

The effect of spatial disorder on the temperature dependence of charge carrier mobility in disordered organics

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Abstract. The effective transport level is defined from the Monte-Carlo modelling in energetically and spatially disordered system, and the temperature dependence of the low-concentration and low-field mobility is calculated. The spatial disorder leads to the slight decreasing of the coefficient C in the temperature dependence, the same as decreasing of localization.

1. Introduction

Organic semiconductors, the conjugated polymers predominantly, are considered as perspective materials for spintronics [1]. Charge transport, characterized by the mobility of charge carriers (electrons and holes), is one of the key physical processes in organic spintronic devices, and also in organic light-emitting diodes [2], photovoltaic devices, field-effect transistors, etc. Transport of charge carriers and excitations in disordered organics occurs by means of uncorrelated phonon-assisted tunneling jumps of a carrier in the manifold of the localized states – the hopping sites (the hopping transport). It is the concept of the Gaussian Disorder Model (GDM) [3]. One has to note that the basic results of this model were obtained originally from the Monte-Carlo (MC) simulations [3]. However, one can obtain these results by the analytic methods [4], like transport level concept (TLC) [5-10], percolation theory [4,6,11], the average hopping parameter [12], etc. These methods are very suitable for the analysis of the mobility dependence on numerous parameters, namely the temperature, electric field strength, concentration of charge carriers and hopping sites. The energetic disorder, caused by the spatial disorder in the molecular structure of a given material, is the principal factor for the temperature dependence of the mobility [3]. Due to this reason, the Monte-Carlo (MC) modeling and other numerical methods often employ ordered structures of the hopping sites (for example, the simple cubic lattice) [9,11]. However, it is shown by MC modeling that the spatial disorder itself influences strongly the field dependence of mobility [3, 4]. In this work, we study the effect of a spatial disorder on the temperature dependence of mobility. We apply the modified method of the work [9] in order to find the transport level by MC modeling, and find the temperature dependence of mobility by the use of the transport level concept [8,9], comparing the cases of a pure energetic disorder (E-disorder) and the combined energetic and spatial disorder (E-r disorder).

2. The method of simulations

We use a MC simulation model similar to works [9,13]. The model is based on the well-known Gaussian disorder model [3]. Energies of the hopping sites are distributed as a Gaussian function with



the variance σ . The Miller-Abrahams (MA) model is used to describe the hopping rates ν_{ij} from a state i to the state j :

$$\nu_{ij} = \nu_0 \exp\left(-2\gamma r - \left(\frac{\Delta\epsilon_{ij} + |\Delta\epsilon_{ij}|}{2kT}\right)\right), \quad (1)$$

where r is the distance between the states i and j , γ is the inverse localization radius of the wave function, ν_0 is the frequency factor, T is the absolute temperature, k is the Boltzmann's constant, $\Delta\epsilon_{ij} = E_j - E_i$.

The escape of a carrier from a rather deep initial state is a multi-step process, because the first jump most probably will be followed by return to the initial state, hence one has to model the random walk of a carrier in vicinity of the initial state [9] (the case of a rather small electric field is considered). The condition of an electron's escape from the initial state of a given energy E is the escape from the spherical volume of a certain radius around this state. This radius is set so that the mean number of states of the energy deeper than E inside this volume is less than unity [9]. The escape time is the time until the last jump from the initial state, after which a carrier does not return to this state.

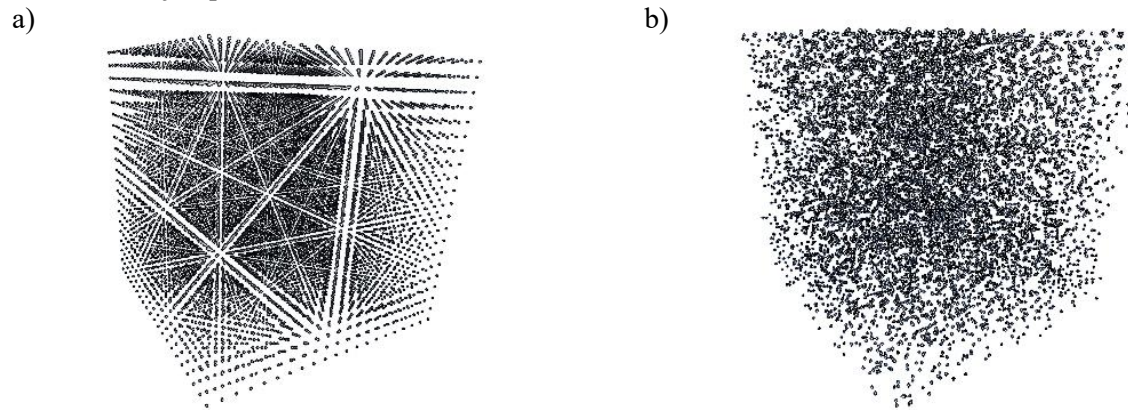


Figure 1. Visualization of the material structure: a) the hopping centers form the cubic lattice and b) randomly distributed in space (the minimal distance is $d = 0.15$ nm). The lattice constant is 1 nm.

