GENERALIZED PARAMETERS OF A POLYDISPERSED INTEGRAL LIGHT SCATTERING PHASE FUNCTION

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The generalized parameters of an integral light scattering phase function for a polydispersed suspension of the spherical particles are considered. The application of these parameters to simplify this problem to that for monodispersed suspension is analyzed on the basis of the obtained analytic expression for polydispersed integral scattering phase function in the Rayleigh–Gans–Debye approximation. The calculated results are presented.

Of special interest in different fields of astronomy, biophysics, chemistry, and medicine is light scattering by optically "soft" particles whose refractive index is close to that of a surrounding medium (their relative refractive index $m \to 1$ or $|m-1| \ll 1$, see Refs. 1–3). A whole class or group of "soft" anomalous diffraction (AD), i.e., large particles with the diffraction parameter $\rho \gg 1$, was considered in close detail in Ref. 4. There are practically no papers aimed at the application of the generalized parameters of polydispersed suspensions of spherical particles in the Rayleigh–Gans–Debye (RGD) range of small phase shift $\Delta = k\,d\,|m-1| \ll 1$, where d is the maximum diameter of particles.

The present paper is aimed at search for the generalized parameters of an integral phase function of light scattering by spherical particles in the RGD approximation. By the term *integral scattering phase function* is meant the relative fraction of energy flux, scattered within a cone with the vertex angle θ, of total light scattering.

We use the equation obtained in Ref.5 for the light intensity scattered within a cone with the vertex angle θ (by an individual spherical particle)

$$\frac{I_\theta}{\pi a^2} = I_0\,|m-1|^2\left\{A + 2\rho^2 + (4b^2 - 2A)\frac{\sin(4\rho b)}{4\rho b} + \right.$$
$$\left. + \left[\frac{\cos(4\rho b) - 1}{(4\rho b)^2}\right](12b^2 - 2A) + \left(\frac{1}{2\rho^2} - 2\right)S_1(4\rho b)\right\}, \quad (1)$$

where $A = 2 + b^2 - \dfrac{1}{2b^2}$, $S_1(x) = \displaystyle\int_0^x \frac{1-\cos u}{u}\,\mathrm{d}u = \ln(x) + \gamma - Ci(x)$, $Ci(x) = \displaystyle\int_\infty^x \frac{\cos u}{u}\,\mathrm{d}u$ is the integral cosine, $b = \sin(\theta/2)$, $\gamma = 0.577216$ is the Euler–Maskeroni constant, and a is the particle radius. From Eq. (1) we may derive the scattering efficiency factor K_s since the vertex angle $\theta = \pi$ and hence $b = 1$, i.e.,

$$K_s = \frac{k_s}{\pi a^2} = \frac{I_\pi}{\pi a^2} = I_0\,|m-1|^2\left\{\frac{5}{2} + 2\rho^2 - \frac{\sin(4\rho)}{4\rho} + \right.$$
$$\left. + \frac{7}{16\rho^2}[\cos(4\rho) - 1] + \left(\frac{1}{2\rho^2} - 2\right)S_1(4\rho)\right\}, \quad (2)$$

coinciding with the formula which has already been obtained by Rayleigh.[2,4,6]

The integral scattering phase function $F(2\rho b)$ for an RGD sphere is the ratio of Eq. (1) to Eq. (2):

$$F[2\rho\sin(\theta/2)] = \frac{I_\theta}{k_s}. \quad (3)$$

We hereafter use the probability density functions in the following form[4,8]:

a) gamma distribution

$$f(a) = \begin{cases} 0, & a < 0, \\ \dfrac{\beta^{\mu+1}}{\Gamma(\mu+1)}\,a^\mu \exp(-\beta a), & a \geqslant 0, \quad (\mu > -1,\ \beta > 0), \end{cases} \quad (4)$$

where $\beta = (\mu + 1)/\overline{a}$ and $\overline{a}$ is the mean radius of particles in polydispersed suspension;

