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Electromagnetic Theory of Light Scattering from an Inhomogeneous Fluid

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The methods of geometric optics are employed to determine the spectral distribution of the time averaged intensity of light scattered by thermal fluctuations in an inhomogeneous fluid. It is assumed that all average quantities undergo little change over distances comparable to a wavelength of the incident or scattered light. An elementary example corresponding to gravitational effects is considered to indicate the use of the method.

1. INTRODUCTION

In the last decade interest in the theory of light scattering from fluctuations has rapidly accelerated due to breakthroughs in instrument development. The high spectral resolution of modern detectors and the narrow bandwidth of the laser has made it possible to observe both the temporal and spatial properties of correlations in transparent media. One significant limitation in the present theory of light scattering is the assumption that the system supporting the fluctuations which produce the scattering is spatially homogeneous. Consider, for example, the scattering of light from a system near the critical point. As the system approaches the critical point, the compressibility becomes very large. Consequently, the gravitational field, which is so weak that it may be neglected far from the critical point, produces a large density gradient near the critical point. The effect of the large density gradient has been ignored in both the electromagnetic and thermodynamic theory of light scattering from a system near the critical point [1] There are many other examples relating to biological and chemically reactive systems.

The present electromagnetic theory of light scattering from homogeneous systems has evolved along two distinct lines. The Einstein approach [2–4] assumes that the fluctuations are small and linearizes the appropriate Maxwell equations governing the response of the system. The result is identical to the Born approximation. The method used by Landau and Placzek, [5–8] applies similar approx-

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imations to the integral equations governing the electric and magnetic fields. If, in addition to the Born approximation, we are willing to assume that there is very little variation in the ensemble average of thermodynamic quantities associated with the system over distances comparable to a wavelength of the incident or scattered light, we may employ the methods of geometric optics [9] to obtain the spectral distribution of the scattered light from an inhomogeneous system.

In this article we develop a method for determining the spectral distribution of light scattered by an inhomogeneous fluid. We first apply the Born approximation to Maxwell's equations to obtain a second-order partial differential equation for the scattered electric field and another for the magnetic field. We then use the methods of geometric optics to obtain a Green's function dyadic [10] for each field. Finally, we consider an elementary example to indicate some of the steps in the application of the procedure to a particular problem.

2. Electromagnetic Theory: Differential Equations Governing Fields

Our purpose is to determine the electric and magnetic fields $\mathbf{E}_1$ and $\mathbf{H}_1$ produced by the scattering of light by fluctuations in an isotropic spatially inhomogeneous system composed of isotropic molecules. We shall restrict our study to those Fourier components of $\mathbf{E}_1$ and $\mathbf{H}_1$ which are characterized by wavelengths greatly in excess of the distance between adjacent molecules. The same considerations, of course, apply to the unscattered light. The response of the fluid is determined by an instantaneous permitivity ϵ, an instantaneous permeability μ, and the appropriate Maxwell equations:

$$\nabla \times \mathbf{H} = \frac{1}{c} \frac{\partial}{\partial t} \epsilon \mathbf{E}, \tag{2.1}$$

$$\nabla \times \mathbf{E} = -\frac{1}{c} \frac{\partial}{\partial t} \mu \mathbf{H}, \tag{2.2}$$

$$\nabla \cdot \mu \mathbf{H} = 0, \tag{2.3}$$

$$\nabla \cdot \epsilon \mathbf{E} = 0, \tag{2.4}$$

where $\mathbf{E}$ and $\mathbf{H}$ are the total electric and magnetic fields.

We may regard ϵ as the sum of two terms, an average term and a fluctuation, and similarly for μ:

$$\epsilon = \epsilon_0 + \epsilon_1 = \epsilon_0(1 + \xi), \tag{2.5}$$

$$\mu = \mu_0 + \mu_1 = \mu_0(1 + \eta), \tag{2.6}$$

$$\epsilon_0 = \langle \epsilon \rangle = \epsilon_0(\mathbf{x}), \tag{2.7}$$

$$\mu_0 = \langle \mu \rangle = \mu_0(\mathbf{x}), \tag{2.8}$$

$$|\xi| \ll 1, \qquad |\eta| \ll 1. \tag{2.9}$$

The brackets in Eqs. (2.7) and (2.8) represent the ensemble average. We shall only consider systems which obey the relationships (2.9).

The equations governing the scattered fields $\mathbf{E}_1$ and $\mathbf{H}_1$ are found by writing

$$\mathbf{E} = \mathbf{E}_0 + \mathbf{E}_1 , \tag{2.10}$$

$$\mathbf{H} = \mathbf{H}_0 + \mathbf{H}_1 , \tag{2.11}$$

where $\mathbf{E}_0$ and $\mathbf{H}_0$ are the fields in the absence of fluctuations. We only retain terms of the first order in small quantities after substitution of Eqs. (2.5), (2.6), (2.10), and (2.11) into Eqs. (2.1)–(2.4).

