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Inversion of light scattering measurements for particle size and optical constants

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University of Illinois at Urbana-Champaign, 1993
INVERSION OF LIGHT SCATTERING MEASUREMENTS FOR PARTICLE SIZE
AND OPTICAL CONSTANTS

BY

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B.S., Brigham Young University, 1988
M.S., University of Illinois, 1990

THESIS

Submitted in partial fulfillment of the requirements
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DOCTOR OF PHILOSOPHY

Director of Thesis Research

Head of Department

Committee on Final Examination†

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† Required for doctor's degree but not for master's.
Abstract

Light scattering techniques have proven to be valuable tools in determining the size and optical properties of aerosol particles. Laser light scattering techniques can be used unobtrusively in harsh environments, so the development and improvement of these techniques is of great interest. The primary difficulty associated with light scattering techniques is inverting the data or obtaining the desired information from the measurements.

The mathematical formulation of the light scattering problem results in a linear inverse problem with discrete data. Solution techniques for this class of problems are classified as either analytical or empirical. In this study, the inverse light scattering problem was solved for the particle size distribution function (PSDF) and the optical properties of aerosol particles using a combination of analytical and empirical techniques. The analytical solution was constrained using information obtained from the empirical solution method. Using synthetic data sets, it was demonstrated that the inversion technique is capable of inverting measurements of the light scattered by a weakly absorbing sphere. The technique was also used to retrieve the PSDF and refractive index from synthetic measurements of the light scattered by log normal distributions of non-absorbing spheres in a blind test. Attempts to retrieve the PSDF were unsuccessful when the distributions were broad, but the refractive index was accurately retrieved in all cases. The results of this study indicate that because of the ill-posed nature of the inverse light scattering problem, constraints based on a priori information regarding the unknown PSDF must be available to successfully apply the analytical inversion technique. If such information cannot be obtained from analysis of the experiment or use of the empirical method, the complementary use of collection techniques is recommended.

Light scattering measurements at 15 polar angles were obtained using a multi-channel polar nephelometer. Measurements of the light scattered by Freon-12 were used to calibrate the nephelometer for absolute scattering measurements. The light scattered by two different size polystyrene spheres was measured and inverted. In both cases, the particle size and the refractive index was accurately retrieved. Because the scattering measurements are fairly insensitive to variations in the absorption index when the particles are weakly absorbing, only an upper bound on the value of absorption index could be determined. Including a total extinction measurement or an absorption measurement in the inversion procedure may provide the additional information necessary to determine the absorption index.
Metallic oxide smoke produced in the combustion of metalized solid rocket propellants is spherical, so the inversion technique developed in this study can be used to analyze propellant smoke. The light scattered by Al$_2$O$_3$ smoke from an aluminized propellant was also measured and inverted. The optical properties retrieved through the inversion process agreed well with previously published results. Analysis of the smoke with the polar nephelometer required that the smoke be sampled and sent through tubing. Since the sampling method was not isokinetic and particle losses in the tubing were not accounted for, the PSDF of the particles in the nephelometer was not representative of the PSDF of the particles produced by the combustion process. However, histograms of the particle sizes in the sampled smoke were obtained using a laser aerosol spectrometer, and the PSDF retrieved by the inversion procedure compared well with the measured histograms. The results of these experiments demonstrated that the inversion technique developed in this study is a practical method of analyzing the smoke produced by solid propellants.
Acknowledgments

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I am most grateful for the unfailing kindness shown me by my parents, Mrs. Nancy S. Jones and Dr. Jerold W. Jones. I am particularly grateful to my father for encouraging me to follow in his footsteps and become an engineer.
# Table of Contents

<table>
<thead>
<tr>
<th>Chapter</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Important Symbols</strong></td>
<td></td>
<td>viii</td>
</tr>
<tr>
<td><strong>Chapter 1. Introduction</strong></td>
<td></td>
<td>1</td>
</tr>
<tr>
<td><strong>Chapter 2. Light Scattering Theory</strong></td>
<td></td>
<td>3</td>
</tr>
<tr>
<td>Extinction of an Electromagnetic Wave</td>
<td>3</td>
<td></td>
</tr>
<tr>
<td>Light Scattering by an Arbitrary Particle</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>Mie Theory</td>
<td>6</td>
<td></td>
</tr>
<tr>
<td>The Light Scattering Equation</td>
<td>6</td>
<td></td>
</tr>
<tr>
<td><strong>Chapter 3. Inverse Problems</strong></td>
<td></td>
<td>10</td>
</tr>
<tr>
<td>Ill-posed Nature of Inverse Problems</td>
<td>11</td>
<td></td>
</tr>
<tr>
<td>Analytical Solutions to Inverse Problems</td>
<td>12</td>
<td></td>
</tr>
<tr>
<td>Empirical Solutions to Inverse Problems</td>
<td>24</td>
<td></td>
</tr>
<tr>
<td>Summary</td>
<td>25</td>
<td></td>
</tr>
<tr>
<td><strong>Chapter 4. Inversion Procedure</strong></td>
<td></td>
<td>27</td>
</tr>
<tr>
<td>Preliminary Analysis of the Measurements</td>
<td>28</td>
<td></td>
</tr>
<tr>
<td>Input Selection</td>
<td>31</td>
<td></td>
</tr>
<tr>
<td>Retrieval of the Refractive Index</td>
<td>32</td>
<td></td>
</tr>
<tr>
<td>Retrieval of the PSDF</td>
<td>33</td>
<td></td>
</tr>
<tr>
<td>Retrieval of the Absorption Index</td>
<td>38</td>
<td></td>
</tr>
<tr>
<td>Summary of the Example Inversion</td>
<td>38</td>
<td></td>
</tr>
<tr>
<td>Inversion of More Synthetic Data Sets</td>
<td>41</td>
<td></td>
</tr>
<tr>
<td>Summary</td>
<td>42</td>
<td></td>
</tr>
<tr>
<td><strong>Chapter 5. Nephelometer Calibration</strong></td>
<td></td>
<td>45</td>
</tr>
<tr>
<td>Calibration for Absolute Scattering Measurements</td>
<td>46</td>
<td></td>
</tr>
<tr>
<td>Aerosol Light Scattering Measurements</td>
<td>68</td>
<td></td>
</tr>
</tbody>
</table>
## Important Symbols

- **$A_b$**: Cross sectional area of the incident laser beam (cm$^2$)
- **$A$**: Area of the detectors (cm$^2$)
- **$a$**: Expansion coefficients for the generalized or unconstrained solution
- **$a^c$**: Expansion coefficients for the constrained solution
- **$a^t$**: Expansion coefficients for the trial function
- **$BD$**: Laser power measured by the beam dump detector (V)
- **$C_{avg}$**: Average of the angular scattering cross section (cm$^2$)
- **$c_{avg}$**: Average of the normalized and imprecision weighted angular scattering cross sections (cm$^{-2}$)
- **$C$**: Angular scattering cross sections (cm$^2$)
- **$c$**: Normalized and imprecision weighted angular scattering cross sections (cm$^{-2}$)
- **$C_{sca}$**: Scattering cross section or the ratio of the power scattered by a particle to the incident irradiance (cm$^2$)
- **$d_{A1}$**: Diameter of the collimators (cm)
- **$d_{A2}$**: Diameter of the detector holder (cm)
- **$d_b$**: Diameter of the incident beam (cm)
- **$d_c(x^d)$**: Diameter of the acceptance cone (cm)
- **$\frac{d\tilde{C}}{d\Omega}(\Omega,x,n,k)$**: Differential scattering cross sections (cm$^2$)
- **$\frac{dC_{avg}}{d\Omega}(x,n,k)$**: Differential scattering cross sections that have been averaged over the solid angle subtended by the detectors (cm$^2$)
- **$\frac{dC_{avg}}{d\Omega}(x,n,k)$**: Imprecision weighted average differential scattering cross sections (cm$^2$)
- **$d_d$**: Detector diameter (cm)
- **$d_F$**: Effective scattering diameter for Freon-12 (cm)
- **$d_j$**: Aerosol jet diameter (cm)
- **$\tilde{f}(x,n,k)$**: Distribution function
- **$f(x)$**: Particle size distribution function (PSDF)
- **$f'(x,n)$**: 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
- **$h_b(x^b)$**: Height of the laser beam (cm)
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>$h_c(x_d,y_d)$</td>
<td>Height of the acceptance cone (cm)</td>
</tr>
<tr>
<td>$I_i$</td>
<td>Incident irradiance (W/cm²)</td>
</tr>
<tr>
<td>$k$</td>
<td>Absorption index</td>
</tr>
<tr>
<td>$l$</td>
<td>Path length (cm)</td>
</tr>
<tr>
<td>$m$</td>
<td>Number of inputs</td>
</tr>
<tr>
<td>$M_{ij}$</td>
<td>Measurement obtained from the $j^{th}$ detector at the $i^{th}$ laser power level (V)</td>
</tr>
<tr>
<td>$M^c$</td>
<td>Corrected measurement</td>
</tr>
<tr>
<td>$M$</td>
<td>Kernel covariance matrix</td>
</tr>
<tr>
<td>$n$</td>
<td>Complex refractive index = $n - ik$</td>
</tr>
<tr>
<td>$n$</td>
<td>Real part of the refractive index</td>
</tr>
<tr>
<td>$N_c$</td>
<td>Particle number concentration (cm⁻³)</td>
</tr>
<tr>
<td>$N_A$</td>
<td>Avogadro's number = $6.02 \times 10^{23}$</td>
</tr>
<tr>
<td>$p$</td>
<td>Number of laser power levels</td>
</tr>
<tr>
<td>$r$</td>
<td>Position vector (cm)</td>
</tr>
<tr>
<td>$R_i$</td>
<td>Distance from the center of the scattering volume to the opening of the collimator (cm)</td>
</tr>
<tr>
<td>$R_o$</td>
<td>Distance from the center of the scattering volume to the detectors (cm)</td>
</tr>
<tr>
<td>$r^*$</td>
<td>Maximum distance from the $x_d$ axis at which the entire detector is visible (cm)</td>
</tr>
<tr>
<td>RRV</td>
<td>Residual relative variance</td>
</tr>
<tr>
<td>SCF</td>
<td>Scattering correction factors</td>
</tr>
<tr>
<td>SL</td>
<td>Stray light (V)</td>
</tr>
<tr>
<td>$u$</td>
<td>Eigenvector of the kernel covariance matrix</td>
</tr>
<tr>
<td>$V$</td>
<td>Scattering Volume (cm³)</td>
</tr>
<tr>
<td>$V_F$</td>
<td>Volume of the Freon-12 molecules (cm³)</td>
</tr>
<tr>
<td>$W_{ext}$</td>
<td>Power removed from the incident laser beam (W)</td>
</tr>
<tr>
<td>$W_i$</td>
<td>Power of the incident laser beam (W)</td>
</tr>
<tr>
<td>$W_s$</td>
<td>Power scattered from the incident laser beam (W)</td>
</tr>
<tr>
<td>$x$</td>
<td>Size parameter</td>
</tr>
</tbody>
</table>

**Greek**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha$</td>
<td>Acceptance angle of the detectors (rad)</td>
</tr>
</tbody>
</table>
\( \beta \)  
Attenuation coefficient for the medium (1/cm)

\( \delta C \)  
Experimental errors (cm²)

\( \delta c \)  
Normalized and imprecision weighted experimental errors (cm⁻²)

\( \delta(x) \)  
Dirac delta function

\( \Delta C \)  
Estimate of the experimental errors (cm²)

\( \Delta f \)  
Relative error in the retrieved distribution function

\( \Delta \Omega \)  
Solid angle subtended by a detector (sr)

\( \Phi(x,n) \)  
Basis functions

\( \phi(x,n) \)  
Supplemental basis functions

\( \tilde{\phi}(x,n) \)  
Orthogonalized supplemental basis functions

\( \phi \)  
Azimuthal angle (rad)

\( \lambda \)  
Wavelength of the laser (µm) or eigenvalues of the kernel covariance matrix

\( \theta \)  
Polar or scattering angle (rad)

\( \rho \)  
Depolarization factor

\( \Omega \)  
Direction

Superscripts

<table>
<thead>
<tr>
<th>Superscript</th>
<th>Meaning</th>
</tr>
</thead>
<tbody>
<tr>
<td>b</td>
<td>Base coordinates</td>
</tr>
<tr>
<td>d</td>
<td>Detector coordinates</td>
</tr>
<tr>
<td>p</td>
<td>Particle coordinates</td>
</tr>
<tr>
<td>t</td>
<td>Trial</td>
</tr>
<tr>
<td>^</td>
<td>Unit vector</td>
</tr>
</tbody>
</table>

Subscripts

<table>
<thead>
<tr>
<th>Subscript</th>
<th>Meaning</th>
</tr>
</thead>
<tbody>
<tr>
<td>F</td>
<td>Freon-12</td>
</tr>
<tr>
<td>i</td>
<td>Index or laser power level</td>
</tr>
<tr>
<td>j</td>
<td>Index or detector number</td>
</tr>
<tr>
<td>p</td>
<td>Particle</td>
</tr>
<tr>
<td>s</td>
<td>Scattering</td>
</tr>
<tr>
<td>t</td>
<td>Trial</td>
</tr>
<tr>
<td>t\text{stp}</td>
<td>Standard temperature (25°C) and pressure (1 atm)</td>
</tr>
<tr>
<td>ref</td>
<td>Reference</td>
</tr>
</tbody>
</table>

Prefix

<table>
<thead>
<tr>
<th>Prefix</th>
<th>Meaning</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \delta )</td>
<td>Uncertainty</td>
</tr>
</tbody>
</table>
Chapter 1
Introduction

Measurements of the radiation scattered by a sample contains 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 fact that a recent issue of Applied Optics [Bohren and Hirlaman, 1991] was devoted entirely to optical particle sizing techniques illustrates the tremendous interest in developing and improving these techniques. 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 using 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. Since mathematically similar problems occur in many scientific disciplines, techniques for solving linear inverse problems have been thoroughly investigated. Twomey (1977) gave a detailed development of the mathematics of inversion and described many of the schemes used to solve inverse problems. More recently, Bertero et al. (1985b, 1988) published a discussion of the general formulation of this class of problems and reviewed more solution methods.

In general, techniques used to invert light scattering measurements can be classified as either analytical or empirical. Analytical techniques involve formal solutions of an integral equation which describes the light scattering or extinction process to obtain the unknown distribution function. Since the information content in a set of light scattering or extinction measurements is limited [Twomey 1974, 1977; Capps et al., 1982; Ben-David et al., 1988], inverse problems do not usually 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 and/or careful optimization of the inputs. Indeed, the primary difference between
most analytic inversion schemes is the way the *a priori* information is incorporated or the inputs are optimized. Also, most of these 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 to obtain a least squares fit of the measured data. A unique empirical inversion technique known as the optical strip-map technique has been developed by Quist and Wyatt (1985). 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 [Quist and Wyatt, 1985; Bottiger, 1991]. However, this technique does not provide any information regarding the absorption index, and it cannot be extended to measurements of the light scattered by polydispersions.

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 and retrieving the particle size distribution function (PSDF) and optical properties. After a brief discussion of the fundamentals of light scattering theory in Chapter 2, Chapter 3 presents methods for solving linear inverse problems with discrete data without reference to a particular application. A combination of the techniques described in Chapter 3 are then applied to the specific problem of determining the PSDF and optical properties of aerosols in Chapter 4. The capabilities of the inversion technique are investigated using synthetic data sets, and the implications of these results are discussed. The ability to make absolute light scattering measurements is necessary to apply the inversion technique described in Chapter 4, so a procedure to calibrate multi-channel polar nephelometers for absolute light scattering measurements was developed. A description of the multi-channel polar nephelometer and the details of the calibration procedure are presented in Chapter 5. In Chapter 6, the inversion technique is applied to measurements of the light scattered by polystyrene spheres and to measurements of the light scattered by Al₂O₃ smoke that was produced by burning an aluminized solid rocket propellant.
As an electromagnetic wave propagates through a medium, the wave is attenuated due to the heterogeneity of the medium. Since all media are heterogeneous on some scale, all media will attenuate electromagnetic radiation. The attenuation of electromagnetic energy is known as extinction and is the result of two different mechanisms: absorption and scattering. Energy that is absorbed is converted into some other form such as thermal or chemical energy, while the energy that is scattered is merely redirected and remains electromagnetic in form. Practical applications of light scattering and absorption exist in most scientific disciplines, so the study of the extinction of electromagnetic radiation is of enormous importance and has been thoroughly treated in the literature. This dissertation presents a technique for determining the size and optical properties of homogeneous, spherical particles from their light scattering patterns, so a brief introduction to the basics of light scattering theory is necessary. More complete developments of the theory of light scattering are given in the classic book by van de Hulst (1981), and in a more recent book by Bohren and Huffman (1983).

Extinction of an Electromagnetic Wave

A basic understanding of the physics of extinction can be obtained by considering the interaction of an electromagnetic wave with an arbitrary medium. From a classical perspective, all matter contains discrete electric charges which are set in motion by the oscillations of an electric field. Some of the electromagnetic energy in the beam will be lost due to absorption as the charged particles collide with each other and with uncharged particles, and some will be scattered or reradiated in different directions due to the acceleration of the charged particles. The classical theory of emission of electromagnetic radiation by an accelerated charge was first developed by J. J. Thomson in 1903 and is presented in Appendix A.

The frequency of the scattered electromagnetic wave is generally the same as the frequency of the incident wave. This is referred to as elastic scattering. However, there are phenomena such as Raman scattering or Compton scattering where the scattering is inelastic, or the frequency of the scattered wave is not the same as that of the incident wave. The Raman effect is observed when intense and highly monochromatic light is scattered by molecules. Measurements show the scattered light is composed mainly of radiation with the same frequency as the incident light, but the scattered light also contains low levels of radiation with slightly different frequencies. During the scattering process, some of the
molecules may change rotational or vibrational energy states, and the energy of the scattered radiation must change by an equal and opposite amount in order to conserve energy. Compton scattering is most easily understood if the incident light is viewed as a stream of photons. The Compton effect occurs when a high energy photon collides with a free electron. Since a photon carries momentum, conservation of momentum requires that the electron recoil after deflecting the photon. Conservation of energy then requires that the energy of the photon be reduced by an amount equal to the kinetic energy of the recoiling electron. Thus, the frequency of the scattered electromagnetic wave must be lower than that of the incident wave. Both Raman and Compton scattering illustrate that scattering and absorption are not mutually independent processes. Although one process may be much more significant than the other, both are always present. Both the Raman and Compton effects are commonly referred to as scattering processes because most of the energy is scattered or reradiated, but absorption processes also always occur.

Light Scattering by an Arbitrary Particle

Consider a beam of electromagnetic energy incident on an arbitrary particle. The superposition of the incident wave and of all the secondary, reradiated waves gives the total scattered electric field. Naturally, the differences in phase and amplitude between the incident wave and all the scattered waves must be properly accounted for when superposing the various waves to obtain the total scattered field. In general, the phase differences and amplitudes will vary with the scattering direction, so the scattering pattern will vary with direction. The total scattered field also depends on the size of the particle. The larger the particle, the greater the possibility of mutual enhancement or cancellation due to phase differences between the incident wave and the various scattered waves. Therefore, the larger the particle, the more maxima and minima in the scattering pattern. When a particle is small relative to the wavelength of the incident beam, the oscillations of the discrete charges within the particle are approximately in phase, and the scattered field does not vary as much with direction. Clearly, the shape of the particle will affect these phase relationships as well, and, if the particle is not spherical, the orientation of the particle will affect the phase relationships. Hence, the scattering pattern will depend on the shape and orientation of the particle. Since the reradiated energy flux is proportional to the square of the acceleration of the charged particle (See Appendix A), the strength of the bonds holding the charged particle in place clearly affect the amplitude and the phase of the scattered radiation. Thus, the scattered field will depend on the composition of the particle. This dependency is expressed through the optical properties of the material which, in general, depend on the frequency and state of polarization of the incident beam. In
summary, the scattered field varies with the scattering direction, with the size, shape, orientation, and optical properties of the particle and with the frequency, irradiance, and polarization of the incident beam.

Since the problem of determining the light scattered by a particle depends in a detailed way on the intensity, wavelength, and polarization state of the incident wave as well as the size, shape and composition of the particle, the problem is hopelessly complex unless several restrictive assumptions are made. The development of the light scattering theory used in this dissertation will rely on the following assumptions. The first restriction imposed is that only elastic scattering will be considered. A second assumption is that if the scattering is the result of more than one particle, the particles are independent. Since waves scattered in the same direction from different particles have the same wavelength, they must interfere constructively or destructively. However, if there is no systematic relationship between the position of the particles, the net effect is that the waves scattered by the various particles may simply be added without regard to their phase. This is the definition of independent scattering. The third restriction is that if the scattering is the result of many particles, the multiple scattering effects can be neglected. This assumption requires that the amount of scattered light incident on each particle be negligible or that the electromagnetic wave incident on each particle is essentially the original electromagnetic wave. Although the physics involved in multiple scattering is the same as in single scattering, multiple scattering results in coupled equations and a tremendous increase in the mathematical complexity of the problem. The fourth and last assumption is that the incident beam is monochromatic or that the incident wave is a plane harmonic wave. Since Fourier analysis can decompose any arbitrary wave into plane harmonic waves and all the operations involved are linear, this assumption is not as restrictive as it might initially appear. The light scattering problem can be solved for an arbitrary incident wave by decomposing the incident wave into its Fourier components and superimposing the results obtained for each component. These assumptions reduce the light scattering problem to that of finding the electromagnetic field inside a particle of known properties and in the medium surrounding the particle when the particle is illuminated by a monochromatic light wave from a known source.

Due to the mathematical complexity, analytical solutions to the light scattering problem have only been obtained for particles with relatively simple geometries and properties. The most important of the analytical solutions, Mie theory, gives the electromagnetic field scattered by a homogeneous, isotropic sphere of arbitrary radius and refractive index.
Mie Theory

In 1908 Gustav Mie developed a theory in an effort to explain the various colors present in the light scattered by colloidal particles of gold suspended in water. The result of these efforts is the mathematical formalism necessary to describe the light scattered by a sphere and is commonly referred to as Mie theory in his honor. However, L. Lorenz may have actually been the first person to construct a solution to the problem of light scattering by a sphere, so the theory is sometimes referred to as Lorenz-Mie theory. Mie theory gives the internal and scattered fields as infinite series expansions. A complete derivation is presented in chapter 4 of Bohren and Huffman (1983), and a helpful overview is presented in chapter 9 of van de Hulst (1981). A brief overview of Mie theory and the equations necessary to calculate differential scattering cross sections and the phase function are given in Appendix B. Differential scattering cross sections serve as kernels in the integral equation that relates the light scattering measurements to the distribution of particle sizes and refractive indices. The light scattering equation is discussed in the following section.

The Light Scattering Equation

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, 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.

$$C_j = \int \int \int \int \int N_c \tilde{f}(x, n, k) \frac{d\tilde{C}_j}{d\Omega}(\Omega, x, n, k) dx dkd\Omega dV + \delta C_j \quad (2.1)$$

If the particles are homogenous spheres, the differential scattering cross sections, $\frac{d\tilde{C}_j}{d\Omega}(\Omega, x, n, k)$, can be calculated from Mie theory. The following assumptions reduce the complexity of Equation 2.1.

1. Figure 2.1 shows that for values of $k$ less than approximately $10^{-2}$, the shape the scattering pattern does not change as $k$ is varied.
Figure 2.1. Effect of the Absorption Index on the Differential Scattering Cross Sections

Size parameter = 25
Refractive index = 1.5
Therefore, 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 if the particles are weakly absorbing.

\[
\frac{d\tilde{C}_i}{d\Omega}(\Omega, x, n, k) \approx h(k)\frac{d\tilde{C}_i}{d\Omega}(\Omega, x, n, 0)
\]  

(2.2)

(2) If a distribution of particles is present, all the particles have the same optical properties.

\[
\tilde{f}(x, n, k) = f(x)\delta(n - n_s)\delta(k - k_s)
\]  

(2.3)

(3) Again, if a distribution of particles is present, the particle number density is uniform over the scattering volume.

(4) The solid angles subtended by the detectors are small, so the integral over \(\Omega\) can be replaced by the product of the average of the differential scattering cross sections and \(\Delta\Omega\).

\[
\int_{\Delta\Omega_j} \frac{d\tilde{C}_i}{d\Omega}(\Omega, x, n, k)d\Omega = \frac{d\tilde{C}_{i\text{avg}}}{d\Omega}(x, n, k)\Delta\Omega_j
\]  

(2.4)

These assumptions simplify Equation 2.1 to

\[
C_j = N_c V_j \Delta\Omega_j h(k_s) \int_{n_i}^{n_f} \int f(x)\delta(n - n_s) \frac{d\tilde{C}_{i\text{avg}}}{d\Omega}(x, n, 0)dxdn + \delta C_j
\]  

(2.5)

Neglecting the error in the measurements, the average of the measurements can be approximated by the average of the right hand side of Equation 2.5.

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

\[
C_{\text{avg}} = \frac{N_c V_j h(k_s)}{m} \sum_{j=1}^{m} \Delta\Omega_j \int_{n_i}^{n_f} \int f(x)\delta(n - n_s) \frac{d\tilde{C}_{i\text{avg}}}{d\Omega}(x, n, 0)dxdn
\]  

(2.6)
The unknown function, \( h(k) \), can be eliminated from Equation 2.5 by normalizing by the average of the measurements. Earlier investigations have also shown that it is beneficial to weight each measurement and scattering kernel by the corresponding imprecision estimate [Curry, 1989; King, 1978]. Imprecision weighting makes the inversion process more stable by making the magnitude of each measurement and the magnitude of each scattering kernel approximately equal. After these modifications, Equation 2.5 reduces to

\[
c_j = \frac{1}{c_{\text{avg}} n_i x_i} \int f(x) \delta(n - n_s) \frac{dC_{\text{avg}}}{d\Omega}(x, n, 0) dx dn + \delta c_j \tag{2.7}
\]

where

\[
c_j = \frac{C_j}{C_{\text{avg}} \Delta C_j} \tag{2.8}
\]

\[
\delta c_j = \frac{\delta C_j}{C_{\text{avg}} \Delta C_j} \tag{2.9}
\]

\[
\frac{dC_{\text{avg}}}{d\Omega}(x, n, 0) = \frac{\delta c_j}{\Delta C_j} \tag{2.10}
\]

\[
c_{\text{avg}} = \frac{1}{m} \sum_{j=1}^{m} \int f(x) \delta(n - n_s) \frac{dC_{\text{avg}}}{d\Omega}(x, n, 0) dx dn \tag{2.11}
\]

Equation 2.7 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 proceeding section fits the definition of a linear inverse problem with discrete data [Bertero et al., 1985b, 1988]. Linear inverse problems occur in wide variety of scientific disciplines, so practical techniques for solving these problems are of great interest. Chapter 3 discusses the general mathematical formulation of inverse problems and describes several of the inversion techniques that have been used in optical particle sizing.
Chapter 3
Inverse Problems

From the discussion of light scattering theory in the previous chapter, it is clear that given the size, shape, orientation, and optical properties of a particle that is illuminated by an electromagnetic wave of known wavelength, irradiance, and polarization, the angular scattering pattern can be determined. This is known as the direct light scattering problem. It also seems reasonable to expect that a solution of the inverse problem can be found or that the characteristics of the particle can be found from measurements of the angular scattering pattern. The direct problem has been compared to describing the tracks left by a dragon given a complete description of the dragon. The inverse light scattering problem would then be compared to finding a description of the dragon based on an analysis of its tracks [Bohren and Huffman, 1983].

The inverse light scattering problem is one application of a broad range of problems known generally as linear inverse problems with discrete data. These problems are characterized by the following equation.

\[ y_j = O_j g + \delta y_j \quad \text{where } j = 1, \ldots, m \]  

(3.1)

In Equation 3.1, the measurement set, \( \{y_j\} \), is related to an unknown function \( g \) by an operator \( O_j \), and the \( \delta y_j \) represent errors in the measurements. Experiments in engineering or the natural sciences often result in data that is indirectly related to the unknown characteristic or property of a system. These indirect relationships can usually be represented by equations identical in form to Equation 3.1, so linear inverse problems with discrete data occur in most scientific disciplines. In general, these problems do not possess a unique solution, so they are ill-posed.

Comparison of Equation 3.1 with Equation 2.7 shows that the light scattering equation derived in Chapter 2 fits the definition of a linear inverse problem with discrete data if \( O_j \) is the following linear integral operator.

\[ O_j g = \frac{1}{c_{avg}} \int \int_{n_1 x_1} \frac{dC_{avg}}{d\Omega}(x,n,0)dxdn \]  

(3.2)

Due to the wide variety of applications, techniques for solving inverse problems have received and continue to receive considerable attention. Even a cursory discussion of the majority of inversion schemes would be prohibitively long, so only a representative
sample of inversion techniques that have been applied to optical particle sizing techniques will be discussed in this chapter. Turchin et al. (1971), Twomey (1977), Ishimaru (1978) and Bertero et al. (1985b, 1988) provide a thorough development of the mathematics of inverse problems and detailed discussions of many more inversion techniques. The literature is full of examples of applications of inversion techniques to various specific problems. A recent edition of Applied Optics was devoted entirely to optical particle sizing [Bohren and Hirleman, 1991], and the articles in that feature issue are representative of the current state of the art in this area.

In general, techniques for solving inverse problems can be categorized as either analytical or empirical. Analytical techniques involve formal solutions of Equation 3.1. The analytical inversion schemes that are commonly used in particle sizing applications are discussed in the following section. Empirical inversion techniques are generally iterative schemes that involve the development of a model of the physical process represented by Equation 3.1. Calculations based on this model are repeated until the calculations match the measurements. A unique non-iterative empirical particle sizing technique known as the optical strip-map technique has been developed by Quist and Wyatt (1985). A description of the optical strip-map technique will be given later in this chapter.

**Ill-posed Nature of Inverse Problems**

The ill-posed nature of the problem is illustrated by considering the case when \( O_j \) is a linear integral operator. Equation 3.1 then becomes

\[
y_j = \int_{z_i}^{z_f} g(z) K_j(z) dz + \delta y_j \quad \text{where } j = 1, ..., m
\]  

(3.3)

Assuming there are no singularities in \( K_j(z) \) is on the interval \([z_i, z_f]\), the Riemann-Lebesgue lemma [Rudin, 1966; Provencher, 1982] says that

\[
\lim_{\omega \to \infty} \int_{z_i}^{z_f} \sin(\omega z) K_j(z) dz = 0
\]  

(3.4)

Therefore,

\[
y_j = \lim_{\omega \to \infty} \int_{z_i}^{z_f} \left[ (g(z) + A \sin(\omega z)) K_j(z) dz + \delta y_j \right]
\]  

(3.5)
Equation 3.5 shows that even for arbitrarily small measurement errors and arbitrarily large A, there is a $\omega$ such that $g(z) + A\sin(\omega z)$ will satisfy Equation 3.3. Thus, it is clear that there are an infinite number of solutions to Equation 3.3, and each of these solutions can have arbitrarily large deviations from one another and from the true solution.

In a well designed experiment, the signal to noise ratio is large, so the $\delta y_j$ are negligible compared to $y_j$. If the $\delta y_j$ can be neglected, the inverse operator for $f_j$ can be found in most cases, and an analytic solution can be obtained. However, considering the infinite number of possible solutions and the possibility that each of these solutions may deviate substantially from the true solution, it is unlikely that the retrieved function will bear much resemblance to $g$. Each of the possible solutions satisfy Equation 3.1 to within the uncertainty in the measurements, so they are mathematically correct. However, many of the possible solutions will demonstrate behavior that allows them to be rejected on the basis of physical considerations. Using these physical considerations, a priori constraints can be formulated and used to eliminate many possible solutions. For example, if $g$ corresponds to a particle size distribution function, it is reasonable to require that the solution be smooth and non-negative. Schemes which do not require the use of such a priori information have been used in particle sizing applications. The Backus-Gilbert method is one such technique, and a description of this technique is included for this reason. Although the Backus-Gilbert method has been successfully applied to particle sizing by at least one research group [Westwater and Cohen, 1973], other researchers have found the Backus-Gilbert technique tends to be less stable in the presence of experimental errors than other methods [Chow and Tien, 1976; Twomey, 1977]. Therefore, the key to successfully solving an inverse problem with an analytical inversion scheme is properly applying one or more constraints based on a priori information that will lead to the selection of the solution from the set of possible solutions that most closely resembles $g$.

Analytical Solutions to Inverse Problems

It has been shown that mathematical solutions to the inverse problems encountered in practice are usually not unique, so direct solution methods will probably lead to a physically unrealistic solution. Therefore, most analytic inversion techniques require the use of a priori information regarding the unknown function $g$ and careful optimization of the measurements $y_j$. Indeed, the primary difference between most analytic inversion schemes is the way the a priori information is incorporated or the inputs are optimized. The first analytical inversion technique described in this chapter, the Backus-Gilbert method, does not require a priori information regarding the unknown
function. The other inversion schemes discussed in this chapter illustrate how a priori information regarding the unknown function and the measurements can be incorporated into the inversion process.

