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Spectral solution of the inverse Mie problem

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ABSTRACT

We developed a fast method to determine size and refractive index of homogeneous spheres from the power Fourier spectrum of their light-scattering patterns (LSPs), measured with the scanning flow cytometer. Specifically, we used two spectral parameters: the location of the non-zero peak and zero-frequency amplitude, and numerically inverted the map from the space of particle characteristics (size and refractive index) to the space of spectral parameters. The latter parameters can be reliably resolved only for particle size parameter greater than 11, and the inversion is unique only in the limited range of refractive index with upper limit between 1.1 and 1.25 (relative to the medium) depending on the size parameter and particular definition of uniqueness. The developed method was tested on two experimental samples, milk fat globules and spherized red blood cells, and resulted in accuracy not worse than the reference method based on the least-square fit of the LSP with the Mie theory. Moreover, for particles with significant deviation from the spherical shape the spectral method was much closer to the Mie-fit result than the estimated uncertainty of the latter. The spectral method also showed adequate results for synthetic LSPs of spheroids with aspect ratios up to 1.4. Overall, we present a general framework, which can be used to construct an inverse algorithm for any other experimental signals.

1. Introduction

Light scattering is ubiquitous in nature and technology and is often the only or the most feasible approach to characterize particles or particle systems [1]. Many characterization techniques address the particle ensemble as a whole, due either to a large scattering volume [2,3] or to the dense packing of individual components [4]. However, they are inherently ill-posed in trying to retrieve the distribution of the ensemble over the particle characteristics [2,5]. Single-particle techniques show greater promise in detailed and robust characterization, at least in the controlled laboratory conditions [6,7].

Successful single-particle characterization requires three ingredients: measurement, simulation, and inversion. The typical measured signals consist of a few scalar values [8,9], an angle-resolved light-scattering pattern (LSP), [10,11] or a two-dimensional LSP [12,13]. The simulation part benefits from several well-established methods and open-source codes [4,14]. It is now easy to simulate light scattering by almost any complex inhomogeneous particle, while the main complexity comes from the vastness of the multidimensional space of possible particle characteristics [15,16]. The ultimate solution for the inverse problem would be a direct imaging (tomography) approach with no prior assumptions about the object. However, all existing attempts employ 2D LSPs for many orientations of the same particle and either assume weak scattering (Rayleigh-Debye-Gans approximation) [17,18] or require phase of the scattered field to be measured as well [19,20]. Otherwise, one has to assume a particle shape model a priori, reducing the problem to determining several characteristics of this model. Such characterization methods can be tentatively divided into 3 broad categories: nonlinear regression, machine learning, and parametric (compression) techniques.

Nonlinear regression is based on the direct comparison of experimental signals (typically, LSPs) with simulated ones, using some norm of the difference (residual). Global minimization of this residual is a challenging task with computational complexity rapidly increasing with the number of shape characteristics [15,21]. This complexity can be partly concentrated into a one-time investment of computational power using precalculated (look-up) database of LSPs [22–24]. This makes it possible to apply

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nonlinear regression routinely for particles without spherical symmetry, such as red blood cells [25], platelets [26], and rod-shaped bacteria [27]. Moreover, in addition to the best-fit particle characteristics one can construct confidence intervals for these characteristics, i.e. evaluate the characterization errors [23,26,28].

Machine learning is an extremely broad class of methods, but we discuss only neural networks as the most representative example. Ideally, this approach should be able to automatically train itself on a large set (database) of LSPs with known characteristics and handle high-dimensional problems [29]. But, practically, its performance is hard to predict and it may require a lot of fine tuning. So far the neural networks has only been used for light-scattering characterization of spherical particles [30,31] and red blood cells [32], as recently reviewed in [33]. Moreover, those methods do not use the whole LSP, but only a number of parameters derived from it.

This compression of an experimental signal into several (two-three) parameters is central to the third class of characterization methods. The parameters are extracted either directly from the measured signal, e.g. the LSP [34,35] or the time-resolved signal [36], or from its Fourier [37,38] or Gegenbauer [39] spectrum. Alternatively the amount of experimental data may originally be limited to only a couple of numbers [8,9]. The specific way to process the signal parameters can vary a lot, but all parametric methods have very high speed in determining a small number of particle characteristics (typically, only the size and, sometimes, the refractive index). They are also potentially more stable to instrumental noise and distortions, as well as to distortions of the optical model. The most popular parametric method is the spectral sizing [40–42], which is based on almost linear relation between the size of a sphere and characteristic frequency of its LSP or, equivalently, the position of the main peak in the Fourier spectrum of the LSP. Interestingly, the spectral sizing of homogeneous spheres [43] can be extended with virtually no changes to estimation of diameters of leukocytes [44] and red blood cells [45], although the accuracy of this estimation has not been thoroughly tested. Moreover, estimation of sphere refractive index has been proposed (without assessing the accuracy) in a limited range of size and refractive index, using the integral of the LSP as a second parameter [46].

