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Advanced percolation solution for hopping conductivity

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Hopping of carriers between localized states dominates charge transport in amorphous organic and inorganic semiconductors. We suggest a comprehensive description of this transport regime based on the percolation approach that allows one to determine not only very pronounced exponential dependencies of the hopping conductivity on material parameters, but also the more weakly dependent pre-exponential factors. The problem of the variable-range hopping (VRH) via sites with exponential energy distribution is mapped onto a universal geometrical problem of percolation via spheres with distributed sizes. An exact solution of the latter problem provides accurate results for the VRH in systems with exponential density-of-states (DOS). Our analytical results are confirmed by straightforward computer simulations and compared to former results present in the literature. We also discuss the case of nearest-neighbor hopping on a lattice, where the pre-exponential factors are provided by the percolation approach for any shape of the DOS.

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I. INTRODUCTION

Interest in optoelectronic properties of amorphous organic and inorganic semiconductors has been steeply growing in the last decades. This is caused by successful applications of such materials in various devices and by their promises for future applications. Manufacturability and low production costs of amorphous materials, along with their specific charge transport properties, make such materials favorable and in some cases unique for various applications, particularly for large-area devices, where demands to the mobilities of charge carriers are not very high. Already at an early stage of the research on disordered semiconductors it was recognized that within wide ranges of parameters charge transport is dominated by incoherent tunneling transitions (hopping) of carriers between spatially localized states with a broad energy distribution. For the energy spectrum, also called the density of states (DOS), two shapes are usually discussed: (i) an exponential DOS,

$$g(\varepsilon) = \begin{cases} 0 & \text{when } \varepsilon > 0, \\ \frac{N}{\sigma} \exp(\varepsilon/\sigma) & \text{when } \varepsilon \leq 0, \end{cases} \quad (1)$$

is often assumed for inorganic amorphous materials,^{1,2} and (ii) a Gaussian DOS,

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

is assumed for organic disordered semiconductors,^{3,4} although the latter materials are sometimes claimed to possess an exponential DOS.^{5,6}

The parameter σ in Eqs. (1) and (2) characterizes the width of the DOS varying from $\sigma \simeq 0.025$ eV for inorganic amorphous materials² to $\sigma \simeq 0.1$ eV for organic systems.³ In this report the hopping of carriers through randomly distributed sites with concentration N will be considered. For the sake of simplicity, no correlations between positions and energies of sites will be assumed.

The rate Γ_{ij} of carrier transitions from an occupied site i to an empty site j , separated by the distance d_{ij} , is assumed to follow the Miller-Abrahams expression,

$$\Gamma_{ij} = v_0 \exp\left[-\frac{2d_{ij}}{a} - \frac{\max(\Delta\varepsilon_{ij}, 0)}{kT}\right], \quad (3)$$

where v_0 is the attempt-to-escape frequency, a is the localization length, k is the Boltzmann constant, T is temperature, and $\Delta\varepsilon_{ij} = \varepsilon_j - \varepsilon_i$ is the difference between carrier energies ε_j and ε_i on sites j and i , respectively. The latter quantities depend on the applied electric field.

Theoretical studies of hopping transport in disordered semiconductors are often performed by empirical fitting of numerical results, which leads to analytical equations that can hardly be valid in a broad range of parameters. This is particularly true for organic materials, for which analytical studies are believed to be very difficult.³ The aim of the current report is to present an analytical theoretical approach that allows one to determine not only the very pronounced exponential dependencies of carrier mobility on various material parameters, but also the more weakly dependent pre-exponential factors. This approach is based on classical percolation theory.

The percolation approach for the description of variable-range-hopping (VRH) has been known for about 40 years.⁷⁻⁹ Grünewald and Thomas¹⁰ used this approach for studying hopping transport in the exponential DOS given by Eq. (1). Almost two decades later, the same problem in the framework of the percolation approach was addressed by Vissenberg and Matters,⁵ who used a somewhat different percolation criterion than Grünewald and Thomas.¹⁰ Since no comparison with the results of Grünewald and Thomas has been performed by Vissenberg and Matters, it is not clear which percolation approach is correct or more accurate.

In Sec. II it is shown that the VRH problem in the exponential DOS can be mapped onto a purely geometrical problem that allows a numerical solution described in Sec. III.

83 This numerical solution provides results for the conductivity
84 of the system and for the carrier mobility, accurate up to a
85 dimensionless numerical coefficient. The value of this coeffi-
86 cient is determined via straightforward simulations described
87 in Sec. IV. In Sec. V the new analytical approach is compared
88 to previous results from the literature. It is shown that the
89 result of Grünwald and Thomas¹⁰ is more accurate than that
90 of Vissenberg and Matters.⁵ Furthermore, the obtained exact
91 solution for the exponential DOS is used to check in Sec. V
92 the validity of the recently suggested theoretical approach for
93 calculating hopping mobilities.⁴

94 The percolation approach to hopping transport in the
95 Gaussian DOS [Eq. (2)]^{11,12} appears less transparent than in
96 the case of an exponential DOS. For the Gaussian DOS the
97 exact mapping on a purely geometrical model is not possible.
98 However, in the case of carrier hopping via neighboring sites
99 on a regular lattice, suggested recently in the frame of empirical
100 fitting for disordered organic semiconductors,¹³ a theoretical
101 approach similar to that described in Sec. II is feasible also
102 for a Gaussian DOS. This approach is presented in Sec. VI.
103 Concluding remarks are given in Sec. VII.

