THE SCATTERING OF LIGHT BY SEAWATER

EXPERIMENTAL RESULTS AND THEORETICAL APPROACH

by

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Translated by:

George Halikas
University of California, San Diego
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La Jolla, California 92037
(August 1975)
Preface to the present English version

The material of this paper was originally published in French. It was orally presented at the Scripps Institution of Oceanography in 1973, during a stay at the Visibility Laboratory at the invitation of S. Q. Duntley, J. E. Tyler, R. C. Smith and R. W. Austin. The scientists at the « VisLab », and particularly Ros Austin, suggested and then decided that an English translation of this work should be made. George Halikas, a Greek student knowledgeable in French and English, was in charge of making this translation, which represented a considerable effort. In his handwritten letter of January, 25, 1976, he informed me that he did not try to polish the language in this first step, but rather strived to remain close to the original by making a word for word translation. Any reader should understand this background, which makes the style somewhat difficult to read. I am sure that George certainly had a hard time in transforming the long, convoluted, French sentences, with subjunctives scattered here and there, and intricate subordinate clauses. However, he did very well, and I’m deeply grateful to him for the tremendous job that he achieved at that time.

He proposed, as a second step, to assist me in producing a « more English » version, although in his letter, he added a sentence “although as time progresses, it appears increasingly doubtful ». I did not understand this well at the time, and found out that unfortunately, he died a short time later.

I am pleased, 25 years later, to have this opportunity of recalling his memory, and to see his effort acknowledged and hopefully used by the broader scientific community.

André Morel

20 Novembre 2001
TRANSLATORS NOTE

I have attempted herein to remain as close to the original French wording as possible, which in several cases resulted in awkward English. I hope that in cases of uncertainties in meaning, the reader will refer to the original for clarification.

I have taken into account the page with Errata supplied with the original paper, and I take this opportunity to point out some additional misprints appearing in the original, which were discovered during the translation.

Pagination follows both the original numbers and the present ones for easy reference:

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| 3.1-60       | 133              | 18 "V(m^3/m^3)" instead of "V(m^2/m^3)"
| 3.1-63       | 137              | "bottom "dI(\theta)" instead of "di(\theta)"
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| 3.1-64       | 141              | 14 "\int P(\theta) d\theta = 4\pi" instead of \( \int P(\theta) = 4\pi \) " \( \int \) " |
| 3.1-64       | 141              | 19 "\bar{\beta}_2(\theta) = \frac{i2}{\pi \Omega \alpha^2}" instead of \( \bar{\beta}_2(\theta) = \frac{i2}{\eta \Omega \alpha^2} \) " \( \frac{2}{2} \) " |
| 3.1-66       | 144              | 8 "(Sec. 2.1 and 3.2, ...)" instead of "(Sec. 5 and 1.7 ...)"

In addition I caught three of my own typographical errors, namely:

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| 3.1-15       | 26               | "Beardsley" instead of "Beardley"
| 3.1-39       | 80               | "two points" instead of "two point"
| 3.1-40       | 84               | "on the one hand" instead of "on the one had"
INTRODUCTION

Speaking in terms which appear sometimes speculative, the study of light scattering by seawater has always had and constantly has, practical motivations. This phenomenon completely reflects a characteristic of seawater, which is essential from the point of view of sedimentology, or geochemistry, or even biology, to know the concentration of particulate matter in suspension. Scattering is in fact a spatial redistribution of energy which does not take place at random, but on the contrary according to a law which is accurately described "scattering function". Its determination, together with its theoretical interpretation, permits the hope of obtaining information on the particles and their properties, which would complement that obtained, sometimes with greater difficulty, by direct observation.

In addition, all the problems of optical oceanography, those of propagation or more generally of radiative transfer, such as those of contrast reduction, i.e. of imagery, whether attacked experimentally or theoretically, imply the knowledge of the volume scattering function. In some cases which are difficult to access through simple experimentation, such as backscattering or scattering in the neighborhood of the propagation direction, theory may provide an equivalent or even better route.

The text that follows is divided into three principal parts. The first treats only the experimental facts, while the second indicates how the theory can be utilized and what the expressions are. Finally the third part considers both the experimental results and those derived from theory, in a research directed towards interpretation and applications.

At the end of the text itself, the definitions and certain numerical aspects relating to the computations on a computer, form the subject of two appendices. In addition, in order not to overextend the bibliography, references to original works relating to the theories of Rayleigh, Rayleigh-Gans and Mie have been omitted. All these references are practically replaced by a single one: that of the by now classical work of Van deHulst (1957) which includes them all.

NOTE: The figures are numbered with two numbers, the first being I, II or III corresponding to Part I, Part II or Part III. The equations are not numbered, except in Part II, also with two numbers, being 1, 2 or 3 according to whether they appear in sections 1, 2 or 3 of this Part.
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PART I

PREFACE

The propagation of light in all media other than the vacuum is accompanied by two phenomena which determine the reduction of flux: absorption and scattering. The absorbed energy is transformed into heat, being eventually partially re-emitted as fluorescence through the Raman effect but with a change in wavelength. On the contrary scattered energy is not transformed but simply dispersed spatially. In the case of an "optically pure" medium it is the molecules themselves and only these which produce the scattering. In the case of "turbid" media, in addition to molecular scattering there occurs scattering by suspended particles. To visible light, the purest sea water behaves like an optically turbid medium, therefore for any question regarding the total scattering coefficient, the volume scattering function, the shape of the scattering function, the (wavelength) selectivity or even the polarization, it is necessary to examine the respective roles of molecular scattering and particle scattering.

All the definitions used are collected in Appendix 1.

1. SCATTERING FUNCTION OF SEAWATER

1.1 GENERAL SHAPE.

During the same year (1963) N. G. Jerlov and S. Q. Duntley independently made a systematic revue of all measurements of the volume scattering function performed up to that time. The results presented by these authors have been obtained with different instruments, some taken in situ, others requiring removal and transfusion of samples, some using white light and others filtered light (blue, blue-green, green, yellow). In addition, these results involve different types of water with respect to turbidity and origin (Pacific Ocean, Atlantic Ocean, English Channel, China Sea and even lakes). During this comparison, N. G. Jerlov and S. Q. Duntley, in arriving at identical conclusions, underline the remarkable fact illustrated in Figure II. All measurements agree for angles less than 90°, demonstrating that the shape of the scattering function varies only slightly from one water to another, while in all cases demonstrating a very pronounced asymmetry.
Figure I.1: Scattering functions obtained by various authors and normalized at $90^\circ$.

$H$: E. O. Hulburt (1945) Chesapeake Bay;
$K$: M. Kozljaninov (1957) China Sea;
$S$: T. Sasaki (1960) of Japan;
$T_1/T_2$: J. E. Tyler (1961) Pacific (San Diego);
$D$: S. Q. Duntley (1963) Lake Winnipesakee;
$J$: N.G. Jerlov (1961) Atlantic (Madeira)

For angles greater than $90^\circ$ a much greater scattering of the curves occurs. The angle at which minimum scattering occurs also varies between approximately $100^\circ$ and $130^\circ$. This minimum being more or less accentuated.

It must be noted that the measurements of Pickard and Giovando (1960) on rather special waters (fjord in British Columbia), are markedly different from the group, the increase at small angles being more pronounced. The measurements by Sasaki, et.al. (1960) are in a way also different from the others indicating small oscillations. Mankovsky et.al. (1970) have also produced such peculiarities. However in a
general sense, all subsequent studies have always confirmed the major trends which can be discerned by examining these initial measurements.

1.2 POSSIBLE VARIATIONS AT MEDIUM ANGLES (θ > 30°).

In reality, numerous studies performed on waters of considerable variety from the point of view of their turbidity, demonstrate that the shape of the scattering function does vary noticeably in the region of medium angles. Figure I.2 provides an example (the curves are normalized at 90° in order to demonstrate variations in their shape without including variations in the magnitude of the scattering function). Without increasing the examples, Figure I.2 summarizes in a way the variations which the scattering function may undergo in this angular domain. Curve 3 has the most apparent symmetric shape

![Figure I.2: Scattering functions normalized at 90°.](image)

1) English Channel (surface, Roscoff, 6/29/63, b=0.6m⁻¹, λ=546 nm)
2) Mediterranean (surface, bay of Villefranche, 11/16/63, b=0.1m⁻¹, λ=546nm).
3) Mediterranean (1500 meters, Trrhenian Sea, 7/21/64, b=0.015m⁻¹, λ=546nm). (also Mixel, 1965)
4) Baltic (5 meters, station 2, 6/13/67, b=0.76m⁻¹, λ=525nm, G. Kullenberg, 1967).
5) Atlantic (1500 meters, Tongue of the Ocean, 7/13/71, b=0.037m⁻¹, λ=510 nm, T. J. Petzold, 1972).

Full and open circles, respectively station 1 and 2 (T₁ and T₂ from preceding figure) (Pacific, J. E. Tyler 1961).

and corresponds to the case of very clear deep water (identical slopes have been obtained at 1000 m or 2000 m in the Eastern Mediterranean - lab. group 1969 - as well as in the region of Madeira),
curve 2 is an intermediate curve frequently observed for ocean surface "blue" water, curve 1 is finally that which predominates for numerous surface waters and coastal waters. The character of the asymmetry is slightly enhanced for Baltic waters (station 2, Kullenberg (1967), \( b_{525} = 0.76 \text{ m}^{-1} \)). Curve 5 taken from the measurements of T. J. Petzold (1972) lies between cases 2 and 3.

The systematic study of the shape of the scattering function leads to the following statement: between the extreme cases presented, i.e., from the most asymmetric form of curve 1 to the most symmetric one of curve 3, the various intermediate forms are not positioned in a random fashion. On the contrary the tendency towards symmetry occurs gradually, as the water becomes less scattering and in addition, in terms of absolute value, all the scattering functions spread. With a good approximation, and considering the experimental uncertainty, this orderly behavior of the volume scattering function has been verified on more than 150 samples taken from very different areas (English Channel, Atlantic, Mediterranean, Indian Ocean). The most noticeable irregularities, which certainly occur, concern backscattering almost exclusively (\( \theta > 120^\circ \) to be more precise). This progressive change is equally well shown (Fig. I.3)

Figure I.3: Ratio \( \beta(45^\circ)/\beta(135^\circ) \)

characterizing the asymmetry of the scattering function of sea water, as a function of \( \beta(90^\circ) \) for various sea waters \( (\lambda = 546 \text{ nm}) \). From the expression relating \( \beta(\theta) \) to \( \beta(90^\circ) \) it is easily deduced that this ratio may be expressed by

\[
\frac{\beta(45)}{\beta(135)} = \frac{R(45)(\beta(90)) + c}{R(135)(\beta(90)) + c}
\]

and varies with \( \beta(90) \) according to a hyperbolic law. Two hyperbolas are drawn with medium values (upper curve) or extremes (lower curve) attributed to \( R(45) \) and \( R(135) \). Note: \( c = \text{constant} \).
by the almost regular variation of the ratio of asymmetry $\beta(45^\circ)/\beta(135^\circ)$. This ratio decreases from a value of about 12 to reach values as low as 2 following the change from the most scattering waters towards the clearest. In Fig. I.3 and in all that follow the unit of the angular volume scattering function is $m^{-1}$ and not $m^{-1}$ steradian$^{-1}$ as is usual (which will be implied and is omitted for convenience).

Since the turbid waters indicate scattering functions quite similar and strongly asymmetric, and only the "clear" waters have more variable and more symmetric shapes, it is logical to attribute this modification at least partially to the more or less important function of molecular scattering. Therefore, the latter effect must be known, in order to subsequently study the residual variations which may then be strictly attributed to particles.

This point may be simply studied, without making it necessary to know the characteristic values of molecular scattering (A. Morel 1965). Using the subscripts $p$ and $o$ for the portion attributed to the particles and to the water itself, respectively, the angular volume scattering coefficient may be decomposed at angles $\theta$ and $90^\circ$ as:

$$\beta(\theta) = \beta_p(\theta) + \beta_o(\theta)$$

$$\beta(90) = \beta_p(90) + \beta_o(90)$$

and formulate the relation

$$\beta(\theta) - \beta_o(\theta) = \frac{\beta_p(\theta)}{\beta_p(90)} \left[ \beta(90) - \beta_o(90) \right]$$

If one obtains $\beta(\theta)$ as a function of $\beta(90)$ for several samples it is determined that the distribution of experimental points behaves roughly in a linear manner. This indicates that as a first approximation the
ratio $\beta_p(\theta)/\beta_p(90) = R(\theta)$ is quite constant and is characteristic of the scattering due to particles only. It should be added that this straight line, if the linearity does not change for waters that are only very slightly scattering, must pass through the assumed point for water which is optically pure, with coordinates $\beta_o(\theta) - \beta_o(90)$ (thus the value of $\beta_o(90)$ can be directly and approximately estimated). Figures I.4 show examples of this procedure.

Figure I.4: Points, representative of various samples, obtained by plotting $\beta(\theta)$ against $\beta(90)$ for wavelengths 436 and 366 nm, and two angles $\theta$, 45° and 135°. These figures complement those presented previously for $\lambda = 546$ nm (A. Morel, 1965). They refer to various sea waters: 7 from the English Channel, 15 from the Mediterranean (both east and central), 5 from the Atlantic (region of Madeira) 9 from the Indian Ocean (region of Madagascar). The open circles refer to optically pure water (cf. Section 2.1).
The regression calculation* shows that the correlation coefficient which is high for small angles (greater than 0.96) drops considerably for the large angles (0.86 at 150°) demonstrating that the scattering function, not of sea water (water with particles) but of the particles alone, possesses an important variability. On the contrary the values of mean slope, i.e., the various values of $R(\theta)$, are not significantly different for the three wavelengths used. Plotting $R(\theta)$ as a function of $\theta$ one obtains the "mean" scattering function for the particles alone. (See Figs. I.6, I.9 and III.7 of the 3rd Section). It is towards this form of maximum asymmetry that the scattering functions of turbid waters tend; the examples taken from V. W. Reese and S. P. Tucker (1970) are a good illustration of this point of view. (Fig. I.6). Two remarks are in order concerning this curve: as regards the mean curve, it only very imperfectly describes certain particular cases, and in addition this mean curve in a way favors the particles present in turbid waters. (In effect the computed ratios $R(\theta)$ are highly dependent on points that are farthest away corresponding to the most scattering waters.) These points will be reexamined further below (Sec. 3.1).

*Performed for a maximum of 112 values at $\lambda=546$ and a maximum of 26 values for $\lambda=436$ and 366 nm (on 36 for the particular angles 45° and 135°). Certain points depart very noticeably from the linear law. Most often, but not necessarily, these correspond to coastal water samples carrying terrigenous particles. A systematic study of these departures remains to be done.
Figure I.6: Scattering functions determined by J. W. Reese and S. P. Tucker (1970) in San Diego Bay between 1 and 15 meters, on 6/30/1967, the attenuation coefficient $C$ being greater than $1.70 \text{ m}^{-1}$; the absolute values are read on the left hand scale. With the scale on the right, the scattering functions normalized at 90°, (A. Morel 1965) with the full line; with the broken and dotted line particle scattering functions measured by T. J. Petzold (1972) for the same two stations as previously, respectively station 8 (Atlantic) and 5 (Pacific).

1.3 SMALL ANGLE CASE. ($\theta<30^\circ$)

Considering the very high values of the scattering function in this angular region, (Fig. I.1), molecular scattering has only a negligible role (in effect it does not vary with angle by more than a factor of 2, see later Section 2.1). Figure I.5 collects several examples of the scattering function in the region of small angles. Two among these are mean curves, that obtained by D. Bauer and A. Morel (1967) from 58 measurements in the English Channel and the Mediterranean (at a wavelength of 550 nm), and that by F. Nyffeler (1969-1970) calculated from 66 measurements in the Mediterranean (in situ at wavelengths 455, 559 and 599 nm with the same instrument and in vitro at wavelengths 488.0 and 632.8 nm). In a general way it may be stated that the vari-
Figure I.5: Scattering functions at small angles (the two scales are logarithmic). The positioning of the curves relative to each other is arbitrary:
1: S.Q. Duntley (1963);
2: Y.E. Ochakovskii (1965);
3: M. Kozlianinov (1957);
4: G. Kullenberg (1968);
5: D. Bauer, A. Morel (1967)
6: F. Nyffeler (1970);
7 & 8: T. J. Petzold (1972), station 5 (Pacific) and station 8 (Atlantic) respectively. i.e. with respect to the most scattering water (b=0.27 m$^{-1}$) and the most clear (b=0.037 m$^{-1}$) presented by this author.

ability of this portion of the scattering function is not large (cf. references below) and also that the influence of wavelength which is not obvious, is small if it exists at all. The agreement among the various measurements is satisfactory, and more so considering that in such delicate measurements, systematic errors and others accompanying the experimental procedure used are inevitable. Also the slight curvature and decrease of slope around 30°-10° which some of the curves show appear to reflect such an instrumental effect. The recent measurements of R. Morrison (1970) and T. J. Petzold (1972) taken with the instrument of the Visibility Laboratory (Scripps Institution of Oceanography) demon-
strate that the increase of $\beta(\theta)$, approximately according to a $\theta^{-1.6}$ law between $10^o$ and $1^o$, continues for angles less than $1^o$. The remarkable fact that these recent measurements demonstrate is exactly this rapid increase, the volume scattering function growing by about 2 orders of magnitude between $1^o$ and $0.1^o$. As will be seen later (Part III, section, Section 3.1.3) the value itself does not appear to be truly independent of the scale on which it is drawn (or if one desires on the scattering volume considered, which according to its cross-section permits one to consider or not a particle as a scattering center - cf. 3.1.1, 3.1.3, Part III).
2.0 ROLE OF MOLECULAR SCATTERING.

