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The macroscopic description of the effective field in certain dielectrics

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Macroscopic formulas which essentially generalize the Lorentz-Lorenz relation are found for the effective field and its first derivatives. It can be seen from the derived equations that the quantity g is in general not equal to $4\pi/3$, but is related to the dielectric constant and its derivatives with respect to density and shear deformations. The proposed derivation uses results of the macroscopic theory for the volume density of electric forces in dielectrics.

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1. INTRODUCTION

The effective field \mathbf{E}_{eff} acting on a certain particle in a condensed medium is the average of the microscopic field under the condition that the given particle is in a definite state. On the other hand the macroscopic field \mathbf{E} that appears in Maxwell's equations is obtained from the microscopic field by averaging over the states of all the particles. Consequently, when the correlations between the particles in their motion or spatial distribution are taken into account the effective field \mathbf{E}_{eff} and the macroscopic field \mathbf{E} are not equal. This difference leads to the so-called local-field effects in condensed media, which are well known and have been studied for crystals^{1,2} and also for liquids³ and plasmas.^{4,5}

The macroscopic description of effective fields is by no means always possible. This is clear already from the fact that the field \mathbf{E}_{eff} is in general a function of the state of the particle and is thus basically a microscopic quantity. The difference between the fields acting on different particles is often important. It is well known, for example, that the Davydov splitting in molecular crystals with several molecules in the elementary cell is due to the differences between the fields acting on these molecules; even in isotropic cubic crystals with several molecules in an elementary cell the fields acting on these molecules differ considerably.¹ There are also differences between the fields acting on different components in many-component liquids, and similar differences in a number of other cases. Other cases are also encountered in which with reasonable accuracy

one can neglect the differences between the effective fields acting on the various particles, and thus it makes sense to consider a single average effective field for all particles. This applies, for example, to cubic crystals with one molecule in the elementary cell, to some one-component simple liquids, and also, under certain circumstances, to plasmas. It is clear from the start that in a macroscopic approach, with the properties of the medium described with one dielectric constant, we have the only sort of case in which a description in terms of effective fields may be possible. If this condition holds, together with certain definite supplementary assumptions, it is well known that the effective field is connected with the macroscopic field by the Lorentz-Lorenz formula

$$\mathbf{E}_{\text{eff}} = \mathbf{E} + \frac{4\pi}{3} \mathbf{P} = \frac{\epsilon + 2}{3} \mathbf{E}. \quad (1)$$

This formula is an approximate one, and only in cases for which its use is justified (cubic crystals,² nonpolar liquids³) it usually provides only a qualitative account of the difference between the effective and the macroscopic fields.

In many cases, however, the Lorentz-Lorenz formula is incorrect even in a qualitative description of the effective field. The best known example of this is a plasma with the dielectric constant

$$\epsilon = 1 - 4\pi e^2 N / m\omega^2,$$

for which, as is clear from the microscopic theory, the

difference between the effective and macroscopic fields is zero,⁶ whereas for this case the Lorentz-Lorenz formula gives the value

$$|E_{eff} - E|/E = 4\pi e^2 N / 3m\omega^2$$

which is not zero, and in general not small compared with unity.

Among the conditions for the applicability of the Lorentz-Lorenz formula, an important one is that the dielectric constant depend on the density in a definite way. Here, in Sec. 2, we find a simple macroscopic formula which connects the effective field with the macroscopic field but is derived without any assumption about the way the dielectric constant depends on the density. This result agrees with the Lorentz-Lorenz formula when the latter can be applied. In the case of the plasma with dielectric constant $\epsilon = 1 - 4\pi e^2 N / m\omega^2$, however, and also for rarefied gases, where the dielectric constant is a linear function of the density, it follows from this result that the effective and macroscopic fields are identical. Thus the result in question, although approximate, has a broader range of applicability than the Lorentz-Lorenz formula.

In the third section we consider the effective field in nonuniform media, and also in nonstationary fields. This problem, like every question about the effective field and its macroscopic description, has recently attracted attention in connection with papers by Peierls.⁷ An expression is found for the derivative of the effective field in a nonuniform medium. For nonstationary fields, the expression found for the spatial derivative of the effective field is the same as Peierls' phenomenological formula, with more general expressions for the coefficients σ and τ that were introduced in Ref. 7. In the nonstationary case the treatment of the time derivative of the effective field must be conducted in analogy with the consideration of the spatial derivatives. It turns out that certain results of Ref. 7 are, in general, incorrect, which explains their failure to agree with experiment.