We quickly obtain

$$\nabla \times \mathbf{H}_0 = \frac{1}{c} \frac{\partial}{\partial t} \epsilon_0 \mathbf{E}_0 , \tag{2.12}$$

$$\nabla \times \mathbf{E}_0 = - \frac{1}{c} \frac{\partial}{\partial t} \mu_0 \mathbf{H}_0 , \tag{2.13}$$

$$\nabla \cdot \mu_0 \mathbf{H}_0 = 0, \tag{2.14}$$

$$\nabla \cdot \epsilon_0 \mathbf{E}_0 = 0, \tag{2.15}$$

$$\nabla \times \mathbf{H}_1 = \frac{1}{c} \frac{\partial}{\partial t} \epsilon_0 (\mathbf{E}_1 + \xi \mathbf{E}_0), \tag{2.16}$$

$$\nabla \times \mathbf{E}_1 = - \frac{1}{c} \frac{\partial}{\partial t} \mu_0 (\mathbf{H}_1 + \eta \mathbf{H}_0), \tag{2.17}$$

$$\nabla \cdot \mu_0 (\mathbf{H}_1 + \eta \mathbf{H}_0) = 0, \tag{2.18}$$

$$\nabla \cdot \epsilon_0 (\mathbf{E}_1 + \xi \mathbf{E}_0) = 0. \tag{2.19}$$

Equations (2.16)–(2.19) are simply the differential form of the Born approximation for $\mathbf{E}_1$ and $\mathbf{H}_1$.

Our prescription is to construct a second-order partial differential equation obeyed by $\mathbf{E}_1$ alone, and another obeyed by $\mathbf{H}_1$. We shall then determine the appropriate Green's dyadics for these inhomogeneous equations in the assymptotic limit of geometric optics. Many of the required mathematical manipulations are given in great detail by Born and Wolf [11]. We shall refer to this treatise frequently and, in particular, to the 1970 edition. Chapters 1 and 3 of all the editions starting with 1959 are substantially alike. All references to particular equations and paragraphs in Born and Wolf will be preceded by an S.

We apply the procedures used by Born and Wolf to obtain Eq. (S 1.2.5) from

Eq. (S 1.2.1) (their notation) to our Eqs. (2.16)–(2.19), making use of Eqs. (2.14) and (2.15), to obtain

$$D_1\mathbf{E}_1 = -4\pi\mathbf{P}, \tag{2.20}$$

$$D_2\mathbf{H}_1 = -4\pi\mathbf{Q}. \tag{2.21}$$

In Eqs. (2.20) and (2.21),

$$D_1(\mathbf{x}, \epsilon_0, \mu_0) = \nabla^2 - \frac{n^2}{c^2}\frac{\partial^2}{\partial t^2} + \nabla[(\nabla \log \epsilon_0)] \cdot + (\nabla \log \mu_0) \times [\nabla \times \quad , \tag{2.21}$$

$$D_2 = D_1(\mathbf{x}, \mu_0, \epsilon_0), \tag{2.22}$$

$$4\pi\mathbf{P} = \nabla[\nabla + (\nabla \log \epsilon_0)] \cdot \mathbf{A} - \frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{A} - \frac{\mu_0}{c}\frac{\partial}{\partial t}\nabla \times \mathbf{B}, \tag{2.24}$$

$$4\pi\mathbf{Q} = \nabla[\nabla + (\nabla \log \mu_0)] \cdot \mathbf{B} - \frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{B} + \frac{\epsilon_0}{c}\frac{\partial}{\partial t}\nabla \times \mathbf{A}, \tag{2.25}$$

$$\mathbf{A} = \xi\mathbf{E}_0, \qquad \mathbf{B} = \eta\mathbf{H}_0, \qquad n^2 = \epsilon_0\mu_0. \tag{2.26}$$

A and **B** vanish outside of the scattering volume, and in particular in the radiation zone, because ξ and η are assumed to be different than zero only in the finite scattering volume V. In an actual experiment this is accomplished by allowing $\mathbf{E}_0$ and $\mathbf{H}_0$ to be different than zero only in V.

To determine the spectral distribution of the time averaged radiated power, we need to know the temporal Fourier transforms of $\mathbf{E}_1$ and $\mathbf{H}_1$. To insure that the transforms exist we replace ξ by ξ_T,

$$\begin{aligned} \xi_T &= 0, \qquad |t| > T, \\ \xi_T &= \xi, \qquad |t| < T \end{aligned} \tag{2.27}$$

and similarly for all other quantities so labeled. We will eventually determine the spectral distribution of the time averaged intensity in the limit $T \to \infty$. Writing

$$\mathbf{E}_1{}^\dagger(\mathbf{x}, \omega) = \frac{1}{2\pi}\int dt\, e^{i\omega t}\mathbf{E}_1(\mathbf{x}, t), \tag{2.28}$$

and similarly for all quantities so indicated we obtain, after taking the transform of Eqs. (2.10) and (2.21),

$$D_1{}^\dagger\mathbf{E}_1{}^\dagger = -4\pi\mathbf{P}_T{}^\dagger, \tag{2.29}$$

$$D_2{}^\dagger\mathbf{H}_1{}^\dagger = -4\pi\mathbf{Q}_T{}^\dagger, \tag{2.30}$$

where

$$D_1{}^\dagger = D_1{}^\dagger(\mathbf{x}, k; \epsilon_0, \mu_0)$$
$$= \nabla^2 + n^2k^2 + \nabla(\nabla \log \epsilon_0) \cdot +(\nabla \log \mu_0) \times \nabla \times \quad , \tag{2.31}$$

