

TEKST NR 224

1992

UNIVERSAL AC CONDUCTIVITY OF NON-METALLIC SOLIDS AT LOW
TEMPERATURES

By: Jeppe C. Dyre

TEKSTER fra

IMFUFA

ROSKILDE UNIVERSITETSCENTER
INSTITUT FOR STUDIET AF MATEMATIK OG FYSIK SAMT DERES
FUNKTIONER I UNDERVISNING, FORSKNING OG ANVENDELSER

IMFUFA. Roskilde Universitetscenter, Postboks 260, 4000 Roskilde
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IMFUFA tekst nr. 224/92 16 pages

ISSN 0106-6242

ABSTRACT

It is shown that, in the low temperature limit, the effective medium approximation predicts a universal frequency-dependence of the conductivity of non-metallic disordered solids. The calculation is based on a macroscopic approach to AC conduction and is valid in more than one dimension. The universality prediction is confirmed by simulations in two dimensions.

For many years AC conduction has been studied in disordered solids like amorphous semiconductors, glasses, polymers, nonstoichiometric solids, or metal-cluster compounds [1-8]. All disordered solids show similar AC behavior, whether the conduction is electronic, polaronic, or ionic. The frequency-dependent conductivity follows an approximate power-law with an exponent between 0.7 and 1.0. At lower frequencies there is a gradual transition to constant conductivity. The standard models for this are hopping models which deal with the random walk of non-interacting charge carriers in a random environment [9-13]. While hopping models are rather successful, the importance of Coulomb interactions has recently come into focus [14,15]. Maass and coworkers have shown that the Coulomb repulsion may have significant effect on the frequency-dependence of certain hopping models [15]. Unfortunately, hopping models with interactions are not amenable to simple analytic treatment. One way to include the effect of Coulomb interactions between charge carriers is to, instead of using hopping models, adopt a macroscopic point of view [16-19]. This is done here where conduction in inhomogeneous media is discussed by exploring Maxwell's equations.

Consider a solid with spatially varying thermally activated conductivity $g(\mathbf{r}) = g_0 e^{-\beta E(\mathbf{r})}$. Here β is the inverse temperature and the activation energy $E(\mathbf{r})$ is assumed to vary randomly in space with a finite correlation length. If ϵ denotes the dielectric constant and ω the angular frequency, the continuity equation and Gauss' law imply

for the electrostatic potential ϕ

$$\nabla \cdot (i\omega\epsilon + g) \nabla \phi = 0. \quad (1)$$

This equation is discretized [20,21] by regarding the potential ϕ as defined on the points of a simple cubic lattice and the quantity $i\omega\epsilon + g$ as defined on nearest neighbor links. In this way Eq. (1) becomes the Kirchhoff current conservation law for a lattice where each link is a resistor in parallel with a capacitor. If a is the lattice constant and D the dimension, the correct continuum limit is ensured if each link admittance y is given by

$$y = a^{D-2} (i\omega\epsilon + g) . \quad (2)$$

The circuit is not to be interpreted literally as a physical model of the solid because the real physical currents run through the resistors only. However, the circuit is useful for calculating the macroscopic frequency-dependent conductivity $\sigma(\omega)$, i. e., the ratio between average current and average electrical field. If L is the linear circuit dimension and $G(\omega)$ is the admittance between opposing faces, it is straightforward to show that, whenever ϵ is space-independent, $\sigma(\omega)$ is given [22] by

$$\sigma(\omega) = \frac{G(\omega)}{L^{D-2}} - i\omega\epsilon . \quad (3)$$

If the discretization length a is chosen to be the correlation length for $E(x)$ and correlations beyond a are ignored [20,23], the effective medium approximation (EMA) may be applied to calculate G [16,20]. The EMA equation for the effective link admittance, y_m , is $\langle (y - y_m) / [y + (D-1)y_m] \rangle_y = 0$

