



Influences of refractive index on forward light scattering

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ABSTRACT

The influence of the relative refractive index (RRI) of the particles to the surrounding medium on the small-angle forward scattering signals is studied, based on the Mie theory, the Debye series expansion (DSE) and the Fraunhofer diffraction theory. It comes to the conclusion that, for small particles, the influence on the forward scattering signals is mainly due to the part of the internal reflection if the RRI deviates from 1. However, when the RRI is close to 1, the effects on the forward scattered light from both the surface reflection and the internal reflection are great. For large particles, the contributions of the surface reflection and the internal reflection to the forward scattered light are much weaker than the diffraction when the RRI deviates from 1. When the RRI is very close to 1, the effects on the forward scattered light from the internal reflection are great. To determine the influence of the RRI in detail, the modified Chahine algorithm is employed. The inversion results cannot give the correct PSD for small particles if the RRI used in the inversion procedure does not match the one of the sample. The result shows that it is necessary to determine the exact value of the RRI and one should avoid the RRI close to 1 by choosing dispersion with proper refractive index in practice.

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1. Introduction

Information of particle size and particle size distribution (PSD) is valuable in the production of particles of specific sizes to control process efficiency and product quality [1–3]. Light scattering has proved to be a powerful technique for particle sizing. The forward light scattering technique (FLST) has been one of the most well-issued and thus widely-used techniques, which employs Fraunhofer diffraction (FD) or the Mie theory. During the past decades, much work concerned with the comparison between the FD and the Mie theory has been published [4–9]. It was concluded that the measurement on the particle size is best described by the Mie theory rather than by the FD, especially when the particles are small. As a result, most of the particle size analyzers employ the Mie theory.

So far, it seems that all the problems in the FLST have been solved satisfactorily. The particle size distribution can be measured accurately with the Mie theory supposing that the refractive index of the particles is known. But, when the refractive index of the particles is unknown, one would have to use a “guessed” value of the refractive index in the inversion procedure, which might not match the real one of the measured sample. In this case, the measurement error would inevitably occur. In this paper, the effects on the measurement results in the FLST caused by the

mismatch between the input value of the refractive index and the real one of the sample are studied. In the first part, the dependence of the scattered light on the refractive index of the particles is studied with numerical calculations of the Mie theory and its DSE [10–13]. In the second part, the scattered light of a pre-determined particle size distribution and a fixed refractive index is simulated, which is taken as the signal. Different values of the refractive index are used in the inversion problem and the corresponding particle size distributions are obtained.

2. Dependence of the scattered light on the refractive index

2.1. Mie theory and Debye series expansion

Scattering of a parallel light beam by a spherical particle can be rigorously described by the Mie theory. However, the Mie theory gives a global description of the light scattering only. It does not give much insight into the physical processes involved in light scattering. The DSE, by contrast, allows for the decomposition of the global physical process into a series of local interactions, which can bring a better physical understanding. When the light interacts with a particle a number of processes can occur, including surface reflection, refraction together with multiple internal reflections, absorption and diffraction, as shown in Fig. 1 [10–15].

According to the Mie theory, when a parallel light beam of light hits the spherical particles, if the incident light is unpolarized with

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intensity I_0 , the scattering intensity I is [8]

$$I = \frac{\lambda^2 I_0}{8\pi^2 r^2} [|s_1(\theta)|^2 + |s_2(\theta)|^2] \quad (1)$$

where λ is the wavelength of the light, s_1 and s_2 are the amplitudes of the scattering field and are given by

$$s_1(\theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (a_n \pi_n + b_n \tau_n)$$

$$s_2(\theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (a_n \tau_n + b_n \pi_n) \quad (2)$$

where a_n and b_n are the Mie coefficients related to the Bessel and Hankel functions whose variables are the particle size parameter and the refractive index of the particle, π_n and τ_n are the Mie angular functions.

The Mie coefficients a_n and b_n can be written as a decomposition of Debye series so that they can represent the separated contributions to the scattered light. The Debye series expansion of the Mie amplitudes is thus given as [10–15]

$$\begin{bmatrix} a_n \\ b_n \end{bmatrix} = \frac{1}{2} \left[1 - R_n^{212} - \sum_{p=1}^{\infty} T_n^{21} (R_n^{212})^{p-1} T_n^{12} \right] \quad (3)$$

where R_n^{121} expresses the reflection coefficient and T_n^{21} is the transmission coefficient of the incoming spherical wave on the particle surface, R_n^{121} is the reflection coefficient and T_n^{12} is the transmission coefficient of the outgoing spherical wave. Here, the particle is labeled with 1 and the surrounding medium is labeled with 2. All these parameters depend on the RRI of the particle to its surrounding medium and the particle size parameter.

