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EFFECTIVE LINEAR RESPONSE IN STRONGLY HETEROGENEOUS MEDIA—SELF-CONSISTENT APPROACH

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Abstract—The present paper is concerned with the effective (or mean-field) response to a non-stochastic source in heterogeneous media. A self-consistent method is proposed which is analogous to the well known CPA. It permits to determine the non-locality of effective properties or, if the Fourier transforms are considered, their dependence on the wave vector k. The method is applied to the electrostatic field in a heterogeneous body consisting of nearly spherical grains and an equation determining the effective dielectricity is obtained. The results are also valid for other parameters which are mathematically equivalent, like conductivity and permeability. Numerical computations are carried out for a two-component mixture. They show that in strongly heterogeneous media a non-locality of very long range can occur.

1. INTRODUCTION

During the last decade considerable attention has been paid to the study of heterogeneous media, e.g. [1, 2]. By a heterogeneous medium we understand a material whose properties are random functions of position. In this paper we are dealing with a field $u(\mathbf{r})$ related to certain sources $\rho(\mathbf{r})$ by means of a linear equation

$$Lu(\mathbf{r}) = \rho(\mathbf{r}) \tag{1.1}$$

where the linear operator L contains stochastically varying material parameters $c(\mathbf{r})$ which give rise to a random space dependence. In the special case of electrostatics $u(\mathbf{r})$ and $\rho(\mathbf{r})$ are the potential and the charge density, respectively and eqn (1.1) takes the form

$$L = -\frac{\partial}{\partial \mathbf{r}} \cdot \epsilon(\mathbf{r}) \frac{\partial}{\partial \mathbf{r}} \tag{1.2}$$

where ϵ is the dielectric parameter.

In order to describe the macroscopic behaviour of the considered heterogeneous body, we do not need the field $u(\mathbf{r})$ itself, which exhibits random fluctuations, but rather some of its averages. In the simplest case we may ask for the mean field $\langle u(\mathbf{r})\rangle$. The average has to be understood as an ensemble average over many samples which are produced in the same manufacturing process, but exhibit different fluctuations.

In general, the source distribution ρ may also be random. But in the present paper we shall confine ourselves to the special case of non-stochastic sources being the same for all samples

$$\rho = \langle \rho \rangle. \tag{1.3}$$

This has been done in the majority of papers on this subject. A subsequent paper will be devoted to the problem of random sources in heterogeneous media.

Our aim is to derive an equation determining the mean field $\langle u(\mathbf{r})\rangle$. We start with the formal solution of eqn (1.1)

$$u(\mathbf{r}) = \int \mathrm{d}\mathbf{r}'\, g_{st}(\mathbf{r}, \mathbf{r}')\rho(\mathbf{r}') \tag{1.4}$$

where g_{st} is the stochastic Green function of the operator L belonging to a single sample. In order to define the Green function unambiguously, boundary conditions are in need. Complications caused by boundary effects can be eluded in considering an infinite body in connection with a field vanishing at infinity. Accordingly we choose that retarded Green function which tends to zero at large distances. Taking into account (1.3) and the definition

$$\langle g_{st}\rangle \equiv g \tag{1.5}$$

we obtain from (1.4)

$$\langle u(\mathbf{r})\rangle = \int d\mathbf{r}'\, g(\mathbf{r},\mathbf{r}')\rho(\mathbf{r}'). \tag{1.6}$$

It is useful to associate an operator L_{eff} to the average—or effective—Green function by

$$L_{\text{eff}}\, g(\mathbf{r},\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}'). \tag{1.7}$$

Applying the effective operator L_{eff} to (1.6), we get the desired equation for the mean field in a formal manner

$$L_{\text{eff}}\langle u\rangle = \rho. \tag{1.8}$$

If statistical homogeneity is assumed, as will be done in the following, the effective Green function depends only on the difference coordinate

$$g = g(\mathbf{r}-\mathbf{r}'). \tag{1.9}$$

Consequently, a Fourier transformation of the space variables reduces the convolution integral (1.6) to a simple product

$$\langle \tilde{u}(\mathbf{k})\rangle = \tilde{g}(\mathbf{k})\tilde{\rho}(\mathbf{k}) \tag{1.10}$$

where $\mathbf{k}$ is the wave vector. For the sake of clarity, quantities in the Fourier space are marked by a tilda. In this representation the transition to (1.8) is easily achieved by an ordinary division

$$\tilde{L}_{\text{eff}}(\mathbf{k}) = \tilde{g}(\mathbf{k})^{-1}, \quad \tilde{L}_{\text{eff}}(\mathbf{k})\langle \tilde{u}(\mathbf{k})\rangle = \tilde{\rho}(\mathbf{k}). \tag{1.11}$$

Turning again to electrostatics, we can introduce the notion of effective dielectricity

$$\tilde{L}_{\text{eff}}(\mathbf{k}) = \mathbf{k}\,.\,\tilde{\epsilon}_{\text{eff}}(\mathbf{k})\mathbf{k} \tag{1.12}$$

which relates the mean electric field $\langle \mathbf{E}\rangle$ to the mean electric displacement $\langle \mathbf{D}\rangle$ according to

$$\langle \tilde{\mathbf{D}}(\mathbf{k})\rangle = \tilde{\epsilon}_{\text{eff}}(\mathbf{k})\langle \tilde{\mathbf{E}}(\mathbf{k})\rangle. \tag{1.13}$$

Since, in general, $\tilde{\epsilon}_{\text{eff}}$ turns out to be a function of the wave vector $\mathbf{k}$, the transformation into the physical space furnishes non-local relations

$$\begin{gathered}\langle \mathbf{D}(\mathbf{r})\rangle = \int d\mathbf{r}'\, \epsilon_{\text{eff}}(\mathbf{r}-\mathbf{r}')\langle \mathbf{E}(\mathbf{r}')\rangle \\ L_{\text{eff}}\langle u(\mathbf{r})\rangle = -\frac{\partial}{\partial \mathbf{r}}\cdot \int d\mathbf{r}'\, \epsilon_{\text{eff}}(\mathbf{r}-\mathbf{r}')\frac{\partial}{\partial \mathbf{r}'}\langle u(\mathbf{r}')\rangle.\end{gathered} \tag{1.14}$$

This non-locality will be calculated by means of a self-consistent method which is appropriate to the case of strongly heterogeneous materials when perturbation techniques fail.

