

FLUCTUATIONS IN DOUBLY SCATTERED LASER LIGHT

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Fluctuations in laser light, doubly scattered by brownian particles, were analysed by measuring the spectral noise power of the photodetector current. Scattering took place at two spatially separated systems of spherical particles. Analytic expressions for the field and intensity correlations are derived. The analytic expressions for the spectrum of the intensity fluctuations of the doubly scattered laser light demonstrate that the frequency dependence of the spectrum depends strongly on the geometry of the experimental arrangement. This is not the case for singly scattered light where in good approximation the spatial and temporal correlations can be separated analytically.

Our measurements show that the noise spectrum of the doubly scattered radiation may have the same frequency dependence as the spectrum of the singly scattered light. However, there are conditions where the frequency dependence of the noise of the doubly scattered light diverges markedly from that of the singly scattered light.

1. Introduction

Light beating spectroscopy of scattered laser light is a current technique used to investigate kinetic properties of the scattering medium^{1,2,3}). Most studies on laser light scattering concern only single scattering. There are, however, scattering systems that are likely to produce double or even multiple scattering. At present it is not quite clear whether light beating spectroscopy in the case of double (or multiple) scattering can be a useful method in the study of the scattering medium. To solve this problem, however, a further theoretical and experimental study into the statistical properties of doubly scattered light is required.

We have investigated doubly scattered light in a very simple experimental set-up (see fig. 1). Scattering took place in two spatially separated cells each filled with the same dilute suspension of polystyrene spheres. A laser beam was focussed in one cell. The scattered light from this first cell was scattered for the second time in the second cell. The concentration of the scattering particles was so low that in each cell single scattering of the radiation dominated. A photomultiplier tube received only the radiation coming from the second cell. The particles in the cells undergo a brownian motion and the fluctuations in the light scattered by such a

system of independent particles have been studied extensively¹⁻¹⁴). In sec. 2 we shall give the field and intensity correlation functions of singly scattered light, referring to the literature.

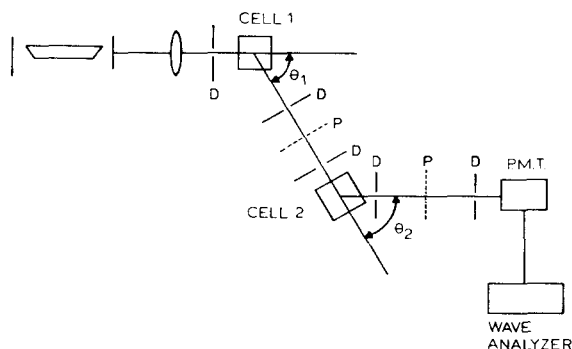


Fig. 1. Experimental set-up for the measurement of the noise spectral density of doubly scattered light. *D* are diaphragms and *P* are polarization analyzers. P.M.T. is the photomultiplier tube.

In the derivation of the correlation functions and spectral noise density of doubly scattered light, also given in sec. 2, we shall use certain assumptions that appeared to be satisfactory in the analysis of single scattering¹⁻³). The incident laser beam is represented as a monochromatic plane wave with a constant amplitude across the scattering volume. Noise in the incident laser beam is neglected. The brownian particles are supposed to be dipole oscillators with dimensions less than the wavelength of the laser light (Rayleigh scattering) and the number of scatterers is assumed to be constant. Only experimental geometries are considered so that the direction of polarization of the scattered waves is nearly equal to that of the incident beam.

In sec. 3 the theoretical and experimental results for the noise spectral density of doubly scattered laser light (homodyne detection) are compared. In sec. 4 we shall discuss our theory and experiments and shall compare them with the results obtained by others¹⁵⁻¹⁷).

2. Theory

2.1. Single scattering

Let a linearly polarized plane monochromatic wave be singly scattered by a system of independent particles. For the cross-correlation of the scattered field (with same polarization as incident field) Tartaglia and Chen¹¹) have derived

that:

$$\begin{aligned} \Gamma_1(\mathbf{R}_P, \mathbf{R}_Q; \tau) &\equiv \langle E_1^*(\mathbf{R}_P, t) E_1(\mathbf{R}_Q, t + \tau) \rangle \\ &= \langle E_1^*(\mathbf{R}_P, t) E_1(\mathbf{R}_Q, t) \rangle \langle E_1^*(\mathbf{R}_P, t) E_1(\mathbf{R}_P, t + \tau) \rangle / \langle I_1(\mathbf{R}_P, t) \rangle, \end{aligned} \quad (1)$$

where \mathbf{R}_P and \mathbf{R}_Q denote the position of P and Q respectively on the surface of a photodetector, $E_1(\mathbf{R}_P, t)$ is the electric field strength at point P at time t due to scattering by the particles and $I_1(\mathbf{R}_P, t)$ is the intensity defined by:

$$I_1(\mathbf{R}_P, t) = E_1^*(\mathbf{R}_P, t) E_1(\mathbf{R}_P, t). \quad (2)$$

The origin of the coordinate system is at the centre of the scattering cell. Note that in eq. (1) the spatial and the temporal correlations appear separately. For a system of N independent spherical brownian particles the temporal correlation function has been found to be^{2,7}:

$$\begin{aligned} \langle E_1^*(\mathbf{R}_P, t) E_1(\mathbf{R}_P, t + \tau) \rangle &= (N\mu |E_0|/R_0)^2 \exp(-i\omega_0\tau - D|\mathbf{k}_0 - \mathbf{k}_P|^2|\tau|), \end{aligned} \quad (3)$$

where E_0 is the amplitude of the incident wave, which has an angular frequency ω_0 and a wave vector in the medium \mathbf{k}_0 . The wave vector of the scattered field along \mathbf{R}_P is denoted by \mathbf{k}_P . Note that $k_0 = k_P = 2\pi n/\lambda$, where n is the index of refraction of the medium and λ is the wavelength in vacuum. The distance from the centre of the cell to the centre of the detector surface is R_0 , the plane detector surface is supposed to be normal to \mathbf{R}_0 . D is the diffusion constant, which for spherical particles at temperature T is¹⁸):

$$D = k_B T / 6\pi\eta\varrho. \quad (4)$$

Here k_B is Boltzmann's constant, η is the viscosity of the solvent and ϱ is the radius of the particle. In eq. (3) μ depends on wavelength and the polarizability of a scatterer¹⁹). The explicit expression of μ is of no interest here.

If in eq. (3) we are allowed to put:

$$|\mathbf{k}_0 - \mathbf{k}_P| \approx |\mathbf{k}_0 - \mathbf{k}_Q| \approx |\mathbf{k}_0 - \mathbf{k}_s|, \quad (5)$$

where \mathbf{k}_Q and \mathbf{k}_s are the wave vectors along the directions of \mathbf{R}_Q and \mathbf{R}_0 , respectively, the temporal coherence function in eq. (3) is independent of the position of the point P on the detector surface. Eq. (5) generally holds if the linear dimensions of the detector surface are much less than the distance between detector and scattering cell. We should note that eq. (1) is only correct if eq. (5) holds¹¹).

The spatial coherence function $\langle E_1^*(\mathbf{R}_P, t) E_1(\mathbf{R}_Q, t) \rangle$ depends on the geometry of the scattering medium^{2,3,11}) and in special cases expressions for it are given in the literature^{11,20}). We shall not discuss this spatial coherence function here as we shall deal with spatial coherence functions in the derivation of doubly scattered light.

