PHYSICAL REVIEW A VOLUME 16, NUMBER 3 SEPTEMBER 1977

Electromagnetic wave propagation in an inhomogeneous medium: A perturbative approach

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(Received 12 November 1976)

This paper is concerned with the scattering of electromagnetic radiation in an inhomogeneous polarizable medium. In particular, we are interested in scatterers whose positions are correlated and whose characteristic size is on the order of or less than a wavelength. The central problem is that of determining the electric and magnetic fields acting on a given scatterer in the medium. This is the effective wave that polarizes the scatterer, and its calculation is complicated by what are generally referred to as local field effects. Assuming that the scattering is weak, i.e., that the system deviates only slightly from being homogeneous, we are able to treat local field effects through second order in perturbation theory, and to derive explicit expressions for the scattered effective field and the dispersion relation satisfied by the coherent wave. The scattering cross section is derived and related to the attenuation of the coherent wave in an infinite medium. In addition, the diffractive effects due to scattering from a finite medium are discussed.

I. INTRODUCTION

Conventional treatments of wave propagation in dielectric media are based on self-consistent integral equations[1-3] that relate the effective field at a given point to the scattered waves emitted at all other points in the sample. This analysis is usually applied to molecular systems with fields whose wavelengths are in the optical regime. In this case the distances characterizing the inhomogeneities (molecular dimensions) are much smaller than a typical wavelength and the polarizability may therefore be assumed to be constant. By contrast, in the present case the size of the inhomogeneities is comparable to the wavelength and a more careful treatment is required. In particular, the polarizability must now be allowed to vary continuously.

Two complementary approaches have been used in connection with the present problem. The first approach is based on multiple scattering theory[4-6] and involves the development of a hierarchy of integral equations. The first equation expresses the total field in terms of the effective field with one scatterer held fixed, the second relates the effective field with one scatterer held fixed to the effective field with two scatterers held fixed, etc. In practice, this hierarchy must be truncated at an early level, for example, by assuming that the effective fields with one and two sites held fixed are equal.[5] While approximations of this kind are physically reasonable, their precise range of validity is difficult to estimate. An alternative procedure has been developed by Vezzetti and Keller.[6] These authors solve for the natural modes of an infinite medium to obtain a dispersion relation as a power series in the polarizability. However, an explicit relation for the index of refraction is found by considering only the first-order term and assuming that the scatterers are uncorrelated.

The second method, which is employed in this paper, is based on perturbation theory and is certainly valid as long as the fluctuations in the random polarizability are small. This approach has been considered in some detail by Keller and coworkers.[7-9] While the basic formalism developed in Sec. III of the present paper is closely related to that of Ref. 8, our approach differs from that of previous authors in several respects. First, these papers[7-9] start from the wave equation, in which the macroscopic parameters are allowed to vary. Our integral equation for the effective field is microscopic, with the random variable being the polarizability at a site. This procedure is conceptually superior because (i) it follows the procedure used in a uniform medium to relate the macroscopic dielectric constant to the polarizability (which is reviewed in Sec. II), and (ii) it more obviously treats the inhomogeneities as perturbations from uniformity. Essentially, the previous authors allow the dielectric constant to vary. They then derive an expression for the change in the macroscopic dielectric constant in terms of the correlation of these fluctuations. However, the dielectric constant, as well as its fluctuations, must fundamentally involve a summation over dipole sites. This is the approach taken in the present paper; we deal directly with the microscopic properties of an inhomogeneous medium. Second, we treat the self-field singularities by noting that each integral over the dipole sites must exclude the small volume occupied by the dipole at the point where the effective field is being calculated. Since each of these integrals involves the differential operator ($\nabla\times\nabla\times$) acting on a Green's function, whenever the derivatives are removed from an integral a self-field term is produced. Of course, one removes the derivatives so that Fourier transforming reduces the integral equation to an algebraic equation. However, the fundamental expression for the effective field involves a summation of fields ($\nabla\times\nabla\times$ inside the integral) which differs from the field of a summation of polarization potentials ($\nabla\times\nabla\times$ outside) by the self-field term. The exact treatment of these terms changes the corrections to the Lorentz-Lorenz relation from those obtained by previous authors.[10]

II. EFFECTIVE FIELD EQUATIONS

We consider the propagation of an electromagnetic wave in an infinite, inhomogeneous nonmagnetic medium. The effective electric and magnetic fields $\vec{E}'(\vec{r}_n, t)$ and $\vec{H}'(\vec{r}_n, t)$, defined as the fields acting on the nth dipole of the medium can be divided into the incident (external) fields $\vec{E}_0$, $\vec{H}_0$, and a contribution due to scattered waves emitted by all of the other dipoles,

$$\vec{E}'(\vec{r}_n, t) = \vec{E}_0(\vec{r}_n, t) + \sum_{m\neq n} \vec{E}_m(\vec{r}_n, t)\,, \tag{2.1a}$$

$$\vec{H}'(\vec{r}_n, t) = \vec{H}_0(\vec{r}_n, t) + \sum_{m\neq n} \vec{H}_m(\vec{r}_n, t)\,. \tag{2.1b}$$

Here $\vec{E}_m(\vec{r}_n, t)$ and $\vec{H}_m(\vec{r}_n, t)$ are the values at the point $\vec{r}_n$ and the time t of the fields radiated by the mth dipole. In terms of the moment $\vec{p}(\vec{r}_m, t)$, these fields are given by[11]

$$\vec{E}_m(\vec{r}_n, t) = \nabla\times\nabla\times[\vec{p}(\vec{r}_m, t - R_{nm}/c)/R_{nm}]\,, \tag{2.2a}$$

$$\vec{H}_m(\vec{r}_n, t) = \nabla\times[\dot{\vec{p}}(\vec{r}_m, t - R_{nm}/c)/R_{nm}]\,, \tag{2.2b}$$

where $R_{nm} = |\vec{r}_n - \vec{r}_m|$, the $\nabla\times$ operation is taken with respect to the coordinates x_n, y_n, z_n of the nth dipole, and the dot in Eq. (2.2b) denotes a time derivative. We will assume that the medium is a continuous distribution of dipoles. Neglecting effects due to magnetic forces, the total dipole moment per unit volume $\vec{P}(\vec{r}, t)$ and the effective field $\vec{E}'(\vec{r}, t)$ are then related by the equation

$$\vec{P}(\vec{r}, t) = \alpha(\vec{r}, t)\vec{E}'(\vec{r}, t)\,. \tag{2.3}$$

Note that the polarizability $\alpha(\vec{r}, t)$ as defined by Eq. (2.3) contains a factor of the dipole number density, and therefore is dimensionless. We allow for nonrigid systems by letting $\alpha(\vec{r}, t)$ be time dependent. Combining Eqs. (2.1)–(2.3), and replacing the sums in (2.1) by volume integrations, we obtain the closed system of integral equations

$$\vec{E}'(\vec{r}, t) = \vec{E}_0(\vec{r}, t) + \int_\sigma \nabla_r\times\nabla_r\times \frac{\alpha(\vec{r}_1, t - R/c)\vec{E}'(\vec{r}_1, t - R/c)}{R}\, d^3r_1\,, \tag{2.4a}$$

$$\vec{H}'(\vec{r}, t) = \vec{H}_0(\vec{r}, t) + \int_\sigma \nabla_r\times \frac{(\partial/\partial t)[\alpha(\vec{r}_1, t - R/c)\vec{E}'(\vec{r}_1, t - R/c)]}{R}\, d^3r_1\,, \tag{2.4b}$$

where $R = |\vec{r} - \vec{r}_1|$ and the subscript σ on the r_1 integration indicates that an infinitesimal sphere of radius ϵ centered at the point $\vec{r}_1 = \vec{r}$ (representing the volume occupied by the dipole at $\vec{r}$) has been excluded from the volume of integration. The limit $\epsilon \to 0$ is taken at the end of every calculation.