2. SELF-CONSISTENT APPROACH TO L_{eff}

Let us consider a medium consisting of a great number of grains which are distributed at random with respect to position, size and material properties. Inside the grain No. i the material parameter $c(\mathbf{r})$ takes the constant value c_i. Then, at an arbitrary point, it may be written as

$$c(\mathbf{r}) = \sum_i c_i\theta_i(\mathbf{r}). \tag{2.1}$$

Here θ_i is a step function defined by

$$\theta_i(\mathbf{r}) = \begin{cases} 1 & \text{inside the } i\text{th grain} \\ 0 & \text{otherwise} \end{cases}, \sum_i \theta_i(\mathbf{r}) = 1. \tag{2.2}$$

The second equation expresses the fact that each point $\mathbf{r}$ belongs to exactly one of the grains. Furthermore, we suppose that correlations between the properties of adjacent grains can be disregarded.

If the non-random source takes a harmonic form

$$\begin{aligned} \rho(\mathbf{r}) &= \rho_0 \exp i\mathbf{k}\,.\,\mathbf{r} \\ \tilde{\rho}(\mathbf{k}') &= \rho_0 \delta(\mathbf{k}-\mathbf{k}') \end{aligned} \tag{2.3}$$

it follows from (1.11) that

$$\begin{aligned} \langle u(\mathbf{r})\rangle &= u_0 \exp i\mathbf{k}\,.\,\mathbf{r} \\ u_0 &= \tilde{L}_{\text{eff}}^{-1}(\mathbf{k})\rho_0, \end{aligned} \tag{2.4}$$

Comparison with (1.8) shows that application of the operator L_{eff} to a harmonic function amounts to multiplication with the c-number $\tilde{L}_{\text{eff}}(\mathbf{k})$. There is no loss of generality, if we restrict our considerations to the special source term (2.3), since an arbitrary source distribution can always be generated by a superposition of harmonic functions. Because of the linearity of eqns (1.1) and (1.8) the corresponding fields u and $\langle u\rangle$ are easily obtained from the superposition of those belonging to harmonic sources.

In order to determine L_{eff}, the stochastic operator L will be decomposed into a homogeneous part L_0 and deviations L_i due to the ith grain

$$L = L_0 + \sum_i L_i. \tag{2.5}$$

L_0 is required to give the same results as L_{eff}, if applied to the harmonic function $\langle u\rangle$ (2.4), i.e.

$$L_0\langle u\rangle = L_{\text{eff}}\langle u\rangle = \tilde{L}_{\text{eff}}(\mathbf{k})\langle u\rangle = \rho \tag{2.6}$$

whereas L_i depends only on the properties of the ith grain and the effective properties contained in L_0. Similarly, we split up the stochastic field u into the mean field $\langle u\rangle$ and fluctuations u_i due to the presence of the ith grain

$$u = \langle u\rangle + \sum_i u_i \tag{2.7}$$

where the fluctuation u_i may be defined by the equation

$$Lu_i = -L_i\langle u\rangle. \tag{2.8}$$

Up to this state the set of eqns (2.5)–(2.8) is exactly equivalent to the basic eqn (1.1). But to find a solution of eqn (2.8) is, of course, as impossible as to solve eqn (1.1). So we are forced to introduce a decisive approximation.

Rewriting (2.8) in the form

$$(L_0 + L_i)u_i = -L_i\langle u\rangle - \sum_{j(\neq i)} L_j u_i, \tag{2.9}$$

we drop the last term on the right hand side which describes the influence of the other grains on the field fluctuation produced by the ith grain. The remaining equation

$$(L_0 + L_i)u_i = -L_i\langle u\rangle \tag{2.10}$$

or

$$(L_0 + L_i)(u_i + \langle u \rangle) = L_0\langle u \rangle = \rho$$

can eventually be solved for simple grain shapes, especially for spheres. This approximation means that, in order to calculate u_i, the ith grain is taken into account exactly, whereas all the other grains are replaced by a homogeneous material described by the operator L_0. But, according to eqn (2.6), L_0 is related to the unknown effective quantity $\tilde{L}_{\text{eff}}(\mathbf{k})$ which, therefore, will be contained in the solution of (2.10). Inserting this solution into eqn (2.7) and performing the ensemble average, we obtain the relation

$$\left\langle \sum_i u_i \right\rangle = 0 \tag{2.11}$$

which may serve as an implicit equation for the unknown effective quantity $\tilde{L}_{\text{eff}}(\mathbf{k})$. A Fourier transformation leads us to the effective operator L_{eff} in the ordinary space.

The validity of the self-consistent method proposed in this section clearly depends on the special problem being considered, and on the choice of the decomposition (2.5). This approximation will be applicable only if there is a natural choice for the operators L_0 and L_i. Certainly, a rigorous justification for neglecting the coupling terms in eqn (2.9) cannot be given. We can only refer to similar self-consistent approaches which have been applied with more or less success in different fields of physics, and to eventual experimental verifications. The approximation scheme outlined above generalizes the basic ideas developed in the case of homogeneous mean fields[3]. Moreover, it is completely analogous to the so-called CPA (coherent potential approximation) which has been used successfully in the theory of disordered alloys[4]. There, the effective Hamiltonian and properties like the density of states are obtained in good agreement with experimental data. The main difference in comparison with the present problem consists in the discreteness and the regularity of the lattice whereas we are concerned with continuous space coordinates and random geometries.

In the frame of the self-consistent approximation outlined above correlations between the material properties of neighbouring grains cannot be taken into account. Consequently, the results are presumed reliable, if at all, only for materials without such correlations.

Further, the results of this approximation can be shown to agree with the perturbation treatment up to third order in the variation of material parameters provided that the geometry of the grains is treated exactly and that there are no correlations as mentioned above (see Appendix).

