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EQUILIBRIUM CHARGE CARRIER MOBILITY IN DISORDERED HOPPING SYSTEMS

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It is shown that the charge carrier mobility in a positionally and energetically disordered hopping system can be evaluated by averaging either hopping rates or hopping times over the thermally equilibrium energy distribution of localized carriers. However, at variance with averaging hopping rates, averaging hopping times can be correct only if the energy dependence of the carrier energy relaxation time is also taken into consideration. The equilibrium carrier mobility was calculated by averaging hopping rates as a function of the temperature and concentration of localized sites. The obtained results prove that, in good quantitative agreement with both Monte Carlo simulations and experimental data, the temperature and concentration dependences of the mobility can be factorized, i.e. that the mobility can be represented as a product of two functions. The first function depends almost solely upon the temperature and reveals only a weak concentration dependence while the second one mainly governs the concentration dependence of the mobility and is almost independent of the temperature.

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1. Introduction

Basic characteristics of charge carrier transport in non-crystalline semiconducting materials are well known to be controlled by positional and energy disorder inherent in these materials [1-6]. Due to the disorder effects, most carriers are permanently localized in amorphous materials and the only feasible mode of charge transport is carrier jumps either directly between localized states or via a band of extended states if such states do occur in a given material. It is worth noting that the occurrence of the positional disorder will almost inevitably give rise to the energy disorder via the dependence of the potential energy of interaction upon the distance between interacting particles [7,8]. Nonetheless, charge carrier hopping can be simulated as *r*-hopping in a random system of localized states ignoring the energy disorder [9]. Trap-controlled transport implies a negligible contribution of direct tunneling jumps of carriers between localized states and, therefore, this transport mode can be considered as the energy-controlled hopping or ε-hopping. In most disordered materials both positional and energy disorder affect the carrier jump rate and this transport mode is described by models of $r\varepsilon$-hopping.

Most such models are based on the Miller-Abrahams [10] expression for the rate of carrier jumps, $\nu(r,E_{st},E_t)$, over the distance r between a starting state of the energy E_{st} and the target site of the energy E_t . This expression can be written as,

$$\nu(r,E_{st},E_t)=\nu_0\exp\left[-u(r,E_{st},E_t)\right],\quad u(r,E_{st},E_t)=2\gamma r+\frac{\eta(E_t-E_{st})}{kT}\quad , \tag{1}$$

where u is the hopping parameter, ν_0 the attempt-to-jump frequency, γ the inverse localization radius, T the temperature, k the Boltzmann constant, and η the unity step-function. Although the distance and

the energy difference between the starting and target sites almost similarly affect the jump rate there is one important difference that makes very different *r*- and ε-hopping modes.

The distance-dependent factor in Eq. (1) is completely symmetric, i.e. the distance between hopping sites similarly affects the rate of forth and back jumps. If a site, remote from any other site in a positionally random system of monoenergetic hopping sites, could be considered as a trap for carriers, the trapping time will be exactly equal to the release time. It means that it is equally difficult for a carrier to be released and to be trapped by such an isolated localized state. However, it is not the case for an energetically random system. While energetically upward jumps require thermal activation downward jumps imply dissipation of the excess energy via phonon emission. The former process is much slower than the latter and, therefore, the rates of forth and back jumps between two fixed states of different energies are normally very different. Consequently, a deep localized state can promptly capture a carrier and keep it localized over a long time. It is the strong asymmetry of trapping and release times that makes the energy disorder much more important as far as charge transport characteristics are concerned.

In the present paper we concentrate on the role of energy disorder in ε- and $r\varepsilon$-hopping. It will be shown that the carrier release time from deep localized states does control the effective carrier mobility in an energetically disordered system with a broad density-of-states (DOS) distribution under the non-equilibrium dispersive transport conditions. However, it is not the case for the equilibrium non-dispersive transport regime. Once the thermally equilibrium energy distribution of localized carriers is established the transport is controlled by both trapping and release of carriers whose energies are around the maximum of this distribution. Averaging the carrier hopping rates yields the equilibrium mobility which is in agreement with the exact analytic results obtained for ε-hopping [11] as well as with the results of Monte-Carlo simulations [12] and predictions of the effective medium theory [13-15] for $r\varepsilon$-hopping.

