

Particle-Energy Distribution and Effective Temperature for the Hopping Transport in One-dimensional Disordered System

Tutiš, Eduard; Jurić, Ivan; Batistić, Ivo

Source / Izvornik: **Croatica Chemica Acta, 2010, 83, 87 - 94**

Journal article, Published version

Rad u časopisu, Objavljena verzija rada (izdavačev PDF)

Permanent link / Trajna poveznica: <https://urn.nsk.hr/urn:nbn:hr:217:539584>

Rights / Prava: [Attribution 4.0 International](#)/[Imenovanje 4.0 međunarodna](#)

Download date / Datum preuzimanja: **2023-09-26**



Repository / Repozitorij:

[Repository of the Faculty of Science - University of Zagreb](#)



Particle-Energy Distribution and Effective Temperature for the Hopping Transport in One-dimensional Disordered System*

Eduard Tutiš,^{a,**} Ivan Jurić,^a and Ivo Batistić^b

^aLaboratory for the Physics of Transport Phenomena, Institute of Physics, Bijenička c. 46,
 P. O. Box 304, HR-10001 Zagreb, Croatia

^bDepartment of Physics, Faculty of Science, University of Zagreb, Bijenička c. 32, HR-10000 Zagreb, Croatia

RECEIVED JUNE 1, 2009; REVISED JANUARY 5, 2010; ACCEPTED JANUARY 12, 2010

Abstract. Recent numerical simulations of the electronic hopping conduction in disordered organics have indicated that the effective temperature may be suitable to describe the ensemble of electrons in the electric field. However, the reasons for the emergence of the effective temperature have not been clarified, and only phenomenological expressions for the effective temperature have been proposed based on numerical simulations. We address these questions here for an analytically tractable one-dimensional model with uncorrelated Gaussian disorder. The exact relation for the distributions of particles in energy is obtained, the analytical expression for the effective temperature is derived, and the limits of the applicability of the concept of the effective temperature are drawn.

Keywords: energy disorder; hopping charge transport; non-equilibrium state; electronic ensemble; organic disordered materials

INTRODUCTION

Last several decades have witnessed the birth and the fast expansion of the field of organic electronics.^{1,2} The related devices are based on amorphous materials consisting either of small organic molecules or conjugated polymers. The development of these devices is paralleled by the fundamental research on the electronic processes in amorphous organics, with much effort invested into the understanding of the mechanism of the electric conduction.^{3–8} In general, the mobility of electrons in the amorphous organic materials is orders of magnitude lower than in related crystals. It is also rather strongly dependent on the electric field, which is usually in the range of one megavolt per centimetre. It is established that the electric conduction in disordered organics is realized by short-range hopping of electrons between molecular polaronic states, with each molecule acting as a trap (in the case of conjugated polymers this role is taken by the conjugated segments).^{6,9–11} The relevant parameters place the polaronic transport into the adiabatic regime. The amorphousness of the media is experienced by the electrons as a disorder in the polaronic energy levels and the hopping probabilities. In the past, the effect of this disorder on the electronic transport was mostly studied through numerical simulations, with

Monte Carlo and master equation methods being most frequently used.^{3,4,12–14} Important efforts were also made to address the qualitative picture beyond the numerical results.¹⁵ The most obvious question is one of the properties of the ensemble of carriers in the current-carrying state. Several recent papers suggest that the concept of an effective temperature may be applicable, with the ensemble in an electric field being equivalent to the ensemble in equilibrium, but at an elevated temperature.^{16,17} The very concept also proved useful in the previous studies of the conduction in doped inorganic semiconductors at low temperatures.^{18–21}

In the present communication we scrutinize the hypothesis of the effective temperature through analytical and numerical solutions of the hopping transport in one-dimension. An exact expression is derived for the distribution of particles in energy for the system far from equilibrium. From this distribution a formula for the effective temperature in the current-carrying case is derived. We also discuss the region of the applicability of the concept of the effective-temperature in the parameter space determined by the strength of the electric field and the disorder.

The paper is structured as follows. The model and the analytic solution for the particle distribution in the current-carrying state are introduced in section 2.

* Presented at the EU Workshop "Frontiers in Complex Metallic Alloys", Zagreb, October 2008.

Dedicated to Professor Boran Leontić on the occasion of his 80th birthday.

** Author to whom correspondence should be addressed. (E-mail: edo@ifs.hr)

In section 3. we present the numerical results for the distribution of particles in energy. In section 4. we derive the exact formulae for this distribution and for the effective temperature.

It should be emphasized that the present paper deals with the effective temperature that rises from and is confined to the shape of the particle-energy distribution of the ensemble. At present we do not pursue further practical usage of this parameter for calculating other properties of the system. Nor we investigate or compare the alternative definitions of the effective temperature. Such questions, although interesting, are left for the future.

