Optical classification of isotropic ensembles of "soft" ellipsoidal particles

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We propose a classification of isotropic ensembles of "soft" ellipsoidal particles based on optical equivalence approach. The results are illustrated by numerically calculated angular dependence of scattering phase matrix elements of randomly oriented "soft" ellipsoidal particles.

Introduction

The proposed classification of isotropic ensembles of "soft" ellipsoidal particles is based on the optical equivalence of the ensembles of "soft" randomly oriented ellipsoidal particles[1] in the Rayleigh–Gans–Debye approximation.[2] According to Ref. 1, randomly oriented ellipsoidal particles are optically equivalent to three polydisperse ensembles of randomly oriented spheroidal particles. It is shown that the optical equivalence has more extensive range of applicability, if calculations of the integral kernel are being carried out using strict methods, such as Mie theory and the method of T-matrices. The working hypothesis is stated and examined, that the equivalent ensembles of particles have close values of the optical characteristics. The correctness of such a classification is confirmed by calculations of the angular dependence of the scattering phase matrix elements using strict methods. Solution of the inverse problems on the classes of equivalence is discussed.

1. Optical equivalence

Following Ref. 1, let us consider the single scattering of light by an ensemble of independent randomly oriented ellipsoidal particles in the Rayleigh–Gans–Debye approximation (RGD). In this case the randomly oriented ellipsoidal particles are optically equivalent:

1) to three different (due to permutation of a, b, and c) polydisperse ensembles of spheroidal particles:

$$< Z_{11}(\theta;a,b,c) > = \frac{2a^2b^2}{\pi}\int\limits_a^b \mathrm{d}\hat{a} < Z_{11}(\theta;\hat{a},\hat{a},c) > \times$$

$$\times \frac{\hat{a}^{-3}}{\sqrt{(b^2-\hat{a}^2)(\hat{a}^2-a^2)}}, \qquad (1)$$

where a, b, c are the half-axes of an ellipsoidal particle; $< Z_{11}(\theta;a,b,c) >$ is the intensity of the scattered radiation by randomly oriented ellipsoidal particles at nonpolarized incident radiation of unit intensity, brackets < > denote the averaging over the ensemble;

2) to polydisperse ensemble of spherical particles with the weighting function invariant relative to the permutation of a, b, and c:

$$\rho_{\mathrm{eq}}(r) = \Theta(c-r)\Theta(r-a)\frac{2a^2b^2c^2}{\pi r^5}\times$$

$$\times \int\limits_a^{\min(r,b)} \mathrm{d}\hat{a}\,\frac{\hat{a}}{\sqrt{-(b^2-\hat{a}^2)(a^2-\hat{a}^2)(c^2-\hat{a}^2)(r^2-\hat{a}^2)}}, \qquad (2)$$

here $\Theta(x)$ is the Heaviside function.

After the corresponding substitution $x = \hat{a}^2$, the obtained integral is reduced to the complete ellipsoid integral of the first kind in the Legendre form.[3]

Direct test shows that all the considered equivalent ensembles of particles have equal ensemble average: 1) volumes $<V>$; 2) areas of projections $<S>$ onto the plane, orthogonal to the direction of the radiation incidence, and, as the particles are convex bodies, the surface areas equal to $4<S>$ (Ref. 4); 3) squares of the volumes $<V^2>$.

2. Classification of isotropic ensembles of optically "soft" ellipsoidal particles

The problem is considered in this section of determination of the most significant parameters of microstructure of the ensemble of particles determining the angular dependence of the elements of the scattering phase matrix aimed at classification of isotropic ensembles of optically "soft" randomly oriented ellipsoidal particles.

The authors of Ref. 5 stated that the principal parameters of the spherical particle size distribution determining the angular dependence of the elements of the scattering phase matrix are second, third, and fourth central moments of the distribution, but the type of the distribution is not very important. Following such an approach, one can perform the classification of polydisperse ensembles of spherical

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particles by introducing the equivalence ratio as the equality of the aforementioned moments of the distribution. Then the ensembles of particles with three equal moments of the distribution and, in general case, with different size distributions, fall to the same class. However, the "bridge" connecting spherical and non-spherical particles is absent.

