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A GENERALIZED DIFFERENTIAL EFFECTIVE MEDIUM THEORY

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ABSTRACT

A GENERALIZATION of the Differential Effective Medium approximation (DEM) is discussed. The new scheme is applied to the estimation of the effective permittivity of a two phase dielectric composite. Ordinary DEM corresponds to a realizable microgeometry in which the composite is built up incrementally through a process of homogenization, with one phase always in dilute suspension and the other phase associated with the percolating backbone. The generalization of DEM assumes a third phase which acts as a backbone. The other two phases are progressively added to the backbone such that each addition is in an effectively homogeneous medium. A canonical ordinary differential equation is derived which describes the change in material properties as a function of the volume concentration ϕ of the added phases in the composite. As $\phi \to 1$, the Effective Medium Approximation (EMA) is obtained. For $\phi < 1$, the result depends upon the backbone and the mixture path that is followed. The approach to EMA for $\phi \cong 1$ is analysed and a generalization of Archie's law for conductor–insulator composites is described. The conductivity mimics EMA above the percolation threshold and DEM as the conducting phase vanishes.

1. INTRODUCTION

THERE are essentially three different ways of approaching the effective medium problem for a composite made up of several materials. The first is to define a particular microgeometry and then proceed to solve the field equations in this geometry. The second approach is to obtain bounds on the possible range of the effective properties. The Hashin–Shtrikman bounds (HASHIN, 1962; HASHIN AND SHTRIKMAN, 1963) are the most general that depend only upon the volume fractions of each material. More restrictive bounds require knowledge of correlation functions in addition to the volume fractions (MILTON, 1981). The third approach estimates the effective properties by means of some sort of self-consistent scheme (HASHIN, 1968), or effective medium "theory". Among the latter are the Effective Medium Approximation (EMA) of BRUGGEMAN (1935) (e.g. LANDAUER, 1952; STROUD, 1975; MENDELSON and COHEN, 1982), the Differential Effective Medium Approximation (DEM) also due to BRUGGEMAN (e.g. ROSCOE, 1952; MCGLAUGHLIN, 1977; SEN, SCALA and COHEN, 1981; YONEZAWA and COHEN, 1983; SHENG and CALLEGARI, 1984) and the cumulant approximation of Hori (HORI and YONEZAWA, 1975). We note that EMA is also known as the Coherent Potential Approximation (CPA) (STROUD, 1977) and DEM as the Iterated Dilute Approximation (IDA). An informative historical account of the various effective medium theories is given by LANDAUER (1978).

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The above approaches can be put into three categories. This is by no means exhaustive, but is useful in order to define the way one attacks the effective medium problem. The approach of this paper will be different from the three above, but will most closely resemble the third. We start by considering the inverse problem. Thus, instead of asking what are the effective properties of a given sample of composite, we construct a class of composites with known effective properties. The original question can then be answered by selecting from the class of composites the one whose microstructure most resembles the given sample. Recently, MILTON (1984) has shown that EMA gives the exact solution for a class of realizable microgeometries. It is also known (SEN *et al.*, 1981) that DEM gives the correct solution for a different class of microgeometries. The common characteristic in both these realizations is the idea of incremental homogenization. Starting with a given homogeneous material, one successively adds small amounts of inhomogeneity. At each addition of inhomogeneity, the incremental changes in the effective properties are calculated using the known dilute concentration results.

In this paper we generalize the idea of incremental homogenization to define a generalized version of DEM. The theory is developed for the effective dielectric permittivity of the medium. The generalization contains a certain amount of non-uniqueness in that the effective permittivity depends upon the way in which we construct the final material from an initially homogeneous material. This path-dependent aspect of the theory is novel. In particular, the effective permittivity estimates of EMA and DEM correspond to special limiting cases of paths and endpoints, respectively. Each different path corresponds to a definite type of underlying structure for the "random composite". The answer to the forward problem of predicting the properties of a given composite can thus be solved by finding which particular microgeometry or path best fits the given microgeometry.

We begin by defining a specific construction process in section 2. The homogenization process is introduced in section 3 and an ordinary differential equation for the homogenization process is derived. It is shown in section 4 that the differential equation is canonical in the sense that it is independent of the specific construction process. The equation is integrated and several example media are discussed in section 5. The case of spherical grains is considered in section 6. In section 7 the percolation properties of the generalized theory are analyzed. Numerical results are presented in section 8. The analogous equations for the bulk and shear moduli of a composite elastic medium will be presented in a forthcoming paper (NORRIS, 1985).

2. FIXED VOLUME PROCESS

The Fixed Volume Process (FVP) begins with a volume v_0 of material 0. The constitution of the material is then altered by incrementally taking out a small fraction of the total volume and replacing it with an equivalent volume of new material. At each step the total volume is kept fixed at v_0. We consider the case that the new material that replaces the removed material is composed of discrete volume elements of materials 1 and 2: define the volume fractions ϕ_0, ϕ_1 and ϕ_2 of the three constituent materials such

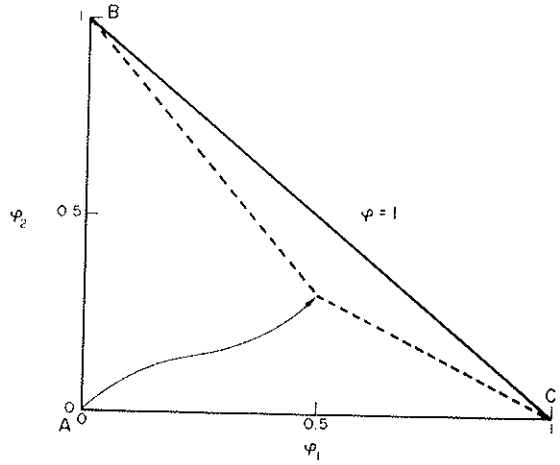

FIG. 1. A possible homogenization path in the (ϕ_1, ϕ_2) plane. The small triangle represents the range of further possible points.

that $\phi_0 + \phi_1 + \phi_2 = 1$. Thus, ϕ_1 and ϕ_2 are two independent parameters which may be used to describe the current configuration. To each FVP there is a corresponding path in the (ϕ_1, ϕ_2) plane (see Fig. 1). Introduce an arc length parameter t, so that on the path we have $\phi_1 = \phi_1(t)$, $\phi_2 = \phi_2(t)$. At each t the material is assumed to be homogeneous, i.e. any given volume v of the composite contains $\phi_0 v$ of material 0, $\phi_1 v$ of material 1 and $\phi_2 v$ of material 2.

