Theoretical description of hopping transport in disordered materials

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Abstract

Incoherent hopping conduction has been revealed as the electronic transport mechanism in lightly doped microcrystalline semiconductors at low temperatures. Although microcrystalline materials should be considered as geometrically inhomogeneous systems, essential features of the photoconductivity and dark conductivity in such systems can be successfully described by theoretical models based on a homogeneous distribution of localized states. We review several theoretical approaches suggested so far for the description of hopping transport in disordered materials. In order to verify the validity of analytical approaches, we have performed a series of straightforward computer simulations for the percolation problem in hopping conduction. The simulation results support the analytical approaches based on the transport energy concept and on percolation arguments for the description of the dark conductivity at low temperatures.
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1. Introduction

Microcrystalline and polycrystalline materials are intensively studied experimentally and theoretically in the last decade because of their potential for applications in high-stability and high-efficiency low cost electronic devices, such as solar cells and thin-film transistors. Among other materials hydrogenated microcrystalline silicon (μc-Si:H) attracts much attention due to its compatibility with various device elements based on silicon technology. In particular, transport properties of charge carriers in μc-Si:H is a subject of intensive study. While quite a number of experimental studies of electronic transport in μc-Si:H have been carried out, the theoretical literature on this subject is comparatively scarce and we are not aware of a consistent theoretical description of the transport mechanism in μc-Si:H. In several previous studies it has been claimed that transport properties of not heavily doped μc-Si:H closely resemble those of the hydrogenated amorphous silicon (a-Si:H) [1–5]. Indeed, at very low temper-

atures $T<50$ K the photoconductivity (PC) in μc-Si:H does not depend on temperature while at temperatures above $T_c \sim 50$ K the PC strongly increases with T [1,2,4,5]. Such behaviour is well known for years for a-Si:H [6,7]. Moreover such temperature dependence of the PC has been observed also for other disordered materials, such as a-C/H [8], a-Se and many others [9]. One of the most important characteristics of a material with respect to its applications in optoelectronic devices, such as solar cells, is the mobility (μ)–lifetime (τ)–efficiency (η) product $\mu\tau\eta$ which is determined as the magnitude of the PC σ_{ph} divided by the elementary charge e and by the generation rate of electron–hole pairs G: $\eta\mu\tau=\sigma_{ph}/eG$. The value of the $\eta\mu\tau$ product in μc-Si:H at low temperatures has been reported to be close to 10^{-11} cm^2 V^{-1} [2], which is remarkably close to the value of this product in a-Si:H [6,7]. Another important characteristics of the PC is its dependence on the generation rate G. In μc-Si:H this dependence has a power form $\sigma_{ph} \propto G^{\gamma}$, where the power exponent γ is close to unity at low temperatures and it decreases with T toward values around 0.5 to 0.7 at room temperatures [2,4]. This behaviour is absolutely similar to that in a-Si:H as has been already mentioned in the literature [2,4]. Of course, such a remarkable similarity

between various characteristics and even numerical values for the PC in μc-Si:H and in a-Si:H leads to the conclusion that a similar transport mechanism is active in both materials [1–5]. This conclusion is not trivial since μc-Si:H is a geometrically strongly inhomogeneous material containing both microcrystalline and amorphous regions, while a-Si:H is rather believed to be spatially homogeneous. Claiming that charge carrier transport in μc-Si:H at least at low temperatures can be described in a homogeneous model [1–5,10] one assumes that transport is via hopping transitions between localized states in amorphous tissue regions on the boundaries of micro-crystalline regions. This must be true even for materials with rather high crystallinity above 90% [1,2]. Below we describe this transport mechanism using several concepts suggested so far for description of charge carrier transport in homogeneous disordered semiconductors. We also show the results of new computer simulations performed in order to verify the analytical approaches. Our computer simulations strongly support the analytical approach based on percolation theory and that based on the transport energy concept for the description of the dark conductivity at low temperatures.

The paper is organized as follows. In Section 2 the theory for PC at very low temperatures is given. In Section 3 the concept of transport energy is described with its application to the PC. Several other analytical approaches are also discussed in this section. In Section 4 the percolation approach to the calculation of the dark conductivity is described and the results of computer simulations are presented and compared with those of analytical theories. Conclusions are gathered in Section 5.

