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Microscopic derivation of the force on a dielectric fluid in an electromagnetic field

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The force acting on a Clausius-Mossotti fluid in an electromagnetic field is evaluated microscopically. Owing to the modification of the two-particle density by the electric field, an additional mechanical force $\Delta\vec{f}^{(M)}$ is found. When this is added to the electrical force $\vec{f}^{(E)}$, the total force in the static case becomes identical to that deduced macroscopically by Helmholtz. The analysis is extended to various time-dependent cases, and it is pointed out that $\Delta\vec{f}^{(M)}$ essentially assumes its static value on time scales longer than T_c, the relaxation time of the two-particle density, but is otherwise negligibly small. Thus Peierls's theory of the momentum of light is valid only for pulses much shorter than T_c; the necessary correction due to $\Delta\vec{f}^{(M)}$ in other cases is given and discussed.

I. INTRODUCTION

The force density in a dielectric fluid under the action of an arbitrary electromagnetic field is the subject of a long controversy. Many different points of view and different results, derived either macroscopically or microscopically, have appeared in the literature and can be found in recent reviews.[1,2]

Among the macroscopically derived results, the one first obtained by Helmholtz[3] for the electrostatic case has strong support from both theory[4] and experiment,[2,5] and is now widely accepted. The result may be stated as the following condition for equilibrium:

$$\vec{\nabla}\pi_0(\rho,T) = -\tfrac{1}{2}(\vec{\nabla}\epsilon)\vec{E}^2 + \tfrac{1}{2}\vec{\nabla}\left[\rho\frac{\partial\epsilon}{\partial\rho}\vec{E}^2\right] \equiv \vec{f}^{(H)}, \tag{1}$$

where ϵ and ρ are, respectively, the dielectric constant and the number density of the fluid, $\vec{E}$ is the macroscopic field, $\vec{f}^{(H)}$ is the so-called Helmholtz force density, and $\pi_0(\rho,T)$ should be taken to be the same pressure function of ρ and the temperature T as in the absence of the electric field[4]—a point which is sometimes not made explicit,[6] but which will turn out to be of crucial significance. Generalization of (1) to time-varying situations[4,7,8] has been attempted, but the results often lack a sound justification[9] and a clear statement of the regimes of applicability.

Parallel to the macroscopic consideration, the microscopic approach to the problem, which starts from the fundamental laws at the molecular or atomic level, has been adopted by several authors. For the electrostatic case, attempts to derive (1) microscopically have met with little success and doubts have been raised whether this can really be done microscopically.[1] For the time-varying case, Gordon[8] has succeeded in solving the problem to first order in $(\epsilon-\epsilon_0)$, which amounts to neglecting dipole-dipole interactions. These interactions were later treated by Peierls[10] and others,[11] who also discussed the associated problem of the momentum of light.

The present paper presents a microscopic theory of force density in a dielectric fluid for both the static case and the time-varying case. The work is motivated by the observation that the microscopic viewpoint of Peierls and others[10,11] is also applicable to the static case but does not by itself lead to (1). We show that the presence of an electric field in a dielectric makes two distinct contributions to the force density, both proportional to $\vec{E}^2$: (i) $\vec{f}^{(E)}$, the electrical force density which is essentially that of Peierls generalized to include density inhomogeneity, and (ii) $\Delta\vec{f}^{(M)}$, which is the additional mechanical intermolecular force density due to the modification of the two-particle density by the electric field. We are able to show that, in the static case,

$$\vec{f}^{(H)}=\vec{f}^{(E)}+\Delta\vec{f}^{(M)} ,$$

and thus provide, for the first time, a microscopic justification of (1). This demonstration is given in Sec. II. With the extra term $\Delta\vec{f}^{(M)}$ which was previously neglected,[10,11] a number of time-dependent cases are studied in Sec. III, followed by a discussion on the momentum of light in Sec. IV. Some discussion and concluding remarks are given in Sec. V. The whole paper is restricted to a linear, isotropic, nonpolar, and nonmagnetic fluid which obeys the Clausius-Mossotti equation, and only terms up to $\vec{E}^2$ will be considered. Furthermore, molecular multipole moments of higher order than the dipole shall be neglected.

II. ELECTROSTATIC CASE

We first consider a dielectric fluid with a single species of molecules under the action of an electrostatic field. The dynamical response is described by the exact momentum balance equation

$$\rho\frac{d}{dt}(m\vec{V})+\vec{\nabla}\cdot\overleftrightarrow{\pi}^{(K)}=\rho\vec{F}(\vec{r}) + \int\vec{X}_{12}\rho^{(2)}(\vec{r},\vec{r}\,')d\vec{r}\,' , \qquad (2)$$

where ρ is the number density, m is the mass of a molecule, $\vec{V}$ is the macroscopic fluid velocity, $\overleftrightarrow{\pi}^{(K)}$ is the kinetic pressure stress, $\vec{F}(\vec{r})$ is the external force on a particle at $\vec{r}$, $\vec{X}_{12}$ is the intermolecular force acting on a molecule at $\vec{r}$ due to another molecule at $\vec{r}\,'$, and $\rho^{(2)}$ is the two-particle density, normalized to

$$\int\rho^{(2)}(\vec{r},\vec{r}\,')d\vec{r}\,'=(N-1)\rho(\vec{r}) ,$$

with N being the total number of particles in the system.