2. THE EFFECTIVE FIELD IN A HOMOGENEOUS DIELECTRIC

The effective field is directly connected with the force acting on a particle of the medium. Therefore in a macroscopic approach to the determination of the effective field it is natural to use the results of macroscopic theory for the volume density of electric forces in dielectrics. The expression for the volume density of electric forces in terms of the macroscopic field is well known and is found either by variation of the free energy of the field in the medium (cf. Ref. 8, Secs. 15, 16), or from an analysis of the conservation laws for the electromagnetic field in a moving medium (cf. Ref. 9, Chap. 12). On the other hand, to a first approximation it is not hard to write an expression for the volume density of electric forces which involves the effective field. We find a formula for the effective field by comparing the two expressions.

We shall apply this method first to a homogeneous liquid dielectric in a static electric field. In this case

the expression connecting the volume force density with the macroscopic field [see Ref. 8, Sec. 15, Eq. (15.12)] can be written in the form

$$f = \frac{1}{4\pi} \rho \left(\frac{\partial \epsilon}{\partial \rho} \right)_T (\mathbf{E} \nabla) \mathbf{E}. \quad (2)$$

Equation (2) holds for a nonabsorbing medium with spatial dispersion effects neglected, and for not too strong fields, for which we can use the results of linear macroscopic theory. On the other hand, since the force with which the field \mathbf{E}_{micro} acts on a dipole \mathbf{d} is $(\mathbf{d} \nabla) \mathbf{E}_{micro}$, in the dipole approximation the volume force density is $\langle (\mathbf{P}_{micro} \nabla) \mathbf{E}_{micro} \rangle$, where the quantities \mathbf{E}_{micro} and \mathbf{P}_{micro} are the microscopic values of the electric field and the density of dipole moment, and the angle brackets denote averaging. According to the definition of the effective field the averaging of the force acting on an individual charged particle reduces to the replacement of the microscopic field by the effective field. Confining ourselves to those cases in which the mean effective fields are the same for all particles, we can write the following expression for the volume force density:

$$f = (\mathbf{P} \nabla) \mathbf{E}_{eff}. \quad (3)$$

From microscopic theory it is well known that the calculations of the effective field and of the dielectric constant are closely related, and that for consistent results these quantities must be found with the same assumptions. In this connection it is important that the formula (3) has been found in the dipole approximation. Therefore when it is used inclusion of higher multipoles either in the effective field or in the dielectric constant is in general exaggerated accuracy. As is well known, a similar restriction is also essential for the Lorentz-Lorenz formula (for crystal lattices the matter of the higher multipoles is considered in Refs. 10 and 1).

Comparing Eqs. (2) and (3), we find that the effective and macroscopic fields are connected by the formula

$$\mathbf{E}_{eff} = \rho \left(\frac{\partial \epsilon}{\partial \rho} \right)_T \frac{\mathbf{E}}{\epsilon - 1}. \quad (4)$$

This corresponds to the Lorentz-Lorenz formula (1) if the condition

$$\rho (\partial \epsilon / \partial \rho)_T = (\epsilon - 1)(\epsilon + 2) / 3. \quad (4')$$

is fulfilled. This last equation is well known and has long been used to calculate the quantity $\rho (\partial \epsilon / \partial \rho)_T$ in liquids (see, for example, Ref. 11); it follows directly from the condition that the Clausius-Mossotti function be independent of the density:

$$\Phi_{C-M} = \frac{1}{\rho} \frac{\epsilon - 1}{\epsilon + 2} = \text{const.} \quad (5)$$

This condition (5) for the applicability of the Lorentz-Lorenz formula to liquids is also used in implicit form in the usual method for deriving the formula (see, for example, Ref. 12, Sec. 28). In fact, in the usual derivation of the Lorentz-Lorenz formula it is assumed that the influences of near neighbors on a particular particle are unimportant and that the difference between the effective and macroscopic fields is due only to particles outside a macroscopic sphere. In this sense it is immaterial whether there are or are not particles filling

this sphere. Accordingly, in the usual derivation of the Lorentz-Lorenz formula it is in practice assumed that the "internal" polarizability β of the medium, normalized to one particle as given by the relation

$$N\beta E_{eff} = P = (\epsilon - 1)E/4\pi,$$

does not depend on the density of the particles. If the Lorentz-Lorenz formula is correct, the value of β is connected with the dielectric constant by the relation

$$\beta = \frac{3}{4\pi N} \frac{\epsilon - 1}{\epsilon + 2},$$

where N is the density of the particles. This makes clear the physical meaning of the condition (5). It follows from Eq. (4) that

$$\beta = \frac{1}{4\pi N} \frac{(\epsilon - 1)^2}{\rho(\partial\epsilon/\partial\rho)_T},$$

and that if the dielectric constant is a linear function of the density, $\beta = 1 + b\rho$, then we get $\beta = (\epsilon - 1)/4\pi N$, which, for a dilute nonpolar gas corresponds, as it should, to the polarizability of an individual particle.

