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Coherent Properties of Light Scattered by Density Fluctuations in Pure Liquids

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The second-order correlation function for light scattered in a pure liquid is calculated theoretically. It is shown that this function is related to the probability distribution function for a three-molecule configuration. An algorithm for setting up the three-particle function on the basis of experimental data on the measurement of second order coherency is found.

To describe the internal structure of a liquid it is necessary to know the probability distribution functions of different numbers of molecules. The two-particle distribution function can be determined experimentally from the angular distribution of the intensity of light scattered in the liquid. To determine experimentally other distribution functions, it is necessary to measure higher correlation characteristics of the scattered light. To determine the three-particle distribution function it is necessary, as shown below, to measure the second-order spatial coherence and the scattered light.

Unlike the first-order coherence, the second-order coherence can differ from zero also when the scattering object is uniformly polarized. We therefore express the polarization of the medium in the form

$$P(\mathbf{r},\ t) = \alpha\mathscr{E}n(\mathbf{r})\exp\{i\ (\mathbf{k}_0\mathbf{r} - \omega_0 t)\}.$$

where α is the average polarizability of one molecule, $\mathscr{E}$ the amplitude of the effective field in the medium, and n(r) the density of the number of molecules in the medium. We shall assume the volume V of the scattering medium to be large enough to permit the function n(r) to be represented by a Fourier integral, i.e.,

$$n(\mathbf{r}) = \int n(\mathbf{q})e^{i\mathbf{q}\mathbf{r}}\frac{d\mathbf{q}}{(2\pi)^3}, \qquad n(\mathbf{q}) = \int n(\mathbf{r})e^{-i\mathbf{q}\mathbf{r}}\,d\mathbf{r}.$$

The electric field intensity of the scattered light at a point **r** remote from the scattering region is then equal to[1]

$$E(\mathbf{r}) = \alpha\mathscr{E}k^2\frac{e^{-ikr}}{r}\sin\theta n(\mathbf{q}), \tag{1}$$

where k = |**k**| = |**k**$_0$| = ω_0/c, the vector **k** is directed towards the observation point, θ is the angle between the direction of the polarization of the light and the vector **k**, and **q** = **k** − **k**$_0$. Light scattering is usually investigated near the critical point of a phase transition, where it is particularly strong. In this region it is possible to disregard with assurance the fluctuations of the liquid temperature and the scattered-light depolarization associated with these fluctuations.

With the aid of (1) we construct the second-order correlation function for the scattered light:

$$\Gamma^{(2)}\{\mathbf{R}\} = \langle E(\mathbf{r}_1)E^*(\mathbf{r}_2)E(\mathbf{r}_3)E^*(\mathbf{r}_4)\rangle = \alpha^4\mathscr{E}^4k^8G^{(2)}(\mathbf{q}_1,\ -\mathbf{q}_2,\ \mathbf{q}_3,\ -\mathbf{q}_4)$$
$$\times\frac{\exp\{-i(\mathbf{k}_1\mathbf{r}_1 - \mathbf{k}_2\mathbf{r}_2 + \mathbf{k}_3\mathbf{r}_3 - \mathbf{k}_4\mathbf{r}_4)\}}{r_1r_2r_3r_4}\sin\theta_1\sin\theta_2\sin\theta_3\sin\theta_4, \tag{2}$$

where

$$G^{(2)}\{\mathbf{K}\} = \int G^{(2)}\{\mathbf{R}\}\exp\{-i(\varkappa_1\mathbf{r}_1 + \varkappa_2\mathbf{r}_2 + \varkappa_3\mathbf{r}_3 + \varkappa_4\mathbf{r}_4)\}\,d\mathbf{r}_1\,d\mathbf{r}_2\,d\mathbf{r}_3\,d\mathbf{r}_4,$$
$$G^{(2)}\{\mathbf{R}\} = \langle n(\mathbf{r}_1)n(\mathbf{r}_2)n(\mathbf{r}_3)n(\mathbf{r}_4)\rangle.$$

