

FEW-PARAMETER MODELS FOR EVALUATION OF CROSS SECTION OF EXTINCTION, SCATTERING, AND ABSORPTION OF ATMOSPHERIC AEROSOL

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In this paper we discuss few-parameter models proposed for estimation of cross section of extinction, scattering, and absorption of light by atmospheric aerosol. Estimation of the accuracy of approximate formulas are presented for the case of polydisperse ensembles of spherical particles with quasi-Gaussian and gamma size distributions as well as for ensembles of randomly oriented spheroids with the refractive index $m_r = 1.5 + i 0.02$.

INTRODUCTION

To estimate effects of atmospheric aerosol on the processes of radiation propagation through the atmosphere, quantitative information on the scattering properties of aerosol particles is needed. Among great variety of aerosol particles of various origin certain contribution to the radiation processes is made by the particles of mineral origin which have nonspherical shapes and are characterized by complicated distribution by size, shape, and orientation.

Exact calculations for the ensembles of nonspherical particles require a complex mathematical means, they are effective for the axially symmetric particles with smooth surface, and limited by the range of particle size comparable to the wavelength of incident radiation¹⁻³; for ellipsoidal particles⁴ computer time increases by several orders as compared with this for spheroidal particles.⁵

This paper presents an approximate estimation of the cross section of extinction, scattering, and absorption for an elementary volume containing randomly oriented nonspherical particles. The data of an approximate estimation are compared with the results of the exact theory for polydisperse ensembles of spherical particles with the quasi-Gaussian and gamma size-distributions and randomly oriented spheroidal particles. Refractive index of the particles $m_r = 1.5 + i 0.02$ corresponds to the continental aerosol.⁶

Creation of few-parameter models is based on conformity of the initial and approximating ensembles. Some microstructural parameters of these ensembles are equal, for example, the volume, projection square, and particle surface which are averaged over an ensemble (for the convex particles equality of the projection squares means the equality of the surfaces⁷); in this case for the approximate estimation the simplest representatives of the class of approximating ensembles with the same microstructural parameters, for example, ensembles of polydisperse spherical particles are chosen.

1. FEW-PARAMETER MODELS. GENERAL APPROACH

Cross sections of extinction, scattering and absorption for a polydisperse ensemble of particles have the form

$$\langle C(m_r, \lambda) \rangle = \int_{r_{\min}}^{r_{\max}} C(m_r, \lambda, r) f(r) dr, \quad (1)$$

where $f(r)$ is the particle size-distribution density function; $[r_{\min}, r_{\max}]$ is the interval of change of dimensional parameters; λ is the wavelength of incident radiation. In the below discussion, the refractive index and wavelength are omitted in the expressions.

In the case of a polydisperse ensemble of nonspherical particles an integral of the type (1) has a more complicated form and it is necessary to integrate over particle orientations. Conventionally the integrals of the type (1) are estimated using the quadrature formulas ignoring microstructure of a suspension. Our purpose is to make a few-parameter estimation of integrals of the type (1) allowing for microstructure of a suspension and to derive corresponding quadrature formulas.

In the general case creation of mathematical models is based on a mathematical concept of equivalence ratio or "equality". When dealing with mathematical objects it is necessary to introduce a concept of equivalence or "equality". A relation between two mathematical objects ($a \overset{r}{\sim} b$) is named as the equivalence only when the following conditions are fulfilled⁸:

- 1) $a \overset{r}{\sim} a$ (reflectivity);
- 2) if $a \overset{r}{\sim} b$ means $b \overset{r}{\sim} a$ (symmetry);
- 3) if $a \overset{r}{\sim} b$ and $b \overset{r}{\sim} c$ mean $a \overset{r}{\sim} c$ (transitivity).

In this case all mathematical objects of the model are distributed among equivalence classes without common elements.

