

Effective temperature for hopping transport in a Gaussian density of states

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(Received 10 April 2008; published 28 May 2008)

For hopping transport in disordered materials, the mobility of charge carriers is strongly dependent on the temperature and the electric field. Our numerical study shows that both the energy distribution and the mobility of charge carriers in systems with a Gaussian density of states, such as organic disordered semiconductors, can be described by a single parameter—effective temperature, which is dependent on the magnitude of the electric field. Furthermore, this effective temperature does not depend on the concentration of charge carriers, while the mobility does depend on the charge carrier concentration. The concept of the effective temperature is shown to be valid for systems with and without space-energy correlations in the distribution of localized states.

DOI: [10.1103/PhysRevB.77.195211](https://doi.org/10.1103/PhysRevB.77.195211)

PACS number(s): 72.20.Ht, 72.20.Ee, 72.80.Le, 72.80.Ng

I. INTRODUCTION

For many years, much of the attention of researchers has been devoted to the effect of high electric fields on the hopping mobility of charge carriers in organic disordered materials, such as conjugated and molecularly doped polymers and semiconducting organic glasses.^{1–4} Two peculiarities in the dependence of the carrier mobility on the electric field are usually discussed in literature. One of them is the apparent decrease in the mobility with a rising field at relatively low fields and high temperatures reported in several time-of-flight studies.^{1,5–7} The other one is the very strong nonlinear increase in the carrier mobility with the electric field at low temperatures and high fields.^{1–4} While the former peculiarity has been attributed^{3,8} to the misinterpretation of experimental data, the latter one has been confirmed in numerous experimental studies and it is currently the focus of an intensive theoretical research. Before we turn to discussing this research, it is worth noting that very similar nonlinear effects with respect to the applied electric field have been known since decades for transport phenomena in inorganic noncrystalline materials, such as amorphous semiconductors. Indeed, strong nonlinearities in amorphous semiconductors were observed for the field dependence of the dark conductivity,^{9,10} of the photoconductivity,¹¹ and of the charge carrier drift mobility^{9,12,13} at high electric fields. While the theoretical description of the field-dependent hopping conductivity at low temperatures was always a challenge, the theories for the temperature dependence of the hopping conductivity at low electric fields were successfully developed for all transport regimes listed above (see, for instance, Ref. 3 and references therein). Furthermore, it has been shown that the effect of a strong electric field on transport coefficients in amorphous semiconductors can be effectively described by replacing the temperature parameter in the formulas for the low-field temperature-dependent mobility and conductivity by an effective temperature $T_{\text{eff}}(T, F)$, which is dependent on the magnitude of the electric field F . Shklovskii¹⁴ was the first who recognized that for hopping conduction, a strong electric field plays a role similar to that of temperature. In the presence of the field, the number of sites available for charge

transport is essentially enhanced in the direction prescribed by the field. The distance between sites available for hopping transport shortens; hence, electrons can move faster.¹⁴ The concept of the effective temperature has been studied in detail for systems with an exponential energy distribution of localized states usually assumed for inorganic noncrystalline materials as follows:

$$g(\varepsilon) = \frac{N}{\sigma} \exp\left(\frac{\varepsilon}{\sigma}\right), \quad (1)$$

where N is the concentration of localized states and σ is the energy scale of the distribution. By studying the steady-state energy distribution of electrons in numerical calculations and computer simulations^{15,16} and by computer simulations of the steady-state hopping conductivity and the transient energy relaxation of carriers,¹⁷ the following result has been found: The whole set of transport coefficients can be represented as a function of a single parameter, $T_{\text{eff}}(T, F)$,

$$T_{\text{eff}} = \left[T^\beta + \left(\gamma \frac{eFa}{k} \right)^\beta \right]^{1/\beta}, \quad (2)$$

with $\beta=2$ and values of γ in the range 0.5–0.9 depending on which transport coefficient is considered.¹⁷ In this expression, a is the localization length of charge carriers in the localized states, e is the elementary charge, and k is the Boltzmann constant. Herewith, the problem of the nonlinearities of transport coefficients with respect to the applied electric field for inorganic noncrystalline materials with the density of states (DOS) described by Eq. (1) has been solved.