b) power–law distribution

$$f(a) = \begin{cases} 0, & \text{for } a < a_{\min} \text{ and } a > a_{\max} \\ \dfrac{(\nu-1)\,a_{\min}}{1 - R^{1-\nu}}\,a^{-\nu} = C_\nu\,a^{-\nu}, & \text{for } a_{\min} \le a \le a_{\max}, \end{cases} \quad (5)$$

where $R = a_{\max}/a_{\min}$; $a_{\min}$, and $a_{\max}$ are the minimum and maximum radii of particles in suspension. To obtain the integral scattering phase function of polydispersed suspension of spherical particles, Eqs. (1) and (2) must be integrated taking into account the probability density function and particle cross section. In such a manner,

$$\langle F(\theta)\rangle = \frac{\pi\displaystyle\int_0^\infty I_\theta\,f(a)\,a^2\,\mathrm{d}a}{\pi\displaystyle\int_0^\infty K_s\,f(a)\,a^2\,\mathrm{d}a} = \frac{\langle I_\theta\rangle}{\langle k_s\rangle}. \quad (6)$$

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To obtain the integral scattering phase function $\langle F(\theta)\rangle$, we need only to calculate $\langle I(\theta)\rangle$ in Eq. (6) taking into account that $\langle k_s\rangle = \langle I_\pi\rangle$. Therefore, using Eqs. (1) and (4), we obtain

$$\langle I_\theta\rangle = \frac{\pi I_0 |m-1|^2 \beta^{\mu+1}}{k^{\mu+3}\Gamma(\mu+1)} \int\limits_0^\infty \Bigg[A\rho^2 + 2\rho^4 + (2A - 4b^2) \times$$

$$\times \frac{\sin(4\rho b)}{4b}\rho + \left(\frac{12b^2 - 2A}{16b^2}\right)(\cos(4\rho b) - 1) +$$

$$+ (1/2 - 2\rho^2) S_1(4\rho b) \Bigg] \rho^\mu \exp[(-\beta/k)\rho] \, d\rho . \qquad (7)$$

The substitution $\rho = ka$ was employed in Eq. (7), i.e., it was integrated over ρ rather than over the radius a.

After integration of Eq. (7) we finally obtain the relation for $\langle I_\theta\rangle$ in the form of a finite sum of elementary functions.[5] The integral scattering phase function $\langle I_\theta\rangle/\langle k_s\rangle$ for the power–law distribution can be obtained in a similar way (see Ref. 5).

Four different parameters were chosen for generalized ones. In the case of gamma distribution they have the following form:

$$\begin{aligned} &1.\ \langle\rho\rangle = (\mu+1)(k/\beta) = t_1 , \\ &2.\ \sqrt{\langle\rho^6\rangle/\langle\rho^4\rangle} = \sqrt{(\mu+5)(\mu+6)}\,(k/\beta) = t_2 , \\ &3.\ \sqrt{\langle\rho^4\rangle/\langle\rho^2\rangle} = \sqrt{(\mu+3)(\mu+4)}\,(k/\beta) = t_3 , \\ &4.\ \frac{3}{4} k\langle V\rangle/\langle S\rangle = (\mu+3)(k/\beta) = t_4 , \end{aligned} \qquad (8)$$

where k is the wave number.

The generalized coordinates are denoted by $t_i\theta$. In the case of the power–law distribution the generalized parameters, in analogy with the gamma distribution, have the following form:

$$\begin{aligned} &1.\ \langle\rho\rangle = \rho_{min}\frac{(3-\nu)(R^{4-\nu}-1)}{(2-\nu)(R^{1-\nu}-1)} = t_1 , \\ &2.\ \sqrt{\langle\rho^6\rangle/\langle\rho^4\rangle} = \rho_{min}\sqrt{\frac{(5-\nu)(R^{7-\nu}-1)}{(7-\nu)(R^{5-\nu}-1)}} = t_2 , \\ &3.\ \sqrt{\langle\rho^4\rangle/\langle\rho^2\rangle} = \rho_{min}\sqrt{\frac{(3-\nu)(R^{5-\nu}-1)}{(5-\nu)(R^{3-\nu}-1)}} = t_3 , \\ &4.\ \frac{3}{4} k\langle V\rangle/\langle S\rangle = \rho_{min}\frac{(3-\nu)(R^{4-\nu}-1)}{(4-\nu)(R^{3-\nu}-1)} = t_4 . \end{aligned} \qquad (9)$$

