

Phenomenological equations for the electrical conductivity of microscopically inhomogeneous materials*

A. Davidson[†] and M. Tinkham

Division of Engineering and Applied Physics and Department of Physics, Harvard University, Cambridge, Massachusetts 02138

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Phenomenological equations for the electrical conductivity of microscopically inhomogeneous materials are proposed. These equations combine ideas from the effective-medium theory and from percolation theory and, for some interesting cases, improve on the effective-medium theory in the vicinity of the percolation threshold. The equations are compared, without adjustable parameters, to three different experimental situations and agree with data remarkably well.

I. INTRODUCTION

We propose phenomenological equations for the electrical conductivity of microscopically inhomogeneous materials. The equations remain valid if the material is drawn into wire, and for some interesting cases improve on the effective-medium-theory¹⁻³ (EMT) result near the percolation threshold. The equations were initially constructed to reconcile data on a drawn superconducting composite wire (made by Tsuei's process⁴) with both the EMT and with percolation⁵ theory; hence our phenomenological ideas are an amalgamation of the results of these theories. The EMT, which works poorly for drawn wire, defines a homogeneous effective medium to replace the real heterogeneous mixture by demanding that a suitable average of the effects of inclusions in the effective medium must be zero. For the case where one constituent material is insulating the EMT predicts that a critical concentration of conducting material is necessary for bulk conduction. Percolation theory also predicts a critical volume fraction, but the two theories predict different values; the difference is enormous for drawn wires, in which the inclusions are highly elongated in the drawing direction. Our phenomenological equations are constructed to satisfy the percolation result for the threshold while resembling the EMT result far from the critical region. This approach generates two simple equations that fit data from three diverse experiments without any adjustable parameters, so we present it here as a guide to both experimenters and theorists concerned with disordered systems.

II. EFFECTIVE-MEDIUM THEORY FOR ALIGNED PROLATE SPHEROIDS

The effective-medium theory for electrical conduction in disordered materials attempts to define self-consistently a homogeneous effective medium that replaces the real heterogeneous medium surrounding a particular inhomogeneity. The problem is simplified (but still physically inter-

esting) if we confine our attention to real heterogeneous media composed of only two materials. We have then a mixture of two real media, material 1 with bulk resistivity ρ_1 and material 2 with resistivity ρ_2 , and an imaginary effective medium whose bulk resistivity ρ_m is to be calculated.

The condition we used to set the value of ρ_m is generated as follows: (i) Let the homogeneous effective medium surround an inclusion of either real material. (ii) Calculate the total current flowing through the midplane of the inclusion and subtract off the total current that would have flowed there if the inclusion were replaced by the effective medium. (iii) We demand that the average value of this excess current be zero, where the average is taken over the different possible compositions and orientations of the inclusions. ρ_m is adjusted until this condition is met.

We have already simplified the problem by allowing only two possible compositions for the inclusions, and ordinarily the problem is further simplified by assuming the inclusions are all spherically shaped, eliminating the orientational average. We will be slightly less simple, and assume that the inclusions are prolate spheroids with their symmetry axes aligned in the direction of the applied field. This removes the isotropic degeneracy of the problem but does not introduce an orientational average. It is also physically relevant for inhomogeneous material drawn into wire, as in Tsuei's process.

The calculation then proceeds as follows. The total current I_i flowing through an inclusion is

$$I_i = \frac{1}{\rho_m} \pi b^2 E \frac{u_i}{1 + (u_i - 1)X}, \quad (1)$$

where b is the semiminor axis of the ellipsoid; u_i is ρ_m/ρ_i , $i = 1, 2$; E is the applied electric field; X is the depolarization factor for the ellipsoid. The value of X is given by⁶

$$X = \frac{1 - \epsilon^2}{\epsilon^3} \left(\frac{1}{2} \ln \frac{1 + \epsilon}{1 - \epsilon} - \epsilon \right), \quad (2)$$

where ϵ is defined as the eccentricity of ellipsoid. Note that for a sphere, the eccentricity is zero, b is the radius, and $X = \frac{1}{3}$. If we let $u_i = 1$ (the inclusion has the same resistivity as the effective medium) the current would be $(1/\rho_m)\pi b^2 E$, so that the excess current is

$$\Delta I_i = \frac{\pi b^2 E}{\rho_m} \frac{(u_i - 1)(X^{-1} - 1)}{(X^{-1} - 1 + u_i)}. \quad (3)$$