MODEL

We consider the hopping conduction in one dimension, between localized states of energy ε_i (i is the index that counts the sites), separated by a distance a . The model is illustrated in Figure 1. The current flows from one side of an infinite system to another when the external electric field F is applied. The hopping probability from site i to site j is denoted by $W_{i,j}$ and is generally of the form

$$W_{i,j} = \Omega_{i,j} \exp\left(\frac{E_i - E_j}{2T}\right), \quad (1)$$

where E_i, E_j are the site energy levels shifted by the applied electric field, $E_i = \varepsilon_i - Fqai$, with q denoting the unit charge. T denotes the ambient temperature in energy units (*i.e.* the Boltzmann constant equals to 1). The frequency $\Omega_{i,j}$ collects all the remaining factors that differ from one hopping law to another. Generally, $\Omega_{i,j}$ has to obey the symmetry relation $\Omega_{i,j} = \Omega_{j,i}$ in order to meet the micro-balance requirement for the electron transfer between sites. We consider the model with the nearest-neighbour hopping which dominates in disordered organics at room temperature.¹¹ Then specifically, $\Omega_{i,i+1} = \Omega_0$ for the "symmetric hopping law",^{6,7}

$\Omega_{i,i+1} = \Omega_0 \exp\left(-\frac{|E_i - E_{i+1}|}{2T}\right)$; for the Miller-Abrahams phonon-assisted hopping^{22,3} and

$$\Omega_{i,i+1} = \Omega_0 \exp\left(-\frac{(E_i - E_{i+1})^2}{8\varepsilon_b T}\right), \quad (2)$$

for the small-polaron hopping,^{23,24} where ε_b is the polaron binding energy. In all of the cases the frequency Ω_0 is independent of site energies, containing the combination of parameters such as the electron-phonon coupling, the electron-wave-function overlaps *etc.* In most aspects the conclusions about the effect of disorder on the conduction do not depend on the type of the

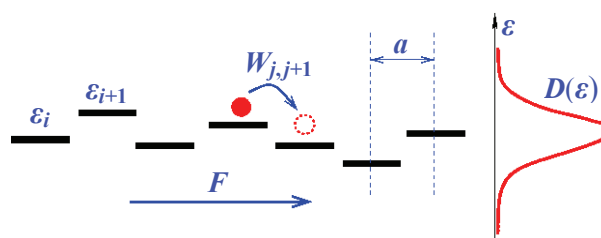


Figure 1. The schematics of the one-dimensional model for the hopping conduction in a molecular material with Gaussian disorder in the energy levels ε_i . The distribution of levels in energy is given by the density of states, $D(\varepsilon)$. The levels are not correlated in space. The net electric current arises as the external electric field F introduces a preferred direction for hopping.

hopping considered.^{7,11} In the calculations presented in this paper we opt for the "symmetric" version with $\Omega_{i,i+1} = \Omega_0$.

Disordered organics are known to have a Gaussian distribution of the molecular energy levels (*i.e.* a Gaussian density of states, D.O.S.),³ with the probability of the energy level ε_i laying between ε and $\varepsilon + d\varepsilon$ being given by

$$D(\varepsilon)d\varepsilon = \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(-\frac{\varepsilon^2}{2\sigma^2}\right) d\varepsilon \equiv G(\varepsilon, \sigma, 0)d\varepsilon, \quad (3)$$

where $G(\varepsilon, \sigma, s)$ denotes the Gaussian (normal) distribution in variable ε , centred at $\varepsilon = s$, and with a standard deviation σ . The energies ε_i are assumed to be spatially uncorrelated, *i.e.*

$$\langle \varepsilon_i \varepsilon_j \rangle = \sigma^2 \delta_{ij}. \quad (4)$$

In this paper we consider the limit of the low density of carriers (electrons, holes), which is appropriate for devices such as organic light emitting diodes (OLED), usually operating at 10^{-6} to 10^{-5} carriers per molecule in the interior of the organic film. The effect of the short range coulomb interaction as well as of the Pauli exclusion principle tends to be negligible in that concentration range. The master equation describing the evolution of the ensemble of carriers in the system out of equilibrium then has the form

$$\frac{dn_i}{dt} = n_{i-1}W_{i-1,i} - n_i(W_{i,i+1} + W_{i,i-1}) + n_{i+1}W_{i+1,i}, \quad (5)$$

with n_i denoting an ensemble average for the occupancy of the site i . In the following, we shall consider the steady state solutions of this equation for the system in the electric field. In the steady state, $dn_i/dt = 0$ and the

particle current J is constant throughout the system, with the relation

$$J = J_{i,i+1} = \Omega_i \left\{ n_i \exp\left(\frac{E_i - E_{i+1}}{2T}\right) - n_{i+1} \exp\left(\frac{E_{i+1} - E_i}{2T}\right) \right\}, \quad (6)$$

holding for every site i (the abbreviation $\Omega_i \equiv \Omega_{i,i+1}$ is used in this and the following formulae). These equations may be inverted to find n_i in the steady state. Numerically this is done by solving a large system of Eq. (6) where each site contributes with one equation. In one dimension and for the nearest-neighbour hopping, the system can be solved analytically. The derivation is given in Appendix I, resulting in the relation

$$n_i = \exp\left(\frac{-\varepsilon_i}{T}\right) \left\{ J \exp\left(-\frac{Fqa}{2T}\right) \times \sum_{k=0}^{\infty} \Omega_{i+k}^{-1} \exp\left(\frac{\varepsilon_{i+k+1} + \varepsilon_{i+k}}{2T} - k \frac{Fqa}{T}\right) \right\}. \quad (7)$$

The factor $\exp(-\varepsilon_i/T)$ in the front determines the occupancy of site i in equilibrium. In that limit, *i.e.* for $F \rightarrow 0$, the factor in the curly brackets does not depend on ε_i and becomes a mere normalization factor, related to the average concentration of the carriers in the system.

