



## The effective medium approximations: Some recent developments

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We provide a brief and highly selective review of the Landauer–Bruggeman effective-medium approximation as applied to random composite media. We first discuss this approximation as applied to linear composites, i.e., to those materials in which there is a linear relation between a curl-free electric field and a divergence-free current density. We then describe extensions of this approach to random composites with cubic nonlinearities in addition to a dominant linear term, and to composites in which the components have a fluctuating conductivity ('conductivity noise'). Finally, we mention one novel application: to conductivity noise in a random composite of normal metal ( $N$ ) and perfect conductor ( $S$ ). It is shown that the extension of the EMA leads to a prediction of a frequency range in which the conductivity noise has a  $1/\omega$  frequency dependence, even if the noise in the individual components is frequency independent.

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### 1. Introduction

This paper gives a brief review of the effective-medium approximation in the theory of inhomogeneous materials. The EMA was first proposed by Bruggeman [1], and then, in a different context, by Landauer [2]. Since its invention, it has been the basis for a vast number of studies of macroscopically inhomogeneous media, and has been generalized by numerous authors to treat a wide variety of problems. As more and more composite microstructures are developed on an ever smaller scale, it is clear that this type of approach will become even more valuable in the future than it has in the recent past.

### 2. Effective-medium approximation for linear media

The effective-medium approximation is a method of treating a *macroscopically inhomogeneous medium*, i.e., a medium in which quantities such as the conductivity  $\sigma$ , dielectric function  $\epsilon$ , or elastic modulus vary in space. Many materials fall into this broad category. One example is a metal–dielectric composite, consisting of a collection of metallic and dielectric grains arranged in some ordered or random fashion. Another example is a porous rock. If that rock is filled with salt water, then it is also a composite of an electrical insulator (the rock matrix) and an electrical conductor (the salt water) [3]. Yet a third example is a polycrystalline sample of an anisotropic material. Each grain in that polycrystal is, in essence, a different material, since it has a different conductivity or dielectric tensor [4]. (Of course, the tensors in the different grains are generally related by a similarity transformation). A polycrystalline elastic material is a more complicated version of the same problem, since each grain has a different *fourth-rank* tensor.

We begin by describing the EMA in the simplest case, as envisioned by Landauer and Bruggeman: a random mixture of two types of grains, which we denote A and B, present in relative volume fractions  $p$  and  $1 - p$ , and characterized by different scalar conductivities  $\sigma_A$  and  $\sigma_B$ . We desire to calculate the effective conductivity  $\sigma_e$  of this composite. To accomplish this, we imagine that each grain, instead of being embedded in its actual random environment, is instead immersed in a homogeneous *effective medium* of conductivity  $\sigma_e$  which will be determined self-consistently. Further, we approximate each grain as having a spherical shape. This may not be unreasonable if the grains are reasonably compact in shape. Then the electric field inside grain  $i$  is uniform and given by an elementary result of electrostatics:

$$E_{in} = E_0 \frac{3\sigma_e}{\sigma_i + 2\sigma_e}, \quad (1)$$

where  $\sigma_i = \sigma_A$  or  $\sigma_B$  and  $E_0$  is the electric field far from the grain.

The self-consistency condition required by the EMA is that the average electric field within the grain shall equal  $E_0$ , or equivalently

$$pE_0 \frac{3\sigma_e}{\sigma_A + 2\sigma_e} + (1 - p)E_0 \frac{3\sigma_e}{\sigma_B + 2\sigma_e} = E_0, \quad (2)$$

The common factor of  $E_0$  may be divided out, and this equation simplified to the well-known form

$$p \frac{\sigma_A - \sigma_e}{\sigma_A + 2\sigma_e} + (1 - p) \frac{\sigma_B - \sigma_e}{\sigma_B + 2\sigma_e} = 0. \quad (3)$$

This quadratic equation has two solutions. Of these, one can generally be identified as the physically sensible solution; the other is typically negative, or has other unphysical features such as a discontinuous variation with the volume fraction  $p$ , or incorrect limiting behavior as  $p \rightarrow 1$  or  $p \rightarrow 0$ .