Let us now turn to organic disordered materials. In such systems, the density of states involved in the hopping transport of charge carriers is believed to be not purely exponential as in Eq. (1) but rather to be described by a Gaussian energy distribution^{1–4,18,19} as follows:

$$g(\varepsilon) = \frac{N}{\sigma\sqrt{2\pi}} \exp\left(-\frac{\varepsilon^2}{2\sigma^2}\right). \quad (3)$$

Numerous computer simulations have been devoted to studying the field nonlinearities of transport coefficients in the hopping regime in such systems. Two models of disor-

dered organic materials were considered: the so-called Gaussian disorder model (GDM) suggested by BäSSLer¹ and the so-called correlated disorder model (CDM) considered by Gartstein and Conwell,²⁰ by Dunlap *et al.*,²¹ and by Novikov *et al.*^{22,23} In both models, the field dependence of carrier mobility has been studied by computer simulations. While analytical calculations have been carried out in order to justify the CDM,^{21–23} a consistent analytical theory for the field dependence of the hopping mobility in a Gaussian DOS is still missing.

It is tempting to try to apply the concept of the effective temperature to organic disordered systems with a Gaussian DOS described by Eq. (3) since this concept has proven to be very successful for inorganic systems with an exponential DOS described by Eq. (1). The idea that a strong electric field leads to heating of the charge carriers in organic materials has already been considered in several theoretical studies. Recently, Preezant and Tessler²⁴ performed such a study within the framework of an analytical approach that artificially decouples the energy-dependent factors in the hopping transport from the space-dependent factors. However, this approach has been shown^{3,25} to be unsuitable for describing hopping transport processes in disordered materials. Moreover, such an approach leads to an effective temperature that essentially differs from the one considered in previous studies for inorganic materials,^{3,14–17} wherein the effective temperature arises from the interplay between space- and energy-dependent factors in hopping processes. Li *et al.*²⁶ recently approached this problem by inserting the dependence of transition rates on the electric field and that of the percolation threshold into the percolation theory of Vissenberg and Matters.²⁷ More recently, Limketkai *et al.*²⁸ exploited the concept of the effective temperature in order to account for the strong field nonlinearity of the conductivity and carrier mobility observed in organic semiconductors.²⁹ Limketkai *et al.*²⁸ chose the expression for the effective temperature $T_{\text{eff}}(T, F)$ in the form of Eq. (2) with $\beta=1$ and $\gamma=0.5$. It has been proven,¹⁷ however, that such an expression for the effective temperature with $\beta=1$ cannot be considered as suitable. Indeed, suppose the conductivity G is dependent on $T_{\text{eff}}(T, F)$ solely. Then

$$\frac{dG}{dF} = \frac{dG}{dT_{\text{eff}}} \frac{dT_{\text{eff}}}{dF}. \quad (4)$$

In the Ohmic transport regime at $F \ll kT/ea$, the conductivity G must be field independent, implying that

$$\frac{dT_{\text{eff}}}{dF} \rightarrow 0 \text{ as } F \rightarrow 0. \quad (5)$$

The function described by Eq. (2) with $\beta=1$ obviously does not fulfill this condition. However, any function of this kind with $\beta > 1$ is consistent with Eq. (5) along with the necessary condition $T_{\text{eff}}=T$ at $F=0$ and $T_{\text{eff}} \propto F$ at $T=0$.