Each of the three above classes has its pros and cons, and occupies a certain application niche. This paper is devoted to the systematic development of the spectral method with the goal to fully characterize a homogeneous sphere, i.e. to solve the inverse Mie problem both quickly and robustly. For that we compress the whole measured LSP into two parameters of its power Fourier spectrum, which are further transformed into two characteristics of the particle. In Section 2 (and Appendix A) we construct this characterization method starting with a LSP measured with the scanning flow cytometer (SFC) [11,43]. However, the provided details should enable one to repeat the whole procedure for any other experimental set-up and/or signal parameters. We also perform a detailed theoretical analysis of the underlying map and applicability (uniqueness) domain of the developed method. In Section 3 we describe two sets of experimental measurements, namely milk fat globules and spherized red blood cells, and a set of synthetic data for spheroids. Those data, affected by both instrumental noise and optical-model distortions, are used for thorough testing of the characterization method in Section 4. We conclude the paper in Section 5.

2. Spectral characterization method

In this Section we construct a method to determine both diameter d and refractive index n of a spherical particle from the spectrum of its LSP. The main idea is to compress the whole LSP spectrum into two parameters, to describe the direct problem as a map of particle characteristics into those parameters ($G: \mathbb{R}^2 \to \mathbb{R}^2$), and to invert this map by constructing an interpolant. To abstract from specific wavelength of the incident light λ and medium refractive index n_0 we further describe the particle by its size parameter $x = \pi d n_0/\lambda$ (d – sphere diameter) and relative refractive index $m = n/n_0$. We also limit ourselves to non-absorbing particles, i.e. assume real m. Moreover, we consider only $m > 1$, while the case of $0 < m < 1$ is expected to be qualitatively similar. Allowing m to take values on both sides of unity will, most probably, break the uniqueness of the inverse problem in the whole range of particle characteristics.

2.1. Power spectrum and its parameters

Let us define the specific form of a LSP spectrum. We start with the standard LSP, measured by the SFC:

$$I(\theta) = \int_0^{2\pi} \mathrm{d}\varphi [S_{11}(\theta,\varphi) + S_{14}(\theta,\varphi)], \tag{1}$$

where S is the Mueller scattering matrix [47], θ and φ are the polar and azimuthal scattering angles, and $S_{14} \equiv 0$ for ideal spheres. To keep the discussion manageable we further only consider the LSP in the range from $\theta_1 = 10°$ to $\theta_2 = 65°$ and apply the same spectral transformation as in [43]. Specifically, the LSP is multiplied by the Hanning window function

$$w(\theta) = \sin^2\left(\pi \frac{\theta - \theta_1}{\theta_2 - \theta_1}\right), \tag{2}$$

and its power Fourier spectrum on this finite range is computed:

$$P(q) = \left| \frac{1}{\theta_2 - \theta_1} \int_{\theta_1}^{\theta_2} \mathrm{d}\theta\, w(\theta) I(\theta) \exp(-2\pi \mathrm{i} q\theta) \right|^2, \tag{3}$$

which is normalized to be only weakly dependent on a particular choice of the angular range. The practical calculations are performed with the fast Fourier transform using uniform discretization over $N = 256$ intervals with further zero-padding up to $M = 4096$ points (to increase the spectral resolution):

$$P(q_k) = \left| \frac{1}{N} \sum_{j=0}^{N-1} w(\theta_1 + j\Delta\theta) I(\theta_1 + j\Delta\theta) \exp\left(-\mathrm{i}\frac{2\pi}{M} kj\right) \right|^2, \tag{4}$$

where $\Delta\theta = (\theta_2 - \theta_1)/N$ and $q_k = k/(M\Delta\theta)$. This procedure is illustrated in Fig. 1, where we also defined the spectral parameters: location (L) and amplitude (A_p) of non-zero spectral peak and amplitude of zero frequency (A_0). L can also be called the main (angular) frequency, while A_0 is the squared average value of the windowed LSP – similar to the parameter used in [46]. The peak parameters are determined by the quadratic fit with a window width of 15 points ($0.98\,\mathrm{rad}^{-1}$). While we conventionally use units of degree for θ, we employ the dimensional SI units (rad^{-1}) for q and L and omit it further. Note also, that the Mueller matrix S and, hence, I and P are dimensionless. However, their scales (used in all figures) are unambiguously defined by Eqs. (1)–(4). In other words, arbitrary units are not used anywhere.

Almost linear relation between x and L, with x/L roughly equal to π, is well-known [43,48] and can be understood using simple arguments of diffraction gratings or the Rayleigh-Debye-Gans (RDG) approximation [47]. The choice of the second parameter (to deduce m as well) is less obvious. Relative peak amplitude A_p/A_0 has been briefly discussed previously [43,45]; here we also consider both A_0 and A_p separately. However, as shown in Section 4.1, A_0 is the most robust (insensitive) with respect to experimental distortions of the LSP. Therefore, we base the production algorithm on this parameter and use it as a primary example in the following discussion. The corresponding analysis for other two parameters is