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ON THE PHYSICS OF A.C. HOPPING CONDUCTIVITY

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Abstract:

The physics of A.C. Hopping conductivity in disordered solids is discussed in the light of a recently proposed model.

In a recent paper (Dyre 1985) a simple model of a.c. hopping conductivity in disordered solids was proposed. It was shown that the model agrees qualitatively with experiments. This letter comments on the physics of the model and an electrical network analogue which reveals a one-dimensional aspect of the model, is suggested.

The proposed model applies to both ionic and electronic conductivity in amorphous solids. This is because the jump frequency distribution $p(\gamma)$ under reasonable assumptions of randomness in both cases is given by $p(\gamma) \propto \gamma^{-1}$ (Dyre 1985). Electronic and ionic conductivity will thus be similar in a Scher-Lax type of model (Scher and Lax 1973) where the disordered solid is represented by a lattice with jump frequencies varying randomly according to the distribution $p(\gamma)$.

In order to calculate the frequency-dependent conductivity, $\sigma(\omega)$, it is necessary to normalize $p(\gamma)$ by introducing two cut-off's: γ_{min} and γ_{max}. The low-frequency cut-off, γ_{min}, is clearly seen experimentally as defining the transistion from d.c. to a.c. conductivity. On the other hand, γ_{max}, is only seldomly observed as a leveling off of the conductivity at high frequencies, so γ_{max} should be eliminated from the model. Naively, $\sigma(\omega)$ diverges as $\gamma_{max} \to \infty$, but it is possible to renormalize the model in the same way as it is done in particle physics. As γ_{max} goes to infinity one finds for the renormalized conductivity in the CTRW approximation (Dyre 1985)

$$\sigma(\omega) = \sigma(0)\,\frac{i\omega\tau}{\ln(1+i\omega\tau)}\,, \qquad (1)$$

where ω is the angular frequency, and $\tau = \gamma_{min}^{-1}$. Equation

(1) predicts a universal frequency-dependence of $\sigma(\omega)$ (except for scale transformations), independent of temperature, chemical composition, and conductivity mechanism, in reasonably good agreement with experiments (Dyre 1985).

There are several ways to think of the renormalization: One may imagine that the density of charge carriers goes to zero while at the same time γ_{max} goes to infinity in such a way that $\sigma(0)$ is kept fixed, or it is possible simply to assume that γ_{max} is finite but <u>very</u> large (e. g. placed well within the region of phonon frequencies where a stochastic model breaks down anyway). In any case, the purpose of the renormalization is to eliminate any influence of the high-frequency cut-off. At this point it is interesting to compare with the pair-approximation (Mott and Davis 1979, Long 1982). Here one finds an approximate power-law frequency-dependence of the conductivity, $\sigma(\omega) \propto \omega^s$, where s is given by

$$s = 1 + \frac{4}{\ln(\omega \tau_{ph})} \qquad (2)$$

and τ_{ph} is of order 10^{-12} s. From eqn. (2) it is clear that the physics of the pair-approximation is a consequence of the physical <u>high-frequency</u> cut-off at τ_{ph}^{-1} : s is a function of the logarithmic "distance" from ω to τ_{ph}^{-1}. In the above model (eqn. (1)) , the exponent s of the a.c. conductivity at high frequencies is given by (Dyre 1985)

$$s = 1 - \frac{2}{\ln(\omega \tau)} \qquad (3)$$

and the physics is a consequence of the <u>low-frequency</u> cut-off at τ^{-1}. In particular, the model does not reduce to the pair-

approximation at high frequencies. This contradicts a theorem of Butcher and Morys (1973), but their result applies only to unrenormalized hopping models (i. e. with a fixed $p(\gamma)$).

The jump frequency distribution $p(\gamma) \propto \gamma^{-1}$ was originally based on microscopic considerations (randomly varying electron jump distances resp. ion jump activation energies), but it is possible to derive this distribution on purely phenomenological grounds. Suppose a hopping model is to be constructed which reproduces the following experimentally well-established facts: The shape of most dielectric loss-peaks (in log-log plots) is independent of temperature (universality) (Hill and Jonscher 1983, Long, Hogg, and Balkan 1983), and as the temperature goes to zero, the dielectric loss-peak frequency goes to zero and the exponent of the a.c. conductivity goes to one (Hill and Jonscher 1983, Long 1982). Then s must go to one as ω goes to infinity on the universal conductivity curve. At extremely large frequencies we thus have $\sigma(\omega) \propto \omega$. Since the conductivity has dimension of frequency and there can be no effects of γ_{min} on $\sigma(\omega)$ as $\omega \to \infty$, it can be concluded that $p(\gamma)$ contains no constant of dimension. But then $p(\gamma)$ must be proportional to γ^{-1} . Furthermore, the effect of γ_{min} at finite ω must be to reduce the frequency-dependence, i. e. to make s smaller than one. Because the jump frequency distribution gives equal weight to each decade of frequencies, one expects s to converge logarithmically to one as ω goes to infinity. Except for a factor of 2 these considerations lead to eqn. (3).

It is possible to construct an electrical equivalent-circuit of the model which sheds light on the physical assumptions involved. Consider an infinite network as shown in fig. 1. Each

capacitance is equal to a constant C, while the resistances R_n vary according to a probability distribution $p(R)$. The electrical properties of the network is determined by the average impedance per RC-unit, $Z(\omega)$, which is given by

$$Z(\omega) = \left\langle \frac{1}{R^{-1} + i\omega C} \right\rangle = \frac{1}{C} \int_0^\infty \frac{p(R)\, dR}{(RC)^{-1} + i\omega} . \quad (4)$$

Suppose each RC-unit somehow corresponds to an activated process in the solid. If the free energies of activation vary randomly, the characteristic time $t = RC$ is distributed according to $p(t) \propto t^{-1}$. Equation (4) then becomes

$$Z(\omega) = \frac{K}{C} \int_0^\tau \frac{1}{t^{-1} + i\omega} \frac{dt}{t} = \frac{K}{C} \int_0^\tau \frac{dt}{1 + i\omega t} , \quad (5)$$

where K is a constant, and τ is the maximum value of t, the existence of which follows if a finite average d.c. conductivity is required. Because $p(t) \propto t^{-1}$ is not normalizable, the constant K is unknown and must be determined selfconsistently. When this is done after the integration has been carried out, eqn. (5) reduces to eqn. (1).

