Energy position of the transport path in disordered organic semiconductors

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Abstract

The concept of *transport energy* is the most transparent theoretical approach to describe hopping transport in disordered systems with steeply energy dependent density of states (DOS), in particular in organic semiconductors with Gaussian DOS. This concept allows one to treat hopping transport in the framework of a simple multiple-trapping model, replacing the mobility edge by a particular energy level called the transport energy. However, there is no consensus among researchers on the position of this transport level. In this article, we suggest a numerical procedure to find out the energy level most significantly contributing to charge transport in organic semiconductors. The procedure is based on studying the effects of DOS modifications on the charge carrier mobility in straightforward computer simulations. We also show why the *most frequently visited energy*, computed in several numerical studies to determine the transport energy, is not representative for charge transport.

Keywords: organic semiconductors, transport energy, electronic transport

(Some figures may appear in colour only in the online journal)

1. Introduction

Although the description of charge transport in disordered organic semiconductors is of high importance for device applications and for the fundamental understanding of material properties, a comprehensive, transparent and self-consistent theory that can accurately predict transport properties is yet to be found. This is particularly unsatisfactory because a simple and clearly defined model framework existed for years: Charge carriers move via incoherent tunneling (*hopping*) between spatially and energetically distributed localized states with a Gaussian energy spectrum [1–3]:

$$g(\varepsilon) = \frac{N}{\sigma\sqrt{2\pi}}\exp\left(-\frac{\varepsilon^2}{2\sigma^2}\right), \qquad (1)$$

where σ is the energy scale of the *density of states* (DOS), usually estimated as $\sigma \approx 0.1$ eV [1] and N is the concentration of randomly distributed localized states (sites) that charge

carriers can use for hopping transport. The rate for a carrier transition from an occupied site i to an empty site j, separated by a distance r_{ij}, is usually assumed [1, 4] to be described by the Miller–Abrahams expression [5]

$$\nu_{ij} = \nu_0 \exp\left(-\frac{2r_{ij}}{\alpha} - \frac{\varepsilon_j - \varepsilon_i + |\varepsilon_i - \varepsilon_j|}{2kT}\right). \qquad (2)$$

Here, α is the localization length of charge carriers in the localized states, usually of the order of $10^{(-8)}$ cm [6, 7], ε_i and ε_j are the carrier energies on sites i and j, respectively, k is the Boltzman constant and T is the temperature. The energy difference between states i and j must be compensated, for example, by absorption or emission of phonons. The prefactor ν_0, often called the *attempt-to-escape* frequency, depends on the interaction mechanism that causes transitions. In the case of interaction with phonons, its value is usually assumed close to the phonon frequency [4], $\nu_0 \approx 10^{12}$ s^{-1}. Due to the interplay between the spatial term in equation (2), dependent on
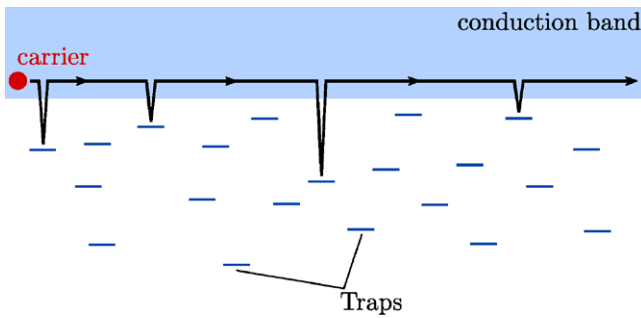
Figure 1. Schematic view of the MT transport mode. The carrier motion in the conduction band is interrupted by trapping into and subsequent activation from localized states (traps).

Figure 2. Schematic view of charge transport via the transport energy in a system with Gaussian DOS. The carrier distribution, with its maximum at the equilibration energy $-\sigma^2/kT$, is drawn with a red background.

parameters characterized by the product $N\alpha^3$ and the energy term, dependent on temperature and site energies characterized by the ratio σ/kT, charge transport at low temperatures is dominated by more distant transitions than at higher temperatures, i.e., transport takes place in the *variable-range-hopping* (VRH) mode [8]. If spatial positions and energies of localized states can be considered independent from each other, this model is traditionally called the *Gaussian Disorder Model* (GDM).

The most comprehensive theoretical tool to describe hopping transport in disordered materials is the percolation theory [8–11]. Particularly simple is its application to lattice models with transitions only between nearest neighboring sites [8]. Such a transport mode is called the *nearest-neighbor-hopping* (NNH) on a lattice. Percolation theory for this kind of transport was developed in the 1970s [8]. For the NNH on a cubic lattice with Gaussian DOS given by equation (1), it predicts that thermal activation of carriers to a particular energy level E_c dominates charge transport [12]. The energy E_c is determined by the condition [12]

$$\int_{-\infty}^{E_c} g(\varepsilon)\,\mathrm{d}\varepsilon = x_c, \tag{3}$$

where x_c is the percolation threshold for the site percolation problem. Using the value $x_c = 0.31$, Schönherr *et al* [12] obtained $E_c \approx -0.6\sigma$. A very close result $E_c \approx -0.5\sigma$ for the same problem has recently been published by Cottaar *et al* [13]. Since the value of x_c is sensitive to the geometrical structure of the chosen regular lattice, the value of the energy E_c determined via equation (3) must also depend on the lattice structure. Therefore, the NNH on a lattice can hardly be suitable to study charge transport in a spatially disordered organic material, where transport was shown to take place via VRH [1]. While the application of the percolation theory to NNH on a lattice is a straightforward exercise [8], the derivation of the percolation approach for the VRH is a much more complicated task [8]. Aside from the uniform DOS [8], only in the case of an exponential DOS, $g(\varepsilon) = g_0\exp(\varepsilon/\varepsilon_0)$, precise results not only for the exponentially strong dependences of transport coefficients on material parameters, but also for the slowly varying preexponential factors, could recently be obtained by percolation theory applied to VRH [14].

An alternative powerful theoretical method to describe hopping transport in systems with steeply energy-dependent DOS, complementary to the p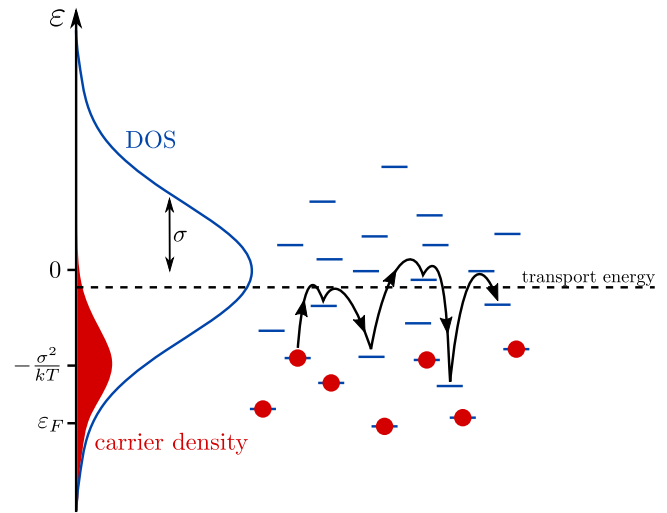ercolation theory, is the concept of the *transport energy* (TE). The essence of the TE approach is the ability to describe hopping transport via sites distributed in space and energy in full analogy with the multiple-trapping (MT) model [15]. The MT model has been developed to describe charge transport in inorganic disordered materials, such as chalcogenide glasses and amorphous silicon [16–18], which possess a so-called *mobility edge*, i.e., the energy level that separates extended states with rather high carrier mobility from the localized states, which can be considered as traps. In the MT model a charge carrier moves only via delocalized states with energies above the mobility edge. This motion is interrupted by trapping into localized states with subsequent activation of the carrier back into the conducting states at the mobility edge. Transport in the framework of the MT model is shown schematically in figure 1. The advantage of the MT charge transport mode as compared to the VRH, is the possibility of exact theoretical treatment by analytical equations under equilibrium and non-equilibrium conditions and for various shapes of the DOS [19–21].

