

## Studies of AC hopping conduction at low temperatures

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*Publication date:*  
1993

*Document Version*  
Publisher's PDF, also known as Version of record

*Citation for published version (APA):*  
Dyre, J. (1993). *Studies of AC hopping conduction at low temperatures*. Roskilde Universitet. Tekster fra IMFUFA No. 259 <http://milne.ruc.dk/lmfufaTekster/>

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**TEKST NR 259**

**1993**

**STUDIES OF AC  
HOPPING CON-  
DUCTION 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

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STUDIES OF AC HOPPING CONDUCTION AT LOW TEMPERATURES

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IMFUFA tekst nr. 259/93

61 pages

ISSN 0106-6242

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Abstract.

A new method is presented that makes computer simulations of hopping conduction in symmetric hopping models with thermally activated jump rates possible at arbitrarily low temperatures. The method utilizes the AC Miller-Abrahams electrical equivalent circuit which is systematically reduced till one ends up with an admittance matrix referring to the voltage generators; from this matrix the conductivity is easily calculated. The results of simulations of hopping in two dimensions are presented and compared to the predictions of the effective medium approximation (EMA). It is shown that the frequency dependent conductivity as the temperature goes to zero in the EMA becomes **universal**, i. e. , independent of the activation energy probability distribution. The computer simulations confirm the existence of universality, although there is not a quantitative agreement between the simulations and the EMA universality prediction.

## 1. INTRODUCTION

AC conduction in disordered solids like amorphous semiconductors, ionic conductive glasses, polymers, or metal-cluster compounds show a number of common features [1-8]. Above a characteristic frequency,  $\omega_m$ , the conductivity becomes strongly frequency dependent, varying as an approximate power-law with exponent between 0.7 and 1.0. The AC conductivity is always less temperature dependent than the DC conductivity, and at low temperatures the AC conductivity becomes almost temperature independent. A final ubiquitous observation is the BNN relation [9-13] which expresses the fact that the characteristic frequency  $\omega_m$  is proportional to the DC conductivity  $\sigma(0)$ . In particular, these two quantities always have the same activation energy.

The most thoroughly studied models for AC conduction in disordered solids are the so-called hopping models [14-16]. A hopping model describes jumps of charge carriers in a classical stochastic framework. The disorder is usually mimicked by assuming that the transition rates vary randomly according to some probability distribution. Only linearized hopping models are amenable to analytic treatment (which is still approximate). Linearized hopping models, henceforth just referred to as hopping models, describe the motion of non-interacting charge carriers. Thus, self-exclusion as well as Coulomb interactions are ignored.

Recently, the role of Coulomb interactions has come into focus [17,18]. To include the effects of Coulomb repulsion, a "macroscopic" model [19-20] was studied by the present author,

following previous work by Springett, Webman et al, Sinkkonen and Fishchuk [21-24]. When Maxwell's equations for an inhomogeneous semiconductor are discretized, one arrives at an electrical equivalent circuit with nodes placed on a cubic lattice and links between neighboring nodes consisting of a resistor and a capacitor in parallel [20,22,24]. The capacitor currents are Maxwell's displacement currents while the resistor currents are the free charge currents [20]. Computer simulations of this model in 2-D and in 3-D have shown [20] that the effective medium approximation (EMA) [25] works very well, even at low temperatures where the disorder is large and the EMA was not previously believed to be reliable. In particular, the simulations confirmed the EMA prediction [19,20] of a universal frequency dependence for the conductivity at low temperatures, independent of the probability distribution for the activation energy of the local conductivity. If one defines  $\tilde{\sigma} = \sigma(\omega) / \sigma(0)$  and  $\tilde{S} = i\tilde{\omega}$  where  $\tilde{\omega}$  is a suitable dimensionless frequency, the EMA equation for the universal frequency dependent conductivity in the macroscopic model [19,20] is

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

Ref. 19 gave the first general derivation of Eq. (1) and presented the first computer simulations confirming it. Equation (1), however, appeared in the literature already in 1980 in a paper by Bryksin studying a hopping model with electrons tunneling between positionally disordered sites [26]. The recent results for the macroscopic model therefore raises a number of questions: Is the EMA for hopping models as reliable as it is

for the macroscopic model? In particular, as  $T \rightarrow 0$ , does the EMA predict Eq. (1) as the universal low temperature frequency dependence of the conductivity even for hopping models with thermally activated jump rates? If this is the case: Is the universality confirmed by computer simulations? To answer these questions, new numerical methods must be developed since neither the standard Monte Carlo method nor, e. g., the Gauss-Seidel relaxation method allows one to go to low temperatures where the jump rates vary over several decades (often more than 50 decades!).

The present paper introduces a new method for computer simulation of hopping models. The method allows one to go to arbitrarily low temperatures without any computational "slowing down". It utilizes the Miller-Abrahams equivalent circuit which is systematically reduced by eliminating nodes according to a transformation well-known from electrical engineering. Before this new method is presented in Sec. 3, Section 2 briefly reviews the basic equations of hopping models and the EMA equation for the frequency dependent conductivity in hopping models. Only symmetric hopping models are dealt with in the present paper, i. e. , models where the rate for jumps between two given sites depends on a common energy barrier. Section 4 reports the results of computer simulations in 2-D and compares the simulations to the EMA predictions. In Sec. 5 the low temperature limit of the EMA is studied. It is shown that Eq. (1) even for hopping models is predicted to be the universal low temperature conductivity. Then Eq. (1) is compared to the results of computer simulations. Finally, Sec. 6 contains a

discussion.

## 2. SYMMETRIC LINEAR HOPPING MODELS

This section briefly reviews symmetric (linear) hopping models and their approximate solution in the EMA. Since several reviews have been written on this subject [14-16,27-30], only the most necessary background is given here.

For simplicity we consider first hopping in one dimension in thermal equilibrium, i. e., with no external electric field. A model solid is regarded in which the charge carriers can be only at regularly spaced discrete sites. Let  $N_i$  denote the number of charge carriers at site  $i$ . This number fluctuates in time because some particles leave site  $i$  and some arrive from the neighboring sites  $i-1$  and  $i+1$  (only nearest-neighbor jumps are allowed). If  $\Gamma(i \rightarrow j)$  denotes the probability per unit time of a jump from site  $i$  to site  $j=i\pm 1$ , the basic equation for the  $N_i$ 's is

$$\frac{dN_i}{dt} = - [\Gamma(i \rightarrow i+1) + \Gamma(i \rightarrow i-1)] N_i + \Gamma(i-1 \rightarrow i) N_{i-1} + \Gamma(i+1 \rightarrow i) N_{i+1} . \quad (2)$$

This equation is a simple example of a master equation [31,32].

In symmetric hopping models one has symmetric equilibrium jump probabilities,

$$\Gamma(i \rightarrow j) = \Gamma(j \rightarrow i) \equiv \Gamma_o(i, j) \quad [j=i\pm 1] \quad . \quad (3)$$

Introducing the probability of finding a particle at site  $i$ ,  $P_i = N_i/N$  where  $N$  is the total number of particles, Eq. (2)

becomes when Eq. (3) is taken into account

$$\frac{dP_i}{dt} = \Gamma_o(i, i-1) [P_{i-1} - P_i] - \Gamma_o(i, i+1) [P_i - P_{i+1}] \quad . \quad (4)$$

The stationary solution of Eq. (4) corresponds to all sites being equally probable. Any initial non-homogeneous distribution of charge carriers will eventually equilibrate through "diffusion" of particles away from densely populated sites.

Equation (4) is linear. This is the mathematical expression of the fact that the equation deals with non-interacting particles. The particles cannot feel each other: neither is Coulomb repulsion taken into account nor is "self-exclusion" [i. e., that there is room for only one particle at each site]. In some papers dealing with hopping models, more general non-linear hopping models are formulated taking these effects into account. Equation (4) is then arrived at by linearizing the general hopping equation. This linearization, however, is not exact, but involves uncontrolled approximations [33].

For systems of tunneling electrons the transition rates depend exponentially on the tunneling distance (as well as on the energy difference). In the present paper we are concerned with the "classical" case when the jump rates are thermally activated, i. e., where one has



$$\Gamma_o(i, j) = \Gamma_o e^{-\beta E_{i,j}} \quad (j=i\pm 1) \quad . \quad (5)$$

Here  $\beta=1/k_B T$ , while  $E_{i,j}$  is the so-called activation energy - the barrier to be overcome. The prefactor  $\Gamma_o$  is the "attack frequency" which is usually of order  $10^{12}$  Hz (a typical phonon frequency). Equation (5) is relevant for ionic conduction and for certain cases of polaronic conduction. Figure 1 shows an example of the potential for a symmetric hopping model in 1-D with thermally activated jumps. In hopping models one usually assumes that the jump rates vary randomly, i. e., are not subject to any spatial correlations. The model is then completely characterized by the activation energy probability distribution,  $p(E)$ .

Before considering the effect of an electric field, we briefly discuss the boundary conditions. The bulk behavior of the model is calculated by letting the volume go to infinity. In any computer simulation only a finite sample is present and one has to specify the boundary conditions. The case of periodic boundary conditions is characterized by requiring  $P_0=P_N$  if the sites are numbered from  $i=0$  to  $i=N$ . An alternative to this is the case of blocking electrodes, i. e., to modify Eq. (4) at the endpoints so that no particles may pass beyond these. A third possible boundary condition is the case of perfectly conducting electrodes. This is arrived at by imposing a fixed probability at the endpoints:

$$P_0 = P_N = \text{Const.} \quad (6)$$

Equation (6) corresponds to having electrodes that allow free passage for the charge carriers; the electrodes are particle reservoirs, each having a fixed chemical potential. This boundary condition is used below in the computer simulations.

In an external electric field the symmetry of the jump rates is broken, since it becomes more favorable to jump in the direction of the field than opposite to it (if the particle charge  $q$  is positive). If the barrier top is placed symmetrically between the two particle sites and  $a$  denotes the distance between neighbouring sites, the jump rates are modified [14] according to (note that  $E$  is now the electric field and not an energy barrier)

$$\begin{aligned} \Gamma(i \rightarrow i+1, E) &= \Gamma_0(i, i+1) e^{\beta qaE/2} \\ \Gamma(i+1 \rightarrow i, E) &= \Gamma_0(i, i+1) e^{-\beta qaE/2} \end{aligned} \quad (7)$$

In the linear limit which defines the ordinary field-independent conductivity, Eq. (7) is expanded to first order in the electric field:

$$\begin{aligned} \Gamma(i \rightarrow i+1, E) &= \Gamma_0(i, i+1) \left[ 1 + E \frac{\beta qa}{2} \right] \\ \Gamma(i+1 \rightarrow i, E) &= \Gamma_0(i, i+1) \left[ 1 - E \frac{\beta qa}{2} \right] \end{aligned} \quad (8)$$

Consequently, Eq. (2) is modified and becomes

$$\begin{aligned} \frac{dP_i}{dt} = & \Gamma_o(i-1, i) [P_{i-1} - P_i] - \Gamma_o(i, i+1) [P_i - P_{i+1}] \\ & + E \frac{\beta qa}{2} [\Gamma_o(i-1, i) (P_{i-1} + P_i) - \Gamma_o(i, i+1) (P_i + P_{i+1})] \end{aligned} \quad (9)$$

Note that the electric field may depend on time in an arbitrary way in this equation.

