

Some remarks on ac conduction in disordered solids

Jeppe C. Dyre

IMFUFA, Roskilde Universitetscenter, Postbox 260, DK-4000 Roskilde, Denmark

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Alternating current conduction in disordered solids is discussed, from a general point of view. As regards experiment, it is argued that the observed power-law behavior of the frequency-dependent conductivity, $\sigma(\omega)$, is probably not fundamental, that the Ngai relation between dc and ac conductivity activation energies follows from independent experimental facts, that the shape of the modulus peak has no fundamental significance, and that there are interesting mechanical analogies to the observed ac electrical behavior. As regards hopping models for ac conduction, it is shown that three commonly used arguments against the existence of a distribution of activation energies are all incorrect. Also, it is shown that $\sigma(\omega) \neq \sigma(0)$ only if there are correlations in the directions of different charge carrier jumps; in particular this result implies $\sigma(\omega) = \sigma(0)$ for all frequencies in the continuous time random walk (CTRW) model. In the final section a number of open problems are listed, and suggestions are made for future work.

1. Introduction

This paper discusses ac conduction in non-metallic disordered solids. A number of remarks are made, most of which are not new but are still not generally appreciated. The class of disordered solids with interesting ac behavior is very large, including amorphous semiconductors [1,2], ionic conductive glasses [3,4], conducting polymers [5,6], various defective or doped crystals [7–9], and many polycrystals [10,11].

Several different representations of ac data are used. One possibility is the complex frequency-dependent conductivity, $\sigma(\omega) = \sigma'(\omega) + i\sigma''(\omega)$. A common alternative is the complex electric modulus, $M(\omega) = M'(\omega) + iM''(\omega)$, defined [12] by

$$M(\omega) = \frac{i\omega}{\sigma(\omega)}. \quad (1)$$

Data may also be presented in terms of the complex impedance [13,14], or in terms of the

complex dielectric constant which is defined by

$$\epsilon_0 \epsilon(\omega) = \frac{\sigma(\omega) - \sigma(0)}{i\omega}. \quad (2)$$

Here, ϵ_0 is the vacuum permittivity. The negative imaginary part of $\epsilon(\omega)$, $\epsilon''(\omega)$, is referred to as the dielectric loss.

AC conduction in quite different disordered solids shows a number of common features, a surprising fact which is often overlooked. For each of the above listed classes of glassy solids one observes, almost without exception [15–19], that at high frequencies $\sigma'(\omega)$ follows a power-law with an exponent s in the range 0.7–1.0; s goes to 1 as the temperature goes to zero. Around the dielectric loss peak frequency, ω_m , there is a transition to a frequency-independent conductivity below ω_m . The Barton–Nakajima–Namikawa (BNN) relation [20–22] is satisfied:

$$\sigma(0) = p \Delta\epsilon \epsilon_0 \omega_m, \quad (3)$$

where $\Delta\epsilon = \epsilon(0) - \epsilon(\infty)$ and p is a numerical constant of order one. Finally, the time-temperature

superposition principle is usually obeyed, i.e., the fact that the shape of the $\sigma'(\omega)$ curve is temperature-independent when plotted in a log–log plot.

The BNN relation signals an important correlation between dc and ac properties. It applies to most disordered solids with a large charge carrier concentration, the solids which are of interest here. If the loss is not due to migrating charge carriers but is dipolar in origin, one does not expect the BNN relation to be obeyed; clearly in such solids any dc conduction would have nothing to do with the dielectric loss due to the dipoles.

The outline of the paper is the following. In section 2, some points relating to experiment are discussed. Section 3 is devoted to hopping models for ac conduction. Section 4 deals with a number of open problems and gives suggestions for future work. Finally, section 5 is the conclusion.

2. Remarks relating to experiment

2.1. The observed power-law frequency dependence of the conductivity is hardly fundamental

The commonly observed large-frequency power-law

$$\sigma'(\omega) \propto \omega^s \quad (4)$$

is deduced from $[\log \sigma'(\omega)]$ having a linear dependence on $\log \omega$. Since both frequency and conductivity usually vary several decades, it is reasonable to plot data in a log–log plot. However, log–log plots are dangerous; an old saying [23] warns: “Almost anything is a straight line in a log–log plot”. The term “anything” refers to any function $f(x)$ which changes several decades when x changes several decades. To illustrate this point, fig. 1 shows a log–log plot of $\sigma'(\omega)$ where $\sigma(\omega)$ is given by

$$\sigma(\omega) = \sigma(0) \frac{i\omega\tau}{\ln(1 + i\omega\tau)}. \quad (5)$$