$$D_2{}^\dagger = D_1{}^\dagger(\mathbf{x}, k; \mu_0, \epsilon_0), \tag{2.32}$$

$$k = \frac{\omega}{c}, \tag{2.33}$$

$$4\pi \mathbf{P}_T{}^\dagger = \nabla[\nabla + (\nabla \log \epsilon_0)] \cdot \mathbf{A}_T{}^\dagger + k^2n^2\mathbf{A}_T{}^\dagger + ik\mu_0\nabla \times \mathbf{B}_T{}^\dagger, \tag{2.34}$$

$$4\pi \mathbf{Q}_T{}^\dagger = \nabla[\nabla + (\nabla \log \mu_0)] \cdot \mathbf{B}_T{}^\dagger + k^2n^2\mathbf{B}_T{}^\dagger - ik\epsilon_0\nabla \times \mathbf{A}_T{}^\dagger. \tag{3.35}$$

If the molecules are not isotropic, the fluctuations ϵ_1 and μ_1 are dyadics, $\boldsymbol{\varepsilon}_1$ and $\boldsymbol{\mu}_1$. For such a system the only modification of the above is to replace $\mathbf{A}$ by $\boldsymbol{\xi} \cdot \mathbf{E}_0$ and $\mathbf{B}$ by $\boldsymbol{\eta} \cdot \mathbf{H}_0$, where

$$\boldsymbol{\xi} = \boldsymbol{\varepsilon}_1/\epsilon_0, \qquad \boldsymbol{\eta} = \boldsymbol{\mu}_1/\mu_0. \tag{2.36}$$

3. Green's Dyadics for $D_1{}^\dagger$ and $D_2{}^\dagger$

The Green's dyadics for the operators $D_1{}^\dagger$ and $D_2{}^\dagger$ are, in general, very difficult to obtain. However, if we are interested in $\mathbf{E}_1{}^\dagger$ and $\mathbf{H}_1{}^\dagger$ for values of k such that

$$\left| \frac{1}{k} \nabla \log \mu_0 \right| \ll 1, \qquad \left| \frac{1}{k} \nabla \log \epsilon_0 \right| \ll 1, \tag{3.1}$$

we may apply the techniques of geometric optics (see Ref. 11, Chap. 3), or what is more generally referred to as the Eikonal method [6], to obtain the Green's dyadic $\mathsf{G}_1{}^\dagger(\mathbf{x}, \mathbf{x}', k; \epsilon_0, \mu_0)$ associated with the operator $D_1{}^\dagger$. The dyadic $\mathsf{G}_2{}^\dagger$ corresponding to $D_2{}^\dagger$ is obtained by interchanging ϵ_0 and μ_0 in $\mathsf{G}_1{}^\dagger$. We are, of course, seeking the fundamental Green's dyadics corresponding to outgoing waves at infinity. The differential equation satisfied by $\mathsf{G}_1{}^\dagger$ is

$$D_1{}^\dagger \mathsf{G}_1{}^\dagger = -4\pi\delta(\mathbf{x} - \mathbf{x}')\mathsf{I}, \tag{3.2}$$

where I is the identity dyadic and $\delta(\mathbf{x} - \mathbf{x}')$ is the three-dimensional Dirac delta function.

We write

$$\mathsf{G}_1{}^\dagger = e^{ik\mathscr{L}} \sum_{j=1}^{3} \mathbf{e}_j \hat{\boldsymbol{\chi}}_j, \tag{3.3}$$

where the $\hat{\boldsymbol{\chi}}_j$ are an orthonormal basis set for a three-dimensional system of orthogonal coordinates χ_j. When $\mathbf{x} \neq \mathbf{x}'$ we observe that each $\mathbf{e}_j$ satisfies Eq. (S 3.1.41) and $\mathscr{L}$ satisfies Eq. (S 3.1.15a) through first order in small quantities. We obtain $\mathscr{L}$ immediately from Fermat's principle (Ref. 11 Chap. 3.3.2)

$$\mathscr{L}(\mathbf{x}, \mathbf{x}') = \int_{C\,\mathbf{x}'}^{\mathbf{x}} ds\, n, \tag{3.4}$$

where C is a contour that minimizes $\mathscr{L}$ and ds is an element of arc along C. In the development presented here we shall assume that n is everywhere continuous so that the solution to Eq. (3.4) is unique. Discontinuities in n will give rise to multiple contours and will complicate the solutions. Hence, the effects of any reflecting surfaces must be dealt with by the experimentalist.

