to right electrode layer, event is over. Drift mobility is calculated from the average drift velocity, \( \mu = L/F \langle 1/t_\mu \rangle \), where \( L \) – length of the volume, \( F \) – strength of the field applied, \( t_\mu \) – transition time. It is also important to notice, that hopping rates that we use in our calculations, are determined by usual Miller-Abraham’s expression: 
\[ v_{ij} = \alpha_0 \exp \left[ -2\gamma r - \left( \Delta E_{ij} + |\Delta E_{ij}| \right)/2kT \right], \]
where \( \alpha_0 \) is frequency factor, \( \gamma \) is inverse radius of localization, \( r \) is a distance between two sites, \( \Delta E_{ij} = E_i - E_j - eF_0 \cos \theta \), \( T \) is the absolute temperature, \( k_B \) is Boltzmann’s constant and \( \theta \) is the angle of directions of electric field \( F \) and jump \( r \). The value \( 2\gamma a = 10 \) is assumed. The main difference between modeling SC and, for example, FC lattice is that for face-centered lattice option we have additional sites that an electron can visit. To make it simple from the programming point of view, one should subdivide an original cell into eight elementary cells with a constant lattice \( a' \), which is twice smaller than the original one, \( a \), and allow visiting only those sites, that provide a cell of concrete shape needed (see Fig. 1, d). Although this procedure is simple, but it takes much more time to get required statistics because of considering more sites for hopping.
The analytic expression for drift mobility has the form [7]:

\[
\mu(T) = B \frac{e \omega_h}{N_0^{\beta_0}} \sigma^{\lambda-2} \exp \left( -\frac{1}{2} \sigma^{-2} - \frac{E_{\text{crit}}}{k_B T} \right),
\]

in case of low relative concentration of charge carriers, that is true in MC-modelling,

\[
c = \frac{n}{N_i} \equiv \left( \frac{1}{2} \right) \exp \left[ -\left( \frac{\sigma}{k_B T} \right)^2 / 2 \right],
\]

where \(B\) is the numerical prefactor, \(e\) – the electron charge, \(\lambda\) – the exponent, \(\sigma \equiv \sigma/k_B T\), \(E_{\text{crit}}\) – the critical (transport) energy, \(N_i\) – the site density, \(\omega_h\) is the frequency factor. In [6] authors use both Miller-Abrahams and Marcus hopping rates [8], [8]. Parameters \(\lambda\) and \(E_{\text{crit}}\) are claimed to be influenced by lattice type. It has to emphasize, that in [7] Eq. (1) is taken to fit obtained mobility data, that are calculated for low field strength and relative carrier concentration, \(c\), being in range of \(\sim 10^{-5} - 10^{-1}\), see also [4]. Numerical values of \(B\), \(\lambda\) and \(E_{\text{crit}}\), that depends on \(\sigma\), are given in [7] for different lattice types (except BC lattice, that is not considered there).

3. Results and discussion

The results in [7] were obtained using periodic boundary conditions and master equation approach, modeling the infinite media with applied small electric field for SC and FC lattice types. The values of \(\sigma\) in [7] are considered in the whole range of disorder, which is technically gives us the opportunity to compare our results for SC and FC with [7] for \(L = 100 \text{ nm}\) and \(F = 10^7 \text{ V/m}\), but one has to remember what conditions Eq. (1) was obtained under, plus authors in [7] do not calculate mobility in low disorder. The brief comparison is summarized in Table 1, where values of the mobility, calculated according to Eq. (1) with all the variables used for MC modeling, are divided by MC results themselves.

| \(F = 10^7 \text{ V/m}, L = 100 \text{ nm}\) | \(\sigma = 0.1 kT\) | \(\sigma = 3 kT\) | \(\sigma = 4 kT\) |
|---|---|---|---|
| Eq. (1) / MC, SC | 4.766 | 0.516 | 0.189 |
| Eq. (1) / MC, FC | 7.963 | 1.339 | 0.651 |