The angle brackets denote here averaging over the ensemble of the scattering centers, and we have introduced the following arbitrary notation for the sets of variables:

$$\{\mathbf{R}\} = (\mathbf{r}_1,\ \mathbf{r}_2,\ \mathbf{r}_3,\ \mathbf{r}_4), \quad \{\mathbf{K}\} = (\varkappa_1,\ \varkappa_2,\ \varkappa_3,\ \varkappa_4).$$

At each given molecule configuration, the density of the number of particles at the point $\mathbf{r}_k$ is

$$n(\mathbf{r}_k) = \sum_{i=1}^{N}\delta(\mathbf{r}_k - \mathbf{x}_i) = \sum_{i=1}^{N}\delta_i^k.$$

Then, as can be readily seen,

$$G^{(2)}\{\mathbf{R}\} = \int\sum_{i,j,k,l=1}^{N}\delta_i^1\delta_j^2\delta_k^3\delta_l^4W_N(\mathbf{x}_1,\ldots,\mathbf{x}_N)\,d\mathbf{x}_1\ldots d\mathbf{x}_N,$$

where W_N is the Gibbs distribution function of the total number of molecules in the medium. From the sum under the integral sign we can separate four groups of terms. In the first group, all the numbers out of the quartet (i, j, k, l) coincide, in the second they assume only two different values, in the third three, and in the fourth they are all different. We denote the integrals of the sums of these groups by $G_1^{(2)}$, $G_2^{(2)}$, $G_3^{(2)}$, and $G_4^{(2)}$, respectively.

Following Bogolyubov, we introduce a sequence of normalized s-particle distribution functions

$$w_s(\mathbf{x}_1,\ldots,\mathbf{x}_s) = V^s\int W_N(\mathbf{x}_1,\ldots,\mathbf{x}_N)\,d\mathbf{x}_{s+1}\ldots d\mathbf{x}_N. \tag{3}$$

Then we obtain after straightforward but rather cumbersome manipulations, taking into account the identity of the molecules ($\delta^{ij} = \delta(\mathbf{r}_i - \mathbf{r}_j)$)

$$G_1^{(2)}\{\mathbf{R}\} = \frac{N}{V}\delta^{21}\delta^{31}\delta^{41}w_1(\mathbf{r}_1), \qquad w_1(\mathbf{r}) = 1, \tag{4.1}$$

$$G_2^{(2)}\{\mathbf{R}\} = \frac{N(N-1)}{V^2}[w_2(\mathbf{r}_1,\mathbf{r}_2)(\delta^{13}\delta^{14} + \delta^{23}\delta^{34} + \delta^{13}\delta^{24} + \delta^{23}\delta^{14})$$
$$+ w_2(\mathbf{r}_2,\mathbf{r}_3)(\delta^{21}\delta^{41} + \delta^{12}\delta^{43}) + w_2(\mathbf{r}_3,\mathbf{r}_4)\delta^{31}\delta^{32}], \tag{4.2}$$

$$G_3^{(2)}\{\mathbf{R}\} = \frac{N(N-1)(N-2)}{V^3}[w_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)(\delta^{41} + \delta^{42} + \delta^{43})$$
$$+ w_3(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_4)(\delta^{31} + \delta^{32} + \delta^{34}) + w_3(\mathbf{r}_1,\mathbf{r}_3,\mathbf{r}_4)(\delta^{21} + \delta^{23} + \delta^{24})$$
$$+ w_3(\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4)(\delta^{12} + \delta^{13} + \delta^{14})], \tag{4.3}$$

$$G_4^{(2)}\{\mathbf{R}\} = \frac{N(N-1)(N-2)(N-3)}{V^4} w_4(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3, \mathbf{r}_4). \quad (4.4)$$