. Since $G = N^{D-2} y_m$ where $N = L/a$, the EMA equation and Eqs. (2) and (3) imply (where $s = i\omega\epsilon$ is the Laplace frequency)

$$\frac{1}{D(\sigma + s)} = \left\langle \frac{1}{g(E) + (D-1)\sigma + Ds} \right\rangle_E . \quad (4)$$

This equation has a simple solution in the limit $\beta \rightarrow \infty$. The root $E = E_g(s)$ of $g(E) = (D-1)\sigma + Ds$ is given by

$$E_g(s) = -\frac{1}{\beta} \ln \left(\frac{(D-1)\sigma + Ds}{g_0} \right) . \quad (5)$$

If $p(E)$ is the activation energy probability distribution, Eq. (4) at low temperatures becomes

$$\frac{1}{D(\sigma + s)} = \frac{1}{(D-1)\sigma + Ds} \int_{E_g(s)}^{\infty} p(E) dE , \quad (6)$$

or

$$\frac{D-1}{D} + \frac{s}{D(\sigma + s)} = \int_{E_g(s)}^{\infty} p(E) dE . \quad (7)$$

For large β subtracting the $s=0$ case of Eq. (7) from Eq. (7) itself leads to

$$\frac{s}{D(\sigma+s)} = \int_{E_g(s)}^{E_g(0)} p(E) dE = p(E_g(0)) \frac{1}{\beta} \ln \left(\frac{\sigma}{\sigma(0)} + \frac{D}{D-1} \frac{s}{\sigma(0)} \right) \quad (8)$$

Introducing the dimensionless variables

$$\tilde{\sigma} = \frac{\sigma}{\sigma(0)} \quad , \quad \tilde{s} = \frac{\beta}{D p(E_g(0)) \sigma(0)} s \quad , \quad (9)$$

Eq. (8) for $\beta \rightarrow \infty$ reduces to

$$\tilde{\sigma} \ln(\tilde{\sigma}) = \tilde{s} \quad . \quad (10)$$

Equation (10) was derived by Fishchuk for the uniform energy barrier distribution with cut-off's where the average in Eq. (4) can be calculated explicitly [24]. Here it has been shown that, in the low temperature limit, the EMA predicts a universal frequency-dependence of the conductivity (in any dimension $D > 1$). There is, however, some doubt whether the EMA is reliable for systems with extremely broad distributions of admittances [25,26]. Therefore, computer simulations were carried out to test Eq. (10). At low temperatures large lattices are needed to obtain reasonable statistics, and the simulations are quite demanding. Only the two-dimensional case was studied where the highly efficient Frank-Lobb algorithm is available [27]. For simplicity the simulations were carried out for real \tilde{s} . Several different activation energy distri-

butions were used. The results are shown in Fig. 1. They confirm the EMA prediction of universality as the temperature is lowered. The universality represents a new type of regularity, appearing gradually as the "relaxation time distribution" becomes extremely broad. The universality is not a consequence of a diverging correlation length, as for a second order phase transition, and there are no critical exponents.

The EMA equation (10) was first derived by Bryksin for a model of non-interacting electrons tunneling between positionally disordered sites [28]; it has also been shown to apply for a hopping model with a box type distribution of energy barriers [5]. Hopping models are neither physically nor mathematically equivalent to the macroscopic approach taken here. But both types of models lead to large sparse matrix equations expressing local current conservation. It is likely that, in the limit of severe disorder, the EMA for any problem of this type leads to Eq. (10) for the frequency-dependent conductivity (or diffusion constant).

An important and well established experimental fact is the Barton-Nakajima-Namikawa (BNN) relation [29-31], i. e. , the rule that the characteristic frequency for onset of AC conduction has the same activation energy as $\sigma(0)$ [32]. This follows directly from Eqs. (9) and (10) (a reduced frequency definition similar to Eq. (9) was used for hopping models by Scher and Lax [33] and by Summerfield [34]). It is easy to understand qualitatively why the BNN relation is valid here. In the DC limit the current follows the "critical" percolation paths giving the easiest ways between the electrodes [35]. As

the frequency increases there is little effect until, for $S \sim S_c$, S is of order the lowest admittance Y_{\min} met on a critical path. On the other hand, the DC conductivity is also determined by Y_{\min} [35,36], and thus one expects $\sigma(0) \sim S_c$ which is the essence of the BNN relation.

