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Influence of spheroidal particle shape on particle size characterization by multi-wavelength light extinction method

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ABSTRACT

In order to improve the applicability, spheroidal particles are substituted for the spherical particles in the traditional modeling of multi-wavelength light extinction method. The extinction coefficient and extinction spectrum are calculated at different axial ratios under the configuration of the generalized Lorenz-Mie theory. Combining extinction spectrum with regularization inversion algorithm, the influence of particle shape on particle size characterization by multi-wavelength light extinction method is discussed in detail. That is, the arithmetic statistical average and stochastic incidence statistics are employed simultaneously to calculate the extinction spectrum in the visible light range and carry out a validation for submicron particles with a refractive index of 1.33. The corresponding comparison between inversed particle sizes and those of spherical particles yields the resulting inversion deviation within 10% for mono-sized spheroids and 20% for a polydisperse particle system with an axial ratio less than 3, but then the relative deviation may increase significantly with the axial ratio, even up to 50%. It can be seen that for particles of equivalent volume, the spheroidal shape exerts a great influence on particle size characterization, and the assumption of spheroidal particle would become crucial in the cases of axial ratio above 3.

Introduction

Particulate matter is widely used in the industrial processes, such as coal or liquid fuel combustion, crystallization, and food production, where particle size distribution is directly related to the technical process, product quality, energy consumption, and the safety of production. Therefore, the demand of high accuracy particle size characterization method has become increasingly urged. Accordingly, quite a few methods have been developed in recent years, from ultrasonic spectrometry method, image method to light scattering method. The latter is usually characterized by fast response, high accuracy and non-invasion [1], and subdivided into various techniques. Among them, light extinction method (LEM) has shown its advantages as it is simple to use and its cost is low [2].

In the early study of light extinction method, the monochromatic light has been frequently used for measuring the colloidal concentration. However, it has a very obvious drawback that the measurement results are prone to multi-valued, limiting the size upper range of 1 ~ 2 μm or less. Instead of using monochromatic light, the multi-wavelength extinction method presents a spectra analysis-based technique to determine the particle size distribution and concentration simultaneously. It also demonstrates higher measurement reliability, and wider measurement range, and thus shows great potential in particle size measurement [3]. Moreover, it can be applied in the aerosol size distribution [4], city dust measurement [5], and gas-solid two-phase flow field [6] when it is combined with imaging method. Depending on the wavelength range of light beam, the measurable particle typically ranges from submicron to several microns.

The Lorenz–Mie theory is usually used to explain the phenomenon of light scattering by particles. Thus, the particle size distribution in the laboratory is obtained by analyzing multi-wavelength extinction spectra which contains two implicit conditions: spherical particles are assumed for physical modeling, and transmitted light rather than scattered light information is used for particle characterization. However, in an actual application, the size distribution and the shape of the particle cannot be known in advance, which may bring great difficulties and errors to the inversion of particle size distribution [7]. For example, the ideal spherical particles are very rare no matter in natural environment or in industrial products. Obviously, a large number of irregular particles have brought great difficulties to the mathematical description, which also involves almost all kinds of light scattering methods. Fortunately, the spheroidal particle assumption can be used to

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seek a balance between over-simplified shape descriptions and mathematical modeling difficulties. In modeling, it can undoubtedly provide a more rational characterization for the actual particles with various aspect ratios as a flexible shape adjustment parameter, provided that the mathematical modeling and solution can be implemented.

The investigation of light scattering of spheroidal particles has yielded many encouraging achievements. Asano et al. [8] studied the scattering of spheroidal particles on plane waves by separating variable method, proposing a theoretical method to deal with the boundary conditions of the electromagnetic field of spheroidal particles, which was verified to be better in solving the problem of boundary condition. Schulz [9–11] studied the scattering problem of spheroidal particles by using T-matrix method in the field of plane beam incidence. In the subsequent study, Han [12] improved the derivation of boundary conditions and corrected the error parameters in Asano's literatures. Xu et al. [13–22] further investigated the scattering of beams of arbitrary shape, position, and incident angle by the homogeneous spheroid particles. These thorough investigations make the possibility emerge for the calculation of spheroid scattering parameters and even for the further application of extinction spectrum.