The force $\vec{X}$ consists of an electric part $\vec{X}^{(E)}$ due to the dipole-dipole interaction and a mechanical short-range part $\vec{X}^{(S)}$ which may be described by a potential $\Phi^{(S)}(\,|\vec{r}-\vec{r}\,'|\,)$ satisfying $\Phi^{(S)}(0)=\infty$ and $\Phi^{(S)}(\infty)=0$. The external force is $(\vec{\mu}\cdot\vec{\nabla})\vec{E}^{(0)}$, where $\vec{\mu}$ is the dipole moment of the molecule at $\vec{r}$, and $\vec{E}^{(0)}$ is the external field. (Nonelectromagnetic external forces, if any, do not affect the following derivation and will be ignored for convenience.) We write the two-particle density as

$$\rho^{(2)}=\rho_0^{(2)}+\Delta\rho^{(2)} ,$$

where $\rho_0^{(2)}$ is the two-particle density taken to be the same function of ρ and T as in the absence of the electromagnetic field, and $\Delta\rho^{(2)}$ is the change caused by the field. Then (2) can be put into the following form:

$$\rho\frac{d}{dt}(m\vec{V})+\left[\vec{\nabla}\cdot\overleftrightarrow{\pi}^{(K)}-\int\vec{X}_{12}^{(S)}\rho_0^{(2)}d\vec{r}\,'\right]=\vec{f}^{(E)}+\Delta\vec{f}^{(M)} \qquad (3)$$

where

$$\vec{f}^{(E)}=(\vec{P}\cdot\vec{\nabla})\vec{E}^{(0)}+\int\vec{X}_{12}^{(E)}\rho_0^{(2)}d\vec{r}\,' , \qquad (4)$$

with the polarization $\vec{P}=\rho\vec{\mu}$, and

$$\Delta\vec{f}^{(M)}=\int\vec{X}_{12}^{(S)}\Delta\rho^{(2)}d\vec{r}\,' . \qquad (5)$$

In the derivation, we have neglected terms of the order higher than $\vec{E}^2$, noting that $\vec{\mu}$ is first order in $\vec{E}$, and $\vec{X}^{(E)}$ is second order in $\vec{E}$. Both $\vec{f}^{(E)}$ and $\Delta\vec{f}^{(M)}$ are caused by the field, but it is clear that $\vec{f}^{(E)}$ is entirely an electric force, while $\Delta\vec{f}^{(M)}$ is the additional mechanical force density due to the modification of the two-particle density by the field.

To evaluate $\vec{f}^{(E)}$, we note that the equilibrium field-free $\rho_0^{(2)}$ may be written as

$$\rho_0^{(2)}=\rho(\vec{r})\rho(\vec{r}\,')\exp[-\beta U(\,|\vec{r}-\vec{r}\,'|\,)] , \qquad (6)$$

where $\beta\equiv(kT)^{-1}$, and U is the potential of the mean force on a molecule at $\vec{r}$ with a second molecule at $\vec{r}\,'$. U is not equal to $\Phi^{(S)}$, in general. If we split it into

$$\rho_0^{(2)}=\rho(\vec{r})\rho(\vec{r}\,') -\rho(\vec{r})\rho(\vec{r}\,')[1-\exp(-\beta U)] , \qquad (7)$$

and note that the explicit form for $\vec{X}^{(E)}$ is

$$X_i^{(E)}=-\frac{\mu_j(\vec{r})\mu_k(\vec{r}\,')}{4\pi\epsilon_0}\nabla_k'\nabla_j\left[\frac{\xi_i}{\xi^3}\right] , \qquad (8)$$

where

$$\vec{\xi}\equiv\vec{r}\,'-\vec{r} , \qquad (9)$$

Eq. (4) becomes

$$f_i^{(E)}=(\vec{P}\cdot\vec{\nabla})E_i +\frac{P_j}{4\pi\epsilon_0}\int(1-e^{-\beta U})\vec{P}(\vec{r}\,')\cdot\vec{\nabla}'\nabla_j\left[\frac{\xi_i}{\xi^3}\right]d\vec{r}\,' , \qquad (10)$$

where

$$\vec{E}=\vec{E}^{(0)}+\frac{1}{4\pi\epsilon_0}\int\vec{P}(\vec{r}\,')\cdot\vec{\nabla}'\frac{\vec{r}-\vec{r}\,'}{|\vec{r}-\vec{r}\,'|^3}d\vec{r}\,' \tag{11}$$

is the field produced by external charges and the average polarization, and is therefore just the usual macroscopic field. The separation of $\vec{f}^{(E)}$ into two terms in (10), corresponding to the separation of $\rho_0^{(2)}$ in (7), is nothing but the usual "plug subtraction" prescription for calculating the effective field $\vec{E}_{\text{eff}}$ here established with greater precision and moreover generalized to the effective-field gradient. The important point is that by using the macroscopic field $\vec{E}$ rather than the external field $\vec{E}^{(0)}$ [compare the first term of (4) and (10)], all long-range effects are already taken into account, and the remaining plug contribution is from nearby sources only. This observation will be of some importance when we come to time-dependent cases.