Discrete Formulation of Inverse Problems

When discussing analytical inversion techniques, it is often helpful to reformulate equations similar to Equation 3.3 as matrix equations. In practice, integrals are usually evaluated using numerical quadrature, so they are generally rewritten as sums. The discrete formulation of Equation 3.3 is

\[ y_j - \delta y_j = y_j = \sum_{i=1}^{m} t_i g(z_i) K_j(z_i) \]  

where the \( t_i \) depend on the quadrature formula that is used. In vector notation, Equation 3.6 simplifies to

\[ y^d = T g \]  

where

\[ T_{ij} = t_i K_j(z_i) \]  

The Backus-Gilbert Inversion Technique

The application of the Backus-Gilbert technique to particle sizing has been discussed by several authors [Westwater and Cohen, 1973; Chow and Tien, 1976; Twomey, 1977; Ishimaru, 1978]. Rather than determining the function \( g(z) \), the Backus-Gilbert method calculates weighted local averages of \( g \) at a discrete number of \( z_j \).

\[ \overline{g}(z_j) = \int_{z_j}^{z_f} S(z_j, z) g(z) dz \]  

where \( j = 1, \ldots, m \)

The function \( S(z_j, z) \) is known as a scanning function or as an averaging kernel. The scanning function is small except near \( z = z_j \) and is normalized on the interval \([z_i, z_f]\).

\[ \int_{z_i}^{z_f} S(z_j, z) dz = 1 \]  

13
Ideally, the scanning function would equal $\delta(z_j - z)$, and then $g(z_j)$ would then be exactly equal to $g(z_j)$. In the Backus-Gilbert technique, the scanning function is given by an linear expansion of the kernels in Equation 3.3.

$$S(z_j, z) = \sum_{i=1}^{m} a_i(z_j)K_i(z)$$  \hspace{1cm} (3.11)

The expansion coefficients, $a_i(z_j)$, are calculated in the following manner. Substitution of Equation 3.11 into Equation 3.9 gives

$$g(z) = \int_{z_1}^{z_f} \sum_{i=1}^{m} a_i(z_j)K_i(z)g(z)dz$$

$$g(z_j) = \sum_{i=1}^{m} a_i(z_j)y_i^d$$

$$\overline{g} = Ay^d = A(y - \delta y)$$  \hspace{1cm} (3.12)

where $A_{ij} = a_i(z_j)$. From Equation 3.12, it is clear that if the magnitude of the $a_i(z_j)$ are large, the errors in $\overline{g}$ due to errors in the measurements will be greatly amplified. Therefore, the following constraint is imposed on the sum of the squares of the $a_i(z_j)$.

$$a^*a = a_{\text{max}}^2$$  \hspace{1cm} (3.13)

Substitution of Equation 3.11 into Equation 3.10 gives another constraint on the $a_i(z_j)$.

$$\int_{z_1}^{z_f} \sum_{i=1}^{m} a_i(z_j)K_i(z)dz = 1$$

$$a^*b = b^*a = 1$$  \hspace{1cm} (3.14)

where
Finally, the expansion coefficients must be selected such that the scanning function is concentrated near \( z_i \). A quantity known as the spread is defined as a measure of the concentration of \( S(z_i, z) \) near \( z_i \). The smaller the spread, the greater the concentration of the scanning function near \( z_i \).

\[
s(z_j) = 12 \int_{z_i}^{z_f} (z_j - z)^2 S^2(z_j, z) \, dz
\]

\[
s(z_j) = 12 \int_{z_i}^{z_f} (z_j - z)^2 \sum_{i=1}^{m} a_i(z_j) K_i(z) \sum_{p=1}^{m} a_p(z_j) K_p(z) \, dz
\]

\[
s(z_j) = a^* \tilde{S} a
\]

where

\[
\tilde{S}_{pq}(z_j) = 12 \int_{z_i}^{z_f} (z_j - z)^2 K_p(z) K_q(z) \, dz
\]

The \( a_i(z_j) \) are now determined by minimizing Equation 3.16 subject to the constraints imposed by Equations 3.13 and 3.14. The minimum in the spread is found using the method of Lagrange multipliers. A performance function is defined as

\[
Q(z_j) = a^* \tilde{S} a + \alpha (a^* a - a_{\max}^2) + 2\beta (a^* b - 1)
\]

Setting \( \frac{\partial Q}{\partial a_i} \) equal to zero gives

\[
a = -\beta (\tilde{S} + \alpha I)^{-1} b
\]

The second Lagrange multiplier, \( \beta \), is found by substituting Equations 3.11 and 3.19 into Equation 3.10.
The first Lagrange multiplier, $\alpha$, is determined by balancing the spread with the amplification of the experimental errors. As $\alpha$ is increased, the magnitude of the $a_i(z_j)$ decreases, so the magnification of the errors in the measurements decreases (see Equation 3.12). However, the spread is proportional to $\alpha$, so the increase in accuracy is paid for by a degradation in the resolution. Twomey (1977) found that for values of $\alpha$ below a certain cutoff value, the decrease in the spread was insignificant compared to the increase in the error magnification. Therefore, this optimal or cutoff value of $\alpha$ can be determined by calculating the spread from Equation 3.16 and $a^*a$ for several values of $\alpha$. Comparison of these values will serve as a guide in selecting the proper value of $\alpha$.

The Backus-Gilbert technique is advantageous in that no a priori information is required, and information regarding the resolution of the retrieved values is obtained. However, this technique is computationally intensive since $(S+\alpha I)$ must be inverted for each value of $\alpha$ and for each $z_j$ at which the solution is obtained. Chow and Tien (1976) found that the Backus-Gilbert method is more sensitive to errors in the measurements than other inversion schemes. Therefore, this method is only recommended if it is not possible to obtain a priori information, and the measurement errors are expected to be small. The Backus-Gilbert method may also be useful in obtaining information needed to select constraints for other analytical inversion techniques.

**Statistical Inversion Techniques**

Due to the interdependence of the kernels encountered in most practical problems, the matrix, $T$, defined in Equation 3.8 is generally nearly singular. Therefore, simply solving for $g$ by calculating $T^{-1}y^d$ does not yield satisfactory results. However, if information regarding the statistical nature of the unknown $g$ and the measurement errors $\delta y$ is available, a matrix $B$ can be found such that

$$g = By^d$$

(3.21)

$B$ is found by minimizing the performance function given in Equation 3.22.

$$Q = \{(g - By^d)^*v\}^\dagger\{(g - By^d)^*v\}$$

(3.22)
where \( v \) is an arbitrary vector. Expanding the right hand side of Equation 3.22 gives

\[
Q = v^*R_{gg}v - v^*R_{gg}w - w^*R_{gg}v + w^*R_{gg}w
\]  
(3.23)

where

\[
w = B^*v
\]  
(3.24)

\[
R_{gg} = gg^*
\]  
(3.25)

\[
R_{gg} = R_{gg}^* = gy^d^*
\]  
(3.26)

\[
R_{g} = y^d_y^d^*
\]  
(3.27)

Equation 3.23 is now rewritten as

\[
Q = \left[w - R^{-1}_{yy}R_{gg}v\right]^*R_{yy}^*\left[v - R^{-1}_{yy}R_{gg}w\right] + v^*\left(R_{gg} - R_{gg}^*R^{-1}_{yy}R_{yy}^*R_{gg}\right)
\]  
(3.28)

It can be shown that first term in Equation 3.28 is non-negative, and the second term is always positive [Ishimaru, 1978]. Therefore, Equation 3.28 is a minimum when \( w \) is chosen such that the first term is equal to zero.

\[
w = R^{-1}_{yy}R_{gg}v
\]  
(3.29)

The desired matrix \( B \) is now obtained by combining Equations 3.24 and 3.29.

\[
B = R_{gg}^*\left(R_{yy}^*\right)^{-1}
\]  
(3.30)

Note that

\[
R_{gg}^* = R_{gg}^* = g(T_g + \delta_y)^*
\]
\[ R_{y'y'}^* = R_{y'y'} + R_{\delta y} \] (3.31)

and

\[ R_{y'y'} = (Tg + \delta y)(Tg + \delta y)^* \]

\[ R_{y'y'}^* = TR_{y'y'}^* + R_{\delta y} + TR_{\delta y} + R_{\delta y} \] (3.32)

where

\[ R_{\delta y} = R_{\delta y}^* = g\delta y^* \] (3.33)

If the unknown function and the measurement errors are uncorrelated, the covariance matrix defined in Equation 3.33 is 0, and \( B \) can be rewritten as

\[ B = R_{y'y'}(TR_{y'y'}^* + R_{\delta y\delta y})^{-1} \] (3.34)

Equation 3.34 shows that the covariance matrices \( R_{y'y'} \) and \( R_{\delta y\delta y} \) are required to implement this statistical inversion technique. If information regarding the statistical properties of the unknown function and the measurement errors is available, Equation 3.34 provides a simple, direct method of obtaining a solution.

The Phillips-Twomey Inversion Technique

The Phillips-Twomey inversion technique is often referred to as constrained linear inversion [Phillips, 1962; Twomey, 1977]. In order to implement this technique, an upper bound on the sum of the squares of the measurement errors is assumed.

\[ \epsilon^2 = |\delta y|^2 \geq |Tg - y|^2 \] (3.35)

Of all the possible \( g \) which satisfy Equation 3.35, the \( g \) that best satisfies an \textit{a priori} constraint is selected as the solution. The constraint is imposed by minimizing a performance function using the method of Lagrange multipliers. The performance function is defined by
\[ Q = |Tg - y|^2 + \gamma g'Hg \]  

\[ (3.36) \]

where \( H \) depends on the constraint that is enforced. Although there are many possible constraints, a commonly imposed constraint is that \( g \) be the smoothest of all the possible solutions. The second differences of \( g \) are generally used as a measure of the smoothness of the solution. \( H \) would then be given by

\[ H = K'K \]  

\[ (3.37) \]

where \( K \) is the following tri-diagonal matrix.

\[ K = \begin{bmatrix} 0 & 1 & 0 & \ldots & 0 \\ 1 & -2 & 1 & \ldots & 0 \\ 0 & 1 & -2 & 1 & 0 \\ 0 & 0 & 1 & -2 & 1 \end{bmatrix} \]  

\[ (3.38) \]

The solution is then obtained by requiring that

\[ \frac{\partial Q}{\partial g_j} = \frac{\partial}{\partial g_j} \left\{ g'T'Cg - g'T'y - y'Tg + y'y + \gamma g'Hg \right\} = 0 \]  

\[ (3.39) \]

Solving Equation 3.39 and assuming \( y \sim y^d \) gives the following expression for \( g \).

\[ g = (T'T + \gamma H)^{-1}T'y^d \]  

\[ (3.40) \]

Equation 3.40 is the basic equation for the Phillips-Twomey method or constrained linear inversion. Clearly the value of \( \gamma \) will play a significant role in the quality of the inversion. Twomey (1977) recommends that solutions be obtained for several values of \( \gamma \), and then the residual error, \( ||T^*g - y^d|| \), be calculated for each of the solutions. The \( g \) that gives residual error closest to the rms of the expected experimental errors is taken to be the retrieved solution. Since \( (T^*T + \gamma H) \) must be inverted for each
different value of \( \gamma \), this method can also require a large number of calculations if \( \gamma \) is not well chosen.

Eigenfunction Method

The eigenfunction method has been applied to particle sizing by several authors and is thoroughly discussed in the literature [Capps et al., 1982; Bertero and Pike, 1983; Viera and Box, 1985; Ben-David et al., 1988; Curry, 1989; Box et al., 1992]. The eigenfunction method makes use of Schmidt-Hilbert theory for linear integral operators [Mathews and Walker, 1964; Arfken, 1966] to solve inverse problems. Since Schmidt-Hilbert theory is the singular value decomposition of the integral operator defined by Equation 3.3, this approach is sometimes referred to as singular value analysis [Bertero et al., 1985; Bertero et al., 1986; Arridge et al., 1989].

The kernels of the integral equation given by Equation 3.3 are used to define a function, \( M \).

\[
M(z, z') = \sum_{j=1}^{m} K_j(z)K_j(z')
\]

(3.40)

The eigenfunctions of \( M(z, z') \) are defined by

\[
\Phi_j(z') = \frac{1}{\lambda_j} \int_{z_i}^{z_f} M(z, z') \Phi_j(z) dz
\]

(3.41)

where the \( \lambda_j \) are the eigenvalues. The eigenfunctions of \( M \) are orthonormal on the interval \([z_i, z_f]\).

\[
\int_{z_i}^{z_f} \Phi_i(z) \Phi_j(z) dz = \delta_{ij}
\]

(3.42)

Using Equation 3.42, it can be shown that the eigenvalues of \( M(z, z') \) are also the eigenvalues of the kernel covariance matrix, which is defined by

\[
\tilde{M}_{ij} = \int_{z_i}^{z_f} K_i(z)K_j(z) dz
\]

(3.43)
The kennel covariance matrix is also known as the Gram matrix [Bertero et al., 1985]. Since $\tilde{M}$ is a real, symmetric matrix all the eigenvalues are real and positive, and the normalized eigenvectors, $u$, obey the following orthogonality relationships.

$$\sum_{i=1}^{m} u_i^i u_i^q = \sum_{i=1}^{m} u_i^p u_i^q = \delta_{pq} \tag{3.44}$$

In Equation 3.44, a superscript refers to a particular eigenvector, and a subscript refers to a component of an eigenvector. Equations 3.42 and 3.44 are used to express the scattering kernels as linear combinations of the eigenvalues, eigenvectors, and eigenfunctions.

$$K_i(z) = \sum_{j=1}^{m} \lambda_j^{1/2} u_i^j \Phi_j(z) \tag{3.45}$$

The inverse of Equation 3.45 can be obtained using Equations 3.42 and 3.44. This is the relationship that is used to generate the eigenfunctions in practice.

$$\Phi_j(z) = \frac{1}{\lambda_j^{1/2}} \sum_{i=1}^{m} u_i^j K_i(z) \tag{3.46}$$

Next, an approximation of the unknown function is obtained by combining the eigenfunctions. It is important to note that only the portion of the unknown function that lies in the space spanned by the kernels, $K_j(z)$, is represented by this expansion. In general, the kernels are not orthogonal and do not form a complete set. Therefore, it is necessary to supplement the eigenfunctions with supplemental basis functions in order to obtain a satisfactory representation of the unknown function. The use of the supplemental basis functions will be discussed in more detail later in this section.

$$g(z) = \sum_{j=1}^{m} a_j \Phi_j(z) \tag{3.47}$$

The expansion coefficients needed in Equation 3.47 are calculated by substituting 3.45 and 3.47 into Equation 3.3.
\[
a_j = \frac{1}{\lambda_j^2} \sum_{i=1}^{m} y_i^j u_i^j
\]  

(3.48)

Due to errors in the measurements and to the fact that the eigenfunctions do not form a complete set, the solution obtained from Equation 3.47 generally exhibits unphysical behavior such as high frequency oscillations and negative values. Therefore, it is necessary to impose \textit{a priori} constraints to obtain a physically realistic solution. These \textit{a priori} techniques are also known as smoothing or regularization methods [Bertero et al., 1988]. Although there are many possible constraints (smoothness or positivity, for example), a trial function constraint was particularly successfully in a previous study [Curry, 1989]. The use of the trial function constraint also allows the Lagrange multiplier or weighting parameter for the trial function to be obtained in a simple and direct manner [Curry, 1989].

An initial guess for the unknown function is used as a trial function, \( g_l \). In order to impose the trial function constraint, the trial function must be expressed as a linear combination of a set of orthonormal basis functions that span the interval \([z_i,z_f]\). Since the eigenfunctions do not form a complete set, a set of supplemental basis functions must be introduced. The supplemental basis functions are obtained by applying the Gram-Schmidt orthogonalization procedure to a set of supplemental functions, \( \phi_i(z) \). Any set of orthogonal functions (trigonometric functions, Legendre polynomials, etc.) may be used as supplemental functions. First the components of the supplemental functions that are not orthogonal to the eigenfunctions are eliminated.

\[
\tilde{\phi}_j(z) = \phi_j(z) - \sum_{i=1}^{j-1} \phi_i(z) \int_{z_i}^{z_j} \phi_i(z') \phi_j(z') dz'
\]  

for \( j = 1,\ldots,p' \)  

(3.49)

Then, the supplemental basis functions are calculated by normalizing the \( \tilde{\phi}_j(z) \) on the interval \([z_i,z_f]\).

\[
\Phi_j(z) = \frac{\tilde{\phi}_{j-m}(z)}{\left\{ \int_{z_i}^{z_f} \tilde{\phi}_{j-m}^2(z') dz' \right\}^{1/2}}
\]  

for \( j = m,\ldots,m+p \)  

(3.50)

Note that since some of the supplemental functions will be eliminated in the orthogonalization process, the number of supplemental basis functions, \( p \), will be less
than the number of supplemental functions, \( p' \). The trial function can now be accurately written as an expansion of the basis functions.

\[
g'(z) = \sum_{j=1}^{m+p} a_j \Phi_j(z) \quad (3.51)
\]

The trial function expansion coefficients are calculated from

\[
a_j = \int_{x_1}^{x_f} g'(z) \Phi_j(z) \, dz \quad (3.52)
\]

The trial function constraint can now be imposed. It is assumed that the unknown function can also be expressed as a linear combination of the eigenfunctions and the supplemental basis functions.

\[
g(z) = \sum_{j=1}^{m+p} a_j^c \Psi_j(z) \quad (3.53)
\]

The constrained expansion coefficients, \( a_j^c \), are obtained by minimizing a performance function. The performance function is defined in Equation 3.54.

\[
Q = \sum_{j=1}^{m} \left\{ \int_{x_1}^{x_f} g(z) K_j(z) \, dz - y_{j_1} \right\}^2 + \gamma \int_{x_1}^{x_f} \left\{ g'(z) - g(z) \right\}^2 \, dz \quad (3.54)
\]

Equations 3.45, 3.47, 3.51 and 3.53 are substituted into Equation 3.54, and the performance function is rewritten as

\[
Q = \sum_{j=1}^{m} a_j^c \lambda_j - 2 \sum_{j=1}^{m} a_j^c a_j \lambda_j + \sum_{j=1}^{m} y_{j_1}^2 + \gamma \sum_{j=1}^{m} (a_j^c - a_j^c)^2 \quad (3.55)
\]

Using Equation 3.55, it can be shown that \( Q \) is a minimum when

\[
a_j^c = \frac{a_j \lambda_j + \gamma a_j}{\lambda_j + \gamma} \quad (3.56)
\]
A convenient measure of the error in the solution is given by the square norm of the difference between the actual unknown function and the approximation of the unknown function that is obtained from Equation 3.53.

\[
\delta g^2 = \int \left( g(z) - \sum_{j=1}^{m+p} a_j^* \Phi_j(z) \right)^2 \, dz \tag{3.57}
\]

The optimal value of gamma is obtained by minimizing the \( \delta g^2 \). If supplemental basis functions are not used, Curry (1989) has shown that \( \delta g^2 \) can be approximated by a parameter known as the residual relative variance or RRV.

\[
RRV = \sum_{j=1}^{m} \frac{1}{\left( \lambda_j + \gamma \right)^2} \left[ \lambda_j \Delta y_j^2 + \gamma^2 (a_j^* - a_j)^2 \right] \tag{3.58}
\]

When supplemental basis functions are used, approximating \( \delta g^2 \) with the RRV is no longer strictly valid. However, the dependence of the RRV on \( \gamma \) is the same as the dependence of on \( \gamma \), so setting \( \frac{\partial RRV}{\partial \gamma} \) equal to zero and solving for \( \gamma \) will give the value that minimizes \( \delta g^2 \).

Although the mathematics of the eigenfunction method is more complex than other analytical techniques, it has several advantages. Primarily, the eigenfunction technique allows for simple and direct method of determining the weight assigned to the trial function. In Phillips-Twomey method, the Lagrange multiplier is determined by trial and error where each iteration requires a matrix inversion. The use of a trial function constraint is also advantageous in that it incorporates two commonly used constraints: smoothness and positivity. Since the trial function is smooth and positive everywhere, constraining the retrieved solution towards the trial function accomplishes both of these objectives with a single constraint.

**Empirical Solutions to Inverse Problems**

Empirical inversion techniques generally require that a parametric model of the physical process be developed. The parameters are then adjusted within physically realistic bounds to obtain a least squares fit of the measured data. In general these techniques are tedious, computationally intensive and do not provide information regarding other possible solutions. Also, the extension of these techniques to cases where there are multiple unknowns presents considerable problems. However, an empirical
inversion technique that overcomes several of these difficulties has been developed by Quist and Wyatt (1985). This technique is known as the optical strip-map technique, and a brief description of this method is given in the next section.

Optical Strip-Map Technique

This technique is attractive in that it does not involve repeated calculations and requires much less data than other analytical or empirical inversion techniques. In this technique, optical observables are calculated for particles of different sizes and optical properties. An optical observable is defined as any dimensionless quantity such as the number of peaks in the scattering pattern or the ratio of the intensities scattered in two different directions. The range of values assumed by an optical observable is divided into intervals, and strip-map catalogs are created by assigning a value of 1 to each combination of particle size and optical properties that results in an optical observable that lies within the specified interval. A value of 0 is assigned to all the combinations of particle sizes and refractive indices that lie outside the specified interval. Similar catalogs are created for each optical observable. The particle size and refractive index is then determined by retrieving the maps from the catalogs that correspond to the measured optical observables, and overlaying the various maps. The region in the \( x-n \) plane that is contained within the strip maps for each optical observable specifies the size and refractive index of the particle.

The optical strip-map technique has been successfully used to retrieve the size and real refractive index from single particle scattering light scattering measurements [Quist and Wyatt, 1985; Bottiger, 1991]. However, this technique does not provide any information regarding the absorption index, and it cannot be extended to measurements of the light scattered by polydispersions.

Summary

The mathematical formulation of inverse problems and several solution methods have been discussed. The solution techniques discussed in this chapter are classified as either analytical or empirical methods. Analytical techniques involve formal solutions of Equation 3.1, while empirical techniques find solutions using direct searches. The analytical inversion schemes that are commonly used in particle sizing applications have been discussed, along with one empirical technique.

Four analytical inversion methods were discussed. The Backus-Gilbert technique is advantageous in that no a priori information is required, and information regarding the resolution of the retrieved values is obtained. However, this technique is computationally
intensive and is more sensitive to errors in the measurements than other inversion schemes. Therefore, this method is only recommended when there is no way to obtain \textit{a priori} information and the measurement errors are expected to be small. This method could also be used to obtain preliminary solutions that will help in selecting constraints for other analytical inversion techniques. The statistical method provides a simple and direct way of obtaining a solution to inverse problems. However, statistical methods require information regarding the statistical properties of the unknown function and the measurement errors which may not be readily available. The Phillips-Twomey method or constrained linear inversion is a fairly simple technique to apply. The most significant difficulty with this method is determining the proper weight to assign to the constraint. This inversion process can become computationally intensive if the weighting parameter for the constraint is not well chosen. Due to the complex mathematics involved in the eigenfunction method, this method is more difficult to apply than other analytical techniques. The primary advantage of the eigenfunction technique is that the proper weight for the constraint can be determined in a relative simple and direct manner.

The one empirical inversion technique discussed in this chapter, the optical strip-map technique, overcomes many of the difficulties that plague empirical techniques. However, this technique does not provide any information regarding the absorption index, and it cannot be extended to measurements of the light scattered by polydispersions.

The inversion technique used in this study makes use of both analytical and empirical inversion methods. A simplified version of the optical strip-map technique is used to select a trial function. The eigenfunction method is then used to retrieve the real part of the refractive index and the PSDF. An estimated of the absorption index is obtained by comparing the measured scattering pattern with scattering patterns that are calculated using the retrieved PSDF and refractive index. Chapter 4 gives a complete description of this inversion process.
Chapter 4
Inversion Procedure

In Chapter 2 the integral equation that represents measurements of the light scattered by spherical particles was discussed. In Chapter 3 it was shown that the scattering equation is categorized as a linear inverse problem with discrete data, and methods of solving this broad class of problems were discussed. This chapter will describe how a combination of the methods described in Chapter 3 can be used to retrieve the PSDF and optical properties of aerosol particles from light scattering measurements. The results from the inversion of an example data set are presented to illustrate the actual mechanics of the inversion process. The synthetic data set used in the example inversion is representative of measurements obtainable with the 36 channel nephelometer which will be described in Chapter 5. Application of the inversion technique to single particles and to polydispersions is examined using synthetic data sets.

In the development of this inversion process, attention was focused on simulating experiments in which multi-channel polar nephelometers are used to measure the light scattered from 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 a light source. The second nephelometer has detectors 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. As discussed in Chapter 2, 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. Equation 2.1 shows the relationship between the angular scattering cross sections and the unknown distribution of particle sizes and refractive indices. Assuming that the particles are weakly absorbing, that all the particles have the same optical properties, and that the particle number density is uniform over the scattering volume, the scattering equation simplifies to Equation 2.6, which is repeated here for convenience.

$$c_j = \frac{1}{c_{\text{avg}}} \int f(x) \delta(n - n_j) \frac{dC_{\text{avg}}}{d\Omega}(x, n, 0) dx dn + \delta c_j$$

(4.1)

The eigenfunction method is initially used without a constraint to obtain the refractive index. Although the unconstrained solution exhibits physically unrealistic
behavior, 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. A simplified version of the optical strip-map technique [Quist and Wyatt, 1985] is used to select the trial function if the PSDF corresponds to a single particle or a narrow distribution. If the PSDF is broad, selection of the technique must be based on an analysis of the experiment or other means. The constrained eigenfunction method is then used to retrieve the PSDF. The RRV is used to select the optimal weighting for the trial function constraint [Curry, 1989]. The value of the absorption index is obtained by comparing the measured scattering pattern with the scattering pattern calculated using 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 summary, there are five major steps in the inversion process: (1) preliminary analysis of the measurements, (2) selection of the inputs from the available measurement set, (3) retrieval of the refractive index using the unconstrained solution, (4) retrieval of the PSDF using the constrained solution, and (5) retrieval of the absorption index by matching the measured and calculated scattering patterns.

Preliminary Analysis of the Measurements

If the PSDF is monodisperse, angular scattering cross sections should be calculated for several different sizes and optical properties within the expected ranges before attempting to 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 selecting the trial function. If the PSDF is not monodisperse, an analysis of the experiment may provide the information needed to select the trial function. If the analysis of the experiment does not provide enough information to select the trial function, complementary use of probe sampling may be required.

The range of sizes and optical properties considered in the example inversion are listed in Table 4.1. Angular scattering cross sections were calculated for the 36 channel nephelometer at 12 different sizes and 3 sets of optical properties. Table 4.2 lists the average and 20° angular scattering cross sections. Table 4.2 can be considered to be a low resolution optical strip-map [Quist and Wyatt, 1985]. For the example problem, synthetic measurements representing the light scattered by a single spherical particle were calculated using Equation 2.1. The size and the optical properties of the particle were randomly selected from the ranges specified in Table 4.1, so the actual parameters of the distribution
function were not known until after the inversion had been completed. Gaussian distributed random noise was added to each synthetic measurement in order to simulate actual experimental conditions. The angular scattering cross sections are plotted in Figure 4.1, 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 \( \mu \text{m}^2 \). Comparison of the 20° angular scattering cross section and the average of the angular scattering cross sections with the values in Table 4.2 indicates that the particle has a diameter of approximately 4 to 5 \( \mu \text{m} \) (a size parameter between 19 and 23).

Table 4.1. Range of Sizes and Optical Properties

<table>
<thead>
<tr>
<th>Diameter Range (( \mu \text{m} ))</th>
<th>Size Parameter Range</th>
<th>Refractive Index Range</th>
<th>Absorption Index Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1 - 10.1 ( \mu \text{m} )</td>
<td>0.5 - 47.0</td>
<td>1.1 - 2.0</td>
<td>( \leq 10^{-3} )</td>
</tr>
</tbody>
</table>

Table 4.2. Average and 20° Angular Scattering Cross Sections for the 36 Channel Nephelometer

<table>
<thead>
<tr>
<th>Diameter (( \mu \text{m} ))</th>
<th>Size Parameter</th>
<th>Optical Constants</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>1.1 + i 10^{-3}</td>
</tr>
<tr>
<td></td>
<td></td>
<td>( \sigma_{\text{avg}}^{\text{sca}} ) (( \mu \text{m}^2 ))</td>
</tr>
<tr>
<td>0.1</td>
<td>0.5</td>
<td>3e-7\text{e-7}</td>
</tr>
<tr>
<td>0.5</td>
<td>2.3</td>
<td>3e-4\text{e-3}</td>
</tr>
<tr>
<td>1.0</td>
<td>4.7</td>
<td>4e-3\text{e-1}</td>
</tr>
<tr>
<td>2.0</td>
<td>9.4</td>
<td>1e-2\text{e-0}</td>
</tr>
<tr>
<td>3.0</td>
<td>14</td>
<td>3e-2\text{e-0}</td>
</tr>
<tr>
<td>4.0</td>
<td>19</td>
<td>0.1\text{e-0}</td>
</tr>
<tr>
<td>5.0</td>
<td>23</td>
<td>0.1\text{e-0}</td>
</tr>
<tr>
<td>6.0</td>
<td>28</td>
<td>0.2\text{e-0}</td>
</tr>
<tr>
<td>7.0</td>
<td>33</td>
<td>0.3\text{e-0}</td>
</tr>
<tr>
<td>8.0</td>
<td>38</td>
<td>0.3\text{e-0}</td>
</tr>
<tr>
<td>9.0</td>
<td>42</td>
<td>0.6\text{e-0}</td>
</tr>
<tr>
<td>10.0</td>
<td>47</td>
<td>0.6\text{e-0}</td>
</tr>
</tbody>
</table>
Angular Scattering
Cross Sections (μm²)

Figure 4.1 Synthetic Angular Scattering Cross Sections
Input Selection

The scattering kernels are not mutually orthogonal functions, so a large number of measurements may contain relatively few independent measurements [Twomey 1974, 1977]. Therefore, it is necessary to select an optimized set of inputs from the available set of measurements. An algorithm similar to that used by Capps et al. (1982) is used to determine the input set. The kernel covariance is calculated using the scattering kernels corresponding to the complete set of measurements, and the eigenvalues and eigenvectors of the kernel covariance matrix are calculated. As in Equation 3.43, the elements of the kernel covariance matrix are defined by

\[
\tilde{M}_{ij} = \int \frac{dC^{rvg}_{i}}{d\Omega} (x,n,0) \frac{dC^{rvg}_{j}}{d\Omega} (x,n,0) dx dn
\]  

(4.2)

The eigenvalues of \( \tilde{M} \) are calculated, and an expression derived by Twomey (1974) for the relative error is used to determine whether or not the selected inputs suitable for use in the inversion process. The relative error is defined by Equation 4.3 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 4.3 is dominated by the condition number, which is the ratio of the largest eigenvalue to the smallest eigenvalue [Turchin et al., 1971; Twomey, 1974; Bertero et al., 1985b; Ben-David et al., 1988].

\[
\Delta f = \frac{1}{m} \left( \sum_{j=1}^{m} \lambda_j \sum_{j=1}^{m} \Delta C_j^2 \right)^{1/2} \sum_{j=1}^{m} C_j^2
\]  

(4.3)

If the relative error given by Equation 4.3 is too large, the largest off diagonal element of the kernel covariance matrix is used 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 Equation 4.3 is small enough. In this study, the best results were obtained when the relative error is slightly less than 1. For the angular scattering cross sections shown in Figure 4.1, 27 to 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 discussed in the next section.

**Retrieval of the Refractive Index**

Schmidt-Hilbert theory [Mathews and Walker, 1964; Arfken, 1966] is used to
obtain a set of orthonormal functions which are known as Schmidt-Hilbert eigenfunctions.
The unconstrained solution is obtained by expanding the distribution function as a linear
combination of the Schmidt-Hilbert eigenfunctions.

\[ f(x)\delta(n - n_s) = \sum_{j=1}^{m} a_j \Phi_j(x, n) \]  

(4.4)

The unconstrained expansion coefficients are calculated from

\[ a_j = \frac{\sum_{j=1}^{m} c_j u_j^i}{\sqrt{\lambda_j}} \]  

(4.5)

where \( u_j^i \) is the \( i \)th term of the \( j \)th eigenvector of the kernel covariance matrix and \( \lambda_j \) is the
eigenvalue which corresponds to the \( j \)th eigenvector. The Schmidt-Hilbert eigenfunctions
are obtained by applying the Gram-Schmidt orthogonalization procedure to the differential
scattering cross sections.

\[ \Phi_j(x, n) = \frac{1}{\sqrt{\lambda_j}} \sum_{i=1}^{m} u_j^i \frac{dC_{\text{wgs}}}{d\Omega}(x, n, 0) \]  

for \( 1 \leq j \leq m \)  

(4.6)

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

\[ W(n_s) = \frac{\int \int W(n) \sum_{j=1}^{m} a_j \Phi_j(x, n) dx dn}{\int \int \sum_{n, j=1}^{m} a_j \Phi_j(x, n) dx dn} \]

\[ n_s = W^{-1}(W(n_s)) \]  

(4.7)
f(x) = \sum_{j=1}^{m} a_j \Phi_j(x,n) dn  

\text{(4.8)}

The weighting function, \( W(n) \), is used 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 [van de Hulst, 1981].

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, the entire range of refractive indices (1.1 - 2.0) was first considered, 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 the retrieved value of the real refractive index converged to 1.42. The unconstrained PSDF is shown in Figure 4.2.

\textbf{Retrieval of the PSDF}

The unconstrained solution shown in Figure 4.2 satisfies Equation 4.1 for the set of inputs, and therefore, is a mathematically correct solution. However, the unconstrained PSDF displays characteristics such as high frequency oscillations and negative values which make it physically unrealistic. As discussed in Chapter 3, these unrealistic characteristics are due to ill-posed nature of the problem and are eliminated through the use of a priori constraints. A trial function constraint proved to be successful in a similar study [Curry, 1989], so a trial function constraint was chosen for use in the present 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. If the PSDF is broad or contains more than one mode, choosing the trial function well is essential in successfully retrieving the PSDF. However, 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 provide enough information to successfully chose a trial function.
In the example inversion, it is known that the measurements are of light scattered by a single particle. Based on this fact, the form of the trial function is chosen to be

\[ f^t(x, n) = \delta(x - x_t)\delta(n - n_s) \]  

(4.9)

The preliminary analysis of the measurements indicated that \( x_t \) should be in the range of 19 to 23. The initial value of \( x_t \) is obtained by examining the unconstrained PSDF shown in Figure 4.2. The most prominent peak of the unconstrained PSDF in or near the expected size ranges occurs at \( x = 25.1 \). Therefore, \( x_t \) is chosen to be 25.1.

As discussed in Chapter 3, a set of basis functions are necessary to supplement the Schmidt-Hilbert eigenfunctions and impose the trial function constraint. The additional basis functions are obtained by orthogonalizing a set of orthonormal functions with respect to the Schmidt-Hilbert eigenfunctions. The supplemental orthonormal functions used in the example inversion are

\[ \phi_j(x, n) = \delta(x - x_j)\delta(n - n_s) \text{ for } 1 \leq j \leq m \]  

(4.10)

One hundred and one supplemental orthonormal basis functions were used, and the \( x_j \) were evenly spaced throughout the range of size parameters. The supplemental orthonormal functions are orthogonalized with respect to the Schmidt-Hilbert eigenfunctions, and the \( \phi_j(x, n) \) that lie entirely in the space spanned by the eigenfunctions are eliminated by the orthogonalization procedure. The number of supplemental basis functions used in the example inversion was \( p = 93 \).