As a rule, parameters of mathematical models coincide with some major parameters of a real object. An equality relation of the model parameters determines the equivalence and thus divides all mathematical objects into nonoverlapping equivalence classes. In this case, within the limits of one of these classes, the objects can not be distinguished. Therefore, any representative of the class characterizes this class as a whole. Based on theoretical or other considerations, the simplest element can be taken for such a representative. The relation between the optical equivalence classes and equivalence classes being assigned using the ratio of parameter equality is of interest in the optics of disperse media.

The elementary scattering volume is assumed to contain randomly oriented particles and characterized by the following microstructural parameters averaged over an ensemble: the projection square ($\langle S \rangle$), volume ($\langle V \rangle$), the square of the projection square ($\langle S^2 \rangle$), and the square of the volume ($\langle V^2 \rangle$). The choice of these parameters is based on the following considerations:

- 1) for particles which are smaller than the wavelength the absorption cross section is proportional to $\langle V \rangle$ and the scattering cross section is proportional to $\langle V^2 \rangle$;
- 2) for large particles the cross sections of scattering and absorption are equal to $\langle S \rangle$ in the size region where the efficiency factors of scattering and absorption are equal to unity, the scattering intensity within small angles is proportional to $\langle S^2 \rangle$ in the Fraunhofer diffraction region.

An ensemble approximating the elementary volume is chosen among the simplest ones in the form of discrete distribution of spherical particles with weights which are considered as concentration coefficients. The cross sections of extinction, scattering, and absorption of the elementary volume are approximated by the sections of the discrete distribution of spherical particles. Let us consider the families of the few-parameter models produced by four microstructural parameters.

1.1. Single-parameter models

Each of the microstructural parameters mentioned above creates a single-parameter model which has one of its parameters equal to the elementary volume. In particular, the models of equivolume and equisurface spherical particle are among the single-parameter models.

1.2. Two-parameter models

A set of two-parameter models consists of six elements, i.e., a number of the elements equals to C_4^2 (a number of combinations of four elements taken two at a time). In this case the cross sections of extinction, scattering, and absorption are estimated proceeding from an equality of two microstructural parameters of the elementary volume and approximating ensemble consisting of spherical particles of the same size with some weighting factor:

$$\langle S \rangle, \langle V \rangle; \langle C \rangle = \frac{16 \langle S \rangle^3}{9 \pi \langle V \rangle^2} C(r_{ef}), \quad r_{ef} = \frac{3 \langle V \rangle}{4 \langle S \rangle}; \quad (2)$$

$$\langle S \rangle, \langle S^2 \rangle; \langle C \rangle = \frac{\langle S \rangle^2}{\langle S^2 \rangle} C(r_{ef}), \quad r_{ef} = \left(\frac{\langle S^2 \rangle}{\pi \langle S \rangle} \right)^{1/2}; \quad (3)$$

$$\langle S \rangle, \langle V^2 \rangle; \langle C \rangle = \left(\frac{16 \langle S \rangle^3}{9 \pi \langle V^2 \rangle} \right)^{1/2} C(r_{ef}),$$

$$r_{ef} = \left(\frac{9 \langle V^2 \rangle}{16 \pi \langle S \rangle} \right)^{1/4}; \quad (4)$$

$$\langle V \rangle, \langle S^2 \rangle; \langle C \rangle = \frac{81 \pi^2 \langle V \rangle^4}{256 \langle S^2 \rangle^3} C(r_{ef}), \quad r_{ef} = \frac{4 \langle S^2 \rangle}{3 \pi \langle V \rangle}; \quad (5)$$

$$\langle V \rangle, \langle V^2 \rangle; \langle C \rangle = \frac{\langle V \rangle^2}{\langle V^2 \rangle} C(r_{ef}), \quad r_{ef} = \left(\frac{3 \langle V^2 \rangle}{4 \pi \langle V \rangle} \right)^{1/3}; \quad (6)$$

$$\langle S^2 \rangle, \langle V^2 \rangle; \langle C \rangle = \frac{256 \langle S^2 \rangle^3}{81 \pi^2 \langle V^2 \rangle^2} C(r_{ef}),$$

$$r_{ef} = \left(\frac{9 \langle V^2 \rangle}{16 \pi \langle S^2 \rangle^{1/2}} \right)^{1/4}; \quad (7)$$