In three dimensions the EMA has the percolation threshold somewhat wrong [20], so the predicted DC conductivity activation energy is also wrong. However, Eq. (10) may still be valid in three dimensions at low temperatures. Summerfield has conjectured a "quasi-universality" for the frequency-dependence of the conductivity [34]. This idea fits nicely into the present work that predicts true universality only in the zero temperature limit. Comparing to experiments, it has been shown elsewhere [5] that all qualitative features of experiment follow the equation

$$\tilde{\sigma} = \frac{\tilde{S}}{\ln(1+\tilde{S})} \quad (11)$$

Equation (11), which clearly is an approximate solution of Eq. (10), represents the admittance along a critical path [22]. Both equations predict an approximate power-law frequency-dependence of the real part of the conductivity where the exponent at the real frequency $\tilde{\omega} = -i\tilde{S}$ is equal to $1 - 2/\ln(\tilde{\omega})$ [5,28]. A few decades above the onset of AC conduction, the exponent is predicted to be close to 0.8, in agreement with most experiments. Thus, there are certainly no experimental reasons to reject Eq. (10) as a low temperature limiting

universal frequency-dependence of the conductivity in three dimensions.

Some time ago Pollak and Pike suggested that details of the conduction mechanism should be contained in deviations from linear frequency-dependence of the conductivity [37]. While Eq. (10) predicts proportionality $\tilde{\sigma} \propto \tilde{\omega}$ for $\tilde{\omega} \rightarrow \infty$, there is a non-trivial frequency-dependence in a very large frequency range. If the predicted universality is indeed valid also in three dimensions, there is little information in a conductivity that follows Eq. (10). It seems therefore that experiments could naturally be interpreted in terms of deviations from Eq. (10), which represents the low temperature fixpoint.

In conclusion, it has been shown that the EMA predicts a universal frequency-dependence of the conductivity for disordered non-metals at low temperatures, and simulations in two dimensions have confirmed the prediction. Since hopping models often also follow Eq. (10), one cannot from AC measurements alone distinguish these two approaches to the modelling of AC conduction in disordered solids.

ACKNOWLEDGEMENTS

The author wishes to thank N. B. Olsen and P. V. Christensen for useful discussions and I. H. Petersen for technical assistance. This work was supported by the Danish Natural Science Research Council.

FIGURE CAPTION

Log-log plot (base 10) of the dimensionless conductivity $\tilde{\sigma}$ as function of the real dimensionless Laplace frequency \tilde{S} (both quantities defined in Eq. (9)). The full curve is the EMA prediction for the low temperature limit of $\tilde{\sigma}(\tilde{S})$ (Eq. (10)), while the symbols represent simulations in two dimensions for different activation energy distributions at different temperatures. Each point represents the average of 20 simulations of a 100x100 lattice. The total admittance was determined by the Frank-Lobb algorithm [27] and $\tilde{\sigma}$ subsequently found from Eq. (3). Beta is the inverse dimensionless temperature. Results are shown for the following activation energy distributions:

a)
$$p(E) = \frac{1}{\sqrt{2\pi}} e^{-E^2/2} \quad (-\infty < E < \infty) \quad (\blacktriangle).$$

b)
$$p(E) = \frac{1}{2} \quad (-1 < E < 1) \quad (\bullet).$$

c)
$$p(E) = \frac{2}{\pi} \frac{1}{1+E^2} \quad (0 < E < \infty) \quad (\blacktriangle).$$

d)
$$p(E) = e^{-E} \quad (0 < E < \infty) \quad (\circ).$$

e)
$$p(E) = 2E \quad (0 < E < 1) \quad (\dagger).$$

In each case the distribution could be thought of realistically as centered around an energy E_0 ; this would give an extra

factor $e^{-\beta E_0}$ to both conductivity and frequency without changing $\tilde{\sigma}$ or \tilde{S} . The quantity $E_g(0)$ in Eq. (9) is the DC conductivity activation energy (Eq. (5)) which is easily determined from the fact that the percolation threshold is $1/2$ in two dimensions [20,22,35,36].