104 II. VRH IN THE EXPONENTIAL DOS

105 The standard method for studying the problem of hopping
106 transport in small electric fields (within the Ohmic regime) is
107 the method of a resistance network.^{7-9,14,15} In this method,
108 a resistance R_{ij} is assigned to each pair of sites with
109 nonvanishing hopping rate Γ_{ij} given by Eq. (3). The expression
110 for R_{ij} becomes especially simple when the Fermi level ε_F is
111 far below the site energies $\varepsilon_i, \varepsilon_j$. In this case,

$$R_{ij} = \frac{kT}{e^2 v_0} \exp \left[\frac{2d_{ij}}{a} + \frac{\max(\varepsilon_i, \varepsilon_j) - \varepsilon_F}{kT} \right]. \quad (4)$$

112 We will assume that the carrier concentration is low enough,
113 so that Eq. (4) holds for all pairs of sites significant for the
114 conductivity. The quantitative meaning of this “low enough
115 concentration” constraint will be described below. Also, low
116 temperatures will be considered, $kT \ll \sigma$.

117 Since the resistances R_{ij} are distributed in an exponentially
118 broad region, the percolation approach can be used for
119 determining the net resistivity. The main part of this approach
120 consists of seeking the *critical* value R_{crit} of the resistance,
121 i.e., the value at which the opposite sides of a sample become
122 connected, when resistances are “switched on” in ascending
123 order. The system of connected site with bond resistances R_{ij}
124 defined by the bonding criterion

$$R_{ij} \leq R_{\text{crit}} \quad (5)$$

125 is at the percolation threshold, by definition of R_{crit} . It is
126 convenient to write R_{crit} in a way similar to Eq. (4),

$$R_{\text{crit}} = \frac{kT}{e^2 v_0} \exp \left(\frac{\varepsilon^* - \varepsilon_F}{kT} \right), \quad (6)$$

127 introducing a “critical energy” ε^* . The bonding criterion (5)
128 can be expressed via ε^* as

$$\frac{2d_{ij}}{a} + \frac{\varepsilon_i}{kT} \leq \frac{\varepsilon^*}{kT}, \quad \frac{2d_{ij}}{a} + \frac{\varepsilon_j}{kT} \leq \frac{\varepsilon^*}{kT}. \quad (7)$$

Since $d_{ij} > 0$, only sites with energies $\varepsilon < \varepsilon^*$ can have
connections satisfying the criterion (7). The concentration N^*
of such sites is

$$N^* = \int_{-\infty}^{\varepsilon^*} g(\varepsilon) d\varepsilon = N \exp \left(\frac{\varepsilon^*}{\sigma} \right). \quad (8)$$

This implies that $\varepsilon^* < 0$, which puts a constraint on the density
 N discussed below. Introducing for each site i with $\varepsilon_i < \varepsilon^*$ a
“radius” r_i according to

$$r_i = \frac{a}{2kT} (\varepsilon^* - \varepsilon_i), \quad (9)$$

one can express the bonding criterion in a purely geometrical
form:

$$d_{ij} \leq r_i, \quad d_{ij} \leq r_j. \quad (10)$$

One can find the distribution function $g(r)$ of the “radii” of
sites, using the relation $|g(r) dr| = |g(\varepsilon) d\varepsilon|$:

$$g(r) = \frac{g(\varepsilon)}{|dr/d\varepsilon|} = \frac{N}{\sigma} \exp(\varepsilon/\sigma) \frac{2kT}{a}. \quad (11)$$

Expressing ε via r in accordance with Eq. (9) and using Eq. (8)
one obtains

$$g(r) = \frac{N^*}{L^*} \exp(-r/L^*), \quad (12)$$

where the quantity

$$L^* = \frac{a\sigma}{2kT} \quad (13)$$

has the meaning of the average “radius.”

Since L^* represents a natural length scale for the “radii,” it
is convenient to measure d_{ij} and r_i in units of L^* , introducing
dimensionless quantities

$$\tilde{d}_{ij} = d_{ij}/L^*, \quad \tilde{r}_i = r_i/L^*. \quad (14)$$

Then, the bonding criterion becomes

$$\tilde{d}_{ij} \leq \tilde{r}_i, \quad \tilde{d}_{ij} \leq \tilde{r}_j, \quad (15)$$

and the distribution of “dimensionless radii” becomes expo-
nential with the mean value 1:

$$g(\tilde{r}) = n \exp(-\tilde{r}), \quad (16)$$

where n is a dimensionless concentration of sites having
positive “radii,”

$$n = N^*(L^*)^3. \quad (17)$$

Now, it will be shown that n does not depend on physical
parameters (such as σ, T, a , etc.), and has a universal value. For
this purpose, a geometrical percolation problem is formulated
that will provide the desired quantity. Consider a set of
spheres of different radii in an infinite three-dimensional space
[Fig. 1(a)]. The distribution of their radii is exponential with
the mean value 1. Centers of spheres are randomly placed,
uncorrelated with their radii. The average number of spheres
per unit cube is n . If the center of sphere i lies inside the
sphere j , and simultaneously the center of sphere j lies inside
the sphere i , then we call these spheres connected to each other
[Fig. 1(b)]. If there is a path from one sphere to another one via
a connected pairs of spheres, we call these spheres belonging
to the same cluster. The percolation problem is to find the

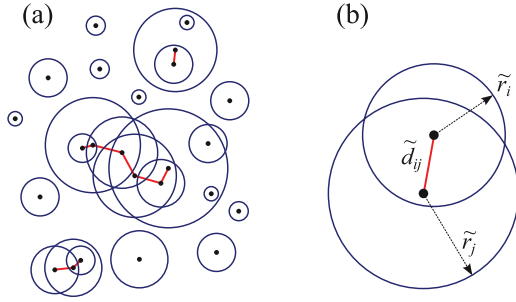


FIG. 1. (Color online) The system of spheres of different radii (a) and the connection criterion (b). Lines join centers of connected spheres.

critical concentration n_c such that there exists no infinite cluster when $n < n_c$ and there is an infinite cluster when $n > n_c$. The numerical solution of this problem with the result $n_c \approx 0.219$ is described in Sec. III.

