EFFECT OF SEPARATION DISTANCE ON THE OPTICAL PROPERTIES 
OF DENSE DIELECTRIC PARTICLE SUSPENSIONS

by

IACOVOS A. VASALOS

Diploma of Chem. Eng., Technical University of Athens 
(1963)
S. M., Massachusetts Institute of Technology 
(1967)

Submitted in Partial Fulfillment 
of the Requirements for the 
Degree of Doctor of Philosophy 
at the 
Massachusetts Institute of Technology 
August 1969

Signature of Author

Department of Chemical Engineering

Certified by 

Thesis/Supervisors

Accepted by 

G. C. Williams, Chairman Dept.Comm. 
on Graduate Theses

Archives 

SEP 15 1969 
M.I.T. 
LIBRARIES
ABSTRACT

EFFECT OF SEPARATION DISTANCE ON THE OPTICAL PROPERTIES
OF DENSE DIELECTRIC PARTICLE SUSPENSIONS

by

IACOVOS A. VASALOS

Submitted to the Department of Chemical Engineering in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

In the solution of the transport equation for scattering media it is usually assumed that the particles scatter independently, and that polarization effects may be neglected. The validity of these assumptions has been here tested by measurement of the bidirectional reflectance and transmittance of suspensions of monodisperse polystyrene particles confined between two parallel glass slides, followed by comparison of the results with the values predicted from theory. For the range of conditions studied - particle sizes of 0.106, 0.248, and 0.530μ, wavelengths of 0.436μ and 0.546μ, and optical thicknesses of 0.25 to 3000 - the agreement between theory and experiment is excellent, provided that the clearance between particles, assumed uniformly distributed, exceeds 0.33 wavelengths and the ratio of clearance to diameter exceeds 0.4.

For those cases of high pigment volume concentrations (up to 30%) in which the average clearance c between particles fell below 0.3/wavelengths, the effective scattering eff. X_se was found to be smaller than the value X_s obtained from the solution of the Mie equations. The ratio X_s/X_se, for the three particle sizes studied, could be correlated by the relation given below:

\[ \log \log \frac{X_s}{X_{se}} = -5.1 \frac{c}{\lambda} + 0.25 \]
This correlation extends the range of application of the equation of transfer and allows one to find the optimum conditions for maximum hiding power in the case of monodisperse systems. Additional studies are recommended for polydisperse systems.

Thesis Supervisors:

Hoyt C. Hottel
Professor of Chemical Engineering

Adel F. Sarofim
Associate Professor of Chemical Engineering

William H. Dalzell
Assistant Professor of Chemical Engineering
To my father's memory
ACKNOWLEDGEMENTS

The author wishes to thank Professors H. C. Hottel and A. F. Sarofim for the many helpful discussions during the course of this work.

The encouragement and moral support offered by Professor A. F. Sarofim in numerous occasions will long be remembered.

The many helpful ideas suggested by Professor W. H. Dalzell during the early stages of this work are sincerely appreciated.

The author is also indebted to Dr. W. Richards of Cabot Corporation for his suggestions and interest in this work.

I would wish to express my appreciation to all those who have helped me during this undertaking. The encouragement and advice of Dr. A. Witt and Mr. E. Matulevicius are largely responsible for my attempting to do doctorate work.


The author wishes to thank D. Swanson for his able assistance in obtaining part of the data, S. Mitchell for producing the electromicrographs and Mrs. E. Kehoe for her patience in typing the thesis manuscript.

The author would like to acknowledge the financial support provided by the National Science Foundation and the Godfrey L. Cabot Solar Energy Fund of M.I.T. Services of the MIT Information Processing Center are also noted.
# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>I. SUMMARY</td>
<td>1</td>
</tr>
<tr>
<td>II. INTRODUCTION</td>
<td>11</td>
</tr>
<tr>
<td>III. SINGLE SCATTERING THEORY</td>
<td>33</td>
</tr>
<tr>
<td>3.1 Physical description of a single scattering process</td>
<td>33</td>
</tr>
<tr>
<td>3.2 Mie theory</td>
<td>36</td>
</tr>
<tr>
<td>3.3 More definitions</td>
<td>37</td>
</tr>
<tr>
<td>IV. MULTIPLE SCATTER</td>
<td>41</td>
</tr>
<tr>
<td>4.1 The specific intensity</td>
<td>41</td>
</tr>
<tr>
<td>4.2 Geometrical system</td>
<td>42</td>
</tr>
<tr>
<td>4.3 Optical thickness</td>
<td>44</td>
</tr>
<tr>
<td>4.4 Equation of transfer</td>
<td>44</td>
</tr>
<tr>
<td>4.4.1 Photon particle interactions</td>
<td>44</td>
</tr>
<tr>
<td>4.4.2 Mathematical formulation</td>
<td>45</td>
</tr>
<tr>
<td>4.4.2.1 Non-reflecting boundaries</td>
<td>46</td>
</tr>
<tr>
<td>4.4.2.2 Reflecting boundary conditions</td>
<td>48</td>
</tr>
<tr>
<td>4.4.2.2.1 One step change in refractive index</td>
<td>48</td>
</tr>
<tr>
<td>4.4.2.2.2 Two step change in refractive index</td>
<td>52</td>
</tr>
<tr>
<td>4.5 Numerical results of multiple scatter calculations</td>
<td>53</td>
</tr>
<tr>
<td>4.6 Effect of phase function on the angular distribution of multiple scatter radiation</td>
<td>55</td>
</tr>
<tr>
<td>4.7 Simplified models</td>
<td>58</td>
</tr>
<tr>
<td>4.7.1 Evaluation of two flux model</td>
<td>60</td>
</tr>
<tr>
<td>V. EXPERIMENTAL APPARATUS AND PROCEDURE</td>
<td>65</td>
</tr>
<tr>
<td>5.1 Apparatus</td>
<td>65</td>
</tr>
<tr>
<td>5.2 Procedure</td>
<td>69</td>
</tr>
<tr>
<td>5.3 Samples</td>
<td>70</td>
</tr>
<tr>
<td>VI. RESULTS</td>
<td>73</td>
</tr>
<tr>
<td>6.1 Single scatter</td>
<td>73</td>
</tr>
<tr>
<td>6.2 Multiple scatter</td>
<td>78</td>
</tr>
</tbody>
</table>
TABLE OF CONTENTS (Continued)

VII. DISCUSSION OF RESULTS                                                                 95
    7.1 Single scatter 95
    7.2 Multiple scatter – No interference present 100
       7.2.1 Polarization effects 101
       7.2.2 Comparison of theory and experiment 110
    7.3 Multiple scatter – Interference effect 111
    7.4 Practical applications 127

IX. CONCLUSIONS                                                                            134

IX. RECOMMENDATIONS                                                                        136

APPENDICES

A. Description of the Phase Function by a Legendre Polynomial Series 137
B. Development and Solution of the Equation of Transfer 145
C. Simplified Representation of Mie Scattering Diagram (Phase Function) 164
D. The b Factor for Different Values of x, m and Matrix Refractive Index 169
E. Details of Apparatus 176
F. Data 182
G. Errors in Multiple Scatter Experiments 207

REFERENCES 212

BIOGRAPHICAL NOTE 219
# LIST OF FIGURES

<table>
<thead>
<tr>
<th>Figure</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.1</td>
<td>Scattering of Light by white paint</td>
<td>13</td>
</tr>
<tr>
<td>3.1</td>
<td>Single scattering geometry</td>
<td>34</td>
</tr>
<tr>
<td>4.1</td>
<td>Geometrical representation</td>
<td>43</td>
</tr>
<tr>
<td>4.2</td>
<td>The relation between the scattering angle and the coordinate angles $\psi$, $\psi'$, $\theta$ and $\theta'$</td>
<td>43</td>
</tr>
<tr>
<td>4.3</td>
<td>Trace of unscattered component of incident beam</td>
<td>50</td>
</tr>
<tr>
<td>4.4</td>
<td>Bidirectional transmittance and reflectance of a suspension of pure scatterers in a layer of optical density one, with incident beam 60.2° off normal. Comparison of results obtained by Bellman with calculations performed with present method.</td>
<td>54</td>
</tr>
<tr>
<td>4.5</td>
<td>Replacement of the exact phase function ($x=4$, $m=2.7$) with one having the same peakedness and forward fraction</td>
<td>56</td>
</tr>
<tr>
<td>4.6</td>
<td>Comparison of the angular distribution of transmitted and reflected radiation for two phase functions having the same $f$ and $P_e$</td>
<td>57</td>
</tr>
<tr>
<td>4.7</td>
<td>The two flux model for different values of $x$, $m$ and $n$</td>
<td>62</td>
</tr>
<tr>
<td>5.1</td>
<td>Schematic of apparatus used for scattering measurements</td>
<td>66</td>
</tr>
<tr>
<td>6.1</td>
<td>Electron micrograph of polystyrene spheres $d=0.530\mu$ and $d=0.106\mu$</td>
<td>74</td>
</tr>
<tr>
<td>6.2</td>
<td>Electron micrograph of polystyrene spheres $d=0.248\mu$</td>
<td>75</td>
</tr>
<tr>
<td>6.3</td>
<td>Mie scattering coefficients, $i_{m}(\theta)$ and $i_{w}(\theta)$: $d=0.106\mu$, $n_p/n_w=1.19$. Data points represent experimental results, solid lines theory.</td>
<td>76</td>
</tr>
<tr>
<td>Figure</td>
<td>Title</td>
<td></td>
</tr>
<tr>
<td>--------</td>
<td>-------</td>
<td></td>
</tr>
<tr>
<td>6.4</td>
<td>Mie scattering coefficients, $i_m(\theta)$ and $i_n(\theta)$: $d=0.530\mu$. Data points represent experimental results, solid lines theory.</td>
<td></td>
</tr>
<tr>
<td>6.5</td>
<td>Bidirectional reflectance and transmittance as a function of PVC of 0.106$\mu$ polystyrene spheres, $L=0.147$ cms. Data points represent experimental results, solid lines theory.</td>
<td></td>
</tr>
<tr>
<td>6.6</td>
<td>Bidirectional reflectance and transmittance as a function of PVC of 0.530$\mu$ polystyrene spheres, $L=0.147$ cms. Data points represent experimental results, solid lines theory.</td>
<td></td>
</tr>
<tr>
<td>6.7</td>
<td>Bidirectional reflectance and transmittance as a function of PVC of 0.120$\mu$ polystyrene spheres. Data points represent experimental results, drawn lines theory.</td>
<td></td>
</tr>
<tr>
<td>6.8</td>
<td>Integrated diffuse reflectance and transmittance as a function of optical thickness for a polystyrene latex 0.530$\mu$ in diameter and a $\lambda=0.4358\mu$. Data points experimental solid lines theory.</td>
<td></td>
</tr>
<tr>
<td>6.9</td>
<td>Integrated diffuse reflectance and transmittance as a function of optical thickness: $d=0.530\mu$, $\lambda=0.5461\mu$. Data points experimental solid lines theoretical.</td>
<td></td>
</tr>
<tr>
<td>7.1</td>
<td>Effect of particle diameter on Mie intensity functions.</td>
<td></td>
</tr>
<tr>
<td>7.2</td>
<td>Components of polarization of bidirectional reflectance and transmittance of polystyrene latex spheres; $L=0.147$ cms. Data points represent experimental values, solid lines theory.</td>
<td></td>
</tr>
<tr>
<td>7.3</td>
<td>Components of polarization of bidirectional reflectance and transmittance of polystyrene spheres; $L=0.147$ cms, $\lambda=0.436\mu$. Data points represent experimental values, solid lines theory.</td>
<td></td>
</tr>
<tr>
<td>Figure</td>
<td>Title</td>
<td>Page</td>
</tr>
<tr>
<td>--------</td>
<td>--------------------------------------------------------------------------------------------------------------------------------------------</td>
<td>------</td>
</tr>
<tr>
<td>7.4</td>
<td>Components of polarization of bidirectional reflectance and transmittance of polystyrene latex spheres; upper set of curves; L=0.147 cms, ( \lambda = 0.436 \mu ); Data points represent experimental values, solid lines theory.</td>
<td>107</td>
</tr>
<tr>
<td>7.5</td>
<td>Reflectivity of water-glass-air interface as a function of angle ( \theta ) (the angle to the normal, forward).</td>
<td>108</td>
</tr>
<tr>
<td>7.6</td>
<td>Bidirectional transmittance and reflectance calculated with (solid line) and without (dashed line) allowance for polarization; Rayleigh scatterers, ( \mu_o = 1.0 ), ( \tau_1 = 1.0 )</td>
<td>109</td>
</tr>
<tr>
<td>7.7</td>
<td>Comparison of theory with measurements on polystyrene spheres 0.530( \mu ) in diameter. Continuous lines are based on multiple scatter of non-interfering particles</td>
<td>117</td>
</tr>
<tr>
<td>7.8</td>
<td>Comparison of theory with measurements on polystyrene spheres 0.248( \mu ) in diameter. Continuous lines are based on multiple scatter of non-interfering particles</td>
<td>118</td>
</tr>
<tr>
<td>7.9</td>
<td>Comparison of theory with experiments on polystyrene spheres 0.102( \mu ) in diameter. Continuous lines are based on multiple scatter of non-interfering particles</td>
<td>119</td>
</tr>
<tr>
<td>7.10</td>
<td>Variation of scattering efficiency with c/( \lambda )</td>
<td>123</td>
</tr>
<tr>
<td>7.11</td>
<td>( X_s/X_{se} ) correlation plot</td>
<td>125</td>
</tr>
<tr>
<td>7.12</td>
<td>Variation of total backscattering per unit volume of polystyrene latex as a function of PVC. The solid lines represent theory, data points experimental</td>
<td>129</td>
</tr>
<tr>
<td>7.13</td>
<td>Variation of total backscattering per unit volume of polystyrene latex with PVC. Solid lines calculated assuming independent scatter, dotted lines experimental</td>
<td>130</td>
</tr>
<tr>
<td>Figure</td>
<td>Title</td>
<td>Page</td>
</tr>
<tr>
<td>--------</td>
<td>----------------------------------------------------------------------</td>
<td>------</td>
</tr>
<tr>
<td>7.14</td>
<td>Variation in backscattering coefficient per unit volume of TiO₂ paint film as a function of PVC. Refractive index of vehicle 1.5; ( \lambda = 0.55 \mu )</td>
<td>132</td>
</tr>
<tr>
<td>A.1</td>
<td>Program flow chart for Legendre polynomials coefficients</td>
<td>141</td>
</tr>
<tr>
<td>B.1</td>
<td>Program flow chart for transfer equation</td>
<td>153</td>
</tr>
<tr>
<td>E.1</td>
<td>Average spectral distribution from a GE B-H6 mercury arc lamp</td>
<td>179</td>
</tr>
<tr>
<td>E.2</td>
<td>Spectral sensitivity characteristic of an RCA 931-A multiplier phototube for equal values of radiant flux at all wavelengths</td>
<td>180</td>
</tr>
<tr>
<td>E.3</td>
<td>Arrangement of electronic equipment for light scattering apparatus</td>
<td>181</td>
</tr>
<tr>
<td>F.1</td>
<td>Optical system for measuring the incident light intensity</td>
<td>182</td>
</tr>
<tr>
<td>F.2</td>
<td>Traces of contributions to reflectance and transmittance by beams that have been scattered more than once</td>
<td>192</td>
</tr>
<tr>
<td>G.1</td>
<td>Bidirectional reflectance and transmittance of polystyrene spheres; upper curves: ( L = 0.0508 ) cm; lower curves: ( L = 0.1473 ) cm. Data points represent experimental results, solid lines theory.</td>
<td>211</td>
</tr>
</tbody>
</table>
# LIST OF TABLES

<table>
<thead>
<tr>
<th>Table</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.1</td>
<td>Refractive index of polystyrene</td>
<td>71</td>
</tr>
<tr>
<td>5.2</td>
<td>Range of variables studied</td>
<td>71</td>
</tr>
<tr>
<td>6.1-6.8</td>
<td>Tabulation of experimental results</td>
<td>86-94</td>
</tr>
<tr>
<td>7.1</td>
<td>Variation in $n_w \pi d/\lambda$</td>
<td>97</td>
</tr>
<tr>
<td></td>
<td>Normalized hemispherical diffuse reflectance and transmittance for polystyrene latex spheres 0.530$\mu$m in diameter</td>
<td></td>
</tr>
<tr>
<td>7.2</td>
<td>$\lambda=0.4358\mu$m</td>
<td>113</td>
</tr>
<tr>
<td>7.3</td>
<td>$\lambda=0.5461\mu$m</td>
<td>113</td>
</tr>
<tr>
<td>7.4</td>
<td>$X_s/X_{se}$ as a function of $c/\lambda$ for $d=0.102\mu$m</td>
<td>121</td>
</tr>
<tr>
<td>7.5</td>
<td>$X_s/X_{se}$ as a function of $c/\lambda$ for $d=0.248\mu$m</td>
<td>121</td>
</tr>
<tr>
<td>A.1</td>
<td>Comparison of Legendre polynomial coefficients of this study with Churchill's values</td>
<td>139</td>
</tr>
<tr>
<td>A.2</td>
<td>Comparison of exact phase function values with series approximation</td>
<td>140</td>
</tr>
<tr>
<td>Appendix</td>
<td></td>
<td></td>
</tr>
<tr>
<td>D</td>
<td>The b factor for different values of size parameter, refractive index and boundary conditions in case of conservative scattering</td>
<td>169-175</td>
</tr>
<tr>
<td>F.1-F.20</td>
<td>Tabulation of the measured bidirectional intensity function for different set of conditions</td>
<td>197-206</td>
</tr>
<tr>
<td>G.1</td>
<td>A comparison of the theoretical and experimental results for a particle size 0.530$\mu$m and a wavelength 0.4358$\mu$m.</td>
<td>210</td>
</tr>
</tbody>
</table>
NOTATION

\( d \) particle diameter
\( e(\theta) \) measure voltage at angle \( \theta \)
\( i_\perp \) light-scattering function for perpendicular polarization
\( i_\parallel \) light-scattering function for parallel polarization
\( k \) wave number, defined as \( 2\pi/\lambda \)
\( L \) optical path through the particles suspension or number of terms in Legendre polynomial series
\( m \) relative index of refraction of the particles with respect to surrounding medium
\( n \) refractive index of medium relative to vacuum, \( n_F = \)
refractive index of particle, \( n_w = \) refractive index of aqueous dispersed phase
\( r \) distance from the scattering particles to the point of measurement of the scattered-light intensity
\( s \) sensitivity of the multiplier phototube system
\( x \) size parameter, defined as \( \pi d/\lambda \)
\( y \) product of \( x \) and \( m \)
\( a_K \) weighting factor in Legendre polynomial series
\( \chi_s \) scatter efficiency
\( I(\tau,\mu) \) intensity at position \( \tau \) and in direction \( \mu \)
\( N \) number of abscissas used in integration formula
\( N_P \) number of particles per unit volume
\( P(\Theta) \) phase function, normalized intensity distribution for single scatter
$P_K(\mu)$ kth Legendre polynomial

PVC percent volume concentration of particles

$R$ hemispherical reflectance, $R_D$ = diffuse component of hemispherical reflectance

$T$ hemispherical transmittance, $T_D$ = diffuse component of hemispherical transmittance

c clearance between particles

$f$ fraction of radiation into forward direction

$b$ fraction of radiation into backward direction

$\theta$ angle between scattered and incident beam, polar angle

$\Theta$ angle between reference beam and in scattered beam

$\lambda$ wavelength measured in vacuo

$\rho$ reflectance of water-glass-air interface system,

$\rho_1$ = reflectivity of water-glass interface,

$\rho_2$ = reflectivity of glass-air interface

$\mu$ cosine of polar angle

$\tau$ optical depth in mean free paths, $\tau_1 = K_s L$

$\Omega$ solid angle

$\delta$ center-to-center distance between particles

$\omega$ albedo for single scatter
I. SUMMARY

Interests in problems as diverse as the conductance of powder or fibrous insulators, the reflectance of pigmented surface coatings, and the development of solar energy collectors with special radiative properties has generated an enormous literature on multiple scatter phenomena. Most of this work is either devoted to the mathematics of solving the transport equation and comparing rigorous solutions with approximations or making experimental measurements in order to find when interference effects become appreciable, without giving a quantitative description of these effects.

This work presents studies on the radiative properties of unidimensional suspensions of non-absorbing but scattering spheres having perimeter-to-wavelength ratios in the range of practical interest and dispersed in solutions in concentrations embracing both multiple scatter and interference phenomena.

The radiative properties of interest for non-absorbing particle systems, such as TiO₂ surface coatings, are the reflectance and transmittance. The angular distribution of the emerging intensities from the bounded surfaces of a unidimensional non-absorbing TiO₂ surface coating depends on the following parameters: particle size distribution, wavelengths of illumination, film thickness, refractive indices of particles and vehicle matrix and clearance between
the particles. Multiple scatter and interference phenomena depend on all parameters listed above. Since it is impossible to include all variables in a comprehensive study it was decided to choose a well defined system of polystyrene spherical particles, since these particles have a known refractive index and are manufactured as a monodisperse latex. Measurements of the angular distribution of reflected and transmitted intensities from polystyrene particles contained in a unidimensional cell for various particle sizes, wavelengths and clearances between particles will provide checks on the validity of the existing multiple scatter theories, the range of their applicability, and the effect of interference.

The polystyrene particles, supplied by Dow Chemical Company in volume concentrations of 0.10 to 0.30, were diluted to the desired level with de-ionized millipore distilled water and the resultant solution was placed in a rectangular cell constructed from parallel microscope slides cemented with epoxy resin to a lucite frame. The cell faces were 5 cms square and the clearance between the inner glass surfaces was varied from 0.05 to 0.20 cms. The range of variables studies is tabulated below:
<table>
<thead>
<tr>
<th>Particle Diameter (μ)</th>
<th>Wavelength (μ)</th>
<th>PVC</th>
<th>Optical Thickness</th>
</tr>
</thead>
<tbody>
<tr>
<td>.102±.003</td>
<td>.436</td>
<td>.0284</td>
<td>.2893</td>
</tr>
<tr>
<td>.102±.003</td>
<td>.546</td>
<td>.0284</td>
<td>.2893</td>
</tr>
<tr>
<td>.106±.003</td>
<td>.436</td>
<td>3.53×10⁻⁵</td>
<td>.10</td>
</tr>
<tr>
<td>.106±.003</td>
<td>.546</td>
<td>3.08×10⁻⁴</td>
<td>.10</td>
</tr>
<tr>
<td>.248±.003</td>
<td>.436</td>
<td>.105</td>
<td>.293</td>
</tr>
<tr>
<td>.248±.003</td>
<td>.546</td>
<td>.105</td>
<td>.293</td>
</tr>
<tr>
<td>.530±.003</td>
<td>.436</td>
<td>1.31×10⁻⁶</td>
<td>.295</td>
</tr>
<tr>
<td>.530±.003</td>
<td>.546</td>
<td>5.2×10⁻⁵</td>
<td>.295</td>
</tr>
</tbody>
</table>

The bidirectional reflectance and transmittance were measured for the case of normal incidence only. The scattered radiation in this study is independent of azimuthal angle, and its directional distribution can be obtained by a traverse in a single plane. The equipment consisted of (i) a mercury arc and optics to produce an unpolarized beam of 0.76 degrees divergence and 0.12 cms image diameter at the sample cell, (ii) a RCA 931A photomultiplier, and (iii) optics to confine the received beam to 0.66 degree divergence angle. The state of polarization of the scattered radiation was measured by use of a Polaroid HN22 polarizer. The area viewed by the collector along the normal was 1.6 cms in diameter; it was made larger than the irradiated area in order to collect all the energy scattered in a given direction.
The results obtained are discussed in the following sequence:

(i) Single scatter measurements
(ii) Multiple scatter measurements in the absence of interference
(iii) Experimental data in close-packed systems

(i) Scatter intensities may be calculated from Mie theory provided that the attenuation of the scattered light is less than 10 percent. With \( \pi d/\lambda - d \) particle diameter, \( \lambda \) wavelength of illumination - and relative refractive index \( m \) of the particles with respect to surrounding medium known, the Mie equations yield the angular distribution of the energy scattered around a particle and the scattering coefficient \( X_s \) (defined as the ratio of the energy extinguished to that intercepted by the particle geometrical cross section). When \( N_p \) particles are present the resulting intensities are \( N_p \)-fold that form a single particle.

Measurements from very dilute polystyrene lattices were performed for two reasons: First in order to get some confidence in the equipment and second to check the particle diameters in the particle suspensions as received. Discrepancies of 3 to 6 percent have been observed between the sizes reported by the manufacturer and the sizes required to match the experimental intensity distributions with
those calculated from the Mie equations. However, no differences were found between the effective scattering diameter of particles so measured and those obtained from electron micrographs of the particles.

Comparison of calculated and measured intensities for single scatter showed that the agreement between theory and experiment is good, the small discrepancies resulting from a combination of the following factors: (a) the spread in particle sizes, (b) the finite bandwidth of the monochromatic filter (65 Å at half peak transmission), (c) finite source (0.76°) and collector (0.66°) angles, (d) any difference in the effective diameter for scattering and that observed in electron micrographs, (e) spurious scatter introduced by the apparatus, which becomes significant in the angular region in which the scattered intensities are very low. Estimation of the errors introduced by the different factors indicates that the spread in particle sizes (factor a) is largely responsible in this study for the deviations between theory and experiments.

(ii) Multiple scatter experimental data were compared with the solution of transfer equation. The equation of transfer, which describes the multiple scatter process in a suspension of particles, was solved by the method of discrete ordinates (discrete conical sheet for polar symmetry) modified to allow for the double refraction and multiple
reflection occurring at the glass surfaces of the test cell. The solution, which gives the intensity at the bounding surfaces and at any depth within the slab, depends on the following parameters: \( \pi d/(\lambda/n_w) \), where \( (\lambda/n_w) \) is the wavelength in the aqueous medium, \( m \) the relative refractive index of the particle with respect to surrounded medium and on the optical thickness \( \tau_1(=K_sL) \), where \( K_s \) is the scattering coefficient per unit volume and \( L \) is the slab thickness.

The validity of the final solutions are qualified by the introduction in the analysis of many assumptions. Most of them were satisfied by the design of the experiments. The assumptions to be tested by experiment were:

(a) that polarization effects are negligible

(b) that the particles in dilute and moderately concentrated suspensions act as independent scatterers.

During this phase of the work the slab thickness was kept constant. For fixed particle size and wavelength the optical thickness was increased by increasing the pigment volume concentration (PVC).

Measurements of the perpendicular and parallel component showed that polarization effects are negligible for optical thicknesses larger than 1.0. Any difference between the two components is attributed to the difference in the interface reflectance for perpendicular and parallel polarization. In case of small optical thicknesses
(<1.0) the differences observed between the two components are not severe even in the case of 0.106μ particles.

The above results motivated comparison of experimental data with theoretical values based on the assumption that polarization effects may be ignored and that the particles scatter independently. Comparisons of calculations and experiment were made for 0.102μ, 0.106μ, 0.248μ, and 0.530μ particles. Excellent agreement has been found between theory and experiment for all levels of optical thickness, provided that the ratio of the clearance and the wavelength (c/λ) is greater than 1/3.

However, when c/λ is less than 1/3 the equation of transfer was found inadequate to describe the radiative properties. Large discrepancies were observed which could be attributed to either a change in the scattering diagram of a particle or a change in scattering coefficient X_s as a result of interference or both.

A theoretical study showed that the angular distribution of emerging intensities from the surfaces of the slab are insensitive to the exact shape of the phase diagram provided that the forward scattered fraction is preserved. Consequently no effort was made to find an effective phase function and all changes were attributed to the change in scattering coefficient.
The scattering coefficient has been found by using the two flux model which in case of non-absorbing particles contained between two glass slides relates the total transmittance $T$ and the optical thickness $\tau_1$ in the following form:

$$\frac{1}{T} = 1.09 + b \, \tau_1 \quad (1-1)$$

where $b$ is the backwards scattered fraction.

A study was undertaken to test the validity of the two flux model. Inverse transmittances calculated using the transfer equation were plotted as a function of $\tau_1$. It was found that the $b$ factor was constant whenever $\tau_1 < 0.1$ and $\tau_1 > 100$. In the intermediate range the two flux model was not valid.

Based on the two flux model, experiments were performed at three to four different large values of $\tau_1$ at a fixed PVC. Inverse experimental transmittances were plotted as a function of $\tau_1$. Equation (1-1) was obeyed even though $c/\lambda < 1/3$. As a consequence of this it was concluded that for constant $c/\lambda$ ratio the interference effects were independent of $\tau_1$. This enabled the search for an effective scattering coefficient $X_{se}$ such that a plot of $1/T$ versus $\tau_1$ (based on $X_{se}$) brings theoretical and experimental lines together.

The effective scattering coefficient $X_{se}$ was compared to the one for isolated scatterers. Calculation of the $X_s/X_{se}$ ratio led to the conclusion that no interference
exists for 0.530μ polystyrene particle up to δ/d=1.36. (δ, center-to-center distance between two particles; calculated assuming particles distributed in a rhombohedral array.) For the same range of δ/d values but a small particle size (d=0.102μ) the ratio X_s/Xse was much larger than one. This important finding enabled us to conclude that in the range studied δ/d is not an important parameter. Interference results as a consequence of small values of c/λ.

The proposed correlation which describes the decrease in the scattering coefficient as a function of c/λ is:

\[ \log \log \frac{X_s}{X_{se}} = -5.1 \frac{C}{\lambda} + 0.25 \] (1-2)

The practical implications of the above equation were investigated by calculating the back-scattering coefficient per unit volume S(=bK_s) of a TiO₂ pigment suspended in a medium of 1.5 as a refractive index. The film was illuminated with light of a wavelength of 0.55μ. The constant S was calculated as a function of PVC for three different particle sizes (0.16, 0.22, and 0.28μ), using b factors derived from the transfer equation and particle scattering efficiencies derived from Mie theory and equation 1.2.

A plot of S versus PVC showed the following features:
(a) The hiding power per unit volume (S) is proportional to PVC over the concentration range in which single scatter prevails. Departures from linearity occurs at lower PVC's for smaller particles.
(b) For each particle size there is an optimum PVC which yields the maximum S. The optimum value of PVC moves to higher values with increasing diameter.

(c) At low PVC's (up to 0.18) a particle size of 0.22μ gives the highest S.

It was thus concluded that the radiative properties of particle suspensions in which the clearance between particles, assumed uniformly distributed exceeds 0.3 wavelengths, and in which the ratio of clearance to diameter exceeds 0.4 may be calculated with confidence from theory. For the cases in which the above criteria were not satisfied an effective scattering efficiency correlation was derived as a function of the clearance to wavelength ratio. Solutions to the equation of transfer based on the effective scattering coefficient that can be calculated using this correction factor yield a good description of radiative properties of particle suspensions.

It is recommended that additional work be carried out in order to test the validity of the correction factor for interference in systems with other refractive indices, both real and complex. In addition experiments with commercial pigments are required in order to test the validity of the correction factors for polydisperse systems for which a single c/λ cannot be defined.
II. INTRODUCTION

When particle suspensions are irradiated, part of the incident radiation is transmitted, part is reflected and part is absorbed. The prediction of these quantities for a system of fixed geometry is possible only when the amount of energy absorbed and scattered by a particle and the directional distribution of the energy scattered are known. Theories exist to predict all the above parameters in the case of an isolated particle (1). No theory exists, however, for revealing the effect of the proximity of two particles upon their scattering behaviour. The only source of information in the last case is experiment.

The results of an experimental study of scatter from a system of particles variably spaced will be useful to describe any system which can be modeled as a dispersion of particles in a continuous matrix. Typical systems are the following:

(a) Lightweight insulations such as polystyrene, polyurethane, wood fibers, all of them possessing a relatively large amount of void space.

From a geometrical point of view, these insulations are of one of two types: In the first, the solid is the continuous medium, the void being dispersed gas bubbles; the other type of insulation consists of a matrix of fibers
held together by a suitable bonding agent. The results of this study will be useful in both types. However, only in the first type will they be directly applicable for quantitative results. Larking and Churchill (2) studied the attenuation of radiation through both types of insulation, and found that scattering is mainly responsible for the blocking of radiation in foam-type insulators.

(b) Coatings used to impart special radiative or aesthetic qualities to a surface. These properties are obtained through the use of pigments which absorb or scatter light. High reflectance is achieved by use of a white pigment such as TiO₂. The true measure of a white pigment is its ability to scatter light. Fig. 2.1 schematically shows how a ray of light falling upon a paint film is reflected.

The optical properties of thin paint films of white pigments depend on the following parameters: Thickness L, diameter of particle, refractive indices of both particles and continuous matrix, wavelength of incident light and center-to-center distance between two particles (δ). In particular, the greater the difference in refractive index between pigment and vehicle the greater will be the scattering power developed. The high refractive index of rutile TiO₂ relative to most organic vehicles (2.73 vs 1.50 - 1.60), accounts for its great scattering power.
Incident Light
Equal parts of short and long wavelength

Reflected Light
Short $\lambda$ Long $\lambda$

White paint film

White pigment particles

Substrate

Fig. 2.1 Scattering of Light by White Paint
In practice the physical properties of the vehicle and pigment particles are known, and the problem arises as to how to determine the influence of the pigment volume concentration (volume fraction occupied by pigment particles, hereafter referred to as PVC) of TiO₂ on particle size for maximum light-scattering efficiency. Previous work relevant to this problem will be reviewed later.

(c) Solar energy collectors designed to absorb solar radiation selectively and to emit amounts of longwave radiation which is small relative to black-body emission at the collector temperature. This may be accomplished by either a homogeneous or a heterogeneous surface. In this study we are interested in the surface formed by absorbing particles dispersed in a continuous matrix. In contrast to the case of white TiO₂ paints, we are interested in obtaining the highest absorbance and lowest emittance. This problem is far more complicated than the one considered previously, and its solution again requires determination of the scattering and absorbing efficiencies of particles in very concentrated suspensions.

Because of its practical importance in so many diversified problems, radiant transfer in scattering media has received considerable attention since the turn of the century.
Work has been devoted to optically thin media containing well-separated particles, for which radiant transfer theories apply, as well as to close-packed systems where information can be obtained only by experimentation.

Mie (1) in 1908 treated the problem of the scattering of a plane wave by a homogeneous sphere. Many authors - Chu and Churchill (4), Penndorf (5), and others - have presented reviews of the Mie theory for computation purposes. An extensive summary of these computations is presented by Van de Hulst (6).

The experimental verification of Mie's theory has been repeated many times. Typical results showing comparison between theory and experiment were reported by Heller (7). More recently, Kratochvil and Smart (8), using a Brice-Phoenix photometer, found excellent agreement between the theoretical Mie intensity function and the corresponding experimental absolute intensities. The use of Mie's theory for the calculation of particle concentration and size from scatter measurements is so common today that it would be impossible to review all the work done in a reasonable space. Several examples include:

(a) Application of Mie's equations for calculation of soot concentration in flames by Erickson (9) and Dalzell (10).

(b) Use by Heller and others (11) of specific turbidity measurements in order to find the size of polystyrene particles. They compared the "Optical diameter" with that
of electron microscopy, and discovered that the latter is higher; this is possibly accounted for by minute flattening out of the spheres in electron microscopy. The systematic error in electron microscopy has been as large as \( \pm 5 \) per cent.

(c) Measurements by Hepplestone and Lewis (12) of the efficiency factor \((X_g)\) of polystyrene monodisperse suspensions of latex spheres under rigorously controlled experimental conditions: They found good agreement with Mie theory. The polystyrene spheres were supplied by Dow Chemical Company, which supplied values of the mean sphere diameter in each suspension together with the standard deviation of their measurements. The authors report, however, that one size has been increased by 8\% in order to match the experimental and theoretical curves.

The obvious conclusion is that the sizes as supplied by Dow Chemical Company are not always in agreement with light-scattering measurements. It is interesting to note also the work of Davidson (13), who found significant differences in the diameter of the polystyrene particles, measured by flow ultramicroscopy, and the values reported by Dow.