The [complex] frequency dependent conductivity,  $\sigma(\omega)$ , is defined as the ratio between the spatially averaged current density and the electric field in a steady state, where the electric field has a periodic time variation given by the factor  $e^{i\omega t}$ . The Kubo formula [34] expresses the frequency dependent conductivity in terms of the current-current time autocorrelation function. For hopping models it is convenient to rewrite the Kubo formula by performing two partial integrations. This leads [35] to the following expression

$$\sigma(\omega) = - \frac{nq^2\omega^2}{2k_B T} \int_0^\infty \langle \Delta x^2(t) \rangle_0 e^{-i\omega t} dt \quad (10)$$

Here,  $n$  is the average particle density,  $\langle \Delta x^2(t) \rangle_0$  denotes the equilibrium mean square displacement of a particle in time  $t$ , and a convergence factor  $\lim_{\epsilon \rightarrow 0} e^{-\epsilon t}$  is implicitly understood in the integral.

For a homogeneous system (i. e., with all jump rates equal) the mean square displacement is determined from the diffusion constant  $D$  via Einstein's equation  $\langle \Delta x^2(t) \rangle_0 = 2Dt$ , and as is easy to see Eq. (10) gives the frequency independent conductivity  $\sigma = nq^2\mu$

where  $\mu$  is the mobility which is given by the Nernst-Einstein relation  $\mu = \frac{D}{k_B T}$ . In the time  $t$  a particle in a homogeneous system with equilibrium neighbor jump rate  $\Gamma$  performs on the average  $N=2\Gamma t$  jumps. Therefore one has  $\langle \Delta x^2(t) \rangle_0 = Na^2 = 2\Gamma a^2 t$ , which implies  $D = \Gamma a^2$ . Combining these equations one finds for the frequency independent conductivity of a homogeneous system

$$\sigma = \frac{nq^2 a^2}{k_B T} \Gamma \quad (11)$$

For any inhomogeneous system the high-frequency limit of the conductivity is given by a similar expression. It can be shown [36] that  $\sigma(\infty)$  is given by the average jump frequency:

$$\sigma(\infty) = \frac{nq^2 a^2}{k_B T} \langle \Gamma \rangle \quad (12)$$

It is convenient to redefine the conductivity and absorb the trivial "prefactor" so that the conductivity is given in terms of an equivalent jump rate. In this "rationalized" unit system, which will be used henceforth, Eq. (12) simply becomes  $\sigma(\infty) = \langle \Gamma \rangle$ .

For one-dimensional hopping models it is also possible to calculate the DC conductivity exactly [36]. To summarize, in 1-D the high and low frequency limits are given by

$$\begin{aligned} \sigma_{1-D}(0) &= \langle \Gamma^{-1} \rangle^{-1} \\ \sigma_{1-D}(\infty) &= \langle \Gamma \rangle \end{aligned} \quad (13)$$

The inequality  $1 \leq \langle \Gamma \rangle \langle \Gamma^{-1} \rangle$  may be derived from the Cauchy-Schwartz inequality. It implies that one always has  $\sigma_{1-D}(0) \leq \sigma_{1-D}(\infty)$ . In fact, it can be shown [37] that in any hopping model in any dimension the real part of the conductivity is an increasing function of frequency.

Hopping models in D dimensions for  $D > 1$  are straightforward generalizations of the one-dimensional case. If  $P_{\mathbf{s}}$  denotes the probability of finding a particle at site  $\mathbf{s}$ , Eq. (4) is replaced by [for symmetric hopping models]

$$\frac{d}{dt} P_{\mathbf{s}}(t) = \sum_{\mathbf{s}'} \Gamma_0(\mathbf{s}, \mathbf{s}') [P_{\mathbf{s}'} - P_{\mathbf{s}}] \quad (14)$$

In the present paper the sites  $\mathbf{s}$  are assumed to lie on a simple cubic lattice and the sum is restricted to nearest neighbors. The Kubo formula Eq. (10) is also valid for  $D > 1$  where the mean-square displacement  $\langle \Delta x^2(t) \rangle_0$  is in any [fixed] direction. The three above mentioned boundary conditions also may be applied in several dimensions. In an external field the concepts of blocking or perfectly conducting electrodes makes sense only for the sample faces perpendicular to the field; it is natural to apply periodic boundary conditions to all remaining faces.

Even for  $D > 1$  is it convenient to use the rationalized unit system representing the conductivity in terms of an equivalent jump frequency. In this unit system the high frequency limit of the conductivity is given [28] by

$$\sigma(\infty) = \langle \Gamma \rangle . \quad (15)$$

For the DC conductivity, on the other hand, there is no general analytical expression similar to Eq. (13) in one dimension. However, the temperature dependence of  $\sigma(0)$  is known; it is given [38-40] by (for  $D > 1$ )

$$\sigma(0) \propto e^{-\beta E_c} , \quad (16)$$

where the so-called percolation energy  $E_c$  in terms of the activation energy probability distribution  $p(E)$  and the link percolation threshold  $p_c$  is given by

$$\int_{-\infty}^{E_c} p(E) dE = p_c . \quad (17)$$

For  $D=2$  one has  $p_c=1/2$  exactly while for  $D=3$  simulations have shown that  $p_c=0.2488$  [41].

The calculation of the frequency-dependent conductivity in hopping models is a complicated problem and suitable approximations have to be made. The standard approximation for disordered systems is the coherent potential approximation (CPA) [42,43]. The CPA is a mean-field approximation that gives an estimate of the relevant Green's function with a number of attractive analyticity properties. For hopping models the CPA is known as the effective medium approximation (EMA) because it is derived by considering one "link" (i. e. , jump frequency) of the lattice as embedded in an "effective medium" mimicing the average surroundings. Several papers [26,44-46] discuss the derivation of the EMA equation and here the result is just quoted. If the complex frequency,  $S=i\omega$ , is introduced - referred to below as

the "Laplace frequency" - the conductivity  $\sigma = \sigma(S)$  in the rationalized unit system introduced above is given [15] as the solution of the equation

$$\left\langle \frac{\Gamma - \sigma}{D\sigma + [1 - s\tilde{G}](\Gamma - \sigma)} \right\rangle_{\Gamma} = 0 \quad . \quad (18)$$

In Eq. (18) the average is over the jump frequency probability distribution and the quantity  $\tilde{G}$  is a function of  $S$  and  $\sigma$  which is defined by

$$\tilde{G} = \int_{1-BZ} \frac{d^D \mathbf{k}}{(2\pi)^D} \frac{1}{s + 2D\sigma [1 - p(\mathbf{k})]} \quad , \quad (19)$$

where the integral is over the first Brillouin zone ( $-\pi < k_i < \pi$ ) and

$$p(\mathbf{k}) = \frac{1}{D} \sum_{i=1}^D \cos(k_i) \quad . \quad (20)$$

For  $S \rightarrow \infty$  one has  $s\tilde{G} \rightarrow 1$ . Thus, as is easy to see, Eq. (18) implies the correct high frequency limit Eq. (15). In the limit  $S \rightarrow 0$  one has  $s\tilde{G} \rightarrow 0$ . Thus, the EMA equation for the DC conductivity in  $D$  dimensions is

$$\left\langle \frac{\Gamma - \sigma(0)}{\Gamma + (D-1)\sigma(0)} \right\rangle_{\Gamma} = 0 \quad . \quad (21)$$

For  $D=1$  Eq. (21) gives the correct result Eq. (13). In two dimensions the EMA prediction for the DC conductivity has the correct activation energy. This is because the EMA gives the correct percolation threshold in 2-D [47]. In three dimensions

the EMA predicts  $p_c=1/3$  which is substantially different from the  $p_c=1/4$  found from simulations. Therefore the EMA predicted DC conductivity activation energy is also inaccurate in 3-D. As  $D \rightarrow \infty$  the EMA becomes exact [48].

For the numerical solution of Eq. (18) one needs to calculate the quantity  $\tilde{G}$  of Eq. (19). This is a standard exercise in calculating the diagonal element of the Green's function for a random walk on a cubic lattice or, equivalently, for the quantum mechanical tight-binding model [49]. In 1-D one finds [49,50]

$$s \tilde{G}_{1-D} = \left(1 + \frac{4}{x}\right)^{-\frac{1}{2}} \quad (x=s/\sigma) \quad . \quad (22)$$

In two dimensions one finds [49,50]

$$s \tilde{G}_{2-D} = \frac{2}{\pi} \frac{x}{4+x} K\left(\frac{4}{4+x}\right) \quad (x=s/\sigma) \quad , \quad (23)$$

where  $K$  is the complete elliptic integral of the first kind. In three dimensions one finds [49]

$$s \tilde{G}_{3-D} = \frac{x}{2\pi^2} \int_0^\pi t(\phi) K[t(\phi)] d\phi \quad (24)$$

$$\text{where: } x=s/\sigma \quad , \quad t(\phi) = \frac{4}{x+6-2\cos(\phi)}$$

Note that Eqs. (22), (23) and (24) all imply  $s\tilde{G} \rightarrow 1$  as  $s \rightarrow \infty$ , as required by Eq. (19). A rough analytical approximation to Eq. (24) [46] is



$$s\tilde{G}_{3-D} = \frac{2}{1 + \frac{6}{x} + \sqrt{1 + \frac{12}{x}}} \quad (x=s/\sigma) \quad , \quad (25)$$

that works best for  $x$  larger than one.

### 3. A NEW METHOD FOR THE NUMERICAL EVALUATION OF THE FREQUENCY DEPENDENT CONDUCTIVITY IN HOPPING MODELS

The frequency dependent conductivity of a hopping model may be evaluated numerically in several ways. In principle the problem is straightforward, namely to solve a large system of linear equations. But in practice this is not easy and several methods exist that are not all equally useful. For the present work, where the interest is focussed on the low temperature regime, it was found necessary to develop a new method. Before this method is presented, a brief review is given of the previously available methods.