2. Equilibrium charge carrier mobility in an energetically disordered system

Mathematically, ε-hopping can be described as a set of multiple-trapping equations which relate the density of localized carriers to the density of carriers in extended states [11]. This is possible because the time carrier spends in extended states is typically by orders of magnitude shorter than the total jump time such that the sequence of carrier release, motion in the extended states, and trapping can be considered as a single jump. The ε-hopping equations for the localized carrier distribution, ρ, and the free carrier density, p_c , then read,

$$\frac{\partial \rho}{\partial t} = \frac{1}{\tau_0} p_c g(E) - \nu_0 \rho \exp\left(\frac{E}{kT}\right) \quad , \tag{2}$$

$$\frac{\partial p}{\partial t} + \mu_c F \frac{\partial p_c}{\partial x} = 0 \quad , \tag{3}$$

where p is the total density of carriers which is practically equal to the density of localized carriers,

$$p = p_c + \int_{-\infty}^{0} dE\, \rho(E) \approx \int_{-\infty}^{0} dE\, \rho(E) \quad , \tag{4}$$

t the time, x the coordinate, $g(E)$ the normalized DOS energy distribution, μ_c the mobility of carriers in extended states, F the electric field, ν_0 the attempt-to-jump frequency, and τ_0 the lifetime of carriers in extended states. Solving Eq. (2) under the thermal equilibrium conditions ($\partial\rho/\partial t = 0$) yields the equilibrium distribution ρ_{eq} as,

$$\rho_{eq} = \frac{1}{\nu_0 \tau_0} p_c g(E) \exp\left(-\frac{E}{kT}\right) \quad . \tag{5}$$

Substituting Eq. (5) into Eq. (4) leads to the following relation between the total density of carriers and the density of carriers in extended states:

$$p_c = \nu_0 \tau_0 \left[\int_{-\infty}^{0} dE\, g(E) \exp\left(-\frac{E}{kT} \right) \right]^{-1} p \quad . \tag{6}$$

This relation together with Eq. (3) allows to obtain a transport equation for the total density of carriers:

$$\frac{\partial p}{\partial t} + \mu F \frac{\partial p}{\partial x} = 0 \quad . \tag{7}$$

with the equilibrium carrier mobility in an energetically disordered system of localized states, μ, defined as [11],

$$\mu = \mu_c \nu_0 \tau_0 \left[\int_{-\infty}^{0} dE\, g(E) \exp\left(-\frac{E}{kT} \right) \right]^{-1} \quad . \tag{8}$$

In the following, we shall use this *exact* expression for the mobility as a reference for different approximate methods.

Now we consider two possible approaches to estimating the equilibrium mobility. The carrier mobility appears in the expressions for the current, $j = \mu F p$, and for the carrier transit time, $t_T = L / \mu F$, where L is the distance crossed by carriers over the time t_T. These two definitions are absolutely equivalent to each other once carriers are thermally equilibrated. Experimentally, it is rather difficult to evaluate the mobility from the current measurements because the density of drifting carriers is normally a poorly known value. Therefore, the mobility is usually determined from the time-of-flight measurements. However, theoretical evaluation of the mobility may be based on either of these two definitions and both must, of course, yield the same result.

The trap-controlled current can be estimated as the total rate of carrier jumps, ν_t, multiplied by the average distance crossed by a carrier during a single jump. The former should be calculated as the jump rate averaged over the energy distribution of localized carriers, while the latter is the mean free path of carriers in extended states, $\mu_c F \tau_0$. The normalized thermally equilibrium distribution function, f_{eq}, takes the form,

$$f_{eq}(E) = g(E) \exp\left(-\frac{E}{kT} \right) \left[\int_{-\infty}^{0} dE\, g(E) \exp\left(-\frac{E}{kT} \right) \right]^{-1} \quad . \tag{9}$$

Averaging the hopping rate over this distribution leads to the following expression for the equilibrium current,

$$j = \mu F p = \mu_c F \tau_0 \nu_0 \left[\int_{-\infty}^{0} dE\, g(E) \exp\left(-\frac{E}{kT} \right) \right]^{-1} p \quad , \tag{10}$$

where σ is the surface carrier density. Equation (10) yields exactly the same formula for the equilibrium mobility as was obtained from the exact analytic solution of the multiple-trapping equations. For a Gaussian DOS distribution,

$$g(E) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{E^2}{2\sigma^2} \right) \quad , \tag{11}$$

with the variance $\sigma >> kT$ both Eq. (8) and Eq. (10) yield,

$$\mu = \mu_c \nu_0 \tau_0 \exp\left[-\frac{\sigma^2}{2(kT)^2} \right] \quad . \tag{12}$$