In the case of low concentrations of scattering particles such as a colloidal solution of \(10^{-6}\) cm\(^3\) of gold in 1 cm\(^3\) of water as investigated by Mie, one can assume that
the mutual interaction of the particles with the scattered light is negligibly small. The action of the colloidal solution can be evaluated as $N_p$-fold that of a single particle; $N_p$ is the number of particles in the solution.

In the typical case of multiple scattering one must reckon with an optical thickness - defined as the product of the extinction coefficient ($K_L$) times the light path (L) - larger than .10 (30). One expects that here, in contrast to the case treated by Mie, multiple scatter will predominate.

A comprehensive review, as for single scatter, is not possible in the space available; and emphasis will be placed on the studies most pertinent to the present investigation. These include:

(a) An extensive and outstanding work in the area of radiative transfer by Chandrasekhar (14). As an astrophysicist, Chandrasekhar is concerned with two problems, (a) the reflection of planetary atmospheres and (b) radiation from stellar atmospheres. He has formulated integro-differential equations the solution of which gives the monochromatic intensities as a function of the angle of exit from the atmosphere. Chandrasekhar presents solutions to this problem for the case in which scatter of a single particle follows Rayleigh's law, or in which scatter can be expressed as a Legendre polynomial series. He presents
results for two and three-term descriptions of the single scatter diagram. The method he used involved reiterative numerical solution of integral equations.

(b) Numerical values for other problems presented by Evans et al (16): They used the set of integral equations derived by Chandrasekhar. They found that integrated reflectance and transmittance are characterized almost completely by the fraction of radiation scattered into the forward hemisphere. This suggests that a complicated phase diagram associated with a large circumference-to-wavelength ratio can be approximated satisfactorily by a simpler function with the same forward-scattered fraction. This result is important from a scientific and economic point of view, since the cost and accuracy of the results increase with the number of terms used to represent the phase function. It should be noted however that the authors varied their phase function, for a given fraction scattered forward, only over a finite limited range. Extrapolation of their conclusion to the limit of the two-flux method can introduce unacceptable errors.

(c) A solution of the equation of transfer for a scattering, absorbing, and emitting medium by Love and Gross (17). Their method, which is restricted to axially symmetric plane parallel geometries, is based on Chandrasekhar's suggestion that the integrodifferential equation for the isotropic
case may be reduced to a system of ordinary linear differential equations by substituting Gaussian or other quadrature formulae for the integral term. They further made use of Sykes (18) remark that the discontinuity in the intensity function for values of \( \mu = 0 \) suggests dividing the problem into a forward component \( I(\tau, +\mu) \) where \( 0 < \mu \leq 1 \) and a backward component \( I(\tau, -\mu) \) where \( -1 \leq \mu < 0 \), and then utilizing the so-called "double Gaussian quadrature". Their final solution accounts for diffusely emitting and reflecting bounding surfaces.

(d) Total and directional reflectances for semi-infinite diffusers with matrices of various refractive indices by Giovanelli (19). The phase function considered was

\[ \omega(1 + x \cos \theta) \]

where \( \omega \) is the albedo for single scatter (\( \omega = K_s / K_t \), \( K_s \) and \( K_t \) scattering and total extinction coefficient respectively) and \( \theta \) is the polar angle between scattered and incident beam. Giovanelli reported that the refractive index of the matrix of a diffuser affects its reflecting properties because not all the radiation incident on the surface from within passes through, some being internally reflected. Hence, he found that the reflectance depends not only on the albedo and phase function but on the value of the refractive index. His results can be used to show that the reflection at the boundaries increases the absorptance of the radiation incident on the diffuser.
(e) A solution of the equation of transfer for an absorbing and scattering medium with bounding surfaces reflecting according to Fresnel's law, by Hottel and Sarofim and others (20, 43). They have developed a computer program which calculates total and directional transmissivities and reflectivities of a plane-parallel dispersion of finite optical thickness, irradiated by an axially symmetrical flux. The phase function was assumed to be expressible as an n-term series of Legendre polynomials, but the numerical calculations were based on a phase function represented by a Legendre series of a few terms. They found that approximation of the integral term of the transfer equation by a twelve-term series was adequate to give convergence. Furthermore, the scheme of numerical quadrature did not have any effect on the accuracy of the results. During their work Giovanelli's (19) earlier finding was confirmed that if the plane boundaries of the scattering medium were assumed to reflect according to Fresnel's law, an absorbance was calculated which was several-fold that of a medium with no reflecting boundaries. However, rigorous calculation of absorbance for systems of engineering interest, using the energy transport equation, is time-consuming and approximate methods become desirable.

One such approximate method is Theissig's two-flux model (23). Theissig formulated a model for spherical particles in which the scattered radiation is confined
to two directions, forward and backward. The method was discussed by Woodward (22) and generalized to include the effect of polarization. The method divides a one-dimensional system into layers and considers successive angular intensity distributions for each successive order of scattering as it occurs in each successive layer. At each particular level inside the scattering slab it considers quantities of light $Q_k$, which have undergone different orders $k$ of scattering and angular distribution functions $f_k(\theta)$ which give a qualitative statement about the distribution of scattered light. The forward and backward components of the distribution function for any order of scattering, $k$, depends on $f_k(\theta)$. The exact form of the $Q_k$ and $f_k(\theta)$ functions was described by Hartel (21). Theissig (23) presents corrections to $Q_k$ for the cases in which the plane scattering layer cannot be considered to be infinite in the plane normal to the direction of propagation.

Woodward considers only the flux in the direction of propagation of light, and as a consequence of this simplification the model must be restricted to particles which are predominately forward scatterers. The same investigator has performed a series of multiple-scatter experiments using two aqueous dispersions of polystyrene latex spherical particles of average diameters $d=2.12$ and $2.89\mu$. Woodward reported agreement between his experimental results and the
multiple-scattering theory of Hartel. He did not correct, however, for the reflection of the emergent beam at the water-glass-air interface.

Smart et al (24) have carried out similar but more extensive experiments and computations of multiple scattering by polystyrene latex particles with \( d=1.277 \mu \)m. They used a special rectangular cell to correspond to an infinite plane-parallel slab. Measurements were made in the range \( 0^\circ \) to \( 90^\circ \). The higher angles were accessible by placing the scattering cell into a cylindrical jacket containing Nujol. The authors claim excellent agreement between the experimental results and Hartel's theory, but in fact there were discrepancies between theory and experiment which increased with increased optical thickness. The reason for the discrepancy, as noted by Orchard (52), is due to the breakdown in the Hartel theory which in the limit of optically thick media predicts a transmittance of 0.5 in place of the correct value of zero.

Approximate methods that are less complicated than the Hartel theory are reviewed by Chu and Churchill (25). In 1905 Schuster (27) developed a set of simultaneous differential equations to describe the transmission of light through fog. Many years later the same set of equations was applied by Kubelka and Munk (29) in discussing the optical properties of layers of paint. The two-flux theory was
extended by Hamaker (28) to include temperature radiation and the combined effect of radiation and heat conduction. Hamaker points out that observations to which the two flux model is applicable must be made under completely diffuse illumination. Comparison of values of reflectance and transmittance calculated by the two flux method were given by Churchill and Chu (51). They confirmed Hamaker's earlier finding that the two-flux model gives good agreement for integrated reflectance and transmittance for diffuse incident radiation but does not provide a good measure of the effect of angle of incidence for plane-parallel incident beams.

An improvement over the two-flux model is the six-flux model developed by Chu and Churchill (25). The radiation is considered to be scattered by a particle in six mutually perpendicular directions — forward, backward, and to the four sides. A system of six simultaneous linear differential equations result the solution of which give an answer for the reflection and transmission. The transmission and reflection from plane parallel dispersions are much more closely approximated by the six-flux than the two-flux model. In case the incident light is a parallel plane wave normally falling upon a parallel plane region of scattering the six-flux model degenerates to a simple two-flux model.
Another approximate model was discussed by Blevin and Brown (34). They used the solution of the transfer equation in the isotropic case together with an effective scattering coefficient in order to handle complex diffuser problems. They checked the validity of their model by measuring the reflectance of a semi-infinite medium composed of zinc oxide or barium sulfate pigment particles. In view of the fact that the pigment particles were non-spherical and polydisperse, with apparent aggregate structures, it was impossible to check the validity of the approximate theory. Blevin and Brown concluded, however, that their theory provides a basis for an understanding of the behavior of pigment suspensions. The same authors (35) made a comparison between the Kubelka-Munk theory (29) and Giovanelli's (19) more detailed model for the case of a semi-infinite scattering medium. They concluded that both theories give the same degree of accuracy unless Giovanelli's diffuser theory is based on a scattering distribution which is more realistic than isotropic scattering. Their theoretical findings were confirmed by measuring the total reflectance of enamel paints in distilled mineral turpentine in which absorption was introduced by adding a black enamel paint of very low diffuse reflectance. They point out, however, that a more elaborate theory than the previous one is necessary when diffuser properties other than total reflectances are being considered.
The multiple-scatter models discussed above consider independent scattering by neighboring particles. We might expect, however, that waves scattered by two neighboring particles will interfere at some distance away. Close proximity will therefore affect the distribution of scattered radiation and the amount of radiation scattered by a particle.

In the following section some results of close-packed systems are discussed. Theoretical and experimental findings are examined to determine those cases in which the multiple-scatter models no longer describe the optical properties of very dense particles suspensions. First, rough criteria for deciding when interference is important are considered.

Van de Hulst (30) states that independent scatter usually can be expected when the separation distance of particles exceeds 1.5 diameters; this criterion may be inadequate in case the particle diameter is smaller than the wavelength. Rosenberg (32) indicates that interference between the wavelets scattered by different particles can be neglected when there is no systematic relation between the positions of the scattering centers and when the interparticle distance exceeds a wavelength. Trinks (33) gives an explicit expression of the effect of interference in case of two adjacent Rayleigh scatterers and in the
special case the incident radiation is perpendicular to the connecting line between the two spheres; the radiation is assumed to be linearly polarized with its electric vector perpendicular to the line.

Any effort to allow for interference between particles on the basis of exact multiple-scattering theory leads to formidable mathematical expressions very hard to handle even with the fastest computing machines.

The difficulty encountered in a theoretical solution of interference has initiated a large amount of experimental work. In most of the experiments an interparticle distance is established at which interference starts. No effort is made, however, to predict the effect of interference on the intensity distribution of the scattered radiation.

There are two different types of systems in which interference may influence scattering properties;
(a) Layers of particles that are characterized by very small separation distances between the particles. Fahimian (31) has presented an excellent review of the literature in this case.
(b) Pigments suspensions where the particulate material is distributed inside a continuous matrix (binder); this is the type of system of present interest.
Relevant to the last system is the work done in measuring the scattering parameters of concentrated lattices of polystyrene spheres. Churchill (36) studied the effect of particle separation distance on the scattering properties of such a system using a modified two-flux model to correlate his data. The constant in this model remained constant down to a centre-to-centre distance of about 1.7 diameters between particles and changed less than 10% down to a centre-to-centre distance of 1.4 diameters, corresponding to 28% solids by volume. Two particle diameters were used, .814 and 1.171 microns, and a wavelength of illumination of 0.546 microns. The \(\frac{\sigma - \delta}{\lambda}\) ratios in the extreme case of a 28% concentration were .6 and .7.

Fahimian (31) measured the transmittance of multilayers of PV/VT spheres 3.49 microns in diameter. He found that thick layers exhibit diffuse, Lambertian transmittance and reflectance behavior. The total transmittance was correlated in terms of a two-flux model. The coefficient in this model could be calculated assuming that the voids rather than the closely spaced particles are the scattering centers and that these are far enough apart to be treated as independent scatterers. Measurements of alumina multilayers could not be correlated, however, in terms of a two-flux model, due possibly to a changing void character with increasing layer
thickness or to a breakdown in the applicability of the two-flux model. (The restrictions on the use of the two-flux model will be discussed in Chapter III.)

Instead of using idealized systems many investigators have made measurements of suspensions approaching the physical characteristics of pigments. Some important findings are reviewed below.

Berry (37) measured the optical transmittance of suspensions of precipitates of AgBr having average grain sizes in the range of 0.1 to 1.0μ. Water suspensions and gelatin coatings were measured with visible radiation. Measured turbidities were compared with that predicted by Rayleigh and Mie theories. Satisfactory agreement between theory and experiment was found in case of very dilute suspensions but Xs decreased significantly for concentrated suspensions. He concluded that the decreased scattering efficiency can be explained if the interstices are taken as the scattering centers. No interference was present, however, so long as the spacing was greater than two diameters.

Because of their practical importance, TiO2 pigment suspensions have been extensively studied. Harding et al (38) considered the problem of reduced scattering efficiency at high concentrations. The "two-constant" Kubelka-Munk theory was used in order to determine the
effective scattering coefficient. The latter was found to decrease enormously in case of large loadings of TiO₂ in the pigment. Harding et al proposed a correlation for the ratio of the theoretical and experimental scatter efficiencies, but they did not extend their measurements to low PVC in order to test whether the measured Xₚ values approached the theoretical values calculated from the Mie equation. It is not expected that the effective scatter cross section that fits the two-flux solution to experiment will equal the value calculated from the Mie equations, and Harding et al's conclusion is therefore based on a combination of two factors - possible interference between particles and the difference between the two-flux and the rigorous scatter theories.

Several investigators (49, 40, 41, 42, 3) studied TiO₂ pigment and tried to find the critical volume concentration above which the scatter efficiency of TiO₂ particles drops. Stieg (39) cites that the hiding power (expressed in square feet per pound of TiO₂) is a multiple of the amount of pigment present up to a PVC of 10 percent; beyond this limit he found that the hiding power was greatly reduced. Assuming a monodisperse system of TiO₂ spherical particles in a rhombohedral arrangement he concluded that reduced scattering efficiency starts when the separation distance becomes one-half the wavelength.
Marchetti and Willets (40) have also observed that the scattering efficiency of a TiO\textsubscript{2} system starts falling at a PVC of about 9.25%, corresponding to a separation distance of about one particle diameter or one-half the wavelength of visible light i.e., 0.2 to 0.4\(\mu\). Hence reduced scattering efficiency is the result of insufficient spacing between the particles. With the conclusions stated above the concentration of particles at which maximum reflectance is obtained is determined by the need for a clearance of about half a wavelength between particles; the PVC at maximum reflectance will therefore increase with increased particle size (3).

Craker et al (42) measured the distribution of scattered radiation from TiO\textsubscript{2} pigment films. They continuously varied the product of PVC times film thickness in mils. Single-scatter theory was valid for a product less than 0.5. For a product greater than 7.5 the Kubelka-Munk approach becomes satisfactory. They could not correlate their data with any existing model in the intermediate range. They confirmed Mitton et al’s (41) previous remark that the Kubelka-Munk model is applicable only when the light incident upon the film and everywhere within the film is diffuse.

Blevin and Brown (45) followed an entirely experimental approach in determining the effect of particle separation
on the reflectance of semi-infinite diffusers consisting of TiO$_2$ aqueous suspensions. They observed the curious phenomenon of an increase in the reflectance with PVC of TiO$_2$ at a wavelength of 365μm. They regarded the increase in the reflectance as due to an increase in the scattering coefficient per unit pigment volume concentration. However a decrease in the scattering coefficient was observed at higher concentrations for all wavelengths.

Hughes et al (46) in a recent article verified Blevin's discovery that there is an optimum concentration of TiO$_2$ at which the scattering efficiency is high. They constructed a special goniometer in order to measure the angular and unscattered radiation of paint films based on a pigment of TiO$_2$. Using films of 1-3 microns in thickness they were able to keep the percentage of unscattered radiation high and, by applying Lambert-Bier's law, to calculate the scattering efficiency of TiO$_2$. Their work is unique in determining $Q_{sca}$. All previous investigations give $bQ_{sca}$ as a function of the system parameters.

The above survey indicates that although much has been done on scatter from a suspension, the approaches to the problem have tended to be either mathematical exercises unsupported by data, or incomplete or approximate theoretical treatment bolstered by empirical
determination of constants. Furthermore the effect of particle spacing has been left hanging, with quite inadequate description of the extent to which departure from non-interaction between particles is due to spacing measured in particle size or spacing measured in wavelengths. Accordingly it was decided to design an experiment to measure the effect of optical density and concentration on the scatter characteristics of suspensions, to compare the results with predictions based on a model as free from approximations as is possible for well-separated particles, and to search for a method of correlating experimental results on suspensions too concentrated to justify assumption of absence of particle interaction.
III. SINGLE-SCATTERING THEORY

The deflection and dispersion of radiation by variations in refractive index within a medium is defined as scatter. Qualitatively scatter by a particle is a consequence of the oscillating electric field associated with any kind of electromagnetic radiation, which sets the electric charges contained within the particle into forced oscillation with a frequency equal to that of the incident radiation. These oscillating charges are sources of scattered radiation.

3.1 Physical description of a single scattering process

The purpose of a scattering theory is to provide a description of the directional distribution of the radiation scattered by a particle and of the energy absorbed by a scattering particle. Before considering specific theories let us consider the simple case of a beam of parallel light incident on a single particle much smaller than the wavelength of the incident light. In order to describe the scattering wave it is sufficient to know the amplitude of its components in two mutually perpendicular directions. First define a plane of reference for the scattering problem as defined by the unit vector $s_0$ in the direction of propagation of the incident plane wave and the unit vector $s$ in the direction of a scattered beam. Let $\theta$ be the angle formed by the vectors $s_0$ and $s$. Let $r_0$ be a unit vector perpendicular to the scatter (observation) plane
and \( l_0 \) be a unit vector in the scattering plane and perpendicular to \( s_0 \). Thus \( s_0 = r_0 \times l_0 \) as shown in Figure 3.1.

The components \( E_{r0} \) and \( E_{l0} \) of the vector which defines the direction and amplitude of the periodic motion of the oscillating charges are also indicated in Figure 3.1. If the incident light is unpolarized the components \( E_{r0} \) and \( E_{l0} \) are equal. On the other hand, if the incident light is plane polarized with its electric vector perpendicular or parallel to the plane of observation, the amplitude \( E_{l0} \) or \( E_{r0} \) will be respectively zero. The amplitude of the two components of the electric vector associated with the scattered light \( E_r \) and \( E_l \) will be proportional to the projected amplitudes of \( E_{r0} \) and \( E_{l0} \) on the direction of observation OP. Referring to Figure 3.1, the amplitude \( r_0 \) being perpendicular to the plane of observation
does not vary with angle $\theta$. Since the intensity of the light is proportional to the square of the amplitude of the associated electric field the plane polarized scattered intensity $I_\perp$ whose electric field vector is perpendicular to the plane of observation is independent of the angle of observation $\theta$. If the incident light is plane polarized in the scattering plane, the observer sees the projection of $E_{10}$ onto a line normal to OP, that is $E_{10} \cos\theta$. The intensity associated with the last case is $I_\parallel = A \cos^2 \theta$. In case of randomly polarized incident light the scattering intensity is $I = \frac{A}{2} (1 + \cos^2 \theta)$. The analytical expression for the intensity of radiation by particles much smaller than the wave length was developed by Lord Rayleigh in 1899. The intensity of the scattered light polarized in and perpendicular to the observation plane are given by the following equations:

$$I_\perp = \frac{9\pi^2}{r^2} \left[ \frac{m^2-1}{m^2+2} \right] \frac{2V^2}{\lambda^4} I_{o,\perp} \tag{3.1}$$

$$I_\parallel = \frac{9\pi^2}{r^2} \left[ \frac{m^2-1}{m^2+2} \right] \frac{2V^2}{\lambda^4} \cos^2 \theta I_{o,\parallel} \tag{3.2}$$

for the perpendicular component and,

The intensities $I_\perp$ and $I_\parallel$ are inversely proportional to $r^2$, where $r$ is the distance from the scatterer to the point of observation; $I_{o,\perp}$ and $I_{o,\parallel}$ are the corresponding incident light intensities. The refractive index of the particle with respect to the surrounding medium is $m$, the volume of the scatterer is $V$ and the wavelength of incident light is denoted by $\lambda$. 
For unpolarized incident light of intensity $I_o$ the total intensity of scattered light $I$ is given by:

$$I = \frac{9\pi}{2r^2} \left( \frac{m^2 - 1}{m^2 + 2} \right)^{\frac{3}{4}} \frac{\nu^2}{\lambda^4} (1 + \cos^2 \theta) I_o \tag{3.3}$$

3.2 Mie theory

The objective of the Mie theory discussion is to give the mathematical quantities which can be compared with experimental data. Such quantities are: the intensity of scattered light for the two plane-polarized components and the total scattered light in the case the particle size is of the same order of magnitude as the wavelength of incident light. Thus we have:

For incident linear perpendicular polarization

$$I_\perp = \frac{k^2}{k^2 - 2} I_o, \perp \tag{3.4}$$

for incident linear parallel polarization

$$I_\parallel = \frac{k^2}{k^2 - 2} I_o, \parallel \tag{3.5}$$

and for the total scattered light when the incident light is unpolarized

$$I = \frac{1}{2} I_o \tag{3.6}$$

where $k = 2\pi/\lambda$ is the wave number, $i_\perp$ and $i_\parallel$ are functions of the following form:

$$i_\perp = f(x, y, \theta) \tag{3.7}$$

$$i_\parallel = f(x, y, \theta) \tag{3.8}$$
where $x$ is the size parameter defined as the ratio of the circumference of a particle to the wavelength of light, $y$ is the product of $x$ and the index of refraction ($m$) of the scattering particle relative to the surrounding medium, and $\theta$ is the angle of observation relative to the forward direction of the incident light. The nature of the functions is developed elsewhere (9.10,31). It is seen from equations 3.4 to 3.8 that for a fixed wavelength and refractive index $m$ the intensities $I_\perp, I_\parallel$ are functions of the particle size.

We have so far discussed scattering by a single particle. In our work we shall be involved with the scattering of suspensions of many particles. When the concentration of the particles and the distance through which the incident and scattered light must travel are small the intensity of the light scattered by a cloud of particles is directly proportional to the number of scattering particles. This is always true when the amount of light extinguished by the sample containing the particles is negligible. Generally this condition is satisfied if the sum of the pathlength traversed by the incident and scattered beams have a transmissivity greater than 0.9.

3.3 **More definitions**

When a ray travels in a medium containing particles which absorb and scatter radiation, the intensity of the ray decreases in a way described by the equation:
\[- \frac{dT}{I} = K \cdot dx \quad (3.9)\]

The proportionality constant in the absence of scatter is the absorption coefficient, \(K_a\); in the absence of absorption is the scatter coefficient \(K_s\) and in the presence of absorption and scatter, it is the total attenuation or extinction coefficient \(K_t\), equal to the sum of \(K_a\) and \(K_s\). The unit \(\frac{K}{a}\) of \(K\) is area per unit volume. If we divide \(K\) by the number of particles per unit volume, the resulting area is called the particle cross section \(C\); the ratio of \(C\) to the geometrical cross section is known as the efficiency factor \(\gamma\). \(K, C, X\) carry subscripts \(a, s, t\) to indicate relation to absorption, scatter, or total extinction.

For a Mie scatterer the scattering efficiency is equal to:

\[X_s = \frac{1}{x^2} \int_0^\pi \left[ (i_\perp(\theta) + i_\parallel(\theta)) \right] \sin \theta \, d\theta \quad (3.10)\]

where \(x = \frac{2\pi r}{\lambda}\) is the size parameter and the remaining quantities have the same notation as before. The ratio of the scatter coefficient to the total extinction coefficient is commonly referred to as the albedo for single scatter \(\omega = K_s/(K_s + K_a)\).

The scattering cross section gives the total amount of energy intercepted by a single particle but does not provide any information as to how this energy is distributed around the particle. This is determined by the phase function. Let a direction in space be defined by its polar angle \(\theta\) and its azimuthal angle \(\psi\) and let the scattered flux in the direction
$(\theta, \psi)$, per unit angle of divergence of the incident beam and per unit angle of divergence in scatter, be represented by $Q(\theta, \psi)$. The phase function, the ratio of intensity scattered in the direction $\theta, \psi$ to that scattered by an isotropic scatterer is defined as follows:

$$p(\theta, \psi) = \frac{4\pi Q(\theta, \psi)}{\int_{4\pi} Q(\theta, \psi) d\Omega_s} \tag{3.11}$$

For symmetrical particles the phase function is independent of $\psi$ and is presented as a function of $\theta$ in polar coordinates.

Replacing $d\Omega_s = 2\pi \sin \theta \, d\theta$ where $\theta$ runs from 0 to $\pi$ we obtain:

$$p(\theta) = \frac{2Q(\theta)}{\int_0^{\pi} Q(\theta) \sin \theta \, d\theta} \tag{3.12}$$

Mie theory gives the following equation for $Q(\theta)$:

$$Q(\theta) = \frac{[i_\perp(\theta) + i_\parallel(\theta)]I}{2k^2} \tag{3.13}$$

Combining equations 3.10, 3.12, and 3.13 yields:

$$p(\theta) = \frac{2[i_\perp(\theta) + i_\parallel(\theta)]}{x_s x^2} \tag{3.14}$$

or

$$p(\theta) = \frac{4i(\theta)}{x_s x^2} \tag{3.15}$$

One can show that the Mie-theory angular distribution of scattered intensity for incident unpolarized light may be approximated to a high degree of accuracy by a sum of Legendre
polynomials with constant coefficients. Hence the phase
function \( p(\theta) \) may be written:

\[
p(\theta) = \sum_{n=0}^{\infty} a_n P_n(\cos \theta)
\]

(3.16)

where \( P_n \) is the Legendre polynomial of order \( n \). The coeffi-
cients \( a_n \) depend on the wavelength \( \lambda \) of the incident light,
the particle diameter \( D \), and the relative refractive index \( m \).
Appendix A gives the mathematical and computational scheme for
determining the coefficients \( a_n \).
4.2 Geometrical system

The system under study is a uniform dispersion of spherical particles, embedded in a clear matrix forming a slab of homogeneous material. The dispersion has a thickness L and is irradiated by a collimated flux $I_0$, which is polar symmetrical, incident at an angle $\theta_0$ with respect to the z-axis as shown in Figure 4.1.

The position of an element of mass dm at any point will be specified by the distance $z$, the cosine of the polar angle and the azimuthal angle $\Psi$. The polar angle $\theta$ is measured positively from zero (positive z-axis) to $\pi$ (negative z-axis). The azimuthal angle $\Psi$ is measured through $2\pi$ radians in the plane of stratification relative to some arbitrary angle. In general, directions will be indicated by the symbol $(\mu, \Psi)$ where $\mu = \cos \theta$. For example, $I(z, \mu, \Psi)$ is the intensity at level $z$ and in direction $(\mu, \Psi)$.

Figure 4.2 may be used to derive the relation between the angle of two scattered beams and the azimuthal angles: The two beams form with the z-axis. Radiation is incident on a mass element dm at 0 in the direction $O_1P_1$ and is scattered at 0 through an angle $\theta$ into the direction $O_2P_2$. From the spherical triangle $z, P_1, P_2$ we obtain the following relation:

$$\cos \Theta = \cos \theta \cos \theta' + \sin \theta \sin \theta' \cos (\Psi' - \Psi)$$

$$= \mu \mu' + (1 - \mu^2)^{1/2} (1 - \mu'^2)^{1/2} \cos (\Psi' - \Psi)$$ (4.2)
Figure 4.1  Geometrical representation

Figure 4.2  The relation between the scattering angle $\theta$ and the coordinate angles $\psi, \psi', \theta$ and $\theta'$. 
where \( \mu = \cos \theta \) and \( \mu' = \cos \theta' \). The angles \( \theta \) and \( \theta' \) are, respectively, the polar angles of incidence and scattering and \( \psi \) and \( \psi' \) are corresponding azimuthal angles.

### 4.3 Optical thickness

The intensity of radiation emerging from the bounding surfaces of a particulate atmosphere are functions of the size parameter, the relative refractive index of the particles \( m \) and the optical thickness, \( \tau \), defined as the product of the total extinction coefficient times the slab thickness \( L \):

\[
\tau = (k_a + k_s)L
\]

where \( k_a \) and \( k_s \) are the extinction coefficient for absorption and scatter, respectively.

For a non-absorbing medium the absorption coefficient is zero; and the scattering coefficient is defined as:

\[
k_s = N \frac{\pi d^2}{p} X_s = 1.5 \frac{(P.V.C.)X_s}{d} \quad (4.3)
\]

where \( N \) number of particles per cc and PVC = \( \frac{\pi d^3}{p} \) pigment volume concentration.

### 4.4 Equation of transfer

### 4.4.1 Photon-particle interactions

The following interactions are possible when a ray of radiation hits an element of mass \( dm \), composed of spherical absorbing particles of the same size:
(a) Photons incident upon dm in time dt and in direction \((\mu, \psi)\) contained in the solid angle \(d\Omega\) are (singly) scattered into the solid angle \(d\Omega'\) in the direction \((\mu', \psi')\) by interactions of dm with particles. This scattering process may be considered as a redirection of an amount \(d[\Delta Q(z, \mu', \psi')]\) of the incident energy \(Q(z, \mu, \psi)\) into \(d\Omega'\).

(b) Photons incident upon dm in time dt and in the direction \((\mu, \psi)\) contained in the solid angle \(d\Omega\) are absorbed by the particles under consideration in dm.

(c) Photons incident on dm in time dt and in the direction \((\mu', \psi')\) contained in the solid angle \(d\Omega'\) are (singly) scattered into the solid angle \(d\Omega\) in the direction \((\mu, \psi)\). Again, this scattering process may be considered as a redirection of an amount \(d[\Delta Q(z, \mu, \psi)]\) of the incident energy \(Q(z, \mu', \psi')\) into \(d\Omega\).

The first two classes of photons are those which are lost from the radiation field in the direction \((\mu, \psi)\) by scattering and absorption, respectively. The last class consists of photons which are gained by the radiation field in the direction \((\mu, \psi)\) by scattering alone.

4.4.2 Mathematical formulation

Any equation which quantitatively describes all three photon particle interactions must account also the fact that at any optical depth there are some unscattered photons.

For the system under study - plane parallel dispersion of optical thickness \(\tau_1\), an albedo \(\omega\), a phase function \(P(\theta)\) and a
refractive index ratio of the matrix to the surrounding $n$ -
the equation of transfer is first formulated for non-reflecting
boundary conditions and then modified to allow for Fresnel re-
fection at the boundaries. The derivation of the equation is
given in numerous references and is not repeated here.

4.4.2.1 Non-reflecting boundaries

The intensity $I_{total}(z, \theta, \psi)$ at plane $z$ is conveniently
considered in two parts: $I_{o}(z, \theta, \psi)e^{-T/U_o}$ due to attenuation of
the unscattered part of the incident intensity $I_{o}$, and $I(z, \theta, \psi)$
due to the scattered part. On this basis, for unit incident
flux density at the bounding surface, the transport equation
takes the form:

$$\cos\theta \frac{dI(z, \theta, \psi)}{dz} = -(K_a + K_s)I(z, \theta, \psi) + \frac{K_s}{4\pi} \int_{4\pi} p(\Theta)I(z, \theta', \psi'; d\Omega')$$

$$+ \frac{K_s}{4\pi} \int_{2\pi} \frac{p(\Theta)}{\mu_o} e^{-(K_a + K_s)\frac{z}{\mu_o}} d\psi$$

(4.4)

The term on the left of the equation represents the rate
of change of the intensity with respect to distance along the
ray having a polar direction $\theta$ and an azimuthal angle $\psi$ with
respect to reference direction and located at some distance
dz/$\cos\theta$ along the ray. The first term on the right represents
the attenuation due to absorption and scatter. The second term
on the right represents the contribution, by scatter into the
reference beam, from beams that have been scattered one or more
times. The last term represents the scatter into the reference
beam out of the partially attenuated incident beam.

It is convenient to make the following changes in equation
(4.4). Express distance in terms of the optical thickness,
directions in terms of \((\mu, \psi)\) and introduce the albedo ratio \(\omega\). Then, equation (4.4) becomes:

\[
\mu \frac{dI(\tau, \mu, \psi)}{d\tau} = -I(\tau, \mu, \psi) + \frac{\omega}{4\pi} \int_{4\pi} p(\Theta) I(\tau, \mu', \psi') d\Omega'
\]

\[
+ \frac{\omega}{4\pi} \int_{2\pi} \frac{p(\theta_o)}{\mu_o} e^{-\tau/\mu_o} \frac{d\psi}{2\pi}
\]  

(4.5)

Substitution into equation (4.5) of the relation for \(p(\Theta)\)
given by equation (4.2) and of the identity \(d\Omega' = d\mu'd\psi'\),
replacement of the azimuth-independent intensity terms by
\(I(\tau, \mu)\) and \(I(\tau, \mu')\), and integration over the azimuth angles
yields (see Appendix B):

\[
\mu \frac{dI(\tau, \mu)}{d\tau} = -I(\tau, \mu) + \frac{\omega}{2} \int_{-1}^{1} I(\tau, \mu') \sum_{i=1}^{L} \alpha_i P_i(\mu) P_i(\mu') d\mu'
\]

\[
+ \frac{\omega}{4\pi \mu_o} e^{-\tau/\mu_o} \sum_{i=1}^{L} \alpha_i P_i(\mu) P_i(\mu_o)
\]  

(4.6)

The associated boundary conditions are \(I(0, \mu) = 0\) for
\(0 < \mu < 1\) and \(I(\tau_1, \mu) = 0\) for \(-1 < \mu < 0\) \((\tau_1 = \text{full slab thickness})\).
Equation (4.6) was solved by what may be called the method of
discrete conical sheets. The integral is replaced by a
weighted sum of the intensities at \(N\) discrete values of \(\mu'\)
yielding the following equation (see Appendix B):
\[
\frac{dI(\tau, \mu_i)}{d\tau} = -I(\tau, \mu_i) + \frac{\omega}{N} \sum_{j=1}^{N} W_j I(\mu_j) \sum_{k=1}^{L} a_k P_k(\mu_i) P_k(\mu_j) \\
+ \frac{\omega}{4\pi\mu_o} e^{-\tau/\mu_o} \sum_{k=1}^{L} a_k P_k(\mu_o) P_k(\mu_i)
\] (4.7)

Equation (4.7) is further simplified to:
\[
\mu_i \frac{dI_i}{d\tau} = -I_i + \frac{\omega}{N} \sum_{j=1}^{N} g_{ij} I_j + \frac{\omega}{4\pi\mu_o} e^{-\tau/\mu_o} g_{i,N+1}
\] (4.8)

As developed in Appendix B equation (4.8) has a solution of the form:
\[
I(\tau, \mu_j) = \sum_{i=1}^{N} c_i a_{ij} \gamma_i^\tau + r_j e^{-\tau/\mu_o}
\] (4.9)

For an albedo ratio \( \omega=1 \) (conservative scattering) two eigenvalues of the matrix in the Appendix B are equal (see Reference 43) giving rise to a solution of the following form:
\[
I(\tau, \mu_j) = \sum_{i=1}^{N-1} c_i a_{ij} \gamma_i^\tau + c_N (B_{Nj} + \tau A_{N-1,j}) e^\gamma_N^\tau \\
+ r_j e^{-\tau/\mu_o}
\] (4.10)

4.4.2.2 Reflecting boundary conditions

4.4.2.2.1 One step-change in refractive index

The reflectivity \( \rho \) at the boundary interface may be calculated from Fresnel's equation. For radiation externally incident on the surface at an angle \( \theta \) with the normal, the reflectivity averaged over the two components of polarization is given by:
where \( n \) is the ratio of the refractive indices of the matrix and the surroundings. When a beam is incident on the interface from the matrix side \( \rho \) may be calculated from equation (4.11) by substituting \( 1/n \) for \( n \), for all \( \theta \)'s less than \( \sin^{-1}(1/n) \); above this angle the beams undergo total internal reflection and \( \rho=1 \).

