

The low frequency conductivity of the Fermi glass

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1992 J. Phys.: Condens. Matter 4 6957

(<http://iopscience.iop.org/0953-8984/4/33/009>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.96

The article was downloaded on 11/05/2010 at 00:25

Please note that [terms and conditions apply](#).

The low-frequency conductivity of the Fermi glass

A Hunt

Earth Sciences Department, University of California, Riverside, CA 92521, USA

Received 11 July 1991, in final form 16 April 1992

Abstract. The low-frequency (below the loss peak, or critical frequency, ω_c) conductivity, $\sigma(\omega)$, of the Fermi glass is evaluated in an approximation replacing cluster currents with chain currents. Long chains with relatively low resistance values but large relaxation times make an important contribution to the polarization current density at low frequencies leading to a non-analytical frequency dependence on ω of $\sigma(\omega)$ in the limit $\omega \rightarrow 0$. The statistics of such chains are derived from the cluster statistics near percolation, while the AC currents of such chains are taken from an existing model for conduction on polymers.

1. Introduction

The Fermi glass refers to disordered systems where the dominant transport is by electronic 'hopping' (phonon-assisted tunnelling) between localized states, and at temperatures high enough that inter-site Coulomb interactions can be neglected. In linear response theory, $\sigma(\omega)$ is calculated from a random network of resistors R_{ij} (Miller and Abrahams 1960) connected between each pair of sites i, j and capacitors C_i (Pollak 1974) from each site i to a generator of the potential $F_0 \cos \omega t \cdot r_i$. F_0 is the external field, ω its frequency, and r_i the position of site i . Expressions for R_{ij} and C_i are

$$R_{ij}^{-1} = (e^2/kT)w_{ij}f_i(1-f_j) = (e^2\nu_{\text{ph}}/kT)\exp[(2r_{ij}/a) + (\Delta_{ij}/kT)]$$
$$C_i = (e^2/kT)f_i(1-f_i) \approx (e^2/kT)\exp(-|E_i|/kT). \quad (1.1)$$

The distributions of R s and C s depend exponentially (through the Fermi function $f \approx \exp(-|E_i|/kT)$, with the Fermi energy, $E_F \equiv 0$, and the quantum mechanical transition rates $w_{ij} = \nu_{\text{ph}} \exp[(2r_{ij}/a) + (E_{ij}/kT)]$) on the distributions of the site energies, E_i , and separations, r_{ij} . Here ν_{ph} is a fundamental rate constant, about 10^{12} Hz, e the electronic charge, a the localization length, E_{ij} the larger of $E_j - E_i$ and 0, and $\Delta_{ij} \approx |E_i| + |E_j| + |E_j - E_i|$. A constant density of states $N(E_F)$ near E_F is assumed; positions and energies of electronic states are assumed uncorrelated. Nearest neighbour statistics are Poisson. These assumptions are usual, and lead to Mott (1969) variable-range hopping (VRH) for the DC conductivity, $\ln \sigma_{\text{DC}} \propto -(T_0/T)^{1/(d+1)}$ with $kT_0 \propto [N(E_F)a^d]^{-1}$ and d the dimensionality. VRH is (e.g. Pollak 1972) equivalent to percolation theoretical calculations.

At high frequencies ω , pairs of sites connected to large capacitors C_i and C_j , and connected by resistances R_{ij} with $\tau_{ij} = R_{ij}C_iC_j/(C_i + C_j) = 1/\omega$, dominate

$\sigma(\omega)$ (Austin and Mott 1969, Pollak and Geballe 1961) and decouple from the network. $\sigma(\omega)$ determined from the distribution of these pairs of sites is known as the pair approximation and yields $\sigma(\omega) \propto \omega^s$ with s approximately 0.8. A new treatment with energies E (Hunt 1991a) of relevant sites determined by optimization (as a function of frequency) yields $\sigma(\omega)$ in agreement with experiment for $\omega > \omega_c$ (the loss peak frequency). At ω_c included-pair processes (for which $\tau \leq \omega_c^{-1}$) percolate, as typical pair lengths and separations scale as $a(T_0/T)^{1/(d+1)}$. And at ω_c $|E| < 0.2kT(T_0/T)^{1/4}$ (in three dimensions) as in percolation theory (Friedman and Pollak 1978).

The DC current flows in paths defined by resistors $R \leq R_{\text{opt}} = R_c e^{2\nu}$ (Friedman and Pollak 1978) with R_c the critical resistance and ν the critical exponent of the correlation length. The representation (discussed below)

$$\sigma_{\text{DC}} = l e^{-2\nu} / L^2 R_c \quad (1.2)$$

with L the correlation length of the DC cluster (at $R = R_c e^{2\nu}$), and l the typical separation of maximal resistors on the percolation path, is based on optimization with respect to R_{opt} of a homogeneous network with L and l derived in terms of R_{opt} near percolation. Though a homogeneous network is sufficient to calculate σ_{DC} , large regions of lower resistance induce polarization currents contributing to $\sigma(\omega)$ at low ω (since they need not connect the electrodes). The possibility of significant further charge transport between pairs (below ω_c) was suggested by Long *et al* (1988). Although the pre-exponential of (1.2) is not compatible with experiment in a-Si (Long *et al* 1988), it is so (Long and Hansmann 1990) in a-Si:H:Au; the result implies that a-Si is special, i.e. the resulting criticism of percolation theory is inappropriate.