We now examine the incremental replacement process. Let Δu be the volume of material removed at point t in the FVP. The same volume is replaced with volume Δu_1 of material 1, and volume Δu_2 of material 2 such that $\Delta u_1 + \Delta u_2 = \Delta u$. The volume of material 1 in the solid before the replacement was $\phi_1 v_0$. After replacement, the volume of material 1 is

$$v_0(\phi_1 + \Delta\phi_1) = v_0\phi_1\left(1 - \frac{\Delta u}{v_0}\right) + \Delta u_1. \tag{1}$$

This relates the increment in ϕ_1 for the FVP to the corresponding increments in u_1 and u_2. A similar relation follows for $\Delta\phi_2$. Note that u_1 and u_2 measure the total volumes of materials 1 and 2, respectively, that have been added during the FVP up to the current value of t. Therefore u_1 and u_2 describe a path $u_1 = u_1(t)$, $u_2 = u_2(t)$ in the (u_1, u_2) plane as t varies. Define the derivative $\dot{\phi}_1$ as

$$\dot{\phi}_1(t) = \frac{\mathrm{d}\phi_1}{\mathrm{d}t}. \tag{2}$$

Similarly, we define the derivatives $\dot{\phi}_2$, $\dot{u}_1$ and $\dot{u}_2$ so that (1) becomes

$$v_0\dot{\phi}_1 = (1 - \phi_1)\dot{u}_1 - \phi_1\dot{u}_2 \tag{3}$$

and the corresponding relationship for the change in the volume of material 2 gives

$$v_0\dot{\phi}_2 = (1 - \phi_2)\dot{u}_2 - \phi_2\dot{u}_1. \tag{4}$$

These relationships can be inverted to give $\dot{u}_1$ and $\dot{u}_2$ as

$$\frac{\dot{u}_1}{v_0} = \frac{(1-\phi_2)\dot{\phi}_1 + \phi_1\dot{\phi}_2}{1-\phi}. \tag{5}$$

$$\frac{\dot{u}_2}{v_0} = \frac{(1-\phi_1)\dot{\phi}_2 + \phi_2\dot{\phi}_1}{1-\phi}, \tag{6}$$

where $\phi \equiv \phi_1 + \phi_2$. Thus, the FVP can be described by a path in the (ϕ_1, ϕ_2) plane or its equivalent path in the (u_1, u_2) plane. However, there are restrictions on the permissible paths that may be considered. First, all paths are required to begin at the origin of both planes. Without loss of generality, put $\phi_1(0) = \phi_2(0) = u_1(0) = u_2(0) = 0$ and let $0 < t \leqslant t_0$ define the paths. Secondly, we require that materials 1 and 2 are added at each stage. This implies that $\dot{u}_1 \geqslant 0$ and $\dot{u}_2 \geqslant 0$. So the permissible range of u_1 and u_2 is the quarter space $u_1 \geqslant 0$, $u_2 \geqslant 0$. Thirdly, we must have $\phi_1 \geqslant 0$, $\phi_2 \geqslant 0$ while the sum $\phi \equiv \phi_1 + \phi_2$ must be less than unity. The permissible range of ϕ_1 and ϕ_2 is thus the interior and edge of the triangle in the (ϕ_1, ϕ_2) plane with vertices at the points (0, 0), (0, 1) and (1, 0). Now consider the identity:

$$\frac{(1-\phi_2)\dot{\phi}_1 + \phi_1\dot{\phi}_2}{(1-\phi)^2} = \frac{\mathrm{d}}{\mathrm{d}t}\left[\frac{\phi_1}{1-\phi}\right]. \tag{7}$$

Referring to (5) and (7), we see that the second and third conditions above imply that the two quantities $\phi_1/(1-\phi)$ and $\phi_2/(1-\phi)$ must both be monotonically increasing functions of t. This result can be interpreted geometrically as follows: at stage t in the FVP when ϕ_1 and ϕ_2 are at $\phi_1(t)$ and $\phi_2(t)$, the direction vector $(\dot{\phi}_1, \dot{\phi}_2)$ can only point from the current point (ϕ_1, ϕ_2) into the triangle with vertices at the points (1, 0), (0, 1) and (ϕ_1, ϕ_2) (see Fig. 1). Thus, the triangle which defines the permissible range in the (ϕ_1, ϕ_2) plane decreases in size as t increases. Similarly, the permissible range in the (u_1, u_2) plane at t is the quarter plane $u_1 \geqslant u_1(t)$, $u_2 \geqslant u_2(t)$.

Note that (1) is not to be thought of as defining the partial derivatives $\partial\phi_1/\partial u_1$ and $\partial\phi_1/\partial u_2$ of a function $\phi_1(u_1, u_2)$. If this were so, then (1) would give

$$\partial\phi_1/\partial u_1 = \frac{1}{v_0}(1-\phi_1), \tag{8}$$

$$\partial\phi_1/\partial u_2 = \frac{1}{v_0}(-\phi_1). \tag{9}$$

However, a necessary and sufficient condition that ϕ_1 be a function of u_1 and u_2 is that the mixed partial derivatives of second order be equal. Differentiating $\partial\phi_1/\partial u_1$ of (8) with respect to u_2 and subtracting from this the derivative of $\partial\phi_1/\partial u_2$ of (9) with respect to u_1, gives the quantity $1/v_0^2$, which is not zero. Therefore, ϕ_1 and ϕ_2 are not functionally related to u_1 and u_2. They are related only along curves in the (ϕ_1, ϕ_2) and (u_1, u_2) planes. The relationship is defined by (3) and (4), or (5) and (6). This means, for instance, that if two different curves in the (ϕ_1, ϕ_2) plane both go from (0, 0) to some point (ϕ_{10}, ϕ_{20}) then the values of u_1 and u_2 at this point can be different for the two curves. In other words, if (ϕ_1, ϕ_2) describe a path which is a closed curve (this is not permissible physically, but there is nothing against it mathematically) there is no reason why (u_1, u_2) must also describe a closed curve.

Define $u(t) = u_1(t) + u_2(t)$, the total volume of materials 1 and 2 at stage t that have

been used up in the FVP. Adding (3) and (4) gives

$$v_0\dot{\phi} = (1-\phi)\dot{u} \tag{10}$$

which, with the initial conditions $\phi(0) = u(0) = 0$, integrates to

$$1-\phi = e^{-(u/v_0)}, \tag{11}$$

This result implies that if $\phi \to 1$, then $u \to \infty$, and vice versa. Dividing (6) by (5) gives

$$\frac{\dot{u}_2}{\dot{u}_1} = \frac{\phi_2\dot{\phi}+(1-\phi)\dot{\phi}_2}{\phi_1\dot{\phi}+(1-\phi)\dot{\phi}_1}. \tag{12}$$

As $\phi \to 1$, (12) implies that u_1 and u_2 go to infinity such that $du_2/du_1 = \phi_2/\phi_1$.

Denote the points (0, 0), (0, 1) and (1, 0) in the (ϕ_1, ϕ_2) plane by A, B and C, respectively (see Fig. 1). The above results show that there is a one-to-one correspondence between the points on the perimeter of the triangle ABC and the points on the perimeter of the (u_1, u_2) quarter plane. Thus, between A and B we have $\phi_1 = 0$ and $0 \leqslant \phi_2 \leqslant 1$. The corresponding point in the (u_1, u_2) plane is $u_1 = 0$, $u_2 = -v_0 \ln(1-\phi_2)$. Along BC we have $\phi_1 + \phi_2 = 1$ which corresponds to $u_1, u_2 \to \infty$ such that $u_2/u_1 = \phi_2/\phi_1$.

It is clear from (3) and (4), or (5) and (6), that straight-line segments in the (ϕ_1, ϕ_2) plane go over to straight-line segments in the (u_1, u_2) plane. For example, the line $\phi_1 = f_1(1-e^{-t})$, $\phi_2 = f_2(1-e^{-t})$ becomes the line $u_1 = f_1 t v_0$, $u_2 = f_2 t v_0$.