1.3. Three-parameter models

A number of three-parameter models being created by four microstructural parameters equals to $C_4^3 = 4$ (a number of combinations of four elements taken three at a time). There is a certain arbitrariness in choice of approximating ensemble, and, on the contrary to the two-parameter models, where a solution is unique, choice of the model having three microstructural parameters equal to the elementary volume is not unique.

Let us consider an estimation of integral of the type (1) in the form

$$\langle C \rangle = [p_1 C(r_{ef}^{(1)}) + p_2 C(r_{ef}^{(2)})] / 2, \quad (8)$$

where $r_{ef}^{(i)} = r_{ev} / p_i^{1/3}$, r_{ev} is the radius of a particle having an elementary volume $\langle V \rangle$. For this choice the automatically approximating ensemble has a volume equal to the elementary volume and weighting factors p_i can be found from the condition of equality of other two microstructural parameters.

$$\langle S \rangle, \langle V \rangle, \langle V^2 \rangle; \begin{cases} p_1^{1/3} + p_2^{1/3} = 2 \langle S \rangle / \pi r_{ev}^2; \\ p_1^{-1} + p_2^{-1} = 2 \langle V^2 \rangle / \langle V \rangle^2; \end{cases} \quad (9)$$

$$\langle S \rangle, \langle V \rangle, \langle S^2 \rangle; \begin{cases} p_1^{1/3} + p_2^{1/3} = 2 \langle S \rangle / \pi r_{ev}^2; \\ p_1^{-1/3} + p_2^{-1/3} = 2 \langle S^2 \rangle / \pi^2 r_{ev}^4; \end{cases} \quad (10)$$

$$\langle V \rangle, \langle S^2 \rangle, \langle V^2 \rangle; \begin{cases} p_1^{-1/3} + p_2^{-1/3} = 2 \langle S^2 \rangle / \pi^2 r_{ev}^4; \\ p_1^{-1} + p_2^{-1} = 2 \langle V^2 \rangle / \langle V \rangle^2; \end{cases} \quad (11)$$

For the next model $r_{ef}^{(i)} = r_{es} / p_i^{1/2}$, r_{es} is the radius of a spherical particle having the projection square $\langle S \rangle$

$$\langle S \rangle, \langle S^2 \rangle, \langle V^2 \rangle; \begin{cases} p_1^{-1} + p_2^{-1} = 2 \langle S^2 \rangle / \langle S \rangle^2; \\ p_1^{-2} + p_2^{-2} = 9 \langle V^2 \rangle / 8 \pi^2 r_{es}^6; \end{cases} \quad (12)$$

Each of these systems of equations has an analytical solution. However, for certain relationships between the microstructural parameters this solution can take negative values. In this case it is necessary to choose some other values of p_i , $r_{ef}^{(i)}$ assuming that the solution is positive and microstructure parameters are equal.

In the present paper we consider the few-parameter models only which admit the analytical determination of the parameters of approximating ensemble.

2. CROSS SECTIONS FOR SPHERICAL PARTICLES

Cross sections of extinction, scattering, and absorption for spherical particles are calculated by the formulas from Ref. 9:

$$C_{ext} = 2\pi/k^2 \sum_{n=1}^{\infty} (2n + 1) \operatorname{Re} (a_n + b_n);$$

$$C_{sca} = 2\pi/k^2 \sum_{n=1}^{\infty} (2n + 1) (|a_n|^2 + |b_n|^2); \tag{13}$$

$$C_{abs} = C_{ext} - C_{sca},$$

where $k = 2\pi/\lambda$ is the wave number; a_n and b_n are the Mie coefficients.⁹ The cross section of a ensemble of polydisperse particles is described by Eq. (1). Relative error of a few-parameter model for the estimation (1) is calculated by the formula

$$F = [(\langle C \rangle - \langle C \rangle (I)) / \langle C \rangle] 100\%, \tag{14}$$

where I is the number of the formula of a few-parameter model.