At first glance this result seems to be puzzling if one tries to rationalize it as a carrier packet drift mobility. According to Eq. (9) the equilibrium distribution of localized carriers is also described by a Gaussian distribution of the same variance σ with the maximum of the distribution located at the energy $E_{max} = -\sigma^2/(kT)^2$. The value of E_{max} is two times larger than the activation energy of the equilibrium drift mobility $E_a = \sigma^2/2(kT)^2$. The implication is that jumps of an *absolute minority* of carriers, which are localized in states with energies around $-E_a$, seem to be responsible for the drift of

a carrier packet as a whole while the majority of carriers, localized around E_{max} , do not apparently participate in this process.

To resolve this puzzle one should consider the carrier release time *together with* following up equilibration. The average carrier release time, $\langle t_r \rangle$, is determined as,

$$\langle t_r \rangle = \frac{1}{\nu_0} \int_0^\infty dE \exp\left(-\frac{E}{kT}\right) f_{eq}(E) \quad . \tag{13}$$

For a Gaussian DOS distribution Eq. (13) yields,

$$\langle t_r \rangle \propto \exp\left[\frac{3}{2}\left(\frac{\sigma}{kT}\right)^2\right] \quad . \tag{14}$$

The average distance, Δx, which a carrier have crossed before it got trapped again must be proportional to the equilibration time, t_{eq}. The latter can be estimated as the carrier release time from a localized state of the energy E_m as,

$$\Delta x \propto t_{eq} \propto \exp\left[\left(\frac{\sigma}{kT}\right)^2\right] \quad . \tag{15}$$

The average carrier velocity during the round trip, $\langle v \rangle$, can be written as,

$$\langle v \rangle = \Delta x / \langle t_{eq} \rangle \propto \exp\left[-\frac{1}{2}\left(\frac{\sigma}{kT}\right)^2\right] \quad . \tag{16}$$

Equation (16) indicates that the average round-trip velocity is determined by the interplay of the carrier release and equilibration times. The result of this interplay strongly depends upon the DOS function. For an exponential DOS, $g(E) = (1/E_0)\exp(E/E_0)$ the round-trip velocity monotonously decreases with increasing depth of the starting site. This implies that carriers are always accumulated in deep states and that the regime of thermally equilibrium transport can never be established. This proves that, under equilibrium transport conditions, the drift mobility is controlled not only by the time of thermally activated jumps of carriers but also by the time of carrier trapping by states that are equally deep or deeper than the starting site. The latter time increases with energy even steeper than an exponential function. This is in contrast with predictions of *r*-hopping models in which the mobility and the current are shown to be controlled by the most difficult single carrier jump. In models of ε- and εr-hopping in disordered systems, neglecting the trapping time (or, equivalently, the time of downward jumps) as compared to the release time (or the time of upward jumps) is possible only under the dispersive transport conditions. The use of this approximation for the analysis of equilibrium transport characteristics leads to exponentially incorrect results [16].

It is worth noting that the integral in the right-hand side of Eq. (8) is formally equivalent to the carrier release time averaged over the DOS distribution rather than over the distribution of localized carriers. In the first place, it should be emphasized that such averaging has no physical meaning. The similarity is simply caused by the fact that the carrier release time is governed by the same Boltzmann exponential as the thermally equilibrium energy distribution of localized carriers. This similarity is specific for ε-hopping but is not valid in systems with both energy and positional disorder. For instance, an attempt to use such averaging in variable range hopping [16] is hardly justifiable. The results of this section prove that the equilibrium carrier mobility can be evaluated by averaging either hopping rates or hopping times. However, averaging hopping times should also account for the energy relaxation time while averaging hopping rates is not subject to this additional complication. In the following section we apply the latter approach for calculating the equilibrium carrier mobility in a positionally and energetically disordered hopping system.