According to the DSE, each term on the right-hand side of Eq. (3) has a clear physical interpretation. The first term describes the diffraction of light around the particle, which is independent of

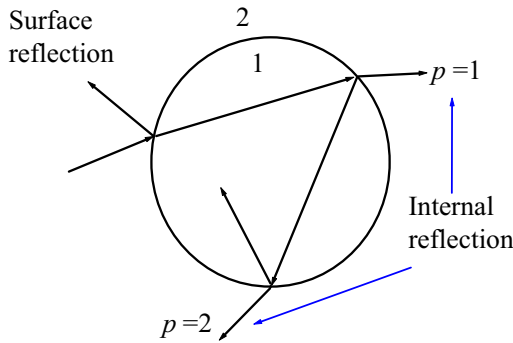


Fig. 1. Schematic of the Debye series.

the RRI. The second term represents the outgoing waves that have reflected on the particle surface and the third term is the sum of the outgoing waves with $p-1$ internal reflections. Both the surface reflection and the multiple internal reflections depend on the RRI. If all the Debye series terms are summed, the Debye series calculation can give the same result as that from the Mie calculation.

The diffraction part of the scattered light can be described by the first term of Eq. (3), which leads to an approximate expression [16]

$$i_{\text{diff}} = \frac{\lambda^2 I_0 \alpha^2}{8\pi^2 r^2} \frac{\theta}{\sin \theta} \left[\frac{2J_1(\alpha\theta)}{\alpha\theta} \right]^2 \quad (4)$$

where $\alpha = \pi d/\lambda$ is the particle size parameter and d is the particle diameter.

If the particle is strongly absorptive, contribution from the multiple internal reflections is very small due to the attenuation of the light while propagating in the particle so that it can be omitted. In this case, the scattered light can be approximately described by the diffraction and surface reflection

$$\begin{bmatrix} a_n \\ b_n \end{bmatrix} = \frac{1}{2} (1 - R_n^{212}) \quad (5)$$

It should be noted that in its present form the Debye series is not amenable to separation of the diffraction term from the surface reflection term [15].

2.2. Numerical results on the scattered light

In this section, the effects of the RRI on the angular distribution of the scattered light are studied with numerical calculation. The particle size parameters in the calculation are $\alpha = 10$ and $\alpha = 100$ respectively.

First, we study the dependence of the angular distributions of scattered light on the refractive index of non-absorbing particles. The RRIs of the particles to the surrounding medium are $m = 0.60, 0.75, 1.05, 1.63$ and 2.5 . Fig. 2 shows the angular distributions of the total scattered light calculated with the Mie theory. It can be seen that there are visible differences between all the results, no matter small or large particles. For large particles (see Fig. 2b), the angular distributions of the scattered light for the RRIs 0.60, 0.75, 1.63 and 2.5 differ from each other slightly. However, for the RRI 1.05, the scattered light deviates largely from all the other cases. This indicates that the refractive index has a great effect on the distribution of the total scattered light when the particle is small and when the RRI is close to 1.

To look more into the detail, the angular distributions of the scattered light which contains the diffraction and the surface reflection are calculated (i.e. the multiple internal reflections are

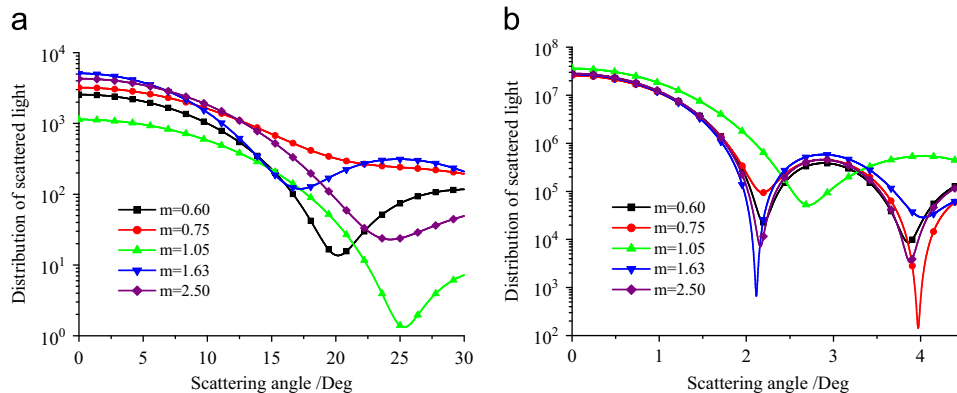


Fig. 2. Angular distributions of the total scattered light with different refractive indices. (a) $\alpha = 10$ and (b) $\alpha = 100$.

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