Equation (2.4a) is an integro-differential equation for the effective field $\vec{E}'(\vec{r}, t)$. Since the medium is nonmagnetic, only one equation is required, that is, given an approximate solution for $\vec{E}'(\vec{r}, t)$, the corresponding approximation for $\vec{H}'(\vec{r}, t)$ may be obtained directly for Eq. (2.4b). We emphasize that these equations are equivalent to Maxwell's equations for the present system.[11] Equations (2.4) simplify if the incident and effective fields are assumed to vary monochromatically in time. In this case (2.4a) may be rewritten

$$\vec{E}'(\vec{r}) = \vec{E}_0(\vec{r}) + \int_\sigma \nabla_r\times\nabla_r\times[G_0(\vec{r} - \vec{r}_1)\alpha(\vec{r}_1)\vec{E}'(\vec{r}_1)]\, d^3r_1\,, \tag{2.5a}$$

where

$$G_0(\vec{r} - \vec{r}) = \exp(ik_0|\vec{r} - \vec{r}_1|)/|\vec{r} - \vec{r}_1| \tag{2.5b}$$

is the usual vacuum Green's function ($k_0 = \omega/c$). (We use a subscript zero to signify quantities characteristic of the vacuum.) In principle, the effective field remains time dependent through the random variable $\alpha(\vec{r}, t - R/c)$. However, for nonrelativistic systems the time R/c is very much smaller than the interval over which the polarizability will vary. Thus we can assume that $\alpha(\vec{r}, t) \simeq \alpha(\vec{r}, t - R/c)$. Though the medium may change in time, we will calculate the effective field at the time t as if the medium froze at the configuration assumed at t. Accordingly the explicit dependence on t in Eq. (2.5a) has been dropped.

Equation (2.5a) is the central equation of the present section. Before developing the formalism required to treat the general inhomogeneous system, we consider the simpler case of a uniform medium, i.e., $\alpha(\vec{r}) \to \bar{\alpha}$ (a constant). (We use a bar to signify quantities characteristic of the average medium.) This will allow us to derive the familiar Lorentz-Lorenz equation. To begin, we employ the identity[12]

$$\int_\sigma \nabla_r\times\nabla_r\times G_0(\vec r-\vec r_1)\vec f(\vec r_1)\,d^3r_1$$
$$=-\tfrac{8}{3}\pi\vec f(\vec r)+\nabla_r\times\nabla_r\times\int_\sigma G_0(\vec r-\vec r_1)\vec f(\vec r_1)\,d^3r_1 \quad (2.6)$$

to write Eq. (2.5a) as

$$\vec{\bar{\tilde E}}'(\vec r)=\vec{\tilde E}_0(\vec r)+\nabla_r\times\nabla_r\times\int_\sigma G_0(\vec r-\vec r_1)\tilde\alpha\vec{\bar{\tilde E}}'(\vec r_1)\,d^3r_1\,, \quad (2.7)$$

where

$$\vec{\tilde E}_0(\vec r)=\vec E_0(\vec r)/(1+\tfrac{8}{3}\pi\bar\alpha)\,, \quad (2.8a)$$

and

$$\tilde\alpha=\bar\alpha/(1+\tfrac{8}{3}\pi\bar\alpha)\,. \quad (2.8b)$$

Introducing the Fourier transforms

$$\vec{\bar{\tilde E}}'(\vec q)=\int d^3r\,e^{-i\vec q\cdot\vec r}\vec{\bar{\tilde E}}'(\vec r)\,, \quad (2.9a)$$

$$\vec{\tilde E}_0(\vec q)=\int d^3r\,e^{-i\vec q\cdot\vec r}\vec{\tilde E}_0(\vec r)\,, \quad (2.9b)$$

and

$$G_0(q)=\int d^3r\,e^{-i\vec q\cdot\vec r}G_0(r)=4\pi/(q^2-k_0^2)\,, \quad (2.9c)$$

Eq. (2.7) reduces to the linear algebraic equation[13]

$$\vec{\bar{\tilde E}}'(\vec q)=\vec{\tilde E}_0(\vec q)+[4\pi\tilde\alpha/(k_0^2-q^2)]\vec q\times\vec q\times\vec{\bar{\tilde E}}'(\vec q)\,, \quad (2.10)$$

which can be solved by iterating and then summing the resulting geometric series. After some straightforward manipulation the result may be written

$$\vec{\bar{\tilde E}}'(\vec q)=\vec{\tilde E}_0(\vec q)+\left(\frac{4\pi\tilde\alpha}{1-4\pi\tilde\alpha}\right)\frac{\vec q\times\vec q\times\vec{\tilde E}_0(\vec q)}{\bar k^2-q^2} \quad (2.11a)$$

or, more simply, as

$$(\bar k^2-q^2)\vec{\bar E}'(\vec q)=[(k_0^2-q^2)/(1-\tfrac{4}{3}\pi\bar\alpha)]\vec E_0(\vec q)\,, \quad (2.11b)$$

where

$$\bar k^2=\frac{k_0^2}{1-4\pi\tilde\alpha}\equiv\bar\epsilon k_0^2\,. \quad (2.12)$$

(Note that $\overline{k^2}=\bar k^2$.) In Eq. (2.11b) we have used the fact that, since $\vec E_0$ propagates in the vacuum, $\vec q\times\vec q\times\vec E_0=-q^2\vec E_0$.

Equations (2.11) and (2.12) represent the solution of the effective-field equation (2.5a) in the case of a uniform medium. Physically, the principal effect of the medium is to change the velocity of propagation of the wave from c to $c/\bar\epsilon^{1/2}$. While this point is perhaps seen most clearly in connection with the extinction theorem,[11] it is already apparent in Eqs. (2.11b) and (2.12) where the vacuum dispersion relation $q^2=k_0^2$ has been changed to $q^2=\bar k^2=\bar\epsilon k_0^2$. Equation (2.12), relating $\bar\epsilon$ and $\bar\alpha$, is clearly equivalent to

$$\bar\epsilon=(1+\tfrac{8}{3}\pi\bar\alpha)/(1-\tfrac{4}{3}\pi\bar\alpha) \quad (2.13a)$$

or

$$\tfrac{4}{3}\pi\bar\alpha=(\bar\epsilon-1)/(\bar\epsilon+2)\,, \quad (2.13b)$$

the usual Lorentz-Lorenz equation. Returning to the coordinate representation, Eq. (2.11) leads to the following expression for the effective field

$$\vec{\bar{\tilde E}}'(\vec r)=\vec{\tilde E}_0(\vec r)+\frac{\bar\epsilon-1}{4\pi}\nabla_r\times\nabla_r\times\int_\sigma\bar G(\vec r-\vec r_1)\vec{\tilde E}_0(\vec r_1)\,d^3r_1\,, \quad (2.14a)$$

where

$$\bar G(\vec r-\vec r_1)=\exp(i\bar k|\vec r-\vec r_1|)/|\vec r-\vec r_1| \quad (2.14b)$$

is the Green's function corresponding to the uniform medium.

III. SPATIALLY VARYING POLARIZABILITY

In this section we develop techniques for the approximate solution of the general effective-field equation (2.5a). Our basic assumption is that the system differs only slightly from a homogeneous medium. Accordingly, the normalized polarizability will be written as

$$\alpha(\vec r)=\bar\alpha+\eta\alpha_1(\vec r)\,, \quad (3.1)$$

when η is a dimensionless parameter that characterizes the strength of the medium's departure from homogeneity. Our aim is to develop a perturbative expression for the effective field. Due to the random variable $\alpha(\vec r)$, the effective field is a statistical quantity and is thus an exceedingly complicated function that requires the detailed knowledge of $\alpha(\vec r)$. However, we may assume that the macroscopic behavior of the effective field (e.g., the dispersion relation it obeys) is shared by the coherent wave, the average of the effective field. If the scattering medium is in equilibrium, the average may be performed either over an ensemble of configurations frozen at the instant t or over a period of time long compared to the interval over which α varies.