The remaining integral in (10) is evaluted in Appendix A. The result is

$$\frac{1}{5\epsilon_0}(P_i\nabla_jP_j+P_j\nabla_iP_j+P_j\nabla_jP_i)\ , \tag{12}$$

which is independent of U provided that the range of U is macroscopically negligible. Putting this into (10) and using $\vec{P}=(\epsilon-\epsilon_0)\vec{E}$, we obtain after a little algebra,

$$f_i^{(E)}=-\tfrac{1}{2}(\nabla_i\epsilon)\vec{E}^2+\nabla_j\left[\tfrac{1}{2}(\epsilon-\epsilon_0)\vec{E}^2\delta_{ij}+\frac{(\epsilon-\epsilon_0)^2}{\epsilon_0}(\tfrac{1}{5}E_iE_j+\tfrac{1}{10}\vec{E}^2\delta_{ij})\right]-(\epsilon-\epsilon_0)[\vec{E}\times(\vec{\nabla}\times\vec{E})]_i\ . \tag{13}$$

The last term is, of course, zero in the static case, but we nevertheless retain it here for the purpose of later generalizing to the time-dependent case.

This force density is the same as the electrical force arising from $(\vec{\mu}\cdot\vec{\nabla})\vec{E}_{\text{eff}}$ found by Peierls[10] in the homogeneous case. However, the E_iE_j term in (13) implies that there can be a tangential surface force on the surface of a dielectric medium. This is already a signal that $\vec{f}^{(E)}$ cannot be the only force caused by the presence of the field, since such a tangential force, if not canceled by some other contribution, would be unphysical since it cannot be balanced by pressure forces.

To evaluate $\Delta\vec{f}^{(M)}$, we note that $\rho^{(2)}$ is an equilibrium two-particle density. Furthermore, $\vec{X}^{(S)}$ in (5) is a short-range force which is only significant at a distance of the order of a molecular size, and at such small distances the static two-particle density is determined mainly by the bare interparticle potential without modification by the presence of other particles. We may, therefore, write

$$\rho^{(2)}(\vec{r},\vec{r}\,')=\rho(\vec{r})\rho(\vec{r}\,')\exp[-\beta(\Phi^{(S)}+\Phi^{(E)})] \tag{14}$$

so that, to order $\vec{E}^2$,

$$\Delta\rho^{(2)}=-\beta\rho(\vec{r})\rho(\vec{r}\,')\Phi^{(E)}\exp(-\beta\Phi^{(S)})\ , \tag{15}$$

where

$$\Phi^{(E)}=\mu_j(\vec{r})\mu_k(\vec{r}\,')(\xi^2\delta_{jk}-3\xi_j\xi_k)/4\pi\epsilon_0\xi^5 \tag{16}$$

is the potential due to the dipole-dipole interaction. Putting (15) and (16) into (5), and changing $\vec{r}\,'$ to $\vec{r}+\vec{\xi}$, we have

$$\Delta f_i^{(M)}=\frac{P_j(\vec{r})}{4\pi\epsilon_0}\int P_k(\vec{r}+\vec{\xi})(\xi^2\delta_{jk}-3\xi_j\xi_k)\times\left[\frac{\xi_i}{\xi^6}\right]\frac{d}{d\xi}e^{-\beta\Phi^{(S)}}d\vec{\xi}\ . \tag{17}$$

The integral can be performed in a similar way as in Appendix A, and leads to

$$\Delta f_i^{(M)}(\vec{r})=\nabla_j\left[\frac{(\epsilon-\epsilon_0)^2}{\epsilon_0}(-\tfrac{1}{5}E_iE_j+\tfrac{1}{15}\vec{E}^2\delta_{ij})\right] \tag{18}$$

which again contains a tangential surface force across dielectric boundaries but is nevertheless expressible as a divergence of a tensor, the latter property being a consequence of the internal origin of the force density.

It is of interest to point out that $\Delta\vec{f}^{(M)}$, in its final form, is independent of the details of $\Phi^{(S)}$. This may at first appear surprising since short-range collisional effects would normally go as R^3T, R being a typical molecular dimension (see, for example, Appendix B). The point is that the dipole-dipole interaction modifies $\rho^{(2)}$ by an extra term proportional to $\beta\Phi^{(E)}\sim(R^3T)^{-1}$, and this is the basic physical reason why $\Delta\vec{f}^{(M)}$ assumes such a simple form.

With the explicit form in (18), we now go back to (3). In the static situation the momentum distribution is Maxwellian, so by definition of $\rho_0^{(2)}$ the quantity in large parentheses in (3) has the property that it is the same function of ρ and T as in the case of zero field. In this trivial case ($\vec{f}^{(E)}=\Delta\vec{f}^{(M)}=0$), the large parentheses must be the gradient of the total pressure, namely, $\vec{\nabla}\pi_0$, and by the property just mentioned, it remains $\vec{\nabla}\pi_0$ even when there is an electromagnetic field. Although this argument is completely general, we present an independent derivation in Appendix B, where π_0 is explicitly evaluated and shown to be in agreement with the standard formula. The right-hand side of (3) can now be obtained by adding (13) and (18) and dropping the $\vec{\nabla}\times\vec{E}$ term; note that the E_iE_j term, and with it the tangential surface force, exactly cancels:

$$\vec{f}^{(E)}+\Delta\vec{f}^{(M)}=-\tfrac{1}{2}(\vec{\nabla}\epsilon)\vec{E}^2+\nabla\left[\left[\tfrac{1}{2}(\epsilon-\epsilon_0)+\frac{1}{6}\frac{(\epsilon-\epsilon_0)^2}{\epsilon_0}\right]\vec{E}^2\right] . \tag{19}$$

For a fluid satisfying the Clausius-Mossotti relation, it is now a straightforward matter to show that (19) is, in fact, identical to $\vec{f}^{(H)}$ in (1). Thus, we have achieved a microscopic understanding of the Helmholtz force density. This is of considerable interest in itself, but more importantly, the success of the derivation validates the microscopic point of view and allows us to go on to time-dependent cases with confidence.

