

## Statistical properties of radiation scattered by a fluid

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## Statistical properties of radiation scattered by a fluid†

**Abstract.** The statistical properties of light scattered by a fluid whose density fluctuations follow a normal distribution are examined.

Modern theory of optical coherence shows that the characterization of a stochastic electromagnetic field requires the knowledge of every-order correlation functions  $G^{(n)}$  of the field (see, for example, Glauber 1965, Mandel and Wolf 1965). If one is concerned with the field associated with linear incoherent scattering of monochromatic light by a fluid, the  $G^{(n)}$  can be connected with the higher-order correlation functions of the scattering medium (Bertolotti *et al.* 1967, Shen 1967). On this line we wish to recover these relations in a very direct way and to specialize them for a fluid whose density fluctuations follow a normal distribution. Photon-counting experiments, which, as well known, can be discussed in terms of the  $G^{(n)}$ , would thus provide a test for this hypothesis.

In the case of visible radiation a macroscopic form of the scattered electric field  $E_1$  can be given in terms of the variations of the dielectric constant  $\epsilon(\mathbf{r}, t)$  (see, for example, van Kampen 1967). The single-scattering electric field reads, at a point  $\mathbf{R}$  far from the scattering volume,

$$E_1(\mathbf{R}, t) = -\left(\frac{1}{4\pi R^2}\right) \exp\left\{-i\omega_0\left(t - \frac{R}{c}\right)\right\} \mathbf{k}_1 \times (\mathbf{k}_1 \times \mathbf{A}) \frac{d\epsilon}{d\rho} \rho_1\left(\mathbf{k}, t - \frac{R}{c}\right) \quad (1)$$

where  $\mathbf{k}_1 = (\omega_0/c)(\mathbf{R}/R)$ ,  $\mathbf{k} = \mathbf{k}_0 - \mathbf{k}_1$ , on the assumption of an incident plane wave  $\mathbf{A} \exp\{i(\mathbf{k}_0 \cdot \mathbf{r} - \omega_0 t)\}$ . Here  $\rho(\mathbf{r}, t) = \rho_0 + \rho_1(\mathbf{r}, t)$  is the number density, the derivative  $d\epsilon/d\rho$  is taken at the average density  $\rho_0$  of the medium supposed homogeneous and  $\rho_1(\mathbf{k}, t - R/c)$  represents the spatial Fourier transform of the density fluctuations, whose typical frequencies are assumed to be much smaller than  $\omega_0$ .

We note that equation (1) is consistent with an approximate expression of the scattered intensity which can be obtained on pure microscopic bases (Bullough and Hynne 1968). We refer to equation (14) of these authors, where the quantity  $d\epsilon/d\rho$  must be evaluated by means of an extension of the Lorentz-Lorenz equation involving every-order correlations of density fluctuations (see, for example, Bullough *et al.* 1968). Equation (14) is, however, approximate (about 10%) because it does not contain depolarization effects.

The correlation functions of the electric field are defined, if we assume the field to possess a specific polarization (which can be accomplished in practice by putting a polarization filter in front of the detector), as

$$G^{(n)}(x_1, x_2, \dots, x_{2n}) = \langle E^*(x_1) \dots E^*(x_n) E(x_{n+1}) \dots E(x_{2n}) \rangle \quad (2)$$

where the angular brackets stand for ensemble average,  $x_j = (\mathbf{r}_j, t_j)$  and  $E(\mathbf{r}, t)$  represents the classical 'analytic signal' of the electric field. In an actual photon-counting experiment the  $G^{(n)}$  can be evaluated using the special conditions  $\mathbf{r}_j = \mathbf{R}$  (with  $\mathbf{R}$  the mean position of the counter) and  $t_j = t_{2n+j-1}$  (which corresponds to assuming a detector of the broad band variety). In this case, one has immediately

$$G^{(n)}(t_1, t_2, \dots, t_n, t_n, \dots, t_2, t_1) = B^{2n} \left\langle \left| \rho_1\left(\mathbf{k}, t_1 - \frac{R}{c}\right) \right|^2 \dots \left| \rho_1\left(\mathbf{k}, t_n - \frac{R}{c}\right) \right|^2 \right\rangle. \quad (3)$$

Equation (3) furnishes the relation between the  $G^{(n)}$  and the  $2n$ th-order correlation functions of the medium.

An explicit expression for the higher-order density correlations would be now in order. While the exact solution of this problem is a formidable task, a suitable assumption may be made since the wavelength of the incident radiation is much greater than the mean

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intermolecular distance  $d$ . Then only Fourier components with  $k \leq 2k_0 \ll d^{-1}$  matter, and hence one may use a hydrodynamical description for all relevant quantities. To this end, we remember that the hypothesis of normality for the ensemble averages of significant quantities is usually made in the theory of isotropic turbulence (Batchelor 1951, Chandrasekhar 1951). Accordingly, the distribution function of a set of real stochastic variables  $y_1, y_2, \dots, y_m$  is termed as normal if it is the Fourier transform of the characteristic function

$$\Psi(\lambda_1, \lambda_2, \dots, \lambda_m) = \exp\left(-\frac{1}{2} \sum_{i,l} \langle y_i y_l \rangle \lambda_i \lambda_l\right). \quad (4)$$

As a property of normal distribution equation (4) yields, for example,

$$\langle y_1 y_2 y_3 y_4 \rangle = \langle y_1 y_2 \rangle \langle y_3 y_4 \rangle + \langle y_1 y_3 \rangle \langle y_2 y_4 \rangle + \langle y_1 y_4 \rangle \langle y_2 y_3 \rangle. \quad (5)$$

We extend by analogy this hypothesis to our case by assuming its validity for the (real) quantities  $y_i = \rho_1(\mathbf{r}_i, t_i)$ . Thus, by a suitable multiple Fourier transform of both sides of equation (5), we obtain

$$\begin{aligned} \langle \rho_1(\mathbf{k}, t_1) \rho_1(-\mathbf{k}, t_1) \rho_1(\mathbf{k}, t_2) \rho_1(-\mathbf{k}, t_2) \rangle &= \langle |\rho_1(\mathbf{k}, t_1)|^2 \rangle \langle |\rho_1(\mathbf{k}, t_2)|^2 \rangle \\ &+ \langle \rho_1(\mathbf{k}, t_1) \rho_1(-\mathbf{k}, t_2) \rangle \langle \rho_1(-\mathbf{k}, t_1) \rho_1(\mathbf{k}, t_2) \rangle \\ &+ \langle \rho_1(\mathbf{k}, t_1) \rho_1(\mathbf{k}, t_2) \rangle \langle \rho_1(-\mathbf{k}, t_1) \rho_1(-\mathbf{k}, t_2) \rangle. \end{aligned} \quad (6)$$

If the further assumption of a homogeneous scattering medium is made, it is immediately seen that the last term on the right-hand side of equation (6) vanishes. In this case equation (3) gives rise to the following factorization for the second-order correlation function:

$$G^{(2)}(t_1, t_2, t_2, t_1) = G^{(1)}(t_1, t_1) G^{(1)}(t_2, t_2) + G^{(1)}(t_1, t_2) G^{(1)}(t_2, t_1). \quad (7)$$

Otherwise a further term is present which takes into account the vector properties of the fluid. We conclude that the normal distribution hypothesis could be actually tested through a photon-counting experiment.

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