3. Equilibrium mobility in a positionally random and energetically disordered hopping system

In a diluted random system of hopping sites most carriers will jump to nearest hopping neighbors i.e. to target sites characterized by minimum values of the hopping parameter *u* as counted

from starting sites. For a starting site of energy E_{st} , the average number, $n(E_{st},u)$, of hopping neighbors, whose hopping parameters are not larger than a given value u , is given by,

$$n(E_{st},u) = 4\pi N_t \int_0^{u/2\gamma} dr\, r^2 \int_{-\infty}^{E_{st}+kT(u-2\gamma r)} dE_t\, g(E_t) = \\ = \frac{\pi N_t}{6}\left(\frac{u}{\gamma}\right)^3 \left[\int_{-\infty}^{E_{stt}} dE_t\, g(E_t) + \int_{E_{st}}^{E_{st}+kTu} dE_t\, g(E_t) \left(1 - \frac{E_t - E_{st}}{kTu}\right)^3 \right] , \quad (17)$$

where N_t is the total density of hopping sites. The probability density, $w(E_{st},u)$, of finding a nearest hopping neighbor of the hopping parameter u is determined by the Poisson distribution as,

$$w(E_{st},u) = \exp\left[-n(E_{st},u)\right] \frac{\partial n(E_{st},u)}{\partial u} \quad . \quad (18)$$

Equation (18) forms a basis for calculating the average hopping parameter, $\langle u \rangle(E_{st})$, for carrier jumps from a starting site of energy E_{st}. Using Eq. (18) as a distribution function for averaging the hopping parameter yields,

$$\langle u \rangle(E_{st}) = \int_0^{\infty} du\, u \exp\left[-n(E_{st},u)\right] \frac{\partial n(E_{st},u)}{\partial u} = \int_0^{\infty} du \exp\left[-n(E_{st},u)\right] \quad . \quad (19)$$

Evaluating the typical jump distance as $\langle u \rangle / 2\gamma$ and averaging over E_{st} under thermal equilibrium conditions leads to the following expression for the equilibrium mobility,

$$\mu = \frac{e\nu_0}{kT(2\gamma)^2} \left[\int_{-\infty}^{\infty} dE_{st}\, g(E_{st}) \exp\left(-\frac{E_{st}}{kT}\right) \right]^{-1} \\ \times \int_{-\infty}^{\infty} dE_{st} \exp\left[-\langle u \rangle(E_{st})\right] \left[\langle u \rangle(E_{st})\right]^2 g(E_{st}) \exp\left(-\frac{E_{st}}{kT}\right) \quad . \quad (20)$$

Temperature dependence of the mobility, calculated from Eq. (20) for a Gaussian DOS distribution, is shown in Fig. 1 for different values of the total density of localized states. All the curves feature almost perfect straight lines if plotted as log μ *vs* $1/T^2$. Although the absolute value of the mobility does strongly decrease with decreasing N_t the slopes of log μ *vs* $1/T^2$ curves increase by only less than 15 % when the density of hopping sites decreases by four orders of magnitude. This implies that the equilibrium mobility can be rather accurately represented in a factorized form as,

$$\mu = \mu_0 \varphi(N_t) \exp\left[-\left(\frac{c\sigma}{kT}\right)^2\right] \quad , \quad (21)$$

with the numeric parameter c changing from 0.59 at $N_t = 10^{22}$ cm^{-3} to 0.68 at $N_t = 10^{18}$ cm^{-3}. Both the form of Eq. (21) and the value of the parameter c are in good quantitative agreement with the results of Monte Carlo simulations (c = 0.67) and predictions of the effective medium model (c = 0.64). It is worth noting that a simpler model, based on averaging hopping rates and employing configurational averaging, also yielded the mobility of the form given by Eq. (21) with c = 0.5. This value seems to be the lower limit of c that can be reached at high densities of localized states.

The dependence of the equilibrium mobility upon the total density of hopping sites is illustrated in Fig. 2 parametric in the temperature. The curves plotted in this figure indicate that the concentration dependence of the mobility does obey typical for hopping transport an exponential law,

$$\varphi(N_t) = \exp\left(-\frac{b\gamma}{N_t^{1/3}}\right) \quad , \quad (22)$$

with the numeric parameter b being rather weakly dependent upon the temperature. The values of this parameter are indicated for each curve in Fig. 2.