A beam incident at an angle corresponding to \( \mu_o \) will be partially reflected (see Figure 4.3). The direction \( \mu_o' \) of the component entering the matrix may be calculated from Shell's law,

\[
\sqrt{1-\mu_o'^2} = \sqrt{(1-\mu_o^2)/n}
\]  

(4.12)

Figure 4.3 traces the path of the unscattered component of the incident beam through a number of internal reflections. The flux density at a level \( \tau \) below the surface due to the unscattered and multiply-reflected part of an externally incident beam of unit flux density \((I_o d\Omega_o = 1)\) can be readily calculated (by series summation or by identification of sinks in a repeating cycle). The total flux density, in the direction and at plane \( \tau \), of the component of the incident beam is thus found to be \((1-\rho) e^{-\tau/\mu_o'/(1-\rho^2 e^{-2\tau l/\mu_o'})} \), and that directed away from the bottom surface in a direction \(-\mu_o'\) is \(\rho(1-\rho) e^{-(2\tau l-\tau)/\mu_o'}/(1-\rho^2 e^{-2\tau l/\mu_o'})\). Allowance in equation (4.6) for these differ-
Figure 4.3  Trace of Unscattered Component of Incident Beam
ences from the no-reflection boundary condition may be made by replacing the last term on the right hand side of the equation by:

\[
\frac{(1-\rho)e^{-\tau/\mu_o}}{1-\rho^2e^{-2\tau/\mu_o}} \sum_{i=1}^{L} a_i p_i(\mu) p_i(\mu_o')
\]

\[
+ \frac{\rho(1-\rho)e^{-(2\tau_1-\tau)/\mu_o}}{1-\rho^2e^{-2\tau_1/\mu_o}} \sum_{i=1}^{L} a_i p_i(\mu) p_i(-\mu_o')
\]

(4.13)

The associated boundary conditions are \( I(\sigma, \mu) = \rho I(\sigma, -\mu) \) for \( \sigma < \mu < 1 \) and \( I(\tau_1, \mu) = \rho I(\tau_1, -\mu) \) for \( -1 < \mu < \sigma \). The method of discrete conical sheets used to solve the intensity distribution for the no-reflecting boundary conditions is easily adapted to allow for the above modifications in the transport equation and the boundary conditions (see Appendix B). The calculated intensity \( I(\tau, \mu) \) will correspond to the value within the matrix. The values \( I(\sigma, \mu_e) \) and \( I(\tau, \mu_e) \) emerging through the interface may be calculated from the values for \( I(\sigma, \mu) \) and \( I(\tau_1, \mu) \) within the matrix by correcting the latter values for surface reflection, refraction and change in divergence. Thus the direction of emergence \( \mu_e \) is related to \( \mu \) by Snell's law, equation (4.12), and the ratio of intensities outside and inside the matrix are given by:

\[
\frac{I(\tau_1, \mu_e)}{I(\tau_1, \mu)} = \frac{I(\sigma, \mu_e)}{I(\sigma, \mu)} = \frac{(1-\rho)}{n^2}
\]
4.4.2.2.2 Two-step change in refractive index

For a system of particles confined between two parallel glass slides there is a two-step change in the refractive index in the water-glass-air interface. The mathematical derivation developed in the preceding section is again applicable.

The water-glass air interface reflectance $\rho$ can be expressed in terms of the reflectivities $\rho_1$ and $\rho_2$ of water-glass and air-glass interfaces:

$$\rho = \frac{\rho_1 + \rho_2 - 2\rho_1 \rho_2}{1 - \rho_1 \rho_2}$$

where $\rho_1$ and $\rho_2$ are obtained from Fresnel's equation (4.11). For the water-glass interface $n$ equals the ratio of refractive indices of water and glass, and $\theta$ is the angle with the normal made by the beam in the water phase; and where for grass-air interface $n$ equals the ratio of refractive indices of glass and air, and is the angle made with the surface normal by the beam in the glass phase. The refraction of the beam across an interface is given by Snell's law:

$$n_1 \sin \theta_1 = n_2 \sin \theta_2$$

The reflectivity $\rho$ is unity for angles $\theta$'s greater than $\sin^{-1}(1/n_w)$. Furthermore, the ratio of intensities outside and inside the matrix are given by:

$$\frac{I(\tau_1, \mu_e)}{I(\tau_1, \mu)} = \frac{I(0, \mu_e)}{I(0, \mu)} = \frac{1 - \rho}{n_w^2}$$
where $\mu_e$ is the direction of emergence related to $\mu$ by Snell's law:

$$\sqrt{1-\mu^2} = \sqrt{1-\mu_e^2}/n_w$$

### 4.5 Numerical results of multiple scatter calculations

The number of independent variables upon which the final solution of transfer equation depends is too large to allow a comprehensive coverage here. The effect of the optical thickness $\tau_1$, albedo for single scatter $\omega$, cosine of angle of incidence $\mu_o$, shape of phase diagram $p(\theta)$, refractive index of continuous matrix has been discussed elsewhere (43). The discrete ordinates method using an equal quadrature formula has been established as a powerful tool in giving answers in any complex multiple scatter problem.

However, no results have been presented using the above method in the case the albedo for single scattering is equal to one.

It has already been established (43) that two eigenvalues are equal to zero when $\omega=1$. It is further shown in Appendix B that the general solution has a different form from the one corresponding in the absorbing medium case.

Results obtained following the numerical procedure in Appendix B are compared with Bellman's calculations for a $\tau_1 = 1.0$, $\omega=1.0$ and isotropic scattering. As shown in Figure 4.4, the agreement is excellent.
Figure 4.4 Bidirectional Transmittance and reflectance of a suspension of pure scatterers in a layer of optical density one, with incident beam $13^\circ$ off normal. Comparison of results obtained by Bellman with calculations performed with present method.
4.6 Effect of phase function on the angular distribution of multiple scatter radiation

The significance of phase function on a multiple scatter problem has been discussed by many investigators (51,20). Churchill and Evans (51) have noticed that the total transmittance and reflectance of radiation through a medium of finite thickness depend primarily on the fraction of forward scatter $f$ and not on the detailed shape of the phase function.

The fraction scattered forward and the peakedness $p_e$ of a Mie scatterer are given by (see Appendix C):

$$ f = \frac{1}{2} + \frac{1}{2} L \sum_{i=1,3,5} (-1)^{i-1} \frac{1}{i(i+1)} \frac{1}{i!} (\frac{1}{3}.5..i)^2 a_i \quad (4.14) $$

$$ p_e = \frac{1}{8f} L \sum_{i=0}^{\lambda} \frac{\Gamma(\frac{1}{2})}{\Gamma(2+\frac{1}{2}i, \Gamma(\frac{5}{2}+\frac{1}{2}i)} a_i \quad (4.15) $$

where $a_i$ are the Legendre polynomial coefficients in which the phase function is expanded according to equation (3.16).

It is possible to replace the phase function corresponding to a Mie scatterer by another simple one having the same $f$ and $p_e$ (see Appendix C). The conditions under which such a replacement is feasible have been presented by Churchill (51). Figure 4.5 shows a graphical representation of a Mie phase diagram, $x=4.0 \ m=2.7$, described by a Legendre polynomial series of 17 terms. In the same plot a smooth curve, described by a Legendre series of 3 terms, depicts a phase diagram of the same $f$ and $p_e$. 
Figure 4.5 Replacement of the exact phase function ($x=4$, $m=2.7$) with one having the same peakedness and forward fraction.
Figure 4.6 Comparison of the angular distribution of transmitted and reflected radiation for two-phase functions having the same $f$ and $Pe$. 

\[ \omega = 1 \]
\[ \mu_0 = 1 \]
\[ \tau_1 = 100 \quad n = 1.5 \]
\[ x = 4. \quad m = 2.7 \]
\[ c_0 = 1. \quad c_1 = 0.5562 \quad c_2 = 1.262 \]
Calculations based on the two phase functions described above are presented in Figure 4.6. Other conditions introduced into the equation of transfer were \( \omega = 1.0 \) (conservative scattering), \( \mu_o = 1.0 \) (normal incidence), \( \tau_1 = 100 \) and refractive index of continuous matrix \( n = 1.5 \). As seen from Figure 4.6, there is a remarkable agreement in the angular distribution of the emerging radiation.

The implications of this finding are very valuable in any experimental study. There is a growing body of literature in an effort to find the scattering parameters of a cloudy atmosphere by measuring the transmitted and reflected radiation from the bounding surfaces. This is the so-called inverse scattering problem. Figure 4.6 implies that a certain angular distribution may correspond to more than one \( p(\mu) \). The significance of this finding will be discussed in the results section.

4.7 Simplified models

Though there are many simplified models in handling multiple scatter problems, only the two flux model is considered here, due to its importance in engineering applications.

The two flux model was proposed by Schuster (27) and extended by Hamaker (28).

An energy balance is written inside a slab of material in terms of two fluxes, one in the direction of propagation and the other opposite to it. The following equations are obtained:
\[ \mu_o \frac{dI_+}{d\tau} = \omega t - 1)I_+ + \omega bI_- \quad (4.16) \]

\[ \mu_o \frac{dI_-}{d\tau} = - \omega bI_+ + (1 - \omega t)I_- \quad (4.17) \]

where \( f \) and \( b \) are the fractions of the radiation that would normally be scattered into the hemispheres facing and opposed to the direction of propagation.

The boundary conditions are \( I_+(0) = 1 \) and \( I_-(\tau_1) = 0 \) where \( \tau_1 \) is the optical thickness of the entire slab. The fractions of the radiation reflected and transmitted are given by:

\[ \frac{I_-(0)}{I_+(0)} = \frac{(1 - \omega t - g)(1 - e^{-2g\tau_1 / \mu_o})}{\omega b(1 - \frac{1 - \omega f - g}{1 - \omega f + g})e^{-2g\tau_1 / \mu_o}} \quad (4.18) \]

and

\[ \frac{I_+(\tau_1)}{I_+(0)} = \frac{2ge^{-g\tau_1 / \mu_o}}{(1 - \omega t + g)[1 - \frac{1 - \omega f - g}{1 - \omega f + g}e^{-2g\tau_1 / \mu_o}]} \quad (4.19) \]

where \( g = \left[ (1 - \omega f)^2 - (\omega b)^2 \right]^{1/2} \)

In the case of \( \mu_o = 1 \) and \( \omega = 1 \), \( g = 0 \) and equation (4.19) becomes indeterminate. Taking the limit of (4.19) as \( g \to 0 \), we find:

\[ \frac{I_+(\tau_1)}{I_+(0)} = \frac{1}{1 + b\tau_1} \quad (4.20) \]

Or, using equation (4.3):

\[ \frac{1}{I_+(\tau_1)} = 1 + 1.5b \frac{X_s(P.V.C.)L}{d} \quad (4.21) \]

where \( L \) is the slab thickness.
For an absorbing system equation (4.19) may still be simplified by rewriting as follows:

\[
\frac{I_+^r(t_1)}{I_+^r(0)} = \frac{2q}{1 - \omega^2 + g} e^{-g t_1 / \mu_0} \sum_{n=0}^{\infty} \left( \frac{1 - \omega^2 - q}{1 - \omega^2 + g} \right)^n e^{-2 n g t_1 / \mu_0} \quad (4.22)
\]

The series portion can be reduced to unity if:

\[
g t_1 = \left[ (1 - \omega f)^2 - (\omega c)^2 \right]^{1/2} t_1 >> 1
\]

or

\[
1 \omega f + \omega b >> \frac{1}{t_1^2 (1 - \omega)} \quad (4.23)
\]

For scatterers with \( f > b \):

\[
1 >> 1 \cdot \omega (f - b) >> \frac{1}{t_1^2 (1 - \omega)} \quad (4.24)
\]

Hence:

\[
\frac{I_+^r(t_1)}{I_+^r(0)} = \frac{2q}{1 - \omega^2 + g} e^{-g t_1 / \mu_0} \quad (4.25)
\]

provided that \( t_1^2 (1 - \omega) >> 1 \).

Equation (4.25) has found widespread use in determining the absorption and scattering cross section of a system. A plot of \( I_+^r(t_1) \) versus slab thickness in semilogarithmic paper results in a straight line the slope and intercept of which may be used to find \( \chi_1 \) and \( \chi_2 \).

4.7.1 Evaluation of two flux model

The equation of transfer, as discussed earlier, describes exactly a multiple scatter problem. The calculations, however,
are lengthy and expensive and the final solution cannot be expressed in a closed form. Approximate models, therefore, such as the two flux model, have considerable value, since they give the reflectance and transmittance in a closed form. For the two flux model, equations (4.18) through (4.25) have been extensively used in many practical applications.

It is the purpose of this section to test the validity of this method and suggest conditions under which it may be used in case of non-absorbing media (\(\omega=1\)), where the applicable equation for the transmission is:

(a) In case non-absorbing particles are suspended in a matrix of unit refractive index:

\[
\frac{1}{I_+^e(t_1)} = 1 + \omega t_1
\]

(4.26)

(b) In case non-absorbing particles are suspended in a matrix of a refractive index (\(n\)) different than unity:

\[
\frac{1}{I_+^e(t_1)} = \frac{1+\rho}{1-\rho} + b t_1
\]

(4.27)

where \(\rho\) is for normal incidence, e.g.:

\[
\frac{1}{I_+^e(t_1)} = 1.0833 + b t_1 \text{ in case } n=1.5
\]

There is a growing body of literature concerning the application of equation (4.26) or (4.27). Most of it has been devoted in experimental measurements of transmittance versus slab thickness. Equation (4.21) is then used in calculating the \(\kappa X_s\) con-
Figure 4.7 The two-flux model for different values of $x$, $m$ and $n$
stant, which for a system of given P.V.C. depends on the particle size and the wavelength of incident radiation. The scattering efficiency $X_s$ is derived by assuming that the $b$ factor is the fraction scattered backwards by a single particle according to Mie theory:

$$b_o = \frac{1}{2} \int_{-1}^{0} p(\mu) d\mu = 2 \int_{-1}^{0} \frac{i(\theta)}{X_s x^2} d\cos \theta \quad (4.28)$$

The simplification introduced by equation (4.28) has been examined in the present work. The equation of transfer was solved for different values of $x$, $m$, and boundary conditions.

The results are listed in Appendix D. Numerical values are plotted according to equations (4.26) or (4.27) in Figure 4.7. The last plot suggests that, though it is justified to use the two flux approximate model in the extreme cases, when $\tau_1 > 10$ and $\tau_2 > 10$, there is an intermediate regime, where it breaks down. The range of the buffer region depends on $x$, $m$ and the nature of the bounding surfaces.

Furthermore, the following conclusions can be drawn from Appendix D:

1. For a system of non-absorbing spheres suspended in a matrix of unit refractive index:

   (a) In the optically thin region ($\tau_1 < 10$) $b = b_o$ and the two flux model is exact.

   (b) In the intermediate region $10 < \tau < 10$, $b$ varies with $\tau_1$. Consequently, the two flux model (e.g. 4.26) is not applicable.
(c) $b$ increases with $\tau_1$ taking a constant value for a $\tau_1 > 10$.

2. For a system of non-absorbing spheres suspended in a matrix of a 1.5 refractive index:

(a) In the optically thin region the two flux model is applicable, but $b$ is different than $b_0$.

(b) For intermediate values of $\tau_1$, i.e., $10 < \tau_1 < 100$ the two flux model is not applicable ($b$ varies with $\tau_1$).

(c) $b$ decreases with increasing $\tau_1$ and approaches a constant value for a $\tau_1 > 100$ (see Table D...0.15).

The above conclusions are in accordance with Churchill's finding that the two flux model is poor and unable to provide good absolute values for the transmittance and reflectance (51).

It is also interesting to note that Craker (42) who measured the transmission radiation from $\text{TiO}_2$ pigment films, has found that for a product of P.V.C. times film thickness (in mils) between .5 and 7.5 the two flux model is not applicable, in agreement with the conclusion arrived at theoretically in the present study.
V. EXPERIMENTAL APPARATUS AND PROCEDURE

5.1 Apparatus

The light scattering apparatus used for the measurement of the optical properties of dense polystyrene particle suspensions consists of a light source, a collimating system for the incident light beam, the particle system to be examined, a collimation system for the scattered radiation, and an electronic system for measuring the signal from the scattered radiation. The light scattering apparatus is shown schematically in Figure 5.1. In the following paragraphs a brief description of the various parts is given:

(a) The light source is a high brightness mercury arc used to produce an incident radiant field of high intensity.

(b) The collimation system for the incident light beam has as a primary purpose the production of an intense beam at the center of the system. For that reason a circular aperture is located next to the surface of the source. The light passing through the aperture and a series of stops, which define the usable solid angle from the source, hits a short focal length achromatic lens located at a distance from the source equal to its focal length. This lens renders, therefore, parallel light, which is received by a long focal length lens, placed at a distance from the center of the system equal to its focal length. A light stop located at the second lens determines the angle of
Figure 5.1  Schematic of apparatus used for scattering measurements
convergence of the incident light. The convergent beam produced by the second lens, is reflected from a first surface, aluminum-coated, partially transmitting mirror, it passes through a UV filter which filters out the radiation damaging to the eyes and is then reflected off a second mirror. The two mirrors placed in the path of the incident beam make the angles close to 180° accessible to measurement and they further allow the placing of a monitor behind the first mirror. This monitor measures the intensity of the radiation, transmitted through the first partially transparent mirror, and records changes in intensity with time. An iris diaphragm located between the second mirror and the target removes all diffuse radiation produced by the collimation system.

(c) The particle system is composed of polystyrene sphere lattices confined between two parallel glass slides and is located at the center of the system. The incident beam, which at the center of the system resembles a light tube of .12 cm in diameter, is normally incident on the front glass plate. Light is scattered in all directions.

(d) The collimation system for the scattered radiation collects part of the scattered light by an optical system, which has its various components on an axis passing through the intersection of the center of the sample holder and the axis of the collimation system for the incident light.
The various components of the optical system for collecting the scattered light are located on a rotating arm. The scattering angle $\theta$ is marked on a graduated circle. A scattered beam passes through an optical interference filter used to choose the desired wavelength. The divergence of the collection system is determined by a light stop located at the front of the lens tube. The divergence is selected to include only a narrow angle range but to yet yield measurable signals at all angles. The portion of the scattered light collected by the lens tube is passed through a series of light stops, and it is received by a lens, which focuses the light on a plane of a circular aperture. A polarization filter, located between this aperture and the lens, allows the measurement of either component of the scattered light.

(e) The electronic system is composed from a phototube, a high voltage power supply and a DC voltmeter. The multiplier phototube is located a short distance behind the circular aperture and is aligned with the optical system such that a spot of light strikes the photosensitive surface of the multiplier phototube. The current generated by the phototube is proportional to the intensity of the spot.

Details of the various components are presented in Appendix E.
5.2 Procedure

The mercury arc lamp is started and the system is optically aligned. The alignment procedure fixes the axis of the incident beam and the axis of the collection system for the scattered light in the same horizontal plane. In addition, the two beams intersect at the center of rotation of the rotating arm, where the test cell is located. The procedure for aligning the optical system is given in Appendix G of Ref. (9) and in Appendix C of Ref. (31).

The light source, the high voltage supply and the DC voltmeter are allowed to operate for a time sufficient to reach steady conditions. At least one hour is necessary for this phase before collecting data.

Each run starts with a measurement of the intensity of the incident light for both components of polarization. The steps necessary are to:

(a) Place a neutral filter and an optical interference filter in the rotating arm. The optical interference filter selects the desired wavelength. The neutral filter reduces the measured intensity by a known factor and it allows the RCA 931-A phototube to keep the anode current under 10 microamperes, a necessary condition for the operation of the phototube with maximum stability (53). With the resistors used across the multiplier phototube,
the above current limitation and the 1000 volts setting at
the high voltage supply used during all experiments, the
output voltage as measured by the DC voltmeter should not
exceed 3000 millivolts.

(b) Remove the diaphragm in front of the lens tube.

(c) Place the rotating arm at 0° degrees.

The alignment of the entire light-scattering apparatus,
and the voltage setting at the high voltage supply is un-
changed over the course of a run. During a measurement
the diaphragm is placed in front of the lens tube. Only
the angular position of the rotating arm θ and the setting
of the polarization filter is changed during a run.

5.3 Samples

The desire to have a well defined system necessitates
the use of spherical particles of known refractive index.
The spherical polystyrene latex particles, produced by Dow
Chemical Co., are ideal for this purpose. Refractive index
versus wavelength data are available (54). The reported
values at the two temperatures are listed in Table 5.1. A
description of the particles on which studies were made is
given in Table 5.2. The particle diameters, listed on
Table 5.2, were determined by electron microscopy and dif-
fered by 3 to 6 percent from the values supplied by the
manufacturer.
**TABLE 5.1**

Refractive Index of Polystyrene

<table>
<thead>
<tr>
<th>Wavelength (Å)</th>
<th>$n_{\text{p15°C}}$</th>
<th>$n_{\text{p35°C}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4358</td>
<td>1.617</td>
<td>1.614</td>
</tr>
<tr>
<td>4801</td>
<td>1.606</td>
<td>1.603</td>
</tr>
<tr>
<td>5896</td>
<td>1.592</td>
<td>1.589</td>
</tr>
<tr>
<td>6503</td>
<td>1.587</td>
<td>1.584</td>
</tr>
<tr>
<td>7679</td>
<td>1.581</td>
<td>1.578</td>
</tr>
</tbody>
</table>

**TABLE 5.2**

Range of Variables Studied

<table>
<thead>
<tr>
<th>Particle Diameter (μ)</th>
<th>Wavelength (μ)</th>
<th>PVC</th>
<th>Optical Thickness</th>
</tr>
</thead>
<tbody>
<tr>
<td>.102+.003</td>
<td>.436</td>
<td>.0284 - .2893</td>
<td>14.3-292.</td>
</tr>
<tr>
<td>.102+.003</td>
<td>.546</td>
<td>.0284 - .2893</td>
<td>5.4-110.</td>
</tr>
<tr>
<td>.106+.003</td>
<td>.436</td>
<td>3.53×10⁻⁵ - .10</td>
<td>.02-80.2</td>
</tr>
<tr>
<td>.106+.003</td>
<td>.546</td>
<td>3.08×10⁻⁴ - .10</td>
<td>.10-30.1</td>
</tr>
<tr>
<td>.248+.003</td>
<td>.436</td>
<td>.105 - .293</td>
<td>290-1096.</td>
</tr>
<tr>
<td>.248+.003</td>
<td>.546</td>
<td>.105 - .293</td>
<td>138-719.</td>
</tr>
<tr>
<td>.530+.003</td>
<td>.436</td>
<td>1.31×10⁻⁶ - .295</td>
<td>.01-3211.</td>
</tr>
<tr>
<td>.530+.003</td>
<td>.546</td>
<td>5.2×10⁻⁵ - .295</td>
<td>.25-1969.</td>
</tr>
</tbody>
</table>
A suspension of the desired PVC is produced by diluting the concentrated latex with de-ionized Millipore-filtered distilled water. The exact concentration is determined after a multiple-scatter run by evaporating under reduced pressure a known volume of the suspension and then weighing the residue.

The scattering test cells for the solutions of polystyrene latex spheres in water are constructed from parallel microscope cover classes cemented with epoxy resin to a lucite frame. The cell faces 5 cms square, and the clearance between the inner glass surfaces is varied from 0.05 to 0.20 cms. The thickness of the cover glasses is 0.15 cms.
VI. RESULTS

Measurements were made of the directional distribution and state of polarization of radiation scattered by polystyrene spheres for perimeter-to-wavelength ratios of .78 to 5.1, optical thickness up to 3000. The fixed parameters were particle refractive index (1.59-1.61), refractive index of the dispersion phase (1.333-1.34) and the refractive index of the confining glass slides (1.5). The results are presented in two sections: those for single scatter in section 1 and for multiple scatter in section 2.

6.1 Single Scatter

Single scatter measurements were performed on optically thin suspensions of particles in a Brice-Phoenix cylindrical cell. Unpolarized incident radiation was used. The two components (perpendicular and parallel) of the scattered beam were measured separately. The values of \( i_\perp(\theta) \) and \( i_\parallel(\theta) \) for the different angles \( \theta \) were calculated from the phototube output as outlined in Appendix F. Figures 6.3 and 6.4 show experimental data for two particle sizes \( d = .106\mu \) and \( .530\mu \) and two wavelengths \( \lambda = .4358\mu \) and \( .5461\mu \). The continuous curves correspond to the prediction of Mie equations using the diameters obtained from electron micrographs of samples of the lattices supplied by Dow Chemical Company (Figures 6.1 and 6.2).
Figure 6.1 Electron Micrograph of polystyrene spheres
Figure 6.2 Electron Micrograph of polystyrene spheres; $d = 0.248\mu$
Figure 6.3 Mie scattering coefficients, $i_\perp(\theta)$ and $i_\parallel(\theta)$: $d = 0.106\mu$, $n_2/n_1 = 1.19$. Data points represent experimental results, solid lines theory.
Figure 6.4 Mie scattering coefficients, $i_L(\theta)$ and $i_H(\theta)$: $d = 0.530 \mu$. Data points represent experimental results, solid lines theory.
6.2 Multiple Scatter

Measurements were performed on four particle sizes of diameter .102μ, .106μ, .248μ, .530μ, and two wavelengths .4358μ and .5461μ. The bidirectional reflectance and transmittance from suspensions of polystyrene spheres were calculated for normal incidence and unpolarized incident radiation. The phototube output was converted into the bidirectional intensity distribution using the procedure outlined in Appendix F.

The optical thicknesses (τ₁) studied were extended from very low values to several hundred by changing the distance between the glass plates of the scattering cell L (0.05-0.2cm) and the pigment volume concentration (10⁻⁶ to 0.3). The 0.3 pigment volume concentration result in separation distances between the particles of 0.36d. The literature survey has showed that interference effects depend on the center-to-center distance (δ) between the particles and on the ratio δ/λ. Wave interference of scattered radiation will change the angular distribution of the energy emerging from the boundaries of the slab. In the present work interference was checked by comparing bidirectional intensity data with values predicted by the transfer equation. Figure 6.5-6.6 show excellent agreement between theory and experiment. Experimental results in Figure 6.7 shows, however, significant discrepancies from theory. In all plots an abscissa of sin²θ is selected to give equal-area
weightings to equal energy increments. The left hand side of the plots correspond to the forward scattering and the right hand side to backward scattering. The area under the curves correspond to the sum of hemispherical diffuse reflectance \( R_D \) and transmittance \( T_D \). The amounts of radiation which are reflected at the entrance window of the cell (DR) and transmitted at 0 degrees DT are given by the following equations:

\[
\text{DR} = R_1 + \frac{(1-R_1)^2 R_1 e^{-2T_1}}{1-R_1^2 e^{-2T_1}}
\]

\[
\text{DT} = \frac{(1-R_1)^2 e^{-T_1}}{1-R_1^2 e^{-2T_1}}
\]

where

\[
R_1 = \rho_1 + \frac{(1 - \rho_1)^2 \rho_2}{1 - \rho_1 \rho_2}
\]

and \( \rho_1, \rho_2 \) are the reflectances at the air-glass, glass-water interfaces for normal incidence. For dielectric particles the sum of DT, DR, \( R_D \), \( T_D \) should equal to one and is given for all experiments performed in Tables 6.1-6.8.

A summary of the results of the integrated diffuse components of reflectance \( R_D \) and transmittance \( T_D \) (excluding the energy spikes along \( \theta = 0^o \) and \( 180^o \)) are shown for a particle size diameter of 0.530\( \mu \) and wavelengths of 0.4358\( \mu \), 0.5461\( \mu \) in Figure 6.8-6.9. The data points correspond to the measured
values, the solid line to calculated values; and both omit non-scattered contributions to \( R \) and \( T \).

The contribution \( T_D \) to the total transmittance is seen to go through a maximum at an optical density of about 3. Agreement would have been poor if allowance had not been made for suspension-glass and glass-air interfaces which trap radiation scattered to angles greater than of total internal reflection.
Figure 6.5 Bidirectional reflectance and transmittance as a function of P.V.C. of 0.106μ polystyrene spheres, L = 0.147cms. Data points represent experimental results, solid lines theory.
Figure 6.6 Bidirectional reflectance and transmittance as a function of P.V.C. of 0.530μ polystyrene spheres, L = 0.147cms. Data points represent experimental results, solid lines theory.
Figure 6.7 Bidirectional reflectance and transmittance as a function of P.V.C. of 0.102μ polystyrene spheres. Data points represent experimental results, drawn lines theory.
Figure 6.8 Integrated diffuse reflectance and transmittance as a function of optical thickness for a polystyrene latex 0.530μ in diameter and a λ = 0.4358μ. Data points experimental, solid lines theoretical.
Figure 6.9 Integrated diffuse reflectance and transmittance as a function of optical thickness: $d = 0.530\mu, \lambda = 0.546\mu$. Data points experimental, solid lines theoretical.
<table>
<thead>
<tr>
<th>RUN</th>
<th>PVC</th>
<th>δ/d</th>
<th>L</th>
<th>t_L</th>
<th>T_D</th>
<th>R_D</th>
<th>DT</th>
<th>DR</th>
<th>SUM</th>
</tr>
</thead>
<tbody>
<tr>
<td>74</td>
<td>3.53x10^{-5}</td>
<td>28</td>
<td>.0965</td>
<td>.0185</td>
<td>.0086</td>
<td>.0042</td>
<td>.9000</td>
<td>.08161</td>
<td>.9945</td>
</tr>
<tr>
<td>87</td>
<td>1.24x10^{-4}</td>
<td>18</td>
<td>.1473</td>
<td>.0995</td>
<td>.0324</td>
<td>.0208</td>
<td>.8305</td>
<td>.07520</td>
<td>.959</td>
</tr>
<tr>
<td>90</td>
<td>6.69x10^{-4}</td>
<td>10.3</td>
<td>.1473</td>
<td>.5371</td>
<td>.1842</td>
<td>.1500</td>
<td>.5352</td>
<td>.05688</td>
<td>.9263</td>
</tr>
<tr>
<td>91</td>
<td>2.77x10^{-3}</td>
<td>6.4</td>
<td>.1473</td>
<td>2.222</td>
<td>.4046</td>
<td>.4372</td>
<td>.0993</td>
<td>.04339</td>
<td>.9844</td>
</tr>
<tr>
<td>102</td>
<td>3.48x10^{-3}</td>
<td>6</td>
<td>.1473</td>
<td>2.795</td>
<td>.418</td>
<td>.486</td>
<td>.05565</td>
<td>.0433</td>
<td>1</td>
</tr>
<tr>
<td>113</td>
<td>5.91x10^{-3}</td>
<td>5</td>
<td>.1473</td>
<td>4.74</td>
<td>.3853</td>
<td>.6321</td>
<td>.00795</td>
<td>.04333</td>
<td>1.07</td>
</tr>
<tr>
<td>104</td>
<td>1.2x10^{-2}</td>
<td>4</td>
<td>.1473</td>
<td>9.62</td>
<td>.273</td>
<td>.808</td>
<td>0</td>
<td>.0433</td>
<td>1.123</td>
</tr>
<tr>
<td>105</td>
<td>2.72x10^{-2}</td>
<td>3</td>
<td>.1473</td>
<td>21.82</td>
<td>.142</td>
<td>.9022</td>
<td>0</td>
<td>.0429</td>
<td>1.087</td>
</tr>
<tr>
<td>116</td>
<td>0.092</td>
<td>2</td>
<td>.1473</td>
<td>73.88</td>
<td>.068</td>
<td>1.05</td>
<td>0</td>
<td>.0433</td>
<td>1.161</td>
</tr>
<tr>
<td>92</td>
<td>10^{-1}</td>
<td>1.94</td>
<td>.1473</td>
<td>80.23</td>
<td>.0571</td>
<td>.922</td>
<td>0</td>
<td>.0433</td>
<td>1.022</td>
</tr>
<tr>
<td>124</td>
<td>10^{-1}</td>
<td>1.94</td>
<td>.0511</td>
<td>27.81</td>
<td>.152</td>
<td>.847</td>
<td>0</td>
<td>.0433</td>
<td>1.042</td>
</tr>
<tr>
<td>123</td>
<td>10^{-1}</td>
<td>1.94</td>
<td>.0983</td>
<td>53.5</td>
<td>.0838</td>
<td>.915</td>
<td>0</td>
<td>.0433</td>
<td>1.042</td>
</tr>
</tbody>
</table>
### TABLE 6.2

d = 0.102 μ  λ = 0.4358 μ

<table>
<thead>
<tr>
<th>RUN</th>
<th>PVC</th>
<th>δ/d</th>
<th>L</th>
<th>τ₁</th>
<th>T_D</th>
<th>R_D</th>
<th>DT</th>
<th>DR</th>
<th>SUM</th>
</tr>
</thead>
<tbody>
<tr>
<td>207</td>
<td>0.0284</td>
<td>3</td>
<td>0.1012</td>
<td>14.26</td>
<td>0.1714</td>
<td>0.8286</td>
<td>0</td>
<td>0.0433</td>
<td>1.043</td>
</tr>
<tr>
<td>205</td>
<td>0.0284</td>
<td>3</td>
<td>0.1473</td>
<td>20.76</td>
<td>0.1393</td>
<td>0.8643</td>
<td>0</td>
<td>0.0433</td>
<td>1.046</td>
</tr>
<tr>
<td>203</td>
<td>0.0284</td>
<td>3</td>
<td>0.2032</td>
<td>28.64</td>
<td>0.1178</td>
<td>0.9464</td>
<td>0</td>
<td>0.0433</td>
<td>1.11</td>
</tr>
<tr>
<td>186</td>
<td>0.0909</td>
<td>2</td>
<td>0.061</td>
<td>27.53</td>
<td>0.1357</td>
<td>0.8357</td>
<td>0</td>
<td>0.0433</td>
<td>1.014</td>
</tr>
<tr>
<td>183</td>
<td>0.0909</td>
<td>2</td>
<td>0.1012</td>
<td>45.85</td>
<td>0.07678</td>
<td>0.9286</td>
<td>0</td>
<td>0.0433</td>
<td>1.048</td>
</tr>
<tr>
<td>181</td>
<td>0.0909</td>
<td>2</td>
<td>0.1473</td>
<td>66.48</td>
<td>0.06107</td>
<td>0.9428</td>
<td>0</td>
<td>0.0433</td>
<td>1.047</td>
</tr>
<tr>
<td>179</td>
<td>0.0909</td>
<td>2</td>
<td>0.2032</td>
<td>91.7</td>
<td>0.04286</td>
<td>0.8821</td>
<td>0</td>
<td>0.0433</td>
<td>0.968</td>
</tr>
<tr>
<td>193</td>
<td>0.1356</td>
<td>1.76</td>
<td>0.061</td>
<td>41.1</td>
<td>0.12143</td>
<td>0.875</td>
<td>0</td>
<td>0.0433</td>
<td>1.04</td>
</tr>
<tr>
<td>191</td>
<td>0.1356</td>
<td>1.76</td>
<td>0.1012</td>
<td>68.1</td>
<td>0.065</td>
<td>0.90357</td>
<td>0</td>
<td>0.0433</td>
<td>1.01</td>
</tr>
<tr>
<td>189</td>
<td>0.1356</td>
<td>1.76</td>
<td>0.1473</td>
<td>99.2</td>
<td>0.04914</td>
<td>0.9093</td>
<td>0</td>
<td>0.0429</td>
<td>1.001</td>
</tr>
<tr>
<td>187</td>
<td>0.1356</td>
<td>1.76</td>
<td>0.2032</td>
<td>137</td>
<td>0.03678</td>
<td>0.93214</td>
<td>0</td>
<td>0.0433</td>
<td>1.012</td>
</tr>
<tr>
<td>201</td>
<td>0.181</td>
<td>1.6</td>
<td>0.061</td>
<td>54.82</td>
<td>0.1107</td>
<td>0.8464</td>
<td>0</td>
<td>0.0433</td>
<td>1.0</td>
</tr>
<tr>
<td>199</td>
<td>0.181</td>
<td>1.6</td>
<td>0.1012</td>
<td>90.94</td>
<td>0.06428</td>
<td>0.9071</td>
<td>0</td>
<td>0.0433</td>
<td>1.014</td>
</tr>
<tr>
<td>197</td>
<td>0.181</td>
<td>1.6</td>
<td>0.1473</td>
<td>132.4</td>
<td>0.05071</td>
<td>0.9214</td>
<td>0</td>
<td>0.0433</td>
<td>1.015</td>
</tr>
<tr>
<td>195</td>
<td>0.181</td>
<td>1.6</td>
<td>0.2032</td>
<td>182.6</td>
<td>0.035</td>
<td>0.9036</td>
<td>0</td>
<td>0.0433</td>
<td>0.9816</td>
</tr>
<tr>
<td>213</td>
<td>0.2195</td>
<td>1.5</td>
<td>0.1012</td>
<td>110.3</td>
<td>0.07357</td>
<td>0.95714</td>
<td>0</td>
<td>0.0433</td>
<td>1.074</td>
</tr>
<tr>
<td>211</td>
<td>0.2195</td>
<td>1.5</td>
<td>0.1473</td>
<td>160.5</td>
<td>0.05821</td>
<td>0.975</td>
<td>0</td>
<td>0.0433</td>
<td>1.076</td>
</tr>
<tr>
<td>209</td>
<td>0.2195</td>
<td>1.5</td>
<td>0.2032</td>
<td>221.4</td>
<td>0.03857</td>
<td>0.900</td>
<td>0</td>
<td>0.0433</td>
<td>0.9815</td>
</tr>
<tr>
<td>250</td>
<td>0.2893</td>
<td>1.37</td>
<td>0.1012</td>
<td>145.4</td>
<td>0.07821</td>
<td>0.75</td>
<td>0</td>
<td>0.0433</td>
<td>0.8712</td>
</tr>
<tr>
<td>248</td>
<td>0.2893</td>
<td>1.37</td>
<td>0.1372</td>
<td>197.1</td>
<td>0.0575</td>
<td>0.7321</td>
<td>0</td>
<td>0.0433</td>
<td>0.8326</td>
</tr>
<tr>
<td>246</td>
<td>0.2893</td>
<td>1.37</td>
<td>0.2032</td>
<td>292</td>
<td>0.03957</td>
<td>0.7622</td>
<td>0</td>
<td>0.0429</td>
<td>0.8447</td>
</tr>
</tbody>
</table>
TABLE 6.3

d = .106µm  λ = .5461µm

<table>
<thead>
<tr>
<th>RUN</th>
<th>PVC</th>
<th>δ/d</th>
<th>L</th>
<th>T₁</th>
<th>TD</th>
<th>RD</th>
<th>DT</th>
<th>DR</th>
<th>SUM</th>
</tr>
</thead>
<tbody>
<tr>
<td>93</td>
<td>3.08x10⁻⁴</td>
<td>13.4</td>
<td>.1473</td>
<td>.0941</td>
<td>.03010</td>
<td>.02410</td>
<td>.8345</td>
<td>.07602</td>
<td>.9647</td>
</tr>
<tr>
<td>107</td>
<td>3.5x10⁻³</td>
<td>6</td>
<td>.1473</td>
<td>1.070</td>
<td>.292</td>
<td>.293</td>
<td>.3142</td>
<td>.0479</td>
<td>.9470</td>
</tr>
<tr>
<td>85</td>
<td>4.5x10⁻³</td>
<td>5.5</td>
<td>.0965</td>
<td>.9008</td>
<td>.2749</td>
<td>.2704</td>
<td>.3719</td>
<td>.04987</td>
<td>.9671</td>
</tr>
<tr>
<td>108</td>
<td>5.9x10⁻³</td>
<td>5</td>
<td>.1473</td>
<td>1.78</td>
<td>.346</td>
<td>.386</td>
<td>.1543</td>
<td>.0445</td>
<td>.931</td>
</tr>
<tr>
<td>109</td>
<td>1.2x10⁻²</td>
<td>4</td>
<td>.1473</td>
<td>3.61</td>
<td>.364</td>
<td>.547</td>
<td>.025</td>
<td>.0433</td>
<td>.9743</td>
</tr>
<tr>
<td>110</td>
<td>2.7x10⁻²</td>
<td>3</td>
<td>.1473</td>
<td>8.25</td>
<td>.260</td>
<td>.7037</td>
<td>0</td>
<td>.0432</td>
<td>1.007</td>
</tr>
<tr>
<td>111</td>
<td>.092</td>
<td>2</td>
<td>.1473</td>
<td>27.72</td>
<td>.146</td>
<td>.822</td>
<td>0</td>
<td>.0433</td>
<td>1.011</td>
</tr>
<tr>
<td>97</td>
<td>10⁻¹</td>
<td>1.94</td>
<td>.1473</td>
<td>30.1</td>
<td>.137</td>
<td>.784</td>
<td>0</td>
<td>.0433</td>
<td>.96</td>
</tr>
</tbody>
</table>
### TABLE 6.4