The magnitude of $\mathbf{e}_j$ is given by Eq. (S 3.1.45) and is seen to be independent of the subscript j. Thus, we may write

$$\mathbf{e}_j = \mathscr{E}\hat{\mathbf{u}}_j, \tag{3.5}$$

where $\hat{\mathbf{u}}_j$ is a unit vector. From Eq. (S 3.1.45),

$$(\mathscr{E})_2 = (\mathscr{E})_1 \exp\left[-\frac{1}{2}\int_{C\,s_1}^{s_2} ds \frac{n}{\epsilon_0} \nabla \cdot \left(\frac{1}{\mu_0} \nabla \mathscr{L}\right)\right], \tag{3.6}$$

where the subscripts 1 and 2 signify two points on the ray trajectory C. Let $\hat{\mathbf{s}}$ denote the tangent vector to C,

$$\hat{\mathbf{s}} = \frac{d\mathbf{x}}{ds}. \tag{3.7}$$

We must choose $\hat{\mathbf{s}}$ pointing away from $\mathbf{x}'$ and toward $\mathbf{x}$, that is distance must be measured along C going from $\mathbf{x}'$ to $\mathbf{x}$, to insure that $\mathbf{G}_1^{\dagger}$ corresponds to an outgoing wave. Hence

$$\nabla \mathscr{L} = n\hat{\mathbf{s}} \tag{3.8}$$

and the $\hat{\mathbf{u}}_j$ satisfy the differential equation (see Eq. (S 3.1.48))

$$\frac{d\hat{\mathbf{u}}_j}{ds} = -(\hat{\mathbf{u}}_j \cdot \nabla \log n)\, \hat{\mathbf{s}}. \tag{3.9}$$

To illuminate the behavior of $\mathbf{G}_1^{\dagger}$ as $\mathbf{x}$ approaches $\mathbf{x}'$, we write Eq. (3.6) in the form

$$(\mathscr{E}m^{1/2})_2 = (\mathscr{E}m^{1/2})_1 \exp\left[-\frac{1}{2}\int_{C\,s_1}^{s_2} ds\, \nabla \cdot \hat{\mathbf{s}}\right], \tag{3.10}$$

where

$$m = (\epsilon_0/\mu_0)^{1/2}. \tag{3.11}$$

Writing $\mathbf{r} = \mathbf{x} - \mathbf{x}'$, $r = |\mathbf{r}|$, $\hat{\mathbf{r}} = \mathbf{r}/r$, we observe that if $0 < \ell r < 1$, then $\hat{\mathbf{s}} \simeq \hat{\mathbf{r}}$ as a result of inequalities (3.1). Hence, from Eq. (3.10), as r approaches zero,

$$\mathscr{E} \cong \frac{1}{r}\mathscr{E}', \tag{3.12}$$

where $\mathscr{E}'$ is nonsingular at $r = 0$. If we multiply Eq. (3.2) by r^2 (r^2 factors the Jacobian in spherical coordinates) we observe that only the contribution from

$$e^{i\ell\mathscr{L}}(\nabla^2\mathscr{E}) \sum \hat{\mathbf{u}}_j\hat{\boldsymbol{\chi}}_j$$

is singular on the left side. Recalling that

$$\delta(\mathbf{r}) = \frac{1}{2\pi r^2}\,\delta(r),$$

we obtain two results: First of all, $(\mathscr{E})_1$ in Eqs. (3.6) and (3.10) must be constructed in such a manner that $\mathscr{E}'$ approaches unity as r approaches zero; secondly, at $\mathbf{x} = \mathbf{x}'$, $\hat{\mathbf{u}}_j = \hat{\boldsymbol{\chi}}_j$. These two results coupled with Eqs. (3.6) and (3.9) completely determine $\mathscr{E}$ and $\hat{\mathbf{u}}_j$.

Carrying out the above procedures for the dyadic $\mathsf{G}_2{}^\dagger$, we quickly find

$$\mathsf{G}_2{}^\dagger = e^{i\ell\mathscr{L}}\mathscr{H} \sum \hat{\mathbf{u}}_j\boldsymbol{\chi}_j\,, \tag{3.13}$$

where

$$\mathscr{H} = \mathscr{E}m(\mathbf{x})/m(\mathbf{x}'). \tag{3.14}$$

In the asymptotic limit of geometric optics, we obtain

$$\mathbf{E}_1{}^\dagger(\mathbf{x}, \omega) = \int d^3\chi'\, \mathsf{G}_1{}^\dagger \cdot \mathbf{P}_T{}^\dagger(\mathbf{x}', \omega)(1 + \text{order } \delta), \tag{3.15}$$

$$\mathbf{H}_1{}^\dagger(\mathbf{x}, \omega) = \int d^3\chi'\, \mathsf{G}_2{}^\dagger \cdot \mathbf{Q}_T{}^\dagger(\mathbf{x}', \omega)(1 + \text{order } \delta), \tag{3.16}$$

where δ is on the order of $|(1/\ell)\, \nabla \log \epsilon_0|$ or $|(1/\ell)\, \nabla \log \mu_0|$. There are, of course, other corrections due to the fact that we started with the Born approximation.

The time averaged energy flux is given by

$$\mathbf{S} = \frac{c}{4\pi} \lim_{T\to\infty} \frac{1}{2T} \int dt\, \mathbf{E}_1 \times \mathbf{H}_1 = \lim_{T\to\infty} \frac{c}{2T} \int_0^\infty d\omega\, \mathscr{R}(\mathbf{E}_1^{\dagger *} \times \mathbf{H}_1{}^\dagger). \tag{3.17}$$

Hence, the spectral distribution of the time averaged intensity is given by

$$\mathscr{I}(\mathbf{x}, \omega) = \lim_{T\to\infty} \frac{c}{2T} \left| \mathscr{R}(\mathbf{E}_1^{\dagger *} \times \mathbf{H}_1{}^{\dagger}) \right|. \tag{3.18}$$

We mention that, to the same order of accuracy, the usual methods of geometric optics may be employed to determine the unperturbed fields $\mathbf{E}_0$ and $\mathbf{H}_0$. We shall perform the details of this procedure in the following example.