It is easy to see that the dimensionless concentration n defined by Eq. (17) is equal to n_c . Indeed, consider sites with energies $\varepsilon_i < \varepsilon^*$, using L^* as a unit of length, and for each of such sites i draw a sphere with radius \tilde{r}_i centered at this site. According to Eq. (16), the distribution of the spheres' radii is exponential with the mean value of unity. The concentration of spheres is n . Since the distance between spheres i and j is \tilde{d}_{ij} , the bonding criterion given by Eq. (15) is equivalent to these spheres being connected as shown in Fig. 1(b). Therefore, the system of connected resistors with bonding criterion Eq. (5) perfectly maps onto the system of connected dimensionless spheres described above. Moreover, by definition of R_{crit} , the system of resistors is exactly at the percolation threshold, hence the same holds true for the corresponding system of spheres, which means that the concentration of spheres n is equal to the critical one:

$$n = n_c \approx 0.219. \quad (18)$$

Using Eqs. (8), (13), (17), and (18), one obtains the following equation for the (yet unknown) “critical energy” ε^* :

$$N \exp\left(\frac{\varepsilon^*}{\sigma}\right) \left(\frac{a\sigma}{2kT}\right)^3 = n_c. \quad (19)$$

For the case of low temperatures, the Fermi energy ε_F is related to the carrier concentration n_e via

$$\int_{-\infty}^{\varepsilon_F} g(\varepsilon) d\varepsilon = N \exp\left(\frac{\varepsilon_F}{\sigma}\right) = n_e. \quad (20)$$

Expressing ε^* and ε_F from the last two equations, and substituting their values into Eq. (6), one obtains the following result for the critical resistance R_{crit} :

$$R_{\text{crit}} = \frac{kT}{e^2 v_0} \left[n_c \left(\frac{2kT}{a\sigma}\right)^3 n_e^{-1} \right]^{\sigma/kT}. \quad (21)$$

Due to its exponential form given by Eq. (6), the critical resistance R_{crit} represents the most strongly varying factor in the volume resistivity ρ as a function of the model parameters. The other (pre-exponential) factors in ρ are more slowly varying. Nevertheless, the latter are also important,

primarily because they provide the correct dimensionality of ρ . Therefore, the resistivity ρ must be calculated, taking into account also the pre-exponential factors.

As explained in Section 5.6 of the monograph by Shklovskii and Efros,¹⁶ the conductivity of a strongly inhomogeneous medium is defined almost completely by the so-called *critical subnetwork*. This subnetwork is an infinite cluster formed by resistances R_{ij} that are smaller than *or comparable with* the critical value R_{crit} :

$$R_{ij} \leq R_{\text{crit}} \exp(\delta), \quad (22)$$

where δ is a quantity of order unity which we can consider as a constant.

In order to obtain the resistivity of the resistor network, cut out from the resistor network a cube of some given size L and consider the resistance $R(L)$ between opposite faces of this cube. There are two limiting cases depending on how L compares with L_{corr} , the correlation length of the critical subnetwork. If $L \gg L_{\text{corr}}$, then the cube content can be treated as a continuous medium, yielding

$$R(L) = \rho/L, \quad \text{if } L \gg L_{\text{corr}},$$

where ρ is the macroscopic resistivity. In the opposite case, $L \ll L_{\text{corr}}$, the cube resistance is determined by the “local” critical resistance \tilde{R}_{crit} , i.e., a resistance at which the opposite sides of the cube become connected when resistors are “switching on” in ascending order:

$$R(L) = \tilde{R}_{\text{crit}}, \quad \text{if } L \ll L_{\text{corr}}.$$

The two cases meet each other at $L \simeq L_{\text{corr}}$. Moreover, the values of \tilde{R}_{crit} are of the same order of magnitude as $R_{\text{crit}} \equiv R_0 \exp(\xi_c)$ for nearly all cubes of the size L_{corr} . Therefore

$$R(L_{\text{corr}}) \simeq \rho/L_{\text{corr}} \simeq R_{\text{crit}},$$

which gives

$$\rho \simeq L_{\text{corr}} R_{\text{crit}}. \quad (23)$$

Here and below we use the sign \simeq to indicate equality up to some dimensionless factor of order unity.

In order to find L_{corr} , it is useful to start from the bonding criterion for the critical subnetwork given by Eq. (22). Expressed via ε^* in Eq. (6), this criterion reads

$$\frac{2d_{ij}}{a} + \frac{\varepsilon_i}{kT} \leq \frac{\varepsilon^*}{kT} + \delta, \quad \frac{2d_{ij}}{a} + \frac{\varepsilon_j}{kT} \leq \frac{\varepsilon^*}{kT} + \delta. \quad (24)$$

The difference between Eq. (7) and Eq. (24) can be eliminated by the following increase of ε^* :

$$\varepsilon^* \rightarrow \varepsilon^* + \delta kT. \quad (25)$$

Therefore the renormalization (25) allows us to turn the consideration of the percolation threshold into a consideration of the critical network, while the derivation of Eqs. (8)–(17) remains unchanged. According to Eq. (13), the value of L^* for the critical subnetwork does not depend on ε^* , and therefore stays the same as for the percolation threshold. However, the values of N^* and n are increased by a factor $\exp(\delta kT/\sigma)$, because they depend on ε^* due to Eqs. (8) and (17). Hence, the dimensionless concentration $n_{\text{subnetwork}}$ of sites necessary to form the critical subnetwork is

$$n_{\text{subnetwork}} = n_c \exp(\delta kT/\sigma), \quad (26)$$

where n_c is the corresponding concentration at the percolation threshold.