In the case when the dielectric constant is a linear function of the density, Eq. (4) shows that the effective field and the macroscopic fields are equal. By using the results of Pitaevskii,¹³ who found an expression connecting the volume force density, averaged over frequency, with a macroscopic monochromatic field in a dielectric with frequency dispersion, far from absorption lines, we can verify that Eq. (4) relates not only to static fields, but also to the amplitudes of monochromatic fields in a nonabsorbing dielectric with frequency dispersion. Thus Eq. (4) implies that the effective and macroscopic fields are identical for a plasma with dielectric constant

$$\epsilon(\omega) = 1 - 4\pi e^2 N / m\omega^2.$$

If for a liquid we deal with the difference between the effective and macroscopic fields with small-correction perturbation theory, so that $E_{eff} \approx (1 - a\rho^\gamma)E$, where a and γ do not depend on the density, we find from Eq. (4) that in this approximation

$$\epsilon \approx 1 + c\rho(1 - a\rho^\gamma/\gamma),$$

where the quantity c does not depend on the density and is connected with the value of the dielectric constant with the difference between E_{eff} and E neglected.

In solids the tensor dielectric constant depends not only on the density, but also changes with shear deformations. Therefore solids differ from liquids not only in the conditions for the applicability of the Lorentz-Lorenz formula, but also in a more general relation between the effective and macroscopic fields than is given by that formula. We find the appropriate expressions by the same method as we used to derive Eqs. (4) and (5).

In a homogeneous isotropic solid which is in a static electric field, the expression which relates the volume force density with the macroscopic field (see Ref. 8, Sec. 16) can be written in the form

$$f = -\frac{1}{4\pi} \left(\frac{1}{2} a_1 + a_2 \right) (E\nabla)E. \quad (6)$$

The quantities a_1 and a_2 in Eq. (6) are related to the tensor dielectric constant ϵ_{ik} of the deformed solid, which in the approximation linear in the deformations has the following form:

$$\epsilon_{ik} = \epsilon\delta_{ik} + a_1 u_{ik} + a_2 u_{ii}\delta_{ik}, \quad (7)$$

where u_{ik} is the strain tensor and ϵ is the dielectric constant of the undeformed isotropic solid.

The expression for the volume density of electric force in terms of the effective field is given, as before, by Eq. (3). Comparing Eqs. (3) and (6), we find that the effective and macroscopic fields are connected by the relation

$$E_{eff} = \frac{\rho(\partial\epsilon/\partial\rho)_T - a_1/6}{\epsilon - 1} E. \quad (8)$$

Here use has been made of the fact that a_1 and a_2 satisfy the relation

$$\rho(\partial\epsilon/\partial\rho)_T = -1/3 a_1 - a_2. \quad (9)$$

This follows from Eq. (7) if we note that for isotropic compression

$$u_{ik} = \frac{1}{3} \delta_{ik} \frac{\delta\rho}{\rho}.$$

It can be seen from Eq. (7) that the quantity a_1 characterizes the change of the dielectric properties of the solid under shearing deformations. Therefore it is natural that if we set $a_1 = 0$ in Eq. (8) we get the formula (4) found before for liquids. It is also convenient to write Eq. (8) in the form

$$E_{eff} = E + gP, \quad (10)$$

where

$$g = \frac{4\pi}{\epsilon - 1} \left[\frac{\rho(\partial\epsilon/\partial\rho)_T - a_1/6}{\epsilon - 1} - 1 \right]. \quad (11)$$

The Lorentz-Lorenz formula corresponds to $g = 4\pi/3$, and it can be seen from Eq. (11) that the condition for the Lorentz-Lorenz formula's validity is the following relation between a_1 and ϵ :

$$a_1 = 6[\rho(\partial\epsilon/\partial\rho)_T - 1/3(\epsilon - 1)(\epsilon + 2)]. \quad (12)$$

We also have from Eqs. (9) and (12)

$$a_2 = -3\rho(\partial\epsilon/\partial\rho)_T + 2/3(\epsilon - 1)(\epsilon + 2). \quad (13)$$

It follows from Eq. (12) that if the Lorentz-Lorenz formula holds a nonvanishing value of a_1 , i.e., a dependence of the tensor dielectric constant on shearing deformations, implies a dependence of the Clausius-Mosotti function Φ_{C-M} on the density [in other words, a violation of the condition (5)].