2.1. Quasi-Gaussian distribution

A shape of the distribution curve is similar to the curve for normal distribution, and it has the density function which differ from zero on a finite interval. Moreover, this distribution makes it possible to simulate nonsymmetric distributions.

The density function has the form¹⁰

$$f(r) = \begin{cases} (1 - z^2)^2 & \text{at } -1 \leq z \leq 1, \\ 0 & \text{at } |z| > 1, \end{cases} \tag{15}$$

$$z = c (r - r_0) / \Delta r_0, \quad c = 2(1 - 2^{-1/2})^{1/2}$$

(where r_0 is the modal or mean size for a symmetric distribution; Δ is the distribution full width at half maximum of the density function) and characterizes the distribution variance $\Delta \approx 3\sigma/r_0$, σ is the rms deviation. A symmetric distribution shape (15) is assigned by the distribution widths Δ_L and Δ_R on the left and on the right of the modal size, respectively. It should be noted that the normalizing constant for the distribution (15) has the form $15/8c / (a_0(\Delta_L + \Delta_R))$. For a symmetric distribution ($\Delta = \Delta_L = \Delta_R$) the values averaged over an ensemble have the form

$$\langle S \rangle = \pi r_0^2 (1 + \Delta^2 / 7c^2),$$

$$\langle V \rangle = (4\pi / 3) r_0^3 (1 + 3\Delta^2 / 7c^2),$$

$$\langle S^2 \rangle = \pi^2 r_0^4 (1 + 6\Delta^2 / 7c^2 + \Delta^4 / 21c^4), \tag{16}$$

$$\langle V^2 \rangle = (16\pi^2 / 9) r_0^6 (1 + 15\Delta^2 / 7c^2 + 5\Delta^4 / 7c^4 + 5\Delta^6 / 231c^6).$$

2.2. Two-parameter gamma-distribution

The density function for the gamma-distribution is written in the form¹¹

$$f(r) = \begin{cases} C r^\mu \exp(-\beta r), & r \geq 0, \quad \beta > 0, \quad \mu > -1, \\ 0, & r < 0, \end{cases} \tag{17}$$

where C is the normalizing constant equal to $\beta^{\mu+1} / \Gamma(\mu + 1)$; $\Gamma(\mu)$ is the gamma-function. The mean radius $r_0 = (\mu + 1)/\beta$, the parameter μ characterizes the distribution width, the relative rms deviation (relative to r_0) is $\sigma = (\mu + 1)^{-1/2}$, and the values averaged over an ensemble are as follows¹¹

$$\langle S \rangle = \pi ((\mu + 1) (\mu + 2) / \mu^2) r_0^2,$$

$$\langle V \rangle = (4\pi/3) ((\mu + 1) (\mu + 2) (\mu + 3) / \mu^3) r_0^3,$$

$$\langle S^2 \rangle = \pi^2 ((\mu + 1) (\mu + 2) (\mu + 3) (\mu + 4) / \mu^4) r_0^4, \tag{18}$$

$$\langle V^2 \rangle = (16\pi^2 / 9) ((\mu + 1) (\mu + 2) (\mu + 3) (\mu + 4) (\mu + 5) \times (\mu + 6) / \mu^6) r_0^6.$$

