Effective one-dimensionality of AC hopping conduction in the extreme disorder limit

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Abstract

It is argued that in the limit of extreme disorder AC hopping is dominated by "percolation paths". Modelling a percolation path as a one-dimensional path with a sharp jump rate cut-off leads to an expression for the universal AC conductivity, that fits computer simulations in two and three dimensions better than the effective medium approximation.

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While ordered solids show no frequency-dependence of their conductivity at frequencies below phonon frequencies, disordered solids are characterized by AC conductivity that varies as an approximate power-law of frequency $[1–6]$. The exponent is usually less than one, but often quite close to one. As the frequency goes to zero the conductivity stabilizes and becomes frequency-independent. The characteristic frequency marking the onset of DC conduction has roughly the same temperature-dependence as the DC conductivity. These features are observed universally for electronically conducting disordered solids like amorphous semiconductors $[1,2,4,7]$, polymers $[8,9]$, doped crystalline semiconductors at helium temperatures $[10]$ (where the random positions of the dopant atoms becomes important), or high temperature superconductors above $T_c$ $[11]$, as well as for ionically conducting disordered solids like glasses or polymers $[2,3,5,6]$.

The standard model for AC conduction in disordered solids is the hopping model $[12–18]$. The simplest version is hopping of non-interacting charge carriers on a regular lattice with random symmetric nearest neighbor jump rates; this is the model to be studied below. Alternatively, a macroscopic approach may be adopted by considering Maxwell’s equations for a solid with spatially randomly varying conductivity $[19–21]$. For both models the limit of extreme disorder may be studied by letting the temperature go to zero when the jump rates/the macroscopic conductivities are thermally activated with randomly varying activation energies. It has recently been shown by computer simulations $[20–22]$ that in the low temperature limit, the AC conductivity becomes universal in both models, i.e., independent of the activation energy probability distribution, $p(E)$. The effective medium approximation (EMA) for both models predicts the same universal AC conductivity in the extreme disorder limit (in more than one dimension). If $\tilde{\sigma}$ denotes the conductivity relative to the DC conductivity and $\tilde{s}$ is a suitably scaled dimensionless imaginary frequency (“Laplace frequency”), the EMA universality equation $[21, 22]$, first derived by Bryksin for the model of electrons tunnelling between randomly localized positions $[13, 23]$, is

$$\tilde{\sigma} \ln \tilde{\sigma} = \tilde{s}.$$ (1)
Computer simulations of the macroscopic model \[20,21\] showed that this equation works very well in two and three dimensions. The situation is different for hopping. While the existence of universality was confirmed in two dimensions, it was found \[22\] that the onset of AC conduction is considerably smoother than predicted by Eq. (1). No simulations of AC hopping in three dimensions have so far been reported studying the extreme disorder limit.

For hopping, the EMA is thus **qualitatively** correct by predicting universality in the extreme disorder limit, but **quantitatively** inaccurate. This is perhaps not surprising, since universality at extreme disorder is due to the dominance of percolation effects \[22\] and the EMA is a mean-field theory for percolation (which is a critical phenomenon). The EMA replaces the disordered solid by an “effective” homogeneous solid with characteristics determined by a self-consistency condition. Such an ordered medium cannot \textit{a priori} be expected to accurately represent conduction along the optimal “percolation” paths of an extremely disordered medium \[24\].

An alternative to the EMA is the “percolation path approximation”(PPA) proposed for the macroscopic model \[21\]. According to the PPA, in the extreme disorder limit the AC conductivity is equal to that of a one-dimensional model with a sharp activation energy cut-off; this explains the existence of universality \[21\]. The idea is that percolation paths, which at extreme disorder dominate conduction in more than one dimension, have two characteristics: They are essentially one-dimensional and they only involve activation energies up to the “percolation energy”, \(E_c\), defined from the bond percolation threshold, \(p_c\), by

\[
p_c = \int_{-\infty}^{E_c} p(E) dE.
\]  
(2)

The percolation energy is the activation energy of the DC conductivity \[25,26\]. For the macroscopic model the PPA leads to \(\tilde{\sigma} = \tilde{s}/\ln(1 + \tilde{s})\), which is close to Eq. (1) and gives a good fit to simulations in more than one dimension \[21\].

The hopping version of PPA is not analytically solvable. Below we derive an approximation to the hopping PPA utilizing the **one-dimensional** EMA, which is known to work well \[27\]. Despite the above objections against the EMA, this procedure does make sense,
because in one dimension the “effective” homogeneous medium still is one-dimensional, of course. We show by computer simulations that the one-dimensional EMA, henceforth referred identified with the PPA, gives a better representation of low-temperature AC hopping in two and three dimensions than Eq. (1). Finally, we briefly discuss the consequences of these findings.