This function gives a reasonably good fit to many data [19]. (There is, of course, always an additional purely imaginary contribution to the conductivity from the infinite frequency dielectric

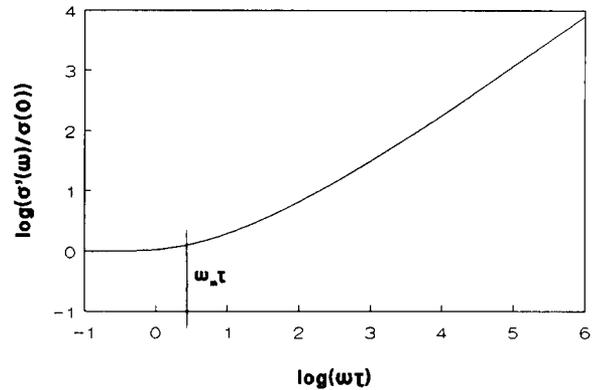


Fig. 1. Log–log plot of the real part of the function $\sigma(\omega)$ given by eq. (5). Although there is no power-law in this function, it follows closely a power-law at high frequencies. The function gives a good fit to many data [19]; this illustrates that one cannot conclude from experiment that a power-law frequency dependence of the ac conductivity of disordered solids is fundamental. The vertical line marks the dielectric loss peak frequency which is always found where the conductivity starts increasing.

constant.) At high frequencies, $\sigma'(\omega)$ follows closely a straight line in the log–log plot, although there is no power-law hidden in eq. (5). Most workers would report an exponent around 0.8 for data following fig. 1. There is no problem with this as long as one speaks only about *approximate* power-laws. However, there is no basis for concluding from $\sigma'(\omega)$ measurements that power-laws are *fundamental*, as is sometimes done [24–26]. If one wants to look into the existence of exact power-laws in data, a much more sensitive method is to study the inverse loss tangent, as shown recently by Niklasson [27].

2.2. The Ngai relation is a consequence of the BNN relation and the time-temperature superposition principle

The Ngai relation [28] correlates three quantities, the activation energy of the dielectric loss peak frequency, ΔE_m , the ac conductivity activation energy, ΔE_{ac} , and the exponent s of eq. (4), as follows:

$$\Delta E_{ac} = (1 - s)\Delta E_m. \quad (6)$$

The quantity ΔE_{ac} is defined as minus the slope of the tangent in the plot of $\log(\sigma'(\omega))$ versus T^{-1} . In general, this quantity, as well as the exponent s , depend on temperature. Equation (6) is confirmed by experiment [28] (ΔE_{ac} is sometimes derived from NMR experiments which, however, give the same activation energy as that of ac conduction [4,29]). As illustrated in fig. 1, the loss peak frequency, ω_m , is the characteristic frequency for the onset of ac conduction. Since the dielectric loss strength, $\Delta\epsilon$, is only weakly temperature-dependent, the essence of the BNN relation is an approximate proportionality between $\sigma(0)$ and ω_m . Remember that the time-temperature superposition principle is the fact that, at different temperatures, one observes in the log-log plot parallel displacements of the same $\sigma'(\omega)$ curve. Because of the proportionality between $\sigma(0)$ and ω_m , as the temperature changes, the $\sigma'(\omega)$ curve is displaced in a direction 45° to the x - and y -axis. Since activation energies are given as derivatives of the logarithm, it is now straightforward to show that eq. (6) is automatically obeyed whenever $\sigma'(\omega)$ follows an approximate power-law with exponent s .

2.3. The shape of the modulus peak has no fundamental significance

For all disordered solids the imaginary part of the electric modulus, $M''(\omega)$, has a peak at a frequency which is usually of the same order of magnitude as ω_m . The shape of the modulus peak is often attributed to a spectrum of relaxation times [12,30]. This spectrum, however, has no significance relative to the motion of the mobile charge carriers. This is because there is always, in parallel to the charge transport due to the mobile charge carriers, the current due to the infinitely fast dielectric displacement. The strength of the latter current is given by the infinite frequency dielectric constant, ϵ_∞ . If ϵ_∞ is changed, the shape of $M''(\omega)$ is affected [31]. This dependence is illustrated in fig. 2 where the loss modulus is plotted in a log-log plot for hypothetical solids with charge carrier contribution to the conductivity given by eq. (5) but with different values of ϵ_∞ .