It is known¹⁶ that the correlation length of the infinite cluster scales with the site concentration n as $(n - n_c)^{-\nu}$, where ν is the known critical index of the correlation length. This allows estimating the dimensionless correlation length of the critical subnetwork, L_{corr}/L^* , as

$$L_{\text{corr}}/L^* \simeq (n_{\text{subnetwork}} - n_c)^{-\nu}. \quad (27)$$

The value of ν depends only on the dimensionality of space. In 3D, $\nu = 0.875 \pm 0.008$.¹⁷ Since

$$n_{\text{subnetwork}} - n_c = n_c [\exp(\delta kT/\sigma) - 1] \simeq \frac{kT}{\sigma}, \quad (28)$$

it follows that

$$L_{\text{corr}} \simeq L^* \left(\frac{\sigma}{kT} \right)^\nu. \quad (29)$$

Note that the factor n_c in Eq. (28) is dropped in the Taylor expansion since all constant prefactors are described via a single fitting coefficient. Combining Eqs. (13), (23), and (29), one obtains the prefactor of the resistivity ρ :

$$\rho \simeq \frac{a\sigma}{kT} \left(\frac{\sigma}{kT} \right)^\nu R_{\text{crit}}. \quad (30)$$

Finally, substituting Eq. (21) for R_{crit} one obtains for the resistivity ρ :

$$\rho = A \frac{a\sigma}{e^2 v_0} \left(\frac{\sigma}{kT} \right)^\nu \left[8n_c \left(\frac{kT}{a\sigma} \right)^3 n_e^{-1} \right]^{\sigma/kT}, \quad (31)$$

where A is a numerical constant. This equation is the central result of the current report for the resistivity of a system with exponential DOS in the frame of the VRH that contains only one unknown dimensionless constant A . The latter is determined via fitting to computer simulation results described in Sec. IV.

The above results are applicable under the following conditions.

(i) The temperature T must be low enough:

$$kT \ll \sigma. \quad (32)$$

This requirement is used in Eqs. (27) and (28), where it is important that $(n_{\text{subnetwork}} - n_c) \ll 1$. It is also used for the evaluation of the Fermi energy ε_F via Eq. (20).

(ii) The electron concentration n_e should be low enough to guarantee that the Fermi level ε_F is far below the ‘‘critical energy’’ ε^* :

$$\left[8n_c \left(\frac{kT}{a\sigma} \right)^3 n_e^{-1} \right]^{\sigma/kT} \gg 1. \quad (33)$$

This condition allows one to use Eq. (4) for the resistances R_{ij} . One can treat condition (33) also as a restriction on temperature T from below. The condition is satisfied if the bracket in the l.h.s. is larger than unity, i.e., if $kT/\sigma > (n_e a^3 / 8n_c)^{1/3}$. Furthermore, one can show that condition (33) determines temperatures far above those, at which the well-known Mott law for VRH is expected: The Mott law is valid at temperatures satisfying the strong inequality $kT(T_0/T)^{1/4} \ll \sigma$, where $T_0 = 1/[kg(\varepsilon_F)a^3]$. Using the estimate $g(\varepsilon_F) \approx n_e/\sigma$, one obtains the condition $kT/\sigma \ll (n_e a^3)^{1/3}$ for the validity of Mott law.

Apparently, the latter condition is fulfilled at much lower temperatures than those determined by condition (33). In other words, the range of applicability for our theory determined by condition (33) does not overlap with the range of applicability for Mott VRH.

(iii) There should be enough sites for variable-range hopping:

$$N > 8n_c \left(\frac{kT}{a\sigma} \right)^3. \quad (34)$$

This requirement is used in Eq. (8), which implies that $\varepsilon^* < 0$. One can also consider it as a restriction on temperature, rewriting it as $kT/\sigma < (Na^3/8n_c)^{1/3}$. The latter condition does not contradict to condition (33) as long as the number of charge carriers is much smaller than the total number of localized states available for hopping transport: $n_e \ll N$.

Finally, a few remarks are in order for the two-dimensional case. There are a few differences between 2D and 3D VRH in a system with exponential DOS.

(i) The dimensionless concentration is $n = N^*(L^*)^2$ instead of $n = N^*(L^*)^3$ as in Eq. (17).

(ii) A different value $n_{c2} \approx 1.303$ of the critical dimensionless concentration applies, which is obtained numerically in Sec. III. (iii) $\rho \simeq R_{\text{crit}}$ must be used instead of $\rho \simeq L_{\text{corr}} R_{\text{crit}}$ in Eq. (23).^{18,19}

As a result, one obtains [instead of Eq. (31), valid in 3D] the following expression in 2D:

$$\rho = A_2 \frac{kT}{e^2 v_0} \left[4n_c \left(\frac{kT}{a\sigma} \right)^2 n_e^{-1} \right]^{\sigma/kT}, \quad (35)$$

where A_2 is a numerical constant.

III. PERCOLATION VIA SPHERES WITH DISTRIBUTED SIZES

The percolation threshold n_c was found using the algorithm described by Newman and Ziff.²⁰ According to this algorithm, spheres are generated randomly and added to the system one by one. For each sphere, one keeps track of the cluster to which it belongs. Initially, each sphere forms its own cluster. When a sphere added to the system is connected with already existing spheres, the clusters are merged. An efficient so-called union/find algorithm is used to keep track of the clusters.^{20,21} Spheres are added until a connected cluster appears that spans the system in a given direction. Periodic boundary conditions are used, so that the system is considered to percolate when a cluster wraps around the system in a given direction and connects to itself. The linked cell method²² is used to efficiently find the neighbors of each newly added sphere, in order to avoid iterating through a list of all spheres for every sphere that is added.

When determining the percolation threshold numerically, two kinds of errors affect the result. First, the concentration required for a spanning cluster will be different from one realization to the next. This error can be reduced by averaging over many realizations. The second error is a finite size effect: the critical concentration in a finite system depends²³ on the system size L ,

