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Inversion of light-scattering measurements for particle size and optical constants: theoretical study

Matthew R. Jones, Bill P. Curry, M. Quinn Brewster, and Keng H. Leong

We invert the Fredholm equation representing the light scattered by a single spherical particle or a distribution of spherical particles to obtain the particle size distribution function and refractive index. We obtain the solution by expanding the distribution function as a linear combination of a set of orthonormal basis functions. The set of orthonormal basis functions is composed of Schmidt–Hilbert eigenfunctions and a set of supplemental basis functions, which have been orthogonalized with respect to the Schmidt–Hilbert eigenfunctions by using the Gram–Schmidt orthogonalization procedure. We use the orthogonality properties of the basis functions and of the eigenvectors of the kernel covariance matrix to obtain the solution that minimizes the residual errors subject to a trial function constraint. The inversion process is described, and results from the inversion of several simulated data sets are presented.

Key words: Optical particle sizing, inverse problems.

Introduction

Measurements of the radiation scattered by a sample contain information regarding the physical properties of the sample. Therefore, the properties of a sample can often be determined from the interaction of the sample with radiation from a known source. Laser light-scattering measurements can be made unobtrusively in environments that are inaccessible to other types of measurements, so light-scattering techniques have become an important tool in areas as diverse as astronomy, combustion, meteorology, geology, medicine, and bioengineering. A promising application of light-scattering techniques is the determination of the size and the optical properties of a particle or a collection of particles from their light-scattering patterns. The primary difficulty associated with these techniques is inverting the measurements or extracting the desired information from the data.

Experiments in engineering or the natural sciences often result in data that are indirectly related to the desired characteristic or property of a system. These experiments can often be modeled through the use of a set of linear integral equations in which the unknown property of the system is embedded inside the integral. The inverse light-scattering problem is a specific example of this broad class of problems, which are known generally as linear inverse problems with discrete data. Because mathematically similar problems occur in many scientific disciplines, techniques for solving linear inverse problems have been thoroughly investigated. Twomey¹ gave a detailed account of the mathematics of inversion and described many of the schemes used to solve inverse problems. More recently, Bertero *et al.*^{2,3} published a discussion of the general formulation of this class of problems and reviewed more solution methods. The fact that a recent issue of *Applied Optics*⁴ was devoted entirely to optical particle sizing techniques illustrates the tremendous interest in developing and improving these techniques.

In general, techniques for solving the inverse light-scattering problem are classified as either analytical or empirical. Analytical techniques involve formal solutions of integral equations that describe the light-scattering or extinction process. Because the information content in a set of light-scattering or extinction measurements is limited,^{1,5–7} inverse prob-

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lems usually do not possess a unique solution and are therefore ill-posed problems. Because of the ill-posed nature of inverse problems, most analytic inversion techniques require the use of *a priori* information regarding the distribution function or a careful optimization of the inputs. Indeed, the primary difference among most analytic inversion schemes is the way the *a priori* information is incorporated or the inputs are optimized. In addition, most analytic inversion techniques are limited by the fact that the complex refractive index of the particles must be known.

Empirical inversion techniques generally require that a parametric model of the light-scattering or extinction process be developed. The parameters are then adjusted within physically realistic bounds so that a least-squares fit of the measured data is obtained. An empirical inversion technique known as the optical strip-map technique has been developed by Quist and Wyatt.⁸ This technique is attractive in that it does not involve repeated calculations and requires much less data than other analytical or empirical inversion techniques. The optical strip-map technique has been successfully used to retrieve the size and real refractive index from single particle light-scattering measurements.^{8,9} However, this technique does not provide any information regarding the absorption index, and it cannot be applied to measurements of the light scattered by distributions of particles.

The purpose of this study was to develop a practical inversion technique capable of inverting measurements of the light scattered by a single particle or by a distribution of particles for the particle size distribution function (PSDF) and optical properties. This paper presents the theoretical development of the inversion process, and a subsequent paper¹⁰ describes the application of the inversion procedure to light-scattering patterns obtained with a multichannel polar nephelometer. The technique developed in this study is most successful when applied to single particles or narrow distributions, but the application of the technique to broader distributions is also examined. A description of the inversion technique is given, with an emphasis on the mechanics of the inversion process. The results from several inversions of simulated data sets are also presented. Symbols used in this paper are defined in Appendix A.