Now let the concentration of material 1 be C_1 and that of material 2 be C_2 . Then the average value of the excess current is

$$\langle \Delta I \rangle = C_1 \Delta I_1 + C_2 \Delta I_2,$$

where

$$C_1 + C_2 = 1.$$

The condition that this average be zero leads to a quadratic equation for ρ_m whose solution is the effective-medium-theory result;

$$2\rho_m = \rho_1 \left(1 - \frac{C_2}{X}\right) + \rho_2 \left(1 - \frac{C_1}{X}\right) + \left\{ \left[\rho_1 \left(1 - \frac{C_2}{X}\right) + \rho_2 \left(1 - \frac{C_1}{X}\right) \right]^2 - 4\rho_1\rho_2 \left(1 - \frac{1}{X}\right) \right\}^{1/2}. \quad (5)$$

Equation (5) may be put into an equivalent form by changing from resistivity to conductivity as the parameter describing the constituents of the system. If $\sigma_i = 1/\rho_i$, where $i = 1, 2, m$, then

$$2\sigma_m \left(\frac{1}{X} - 1\right) = \sigma_1 \left(\frac{C_1}{X} - 1\right) + \sigma_2 \left(\frac{C_2}{X} - 1\right) + \left\{ \left[\sigma_1 \left(\frac{C_1}{X} - 1\right) + \sigma_2 \left(\frac{C_2}{X} - 1\right) \right]^2 + 4 \left(\frac{1}{X} - 1\right) \sigma_1 \sigma_2 \right\}^{1/2}. \quad (6)$$

Equations (5) and (6) emphasize two limits in the behavior of ρ_m or σ_m . If the conductivity of one constituent goes to zero the system is best described by the conductivity form (6). If the resistivity of one constituent goes to zero, then we are served better by (5). This distinction will be useful to us later.

Equations (5) and (6) fit data in situations where the conductivity ratio σ_1/σ_2 is close to unity,^{1,5,7} but when this ratio becomes very large or very small a serious deficiency develops for the EMT. If, for example, we let σ_1 go to zero in (6) (we adopt the convention that material 2 is always the material with the higher conductivity) we have

$$\frac{\sigma_m}{\sigma_2} = \begin{cases} \frac{1}{(1/X - 1)} \left(\frac{C_2}{X} - 1\right), & X < C_2 \leq 1 \\ 0, & 0 \leq C_2 \leq X. \end{cases} \quad (7)$$

Hence the EMT predicts that the composite will act like an insulator until a critical volume frac-

tion of conducting material is reached, and that the critical value is exactly the depolarizing factor for the particular shape inclusion. For spherical inclusions, where $X = \frac{1}{3}$, this threshold comes at a not unreasonable value. If the material is drawn into wire, however, so that the depolarizing factor for the inclusions approaches zero without a change in topology, the EMT predicts the unlikely result that the threshold approaches zero. A similarly unphysical result occurs if, instead of letting σ_1 go to zero, we let ρ_2 go to zero. From (5) we have

$$\frac{\rho_m}{\rho_1} = \begin{cases} 1 - C_2/X, & 0 \leq C_2 \leq X \\ 0, & X < C_2 \leq 1. \end{cases} \quad (8)$$

Equation (8) is the EMT description of resistivity in a composite of normal and superconducting materials. Again, there is a critical volume fraction of superconducting material required for bulk superconducting behavior, and this concentration threshold is none other than the depolarizing factor of the inclusions. This false prediction of close coupling of the critical volume fraction to the shape of the inclusions is a serious problem for the EMT, and one must go outside the EMT to resolve the dilemma.