To arrive at the PPA, hopping in one dimension with a sharp energy barrier cut-off, \( p(E) = 0 \) for \( E > E_c \) while \( p(E_c) > 0 \), is addressed. In the “rationalized” unit system where the conductivity for a homogeneous system is equal to the jump rate \([14]\), the EMA equation for the AC conductivity \( \sigma(s) \) in one dimension \([18, 22, 27–32]\) is \([where s = i \omega \) is the Laplace frequency, \( \Gamma \) is the jump rate, and the brackets denote an average over the jump rate probability distribution\]

\[
\left\langle \frac{\Gamma - \sigma}{\sigma + (1 - s\tilde{G})(\Gamma - \sigma)} \right\rangle = 0. \tag{3}
\]

The quantity \( s\tilde{G} \) is \( s \) times the diagonal element of the Green’s function for a random walk on a one-dimensional lattice with uniform jump rate \( \sigma \) [the “effective medium”]; \( s\tilde{G} \) is given \([22, 27, 32]\) by

\[
s\tilde{G} = \left(1 + \frac{4\sigma}{s}\right)^{-1/2}. \tag{4}
\]

We are only concerned here with relatively low frequencies where \( s\tilde{G} \ll 1 \). To lowest order in \( s\tilde{G} \) Eq. (3) may be rewritten \([22]\)

\[
\frac{1}{\sigma} = \left\langle \frac{1}{\Gamma + s\tilde{G}\sigma} \right\rangle. \tag{5}
\]

The right hand side may be expanded as a power series in \( s\tilde{G}\sigma \), leading to

\[
\frac{1}{\sigma} = \sum_{n=0}^{\infty} (-s\tilde{G}\sigma)^n \left\langle \Gamma^{-(n+1)} \right\rangle. \tag{6}
\]

Since \( \Gamma = \Gamma_0 \exp(-\beta E) \), where \( \beta \) is the inverse temperature, the average \( \left\langle \Gamma^{-(n+1)} \right\rangle \) is easily evaluated in the low temperature limit: If \( \tilde{\beta} = \beta/p(E_c) \), one finds to leading order in \( 1/\tilde{\beta} \)

\[
\left\langle \Gamma^{-(n+1)} \right\rangle = \Gamma(E_c)^{-(n+1)}/[(n + 1)\tilde{\beta}].
\]

When this is substituted into Eq. (5) the following equation is obtained.
\[
\frac{1}{\sigma} = \frac{1}{\beta sG\sigma} \ln \left(1 + \frac{sG\sigma}{\Gamma(E_c)}\right).
\]

Letting \( s \) go to zero we find \( \sigma(0) = \tilde{\beta} \Gamma(E_c) \). Introducing the dimensionless Laplace frequency,

\[
\tilde{s} = \frac{\tilde{\beta}^2}{4 \sigma(0)} s,
\]

whenever \( sG << 1 \) Eq. (1) implies \( \tilde{\beta} s\tilde{G} = \sqrt{\tilde{s}/\tilde{\sigma}}, \) where \( \tilde{\sigma} = \sigma/\sigma(0) \). Substituting this and \( \Gamma(E_c) = \sigma(0)/\tilde{\beta} \) into Eq. (7) finally leads to the PPA expression for hopping,

\[
\sqrt{\tilde{\sigma}} \ln \left[1 + \sqrt{s\tilde{\sigma}}\right] = \sqrt{\tilde{s}}.
\]

Due to the factor \( \tilde{\beta}^2 \) in Eq. (8), as the temperature is lowered towards zero the condition \( sG << 1 \) is obeyed in a wider and wider range of frequencies around the onset of AC conduction.

We have carried out computer simulations of low-temperature AC hopping in one, two and three dimensions using the Fogelholm algorithm [33] to reduce the AC Miller-Abrahams electrical equivalent circuit of hopping [34–36] according to a recently proposed scheme [22]. To speed up the calculations the lowest jump rates were set to zero; it was carefully checked that this does not affect the conductivity. Figure one shows the results of computer simulations of low-temperature AC hopping in three dimensions. Results are shown for averages of 100 simulations of the AC conductivity at real Laplace frequencies for four different activation energy probability distributions at the following “reduced” inverse temperatures: (a) \( \tilde{\beta} = 80 \); (b) \( \tilde{\beta} = 160 \); and (c) \( \tilde{\beta} = 320 \). The full curve is the PPA (Eq. (1)) while the dashed curve is the EMA (Eq. (1)). Empirical rescalings of the data were allowed in order to focus only on the shape of the conductivity curves. Figure one shows that universality is approached as the temperature goes to zero and that the PPA gives a good fit to the universal AC conductivity in three dimensions.

The universality may be studied without use of empirical rescalings by plotting the slope of the Log-Log plot, \( d \log \tilde{\sigma}/d \log \tilde{s} \), as function of \( \tilde{\sigma} \). This is done in Fig. 2 for data from
computer simulations in one, two and three dimensions. The computer simulations in one dimension were carried out for systems with a sharp activation energy cut-off, to check the validity of Eq. (9). The full curve is the PPA prediction and the dashed curve is the EMA prediction in more than one dimension (Eq. (1)). Clearly, the PPA works better than the EMA in two and three dimensions; in fact, the PPA works very well in three dimensions.