The basic elements of the reticulation of the model are of course the CTRW approximation and the assumption $p(t) \propto t^{-1}$. While an electrical equivalent-circuit of the CTRW approximation usually involves a capacitance in parallel to the infinite network of fig. 1, this capacitance disappears as $\gamma_{max} \to \infty$ (Dyre 1985). The circuit of fig. 1 thus only applies to renormalized versions of the CTRW. Actually, the circuit is equal to that proposed by Macedo, Moynihan, and Bose (1972), thereby establishing a connection between their approach and the CTRW. Note that in our case, the distribution of resistances is loga-

rithmic just as the ordinary resistance scale, so it should be easy to build (a finite version of) the equivalent-circuit in the laboratory.

The circuit of fig. 1 is one-dimensional. How can hopping conductivity in three-dimensional solids be satisfactory represented by a one-dimensional model? The key to answering this question is the fact that very broad distributions of relaxation times (or jump frequencies) are involved. This means that the vast majority of links of the Scher-Lax lattice are only seldomly used for jumps. Therefore, the number of possible paths between two given sites of the lattice is effectively very much smaller than the corresponding number for a random walk on a homogenous lattice. The dimensionality is effectively reduced, and one ends up with a picture of conduction in disordered solids as mediated by certain "conducting paths".The solid is viewed as a spaghetti-like compound of conducting paths which cross each other here and there (like a polymer).

The above picture of the physics of hopping conductivity in disordered solids is closely related to the ideas of several other authors. The fact that most links are unlikely to promote jumps is the starting point of the pair-approximation (Pollak and Geballe 1961, Austin and Mott 1969). The concept of conducting paths was introduced by Nakajima (1972) in the context of ionic conductivity in oxide glasses, but the idea that certain paths are by far the most important goes back to Miller and Abrahams (1960). On this basis it seems a good idea to apply a suitable version of percolation theory to find the frequency-dependence of the conductivity. This was done by Böttger, Bryskin and Yashin (1979) who found

$$\sigma(\omega) \propto i\omega \left\{ \ln(i\omega\tau) \right\}^{-0,93} \qquad (6)$$

which is valid at moderately high frequencies in three dimensions. Notice the similarity between eqn. (1) and eqn. (6) when $\omega\tau >> 1$.

In conclusion, ideas of an effective low dimensionality for conduction in disordered solids have been discussed for several years. Equation (1) summarizes these ideas into a simple analytical formula, which gives a qualitatively satisfactory description of experiments (Dyre 1985).

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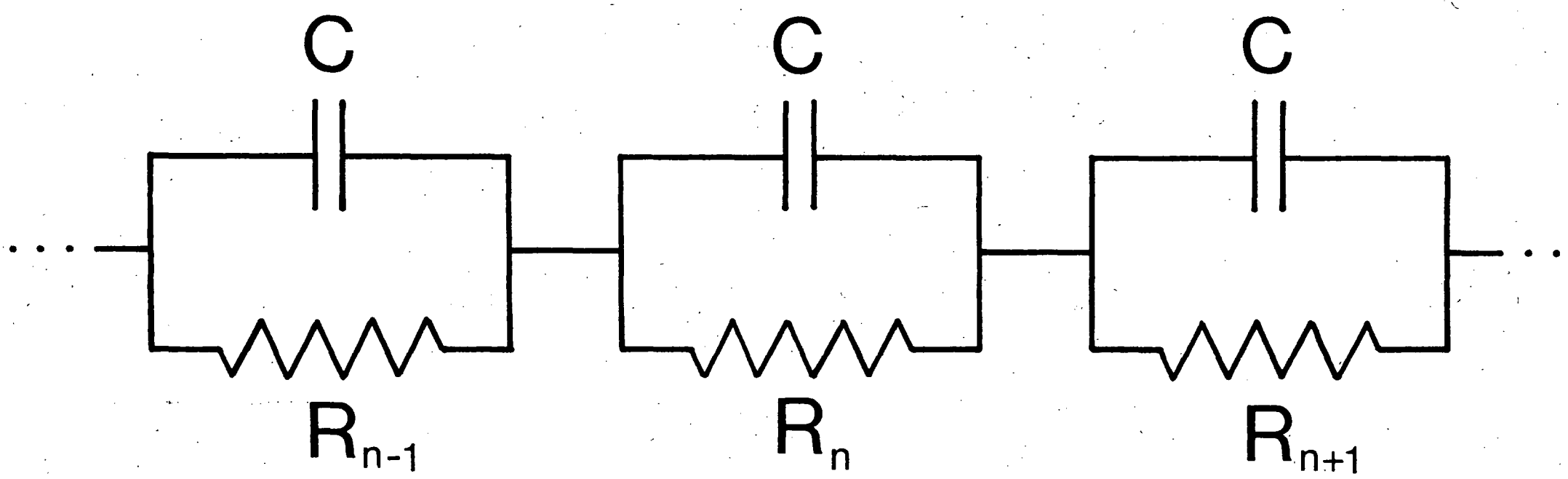
Figure Caption

Fig. 1: Electrical equivalent-circuit of the renormalized CTRW approximation.

FIG. 1



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