EFFECTIVE TEMPERATURE FROM NUMERICAL SIMULATIONS

In this section we present the results of the numerical simulations for the particle distribution in steady state in the external electric field. Our aim is to draw a parallel to the numerical simulations in three dimensions by showing the same signature of the effective temperature.¹⁶⁻²¹ For the purpose of the simulation we generate long chains of sites with random energy levels ε_i , with the properties described in the previous section, *i.e.*

$$\sum_i \delta(\varepsilon - \varepsilon_i) d\varepsilon \sum_i 1 = \sum_{i: \varepsilon < \varepsilon_i < \varepsilon + d\varepsilon} 1 \sum_i 1 = D(\varepsilon) d\varepsilon \quad (8)$$

The separation between sites is chosen to be $a = 0.6$ nm, a typical size for a molecule used in the organic devices. For a given electric field F , we solve numerically the system of equations for n_i . These are subsequently used to derive the statistical properties of the ensemble.

Our first quantity of interest is the distribution of particles in energy. The probability density $P_F(\varepsilon, T)$ that a particle is found at a site with the site-energy in the range $(\varepsilon, \varepsilon + d\varepsilon)$ is given by

$$P_F(\varepsilon, T) d\varepsilon \equiv \sum_{i: \varepsilon < \varepsilon_i < \varepsilon + d\varepsilon} n_i \sum_i n_i. \quad (9)$$

In equilibrium, *i.e.* in the $F \rightarrow 0$ limit, n_i follows the Boltzmann distribution,

$$n_i \propto \exp(-\varepsilon_i / T), \quad (10)$$

and $P_F(\varepsilon, T)$ reduces to

$$P_0(\varepsilon, T) \propto \exp(-\varepsilon / T) \sum_i \delta(\varepsilon_i - \varepsilon) \propto \exp(-\varepsilon / T) D(\varepsilon). \quad (11)$$

Upon normalization this gives the Gaussian distribution,

$$P_0(\varepsilon, T) = G\left(\varepsilon, \sigma - \frac{\sigma^2}{T}\right), \quad (12)$$

which has the center shifted from the center of the D.O.S. $D(\varepsilon)$ by the amount³

$$\langle \varepsilon \rangle_0 = -\frac{\sigma^2}{T}. \quad (13)$$

Note that the position of the centre of the distribution in equilibrium depends on the ambient temperature T . On the contrary, the width of the distribution is fixed and equals σ , the width of the density-of-states distribution $D(\varepsilon)$.

The hypothesis of the effective temperature at finite electric field F can then be formulated as the claim that the distribution $P_F(\varepsilon, T)$ at finite electric field has the same form as P_0 in equation (12),

$$P_F(\varepsilon, T) \approx G\left(\varepsilon, \sigma - \frac{\sigma^2}{T^*}\right). \quad (14)$$

with an effective temperature $T^* = T^*(F)$ replacing the ambient temperature.

This hypothesis may be tested through numerical simulations of the steady state in large systems. (*Note:* Recently Preezant and Tessler also concluded on the existence of the effective temperature in the $P_F(\varepsilon)$ distribution.¹⁷ Interestingly, their "effective" master equation deals directly with that distribution.) An example of our numerical results for $P_F(\varepsilon, T)$ in one dimension is shown in Figure 2. The central panel of this figure indeed suggests that, to a good approximation, the distribution $P_F(\varepsilon, T)$ may be replaced by the distribution in equilibrium at some elevated temperature. The corres-