In the more general case when a_1 and ϵ do not satisfy Eq. (12), the factor g is not equal to $4\pi/3$ and is given by Eq. (11).

3. THE EFFECTIVE FIELD IN INHOMOGENEOUS MEDIA AND FOR NONSTATIONARY FIELDS

The sources of the field acting on a particle at a point r_0 include external charges and also all the particles of the medium except the one under consideration. The field produced by these sources at points $r \neq r_0$ is in general not the effective field for particles at points r .

Therefore it is convenient to consider the effective field as a function of two coordinates: $\mathbf{E}_{\text{eff}} = \mathbf{E}_{\text{eff}}(\mathbf{r}_0, \mathbf{r})$, where \mathbf{r}_0 is the position of a particle for which the field in question is the effective field, and \mathbf{r} denotes a point at which we consider this field. Accordingly, the fields $\mathbf{E}_{\text{eff}}(\mathbf{r}_{01}, \mathbf{r})$ and $\mathbf{E}_{\text{eff}}(\mathbf{r}_{02}, \mathbf{r})$ are in general produced by different sources. The quantity found in the preceding section is the field $\mathbf{E}_{\text{eff}}(\mathbf{r}_0, \mathbf{r}_0)$, taken at the point where it is the effective field. The derivative of the effective field that appears in the expression for the volume force density is obviously the quantity

$$\left. \frac{\partial}{\partial r_j} \mathbf{E}_{\text{eff},i}(\mathbf{r}_0, \mathbf{r}) \right|_{\mathbf{r}=\mathbf{r}_0}$$

In a homogeneous medium in a stationary field the effective field $\mathbf{E}_{\text{eff}}(\mathbf{r}_0, \mathbf{r})$ actually does not depend on the first coordinate, and therefore one can set $\mathbf{r} = \mathbf{r}_0$ in the second argument before differentiating, as was done implicitly in the preceding section. In an inhomogeneous medium, however, one must not do this, and the quantities

$$\mathbf{E}_{\text{eff}}(\mathbf{r}_0, \mathbf{r}_0), \left. \frac{\partial}{\partial r_j} \mathbf{E}_{\text{eff},i}(\mathbf{r}_0, \mathbf{r}) \right|_{\mathbf{r}=\mathbf{r}_0}$$

are to a considerable degree independent of each other, since

$$\left. \frac{\partial}{\partial r_j} \mathbf{E}_{\text{eff},i}(\mathbf{r}_0, \mathbf{r}) \right|_{\mathbf{r}=\mathbf{r}_0} \neq \frac{\partial}{\partial r_{0j}} \mathbf{E}_{\text{eff},i}(\mathbf{r}_0, \mathbf{r}_0).$$

The connection between the effective and macroscopic fields is a local relation when spatial dispersion can be neglected, so that even in an inhomogeneous medium the effective field $\mathbf{E}_{\text{eff}}(\mathbf{r}_0, \mathbf{r}_0)$ is connected at each point \mathbf{r}_0 with the macroscopic field $\mathbf{E}(\mathbf{r}_0)$ by the relation (8), in which the dielectric constant ε and the quantity a_1 are now functions of \mathbf{r}_0 . Noting this and using a method for inhomogeneous solids analogous to that used in Sec. 2, we have to find an expression for the quantity

$$\left. \frac{\partial}{\partial r_j} \mathbf{E}_{\text{eff},i}(\mathbf{r}_0, \mathbf{r}) \right|_{\mathbf{r}=\mathbf{r}_0}$$