The obvious method for calculating the frequency dependent conductivity is to solve the master equation numerically. In a periodic external field this equation generates a system of linear equations with complex coefficients; from the solution it is straightforward to calculate the conductivity. The Gauss-Seidel or the Jacobi relaxation methods [51] are usually applied to such a problem. Unfortunately, they converge much too slowly if the coefficients vary over several orders of magnitude, as is the case when one wants to study the hopping at low temperatures. Overrelaxation methods [51] may be faster, but are still not fast enough.

The most common way of evaluating the frequency dependent conductivity in hopping models is to use an equivalent of Monte Carlo simulations. This method works fine at high temperatures, but for  $\beta$  larger than about 10 any charge carrier gets caught in places where it tends to jump to and fro the same place without

moving away until after thousands of Monte Carlo steps. This behaviour reflects the real physics at low temperatures in a solid described by a hopping model. But it makes the method highly inefficient at these low temperatures.

We now proceed to describe an alternative method for evaluating  $\sigma(\omega)$  for a hopping model. The method applies a systematic reduction to the AC Miller-Abrahams (ACMA) equivalent circuit. The reduction ends up with an admittance matrix from which the conductivity is easily calculated. This matrix depends on the frequency so the reduction must be repeated for each frequency. To describe the method we first review the one-dimensional ACMA equivalent circuit, and then show how to reduce the circuit. Finally, the generalization of the method to higher dimensions is considered.

We want to solve Eq. (9) for small electrical fields. For these small fields the probabilities  $P_i$  are only slightly different from the average probability  $\langle P \rangle$  and we write  $P_i = \langle P \rangle + \delta P_i$ . When substituted into Eq. (9) this gives to first order  $\epsilon$  where  $\epsilon = \beta q a \langle P \rangle E$  is a dimensionless electric field

$$\begin{aligned} \frac{d}{dt} \delta P_i = & \Gamma_o(i-1, i) [\delta P_{i-1} - \delta P_i] - \Gamma_o(i, i+1) [\delta P_i - \delta P_{i+1}] \\ & + e [\Gamma_o(i-1, i) - \Gamma_o(i, i+1)] \end{aligned} \quad (26)$$

In this equation  $\epsilon$  may depend on time in any arbitrary way.

Consider now the electrical circuit shown in Fig. 2a. The capacitors all have capacity equal to one while the (real) conductance between site  $i$  and  $i+1$  is  $\Gamma_o(i, i+1)$ . The voltage

generators impose the potential drop  $-i\epsilon$  from the capacitors to the ground. If the voltage at site  $i$  is denoted by  $U_i$ , the Kirchhoff law expressing current conservation is

$$\frac{d}{dt} [U_i + i\epsilon] = \Gamma_o(i-1, i) [U_{i-1} - U_i] - \Gamma_o(i, i+1) [U_i - U_{i+1}] \quad . \quad (27)$$

This equation transforms into Eq. (26) if one makes the identification

$$U_i = \delta P_i - i\epsilon \quad . \quad (28)$$

Solving Eq. (26) therefore becomes equivalent to "solving" the ACMA circuit [25, 52-54]. To completely specify the problem the boundaries must be considered [14]. We use perfectly conducting electrodes for which the boundary conditions are  $\delta P_0 = \delta P_N = 0$  (Eq. (6)). For  $i=0$  this implies  $U_0 = 0$  while for  $i=N$  the condition is  $U_N = -N\epsilon$ . These two boundary conditions correspond to the circuit endings of Fig. 2b.

Before it is shown how to "solve" the ACMA circuit, let us consider the calculation of the frequency dependent conductivity from the solution. By definition,  $\sigma(\omega)$  is the [complex] ratio between the spatially averaged current and the electric field in a steady periodic situation. If  $\langle J \rangle$  denotes the spatially averaged current in the "rationalized" unit system behind Eqs. (13) and (15), we have (where  $K$  is a proportionality constant)

$$\langle J \rangle_{\epsilon} = K \sum_{i=0}^{N-1} [\Gamma(i \rightarrow i+1) P_i - \Gamma(i+1 \rightarrow i) P_{i+1}] \quad (29)$$

To first order in the electric field this expression via Eqs. (8) and (28) reduces to

$$\begin{aligned} \langle J \rangle_{\epsilon} &= K \sum_{i=0}^{N-1} \Gamma_o(i, i+1) \left[ 2 \frac{e}{2 \langle P \rangle} \langle P \rangle + \delta P_i - \delta P_{i+1} \right] \\ &= K \sum_{i=0}^{N-1} \Gamma_o(i, i+1) [U_i - U_{i+1}] \\ &= K \sum_{i=0}^{N-1} I_R(i \rightarrow i+1) \end{aligned} \quad (30)$$

Here  $I_R(i \rightarrow i+1)$  is the current in the resistor from site  $i$  to site  $i+1$  in Fig. 2a.

At very high frequencies the capacitors may be ignored and the potential drop across each resistor simply becomes  $e$ . In order to reproduce Eq. (15) for the high frequency conductivity the general expression for the conductivity in the rationalized unit system must therefore be

$$\sigma = \frac{1}{N} \sum_{i=0}^{N-1} \Gamma(i, i+1) [U_i - U_{i+1}] \quad (\epsilon=1) \quad , \quad (31)$$

or

$$\sigma = \frac{1}{N} \sum_{i=0}^{N-1} I_R(i \rightarrow i+1) \quad (\epsilon=1) \quad . \quad (32)$$

In the limit  $\omega \rightarrow 0$  the capacitors are completely blocking and only the voltage generator at the site  $i=N$  matters (where there is no capacitor, compare Fig. 2b). Thus, the ACMA circuit effectively reduces to resistances in series and the current is

the same in each resistor. This current is determined by the total resistance from  $i=0$  to site  $i=N$ . Each resistor has the value  $1/\Gamma(i, i+1)$  and the total resistance is the sum of all resistors. When the current thus determined is substituted into Eq. (32) one finds the expression given in Eq. (13) for the one-dimensional DC conductivity of a hopping model.

Returning now to the case of an arbitrary frequency (but still in 1-D), it is convenient to rewrite Eq. (32) in terms of the current through each voltage generator. If  $I_V(i)$  denotes the current "upwards" through the  $i$ 'th voltage generator towards site  $i$ , current conservation implies

$$\begin{aligned} I_R(0-1) &= I_V(0), \\ I_R(1-2) &= I_R(0-1) + I_V(1) = I_V(0) + I_V(1) \quad ; \end{aligned} \quad (33)$$

in general

$$I_R(i-i+1) = I_V(0) + \dots + I_V(i) \quad (34)$$

When substituted into Eq. (32) this gives

$$\sigma = \frac{1}{N} \sum_{i=0}^N (N-i) I_V(i) \quad (\epsilon=1) \quad (35)$$

Equation (35) suggests regarding the ACMA circuit as an  $nN$  port consisting of all the capacitors and the resistors as "internal" elements with external nodes that are to be subjected to the potentials  $-\epsilon, \dots, -N\epsilon$ . Such a circuit is characterized by a [frequency dependent] symmetric matrix of admittances,  $Y[i, j]$  ( $i, j=0, \dots, N$ ) (the index 0 refers to the ground). In particular, it is possible from this matrix to calculate the

generator currents  $I_V(i)$  corresponding to  $\epsilon=1$  which is given by (the value of  $Y[i, i]$  may be any number in the below expression)

$$\epsilon=1: \quad I_V(i) = \sum_{j=0}^N (j-i) Y[i, j] \quad . \quad (36)$$

Substituting this into Eq. (35) one finally arrives at

$$\sigma = \frac{1}{N} \sum_{i=0}^N \sum_{j=0}^N (N-i) (j-i) Y[i, j] \quad . \quad (37)$$

The problem is now reduced to calculating the admittance matrix. This is done by utilizing a transformation which is well-known from electrical engineering. This transformation, which was introduced into the random resistor network community by Fogelholm [55], is a prescription of how to remove nodes from a circuit without changing the "external" properties of the circuit. Consider any node in an electrical circuit connected to  $m$  other nodes by the admittances  $Y_1, \dots, Y_m$ . This is illustrated in Fig. 3 for the case  $m=5$ . The central node may be removed by introducing new admittances between all possible pairs of the  $m$  neighbor nodes. The new admittance between the neighbor nodes  $i$  and  $j$ ,  $Y_{ij}$ , is given by

$$Y_{ij} = \frac{Y_i Y_j}{Y_1 + \dots + Y_m} \quad . \quad (38)$$

If some of the  $m$  neighbor nodes are already connected by an admittance, this admittance is increased by the amount given by Eq. (38). Before proceeding, let us briefly reflect on the question: What does it actually mean physically that the new

circuit is "equivalent" to the old? A little thought reveals that this means that, for all possible choices of potentials applied to the  $m$  neighbor nodes, the same currents run from each of these nodes into the circuit. In this sense, the  $m$  neighbor nodes cannot detect any difference between the circuits before and after the reduction. Once this condition has been specified, it is straightforward to derive Eq. (38).

When this transformation is applied to the ACMA circuit each of the "internal" nodes indexed  $i=1, \dots, N$  is removed. One is then left with all possible connections between the "external" nodes that are directly connected to the voltage generators. Each connection has an admittance which specifies the corresponding matrix element in the admittance matrix  $Y[i, j]$ . With this method for evaluating  $\sigma(\omega)$  of a hopping model the number of calculations that are to be performed is independent of the temperature. This is in contrast to the two "standard" methods, solving the linear equations or performing a Monte Carlo simulation. A further advantage is that the present method (which proceeds through a number of simple algebraic operations on the circuit admittances) introduces virtually no numerical inaccuracies. Thus, the conductivity is evaluated with high precision.

The method is easily generalized to higher dimensions. Considering the case  $D=2$ , the ACMA circuit is a square lattice whose nodes are indexed by  $(i, k)$  where  $i=0, \dots, N$  and  $k=1, \dots, N$ . Each node on the lattice is connected to the ground via a capacitor and a voltage generator [14,54]. Neighboring



nodes are connected by a resistor whose conductance (admittance) is the jump frequency. The external electric field is assumed to be in the direction of the x-axis (indexed by  $i=0, \dots, N$ ). This means that the voltage generators (just as in 1-D) have a voltage equal to  $-i\epsilon$ , and this voltage is independent of  $k$ . In effect there is thus only one voltage generator for each  $i$ . In the y-direction we use periodic boundary conditions so that the point  $(i, N+1)$  is identified with the point  $(i, 1)$ . In the x-direction the "perfect electrode" boundary condition is used. For calculating the conductivity, all nodes  $(i, k)$  with  $i=1, \dots, N-1$  are removed according to the recipe of Eq. (38). Since all nodes with the same  $i$ -coordinate below the capacitors are connected to the same voltage generator, one after the reduction ends up with (just as in 1-D) an  $(N+1) \times (N+1)$  symmetric admittance matrix  $Y[i, j]$  (where both indexes refer to x-coordinates). The calculation of the conductivity from the matrix proceeds as in Eq. (37), except that a further division by  $N$  is necessary to compensate for the fact that each "layer" perpendicular to the x-axis has  $N$  parallel channels. We thus get

$$\sigma(s) = \frac{1}{N^2} \sum_{i=0}^N \sum_{j=0}^N (N-i)(j-i) Y[i, j; s] \quad . \quad (39)$$

An  $s=i\omega$  has been introduced here to remind of the fact that the conductivity is frequency dependent.