4. An Application: General Considerations

We shall consider the effect of the gravitational field on $\mathsf{G}_1{}^{\dagger}$, $\mathsf{G}_2{}^{\dagger}$, $\mathbf{E}_0$, and $\mathbf{H}_0$ under very special restrictions for a nonmagnetic ($\mu_0 = 1$, $\eta = 0$) fluid. Before we make any restrictions let us consider some aspects the general problem. In the remainder of the discussion we shall use (χ, y, z), a Cartesian coordinate system with basis set $(\hat{\boldsymbol{\chi}}, \hat{\mathbf{y}}, \hat{\mathbf{z}})$. We take $\hat{\mathbf{z}}$ in the vertical direction with the gravitational field in the $-\hat{\mathbf{z}}$ direction. The scattering medium is confined to a cylinder with its centroid at the origin, axis of symmetry prallel to $\hat{\mathbf{z}}$, height $2L$, and radius L. Within the cylinder $\epsilon_0 = \epsilon_0(z)$. In order to avoid introducing any discontinuities in ϵ_0 we divide space into three regions (see Fig. 1) and set

$$\begin{aligned} \epsilon_0 &= \epsilon_0(L), && z > L, \\ \epsilon_0 &= \epsilon_0(z), && |z| \leqslant L, \\ \epsilon_0 &= \epsilon_0(-L), && z < -L. \end{aligned} \tag{4.1}$$

We are interested in constructing ray trajectories diverging from some point $\mathbf{x}'$ within the cylinder. Assuming that ϵ_0 is a monotonically decreasing function of z, the rays can belong to one of three classes (see Fig. 1). If, at $\mathbf{x} = \mathbf{x}'$, $\hat{\mathbf{s}} \cdot \hat{\mathbf{z}} < 0$, then $\hat{\mathbf{s}} \cdot \hat{\mathbf{z}} < 0$ along the complete trajectory (class I); if $\hat{\mathbf{s}} \cdot \hat{\mathbf{z}} > 0$ at $\mathbf{x} = \mathbf{x}'$, and if $\hat{\mathbf{s}} \cdot \hat{\mathbf{z}} > 0$ at $z = L$, then $\hat{\mathbf{s}} \cdot \hat{\mathbf{z}} > 0$ along the complete trajectory (class II); finally, if at $\mathbf{x} = \mathbf{x}'$, $\hat{\mathbf{s}} \cdot \hat{\mathbf{z}} > 0$ but for some $|z| < L$, $\hat{\mathbf{s}} \cdot \hat{\mathbf{z}} = 0$, then for the remainder of the trajectory $\hat{\mathbf{s}} \cdot \hat{\mathbf{z}} < 0$. By performing all of our measurements in the region $z > L$ we need only consider trajectories of class II and eliminate the necessity of dealing with multiple valued functions. If ϵ_0 is monotonically increasing the situation is naturally reversed.

Directing the incident light parallel to the z axis allows another simplification in that the ray trajectories of the incident light will not be bent. The calculation of $\mathbf{E}_0$ and $\mathbf{H}_0$ within V is then greatly simplified.

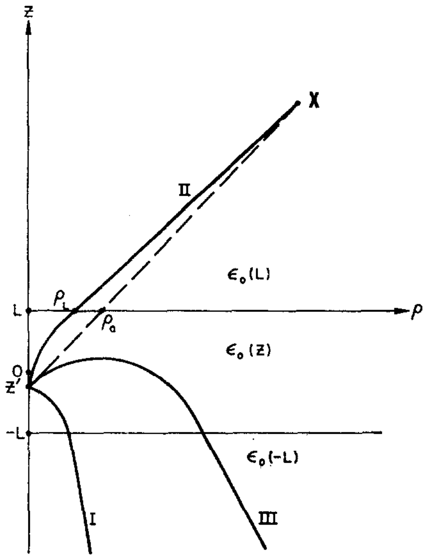

FIG. 1. Classification of ray trajectories.

We may take advantage of the cylindrical symmetry of the experimental geometry by defining the vector $\boldsymbol{\rho}$:

$$\boldsymbol{\rho} = (\mathbf{I} - \hat{\mathbf{z}}\hat{\mathbf{z}}) \cdot (\mathbf{x} - \mathbf{x}') = -\hat{\mathbf{z}} \times [\hat{\mathbf{z}} \times (\mathbf{x} - \mathbf{x}')], \tag{4.2}$$

$$\rho = |\boldsymbol{\rho}|, \qquad \hat{\boldsymbol{\rho}} = \boldsymbol{\rho}/\rho. \tag{4.3}$$

Clearly, $\mathbf{G}_1{}^\dagger$ and $\mathbf{G}_2{}^\dagger$ depend solely on the three coordinates ρ, z and, z', as opposed to six coordinates in the general case.

5. Specific Example

For the remainder of this article we address ourselves to what is, perhaps, the most elementary specific case. We assume that, for $|z| < L$, ϵ_0 varies so slowly that it is adequately represented by the first two terms of a Taylor series about $z = 0$:

$$\epsilon_0(z) = a^2(1 - 2\alpha z), \qquad |z| < L, \tag{5.1}$$

where a and α are both positive definite and $\alpha L \ll 1$.