Scattering Equation

In the development of the inversion technique, attention was focused on simulating experiments in which we used multichannel polar nephelometers to measure the light scattered by homogeneous spherical particles. Two nephelometers were considered. The first nephelometer has 15 detectors positioned between 23° and 128° and uses a GaAlAs laser diode with a wavelength of 0.840 μm as the light source. The detectors are positioned so they are equidistant in $\sin(\theta/2)$. The second nephelometer has 36 detec-

tors positioned every 4° from 20° to 160° and uses a 0.67-μm diode laser as a light source. Therefore, the available set of measurements consists of simultaneous measurements of the power scattered into the solid angles subtended by detectors located at several polar angles.

The ratio of the power scattered in the direction of a particular detector to the incident irradiance is defined as an angular scattering cross section. Assuming single scattering, we find that the angular scattering cross section measured by the *j*th detector C_j is related to the distribution of sizes and optical properties by an inhomogeneous Fredholm equation of the first kind:

$$C_j = \int_{V_j} \int_{\Delta\Omega_j} \int_{k_i}^{k_f} \int_{n_i}^{n_f} \int_{x_i}^{x_f} N_c \tilde{f}(x, n, k) \times \frac{d\tilde{C}_j}{d\Omega}(\Omega, x, n, k) dx dn dk d\Omega dV + \delta C_j. \quad (1)$$

The δC_j in Eq. (1) are the unknown errors in the measurements and are assumed to be Gaussian distributed. If the particles are homogeneous spheres, the differential scattering cross sections ($d\tilde{C}_j/d\Omega$) (Ω, x, n, k) can be calculated from Mie theory.^{11,12} The following assumptions reduce the complexity of Eq. (1).

First, for weakly absorbing particles, the scattering kernels can be approximated by the product of a function that depends only on k and the scattering kernel with k set equal to zero:

$$\frac{d\tilde{C}_j}{d\Omega}(\Omega, x, n, k) \sim h(k) \frac{d\tilde{C}_j}{d\Omega}(\Omega, x, n, 0). \quad (2)$$

Second, if a distribution of particles is present, all the particles have the same optical properties:

$$\tilde{f}(x, n, k) = f(x)\delta(n - n_s)\delta(k - k_s). \quad (3)$$

Third, if a distribution of particles is present, the particle number density is uniform over the scattering volume. Fourth, the solid angles subtended by the detectors are small, so the integral over Ω can be replaced by the product of the average of the differential scattering cross sections and $\Delta\Omega$:

$$\int_{\Delta\Omega_j} \frac{d\tilde{C}_j}{d\Omega}(\Omega, x, n, k) d\Omega = \frac{d\tilde{C}_j^{\text{avg}}}{d\Omega}(x, n, k) \Delta\Omega_j. \quad (4)$$

These assumptions simplify Eq. (1) to

$$C_j = N_c V_j \Delta\Omega_j h(k_s) \int_{n_i}^{n_f} \int_{x_i}^{x_f} f(x)\delta(n - n_s) \times \frac{d\tilde{C}_j^{\text{avg}}}{d\Omega}(x, n, 0) dx dn + \delta C_j. \quad (5)$$

Neglecting the error in the measurements, we can

approximate the average of the angular scattering cross sections by the average of the right-hand side of Eq. (5):

$$C_{\text{avg}} = \frac{1}{m} \sum_{j=1}^m C_j$$

$$C_{\text{avg}} \approx \frac{N_c V_j h(k_s)}{m} \sum_{j=1}^m \Delta \Omega_j \int_{n_i}^{n_f} \int_{x_i}^{x_f} f(x) \delta(n - n_s) \times \frac{d\tilde{C}_j^{\text{avg}}}{d\Omega}(x, n, 0) dx dn. \quad (6)$$

We can eliminate the unknown function $h(k)$ from Eq. (5) by normalizing by the average of the measurements. Earlier investigations have shown that it is beneficial to weight each measurement and scattering kernel by the corresponding estimate of the experimental error.^{13,14} It is also assumed that the solid angle subtended by each detector is the same. These modifications simplify Eq. (5) to

$$c_j = \frac{1}{C_{\text{avg}}} \int_{n_i}^{n_f} \int_{x_i}^{x_f} f(x) \delta(n - n_s) \times \frac{dC_j^{\text{avg}}}{d\Omega}(x, n, 0) dx dn + \delta c_j, \quad (7)$$

where

$$c_j = \frac{C_j}{C_{\text{avg}} \Delta C_j}, \quad (8)$$

$$\delta c_j = \frac{\delta C_j}{C_{\text{avg}} \Delta C_j}, \quad (9)$$

$$\frac{dC_j^{\text{avg}}}{d\Omega}(x, n, 0) = \frac{(d\tilde{C}_j^{\text{avg}}/d\Omega)(x, n, 0)}{\Delta C_j}, \quad (10)$$

$$C_{\text{avg}} = \frac{1}{m} \sum_{j=1}^m \int_{n_i}^{n_f} \int_{x_i}^{x_f} f(x) \delta(n - n_s) \times \frac{d\tilde{C}_j^{\text{avg}}}{d\Omega}(x, n, 0) dx dn. \quad (11)$$