On this basis, we propose a modeling method based on the light extinction cross-section of spheroidal particles and explore the influence of shape factors on light extinction spectrum. The contents of this paper are organized as follows: in Section "Principle", the analytical expression of light extinction cross-section and light extinction spectra are derived by using Lambert-Beer's law, as well as the introduction to spheroid coordinate and key scattering parameters calculation. Section "Numerical results of extinction characteristics" is devoted to calculating the effect of the change of the equivalent volume spherical particle axial ratio and discussing the influence of the incident azimuth angle on the light extinction spectrum. In Section "Inversion and results", we obtain the particle size information in the extinction spectra via using the regularization inverse algorithm. The effect of the axis ration, equivalent volume and incident angle on the particle size measurement are consequently studied by comparing inversed sizes with the given nominal ones.

Principle

Light extinction method

By measuring the extinction spectra of light beam through the discrete particles medium, the particle size distribution is obtained by inversion of the spectral information. The core of the theory is the Lambert-Beer law. Fig. 1 shows that when a collimated incident monochromatic light beam with the strength I_0 and wavelength λ, whose diameter is greatly larger than that of measured particles to meet the conditions of static light scattering, goes through the medium containing numerous spatially stochastically distributed particles, the intensity of the transmitted light due to the scattering and absorption by particles is expressed as follows:

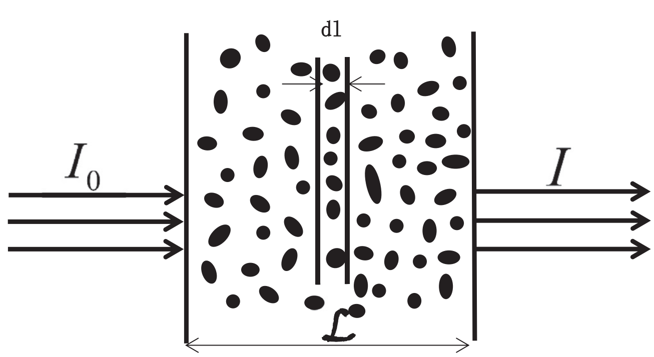

Fig. 1. Schematic diagram of the principle of light extinction method.

$$I = I_0 \exp(-\tau L) \tag{1}$$

where, L is the light path, and τ is the turbidity. If the light scattering of each particle satisfies the non-correlation single scattering, the turbidity of a single scattering particle system in the unit volume is

$$\tau = NC_{ext} = N\frac{\pi}{4}D^2Q_{ext} \tag{2}$$

where, the extinction cross-section C_{ext} and the extinction coefficient Q_{ext} are the functions of the wavelength λ of the incident light, the diameter of the particles D and the refractive index of the particles relative to the surrounding medium m. N indicates the particle number in the unit volume. Substituting Eq. (2) into Eq. (1)

$$\ln\left(\frac{I}{I_0}\right) = -NLC_{ext} \tag{3}$$

where, the ratio I/I_0 is the transmittance of Light. Obviously, light extinction can be expressed by $1-I/I_0$. The Lorenz–Mie scattering theory, in which the spherical particles are usually assumed, is frequently used to analyze light scattering phenomena and to determine extinction characteristic parameters such as extinction cross-section. However, as shown in Fig. 1, the spherical and non-spherical particles may coexist, the Lorenz–Mie theory and spherical particles assumption would then yield errors, but the degree of influence needs to be investigated and quantified if the light extinction method is expected to be well applied in a real particle system. It should be pointed out that a common simplification operation for spheroids is to used here to define the equivalent size as the diameter of a sphere with the same volume. Then the modeling method based on the extinction cross-section of spheroid particles can be implemented during exploring the influence of shape factors on extinction spectrum. The light extinction characteristic parameters based on the spheroid assumption will be introduced in the next section.

Spectral calculation of spheroidal particles

Spheroidal coordinates

In order to discuss the shape features of spheroidal particles, the spheroidal coordinates are defined first with the rotation around the oval major axis to form the coordinate system and then the ellipsoidal surface, in which, a point on the spheroid is defined by the angular coordinates η, the radial coordinates r and the azimuth coordinates φ. Their ranges are defined as

$$-1 \leqslant \eta \leqslant 1, \quad 1 \leqslant r \leqslant \infty, \quad 0 \leqslant \varphi \leqslant 2\pi \tag{4}$$

Define f as the half-focal length of the spheroid, i.e. the distance between the two focal points of a spheroid.

$$f = a\left[1-\left(\frac{b}{a}\right)^2\right]^{\frac{1}{2}} \tag{5}$$

where, a represents the half-length axis of a rotating spheroid, and b represents the half shaft of the rotating spheroid. Obviously, when f tends to be 0, that is, the focal points of the rotating spheroid coincide, the spheroidal coordinates degenerate into the spherical coordinate system.