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

\[ f^t(x, n) = \sum_{j=1}^{m+p} a_j^t \phi_j(x, n) \]  

(4.11)

\[ f(x)\delta(n - n_s) = \sum_{j=1}^{m+p} a_j^x \phi_j(x, n) \]  

(4.12)

where

\[ a_j^t = \int_{n_s}^{n_s} \int_{x_s}^{x_s} f^t(x, n) \phi_j(x, n) dx dn \]  

(4.13)
As before, a performance function is defined, and the constrained expansion coefficients are found by minimizing the performance function.

\[
Q = \sum_{j=1}^{m} \left[ \frac{1}{c_j} \int \left( \sum_{i=1}^{m} a_{j}^c \Phi_i(x, n) \frac{dC_{ij}}{d\Omega} (x, n_s, 0) dx dn - c_j \right)^2 \right] 
+ \gamma \int \left( \sum_{i=1}^{m} a_{j}^c \Phi_i(x, n) - \sum_{i=1}^{m} a_{j}^t \Phi_i(x, n) \right) dx dn
\tag{4.14}
\]

where

\[
a_j^c = \frac{a_j \lambda_j + \gamma a_j^t}{\lambda_j + \gamma} \quad \text{for } 1 \leq j \leq m
\]

\[
a_j^t = a_j^t \quad \text{for } m < j \leq m + p
\tag{4.15}
\]

The weighting parameter for the trial function constraint is determined from the value of the partial derivative of the RRV with respect to \( \gamma \) as discussed in Chapter 3.

\[
\frac{\partial \text{RRV}}{\partial \gamma} = \sum_{j=1}^{m} \frac{\lambda_j \left\{ \gamma (a_j - a_j^t)^2 - \frac{c_{\text{sca}}^2}{C_{\text{sca}}^2} \right\}}{(\lambda_j + \gamma)^3}
\tag{4.16}
\]

In the example inversion, \( \frac{\partial \text{RRV}}{\partial \gamma} \) was -5.0E-08 when \( \gamma \) equaled 86.3.

After the optimal value of \( \gamma \) is determined, the constrained solution can be calculated from Equation 4.16. The PSDF is then obtained by integrating the constrained solution over the range of refractive indices. The constrained PSDF for the example inversion is plotted in Figure 4.3.

\[
f(x) = \int_{n_i}^{n_{i+p}} \sum_{j=1}^{m} a_j^c \Phi_j(x, n) dn
\tag{4.17}
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 using Equation 2.1. Calculation of the angular scattering cross sections from Equation 2.1 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 using a different trial function.

In the example inversion, the absorption index that gave the rms residual error closest to the rms of the estimated experimental errors, \( \Delta C_j \), was \( 10^{-3} \). The measured and calculated scattering patterns are compared in Figure 4.4. The relatively large discrepancy between the calculated and measured 20° angular scattering cross section indicated that the size parameter selected for the trial function is too large. The relatively good agreement between the calculated and measured scattering cross sections in the 100° to 150° range indicates that the retrieved value of the real refractive index is close to the actual value. Figure 4.2 is again used to select a \( x_t \) 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, the closest agreement between the measured and calculated scattering patterns is obtained 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 Figure 4.5.

Summary of the Example Inversion

The retrieved and actual size and optical properties of the particle in the example inversion are compared in Table 4.3. These results are representative of a number of inversions performed using synthetic data sets. Similar results are expected from inversions of data collected with the 36 channel polar nephelometer.
Figure 4.4. Comparison of the Synthetic Measurements and the Calculated Scattering Pattern with $x_t = 25.1$
Figure 4.5. Comparison of the Synthetic Measurements and the Calculated Scattering Pattern with $x_t = 20.1$
Table 4.3. Typical Results from the Inversion of a 36 Channel Synthetic Data Set

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Actual Value</th>
<th>Retrieved Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diameter (µm)</td>
<td>4.5</td>
<td>4.3</td>
</tr>
<tr>
<td>Size Parameter</td>
<td>21.2</td>
<td>20.1</td>
</tr>
<tr>
<td>Optical Properties</td>
<td>$1.45 + i 7.5E-5$</td>
<td>$1.42 + i 1.0E-4$</td>
</tr>
</tbody>
</table>
The actual particle size distribution functions used to generate the synthetic data in the blind test are plotted in Figure 4.6. Although the PSDF for case 4 has the same geometric standard deviation as the distributions in cases 1 and 2, the PSDF for case 4 is actually broader than in 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). This is due to the fact that as the distributions become broader, the scattering pattern becomes smoother, and probability of finding another PSDF that will produce a similar scattering pattern increases. These results also show the need to obtain reliable a priori information regarding the PSDF in order to successfully invert light scattering measurements. In this study, it was assumed that the height of each distribution is greater than its width, and the trial functions used to constrain the solution were selected accordingly. In cases 3 - 5, assumption of a narrow PSDF was not valid, and the retrieved PSDF did not resemble the actual PSDF. Finally, it is interesting to note that even when the retrieved distributions differed from the actual distributions, the refractive index was retrieved accurately.

Summary

An inversion technique that retrieves the particle size distribution function and the refractive index of weakly absorbing spherical particles from synthetic measurements of scattered light has been developed. The solution is obtained by expanding the distribution function as a linear combination of orthonormal basis functions. The orthogonality properties of the basis functions are used to find the expansion coefficients which minimize the residual errors subject to a trial function constraint. Fortran programs were written to implement the inversion technique. A program users guide and a listings of the programs of the programs are provided in Appendix C. The technique is shown to be capable of retrieving the size and optical properties from measurements of the light scattered by a weakly absorbing sphere. The technique was also used to retrieve the PSDF and refractive index from synthetic measurements of the light scattered by narrow log normal distributions of non-absorbing 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. The results of this study show that because there is not a unique solution to the inverse light scattering problem, accurate a priori information regarding the unknown PSDF must be available. If such information cannot be obtained from analysis of the particular environment in which the light scattering measurements are made, the complementary use of probe sampling techniques is recommended. This conclusion is in agreement with Koo (1987), who after reviewing particle sizing techniques
Figure 4.6. Particle Size Distribution Functions
used to analyze the metallic oxide smoke produced by the combustion of solid rocket propellants recommended the complementary use of laser/optical and probe sampling techniques. Similar conclusions were reached by Bottiger (1985) after comparing five different inversion techniques. Further research is needed to investigate the possibility that the trial function could be selected without the use of a priori information. One possibility is to use an iterative procedure that begins with a non-prejudical trial function. Another possibility is to use the Backus-Gilbert inversion method to obtain a preliminary solution that could serve as a trial function. Further modifications of the inversion process will focus in this area.
Chapter 5
Nephelometer Calibration

Multi-channel nephelometers capable of measuring the light scattered by aerosol particles at 36 polar angles evenly spaced every 4° from 20° to 160° have been designed and are currently being constructed. A prototype nephelometer with 15 detectors located between 23° and 128° is currently in use in the Laser Applications Laboratory at Argonne National Laboratory. Figure 5.1 shows the primary components of the 36 channel nephelometers.

![Figure 5.1. Schematic Diagram for a Multi-Channel Polar Nephelometer](image)

Aerosol particles are entrained in the air stream that passes through the center of the scattering chamber and intersects the laser beam. The laser light scattered within the acceptance cone of each detector is collected and transmitted along fiber optics to a CCD detector array. The prototype nephelometer uses photodiodes rather than a CCD array. The output from the CCD array or the photodiodes is digitized and stored on a personal computer.

Angular light scattering measurements are classified as either relative measurements or absolute measurements. A relative measurement is normalized by another measurement or set of measurements. The normalization eliminates the need to characterize the optical system in detail or to have calibrated detectors. However, since the measurements are made simultaneously, the relative sensitivity of each detector must still be known. A simple technique to determine relative sensitivity of the detectors has been developed and is described in Appendix D. In order to make absolute scattering measurements, the performance of the light source and all the optical components and detectors must be well...
characterized. One method of calibrating a nephelometer or other light scattering instrument is to measure the light scattered by spheres of known size, refractive index, and concentration. Although aerosol streams containing spherical particles of known size and refractive index can be generated using commercially available polystyrene spheres, it is difficult to determine the particle number concentration accurately enough to use in a calibration procedure. However, since the molecular concentration of a gas can easily be calculated at a given temperature and pressure, the use of a gas with a relatively high refractive index for a scattering medium is a promising alternative. A method that uses dichlorodifluoromethane (CCl₂F₂), commonly known as Freon-12, as the scattering medium is described in this chapter. All of the results described in this chapter were obtained using the 15 channel nephelometer, but the techniques are equally applicable to the 36 channel nephelometers. Examples of the type of measurements obtainable with the nephelometers are also presented.

Calibration for Absolute Scattering Measurements

Calibration for absolute scattering measurements requires theoretical values of the ratio of the power received by each detector to the incident irradiance be known. In Chapter 2, the ratio of the power received by a detector to the incident irradiance was defined as an angular scattering cross section. Assuming single scattering, the relationship between the angular scattering cross section measured by the jth detector, C_j, and the distribution of particle sizes and optical properties is given by Equation 2.1, which is repeated here for convenience.

\[
C_j = \int \int \int \int N_c \tilde{f}(x,n,k) \frac{d\tilde{C}_j}{d\Omega}(\Omega,x,n,k) dx dv d\Omega dV \quad (5.1)
\]

When the following conditions are satisfied, Equation 5.1 can be greatly simplified.

1. The particle number concentration is uniform over the scattering volume.
2. The distribution of particle sizes and optical properties can be replaced by a monodispersion with an average size and optical properties.

\[
\tilde{f}(x,n,k) = \delta(x-x_{\text{avg}})\delta(n-n_{\text{avg}})\delta(k-k_{\text{avg}}) \quad (5.2)
\]

3. The differential scattering cross sections can be replaced by their average over the solid angle subtended by each detector.
The third condition needs clarification. The integral shown in Equation 5.3 is calculated using the trapezoidal rule, so in practice the integral is replaced by a sum. Therefore, Equation 5.3 is implying that the average of a product is equal to the product of the averages. This is only absolutely true if one of the terms in the product is a constant. Therefore, Equation 5.3 will only be valid if the solid angles subtended by the detectors are sufficiently small that the differential scattering cross sections are relatively constant over the subtended solid angles. This approximation will be valid near 90° where the slope of the scattering pattern is small, but may be suspect in the forward scattering angles where the scattering pattern varies rapidly with angle. In order to investigate the validity of assuming that the differential scattering cross sections were constant over the subtended solid angles, the following functional form was assumed for the differential scattering cross sections.

\[
\frac{d\tilde{C}_j}{d\Omega}(\Omega, x_{avg}, n_{avg}, k_{avg}) = \theta^{-m} \quad \text{where } m = 1, 2, 3 \tag{5.4}
\]

Assuming a monodispersion and particle number density of 1 cm\(^{-3}\), Equation 5.4 can be substituted into Equation 5.1, and the integrals can be calculated analytically. The exact results are compared with the numerical results in Table 5.1. These results indicate that the third assumption is accurate to within ~1% as long as the scattering pattern does not vary with \(\theta\) more rapidly than \(\theta^{-3}\). The detectors on the 36 channel nephelometers are smaller than those on the 15 channel nephelometer, so the third assumption is also valid for the 36 channel nephelometers.

These assumptions eliminate the integrals, and Equation 5.1 simplifies to

\[
C_j = N_c V_j \frac{d\tilde{C}_{avg}}{d\Omega} \Delta\Omega_j \tag{5.5}
\]

Equation 5.5 is the working equation for the nephelometer. Theoretical values of the parameters on the right hand side of Equation 5.5 can be determined if the characteristics of the aerosol stream (concentration, particle size and optical properties) and the geometry of the nephelometer are known. The theoretical values of \(C_j\) are directly
proportional to the ratio of the measured scattered power to the incident laser power. This
relationship is used to define the scattering correction factors.

\[
C_j = SCF_j M_j
\]  

(5.6)

Therefore, measurements of the light scattered by a well characterized aerosol stream can be
used to calculate absolute scattering correction factors for each detector. The following
sections describe how the parameters needed to determine the scattering correction factors
are measured or calculated.

Table 5.1. Validation of the Third Assumption

<table>
<thead>
<tr>
<th>( \theta_j )</th>
<th>( C_j ) for ( m = 1 )</th>
<th>( C_j ) for ( m = 2 )</th>
<th>( C_j ) for ( m = 3 )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Numerical</td>
<td>Exact</td>
<td>% Error</td>
</tr>
<tr>
<td>23.07</td>
<td>9.40e-05</td>
<td>9.39e-05</td>
<td>0.12</td>
</tr>
<tr>
<td>28.96</td>
<td>7.62e-05</td>
<td>7.61e-05</td>
<td>0.14</td>
</tr>
<tr>
<td>34.92</td>
<td>9.71e-05</td>
<td>9.70e-05</td>
<td>0.11</td>
</tr>
<tr>
<td>40.97</td>
<td>8.29e-05</td>
<td>8.28e-05</td>
<td>0.10</td>
</tr>
<tr>
<td>47.16</td>
<td>7.21e-05</td>
<td>7.20e-05</td>
<td>0.11</td>
</tr>
<tr>
<td>53.49</td>
<td>6.37e-05</td>
<td>6.37e-05</td>
<td>0.09</td>
</tr>
<tr>
<td>60.00</td>
<td>5.69e-05</td>
<td>5.69e-05</td>
<td>0.07</td>
</tr>
<tr>
<td>66.73</td>
<td>5.12e-05</td>
<td>5.12e-05</td>
<td>0.04</td>
</tr>
<tr>
<td>73.74</td>
<td>4.64e-05</td>
<td>4.64e-05</td>
<td>0.06</td>
</tr>
<tr>
<td>81.08</td>
<td>4.23e-05</td>
<td>4.22e-05</td>
<td>0.03</td>
</tr>
<tr>
<td>88.85</td>
<td>3.85e-05</td>
<td>3.85e-05</td>
<td>0.05</td>
</tr>
<tr>
<td>97.18</td>
<td>3.52e-05</td>
<td>3.52e-05</td>
<td>0.04</td>
</tr>
<tr>
<td>106.30</td>
<td>3.22e-05</td>
<td>3.22e-05</td>
<td>0.08</td>
</tr>
<tr>
<td>116.40</td>
<td>2.93e-05</td>
<td>2.93e-05</td>
<td>0.01</td>
</tr>
<tr>
<td>128.30</td>
<td>2.65e-05</td>
<td>2.65e-05</td>
<td>0.03</td>
</tr>
</tbody>
</table>

Measurement of the Light Scattered by Freon-12

A stream of scattering particles of known size and optical properties is most easily
produced using a pure gas. Completely closing the sheath flow bypass valve on the
nephelometer pump unit forces air back through the nephelometer and out the inlet
tube. Once the nephelometer is thoroughly purged, the stray light received by each detector
is measured at several laser power levels. Freon-12 from an air conditioner recharging kit
is then used to fill the nephelometer as shown in Figure 5.2. The Freon-12 was sent
through a long coil of tubing before being introduced into the nephelometer to allow the gas
to reach room temperature, and the temperature of the gas was measured with a
thermocouple just before it entered the nephelometer. The gas was also sent through a 0.2
\( \mu \)m filter to remove any impurities. The flow of the Freon-12 was regulated such that a
slight amount of gas was spilling out of the nephelometer's inlet tube. Recently, legislation
has been enacted that makes it illegal to vent chlorofluorocarbons, so future use of this
technique will require that the Freon-12 be recaptured. The light scattered by the Freon-12 was then measured at the same laser power levels as the stray light measurements. Varying the laser power levels shows that the detectors respond linearly to variations in the scattered light.

![Figure 5.2. Schematic Diagram of Experimental Set-up for the Calibration Measurements](image)

The raw data was corrected by subtracting out the stray light, dividing by the incident laser power which was measured by the beam dump detector and averaging over the different laser power settings.

\[
M_j^c = \frac{1}{p} \sum_{i=1}^{p} \frac{M_{ji} - SL_{ji}}{BD_i}
\]  

(5.7)

A more accurate, but slightly more involved method of reducing the raw data is to calculate the linear least squares fit of \((M_{ji} - SL_{ji})\) vs. BD for each of the 15 channels. The slope of the linear fit then equals the corrected measurement, \(M_j^c\).

![Figure 5.3. Least Squares Data Reduction Technique](image)
Particle Number Concentration

The particle number concentration is obtained from the ideal gas law.

\[ N_c = \frac{PN_A}{RT} \]  \hspace{1cm} (5.8)

Scattering Volume

When the nephelometer is filled with a gas such as Freon-12, the scattering volume is different for each detector and is defined by the intersection of the laser beam with acceptance cone. The shaded areas in Figures 5.4a - 5.4c illustrate the scattering volume from several different perspectives.

![Figure 5.4a. Calibration Scattering Volume in the x - y Plane](image)

![Figure 5.4b. Calibration Scattering Volume in the y^b - z^b Plane](image)
As shown in Figures 5.4a - 5.4c, there are two relevant sets of coordinates. One coordinate system is aligned with the laser beam and is referred to as the base coordinate system. The other coordinate system is aligned with the detector and is referred to as the detector coordinate system. Simple rotation matrices are used to convert from detector coordinates to base coordinates and visa versa.

\[
\begin{pmatrix}
    x^b \\
    y^b
\end{pmatrix} =
\begin{pmatrix}
    \sin \theta_j & \cos \theta_j \\
    -\cos \theta_j & \sin \theta_j
\end{pmatrix}
\begin{pmatrix}
    x^d \\
    y^d
\end{pmatrix}
\]  

(5.9)

\[
\begin{pmatrix}
    x^d \\
    y^d
\end{pmatrix} =
\begin{pmatrix}
    \sin \theta_j & -\cos \theta_j \\
    \cos \theta_j & \sin \theta_j
\end{pmatrix}
\begin{pmatrix}
    x^b \\
    y^b
\end{pmatrix}
\]  

(5.10)

The geometry of channels 1 and 2 on the 15 channel nephelometer is shown in Figure 5.5. The geometry of channels 3 to 15 is identical except the collimator diameter is slightly larger. The acceptance angle, \( \alpha \), is calculated from

\[
\alpha = 2 \tan^{-1}\left( \frac{d_d + d_{A1}}{2(R_o - R_i)} \right)
\]  

(5.11)
The scattering volume for calibration is obtained from

\[ V_j = 2 \int_{-\frac{d_j}{2} y_{\text{min}}^b}^{\frac{d_j}{2} y_{\text{max}}^b} h(x^b) dx^b \int_{-\frac{d_j}{2} y_{\text{min}}^b}^{\frac{d_j}{2} y_{\text{max}}^b} dy^b \]  

(5.12)

where

\[ y_{\text{max}}^b = \frac{x^b \{ \tan(\alpha/2) \sin^2 \theta_j - \cos \theta_j \sin \theta_j \} + R_o \tan(\alpha/2) - d_d/2}{1 + \tan(\alpha/2) \cos \theta_j \sin \theta_j - \cos^2 \theta_j} \]  

(5.13)

\[ y_{\text{min}}^b = \frac{x^b \{ - \tan(\alpha/2) \sin^2 \theta_j - \cos \theta_j \sin \theta_j \} - R_o \tan(\alpha/2) + d_d/2}{1 - \tan(\alpha/2) \cos \theta_j \sin \theta_j - \cos^2 \theta_j} \]  

(5.14)

\[ h_c(x^d, y^d) = \sqrt{\frac{d_c(x^d)^2}{4} - y^d} \]  

(5.15)

\[ h_b(x^b) = \sqrt{\frac{d_b}{4} - x^b} \]  

(5.16)

\[ h = \min\{h_c(x^d, y^d), h_b(x^b)\} \]  

(5.17)

During normal operation, the scattering volume is determined by the intersection of the aerosol jet with the laser beam, and the scattering volume is the same for each detector. The scattering volume for normal operation is illustrated in Figure 5.6. The scattering volume for normal operation is calculated using Equation 5.18.

\[ V_j = 2 \int_{-\frac{d_j}{2} y_{\text{min}}^b}^{\frac{d_j}{2} y_{\text{max}}^b} h_b(x^b) dx^b \int_{-\frac{d_j}{2} y_{\text{min}}^b}^{\frac{d_j}{2} y_{\text{max}}^b} dy^b \]  

(5.18)

where

\[ y_{\text{max}}^b = -y_{\text{min}}^b = \sqrt{\frac{d_j^2}{4} - x^b} \]  

(5.19)
For the 36 channel nephelometer, the diameter of the laser beam is less than the diameter of the aerosol jet, but Equations 5.18 and 5.19 can still be used to calculate the scattering volume if $d_b$ and $d_j$ are interchanged.

A FORTRAN subroutine was written to carry out the integration for the scattering volume using the trapezoidal rule. The number of grid points was doubled until the percent change in the calculated volume was less than 0.1% to prove grid independence. A listing of the code for this subroutine is included in Appendix E. The values of the geometric parameters for each of the 15 channels needed to calculate the scattering volumes are listed in Table 5.2.

Differential Scattering Cross Sections

Differential scattering cross sections can be calculated from Mie theory only if the scattering particles are spherical. However, when Rayleigh scattering prevails, as in the case of the Freon calibration, the scattering pattern is fairly insensitive to the particle shape and depends mainly on the particle volume. The weak dependency of the scattered light on the particle shape is accounted for with shape depolarization factors. Therefore, differential scattering cross sections for Freon-12 molecules can be calculated from Mie theory using an effective scattering diameter. Although much simpler and faster approximations to Mie theory can be used for molecular scattering, Mie theory was used, so the nephelometer's response to scattering by larger spheres could be calculated using the same code. The depolarization and the refractive index of Freon-12 have been measured at several wavelengths [Bodhaine, 1979], and these measurements can be used to determine an effective diameter and refractive index at the wavelengths of interest.
Table 5.2. Geometric Parameters for the 15 Channel Polar Nephelometer

<table>
<thead>
<tr>
<th>Channel</th>
<th>θ_i(°)</th>
<th>d_d(cm)</th>
<th>d_A1(cm)</th>
<th>α(°)</th>
<th>R_o(cm)</th>
<th>R_i(cm)</th>
<th>d_b(cm)</th>
<th>d_l(cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>23.1</td>
<td>0.235</td>
<td>0.235</td>
<td>6.30</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
<tr>
<td>2</td>
<td>29.0</td>
<td>0.235</td>
<td>0.235</td>
<td>6.30</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
<tr>
<td>3</td>
<td>34.9</td>
<td>0.282†</td>
<td>0.342</td>
<td>8.35</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
<tr>
<td>4</td>
<td>41.0</td>
<td>0.282†</td>
<td>0.342</td>
<td>8.35</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
<tr>
<td>5</td>
<td>47.2</td>
<td>0.282†</td>
<td>0.344</td>
<td>8.38</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
<tr>
<td>6</td>
<td>53.5</td>
<td>0.282†</td>
<td>0.342</td>
<td>8.35</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
<tr>
<td>7</td>
<td>60.0</td>
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<td>0.342</td>
<td>8.35</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
<tr>
<td>8</td>
<td>66.7</td>
<td>0.282†</td>
<td>0.344</td>
<td>8.38</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
<tr>
<td>9</td>
<td>73.7</td>
<td>0.282†</td>
<td>0.343</td>
<td>8.37</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
<tr>
<td>10</td>
<td>81.1</td>
<td>0.282†</td>
<td>0.342</td>
<td>8.35</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
<tr>
<td>11</td>
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<td>0.344</td>
<td>8.38</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
<tr>
<td>12</td>
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<td>0.344</td>
<td>8.38</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
<tr>
<td>13</td>
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<td>0.344</td>
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<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
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<td>1.90</td>
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<tr>
<td>15</td>
<td>128.3</td>
<td>0.282†</td>
<td>0.344</td>
<td>8.38</td>
<td>6.17</td>
<td>1.90</td>
<td>0.39</td>
<td>0.38</td>
</tr>
</tbody>
</table>

†These detectors are actually 0.25 cm squares. A circular detector with an equivalent area would have a diameter of 0.282 cm.

The qualitative features of the spectral dependence of the refractive index can be described by the classical damped oscillator or Lorentz model [Brewster, 1992; Ditchburn, 1963]. The key assumptions of the Lorentz model are that the medium consists of charged particles that are held in equilibrium by perfectly elastic or Hooke's law forces and are subject to isotropic, linear damping forces. From the Lorentz model, the fundamental equation of classical dispersion theory can be derived.

\[
\hat{n} = 1 + \sum \frac{\omega_p^2}{\omega_j^2 - \omega^2 + i\gamma_j \omega} \quad (5.20)
\]

For gases, empirical dispersion curves show that only the first term in the summation needs to be included [Ditchburn, 1963]. Further simplifications can be made if the frequencies of interest lie in regions that are remote from resonant frequencies. In these regions, k \sim 0 and \left| \omega_o^2 - \omega^2 \right| \gg \gamma_\omega, and Equation 5.20 simplifies to

\[
\frac{n^2 + 2}{n^2 - 1} = \frac{3\omega_o^2}{\omega_p^2} + 1 - \omega^2 = c_0 + \frac{c_1}{\omega^2} \quad (5.21)
\]
where \( c_0 \) and \( c_1 \) are constants. Measured values of the refractive index of Freon-12 at 0°C and 1 atm were obtained from Bodhaine (1979) and are plotted in Figure 5.7. The constants \( c_0 \) and \( c_1 \) are obtained from the least squares fit shown in the figure. Equation 5.22 can then be used to calculate the refractive index at 0°C and 1 atm for the wavelengths of interest.

\[
n_{\text{ref}} = \sqrt{\frac{c_0 + c_1 / \lambda^2 + 2}{c_0 + c_1 / \lambda^2 - 1}} \quad (5.22)
\]

Values for the refractive index at other temperatures and pressures can be obtained from the Gladstone-Dale equation [Brewster, 1992].

\[
n = 1 + (n_{\text{ref}} - 1) \frac{T_{\text{ref}} P}{T P_{\text{ref}}} \quad (5.23)
\]

Now that the refractive index is know, the effective scattering diameter needs to be determined. The following derivation shows how Rayleigh-Gans scattering theory can be used to obtain a relationship between the refractive index and the effective scattering diameter.

Consider a laser beam passing through an attenuating medium. The rate at which energy is removed from a laser beam as it propagates along a path length \( l \) is given by

\[
W_{\text{ext}} = W_i (1 - \exp(-\beta l)) \quad (5.24)
\]

Assuming that the medium is non-absorbing, the extinction is equal to the scattering, and assuming that \( \beta l \ll 1 \), the power scattered in all directions is given by

\[
W_s = W_{\text{ext}} = I_i A \beta l \quad (5.25)
\]

The ratio of the power scattered by a single particle to the irradiance of the incident laser beam defines the scattering cross section of each particle. Assuming single scattering, the ratio of the total power scattered by all the illuminated particles to the incident irradiance is given by

\[
\frac{W_s}{I_i} = N_c V C_{\text{scat}} \quad (5.26)
\]
\( \frac{n_{ref}^2 + 2}{n_{ref}^2 - 1} = 1360 - 10.7/\lambda^2 \)

Figure 5.7. Measured Refractive Indices of Freon-12 at \( T_{ref} = 0 \) C and \( P_{ref} = 1 \) atm [Bodhaine, 1979]
Combining Equations 5.26 and 5.27 gives an expression for the attenuation coefficient of the medium.

\[ \beta = \frac{N_e V C_{sca}}{A_b I} = N_c C_{sca} \quad (5.27) \]

The scattering cross section is obtained by integrating the differential scattering cross sections over all directions, so Equation 5.27 can be rewritten as

\[ \beta = N_c \int \frac{d\tilde{C}}{4\pi d\Omega}(\Omega, x, \bar{n})d\Omega \quad (5.28) \]

Since Freon-12 is non-absorbing in the spectral region of interest, \( \bar{n} = n \). Also, for scattering by a gas, \( n \) is approximately 1 and \( x\ln - 1 \) is much less than 1. Therefore, the differential scattering cross sections can be obtained from Rayleigh-Gans scattering. For monochromatic, unpolarized incident light the differential scattering cross sections are given by [van de Hulst, 1981]

\[ \frac{d\tilde{C}}{d\Omega}(\Omega, x, \bar{n}) = \frac{2\pi^2 V_F^2}{\lambda^4} (n - 1)^2 |R(\Omega)|^2 (1 + \cos^2 \theta) \quad (5.29) \]

where

\[ R(\Omega) = \frac{1}{V_F} \int \exp(i\delta) dV_F \quad (5.30) \]

and \( \delta \) is the phase difference between rays scattered from different points in the particle at an infinite distance from the particle in the \( \Omega \) direction. Since the scattering particles are small, \( \delta \sim 0 \) and \( R(\Omega) \sim 1 \). Substituting for the differential scattering cross sections in Equation 5.28 gives

\[ \beta = \frac{2\pi^2 N_s V_F^2}{\lambda^4} (n - 1)^2 \int_0^{2\pi} \int_0^\theta (1 + \cos^2 \theta) \sin \theta d\theta d\phi \quad (5.31) \]

Equation 5.31 can be solved for an effective scattering diameter of the Freon-12 molecules.
The attenuation coefficient can also be calculated for particles that are small compared to the wavelength from

\[
\beta = \frac{8\pi^3 (n^2 - 1)^2 6 + 3\rho}{3\lambda^2 N_e 6 - 7\rho}
\]  

(5.33)

where \( \rho \) is the depolarization factor [van de Hulst, 1981]. At first glance, it may seem incorrect that the particle number density appears in the denominator of Equation 5.33, since the attenuation coefficient should be proportional to the number density. However, remembering that the refractive index is also proportional to the number density, it can be seen that the attenuation coefficient given by Equation 5.33 is proportional to the number density as it should be. Substituting Equation 5.33 into Equation 5.32 gives the effective diameter of the Freon-12 molecules as a function of the refractive index and the particle number concentration. It should noted that the effective scattering diameter is calculated as a computational convenience, and it is not representative of any physical dimension of a Freon-12 molecule.

\[
d_F = \left( \frac{9(n+1)^2 6 + 3\rho}{\pi^2 N_e^2 6 - 7\rho} \right)^{\frac{1}{6}}
\]  

(5.34)

The refractive indices and effective scattering diameters were calculated at 1 atm and 25°C for the wavelengths used in the 15 and 36 channel nephelometers and are listed in Table 5.3. The depolarization factor for Freon-12 is 0.048 [Bodhaine, 1979].

<table>
<thead>
<tr>
<th>Wavelength ((\mu m))</th>
<th>Refractive Index</th>
<th>Effective Scattering Diameter ((\mu m))</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.670*</td>
<td>1.001029</td>
<td>0.00423</td>
</tr>
<tr>
<td>0.840†</td>
<td>1.001023</td>
<td>0.00423</td>
</tr>
<tr>
<td>1.064*</td>
<td>1.001018</td>
<td>0.00423</td>
</tr>
</tbody>
</table>

†15 Channel Nephelometer  * 36 Channel Nephelometers

Using the refractive index and the effective scattering diameter listed in Table 5.3, differential scattering cross sections were calculated every 0.1° from 0° to 180° using the
bhmie subroutine. These differential scattering cross sections were stored in an array and used to calculate the average differential scattering cross sections for each detector, \( \frac{d\sigma_{\text{avg}}}{d\Omega_j} \).

The details pertaining to the calculation of the average differential scattering cross sections and the average solid angle subtended by each detector are described in the next section.

Average Differential Scattering Cross Section and Subtended Solid Angle for each Detector

The average differential scattering cross section for each detector and the average solid angle subtended by each detector is calculated by determining the portion of the detector that can be seen by a particle from various locations in the scattering volume. The geometry of the problem is illustrated in Figures 5.8a and 5.8b. These figures show there are three relevant coordinate systems: base coordinates, detector coordinates, and particle coordinates. The base coordinate system is aligned with the laser beam, and the detector coordinate system is aligned with the detector. The particle coordinate system is parallel to the base coordinate system, but its origin is located at \((x_p^b, y_p^b, z_p^b)\).

![Figure 5.8a. Scattering Geometry in the x-y Plane](image)
Equations 5.9 and 5.10 give the relationship between the base and detector coordinate systems. Base coordinates can be transformed into particle coordinates using Equations 5.35 through 5.37.

\[
x^p = x^b - x^b_p \quad (5.35)
\]

\[
y^p = y^b - y^b_p \quad (5.36)
\]

\[
z^p = z^b - z^b_p \quad (5.37)
\]

When viewed from the point \((x^b_p, y^b_p, z^b_p)\), the detector is roughly oval shaped. Figure 5.9 illustrates the portion of the detector that can be seen from an arbitrary point within the scattering volume.

