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# Application of simulated annealing for simultaneous retrieval of particle size distribution and refractive index

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ABSTRACT

This paper describes the application of the simulated annealing technique for the simultaneous retrieval of particle size distribution and refractive index based on polarization modulated scattering (PMS) measurements. The PMS technique is a well-established method to measure multiple elements of the Mueller scattering matrix. However, the inference of the scatterers' properties (e.g., the size distribution function and refractive index) from such measurements involves solving an ill-conditioned inverse problem. In this paper, a new inversion technique was demonstrated to infer particle properties from PMS measurements. The new technique formulated the inverse problem into a minimization problem, which is then solved by the simulated annealing technique. Both numerical and experimental investigation on the new inversion technique was presented in the paper. The results obtained demonstrated the robustness and reliability of the new algorithm, and supported its expanded applications in scientific and technological areas involving particulates/aerosols.



## 1. Introduction

The use of light scattering as a noninvasive diagnostic tool for small particles (or aerosols) has a long history and encompasses an incredibly wide spectrum of applications, ranging from the remote sensing of interstellar dust (Wickramasinghe, 1991), to the detection of atmospheric particles (Bohren & Huffman, 1983), and to the studies of marine organisms (Quinby-hunt, Hunt, Lofftus, & Shapiro, 1989). All these applications essentially involve solving the inverse scattering problem, i.e., the inference of the scatterers' properties (size distribution function and refractive index) from scattering measurements. Light scattering is fully described by the 4-by-4 Mueller scattering matrix (Kerker, 1969). Consequently, all information about the scatterer properties that can be extracted from scattering measurements is contained in the elements of the Mueller matrix. Therefore, different techniques and instruments have been developed to measure these elements (Ding et al., 2007; Hull, Shepherd, & Hunt, 2004; Hunt & Huffman, 1973; Hunt, Quinby-Hunt, & Shepard, 1998; Perry, Hunt, & Huffman, 1978). These techniques typically utilize polarization modulated scattering (PMS) to simultaneously measure multiple elements of the Mueller matrix.

However, the retrieval of the scatterers' properties from these measurement elements (either multi-angular or multi-spectral) is not trivial, because of the ill-posedness of the inverse scattering problem. Due to decades of research efforts in this area, numerous inversion methods have been developed, each with its own advantages and limitations. Many of these methods are developed for a specific application, where certain simplifications can be assumed (e.g., the anomalous diffraction approximation). An extensive review of these methods is beyond the scope of this paper, and the readers are referred to monographs and papers

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dedicated for this purpose (e.g., Baltes, 1980; Shchyogolev, 1999; Shifrin & Tonna, 1993). It is highly desirable to have a robust inversion method that can be applied over a relatively wide range of conditions.

This paper describes such an attempt. Here, a new inversion method was developed and demonstrated to inverse multi-angular PMS data to simultaneously obtain the size distribution function and refractive index. The new method cast the inversion problem into a minimization problem, which was then solved by the simulated annealing algorithm (Corana, Marchesi, Martini, & Ridella, 1987; Kirkpatrick, Gelatt, & Vecchi, 1983). This technique was experimentally demonstrated with polystyrene spheres with a narrow size distribution, and showed good agreement when compared with other measurement techniques. Though demonstrated for spheres here, the inversion technique can also be extended to more complicated particles (e.g., nonspherical particles and composite particles), which will be discussed in more detail in Section 5.

The rest of the paper is organized as follows. Section 2 introduces the theoretical background of the measurement technique and the inversion algorithm. Section 3 describes the experimental arrangement, with results reported in Section 4. Finally, Section 5 summarizes the paper with discussions.