III. PERCOLATION THEORY

The problem of correctly assigning this critical volume fraction can be addressed from a point of view entirely different from that of the EMT; this independent body of science is percolation theory.^{5,8} A self-contained discussion of percolation theory is beyond the scope of this paper, but we wish to point out various of the concepts involved, especially the idea of the percolation probability. This quantity may be defined for our purposes with the aid of a regular cubic network of equivalent resistors. Let a voltage be imposed across this network so that a current flows in the resistors along a cubic axis. Now begin removing resistors at random from the network leaving open circuits in place of each removed resistor. The percolation probability is defined as the probability that a given node is connected by resistors to infinitely many other nodes (in an infinite network). As more and more resistors are removed the percolation probability grows smaller, and when it finally vanishes, conduction ceases. Clearly this problem is related to the problem considered by the EMT, and the critical volume fraction corresponds to the situation where the percolation probability changes from some finite value to zero.

It has been shown⁵ for resistor lattices that there is a finite density of remaining resistors at

which the percolation probability indeed becomes zero. This critical density has been subjected to a number of analytical and numerical studies for various sorts of regular lattices in both two and three dimensions.^{5,6} The results of these studies are not of direct relevance to us because they apply only to these regular lattices where individual resistors have been randomly removed. However, Scher and Zallen⁹ pointed out that one could construct from the critical resistor density a critical volume fraction of conducting material that is nearly independent of the particular lattice used, and which depends primarily on the dimensionality of the lattice. The critical volume fraction that they suggested should apply to any three-dimensional heterogeneous *continuum*¹⁰ is approximately $(15 \pm 2)\%$. This should be contrasted to the value of 33% predicted by the EMT for spherical inclusions. Furthermore, we expect, in contradiction of the EMT, that this critical volume fraction is largely independent of the shape of the inclusions, so long as they are randomly arranged and there is rough topographical symmetry¹⁰, between the two phases.

The concept of an invariant critical volume fraction for given dimensionality has been criticized by Pike and Seager,¹¹ among others. However, their objections seem to apply primarily to powder mixture composites (where there is indeed experimental evidence against an invariant critical volume fraction). Here we consider composites in the continuum limit, and there is a measure of experimental evidence^{7,12} in that limit to support Scher and Zallen's contention. Hence, we tentatively accept the critical volume fraction of $\sim 15\%$ as correct for biphase continuous media.

The EMT fails to predict this percolation threshold correctly because there is nothing in it to take account of large clusters of like inclusions—its only independent parameter is the shape of the inclusions, and changing their shape by drawing the material into wire reveals the shortcoming of this single parameter. To do the calculation correctly in the spirit of the EMT one would have to include terms that average the excess current through pairs of inclusions, and triplets and so on. Such a calculation¹³ is very difficult and has not yet borne fruit.⁷ On the other hand we already have independent knowledge of the critical volume fraction from percolation theory, and by phenomenologically combining these two theories we can produce simple equations that describe diverse data.

IV. TSUEI'S SUPERCONDUCTING COMPOSITES AND OUR PHENOMENOLOGICAL EQUATIONS

The phenomenological amalgamation of the EMT and of percolation theory was necessitated by our

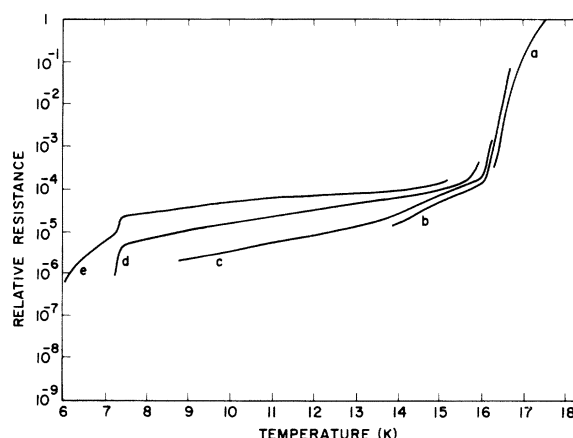


FIG. 1. Resistive transition of the 7½%-superconductive wire made by Tsuei's process. The resistance of the sample relative to its normal-state resistance is plotted as a function of temperature. Curves *a* through *e* are measurements made with different current levels, ranging in decades from 6×10^{-5} A for curve *a* to 0.6 A for curve *e*. The long plateau below T_c (~ 15 K) at high currents indicates that the filaments are essentially isolated from one another.