It is not intended here to determine how to modify the model to bring σ_{DC} into agreement with experiment (in a-Si) but rather to determine the low ω response of the Fermi glass using the simplest model assumptions. (Although the results have already been published in Hunt (1991a) the details for low ω were referred to this work.) Such a theory should apply to many systems. Indeed the T dependences of $\sigma(\omega)$ in a-Si:H:Au also agrees (Hunt 1991a) with this theory (equation (3.18)). Moreover (Hunt 1991a) close agreement with experiment for $\sigma(\omega)/\sigma_{\text{DC}}$ results in a-Si.

Specific theoretical advances resulting from this theory are that (i) it is the first to describe cluster-length dependent enhancements of relaxation times of clusters of impedances (compared with the relaxation time of the slowest element, $\tau_c \approx \omega_c^{-1}$), (ii) it is the first to show that the dominant effects on $\text{Re } \sigma(\omega)$ arise from processes with $\tau \approx 1/\omega$ at all frequencies, and (iii) as a consequence of (i) and (ii) it shows that the effects of critical exponents of percolation theory on the exponent of ω in $\sigma(\omega)$ is displaced from ω_c to 0, where relaxations extend to infinite size. Non-integral powers to ω are generated on both sides of ω_c without denying the relevance of wide distributions of relaxation times to the power s in $\sigma(\omega) \propto \omega^s$. Divorcing s (which is system- as well as temperature-dependent) from percolation arguments (Niklasson 1987) involving the fractal dimensionality, d_f , also keeps d_f independent of T allowing its meaningful experimental determination.

The above advances in understanding rest on the following improvements in agreement between theory and experiment. First $\sigma(\omega)$ at low ω in VRH systems as determined by experiment (in a-Si (Long *et al* 1988), in a-Si:H:Au (Long and Hansmann 1990) and in polyacetylene (Summerfield and Chroboczek 1985)) is always enhanced

over quadratic in ω (from a maximum relaxation time τ_c) predicted in other cluster theories (e.g. Boettger and Bryksin 1985) and in effective medium theories (Summerfield and Butcher 1982, Movaghar *et al* 1980). This is rectified by the sub-quadratic dependence predicted here; moreover in a-Si the power of the low-frequency conductivity has been demonstrated explicitly to be in accord with this theory. Second the placement of ω_c is in agreement with experiment. Third, as noted, the temperature dependence of $\sigma(\omega)/\sigma_{DC}$ is brought into much closer agreement with experiment. Finally, the numerical pre-factor of $\sigma(\omega)/\sigma_{DC}$ is within a factor of three of the experimentally observed value.

2. Temperature-dependent cluster statistics

First, results for temperature-dependent lengths and for scaling variables of cluster statistics are found. Since Summerfield (1985) and Long *et al* (1988) discuss the ratio $\sigma(\omega)/\sigma_{DC}$, the pre-exponential σ_0 of σ_{DC} is found also. Besides, σ_0 depends on lengths relevant for calculating $\sigma(\omega)$, for which published values are controversial.

The impedance network contains elements whose values are exponential functions of random variables with enormous spreads. We replace the continuous distribution of resistance values by a discrete one. All R_{ij} with $e^{-1/2}R < R_{ij} < e^{1/2}R$, $e = 2.718\dots$, are assigned the value R , and R takes on discrete values of powers of e . Emplacement in the network of R s with $R \leq R_m$ (R_m arbitrary) connects isolated pairs when R_m is small. Increasing R_m connects more pairs until pairs coalesce into clusters. $R_m \equiv R_c$ when the largest cluster just reaches infinite size. If R_m is not too different from R_c , cluster size distributions are given in terms of critical exponents.

The temperature dependence of σ_{DC} arises chiefly through R_c . The current avoids resistors with $R > R_c$, and resistors with $R < R_c$ can be replaced by shorts. If σ_{DC} were defined by $R_m = R_c$, the infinite correlation length χ (at critical percolation) would require infinite separation of current-carrying paths and vanishing σ_{DC} . So σ_{DC} is calculated from $R_m > R_c$; the resulting resistance of the relevant paths is increased, but beyond critical percolation their density is non-zero. Since $\sigma_{DC} \propto l/R_m \chi^{d-1}$, with l the typical separation of resistors (with $R = R_m$), and $\chi = \chi(R_m)$, this quotient can be optimized. The subscript m is now dropped. From percolation theory (Stauffer 1979)

$$\chi = \chi_0 |p_c - p|^{-\nu} \tag{2.1}$$

with χ_0 a number and p_c the critical value of the site (or bond) probability p . p_c relates to R_c , p to R . The percolation condition for constant $N(E_F)$ is

$$\xi_c^{d+1}(T/T_0) = \alpha_c. \tag{2.2}$$

$\alpha_c \approx 2.7$ in 3D and $R_c = R_0 \exp \xi_c$. Assume $p_c \propto \alpha_c$, $p \propto \alpha$, so $\xi^{d+1}(T/T_0) = \alpha$,

$$|p_c - p| \propto |\xi_c^{d+1} - \xi^{d+1}| T/T_0 \simeq \xi_c^d |\xi_c - \xi| T/T_0 = |\ln(R_c/R)| (T/T_0)^{1/(d+1)}. \tag{2.3}$$