The key to determining the solid angle subtended by the detector when viewed from \((x^b_p, y^b_p, z^b_p)\) is determining the vectors \(r_1, r_2, r_3, \) and \(r_4\) in particle coordinates. The first
The step is to determine whether the entire detector can be seen from \((x^b_p,y^b_p,z^b_p)\). The parameter \(r^*\) determines the maximum distance from the \(x^d\) axis at which the entire detector is visible.

\[
r^* = \frac{R_p d_{A1} - R_d d_d}{2(R_o - R_i)}
\]  

(5.38)

If \(|y_p^d|\) is less than or equal to \(r^*\), \(r_1\) is determined by the point 1' shown in Figure 5.8a. In detector coordinates, point 1' is located at \((-R_o, d_d/2, 0)\) if \(y_p^d\) is greater than or equal to zero or \((-R_o, -d_d/2, 0)\) if \(y_p^d\) is less than zero. If \(|y_p^d|\) is greater than \(r^*\), \(r_1\) is determined by point 1, which has detector coordinates of \((-R_i, d_{A1}/2, 0)\) if \(y_p^d\) is greater than or equal to zero or \((-R_i, -d_{A1}/2, 0)\) if \(y_p^d\) is less than zero. Vector \(r_2\) is determined by point 2 in Figure 5.8a. In detector coordinates, point 2 is located at \((-R_o, -d_d/2, 0)\) if \(y_p^d\) is greater than or equal to zero or \((-R_o, d_d/2, 0)\) if \(y_p^d\) is less than zero. Vector \(r_3\) is determined by point 3' in Figure 5.8b if \(|z_p^d|\) is less than or equal to \(r^*\), and by point 3 if \(|z_p^d|\) is greater than \(r^*\). Point 3' has coordinates of \((-R_o, 0, d_{A1}/2)\) in detector coordinates, and point 3 has coordinates of \((-R_i, 0, d_{A1}/2)\) in detector coordinates. Vector \(r_4\) is determined by point 4 in Figure 5.8b. In detector coordinates, point 4 is located at \((-R_o, 0, -d_d/2)\) if \(z_p^d\) is greater than or equal to zero or \((-R_o, 0, d_d/2)\) if \(z_p^d\) is less than zero. Once \(r_1, r_2, r_3,\) and \(r_4\) are transformed into particle coordinates, \(\Delta \theta\) and \(\Delta \phi\) can be calculated.

\[
\Delta \theta = \cos^{-1} \left( \frac{r_1 \cdot r_2}{|r_1| |r_2|} \right) \cos \left( \frac{\pi |y_p^d|}{2 y_{\text{max}}} \right)
\]  

(5.39)

\[
\Delta \phi = \cos^{-1} \left( \frac{r_1 \cdot r_2}{|r_1| |r_2|} \right) \cos \left( \frac{\pi |z_p^d|}{2 y_{\text{max}}} \right)
\]  

(5.40)

The \(\Delta \theta\) and \(\Delta \phi\) are used to calculate the solid angle subtended by a detector when viewed from \((x^b_p,y^b_p,z^b_p)\). Figure 5.9 helps to determine the relationship between \(\Delta \theta\) and \(\Delta \phi\) and \(\Delta \Omega_p\).

\[
\Delta \Omega_p = \frac{\pi ab}{R_o^2}
\]  

(5.41)
\[ \Delta \theta = \frac{2a}{R_o} \]  
(5.42)

\[ \Delta \phi = \frac{2b}{R_o} \]  
(5.43)

Therefore,

\[ \Delta \Omega_p = \frac{\pi \Delta \theta \Delta \phi}{4} \]  
(5.44)

Vectors \( r_1 \) and \( r_2 \) are also used to determine the actual scattering angle, \( \theta_s \).

\[ \theta_s = \frac{\cos^{-1} \left( \frac{r_1 \cdot \hat{y}^p}{|r_1|} \right) + \cos^{-1} \left( \frac{r_2 \cdot \hat{y}^p}{|r_2|} \right)}{2} \]  
(5.45)

The average differential scattering cross section for a particle located at \((x_p^b, y_p^b, z_p^b)\) is calculated from

\[ \frac{d\overline{C}_{av}}{d\Omega} = \frac{1}{\Delta \Omega} \int_{\theta_1}^{\theta_2} \frac{d\overline{C}}{d\Omega}(\Omega, x, \tilde{n})d\theta \]  
(5.46)

The average solid angle and differential scattering cross section for each detector is determined by considering a large number of particle positions within the scattering volume. For each particle position, the vectors \( r_1, r_2, r_3, \) and \( r_4 \) are determined in particle coordinates. Then, \( \Delta \Omega_p \) is calculated from Equation 5.44 and \( \frac{d\overline{C}_{av}}{d\Omega} \) is calculated from Equation 5.46. The average solid angle subtended by each detector, \( \Delta \Omega_j \), and the average differential scattering cross section for each detector, \( \frac{d\overline{C}_{av}}{d\Omega} \), are determined by averaging \( \Delta \Omega_p \) and \( \frac{d\overline{C}_{av}}{d\Omega} \). The angular scattering cross sections for Freon-12 can then be calculated from Equation 5.5, and the scattering correction factors are obtained from Equation 5.6. A FORTRAN program titled nephelometer.f was written to carry out these calculations, and a complete listing of the code is contained in the Appendix E. The number of particle
positions is doubled until $\Delta \Omega_j$ and $\frac{dC_{ij}}{d\Omega}$ converge to within 1.0% to prove grid independence.

Pressure and Temperature Dependence of the Scattering Correction Factors

From Equations 5.23 and 5.24, it can be seen that refractive index and the effective scattering diameter of Freon-12 depend on the temperature and pressure. Therefore, the scattering correction factors will also depend on the temperature and pressure. This dependency needs to be determined and corrected when operating the nephelometer under conditions other than standard temperature and pressure. In order to determine the pressure and temperature dependence, Equations 5.8 and 5.23 are substituted into Equation 5.34.

$$d_F = \left[ \frac{\left\{ 2 + (\bar{n}_{ep} - 1) \frac{T_{ep} P}{P_{ep} T} \right\}^2}{\left( \frac{P N_A}{RT} \right)^2} \right]^{1/6}$$

(5.47)

where

$$A_1 = \frac{9}{\pi^2} \frac{6 + 3\rho}{6 - 7\rho}$$

(5.48)

Equation 5.47 can be simplified to

$$d_F = \left[ A_2 \left( A_3 + \frac{T}{P} \right)^2 \right]^{1/6}$$

(5.49)

where

$$A_2 = \frac{4A_1 R^2}{N_A^2}$$

(5.50)

$$A_3 = \frac{\bar{n}_{ep} - 1}{2} \frac{T_{ep}}{P_{ep}}$$

(5.51)

From Equation 5.29, it is seen that
\[ \frac{d\hat{C}}{d\Omega} \propto d_r^2 (n-1)^2 \] (5.52)

Substituting Equations 5.23 and 5.49 into Equation 5.52 gives

\[ \frac{d\hat{C}}{d\Omega} \propto 1 + 2A_3 \frac{P}{T} + \left( A_3 \frac{P}{T} \right)^2 \] (5.53)

Since \( A_3 \) is approximately 0.1, \( P \) is approximately 1 atm, and \( T \) is approximately 300 K, the dependence of the differential scattering cross sections on \( P \) and \( T \) is negligible. Therefore, only the change in the particle number density with temperature and pressure needs to be accounted for. Since the scattering correction factors depend linearly on the particle number density, the scattering correction factors need only be calculated using nephelometer.at standard temperature and pressure. At conditions other than standard temperature and pressure, the scattering correction factors should be obtained from Equation 5.54 rather than 5.6

\[ \text{SCF}_j = \frac{C_j T_{sp} P}{M_j T P_{sp}} \] (5.54)

Uncertainty in the Scattering Correction Factors

The uncertainty in the scattering correction factors is determined by the uncertainty in the calculation of the theoretical angular scattering correction factors and the uncertainty in the light scattering measurements used in the calibration process. The theoretical angular scattering cross sections are obtained from Equation 5.5 which is repeated here for convenience.

\[ C_j = N_e V_j \frac{d\hat{C}^{\text{av}}}{d\Omega} \Delta\Omega_j \] (5.55)

Standard error analysis gives the relative uncertainty in the theoretical \( C_j \) for Freon-12. When calculating the uncertainties in this manner, it is assumed that all the possible errors make positive contributions to the total uncertainty. Therefore, these are conservative estimates of the experimental errors, and the actual errors may be somewhat less than the calculated uncertainties.
\[
\frac{\delta C_j}{C_j} = \left( \frac{\delta N_c}{N_c} + \frac{\delta V_j}{V_j} + \frac{\delta (\Delta \Omega_j)}{\Delta \Omega_j} + \frac{\delta \left( \frac{d\tilde{C}_{jav}}{d\Omega} \right)}{\frac{d\tilde{C}_{jav}}{d\Omega}} \right) \tag{5.56}
\]

For the 15 channel nephelometer, the parameters on the right hand side of Equation 5.56 are obtained from the following calculations.

\[
\frac{\delta N_c}{N_c} = \frac{\delta P}{P} + \frac{\delta T}{T} \approx 3.6 \times 10^{-4} \tag{5.57}
\]

\[
\frac{\delta V_j}{V_j} = \frac{3d^2 \delta b}{V_j} \tag{5.58}
\]

\[
\frac{\delta (\Delta \Omega_j)}{\Delta \Omega_j} = \frac{\delta A_i}{A_j} + \frac{\delta R_n}{R_o} \tag{5.59}
\]

Determining the uncertainty in the average differential scattering cross sections for each detector is more difficult. The average differential scattering cross sections are a function of the scattering angle, size parameter, and the refractive index. Therefore, the uncertainty in the average differential scattering cross sections can be expressed as

\[
\delta \left( \frac{d\tilde{C}_{jav}}{d\Omega} \right) = \left\{ \delta \Theta \frac{\partial}{\partial \Theta} + \delta x \frac{\partial}{\partial x} + \delta n \frac{\partial}{\partial n} + \delta k \frac{\partial}{\partial k} \right\} \left( \frac{d\tilde{C}_{jav}}{d\Omega} \right) \tag{5.60}
\]

Since \( \frac{\partial}{\partial n} \left( \frac{d\tilde{C}_{jav}}{d\Omega} \right) \) and \( \frac{\partial}{\partial k} \left( \frac{d\tilde{C}_{jav}}{d\Omega} \right) \) are several orders of magnitude less than \( \frac{\partial}{\partial \Theta} \left( \frac{d\tilde{C}_{jav}}{d\Omega} \right) \)
and \( \frac{\partial}{\partial x} \left( \frac{d\tilde{C}_{jav}}{d\Omega} \right) \), only the first two terms in Equation 5.52 need to be considered. For molecular scattering [van de Hulst, 1981]

\[
\frac{d\tilde{C}_{jav}}{d\Omega} = \frac{d\tilde{C}}{d\Omega} = g(\bar{n})x^4(1 + \cos^2 \theta) \tag{5.61}
\]
Using the effective scattering diameter and the refractive index obtained earlier for Freon-12, \( g(\bar{n}) \) can be calculated from Mie theory, and expressions for \( \frac{\partial}{\partial \theta} \left( \frac{\partial \bar{C}_{j}}{\partial \Omega} \right) \) and

\[
\frac{\partial}{\partial \theta} \left( \frac{\partial \bar{C}_{j}}{\partial \Omega} \right) \text{ can be obtained.}
\]

\[
\frac{\partial}{\partial \theta} \left( \frac{\partial \bar{C}_{j}}{\partial \Omega} \right) = -2g(\bar{n})x^6 \cos \theta \sin \theta = -1.3 \times 10^{-27} \cos \theta \sin \theta \quad \text{(cm}^2) \quad (5.62)
\]

\[
\frac{\partial}{\partial \theta} \left( \frac{\partial \bar{C}_{j}}{\partial \Omega} \right) = 6g(\bar{n})x^5(1 + \cos^2 \theta) = 9.25 \times 10^{-25}(1 + \cos^2 \theta) \quad \text{(cm}^2) \quad (5.63)
\]

From Equation 5.34 it is seen that the effective scattering size parameter of Freon-12 is primarily a function of the particle concentration, \( N_c \). Therefore, the uncertainty in the effective size parameter is given by

\[
\delta x = \frac{x \delta N_c}{3N_c} = 5.1 \times 10^{-6} \quad (5.64)
\]

The uncertainty in the position of the polar angle, \( \delta \theta \), is estimated to be 0.1 degree. Equation 5.60 is now used to calculate the uncertainty in the average differential scattering cross sections, and Equation 5.56 is used to calculate the relative uncertainty in the angular scattering cross sections. The relative uncertainties in the solid angle, in the average differential scattering cross sections, and in the angular scattering cross sections for the 15 channel nephelometer are listed in Table 5.4.

The relative uncertainty in the scattering correction factors is calculated using the uncertainty in the theoretical angular scattering cross sections and the uncertainty in the measurements of the light scattered by Freon-12.

\[
\frac{\delta \text{SCF}}{\text{SCF}} = \frac{\delta C_j}{C_j} + \frac{\delta M_j^c}{M_j^c} \quad (5.65)
\]

Summary of the Nephelometer Calibration Procedure

Theoretical values of the angular scattering cross sections for Freon-12 were calculated at standard temperature and pressure for both the 15 and 36 channel nephelometers. The Freon-12 angular scattering cross sections for the 15 channel
nephelometer are listed in Table 5.5. These values and measurements of the light scattered by Freon-12 can be used to calculate the scattering correction factors. Figure 5.10 shows a set of typical scattering correction factors. The scattering correction factors depend on various nephelometer settings such as the laser alignment, so these factors will change slightly each time the nephelometer is calibrated.

Table 5.4. Relative Uncertainties for the 15 Channel Polar Nephelometer

<table>
<thead>
<tr>
<th>Channel</th>
<th>$\theta_j$</th>
<th>$\frac{\delta N_c}{N_c}$</th>
<th>$\frac{\delta V_j}{V_j}$</th>
<th>$\frac{\delta \Delta \Omega_j}{\Delta \Omega_j}$</th>
<th>$\delta \left( \frac{\delta C_j^{\text{avg}}}{\delta \Omega} \right)$</th>
<th>$\frac{\delta C_j}{C_j}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>23.1</td>
<td>0.00036</td>
<td>0.010</td>
<td>0.026</td>
<td>0.052</td>
<td>0.092</td>
</tr>
<tr>
<td>2</td>
<td>29.0</td>
<td>0.00036</td>
<td>0.012</td>
<td>0.026</td>
<td>0.061</td>
<td>0.103</td>
</tr>
<tr>
<td>3</td>
<td>34.9</td>
<td>0.00036</td>
<td>0.010</td>
<td>0.022</td>
<td>0.070</td>
<td>0.105</td>
</tr>
<tr>
<td>4</td>
<td>41.0</td>
<td>0.00036</td>
<td>0.011</td>
<td>0.022</td>
<td>0.077</td>
<td>0.113</td>
</tr>
<tr>
<td>5</td>
<td>47.2</td>
<td>0.00036</td>
<td>0.012</td>
<td>0.022</td>
<td>0.082</td>
<td>0.120</td>
</tr>
<tr>
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<td>0.022</td>
<td>0.084</td>
<td>0.123</td>
</tr>
<tr>
<td>7</td>
<td>60.0</td>
<td>0.00036</td>
<td>0.015</td>
<td>0.022</td>
<td>0.083</td>
<td>0.123</td>
</tr>
<tr>
<td>8</td>
<td>66.7</td>
<td>0.00036</td>
<td>0.016</td>
<td>0.022</td>
<td>0.076</td>
<td>0.117</td>
</tr>
<tr>
<td>9</td>
<td>73.7</td>
<td>0.00036</td>
<td>0.016</td>
<td>0.022</td>
<td>0.063</td>
<td>0.105</td>
</tr>
<tr>
<td>10</td>
<td>81.1</td>
<td>0.00036</td>
<td>0.017</td>
<td>0.022</td>
<td>0.043</td>
<td>0.085</td>
</tr>
<tr>
<td>11</td>
<td>88.9</td>
<td>0.00036</td>
<td>0.017</td>
<td>0.022</td>
<td>0.017</td>
<td>0.060</td>
</tr>
<tr>
<td>12</td>
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<td>0.017</td>
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<td>0.037</td>
<td>0.080</td>
</tr>
<tr>
<td>13</td>
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<td>0.00036</td>
<td>0.016</td>
<td>0.022</td>
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<td>0.105</td>
</tr>
<tr>
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</tr>
<tr>
<td>15</td>
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<td>0.013</td>
<td>0.022</td>
<td>0.084</td>
<td>0.122</td>
</tr>
</tbody>
</table>

Table 5.5. Solid Angles, Scattering Volumes, and Freon Angular Scattering Cross Sections for the 15 Channel Nephelometer.

<table>
<thead>
<tr>
<th>Channel</th>
<th>$\theta_j$</th>
<th>$\Delta \Omega_j * 10^3$ (sr) for calibration</th>
<th>$V_j (cm^3)$ for calibration</th>
<th>$\Delta \Omega_j * 10^3$ (sr) for normal operation</th>
<th>$V_j (cm^3)$ for normal operation</th>
<th>$C_j * 10^{12}$ (cm$^2$) for Freon-12</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>23.1</td>
<td>0.92</td>
<td>0.123</td>
<td>1.00</td>
<td>0.038</td>
<td>3.35</td>
</tr>
<tr>
<td>2</td>
<td>29.0</td>
<td>0.92</td>
<td>0.099</td>
<td>1.00</td>
<td>0.038</td>
<td>2.59</td>
</tr>
<tr>
<td>3</td>
<td>34.9</td>
<td>1.32</td>
<td>0.124</td>
<td>1.54</td>
<td>0.038</td>
<td>4.39</td>
</tr>
<tr>
<td>4</td>
<td>41.0</td>
<td>1.32</td>
<td>0.108</td>
<td>1.55</td>
<td>0.038</td>
<td>3.60</td>
</tr>
<tr>
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<td>0.097</td>
<td>1.55</td>
<td>0.038</td>
<td>3.01</td>
</tr>
<tr>
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<td>1.33</td>
<td>0.088</td>
<td>1.55</td>
<td>0.038</td>
<td>2.53</td>
</tr>
<tr>
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<td>1.56</td>
<td>0.038</td>
<td>2.17</td>
</tr>
<tr>
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<td>0.077</td>
<td>1.56</td>
<td>0.038</td>
<td>1.90</td>
</tr>
<tr>
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<td>73.7</td>
<td>1.33</td>
<td>0.073</td>
<td>1.56</td>
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<td>1.69</td>
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<td>81.1</td>
<td>1.33</td>
<td>0.071</td>
<td>1.56</td>
<td>0.038</td>
<td>1.56</td>
</tr>
<tr>
<td>11</td>
<td>88.9</td>
<td>1.33</td>
<td>0.071</td>
<td>1.56</td>
<td>0.038</td>
<td>1.51</td>
</tr>
<tr>
<td>12</td>
<td>97.2</td>
<td>1.33</td>
<td>0.071</td>
<td>1.56</td>
<td>0.038</td>
<td>1.54</td>
</tr>
<tr>
<td>13</td>
<td>106.3</td>
<td>1.33</td>
<td>0.073</td>
<td>1.56</td>
<td>0.038</td>
<td>1.68</td>
</tr>
<tr>
<td>14</td>
<td>116.4</td>
<td>1.33</td>
<td>0.079</td>
<td>1.56</td>
<td>0.038</td>
<td>2.02</td>
</tr>
<tr>
<td>15</td>
<td>128.3</td>
<td>1.33</td>
<td>0.090</td>
<td>1.55</td>
<td>0.038</td>
<td>2.66</td>
</tr>
</tbody>
</table>
Figure 5.10. Typical Scattering Correction Factors.
Aerosol Light Scattering Measurements

As a check on the calibration procedure, absolute measurements of the light scattered by different media were obtained and compared to expected results. The measured angular scattering cross sections were converted into the product of the particle concentration and the average differential scattering cross sections to aid these comparisons.

\[ N_e \frac{d\tilde{C}_{\text{avg}}}{d\Omega} = \frac{C_j}{\Delta\Omega_j V_j} \quad (5.66) \]

Figure 5.11 shows the measurements of the light scattered by Dust-Off®, a mixture of chlorodifluoromethane and dimethyl ether that is used in cleaning optics. The measured scattering pattern is proportional to \(1 + \cos^2\theta\) as expected. Figure 5.12 shows the measurements of the light scattered by aerosols suspended in room air. As expected, the scattering pattern is smooth due to the broad size distribution and peaks in the forward scattering angles.
Figure 5.11. Light Scattering Pattern for Dust-Off®
Figure 5.12. Light Scattering Pattern for Room Air
Chapter 6
Application of the Inversion Procedure

The results presented in Chapter 4 indicated that the inversion scheme is capable of accurately retrieving the PSDF and optical properties when the distributions are narrow and that the refractive index could be obtained in all cases. However, the calculation of the synthetic data used in Chapter 4 was based on the assumptions that the scattering particles were perfectly spherical and homogeneous and that there were no systematic errors in the measurements. Since these conditions are difficult to produce in the laboratory and are never encountered in practical systems, it must be verified that the inversion of experimentally obtained scattering measurements will still yield valid results. The capabilities of the inversion technique were further investigated by inverting the measurements of the light scattered by polystyrene spheres and by Al₂O₃ smoke produced by the combustion of an aluminized solid propellant.

Light Scattering by Polystyrene Spheres
Measurement and inversion of the light scattered by polystyrene spheres with known sizes and optical properties will further demonstrate that the nephelometer is properly calibrated. These results will also show whether slight asphericity or inhomogeneity of the particles significantly affects the inversion process. Figure 6.1 is a schematic diagram of the experimental setup for the polystyrene sphere light scattering measurements.

![Figure 6.1. Experimental Set-up for Polystyrene Sphere Light Scattering Measurements](image)

The polystyrene spheres were diluted using distilled, filtered water and nebulized using a medical nebulizer. The aerosol stream produced by the nebulizer was diluted using filtered (0.2 μm filter) compressed air. The water evaporated as the aerosol stream passed.
through a coil of heated copper tubing, and the moisture was then removed from the aerosol stream using a diffusion dryer. After passing through the dryer, the samples from the aerosol stream were drawn into the nephelometer and the optical particle counter (Particle Measuring Systems, Model LAS-X CRT) denoted by PMS-LAS in Figure 6.1.

In order to ensure that all the moisture was removed from the aerosol stream and that the sediments contained in the water did not significantly affect the light scattering measurements, clean water was nebulized and analyzed using the nephelometer and the PMS-LAS. The histogram shown in Figure 6.2 indicates that there were still some very small ($\leq 0.45 \mu m$) water droplets or sediments remaining in the aerosol stream. The light scattered by these background particles was monitored for five minutes and found to be very steady. Comparison of these measurements with stray light measurements (Figure 6.3) shows that the difference between the background and the stray light is at most 0.03 V. Since the average signal is approximately 2 V, the scattering due to the background particles will not significantly affect the light scattering measurements. The background measurements were consistent with time and were used in place of the stray light measurements when reducing the data (see Equation 5.7).

Light scattering measurements using polystyrene spheres with nominal diameters of 1.06 $\mu m$ and 2.02 $\mu m$ were obtained and inverted. The PSDF retrieved using the inversion process is compared to particle size measurements obtained using the optical particle counter. Since no attempt was made to account for the difference in the sampling losses in the tubing leading to the nephelometer and the optical particle counter, comparison of the PMS-LAS histograms and the retrieved PSDF gives only a rough indication of the accuracy of the inversion technique. Figure 6.4a shows the retrieved PSDF for the 1.06 $\mu m$ spheres, and Figure 6.4b is the corresponding histogram. Figures 6.5a and 6.5b are the retrieved PSDF and histogram for the 2.02 $\mu m$ spheres. The retrieved optical properties are compared with published values for polystyrene spheres in Table 6.1 [Devon and Rudin, 1987; Inagaki et al., 1977]. Since smaller values of the absorption index give essentially the same rms errors in the calculated scattering patterns, the retrieved values for the absorption index represent upper bounds. These results indicate light scattering measurements alone are not sufficiently sensitive to variations in the absorption index to allow the accurate retrieval of this parameter. Incorporation of an absorption or total extinction measurement into the inversion procedure may provide sufficient information to allow the accurate retrieval of the absorption index. Shaw (1979) described how to use both spectral extinction measurements and angular scattering measurements in an inversion technique.
Figure 6.3. Comparison of the Detector Voltages due to Stray Light and Background Particles
The scattering pattern based on the retrieved PSDF and optical properties was calculated using the Mie code written by Bohren and Huffman (1983). Figure 6.6 compares the measured scattering pattern to the calculated scattering pattern for the 1.06 μm polystyrene spheres. The measured and calculated scattering patterns for the 2.02 μm polystyrene spheres are compared in Figure 6.7.

Light Scattering by Al₂O₃ Smoke

Additives such as aluminum, magnesium and zirconium carbide are included in formulations of solid propellant rocket motors to increase motor performance and suppress high frequency combustion instabilities. Combustion of these additives results in metallic oxide (Al₂O₃, MgO, and ZrO₂) smoke particles which dramatically effect the optical characteristics of the plume and the motor performance. Radiative emissions from rocket plumes plays a significant role in the rocket design due to the radiant heat transfer to the nozzle, internal insulator surfaces, external equipment, and to the surface of the propellant. Radiation emitted from the exhaust plumes also plays a significant role in the detection and identification of missiles. The PSDF of the condensed phase particulates is one of the most important parameters needed to predict the two-phase flow losses in a rocket nozzle. In order to assess the effects of smoke on the radiative heat transfer and the overall motor performance and to characterize the radiation emitted from plumes, the PSDF and the complex refractive index of the metallic oxide smoke need to be determined. Brewster (1992a) included a discussion of measurements of the PSDF and the optical properties of propellant smoke in a recent review of heat transfer in heterogeneous propellant combustion systems, so measurements of the PSDF and optical properties of these particles are available in the literature for comparison.
Figure 6.4a. Retrieved PSDF for the 1.06 μm Spheres
Figure 6.5a. Retrieved PSDF for the 2.02 μm Spheres
Figure 6.6. Comparison of the Measured and Calculated Scattering Patterns for the 1.06 μm Spheres
Figure 6.7. Comparison of the Measured and Calculated Scattering Patterns for the 2.02 μm Spheres
Unfortunately, it is not possible to obtain \textit{in situ} measurements of combustion generated particles with the available nephelometer. In order to use the 15 channel nephelometer, the smoke had to be sampled and diluted. Figure 6.8 illustrates an experimental set up in which the nephelometer could be used.

![Diagram of Experimental Set Up](image)

Figure 6.8. Schematic Diagram for the Propellant Smoke Measurements

Sampling, diluting, and sending the smoke through tubing to the nephelometer will bias the measurements, so the retrieved properties will not be representative of properties of the particles in the flame. Previous investigations have determined that the PSDF of $\text{Al}_2\text{O}_3$ smoke produced in propellant combustion is bimodal. A submicron mode is produced by detached, vapor-phase oxidation of the aluminum particles, and a larger mode ($10 - 100 \mu\text{m}$) is produced by surface oxidation and condensation of the submicron smoke on the surface [Brewster, 1992b]. The large particles will settle out of the sampled stream before reaching the nephelometer, so the retrieved PSDF will be biased toward the smaller particles. The remaining particles will cool before reaching the nephelometer, so the optical properties of the particles will also change. Although, the retrieved PSDF will not be representative of that in the flame, the optical properties of the particles will be representative of the properties of smoke found in rocket plumes. The optical properties of cooled smoke are significant in determining the plume signature, so these measurements should still be of interest to the scientific community.

Ammonium perchlorate solid propellant containing 10% aluminum by weight was used in the experiments. The detailed formulation of the propellant is described by Ishihara (1991). Three different runs were made. Since the PSDF is not monodisperse, the method used in Chapter 4 to select the trial function will not work. However, analysis of the experimental setup indicated that the particles were primarily submicron, and this
information was used to constrain the inversion process. The optical particle counter was used to obtain a histogram of particle sizes to compare to the retrieved PSDF. The histograms of particle sizes measured with the PMS-LAS for each run are shown in Figures 6.9a - 6.9c. The retrieved size distributions for all 3 runs are plotted in Figure 6.10. Again, it should be noted that the sampling method used in this study is not isokinetic and no attempt was made to account for the particle losses in the tubing. Therefore, the histogram of particle sizes measured by the PMS-LAS and the PSDF retrieved by the inversion process are not representative of the size distribution of the smoke produced by the propellant.

The optical properties retrieved from the inversion of the measurements of the light scattered by the aluminized propellant are listed in Table 6.2. Plass (1965) reports the refractive index of Al₂O₃ at 1.0 μm varies between 1.78 at 1200 C and 1.81 at 2020 C, so the retrieved values of the refractive index agree fairly well with previous measurements. However, Plass (1965) reports the absorption index at 1.0 μm to range between 6.0 x 10⁻⁸ at 1200 C to 1.6 x 10⁻⁶ at 2020 C, which is much lower than the retrieved values. Brewster (1992b) found absorption indices at 1.0 μm as high as ~10⁻⁴ reported in the literature. Brewster points out that substoichiometric aluminum oxide (Al₂O₃-x) may have a significantly higher absorption since free aluminum atoms can act as free charge carriers.

Figures 6.11a through 6.11c show the measured and calculated scattering patterns for each run. The large variations in the magnitudes of the scattering patterns from run to run are due to variations in the flow rate of the dilution stream.

Table 6.2. Optical Properties of Al₂O₃ Smoke Particles at 0.84 μm

<table>
<thead>
<tr>
<th>Run</th>
<th>Refractive Index</th>
<th>Absorption Index</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.81</td>
<td>1 x 10⁻²</td>
</tr>
<tr>
<td>2</td>
<td>1.82</td>
<td>1 x 10⁻²</td>
</tr>
<tr>
<td>3</td>
<td>1.82</td>
<td>2 x 10⁻²</td>
</tr>
</tbody>
</table>
Counts/Total Number of Counts

Figure 6.9a. Histogram of Al₂O₃ Smoke Sizes, Run 1 (μm)

- 0.01-0.12
- 0.12-0.15
- 0.15-0.20
- 0.20-0.25
- 0.25-0.35
- 0.35-0.45
- 0.45-0.60
- 0.60-0.75
- 0.75-1.00
- 1.00-1.50
- 1.50-2.00
- 2.00-3.00
- 3.00-4.50
- 4.50-6.00
- 6.00-7.50
- >7.50
Counts/Total Number of Counts

Figure 6.9c. Histogram of Al2O3 Smoke Sizes, Run 3 (µm)

- 0.10-0.12
- 0.12-0.15
- 0.15-0.20
- 0.20-0.25
- 0.25-0.35
- 0.35-0.45
- 0.45-0.60
- 0.60-0.75
- 0.75-1.00
- 1.00-1.50
- 1.50-2.00
- 2.00-3.00
- 3.00-4.50
- 4.50-6.00
- 6.00-7.50
- >7.50
Figure 6.10. Retrieved PSDF of Al$_2$O$_3$ Smoke
Figure 6.11a. Comparison of the Measured and Calculated Scattering Patterns for Run 1
Figure 6.11b. Comparison of the Measured and Calculated Scattering Patterns for Run 2
Figure 6.11c. Comparison of the Measured and Calculated Scattering Patterns for Run 3
Summary

In this chapter, the inversion technique was applied to measurements of the light scattered by two monodispersions of polystyrene spheres. The results of these inversions demonstrates that the inversion scheme developed in this study can successfully retrieve the PSDF and refractive index of monodisperse polystyrene spheres. Attempts to retrieve the absorption index were less successful. Due to the relative insensitivity of light scattering measurements to variations in the absorption index of weakly absorbing particles, only an upper bound on the absorption index could be determined. Increased sensitivity to the absorption index may be obtained by incorporating an absorption measurement or a total extinction measurement into the inversion scheme. Shaw (1979) discussed the combined use of angular scattering and spectral extinction measurements in an inversion technique. The possibility that spectral extinction measurements may provide the additional information needed to accurately retrieve the absorption index should be further examined.

Application of the inversion technique to measurements of the light scattered by smoke from an aluminized propellant indicate that the technique could be a useful tool in analyzing propellant smoke. If an experimental facility capable of measuring the light scattered by metallic oxide smoke particles in situ were developed, the inversion technique could be used to determine the PSDF and optical properties of metallic oxide smokes produced in the combustion of solid rocket propellants. A review of heat transfer in heterogeneous propellant systems discussed the variability of these parameters from motor to motor [Brewster, 1992b]. Because of the many factors which influence the size and optical properties of propellant smoke, further systematic studies are required to predict these properties with confidence. Accurate measurements or predictions of smoke properties would greatly improve radiative transfer calculations in propellant flames and in rocket plumes that are obtained using codes such as the JANNAF Standardized Infrared Radiation Model.
Chapter 7
Summary and Conclusions

Techniques for solving linear inverse problems with discrete data have received considerable attention in the literature and are of great interest due to the wide range of potential applications. The wide variety of applications is primarily due to the fact that properties of a physical sample can be determined from the interaction of the sample with radiation from a known source. This dissertation focused on a technique for solving the inverse light scattering problem and determining the properties of aerosol particles.

The fundamentals of light scattering theory and the mathematical formulation of the inverse light scattering problem were presented in Chapter 2. It was shown that the inverse light scattering problem is classified as a linear inverse problem with discrete data. Several techniques for solving this class of problems were discussed in Chapter 3. In general, solution techniques are either analytical or empirical methods. Analytical methods involve formal solutions of the integral equation. Constraints based on a priori information are required to successfully apply analytical solution techniques. 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 to obtain a least squares fit of the measured data. Empirical methods are usually tedious and computationally intensive.

Four commonly used analytical inversion methods were discussed in Chapter 3. The Backus-Gilbert technique is advantageous in that no a priori information is required, and information regarding the resolution of the retrieved values is obtained. However, this technique is computationally intensive and is more sensitive to errors in the measurements than other inversion schemes. This method could be used to obtain preliminary solutions that would serve as constraints for other analytical inversion techniques. The statistical method provides a simple and direct way of obtaining a solution to inverse problems. However, statistical methods require information regarding the statistical properties of the unknown function and the measurement errors which may not be readily available. The Phillips-Twomey method or constrained linear inversion is a fairly simple technique to apply. The most significant difficulty with this method is determining the proper weight to assign to the constraint. This inversion process can become computationally intensive if the weighting parameter for the constraint is not well chosen. Due to the complex mathematics involved in the eigenfunction method, this method is more difficult to apply than other analytical techniques. The primary advantage of the eigenfunction technique is
that the proper weight for the constraint can be determined in a relative simple and direct manner.

One empirical inversion technique, the optical strip-map technique, was discussed in this dissertation. The optical strip-map technique overcomes many of the difficulties that plague empirical techniques. However, it does not provide any information regarding the absorption index, and it cannot be extended to measurements of the light scattered by polydispersions. It was found that low resolution strip-maps are helpful in selecting constraints for analytical inversion methods when the PSDF is narrow.

The inversion technique described in Chapter 4 is a combination of the eigenfunction method and the optical strip-map method. Using synthetic data sets, it was demonstrated that the technique is capable of retrieving the PSDF and optical properties of a weakly absorbing spherical particle or of a narrow distribution of non-absorbing spherical particles from measurements of the angular light scattering pattern. Application of the technique to polydispersions was also examined. It was found that unless accurate a priori information regarding the PSDF is available, the retrieved PSDF may be inaccurate. If a priori information cannot be obtained from analysis of the particular environment in which the light scattering measurements are made, the complementary use of probe sampling techniques is recommended. Further research is needed to investigate the possibility that the trial function could be selected without the use of a priori information. Further modifications of the inversion technique will focus in this area.

Absolute scattering measurements are required to use the inversion technique described in Chapter 4. A procedure for calibrating multi-channel polar nephelometers for absolute scattering measurements has been developed and was described in Chapter 5. Theoretical values of the light scattered by Freon-12 are determined from an analysis of the geometry of the nephelometer and the properties of Freon-12. Measurements of the light scattered by Freon-12 and the theoretical angular scattering cross sections are used to calculate scattering correction factors for each detector.

