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An optimization method for solving the inverse Mie problem based on adaptive algorithm for construction of interpolating database

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ABSTRACT

We introduce a numerical solution of the inverse light-scattering problem for a single non-absorbing spherical particle. The solution is implemented by global optimization at preliminary constructed database of light-scattering patterns. We propose an adaptive method for database construction, which aims both at providing satisfactory local accuracy and at avoiding large errors of the inverse map. Several databases were constructed varying the required accuracy of solution of the inverse problem and parameters used to characterize a sphere. We tested accuracy of the method on synthetic data for spheres with and without noise, on synthetic data for slightly prolate and oblate spheroids, and on experimental data of polystyrene microspheres measured with a scanning flow cytometer. The constructed databases have shown appropriate results in determination of the size and refractive index of a sphere from the angle-resolved light scattering with given accuracy.

1. Introduction

The problem of single particle characterization from light scattering arises in different fields, including astronomy, remote sensing, and analysis of aerosols and emulsions [1]. Optical methods play an important role in solution of this problem. These methods in analysis of individual particles, especially biological cells, can be divided into the following categories: (1) probe-field methods forming an electromagnetic field in smallest volume (confocal microscopy) and (2) full-field methods based on a detailed analysis of light scattered by a particle. Most powerful method that may be considered to belong to the second category is flow cytometry. A flow cytometer allows the measurements of light scattering from a single particle in fixed solid angles [2] or in a form of an entire angle-resolved light-scattering pattern (LSP) [3]. Recently Strokotov et al [4] modernized the flow cytometer for measurement of regular and polarized LSPs of individual particles simultaneously. New facilities of flow cytometry in the measurement of ample light-scattering data force us to develop new approaches in a solution of the inverse light-scattering (ILS) problem. Generally flow cytometers measure individual particles with a rate of 1000 particles per second that forms an extra requirement for the solution of the ILS problem—the fast determination of particle parameters (characteristics) from LSPs [5].

At present a practical analytical solution of the ILS problem for a single particle is unavailable. The most successful attempt is the result for spheres that allows determination of its parameters from the Mie scattering amplitudes, which include both the amplitude and phase components of a scattering field, measured over the

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whole angular interval [0°,180°] [6]. However, such complete measurements cannot be performed with flow cytometry.

The alternative approaches are to solve the inverse problem numerically. Spectral decomposition methods [7–9] allow one to determine the size of a sphere from the Gegenbauer of Fourier spectrum of the LSP. However, it provides no information about the refractive index. A parametric solution [10] is based on dependence of particle parameters on the phase-shift and fringe pitch of LSPs. It provides satisfactory accuracy only for spheres and is sensitive to the experimental noise [5]. Neural networks were also used to characterize single spheres [11–13]. This method requires only a learning sample of calculated LSPs and it was successfully used on experimental data of polystyrene spheres and spherized red blood cells [11]. However, application of neural networks is still an art, which requires fine-tuning of internal parameters for the particular scattering problem. That is why it has not been yet applied to non-spherical particles.

The most general approach for characterization of particles with relatively simple shape is optimization, i.e. direct fitting of the experimental signal to the computed LSPs. Oscillatory nature of the LSPs with strong dependence on particle parameters calls for robust global optimization techniques. In particular, stochastic global optimization techniques [14,15] and the DiRect method [15,16] were applied to spheres.

A similar problem, characterization of multi-layered concentric spheres, was approached by multi-start Levenberg-Marquardt [17] and the DiRect [18] methods. However, optimization methods come at a great computational cost which becomes unbearable for non-spherical particles, such as blood platelets and red blood cells, due to increasing number of parameters to explore and much slower computation of the LSP. New optimization methods have to allow determination of cell characteristics in reasonable time to be included in common routine of hematological analysis. Therefore, there is demand for acceleration of the optimization methods using some kind of preliminary exploration of a particular scattering problem. Since the problem of single particle characterization usually has to be solved multiple times in a row (in flow cytometry applications at least several thousand times), one-time investment of large computational time should be acceptable.

Conceptually the simplest approach for such acceleration is to calculate a large database of LSPs and to solve the ILS problem by the nearest-neighbor interpolation. In particular, this method was applied to spheres [19], spheroids [20] and biconcave disks [21,22]. However, in all cases of non-spherical particles the accuracy of the solution was not reliably assessed. The main problem of this approach, not addressed in the mentioned papers, is finding the optimum structure and size of the database. One should strike a compromise between computational time, both for the construction of the database and for each interpolation, and the accuracy of solution of the inverse problem. While a 'large enough' database may work for spheres, a careful compromise is required for practical feasibility of characterization of non-spherical particles. It is complicated by variability of sensitivity of the LSP to the particle parameters over their domain, calling for variable density of the database. Moreover, there may be a certain threshold in the dependence of interpolation error on grid density due to oscillatory nature of the LSP. Above this threshold the error continuously decreases with increasing density, as typical for interpolation. But below the threshold the errors abruptly increase to the values comparable with the size of the whole parameter domain (see Section 3). A related question is how large experimental noise is acceptable for a particular database.

Optimality of the interpolation database was previously addressed for a different problem, detecting defects in 2D systems [23,24]. However, the proposed adaptive algorithm of database construction focused only on interpolation error within the elementary cell, thus implicitly assuming that database density is large enough. There are also sampling methods based on nearest-neighbor interpolation [25,26], which share similar ideas. However, they construct a set of samples for a particular experimental signal instead of an universal database.

In this paper we propose a method to adaptively construct a database to characterize single particles from measured LSPs. This method aims at providing satisfactory uniqueness and local accuracy, avoiding large errors of the inverse map and limitation of the database constructed. After a general description of the problem and the method in Sections 2 and 3 respectively, we consider the simplest model, a homogeneous sphere, for a practical example in Section 4. In a recent paper [27] the inverse Mie problem is solved by constructing a set of starting points for the gradient-based optimization. This approach is based on detailed and rigorous analysis of the map, implementing LSP calculation, and its derivative, which guarantees perfect accuracy at least for the noise-free data. In contrast, this paper is based on the implicit assumption that a relatively small number of LSPs is the most complete information about the map one can obtain. From the current practical viewpoint this assumption seems to be valid for all non-spherical particles. Therefore, the method proposed in this paper should be applicable to many particle models described by a few parameters, but with no complete rigor due to the discreteness of the analysis.

2. Problem statement

The direct scattering problem under consideration consists in determination of the LSP from given parameters of a particle. It is implemented by a map f: $X \to Y$, where $X \subset \mathbf{R}^p$ is a domain of parameters of a particle, $Y \subset \mathbf{R}^d$ is a domain of LSP and $d \geq p$. We assume that f is a C^1-smooth and one-to-one map, whose value can be obtained (with sufficient accuracy) for any point in X. In particular, for a sphere the map is implemented using the Mie theory [28], which provides an analytical solution in terms of infinite series. Specifics of the Mie theory are also discussed in [27]. Typical LSP's for spherical particles, defined by Eq. (12) below, are presented in Fig. 1.