3. THE ABSORPTION CROSS SECTION OF THE RANDOMLY ORIENTED SPHEROIDAL PARTICLES

To calculate the absorption cross sections for the suspension of randomly oriented spheroidal particles the analytical formulas for the cross section of absorption and scattering expressed in terms of the elements of the T -matrix were used^{12,13}

$$\langle C_{ext} \rangle = -\frac{2\pi}{k^2} \operatorname{Re} \sum_{m=0}^{\infty} \sum_{n=\max(m,1)}^{\infty} (2 - \delta_{m0}) (t_{omn, omn}^{11} + t_{emn, emn}^{22}),$$

$$\langle C_{sca} \rangle = \frac{2\pi}{k^2} \sum_{n=1}^{\infty} \sum_{n'=1}^{\infty} \sum_{m=\min(n, n')}^{m=\min(n, n')} (2 - \delta_{m0}) D_{mn} D_{mn'}^{-1} \times$$

$$\times (|t_{emn, emn}^{11}|^2 + |t_{emn, omn}^{12}|^2 + |t_{omn, emn}^{21}|^2 + |t_{omn, omn}^{22}|^2), \tag{19}$$

$$\langle C_{abs} \rangle = \langle C_{ext} \rangle - \langle C_{sca} \rangle.$$

The spheroid surface in the spherical coordinate system obeys the following equation:

$$r(\theta, \varphi) = a (\sin^2\theta + (a^2 / b^2) \cos^2\theta)^{-1/2}, \tag{20}$$

where θ and φ are the zenith and azimuth angles; b is the vertical half-axis of rotation; a is the horizontal half-axis. The shape parameter ϵ is determined as a ratio of the larger parameter to the smaller one

$$\epsilon = \begin{cases} b/a & \text{for the stretched spheroids,} \\ a/b & \text{for the compressed spheroids.} \end{cases} \tag{21}$$

By varying the shape parameter we can simulate the change of a particle shape in a wide range from the stick-shaped to disk-shaped.

For a monodisperse ensemble of randomly oriented spheroidal particles the values averaged over the ensemble have the form

$$\langle S \rangle = \pi/2 [a^2 + ab (\arcsin e/e)] \text{ for the stretched spheroids,}$$

$$\langle S \rangle = \pi/2 [a^2 + (b^2/2e) \ln((1+e)/(1-e))] \text{ for the compressed spheroids,}$$

$$\langle V \rangle = 4\pi/3 ba^2, \tag{22}$$

$$\langle S^2 \rangle = \pi^2 a^2 b^2 [(\epsilon^{-2} + 2)/3] \text{ for the stretched spheroids,}$$

$$\langle S^2 \rangle = \pi^2 a^2 b^2 [(\epsilon^2 + 2)/3] \text{ for the compressed spheroids,}$$

$$\langle V^2 \rangle = (16\pi^2/9) b^2 a^4,$$

where $e = (\epsilon^2 - 1)^{1/2}/\epsilon$.

3.1. Optical equivalence of an ensemble of randomly oriented spheroidal particles and a polydisperse ensemble of spherical particles

In Ref. 14 it was shown in the Rayleigh–Gans–Debye approximation¹⁵ that a monodisperse ensemble of randomly oriented spheroids is optically equivalent to a polydisperse ensemble of spherical particles with the weighting function

$$f(r) = \begin{cases} a^4 b e^{-1} (r^2 - a^2)^{-1/2} r^{-5}, & a \leq r \leq b, \\ \text{for the stretched spheroids,} \\ a^3 b^2 e^{-1} (a^2 - r^2)^{-1/2} r^{-5}, & b \leq r \leq a, \\ \text{for the compressed spheroids.} \end{cases} \quad (23)$$

In the anomalous diffraction approximation¹⁵ a direct test shows that the cross sections of extinction, scattering, and absorption of a monodisperse ensemble of randomly oriented spheroids are identical to corresponding cross sections of a polydisperse ensemble of spherical particles with the weighting function (23). Note, also that the volume, projection squares, surface squares, and squares of the volumes, which are averaged over an ensemble of the suspension mentioned above, are the same. Therefore, the estimations of these cross sections by the few-parameter models (2), (4), (6), and (9) are the same too. Under the conditions of anomalous diffraction¹⁵ for the calculations of the cross sections of extinction, scattering, and absorption of randomly oriented spheroids the above noted optical equivalence ought to be used changing the calculations by *T*-matrix method to simpler ones using Eq. (23) and the Mie theory.