Application of the inversion technique to actual light scattering measurements was discussed in Chapter 6. The light scattered by two monodispersions of polystyrene spheres was measured and inverted. The results of these inversions demonstrated that the inversion scheme developed in this study can successfully retrieve the PSDF and refractive index of monodisperse spheres. Attempts to retrieve the absorption index were less successful. Due to the relative insensitivity of light scattering measurements to variations in the absorption index, only an upper bound on the absorption index of the polystyrene spheres could be determined. Increased sensitivity to the absorption index may be obtained
by incorporating an absorption measurement or a total extinction measurement into the inversion scheme. The possibility that such a measurement may provide the additional information needed to accurately retrieve the absorption index should be investigated.

Application of the inversion technique to measurements of the light scattered by $\text{Al}_2\text{O}_3$ smoke produced by combustion of an aluminized solid propellant demonstrated that the technique has the potential of becoming a useful tool. Determining the properties of smoke produced by metalized propellants is necessary to calculate the radiative transfer in rocket motors and plumes, so suitable measurement techniques are of considerable interest. If an experimental facility capable of \textit{in situ} measurements were developed, the inversion technique could be used to determine the PSDF and optical properties of metallic oxide propellant smoke. Because of the many factors which influence the size and optical properties of propellant smoke, systematic studies are needed to predict these properties with confidence. Accurate measurements of smoke properties would greatly improve radiative transfer calculations in propellant flames and in rocket plumes which are obtained using codes such as the JANNAF Standardized Infrared Radiation Model.
References


Appendix A
Radiation from an Accelerated Charge

The classical theory of emission of electromagnetic radiation by an accelerated charge was first developed by J.J. Thomson in 1903 and is briefly explained in this appendix. Similar derivations are found in many texts on electromagnetics or physics [See, for example, Lorrain et al. (1988) or Eisberg and Resnick (1985)].

The electric field strength of a point charge, Q, at rest can be obtained from Gauss's law.

$$E = \frac{Q\hat{r}}{4\pi \epsilon_0 r^2}$$  \hspace{1cm} (A.1)

The field is radial and static, so no energy is radiated away. If the charged particle moves at a constant velocity, there is a magnetic as well as an electric field, so the total energy of the system is greater than in the static case. When the charged particle moves with constant velocity, no energy is radiated away. This can be shown to be true by transforming to a reference frame where the particle is stationary. Since the behavior of the particle, including whether or not it radiates, cannot depend on the reference frame, the electric and magnetic fields created by the moving charge must adjust themselves so the total energy stored in the fields remains constant. However, if the charge is accelerated, the fields develop kinks that propagate radially outward at the speed of light and the fields can no longer adjust themselves so that no energy is radiated away. An understanding of this behavior can be obtained by considering the electric field lines shown in Figure A.1. Such an electric field could be created at time t' by accelerating a point charge that is initially at rest at the origin with a constant acceleration, a, to a constant velocity, v, which is much less than the speed of light, c. The time during which the charge is accelerated, t, is assumed to be extremely short, so the field at distances greater than ct' from origin corresponds to the original static field. The field at distances less than c(t'-t) corresponds to the field of the charge moving at the constant velocity v when it is located a distance vt' from the origin. In the region between ct' and c(t'-t), the electric field clearly has a component in the \( \hat{\theta} \) direction. Assuming the electric field is straight in the transition region, the geometry illustrated in Figure A.1 can be used to determine the ratio of the polar component to the radial component of the electric field vector.
Multiplying Equation A.2 by the magnitude of the radial component of the electric field obtained from Equation A.1 gives

\[
E_r = \frac{Q a \sin \theta}{4\pi \varepsilon_0 c^2 r}
\]  
(A.3)

Since \( E_r \) is proportional to \( 1/r^2 \) and \( E_\theta \) is proportional to \( 1/r \), the electric field is polarized in the \( \hat{\theta} \) direction for sufficiently large \( r \). Therefore,

\[
E = \frac{Q a \sin \theta}{4\pi \varepsilon_0 c^2 r} \hat{\theta}
\]  
(A.4)

The magnetic flux density in the transition region can be calculated from Ampere's law,

\[
\oint_C \mathbf{B} \cdot d\mathbf{l} = \varepsilon_0 \mu_0 \int_A \frac{\partial \mathbf{E}}{\partial t} \cdot d\mathbf{A} = \varepsilon_0 \mu_0 \frac{d\Phi_E}{dt}
\]  
(A.5)
where C is the circle perpendicular to the plane of the paper shown in Figure A.1. Since the magnetic field is created by the motion of the charge in the plane of the page, B will be everywhere parallel to the line element dl. Therefore, the integral in Equation A.5 can be evaluated, and the following expression is obtained for B.

$$B = \frac{\epsilon_0 H_0}{2\pi r \sin \theta} \frac{d\Phi_E}{dt} \vec{\phi}$$  \hspace{1cm} (A.6)

The rate of change of the electric flux can be found from the following considerations. At \( t' \), just before the kink in the electric field passes through C, the electric flux through C is due to a point charge at the origin. Therefore, the electric field vector is obtained from Equation A.1, and the flux of E through C is given by

$$\Phi_E(t') = \int_{\text{A}} E \cdot dA = \frac{Q}{4\pi \epsilon_0} \int_0^\theta \frac{2\pi r \sin \theta' r d\theta'}{r^2} = \frac{Q(1 - \cos \theta)}{2\epsilon_0}$$  \hspace{1cm} (A.7)

After \( t \) seconds, the kink has passed through C, and the electric flux through C is now due to a point charge that is a distance \( vt \) from the origin and is moving with velocity \( v \). The electric flux through C at \( t'+t \) is given by

$$\Phi_E(t'+t) = \frac{Q(1 - \cos(\theta + \Delta \theta))}{2\epsilon_0}$$  \hspace{1cm} (A.8)

Therefore,

$$\frac{d\Phi_E}{dt} = \lim_{t \to 0} \frac{Q \cos \theta - \cos(\theta + \Delta \theta)}{t} = \frac{Q \sin \theta}{2\epsilon_0} \frac{d\theta}{dt}$$  \hspace{1cm} (A.9)

From Figure A.1, it can be seen that

$$\frac{d\theta}{dt} = \frac{v t \sin \theta}{a} = \frac{a \sin \theta}{c}$$  \hspace{1cm} (A.10)

Combining Equations A.6, A.9, and A.10 gives the final expression for the magnetic flux density
The Poynting vector or the intensity of the radiation emitted by the accelerated charge can now be calculated using Equations A.4 and A.11.

\[ B = \frac{Qa \sin \theta}{4\pi \varepsilon_0 c^3 r} \hat{\phi} = \frac{E_\theta}{c} \hat{\phi} \quad (A.11) \]

\[ S = \frac{1}{\mu_0} E \times B \]

\[ S = \frac{Q^2 a^2 \sin^2 \theta}{16\pi^2 \varepsilon_0 c^3 r^2} \hat{\phi} \quad (A.12) \]
Appendix B
Calculation of the Differential Scattering Cross Sections from Mie Theory

When particles are illuminated by a beam of light, the amount of energy absorbed and the amount and angular distribution of the scattered energy depends in a detailed way on the shape, size, and composition of the particle. Assuming elastic, single scattering of monochromatic light by independent particles, the problem of determining the absorption and scattering due to the particles simplifies to that of finding the electromagnetic field inside the particles and in the medium surrounding the particle. Following the notation used by Bohren and Huffman (1983), the electromagnetic field inside the particle will be denoted by \((E_1,H_1)\), the field in the surrounding medium by \((E_2,H_2)\), the scattered field by \((E_s,H_s)\) and the incident field by \((E_i,H_i)\) as shown in Figure B.1.

Extinction due to an Arbitrarily Shaped Particle

The electromagnetic fields inside an arbitrarily shaped particle and in the surrounding medium must satisfy Maxwell's equations for a homogenous, isotropic, linear and stationary medium.

\[
\begin{align*}
\nabla \cdot E &= 0 \quad \text{(B.1)} \\
\nabla \cdot H &= 0 \quad \text{(B.2)} \\
\n\nabla \times E &= -\mu \frac{\partial H}{\partial t} \quad \text{(B.3)} \\
\n\nabla \times H &= \varepsilon \frac{\partial E}{\partial t} \quad \text{(B.4)}
\end{align*}
\]

It is assumed that the electric wave and the magnetic wave are plane harmonic waves,

\[
\begin{align*}
E &= E(z) \exp(ik \cdot z - i\omega t) \quad \text{(B.5)} \\
H &= H(z) \exp(ik \cdot z - i\omega t) \quad \text{(B.6)}
\end{align*}
\]

where \(k\) is the wave vector for the surrounding medium. Since an \(\exp(-i\omega t)\) time dependence is assumed for all the fields, all the time derivatives can be replaced by a factor
of -iw. Taking the curl of Equations B.3 and B.4 and using the vector identity \( \nabla \times (\nabla \times \mathbf{A}) = (\nabla \cdot \mathbf{A}) - \nabla \cdot (\nabla \mathbf{A}) \) gives

\[
\begin{align*}
\nabla^2 \mathbf{E} + \kappa^2 \mathbf{E} &= 0 \\
\nabla^2 \mathbf{H} + \kappa^2 \mathbf{H} &= 0
\end{align*}
\]  

(B.7)  

(B.8)

where \( \kappa = \omega^2 \varepsilon \mu = \frac{2\pi}{\lambda} \). Therefore, the electric and magnetic fields inside the particle and in the surrounding medium must satisfy the vector wave equation and be divergence free.

The matching conditions at the boundary between the particle and the surrounding medium are determined from the following considerations. Integrating Equations B.3 and B.4 over the area, \( \mathcal{A} \), enclosed by the path \( \mathcal{C} \) shown in Figure B.1 gives

\[
\begin{align*}
\oint \nabla \times \mathbf{E} \cdot d\mathbf{A} &= -\varepsilon \frac{\partial}{\partial t} \oint \mathbf{H} \cdot d\mathbf{A} \\
\oint \nabla \times \mathbf{H} \cdot d\mathbf{A} &= \mu \frac{\partial}{\partial t} \oint \mathbf{E} \cdot d\mathbf{A}
\end{align*}
\]  

(B.9)  

(B.10)

The left hand sides of Equations B.9 and B.10 can be reduced to integrals over the closed path \( \mathcal{C} \) by application of Stokes theorem. The right hand sides of Equations B.9 and B.10 are eliminated by letting the height of path \( \mathcal{C} \) become infinitesimal.

\[
\begin{align*}
\oint \mathbf{E} \cdot d\mathbf{l} &= (\mathbf{E}_2 - \mathbf{E}_1) \cdot \hat{n} = 0 \\
\oint \mathbf{H} \cdot d\mathbf{l} &= (\mathbf{H}_2 - \mathbf{H}_1) \cdot \hat{n} = 0
\end{align*}
\]  

(B.11)  

(B.12)

Therefore, the boundary conditions for the electric and magnetic fields at the interface between the particle and the surrounding medium can be written

\[
\begin{align*}
(\mathbf{E}_2 - \mathbf{E}_1) \times \hat{n} &= 0 \\
(\mathbf{H}_2 - \mathbf{H}_1) \times \hat{n} &= 0
\end{align*}
\]  

(B.13)  

(B.14)
In summary, the electromagnetic field scattered by an arbitrary particle is obtained by solving the vector wave equations (Equations B.7 and B.8) for the electric and magnetic fields inside and outside the particle subject to Maxwell's equations (Equations B.1 through B.4) and the conditions that the tangential components of the electric and magnetic fields are continuous at the boundary between the particle and the surrounding medium (Equations B.13 and B.14). Because of the mathematical complexity, analytical solutions have only been obtained for particles with relatively simple geometries and properties. The most important of the analytical solutions gives the electromagnetic field scattered by a homogeneous, isotropic, linear sphere of arbitrary radius and refractive index. An overview of the mathematical description of the light scattered by a sphere is given in the next section.

**Mie Theory**

The following is an overview of Mie theory. A complete derivation is presented in chapter 4 of Bohren and Huffman (1983), and a helpful overview is presented in chapter 9 of van de Hulst (1981).

Both the internal and scattered electromagnetic fields must satisfy Equations B.7 and B.8 and the boundary conditions specified by Equations B.13 and B.14 as well as Equations B.1 through B.4. Vector fields satisfying these equations are found in the following manner. Construct a vector function \( \mathbf{M} \) such that
\[ M = \nabla \times (r \psi) \quad (B.15) \]

where \( r \) is a constant radius vector and \( \psi \) is a scalar function. Now, substituting \( M \) into the vector wave equation gives

\[ \nabla^2 M + \kappa^2 M = \nabla \cdot \nabla (\nabla \times r \psi) + \kappa^2 \nabla \times (r \psi) \quad (B.16) \]

Using tensor notation, the right hand side of Equation B.16 can be rewritten as

\[ \text{RHS} = \frac{\partial}{\partial x_i} \frac{\partial}{\partial x_i} (\epsilon_{jkl} \frac{\partial}{\partial x_k} r_i \psi) + k^2 (\epsilon_{jkl} \frac{\partial}{\partial x_k} r_i \psi) \quad (B.17) \]

Factoring out common terms and returning to vector notation gives

\[ \nabla^2 M + \kappa^2 M = \nabla \times (\nabla^2 \psi + \kappa^2 \psi) \quad (B.18) \]

Therefore, \( M \) satisfies the vector wave equation if \( \psi \) satisfies the scalar wave equation. That is if

\[ \nabla^2 \psi + \kappa^2 \psi \quad (B.19) \]

then

\[ \nabla^2 M + \kappa^2 M \quad (B.20) \]

Now, construct another vector function such that

\[ N = \frac{\nabla \times M}{\kappa} \quad (B.21) \]

Substituting \( N \) into the vector wave equation gives

\[ \nabla^2 N + \kappa^2 N = \frac{\nabla \cdot \nabla (\nabla \times M)}{\kappa} + \kappa \nabla \times M \quad (B.22) \]
Multiplying through by $k$ and using tensor notation, the right hand side of Equation B.22 can be rewritten as

$$\text{RHS} = \frac{\partial}{\partial x_i} \left( \varepsilon_{jkl} \frac{\partial}{\partial x_k} M_j \right) + \kappa^2 \left( e_{jkl} \frac{\partial}{\partial x_k} M_j \right)$$

(B.23)

Again, factoring out common terms and returning to vector notation gives

$$\kappa \left( \nabla^2 N + \kappa^2 N \right) = \nabla \times \left( \nabla^2 M + \kappa^2 M \right)$$

(B.24)

and it is can be seen that $N$ satisfies the vector wave equation if $M$ satisfies the vector equation.

Now, consider the curl of $N$.

$$\nabla \times N = \frac{\nabla \times \nabla \times M}{\kappa}$$

(B.25)

Rewriting the right hand side in tensor notation gives

$$\text{RHS} = \frac{1}{\kappa} \left\{ \varepsilon_{ijk} \frac{\partial}{\partial x_j} \varepsilon_{klm} \frac{\partial}{\partial x_k} M_m \right\}$$

$$\text{RHS} = \frac{1}{\kappa} \left\{ \varepsilon_{ijkl} \varepsilon_{kln} \frac{\partial}{\partial x_j} \frac{\partial}{\partial x_k} M_n \right\}$$

$$\text{RHS} = \frac{1}{\kappa} \left\{ (\delta_{i}^{j} \delta_{k}^{m} - \delta_{i}^{m} \delta_{k}^{j}) \frac{\partial}{\partial x_j} \frac{\partial}{\partial x_k} M_m \right\}$$

$$\text{RHS} = \frac{1}{\kappa} \left\{ \frac{\partial}{\partial x_j} \frac{\partial}{\partial x_k} M_j - \frac{\partial}{\partial x_j} \frac{\partial}{\partial x_i} M_i \right\}$$

(B.26)

Rewriting Equation B.26 in vector notation and substituting it back into Equation B.25 gives

$$\nabla \times N = \frac{1}{\kappa} \nabla (\nabla \cdot M) - \frac{1}{\kappa} \nabla^2 M$$

(B.27)
Since the divergence of a curl is zero, the first term on the right hand side of Equation B.27 drops out. Substituting Equation B.20 into Equation B.27 gives

$$\nabla \times \mathbf{N} = \kappa \mathbf{M} \quad (B.28)$$

It has been shown that $\mathbf{M}$ and $\mathbf{N}$ satisfy the vector wave equation if $y$ is chosen so that it satisfies the scalar wave equation. It has also been demonstrated that the curl of $\mathbf{N}$ is proportional to $\mathbf{M}$, and the curl of $\mathbf{M}$ is proportional to $\mathbf{N}$ by definition. Therefore $\mathbf{M}$ and $\mathbf{N}$ have all the required properties of an electromagnetic field. The scalar function $y$ is known as the generating function for the vector harmonics $\mathbf{M}$ and $\mathbf{N}$, and it is determined by the geometry of the problem. In spherical coordinates, the scalar wave equation is given by

$$\frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial \psi}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left( \sin \theta \frac{\partial \psi}{\partial \theta} \right) + \frac{1}{r^2 \sin^2 \theta} \frac{\partial^2 \psi}{\partial \phi^2} + \kappa^2 \psi = 0 \quad (B.29)$$

Solutions to the scalar wave equation are obtained by separation of variables and are given by

$$\psi_{emn} = \cos m \phi P_n^m(\cos \theta) z_n(\kappa r) \quad (B.30)$$

$$\psi_{omn} = \sin m \phi P_n^m(\cos \theta) z_n(\kappa r) \quad (B.31)$$

The $P_n^m$ are the associated Legendre functions of the first kind of degree $n$ and order $m$, and the $z_n(\kappa r)$ is any of the four spherical Bessel functions [Bohren and Huffman, 1983].

$\mathbf{M}_{yemn}$, $\mathbf{N}_{yemn}$, $\mathbf{M}_{yomn}$, $\mathbf{N}_{yomn}$ are the spherical vector harmonics associated with the solutions $y_{emn}$ and $y_{omn}$ of the scalar wave equation when $z_n(\kappa r)$ is given by the spherical Hankel function that corresponds to an outgoing spherical wave. For these conditions, the following electric and magnetic field vectors satisfy Equations B.7 and B.8 in the region surrounding a homogeneous spherical particle and are divergence free [Bohren and Huffman, 1983].

$$\mathbf{E}_n = \sum_{n=1}^{\infty} E_n \left( i a_n \mathbf{N}_{e1n} - b_n \mathbf{M}_{e1n} \right) \quad (B.32)$$
The $E_n$ in Equations B.32 and B.33 is determined by the polarization and amplitude of the incident beam. The $a_n$ and $b_n$ are given by

\[ a_n = \frac{\psi'_n(\hat{n}x)\psi_n(x) - \tilde{n}\psi'_n(\hat{n}x)\psi'_n(x)}{\psi'_n(\hat{n}x)\xi_n(x) - \tilde{n}\psi'_n(\hat{n}x)\xi'_n(x)} \]  

\[ b_n = \frac{\tilde{n}\psi'_n(\hat{n}x)\psi_n(x) - \psi_n(\hat{n}x)\psi'_n(x)}{\tilde{n}\psi'_n(\hat{n}x)\xi_n(x) - \psi_n(\hat{n}x)\xi'_n(x)} \]

where $\psi_n$ and $\xi_n$ are Riccatti-Bessel functions. That Equations B.32 and B.33 are in fact solutions to Equations B.7 and B.8 and are divergence free is easily verified through direct substitution.

Now that expressions for the scattered fields have been obtained, the power scattered in each direction is determined by calculating the Poynting vector as a function of the scattering angle. The scattered power is generally normalized by the incident irradiance to give differential scattering cross sections or by the incident irradiance and the total scattering cross section to give the phase function. For an unpolarized incident beam, the differential scattering cross sections are given by [Bohren and Huffman, 1983]

\[ \frac{d\tilde{C}}{d\Omega}(\theta, x, \tilde{n}) = \frac{1}{2\kappa^2} \left( |S_1|^2 + |S_2|^2 \right) \]  

where

\[ S_1(\theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (a_n \pi_n + b_n \tau_n) \]  

\[ S_2(\theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} (a_n \tau_n + b_n \pi_n) \]  

\[ \pi_n(\cos \theta) = \frac{1}{\sin \theta} P_n^1(\cos \theta) \]
The phase function can be calculated by dividing the differential scattering cross sections by the total scattering cross section. The phase function defined by Equation B.41 differs from the phase function commonly used in heat transfer calculations by a factor of $4\pi$ [Brewster, 1992a].

$$p(\theta, x, \bar{n}) = \frac{1}{C_{\text{sc}}} \frac{d\tilde{C}}{d\Omega}(\theta, x, \bar{n}) \quad \text{(B.41)}$$
Appendix C
Inversion Programs User's Guide

This appendix demonstrates the use of the FORTRAN program, invert.f, and two auxiliary programs that have been written to implement the inversion technique described in Chapter 4. The inputs required by invert.f are measured angular scattering cross sections and corresponding imprecision estimates as a function of the scattering angle. The program also requires a library of Mie intensity functions be created by the first auxiliary program, mie.f. The second auxiliary program, datgen.f, can be used to create synthetic data sets for practice inversions and to generate scattering patterns that serve as guide in the inversion process.

Mie.f

Prior to using the inversion code, a library of Mie intensity functions must be created using mie.f. This program is based on the mie code written by Bohren and Huffman (1983). The mie code was validated by comparing the calculated mie intensity functions with those calculated by Wiscombe (1979). The inputs to mie.f are the wavelength of the incident laser beam, the range of particle diameters to be considered, the range of real refractive indices to be considered, the number of detectors, and the scattering angle and the angular resolution for each detector. The scattering angle of a detector, $\theta_j$, is the angle between the direction of incidence and the line that passes through the center of the scattering volume and the center of the detector. The angular resolution of a detector, $\Delta \theta$, is the angle between the line that passes through the center of the scattering volume and the center of the detector and the line that passed through the center of the scattering volume and the edge of the detector. Therefore, the detector with a scattering angle of $\theta_j$ will receive the light scattered by a particle at the center of the scattering volume in any direction between the $\theta_j \pm \Delta \theta$ directions. For each real refractive index and particle size in the specified ranges, mie.f calculates the Mie intensity functions every 0.1° between $\theta \pm \Delta \theta$ for each detector. The average of these Mie intensity functions are written to an unformatted data file. The name of the file corresponds to the value of the real refractive index, so a new file is created for each value of the real refractive index. Therefore, the library consists of files containing the averaged Mie intensity functions with names such as n133.dat, n157.dat, etc. Mie.f also creates a data file called miecntrl.dat that contains the information used to generate the library. Table C.1 shows the contents of a typical miecntrl.dat file. The lines in miecntrl.dat indicating the scattering angles and the angular resolution of each

110
detector are specific to the 15 channel nephelometer in use in the Laser Applications Laboratory at Argonne National Laboratory.

Table C.1. Typical Miecntrl.dat File

| .855000 | .100000 | 10.1000 | .100000 | 101 | (wavelength in μm) |
| .402716 | .505361 | .609385 | .715142 | 15 | (initial, final, and step size for the particle size range in μm, the number of particles considered) |
| .823034 | .933531 | 1.04720 | 1.16473 | | (number of detectors) |
| 1.28700 | 1.41517 | 1.55079 | 1.69612 | | (scattering angle for each detector in radians) |
| 1.85459 | 2.03197 | 2.23954 | | | |
| 1.867502E-02 | 21 | | | | (angular resolution of detectors 1&2 and the number of calculations in the averaging process) |
| 2.024582E-02 | 23 | | | | (angular resolution of detectors 3-15 and the number of calculations in the averaging process) |
| 1.10 | 2.00 | 0.01 | 91 | | (initial, final, and step size for the real refractive index range, the number of indices considered) |

Datgen.f

Datgen.f calculates the differential scattering cross sections that would be measured by detectors with the scattering angles and angular resolutions listed in miecntrl.dat for a given particle size and a given set of optical properties. Datgen.f can be used to create synthetic light scattering data for blind inversion tests. The particle size and optical properties are randomly selected from within the ranges specified in miecntrl.dat, and the differential scattering cross sections are calculated for a particle with these properties. Gaussian distributed random noise can be added to the calculated values in order to simulated actual measurements. The results of datgen.f are written to a file called decrat.dat. Table C.2 shows the decrat.dat file used in the example inversion. The same format is required for measured data sets that are used as inputs to invert.f.

Datgen.f should be used to calculate the average of the differential scattering cross sections and the differential scattering cross section at the detector closest to 0° for several sizes and real refractive indices with in specified ranges before attempting to invert any data set. A table similar to Table C.3 should be compiled and used as a guide in selecting the trial function as required in the inversion process.
Table C.2. Typical Decrat.dat File

<table>
<thead>
<tr>
<th>Diameter (um)</th>
<th>Size Parameter</th>
<th>Optical Constants</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>1.1 + i 10^{-3}</td>
</tr>
<tr>
<td></td>
<td></td>
<td>C_{sca}^{avg} (um^2)</td>
</tr>
<tr>
<td>0.1</td>
<td>0.4</td>
<td>1.3e-7</td>
</tr>
<tr>
<td>0.5</td>
<td>1.8</td>
<td>1.1e-3</td>
</tr>
<tr>
<td>1.0</td>
<td>3.7</td>
<td>2.4e-2</td>
</tr>
<tr>
<td>2.0</td>
<td>7.3</td>
<td>0.1</td>
</tr>
<tr>
<td>3.0</td>
<td>11.0</td>
<td>0.2</td>
</tr>
<tr>
<td>4.0</td>
<td>14.7</td>
<td>0.6</td>
</tr>
<tr>
<td>5.0</td>
<td>18.4</td>
<td>0.7</td>
</tr>
<tr>
<td>6.0</td>
<td>22.0</td>
<td>1.6</td>
</tr>
<tr>
<td>7.0</td>
<td>25.7</td>
<td>1.4</td>
</tr>
<tr>
<td>8.0</td>
<td>29.4</td>
<td>3.6</td>
</tr>
<tr>
<td>9.0</td>
<td>33.1</td>
<td>2.6</td>
</tr>
<tr>
<td>10.0</td>
<td>36.7</td>
<td>4.8</td>
</tr>
</tbody>
</table>

Table C.3. Average and 23° Angular Scattering Cross Sections for the 15 Channel Nephelometer
Invert.f

The use of invert.f is demonstrated with a step by step example of an inversion process. The data for the example inversion was generated using datgen.f and assumes the use of the 15 channel nephelometer in use at the Laser Applications Laboratory at Argonne National Laboratory.

Invert.f first reads miecntrl.dat to obtain the necessary information regarding the library of Mie intensity functions. The program then reads the file containing the measurements. This file must be titled decrate.dat and must be in the form previously specified. The measurements and corresponding imprecision estimates are converted from cm$^2$ to $\mu$m$^2$, and the user is given the option of plotting the measurement set. The user should make a note of the value of the differential scattering cross section closest to 0°, of the value of the average of the differential scattering cross sections, and of the percent error in the measurements. Figure C.1 is a plot of the example data set. In the example data set, the 23° differential scattering cross section is the closest measurement to 0° and has a value of 15.2 $\mu$m$^2$, the average of the differential scattering cross sections is 6.2 $\mu$m$^2$, and the percent error in the measurements is 10%.

The program then asks the user to specify the real refractive index range by entering the initial and final values and the step size. The maximum number of indices that can be considered at one time is 11. Experience has show that it is best to first consider the entire range of refractive indices specified in miecntrl.dat and to gradually reduce the range and step size. When inverting the example data set, the range was initially set at 1.1-2.0 with a step size of 0.1. The names of the data files from the library are displayed on the screen as they are read into the program.

After the Mie intensity files are read, the user is prompted for the maximum allowable average relative error. Experience has show that a maximum allowable average relative error equal to one tenth of the percent error generally gives good results. The percent error in the example data set is 10%, so the maximum allowable relative error was set to 1. The program then calculates the kernel covariance matrix, finds the eigenvalues and eigenvectors of the kernel covariance matrix, and calculates the average relative error. If the average relative error is greater than the maximum allowable average relative error, the most redundant measurement is removed from the set of inputs, and a new kernel covariance matrix is calculated. In the example inversion, the average relative error is 0.724 which is less than the maximum allowable average relative error, so no measurements were removed from the input set.
Once the set of inputs is determined, the eigenvalues of the final kernel covariance matrix are displayed on the screen. The program then calculates the eigenfunctions, the unconstrained expansion coefficients, and the unconstrained distribution. The unconstrained distribution is used to retrieve a value for the real refractive index. In the example inversion, the retrieved real refractive index was 1.62. The user is then asked whether or not the range of real refractive indices should be changed. Experience has shown that it is best to choose a narrower range that is centered around the retrieved value and repeat the process until the retrieved real refractive index converges to a single value. In the example inversion, the second range of refractive indices selected was 1.37-1.87 with a step size of 0.05. The retrieved value of the refractive index was then 1.62 again. A new range, 1.47-1.77, with a step size of 0.03 was then selected, and the retrieved value was 1.60. This process was repeated until the retrieved value converged to 1.58. Finally, the eigenvalues and eigenfunctions were calculated for a single refractive index by specifying the range of refractive indices to be 1.58-1.58 and setting the step size equal to zero.

After retrieving a value for the real refractive index, the unconstrained expansion coefficients and the eigenfunctions are used to calculated the unconstrained particle size distribution function (PSDF). The unconstrained PSDF is highly oscillatory and assumes negative values, so it is not a physically realistic solution. However, when the measurements correspond to light scattering by a single particle or a narrow distribution of particles, the unconstrained solution provides enough information to successfully select a trial function. Often, the oscillations in the unconstrained PSDF can be reduced by suppressing the highest order eigenfunctions, so the program asks the user to specify the number of eigenfunctions to suppress. Experience has show that the number of suppressed eigenfunctions should be increased until the rms error in unconstrained solution is approximately equal to the expected rms error.

After suppressing the appropriate number of eigenfunctions, examination of the unconstrained PSDF will help in selecting the trial function. Recall that the average of the measurements in the example data set is 6.2 \( \mu m^2 \) and the value of the 23° measurement is 15.2 \( \mu m^2 \). Comparison of these values with the values listed in Table C.1, indicates that the size parameter of the particle is probably somewhere between 22 and 29. Figure C.2 shows the unconstrained PSDF for the example inversion. The most prominent peak in the unconstrained PSDF in the expected range is at a size parameter of 28, so 28 is chosen as the value to use as the peak value in the trial function.
Figure C.2. Unconstrained PSDF with 2 Eigenfunctions Suppressed
After displaying the unconstrained PSDF, the program calculates the supplemental eigenfunctions. The Gram-Schmidt orthogonalization procedure is used to orthogonalize the supplemental eigenfunctions with respect to the natural eigenfunctions. This procedure requires a minute or two to complete. Once the orthogonalization procedure is completed, the number of supplemental eigenfunctions is displayed on the screen. In the example inversion, the number of supplemental eigenfunctions is 86. The user is then asked to input the peak size parameter needed to specify the trial function. A size parameter of 28 is input to the program to continue the example inversion.

The expansion coefficients for the trial function are then calculated, the integral of the trial function is displayed, and the user is given the option of re-normalizing the trial function. The trial function should be re-normalized if the normalization integral is different from one, but this is rarely necessary. The user may then view the trial function. After viewing the trial function the average of the measurements and the normalization constant are displayed on the screen and the user is asked whether or not the selected trial function should be used in the constrained inversion process for the unknown size. The normalization constant should be slightly larger than the average of the measurements, so if the normalization constant is much smaller or larger than the average of the measurements, another trial function should be selected. In general, the normalization constant can be reduced by selecting a small size parameter for the trial function or increased by selecting a larger size parameter for the trial function. In the example inversion, the average of the measurements was 6.2 μm² and the normalization constant had a value of 6.3 μm², so the selected trial function was used in the constrained inversion process.

After determining the trial function, the weighting parameter for the trial function constraint, γ, is determined by minimizing the RRV. The appropriate value of γ is determined by increasing γ from zero until \( \frac{\partial RRV}{\partial \gamma} \) is approximately zero. Experience has show that γ should be increased until \( \frac{\partial RRV}{\partial \gamma} \) is greater than -10^{-4}, but still negative. In the example inversion, γ = 37.45 resulted in \( \frac{\partial RRV}{\partial \gamma} = -1.35 \times 10^{-4} \).

Once the value of gamma is obtained, the constrained expansion coefficients are calculated and the constrained PSDF is calculated and displayed on the screen.

After displaying the constrained PSDF, the program asks the user to window the retrieved size distribution. In order to obtain an estimate of the imaginary part of the refractive index, the program must calculated the scattering pattern for every size parameter at which the constrained PSDF has a non-zero value. The time required for these computations can be greatly reduced if the range of sizes is restricted to a narrow region.
around the peak in the constrained PSDF. The program asks the user to enter a lower and an upper limit for the size range, and a windowing function sets the PSDF equal to zero outside of the selected range. The windowed PSDF is then displayed on the screen.

Next, the rms error of the retrieved solution and the expected rms error are shown on the screen. If the rms error of the retrieved solution is less than or equal to the expected rms error, the constrained PSDF is a possible solution and can be used to estimate the value of the imaginary part of the refractive index. If the rms error of the retrieved solution is much larger than the expected rms error, a new trial function should be selected. In the example inversion, the rms error of the constrained solution was 0.66 and the expected rms error was 0.93, so the PSDF shown in Figure C.3 is a possible solution and can be used to find an estimate of the imaginary part of the refractive index.

The program then prompts the user to guess a value of the imaginary part of the refractive index. The differential scattering cross sections that would be produced by the constrained PSDF and the retrieved real refractive index and the assumed imaginary refractive index are then calculated and displayed on the screen. The program also allows the user to plot and compare the measured and calculated scattering patterns. Different values are assumed for the imaginary part of the refractive index until the rms error of the calculated scattering pattern is approximately equal to the rms of the imprecision estimates. If it is not possible to reconcile the measured and calculated scattering patterns by adjusting the value of the imaginary part of the refractive index, the inversion process should be repeated. As a general rule, if the calculated and measured differential scattering cross sections in the backward hemisphere agree well, only the trial function should be changed. If there is poor agreement in the backward hemisphere, a new value for the real refractive index should be obtained using a different value for the maximum allowable average relative error. However, the disagreement between the measured and calculated scattering patterns generally occurs in the forward hemisphere, and it is rarely necessary to obtain a new value for the refractive index. Another general rule to follow in selecting a new trial function, is that the scattering in the forward angles increases as the size increases. Therefore, the peak size of the trial function should be increased if the calculated differential scattering cross sections are less than the measured differential scattering cross sections in the forward directions and reduced if they are greater. In the example inversion, the rms error of the calculated scattering pattern (0.82) was close to the expected rms error (0.93) when the value of the imaginary part of the refractive index was assumed to be $10^{-5}$. The measured and calculated scattering patterns are compared in Figure C.4.
Figure C.3. Constrained PSDF
Figure C.4. Comparison of the Measured and Calculated Scattering Patterns
The results of the example inversion were presented in Chapter 4, but are repeated here for convenience. The retrieved size and optical properties are compared with the actual values in Table C.2.