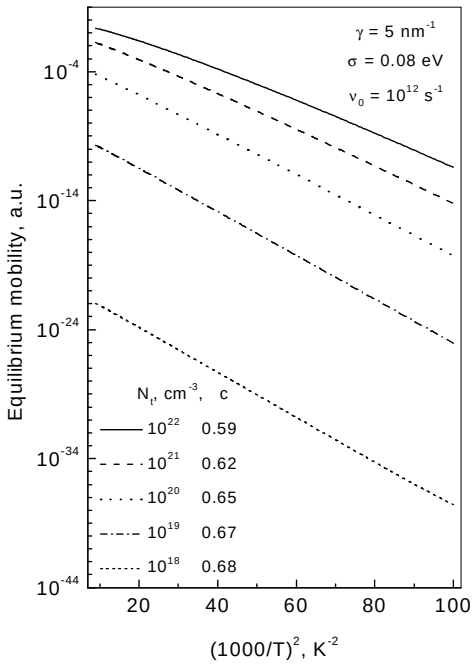

Fig. 1. Temperature dependence of the equilibrium drift mobility calculated from Eq. (20) for a hopping system with a Gaussian DOS distribution of localized states.

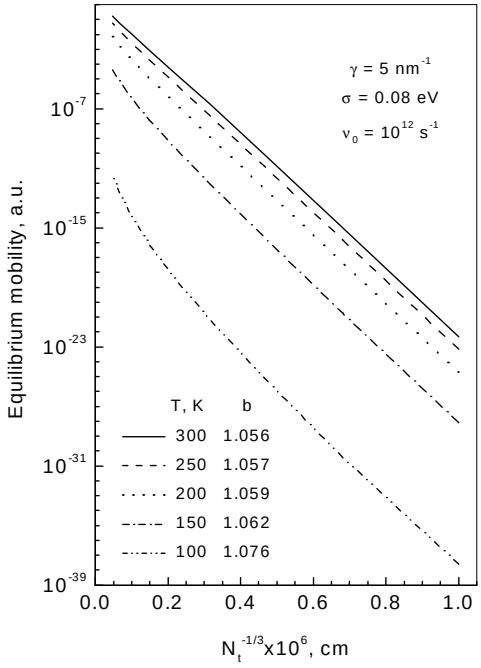

Fig. 2. Concentration dependence of the equilibrium drift mobility calculated from Eq. (20) for a hopping system with a Gaussian DOS distribution of localized states.

The above consideration disregards repeated carrier jumps between two accidentally close hopping neighbors both of which are isolated from other localized states. Although such jumps are important as far as ac conductivity is concerned they do not contribute to the dc current and, concomitantly, to the equilibrium carrier mobility. Accounting for isolated pairs and, more general, for isolated clusters of hopping sites would result in decreasing effective density of states N_t and, therefore, to increasing values of the coefficients *b* and *c*.

4. Effective transport energy

The first term in the right-hand side of Eq. (17) gives the number of target states which are deeper than the starting site and the second one describes the number of shallower states. The former is important as far as downward carrier jumps are concerned while the latter governs the rate of upward jumps. In the present paper we consider the equilibrium transport conditions under which both drift and diffusion are controlled by upward hopping and, concomitantly, in the following we concentrate on consideration of this hopping mode. Making out the replacement of variables,

$$u = \frac{E_{tr} - E_{st}}{kT} \quad , \tag{23}$$

yields the following expression for the number of shallower hopping neighbours of a starting site of energy E_{st} :

$$n(E_{st}, E_{tr}) = \frac{\pi}{6}(\gamma kT)^{-3} \int_{E_{st}}^{E_{tr}} dE_t \, g(E_t)(E_{tr} - E_t)^3 \quad . \tag{24}$$

An upward carrier jump from a starting site is possible if there is at least one such hopping neighbour, i.e. from $n(E_{st}, E_{tr}) = 1$ on. The use of this condition in Eq. (24) leads to the following transcendental equation for the energy of the most probable upward jumps:

$$\int_{E_{st}}^{E_{tr}} dE_t \, g(E_t)(E_{tr} - E_t)^3 = \frac{6}{\pi}(\gamma kT)^3 \quad . \tag{25}$$