3. APPLICATION TO ELECTROSTATICS

In the case of electrostatics (1.2), it seems reasonable to choose the operator L_0 in the same form as the operator (1.2), i.e.

$$L_0 = -\frac{\partial}{\partial \mathbf{r}} \cdot \epsilon_0 \frac{\partial}{\partial \mathbf{r}} \tag{3.1}$$

where ϵ_0 is independent of position because of the required homogeneity of L_0. According to eqns (2.3) and (2.4) the mean field and the charge density are chosen as harmonic functions of position. Then the requirement of eqn (2.6) together with the definition (1.12) leads to

$$\epsilon_0 = \tilde{\epsilon}_{\text{eff}}(\mathbf{k}), \qquad L_0 = -\frac{\partial}{\partial \mathbf{r}} \cdot \tilde{\epsilon}_{\text{eff}}(\mathbf{k}) \frac{\partial}{\partial \mathbf{r}}. \tag{3.2}$$

With the aid of (2.1) and (2.2), L can be written as

$$\begin{aligned} L &= -\frac{\partial}{\partial \mathbf{r}} \cdot \epsilon(\mathbf{r}) \frac{\partial}{\partial \mathbf{r}} = -\frac{\partial}{\partial \mathbf{r}} \cdot \sum_i \epsilon_i \theta_i(\mathbf{r}) \frac{\partial}{\partial \mathbf{r}} \\ &= L_0 - \frac{\partial}{\partial \mathbf{r}} \cdot \sum_i (\epsilon_i - \tilde{\epsilon}_{\text{eff}}(\mathbf{k}))\theta_i(\mathbf{r}) \frac{\partial}{\partial \mathbf{r}}. \end{aligned} \tag{3.3}$$

This expression suggests the following definition of L_i

$$L_i \equiv -\frac{\partial}{\partial \mathbf{r}} \cdot (\epsilon_i - \tilde{\epsilon}_{\text{eff}}(\mathbf{k}))\theta_i(\mathbf{r}) \frac{\partial}{\partial \mathbf{r}}. \tag{3.4}$$

But it must be emphasized that the requirements made in section 2 do not determine the decomposition of L into L_0 and L_i uniquely. Although the expressions (3.1), (3.2) and (3.4) fulfill these requirements, different, but more complicated decompositions would be possible.

If we insert the settlements (3.2), (3.4) into eqn (2.10) it takes the form

$$\frac{\partial}{\partial \mathbf{r}} \cdot \{\tilde{\epsilon}_{\text{eff}} + (\epsilon_i - \tilde{\epsilon}_{\text{eff}})\theta_i(\mathbf{r})\} \frac{\partial}{\partial \mathbf{r}} u_i = -\frac{\partial}{\partial \mathbf{r}} \cdot (\epsilon_i - \epsilon_{\text{eff}})\theta_i(\mathbf{r}) \frac{\partial}{\partial \mathbf{r}} u_0 \, \mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}}$$

or

$$\frac{\partial}{\partial \mathbf{r}} \cdot \{\tilde{\epsilon}_{\text{eff}} + (\epsilon_i - \tilde{\epsilon}_{\text{eff}})\theta_i(\mathbf{r})\} \frac{\partial}{\partial \mathbf{r}} \{\langle u \rangle + u_i\} = \frac{\partial}{\partial \mathbf{r}} \cdot \tilde{\epsilon}_{\text{eff}} \frac{\partial}{\partial \mathbf{r}} \langle u \rangle = -\rho_0 \, \mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}}. \tag{3.5}$$

The solution u_i must tend to zero at great distances from the ith grain. Since only in the case of spherical grains a relatively simple solution can be expected, we restrict ourselves to the case that all grains can be approximated by spheres, similarly, for example, to the approximation of Wigner-Seitz-cells by spheres which is frequently used in solid state physics. It is perhaps possible to justify this approximation from the fact that spheres represent a certain average grain shape. Obviously, the approximation scheme allows dealing with other grain shapes, but this would certainly demand numerical methods.

If we single out the ith grain with radius R_i and attach the origin of coordinates to its center (see Fig. 1), the step function θ_i (2.2) reads

$$\theta_i(\mathbf{r}) = \theta(R_i - r) = \begin{cases} 1 \\ 0 \end{cases} \quad r \lesseqgtr R_i. \tag{3.6}$$

Let us split up u_i into two parts

$$u_i = -\theta_i(\mathbf{r}) u_0 \frac{\epsilon_i - \tilde{\epsilon}_{\text{eff}}}{\epsilon_i} \mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}} + u_i'. \tag{3.7}$$

Then eqn (3.5) becomes

$$\Delta u_i' = 0 \qquad \text{for} \qquad r \gtreqless R_i. \tag{3.8}$$

At the surface $r = R_i$, where the dielectric parameter in eqn (3.5) suffers a jump, the potential as well as the normal component of the dielectric displacement have to be continuous

$$\begin{gathered} \{\langle u \rangle + u_i\}_{\text{ext}} = \{\langle u \rangle + u_i\}_{\text{int}} \\ \tilde{\epsilon}_{\text{eff}} \frac{\partial}{\partial \mathbf{r}} \{\langle u \rangle + u_i\}_{\text{ext}} = \epsilon_i \frac{\partial}{\partial \mathbf{r}} \{\langle u \rangle + u_i\}_{\text{int}}. \end{gathered} \tag{3.9}$$

The labels ext and int indicate the exterior and interior sides of the spherical surface, respectively. Equations (3.9) lead together with eqn (3.7) to the following boundary conditions for u_i'

$$\begin{gathered} u_i'\big|_{\text{ext}} = u_i'\big|_{\text{int}} - u_0 \frac{\epsilon_i - \tilde{\epsilon}_{\text{eff}}}{\epsilon_i} \mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}} \\ \tilde{\epsilon}_{\text{eff}} \left.\frac{\partial u_i'}{\partial \mathbf{r}}\right|_{\text{ext}} = \epsilon_i \left.\frac{\partial u_i'}{\partial \mathbf{r}}\right|_{\text{int}}. \end{gathered} \tag{3.10}$$

In consequence of the spherical boundary it is convenient to expand the fields $\langle u \rangle$ and u_i into