2.1 COLLECTION OF USEFUL VALUES FOR WATER AND SEA-WATER.

Measurements performed on optically purified sea-water, through repeated filtration, have furnished values of the molecular scattering function. Confirmation of these values has been sought, by conducting similar measurements on artificial sea-water, on sodium chloride solutions (which are easier purified than natural sea-water) and also on pure water obtained by evaporation under vacuum. (A. Morel 1966, 1968a). The scattering functions of these different solutions and of water itself are effectively absolutely symmetric (which can be used as a criterion of optical purity) and obey Rayleigh's theoretical law - effectively the modified law which takes into account the molecular anisotropy of water, producing a partial depolarization:

\[ \beta_o(\theta) = \beta_o(90^\circ)(1 + p_{90}\cos^2 \theta) \]

\( p_{90} \) being the degree of polarization at 90° whose experimental value is 0.84 corresponding to 0.09 for the polarization defect \( \delta \) (\( \delta \) is the ratio \( (1-p)/(1+p) \)). In the case of water the absolute values of \( \beta_o(90) \) obtained at 5 wavelengths are in good agreement with a certain number of other experimental investigations (from 546 to 436 nm) and also with those calculated from the theory of fluctuations in density. A detailed revue of these questions has been recently made (A. Morel, 1973b) from which the following table has been taken giving at 25 nm steps the theoretical values of \( \beta_o(90) \) and \( b_o \) (the total scattering coefficient) for optically pure water and sea-water.

It should be noted that if a power law is sought for the molecular scattering of water (or of sea-water) the best expression is \( \lambda^{-4.32} \). The fact that the exponent differs from 4 essentially proves the dispersion of refractive index.
<table>
<thead>
<tr>
<th></th>
<th>λ (nm)</th>
<th>350</th>
<th>375</th>
<th>400</th>
<th>425</th>
<th>450</th>
<th>475</th>
<th>500</th>
<th>525</th>
<th>550</th>
<th>575</th>
<th>600</th>
</tr>
</thead>
<tbody>
<tr>
<td>water</td>
<td>6.47</td>
<td>4.80</td>
<td>3.63</td>
<td>2.80</td>
<td>2.18</td>
<td>1.73</td>
<td>1.38</td>
<td>1.12</td>
<td>0.93</td>
<td>0.78</td>
<td>0.68</td>
<td></td>
</tr>
<tr>
<td></td>
<td>103.5</td>
<td>76.8</td>
<td>58.1</td>
<td>44.7</td>
<td>34.9</td>
<td>27.6</td>
<td>22.2</td>
<td>17.9</td>
<td>14.9</td>
<td>12.5</td>
<td>10.9</td>
<td></td>
</tr>
<tr>
<td>sea-water</td>
<td>8.41</td>
<td>6.24</td>
<td>4.72</td>
<td>3.63</td>
<td>2.84</td>
<td>2.25</td>
<td>1.80</td>
<td>1.46</td>
<td>1.21</td>
<td>1.01</td>
<td>0.88</td>
<td></td>
</tr>
<tr>
<td>s=35/38f_{oo}</td>
<td>134.5</td>
<td>99.8</td>
<td>75.5</td>
<td>58.1</td>
<td>45.4</td>
<td>38.9</td>
<td>28.8</td>
<td>23.3</td>
<td>19.3</td>
<td>16.2</td>
<td>14.1</td>
<td></td>
</tr>
</tbody>
</table>

Table I: Theoretical values of β₀(90) and of b₀ as a function of wavelength. The other angular coefficients \( \beta(\theta) \) are deduced from \( \beta₀(90) \) by means of the following expression. The integration over all space of this same expression,

\[
B = \frac{8\pi}{3} \beta₀(90) \frac{2+\delta}{1+\delta}
\]

permits calculation of \( b₀ \) (\( \delta \) is taken equal to 0.09).

Scattering is higher in the case of solutions by an additive term explained by fluctuations in concentration. This term may be calculated at least for ideal dilute solutions from the molecular mass of the solute and the increase in index which its presence entails. Therefore the measurements on sodium chloride solutions must be interpreted so as to produce a comparison term for the relative values of sea-water. Sea-water demonstrated values by 6% higher than sodium chloride solution of the same concentration in chloride ions, and by 30% higher than those of water.

2.2 RELATIVE IMPORTANCE OF MOLECULAR SCATTERING.

Given the accentuated asymmetry of the particle scattering function, the molecular scattering function is expected to play a non-negligible role in the backward direction, particularly where the scattering function demonstrates a minimum, i.e., at about 120° to 140°. Whenever the waters are turbid the relative importance of molecular scattering is evidently negligible at all angles; on the other hand for clear waters it is perhaps interesting to determine its maximum influence (at least from the existing measurements, not excluding the possibility of existence of even clearer waters).
Figure I. 7: Scattering functions normalized at 90°. Measurements made at three wavelengths on a turbid sea water sample (English Channel) and on another very clear one (Tyrrhenian Sea).

Curve 3 of Figure I. 2 or curves 1, 2, 3 of Figure I. 7 are representative examples of deep clear waters. Thus the ratio of \( \beta_o(\theta)/\beta(\theta) \) of molecular scattering to total scattering observed for one of these typical cases (that of Fig. I. 7) must be formed, where the scattering function has been determined at three wavelengths.

*To obtain the ratio the experimental values on purified sea-water have been used (A. Morel 1968) which are slightly lower (about 10%) rather than the theoretical values of table I. This is judged as preferable since the measurements on both the purified water and the sample have been made using the same instrument. The uncertainty on the absolute values disappears through the ratio.
The ratios in Figure I.8 are obtained in this way, plotted against scattering angle. Due to the pronounced selectivity of molecular scattering, its relative importance increases with shorter wavelengths, and in all cases it is effectively maximal for angles between 90° and 140°. The total molecular scattering coefficient $b_0$ must also be compared to the coefficient $b$ of the sample (calculated through $\beta(30^\circ)$, cf. Section 6).

**Figure I.8**: Relative importance of molecular scattering as a function of angle. The curves for the three wavelengths refer to the Tyrrhenian Sea sample (cf. Fig. I.7). The open circles are deduced from the measurements of G. Kullenberg (1968) in the Sargasso Sea (10 meters) at 460 nm wavelength.

<table>
<thead>
<tr>
<th>$\lambda$ (nm)</th>
<th>$b$ $10^{-3}$m$^{-1}$</th>
<th>$b_0/b$ %</th>
</tr>
</thead>
<tbody>
<tr>
<td>546</td>
<td>1.6</td>
<td>10</td>
</tr>
<tr>
<td>436</td>
<td>2.5</td>
<td>18</td>
</tr>
<tr>
<td>366</td>
<td>3.1</td>
<td>30</td>
</tr>
<tr>
<td>o 460</td>
<td>2.5</td>
<td>14</td>
</tr>
</tbody>
</table>

The ratio $b_0/b$ increases in the case of this very pure water by 10% for $\lambda = 546$ nm and by 30% for $\lambda = 366$ nm. It should be added that similar results have also been obtained in the eastern Mediterranean (Lab. group 1969) and in the Atlantic in the vicinity of Madeira, generally at depths greater than 1000 meters, while from the measurements of G. Kullenberg (1968) at 10 meters only, but in the Sargasso Sea, similar values can be deduced, proving once more the great clarity of this sea.
3.0 SCATTERING FUNCTION OF THE PARTICLES.

3.1 RESULTS.

A mean scattering function has been obtained through an indirect procedure which, it must be reminded, is probably quite representative of particles in turbid waters. (Section 1.2) The particle scattering functions must be determined, in each case separately, by subtracting the scattering functions $\beta_o(\theta)$. The figures I.9 demonstrate, for the three wavelengths indicated, the regions within which all the particle scattering functions computed in this manner are contained. (112 for $\lambda = 546$ nm, of which 26 correspond to measurements at three wavelengths 546, 436 and 366 nm). The backward protons of only 436 nm are drawn as an example.

Figure I.6 provides another illustration, the computations made from measurements by T. J. Petzold (1972), already given (Fig. I.2 and I.5); chosing the two scattering functions which correspond to the clearest water (Atlantic, 1500 m) and the most turbid oceanic water (coastal California) respectively. After deducting the portion due to molecular scattering the resulting scattering functions are drawn together with the mean scattering function.

Without further modifications, considering the uncertainty for clear waters, it appears that in precisely these waters the suspended particles demonstrate in general a scattering function with a well marked minimum, rapidly followed by an abrupt rise towards $150^\circ$. The mean scattering function, drawn with broken lines

*The case of clear waters still remains difficult to study. According to what has been said, the part $\beta_o(\theta)$ to be subtracted, is predominant for medium angles. Thus the experimental errors of $\beta(\theta)$ are wholly included in $\beta_o(\theta)$, producing a relatively important error. Consequently the backward portion of the scattering function perhaps appears to be more variable than it really is. Thus certain "aberrant" scattering functions have been removed.
Figure I.9: See the text. These curves refer to 26 measurements performed simultaneously at three wavelengths (2 samples from the English Channel, 13 from the Mediterranean, 11 from the Indian Ocean).
on these figures, is almost confined to the curve constituting the lower limit of this region, for angles greater than 90°; for angles less than 90° it is located almost along the center of the region.

3.2 ADOPTION OF A "TYPICAL" PARTICLE SCATTERING FUNCTION.

In order to resolve several problems, especially those concerning the relations between scattering properties and the content of suspended material, which affects the visibility of submerged objects or the propagation of daylight or artificial light, it is necessary to know the particle scattering function. The study of the asymptotic regime of underwater luminance (L. Prieur, A. Morel, 1971) or more generally of all problems related to radiative transfer, make this necessity evident. For these computations or for predictive models, it is useful to be able to use a "typical" particle scattering function, i.e., which provides a satisfactory approximation. For this reason, the "mean" curve obtained between 30° and 150° will be adopted and connected with the "mean" curve between angles 1.5° and 14°. The agreement between these two portions has been made (D. Bauer, A. Morel, 1967) using the measurements of N. G. Jerlov (1961) which have been taken down to 10°. The subsequent measurements by F. Nyffeler (1969-1970) between 1° and 25°, have demonstrated that this interpolation is correct. Under these conditions the scattering coefficients for this "typical" scattering function are as follows:

Table II: "Typical" particle scattering function. Column 1: Volume scattering function normalized at 90°. Column 2: Normalized coefficients β(θ), i.e. the ratio to the given integral b (cf. Appendix 1). For the computation of the integral an exponential extrapolation has been performed - of the form β(θ) = exp [-kθ] - between 1° and 0°.
This produces a value of the integral between 0° and 1° equal to 9.7% of the total integral between 0° and 180°. This value is perhaps small, see the results of T. J. Petzold (1972). This author estimates that between 0.1° and 1° the values constitute 20% to 30% of the final integral between 0° and 180°. Final columns: Perpendicular $\beta_1$ and parallel $\beta_2$ polarization components to the scattering plane, computed from the degree of polarization:

$$p(\theta) = \frac{\bar{\beta}_1(\theta) - \bar{\beta}_2(\theta)}{\bar{\beta}_1(\theta) + \bar{\beta}_2(\theta)} = \frac{1}{2} \frac{\bar{\beta}_1(\theta) - \bar{\beta}_2(\theta)}{\bar{\beta}(\theta)} ;$$

the values of $p(\theta)$ used are presented in Figure I.13 and the "typical" scattering function is presented graphically in Figure III.7 (Part III).

**TABLE II**

<table>
<thead>
<tr>
<th>$\theta$</th>
<th>$\beta(\theta)$</th>
<th>$\bar{\beta}(\theta)$</th>
<th>$\bar{\beta}(90)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>34.400</td>
<td>77.5</td>
<td></td>
</tr>
<tr>
<td>1.5</td>
<td>26.100</td>
<td>58.8</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>19.100</td>
<td>43.0</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>8.500</td>
<td>19.2</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>47.500</td>
<td>10.7</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>29.900</td>
<td>6.7</td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>19.200</td>
<td>4.32</td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>13.000</td>
<td>2.93</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>9.15</td>
<td>2.06</td>
<td></td>
</tr>
<tr>
<td>9</td>
<td>6.70</td>
<td>1.14</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>5.00</td>
<td>1.12</td>
<td></td>
</tr>
<tr>
<td>11</td>
<td>3.14</td>
<td>0.71</td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>2.24</td>
<td>0.505</td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>1.68</td>
<td>0.378</td>
<td></td>
</tr>
<tr>
<td>14</td>
<td>1.30</td>
<td>0.298</td>
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<tr>
<td>15</td>
<td>1.02</td>
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<td></td>
</tr>
<tr>
<td>16</td>
<td>0.75</td>
<td>0.169</td>
<td></td>
</tr>
<tr>
<td>17</td>
<td>0.58</td>
<td>0.130</td>
<td></td>
</tr>
<tr>
<td>18</td>
<td>0.47</td>
<td>0.106</td>
<td></td>
</tr>
<tr>
<td>19</td>
<td>0.38</td>
<td>0.0656</td>
<td></td>
</tr>
</tbody>
</table>

*The total integral has a value of 444 if $\beta(90)$ is given a value identically 1. Since the computation was presented, (A. Morel 1968) an error has been discovered, that stated value being 444/$\pi$ instead of 444. The coefficients of the relation between $\beta(\theta)$ and $\beta$ are wrong by the same ratio (cf. correction 1970). The possible variations of the scattering function, particularly in the forward part, result in possible variations in the integral estimated at ±17% (cf. Section 2.2 and 2.3, Part III).
It must be underlined that such a "typical" scattering function, by its very nature, is simply a mean which is considered to be satisfactory. It is precisely the variations around this mean which will be used during the theoretical interpretation (Part III, Section 2) to deduce information on the nature of the particles (more accurately on their index of refraction) as well as on the law which governs the distribution.

4.0 THE INFLUENCE OF WAVELENGTH.

One must distinguish the qualitative aspect of the problem, i.e. the influence on the shape of the scattering function, from the quantitative aspect, i.e. the influence on the magnitude of the scattering phenomenon.

4.1 VARIATION OF SHAPE OF THE SCATTERING FUNCTION.

This question has actually been implicitly examined previously. To briefly recall the principal points: When the waters are clear, the scattering function which is already symmetric becomes more so with decreasing wavelength, due to the very selectively increasing importance of molecular scattering. On the contrary, in turbid waters, the shape of the scattering function is not practically modified by changes in wavelength, indicating that molecular scattering is negligible and that in addition the particle scattering function is insensitive to this factor. Figure I.7 provides one example of this case.

After removing the molecular scattering, the shape of the particle scattering function actually varies with wavelength, but without evidence of a systematic law dependence*. On the average,

*At least in the case of our measurements, because Hinzpeter (1962) notes that the scattering function at 400 nm is less asymmetric than at 700 nm and it probably involves a particle scattering function since these results correspond to turbid waters from the Baltic.
the scattering functions normalized at 90° are practically similar to those in Figure I. 9. The measurements of F. Nyffeler (1970) are also in favor of a lack of significant influence in the small angle region.

4.2 (WAVELENGTH) SELECTIVITY OF SCATTERING.

4.2.1 Volume scattering functions.

In contrast, after removing molecular scattering, it appears that \( \beta_p(\theta) \) (considering now absolute values) varies systematically with wavelength, slowly increasing with decreasing wavelength. Figure I.10 shows the variations of the ratio of \( \beta_p(\theta, \lambda) \) over \( \beta_p(\theta, 546) \) plotted against angle \( \theta \), where \( \lambda \) is 436 nm and 366 nm. For reasons

Figure I.10: Ratio of \( \beta_p(\theta, \lambda) \) over \( \beta_p(\theta, 546) \) as a function of \( \theta \), with \( \lambda = 436 \text{ nm, 366 nm} \); full line is mean value; vertical bars correspond to standard deviation calculated from N measurements, the values of N indicated. Maximum N is 26, corresponding to 26 samples on which measurements have been performed consecutively at 3 wavelengths (See legend of Fig. I. 9).
already stated, the ratios are better determined at small angles, the variability of the scattering function at large angles resulting in the increase of the standard deviation. Analogous results have been obtained in the Atlantic (for $\theta = 30^\circ$ and $\lambda$ of 436 and 546 nm) presented elsewhere (A. Morel, 1970). J. R. Zaneveld and H. Pak (1973) made a table of measurements at 45°; the ratio of the total scattering coefficients at the two wavelengths 436 nm and 546 nm, varying according to these authors by approximately 1.10 and 1.45 respectively (it does not appear that they obtained the relative coefficients of the particles alone, but the directly measured coefficients, thus the ratio may have been slightly affected).