Table 2. Comparison of the Particle Size and Optical Properties with the Retrieved Values

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Actual Value</th>
<th>Retrieved Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diameter (µm)</td>
<td>7.6</td>
<td>7.6</td>
</tr>
<tr>
<td>Size Parameter</td>
<td>27.9</td>
<td>28.0</td>
</tr>
<tr>
<td>Real Refractive Index</td>
<td>1.58</td>
<td>1.58</td>
</tr>
<tr>
<td>Imaginary Refractive Index</td>
<td>5.0x10⁻⁴</td>
<td>1.0x10⁻⁵</td>
</tr>
</tbody>
</table>

Inversion Codes

The following are complete listings of each of the programs. Some portions of the code are specific to the nephelometers in use at the Laser Applications Laboratory at Argonne National Laboratory. The plotting routines require the use of SuperPlot on an Apple MacIntosh, so these portions of the code will not work on other systems. The portions of the code that need to be changed in order to accommodate a particular nephelometer or to allow for interactive plotting on another computer system are clearly marked.

Mie.f

```fortran
program mie_library
implicit none
real*4 detct(36), theta(100)
real*1 wavei, pi, diai, dafi, ddia, refi, reff, dref, ipor, iper,
idia, refirefox, refim, dtheta, dtheta1, dtheta3, alpha, alphai, alpha3
integer ndia, nref, ndetct, idetct, ione, item, ihund, j, i, iref, nangle,
1 ntheta, nangle1, nangle3
character fname*25, c1*1, c2*1, c3*1
pi=4.0*atan(1.0)
print*, 'enter the wavelength'
read(*, *)wavei
print*, 'enter the initial, final, and step size for diameter'
```
read(*,*)dia1, diaf, ddia
ndia=nint((diaf-dia1)/ddia)+1

print*, 'enter the initial, final, and step size for n'
read(*,*)refi,reff,dref
if(dref.eq.0.)then
  nref=1
else
  nref=nint((reff-refi)/dref)+1
endif

This portion of the code is specific to the nephelometers used in the Laser Applications Lab at Argonne.
ndetct = number of detectors
detct(idetct) = scattering angles in radians
alpha = angular resolution of the detectors in radians. A detector with a scattering angle of detct(idetct) will receive light from a particle at the center of the scattering volume that is scattered in any direction between the detct(idetct)±alpha directions.
nangle = number of angles used in averaging the Mie intensity functions over the solid angle subtended by each detector. The number of angles is chosen so that the mie intensity functions are calculated every 0.1 degree.

print*, 'enter the number of channels (15 or 36)'
read(*,*)ndetct
if(ndetct.eq.15)then
  do 100 idetct=1,ndetct
     detct(idetct)=2.*asin(float(idetct-1)*0.05+0.2)
   100 continue
  alpha1 = 1.07*pi/180.
  nangle1=(nint(2.*1.07/0.1)/2)*2+1
  dtheta1=2.*alpha1/float(nangle1-1)
  alpha3=1.16*pi/180.
  nangle3=(nint(2.*1.16/0.1)/2)*2+1
  dtheta3=2.*alpha3/float(nangle3-1)
  ntheta=2*nangle1+13*nangle3
elseif(ndetct.eq.36)then
  do 200 idetct=1,ndetct
     detct(idetct) = (20.+float(idetct-1)*4.)*pi/180.
  200 continue
  alpha=0.82*pi/180.
  nangle=(nint(2.*0.82/0.1)/2)*2+1
  dtheta=2.*alpha/float(nangle-1)
  ntheta=ndetct*nangle
endif

open(unit=13, file='miecntrl.dat', status='unknown')
write(13,*)wave
write(13,*)dia1, diaf, ddia, ndia
write(13,*)ndetct
write(13,*)(detct(idetct),idetct=1,ndetct)
Calculating the Mie intensity functions. A file containing
the smoothed intensity functions is created for each real
refractive index.

refim=0.
do 700 iref=1,nref
  refre=float(iref-l)*dref+refi
  ione=ifix(refre+0.0005)
  iten=ifix(amod(refre*10.,10.)+0.0005)
  ihund=nint(amod(refre*100.,10.)+0.0005)
  if(ihund.eq.10)ihund=0
  cl=char(48+ione)
  c2=char(48+iten)
  c3=char(48+ihund)
  fname=' n'//d//c2//c3//'.dat'
  write(*,300)fname
  format(a25)
  open(unit=4,file=fname,form='unformatted',status='unknown')
do 600 i=1,ndia
  dia=float(i-1)*ddia+diai
do 500 idetct=1,ndetct
  if(ndetct.eq.15)then
    if(idetct.eq.1.or.idetct.eq.2)then
      nangle=nangle1
      dtheta=dtheta1
      alpha=alpha1
    else
      nangle=nangle3
      dtheta=dtheta3
      alpha=alpha3
    endif
  endif
  do 400 j=1,nangle
    theta(j)=detct(idetct)-alpha+float(j-1)*dtheta
  continue
  call bhmie(wavel,dia,refre,refim,ipar,iper,theta,nangle)
  write(4)ipar,iper
  continue
  continue
  continue
close(unit=4)

700 continue

stop
end

subroutine bhmie(wavel, dia, refre, refim, ipar, iper, theta, nangle)

dimension amu(100), theta(100), pi(100), tau(100), pi0(100), pi1(100)
real*4 ipar, iper
complex d(3000), y, refrel, xi, x10, x11, an, bn, s1(200), s2(200)
real*8 psi0, psi1, psi, dn, dx

pie=4.0*atan(1.0)
refmed=1.0

do 50 j=1, nangle
    amu(j)=cos(theta(j))
50 continue

refrel=complx(refre, refim)/refmed

x=pie*dia/wavel
dx=x
y=x*refrel
xstop=x+4.*x**.3333+2.
nstop=xstop
ymod=cabs(y)
nmx=amax1(xstop, ymod)+15

d(nmx)=complx(0., 0.)
nn=nmx-1

do 100 n=1, nn
    mn=nmx-n+1
    d(mn-n)=(rn/y)-(1./(d(mn-n+1)+rn/y))
100 continue

do 200 j=1, nangle
    pi0(j)=0.
    pi1(j)=1.
200 continue

nn=2*nangle-1

do 300 j=1, nn
    s1(j)=complx(0., 0.)
    s2(j)=complx(0., 0.)
300 continue

psi0=dcos(dx)
psi1=dain(dx)
chi0=-sin(x)
chi1=cos(x)
apsi0=psi0
apsi1=psi1
x10=complx(apsi0, -chi0)
xi1=complex(apsi1,-chi1)
n=1

400  
dn=n  
nn=n  
fn=(2.*rn+1.)/(rn*(rn+1.))  
ln=(rn*(rn+2.))/(rn+1.)  
psi=(2.*dn-1.)*psi1/dx-psi0  
apsi=psi  
chi=(2.*rn-1.)*chi1/x-chi0  
xi=complex(apsi,-chi)  
an=(dn/refrel+rn/x)*apsi-apsi  
an=an/(dn/refrel+rn/x)*xi-xi1  
bn=(refrel*d(n)+rn/x)*apsi-apsi  
bn=bn/(refrel*d(n)+rn/x)*xi-xi1

      do 500 j=1,nangle
         jj=2*nangle-j  
         pl(j)=pl1(j)  
         tau(j)=rn*pi(j)*amu(j)-(rn+1.)*pi0(j)  
         p=(-1.)**(n-1)  
         s1(j)=s1(j)+fn*(an*pi(j)+bn*tau(j))  
         t=(-1.)**n  
         s2(j)=s2(j)+fn*(an*tau(j)+bn*pi(j))  
         if(j.ne.jj) then  
            s1(jj)=s1(jj)+fn*(an*pi(j)*p+bn*tau(j)*t)  
            s2(jj)=s2(jj)+fn*(an*tau(j)*t+bn*pi(j)*p)  
         endif
      500 continue

      psi0=psi  
      psi1=psi  
      apsi1=apsi  
      chi0=chi  
      chi1=chi  
      xi1=complex(apsi1,-chi1)
      n=n+1  
nn=n

      do 600 j=1,nangle
         pl1(j)=((2.*rn-1.)/(rn-1.))*pi(j)*amu(j)  
         pl1(j)=pi1(j)-rn*pi0(j)/(rn-1.)  
         pi0(j)=pi(j)
      600 continue

      if (n-1-nstop) 400,700,700

700 continue

      ipar=0.0  
      iper=0.0
      do 800 j=1,nangle
         ipar=cabs(s2(j))**2+ipar  
         iper=cabs(s1(j))**2+iper
      800 continue

      ipar=ipar/float(nangle)  
      iper=iper/float(nangle)
return
end

Datgen.f

This program generates synthetic light scattering in the form
needed to test the inversion programs

program datgen

implicit none

real*4 g(36),dg(36),sd(36),detct(36),theta(100)
real*4 pi,wavel,diai,diaf,ddia,refi,reff,refs,dias,iper,
1dsizep,gsum,pererr,sec,n0,refims,seends,ran,alpa,alpa1,alpa3,
2waven,dtheta,dtheta1,dtheta3,ipar

integer ndia,ndetct,nref,j,iseed,1,1s,rs,nangle,nangle1,nangle3,
idetct

character entyn*1,yn*1,tab*1

pi=4.*atan(1.0)
tab=char(9)

Open and read file created by mie.f that contains information
regarding the library of mie intensity functions. The open
statement must be modified so the program can find the file
'miecntrl.dat'.
wavel= wavelength of the incident beam in μm
diai= initial particle diameter in μm
diaf= final particle diameter in μm
ddia= step size in μm
ndia= number of sizes in the library
ndetct= number of detectors
detct(idetct)= scattering angle of the detectors in radians
alpa= angular resolution for the detectors
nangle= number of angle used in integrating over the solid angle
subtended by the detectors
Alpha and nangle depend on the specific nephelometer used. The
code is currently set up for the nephelometers used in the Laser
Applications Lab at Argonne.
refi= initial real refractive index
reff= final real refractive index
dref= step size
nref= number of real refractive indices in the library

open(unit=13, file="Macintosh HD:MPU:miedata:miecntrl.dat", status=
1'old')
read(13,*),wavel
read(13,*),diai,diaf,ddia,ndia
read(13,*),ndetct
read(13,*),detct(idetct),ndetct=1,ndetct

Nephelometer specific code.
These lines should be modified according to the number of detectors available and the angular resolution of the detectors. The number of angles, nangle, is set so that Mie intensity functions are calculated every 0.1 degree.

if(ndetct.eq.36) then
   read(13,*)alpha, nangle
   dtheta=2.*alpha/float(nangle-1)
elseif(ndetct.eq.15) then
   read(13,*)alpha1, nangle1
   read(13,*)alpha3, nangle3
   dtheta1=2.*alpha1/float(nangle1-1)
   dtheta3=2.*alpha3/float(nangle3-1)
endif

read(13,*)refi, reff, dref, nref
write(*,100)
write(*,200)wave
write(*,300)diai, diaf, ddia, ndia
write(*,400)ndetct
write(*,500)refi, reff, dref, nref
if(ndetct.eq.36) then
   write(*,600)alpha*180./pi, nangle
elseif(ndetct.eq.15) then
   write(*,700)alpha1, nangle1
   write(*,800)alpha3, nangle3
endif

format(2x,'scattering data available for these conditions')
format(2x,'wavelength(microns)=',f7.4)
format(2x,'diameter(microns)=',f10.4, ' to ', f10.4, '/', f6.4, ' step size ', f6.4, ' total number of sizes=',i4)
format(2x,'number of angles=',i3)
format(2x,'real part of the refractive index=',f4.2, ', step size=',f6.4, ' total number of indices=',i3)
format(2x,'angular resolution of each detector=','e12.4,/', e12.4, ' kernels are smoothed over ','i3,' angles')
format(2x,'angular resolution of channels 1&2=','e12.4,/', e12.4, ' kernels are smoothed over ','i3,' angles')
format(2x,'angular resolution of channels 3-15=','e12.4,/', e12.4, ' kernels are smoothed over ','i3,' angles')

sec=secnds(28800.)
iseed=nint(sec*100.)
iseed=ifix(float(iseed)/2.)*2+1
wave=(2.*pi/wave)*1.0e4
dsiz=pi*dia/wave

print*, 'do you want to enter the particle size and ref. index?'
read(*,900)entyn
if(entyn.eq.'y') then
   print*, 'enter the particle size'
   read(*,*)dias
   print*, 'enter the real part of the refractive index(n)'
   read(*,*)refs
   print*, 'enter the imaginary part of the refractive index(k)'
   read(*,*)refims
else
    print*, 'use only library values (y/n)?'
    read(*, 900) yn
    dias = dials + ran(iseed) * (dial - dials)
    refs = rfei + ran(iseed) * (reff - refi)
    refims = ran(iseed) * 0.001
    if (yn.eq.'y') then
      is = nint((dias - dials)/ (dial - dials)) + 1
      dias = dials + float(is - 1) * dia
      if (nref.gt.1) then
        rs = nint((refs - refi)/ dref) + 1
      else
        rs = 1
      endif
      refs = refi + float(rs - 1) * dref
      refims = 0.
    endif
    endif
    print*, 'enter the amount of error (%)'
    read(*, *) pererr
    pererr = pererr/100.
900 format(a1)
c
Calculate the error free differential scattering cross sections
in units of cm2
c
do 1400 idetct = 1, ndetct
  g(idetct) = 0.
  dg(idetct) = 0.
  if (ndetct.eq.15) then
    if (idetct .eq. 1 .or. idetct .eq. 2) then
      nangle = nangle1
      dtheta = dtheta1
      alpha = alpha1
    else
      nangle = nangle3
      dtheta = dtheta3
      alpha = alpha3
    endif
  endif
  do 1200 j = 1, nangle
    theta(j) = idetct(idetct) - alpha + float(j - 1) * dtheta
  1200 continue
  call bhmie(wavel, dias, refs, refims, ipar, iper, theta, nangle)
  g(idetct) = (ipar + iper)/2./waven**2
1400 continue

c Add gaussian distributed random noise to the error free
c differential scattering cross sections
The algorithm for generating the noise is described in
"Statistical Distributions" by Hastings and Peacock

c
128
gs\text{sum}=0.
da 1600 \quad j=1,ndetct
\quad \text{if}(\text{pererr} \lt 0.0001)\text{then}
\quad \quad \text{sd}(j)=1.0
\quad \text{else}
\quad \quad \text{sd}(j)=\text{pererr} \times \text{g}(j)
\quad \quad \text{do} 1500 \quad i=1,12
\quad \quad \quad \text{dg}(j)=\text{dg}(j)+\text{ran}(\text{i seed}) \times \text{sd}(j)
\quad \quad \quad \text{continue}
\quad \quad \quad \text{dg}(j)=(\text{dg}(j)-6. \times \text{sd}(j))
\quad \quad \quad \text{g}(j)=\text{g}(j)+\text{dg}(j)
\quad \quad \text{endif}
\quad \text{gsum} = \text{gsum} + \text{g}(j)
1600 \text{continue}
\text{n0} = \text{gsum} / \text{float}(\text{ndetct})
c
c Write the differential scattering cross and the corresponding
c imprecision estimates to a file titled 'decrat.dat'
c
open(unit=15, file='decrat.dat', status='unknown')
write(15,*)\text{dias}, \text{dias} \times \text{pi/wavel}, \text{refs}, \text{refim3}, \text{pererr} \times 100.
write(15,*)\text{n0}
\text{do} 1700 \quad j=1,ndetct
\quad \text{write}(15,1800)\text{g}(j), \text{tab}, \text{sd}(j)
1700 \text{continue}
1800 \text{format}(g10.4, a1, g10.4)
close(unit=15)
stop
c end
c This subroutine uses the mie code written by Bohren and Huffman
c to average the Mie intensity functions over the solid angle
subtended by each detector
c
\begin{verbatim}
 subroutine bhmie(wavel,dia,refre,refim,ipar,iper,theta,nangle)
 dimensional anu(100),theta(100),pi(100),tau(100),pi0(100),pi1(100)
 real*4 ipar,iper
 complex d(3000),y,refrel,xi,xi0,xi1,an,bn,s1(200),s2(200)
 real*8 psi0,psi1,psi,dn,dx

 pie=4.0*atan(1.0)
 re\text{fmed}=1.0

 do 50 \quad j=1,nangle
 \quad anu(j)=cos(\text{theta}(j))
50 \text{continue}

 refrel=\text{cmplx}(\text{refre},\text{refim})/\text{re\text{fmed}}
x=\text{pie} \times \text{dia/wavel}
\end{verbatim}
dx=x
y=x*refrel
xstop=x+4.*x**.3333+2.
nstop=xstop
ymod=cabs(y)
nmax=anaxl(xstop,ymod)+15
d(nmx)=cmplx(0.,0.)
nn=nmax-1
do 100 n=1,nn
   nn=nmx-n+1
   d(nmx-n)=(rn/y)-(1./(d(nmx-n)+rn/y))
100 continue

   do 200 j=1,nangle
      pi0(j)=0.
      pi1(j)=1.
200 continue

   nn=2*nangle-1
   do 300 j=1,nn
      a1(j)=cmplx(0.,0.)
      a2(j)=cmplx(0.,0.)
300 continue

   psi0=dcos(dx)
   psi1=dsin(dx)
   chi0=-sin(x)
   chi1=cos(x)
   apsi0=psi0
   apsi1=psi1
   xi0=cmplx(apsi0,-chi0)
   xi1=cmplx(apsi1,-chi1)

   n=1
   do 400 n=1
      nn=nn
      fn=(2.*rn+1.)/(rn*(rn+1.))
      gn=(rn*(rn+2.))/(rn+1.)
      psi=(2.*dn-1.)*psi1/dx-psi0
      apsi=psi
      chi=(2.*rn-1.)*chi1/x-chi0
      xi=cmplx(apsi,-chi)
      an=(d(n)/refrel+rn/x)*apsi-apsi1
      an=an/((d(n)/refrel+rn/x)*xi-xi1)
      bn=(refrel*d(n)+rn/x)*apsi-apsi1
      bn=bn/((refrel*d(n)+rn/x)*xi-xi1)
   400 continue

   do 500 j=1,nangle
      jj=2*nangle-j
      pi(j)=pi1(j)
      tau(j)=rn*pi(j)*amu(j)-(rn+1.)*pi0(j)
      p=(-1.)**(n-1)
      s1(j)=s1(j)+fn*(an*pi(j)+bn*tau(j))
      t=(-1.)**n
      s2(j)=s2(j)+fn*(an*tau(j)+bn*pi(j))
500 continue
if(j.ne.jj) then
   s1(jj)=s1(jj)+fn*(an*pi(j)*p+bn*tau(j)*t)
   s2(jj)=s2(jj)+fn*(an*tau(j)*t+bn*pi(j)*p)
endif

500 continue

psi0=psi1
psi1=psi
apsi1=psii1
chi0=chii1
chii1=chii
xi1=cmplx(apsi1,-chii1)
n=n+1
rn=n

do 600 j=1,nangle
   pi1(j)=((2.*rn-1.)/(rn-1.))*pi(j)*amu(j)
   pi1(j)=pi1(j)-rn*pi0(j)/(rn-1.)
   pi0(j)=pi(j)
600 continue

if (n-1-nstop) 400,700,700
700 continue

ipar=0.0
iper=0.0

do 800 j=1,nangle
   ipar=cabs(s2(j))**2+ipar
   iper=cabs(s1(j))**2+iper
800 continue

ipar=ipar/float(nangle)
iper=iper/float(nangle)

return
end

Invert.f

This program inverts single particle light scattering data for
the particle size and optical properties.

Inputs are measured differential scattering cross sections(cm2)
and corresponding imprecision estimates. These inputs should be
placed in a file entitled 'decrat.dat' in the following form:
1 line header
average of the differential scattering cross sections
ith differential scattering cross section, ith imprecision
estimate

A library of Mie intensity functions must also be created using
mie.f before running invert.f

program invert
implicit none
Read the parameters for the library of Mie intensity functions

```fortran
    pi=4*atan(1.)
    tab=char(9)

    read(11,*)waveI
    read(11,*)diai,diaf,ddia,ndia
    read(11,*)ndetct
    read(11,*)detct(idetct),idetct=1,ndetct)
    if(ndetct.eq.36)then
        read(11,*)alpha,nangle
        dtheta=2.*alpha/float(nangle-1)
    elseif(ndetct.eq.15)then
        read(11,*)alpha1,nangle1
        read(11,*)alpha3,nangle3
        dtheta1=2.*alpha1/float(nangle1-1)
        dtheta3=2.*alpha3/float(nangle3-1)
    endif
    read(11,*)refi,reff,dref,nref
    close(unit=11)

    open(unit=12,file='inversion.out',status='unknown')
    write(*,100)
    write(*,200)waveI
    write(*,300)diai,diaf,ddia,ndia
    write(*,400)ndetct
    write(*,500)refi,reff,dref,nref
    write(12,100)
    write(12,200)waveI
    write(12,300)diai,diaf,ddia,ndia
    write(12,400)ndetct
    write(12,500)refi,reff,dref,nref
    if(ndetct.eq.36)then
        write(*,600)alpha*180./pi,nangle
        write(12,600)alpha*180./pi,nangle
    elseif(ndetct.eq.15)then
        write(*,700)alpha1,nangle1
        write(*,800)alpha3,nangle3
```

---

```
real*8 evale(36),evector(36,36),efunct(36,101,11),
supef(101,101,11)
real*4 g(36),dg(36),dia(101),x(101),kernel(36,101,11),ft(101,11),
lap(36),at(137),ac(137),f(101,11),optk(36,101,11),input(36),
2detct(36),dinput(36),fplot(101),dfplot(101)
real*4 waveI,diai,diaf,ddia,refi,reff,dref,pi,waven,experr,dx,
lgmean,gamma,refs,junk,alpha,norm,cnorm,error,referr,dtheta,
2dtheta1,dtheta3,alpha1,alpah3,rserr,auginpt,pererr
integer ndia,ndetct,nref,ninput,j,i,nsupef,rs,nout,nangle,r,nsup,
idetct,nangle1,nangle3,k
character yn*1,tab*1
```
write(12,700)alpha1,nangle1
write(12,800)alpha3,nangle3
endif

100 format(2x,'scattering data available for these conditions')
200 format(2x,'wavelength(µm)=',f7.4)
300 format(2x,'diameter(µm)=',f10.4,' to ',f10.4,/,step size 1=',f6.4,','total number of sizes=',i4)
400 format(2x,'number of measurements=',i13)
500 format(2x,'real part of the refractive index=',f4.2,' to ',f4.2,1/,step size=',f6.4,'total number of indices=',i13)
600 format(2x,'angular resolution of each detector=',e12.4,/,kernels are smoothed over ',i13,'angles')
700 format(2x,'angular resolution of channels 1&2=',e12.4,/,kernels are smoothed over ',i13,'angles')
800 format(2x,'angular resolution of channels 3-15=',e12.4,/,kernels are smoothed over ',i13,'angles')

pause

waven=2.*pi/wavel

print*,'reading the scattering measurements'
ninput=ndetct

c Read the measured differential scattering cross sections
The units are changed from cm2 to µm2

c
open(unit=13,file='decrat.dat',status='old')
read(13,*)junk
read(13,*)gmean

gmean=gmean*1.e8

write(12,*)'differential scattering cross sections (µm2)'
print*,'differential scattering cross sections (µm2)'
experr=0.
pererr=0.
do 900 j=1,ndetct
    read(13,*)g(j),dg(j)
    g(j)=g(j)*1.e8
    dg(j)=dg(j)*1.e8
    pererr=pererr+dg(j)/g(j)+pererr
    experr=experr+(dg(j))**2
    print*,g(j),dg(j)
    write(12,*)g(j),dg(j)
    if(amod(float(j),30.).eq.0.0.or.j.eq.ndetct)pause
900 continue

close(unit=13)

experr=sqrt(experr/float(ndetct))
pererr=100.*pererr/float(ndetct)
write(12,*),'rms error = ',experr
print*,'rms error = ',experr
write(12,*),'percent error = ',pererr
print*,'percent error = ',pererr

do 1000 j=1,ndetct
    input(j)=g(j)/dg(j)/gmean
    dinput(j)=dg(j)
    detct(j)=180.*detct(j)/pi
1000 continue
continue

print*, 'average angular scattering cross section = ', gmean
write(12,*) 'average angular scattering cross section = ', gmean
print*, 'plot the scattering pattern? (y/n)'
read(*,1100) yn

ndetct=number of points to plot
detct*array containing the scattering angles
g*array containing the measured differential scattering cross sections
if(yn.eq.'y') call plotg(ndetct,detct,g,g)

dx=ddia*pi/wavel
do 1200 i=1,ndia
   dia(i)=float(i-1)*ddia+diai
   x(i)=dia(i)*pi/wavel
1200 continue

cnorm=gmean

c Calculate the eigenvalues and eigenvectors
c
1300 call eigen(evalue,evector,efunct,kernel,ninput,ndia,nref,dx,dref,
   crefi,reff,g,dg,wavel,ndetct,optk,input,dinput,nout,gmean)
c Find the real part of the refractive index
c
print*, 'retrieving the real part of the refractive index'
call findn(evalue,evector,efunct,ap,ninput,ndia,pi,nref,refi,reff,f,
   crefi,reff,g,dg,wavel,input,nout,cnorm)
print*, 'current refractive index range', crefi,'to',reff
print*, 'retrieved refractive index =', refs
print*, 'change the range of refractive indices (y/n)?'
read(*,1400) yn
1400 format(a1)
if(yn.eq.'y')then
do 1500 j=1,ninput
   input(j)=g(j)/dg(j)/gmean
   dinput(j)=dg(j)
1500 continue
goto 1300
dendif

norm=0.
do 1700 i=1,ndia
fplot(i)=0.
do 1600 r=1,nref
   fplot(i)=fplot(i)+f(l,r)
end do
1600 continue
   if(nref.ne.1)fplot(i)=fplot(i)*dref
   norm=norm+fplot(i)
end if
1700 continue
   norm=norm*dx
   print*, 'return to view the unconstrained size distribution'
   pause
nsup=0
1800 continue
ccc

This portion of code requires the use of Superplot on an
Apple Macintosh
ndia=number of points to plot
x=size parameter array
fplot=array containing the unconstrained PSDF

call plot(ndia,x,fplot,fplot)

ccc

Calculating the rms residual error of the unconstrained solution
Often it is helpful to suppress some of the highest order eigen-
functions and smooth out some of the oscillations in the
unconstrained solution. The number of suppressed eigenfunctions
should be increased until the rms error of the unconstrained
solution is approximately equal to the expected rms error

rmserr=0.
do 1820 j=1,ninput-nout-nsup
   error=0.
do 1810 k=1,ninput-nout-nsup
      error=error+dsqrt(evalue(k))*evector(j,k)*dble(ap(k))
end do
1810 continue
   rmserr=rmserr+((input(j)-error/cnorm)*dinput(j)*gmean)**2
end do
1820 continue
   rmserr=sqrt(rmserr/float(ninput-nout-nsup))
yn='n'
print*, 'rms error of the unconstrained solution = ',rmserr
print*, 'expected rms error = ',experr
print*, 'suppress the higher order eigenfunctions? (y/n)'
print*, 'number of eigenfunctions = ',ninput-nout
read(*,1400)yn
if(yn.eq.'y') then
   print*, 'enter the number of eigenfunctions to suppress'
   read(*,*)nsup
   if(nsup.ge.ninput-nout) then
      nsup=ninput-nout-1
   end if
end if

135
print*,nsup,' is the maximum number of eigenfunctions that'  
print*, 'can be suppressed'  
print*, 'return to continue'
pause
endif

do 2100 i=1,ndia
   do 2000 r=1,nref
      f(i,r)=0.
      do 1900 j=1,ninput-nout-nsup
         f(i,r)=ap(j)*efunct(j,i,r)+f(i,r)
      1900 continue
   2000 continue
2100 continue
norm=0.
   do 2300 i =1,ndia
      fplot(i)=0.
      do 2200 r=1,nref
         fplot(i)=fplot(i)+f(i,r)
      2200 continue
   if(nref.ne.1)fplot(i) = fplot(i)*dref
   norm=norm+fplot(i)
2300 continue
norm=norm*dx
goto 1800
endif
write(12,*)'unconstrained PSDF'
   do 2400 i=1,ndia
      write(12,2500)x(i),tab,fplot(i)
2400 continue
2500 format(g12.4,a1,g12.4)
print*, 'normalization integral of the unconstrained solution =', 
   lnorm
   write(12,*)'normalization integral of the unconstrained solution 
1 =',norm

Calculating the supplemental eigenfunctions

call supefcal(efunct,supef,rs,ndia,dx,dref,ninput,nout,nref, 
   insupef)
2600 continue

auginpt=0.
   do 2700 j=1,ninput-nout
      auginpt=auginpt+input(j)*dinput(j)*gmean
      ap(j)=ap(j)/cnorm
2700 continue
auginpt=auginpt/float(ninput-nout)
2750 continue

Finding the expansion coefficients for the trial function

call findat(at,x,dx,ndia,rs,efunct,supef,nsupef,pi,wavel,dref,
Calculating the normalization constant. The normalization constant should be slightly larger than the mean of the inputs.

cal1 cnrncal(optk,dinput,ft,cnorm,dx,ndia,ra,ninput,nout,nref,ldref)

print*,'average of the measurements =',avginpt
print*,'normalization constant =',cnorm
write(12,*),'normalization constant =',cnorm
write(12,*),'average of the measurements =',avginpt
print*,'change the trial function?(y/n)?'
read(*,1400)yn
if(yn.eq.'y')goto2750
do 2800 j=1,ninput-nout
   ap(j)=ap(j)*cnorm
2800 continue

Calculating the weighting parameter for the trial function

print*,'finding gamma'
call rrvcal(evalue,ac,at,gamma,ninput-nout,cnorm)

Calculating the constrained solution

print*,'calculating the constrained solution'
call findf(efunct,superf,ap,at,ac,refs,gamma,ndia,ninput,nout,nref,1nsuperf,rs,norm,dx,dref,f,evalue,x,fplot,dfplot,dinput,evector,2cnorm,optk,error,input,gmean,referr)

print*,'rms error of the retrieved solution',error
print*,'expected rms error',experr
write(12,*),'rms error of the retrieved solution',error
write(12,*),'expected rms error',experr

write(12,*),' particle size distribution function'
do 2900 i=1,ndia
   write(12,3000)x(i),tab,fplot(i),tab,dfplot(i)
2900 continue
3000 format(2x,g10.3,2(a1,g10.3))
write(12,3100)refs,referr
write(12,3300)norm
write(*,3100)refs,referr
write(*,3300)norm
3100 format(2x,'refractive index=',g10.3,' +/- ',f5.2)
3300 format(2x,'normalization integral = ',g10.4)

yn='n'
print*,'change the trial function (y/n)?'
read(*,1400)yn

yn='n'
print*,'change the trial function (y/n)?'
read(*,1400)yn
If(yn.eq.'y')goto 2600
print*, 'change the range of refractive indices (y/n)?'
read(*,1400)yn
if(yn.eq.'y')then
   do 3500 j=1,ninput
      input(j)=g(j)/gmean/dg(j)
      dinput(j)=dg(j)
   continue
   goto 1300
endi f
Finding an estimate of the imaginary part of the refractive index
by comparing the measured and calculated scattering patterns

   call findk(fplot,g,dg,gmean,refs,detct,ndetct,alpha,nangle,wavel,
      india,dx,dia,experr,waven,alpaha1,alpaha3,nangle1,nangle3,dtheta,
      2dtheta1,dtheta3)
   yn='n'
print*, 'change the trial function (y/n)?'
read(*,1400)yn
if(yn.eq.'y')goto 2600
print*, 'change the range of refractive indices (y/n)?'
read(*,1400)yn
if(yn.eq.'y')then
   do 3600 j=1,ninput
      input(j)=g(j)/gmean/dg(j)
      dinput(j)=dg(j)
   continue
   goto 1300
endif
close(unit=12)
stop
end
subroutine eigen(evalue,evector,efunct,kernel,ninput,ndia,nref,dx,
   l1dref,refi,reff,g,dg,waven,ndetct,optk,input,dinput,nout,gmean)
    implicit none
real*8 evalue(36),evector(36,36),efunct(36,101,11),kconv(36,36)
real*4 g(36),dg(36),kernel(36,101,11),optk(36,101,11),input(36),
   l1dinput(36)
real*4 refi,reff,dref,dx,ref,waven,nmax,sumlam,invlam,magimp,magg,
   linverr,relac,sumj,sumk,gmean,ipar,iper
Remove these comments to verify the orthogonality of the eigen-
 vectors
real*4 uiuj,uiuk
integer kk
integer ninput, ndia, nref, l, j, r, k, lbound, l, i, kmax, nout, l

character c1*, c2*, c3*, fname*25

print*, 'enter the initial and final refractive index'
read(*, *) refi, reff
print*, 'enter the step size for n'
read(*, *) dref
if(dref.lt.0.00001) then
  nref=1
else
  nref=nint((reff-refi)/dref)+1
endif
write(*, 100)
write(12, 200) refi, reff, dref, nref
100 format('calculating eigenfunctions for the following range of refractive indices')
200 format(2x,'real part of the refractive index=', f4.2, ' to ', f4.2, '1/,' step size=', f4.2, ' total number of real indices=', i3)

c Reading the library of Mie intensity functions

c print*, 'reading the scattering kernels'
do 600 r=1, nref
  ref=float(r-1)*dref+refi
  ref=float(nint(ref*100.))/100.
  l=ifix(ref+0.0005)
  iten=ifix(amod(ref*10.,10.)+0.0005)
  lbound=nint(amod(ref*100.,10.)+0.0005)
  if(lbound.eq.10) lbound=0
  c1=char(48+l)
  c2=char(48+l)
  c3=char(48+l)
  fname='n'//c1//c2//ce3//'.dat'
write(*, 300) fname
300 format(2x,a25)
onopen(unit=15, file="Macintosh HD:MPU:miedata:"//fname, status=1 'old', form='unformatted')
do 500 i=1, ndia
  do 400 j=1, ndetct
    read(15) ipar, iper
    kernel(j, i, r)=(iper+ipar)/2./waven**2/dg(j)
    optk(j, i, r)=kernel(j, i, r)
 400   continue
500   continue
600   continue

c The average relative error was derived by Twomey (Appl. Opt. 13,
942 (1974). I found that a maximum allowable average relative error equal to one tenth of the percent error of the measurements worked best.

print*, 'enter the maximum allowable average relative error'
read(*,*)relec
nout=0
max=0.
print*, 'calculating the kernel covariance matrix'
do 1100 j=1,ninput-nout
do 1000 k=1,j
evector(j,k)=0.
do 900 r=1,nref
do 800 i=1,ndia
   evector(j,k)=evector(j,k)+dble(optk(j,i,r)*optk(k,i,r))
800 continue
900 continue
   if(nref.eq.1)then
      evector(j,k)=evector(j,k)*dble(dx)
   else
      evector(j,k)=evector(j,k)*dble(dx*dref)
   endif
   evector(k,j)=evector(j,k)
   kcov(j,k)=evector(j,k)
   kcov(k,j)=evector(j,k)
   if(k.ne.j.and.evector(k,j).gt.nmax)then
      nmax=evector(k,j)
jmax=j
kmax=k
   endif
1000 continue
1100 continue
print*, 'finding the eigenvalues and eigenvectors'
call tred2(evalue,evector,ninput-nout)
sumlam=0.
invalam=0.
do 1200 j=1,ninput-nout
   magimp=magimp+input(j)**2
   magg=magg+(input(j)*input(j)*gmean)**2
   sumlam=abs(evalue(j))+sumlam
   invalam=1./abs(evalue(j))+invalam
1200 continue
inverr=sqrt(magimp*invalam*sumlam/magg)/float(ninput-nout)
print*, 'average relative error = ',inverr
write(12,*)'average relative error = ',inverr
if(inverr.gt.relec)then
   nout=nout+1
sumj=0
sumk=0
do 1300 j=1,ninput-nout
   if(j.ne.jmax)sumj=kcov(j,jmax)+sumj
   if(j.ne.kmax)sumk=kcov(j,kmax)+sumk
1300 continue

140
if(sumj.gt.sumk)then
  jout=jmax
else
  jout=kmax
endif

do 1600 j=jout,ninput-nout
  do 1500 i=1,ndia
    do 1400 r=1,nref
      optk(j,i,r)=optk(j+1,i,r)
    continue
  continue
  input(j)=input(j+1)
  dinput(j)=dinput(j+1)
1600 continue

1700 write(12,*)'eigenvalues'
   print*, 'eigenvalues'
   do 1700 j=1,ninput-nout
     write(12,*)evalue(j)
     print*,evaluate(j)
   continue

1800 print*, 'calculating the eigenfunctions'
   do 2400 r=1,nref
     do 2300 i=1,ndia
       do 2200 j=1,ninput-nout
         efunct(j,i,r)=0.0
         do 2100 k=1,ninput-nout
           efunct(j,i,r)=efunct(j,i,r)*evector(k,j)*
                       dble(optk(k,i,r))
         continue
       continue
     continue
2400 continue
   efunct(j,i,r)=efunct(j,i,r)/dsqrt(evalue(j))
SUBROUTINE TRED2(lambda,n,number)
implicit none
real*8 lambda(36),n(36,36),offdia(36)
real*8 h,scale,f,g,hh
integer i,j,k,l,number

This routine is taken from Section 11.2 in Numerical Recipes by
Press et al.
Householder reduction of a real symmetric matrix n, stored in a
number by number array. On output, n is replaced by the
orthogonal matrix effecting the transformation. lambda returns
the diagonal elements of the tridiagonal matrix, and offdia the
off diagonal elements, with offdia(1)=0.

