Retrieving the refractive index of a sphere from the phase spectrum of its light-scattering profile

A V Romanov\textsuperscript{1, 2, 3} and M A Yurkin\textsuperscript{1, 2}

\textsuperscript{1}Voevodsky Institute of Chemical Kinetics and Combustion SB RAS, Institutskaya Str. 3, 630090, Novosibirsk, Russia
\textsuperscript{2}Novosibirsk State University, Pirogova Str. 2, 630090, Novosibirsk, Russia
\textsuperscript{3}Corresponding author: avm.romanov@gmail.com

Abstract. We studied the Fourier spectrum of the light-scattering profiles of single particles in the Rayleigh–Gans–Debye (RGD) and Wentzel–Kramers–Brillouin (WKB) approximations. In the case of a homogeneous sphere, we found the relationship between the key parameters of the spectrum (including its phase) and the sphere characteristics – both analytically and numerically in the framework of the approximations and the rigorous Lorentz–Mie theory, respectively. Based on these results, we have improved the existing spectral characterization method for spheres extending the applicability range to particles with a higher refractive index.

1. Introduction
Light scattering is one of the most common approaches to the non-invasive characterization of microparticles. Among multitude of existing methods, the most promising ones are those working with single particles, since they are more reliable than ensemble ones (the corresponding inverse problem is typically well posed). There are many approaches to solving such inverse light-scattering problems (Ref. [1] and references therein), one of which is the compression of information in the measured light-scattering profiles or patterns (LSP) into several parameters that determine the characteristics of the particle under study. An important common advantage of such methods is the resistance to various distortions introduced both by experiment and by the imperfection of the particle model. Several recent examples include spectral methods for determining the size and refractive index of spheres [2, 3] and for non-sphericity estimation [4, 5].

In particular, an accurate and robust characterization of spheres was demonstrated in a limited range of size and refractive index [2]. This method extracted two parameters from the Fourier spectrum of the one-dimensional LSP: the main peak position and the zero-frequency amplitude, which strongly correlated with the size and refractive index, respectively. However, the ambiguity of the latter parameter for high refractive indices limited its use for widespread polystyrene beads. In this work we show the reasons for this ambiguity and propose a way to improve the capabilities of the spectral method based on the usage of the phase spectrum.

2. Main analytical results
We focus on the versatility of our approach for variety of applications and mostly consider the scattered intensity of unpolarized light. However, one of the prominent examples, the scanning flow cytometer, measures the following combination of Mueller scattering matrices:
\[ I(\theta) = \frac{1}{2\pi} \int_0^{2\pi} [S_{11}(\theta, \phi) + S_{14}(\theta, \phi)] d\phi, \]  

(1)

where \( \theta \) is polar angle from \( \theta_1 = 10^\circ \) to \( \theta_2 = 65^\circ \). Note that the integral of \( S_{14} \) is exactly zero for any axisymmetric particle [6]. We apply the FFT-based spectral transformation, described in [4], to the discrete measured data, in order to have a discrete representation of the following continuous transform:

\[ F(v) = \frac{1}{2\pi} \int_{-\infty}^{\infty} w(\theta) I(\theta) \exp(-iv\theta) \, d\theta \]  

(2)

where \( w(\theta) \) is the Hann window function for the range \([10^\circ, 65^\circ]\).

Analyzing the light-scattering problem in the framework of the RGD approximation, we have managed to explain the origin of the main peak in the Fourier spectrum and its relation to the particle size. Furthermore, analyzing the WKB approximation, we found that the given Fourier transform of the scattering intensity has a phase factor linearly proportional to both the refractive index and the frequency. Such a transformation result can be expected already proceeding from the given formula for the intensity

\[ |I(\theta)|^2 = \frac{1}{4\pi} \int_{-\infty}^{\infty} \exp \left( i2 \sin \frac{\theta}{2} kp + i(m-1)kp \right) F(p, \theta) \, dp \]  

(3)

where \( (m-1)kp \) is a phase factor, and \( F \) is some function defined by particle geometry, the spectrum of which we obtain as a result of the Fourier transformation. This will be discussed in more detail at the conference.

A factor of this type in the Fourier image indicates a shift of the original function (LSP) along the coordinate axis (\( \theta \)) by a value proportional to the refractive index, which was observed and used earlier for spheres [7,8].