**Indirect Method.** A precise but indirect method (A. Morel 1967) permits the determination of selectivity for a given angle. It is applied for five wavelengths (578, 546, 436, 405 and 366 nm) to a large number of values at 90° (Figure 1.11) and to a smaller number at 30°, providing essentially the same results, if the confidence interval is considered (Figure 1.12). It is based on the following principle: If measurements are made relative to optically pure benzene, used as a standard, the selectivity of scattering in a sample may be deduced from the selectivity of scattering in the benzene, which is known experimentally and computed theoretically. In practice one must proceed as follows: denoting by $r$ the ratio of the scattering coefficient of the sample to that of the benzene (for the same angle $\theta$, not included for simplicity), one obtains for $\lambda = 546$ nm,

$$
    r(546) = r_0(546) + r_p(546),
$$

and for any other wavelength

$$
    r(\lambda) = r_0(\lambda) + r_p(\lambda),
$$
Figure I.11: $r(90, \lambda)$ as a function of $r(90, 546)$ for various samples (see text). The open symbols correspond to the English Channel, the black symbols to the Mediterranean.

Figure I.12: Results obtained by the method illustrated in the previous figure (Fig. I.10) concerning selectivity (at 30° and 90°) of particle scattering. The vertical bars correspond to the standard deviation of the values, the curves correspond to the indicated power laws.
the subscripts \( o \) and \( p \) denoting as previously, the part due to water and that due to particles. The index \( b \) in the following refers to benzene.

By expanding

\[
\beta'(546) = \frac{\beta_o(546) + \beta_p(546)}{\beta_b(546)}
\]

and supposing that as wavelength changes, each scattering coefficient is multiplied by a specific coefficient \( K \), i.e. by \( K_o, K_p, K_b \) respectively one may write:

\[
r(\lambda) = \frac{K_o \beta_o(546) + K_p \beta_p(546)}{K_b \beta_b(546)} = \frac{K_o}{K_b} r_o(546) + \frac{K_p}{K_b} r_p(546)
\]

and finally by reintroducing \( r(546) \)

\[
r(\lambda) = \text{constant} + \frac{K_p}{K_b} \left[ r(546) - r_o(546) \right]
\]

If the selectivity of particle scattering was zero (\( K_p = 1 \)), a linear relation between \( r(\lambda) \) and \( r(546) \) would be obtained, with slope the inverse of \( K_b \).* Actually, plotting \( r(\lambda) \) as a function of \( r(546) \) for each sample, approximately linear behavior appears, but with slopes different from \( 1/K_b \), which permit evaluation of \( K_p \). These values obtained for \( \theta = 90^\circ \) and \( \theta = 30^\circ \), are shown as a function of wavelength in Figure I.12 and are compared to curves corresponding to selectivities of \( \lambda^{-0.8} \) and \( \lambda^{-1.2} \). It is noted that they are very close to the values obtained directly, by the first method.

*The values used for the selectivity of scattering of benzene are as follows (A. Morel, 1966).

\[
\lambda_{nm} = 578, 546, 436, 405, 366
\]

\[
K_b = 0.79, 1, 2.78, 3.88, 6.36
\]
4.2.2 **Total scattering coefficient.**

It does not appear that direct measurements of \( b \) have been performed at several wavelengths on the same sample or at the same location and depth. The conclusions which in any case are quite controversial cannot be produced without resorting to hypotheses. If the shape of the particle scattering function can be considered to be insensitive to wavelength, the selectivity found for various angles (which behaves very roughly as \( \lambda^{-1} \)) is wholly included in the total coefficient \( b \). This is for example the hypothesis made by J. R. Zaneveld and H. Pak (1973), which is probably well justified.

Another hypothesis consists in accepting that in the attenuation due to particles, the term due to scattering is largely dominant* over the term absorption. This is the hypothesis of W. V. Burt, 1955. If dispersion is observed for the attenuation coefficient \( c \), it is totally attributed to the coefficient \( b \). Burt interprets in this way the measurements made with a spectrophotometer and the resulting spectra, corresponding to a law of \( \lambda^{-1} \) or \( \lambda^{-2} \) approximately.

---

*This hypothesis is probably and partially erroneous for certain spectral bands in the case of strongly colored particles, while it appears realistic as the following experiment demonstrates in an approximate way: if the spectrum of a sample of sea-water is determined with a spectrophotometer, as a ratio to the same water after filtration, the particles act in each case as scattering and absorbing centers. These same particles concentrated on a filter of the same diameter as the container behave mostly as absorbing centers when the filter is "clarified" with immersion oil (using as reference a fresh filter clarified in the same way, method of C. Yentsh 1957). Hence in this manner, one does not measure something comparable to that measured in the preceding experiment, as when one filters a volume 20 times greater than that of the container. This demonstrates that scattering largely dominates over absorption. The approximations made: A spectrophotometer does not measure \( c \) but a number intermediate between \( c \) and \( a \), the particles on the filters still being scatterers, thus the approximations are in the same direction reinforcing the conclusion.
5.0 POLARIZATION.

5.1 RESULTS.

H. Hinzpeter (1962) apparently for the first time demonstrated that the angular partition of the degree of polarization of incident to natural light, is very nearly symmetric around 90° with a maximum value at this angle of the order of 0.40 to 0.65 (slightly less at 700 nm than at 400 nm) and values of the order* of 0.10 at 30° and 150°. Beardsley (1968) has measured the polarization in all possible configurations, with combinations of polarizer and of quarter wavelength plate, in each case on the direct and scattered beams. In addition it was possible to determine, for various angles, the sixteen elements of the matrix for application to the four Stokes parameters which characterize the incident light (cf. Eq. 1.12, Part II). This matrix contains small values for the non-diagonal elements which must theoretically be zero if the particles were spheres. Thus the strongly diagonal character of the matrix, determined experimentally, demonstrates that the assumption for spheres constitutes a good approximation (cf. 1.2, Part II).

The variation of the degree of polarization with angle θ, with natural incident light, is approximately symmetric as shown in Figure 1.13, where on the left side is a mean of 5 measurements from the English Channel & of 3 on very turbid waters** of the Mediterranean (bay of Villefranche). The extreme values observed at 90° (60% and 79%) are denoted by the vertical bar. The stippled line corresponds to a curve determined for optically pure water (A. Morel, 1966) with \( \rho(90°) = 84\% \). On the right hand side are

* For very turbid waters of the Baltic.

** Measurements on clear waters, from which however the portion of molecular scattering must be deducted in order to calculate the polarization due to particles only, are subject to caution, because of the loss of sensitivity following the positioning of an analyzer. The 8 measurements used here are relative to waters that are sufficiently scattering (b ≈ 1 m\(^{-1}\) ) to make the influence of molecular scattering negligible.
traced curves obtained by G. F. Beardsley (1968) (with exception of station "Atlantique 4"); they demonstrate that the maximum degree of polarization (at $90^\circ$) cannot exceed 50% and in addition that the angular dependence is more complex in the zone $20^\circ$-$40^\circ$.

5.2 VARIABILITY OF THE DEGREE OF POLARIZATION AT $90^\circ$.

The systematic measurements of A. Ivanoff (1961) performed at $90^\circ$ demonstrate, for the samples of sea-water studied, that the degree of polarization at this angle, varies in a very broad manner from a value (80%) approaching that of pure water to low values just greater than 40%. This variation occurs in a very regular manner, those waters with the largest scattering coefficient having grosso modo (roughly) the least degree of polarization. It must be attempted to determine whether this effect is due only to the fact that molecular scattering, for which the degree of polarization is high, has a progressively diminishing role, or on the contrary whether the degree of polarization of light diffused by the particles is itself variable, and if so in what manner. By definition (cf. Part II, Section 1.1) the degree of polarization is the ratio of the intensity of the polarized light $I_\pi$ to the total intensity
$I_\pi + I_\sigma$ of natural light and polarized light. Decomposing into that which is produced by particle scattering and by the water itself (indices $p$ and $o$ respectively)

\[ p = \frac{I_\pi}{I_\pi + I_\sigma} = \frac{I_\pi p + I_\sigma o}{I_\pi p + I_\sigma o + I_{np} + I_{no}}, \]

and in a similar way

\[ p_o = \frac{I_\pi o}{I_\pi o + I_{no}} \quad \text{and} \quad p_p = \frac{I_\pi p}{I_\pi p + I_{np}}, \]

The denominators of these three expressions are proportional to $\beta$, $\beta_o$ and $\beta_p$ respectively. Therefore

\[ p = \frac{\beta}{\beta} \quad p_o = \frac{\beta}{\beta} \quad p_p \]

i.e., an expression corresponding to a "law of mixtures" where $p$ varies according to a hyperbolic law with $\beta$:

\[ p = p_o + \frac{\beta}{\beta} (p_o - p_p) ; \]

$p$ tends to the limiting value $p_p$ as the scattering coefficient $\beta$ rises, and towards $p_o$ as the scattering is reduced to molecular scattering (i.e. $\beta_o / \beta = 1$)*

**Figure I.14**: For various values $p_o$ of the degree of polarization for particles at $90^\circ$; variation of the degree of polarization of a sample versus the value of its scattering coefficient at $90^\circ$, with $p_p$ taken to be molecular scattering $\beta_o(90^\circ)$ at the same angle. The ordinate scale is logarithmic.

*There is no assumption in this computation, excepting that which is justified by the additivity of the scattered waves.
In order to compare with the experimental values of A. Ivanoff the logarithm of $\beta/\beta_0$ is plotted (not the $\beta/\beta_0$) as a function of $p$ --- see Figure I.14 --- and the two straight lines are redrawn delimiting the region wherein the experimental points are concentrated, as well as the mean line. The curves corresponding to various values are denoted a priori as $p^*_p$, the value of $p_0$ being 0.84. It appears that the distribution of the experimental points corresponds to a variable degree of polarization for the particles from 0.7 to 0.4 and that in addition this variation has a quite systematic character. The particles of the more turbid waters are actually the least polarizing, those of the clear waters demonstrate on the contrary a variable degree and may reach up to values of the order of 0.8 (this of course explains how the extrapolation made by A. Ivanoff produces a value, 0.88, a little large for molecular scattering).

Finally these values, particularly those relative to $90^\circ$, are in their diversity consistent, and confirm the variability of $p(90)$. This, as will be seen in Part III, is theoretically plausible, but among the reasons which one may find for it, several, as the influence of the shape of the particles remain difficult to isolate.

6.0 RELATIONS AMONG THE SCATTERING COEFFICIENTS

The use of certain ratios of angular scattering functions has been destined to serve as the descriptive element of the scattering function (for example the ratio $\beta(30)/\beta(45)$).

A. F. Spilhaus, 1968). More generally, and confronting the difficulty of directly measuring the total scattering coefficient $b$, it has been attempted to evaluate it from measurements of this or that scattering function $\beta(\theta)$. N. G. Jerlov has since 1953 used this method and deduced $b$ from $\beta(45^\circ)$. Such methods assume at least approximately the ratio $\beta(\theta)/b$, i.e. that which is called the normalized coefficient $\beta(\theta)$, to be sufficiently constant. The precaution is not always taken in using these methods, to remove from $\beta(\theta)$ the part produced by molecular scattering, which is addition introduces uncertainties when $\theta$ is greater than $45^\circ$ (and even at $45^\circ$ if the waters are very clear).
Such relations may be written from relative values of $\bar{A}(\theta)$ of the "typical" particle scattering function and also taking into consideration molecular scattering (A. Morel, 1968). Thus one obtains

$$b = \frac{1}{\bar{A}_p(\theta)} \left[ \beta(\theta) - \beta_o(\theta) \right] + b_o;$$

$\bar{A}(\theta)$ being the measured value on the sample, from which the molecular term $\beta_o(\theta)$ is removed. With the values of Table II, this relation may be written for $30^\circ$, $45^\circ$ and $90^\circ$:

$$b \approx 14 \left[ \beta(30) - \beta_o(30) \right] + b_o$$

$$b \approx 50 \left[ \beta(45) - \beta_o(45) \right] + b_o$$

$$b \approx 445 \left[ \beta(90) - \beta_o(90) \right] + b_o$$

Often $\beta_o(30)$ can be neglected, but $\beta(45)$ cannot in the case of clear waters (cf. Figure I.8) and finally $\beta_o(90)$ cannot either except in very turbid waters. The possibility of neglecting these terms exists, the less so obviously, as the wavelength becomes shorter. The value $b_o$ which varies with wavelength appears in Table I. The same remark as above applies regarding the possibility of taking it into consideration.

The review of experimental values obtained by numerous authors for these ratios, either between two angles, or between angular coefficient and total coefficient, will be made elsewhere in more detail (Part III, Section 2). In effect the variability of these ratios, which in reality reflect the variability of the particle scattering function, may be theoretically interpreted in terms of the variability of the particles themselves.
PART II

PREFACE

The search for a theoretical explanation of the experimental results has two aspects: on the one hand; is the existing knowledge of the nature of suspended marine particles and of their properties compatible? In other words does one explain the other? on the other hand; is it possible, through observation of their scattering properties, to deduce information on the particles which would be difficult to obtain by other means? These questions have already motivated several studies, notably those of Y. Otchakovsky (1965), T. Sasaki (1967), T. Sasaki et al. (1968), and more recently H. Pak et al. (1971), G. Kullenberg (1972), H. R. Gordon and O. B. Brown (1972) the list not being exhaustive. The last three were made not from published and tabulated values of Mie functions, but instead using values computed from them. It does not appear that the possibilities offered by theory have been systematically explored, nor have the comparisons between theoretical and experimental results been performed methodically. Thus the results of theoretical calculations must be interpreted in order to be in a position to predict the influence of diverse parameters, without however increasing the calculations. Under this condition a guideline may be adopted, which will serve as a guide when these comparisons are undertaken. What is presented here is tentative in this sense. The purpose of the present PART II is thus to provide the necessary theoretical basis for solving the problem, and to demonstrate what, and under which conditions, can possibly be predicted from the calculations. The comparison of these theoretical calculations and of the experimental results is left for PART III.

Initially (Section I) the general relationships between the polarizations of the incident and scattered waves is briefly exposed. The studies most frequently cited are those of F. Perin (1942), as well as chapters dedicated to this subject in the works of Van de Hulst (1957), Preisendorfer (1965) and Deirmendjian (1969) which provide more penetrating developments. It is intended here to simply determine the framework within which is situated the most restricted case to be further examined, namely that in which the incident light is considered natural or linearly polarized (perpendicular or parallel to the scattering plane).
The suspended marine particles whose sizes vary over an extensive range obey Mie's theory (1908). This furnishes the rigorous analytical solution to the scattering problem of a plane wave by a spherical particle of any size by applying electromagnetic theory using Maxwell's equations. From this point of view, it agrees with Rayleigh's theory when the particles become very small compared to the wavelength of light, and on the other extreme it tends toward the limiting solution of geometrical optics (reflection, refraction) and of diffraction theory when the particles become large.

The exposition of Mie theory, which became classic particularly after the publication of Van de Hulst's work* will not be repeated in the context of this study, so Section 2 only recounts the formulation necessary to the subsequent development. In contrast it appears useful to trace the evolution (functional dependence) of the scattering function by varying the parameters size and index using the numerical results provided by this theory. Actually it is not easy to obtain an accurate idea of this dependence within the literature. Therefore for a "practical user" the possibility of approximately predicting the result leads to the choice of calculations to be made, just as well for the scattering functions of individual particles as, following that, for the scattering functions of polydisperse systems. The interpretation which must be sought by this dependence, passes in practice through the theories which apply at the extremities of the Mie domain, i.e. on the one side by the theories of Rayleigh and Rayleigh-Gans and on the other by diffraction and geometric optics. The Rayleigh-Gans approximation provides in addition very important elements for predicting the scattering properties of a polydisperse medium.

Section 3 demonstrates how the scattering properties can be computed for a population of particles of varying size but assumed to be of the same nature ("polydisperse ensemble"). The physical significance of the computation is discussed, keeping in mind that the mathematical limits corresponding to maximum and minimum size have been set somewhat arbitrarily. This is a delicate problem which reappears on several occasions in PART III.