Using (2.3) in (2.1)

$$\chi = \chi_0 (T_0/T)^{\nu/(d+1)} |\ln(R_c/R)|^{-\nu} \tag{2.4}$$

assuming unit bond length. But the average bond length in VRH is also temperature-dependent, $r_{\text{VRH}} \approx a(T_0/T)^{1/(d+1)}$, so

$$\chi = \chi_0 a (T_0/T)^{(1+\nu)/(d+1)} |\ln(R_c/R)|^{-\nu}. \quad (2.5)$$

χ_0 , assumed of order 1, is dropped. The factor l , slowly varying in R (justified later), is neglected in optimization. Substituting (2.5) into (1.1) and optimizing gives

$$R_{\text{opt}} = R_c e^{(d-1)\nu} \quad (2.6)$$

and for the correlation length of the DC cluster

$$\chi(R_{\text{opt}}) \equiv L = a [e/(d-1)\nu]^{(d-1)\nu} (T_0/T)^{(1+\nu)/(1+d)}. \quad (2.7)$$

$\chi(R_{\text{opt}})$ defines the characteristic distance between branches of the DC cluster, and is given the symbol L . Note that this value for L is the same as in Reikh and Ruzhin (1991), and nearly identical to that of Friedman and Pollak (1979). σ_{DC} is (dropping numerical factors)

$$\sigma_{\text{DC}} = l/R_{\text{opt}} L^{d-1} \approx l/R_c L^{d-1}. \quad (2.8)$$

Consider now l . We assume it to be the average separation of maximal resistors on the finite clusters also. We assume that the ratio of the number of maximally valued resistances to the number of all smaller resistances is the same on clusters as in the bulk (because clusters are formed by random selections of resistors; although some maximal R s may be shorted on large clusters, it turns out to be unnecessary to take this complication into account). This ratio scales as (Apsley and Hughes 1975)

$$a r_{\text{VRH}}^d / r_{\text{VRH}}^{d+1} = a / r_{\text{VRH}} = (T/T_0)^{1/(d+1)}. \quad (2.9)$$

Resistances with $\xi \leq \xi_c$ fill a $(d+1)$ -dimensional space to length r_{VRH} ; those with $\xi \approx \xi_c$ are located on the surface of this volume. The linear dimension, r_s , of a cluster of s elements (volume $\propto s$) of unit length is

$$r_s = s^{\sigma\nu} h(z) = s^{1/d_t} h(z) \quad (2.10)$$

with h some function of $z \equiv |p_c - p|s^\sigma$, and with the product, $\sigma\nu$, equal to the inverse of the fractal dimensionality, d_t , of the percolation cluster (Stauffer 1979). Since individual bonds have $r_{\text{VRH}} = a(T_0/T)^{1/(d+1)}$

$$r_s = a(T_0/T)^{1/(d+1)} s^{\sigma\nu} h(z). \quad (2.11)$$

Selectively severing bonds perpendicular to F converts clusters to bundles of chains. r_s is defined as Nl with N the number of maximal R s on the longest chain. Then

$$Nl = a(T_0/T)^{1/(d+1)} s^{\sigma\nu}. \quad (2.12)$$

But since $N^{1/\sigma\nu} = N^{d_t}$ is the total number of maximal R s on the cluster

$$(N^{1/\sigma\nu})/s = (T/T_0)^{1/(d+1)}. \tag{2.13}$$

Simultaneous solution of (2.11)–(2.13) yields

$$l = a(T_0/T)^{(1+\sigma\nu)/(1+d)}. \tag{2.14}$$

l^{-d} is $a^{-d}(T/T_0)^{(d+d/d_t)/d(d+1)}$, which, if $d = d_t$, is the typical separation of maximal R s in the bulk. Even so, 0.35 is obtained (with $d_t = 2.5$) for the exponent instead of 0.33 ($= \frac{1}{3}$). This is reasonable as association of resistances through a fractal network should not alter their effective separations greatly.

The density of s -clusters (unit bond length) is (Stauffer 1979)

$$n_s = K(d)s^{-\tau} f(z) \tag{2.15}$$

where $K(d)$ a dimensionally dependent numerical factor and $z = |p_c - p|s^\sigma$ again. $f(z)$ is approximated by $\exp(-z)$ or $\exp(-z^2)$ (the latter expression valid on both sides of p_c), and involves a sharp cut-off of cluster sizes except at $p = p_c$. Clusters with bond lengths proportional to r_{VRH} have volumes proportional to $a^d(T_0/T)^{d/(d+1)}$, so

$$n_s(T) = a^{-d}(T_0/T)^{-d/(d+1)}s^{-\tau} f(z). \tag{2.16}$$

The distribution of maximum chain lengths, $N = r_s/l$, is derived using $n_N dN = n_s ds$ and assuming (Hunt and Pollak 1990) that only one chain of length N with a given orientation exists on any cluster. Then

$$n_N = K(d)N^{-(d+1)}l^{-d} f(z) = 1.6N^{-4}l^{-3} \exp\{-[(Nl/L)^{1/\nu} \ln R_c/R]^2\} \tag{2.17}$$

after applying (Stauffer 1979) $(\tau - 1 + \sigma\nu)/\sigma\nu = d + 1$. Some poorly known numerical constants are suppressed. The latter equality is for 3D.

Note that (2.11) for $r_s(T)$ and (2.16) for $n_s(T)$ are consistent with (2.5) for $\chi(T)$ according to (Stauffer 1979)

$$\chi = \int r_s s^k n_s ds / \int s^k n_s ds \tag{2.18}$$

for $k > 1$ with $z = |p_c - p|s^\sigma$, unchanged by introducing the parameter T .

For r -percolation, L , l , and χ are all independent of T .