To begin, let us write the basic equation (2.5a) in symbolic form

$$\vec E'=\vec E_0+L^0(\alpha)\vec E'\,. \quad (3.2)$$

Each term in this equation is understood to be a function of $\vec r$. The operator $L^0(\alpha)$ acting on an arbitrary function $\vec f(\vec r)$ is defined as

$$L^0(\alpha)\vec{f} = \int_\sigma \nabla\times\nabla\times G_0(\vec{r}-\vec{r}_1)\alpha(\vec{r}_1)\vec{f}(\vec{r}_1)\,d^3r_1 . \qquad (3.3)$$

In the limit $\alpha \to \overline{\alpha}$ (i.e., $\eta \to 0$), the operator $L^0(\alpha) \to L^0(\overline{\alpha})$ and we regain the uniform effective field $\overline{\vec{E}}'$:

$$\overline{\vec{E}}' = \vec{E}_0 + L^0(\overline{\alpha})\overline{\vec{E}}' . \qquad (3.4)$$

In general, let us define

$$\vec{E}' = \overline{\vec{E}}' + \Delta\vec{E}' \qquad (3.5a)$$

$$\equiv \overline{\vec{E}}' + \eta\vec{E}_1' + \eta^2\vec{E}_2' + \cdots \qquad (3.5b)$$

and derive an equation for the quantity $\Delta\vec{E}'$ that describes the change in the effective wave due to the inhomogeneities. Using Eqs. (3.5a) and (3.1) in the right-hand side of (3.2), we find

$$\vec{E}' = \vec{E}_0 + L^0(\overline{\alpha})\overline{\vec{E}}' + L^0(\eta\alpha_1)\overline{\vec{E}}' + L^0(\overline{\alpha})\Delta\vec{E}' + L^0(\eta\alpha_1)\Delta\vec{E}' . \qquad (3.6)$$

Introducing the abbreviated notation

$$L_0^0 \equiv L^0(\overline{\alpha}) , \qquad (3.7a)$$

$$\eta L_1^0 \equiv L^0(\eta\alpha_1) , \qquad (3.7b)$$

and recalling Eqs. (3.4) and (3.5a), we can obtain an expression for $\Delta\vec{E}'$ from Eq. (3.6):

$$\Delta\vec{E}' = \eta L_1^0\overline{\vec{E}}' + L_0^0\Delta\vec{E}' + \eta L_1^0\Delta\vec{E}' = \eta L_1\overline{\vec{E}}' + \eta L_1\Delta\vec{E}' , \qquad (3.8)$$

where

$$L_1 \equiv (1-L_0^0)^{-1}L_1^0 . \qquad (3.9)$$

Iterating Eq. (3.8) we obtain the desired perturbation series in η

$$\vec{E}' = (1 + \eta L_1 + \eta^2 L_1L_1 + \eta^3 L_1L_1L_1 + \cdots)\overline{\vec{E}}' . \qquad (3.10)$$

Equation (3.10) is the principal equation of this section; it expresses the effective field $\vec{E}'$ in terms of successive powers of the perturbation parameter η and the operator L_1 defined by Eqs. (3.9) and (3.7). All of the dependence on the random function α_1 is contained in L_1. We may now average equation (3.10) to obtain an expression for the coherent wave:

$$\langle\vec{E}'\rangle = (1 + \eta\langle L_1\rangle + \eta^2\langle L_1L_1\rangle + \cdots)\overline{\vec{E}}' . \qquad (3.11)$$

By analogy with the uniform medium, Eq. (3.4), let us define an operator L such that

$$(1-L)\langle\vec{E}'\rangle = \overline{\vec{E}}' . \qquad (3.12)$$

In view of (3.11), L must satisfy the equation

$$1 - L = (1 + \eta\langle L_1\rangle + \eta^2\langle L_1L_1\rangle + \cdots)^{-1} . \qquad (3.13)$$

Expanding the right-hand side of this equation and keeping terms through second order in η we find

$$L = \eta\langle L_1\rangle + \eta^2(\langle L_1L_1\rangle - \langle L_1\rangle\langle L_1\rangle) + O(\eta^3) . \qquad (3.14)$$

For convenience, let us assume that the constant $\overline{\alpha}$ in Eq. (3.1) has been chosen such that $\langle\alpha\rangle = \overline{\alpha}$, i.e., such that $\langle\alpha_1\rangle = 0$. In this case $\langle L_1\rangle$ vanishes and Eqs. (3.12) and (3.14) reduce to

$$(1 - \eta^2\langle L_1L_1\rangle)\langle\vec{E}'\rangle = \overline{\vec{E}}' . \qquad (3.15)$$

In Sec. IV this result will be used to derive a dispersion relation correct to second order.

IV. EFFECTIVE FIELDS AND THE DISPERSION RELATION OF THE COHERENT FIELD

We shall now apply the formalism developed in Sec. III to write expressions for the effective wave in the first and second orders of perturbation theory. It should be emphasized that, at the present stage, the results of Sec. III are entirely formal in the sense that we have not yet derived an explicit representation of the operator L_1. This point is discussed in Appendix A where the definition (3.9) is related to the solution of a linear integral equation. Within this framework, the analysis is quite similar to that used to solve for the uniform effective field $\overline{\vec{E}}'$ [i.e., to derive Eqs. (2.11) and (2.14)]. From Eqs. (A11), in the coordinate representation, the vector obtained by allowing L_1 to act on an arbitrary vector field $\vec{F}(\vec{r})$ is found to be

$$L_1\vec{F} = c_1\alpha_1(\vec{r})\vec{F}(\vec{r}) + c_2\nabla_r\times\nabla_r\times\int_\sigma \overline{G}(\vec{r}-\vec{r}_1)\,\alpha_1(\vec{r}_1)\vec{F}(\vec{r}_1)\,d^3r_1 , \qquad (4.1)$$

where c_1 and c_2 are the constants

$$c_1 = -\tfrac{8}{3}\pi(1+\tfrac{8}{3}\pi\overline{\alpha})^{-1} , \qquad (4.2a)$$

$$c_2 = (1+\tfrac{8}{3}\pi\overline{\alpha})^{-1}(1-\tfrac{4}{3}\pi\overline{\alpha})^{-1} . \qquad (4.2b)$$

Note that, once again, the vacuum Green's function $G_0(r)$, which appeared in the definition of the original operators L_0^0 and L_1^0 [Eqs. (3.7) and (3.3)], has been replaced by the medium Green's function $\overline{G}(r)$ defined by Eq. (2.14b).

Using Eqs. (4.1) in (3.10), the first- and second-order corrections to the effective wave are given by

$$\vec{E}_1'(\vec{r}) \equiv L_1\overline{\vec{E}}' = c_1\alpha_1(\vec{r})\overline{\vec{E}}'(\vec{r}) + c_2\nabla_r\times\nabla_r\times\int_\sigma d^3r_1\overline{G}(\vec{r}-\vec{r}_1)\alpha_1(\vec{r}_1)\overline{\vec{E}}'(\vec{r}_1) , \qquad (4.3a)$$

$$\vec{E}'_2(\vec{r}) \equiv (L_1)^2\overline{\vec{E}}' = c_1\alpha_1(\vec{r})\vec{E}'_1(\vec{r}) + c_2\nabla_r\times\nabla_r\times\int_\sigma d^3r_1\overline{G}(\vec{r}-\vec{r}_1)\alpha_1(\vec{r}_1)\vec{E}'_1(\vec{r}_1)$$

$$= c_1^2\alpha_1^2(\vec{r})\overline{\vec{E}}'(\vec{r}) + c_1c_2\left(\alpha_1(\vec{r})\nabla_r\times\nabla_r\times\int_\sigma d^3r_1\overline{G}(\vec{r}-\vec{r}_1)\alpha_1(\vec{r}_1)\overline{\vec{E}}'(\vec{r}_1) + \nabla_r\times\nabla_r\times\int_\sigma d^3r_1\overline{G}(\vec{r}-\vec{r}_1)\alpha_1^2(\vec{r}_1)\overline{\vec{E}}'(\vec{r}_1)\right)$$

$$+ c_2^2\nabla_r\times\nabla_r\times\int_\sigma d^3r_1\overline{G}(\vec{r}-\vec{r}_1)\alpha_1(\vec{r}_1)\nabla_{r_1}\times\nabla_{r_1}\times\int_\sigma d^3r_2\overline{G}(\vec{r}_1-\vec{r}_2)\alpha_1(\vec{r}_2)\overline{\vec{E}}'(\vec{r}_2). \tag{4.3b}$$

Of course, with an explicit representation for the operator L_1 one may solve for the effective field to any order, where successive corrections would involve higher moments of α_1. Note that the resulting set of equations is more useful than the hierarchy of equations developed from the multiple scattering viewpoint.[4,5] These are explicit in the sense that the known field $\overline{\vec{E}}'$ appears on the right-hand side of Eqs. (4.3) and one can readily estimate the range of validity of any truncated series.