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*This is the reason why references to the original works of Mie, Rayleigh, Debye and Gans have been omitted. The reader will find at the end of each chapter in Van de Hulst's book an annotated bibliography and commentary from a historical point of view.
Finally, the initial hypothesis of considering that the marine particles can be compared to transparent spheres, needs some comments. Extensions of Mie theory to non-homogeneous spheres or to particles of different shape (ellipsoids, cylinders, etc.) already exist but lead in general to more complicated calculations. This is one reason for restricting the analysis to spheres. A second more convincing reason is that marine particles have a variety of shapes, and due to their perfectly random orientation, the sphere offers the best approximation. Confirmation of this hypothesis can be found in the experimental work of Hodkinson (1963) and of Holland and Gagne (1970). These last authors demonstrate that irregular particles behave practically as equivalent spherical particles in scattering natural light, (except perhaps for backscattering), but in contrast the polarization is quite affected. This possible result will necessitate small changes in conclusions with respect to polarization (PART III)*.

The choice of uniquely real indices, must also be explained. The introduction of an imaginary part to account for the absorption by particles, does not practically change the results, since the term to be introduced, actually quite hypothetical, must be in any case quite small. Its influence thus becomes negligible, as seen in Fig. II.17, where all other things being equal, the scattering function calculated for an index of refraction of 1.05 is compared to that calculated by O. B. Brown and H. R. Gordon (1972) for an index of 1.05-0.01i. The values are approximately overlapping, even though the imaginary term corresponds to quite strong absorption (92% for traversing a layer 10 microns thick).

In conclusion, by retaining the hypothesis, the model is without doubt idealized, but it constitutes an initial approach or, if you like, a reference, in comparison to which the deviations can be interpreted.

*There is actually a difference between the polarization caused by marine particles from that which would be caused by spherical particles, but it is quite small. (G. F. Beardsley, 1968). (cf. PART I, Section 5).
1. STATE OF POLARIZATION AND SCATTERING

1.1 DESCRIPTION OF THE STATE OF POLARIZATION BY THE STOKES PARAMETERS.

To a beam of totally polarized monochromatic light corresponds an electric field vector \( \vec{E} \) whose components in two perpendicular axes \((\vec{r}, \vec{l})\) in the plane of the wave are written as:

\[
\begin{align*}
E_r &= p_r e^{i(\omega t - \varphi)} \\
E_l &= p_l e^{i(\omega t - \varphi_2)}
\end{align*}
\]  

\((1.1)\)

\(\vec{r}\) and \(\vec{l}\) may be any axes but are chosen so that their vector product \(\vec{r} \times \vec{l}\) is oriented in the direction of propagation of light (incident or scattered); \(\vec{r}\) is perpendicular to the scattering plane and \(\vec{l}\) is in the plane (this plane being defined by the directions of incidence and scattering).

The four Stokes parameters are defined by

\[
\begin{align*}
I &= E_r^* E_r + E_l^* E_l \\
Q &= E_r^* E_l - E_l^* E_r \\
U &= E_r^* E_l + E_l^* E_r \\
V &= i (E_r^* E_l - E_l^* E_r)
\end{align*}
\]  

\((1.2)\)

According to (1.1) three independent parameters suffice to describe the state of polarization: \(p_r, p_l\) and the phase difference \(\delta = \varphi - \varphi_2\); there effectively exists a relation between the four Stokes parameters (see (1.6) below). Interpreting (1.2) using (1.1) one obtains

\[
\begin{align*}
I &= p_r^2 + p_l^2 \\
Q &= p_r^2 - p_l^2 \\
U &= 2 p_r p_l \cos \delta \\
V &= -2 p_r p_l \sin \delta
\end{align*}
\]  

\((1.3)\)

* Translator's note: The original paper uses the notation \(\vec{r} \wedge \vec{l}\) to denote the vector cross-product, which is popular in Europe. Here I revert to the notation \(\vec{r} \times \vec{l}\) customarily used in the U.S.
A simple geometrical interpretation may be given: the vector \( \vec{E} \) in the general case describes an ellipse whose axes are different from the system \((1, r)\). Setting \( \tan \epsilon = e \), where \( e \) is the ellipticity (the ratio of the short to the long axis) and denoting by \( \alpha \) the angle between the long axis and \( \vec{P} \), \( \alpha \) may be found by reducing the ellipse onto its principal axes.

\[
\tan 2\alpha = 2 \cos \delta \frac{P_1 P_r}{2} \frac{2}{P_r - P_1} \tag{1.4}
\]

The energy of the oscillation, which does not depend on the phase is \( I = p_1^2 + p_r^2 \) which is set equal to \( p^2 \). Through geometric considerations the following new expressions are found.

\[
I = p^2, \quad Q = p^2 \cos 2\epsilon \cos 2\alpha, \quad U = p^2 \cos 2\epsilon \sin 2\alpha, \quad V = p^2 \sin 2\epsilon. \tag{1.5}
\]

which permit interpretation of the following particular cases.

- If \( p_1 = p_r \), then \( \alpha = \pi/4 \) from (1.4) and the oscillation is:
  - linear, on the first bisector (\( \delta = 0 \)) or on the second (\( \delta = \pi \)). \( Q \) and \( V \) are zero, \( U \) is not zero, the Stokes vector being written \((1, 0, \pm 1, 0)\) assuming \( I \) unitary.
  - circular, left or right, depending on the sign of \( \delta = \pm \frac{\pi}{2} \); in this case \( Q \) and \( U \) are zero, \( V \) is not and one obtains \((1, 0, 0, \pm 1)\).

- If \( p_1 \) or \( p_r \) are zero, then \( I = \pm Q \), the polarization is rectilinear, perpendicular or parallel to the scattering plane respectively and the parameters are \((1, \pm 1, 0, 0)\).

Finally in the general case, \( \delta \) differs by multiples of \( \pi/2 \), the oscillation is elliptical and the Stokes parameters attain various values. In any case the following relation is obtained from (1.5)

\[
I^2 = Q^2 + U^2 + V^2 \tag{1.6}
\]

which constitutes the criterion of total polarization.
If on the contrary the light is completely unpolarized, it must be considered that $E_r$ and $E_l$ exist but have no phase coherence: $p_r$ and $p_l$ are equal and $I$ is defined in the same way. In contrast none of the values $Q, U, V$ are statistically different from zero, in the common time scale, and the Stokes vector for natural light is written $(I, 0, 0, 0)$.

Consider partially polarized light as the superposition of natural light $I_N$ and totally polarized light $I_P$, interpreted as

$$I = I_N + I_P$$

where

$$I_P = (Q^2 + U^2 + V^2)^{1/2},$$

producing the inequality

$$I^2 > Q^2 + U^2 + V^2$$

In its most general form, the degree of polarization (restricted between 0 and 1) is the ratio:

$$\frac{(Q^2 + U^2 + V^2)^{1/2}}{I}$$

(1.7)

1.2 SCATTERING MATRICES.

When two waves uncorrelated in phase are superposed, the Stokes parameters are additive. An optical medium or equally well a scattering medium penetrated by an incident wave $(I_0, Q_0, U_0, V_0)$ and producing an emergent wave $(I, Q, U, V)$, results in linear combinations of the initial parameters; in other words the medium is characterized by a transformation matrix of $4 \times 4$ coefficients:

$$\begin{pmatrix}
I \\
Q \\
U \\
V \\
\end{pmatrix} = \begin{pmatrix}
a_{11} & \cdots & \cdots & a_{14} \\
\cdots & \cdots & \cdots & \cdots \\
a_{14} & \cdots & \cdots & a_{44} \\
\end{pmatrix}
\begin{pmatrix}
I_0 \\
Q_0 \\
U_0 \\
V_0 \\
\end{pmatrix}$$

(1.8)
The modification of the field vector $\mathbf{E}$ relative to the incident wave, is expressed by the following linear relations, relating the components

$$
E_r = A_1 E_{r,0} + A_4 E_{1,0} \quad \quad E_1 = A_3 E_{r,0} + A_2 E_{1,0}
$$

(1.9)

where

$$
\begin{vmatrix}
A_1 & A_4 \\
A_3 & A_2
\end{vmatrix}
$$

constitutes thus the matrix of the transformation of the amplitudes, the coefficients of the matrix being complex. Introducing (1.9) in the definitions (1.2) and rearranging, the 16 elements of the matrix (1.8) are calculated, which when applied to the initial parameters, permit calculation of the final parameters. These 16 coefficients are all real and form quadratic expressions of the type $A_j A_k$. For scattering problems the "modified" Stokes parameters ($I_1', I_2', U, V$) are used most often, deduced simply by:

$$
I_1' \text{ (or also } I_r) = \frac{1}{2} (I+Q)
$$

$$
I_2' \text{ (or also } I_1) = \frac{1}{2} (I-Q)
$$

$U$ and $V$ remaining unchanged. The criterion for total polarization (1.6) is written with this notation:

$$
4I_1 I_2 = U^2 + V^2
$$

The parameters characterizing natural light are then $(1/2, 1/2, 0, 0)$ and the degree of polarization defined by (1.7) reduces to

$$
\frac{I_1 - I_2}{I_1 + I_2}
$$

(1.10)

Scattering by isotropic spheres.

If the particles possess certain symmetries the number of independent coefficients becomes smaller than 16; a particularly simple case being that of isotropic spheres for which the matrix for the amplitudes is diagonal ($A_3 = A_4 = 0$), the deduced matrix for intensities becoming quasi-symmetric:
\[
\begin{align*}
\begin{bmatrix}
I_1 \\ I_2 \\ V \\
\end{bmatrix} &= 
\begin{bmatrix}
I_{1,0} \\ I_{2,0} \\ U_0 \\
\end{bmatrix}
\begin{bmatrix}
A_1 A_1^* & 0 & 0 \\
0 & A_2 A_2^* & 0 \\
0 & 0 & 1/2(A_1 A_2^* + A_2 A_1^*) \\
\end{bmatrix}
\begin{bmatrix}
0 \\
0 \\
i/2(A_1 A_2^* - A_2 A_1^*) \\
\end{bmatrix}
\begin{bmatrix}
I_{1,0} \\ I_{2,0} \\ U_0 \\
\end{bmatrix} \\

\text{Which becomes}
\begin{align*}
\begin{bmatrix}
I_1 \\ I_2 \\ V \\
\end{bmatrix} &= 
\begin{bmatrix}
I_{1,0} \\ I_{2,0} \\ U_0 \\
\end{bmatrix}
\begin{bmatrix}
i_1 & 0 & 0 \\
0 & i_2 & 0 \\
0 & 0 & i_3 + i_4 \\
\end{bmatrix}
\begin{bmatrix}
I_{1,0} \\ I_{2,0} \\ U_0 \\
\end{bmatrix} \\
\end{align*}
\tag{1.11}
\]

using the notation of the intensity function \( i \) (see definition in Appendix I). The factor \( 1/k^2 \) (where \( k = 2\pi/\lambda \)) comes from the fact that the dimensionless functions of intensity are defined from the functions of amplitude \( S = kA \) by \( i_1 = S_1 S_1^* \), \( i_2 = S_2 S_2^* \) etc; the terms of the matrix of \( A \)'s (1.11) being the partial effective cross-sections \( \sigma_1, \sigma_2, \ldots \) (cf. definitions, Appendix I). This matrix contains 4 non-zero elements, of which only 3 are independent, since there exists among them a relation originating from the expressions contained in (1.11), which is written:

\[
i_{12} = i_3^2 + i_4^2
\tag{1.12}
\]

In applying the matrix Eq. (1.12) the following differing cases must be considered:

- If the incident light is elliptically \((I_1, I_2, U, V)\) or circularly \((I_1, I_2, 0, V)\) polarized or rectilinearly but not along \( \mathbf{r} \) or \( \mathbf{T} \) \((I_1, I_2, U, 0)\), the scattered light is in general elliptic since \( i_3 \neq i_4 \neq 0 \); in addition it is totally polarized if so is the incident light, which can be easily verified from the criterion of polarization (1.10) for the scattered light, taking into consideration the relation (1.12).

---

*Perrin and Abragam (1951) have demonstrated that in the case of simultaneous existence of spheres of different size, this equality is not true except for the angles 0° and 180°, the inequality which replaces it being \( i_1 i_2 > i_3^2 + i_4^2 \). The criterion of polarization cannot be satisfied any longer and a partial depolarization of the scattered light appears even if the incident light is totally polarized. Perrin and Abragam, as well as Deirmeindjian (1969) have suggested using this factor of depolarization as an indicator of the heterogeneity of sizes.*
- If the light is rectilinearly polarized along \( \vec{r} \) or \( \vec{l} (I_1, 0, 0, 0) \) or \( (I_2, 0, 0, 0) \) the scattered light completely conserves this property.

- Finally if the light is natural \((1/2, 1/2, 0, 0)\) the scattered light is partially polarized, since in general \( i_1 \) differs from \( i_2 \); its total intensity is \( I = I_1 / 2 (i_1 + i_2) \) and its degree of polarization is

\[
\frac{i_1 - i_2}{i_1 + i_2}
\]

as shown above.

---

The case of angles 0° and 180°.

It will be seen later (Section 2.1.2) that \( S_1(0) = S_2(0) \); it follows immediately, through (1.11), that \( i_1 = i_2 = i_3 \) and that \( i_4 = 0 \); the scattering matrix is diagonal and the state of polarization, whatever it is, is strictly retained; the same is true for the state of non-polarization. This can be anticipated from considerations of symmetry: in essence the "scattering plane" previously defined, does not exist any longer, and since the particles are isotropic spheres no preferred direction is privileged in this system of axes. For scattering at 180° the same reasoning holds but with one difference: the scattering plane is not defined, but by continuity i.e. as \( \theta \) approaches 180° (see previous figure) it is seen that the vector \( \vec{l} \) for the scattered light is opposed to the vector \( \vec{l} \) of the incident oscillation. In this case (cf. Section 2.1.2) \( S_1(180) = -S_2(180) \), thus \( i_1 = i_2 = -i_3 \), while \( i_4 \) remains zero. Every linear polarization remains unchanged: the rotation of \( \vec{l} \) cancelling the sign of \( U(= -iU_0) \). In contrast, considering this rotation, \( V(= iV_0) \) retains its sign, the sense of any circular (or elliptic) polarization being inverted. Applications of these last two points have been examined: the use of lighting with polarized light has been conceived in order to reduce the glow due to backscattering in underwater viewing ports. (R. O. Briggs and G. L. Hatchett, 1965, and G. D. Gilbert and J. C. Pernicka, 1966, to name only the initiators of this technique).
2. SCATTERING BY A SPHERICAL PARTICLE

2.1 FORMULATION OF THE MIE THEORY

In the case of an isotropic particle, spherical and optically inactive, the scattering phenomenon possesses an axial symmetry, the axis being the direction of propagation of the incident wave, on the condition that the incident wave itself possesses this symmetry, which is the case with natural light. Any plane containing this direction is a plane of symmetry; and any other which also contains the direction of observation of scattering is the "scattering plane" (often called "horizontal" due to the experimental configuration usually adopted). In this plane, $\theta$ is the angle between the directions of incidence and scattering; effectively a single angular parameter suffices by reason of the symmetry of revolution. As has been already seen (Section 1.2), the components of the electric field follow two rectangular axes taken in the plane of scattered light, the one perpendicular (l or r), the other parallel (2 or l), to the scattering plane, having amplitudes $S_1(\theta)$ and $S_2(\theta)$ ($S_3$ and $S_4$ being zero). The corresponding intensity functions (dimensionless, cf. Appendix I) are obtained by taking the square of the modulus

$$i_1(\theta) = S_1(\theta) S_1^*(\theta)$$
$$i_2(\theta) = S_2(\theta) S_2^*(\theta) .$$  \hspace{1cm} (2.1)

Mie theory supplies the exact solution for the amplitudes which in addition to the angle $\theta$, also depend on:

- the size of the particle, or more accurately the relative size $\alpha = \frac{2\pi r}{\lambda}$ where $r$ is the spherical particle radius and $\lambda$ is the wavelength in the medium surrounding the particle.

- the relative index of refraction $m$, i.e. the ratio of the real or complex index of the particle relative to that of the surrounding medium.

This solution is expressed in the form of converging series:
\[ S_1(a, m, \theta) = \sum_{n=1}^{n=\infty} \left( \frac{2n+1}{n(n+1)} \left( a_n r_n(\cos \theta) + b_n r_n(\cos \theta) \right) \right) \]

\[ S_2(a, m, \theta) = \sum_{n=1}^{n=\infty} \left( \frac{2n+1}{n(n+1)} \left( b_n r_n(\cos \theta) + a_n r_n(\cos \theta) \right) \right) \]

for \( n = 1, 2, 3, \ldots \) positive integer

The parameters \( a, m \) and \( \theta \) have different roles: actually each term of either series combines functions uniquely dependent on the angle \( \theta \) (by \( \cos \theta \)) with the functions \( a_n \) and \( b_n \) which depend only on the size and on the relative index. \( (\alpha \text{ and } m) \). The final amplitude appears as a sum of all the partial amplitudes corresponding to the different orders \( n \). The relative importance of the partial amplitudes is determined by the "Mie coefficients" \( a_n \) and \( b_n \). For a given \( m \), the values of these coefficients increase with increasing \( \alpha \), and consequently the number of the partial waves (which may be associated with those emitted by oscillating multipoles) to be considered is large and the series converge slowly. On the contrary when \( \alpha \) is sufficiently small, the coefficients beyond the second order are negligible and as will be seen later, the formulae reduce to those of a Hertz dipole; or put in another way this limited case is Rayleigh scattering.