Therefore, a challenging problem arises on finding out whether the concept of the effective temperature is applicable to systems with a Gaussian DOS and if so, what is the expression for $T_{\text{eff}}(T, F)$. We try to answer these questions in the present study. For this purpose, we follow the idea of

Marianer and Shklovskii¹⁵ to look at the energy distribution of charge carriers using the numerical method of nonlinear balance equations suggested by Yu *et al.*^{30,31} Pasveer *et al.*,³² and Cottaar and Bobbert³³ and apply it to electron transitions in a Gaussian DOS. The preliminary data for the energy distribution of charge carriers in the GDM at low carrier concentrations confirmed the concept of the effective temperature.³⁴

The paper is organized as follows: In Sec. II, the numerical method used in our study is described. In Sec. III, the results for the energy distribution function of charge carriers in the GDM are presented. It is shown that at finite temperatures and electric fields the energy distribution of charge carriers in a Gaussian DOS can be well described by the Fermi–Dirac distribution function characterized by $T_{\text{eff}}(T, F)$ given by Eq. (2) with $\beta=1.54 \pm 0.2$ and $\gamma=0.64 \pm 0.2$, thus confirming the applicability of the concept of the effective temperature to organic disordered solids. Furthermore, we show that the expression for $T_{\text{eff}}(T, F)$ is stable against changes in the concentration of charge carriers. Numerical results obtained for the mobility of charge carriers also confirm the concept of the effective temperature for the GDM. In Sec. IV, the corresponding results for the CDM are presented. They prove that the concept of the effective temperature is valid also for correlated systems. Concluding remarks are gathered in Sec. V.

II. NUMERICAL METHOD

In order to check the validity of the concept of the effective temperature, we solved a system of nonlinear balance equations with respect to the steady-state occupation probabilities p_i of sites and checked, following the idea of Marianer and Shklovskii,¹⁵ whether this distribution can be fitted by a Fermi–Dirac distribution,

$$p(\varepsilon) = \frac{1}{1 + e^{(\varepsilon - \mu_c)/kT_{\text{eff}}}}, \quad (6)$$

with a single parameter, $T_{\text{eff}}(T, F)$. We also calculated the carrier drift velocity along the field direction and the corresponding mobility at different temperatures and electric fields and checked whether the mobility can be described as a function of a single parameter, $T_{\text{eff}}(T, F)$, combined from the temperature T and the field strength F .

The balance equation for the occupation probability p_i of a site i has the form^{30–33}

$$\sum_{j \neq i} p_i \Gamma_{ij}(1 - p_j) = \sum_{j \neq i} p_j \Gamma_{ji}(1 - p_i), \quad (7)$$

where the rate of jumping from site i to site j is given by the Miller–Abrahams formula,

$$\Gamma_{ij} = \nu_0 e^{-2(\Delta R_{ij}/a)} \begin{cases} e^{-\Delta \varepsilon_{ij}/kT}, & \Delta \varepsilon_{ij} > 0 \\ 1, & \Delta \varepsilon_{ij} \leq 0. \end{cases} \quad (8)$$

Here, $\Delta \varepsilon_{ij}$ is the difference between the energies of states j and i , ΔR_{ij} is the distance between these states, and ν_0 is the attempt-to-escape frequency. While in the initial work of Marianer and Shklovskii¹⁵ the limit of a low concentration of

charge carriers was considered and the balance equations were linearized, we study a system with a finite number of charge carriers n . In order to solve the system of the nonlinear balance equations, we use the iterative numerical procedure suggested by Yu *et al.*,^{30,31} Pasveer *et al.*,³² and Cottaar and Bobbert.³³ Balance equation (7) is rewritten in the form

$$p_i = \frac{\sum_{j \neq i} \Gamma_{ji} p_j}{\sum_{j \neq i} \Gamma_{ij} - \sum_{j \neq i} (\Gamma_{ij} - \Gamma_{ji}) p_j}, \quad (9)$$

where the right hand side does not contain p_i . This expression is iteratively used to find a solution to Eq. (7). In each iteration step, all p_i are updated. If p_j has already been calculated in the current step, this value is used; otherwise, the result from the previous step is used. This so-called implicit iteration is necessary for obtaining convergence.³⁰ In the sums over j , only the most important transitions are needed. We have considered jumps shorter than a cutoff length R_{cutoff} , chosen to be so large that it does not affect the transport parameters.