3. Calculations of $\sigma(\omega)$ at low frequencies

3.1. General d -dimensionality calculations

$\sigma(\omega)$ can be expressed (justified in appendix A) as a sum of σ_{DC} and a polarization current contribution, $\sigma_p(\omega)$

$$\sigma_p(\omega) = \sum I_{\text{cluster}}(\omega)/\Omega F_0 = \sum n_s I_s(\omega)/F_0 \tag{3.1}$$

with Ω the volume of the system, and $I_{\text{cluster}}(I_s)$ the polarization current in an s -cluster. This was also found (Pollak 1971) for $\sigma(\omega)$ in the pair-approximation regime using a dielectric representation. The complexity of large clusters requires simplification. That long chains of resistances with similar R have strongly enhanced τ s and large cluster currents suggests that backbone chains of clusters (with $R \leq R_c$) should contribute most to $\sigma_p(\omega)$ for $\omega < \omega_c$. The following approximations are applied.

- (A) Cut selected maximal R s oriented perpendicular to F
- (B) Replace all resistances $R_{ij} < Re^{-1/2}$ by shorts
- (C) Merge capacitors joined by shorts into single capacitors $C = \sum_i C_i$
- (D) Replace all C s with their average value
- (E) Make separations of resistors equal to their average value l
- (F) Treat the remaining elements as an untwisted polymer
- (G) Treat only the longest chain in a given cluster.

A has been used for the DC cluster, and is certainly appropriate for small clusters (with a single maximal R). B assumes the chief contribution of sites connected by very small resistors to be charge generation. Since their contributions are parallel, the capacitances add, justifying C. D is justified since the average number, n_c , of shorted R s per maximal R is $(T_0/T)^{1/4} \approx 30$ at typical (measured) T , making fluctuations in n_c small; $30^{-1/2} \approx 20\%$. E is in keeping with the result that the length of the longest chains of large clusters is most important, not irregularities in the positions of individual elements along the chain. Ignoring the tortuosity (F) may seem risky, but it is argued that it at most changes values of frequency-dependent exponents by 3%; these exponents describe charge transport on long chains and the number of such chains per unit volume. G, adequate for $\text{Re } \sigma(\omega)$ (see (3.4) and following discussion), is inadequate for direct calculation of $\text{Im } \sigma(\omega)$; $\text{Im } \sigma(\omega)$ is best determined from $\text{Re } \sigma(\omega)$ by Kramers-Kronig relations.

At ω for which the longest chain contributes most to $\text{Re } \sigma(\omega)$ the remaining chains are much shorter (with smaller I_N and much smaller τ) and contribute mainly to $\text{Im } \sigma(\omega)$. While smaller chains may be important in screening longer chains, this effect is ignored here (because of its complexity and because such screening effects are of minimal importance at large ω and at $\omega = 0$).

The average capacitance per maximal R is evaluated at critical percolation; the variation of C with R is minimal if R is not too different from R_c . This value is the product of the average capacitance of sites within $kT(T_0/T)^{1/(d+1)}$ of E_F , and the average number of such sites connected by shorts to resistors with R_c . The latter is given by the reciprocal of (2.13), the former is

$$\langle C \rangle = \int N(E_F)(e^2/kT) \exp(-E/kT) dE \times \left(\int N(E_F) dE \approx (e^2/kT)(T/T_0)^{1/(d+1)} \right)^{-1} \quad (3.2)$$

so that

$$C = n_c \langle C \rangle = e^2/kT. \quad (3.3)$$

The characteristic R of a chain is *arbitrary*! Its polarization current (induced by an external field) is (Pollak and Pohl 1975)

$$\text{Re } I_N/F \approx (2/3\pi^2)(Nl^2/R)[\omega^2\tau_N^2/(1 + \omega^2\tau_N^2)] \quad \tau_N = N^2RC/\pi^2. \quad (3.4)$$

The exact result is a sum over modes, but $\text{Re } I_N$ is given to within 20% by (3.4). Note that I_N has the form of a pair approximation with $R \rightarrow NR$, $C \rightarrow NC$, and

number of chains on a given cluster is taken to justify treating only the longest chain in the cluster. Further implications are discussed in Hunt (1991b). Converting the sum over cluster sizes to an integral over chain lengths and substituting (3.4) and (2.17) into (3.1) yields

$$\sigma_p(\omega) = (3.2/3\pi^2) \int (dN/N^{d+1}) \exp\{-[Nl/L(\ln R_c/R)^{1\nu}]\}(Nl^2/R) \times (\omega^2 R^2 C^2 N^4/\pi^4)/[1 + (\omega^2 R^2 C^2 N^4/\pi^4)]. \tag{3.5}$$

A common practice is to treat R as a parameter and optimize $\sigma_p(R, \omega)$. This ‘finds’ the dominant relaxation units at ω . Such a procedure (Hunt 1990) leads here to $R = R_c$. The physical reason is the exponential cut-off in n_N (eliminated when $R = R_c$ i.e. the cluster sizes decay according to a simple power law at critical percolation) and that as $\omega \rightarrow 0$ the relaxation time of the dominant process diverges, i.e. $N \rightarrow \infty$. What is surprising is that $R = R_c$ holds also for finite ω . Note, however, that the simple condition $\omega\tau \approx 1$ leads to

$$N^2 RC/\pi^2 = 1 \tag{3.6}$$

which implies

$$N = (\pi^2/\omega RC)^{1/2} \tag{3.7}$$

so that

$$I_N \propto (1/2)Nl^2/R \tag{3.8}$$

and selection of longer chains with smaller R s enhances I_N in both respects. This argument is related to (in 3D)