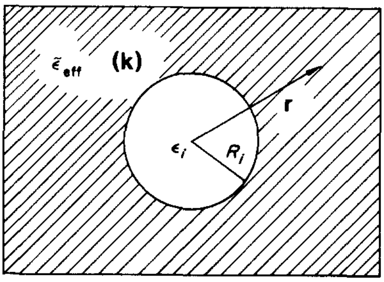


Fig. 1.

series of spherical harmonics. The mean field then becomes

$$\frac{\langle u\rangle}{u_0} = \mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}} = \sum_{l=0}^{\infty} U_l(kr)P_l(\zeta) \tag{3.11}$$

with

$$U_l(x) \equiv (2l+1)i^l\sqrt{\left(\frac{\pi}{2x}\right)}J_{l+(1/2)}(x) \quad \text{and} \quad \zeta = \frac{\mathbf{k}\cdot\mathbf{r}}{kr} = \cos(\sphericalangle\mathbf{k},\mathbf{r})$$

whereas the solution of (3.8) can be written as

$$u_i' = \begin{cases} \displaystyle\sum_{l=0}^{\infty} A_l\left(\frac{R_i}{r}\right)^{l+1} P_l(\zeta) \\ & r \gtrless R_i. \\ \displaystyle\sum_{l=0}^{\infty} B_l\left(\frac{r}{R_i}\right)^{l} P_l(\zeta) \end{cases} \tag{3.12}$$

This expression takes into account the rotational symmetry with respect to the axis **k** as well as the fact that u_i' has to remain finite in the origin and to vanish in the limit $r \to \infty$.

The boundary conditions (3.1) lead to a set of equations for the coefficients A_l and B_l

$$A_l - B_l = -\frac{\epsilon_i - \tilde{\epsilon}_{\mathrm{eff}}}{\epsilon_i}u_0U_l(kR_i) \quad (l+1)\,\tilde{\epsilon}_{\mathrm{eff}}A_l + l\epsilon_iB_l = 0 \tag{3.13}$$

whose solution reads

$$A_l = -\frac{\epsilon_i}{\tilde{\epsilon}_{\mathrm{eff}}}\cdot\frac{l}{l+1}B_l = -\frac{(\epsilon_i - \tilde{\epsilon}_{\mathrm{eff}})l}{(l+1)\tilde{\epsilon}_{\mathrm{eff}} + l\epsilon_i}u_0U_l(kR_i). \tag{3.14}$$

Equation (3.7), (3.12) and (3.14) represent the solution $u_i(\mathbf{r})$ of eqn (3.5) for a grain centered at the origin. In order to obtain the solution $u_i(\mathbf{r}, \mathbf{r}_i)$ for an arbitrary grain position $\mathbf{r}_i$, we only have to replace the coordinate $\mathbf{r}$ in $u_i(\mathbf{r})$ by the difference vector $\mathbf{r} - \mathbf{r}_i$, and to multiply it by a factor $\exp\mathbf{k}\cdot\mathbf{r}$

$$u_i(\mathbf{r}, \mathbf{r}_i) = u_i(\mathbf{r} - \mathbf{r}_i, 0)\,\mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}_i} \equiv u_i(\mathbf{r} - \mathbf{r}_i)\,\mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}_i}. \tag{3.15}$$

This additional factor arises from the exponential term on the right hand side of eqn (3.5) which can be split up into two factors $\exp i\mathbf{k}\cdot\mathbf{r} = \exp i\mathbf{k}\cdot(\mathbf{r} - \mathbf{r}_i)\exp i\mathbf{k}\cdot\mathbf{r}_i$.

Now, substituting the results for u_i into eqn (2.11) and ensemble averaging yields an implicit equation on $\tilde{\epsilon}_{\mathrm{eff}}$. The ensemble is characterized by a probability distribution for the positions $\mathbf{r}_i$, the dielectricity constants ϵ_i, and the sizes R_i. Because of statistical homogeneity all positions $\mathbf{r}_i$ of a grain have equal probability independently of the other stochastic parameters ϵ_i, R_i. Thus, taking into account eqn (3.15), we can perform the position average of u_i separately to obtain

$$\begin{aligned} \bar{u}_i^{\mathbf{r}_i} &= \frac{1}{V}\int \mathrm{d}\mathbf{r}_i\, u_i(\mathbf{r}, \mathbf{r}_i) = \frac{1}{V}\int \mathrm{d}\mathbf{r}_i\, \mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}_i}u_i(\mathbf{r} - \mathbf{r}_i) \\ &= \frac{\mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}}}{V}\int \mathrm{d}\mathbf{r}'\, \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}'}u_i(\mathbf{r}'). \end{aligned} \tag{3.16}$$

In order to express the remaining average with respect to ϵ_i and R_i, we introduce the mean number N_α of grains with determinate values ϵ_α and R_α or, in other terms, the probability $p_\alpha = N_\alpha / N (N = \sum_\alpha N_\alpha)$ for an arbitrary grain to be of the kind α. With help of these quantities we finally obtain

$$\begin{aligned}\left\langle \sum_i u_i \right\rangle &= \mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}} \sum_\alpha \frac{N_\alpha}{V} \int \mathrm{d}\mathbf{r}'\, \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}'}\, u_\alpha(\mathbf{r}') \\ &= \frac{N}{V} \mathrm{e}^{i\mathbf{k}\cdot\mathbf{r}} \sum_\alpha p_\alpha \int \mathrm{d}\mathbf{r}'\, \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}'} u_\alpha(\mathbf{r}') = 0.\end{aligned} \tag{3.17}$$

Let us recall that the field contribution $u_\alpha(\mathbf{r})$ belongs to a grain of the kind α (i.e. ϵ_α, R_α) located at the origin.