For an electrostatic field in an inhomogeneous isotropic dielectric the expression connecting the volume force density with the macroscopic field (see Ref. 8, Secs. 15, 16) can be put in the form

$$\mathbf{f} = \mathbf{f}^{(0)} + \frac{1}{4\pi} \left[\rho \left(\frac{\partial \varepsilon}{\partial \rho} \right)_{\mathbf{r}} - \frac{1}{6} a_1 \right] (\mathbf{E}\nabla) \mathbf{E} + \frac{E^2}{8\pi} \nabla \left[\rho \left(\frac{\partial \varepsilon}{\partial \rho} \right)_{\mathbf{r}} - \varepsilon + \frac{1}{3} a_1 \right] - \frac{\mathbf{E}}{8\pi} (\mathbf{E}\nabla) a_1 + \frac{\mathbf{E}}{8\pi \varepsilon} a_1 (\mathbf{E}\nabla) \varepsilon, \quad (14)$$

where $\mathbf{f}^{(0)}$ is the force density that would exist in the medium in the absence of the electric field for the given values of the density, the temperature, and other parameters of the medium. In the case of a liquid the quantity $\mathbf{f}^{(0)}$ is connected with the pressure, $\mathbf{f}^{(0)} = -\nabla p_0(\rho, T)$, and for solids it is determined in terms of the bulk modulus and the modulus of rigidity by the usual formulas of the theory of elasticity.

We can write the connection between the force density and the effective field in an inhomogeneous medium in the form

$$\mathbf{f} = \mathbf{f}^{(0)} + (\mathbf{P}\nabla) \mathbf{E}_{\text{eff}} + \rho_{\text{ind}} \mathbf{E}_{\text{eff}}, \quad (15)$$

and for the bound-charge density ρ_{ind} and the polariza-

tion \mathbf{P} we use the usual relations

$$\rho_{\text{ind}} = \frac{1}{4\pi} \text{div } \mathbf{E} = -\frac{1}{4\pi \varepsilon} (\mathbf{E}\nabla) \varepsilon, \quad \mathbf{P} = \frac{\varepsilon - 1}{4\pi} \mathbf{E}.$$

The force density $\mathbf{f}^{(0)}$ that appears in Eqs. (14) and (15) does not affect the expression for the effective field, so that we shall omit it throughout our further treatment. We find the expression for

$$\left. \frac{\partial}{\partial r_j} \mathbf{E}_{\text{eff},i}(\mathbf{r}_0, \mathbf{r}) \right|_{\mathbf{r}=\mathbf{r}_0}$$

by comparing Eqs. (14) and (15) and using the relation (8). The answer is unique when we take into account the equation $\nabla_{\mathbf{r}} \times \mathbf{E}_{\text{eff}}(\mathbf{r}_0, \mathbf{r}) = 0$. The result is

$$\begin{aligned} \left. \frac{\partial \mathbf{E}_{\text{eff},i}(\mathbf{r}_0, \mathbf{r})}{\partial r_j} \right|_{\mathbf{r}=\mathbf{r}_0} &= \frac{\rho (\partial \varepsilon / \partial \rho)_{\mathbf{r}} - a_1 / 6}{\varepsilon - 1} \frac{\partial E_i}{\partial r_{0j}} + \frac{1}{2(\varepsilon - 1)} \\ &\times \left[E_i \frac{\partial}{\partial r_{0j}} \left(\rho \left(\frac{\partial \varepsilon}{\partial \rho} \right)_{\mathbf{r}} - \varepsilon + \frac{a_1}{3} \right) + E_j \frac{\partial}{\partial r_{0i}} \left(\rho \left(\frac{\partial \varepsilon}{\partial \rho} \right)_{\mathbf{r}} - \varepsilon + \frac{a_1}{3} \right) \right] \\ &+ \frac{\delta_{ij}}{\varepsilon - 1} \left[\frac{\rho (\partial \varepsilon / \partial \rho)_{\mathbf{r}} - a_1 / 6}{\varepsilon (\varepsilon - 1)} E_k \frac{\partial \varepsilon}{\partial r_{0k}} - \frac{1}{2} E_k \frac{\partial}{\partial r_{0k}} \left(\rho \left(\frac{\partial \varepsilon}{\partial \rho} \right)_{\mathbf{r}} - \varepsilon + \frac{a_1}{3} \right) \right. \\ &\left. + \frac{a_1}{2\varepsilon} E_k \frac{\partial \varepsilon}{\partial r_{0k}} - \frac{1}{2} E_k \frac{\partial a_1}{\partial r_{0k}} \right]. \quad (16) \end{aligned}$$