$$\sigma_p(\omega) \propto (1/N^2 R)(\omega^2 R^2 C^2 N^4/\pi^4)F(z)/[1 + (\omega^2 R^2 C^2 N^4/\pi^4)] \tag{3.9}$$

where the integral (over only those N for which $\tau_N\omega \approx 1$)

$$\int dN f(z)/N^3 \equiv F(z)/N^2 \tag{3.10}$$

and $F(z)$ is known only through the definition (3.10), but contains the same nature exponential cut-off. The maximum of $\sigma_p(\omega)/F(z)$ clearly occurs at $N^2 R = 1/\omega C\pi^2$ (by analogy to the pair approximation i.e. $R \rightarrow N^2 R/\pi^2$). However, pair distributions are slowly varying functions of R , whereas $F(z)$ is not. The aim of this paragraph is, however, not a direct calculation of $\sigma_p(\omega)$, but a suggestion for the decomposition of clusters. The factor mitigating against merely choosing (3.8) is the number of such chains. Chains with large N and small R (significantly less than R_c) are very rare. The particular decomposition of the clusters is very important.

It is clear that one can sum clusters of all sizes for any arbitrary R . One might ask if it is legitimate to sum over R also. This method is unusual, but not inconsistent.

By (3.6) one has that for a given frequency, long chains of small resistors and/or short chains of large resistors contribute to $\sigma_p(\omega)$. Since such clusters are mutually exclusive, no redundancy in counting results from a double sum. It will turn out that for $\omega \approx \omega_c$ the result of such a double sum (approximated as a double integral) is larger than the contribution from $R = R_c$, while an approximation including only $R = R_c$ is larger when $\omega \ll \omega_c$. An important question is why a sum over R is preferable to, say, a sum over smaller chains on clusters with $R = R_c$. First note that in clusters with $R < R_c$ contributions from smaller chains are also neglected. Besides, in clusters with one maximal R , only the longest chain is relevant to $\text{Re } \sigma(\omega)$, and in the limit $\omega \rightarrow 0$, $\sigma(\omega)$ is also calculated from the backbone cluster. Thus, the treatment is correct in these two limits (though this does not necessarily imply accuracy in the limits $\omega \rightarrow 0$ and $\omega \rightarrow \omega_c$). Also chains with only one R have already been accounted for in the pair approximation regime. Perhaps the most important theoretical argument for neglect of shorter chains is that a thermodynamically motivated (Hunt 1992) method to determine a long time limit of cluster charge transport yields the same length dependence for a dominant cluster current and relaxation time as in the longest chain. However, unlike for chains, a quantitative discussion of charge transport due to all modes is unavailable. Nevertheless, the method chosen gives agreement with experiment, and is appropriate for and yields the correct result in 1D systems.

The problem of summing a series with (as it turns out) from 1–10 meaningful terms (depending on the frequency) is non-trivial. The method here is to treat the first term separately, i.e. $R = R_c$, and to sum the remainder by integration. In the integration $R = R_c$ ($j = 0$ below) is excluded; to perform the integral analytically requires, however, the extension of the lower limit on j to 0. This seems inconsistent, but in the limit $\omega \rightarrow 0$ where $R = R_c$ dominates, the integral is insignificant, while near ω_c , where the integral dominates, the term with $R = R_c$ is insignificant. However, changing the lower limit of integration introduces corrections in the frequency dependence at higher powers of ω (which become important by $\pi^2\omega_c$, and which, together with the neglected quadratic terms in the integral over N for R_c cause the sum over cluster currents to start to decrease above $\pi^2\omega_c$, thereby generating the loss peak).

The terms with $R = R_c$ are defined as $(3.2l^{2-d}/3\pi^2) \sum_N I_{N,R_c}$ with

$$\begin{aligned} \sum_N I_{N,R_c} &= \int_1^\infty (dN/N^{d+1} R_c) [\omega^2 R_c^2 C^2 N^4 / \pi^4 / (1 + \omega^2 R_c^2 C^2 N^4 / \pi^4)] \\ &= (\omega C / 2\pi^2) [\pi/2 - \tan^{-1}(\omega / \pi^2 \omega_c)] \quad (d = 3) \end{aligned} \quad (3.11)$$

and extension of the lower limit of integration in I_{N,R_c} from 1 to 0 omits the \tan^{-1} corrections of order ω^2 . For integration over R define

$$R \equiv R_c \exp -j \quad \ln R_c / R = j \quad (3.12)$$

and the sum over j can be approximated directly as ($\Delta j = 1$)

$$\sum_{N,R} = (3.2/3\pi^2 l) I_{N,R}$$

$$I_{N,R} = \int_1^\infty \int_1^\infty dj dN N^{-3} \exp[-[j(Nl/L)^{1/\nu}]^2](\omega/\omega_c)^2(N/\pi)^4 \times \exp -j/[1 + (\omega/\omega_c)^2(N/\pi)^4 \exp -2j] \quad (3.13)$$

where $\omega_c \equiv \nu_{ph} \exp[-(T_0/T)^{1/(d+1)}]$. Clusters with $j \gg 1$; $N \gg 1$ are rare. But

$$(l/L)^2 = (T/T_0)^{2(\nu-\sigma\nu)/(d+1)} = (T/T_0)0.25 \quad (3.14)$$

is very small ($(T/T_0)^{0.25} \approx 0.03$ at typically measured temperatures). This is the reason why the temperature dependences of l and L are important. If values from Friedman and Pollak (1978) are used $l \approx L$ and clusters with $j > 1$ and $N > 1$ would be so rare as to be irrelevant. (This question has implications on existing derivations of the conductivity of narrow 2D strips and on non-ohmic effects, which both depend on $l \approx L$.) But the derivation here is a simultaneous solution of three 'well-known' equations.