For arbitrary curves, the exact integral of (11) reduces the system of two equations (3) and (4) to a single ordinary differential equation along the path. This equation is found by subtracting (4) from (3) and using (10) to get

$$\frac{d}{dt}\left[\frac{\phi_1-\phi_2}{1-\phi}\right](t) = \frac{1}{1-\phi}(\dot{u}_1-\dot{u}_2). \tag{13}$$

Integrating, and using the initial conditions, gives

$$u_1(t)-u_2(t) = (1-\phi)\int_0^t \frac{\phi_1(s)-\phi_2(s)}{1-\phi(s)}\,ds. \tag{14}$$

3. The Homogenization Process

We begin with the basic result for a dilute concentration of inhomogeneities in an otherwise homogeneous medium. Let the permittivity of the current medium be ε. In the context of the FVP, add a small volume $\Delta u = \Delta u_1 + \Delta u_2$ of two dissimilar materials with permittivities ε_1 and ε_2. Let $\bar{E}$ and $\bar{D}$ be the averaged electric and displacement fields, where the average is over all of v_0 after the incremental addition. The new permittivity $\varepsilon+\Delta\varepsilon$ is defined by the averaged relation

$$\bar{D} = (\varepsilon+\Delta\varepsilon)\bar{E}. \tag{15}$$

We have the exact relation (Landau and Lifshitz, 1960),

$$\bar{E}\Delta\varepsilon = \frac{1}{v_0}(\varepsilon_1-\varepsilon)\int_{\Delta u_1} E\,dv + \frac{1}{v_0}(\varepsilon_2-\varepsilon)\int_{\Delta u_2} E\,dv. \tag{16}$$

We now invoke the dilute concentration approximation to calculate the field in Δu_1 and Δu_2. Assume that the volumes Δu_1 and Δu_2 are each made up of similarly shaped ellipsoids that are randomly oriented in space. It is known (POLDER and VAN SANTEN, 1946; LANDAU and LIFSHITZ, 1960) that

$$\int_{\Delta u_j} E \, dv = E_0 g_j(\varepsilon) \Delta u_j, \quad j = 1 \text{ or } 2, \tag{17}$$

where E_0 is the average electric field in the unperturbed volume and the quantity $g_j(\varepsilon)$ is

$$g_j(\varepsilon) = \frac{\varepsilon}{3} \sum_{i=1,3} \frac{1}{(1-L_i^{(j)})\varepsilon + L_i^{(j)}\varepsilon_j}, \quad j = 1 \text{ or } 2. \tag{18}$$

Here, the quantities $L_i^{(j)}$, $i = 1, 2, 3$ are the depolarization coefficients of the ellipsoids associated with material j, where $j = 1$ or 2. The depolarization coefficients are real and positive and satisfy

$$\sum_{i=1,3} L_i^{(j)} = 1, \quad j = 1, \text{ or } 2. \tag{19}$$

The depolarization coefficients are all equal to 1/3 in the case that the ellipsoids are spheres. The average electric field follows from (17) as

$$\bar{E} = E_0 \left[\left(1 - \frac{\Delta u}{v_0} \right) + \frac{\Delta u_1}{v_0} g_1(\varepsilon) + \frac{\Delta u_2}{v_0} g_2(\varepsilon) \right]. \tag{20}$$

The incremental change in ε follows from (16), (17) and (18) as

$$\Delta\varepsilon = -\left[\frac{\Delta u_1 G_1(\varepsilon) + \Delta u_2 G_2(\varepsilon)}{v_0 - \Delta u + \Delta u_1 g_1(\varepsilon) + \Delta u_2 g_2(\varepsilon)} \right], \tag{21}$$

where

$$G_j(\varepsilon) = (\varepsilon - \varepsilon_j) g_j(\varepsilon), \quad j = 1, 2. \tag{22}$$

The homogenization process is defined by (21). In the limit $\Delta u_1, \Delta u_2 \to 0$, we get

$$\Delta\varepsilon = \frac{-1}{v_0} \{ \Delta u_1 G_1(\varepsilon) + \Delta u_2 G_2(\varepsilon) \}. \tag{23}$$

This equation must be interpreted as defining the increment in ε as u_1 and u_2 progress incrementally along a path in the (u_1, u_2) plane. Let t parametrize the path, then (23) becomes

$$\dot{\varepsilon}(t) = \frac{-1}{v_0} \{ G_1(\varepsilon)\dot{u}_1(t) + G_2(\varepsilon)\dot{u}_2(t) \}. \tag{24}$$

This ordinary differential equation for $\varepsilon(t)$ reflects the change in ε as u_1 and u_2 progress along their path. We need to define an initial value to ε. At $t = 0$ we have $u_1 = u_2 = 0$ and $\varepsilon = \varepsilon_0$, where ε_0 is the permittivity of the original volume. Therefore, (24) with $\varepsilon(0) = \varepsilon_0$ defines an initial value problem for ε.

We can rewrite (24) for $\dot{\varepsilon}$ in terms of the variables $\phi_1(t)$ and $\phi_2(t)$ by using (5) and (6)

$$\dot{\varepsilon}(t) = -(G_1\dot{\phi}_1 + G_2\dot{\phi}_2) - (G_1\phi_1 + G_2\phi_2)\frac{\dot{\phi}}{1-\phi}. \tag{25}$$

As we shall see in the next section (25) is the canonical equation for the homogenization process. Equation (24) is not incorrect, but it is phrased in terms of variables that do not have intrinsic significance to the current configuration.

Referring to (23), we note that this relation does not define an exact differential $\Delta\varepsilon$. If it did, we would have

$$\partial\varepsilon/\partial u_1 = \frac{-1}{v_0} G_1(\varepsilon), \tag{26}$$

$$\partial\varepsilon/\partial u_2 = \frac{-1}{v_0} G_2(\varepsilon). \tag{27}$$

Taking the mixed partial second derivatives of (26) and (27), we see that in general they do not agree. Thus, ε is not simply a function of u_1 and u_2. This is similar to the relationship we found between the variables (u_1, u_2) and (ϕ_1, ϕ_2). Mathematically, it means that given ε at one point in the (u_1, u_2) plane, there does not exist a unique integral surface that defines ε at every other point. Physically, it means that if we follow two different paths in the (u_1, u_2) plane from an initial point to a final point, such that ε is the same at the initial point, then in general the two paths will give different values of ε at the final point. Similarly, ε is not functionally related to ϕ_1 and ϕ_2, so that different paths in the (ϕ_1, ϕ_2) plane also give different results. In general, the homogenization equations (24) and (25) should be thought of as defining a path-dependent process.

4. Variable Volume Process

The FVP process is characterized by taking a piece of the current material out and replacing it with materials 1 and 2 so that the total volume is kept constant. Now consider the case where no material is taken out, but only put in. The volume will thus increase at each step in the process, so we call it a Variable Volume Process (VVP).

Begin with an initial volume, v_0. At each step in the homogenization, incremental volumes Δv_1 and Δv_2 of materials 1 and 2 are added to the current total volume $V \equiv v_0 + v_1 + v_2$. Define the current volume fractions ϕ_0, ϕ_1 and ϕ_2 such that $\phi_0 + \phi_1 + \phi_2 = 1$ as

$$\phi_1 = \frac{v_1}{V}, \quad \phi_2 = \frac{v_2}{V}. \tag{28}$$

Thus, ϕ_0 is the volume fraction of the original material and ϕ_1 and ϕ_2 are the volume fractions of the added materials. At each stage the material is assumed to be homogeneous, in that arbitrary volumes have volume fractions ϕ_0 of material 0, ϕ_1 of material 1 and ϕ_2 of material 2. Note that (28) defines a one-to-one relationship between points in the (v_1, v_2) plane and points in the (ϕ_1, ϕ_2) plane. This contrasts with the situation for the FVP where there did not exist such a functional relationship. The difference is trivially apparent, since in the VVP we are retaining all the added material within the mixture. There is thus no indeterminacy in how the new material is

distributed in terms of volume fractions. The inverse of (28) is

$$\frac{v_1}{v_0} = \frac{\phi_1}{1-\phi}, \quad \frac{v_2}{v_0} = \frac{\phi_2}{1-\phi}. \tag{29}$$

In particular, note that ϕ can only approach unity if v_1 and/or v_2 go to infinity. This is obvious since the volume fraction of the original material, ϕ_0, can go to zero only if the added material occupies a volume much greater than v_0. The VVP is inefficient in this sense compared with the FVP. From (11) we see that for the original material to achieve a volume fraction of $\phi_0 \equiv 1-\phi$, the FVP requires adding a volume $u = v_0 \ln|\phi_0|$. With the same starting volume v_0, the VVP requires adding a volume of $v = v_0(1-\phi_0)/\phi_0$. Thus, v always exceeds u and the ratio v/u becomes infinite as $\phi \to 1$.