The procedure is repeated until the relative change in any one of the probabilities is smaller than 10^{-10} . We note that the solution seems to converge faster if the sites are placed on a lattice than at random. When the localization length is small, more iterations are needed, but the cutoff length can be reduced.

If an electric field F is applied in the z direction, the difference in energy between sites j and i is given by

$$\Delta \varepsilon_{ij} = \varepsilon_j - \varepsilon_i - F e (z_j - z_i). \quad (10)$$

When the probability distribution of the charge carriers over the sites is known, the drift velocity of the carriers along the field and their mobility are given, respectively, by

$$v_z = \sum_{i,j \neq i} p_i \Gamma_{ij} (1 - p_j) (z_j - z_i) / n, \quad \mu = \frac{v_z}{F}. \quad (11)$$

We studied a system of M sites randomly distributed within a cube with a side length L . Each site has a random energy ε , from a Gaussian distribution with the width σ . The density of states is given by Eq. (3). Periodic boundary conditions were applied in all directions. The calculations were performed in dimensionless units, where the width of the DOS, σ , the site concentration $N = M/L^3$, the Boltzmann constant k , elementary charge e , and the attempt-to-jump frequency ν_0 are equal to unity.

We used a system containing $M = 8000$ sites inside a cube with side length $L = 20$. The localization length was varied between 0.2 and 1, while the particle concentration $c = n/M$ was varied in the interval between $c = 10^{-5}$ and $c = 0.1$.

For each choice of T , F , concentration c , and localization length a , a number of realizations of the system were generated and the steady state occupation probabilities were determined. For each realization, the carrier mobility and effective temperature were calculated. The magnitudes of the effective temperatures obtained with different realizations of the system were equal to each other within the accuracy of 1%.

The effective temperature was determined from the occupation probabilities p_i with a linear fit of $\ln(1/p_i)$ as a function of ε_i ,

$$\ln\left(\frac{1}{p_i}\right) = \frac{1}{kT_{\text{eff}}} \varepsilon_i - \frac{\mu_c}{kT_{\text{eff}}}. \quad (12)$$

This approach works well when the variation in the occupation probabilities for sites with similar energies is small. When the variation is larger, we found it better to produce a histogram of $p(\varepsilon)$ and then fit to this histogram as above. The histogram approach seems more correct since it uses the average occupation probability at a given energy, in contrast to the direct fit that uses the average of $\ln(1/p)$. All of the results for effective temperature presented below were obtained with the histogram method. The parameters β and γ in Eq. (2) were determined by simultaneous fitting of all data points to the surface given by this equation.

In Sec. III, we present the results of calculations for a system without any correlation between site energies and their spatial positions (GDM). In Sec. IV we present the results of calculations for the CDM—a system in which the energies of neighboring sites are correlated. In order to introduce correlations, we generate an initial energy for each site with a Gaussian distribution. We then calculate the real energy for each site by averaging the initial energies of all sites inside the correlation radius R_{corr} , in accord with the recipe suggested by Gartstein and Conwell.²⁰ This averaging procedure has two effects. It will introduce a correlation in energy between spatially close sites and it will decrease the width of the energy distribution. In order to keep the width of the energy distribution independent of the correlation length, the site energies were rescaled to provide the initial energy width σ . We discuss these effects of space-energy correlations in more detail in Sec. IV.

At very high electric fields, when the energy landscape is strongly sloped along the field direction and the energy differences between the successive states become negative as given by Eq. (10), the carrier drift velocity saturates and becomes field independent.¹ We study the range of electric fields, which are less than the field at which the velocity saturates.