## 2. Theoretical background

The single scattering of light can be described by the 4×4 Mueller matrix (Bohren & Huffman, 1983), which reduces to the following form due to symmetry relationships when the scatterers are spherical or consist of an ensemble of randomly aligned particles:

$$M = \begin{pmatrix} S_{11} & S_{12} & 0 & 0 \\ S_{12} & S_{11} & 0 & 0 \\ 0 & 0 & S_{33} & S_{34} \\ 0 & 0 & -S_{34} & S_{33} \end{pmatrix} \quad (1)$$

The elements of the Mueller matrix depend on the scattering angle and the scatterers' properties (size distribution, complex refractive index, shape, etc.), and contain all the information available from elastic light scattering. The $S_{11}$ element is proportional to the total scattered intensity, a quantity that most scattering techniques measure. Other elements of the matrix can be measured via various polarization modulated techniques (Hunt & Huffman, 1973; Perry et al., 1978). When the scatterers are polydispersed, these elements need to be averaged by the size distribution function. In this paper, these notations ($S_{11}$, $S_{12}$, etc.) represent the size-averaged elements. Generally, any one of the Mueller elements is insufficient to characterize a particle system. For example, the $S_{11}$ element of two particles with different sizes and shapes can be experimentally indistinguishable over the entire range of scattering angles (from 0° to 180°). Therefore, measurement of multiple elements is usually necessary.

To infer the size distribution function and refractive index of the scatterers from the measured Mueller elements, a least-square problem was formulated. For example, in this study, where two of the Mueller elements ($S_{11}$ and $S_{12}$) were measured at multiple scattering angles, the following object function, $F$, is constructed:

$$F(f, m) = \sum_{i=1}^{N} \left[ \frac{S^m_{12}(\theta_i)}{S^m_{11}(\theta_i)} - \frac{S^c_{12}(\theta_i)}{S^c_{11}(\theta_i)} \right]^2 \quad (2)$$

where $f$ and $m$ represent the size distribution function and refractive index of the scatterers, respectively; $N$ is the total number of angular positions; $S^m_{11}(\theta_i)$ and $S^m_{12}(\theta_i)$ are the measured $S_{11}$ and $S_{12}$ at a scattering angle of $\theta_i$ ($i = 1, 2, \ldots, N$), respectively; and $S^c_{11}(\theta_i)$ and $S^c_{12}(\theta_i)$ are the calculated $S_{11}$ and $S_{12}$ at a certain $f$ and $m$ at $\theta_i$, respectively. Note that (1) only $S_{11}$ and $S_{12}$ appear in Eq. (2) because only these Mueller elements were measured in this study, and incorporation of additional elements in the objective function is straightforward and (2) the ratio of $S_{12}$ to $S_{11}$ was used to eliminate the influence of the number density of particles. Minimization of $F$ with respect to $f$ and $m$ essentially solves the inverse scattering problem in the least-square sense, which is a rather "standard" technique.

However, minimizing $F$ is not trivial due to the complicated behavior of $S_{11}$ and $S_{12}$ with respect to $f$ and $m$. Such complication leads to many local minima in $F$, which poses a significant challenge to many optimization algorithms, especially those based on the gradient of the objective function (Press, Teukolsky, Vetterling, & Flannery, 1992). These local minima can easily trap the algorithm into a false solution. Therefore, a robust minimization algorithm must be carefully chosen. Here, we found that the simulated annealing algorithm (Corana et al., 1987; Kirkpatrick et al., 1983) provides a robust method to minimize the objective function. The simulated annealing algorithm roots from an analogy to the way liquids are annealed, i.e., cooled slowly to crystallize. During the annealing process, the molecules in the liquid rearrange themselves in such a way that a state of minimum energy is reached when the liquid crystallizes. In function minimization, function values are the counterpart of energy states in liquid annealing; and the simulated annealing algorithm minimizes a function by "annealing" it. The simulated annealing technique has been successfully tested on many difficult functions to find the global minimum in the presence of interfering local minima. In our application, the simulated annealing algorithm is able to overcome the complications in the objective function, and yield stable solutions of $f$ and $m$. Based on the above understanding, an inversion algorithm was developed according to the flow chart shown in Fig. 1.

Another well-proven key virtue of the simulated annealing algorithm is its insensitivity to the initial guesses. Past experience (Hunt et al., 1998) with diesel soot measurement by the PMS technique has suggested that the minimization of the objective