\( d = 0.102 \mu \quad \lambda = 0.546 \mu \)

<table>
<thead>
<tr>
<th>RUN</th>
<th>PVC</th>
<th>( \delta/d )</th>
<th>L</th>
<th>( T_1 )</th>
<th>( T_D )</th>
<th>R_D</th>
<th>DT</th>
<th>DR</th>
<th>SUM</th>
</tr>
</thead>
<tbody>
<tr>
<td>208</td>
<td>.0284</td>
<td>3</td>
<td>.1012</td>
<td>5.38</td>
<td>.31428</td>
<td>.675</td>
<td>.00502</td>
<td>.0433</td>
<td>1.032</td>
</tr>
<tr>
<td>206</td>
<td>.0284</td>
<td>3</td>
<td>.1473</td>
<td>7.83</td>
<td>.2786</td>
<td>.7357</td>
<td>.00041</td>
<td>.0433</td>
<td>1.058</td>
</tr>
<tr>
<td>204</td>
<td>.0284</td>
<td>3</td>
<td>.2032</td>
<td>10.8</td>
<td>.225</td>
<td>.7893</td>
<td>0</td>
<td>.0433</td>
<td>1.057</td>
</tr>
<tr>
<td>185</td>
<td>.0909</td>
<td>2</td>
<td>.061</td>
<td>10.38</td>
<td>.2893</td>
<td>.6821</td>
<td>0</td>
<td>.0433</td>
<td>1.014</td>
</tr>
<tr>
<td>184</td>
<td>.0909</td>
<td>2</td>
<td>.1012</td>
<td>17.29</td>
<td>.1857</td>
<td>.7821</td>
<td>0</td>
<td>.0433</td>
<td>1.01</td>
</tr>
<tr>
<td>182</td>
<td>.0909</td>
<td>2</td>
<td>.1473</td>
<td>25.07</td>
<td>.1571</td>
<td>.8321</td>
<td>0</td>
<td>.0433</td>
<td>1.03</td>
</tr>
<tr>
<td>180</td>
<td>.0909</td>
<td>2</td>
<td>.2032</td>
<td>34.58</td>
<td>.1286</td>
<td>.8786</td>
<td>0</td>
<td>.0433</td>
<td>1.05</td>
</tr>
<tr>
<td>194</td>
<td>.1356</td>
<td>1.76</td>
<td>.061</td>
<td>15.48</td>
<td>.26785</td>
<td>.6893</td>
<td>0</td>
<td>.0433</td>
<td>1.0</td>
</tr>
<tr>
<td>192</td>
<td>.1356</td>
<td>1.76</td>
<td>.1012</td>
<td>25.69</td>
<td>.175</td>
<td>.800</td>
<td>0</td>
<td>.0433</td>
<td>1.02</td>
</tr>
<tr>
<td>190</td>
<td>.1356</td>
<td>1.76</td>
<td>.1473</td>
<td>37.39</td>
<td>.1429</td>
<td>.8214</td>
<td>0</td>
<td>.0433</td>
<td>1.0</td>
</tr>
<tr>
<td>188</td>
<td>.1356</td>
<td>1.76</td>
<td>.2032</td>
<td>51.58</td>
<td>.1143</td>
<td>.8536</td>
<td>0</td>
<td>.0433</td>
<td>1.01</td>
</tr>
<tr>
<td>202</td>
<td>.181</td>
<td>1.6</td>
<td>.061</td>
<td>20.67</td>
<td>.2821</td>
<td>.6928</td>
<td>0</td>
<td>.0433</td>
<td>1.02</td>
</tr>
<tr>
<td>200</td>
<td>.181</td>
<td>1.6</td>
<td>.1012</td>
<td>34.29</td>
<td>.1857</td>
<td>.7786</td>
<td>0</td>
<td>.0433</td>
<td>1.01</td>
</tr>
<tr>
<td>198</td>
<td>.181</td>
<td>1.6</td>
<td>.1473</td>
<td>49.91</td>
<td>.1511</td>
<td>.8268</td>
<td>0</td>
<td>.0432</td>
<td>1.021</td>
</tr>
<tr>
<td>196</td>
<td>.181</td>
<td>1.6</td>
<td>.2032</td>
<td>68.85</td>
<td>.1214</td>
<td>.8571</td>
<td>0</td>
<td>.0433</td>
<td>1.02</td>
</tr>
<tr>
<td>214</td>
<td>.2195</td>
<td>1.5</td>
<td>.1012</td>
<td>41.58</td>
<td>.2036</td>
<td>.8071</td>
<td>0</td>
<td>.0433</td>
<td>1.05</td>
</tr>
<tr>
<td>212</td>
<td>.2195</td>
<td>1.5</td>
<td>.1473</td>
<td>60.53</td>
<td>.175</td>
<td>.8610</td>
<td>0</td>
<td>.0433</td>
<td>1.08</td>
</tr>
<tr>
<td>210</td>
<td>.2195</td>
<td>1.5</td>
<td>.2032</td>
<td>83.5</td>
<td>.1214</td>
<td>.7964</td>
<td>0</td>
<td>.0433</td>
<td>.96</td>
</tr>
<tr>
<td>251</td>
<td>.2893</td>
<td>1.37</td>
<td>.1012</td>
<td>54.8</td>
<td>.2357</td>
<td>.6786</td>
<td>0</td>
<td>.0433</td>
<td>.957</td>
</tr>
<tr>
<td>249</td>
<td>.2893</td>
<td>1.37</td>
<td>.1372</td>
<td>74.3</td>
<td>.2000</td>
<td>.7071</td>
<td>0</td>
<td>.0433</td>
<td>.95</td>
</tr>
<tr>
<td>247</td>
<td>.2893</td>
<td>1.37</td>
<td>.2032</td>
<td>110</td>
<td>.1389</td>
<td>.7531</td>
<td>0</td>
<td>.0432</td>
<td>.935</td>
</tr>
</tbody>
</table>
### TABLE 6.5

\(d = 0.248 \mu \quad \lambda = 0.4358 \mu\)

<table>
<thead>
<tr>
<th>RUN</th>
<th>PVC</th>
<th>(\delta/d)</th>
<th>L</th>
<th>(T_1)</th>
<th>(T_D)</th>
<th>(R_D)</th>
<th>DT</th>
<th>DR</th>
<th>SUM</th>
</tr>
</thead>
<tbody>
<tr>
<td>264</td>
<td>0.1046</td>
<td>1.92</td>
<td>0.1092</td>
<td>296</td>
<td>0.02745</td>
<td>0.8325</td>
<td>0</td>
<td>0.0433</td>
<td>0.9029</td>
</tr>
<tr>
<td>263</td>
<td>0.1046</td>
<td>1.92</td>
<td>0.1473</td>
<td>391</td>
<td>0.02714</td>
<td>0.87142</td>
<td>0</td>
<td>0.0433</td>
<td>0.942</td>
</tr>
<tr>
<td>260</td>
<td>0.1046</td>
<td>1.92</td>
<td>0.2032</td>
<td>551</td>
<td>0.01551</td>
<td>0.85190</td>
<td>0</td>
<td>0.0433</td>
<td>0.9104</td>
</tr>
<tr>
<td>259</td>
<td>0.293</td>
<td>1.36</td>
<td>0.0480</td>
<td>357</td>
<td>0.03571</td>
<td>0.85356</td>
<td>0</td>
<td>0.0433</td>
<td>0.932</td>
</tr>
<tr>
<td>256</td>
<td>0.293</td>
<td>1.36</td>
<td>0.1092</td>
<td>812</td>
<td>0.01536</td>
<td>0.86785</td>
<td>0</td>
<td>0.0433</td>
<td>0.926</td>
</tr>
<tr>
<td>255</td>
<td>0.293</td>
<td>1.36</td>
<td>0.1473</td>
<td>1096</td>
<td>0.01134</td>
<td>0.8838</td>
<td>0</td>
<td>0.0433</td>
<td>0.938</td>
</tr>
</tbody>
</table>

### TABLE 6.6

\(d = 0.248 \mu \quad \lambda = 0.5461 \mu\)

<table>
<thead>
<tr>
<th>RUN</th>
<th>PVC</th>
<th>(\delta/d)</th>
<th>L</th>
<th>(T_1)</th>
<th>(T_D)</th>
<th>(R_D)</th>
<th>DT</th>
<th>DR</th>
<th>SUM</th>
</tr>
</thead>
<tbody>
<tr>
<td>265</td>
<td>0.1046</td>
<td>1.92</td>
<td>0.1092</td>
<td>138</td>
<td>0.04893</td>
<td>0.92142</td>
<td>0</td>
<td>0.0433</td>
<td>1.013</td>
</tr>
<tr>
<td>262</td>
<td>0.1046</td>
<td>1.92</td>
<td>0.1473</td>
<td>186</td>
<td>0.04036</td>
<td>0.90713</td>
<td>0</td>
<td>0.0433</td>
<td>0.9905</td>
</tr>
<tr>
<td>261</td>
<td>0.1046</td>
<td>1.92</td>
<td>0.2032</td>
<td>257</td>
<td>0.02857</td>
<td>0.93213</td>
<td>0</td>
<td>0.0433</td>
<td>1.004</td>
</tr>
<tr>
<td>258</td>
<td>0.293</td>
<td>1.36</td>
<td>0.048</td>
<td>170</td>
<td>0.04678</td>
<td>0.90713</td>
<td>0</td>
<td>0.0433</td>
<td>0.9669</td>
</tr>
<tr>
<td>257</td>
<td>0.293</td>
<td>1.36</td>
<td>0.1092</td>
<td>386</td>
<td>0.02357</td>
<td>0.92856</td>
<td>0</td>
<td>0.0433</td>
<td>0.9951</td>
</tr>
<tr>
<td>254</td>
<td>0.293</td>
<td>1.36</td>
<td>0.1473</td>
<td>521</td>
<td>0.01714</td>
<td>0.87142</td>
<td>0</td>
<td>0.0433</td>
<td>0.932</td>
</tr>
<tr>
<td>266</td>
<td>0.293</td>
<td>1.36</td>
<td>0.2032</td>
<td>719</td>
<td>0.01159</td>
<td>0.8582</td>
<td>0</td>
<td>0.0433</td>
<td>0.9131</td>
</tr>
<tr>
<td>267</td>
<td>0.293</td>
<td>1.36</td>
<td>0.1473</td>
<td>521</td>
<td>0.01728</td>
<td>0.8501</td>
<td>0</td>
<td>0.0433</td>
<td>0.9107</td>
</tr>
<tr>
<td>268</td>
<td>0.293</td>
<td>1.36</td>
<td>0.1092</td>
<td>386</td>
<td>0.02243</td>
<td>0.8652</td>
<td>0</td>
<td>0.0433</td>
<td>0.931</td>
</tr>
</tbody>
</table>
### TABLE 6.7

\(d = .530\mu \quad \lambda = .4358\mu\)

<table>
<thead>
<tr>
<th>RUN</th>
<th>PVC</th>
<th>(\delta/d)</th>
<th>L</th>
<th>(t^1)</th>
<th>TD</th>
<th>RD</th>
<th>DT</th>
<th>DR</th>
<th>SUM</th>
</tr>
</thead>
<tbody>
<tr>
<td>40</td>
<td>1.316\times10^{-6}</td>
<td>83</td>
<td>.1473</td>
<td>.0102</td>
<td>.01017</td>
<td>.00092</td>
<td>.9083</td>
<td>.08152</td>
<td>1.001</td>
</tr>
<tr>
<td>51</td>
<td>2.603\times10^{-6}</td>
<td>65</td>
<td>.1473</td>
<td>.0202</td>
<td>.02112</td>
<td>.00234</td>
<td>.8985</td>
<td>.08148</td>
<td>1.003</td>
</tr>
<tr>
<td>36</td>
<td>3.76\times10^{-6}</td>
<td>58</td>
<td>.0508</td>
<td>.01</td>
<td>.00933</td>
<td>.00095</td>
<td>.9077</td>
<td>.08226</td>
<td>1.0</td>
</tr>
<tr>
<td>52</td>
<td>3.452\times10^{-5}</td>
<td>27.5</td>
<td>.1473</td>
<td>.2667</td>
<td>.239</td>
<td>.024</td>
<td>.7023</td>
<td>.06602</td>
<td>1.031</td>
</tr>
<tr>
<td>41</td>
<td>3.645\times10^{-5}</td>
<td>28</td>
<td>.1473</td>
<td>.2828</td>
<td>.2286</td>
<td>.02446</td>
<td>.6918</td>
<td>.0659</td>
<td>1.01</td>
</tr>
<tr>
<td>16</td>
<td>9.52\times10^{-5}</td>
<td>19.8</td>
<td>.094</td>
<td>.499</td>
<td>.350</td>
<td>.0309</td>
<td>.5551</td>
<td>.05787</td>
<td>.9938</td>
</tr>
<tr>
<td>37</td>
<td>1.04\times10^{-4}</td>
<td>19.5</td>
<td>.0508</td>
<td>.2799</td>
<td>.2036</td>
<td>.02375</td>
<td>.6918</td>
<td>.0659</td>
<td>.985</td>
</tr>
<tr>
<td>48</td>
<td>1.402\times10^{-4}</td>
<td>17.4</td>
<td>.1473</td>
<td>1.088</td>
<td>.5701</td>
<td>.0642</td>
<td>.3087</td>
<td>.04739</td>
<td>.9905</td>
</tr>
<tr>
<td>47(B)</td>
<td>3.69\times10^{-4}</td>
<td>12</td>
<td>.056</td>
<td>1.086</td>
<td>.5731</td>
<td>.0687</td>
<td>.3090</td>
<td>.04785</td>
<td>.9987</td>
</tr>
<tr>
<td>49</td>
<td>3.69\times10^{-4}</td>
<td>12</td>
<td>.1473</td>
<td>2.8602</td>
<td>.675</td>
<td>.125</td>
<td>.05242</td>
<td>.0433</td>
<td>.895</td>
</tr>
<tr>
<td>53</td>
<td>3.452\times10^{-4}</td>
<td>13</td>
<td>.1473</td>
<td>2.667</td>
<td>.7144</td>
<td>.1586</td>
<td>.06363</td>
<td>.04312</td>
<td>.9797</td>
</tr>
<tr>
<td>58</td>
<td>4.818\times10^{-3}</td>
<td>5.4</td>
<td>.1473</td>
<td>37.38</td>
<td>.3681</td>
<td>.6197</td>
<td>0</td>
<td>.0429</td>
<td>1.03</td>
</tr>
<tr>
<td>117</td>
<td>2.784\times10^{-2}</td>
<td>3</td>
<td>.1473</td>
<td>215.9</td>
<td>.090</td>
<td>.827</td>
<td>0</td>
<td>.0433</td>
<td>.96</td>
</tr>
<tr>
<td>118</td>
<td>9.208\times10^{-2}</td>
<td>2</td>
<td>.1473</td>
<td>713.9</td>
<td>.03</td>
<td>.880</td>
<td>0</td>
<td>.0433</td>
<td>.953</td>
</tr>
<tr>
<td>125</td>
<td>.944\times10^{-1}</td>
<td>1.98</td>
<td>.0508</td>
<td>252.9</td>
<td>.09089</td>
<td>.925</td>
<td>0</td>
<td>.0433</td>
<td>1.059</td>
</tr>
<tr>
<td>128</td>
<td>.944\times10^{-1}</td>
<td>1.98</td>
<td>.1067</td>
<td>529.9</td>
<td>.0435</td>
<td>.8950</td>
<td>0</td>
<td>.0433</td>
<td>.9815</td>
</tr>
<tr>
<td>129</td>
<td>.944\times10^{-1}</td>
<td>1.98</td>
<td>.1473</td>
<td>731.5</td>
<td>.0335</td>
<td>.940</td>
<td>0</td>
<td>.0433</td>
<td>1.0165</td>
</tr>
<tr>
<td>131</td>
<td>.944\times10^{-1}</td>
<td>1.98</td>
<td>.2032</td>
<td>1010</td>
<td>.0234</td>
<td>.9403</td>
<td>0</td>
<td>.0429</td>
<td>1.007</td>
</tr>
<tr>
<td>119</td>
<td>10^{-1}</td>
<td>1.94</td>
<td>.1473</td>
<td>775.3</td>
<td>.028</td>
<td>.919</td>
<td>0</td>
<td>.0433</td>
<td>.99</td>
</tr>
</tbody>
</table>
| 133  | .111 | 1.88 | .0508 | 297   | .0793 | .9285 | 0 | .0433 | 1.0508
| 135  | .111 | 1.88 | .1067 | 623.7 | .04143 | 1.0286 | 0 | .0433 | 1.113
| 137  | .111 | 1.88 | .1473 | 861   | .03143 | 1.0464 | 0 | .0433 | 1.1208
| 140  | .111 | 1.88 | .2032 | 1188  | .01821 | .86071 | 0 | .0433 | .9219
| 141  | .1267| 1.8  | .0560 | 373   | .06536 | .975  | 0 | .0433 | 1.0834
| 144  | .1267| 1.8  | .1067 | 713.4 | .0325  | .975  | 0 | .0433 | 1.051
| 145  | .1267| 1.8  | .1473 | 983   | .02607 | .98928 | 0 | .0433 | 1.058
| 147  | .1267| 1.8  | .2032 | 1356  | .01571 | .83125 | 0 | .0433 | .89086
| 156  | .1504| 1.7  | .061  | 482   | .04286 | .9367 | 0 | .0433 | 1.0216
| 153  | .1504| 1.7  | .1067 | 845   | .02928 | 1.0393 | 0 | .0433 | 1.116
| 152  | .1504| 1.7  | .1473 | 1167  | .02286 | 1.0857 | 0 | .0433 | 1.1516
| 149  | .1504| 1.7  | .2032 | 1610  | .01571 | 1.0678 | 0 | .0433 | 1.1265
| 161  | .177 | 1.61 | .1012 | .943  | .025  | 1.0071 | 0 | .0433 | 1.0751
| 159  | .177 | 1.61 | .1473 | 1373  | .01678 | .9321 | 0 | .0433 | .9919
| 157  | .177 | 1.61 | .2032 | 1894  | .01143 | .96785 | 0 | .0433 | 1.0223
| 168  | .2172| 1.5  | .1012 | 1158  | .01857 | .90357 | 0 | .0433 | .9651
| 166  | .2172| 1.5  | .1473 | 1685  | .01428 | .9607 | 0 | .0433 | 1.018
| 172  | .2172| 1.5  | .2032 | 2324  | .00911 | .9286 | 0 | .0433 | .9807
| 237  | .2248| 1.49 | .1012 | 1198  | .01893 | .95714 | 0 | .0433 | 1.02
| 235  | .2248| 1.49 | .1372 | 1624  | .01536 | .98214 | 0 | .0433 | 1.04
| 234  | .2248| 1.49 | .2032 | 2406  | .00732 | .81428 | 0 | .0433 | .8646
| 243  | .2695| 1.4  | .1012 | 1436  | .01446 | .83571 | 0 | .0433 | .89317
| 241  | .2695| 1.4  | .1372 | 1947  | .01143 | .88928 | 0 | .0433 | .9437
| 177  | .2947| 1.36 | .1012 | 1571  | .01464 | .91428 | 0 | .0433 | .9719
| 175  | .2947| 1.36 | .1473 | 2286  | .01035 | .9488 | 0 | .0433 | 1.002
<table>
<thead>
<tr>
<th>RUN</th>
<th>PVC</th>
<th>$\delta/d$</th>
<th>$L$</th>
<th>$\tau_L$</th>
<th>$\tau_D$</th>
<th>$R_D$</th>
<th>DT</th>
<th>DR</th>
<th>SUM</th>
</tr>
</thead>
<tbody>
<tr>
<td>99</td>
<td>5.18x10^{-5}</td>
<td>25</td>
<td>.1473</td>
<td>.2473</td>
<td>.2239</td>
<td>.02448</td>
<td>.7157</td>
<td>.0673</td>
<td>1.031</td>
</tr>
<tr>
<td>100</td>
<td>8.62x10^{-4}</td>
<td>9.5</td>
<td>.1473</td>
<td>4.1</td>
<td>.7238</td>
<td>.2315</td>
<td>.01487</td>
<td>.04321</td>
<td>1.013</td>
</tr>
<tr>
<td>120</td>
<td>2.78x10^{-2}</td>
<td>3</td>
<td>.1473</td>
<td>132.3</td>
<td>.1068</td>
<td>.8162</td>
<td>0</td>
<td>.0433</td>
<td>.9663</td>
</tr>
<tr>
<td>121</td>
<td>9.21x10^{-2}</td>
<td>2</td>
<td>.1473</td>
<td>438.1</td>
<td>.0365</td>
<td>.836</td>
<td>0</td>
<td>.0433</td>
<td>.9155</td>
</tr>
<tr>
<td>126</td>
<td>.0944</td>
<td>1.98</td>
<td>.0508</td>
<td>154.8</td>
<td>.1098</td>
<td>.925</td>
<td>0</td>
<td>.0433</td>
<td>1.078</td>
</tr>
<tr>
<td>127</td>
<td>.0944</td>
<td>1.98</td>
<td>.1067</td>
<td>324.9</td>
<td>.056</td>
<td>.94</td>
<td>0</td>
<td>.0433</td>
<td>1.039</td>
</tr>
<tr>
<td>130</td>
<td>.0944</td>
<td>1.98</td>
<td>.1473</td>
<td>449.4</td>
<td>.0421</td>
<td>.94</td>
<td>0</td>
<td>.0433</td>
<td>1.025</td>
</tr>
<tr>
<td>132</td>
<td>.0944</td>
<td>1.98</td>
<td>.2032</td>
<td>618.8</td>
<td>.0316</td>
<td>.96</td>
<td>0</td>
<td>.0433</td>
<td>1.035</td>
</tr>
<tr>
<td>122</td>
<td>10^{-1}</td>
<td>1.94</td>
<td>.1473</td>
<td>475.7</td>
<td>.03714</td>
<td>.92</td>
<td>0</td>
<td>.0433</td>
<td>1</td>
</tr>
<tr>
<td>134</td>
<td>.111</td>
<td>1.88</td>
<td>.0508</td>
<td>182</td>
<td>.1023</td>
<td>1.004</td>
<td>0</td>
<td>.0433</td>
<td>1.149</td>
</tr>
<tr>
<td>136</td>
<td>.111</td>
<td>1.88</td>
<td>.1067</td>
<td>382.4</td>
<td>.0532</td>
<td>1.046</td>
<td>0</td>
<td>.0433</td>
<td>1.143</td>
</tr>
<tr>
<td>138</td>
<td>.111</td>
<td>1.88</td>
<td>.1473</td>
<td>524.7</td>
<td>.0359</td>
<td>.9420</td>
<td>0</td>
<td>.0433</td>
<td>1.021</td>
</tr>
<tr>
<td>139</td>
<td>.111</td>
<td>1.88</td>
<td>.2032</td>
<td>728.3</td>
<td>.0261</td>
<td>.9143</td>
<td>0</td>
<td>.0433</td>
<td>.9834</td>
</tr>
<tr>
<td>142</td>
<td>.1267</td>
<td>1.8</td>
<td>.0511</td>
<td>209.1</td>
<td>.0746</td>
<td>.9214</td>
<td>0</td>
<td>.0433</td>
<td>1.039</td>
</tr>
<tr>
<td>143</td>
<td>.1267</td>
<td>1.8</td>
<td>.1048</td>
<td>428.8</td>
<td>.0421</td>
<td>.9928</td>
<td>0</td>
<td>.0433</td>
<td>1.078</td>
</tr>
<tr>
<td>146</td>
<td>.1267</td>
<td>1.8</td>
<td>.1473</td>
<td>6027</td>
<td>.0353</td>
<td>1.032</td>
<td>0</td>
<td>.0433</td>
<td>1.11</td>
</tr>
<tr>
<td>148</td>
<td>.1267</td>
<td>1.8</td>
<td>.2032</td>
<td>831.4</td>
<td>.0243</td>
<td>.9678</td>
<td>0</td>
<td>.0433</td>
<td>1.035</td>
</tr>
<tr>
<td>154</td>
<td>.1504</td>
<td>1.7</td>
<td>.1016</td>
<td>493.4</td>
<td>.0336</td>
<td>.8964</td>
<td>0</td>
<td>.0433</td>
<td>.973</td>
</tr>
<tr>
<td>151</td>
<td>.1504</td>
<td>1.7</td>
<td>.1473</td>
<td>714</td>
<td>.0268</td>
<td>.925</td>
<td>0</td>
<td>.0433</td>
<td>.9948</td>
</tr>
<tr>
<td>150</td>
<td>.1504</td>
<td>1.7</td>
<td>.2032</td>
<td>984</td>
<td>.0196</td>
<td>.925</td>
<td>0</td>
<td>.0433</td>
<td>.9876</td>
</tr>
<tr>
<td>164</td>
<td>.177</td>
<td>1.61</td>
<td>.061</td>
<td>353</td>
<td>.0554</td>
<td>.8893</td>
<td>0</td>
<td>.0433</td>
<td>.9877</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td>---</td>
<td></td>
<td></td>
</tr>
<tr>
<td>162</td>
<td>0.177</td>
<td>1.61</td>
<td>0.1012</td>
<td>578</td>
<td>0.0296</td>
<td>0.9107</td>
<td>0</td>
<td>0.0433</td>
<td>0.9833</td>
</tr>
<tr>
<td>160</td>
<td>0.177</td>
<td>1.61</td>
<td>0.1473</td>
<td>842</td>
<td>0.0239</td>
<td>0.9607</td>
<td>0</td>
<td>0.0433</td>
<td>1.027</td>
</tr>
<tr>
<td>158</td>
<td>0.177</td>
<td>1.61</td>
<td>0.2032</td>
<td>1161</td>
<td>0.0184</td>
<td>1.026</td>
<td>0</td>
<td>0.0432</td>
<td>1.088</td>
</tr>
<tr>
<td>171</td>
<td>0.2172</td>
<td>1.5</td>
<td>0.061</td>
<td>428</td>
<td>0.04964</td>
<td>0.9357</td>
<td>0</td>
<td>0.0433</td>
<td>0.9962</td>
</tr>
<tr>
<td>169</td>
<td>0.2172</td>
<td>1.5</td>
<td>0.1012</td>
<td>710</td>
<td>0.025</td>
<td>0.91428</td>
<td>0</td>
<td>0.0433</td>
<td>0.9823</td>
</tr>
<tr>
<td>167</td>
<td>0.2172</td>
<td>1.5</td>
<td>0.1473</td>
<td>1033</td>
<td>0.02143</td>
<td>1.0035</td>
<td>0</td>
<td>0.0433</td>
<td>1.068</td>
</tr>
<tr>
<td>238</td>
<td>0.2248</td>
<td>1.49</td>
<td>0.1012</td>
<td>734.6</td>
<td>0.02428</td>
<td>0.86785</td>
<td>0</td>
<td>0.0433</td>
<td>0.935</td>
</tr>
<tr>
<td>236</td>
<td>0.2248</td>
<td>1.49</td>
<td>0.1372</td>
<td>996</td>
<td>0.01928</td>
<td>0.82142</td>
<td>0</td>
<td>0.0433</td>
<td>0.8837</td>
</tr>
<tr>
<td>233</td>
<td>0.2248</td>
<td>1.49</td>
<td>0.2032</td>
<td>1475</td>
<td>0.01268</td>
<td>0.79642</td>
<td>0</td>
<td>0.0433</td>
<td>0.8521</td>
</tr>
<tr>
<td>244</td>
<td>0.2695</td>
<td>1.4</td>
<td>0.1012</td>
<td>880.7</td>
<td>0.02107</td>
<td>0.934</td>
<td>0</td>
<td>0.0433</td>
<td>0.998</td>
</tr>
<tr>
<td>242</td>
<td>0.2695</td>
<td>1.4</td>
<td>0.1372</td>
<td>1194</td>
<td>0.0175</td>
<td>0.938</td>
<td>0</td>
<td>0.0433</td>
<td>0.9988</td>
</tr>
<tr>
<td>239</td>
<td>0.2695</td>
<td>1.4</td>
<td>0.2032</td>
<td>1768</td>
<td>0.01143</td>
<td>0.9400</td>
<td>0</td>
<td>0.0433</td>
<td>0.9947</td>
</tr>
<tr>
<td>232</td>
<td>0.2947</td>
<td>1.36</td>
<td>0.1012</td>
<td>980</td>
<td>0.02071</td>
<td>0.85357</td>
<td>0</td>
<td>0.0433</td>
<td>0.9173</td>
</tr>
<tr>
<td>230</td>
<td>0.2947</td>
<td>1.36</td>
<td>0.1372</td>
<td>1329</td>
<td>0.01643</td>
<td>0.88571</td>
<td>0</td>
<td>0.0433</td>
<td>0.9451</td>
</tr>
<tr>
<td>228</td>
<td>0.2947</td>
<td>1.36</td>
<td>0.2032</td>
<td>1934</td>
<td>0.01118</td>
<td>0.9027</td>
<td>0</td>
<td>0.0432</td>
<td>0.9571</td>
</tr>
</tbody>
</table>
VII. DISCUSSION OF RESULTS

The results will be discussed in four parts: (a) single scatter experiments in section 7.1, (b) multiple scatter experiments in agreement with the transfer equation in section 7.2, (c) multiple scatter where interference is present in section 7.3 and practical applications in section 7.4.

7.1 Single Scatter

Mie theory \(^1\) has been successfully used several times in the past in order to find the dimensions and concentration of spherical particles in a colloidal or aerosol system. Its validity has been established with no exceptions. This argument led to single scatter experiments in order to check the particles sizes of polystyrene spheres produced by Dow Chemical Company.

The theory behind a single scatter experiment was outlined in Chapter III. For a colloidal suspension of \( N_p \) scattering monodispersed particles per unit volume, irradiated with a beam attenuated less than 10\% the two plane polarized light-scattering intensities, \( I_\perp \) and \( I_{\parallel} \), follow from equations:

\[
I_\perp = N_p \frac{i_\perp}{k^2 r^2} I_{o\perp} \tag{7-1}
\]

\[
I_{\parallel} = N_p \frac{i_{\parallel}}{k^2 r^2} I_{o\parallel} \tag{7-2}
\]
Equations (7-1) and (7-2) have been modified to take into account the geometrical characteristics of the scattering cell and the equipment. Details are given in Appendix F.

In any given experiment the following parameters are fixed: $N_p$, $r$, $k(=2\pi/\lambda)$ ($\lambda$ is the wavelength transmitted through the narrow band interference filter), the state of polarization (known from the setting of the polarization filter), $I_{o\perp}$, and $I_{o\parallel}$. $I_{\perp}$ and $I_{\parallel}$ are measured. If the relative refractive index $m$ is known the shape of the scattering pattern of $I_{\perp}$ and $I_{\parallel}$ against $\theta$ (the angle to the normal, forward) will be determined by particle diameter $d$ only, since $i_{\perp}$ and $i_{\parallel}$ are functions of $\pi d/\lambda$, $m$ and $\theta$. A comparison of the measured scattered light intensity function $i_{\perp}$ and $i_{\parallel}$ with the various calculated theoretical distributions of $i_{\perp}$ and $i_{\parallel}(31)$ for a given value of $m$ but various value of $x$ gives the particle diameter $d$, since $\lambda$ is known.