As the present investigation shows, second, third, and sixth central moments of the distribution are the most significant for the optically "soft" particles. Let us consider the mean area of projection of particle to the plane perpendicular to the direction of the radiation incidence, mean volume, and mean square of the volume to the means of the second, third, and sixth moments of the distribution of isotropic ensembles of non-spherical particles.

Let us call these parameters the parameters of microstructure of the suspension (ensemble) of particles, and they will be used for classification of isotropic ensembles of ellipsoidal particles with the relative refractive indices corresponding to biological particles.

The equality of three aforementioned parameters of the microstructure is the equivalence ratio and divides all the isotropic ensembles to the equivalence classes. Let us call the isotropic ensembles of particles belonging to the same class, equivalent.

Let us consider the power-law size distribution as a representative characterizing the class. Let the quantities:

$$< S > = \int_a^b \pi r^2 f(r)\mathrm{d}r, \quad < V > = \int_a^b \frac{4\pi}{3} r^3 f(r)\mathrm{d}r,$$

$$< V^2 > = \int_a^b \frac{16\pi^2}{9} r^6 f(r)\mathrm{d}r$$

be the second, third, and sixth moments of the particle size distribution with the density $f(r)$, respectively, $[a, b]$ is the range of variation of the spherical particle size.

If $<S>$, $<V>$, and $<V^2>$ are known, one can find the parameters of the power-law size distribution of particles with the density function of the form:

$$f(r) = K r^{-5}, \; r_{\min} \le r \le r_{\max}. \tag{3}$$

in the explicit form.

Let us note that this function does not satisfy the normalizing condition, the coefficient K is considered as the concentration of particles.

In considering the distributions different from the aforementioned power-law size distribution, let us find the moments of the distribution and the corresponding power-law size distribution with the same equal three moments, and compare the calculated results on the optical characteristics of these ensembles.

Let us consider optically "soft" particles, i.e., the particles (with the relative refractive index close to 1), which at random orientation have the scattering phase matrix with four independent elements, i.e., the same structure of the scattering phase matrix as that of spherical particles.

To meet the second condition, it is necessary and sufficient that the values of the normalized elements of the scattering phase matrix F_{22} and F_{11} differ insignificantly, while the consequence of the inequality $|F_{33} - F_{44}| \le |F_{11} - F_{22}|$ and the normalized elements of the scattering phase matrix F_{33} and F_{44} also insignificantly differ from each other.

In this section we consider the optical characteristics of the equivalent randomly oriented ensembles of ellipsoidal particles, namely, three different polydisperse ensembles of spheroidal particles (1) and two polydisperse ensembles of spherical particles with the weighting function (2) and the equivalent power-law size distribution (3). Formally, there are 6 = 3! permutations in the set of three half-axes (a, b, c), however, calculations according to Eq. (1) depend on the value at the third place and do not depend on permutations of two first numbers. Thus, we have three different equivalent polydisperse ensembles of spheroidal particles. These are the polydisperse elongated particles ($a < c$, $b < c$), oblate particles ($c < a$, $c < b$), and the ensemble containing oblate and elongated particles ($a < c < b$ or $b < c < a$). One should note that the permutation (or the change of the coordinate axes) of the size of the half-axes does not affect the result, but the choice of the equivalent polydisperse ensemble of spheroidal particles depends on it.

All the aforementioned ensembles of particles are equivalent, and the optical properties of each of them can be considered as an estimate of the optical characteristics of the ellipsoidal particles.