IF(number.GT.1)THEN
  DO 10 l=number,2,-1
    L=L-1
    H=0.
    SCALE=0.
    IF(L.GT.1)THEN
      DO 11 K=1,L
        SCALE=SCALE+ABS(n(l,K))
      11 CONTINUE
      IF(SCALE.EQ.0.)THEN
        offdia(l)=n(l,L)
      ELSE
        DO 12 K=1,L
          n(l,K)=n(l,K)/SCALE
          H=H+n(l,K)**2
        12 CONTINUE
        F=n(l,L)
        G=-SIGN(SQRT(H),F)
        offdia(l)=SCALE*G
        H=H-F*G
        n(l,L)=F-G
        F=0.
        DO 15 J=1,L
          n(J,l)=n(l,J)/H
          G=0.
          DO 13 K=1,J
            G=G+n(J,K)*n(l,K)
          13 CONTINUE
          IF(L.GT.J)THEN
            DO 14 K=J+1,L
              G=G+n(K,J)*n(l,K)
            14 CONTINUE
          ENDIF
          offdia(J)=G/H
          F=F+offdia(J)*n(l,J)
        15 CONTINUE
        HH=F/(H+H)
        DO 17 J=1,L
          n(J,L)=n(L,J)/HH
          G=0.
          DO 16 K=1,L
            G=G+n(K,L)*n(J,K)
          16 CONTINUE
          IF(J.GT.L)THEN
            DO 18 K=J+1,L
              G=G+n(K,J)*n(J,K)
            18 CONTINUE
          ENDIF
          offdia(J)=G/H
          F=F+offdia(J)*n(J,J)
        17 CONTINUE
      ENDIF
    10 CONTINUE
  ELSE
    DO 8 L=1,number
      H=0.
      SCALE=0.
      DO 9 K=1,L
        SCALE=SCALE+ABS(n(l,K))
      9 CONTINUE
      IF(SCALE.EQ.0.)THEN
        offdia(l)=n(l,L)
      ELSE
        DO 12 K=1,L
          n(l,K)=n(l,K)/SCALE
          H=H+n(l,K)**2
        12 CONTINUE
        F=n(l,L)
        G=-SIGN(SQRT(H),F)
        offdia(l)=SCALE*G
        H=H-F*G
        n(l,L)=F-G
        F=0.
        DO 15 J=1,L
          n(J,l)=n(l,J)/H
          G=0.
          DO 13 K=1,J
            G=G+n(J,K)*n(l,K)
          13 CONTINUE
          IF(L.GT.J)THEN
            DO 14 K=J+1,L
              G=G+n(K,J)*n(J,K)
            14 CONTINUE
          ENDIF
          offdia(J)=G/H
          F=F+offdia(J)*n(J,J)
        15 CONTINUE
        HH=F/(H+H)
        DO 17 J=1,L
          n(J,L)=n(L,J)/HH
          G=0.
          DO 16 K=1,L
            G=G+n(K,L)*n(J,K)
          16 CONTINUE
          IF(J.GT.L)THEN
            DO 18 K=J+1,L
              G=G+n(K,J)*n(J,K)
            18 CONTINUE
          ENDIF
          offdia(J)=G/H
          F=F+offdia(J)*n(J,J)
        17 CONTINUE
      ENDIF
    8 CONTINUE
  ENDIF
END
F = n(1,J)  
G = offdia(J) - HH*F  
offdia(J) = G  
DO 16 K = 1, J  
n(J,K) = n(K,J) - F*offdia(K) - G*n(1,K)  
16 CONTINUE  
17 CONTINUE  
ENDIF  
ELSE  
offdia(1) = n(1,L)  
ENDIF  
lambda(1) = H  
18 CONTINUE  
ENDIF  
lambda(1) = 0.  
offdia(1) = 0.  
DO 23 I = 1, number  
L = I - 1  
IF(lambda(I).NE.0.) THEN  
DO 21 J = 1, L  
G = 0.  
DO 19 K = 1, L  
G = G + n(I,K)*n(K,J)  
19 CONTINUE  
DO 20 K = M  
n(K,J) = n(K,J) - G*n(K,I)  
20 CONTINUE  
21 CONTINUE  
ENDIF  
lambda(I) = n(I,1)  
n(I,1) = 1.  
IF(L.GE.1) THEN  
DO 22 J = 1, L  
n(I,J) = 0.  
n(J,1) = 0.  
22 CONTINUE  
ENDIF  
23 CONTINUE  
call tqli(offdia, lambda, n, number)  
RETURN  
END  

SUBROUTINE TQLI(offdia, lambda, n, number)  
imPLICIT none  
real*8 lambda(36), n(36,36), offdia(36)  
real*8 f, g, rr, s, c, p, b, dd  
integer i, k, l, mm, iter, number  

c This routine was taken from Section 11.3 of Numerical Recipes by  
c Press et al.  
c QL algorithm with implicit shifts, to determine the eigenvalues  
c and eigenvectors of a real symmetric, tridiagonal matrix, or of a  
c real, symmetric matrix previously reduced by subroutine  
c TRED2(Sec.11.2).  

143
lambda is a vector of length number. On input, its first number
elements are the diagonal elements of the tridiagonal matrix. On
output, it returns the eigenvalues. The vector offdia inputs the
subdiagonal elements of the tridiagonal matrix, with offdia(1)
arbitrary. On output E is destroyed. The kth column of n returns
the normalized eigenvector corresponding to kth eigenvalue.

IF (number.GT.1) THEN
  DO 11 l=2,number
     offdia(l-1)=offdia(l)
  11 CONTINUE
  offdia(number)=0.
  DO 15 l=1,number
     ITER=0
     DO 12 mm=L,number-1
        DD=ABS(lambda(mm))+ABS(lambda(mm+1))
        IF (ABS(offdia(mm))+DD.EQ.DD) GO TO 2
     12 CONTINUE
     mm=number
     IF(mm.NE.L)THEN
        IF(ITER.EQ.30)print*,'too many iterations'
        ITER=ITER+1
        G=(lambda(L+1)-lambda(L))/(2.*offdia(L))
        rr=SQRT(G**2+1.)
        G=lambda(mm)-lambda(L)+offdia(L)/(G+SIGN(rr,G))
        S=1.
        C=1.
        P=0.
        DO 14 l=mm-1,L,-1
           F=S*offdia(l)
           B=C*offdia(l)
           IF(ABS(F).GE.ABS(G))THEN
              C=G/F
              rr=SQRT(C**2+1.)
              offdia(l+1)=F*rr
              S=1./rr
              C=C*S
           ELSE
              S=F/G
              rr=SQRT(S**2+1.)
              offdia(l+1)=G*rr
              C=1./rr
              S=S*C
           ENDIF
           G=lambda(l+1)-P
           rr=(lambda(1)-G)*S+2.*C*B
           P=S*rr
           lambda(l+1)=G+P
           G=C*rr-B
           DO 13 K=1,number
              F=n(K,l+1)
              n(K,l+1)=S*n(K,l)+C*F
              n(K,1)=C*n(K,l)-S*F
           13 CONTINUE
  15 CONTINUE
  lambda(L)=lambda(L)-P
  offdia(L)=G
offdia(mm)=0.
GO TO 1
ENDIF
CONTINUE
ENDIF

call eigrt(lambda,n,number)
RETURN
END

SUBROUTINE EIGRT(lambda,n,number)
implicit none
real*8 lambda(36),n(36,36),p
integer i,j,k,number

This routine was taken from Section 11.1 in Numerical Recipes by Press et al.
Given the eigenvalues lambda and the eigenvectors n, this routine sorts the eigenvalues into descending order, and rearranges the eigenvectors correspondingly. The method is straight insertion.

DO 13 l=1,number-1
    K=l
    P=lambda(l)
    DO 11 J=l+1,number
        IF(lambda(J).ge.P)THEN
            K=J
            P=lambda(J)
        ENDIF
    11 CONTINUE
    IF(K.NE.I)THEN
        lambda(l)=lambda(l)
        lambda(l)=p
        DO 12 J=1,number
            P=n(J,l)
            n(J,l)=n(J,K)
            n(J,K)=P
        12 CONTINUE
    ENDIF
  13 CONTINUE
RETURN
END

subroutine findn(evalue,evector,efunct,ap,ninput,ndia,pi,nref,loptk,f,ref,dia,x,refl,reff,dref,gmean,refs,wavel,input,nout,2cnorm)
implicit none
real*8 efunct(36,101,11),evector(36,36),evalue(36)
real*4 f(101,11),ap(36),x(101),dia(101),input(36),loptk(36,101,11)
Calculating the unconstrained expansion coefficients

do 200 j=1,ninput-nout
   ap(j)=0.0
   do 100 k=1,ninput-nout
      ap(j)=input(k)*evector(k,j)+ap(j)
   100 continue
   ap(j)=ap(j)*cnorm/sqrt(evalue(j))
200 continue

Calculating the unconstrained distribution

do 500 r=1,nref
   do 400 i=1,ndia
      f(i,r)=0.0
      do 300 j=1,ninput-nout
         f(i,r)=ap(j)*efunct(j,i,r)+f(i,r)
      300 continue
   400 continue
500 continue

Calculating the real part of the refractive index

refs=0.
intgrl=0.
do 700 r=1,nref
   ref=refi+float(r-1)*dref
   x2bar=0.
   do 600 i=1,ndia
      x2bar=x2bar+x(i)**2*f(i,r)
   600 continue
   intgrl=intgrl+x2bar
   refs=refs+x2bar*(ref-1.)**2
700 continue
refs=abs(refs/intgrl)
refs=sqrt(refs)+1.0
if(nref.eq.1)then
   rs=1
else
   rs=nint((refs-refi)/dref)+1
endif
write(12,800)refs
800 format(2x,'refractive index=',gl0.3)
return
end

subroutine supefcal(efunct, supef, rs, ndia, dx, dref, ninput, nout, nref, insupef)
implicit none

real*8 efunct(36,101,11),supef(101,101,11)
real*8 supnrm,lnprd
real*4 dx,dref

integer rs,j,i,r,l,nsupef,ndia,nref,m,ninput,nout

remove these comment marks to verify the orthogonality of the eigenfunctions
real*8 phil2
integer k

nsupef=ndia

print*, 'calculating the supplemental eigenfunctions'
do 300 j=1,nsupef
   do 200 i=1,ndia
      do 100 r=1,nref
         if(j.eq.i.and.r.eq.rs)then
            if(nref.ne.1)then
               supef(j,i,r)=dble(sqrt(1./dx/dref))
            else
               supef(j,i,r)=dble(sqrt(1./dx))
            endif
         else
            supef(j,i,r)=0.d0
         endif
      100 continue
   200 continue
300 continue

print*, 'orthogonalizing the supplemental eigenfunctions'
m=0
do 1600 l=1,nsupef
   do 1500 j=1,ninput-nout+l-1
      inprd=0.d0
      do 600 i=1,ndia
         do 500 r=1,nref
            if(j .le. ninput-nout)then
               inprd=inprd+supef(l,i,r)*efunct(j,i,r)
            else
               inprd=inprd+supef(l,i,r)*supef(j-(ninput-nout),i,r)
            endif
500 continue
600 continue
         if(nref.eq.1)then
            inprd=inprd*dble(dx)
         else
            inprd=inprd*dble(dx*dref)
         endif
      do 800 i=1,ndia
         do 700 r=1,nref
147
if (j .le. ninput-nout) then
   super(I,i,r)=super(I,i,r)-inprd*efunct(j,i,r)
else
   super(I,i,r)=super(I,i,r)-inprd*
1 super(j-(ninput-nout),i,r)
endif
700 continue
800 continue
supnrm=0.d0
do 1000 i=1,ndia
   do 900 r=1,nref
      supnrm=supnrm+super(I,i,r)**2
900 continue
1000 continue
if(nref.eq.1)then
   supnrm=3*supnrm*dble(dx)
else
   supnrm=supnrm*dble(dx*dref)
endif
1100 continue
1200 continue
if(supnrm.lt.0.1d0)m=m+1
1300 continue
print*,'number of supplemental eigenfunctions =',nsupef-m
write(12,*)'number of supplemental eigenfunctions =',nsupef-m
Remove the comment marks to verify the orthogonality of
the eigenfunctions
print*,'checking the orthogonalization of the'
print*, 'supplemental eigenfunctions'
do 2000 I=1,nsupef+ninput-nout
do 2000 I=ninput-nout + 1,nsupef+ninput-nout
do 1900 k=1,nsupef+ninput-nout
   phi2=0.d0
1800 continue
if(k .le. ninput-nout)then
   phi2=phi2+efunct(k,i,r)*efunct(I,i,r)
elseif(k.gt.ninput-nout)then
   phi2=phi2+efunct(I,i,r)*super(k-(ninput-nout),i,r)
elseif(I.gt.ninput-nout)then
   phi2=phi2+efunct(I,i,r)*super(k-(ninput-nout),i,r)
elseif(I.gt.ninput-nout)then
   phi2=phi2+efunct(I,i,r)*super(I-(ninput-nout),i,r)
else
   phi2=phi2+super(I-(ninput-nout),i,r)
C 1 super(k-(ninput-nout), i, r)
C endif
C1700 continue
C1800 continue
C if(nref.eq.1)then
C phi2=phi2*db!a(dx)
C else
C phi2=phi2*dble(dx*dref)
C endif
C if(dabs(phi2).gt.0.01d0)then
C print*, I, k, phi2
C endif
C1900 continue
C pause
C2000 continue
C return
Cend
C subroutine cnrmcal(optk, dinput, ft, cnorm, dx, ndia, rs, ninput, nout, inref, dref)
imPLICIT NONE
REAL*4 optk(36, 101, 11), dinput(36), ft(101, 11)
REAL*4 cnorm, dx, dref
INTEGER ninput, I, j, ndia, rs, nout, nref, r
PRINT*, 'calculating the normalization constant'
cnorm=0.
DO 300 j=1, ninput-nout
   DO 200 i=1, ndia
      DO 100 r=1, nref
         cnorm=cnorm+ft(i, rs)*optk(j, i, rs)*dinput(j)
   100 continue
200 continue
300 continue
IF(nref.eq.1) THEN
   cnorm=cnorm*dx/REAL(ninput-nout)
ELSE
   cnorm=cnorm*dx*dref/REAL(ninput-nout)
ENDIF
RETURN
END
C subroutine findat(at, x, dx, ndia, rs, efunct, superf, nsuperf, pi, wavel, idref, nref, ft, f, ninput, nout)
imPLICIT NONE
REAL*8 superf(101, 101, 11), efunct(36, 101, 11)
REAL*4 f(101, 11), at(137), x(101), ft(101, 11), fplot(101)
real*4 dx, intgrl, pi, wavel, dref, xt

integer rs, imp, i, j, nsupef, ndla, ninput, nref, nout, r

character yn*1

100 continue

print*, 'previous peak size parameter = ', xt
print*, 'enter the peak size parameter'
read(*,*) xt
imp=nint((xt-x(1))/dx)+1

do 100 i=1, ndla
  do 200 r=1, nref
    if (i.eq.imp .and. r.eq.rs) then
      if (nref.eq.1) then
        ft(i,r)=1./dx
      else
        ft(i,r)=1./dx/dref
      endif
    else
      ft(i,r)=0.
    endif
  enddo 200
  continue 100

print*,'calculating trial function expansion coefficients'

1200 continue
1300 continue
1400 continue

print*, 'calculating the trial function'
1500 intgrl=0.

1600 continue
integral=integral+ft(i,r)
1700 continue
1800 continue
if(nref.eq.1)then
  integral=integral*dx
else
  integral=integral*dx*dref
endif
print*, 'integral of the trial function = ', integral
print*, 'renormalize the trial function? (y/n)'
read(*,1900)yn
1900 format(a1)
if(yn.eq.'y')then
  do 2000 j=1,ninput-nout+nsuper
    at(j)=at(j)/integral
  2000 continue
  goto 1500
endif

do 2100 i=1,ndia
  fplot(i)=ft(i,rs)
  yn='n'
  print*, 'view the trial function? (y/n)'
  read(*,1900)yn
cccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
  c
  This portion of code requires the use of Superplot on an
  c Apple Macintosh
  c ndia=number of points to plot
  c x=size parameter array
  c fplot=array containing the trial function
  c if(yn.eq.'y')call plot(ndia,x,fplot,fplot)
cccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
return
end

subroutine rruca1(evalue,ac,at,gamma,ninput,nout,conrm)
  implicit none
  real*8 evalue(36)
  real*4 ac(137),at(137)
  real*4 gamma,drrv,temp,conrm
  integer ninput,j,nout
  character*1 chgyn,tab

151
tab=char(9)
write(12,*)'gamma,drrv'

100 print*, 'enter a value for gamma'
read(*,*)gamma
print*, 'gamma=',gamma

drrv=0.0

do 200 j=1,ninput-nout
   temp=evalue(j)*((gamma*(ac(j)-at(j)))**2-cnorm**2)
   temp=temp/(evalue(j)+gamma)
   temp=temp/(evalue(j)+gamma)
   temp=temp/(evalue(j)+gamma)
   drrv=drrv+temp

200 continue

drrv=2*drrv

write(12,300)gamma,tab,drrv
300 format(g10.3,a1,g10.3)
write(*,400)gamma
write(*,600)drrv
400 format(2x,'gamma = ',g12.4)
600 format(2x,'slope of the rrv.us.gamma = ',g12.4)

print*, 'change the value of gamma?(y/n)'
read(*,700)chgyn
700 format(a1)
if(chgyn.eq.'y')goto100

return
end

subroutine findf(efunct,supef,ap,at,ac,refs,gamma,ndia,ninput,
inout,nref,nsupef,rs,norm,dx,dref,f,evalue,x,fplot,dfplot,dinput,
2evector,cnorm,optk,error,input,gmean,referr)
implicit none
real*8 efunct(36,101,11),supef(101,101,11),evector(36,36),evalue(36)
real*4 ap(36),at(137),ac(137),f(101,11),x(101),fplot(101),
df(101,11),dfplot(101),dac(137),dinput(36),optk(36,101,11),
2reserr(36),input(36)
real*4 gamma,refs,norm,dx,dref,cnorm,error,gmean,referr
integer i,j,rs,ninput,nout,nsupef,nref,ndia
character wndwyn*1,tab*1

152
Calculating the constrained expansion coefficients and the uncertainty in the constrained expansion coefficients.
The uncertainty in the constrained expansion coefficients is calculated in the manner described by Curry, Appl. Opt., 28, 1345 (1989).

```
do 100 j = 1, ninput-nout+nsupef
  if(j.le.ninput-nout)then
    ac(j) = (ap(j)*evalue(j)+at(j)*gamma)/(gamma+evalue(j))
    dac(j) = 1./ (evalue(j)+gamma)
  else
    ac(j) = at(j)
    dac(j) = 1./gamma
  endif
100 continue
```

Calculating the constrained solution and the uncertainty in the constrained solution.

```
200 continue
norm=0.
do 500 r=1,nref
do 400 i=1,ndl
  f(i,r)=0.
  df(i,r)=0.
do 300 j=1,ninput-nout+nsupef
  if(j.le.ninput-nout)then
    f(i,r)=f(i,r)+ac(j)*efunct(j,i,r)
    df(i,r)=df(i,r)+dac(j)*efunct(j,i,r)**2
  else
    f(i,r)=f(i,r)+ac(j)*supef(j-(ninput-nout),i,r)
    df(i,r)=df(i,r)+dac(j)*supef(j-(ninput-nout),i,r)**2
  endif
300 continue
norm=norm+f(i,r)
  df(i,r)=sqrt(df(i,r))
do 400 continue
500 continue
if(nref.eq.1)then
  norm=norm*dx
else
  norm=norm*dx*dref
endif
write(12,*)'constrained PSDF'
do 700 i=1,ndla
  fplot(i)=0.
  dfplot(i)=0.
do 600 r=1,nref
  fplot(i)=f(i,r)+fplot(i)
  dfplot(i)=df(i,r)+dfplot(i)
do 700 continue
if(nref.ne.1)then
  fplot(i)=fplot(i)*dref
```

153
dfplot(i)=dfplot(i)*dref
endif
write(12,800)x(i),tab,fplot(i)
700 continue
800 format(g12.4,a1,g12.4)
referr=0.
do 900 i=1,ndia
   referr=referr+df(i,rs)
900 continue
referr=referr*dx/(x(ndia)-x(1))
print*,'return to view the retrieved size distribution'
pause
cccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
This portion of code requires the use of Superplot on an
Apple Macintosh
ndia=number of points to plot
x=size parameter array
fPlot=array containing the constrained PSDF
ccall pIot(ndia,x,fplot,fplot)
cccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc
print*,'window the retrieved size distribution?(y/n)'
read(*,1000)wndwyn
1000 format(a1)
if(wndwyn.eq.'y')then
   call window(fplot,ndia,x,dx)
endif
do 1200 j=1,ninput-nout
   reserr(j)=0.0
do 1100 i=1,ndia
      reserr(j)=reserr(j)+fplot(i)*optk(j,i,rs)
1100 continue
reserr(j)=reserr(j)*dx/cnorm
1200 continue
error=0.
do 1300 j=1,ninput-nout
   reserr(j)=(reserr(j)-input(j))*gmean
   error=error+reserr(j)**2
1300 continue
error=sqrt(error/float(ninput-nout))
return
end
cc subroutine pIot(ndia,x,f1,f2)
implicit none
include "Quickdraw.inc"
include "Types.inc"

integer ndia
real*4 x(101), f1(101), f2(101)
character*256 str255, ptype1, ptype2, template
logical firstTime, notFirstTime, drawlt, dontDrawlt
PASCAL EXTERNAL SuperPlotReal

c Setup the SuperPlot logicals
firstTime=.true.
notFirstTime=.false.
drawlt=.true.
dontDrawlt=.false.

template=str255('PSDFTemplate')

c If you pass a template name, the template can be located in one of
c three places.
c In the same folder as the application, in a folder called SPtemplates
c located
c in the same folder as the application, or in a folder called
SPtemplates located in the system folder.
c
ptype1=str255('Line')
ptype2=str255('DoubleY')
ptype2=''

if(ptype2 .eq. '')then
  call SuperPlotReal(VAL2(ndia), x, f1, ptype1, template,
                   1  VAL1(firstTime), VAL1(drawlt))
else
  call SuperPlotReal(VAL2(ndia), x, f1, ptype1, template,
                   1  VAL1(firstTime), VAL1(dontDrawlt))
  call SuperPlotReal(VAL2(ndia), x, f2, ptype2, template,
                   1  VAL1(notFirstTime), VAL1(drawlt))
endif

return
end

subroutine plotg(ninput, detct, g, dg)
implicit none
include "Quickdraw.inc"
include "Types.inc"

integer ninput
real*4 detct(36), g(36), dg(36)
character*256 str255, ptype1, ptype2, template
logical firstTime, notFirstTime, drawlt, dontDrawlt
PASCAL EXTERNAL SuperPlotReal

c Setup the SuperPlot logicals
firstTime=.true.
notFirstTime=.false.
drawlt=.true.
dntDrawlt=.false.

template=str255('ScatteringTemplate')

ptype1=str255('Line')
ptype2=str255('DoubleV')
ctype=''

if(ptype2 .eq. ' ')
    call SuperPlotReal(UAL2(ninput),detct,g,ptypel,template,
        1 UAL1(firstTime),UAL1(drawlt))
else
    call SuperPlotReal(UAL2(ninput),detct,g,ptypel,template,
        1 UAL1(firstTime),UAL1(dontDrawlt))
    call SuperPlotReal(UAL2(ninput),detct,dg,ptype2,template,
        1 UAL1(notFirstTime),UAL1(drawlt))
endif
return

end

subroutine plotg2(ninput,detct,g,dg)

implicit none
include "Quickdraw.inc"
include "Types.inc"
integer ninput
real*4 detct(36),g(36),dg(36)
character*256 str255,ptypel,ptype2,tempate
logical firstTime,notFirstTime,drawlt,dontDrawlt

PASCAL EXTERNAL SuperPlotReal

c Setup the SuperPlot logicals
firstTime=.true.
notFirstTime=.false.
drawlt=.true.
dontDrawlt=.false.

template=str255('CompScatPatTemplate')

ptype1=str255('Line')
ptype2=str255('DoubleV')
ctype=''

if(ptype2 .eq. ' ')
    call SuperPlotReal(UAL2(ninput),detct,g,ptypel,template,
        1 UAL1(firstTime),UAL1(drawlt))
else
    call SuperPlotReal(UAL2(ninput),detct,g,ptypel,template,
        1 UAL1(firstTime),UAL1(dontDrawlt))
    call SuperPlotReal(UAL2(ninput),detct,dg,ptype2,template,
        1 UAL1(notFirstTime),UAL1(drawlt))
endif
return
end

---

`str255: converts a FORTRAN string to a Pascal LSTRING`

`function str255(string)`
`character*(*) string`
`str255 = char(len(trim/string)))//string`  
`end`

```
subroutine findk(fplot,g,dg,gmean,refs,detct,ndetct,alpha,nangle,
  IwaveI,ndia,dx,dia,experr,waven,alphal,alphal3,nangle1,nangle3,
  2dtheta,dtheta1,dtheta3)
  implicit none
  real*4 fplot(101),g(36),detct(36),dg(36),decrat(36),dia(101),
    ltheta(100)
  real*4 pi,wavel,ref3,dx,refims,gmean,experr,error,alpha,waven,
    liper,ipar,alphal,alphal3,dtheta,dtheta1,dtheta3
  integer ndia,ndetct,i,j,nangle,nangle1,nangle3,ndetct
  character tab*1,yn*1
  tab=char(9)
  pi=4.*atan(1.0)
  print*,'finding the imaginary part of the refractive index'
  continue
  print*,'enter a value for the imaginary part of the refractive index'
    read(*,*)refims
  print*,'calculating the scattering pattern'
  do 400 idetct=1,ndetct
    decrat(idetct)=0.
  c This portion of the code is specific to the nephelometers used in
  c the Laser Applications Laboratory at Argonne
  c ndetct=number of detectors
  cfunctions over the solid angle subtended by each detector
  calpha=angular resolution of the detectors. A detector
  cwith a scattering angle of detct(idetct) will receive light from a
  cparticle at the center of the scattering volume that is scattered
  cin any direction between the detct(idetct)alpha directions.
  cnangle=number of angles used in averaging the Mie intensity
  cfunctions over the solid angle subtended by each detector. The
  cnumber of angles is chosen so that the Mie intensity functions are
  cacculated at least every 0.1 degree.
  cdtheta=angular step size in averaging the Mie intensity functions
  cdtheta - 0.1 degree
```

157
IF (NDETCT .EQ. 15) THEN
  IF (IDETCT .EQ. 1 .OR. IDETCT .EQ. 2) THEN
    NANGLE = NANGLE1
    DTHETA = DTHETA1
    ALPHA = ALPHA1
  ELSE
    NANGLE = NANGLE3
    DTHETA = DTHETA3
    ALPHA = ALPHA3
  ENDIF
ENDIF

DO 200 J = 1, NANGLE
  THETA(J) = (DETCT(IDETCT) * PI / 180.) - ALPHA + FLOAT(J-1) * DTHETA
  CONTINUE
DO 300 I = 1, NDATA
  IF (FPLT(I) .GT. 0.01) THEN
    CALL BMIE(WAVE, DIA(I), REFS, REFSIM, IPAR, IPER, THETA, NANGLE)
    DECROT(IDETCT) = FPLT(I) * (IPAR + IPER) + DECROT(IDETCT)
  ENDIF
  CONTINUE
DECROT(IDETCT) = DECROT(IDETCT) * DX / 2. / WAVE**2
  CONTINUE
WRITE(12, 500)
WRITE(*, 500)
500  FORMAT(2X, 'ANGULAR SCATTERING CROSS SECTIONS', /,
     12X, 'CALCULATED', 4X, 'MEASURED', 4X, 'CALCULATED ERROR', 4X, 'EXPECTED ERROR')
ERROR = 0.
DO 700 J = 1, NDCT
  PRINT*, DETCT(J), DECROT(J), G(J), ABS(DECROT(J) - G(J)), DG(J)
  ERROR = ERROR + (DECROT(J) - G(J))**2
  WRITE(12, 600) DETCT(J), TAB, DECROT(J), TAB, G(J), TAB,
1       ABS(DECROT(J) - G(J)), TAB, DG(J)
600  FORMAT(E12.4, A1, E12.4, 01, E12.4, 01, E12.4, 01, E12.4)
  IF (AMOD(FLOAT(J), 30.) .EQ. 0.0 .OR. J .EQ. NDCT) THEN
    PRINT*, 'RETURN TO CONTINUE'
    PAUSE
  ENDIF
  CONTINUE
ERROR = SQRT(ERROR / FLOAT(NDCT))
PRINT*, 'IMAGINARY PART OF THE REFRACTIVE INDEX =', REFSIM
WRITE(12, *) 'IMAGINARY PART OF THE REFRACTIVE INDEX =', REFSIM
PRINT*, 'RMS ERROR = ', ERROR
WRITE(12, *) 'RMS ERROR = ', ERROR
PRINT*, 'EXPECTED RMS ERROR = ', EXPRR
WRITE(12, *) 'EXPECTED RMS ERROR = ', EXPRR
PRINT*, 'COMPARE THE CALCULATED AND MEASURED SCATTERING PATTERNS?(Y 1/N)'
READ(*, 800) YN
This portion of code requires the use of Superplot on an Apple Macintosh.

ndetct=number of points to plot
detct=array containing the scattering angles
g=array containing the measured differential scattering cross sections
decrat=array containing the calculated differential scattering cross sections

if(yn.eq.'y')call plotg2(ndetct,detct,g,decrat)

print*,'change the value of k?(y/n)' read(*,800)yn
800 format(a1)
if(yn.eq.'y')goto 100
return
end

subroutine bhmie(wavel,dia,refre,refim,ipar,iper,theta,nangle)
dimension amu(100),theta(100),pi(100),tau(100),pi0(100),pi1(100)
real*4 ipar,iper
complex d(3000),y,refrel,xi,xi0,xi1,an,bn,s1(200),s2(200)
real*8 psi0,psi1,psi,dn,dx
	pie=4.0*atan(1.0)
	refmed=1.0

do 50 j=1,nangle
	amu(j)=cos(theta(j))
50 continue

refrel=cmplx(refre,refim)/refmed

x=pie*dia/wavel
dx=x
y=x*refrel
xstop=x+4.*x**.3333+2.
nstop=xstop
ymod=cabs(y)
nnx=amax1(xstop,ymod)+15

d(nx)=cmplx(0.,0.)
nn=nnx-1

do 100 n=1,nn
	nn=nnx-n+1
	d(nx-n)=(rn/y)-(1./(d(nx-n)+rn/y))
100 continue

do 200 j=1,nangle
	pi0(j)=0.
	pi1(j)=1.
200 continue
nn=2*nangle-1

do 300 j=1,nn
   s1(j)=cmplx(0.,0.)
   s2(j)=cmplx(0.,0.)
300 continue

psi0=dcos(dx)
psi1=dsin(dx)
chi0=-sin(x)
chi1=cos(x)
apsi0=psi0
apsi1=psi1
x10=cmplx(apsi0,-chi0)
x1l=cmplx(apsi1,-chi1)
n=1