It is not clear how to find the optimal strategy for choosing in which order the nodes should be removed. This point is important because the removal of one node introduces several

new connections. And the more connections there are to a given node, the more calculations are required for removing it. The nodes should therefore be removed so that as few new connections as possible are created. In the original Fogelholm algorithm at any time one removes the node with fewest connections to its surroundings [56]. This works very well for a system where most neighboring nodes are not connected, as is the case close to the percolation threshold. In the present case, however, where all neighbors are connected, this procedure becomes inefficient, because the last nodes to be removed become excessively costly. We found it better to "contain the damage" by removing one column at a time (i. e., the nodes with same index  $i$ ). After the first  $p$  columns have been removed one has a situation where the  $p+1$  "electrodes" connected to the voltages  $0, -1, \dots, -p$  are all connected to each other (i. e.,  $Y[i, k] \neq 0$  for  $i, k=0, \dots, p$ ). Furthermore, all possible connections exist from these  $p+1$  "electrodes" to the  $N$  nodes of the  $(p+1)$ 'th column that is to be removed next, and all nodes in the  $(p+1)$ 'th column are also connected to each other. Clearly, during the reduction a large number of connections are created. A further optimization of the algorithm is obtained by, after removing the first  $N/2$  columns, starting from the other end of the circuit by removing columns in decreasing order of the  $i$ -index. Thus, the last column to be removed is the  $N/2+1$ 'th.

#### 4. COMPUTER SIMULATIONS

The algorithm derived in Sec. 3 was applied to a study of hopping conduction in two dimensions. At low temperatures large lattices are needed to obtain reasonable statistics. We chose to simulate hopping on a 100X100 lattice. For this system a standard workstation calculates the conductivity (at one particular frequency) in about one minute. Even this large lattice is not selfaveraging at low temperatures, however, and it was necessary to average over several simulations to obtain reproducible results.

In each simulation a 100X100 lattice was generated by, for each link, choosing a random activation energy according to the probability distribution under study. Five different probability distributions [20] were used: The asymmetric Gaussian, the Cauchy, the Exponential, the Power law with exponent -4, the Box. The details of how the energies were generated is described in Ref. 20; to avoid spurious correlations the random numbers were generated according to the RAN0 algorithm of Ref. 51.

For each lattice the frequency dependent conductivity was evaluated from Eq. (39) at a number of frequencies, where the admittance matrix  $Y[i,j;s]$  results from the reduction of the ACMA circuit. For simplicity, the simulations were carried out at real Laplace frequencies, corresponding to purely imaginary frequencies. This trick simplifies the computations and, since the conductivity is real for real Laplace frequencies, makes it possible to present the frequency dependent conductivity in one single curve.

In order to identify the frequency range marking the onset of AC conduction, the BNN relation [9-13] was used. This relation is found in experiment [9-12] as well as in any hopping model [13,35,57]. The BNN relation expresses the fact that the frequency marking the onset of AC conduction,  $\omega_m$ , is proportional to the DC conductivity. The value of the DC conductivity is known roughly because the percolation energy (which according to Eq. (16) is the DC conductivity activation energy) can be calculated analytically for each probability distribution [20].

Figure 4 shows a log-log plot of the results for the five activation energy distributions at the dimensionless inverse temperatures  $\beta=5,10,20,40$ , where each point represents the average of 40 lattices. If  $p(E)$  is the normalized energy probability distribution under study and one introduces  $\beta=\beta/[8\pi p(E_c)]$ , the quantities  $s'$  and  $\tilde{\sigma}$  in Fig. 4 are defined by

$$s' = \frac{\beta}{\sigma(0)} s \quad , \quad \tilde{\sigma} = \frac{\sigma(s')}{\sigma(0)} \quad . \quad (40)$$

In Fig. 4 the full curves are the EMA predictions. These were found by solving Eq. (18) numerically. In two dimensions the quantity  $s\tilde{\sigma}$  is given by an elliptic function (Eq. (23)). A numerical approximation to this function was used [58], Eq. (18) was discretized into 10.000 terms and then solved by the bisection method.

The EMA is usually derived from a perturbation expansion

around an ordered state. As such, there is no a priori reason to believe in the EMA predictions for a severely disordered system as the low-temperature hopping model studied here. This model is really quite extreme: It involves jump frequencies that for the  $\beta=40$  case vary 20-60 decades! This extreme variation implies that the DC conductivity is very small and that the scaling of the frequency introduced in Eq. (39) shifts the frequency by in some cases more than 15 decades. In this light it must be said that the EMA after all is doing well in Fig. 4.

##### 5. THE LOW TEMPERATURE LIMIT OF THE EMA: THE APPEARANCE OF UNIVERSALITY

This section studies the EMA prediction for the  $T \rightarrow 0$  limit of the frequency dependent conductivity in symmetric hopping models in more than one dimension. It was recently shown [19,20] that in the "macroscopic" model for AC conduction the frequency dependent conductivity follows Eq. (1) at low temperatures, independently of the activation energy probability distribution. It is shown below, using a method similar to that of Refs. 19 and 20, that the same equation appears at low temperatures by the EMA treatment of hopping models, despite the considerable physical difference there is between these two models.

The derivation starts by noting that

$$x = \frac{S}{\sigma} \ll 1 \quad (\beta \rightarrow \infty) \quad (41)$$

for all frequencies in an increasingly large range around the frequency marking the onset of AC conduction. This observation, which is crucial to the below derivation, was first made by Bryksin in paper from 1980 [26] dealing with electrons tunneling between random positions. In the derivation given below Eq. (41) will be first assumed and then Eq. (41) is shown to be consistent with the result derived.

Because of Eq. (41), when Eq. (18) is rewritten

$$0 = \left\langle \frac{\Gamma - \sigma}{\Gamma - \sigma + \frac{D}{1 - S\tilde{G}} \sigma} \right\rangle_{\Gamma} \quad (42)$$

one has to first order in  $S\tilde{G}$  (where the numerator is rewritten for convenience below)

$$0 = \left\langle \frac{\Gamma + [(D-1) + Ds\tilde{G}] \sigma - D(1 + S\tilde{G}) \sigma}{\Gamma + [(D-1) + Ds\tilde{G}] \sigma} \right\rangle_{\Gamma} \quad (43)$$

If we introduce the notation  $\Gamma(E) = \Gamma_0 e^{-\beta E}$  to emphasize the activation energy dependence of the jump frequency and if the jump frequency average is converted into an average over the activation energy probability distribution, Eq. (43) becomes

$$\frac{1}{D(1 + S\tilde{G}) \sigma} = \left\langle \frac{1}{\Gamma(E) + [(D-1) + Ds\tilde{G}] \sigma} \right\rangle_E \quad (44)$$

In the  $\beta \rightarrow \infty$  limit of Eq. (44) the jump frequency  $\Gamma(E)$  varies extremely rapidly and, for given  $\sigma$  and  $S$ , there are essentially just two extreme possibilities, depending on  $E$ :

Either one has  $\Gamma(E) \ll [(D-1) + Ds\tilde{G}] \sigma$  or the opposite extreme. In the former case  $\Gamma(E)$  may be ignored altogether from the denominator, while in the latter case the denominator becomes very large and gives little contribution to the right hand side of Eq. (44). The energy separating the two cases,  $E_g(s)$ , is given as the solution of  $\Gamma(E) = [(D-1) + Ds\tilde{G}] \sigma$ , thus

$$E_g(s) = \frac{-1}{\beta} \ln \left( \frac{[(D-1) + Ds\tilde{G}] \sigma}{\Gamma_0} \right) . \quad (45)$$

Accordingly, the right hand side of Eq. (44) for  $\beta \rightarrow \infty$  becomes

$$\left\langle \frac{1}{\Gamma(E) + [(D-1) + Ds\tilde{G}] \sigma} \right\rangle_E = \frac{1}{[(D-1) + Ds\tilde{G}] \sigma} \int_{E_g(s)}^{\infty} P(E) dE \quad (46)$$

and Eq. (44) reduces to

$$\frac{D-1}{D} + \frac{s\tilde{G}}{D(1+s\tilde{G})} = \int_{E_g(s)}^{\infty} P(E) dE . \quad (47)$$

Evaluating Eq. (47) at  $s=0$  gives an expression for  $(D-1)/D$ .

When this expression is subtracted from Eq. (47), one gets

$$\frac{s\tilde{G}}{D(1+s\tilde{G})} = \int_{E_g(s)}^{E_g(0)} P(E) dE . \quad (48)$$

For large  $\beta$ ,  $E_g(0)$  is close to  $E_g(s)$  and therefore the integral on the right hand side can easily be evaluated. If  $p(E_g(0))$  is denoted by  $p_0$ , the integral is simply  $p_0 [E_g(0) - E_g(s)]$  at low temperatures. If the symbol  $\tilde{\sigma} = \sigma/\sigma(0)$  is introduced into Eq. (48) it thus becomes

$$\frac{s\tilde{G}}{D(1+s\tilde{G})} = \frac{P_0}{\beta} \ln \left[ \left( 1 + \frac{D}{D-1} s\tilde{G} \right) \tilde{\sigma} \right] . \quad (49)$$

To leading order in the small quantity  $s\tilde{G}$  Eq. (49) reduces to

$$\frac{\beta}{D P_0} s\tilde{G} = \ln[\tilde{\sigma}] . \quad (50)$$

Since for  $D=1$  one has  $E_g(0) = \infty$ , the derivation assumes  $D > 1$ .

For any dimension  $D > 1$ , however, as  $\beta \rightarrow \infty$   $E_g(0)$  approaches the DC conductivity activation energy which is equal to the percolation energy defined by Eq. (17); thus

$$P_0 = p(E_c) . \quad (51)$$

In the further development one has to distinguish between the cases  $D=2$  and  $D > 2$ . In the latter case, which is the simplest, the quantity  $s\tilde{G}$  as function of  $x$  contains a regular first-order term (after this term there are, e. g. in 3-D, non-analytic terms which are of no concern here). Writing

$$D > 2: \quad s\tilde{G} = \alpha_D x \quad (x \rightarrow 0) , \quad (52)$$

Eq. (19) implies

$$\alpha_D = \frac{1}{2} \frac{1}{(2\pi)^D} \int_{1-BZ} \frac{d^D \mathbf{k}}{D - (\cos k_1 + \dots + \cos k_D)} . \quad (53)$$

Note that this integral is defined only for  $D > 2$ . For  $D=3$  one has  $\alpha_3 = 0.253$  [59]. Substituting now the expansion Eq. (52) into

Eq. (50) leads to Eq. (1):  $\tilde{\sigma} \ln(\tilde{\sigma}) = \tilde{S}$ , where



$$D > 2: \quad \tilde{s} = \frac{\beta \alpha_D}{D p_0 \sigma(0)} s \quad (54)$$

Finally, the consistency of the derivation is checked: The assumption Eq. (41) is indeed satisfied, since for  $\beta \rightarrow \infty$  one has  $x = \frac{s}{\sigma(0)} \rightarrow 0$  for fixed  $\tilde{\sigma}$  and  $\tilde{s}$ .