The values of $i_{\perp}(\theta)$ and $i_{\parallel}(\theta)$ calculated from the scatter measurements are compared with the predictions of the Mie equations in Figures 6.3 and 6.4 for two particle sizes ($d=0.106$ and 0.530$\mu$) and two wavelengths ($\lambda=0.4358$ and 0.5461$\mu$). The agreement between theory and experiment is considered to be good, the small discrepancies resulting from a combination of the following factors: (a) the spread in particle sizes, (b) the finite bandwidth of the monochromatic filter 65$\AA$ at half peak transmission, (c) finite source (0.76$^\circ$) and collector
(0.66°) angles, (d) any difference in the effective diameter for scattering and that observed in electron micrographs, (e) spurious scatter introduced by the apparatus, which becomes significant in the angular regions in which the scattered intensities are very low.

Fahimian (31) has noticed that factor (c) results in a smoothing out of all sharp maxima and minima in the Mie diagram. Factors (a) and (b) yield a range of values of $\pi d/\lambda$ and their effect can be predicted by a suitable integration of the prediction of Mie equations. The standard deviation of the particle diameter of 0.106μ and 0.530μ reported by the manufacturer is 0.0027μ and the bandpass of the interference filters used is approximately ±30Å. These values were used to calculate the variations in $x$ shown in Table 7.1. No appreciable variation in $x$ is introduced and therefore no integration of Mie equations was considered necessary.

<table>
<thead>
<tr>
<th>d(μ)</th>
<th>λ(Å)</th>
<th>$n_w\pi d/\lambda$</th>
<th>m</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5327</td>
<td>4388</td>
<td>5.11</td>
<td>1.2</td>
</tr>
<tr>
<td>0.5300</td>
<td>4358</td>
<td>5.12</td>
<td>1.2</td>
</tr>
<tr>
<td>0.5273</td>
<td>4328</td>
<td>5.13</td>
<td>1.2</td>
</tr>
</tbody>
</table>
Figure 7.1 Effect of Particle Diameter on Mie Intensity Functions
Factor (e) is mainly responsible for the failure to present results for angles larger than 120° especially in the case of 0.530μ particles where large dissymmetries exist in the light scattered at θ and 180-θ directions.

The difference in the effective diameter for scattering and that observed in electron micrographs (factor d) is the main source of discrepancies in a single scatter experiment. Figure 7.1 shows the effect of a 5% error in the particle diameter with 0.530μ representing the diameter observed in the electronmicrograph and 0.500μ the nominal particle size reported by the manufacturer. Failure to have adjusted the particle size from 0.500μ to 0.530μ in Figures 6.3 and 6.4 would have resulted in unacceptably large deviations of the experimental values from theory.

It is interesting to note also that previous investigators have observed discrepancies in the particle sizes as supplied by Dow Chemical Company. Hepplestone and Lewis (12) have reported that a particle size has been increased by 8% in order to match the experimental and theoretical values of turbidity. Literature survey summarizes additional work on this subject.
7.2 Multiple Scatter—No Interference Present

The discussion of the experimental data of concentrated latex solutions is based on an exact mathematical equation which describes the general case of transfer of radiation through an absorbing and scattering medium.

The equation of transfer for a one-dimensional system composed of spherical dielectric particles contained between two parallel glass plates and illuminated by a polar symmetrical flux was solved as outlined in Chapter IV. The Legendre polynomial coefficients $a_i$ contained in the equation of transfer were found by Mie theory. (Appendix A) Solutions of the equation of transfer yield the angular distribution of diffusely reflected and transmitted radiation at the surfaces or any depth of a plane parallel atmosphere. In this investigation solutions were obtained by the method of discrete conical sheets (20). (Appendix B) Convergence and accuracy of the method has been established previously (43).

The validity of the final solutions are qualified by the introduction in the analysis of the following eight major assumptions:

(a) The system is a one-dimensional plane-parallel dispersion.

(b) Polarization effects are negligible.

(c) The particles are spherical and monodisperse.
(d) The particles are sufficiently far apart.

(e) The phase function for single scattering is independent of optical depth.

(f) A finite number of terms in a Legendre-polynomial expansion yields an adequate description of the phase function for single scattering.

(g) A finite number of ordinates is adequate in the solution of the equation of transfer.

(h) The particle suspension may be treated as a continuum, i.e., any volume dv large enough to contain many particles is still small compared to the system dimensions.

Of the above assumptions (a,c,e,f,g,h) were satisfied by the design of the experiment. An experimental investigation was set up to test the validity of the remaining assumptions (b,d), introduced in the solution of the equation of transfer and to determine the conditions under which the solution is valid.

7.2.1 **Polarization effects**

Polarization effects were studied by measuring separately the two components of the scattered beam. When the optical depth is small it is expected that the two components will be different. The difference will depend on the particle size, the refractive indices of the particles and continuous matrix and on the geometry of the scattering system. A large difference between the
two components is expected in case of single scatter by particles contained in a cylindrical cell. The difference here is the same observed in a Mie scattered diagram (Figures 6.3-6.4).

In case the light travels through long distances of concentrated solutions the effect of polarization can be tested by comparison of properties calculated without allowance for polarization effects either with corresponding experimental values or rigorous calculations. Both comparisons will be presented.

In the experimental study, the bidirectional intensity was calculated for radiation polarized in and perpendicular to the scatter plane. Multiple scatter bidirectional intensity data for the two components of polarization are shown for several cases in Figures 7.2-7.4. Figure 7.2 shows that small particle sizes and small optical thicknesses favor large differences between the perpendicular and parallel component. However, Figure 7.2 shows that the difference is not as large as it might be expected even though the particle diameter of the latex is \( d = .106 \mu \) and \( \tau_1 = .537 \). This is due to the interface reflection. The interface reflectance largely establishes the scattered pattern for the two components. For the system studied in this work all energy at scattered internal angles larger than \( \text{arc} \sin(1/1.33) = 48^\circ 45' \) has been totally reflected. Multireflections at the interface, inasmuch as they increase
the number of times radiation is scattered before it escapes through either boundary, process smooth out large differences between the two components. It is also interesting to notice from Figure 7.2 that the intensity of perpendicular component is higher than that of the parallel, as might be expected from the results for single scatter.

An increase in the optical thickness changes the polarization of scattered lights as observed in Figure 7.3. For a particle size \( d=0.106\mu \) and \( \tau_1=2.8 \) the intensities for the two components of polarization are essentially equal once again demonstrating that multiple scatter tends to eliminate polarization effects. A small difference between the parallel and perpendicular component of the reflected light is observed for \( d=0.530\mu \) and \( \tau_1=1.088 \) (Figure 7.3) although no appreciable polarization exists in the transmitted radiation. Polarization effects being a measure of the number of times the radiation has been scattered it is tentatively concluded that for this case the transmitted radiation has on the average been scattered more times than the reflected radiation.

Figure 7.4 is indicative of what happens in case of very large optical thicknesses. It is seen that no difference exists between the two components in the range of small angles. An increase in the angle of observation,
however, shows a higher scattering efficiency for the parallel setting of the polarizer contrary to what would be expected from single scatter results. This is explained by the fact that the interface reflectance for the parallel component ($\rho_{//}$) is lower than for the perpendicular ($\rho_{\perp}$) (Figure 7.5) in the case of large angles of observation. Consequently radiation scattered at large angles is attenuated differently for the two components. It is concluded therefore that no polarization effects are present at large optical thicknesses except the ones due to interface reflection.

The above conclusions could have been arrived at theoretically. One test example will be given. A comparison of theoretical values calculated with and without allowance for polarization effect is shown on Figure 7.6. The exact calculations were performed by Coulson et al (57) on Rayleigh scatterers ($\pi d/\lambda < 0.3$) and are shown for the case of $\tau_{//} = 1.0$. Figure 7.6 shows the differences between the bidirectional transmittance and reflectance obtained with (solid line) and without (dashed line) allowance for polarization; however the mean transmittances and reflectances as given by the areas under the curves are essentially equal for the two cases. Thus under the most extreme conditions the differences between the approximate and more rigorous analysis is seen to be restricted to the directional distribution of the energy transferred and not to the total amount.
Figure 7.2 Components of polarization of bidirectional reflectance and transmittance of polystyrene latex spheres; L=0.147 cms. Data points represent experimental values, solid lines theory.
**Figure 7.3** Components of polarization of bidirectional reflectance and transmittance of polystyrene latex spheres; $L=0.147$ cms, $\lambda=0.436$ $\mu$. Data points represent experimental values, solid lines theory.
Figure 7.4 Components of polarization of bi-directional reflectance and transmittance of polystyrene latex spheres; upper set of curves: L=.147cms, λ=0.436μ; lower set: L=0.1012 cm, λ=0.436μ; Data points represent experimental values, solid lines theory.
Figure 7.5 Reflectivity of water-glass-air interface as a function of angle $\theta$ (the angle to the normal, forward)
Figure 7.6 Bidirectional transmittance and reflectance calculated with (solid line) and without (dashed line) allowance for polarization; Rayleigh scatterers, $\mu_s = 1.0$, $\tau_1 = 1.0$.

Computed without allowance for polarization

Computed with allowance for polarization

$\theta = 0$ to $\theta = 180^\circ$
In summary it has been established that for a plane parallel multiple scattering medium with a refractive index different than one, neglecting any difference between the two components of a scattered beam, is a valid approximation.

7.2.2 Comparison of theory and experiment

The following section describes the comparison between bidirectional intensity data calculated neglecting polarization and similar data obtained by summation of the two components from experimental quantities. Figures 7.2-7.4 form the basis for this comparison. Theoretical values containing no adjustable parameters but based on the assumption that polarization effects may be ignored and that the particles scatter independently are shown as solid lines; with minor discrepancies they can be seen to constitute an excellent representation of the experimental values (total component). The discrepancies are greater at lower optical thicknesses. The source of errors is discussed in Appendix G.

The above comparison justifies the relative unimportance of polarization effects in multiple scatter problems and is based on experimental results that encompassed the entire range of optical thickness of interest and a \( \pi d/\lambda \) range of 0.78 to 5.1
From consideration of the state of polarization for single scatter it is expected that polarization effects will decrease with increasing particle size; this has already been observed in Figure 7.2-7.4.

Additional evidence of the good agreement to be expected between calculated and measured values of the energy scatter has been presented in Figures 6.5 and 6.6, which include results for two particle sizes and various optical thicknesses. It is to be emphasized that the theoretical development here, unlike the two flux method contains no adjustable parameters.

It is concluded that the solution of the equation of transfer based on the assumptions introduced in section 7.2 is adequate to describe the emerging intensities from a non-absorbing latex solution, provided that we know the refractive indices of the particle and continuous matrix and the latex particle size.

7.3 Multiple Scatter-Interference Effect

For particles not widely separated, the assumption of independent scatter is not valid. Radiation from neighboring particles interfere thereby causing changes in the scattering efficiency of the particles $X_s$, absolute intensity values and the angular distribution of emergent beam. The magnitude of changes obtained is demonstrated by comparison of experiment and theory based on independent scatter.
The degree of interference will depend on interparticle separation. For dielectric charged particles the center-to-center distance ($\delta$) between particles is calculated from the following expression for a rhombohedral array:

$$\delta = \left[\frac{\pi}{3\sqrt{2}} (\text{PVC})\right]^{1/3}d$$

(7-3)

In the present study the distance $\delta$ was varied and the angular distribution determined. The results for 0.530$\mu$m diameter spheres will be discussed first. Figure 6.6 shows data of 0.530$\mu$m particle diameter polystyrene lattices for a PVC=$10^{-1}$ — corresponding to a $\delta=1.94d$— in excellent agreement with theoretical values at two wavelengths ($\lambda=0.436$ and 0.546$\mu$m). A summary of the results of the integrated diffuse components of reflectance $R_D$ and transmittance $T_D$ have been shown for a particle diameter (0.530$\mu$m) and two wavelengths in Figures 6.8 and 6.9. There exists a remarkable agreement between theory (solid line) and experiment with the exception of slight discrepancies such as observed in Figure 6.9 where $d=0.530\mu$m and $\lambda=0.5461\mu$m. It is very important to note that data points with as close a spacing as $\delta=1.36d$ are included in Figures 6.8 and 6.9.

Figures 6.8 and 6.9 give evidence of absence of interference in the cases considered. Further support for this conclusion is offered in the Tables 7.2 and 7.3. The diffuse reflectance $R_D$ and transmittance $T_D$-normalized so
that the total quantities have a sum equal to one - are compared at the same values of \( \tau_1 \) but different \( \delta' \)s. Tables 7.2 and 7.3 suggest that \( T_D \) and \( R_D \) are independent of \( \delta \). Hence it is concluded that for a particle size \( d=0.530\mu \) and \( \lambda=0.4358 \) and \( 0.5461\mu \) the equation of transfer based on the assumption of independent scatter is an applicable model to predict \( T_D \) and \( R_D \) of lattices of particles up to a \( \delta=1.36d \)

Normalized hemispherical diffuse reflectance and transmittance for polystyrene latex spheres 0.530\( \mu \) in diameter

**TABLE 7.2**

\( \lambda=0.4358\mu \)

<table>
<thead>
<tr>
<th>( \delta/d )</th>
<th>( L(\text{cm}) )</th>
<th>( \tau_1 )</th>
<th>( T_D )</th>
<th>( R_D )</th>
<th>( DT )</th>
<th>( DR )</th>
</tr>
</thead>
<tbody>
<tr>
<td>83</td>
<td>.1473</td>
<td>.0102</td>
<td>.01017</td>
<td>.00092</td>
<td>.9083</td>
<td>.08152</td>
</tr>
<tr>
<td>58</td>
<td>.0508</td>
<td>.01</td>
<td>.00933</td>
<td>.00095</td>
<td>.9077</td>
<td>.08226</td>
</tr>
<tr>
<td>17</td>
<td>.1473</td>
<td>1.088</td>
<td>.5787</td>
<td>.0652</td>
<td>.3087</td>
<td>.04739</td>
</tr>
<tr>
<td>12</td>
<td>.056</td>
<td>1.086</td>
<td>.5742</td>
<td>.0688</td>
<td>.3090</td>
<td>.04785</td>
</tr>
<tr>
<td>2</td>
<td>.1473</td>
<td>713.9</td>
<td>.0315</td>
<td>.9254</td>
<td>0</td>
<td>.0433</td>
</tr>
<tr>
<td>1.8</td>
<td>.1067</td>
<td>713.4</td>
<td>.0309</td>
<td>.9262</td>
<td>0</td>
<td>.0433</td>
</tr>
<tr>
<td>1.7</td>
<td>.2032</td>
<td>1610</td>
<td>.0139</td>
<td>.9431</td>
<td>0</td>
<td>.0433</td>
</tr>
<tr>
<td>1.36</td>
<td>.1012</td>
<td>1571.</td>
<td>.0151</td>
<td>.9417</td>
<td>0</td>
<td>.0433</td>
</tr>
</tbody>
</table>

**TABLE 7.3**

\( \lambda=0.5461\mu \)

<table>
<thead>
<tr>
<th>( \delta/d )</th>
<th>( L(\text{cms}) )</th>
<th>( \tau_1 )</th>
<th>( T_D )</th>
<th>( R_D )</th>
<th>( DT )</th>
<th>( DR )</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>.1473</td>
<td>438.</td>
<td>.0400</td>
<td>.9162</td>
<td>0</td>
<td>.0433</td>
</tr>
<tr>
<td>1.8</td>
<td>.1048</td>
<td>429.</td>
<td>.0389</td>
<td>.9180</td>
<td>0</td>
<td>.0433</td>
</tr>
<tr>
<td>1.61</td>
<td>.2032</td>
<td>1161.</td>
<td>.0168</td>
<td>.9401</td>
<td>0</td>
<td>.0433</td>
</tr>
<tr>
<td>1.4</td>
<td>.1372</td>
<td>1194.</td>
<td>.0175</td>
<td>.9395</td>
<td>0</td>
<td>.0433</td>
</tr>
<tr>
<td>1.7</td>
<td>.2032</td>
<td>984.</td>
<td>.0198</td>
<td>.9372</td>
<td>0</td>
<td>.0433</td>
</tr>
<tr>
<td>1.36</td>
<td>.1012</td>
<td>980.</td>
<td>.0227</td>
<td>.9342</td>
<td>0</td>
<td>.0433</td>
</tr>
</tbody>
</table>
Results shown in Figure 6.7, however, imply that there is a large change in the absolute angular distribution of emerging radiation in case of a latex solution of a PVC=0.2195 composed of particles .102µ in diameter. Comparison of this finding with previous results based on a particle size 0.530µ implies that interference originates from low values of c/λ (where c is defined as the separating distance between two particles and is equal to δ-d).

It would be possible to search for an effective phase function and scattering efficiency which bring theory and experiment together. However, it has been established in section 4.6 that bidirectional intensity data are insensitive to the detailed shape of the phase diagram and only depend on the fraction of the energy scattered backward (b). Consequently a measured scattered intensity distribution may correspond to more than one phase function. Following this finding the effort in this thesis is restricted in determining the effective particle scattering coefficient X_{se}.

The scattering coefficient has been found by using the two flux model which for a system of dielectric particles contained between two cover glasses takes the following form:

\[ \frac{1}{T} = 1.09 + b \tau_1 \]  

(7-4)
Equation (7-4), however, gives no basis for selecting \( b \). Indeed there is even no guarantee that \( b \) should be independent of \( \tau \). This has been discussed in some detail in Section 4.7.1, where it was shown by using the more rigorous solution of the transfer equation that \( b \) varies with \( \tau \) at intermediate optical thicknesses but that it was approximately constant at large \( \tau \)'s. With this background it was decided to correlate the data on \( 1/T \) versus \( \tau_1 \) plots since at large optical thicknesses these would be expected to yield straight lines. The theoretical values of \( T \) calculated from the solution of the transport equation assuming that the particles scattered independently were also plotted in this manner. Departures of the data points from the theoretical line provides a measure of interference effects.

Interference can cause effective changes in the phase function, the efficiency for scatter, or in both. Since the diffuse reflectance and transmittance are insensitive to changes in the phase function (see section 4.6) it was decided to interpret departures from theory as being due to a change in the effective scattering efficiency.

Following the above discussion a number of experiments were performed based on equation 7.4. For fixed PVC the transmittance was measured at 3 to 4 different values of \( \tau_1 \). Experimental data plotted on a \( 1/T \) versus \( \tau_1 \) line were compared with theory. Data for 0.530\( \mu \) particles based on
such a comparison are shown in Figure 7.7. The ordinate is $1/T-1.09$ and the abscissa is the optical thickness $\tau_1$ based on independent scatter. The plot supports previous findings that the polystyrene latex data are fitted quite well by the theory (solid line) for both wavelengths in the whole range of PVC. A slight discrepancy is only observed for a PVC of 0.2947 and a $\lambda$ of 0.5461$\mu$, i.e. for $c/\lambda=0.35$.

Experimental results for smaller particle sizes but the same range of PVC and same $\lambda$'s, as in the previous case, are presented next.

Figure 7.5 shows results of 0.248$\mu$ particles for two $\lambda$'s and two PVC's (Tables 6.5-6.6). Measured transmittances for a PVC=0.1046 and both wavelengths are in good agreement with theory (solid line). For a PVC of 0.293, however, the inverse transmittances calculated using the transfer equation (solid line) are significantly higher than the experimental values for both wavelengths (dotted line). The dotted lines were fitted according to least square method. It is important to note that experimental values of $1/T$ for PVC=0.293 lie on a straight line.

From qualitative consideration of the effect of interference it may be expected that the decrease in effective cross-section for scatter would be a function of the total number of particles present or of the optical thickness. However, because of the linearity of $1/T$ versus $\tau_1$ plot shown in Figure 7.8 the fractional decrease appears
Figure 7.7 Comparison of theory with measurement on polystyrene spheres .530µ in diameter. Continuous lines are based on multiple scatter of non-interfering particles.
Figure 7.8 Comparison of theory with measurements on polystyrene spheres .248\(\mu\) in diameter. Continuous lines are based on multiple scatter of non-interfering particles.
Figure 7.9 Comparison of theory with experiments on polystyrene spheres 0.102μ in diameter. Continuous lines are based on multiple scatter of non-interfering particles.
to be independent of $\tau_1$. This important finding enables us to define an effective scatter coefficient $X_{se}$, so that experimental data based on an optical thickness calculated using $X_{se}$ fit the theoretical $1/T$ versus $\tau_1$ line. The effective scatter coefficient $X_{se}$ thus derived will equal the value for isolated particles for the cases in which independent scatter prevails (large $c/\lambda$ and $\delta/D$) but will show increasing departure from the isolated particle solution as $c/\lambda$ or $\delta/D$ is reduced to values where interference is important.

Further evidence of the effect of $c/\lambda$ on the $X_{se}$ value is shown in Figure 7.9 which is based on Tables 6.2 and 6.4. Data of 0.102$\mu$ polystyrene particles are compared with theory for different values of PVC and two wavelengths. It is shown that for both wavelengths and a PVC=0.0284 experimental transmittances are in absolute agreement with theory. For all other cases the experimental transmittances (dotted line) are different from the predicted theoretical values. The discrepancies increase with increasing PVC. The dotted lines in Figure 7.9 have been fitted according to the least square method. The ratio of the slope of these lines to the line obtained, either theoretically or experimentally, for widely spaced particles equals the $X_{se}/X_s$, since the use of $X_{se}$ thus derived in place of $X_s$
in evaluating $\tau_1$ will bring all the data together. The data shown in Figures 7.8-7.9 are used to derive the list of constants shown in Tables 7.4-7.5.

### TABLE 7.4

<table>
<thead>
<tr>
<th>PVC</th>
<th>$\delta/d$</th>
<th>$c$</th>
<th>$\lambda$</th>
<th>$X_{se}$</th>
<th>$X_s/X_{se}$</th>
<th>$\lambda=.4358\mu$</th>
<th>$\lambda=.5461\mu$</th>
</tr>
</thead>
<tbody>
<tr>
<td>.0284</td>
<td>2.98</td>
<td>.463</td>
<td>3287×10^{-1}</td>
<td>.995</td>
<td>.370</td>
<td>1332×10^{-1}</td>
<td>.996</td>
</tr>
<tr>
<td>.0909</td>
<td>2.02</td>
<td>.239</td>
<td>2586×10^{-1}</td>
<td>1.305</td>
<td>.190</td>
<td>0812×10^{-1}</td>
<td>1.567</td>
</tr>
<tr>
<td>.1356</td>
<td>1.76</td>
<td>.178</td>
<td>2082×10^{-1}</td>
<td>1.621</td>
<td>.142</td>
<td>0595×10^{-1}</td>
<td>2.139</td>
</tr>
<tr>
<td>.181</td>
<td>1.6</td>
<td>.140</td>
<td>1577×10^{-1}</td>
<td>2.141</td>
<td>.112</td>
<td>0421×10^{-1}</td>
<td>3.023</td>
</tr>
<tr>
<td>.2195</td>
<td>1.5</td>
<td>.117</td>
<td>1192×10^{-1}</td>
<td>2.832</td>
<td>.093</td>
<td>0319×10^{-1}</td>
<td>3.990</td>
</tr>
<tr>
<td>.2893</td>
<td>1.37</td>
<td>.086</td>
<td>0728×10^{-1}</td>
<td>4.637</td>
<td>.069</td>
<td>0191×10^{-1}</td>
<td>6.664</td>
</tr>
</tbody>
</table>

### TABLE 7.3

<table>
<thead>
<tr>
<th>PVC</th>
<th>$\delta/d$</th>
<th>$c$</th>
<th>$\lambda$</th>
<th>$X_{se}$</th>
<th>$X_s/X_{se}$</th>
<th>$\lambda=.4358\mu$</th>
<th>$\lambda=.5461\mu$</th>
</tr>
</thead>
<tbody>
<tr>
<td>.1046</td>
<td>1.92</td>
<td>.523</td>
<td>.36</td>
<td>1.085</td>
<td>.417</td>
<td>1.765</td>
<td>1.069</td>
</tr>
<tr>
<td>.293</td>
<td>1.36</td>
<td>.205</td>
<td>.287</td>
<td>1.46</td>
<td>.163</td>
<td>1.392</td>
<td>1.44</td>
</tr>
</tbody>
</table>
The results that have been presented so far demonstrate that over the range of PVC's studied the $\delta/d$ ratio was larger than the value at which spatial interference sets in. Since the measurements shown in Figure 7.7 include $\delta/d$ ratios as low as 1.36 it is concluded that the critical spacing at which interference becomes significant is less than 1.36. Churchill et al (36), however, conclude that $X_{se}$ decreases by amounts up to 10% when $c/\lambda = 0.6 - 0.7$ and $\delta/d=1.4$ (corresponding to a PVC=0.28). It is not clear that the discrepancy in the conclusions of the two studies is not within the bounds of experimental error. Further work is needed to establish the decrease in the scattering efficiency $X_{se}$ with decreases in $\delta/D$ below the value of 1.36.

Following the findings above and the results presented before it is concluded that the important parameter is $c/\lambda$. Tables 7.4 and 7.5 show a complete picture of the total scattering variation of the polystyrene lattices as a function of $c/\lambda$. The $\delta/d$ and $c/\lambda$ values were calculated assuming that the particles were uniformly distributed in a rhombohedral array (equation 7.3). The variation of $X_s/X_{se}$ (the ratio of the expected Mie scattering coefficient $X_s$ at infinite dilution to the observed experimental one $X_{se}$) as a function of $c/\lambda$ is shown in Tables 7.4 and 7.5 for two of the three particle sizes studied. The values for $X_s/X_{se}$ for the 0.530µ
Figure 7.10 Variation of scattering efficiency with c/λ.
particles are not tabulated since they were essentially equal to one (For this particle size the minimum value of c/\( \lambda \) achievable with visible light was only 0.35).

Tables 7.4 and 7.5 have been used to prepare Figure 7.10. This figure suggests that the efficiency of dense polystyrene particles remains the same as that of isolated scatterers if the clearance between the particles is larger than 1/3 the wavelength of incident radiation.

The investigation of \( X_s/X_{se} \) as a function of c/\( \lambda \) constitutes the next task of this section. The work performed by Harding et al (38) has been used as a guideline in searching for the equation to fit the data shown in Figure 7.10. Harding has estimated the effect of concentration in TiO_2 pigments. The functions he used to correlate the \( X_s/X_{se} \) ratio takes the following form for the case of spherical particles:

\[
\frac{X_{se}}{X_s} = \exp\left[-a(d/\lambda)^b(PVC)^{1/3}X_s^{1/2}\right]
\]  

(7-5)

where \( a, b \) are constants to be determined.

In this study it has been found that c/\( \lambda \) is the critical parameter and therefore the data were fitted to an appropriate modification of eq.(7-5). The resulting correlation (Figure 7.11) is given by

\[
\log\log\frac{X_s}{X_{se}} = -5.1 \frac{c}{\lambda} + 0.25
\]  

(7-6)
\[ \log \log \frac{X_S}{X_{Se}} = 0.2511 - 5.09 \frac{C}{\lambda} \]

- $d = 0.102 \mu \quad \lambda = 0.4358 \mu$
- $d = 0.102 \mu \quad \lambda = 0.5461 \mu$
- $d = 0.248 \mu \quad \lambda = 0.4358 \mu$

Figure 7.11 $X_S/X_{Se}$ correlation plot
Equation (7-6) has been derived for spherical non-absorbing particles, suspended in a medium confined between glass slides. It must be recognized that the relationship is restricted to the conditions of the present study and that its extension to systems in which the particles are absorbing, the refractive index ratio of particles to the medium differs from 1.2, or in which a significant particle size distribution exists must be tested by further experimentation. In the absence of additional data, however, Eq. (7-6) should provide a useful first estimate of interference effects.
7.4 Practical Application

The results of this investigation are valuable for the formulation of the optical properties of pigment coatings. A paint film is basically composed of pigment particles and a vehicle matrix which carries the pigment. For non-absorbing paint films it is possible to evaluate the practical application of this work. As an example we refer to TiO$_2$ paint films used extensively in paper or other white coatings.

Paper coatings are pigmented to get opacity and brightness. The coating efficiency of a pigment depends on three fundamental characteristics; namely, refractive index, particle size and pigment concentration. The refractive index is a property inherent in the structure of the crystal of each material. TiO$_2$ has the highest refractive index and consequently hiding power of any white pigment.

Hiding power of paint films is usually measured by the Kubelka-Munk constant $S$, which is defined as follows:

$$S = bK_s = 1.5 \frac{b}{d} \frac{X_{se}}{PVC} \quad (7-7)$$

where $b$ is defined by equation (7-4).

For maximum hiding power for a fixed coating thickness we need to maximize $S$. It has been found in Section 4.7.1 that for constant $d$ and $\lambda$, $b$ is constant at large $\tau_1$'s.
Our attention is therefore restricted in the variation of \( X_{se} \) with PVC. One of the important findings of this study is that, since \( X_{se} \) is a function of \( c(=\delta-d) \), there is a value of PVC increases beyond which cause the hiding power to decrease.

As an example for polystyrene lattices of 0.530\( \mu \) in diameter and two wavelengths, it has been found that \( X_{se} \) is equal to \( X_s \) (scattering efficiency for isolated particles) for all PVC's studied. It is therefore expected that \( S \) will vary linearly with PVC as demonstrated in Figure 7.12. For smaller particle sizes, where the proximity of the particles give rise to a reduction in their scattering efficiency \( X_{se} \), \( S \) no longer varies in proportion to PVC. A plot of \( S \) versus PVC departs from a straight line at the PVC at which interference becomes significant and then passes through a maximum (Figure 7.13).

If the correlation obtained for \( X_s/X_{se} \) is tentatively accepted we now have a mean for optimizing the optical properties of pigment coatings. Many other factors affect the formulation of a coating and therefore no attempt will be made to calculate an economic optimum. However, the utility of the results of this study will be illustrated by one example. Suppose it is desired to obtain the maximum hiding power of TiO\(_2\) paint films composed of spherical particles carried by a vehicle of 1.5 refractive index.
Figure 7.12 Variation of total backscattering per unit volume of polystyrene latex as a function of PVC. The solid lines represent theory, data points experimental.
Figure 7.13 Variation of total backscattering per unit volume of polystyrene latex with PVC. Solid lines calculated assuming independent scatter, dotted lines experimental.
The wavelength of illumination is assumed to be 0.55μ. The particle sizes selected were of 0.16, 0.22 and 0.28μ in diameter. Using the solution of the equation of transfer and equation (7-4) the b factor was found for all three cases. The variation of the back-scattering power per unit volume as expressed by equation (7-7) was found for different PVC's using equation (7-6) to calculate the effect of particle close spacing upon \( X_{se} \). The results of such calculations are illustrated in Figure 7.14. It is observed that a 0.22μ TiO₂ suspension gives higher hiding power up to a PVC=0.18.

The purpose of the above illustration was to demonstrate how the correlation for \( X_s/X_{se} \) provides the missing link needed for the prediction of coating optical properties. In case of commercial coatings where the systems used are poly-disperse, it is suggested that an average distance of separation be used to calculate \( c/\lambda \) and \( X_s/X_{se} \).

The effect of \( c/\lambda \) on \( X_s/X_{se} \) causes some interesting changes on \( S \) (scattering power per unit volume). Those variations are:

(i) The PVC at which the dependence of \( S \) on PVC departs from one of direct proportionality is a function of both particle size and wavelength. For fixed particle size (PVC)\(_{crit}\) increases with decreasing wavelength.

(ii) \( S \) passes through a maximum at a certain value of PVC. For fixed \( \lambda \) the value of PVC at \( S_{max} \) increases with increasing particle diameter.
Figure 7.14 Variation in backscattering coefficient per unit volume of TiO$_2$ paint film as a function of PVC. Refractive index of vehicle equals 1.5; $\lambda=0.55\mu$
The conclusions above are in agreement with the results of previous investigators working with the TiO₂ system. They usually state that the size for optimum scattering gradually becomes larger as PVC increases; smaller particle size pigments are required for development of maximum light scattering power at low PVC as shown by the crossover of 0.22 and 0.28μ curves in Figure 7.14.
IIX. CONCLUSIONS

A comparison of experimental results with calculations based on the equation of transfer showed that:

1) The radiative properties of particle suspensions may be calculated with confidence from theory provided that we know the particle diameter, the refractive index of the particles and the matrix, and the thickness of the irradiated surface.

2) Polarization effects are unimportant

3) When the clearance between particles, assumed uniformly distributed exceeds 0.33 wavelengths and when the ratio of clearance to diameter exceeds 0.4 the assumption of independent scatter is valid.

A theoretical study showed that:

1) The bidirectional intensity data are insensitive to the phase function and depend only on the fraction of radiation scattered forward.

2) In case of non-absorbing particles the total transmittance is related to the optical thickness \( \tau_1 \) in the following form:

\[
\frac{1}{T} = 1 + b \tau_1
\]

where \( b \) is the backscattered fraction. It was shown that \( b \) is constant at large \( \tau_1 \)'s.

An experimental study on dense polystyrene latices led to the following conclusions:
1) Large discrepancies exist between experiments and theoretical calculations based on the assumption of independent scatter.

2) The discrepancies are caused by a change in the particle scattering efficiency.

3) The ratio of the theoretical scattering efficiency, $X_s$, to the experimental one $X_{se}$ does not depend on $\delta/d$ in the range studied (minimum $\delta/d=1.36$).

4) $X_s/X_{se}$ is a function of only $c/\lambda$ for different particle sizes.

5) The ratio $X_s/X_{se}$ is related to $c/\lambda$ as follows:

$$\log \log \frac{X_s}{X_{se}} = -5.1 \frac{c}{\lambda} + 0.25$$

6) At low PVC’s a smaller particle size is required for the maximum scattering efficiency, but at high PVC’s large particle sizes are necessary.

7) For fixed wavelength the optimum for maximum scattering efficiency moves toward larger values.
IX. RECOMMENDATIONS

The following recommendations are offered:

1) Additional measurements be made on other dielectric particles of very concentrated lattices in order to test the validity of the correlation found in this work for other systems.

2) A study to find the effect of $\delta/d$ ratio on the particle scattering efficiency. A system of large particle sizes not subjected to fast agglomeration is recommended.

3) Polydisperse systems of known particle size distribution be studied. For that purpose monodisperse particle suspension of different diameters could be mixed and the effect of concentration on $X_s/X_{se}$ could be studied.

4) Paint films prepared from commercially available pigments be studied. This should offer a means to determine if correlations found for idealized systems are of any value in practical applications.
APPENDIX A

Description of the Phase Function by a Legendre Polynomial Series

It is desired to express the phase function by a Legendre polynomial series:

\[ p(\cos \theta) = \sum_{n=0}^{\infty} a_n P_n(\cos \theta) \]  

(A-1)

Using the properties of the orthogonal Legendre polynomials (47) we obtain:

\[ a_n = \frac{(2n+1)}{2} \int_{-1}^{1} p(\cos \theta) P_n(\cos \theta) \, d\cos \theta \]  

(A-2)

If the phase function is replaced by \(4i(\theta)/x_s x^2\) according to 3.15 and integration is over \(\theta\) instead of \(\cos \theta\) we have:

\[ a_n = \frac{2(2n+1)}{x_s x^2} \int_{0}^{180} i(\theta) P_n(\cos \theta) \sin \theta \, d\theta \]  

(A-3)

If the intensity parameter \(i(\theta)\) is available at equal increments of \(\Delta \theta\) between \(0^\circ\) and \(180^\circ\), this integral can be approximated by Simpson's rule. This procedure was followed in the present study.