4. CALCULATIONAL RESULTS

In this section we present some results of calculations of the cross sections of extinction, scattering, and absorption by approximation formulas (2), (8)–(10), and also using the exact theory by the formulas (1), (19), and (23). The relative error is estimated by formula (14).

TABLE I. The upper boundary of the relative error (9) and (10) for the quasi-Gaussian distribution.

ρ_0	Δ			
	0.1	0.2	0.5	1
0.1	< 0.1	< 0.1	0.7	3.3
1	< 0.1	< 0.1	0.5	2.0
2	< 0.1	< 0.1	3.4	1.3
5	1.3	5.4	5.8	11
10	2.0	2.5	4.8	7.8
50	0.1	0.4	1.0	0.6
100	0.1	< 0.1	0.1	0.2
200	0.05	0.1	0.1	0.1

Table I presents the upper boundary of the absolute value of the relative error (14) for the three-parameter models (9) and (10) simultaneously for the cross sections of extinction, scattering, and absorption as a function of the modal diffraction parameter ρ_0 ($\rho_0 = k r_0$) and distribution width Δ . The maximum error is not more than 11%, and it characterizes the particles which have the mean size comparable with the wavelength of incident radiation in the visible region. The maximum error for the two-parameter model (2) frequently used in the optics of biological media¹⁶ is

not more than 27% and, for the scattering cross sections in the region of small particles, compared to the wavelength of incident radiation. It is connected with the difference between the microstructural parameter $\langle V^2 \rangle$ of the approximated and approximating ensembles. In this case the error decreases but the size of particles ($\rho_0 > 2$) increases and has the order presented in Table I.

Table II presents the relative error for three-parameter model (9) when estimating the cross sections of extinction (the values in the numerator) and scattering (the values in the denominator) for a polydisperse ensemble of spherical particles and gamma-distribution as a function of the parameters μ and β .

TABLE II. The relative error (9) for gamma-distribution.

μ	β					
	0.5	1	2	5	10	20
0.5	-20/-26	13/15	5.5/5.8	-6.6/-8.4	-2.2/-4.3	-0.1/-0.1
1	-6.5/-9.5	1.7/2.6	6.7/6.9	-4.3/-5.0	-2.8/-5.0	-0.1/-0.1
2	16/21	-13/-15	4.9/5.9	-5.7/-5.9	-3.8/-6.0	-0.2/-0.3
5	-4.8/-7.3	-4.9/-6.1	3.6/3.8	2.8/2.7	-2.6/-3.5	-0.5/-0.9
10	3.2/5.4	-9.1/-13	-13/-15	-2.1/-2.2	-3.5/-3.5	-1.3/-1.9
20	0.6/1.1	0.1/0.3	2.4/4.3	-0.2/-0.4	-2.6/-2.6	0.9/1.1

Note, that the estimations (9) and (10) are close, and they differ by less than 5%. The absolute value of the relative error (9) and (10) is less than 5% when estimating the absorption cross section. For the two-parameter model (2) this difference is 13%. At the same time the latter model can not be used for estimation of the cross sections of extinction and scattering (1), and corresponding values differ by several times.