In the following section 2, various approaches and different definitions of the TE are summarized in order to point out the ambiguity of the term *transport energy*. In section 4 the popular numerical approach to the TE as the *most frequently visited energy* in Monte Carlo simulations is proven not to be related to the long-range transport. We suggest an alternative numerical procedure in section 5 and show that its results agree well with the analytic derivation of the transport energy by Oelerich *et al* [22], the important steps of which are briefly presented in section 3. In section 6 we address the issue of using the TE for description of non-equilibrium processes in the GDM. Concluding remarks are gathered in section 7.

2. The transport energy

In 1985 Monroe [23] has shown that in the case of the exponential DOS one can describe VRH in the framework of the

MT formalism by replacing the mobility edge in the MT model by some particular energy level ε_t, that he called the *transport energy* (TE). A slightly different derivation of ε_t was later suggested by Baranovskii et al [24]. The latter approach was soon extended [25] for systems with other than exponential energy spectra, in particular with a Gaussian DOS described by equation (1). Schematic movement of charge carriers via activation to the TE in a system with Gaussian DOS is shown in figure 2. It is worth emphasizing that the value ε_t of the TE is determined by the interplay between the term in equation (2) dependent on spatial parameters and the term dependent on temperature and site energies, i.e., the TE reflects the essence of the VRH transport mechanism [23–25].

Numerous derivations of the value of ε_t have been suggested in the literature and there is a lot of confusion in this respect. Studying VRH in systems with exponential DOS in the framework of the percolation theory, Grünewald and Thomas [26] have shown that in thermal equilibrium the dc conductivity σ_{dc} can be represented in the Arrhenius form,

$$\sigma_{dc} \propto \exp\left(-\frac{\varepsilon^* - \varepsilon_F}{kT}\right), \qquad (4)$$

where ε_F is the Fermi energy and ε^* is some energy level dependent on the parameters $N\alpha^3$ and kT/ε_0, where ε_0 denotes the energy scale of the exponential DOS. Later, the description of the VRH in the exponential DOS using percolation theory has been attempted by Vissenberg and Matters [27], though with a lower accuracy as compared to the initial derivation of Grünewald and Thomas [14]. The lower accuracy of the approach by Vissenberg and Matters is due to the different interpretation of the percolation criterion as compared to the classical one formulated by Pollak [11] and used by Grünewald and Thomas: In order to establish infinite percolation, the classical recipe demands an average number of $B_c \approx 2.7$ neighbors around each site, to which the transition rates are the highest and which *belong to the percolation cluster*. Vissenberg and Matters [27] instead averaged over *all* sites in the system, which leads to less accurate results as evidenced by comparison to the exact solution of the VRH problem for the exponential DOS obtained by Nenashev et al [14].

Percolation theory based on the classical recipe [11] has also been developed to describe the VRH in the GDM [28–30], i.e., in a random system of sites with a Gaussian energy distribution. It was shown that in thermal equilibrium at low temperatures or at high concentrations of carriers, cases in which the Fermi level ε_F is situated above the equilibration energy $\varepsilon_\infty = -\sigma^2/kT$, the temperature dependence of the dc conductivity σ_{dc} exhibits a simple activated behavior similar to equation (4) with ε^* determined by the relation

$$\varepsilon^* = -\sqrt{2\ln(Q)}\,\sigma, \quad Q = \beta\frac{N\alpha^3}{s^2}\left(\frac{\sigma}{kT}\right)^3, \qquad (5)$$

where β is a constant and $s \approx -\ln^{1/2}(Q)$ is a logarithmic function of $N\alpha^3$ and σ/kT.

The simple forms of equations (4) and (5) might lead to the conclusion that in systems with exponential or Gaussian DOS the VRH takes place by activation of charge carriers towards the energy ε^*. However, the r-dependent term in equation (2) plays an equally important role for VRH as the thermal activation. Therefore, one can hardly assume that the description of the VRH as simple activation of carriers towards the energy level ε^* would mean that carriers actually conduct moving around that energy level. One should instead interpret the energy level ε^* in equation (4) as an 'effective' energy level serving formally to mimic the combined effects of the spatial tunneling and of the activation in energy. Therefore, percolation theory is not a suitable tool to determine the 'real' energy level that charge carriers have to visit in order to provide transport in the VRH regime. Mixing up the effective TE ε^* with the real TE ε_t is probably the main reason for the confusion with respect to the term 'transport energy'. While the effective TE ε^* serves as a formal tool to describe the VRH as a simple activation, the real TE ε_t characterizes the actual transport level at which carriers conduct via spatial tunneling.

The difference between these effective and real TEs becomes more transparent if one compares the expressions for the mobility of charge carriers μ derived in various approaches. After deriving ε_t for the GDM [25] in full analogy with the derivations for the exponential DOS, Baranovskii et al [31] suggested the following expression for the carrier mobility:

$$\mu = \mu_0 \exp\left[-\frac{2R(\varepsilon_t)}{\alpha} - \frac{\varepsilon_t}{kT} - \frac{1}{2}\left(\frac{\sigma}{kT}\right)^2\right], \qquad (6)$$

where $R(\varepsilon_t)$ is the typical distance between localized states with energies below ε_t. Working with the effective TE ε_{eff}, Arkhipov et al [32] and Nikitenko et al [33] suggested a slightly different expression for the carrier mobility:

$$\mu = \widetilde{\mu_0} \exp\left[-\frac{\varepsilon_{eff}}{kT} - \frac{1}{2}\left(\frac{\sigma}{kT}\right)^2\right]. \qquad (7)$$

Taking into account that the values of μ_0 in equation (6) and $\widetilde{\mu_0}$ in equation (7) are very close to each other [31–33], it is apparent that the meaning of the effective TE ε_{eff} is different to that of the TE ε_t. The effective TE ε_{eff} in equation (7) is denoted differently to the energy ε^* in equation (4) particularly because the percolation nature of the VRH has not been taken into account in the derivation of ε_{eff} by Arkhipov et al [32] and Nikitenko et al [33]. The same is true for the initial derivation of the ε_t in the GDM by Baranovskii et al [25]. Later Oelerich et al [22] derived the equation for ε_t in the GDM taking into account the percolation criterion and extending this concept for finite carrier concentrations n. This approach is outlined in section 3. The dependence of the 'effective' transport energy ε_{eff} on n has been derived by Arkhipov et al [34].