According to eq. (3):

$$\langle I_1 \rangle = \langle I_1(\mathbf{R}_P, t) \rangle = \langle I_1(\mathbf{R}_Q, t) \rangle = (N\mu |E_0|/R_0)^2. \quad (6)$$

For the cross-correlation of the intensity of the single scattered field, defined by:

$$\Phi_1(\mathbf{R}_P, \mathbf{R}_Q; \tau) = \langle I_1(\mathbf{R}_P, t) I_1(\mathbf{R}_Q, t + \tau) \rangle, \quad (7)$$

Tartaglia and Chen derived¹¹):

$$\Phi_1(\mathbf{R}_P, \mathbf{R}_Q; \tau) = \langle I_1(\mathbf{R}_P, t) \rangle^2 (1 + |I_1'(\mathbf{R}_P, \mathbf{R}_Q; \tau)|^2) \quad (8)$$

assuming that $N \gg 1$. Eq. (8) generally holds for gaussian fields²¹). From eqs. (1), (3) and (6) it now follows that:

$$\begin{aligned} \Phi_1(\mathbf{R}_P, \mathbf{R}_Q; \tau) = \langle I_1 \rangle^2 \{ & 1 + \exp(-2D|\tau||\mathbf{k}_0 - \mathbf{k}_s|^2) \\ & \times |\langle E_1^*(\mathbf{R}_P, t) E_1(\mathbf{R}_Q, t) \rangle|^2 \}. \end{aligned} \quad (9)$$

2.2. Double scattering

In fig. 1 an outline is given of the experimental set-up and the relevant vectors and distances in the double scattering experiment can be found in fig. 2. The origin of the coordinate system is located at the centre O_1 of the first cell. The centre of the second scattering cell O_2 is at position \mathbf{R}_1 with respect to O_1 . The centre of the detector is at position \mathbf{R}_2 , which vector has O_2 as origin and is normal to the detector surface. The wave vector of the incident plane wave is denoted by \mathbf{k}_0 ; \mathbf{k}_1 and \mathbf{k}_2 are wave vectors along \mathbf{R}_1 and \mathbf{R}_2 , respectively. The first scattering angle θ_1 is the angle between \mathbf{k}_0 and \mathbf{k}_1 and the second scattering angle θ_2 is that between \mathbf{k}_1 and \mathbf{k}_2 . The position of particle a (or b, c, d) in the first cell with respect to O_1 is indicated by the vector \mathbf{r}_a (or $\mathbf{r}_b, \mathbf{r}_c, \mathbf{r}_d$) whereas s_α (or $s_\beta, s_\gamma, s_\delta$) gives the position of particle α (or β, γ, δ) in the second cell with respect to O_2 . The distance between a particle a in the first and a particle α in the second cell is denoted by $r_{a\alpha}$. P is a point at the detector surface. Its position with respect to O_1 is indicated by the vector \mathbf{R}_P whereas s_P gives the position of P with respect to O_2 . The distance between particle α belonging to the second cell and P is given by $r_{\alpha P}$.

is negligible with respect to λ in time intervals of length $\tau = R_1/c$, where c is the velocity of light in the medium. In the sequel we shall denote the position of a particle at time t by \mathbf{r} (or s) and the position of a particle at time $t + \tau$ by \mathbf{r}' (or s').

The field at point P of a wave scattered by particle a and subsequently by particle α is:

$$(\mu^2 E_0 / r_{a\alpha} r_{\alpha P}) \exp \{i (\mathbf{k}_0 \cdot \mathbf{r}_a + k_0 r_{a\alpha} + k_0 r_{\alpha P} - \omega_0 t)\}. \quad (13)$$

Fluctuations in the total field at P will be primarily due to interference of waves with different phases. Therefore small differences in wave amplitude can be neglected since the linear dimensions of the scattering volumes are supposed to be small with respect to their distance R_1 . So in the amplitude of the wave in eq. (13) we put:

$$1/r_{a\alpha} r_{\alpha P} = 1/R_1 R_2. \quad (14)$$

The total field at P at time t due to double scattering is now found to be:

$$E_2(\mathbf{R}_P, t) = (\mu^2 E_0 / R_1 R_2) \exp(-i\omega_0 t) \times \sum_{a=1}^{N_1} \sum_{\alpha=1}^{N_2} \exp \{i (\mathbf{k}_0 \cdot \mathbf{r}_a + k_0 r_{a\alpha} + k_0 r_{\alpha P})\}, \quad (15)$$

where N_1 and N_2 are the total number of scatterers in cells 1 and 2, respectively.

Defining the intensity of the field at P by:

$$I_2(\mathbf{R}_P, t) = E_2^*(\mathbf{R}_P, t) E_2(\mathbf{R}_P, t), \quad (16)$$

we obtain:

$$I_2(\mathbf{R}_P, t) = (\mu^2 |E_0|^2 / R_1 R_2)^2 \times \sum_{a,b}^{N_1} \sum_{\alpha,\beta}^{N_2} \exp [i \{ \mathbf{k}_0 \cdot (\mathbf{r}_b - \mathbf{r}_a) + k_0 (r_{b\beta} - r_{a\alpha} + r_{\beta P} - r_{\alpha P}) \}]. \quad (17)$$

The positions of the particles are randomly distributed over the scattering volumes. The dimensions of the scattering volumes are supposed to be much greater than the wavelength. In the averaging over the positions of the particles only terms in eq. (17) with indices $a = b$ and $\alpha = \beta$ are nonzero. Thus:

$$\langle I_2 \rangle = \langle I_2(\mathbf{R}_P, t) \rangle = (\mu^2 |E_0|^2 / R_1 R_2)^2 N_1 N_2. \quad (18)$$

For the cross-correlation function of the field at P and Q we obtain with the help of eq. (15):

$$\begin{aligned} I_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) \\ \equiv \langle E_2^*(\mathbf{R}_P, t) E_2(\mathbf{R}_Q, t + \tau) \rangle = (\mu^2 |E_0| / R_1 R_2)^2 \exp(-i\omega_0 \tau) \\ \times \left\langle \sum_{a,b}^{N_1} \sum_{\alpha,\beta}^{N_2} \exp[i\{\mathbf{k}_0 \cdot (\mathbf{r}'_b - \mathbf{r}_a) + k_0(r'_{b\beta} - r_{a\alpha} + r'_{\beta Q} - r_{\alpha P})\}] \right\rangle. \quad (19) \end{aligned}$$

And for the cross-correlation of the intensity at points P and Q we find:

$$\begin{aligned} \Phi_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) \equiv \langle I_2(\mathbf{R}_P, t) I_2(\mathbf{R}_Q, t + \tau) \rangle \\ = (\mu^2 |E_0| / R_1 R_2)^4 \left\langle \sum_{a,b,c,d}^{N_1} \sum_{\alpha,\beta,\gamma,\delta}^{N_2} \exp[i\{\mathbf{k}_0 \cdot (\mathbf{r}_b - \mathbf{r}_a + \mathbf{r}'_d - \mathbf{r}'_c) \right. \\ \left. + k_0(r_{b\beta} - r_{a\alpha} + r'_{d\delta} - r'_{c\gamma} + r_{\beta P} - r_{\alpha P} + r'_{\delta Q} - r'_{\gamma Q})\}] \right\rangle. \quad (20) \end{aligned}$$

In general the ensemble averaging in eqs. (19) and (20) does not lead to amenable expressions. Therefore we shall make the following approximations. First we suppose the dimensions of the second scattering volume to be much less than R_2 . So:

$$r_{\alpha P} = |\mathbf{s}_P - \mathbf{s}_\alpha| \simeq s_P - \mathbf{s}_\alpha \cdot \hat{\mathbf{s}}_P, \quad (21)$$

where $\hat{\mathbf{s}}_P$ is a unit vector in the direction \mathbf{s}_P . If we use the notation $\mathbf{k}_P = k_0 \hat{\mathbf{s}}_P$ and $\mathbf{k}_Q = k_0 \hat{\mathbf{s}}_Q$ we obtain with the help of eq. (21):

$$\begin{aligned} I_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) \\ = (\mu^2 |E_0| / R_1 R_2)^2 \exp\{-i\omega_0 \tau + ik_0(s_Q - s_P)\} \\ \times \left\langle \sum_{a,b}^{N_1} \sum_{\alpha,\beta}^{N_2} \exp[i\{\mathbf{k}_0 \cdot (\mathbf{r}'_b - \mathbf{r}_a) + k_0(r'_{b\beta} - r_{a\alpha}) - (\mathbf{k}_Q \cdot \mathbf{s}'_\beta - \mathbf{k}_P \cdot \mathbf{s}_\alpha)\}] \right\rangle, \quad (22) \end{aligned}$$

and

$$\begin{aligned} \Phi_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) \\ = (\mu^2 |E_0| / R_1 R_2)^4 \left\langle \sum_{a,b,c,d}^{N_1} \sum_{\alpha,\beta,\gamma,\delta}^{N_2} \exp[i\{\mathbf{k}_0 \cdot (\mathbf{r}_b - \mathbf{r}_a + \mathbf{r}'_d - \mathbf{r}'_c) \right. \\ \left. + k_0(r_{b\beta} - r_{a\alpha} + r'_{d\delta} - r'_{c\gamma}) - \mathbf{k}_P \cdot (\mathbf{s}_\beta - \mathbf{s}_\alpha) - \mathbf{k}_Q \cdot (\mathbf{s}'_\delta - \mathbf{s}'_\gamma)\}] \right\rangle. \quad (23) \end{aligned}$$

Next we distinguish two cases:

- (i) the cells are close together but the linear dimensions of the scattering volumes are less than R_1 by about a factor 10;
- (ii) the cells are far apart. Now the linear dimensions of the scattering volumes are supposed to be much smaller than R_1 . Thus:

$$r_{a\alpha} = |\mathbf{r}_\alpha - \mathbf{r}_a| \cong r_\alpha - \mathbf{r}_a \cdot \hat{\mathbf{r}}_\alpha. \quad (24)$$

We shall calculate the field and intensity correlations for both cases separately.