Inversion Process

Equation (6) represents an integral operator that transforms a function $f(x)\delta(n - n_s)$ into a vector $\{c_j\}$, so the problem presented in the previous fits the definition of a linear inverse problem with discrete data.^{2,3} The inversion process used in this study is a combination of analytical and empirical inversion techniques. The eigenfunction method is initially used for the retrieval of the unconstrained or generalized solution. The eigenfunction method has been applied to particle sizing by several authors and is thoroughly discussed in the literature.^{3,6,7,13,15-17} Curry¹³ also discussed the similarities between the

eigenfunction method and the Phillips–Twomey constrained linear inversion method.^{1,18} The eigenfunction method uses the Schmidt–Hilbert theory for linear integral operators¹⁹ to solve inverse problems. Because the Schmidt–Hilbert theory is essentially the singular value decomposition of the integral operator defined by Eq. (6), methods similar to the eigenfunction method are sometimes referred to as singular value analyses.^{2,20-22}

It is well known that the problem presented by Eq. (6) is ill posed, so even small errors in the measurements can lead to a physically unrealistic generalized or unconstrained solution. However, the results of this study show that the real part of the refractive index is accurately obtained from the unconstrained solution if the inputs are carefully selected from the available measurement set. We use information obtained from the unconstrained solution in conjunction with a preliminary analysis of the data to choose a trial function constraint. The preliminary analysis of the data is a simplified version of the optical strip-map technique developed by Quist and Wyatt.⁸ We then use the constrained eigenfunction method described by Curry¹³ to retrieve the PSDF. We obtain the value of the absorption index by comparing the measured scattering pattern with the scattering pattern calculated by use of the retrieved refractive index and PSDF. The value of the absorption index that results in a rms value of the residual errors that is closest to the rms of the imprecision estimates is taken to be the retrieved absorption index. In experiments in which spectral extinction measurements are made, it may be possible for one to retrieve the absorption index in a more direct manner. Shaw²³ discussed an inversion procedure that uses a combination of spectral extinction and angular scattering measurements.

In summary, there are five major steps in the inversion process: (a) preliminary analysis of the measurements, (b) selection of the optimal inputs from the available measurement set, (c) retrieval of the refractive index through the use of the unconstrained solution, (d) retrieval of the PSDF through the use of the constrained solution, and finally (e) retrieval of the absorption index by the matching of the measured and calculated scattering patterns. We present an example inversion with the description of the mathematical formulation to illustrate the mechanics of the inversion process. An effort has been made to keep the description of the mathematics brief but still provide the reader with all the equations needed to perform an inversion. The simulated data set used in the example inversion is representative of measurements obtainable with the 36-channel nephelometer.

Preliminary Analysis of the Simulated Measurements

One should calculate angular scattering cross sections for several different sizes and optical properties within the expected ranges before attempting to

Table 1. Range of Sizes and Optical Properties

Diameter Range (μm)	Size Parameter Range	Refractive Index Range	Absorption Index Range
0.1–10.1	0.5–47.0	1.1–2.0	$\leq 10^{-3}$

invert any measured values. Particular attention should be given to the value of the most forward angular scattering cross section available and the average of the angular scattering cross sections. These parameters will serve as a guide in the selection of the trial function used in obtaining the constrained solution. The ranges of sizes and optical properties considered in the example inversion are listed in Table 1. Angular scattering cross sections were calculated for the 36-channel nephelometer at 12 different sizes and three sets of optical properties. Table 2 lists the average and 20° angular scattering cross sections. Table 2 can be considered to be a low-resolution optical strip map.⁸

We calculated simulated measurements representing the light scattered by a single spherical particle by using Eq. (1). The size and the optical properties of the particle were randomly selected from the ranges specified in Table 1, so the actual parameters of the distribution function were not known until after the inversion was completed. We added Gaussian distributed random noise to each angular scattering cross section to simulate experimental conditions. The angular scattering cross sections are plotted in Fig. 1, and the imprecision estimates shown in the figure are equal to the standard deviation of the random noise. The standard deviation of the random noise was equal to 10% of the error-free measurements. The average of these angular scattering cross sections is $0.67 \mu\text{m}^2$, and the 20° angular scattering cross section has a value of $2.6 \mu\text{m}^2$. A comparison of the 20° angular scattering cross section and the average of the angular scattering cross sections with the values

Fig. 1. Simulated angular scattering cross sections.

in Table 2 indicates that the particle has a diameter of approximately $4\text{--}5 \mu\text{m}$ (a size parameter between 19 and 23).