own experiments on samples of a disordered superconducting composite wire made by Tsuei's process.⁴ This process results in a multifilamentary composite of Nb_3Sn filaments in a copper matrix where the filaments form a disordered array within the copper, and each filament is the order of 1 cm long and 1 μm in diameter. The depolarizing factor for this wire may be calculated from Eq. (2) by using the relation between the eccentricity ϵ and the area reduction ratio R of the drawn wire: $1 - \epsilon^2 = 1/R^3$. For typical wire with a reduction ratio of 600, $X \cong 10^{-7}$. Of course, this calculation involves a physical approximation since the filaments are not in the shape of perfect prolate spheroids. Our measurements have included low-field magnetization tests, high-field critical current tests, and resistance measured as a function of temperature. We have had available for test wires with two different compositions: one about 7.5% superconductor by volume, and the other about 15%. Each kind of measurement shows a qualitative difference between the two kinds of wire, but for the purposes of this paper we shall concentrate on the resistance results. These results are displayed in Figs. 1 and 2. The long plateau in the data from the 7.5% sample on the higher current curves is consistent with the resistance levels estimated by Davidson, Beasley, and Tinkham¹⁴ (DBT). Their estimation assumed that the filaments were typically isolated from each other by normal copper; hence this assumption would appear to be justified in this sample. The 15% sample, however, shows no such

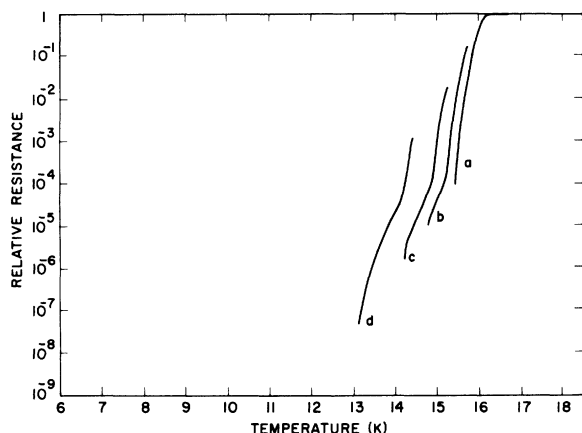


FIG. 2. Resistive transition of the 15%-superconductive wire made by Tsuei's process. The resistance of the sample relative to its normal-state resistance is plotted as a function of temperature, as in Fig. 1. Curves *a* through *d* are measurements made with different current levels, ranging from 6×10^{-4} A for curve *a* to 0.6 A for curve *d*. The geometry of this wire and the $7\frac{1}{2}\%$ wire are about the same, both being about 0.5 mm in diameter, and with reduction ratios in the range of 200 to 300. The relatively sharp transition indicates that the filaments are in some sense continuous.

plateau at the same current levels, so that one would conclude that the filaments are not so isolated, and either form genuine connections or else very good proximity-effect junctions. These measurements, then, are consistent with the theory of percolation in a continuum but not with the EMT (i. e., the EMT predicts a threshold at about $\sim 10^{-5}\%$ by volume of superconductor since the demagnetizing factor for these well drawn out filaments is $\sim 10^{-7}$). We have assumed here that the Tsuei wire system is well approximated by a continuum because the Cu and Nb_3Sn should be in excellent electrical contact over their entire interfacial area.

Thus we are faced with reconciling these facts: The percolation threshold should be about 15%, and below this threshold the estimates of DBT are approximately right. A function that meets the test of these conditions may readily be constructed, however, in the spirit of Padé approximations.¹⁵ That is, we shall try to construct a function as the ratio of two polynomials that satisfies the basic facts known about the Tsuei wire system.