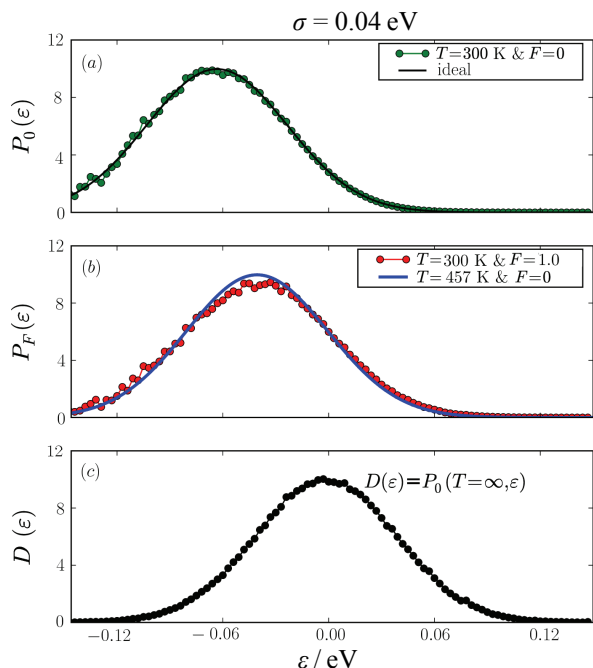


Figure 2. The distribution of the carriers in energy, as obtained in the numerical simulations for the chain of 2^{18} sites. Electric field F is measured in units of MV cm^{-1} . Panel (a) shows the distribution of particles in energy in equilibrium at $T = 300$ K; (b) shows the distribution of particles in the current-carrying state at $F = 1.0 \text{ MV cm}^{-1}$ at the same temperature, compared to the distribution of particles in equilibrium at much higher temperature of 457 K. The comparison suggests that 457 K may be used as the effective temperature to describe the current-carrying state at 300 K in the given electric field; (c) shows the density of states in the system, equivalent to the distribution of particles in the limit of the infinite temperature. The numerical results are shown with circles, while the full lines represent the theoretical Gaussian distributions.

ponding "effective temperature" T^* may be calculated from the position of the centre of the distribution $P_F(\epsilon)$,

$$-\frac{\sigma^2}{T^*} \equiv \langle \epsilon \rangle_F = \int d\epsilon P_F(\epsilon) \epsilon = \sum_i \epsilon_i n_i \sum_i n_i, \quad (15)$$

An alternative way of stating the effective temperature hypothesis is by saying that the average occupancy of the site with energy ϵ , defined as

$$n(\epsilon) \equiv \langle n_m \rangle_{\epsilon_m = \epsilon} = \sum_{i, \epsilon < \epsilon_i < \epsilon + d\epsilon} n_i \sum_{i, \epsilon < \epsilon_i < \epsilon + d\epsilon} 1, \quad (16)$$

depends on energy as

$$n(\epsilon) \propto \exp\left(-\frac{\epsilon}{T^*}\right). \quad (17)$$

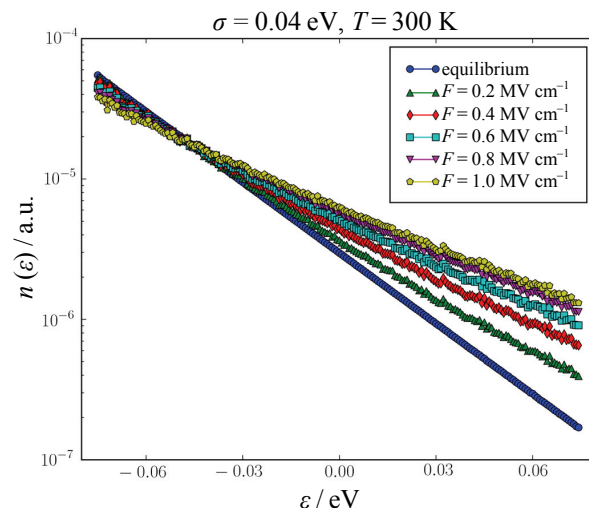


Figure 3. The mean occupancy $n(\epsilon)$ in the current-carrying state, with $n(\epsilon)$ numerically calculated through Eq. (16). Note the logarithmic scale on the vertical axis. The energy range shown in the figure covers the energies with reliable statistics. The data-set with the highest slope corresponds to the system in equilibrium at the temperature T , with the slope given by $-1/T$. The absolute value of the slope decreases as the strength of the applied electric field increases. This slope is identified as $1/T^*$, with $T^* > T$, being the effective temperature of the system out of equilibrium.

This definition of T^* is equivalent to Eq. (15) since the relation

$$P_F(\epsilon, T) \propto D(\epsilon)n(\epsilon), \quad (18)$$

follows from these Eqs. (8), (9), and (16). The dependence $n(\epsilon)$ in the current-carrying state has been recently used in Ref. 18 to identify the effective temperature in numerical simulations of the hopping conduction in three dimensions. The example of numerical results for $n(\epsilon)$ that we obtain from our numerical simulation in one dimension is shown in Figure 3. This figure also supports the concept of the effective temperature: to a good approximation the logarithm of $n(\epsilon)$ turns to be linear in ϵ . The slope of this linear dependence is identified as $1/T^*$. Obviously, T^* depends on the strength of the applied electric field. In Figure 4 we plot the results for the dependence of the effective temperature on the electric field, for several values of disorder strength σ .

THE ANALYTICAL EXPRESSION FOR THE EFFECTIVE TEMPERATURE

We start from the exact relation (7) that applies for arbitrary disorder. For an uncorrelated Gaussian disorder this relation may be easily averaged over all energy configurations, leading to an exact expression for $n(\epsilon)$.