Another interesting definition of the TE was suggested by Schmechel [35], who, in contrast to all other derivations of the TE, did not use this quantity to calculate the carrier mobility, but instead derived the TE *from* the previously calculated dc conductivity by introducing a differential energy-dependent dc conductivity. The percolation nature of the cluster of sites responsible for the VRH has not been taken into account in Schmechel's derivation.

Martens *et al* [36] essentially repeated the derivation by Baranovskii *et al* [25] also not taking into account the percolation nature of the VRH transport.

Sometimes the following arguments are used to determine the position of the TE in the GDM [1, 37–39]. First one assumes that the temperature dependence of the carrier mobility $\mu(T)$ is completely decoupled from the dependence on the concentration of sites N. Then one uses the expression for $\mu(T)$ in the form obtained in computer simulations [1]

$$\mu \propto \exp\left[-\left(C\frac{\sigma}{kT} \right)^2 \right], \qquad (8)$$

with $C = \frac{2}{3}$. Assuming that transport is governed by activation of carriers from the equilibration energy $\varepsilon_\infty = -\sigma^2/kT$ towards some transport level ε_B, one obtains from equation (8) $\varepsilon_B \approx -\left(\frac{\sigma^2}{kT} \right) + 4/9\left(\frac{\sigma^2}{kT} \right) = -5/9\left(\frac{\sigma^2}{kT} \right)$. Oelerich *et al* [22] argued that the level ε_B can hardly be considered as a good candidate for the TE since in thermal equilibrium not all carriers occupy energies equal to $\varepsilon_\infty = -\sigma^2/kT$, but are instead distributed around ε_∞ in a broad energy range with the width [1] of the order σ. Carriers with energies in the upper part of that distribution contribute significantly more to transport than carriers from the lower part of the distribution, since activation to the transport level ε_t is exponentially easier for carriers with higher energies. The effective activation energy $\sigma^2/2kT$ is therefore essentially smaller than σ^2/kT.

Several algorithms have been developed and applied to determine the value of the TE by Monte Carlo computer simulations. Schönherr *et al* [12] assumed that $\varepsilon_\infty(T)$ must ultimately approach an asymptotic value as a function of temperature because of the Gaussian DOS profile. Their computer simulations suggest that for $\sigma/kT > 6$, the value $\varepsilon_\infty(T) \approx -1.8\,\sigma$ is attained. The activation energy of the mobility obtained in those simulations caused the assumption that the transport level is situated at an energy 0.70σ *above* the center of the Gaussian DOS [12]. The drastic discrepancy between this result and the value $E_c \approx -0.6\sigma$ obtained using equation (3) ultimately led to the conclusion that percolation theory is not suitable to account for hopping transport in the GDM [12]. In fact, this conclusion is due to the small size of the simulated array of sites. At small temperatures sites with energies around $\varepsilon_\infty = -\sigma^2/kT$ were statistically not present in the system and the simulated position of $\varepsilon_\infty(T)$ at low temperatures corresponded simply to the smallest energy still present in the simulated array of sites. Therefore this value did not depend on T in the simulations by Schönherr *et al* [12]. No saturation would have been recognized if the size of the simulated system had been increased.

Another computer algorithm to determine the position of the TE was used by Cleve *et al* [40], who suggested to trace the energies of sites targeted by carriers in hopping transitions during Monte Carlo simulations and to determine the position of the TE as the energy of the most frequently targeted sites. This algorithm has been used in several numerical studies [37, 41, 42]. Hartenstein and Bässler [37] recognized,

however, that due to oscillations of carriers within pairs of spatially and energetically close sites the most frequently targeted energy is not the energy essential for charge transport. The effect of such oscillations for the most frequently targeted sites has been addressed in analytical studies [43] and in computer simulations [42]. Very recently a comprehensive study of this effect has been provided by Mendels and Tessler [44], who suggested the analytical and numerical methods how to exclude the effect of frequent oscillations. The obtained distribution of targeted energies have not been related though to long-range charge transport [44]. In section 4 we address the issue of frequent carrier oscillations in detail, also showing that the algorithm suggested by Cleve *et al* is not suitable to find the position of the TE.

In section 5 we suggest a novel numerical algorithm as an alternative approach, which allows one to determine the position of the TE responsible for the long-range VRH transport in the GDM. The algorithm is checked by application to the exactly solvable model of the NNH on a lattice. Straightforward computer simulations of the VRH in the GDM carried out in the framework of this algorithm reveal the analytical estimate of the TE by Oelerich *et al* [22] as valid. In that analytical estimate one assumes that the TE corresponds to the target energy which maximizes the rate of carrier transitions upward in energy accompanied by tunneling over a distance typical for sites with energies below TE. This approach is outlined in section 3.

3. Analytic estimate of the transport energy

Let us briefly repeat the important steps in the analytical derivation of the TE following [22, 24, 25, 31, 45]. The obtained value for the TE will be compared with numerical results in section 5.

Consider a carrier in a state with energy ε_i. According to equation (2), the typical rate of a downward hop of such a carrier to a localized state deeper in energy is [22]

$$\nu_\downarrow = \nu_0 \exp\left[-\frac{2r(\varepsilon_i)}{\alpha} \right], \qquad (9)$$

where

$$r(\varepsilon) = \left[\frac{4\pi}{3} \int_{-\infty}^{\varepsilon} g(\varepsilon')\left[1 - f(\varepsilon', \varepsilon_F) \right] d\varepsilon' \right]^{-1/3}. \qquad (10)$$

The typical rate of an upward hop of such a carrier to a localized state with energy $\varepsilon_x > \varepsilon_i$ is

$$\nu_\uparrow(\varepsilon_i, \varepsilon_x) = \nu_0 \exp\left[-\frac{2r(\varepsilon_x)}{\alpha} - \frac{\delta}{kT} \right], \qquad (11)$$

where $\delta = \varepsilon_x - \varepsilon_i > 0$. Note, that this expression is not exact: The considered distance $r(\varepsilon_x)$ is based on all empty states with energies deeper than ε_x. For the Gaussian DOS this is equivalent to considering a slice of energy with the width of the order σ. This works for a DOS that varies slowly compared to kT, but not in general. It is also assumed for simplicity that the localization length α does not depend on energy. The latter

assumption can be released easily on the cost of a somewhat more complicated form of the equations. We will analyze the hopping rates at a given temperature T and try to find out the energy level ε_x, which provides the fastest typical hopping rate for a charge carrier initially placed at a site with the energy ε_i. The corresponding energy ε_x is determined by the condition

$$\frac{\partial \nu_\uparrow(\varepsilon_i, \varepsilon_x)}{\partial \varepsilon_x} = 0. \quad (12)$$

It is easy to show [24, 25] that the target energy ε_x determined by equation (12) does not depend on the initial energy ε_i and hence is universal for given parameters $N\alpha^3$ and σ/kT and can be considered as the transport energy ε_t. It means that from states below ε_t carrier transitions essential for charge transport occur to the vicinity of ε_t, while transitions from states with energies higher than ε_t occur downwards in energy. This is the essence of the TE concept. After relaxation towards ε_t the behavior of carriers resembles the MT mode [15] with ε_t playing the role of the mobility edge [23–25].