Since the first requirement is that the resistivity of the composite must be zero for concentrations above the percolation threshold, we choose the numerator of our Padé approximant to be the simplest function which has a zero at that point:

$$(\text{numerator}) = \rho_1(1 - C_2/C_2^*),$$

where $C_2^* \cong 0.15$ and is the percolation threshold, but we make the convenient approximation that $C_2^* = \frac{1}{6}$. Having accounted for near-neighbor connectivity in this brute-force manner, we may now choose the denominator so that the elongated shape of the inclusions is accounted for:

$$(\text{denominator}) = (1 + C_2/X).$$

Hence, we propose for the resistivity,

$$\begin{aligned} \frac{\rho_m}{\rho_1} &= \frac{(1 - 6C_2)}{(1 + C_2/X)}, & 0 \leq C_2 < \frac{1}{6} \\ &= 0, & \frac{1}{6} < C_2 \leq 1. \end{aligned} \quad (9)$$

For $C_2 < X \ll 1$ this function is dominated by the denominator, and therefore agrees to first order with the EMT; but close to the threshold, clustering takes over and the numerator provides a manifest zero at the right place. Further, still in the limit of a small depolarizing factor, (9) has the form

$$\rho_m \cong \rho_1 \frac{\ln(4R^3)}{2R^3 C_2}, \quad X < C_2 < \frac{1}{6} \quad (9a)$$

which is very close to DBT's estimate, and also suggestive of some results from Callaghan and Toth.¹⁶ Equation (9) is therefore consistent with our measurements, although the details of that equation are not confirmed by the two data points supplied by Figs. 1 and 2. However, there are already published experiments with more detailed results.^{7,12} This simple theory works well for them too, even though the appropriate value of X is $\frac{1}{3}$ and not 10^{-7} , so that there is no longer first-order agreement with the EMT.

Before we can make a comparison with these experiments, we must generalize our approach to cover both ends of the C_2 scale. Equation (9) applies only to a normal-superconductor (NS) composite, so we must now work out the analogous case for a metal-insulator composite, where the conductivity (rather than the resistivity) of one constituent goes to zero. For this case we refer to the simple EMT result, (7). In contrast to the NS case, we expect (7) to be valid near $C_2 = 1$ rather than for $C_2 = 0$. Evidently, to make a first-order expansion we would be better off to write (7) in terms of $C_1 (= 1 - C_2)$, which is small in the region of validity:

$$\frac{\sigma_m}{\sigma_2} = 1 - \frac{C_1}{1 - X}, \quad 0 \leq C_1 < 1 - X. \quad (10)$$

Notice that (10) has the same form as (8). We now generalize this EMT result with our Padé approach. The threshold should occur when the highly conducting material forms a connected network at a volume fraction of about $\frac{1}{6}$ so that $C_2^* = \frac{1}{6}$ or $C_1^* = \frac{5}{6}$. Hence the analog to (9) is

$$\frac{\sigma_m}{\sigma_2} = \frac{1 - \frac{5}{3} C_1}{1 + C_1/(1-X)} \quad (11a)$$

Rewriting (11a) in terms of C_2 for convenience in comparison, we have

$$\frac{\sigma_m}{\sigma_2} = \frac{1-X}{5} \frac{6C_2-1}{2-X-C_2}, \quad \frac{1}{6} < C_2 \leq 1. \quad (11b)$$

Finally, we also rewrite (9) in terms of conductivity:

$$\frac{\sigma_m}{\sigma_1} = \frac{1+C_2/X}{1-6C_2}, \quad 0 \leq C_2 < \frac{1}{6}. \quad (12)$$

These equations, (11b) and (12), are the fundamental equations of our phenomenological approach, and they are in a sense complementary. That is, although they both assume that the conductivity of material 1 is quite small compared to material 2, (11b) should be valid for $C_2 \approx 1$, while (12) will be valid near $C_2 \approx 0$. The two equations diverge in opposite directions at the percolation threshold, but, as we shall see, they remain valid until very near the critical region provided $\sigma_1/\sigma_2 \ll 1$. We note that there is first-order agreement with the EMT in (11a) only if $X \approx 1$, and in (12) only if $X \approx 0$. It would be simple to add a linear term to the denominator of (11a) or the numerator of (12) that would guarantee first-order agreement with the EMT for all X in both equations, but the over-all fit to the data in the following is best if (11) and (12) are used as written above. That is, we shall see empirically that (11) and (12) work well for all values of C_2 except in the immediate vicinity of $C_2 = \frac{1}{6}$. The EMT with $X = \frac{1}{3}$ works well only for C_2 near 1, and it is apparently a mistake to demand first-order agreement with the EMT on both sides of the threshold.