400 dn=n
rn=n
fn=(2.*rn+1.)/(rn*(rn+1.))
gn=(rn*(rn+2.))/(rn+1.)
psi=(2.*dn-1.)*psi1/dx-psi0
apsi=psi
chi=(2.*rn-1.)*chi1/x-chi0
xi=cmplx(apsi,-chi)
an=(d(n)/refrel+rn/x)*apsi-apsi1
an=an/((d(n)/refrel+rn/x)*xi-xi1)
bn=(refrel*d(n)+rn/x)*apsi-apsi1
bn=bn/((refrel*d(n)+rn/x)*xi-xi1)
do 500 j=1,nangle
   jj=2*nangle-j
   pi(j)=pi1(j)
   tau(j)=rn*pi(j)*amu(j)-(rn+1.)*pi0(j)
   p=(-1.)**(n-1)
   s1(j)=s1(j)+fn*(an*pi(j)+bn*tau(j))
   t=(-1.)**(n-1)
   s2(j)=s2(j)+fn*(an*tau(j)+bn*pi(j))
if (j.ne.jj) then
   s1(jj)=s1(j)+fn*(an*pi(j)*p+bn*tau(j)*t)
   s2(jj)=s2(j)+fn*(an*tau(j)*t+bn*pi(j)*p)
endif
500 continue

psi0=psi1
psi1=psi
apsi1=apsi
chi0=chi1
chi1=chi
x1l=cmplx(apsi1,-chi1)
\n
n=n+1
rn=n

do 600 j=1,nangle
   pi1(j)=((2.*rn-1.)/(rn-1.))*pi(j)*amu(j)
   pi1(j)=pi1(j)-rn*pi0(j)/(rn-1.)
   pi0(j)=pi(j)
600 continue
if (n-1-nstop) 400,700,700

700 continue

ipar=0.0
iper=0.0

do 800 j=1,ngangle
   ipar=cabs(s2(j))**2+ipar
   iper=cabs(s1(j))**2+iper
800 continue

ipar=ipar/float(ngangle)
iper=iper/float(ngangle)

return
end

cc

subroutine window(fplot,ndia,x,dx)
implicit none
real*4 fplot(101),fwnd(101),x(101)
real*4 dx,xmin,xmax,norm,pi
integer i,ndia
pi=4.*atan(1.0)

print*, 'enter the range of the window'
read(*,*)xmin,xmax

if(xmax-xmin.le.dx)then
   xmin=xmin-dx/2.
   xmax=xmax+dx/2.
endif

norm=0.
do 100 i=1,ndia
   if(x(i).ge.xmin.and.x(i).le.xmax)then
      fwnd(i)=1.
   else
      fwnd(i)=0.
   endif
   fplot(i)=fplot(i)*fwnd(i)
   norm=norm+fplot(i)
100 continue

norm=norm*dx
print*, 'normalization integral of the windowed solution =',norm
write(12,*)'normalization integral of the windowed solution =',
inorm

do 200 i=1,ndia
   fplot(i)=fplot(i)/norm
200 continue

161
print*, 'return to view the windowed solution'
pause

cccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc

This portion of code requires the use of Superplot on an
Apple Macintosh.

ndia = number of points to plot
x = size parameter array
fplot = array containing the windowed PSDF

call plot(ndia, x, fplot, fplot)

cccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccccc

return
end
Appendix D
Calibration for Relative Scattering Measurements

The goal of a relative scattering calibration is to determine the relative sensitivity of each of the detectors. Once the relative sensitivity of each detector is known, relative scattering correction factors can be determined for each detector and used to correct the measured scattering pattern.

\[ RM_j^c = RSCF_j \cdot M_j^c \]  \hspace{1cm} (D.1)

where

\[ M_j^c = \frac{M_{ij} - SL_{ij}}{BD_i} \]  \hspace{1cm} (D.2)

Calibration for relative scattering measurements requires an isotropic light source. A preliminary study was conducted to determine if the light leaking radially outward from a plastic optical fiber can be used as the light source. An Eska\textsuperscript{TM} CK-20 plastic optical fiber was fed through the inlet tube and fixed with a rubber stopper such that the fiber passed through the center of the scattering volume. This type of optical fiber has a relatively high attenuation rate and is primarily used in decorations, signs, and displays. The end of the fiber was then placed in front of a white light source, and the output of each channel was recorded. The measurements were repeated sixteen times, and after each measurement, the fiber was rotated approximately 45°. The relative scattering correction factors were calculated for each fiber position by subtracting the dark currents from each measurement, normalizing the measurements by their mean and then inverting the result. Normalizing by the mean of the measurements eliminates any error due to fluctuations in the power output of the white light source. Also, the acceptance angles of the first two detectors on the 15 channel nephelometer are smaller than the acceptance angles of the other thirteen channels, so the output from the first two channels was multiplied by the ratio of the acceptance angle of channels 3 through 13 to the acceptance angle of channels 1 and 2. The acceptance angles are listed in Table 5.2. The relative scattering correction factors for channels 1 and 2 are calculated from
The correction factors for channels 3 through 15 are given by

\[
\text{RSCF}_{1,2} = \frac{\frac{1}{15} \left\{ (\alpha_3 / \alpha_1)(\text{RM}_1 - \text{DC}_1) + (\text{RM}_2 - \text{DC}_2) \right\} + \sum_{k=3}^{15} \text{RM}_k - \text{DC}_k}{(\alpha_3 / \alpha_1)(\text{RM}_{1,2} - \text{DC}_{1,2})}
\]

The relative scattering correction factors were calculated for each channel at each fiber location. Then, the effects of any anisotropy in the amount of energy leaking out of the fiber in a particular direction were eliminated by averaging the relative scattering correction factors obtained for each fiber position. The averaged relative scattering correction factors obtained on two different days using different fibers are shown in Figure D.1. The error bars show one standard deviation. Although the error bars indicate considerable scatter in the measurements, the average values are fairly consistent between the data sets and are in agreement with the absolute scattering correction factors shown in Figure 5.10. It is believed that the scatter is largely due to the difficulty of placing the fiber exactly in the center of the scattering volume. Once a better method of positioning the fiber is developed, this technique will provide a simple way of calibrating a polar multi-channel nephelometer for relative scattering measurements.
Figure D.1. Relative Scattering Correction Factors
Appendix E
Listing of Nephelometer.f

program nephelometer

implicit none

pt3d(3),pt4d(3),dect(36),theta(1801),preWall(36),intens(1801),
volume(36),dd15(15),do15(15),dd36(36),do36(36)

real*8 pi,dia,refre,refim,wavel,press,temp,concen,waven,ro,ri,dd,
d1,angres,hcone,hbeam,a,b,c,domega,dphi,thetas,thetad,dangle,
2dtheta,magr1,magr2,magr3,magr4,r1r2,r3r4,exp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
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3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
3dz,xp,yp,zp,change,dc,dx,dy,
calculating the differential scattering cross sections at
0.1 degree intervals

print*, 'calculating the differential scattering cross sections'
open(unit=45, file='scatteringpattern.out', status='unknown')
write(45,*) 'diameter = ', dia, ', n = ', refre, ', k = ', refim

dangle = 1*pi/180.
do 110 j = 1, 1801
   theta(j) = float(j-1)*dangle
110 continue

call bhmie(dia, theta, refre, refim, intens, 1801, wave1, waven)
do 120 j = 1, 1801
   if(amod(float(j-1),10.).eq.0.0)then
      write(45,*) theta(j)*180./pi, intens(j), intens(j)*waven**2
   endif
120 continue

close(unit=45)

gemetric parameters for the nephelometer
all lengths are in centimeters

if(ndetct.eq.36)then
   ro=6.262
   ri=3.048
   db=0.3
   dj=.38
   do 130 idetct = 1, ndetct
      dd36(idetct)=.18
      da136(idetct)=.178
   130 continue
elseif(ndetct.eq.15)then
   ro=6.17
   ri=1.905
   db=.39
   dj=.38
   dd15(1)=.235
   dd15(2)=.235
   dd15(3)=.282
   dd15(4)=.282
   dd15(5)=.282
   dd15(6)=.282
   dd15(7)=.282
   dd15(8)=.282
   dd15(9)=.282
   dd15(10)=.282
   dd15(11)=.282
   dd15(12)=.282
   dd15(13)=.282
   dd15(14)=.282
   dd15(15)=.282
   da115(1)=.235
   da115(2)=.235
   da115(3)=.342
   da115(4)=.342
   da115(5)=.342
   da115(6)=.342
   da115(7)=.342
   da115(8)=.342
   da115(9)=.342
   da115(10)=.342
   da115(11)=.342
   da115(12)=.342
   da115(13)=.342
   da115(14)=.342
   da115(15)=.342

167
calculating the nominal scattering angle for each detector

if(ndetct.eq.15) then
  do 200 idetct=1,ndetct
     detct(idetct)=2.*asin(float(idetct-1)*0.05+0.2)
  continue
else if(ndetct.eq.36) then
  do 300 idetct=1,ndetct
     detct(idetct)=(20.+float(idetct-1)*4.)*pi/180.
  continue
endif

calculating the scattering volumes

print*, 'calculating the scattering volumes'
if(calyn.eq.'y') then
  do 350 idetct=1,ndetct
     if(ndetct.eq.15) then
       dd=dd15(idetct)
       da1=da115(idetct)
     else
       dd=dd36(idetct)
       da1=da136(idetct)
     endif
     alpha=2.*atan((dd+dat)/2./(ro-ri))
     angres=atan(dd/2./ro)
     rstar=(ro*da1-ri*dd)/2./(ro-ri)
     call vcal(alpha,dd,db,ro,detct(idetct),volume(idetct))
     print*, 'detector=', idetct, ' volume=', volume(idetct)
  continue
else
  if(ndetct.eq.36) then
    call veal(ndetct,db,dj,vol)
  else
    call veal(ndetct,dj,db,vol)
  endif
  print*, 'scattering volume=', vol
endif

open(unit=44,file='nephelometer.out',status='unknown')
write(44,*)' wavelength = ', wavel
write(44,*)' particle size = ',dia
write(44,*)' refractive index = ',refre,refim
write(44,*)' distance to the detectors = ',ro
write(44,*)' aerosol jet diameter = ',dj
write(44,*)' beam diameter = ',db
write(44,*)' number of detectors = ',ndetct
write(44,*)' concentration = ',concen

c calculating the angular scattering cross sections
c
print*,'calculating the angular scattering cross sections'
do 1500 idetct=1,ndetct
  thetad=detct(idetct)
  preUsi(idetct)=0.
  print*,'detector*=', idetct, ' scattering angle=',thetad*180/pi
  nx=11
  ny=23
  nz=11
  continue
write(44,*)'number of steps : ',nx,ny,nz
print*,'number of steps : ',nx,ny,nz
write(44,450)tab,tab,tab,tab,tab
write(*,450)tab,tab,tab,tab,tab
450 format('angle','a1,Csca','a1,change','a1,'N*dCsca/dOmega'

1 ,a1,'volume')
Wali(idetct)=0.
avgomi=0.

if(calyn.eq.'y')then
  npart=nx*ny*nz
  if(ndetct.eq.15)then
    dd=dd15(idetct)
    da1=da115(idetct)
  else
    dd=dd36(idetct)
    da1=da136(idetct)
  endif
  alpha=2.*atan(((dd+da1)/2.)/(ro-ri))
  angres=atan(dd/2./ro)
  rstar=(ro*da1-ri*dd)/2./(ro-ri)
  a=tan(alpha/2.)*dsin(thetad)-dcos(thetad)
  b=(ro*tan(alpha/2.)-dd/2.)*dsin(thetad)
  c=-tan(alpha/2.)*dsin(thetad)-dcos(thetad)
  if(nx.ne.1)then
    dx=db/float(nx-1)
  else
    dx=0.
  endif
  do 900 ix=1,nx
    if(nx.ne.1)then
      xp=-db/2.+float(ix-1)*dx
    else
      xp=0.
    endif
    ymax=(xp*a*dsin(thetad)+b)/(1.+a*dcos(thetad))
 169
ymin=(xp*c*d\sin(\theta_d)-b)/(1.+c*d\cos(\theta_d))

if(ny.ne.1)then
    dy=(ymax-ymin)/float(ny-1)
else
    dy=0.
endif

do 800  iy=1,ny
if(ny.ne.1)then
    yp=ymin+float(iy-1)*dy
else
    yp=0.
endif

xd=d\sin(\theta_d)*xp-d\cos(\theta_d)*yp
yd=d\cos(\theta_d)*xp+d\sin(\theta_d)*yp
ydm\ax=d(xd, alpha, dd, ro)
pt2d(1)=-ro
pt1d(3)=0.
pt2d(3)=0.
if(abs(yd).le.rstar)then
    pt1d(1)=-ro
    if(yd.ge.0.)then
        pt1d(2)=dd/2.
        pt2d(2)=-dd/2.
    else
        pt1d(2)=-dd/2.
        pt2d(2)=dd/2.
    endif
else
    pt1d(1)=-ri
    if(yd.ge.0.)then
        pt1d(2)=da/2.
        pt2d(2)=-dd/2.
    else
        pt1d(2)=-da/2.
        pt2d(2)=dd/2.
    endif
endif
hcone=dsqrt(dabs(ydm\ax**2/4.-yd**2))
hbeam=dsqrt(dabs(db**2/4.-xp**2))
h=min(hcone,hbeam)
if(nz.ne.1)then
    dz=h/float(nz-1)
else
    dz=0.
endif

do 700  iz=1,nz
    zp=float(iz-1)*dz
    pt4d(1)=-ro
    pt3d(2)=0.
    pt4d(2)=0.
    pt4d(3)=-dd/2.
    if(zp.le.rstar)then
        pt3d(1)=-ro
        pt3d(3)=dd/2.
    else
        pt3d(1)=-ri
        pt3d(3)=da/2.
    endif
\( pt1(1) = pt1d(1) \cdot \sin(\text{thetad}) + pt1d(2) \cdot \cos(\text{thetad}) - xp \)
\( pt1(2) = -pt1d(1) \cdot \cos(\text{thetad}) + pt1d(2) \cdot \sin(\text{thetad}) - yp \)
\( pt1(3) = pt1d(3) - zp \)
\( pt2(1) = pt2d(1) \cdot \sin(\text{thetad}) + pt2d(2) \cdot \cos(\text{thetad}) - xp \)
\( pt2(2) = -pt2d(1) \cdot \cos(\text{thetad}) + pt2d(2) \cdot \sin(\text{thetad}) - yp \)
\( pt2(3) = pt2d(3) - zp \)
\( pt3(1) = pt3d(1) \cdot \sin(\text{thetad}) + pt3d(2) \cdot \cos(\text{thetad}) - xp \)
\( pt3(2) = -pt3d(1) \cdot \cos(\text{thetad}) + pt3d(2) \cdot \sin(\text{thetad}) - yp \)
\( pt3(3) = pt3d(3) - zp \)
\( pt4(1) = pt4d(1) \cdot \sin(\text{thetad}) + pt4d(2) \cdot \cos(\text{thetad}) - xp \)
\( pt4(2) = -pt4d(1) \cdot \cos(\text{thetad}) + pt4d(2) \cdot \sin(\text{thetad}) - yp \)
\( pt4(3) = pt4d(3) - zp \)

\( \text{magr1} = 0 \)
\( \text{magr2} = 0 \)
\( \text{magr3} = 0 \)
\( \text{magr4} = 0 \)
\( r1r2 = 0 \)
\( r3r4 = 0 \)

\[ \text{do } 500 \text{ i}=1,3 \]
\[ \text{magr1} = \text{magr1} + pt1(i)**2 \]
\[ \text{magr2} = \text{magr2} + pt2(i)**2 \]
\[ \text{magr3} = \text{magr3} + pt3(i)**2 \]
\[ \text{magr4} = \text{magr4} + pt4(i)**2 \]
\[ r1r2 = pt1(i) \cdot pt2(i) + r1r2 \]
\[ r3r4 = pt3(i) \cdot pt4(i) + r3r4 \]
\[ \text{continue} \]
\[ \text{magr1} = \text{dsqrt(magr1)} \]
\[ \text{magr2} = \text{dsqrt(magr2)} \]
\[ \text{magr3} = \text{dsqrt(magr3)} \]
\[ \text{magr4} = \text{dsqrt(magr4)} \]

\[ \text{arg} = \text{min}(|\text{r1r2}/\text{magr1}/\text{magr2}|, 1.0) \]
\[ \text{dtheta} = \text{dacos(arg)} \]
\[ \text{dtheta} = \text{dtheta} \cdot \cos(\pi \cdot zp/2./ydiv) \]
\[ \text{arg} = \text{min}(|\text{r3r4}/\text{magr3}/\text{magr4}|, 1.0) \]
\[ \text{dphi} = \text{dacos(arg)} \]
\[ \text{dphi} = \text{dphi} \cdot \cos(\pi \cdot \text{abs(yd)}/2./ydiv) \]
\[ \text{thetas} = (\text{dacos}(pt1(2)/\text{magr1}) + \text{dacos}(pt2(2)/\text{magr2}))/2. \]
\[ \text{ntheta} = (\text{nint}(\text{dtheta}/\text{dangle}))/2 + 1 \]
\[ \text{js} = \text{nint}(\text{thetas}/\text{dangle}) \]
\[ \text{ddcmg} = 0. \]
\[ \text{do } 600 \text{ j}=\text{js}-(\text{ntheta}-1)/2,\text{js}+(\text{ntheta}-1)/2 \]
\[ \text{ddcmg} = \text{ddcmg} + \text{intens}(j) \]
\[ \text{continue} \]
\[ \text{ddcmg} = \text{ddcmg}/\text{float(ntheta)} \]
\[ \text{domega} = \text{dtheta} \cdot \text{dphi} \cdot \pi/4. \]
\[ \text{augong} = \text{augong} + \text{domega} \]
\[ \text{Wall}(\text{idetct}) = \text{Wall}(\text{idetct}) + \text{ddcmg} \cdot \text{domega} \]

\[ \text{continue} \]
\[ \text{continue} \]
\[ \text{continue} \]
\[ \text{else} \]
\[ \text{if(ndetct.eq.15)} \text{then} \]

171
\[
\begin{align*}
\text{dd} &= \text{dd15(idetct)} \\
\text{dai} &= \text{dai15(idetct)} \\
\text{else} & \\
\text{dd} &= \text{dd36(idetct)} \\
\text{dai} &= \text{dai36(idetct)} \\
\text{endif} \\
\text{alpha} &= 2. \cdot \text{atan}(\text{dd} / 2. / \text{ro} - \text{ri}) \\
\text{angres} &= \text{atan}(\text{dd} / 2. / \text{ro}) \\
\text{rstar} &= (\text{ro} \cdot \text{dai} - \text{ri} \cdot \text{dd}) / 2. / (\text{ro} - \text{ri}) \\
\text{if} (\text{nx} \cdot \text{ne} . 1. ) & \text{then} \\
\text{dx} &= \text{db} / \text{float(nx-1)} \\
\text{else} & \\
\text{dx} &= 0. \\
\text{endif} \\
\text{npart} &= 0 \\
\text{do 1400} & \text{ ix=1,nx} \\
\text{if} (\text{nx} . 1. ) & \text{then} \\
\text{xp} &= -\text{db} / 2. + \text{float(ix-1)} \cdot \text{dx} \\
\text{else} & \\
\text{xp} &= 0. \\
\text{endif} \\
\text{if} (\text{abs(xp)} . \leq \text{dj}/2.) & \text{then} \\
\text{ymax} &= \text{dsqrt(\text{dj}**2/4.-xp**2)} \\
\text{ymin} &= -\text{ymax} \\
\text{if} (\text{ny} . 1. ) & \text{then} \\
\text{dy} &= (\text{ymax} - \text{ymin}) / \text{float(ny-1)} \\
\text{else} & \\
\text{dy} &= 0. \\
\text{endif} \\
\text{do 1300} & \text{ iy=1,ny} \\
\text{if} (\text{ny} . 1. ) & \text{then} \\
\text{yp} &= \text{ymin} + \text{float(iy-1)} \cdot \text{dy} \\
\text{else} & \\
\text{yp} &= 0. \\
\text{endif} \\
\text{if} (\text{xp}^2 + \text{yp}^2 \leq \text{dj}^2/4.) & \text{then} \\
\text{xd} &= \text{dsin(thetad)} \cdot \text{xp} - \text{dcos(thetad)} \cdot \text{yp} \\
\text{yd} &= \text{dcos(thetad)} \cdot \text{xp} + \text{dsin(thetad)} \cdot \text{yp} \\
\text{ydamx} &= \text{dc} (\text{xd}, \text{alpha}, \text{dd}, \text{ro}) \\
\text{pt2d}(1) &= -\text{ro} \\
\text{ptld}(3) &= 0. \\
\text{pt2d}(3) &= 0. \\
\text{if} (\text{abs(yd)} . \leq \text{rstar}) & \text{then} \\
\text{ptld}(1) &= -\text{ro} \\
\text{if} (\text{yd} . \geq 0.) & \text{then} \\
\text{ptld}(2) &= \text{dd}/2. \\
\text{pt2d}(2) &= -\text{dd}/2. \\
\text{else} & \\
\text{ptld}(2) &= -\text{dd}/2. \\
\text{pt2d}(2) &= \text{dd}/2. \\
\text{endif} \\
\text{else} & \\
\text{ptld}(1) &= -\text{ri} \\
\text{if} (\text{yd} . \geq 0.) & \text{then} \\
\text{ptld}(2) &= \text{dai}/2. \\
\text{pt2d}(2) &= -\text{dai}/2. \\
\text{else} & \\
\text{ptld}(2) &= -\text{dai}/2. \\
\end{align*}
\]
pt2d(2)=dd/2.
endif
endif

h=dsqrt(db**2/4.-xp**2)
if(nz.ne.1)
  dz=h/float(nz-1)
elset
  dz=0.
endif

do 1200 iz=1,nz
  npart=npart+1
  zp=float(iz-1)*dz
  pt4d(1)=-ro
  pt3d(2)=0.
  pt4d(2)=0.
  pt4d(3)=-dd/2.
  if(zp.Ie.rstar)then
    pt3d(1)=-ro
    pt3d(3)=dd/2.
  else
    pt3d(1)=-ri
    pt3d(3)=da1/2.
  endif
  pt1(1)=pt1d(1)*dsin(thetad)+pt1d(2)*
  dcos(thetad)-xp
  pt1(2)=-pt1d(1)*dcos(thetad)+pt1d(2)*
  dsin(thetad)-yp
  pt1(3)=pt1d(3)-zp
  pt2(1)=pt2d(1)*dsin(thetad)+pt2d(2)*
  dcos(thetad)-xp
  pt2(2)=-pt2d(1)*dcos(thetad)+pt2d(2)*
  dsin(thetad)-yp
  pt2(3)=pt2d(3)-zp
  pt3(1)=pt3d(1)*dsin(thetad)+pt3d(2)*
  dcos(thetad)-xp
  pt3(2)=-pt3d(1)*dcos(thetad)+pt3d(2)*
  dsin(thetad)-yp
  pt3(3)=pt3d(3)-zp
  pt4(1)=pt4d(1)*dsin(thetad)+pt4d(2)*
  dcos(thetad)-xp
  pt4(2)=-pt4d(1)*dcos(thetad)+pt4d(2)*
  dsin(thetad)-yp
  pt4(3)=pt4d(3)-zp
  magr1=0.
  magr2=0.
  magr3=0.
  magr4=0.
r1r2=0.
r3r4=0.
do 1000 i=1,3
  magr1=magr1+pt1(i)**2
  magr2=magr2+pt2(i)**2
  magr3=magr3+pt3(i)**2
  magr4=magr4+pt4(i)**2
  r1r2=pt1(i)*pt2(i)+r1r2
  r3r4=pt3(i)*pt4(i)+r3r4
  continue
1000
mAGR1=dsqrt(magr1)
magr2 = dsqrt(magr2)
magr3 = dsqrt(magr3)
magr4 = dsqrt(magr4)
arg = min(dabs(r1r2/magr1/magr2), 1.d0)
dtheta = dacos(arg)
dtheta = dtheta * dacos(pi*zp/2./yndmax)
arg = min(dabs(r3r4/magr3/magr4), 1.d0)
dphi = dacos(arg)
dphi = dphi * dacos(pi*abs(yd)/2./yndmax)

thetas = (dacos(pt1(2)/magr1) +
dacos(pt2(2)/magr2))/2.
ntheta = (nint(dtheta/dangle)/2)*2+1
js = nint(thetas/dangle)+1
dcdomg = 0.
do 1100 j = js-(ntheta-1)/2, js+(ntheta-1)/2
   dcdomg = dcdomg + intens(j)
1100 continue
   dcddomg = dcdomg / float(ntheta)
domega = dtheta*dphi*pi/4.
augomg = augomg + domega
   Wall(idetct) = Wall(idetct) + dcdomg*domega
1200 continue
endif
continue
endif
1300 continue
endif
1400 continue
volume(idetct) = vol
endif
Wall(idetct) = Wall(idetct) * concen * volume(idetct) / float(npart)
change = abs(Wall(idetct) - preWall(idetct)) / Wall(idetct)
augomg = augomg / float(npart)
write(44, 1600) detct(idetct) * 180./pi, tab, Wall(idetct), tab,
1 change, tab, augomg, tab, Wall(idetct) / augomg, tab, volume(idetct)
write(*, 1600) detct(idetct) * 180./pi, tab, Wall(idetct), tab,
1 change, tab, augomg, tab, Wall(idetct) / augomg, tab, volume(idetct)
if (change .gt. convrg) then
   preWall(idetct) = Wall(idetct)
   nx = nx * 2+1
   ny = ny * 2+1
   nz = nz * 2+1
   goto 400
endif
1500 continue
1600 format(g12.4, 4(a1, g12.4))
stop
end

function dc(x, alpha, dd, ro)
implicit none
real*8 dc, x, alpha, dd, ro

This function calculates the diameter of the acceptance cone as a function of the distance from the detector.
dc=2.*((x+ro)*tan(alpha/2.)-dd/2.)
return
end

Mie Code by Bohren and Huffman, 1983
This subroutine was validated through comparison with published results of Mie calculations. See U.J. Wiscombe, "Mie scattering calculations: advances in technique and fast, vector-speed computer codes," NCAR/TH-140+STR, National Center for Atmospheric Research, Boulder, CO.

subroutine bhmie(dia,theta,refre,refim,intens,ntheta,wavel,waven)
dimension amu(1801),theta(1801),pi(1801),tau(1801),pio(1801),
lpio(1801),ipar(1801),iper(1801),intens(1801)
complex d(6000),y,refrel,xi,xi0,xi1,an,bn,s1(3602),s2(3602)
real*8 psi0,psi1,psi,dn,dx
real*8 ipar,iper,intens,dia,theta,refre,refim,wavel,waven
refmed=1.0
refrel=cmplx(refre,refim)/refmed
pie=4.*atan(1.)

calculating the mie intensity functions

do 100 itheta=1,ntheta
   ipar(itheta)=0.0
   iper(itheta)=0.0
100 continue
x=pie*dia/wave I
dx=x
y=x*refrel
xstop=x+4.*x**.3333+2.
nstop=xstop
ymod=cabs(y)
nmx=amax1(xstop,ymod)+15

do 200 j=1,ntheta
   amu(j)=dcos(theta(j))
200 continue
d(nmx)=cmplx(0.,0.)
nn=nmx-1

do 300 n=1,nn
   rn=nn+1
   d(nmx-n)=(rn/y)-(1./((nmx+n+1)+rn/y))
300 continue

do 400 j=1,ntheta
pi0(j)=0.
pi1(j)=1.

400 continue

nn=2*ntheta-1

do 500 j=1,nn
   a1(j)=cmplx(0.,0.)
   a2(j)=cmplx(0.,0.)
500 continue

psi0=dcos(dx)
psi1=dsin(dx)
chi0=-sin(x)
chi1=cos(x)
apsi0=psi0
apsi1=psi1
x10=cmplx(apsi0,-chi0)
xi1=cmplx(apsi1,-chi1)
qsc=0.0
n=1

600 do n=1,n
   dn=n
   rn=n
   fn=(2.*rn+1.)/(rn*(rn+1.))
   gn=(rn*(rn+2.))/(rn+1.)
   psi=(2.*dn-1.)*psi1/dx-psi0
   apsi=psi
   chi=(2.*rn-1.)*chi1/x-chi0
   xi=cmplx(apsi,-chi)
   an=(d(n)/refrel+rn/x)*apsi-apsi1
   an=an/((d(n)/refrel+rn/x)*xi-xi1)
   bn=(refrel*d(n)+rn/x)*apsi1-apsi
   bn=bn/((refrel*d(n)+rn/x)*xi-xi1)
   qsc=qsc+(2.*rn+1.)*(cabs(an)*cabs(an)+cabs(bn)*cabs(bn))
700 continue

psi0=psi1
psi1=psi
apsi1=psi1
chi0=chi1
chi1=chi
xi1=cmplx(apsi1,-chi1)
n=n+1
rn=n

do 800 j=1,ntheta

176
pi(l(j))=((2.*rn-1.)/(rn-1.))*pi(j)*amu(j)
pi(l(j))=pi(l(j))-rn*pi(0(j))/(rn-1.)
pi(0(j))=pi(j)

800 continue

if (n-1-nstop) 600,900,900

900 continue

c

c calculating the Mie intensity functions for each scattering angle

do 1000 itheta=1,ntheta

ipar(itheta)=cabs(s2(itheta))**2
iper(itheta)=cabs(s1(itheta))**2
intens(itheta)=(iper(itheta)+ipar(itheta))/2./waven**2

1000 continue
return
end

c
This subroutine calculates the scattering volume for regular
operation

subroutine vcal(ndetct,db,dj,volume)

implicit none

real*8 db,dj,volume,dz,prevol,x,y,z,psi,change

integer ndetct,iz,nz

prevol=0.
nz=5

100 continue

volume=0.
if(nz.ne.1)then
  dz=db/2./float(nz-1)
elself
  dz=0.
endif

do 200 iz=1,nz
  z=float(iz-1)*dz
  y=dsqrt(dj**2/4.-z**2)
  psi=dacos(2.*z/db)
  x=db*dsin(psi)
  volume=x*y+volume

200 continue

volume=4.*volume*dz
change=abs(volume-prevol)/volume
if(change.gt.0.01)then
  prevol=volume
  nz=2*nz+1
  goto 100
endif
This subroutine calculates the scattering volume when the nephelometer is operating in calibration mode.

Subroutine vccal(alpha, dd, db, ro, thetad, volume)

Implicit None

Real*8 alpha, dd, ro, thetad, volume, ymax, ymin, xp, yp, hcone, hbeam, h,
prevol, xchange, a, b, c, dx, db, dy, xd, dc, yd, arg

Integer nx, ny, ix, iy

prevol = 0.

nx = 5

ny = 11

a = tan(alpha/2.)*dsin(thetad) - dcos(thetad)
b = (ro*tan(alpha/2.) - dd/2.)*dsin(thetad)
c = - tan(alpha/2.)*dsin(thetad) - dcos(thetad)

100 Continue

If nx .ne. 1 then

dx = db/float(nx-1)

Else

dx = 0.

End If

Volume = 0.

Do 300 ix = 1, nx

If nx .ne. 1 then

xp = - db/2. + float(ix-1)*dx

Else

xp = 0.

End If

Ymax = (xp*a*dsin(thetad) + b)/(1. + a*dcos(thetad))

Ymin = (xp*c*dsin(thetad) - b)/(1. + c*dcos(thetad))

If ny .ne. 1 then

dy = (ymin - ymin)/float(ny-1)

Else

dy = 0.

End If

Do 200 iy = 1, ny

If ny .ne. 1 then

yp = ym + float(iy-1)*dy

Else

yp = 0.

End If

Xd = dsin(thetad)*xp - dcos(thetad)*yp

Yd = dcos(thetad)*xp + dsin(thetad)*yp

Arg = dc(xd, alpha, dd, ro)**2/4. - yd**2

If(arg .lt. 0.d0) arg = 0.d0

Hcone = sqrt(arg)

Arg = db**2/4. - xp**2

If(arg .lt. 0.d0) arg = 0.d0

Hbeam = sqrt(arg)

H = min(hcone, hbeam)
if(nx.eq.1.and.ny.ne.1) then
    volume = h*dy*volume
elseif(ny.eq.1.and.nx.ne.1) then
    volume = h*dx*(ymax-ym) + volume
elseif(ny.eq.1.and.nx.eq.1) then
    volume = h*dy*(ymax-ym) + volume
else
    volume = h*dx*dy + volume
endif

200 continue
300 continue
volume = 2.*volume
vchange = abs(volume-prevol)/volume
print*, volume, vchange
if(vchange.gt.0.001) then
    prevol = volume
    nx = 2*nx+1
    ny = 2*ny+1
    goto 100
endif

return
end
Vita

Matthew R. Jones was born in San Jose, California on January 18, 1964 to Jerold W. and Nancy S. Jones. His family moved from California to Salt Lake City, Utah, and then to Columbus, Ohio. In 1973, his family settled in Austin, Texas. He graduated from L. C. Anderson High School in June, 1982. Matthew received a Trustee's scholarship from Brigham Young University where he studied for a year before serving a mission for the Church of Jesus Christ of Latter-Day Saints in Tokyo, Japan. After returning from Japan, he continued his studies at Brigham Young University. From June to December of 1986, he went to London, England on a study abroad program. During this time, he had the opportunity to travel throughout Europe and to visit Russia and Israel. He graduated magna cum laude with a B. S. in Mechanical Engineering in April, 1988. Matthew began graduate school at the University of Illinois at Urbana-Champaign in August, 1988. While a student at Illinois, he worked as a research assistant and received a Chevron Graduate Fellowship Award and a Dupont University Science & Engineering Grant. He received a M. S. in Mechanical Engineering in August, 1990. From April, 1991 to March, 1993, he work as a research associate in the Engineering Physics Division of Argonne National Laboratory while completing the research for his Ph.D. Matthew is a member of the American Society of Mechanical Engineers, Tau Beta Pi, and Phi Kappa Phi.