Relationship between the conductivity and the glass temperature for hopping systems

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A relationship between the glass temperature and the steady-state conductivity is derived for hopping-conduction systems generally. The glass temperature is given simply in terms of experimentally obtainable quantities. The steady-state conductivity just above the glass temperature depends only on the density of states and the relaxation time to steady state.

The problem of hopping conductivity in disordered systems is generally treated within the framework of percolation theory. Hopping rates u between initial and final states i and f are written, in general,

$$u_{if} = u_0 \exp(-\xi_{if}) = u_0 \exp(-2r_{if}/a - \Delta_{if}/kT)$$
 (1)

with r_{if} a hopping distance, Δ_{if} an energy, a the localization radius, u_0 a constant frequency related to the socalled phonon frequency, and kT the Boltzmann constant times the temperature T. The applicability of this relationship is not restricted to systems in which the intersite Coulomb interaction can be neglected, such as amorphous semiconductors, but it may be applied to systems where this interaction is demonstrably important, such as impurity conduction systems, as well. When successive correlations are important a more sophisticated treatment is necessary.¹ In the former case $u_0 = v_{ph}$, the phonon frequency. As is discussed in the book by Mott and Davis² this phonon frequency may often be taken as the Debye frequency. This frequency varies from material to material but it has been assumed for over a decade to lie between 10^{12} and 10^{13} Hz. In the latter case u is given as a product of the phonon frequency and a constant factor γ dependent on disorder energy, Coulomb interaction energy, and the resonant energy. The applicability of percolation theories requires a very wide distribution of the values of the u_{if} , provided for by the exponential dependence of the u_{if} on the random variables r_{if} and Δ_{if} .

The dc conductivity is then usually written

$$\sigma = \sigma_0 \exp(-\xi_m) = \sigma_0 \exp[-f(T_0/T)] . \tag{2}$$

Here the exponent ξ_m corresponds to optimal percolation.³ Critical percolation occurs at $\xi_c = \xi_m - 2\nu$. The reason for the distinction between critical and optimal percolation is discussed in detail by Pollak³ and is related to the areal density of current paths. At critical percolation this density vanishes as the inverse square of the correlation length, $(\xi_m - \xi_c)^{2\nu}$. The constant $\nu = 0.9$ gives the critical behavior of the correlation length. Values for ν given in the literature⁴ are in the range $0.7 < \nu < 0.9$. The value ξ_c then corresponds to the slowest transition $[u_m = u_0 \exp(-\xi_c)]$ on the critical path. The function $f(T_0/T)$ often has the form of a power law,

$$f(T_0/T) = (T_0/T)^p, \quad 0.25 (3)$$

A plot of $\ln \sigma$ versus T^{-p} yields in this case a straight line with slope $-T_0^p$. It is very important to note here that although results of percolation theories for interacting systems are still not generally accepted, the experimental form for the relationship $f(T_0/T)$ still seem to be given quite generally by Eq. (3).

Recently some authors 1,5,6 have asked the question of what happens when the slowest rate, u_m , on the percolation path is smaller than the inverse of the experimental time t. It is evident that in this case one cannot measure a steady-state dc conductivity. This situation occurs below a finite temperature T_e . Calculations of T_e , called the glass temperature, have been obtained for both interacting and noninteracting systems, and T_e has in each case been shown to depend weakly on the experimental time through the factor $\ln(tu_0)$. It is the purpose of this note to exploit the relationship between transport and equilibrium properties to develop a general relationship between T_0 and T_e . This can be used to determine T_e even when T_0 can only be determined experimentally. This fact is particularly useful as the result derived here may be used even in interacting systems where the percolation theories available at this time do not give unambiguous results for f. It should be noted that the problem of nonergodic behavior in hopping systems with intersite Coulomb interactions has been also treated by some other authors.^{7,8} In their investigation the applicability of the ergodic hypothesis to the "electron glass" was considered in the limit of infinite time. Their approach has been to investigate the behavior of an order parameter (corresponding to the modified Edwards-Anderson order parameter in spin glasses). They have found strong evidence for a phase transition to a glassy state at a finite temperature. In the treatment discussed here, however, we consider the applicability of the ergodic hypothesis for experimentally feasible times and it proves unnecessary to invoke the Coulomb interaction in order to produce nonergodic behavior. Thus a temperature is determined which describes a transition to glassy behavior even for noninteracting systems. For this reason it seems evident that the glass temperature as calculated here will also for interacting systems be significantly higher than that calculated rigorously in the

<u>39</u> 11 154

thermodynamic limit. The work of these authors depends on the applicability of the ergodic hypothesis, but even at temperatures above the glass temperature which they calculate it is likely that an infeasibly long time will be required for the approach to thermodynamic equilibrium.

To calculate T_e we first write ξ_m as

$$\xi_m = f(T_0/T) \ . \tag{4}$$

For the condition $T = T_e$ we have

$$t^{-1} = u_m = u_0 \exp(-\xi_c)$$
 (5)

or

$$\xi_c = \ln(tu_0) \ . \tag{6}$$

In this case we also have

$$\xi_m = f(T_0 / T_e) . (7)$$

Thus

$$2\sigma + \ln(tu_0) = f(T_0/T_e) \tag{8}$$

or

$$T_e = T_0 / f^{-1} (\ln(tu_0) + 2\nu) = T_0 / [\ln(tu_0) + 2\nu]^{1/p} , \qquad (9)$$

the latter equality following if $f(x) = x^p$. This result gives the glass temperature T_e purely in terms of experimental quantities, p and T_0 . Even if it is not possible to represent f(x) as a power law it is possible to make a prediction of the glass temperature. Using this we have, for $\sigma(T_e)$,

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$$\sigma(T_e) = \sigma_0 \exp[-f(T_0/T_e)]$$

= $\sigma_0 e^{-2\nu}/tu_0$
= $7[N(E_f)]^{0.58} a^{0.74} (kT)^{-0.42} e^{-2\nu}/tu_0$. (10)

The last equality uses a result for σ_0 without interactions from Pollak.³ It can be seen that $\sigma_0 E e^{-2\nu} / t_c u_0$ is the value of the steady-state current at optimal percolation, which occurs at a time t_c after turning on an electric field *E*. For earlier times a calculation of the time dependence of the polarization current has yielded⁹

$$j(t) \sim (t)^{-1} [\ln(t_c u_0)]^{-2.3}$$
 (11)

(see Ref. 6 for calculation which would lead to another exponent on the ln factor). The time dependence of the current due to the evolution of the percolation cluster itself can be shown to be given by

$$j(t) \sim (t)^{-1} [\ln(tu_0)]^2$$
 (12)

The results for the time-dependent current and their implications on experiment near the onset of percolation as well as on the ac conductivity will be considered in depth in a forthcoming paper.¹⁰ We mention them here, however, in order to describe roughly the behavior of the dc response near the onset of percolation.

If σ_0 is obtained from a plot of σ versus T it is then possible (using $u \sim 10^{13}$ Hz) to obtain the critical value of t from Eq. (10). Conversely, an observation of the time required for percolation yields, with the aid of Eq. (10), information on the density of states.

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¹⁰A. Hunt (unpublished).