Turning now to the two-dimensional case, we use the asymptotic expansion of the complete elliptic integral of the first kind [60]: For  $k \rightarrow 1$  one has  $K(k) = \ln(4/k')$  where  $k^2 + k'^2 = 1$ . This implies that  $K(k) = \frac{-1}{2} \ln(1-k)$  for  $k \rightarrow 1$ .

Thus,  $K\left(\frac{4}{4+x}\right) = \frac{-1}{2} \ln(x)$  for  $x \rightarrow 0$ . When this is substituted into

Eq. (23) one finds asymptotically

$$s \tilde{G}_{2-D} = \frac{-1}{4\pi} x \ln(x) \quad (x \rightarrow 0) \quad (55)$$

If the quantity

$$\beta = \frac{\beta}{8\pi p_0} \quad (56)$$

is introduced Eq. (50) thus becomes

$$\ln(\tilde{\sigma}) = \beta \frac{s}{\sigma} \ln\left(\frac{\sigma}{s}\right) \quad (57)$$

Defining now

$$D=2: \quad \tilde{s} = \frac{\beta \ln(\beta)}{\sigma(0)} s \quad (58)$$

Eq. (57) becomes

$$\ln(\bar{\sigma}) = \beta \frac{1}{\bar{\sigma}} \frac{\bar{S}}{\beta \ln(\beta)} [ \ln(\bar{\sigma}) - \ln(\bar{S}) + \ln(\beta) + \ln(\ln(\beta)) ] . \quad (59)$$

For fixed  $\bar{\sigma}$  and  $\bar{S}$  as  $\beta \rightarrow \infty$  Eq. (59) reduces to Eq. (1),  $\bar{\sigma} \ln(\bar{\sigma}) = \bar{S}$ . Note that the assumption  $x = S/\sigma \ll 1$  is again satisfied for fixed  $\bar{\sigma}$  and  $\bar{S}$  when  $\beta$  is sufficiently large.

Figure 5 tests the EMA universality prediction against computer simulations. The five different activation energy distributions of Fig. 4 were used, and for each distribution the temperature was chosen so that  $\beta = 4$ . Each point in the figure represents the average of 100 simulations of a 100X100 lattice. There is not a good agreement between prediction and simulations, but the simulations certainly show the existence of universality. A further discussion of Fig. 5 is given in the next section.

## 6. DISCUSSION

In this paper a new method for the numerical simulation of symmetric hopping models has been derived. The method makes use of the AC Miller Abrahams circuit which interpretes the hopping master equation in terms of an electric circuit. Previously, this circuit was mainly used for developing an intuition about hopping problems. However, it was also used by Summerfield and Butcher [54] for deriving the extended pair approximation (EPA), an approximate analytical method for the calculation of the frequency dependent conductivity, that in many respects is similar to the EMA.

The new simulation method allows a faster and more accurate calculation of  $\sigma(\omega)$  for symmetric hopping models for larger lattices and at lower temperatures than previous methods. Thus, the standard Monte Carlo type method is useless if one wants to study low temperatures like the  $\beta=5,10,20,40$  of Fig. 4. The standard relaxation methods for solving linear equations are also too slow in this regime where the coefficients vary many, many orders of magnitude. With the method presented here it is possible in about one minute to calculate the frequency dependent conductivity with a very high accuracy at any one particular frequency for a 100X100 symmetric hopping model on a standard workstation.

The numerical method may be generalized immediately to deal with non-symmetric hopping models; the only change is that in the ACMA circuit the capacitors then vary from site to site.

There exists a clever algorithm for solving linear

equations with coefficients that vary many orders of magnitude. This is the algebraic multigrid algorithm (AMG) [61,62] which is available from the Yale multigrid library in a well-documented and carefully debugged Fortran version, AMG1R5 [63]. The AMG is an algebraic generalization of the multigrid method for solving elliptic partial differential equations. The method has been tested successfully for large random admittance networks with admittances varying many orders of magnitude [20,64]. It solves the Kirchhoff equations in a time only proportional to the number of equations. For the present problem, the AMG solves the problem in  $D$  dimensions in a time  $\propto N^D$ . At first sight, this is much better than the method presented in Sec. 3 which, as is easy to show, calculates the conductivity in a time  $\propto N^{3D-2}$ . However, in the practical use of the AMG it is not superior to the method of Sec. 3. Thus, when applied to a hopping problem at low temperatures, the AMG easily runs into overflow problems, whereas the method of Sec. 3 avoids such problems. At low temperatures, if one wants to calculate the conductivity by solving Eq. (9) or the higher dimensional analogues, the solution must be extremely accurate. The standard double precision real number representation is not enough, since the equations should be solved with an accuracy of 50-100 digits (depending on how low the temperature is). Unfortunately, higher precissions are not hardware implemented today and therefore very slow. Therefore, the method presented in Sec. 3 seems to be the best available at present. On a longer time span it seems to be likely that the AMG will eventually become the best choice. The AMG method is

very impressive, and it is probably the "final" solution to the problem of solving hopping models numerically.

The results of extensive computer simulations of a  $100 \times 100$  lattice in 2-D was reported in Sec. 4. No simulations were performed in 3-D because the method here is too slow for solving large lattices. However, it works fine in 2-D. In order to obtain reproducible results at low temperatures, it was necessary to average over several simulations of different lattices. The main problem in the reproducibility lies at low frequencies; at frequencies where  $\log(\tilde{\sigma}) > 1$  the results [i. e.,  $\tilde{\sigma}$  as function of  $S'$  (Eq. (40))] are quite reproducible.

The results of the computer simulations were compared to the predictions of the EMA at real Laplace frequencies at a number of temperatures in Fig. 4. The use of real Laplace frequencies not only simplifies the calculations, but also makes it possible to represent the results in one curve (instead of two, one for the real part and one for the imaginary part of the conductivity). This curve contains all information about the frequency dependence of the conductivity. This is because the function  $\sigma(S)$  is analytic in the upper half  $S$ -plane so, by analytic continuation, the behavior on the real  $S$ -axis determines the function uniquely. A further virtue of this representation is that deviations from the EMA are here somewhat magnified compared to the use of real frequencies. The "reduced" frequency used in Fig. 4 is not quite the  $\tilde{S}$  of Eq. (58) because, for some of the highest temperatures studied,  $\beta$  becomes less than one, implying that Eq. (58) does not make sense here.

Instead, the related "reduced" frequency  $s' = \beta s / \sigma(0)$  (Eq. (40)) was used in Fig. 4.

As far as is known to the author, these results are the first simulations of a hopping model at low temperatures where the jump frequencies  $\Gamma$  vary over several decades (here up to about 50-60 decades). In general, there is a rather good agreement between the simulations and the EMA, with deviations in the transition region where the EMA at low temperatures underestimates the conductivity. It should be remembered, however, that the EMA is only a mean-field approximation.

At low temperatures the EMA predicts a universal frequency dependence of the conductivity, which should become independent of the activation energy probability distribution (Sec. 5). The equation governing the universal conductivity is  $\tilde{\sigma} \ln(\tilde{\sigma}) = \tilde{S}$ . A special case of this equation was derived by Bryksin [26] for a system of tunnelling electrons in D dimensions. Bryksin's paper from 1980 was the first time Eq. (1) appeared in the literature. The equation was later derived by Fishchuk [24] for the box distribution of activation energies in the "macroscopic" model, and by the present author for a hopping model with the box distribution [13]. Recently, it was shown that Eq. (1) is universal in the low temperature limit of the "macroscopic" model. Here it has been shown that the equation is also universal for symmetric hopping models. What happens is that, at low temperatures the conduction is mainly along the percolation paths, and the only "signature" of the activation energy distribution is the number  $p(E_c)$ . The "effective" activation

energy distribution - the distribution of activation energies that are relevant for the frequencies around the transition frequency - becomes flat, i. e., like the box distribution.

The universality prediction was tested in Fig. 5, which studies five different activation energy probability distributions at the same "reduced" temperature  $\beta=4$ . There is clearly a universality in the sense that the function  $\bar{\sigma}(\bar{\xi})$  is independent of the activation energy distribution. However, the results deviate considerably from the EMA prediction Eq. (1). A careful comparison to Fig. 4, that also contains low temperature data, reveals that one reason for the discrepancy is that the temperature simply is not low enough in Fig. 5. In other words, the EMA predictions for  $\beta=4$  are still not really close to Eq. (1). Unfortunately, it is not possible to go to lower temperatures for a 100X100 lattice without losing reproducibility.

There are interesting differences between the "macroscopic" model [20] and hopping models. Figure 4 indicates a systematic deviation of the simulations from the EMA predictions at low temperatures in the transition region. Here the data give a less sharp transition to frequency dependence than the EMA predicts. In the "macroscopic" model, on the other hand, there is a very good agreement between the EMA predictions and the simulations at all temperatures. It is not clear what is the origin of these differences. Apparently, the hopping model is more complex than the macroscopic model. We thus find larger sample to sample fluctuations in the hopping model than in the macroscopic model

(in 2-D at the same temperature and for the same activation energy distribution). Also, in the derivation of universality for hopping models, one has to distinguish between the case  $D=2$  and  $D>2$ ; this is not necessary for the derivation of Eq. (1) for the macroscopic model [20].

In the simplest possible approximation to hopping models, the continuous time random walk (CTRW) approximation [35], the conductivity for the box distribution of energy barriers is in dimensionless units given by

$$\tilde{\sigma} = \frac{\tilde{S}}{\ln(1+\tilde{S})} \quad (59)$$

As has been shown elsewhere [20], this expression is close to that given by Eq. (1); in particular the two functions  $\tilde{\sigma}(\tilde{S})$  have the same asymptotic expressions for the exponents of the real and imaginary parts of the conductivity. These exponents converge slowly to one as the frequency goes to infinity [20]. A convincing experimental demonstration of this phenomenon has recently been given for metal-cluster compounds [8].