2.1.1 THE FUNCTIONS \( \pi_n, \tau_n \) AND \( a_n, b_n \).

They are expressed by:

\[
\pi_n(\cos \theta) = \frac{1}{\sin \theta} \left| \frac{P_n'(\cos \theta)}{P_n(\cos \theta)} \right| \]

\[
\tau_n(\cos \theta) = \frac{d}{d \theta} \frac{P_n'(\cos \theta)}{P_n(\cos \theta)} ,
\]

where \( P' \) is the first derivative of the Legendre polynomial \( P_n \) of order \( n \); and \( \pi_n \) by

\[
a_n = \frac{\psi_n'(m \alpha) \psi_n(a) - n \psi_n(m \alpha) \psi_n'(a)}{\psi_n'(m \alpha) \zeta_n(a) - n \psi_n(m \alpha) \zeta_n'(a)} \]

\[
b_n = \frac{\psi_n'(m \alpha) \zeta_n(a) - \psi_n(m \alpha) \zeta_n'(a)}{\psi_n'(m \alpha) \zeta_n(a) - n \psi_n(m \alpha) \zeta_n'(a)} ,
\]

where \( \psi_n \) is the Ricatti-Bessel function of order \( n \), \( \psi_n' \) its first derivative; \( \zeta_n \) is the Ricatti-Hankel function of order \( n \), \( \zeta_n' \) its first derivative. \( n \) The arguments are \( \alpha \) and the product \( m \alpha \), which have already been defined.

* From this point on until paragraph 2.2 the relative index will be denoted by \( m \), to be distinguished from the integer \( n \).
These functions $\psi_n$ and $\zeta_n$ are related to the Bessel and Hankel function of order $n + 1/2$ by

$$
\psi_n (x) = \left( \frac{\pi x}{2} \right)^{1/2} J_{n+1/2} (x) \quad \text{and} \quad \zeta_n (x) = \left( \frac{\pi x}{2} \right)^{1/2} H_{n+1/2} (x),
$$

(2.5)
or

$$
H_n (x) = J_n - i Y_n (x). \quad (2.6)
$$

$J_n$ and $Y_n$ being Bessel functions of the first and second kind respectively of order $n$. $H_n$ the Hankel function of the same order.

Introducing the Ricatti-Bessel function of the second kind $\chi_n$:

$$
\chi_n = \left( \frac{\pi x}{2} \right)^{1/2} \chi_{n+1/2} (x), \quad (2.7)
$$

and $\zeta_n (x)$ is then expressed by

$$
\zeta_n (x) = \psi_n (x) + i \chi_n (x). \quad (2.8)
$$

It should be pointed out that $a_n$ and $b_n$ are complex due to the function $\gamma_n$, even when the argument $(mk)$ is real (non-absorbing particles). In the latter case the real and imaginary parts are directly separable which is indicated by substituting (2.8) into (2.4). In practice it is possible to calculate these series, since for each order $n$, the values of the functions $a_n$, $b_n$, $\pi_n$, $\tau_n$ can be deduced from the values of previous orders, using the recurrence relations for the Legendre polynomials and the Bessel functions. This is examined further in Appendix II, "computational procedure and computer application." In the appendix it is shown that in the case just considered, when $m$ is real, $a_n$ and $b_n$ may be obtained after separation of real and imaginary parts in the form of

$$
\text{Re} \{a_n\} = \frac{1}{1 + \frac{p^2}{q^2}} \quad \text{and} \quad \text{im} \{a_n\} = \frac{-p^{1/2} \tilde{g}}{1 + \frac{p^2}{q^2}}.
$$
p and q being themselves real, p/q may take any value between $+\infty$ and $-\infty$ with varying order n. This permits elucidation of an important property of the complex sequences $a_n$ and $b_n$, in a different way than Van de Hulst: the locus of the image of $a_n$ (or of $b_n$) is simply found by setting $x = \text{Re} \{ a_n \}$, $y = \text{Im} \{ a_n \}$, then eliminating p/q between X and Y; One obtains the relation $Y^2 + X^2 - X = 0$ of a circle centered at $I_m = 0$, $\text{Re} = 0.5$ of radius 0.5.

Figure (II.1) shows the images of successive values of the first term $a_1$ as $\alpha$ varies between 1 and 100, the computation having been made for an index of refraction of 1.05.

Figure II.1: Values of the complex coefficient $a_1$ in the case of real index of refraction 1.05. Examples corresponding to complex indices may be found in particular in the work by D. Deirmendjian (1969); the loci of the images of $a_1$ no longer remain a circle but a spiral remaining in the interior of the circle.
As $\theta$ varies from $0^\circ$ to $180^\circ$, the functions $\tau_n$ and $\pi_n$ behave in an increasingly more complex way as the order $n$ is increased; for the first orders:

\begin{align*}
\tau_1(\cos\theta) &= 1 \\
\tau_2(\cos\theta) &= 3 \cos \theta \\
\pi_1(\cos\theta) &= \cos \theta \\
\pi_2(\cos\theta) &= 3 \cos 2\theta
\end{align*}

(2.9A)

At higher orders, more frequent oscillations of increasing amplitude appear, the larger amplitudes corresponding to values $\theta = 0^\circ$ and $\theta = 180^\circ$. From the recurrence formulae (II.7) (II.8)* it is easily established that whatever the order:

\begin{align*}
\text{for } \theta = 0^\circ & \quad \pi_n(1) = \tau_n(1) = \frac{1}{2} n(n+1) \\
\text{and} & \\
\text{for } \theta = 180^\circ & \quad -\pi_n(-1) = \tau_n(-1) = (-1)^n \frac{1}{2} n(n+1)
\end{align*}

(2.10)

(2.11)

Thus, as will be seen further, these relations will become useful where the interpretation of scattering at the particular angles is concerned, and also for calculating the extinction efficiency factor, (it reduces to a scattering efficiency factor in the case of non-absorbing particles - see appendix "definitions").

2.1.2 The Intensity Functions $i_1(\theta)$ and $i_2(\theta)$

Special case of angles $\theta = 0^\circ$ and $\theta = 180^\circ$

Combining (2.2) with (2.10), then (2.11), it is immediately seen that

\begin{align*}
S_1(0) &= S_2(0) = \frac{1}{2} \sum_{n=1}^{\infty} (2n+1) (a_n + b_n) \\
\text{and that} & \\
S_1(180) &= -S_2(180) = \frac{1}{2} \sum_{n=1}^{\infty} (-1)^n (2n+1) (b_n - a_n)
\end{align*}

(2.12)

(2.13)

* Established in Appendix II
Taking the square of the modulus to obtain the intensities, it is seen that both at 0° and 180° \(i_1 = i_2\). The consequences regarding polarization have been considered previously (Section 1.2).

**Arbitrary angle \(\theta\); evolution of the scattering function with size (\(m\) real).**

When \(\alpha\) is sufficiently small (\(< 0.2\) approximately), the series (2.2), expressing \(S_1\) and \(S_2\), reduce to the first term, \(\pi_1\) and \(\tau_1\), having the values given by (2.9A). On the other hand (cf. Appendix II) \(b_1\), numerically of the same size as \(a_2\), is negligible and we have again Rayleigh scattering with:

\[
i_1 = |S_1|^2 = \text{const.} \quad i_2 = |S_2|^2 = \text{const.} \times \cos^2 \theta.
\]

The vertical component is constant, the horizontal component behaving as \(\cos^2 \theta\) vanishes at \(\theta = 90^\circ\), the scattered light is totally polarized. The scattering functions for the two components and for the total intensity \(i_T = 1/2(i_1 + i_2)\) are symmetric. As \(\alpha\) increases, all the values increase but an asymmetry appears to be enhanced; the backscattering \((\theta > 90^\circ)\) is less important than the forward scattering \((\theta < 90^\circ)\), the minimum of \(i_T\) at 90° disappears. In contrast when \(\alpha\) attains the value 2.25 a new minimum of \(i_T\) appears at the rear \((180^\circ)\) and "migrates" towards the central part of the scattering function, while another minimum of \(i_T\) appears at 180° when \(\alpha\) goes beyond the value 4 (cf. figures in Appendix II). The same process is repeated when the parameter of size is increased; the undulations appear at the rear and become narrower. This results from taking into account higher orders of the functions \(\pi_n\), \(\tau_n\), since as \(\alpha\) increases the sequences of Mie coefficients converge all the more slowly towards zero. Thus for example when \(\alpha = 100\) \(\pi_n\) and \(\tau_n\) are introduced, the scattering function having about 100 undulations.

\[\text{This suggests the choice of a criterion:} \quad a_n \quad \text{and} \quad b_n \quad \text{are considered to be zero, thus stopping the computation at a given order, when the modulus of both coefficients becomes less than } 10^{-7} \quad \text{(cf. Appendix II).}\]
Apart from these undulations, the general tendency is towards enhancement of the asymmetry. If now the two intensity functions $i_1$ and $i_2$ are considered instead of $i_T$, the behaviour is analogous but ever more complicated. The degree of polarization $(i_1 - i_2)/(i_1 + i_2)$ also oscillates between +1 and -1 (the negative values correspond to the case when the horizontal vibration has a greater amplitude than the vertical).

Qualitatively this description remains valid as the index of refraction (real) varies within fairly broad limits. However, it should be noted that when the index comes very close to 1, the amplitude of the oscillations of $i_T$ is greatest, and in a general way the difference between the two components $i_1$ and $i_2$ is large, resulting in strong polarization. As the index departs from 1, there occurs "damping" of the undulations (and reduction of polarization) as indicated in Figure II. 2. In the opposite case for a given relative size $\alpha$ the number of undulations is larger, while the global asymmetry is less accentuated.

In what follows, these diverse points will be taken up in greater detail and will be interpreted using the computed numerical values.

Figure II. 2: Scattering functions normalized to zero degrees. For the upper group $\alpha = 10$, for the lower $\alpha = 5$. In both cases, starting from below, scattering functions for index of refraction 1.02, 1.05, 1.075, 1.10, 1.15, 1.20 are found. The ordinate axis is logarithmic.
2.2 NUMERICAL RESULTS AND INTERPRETATION

The computations of the scattering function using Mie theory have been performed for a certain number of cases, listed below. There is a double motivation for this preliminary calculation. On the one hand the published tables of Mie functions are very incomplete mostly where the indices of refraction and the particle size are concerned in the frame work of this study, while on the other hand the introduction into a computer of a large number of tabulated values, if they exist, results mostly in an excessively long procedure. For further application it is in the end more efficient to predict within the program the generation of useful scattering functions. In addition it is found indispensable to be able to chose the increments of the computation, mostly where the index and the size are concerned, rather than the scattering angle.

- The intensity functions $i_1(\theta)$ and $i_2(\theta)$, and consequently the total intensity function $i_I(\theta) = \frac{1}{2} (i_1 + i_2)$, and the degree of polarization

$$p = \frac{i_1(\theta) - i_2(\theta)}{i_1(\theta) + i_2(\theta)}$$

have been computed in $2^\circ$ increments from $0^\circ$ to $180^\circ$, (in exceptional cases a $1^\circ$ step has been used).

- For each index of refraction, the computation is repeated for 60 values of size, namely: $0.2(0.2)2.0, 2.0(0.5)5.0, 5.0(1.0)20.0, 20.0 (2.0)40.0, 40.0(5.0)100.0, 100.0(10.0)140.0, 140.0(20.0)200.0$, the numbers in parentheses indicating the increments of $\alpha$ used.

- Finally the whole group of computations above, is repeated 5 times for the following values of index of refraction; $1.02, 1.05, 1.075, 1.10, 1.15$.

- For each case, i.e., for each pair of values $\alpha$ and $n^*$, both the real and imaginary parts of $S(0)$ are computed, as well as the amplitude at $0^\circ$, and the scattering efficiency factor $Q$ (cf. Appendix I and Sec. 2.2.7).

* The notation $n$ (and no longer $m$) is now re-adopted for the index of refraction, ($m$ will denote the characteristic exponent to the distribution).
Figure II. 3
Plots of the functions $i(\theta) \alpha^{-4}$ for the indicated values of $\theta$. Each figure corresponds to a different index of refraction. The size parameter varies between 0.2 and 200. The figure for index 1.05 has been provided elsewhere (A. Morel, 1972 a).

Figure II. 4

Figure II. 5

Figure II. 6: Graphs of the same function but for $\theta=0^\circ$ and for various values of the index of refraction $n$. 
For reasons explained in Appendix II "computational procedure and computer application" the use of "double precision" (17 significant digits) is indispensable. However, it is the criterion adopted for series convergence, in ending the computation, which determines the precision; in the worst case (large values of $\alpha$) this results in 6 significant digits, whatever the actual value of the number (expressed in floating point).

### 2.2.1 The functions $i_T(\theta)\alpha^{-4}$

In order to compactly present these results, for each value of index of refraction the variations of the functions $i_T(\theta)\alpha^{-4}$ have been plotted for varying size parameters $\alpha$ from 0.2 to 200.0 (figures II.3 and II.5). On each of these figures, six curves are drawn corresponding to the following values of $\theta$: $0^\circ, 2^\circ, 10^\circ, 40^\circ, 90^\circ, 180^\circ$. For clarity the number of curves has been voluntarily curtailed; however the plots demonstrate, schematically at least, how the scattering function varies as size increases.

The choice of the function, more specifically of the $-4$ exponent of $\alpha$ is not arbitrary; as will be seen further (Sec. 2.2.8) it originates in the expression for diffraction, which constitutes a limiting expression for scattering at $0^\circ$ in the case of sufficiently large particles.

The following remarks can be made based on the figures:

a) For the smaller values of $\alpha$, the curves corresponding to symmetric scattering angles $0^\circ, 180^\circ$ and $40^\circ, 140^\circ$ overlap, while the curve for $90^\circ$ is located below them. All these curves in their initial linear portion have a slope of $+2$. Finally for a given value of $\alpha$ (e.g. 0.2) the values $i(\theta)\alpha^{-4}$ increase smoothly with index (cf. figure II.6, where the relative curves corresponding to $\theta=0$ but to various indices are grouped together).

b) As far as angle $0^\circ$ is concerned, the linear portion extends to higher values of the parameter $\alpha$. Figure II.6 also shows that it also corresponds to higher values of $\alpha$ as the index is closer to 1.0 (to a lesser degree a similar behaviour occurs for the curves at $20^\circ$).
c) Beyond the linear portion and after passing through a maximum
the relative curves at 0° demonstrate oscillations whose amplitude
decreases as \( \alpha \) increases. These oscillations occur around a fixed
limiting value of 0.25. (Figure II.6)

d) For angles other than 0°, the curves also pass through a maximum
beyond which the decrease is accompanied by complicated oscillations.*
This maximum appears at higher values of \( \alpha \) as \( \theta \) becomes smaller.
Comparison of the different figures indicates that the location of the
maxima is not practically dependent on the value of index. Thus
contrary to what has been seen for 0°, the corresponding value of \( \alpha \)
depends only on the angle and not on the index.

e) The mean slope of the various curves (\( \theta \neq 0 \)), neglecting the oscillations
is approximately -2.3 for indices of refraction 1.02, 1.05, 1.075
and then about -2.0 for the higher indices 1.10 and 1.15.

f) Finally, the relative curves at 180° are more or less an exception; the
oscillations are more erratic and as long as one can talk about a mean
slope, it is very small or zero, particularly at higher values of index.

These remarks have been made in view of the numerical re-
sults obtained from the mathematical formulation of Mie; they are
statements which by themselves have no interpretative value. The
Mie theory furnishes a rigorous solution covering all cases, from
that of very small particles where Rayleigh theory applies, up to large
enough particles where classical theories of geometrical optics and
diffraction apply. These theories, in fact strictly applicable at the limits
and only approximately in the intermediate region, are capable of more
easily providing the elements of interpretation, and ascribing to the
computational results a clearer physical meaning.

* The curves have been drawn by joining the computed points with
straight lines. The increment of \( \alpha \) in the computations permits only an
approximate demonstration of their behaviour without showing the fine
details.
Thus Rayleigh theory accounts for the first remark, while the Rayleigh-Gans theory permits elucidation of b) and d). The other remarks - only partially for point e) — receive a satisfactory explanation by invoking diffraction theory or geometrical optics, (reflection, refraction). These various interpretations are successively examined; their contribution resting in the possibility they offer of predicting the scattering functions, both for a given particle as well as for an ensemble of polydisperse particles.