In this section we shall determine the appropriate form of $\mathbf{G}_1{}^\dagger$ and $\mathbf{G}_2{}^\dagger$ in the region $|z| < L$ ($|z'| < L$ throughout). Over this region, $\mathscr{L}$, $\mathscr{E}$, $\mathscr{H}$ and $\hat{\mathbf{u}}_j$ possess series expansions in powers of αz and $\alpha z'$. If we agree to ignore all terms of order αL in our final result, we may immediately set

$$\mathscr{E} = \mathscr{H} = 1/r, \tag{5.2}$$

$$\hat{\mathbf{u}}_j = \hat{\boldsymbol{\chi}}_j \,. \tag{5.3}$$

But, in a typical light scattering experiment, we are observing the spectral distribution of the intensity over regions of ω such that $kL \gg 1$. Hence, we may *not* set $\mathscr{L} = ar$ in the exponential in Eqs. (3.3) and (3.13). In the next section we shall show that $\mathscr{L}$ has the form

$$\mathscr{L} = ar(1 + \delta), \tag{5.4}$$

where δ is of order αL and represents the first-order correction to $\mathscr{L}$ due to the fact that $\alpha \neq 0$. As $k\alpha L^2$ may be of order unity, or larger, we many not drop the term $ikra\delta$ from $e^{ik\mathscr{L}}$. To insure that the third and higher order terms in the series expansion of $\mathscr{L}$ in powers of az and $\alpha z'$ are unnecessary we shall require

$$k\, \alpha^2 L^3 \ll 1.$$

To briefly summarize, we have found that $\mathsf{G}_1{}^{\dagger}$ and $\mathsf{G}_2{}^{\dagger}$ have the representation

$$\mathsf{G}_1{}^{\dagger} = \mathsf{G}_2{}^{\dagger} = \frac{1}{r}\, \mathsf{I} \exp[ikar(1 + \delta)], \qquad | \mathbf{r} \cdot \hat{\mathbf{z}} | < L, \tag{5.5}$$

with the restrictions

$$\alpha L \ll 1, \tag{5.6}$$

$$kL \gg 1, \tag{5.7}$$

$$k\alpha^2 L^3 \ll 1, \tag{5.8}$$

$$\frac{\alpha}{k} \ll 1, \tag{5.9}$$

where the last restriction is a consequence of the range of validity of geometric optics, and in this case is clearly redundant.

6. The Eikonal

The Euler Lagrange equation defining the trajectory $C(\mathbf{x}, \mathbf{x}')$ may be written in the form (see Eq. (S 3.2.2))

$$\frac{d}{ds} n\hat{\mathbf{s}} = \nabla n. \tag{6.1}$$

We define ζ by

$$\cos\zeta = \hat{\mathbf{z}}\cdot\hat{\mathbf{s}}, \qquad \sin\zeta = \hat{\boldsymbol{\rho}}\cdot\hat{\mathbf{s}} \tag{6.2}$$

and note that, for trajectories of class II, $0 \leqslant \zeta < \pi/2$ and $z > z'$. In our example n is a function of z so that, from Eq. (6.1)

$$n\sin\zeta = n'\sin\zeta' \tag{6.3}$$

along a ray trajectory, where the primes denote quantities evaluated at $\mathbf{x} = \mathbf{x}'$, the point of origin. Hence,

$$\cos\zeta = \left[1 - \left(\frac{n'}{n}\sin\zeta'\right)^2\right]^{1/2}. \tag{6.4}$$

Expanding the right side through first order in αz and $\alpha z'$ we obtain

$$\cos\zeta = \cos\zeta'[1 - \alpha(z - z')\tan^2\zeta'] \tag{6.5}$$

and, from Eq. (6.3),

$$\sin\zeta = \sin\zeta'[1 + \alpha(z - z')]. \tag{6.6}$$

Along a particular trajectory ρ may be regarded as a function of z. From

$$\frac{d\rho}{dz} = \tan\zeta = \tan\zeta'[1 + \alpha(z - z')\sec^2\zeta'], \tag{6.7}$$

we obtain

$$\begin{aligned} \rho &= (z - z')\tan\zeta'\left[1 + \frac{\alpha}{2}(z - z')\sec^2\zeta'\right], \\ r &= [(z - z')^2 + \rho^2]^{1/2} = \rho\sin\zeta', \end{aligned} \tag{6.9}$$

$$\begin{aligned} \mathscr{L} &= \int_{\mathbf{x}' \atop C}^{\mathbf{x}} ds\, n = \int_{z'}^{z} dz''\left[1 + \left(\frac{d\rho}{dz''}\right)^2\right]^{1/2} n \\ &= ar\left[1 - \frac{\alpha}{2}(z + z')\right] \end{aligned} \tag{6.10}$$

through first order. Equation (6.10) is correct when $|z| < L$ and $|z'| < L$. When $|z'| < L$ but $z > L$ we may write

$$\mathscr{L} = a(1 - 2\alpha L)^{1/2} r_1 + ar_2\left[1 - \frac{\alpha}{2}(L + z')\right], \tag{6.11}$$