Input Selection

The scattering kernels are not mutually orthogonal functions, so a large number of measurements may contain a relatively small number of independent measurements.^{1,5} Therefore, we find it necessary to select an optimized set of inputs from the available set of measurements. We use an algorithm similar to that used by Capps *et al.*⁶ to determine the input set. We calculate the kernel covariance or Gram matrix corresponding to the complete set of measurements, and the eigenvalues and eigenvectors of the kernel covariance matrix are calculated. The elements of the kernel covariance matrix are defined by

$$M_{ij} = \int_{n_i}^{n_f} \int_{x_i}^{x_f} \frac{dC_i^{\text{avg}}}{d\Omega}(x, n, 0) \frac{dC_j^{\text{avg}}}{d\Omega}(x, n, 0) dx dn. \quad (12)$$

The eigenvalues of $\mathbf{M}$ are calculated, and an expression derived by Twomey⁵ for the relative error is used as a way to determine whether the selected inputs are suitable for the inversion process. The relative error

Table 2. Average and 20° Angular Scattering Cross Sections

Diameter (μm)	Size Parameter	Optical Properties					
		$1.1 + i 10^{-3}$		$1.5 + i 10^{-3}$		$2.0 + i 10^{-3}$	
		C_{avg} (μm^2)	C_{20° (μm^2)	C_{avg} (μm^2)	C_{20° (μm^2)	C_{avg} (μm^2)	C_{20° (μm^2)
0.1	0.5	3×10^{-7}	4×10^{-7}	7×10^{-6}	8×10^{-6}	2×10^{-5}	3×10^{-5}
0.5	2.3	3×10^{-4}	3×10^{-3}	1×10^{-2}	8×10^{-2}	4×10^{-2}	0.1
1.0	4.7	4×10^{-3}	0.1	0.1	0.4	0.1	0.3
2.0	9.4	1×10^{-2}	0.2	0.2	0.6	0.2	0.7
3.0	14	3×10^{-2}	0.6	0.3	3	0.4	1
4.0	19	0.1	0.9	0.5	2	0.6	2
5.0	23	0.1	2	0.6	4	0.9	4
6.0	28	0.2	2	1	5	1	5
7.0	33	0.3	4	1	6	2	5
8.0	38	0.3	4	2	8	2	7
9.0	42	0.6	8	2	10	2	10
10.0	47	0.6	7	2	11	3	12

is defined by Eq. (13) and is equal to the square root of the ratio of the square norm of the error in the distribution function to the square norm of the distribution function. Equation (13) is dominated by the condition number, which is the ratio of the largest eigenvalue to the smallest eigenvalue.^{2,5,7,24}

$$\Delta f = \frac{1}{m} \left(\frac{\sum_{j=1}^m \frac{1}{\lambda_j} \sum_{j=1}^m \lambda_j \sum_{j=1}^m \Delta C_j^2}{\sum_{j=1}^m C_j^2} \right)^{1/2}. \quad (13)$$

If the relative error given by Eq. (13) is too large, then we use the largest off-diagonal element of the kernel covariance matrix to identify the two most nearly dependent measurements. The sums of the squares of the off-diagonal matrix elements are calculated for the rows that correspond to the two most redundant measurements. The largest of these sums identifies the measurement that is most nearly dependent on the rest of the measurements, and that measurement is eliminated from the set of inputs. A new kernel covariance matrix is then calculated, and the process is repeated until the relative error calculated from Eq. (13) is small enough. In this study, the best results were obtained when the relative error was slightly less than 1. For the angular scattering cross sections shown in Fig. 1, between 27 and 30 of the 36 measurements were eliminated from the input set before a relative error less than 1 was achieved. The variation in the number of inputs was due to variations in the range of real refractive indices, as we discuss in the next section.

Retrieval of the Refractive Index

We use the Schmidt–Hilbert theory¹⁹ to obtain a set of orthonormal functions that are known as Schmidt–Hilbert eigenfunctions. The unconstrained solution is obtained by the expansion of the distribution function as a linear combination of the Schmidt–Hilbert eigenfunctions.

$$f(x)\delta(n - n_s) \approx \sum_{j=1}^m a_j \Phi_j(x, n). \quad (14)$$

The unconstrained expansion coefficients are calculated from

$$a_j = \frac{\sum_{i=1}^m c_i u_i^j}{\sqrt{\lambda_j}}, \quad (15)$$

where u_i^j is the i th term of the j th eigenvector of the kernel covariance matrix and λ_j is the eigenvalue that corresponds to the j th eigenvector. The following