Calculation of extinction cross-section, coefficient and spectral

When plane waves take the polarization angle $\phi = 0°$ and the incident angle ξ, the extinction cross-section and extinction coefficient of a single prolate spheroidal particle can be written as follows:

$$C_{ext}(\xi) = -\frac{\lambda}{\pi}\mathrm{Re}\sum_{m,n}[\alpha_{mn}\cdot\sigma_{mn}(\xi) + \beta_{mn}\cdot\chi_{mn}(\xi)] \tag{6}$$

$$Q_{ext}(\xi) = C_{ext}(\xi)/G(\xi) \tag{7}$$

$$G(\xi) = \frac{\pi ab^2}{(a^2\cdot\cos^2\xi + b^2\sin^2\xi)^{1/2}} \tag{8}$$

The extinction cross-section C_{ext} (or extinction coefficient Q_{ext}) is the function of spheroid dimension parameter C, $C = 2\pi f/\lambda$, spheroid relative refractive index m and incident angle ξ, where Re represents the real part, α_{mn}, β_{mn} representing the scattering coefficient, σ_{mn}, χ_{mn} representing the infield coefficient. The calculation can be carried out under the generalized Lorenz-Mie theory (GLMT), and more detailed descriptions can be traced back to Xu's work [13]. Further, the extinction section can be computed by substituting the extinction cross-section into Eq. (3). It must be noted that the direction of the beam with respect to particles is actually stochastic rather than fixed. Li [23] has proposed a calculation mode of the stochastic-orientation clustering particles extinction section on the basis of the statistical average method in which the arithmetic mean of the corresponding extinction section was computed when incident angle ξ varies evenly from 0 to 180 degrees. Meanwhile, the arithmetic incidence algorithm is proposed by authors here, i.e. randomly taking 500 different angles incident on spheroidal particles in a certain volume, calculating the corresponding extinction section, and obtaining the final extinction section on average. As shown in Eq. (9), the algorithm is more consistent with the actual situation of the incident beam particle system than the simple average incidence.

$$\overline{C_{ext}} = \frac{1}{500}\sum C_{ext}(\xi) \tag{9}$$

Numerical results of extinction characteristics

Verification of extinction characteristics calculation

In order to verify the accuracy of particle extinction characteristics under the generalized Lorenz-Mie theory, the paper compares the curves of the extinction characteristic curves of individual particles with Asano's [8] and T-matrix's results [24]. Fig. 2 shows the variation of extinction coefficient with the change of spheroid size parameter C. The calculation example was selected with the shape parameter $a/b = 2$, the incident angle $\xi = 0°$, 45° and 90° respectively, the relative refractive index $m = 1.33$ (the imaginary part is zero), which infers that the absorption of light is not taken into account, i.e. $Q_{ext} = Q_{sca}$. The resultant curves in Fig. 2 coincide well with Fig. 19 in the reference [8], which verifies the correctness of the computation code. Meanwhile, the results of GLMT agree well with those of T-matrix in the cases of on-axis incidence. However, numerical deviations between the two algorithms can be observed at incident angle of 45°. Fig. 2 also reveals that the extinction coefficient increases with the increase of the size parameters, but the trend slows down and decreases after reaching the peak. At the same time, under the given dimension parameter, its value decreases with the change of the incident angle ξ.

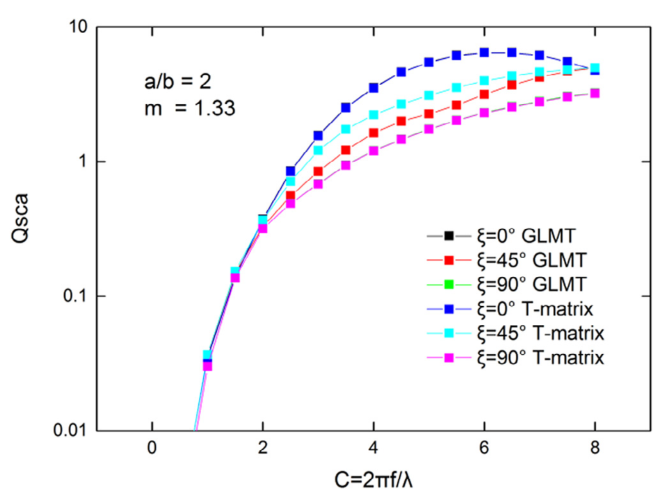

Fig. 2. Relations between extinction coefficient and non-dimensional parameters.