To find the correlation function (2) of the scattered light it is necessary to calculate the Fourier transform $G^{(2)}\{K\}$ of the function $G^{(2)}\{R\}$. Let us prove first one simple property of the Fourier transform of the function w_s. Since w_s does not depend on the location of the group of s molecules inside the homogeneous liquid, it follows that

$$w_s(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_s) = f_s(\Delta_2, \Delta_3, \ldots, \Delta_s),$$

where $\Delta_i = \mathbf{r}_i - \mathbf{r}_1$ and $i = 2, 3, \ldots, s$. Consequently,

$$\begin{aligned} w_s(\varkappa_1, \ldots, \varkappa_s) = \int f_s(\Delta_2, \ldots, \Delta_s) \exp\{-i(\mathbf{K}\mathbf{r}_1 + \varkappa_2\Delta_2 \\ \ldots + \varkappa_s\Delta_s)\} d\mathbf{r}_1\, d\Delta_2, \ldots, d\Delta_s = \delta(\mathbf{K}) f_s(\varkappa_2, \ldots, \varkappa_s), \end{aligned} \quad (5)$$

where $K = \kappa_1 + \kappa_2 + \ldots + \kappa_s$. Thus, by virtue of the translational symmetry of the distribution function, its Fourier transform, and hence also the function $G^{(2)}\{K\}$, differs from zero only when $K = 0$. For formula (2) this condition denotes $q_1 - q_2 + q_3 - q_4 = 0$, or, which is the same, $\mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k}_3 - \mathbf{k}_4 = 0$. The latter is evidently satisfied automatically for the correlation function of the intensities of the scattered light:

$$\Gamma^{(2)}(\mathbf{r}_1, \mathbf{r}_2) = \langle |E(\mathbf{r}_1)|^2 |E(\mathbf{r}_2)|^2 \rangle.$$

Using (5), we obtain by direct calculation the Fourier transforms of expressions (4.1)–(4.4):

$$G_1^{(2)}\{\mathbf{K}\} = \frac{N}{V}\delta(\mathbf{K}), \quad (6.1)$$

$$\begin{aligned} G_2^{(2)}\{\mathbf{K}\} = \frac{N(N-1)}{V^2}\delta(\mathbf{K})\,[f_2(\varkappa_1) + f_2(\varkappa_2) + f_2(\varkappa_3) + f_2(\varkappa_4) \\ + f_2(\varkappa_1 + \varkappa_2) + f_2(\varkappa_1 + \varkappa_3) + f_2(\varkappa_1 + \varkappa_4)], \end{aligned} \quad (6.2)$$

$$\begin{aligned} G_3^{(2)}\{\mathbf{K}\} = 2\frac{N(N-1)(N-2)}{V^3}\delta(\mathbf{K})\,[f_3(\varkappa_1, \varkappa_2) + f_3(\varkappa_1, \varkappa_3) + f_3(\varkappa_1, \varkappa_4) \\ + f_3(\varkappa_2, \varkappa_3) + f_3(\varkappa_2, \varkappa_4) + f_3(\varkappa_3, \varkappa_4)], \end{aligned} \quad (6.3)$$

$$G_4^{(2)}\{\mathbf{K}\} = \frac{N(N-1)(N-2)(N-3)}{V^4}\delta(\mathbf{K}) f_4(\varkappa_2, \varkappa_3, \varkappa_4). \quad (6.4)$$

We consider next the particular case $\kappa_1 = -\kappa_2 = q_1$ and $\kappa_3 = -\kappa_4 = q_2$, corresponding to the correlation of the intensities at two points of the scattered-light field. We see that in this case functions of the type $f_2(q = 0)$, $f_3(q, -q)$, and $f_4(-q_1, q_2, -q_2)$ appear in formulas (6). As can be readily seen, it follows directly from the definition (3) that

$$\delta(\mathbf{K}) f_2(0) = \int w_2(\mathbf{r}_1, \mathbf{r}_2)\, d\mathbf{r}_1\, d\mathbf{r}_2 = V^2, \quad (7.1)$$

