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Effective-Medium Theory for the ac Behavior of a Random System*

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It is shown that the ideas of effective-medium theory can be applied to the evaluation of the ac behavior of a random system. The results for systems of which the components have no dispersion, show dispersion. In particular, it is found that the ω^s behavior of the ac conductivity of many disordered systems is reproduced to a good approximation over a limited frequency range.

The method of approach of substituting an effective medium for the real medium in the treatment of transport problems in inhomogeneous media has been emphasized by several authors,[1-5] most recently by Cohen and Jortner.[6] Here we shall be not so much concerned with relating the model macroscopically inhomogeneous medium to its underlying microscopic structure, as with discussing the ac behavior of the model. The motivation for this comes from experiments which closely resemble the model,[7,8] and the fact that these experiments yield results which closely parallel those in systems such as amorphous semiconductors, transition-metal glasses, and chalcogenide glasses.

When using some measurement system to determine the real and imaginary parts of the conductivity (or alternatively the dielectric constant), one experimentally defines the real part σ_m and imaginary part ϵ_m such that

$$\sigma_m^*(\omega)=\sigma_m(\omega)+i\omega\epsilon_0\epsilon_m(\omega),$$
$$\epsilon_m^*(\omega)=\epsilon_m(\omega)-i\sigma_m(\omega)/\epsilon_0\omega. \quad (1)$$

Since these quantities are experimentally defined, there must exist an experimentally defined average field $E_m^*(\omega)$ within the sample. The problem is to relate the average or measured values of (1) to the values of σ and ϵ of the components of the medium. We choose a medium consisting of two components, one with complex dielectric constant ϵ_1^* and concentration C, the other being defined by ϵ_2^* and $1-C$. We insist that the frequency of the applied field be such that this field is the same at all points in space at any instant of time. For sample thicknesses ~ 1 μm, this limits the present model to frequencies of about 30 GHz, that is, within the frequency range of standard bridge, stand-

ing-wave, or time-domain reflectometry techniques.

We start from a single-component medium, the effective medium, characterized by ϵ_m^*, and replace it piecewise with the original system using spherical volumes. Initially, the field at infinity and within a small spherical volume inside the sample is E_m^*. When the volume is replaced by original material, the new field within the spherical volume is given by

$$E_i^* = E_m^* - \left(\frac{\epsilon_i^* - \epsilon_m^*}{\epsilon_i^* + 2\epsilon_m^*}\right) E_m^*, \quad i = 1, 2. \tag{2}$$

When all replacements are effected and we are back to the original medium, the average difference between E_i^* and E_m^* must be zero, i.e., we must have

$$\langle E_i^* - E_m^* \rangle = 0. \tag{3}$$

Carrying out the averaging process explicit in (3) (which is not strictly limited to a binary distribution such as we have chosen) yields the following set of equations for $\sigma_m(\omega)$ and $\epsilon_m(\omega)$:

$$\begin{aligned} (\sigma_m/\sigma_1 - \sigma/\sigma_1)^2 &= (A^2 + B^2)^{1/2} + A, \\ (\epsilon_m/\epsilon_1 - \epsilon/\epsilon_1)^2 &= [(A^2 + B^2)^{1/2} - A] \tan^2\delta_1, \end{aligned} \tag{4}$$

where

$$2A = \left(\frac{\sigma}{\sigma_1}\right)^2 + \frac{1}{2}\frac{\sigma_2}{\sigma_1} - \left[\left(\frac{\epsilon}{\epsilon_1}\right)^2 + \frac{1}{2}\frac{\epsilon_2}{\epsilon_1}\right](\tan\delta_1)^{-2}, \tag{5}$$

$$B = \left(\frac{1}{4}\frac{\sigma_2}{\sigma_1} + \frac{\epsilon\sigma}{\epsilon_1\sigma_1} + \frac{1}{4}\frac{\epsilon_2}{\epsilon_1}\right)(\tan\delta_1)^{-1}, \tag{6}$$

$$a = \tfrac{1}{4}(3C - 1)a_1 + \tfrac{1}{4}(2 - 3C)a_2, \quad a = \sigma \text{ or } \epsilon, \tag{7}$$

$$\tan\delta_1 = \sigma_1/\omega\epsilon_0\epsilon_1. \tag{8}$$

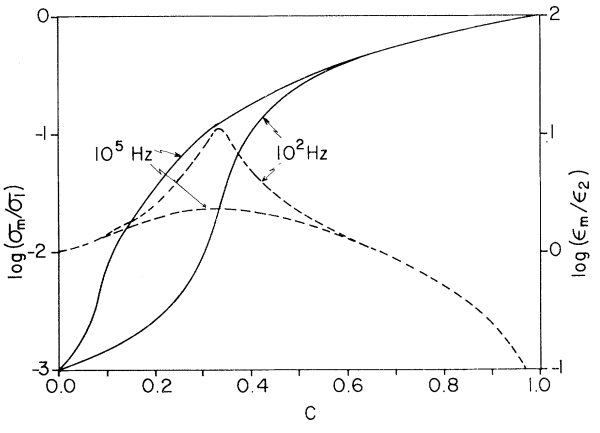

FIG. 1. Variation of σ_m/σ_1 (solid lines) and ϵ_m/ϵ (dashed lines) with C at 10^2 and 10^5 Hz, for the conditions $\epsilon_2 = 10$, $\epsilon_1 = 0$, $\sigma_2/\sigma_1 = 10^{-3}$, and $\sigma_1 = 10^{-5}$ mho cm^{-1}.