Extinction cross-section

Fig. 3 presents the influences of refractive index, incident angle of the beam, and ellipticity on extinction cross-section when the polarization angle $\Phi = 0°$. It shows that, whether the incident direction is fixed or not, the extinction coefficient grows monotonically with the increase of the axial ratio. That is, when the particles are gradually deviated from the sphere in shape, the extinction effect is becoming stronger. Meanwhile, the variation of the extinction coefficient appears almost unchanged when the axial ratio ranges from 1 to 3 in the same incident direction. The extinction coefficient increases with the growing of the imaginary part of the relative refractive index under the same incident direction and axial ratio, reflecting the obvious effect of light absorption on the light extinction (see Fig. 3(a)). It is evident from Fig. 3(b) that with the increase of incident angle ξ, the variation level of the spheroid cross-section with axial ratio reduces dramatically, and the curves tend to flatten. Since the extinction cross-section is more sensitive to the direction of major axis, the curve will gradually flatten when the incident direction is close to the minor axis. At the same time, it can be found from Fig. 3(c) that with the increase of the wavelength of incident light from 400 nm to 800 nm, the extinction cross-section shows a decreasing trend.

Light extinction spectra

In order to study the influence of the spheroidal particle shape, the light extinction spectra of spheroidal particles are calculated with Eq. (3). Because the particle diameter inversion heavily relies on the spectral information, the accurate analysis and comparison of the extinction spectra attach significant importance to determine the particle size. The particle existence of the actual particle system is mostly randomly oriented. Therefore, two statistical methods are proposed to calculate the extinction spectrum of the particle system. The spheroidal particles are selected with the effective volume of spherical particle diameter $D = 0.05$ μm, 0.1 μm, 0.5 μm, 1 μm, the refractive index $m = 1.33$ relative to the air, at volume concentration $C_v = 1e{-5}$ and optical path $L = 0.01$ m. The calculation of light extinction spectra employs simultaneously the arithmetic average and stochastic incidence distribution mentioned in the previous section for comparing and analyzing the effect of average in a specific incident direction. Fig. 4 presents a series of numerical results for arithmetic averaging and stochastic incidence algorithm under different equivalent particle sizes. It reveals that the results of arithmetic averaging algorithm are basically consistent with the stochastic incidence algorithm. In addition, it can be found that in the case of particle diameter $D = 0.05$ μm, 0.1 μm, and 0.5 μm, the light extinction spectra occurs with a downward trend with the increase of incident light wavelength, while the curve is to rise first and then decrease when particle diameter $D = 1$ μm. This actually reflects the characteristics of the particle size (or size of wavelength ratio) in the spectrum. In Fig. 4, the light extinction increases monotonously with the increase of the axial ratio under the condition of the same particle diameter and light wavelength. Moreover, with the change of axial ratio from 1 to 3, the light extinction spectra of particles with identical size yields only a slight change, whereas the deviation of light extinction spectrum becomes apparent as the axial ratio gradually increases. In fact, such a changing trend will cause a noticeable effect on the inversion of particle sizes.

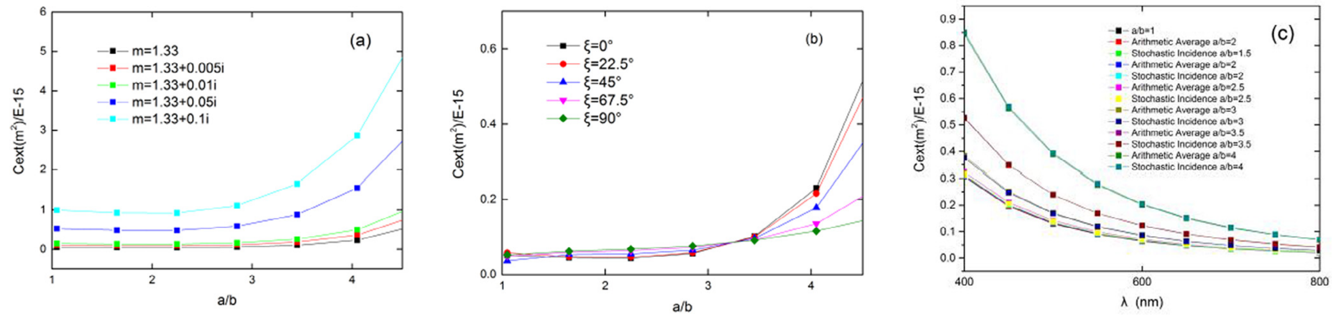

Fig. 3. Change of the extinction cross-section under different refractive index, incident direction and ellipticity. (a) $\xi = 0°$; (b) $\lambda = 632.8$ nm; (c) $m = 1.33$.