$$\begin{aligned} \delta(\mathbf{K}) f_2(\mathbf{q}, -\mathbf{q}) = \int w_3(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) e^{-i\mathbf{q}(\mathbf{r}_2 - \mathbf{r}_3)}\, d\mathbf{r}_1\, d\mathbf{r}_2\, d\mathbf{r}_3 \\ = V\int w_2(\mathbf{r}_2, \mathbf{r}_3) e^{-i\mathbf{q}(\mathbf{r}_2 - \mathbf{r}_3)}\, d\mathbf{r}_2\, d\mathbf{r}_3 = V^2 f_2(\mathbf{q}). \end{aligned} \quad (7.2)$$

In addition, Prigogine's book[2] contains the proof of a theorem concerning the factoring of the Fourier transform of a four-particle distribution function:

$$w_4(\mathbf{q}_1, -\mathbf{q}_1, \mathbf{q}_2, -\mathbf{q}_2) = w_2(\mathbf{q}_1, -\mathbf{q}_1) w_2(\mathbf{q}_2, -\mathbf{q}_2). \quad (8)$$

Therefore

$$\delta(\mathbf{K}) f_4(-\mathbf{q}_1, \mathbf{q}_2, -\mathbf{q}_2) = V^2 f_2(\mathbf{q}_1) f_2(\mathbf{q}_2). \quad (7.3)$$

Using the obtained relations and recognizing that $K = 0$ we have $\delta(K) = V$, we rewrite formulas (6) in the form

$$G_1^{(2)}(\mathbf{q}_1, \mathbf{q}_2) = N,$$

$$G_2^{(2)}(\mathbf{q}_1, \mathbf{q}_2) = \frac{N(N-1)}{V}[2f_2(\mathbf{q}_1) + 2f_2(\mathbf{q}_2) + f_2(\mathbf{q}_1 + \mathbf{q}_2) + f_2(\mathbf{q}_1 - \mathbf{q}_2) + V],$$

$$\begin{aligned} G_3^{(2)}(\mathbf{q}_1, \mathbf{q}_2) = 2\frac{N(N-1)(N-2)}{V^2}[2\,\mathrm{Re}(f_3(\mathbf{q}_1, \mathbf{q}_2) + f_2(\mathbf{q}_1, -\mathbf{q}_2)) \\ + V(f_2(\mathbf{q}_1) + f_2(\mathbf{q}_2))], \end{aligned}$$

$$G_4^{(2)}(\mathbf{q}_1, \mathbf{q}_2) = \frac{N(N-1)(N-2)(N-3)}{V^2} f_2(\mathbf{q}_1) f_2(\mathbf{q}_2).$$

We add the last formulas together, and retain in the distribution function of each order only the maximum degree of the unusually large number N. This is permissible, since the distribution functions of a small ($\ll$ N) number of molecules depends very little on N (the canonical and grand canonical ensembles are equivalent). Introducing the average particle-number density $n = N/V$, we obtain

$$\begin{aligned} G^{(2)}(\mathbf{q}_1, \mathbf{q}_2) = V^2[n^2 + 2n^3(f_2(\mathbf{q}_1) + f_2(\mathbf{q}_2)) \\ + n^4 f_2(\mathbf{q}_1) f_2(\mathbf{q}_2)] + 4Vn^3\,\mathrm{Re}(f_3(\mathbf{q}_1, \mathbf{q}_2) + f_3(\mathbf{q}_1, -\mathbf{q}_2)). \end{aligned} \quad (9)$$

Munster[3] cites the first-order autocorrelation function for the density fluctuations:

$$G^{(1)}(\mathbf{q}, -\mathbf{q}) = G^{(1)}(\mathbf{q}) = V[n + n^2 f_2(\mathbf{q})],$$

which determines the distribution of the scattered-light intensity

$$\Gamma^{(1)}(\mathbf{r}) = \alpha^2 \mathscr{E}^2 k^4 \frac{\sin^2\theta}{r} G^{(1)}(\mathbf{q}).$$