V. COMPARISON TO EXPERIMENT

A table-top experiment performed by Fitzpatrick, Malt, and Spaepen¹² (FMS) is an example of the case where σ_1 may be taken to be effectively zero and $X = \frac{1}{3}$. Random close-packed lattices of insulating and metallic spheres were put together in glass beakers with aluminum foil electrodes and the conductivity as a function of relative densities of the two kinds of spheres was studied. Since one conductivity may be taken as zero we expect (11) to apply. To compare the results we switch to the notation of FMS, and plot the conductivity as a function of the *number fraction* of insulating spheres. That is, if q is the insulating number of fraction, then C_1 , the insulating volume fraction, is $C_1 = 0.65q + 0.35$, where the filling factor of the lattice is taken to be 0.65.¹⁷ Hence we may use (11a) with C_1 put in terms of q , and $X = \frac{1}{3}$, and plot the results direct-

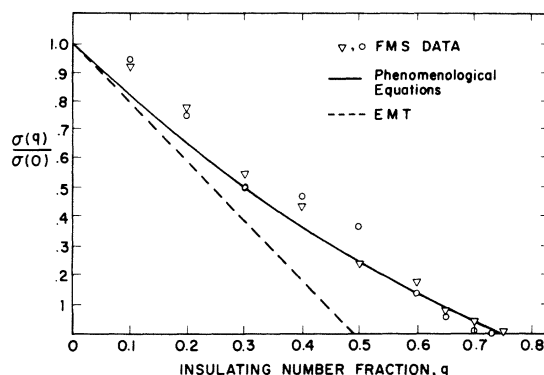


FIG. 3. Conductivity as a function of the fraction of insulating spheres in an experiment by FMS (Ref. 12). To apply our equations we take the interstitial spaces as part of the insulating volume fraction. The results of the EMT are also plotted. Note that the data and both theories have been normalized not at zero volume fraction of insulating spheres, which is unattainable in this system, but at zero *number* fraction.

ly over the data of FMS. This is presented in Fig. 3. The high-conductivity data appear too high in comparison to our equations, but this is consistent with the experimental errors reported by FMS. The low-conductivity data and the threshold fit quite well, and over-all we feel that these experimental data are consistent with our formulas.

A more detailed set of mathematical and experimental data are compiled in a paper by Webman, Jortner, and Cohen⁷ (WJC). They compare conductivity data¹⁸ from a Li-NH₃ system to data derived from computer simulations. The simulations involved calculating the effective conductivity of a cubic resistor network in which each resistor may have one of only two values, and in which the resistor values are correlated over a length larger than the distance separating the nodes but are random on a scale larger than the correlation length. Hence their simulation is just the sort of inhomogeneous system considered in this paper, with $X = \frac{1}{3}$, and they demonstrate convincingly that the metal-ammonia system is described very well by their model network. We shall therefore compare our phenomenological equations both to WJC's numerical simulations and to the Li-NH₃ data which they present in their paper. In Fig. 4, Eqs. (11a) and (12) with $X = \frac{1}{3}$ are compared to the metal-ammonia data and to the EMT result. The phenomenological fit is remarkably good with no adjustable parameters except the conductivity values at either extreme of the C_2 scale. Of course, at the percolation threshold the phenomenological theory diverges, but the fit to the data is good except within $\Delta C_2 = \pm 0.03$

around the threshold. Even within this divergent area it is easy to interpolate a straight line, tangent to both curves, that adequately describes the data.