4. EVALUATION OF EFFECTIVE DIELECTRICITY

In view of the following calculations, it is convenient to replace the self-consistency condition (2.11) by its derivative

$$i\mathbf{k} \cdot \frac{\partial}{\partial \mathbf{r}} \left\langle \sum_i u_i \right\rangle = \left\langle \sum_i i\mathbf{k} \cdot \frac{\partial}{\partial \mathbf{r}} u_i \right\rangle = 0. \tag{4.1}$$

Conformely with eqn (3.17), the ensemble average can be written as

$$\sum_\alpha p_\alpha \int \mathrm{d}\mathbf{r}\, \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}} i\mathbf{k} \cdot \frac{\partial}{\partial \mathbf{r}} u_\alpha = 0. \tag{4.2}$$

$$\sum_\alpha p_\alpha \left\{ k^2 u_0 \frac{\epsilon_\alpha - \epsilon_{\mathrm{eff}}}{\epsilon_\alpha} V_\alpha + \int_{r<R_\alpha} \mathrm{d}\mathbf{r}\, \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}} i\mathbf{k} \cdot \frac{\partial u'_\alpha}{\partial \mathbf{r}} + \int_{r>R_\alpha} \mathrm{d}\mathbf{r}\, \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}} i\mathbf{k} \cdot \frac{\partial u'_\alpha}{\partial \mathbf{r}} \right\} = 0 \tag{4.3}$$

where $V_\alpha = 4\pi R_\alpha^3/3$ is the volume of a grain of kind α. The calculation of the second term on the left hand side may be performed by using eqn (3.8) for u_i, to obtain

$$\begin{aligned} 0 = \int_{r<R_\alpha} \mathrm{d}\mathbf{r}\, \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}} \Delta u'_\alpha &= \oiint_{r=R_\alpha} \mathrm{d}S \left.\frac{\partial u'_\alpha}{\partial r}\right|_{\mathrm{int}} \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}} \\ &+ \int_{r<R_\alpha} \mathrm{d}\mathbf{r}\, i\mathbf{k} \cdot \frac{\partial u'_\alpha}{\partial \mathbf{r}} \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}}. \end{aligned} \tag{4.4}$$

The surface integral can be treated with the help of the expansions (3.11), (3.12). On account of the orthogonality of Legendre polynomials, it follows

$$\int_{r<R_\alpha} \mathrm{d}\mathbf{r}\, i\mathbf{k} \cdot \frac{\partial u'_\alpha}{\partial \mathbf{r}} \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}} = -\oiint \mathrm{d}S \left.\frac{\partial u'_\alpha}{\partial r}\right|_{\mathrm{int}} \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}} = -4\pi R_\alpha \sum_l \frac{lB_l}{2l+1} U_l^*(kR_\alpha) \tag{4.5}$$

and, analogously, for the exterior region

$$\int_{r>R_\alpha} \mathrm{d}\mathbf{r}\, i\mathbf{k} \cdot \frac{\partial u'_\alpha}{\partial \mathbf{r}} \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}} = \oiint \mathrm{d}S \left.\frac{\partial u'_\alpha}{\partial r}\right|_{\mathrm{ext}} \mathrm{e}^{-i\mathbf{k}\cdot\mathbf{r}} = -4\pi R_\alpha \sum_l \frac{(l+1)A_l}{2l+1} U_l^*(kR_\alpha). \tag{4.6}$$

Thus, eqn (4.3) takes the form

$$\begin{aligned} \sum_\alpha p_\alpha k^2 V_\alpha \frac{\epsilon_\alpha - \bar{\epsilon}_{\mathrm{eff}}}{\epsilon_\alpha} &= \sum_\alpha p_\alpha 4\pi R_\alpha \sum_l \frac{(l+1)A_l + lB_l}{(2l+1)u_0} U_l^*(kR_\alpha) \\ &= -\sum_\alpha p_\alpha 4\pi R_\alpha \frac{(\epsilon_2 - \tilde{\epsilon}_{\mathrm{eff}})^2}{\epsilon_\alpha} \sum_l \frac{(l+1)}{(2l+1)} \frac{|U_l(kR_\alpha)|^2}{(l+1)\bar{\epsilon}_{\mathrm{eff}} + l\epsilon_\alpha} \end{aligned} \tag{4.7}$$

where in the last equation the expression (3.14) for the coefficients has been used. If we introduce the abbreviations

$$S(x, y) \equiv \sum_{l=1}^{\infty} \frac{l(l+1)}{l+1+ly} \cdot \frac{|U_l(x)|^2}{2l+1} = \sum_{l=1}^{\infty} \frac{(2l+1)(l+1)l}{l+1+ly} \frac{(J_{l+(1/2)}(x))^2}{2x} \tag{4.8}$$

$$Q(x, y) \equiv \left(1 - \frac{1}{y}\right)\left[1 + 3(y-1)\frac{S(x, y)}{x^2}\right], \tag{4.9}$$

the implicit equation governing the unknown quantity $\tilde{\epsilon}_{\text{eff}}$ can finally be written as

$$\sum_{\alpha} p_\alpha \frac{v_\alpha}{v} Q\left(kR_\alpha, \frac{\epsilon_\alpha}{\tilde{\epsilon}_{\text{eff}}(\mathbf{k})}\right) = \sum_{\alpha} v_\alpha Q\left(kR_\alpha, \frac{\epsilon_\alpha}{\tilde{\epsilon}_{\text{eff}}(\mathbf{k})}\right) = 0. \tag{4.10}$$

Here, v_α denotes the volume part occupied by grains of kind α. Obviously, an approximative replacement of the infinite sum $S(x, y)$ by a closed function is desirable. This can be achieved with the help of the approximation

$$\frac{l(l+1)}{l+1+ly} \approx \frac{2}{1+y}\left[\frac{l(l+1)}{2l+1} + \frac{1}{4}\frac{1-y}{1+y}\left(\frac{2-y}{2+y} \cdot \frac{1}{2l+1} + 1\right)\right]. \tag{4.11}$$

This relation is exact for $l = 1$ as well as for $y = 0, 1, +\infty$ and its maximal error can be shown to amount about 3 per mill. If we employ this approximation, the sum involved in S (4.8) can be carried out by means of the following exact relations (see [5])

$$\begin{aligned} &\sum_{l=0}^{\infty} \frac{|U_l(x)|^2}{2l+1} = \frac{\pi}{2x} \sum_{l=0}^{\infty} (2l+1)\ J^2_{l+(1/2)}(x) = 1 \\ &\sum_{l=0}^{\infty} |U_l(x)|^2 = \frac{1}{2}\left[\frac{\sin 2x}{2x} + \cos 2x + 2x\,si(2x)\right] \equiv 1 + 2x^2\alpha(x) \\ &\sum_{l=0}^{\infty} \left|\frac{U_l(x)}{2l+1}\right|^2 = \frac{si(2x)}{2x} \equiv 1 + x^2\beta(x). \end{aligned} \tag{4.12}$$