2.2.2. The Rayleigh Scattering Domain

It has been seen previously (Sec. 2.1.2) that in the case of small values of the parameter $\alpha$ (e.g. 0.2) the Mie formulae giving $i_1(\theta)$ and $i_2(\theta)$ reduce to simple expressions, revealing an angular dependence which is that of Rayleigh scattering. The Rayleigh theory of a radiating dipole produces for $i_1$ and $i_2$ the expression

\[
\begin{align*}
    i_1 &= k^6 \frac{2}{p^2} \frac{1}{\cos^2 \theta} \\
    i_2 &= \frac{1}{\cos^2 \theta}
\end{align*}
\]

related to physical quantities which are the wavenumber $k = \frac{2\pi}{\lambda}$ and the polarisability of the particle $p^\ast$, having dimensions $L^3$. If a particle is an isotropic sphere of radius $r$, volume $V$ and index of refraction $n$, the polarisability is expressed by the Lorenz-Lorentz relation:

\[
p = \frac{3}{4\pi} \frac{n^2-1}{n^2+2} \frac{V}{r^3} \quad \text{or} \quad \frac{n^2-1}{n^2+2} \frac{1}{r^3} \quad \text{and setting} \quad \alpha = kr \text{ and } \frac{n^2-1}{n^2+2} = \Lambda,
\]

\[
\text{then} \quad i_1 = \alpha^6 \frac{\Lambda^2}{r^2}, \quad i_2 = \alpha^6 \frac{\Lambda^2 \cos^2 \theta}{r^2} \quad .
\]

* Actually an extension of Rayleigh theory, done partially by Rayleigh himself (1914) and developed by Gans (1925). (cf. H.C. van de Hulst, 1957).

** The polarizability is the induced moment $\vec{P}$ when the electric field $\vec{E}$ is unitary. The vectors $\vec{P}$ and $\vec{E}$ are parallel for an isotropic dielectric and in such a case $\vec{P}$ is a scalar; in the general case it is a tensor, $\vec{P}$ and $\vec{E}$ having different directions.
In the domain of applicability of Rayleigh theory ($\alpha \ll 1$) the quantities $i_T(\theta)\alpha^{-4}$ represented graphically are expressed by

$$i_T(\theta) \alpha^{-4} = \frac{1}{2}a^2 \Lambda^2 (1 + \cos^2 \theta).$$

Plotted as functions of $\alpha$ on logarithmic coordinates, they are represented by straight lines of +2 slope, whatever the value of $\theta$. The relative locations of these lines evidently explain the symmetry of the scattering function with respect to $90^\circ$, (cf. remark a)).

Finally the index of refraction enters to determine the magnitude of the phenomenon through $\Lambda^2$. For a given relative size $\alpha$ and for a given angle (for example $\theta=0^\circ$ which is the case drawn in Figure II.6) the scattered intensity is proportional to $\Lambda^2$. If an index close to 1.0 is considered, $\Lambda$ is approximately expressed as

$$\Lambda = \frac{2}{3} (n-1)$$  \hspace{1cm} (2.16)

Denoting by $i$ and $i'$ the intensities scattered by particles of the same size but of indices $n$ and $n'$ respectively, then

$$\frac{i'}{i} = \frac{\Lambda^2}{\Lambda'^2} = \left(\frac{n-1}{n'-1}\right)^2.$$  \hspace{1cm} (2.17)

The intensity functions thus attain increasingly larger values as the index is farther removed from 1.0. Thus as indicated by Figure II.6 for example, the intensities are multiplied by 25 as the index of refraction changes from 1.02 to 1.10.
2.2.3 The Rayleigh-Gans approximation

The condition for applicability of the Rayleigh theory

\[ \alpha = \frac{2\pi}{\lambda} \ll 1 \quad (2.18) \]

has the following physical hypothesis in mind: the particle must be
considered to be small enough so that the applied electric field is every-
where the same. In order to extend in a simple way this theory to a
larger particle and to apply it to each portion of the particle which is
considered an independent dipole, a first approximation must be made
that the electric field is the same over the surface of each of the various
portions comprising the particle. In other words, it is necessary that
both the amplitude and the phase of a wave reaching a given portion, not
be appreciably modified by the presence of the others. This implies that
the scattered energy is very weak and the phase retardation is negligible,
which can be expressed by the condition:

\[ \frac{2\pi r}{\lambda} |n-1| \ll 1 \]

\( \lambda \) being the wavelength in the medium surrounding the particle , \( n \) the
index relative to the external medium (the expression corresponds to the
maximum phase change, i.e., that of a ray crossing the particle along a
diameter). This condition is generally written as

\[ \rho \ll 1 \quad (2.19) \]

the parameter \( \rho \) combining relative size and relative index of refraction,
being equal to \( 2\alpha |n-1| \). This condition can be evidently satisfied even
by large values of \( \alpha \), as long as \( n \) only slightly differs from 1.

Case of Scattering at Zero Degrees

In a general way, the waves scattered from the various portions
of a particle are not simply additive since they interfere. However, the
initial hypothesis (that the particle by its physical presence does not modify the
state of the wave) suggests that in the particular direction of $0^\circ$, the scattered waves are necessarily in phase; the lengthening of the path of the incident wave is compensated exactly by a shortening of the path of the scattered wave and conversely, whatever the position of the various portions in space. In this particular case, in which strict additivity holds, the intensity expression given by Rayleigh theory holds even though $\alpha$ is not small, the formulae (2.15) remaining

$$i_T(0^\circ) = \alpha^6 \wedge^2$$

This suggests an explanation to remark b). Actually this is only a partial one since the linearity (on the log-log graphs) extends beyond the domain of applicability, so that the condition $\rho < 1$ ceases to be true. This extension justifies the name given by Penndorf (1960) of "extended Rayleigh region". The limit of this zone is fixed as will be seen later (Part II, Sec 2.2.8) by the value 4.09 of $\rho$; the corresponding value of $\rho$ thus being higher as the index approaches 1.

**General Case:**

For angles other than zero, the geometric compensation of the path differences does not hold; thus the amplitude of the scattered wave is obtained by multiplying the amplitude deduced from Rayleigh theory by a factor $F(\theta)$ describing the interferences which depends on $\theta$ (or on $\theta$ and $\varphi$ if the phenomenon does not have symmetry of revolution)

$$i_T(\theta) = \frac{i_1(\theta) + i_2(\theta)}{2} = \frac{1}{2} \alpha^6 \wedge^2 (1 + \cos^2 \theta) F(\theta)$$

(2.20)

$F(\theta)$ being 1 at $\theta = 0^\circ$ and less than 1 for all other angles. This function is obtained by integration extended over the whole particle; the differential element being composed of the elementary "troughs" representing constant differences in path. The integral can be expressed by the common functions if the particles have simple geometric shapes; in the case of a sphere:
(2.21)

\[ F(\theta) = (G(\bar{u}))^2 \]

with

\[ \bar{u} = 2\alpha \sin \theta/2 \]

and

\[ G(u) = \left(\frac{\pi}{2}\right)^{1/2} \frac{J_{3/2}(u)}{u^{3/2}} \]

where \( J_{3/2}(U) \) is a Bessel function of the first kind of order 3/2. This function multiplies separately the components \( i_1 \) and \( i_2 \); as a result the total polarization at \( \theta = 90^\circ \) characteristic of Rayleigh scattering is conserved. The scattering function becomes asymmetric by decreasing the values at large angles. The effect is all the more prominent as \( \alpha \) increases. As the first zero of the function \( J_{3/2} \) is reached, i.e. when the argument \( U \) attains the value 4.49, a minimum appears corresponding to extinction by interference: It appears at \( 180^\circ \) for \( \alpha = 2.25 \). As \( \alpha \) increases this minimum moves to smaller angles, while a second minimum appears corresponding to the second zero (when \( \alpha = 3.86 \)) and so forth. Minima are successively born in the rear and the scattering function presents a system of oscillations which gets continuously narrower while the predominance of forward scattering (at 0° and at neighboring angles) becomes more accentuated. The "shape" of scattering resembles the shape of Fraunhofer diffraction but the angular rays of the rings are different. As the particle size increases, there is a progressive transformation from one shape to another, this transition zone having been named "anomalous diffraction" in the Anglo-saxon literature. In any case the scattering function determined by the function \( G(U) \) is thus independent of the value of index of refraction (as long as \( n-1 \) remains small) and the argument of \( U \) depends only on \( \theta \) and \( \alpha \). If \( u_1, u_2, \ldots \) denote the successive values of the argument for which the function \( J_{3/2} \) vanishes, the equations

\[ 2\alpha \sin \theta/2 = u_1 (= 4.49) \]

\[ = u_2 (= 7.73) \ldots \]

are those of the loci of the successive minima in the plane \((\alpha, \theta)\). The curves corresponding to the first six minima are drawn on Figure II. 7.*

As an example it is seen from the figure that when \( \alpha = 5 \), the scattering function must have two minima at 55° and 110° and the indication of one at 180°; when \( \alpha = 10 \), there are 5 minima and an indication of a sixth.

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* Translator's Note: The Figure II. 7 has not been drawn very accurately.
Figure II. 7: Angular position of the first six minima according to the Rayleigh Gans theory (equation 2.23); the first two diffraction minima are shown with dashed lines (see further equations 2.24).

Figure II. 2 demonstrates that they clearly exist for the lowest value of index (1.02) but the system of minima migrates towards smaller angles as the index increases.

Returning to remark d) the relative curves at various angles $\theta$ become detached from that at $0^\circ$, this departure being due to the appearance of the first minimum at the angle $\theta$ under consideration (first equation 2.23); the smaller the angle $\theta$, the larger the corresponding value of $\alpha$, independently of the index of refraction, since it does not appear in the equation. The following table (first line) presents the theoretical values of $\alpha$ for which this minimum appears at the indicated angles $\theta$:

<table>
<thead>
<tr>
<th>$\theta$ (°)</th>
<th>180°</th>
<th>140°</th>
<th>90°</th>
<th>40°</th>
<th>20°</th>
<th>10°</th>
<th>2°</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rayleigh Gans</td>
<td>2,24</td>
<td>2,42</td>
<td>3,17</td>
<td>6,65</td>
<td>12,9</td>
<td>25,8</td>
<td>(128)</td>
</tr>
<tr>
<td>Diffraction</td>
<td>2.04</td>
<td>3,17</td>
<td>3,83</td>
<td>5,95</td>
<td>11,9</td>
<td>22,0</td>
<td>109</td>
</tr>
</tbody>
</table>
2.2.4 Approximation by diffraction theory

It must be noted that whenever the minimum is located at $10^\circ$, i.e., whenever $\alpha = 25.8$, the value of $\rho$ is 1 if the index is 1.02, but $\rho$ becomes 5 if $n=1.10$, in other words it is no longer justifiable to use the Rayleigh-Gans theory, and even less so for an angle of $2^\circ$. For these larger sizes, diffraction theory furnishes an approximation in computing the angular distribution of intensities, which in the case of a diffracting circular aperture is expressed by

$$f'(b) = \frac{2 J_1(U)}{U}, \quad (2.24)$$

with $U=\alpha \sin \theta$. The first zero of the Bessel function $J_1(U)$ is obtained when the argument $U$ becomes 3.83. This permits computation of the values which the size parameter $\alpha$ must have so that the angular ray of the first dark ring attain the values $2^\circ, 10^\circ, \ldots$; the second line of the previous table presents these values. This approximation by diffraction theory is more justifiable than that of the Rayleigh-Gans theory in explaining the shape of the scattering function in the small angle region*, and the same holds for the other angles if the index is far from 1. This in particular explains the increase in the number and the density of the oscillations in the scattering function when the index increases (Figure II.2), as can be anticipated from Figure II.7.

In conclusion it must be however noted that the position of the first minimum for a given angle $\theta$ corresponds to values of $\alpha$ derived from one or the other list, depending on the approximation used; but these values are sufficiently close to account for remark d.

---

* The diffraction minimum at $\alpha=109$ appears in all the relative curves for $2^\circ$ in Figures II.3 to II.5. However other minima appear ahead of it (except when $n=1.02$); they correspond to minima observed at $0^\circ$ or as will be seen later to those of the efficiency factor $Q$. This is explained as the result of constructive or destructive interference between the wave traversing the particle (refracted wave) and the diffracted wave formed by the central lobe.
2.2.5 The role of refraction and reflection

Independently of diffraction, the geometric optics approach takes into consideration reflection and refraction: the combination of three phenomena and of the possible interferences between three types of waves totally account for scattering. The function $F'(\theta)$ may attain the value of zero, and the minima may be produced by total extinction if the light scattered by the interplay of reflections and refractions is not added to the diffracted light. The importance of energy scattered in this manner is determined by the value of the Fresnel reflection coefficients and increases as the particle becomes more refractive; the minima are then attenuated (cf. figure II.2), the general asymmetry diminishes (Figures II.3 to II.5 and remark e)), but in contrast the backscattering (for angles close to $180^\circ$) becomes more accentuated (remark f). It must be noted that the total extinction which implies the vanishing of $F(\theta)$ in the Rayleigh-Gans theory does not hold any longer. The minima in this region are equally strongly accentuated if the index is close to 1, and they are attenuated when it departs from this value. The preceding physical interpretation, is only used here for the sake of continuity: the relative dimensions of the particle as compared to the wavelength no longer permit rigorous discussion in terms of reflection and refraction.

Figure II.8: Angular position of the "rainbow" according to the index of refraction.
Finally the application of geometric optics laws permits explanation of the rainbow (admittedly Descartes himself provided the correct explanation) which is due to a concentration of energy following an internal reflection (or two for a "second" rainbow) within the drop. The corresponding emergence angle (which is that of minimum deviation for the first rainbow) has the value of 138° for water droplets. Repeating the same calculation with other values of index of refraction, shows that the rainbow is shifted to smaller angles as the index approaches 1.0 (Figure II.8). The figures of Appendix II show that a stable maximum clearly appears (at about 75° for index 1.075) when the size is sufficiently large; other scattering functions of particle ensembles clearly demonstrate this phenomenon, in the same angular region, taking into account the value of the index of refraction (See Fig. II.17 and in Part III, Figures III.2, III.7, III.12).

2.2.6 General remark on the asymmetry of the scattering function.

To summarize, for the previously stated reasons, each one of the functions $i(\theta)\alpha^{-4}$ passes through a maximum for a practically fixed value of $\alpha$, whatever the index. In contrast the maximum through which pass the relative curves for 0° shifts to a larger value of $\alpha$ as the index approaches 1.0. Consequently the departure between the curve for 0° and the others, which determines the global asymmetry, is all the greater as the index is closer to 1.0. As an example, when $\alpha=200$ the scattering function covers 8 orders of magnitude for $n=1.02$ but only 5 or 6 when $n=1.15$ (cf. figure II.3 and II.5). This remark will attain its importance where prediction of the result of weighted additions of scattering functions is involved.

2.2.7 Total scattering coefficient. Efficiency factor

The efficiency factor for scattering $Q_{\text{scat}}^*$ is defined as the ratio of the effective cross-section for scattering to the particle geometrical cross-section, $(\pi r^2)$ for a spherical particle of radius $r), or equally well, as the ratio of the total scattered flux to that incident on the particle, i.e. on the section of area $\pi r^2$, for a sphere. These different definitions are given more precisely in Appendix I. Similarly an efficiency factor for attenuation is defined which does not differ from the previous except if the particle is absorbing, in which case one obtains

*Translator's Note: The author uses $Q_{\text{diff}}$ for $Q_{\text{scat}}$ since in French the word for scattering is "diffusion". However here I will use $Q_{\text{scat}}$ for convenience and simplicity.
\[ Q_{\text{attenuation}} = Q_{\text{absorption}} + Q_{\text{scattering}} \]

This relative attenuation factor is related to the amplitude \( S(0) \) by a relation established in particular by VandeHulst (1949, 1957) (and known by the name of "extinction formula"):

\[
Q_{\text{att.}} = \frac{4}{\alpha^2} \operatorname{Re} \left\{ S(0) \right\}, \quad (2.25)
\]

with \( S_1(0) = S_2(0) \), which is expressed through (2.12):

\[
Q_{\text{att.}} = \frac{2}{\alpha^2} \sum_{n=1}^{\infty} (2n+1) \operatorname{Re} \{ a_n + b_n \} \quad (2.26)
\]

The integral over \( 4\pi \) steradians producing the relative efficiency factor for scattering is (cf. Appendix I):

\[
Q_{\text{scat.}} = \frac{1}{2\pi} \int_{-\pi}^{\pi} \left( i_1 (\theta, \phi) + i_2 (\theta, \phi) \right) \sin \theta \, d\theta \quad (2.27)
\]

which for a geometry of revolution:

\[
Q_{\text{scat.}} = \frac{1}{\alpha^2} \int_{0}^{\pi} \left( i_1 (\theta) + i_2 (\theta) \right) \sin \theta \, d\theta \quad (2.27)
\]

It has been shown by Debye (1909) that this integral carried to a series finally reduces to (see Van de Hulst p. 128):

\[
Q_{\text{scat.}} = \frac{2}{\alpha^2} \sum_{n=1}^{\infty} (2n+1) \left( |a_n|^2 + |b_n|^2 \right) \quad (2.28)
\]

where \( |a_n|^2, |b_n|^2 \) are the squares of the moduli of the Mie coefficients.