The EMA is of particular interest in two special cases. One is the limit  $\sigma_B = 0$ ,  $\sigma_A$  finite. This corresponds to a metal-insulator composite. The other limit is  $\sigma_B$  finite,  $\sigma_A \rightarrow \infty$ , corresponding to a composite of normal metal and perfect conductor. (We do not say 'superconductor' because the magnetic properties of component 1 are unspecified.) In these cases, as first pointed out by Landauer [5]  $\sigma_e$  is predicted by the EMA to have singular properties near a percolation threshold at  $p = p_c \equiv \frac{1}{3}$ . In the first case, we readily find that

$$\sigma_e = 0 \quad p < p_c \quad (4)$$

$$\sigma_e = \frac{3}{2}\sigma_A(p - p_c) \quad p > p_c. \quad (5)$$

In the second case,

$$\sigma_e = \frac{1}{3}\sigma_B(p_c - p)^{-1} \quad p < p_c \quad (6)$$

$$\sigma_e = \infty \quad p > p_c. \quad (7)$$

The special concentration  $p_c = \frac{1}{3}$  has a natural physical interpretation: it is the 'connectivity threshold,' above which there is a connected path of material 1 extending throughout the sample. The EMA predicts, the three-dimensional sample and approximately spherical grains, that  $p_c = \frac{1}{3}$ . In general, in  $d$  dimensions, and compact grains, it is found that the EMA predicts  $p_c = \frac{1}{d}$ . For other grain shapes, or for a distribution of shapes, the EMA predictions can vary widely [6].

As specified so far, the EMA applies only at zero frequency. It is not, however, restricted only to the electrical conductivity of an inhomogeneous conductor. In fact, it applies to any effective transport coefficient connecting a curl-free driving field to a divergence-free current density. In the case of electrical conductivity, these quantities are the electric field and the electrical-current density, but numerous other mathematically equivalent cases can be written down. For example, a formally identical equation describes the thermal conductivity of an inhomogeneous medium. In this case, the driving field is the negative of the temperature gradient, and the divergence-free quantity is the heat-current density. For an inhomogeneous magnetic material,

the curl-free quantity is the magnetic field  $\mathbf{H}$  (assuming no free electrical current density  $\mathbf{J}$ ), and the divergence-free ‘current density’ is the magnetic induction  $\mathbf{B}$ . The effective transport coefficient in this case is the effective magnetic permeability  $\mu$ .

The EMA can also be generalized to finite frequencies, provided that the relevant fields are of sufficiently long wavelengths. The wavelength condition is generally that the size of the inhomogeneity be small relative to the wavelength of the electric field in the composite medium. The physical requirement is that the electric fields and displacements should be approximately uniform within any given grain of the inhomogeneous medium. As has been noted by several authors (see, for example, [7]), this condition is not adequate in certain special cases, such as the calculation of far infrared absorption by small metallic grains embedded in a dielectric host; but it is quite adequately satisfied in many composite materials, even in the visible regime, provided that the grains are no larger than 100–200 Å.

To accomplish the finite-frequency generalization, one describes the components in terms of *complex dielectric functions*  $\epsilon_A(\omega)$  and  $\epsilon_B(\omega)$ . The dielectric functions include the free-current densities in the usual way:

$$\epsilon_j(\omega) = \epsilon_{jb}(\omega) + \frac{4\pi i \sigma_j(\omega)}{\omega}, \quad (8)$$