The relevant homogenization equation for the VVP is now briefly derived. Let ε be the current permittivity at the point (ϕ_1, ϕ_2) in the process. The current volume is $V = v_0 + v_1 + v_2$. After the addition of the incremental volumes Δv_1 and Δv_2 the permittivity becomes $\varepsilon + \Delta\varepsilon$, where in analogy with (16) we now have

$$\bar{E}\Delta\varepsilon = \frac{1}{V+\Delta v}(\varepsilon_1 - \varepsilon)\int_{\Delta v_1} E \, dv + \frac{1}{V+\Delta v}(\varepsilon_2 - \varepsilon)\int_{\Delta v_2} E \, dv, \tag{30}$$

where $\Delta v = \Delta v_1 + \Delta v_2$. Using the dilute concentration approximation,

$$\int_{\Delta v_j} E \, dv = E_0 g_j(\varepsilon)\Delta v_j, \quad j = 1 \text{ or } 2, \tag{31}$$

where E_0 is the average electric field in the material exterior to Δv_1 and Δv_2, and g_j are defined in (18). The average field $\bar{E}$ is then

$$\bar{E} = \frac{E_0}{V+\Delta v}\{V + \Delta v_1 g_1(\varepsilon) + \Delta v_2 g_2(\varepsilon)\}. \tag{32}$$

The increment in permittivity follows from (31), (32) as

$$\Delta\varepsilon = -\left[\frac{\Delta v_1 G_1(\varepsilon) + \Delta v_2 G_2(\varepsilon)}{V + \Delta v_1 g_1(\varepsilon) + \Delta v_2 g_2(\varepsilon)}\right], \tag{33}$$

where G_j are defined in (22). In the limit as Δv_1 and $\Delta v_2 \to 0$, (33) becomes

$$\Delta\varepsilon = \frac{-1}{V}\{\Delta v_1 G_1(\varepsilon) + \Delta v_2 G_2(\varepsilon)\}. \tag{34}$$

Now consider an homogenization path $\phi_1 = \phi_1(t)$, $\phi_2 = \phi_2(t)$. This is equivalent to a path in the (v_1, v_2) plane defined by the relations in (29). So we have $v_1(t)$ and $v_2(t)$, with initial conditions $v_1(0) = v_2(0) = 0$. The incremental relationship (34) then defines the process $\varepsilon = \varepsilon(t)$, where

$$\dot{\varepsilon}(t) = \frac{-1}{V}\{G_1(\varepsilon)\dot{v}_1(t) + G_2(\varepsilon)\dot{v}_2(t)\}, \tag{35}$$

with initial condition $\varepsilon(0) = \varepsilon_0$. Note that (35) is similar to (24), but not quite the same. The difference lies with the variables (v_1, v_2) and (u_1, u_2) which have different physical

interpretations. However, eliminating v_1 and v_2 from (35) using (29) gives an ordinary differential equation for $\varepsilon(t)$ identical in form to (25). This happens because the variables ϕ_1 and ϕ_2 have the same physical meaning in both the FVP and the VVP. Thus, (25) is a canonical equation for this type of homogenization, independent of the specific material replacement process.

5. Integrating the Homogenization Equation

In general the homogenization equation (25) cannot be integrated directly. However, it may be integrated along straight-line segments. Consider the straight-line in the (ϕ_1, ϕ_2) plane between the two points (ϕ_{1A}, ϕ_{2A}) and (ϕ_{1B}, ϕ_{2B}). Define the following quantities

$$\Delta\phi_1 = \phi_{1B} - \phi_{1A}, \tag{36}$$

$$\Delta\phi_2 = \phi_{2B} - \phi_{2A}, \tag{37}$$

$$\Delta\phi = \Delta\phi_1 + \Delta\phi_2, \tag{38}$$

and

$$p_1 = \{\Delta\phi_1(1-\phi_{2A}) + \Delta\phi_2\phi_{1A}\}/\Delta\phi, \tag{39}$$

$$p_2 = \{\Delta\phi_2(1-\phi_{1A}) + \Delta\phi_1\phi_{2A}\}/\Delta\phi. \tag{40}$$

Thus, $p_1 + p_2 = 1$. Note that $\Delta\phi_1$, $\Delta\phi_2$ and $\Delta\phi$ are not infinitesimal quantities. Let ε_A and ε_B be the permittivities at the two endpoints of the line segment. Then by any parametrization of the line segment, (25) can be reduced to

$$\int_{\varepsilon_A}^{\varepsilon_B} \frac{d\varepsilon}{p_1 G_1(\varepsilon) + p_2 G_2(\varepsilon)} = \ln\left[\frac{1-\phi_B}{1-\phi_A}\right], \tag{41}$$

where $\phi_A = \phi_{1A} + \phi_{2A}$ and $\phi_B = \phi_{1B} + \phi_{2B}$. For any p_1 (and $p_2 = 1 - p_1$), define the indefinite integral $F(\varepsilon, \varepsilon_1, \varepsilon_2, p_1)$ by

$$F(\varepsilon, \varepsilon_1, \varepsilon_2, p_1) = \exp\left\{\int^{\varepsilon} \frac{d\gamma}{p_1 G_1(\gamma) + p_2 G_2(\gamma)}\right\}. \tag{42}$$

Then (41) becomes

$$\frac{F(\varepsilon_B, \varepsilon_1, \varepsilon_2, p_1)}{F(\varepsilon_A, \varepsilon_1, \varepsilon_2, p_1)} = \frac{1-\phi_B}{1-\phi_A}. \tag{43}$$

Consider the function $F(\varepsilon, \varepsilon_1, \varepsilon_2, p_1)$ of (42). For simplicity, we assume that the complex numbers ε_1 and ε_2 have the same phase. Then, without loss of generality, we can take that phase as zero. The case of different phases follows by analytic continuation. First, we define $\tilde{\varepsilon}_i$, $i = 1, 6$ to be the six non-zero roots of $G(\varepsilon) = 0$, where

$$G(\varepsilon) = p_1 G_1(\varepsilon) + p_2 G_2(\varepsilon)$$

$$= \frac{-\varepsilon}{3} \sum_{j=1,2} p_j \sum_{i=1,3} [L_i^{(j)} + \varepsilon/(\varepsilon_j - \varepsilon)]^{-1}. \tag{44}$$

Since ε_1 and ε_2 are both real and positive, it follows from the form of $G(\varepsilon)$ through (44) that five of the roots are negative and one is positive. The positive root lies between ε_1 and ε_2 and we denote it as $\varepsilon_E(p_1)$ since it is the permittivity predicted by EMA (STROUD, 1975; MENDELSON and COHEN, 1982) at volume fraction p_1 of material 1 and volume fraction $p_2 = 1-p_1$ of material 2. The five negative roots are located between the six points $-\varepsilon_j L_i^{(j)}/\{1-L_i^{(j)}\}$, $i = 1,3$, $j = 1,2$ on the negative real axis.