The system of Eqs. (4) – (8) describes the frequency dependence of the effective conductivity and dielectric constant. They can be shown to reduce to the familiar expression[4-6] for the effective dc conductivity σ_{0m} for $\epsilon_1 = \epsilon_2 = 0$, and an exactly parallel result for ϵ_{0m} in the case that $\sigma_1 = \sigma_2 = 0$. This last result is of interest because many studies have been made of liquid mixtures. Such a situation is not easy to make comparisons with because of the strong dependence of ϵ on density. The most obvious area of applicability is this context is that of the critical point of binary liquid mixtures, where one has a theoretical prescription for the concentration fluctuations.

Figure 1 displays the variation of σ_m/σ_1 and ϵ_m/ϵ_2 versus C at two low frequencies. The conditions correspond roughly with some experiments performed by Bueche,[7] and the curves give a reasonable explanation of those results. The striking effect occurs in Fig. 2, where $\sigma_m(\omega)/\sigma_1$ and $\epsilon_m(\omega)/\epsilon_2$ are plotted versus frequency. Over the frequency range covered by an audio-frequency bridge, and a bit beyond (where measurements are most plentiful), we see a dependence of the type[9] $\sigma_m(\omega) \propto \omega^s$. Such a dependence is common in the materials mentioned in the beginning over this frequency range. Hauser[8] recently found a similar dependence in sputtered metal–metal-oxide films, which he quoted as evidence for a hopping mechanism.

It would seem that if the materials mentioned possess a microscopic aggregatelike structure, whether it be a physical clumping or complicated

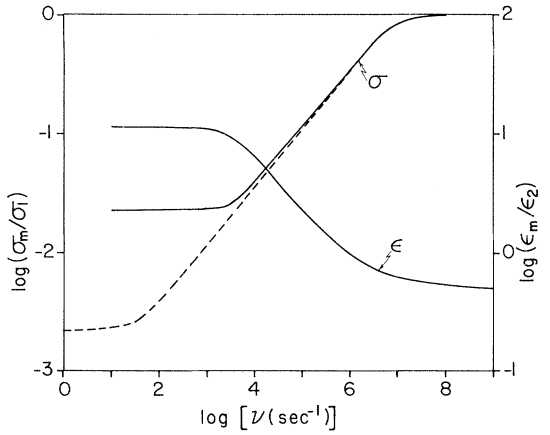

FIG. 2. Variation of σ_m/σ_1 and ϵ_m/ϵ_2 for the same conditions as in Fig. 1, but for $C = \frac{1}{3}$ (solid curves). Over the straight parts of the curves $\sigma_m/\sigma_1 \propto \omega^{0.46}$, $\epsilon_m/\epsilon_2 \propto \omega^{-0.44}$. The dashed curve is for σ_m/σ_1 with $\sigma_2/\sigma_1 = 10^{-5}$. The slope gives $\sigma_m/\sigma_1 \propto \omega^{0.49}$.

network of variable complex conductances such as envisioned in percolation theory, this alone could produce the sublinear frequency dependence of the ac conductivity. Previous attempts at analyzing this sort of situation have been confined to low concentrations and have been reviewed by Van Beek.[10] The results displayed in Fig. 2 are for components which individually are nondispersive. The present development, as given by Eqs. (4)–(8), is not confined to low concentrations, and also includes the case in which the individual components may be dispersive. The results in this latter case are not too dissimilar to those shown in Fig. 2. That is, it is easy to find cases in which an apparent $\sigma \propto \omega^s$, with $s < 1$, behavior occurs over a significant frequency range. Experimental results are often presented with the dc conductivity σ_{0m} subtracted out. This process, because of experimental uncertainties, often obscures the very low-frequency behavior, which in the present case is proportional to ω^2. Similarly, the ω^s behavior is often observed to hold to the highest frequency used experimentally. If, in fact, such behavior holds for all frequencies, one finds from the Kramers-Kronig relations that $\sigma/\epsilon \propto \omega$. Over the range of frequencies where the ω^s behavior occurs in the present case, this relationship is not obeyed because the behavior is not true for all frequencies. Thus it would seem that from an experimental point of view both components of the complex conductivity need to be measured over a very large frequency range in order to understand better the amorphous or random systems discussed to date.

Commonly, varying the temperature of the system is somewhat equivalent to varying the measurement frequency because of the fact that relaxation times are usually activated. In the present context, temperature variation could be expected to have the effect of changing the value of σ_2/σ_1. The effect of such a change is illustrated in Fig. 2. The shift in the value of s is much more dramatic if the dc conductivity is subtracted out. If both σ_1 and σ_2 are activated, with σ_2 more strongly so than σ_1, σ_2/σ_1 decreases as the temperature decreases, with a concomitant increase in the value of the index s. This behavior also is typical of that observed in several glassy systems.

To conclude, it has been shown that for a binary mixture of two materials which each have both a constant dielectric constant and a dc conductivity, the ac behavior exhibits dispersion, as shown in Fig. 1, and also displays a power-law dependence on frequency, with an exponent less than 1, over a considerable frequency range. The expressions developed for the real and imaginary parts of the complex conductivity are equally applicable to a binary mixture whose individual components exhibit dispersion. The present results indicate once again the extreme importance of determining the structural characteristics of an amorphous material at the same time as other measurements are made.

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