A flow chart of the computer programming is shown in Figure A.2. In Table A.1 results of the present program are compared with those obtained by Churchill et al (48).
A more critical test is the comparison of phase function values with series approximation shown in Table A.2. Table A.2 demonstrates that the approximate representation of phase function by a Legendre polynomial series does not introduce appreciable errors in the phase function values at any angle.
TABLE A.1

\[ \frac{\pi^D}{\lambda} = 10. \quad m = 1.2 \]

<table>
<thead>
<tr>
<th>( a_n ) (Churchill)</th>
<th>( a_n ) (Present Work)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.0</td>
</tr>
<tr>
<td>2</td>
<td>2.78197</td>
</tr>
<tr>
<td>3</td>
<td>4.25856</td>
</tr>
<tr>
<td>4</td>
<td>5.38683</td>
</tr>
<tr>
<td>5</td>
<td>6.19015</td>
</tr>
<tr>
<td>6</td>
<td>6.74492</td>
</tr>
<tr>
<td>7</td>
<td>7.06711</td>
</tr>
<tr>
<td>8</td>
<td>7.20999</td>
</tr>
<tr>
<td>9</td>
<td>7.20063</td>
</tr>
<tr>
<td>10</td>
<td>7.03629</td>
</tr>
<tr>
<td>11</td>
<td>6.76587</td>
</tr>
<tr>
<td>12</td>
<td>6.35881</td>
</tr>
<tr>
<td>13</td>
<td>5.83351</td>
</tr>
<tr>
<td>14</td>
<td>5.22997</td>
</tr>
<tr>
<td>15</td>
<td>4.47918</td>
</tr>
<tr>
<td>16</td>
<td>3.69000</td>
</tr>
<tr>
<td>17</td>
<td>2.81577</td>
</tr>
<tr>
<td>18</td>
<td>1.92305</td>
</tr>
<tr>
<td>19</td>
<td>1.11502</td>
</tr>
<tr>
<td>20</td>
<td>.50766</td>
</tr>
<tr>
<td>21</td>
<td>.20927</td>
</tr>
<tr>
<td>22</td>
<td>.07138</td>
</tr>
<tr>
<td>23</td>
<td>.02090</td>
</tr>
<tr>
<td>24</td>
<td>.00535</td>
</tr>
<tr>
<td>25</td>
<td>.00120</td>
</tr>
<tr>
<td>26</td>
<td>.00024</td>
</tr>
</tbody>
</table>
### TABLE A.2

**Comparison of Exact Phase Function Values With Series Approximation**

\[ \frac{\pi d}{\lambda} = 5.1212 \]

\[ m = 1.201 \]

<table>
<thead>
<tr>
<th>( \theta ) (degrees)</th>
<th>( \frac{i(\theta)}{X_s x^2} )</th>
<th>( \sum_{i=0}^{\infty} a_n P_n (\cos \theta) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>27.23</td>
<td>27.23</td>
</tr>
<tr>
<td>10</td>
<td>22.40</td>
<td>22.40</td>
</tr>
<tr>
<td>20</td>
<td>12.14</td>
<td>12.13</td>
</tr>
<tr>
<td>30</td>
<td>3.872</td>
<td>3.873</td>
</tr>
<tr>
<td>40</td>
<td>.5598</td>
<td>.5599</td>
</tr>
<tr>
<td>50</td>
<td>.1731</td>
<td>.1736</td>
</tr>
<tr>
<td>60</td>
<td>.2606</td>
<td>.2601</td>
</tr>
<tr>
<td>70</td>
<td>.1444</td>
<td>.1439</td>
</tr>
<tr>
<td>80</td>
<td>.03704</td>
<td>.03651</td>
</tr>
<tr>
<td>90</td>
<td>.03286</td>
<td>.03262</td>
</tr>
<tr>
<td>100</td>
<td>.04859</td>
<td>.04842</td>
</tr>
<tr>
<td>110</td>
<td>.03623</td>
<td>.03625</td>
</tr>
<tr>
<td>120</td>
<td>.01753</td>
<td>.01783</td>
</tr>
<tr>
<td>130</td>
<td>.01352</td>
<td>.01374</td>
</tr>
<tr>
<td>140</td>
<td>.01928</td>
<td>.01929</td>
</tr>
<tr>
<td>150</td>
<td>.02700</td>
<td>.02667</td>
</tr>
<tr>
<td>160</td>
<td>.03923</td>
<td>.03896</td>
</tr>
<tr>
<td>170</td>
<td>.05525</td>
<td>.05532</td>
</tr>
<tr>
<td>180</td>
<td>.06324</td>
<td>.06331</td>
</tr>
</tbody>
</table>
Programm Flow Chart for Legendre Polynomials Coefficients

1. Read n/a
2. Read U, V
3. NTEST
4. Calculate \( s_1(\theta) \)
5. \( s_1(\theta), s(\theta), x_3 \) from 0° to 180°
6. Print \( s(\theta), s(\theta), x_3 \)
7. YES
8. NTEST
9. NO
10. Calculate \( \sin \theta \)
11. from 0° to 180°
12. Start calculating \( a_n \) for \( n = 1, 100 \)
13. Calculate \( \cos \theta \), \( p(n(\cos \theta)) \) for all angles
14. Calculate \( a_n \) according to A.4
15. If \( a_n \) less than 0.0000
16. CALL EXIT
17. Write \( a_n \)
Key to the Fortran Symbols

<table>
<thead>
<tr>
<th>Variable</th>
<th>Fortran Symbol</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\pi d/\lambda)</td>
<td>ALPHA</td>
</tr>
<tr>
<td>m</td>
<td>U</td>
</tr>
<tr>
<td>k</td>
<td>v</td>
</tr>
<tr>
<td>(\theta)</td>
<td>ANGLE</td>
</tr>
<tr>
<td>(i_L(\theta))</td>
<td>PRI</td>
</tr>
<tr>
<td>(i_{II}(\theta))</td>
<td>PLI</td>
</tr>
<tr>
<td>(X_s)</td>
<td>QSCA</td>
</tr>
<tr>
<td>(X_a)</td>
<td>QABS</td>
</tr>
<tr>
<td>(X_t)</td>
<td>QEXT</td>
</tr>
<tr>
<td>(a_n)</td>
<td>A</td>
</tr>
</tbody>
</table>

Number to test for convergence of \(i_L(\theta), i_{II}(\theta)\) \hspace{1cm} CONV

**IMPUT**

<table>
<thead>
<tr>
<th>ALPHA, U, V</th>
<th>3F10.4</th>
</tr>
</thead>
<tbody>
<tr>
<td>CONV</td>
<td>F10.9</td>
</tr>
<tr>
<td>NTEST</td>
<td>I3</td>
</tr>
</tbody>
</table>
APPENDIX B

Development and Solution of the Equation of Transfer

For azimuth independent boundary conditions

\[ I(\tau, \mu, \psi) \text{ in equation 4.5 may be replaced by } I(\tau, \mu) \text{ to yield} \]

\[ \mu \frac{dI(\tau, \mu)}{d\tau} = -I(\tau, \mu) \]

\[ + \frac{\omega}{2\pi} \int_{-1}^{1} I(\tau, \mu') \int_{0}^{\pi} P(\Theta) d\psi' d\mu' \]

\[ + \frac{\omega}{4\pi\mu_o} e^{-\tau/\mu_o} \int_{0}^{\pi} P(\theta_o) \frac{d\psi}{\pi} \]

The integrals over \( \psi \) are evaluated by the following procedure: \( P(\Theta) \) is expanded in a Legendre polynomial series (eq. 3.16); \( \cos \Theta \) replaced by the function of \( \mu, \mu' \), and \( \psi-\psi' \) given in equation 4.2; and the addition theory for Legendre polynomials is utilized to yield the following relation for the \( i \)th term.

\[ P_i(\cos \Theta) = P_i(\mu)P_i(\mu') + \sum_{k=4}^{i} f(\mu, \mu', k, i) \cos k(\psi-\psi') \]

where \( f \) is a function the knowledge of whose structure is not needed here. The integrals over azimuth angles then yield:

\[ \int_{0}^{\pi} P(\Theta) d\psi' = \pi \sum_{i=1}^{L} a_i P_i(\mu) P_i(\mu') \]

since \( \int_{0}^{\pi} \cos k\psi' d\psi' \) is zero for integral values of \( k \).
With equation 4.6 thus derived the integral is replaced by a weighted sum of intensities evaluated at $N$ discrete ordinates. $N$ linear differential equations result. For the intensity in the direction $\mu_i$

$$\mu_i \frac{dI(\tau, \mu_i)}{d\tau} = -I(\tau, \mu_i)$$

$$+ \frac{\omega}{N} \sum_{j=1}^{N} W_j I(\tau, \mu_j) \sum_{k=1}^{L} a_k P_k(\mu_i) P_k(\mu_j)$$

$$+ \frac{\omega}{4\pi \mu_o} e^{-\tau/\mu_o} \sum_{k=1}^{L} a_k P_k(\mu_o) P_k(\mu_i)$$

(B-1)

where $W_j = 1$ for equal interval integration formula

$$W_j = \frac{H_j N}{2}$$ for non-equal integral integration formula.

$H_j$ as well as $\mu_j$ represent the weights and ordinates for a particular quadrature.

For future simplification the following definitions are introduced:

$$g_{ij} = \{ \sum_{k=1}^{L} a_k P_k(\mu_i) P_k(\mu_j) \} W_j$$

(B-2)

$$g_{i,N+1} = \sum_{k=1}^{L} a_k P_k(\mu_i) P_k(\mu_o)$$

$$g_{i,N+2} = \sum_{k=1}^{L} a_k P_k(\mu_i) P_k(-\mu_o)$$
Introducing the so defined quantities into the equation B-1 we get

\[ \mu_i \frac{dI_i}{d\tau} = -I_i + \frac{\omega}{N} \sum_{j=1}^{N} g_{ij} I_j + \frac{\omega}{4\pi} e^{-\tau/\mu_0} g_{i,N+1} \]  

(B-3)

Solution of the Homogeneous Set

Let \( \omega/N = \omega_i \) and let us rewrite the homogeneous set of equations related to B-3 as follows:

\[ \frac{dI_i}{d\tau} = \sum_{j=1}^{N} \left( \frac{\omega_i g_{ij} - \delta_{ij}}{\mu_i} \right) I_j \]  

(B-4)

Assume a solution vector of the form:

\[ I_i = A_i e^{\gamma \tau} \]

where \( A_i \) is independent of \( \tau \)

Introducing the assumed form of \( I_i \) into B-4 we obtain:

\[ \sum_{j=1}^{N} \left( \frac{\omega_i g_{ij} - \delta_{ij}}{\mu_i} - \delta_{ij} \gamma \right) A_j = 0 \]  

(B-5)

For a non-zero solution for \( A_j \) the determinant of the coefficient matrix of B-5 must equal zero, i.e.,

\[ \begin{vmatrix} \frac{\omega_i g_{ij} - \delta_{ij}}{\mu_i} - \delta_{ij} \gamma \end{vmatrix} = 0 \]  

(B-6)
Equation B-6 has in general N roots $\gamma_j$. Special cases include:

(i) Distinct roots

The complementary function for system B-4 is:

$$I_i = \sum_{j=1}^{N} c_j A_{ji} e^{\gamma_j \tau} \quad (B-7)$$

where $A_{ji}$ are the eigenvector of the matrix B-6.

(ii) Two roots are equal. Assume $\gamma_{N-1} = \gamma_N$.

For all eigenvalues $\gamma_j$ where $j = 1, N-1$ the corresponding solution vector is $I_i = A_{i} e^{\gamma \tau}$

For the eigenvalue $\gamma_N$ a solution vector of the following form is assumed:

$$I_i = (B_i + \tau D_i)e^{\gamma \tau} \quad (B-8)$$

If we introduce B-9 into B-4 we get:

$$\sum_{j=1}^{N} \left( \frac{\omega_{ij} - \delta_{ij} - \gamma \delta_{ij}}{\mu_i} \right) B_j + \tau \sum_{j=1}^{N} \left( \frac{\omega_{ij} - \delta_{ij} - \gamma \delta_{ij}}{\mu_i} \right) D_j = D_i \quad (B-9)$$

Equation B-9 implies:

$$\sum_{j=1}^{N} \left( \frac{\omega_{ij} - \delta_{ij} - \gamma \delta_{ij}}{\mu_i} \right) B_j = D_i \quad (B-10)$$

and

$$\sum_{j=1}^{N} \left( \frac{\omega_{ij} - \delta_{ij} - \gamma \delta_{ij}}{\mu_i} \right) D_j = 0 \quad (B-11)$$
Equation B-11 implies that $D_j$ is the eigenvector corresponding to $\gamma = \gamma_{N-1} = \gamma_N$. Hence $D_j = A_j$ for $j = A_{N-1}$. The components of the vector $[B_j]$ are found by letting $B_N = 1$ and solving the system of equations:

$$\sum_{j=1}^{N-1} \left( \frac{\omega g_{ij} - \delta_{ij}}{\mu_i} - \gamma \delta_{ij} \right) B_j = \binom{A_{N-1}}{\mu_i} - \frac{\omega g_{iN}}{\mu_i} \quad (B-12)$$

The general solution in this case is

$$I_i = \sum_{j=1}^{N-1} c_j A_{j(i)} e^{\gamma_j T} + c_N (B_{N(i) + \tau A_{N-1},i}) e^{\gamma N T} \quad (B-13)$$

**Particular Solution**

(a) Non-reflecting boundary conditions.

A solution vector of the following form is assumed:

$$I_i = r_i e^{-\tau/\mu_0} \quad (B-14)$$

The constants $I_i$ are found by solving the system of equations resulting by introducing B-14 into B-3:

$$\sum_{j=1}^{N} \left[ -\omega g_{ij} + (1 - \frac{\mu_i}{\mu_0}) \delta_{ij} \right] R_j = \frac{\omega}{4\pi \mu_0} g_i, N+1 \quad (B-15)$$

(b) Reflecting boundary conditions:

The constant term in B-3 is replaced by the term 4.13. The term 4.13 implies that there are two particular solutions. One has the form $I_i = r_i e^{-\tau/\mu_0}$ where $[r_i]$ is found by solving
the system of equations B-15 after we replace the second member with
\[ \frac{1-\rho}{1-\rho^2e^{-2\tau_1/\mu_0}} \cdot \frac{\omega}{4\pi \mu_0} g_{i,N+1} \]

The second particular solution vector \( I_i = s_i e^{\tau/\mu_0} \) where \( [s_i] \) is found from the system of equations:

\[
\sum_{j=1}^{N} \left[ -\omega_1 g_{ij} + (1 + \frac{\mu_i}{\mu_0}) \delta_{ij} \right] s_j = \frac{\rho(1-\rho)e^{-2\tau_1/\mu_0}}{1-\rho^2e^{-2\tau_1/\mu_0}} \cdot \frac{\omega}{4\pi \mu_0} g_{i,N+2}
\]

(B-16)

**General Solution**

In case the eigenvalues are distinct and the boundary conditions are non-reflecting the general solution is:

\[
I_i = \sum_{j=1}^{N} c_j A_{ji} e^{\gamma_j \tau} + r_i e^{-\tau/\mu_0}
\]

(B-17)

The boundary conditions are:

\[
I_i(i=1,\ldots,N) = 0 \quad \text{at} \quad \tau = 0
\]

(B-18)

\[
I_i(i=N/2+1,\ldots,N) = 0 \quad \text{at} \quad \tau = \tau_1
\]

The constants \( [c_j] \) are found by solving the system of linear equations:

\[
\sum_{j=1}^{N} c_j A_{ji} = -R_i \quad i=1,\ldots,N/2
\]

(B-19)

\[
\sum_{j=1}^{N} c_j A_{ji} e^{\gamma_j \tau_1} = -R_i e^{-\tau_1/\mu_0} \quad i=N/2+1,\ldots,N
\]
In case two eigenvalues are equal and the boundary conditions are reflecting the general solution takes the following form:

\[ I_i = \sum_{j=1}^{N-1} c_j A ji e^{\gamma j \tau} + c_N (B_{N i}^r + A_{N-1, i}) e^{\gamma_N r_i + r_i e^{-\tau/\mu_0} + \mu_1 e^{\tau/\mu_0}} \]  

(B-20)

The boundary conditions are:

\[ I_i (i=1, \ldots, N/2) = \rho_i I_{N+1-i} \text{ at } \tau = 0 \]  

(B-21)

\[ I_i (i=N/2+1, \ldots, N) = \rho_{N+1-i} I_{N+1-i} \text{ at } \tau = \tau_1 \]

The application of the boundary conditions B-21 lead to the following system of equation:

\[ \sum_{j=1}^{N-1} (A_{ji} - \rho_i A_{ji, N+1-i}) c_j + (B_{Ni} - \rho_i B_{N, N+1-i}) c_N \]

\[ = (-R_{i} + \rho_i R_{N+1-i}) + (-S_{i} + \rho_i S_{N+1-i}) (i=i, \ldots, \frac{N}{2}) \]

(B-22)
The calculated intensities are used to find the diffuse reflectance \( R_D \) and transmittance using the relevant equations:

\[
R_D = -2\pi \int_{-1}^{0} I(\alpha, \mu) \mu d\mu = -2\pi \sum_{j=1}^{N} W_j I_{j, \mu_j} \quad (B-23)
\]

\[
T_D = 2\pi \int_{0}^{1} I(\tau, \mu) \mu d\mu = 2\pi \sum_{j=1}^{N} W_j I_{j, \mu_j} \quad (B-24)
\]
FIGURE B.1
PROGRAMM FLOW CHART FOR TRANSFER EQUATION
### Key to the Fortran Program

<table>
<thead>
<tr>
<th>Variable</th>
<th>Fortran Symbol</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \omega )</td>
<td>( W )</td>
</tr>
<tr>
<td>( \theta_0 )</td>
<td>( \text{PHIO} )</td>
</tr>
<tr>
<td>( \tau_1 )</td>
<td>( \text{TAUL} )</td>
</tr>
<tr>
<td>( a_i )</td>
<td>( c(I) )</td>
</tr>
<tr>
<td>( I_0 )</td>
<td>( E0 )</td>
</tr>
<tr>
<td>( I(\tau,\mu,\psi) )</td>
<td>( \text{SINT(LL,I)} )</td>
</tr>
<tr>
<td>( n )</td>
<td>( RN )</td>
</tr>
<tr>
<td>( p_i(\mu) )</td>
<td>( P(I,J) )</td>
</tr>
<tr>
<td>( g_{ij} )</td>
<td>( G(I,J) )</td>
</tr>
<tr>
<td>( \rho )</td>
<td>( RO )</td>
</tr>
<tr>
<td>( \gamma )</td>
<td>( \text{EIGEN} )</td>
</tr>
<tr>
<td>( n_{\text{W}} )</td>
<td>( \text{RNW} )</td>
</tr>
<tr>
<td>( n_{\text{G}} )</td>
<td>( \text{RNG} )</td>
</tr>
</tbody>
</table>
### Input to Program

<table>
<thead>
<tr>
<th>Order</th>
<th>Item</th>
<th>Format</th>
<th>Location and Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>W, PHI0, TAUL</td>
<td>9E8.4</td>
<td>Main</td>
</tr>
<tr>
<td>2</td>
<td>N, NTEST</td>
<td>5I3</td>
<td>Main</td>
</tr>
<tr>
<td></td>
<td>NTEST=1</td>
<td>No Print</td>
<td>Intermediate Results</td>
</tr>
<tr>
<td></td>
<td>NTEST=2</td>
<td>Print</td>
<td>Intermediate Results</td>
</tr>
<tr>
<td>3</td>
<td>EO</td>
<td>E8.4</td>
<td>Main</td>
</tr>
<tr>
<td>4</td>
<td>IRFK, IREFL, IGLASS</td>
<td>3I3</td>
<td>Main</td>
</tr>
<tr>
<td></td>
<td>(a) IRFK=1, VOID, VOID</td>
<td></td>
<td>No reflection at the boundaries</td>
</tr>
<tr>
<td></td>
<td>(b) (i) IRFK=2, VOID, VOID</td>
<td></td>
<td>Reflection at boundaries. One step change in refractive index.</td>
</tr>
<tr>
<td></td>
<td>(ii) IRFK=2, IREFL=1, IGLASS=1</td>
<td></td>
<td>Reflection at boundaries. Two step change in refractive index.</td>
</tr>
<tr>
<td></td>
<td>(iii) IRFK=2, IREFL=2, VOID</td>
<td></td>
<td>Continuous medium is water. Two step change in refractive index.</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>Slab is surrounded by water cylindrical jacket.</td>
</tr>
<tr>
<td>5</td>
<td>MM,N</td>
<td>3I3</td>
<td>PHIANG</td>
</tr>
<tr>
<td></td>
<td>MM=1</td>
<td></td>
<td>Equal Δµ</td>
</tr>
<tr>
<td></td>
<td>MM=2</td>
<td></td>
<td>Gaussian</td>
</tr>
<tr>
<td>6</td>
<td>RN</td>
<td>E8.4</td>
<td>RFK</td>
</tr>
<tr>
<td></td>
<td>VOID</td>
<td></td>
<td>IRFK=1, VOID, VOID</td>
</tr>
<tr>
<td></td>
<td>If</td>
<td></td>
<td>IRFK=2, IREFL=2, VOID</td>
</tr>
<tr>
<td>7</td>
<td>M,N,L</td>
<td>3I3</td>
<td>GNM</td>
</tr>
<tr>
<td></td>
<td>If M=1 Read</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>((G(I,J), J=1, N+2), I=1,N)</td>
<td>9E8.4</td>
<td></td>
</tr>
<tr>
<td></td>
<td>M=2</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>((C(K), K=1,L)</td>
<td>9E8.4</td>
<td></td>
</tr>
</tbody>
</table>
## Input to Program (Continued)

<table>
<thead>
<tr>
<th>Order</th>
<th>Item</th>
<th>Format</th>
<th>Location and Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>M, IPRINT</td>
<td>2I3</td>
<td>UNSEIG</td>
</tr>
<tr>
<td></td>
<td>M=N In this case</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>IPRINT=0</td>
<td>No Print</td>
<td>Intermediate results</td>
</tr>
<tr>
<td></td>
<td>=L</td>
<td>Print</td>
<td>Intermediate results</td>
</tr>
<tr>
<td>9</td>
<td>LL</td>
<td>I3</td>
<td>CAOFIN</td>
</tr>
<tr>
<td></td>
<td>LL=Number of optical depths we want to calculate and print intensities</td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>(TAU(I), I=1, LL)</td>
<td>9E8.4</td>
<td>CAOFIN</td>
</tr>
</tbody>
</table>
C

SUBROUTINE FOR MIGRATION FUNCTION FOR DIFFERENT SETS

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE

200 CONTINUE

300 CONTINUE

400 CONTINUE

SUBROUTINE MIGRATION

DIMENSION MIGRATION(9)

100 CONTINUE
158
SUBROUTINE FOR PRINTING FINAL RESULTS AT THE INTERFACE OF THE
C LAYERS

SUBROUTINE PRINT

COMMON / LAYERS / ANGLE, VS, SQUARE, WAVE, INTENSITY, TH, LAYERS

COMMON / INTERFACE / INTERFACE

COMMON / OUTFILE / OUTFILE

COMMON / PAPER / PAPER

COMMON / PLOT / PLOT

COMMON / SCALE / SCALE

COMMON / TITLE / TITLE

COMMON / XMAX / XMAX

COMMON / YMAX / YMAX

COMMON / ZMAX / ZMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMAX

COMMON / ZYMAX / ZYMAX

COMMON / ZMIN / ZMIN

COMMON / ZXMAX / ZXMA
APPENDIX C

Simplified Representation of Mie Scattering

Diagram (Phase Function)

Because of the difficulty in solving the transport equation for large \( \pi d/\lambda \)'s, it is desired to replace an exact phase function of many terms with a hypothetical one of very few terms. There are many ways that this may be achieved. Work done by Churchill and Evans (16) has shown that the transmission and reflectance are dependent primarily on the fraction of the radiation scattered into the backward (or forward) hemisphere. Hence, any substitution of a complicated phase function with a simple one must preserve the forward (or backward) fraction.

The forward fraction is given by (Ref. 26):

\[
\frac{1}{2} \int_{0}^{1} p(\mu) d\mu = \frac{1}{2} \int_{0}^{1} \left\{ \sum_{i=0}^{L} a_i p_i(\mu) \right\} d\mu \quad (C-1)
\]

or

\[
f = \frac{1}{2} \sum_{i=0}^{L} a_i \int_{0}^{1} p_i(\mu) d\mu \quad (C-2)
\]

From Hilderbrand we get:

\[
\int_{0}^{1} p_i(\mu) d\mu = (-1)^{i-1}/2 \frac{(1.3.5\ldots-i)^2}{i!} \quad (C-3)
\]
Introducing equation C-3 into C-2 yields:

\[
\frac{1}{2} + \frac{1}{2} \sum_{i=1,3,5,\ldots}^{L} (-1)^{ \frac{i-1}{2} } \frac{1}{i(i+1)} \frac{(1-3-5-\ldots-i)}{i!} a_i \quad (C-4)
\]

In a similar way the peakedness defined by:

\[
\frac{1/2}{f} \int_0^1 P(\mu) \mu^2 d\mu = \frac{1}{f} \int_0^1 P_i(\mu) \mu^2 d\mu = \frac{1}{2f} \sum_{i=0}^{L} a_i \int_0^1 P_i(\mu) \mu^2 d\mu \quad (C-5)
\]

is equal to

\[
P_{\text{e}} = \frac{1}{2f} \int_0^1 \sum_{i=0}^{L} a_i P_i(\mu) \mu^2 d\mu = \frac{1}{2f} \sum_{i=0}^{L} a_i \int_0^1 P_i(\mu) \mu^2 d\mu \quad (C-6)
\]

From the Handbook of Mathematical Functions (50) we have:

\[
\int_0^1 P_i(\mu) \mu^2 d\mu = \frac{\Gamma(1/2)2^{-2}}{\Gamma(2-1/2)(5/2+i/2)} \quad (C-7)
\]

Hence:

\[
P_{\text{e}} = \frac{1}{8f} \sum_{i=0}^{L} \frac{\Gamma(1/2)}{\Gamma(2-1/2)\Gamma(5/2+i/2)} a_i \quad (C-8)
\]

The height of the forward peak (intensity at \(\theta=0\)) is given by:

\[FP = 1 + a_1 + a_2 + a_3 + a_4 + \ldots \quad (C-9)\]

In the following consider the problem of finding the coefficients \(c_i\) of a simple phase function in some simplified cases:
(a) It is desired to obtain a simple phase function of
the form \( P(\mu) = l + c_1 P_1(\mu) + c_2 P_2(\mu) \) that matches the forward
fraction \( f \) and Peakedness \( Pe \) of a Mie scatterer:

\[
f = \frac{1}{2} + \frac{c_1}{4}
\]

\[
Pe = \frac{1}{6} + \frac{c_1}{8} + \frac{c_2}{15}
\]

From equations C-10 we get:

\[
c_0 = 1
\]

\[
c_1 = 4f - 2
\]

\[
c_2 = 15f \cdot Pe - \frac{15}{8} \cdot c_1 - \frac{15}{6}
\]

(b) For the case in which the forward fraction and
forward peak are to be matched, the coefficients \( c_1, c_2 \)
are given by:

\[
c_0 = 1
\]

\[
c_1 = 4f - 2
\]

\[
c_2 = FP - 1 - c_1
\]

(c) For a four term phase function which has the same
\( f, Pe \) and \( FP \) as the original one the coefficients are:

\[
c_0 = 1
\]

\[
c_1 = \frac{(16f + 48Pe - 16)}{10} - \frac{48}{150} \cdot c_2
\]

\[
c_2 = \frac{15}{18} \left\{ (16f - 8) + 2(FP - 1) - (48Pe - 8) \right\}
\]

\[
c_3 = (FP - 1) - c_1 - c_2
\]
Key to the Program for Finding the Coefficients of a Simplified Phase Function

<table>
<thead>
<tr>
<th>Variable</th>
<th>Symbol</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\pi d/\lambda$</td>
<td>ALPHA</td>
</tr>
<tr>
<td>m</td>
<td>U</td>
</tr>
<tr>
<td>f</td>
<td>F</td>
</tr>
<tr>
<td>$a_n$</td>
<td>C</td>
</tr>
<tr>
<td>FP</td>
<td>FP</td>
</tr>
<tr>
<td>Pe</td>
<td>PE</td>
</tr>
<tr>
<td>$\theta$</td>
<td>ANGLE</td>
</tr>
</tbody>
</table>

Input to the Program

ALPHA, U
L
$C(I), I=1, L$

2F10.5
I3
9E8.4
C PROGRAM FOR FINDING THE COEFFICIENTS OF A SIMPLIFIED
C PHASE FUNCTION
C MAIN
C
C SUBROUTINE PHASE (E, PHA)
C
C SIMPLE FORM OF PHASE FUNCTION OF THE TYPE
C PHASE = E \* (1 + E \* EXP(-D/E))
C
C WHERE E IS THE ENERGY, PHA IS THE PHASE FUNCTION
C
C SUBROUTINE AMPLITUDE (E, AMP)
C
C SIMPLE FORM OF AMPLITUDE FUNCTION OF THE TYPE
C AMPLITUDE = E \* (1 + E \* EXP(-D/E))
C
C WHERE E IS THE ENERGY, AMP IS THE AMPLITUDE FUNCTION
C
C DATA N=5, D=5.0
C
C COMMON AMPLITUDE, PHASE, N, D
C
C SUBROUTINE FILL (E, PHA, AMP)
C
C FILL THE DATA TABLES WITH PHASE AND AMPLITUDE VALUES
C
C FOR E = 0.0 TO 5.0 IN STEPS OF 0.5
C
C SUBROUTINE INTEGRATE (E, PHA, AMP)
C
C INTEGRATE THE PHASE AND AMPLITUDE DATA TABLES
C
C FOR E = 0.0 TO 5.0 IN STEPS OF 0.5
C
C SUBROUTINE DETERMINE (E, PHA, AMP)
C
C DETERMINE THE COEFFICIENTS OF THE PHASE FUNCTION
C
C USING THE INTEGRATED DATA
C
C FOR E = 0.0 TO 5.0 IN STEPS OF 0.5
C
C SUBROUTINE PRINT (E, PHA, AMP)
C
C PRINT THE DATA TABLES AND THE DETERMINED COEFFICIENTS
C
C FOR E = 0.0 TO 5.0 IN STEPS OF 0.5
C
C END

100 FORMAT (1X) 168
APPENDIX D

The b factor for different values of size parameter, refractive index and boundary conditions in case of conservative scattering ($\omega=1.0$).

**TABLE D.1**

$x=.1$  $m=1.5$  $n=1.0$

\[ b_o = \frac{1}{T} - 1 \]

<table>
<thead>
<tr>
<th>$\tau_1$</th>
<th>$R_D$</th>
<th>$T_D$</th>
<th>$DT$</th>
<th>$DR$</th>
<th>$T_\frac{T_D+DT}{T}$</th>
<th>$1/T$</th>
<th>$b_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>.02</td>
<td>.00987</td>
<td>.00992</td>
<td>.98019</td>
<td>0</td>
<td>.99011</td>
<td>1.00998</td>
<td>.4994</td>
</tr>
<tr>
<td>.05</td>
<td>.02432</td>
<td>.02442</td>
<td>.95123</td>
<td>0</td>
<td>.97565</td>
<td>1.02496</td>
<td>.4992</td>
</tr>
<tr>
<td>.08</td>
<td>.03837</td>
<td>.03847</td>
<td>.92312</td>
<td>0</td>
<td>.96159</td>
<td>1.03994</td>
<td>.4992</td>
</tr>
<tr>
<td>.10</td>
<td>.04752</td>
<td>.04758</td>
<td>.90484</td>
<td>0</td>
<td>.95242</td>
<td>1.04995</td>
<td>.4995</td>
</tr>
<tr>
<td>50</td>
<td>.2017</td>
<td>.1916</td>
<td>.60653</td>
<td>0</td>
<td>.79813</td>
<td>1.253</td>
<td>.5060</td>
</tr>
<tr>
<td>1.</td>
<td>.3398</td>
<td>.2920</td>
<td>.36788</td>
<td>0</td>
<td>.65988</td>
<td>1.515</td>
<td>.5150</td>
</tr>
<tr>
<td>2.</td>
<td>.5154</td>
<td>.3488</td>
<td>.13533</td>
<td>0</td>
<td>.48413</td>
<td>2.065</td>
<td>.5325</td>
</tr>
<tr>
<td>3.</td>
<td>.6204</td>
<td>.3293</td>
<td>.04978</td>
<td>0</td>
<td>.37908</td>
<td>2.638</td>
<td>.5460</td>
</tr>
<tr>
<td>5.</td>
<td>.737</td>
<td>.2557</td>
<td>.00674</td>
<td>0</td>
<td>.26244</td>
<td>3.810</td>
<td>.5620</td>
</tr>
<tr>
<td>10</td>
<td>.8518</td>
<td>.1477</td>
<td>.00004</td>
<td>0</td>
<td>.1477</td>
<td>6.770</td>
<td>.5770</td>
</tr>
<tr>
<td>100</td>
<td>.9829</td>
<td>.01664</td>
<td>0</td>
<td>0</td>
<td>.01664</td>
<td>60.09</td>
<td>.6009</td>
</tr>
<tr>
<td>1000</td>
<td>.9978</td>
<td>.00168</td>
<td>0</td>
<td>0</td>
<td>.00168</td>
<td>593.5</td>
<td>.5935</td>
</tr>
</tbody>
</table>

**TABLE D.2**

$x=.1$  $m=2.7$  $n=1.0$

\[ b_o = \frac{1}{T} - 1 \]

<table>
<thead>
<tr>
<th>$\tau_1$</th>
<th>$R_D$</th>
<th>$T_D$</th>
<th>$DT$</th>
<th>$DR$</th>
<th>$T_\frac{T_D+DT}{T}$</th>
<th>$1/T$</th>
<th>$b_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>.05</td>
<td>.02426</td>
<td>.02448</td>
<td>.95123</td>
<td>0</td>
<td>.97571</td>
<td>1.02489</td>
<td>.4978</td>
</tr>
<tr>
<td>.08</td>
<td>.03828</td>
<td>.03856</td>
<td>.92312</td>
<td>0</td>
<td>.96168</td>
<td>1.03965</td>
<td>.4981</td>
</tr>
<tr>
<td>.10</td>
<td>.04742</td>
<td>.04769</td>
<td>.90484</td>
<td>0</td>
<td>.95253</td>
<td>1.04983</td>
<td>.4983</td>
</tr>
<tr>
<td>.50</td>
<td>.2013</td>
<td>.1919</td>
<td>.60653</td>
<td>0</td>
<td>.79843</td>
<td>1.25245</td>
<td>.5049</td>
</tr>
<tr>
<td>1.</td>
<td>.3393</td>
<td>.2924</td>
<td>.36788</td>
<td>0</td>
<td>.66028</td>
<td>1.514</td>
<td>.5140</td>
</tr>
<tr>
<td>2.</td>
<td>.5150</td>
<td>.3492</td>
<td>.13533</td>
<td>0</td>
<td>.48453</td>
<td>2.064</td>
<td>.5320</td>
</tr>
<tr>
<td>3.</td>
<td>.6200</td>
<td>.3297</td>
<td>.0497</td>
<td>0</td>
<td>.37948</td>
<td>2.635</td>
<td>.5450</td>
</tr>
<tr>
<td>5.</td>
<td>.7367</td>
<td>.2561</td>
<td>.00674</td>
<td>0</td>
<td>.26284</td>
<td>3.805</td>
<td>.5610</td>
</tr>
<tr>
<td>10</td>
<td>.8629</td>
<td>.1342</td>
<td>.00004</td>
<td>0</td>
<td>.13424</td>
<td>7.45</td>
<td>.645</td>
</tr>
</tbody>
</table>
TABLE D.3

\( x = .1 \)  \( m = 1.2 \)  \( n = 1.5 \)