$j = A, B$ . Here  $\epsilon_{jb}$  is the contribution to the dielectric function of the  $j$ th component due to bound charge, and  $\sigma_j(\omega)$  is the conductivity arising from free charges. In general, both  $\epsilon_{jb}(\omega)$  and  $\sigma_j(\omega)$  will be complex quantities, as will  $\epsilon_j(\omega)$ . In the  $j$ th component, the electric field  $\mathbf{E}$  and displacement  $\mathbf{D}$  are related by

$$\mathbf{D}(\omega) = \epsilon_j(\omega)\mathbf{E}(\omega). \quad (9)$$

Since  $\nabla \times \mathbf{E} = 0$  in the quasistatic approximation and  $\nabla \cdot \mathbf{D} = 0$ , it follows that exactly the same equations used to obtain  $\sigma_e$  at zero frequency can be applied to find  $\epsilon_e$  at finite frequencies. In other words,  $\epsilon_e$  satisfies

$$p \frac{\epsilon_e - \epsilon_A}{\epsilon_A + 2\epsilon_e} + (1 - p) \frac{\epsilon_e - \epsilon_B}{\epsilon_B + 2\epsilon_e} = 0. \quad (10)$$

In a metal–insulator composite, the EMA has a variety of striking predictions. If one takes for the metallic component the Drude form  $\epsilon_A = 1 - \omega_p^2/[\omega(\omega + i/\tau)]$ , where  $\omega_p$  is the plasma frequency and  $\tau$  a relaxation time, and for the insulating component simply  $\epsilon_B = 1$ , then  $\epsilon_e(\omega)$ , in the EMA, is found to behave as follows.

1. Above the percolation threshold, there is a Drude peak in  $\epsilon_e$ , whose integrated strength is approximately equal to the zero-frequency effective conductivity  $\sigma_e(0) = \text{Lim}_{\omega \rightarrow 0}(\omega \text{Im}\epsilon_e(\omega)/(4\pi))$ .
2.  $\text{Im}\epsilon_e(\omega)$  shows a broad ‘surface plasmon’ band in the range  $0 < \omega < \omega_p$ . This band corresponds to mostly localized impurity modes in the composite, which represent oscillations of the electron gas in clusters of small metal particles, or in the vicinity of voids in the composite (‘small particle resonances’ or ‘void resonances’). These modes are spread over a broad frequency range.

It is often difficult to make a direct comparison between these predictions and experiment, since real materials hardly ever perfectly satisfy the geometrical expectations of the EMA in its simplest form (symmetric arrangement of the two components in the form of compact, nearly spherical grains, with no short-range order—i.e., no excess or deficit of neighbors of type A around grains of type A, above or below that expected in a random composite). Nevertheless, one can compare EMA predictions for a random impedance network with the calculated effective impedance obtained by direct numerical solution of the impedance network. A number of such calculations have been carried out [8, 9] in both two and three dimensions (2D and 3D). The results show that the EMA is in excellent agreement with computer ‘experiment.’ In particular, the broad surface plasmon band predicted by the EMA is very similar to that found in the computer results, as a function of concentration. The main discrepancies arise near  $p_c$ . These are not surprising, since it is known that the EMA, being a mean-field approximation, must give incorrect power-law relations between  $\sigma_e$  and  $|p - p_c|$

near the percolation threshold. The EMA also gives a percolation threshold which is a little too high in 3D, though it happens to be exact in 2D [10].

### 3. EMA-like approximations for nonlinear composites

Up to now, we have discussed the application of the EMA to *linear* transport problems in composite media. Of equal interest is *nonlinear* transport, i.e., transport in which there is a nonlinear relationship between fields and current densities. The range of such problems is enormous. Moreover, they are becoming of ever greater importance in practice, because the fields and current densities in typical submicron devices can be very large. Thus, even though nonlinear problems are much more difficult than linear ones, they cannot be ignored. In this section, we briefly sketch one approach to one such problem—cubic nonlinearity—and show how an effective-medium approximation can be developed in this case. The EMA for nonlinear problems is on a less firm footing than in the linear case; but it leads to some remarkable predictions, one of which we will describe below.