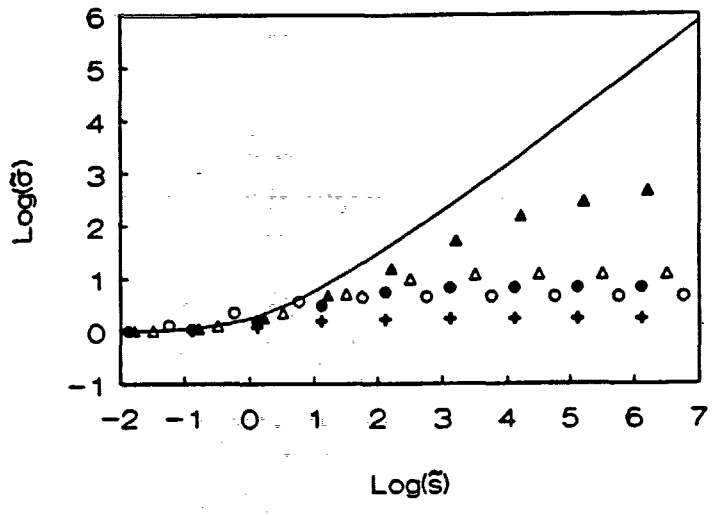
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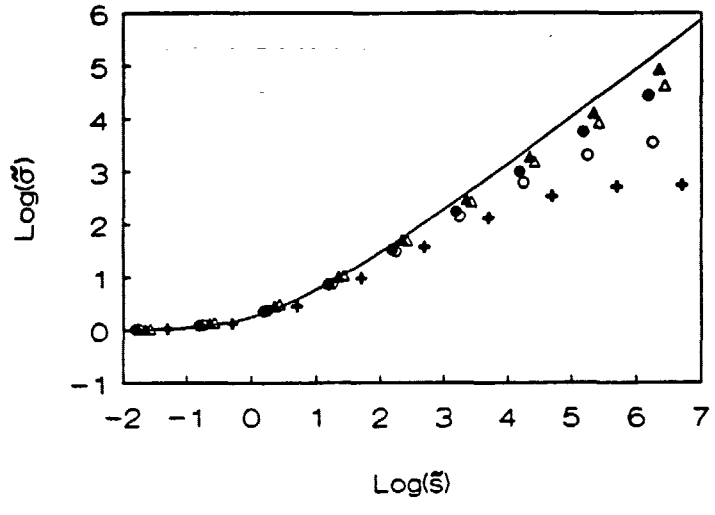
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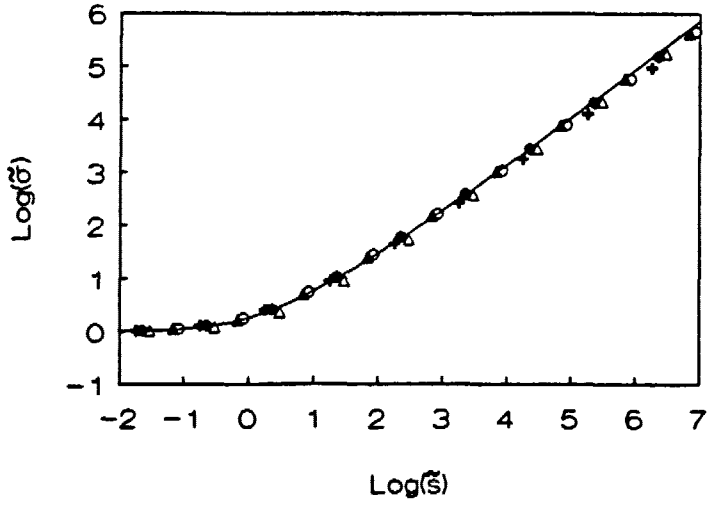
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