As mentioned in the introduction, for the macroscopic model the EMA works very well in two and three dimensions [it is exact in one dimension]. For the macroscopic model the EMA universality prediction (Eq. (1)) is very close to the macroscopic PPA [21]. In view of the above presented results for hopping, it now appears that the EMA works well for the macroscopic model because the EMA happens to be close to the PPA, in contrast to what is the case for hopping. The two models have the common characteristic that, in the extreme disorder limit, conduction becomes essentially one-dimensional and that consequently the PPA gives a good description of the universal low-temperature AC conductivity for both models.

To summarize, we have presented evidence showing that the PPA, despite being based on a naive one-dimensional picture of percolation, is a good model for AC hopping conduction in the extreme disorder limit in two and three dimensions. These findings have important experimental consequences. The low-temperature universal conductivity of the macroscopic model is different from that of the hopping model. The macroscopic model incorporates Coulomb interactions via Maxwell’s equations, while hopping traditionally is concerned with non-interacting particles. Consequently, it is in principle possible to determine the relevance of Coulomb interactions by measuring the low-temperature AC conductivity.

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FIGURES

FIG. 1.

Log-Log plot (base 10) of computer simulations (symbols) of low-temperature AC conductivity in three dimensions at real Laplace frequencies for four different activation energy probability distributions, compared to the PPA (Eq. (3), full curve) and the EMA (Eq. (1), dashed curve). The dimensionless “reduced” Laplace frequency, $\tilde{s}$, is defined by Eq. (8) - however, an empirical rescaling was allowed to focus exclusively on the shape of the conductivity curve; $\tilde{\sigma}$ is defined by $\tilde{\sigma} = \sigma(s)/\sigma(0)$. The jump rates are $\Gamma = \Gamma_0 \exp(-\beta E)$, where the activation energy $E$ is chosen randomly according to the following probability distributions [21]: Asymmetric Gaussian [$p(E) \propto \exp(-E^2/2)$, $0 < E < \infty$] ($\times$); Cauchy [$p(E) \propto 1/(1+E^2)$, $0 < E < \infty$] (+); Exponential [$p(E) \propto \exp(-E)$, $0 < E < \infty$] ($\Diamond$); and Box [$p(E) = 1$, $0 < E < 1$] ($\Box$). To speed up the calculations all jump rates with activation energy larger than $E_c + 6.4/\beta$ were set to zero [$E_c$ is defined from the bond percolation threshold in Eq. (4)]. In terms of the dimensionless inverse temperature $\tilde{\beta} = \beta/p(E_c)$ the figure shows data for 100 averages of simulations of cubic lattices with sidelength $N$ where (a) $\tilde{\beta} = 80$ [$N = 29$]; (b) $\tilde{\beta} = 160$ [$N = 54$]; (c) $\tilde{\beta} = 320$ [$N = 100$].

FIG. 2.
The slope of the Log-Log plot of \( \tilde{\sigma}(\tilde{s}) \) at real Laplace frequencies, \( d \log_{10}\tilde{\sigma}/d \log_{10}\tilde{s} \), as function of \( \log_{10}\tilde{\sigma} \) for simulations (symbols, as in Fig. 1) at the inverse dimensionless temperature \( \tilde{\beta} = 320 \) in: (a) one dimension [100 averages of 8192 lattices]; (b) two dimensions [10 averages of 880X880 lattices]; (c) three dimensions [100 averages of 100X100X100 lattices] for the four activation energy probability distributions of Fig. 1. The simulations in one dimension were carried out with a sharp activation energy cut-off at \( E = 1 \) in order to show the validity of Eq. (9) in one dimension. In two and three dimensions, to speed up the calculations, all jump rates with activation energy larger than \( E_c + 6.4/\beta \) were set to zero. The simulations are compared to the predictions of the PPA (full curve, Eq. (9)) and the EMA (dashed curve, Eq. (10)). Both the EMA and the PPA predicts that the slope of the Log-Log plot goes to one as \( \tilde{s} \to \infty \), but the PPA works better than the EMA in two dimensions and much better than the EMA in three dimensions.
(b) 

\[ \log_{10}(\tilde{\sigma}) \sim \log_{10}(\tilde{s}) \]
$\log_{10}(\tilde{\sigma})$ vs. $\log_{10}(\tilde{s})$
\[
\frac{d \log(\tilde{\sigma})}{d \log(s)} \sim \log_{10}(\tilde{\sigma})
\]

(a)
\[
\frac{d \log_{10}(\bar{\sigma})}{d \log_{10}(\bar{s})}
\]
\[ \frac{d \log_{10}(\tilde{\sigma})}{d \log_{10}(\tilde{s})} \]