Equation (4.3a) will be used in Sec. V to derive an explicit equation for the angular dependence of the scattered radiation correct to second order. The remainder of this section is concerned with the derivation of the dispersion relation for the coherent wave $\langle\vec{E}'\rangle$. To derive the dispersion relation from Eq. (3.15) the action of the operator $\langle(L_1)^2\rangle$ on the coherent wave must be examined. Combining the explicit representation of $(L_1)^2$ given in equation (4.3b) with the identity (2.6), we obtain

$$(L_1)^2\langle\vec{E}'\rangle = (c_1^2+\tfrac{8}{3}\pi c_1c_2)\alpha_1^2(\vec{r})\langle\vec{E}'(\vec{r})\rangle + c_1c_2\int_\sigma d^3r_1\alpha_1(\vec{r})\alpha_1(\vec{r}_1)\nabla_r\times\nabla_r\times\overline{G}(\vec{r}-\vec{r}_1)\langle\vec{E}'(\vec{r}_1)\rangle$$

$$+ (c_1c_2+\tfrac{8}{3}\pi c_2^2)\,\nabla_r\times\nabla_r\times\int_\sigma d^3r_1\alpha_1^2(\vec{r}_1)\overline{G}(\vec{r}-\vec{r}_1)\langle\vec{E}'(\vec{r}_1)\rangle$$

$$+ c_2^2\nabla_r\times\nabla_r\times\int_\sigma d^3r_1\overline{G}(\vec{r}-\vec{r}_1)\int_\sigma d^3r_2\alpha_1(\vec{r}_1)\alpha_1(\vec{r}_2)\nabla_{r_1}\times\nabla_{r_1}\times\overline{G}(\vec{r}_1-\vec{r}_2)\langle\vec{E}'(\vec{r}_2)\rangle. \tag{4.4}$$

To average the right-hand side of Eq. (4.4) over the various configurations of the random medium we introduce the two-site correlation function

$$S(|\vec{r}_1-\vec{r}_2|) = \langle\alpha_1(\vec{r}_1)\alpha_1(\vec{r}_2)\rangle. \tag{4.5}$$

$S(|\vec{r}_1-\vec{r}_2|)$ has been written as a function of the argument $|\vec{r}_1-\vec{r}_2|$ to indicate that on the average the system is both translationally invariant and isotropic. The configuration average of Eq. (4.4) is then

$$\langle(L_1)^2\rangle\langle\vec{E}'\rangle$$

$$= (c_1^2+\tfrac{8}{3}\pi c_1c_2)S(0)\langle\vec{E}'(\vec{r})\rangle + c_1c_2\vec{K}(\vec{r})$$

$$+ (c_1c_2+\tfrac{8}{3}\pi c_2^2)S(0)\nabla_r\times\nabla_r\times\int_\sigma d^3r_1\overline{G}(\vec{r}-\vec{r}_1)\langle\vec{E}'(\vec{r}_1)\rangle$$

$$+ c_2^2\nabla_r\times\nabla_r\times\int_\sigma d^3r_1\overline{G}(\vec{r}-\vec{r}_1)\vec{K}(\vec{r}_1), \tag{4.6a}$$

where

$$\vec{K}(\vec{r}) = \int_\sigma d^3r_1\, S(|\vec{r}-\vec{r}_1|)\nabla_r\times\nabla_r\times\overline{G}(\vec{r}-\vec{r}_1)\langle\vec{E}'(\vec{r}_1)\rangle. \tag{4.6b}$$

Note that $\langle\vec{E}'(\vec{r}_1)\rangle$ is also translationally invariant and therefore does not enter the averaging process. The vector field $\vec{K}(\vec{r})$ is analyzed in detail in Appendix B. Assuming that the coherent effective field $\langle\vec{E}'(\vec{r})\rangle$ may be represented as a linearly polarized plane wave, $\vec{K}(\vec{r})$ can be reduced to

$$\vec{K}(\vec{r}) = \langle\vec{E}'(\vec{r})\rangle K(\bar{k})$$

$$\equiv \langle\vec{E}'(\vec{r})\rangle 4\pi\bar{k}\int_0^\infty d\rho\, S(\rho)e^{i\bar{k}\rho}F(\bar{k}\rho), \tag{4.7a}$$

where

$$F(x) = [(1-2x^{-2}+3x^{-4})\sin(x) + (x^{-1}-3x^{-3})\cos(x)]$$
$$+ i[(x^{-1}-3x^{-3})\sin(x) + 3x^{-2}\cos(x)]. \tag{4.7b}$$

It should be emphasized that the transverse character of $\vec{K}(\vec{r})$ (and hence of $\langle\vec{E}'(\vec{r})\rangle$) is a consequence not only of a scalar polarizability $\alpha(\vec{r})$, but also of an isotropic correlation function $S(r)$.

We now use equations (4.7a) and (4.6a) in (3.15), and the resulting expression is Fourier transformed to the q representation to obtain

$$[1-\eta^2\langle(L_1)^2\rangle]\langle\vec{E}'\rangle = \left(1-\eta^2[(c_1^2+\tfrac{8}{3}\pi c_1c_2)S(0)+c_1c_2K(\bar{k})] - 4\pi\eta^2[(c_1c_2+\tfrac{8}{3}\pi c_2^2)S(0)+c_2^2K(\bar{k})]\frac{\vec{q}\times\vec{q}\times}{\bar{k}^2-q^2}\right)\langle\vec{E}'(\vec{q})\rangle = \overline{\vec{E}}'(\vec{q}). \tag{4.8}$$

Multiplying by $(q^2-\bar{k}^2)$ and retaining terms of order η^2, we obtain[14]

$$\{q^2 - \bar{k}^2 - 4\pi\eta^2\bar{k}^2[(c_1c_2 + \tfrac{8}{3}\pi c_2^2)S(0) + c_2^2K(\bar{k})]\}\langle\vec{\mathrm{E}}'(\vec{\mathrm{q}})\rangle = (q^2 - \bar{k}^2)\bar{\vec{\mathrm{E}}}'(\vec{\mathrm{q}}). \tag{4.9}$$

Since $\bar{\vec{\mathrm{E}}}$ propagates with wave number $\bar{k}$, the right-hand side of Eq. (4.9) is zero for all values of q. Therefore, the dispersion relation for the coherent wave to η^2 is

$$q^2 = \bar{k}^2[1 + 4\pi\eta^2\{(c_1c_2 + \tfrac{8}{3}\pi c_2^2)S(0) + c_2^2K(\bar{k})\}]. \tag{4.10}$$

Equation (4.10) expresses the wave number q of the coherent effective wave in terms of three parameters; the incident wavelength λ, the mean polarizability $\bar{\alpha}$ or the mean dielectric constant $\bar{\epsilon}$, and $S(0) = \langle\alpha_1^2\rangle$. In addition, q depends upon the shape of the correlation function $S(\rho) = \langle\alpha_1(\vec{\mathrm{r}})\alpha_1(\vec{\mathrm{r}} + \vec{\rho})\rangle$ through the function $K(k)$. The term proportional to $S(0)$ in (4.10) is real and independent of λ. Physically, this contribution leads to a uniform slowing down of the coherent wave brought about by the mean square fluctuations in $\alpha_1(\vec{\mathrm{r}})$. By contrast, the term proportional to $K(\bar{k})$ has both real and imaginary parts and is clearly λ dependent. From Eqs. (4.10) and (4.7a) we can write an expression for the absorption coefficient of the coherent wave due to scattering:

$$2\,\mathrm{Im}(q) = \eta^2(4\pi c_2)^2\bar{k}^2 \int_0^\infty d\rho\, S(\rho)\,\mathrm{Im}[e^{i\bar{k}\rho}F(\bar{k}\rho)], \tag{4.11}$$

where $\mathrm{Im}(x)$ means the imaginary part of x.