Two models of the geometric distribution of localized states are considered. In the first model, there is no spatial disorder, the same as in the ref. [9] (Fig. 1a), i.e. the hopping centers are located at the nodes of the simple cubic lattice with the lattice constant a (1 nm in this work). Spatial disorder is present in the second model (Fig. 1b), so that localized states are randomly distributed in space. The mean concentration of these states is the same as in the case of an ordered lattice. The parameter of this model is the minimal inter-site distance d (0.15nm and 0.03nm in this work).

3. Results and discussion

Energy dependence of the escape time obeys the exponential law, $t_{esc} = \omega_0^{-1} \exp((E_c - E)/kT)$ both for the case of E- and E-r disorder, see the figure 2, $\omega_0 = \nu_0 \exp(-2\gamma a)$, hence one can define the effective transport level E_c from the condition $t_{esc} = \omega_0^{-1}$, see the figure 2b. Anyway, the spatial disorder accelerates the charge escape from a deep state, this effect is more pronounced for the case of the smaller localization parameter $2\gamma a$, and for the smaller energy disorder parameter σ/kT . Variation of the minimal hopping distance d , gives negligible effect on the escape time, see figure 2a.

Dependencies $E_c(\sigma/kT)$ are shown in figures 3a and 3b in units of kT and $\sqrt{2}\sigma$, respectively. The ratio E_c/kT approaches to zero at vanishing disorder, while the ratio E_c/kT becomes constant, if the parameter σ/kT is rather large. This constant value increases with increasing of the parameter $2\gamma a$ and approaches to the value 0.35, which was obtained in the ref. [11] from the percolation theory for the case of nearest-neighbor hopping, $2\gamma a = 20$.

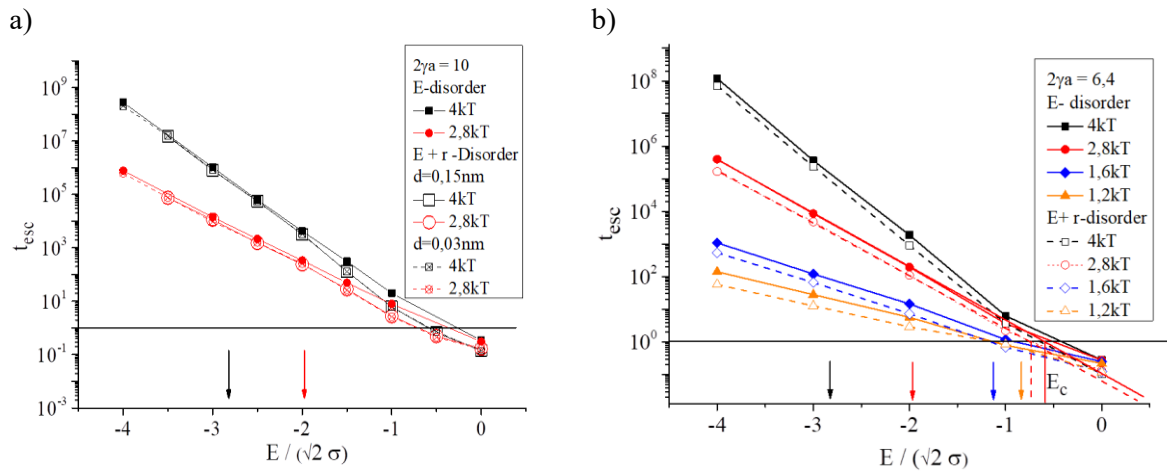


Figure 2. Escape time from the site of the energy E (in units of ω_0^{-1}) at various parameters of energetic and spatial disorder. The values of σ (in units of kT) are shown in the figures. $T=290$ K, $2\gamma a=10$ (a) and $2\gamma a=6.4$ (b). Arrows mark the energies of the energy distribution of carriers, $-\sigma^2/kT$.