Integral (3.13) can be transformed to

$$I_{N,R} = (1/2\pi^2) \int dj \int dt \sin t [t + A_0 j^{2\nu} e^j]^{-1} \quad A_0 \equiv \pi^2(l/L)^2(\omega_c/\omega). \quad (3.15)$$

In (3.15) the integral over j is done first; the lowest-order term in $\omega/\pi^2\omega_c$ is found when the lower limit is extended to 0 and e^j is neglected

$$I_{N,R} = (1/2\pi^2) \int dt (\sin t) t^{(1/2\nu-1)} (\pi/2\nu) \operatorname{cosec}(\pi/2\nu) / A_0^{1/2\nu}. \quad (3.16)$$

By $\omega \approx \omega_c \pi^2$ higher-order terms reduce the value of the conductivity. Subsequent integration over N yields

$$I_{N,R} = (1/4\pi\nu) \operatorname{cosec}(\pi/2\nu) \Gamma(1/2\nu) \pi^{-1/\nu} [\sin \pi/4\nu] [(\omega/\omega_c)(L/l)^2]^{1/2\nu} \quad (3.17)$$

and combined with (3.11) and the definitions of $I_{N,R}$,

$$\operatorname{Re} \sigma(\omega) = \sigma_{DC} + (0.008\omega C/l)[1 + (T_0/T)^{0.14}(\omega/\omega_c)^{0.55}]. \quad (3.18)$$

Although the integrations are neither particularly straightforward nor trivial, they are not described here in detail because of space limitations. Performed for arbitrary d they yield

$$\sigma(\omega) = \sigma_{DC} \{1 + [(\omega/\omega_c)(l/L)^2]^{(d-1)/2} + [(\omega/\omega_c)(l/L)^2]^{(d-1+1/\nu)/2}\} \quad (3.19)$$

where numerical constants have been suppressed. The latter yields

$$\sigma(\omega)/\sigma_{DC} = f(\omega/\omega_c T^p) \quad (3.20)$$

with

$$p = 2(\nu - \sigma\nu)/(d + 1). \quad (3.21)$$

For $d = 1$ (when distributions with infinite resistance values are excluded) known results are obtained (Zwanzig 1982)

$$\sigma(\omega) \propto \omega^0 + \omega^{1/2} \quad p = 0 \quad (3.22)$$

since in 1D $\nu = \sigma = 1$ and l and L do have the same temperature dependence. Moreover, the frequency-independent term is known to arise from critical R_s and the frequency-dependent term from 'off-critical' R_s . For $d = 3$ one has $p = 0.5$. This value agrees closely with experiment in both a-Si and a-Si:H:Au (if $\omega_c = \nu_{ph} \exp[-(T_0/T)^{1/4}] = (R_c C)^{-1}$). It is worth noting here that p arises from the quotient $(l/l^2)/(l/L^2) = (L/l)^2$, i.e. from the difference in the densities of AC- and DC-current-carrying paths. Systems, therefore, for which scaling of $\sigma(\omega)/\sigma_{DC}$ as a function of ω/ω_c does not require an extra temperature-dependent factor (such as 1D VRH systems and ionic conducting glasses), must have the same densities of AC- and DC-current-carrying paths. Also, in order for the same power of T to apply to both terms, the exponential in the cluster distribution function, n_s , must be a function of an argument with l/L , a ratio which only arises when cluster currents are expressed in terms of chain currents. At this point, however, neither experimental nor theoretical results are so accurate or extensive that it is certain that two different terms in the low ω response exist; it cannot be claimed that the same T -dependent scaling must be found in each (expressed as a ratio of $\sigma(\omega)$ to σ_{DC}).

Experimental results for $\text{Re } \sigma(\omega)$ and $\text{Im } \sigma(\omega)$ are reported in terms of the scaled loss functions

$$F_R(\omega) = [\sigma(\omega) - \sigma_{DC}]\omega_c/\sigma_{DC}\omega \quad F_I(\omega) = (\omega_c\omega/\sigma_{DC})(d/d\omega)[\text{Im } \sigma(\omega)/\omega]. \quad (3.23)$$

Results for $F_R(\omega)$ are

$$F_R(\omega) \approx 0.008(L/l)^2 + 0.008(\omega/\omega_c)^{1/2\nu}(L/l)^{2+1/\nu}. \quad (3.24)$$

$\text{Im } \sigma(\omega)$ has not been calculated here directly. Several approximations are based on the fact that processes with $\tau \approx 1/\omega$ contribute most to $\text{Re } \sigma(\omega)$; these approximations are inappropriate for calculation of $\text{Im } \sigma(\omega)$. Thus $\text{Im } \sigma(\omega)$ is best obtained by Kramers-Kronig relations. If

$$\sigma(\omega) = A\omega^s \quad (3.25)$$

then it is known that

$$\text{Im } A/\text{Re } A = \tan s\pi/2. \quad (3.26)$$

While this relationship is strictly valid only if the power s holds for all frequencies, introduction of a cut-off frequency ω_c introduces corrections linear in the ratio ω/ω_c .