$$n_c(L) - n_c \propto L^{-1/\nu}. \quad (36)$$

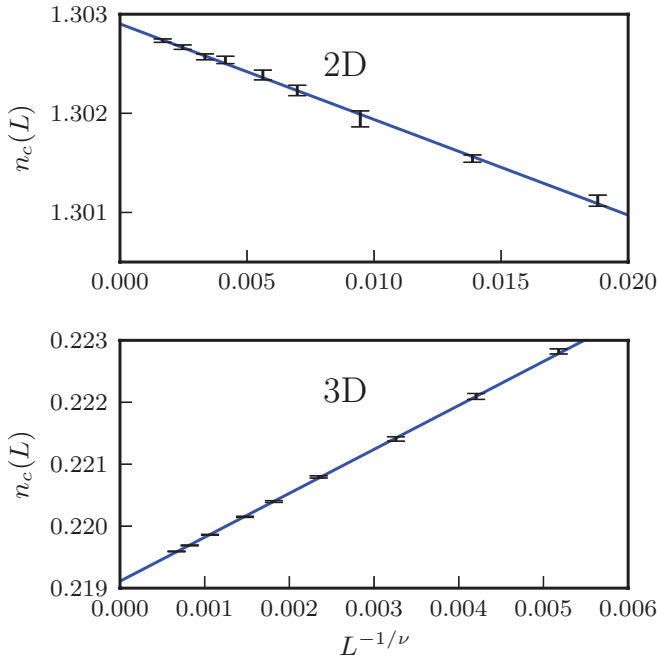


FIG. 2. (Color online) Finite size scaling of the percolation threshold n_c for disks (2D) and spheres (3D) with exponentially distributed radii. The scaling exponent ν is 0.875 in 3D,¹⁷ and $4/3$ in 2D.²³

To obtain the limit of infinite system size one can plot $n_c(L)$ against $L^{-1/\nu}$, perform a linear fit, and extrapolate n_c for $1/L \rightarrow 0$. Figure 2 shows these plots for percolation in two and three dimensions, and demonstrates that the scaling law (36) applies in both cases. Between 20 000 and 40 000 realizations for each system size L were evaluated in 3D, and over 70 000 for each L in 2D. By extrapolating to $1/L \rightarrow 0$, the critical concentration of spheres in 3D,

$$n_c \approx 0.219, \quad (37)$$

was found. In two dimensions, the corresponding critical concentration of disks is $n_{c2} \approx 1.303$. To verify the program, it was applied to the case of equally sized spheres, where the percolation threshold is accurately known,²⁴ $n_c^{\text{uni}} \approx 0.65296$. The known value was reproduced with an accuracy of four digits.

IV. STRAIGHTFORWARD SIMULATION OF THE VRH IN THE EXPONENTIAL DOS

In order to test the resistivity expression (31) derived in Sec. II, a numerical simulation of the charge transport was performed. The simulation also provides a value for the prefactor A in Eq. (31). The numerical results were obtained using the method of balance equations,^{25–28} linearized for the limit of a small electric field.²⁹ Details of the procedure are given in the Appendix.

Simulation results for the temperature dependence of the mobility are shown in Fig. 3, and for the concentration dependence in Fig. 4. The data were obtained for a system with 125 000 sites with the localization length chosen as $a = 0.3N^{-1/3}$. The simulation was repeated for five realizations of the system, yielding error bars smaller than the symbol size.

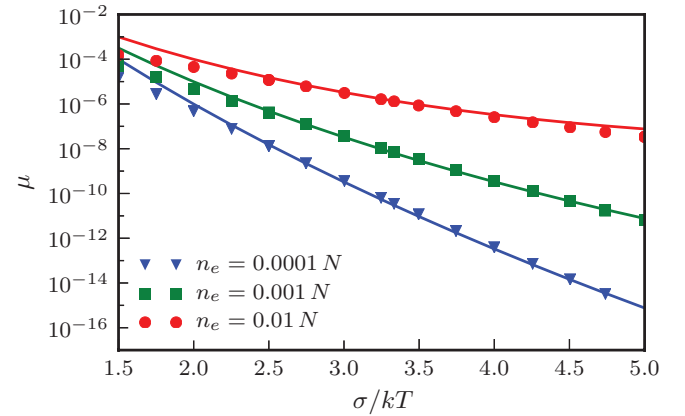


FIG. 3. (Color online) Temperature dependence of the mobility. The symbols show simulation results, while the curves are calculated with Eq. (31), for $N^{1/3}a = 0.3$. Deviations between the simulation results and the theory can be seen at high temperatures, when conditions (32) and (34) are not satisfied. For the highest concentration, a deviation is seen also at low temperatures where condition (33) is violated.

Fitting Eq. (31) against the simulation results, the value

$$A = 0.36 \quad (38)$$

was obtained for the constant in the resistivity expression (31). Figures 3 and 4 show good agreement between expression (31) and the simulation results, within the limits given by the conditions (32)–(34). Note that A is the only fitted parameter.

V. COMPARISON WITH LITERATURE RESULTS FOR EXPONENTIAL DOS

The result in the form of Eq. (31) for the resistivity of a 3D system in the variable-range-hopping (VRH) regime and the computer simulations described in Sec. IV allows for checking the validity of theoretical approaches tried in the literature for the same problem or similar ones. After the basics for theoretical description of the VRH were clarified in

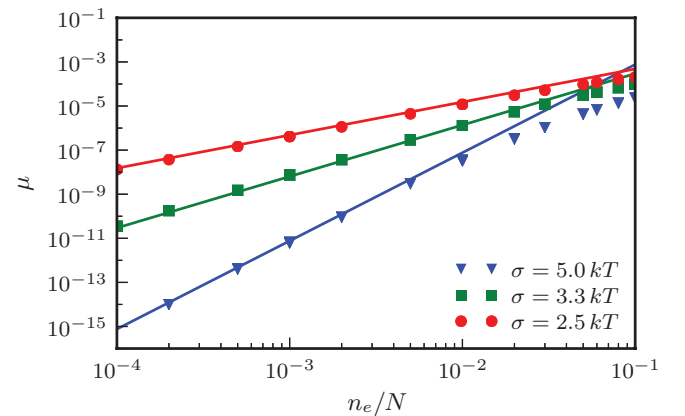


FIG. 4. (Color online) Concentration dependence of the mobility. The symbols show simulation results, while the curves are calculated with Eq. (31), for $N^{1/3}a = 0.3$. Deviations between the simulation results and the theory appears at high concentrations when condition (33) is violated.