2. Steady-state PC at low temperatures

In order to formulate a theoretical picture for the PC, one should study the fate of an electron–hole pair generated by light. Let us consider such a pair generated at or just below the mobility edge. Electron and hole are generated quite close together because of the exponential decay of their overlap integral with distance. An electron, which we consider as more mobile than a hole, can take part in two competing processes. It can hop down in energy (upward hops are not possible at $T=0$) to a nearest localized state in the band tail at distance r with the rate

$$v_{\rm h}(r) = v_0 \exp\left(-\frac{2r}{a}\right), \tag{1}$$

where v_0 is the attempt-to-escape frequency of the order of the phonon frequency, or it can recombine with the hole at a rate

$$v_{\rm r}(R) = \tau_0^{-1} \exp\left(-\frac{2R}{a}\right), \tag{2}$$

where R is the electron-hole separation, a is the local-ization length and τ_0 is some preexponential factor for the recombination process. If this process is radiative, then $\tau_0 \approx 10^{-8}$ s is given by the typical dipole radiation lifetime. If the recombination is nonradiative, the only fact we know is that $\tau_0 \gg v_0^{-1}$ because of the multiphonon nature of the recombination event. We assume that the localization radius of the hole is smaller than that of the electron, so both rates $v_{\rm h}$ and $v_{\rm r}$ are determined by the same quantity a. It has been shown [11,12] that the probability for an electron–hole pair to avoid recombination and to survive in the hopping relaxation process to the electron–hole separation R is described at $R \gg R_{\rm c}$ by a power law

$$\eta(R) = A\left(\frac{R_{\rm c}}{R}\right)^{\beta} \tag{3}$$

with $\beta \approx 1.2$ as a critical index, $A \approx 3$, and the character-istic length $R_{\rm c}$ determined as

$$R_{\rm c} = \frac{a}{2} \ln(v_0 \tau_0). \tag{4}$$

Charge carriers that recombine geminately do not contribute to the steady-state PC [6]. Non-geminate recom-bination arises when an electron–hole pair succeeds to separate by hopping to a distance $\sim n^{-1/3}/2$, where n is the steady-state concentration of electrons (or holes) under the given generation rate G.

The steady-state concentration n is determined by the equation [11,12]

$$G\eta\left(\frac{n^{-1/3}}{2}\right) = \frac{n}{\tau_0} \exp\left(-\frac{n^{-1/3}}{a}\right) \tag{5}$$

with the solution

$$n(G) = [aL(G)]^{-3}, \tag{6}$$

where $L(G)$ satisfies the equation

$$L = \ln\left\{ \left[\tau_0 G L^2 a^3 \ln(v_0\tau_0)\right]^{-1} \right\}. \tag{7}$$

The left-hand side of Eq. (5) represents the generation rate of those carriers which survive through hopping to the distance $\sim n^{-1/3}/2$ and avoid, therefore, the geminate recombination and hence contributing to photocurrent. The photocurrent density is equal to

$$j = G\eta\left(n^{-1/3}/2\right)d, \tag{8}$$

where d is the typical dipole moment of a pair at the recombination event induced by the external electric field with strength E [11,12]. Assuming for simplicity the exponential shape of the density of states (DOS) in the band tail

$$g(\varepsilon) = \frac{N_0}{\varepsilon_0} \exp\left(\frac{\varepsilon}{\varepsilon_0}\right) \tag{9}$$

with the total concentration of localized states N_0 and the energy ε assumed zero at the mobility edge, one obtains [11,12]

$$d = \frac{1}{3} \frac{e^2 E n^{-2/3}}{4\varepsilon_0} . \tag{10}$$

Combining these equations one obtains for the PC $\sigma_{ph}=j/E$ the following expression

$$\sigma_{ph} = G\eta \left(\frac{n^{-1/3}}{2}\right) \frac{e^2 n^{-2/3}}{12\varepsilon_0}, \tag{11}$$

where the steady-state concentration of electrons (holes) n is determined by Eq. (5). Inserting Eqs. (5)–(7) into Eq. (11) one obtains the power law dependence of the PC on the generation rate $\sigma_{ph}=G^{\gamma}$ with $\gamma=1-(1/L)$. Comparing these results with experimental data [1,2,4–8] we obtain $\sigma_{ph}/eG \approx 10^{-11}$ cm²/V, $\gamma=0.94$ for $G=10^{20}$ cm⁻³ s⁻¹ for a reasonable choice of material parameters $a=1$ nm, $\varepsilon_0=0.025$ eV and $\nu_0\tau_0=10^4$. Experimentally one finds for µc-Si:H $\sigma_{ph}/eG \approx 10^{-11}$ cm²/V and $\gamma=0.96$ [1,2,4,5] and also rather close values for *a*-Si:H [6,7].