III. EFFECTIVE TEMPERATURE FOR SYSTEMS WITHOUT SPACE-ENERGY CORRELATIONS

In Fig. 1, the numerical results are shown for the occupation probability for each site as a function of site energy for three different charge concentrations. The inset shows the same data on a logarithmic scale.

At low concentrations, the data can be fitted by a Boltzmann distribution with the parameter $T_{\text{eff}}(T, F)$, while the data at high concentrations show a Fermi–Dirac shape, remarkably corresponding to the same parameter $T_{\text{eff}}(T, F)$. Furthermore, Fig. 2 shows that the value of $T_{\text{eff}}(T, F)$ does not depend on the carrier concentration c . These results are really remarkable because they mean that the concept of the effective temperature is valid for systems with a Gaussian density of states [so far it has been proven only for exponential DOS described by Eq. (1)] and that the parameter

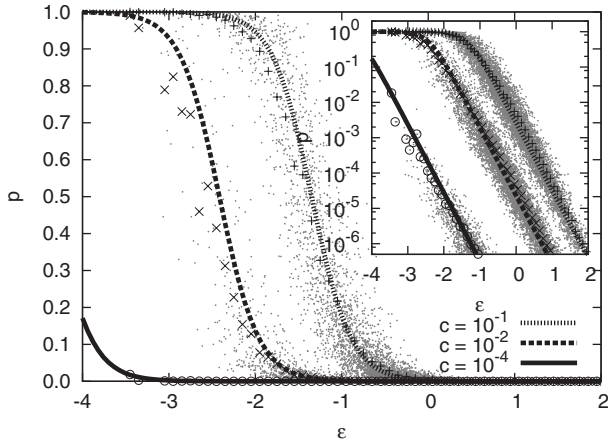


FIG. 1. Occupation probability as a function of site energy for three different charge carrier concentrations c . The temperature and the field are $T=0.2$ and $F=0.4$, respectively. The dots show the occupation probabilities of individual sites, while the symbols show a histogram, i.e., average occupation probability for sites in an energy interval. The curves show the Fermi–Dirac functions fitted to the histogram.

$T_{\text{eff}}(T, F)$ is universal with respect to the concentration of charge carriers in the system under study.

In Fig. 3, we bring together all of the calculation results obtained for various field strengths in the range $0 < F < 3$ and for various temperatures in the range $0.1 < T < 0.5$ for concentration of carriers $c=10^{-2}$ and 10^{-5} , in the form of a three-dimensional plot of $T_{\text{eff}}(T, F)$, obtained by the best fit of the calculated energy distributions by a Fermi–Dirac function. Also, the surface determined by Eq. (2) with the parameters $\beta=1.54$ and $\gamma=0.64$ is shown in Fig. 3. One can see excellent agreement between the calculated results and Eq. (2) with these parameters.

As described in Sec. II, we also studied the mobility of charge carriers with respect to the applicability of the concept of the effective temperature. In Fig. 4, the dependences of the carrier mobility as a function of $T_{\text{eff}}(T, F)$ determined from the fits of the energy distribution function are shown for different concentrations c of charge carriers. These data were

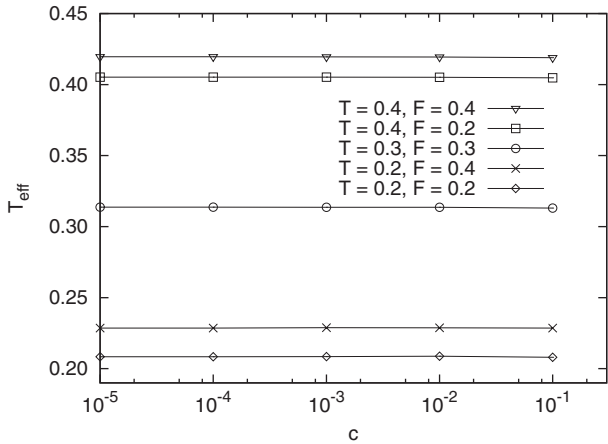


FIG. 2. Calculated T_{eff} as a function of charge carrier concentration for different values of T and F .