Subsequent calculations were carried out for the relative refractive index of particles equal to 1.05. An ellipsoidal particle is characterized by the size of the half-axes with respect to the wavelength of the incident radiation:

$$\rho_a = \frac{2\pi a}{\lambda}, \; \rho_b = \frac{2\pi b}{\lambda}, \; \rho_c = \frac{2\pi c}{\lambda}.$$

Figures 1–4 show the angular dependence of the elements of the normalized scattering phase matrices of five aforementioned equivalent ensembles of particles enumerated as follows: (*1*) polydisperse elongated and (*2*) oblate spheroids; (*3*) ensemble consisting of elongated and oblate spheroids; (*4*) polydisperse ensemble of spherical particles (Eq. (2)), and (*5*) equivalent ensemble of spherical particles with the power-law size distribution. The representatives of this class of equivalence are corresponding randomly oriented ellipsoidal particles.

The figures presented confirm the working hypothesis on the significance of the three moments of the size distribution determining the angular dependence of the scattering phase function of "soft" ellipsoidal particles. The agreement between the results obtained for five different distributions also makes it possible to assume that the true values of the scattering phase function of ellipsoidal particles agree with the calculated results.

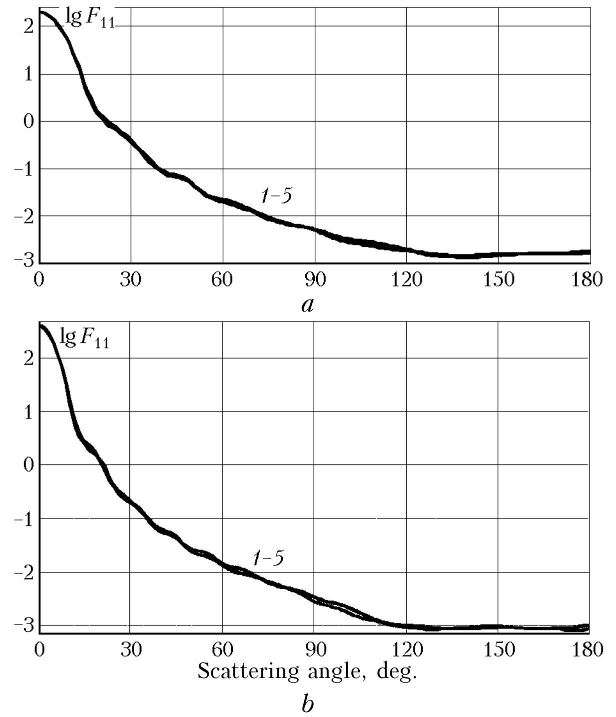

Fig. 1. Angular dependence of the element F_{11} of the scattering phase matrix of the equivalent ensembles of particles. The values are presented on the logarithmic scale. $\rho_a = 10$, $\rho_b = 15$, $\rho_c = 20$ (*a*), $\rho_a = 150$, $\rho_b = 20$, $\rho_c = 30$ (*b*). Enumeration is explained in text.