TABLE I. Values for **A** and **B** in Eq. (40) for the different compared expressions of the mobility μ . C is an unknown numerical coefficient, $\nu \approx 0.875$ and $n_c \approx 0.219$ are known from percolation theory (see Sec. III), and $B_c = 2.7$ is a percolation threshold taken from Ref. 16.

Article	A	B
This paper, Eq. (31)	$0.36 \left(\frac{kT}{\sigma}\right)^\nu$	$\frac{1}{n_c}$
Oelerich <i>et al.</i> (Ref. 4)	$\frac{B_c}{2\pi}$	$\frac{4\pi}{3B_c} \frac{27}{\exp(3)}$
Vissenberg and Matters (Ref. 5)	C	$\frac{\pi}{B_c}$
Grünewald and Thomas (Ref. 10)	$\frac{1}{3}$	$\frac{4\pi}{3B_c} \frac{68}{27}$

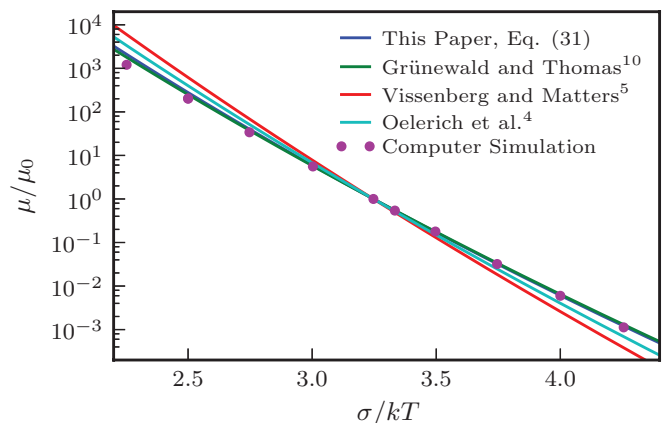


FIG. 5. (Color online) Normalized mobility values obtained from computer simulation and Eq. (40) for the different analytical solutions, for $N^{1/3}a = 0.3$ and $n_e/N = 0.001$.

Each data set is normalized to its value at $kT \approx 0.3\sigma$ so that only deviations in the temperature dependencies are shown.

It is clear from the figure that the result given by Eq. (31) and the one of Grünewald and Thomas¹⁰ show very similar dependencies on temperature, and they match the simulation results. This is particularly remarkable not only because the result of Grünewald and Thomas¹⁰ was the first obtained for the VRH in the exponential DOS, but mainly because it was based on a simplified approach with percolation threshold $B_c = 2.7$ for overlapping spheres with equal sizes. The result of Eq. (31) based on the geometrical problem with distributed sizes of spheres agrees with the simulation results even better. The comparison shown in Fig. 5 clearly confirms the validity of the approach suggested by Oelerich *et al.*⁴ for description of the VRH based on the transport-energy concept.

The worst performance in comparison with the simulation data in Fig. 5 is demonstrated by the results of Vissenberg and Matters.⁵ Therefore, it is worthwhile to discuss why the two percolation results of Grünewald and Thomas and of Vissenberg and Matters differ from each other. In both approaches, the average number of bonds per site is used as a percolation criterion. The system is assumed to percolate, when the sites in the largest cluster in average have $B_c \approx 2.7$ bonds each. However, in the two approaches the average is evaluated in different ways. Vissenberg and Matters evaluate the average number of bonds as the total number of bonds in the system divided by the number of sites with energies low enough to have bonds. Grünewald and Thomas^{10,30} (following Pollak⁹) performed weighted averaging for these two quantities, where the weight of each site is the probability for that site to belong to the largest cluster. This probability is taken to be proportional to the number of bonds connected to the site.

This difference in the averaging procedure could be the reason for the different temperature dependencies obtained. While our result is very close to that of Grünewald and Thomas, our approach is different to theirs. We do not use the number of bonds per site as the percolation criterion, making the derivation more transparent. As the difference between the results of the two percolation approaches show, the number

the early 1970s⁷⁻⁹ the main effort of researchers was directed to studying the effect of the DOS for the VRH charge transport. Grünewald and Thomas¹⁰ were the first who studied hopping transport in the exponential DOS given by Eq. (1) within the percolation approach. Almost two decades later the same problem in the framework of the percolation approach was addressed by Vissenberg and Matters.⁵ Vissenberg and Matters claimed to use a somewhat different percolation criterion as compared to that used by Grünewald and Thomas,¹⁰ although no comparison with the result of Grünewald and Thomas has been performed by Vissenberg and Matters, and it is not clear which percolation approach is more accurate.

A percolation approach has been derived also for the VRH in systems with Gaussian DOS, valid for organic disordered semiconductors.^{11,12} In this derivation, the percolation criterion from classical papers^{9,10} and not that of Vissenberg and Matters⁵ has been used. It is therefore highly desirable to check the validity of the classical recipe.^{9,10} Furthermore, aiming at a description of the VRH in disordered organic materials with Gaussian DOS, a transparent approach based on the concept of transport energy has recently been put forward.⁴ The latter approach should work also for the case of the exponential DOS, and herewith its validity can be checked by comparison with Eq. (31). Therefore, the results of Grünewald and Thomas,¹⁰ the results of Vissenberg and Matters,⁵ and the results of Oelerich *et al.*⁴ for the VRH in the exponential DOS are compared with that in Eq. (31) based on the mapping of the problem onto a geometrical problem.

For the sake of clarity, the results obtained for the charge carrier mobility μ are compared, which is related to the resistivities ρ via

$$\mu^{-1} = en_e \rho. \quad (39)$$

All analytical solutions mentioned above can be represented in the following form:

$$\mu = \mathbf{A} \frac{v_0 e}{n_e \sigma a} \left[\mathbf{B} \frac{n_e}{8} \left(\frac{a \sigma}{kT} \right)^3 \right]^{\sigma/kT}, \quad (40)$$

where **A** and **B** are different factors for different approaches to be checked. Table I lists the different approaches and the corresponding values of **A** and **B**. Note that in all cases, these values are either constants or slowly varying functions of σ/kT so that the strong dependencies on system parameters are equal in all solutions.