Solving equation (12) for ε_x, the TE ε_t is then determined by [22]

$$\frac{2kT}{3\alpha}\left(\frac{4\pi}{3B_c}\right)^{-1/3}\left[\int_{\varepsilon_t}^{\infty} g(\varepsilon)\left[1 - f(\varepsilon, \varepsilon_F)\right]d\varepsilon\right]^{-4/3}$$
$$\times \left[1 - f(\varepsilon_t, \varepsilon_F)\right]g(\varepsilon_t) = 1. \quad (13)$$

The factor B_c in this expression accounts for the percolation nature of the hopping transport, i.e., for the necessity to provide a continuous path via sites separated by appropriate distances. Accurate estimates give [46] the value $B_c = 2.735$.

4. On the most frequently visited energy

In this Section we highlight why computer algorithms based on tracing the energies of most frequently visited sites are not suitable to determine the position of the TE. In such algorithms one counts the number of carrier transitions, which, in course of a standard Monte Carlo simulation, bring the carrier into a particular energy interval. The statistics of such events is then plotted as a histogram with number of hops per energy interval. The peak of this histogram, i.e., the *most frequently visited energy* is interpreted as pointing at the position of the TE [40–42].

Although it has been doubted that such an algorithm is accurate enough to determine the position of the TE [37, 42], the most frequently visited energy is often used not only to determine the TE, but even for conclusions on the existence or non-existence of the TE [41]. Therefore we find it instructive to analyze the approach based on the most frequently visited energy in more detail. Below, we derive an analytic expression for the most frequently visited energy and validate it via comparison with computer simulations. We will focus on the Gaussian DOS, although the treatment of the exponential DOS is very similar. The derivation shows which hops determine the energy in question and that these hops *cannot* be the ones decisive for mobility or other long-range transport coefficients.

We will determine the frequency of hops $f(\varepsilon_j)\,d\varepsilon_j$ with the target energy within the interval $[\varepsilon_j, \varepsilon_j+d\varepsilon_j]$. Let us assume that the electron concentration is low and that only a weak external electric field is applied. The occupation probability for any site with energy ε is then given by a Boltzmann distribution

$$P_B(\varepsilon) \approx e^{\varepsilon_F/kT}e^{-\varepsilon/kT} \equiv \mathcal{N}e^{-\varepsilon/kT}, \quad (14)$$

where the normalization constant $\mathcal{N}$ is determined by the concentration of electrons. The expected frequency of hops to a particular site with the energy ε_j is determined by integrating the hopping rate $\nu_{ij} = \nu(\varepsilon_i, \varepsilon_j, r)$ (see equation (2)) over all possible initial energies ε_i and hopping distances r. The contributions from sites with the energy ε_i and distance r are weighted by the expected number of such sites $g(\varepsilon_i)$ and by the probability $P_B(\varepsilon_i)$ that the initial site is occupied. The resulting frequency of hops $f(\varepsilon_j)$ to one site with energy ε_j, weighted by the concentration of such sites $g(\varepsilon_j)$, gives the frequency of hops to the energy ε_j.

It is convenient to evaluate the hop frequency separately for hops upward and downward in energy, since the form of the hopping rates equation (2) is different for upward and downward hops. The frequency of upward hops to the energy interval $[\varepsilon_j, \varepsilon_j+d\varepsilon_j]$ per unit system volume and energy, is

$$f_{up}(\varepsilon_j)\,d\varepsilon_j = g(\varepsilon_j)\,d\varepsilon_j \times \int_{-\infty}^{\varepsilon_j} d\varepsilon_i \int_0^{\infty} dr\, 4\pi r^2\, g(\varepsilon_i)\, P_B(\varepsilon_i)\, \nu(\varepsilon_i, \varepsilon_j, r). \quad (15)$$

The integral over r can be evaluated as

$$\int_0^{\infty} dr\, 4\pi r^2\, e^{-2r/\alpha} = \pi\alpha^3. \quad (16)$$

By inserting the Gaussian DOS $g(\varepsilon)$ from equation (1), we obtain

$$f_{up}(\varepsilon_j) = \frac{\pi\nu_0 \mathcal{N} N\alpha^3}{2} \times g(\varepsilon_j)e^{-\varepsilon_j/kT}\left[1 + \text{erf}\left(\frac{\varepsilon_j}{\sqrt{2}\,\sigma}\right)\right]. \quad (17)$$

Similarly, by adjusting the limits of the energy integral in equation (15) and using the right form of the hopping rates $\nu(\varepsilon_i, \varepsilon_j, r)$ (see equation (2)), the frequency of downward hops arriving at ε_j can be evaluated as

$$f_{dn}(\varepsilon_j) = \frac{\pi\nu_0 \mathcal{N} N\alpha^3}{2} \times g(\varepsilon_j)e^{\sigma^2/2(kT)^2}\left[1 - \text{erf}\left(\frac{\varepsilon_j + \sigma^2/kT}{\sqrt{2}\,\sigma}\right)\right]. \quad (18)$$

The total frequency of hops to the energy ε_j is obtained by adding the frequencies of upward and downward hops

$$f(\varepsilon_j) = f_{up}(\varepsilon_j) + f_{dn}(\varepsilon_j). \quad (19)$$

The energy ε_{peak}, which maximizes $f(\varepsilon_j)$, is our desired quantity, the most frequently visited energy. It has previously been calculated in Monte Carlo simulations [37, 42]. In addition to the peak energy of $f(\varepsilon_j)$, we also calculate its average energy,

$$\langle \varepsilon_j \rangle = \int_{-\infty}^{\infty} \varepsilon_j f(\varepsilon_j)\,d\varepsilon_j \,/\, \int_{-\infty}^{\infty} f(\varepsilon_j)\,d\varepsilon_j. \quad (20)$$

Both $\langle \varepsilon_j \rangle$ and ε_{peak} are determined numerically.