3. Numerical calculations and discussions

The above theoretical analysis suggests that the phase of the spectrum at peak frequency (further denoted as peak phase) strongly depends on the refractive index, so it is natural to combine it with the peak location, which strongly depends on size and only weakly on refractive index. In contrast to the zero-frequency amplitude, used in [4], the peak phase is equally applicable for any refractive index at the cost of being cyclic, i.e. inherently non-invertible in any wide range. Thus, its natural application niche is characterization of particles in a relatively narrow range of size and refractive indices. In particular, we further apply it to characterization of 4-μm polystyrene beads. Note, however, that similar calculations can be performed for other ranges of characteristics.

We used the Lorenz–Mie theory to calculate the LSPs in the ranges \([3.7 \, \mu m, 4.3 \, \mu m]\) and \([1.56, 1.62]\) for size and refractive index, respectively, and Fourier-transformed them according to Eq. (2). We assumed the wavelength 0.66 μm and host-medium refractive index equal to 1.333. In the resulting spectra, we determined the position of the main peak and the phase value at this point. The dependence of these parameters on the sphere characteristics are shown in Fig. 1. As can be seen from the figure, the phase has an almost linear dependence on the refractive index; however, there is some ripple associated with effects not accounted for in the WKB approximation.
Figure 1. The dependence of peak location (a) and its phase value (b) on size and refractive index calculated by using the Lorentz–Mie theory.

Inverting this dependence by plotting the same data points in other coordinates (characteristics of the sphere as functions of the parameters of the spectrum), we solve the inverse problem using interpolation. Despite the underlying linear structure, ripples present significant problems by compromising uniqueness and affecting interpolation accuracy. Note that these ripples present problems for any other characterization methods as well, even for the least-square fit. For example, the LSPs of two spheres with size different by a ripple period (approximately 0.15 μm in this case) maybe more similar to each other than to LSP of spheres with sizes in between. To assess the influence of these ripples on our method, we tested it on synthetic LSPs of spheres, computed in the same range of characteristics but different from the interpolation grid nodes. While the maximum errors are 10 nm and 10⁻³ for size and refractive index, the mean errors do not exceed 1 nm and 10⁻⁴, respectively. The results of processing experimental LSPs of polystyrene beads will be presented at the conference.

4. Conclusion
We have analyzed the application of the phase spectrum of 1D LSP to the characterization of spheres, both analytically using the WKB approximation and numerically using the Mie theory. Moreover, we constructed an interpolant to characterize homogeneous spheres using the position and phase of the main peak in the Fourier spectrum. This interpolant works in the limited range of size and refractive indices (due to the cyclicality of the phase parameter), but proved to be fast, robust, and accurate for characterization of latex beads.

Acknowledgments
The research has been supported by the Russian Foundation for Basic Research (grant No. 19-32-90073).

References
[1] Romanov A V and Yurkin M A 2021 Single-particle characterization by elastic light scattering Laser & Photon. Rev. 15 2000368
[2] Romanov A V, Konokhova A I, Yastrebova E S, Gilev K V, Strokotov D I, Chernyshev A V, Maltsev V P and Yurkin M A 2017 Spectral solution of the inverse Mie problem J. Quant. Spectrosc. Radiat. Transf. 200 280–94
[3] Konokhova A I, Yastrebova E S, Strokotov D I, Chernyshev A V, Karpenko A A and Maltsev V P 2019 Ultimate peculiarity in angular spectrum enhances the parametric solution of the inverse Mie problem J. Quant. Spectrosc. Radiat. Transfer 235 204–8

[4] Romanov A V, Konokhova A I, Yastrebova E S, Gilev K V, Strokotov D I, Maltsev V P and Yurkin M A 2019 Sensitive detection and estimation of particle non-sphericity from the complex Fourier spectrum of its light-scattering profile J. Quant. Spectrosc. Radiat. Transf. 235 317–31

[5] Yastrebova E S, Dolgikh I, Gilev K V, Vakhrusheva I V, Liz E, Litvinenko A L, Nekrasov V M, Strokotov D I, Karpenko A A and Maltsev V P 2021 Spectral approach to recognize spherical particles among non-spherical ones by angle-resolved light scattering Opt. Laser Technol. 135 106700

[6] Yurkin M A, Semyanov K A, Tarasov P A, Chernyshev A V, Hoekstra A G and Maltsev V P 2005 Experimental and theoretical study of light scattering by individual mature red blood cells with scanning flow cytometry and discrete dipole approximation Appl. Opt. 44 5249–56

[7] Maltsev V P and Lopatin V N 1997 Parametric solution of the inverse light-scattering problem for individual spherical particles Appl. Opt. 36 6102–8

[8] Shepelevich N V, Lopatin V V, Maltsev V P and Lopatin V N 1999 Extrema in the light-scattering indicatrix of a homogeneous sphere J. Opt. A 1 448–53