V. SCATTERED INTENSITY

Consider an infinite medium in which the mean polarizability is equal to $\bar{\alpha}$. Then in the far-field region Eqs. (3.5b) and (4.3a) give a first-order expression for the effective wave:

$$\vec{\mathrm{E}}'(\hat{m}X) = [1 + \eta c_1\alpha_1(\hat{m}X)]\bar{\vec{\mathrm{E}}}'(\hat{m}X) + \eta c_2\nabla_X\times\nabla_X\times\int_V d^3r_1\bar{G}(\hat{m}X - \vec{\mathrm{r}}_1)\alpha_1(\vec{\mathrm{r}}_1)\bar{\vec{\mathrm{E}}}'(\vec{\mathrm{r}}_1), \tag{5.1}$$

where $\vec{\mathrm{r}} \to \hat{m}X$ ($\hat{m}$ being a unit vector). Here we have recognized that the integral extends over the finite volume V in which both $\bar{\vec{\mathrm{E}}}'$ and α_1 are nonzero (i.e., the volume containing the coherently illuminated inhomogeneities). Making the far-field approximation

$$\bar{G}(\hat{m}X - \vec{\mathrm{r}}_1) \simeq (e^{i\bar{k}X}/X)e^{-i\bar{k}\vec{\mathrm{r}}_1\cdot\hat{m}} \tag{5.2a}$$

and assuming that $\bar{E}'$ is linearly polarized,

$$\bar{\vec{\mathrm{E}}}'(\vec{\mathrm{r}}_1) = \bar{E}'\hat{e}e^{i\bar{\vec{k}}\cdot\vec{\mathrm{r}}_1}, \tag{5.2b}$$

the portion of the effective field propagating in the $\hat{m}$ direction is

$$\vec{\mathrm{E}}_s(\hat{m}X) = \vec{\mathrm{E}}'(\hat{m}X) - [1 + \eta c_1\alpha_1(\hat{m}X)]\bar{\vec{\mathrm{E}}}'(\hat{m}X) = \eta c_2\bar{E}'\bar{k}^2\frac{e^{i\bar{k}X}}{X}\,\hat{m}\times\hat{e}\times\hat{m}\int_V d^3r_1\,\alpha_1(\vec{\mathrm{r}}_1)e^{i\vec{\mathrm{r}}_1\cdot(\bar{\vec{\mathrm{k}}} - \hat{m}\bar{k})}. \tag{5.3}$$

Far from the volume V, $\vec{\mathrm{E}}_s$ represents the scattered field. In most experimental situations V is a cylinder aligned with $\bar{\vec{\mathrm{k}}}$ direction (taken to be the z axis). Defining the scattering vector $\vec{\mathrm{s}}$ equal to the wave-vector difference

$$\vec{\mathrm{s}} = \bar{\vec{\mathrm{k}}} - \bar{k}\hat{m}, \tag{5.4}$$

the integral in Eq. (5.3) may be expanded to rewrite the scattered field

$$\vec{\mathrm{E}}_s(\hat{m}X) = \eta c_2\bar{E}'\bar{k}^2\frac{e^{i\bar{k}X}}{X}\,\hat{m}\times\hat{e}\times\hat{m}\int_{-b}^{b}dx\,e^{ixs_x}\int_{-(b^2-x^2)^{1/2}}^{(b^2-x^2)^{1/2}}dy\,e^{iys_y}\int_{-l/2}^{l/2}dz\,\alpha_1(xyz)e^{izs_z} \tag{5.5a}$$

where

$$s_x = -\bar{k}\sin\theta\cos\phi,\quad s_y = -\bar{k}\sin\theta\sin\phi,\ \text{and}\ s_z = \bar{k}(1 - \cos\theta). \tag{5.5b}$$

The angles θ and ϕ are the polar and azimuthal angles of $\hat{m}$, b is the cylinder radius, and l is its length.

Equation (5.5a) can be solved easily if

$$\int_{-l/2}^{l/2}dz\,\alpha_1(xyz)e^{izs_z} \equiv \alpha_1(x,y;s_z) \to \alpha_1(s_z), \tag{5.6}$$

where $\alpha_1(s_z)$ is independent of x and y. This approximation is valid in the limit $l\to\infty$, where all possible random arrangements are represented in the z integration. Using Eqs. (5.6) in (5.5a), and orienting the scattering plane so that $\phi = \frac{1}{2}\pi$

$$\vec{\mathrm{E}}_s(\hat{m}X) = \eta c_2\bar{E}'\bar{k}^2\frac{e^{i\bar{k}X}}{X}\,\hat{m}\times\hat{e}\times\hat{m}\,\alpha_1(s_z)\,2\pi b^2\,\frac{J_1(b\bar{k}\sin\theta)}{b\bar{k}\sin\theta}, \tag{5.7}$$

where $J_1(x)$ is a Bessel function of the first kind and order. The function $\alpha_1(s_z)$ gives the angle-dependent background distribution of the scattered field, which is modulated by a rapidly oscillating function, $J_1(b\bar{k}\sin\theta)/b\bar{k}\sin\theta$ (since laser beams are difficult to focus, $b \gg \lambda/\bar{\epsilon}^{1/2}$, so for $\sin\theta > 0$, $b\bar{k}\sin\theta \gg 1$). This oscillatory function is the same as that due to Fraunhofer diffraction from a circular aperture.

In a finite random system, particularly a disperse, weakly scattering system, $\alpha_1(x, y; s_z)$ depends explicitly on x and y. This breaks the circular symmetry. Thus the diffraction pattern from a random screen is speckled and if the inhomogeneities are mobile, the pattern sparkles. In a disperse system, we may take the view that comparatively few of the differential volumes $dxdydz$ in Eq. (5.5a) are significant, and the the field amplitude at a particular observation point depends upon the interference between waves from these few. Thus we should expect a significant change in the amplitude when the scattering angle varies

$$\delta\theta = \tfrac{1}{2}\pi/r\bar{k}\cos\theta \quad \text{or} \quad \delta\theta = \tfrac{1}{2}\pi/z\bar{k}\sin\theta, \tag{5.8a}$$

where $r \lesssim b$ and $z \lesssim l$. Similarly, in nonrigid systems, we should expect a significant change in the time interval

$$\delta t = \tfrac{1}{2}\pi/v_\perp\bar{k}\sin\theta \quad \text{or} \quad \delta t = \tfrac{1}{2}\pi/v_\parallel\bar{k}(1-\cos\theta), \tag{5.8b}$$

where $v_\perp$ and $v_\parallel$ are the scatterer velocities in the scattering plane, perpendicular and parallel to $\vec{\bar{k}}$ (to this order of the far-field approximation there is no dependence on the coordinate perpendicular to the scattering plane).

Returning to Eq. (5.3), we can write an expression for the scattered Poynting vector

$$\begin{aligned}\vec{I}_s &= \eta^2 c_2^2 \bar{I}(\bar{k}^4/X^2)\hat{m}|\hat{e}\times\hat{m}|^2 \\ &\times \int_V d^3r \int_V d^3r_1\, \alpha_1(\vec{r})\alpha_1(\vec{r}_1)e^{i\vec{s}\cdot(\vec{r}-\vec{r}_1)} \\ &= \eta^2 c_2^2 \bar{I}(\bar{k}^4/X^2)\hat{m}|\hat{e}\times\hat{m}|^2 V\int_{-\infty}^{\infty} d^3\rho\, S(\rho)e^{i\vec{s}\cdot\vec{\rho}},\end{aligned} \tag{5.9a}$$

where $\bar{I}$ is the irradiance of the average wave and $\vec{\rho} = \vec{r} - \vec{r}_1$. We have also approximated the two-point correlation function

$$\begin{aligned}S(\rho) &= \langle \alpha_1(\vec{r})\alpha_1(\vec{r}+\vec{\rho})\rangle \\ &\simeq \frac{1}{V}\int_V d^3\rho\, \alpha_1(\vec{r}_1)\alpha_1(\vec{r}_1+\vec{\rho}),\end{aligned} \tag{5.9b}$$

and allowed the first integration to extend to infinity. The degree to which these approximations are inexact in a finite random medium lead to the diffractive effects discussed above. Within the approximations, one has a description of the scattered irradiance that is observed by a detector of angular width much greater than $\delta\theta$ in Eq. (5.8a) and with a time constant much longer than δt in (5.8b).