The two energies ε_{peak} and $\langle \varepsilon_j \rangle$ turn out to be very close to each other, since the distribution of $f(\varepsilon_j)$ is almost symmetric. This will become clear from the results in figure 3. We prefer $\langle \varepsilon_j \rangle$ as compared to ε_{peak}, since the average is easier to determine in a simulation. Furthermore, a numeric evaluation of equation (20) shows that
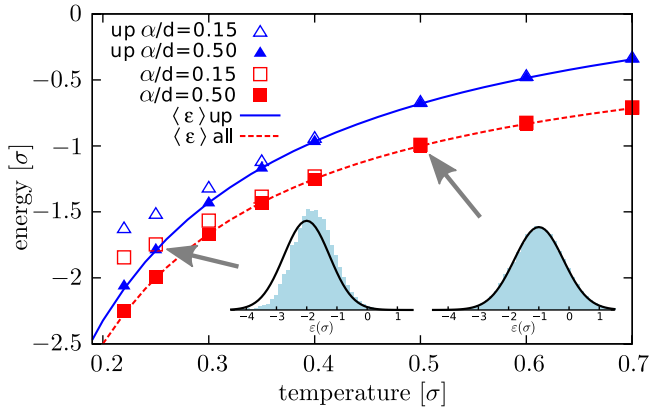
Figure 3. The average visited energy (squares) as a function of temperature, obtained by Monte Carlo simulation in a system with 10^6 randomly placed sites at zero electric field. The triangles show the average energy of upward hops. The curves show the predicted average of the visited energies equation (20) and the corresponding prediction for upward hops. The insets show histograms of the number of visits as a function of energy, for $\alpha = 0.15\,d$ at two different temperatures (arrows). The curves show the function $f(\varepsilon_j)$, given by equation (19).

$$\langle \varepsilon_j \rangle \approx \frac{-\sigma^2}{2kT}, \tag{21}$$

to an accuracy of 10 digits. Note that $f(\varepsilon_j)$ depends on the localization length α only via the prefactor α^3. This means that both ε_{peak} and $\langle \varepsilon_j \rangle$ are independent of α, which is a first hint to why this approach is insufficient for transport energy calculations.

The integral in equation (16) reveals the hops contributing mostly to the frequency of visits. The integrand has a maximum at $r = \alpha$, which means that hops of this length are the most important. Thus the most frequently visited energy is determined by pairs of spatially close sites, separated by the localization length α which is much smaller than the typical site separation. Such pairs are not important for the charge carrier mobility, [8, 47] since a charge carrier cannot move any significant distance using only such pairs.

In order to verify the results of the calculation above, we perform a Monte Carlo computer simulation of VRH and calculate the average of the visited energies. The algorithm is similar to that of Hartenstein and Bässler, [37] except that we consider sites randomly placed in space rather than distributed on a lattice. The average of the visited energies is shown in figure 3. The average over all upward hops is also shown since the transport energy is usually derived taking into account only upward hops in the hopping-rate optimization approach (see section 3). The insets in figure 3 show histograms of the number of visits to different energy intervals and a comparison with the function $f(\varepsilon_j)$. Good agreement is seen between the simulation results and equation (20), except for the small localization length at low temperatures. The latter is due to the finite-size effect, which comes in play at small localization lengths. In order to warrant representative results, sites in the low-energy tail of the DOS must not only be present in the system, but be present in pairs separated by the distance of

the order of α. This is apparently not the case in the left-hand histogram, where the number of visits in the low-energy part of the histogram is lower than predicted.

The data was obtained for a single electron hopping in an otherwise empty system. The electron was started from a random position and allowed to perform 10^8 hops; this procedure was repeated 1000 times. No electric field was applied. The simulation was repeated for five realizations of the disorder and the results were averaged. The hops in the simulation were restricted to $r_{cut} = 3d$, where $d = N^{-1/3}$ gives the typical site separation. Increasing the cut-off length r_{cut} did not alter the results.

In summary, the *most frequently visited energy* in the GDM can be determined analytically. The result agrees with those of a Monte Carlo simulation, yielding that the most frequent hops lead to an energy in the vicinity of $-\sigma^2/2kT$. However, these hops are much shorter than the typical distance between sites responsible for the charge carrier mobility. Therefore, the most frequently visited energy cannot be considered as relevant for charge transport and cannot serve as a transport energy.

Very recently, Mendels and Tessler [44] have attempted to overcome the problem by removing oscillatory hops between adjacent sites *before* the statistical analysis. Although the resulting distribution peaks are significantly shifted towards higher energies as compared to statistics including oscillations, it is not sufficient to eliminate cycles between only two sites in order to mimic the long-range hopping transport. In fact, the approach of tracing the most frequently visited energy becomes useful only when *all* closed cycles in the paths of charge carriers are removed before the statistical analysis.

However, removing all closed cycles from a directed graph is a very computationally demanding task, especially considering the large amount of data that is needed for a sufficiently accurate numerical study of the long-range hopping transport.

In the following Section, we suggest another approach which allows one to determine the transport energy using computer simulations. In that approach, the TE will be deduced from the results of carrier mobility and is therefore directly related to the long-range charge transport. It will be shown, that the transport energy is situated much higher in the DOS than $-\sigma^2/2kT$ given by equation (20).

5. How to find the transport energy in numerical simulations

We suggest to determine the TE by studying the effect of a DOS modifications on the charge carrier mobility. Thereby, it is ensured that the determined energy range is decisive for long-range transport. The idea is to cut out sites with energies in an interval $[\varepsilon_{cut}-w, \varepsilon_{cut}]$ from the system and to check, whether and how the resulting mobility, determined in straightforward computer simulations, is affected by such a modification of the DOS.

In figure 4, the corresponding modification is depicted. Sites in the interval $[\varepsilon_{cut}-w, \varepsilon_{cut}]$ are cut out from the DOS and are hence unaccessible for carriers during charge transport. Charge carriers must avoid the sites with energies in this
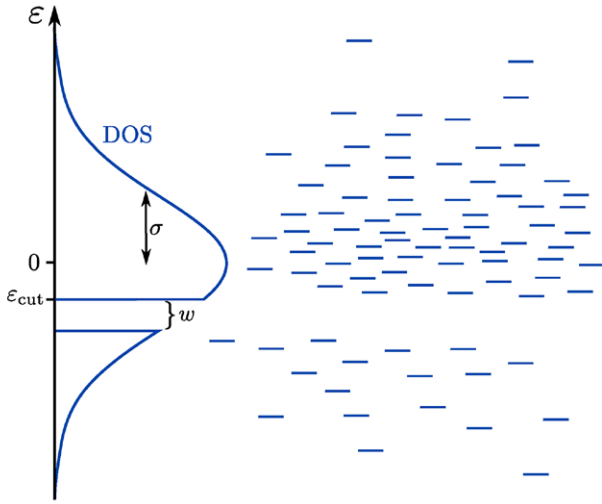
Figure 4. Schematic picture of the modified DOS $g(\varepsilon)$. Sites with energies in the interval $[\varepsilon_{cut}-w, \varepsilon_{cut}]$ are removed from the system and therefore not available for charge transport.
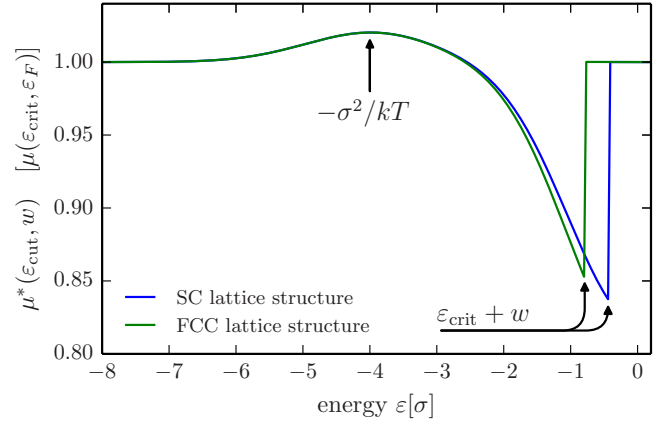

Figure 5. The mobility value of the modified DOS relative to the unmodified one as a function of the upper cutout interval boundary. Temperature $kT = 0.25\sigma$ and the cutout interval width $w = 0.05\sigma$. Evaluation is stopped when $\varepsilon_{crit} < \varepsilon_{cut} -w$, hence the discontinuity in the curves.

region and find other transport paths. Note, that the DOS is *not* renormalized after the modification. Depending on how important the withdrawn sites are for charge transport, the cutting will affect the mobility of the system more or less severely. We expect the highest drop in the carrier mobility when the cut-out sites have been those of highest importance for the long-range transport.