2.3. Case (i), cells close together

The ensemble averaging in eq. (22) over the random positions of the particles cannot be carried out separately over both scattering volumes. Because of the statistical independence of the positions of the particles only terms in eq. (22) with indices $a = b$ and $\alpha = \beta$ are nonzero. The displacements $\Delta \mathbf{r}_a$ and $\Delta \mathbf{s}_\alpha$ in a time-interval $\tau \cong t_c$, where t_c is the correlation time of the singly scattered field, are of the order of the magnitude of the wavelength λ of the laser light (see appendix). So we can use the following approximation:

$$\mathbf{r}'_{a\alpha} = |\mathbf{R}_1 + \mathbf{s}'_\alpha - \mathbf{r}'_a| = r_{a\alpha} + (\Delta \mathbf{s}_\alpha - \Delta \mathbf{r}_a) \cdot \hat{\mathbf{r}}_{a\alpha}, \quad (25)$$

where $\hat{\mathbf{r}}_{a\alpha}$ is a unit vector along $r_{a\alpha}$ in the direction of particle α . Eq. (22) now becomes:

$$\begin{aligned} & \Gamma_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) \\ &= N_1 N_2 (\mu^2 |E_0| / R_1 R_2)^2 \exp \{ -i\omega_0 \tau + ik_0 (s_Q - s_P) \} \\ & \quad \times \langle \exp [i \{ (\mathbf{k}_0 - \mathbf{k}_{a\alpha}) \cdot \Delta \mathbf{r}_a + (\mathbf{k}_{a\alpha} - \mathbf{k}_Q) \cdot \Delta \mathbf{s}_\alpha - (\mathbf{k}_Q - \mathbf{k}_P) \cdot \mathbf{s}_\alpha \}] \rangle, \quad (26) \end{aligned}$$

where $\mathbf{k}_{a\alpha} = k_0 \hat{\mathbf{r}}_{a\alpha}$. Note that in eq. (26) we must average over particle positions at t and over the displacement of a particle in a time-interval of length τ . As is well known from the theory of brownian motion the probability of occurrence of a particle at $\mathbf{r} + \Delta \mathbf{r}$ at time $t + \tau$ when it was at \mathbf{r} at time t is given by²²:

$$P(\mathbf{r} + \Delta \mathbf{r} | \mathbf{r}) = (4\pi D\tau)^{-3/2} \exp \{ -(\Delta \mathbf{r})^2 / 4D\tau \}. \quad (27)$$

Carrying out the ensemble averaging over $\Delta \mathbf{r}_a$ and $\Delta \mathbf{s}_\alpha$ in eq. (26) with the help of eq. (27) we obtain:

$$\begin{aligned} & \Gamma_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) \\ &= N_1 N_2 (\mu^2 |E_0| / R_1 R_2)^2 \exp \{ -i\omega_0 \tau + ik_0 (s_Q - s_P) \} \\ & \quad \times \langle \exp \{ -D|\tau| (|\mathbf{k}_0 - \mathbf{k}_{a\alpha}|^2 + |\mathbf{k}_{a\alpha} - \mathbf{k}_Q|^2) + i(\mathbf{k}_P - \mathbf{k}_Q) \cdot \mathbf{s}_\alpha \} \rangle. \quad (28) \end{aligned}$$

In eq. (28) ensemble averaging still has to be performed over the statistical positions of the scatterers a and α . Note that even if:

$$|\mathbf{k}_{a\alpha} - \mathbf{k}_Q| \simeq |\mathbf{k}_{a\alpha} - \mathbf{k}_2|, \quad (29)$$

the spatial and the temporal correlations in $\Gamma_2(\mathbf{R}_P, \mathbf{R}_Q; \tau)$ cannot be separated (cross-spectrally impure²³). But if we are allowed to assume that the factor between ensemble brackets in eq. (28) is about equal to:

$$\begin{aligned} \langle \dots \rangle &\simeq \langle \exp \{i(\mathbf{k}_P - \mathbf{k}_Q) \cdot \mathbf{s}_\alpha\} \rangle \\ &\times \exp \{-D|\tau|(|\mathbf{k}_0 - \mathbf{k}_1|^2 + |\mathbf{k}_1 - \mathbf{k}_2|^2)\}, \end{aligned} \quad (30)$$

separation of temporal from spatial correlation is obtained:

$$\begin{aligned} \Gamma_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) &= N_1 N_2 (\mu^2 |E_0| / R_1 R_2)^2 \\ &\times \exp \{ik_0(s_Q - s_P)\} \langle \exp \{i(\mathbf{k}_P - \mathbf{k}_Q) \cdot \mathbf{s}_\alpha\} \rangle \\ &\times \exp \{-i\omega_0\tau - D|\tau|(|\mathbf{k}_0 - \mathbf{k}_1|^2 + |\mathbf{k}_1 - \mathbf{k}_2|^2)\}. \end{aligned} \quad (31)$$

Whether eq. (30) holds or not depends on the dimensions of both scattering volumes. But if $(|\mathbf{k}_0 - \mathbf{k}_{a\alpha}|^2 + |\mathbf{k}_{a\alpha} - \mathbf{k}_Q|^2)$ can be approximated by $(|\mathbf{k}_0 - \mathbf{k}_1|^2 + |\mathbf{k}_1 - \mathbf{k}_2|^2)$ within 1% for all values of the index $a\alpha$, eq. (30) can be accepted.

Next we shall calculate the cross-correlation of the light intensity. Only terms in eq. (23) with indices:

$$a = b, \quad c = d \quad \text{and} \quad \alpha = \beta, \quad \gamma = \delta \quad (32a)$$

or

$$a = d, \quad b = c, \quad a \neq b \quad \text{and} \quad \alpha = \delta, \quad \beta = \gamma, \quad \alpha \neq \beta \quad (32b)$$

are nonzero after ensemble averaging. Therefore we can write:

$$\begin{aligned} \Phi_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) &= (\mu^2 |E_0| / R_1 R_2)^4 \left[N_1^2 N_2^2 \right. \\ &+ \sum_{\substack{a, b \\ a \neq b}}^{N_1} \sum_{\substack{\alpha, \beta \\ \alpha \neq \beta}}^{N_2} \langle \exp [i \{ (\mathbf{k}_0 - \mathbf{k}_{a\alpha}) \cdot \Delta \mathbf{r}_a - (\mathbf{k}_0 - \mathbf{k}_{b\beta}) \cdot \Delta \mathbf{r}_b \\ &+ (\mathbf{k}_{a\alpha} - \mathbf{k}_Q) \cdot \Delta \mathbf{s}_\alpha - (\mathbf{k}_{b\beta} - \mathbf{k}_Q) \cdot \Delta \mathbf{s}_\beta \\ &+ (\mathbf{k}_P - \mathbf{k}_Q) \cdot (\mathbf{s}_\alpha - \mathbf{s}_\beta) \}] \rangle \left. \right]. \end{aligned} \quad (33)$$

In the derivation of eq. (33) from eq. (23) we have used eq. (25). Working out the averaging over $\Delta \mathbf{r}_a$, $\Delta \mathbf{r}_b$, $\Delta \mathbf{s}_\alpha$ and $\Delta \mathbf{s}_\beta$ and assuming that $N_1 \gg 1$ and $N_2 \gg 1$, we

obtain:

$$\begin{aligned}\bar{\Phi}_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) &= (N_1 N_2)^2 (\mu^2 |E_0| / R_1 R_2)^4 \\ &\times [1 + |\langle \exp \{-D|\tau| (|\mathbf{k}_0 - \mathbf{k}_{a\lambda}|^2 + |\mathbf{k}_{a\lambda} - \mathbf{k}_Q|^2) \\ &+ i(\mathbf{k}_P - \mathbf{k}_Q) \cdot \mathbf{s}_\lambda \rangle \rangle|^2].\end{aligned}\quad (34)$$