Fig. 2. Unconstrained PSDF.

equation is used to generate the Schmidt–Hilbert eigenfunctions:

$$\Phi_j(x, n) = \frac{1}{\sqrt{\lambda_j}} \sum_{i=1}^m u_i^j \frac{dC_j^{\text{avg}}}{d\Omega}(x, n, 0) \quad \text{for } 1 \leq j \leq m. \quad (16)$$

The value of the refractive index and the unconstrained PSDF are calculated from the unconstrained solution.

$$W(n_s) = \frac{\int_{n_i}^{n_f} \int_{x_i}^{x_f} W(n) \sum_{j=1}^m a_j \Phi_j(x, n) dx dn}{\int_{n_i}^{n_f} \int_{x_i}^{x_f} \sum_{j=1}^m a_j \Phi_j(x, n) dx dn}, \quad (17)$$

$$n_s = W^{-1}[W(n_s)], \quad (18)$$

$$f(x) \approx \int_{n_i}^{n_f} \sum_{j=1}^m a_j \Phi_j(x, n) dn. \quad (18)$$

We use the weighting function $W(n)$ to increase the sensitivity of the unconstrained solution to changes in the real refractive index. In order to be effective, the weighting function should be a physically significant function of the refractive index. One possibility is to use the extinction efficiency or an approximation of the extinction efficiency as the weighting function. A weighting function that proved to be useful in this study is $(n - 1)^2$. This function has the same dependence on the refractive index as the phase shift squared, which is an approximation to the extinction efficiency for large size parameters.¹²

In practice, it is usually necessary to vary n_i and n_f to ensure that the retrieved refractive index is close to the actual value. When inverting the example data set we first considered the entire range of refractive indices (1.1–2.0), and the retrieved real refractive index was 1.47. The range of refractive indices was then narrowed to 1.3–1.6, and the retrieved refractive index was 1.43. This process was continued until

Fig. 3. Constrained PSDF.

the retrieved value of the real refractive index converged to 1.42. The unconstrained PSDF is shown in Fig. 2.

Retrieval of the Particle Size Distribution Function

The unconstrained solution satisfies Eq. (6) for the set of inputs, and therefore it is a mathematically correct solution. However, the PSDF obtained from the unconstrained solution displays characteristics such as high-frequency oscillations and negative values that make it physically unrealistic. These unrealistic characteristics are due to the ill-posed nature of the problem and must be eliminated through the use of a smoothing or regularization method. Although there are a number of possible regularization methods,³ a trial function constraint was used in this study. In general, the nature of the trial function will depend on the particular conditions under which the measurements are made and can only be determined after careful consideration of the particular experiment. The present study will demonstrate that if the data consist of measurements of the light scattered by a single particle or by an ensemble of nearly identical particles, the unconstrained solution and the preliminary analysis of the measurements provided enough information for one to choose a trial function successfully. In our example inversion, it is

Fig. 4. Comparison of the simulated measurements and the calculated scattering pattern with $x_t = 25.1$.

Fig. 5. Comparison of the simulated measurements and the calculated scattering pattern with $x_t = 20.1$.

known that the measurements are of light scattered by a single homogeneous particle. Based on this fact, the form of the trial function is chosen to be

$$f^t(x, n) = \delta(x - x_t)\delta(n - n_s). \quad (19)$$

The preliminary analysis of the measurements indicated that x_t should be in the range 19–23. We obtained the initial value of x_t used in Eq. (19) by examining the unconstrained PSDF shown in Fig. 2. The most prominent peak of the unconstrained PSDF in or near the expected size ranges occurs at $x = 25.1$. Therefore, x_t was chosen to be 25.1.

Imposition of the trial function constraint requires that the trial function and the unknown distribution function be expanded as linear combinations of a set of orthonormal basis functions. However, only the portion of the trial function that lies in the space spanned by the scattering kernels can be represented by the use of the Schmidt–Hilbert eigenfunctions. Therefore, a set of supplemental basis functions or pseudo-empirical eigenfunctions⁷ are introduced as a way to form a more nearly complete set in the solution space. We obtain the additional basis functions by orthogonalizing a set of supplemental orthonormal functions with respect to the Schmidt–Hilbert eigenfunctions. The supplemental orthonormal functions used in the example inversion were

$$\phi_j(x, n) = \delta(x - x_j)\delta(n - n_s) \quad \text{for } 1 \leq j \leq 101, \quad (20)$$

Table 3. Typical Results from the Inversion of a 36-Channel Simulated Data Set

Parameter	Value	
	Actual	Retrieved
Diameter (μm)	4.5	4.3
Size parameter	21.2	20.1
Optical properties	$1.45 + i 7.5 \times 10^{-5}$	$1.42 + i 1.0 \times 10^{-4}$

Table 4. Typical Results from the Inversion of a 15-Channel Simulated Data Set

Parameter	Value	
	Actual	Retrieved
Diameter (μm)	7.6	7.6
Size parameter	27.9	28.0
Optical properties	$1.58 + i 5.0 \times 10^{-4}$	$1.58 + i 1.0 \times 10^{-5}$

where the x_j are evenly spaced throughout the size range.