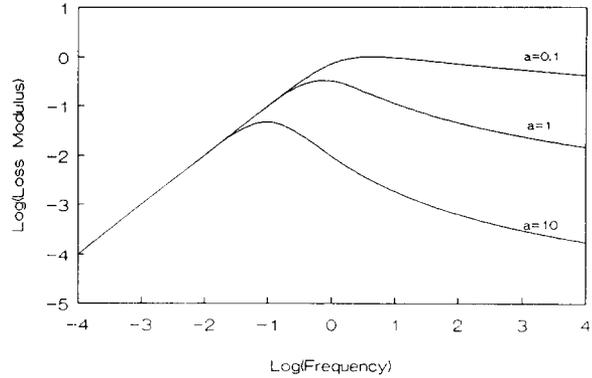


Fig. 2. Log-log plot of $M''(\omega)$ for hypothetical solids with the same hopping contribution to the conductivity but with different high frequency dielectric constant. The conductivity is given as a sum of eq. (5) and the purely imaginary contribution $i\omega\epsilon_\infty\epsilon_0$. The figure shows the dimensionless quantity $M''(\omega)\sigma(0)\tau$ as function of $\omega\tau$ for three different values of $a = \epsilon_0\epsilon_\infty/\sigma(0)\tau$.

2.4. There are close mechanical analogies to the observed ac behavior

(a) Many ionic conductive glasses have an internal friction loss peak at the dielectric loss peak frequency [4,32,33]. This mechanical loss must be due to ionic motion and thus mechanical and electrical properties of ionic glasses are strongly correlated.

(b) The frequency-dependent viscosity, $\eta(\omega)$, of a typical highly viscous liquid, e.g., a polymeric liquid, looks very much like $1/\sigma(\omega)$ for a typical disordered solid. Thus, at low frequencies $|\eta(\omega)|$ is constant whereas at higher frequencies $|\eta(\omega)|$ decreases like an approximate power-law [34]. Now, suppose a foreign microscopic particle is introduced into a viscous liquid. If the particle is described by hydrodynamics, its frequency-dependent mobility (velocity/force) varies as $1/\eta(\omega)$. Thus, the particle moves about in the liquid much as a charge carrier moves about in a disordered solid. Conductivity measurements on ions dissolved in viscous liquids, where the observed conductivity is indeed like that of a disordered solid, confirms this picture [35,36].

3. Remarks relating to hopping models

In hopping models, dc and ac conduction are both due to hopping charge carriers [37]. The solid disorder is usually incorporated by assuming randomly varying transition rates $\Gamma(s' \rightarrow s)$ for transitions from site s' to site s . If $P(s, t)$ is the probability for a particle to be at site s at time t , a hopping model is described by the master equation [37,38]

$$\frac{\partial P(s, t)}{\partial t} = -\gamma_s P(s, t) + \sum_{s'} \Gamma(s' \rightarrow s) P(s', t), \quad (7)$$

where

$$\gamma_s = \sum_{s'} \Gamma(s \rightarrow s'). \quad (8)$$

Equation (7) applies to the most commonly studied case of a system of non-interacting charge carriers. It refers to the zero external field situation. More generally, Γ depends on the external field, but the fluctuation–dissipation theorem allows one to calculate $\sigma(\omega)$ purely from a knowledge of the zero field jump frequencies. It can be shown that in hopping models $\sigma'(\omega)$ is always an increasing function of ω [39]. No exact analytical methods are available for evaluating $\sigma(\omega)$, but various approximate methods exist [37,38,40,41].

3.1. Three common arguments against hopping models are all incorrect

In most hopping models the variation in hopping rates is assumed to derive from a spread in activation free energies, $P(\Delta F)$. The following three arguments have traditionally been put forward against the existence of any $P(\Delta F)$.

(a) “Any distribution of activation energies implies the dc conductivity is non-Arrhenius.” This is not necessarily true; in some models $\sigma(0)$ is indeed Arrhenius with an activation energy which is simply the maximum barrier encountered on any ‘percolation’ path between the electrodes. In one dimension, exact results are available [42,43]. Here, $p(\Delta F) = \text{constant}$, or more generally $P(\Delta F)$ proportional to $\exp(-\Delta F/\Delta F_0)$, give an

exactly Arrhenius $\sigma(0)$ if a sharp cut-off at a maximum activation energy is assumed. Similar results are obtained from the approximate analytical methods available in three dimensions [37,38,40,41].