Inversion and results

Inversion method

In order to reflect the overall influence of particle shape on particle size characterization, the mathematical inversion process must be taken into careful consideration. We adopt a relatively stable regularization algorithm of independent model, when solving the Fredholm integral equation of the first kind (see Eq. (10)) to determine the size distribution of particles. For the polydisperse particle system, the integral form is given as:

$$\ln(I/I_0) = -l\int_{D_{\min}}^{D_{\max}} \overline{C}_{ext}(m,D,\lambda)N(D)dD \tag{10}$$

Among them, D_{min} and D_{max} are respectively the lower and upper limits of the particle size under study. Through the discretization of the mathematical transformation, the following linear equations can be obtained:

$$Tx = y, \quad x \geqslant 0, \tag{11}$$

where, T is called coefficient matrix whose elements are related to extinction cross-section, y a vector indicating the light extinction at different wavelengths, and x the probability frequency distribution of discrete particle size. As Eq. (11) shows, it is evident that the solution can be performed by non-negative least squares (NNLS) algorithm. However, a major drawback of this method is that its solution is greatly affected by the error perturbation of vector y, and the solution vector tends to be too oscillatory to reflect the true distribution. Therefore, the smooth matrix L and regularization parameter γ are employed to improve the property of coefficient matrix as [25],

$$\min_x(||T_x - y||^2 + \gamma||L_x||) \tag{12}$$

The regularization algorithm provided in solving Eq. (12) helps to lead to useful stabilized solutions.