A comparison was made for the power 4, which is most widely used in experimental distributions, as well as in the ranges of variation of the particle size 5–25 and 5–15 (see Refs. 4, 8, and 9).

The results of comparison are given in Table I. The values of the integral phase function of scattering by an individual particle calculated from formula (3) and by an ensemble of spherical particles are compared; in addition, the values of $t_i\theta$ are no more than 3, i.e., the scattering at small angles is analyzed. The relative error is calculated using the formula

$$(F_{approx} - F_{exact}) \cdot 100\% / F_{exact} . \qquad (10)$$

It is seen from the Table I that the relative error in calculating $\langle\rho\rangle$ decreases with increase of μ in the case of gamma distribution. Nevertheless, for "real" suspensions of particles of arbitrary size, i.e., for $\mu < 10$, the error reaches –60%.

The situation is much better for the second generalized parameter $\sqrt{\langle\rho^6\rangle/\langle\rho^4\rangle}$, because the error is no more than 12% even at $\mu = 7$. Naturally, it decreases with increase of μ and becomes less than 8% already at $\mu = 10$ under the same conditions.

TABLE I. Maximum relative error in calculation of the integral phase function of light scattering by an individual particle in comparison with scattering by an ensemble of spherical particles, obeying the gamma and power–law distributions, in the generalized coordinates.

Distribution	Generalized parameter			
	$\langle\rho\rangle$	$\sqrt{\langle\rho^6\rangle/\langle\rho^4\rangle}$	$\sqrt{\langle\rho^4\rangle/\langle r^2\rangle}$	$\frac{3}{4}k\langle V\rangle/\langle S\rangle$
Gamma $\mu = 7$, $\beta = 0.1$	– 59%	12%	– 30%	– 35%
$\mu = 10$, $\beta = 0.1$	– 49%	8%	– 24%	– 29%
Power–law $\nu = 4$ $\rho_{min} = 5$, $\rho_{max} = 25$	– 80%	+ 18%	– 51%	– 32%
$\nu = 4$ $\rho_{min} = 5$, $\rho_{max} = 15$	– 57%	+ 10%	– 33%	+ 17%

The error is less than –30% at $\mu = 7$ for the third generalized parameter $\sqrt{\langle\rho^4\rangle/\langle\rho^2\rangle}$ chosen from the condition of minimization of the error in $\langle k_s\rangle$. For the fourth generalized parameter $k\langle V\rangle/\langle S\rangle$, characterizing the ratio of the mean volume to the mean cross section of suspended particles, the error is small and is no more than –35 to –30% for $\mu < 10$, although it decreases insignificantly with further increase of μ. The behavior of relative error is analogous for the power–law distribution. Obviously, the absolute value of the error decreases considerably with decrease of ρ_{max}, when suspension tends to monodispersed one.

Thus based on the results of comparison we may conclude that only the second generalized parameter

$\sqrt{<\rho^6>/<\rho^4>}$ provides the minimum error in the region of small–angle scattering in comparison with the phase function of scattering by an individual particle. The most "natural" parameter $<\rho>$ (mean particle size) gives the rough estimate. However, it should be noted that the relative refractive index was disregarded in comparison (because we set $m = 1$). In practice, the influence of the last factor results in the considerable decrease of relative error thereby providing the use of the mean particle size $<\rho>$ with an error of about 30%[4].

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