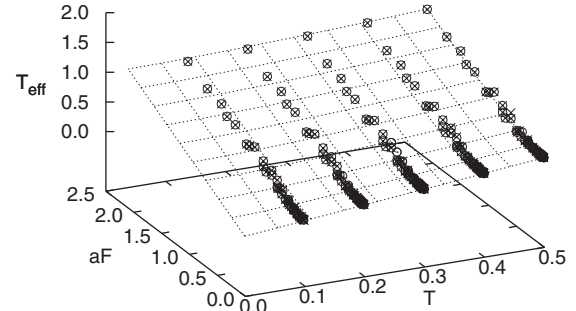


FIG. 3. T_{eff} as a function of temperature T and aF . The concentrations $c=10^{-2}$ and 10^{-5} are shown. The surface is the best fit to Eq. (2), with $\beta=1.54$ and $\gamma=0.64$.

obtained for various field strengths in the range $0 < F < 3$ and for various temperatures in the range $0.1 < T < 0.5$. The data were averaged over three realizations. For the chosen parameters, it appears not important whether the mobilities or the inverse mobilities were averaged. While the magnitude of the mobility appears sensitive to the concentration of carriers, in agreement with the results obtained previously by several research groups,^{25,27,30–33,35–39} the magnitude of the effective temperature for the given pair (T, F) does not indicate any dependence on c .

Therefore, one can conclude that for the GDM, i.e., for a disordered system with a Gaussian distribution of energies and without any correlations between spatial positions of hopping sites and their energies, the concept of the effective temperature is valid. In Sec. IV, we present results for the CDM, i.e., for the correlated system.

IV. EFFECTIVE TEMPERATURE FOR SYSTEMS WITH SPACE-ENERGY CORRELATIONS

In this section, we study the effect of the space-energy correlations on the interplay between the temperature T and the electric field F with respect to the validity of the concept of the effective temperature.

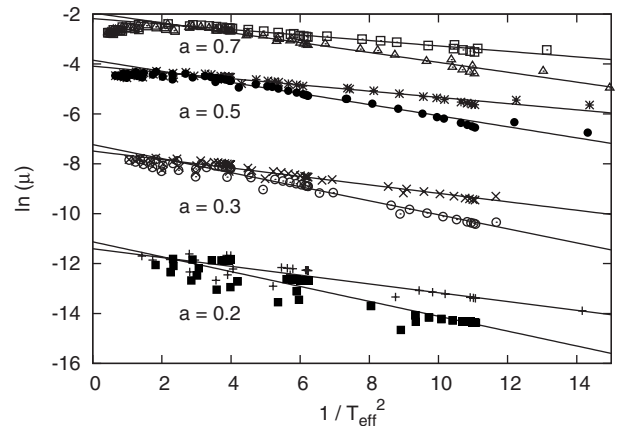


FIG. 4. The dependence of the carrier mobility on the effective temperature for different localization lengths a . In each pair of curves, the upper one is for the charge carrier concentration $c=10^{-2}$ and the lower one is for $c=10^{-5}$.

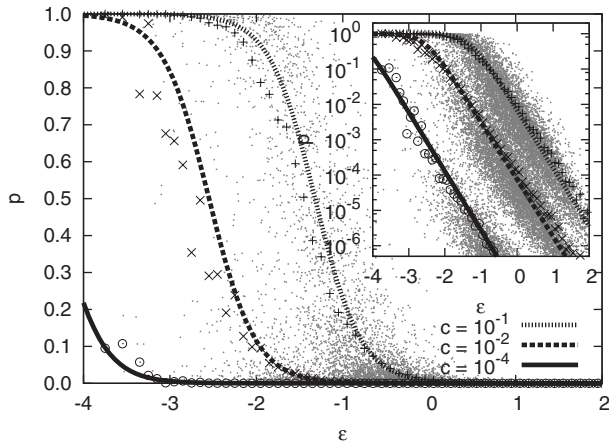


FIG. 5. Occupation probability in the correlated disorder case, with $R_{\text{corr}}=2$. The temperature and the field are $T=0.2$ and $F=0.4$, respectively. The symbols are defined as in Fig. 1.