The function $F(\varepsilon, \varepsilon_1, \varepsilon_2, p_1)$ can now be evaluated by the method of partial fractions. We obtain in a straightforward manner,

$$F(\varepsilon, \varepsilon_1, \varepsilon_2, p_1) = \varepsilon^{-l} \prod_{i=1,6} (\varepsilon - \tilde{\varepsilon}_i)^{R_i}, \tag{45}$$

where

$$R_i = \text{Residue}\left[\frac{1}{G(\varepsilon)}\right]_{\varepsilon=\tilde{\varepsilon}_i}, \quad i = 1,6 \tag{46}$$

and

$$\frac{1}{l} = \frac{1}{3}\sum_{i=1,3}\left[\frac{p_1}{L_i^{(1)}} + \frac{p_2}{L_i^{(2)}}\right]. \tag{47}$$

The six residues, R_i satisfy the constraint

$$\sum_{i=1,6} R_i = (1-L)+l, \tag{48}$$

where L is defined by

$$\frac{1}{1-L} = \frac{1}{3}\sum_{i=1,3}\left[\frac{p_1}{1-L_i^{(1)}} + \frac{p_2}{1-L_i^{(2)}}\right]. \tag{49}$$

Note that l and L depend upon both the inclusion shapes and on p_1, but are independent of p_1 if the shapes are the same for materials 1 and 2. In general, l and L are bounded as follows,

$$0 \leqslant l \leqslant \frac{1}{3} \leqslant L \leqslant 1. \tag{50}$$

We have $l = 0$ when either material is shaped as needles or plates; $l = 1/3$ and $L = 1/3$ when both are spheres, and $L = 1$ when either or both are plates.

It can be shown quite easily that the R_i are all negative except for the one corresponding to $\varepsilon_E(p_1)$, which we denote R_E. Then it follows from the bounds on l and L and (48) that

$$0 \leqslant (1-L)+l \leqslant R_E. \tag{51}$$

Consider the process that follows a straight line in the (ϕ_1, ϕ_2) plane from $(0,0)$ to some point (ϕ_1, ϕ_2) such that $\phi = \phi_1 + \phi_2 < 1$. Let $\varepsilon = \varepsilon_0$ at $(0,0)$. Referring to (39) and (40), we have

$$p_1 = \phi_1/\phi, \tag{52}$$

$$p_2 = \phi_2/\phi. \tag{53}$$

Then, from (43) the value of ε at (ϕ_1, ϕ_2) is given by

$$\frac{F(\varepsilon, \varepsilon_1, \varepsilon_2, \phi_1/\phi)}{F(\varepsilon_0, \varepsilon_1, \varepsilon_2, \phi_1/\phi)} = 1-\phi. \tag{54}$$

It is instructive to rewrite this equation for ε using (45), as

$$\varepsilon - \varepsilon_E(\phi_1/\phi) = \left[(1-\phi)F(\varepsilon_0, \varepsilon_1, \varepsilon_2, \phi_1/\phi)\varepsilon^l \prod_{\substack{i=1,6 \\ i\neq E}} (\varepsilon - \tilde{\varepsilon}_i)^{-R_i}\right]^{1/R_E}, \tag{55}$$

where the product excludes ε_E. Based on the fact that $R_E > 0$, while the other R_i are negative, (55) shows that $\varepsilon \to \varepsilon_E(\phi_1)$ as $\phi \to 1$. This result is independent of the initial value, ε_0. It is not surprising that ε approaches a value independent of ε_0 as $\phi \to 1$, since according to either FVP or VVP, the volume fraction of the initial material tends to zero as $\phi \to 1$. Thus, all properties associated with the starting material are forgotten. Also, it is not really surprising that the value of ε attained is always that of EMA. This is to be expected from the result of MILTON (1984). In his paper, Milton shows that EMA can be realized through a process akin to the present homogenization. Milton's procedure, translated into the present terminology, amounts to the straight-line process from (0, 0) to (ϕ_1, ϕ_2), where $\phi = 1$.

Note from (55) that as $\phi \to 1$, we have $\varepsilon - \varepsilon_E(\phi_1) \sim (1-\phi)^{(1/R_E)}$, apart from a multiplicative constant. In the case of spheres, the "critical exponent", $(1/R_E)$ equals $1/(1+a) \leqslant 1$, where a is defined in (59).

6. Spherical Grains

The particular case in which both material shapes are spheres is of special interest. Then $l = L = 1/3$ and instead of six roots, there are now only two. They are $\tilde{\varepsilon}_1 \geqslant 0 \geqslant \tilde{\varepsilon}_2$, the roots of

$$2\varepsilon^2 + \varepsilon(\varepsilon_1 + \varepsilon_2 - 3\varepsilon_p) - \varepsilon_1\varepsilon_2 = 0, \tag{56}$$

where

$$\varepsilon_p = p_1\varepsilon_1 + p_2\varepsilon_2. \tag{57}$$

Thus, $\varepsilon_E(p_1) = \tilde{\varepsilon}_1$. The function $F(\varepsilon, \varepsilon_1, \varepsilon_2, p_1)$ becomes

$$F(\varepsilon, \varepsilon_1, \varepsilon_2, p_1) = \varepsilon^{-1/3}(\varepsilon - \tilde{\varepsilon}_1)^{(1+a)}(\varepsilon - \tilde{\varepsilon}_2)^{-a}, \tag{58}$$

where $a \geqslant 0$ is

$$a = \frac{\varepsilon_p - \tilde{\varepsilon}_1}{\tilde{\varepsilon}_1 - \tilde{\varepsilon}_2}. \tag{59}$$

We now specialize further to the case that p_1 becomes zero. This happens, for example, if the process only involves putting in material 2. As $p_1 \to 0$, we have ε_p, $\tilde{\varepsilon}_1 \to \varepsilon_2$, $\tilde{\varepsilon}_2 \to -\varepsilon_1/2$ and $a \to 0$, and (58) simplifies to

$$F(\varepsilon, \varepsilon_1, \varepsilon_2, 0) = \varepsilon^{-1/3}(\varepsilon - \varepsilon_2). \tag{60}$$

Suppose the homogenization begins at $\phi_1 = \phi_2 = 0$ with $\varepsilon = \varepsilon_0$. Then as the volume fraction ϕ_2 of material 2 is increased, the current ε is determined from (43) and (60) as the solution to

$$\frac{(\varepsilon-\varepsilon_2)}{(\varepsilon_0-\varepsilon_2)}\left[\frac{\varepsilon_0}{\varepsilon}\right]^{1/3} = 1-\phi_2. \tag{61}$$

This is just the usual Differential Effective Medium theory (DEM) result originally due to Bruggeman (LANDAUER, 1978; see also LAWS and WALPOLE, 1979; SEN *et al.*, 1981). DEM starts with all of ε_0 and material 2 is added incrementally until it has achieved its correct volume fraction.