In eq. (34) the ensemble averaging has to be taken over the positions of one particle a and one particle λ . Spatial and temporal correlations in the intensity at the detector surface appear to be connected. However, if eq. (30) holds, we find:

$$\begin{aligned}\bar{\Phi}_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) &= (N_1 N_2)^2 (\mu^2 |E_0| / R_1 R_2)^2 \langle \exp \{i(\mathbf{k}_P - \mathbf{k}_Q) \cdot \mathbf{s}_\lambda\} \rangle^2 \\ &\times [1 + \exp \{-2D|\tau| (|\mathbf{k}_0 - \mathbf{k}_1|^2 + |\mathbf{k}_1 - \mathbf{k}_2|^2)\}].\end{aligned}\quad (35)$$

By comparison of eq. (34) with eq. (28) or by comparison of eq. (35) with eq. (31) it can be seen that:

$$\bar{\Phi}_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) = \langle I_2 \rangle [1 + |I_2(\mathbf{R}_P, \mathbf{R}_Q; \tau)|^2].\quad (36)$$

We note that for singly scattered light an equation equivalent to eq. (36) holds.

2.4. Case (ii), cells far apart

Noting that $r_a \ll r_\lambda$ we find in addition to eq. (24):

$$r'_{a\lambda} \simeq r'_\lambda - \mathbf{r}'_a \cdot \hat{\mathbf{r}}'_\lambda \simeq r'_\lambda - \frac{\mathbf{r}'_a \cdot (\mathbf{r}_\lambda + \Delta \mathbf{s}_\lambda)}{r_\lambda + \Delta \mathbf{s}_\lambda \cdot \hat{\mathbf{r}}'_\lambda} \simeq r'_\lambda - \mathbf{r}'_a \cdot \hat{\mathbf{r}}'_\lambda, \quad (37)$$

where

$$r'_\lambda \simeq r_\lambda + \Delta \mathbf{s}_\lambda \cdot \hat{\mathbf{r}}'_\lambda.$$

By the use of eqs. (24) and (37) we can write for the cross-correlation of the optical field given in eq. (22):

$$\begin{aligned}I_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) &= (\mu^2 |E_0| / R_1 R_2)^2 \exp \{-i\omega_0 \tau + ik_0(s_Q - s_P)\} \\ &\times \left\langle \sum_{a,b}^{N_1} \sum_{\lambda,\beta}^{N_2} \exp [i \{(\mathbf{k}_0 - \mathbf{k}_\beta) \cdot (\mathbf{r}_b + \Delta \mathbf{r}_b) - (\mathbf{k}_0 - \mathbf{k}_\lambda) \cdot \mathbf{r}_a \right. \\ &\left. + k_0(r_\beta - r_\lambda) + (\mathbf{k}_\beta - \mathbf{k}_Q) \cdot \Delta \mathbf{s}_\beta - (\mathbf{k}_Q \cdot s_\beta - \mathbf{k}_P \cdot s_\lambda)\} \right] \right\rangle, \quad (38)\end{aligned}$$

where $k_\alpha = k_0 \hat{r}_\alpha$ and $k_\beta = k_0 \hat{r}_\beta$. Only terms with indices $b = a$ and $\alpha = \beta$ in eq. (38) give nonzero contributions. After the averaging over the displacements $\Delta \mathbf{r}_a$ and $\Delta \mathbf{s}_\beta$ and over \mathbf{r}_a , which is homogeneously distributed over the first scattering volume, eq. (38) yields:

$$\begin{aligned} I_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) &= N_1 N_2 (\mu^2 |E_0| / R_1 R_2)^2 \exp \{-i\omega_0 \tau + ik_0(s_Q - s_P)\} \\ &\times \langle \exp \{-D|\tau| (|\mathbf{k}_0 - \mathbf{k}_\alpha|^2 + |\mathbf{k}_\alpha - \mathbf{k}_Q|^2) + i(\mathbf{k}_P - \mathbf{k}_Q) \cdot \mathbf{s}_\alpha\} \rangle. \end{aligned} \quad (39)$$

If the linear dimensions of the second scattering volume are (much) smaller than R_1 and R_2 and if the linear dimension of the detector surface is small compared to R_2 we have:

$$|\mathbf{k}_0 - \mathbf{k}_\alpha|^2 + |\mathbf{k}_\alpha - \mathbf{k}_Q|^2 \simeq |\mathbf{k}_0 - \mathbf{k}_1|^2 + |\mathbf{k}_1 - \mathbf{k}_2|^2. \quad (40)$$

So then we find the final result:

$$\begin{aligned} I_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) &= N_1 N_2 (\mu^2 |E_0| / R_1 R_2)^2 \\ &\times \exp \{ik_0(s_Q - s_P)\} \langle \exp \{i(\mathbf{k}_P - \mathbf{k}_Q) \cdot \mathbf{s}_\alpha\} \rangle \\ &\times \exp \{-i\omega_0 \tau - D|\tau| (|\mathbf{k}_0 - \mathbf{k}_1|^2 + |\mathbf{k}_1 - \mathbf{k}_2|^2)\}. \end{aligned} \quad (41)$$

Note that eq. (41) equals eq. (31).

With the help of eqs. (23), (24) and (37) we can write for the cross-correlation function of the intensity:

$$\begin{aligned} \Phi_2(\mathbf{R}_P, \mathbf{R}_Q; \tau) &= (\mu^2 |E_0| / R_1 R_2)^4 \left\langle \sum_{a,b,c,d}^{N_1} \sum_{\alpha,\beta,\gamma,\delta}^{N_2} \exp [i \{k_0 \cdot (\mathbf{r}_b - \mathbf{r}_a + \mathbf{r}'_d - \mathbf{r}'_c) \right. \\ &\quad + k_0 (r_\beta - r_\alpha + r'_\delta - r'_\gamma) - k_0 (\mathbf{r}_b \cdot \hat{r}_\beta - \mathbf{r}_a \cdot \hat{r}_\alpha + \mathbf{r}'_d \cdot \hat{r}_\delta - \mathbf{r}'_c \cdot \hat{r}_\gamma) \\ &\quad \left. - \mathbf{k}_P \cdot (\mathbf{s}_\beta - \mathbf{s}_\alpha) - \mathbf{k}_Q \cdot (\mathbf{s}'_\delta - \mathbf{s}'_\gamma)\} \right] \rangle. \end{aligned} \quad (42)$$

Carrying out the ensemble averaging in eq. (42) we deal only with terms with indices:

$$\begin{aligned} (1) \quad a = b, \quad c = d, \quad (I) \quad \alpha = \beta, \quad \gamma = \delta, \\ (2) \quad a = d, \quad b = c, \quad a \neq b, \quad (II) \quad \alpha = \delta, \quad \beta = \gamma, \quad \alpha \neq \beta. \end{aligned} \quad (43)$$