In Fig. 5, the results from Eq. (40) and Table I are plotted together with data points obtained by computer simulations.

of bonds is a nontrivial quantity to use when all sites are not equivalent.

One can view the good agreement between our numeric and analytic results with those of Grünewald and Thomas as supporting the averaging procedure used by Grünewald and Thomas.¹⁰ This information is important for applying the percolation theory to systems with a DOS different from the exponential one considered in the current report.

VI. HOPPING TRANSPORT ON A LATTICE

So far, variable range hopping over sites randomly distributed in space was considered. With the percolation approach developed in Sec. II, the mobility was determined for the particular case of an exponential distribution of site energies. In another special case, that of nearest-neighbor hopping on a lattice, the mobility can be found for *any* shape of the DOS, using a similar percolation approach. For this reason, a discussion of the lattice case is included here. The only properties of the DOS needed are the characteristic width σ and the critical energy ε^* required for percolation.

Consider a lattice of sites with the lattice constant l_0 , where resistances R_{ij} connect nearest-neighbor sites i and j :

$$R_{ij} = \frac{kT}{e^2 v_0} \exp\left(\frac{2l_0}{a}\right) \exp\left[\frac{\max(\varepsilon_i, \varepsilon_j) - \varepsilon_F}{kT}\right]. \quad (41)$$

The energy-dependent exponent is denoted by ξ_{ij} , so that $R_{ij} = R_0 \exp(\xi_{ij})$. Let $\sigma_{dc}(\xi)$ be the dc conductivity of the lattice, in which resistances with $\xi_{ij} > \xi$ are cut off. There is a critical value ξ_c , such that $\sigma_{dc}(\xi) > 0$ only when $\xi > \xi_c$. The conductivity rapidly increases above ξ_c , and saturates at $\xi \approx \xi_c + O(1)$, determined by the critical subnetwork of resistances with $\xi_{ij} < \xi_c + O(1)$.¹⁶ As in the VRH case, the resistivity of the system is determined using Eq. (23), where L_{corr} and R_{crit} are obtained for the lattice case below. The critical resistance in the lattice case can be written as

$$R_{\text{crit}} = \frac{kT}{e^2 v_0} \exp\left(\frac{2l_0}{a}\right) \exp\left(\frac{\varepsilon^* - \varepsilon_F}{kT}\right), \quad (42)$$

where ε^* again denotes the critical energy. Because the resistance expression (41) depends only on the larger of the two energies ε_i and ε_j , the bond between sites i and j has $\xi_{ij} < \xi_c$ and is present in the percolating network if both inequalities $\varepsilon_i < \varepsilon^*$ and $\varepsilon_j < \varepsilon^*$ hold. This turns the problem of finding ε^* into a *site*-percolation problem, and allows for determining the critical energy using the site-percolation threshold p_s of the lattice,

$$\int_{-\infty}^{\varepsilon^*} g(\varepsilon) d\varepsilon = p_s, \quad (43)$$

with the threshold p_s depending on the lattice type.

The correlation length L_{corr} of the percolating network above the percolation threshold p_c is determined by the scaling relation¹⁶

$$L_{\text{corr}} \simeq l_0 (p - p_c)^{-\nu}, \quad (44)$$

where p is the fraction of bonds with $\xi_{ij} < \xi$ and l_0 is the lattice constant. This expression is similar to Eq. (29), but formulated in quantities appropriate for the lattice problem.

For a distribution of site energies characterized by the width σ , the distribution of ξ_{ij} has a width of order σ/kT . Then,

$$p - p_c = (\xi - \xi_c) \frac{dp}{d\xi} \simeq (\xi - \xi_c) \left(\frac{\sigma}{kT}\right)^{-1} \simeq \frac{kT}{\sigma}, \quad (45)$$

and

$$L_{\text{corr}} \simeq l_0 \left(\frac{\sigma}{kT}\right)^\nu. \quad (46)$$

Using Eqs. (23), (42), and (46), one obtains the resistivity ρ in the form

$$\rho \simeq \frac{kT l_0}{e^2 \omega_0} \left(\frac{\sigma}{kT}\right)^\nu \exp\left(\frac{\varepsilon^* - \varepsilon_F(T, n_e)}{kT}\right), \quad (47)$$

with $\omega_0 = v_0 \exp(-2l_0/a)$ for consistency with the VRH expressions.

A scaling theory for charge transport on a regular lattice with Gaussian distribution of site energies was recently presented¹³ by Cottaar *et al.* The main result is an expression for the charge carrier mobility, as a function of the fraction of occupied sites c and temperature T ,

$$\mu(T, c) = B \frac{e\omega_0}{N^{2/3} kT c} \left(\frac{kT}{\sigma}\right)^\lambda \exp\left[\frac{\varepsilon_F(T, c) - \varepsilon^*}{kT}\right]. \quad (48)$$

Here, B and λ are parameters assumed to be independent of T and c . These two parameters were determined by fitting Eq. (48) to mobility data obtained in numerical simulations.

Expressing the mobility μ as $(en_e \rho)^{-1}$ with ρ given by Eq. (47), the carrier concentration n_e as Nc , and the lattice constant l_0 as $N^{-1/3}$, the expression (48) for the mobility μ is obtained, except that ν replaces λ . Therefore the exponent λ introduced in Ref. 13 is equal to the critical exponent ν of the correlation length of the percolation cluster, with the magnitude $\nu = 0.875 \pm 0.008$ in the 3D case.¹⁷ The exponent ν is independent of the details of the lattice and the hopping rate model, and depends only on the number of spatial dimensions of the problem. The values for λ obtained by fitting in Ref. 13 are very close to ν .

VII. SUMMARY

A percolation approach was developed to solve the variable range hopping (VRH) problem in the exponential DOS. By mapping the physical VRH problem onto a purely geometrical problem using randomly placed spheres with distributed sizes, an expression was derived for the hopping resistivity in the exponential DOS in two and three dimensions, exact up to a single numerical coefficient. The solution contains the pronounced exponential dependencies of the resistivity on material parameters as well as the more weakly dependent prefactors.