where

$$r_1 = [(z - L)^2 + (\rho - \rho_2)^2]^{1/2}, \tag{6.12}$$

$$r_2 = [(L - z')^2 + \rho_L{}^2]^{1/2} \tag{6.13}$$

and ρ_L is the value of ρ at the intersection of the ray trajectory with the line $z = L$ (see Fig. 1). We now set

$$\rho_L = \rho_0 + \Delta\rho, \tag{6.14}$$

where ρ_0 is the intersection of the straight line through (ρ, z) and $(0, z)$ and the line $z = L$ (see Fig. 1). Expanding $\mathscr{L}$ through first order in $\Delta\rho$ and making use of the relationship between similar triangles, we finally obtain

$$\mathscr{L} = a(1 - 2\alpha L)^{1/2} R_0 + \frac{a\alpha}{2}(L - z')^2 R_0/(z - z'), \tag{6.15}$$

where

$$R_0 = [\rho^2 + (z - z')^2]^{1/2}. \tag{6.16}$$

Writing $n_0 = n(L)$, $R = |\mathbf{x}|$, and expanding R_0 about R we obtain

$$\mathscr{L} = n_0 \left\{ R - \hat{\mathbf{n}} \cdot \mathbf{x}' + \frac{\alpha}{2}(L - z')^2 \sec\theta \right\} + \text{order}\,\frac{L^2}{R} \tag{6.17}$$

in the raditation zone ($R \gg L$). In Eq. (5.18),

$$\begin{aligned} \hat{\mathbf{n}} &= \mathbf{x}/R, \\ \cos\theta &= \hat{\mathbf{n}} \cdot \hat{\mathbf{z}}. \end{aligned} \tag{6.18}$$

The direction of propagation of the scattered radiation, $\hat{\mathbf{s}}$ is quickly obtained from

$$\hat{\mathbf{s}} = \frac{1}{n_0} \nabla \mathscr{L} = \hat{\mathbf{n}} + \text{order}\,\frac{L}{R}. \tag{6.20}$$

The Green's dyadics are, from Eqs. (6.17), (5.2), and (5.3),

$$\mathsf{G}_1{}^\dagger = \mathsf{G}_2{}^\dagger = G^\dagger \mathsf{I}, \tag{6.21}$$

$$G^\dagger = \frac{1}{R} \exp\left\{ i k n_0 \left[R - \hat{\mathbf{n}} \cdot \mathbf{x}' + \frac{\alpha}{2}(L - z')^2 \sec\theta \right] \right\}. \tag{6.22}$$

At this point it is appropriate to introduce one further restriction;

$$0 < \theta \leqslant 1.36\ \text{rad}. \tag{6.23}$$

Clearly, if we allow θ to approach $\pi/2$, our method of approximation breaks down. Our choice of 1.36 rad is to insure that

$$\frac{\alpha}{2}(L - z')^2 \sec\theta \leqslant 10\,\alpha L^2,$$

which is admittedly arbitrary.

7. Spectral Distribution

We suppose that the fluid is excited by a monochromatic, linearly polarized plane wave with angular frequency ω_0 propagating in the z direction. Taking $\hat{\chi}$ for the direction of polarization we may write

$$\mathbf{E}_0 = \hat{\chi}\mathscr{E}_0\mathscr{R}e^{i(k_0 n_0 z - \omega_0 t)} \tag{7.1}$$

in the region $z > L$. In order to determine $\mathscr{I}(\mathbf{x}, \omega)$, the spectral distribution of the time averaged radiated intensity, we must construct $\mathbf{E}_0$ in the region $|z| < L$. Applying the arguments of the previous section we may write

$$\mathbf{E}_0 = \hat{\chi}\mathscr{E}_0\mathscr{R}e^{i(k_0\mathscr{L}_0 - \omega_0 t)}, \qquad |z| < L, \tag{7.2}$$

where $k_0 = \omega_0/c$ and

$$\mathscr{L}_0 = \text{const} + \int^z dz'\, a(1 - \alpha z'). \tag{7.3}$$

As $\mathbf{E}_0$ is continuous at $z = L$ we obtain

$$\mathbf{E}_0 = \hat{\chi}\mathscr{E}_0\mathscr{R} \exp\{ik_0 n_0[z - \tfrac{1}{2}\alpha(L - z)^2] - i\omega_0 t\} \tag{7.4}$$

in the region $|z| < L$.

To obtain the scattered fields $\mathbf{E}_1{}^\dagger$ and $\mathbf{H}_1{}^\dagger$, we insert Eq. (7.4) into Eqs. (2.34) and (2.35) ($\mathbf{B}_T \equiv 0$ in this example) and insert Eqs. (6.21) and (6.22) into Eqs. (3.15) and (3.16). We then integrate by parts and drop terms of order αL to obtain

$$\mathbf{H}_1{}^\dagger = \frac{a\mathscr{E}_0}{2R}\left(\frac{ka}{2\pi}\right)^2 e^{ikn_0R}(\hat{\mathbf{n}} \times \hat{\chi})\, F, \tag{7.5}$$

$$\mathbf{E}_1{}^\dagger = -\frac{1}{a}\hat{\mathbf{n}} \times \mathbf{H}_1{}^\dagger, \tag{7.6}$$