There has been relatively little discussion of universality for AC conduction in the literature. In experiments, a number of authors early pointed out that quite different systems like ionic conductive glasses and amorphous semiconductors have surprisingly similar AC responses [1-3]. In theory, Summerfield in 1985 termed the phrase "quasi-universality" for the fact that a number of different models, when solved in the EPA [57], give almost the same frequency dependence of the conductivity. Since then there seems to have been no discussion in the literature of AC conduction universality, except the recent demonstration of



universality in the "macroscopic" model [19,20].

In a recent paper [20] computer simulations of the "macroscopic" model of AC conduction in disordered solids were presented. This model regards the solid as having a spatially varying [frequency independent] conductivity. The model studies the consequences of Maxwell's equations: the macroscopic conductivity becomes frequency dependent. The "macroscopic" model is physically quite different from hopping models. Thus, in hopping models one has non-interacting charge carriers and, consistent with this, there is a spatially homogeneous electric field. But in the "macroscopic" model, the Coulomb forces are crucial, the electric field is screened and it becomes strongly locally varying. An interesting case between these two models, which has recently been studied numerically by Maass et al [18], is the case of a hopping model with Coulomb repulsion between the charge carriers.

The "macroscopic" model leads to an electrical equivalent circuit where the nodes on a cubic lattice are joined by a capacitor and a resistor in parallel [20,24]. The resistors carry the free charge currents while the capacitor currents are Maxwell's displacement currents [20]. In contrast to the circuit of Fig. 2, there are no connections to the ground and no voltage generators; the "solid" is simply subjected to a macroscopic potential drop at the electrodes (=two opposing end-faces). As mentioned already, when solved in the random admittance version of the EMA, the "macroscopic" model leads to Eq. (1). Simulations confirm this [20]. In the DC limit the "macroscopic" model and hopping models both give simple resistance circuits.

Thus, the DC limit of the EMA hopping equation Eq. (21) is nothing but the EMA equation for a resistance circuit.

An interesting connection between the symmetric hopping model and the "macroscopic" model is that the CTRW approximation [28] to hopping models corresponds to the one-dimensional version of the macroscopic model. As has been discussed in detail elsewhere [20], this version becomes realistic at low temperatures where conduction mainly follows the "percolation" paths. Along these lines, an approximation referred to as the "percolation path approximation" (PPA) was proposed in Ref. 20. Since the PPA is equivalent to the CTRW for hopping models, it also leads to Eq. (60).

Throughout this paper the limit of extreme disorder (where the jump frequencies  $\Gamma$  vary several decades) was arrived at by going to low temperatures for a system with thermally activated hopping. The same limit is also arrived at in a system of localized electrons tunnelling between nearest neighbor sites, when the density of electrons becomes very small. The system of tunnelling electrons has been studied extensively in the past [26,54,66]. Though it has not been spelled out in detail in Sec.5, the same universality prediction (Eq. (1)) applies to this system as to the hopping model with activated jump rates.

There are a number of open problems and work that remains to be done in this field. The symmetric hopping model should be studied numerically at low temperatures also in three dimensions. Large lattices are needed at low temperatures to get reasonable statistics, and the method of Sec. 3 is not useful. Regarding the new method, that works well in 2-D, it is not clear what is

the optimal strategy for removing nodes. From the theoretical point of view the main question is: Is there true universality in the extreme disorder limit, or is there only "quasi-universality"? If true universality does exist, as believed by the author, is the universal function  $\tilde{\sigma}(\tilde{S})$  the same in all dimensions (this seems less likely)? If not, analytical methods more accurate than the EMA should be developed to calculate the universal conductivity. A further question that should be looked into is: What is the cause of the difference between the hopping model and the "macroscopic" model in 2-D, where the EMA works significantly better than for the hopping model? Finally, does the EMA also predict universality for non-symmetric hopping models?

## ACKNOWLEDGEMENTS

The author wishes to thank H. Larsen, I. H. Pedersen, and T. Riedel for technical assistance. This work was supported by the Danish Natural Science Research Council.

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## FIGURE CAPTIONS

### Fig. 1:

Potential felt by a charge carrier in a one-dimensional symmetric hopping model with thermally activated jump rates. If  $\beta$  denotes  $1/k_B T$  and  $\Delta E$  is the energy barrier height, the rate for jumps between two sites is  $\Gamma_0 e^{-\beta \Delta E}$ . At low temperatures the jump rates vary over several decades, and a charge carrier makes many jumps to and fro at sites with low barriers to its surroundings. Thus, the fluctuation-dissipation theorem shows that the conductivity depends strongly on frequency. The DC conductivity activation energy is the largest barrier. In more than one dimension one has for the symmetric hopping models studied here a cubic lattice with a similar random variation of the barrier heights. In this case the DC conductivity activation energy is the largest energy met on any "percolation" path (Eqs. (16) and (17)).

### Fig. 2:

a) AC Miller-Abrahams electrical equivalent circuit of a symmetric hopping model in one dimension. All capacitors have capacity equal to one while the conductance of the resistor between site  $i$  and site  $i+1$  is equal to the equilibrium jump frequency  $\Gamma_0(i, i+1)$ . The voltage generators give voltages that

are proportional to the electrical field in the sample for which  $\epsilon$  is a dimensionless measure; the electric field may depend on time in any arbitrary fashion. The currents in the resistors are equal to the particle currents in the hopping model. Similar electrical equivalent circuits exist in higher dimensions. The voltage depends only on the coordinate in the direction of the electric field, and thus the capacitors in a plane perpendicular to the field are all connected to the same voltage. This fact is crucial for the numerical method for calculation of the frequency dependent conductivity in a hopping model derived in Sec. 3.

b) Boundary conditions to the ACMA circuit in one dimension. These boundary conditions correspond to "perfectly conducting electrodes" (Eq. (6)).

**Fig. 3:**

Reduction of an electrical circuit by removal of a node. In this example the node to be removed is connected to five neighboring nodes by admittances  $Y_1, \dots, Y_5$ . When the node is removed, all possible connections between the five nodes are created by, between the  $i$ 'th and the  $j$ 'th node, introducing the admittance  $Y_i Y_j / (Y_1 + \dots + Y_5)$ . If two neighboring nodes were already connected before the reduction their admittance is increased by this amount. Physically, the fact that the new circuit is equivalent to the old means that for any potentials applied to the five nodes the same currents run into each circuit. The transformation may be applied to the ACMA circuit of Fig. 2 (or

higher dimensional analogues). When all nodes have been removed one is left with an admittance matrix  $Y[i,j]$  which directly determines the frequency dependent conductivity [Eq. (37) for 1-D or Eq. (39) for 2-D].

**Fig. 4:**

Log-log plots of the results of computer simulations in two dimensions (points) compared to the EMA predictions (full curves) for symmetric hopping on a 100X100 lattice. The figures show the conductivity as function of the real Laplace frequency (i. e., at imaginary frequencies). The computer simulations were carried out using the algorithm developed in Sec. 3. The points represent averages over 40 different 100X100 lattices, where the jump frequency activation energy varies according to the following distributions (compare Ref. 20): a) Asymmetric Gaussian, b) Cauchy, c) Exponential, d) Power law with exponent -4, e) Box. Each figure shows the following dimensionless inverse temperatures:  $\beta=5$  ( $\square$ ),  $\beta=10$  ( $\circ$ ),  $\beta=20$  ( $\nabla$ ), and  $\beta=40$  ( $\diamond$ ). The "reduced" Laplace frequency  $s'$  on the x-axis is the scaled Laplace frequency defined in Eq. (40), while  $\tilde{\sigma}=\sigma/\sigma(0)$ . The EMA predictions were found by solving Eq. (18) where  $S\tilde{G}$  is given by Eq. (23).

Fig. 5:

Log-log plot comparing the EMA universality prediction (full curve; Eq. (1)) to computer simulations (points) of a 100X100 lattice for the five different activation energy probability distributions of Fig. 4. Each point represents the average of 100 simulations taken at the "reduced" inverse dimensionless temperature  $\beta=4$  where  $\beta$  is defined in Eq. (56); this corresponds to  $\beta=63.91$  for the Asymmetric Gaussian,  $\beta=32.00$  for the Cauchy,  $\beta=50.27$  for the Exponential,  $\beta=119.66$  for the Power law with exponent -4, and  $\beta=100.53$  for the Box distribution.. The "reduced" Laplace frequency  $\tilde{S}$  is defined in Eq. (58) and  $\tilde{\sigma}=\sigma/\sigma(0)$ . The figure shows results for the following distributions: Asymmetric Gaussian ( $\blacktriangle$ ), Cauchy ( $\circ$ ), Exponential ( $\square$ ), Power law with exponent -4 ( $\triangle$ ), and Box ( $\nabla$ ). The figure clearly shows that there is a universality at low temperatures, but there is not a good agreement with the EMA predictions. Part of the disagreement is due to the fact that the EMA predictions at these temperatures are still not close to Eq. (1). Unfortunately, it was not possible to go to lower temperatures without losing reproducibility (even after averaging 100 times).