3. The concept of the transport energy (TE) applied to the steady-state PC

Before starting a discussion on the temperature-dependent PC in µc-Si:H, let us recall the very useful concept of the so-called transport energy, which will help us in calculations of $\sigma_{ph}(T)$. The crucial role of a particular temperature-dependent energy level $\varepsilon_t(T)$ in the band tail of disordered materials was first recognized by Grünewald and Thomas [13] in their numerical analysis of the equilibrium variable-range hopping conductivity. They obtained numerically that carriers with energy close to $\varepsilon_t(T)$ dominate the equilibrium transport of electrons in the band tail. This result was confirmed later by analytical calculations of Shapiro and Adler [14], who gave a formula for $\varepsilon_t(T)$. A rather different problem of non-equilibrium energy relaxation of electrons by hopping through the band tail states was solved in the same year by Monroe [15]. He showed that an electron, starting from the mobility edge, most likely makes a series of hops downward in energy until it reaches some particular energy $\varepsilon_t(T)$, where the relaxation process changes drastically. In the vicinity and below $\varepsilon_t(T)$ the hopping resembles a multiple trapping process with the mobility edge replaced by $\varepsilon_t(T)$. Monroe called $\varepsilon_t(T)$ the transport energy (TE). Later Shklovski et al. [16] showed that the same energy level $\varepsilon_t(T)$ determines both recombination and transport of electrons under steady-state photogeneration and hence this energy determines the temperature dependence of the PC. Baranovskii et al. [17,18] suggested an alternative derivation of $\varepsilon_t(T)$ and they extended this derivation to other shapes of the DOS than the exponential one, for example for a Gaussian DOS [19]. In particular, it has been shown that if

the energy dependence of the DOS is essentially weaker than an exponential one, the concept of the TE fails to describe the hopping process. Unfortunately, statements can be found in the literature that the TE concept is applicable to practically all realistic DOS distributions [20]. One has to emphasize that the latter is not true, as clearly shown in [19] for the DOS of the shape $g(\varepsilon) \propto \exp[(\varepsilon/\varepsilon_0)^{1/2}]$ known to be valid in solid solutions [19]. We assume, following [5], that the DOS in µc-Si:H is exponential and hence the concept of the TE is applicable to this material. The TE is determined for exponential DOS by equation [13–18]

$$\varepsilon_t = -3\varepsilon_0 \ln\left[\frac{3\varepsilon_0(4\pi N_0/3)^{1/3}a}{2kT}\right]. \tag{12}$$

Let us estimate the temperature T_c at which the PC first becomes temperature dependent. At very low temperatures electrons perform the energy-loss hopping described in Section 2 until they recombine at some typical energy ε_F^0 where the hopping rate ν_h is comparable with the recombination rate ν_r. This happens when electrons fill up the bottom of the band tail to a level

$$\varepsilon_F^0 = -\varepsilon_0 \ln\left(\frac{N_0}{n}\right), \tag{13}$$

which can be considered as the quasi-Fermi level at low temperatures. If the TE is deeper in the tail than ε_F^0, the temperature does not influence the relaxation and recombination processes anyhow and the PC does not depend on temperature. With rising T the TE moves toward more shallow states in the band tail and at some particular temperature T_c it crosses the quasi-Fermi level ε_F^0. At this temperature T_c the PC becomes temperature dependent. Equating ε_F^0 to $\varepsilon_t(T_c)$ one obtains from Eqs. (6), (7), (12), (13) the equation [16]

$$T_c = \frac{3}{2}\varepsilon_0\left(\frac{4\pi}{3}\right)^{1/3}\frac{1}{L(G)}. \tag{14}$$

Using the values of material parameters given above in Section 2, one obtains an estimate $T_c \approx 45$ K in excellent agreement with experimental observations for µc-Si:H [1,2,4,5].