For clarity, we describe the nonlinear problem in its simplest form (for a more detailed description, see Stroud and Hui [11]). Thus, we assume that the current density  $\mathbf{J}$  and electric field  $\mathbf{E}$  are related by

$$\mathbf{J}(\mathbf{x}) = \sigma(\mathbf{x})\mathbf{E}(\mathbf{x}) + a(\mathbf{x})|\mathbf{E}(\mathbf{x})|^2\mathbf{E} \quad (11)$$

where  $\sigma(\mathbf{x})$  and  $a(\mathbf{x})$  are respectively the linear conductivity and cubic nonlinear susceptibility the composite at point  $\mathbf{x}$ . We assume that these quantities take on the values  $\sigma_A, a_A$ , and  $\sigma_B, a_B$  if point  $\mathbf{x}$  is in component A or component B, respectively. In order to define an effective conductivity  $\sigma_e$  and effective cubic nonlinear susceptibility  $a_e$ , Stroud and Hui [11] assumed that the composite consisted of a volume  $V$ , surrounded by a surface  $S$  on which the electrostatic potential was fixed at  $\Phi(\mathbf{x}) = \vec{E}_0 \cdot \vec{x}$  (corresponding to an applied electric field  $\mathbf{E}_0$ ).  $\sigma_e$  and  $a_e$  were then shown to be given by

$$\sigma_e E_0^2 = \frac{1}{V} \int d\mathbf{x} \sigma(\mathbf{x}) |\mathbf{E}_{lin}(\mathbf{x})|^2 \quad (12)$$

and

$$a_e E_0^4 = \frac{1}{V} \int d\mathbf{x} a(\mathbf{x}) |\mathbf{E}_{lin}(\mathbf{x})|^4, \quad (13)$$

where the subscript *lin* denotes that the electric field is to be calculated in the related *linear* medium, in which  $a(\mathbf{x}) = 0$ . Thus, the cubic nonlinear properties of the composite depend on (i) the cubic nonlinear susceptibility  $a(\mathbf{x})$  in the actual composite geometry; and (ii) the electric field  $\mathbf{E}_{lin}(\mathbf{x})$  in the related *linear* medium.

Closely related to the effective nonlinear susceptibility of a composite is the *conductivity noise*. To define this noise, assume that the composite is now linear, but that the conductivity  $\sigma(\mathbf{x})$  is fluctuating. Specifically, we imagine that the actual composite is a member of a large ensemble of composites, each of which has the same geometry (i.e. spatial distribution of the two components A and B), but different conductivities. The conductivity in a particular member of this ensemble is  $\sigma(\mathbf{x}) + \delta\sigma(\mathbf{x})$ . We assume that the ensemble-averaged conductivity fluctuations can be written as

$$\langle (\delta\sigma(\mathbf{x})\delta\sigma(\mathbf{x}'))_{ens} \rangle = \lambda(\mathbf{x})\delta(\mathbf{x} - \mathbf{x}'), \quad (14)$$

where the brackets here refer to an average over an ensemble of composites, all of the same geometry, at a particular point  $\mathbf{x}$ . In general, we expect  $\lambda(\mathbf{x})$  to take on one of the two values  $\lambda_A$  and  $\lambda_B$ , depending on whether  $\mathbf{x}$  lies in medium A or medium B.

Because of the nonzero local fluctuations  $\delta\sigma(\mathbf{x})$ , the effective conductivity of this composite will also vary from one member of the ensemble to another. We write the effective conductivity for a particular member of the ensemble as  $\sigma_e + \delta\sigma_e$ . Stroud and Hui [11] showed that there was an exact connection between the

ensemble-averaged mean-square conductivity fluctuations,  $\langle(\delta\sigma_e)^2\rangle_{ens}$ , and the fourth moment of the electric field  $E_{lin}(\mathbf{x})$  in the related *linear* medium. This connection is

$$\langle(\delta\sigma_e)^2\rangle_{ens} E_0^4 = \frac{1}{V^2} \int d\mathbf{x} \lambda(\mathbf{x}) |E_{lin}(\mathbf{x})|^4, \quad (15)$$

where  $E_0$  is once again the applied electric field.