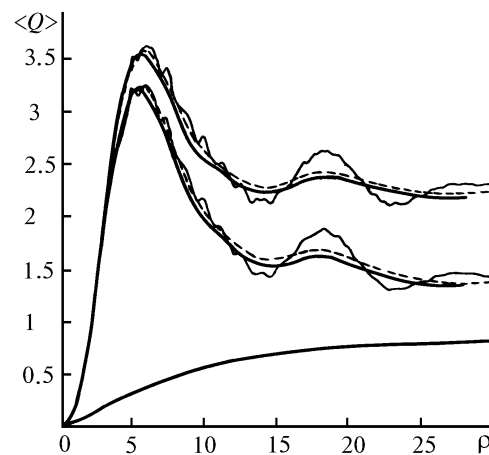


FIG. 1. The efficiency factors of extinction, scattering, and absorption of randomly oriented compressed spheroids ($\epsilon = 2$) which are calculated by the formulas (19) (heavy solid curve), (23) (dashed curve), (8) and (9) (solid curve); the lower solid curve is for the formulas (19), (23), (8), and (9) as functions of the maximum diffractive parameter ρ .

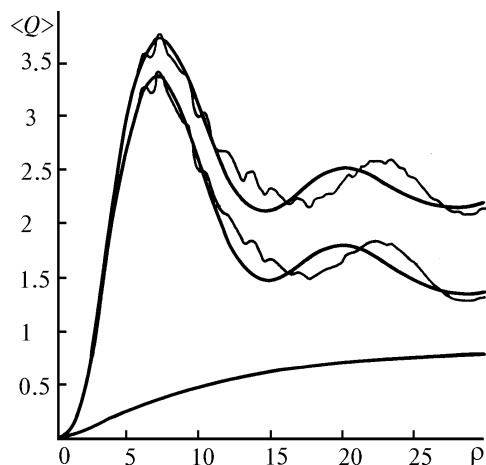


FIG. 2. The same as in Fig. 1 but for stretched spheroids ($\epsilon = 2$).

Figures 1 and 2 present the efficiency factors of extinction, scattering, and absorption

$$\begin{aligned} \langle Q_{\text{ext}} \rangle &= \langle C_{\text{ext}} \rangle / \langle S \rangle; \quad \langle Q_{\text{scat}} \rangle = \langle C_{\text{scat}} \rangle / \langle S \rangle; \\ \langle Q_{\text{abs}} \rangle &= \langle Q_{\text{ext}} \rangle - \langle Q_{\text{scat}} \rangle, \end{aligned} \quad (24)$$

calculated by the exact formulas (19) for randomly oriented spheroidal particles, by the Mie formulas for the equivalent polydisperse suspension of spherical particles with the distribution (23), and with the use of the three-parameter model (9) as a function of the maximum spheroid diffraction parameter ρ .

The estimation of the cross sections of extinction, scattering, and absorption by formula (23) coincides with the calculations by the exact theory. It should be expected that the error will be decrease with increasing size because of fulfilled conditions of the anomalous diffraction approximation ($\rho \gg 1$) under which ensemble of randomly oriented spheroids is equivalent to the polydisperse ensemble of spherical particles (23) what is confirmed by direct calculations.

DISCUSSION

In the cases considered the cross sections of extinction, scattering, and absorption are mainly determined by four microstructure parameters of an elementary volume. It is confirmed by the efficiency of the few-parameter estimations. The equivalence classes which are assigned by an equality of the microstructure parameters can be considered as the classes of optical equivalence with a certain error. In this case the formulation can be possible and solution can be obtained for the inverse problems on the classes of optical equivalence and estimation of the microstructure parameters of an elementary volume.

Use of the few-parameters estimations allows us to simplify essentially the calculation of the cross sections of

extinction, scattering, and absorption for the elementary volume containing randomly oriented particles of different size.

The use of optical equivalence of randomly oriented spheroidal and spherical particles (23) makes it possible to study the optical properties of nonspherical particles in the whole range of the dimensional parameters in spite of the restrictions of T -matrix method.¹⁻³

It should be noted in conclusion that all the above mentioned is justified in the whole measure for the hydrosol particles of ocean suspension, biological suspensions, suspensions of erythrocytes and leukocytes, in particular, the absorption of the biological suspension is determined by $\langle S \rangle$, $\langle V \rangle$, and absorption by the cell substance.¹⁷

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