In order to justify the suggested method, let us check it by application to the exactly solvable hopping model, namely, the nearest-neighbor hopping (NNH) on a lattice. We assume a system of localized states distributed in an fcc or sc lattice structure. The site energies follow a Gaussian DOS. Charge carriers with finite concentration move via nearest neighbor transitions with Miller–Abrahams hopping rates given by equation (2). Percolation theory provides an exact analytical expression for the carrier mobility in this system [48]:

$$\mu(\varepsilon_{crit}, \varepsilon_F) = \mu_0 [\, g(\varepsilon_{crit})\,]^\lambda \exp\left(\frac{\varepsilon_F - \varepsilon_{crit}}{kT} \right), \qquad (22)$$

where μ_0 and λ do not depend on the DOS. The critical energy ε_{crit} is obtained from the DOS via equation (3). Since equation (22) can be applied to any DOS function, it provides a perfect tool to test the approach of searching for the TE by DOS modifications.

In order to perform such a test, let us cut an interval $[\varepsilon_{cut}-w, \varepsilon_{cut}]$ from the Gaussian DOS and check how it affects the carrier mobility. The modified DOS vanishes in the chosen energy interval:

$$g^*(\varepsilon) = \begin{cases} 0 & \text{if } \varepsilon \in [\varepsilon_{cut} - w, \varepsilon_{cut}], \\ g(\varepsilon) & \text{otherwise.} \end{cases} \qquad (23)$$

Both ε_{crit} and ε_F in equation (22) depend on the density of states and therefore need to be recalculated. Since the percolation threshold x_c and the carrier concentration n are independent of the shape of the DOS, we can use the following equations to determine the values ε_{crit}^* and ε_F^* for the modified

DOS from the values ε_{crit} and ε_F valid for the unmodified Gaussian DOS:

$$x_c = \int_{-\infty}^{\varepsilon_{crit}} g(\varepsilon)\,d\varepsilon = \int_{-\infty}^{\varepsilon_{cut}-w} g(\varepsilon)\,d\varepsilon + \int_{\varepsilon_{cut}}^{\varepsilon_{crit}^*} g(\varepsilon)\,d\varepsilon \qquad (24)$$

$$n = \int_{-\infty}^{\infty} g(\varepsilon) f(\varepsilon, \varepsilon_F)\,d\varepsilon \qquad (25)$$

$$= \int_{-\infty}^{\varepsilon_{cut}-w} g(\varepsilon) f(\varepsilon, \varepsilon_F^*)\,d\varepsilon + \int_{\varepsilon_{cut}}^{\infty} g(\varepsilon) f(\varepsilon, \varepsilon_F^*)\,d\varepsilon$$

In the above equations, the cut-out interval $[\varepsilon_{cut}-w, \varepsilon_{cut}]$ was simply removed from the integration ranges. Inserting the values for ε_{crit}^* and ε_F^* into equation (22) one obtains the mobility dependent on the cutout interval:

$$\mu^*(\varepsilon_{cut}, w) = \mu(\varepsilon_{crit}^*, \varepsilon_F^*). \qquad (26)$$

By comparing this result to that for the unmodified DOS one can study the effect of the DOS modification on the carrier mobility.

In figure 5, the ratio $\mu^*(\varepsilon_{cut}, w)/\mu(\varepsilon_{crit}, \varepsilon_F)$ is plotted as a function of the upper boundary of the cutout interval, ε_{cut}. The interval width w is fixed at $w = 0.05\sigma$ and results for two different lattice structures, *fcc* and *sc* are depicted.

The figure clearly demonstrates that the mobility drops significantly when cutting out sites in the vicinity of the critical energy ε_{crit}. This is the expected result.

There is another interesting peculiarity demonstrated by the curves in figure 5: when the cutout interval overlaps with the energy $\varepsilon_\infty = -\sigma^2/kT$ the carrier mobility increases. This happens for the following reason. In the Gaussian DOS and in thermal equilibrium most carriers occupy sites around the so-called equilibration energy ε_∞. Removing sites around this energy pushes carriers to higher energies and hence decreases the activation energy necessary for activation to the transport path, which leads concomitantly to the increase of the carrier mobility.
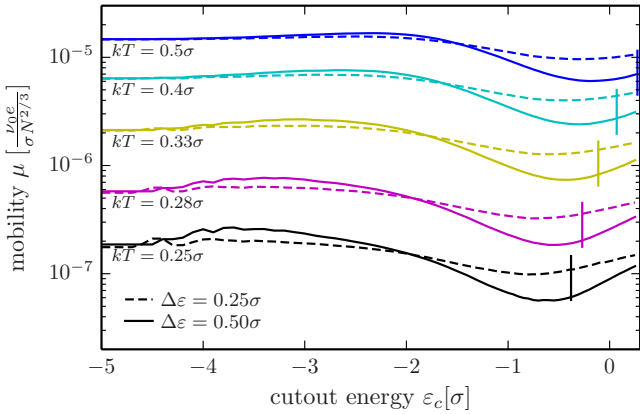
Figure 6. The mobility in a system with absent sites in the interval $[\varepsilon_{\mathrm{cut}}-w, \varepsilon_{\mathrm{cut}}]$. The vertical lines next to the minimum of the mobility are the analytical results from equation (13).

The above results, obtained by the exactly solvable model of nearest-neighbor hopping on a lattice, demonstrate the validity of the suggested approach for revealing the position of the TE: To cut out sites with particular energies and check the effect of such a modification on the calculated carrier mobility. Below we apply this approach to the problem relevant to the charge transport in real systems, namely to the VRH in the Gaussian DOS.

In order to study the effect of the DOS modification for the case of the VRH, we calculate the charge carrier mobilities numerically using the balance equation (BE) approach. [49, 50] By varying the width w and the upper boundary $\varepsilon_{\mathrm{cut}}$ of the cutout interval (see figure 4), one can find at which w and $\varepsilon_{\mathrm{cut}}$ the maximal effect in the decrease of the carrier mobility is achieved, thereby revealing the energy range most important for charge transport. The results are presented in figure 6. Results for two different widths, $w = 0.25\sigma$ and $w = 0.5\sigma$ at different temperatures kT are compared. In all cases, motion of a single charge carrier in an otherwise empty system was simulated. The localization length was chosen as $\alpha = 0.215\,N^{-1/3}$ and the number of sites in the simulated system was equal to 90^3.