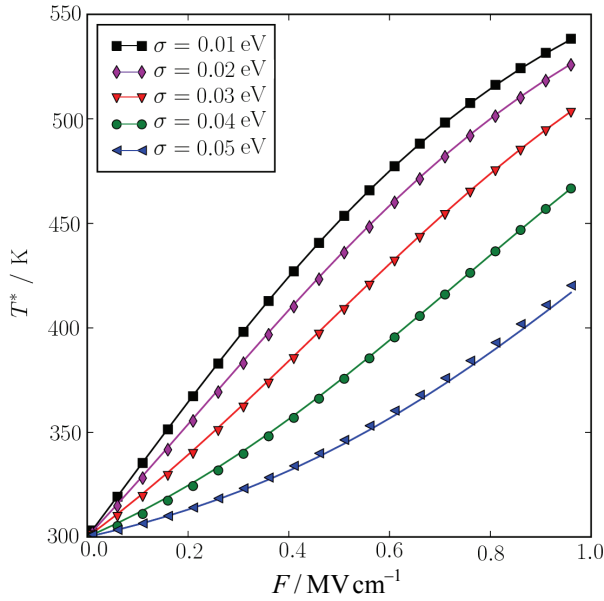


Figure 4. The effective temperature vs. electric field for several strengths of uncorrelated Gaussian disorder. The symbols represent the results of our numerical simulation on the system of 2^{18} sites. The effective temperature in the simulation was calculated using Eq. (15). The full lines correspond to the formula (24) derived in the nextcoming section.

Details of this calculation are given in Appendix II. The result is

$$n(\varepsilon) \propto \left\{ \exp\left(-\frac{\varepsilon}{T}\right) + \exp\left(-\frac{\sigma^2}{8T^2}\right) \left[\exp\left(\frac{Fqa}{T}\right) - 1 \right] \exp\left(-\frac{\varepsilon}{2T}\right) \right\}. \quad (19)$$

Two exponentials in energy ε appear in the expression, instead of one required by the hypothesis of the effective temperature. The first term corresponds to the system in equilibrium (indeed, the factor in front of the second exponential vanishes in the $F \rightarrow 0$ limit).

The presence of two exponentials corresponds to the slight curvature in $\ln n(\varepsilon)$ that can be seen at high fields in our numerical results in Figure 3. However, this effect is much less pronounced than the change of the average slope with increasing field. In other words, it is clear that the change of the average slope is related to the appearance of the second exponential in Eq. (19). This can be traced back to the fact that the occupancy of the particular site i in Eq. (7) is influenced by the energy levels at the neighbouring sites. This spatial correlation in density develops at finite electric field even when the energy levels are uncorrelated in space.

The distribution $P_F(\varepsilon)$ of particles in energy can

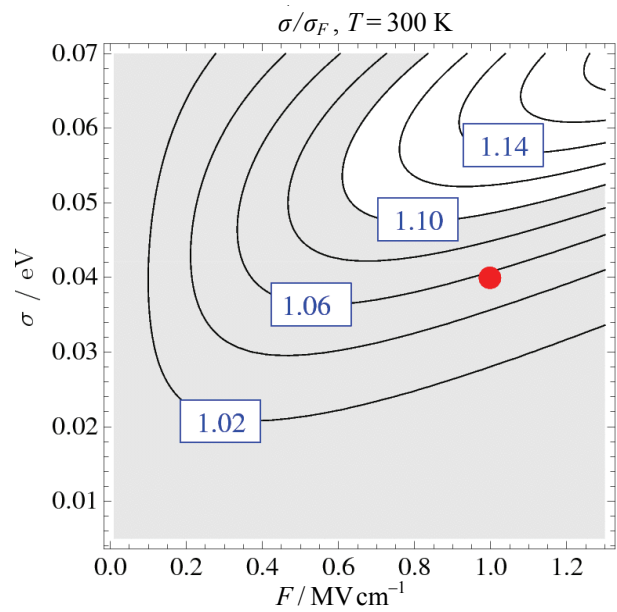


Figure 5. The contour plot of σ_F/σ , as defined in Eq. (22). The axis labels σ and F stand respectively for the strength of disorder and the electric field. In the region painted in grey, where $1 \leq \sigma_F/\sigma < 1.10$, the concept of the effective temperature for the particle-energy distribution $P_F(\varepsilon)$ works rather well. The red spot corresponds to the set of parameters used to obtain Figure 2 b.

be obtained from Eq. (19) using the relation (18). The presence of two exponentials in $n(\varepsilon)$ translates into the expression for $P_F(\varepsilon)$ which is a weighted sum of two Gaussian distributions,

$$P_F(\varepsilon) = \frac{1}{1+z} G\left(\varepsilon, \sigma - \frac{\sigma^2}{T}\right) + \frac{z}{1+z} G\left(\varepsilon, \sigma - \frac{\sigma^2}{2T}\right), \quad (20)$$

with z given by

$$z \equiv \exp\left(-\frac{\sigma^2}{2T^2}\right) \left[\exp\left(\frac{Fqa}{T}\right) - 1 \right]. \quad (21)$$