Other terms are zero in a trivial way. Combination of the cases (1) and (2) with the cases (I) and (II) gives four cases in all. If we neglect the amplitude factor in eq. (42) we find after some arithmetic:

$$\begin{aligned}
 (1I) &= N_1^2 N_2^2, \\
 (2I) &= \left\langle \sum_{a,b}^{N_1} \sum_{\alpha,\gamma}^{N_2} \exp [i \{ (\mathbf{k}_0 - \mathbf{k}_\gamma) \cdot (\Delta \mathbf{r}_a - \Delta \mathbf{r}_b) + (\mathbf{k}_\alpha - \mathbf{k}_\gamma) \cdot (\mathbf{r}_a - \mathbf{r}_b) \}] \right\rangle \\
 &= N_1 (N_1 - 1) N_2^2 \langle \exp \{i (\mathbf{k}_\alpha - \mathbf{k}_\gamma) \cdot (\mathbf{r}_a - \mathbf{r}_b)\} \exp (-2D |\tau| |\mathbf{k}_0 - \mathbf{k}_\gamma|^2) \rangle, \\
 (1II) &= \left\langle \sum_{a,c}^{N_1} \sum_{\alpha,\beta}^{N_2} \exp [i \{ (\mathbf{k}_\alpha - \mathbf{k}_Q) \cdot \Delta \mathbf{s}_\alpha - (\mathbf{k}_\beta - \mathbf{k}_Q) \cdot \Delta \mathbf{s}_\beta \right. \\
 &\quad \left. + (\mathbf{k}_\alpha - \mathbf{k}_\beta) \cdot (\mathbf{r}_a - \mathbf{r}_c) - (\mathbf{k}_\alpha - \mathbf{k}_\beta) \cdot \Delta \mathbf{r}_c + (\mathbf{k}_P - \mathbf{k}_Q) \cdot (\mathbf{s}_\alpha - \mathbf{s}_\beta) \}] \right\rangle \\
 &\text{(The term } (\mathbf{k}_\alpha - \mathbf{k}_\beta) \cdot \Delta \mathbf{r}_c \text{ can be neglected)} \\
 &= N_1^2 N_2 (N_2 - 1) \langle \exp \{i (\mathbf{k}_\alpha - \mathbf{k}_\beta) \cdot (\mathbf{r}_a - \mathbf{r}_b) + i (\mathbf{k}_P - \mathbf{k}_Q) \cdot (\mathbf{s}_\alpha - \mathbf{s}_\beta)\} \\
 &\quad \times \exp \{-D |\tau| (|\mathbf{k}_\alpha - \mathbf{k}_Q|^2 + |\mathbf{k}_\beta - \mathbf{k}_Q|^2)\} \rangle, \\
 (2II) &= \left\langle \sum_{a,b}^{N_1} \sum_{\alpha,\beta}^{N_2} \exp [i \{ (\mathbf{k}_0 - \mathbf{k}_\alpha) \cdot \Delta \mathbf{r}_a - (\mathbf{k}_0 - \mathbf{k}_\beta) \cdot \Delta \mathbf{r}_b \right. \\
 &\quad \left. + (\mathbf{k}_\alpha - \mathbf{k}_Q) \cdot \Delta \mathbf{s}_\alpha - (\mathbf{k}_\beta - \mathbf{k}_Q) \cdot \Delta \mathbf{s}_\beta + (\mathbf{k}_P - \mathbf{k}_Q) \cdot (\mathbf{s}_\alpha - \mathbf{s}_\beta) \}] \right\rangle \\
 &= N_1 (N_1 - 1) N_2 (N_2 - 1) \langle \exp \{i (\mathbf{k}_P - \mathbf{k}_Q) \cdot (\mathbf{s}_\alpha - \mathbf{s}_\beta)\} \\
 &\quad \times \exp \{-D |\tau| (|\mathbf{k}_0 - \mathbf{k}_\alpha|^2 + |\mathbf{k}_0 - \mathbf{k}_\beta|^2 \\
 &\quad + |\mathbf{k}_\alpha - \mathbf{k}_Q|^2 + |\mathbf{k}_\beta - \mathbf{k}_Q|^2)\} \rangle. \tag{44}
 \end{aligned}$$

If we assume that $N_1, N_2 \gg 1$ as before and if within 1% the next equations hold:

$$\begin{aligned}
 |\mathbf{k}_0 - \mathbf{k}_\gamma|^2 &\simeq |\mathbf{k}_0 - \mathbf{k}_1|^2, \\
 |\mathbf{k}_\alpha - \mathbf{k}_Q|^2 + |\mathbf{k}_\beta - \mathbf{k}_Q|^2 &\simeq 2 |\mathbf{k}_1 - \mathbf{k}_2|^2,
 \end{aligned} \tag{45}$$

then we can give the final result in the following form:

$$\begin{aligned}
 \Phi_2 (\mathbf{R}_P, \mathbf{R}_Q; \tau) \\
 &= N_1^2 N_2^2 (\mu^2 |E_0| / R_1 R_2)^4 \\
 &\quad \times [1 + \langle \exp \{i (\mathbf{k}_\alpha - \mathbf{k}_\gamma) \cdot (\mathbf{r}_a - \mathbf{r}_b)\} \rangle \exp (-2D |\tau| |\mathbf{k}_0 - \mathbf{k}_1|^2)]
 \end{aligned}$$

$$\begin{aligned}
& + \langle \exp \{i (\mathbf{k}_\alpha - \mathbf{k}_\beta) \cdot (\mathbf{r}_a - \mathbf{r}_b) + i (\mathbf{k}_\mathbf{p} - \mathbf{k}_\mathbf{Q}) \cdot (\mathbf{s}_\alpha - \mathbf{s}_\beta)\} \rangle \\
& \times \exp (-2D |\tau| |\mathbf{k}_1 - \mathbf{k}_2|^2) + \langle \exp \{i (\mathbf{k}_\mathbf{p} - \mathbf{k}_\mathbf{Q}) \cdot (\mathbf{s}_\alpha - \mathbf{s}_\beta)\} \rangle \\
& \times \exp \{-2D |\tau| (|\mathbf{k}_0 - \mathbf{k}_1|^2 + |\mathbf{k}_1 - \mathbf{k}_2|^2)\}.
\end{aligned} \tag{46}$$

Inspection of eq. (46) shows that the total expression cannot be written as the product of a factor that describes only the spatial correlations and one that describes only the temporal correlations.

2.5. Spectral noise density of the photodetector current

The spectral densities of the optical field and of the intensity fluctuations can easily be derived from the correlation function by the use of the Wiener-Khintchine theorem^{1-3,23}). The spectral noise density of the fluctuations in the photodetector current depends on the instantaneous light intensity integrated over the detector surface with area A . Thus:

$$\begin{aligned}
S(f) &= 2\eta^2 e^2 M^2 \int_{-\infty}^{\infty} d\tau \int_A d\mathbf{R}_\mathbf{p} \int_A d\mathbf{R}_\mathbf{Q} \{ \Phi(\mathbf{R}_\mathbf{p}, \mathbf{R}_\mathbf{Q}; \tau) - \langle I \rangle^2 \} \\
&\times \exp(i2\pi f\tau) + S_0,
\end{aligned} \tag{47}$$

where η is the detection efficiency, e is the electronic charge and M is the mean multiplication factor of the detector. The term S_0 is due to shot noise and fluctuations in the multiplication factor and is supposed to be independent of the frequency f . The mean detector current is:

$$\langle i \rangle = \eta e M A \langle I \rangle. \tag{48}$$

According to eq. (9) the excess noise spectrum in case of single scattering is then found to be a Lorentz spectrum:

$$S_1(f) - S_0 = 4\langle i \rangle^2 \frac{F_1(A)}{A^2} \frac{2D |\mathbf{k}_0 - \mathbf{k}_s|^2}{(2\pi f)^2 + (2D |\mathbf{k}_0 - \mathbf{k}_s|)^2}, \tag{49}$$

where $F_1(A)$ is the spatial coherence factor defined by:

$$F_1(A) = \int_A d\mathbf{R}_\mathbf{p} \int_A d\mathbf{R}_\mathbf{Q} |\langle E_1^*(\mathbf{R}_\mathbf{p}, t) E_1(\mathbf{R}_\mathbf{Q}, t) \rangle|^2. \tag{50}$$

The bandwidth of the excess noise spectrum, defined as the frequency at half-height, is:

$$f_{\frac{1}{2}}(\theta) = D |\mathbf{k}_0 - \mathbf{k}_s|^2 / \pi = 4Dk_0^2 \sin^2 \frac{1}{2}\theta / \pi, \tag{51}$$

where θ is the angle between \mathbf{k}_0 and \mathbf{k}_s .

For the noise spectrum in case of double scattering we distinguish the two cases mentioned before.