Fig. 1

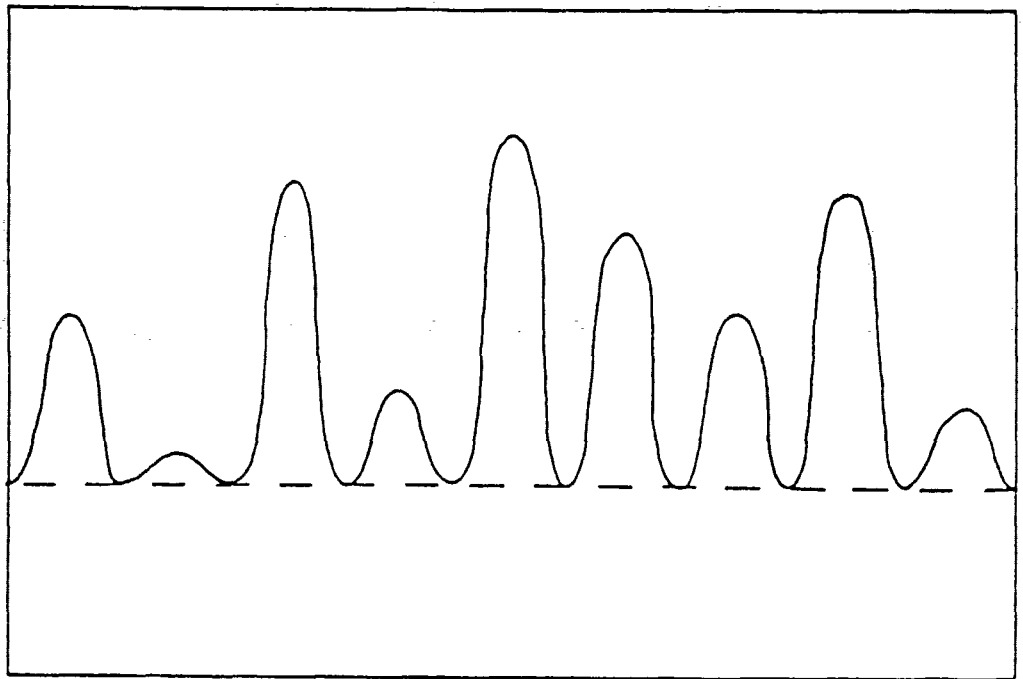


Fig. 2a

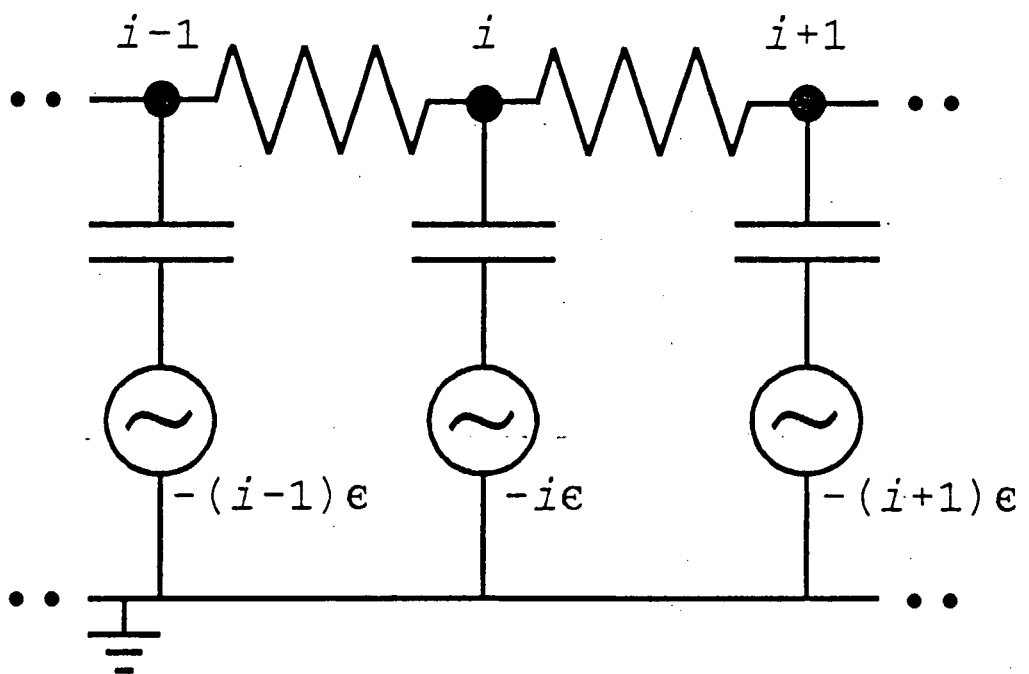


Fig. 2b

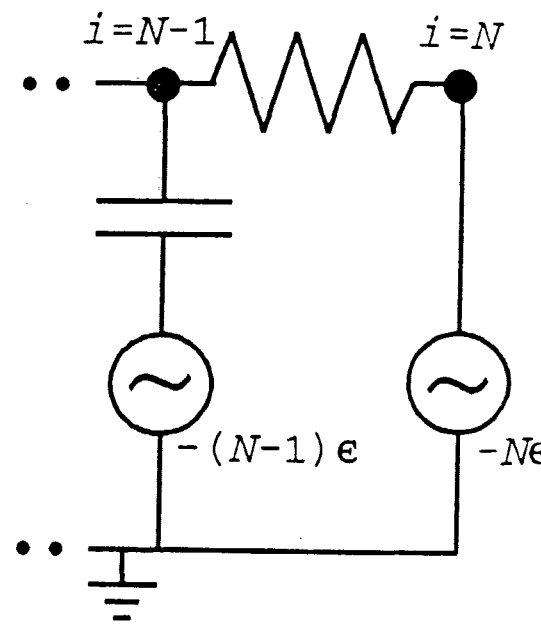
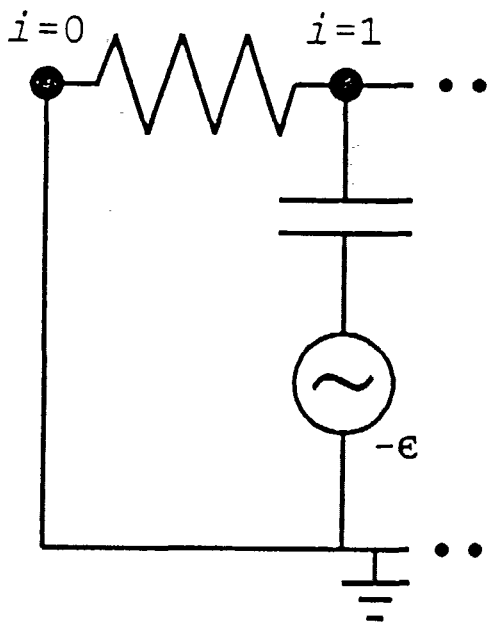


Fig. 3

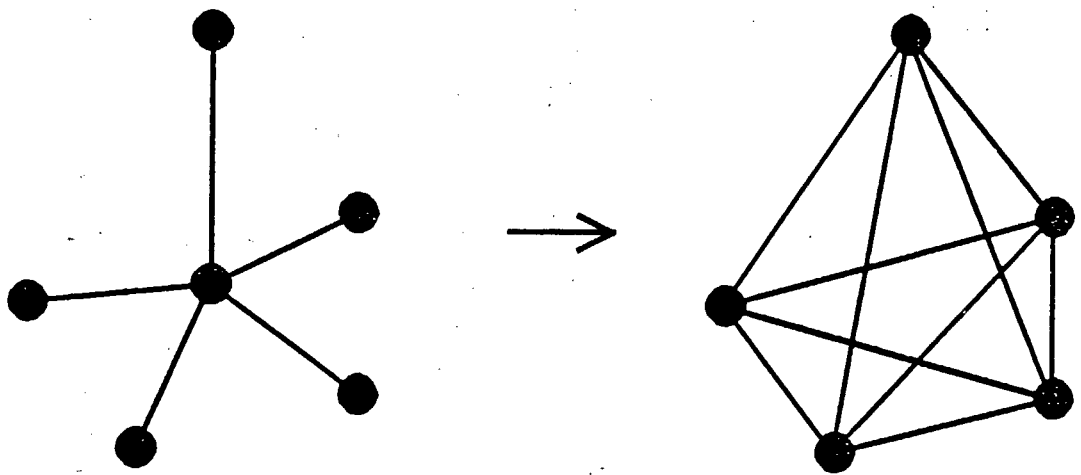




Fig 4 (a)

ASYM GAUSS (INDEX=1) HOP  
Dimension 2

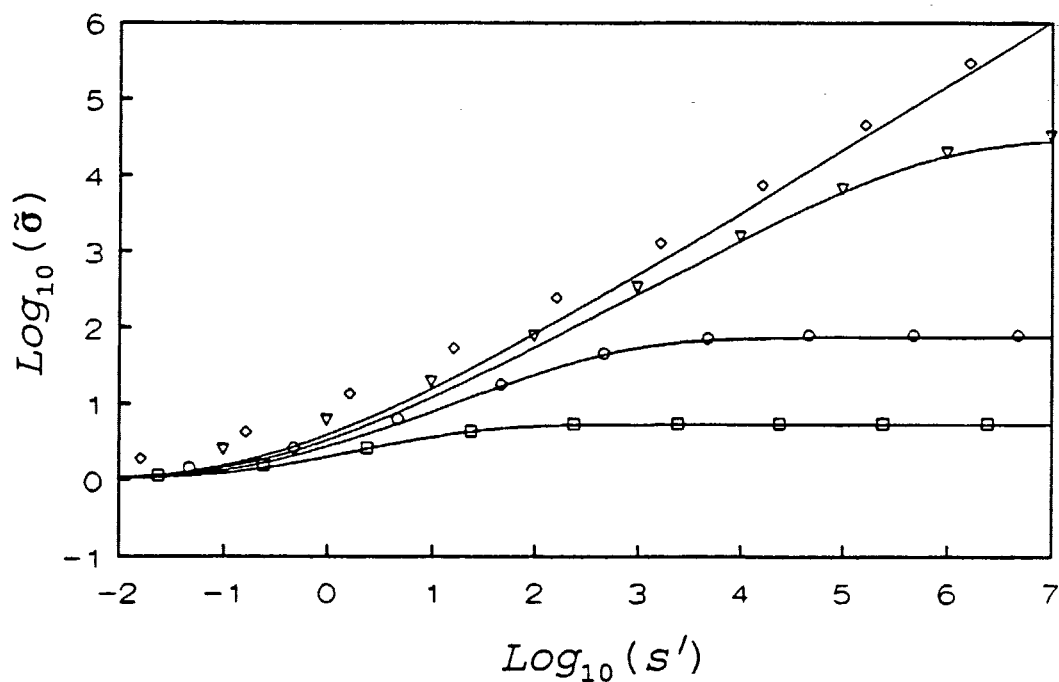


Fig 4 (b)

CAUCHY (INDEX=2) HOP  
Dimension 2

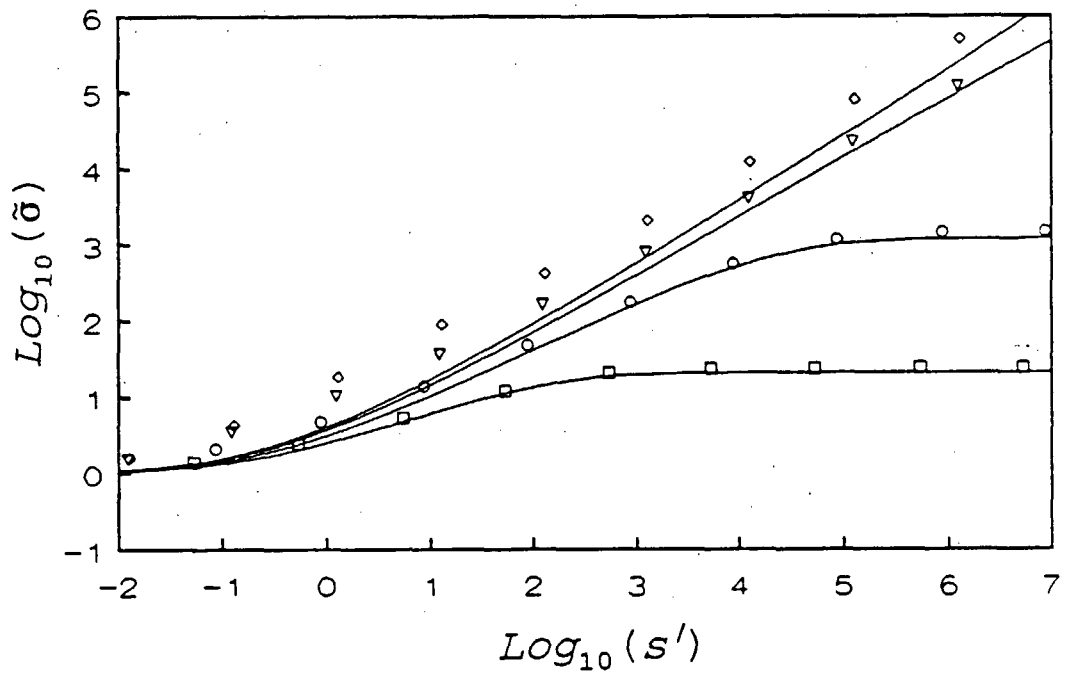


Fig 4 (c)