When the index of refraction is real, it has been remarked (Fig. II.1) that the images of \( a_n \) and \( b_n \) on the complex plane lie on a circle of radius \( 1/2 \). Geometrically, for any \( a_n \) or \( b_n \):

\[
\frac{|a_n|}{\operatorname{Re} (a_n)} = \frac{1}{|a_n|}, \quad \text{or} \quad |a_n|^2 = \operatorname{Re} (a_n).
\]

The real part of \( a_n \) is numerically equal to the square of its modulus. The equations (2.26) and (2.28) are thus mathematically identical, which is necessary in the absence of absorption, since the attenuation is only due to scattering.
In what follows there will be no question of absorption and the factor $Q_{\text{scat}}$, equal to $Q_{\text{atten}}$, will simply be denoted by $Q$.

Following equation (2.25), the variations of this factor with the size parameter $\alpha$ will be studied through the evolution of the function $S(0)\alpha^{-2}$ in the complex plane. This quantity has been computed for all the sizes and indices of refraction already mentioned; as is shown for example in Figure II.9, the function $S(0)\alpha^{-2}$ is represented on the complex plane by a spiral which revolves around a point $\text{Im}=0$, $\text{Re}=0.5$, as the parameter $\alpha$ increases. In contrast to the example given by Van de Hulst relative to index 1.33 (Van de Hulst, 1957, page 176) the spiral is more regular which is characteristic of indices closer to 1.0; oscillations, reversions and finally retrograde curls appear for large values of $\alpha$ as soon as the index appreciably departs from 1.0. The rate of spiraling depends on the index; for example as $\alpha$ increases from 0.2 to 200.0, one turn in the spiral occurs when $n=1.02$ but five when $n=1.10$. In fact the rates of spiraling in the various cases become identical if instead of $\alpha$ the parameter $\rho = 2\alpha(n-1)$ is used (particular values of $\rho$ for various angles of rotation are indicated in the figure). The factor $Q$ is equal to four times the real part of the function $S(0)\alpha^{-2}$; it passes through a first maximum for $\rho$ close to 4, then oscillates around the value 2, the amplitude of the oscillations becoming continuously smaller as the spiral revolves inwards. The limiting value $2^*$ is that which produces diffraction (see immediately later), whose laws are applicable if the particle is sufficiently large $^{**}$.

---

* It does not appear that the convergence of the series (2.28) or (2.26) towards 2 has been proven mathematically.

$^{**}$ The fact that a particle scatters twice the energy which it can geometrically intercept is known by the term "extinction paradox" (or "Babinet's paradox"). This paradox is not apparent and derives from the initial hypothesis that the distance of observation is very large compared to the particle size. Van de Hulst pleasantly illustrates this phenomenon taking as his example a flower pot on a window ledge projecting a shadow, for which the coefficient is 1, while it is 2 for a meteorite of the same size which intercepts light originating from a distant star.
2.2.8 Efficiency factor and intensity at 0°. Limiting values (diffraction)

The intensity function is obtained by taking the squared modulus of the amplitude (2.1): \( S(0)\alpha^{-2} \) tends to 0.5 as \( \alpha \) increases, \( i(0)\alpha^{-4} \) going to 0.25. This has been the object of remark c, which may be simply interpreted: the diffracted intensity in the direction 0° (central spot) is proportional to the square of the surface of the diffracting aperture and the amplitudes of the diffracted wave in any direction are

\[
S_1(\theta) = S_2(\theta) = \frac{a^2}{2} \frac{2J_1(\alpha \sin \theta)}{\alpha \sin \theta}.
\]

As \( \theta \) tends towards 0, \( J_1(U)/U \) tends towards 1/2 and \( i(0) = |S(0)|^2 \) actually goes to \( \alpha^4/4 \). Before converging to their respective limiting values 0.25 and 2.0, the oscillations of the functions \( i(0)\alpha^{-4} = |S(0)|^2\alpha^{-4} \) and \( Q = \text{Re}\{S(0)|^2\alpha^{-2}\} \) are coincident; the maxima and minima of the two functions occur for the same values of \( \alpha \) which correspond in fact to fixed
values of \( \rho \), marked approximately on the spiral. More precisely (computed with equation 2.29 below, applicable if the index is close to 1) these values are

\[
\begin{align*}
\text{1st maximum} & = 4.09 \\
\text{2nd maximum} & = 10.79 \\
\text{1st minimum} & = 7.63 \\
\text{2nd minimum} & = 14.00
\end{align*}
\]

2.2.9 Van de Hulst's approximation

It has been demonstrated by Van de Hulst (1946, 1957) that the efficiency factor may be expressed simply as a function of the parameter \( \rho \) by:

\[
Q = 2 - 4\rho^{-1}\sin\rho + 4\rho^{-2}(1 - \cos\rho)
\]

(2.29)

as long as the index of refraction is very close to 1 (\( 1 \pm \varepsilon \)); in fact even if it differs considerably, this expression remains mostly valid. Figure II.10 demonstrates that the curve obtained by equation 2.29 constitutes a good approximation, even when the index has the value 1.15, the representative points having been computed by the exact expression (2.26). When \( n \) is increased, on top of the oscillations of period 2\( \pi \), many systems of small oscillations are superimposed which correspond to the ondulations or the curls on the spiral. This expression for the efficiency factor \( Q \) is an approximation valid in the whole Mie domain, whatever the value of \( \rho \), the only condition being that \( n - 1 \) be small.

In the limit, \( Q \) approaches 2 as \( \rho \) increases. On the contrary as \( \rho \) goes to zero, i.e. as the particle size decreases, expansion of (2.29) gives

\[
Q_{\rho \rightarrow 0} = (1/2) \rho^2 - (1/36) \rho^4 + \ldots \tag{2.30}
\]

This formula agrees as will be briefly seen below, with the formula resulting from the Rayleigh-Gans theory which also assumes \( n \) close to 1; but it does not agree with the formula corresponding to the Rayleigh theory whose more restrictive condition \( \alpha \ll 1 \) must be respected.
Figure II.10:
Efficiency factor $Q$ plotted on a logarithmic scale as a function of the parameter $\rho = 2\alpha (n-1)$, for two values of index of refraction.

Figure II.11:
Efficiency factor $Q$ in the region of small values of $\rho$ (logarithmic scales) for various values of index of refraction.
2.2.10 Efficiency factor in the Rayleigh and Rayleigh-Gans domains.

Combining equations 2.27 with 2.15 giving the intensity functions in the case of Rayleigh scattering:

\[ q = \frac{1}{a^2} \int_0^\pi a^6 \cdot \lambda^2 \cdot (1 + \cos^2 \theta) \sin \theta \cdot d \theta \quad (2.31) \]

the integral of the function of \( \theta \) having the value 8/3; thus

\[ q = \frac{8}{3} a^4 \cdot \lambda^2 \quad (2.32) \]

an expression in which we find again the \( \lambda^{-4} \) law, but which is in contradiction to the \( \lambda^{-2} \) law which resulted from the development of (2.30).

In the case where \( n \) is assumed close to 1, the Lorentz term is replaced according to (2.16) by \( 2/3(n-1) \), and so

\[ q = \frac{32}{27} s^4 (n-1)^2 \quad (2.33) \]

In the domain where the Rayleigh-Gans approximation is valid, \( \theta \) the intensity functions are multiplied by the factor \( F(\theta) = G^2(2a \sin \frac{\theta}{2}) \) which enters the integral. With \( \Lambda \) replaced by the preceding approximate expression, one obtains

\[ q = \frac{4}{9} (n-1)^2 a^4 \int_0^\pi a^2 (2 \sin \frac{\theta}{2}) (1 + \cos^2 \theta) \sin \theta \cdot d \theta \quad (2.34) \]

the value of the definite integral now depending on \( a \); if \( a \) goes to zero, the previous expression (2.33) is obtained which is valid when \( n \) is close to 1; in contrast if \( a \) is large, it has been shown that the value of the integral is \( 9/2 a^{-2} \) and thus \( Q = 2(n-1)a^2 \) which corresponds well to the limiting expression (2.30) of the Van de Hulst approximation.

As a practical conclusion, the difference between the exact values of \( Q \) from (2.28) and the approximate values from (2.29) is very small in general as has been stated previously, except in the case where \( \rho \) is small. For given values of \( \rho \) it is very important that \( (n-1) \) be far from zero. Figure II.11 demonstrates that for values of \( \rho \) less than 0.5 it is necessary to use the exact expression (2.28).
3. SCATTERING BY AN ENSEMBLE OF POLYDISPERSE PARTICLES.

The expression "polydisperse ensemble" is taken in the general sense in which it is understood. It designates an ensemble of particles of similar shape (spherical perhaps) and of the same nature (thus of the same index of refraction) which differ among themselves only by size. Their number varies with size according to some distribution law.

Initially (Section 3.1) the formulae permitting the computation of the scattering properties of such an ensemble are presented. Then the predictions that may be made concerning the results of the computations are examined (Section 3.2) in the case where a power law for the distribution is adopted. This leads to questions on the significance of the computations, considering that they have been performed by setting mathematical limits whose physical significance is not clear. Finally for comparison with the power law the case of other distributions is considered.

3.1 COMPUTATION OF THE SCATTERING PROPERTIES.

The particle population is characterized by a distribution function established as a function of a geometric parameter characterizing size; in what follows the parameter $\alpha$ is used, i.e. the relative size $2\pi r/\lambda$, since it appeared in the scattering computation and also to retain generality (for applications, the radii of the presumed spherical particles remaining constant, the variation of wavelength is considered as a variation inversely proportional to $\alpha$). The distribution function $F(\alpha)$ corresponds to a frequency distribution or a probability of appearance; if the total population is comprised of $N$ particles whose sizes vary from zero to infinity (or from a minimum size $\alpha_m$ to a maximum size $\alpha_M$), the quantity

$$\frac{1}{N} \int_{\alpha_1}^{\alpha_2} F(\alpha) \, d\alpha$$
is the relative probability of particles with sizes between $\alpha_1$ and $\alpha_2$; the function $F(\alpha)$ is considered continuous and integrable in the interval $0$ to $\infty$ or $\alpha_m$ to $\alpha_M$.

The additivity of the scattered intensities by randomly dispersed particles makes the computation simple from a formal point of view; it is understood that the integrations presented below are in practice replaced by summations, the size increment $d\alpha$ being that which was determined by the previous computations of individual scattering functions (cf. Section 2.2, part II).

3.1.1 Scattering function for a given number of particles

For each angle, the intensity functions $i_1(\theta)$ and $i_2(\theta)$ for the particle ensemble are given by the integral of the products $i_1(\theta, \alpha)F(\alpha)$ and $i_2(\theta, \alpha)F(\alpha)$. By normalizing with the integral of the function $F(\alpha)$ between the same limits, the computation becomes independent of the total number of particles, otherwise it would be dependent in each case on the particular distribution and the limits used. Thus the computation produces

$$i_1(\theta) = \frac{\int_{\alpha_m}^{\alpha_M} i_1(\theta, \alpha) F(\alpha) d\alpha}{\int_{\alpha_m}^{\alpha_M} F(\alpha) d\alpha} \quad \text{(3.1)}$$

and similarly for $i_2(\theta)$. The total intensity function $i_T(\theta) = \frac{1}{2}(i_1(\theta)+i_2(\theta))$ and the degree of polarization $(i_1(\theta)-i_2(\theta))/(i_1(\theta)+i_2(\theta))$ are then deduced. For an application, if $N$ is the total number of particles in unit volume, the angular scattering coefficient is simply obtained by computing (cf. Appendix I, "definitions"):

$$\beta(\theta) = N \frac{\lambda^2}{4 \pi} i_T(\theta)$$

The adoption of any given value for $\lambda$ is reflected on the limits $\alpha_m$ and $\alpha_M$ used.

3.1.2 Mean efficiency factor.

For an ensemble of particles, it is defined as the ratio of the sum of the effective cross-sections to the sum of the areas of the geometric sections. For the spherical particles under consideration, use of the parameter $\alpha$ produces
\[ Q = \frac{\int_{a_m}^{a_0} F(a) \ a^2 \ Q(a) \ da}{\int_{a_m}^{a_0} F(a) \ a^2 \ da} \]

(3.2)

The parameter \( \rho = \varphi(n-1) \) may equally well be used, combining both a relative size and relative index of refraction. This is advantageous when the index is close enough to 1.0 since Van de Hulst's formula (2.29) is applicable; his expression is sufficiently simple for the integral in the numerator to be numerically computed with a very small step in \( \rho \).

3.1.3 Normalized scattering function.

In order to compare the results of varying the distribution law and its limits, it is less practical to fix the total number of particles (as is done in equation 3.1) than to fix, on the contrary, the total scattering coefficient. This corresponds to examining particles whose number and distribution vary but which present the same global scattering effect, for convenience unitary scattering; the normalized scattering coefficients (cf. Appendix I, "definitions") are computed by

\[ \tilde{\beta}_1(\theta) = \frac{1}{n} \frac{\int_{a_m}^{a_0} F(a) \ i_1(\theta, a) \ da}{\int_{a_m}^{a_0} F(a) \ Q(a) \ a^2 \ da} \]

(3.3)

and correspondingly for \( \tilde{\beta}_2(\theta) \); \( \tilde{\beta}(\theta) \) is thus computed as the mean \( \frac{1}{2} (\tilde{\beta}_1(\theta) + \tilde{\beta}_2(\theta)) \). The computation of \( \bar{Q} \) being done simultaneously (3.1.2), the inverse of \( \bar{Q} \) provides the total area of the geometric sections of the particles, necessary to produce this unitary total scattering.

For example, if \( \bar{Q} = 0.5 \), it is necessary that the total cross-sectional area of the particles present in \( 1 \text{m}^3 \) be \( 2 \text{m}^2 \), in order that the total scattering coefficient \( \beta \) be \( 1 \text{m}^{-1} \).

3.2 PREDICTIONS ON THE RESULT OF THE COMPUTATIONS

It is possible to base these on the following two points:
- the evolution of the shape of the Mie scattering functions for increasing particle size is represented in its main characteristics by graphs of the functions \( i(\theta) \propto \alpha^{-4} \); the previous remarks concerning the varying slopes of the curves for various angles permit such predictions.
- the distribution of particles according to size is assumed to follow a Junge law* which is expressed by a power function $F(\alpha) = \text{const.} \cdot \alpha^{-m}$.

3.2.1 Conditions of convergence: influence of the upper limit on the scattering function.

Taking into account the form given to $F(\alpha)$, the functions $i_{\text{T}}(\theta) \alpha^{-4}$ already presented correspond precisely to distributions of $\alpha^{-4}$ and the integration of these functions is that appearing in the numerator of expressions 3.1 or 3.3. It provides (with the normalization) the scattering function for the assumed population (characterized by a $\alpha^{-4}$ law). It is immediately evident that all the integrals for the various angles converge since the mean slopes of the curves are close to -2.3 (cf. Section 2.2.1, e), except for the relative integral for $0^\circ$ angle for which the mean slope of the curve is zero. Put in another way, beyond a certain limit, taking into consideration larger and larger particles does not modify the resulting scattering function except for $0^\circ$ angle and for the immediately neighboring scattering angles where the intensity continues to increase. This fact illustrated by Figure III.2 (Part III) where the upper limit is taken from 50 to 200 without any significant change of the scattering function except at $0^\circ$, and also near $175^\circ - 180^\circ$. The same is not true however if the upper limit becomes less than 50 (A. Morel, 1972a, fig. 2).

In a general way, the functions $i(\theta)$ increase with size according to a $\alpha^{4+p}$ law where $p$ is the mean slope shown in graphs (log-log) such as II. 3; to recapitulate (cf. Section 2.2.1) these slopes have the following values:

*It will be seen later that functions of this type represent quite well the distributions of marine particles actually observed. In any case more complex distributions can be decomposed and approximated in each region by such shapes, at least theoretically.
\[ p = +2 \quad \text{for any } \theta, \text{ if } \alpha \text{ is small} \]
\[ \quad \text{for } \theta = 0^\circ, \text{ also when } \alpha \text{ is not small, under the} \]
\[ \quad \text{condition that } \rho = 2\alpha(n-1) \text{ be less than 4;} \]
\[ p = 0 \quad \text{for } \theta = 0^\circ \text{ when } \alpha \text{ is large } (\rho > 4) \]
\[ p \not\leq -2,3 \text{(or } -2) \quad \text{for } \theta \neq 0^\circ \text{ when } \alpha \text{ is large;} \]
\[ -2 < p < 1 \text{ for } \theta = 180^\circ, \text{ when } \alpha \text{ is large; the value of the slope depends} \]
\[ \quad \text{in fact on the index (it is however difficult to determine} \]
\[ \quad \text{a mean value).} \]

The functions \( F(\alpha) I_{\text{T}}(\alpha, \theta) \) are functions of \( \alpha^{4+p-m} \) if \( -m \) is the
\[ \text{exponent of the distribution; the integrals to be computed are thus functions} \]
\[ \text{whose exponent is } 5+p-m. \text{ These integrals converge absolutely if:} \]
\[ 5+p-m < 0; \quad \text{(3.4)} \]

the equality to zero does not lead to convergence, since the integral is a
logarithm.