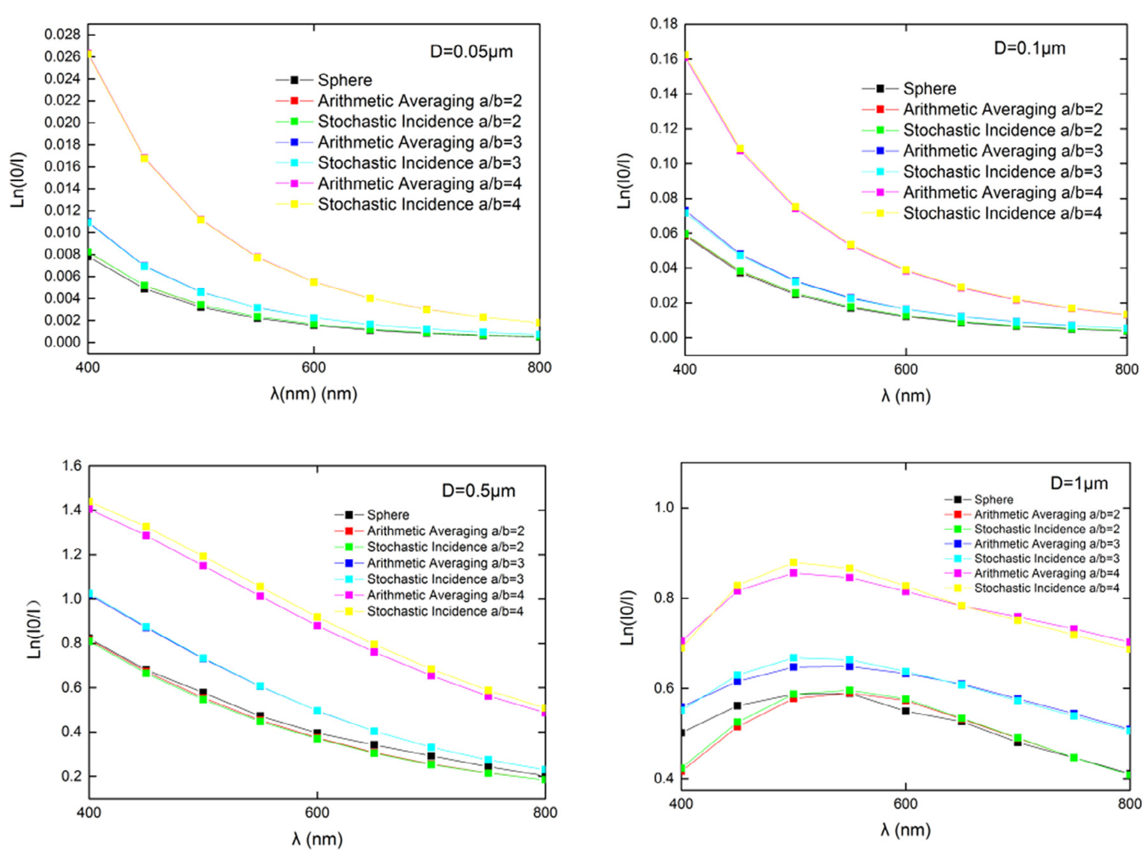

Fig. 4. Spheroid extinction spectra under different equivalent particle diameters.

Table 1
Inversion result of spheroid particles with various axial ratios a/b.

Nominal value D_{n50}/µm		Inversion result D_{n50}/µm			
		a/b = 1	a/b = 2	a/b = 3	a/b = 4
0.05	Arithmetic mean	0.050	0.050	0.051	0.053
	Stochastic incidence	0.050	0.050	0.051	0.053
	0 degree incidence	0.050	0.051	0.081	0.111
	11.25 degree incidence	0.050	0.051	0.066	0.111
	22.5 degree incidence	0.050	0.051	0.056	0.110
	33.75 degree incidence	0.050	0.051	0.052	0.061
	45 degree incidence	0.050	0.050	0.051	0.053
	90 degree incidence	0.050	0.050	0.050	0.050
0.1	Arithmetic mean	0.100	0.101	0.107	0.127
	Stochastic incidence	0.100	0.102	0.109	0.132
0.5	Arithmetic mean	0.500	0.506	0.500	0.646
	Stochastic incidence	0.500	0.506	0.500	0.657
0.8	Arithmetic mean	0.800	0.800	0.863	0.986
	Stochastic incidence	0.800	0.800	0.888	0.958
1	Arithmetic mean	1.000	1.104	1.021	1.048
	Stochastic incidence	1.000	1.095	1.028	1.058

Results and analysis

Through the inversion of the light extinction spectra of spheroids with different equivalent diameters and incident angles, Table 1 shows the retrieval particle sizes of monodispersed particles for various nominal values from 0.05 to 1 µm under different axial ratio conditions. Obviously different statistical methods did not bring apparent difference to the inversion results which implies that they are equally applicable to the modeling assumption. However, the idea of a fixed incident angle may produce relatively evident deviations, especially when the axial ratio is relatively large (say 3 or 4).

As expected, the axial ratio exerts a greater influence on the inversion results, and the relative deviations of the inversion results with the spheres could be quite significant under different equivalent particle sizes, which are also shown in Fig. 5(a). Specifically, when axial ratio equals to 1, the deviations of equivalent particle size and inversion results via two methods (stochastic statistics and arithmetic mean) are within 1%, which can be seen that the inversion procedure can successfully verify the given values. However, it can be observed that the deviation becomes larger with the increase of the axial ratio under the same particle size, and the maximum can even reach about 50%. It should also be noted that, when the axial ratio throughout the range from 1 to 3, the deviation is always within 10%.

In order to investigate the influence of particle shape on size characterization of polydisperse system, a series of particle systems satisfying the Gaussian normal distribution have been assumed to possess the nominal sizes of 0.05 µm, 0.1 µm, 0.5 µm, and 1 µm respectively and the identical standard deviation of 0.005 µm. As shown in Fig. 5(b), the trend of deviation against axis ratio is similar to that of the monodispersed particles, while it should also be noted that, the deviation is possibly increasing to around 20% with respect to the axial ratio throughout the range from 1 to 3, about twice as much as the former. At the same time, we explored the relative error of the given distribution parameter (i.e. standard deviation, 0.005 µm) of the particle system when the axial ratio is 2, and the maximum error is 8% for the system of nominal size of 0.5 µm. The resultant size distribution are narrower than the given ones for four particle systems in Fig. 6. Thus, it can be deduced that the influence of shape and the consequent deviations will be more crucial for a polydisperse system. On the whole, the results can be understood as a great influence of the shape of the spheroid exerting on the inversion of the particle size, particularly when the axial ratio is greater than 3.