EXP (INDEX=3) HOP  
Dimension 2

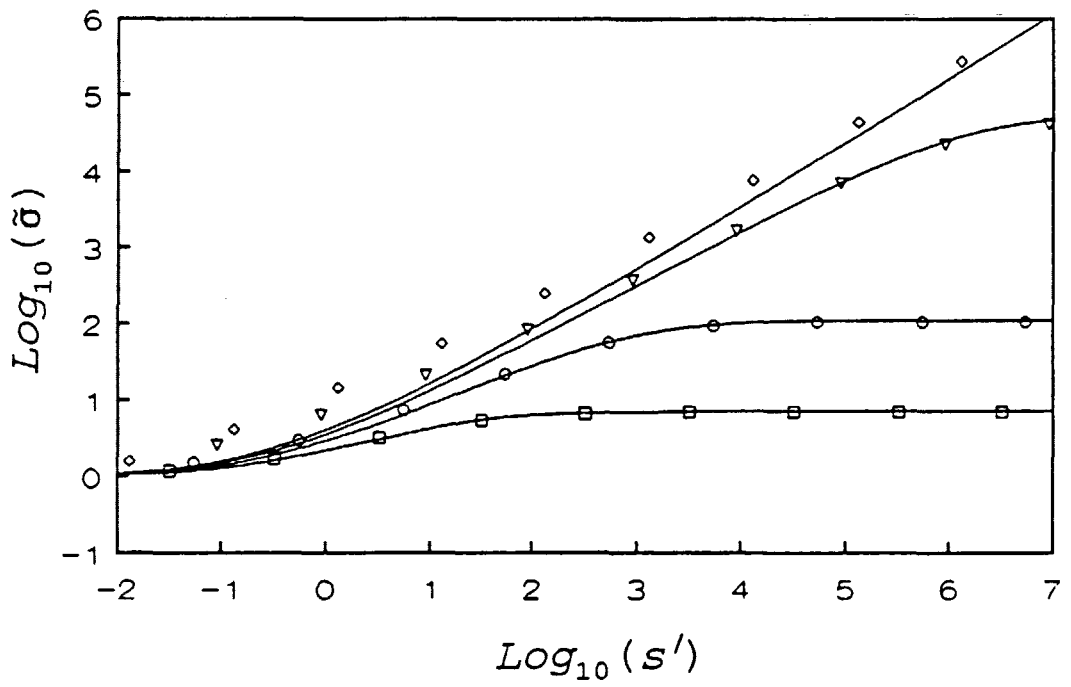


Fig. 4 (d)

POWER (INDEX=4) HOP  
Dimension 2

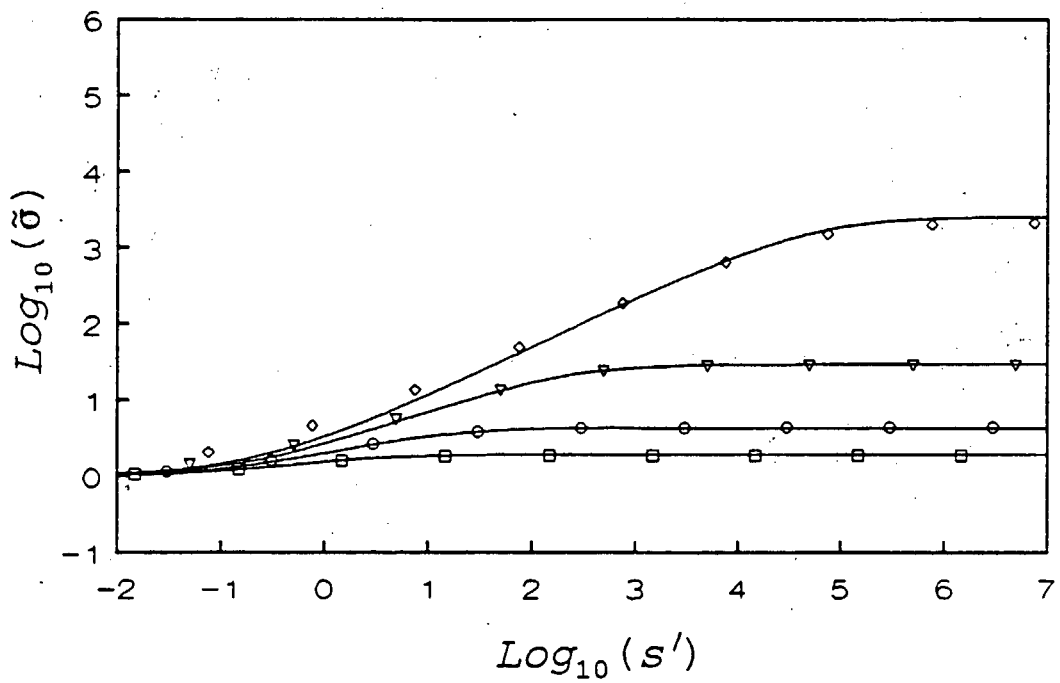


Fig 4 (e)

BOX (INDEX=5) HOP  
Dimension 2

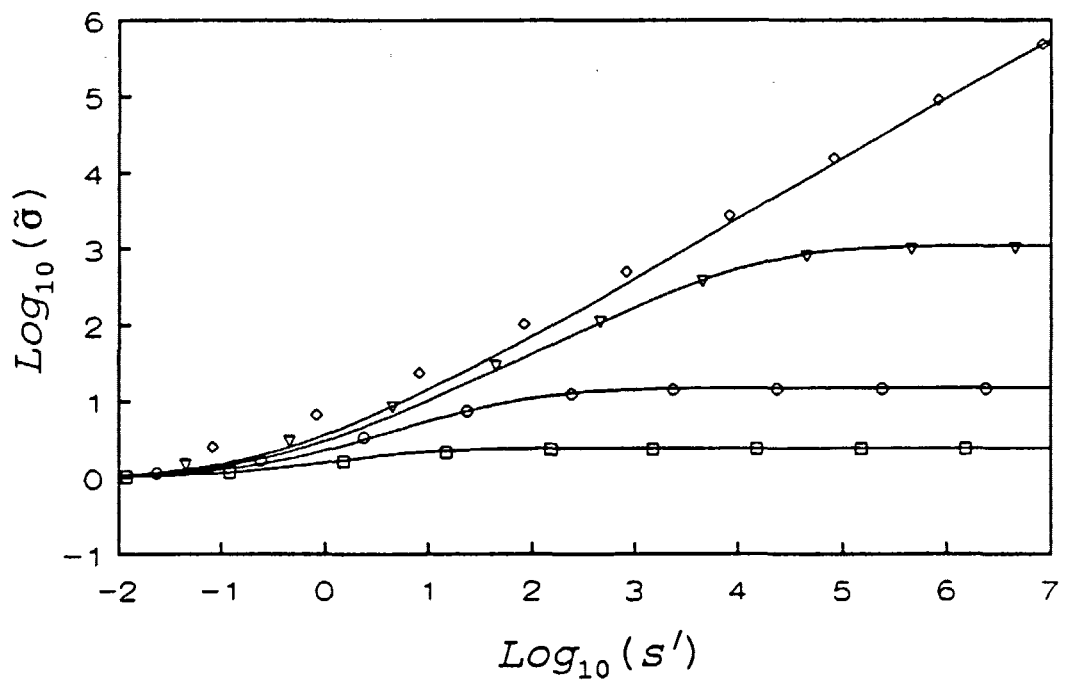
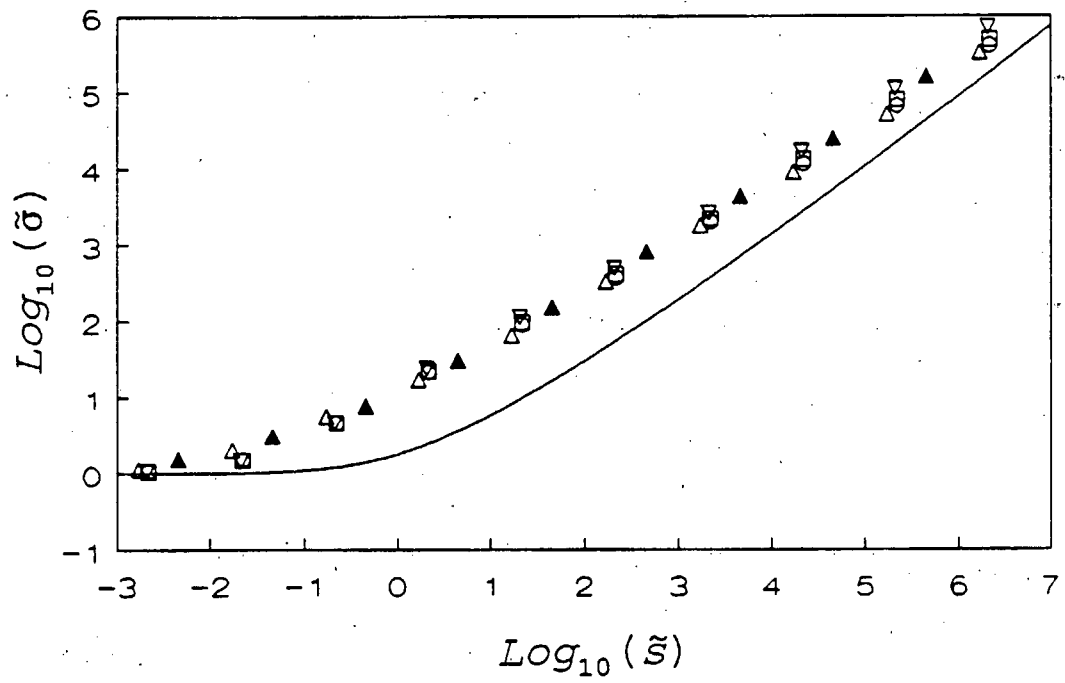


Fig. 5

betatilde=4 (hop)  
D=2



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