It should be noted that the trial function and these supplemental basis functions are not true functions. In practice, $\delta(x - x_j)$ was approximated by the use of a one-dimensional array in which every element except the j th element was zero. The j th element was assigned a value of $(x_j - x_{j-1})^{-1}$. An approximation for $\delta(n - n_s)$ was obtained in the same manner. Some of the $\phi_j(x, n)$ will lie entirely in the space spanned by the lower-order basis functions and will be eliminated during the orthogonalization procedure. Therefore, the number of supplemental basis functions p will be less than the number of $\phi_j(x, n)$. The number of supplemental basis functions used in the example inversion was 93. The first step in the application of the Gram-Schmidt orthogonalization procedure is the removal of the components of the higher-order basis functions that are parallel with lower-order basis functions:

$$\tilde{\phi}_j(x, n) = \phi_j(x, n) - \sum_{i=1}^{j-1} \phi_j(x, n) \int_{n_i}^{n_f} \int_{x_i}^{x_f} \phi_j(x', n') \times \Phi_i(x', n') dx' dn', \quad 1 \leq j \leq 101. \quad (21)$$

The orthogonalized basis functions are then normalized:

$$\Phi_j(x, n) = \frac{\tilde{\phi}_{j-m}(x, n)}{\left[\int_{n_i}^{n_f} \int_{x_i}^{x_f} \tilde{\phi}_{j-m}^2(x', n') dx' dn' \right]^{1/2}}, \quad m < j \leq m + p. \quad (22)$$

Table 5. Comparison of the Actual and Retrieved PSDF and Refractive Indices

Case	Mean Size Parameter		Geometric Standard Deviation		Refractive Index	
	Retrieved	Actual	Retrieved	Actual	Retrieved	Actual
1	10.1	10.0	1.08	1.11	1.33	1.33
2	19.7	20.0	1.11	1.11	1.46	1.45
3	23.2	20.0	1.09	1.35	1.45	1.45
4	48.9	35.0	1.03	1.11	1.56	1.55
5	47.4	35.0	1.04	1.22	1.56	1.55
6	55.0	55.0	1.03	1.02	1.33	1.33

The trial function and the unknown distribution function can now be accurately expressed in terms of the basis functions:

$$f^t(x, n) = \sum_{j=1}^{m+p} a_j^t \Phi_j(x, n), \quad (23)$$

$$f(x) \delta(n - n_s) = \sum_{j=1}^{m+p} a_j^c \Phi_j(x, n). \quad (24)$$

The expansion coefficients for the trial function are obtained from

$$a_j^t = \int_{n_i}^{n_f} \int_{x_i}^{x_f} f^t(x, n) \Phi_j(x, n) dx dn. \quad (25)$$

The constrained expansion coefficients are found by minimizing the residual errors subject to the trial function constraint. A performance function is defined as

$$Q = \sum_{j=1}^m \left[\frac{1}{c_j^{\text{avg}}} \int_{n_i}^{n_f} \int_{x_i}^{x_f} \sum_{i=1}^m a_i^c \Phi_i(x, n) \frac{dC_j^{\text{avg}}}{d\Omega}(x, n_s, 0) \times dx dn - c_j \right]^2 + \gamma \int_{n_i}^{n_f} \int_{x_i}^{x_f} \left[\sum_{i=1}^m a_i^c \Phi_i(x, n) - \sum_{i=1}^m a_i^t \Phi_i(x, n) \right]^2 dx dn, \quad (26)$$

where γ is a Lagrange multiplier. The Lagrange multiplier is sometimes referred to as a weighting or regularization parameter, because it determines the relative importance of the trial function. The performance function is proportional to the residual errors in the retrieved solution and to the square norm of the difference between the unconstrained solution and the trial function. Minimizing the performance function with respect to the expansion coefficients gives the following expression for the constrained expansion coefficients:

$$a_j^c = \frac{a_j \lambda_j + \gamma a_j^t}{\lambda_j + \gamma} \quad \text{for } 1 \leq j \leq m, \\ a_j^c = a_j^t \quad \text{for } m < j \leq m + p. \quad (27)$$

Fig. 6. Particle size distribution functions.