Thus both  $a_e$  and  $\langle(\delta\sigma_e)^2\rangle_{ens}$  depend on the space average of  $|\mathbf{E}|^4$  in each component of the related *linear* medium. There are several possible ways one might attempt to compute this quantity. One approach would be to attempt a direct calculation of this average. Another possibility is to carry out the averaging within some simple approximation. Such an approximation was first suggested by Zeng *et al.* [12] and by Stroud and Hui [11], and later at finite frequencies by Stroud and Wood [13], who called this approach a ‘nonlinear decoupling approximation’ (NDA). The finite frequency problem is of great interest in the context of nonlinear optical properties of composite materials (see, e.g., [14]). The NDA at zero frequency may be written simply

$$\langle|\mathbf{E}|^4\rangle_{lin,i} \sim \langle|\mathbf{E}|^2\rangle_{lin,i}^2, \quad (16)$$

where  $\langle \dots \rangle_{lin,i}$  refers to an average of the quantity in brackets, for a linear medium, within the  $i$ th component. With this decoupling, a simple approximation can be written down for  $a_e$  using an exact relationship for  $\langle|\mathbf{E}|^2\rangle_{lin,i}$  noted by Bergman [15]:

$$\langle|\mathbf{E}|^2\rangle_{lin,i} = \left( \frac{\partial\sigma_e}{\partial\sigma_i} \right)_{\sigma_j, j \neq i} E_0^2. \quad (17)$$

That is, the desired mean-square electric field is related to a partial derivative of the effective conductivity with respect to the conductivity of one of the composites. (This result actually applies to an  $n$ -component composite, not just a two-component one.) With this identification, we may write [11, 13]

$$a_e = \sum_i \frac{1}{p_i} a_i \left( \frac{\partial\sigma_e}{\partial\sigma_i} \right)_{\sigma_j, j \neq i}^2, \quad (18)$$

where the sum is to be carried out over the components  $i$  of the composite, which are present in volume fraction  $p_i$  (this result is not necessarily restricted to a two-component composite). The analogous expression for the conductivity noise is

$$\langle(\delta\sigma_e)^2\rangle_{ens} = \frac{1}{V} \sum_i \frac{\lambda_i}{p_i} \left( \frac{\partial\sigma_e}{\partial\sigma_i} \right)^2. \quad (19)$$

It is straightforward to generalize this procedure to finite frequencies. For example, the mean-square fluctuations in the effective conductivity are given, within the NDA, by

$$\langle(\delta\sigma_e)^2\rangle_{ens} = \frac{1}{V} \sum_i \frac{\lambda_i}{p_i} \frac{\partial\sigma_e}{\partial\sigma_i} \left| \frac{\partial\sigma_e}{\partial\sigma_i} \right|. \quad (20)$$

In general, at finite frequencies, both  $\sigma_e$  and the individual conductivities  $\sigma_i$  will be complex and frequency dependent; the proper expression for the mean-square fluctuations takes this into account via the absolute values as shown.

This result is still not entirely explicit. In order to complete the specification of either  $a_e$  (or  $\langle(\delta\sigma_e)^2\rangle$ ), one must specify an approximation for  $\sigma_e$ . The most obvious and convenient is, of course, the EMA itself. With this choice, one has a simple, fully explicit means of estimating  $a_e$  or  $\langle(\delta\sigma_e)^2\rangle$  in a composite medium, given the properties of the individual components.