Here the functions E_i , ε , ρ , and a_i of the coordinates are taken at the point \mathbf{r}_0 . Therefore, owing to specific properties, such as the dependence of the effective field $\mathbf{E}_{\text{eff}}(\mathbf{r}_0, \mathbf{r})$ on the two spatial coordinates, the derivative

$$\left. \frac{\partial}{\partial r_j} \mathbf{E}_{\text{eff},i}(\mathbf{r}_0, \mathbf{r}) \right|_{\mathbf{r}=\mathbf{r}_0}$$

of the effective field in an inhomogeneous medium is connected linearly, according to Eq. (16), not only with the derivative of the macroscopic field, but also with that field itself. In this sense the connection between the derivative of the effective field and that of the macroscopic field is nonlocal, whereas the quantities $\mathbf{E}_{\text{eff}}(\mathbf{r}_0, \mathbf{r})$ and $\mathbf{E}(\mathbf{r}_0)$ satisfy the local relation (8).

In a homogeneous medium the connection between the derivatives of the effective field and those of the macroscopic field is complicated in the nonstationary case, when the electric and magnetic fields depend on the time. If this dependence is sufficiently slow, the relation (8) between the fields $\mathbf{E}_{\text{eff}}(\mathbf{r}_0, \mathbf{r}_0)$ and $\mathbf{E}(\mathbf{r}_0)$ remains unchanged, but the expression for the volume force density, to and including terms in the first derivatives of the fields with respect to the time, is of the form (see Ref. 8, Secs. 16 and 56, and also Refs. 9 and 14)

$$\mathbf{f} = -\frac{a_1}{8\pi} (\mathbf{E}\nabla) \mathbf{E} - \frac{a_2}{8\pi} \nabla (E^2) + \frac{\varepsilon - 1}{4\pi c} \frac{\partial}{\partial t} [\mathbf{E} \times \mathbf{B}]. \quad (17)$$

Since the difference between the effective magnetic field \mathbf{B}_{eff} and the macroscopic field \mathbf{B} leads to a term in the Lorentz force that is quadratic in the time derivative of the field, including the difference between \mathbf{B}_{eff} and \mathbf{B} would be an exaggeration of the accuracy of Eq. (17). Therefore we take $\mathbf{B}_{\text{eff}} = \mathbf{B}$, and write the expression connecting the force density and the effective field in the form

$$\mathbf{f} = (\mathbf{P}\nabla) \mathbf{E}_{\text{eff}} + \frac{1}{c} \left[\frac{\partial \mathbf{P}}{\partial t} \times \mathbf{B} \right]. \quad (18)$$

Comparing Eqs. (17) and (18) and using (9), we find the following expression for the derivative of the effective field:

$$\frac{\partial E_{eff,i}(\mathbf{r}_0, \mathbf{r})}{\partial r_j} \Big|_{\mathbf{r}=\mathbf{r}_0} = \frac{\rho(\partial\epsilon/\partial\rho)_T - a_1/6}{\epsilon-1} \frac{\partial E_i}{\partial r_{0j}} + \left[1 - \frac{\rho(\partial\epsilon/\partial\rho)_T + a_1/3}{\epsilon-1} \right] e_{ijk} \frac{1}{c} \frac{\partial B_k}{\partial t}, \quad (19)$$

where e_{ijk} is the completely antisymmetric tensor.

The difference between the derivative of the effective field

$$\frac{\partial}{\partial r_j} E_{eff,i}(\mathbf{r}_0, \mathbf{r}) \Big|_{\mathbf{r}=\mathbf{r}_0}$$

and the quantity

$$\frac{\partial}{\partial r_{0j}} E_{eff,i}(\mathbf{r}_0, \mathbf{r}_0)$$

in a homogeneous dielectric in which an electromagnetic wave is propagated was pointed out by Peierls.⁷ In his paper coefficients σ and τ are used, which are defined in the following way:

$$\frac{\partial}{\partial r_j} E_{eff,i}(\mathbf{r}_0, \mathbf{r}) \Big|_{\mathbf{r}=\mathbf{r}_0} = [1 + \tau(\epsilon-1)] \frac{\partial E_i(\mathbf{r}_0)}{\partial r_{0j}} + \sigma(\epsilon-1) \frac{\partial E_j(\mathbf{r}_0)}{\partial r_{0i}}. \quad (20)$$

Comparing Eqs. (19) and (20) and using the equation

$$\text{rot } \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t},$$

we find

$$\tau = -\frac{a_1}{2(\epsilon-1)^2}, \quad \sigma = \frac{1}{\epsilon-1} \left(\frac{\rho(\partial\epsilon/\partial\rho)_T + a_1/3}{\epsilon-1} - 1 \right). \quad (21)$$