III. TIME-DEPENDENT CASES

To generalize to time-dependent cases, it is necessary to take into account (i) time dependence of the electric field, (ii) the effect of the magnetic field, and (iii) the dynamical response of the fluid. The first two effects modify $\vec{f}^{(E)}$ (now defined to be the electromagnetic force density), while the last will be relevant for $\Delta\vec{f}^{(M)}$. Although the first two effects are not novel,[10,11] it is nevertheless profitable to spell out in some detail the physical justifications for the very simple results that are obtained.

A. Time dependence of electric field

When the electric field is time dependent, the effect of one dipole on another [e.g., (8) and (16)] must be evaluated taking retardation into account. However, the contributions from the distant dipoles are treated exactly—including retardation—by expressing the result in terms of the macroscopic field. The remaining effects (i.e., the plug term), being of a short-range nature, do not suffer any appreciable retardation since we shall be concerned exclusively with electromagnetic wavelengths λ much greater than molecular dimensions R. Thus, the contributions of electrical forces to $\vec{f}^{(E)}$ remains the same as in (13).

Of course when the fields are time dependent, the last term in (13) is no longer zero, but becomes

$$(\epsilon-\epsilon_0)\vec{E}\times\frac{\partial\vec{B}}{\partial t} . \tag{20}$$

B. Magnetic field

The magnetic field exerts a force on each dipole given by

$$\frac{\partial\vec{\mu}}{\partial t}\times\vec{B}_{\rm eff} ,$$

where $\vec{B}_{\rm eff}$ is the effective (i.e., actual rather than spatially averaged) field at the position of the dipole, due to all sources other than the dipole itself. We shall shortly see that $\vec{B}_{\rm eff}$ may be replaced by the macroscopic field $\vec{B}$, and this leads to a force density

$$\frac{\partial\vec{P}}{\partial t}\times\vec{B}=(\epsilon-\epsilon_0)\frac{\partial\vec{E}}{\partial t}\times\vec{B} , \tag{21}$$

and when this is combined with (20), we get

$$(\epsilon-\epsilon_0)\frac{\partial}{\partial t}(\vec{E}\times\vec{B}) . \tag{22}$$

The differences between $\vec{B}_{\rm eff}$ and $\vec{B}$ is directly analogous to that between $\vec{E}_{\rm eff}$ and $\vec{E}$ and may be accounted for by performing the plug subtraction, which is due to the currents, namely, $\partial\vec{P}/\partial t$, in the plug. But it is not difficult to see that a plug of dimension R whose polarization $\vec{P}$ is oscillating at a frequency ω produces a near-zone magnetic field proportional to $\omega R/c\sim R/\lambda$, which is therefore negligible. This is the reason why $\vec{B}_{\rm eff}$ can be freely replaced by $\vec{B}$ (while $\vec{E}_{\rm eff}$ cannot be replaced by $\vec{E}$).

C. Response of the fluid

To discuss the fluid response it is necessary to pay attention to several time scales in the problem. Time-dependent fields may, in general, be characterized by the field oscillation period T_p and the intensity variation time of the pulse T_I, with $T_p << T_I$. The particle distributions respond over certain relaxation times. In particular, the two-particle density relaxes in a time which may be identified with the time in a collision T_c. For example, at room temperatures, T_c is typically 10^{-12} s for a liquid. Depending on the magnitude of T_c compared with T_p and T_I, three cases may be distinguished which are amenable to simple treatment: (1) $T_c << T_p << T_I$, (2) $T_p << T_c << T_I$, and (3) $T_p << T_I << T_c$. There is, in fact, a fourth characteristic time, namely, the time required for an individual dipole to respond to the electric field, but for induced dipoles this is limited only by the inertia of electrons and can usually be neglected. This amounts to assuming ϵ is real, or physically that the medium is transparent.

(1) $T_c << T_p << T_I$. Such situations may be described as quasistatic, in the sense that the two-particle density assumes the equilibrium functional form under the instantaneous fields. The results of the previous section [now including the $(\epsilon-\epsilon_0)\partial(\vec{E}\times\vec{B})/\partial t$ term] can be applied at every instant, i.e., the total force density $\vec{f}$ caused by the field or the right-hand side of (3) is

$$\vec{f}=-\tfrac{1}{2}(\vec{\nabla}\epsilon)\vec{E}^2+\vec{\nabla}\left[\left[\tfrac{1}{2}(\epsilon-\epsilon_0)+\frac{1}{6}\frac{(\epsilon-\epsilon_0)^2}{\epsilon_0}\right]\vec{E}^2\right]+(\epsilon-\epsilon_0)\frac{\partial}{\partial t}(\vec{E}\times\vec{B}) . \tag{23}$$

(2) $T_p << T_c << T_I$. When the fields oscillate much faster than the fluid can respond, the molecules are effectively experiencing time averaged dipole-dipole forces which vary on a time scale of T_I. In every time interval T_c, the averaged forces may be considered constant and the two-particle density takes the equilibrium value given by (14), with $\Phi^{(E)}$ replaced by its time-averaged value. The evaluation of $\Delta\vec{f}^{(M)}$ is thus unaffected. The total force density is, therefore,

$$\vec{f}=-\tfrac{1}{2}(\vec{\nabla}\epsilon)\langle\vec{E}^2\rangle_T+\vec{\nabla}\left[\left[\tfrac{1}{2}(\epsilon-\epsilon_0)+\frac{1}{6}\frac{(\epsilon-\epsilon_0)^2}{\epsilon_0}\right]\langle\vec{E}^2\rangle_T\right]+(\epsilon-\epsilon_0)\frac{\partial}{\partial t}\langle\vec{E}\times\vec{B}\rangle_T , \tag{24}$$

where $\langle\ \rangle_T$ denotes averaging over T_c.