The functions α, β and γ

$$|U_0(x)|^2 = \left(\frac{\sin x}{x}\right)^2 \equiv 1 + x^2\gamma(x) \tag{4.13}$$

are introduced for convenience. Eqns (4.11)–(4.13) then lead to

$$\begin{aligned} S(x, y) &\approx \frac{1}{2(y+1)} \sum_{l=0}^{\infty} \left[(2l+1) - \frac{6y}{(1+y)(2+y)} \cdot \frac{1}{2l+1} - \frac{1-y}{1+y}\right] \frac{|U_l(x)|^2}{2l+1} \\ &\quad - \frac{1}{2(y+1)}\left[1 - \frac{6y}{(1+y)(2+y)} - \frac{1-y}{1+y}\right] |U_0(x)|^2 \\ &= \frac{x^2}{(1+y)^2(2+y)} [2\alpha + y(3\alpha - 3\beta + \gamma) + y^2(\alpha - \gamma)]. \end{aligned} \tag{4.14}$$

This approximation enables us to compute $\tilde{\epsilon}_{\text{eff}}(\mathbf{k})$ from (4.10) for an arbitrary composition of the mixture.

Especially in the limiting case of a homogeneous mean field $kR_\alpha \to 0$ the exact expression (4.8) as well as the approximation (4.14) reduce to

$$\begin{aligned} \lim_{x \to 0} \frac{S(x, y)}{x^2} &= \frac{2}{3(2+y)} \\ Q(0, y) &= \frac{3(y-1)}{y+2} \end{aligned} \tag{4.15}$$

so that (4.10) becomes

$$\sum_{\alpha} v_\alpha \frac{\epsilon_\alpha - \tilde{\epsilon}_{\text{eff}}}{\epsilon_\alpha + 2\tilde{\epsilon}_{\text{eff}}} = \left\langle \frac{\epsilon_\alpha - \tilde{\epsilon}_{\text{eff}}}{\epsilon_\alpha + 2\tilde{\epsilon}_{\text{eff}}} \right\rangle = 0, \qquad kR_\alpha = 0 \tag{4.16}$$

in agreement with the result of familiar self-consistent treatments for homogeneous fields[3]. Here the bracket denotes the volume average.

In the other limit of extremely inhomogeneous fields $kR_\alpha \to \infty$, expression (4.14) leads to the exact result known from literature[6]

$$Q(x = \infty, y) = 1 - \frac{1}{y}$$

$$\sum_{\alpha} v_\alpha \left(1 - \frac{\tilde{\epsilon}_{\text{eff}}}{\epsilon_\alpha}\right) = 0 \quad \text{or} \quad \frac{1}{\tilde{\epsilon}_{\text{eff}}} = \left\langle \frac{1}{\epsilon} \right\rangle, \; kR_\alpha \to \infty. \tag{4.17}$$

5. NUMERICAL RESULTS AND DISCUSSION

In Fig. 2 the function $Q(x, y)$ is plotted. Numerical calculations of $\tilde{\epsilon}_{\text{eff}}$ have been carried out for a two-component mixture. All grains are assumed to be of equal size $R_\alpha = R$. Consequently, the sum over α reduces to the sum over the two components where their volume parts v_1 and v_2 appear as weighing factors. Two special volume ratios of the mixture 0·5 : 0·5 and 0·8 : 0·2 have been chosen. These two cases reveal characteristic peculiarities in strongly heterogeneous media in which the two dielectric parameters are extremely different from one another ($\epsilon_2 \gg \epsilon_1$).

To begin with the limiting case $kR = 0$, we readily see from (4.16) that, for strong heterogeneity,

$$\tilde{\epsilon}_{\text{eff}} \approx \begin{cases} \dfrac{1}{1 - 3v_2}\epsilon_1 \\[2ex] \dfrac{3v_2 - 1}{2}\epsilon_2 \end{cases} \quad v_2 \lesseqgtr \frac{1}{3} \quad \text{for } \epsilon_2 \gg \epsilon_1, \; kR = 0 \tag{5.1}$$

holds. Consequently, as long as the second component possessing the greater dielectric parameter ϵ_2 covers more than one third of the body, $\tilde{\epsilon}_{\text{eff}}$ takes a value of the order of magnitude ϵ_2. As soon as this relative volume v_2 decreases below one third, $\tilde{\epsilon}_{\text{eff}}$ suffers a rapid transition to the order of magnitude of the smaller dielectricity ϵ_1. This behaviour is analogous to the percolation phenomena studied in disordered alloys[7].

In the opposite limiting case $kR \to \infty$ we deduce from (4.17) for strong heterogeneity

$$\frac{1}{\tilde{\epsilon}_{\text{eff}}} = \frac{v_1}{\epsilon_1} + \frac{v_2}{\epsilon_2} \approx \frac{v_1}{\epsilon_1} \quad \text{for} \quad \epsilon_2 \gg \epsilon_1, \quad kR \to \infty. \tag{5.2}$$