If we assume that the condition (4') holds, as is done in Ref. 7, then we get from Eq. (21) precisely the same formulas for σ and τ as were found in Ref. 7. We can see from Eqs. (19) and (20) that in general the rotation of the effective field is not equal to the quantity $-c^{-1} \partial \mathbf{B} / \partial t$, and at the same time $\mathbf{B}_{eff} = \mathbf{B}$. It follows from Eq. (20) that the equation

$$\text{rot } \mathbf{E}_{eff}(\mathbf{r}_0, \mathbf{r}) \Big|_{\mathbf{r}=\mathbf{r}_0} = -\frac{1}{c} \frac{\partial \mathbf{B}(\mathbf{r}_0)}{\partial t} \quad (22)$$

is true only in the special case when $\sigma = \tau$, which, according to Eq. (21) corresponds to the condition

$$\frac{5}{6} a_1 = (\epsilon-1) - \rho \left(\frac{\partial \epsilon}{\partial \rho} \right)_T.$$

If we assume that Eq. (4') is correct, then from this last condition it follows that $a_1 = -2(\epsilon-1)^2/5$, and then from Eq. (21) we have $\sigma = \tau = 1/5$.

In the case of a liquid, with $a_1 = 0$, it follows from Eq. (21) that $\tau = 0$, and consequently

$$\text{rot } \mathbf{E}_{eff}(\mathbf{r}_0, \mathbf{r}) \Big|_{\mathbf{r}=\mathbf{r}_0} \neq -\frac{1}{c} \frac{\partial \mathbf{B}(\mathbf{r}_0)}{\partial t}.$$

In the article by Peierls⁷ the requirement that under the condition $\mathbf{B}_{eff} = \mathbf{B}$ Eq. (22) should be satisfied is actually used as a criterion in choosing the expression for the volume force density. Therefore in Ref. 7 the expression used for the volume force density is such that $\sigma = \tau$, and in terms of the model used there $\sigma = \tau = 1/5$. We believe this requirement is in general not correct, and consequently that the quantities σ and τ , connected by Eqs. (21) and (7) with the dielectric properties of the medium, may be unequal to each other.

In fact, the time derivatives of effective fields must be dealt with in the same way as their derivatives with

respect to space coordinates, since the sources of the field acting on the particle which is at the point \mathbf{r}_0 at the time t_0 are in general characterized by both space coordinates and by the time t_0 . Accordingly, it is convenient to consider the fields $\mathbf{E}_{eff}(\mathbf{r}_0, \mathbf{r}, t_0, t)$ and $\mathbf{B}_{eff}(\mathbf{r}_0, \mathbf{r}, t_0, t)$, which are the effective fields at $t = t_0$, $\mathbf{r} = \mathbf{r}_0$. In the stationary case this definition actually does not depend on the value of t_0 , as we have implicitly assumed in our discussion so far. In the nonstationary case to be considered now,

$$\mathbf{B}_{eff}(\mathbf{r}_0, \mathbf{r}_0, t_0, t_0) = \mathbf{B}(\mathbf{r}_0, t_0),$$

but the derivative

$$\frac{\partial}{\partial t} \mathbf{B}_{eff}(\mathbf{r}_0, \mathbf{r}_0, t_0, t) \Big|_{t=t_0}$$

may still be unequal to the quantity $\partial \mathbf{B}(\mathbf{r}_0, t_0) / \partial t$. From the equation

$$\text{rot } \mathbf{E}_{eff}(\mathbf{r}_0, \mathbf{r}, t_0, t) \Big|_{\mathbf{r}=\mathbf{r}_0} = -\frac{1}{c} \frac{\partial \mathbf{B}_{eff}(\mathbf{r}_0, \mathbf{r}_0, t, t_0)}{\partial t} \Big|_{t=t_0}$$

we find, using Eq. (19), that

$$\frac{\partial \mathbf{B}_{eff}(\mathbf{r}_0, \mathbf{r}_0, t_0, t)}{\partial t} \Big|_{t=t_0} = \left(2 - \frac{\rho(\partial\epsilon/\partial\rho)_T}{\epsilon-1} \right) \frac{\partial \mathbf{B}(\mathbf{r}_0, t_0)}{\partial t}. \quad (23)$$