Two Gaussian distributions in Eq. (20) have the same width σ but the positions of their centres differ by $\sigma^2/2T$. The presence of two Gaussian distributions in $P_F(\varepsilon)$ is, of course, contrary to the hypothesis of a single effective temperature in relation (14). It demonstrates that this hypothesis is by no means exact, but is an approximation whose application has certain limits. Upon closer inspection, however, it turns out that in practice the two Gaussian distributions never appear visibly separated. Moreover, the asymmetry in $P_F(\varepsilon)$ is difficult to observe in a wide range of parameters. In fact, the departure of $P_F(\varepsilon)$ from the ideal form (14) may be first observed through its width

$$\sigma_F \equiv \left[\int d\varepsilon P_F(\varepsilon) (\varepsilon - \langle \varepsilon \rangle_F)^2 \right]^{1/2}, \quad (22)$$

that becomes slightly bigger than σ . The result of the numerical simulations presented in Figure 2 b already hints at that, although we can see that such deviation would be difficult to spot using numerical simulations only. Our calculation shows that the ratio σ_F/σ stays very close to unity in the wide range of disorder and field parameters. This is detailed in Figure 5 where the values of the ratio σ_F/σ is shown in the (σ, F) plane. This figure also indicates the region in which the concept of the effective temperature works very well.

The wide range of the applicability of the concept of the effective temperature in the (σ, F) space permits us to proceed with the evaluation of the effective temperature defined through Eq. (15). From Eq. (20) we obtain,

$$\frac{\sigma^2}{T^*} = -\frac{\sigma^2}{T} \frac{1+z/2}{1+z}. \quad (23)$$

Finally, the formula for the effective temperature in the current-carrying state reads as

$$T^* = T \frac{1 + \exp\left(-\frac{\sigma^2}{2T^2}\right) \left[\exp\left(\frac{Fqa}{T}\right) - 1 \right]}{1 + \frac{1}{2} \exp\left(-\frac{\sigma^2}{2T^2}\right) \left[\exp\left(\frac{Fqa}{T}\right) - 1 \right]}. \quad (24)$$

The data in the numerical simulations indeed follow this relation, as shown by full lines in Figure 4. This expression for the effective temperature is exact within the definition given through Eq. (15). Note that the formula is considerably more complicated than those proposed previously in the literature on the basis of numerical simulations and approximate analytical calculations.¹⁷⁻²¹ In the limit of low electric field ($F < 0.3$ MV/cm at room temperature, as may be seen in Figure 4), the relation (24) may be simplified to an approximate one,

$$T^* \approx T + \gamma(\sigma)Fqa, \quad (25)$$

with

$$\gamma(\sigma) \equiv \frac{1}{2} \exp\left(-\frac{\sigma^2}{2T^2}\right), \quad (26)$$

which is of the same level of complexity as the relations for the effective temperature proposed earlier on the phenomenological grounds and in related contexts.¹⁷⁻²¹ However, in contrast to previous studies in one-dimension (Ref. 22 and references therein), the factor γ in Eq. (25) depends on the strength of disorder and that this dependence is strong for σT . Moreover, the dependence of T^* on electric field in Eq. (25) is linear for low

fields, which counters the argument presented by Cleve *et al.* in Ref. 20. In that respect it may be noted that our definition of the effective temperature via particle distribution in energy differs from the definition of the effective temperature through mobility, used in Ref. 20. This allows for possible differences in effective temperatures defined in two manners, including their field dependence at low fields. It may be also noted that the assumptions used in the argument in Ref. 20 are not necessarily fulfilled for the mobility derived using the simple hopping formulae (Appendix III).

CONCLUSION

We address the properties of the ensemble of hopping carriers in a disordered landscape of energy levels, in the limit of one dimension and a uncorrelated disorder. Focusing on that limit allows for an exact analytical treatment that brings additional insight into factors behind the phenomenon of the effective temperature that has been observed in various numerical simulations. In the current communication we derive the exact analytic expression for the distribution of particles in energy and the effective temperature in the current-carrying state. The applicability of the concept of effective temperature is tested through numerical simulations and analytical calculations, and is illustrated in Figure 5.

One can foresee several directions in which the present work could be expanded. First, spatially-correlated disorder seems to be much more relevant for disordered organics than uncorrelated disorder.^{6,7} Moreover, the numerical simulations have shown that the effective temperature is strongly sensitive to correlation.¹⁷ Although the case of a correlated disorder is more difficult to address than the case considered here, the extension of the present approach in that direction seems achievable. Second, the limits of the applicability of the concept of effective temperature on the calculation of the electric current has to be re-examined. Finally, there is a hope that what is learned in one dimension may be also used in higher dimensions. In particular, the spatial density-correlations caused by the electric field, as hinted by Eq. (7), might be related to the filamentary nature of the conduction in higher dimensions.¹²⁻¹⁴

APPENDIX I

Site Occupancy n_i in the Steady State

Here we provide a brief derivation of the expression (7) for n_i in the steady state. This expression is essentially the one derived by Derrida,²⁵ although the final form and the derivation presented below somewhat differ from Derrida's. We start from the equation (6) for the steady state. This equation may be written as a recursion,

$$n_{i+1} = n_i e_i^2 - J \Omega_i e_i, \quad (27)$$

where we have introduced the abbreviation

$$e_i \equiv \exp\left(\frac{E_i - E_{i+1}}{2T}\right) = \exp\left(\frac{\varepsilon_i - \varepsilon_{i+1} + Fqa}{2T}\right). \quad (28)$$