\( b_o = .4988 \)

\[
\begin{array}{cccccccc}
\tau_1 & R_D & T_D & DT & DR & T_{T_D+DT} & \frac{1}{T} & \frac{b}{T} = \left( \frac{1}{\tau_1} - \frac{1}{0.0835} \right)
\
.02 & .00988 & .0099 & .9047 & .07547 & .9146 & 1.09337 & .4935 \\
.05 & .02431 & .02435 & .8775 & .07340 & .90225 & 1.10834 & .4968 \\
.08 & .03828 & .03834 & .8519 & .07146 & .89024 & 1.12329 & .4973 \\
.10 & .04735 & .04741 & .8350 & .07022 & .88241 & 1.13326 & .4976 \\
.50 & .1945 & .1925 & .5593 & .05357 & .7518 & 1.33014 & .4933 \\
1. & .3146 & .3012 & .3391 & .04499 & .6403 & 1.56177 & .4783 \\
2. & .4507 & .3837 & .1247 & .04068 & .5084 & 1.967 & .4417 \\
3. & .5285 & .3853 & .04588 & .04009 & .43118 & 2.319 & .4118 \\
5. & .6230 & .3305 & .00621 & .04000 & .33671 & 2.970 & .3773 \\
10 & .7402 & .2199 & .00004 & .0400 & .21994 & 4.547 & .3463 \\
100 & .9294 & .03033 & 0 & .0400 & .03033 & 32.97 & .3189 \\
1000 & .9566 & .00316 & 0 & .0400 & .00316 & 316.45 & .3154 \\
\end{array}
\]

TABLE D.4

\( x = .1 \)  \( m = 1.8 \)  \( n = 1.5 \)

\( b_o = .4984 \)

\[
\begin{array}{cccccccc}
\tau_1 & R_D & T_D & DT & DR & T_{T_D+DT} & \frac{1}{T} & \frac{b}{T} = \left( \frac{1}{\tau_1} - \frac{1}{0.0835} \right)
\
.02 & .00988 & .0099 & .9047 & .07547 & .9146 & 1.09337 & .4935 \\
.05 & .0243 & .02436 & .8779 & .0734 & .90225 & 1.10834 & .4968 \\
.08 & .03827 & .03835 & .8519 & .07146 & .89024 & 1.12329 & .4973 \\
.10 & .04733 & .04743 & .8350 & .07022 & .88241 & 1.13326 & .4976 \\
.50 & .1944 & .1926 & .5593 & .05357 & .7519 & 1.32996 & .4929 \\
1. & .3145 & .3012 & .3391 & .04499 & .6403 & 1.56177 & .4782 \\
2. & .4506 & .3838 & .1247 & .04068 & .5085 & 1.965 & .4415 \\
3. & .5284 & .3854 & .04588 & .04009 & .43128 & 2.3187 & .4117 \\
5. & .6229 & .3306 & .00621 & .04 & .33681 & 2.969 & .3771 \\
10 & .7402 & .2200 & .00004 & .04 & .22004 & 4.544 & .3460 \\
\end{array}
\]
TABLE D.5

\( x=1 \). \text{ } m=1.5 \text{ } n=1.

\[ b_0 = 0.3577 \]

<table>
<thead>
<tr>
<th>( t _1 )</th>
<th>( R _D )</th>
<th>( T _D )</th>
<th>( DT )</th>
<th>( DR )</th>
<th>( T_\text{D}+DT )</th>
<th>( 1/T )</th>
<th>( b=(\frac{1}{T}-1) )</th>
<th>( t _1 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.02</td>
<td>0.00712</td>
<td>0.01267</td>
<td>0.9802</td>
<td>0</td>
<td>0.99287</td>
<td>1.00718</td>
<td>0.3590</td>
<td></td>
</tr>
<tr>
<td>0.05</td>
<td>0.01769</td>
<td>0.03104</td>
<td>0.95123</td>
<td>0</td>
<td>0.98227</td>
<td>1.01805</td>
<td>0.3610</td>
<td></td>
</tr>
<tr>
<td>0.08</td>
<td>0.02812</td>
<td>0.0487</td>
<td>0.92312</td>
<td>0</td>
<td>0.97182</td>
<td>1.02899</td>
<td>0.3624</td>
<td></td>
</tr>
<tr>
<td>0.10</td>
<td>0.03500</td>
<td>0.06010</td>
<td>0.90484</td>
<td>0</td>
<td>0.96494</td>
<td>1.03633</td>
<td>0.3633</td>
<td></td>
</tr>
<tr>
<td>0.50</td>
<td>0.1590</td>
<td>0.2342</td>
<td>0.60653</td>
<td>0</td>
<td>0.84073</td>
<td>1.18944</td>
<td>0.3789</td>
<td></td>
</tr>
<tr>
<td>1.0</td>
<td>0.2821</td>
<td>0.3496</td>
<td>0.36788</td>
<td>0</td>
<td>0.71748</td>
<td>1.39376</td>
<td>0.3938</td>
<td></td>
</tr>
<tr>
<td>2.0</td>
<td>0.4529</td>
<td>0.4112</td>
<td>0.13533</td>
<td>0</td>
<td>0.54653</td>
<td>1.8297</td>
<td>0.4148</td>
<td></td>
</tr>
<tr>
<td>3.0</td>
<td>0.5622</td>
<td>0.3874</td>
<td>0.04979</td>
<td>0</td>
<td>0.4372</td>
<td>2.2873</td>
<td>0.4291</td>
<td></td>
</tr>
<tr>
<td>5.0</td>
<td>0.6896</td>
<td>0.3031</td>
<td>0.00674</td>
<td>0</td>
<td>0.30984</td>
<td>3.2275</td>
<td>0.4455</td>
<td></td>
</tr>
<tr>
<td>10.0</td>
<td>0.837</td>
<td>0.1608</td>
<td>0.00004</td>
<td>0</td>
<td>0.16084</td>
<td>6.217</td>
<td>0.5217</td>
<td></td>
</tr>
</tbody>
</table>

TABLE D.6

\( x=1 \). \text{ } m=2.7 \text{ } n=1.

\[ b_0 = 0.1131 \]

<table>
<thead>
<tr>
<th>( t _1 )</th>
<th>( R _D )</th>
<th>( T _D )</th>
<th>( DT )</th>
<th>( DR )</th>
<th>( T_\text{D}+DT )</th>
<th>( 1/T )</th>
<th>( b=(\frac{1}{T}-1) )</th>
<th>( t _1 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.02</td>
<td>0.00226</td>
<td>0.01753</td>
<td>0.9802</td>
<td>0</td>
<td>0.99773</td>
<td>1.00227</td>
<td>0.1135</td>
<td></td>
</tr>
<tr>
<td>0.05</td>
<td>0.00587</td>
<td>0.04286</td>
<td>0.95123</td>
<td>0</td>
<td>0.99409</td>
<td>1.00594</td>
<td>0.1188</td>
<td></td>
</tr>
<tr>
<td>0.08</td>
<td>0.00971</td>
<td>0.06712</td>
<td>0.92312</td>
<td>0</td>
<td>0.99024</td>
<td>1.00984</td>
<td>0.1230</td>
<td></td>
</tr>
<tr>
<td>0.10</td>
<td>0.01237</td>
<td>0.08272</td>
<td>0.90484</td>
<td>0</td>
<td>0.92473</td>
<td>1.08139</td>
<td>0.1628</td>
<td></td>
</tr>
<tr>
<td>1.0</td>
<td>0.1586</td>
<td>0.4730</td>
<td>0.36788</td>
<td>0</td>
<td>0.84088</td>
<td>1.18923</td>
<td>0.1892</td>
<td></td>
</tr>
<tr>
<td>2.0</td>
<td>0.3042</td>
<td>0.5599</td>
<td>0.13533</td>
<td>0</td>
<td>0.69523</td>
<td>1.4384</td>
<td>0.2192</td>
<td></td>
</tr>
<tr>
<td>3.0</td>
<td>0.4140</td>
<td>0.5355</td>
<td>0.04979</td>
<td>0</td>
<td>0.58529</td>
<td>1.7085</td>
<td>0.2363</td>
<td></td>
</tr>
<tr>
<td>5.0</td>
<td>0.5588</td>
<td>0.4338</td>
<td>0.00674</td>
<td>0</td>
<td>0.44054</td>
<td>2.270</td>
<td>0.2540</td>
<td></td>
</tr>
<tr>
<td>10.0</td>
<td>0.7607</td>
<td>0.2389</td>
<td>0.00004</td>
<td>0</td>
<td>0.23894</td>
<td>4.185</td>
<td>0.3185</td>
<td></td>
</tr>
</tbody>
</table>
### TABLE D.7

\(x=1\). \(m=1.2\) \(n=1.5\)

\(b_o=.3754\)

<table>
<thead>
<tr>
<th>(\tau_1)</th>
<th>(R_D)</th>
<th>(T_D)</th>
<th>(DT)</th>
<th>(DR)</th>
<th>(T=\frac{T_D+DT}{1/T})</th>
<th>(\frac{1}{T})</th>
</tr>
</thead>
<tbody>
<tr>
<td>.02</td>
<td>.00889</td>
<td>.01088</td>
<td>.9047</td>
<td>.07547</td>
<td>.91558</td>
<td>1.09220</td>
</tr>
<tr>
<td>.05</td>
<td>.02191</td>
<td>.02675</td>
<td>.8775</td>
<td>.07340</td>
<td>.90465</td>
<td>1.10539</td>
</tr>
<tr>
<td>.08</td>
<td>.03454</td>
<td>.04207</td>
<td>.8519</td>
<td>.07146</td>
<td>.89397</td>
<td>1.11860</td>
</tr>
<tr>
<td>.10</td>
<td>.04275</td>
<td>.05201</td>
<td>.8350</td>
<td>.07022</td>
<td>.88701</td>
<td>1.12738</td>
</tr>
<tr>
<td>.50</td>
<td>.1776</td>
<td>.2093</td>
<td>.5593</td>
<td>.05357</td>
<td>.7686</td>
<td>1.30106</td>
</tr>
<tr>
<td>1.</td>
<td>.2901</td>
<td>.3256</td>
<td>.3391</td>
<td>.04499</td>
<td>.6647</td>
<td>1.5044</td>
</tr>
<tr>
<td>2.</td>
<td>.4198</td>
<td>.4145</td>
<td>.1247</td>
<td>.04068</td>
<td>.5392</td>
<td>1.8546</td>
</tr>
<tr>
<td>3.</td>
<td>.4979</td>
<td>.4188</td>
<td>.04588</td>
<td>.04009</td>
<td>.46468</td>
<td>2.1520</td>
</tr>
<tr>
<td>5.</td>
<td>.5882</td>
<td>.3653</td>
<td>.00621</td>
<td>.04000</td>
<td>.37151</td>
<td>2.6917</td>
</tr>
<tr>
<td>10</td>
<td>.7098</td>
<td>.2503</td>
<td>.00004</td>
<td>.0400</td>
<td>.25034</td>
<td>3.9945</td>
</tr>
</tbody>
</table>

### TABLE D.8

\(x=1\). \(m=1.8\) \(n=1.5\)

\(b_o=.3283\)

<table>
<thead>
<tr>
<th>(\tau_1)</th>
<th>(R_D)</th>
<th>(T_D)</th>
<th>(DT)</th>
<th>(DR)</th>
<th>(T=\frac{T_D+DT}{1/T})</th>
<th>(\frac{1}{T})</th>
</tr>
</thead>
<tbody>
<tr>
<td>.02</td>
<td>.00860</td>
<td>.01118</td>
<td>.9047</td>
<td>.07547</td>
<td>.91588</td>
<td>1.09184</td>
</tr>
<tr>
<td>.05</td>
<td>.02118</td>
<td>.02747</td>
<td>.8779</td>
<td>.07340</td>
<td>.90537</td>
<td>1.10452</td>
</tr>
<tr>
<td>.08</td>
<td>.03340</td>
<td>.04321</td>
<td>.8519</td>
<td>.07146</td>
<td>.89511</td>
<td>1.11718</td>
</tr>
<tr>
<td>.10</td>
<td>.04135</td>
<td>.05340</td>
<td>.8350</td>
<td>.07022</td>
<td>.8884</td>
<td>1.12562</td>
</tr>
<tr>
<td>.50</td>
<td>.1724</td>
<td>.2146</td>
<td>.5593</td>
<td>.05357</td>
<td>.7739</td>
<td>1.29215</td>
</tr>
<tr>
<td>1.</td>
<td>.2820</td>
<td>.3336</td>
<td>.3391</td>
<td>.04499</td>
<td>.6727</td>
<td>1.4865</td>
</tr>
<tr>
<td>2.</td>
<td>.4088</td>
<td>.4255</td>
<td>.1247</td>
<td>.04068</td>
<td>.5502</td>
<td>1.8175</td>
</tr>
<tr>
<td>3.</td>
<td>.4824</td>
<td>.4313</td>
<td>.04588</td>
<td>.04009</td>
<td>.47718</td>
<td>2.0956</td>
</tr>
<tr>
<td>5.</td>
<td>.5746</td>
<td>.3788</td>
<td>.00621</td>
<td>.0400</td>
<td>.38501</td>
<td>2.5973</td>
</tr>
<tr>
<td>10</td>
<td>.6973</td>
<td>.2929</td>
<td>.00004</td>
<td>.04</td>
<td>.26294</td>
<td>3.8031</td>
</tr>
</tbody>
</table>
TABLE D.9

\( x=2, \ m=1.5, \ n=1.0 \)

\( b_o = .0668 \)

<table>
<thead>
<tr>
<th>( t_1 )</th>
<th>( R_D )</th>
<th>( T_D )</th>
<th>( DT )</th>
<th>( DR )</th>
<th>( T= )</th>
<th>( 1/T )</th>
<th>( b=(1/T-1) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>.02</td>
<td>.00139</td>
<td>.01840</td>
<td>.9802</td>
<td>0</td>
<td>.9986</td>
<td>1.0014</td>
<td>.0700</td>
</tr>
<tr>
<td>.05</td>
<td>.00363</td>
<td>.0451</td>
<td>.95123</td>
<td>0</td>
<td>.9963</td>
<td>1.00371</td>
<td>.0742</td>
</tr>
<tr>
<td>.08</td>
<td>.00606</td>
<td>.07077</td>
<td>.92312</td>
<td>0</td>
<td>.99389</td>
<td>1.0061</td>
<td>.0762</td>
</tr>
<tr>
<td>.10</td>
<td>.00776</td>
<td>.08734</td>
<td>.90484</td>
<td>0</td>
<td>.9922</td>
<td>1.00786</td>
<td>.0786</td>
</tr>
<tr>
<td>.50</td>
<td>.05077</td>
<td>.3424</td>
<td>.60653</td>
<td>0</td>
<td>.9489</td>
<td>1.05385</td>
<td>.1077</td>
</tr>
<tr>
<td>1.</td>
<td>.1144</td>
<td>.5172</td>
<td>.3679</td>
<td>0</td>
<td>.8851</td>
<td>1.1298</td>
<td>.1298</td>
</tr>
<tr>
<td>2.</td>
<td>.2380</td>
<td>.6260</td>
<td>.1353</td>
<td>0</td>
<td>.7613</td>
<td>1.3135</td>
<td>.1567</td>
</tr>
<tr>
<td>3.</td>
<td>.3405</td>
<td>.6090</td>
<td>.04978</td>
<td>0</td>
<td>.6588</td>
<td>1.5179</td>
<td>.1726</td>
</tr>
<tr>
<td>5.</td>
<td>.5767</td>
<td>.4215</td>
<td>.00674</td>
<td>0</td>
<td>.4282</td>
<td>2.3353</td>
<td>.2671</td>
</tr>
</tbody>
</table>

TABLE D.10

\( x=2, \ m=2.7, \ n=1 \)

\( b_o = .5953 \)

<table>
<thead>
<tr>
<th>( t_1 )</th>
<th>( R_D )</th>
<th>( T_D )</th>
<th>( DT )</th>
<th>( DR )</th>
<th>( T= )</th>
<th>( 1/T )</th>
<th>( b=(1/T-1) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>.02</td>
<td>.01170</td>
<td>.00808</td>
<td>.9802</td>
<td>0</td>
<td>.9883</td>
<td>1.0118</td>
<td>.506</td>
</tr>
<tr>
<td>.05</td>
<td>.02859</td>
<td>.02015</td>
<td>.95123</td>
<td>0</td>
<td>.9714</td>
<td>1.02944</td>
<td>.589</td>
</tr>
<tr>
<td>.08</td>
<td>.04473</td>
<td>.03209</td>
<td>.92311</td>
<td>0</td>
<td>.9552</td>
<td>1.0469</td>
<td>.589</td>
</tr>
<tr>
<td>.10</td>
<td>.05512</td>
<td>.03995</td>
<td>.90484</td>
<td>0</td>
<td>.9448</td>
<td>1.0584</td>
<td>.584</td>
</tr>
<tr>
<td>.50</td>
<td>.2200</td>
<td>.1731</td>
<td>.60653</td>
<td>0</td>
<td>.7796</td>
<td>1.28271</td>
<td>.565</td>
</tr>
<tr>
<td>1.</td>
<td>.3584</td>
<td>.2732</td>
<td>.3679</td>
<td>0</td>
<td>.6411</td>
<td>1.5598</td>
<td>.5598</td>
</tr>
<tr>
<td>2.</td>
<td>.5295</td>
<td>.3344</td>
<td>.1353</td>
<td>0</td>
<td>.4697</td>
<td>2.129</td>
<td>.6099</td>
</tr>
<tr>
<td>3.</td>
<td>.6313</td>
<td>.3181</td>
<td>.04978</td>
<td>0</td>
<td>.3679</td>
<td>2.718</td>
<td>.5727</td>
</tr>
<tr>
<td>5.</td>
<td>.7777</td>
<td>.2151</td>
<td>.00674</td>
<td>0</td>
<td>.2218</td>
<td>4.508</td>
<td>.7016</td>
</tr>
</tbody>
</table>
TABLE D.11

\(x=2. \ m=1.2 \ n=1.5\)

\(b_o = 0.0462\)

<table>
<thead>
<tr>
<th>(\tau_1)</th>
<th>(R_D)</th>
<th>(T_D)</th>
<th>(DT)</th>
<th>(DR)</th>
<th>(T=T_D+DT)</th>
<th>(1/T)</th>
<th>(b=(1/T-1.0835)/\tau_1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.02</td>
<td>0.00592</td>
<td>0.01386</td>
<td>0.9047</td>
<td>0.07547</td>
<td>0.9186</td>
<td>1.08861</td>
<td>0.2555</td>
</tr>
<tr>
<td>0.05</td>
<td>0.01462</td>
<td>0.03403</td>
<td>0.8779</td>
<td>0.07340</td>
<td>0.9119</td>
<td>1.09661</td>
<td>0.2622</td>
</tr>
<tr>
<td>0.08</td>
<td>0.02312</td>
<td>0.05349</td>
<td>0.8519</td>
<td>0.07146</td>
<td>0.9054</td>
<td>1.10448</td>
<td>0.2622</td>
</tr>
<tr>
<td>0.10</td>
<td>0.02867</td>
<td>0.06607</td>
<td>0.8350</td>
<td>0.07022</td>
<td>0.9011</td>
<td>1.10975</td>
<td>0.2625</td>
</tr>
<tr>
<td>0.15</td>
<td>0.1237</td>
<td>0.2632</td>
<td>0.5593</td>
<td>0.05357</td>
<td>0.8225</td>
<td>1.2168</td>
<td>0.2646</td>
</tr>
<tr>
<td>1.0</td>
<td>0.2092</td>
<td>0.4064</td>
<td>0.3391</td>
<td>0.04499</td>
<td>0.7455</td>
<td>1.3414</td>
<td>0.2579</td>
</tr>
<tr>
<td>2.0</td>
<td>0.3129</td>
<td>0.5213</td>
<td>0.1247</td>
<td>0.04068</td>
<td>0.6460</td>
<td>1.5480</td>
<td>0.2322</td>
</tr>
<tr>
<td>3.0</td>
<td>0.3716</td>
<td>0.5420</td>
<td>0.04588</td>
<td>0.04009</td>
<td>0.5879</td>
<td>1.701</td>
<td>0.2058</td>
</tr>
<tr>
<td>5.0</td>
<td>0.4438</td>
<td>0.5100</td>
<td>0.00621</td>
<td>0.04</td>
<td>0.5162</td>
<td>1.937</td>
<td>0.1707</td>
</tr>
</tbody>
</table>

TABLE D.12

\(x=2. \ m=1.8 \ n=1.5\)

\(b_o = 0.1501\)

<table>
<thead>
<tr>
<th>(\tau_1)</th>
<th>(R_D)</th>
<th>(T_D)</th>
<th>(DT)</th>
<th>(DR)</th>
<th>(T=T_D+DT)</th>
<th>(1/T)</th>
<th>(b=(1/T-1.0835)/\tau_1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.02</td>
<td>0.00663</td>
<td>0.01314</td>
<td>0.9047</td>
<td>0.07547</td>
<td>0.9178</td>
<td>1.08956</td>
<td>0.3032</td>
</tr>
<tr>
<td>0.05</td>
<td>0.01637</td>
<td>0.03228</td>
<td>0.8779</td>
<td>0.07340</td>
<td>0.9102</td>
<td>1.09866</td>
<td>0.3032</td>
</tr>
<tr>
<td>0.08</td>
<td>0.02588</td>
<td>0.05073</td>
<td>0.8519</td>
<td>0.07146</td>
<td>0.9026</td>
<td>1.10791</td>
<td>0.3051</td>
</tr>
<tr>
<td>0.10</td>
<td>0.03208</td>
<td>0.06266</td>
<td>0.8350</td>
<td>0.07022</td>
<td>0.8977</td>
<td>1.11396</td>
<td>0.3046</td>
</tr>
<tr>
<td>0.15</td>
<td>0.1375</td>
<td>0.2495</td>
<td>0.5593</td>
<td>0.05357</td>
<td>0.8088</td>
<td>1.2364</td>
<td>0.3058</td>
</tr>
<tr>
<td>1.0</td>
<td>0.2311</td>
<td>0.3845</td>
<td>0.3391</td>
<td>0.04499</td>
<td>0.7236</td>
<td>1.3820</td>
<td>0.2985</td>
</tr>
<tr>
<td>2.0</td>
<td>0.3446</td>
<td>0.4896</td>
<td>0.247</td>
<td>0.04068</td>
<td>0.6143</td>
<td>1.628</td>
<td>0.2722</td>
</tr>
<tr>
<td>3.0</td>
<td>0.4104</td>
<td>0.5032</td>
<td>0.04588</td>
<td>0.04009</td>
<td>0.5491</td>
<td>1.821</td>
<td>0.2458</td>
</tr>
<tr>
<td>5.0</td>
<td>0.4929</td>
<td>0.4608</td>
<td>0.00621</td>
<td>0.04</td>
<td>0.467</td>
<td>2.141</td>
<td>0.2115</td>
</tr>
</tbody>
</table>
### TABLE D.13

\(x = 4, \ m = 1.5, \ n = 1.0\)

\(b \approx 0.0708\)

<table>
<thead>
<tr>
<th>(\tau_1)</th>
<th>(R_D)</th>
<th>(T_D)</th>
<th>(DT)</th>
<th>(DR)</th>
<th>(\frac{T}{T_D + DT})</th>
<th>(1/T)</th>
<th>(b = \left(\frac{1}{T} - 1\right) / \tau_1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.02</td>
<td>0.00142</td>
<td>0.01822</td>
<td>0.98019</td>
<td>0</td>
<td>0.99841</td>
<td>1.00159</td>
<td>0.0795</td>
</tr>
<tr>
<td>0.05</td>
<td>0.00359</td>
<td>0.04477</td>
<td>0.95123</td>
<td>0</td>
<td>0.996</td>
<td>1.00401</td>
<td>0.0802</td>
</tr>
<tr>
<td>0.08</td>
<td>0.00579</td>
<td>0.07044</td>
<td>0.92312</td>
<td>0</td>
<td>0.99356</td>
<td>1.00648</td>
<td>0.0810</td>
</tr>
<tr>
<td>0.10</td>
<td>0.00728</td>
<td>0.08708</td>
<td>0.90484</td>
<td>0</td>
<td>0.99192</td>
<td>1.00814</td>
<td>0.0814</td>
</tr>
<tr>
<td>0.50</td>
<td>0.03881</td>
<td>0.3512</td>
<td>0.60653</td>
<td>0</td>
<td>0.95773</td>
<td>1.04413</td>
<td>0.0882</td>
</tr>
<tr>
<td>1.0</td>
<td>0.08037</td>
<td>0.5459</td>
<td>0.36788</td>
<td>0</td>
<td>0.91378</td>
<td>1.09435</td>
<td>0.0943</td>
</tr>
<tr>
<td>2.0</td>
<td>0.1634</td>
<td>0.6927</td>
<td>0.13533</td>
<td>0</td>
<td>0.828</td>
<td>1.208</td>
<td>0.1040</td>
</tr>
<tr>
<td>3.0</td>
<td>0.2405</td>
<td>0.6998</td>
<td>0.04978</td>
<td>0</td>
<td>0.74958</td>
<td>1.334</td>
<td>0.1113</td>
</tr>
<tr>
<td>5.0</td>
<td>0.3686</td>
<td>0.6142</td>
<td>0.00674</td>
<td>0</td>
<td>0.62094</td>
<td>1.610</td>
<td>0.1220</td>
</tr>
<tr>
<td>10.0</td>
<td>0.5630</td>
<td>0.4262</td>
<td>0</td>
<td>0</td>
<td>0.4267</td>
<td>2.35</td>
<td>0.135</td>
</tr>
<tr>
<td>100.0</td>
<td>0.9259</td>
<td>0.06336</td>
<td>0</td>
<td>0</td>
<td>0.06336</td>
<td>15.8</td>
<td>0.148</td>
</tr>
<tr>
<td>1000.0</td>
<td>0.9826</td>
<td>0.006657</td>
<td>0</td>
<td>0</td>
<td>0.006657</td>
<td>150.0</td>
<td>0.149</td>
</tr>
</tbody>
</table>

### TABLE D.14

\(x = 4, \ m = 2.7, \ n = 1.0\)

\(b \approx 0.3609\)

<table>
<thead>
<tr>
<th>(\tau_1)</th>
<th>(R_D)</th>
<th>(T_D)</th>
<th>(DT)</th>
<th>(DR)</th>
<th>(\frac{T}{T_D + DT})</th>
<th>(1/T)</th>
<th>(b = \left(\frac{1}{T} - 1\right) / \tau_1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.02</td>
<td>0.00712</td>
<td>0.0125</td>
<td>0.98019</td>
<td>0</td>
<td>0.99269</td>
<td>1.00736</td>
<td>0.3680</td>
</tr>
<tr>
<td>0.05</td>
<td>0.01767</td>
<td>0.03064</td>
<td>0.95123</td>
<td>0</td>
<td>0.98187</td>
<td>1.01846</td>
<td>0.3692</td>
</tr>
<tr>
<td>0.08</td>
<td>0.02807</td>
<td>0.04807</td>
<td>0.92312</td>
<td>0</td>
<td>0.97119</td>
<td>1.02966</td>
<td>0.3707</td>
</tr>
<tr>
<td>0.10</td>
<td>0.03493</td>
<td>0.05932</td>
<td>0.90484</td>
<td>0</td>
<td>0.96416</td>
<td>1.03717</td>
<td>0.3717</td>
</tr>
<tr>
<td>0.50</td>
<td>0.1583</td>
<td>0.2313</td>
<td>0.60653</td>
<td>0</td>
<td>0.83783</td>
<td>1.1935</td>
<td>0.3870</td>
</tr>
<tr>
<td>1.0</td>
<td>0.2802</td>
<td>0.3457</td>
<td>0.36788</td>
<td>0</td>
<td>0.71358</td>
<td>1.401</td>
<td>0.401</td>
</tr>
<tr>
<td>2.0</td>
<td>0.4488</td>
<td>0.4072</td>
<td>0.13533</td>
<td>0</td>
<td>0.5425</td>
<td>1.843</td>
<td>0.4215</td>
</tr>
<tr>
<td>3.0</td>
<td>0.5566</td>
<td>0.384</td>
<td>0.04978</td>
<td>0</td>
<td>0.43378</td>
<td>2.305</td>
<td>0.4350</td>
</tr>
<tr>
<td>5.0</td>
<td>0.7275</td>
<td>0.2558</td>
<td>0.00674</td>
<td>0</td>
<td>0.2625</td>
<td>3.809</td>
<td>0.5618</td>
</tr>
</tbody>
</table>
APPENDIX E

Details of Apparatus

The various components of the apparatus were chosen by Erickson (9) and slightly modified by Fahimian (31). Only a brief description is given here.

The mercury arc lamp is of the type GE B-H6, cooled with oil-free air at 20 psia (55), and is characterized by its high brightness and the fact that the radiation from this lamp is concentrated in narrow spectral bands. Thus, by using an appropriate optical interference filter, a suitably measurable scattered-light intensity, having a fairly narrow wavelength band width, is possible. The average spectral distribution is given in Figure E.1. The radiation is mainly concentrated to the mercury lines at 3650Å, 4047Å, 4358Å and 5461Å. In the present work the lines at 4358Å and 5461Å were chosen.

The signal generated by the phototube is proportional to the intensity of the mercury arc lamp and the sensitivity the multiplier phototube has at the particular wavelength Figure E.2 shows the spectral sensitivity of the RCA 931-A multiplier phototube.

A narrow wavelength band is defined about 4358Å or 5461Å by a narrow band optical interference filter. The filters used are of the type B-1 with a peak wavelength
of 4358Å and 5461Å manufactured by Baird Atomic, Inc. These filters have a bandwidth at half of the peak transmission of approximately 65Å and have a peak transmission of about 50 percent.

The achromatic lenses used are of good quality and assure that the alignment of the system remains the same for any wavelength in the visible range. The two lenses used in the collimation system of the incident light have focal lengths of 180 mm and 508 mm. The detector lens has a focal length of 78 mm.

The circular cross section of the incident beam at the center of the system is determined by the aperture in front of the light source, the lenses and the light stops in the collimation system of the incident light. The aperture at the light source is 0.0135 inch diameter drilled in 5 mil stainless steel sheet. The light stop at the end of the lens tube of the incident beam is .75 inch in diameter. The source beam had a divergence of approximately ± 0.75 degrees. As mentioned before, the cross section of the beam on a slide at the center of rotation of the system is .12 cm.

The light stop in front of the lens tube of the collection system is located at a distance from the center of the system equal to 22.125 inches and its cross section has a diameter .508 inch. The divergence of the scattered beam is ± 0.66 degree.
The circular aperture between the multiplier phototube and polarization filter defines the viewing area of the scattering sample seen by the photosensitive surface. The viewing area is measured by placing a lamp behind the aperture and observing the image formed at various aperture sizes used. They are drilled in 5 mil stainless steel sheet. For single scatter experiments the aperture used has a diameter .105 cm. The image formed at the center of the system is .87 cm in diameter. For multiple scatter experiments the diameter of the aperture used is .2 cm and the detector is able to view a circular area normal to it 1.66 cm in diameter.

The light passing through the aperture becomes linearly polarized by a neutral color polarizing filter of type HN22 with a transmittance of 22 percent for white light. When two filters have their axes crossed, they transmit .0005 percent of the incident unpolarized radiation.

The circuit diagram for the multiplier phototube is shown in Figure E.3. The high voltage required to operate the RCA931A multiplier phototube is supplied by a regulated high-voltage supply model RE-1602 manufactured by the Northeast Scientific Corp. The phototube output voltage was measured with a model 400AC Hewlett-Packard Voltmeter.
Figure E.1  Average spectral distribution from a GE B-H6 mercury arc lamp
Figure E.2  Spectral sensitivity characteristic of an RCA 931-A multiplier phototube for equal values of radiant flux at all wavelengths.
Figure E.3 Arrangement of electronic equipment for light-scattering apparatus
APPENDIX F

Data

In the following two sections equations are presented for the reduction of the data. Some additional numerical results are presented also.

F.1 Calculation of Mie Scattering Parameters for Very Dilute Particles Suspensions

In this section the relations for converting the photomultiplier output voltage e(θ) to the Mie scattering coefficients i(θ) are derived. A schematic diagram of the optical equipment and the definition of some of the physical parameters involved are given below:

Figure F.1 Optical System for Measuring the Incident Light Intensity

$I'_i =$ intensity of collimated beam incident on particles

$I'_s =$ intensity of beam picked up by detector

$A_o =$ area of illuminated by incident beam at the center of the system
$A_3 =$ area of aperture at detector during scattering measurements

$A'_3 =$ area of incident beam at detector

$r =$ distance from center of the system to aperture $A_3$

$NF =$ neutral filter

$F =$ optical interference filter

$e_{\perp}, e_{\parallel} =$ voltage measured from the photomultiplier output at $0^\circ$ and with the aperture $A_3$ removed; $e_{\perp}$ for the perpendicular, $e_{\parallel}$ for parallel component of polarization.

$e_0 = e_{\perp} + e_{\parallel}$ total voltage output

$e_{\perp}(\theta), e_{\parallel}(\theta), e(\theta) =$ voltage measured from the photomultiplier output at $\theta$ degrees with the aperture $A_3$ in place

$e_{\perp}(\theta)$ for perpendicular, $e_{\parallel}(\theta)$ for parallel component of polarization; $e(\theta)$ total $e_{st}(\theta)$ monitor reading at $\theta$ degrees.

$e_{\perp}(\theta), e_{\parallel}(\theta), e_w(\theta) =$ voltage measured from millipore distilled water (background scattering)

$e_{wst}(\theta) =$ monitor reading for the water solution at $\theta$ degrees.

$s =$ sensitivity of photomultiplier in volts/light energy.

For scatter out of one cc of an air-suspension of spheres containing $N_p$ particles per cc the Mie equation for the intensity at angle $\theta$ is given by:

$$\frac{I_3(\theta)}{I_0} = N_p \frac{k^2 r^2}{i(\theta)}$$

(F-1)
where $k = \frac{2\pi}{(\lambda/\eta_w)}$; intensity is here defined as the energy flux per unit area; its cgs units are erg per cm² per sec.

Equation F-1 needs to be modified to allow for the characteristics of the equipment and the cylindrical cell used. With reference to Figure F-1 the source beam in the absence of scatter yields a signal $e_0$ given by:

$$e_0 = s \, I_3'(o) \, A_3' \tag{F-2}$$

But since the incident beam is collimated,

$$I_3'(o) \, A_3' = I_o' \cdot A_0 \tag{F-3}$$

$$e_0 = s \cdot I_o' \cdot A_0 \tag{F-4}$$

$$I_o' = \frac{e_0}{s \cdot A_0} \tag{F-5}$$

For a measurement of radiation scattered at $\theta^o$:

$$e(\theta) = s \cdot I_3'(\theta) \cdot A_3 \tag{F-6}$$

$$I_3'(\theta) = \frac{e(\theta)}{s \cdot A_3} \tag{F-7}$$

Combination of (F-5) and (F-7) yields:

$$\frac{I_3'(\theta)}{I_o'} = \frac{e(\theta)}{e_0} \cdot \frac{A_0}{A_3} \tag{F-8}$$

Substitution of (F-8) into (F-1) gives,

$$i(\theta) = \frac{e(\theta)}{e_0} \cdot \frac{A_0}{A_3} \cdot \frac{k^2r^2}{N_p} \tag{F-9}$$
The illuminated volume element and hence the number of particles viewed by the detector changes with the angle of observation $\theta$. Let us call $D_e$ the diameter of the circular area formed on a glass slide at the center of the system when a lamp is placed behind the phototube aperture at $0^\circ$ degrees. The volume element viewed by the phototube at $0^\circ$ equals $V_o = D_e^2 A_o$, while for any other angle of observation $\theta$ is:

$$ V(\theta) = \frac{D_e^2 A_o}{\sin \theta} $$

Correction of equation (F-9) for the volume changed with angle $\theta$ and subtraction from the signal $e(\theta)$ of the background scattering $e_w(\theta)$, obtained by measuring the scatter in the reference direction by millipore distilled water, gives:

$$ i(\theta) = \frac{e_{st}(0)}{e_o} \frac{n_w^2}{A_3 \cdot D_e} \frac{k^2 r^2}{N_p} \sin \theta \left[ \frac{e(\theta)}{e_{st}(\theta)} - \frac{e_w(\theta)}{e_{wst}(\theta)} \right] \quad (F-10) $$

where the factor $n_w^2$ is included to correct for the change in divergence of the collection angle introduced by the transition of a beam from the water to the air phase; correction for variations in the source beam intensity is made by normalizing the readings by the monitor signals $e_{st}(\theta)$, $e_{wst}(\theta)$ and $e_{st}(o)$ (see Figure F.1)
Tominatsu and Palmer (56) have modified equation (F-10) to include corrections for reflection effects from the glass-air interfaces of a cylindrical cell of the Brice-Phoenix type. The following equation results:

\[
i(\theta) = \frac{n_w^2}{A_3 \cdot D_\perp} \cdot \frac{k^2 r^2}{N_p} \frac{1}{1.049(1-f)^2(1-4f^2)} \sin \theta \frac{e_{st}(\theta)}{e_o}
\]

\[
\left(\frac{e(\theta)}{e_{st}(\theta)} - \frac{e_w(\theta)}{e_{wst}(\theta)}\right) - 2f \left[\frac{e(180-\theta)}{e_{st}(180-\theta)} \frac{e_w(180-\theta)}{e_{wst}(180-\theta)}\right]
\]

where \(e(\theta) = e_\perp(\theta) + e_{||}(\theta)\), \(e_\perp(\theta)\) and \(e_{||}(\theta)\) being the output voltage for perpendicular and parallel polarization respectively, and \(f\) is the reflectivity of the air-glass interface.