At higher temperatures, $T>T_c$, the TE lies above the low-temperature recombination energy ε_F^0 and the physical picture of recombination and hopping transport differs drastically from that at low temperatures. At $T>T_c$, the hops of electrons upward in energy to the vicinity of the TE determine both transport and recombination of photo-generated electrons and holes. The corresponding comprehensive theory can be found in [16] resulting in the expression

$$\sigma_{ph} = \frac{e^2 G\eta(r_t)r_t^4}{6\pi\varepsilon_0 R_r a}\left[\frac{4\pi R_r D(r_t)W}{G\eta(r_t)r_t^6\varepsilon_0}\right]^{\frac{kT}{\varepsilon_0+kT}}, \tag{15}$$

where r_t, $D(r_t)$, $W = (6\varepsilon_0 \cdot kT)^{1/2}$ and $R_r = r_t^2/3a$ are the typical distance between the tail states below the TE, the diffusion coefficient of electrons on the TE, the effective width of the TE, and the effective recombination radius on the TE, respectively [16]. A comparison of Eq. (15) with experimental data for a-Si:H gives very nice results [16], which automatically means that this formula is valid for description of the PC in μc-Si:H, since experimental data on the temperature dependent PC in μc-Si:H are very close to those in a-Si:H [1,2,4,5].

4. Theoretical description of the dark conductivity in the hopping regime

The most transparent approach to the dark conductivity (DC) in disordered systems was suggested by Grünewald and Thomas [13], who discovered the significance of the TE for description of variable-range-hopping in band tails with exponential DOS. It was assumed that the jump rate of a charge carrier from a filled site i to an empty site j is of the Miller–Abrahams type

$$v_h(r, \varepsilon_i, \varepsilon_j) = v_0 \exp\left(-\frac{2r}{a}\right) \exp\left[-\frac{(\varepsilon_j - \varepsilon_i + |\varepsilon_j - \varepsilon_i|)}{2kT}\right],$$
(16)

where ε_i and ε_j are the site energies. Taking into account the concept of the TE described in Section 3, the result for σ_d reads

$$\sigma_d = \frac{e^2 v_0 r_t^2}{kT} \exp\left(-\frac{2r_t}{a}\right) \int_{-\infty}^{\varepsilon_t} \frac{d\varepsilon \cdot g(\varepsilon)}{1 + \exp[(\varepsilon - \varepsilon_F)/kT]}$$
$$\times \exp\left(\frac{\varepsilon - \varepsilon_t}{kT}\right) + \frac{Ce^2 v_0}{\varepsilon_0} \int_{\varepsilon_t}^{0} \frac{d\varepsilon \cdot g(\varepsilon)}{1 + \exp[(\varepsilon - \varepsilon_F)/kT]}$$
$$\times \exp\left[-\frac{2r(\varepsilon)}{a}\right],$$
(17)

where ε_F is the Fermi level and the typical hopping distance $r(\varepsilon)$ at energy ε can be estimated as

$$r(\varepsilon) = \left[\left(\frac{4\pi}{3B}\right) \int_{-\infty}^{\varepsilon} g(\varepsilon) d\varepsilon\right]^{-1/3}$$
(18)

with $r_t = r(\varepsilon_t)$ being the hopping distance at and below the TE. The coefficient $C = 2.3$ in Eq. (17) is due to the modified form of the Einstein relation for energy-loss hopping [21]. The coefficient $B = 2.7$ in Eq. (18) takes into account the necessity to fulfil the percolation criterion [22]. The first term in the right-hand side of Eq. (17) was recently suggested for calculation of σ_d by Arkhipov et al. [23], though unfortunately without the very important hopping term $\exp(-2r_t/a)$. Thus the hopping nature of charge transport was ignored in [23].

If the position of the Fermi level is known, one can estimate the total concentration of charge carriers as

$$n = \int_{-\infty}^{\infty} \frac{g(\varepsilon) d\varepsilon}{1 + \exp[(\varepsilon - \varepsilon_F)/kT]}$$
(19)

and calculate the DC as $\sigma_d = e \cdot n \cdot \mu$, where μ is a drift mobility of a charge carrier. The most straightforward way to estimate μ is to average times of hopping transitions over energy states below the TE, since only these states are essential for transport [24]. The mobility is equal to $\mu \approx (e/kT) r_t^2 <t>^{-1}$, where $<t>$ is the hopping time averaged over successive hops of charge carrier. For DOS distributions decaying into the mobility gap more steeply than the exponential function described by Eq. (9) one can estimate $<t>$ as [24]

$$<t> = \left(\int_{-\infty}^{\varepsilon_t} g(\varepsilon) d\varepsilon\right)^{-1} \times \int_{-\infty}^{\varepsilon_t} v_0^{-1} g(\varepsilon) \exp\left[\frac{2r_t}{a} + \frac{\varepsilon_t - \varepsilon}{kT}\right] d\varepsilon.$$
(20)

For exponential DOS described by Eq. (9) and for all DOS functions with a weaker energy dependences one cannot use Eq. (20) for calculations of $<t>$. Instead one should average hopping times taking into account the filling of energy states by the charge carriers.