As one can see, for low disorder the relative comparison shows the biggest deviation. One has to note, that in [7] comparison of analytic (1) and numerical results is absent in case of low disorder. Fig. 3 \(a,b\) shows that both MC and analytic results have only slight dependence on a lattice type, in accord with previous suggestions [3]. However, the difference increases along with the increase of disorder (it reaches 50\% at \(\sigma/k_B T = 4\), see Fig. 1b), so one has to take it into account in quantitative modeling. MC results exceed mobility from [7] at high disorder due to finite-size effect on mobility in thin films [5]. Deviations in low-disorder limit result from absence of applicability of the analytic approach [7]. One has to expect that with field strength increasing all the deviations will be negligible quantities. The truth of this statement is demonstrated on Fig.3,4, where three lattice types are compared with each other in strong field modes, \(2.5 \times 10^7\) and \(5 \times 10^7\) \(\text{V/m}\). For the case of barrier-limited injection, see Fig. 5, it is shown, that this realistic effect smoothens slightly the thickness dependence of drift mobility, which has been reported in [5], but deviations are not principal (values of mobility being slightly reduced).
Fig. 3 Comparison MC results with [7] for two different cases: a) SC and b) FC and BC lattice. MC results were obtained for field strength $10^7 \text{ V/m}$, $\sigma/kT = 0.1, 3, 4$ and $c_1 = c_2$.

Fig. 4 Comparison MC results for different lattice types for two different cases: a) for field strength $2.5 \times 10^7 \text{ V/m}$ and b) for field strength $5 \times 10^7 \text{ V/m}$; $\sigma/kT = 0.1, 3, 4$ and $c_1 = c_2$.

Fig. 5 Comparison MC results for SC lattice type, taking into account Coulomb interaction, for two different cases: a) for low disorder $\sigma/kT = 0.1$ and b) for high disorder $\sigma/kT = 4; F = 1, 2.5, 5 \times 10^7 \text{ V/m}$ and $c_1 = c_2$. 
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References

[1] Bassler H Charge Transport in Disordered Organic Photoconductors a Monte Carlo Simulation Study 1993 Phys. Status Solidi B 175 15

[2] Baranovskii S D Theoretical description of charge transport in disordered organic semiconductors 2014 Phys. Status Solidi B 251 487–525

[3] Shklovskii B I and Efros A L Electronic Properties of Doped Semiconductors 1984 Springer Berlin

[4] Cottaar J Coehoorn R and Bobbert P A Scaling theory for percolative charge transport in molecular semiconductors: Correlated versus uncorrelated energetic disorder 2012 Phys. Rev. B 85 245205

[5] Nikitenko V R Sukharev V M and Sannikova N A Drift mobility in thin organic layers: joint Monte-Carlo and analytic modeling 2014 J. Phys.: Conf. Ser. 541 012028

[6] Arkhipov V I Wolf U and Bässler H Current injection from a metal to a disordered hopping system. II. Comparison between analytic theory and simulation 1999 Phys. Rev. B 59 7514

[7] Cottaar J Koster L J A Coehoorn R and Bobbert P A Scaling theory for percolative charge transport in disordered molecular semiconductors 2011 Phys. Rev. Lett. 107 136601

[8] Miller A and Abrahams E Impurity conduction at low concentrations 1960 Phys. Rev. 120 745

[9] Marcus R A Electron transfer reactions in chemistry. Theory and experiment 1993 Rev. Mod. Phys. 65 599

[10] Zvyagin I P A Percolation Approach to the Temperature and Charge Carrier Concentration Dependence of the Hopping Conductivity in Organic Materials 2008 Phys. Stat. Sol. 5 725

[11] Coehoorn R Pasveer W F Bobbert P A and Michels M A J Charge-carrier concentration dependence of the hopping mobility in organic materials with Gaussian disorder 2005 Phys. Rev. B 72 155206.