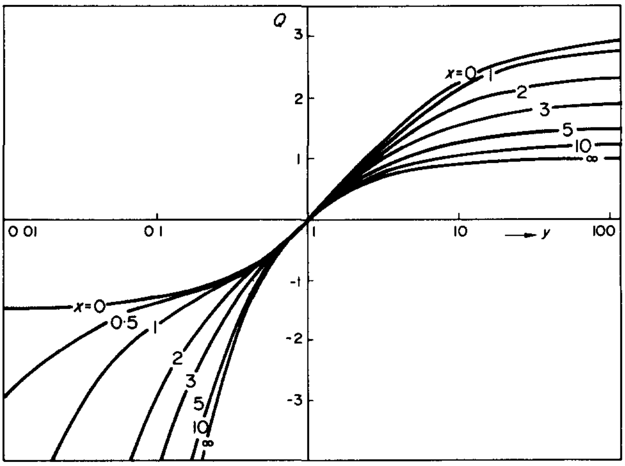
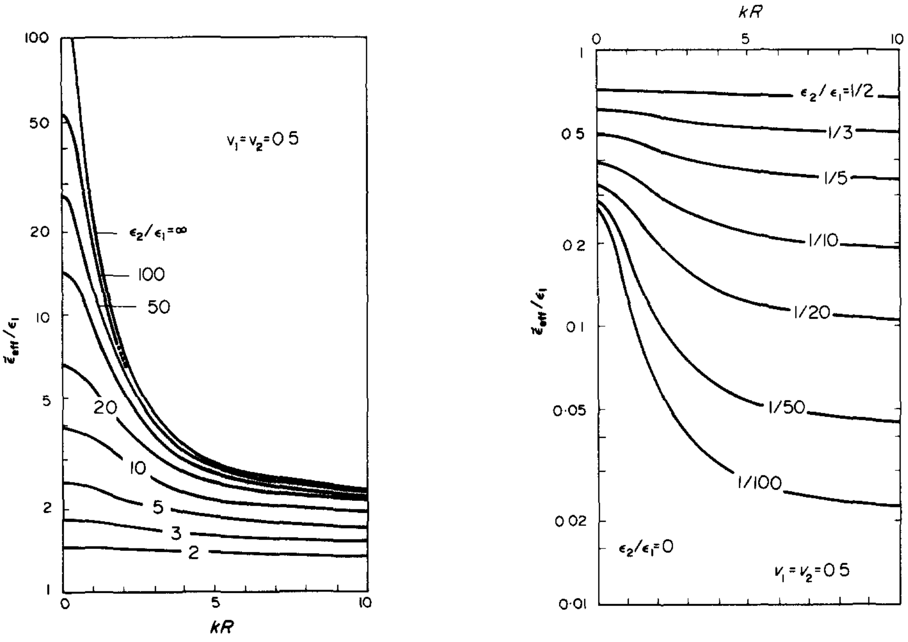


Fig. 2.

We see that in this limit the effective dielectricity is of the order of magnitude of the smaller dielectric parameter ϵ_1 whatever the relative volumes are (except extremely small v_1). Comparison of the two limiting cases suggests that for $v_2 < 1/3$ and independently of kR the effective dielectricity $\tilde{\epsilon}_{\text{eff}}$ remains within the range of ϵ_1. On the contrary, for $v_2 > 1/3$, $\tilde{\epsilon}_{\text{eff}}$ as a function of kR varies between the orders of magnitude ϵ_1 and ϵ_2. As is seen from Fig. 3, this transition takes place at small $kR \ll 1$. Indeed, in the strongly heterogeneous case the $\tilde{\epsilon}_{\text{eff}} - kR$—diagram shows a high peak about $k_R = 0$. Its width turns out to be of the order $kR = 0(7\sqrt{(\epsilon_1/\epsilon_2 v_1)})$. Therefore, if we consider the effective dielectricity $\tilde{\epsilon}_{\text{eff}}$ of a strongly heterogeneous body as a function of wave length $\lambda = 2\pi/k$ and if we diminish λ, starting from infinity, the decrease of $\tilde{\epsilon}_{\text{eff}}$ from the order of ϵ_2 to that of ϵ_1 will appear at wavelengths which are much greater than the grain radius. The peak in the Fourier space necessarily gives rise to a long range of the non-locality within the physical space. In fact, $\epsilon_{\text{eff}}(\mathbf{r})$ vanishes only at distances of the order $r > 0{\cdot}15R\sqrt{(v_1\epsilon_2/\epsilon_1)}$. This can considerably exceed the correlation length being of the order of R.

As it is to be expected, these effects observed at a mixture ratio of 0·5 : 0·5 occur in the case of



(a)

(b)

Fig. 3.

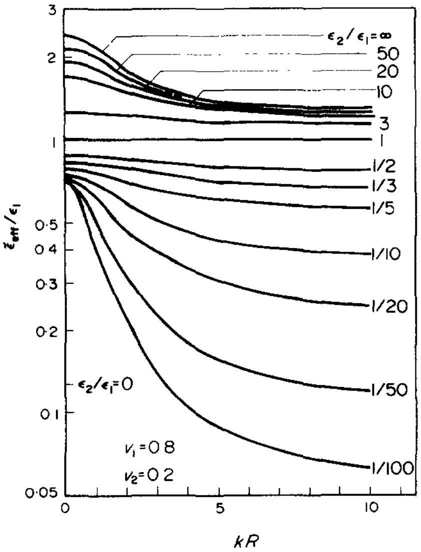


Fig. 4.

a mixture ratio of 0·2 : 0·8 only if the greater value of ϵ belongs to the prevailing component (see Fig. 4).

The long-range non-locality stated above looks very strange. This pecularity depends on the assumptions about the source distribution. If we admit random sources whose fluctuations are correlated with those of the material properties, the effective behaviour can considerably differ from the above results. This also holds for the range of nonlocality. A forthcoming paper will be devoted to these effects.

Obviously, the results of the present paper can be carried over without any change to other properties of composites such as electric and heat conductivity, diffusion, permeability, because these quantities are governed by the same laws (1.1), (1.2) as we have started from. In the case of elasticity the tensorial character involves some computational difficulties.

At the end the reader must be warned of ascribing general validity to our results. As must be noticed, the effective behaviour of a heterogeneous body does not only depend on the ratio of its components but also on their morphology. In this paper all the components have been treated on the same footing. Therefore, we expect the achieved results to describe only the behaviour of such heterogeneous bodies in which, disregarding different volume parts, all the components are of similar geometry. A counter-example is a connected matrix with insulated inclusions of different material. Certainly, such a heterogeneous medium demands other methods for calculating its effective behaviour. However, the restriction to nearly spherical grains is not inherent to the general procedure, and could be removed with an appropriate computational effort.