These corrections are not seen by $F_1(\omega)$ (because of division by ω and subsequent differentiation) so that the relevant terms in $\text{Im } \sigma(\omega)$ yield for $F_1(\omega)$

$$F_1(\omega) = 0.006(L/l)^2 + 0.005(\omega/\omega_c)^{1/2\nu}(L/l)^{2+1/\nu}. \quad (3.27)$$

In the case $s = 1$, $\tan s\pi/2$ diverges (giving rise to a logarithmic divergence in $\text{Im } A$, but not in $\text{Im } \sigma(0)$); L'Hopital's rule has been used to generate the constant factor in the term linear in ω . The results for $F_R(\omega)$ and $F_1(\omega)$ give excellent agreement with experiment in a-Si (Hunt 1991a) if the result for the pre-exponential for σ_{DC} is taken from experiment and not from theory (known to be inapplicable in this particular system).

4. Discussion and conclusions

It is clear that non-local relaxation represented by cluster polarization currents with enhanced relaxation times is responsible for the observed low-frequency AC conductivity of VRH systems which is strongly enhanced over a low-frequency Debye cut-off. The description here of these cluster currents in terms of independent chain currents leads to excellent experimental agreement, but leaves some questions unanswered. One is the question of intra-cluster screening, i.e. do the shorter chains of a given cluster generate a significantly depolarizing field? If so, does this field slow relaxation further (as in Pollak (1971))? Or does it reduce the effective chain capacitance? (In Hunt (1992) it is shown that intra-chain screening does so.) Another is whether significant charge transport between chains occurs. This question cannot be answered without relaxing the approximation severing bonds. It is not clear to what extent various relaxation modes in different chains of impedances can be identified as independent. Thus the particular decomposition of clusters chosen cannot be rigorously justified, i.e. quantitative error estimates are not available.

Questions regarding the relevance of such concepts as anomalous diffusion, etc to transport in glasses have been discussed at some length (Hunt 1991b). It is worth repeating, however, that the general features (a loss peak which is broad and asymmetric in $[\sigma(\omega) - \sigma_{DC}]/\omega$ at a frequency ω_c which is proportional to σ_{DC} (Barton 1966, Nakajima 1972, Namikawa 1975)) of relaxation in glasses are reproduced in this and related treatments, and that more specific system-dependent properties are generated as well.

That $\sigma(\omega)$ should have contributions with two different powers (below ω_c) is a product of the decomposition of the clusters considered and of the different temperature dependences of l and L . The decomposition appears to maximize $\sigma_p(\omega)$ with no redundancy. Moreover agreement of $\sigma(\omega)$ with experiment and of the transverse $\sigma_{DC}(\sigma_\perp)$ in thin films (Hunt 1991c) is good. But if l and L should have the same magnitude, the importance of the term in $\omega^{(1+1/2\nu)}$ diminishes greatly; although it should not be necessary to reformulate the theory, agreement with experiment regarding the frequency dependence would be worsened. However, the scaling relationship for $\sigma(\omega)/\sigma_{DC}$ is a strong indicator that l and L do have different temperature dependences. The question then arises, which agreements with experiment are better established, those with $\sigma(\omega)$ (and σ_\perp in thin films), or those (where it is assumed that $l \approx L$) in non-ohmic effects? And which theoretical approach is more solidly grounded?

In summary, this work appears to answer some questions relating to transport in the Fermi glass and to raise others. The field, while reasonably well understood, is in need of further theoretical advances as well as additional experimental work, particularly in the very-low-frequency limit (where the linear frequency dependence in (3.18) should take over). Further research into these questions will aid in establishing the relevance of deviations from the simplest model assumptions, the appropriateness of the various calculation schemes, and the general physical behaviour of amorphous semiconductors. By extension, the physics of disordered insulators generally will be better understood.

Appendix

The treatment in section 3, where $\sigma(\omega)$ is calculated from the longest cluster chains will ultimately require a dielectric treatment of ellipsoidal inclusions with non-uniform polarizations in a medium ϵ_m . Interactions between such ellipsoidal inclusions in a random assemblage cannot be accurately treated in dipole-dipole form, because in principle their separations may be less than their individual lengths. Nevertheless, the generation of such ellipsoids by taking the longest chains of each individual cluster does guarantee that their minimum separations be greater than their individual lengths because the clusters are isotropic, so that the dipole approximation may be sufficient.

We consider a dielectric cylinder of macroscopic height, h , and radius r , so that the volume $V = \pi r^2 h$. The quantities ρ and h will be allowed to diverge. The bulk of the dielectric has a frequency-dependent dielectric constant, $\epsilon_m(\omega)$, while spherical volumes of radius a_i and dielectric constants $\epsilon_i(\omega)$ are distributed randomly inside, according to the cluster statistics of percolation (Stauffer 1979). A free charge density $\sigma_f = \mathbf{D} \cdot \mathbf{n}$ is placed on the top and bottom surfaces of the cylinder. The field \mathbf{D} induces a uniform polarization of both media; the polarization of the inclusions may be considered uniform for the purpose of calculating the field outside the spheres. The actual non-uniformity will contribute to higher-order terms in the multipole expansion of the field. Although we neglect these terms here, it has been shown that this approximation may not be adequate.