(3) $T_p << T_I << T_c$. In this case, the evaluation of $\vec{f}^{(E)}$ is the same as in case (2). But during the time of the pulse T_I, the two-particle density does not have enough time to respond to the fields but essentially retains its original value in the absence of the fields. In particular, $\Delta\rho^{(2)}=0$ and hence $\Delta\vec{f}^{(M)}=0$, and

$$f_i=f_i^{(E)}=-\tfrac{1}{2}(\nabla_i\epsilon)\vec{E}^2+\nabla_j\left[\tfrac{1}{2}(\epsilon-\epsilon_0)\vec{E}^2\delta_{ij}+\frac{(\epsilon-\epsilon_0)^2}{\epsilon_0}(\tfrac{1}{5}E_iE_j+\tfrac{1}{10}\vec{E}^2\delta_{ij})\right]+(\epsilon-\epsilon_0)\frac{\partial}{\partial t}(\vec{E}\times\vec{B})_i , \tag{25}$$

which agrees with the result of Peierls[10] in the homogeneous case.

The above do not exhaust all possibilities; however, other cases cannot be treated without solving the time-dependent kinetic equations dynamically. Perhaps the most important point of this section is the distinction between cases (2) and (3), which may be summarized as follows: Of the two forces $\vec{f}^{(E)}$ and $\Delta\vec{f}^{(M)}$, the latter depends on a change in the two-particle density and therfore does not assume the value given in (18) until a time of order T_c.

IV. MOMENTUM OF LIGHT

An electromagnetic wave traveling in vacuum carries a momentum density

$$\vec{g}'_{em}=\epsilon_0\vec{E}'\times\vec{B}' , \tag{26}$$

where $\vec{E}'$ and $\vec{B}'$ are the microscopic fields, whose spatial averages are the macroscopic fields $\vec{E}$ and $\vec{B}$. If $\vec{g}_{em}$ is the spatial average (denoted by $\langle\ \rangle_S$) of $\vec{g}'_{em}$, then

$$g_{em}=\epsilon_0\vec{E}\times\vec{B}+\epsilon_0\langle\delta\vec{E}\times\delta\vec{B}\rangle_S ,$$

where $\delta\vec{E}$ and $\delta\vec{B}$ are the fluctuations

$$\delta\vec{E}=\vec{E}'-\vec{E},\quad \delta\vec{B}=\vec{B}'-\vec{B} .$$

If spatial averages are defined over a typical distance L which is microscopically large ($R << L$) and macroscopically small ($L << \lambda$), then it is clear that $\delta\vec{E}$ and $\delta\vec{B}$ are produced only by sources within a distance of order L. But since $L << \lambda$, all such contributions may be evaluated quasistatically, i.e., in a manner which makes no reference to the direction of wave propagation. In the absence of any preferred direction, clearly $\langle \delta\vec{E}\times\delta\vec{B}\rangle_S=0$, and

$$\vec{g}_{em}=\epsilon_0\vec{E}\times\vec{B} . \tag{27}$$

Now consider a plane wave traveling in the z direction,

$$\vec{E}\times\vec{B}=\frac{n}{c}\vec{E}^2\vec{e}_z ,$$

where n is the refractive index. It is convenient to average over several cycles and write the result in terms of the intensity I:

$$I=\epsilon_0 nc\langle\vec{E}^2\rangle_T ,$$

and thus

$$\vec{g}_{em}=(I/c^2)\vec{e}_z . \tag{28}$$

Equation (27), or equivalently Eq. (28), was first proposed by Abraham[12] and is widely accepted as the correct expression for the momentum residing in the electromagnetic field.[8,10,13]

However, there is some momentum $\vec{g}_{med}$ residing in the medium which nevertheless travels along with the wave, and one is often interested in the total momentum density

$$\vec{g}=\vec{g}_{em}+\vec{g}_{med} . \tag{29}$$

With the knowledge of the force acting on the medium, it is now straightforward to compute $\vec{g}_{med}$ and hence $\vec{g}$. The pressure force is of no concern here, because the momentum associated with the acoustic pulse propagates much slower than the electromagnetic pulse and can therefore be separated out.