Case (i), cells close together. From eq. (35) it follows that:

$$S_2(f) - S_0 = 4\langle i_2 \rangle^2 \frac{F_2(A)}{A^2} \frac{2D(|\mathbf{k}_0 - \mathbf{k}_1|^2 + |\mathbf{k}_1 - \mathbf{k}_2|^2)}{(2\pi f)^2 + \{2D(|\mathbf{k}_0 - \mathbf{k}_1|^2 + |\mathbf{k}_1 - \mathbf{k}_2|^2)\}^2}, \quad (52)$$

where

$$F_2(A) = \int_A d\mathbf{R}_P \int_A d\mathbf{R}_Q |\langle \exp \{i(\mathbf{k}_P - \mathbf{k}_Q) \cdot \mathbf{s}_\alpha\} \rangle|^2. \quad (53)$$

As we see in eq. (52) the double scattering spectrum in excess to S_0 has a lorentzian shape with bandwidth:

$$f_{\frac{1}{2}}(\theta_1, \theta_2) = 4Dk_0^2 (\sin^2 \frac{1}{2}\theta_1 + \sin^2 \frac{1}{2}\theta_2)/\pi = f_{\frac{1}{2}}(\theta_1) + f_{\frac{1}{2}}(\theta_2). \quad (54)$$

Case (ii), cells far apart. Eqs. (46) and (47) yield:

$$\begin{aligned} S_2(f) - S_0 = 4\langle i_2 \rangle^2 \left[B_1 \frac{2D|\mathbf{k}_0 - \mathbf{k}_1|^2}{(2\pi f)^2 + (2D|\mathbf{k}_0 - \mathbf{k}_1|^2)^2} \right. \\ + B_2 \frac{2D|\mathbf{k}_1 - \mathbf{k}_2|^2}{(2\pi f)^2 + (2D|\mathbf{k}_1 - \mathbf{k}_2|^2)^2} \\ \left. + B_3 \frac{2D(|\mathbf{k}_0 - \mathbf{k}_1|^2 + |\mathbf{k}_1 - \mathbf{k}_2|^2)}{(2\pi f)^2 + \{2D(|\mathbf{k}_0 - \mathbf{k}_1|^2 + |\mathbf{k}_1 - \mathbf{k}_2|^2)\}^2} \right], \quad (55) \end{aligned}$$

where

$$\begin{aligned} B_1 &= \langle \exp \{i(\mathbf{k}_\alpha - \mathbf{k}_\gamma) \cdot (\mathbf{r}_a - \mathbf{r}_b)\} \rangle, \\ B_2 &= (1/A^2) \int_A d\mathbf{R}_P \int_A d\mathbf{R}_Q \langle \exp \{i(\mathbf{k}_\alpha - \mathbf{k}_\beta) \cdot (\mathbf{r}_a - \mathbf{r}_b) \\ &\quad + i(\mathbf{k}_P - \mathbf{k}_Q) \cdot (\mathbf{s}_\alpha - \mathbf{s}_\beta)\} \rangle, \\ B_3 &= (1/A^2) \int_A d\mathbf{R}_P \int_A d\mathbf{R}_Q \langle \exp \{i(\mathbf{k}_P - \mathbf{k}_Q) \cdot (\mathbf{s}_\alpha - \mathbf{s}_\beta)\} \rangle. \quad (56) \end{aligned}$$

The excess noise spectrum consists now of three terms, each of which is the product of a spatial coherence factor and a Lorentz spectrum. Note that $B_3 = F_2(A)/A^2$.

3. Experimental set-up and results

In the double scattering experiment we used two lasers: a three mode HeNe laser (Spectra Physics model 120) with wavelength $\lambda = 632.8$ nm and a power of 5 mW and a single mode Kr-ion laser (Spectra Physics model 165) with wavelength $\lambda = 647.1$ nm and a power adjustable between 40 and 100 mW. The scattering medium consisted of Latex spheres suspended in water. In every scattering experiment both cells were filled with the same suspension. We found no difference in the interpretation of the experimental results when the spheres had a diameter of 101 nm or 332 nm. So we restrict ourselves to a report of the measurements where we used spheres with diameter 332 nm. We note that the particle size was of the same order of magnitude as the wavelength of the light. So the criterion for Rayleigh scattering $2\varrho \ll \lambda$ was not satisfied. Nevertheless the frequency dependence of the single scattering spectrum is still given by eq. (49)^{2,8)}.

Between the two cells and between the second cell and the detector diaphragms were placed (see fig. 1) in order to limit the solid angles of the light beams. We checked carefully that only light coming from the second cell was seen by the photodetector. Polarization analyzers, inserted in the light beams with axes parallel to the polarization of the incident laser light, did not change the frequency dependence of the measured noise spectra. The light signal after double scattering was always very weak. Normally detector currents were about $0.5 \mu\text{A}$. In order to enhance the detection efficiency of the photomultiplier (EMI 9558A) we stuck a small prism with immersion oil on the window of the detector²⁴⁾. When the two cells were close together we reduced the light losses caused by reflections by sticking the two cells on either side of a glass plate. The spectral noise density of the photocurrent was measured by means of a wave analyzer (Bruel & Kjaer type 2114).

We do not know the concentration of the particles in the suspensions we have used. In order to be sure that in each cell only single scattering occurred we measured the homodyne spectrum of the light scattered by the first cell. The experimental results of two of these measurements can be found in fig. 3 where the experimental data (the dots) are compared with Lorentz spectra (full curves). Experimentally we found the bandwidths: $f_{\frac{1}{2}}(\theta_1 = 60^\circ) = 86$ Hz and $f_{\frac{1}{2}}(\theta_1 = 90^\circ) = 165$ Hz. According to eq. (51) the theory predicts the bandwidths: $f_{\frac{1}{2}}(\theta_1 = 60^\circ) = 85$ Hz and $f_{\frac{1}{2}}(\theta_1 = 90^\circ) = 169$ Hz for particles with diameter $2\varrho = 332$ nm at a wavelength of $\lambda = 647.1$ nm and a temperature $T = 28^\circ\text{C}$. As the error in the experimental determination of the bandwidths in all our experiments was about 4% and the experimental data corresponded to Lorentz spectra over almost five decades we may assume that in spite of possible high particle concentration only single scattering prevails in each cell.

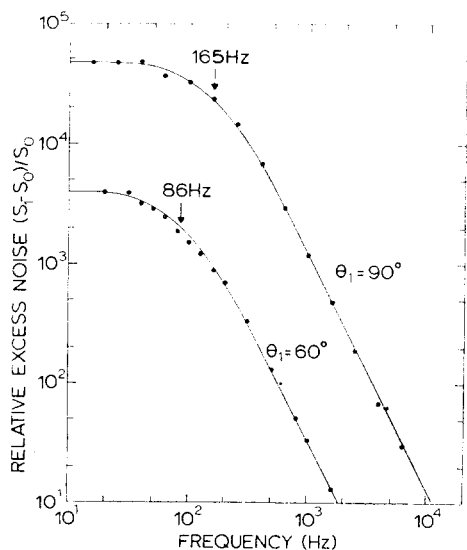


Fig. 3

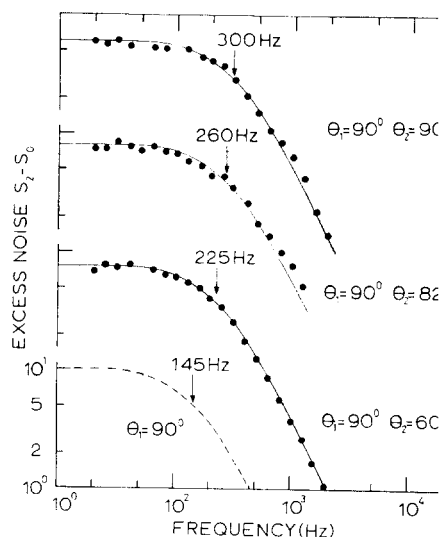


Fig. 4

Fig. 3. Relative excess noise $(S_1 - S_0)/S_0$ versus frequency of singly scattered light at an $\theta_1 = 90^\circ$ and $\theta_2 = 60^\circ$. The dots are the experimental results, the full curves are Lorentz spectra and the arrows indicate the bandwidth of the spectra.

Fig. 4. Double logarithmic plot of $S_2 - S_0$ (in arbitrary units) of doubly scattered light versus frequency. The distance between the scattering cells was 1.5×10^{-2} m. The first scattering angle was $\theta_1 = 90^\circ$ and the second scattering angles were $\theta_2 = 90^\circ, 82^\circ, 60^\circ$. The experimental results (the dots) are compared by Lorentz spectra (full curves). The dashed curve gives the frequency dependence of the noise in singly scattered light at a scattering angle $\theta_1 = 90^\circ$.