We assume that both the dielectric and the spheres are acted on by a macroscopic field \mathbf{E} . This average field is given by

$$\mathbf{E} = \epsilon_m^{-1}(\sigma_f + \sigma_p) - \sum_i (4\pi a_i^3 / \Omega) f(\epsilon_i) \mathbf{E}_i$$

where \mathbf{E}_i is the field acting on sphere i , and with $f(\epsilon_i) = (\epsilon_i - \epsilon_m) / (\epsilon_i + 2\epsilon_m)$, appropriate for spherical volumes. Now $\sigma_f + \sigma_p$, the sum of the free- and polarization-charge densities, would be $\mathbf{E} \cdot \mathbf{n}$ in the absence of the spheres, and one would have $\sigma_p = -\sigma_f + D / \kappa_m$, with $\kappa_m = \epsilon_m / \epsilon_0$. But in our case

$$\sigma_p = -\sigma_f + (\epsilon_0 / \kappa_m) \left[(D / \epsilon_0) - \sum_i (4\pi a_i^3 / \Omega) f(\epsilon_i) \mathbf{E}_i \right] - \sum_i (4\pi a_i^3 / \Omega) f(\epsilon_i) \mathbf{E}_i.$$

The second term in the square brackets represents the interaction between the finite clusters and the DC cluster, as the DC cluster feels a smaller average field when the

effect of the polarization of the clusters is included. Solving the above equation for E gives

$$E = (D_0/\epsilon_0\kappa_m) \left[1 + (1 + \kappa_m^{-1}) \sum_i (4\pi\alpha_i^3/\Omega) f(e_i) \right].$$

Here the average field is taken for the moment to be the local field; later the well known linear relation $E_{\text{loc}}/E = (1 + \chi_m)/(1 + 2\chi_m/3)$ will be applied. The effective dielectric constant, $\epsilon_{\text{eff}} = D_0/E$ is

$$\epsilon_{\text{eff}} = \epsilon_m \left[1 + (1 + \kappa_m^{-1}) \sum_i (4\pi\alpha_i^3/\Omega) f(\epsilon_i) \right].$$

Now if the polarization current in a given cluster is assumed to be spread uniformly throughout the cluster, but expressions for $\sigma_p(\omega)$ are used consistent with the current in the longest chain, one gets for $\sigma(\omega) = \epsilon(\omega)i\omega$:

$$\sigma(\omega) = \sigma_{\text{DC}} \left[1 + (1 + \kappa_m^{-1}) \sum_N n_N I_N / \sigma_{\text{DC}} \right] = \sigma_{\text{DC}} + (1 + i\omega\epsilon_0/\sigma_{\text{DC}}) \sum_N n_N I_N$$

where σ_N is the conductivity of an N -chain, and where the term $i\omega\epsilon_0/\sigma_{\text{DC}}$ now represents the interaction between the DC cluster and the finite clusters. This factor may be neglected if the low-frequency behaviour of the conductivity is desired, so that we arrive at the expression used at the beginning of section 3. Also, if the relation for the local field in terms of the average field is applied, one finds again that correction factors of the order of $1 + i\omega\epsilon_0/\sigma_{\text{DC}}$ are generated, which may be neglected in the low-frequency limit.

References

- Apsley N and Hughes H 1975 *Phil. Mag.* B 31 1325
 Austin I G and Mott N F 1969 *Adv. Phys.* 18 41
 Barton J L 1966 *Verres Refr.* 20 328
 Boettger H and Bryksin V V 1985 *Hopping Conduction in Solids* (Weinheim, Federal Republic of Germany: VCH)
 Dyre J 1988 *J. Appl. Phys.* 64 2456
 Fishchuk I I and Rudko V N 1980 *J. Phys. C: Solid State Phys.* 13 L493
 Friedman L and Pollak M 1978 *Phil. Mag.* B 38 173
 Hunt A 1990 unpublished
 — 1991a *Phil. Mag.* B 65 377
 — 1991b *Solid State Commun.* 80 151
 — 1991c *J. Non-Cryst. Solids* 134 287
 — 1992 *Phil. Mag.* B submitted
 Hunt A and Pollak M 1990 *Hopping and Related Phenomena* ed M Pollak and H Fritzsche (Singapore: World Scientific) p 309
 Long A R, McMillan J, Balkan N and Summerfield S 1988 *Phil. Mag.* B 58 153
 Long A R and Hansmann L 1990 *Hopping and Related Phenomena* ed M Pollak and H Fritzsche (Singapore: World Scientific)
 Miller A and Abrahams E 1960 *Phys. Rev.* 120 745
 Mott, N F 1969 *Phil. Mag.* 19 835

- Movaghar B, Pohlmann B and Schirmacher W 1980 *Phil. Mag.* B 41 49
- Nakajima T 1972 1971 *Ann. Rep., Conf. on Electric Insulation and Dielectric Phenomena* (Washington, DC: National Academy of Sciences) p 168
- Namikawa H 1975 *J. Non-Cryst. Solids* 18 173
- Niklasson G A 1987 *J. Appl. Phys.* 62 R1
- Pollak M 1971 *Proc. R. Soc. A* 325 383
- 1972 *J. Non-Cryst. Solids* 10 1
- 1974 *Amorphous and Liquid Semiconductors: Proceedings* ed J Stuke and W Brenig (New York: Halsted Press) p 127
- 1987 *Disordered Semiconductors* ed M Pollak (Boca Raton, FL: CRC Press) ch 5b
- Pollak M and Geballe T H 1961 *Phys. Rev.* 120 745
- Pollak M and Pohl H A 1975 *J. Chem. Phys.* 63 2980
- Reikh M and Ruzhin I M 1991 *Preprint*
- Stauffer D 1979 *Phys. Rep.* 54 1
- Summerfield S 1985 *Phil. Mag.* B 52 9
- Summerfield S and Butcher P N 1982 *J. Phys. C: Solid State Phys.* 15 703
- Summerfield S and Chroboczek J A 1985 *Solid State Commun.* 53 129
- Zwanzig R 1982 *J. Stat. Phys.* 28 127