Cases (1) and (2). Although case (1) is not normally encountered in the case of light, it is formally the same as case (2) and will therefore be treated together. Inside a uniform medium ($\vec{\nabla}\epsilon=0$) and averaging over several cycles, both (23) and (24) become

$$f_z=[\tfrac{1}{2}(n^2-1)+\tfrac{1}{6}(n^2-1)^2]\frac{1}{nc}\frac{\partial I}{\partial z}+(n^2-1)\frac{1}{c^2}\frac{\partial I}{\partial t}$$
$$=[\tfrac{1}{2}(n^2-1)-\tfrac{1}{6}(n^2-1)^2]\frac{1}{c^2}\frac{\partial I}{\partial t} . \tag{30}$$

This is positive (for n not too large) at the leading edge of the pulse and imparts a positive momentum to the medium. The negative force at the trailing edge of the pulse exactly removes this momentum and (pressure waves aside) returns the medium to rest. Thus, the momentum density in the medium travels with the pulse, and the magnitude of the momentum can be obtained by integrating (30) from the infinite past:

$$\vec{g}_{med}=[\tfrac{1}{2}(n^2-1)-\tfrac{1}{6}(n^2-1)^2](I/c^2)\vec{e}_z , \tag{31}$$

so

$$\vec{g}=[\tfrac{1}{2}(n^2+1)-\tfrac{1}{6}(n^2-1)^2](I/c^2)\vec{e}_z . \tag{32}$$

This result applies to most experimental situations, e.g., nanosecond light pulses incident on a liquid.

Case (3). The force density is now given by (25), but for the force along the direction of propagation, E_iE_j does not contribute since the wave is transverse. The only difference from cases (1) and (2) is that the term $\frac{1}{6}E^2\delta_{ij}$ becomes $\frac{1}{10}E^2\delta_{ij}$, and consequently

$$\vec{g}=[\tfrac{1}{2}(n^2+1)-\tfrac{1}{10}(n^2-1)^2](I/c^2)\vec{e}_z . \tag{33}$$

This was the result obtained by Peierls.[10,11] Our derivations however, makes it clear that (33) is applicable only for very short pulses, e.g., for liquids with $T_c\sim10^{-12}$ s, the pulse length must be shorter than, for example, 10^{-13} s, which, though theoretically possible, is not yet feasible in practice.

All these results are strictly valid only for plane waves. For waves of finite breadth, $\langle\vec{E}^2\rangle_T$ falls off at the edges of the beam and either (23), (24), or (25) implies *lateral* forces in these regions.[8,11,14] These forces result in an extra momentum density in the form of a pressure wave, which is *not* included in (32) or (33). Experiments meant to test these equations must be designed to be able to segregate $\vec{g}$ (which travels at c/n) from pressure waves (which travels at acoustical velocity), a crucial point which is sometimes overlooked.[15] Moreover, in cases where two beams overlap (e.g., a beam together with its own reflection), it is not, in general, correct to simply add up the individual momentum densities[15] on account of interference effects.[10] In such cases it is perhaps more illuminating to work directly from the force density (24) or (25) rather than to resort to the concept of the momentum density of light.

The above derivation of the momentum density relies on the knowledge of terms of the form $\vec{\nabla}(E^2)$ in $\vec{f}$. To close this section we present an alternative derivation which relies on terms of the type $(\vec{\nabla}\epsilon)$ in $\vec{f}$,[11] thus providing a nice self-consistency check on $\vec{f}$.

Consider a beam normally incident onto the dielectric fluid from vacuum. If the incident, reflected and transmitted momentum fluxes are ϕ_i, ϕ_r, and ϕ_{tr}, then

$$\phi_{tr}=\phi_i+\phi_r-F_s \ . \qquad (34)$$

In (34) F_s is the force per unit area pushing into the surface:

$$F_s=-\int_{-\delta}^{\delta}\vec{n}\cdot\vec{f}\,dz \ ,$$

where $z=0$ is the surface and $\vec{n}$ is the outward normal of the fluid. The only relevant terms in f are, for case (2),

$$\vec{f}=\vec{\nabla}\left[-\tfrac{1}{2}\epsilon+\tfrac{1}{2}(\epsilon-\epsilon_0)+\frac{1}{6}\frac{(\epsilon-\epsilon_0)^2}{\epsilon_0}\right]\langle\vec{E}^2\rangle_T$$

leading to

$$F_s=\frac{1}{6}\frac{(\epsilon-\epsilon_0)^2}{\epsilon_0}\langle\vec{E}^2\rangle_T \ ,$$

where now ϵ refers to the value in the fluid. $\langle\vec{E}^2\rangle_T$ is continuous across the boundary and may be thought of as the value in the transmitted beam. In an obvious notation

$$\phi_i=cg_i=\epsilon_0\langle\vec{E}_i^2\rangle_T=\epsilon_0\left[\frac{n+1}{2}\right]^2\langle\vec{E}^2\rangle_T \ ,$$

$$\phi_r=cg_r=\epsilon_0\langle\vec{E}_r^2\rangle_T=\epsilon_0\left[\frac{n-1}{2}\right]^2\langle\vec{E}^2\rangle_T \ ,$$

and putting all these into (34) gives

$$\phi_{tr}=\frac{c}{n}g_{tr}$$

$$=\epsilon_0[\tfrac{1}{2}(n^2+1)-\tfrac{1}{6}(n^2-1)^2]\langle\vec{E}^2\rangle_T \qquad (35)$$

from which (32) for $\vec{g}$ in the transmitted beam follows immediately. The derivation for case (3) is identical, with $\frac{1}{6}$ replaced by $\frac{1}{10}$.

V. DISCUSSION AND CONCLUSION

The Clausius-Mossotti relation is usually derived for a homogeneous medium ($\vec{\nabla}\rho=0$) and a few words on its more general applicability are in order. These remarks should also clarify how our calculation of $\vec{f}^{(E)}$ is related to the effective-field gradient[10,11] and how this is, in turn, related to the concept of the effective field familiar from the usual derivation of the Clausius-Mossotti relation.