Case (i), cells close together. Characteristic values for the distances in this experiment were: $R_1 = 1.5 \times 10^{-2}$ m and $R_2 = 4.2 \times 10^{-1}$ m. The effective diameter of the photodetector was 5×10^{-3} m. If we represent the scattering volumes approximately by cuboids, the first scattering volume has the dimensions: 1×10 by 1×10^{-4} by 2×10^{-3} m and the second scattering volume had the dimensions 2×10^{-3} by 2×10^{-3} by 2×10^{-3} m.

Fig. 4 gives three excess noise spectra (the dots) measured on doubly scattered He-Ne laser light at particles with diameter $2\phi = 332$ nm and at a temperature $T = 21.5^\circ\text{C}$. Lorentz spectra could be fitted well to the experimental points shown by the full curves. The first scattering angle was in all cases $\theta_1 = 90^\circ$ and the second scattering angle was varied as indicated in the figure. The dashed curve shows the theoretical frequency dependence of singly scattered light at a scattering angle $\theta_1 = 90^\circ$. The bandwidths of the double scattering spectra (indicated by the arrows) depart markedly from the bandwidth of the single scattering spectrum. We compare the experimentally obtained values for the bandwidths of the

double scattering spectra with the bandwidths calculated from eq. (54) (within the brackets):

$$f_{\frac{1}{2}}(\theta_1 = 90^\circ, \theta_2 = 90^\circ) = 300 \text{ Hz (290 Hz)},$$

$$f_{\frac{1}{2}}(\theta_1 = 90^\circ, \theta_2 = 82^\circ) = 260 \text{ Hz (270 Hz) and}$$

$$f_{\frac{1}{2}}(\theta_1 = 90^\circ, \theta_2 = 60^\circ) = 225 \text{ Hz (218 Hz)}.$$

Within the experimental error, which was about 4%, agreement between experimentally determined and theoretically calculated bandwidths is found.

Case (ii), cells far apart. Characteristic values of the distances in the experimental set-up were now: $R_1 = 5.0 \times 10^{-1} \text{ m}$ and $R_2 = 3.0 \times 10^{-1} \text{ m}$. The effective diameter of the detector was $5 \times 10^{-2} \text{ m}$. The first scattering volume represented approximately as a cuboid, had the dimensions 1×10^{-4} by 1×10^{-4} by $1 \times 10^{-3} \text{ m}$ and the dimensions of the second scattering volume were 1×10^{-2} by 1×10^{-2} by $1 \times 10^{-2} \text{ m}$. In fig. 5 three spectra (the dots) are shown measured on laser light ($\lambda = 647.1 \text{ nm}$) doubly scattered by particles ($2a = 332 \text{ nm}$) at a temperature

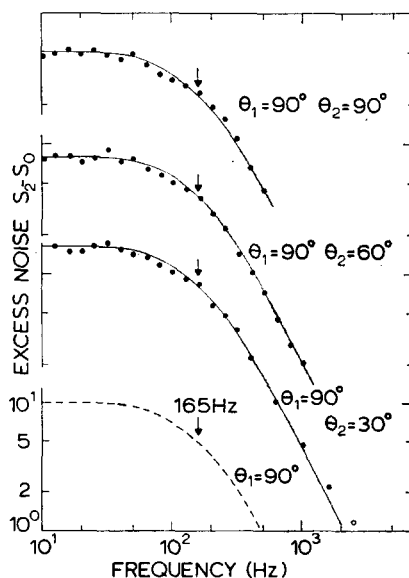


Fig. 5. Excess noise spectra $S_2 - S_0$ in arbitrary units of doubly scattered light. The cells were at a distance of $5.0 \times 10^{-1} \text{ m}$ from each other. The first scattering angle was $\theta_1 = 90^\circ$, the second scattering angles were $\theta_2 = 90^\circ, 60^\circ, 30^\circ$. The full curves are Lorentz spectra fitted on the experimental points. The dashed curve gives the frequency dependence of the spectra for single scattering at a scattering angle $\theta_1 = 90^\circ$.

$T = 28^\circ\text{C}$. The experimental points are well matched by Lorentz spectra (full curves). The dashed Lorentz spectrum gives the frequency dependence of the noise spectral density of singly scattered light at a scattering angle $\theta_1 = 90^\circ$ (see fig. 3). In the double scattering experiment the first scattering angle was $\theta_1 = 90^\circ$ and the second angle θ_2 was varied. However, as the arrows in fig. 5 indicate, all bandwidths of the double scattering spectra were the same. The bandwidths were (within the experimental error of 4%) equal to the spectral bandwidth of the light after the first scattering process: $f_{\frac{1}{2}}(\theta_1 = 90^\circ) = 165 \text{ Hz}$.

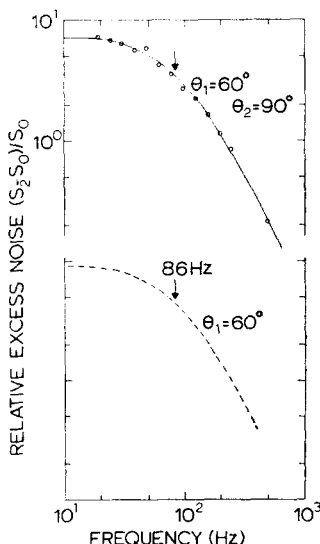


Fig. 6. Relative excess noise $(S_2 - S_0)/S_0$ of doubly scattered light when the distance between the cells was $5.0 \times 10^{-1} \text{ m}$. The first scattering angle is $\theta_1 = 60^\circ$ and the second one $\theta_2 = 90^\circ$. The frequency dependence of the spectrum for single scattering at $\theta_1 = 60^\circ$ is given by the dashed curve.

If we changed the first scattering angle, however, to $\theta_1 = 60^\circ$ the bandwidth of the double scattering spectrum was changed into $f_{\frac{1}{2}}(\theta_1 = 60^\circ, \theta_2 = 90^\circ) = 86 \text{ Hz}$ as can be seen in fig. 6. And this latter bandwidth is again equal to the spectral bandwidth of the intensity fluctuations of singly scattered light at an angle $\theta_1 = 60^\circ$.

4. Discussion

Good agreement between theory and experiment was found when the two scattering cells were close together (case (i)). In the derivation of the intensity cross-

correlation function for this case the assumption given in eq. (30) was used. From the experimental geometry described in sec. 3 it follows that within 1%

$$|\mathbf{k}_0 - \mathbf{k}_{\alpha\alpha}|^2 + |\mathbf{k}_{\alpha\alpha} - \mathbf{k}_Q|^2 \simeq |\mathbf{k}_0 - \mathbf{k}_1|^2 + |\mathbf{k}_1 - \mathbf{k}_2|^2.$$

So the use of eq. (30) is justified.

In the second case we studied, the scattering cells were far apart (case (ii)). From the experimental geometry as mentioned in sec. 3 it follows that eq. (45) holds. In sec. 3 we found that the frequency dependence of the double scattering spectra was the same as that of the single scattered spectrum measured at the first scattering angle. This experimental result can be explained with the help of eq. (55) as a rough calculation of the spatial coherence factors B_1 , B_2 and B_3 gives:

$$B_2/B_1 \simeq 10^{-4} \quad \text{and} \quad B_3/B_1 \simeq 10^{-4}.$$

So the first term in eq. (55) dominates and this term has the same frequency dependence as the spectrum of the singly scattered radiation.

Inspection of eq. (56) makes clear the meaning of the spatial coherence factors B_1 , B_2 and B_3 . The term B_1 describes the spatial coherence of the light beam in the second cell, whereas B_3 describes the spatial coherence at the detector surface. The term B_2 depends on the spatial coherence of the light beam in the second cell as well as on the spatial coherence of the light at the detector surface. The detector area was about a factor 10^6 larger than the "coherence area"¹⁻³) associated with the second cell, whereas the cross section of the beam in the second cell was about a factor 10^2 larger than the coherence area associated with the first cell. Therefore the fluctuations in the light induced by the random movements of the scatterers in the first cell dominate those induced by the scatterers in the second cell. The solid angle within which the light is scattered from the first to the second scattering cell was made small enough to ensure a beam with well-defined properties; however, light scattered from the second cell to the detector was emitted within a large solid angle, so that we had a detectable signal.