Let us consider the difference $D(q_1, q_2) = G^{(2)}(q_1, q_2) - G^{(1)}(q_1) \cdot G^{(1)}(q_2)$. Obviously,

$$D(\mathbf{q}_1; \mathbf{q}_2) = V^2 n^3 [f_2(\mathbf{q}_1) + f_2(\mathbf{q}_2) + 8V^{-1} f_3^{(c)}(\mathbf{q}_1, \mathbf{q}_2)], \quad (10)$$

where

$$f_3^{(c)}(\mathbf{q}_1, \mathbf{q}_2) = \int f_3(\Delta_1, \Delta_2) \cos \mathbf{q}_1\Delta_1 \cos \mathbf{q}_2\Delta_2\, d\Delta_1\, d\Delta_2 \quad (11)$$

(vector cosine) is the Fourier transform of the three-particle distribution function. Comparing (2), (10), and the expression for $\Gamma^{(1)}$, we see that the function $D(q_1, q_2)$ can be determined experimentally from the formula

$$\Gamma^{(2)}(\mathbf{r}_1, \mathbf{r}_2) - \Gamma^{(1)}(\mathbf{r}_1)\Gamma^{(1)}(\mathbf{r}_2) = \alpha^4 \mathscr{E}^4 k^8 \frac{\sin^2\theta_1 \sin^2\theta_2}{r_1^2 r_2^2} D(\mathbf{q}_1, \mathbf{q}_2).$$

Since the function $f_2(q)$ is known from measurements of the angular distribution of the intensity of the static light, formula (10) makes it possible to find the experimental values of the function $f_3^{(C)}(q_1, q_2)$.

The next task is to invert the transformation defined by formula (11). It can be solved by assuming (11) to be an ordinary Fourier transform, but it is necessary for this purpose to know the values of the function $f_3^{(C)}(q_1, q_2)$, in the entire six-dimensional space of variation of the arguments, i.e., to measure the scattering parameters in the entire frequency range. Fortunately, the problem can be simplified if it is noted that the function $f_3(\Delta_1, \Delta_2)$ is isotropic, and is therefore completely defined by only three variables, for example the lengths of the vectors Δ_1 and Δ_2 and the angle γ between them. Then, as will be shown below, its transform $f_3^{(C)}(q_1, q_2)$ is completely determined by the lengths of the vectors q_1 and q_2 and by the angle ψ between them. Consequently, it can be obtained from a scattering experiment at a single frequency. This greatly facilitates the experiment and the reduction of its results.

Let us introduce the angles ϑ_1, ϑ_2, φ_1, and φ_2, which define the orientations of the vectors Δ_1 and Δ_2 in a spherical coordinate system with a polar axis directed along the vector q_1, and the angle β between the vectors Δ_2 and q_2 (see the figure). In terms of the variables $\{\xi\} = (\Delta_1, \Delta_2, \vartheta_1, \vartheta_2, \varphi_1, \varphi_2)$, we obviously have $(d\Delta_1 d\Delta_2)_\xi = \Delta_1^2\Delta_2^2 \sin\vartheta_1 \sin\vartheta_2\, d\Delta_1 d\Delta_2 d\vartheta_1 d\vartheta_2 d\varphi_1 d\varphi_2$. Using the well known formulas of spherical trigonometry

$$\cos\gamma = \cos\vartheta_1\cos\vartheta_2 + \sin\vartheta_1\sin\vartheta_2\cos(\varphi_1-\varphi_2),$$

$$\cos\beta = \cos\psi\cos\vartheta_2 + \sin\psi\sin\vartheta_2\cos\varphi_2,$$

we change over to the set of variables $\{\zeta\} = \{\Delta_1, \Delta_2, \vartheta_1, \vartheta_2, \beta, \gamma\}$. Then, after calculating the Jacobian, we can rewrite the transform (11) in the form