(b) “The BNN relation implies that ac conduction is due to processes with activation energy equal to that of $\sigma(0)$.” The BNN relation implies that the dielectric loss peak frequency has the same activation energy as $\sigma(0)$. However, this does not rule out the possibility of a distribution of activation energies being responsible for the frequency dispersion. Thus, in most hopping models ω_m corresponds to the *lowest* effective jump frequency and this quantity is determined by the *maximum* energy barrier, just as $\sigma(0)$ is itself [44].

(c) “The time–temperature superposition principle contradicts the existence of a distribution of energy barriers.” In some cases it can be clearly excluded that a distribution of activation energies is responsible for the observed frequency dispersion [45]. In general, however, this possibility cannot be ruled out. In particular, it is incorrect to claim, as is often done, that the existence of a distribution of activation energies necessarily implies a broadening of the distribution of relaxation times as the temperature is decreased, thereby violating the time–temperature superposition principle. First, for experimental reasons, the time–temperature superposition principle is usually checked only over a relatively narrow range of temperatures and frequencies; here any sufficiently broad distribution of activation energies will obey the time–temperature superposition principle rather accurately. Second, for the flat distribution of activation energies, $p(\Delta F) = \text{constant}$, the time–temperature superposition principle is obeyed exactly. In this case, the distribution of jump frequencies varies as Γ^{-1} at all temperatures. Thus, as long as one assumes a sufficiently broad distribution of activation energies, approaching the flat distribution, there is no contradiction with experiment.

Traditionally, points (a), (b) and (c) have been thought to imply at most a quite *narrow* $p(\Delta F)$, which obviously cannot account for the observed very *broad* loss peaks. This is why an early model like Stevels’ and Taylor’s random potential en-

ergy model from 1957 [46,47] was never considered a serious candidate for explaining experiments.

3.2. *The conductivity is frequency-dependent only if there are correlations between the directions of charge carrier jumps*

Thus, if each jump occurs in a random direction one has $\sigma(\omega) = \sigma(0)$ at all frequencies [38]. To prove this result, we first recall the fluctuation-dissipation theorem which expresses $\sigma(\omega)$ in terms of the zero-field auto-correlation function of the total current in volume V , $\mathbf{J}(t)$, in the following way [48]:

$$\sigma(\omega) = \frac{1}{3k_B T V} \int_0^\infty \langle \mathbf{J}(0) \mathbf{J}(t) \rangle e^{-i\omega t} dt. \quad (9)$$

Here k_B is the Boltzmann constant and T is the temperature. In hopping models the jumps are instantaneous and $\mathbf{J}(t)$ is a sum of delta functions. If the i th jump occurs at time, τ_i , and displaces a particle by $\Delta \mathbf{r}_i$, one has

$$\mathbf{J}(t) = q \sum_i \Delta \mathbf{r}_i \delta(t - \tau_i), \quad (10)$$

where q is the charge of the carrier. For $\langle \mathbf{J}(0) \mathbf{J}(t) \rangle$ to be non-zero at any $t > 0$, one must have $\langle \Delta \mathbf{r}_i \Delta \mathbf{r}_j \rangle \neq 0$ for at least one pair of $i < j$. However, whenever the direction of the latter jump, $\Delta \mathbf{r}_j$, is random, one has necessarily $\langle \Delta \mathbf{r}_i \Delta \mathbf{r}_j \rangle = 0$. Consequently, $\langle \mathbf{J}(0) \mathbf{J}(t) \rangle$ is proportional to $\delta(t)$ and the conductivity is frequency-independent according to eq. (9). This result has two important consequences.