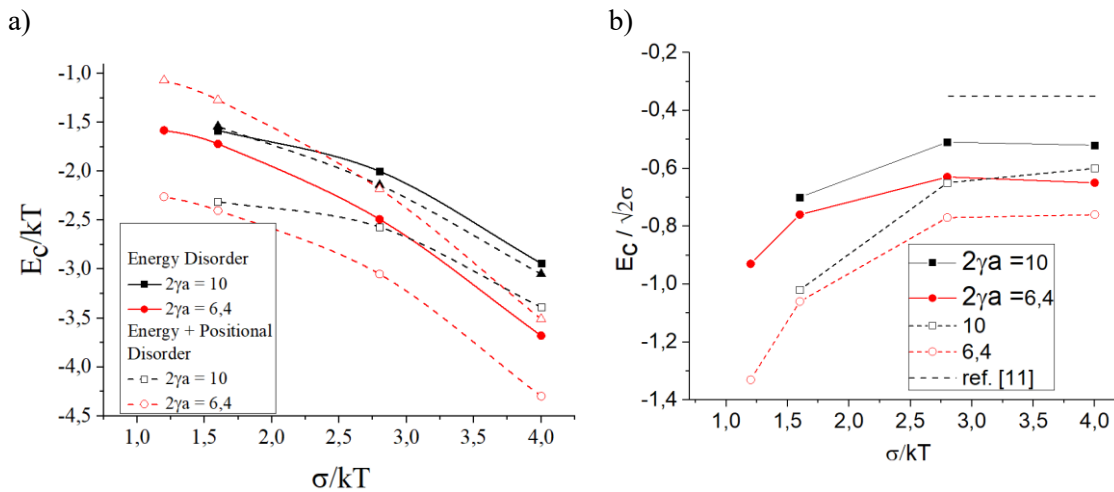


Figure 3. The effective transport level, E_c , in units of kT (a) and $\sqrt{2}\sigma$ (b). Filled ($2\gamma a=10$) and open ($2\gamma a=6.4$) triangles in the fig. (a) show the E_c from the analytic model of the ref. [9], providing the percolation factor $B=2.8$.

The transport level reduces both due to spatial disorder and due to the reduction of the localization parameter, $2\gamma a$. Comparing the dependencies $E_c(\sigma/kT)$, obtained from the MC simulation, and from the analytic model of the ref. [9], see triangles in the figure 3a, one can conclude that, counter intuition, the agreement is better for the case of E-disorder, than for the case of E-r disorder. One has to note that the escape time is defined in this work as the median time of the distribution, obtained from the set of trials, while it was the average escape time in the ref. [9]. The latter approach leads to the overestimation of the t_{esc} due to large contribution of rare events of very numerous round trip jumps. The constant value of the percolation factor [9] $B=2.8$ yields excellent agreement with MC data, if $2\gamma a=10$, while the temperature-dependent factor $(\sigma/kT - 1.5)$, proposed in the ref. [9], works better, if localization parameter decreasing, $2\gamma a=6.4$. However, the value $B=2.8$ provide qualitative agreement also in the latter case; the agreement gets better for the stronger disorder.

In order to study the temperature dependence of mobility, we use the founded values of E_c in the equation for the mobility [10]

$$\mu = \mu_c \frac{1}{2} \operatorname{erfc} \left(\frac{E_c}{\sqrt{2}\sigma} + \frac{\sigma}{\sqrt{2}kT} \right), \quad (2)$$

where $\mu_c \approx (e/kT)\omega_0 a^2$, and erfc is the complementary error function. If the energy disorder is rather strong, $\sigma/kT > 1.5$, eq. (2) yields the ubiquitous [4-11] law

$$\mu \approx \mu_0 \exp \left(-0.5(\sigma/kT)^2 - E_c/kT \right) \approx \mu_0 \exp \left(-C(\sigma/kT)^2 \right), \quad (3)$$

where μ_0 is the high-temperature limit of mobility. We found the coefficient C from eqs (2) and (3) at various values of σ/kT , giving the small variation to the temperature $T=290$ K and assuming E_c to be constant. We obtain for the case $2\gamma a=10$: $C=0.44$ (0.43), 0.44 (0.41), 0.39 (0.29) at $\sigma/kT = 4, 2.8, 1.6$, respectively (results for E-r disorder are in the brackets); $2\gamma a=6.4$: $C=0.42$ (0.41), 0.41 (0.38), 0.36 (0.27), respectively. Anyway, $C \approx \text{const}$, if $\sigma/kT > 1.6$, hence the law (3) is fulfilled, while deviations occurs at $\sigma/kT < 2$. These low-disorder deviations seem to be more pronounced for the case of E-r disorder. The value $C=0.44$ was obtained previously for the case of $2\gamma a=10$ by various methods [3,4], and this value is often considered as universal. However, various analytic methods [4-6,12,13] yield the slight decreasing of C along with $2\gamma a$, in qualitative agreement with our results. The same effect results from introducing the spatial disorder to the system. It is not surprising, because both the decreasing of $2\gamma a$ and introducing the spatial disorder leads to the prevailing of variable-range hopping relative to the nearest-neighbor hopping. Although the effect of E-r disorder seems to be negligible, if the energy disorder is rather strong, the mean values of C within an interval of moderate disorder can decrease by several hundredths, i.e. observably.

4. Conclusions

Monte-Carlo modeling of the electron's escape from rather deep states in the manifold of energetically and spatially disordered hopping sites confirm the applicability of the effective transport level concept, as it was done previously for the case of a pure energetic disorder [9]. Calculation of the temperature dependence of the low-concentration and low-field mobility show, that the effect of spatial disorder qualitatively is the same, as the effect of reduction of localization parameter, i.e. the slight decreasing of the coefficient C in the dependence of $\ln(\mu)$ vs $(\sigma/kT)^2$.

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