It is clearly visible in the figure, that a significant decrease in the resulting mobility appears in each of the curves for a certain energy range of withdrawn sites. The effect is larger for larger energy intervals w, yet more accurate for lower values of w, since in that case the system is affected less severely. The choice of the interval width w should therefore be a compromise between the visibility of the effect and the accuracy in determining the position of the most efficient $\varepsilon_{\mathrm{cut}}$. We interpret the minima on the curves plotted in figure 6 as pointing at the position of the real transport energy (TE) responsible for the long-range charge transport. The position of the TE, as determined from figure 6, shifts upwards in energy with rising temperature. This agrees with all previous analytical and numerical studies. The vertical lines close to the minima of the mobility curves in figure 6 show the positions of the TE calculated from equation (13). Apparently, the optimization approach leading to equation (13) (outlined in section 3) is supported by the results of the straightforward computer simulations of the carrier mobility presented in figure 6.

Figure 6 also shows, that when sites in the vicinity of the average carrier energy $\varepsilon_\infty = -\sigma^2/kT$ are cut out, the mobility is slightly increased. This is literally the effect discussed above with respect to the exactly solvable model of the nearest-neighbor hopping on a lattice; It is clearly visible in figure 5. When sites with energies in the vicinity of the equilibration level $\varepsilon_\infty = -\sigma^2/kT$ are removed from the system, the average carrier energy increases, which diminishes the activation energy to the TE and leads to an increase of the mobility.

For the given temperature range the minima of the $\mu(\varepsilon_{\mathrm{cut}})$ curves in figure 6, which we interpret as the real *transport energy* related to long-range charge transport, are situated close to the center of the DOS. It seems therefore correct to assume in a simplified approach that the TE coincides with the center of the Gaussian DOS, as was suggested before [1, 51]. It means that although charge carriers spend most time in states with energies around the equilibration energy $\varepsilon_\infty = -\sigma^2/kT$, transport takes place via sites with much higher energies than ε_∞.

The data in figure 6 shows another remarkable feature, namely that the removal of sites with energies in the vicinity of the energy $-\sigma^2/2kT$ does not affect the carrier mobility significantly. This shows that it is wrong to interpret the VRH charge transport in the Gaussian DOS as activation of carriers from some particular energy level $\varepsilon_{\mathrm{start}}$ to the transport energy ε_t. If the latter interpretation was correct, the observation of the mobility temperature dependence in the form of equation (8) with $C^2 \approx 1/2$ would yield the value $\varepsilon_{\mathrm{start}} \approx -\sigma^2/2kT$ for $\varepsilon_t \approx 0$. The fact that cutting sites with energies in the vicinity of the level $-\sigma^2/2kT$ does not affect the mobility means that this energy level does not play any significant role for charge transport. The temperature dependence of carrier mobility in the form of equation (8) is instead the result of the time averaging over the upward hops to the TE from deeper energy levels [2, 31].

6. Energy level responsible for non-equilibrium energy relaxation

While it is well established that the percolation nature of the VRH plays a decisive role for long-range conductivity [8], it is not clear whether the percolation features have anything to do with the description of the energy relaxation of charge carriers towards the equilibrium distribution. It is worth noting that the concept of the TE was initially invented to study the relaxation behavior of charge carriers, showing particularly that these dynamics can be described in the framework of the MT model by replacing the mobility edge by the TE [23]. The MT model, in its turn, was initially developed [16–18] also in order to describe the energy relaxation of the non-equilibrium carrier distribution in systems with a mobility edge. During the relaxation phase, before thermal equilibrium is achieved, the carrier mobility depends on time, which is the essence of the so-called *dispersive transport*. In the GDM, dispersive transport takes place before charge carriers have relaxed in energy either to the equilibration energy $\varepsilon_\infty = -\sigma^2/kT$, if $\varepsilon_\infty > \varepsilon_{\mathrm{F}}$, or to the Fermi energy, if $\varepsilon_\infty < \varepsilon_{\mathrm{F}}$.

Let us consider the case of small carrier concentrations n so that $\varepsilon_\infty > \varepsilon_{\mathrm{F}}$. Baranovskii *et al* [31] estimated the time τ_{rel} necessary for carriers to achieve thermal equilibrium around

$\varepsilon_\infty = -\sigma^2/kT$ in the Gaussian DOS: They calculated $\tau_{\rm rel}$ as the time needed for activation from ε_∞ towards the level $\varepsilon_t^{\rm rel}$, calculated without taking percolation arguments into account [25, 31]. This led to a temperature dependence of $\tau_{\rm rel}$ in the form [31]

$$\tau_{\rm rel} \propto \exp\left[\left(B\frac{\sigma}{kT}\right)^2\right], \qquad (27)$$

with $B \approx 1$ resolving the puzzle about the difference between the values of the coefficients $C^2 \simeq 1/2$ and $B \simeq 1$ in equations (8) and (27), respectively, raised by the results of computer simulations [1, 12, 52, 53]. The quantity $\varepsilon_t^{\rm rel}$ was calculated [25, 31] via equation (13) with $B_c = 1$. Apparently, the level $\varepsilon_t^{\rm rel}$ for the energy relaxation of carriers does not coincide with the TE ε_t responsible for the long-range transport calculated with $B_c \simeq 2.7$.

A similar result has recently been obtained by Germs *et al* [54], who performed Monte Carlo (MC) simulations of the carrier energy relaxation in the GDM and found the position of the energy level $E_c^{\rm MC}$ that would describe numerical results in the framework of the MT model with replacing the mobility edge by $E_c^{\rm MC}$. It was found that $E_c^{\rm MC}$ depends on the ratio σ/kT, i.e., depends on temperature T for a given value σ. Although it is not specified explicitly, one can guess that Germs *et al* [54] studied the nearest-neighbor hopping (NNH) process on a lattice. Otherwise the localization length α, not at all specified in the paper by Germs *et al* [54], would have come in play as an additional decisive parameter. On the other hand, equation (3), that determines the position of the TE E_c responsible for the long-range transport in the NNH on a lattice, depends only on the energy scale σ of the DOS and not on temperature T. This clearly illustrates the difference between $E_c^{\rm MC}$ and E_c. It would be desirable to extend the study from the oversimplified model of the NNH on a lattice, that yields the transport energy E_c dependent on the choice of the lattice structure [13], to a more realistic problem of the VRH in the GDM.

Assuming carrier relaxation in the VRH, the interesting question is whether or not to include the percolation nature of transport in such considerations, e.g., when calculating the transport energy ε_t to replace the mobility edge in the MT model. If the average spatial displacement of carriers during relaxation, i.e., before they reach the equilibration energy ε_∞, exceeds the correlation length $\mathcal{L}$ of the percolating cluster [8, 14], percolation criteria are expected to be necessary for the description of the relaxation dynamics; in case the displacement is smaller than $\mathcal{L}$, percolation should probably not play a significant role in the relaxation properties. This topic exceeds the scope of the current report and remains an open question.