The recursion may be re-iterated to relate two arbitrary distant sites,

$$n_{i+1} = n_0 \prod_{j=0}^i e_j^2 - J \sum_{k=0}^i \left[\Omega_k^{-1} e_k^{-1} \prod_{j=k}^i e_j^2 \right], \quad (29)$$

or

$$n_{i+1} = \exp\left(-\frac{E_{i+1}}{T}\right) \left\{ n_0 \exp\left(\frac{E_0}{T}\right) - J \sum_{k=0}^i \left[\Omega_k^{-1} e_k^{-1} \exp\left(\frac{E_k}{T}\right) \right] \right\}. \quad (30)$$

Since the prefactor

$$\exp\left(-\frac{E_{i+1}}{T}\right) = \exp\left[\frac{-\varepsilon_{i+1} + (i+1)Fqa}{T}\right] \quad (31)$$

diverges for $F > 0$ and in the $i \rightarrow 0$ limit, the condition of keeping n_{i+1} finite requires that the expression in curly brackets in (30) vanishes in that limit. This determines the relation between the steady state current J and n_0 ,

$$n_0 = \exp\left(-\frac{E_0}{T}\right) \left\{ J \sum_{k=0}^{\infty} \Omega_k^{-1} \exp\left(\frac{E_{k+1} + E_k}{2T}\right) \right\}. \quad (32)$$

Generalizing to an arbitrary site i , and written in terms of molecular energy levels, ε_i the last relation turns into Eq. (7).

APPENDIX II

The Distribution $n(\varepsilon)$ in the Current-carrying Case

We start from Eq. (7). Keeping the energy at site i fixed, $\varepsilon_i = \varepsilon$, we average over the energies of all other sites in the system. For the system with uncorrelated disorder this is done independently for each site. The average is calculated using the density of states in Eq. (8). The calculation leads to the expression

$$n(\varepsilon) = \exp\left(\frac{-\varepsilon}{T}\right) J \Omega_0^{-1} e^{-Fqa/2T} \times \left[e^{\varepsilon/2T} \langle e^{\varepsilon'/2T} \rangle + \sum_{k=1}^{\infty} \langle e^{\varepsilon'/2T} \rangle^2 e^{-kFqa/T} \right]. \quad (33)$$

The average of the exponential is calculated as the following integral,

$$u \equiv \left\langle \exp\frac{\varepsilon'}{2T} \right\rangle = \int d\varepsilon' \exp\frac{\varepsilon'}{2T} \frac{1}{\sqrt{2\pi\sigma^2}} \exp\left(-\frac{\varepsilon'^2}{2\sigma^2}\right) = \exp\frac{\sigma^2}{8T^2}. \quad (34)$$

Upon substituting into the previous expression this gives

$$\begin{aligned} n(\varepsilon) &\propto \exp\left(-\frac{\varepsilon}{T}\right) \left(e^{\varepsilon/2T} u + \sum_{k=1}^{\infty} u^2 e^{-kFqa/T} \right) = \\ &\exp\left(-\frac{\varepsilon}{T}\right) \left(e^{\varepsilon/2T} u + u^2 \frac{e^{-Fqa/T}}{1 - e^{-Fqa/T}} \right) = \\ &\exp\left(-\frac{\varepsilon}{2T}\right) \exp\frac{\sigma^2}{8T^2} + \exp\left(-\frac{\varepsilon}{T}\right) \exp\frac{\sigma^2}{4T^2} \frac{1}{e^{Fqa/T} - 1}, \\ &\propto \exp\left(-\frac{\varepsilon}{T}\right) + \exp\left(-\frac{\sigma^2}{8T^2}\right) \left[\exp\left(\frac{Fqa}{T}\right) - 1 \right] \exp\left(-\frac{\varepsilon}{2T}\right), \end{aligned} \quad (35)$$

which is the expression used in Eq. (19).

APPENDIX III

The argument in Ref. 20 relies on the assumption that $(\partial\mu(F)/\partial F)_{F=0} = 0$. One may check this assumption against the most trivial case of the one-dimensional hopping transport in the ordered medium with Müller-Abrahams hopping rates between nearest-neighbours. The current in the system, J , is then given by the difference of the hopping probabilities (per unit time) for down-hill and up-hill hops, respectively given by v_0 and $v_0 \exp(-Fqa/T)$ (see Eq. (8a) of Ref. 20). This leads to $J \propto |1 - \exp(-Fqa/T)|$, or $\mu(F) \propto |1 - \exp(-Fqa/T)|/F$. It may be readily seen that the above assumption does not hold in this simple case. Including the hopping to further sites, and introducing disorder is not expected to change this, showing that the assumption is inconsistent with simple exponential formulae for the hopping probabilities ordinarily used (Ref. 20 included).

Acknowledgements. This work was supported by the Croatian MoSES project awards 035-0352826-2847 and 119-1191458-0512.