7. CONDUCTOR–INSULATOR COMPOSITES

The case of $\varepsilon_2 \equiv 0$ is of special interest, as it corresponds to the situation that one of the added materials is insulating. The above general analysis becomes singular as $\varepsilon_2 \to 0$. Of the six roots $\tilde{\varepsilon}_i$, $i = 1, 6$, three of them become zero. The remaining roots, say $\tilde{\varepsilon}_i$, $i = 1, 3$ are determined by the cubic equation:

$$\sum_{i=1,3} \frac{p_1(\varepsilon-\varepsilon_1)}{(1-L_i^{(1)})\varepsilon + L_i^{(1)}\varepsilon_1} + \frac{p_2}{1-L^{(2)}} = 0, \tag{62}$$

where

$$\frac{1}{1-L^{(j)}} = \frac{1}{3}\sum_{i=1,3}\frac{1}{1-L_i^{(j)}}, \quad j = 1 \text{ or } 2. \tag{63}$$

Equation (62) has three real roots. Two of them are always negative; the third root is positive or negative, depending as p_1 is greater or less than p_{1c}, respectively, where the percolation threshold p_{1c} is

$$p_{1c} = \frac{\dfrac{1}{1-L^{(2)}}}{\dfrac{1}{l^{(1)}} + \dfrac{1}{1-L^{(2)}}} \tag{64}$$

and

$$\frac{1}{l^{(j)}} = \frac{1}{3}\sum_{i=1,3}\frac{1}{L_i^{(j)}}, \quad j = 1 \text{ or } 2. \tag{65}$$

For spheres we have $p_{1c} = 1/3$; if the material 2 shapes are plate-like while the material 1 shapes are not plate-like then $p_{1c} = 1$, which means that material 1 never percolates in EMA; conversely, if material 1 is needle-like, then 1 always percolates according to EMA. In general, if the material shapes are the same, then p_{1c} is bounded between 0 and 1/3.

The function $F(\varepsilon, \varepsilon_1, 0, p_1)$ follows from the Appendix as:

$$F(\varepsilon, \varepsilon_1, 0, p_1) = \varepsilon^{\beta(p_1)} \prod_{i=1,3} (\varepsilon-\tilde{\varepsilon}_i)^{R_i}, \tag{66}$$

where the R_i are defined by (46) and

$$\beta(p_1) = \frac{p_{1c}\{1-L^{(2)}\}}{p_{1c}-p_1}. \tag{67}$$

Instead of (48), we now have

$$\sum_{i=1,3} R_i = (1-L)-\beta(p_1). \tag{68}$$

Note that $\beta(p_1)$ is negative for $p_1 > p_{1c}$ and positive for $p_1 < p_{1c}$. Also, the result of the process depends upon the shape of material 2 only through the parameter $L^{(2)}$. When $p_1 \ll p_{1c}$, (66) further simplifies to (see Appendix)

$$F(\varepsilon, \varepsilon_1, 0, p_1) \sim \varepsilon^{\{1-L^{(2)}\}}, \quad p_1 \to 0. \tag{69}$$

We shall use this result later.

When both materials are spheres, the cubic equation (62) becomes linear with root

$$\tilde{\varepsilon}_1 = \varepsilon_1(3p_1-1)/2, \tag{70}$$

which is zero at $p_1 = p_{1c} = 1/3$. The function $F(\varepsilon, \varepsilon_1, 0, p_1)$ becomes

$$F(\varepsilon, \varepsilon_1, 0, p_1) = [\varepsilon\{\varepsilon-\varepsilon_1(3p_1-1)/2\}^{-3p_1}]^{\{2/(3(1-3p_1))\}}. \tag{71}$$

Now consider the straight-line process of section 5. Start at (0, 0) with $\varepsilon = \varepsilon_0$ and go to (ϕ_1, ϕ_2) along a straight line. At (ϕ_1, ϕ_2) we have from (54) and (71)

$$\frac{\varepsilon}{\varepsilon_0}\left[\frac{\varepsilon_0-\varepsilon_1(3\phi_1/\phi-1)/2}{\varepsilon-\varepsilon_1(3\phi_1/\phi-1)/2}\right]^{(3\phi_1/\phi)} = [1-\phi]^{\{(3/2)(1-3\phi_1/\phi)\}}. \tag{72}$$

It is clear that the solution to this equation behaves quite differently for $\phi_1/\phi < 1/3$ than for $\phi_1/\phi > 1/3$, especially as $\phi \to 1$. Exactly at $\phi_1/\phi = 1/3$, (72) becomes

$$\frac{1}{2}\left(\frac{\varepsilon_1}{\varepsilon}-\frac{\varepsilon_1}{\varepsilon_0}\right)+\ln\left(\frac{\varepsilon_0}{\varepsilon}\phi_0^{3/2}\right) = 0. \tag{73}$$

In the limit as $\phi \to 1$, ϕ_1 and ϕ_2 become the volume fractions of materials 1 and 2, respectively. The convergence of ε to the solution at $\phi = 1$ can be analysed by taking $\phi_0 \equiv 1-\phi \ll 1$ and solving (72) asymptotically in ϕ_0. As $\phi_0 \to 0$, the "backbone" of original material ε_0 disappears and the solid contains only materials 1 and 2. The asymptotic solution of (72) changes character at $\phi_1/\phi \sim \phi_1 = 1/3$. For $\phi_1 < 1/3$ we have

$$\varepsilon \sim \varepsilon_0[\phi_0]^{\{(3/2)(1-3\phi_1)\}}\left[\frac{1-3\phi_1}{1-3\phi_1+2\varepsilon_0/\varepsilon_1}\right]^{3\phi_1}, \tag{74}$$

while for $\phi_1 > 1/3$ the solution is

$$\varepsilon \sim \varepsilon_1\left(\frac{3\phi_1-1}{2}\right)+[\phi_0]^{\{(3/2)(1-1/3\phi_1)\}}\left[\varepsilon_0-\varepsilon_1\left(\frac{3\phi_1-1}{2}\right)\right]\left[\frac{\varepsilon_1}{\varepsilon_0}\left(\frac{3\phi_1-1}{2}\right)\right]^{(1/3\phi_1)}. \tag{75}$$

At $\phi_1 = 1/3$ we have from (73)

$$\varepsilon \sim \frac{-\varepsilon_1}{3\ln(\phi_0)}. \tag{76}$$

The limiting solution at $\phi_0 = 0$ follows from (74) and (75) as

$$\varepsilon = \begin{bmatrix} 0 & \phi_1 \leqslant 1/3 \\ \varepsilon_1(3\phi_1 - 1)/2, & \phi_1 \geqslant 1/3 \end{bmatrix}. \tag{77}$$

This is the well known Effective Medium Approximation (EMA) solution, or Bruggeman's solution, among various other names (BRUGGEMAN, 1935; LANDAUER, 1952). The physical significance of a zero solution for $\phi_1 < 1/3$ is that material 1 ceases to percolate at this volume fraction. This phenomenon of a percolation threshold at a volume fraction greater than zero is characteristic of EMA. In contrast, DEM is known to have a percolation threshold at zero (YONEZAWA and COHEN, 1983; SHENG and CALLEGARI, 1984). The asymptotic solution (74)–(76) has a zero percolation threshold for $\phi_0 \neq 0$. The solution for $\phi_1 < 1/3$ is similar to Archies' law (SEN *et al.*, 1981), which predicts that $\varepsilon \sim \varepsilon_0 \phi_0^m$ where m is typically of the order of 2 in rocks. We have $m \sim (3/2)(1 - 3\phi_1)$, which is less than 3/2, but approaches 3/2 as $\phi_1 \to 0$. For $\phi_1 > 1/3$, (75) shows the solution to be slightly above the EMA solution. Therefore, the solution overall is very close to the EMA solution, but remains non-zero until $\phi_1 = 0$ and approaches this threshold in an Archie's law fashion.