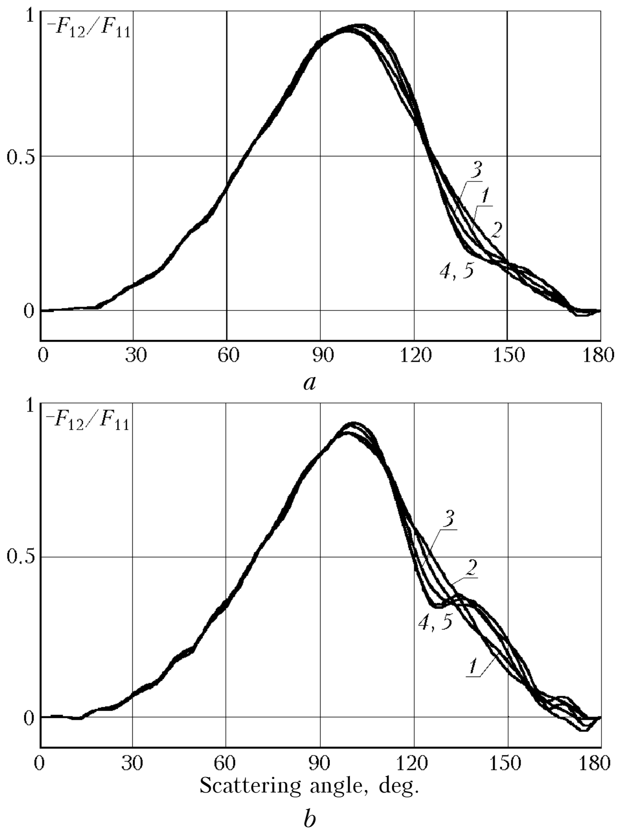

Fig. 2. The same as in Fig. 1 for angular dependence of the linear polarization $-F_{12}/F_{11}$.

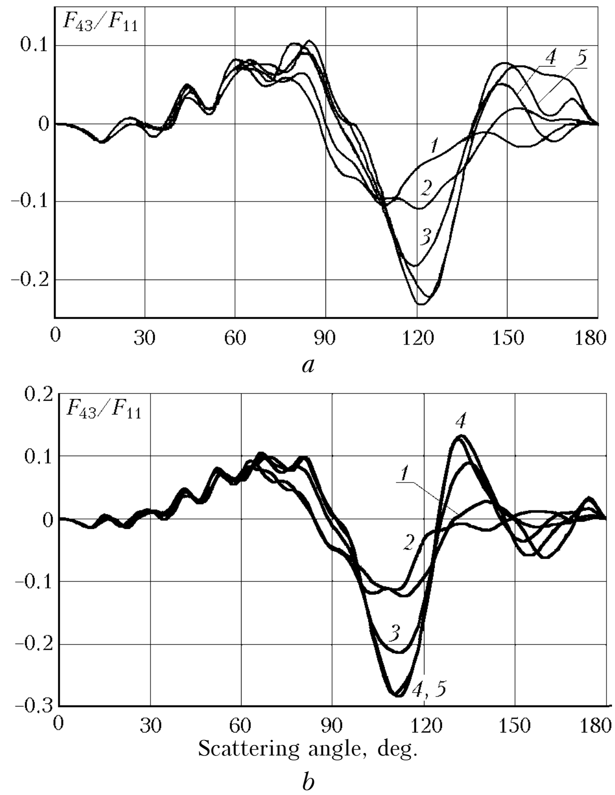

Fig. 3. The same as in Fig. 1 for angular dependence of the ratio F_{43}/F_{11}.

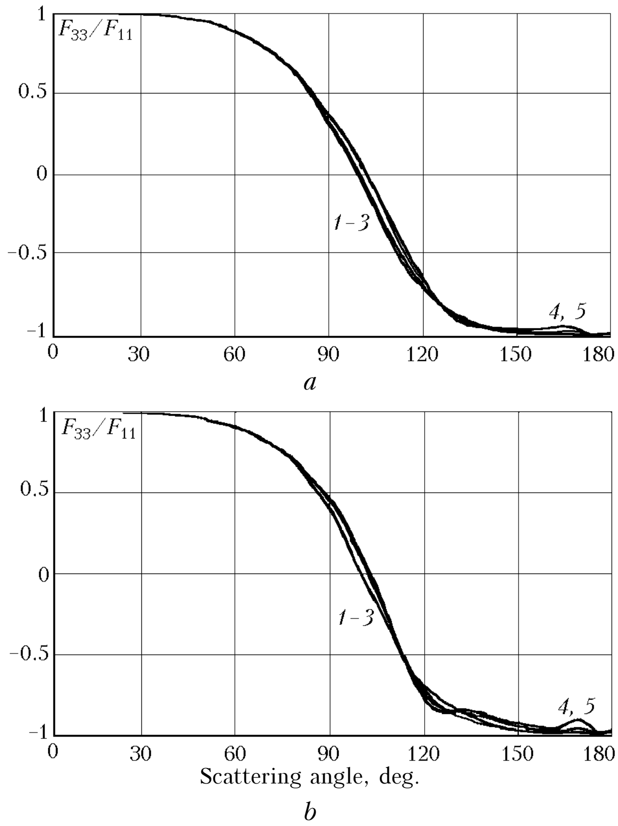

Fig. 4. The same as in Fig. 1 for angular dependence of the ratio F_{33}/F_{11}.