In Fig. 5, the numerical results are shown for the occupation probability for each site as a function of site energy for three different charge concentrations at correlation length $R_{\text{corr}}=2$. The inset shows the same data on a logarithmic scale. In analogy to the uncorrelated system, at low concentrations the data can be fitted by a Boltzmann distribution with the parameter $T_{\text{eff}}(T, F)$, while the data at high concentrations show a Fermi–Dirac shape corresponding to the same parameter $T_{\text{eff}}(T, F)$ as for low concentration.

Figure 6 shows the effective temperature as a function of the correlation length R_{corr} . The effective temperature strongly depends on the correlation length; thus, Eq. (2) is not directly applicable to a system with space-energy correlations.

Before discussing the carrier mobility in a correlated system, one should note the following: When the space-energy correlations are introduced as described in Sec. II, the effective width σ of the energy distribution of localized states decreases. Furthermore, this width becomes smaller with increasing R_{corr} . Therefore, one can expect that with increasing R_{corr} , the mobility would increase just because of the diminishing energetic disorder due to the effect of correlations. In order to compare the carrier mobilities in systems with dif-

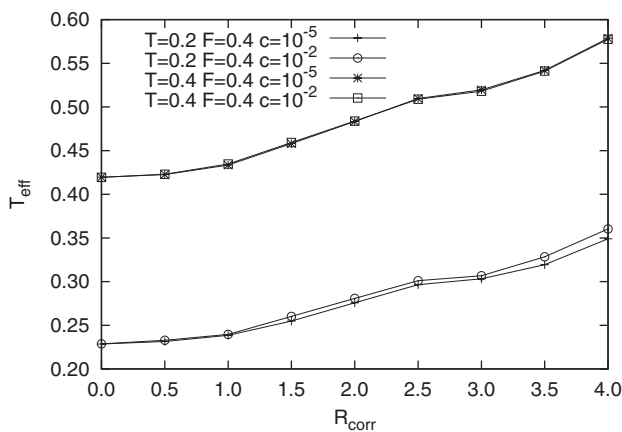


FIG. 6. The effective temperature as a function of the correlation length, for randomly placed sites, with $a=0.5$.

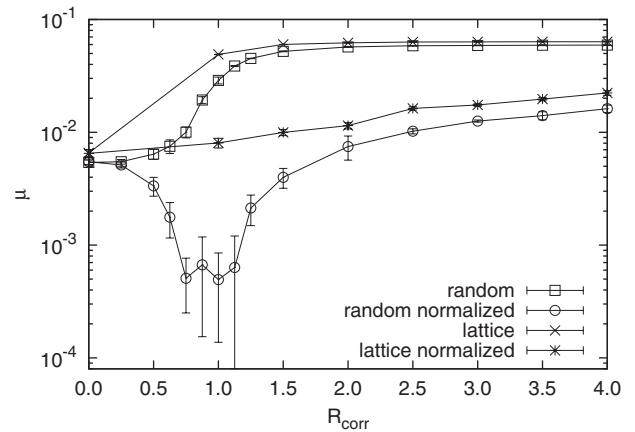


FIG. 7. Mobility as a function of correlation length, with and without rescaling of site energies, for $T=0.4$, $F=0.4$, $a=0.5$, and $L=30$.

ferent correlations and the same energy disorder, one should therefore rescale the width of the energy distribution in the correlated systems and bring it to the value initially ascribed to the uncorrelated system.²⁰ The effect of correlations on the carrier mobility has already been studied by computer simulations, although within the framework of the lattice model and not with respect to the validity of the effective temperature.²⁰ In order to compare our results to those of previous studies, we also have simulated mobilities in correlated systems within a lattice model along with simulations in the random model.