Consider a dipole with its center at $\vec{r}$. The basic ingredient of the Clausius-Mossotti relation and of our derivation of $\vec{f}^{(E)}$ is the effective field (i.e., the field due to all sources except the dipole at $\vec{r}$) at a point $\vec{r}+\vec{\eta}$ near $\vec{r}$:

$$\vec{E}_{eff}(\vec{r}+\vec{\eta})=\vec{E}^{(0)}(\vec{r}+\vec{\eta})+\frac{1}{\rho(\vec{r})}\int\left[-\vec{\nabla}\frac{\vec{\mu}(\vec{r}\,')\cdot(\vec{r}+\vec{\eta}-\vec{r}\,')}{4\pi\epsilon_0|\vec{r}+\vec{\eta}-\vec{r}\,'|^3}\right]\rho^{(2)}(\vec{r},\vec{r}\,')d\vec{r}\,' \ , \qquad (36)$$

where $\vec{E}^{(0)}$ is the external field and the second term is obviously the field due to all the other dipoles at various points $\vec{r}\,'$. Since we are interested in contributions linear in $\vec{E}$, $\rho^{(2)}(\vec{r},\vec{r}\,')$ may be replaced by $\rho_0^{(2)}(\vec{r},\vec{r}\,')$ as given in (6) or (7). Thus (36) reduces to

$$\vec{E}_{eff}(\vec{r}+\vec{\eta})=\vec{E}(\vec{r}+\vec{\eta})+\frac{1}{4\pi\epsilon_0}\int(1-e^{-\beta U(\xi)})\vec{P}(\vec{r}\,')\cdot\vec{\nabla}'\left[\frac{\vec{\xi}-\vec{\eta}}{|\vec{\xi}-\vec{\eta}|^3}\right]d\vec{r}\,' \ , \qquad (37)$$

where $\vec{\xi}=\vec{r}\,'-\vec{r}$, and $\vec{E}$ is the macroscopic field given in (11). For deriving the Clausius-Mossotti equation we only need $\vec{E}_{eff}$ right at the center of the dipole, so putting $\vec{\eta}=0$ and evaluating (37) in the same way as the first integral in (A1), we get the usual result

$$\vec{E}_{eff}(\vec{r})=\vec{E}(\vec{r})+\vec{P}(\vec{r})/3\epsilon_0 \qquad (38)$$

whether or not the medium is homogeneous.

On the other hand, for the purpose of evaluating the electrical force on the dipole, we need $(\nabla_i E_j)_{eff}$ at $\vec{\eta}=0$, which should be calculated by differentiating (37) and evaluating the result at $\vec{\eta}=0$. This is basically what we did in Sec. II. [Compare the last term in (10) with the derivative of the last term in

(37)]. What should not be done[2] is to differentiate (38), since this equation holds at only one point, and to equate the derivatives of the two sides would be totally incorrect. Had one attempted this, one would have obtained the following erroneous result for the static case:

$$\vec{f}^{(E)}=(\vec{P}\cdot\vec{\nabla})(\vec{E}+\vec{P}/3\epsilon_0) \ .$$

Additional points

(1) The momentum of light for a dispersive but nonabsorbing medium can be treated most simply via the surface force as in Eqs. (34) to (35). In such a derivation, there is no need to "switch on" the intensity and one can therefore imagine that the wave is strictly monochromatic, so that dispersion has absolutely no effect. The only difference occurs in (35), where now

$$\phi_{tr}=\frac{c}{n_g}g_{tr} \ ,$$

where n_g is the so-called group refractive index, and c/n_g is the group velocity. This shows that g given in (32) and (33) should, in general, be corrected by multiplying by n_g/n.

To derive the same result by integrating the force density from the infinite past [as in Eq. (30) to (32)] is somewhat more tedious, since it is now no longer permissible to ignore the adiabatic switching on and the consequent nonzero width in the frequency distribution, as well as the variation of ϵ over this distribution. We have, however, verified that exactly the same correction factor is obtained.

(2). The fact that the medium has a momentum density $\vec{g}_{med}$ [e.g., Eq. (31)] means that it must also have a corresponding energy flux

$$\vec{S}_{med}=\vec{g}_{med}c^2$$

representing the transport of rest energy. Since the purely electromagnetic energy flux $\vec{S}_{em}$ and momentum density $\vec{g}_{em}$ satisfy a similar relation, so do the total energy flux $\vec{S}$ and total momentum density $\vec{g}$:

$$\vec{S}=\vec{g}c^2 \ ,$$

and the total energy-momentum tensor is therefore symmetric.

(3) It has been brought to our attention that one-tenth picosecond light pulses has recently been reported,[17] which may eventually permit an experimental test of case (3).