The above analysis for spheres may be generalized to the case that the materials are ellipsoids. Now the percolation threshold is at p_{1c} defined in (64). For $\phi_1 < p_{1c}$ it follows from the Appendix that ε behaves asymptotically like

$$\varepsilon \sim \tilde{\varepsilon}_0 [\phi_0]^{\{1/\beta(\phi_1)\}}, \quad \phi_0 \to 0, \tag{78}$$

where $\tilde{\varepsilon}_0$ is a function of ϕ_1 that tends to ε_0 as $\phi_1 \to 0$. The latter result follows from (69) and the Appendix. Also, from (67) we see that the exponent in (78) is

$$\frac{1}{\beta(\phi_1)} = \frac{1 - \phi_1/p_{1c}}{1 - L^{(2)}}, \tag{79}$$

which increases from 0 to $1/[1 - L^{(2)}]$, as ϕ_1 decreases from the percolation threshold to zero. As $\phi_1 \to 0$, we have the doubly asymptotic result from (78) and (79) that

$$\varepsilon \sim \varepsilon_0 [\phi_0]^{\{1/(1 - L^{(2)})\}}, \quad \phi_0 \to 0, \quad \phi_1 \to 0. \tag{80}$$

This agrees with Archie's law if the shapes are selected such that $L^{(2)}$ of (62) equals 1/2. Note that (80) is independent of the value of ε_1 and the shape of the material 1 inclusions. The result depends only on the permittivity of the vanishing backbone, ε_0, and on the shape of the material 2 inclusions.

When $\phi_1 > p_{1c}$, the solution for small ϕ_0 lies close to, but slightly above, the corresponding EMA solution. The difference is determined by the exact relation (55) which implies that

$$\varepsilon \sim \varepsilon_E(\phi_1) + O[\phi_0^{(1/R_E)}], \quad \phi_0 \to 0, \quad \phi_1 > p_{1c}. \tag{81}$$

8. NUMERICAL EXAMPLES OF CONDUCTOR–INSULATOR COMPOSITES

Two examples are considered in which the starting material is pure conductor, i.e. $\varepsilon_0 = \varepsilon_1$ and $\phi_0 = 1$ at $t = 0$. The homogenizations are achieved using spherical grains

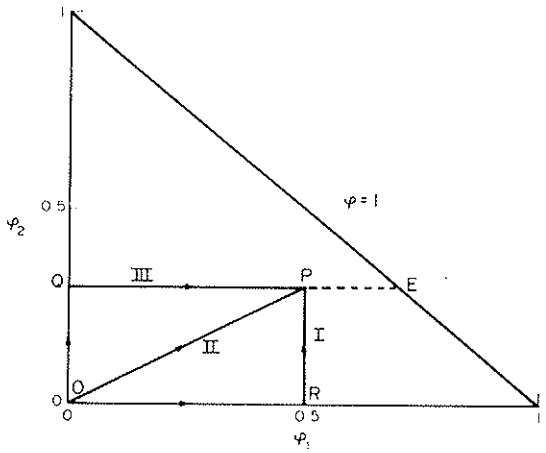

FIG. 2. Three possible paths to the same endpoint $P(\phi_1, \phi_2)$.

for both phases, so that the results of the previous section are relevant. Both of the examples illustrate for given volume fractions of conductor and insulator that different effective conductivities can be realized depending upon the microstructure involved.

The first example considers paths in the (ϕ_1, ϕ_2) plane such that ϕ_1 is specified. The free parameter is ϕ_2 which satisfies $0 \leqslant \phi_2 \leqslant 1 - \phi_1$. Three different paths I, II and III from (0, 0) to the point (ϕ_1, ϕ_2) are considered (see Fig. 2). The homogenization represented by OQ in Fig. 2 is the usual DEM considered by SEN *et al.* (1981) in which the added material is insulator only. This solution follows from (61) with $\varepsilon_0 = \varepsilon_1$ and $\varepsilon_2 = 0$ as

$$\varepsilon = \varepsilon_1(1 - \phi_2)^{3/2}. \tag{82}$$

The first path, I, goes from O to R and then to P. There is no change in ε along OR because the same material (conductor) is put back in as it is taken out. From R to P there is a change in ε because insulator is added. However, conductor is also added between R and P in order to keep the volume fraction ϕ_1 fixed. Note that the total volume fraction of conductor is $\phi_0 + \phi_1$, but that ϕ_0 and ϕ_1 represent distinct regions. In the FVP, as u_2 increases along RP, u_1 must also increase according to (1.3) with $\dot{\phi}_1 = 0$. If we defined ε (P, I) as the value of ε at P due to path I, then we do not expect ε (P, I) to equal ε (Q, III). In fact, we expect ε (P, I) $\leqslant \varepsilon$ (Q, III) because along RP the conductor is thrown back in with the insulator and so some conductor may be entirely surrounded by insulator. The volume fraction of the percolating conductor is $1 - \phi_2$ for ε (Q, III), but is less than this for ε (P, I).

Path II is the straight-line process from O to P. Path III is ordinary DEM (O → Q) followed by the straight-line process Q → P. Note that on all of II and on I between R and P and III between Q and P, both conductor and insulator are added simultaneously.

The results are shown in Fig. 3. The usual DEM result, corresponding to ε (Q, III) and the EMA result (77), corresponding to ε (E) (see Fig. 2) are also plotted for comparison.

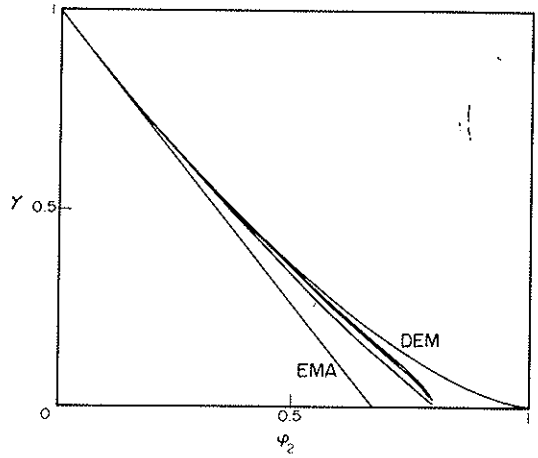

FIG. 3. Plots of $\gamma(\phi_2) = \varepsilon/\varepsilon_1$ for a conductor–insulator composite. The top curve is the usual DEM. The next three curves are for paths I, II and III of Fig. 2 for $\phi_1 = 0.2$. The bottom curve is the EMA result.

We note that ε (E) is independent of how the point E (ϕ_1, ϕ_2) is achieved. The results in Fig. 3 for paths I, II and III are for $\phi_1 \equiv 0.2$. Therefore, the range of ϕ_2 for these paths is 0–0.8. At the upper limit all three paths reduce to the EMA result because as $\phi_2 \to 0.8$, $\phi_0 \to 0$. However, for $\phi_2 < 1-\phi_1$ the results of paths I, II and III are evidently different.