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APPENDIX

Comparison of the self-consistent approach with a perturbation treatment

The effective operator L_{eff} is connected with the stochastic operator L by the operator relation

$$L_{\text{eff}} = g^{-1} = \langle g_{st}\rangle^{-1} = \langle L^{-1}\rangle^{-1}. \tag{A1}$$

Decomposing L according to eqn (2.5)

$$L = L_0 + L' \qquad L' = \sum_i L_i, \tag{A2}$$

we expand expression (A1) into powers of L' to obtain

$$\begin{aligned} O &= (L_{\text{eff}} - L_0)\langle u\rangle \\ &= \{\langle L'\rangle - \langle L'g_0L'\rangle + \langle L'\rangle g_0\langle L'\rangle + \langle L'g_0L'g_0L'\rangle \\ &\quad - \langle L'g_0L'\rangle g_0\langle L'\rangle - \langle L'\rangle g_0\langle L'g_0L'\rangle + \langle L'\rangle g_0\langle L'\rangle g_0\langle L'\rangle + \ldots\}\langle u\rangle \end{aligned} \tag{A3}$$

up to third order terms. The Green operator g_0 represents the inversion of L_0 and is supposed to be known. Transporting the first term of expansion (A3) to the other side of the equation, we can immediatly see that, although L' being of first order, its average $\langle L'\rangle$ is only a second order term. This means that some of the terms written down in (A3) are of higher than third order. If we omit these contributions, eqn (A3) simplifies to

$$\{\langle L'\rangle - \langle L'g_0L'\rangle + \langle L'g_0L'g_0L'\rangle + \text{fourth order}\}.\langle \text{u}\rangle \tag{A4}$$

On the other hand, the self-consistency condition (2.11) can be transformed into

$$L_0\left\langle \sum u_i\right\rangle = -\left\langle \sum L_i(\langle u\rangle + u_i)\right\rangle = 0 \tag{A5}$$

where eqn (2.10) is used. Interserting the solution for u_i in an expanded form

$$\begin{aligned} u_i &= -g_0L_i(\langle u\rangle + u_i) \\ &= -g_0(L_i - L_ig_0L_i + \ldots)\langle u\rangle, \end{aligned} \tag{A6}$$

we can rewrite (A5) as

$$\left[\left\langle\sum_i L_i\right\rangle - \left\langle\sum_i L_ig_0L_i\right\rangle + \sum_i \langle L_ig_0L_ig_0L_i\rangle - \ldots\right]\langle u\rangle = 0. \tag{A7}$$

A comparison with the perturbation series (A4) shows a complete accordance of the first order terms. The second order terms differ by

$$\langle L'g_0L'\rangle - \left\langle\sum_i L_ig_0L_i\right\rangle = \left\langle\sum_{i,j} L_ig_0L_j\right\rangle - \left\langle\sum_i L_ig_0L_i\right\rangle = \left\langle\sum_{i\neq j} L_ig_0L_j\right\rangle. \tag{A8}$$

Turning to the special case of electrostatics (3.2), (3.4), we can easily show this difference to be of fourth order only, provided that the dielectric parameter outside the ith grain is not correlated to the properties of this grain. The last expression in (A6) contains a two point average

$$f(\mathbf{r}, \mathbf{r}') = \left\langle\sum_{i\neq j} (\epsilon_i - \tilde{\epsilon}_{\text{eff}})\theta_i(\mathbf{r})(\epsilon_j - \tilde{\epsilon}_{\text{eff}})\theta_j(\mathbf{r}')\right\rangle. \tag{A9}$$

With the aid of eqns (2.1), (2.2), it may be written in different forms†

$$\begin{aligned} f(\mathbf{r}, \mathbf{r}') &= \left\langle\sum_{i\neq j} (\epsilon_i - \tilde{\epsilon}_{\text{eff}})\theta_i(\mathbf{r})\theta_j(\mathbf{r}')[\epsilon(\mathbf{r}') - \tilde{\epsilon}_{\text{eff}}]\right\rangle \\ &= \left\langle\sum_i (\epsilon_i - \tilde{\epsilon}_{\text{eff}})\theta_i(\mathbf{r})[1 - \theta_i(\mathbf{r}')] \,.\, [\epsilon(\mathbf{r}') - \tilde{\epsilon}_{\text{eff}}]\right\rangle. \end{aligned} \tag{A10}$$

In consequence of the above assumption, the average of $\epsilon(\mathbf{r}')$ can be performed independently of the ith grain

$$f(\mathbf{r}, \mathbf{r}') = \left\langle\sum_i (\epsilon_i - \tilde{\epsilon}_{\text{eff}})\theta_i(\mathbf{r})[1 - \theta_i(\mathbf{r}')]\right\rangle\langle\epsilon - \tilde{\epsilon}_{\text{eff}}\rangle. \tag{A11}$$

Repeating the same arguments with respect to the other point $\mathbf{r}$, we obtain

$$\begin{aligned} f(\mathbf{r}, \mathbf{r}') &= \left\langle\sum_{i\neq j} (\epsilon_i - \tilde{\epsilon}_{\text{eff}})\theta_i(\mathbf{r})\theta_j(\mathbf{r}')\right\rangle\langle\epsilon - \tilde{\epsilon}_{\text{eff}}\rangle \\ &= \left\langle(\epsilon(\mathbf{r}) - \tilde{\epsilon}_{\text{eff}})\sum_j [1 - \theta_j(\mathbf{r})]\theta_j(\mathbf{r}')\right\rangle\langle\epsilon - \tilde{\epsilon}_{\text{eff}}\rangle \\ &= \langle\epsilon - \tilde{\epsilon}_{\text{eff}}\rangle^2\left\langle\sum_j [1 - \theta_j(\mathbf{r})]\theta_j(\mathbf{r}')\right\rangle. \end{aligned} \tag{A12}$$

According to the discussion after (A3), $\langle L'\rangle\langle u\rangle$ and, therefore, $\langle\epsilon - \tilde{\epsilon}_{\text{eff}}\rangle$ are only of second order. Thus, the expressions (A12) and (A8) are only of fourth order, as affirmed above.

An analogous reasoning can be applied to the difference of the third order terms in (A4) and (A7) which turns out to be also of fourth order.

It results that, for weak heterogeneities, the self-consistent and the perturbation treatments are in accordance up to third order, provided that there is no correlation between the material properties of different grains.

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†Mathematical difficulties which could arise from the step function are physically irrelevant. They can always be avoided by imagining the steps as rapid, but continuous transitions.