Clearly, the value used for γ will be important in the inversion process. Shaw²³ demonstrated that rms deviation between the retrieved and actual distributions has a minimum with respect to the Lagrange multiplier. For the current implementation of the eigenfunction method, it can be shown that the square norm of the error introduced by applying the constraint is minimized if the parameter known as the residual relative variance (RRV) is minimized with respect to γ .¹³ The RRV is defined in Eq. (28), and the partial derivative of the RRV with respect to γ is given in Eq. (29).

$$\text{RRV} = \sum_{j=1}^m \frac{\lambda_j (c_{\text{avg}}^2 / C_{\text{avg}}^2) + \gamma^2 (a_j - a_j^f)^2}{(\lambda_j + \gamma)^2}, \quad (28)$$

$$\frac{\partial \text{RRV}}{\partial \gamma} = \sum_{j=1}^m \frac{\lambda_j [\gamma (a_j - a_j^f)^2 - (c_{\text{avg}}^2 / C_{\text{avg}}^2)]}{(\lambda_j + \gamma)^3}. \quad (29)$$

We obtain the optimal value of γ by increasing γ until $\partial \text{RRV} / \partial \gamma$ is approximately zero. In the example inversion, at $\gamma = 86.3$, $\partial \text{RRV} / \partial \gamma = -5.0 \times 10^{-8}$.

Once the optimal value of γ is determined, the constrained expansion coefficients can be calculated from Eq. (27). We then obtain the PSDF by integrating the constrained solution over the range of refractive indices. The constrained PSDF for the example inversion is plotted in Fig. 3.

$$f(x) = \int_{n_i}^{n_f} \sum_{j=1}^{m+p} a_j^c \Phi_j(x, n) dn. \quad (30)$$

Retrieval of the Absorption Index

An estimate of the absorption index can now be obtained. An initial guess of the absorption index is made and the scattering pattern is calculated through the use of Eq. (1). Calculation of the angular scattering cross sections from Eq. (1) requires that the particle number concentration be known. In the example inversion, it is assumed that the scattering pattern is due to a single particle, so N_c is known. When the scattering pattern is due to a distribution of particles, both the value of the absorption index and the value of the particle number concentration are varied until the calculated and measured scattering patterns match. If it is not possible to bring the measured and calculated scattering patterns into agreement by adjusting the value of the absorption index, the inversion process should be repeated with a different trial function. It should be noted that the calculated angular scattering cross sections were required to match the measurements that were not used as inputs in the inversion for the PSDF and the real part of the refractive index as well as the measurements that were used.

In the example inversion, the absorption index that gave the rms residual error closest to the rms of the estimated experimental errors, ΔC_j , was 10^{-3} . The measured and calculated scattering patterns are com-

pared in Fig. 4. The relatively large discrepancy between the calculated and measured 20° angular scattering cross sections indicated that the size parameter selected for the trial function was too large. The relatively good agreement between the calculated and measured scattering cross sections in the 100° – 150° range indicated that the retrieved value of the real refractive index was close to the actual value. Figure 2 is again used in the selection of a x_i value for a new trial function. The largest peak at a size parameter less than 25.1 is at $x = 20.1$. A new constrained solution is calculated as before. Using the new constrained solution, we obtain the closest agreement between the measured and calculated scattering patterns for an imaginary part of the refractive index of 10^{-4} . The new calculated scattering pattern compares well with the measured scattering pattern, as shown in Fig. 5.

The results of the example inversion are summarized in Table 3. A comparison of the retrieved and actual size and optical properties of the particle in the example indicates that the particle size and optical properties can be accurately retrieved from single particle light-scattering measurements. These results are representative of a number of inversions performed with simulated data sets.

Inversion of More Simulated Data Sets

Simulated data sets representative of the measurements obtainable with the 15-channel nephelometer were also inverted. Table 4 shows typical results from these inversions. To test the inversion process further, we were provided with six sets of simulated light-scattering measurements in a blind test.²⁵ We were told that the measurements corresponded to the light scattered by narrow distributions of nonabsorbing spheres, but no other information was given. The results of the inversions are shown in Table 5.