(a) *Any random walk in a spatially homogeneous medium has $\sigma(\omega) = \sigma(0)$.* This is true even for non-Markovian random walks. An important example is the continuous time random walk model (CTRW) of Montroll and Weiss [49]. This model is characterized by the so-called waiting-time distribution function, $\psi(t)$, which is the probability for a particle to jump at time, t , given the particle last jumped at $t = 0$. In 1973, Scher and Lax erroneously calculated $\sigma(\omega)$ in terms of $\psi(t)$ [50]; the error was pointed out by Tunaley who proved by direct calculation that there is no frequency dispersion of the conductivity in the

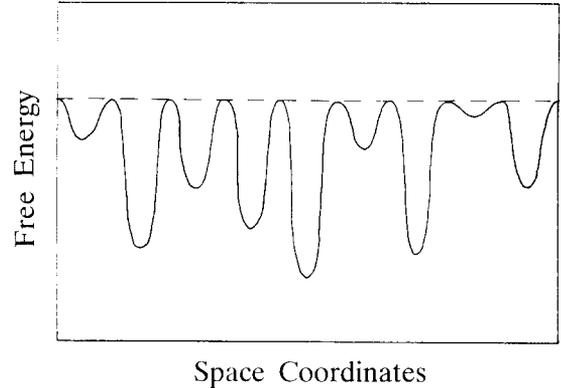


Fig. 3. Free energy surface of a hopping model which, because the direction of each charge carrier jump is random, has no frequency dependence of the conductivity. This example shows that a distribution of waiting times is not enough to ensure frequency dependence of the conductivity. Also, since the model has non-trivial transient behavior [55], the example shows that there is no correlation between ac conduction and transient behavior.

CTRW model [51,52]. While the CTRW model is itself of no use as a model for ac conduction, the formalism developed by Scher and Lax gives rise to a very useful approximation, usually referred to as the CTRW approximation or the Hartree approximation [40].

(b) *The existence of a distribution of relaxation times in a hopping model is not enough to ensure frequency dependence of the conductivity.* Consider hopping in a potential where all maxima are equal but the minima vary (fig. 3). Obviously, in this model there is a distribution of waiting times. However, the direction of each charge carrier jump is random, so $\sigma(\omega) = \sigma(0)$. This has also been shown by explicit calculation [53,54]. In passing, we note that the model of fig. 3 is a useful model for the transient behavior of photoexcited charge carriers in amorphous semiconductors [55]. Here, a brief laser pulse excites the electrons to random states at $t = 0$, and the current in an external field subsequently monitors the thermalization of the charge carriers. This example shows that, in general, transient currents cannot be calculated from $\sigma(\omega)$, as has been predicted from the study of specific models [56,57].

4. Open problems and suggestions for future work

4.1. *Are reported data always reliable and not due to contact effects?*

Electrode effects may cause serious problems for the interpretation of measurements. One might think it could easily be checked, by simply varying the sample size, whether or not the bulk response is measured. This is sometimes possible [58]; more often, however, it is quite difficult to prepare two samples with identical physical properties. The contacts are usually modeled as simple RC elements, implying the bulk response is measured at sufficiently high frequencies. It has never been proved that this procedure is correct. In fact, it has been suggested that contacts and interfaces play a dominant role in the whole range of frequencies measured [59]. While this is probably too drastic a statement, even a quite simple model of the electrode/sample interface predicts a non-trivial frequency dependence of the measured conductivity, varying like $\omega^{1/2}$ [32,60]. In conclusion, it is not obvious that all reported data are bulk, and more work is needed to clarify the role of contacts.

4.2. *Are dc and ac conduction always due to the same mechanism?*

The BNN relation shows that dc and ac conduction in disordered solids are strongly correlated. The simplest possibility is that dc and ac conduction are both due to the same mechanism, as is the case in hopping models. The existence of a loss peak supports this; loss peaks are very hard to explain otherwise. It should be noted that, when there is no dielectric loss peak, a BNN-like relation may still exist between $\sigma(0)$ and the characteristic frequency ω'_m defined by

$$\sigma'(\omega'_m) = 2\sigma(0). \quad (11)$$

(If a loss peak does exist, ω'_m is close to ω_m .) Suppose $\sigma'(\omega) = \sigma(0) + A\omega$. Then clearly ω'_m is proportional to $\sigma(0)$ although this does not reflect any relation between dc and ac conduction. Thus, the existence of a genuine loss peak is

necessary to ensure ac and dc conduction are due to the same mechanism. The existence of loss peaks should always be carefully checked in experiments.

4.3. *There are theoretical reasons to expect $\epsilon''(\omega)$ is proportional to $\omega^{1/2}$ on the low-frequency side of the dielectric loss peak*

In hopping models, one has always $\sigma(\omega) = \sigma(0) + C(i\omega)^{3/2}$ as $\omega \rightarrow 0$ [38,61], an example of the celebrated 'long time tails'. However, there seems to be no experimental evidence for this prediction. As one of the few general predictions in the field, it should be tested carefully on a number of disordered solids. Unfortunately electrode effects cause very serious problems for measuring accurately the low frequency side of the loss peak, and the prediction may be very difficult to verify.