7. Conclusions

A new numerical approach is suggested to determine the transport path of hopping charge carriers in a steeply decreasing DOS. The approach is based on simulating the effect of a DOS modification on charge transport properties. The method is applied to the Gaussian DOS relevant for organic disordered semiconductors. Our numerical calculations confirm the

validity of the analytical approach to calculate the transport energy via optimization of hopping rates [22].

It is also rigorously proven by analytical calculations and numerical simulations that algorithms based on tracing the energies of the most frequently targeted localized states, although widely applied, are not suitable to determine the position of the transport energy.

References

[1] Bässler H 1993 *Phys. Status Solidi* B **175** 15
[2] Oelerich J O, Huemmer D and Baranovskii S D 2012 *Phys. Rev. Lett.* **108** 226403
[3] Baranovskii S D 2014 *Phys. Status Solidi* B **251** 487–25
[4] Bässler H 1981 *Phys. Status Solidi* B **107** 9
[5] Miller A and Abrahams E 1960 *Phys. Rev.* **120** 745–55
[6] Gill W D 1972 *J. Appl. Phys.* **43** 5033
[7] Rubel O, Baranovskii S D, Thomas P and Yamasaki S 2004 *Phys. Rev.* B **69** 014206
[8] Shklovskii B I and Efros A L 1984 *Electronic Properties of Doped Semiconductors* (Heidelberg: Springer)
[9] Shklovskii B and Efros A 1971 *Sov. Phys.—JETP* **33** 468
[10] Ambegaokar V, Halperin B and Langer J 1971 *Phys. Rev.* B **4** 2612
[11] Pollak M 1972 *J. Non-Cryst. Solids* **11** 1–24
[12] Schönherr G, Bässler H and Silver M 1981 *Phil. Mag.* B **44** 369
[13] Cottaar J, Koster L J A, Coehoorn R and Bobbert P A 2011 *Phys. Rev. Lett.* **107** 136601
[14] Nenashev A V, Jansson F, Oelerich J O, Huemmer D, Dvurechenskii A V, Gebhard F and Baranovskii S D 2013 *Phys. Rev.* B **87** 235204
[15] Orenstein J and Kastner M A 1981 *Solid State Commun.* **40** 85–9
[16] Noolandi J 1977 *Phys. Rev.* B **16** 4466
[17] Schmidlin F W 1977 *Phys. Rev.* B **16** 2362
[18] Silver M and Cohen L 1977 *Phys. Rev.* B **15** 3276
[19] Rudenko A I and Arkhipov V I 1982 *Phil. Mag.* B **45** 177
[20] Arkhipov V I and Rudenko A I 1982 *Phil. Mag.* B **45** 189
[21] Nenashev A V, Jansson F, Baranovskii S D, Österbacka R, Dvurechenskii A V and Gebhard F 2009 *Phys. Rev.* B **81** 115204
[22] Oelerich J O, Huemmer D, Weseloh M and Baranovskii S D 2010 *Appl. Phys. Lett.* **97** 143302
[23] Monroe D 1985 *Phys. Rev. Lett.* **54** 146–9
[24] Baranovskii S D, Thomas P and Adriaenssens G J 1995 *J. Non-Cryst. Solids* **190** 283–87
[25] Baranovskii S D, Faber T, Hensel F and Thomas P 1997 *J. Phys. Condens. Matter* **9** 2699–706
[26] Grünewald M and Thomas P 1979 *Phys. Status Solidi* B **125** 125–33
[27] Vissenberg M C J M and Matters M 1998 *Phys. Rev.* B **57** 12964
[28] Baranovskii S D, Zvyagin I P, Cordes H, Yamasaki S and Thomas P 2002 *J. Non-Cryst. Solids* **299** 416
[29] Zvyagin I P 2008 *Phys. Status Solidi* C **5** 725

[30] Baranovskii S D, Zvyagin I P and Cordes H 2002 *Phys. Status Solidi* B **287** 281–7
[31] Baranovskii S D, Cordes H, Hensel F and Leising G 2000 *Phys. Rev.* B **62** 7934
[32] Arkhipov V I, Emelianova E V and Adriaenssens G J 2001 *Phys. Rev.* B **64** 125125
[33] Nikitenko V R, von Seggern H and Bässler H 2007 *J. Phys.: Condens. Matter* **19** 136210
[34] Arkhipov V I, Heremans P, Emelianova E V, Adriaenssens G J and Bässler H 2003 *Appl. Phys. Lett.* **82** 3245
[35] Schmechel R 2002 *Phys. Rev.* B **66** 235206
[36] Martens H C F, Hulea I N, Romijn I, Brom H B, Pasveer W F and Michels M A J 2003 *Phys. Rev.* B **67** 121203
[37] Hartenstein B and Bässler H 1995 *J. Non-Cryst. Solids* **190** 112–6
[38] Tanase C 2005 *PhD Thesis* University of Groningen
[39] Fumagalli L, Binda M, Natali D, Sampietro M, Salmoiraghi E, Gianvincenzo P D and Di Gianvincenzo P 2008 *J. Appl. Phys.* **104** 084513
[40] Cleve B, Hartenstein B, Baranovskii S D, Scheidler M, Thomas P and Bässler H 1995 *Phys. Rev.* B **51** 16705
[41] Novikov S V and Malliaras G G 2006 *Phys. Status Solidi* B **243** 387
[42] Gonzalez-Vazquez J P, Anta J A and Bisquert J 2009 *Phys. Chem. Chem. Phys.* **11** 10359–67
[43] Arkhipov V I, Emelianova E V and Adriaenssens G J 2001 *Phys. Rev.* B **64** 125125
[44] Mendels D and Tessler N 2013 *J. Phys. Chem.* C **117** 24740
[45] Baranovskii S D (ed) 2006 *Charge Transport in Disordered Solids with Applications in Electronics* (Chichester: Wiley)
[46] Lorenz C D and Ziff R M 2001 *J. Chem. Phys.* **114** 3659
[47] Baranovskii S D and Thomas P 1996 *J. Non-Cryst. Solids* **198** 140–45
[48] Cottaar J, Coehoorn R and Bobbert P A 2012 *Phys. Rev.* B **85** 245205
[49] Yu Z G, Smith D L, Saxena A, Martin R L and Bishop A R 2000 *Phys. Rev. Lett.* **84** 721–24
[50] Cottaar J and Bobbert P A 2006 *Phys. Rev.* B **74** 115204
[51] Neumann F, Genenko Y A and von Seggern H 2006 *J. Appl. Phys.* **99** 013704
[52] Ries B, Bässler H, Grünewald M and Movaghar B 1988 *Phys. Rev.* B **37** 5508
[53] Bässler H 1990 *Transport and Relaxation of Excitations in Random Organic Solids Advances in Disordered Semiconductors* (Singapore: World Scientific) 491–520 (Monte Carlo Simulation and Experiment)
[54] Germs W C, van der Holst J J M, van Mensfoort S L M, Bobbert P A and Coehoorn R 2011 *Phys. Rev.* B **84** 165210