Let us note that such a coincidence of the angular dependence of the scattering phase function is possible only for the set of ellipsoidal particles, because spheroidal and ellipsoidal particles can be obtained from a spherical particle by means of a linear transformation. At the same time, such a classification of cylindrical particles does not lead to analogous results; the scattering phase function of these particles has local extrema in contrast to smooth scattering phase functions of spheroidal particles.

The numerically calculated results make it possible to draw the conclusion, important from the standpoint of practical applications. Under conditions of single scattering and in the absence of the particle orientation the solution of some inverse problems of optics of biological suspensions of cells is possible on the classes of equivalence. For the randomly oriented biological particles, when the particles of different shape belong to the same class, given the relative refractive index is known, one can determine only two values from inverting the scattering phase function $\langle V\rangle/\langle S\rangle$ and $\langle V^2\rangle/\langle S\rangle$. To do this, it is necessary to preliminarily determine the class of equivalence. The representatives of the class are the polydisperse ensembles of spherical particles with power-law size distribution with the parameters $r_{\min}$, $r_{\max}$. The third parameter K is not important, because the scattering phase function in the case of single scattering does not depend on the number density of particles. If the angular dependence of the scattering phase function is "sensitive" to the relative refractive index of particles, i.e., the conditions of the RGD approximation are not met the refractive index of particles can be estimated.

Subsequent differentiation of particles is possible by use of the polarization characteristics and in the presence of the *a priori* data on the particle shape.

Conclusion

The effective method is proposed for calculation of the optical characteristics of "soft" randomly oriented ellipsoidal particles based on the optical equivalence of polydisperse ensembles of randomly oriented ellipsoidal and spherical particles in the Rayleigh–Gans–Debye and anomaly diffraction approximations,[1,6] which allows one to reduce the estimation the optical properties of "soft" ellipsoidal particles to a more simple calculation using the Mie theory.

Optical equivalence has a wider area of application, if the kernels of the integral operators have been calculated by the exact methods, such as Mie theory or the method of T-matrix.[7]

Optical classification of isotropic ensembles of optically "soft" ellipsoidal particles upon microstructure parameters $\langle S\rangle$, $\langle V\rangle$, $\langle V^2\rangle$ is proposed. It is necessary to note that there is a freedom in choosing the representatives of the class of equivalence, the optical characteristics of which are the estimates of the class as a whole. In this case the choice of the equivalent power-law size distribution is dictated by mathematical ideas – the parameters of this distribution are determined analytically in the explicit form. Based on, for example, physical ideas or the *a priori* data, the equivalent distribution can be uniform, lognormal, etc. The usefulness of such an approach is confirmed by numerical calculations of the angular dependence of the elements of the scattering phase matrix of a polydisperse ensemble of randomly oriented spheroidal particles by the exact methods.

The necessary condition of the classification is that the scattering phase matrix should have four independent elements, i.e., the linear depolarization of the scattered radiation should be close to zero.

The proposed classification of the ensembles of spherical particles is also correct in the case when the relative refractive index corresponds to the aerosols of mineral origin without restrictions of the particle size.

More detailed and systematic investigations are necessary for determination of the range of correct application of the classification of ensembles of non-spherical particles, including divisions into subclasses taking into account the polarization characteristics.

The classification allows one to parameterize the inverse problem and to reduce its solution in the range of acceptance of the working hypothesis to determination of the class of equivalence.

Calculations of the elements of the scattering phase matrix were carried out using the method of T-matrices[7] by the formulas and algorithms proposed in Ref. 8.

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