The purely geometrical percolation problem of spheres with distributed sizes was studied in detail, and the percolation thresholds were found with high accuracy in two and three dimensions using numerical simulations. The single fitting parameter in the analytical expression for the resistivity was determined via straightforward computer simulations. Comparison of the analytical theory with the simulation results shows a good agreement within the range of parameters, for which the theory is applicable.

548 The obtained expression for the VRH mobility was compared to the literature results. The result of Grünwald and Thomas¹⁰ performs remarkably well in comparison to the analytical solution and to the computer simulations.

552 Finally, the problem of the nearest-neighbor hopping on a lattice was discussed. It is shown that the percolation approach provides a solution for this problem including pre-exponential factors also for DOS functions different from the exponential one.

557 ACKNOWLEDGMENTS

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562 APPENDIX: BALANCE EQUATIONS FOR THE OHMIC REGIME

564 The numerical results for the charge carrier mobility presented in Sec. IV were obtained by solving balance equations for the charge carrier flow. In this method, a system of randomly placed sites with randomly chosen energies is generated. The steady state current in response to an electric field is then calculated for the system. First, the occupation probability for each site is determined by solving a set of balance equations. The occupation probabilities are then used to evaluate the current. The system is considered with periodic boundary conditions in all directions, so that when an electric field is applied, there will be a net current flowing through the system in steady state.

576 In the steady state, the electron flow into each site equals the electron flow out,

$$\sum_j n_i \Gamma_{ij} (1 - n_j) = \sum_j n_j \Gamma_{ji} (1 - n_i), \quad (\text{A1})$$

578 where n_i is the probability that site i is occupied by an electron.

579 Equation (A1) is a nonlinear system of equations, with one equation for each site in the system. When the charge carrier concentration can be considered small, the equations are typically linearized by omitting the factors $(1 - n_i)$, because the resulting linear equation system is much easier to solve than the full nonlinear one.^{31,32}

585 For the present problem, the charge carrier concentration can not be considered small. However, the equations can be linearized for the limit of a small electric field.²⁹ Only the small-field limit is of interest in the current case, because the percolation theory is derived for the Ohmic regime.

590 Following Ref. 29, we define n_i^0 as the occupation probability of site i at zero electric field. These occupation probabilities follow the Fermi-Dirac distribution,

$$n_i^0 = f(\varepsilon_i, \varepsilon_F) = \frac{1}{1 + \exp[(\varepsilon_i - \varepsilon_F)/kT]}, \quad (\text{A2})$$

593 since the hopping rates obey detailed balance. The problem will be solved in the variables

$$p_i = \frac{n_i - n_i^0}{n_i^0(1 - n_i^0)}, \quad (\text{A3})$$

598 which measure the electric-field-induced deviations in the probabilities n_i . Further, the hopping rate Γ_{ij} is split into two parts,

$$\Gamma_{ij} = \Gamma_{ij}^0 + \Delta\Gamma_{ij}, \quad (\text{A4})$$

598 where Γ_{ij}^0 denotes the hopping rate from site i to j in the absence of an electric field and $\Delta\Gamma_{ij}$ is the field-induced change. The zero-field quantities obey the detailed balance relation

$$n_i^0 \Gamma_{ij}^0 (1 - n_j^0) = n_j^0 \Gamma_{ji}^0 (1 - n_i^0). \quad (\text{A5})$$

602 Next, n_i from Eq. (A3) and Γ_{ij} from Eq. (A4) are inserted into Eq. (A1). Terms quadratic in p are omitted, and Eq. (A5) is used to cancel terms without p 's. Finally, $\Delta\Gamma_{ij}$ is expressed by linearizing the hopping rate (3), yielding

$$-p_i \sum_j \gamma_{ij} + \sum_j p_j \gamma_{ij} = \frac{eE}{kT} \sum_j \gamma_{ij} (z_j - z_i), \quad (\text{A6})$$

606 where E is the magnitude of the electric field (the field is assumed to be directed in the z direction), and

$$\gamma_{ij} = n_i^0 \Gamma_{ij}^0 (1 - n_j^0). \quad (\text{A7})$$

608 Equation (A6) is a linear system of equations. The equations are not all independent, since the total number of charge carriers is free to change. One further equation is given by fixing the total number of charge carriers in the system,

$$\sum_j p_j n_j^0 (1 - n_j^0) = 0. \quad (\text{A8})$$

612 One can simply replace one of the equations in (A6) with Eq. (A8), however, for numerical stability it is preferable to spread the additional constraint over the whole system by adding it, with a suitably chosen scaling factor, to each equation in (A6).³³ The zero-field quantities n_i^0 can be determined either by fixing the total concentration n_e of charge carriers in the system or by specifying the Fermi level. To decrease the variations of the current between different realizations of the system, the latter approach was chosen.³³ The Fermi energy ε_F was found numerically by solving

$$\int_{-\infty}^{\infty} g(\varepsilon) f(\varepsilon, \varepsilon_F) d\varepsilon = n_e \quad (\text{A9})$$

622 for the desired n_e .

623 Once the p_i are known from solving Eq. (A6), the current through the system in the z direction is given by

$$I = e \sum_{i>j} \frac{z_j - z_i}{L_z} \gamma_{ij} \left[p_i - p_j + \frac{eE}{kT} (z_j - z_i) \right], \quad (\text{A10})$$

625 where L_z is the length of the system in the z direction. This expression was obtained by evaluating the net charge carrier flow, using the same linearizations as for Eq. (A6).

627 The expression for the current I shows the physical meaning of the quantities p_i and γ_{ij} . Performing the linearization corresponds to considering a resistor network picture, where γ_{ij} plays the role of conductivity between sites i and j , while p_i corresponds to the potential at site i . The resulting equations are then simply Kirchhoff's laws for the network.

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