A feature that is emphasized by the log plot in Fig. 4 is that as the ratio σ_1/σ_2 is varied the shapes of the two curves generated by Eqs. (11b) and (12) are unchanged, but their relative positions slide vertically. It is therefore apparent that for conductivity ratios near 1 the phenomenological equations will be unsatisfactory, but that for more extreme differences the two curves will come closer and closer to approximating a single smooth curve. This feature is clearly visible in Fig. 5, which compares Eqs. (11b) and (12) for the case of $\sigma_1/\sigma_2 = 10^{-5}$ to one of WJC's numerical simulations. The two phenomenological curves now come very close together and still give a nice fit to the numerical data, again with no adjustable parameters. We have also plotted in Fig. 5 two curves generated by the EMT. These curves are plots of (6) with appropriate values of conductivities. One curve uses a depolarizing factor of $\frac{1}{3}$, as would seem appropriate for the isotropic nature of WJC's simulation, and the complete failure at the percolation threshold is apparent. On the other hand, the curve has some appeal because it has the correct qualitative shape, and one might be tempted to improve upon its shape by artificially moving the threshold over to the correct value. The resulting curve, again (6) but with $X = \frac{1}{6}$, is the remaining curve in Fig. 5. Indeed the behavior is better at the threshold and below, but worse above. Furthermore this approach has no hope of coping with the separation of the depolarizing factor and percolation threshold for drawn anisotropic wire.

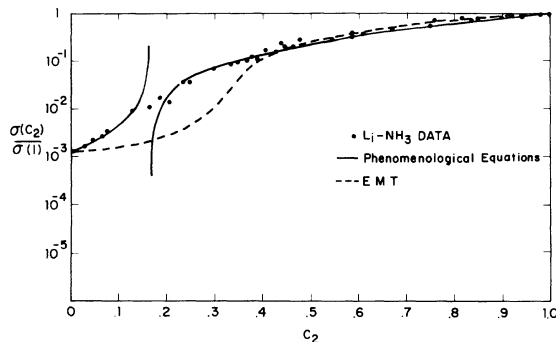


FIG. 4. Comparison of Li-NH₃ data of Refs. 7 and 18 with the EMT and the phenomenological equations. Despite the divergence of the phenomenological curves at the percolation threshold, the theory remains valid to $C_2 = C_2^* \pm 0.03$. The EMT misses the threshold completely.

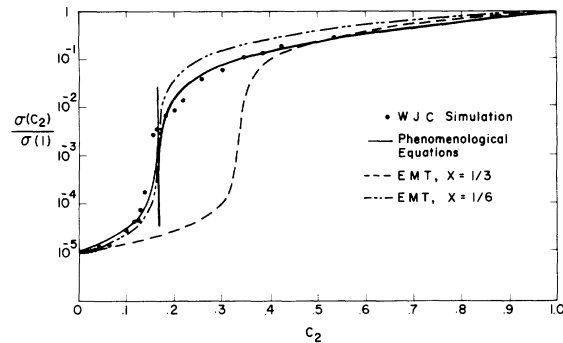


FIG. 5. Comparison of a HJC (Ref. 7) simulation for $\sigma_1/\sigma_2 = 10^{-5}$ with the EMT with $X = \frac{1}{3}$ and $\frac{1}{6}$, and with our phenomenological equations. The phenomenological curves diverge over a very small region for this case of lower conductivity ratio. The EMT with $X = \frac{1}{6}$ fits the critical region and below, but fails above; moreover, it represents use of a physically inappropriate value of X .

We are left then with the phenomenological approach.

VI. CONCLUSION

The phenomenological equations presented here, which were initially generated to reconcile the properties of a highly anisotropic heterogeneous superconductor with the EMT, also work well for isotropic normal microscopically heterogeneous conductors. In their present form the equations are limited to those situations in which the constituent materials differ greatly in conductivity, but a more generalized form should be possible. Even without such a generalization these equations represent a significant improvement on the EMT in building in the proper percolation threshold. Further, we note that the experimental data discussed here support the idea of an invariant critical volume fraction for percolation in some kinds of biphased continua.

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†Present address: IBM Research Laboratory, Yorktown Heights, N. Y. 10598.

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