$$f_2(q_1,q_2,\psi) = \int d\Delta_1\, d\Delta_2\, d\vartheta_1\, d\vartheta_2\, d\gamma\, d\beta f_3(\Delta_1,\Delta_2,\gamma)\Delta_1^2\Delta_2^2\sin\vartheta_1\sin\vartheta_2\sin\beta\sin\gamma$$
$$\times\cos(q_1\Delta_1\cos\vartheta_1)\cos(q_2\Delta_2\cos\beta)[(\cos\beta-\cos(\psi-\vartheta_2))(\cos\beta-\cos(\psi+\vartheta_2))$$
$$\times(\cos\vartheta_1-\cos(\gamma-\vartheta_2))(\cos\vartheta_1-\cos(\gamma+\vartheta_2))]^{-1/2}. \qquad (12)$$

It is seen from the figure that at fixed γ and ϑ_2, the inequalities $\gamma-\vartheta_2 \le \vartheta_1 \le \gamma+\vartheta_2$ and $\psi-\vartheta_2 \le \beta \le \psi+\vartheta_2$ are satisfied. Taking this into account, we integrate with respect to ϑ_1 and β. We have

$$\int\limits_{\psi-\vartheta_2}^{\psi+\vartheta_2}\frac{\cos(q_2\Delta_2\cos\beta)\sin\beta\,d\beta}{[(\cos\beta-\cos(\psi-\vartheta_2))(\cos\beta-\cos(\psi+\vartheta_2))]^{1/2}}$$
$$=\frac{2}{\pi i}\cos(q_2\Delta_2\cos\vartheta_2)J_0(q_2\Delta_2\sin\psi\sin\vartheta_2)\cdot$$

The second integral with respect to ϑ_1 is evaluated in similar fashion, after which, to determine the kernel of the sought integral transformation, it remains to calculate the integral

$$I=\frac{4}{\pi^2}\int\limits_0^\pi J_0(q_1\Delta_1\sin\gamma\sin\vartheta_2)\cos(q_1\Delta_1\cos\gamma\cos\vartheta_2)$$
$$\times J_0(q_2\Delta_2\sin\psi\sin\vartheta_2)\cos(q_2\Delta_2\cos\psi\cos\vartheta_2)\sin\vartheta_2\,d\vartheta_2. \qquad (13)$$

We note for this purpose that the integral ([4], Sec. 19.4, formula (21))

$$a_{2n}(q_2,\psi)=\int\limits_0^\pi\cos(q_2\Delta_2\cos\psi\cos\vartheta_2)J_0(q_2\Delta_2\sin\psi\sin\vartheta_2) \qquad (14)$$
$$\times P_{2n}(\cos\vartheta_2)\sin\vartheta_2\,d\vartheta_2=(-1)^n\left(\frac{2\pi}{q_2\Delta_2}\right)^{1/2}J_{2n+1/2}(q_2\Delta_2)P_{2n}(\cos\psi)$$

determines the coefficients of the expansion of the function

$$\cos(q_2\Delta_2\cos\psi\cos\vartheta_2)J_0(q_2\Delta_2\sin\psi\sin\vartheta_2)$$

in Legendre polynomials $P_n(\cos\vartheta_2)$ on the segment $(0, \pi)$ (the odd coefficients are equal to zero), i.e.,

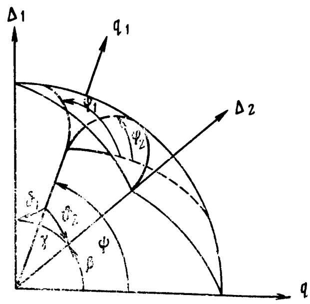
$$\cos(q_2\Delta_2\cos\psi\cos\vartheta_2)J_0(q_2\Delta_2\sin\psi\sin\vartheta_2)$$
$$=\sum_{n=0}^\infty\frac{4n+1}{2}a_{2n}(q_2,\psi)P_{2n}(\cos\vartheta_2).$$