$$F(\hat{\mathbf{n}}, \omega) = \int_V d^3\chi' \int_{-T}^{T} dt'\, \xi e^{-i(k\mathscr{L}_1 - \omega t')} \cos(k_0\mathscr{L}_0 - \omega_0 t'), \tag{7.7}$$

where, in Eq. (7.7),

$$\mathscr{L}_1 = n_0\left[\hat{\mathbf{n}} \cdot \mathbf{x}' - \frac{\alpha}{2}(L - z')^2 \sec\theta\right], \tag{7.8}$$

$$\mathscr{L}_0 = n_0\left[z' - \frac{\alpha}{2}(L - z')^2\right]. \tag{7.9}$$

The spectral distribution is obtained by substituting Eqs. (7.5) and (7.6) into Eq. (3.18):

$$\mathscr{I}(\mathbf{x}, \omega) = ca\left(\frac{\mathscr{E}_0}{2R}\sin\gamma\right)^2\left(\frac{ka}{2}\right)^4 \lim_{T\to\infty}\frac{1}{2T}\,|\,F\,|^2, \tag{7.10}$$

where

$$\cos\gamma = \hat{\mathbf{n}}\cdot\hat{\boldsymbol{\chi}}. \tag{7.11}$$

We define ϵ_2 and ϵ_3 by

$$\epsilon_2(t) = V^{-1/2}\int_V d^3\chi'\, e^{-i(k\mathscr{L}_1 - k_0\mathscr{L}_0)}\epsilon_1(\mathbf{x}', t), \tag{7.12}$$

$$\epsilon_3(t) = V^{-1/2}\int_V d^3\chi'\, e^{-i(k\mathscr{L}_1 + k_0\mathscr{L}_0)}\epsilon_1(\mathbf{x}', t). \tag{7.13}$$

The system is stationary as ϵ_0 is independent of time. Thus, by an application of the Wiener–Khintchine Theorem [13], we obtain

$$\langle\mathscr{I}(\mathbf{x}, \omega)\rangle = \frac{c}{a}\left(\frac{\mathscr{E}_0}{R}\sin\gamma\right)^2\left(\frac{k}{4\pi}\right)^4 VW, \tag{7.14}$$

where

$$W = \int dt\, e^{-i(\omega-\omega_0)t}\langle\epsilon_2{}^*(t)\,\epsilon_2(0)\rangle + \int dt\, e^{-i(\omega+\omega_0)t}\langle\epsilon_3{}^*(t)\,\epsilon_3(0)\rangle. \tag{7.15}$$

Alternatively, defining the correlation function g by

$$g(\mathbf{x}', \mathbf{x}''; \,|\, t'' - t' \,|) = \langle\epsilon_1(\mathbf{x}', t')\,\epsilon_1(\mathbf{x}'', t'')\rangle, \tag{7.16}$$

we obtain

$$\begin{aligned} W = \frac{1}{V}\int_V d^3\chi' \int_V d^3\chi'' \int dt\, g(\mathbf{x}', \mathbf{x}''; \,|\, t\,|) \\ \times \{\cos[k(\mathscr{L}_1' - \mathscr{L}_1'') - k_0(\mathscr{L}_0' - \mathscr{L}_0'') - (\omega - \omega_0)\,t] \\ + \cos[k(\mathscr{L}_1' - \mathscr{L}_1'') + k_0(\mathscr{L}_0' - \mathscr{L}_0'') - (\omega + \omega_0)\,t]\}, \end{aligned} \tag{7.17}$$

where, in Eq. (7.17),

$$\mathscr{L}_1' = \mathscr{L}_1(\mathbf{x}'), \qquad \mathscr{L}_1'' = \mathscr{L}_1(\mathbf{x}''), \tag{7.18}$$

and similarly for $\mathscr{L}_0'$ and $\mathscr{L}_0''$. Both types of representation for W frequently appear in the literature on light scattering from homogeneous systems. A typical molecular collision frequency is on the order of $10^{11}\ \mathrm{sec}^{-1}$ and a typical light frequency is on the order of $10^{14}\ \mathrm{sec}^{-1}$. Hence, except at extraordinarily high temperatures or densities, the second term on the right of Eq. (7.15) and the second term in the curly brackets of Eq. (7.17) may be dropped.

8. Concluding Remarks

While we have outlined a viable method for constructing the spectral distribution of light scattered by fluctuations in an inhomogeneous fluid, this is just the first, and easiest, step towards expressing $\mathscr{I}(\mathbf{x}, \omega)$ in terms of the measurable properties of the fluid. In order to proceed further one must first determine $\epsilon_0(\mathbf{x})$ and/or $\mu_0(\mathbf{x})$ explicitly and then construct the required correlation functions by means of the appropriate transport equations. For the case of an inhomogeneous fluid under the influence of external fields, this requires a great deal more thermodynamic information than is needed for a homogeneous fluid.

From the elementary example considered in the last four sections we can conclude that the spatial transform (represented by ϵ_2 in the example) will no longer be simply a Fourier transform as in the homogeneous case, but rather will be some functional of ϵ_0. Consequently, the dependence of $\mathscr{I}$ on $\mathbf{x}$ (specifically on θ in the example) will no longer be the same as in the homogeneous case. Hopefully, this difference can be utilized to determine how close the critical point may be approached before gravitational effects become significant.

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