Finally, we may write an expression for the scattering cross section per unit volume in the direction $\hat{m}$:

$$\begin{aligned}\mathcal{R}(\hat{m}) &= \frac{I_s(\hat{m}X)X^2}{\bar{I}V} \\ &= \eta^2 4\pi c_2^2 \bar{k}^4|\hat{e}\times\hat{m}|^2 \int_0^\infty \rho\, d\rho\, S(\rho)\sin(s\rho)/s,\end{aligned} \tag{5.10a}$$

where

$$s = 2\bar{k}|\sin\tfrac{1}{2}\theta|, \tag{5.10b}$$

and we have used that $S(\rho)$ is isotropic. Now, letting the polar z axis coincide with the $\vec{\bar{k}}$ direction and the polarization direction coincide with the x axis, the polarization factor is

$$|\hat{e}\times\hat{m}|^2 = \cos^2\theta + \sin^2\theta\sin^2\phi, \tag{5.11}$$

where θ and ϕ are still the polar and azimuthal angles of $\hat{m}$. With Eq. (5.11) we may integrate $\mathcal{R}(\hat{m})$ over all solid angles to obtain the scattering coefficient of absorption,

$$\begin{aligned}\sigma \equiv \int \mathcal{R}(\hat{m})d\Omega = \eta^2(4\pi c_2)^2\bar{k}^2\int_0^\infty d\rho\, S(\rho)[&\sin^2\bar{k}\rho + 2(\bar{k}\rho)^{-1}\sin\bar{k}\rho\cos\bar{k}\rho - 5(\bar{k}\rho)^{-2}\sin^2\bar{k}\rho + 3(\bar{k}\rho)^{-2} \\ &- 6(\bar{k}\rho)^{-3}\sin\bar{k}\rho\cos\bar{k}\rho + 3(\bar{k}\rho)^{-4}\sin^2\bar{k}\rho].\end{aligned} \tag{5.12}$$

With Eqs. (4.7b) and (4.11), we see that

$$\sigma = 2\,\mathrm{Im}(q), \tag{5.13}$$

where q is the wave number of the coherent wave correct to η^2. Hence, making the approximations in Eq. (5.9a), which ignore the diffractive effects of a finite medium, yields a scattering cross section compatible with the attenuation of the average effective field in an infinite medium. Correspondingly, the true effective field for a particular random system at a particular time will propagate with a wave number dependent upon the nature of the medium surface.

VI. DISCUSSION

Let us summarize the results for an infinite isotropic medium characterized by the randomly varying scalar polarizability $\alpha(\vec{r}) = \bar{\alpha} + \alpha_1(\vec{r})$. The average effective field will propagate with the wave number q where to order η^2

$$\mathrm{Re}(q)=\bar{k}\Big(1+\eta^2\tfrac{64}{9}\pi^3c_2^2\bar{\alpha}S(0)+\eta^2 8\pi^2c_2^2\bar{k}\times\int_0^\infty d\rho\, S(\rho)\,\mathrm{Re}(e^{i\bar{k}\rho}F(\bar{k}\rho))\Big) \tag{6.1a}$$

$$\mathrm{Im}(q)=\tfrac{1}{2}\eta^2(4\pi c_2)^2\bar{k}^2\times\int_0^\infty d\rho\, S(\rho)\,\mathrm{Im}(e^{i\bar{k}\rho}F(\bar{k}\rho)), \tag{6.1b}$$

where

$$\mathrm{Re}(e^{i\bar{k}\rho}F(\bar{k}\rho))=[1-5(\bar{k}\rho)^{-2}+3(\bar{k}\rho)^{-4}]\sin\bar{k}\rho\cos\bar{k}\rho+[(\bar{k}\rho)^{-1}-3(\bar{k}\rho)^{-3}]\cos 2\bar{k}\rho \tag{6.1c}$$

and

$$\mathrm{Im}(e^{i\bar{k}\rho}F(\bar{k}\rho))=[1-5(\bar{k}\rho)^{-2}+3(\bar{k}\rho)^{-4}]\sin^2\bar{k}\rho+3(\bar{k}\rho)^{-2}+[(\bar{k}\rho)^{-1}-3(\bar{k}\rho)^{-3}]2\sin\bar{k}\rho\cos\bar{k}\rho\,. \tag{6.1d}$$

$\bar{k}$ is the wave number of the average medium wave,

$$\bar{k}=\bar{\epsilon}^{1/2}k_0=2\pi\bar{\epsilon}^{1/2}/\lambda\,, \tag{6.2}$$

and $S(\rho)$ is the two-point correlation function,

$$S(\rho)=\langle\alpha_1(\vec{r})\alpha_1(\vec{r}+\vec{\rho})\rangle\,. \tag{6.3}$$

This average is either over an ensemble of random systems or over time in nonrigid systems in equilibrium. Finally, c_2 is the constant

$$c_2=(1-\tfrac{4}{3}\pi\bar{\alpha})^{-1}(1+\tfrac{8}{3}\pi\bar{\alpha})^{-1}\,. \tag{6.4}$$

The scattering cross section per unit volume in the $\hat{m}$ direction, which is at the angle θ with respect to the $\bar{k}$ direction, is

$$\mathcal{R}(\hat{m})=\eta^2 4\pi c_2^2\bar{k}^4|\hat{e}\times\hat{m}|^2\int_0^\infty \rho\, d\rho\, S(\rho)\frac{\sin(s\rho)}{s}, \tag{6.5a}$$

where

$$s=2\bar{k}|\sin(\tfrac{1}{2}\theta)|\,, \tag{6.5b}$$

and $\hat{e}$ is a unit vector in the polarization direction of the average medium wave. As expected, the scattering coefficient of absorption for the coherent wave is

$$\sigma=2\,\mathrm{Im}(q)=\int\mathcal{R}\,d\Omega\,. \tag{6.6}$$

The coherent wave is attenuated by intensity scattered into finite angles. The scattered cross section in the direction $\hat{m}$ is proportional to the Fourier components of $S(\rho)$, $\mathcal{R}(\hat{m})\sim\int d\rho^3\, S(\rho)e^{i\vec{s}\cdot\vec{\rho}}$ where $\vec{s}=\vec{k}-\bar{k}\hat{m}$. But the magnitude of $\vec{s}$ is limited, $0\le s\le 2\bar{k}$. Thus only the smaller spatial frequencies of $S(\rho)$ contribute to the scattering. These correspond to the flatter portions of $S(\rho)$. If the medium inhomogeneities are characterized by the length a, then the flatter portions of the correlation function will occur when $\rho<a$ and $\rho>a$.

There are two second-order contributions to the phase velocity retardation. The first is wavelength independent and is a second-order Lorentz-Lorenz correction due to the mean squared fluctuations. It results from the self-field of $\vec{E}'_1(\sim\alpha_1 E')$ acting on the surrounding mean polarizability to induce a second-order effect at the local-field point. Hence the dependence on $\bar{\alpha}S(0)$. The wavelength-dependent contribution to the retardation results from the effective slowing down of the wave due to the increased path length involved in correlated two-point scattering.