In conclusion, we have presented a microscopic theory of the force density in a dielectric fluid under the action of an arbitrary electromagnetic field. The force caused by the field consists of two distinctive parts. The first, denoted by $\vec{f}^{(E)}$, is an electromagnetic force while the second, denoted by $\Delta\vec{f}^{(M)}$ in (5), is an additional mechanical force due to the modification of the two-particle density by the field. In the static case, these two forces add up to give the Helmholtz force in (1). Equation (1) is sometimes interpreted[6] as the balance between a mechanical force $-\vec{\nabla}\pi_0$ and an electrical force $\vec{f}^{(H)}$; the present work shows this to be incorrect. In fact, the balance is between the mechanial force $-\vec{\nabla}\pi_0+\Delta\vec{f}^{(M)}$ and the electrical force $\vec{f}^{(E)}$. To the best of our knowledge, this is the first time that such a microscopic derivation has been given. In the quasistatic case, where the field oscillation period is much longer than the relaxation time T_c for the two-particle density, the force density is given by (23). This expression has, in fact, appeared in the literature,[16] but there seems to be no reliable derivation and no precise specification of the regime of applicability. In the case of a rapidly oscillating field with a slowly varying intensity, the force density is given by (24). Most experiments with optical pulses should fall into this category. Finally, for a wave pulse much shorter than T_c, the two-particle density remains essentially unchanged and the force is simply $\vec{f}^{(E)}$. Peierls's theory of the momentum of light is valid only in this last case.

Thus, the understanding of this age-old problem has been enhanced and unified, not only in terms of the different time scales involved, but also in terms of the two different approaches, i.e., thermodynamic versus microscopic.

APPENDIX A: EVALUATION OF THE INTEGRAL IN (10)

Taking ∇_j out of the integral, we have

$$\frac{P_j}{4\pi\epsilon_0}\left[\nabla_j\int(1-e^{-\beta U})P_k(\vec{r}\,')\nabla'_k\left[\frac{\xi_i}{\xi^3}\right]d\vec{r}\,' - \int\nabla_j(1-e^{-\beta U})P_k(\vec{r}\,')\nabla'_k\left[\frac{\xi_i}{\xi^3}\right]d\vec{r}\,'\right] . \quad \text{(A1)}$$

Now since $U(|\vec{r}-\vec{r}\,'|)$ is a short-range potential we may let

$$\vec{P}(\vec{r}\,')=\vec{P}(\vec{r})+(\vec{\xi}\cdot\vec{\nabla})\vec{P}(\vec{r}) \qquad \text{(A2)}$$

in the integrand, where the neglected terms are of the order of R^2/L^2 or higher, R being the molecular size, and L the characteristic macroscopic length. We then notice that only the first term in (A2) contributes to the first integral in (A1), whereas only the second term contributes to the second integral. Transforming $d\vec{r}\,'$ to $d\vec{\xi}$ and $\vec{\nabla}'$ to $\partial/\partial\vec{\xi}$, the first integral may be simplified by an integration by parts and the second integral may be more easily handled by putting $\nabla_j=-\partial/\partial\xi_j$. Equation (A1) then becomes

$$\frac{P_j}{4\pi\epsilon_0}\left[\nabla_j P_k \int \frac{\xi_i}{\xi^3}\frac{\partial}{\partial\xi_k}e^{-\beta U}d\vec{\xi} - \nabla_l P_k \int \frac{\xi^2\delta_{ik}-3\xi_i\xi_k}{\xi^5}\xi_l\frac{\partial}{\partial\xi_j}e^{-\beta U}d\vec{\xi}\right].$$

Using spherical coordinates, these two integrals can be readily performed if the angular part is carried out before the radial part and noting that

$$\int_0^\infty \frac{d}{d\xi}e^{-\beta U}d\xi=1 .$$

The final result is simply (12).

APPENDIX B: EVALUATION OF π_0

The left-hand side of (3) is

$$\vec{\nabla}\left[\frac{\rho}{\beta}\right]+\int\frac{\partial\Phi^{(s)}}{\partial\vec{r}}\rho_0^{(2)}d\vec{r}\,' \qquad \text{(B1)}$$

in the static case. Since $\partial\Phi^{(s)}/\partial\vec{r}$ is short ranged, $\rho_0^{(2)}$ is mainly determined by the bare interparticle potential, i.e.,

$$\rho_0^{(2)}=\rho(\vec{r})\rho(\vec{r}\,')\exp[-\beta\Phi^{(s)}(\,|\,\vec{r}-\vec{r}\,'\,|\,)] .$$

The integral in (B1) thus becomes

$$\frac{1}{\beta}\rho(\vec{r})\int\rho(\vec{r}\,')\frac{\partial}{\partial\vec{r}}[1-\exp(-\beta\Phi^{(s)})]d\vec{r}\,' . \qquad \text{(B2)}$$

Changing $\partial/\partial\vec{r}$ to $-\partial/\partial\vec{r}\,'$, and after an integration by parts, (B2) reduces to

$$\frac{1}{\beta}\rho(\vec{r})\int[1-\exp(-\beta\Phi^{(s)})]\frac{\partial}{\partial\vec{r}\,'}\rho(\vec{r}\,')d\vec{r}\,' .$$

Since the expansion of $\vec{P}(\vec{r}\,')$ in (A2) applies similarly to $\rho(\vec{r}\,')$, we get

$$kT\rho(\vec{r})\frac{\partial}{\partial\vec{r}}\rho(\vec{r})\int\{1-\exp[-\beta\Phi^{(s)}(\xi)]\}d\vec{\xi} ,$$

where change of $d\vec{r}\,'$ to $d\vec{\xi}$ has been made. Equation (B1) can be finally written as

$$\vec{\nabla}\left[\rho kT+\frac{\rho^2 kT}{2}\int[1-\exp(-\beta\Phi^{(s)})]d\vec{\xi}\right] .$$

The quantity is readily recognized as the pressure in the absence of electric fields including the contribution of the second virial coefficient.

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