4.4. *Does any solid exist which has $\sigma'(\omega) \ll \epsilon_0\omega$?*

A puzzling phenomenon is the fact that, apparently, any solid has a conductivity $\sigma'(\omega)$ which is at least of order $\epsilon_0\omega$ [16]. Thus, at 1 MHz the conductivity is never much less than 10^{-6} ($\Omega \text{ cm}$)⁻¹. This rule seems to apply without exception, even to single crystal insulators. It could be a spurious effect due to contact effects [59], or due to experimental problems in distinguishing properly between $\sigma'(\omega)$ and $\sigma''(\omega)$ ($\sigma''(\omega)$ has always a sizable contribution from the infinite frequency dielectric constant). If the effect is real, an explanation is very much needed. Is it possible that even the most 'perfect' single crystal contains enough defects to account for this observation?

4.5. *What kind of measurements could supplement the measurement of $\sigma(\omega)$?*

The ac conductivity is the $k = 0$ component of the more general quantity $\sigma(\mathbf{k}, \omega)$ (which, by the fluctuation-dissipation theorem, is related to equilibrium fluctuations of the k th Fourier component of $\mathbf{J}(\mathbf{r}, \omega)$). It would be interesting to have measurements of $\sigma(\mathbf{k}, \omega)$. For electronic

systems, it is not obvious how to do these measurements but for ionic conductors neutron scattering can be applied, at least in principle. Other important measurements to supplement $\sigma(\omega)$ are transient current experiments (available only for electronic conductors) [62], excess current noise measurements [63], and large field experiments [17,32,64,65].

4.6. Is the observed ac behavior due to microscopic or macroscopic inhomogeneities?

The mathematical description of, and predictions for, inhomogeneous conductors are quite similar to that of hopping models [11,14]. Therefore, ac measurements alone do not reveal whether macroscopic or microscopic inhomogeneities are responsible for the observed frequency dispersion. It is not unlikely that, in some amorphous systems, there are inhomogeneities with dimensions of several hundred ångströms. One way to distinguish between macroscopic and microscopic inhomogeneities is to measure the large field response; for macroscopic inhomogeneities, one expects non-linearities to set in at much lower fields than for microscopic inhomogeneities [17,32,66,67]. Unfortunately, electrode problems are a serious obstacle for reliable measurements of the non-linear conductivity.

4.7. There are two important open problems relating to hopping models:

4.7.1. How accurate are the presently available approximate analytical solutions of hopping models?

Perhaps the simplest hopping model is the random free energy barrier model which, when solved in the CTRW approximation, yields eq. (5) [19]. Numerical solutions of this model should be undertaken to assess the validity of eq. (5). Preliminary work shows that, in one dimension, eq. (5) works very well [68]. In general, the question 4.7.1. remains unanswered.

4.7.2. What is the cause of the quasi-universality among different models?

As noticed by Summerfield in 1985 [69], different models solved in the extended pair approxi-

mation (EPA) yield almost identical predictions for $\sigma(\omega)$, apart from an overall scaling of σ and ω . In fact, 'quasi-universality' applies not only to EPA models, but to most models studied so far. The cause of quasi-universality is not clear. The agreement between different hopping models is generally much better than the agreement between theory and experiment, where quasi-universality does not really apply. This indicates that the hopping models described by eq. (7) are perhaps too simple. It seems likely that interactions between the charge carriers have to be taken into account to arrive at a realistic model [70,71]. (Contrary to what is sometimes claimed, eq. (7) cannot describe interacting particles [72].)

The most important question relating to hopping models, of course, is as follows.

4.8. Are hopping models the correct framework for describing experiment?

Hopping models are simple and give reasonably good fits to experiments. However, it is possible that other types of models are more appropriate. Thus, the ω^s behavior of the dielectric loss in insulating dielectrics cannot be explained by any hopping model that allows a dc conduction, and the correct model for this phenomenon could be applicable also to describe loss in conducting dielectrics [24,25].

5. Conclusion

There are a number of important unsolved problems in the field of ac conduction. Because of this, measurements of $\sigma(\omega)$ do not yet provide unambiguous insight into the conduction process. More work, for instance along the lines of section 4, is needed before this goal is reached. In this sense, ac conduction is still a field in its infancy.

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