It is worth noting that there are a lot of inconsistencies in the scientific literature on the question what to average and how to average. For example, Arkhipov et al. [23] recently suggested a formula with $\mu \approx (e/kT) r_t^2 <v>$, which is surely at variance with the correct result $\mu \approx (e/kT) r_t^2 <t>^{-1}$ for $v = t^{-1}$, provided the averaging $<\ldots>$ would mean in both cases the same. It has been also claimed by Fishchuk et al. [25] that the averaging of hopping rates can lead to a correct result. We would like to emphasize, however, that the result in [25] for the essential quantity $<t>$ in the linear regime literally coincides with Eq. (20), as the authors remark themselves, so it is difficult to understand why they claim the validity of the rate averaging. The situation with [23] is even more puzzling. The authors simply define the average rate $<v>$ in a new way, namely as $<v> = \mu/(e \cdot r_t^2/kT)$ with $\mu = \sigma_d/(e \cdot n)$, where σ_d is the conductivity and n is described by Eq. (19). This definition of $<v>$ is in contrast to the well accepted terminology from textbooks. We believe that correct equations are more important than the matter of terminology. However, it is worth noting that equations in [26,27] based on the averaging of hopping rates in the traditional meaning of the averaging procedure from textbooks [28] lead to erroneous results, as shown in [28,29].

Although the above approach based on the concept of the TE is transparent, it is not the most accurate for calculations of the DC. There is a more accurate, though less transparent approach based on the percolation theory. The details of the percolation approach can be found, for example, in [28]. Its application to variable-range hopping in exponential band tails has been first studied by Grünewald and Thomas [13] and later developed by Vissenberg and Matters [30]. In this

approach one treats the transition rate v_{ij} in all pairs of sites taking into account both the quantum mechanical transition probability determined by Eq. (16) and also the probability that one of the sites in a pair is filled by a charge carrier, while the other site is empty [28]:

$$v_{ij} = v_0 \exp\left(-\frac{2r}{a}\right) \exp\left[-\frac{|\varepsilon_i - \varepsilon_F| + |\varepsilon_j - \varepsilon_F| + |\varepsilon_j - \varepsilon_i|}{2kT}\right]$$
$$\equiv v_0 \exp(-\xi_{ij}). \qquad (21)$$

Connecting only sites with $\xi_{ij} < \xi$ and increasing the value of ξ, one determines the value ξ_c at which the infinite percolation cluster of interconnected bonds appears, which is responsible for the hopping motion of charged carriers in the dc regime. The critical value ξ_c determines the exponential temperature and concentration dependences of transport coefficients

$$\sigma_d = \sigma_0 \exp(-\xi_c), \qquad (22)$$

where σ_0 is a prefactor. The problem is to calculate the exponent ξ_c. Vissenberg and Matters [30] calculated σ_d using the classical percolation criterion that B_c determined as the number of valid bonds per site at the percolation threshold is equal to 2.8 as explained in [28]. Another criterion was suggested recently by Arkhipov et al. [31], who assumed that one bond per site would be enough. It is very easy to see that the latter approach is erroneous, since it does not reproduce the well-known result for the limit of high temperatures. In such a limit, only spatial disorder is significant, whereas energy disorder does not play any role if $kT \gg \varepsilon_0$. In such a case $\sigma_d \propto \exp\{-1.73(N_0 a^3)^{-1/3}\}$ [28] at variance to the result of Arkhipov et al. [31].

In order to verify the analytical percolation approach used by Vissenberg and Matters [30] and also in order to check the accuracy of the analytical approach based on the TE, we performed a series of straightforward computer simulations of the percolation problem for the DC. The simulation was based on a straightforward Monte Carlo algorithm directed to finding the percolation threshold in a system of randomly placed localization sites with energy distribution described by Eq. (9). The sites were placed in a cube with a length chosen to provide a large number of sites m for a given concentration N_0. No correlation between the spatial positions of sites and their energies were assumed. A hopping parameter ξ_{ij} was calculated for each pair of sites according to Eq. (21) and the percolation threshold $\xi_c(m)$ was found, which provides a continuous path through the system via states with $\xi_{ij} < \xi_c(m)$. Five runs of the algorithm were performed for each set of the system parameters with different random generator seed numbers and the results were averaged over these runs showing good statistics. The accuracy in determination of $\xi_c(m)$ for each run was $\xi_c(m) \pm 0.1$. The standard deviations of the $\xi_c(m)$ values for single runs never exceeded 0.2. Calculations for each set of parameters were performed for $m = 10^5$, $2 \cdot 10^5$ and $5 \cdot 10^5$ and the value of ξ_c

for the infinite system was determined via linear extrapolation of $\xi_c(m)$ as a function of $1/m$.