Of primary importance is the nature of the correlation function. One may take the inverse of Eq. (6.5a), measure $\mathcal{R}(\hat{m})$, and determine $S(\rho)$ experimentally as in x-ray scattering. If the random medium is simulated, the correlation function may be calculated directly.[15] Or a functional form for $S(\rho)$ may be assumed, with an adjustable parameter proportional to the size of the inhomogeneities, to calculate the measurable quantities. Then, by experiment, one determines the physical significance of the size parameter.[15,16]

Finally, the mth term of a perturbation expansion scales with $\eta^m\langle\alpha_1^m\rangle$, where variations in α_1 may be due to density or temperature fluctuations, the presence of suspended particles, etc. In the latter case, if the concentration in volume of the particles is ϕ, then

$$\langle\alpha_1^m\rangle\simeq(\alpha_{1p})^m\phi+(\alpha_{1b})^m(1-\phi)\,, \tag{6.7}$$

where α_{1p} and α_{1b} are the polarizability deviations of a particle and the bath, respectively (recall that $\alpha=\bar{\alpha}+\alpha_1$ contains a factor of the dipole number density, and therefore is dimensionless). If $\phi\ll 1$, then

$$\eta^m\langle\alpha_1^m\rangle\simeq(\eta\alpha_{1p})^m\phi\,, \tag{6.8}$$

so successive perturbation terms are scaled downward by a factor of $\eta\alpha_{1p}$. Equations like (6.8) determine the physical range of validity of the expansion for a measurable quantity in a given type of random material.

APPENDIX A

In this Appendix we derive an explicit representation of the operator

$$L_1=(1-L_0^0)^{-1}L_1^0 \tag{A1}$$

originally defined in connection with Eqs. (3.9), (3.10), and (3.15). To begin,

$$\vec{E}(\vec{r})=\int_\sigma\nabla_r\times\nabla_r\times G_0(\vec{r}-\vec{r}_1)\eta\alpha_1(\vec{r}_1)\vec{E}(\vec{r}_1)\,d^3r_1+\int_\sigma\nabla_r\times\nabla_r\times G_0(\vec{r}-\vec{r}_1)\bar{\alpha}\,\vec{E}(\vec{r}_1)\,d^3r_1\,, \tag{A2}$$

where $\vec{\overline{\mathrm{E}}}(\vec{\mathrm{r}}_1)$ is an arbitrary vector field. Equation (A2) may be rewritten symbolically as

$$\vec{\mathrm{E}} = L_1^0\vec{\overline{\mathrm{E}}} + L_0^0\vec{\mathrm{E}} = (1 - L_0^0)^{-1}L_1^0\vec{\overline{\mathrm{E}}} = L_1\vec{\overline{\mathrm{E}}}\,, \tag{A3}$$

so that a solution of Eq. (A2) provides a closed expression for the operator L_1.

Applying the identity (2.6) to Eq. (A2), and then Fourier transforming the resulting equation to the q representation, we obtain

$$\vec{\mathrm{E}}(\vec{\mathrm{q}}) = \left(\frac{-8\pi}{3} + \frac{4\pi\vec{\mathrm{q}}\times\vec{\mathrm{q}}\times}{k_0^2 - q^2}\right)\vec{\mathrm{f}}_1(\vec{\mathrm{q}}) + \left(\frac{4\pi\bar{\alpha}\vec{\mathrm{q}}\times\vec{\mathrm{q}}\times}{k_0^2 - q^2}\right)\vec{\mathrm{E}}(\vec{\mathrm{q}})\,, \tag{A4}$$

where

$$\vec{\mathrm{f}}_1(\vec{\mathrm{q}}) = \int d^3r\,\eta\tilde{\alpha}_1(\vec{\mathrm{r}})\vec{\overline{\mathrm{E}}}(\vec{\mathrm{r}})e^{-i\vec{\mathrm{q}}\cdot\vec{\mathrm{r}}} \tag{A5a}$$

and

$$\tilde{\alpha}_1(\vec{\mathrm{r}}) = \alpha_1(\vec{\mathrm{r}})(1 + \tfrac{8}{3}\pi\bar{\alpha})^{-1}\,. \tag{A5b}$$

Introducing the longitudinal and transverse projection operators,

$$P_L = \vec{\mathrm{q}}\vec{\mathrm{q}}\cdot/q^2\,, \tag{A6a}$$

$$P_T = 1 - P_L = (q^2 1 - \vec{\mathrm{q}}\vec{\mathrm{q}}\cdot)/q^2 = -\vec{\mathrm{q}}\times\vec{\mathrm{q}}\times/q^2\,, \tag{A6b}$$

then Eq. (A4) may be rewritten as

$$\left(1 + \frac{4\pi\bar{\alpha}q^2P_T}{k_0^2 - q^2}\right)\vec{\mathrm{E}}(\vec{\mathrm{q}}) = -\left(\tfrac{8}{3}\pi 1 + \frac{4\pi q^2 P_T}{k_0^2 - q^2}\right)\vec{\mathrm{f}}_1(\vec{\mathrm{q}})$$

or, since $1 = P_L + P_T$, as

$$\left[P_L + \left(1 + \frac{4\pi\bar{\alpha}q^2}{k_0^2 - q^2}\right)P_T\right]\vec{\mathrm{E}}(\vec{\mathrm{q}}) = -\left[\frac{8\pi}{3}P_L + \left(\frac{8\pi}{3} + \frac{4\pi q^2}{k_0^2 - q^2}\right)P_T\right]\vec{\mathrm{f}}_1(\vec{\mathrm{q}})\,. \tag{A7}$$

Now, given any operator of the form

$$A = a_L(q)P_L + a_T(q)P_T\,, \tag{A8}$$

where $a_L(q)$ and $a_T(q)$ are scalar functions of q, its inverse is immediately given by

$$A^{-1} = [a_L(q)]^{-1}P_L + [a_T(q)]^{-1}P_T \tag{A9}$$

since $P_L^2 = P_L$, $P_T^2 = P_T$, and $P_LP_T = P_TP_L = 0$. Using these identities, we operate on both sides of Eq. (A7) with the inverse of the left-hand side

$$\vec{\mathrm{E}}(\vec{\mathrm{q}}) = \left(\frac{-8\pi}{3}P_L - \frac{\frac{8}{3}\pi + 4\pi q^2/(k_0^2 - q^2)}{1 + 4\pi\bar{\alpha}q^2/(k_0^2 - q^2)}P_T\right)\vec{\mathrm{f}}_1(\vec{\mathrm{q}})$$

or, after some straightforward rearrangement,

$$\vec{\mathrm{E}}(\vec{\mathrm{q}}) = -\left(\frac{8\pi}{3}1 + \frac{4\pi q^2 P_T}{(1 - \frac{4}{3}\pi\bar{\alpha})(\bar{k}^2 - q^2)}\right)\vec{\mathrm{f}}_1(\vec{\mathrm{q}})\,, \tag{A10}$$

where $\bar{k}^2 = \bar{\epsilon}k_0^2$ and $\bar{\epsilon}$ is defined by the Lorentz-Lorenz relation, (2.13).

Transforming back to the r representation, we obtain the desired expression for the operator L_1,

$$\vec{\mathrm{E}}(\vec{\mathrm{r}}) = L_1\vec{\overline{\mathrm{E}}}(\vec{\mathrm{r}}) = \eta\left(c_1\alpha_1(\vec{\mathrm{r}})\vec{\overline{\mathrm{E}}}(\vec{\mathrm{r}}) + c_2\nabla_r\times\nabla_r \times\int_\sigma \overline{G}(\vec{\mathrm{r}} - \vec{\mathrm{r}}_1)\alpha_1(\vec{\mathrm{r}}_1)\vec{\overline{\mathrm{E}}}(\vec{\mathrm{r}}_1)\,d^3r_1\right), \tag{A11a}$$

where $\overline{G}(\vec{\mathrm{r}} - \vec{\mathrm{r}}_1)$ is the average medium Green's function, Eq. (2.14b), and

$$c_1 = -\tfrac{8}{3}\pi(1 + \tfrac{8}{3}\pi\bar{\alpha})^{-1}\,, \tag{A11b}$$

$$c_2 = (1 - \tfrac{4}{3}\pi\bar{\alpha})^{-1}(1 + \tfrac{8}{3}\pi\bar{\alpha})^{-1}\,. \tag{A11c}$$