Accordingly, in dealing with the field $\mathbf{E}_{eff} - \mathbf{E}$ in the present case it is not sufficient to introduce only a scalar potential, and therefore the parameters σ and τ are in general unequal, and according to Eq. (21) are to a considerable degree independent. Using these conclusions and proceeding otherwise in analogy with Peierls' work,⁷ (see also Refs. 14 and 15), we find without difficulty what change these considerations cause in his results. He considered the problem of the force which an electromagnetic wave exerts on a mirror immersed in a liquid on being reflected from it. Besides the usual expression for the pressure, it was found in Ref. 7 that in the case of oblique incidence with the electric vector of the wave perpendicular to the plane of incidence there is an additional force proportional to the quantity τ and given by Eq. (20). From our present results it follows that in simple liquids, in which the dielectric constant is not altered by shearing deformations (in which case $a_1 = 0$, and according to Eq. (21) $\sigma \neq \tau = 0$ and, as we have explained, this inequality does not involve any contradictions), owing to the fact that $\tau = 0$ the additional force vanishes. For some substances it may be that there is a situation such that in the optical range of frequencies we can set $a_1 = 0$, but for low frequencies $a_1 \neq 0$, so that $\tau \neq 0$. In Ginzburg's paper¹⁵ it was pointed out that in this case it is natural to expect that at low frequencies elastic mechanical properties will appear in the medium, and that these will lead to cancellation of the additional force found in Ref. 7. Therefore, according to Ref. 15, in this case also an additional force may not be observed experimentally.

Peierls⁷ also considered the question of the momentum of an electromagnetic wave field in a dielectric. The total momentum associated with the field and propagated in the dielectric along with the electromagnetic wave consists of two parts: The electromagnetic part of the wave's momentum in the medium, owing to the processes of propagation of the polarization in the dielectric, and the momentum of the mechanical motion

caused by the electric forces in the dielectric. For the total momentum density of a transverse wave in a homogeneous dielectric, the expression found in Ref. 7 is

$$\mathbf{G} = \left[\frac{1+\epsilon}{2} - \frac{\sigma}{2}(\epsilon-1)^2 \right] \frac{1}{4\pi c} [\mathbf{E} \times \mathbf{B}]. \quad (24)$$

Written in this way, the expression remains correct also in the light of the results found here, but there is no need to use estimates from models for the quantity σ ; the more general expression (21) must be used. Then, using Eq. (9), we get

$$\mathbf{G} = \left(\epsilon + \frac{a_2}{2} \right) \frac{1}{4\pi c} [\mathbf{E} \times \mathbf{B}] = \left(\epsilon - \frac{1}{2} \rho \left(\frac{\partial \epsilon}{\partial \rho} \right)_r - \frac{a_1}{6} \right) \frac{1}{4\pi c} [\mathbf{E} \times \mathbf{B}]. \quad (25)$$

Though there is in general simply no relation between the force $f^{(0)}$ which appears in Eqs. (14) and (15) and the determination of the effective field, this is no longer the case when we deal with the question of the total momentum density. Therefore the applicability of the results (24) and (25) depends in an essential way on the assumption that the force $f^{(0)}$ can be neglected in finding the mechanical part of the momentum density, which is due to the ponderomotive action of the field.

Peierls' treatment⁷ is conducted on the assumption that the Lorentz-Lorenz law and the relation (4') are valid. According to Eq. (12), it follows from these two assumptions that $a_1 = 0$, and for $a_1 \neq 0$ the two assumptions contradict each other. It is clear from the present discussion, however, that the results of Peierls⁷ can be considered also without resorting to such model-based estimates for the dielectric constant and the effective field.

For nonstationary fields in an inhomogeneous medium the expressions for the derivatives of effective fields can be found without difficulty, if we note that the formula for

$$\operatorname{div} \mathbf{E}_{eff}(\mathbf{r}_0, \mathbf{r}) \Big|_{r=r_0}$$

must be just what follows from Eq. (16), and the formula for

$$\operatorname{rot} \mathbf{E}_{eff}(\mathbf{r}_0, \mathbf{r}) \Big|_{r=r_0}$$

must be given by using Eq. (19). The results so found can easily be extended to the case of magnetic media, but we shall not write out the expressions here.

We note in conclusion that the method used here to find the expressions for the effective field and its derivatives directly clarifies the relation between the Helmholtz and the Einstein-Laub formulas for the volume force density in the electrodynamics of continuous media (in the former case using the Abraham force, in the latter the Lorentz force. On this matter see, for example, Refs. 9 and 14 and literature cited there).

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