The actual PSDF used to generate the simulated data in the blind test is plotted in Fig. 6. Although the PSDF for case 4 has the same geometric standard deviation as the size distributions for cases 1 and 2, the PSDF for case 4 is actually broader than those for cases 1 and 2. These results show that the technique is successful when the distributions are narrow (cases 1, 2, and 6) but has difficulty when the distributions are broad (cases 3–5). These results also show the need to obtain reliable *a priori* information regarding the PSDF in order for one to invert light-scattering measurements successfully. In this study, it was assumed that the height of each distribution was greater than its width, and the trial functions used to constrain the solution were selected accordingly. In cases 3–5 the assumption of a narrow PSDF was not valid, and the retrieved PSDF did not resemble the actual PSDF. If more reliable information regarding the PSDF was available, the inversion process presented in this paper would probably give more accurate results in all cases. In practical applications, reliable *a priori* information may be obtained

from knowledge of the process that generated the particles or from mechanical sampling and analysis of the particles. This conclusion is in agreement with Koo,²⁶ who recommended the complementary use of laser light scattering and mechanical collection techniques after he reviewed particle sizing techniques used in the analysis of the metallic oxide smoke produced by the combustion of solid rocket propellants. Similar conclusions were reached by Bottiger²⁷ after he compared five different inversion techniques. Finally, it is interesting to note that even when the retrieved distributions differed from the actual distributions, the refractive index was retrieved accurately.

Conclusions and Recommendations

An inversion technique that retrieves the particle size distribution function and the refractive index of weakly absorbing spherical particles from simulated measurements of scattered light has been developed. The solution is obtained by the expansion of the distribution function as a linear combination of orthonormal basis functions. We use the orthogonality properties of the basis functions to find the expansion coefficients that minimize the residual errors subject to a trial function constraint. The technique is shown to be capable of retrieving the size and optical properties from simulated measurements of the light scattered by a weakly absorbing sphere.

We also used the technique to retrieve the PSDF and refractive index from simulated measurements of the light scattered by narrow log normal distributions of nonabsorbing spheres in a blind test. Attempts to retrieve the PSDF were less successful when the distributions were not narrow, but the refractive index was accurately retrieved in all cases. Because of the ill-posed nature of the inverse light-scattering problem, accurate *a priori* information regarding the PSDF must be available for the inversion technique developed in this study to be applied successfully to broad size distributions. If *a priori* information cannot be obtained from an analysis of the particular environment in which the light-scattering measurements are made, the complementary use of collection techniques is recommended. Further research is needed to investigate the possibility that the trial function could be selected with minimal reliance on *a priori* information. One possibility is the use of an iterative procedure that begins with a nonprejudicial trial function. Further modifications of the inversion technique will focus in this area.

Appendix A: Symbols Used in This Paper

a ,	Expansion coefficients for the generalized or unconstrained solution
a^c ,	Expansion coefficients for the constrained solution
a^t ,	Expansion coefficients for the trial function

C_{avg} ,	Average of the angular scattering cross section (cm^2)
c_{avg} ,	Normalized and imprecision weighted average angular scattering cross section (cm^{-2})
C ,	Angular scattering cross sections (cm^2)
c ,	Normalized and imprecision weighted angular scattering cross sections (cm^{-2})
$\tilde{f}(x, n, k)$,	Distribution of sizes and optical properties
$f(x)$,	Particle size distribution function
$f^t(x, n)$,	Trial function
$h(k)$,	Ratio of the scattering kernels evaluated at a finite value of k to the scattering kernels evaluated at k equal to zero
k ,	Absorption index
$\frac{d\tilde{C}}{d\Omega}(\Omega, x, n, k)$,	Differential scattering cross sections (cm^2)
$\frac{d\tilde{C}^{\text{avg}}}{d\Omega}(x, n, k)$,	Differential scattering cross sections that have been averaged over the solid angle subtended by the detectors (cm^2)
$\frac{dC^{\text{avg}}}{d\Omega}(x, n, k)$,	Imprecision weighted average differential scattering cross sections
m ,	Number of inputs
$\mathbf{M}$,	Kernel covariance matrix
n ,	Real part of the refractive index
N_c ,	Particle number density (cm^{-3})
p ,	Number of supplemental basis functions
RRV,	Residual relative variance
$\mathbf{u}$,	Eigenvectors of the kernel covariance matrix
V ,	Scattering volume (cm^3)
x ,	Size parameter
Greek	
δC ,	Experimental errors (cm^2)
δc ,	Normalized and imprecision weighted experimental errors (cm^{-2})
$\delta(x)$,	Dirac delta function
ΔC ,	Estimate of the experimental error in the j th measurement (cm^2)
Δf ,	Relative error in the retrieved distribution function
$\Delta\Omega$,	Solid angle subtended by a detector (sr)
λ ,	Eigenvalues of the kernel covariance matrix
$\Phi(x, n)$,	Basis functions
$\phi(x, n)$,	Supplemental basis functions

$\tilde{\phi}(x, n),$	Orthogonalized supplemental basis functions
$\Omega,$	Solid angle (sr)
Subscripts	
$f,$	Final
$i,$	Initial or index
$j,$	Index
$s,$	Solution
$t,$	Trial

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