Figure 7 shows the mobility μ as a function of the correlation length R_{corr} with randomly placed sites and with sites on a lattice for both cases: with and without rescaling (normalization) of the width of the energy distribution. As expected, without rescaling, the mobility always increases with the increase in the correlation length R_{corr} . However, in the rescaled system there is an apparent difference between the lattice model and the random model with respect to the dependence of the mobility on the correlation length. While in the lattice model the mobility monotonously increases with R_{corr} , in accord with the results of previous studies,²⁰ in the random model the dependence of μ on R_{corr} appears nonmonotonous. At $R_{\text{corr}} \leq 1$, the mobility decreases with R_{corr} . The reason for the low mobility at $R_{\text{corr}} \approx 1$ could be the following: The normalization (rescaling) procedure creates a small number of sites with very low energies. While the general shape of the DOS for the correlated system is Gaussian and the variance of the DOS is normalized to be equal to the variance of the uncorrelated system, the DOS of the correlated system has longer tails, and the low-energy tail greatly reduces the carrier mobility. The sites in the tail are those sites that initially had a low energy and have no neighbors within the distance R_{corr} . Thus, they are unaffected by the averaging but are still rescaled with the normalization factor, which is typically close to 2 for $R_{\text{corr}}=1$. This idea is supported by the data shown in Fig. 8. The dip in the mobility disappears when the charge carrier concentration is increased, and the deep states are filled.

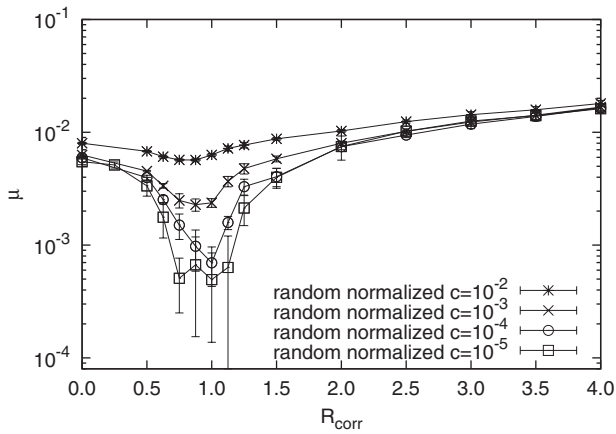


FIG. 8. Mobility as a function of correlation length, with randomly placed sites and rescaling of the site energies, for different concentrations of charge carriers; $T=0.4$, $F=0.4$, $a=0.5$, and $L=30$.

V. CONCLUSIONS

The concept of the effective temperature has been shown to be valid for a system with a Gaussian DOS with and without space-energy correlations in the distribution of local-

ized states. From the numerical results for uncorrelated systems, one can conclude that the effective temperature is described by Eq. (2) with the parameters $\beta=1.54 \pm 0.2$ and $\gamma=0.64 \pm 0.2$. Remarkably, the validity of the concept of the effective temperature with very close numerical parameters has recently been reported on the basis of an experimental study on the inorganic disordered material *a*-Si:H.⁴⁰ This quantitative agreement between experimental data and the results of our calculations could mean that in the material studied in Ref. 40 the distribution of localized states can be described by the Gaussian disorder model with an uncorrelated distribution of localized states. Note that the effective temperature in our numerical calculations has been proven to be independent of the charge carrier concentration.

ACKNOWLEDGMENTS

F.J. wants to thank P. Bobbert for a valuable discussion regarding the iterative solution method. The calculations were performed at CSC, the Finnish IT center for science. The financial support from the Academy of Finland Project No. 107684 and the TEKES NAMU project, from the Deutsche Forschungsgemeinschaft, and that of the Fonds der Chemischen Industrie is gratefully acknowledged.

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