REFERENCES

- G. Malliaras and R. Friend, *Phys. Today* **58** (2005) 53–58.
- R. H. Friend, R. W. Gymer, A. B. Holmes, J. H. Burroughes, R. N. Marks, C. Taliani, D. D. C. Bradley, D. A. Dos Santos, J. L. Bredas, M. Logdlund, W. R. Salaneck, *Nature* **397** (1999) 121–128.
- H. Bässler, *Phys. Status Solidi B* **175** (1993) 15–56.
- P. M. Borsenberger, L. T. Pautmeier, and H. Bässler, *Phys. Rev. B* **48** (1993) 3066–3073.
- Y. N. Gartstein and E. M. Conwell, *Chem. Phys. Lett.* **245** (1995) 351–358.
- D. H. Dunlap, P. E. Parris, and V. M. Kenkre, *Phys. Rev. Lett.* **77** (1996) 542–545.
- S. V. Novikov, D. H. Dunlap, V. M. Kenkre, and P. E. Parris, *Phys. Rev. Lett.* **81** (1998) 4472–4475.
- H. Cordes, S. D. Baranovskii, K. Kohary, P. Thomas, S. Yamasaki, F. Hensel, and J.-H. Wendorff, *Phys. Rev. B* **63** (2001) 094201-1–094201-9
- W. F. Pasveer, J. Cottaar, C. Tanase, R. Coehoorn, P. A. Bobbert, P. W. M. Blom, D. M. de Leeuw, M. A. J. Michels, *Phys. Rev. Lett.* **94** (2005) 206601-1–206601-4.
- R. Coehoorn, P. Bobbert, W. F. Pasveer, and M. A. J. Michels, *Phys. Rev. B* **72** (2005) 155206-1–155206-20.
- H. Houili, E. Tutiš, I. Batistić, and L. Zuppiroli, *J. Appl. Phys.* **100** (2006) 033702.
- Z. G. Yu, D. L. Smith, A. Saxena, R. L. Martin, A. R. Bishop, *Phys. Rev. Lett.* **84** (2000) 721–724.
- E. Tutiš, I. Batistić, and D. Berner, *Phys. Rev. B* **70** (2004) 161202-1–161202-4.
- J. J. M. van der Holst, M. A. Uijttewaai, B. Ramachandhran, R. Coehoorn, P. A. Bobbert, G. A. de Wijs, and R. A. de Groot, *Phys. Rev. B* **79** (2009) 085203-1–085203-11.
- P. E. Parris, D. H. and V. M. Kenkre, *Phys. Status Solidi (b)*, **218** (2000) 47–53.
- Y. Preezant and N. Tessler, *Phys. Rev. B* **73** (2006) 235202-1–235202-5.
- F. Jansson, S. D. Baranovskii, F. Gebhard, and R. Österbacka, *Phys. Rev. B* **77** (2008) 195211-1–195211-7.
- S. Marianer and B. I. Shklovskii, *Phys. Rev. B* **46** (1992) 13100–13103.
- B. Cleve, B. Hartenstein, S. D. Baranovskii, M. Scheidler, P. Thomas, and H. Bässler, *Phys. Rev. B* **51** (1995) 16705–16713.
- S. D. Baranovskii, B. Cleve, R. Hess, and P. Thomas, *J. Non-Cryst. Solids* **166** (1993) 437–440.
- V. I. Arkhipov and H. Bässler, *Philos. Mag. Lett.* **69** (1994) 241–246.
- A. Miller and E. Abrahams, *Phys. Rev.* **120** (1960) 745–755.
- D. Emin, *Adv. Phys.* **24** (1975) 305–348.
- P. E. Parris, V. M. Kenkre, and D. H. Dunlap, *Phys. Rev. Lett.* **87** (2001) 126601-1–126601-4.
- B. Derrida, *J. Stat. Phys.* **31** (1983) 433–450.

SAŽETAK

Energetska raspodjela čestica i efektivna temperatura za transport preskocima elektrona u jednodimenzionalnom neuređenom sustavu

Eduard Tutiš,^a Ivan Jurić^a i Ivo Batistić^b

^aLaboratorij za fiziku transportnih svojstava, Institut za fiziku, Bijenička c. 46, P.P. 304, HR-10001 Zagreb, Hrvatska

^bFizički odsjek, Prirodoslovno-matematički fakultet, Bijenička c. 32, HR-10000 Zagreb, Hrvatska

Nedavno objavljene numeričke simulacije elektronskog transporta u neuređenim organskim materijalima sugeriraju mogućnost opisa elektronskog ansambla u električnom polju preko efektivne temperature. Međutim, ti radovi ne objašnjavaju razloge pojavljivanja efektivne temperature, a izrazi koji se predlažu za efektivnu temperaturu su fenomenološke naravi. U ovom radu su ta pitanja egzaktno riješena za jednodimenzionalni model s nekoreliranim Gaussovima neredom. Nađeni su izrazi za raspodjele čestica po energiji u neravnotežnom stanju, izvedena je formula za efektivnu temperaturu, te su određene granice primjenljivosti samog koncepta.