The second example illustrates the approach to EMA as $\phi_0 \to 0$. We fix ϕ_0 and compute ε by the straight-line processes from (0, 0) to (ϕ_1, ϕ_2) shown in Fig. 4. The values of $\gamma = \varepsilon/\varepsilon_1$ is found by solving (72). Results are shown in Fig. 5 for six values of ϕ_0, ranging from 0.3 (top) to 0 (bottom). Note that the range of ϕ_2 is $0 \leqslant \phi_2 \leqslant 1-\phi_0$

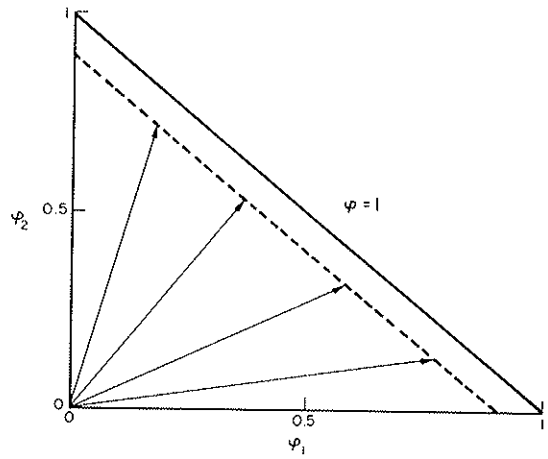

FIG. 4. Straight-line paths to points (ϕ_1, ϕ_2) such that ϕ_0 is constant.

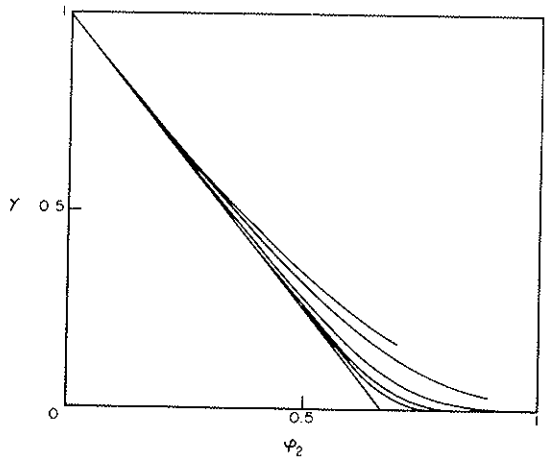

FIG. 5. Results of the straight-line processes of Fig. 4 for different values of ϕ_0. From the top down, the curves are $\phi_0 = 0.3, 0.1, 0.01, 0.001, 0.0001$ and the bottom curve is EMA or $\phi_0 = 0$.

(see Fig. 4). The two distinct forms of solution, (74) and (75), are evident in Fig. 5 for small ϕ_0. It is noteworthy that the curves remain continuous and non-zero at $\phi_1 = 1/3$ even as ϕ_0 gets very small. This is because the approach to EMA at $\phi_1 = 1/3$ is given by (76) which depends upon ϕ_0 only through ln (ϕ_0). The endpoints of the curves in Fig. 5 are at $\phi_2 = 1 - \phi_0$, or equivalently, $\phi_1 = 0$. The solution at the endpoints follows from (72) as

$$\varepsilon = \varepsilon_1 \phi_0^{3/2}, \quad \phi_1 = 0, \tag{83}$$

which is the same as the ordinary DEM result of (82) at the same value of ϕ_2.

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APPENDIX

Some results for $\varepsilon_2 = 0$

In this Appendix the function $F(\varepsilon, \varepsilon_1, \varepsilon_2, p_1)$ of (42), (22) and (18) is derived for the special case of $\varepsilon_2 = 0$. First we let $\varepsilon_2 = \delta$, where $\delta = o(\varepsilon_1)$, then we will take the limit of $\delta \to 0$. With $\delta > 0$, the six non-zero roots of $G = 0$, where G is defined in (44), split up into two sets of three. First there are three $O(1)$ roots determined by (62). Denote these as $\tilde{\varepsilon}_i$, $i = 1, 3$. Next there are three $o(1)$ roots, $\tilde{\varepsilon}_i$, $i = 4, 6$, given by $\varepsilon = c\delta$, where c solves the cubic.

$$\frac{-p_1}{l^{(1)}} + \frac{1}{3}\sum_{i=1,3}\frac{p_2(c-1)}{\{1-L_i^{(2)}\}c + L_i^{(2)}} = 0 \tag{A.1}$$

and $l^{(1)}$ is defined in (65). The algebraic equations (62) and (A.1) arise from the different behavior of $G(\varepsilon)$ in the $O(1)$, or "outer" region, and the $O(\delta)$, or "inner" region.

Now consider the general result of (45). As $\delta \to 0$, this reduces to

$$F(\varepsilon, \varepsilon_1, 0, p_1) = \varepsilon^{(R_4+R_5+R_6-l)}\prod_{i=1,3}(\varepsilon-\tilde{\varepsilon}_i)^{R_i}. \tag{A.2}$$

Therefore it is not necessary to know the $O(\delta)$ roots explicitly. The only thing required is the quantity $(R_4+R_5+R_6-l)$. The latter follows from the "inner" expansion of $G(\varepsilon)$, i.e. (A.1), and Cauchy's residue theorem. We readily obtain

$$R_4+R_5+R_6-l = \frac{1}{\dfrac{p_2}{1-L^{(2)}} - \dfrac{p_1}{l^{(1)}}} = \beta(p_1), \tag{A.3}$$

where $\beta(p_1)$ is defined in (67), and (68) follows as a result of (48) and (A.3).

We now examine the limit of (66) as $p_1 \to 0$. From (62) it follows that the three roots $\tilde{\varepsilon}_i$, $i = 1, 3$ are

$$\tilde{\varepsilon}_i = \frac{-\varepsilon_1 L_i^{(1)}}{1-L_i^{(1)}} + p_1\varepsilon_1\left[\frac{1-L}{1-L_i^{(1)}}\right] + O(p_1^2), \tag{A.4}$$

where L is defined in (49). Using (A.4) to compute the residues R_i, $i = 1, 3$ of (46), we readily find that

$$R_i = 3p_1(1-L)^2\varepsilon_1/\tilde{\varepsilon}_i + O(p_1^2), \quad i = 1, 3. \tag{A.5}$$

Therefore, (66) reduces to

$$F(\varepsilon, \varepsilon_1, 0, p_1) \sim \varepsilon^{\{1-L^{(2)}\}} \tag{A.6}$$

when $p_1 \ll p_{1c}$, which is the same as (69).

The general result for straight-line processes becomes for $\varepsilon_2 = 0$, using (66),

$$\varepsilon = \varepsilon_0[\phi_0]^{\{1/\beta(p_1)\}} \prod_{i=1,3} \left[\frac{\varepsilon_0 - \tilde{\varepsilon}_i}{\varepsilon - \tilde{\varepsilon}_i}\right]^{R_i/\beta(p_1)}. \tag{A.7}$$

Now consider this exact result in the limit as $\phi_0 \to 0$. For $\phi_1 < p_{1c}$ the three roots $\tilde{\varepsilon}_i$, $i = 1, 3$ of (62) are all non-zero and negative. The solution to (A.7) is then given approximately as

$$\varepsilon \sim \tilde{\varepsilon}_0[\phi_0]^{\{1/\beta(\phi_1)\}}, \quad \phi_0 \to 0, \tag{A.8}$$

where

$$\tilde{\varepsilon}_0 = \varepsilon_0 \left[\prod_{i=1,3} (1 - \varepsilon_0/\tilde{\varepsilon}_i)^{R_i}\right]^{1/\beta(\phi_1)}. \tag{A.9}$$

If we consider the additional limit of $\phi_1 \to 0$, then the results of (A.5) and (A.6) show that

$$\tilde{\varepsilon}_0 \to \varepsilon_0, \quad \phi_1 \to 0. \tag{A.10}$$