In Fig. 1 the results of the computer simulation for ξ_c as a function of temperature are shown (squares) in comparison with the results of the percolation analytical approach of Vissenberg and Matters [30] (solid line) and with the results obtained by the TE approach according to Eq. (17) (dashed line). The simulation parameters, such as N_0, n, a, ε_0, were chosen equal to those given in [30]. Since the analytical approach based on the TE concept does not allow us to determine the preexponential factor σ_0 in Eq. (22), we adjusted this factor in order to equate ξ_c in the TE description to that obtained in the simulation for the lowest considered temperature $T = 55$ K. Hence only the slope in the temperature dependence of ξ_c can be found in the framework of the TE approach. Furthermore, the TE approach for the chosen material parameters is self-consistent only at rather low temperatures $T < 70$ K, since for higher temperatures the assumptions used for the derivation of the TE are not valid anymore. Comparison with the computer simulation in Fig. 1 shows that in the range of low temperatures, the TE approach gives a correct temperature dependence of the exponential factor for the DC. The same can be generally claimed for the analytical approach based on the percolation theory [30], although this analytical approach is self-consistent in a much broader temperature range than the TE description. The analytical percolation approach is supposed to be accurate enough at $kT < \varepsilon_0$ and $-\varepsilon_F \gg \varepsilon_0$ [30]. These conditions are fulfilled in a very broad temperature range. Nevertheless an essential deviation between the results of the analytical percolation approach and those of computer simulations can be recognized in Fig. 1 at temperatures above ~100 K, while at low temperatures there is a good agreement between the analytical theory and computer simulations. On the basis of this comparison one should conclude that while analytical approaches based on
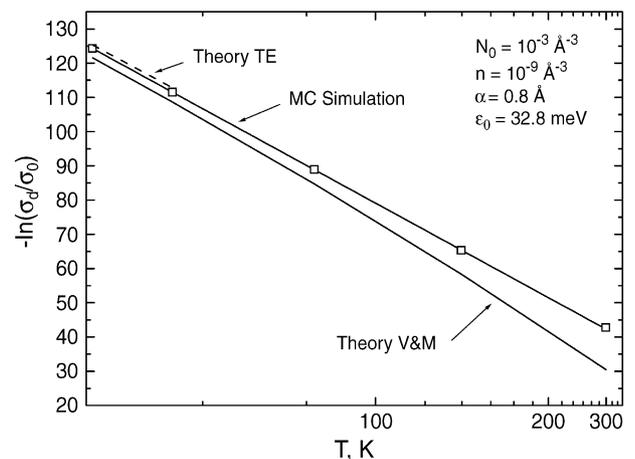

Fig. 1. Temperature dependence of the hopping conductivity for exponential density of states. The squares represent the results of the simulation, the dashed line is according to Eq. (17) for the TE theory, and the solid line represents the analytical percolation theory of Vissenberg and Matters [30].

the TE concept or on the percolation theory reasonably describe the temperature dependence of the dark conductivity, at high enough temperatures only computer simulations provide a sufficiently accurate result.

5. Conclusions

Energy-loss hopping of charge carriers at very low temperatures and the temperature-assisted hopping of charge carriers via localized band-tail states at higher temperatures are known to be the dominant transport mechanisms in disordered materials. Analytical theories for the description of such transport are reviewed with application to amorphous and microcrystalline semiconductors. Particular attention is devoted to the analytical theory based on the concept of the transport energy due to its transparency in description of the most pronounced transport phenomena. A more sophisticated theoretical approach based on the percolation theory is also described with applications to the dark conductivity. Monte Carlo computer simulations have been performed in order to check the validity of these analytical approaches. The results of the comparison show that the analytical approaches provide rather accurate results for the temperature dependence of the dark conductivity at low temperatures, while at high enough temperatures only the results of computer simulations can be considered as reliable.

Acknowledgements

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