Next we compare our theoretical results with those of other authors¹⁵⁻¹⁷). Ivanov *et al.*¹⁵) have calculated the autocorrelation function of the intensity of doubly scattered radiation. Their scattering medium was a layer with moving centres and can be best compared with our case (i) where the cells were close together. Ivanov *et al.* did not take into account the spatial correlation across the detector surface. If we put $\mathbf{k}_p = \mathbf{k}_Q = \mathbf{k}_2$ in eq. (34) we find agreement with their result given in eq. (11) of ref. 15. But where we have simplified eq. (34) to eq. (35), Ivanov *et al.* have performed an integration over the scattering volumes. Their numerical calculation shows that in fairly good approximation the autocorrelation function $\Phi_2(\mathbf{R}_p, \mathbf{R}_p; \tau) - \langle I \rangle^2$ is an exponential function. They have measured the spectral bandwidth of the intensity fluctuations as a function of concen-

tration of the particles. But their results cannot be compared with ours because they detected the light from single, double and multiple scattering simultaneously, whereas we detected doubly scattered light only. Moreover they assumed erroneously that singly and doubly scattered light are statistically independent. In a study of multiple scattering Kelly¹⁶) has calculated the cross-correlation between singly and doubly scattered light. This correlation is not present in our analysis because, as stated before, we investigated double scattering only. Correlation functions for purely doubly scattered light were not given in Kelly's paper.

Bertolotti *et al.*¹⁷) have calculated the photon counting distribution of gaussian light scattered by a gaussian medium. If the medium consisted of randomly moving particles they found for the second factorial moment of the photon counts:

$$\langle n(n-1) \rangle = 4\langle n \rangle^2, \quad (57)$$

where n is the number of photons counted in a time-interval of length τ , which was much shorter than the coherence time of the radiation. This scattering process can be compared with our case (ii) where the cells were far apart, since we may assume that the singly scattered field is gaussian^{1-3,10,13}).

The relation between intensity fluctuations and $\langle n(n-1) \rangle$ is generally given by²⁵) (A less than the coherence area):

$$(\eta A \tau)^2 \langle I^2 \rangle = \langle n(n-1) \rangle. \quad (58)$$

The value of $(\eta A \tau)^2 \langle I^2 \rangle$ can be derived from the noise spectral density of the photocurrent with the help of the relation:

$$(\eta A \tau)^2 \langle I^2 \rangle = (\tau/eM)^2 \int_0^\infty \{S(f) - S_0\} df + (\eta A \tau \langle I \rangle)^2, \quad (59)$$

where $\eta A \tau \langle I \rangle = \langle n \rangle$. Bertolotti *et al.* did not take into account the effect of spatial coherence, so we take $B_1 = B_2 = B_3 = 1$ in eq. (55). Eqs. (48), (55) and (59) now yield:

$$(\eta A \tau)^2 \langle I^2 \rangle = 3(\eta A \tau \langle I \rangle)^2 + \langle n \rangle^2 = 4\langle n \rangle^2, \quad (60)$$

which is in agreement with eq. (57).

Finally we give some thought to the question whether the doubly scattered light is gaussian or not. In case (i) the intensity correlation function obeys eq. (36). And this is a necessary but not a sufficient condition for a gaussian field. In case (ii), however, eq. (36) is not satisfied as inspection of eqs. (41) and (46) makes clear. So in case (ii) the doubly scattered field is certainly not gaussian. This can be explained as follows. If we accept the widely used assumption that the singly scattered field is gaussian, we can interpret the double scattered field as a product

of two gaussian variables. But a product of two gaussian variables never results in a new gaussian variable. Kelly¹⁶⁾ has concluded that an n -fold scattered field does not obey gaussian statistics. But he found that n must be quite large before the departure from gaussian statistics has any experimental significance. His conclusion seems to be correct in the case when the cells are close together but is certainly untrue when the cells are far apart. The non-gaussian character of the light in the latter case is confirmed by the experiments. For, if $E_2(\mathbf{R}_p, t)$ was gaussian, only the third term on the right of eq. (55) would have been found. However, this term in the noise spectral density has not been observed at all in the spectra measured.

Having studied the fluctuations in doubly scattered laser light experimentally and theoretically in this paper, we have come to the following conclusions: in the analysis of double scattering spectra the geometry of the experimental set-up is very important and separation of temporal and spatial correlations cannot always be obtained. In addition we found that a doubly scattered field can have non-gaussian statistics.

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Appendix

From eq. (27) it follows that for a displacement Δr in the time interval τ :

$$\langle (\Delta r_t)^2 \rangle = 6D\tau. \quad (\text{A.1})$$

According to eqs. (3) and (5) the correlation time of the singly scattered field is:

$$t_c = 1/D |\mathbf{k}_0 - \mathbf{k}_s|^2 = \lambda^2/D (4\pi n \sin \frac{1}{2}\theta)^2, \quad (\text{A.2})$$

where λ is the wavelength of the laser light in vacuum, n is the refractive index of the solvent and θ is the angle between \mathbf{k}_0 and \mathbf{k}_s . If we neglect scattering in a forward direction where θ is small, we see that:

$$\langle (\Delta r_{t_c})^2 \rangle^{\frac{1}{2}} \simeq \lambda. \quad (\text{A.3})$$

References

- 1) G.B. Benedek, in: *Polarisation, Matière et Rayonnement*, Livre de Jubilé en l'honneur de Professeur A. Kastler (Presses Universitaires de France, Paris, 1969) p. 49
- 2) H.Z. Cummins and H.L. Swinney, in: *Progress in Optics*, vol. 8, E. Wolf, ed. (North-Holland, Amsterdam, 1970) p. 133.
- 3) B. Chu, *Laser Light Scattering* (Academic Press, New York, 1974).
- 4) R. Pecora, *J. Chem. Phys.* **40** (1964) 1604.
- 5) F.T. Arecchi, M. Giglio and U. Tartari, *Phys. Rev.* **163** (1967) 186.
- 6) R. Foord, E. Jakeman, C.J. Oliver, R.J. Blagrove, E. Wood and A.R. Peacocke, *Nature* **227** (1970) 242.
- 7) N.A. Clark, J.H. Lunacek and G.B. Benedek, *Am. J. Phys.* **38** (1970) 575.
- 8) K. Ohbayashi, S. Kagoshima and A. Ikushima, *Japan. J. Appl. Phys.* **11** (1972) 808
- 9) D.W. Schaefer and B.J. Berne, *Phys. Rev. Letters* **28** (1972) 475.
- 10) S.H. Chen and P. Tartaglia, *Opt. Commun.* **6** (1972) 119.
- 11) P. Tartaglia and S.H. Chen, *Opt. Commun.* **7** (1973) 379.
- 12) T. Aoki, Y. Okabe and K. Sakurai, *Phys. Rev. A* **10** (1974) 259.
- 13) P.N. Pusey, D.W. Schaefer and D.E. Koppel, *J. Phys. A: Gen. Phys.* **7** (1974) 530.
- 14) D.W. Schaefer and P.N. Pusey, *Phys. Rev. Letters* **29** (1972) 843.
- 15) A.P. Ivanov, A. Ya. Khairullina and A.P. Chaikovskii, *Opt. Spectrosc.* **35** (1973) 668.
- 16) H.C. Kelly, *J. Phys. A: Gen. Phys.* **6** (1973) 353.
- 17) M. Bertolotti, B. Crosignani and P. di Porto, *J. Phys. A: Gen. Phys.* **3** (1970) L37.
- 18) C. Kittel, *Elementary Statistical Physics* (John Wiley, New York, 1964).
- 19) H.C. van der Hulst, *Light Scattering by Small Particles* (John Wiley, New York, 1957).
- 20) E. Jakeman, C.J. Oliver and E.R. Pike, *J. Phys. A: Gen. Phys.* **3** (1970) L45.
- 21) L. Mandel, in: *Progress in Optics*, vol. 2, E. Wolf, ed. (North-Holland, Amsterdam, 1970) p. 181.
- 22) S. Chandrasekhar, *Rev. Mod. Phys.* **15** (1943) 1.
- 23) L. Mandel and E. Wolf, *Rev. Mod. Phys.* **37** (1965) 231.
- 24) W.D. Gunter, G.R. Grant and S.A. Shaw, *Appl. Opt.* **9** (1970) 251.
- 25) C.L. Mehta, in: *Progress in Optics*, vol. 8, E. Wolf, ed. (North-Holland, Amsterdam, 1970) p. 373.