If the DOS distribution decreases with energy faster than $|E|^{-4}$ than (i) the value of the integral in the left-hand side of Eq. (25) is practically independent of the lower bound of integration for sufficiently deep starting sites and (ii) a major contribution to the integral comes from states with energies around E_{tr} . Physically, it means that target sites for thermally assisted upward carrier jumps are located around the energy E_{tr} independent of the energy of starting sites and, therefore, Eq. (5) reduces to:

$$\int_{-\infty}^{E_{tr}} dE_t \, g(E_t)(E_j - E_t)^3 = \frac{6}{\pi}(\gamma kT)^3 \quad . \tag{26}$$

In order to illustrate the efficiency of the transport energy concept we apply it to the calculation of the equilibrium carrier mobility, μ , in a positionally random hopping system with a Gaussian DOS distribution at weak external electric fields. Estimating the equilibrium diffusivity as a squared typical jump distance multiplied by the average jump frequency and using the Einstein relation yields,

$$\begin{aligned}\mu_{eq} &= \frac{e\nu_0}{kT}\left[\int_{-\infty}^{\infty} dE g(E)\exp\left(-\frac{E}{kT}\right)\right]^{-1}\left[\int_{-\infty}^{E_{tr}} dE g(E)\right]^{-2/3}\int_{-\infty}^{E_{tr}} dE g(E)\exp\left(-\frac{E}{kT}\right)\exp\left(-\frac{E_{tr}-E}{kT}\right) = \\ &= \frac{e\nu_0}{kT}\left[\int_{-\infty}^{\infty} dE\, g(E)\exp\left(-\frac{E}{kT}\right)\right]^{-1}\left[\int_{-\infty}^{E_{tr}} dE\, g(E)\right]^{1/3}\exp\left(-\frac{E_{tr}}{kT}\right)\end{aligned} \quad . \tag{27}$$

Equation (27) is remarkably similar to the expression for the trap-controlled equilibrium carrier mobility. The only two differences are the occurrence of a temperature-dependent transport energy instead of a fixed mobility edge and a weakly temperature-dependent mean jump distance instead of a fixed mean free path of delocalized carriers. For a Gaussian DOS function Eq. (27) reduces to,

$$\mu_{eq} = \frac{e\nu_0}{\sqrt[3]{2}kTN_t^{2/3}}\exp\left[-\frac{\sigma^2}{2(kT)^2}\right]\left[1+\mathrm{Erf}\left(\frac{E_{tr}}{\sqrt{2}\sigma}\right)\right]^{1/3}\exp\left(-\frac{E_{tr}}{kT}\right) \quad , \tag{28}$$

where Erf is the error function. Further simplification of this equation is possible at high T and/or low N_t . Substituting the high-temperature/low-concentration expression for E_{tr} from Eq. (26) into Eq. (28) yields,

$$\mu_{eq} = \frac{e\nu_0}{kTN_t^{2/3}}\exp\left[-\left(\frac{6\gamma^3}{\pi N_t}\right)^{1/3}\right]\exp\left[-\frac{\sigma^2}{2(kT)^2}\right] \quad . \tag{29}$$

Equation (29) once again proves that the temperature and concentration dependencies of the mobility are factorized at high temperatures and/or in diluted hopping systems. This result does suggest that these dependencies will also be almost factorized at lower temperatures and in systems with higher concentrations of hopping sites.

5. Conclusions

Charge carrier mobility in disordered hopping systems can be evaluated by averaging either hopping rates or hopping times over the thermally equilibrium energy distribution of localized carriers. However, at variance with averaging hopping rates, averaging hopping times can be correct only if the energy dependence of the carrier energy relaxation time is also taken into consideration. Neglecting this relaxation would lead to exponentially incorrect expressions for the equilibrium mobility and diffusivity.

The equilibrium carrier mobility was calculated by averaging hopping rates as a function of the temperature and concentration of localized sites in an energetically disordered and positionally

random hopping system. The obtained results prove that, in good quantitative agreement with both Monte Carlo simulations and experimental data, the temperature and concentration dependences of the mobility can be factorized, i.e. that the mobility can be represented as a product of two functions.

The first one depends almost solely upon the temperature and reveals only a weak concentration dependence while the second one mainly governs the concentration dependence of the mobility and is almost independent of the temperature. These results support the predictions of the simpler models based on the effective medium approximation and configurational averaging.

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