APPENDIX B

In this Appendix we consider the integral

$$\vec{\mathrm{K}}(\vec{\mathrm{r}}) = \int_\sigma d^3r_1\,S(|\vec{\mathrm{r}} - \vec{\mathrm{r}}_1|)\nabla_r\times\nabla_r\times\overline{G}(\vec{\mathrm{r}} - \vec{\mathrm{r}}_1)\langle\vec{\mathrm{E}}'(\vec{\mathrm{r}}_1)\rangle\,, \tag{B1}$$

where $\langle\vec{\mathrm{E}}'\rangle$ is assumed to be a linearly polarized plane wave; i.e., $\langle\vec{\mathrm{E}}'(\vec{\mathrm{r}})\rangle = \langle E'\rangle\hat{e}_x e^{i\vec{\mathrm{q}}\cdot\vec{\mathrm{r}}}$. Changing the variable of integration to $\vec{\rho} = \vec{\mathrm{r}} - \vec{\mathrm{r}}_1$, $\vec{\mathrm{K}}(\vec{\mathrm{r}})$ may be written

$$\vec{\mathrm{K}}(\vec{\mathrm{r}}) = \langle E'\rangle e^{i\vec{\mathrm{q}}\cdot\vec{\mathrm{r}}}\int_\sigma d^3\rho\,S(\rho)e^{-i\vec{\mathrm{q}}\cdot\vec{\rho}} \times\{\nabla_\rho[\nabla_\rho\cdot\overline{G}(\rho)\hat{e}_x] - [\nabla_\rho^2\overline{G}(\rho)\hat{e}_x]\}\,, \tag{B2}$$

where σ is now a small sphere about $\rho = 0$. Now,

$$\nabla_\rho^2[\overline{G}(\rho)\hat{e}_x] = [\nabla_\rho^2\overline{G}(\rho)]\hat{e}_x = -[\bar{k}^2\overline{G}(\rho) + 4\pi\delta(\rho)]\hat{e}_x \to -\bar{k}^2\overline{G}(\rho)\hat{e}_x\,, \tag{B3}$$

since the point $\rho = 0$ is excluded from the volume of integration. Similarly, the first term in the curly braces of (B2) reduces to

$$\nabla_\rho(\nabla_\rho\cdot\overline{G}(\rho)\hat{e}_x) \to \frac{\partial^2\overline{G}(\rho)}{\partial x^2}\hat{e}_x\,, \tag{B4}$$

since the y and z components of the vector are easily shown to integrate to zero, assuming that $S(\rho)$ is isotropic. Combining equations (B3) and (B4), we obtain

$$\vec{\mathrm{K}}(\vec{\mathrm{r}}) = \langle\vec{\mathrm{E}}'(\vec{\mathrm{r}})\rangle\int_\sigma d^3\rho\,S(\rho)e^{-i\vec{\mathrm{q}}\cdot\vec{\rho}} \times\left(\bar{k}^2\overline{G}(\rho) + \frac{\partial^2\overline{G}(\rho)}{\partial x^2}\right)\,. \tag{B5}$$

The derivative $\partial^2\overline{G}/\partial x^2$ is given by

$$\frac{\partial^2\overline{G}(\rho)}{\partial x^2} = \overline{G}(\rho)\left[\frac{i\bar{k} - 1/\rho}{\rho} + \frac{x^2}{\rho^2}\left(-\bar{k}^2 - \frac{3i\bar{k}}{\rho} + \frac{3}{\rho^2}\right)\right]. \tag{B6}$$

Substituting (B6) into (B5) and converting to spherical coordinates,

$$\vec{K}(\vec{r}) = \langle \vec{E}'(\vec{r})\rangle \int_\epsilon^\infty \rho^2\, d\rho\, S(\rho)\bar{G}(\rho) \int_C^\pi \sin\theta\, d\theta\, e^{-iq\rho\cos\theta} \int_0^{2\pi} d\phi \left[\left(\bar{k}^2 + \frac{(i\bar{k} - 1/\rho)}{\rho}\right) + \sin^2\theta\cos^2\phi\left(-\bar{k}^2 - \frac{3i\bar{k}}{\rho} + \frac{3}{\rho^2}\right)\right], \tag{B7}$$

where the z axis has been chosen parallel to the direction of the vector $\vec{q}$. Integrating over the angles and rearranging,

$$\vec{K}(\vec{r}) = \langle \vec{E}'(\vec{r})\rangle 4\pi\bar{k} \int_\epsilon^\infty \rho\, d\rho\, S(\rho)\bar{G}(\rho)\left[\frac{\bar{k}}{q}\sin(q\rho) + [i - (\bar{k}\rho)^{-1}]\frac{\sin(q\rho)}{q\rho} - \{\bar{k}\rho + 3[i - (\bar{k}\rho)^{-1}]\}\left(\frac{\sin(q\rho)}{(q\rho)^3} - \frac{\cos(q\rho)}{(q\rho)^2}\right)\right]. \tag{B8}$$

Anticipating that $\vec{K}$ will appear in a second-order perturbation term of the dispersion relation, Eqs. (4.8)–(4.10), we will only need the zero-order terms of $\vec{K}$ where we will find that $q = \bar{k}[1 + O(\eta^2)]$. Accordingly we may write

$$\vec{K}(\vec{r}) = K(\bar{k})\langle \vec{E}'(\vec{r})\rangle + O(\eta^4), \tag{B9a}$$

where

$$K(\bar{k}) \equiv 4\pi\bar{k} \int_0^\infty d\rho\, S(\rho) e^{i\bar{k}\rho} F(\bar{k}\rho), \tag{B9b}$$

and

$$F(\bar{k}\rho) = \sin(\bar{k}\rho) + [i - (\bar{k}\rho)^{-1}]\frac{\sin(\bar{k}\rho)}{\bar{k}\rho} - \{\bar{k}\rho + 3[i - (\bar{k}\rho)^{-1}]\}\left(\frac{\sin(\bar{k}\rho)}{(\bar{k}\rho)^3} - \frac{\cos(\bar{k}\rho)}{(\bar{k}\rho)^2}\right). \tag{B9c}$$

Expanding $F(x)$, we find it well behaved for small values of its argument, $F(x) = x(1 - \frac{4}{15}) - \frac{1}{15}ix^2 + \cdots$, thus the integral of equation (B9b) may be extended to zero with no difficulty.

ACKNOWLEDGMENT

The authors are grateful to Professor R. Victor Jones for suggesting the topic of this investigation and for providing guidance in its initial stages.

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*Supported in part by Grant No. DAHC15 73 G1 73 with the Advanced Research Projects Agency at Harvard University.

†Supported in part by NSF Grant No. DMR-72-03209.

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[10]In Ref. 8 self-field terms were omitted since the Green's function [Eqs. (92)–(94)] contains no δ function. In Ref. 9, the Green's function does contain a δ function [Eq. (3.3)] and hence the expression for the effective dielectric constant [Eq. (3.13)] does include a self-field term. However, this term is a negative perturbation which is physically impossible since the mean squared fluctuations can only slow the coherent wave.

[11]M. Born and E. Wolf, *Principles of Optics*, 2nd ed. (Pergamon, New York, 1964), Sec. 2.4.

[12]M. Born and E. Wolf, in Ref. 11, Appendix V.

[13]To transform the last term of Eq. (2.7), it is convenient to use the identities; $G_0(\vec{r}) = (2\pi^2)^{-1}\int dk^3 \exp(i\vec{k}\cdot\vec{r})/(k^2 - k_0^2)$ and $\int dr^3 \exp[-i\vec{r}\cdot(\vec{k}-\vec{q})] = (2\pi)^3\delta(\vec{k}-\vec{q})$, where $\delta(\vec{r})$ is the three-dimensional Dirac δ function.

[14]Consider the terms in the large parentheses of Eq. (4.8) after multiplying through by $(q^2 - k^2)$. Since $(q^2 - \bar{k}^2) \sim \eta^2$, the second term is of order η^4 and higher. A second-order contribution from the third term is obtained by allowing $\vec{q} = \vec{k}$ and $\langle \vec{E}'\rangle = \vec{E}'$, so that $\vec{q}\times\vec{q}\times\langle\vec{E}'\rangle \to -\bar{k}^2\vec{E}'$.

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