Substituting this expansion in (13) and again using formula (14), we get

$$I=\frac{4}{\pi^2}\sum_{n=0}^\infty\frac{4n+1}{2}a_{2n}(q_2,\psi)a_{2n}(q_1,\gamma).$$

Thus, (12) takes the final form

$$\sqrt{q_1q_2}f_3(q_1,q_2,\psi)=\frac{8}{\pi}\int\limits_0^\infty d\Delta_1\int\limits_0^\infty d\Delta_2\int\limits_0^\pi d\gamma\Delta_1\Delta_2\sin\gamma \qquad (15)$$
$$\times\sqrt{\Delta_1\Delta_1}f_3(\Delta_1,\Delta_2,\gamma)\sum_{n=0}^\infty\frac{4n+1}{2}J_{2n+1/2}(q_1\Delta_1)J_{2n+1/2}(q_2\Delta_2)P_{2n}(\cos\gamma)P_{2n}(\cos\psi).$$



To inverse this transformation, it is necessary to construct a delta-functional with the aid of its kernel. To this end we use the well known relations

$$\int\limits_0^\infty J_\nu(kx)J_\nu(k'x)x\,dx=\frac{1}{k}\delta(k-k'),$$

$$\int\limits_0^\pi P_n(\cos\vartheta)P_{n'}(\cos\vartheta)\sin\vartheta\,d\vartheta=\frac{2}{2n+1}\delta_{nn'}.$$

In addition, in the space of functions that can be expanded only in even Legendre polynomials in the segment $(0, \pi)$, the δ-functional is defined by the formula

$$\sum_{n=0}^\infty\frac{4n+1}{2}P_{2n}(\cos\vartheta)P_{2n}(\cos\vartheta')=\delta(\cos\vartheta-\cos\vartheta').$$

We then readily obtain

$$\sum_{m=0}^\infty\sum_{n=0}^\infty\int\limits_0^\infty q_1\,dq_1\int\limits_0^\infty q_2\,dq_2\int\limits_0^\pi\sin\psi\,d\psi P_{2n}(\cos\psi)P_{2m}(\cos\psi)$$
$$\times P_{2n}(\cos\gamma)P_{2m}(\cos\gamma')J_{2n+1/2}(q_1\Delta_1)J_{2m+1/2}(q_1\Delta_1')$$
$$\times J_{2n+1/2}(q_2\Delta_2)J_{2m+1/2}(q_2\Delta_2')\frac{4n+1}{2}\frac{4m+1}{2}$$
$$=\frac{1}{\Delta_1\Delta_2}\delta(\Delta_1-\Delta_1')\delta(\Delta_2-\Delta_2')\delta(\cos\gamma-\cos\gamma').$$

Consequently, the sought inverse transform is

$$\sqrt{\Delta_1\Delta_2}f_3(\Delta_1,\Delta_2,\gamma)=\frac{\pi}{8}\int\limits_0^\infty q_1\,dq_1\int\limits_0^\infty q_2\,dq_2\int\limits_0^\pi\sin\psi\,d\psi \qquad (16)$$
$$\times\sqrt{q_1q_2}f_3(q_1,q_2,\psi)\sum_{n=0}^\infty\frac{4n+1}{2}J_{2n+1/2}(q_1\Delta_1)J_{2n+1/2}(q_2\Delta_2)P_{2n}(\cos\psi)P_{2n}(\cos\gamma).$$

Summarizing, we note once more that the fundamental functions of statistical mechanics of many-particle systems, namely the correlation functions of the probability distributions, can be determined experimentally from the correlation characteristics of radiation scattered by these systems. In the present paper we investigated only the second-order coherence of light scattered by density fluctuations, but the formalism can be used without limitations for higher orders of coherence of light, and also for other scattering mechanisms (for example, scattering of light by anisotropy fluctuations in liquids and crystals, scattering of neutrons in magnets, etc.). Hopefully, the experimental investigations in this direction will help explain the general structure of many-particle distribution functions and determine the accuracy of the approximations used in the theory.

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