As an illustration, we consider the behavior of conductivity fluctuations in a composite of normal metal ( $N$ ) and perfect conductor ( $S$ ). The normal metal is assumed to have a constant conductivity  $\sigma_N$ , while the  $S$  component is taken to have a purely *inductive* response,  $\sigma_S = \frac{i\gamma}{\omega}$ , where  $\gamma$  is the so-called inverse kinetic

inductance of the  $S$  component. We will consider the noise only exactly at the percolation threshold  $p_c$  for the *perfectly conducting* ( $S$ ) component. Then, in 2D, the effective EMA conductivity takes the form

$$\sigma_e = \sigma_N^{1/2} \sigma_S^{1/2}, \quad (21)$$

while in 3D, it becomes

$$\sigma_e = \frac{1}{4} \left( \sigma_N + \sqrt{\sigma_N^2 + 8\sigma_N\sigma_S} \right). \quad (22)$$

In the limit  $|\sigma_N/\sigma_S| \ll 1$ , the latter expression reduces approximately to

$$\sigma_e \sim \sqrt{\frac{\sigma_N\sigma_S}{2}}. \quad (23)$$

Now we assume that only the  $N$  component has any fluctuations in the conductivity, i.e., that  $\lambda(\mathbf{x})$  vanishes unless  $\mathbf{x}$  lies within the  $N$  component. We also assume that  $\lambda_N$  is frequency independent. Then according to eqn (20), the *effective* noise coefficient  $\langle(\delta\sigma_e)^2\rangle_{ens}$  takes the form

$$\langle(\delta\sigma_e)^2\rangle_{ens} = \frac{\lambda_N}{2V} \left( \frac{\sigma_S}{\sigma_N} \left| \frac{\sigma_S}{\sigma_N} \right| \right)^{1/2}. \quad (24)$$

in 2D; and

$$\langle(\delta\sigma_e)^2\rangle_{ens} = \frac{\lambda_N}{6V} \left( \frac{\sigma_S}{\sigma_N} \left| \frac{\sigma_S}{\sigma_N} \right| \right)^{1/2} \quad (25)$$

in 3D. Here we have used the fact that the volume fraction of normal metal at which the  $S$  component first percolates, in the EMA, is  $p_c = \frac{1}{2}$  in 2D and  $p_c = \frac{2}{3}$  in 3D.

Since the ratio  $\sigma_S/\sigma_N$  varies as  $1/\omega$  in absolute value, eqns (24) and (25) predict that, precisely at percolation, conductivity fluctuations in the composite should vary in frequency precisely as  $1/\omega$ . In other words, the inhomogeneity of the  $SN$  composite is predicted to produce a  $1/\omega$  conductivity noise, even if the noise in the individual components is *entirely frequency independent*. This is potentially a novel source of  $1/\omega$  noise in granular materials.

We should point out several caveats in this derivation. First, the derivation is an approximation, based on a decoupling approximation which may or may not be accurate near the percolation threshold. Secondly, it remains to be seen whether this noise source, even if accurately  $1/\omega$ , would be experimentally significant in the very-low-frequency range where such noise is usually found [16].

What happens away from the percolation threshold (that is, the regime where the composite is electrically still a normal metal)? We have carried out preliminary numerical studies in this regime. The conclusion is that, for any composition below the percolation threshold for the  $S$  component, the  $1/\omega$  conductivity noise flattens and becomes white noise below a characteristic crossover frequency  $\omega_X$ .  $\omega_X$  itself vanishes, as a power law in  $|p - p_c|$ , as the percolation threshold is approached. Finally, the *relative magnitude* of the noise (that is, of  $V\langle(\delta\sigma_e)^2\rangle_{ens}/\lambda_N$ ) diverges as a power law in  $1/|p - p_c|$ . This behavior is reminiscent of a critical phenomenon, in which  $p$  plays the role of a temperature and  $p_c$ , that of a critical temperature. This analogy deserves further exploration, as does the connection, if any, between the predicted  $1/\omega$  frequency dependence to that observed in many granular materials.

#### 4. Summary

We have given a very brief and selective review of some recent developments in the effective-medium approach to composite media, which was pioneered by Rolf Landauer. There are many additional developments which are just opening up now—questions involving frequency-doubling, frequency-tripling, and optical bistability in composite materials—for all of which Rolf's original ideas stand as an excellent starting point.

The problems in this field remain as challenging scientifically as ever, and are only increasing in technological importance. Thus, further developments in this field are very likely in the next few years.

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