Equation (F-11) includes first order corrections, neglecting the essentially negligible reflections at the glass-solution interfaces, of the following kind:

(a) reflection of the primary beam at the entrance and exit windows; (b) reflection of the light scattered in the \(180 + \theta\) direction; and (c) reflection of the scattered light at the air-glass interface in the direction of the measurement, \(\theta\).

Introduction of the \(e_\perp(\theta)\) and \(e_{||}(\theta)\) values into (F-11) gives \(i_\perp(\theta)\) and \(i_{||}(\theta)\). The logarithms of \(i_\perp(\theta)\) and \(i_{||}(\theta)\) thus calculated were presented in the plots of the results section. In the following a listing
of the FORTRAN Program used to calculate $i_\perp(\theta)$ and $i_\parallel(\theta)$ is presented.
### Key to the Fortran Program

<table>
<thead>
<tr>
<th>Variable</th>
<th>Fortran Symbol</th>
</tr>
</thead>
<tbody>
<tr>
<td>d</td>
<td>D</td>
</tr>
<tr>
<td>λ</td>
<td>WAVELO</td>
</tr>
<tr>
<td>nw</td>
<td>RNW</td>
</tr>
<tr>
<td>D e</td>
<td>DL</td>
</tr>
<tr>
<td>πd/λ</td>
<td>ALPHA</td>
</tr>
<tr>
<td>e₀⊥</td>
<td>EOO</td>
</tr>
<tr>
<td>e₀∥</td>
<td>E9090</td>
</tr>
<tr>
<td>est (θ)</td>
<td>ESTDO</td>
</tr>
<tr>
<td>est (90)</td>
<td>ESTD90</td>
</tr>
<tr>
<td>k</td>
<td>AK</td>
</tr>
<tr>
<td>θ</td>
<td>ANGLE</td>
</tr>
<tr>
<td>e₁(θ)</td>
<td>EO(I)</td>
</tr>
<tr>
<td>e₁₁(θ)</td>
<td>E90(I)</td>
</tr>
<tr>
<td>est(θ)</td>
<td>ESTD(I)</td>
</tr>
<tr>
<td>ew₁(θ)</td>
<td>EWO(I)</td>
</tr>
<tr>
<td>ew₁₁(θ)</td>
<td>EW90(I)</td>
</tr>
<tr>
<td>ewst(θ)</td>
<td>EWSTD(I)</td>
</tr>
<tr>
<td>m_L (mass of Latex per cc of solution)</td>
<td>WPERCC</td>
</tr>
</tbody>
</table>

### Input

<table>
<thead>
<tr>
<th>ITEST</th>
<th>I3</th>
</tr>
</thead>
<tbody>
<tr>
<td>D, WAVELO, WPERCC</td>
<td>3E8.4</td>
</tr>
<tr>
<td>EOO, E9090,ESTDO,ESTD90</td>
<td>4F10.3</td>
</tr>
<tr>
<td>Number of Angles (L)</td>
<td>I3</td>
</tr>
<tr>
<td>ANGLE,EO,E90,ESTD,EWO,EW90,EWSTD</td>
<td>7F10.3</td>
</tr>
</tbody>
</table>
PROGRAM TO CALCULATE SINGLE SCATTERING OF PARTICLES INSIDE
A CRYSTAL

1000 FORMAT(1X, 'TIME=', I4)

100 FORMAT(1X, 'SCATTERED INTENSITY=', F6.2)

200 FORMAT(1X, 'TOTAL SCATTERED INTENSITY=', F6.2)

1000 FORMAT(1X, 'ENERGY=', F6.2)

200 FORMAT(1X, 'ENERGY=', F6.2)

300 FORMAT(1X, 'ENERGY=', F6.2)

1000 FORMAT(1X, 'ENERGY=', F6.2)

200 FORMAT(1X, 'ENERGY=', F6.2)

300 FORMAT(1X, 'ENERGY=', F6.2)

1000 FORMAT(1X, 'ENERGY=', F6.2)

200 FORMAT(1X, 'ENERGY=', F6.2)

300 FORMAT(1X, 'ENERGY=', F6.2)

1000 FORMAT(1X, 'ENERGY=', F6.2)

200 FORMAT(1X, 'ENERGY=', F6.2)

300 FORMAT(1X, 'ENERGY=', F6.2)

1000 FORMAT(1X, 'ENERGY=', F6.2)

200 FORMAT(1X, 'ENERGY=', F6.2)

300 FORMAT(1X, 'ENERGY=', F6.2)

1000 FORMAT(1X, 'ENERGY=', F6.2)

200 FORMAT(1X, 'ENERGY=', F6.2)

300 FORMAT(1X, 'ENERGY=', F6.2)

1000 FORMAT(1X, 'ENERGY=', F6.2)

200 FORMAT(1X, 'ENERGY=', F6.2)

300 FORMAT(1X, 'ENERGY=', F6.2)

1000 FORMAT(1X, 'ENERGY=', F6.2)

200 FORMAT(1X, 'ENERGY=', F6.2)

300 FORMAT(1X, 'ENERGY=', F6.2)

1000 FORMAT(1X, 'ENERGY=', F6.2)

200 FORMAT(1X, 'ENERGY=', F6.2)

300 FORMAT(1X, 'ENERGY=', F6.2)

1000 FORMAT(1X, 'ENERGY=', F6.2)

200 FORMAT(1X, 'ENERGY=', F6.2)

300 FORMAT(1X, 'ENERGY=', F6.2)

1000 FORMAT(1X, 'ENERGY=', F6.2)

200 FORMAT(1X, 'ENERGY=', F6.2)

300 FORMAT(1X, 'ENERGY=', F6.2)
F.2 Calculation of Bidirectional Reflectance and Transmittance of Multiple Scatter Experiments

In this section the bidirectional intensity distribution for multiple scatter experiments is derived from the output voltage of the phototube and the characteristics of the equipment. The optical arrangement of Figure F.1 is again used for this set of experiments.

Let us imagine a scattering slab of a polystyrene latex at the center of the system $P$. The incident flux on the front surface of the slab is:

$$Q_{\text{inc}} = I_0 d\Omega \cdot A_0$$  \hspace{1cm} (F-12)

$$e_0 = s \cdot Q_{\text{in}}$$  \hspace{1cm} (F-13)

where $d\Omega$ is the angle of divergence of the incident beam and $e_0 = e_0 \perp + e_0 \parallel$ measures the total incident flux ($e_0 \perp$ and $e_0 \parallel$ being the incident flux for perpendicular and parallel polarization respectively).

The incident beam is scattered by the latex over the surrounding space as determined by the principles of multiple scatter outlined in Section IV. The radiative flux scattered at angle $\theta$ is:

$$Q(\theta) = I(\theta) \cdot A_0 \cdot \cos \theta \cdot d\Omega_c$$ \hspace{1cm} (F-14)

$$e(\theta) = s \cdot Q(\theta)$$ \hspace{1cm} (F-15)

where $d\Omega_c$ is the divergence of the detected beam.
The bidirectional intensity function is defined as follows:

$$
\rho(\theta) = \frac{I(\theta)}{Q_{\text{inc}}/A_o \pi} \frac{e(\theta)/(s \cdot A_o \cdot \cos \theta \cdot d \Omega_C)}{e_o/(s \cdot A_o \cdot \pi)}
$$

or

$$
\rho(\theta) = \frac{\pi \cdot e(\theta)}{e_o \cos \theta \Delta \Omega_C}
$$

where $\Delta \Omega_C$ is now written for a finite collection angular divergence calculated from the system dimensions.

$r =$ distance between sample and detector aperture $A_o$

$A_o =$ area of detector aperture

$$
\Delta \Omega_C = \frac{A_o}{2} = \frac{\pi \cdot D_1^2}{4 \cdot r^2}
$$

On substituting this expression into equation (F-17) we get:

$$
\rho(\theta) = \frac{4r^2}{D_3^2 \cos \theta} \frac{1}{e_o} \frac{e(\theta)}{e_o}
$$

Replacement of $e(\theta)$ by $e_\perp(\theta)$ or $e_\parallel(\theta)$ the output voltage reading for perpendicular or parallel polarization gives the bidirectional intensity values for the two components of polarization.

Equation (F-19) is now rewritten to allow for the background scattering $e_w$ of the millipore distilled water and possible change in the monitor reading $I_{\text{st}}$ indicating
variation in $e_o$ with time.

\[ n_p = 1.59 - 1.61 \quad n_G = 1.5 \]

\[ n_w = 1.34 \]

![Diagram showing trace of contributions to reflectance and transmittance by beams that have been scattered more than once.]

**Figure F.2** Traces of Contributions to Reflectance and Transmittance by Beams that have been Scattered More Than Once

\[ \rho(\theta) = \frac{4r^2}{D_s^2 \cos \theta} \frac{1}{e_o} \frac{e_{st}(\theta)}{e_{st}(\theta)} \left[ \frac{e(\theta)}{e_{st}(\theta)} - \frac{e_w(\theta) \exp(-\tau_1)}{e_{wst}(\theta)} \right] \]  

(F-20)

Though as shown in Figure F.2 a scattered beam suffers double refraction and multiple reflection occurring at the glass-water and glass-air interfaces of the test cell, no changes are made in equation (F-20) Reflection and refraction effects due to the two-step change in refractive index at the boundaries are introduced into the theoretical solution as discussed in Section IV and in APPENDIX B.
The bidirectional intensity function $\rho(\theta)$ thus calculated is plotted versus $\sin^2\theta$. The area under a plot of either the bidirectional reflectance or transmittance yields the hemispherical values.

The bidirectional parameters were calculated for all experiments. All experimental values were presented in the results section. Some of the angular distribution data are plotted on the Results and Discussion Chapter. A representative portion of the remaining data is presented along with the Fortran Program in the following pages.
**Key to Fortran Program**

<table>
<thead>
<tr>
<th>Variable</th>
<th>Fortran Symbol</th>
</tr>
</thead>
<tbody>
<tr>
<td>d</td>
<td>D</td>
</tr>
<tr>
<td>λ</td>
<td>WAVEL</td>
</tr>
<tr>
<td>L</td>
<td>AL</td>
</tr>
<tr>
<td>PVC</td>
<td>PVC</td>
</tr>
<tr>
<td>$x_s$</td>
<td>QSCA</td>
</tr>
<tr>
<td>$e_{0\perp}$</td>
<td>EOO</td>
</tr>
<tr>
<td>$e_{0\parallel}$</td>
<td>E9090</td>
</tr>
<tr>
<td>$e_{st}(o)$</td>
<td>ESTDO</td>
</tr>
<tr>
<td>$e_{st}(90)$</td>
<td>ESTD90</td>
</tr>
<tr>
<td>$\tau_1$</td>
<td>TAUL</td>
</tr>
<tr>
<td>$\pi d/\lambda$</td>
<td>ALPHA</td>
</tr>
<tr>
<td>$e_\perp(\theta)$</td>
<td>VO(I)</td>
</tr>
<tr>
<td>$e_{\parallel}(\theta)$</td>
<td>V90(I)</td>
</tr>
<tr>
<td>$e_{\parallel}(\theta)$</td>
<td>ESTD(I)</td>
</tr>
<tr>
<td>$e_{\parallel}(\theta)$</td>
<td>EWO(I)</td>
</tr>
<tr>
<td>$e_{\parallel}(\theta)$</td>
<td>EW90(I)</td>
</tr>
<tr>
<td>$e_{\parallel}(\theta)$</td>
<td>EWSTD(I)</td>
</tr>
<tr>
<td>$\theta$</td>
<td>ANGLE</td>
</tr>
</tbody>
</table>

**Imput to the Program**

<table>
<thead>
<tr>
<th>Variable</th>
<th>Type</th>
</tr>
</thead>
<tbody>
<tr>
<td>ITEST</td>
<td>I3</td>
</tr>
<tr>
<td>D, WAVEL, AL, PVC, QSCA</td>
<td>SE8.4</td>
</tr>
<tr>
<td>Number of Angles (L)</td>
<td>I3</td>
</tr>
<tr>
<td>ANGLE, VO, V90, ESTD, EWO, EW90, EWSTD</td>
<td>7F10.3</td>
</tr>
</tbody>
</table>
...
### TABLE F.1

<table>
<thead>
<tr>
<th>EXPERIMENT</th>
<th>ANGLE</th>
<th>SLICE</th>
<th>PENETRATION</th>
<th>PENETRATION</th>
<th>PENETRATION</th>
</tr>
</thead>
<tbody>
<tr>
<td>C-0</td>
<td>0.244</td>
<td>C-0.20</td>
<td>0.037</td>
<td>0.037</td>
<td>0.037</td>
</tr>
<tr>
<td>C-0</td>
<td>0.244</td>
<td>C-0.20</td>
<td>0.037</td>
<td>0.037</td>
<td>0.037</td>
</tr>
<tr>
<td>C-1</td>
<td>0.244</td>
<td>C-0.20</td>
<td>0.037</td>
<td>0.037</td>
<td>0.037</td>
</tr>
<tr>
<td>C-0</td>
<td>0.244</td>
<td>C-0.20</td>
<td>0.037</td>
<td>0.037</td>
<td>0.037</td>
</tr>
<tr>
<td>C-1</td>
<td>0.244</td>
<td>C-0.20</td>
<td>0.037</td>
<td>0.037</td>
<td>0.037</td>
</tr>
</tbody>
</table>

**TOTAL**

<table>
<thead>
<tr>
<th>C-0</th>
<th>0.244</th>
<th>C-0.20</th>
<th>0.037</th>
<th>0.037</th>
<th>0.037</th>
</tr>
</thead>
</table>

### TABLE F.2

<table>
<thead>
<tr>
<th>EXPERIMENT</th>
<th>ANGLE</th>
<th>SLICE</th>
<th>PENETRATION</th>
<th>PENETRATION</th>
<th>PENETRATION</th>
</tr>
</thead>
<tbody>
<tr>
<td>C-0</td>
<td>0.244</td>
<td>C-0.20</td>
<td>0.037</td>
<td>0.037</td>
<td>0.037</td>
</tr>
<tr>
<td>C-1</td>
<td>0.244</td>
<td>C-0.20</td>
<td>0.037</td>
<td>0.037</td>
<td>0.037</td>
</tr>
<tr>
<td>C-0</td>
<td>0.244</td>
<td>C-0.20</td>
<td>0.037</td>
<td>0.037</td>
<td>0.037</td>
</tr>
<tr>
<td>C-1</td>
<td>0.244</td>
<td>C-0.20</td>
<td>0.037</td>
<td>0.037</td>
<td>0.037</td>
</tr>
<tr>
<td>C-0</td>
<td>0.244</td>
<td>C-0.20</td>
<td>0.037</td>
<td>0.037</td>
<td>0.037</td>
</tr>
<tr>
<td>C-1</td>
<td>0.244</td>
<td>C-0.20</td>
<td>0.037</td>
<td>0.037</td>
<td>0.037</td>
</tr>
</tbody>
</table>

**TOTAL**

<table>
<thead>
<tr>
<th>C-0</th>
<th>0.244</th>
<th>C-0.20</th>
<th>0.037</th>
<th>0.037</th>
<th>0.037</th>
</tr>
</thead>
</table>
### TABLE F.3

<table>
<thead>
<tr>
<th>EXPWAVELENGTH</th>
<th>ANGLE</th>
<th>SCALE</th>
<th>PER.</th>
<th>PAR.</th>
<th>TOTAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.24000000 C1</td>
<td>0.26000000 C1</td>
<td>0.28000000 C1</td>
<td>0.30000000 C1</td>
<td>0.32000000 C1</td>
<td>0.34000000 C1</td>
</tr>
<tr>
<td>0.26000000 C1</td>
<td>0.28000000 C1</td>
<td>0.30000000 C1</td>
<td>0.32000000 C1</td>
<td>0.34000000 C1</td>
<td></td>
</tr>
<tr>
<td>0.28000000 C1</td>
<td>0.30000000 C1</td>
<td>0.32000000 C1</td>
<td>0.34000000 C1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.30000000 C1</td>
<td>0.32000000 C1</td>
<td>0.34000000 C1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.32000000 C1</td>
<td>0.34000000 C1</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### TABLE F.4

<table>
<thead>
<tr>
<th>EXPWAVELENGTH</th>
<th>ANGLE</th>
<th>SCALE</th>
<th>PER.</th>
<th>PAR.</th>
<th>TOTAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.24000000 C1</td>
<td>0.26000000 C1</td>
<td>0.28000000 C1</td>
<td>0.30000000 C1</td>
<td>0.32000000 C1</td>
<td>0.34000000 C1</td>
</tr>
<tr>
<td>0.26000000 C1</td>
<td>0.28000000 C1</td>
<td>0.30000000 C1</td>
<td>0.32000000 C1</td>
<td>0.34000000 C1</td>
<td></td>
</tr>
<tr>
<td>0.28000000 C1</td>
<td>0.30000000 C1</td>
<td>0.32000000 C1</td>
<td>0.34000000 C1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.30000000 C1</td>
<td>0.32000000 C1</td>
<td>0.34000000 C1</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.32000000 C1</td>
<td>0.34000000 C1</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**Legend:**
- **PER.** Reflectance
- **PAR.** Transmittance
### Table F.5

<table>
<thead>
<tr>
<th>Exponential Angle Time (sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total</td>
</tr>
<tr>
<td>Exposure</td>
</tr>
<tr>
<td>Direct Reflectance</td>
</tr>
<tr>
<td>Diffuse Reflectance</td>
</tr>
<tr>
<td>Transmittance</td>
</tr>
<tr>
<td>Direct Reflectance / Transmittance</td>
</tr>
</tbody>
</table>

### Table F.6

<table>
<thead>
<tr>
<th>Exponential Angle Time (sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total</td>
</tr>
<tr>
<td>Exposure</td>
</tr>
<tr>
<td>Direct Reflectance</td>
</tr>
<tr>
<td>Diffuse Reflectance</td>
</tr>
<tr>
<td>Transmittance</td>
</tr>
<tr>
<td>Direct Reflectance / Transmittance</td>
</tr>
</tbody>
</table>

---

**Notes:**
- Exponential Angle Time (sec) columns represent different time intervals for each category (Total, Exposure, Direct Reflectance, Diffuse Reflectance, Transmittance, Direct Reflectance / Transmittance).
- The data entries are numerical values indicating the intensity or percentage for each category at each time interval.
TABLE F 7

<table>
<thead>
<tr>
<th>EXPERIMENT</th>
<th>ANGLE</th>
<th>SCALE FACTOR</th>
<th>PM</th>
<th>CD</th>
<th>TOTAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>G4</td>
<td>0.450</td>
<td>0.340</td>
<td>C1</td>
<td>C1</td>
<td>0.722</td>
</tr>
<tr>
<td>G5</td>
<td>0.450</td>
<td>0.400</td>
<td>C1</td>
<td>C1</td>
<td>0.722</td>
</tr>
<tr>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
</tr>
</tbody>
</table>

TABLE F 8

<table>
<thead>
<tr>
<th>EXPERIMENT</th>
<th>ANGLE</th>
<th>SCALE FACTOR</th>
<th>PM</th>
<th>CD</th>
<th>TOTAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>H1</td>
<td>0.200</td>
<td>0.340</td>
<td>C1</td>
<td>C1</td>
<td>0.722</td>
</tr>
<tr>
<td>H2</td>
<td>0.200</td>
<td>0.400</td>
<td>C1</td>
<td>C1</td>
<td>0.722</td>
</tr>
<tr>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
</tr>
</tbody>
</table>

SUM OF TOTAL REFLECTANCE ALL TRANSPARENCY = 0.640 CC
| TABLE F.9 |

<table>
<thead>
<tr>
<th>EXPERIM. ANGLE</th>
<th>WAVELENGTH</th>
<th>DIRECT REFLECTANCE</th>
<th>DIFFUSE TRANSMITTANCE</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.404E+05</td>
<td>0.0706E+00</td>
<td>0.3506E+00</td>
</tr>
<tr>
<td>0.05</td>
<td>0.454E+05</td>
<td>0.1224E+00</td>
<td>0.6124E+00</td>
</tr>
<tr>
<td>0.10</td>
<td>0.494E+05</td>
<td>0.1742E+00</td>
<td>0.8742E+00</td>
</tr>
<tr>
<td>0.15</td>
<td>0.534E+05</td>
<td>0.2260E+00</td>
<td>1.1360E+00</td>
</tr>
<tr>
<td>0.20</td>
<td>0.574E+05</td>
<td>0.2778E+00</td>
<td>1.3978E+00</td>
</tr>
<tr>
<td>0.25</td>
<td>0.614E+05</td>
<td>0.3296E+00</td>
<td>1.6596E+00</td>
</tr>
<tr>
<td>0.30</td>
<td>0.654E+05</td>
<td>0.3814E+00</td>
<td>1.9214E+00</td>
</tr>
<tr>
<td>0.35</td>
<td>0.694E+05</td>
<td>0.4332E+00</td>
<td>2.1832E+00</td>
</tr>
<tr>
<td>0.40</td>
<td>0.734E+05</td>
<td>0.4850E+00</td>
<td>2.4450E+00</td>
</tr>
<tr>
<td>0.45</td>
<td>0.774E+05</td>
<td>0.5368E+00</td>
<td>2.7078E+00</td>
</tr>
<tr>
<td>0.50</td>
<td>0.814E+05</td>
<td>0.5886E+00</td>
<td>2.9696E+00</td>
</tr>
</tbody>
</table>

**SUM OF TOTAL REFLECTANCE AND TRANSMITTANCE** = 1.000E+01

---

| TABLE F.10 |

<table>
<thead>
<tr>
<th>EXPERIM. ANGLE</th>
<th>WAVELENGTH</th>
<th>DIRECT REFLECTANCE</th>
<th>DIFFUSE TRANSMITTANCE</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.404E+05</td>
<td>0.0706E+00</td>
<td>0.3506E+00</td>
</tr>
<tr>
<td>0.05</td>
<td>0.454E+05</td>
<td>0.1224E+00</td>
<td>0.6124E+00</td>
</tr>
<tr>
<td>0.10</td>
<td>0.494E+05</td>
<td>0.1742E+00</td>
<td>0.8742E+00</td>
</tr>
<tr>
<td>0.15</td>
<td>0.534E+05</td>
<td>0.2260E+00</td>
<td>1.1360E+00</td>
</tr>
<tr>
<td>0.20</td>
<td>0.574E+05</td>
<td>0.2778E+00</td>
<td>1.3978E+00</td>
</tr>
<tr>
<td>0.25</td>
<td>0.614E+05</td>
<td>0.3296E+00</td>
<td>1.6596E+00</td>
</tr>
<tr>
<td>0.30</td>
<td>0.654E+05</td>
<td>0.3814E+00</td>
<td>1.9214E+00</td>
</tr>
<tr>
<td>0.35</td>
<td>0.694E+05</td>
<td>0.4332E+00</td>
<td>2.1832E+00</td>
</tr>
<tr>
<td>0.40</td>
<td>0.734E+05</td>
<td>0.4850E+00</td>
<td>2.4450E+00</td>
</tr>
<tr>
<td>0.45</td>
<td>0.774E+05</td>
<td>0.5368E+00</td>
<td>2.7078E+00</td>
</tr>
<tr>
<td>0.50</td>
<td>0.814E+05</td>
<td>0.5886E+00</td>
<td>2.9696E+00</td>
</tr>
</tbody>
</table>

**SUM OF TOTAL REFLECTANCE AND TRANSMITTANCE** = 1.000E+01

---

**DIFFUSE REFLECTANCE** = 0.484E+01 **DIFFUSE TRANSMITTANCE** = 0.516E+01

**SUM OF TOTAL REFLECTANCE AND TRANSMITTANCE** = 1.000E+01
### TABLE F.13

<table>
<thead>
<tr>
<th>EXPERI.</th>
<th>ANGLE SIZE</th>
<th>SLANT TYPE</th>
<th>FEP. COMPONENT</th>
<th>PAR. COMPONENT</th>
<th>TOTAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>C</td>
<td>0.12567101-C</td>
<td>0.12567101-C</td>
<td>0.12567101-C</td>
<td></td>
</tr>
<tr>
<td>c.100C0C0C C1</td>
<td>C.100C0C0C C2</td>
<td>G.100C0C0C C3</td>
<td>G.100C0C0C C4</td>
<td>G.100C0C0C C5</td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>C</td>
<td>0.12567101-C</td>
<td>0.12567101-C</td>
<td>0.12567101-C</td>
<td></td>
</tr>
<tr>
<td>c.100C0C0C C1</td>
<td>C.100C0C0C C2</td>
<td>G.100C0C0C C3</td>
<td>G.100C0C0C C4</td>
<td>G.100C0C0C C5</td>
<td></td>
</tr>
</tbody>
</table>

### TABLE F.14

<table>
<thead>
<tr>
<th>EXPERI.</th>
<th>ANGLE SIZE</th>
<th>SLANT TYPE</th>
<th>FEP. COMPONENT</th>
<th>PAR. COMPONENT</th>
<th>TOTAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>C</td>
<td>C</td>
<td>0.12567101-C</td>
<td>0.12567101-C</td>
<td>0.12567101-C</td>
<td></td>
</tr>
<tr>
<td>c.100C0C0C C1</td>
<td>C.100C0C0C C2</td>
<td>G.100C0C0C C3</td>
<td>G.100C0C0C C4</td>
<td>G.100C0C0C C5</td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>C</td>
<td>0.12567101-C</td>
<td>0.12567101-C</td>
<td>0.12567101-C</td>
<td></td>
</tr>
<tr>
<td>c.100C0C0C C1</td>
<td>C.100C0C0C C2</td>
<td>G.100C0C0C C3</td>
<td>G.100C0C0C C4</td>
<td>G.100C0C0C C5</td>
<td></td>
</tr>
</tbody>
</table>
### TABLE F 17

<table>
<thead>
<tr>
<th>EXPERIMENTAL ANGLE SINE SQUARE THETE</th>
<th>PER-COMPONENT</th>
<th>PAR-COMPONENT</th>
<th>TOTAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>0.01000000 01</td>
<td>0.7961240-07</td>
<td>0.5952380-01</td>
<td>0.485400</td>
</tr>
<tr>
<td>0.02000000 02</td>
<td>0.3151564-01</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.03000000 03</td>
<td>0.6668730-01</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.04000000 04</td>
<td>0.2107107-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.05000000 05</td>
<td>0.1289899-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.06000000 06</td>
<td>0.1317990-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.07000000 07</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.08000000 08</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.09000000 09</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.10000000 10</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.11000000 11</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.12000000 12</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.13000000 13</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.14000000 14</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.15000000 15</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.16000000 16</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.17000000 17</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.18000000 18</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.19000000 19</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
</tbody>
</table>

### TABLE F 18

<table>
<thead>
<tr>
<th>EXPERIMENTAL ANGLE SINE SQUARE THETE</th>
<th>PER-COMPONENT</th>
<th>PAR-COMPONENT</th>
<th>TOTAL</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
</tr>
<tr>
<td>0.01000000 01</td>
<td>0.7961240-07</td>
<td>0.5952380-01</td>
<td>0.485400</td>
</tr>
<tr>
<td>0.02000000 02</td>
<td>0.3151564-01</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.03000000 03</td>
<td>0.6668730-01</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.04000000 04</td>
<td>0.2107107-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.05000000 05</td>
<td>0.1289899-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.06000000 06</td>
<td>0.1317990-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.07000000 07</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.08000000 08</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.09000000 09</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.10000000 10</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.11000000 11</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.12000000 12</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.13000000 13</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.14000000 14</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.15000000 15</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.16000000 16</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.17000000 17</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.18000000 18</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
<tr>
<td>0.19000000 19</td>
<td>0.4000000-00</td>
<td>0.5829071-01</td>
<td>0.477294</td>
</tr>
</tbody>
</table>

### Additional Information
- **TABLE F 17**: This table lists the experimental setup for measuring reflectance and transmittance. The columns include the experimental angle sine square, the per-component, and the par-component with a total value calculated for each entry.
- **TABLE F 18**: Similarly, this table provides the per-component and par-component data for the same experimental setup, with a total value calculated for each entry.
- **General Notes**: Both tables are used to test the reflectance and transmittance properties at different angles, with the sum of total reflectance and transmittance being a constant value.

### Summary
- The sum of total reflectance and transmittance is 0.963800.
- Additional notes for the tables include the direct reflectance and transmittance, as well as the diffuse reflectance and transmittance, which are calculated and presented in the tables.
APPENDIX G

Errors in Multiple Scatter Experiments

The errors introduced in a light scattering experiment largely depend on the arrangement used to measure the scattered intensities. In the present study the arrangement used consisted of a narrow incident beam which illuminated a small portion of the scattering sample (the diameter of the incident beam at the center of the system was 0.12 cms). The field of view of the detector was large enough that for any angle most of the scattered radiation was captured. A small amount of radiation is inevitably scattered outside the detector field of view. Since the area viewed at the center of the system must be large a relatively large circular aperture (0.2 cms) was placed in front of the detector. The sensitivity of the photomultiplier is non-uniform over the exposed area of 0.2 cms and it is expected that errors might be introduced in the experiments if the radiation measured at different angles fell on different parts of the detector.

Another source of error has been described by Theissing and is due to the finite dimensions of the scattering volume. Theissing has noted that whenever we observe light scattered from a finite volume of a unidimensional system a portion is lost in the surroundings along the equatorial section of the cell. Theissing has derived the following equation for the error due to equatorial losses:
Error = Ω·p(90°)·e^{−τ_1/2} \quad (G-1)

where Ω is the solid angle subtended by the equatorial boundaries and p(90°) is the value of the phase function at 90°.

Since the optical thickness τ_1 in most of the experiments in this study was very large the term e^{−τ_1/2} dominated in equation (G-1) and consequently no error was introduced. However, when τ_1<1.0 the error may become appreciable.

Losses due to the finite dimensions of the scattering cell may be also introduced as a result of the total reflection. In the theoretical analysis it is assumed that totally reflected light eventually escapes in the surroundings in the form of diffuse radiation. However, when the dimensions of the cell are finite and the system dilute enough the totally reflected light might reach the boundaries of the cell before it becomes diffusely scattered. As a consequence of this phenomenon it is appropriate to substitute the solid angle within which total internal reflection occurs for Ω in eq. (G-1). The consequence of increased edge losses is the reduction in intensity at other angles. The solid angle subtended by the cell edge is also a factor. This is shown in Figure G.1 where the theoretical bidirectional intensity function is compared
with experimental results for two values of L (slab thickness). It is observed that better agreement is obtained for the smaller L, for which the solid angle subtended by the edge is smaller. Discrepancies due to this cause were observed in Figures 7.2 to 7.4 which presented results for relatively small values of the optical thickness.

The errors due to all reasons stated above can be evaluated easily in this study because the lattices used were non-absorbing. Conservation requires that the sum of the reflectance and transmittance should equal one. Tables 6.1 to 6.8 show no large discrepancies. The average sum of the reflectance and transmittance was found to be 1.003 and the standard deviation 0.0613. The largest deviation observed was ± 0.16.

The experimental data after being normalized so that the sum equals one were compared to the theoretical values. A case of comparison is shown in Table G.1 where it can be seen that the discrepancies are not appreciable. The fractional error defined as (Theoretical value - Experimental) x 100 / Theoretical was found to be larger in case of small optical thicknesses (15% for reflectance, 4% for transmittance). The mean fractional error observed for all runs tabulated in Table G.1 was ± 5% for the transmittance and ± 2% for the reflectance.
TABLE G.1

A comparison of the theoretical and experimental results for a particle size $0.530\mu$ and a wavelength $0.4358\mu$.

<table>
<thead>
<tr>
<th>Optical Thickness</th>
<th>Calculated Diffuse Transmittance</th>
<th>Error</th>
<th>Calculated Diffuse Reflectance</th>
<th>Error</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2667</td>
<td>0.2026</td>
<td>0.00800</td>
<td>0.02469</td>
<td>-0.00365</td>
</tr>
<tr>
<td>1.091</td>
<td>0.566</td>
<td>0.02000</td>
<td>0.07894</td>
<td>-0.01034</td>
</tr>
<tr>
<td>37.36</td>
<td>0.3576</td>
<td>0.00210</td>
<td>0.59900</td>
<td>-0.00190</td>
</tr>
<tr>
<td>215.9</td>
<td>0.1014</td>
<td>-0.00748</td>
<td>0.85550</td>
<td>0.00750</td>
</tr>
<tr>
<td>252.3</td>
<td>0.08835</td>
<td>-0.00289</td>
<td>0.86640</td>
<td>0.00500</td>
</tr>
<tr>
<td>577.7</td>
<td>0.04120</td>
<td>0.00470</td>
<td>0.91570</td>
<td>-0.00460</td>
</tr>
<tr>
<td>713.9</td>
<td>0.03363</td>
<td>-0.00209</td>
<td>0.92330</td>
<td>0.00210</td>
</tr>
<tr>
<td>775.5</td>
<td>0.03104</td>
<td>-0.00275</td>
<td>0.92590</td>
<td>0.00270</td>
</tr>
<tr>
<td>983.</td>
<td>0.02467</td>
<td>-0.00010</td>
<td>0.93220</td>
<td>0.00030</td>
</tr>
<tr>
<td>1009.</td>
<td>0.02405</td>
<td>-0.00062</td>
<td>0.93290</td>
<td>0.00030</td>
</tr>
<tr>
<td>1168.</td>
<td>0.02085</td>
<td>-0.00158</td>
<td>0.93560</td>
<td>0.00210</td>
</tr>
<tr>
<td>1632.</td>
<td>0.01502</td>
<td>-0.00115</td>
<td>0.94190</td>
<td>0.00120</td>
</tr>
<tr>
<td>2327.</td>
<td>0.01059</td>
<td>-0.00011</td>
<td>0.94640</td>
<td>0.00010</td>
</tr>
</tbody>
</table>

Error = Measured Value - Calculated Value
Figure G.1 Bidirectional reflectance and transmittance of polystyrene latex spheres; upper curves: $L=0.0508\text{cm}$; lower curves: $L=0.1473\text{ cms}$. Data points represent experimental results, solid lines theory.
REFERENCES


The author was born November 30, 1938 in Apollonia Sifnos, Greece. He received his elementary education in the school of Apollonia and then moved to Athens where he attended high school. He graduated in June 1958 from the 13th high school for boys located in Athens.

In September 1958 the author entered the Technical University of Athens and he was awarded a Diploma in Chemical Engineering in June 1963. He served in the Greek Air Force from July 15, 1963 to October 15, 1965. During his service he was attached to the Nuclear Research Center Democritos for a period of 18 months working as a research assistant in the Physics Laboratory. In October of 1965 the author was accepted by McGill University where he did graduate work until June 1966. The same month the author was accepted at M.I.T. where he received the degree Master of Science in Chemical Engineering in August 1967. In October 1967 the author began his doctoral thesis. During that period he held various research assistantships and fellowships.

The author has accepted full-time employment with the American Oil Company, Research and Development Department.