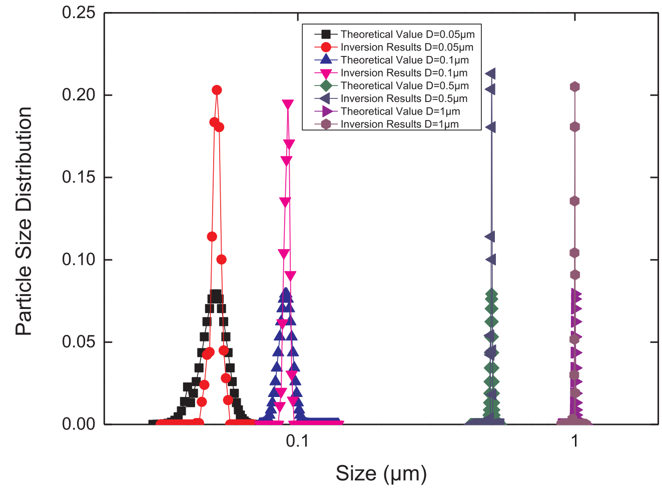

Fig. 6. Distribution of particle size of spheroidal particle system with the axial ratio of 2.

Conclusions

(1) In this paper, the generalized Lorenz-Mie theory is used to calculate the extinction cross-section and predict light extinction spectra of the spheroidal particles under various axial ratios and equivalent volumes. The calculation results of extinction cross-section show that under the same incident direction, the extinction cross-section increases with the increase of axial ratio, which means that under the equivalent volume, the extinction effect becomes stronger with the spheroid deviating from the sphere in shape. The calculation

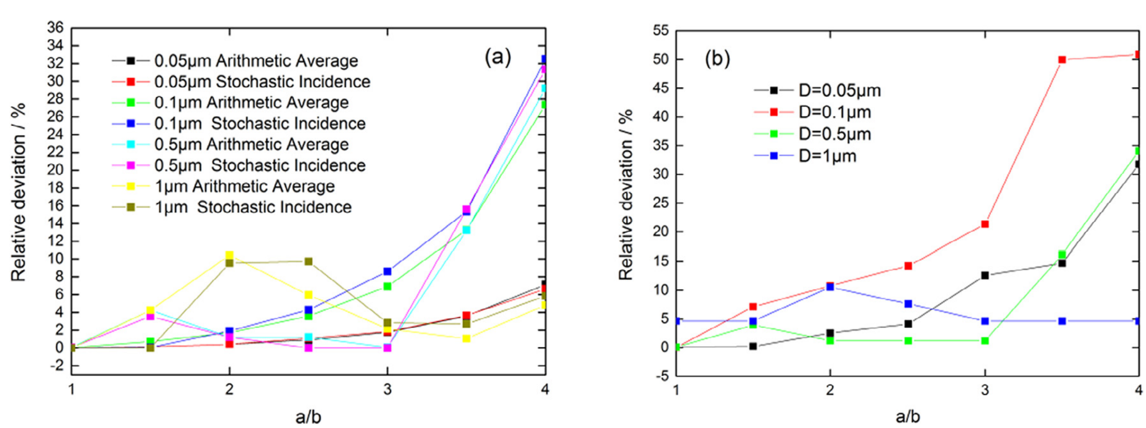

Fig. 5. Relative deviations of the inversion results and the calibration values under different equivalent diameters (a) monodispersion; (b) polydispersion.

results of the extinction spectra indicate that under the condition of the equivalent volume, the deviations of light extinction spectrum curve become more obvious from sphere to spheroid with the increase of axial ratio (especially when the ratio is larger than 3). It will also exert a major influence on the particle size inversion.

(2) After calculating the extinction spectrum of the spheroid, the particle size of spheroid is obtained by using the regularization inversion algorithm. According to the inversion results, the incident direction of the beam has a great influence on the inversion of the particle size. As the axial ratio increases under the equivalent volume, the deviation of particle inversion results also shows a rising trend. In particular when ratios reach 4 to 5, the deviations of the inversion results may be up to 50%. In contrast, when the ratio changes from 1 to 3, the deviations of spheroidal particle size inversion under equivalent volume keep within 20%. That is to say, it could be feasible and effective to characterize the particle size when the spheroidal axis ratio is within the range of 1 to 3, if possible influence and current level of deviation should be taken into account in multi-wavelength light extinction method, while deviation gets larger when beyond such axis ratio range.

Acknowledgments

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