If progressive values of the integrals are plotted as functions
\[ \text{of } \alpha : \]
\[ \int_{\epsilon}^{\alpha} F(\alpha) I_{\text{T}}(\theta, \alpha) \, d\alpha, \quad \text{(3.5)} \]

for various angles \( \theta \) (Fig. II.12) it can be determined that:

a) when \( \alpha \) is small, whatever the value of \( \theta \), all the curves grow with
\[ \quad \text{a slope* of 7-m (being 3.5 and 2 respectively for the two cases pre-} \]
\[ \quad \text{sented);} \]

b) for increasing values of \( \alpha \) as \( \theta \) decreases (i.e. for values of \( \alpha \)
\[ \quad \text{corresponding to the maximum of the } i(\theta)(\alpha)^{-4} \text{ curves, cf. Section 2.2.1 d} \]
\[ \quad \text{and 2.2.3), the integrals converge asymptotically and the curves pre-} \]
\[ \quad \text{sent an asymptotic plateau (when the condition 3.4 is respected);} \]

\* The slight undulations which appear on the ascending portions of the
curves (in the vicinity of \( \alpha=2 \) for example) are artifacts of the computa-
tions due to the change of increment \( (d\alpha) \) in computing the integral. This
increment is in fact determined by the preliminary computations of the
scattering functions (cf. Section 2.2).
c) for the angle 0° (where p=0) there is no plateau except if m > 5, otherwise the integral continues to increase with a slope 5-m (having a slope of 1.5 for the upper figure, and a slope going to zero for the lower figure -- the logarithmic branch of the curve -- which constitutes a limiting case);

d) for the 180° angle the evolution is more complicated by the very fact that the mean slope p has the tendency to vanish as χ grows (all the more so as the index is larger, which enhances reflection, (cf. Section 2.2.5); in general, beyond the fault portion, the partial integral resumes its growth.

In conclusion, since the separation among the various plateaus characterizes the resulting scattering function, the following predictions can be made:

- if all the curves present a plateau (when m > 5), the departure among the final values no longer changes, and thus the scattering function is no longer modified when the upper limit of integration is continuously raised.

- if 2.7 < m < 5, the conclusion remains the same, except where the 0° angle is concerned, in other words taking into consideration larger and larger particles does not change the shape of the scattering function except for very small angles (and also towards 180°) where the growth persists.

- if m < 2.7 no mathematical limit exists and the computation does not have meaning unless there exists a limit having physical reality.
Figure II.12: Progressive values of the integral as a function of the upper limit. The exponent of the distribution has the value -3.5 and -5 in each case, the index of refraction remaining in both cases 1.05. The lower limit for the computations is 0.2 but the curves have been drawn after the first step in the integration, i.e. starting at $\alpha = 0.4$. The upper limit is 200.
3.2.2 The role of small particles: the influence of the lower size limit on the scattering function

It will be seen that under certain conditions which will be determined, the computation retains its significance, even when the upper limit is set arbitrarily. An analogous problem presents itself in relation to the lower size limit. It is physically unknown and for the computation the lower size limit here again cannot be but arbitrary. It is necessary therefore to predict what happens to the scattering function when the very small particles are ignored. Theoretically the computation may be conducted, starting from a hypothetical lower limit determined by particles of "zero" dimensions. In order to determine the effect of truncation* if the integration is begun from a finite non-zero limit, two cases must be distinguished:

Case in which the neglected particles belong to the Rayleigh domain:

This is the case in which the integration is performed beginning from a lower limit \( \alpha = \epsilon \), at most equal to 1. An unfavorable hypothesis consists in assuming that the unknown particles continue to be distributed according to the same \( \alpha^{-m} \) law; this hypothesis leads to continuously increasing numbers of particles (and to an infinite number for size "zero"). The same scattering law applies to all these particles (cf. Section 2.2.2) being:

\[
\begin{bmatrix}
    i_1(\theta) \\
    i_2(\theta)
\end{bmatrix} = c^6 \kappa^2 \begin{bmatrix}
    1 \\
    \cos^2 \theta
\end{bmatrix}
\]

For each value of the angle \( \theta \), the integration over the domain under consideration - for \( i_T = \frac{1}{2} (i_1 + i_2) \) - is:

\[
1/2 \int_0^\epsilon \kappa^2 \alpha^{6-m} (1 + \cos^2 \theta) \, d\alpha.
\]  

(3.6)

This ensemble of particles thus presents a Rayleigh scattering function, expressed by:

\[
i_T(\theta) = \frac{1}{2} \kappa^2 \left( 1 + \cos^2 \theta \right) \frac{1}{\Gamma - m} \epsilon^{\Gamma - m},
\]  

(3.7)

* Since the presence of the small particles cannot be observed, the effect of knowing the distribution law can only be deduced, through continuity.
i.e. a scattering function of finite magnitude under the condition that the inequality

\[ 7 - m > 0 \quad (3.8) \]

is respected, otherwise one obtains \( i_T = \infty \). In other words, in the former case when truncation is made, the neglected quantities \( i_T(\theta) \) are finite and the resulting error can be computed. The error is infinite in the latter case and the computation has no significance, unless a non-zero physical limit exists.

In the former case, the neglected terms vary with angle according to the Rayleigh law (i.e. in the ratio 1 to 2). If they are compared to a very asymmetric scattering function obtained when the upper limit is greater than 200, it is found that the potential error is greater for angles equal to or greater than 90°. A relative value of this error is obtained by forming the ratio of the respective integrals over 0 to 0.2 and over 0.2 to 200. In examining the two examples illustrated in Fig. II.12, the resulting truncation error (to 0.2) is less than 0.1% if the value of the exponent is -3.5, and of the order of 1% if this value is -5 (this holds for the angles 90°, 140° and 180°; it is evidently less for the smaller angles). This order of magnitude remains valid for the other cases. In Appendix II are presented values calculated by means of (3.7) for various indices of refraction and for exponents varying from -3 to -5. These values are always negligible compared to those provided by the integration in the size interval 0.2 - 200.

In conclusion it must be accepted that the lower limit of this interval has been set to a sufficiently small value. The scattering function obtained in this way is significant because the particles whose size is less than this lower limit do not have any essential influence (on the condition that their distribution law obeys the condition 3.8).

Case where the truncation enters for larger sizes.

The effect may be directly observed on figures such as II.12. In particular if the value of the lower limit is such that the curve for any particular angle has reached its plateau then the effect is important; for such an angle the neglected quantities may become greater than
those retained in the integration and the final value (for $\alpha = 200$) is consequently lowered. The plateaus occur at larger values of $\alpha$ the smaller the angle $\theta$ is (cf. Section 3.2.1, b); so this lowering affects initially the value for angle $180^\circ$, then for $140^\circ$ etc. The asymmetry of the final scattering function is all the more pronounced the greater the lower limit $\alpha_m$ becomes:

if $\alpha_m = 1$ it appears that the values at $90^\circ$, $140^\circ$ and $180^\circ$ are reduced by 10% to 20%, compared to those obtained previously with $\alpha_m = 0.2$.

if $\alpha_m = 2$ the reductions at the same angles are about 30% to 50% while at $40^\circ$ a slight departure (-10%) appears.

if $\alpha_m = 10$ the effect is more accentuated and in addition extends over a greater angular domain; the departure is significant beginning at a $20^\circ$ angle.

Figure II.13 presents this enhancement of the global asymmetry by reduction of the values at large angles, the values at small angles ($0^\circ$ and $2^\circ$) not being affected.

Figure II.13: Influence of the lower limit; the dotted curves are taken from Fig. II.12 (upper plot, exponent $-3.5$). The two other groups of curves are obtained when the lower limit of integration is successively 2 & then 10. The three unidentified curves are for the $180^\circ$ angle; they are characterized by a rapid growth for the larger values of $\alpha$. 
This figure recasts the upper graph of Fig. II.12; in addition it represents the curves obtained for lower limits of 2 and 10, all other things being equal. This evolution is more completely represented by the full scattering function plots presented later (Figure III.12, Part III).

3.2.3. Total scattering; influence of the limits on the computation.

As previously the question arises of determining whether total scattering has any meaning when only a portion of the particle population is considered; the upper and lower limits have been determined by the needs of the computation and do not correspond to a physical reality. The argument which is analogous to that presented earlier (3.2.1 and 3.2.2) must be applied now to the integral:

$$\int_{a}^{a_{M}} F(a) \alpha^{2} Q(a) \, da$$

(3.9)

which, to a very close degree, expresses the effective total cross-section of particles distributed according to the law $F(\alpha)$ within the minimum and maximum size range of $\alpha_{m}$ to $\alpha_{M}$. It suffices to recall that the efficiency factor $Q$ varies as $\alpha^{4}$ in the Rayleigh domain (equation 2.32), as $\alpha^{2}$ in the Rayleigh Gans region (equation 2.35) and is finally independent of $\alpha$ on the average in the Mie domain and in the diffraction region (section 2.2.8).

Without recalling in detail, it is seen that as far as the upper limit is concerned (in the Mie domain), the total scattering reaches a limit if the integral of $F(\alpha)\alpha^{2}$ converges, i.e. if the total surface, presented by the particles, itself tends to a limit. This implies that the following condition must hold

$$m > 3$$

(3.10)

if the distribution is, as previously, expressible by $F(\alpha) = C\alpha^{-m}$. The total surface and therefore also the scattering increases as the logarithm of the upper limit $\alpha_{m}$ when $m=3$. As far as the lower limit is concerned (in the Rayleigh domain), the integral between 0 and $\alpha$ corresponding to neglected particles, is taken over an expression of the
form $\alpha^6 m$ just as in the scattering function computation. Thus the same condition (3.8): $m < 7$ assures for the integral a finite value. The estimates for a variety of exponents and indices of refraction show that the value of this integral between the limits 0 and 0.2, remains absolutely negligible compared to the value of the integral for the particles that are taken into account (i.e. between sizes 0.2 to 200.). These values are presented in Appendix II.

As has thus been observed (Section 3.2.2) an infinite number of particles may nevertheless produce a finite scattering function. In the same manner, the total scattering, or more precisely the total effective cross-section produced by particles with sizes between 0 and 0.2 can also be finite even though the total geometrical cross-section is not; this latter is in fact not finite unless $m < 3$; thus, as long as the exponent remains within the limits $3 < m < 7$, the situation which has been envisaged is actually realized.

Just as has been done for the angular values, the progressive value of the integral (3.9) must also be plotted as a function of the upper limit; figure II.14 provides such an example for various values of index and with the exponent being -3.10. The initial slope of the curves has a value $7-m$, it then attains the value $5-m$ and finally it goes to zero, thus the curves present an asymptotic plateau as long as the convergence condition holds.

Figure II.14: Progressive values of the integral providing the effective cross-section (3.9) as the upper limit grows, up to the final value $\alpha=200$. The various curves correspond to the indicated indices of refraction and to a single value of the exponent of the particle distribution.
Figure II.15: Analogous to the preceding figure, with the variable $\rho$ replacing $\alpha$. The various curves correspond to the indicated different exponents; they are normalized to their asymptotic values (100%). The scales are logarithmic.

This beginning of the plateau occurs where $Q$ becomes (on the average) independent of $\alpha$, i.e., for values of $\alpha$ which vary according to the index; these values are related since at this point the parameter $\rho = 2\alpha(n-1)$ is equal to 4.1 (cf. Section 2.2.8 and Figure II.10).

In the situation where the Van de Hulst expression (2.29) is a good approximation (which is the case with the values of index used here) a single computation suffices, with the variable $\rho$ replacing $\alpha$. The various curves of Figure II.14 correspond in fact to a unique curve, however with more or less dilated abscissas.

As a consequence of this remark, the integrals computed this time with the variable $\rho$ are represented by the curves on Figure II.15, each one corresponding to a different value for the exponent. The asymptotic values of these integrals must be computed when the upper limit increases to infinity (as long as condition 3.10 is satisfied), these values being
used to normalize the plots; however the relative curve for exponent -3 can only be placed arbitrarily since it does not have an asymptote. For all the curves the inflection appears when $\rho$ passes the value 4 (See Appendix II, for the computation of the asymptotic values).

### 3.2.4 Summary regarding the "validity domain" of the computation.

The expression "validity domain" is given an exact meaning: it is the domain in which the computation retains its significance independently of the values assigned to the upper and lower size limits. This assumes that the computed quantities would be finite if the limits were extended toward zero or towards infinity. Under these conditions, ignoring the physical limits does not invalidate the computations. If for the sake of continuity the same distribution law (power law) is assumed to govern the population whatever the size, the conditions ensuring validity refer to the exponent of the law. These conditions, as has been observed, differ depending on the quantities to be computed. They are all gathered in the accompanying table below; where the finite or infinite nature of the computed quantities is indicated, in relation to the value of the exponent, between 0 and $\alpha_m$ (influence of the lower limit) and $\alpha_M$ and $\infty$ (influence of the upper limit).

<table>
<thead>
<tr>
<th>Influence of the lower limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Scattering function $i(\theta)$, any $\theta$</td>
</tr>
<tr>
<td>Total scattering effective cross-sections</td>
</tr>
<tr>
<td>Total volume</td>
</tr>
<tr>
<td>Total area Geometric cross-sections</td>
</tr>
<tr>
<td>Total number of particles</td>
</tr>
<tr>
<td>$\Rightarrow$</td>
</tr>
<tr>
<td>Total number of particles</td>
</tr>
<tr>
<td>Total area Geometric cross-sections</td>
</tr>
<tr>
<td>Total volume</td>
</tr>
<tr>
<td>Total scattering effective cross-sections</td>
</tr>
<tr>
<td>Scattering function $i(\theta)$, with $\theta = 0$</td>
</tr>
<tr>
<td>Scattering function $i(\theta)$, with $\theta \neq 0$</td>
</tr>
</tbody>
</table>

Influence of the upper limit
It can be remarked, that where particle number, surface or volume are concerned, the conditions relating to the upper and lower limits are independent of each other; put in another way, the "validity domain" as defined above does not exist; the computation is not possible except if it can be determined that physical limits do exist. This is not the case for the magnitudes relating to scattering: the two categories of conditions may be simultaneously fulfilled in several intervals of the exponent. This is what is summarized by the following diagram, whose "validity domain" is represented by the shaded zones.

3.2.5 Extension to distribution laws other than the Junge law.

The preceding conclusions concerning the "validity domain" are valid only, at least such as they have been presented, for distributions expressed by the power law. They will be reexamined however and extended to other distributions. The previously treated case, mathematically convenient, may because of this constitute a reference case. It does not appear useful to consider cases of serrated type (rectangular, triangular, ...) for which the problem of limits and of the significance of the computation does not arise. The case of the Gaussian normal distribution however leads to such problems as will be seen further.

It is appropriate in contrast to examine the case of continuously decreasing distributions, different however from the power law. Thus plausible distributions are to be chosen, which can approximately account for the observed decrease*, within the size interval accessible to experiment. But beyond the limits of this interval, the hypothesis according

---
*This remark concerns the granulometric study, so that the choice of numerical values for the distributions, to be done later, is an anticipation on what will be presented at the beginning of Part III, to which it is referred.
to which the power law holds is abandoned, the new distributions behaving differently. The exponential function is often used to describe particle populations (natural or artificial):

$$ F_1 (\alpha) = A_1 \exp(-B_1 \alpha) , $$

(3.11)

or more frequently the gausso-logarithmic (log-normal) function:

$$ F_2 (\alpha) = A_2 \exp (-B_2 (\log \alpha / \overline{\alpha})^2) , $$

(3.12)

where $\overline{\alpha}$ is the value corresponding to the maximum of the distribution, and $B_2$ is a "geometric" standard deviation. Exponential laws (or of the exponential type, as Weibull's law) have been proposed for marine particles by Carder et al (1971). In addition J. R. Zaneveld and H. Pak (1973) use this distribution, which proves convenient for any theoretical calculations, due to convergence at the limits as will be seen below.

It is practical for comparisons with the power law $F(\alpha) = A \alpha^{-m}$, to consider the log-log graph of the preceding distributions. The first remains exponential, two point determining it completely. The second simply becomes a parabola, two points not being sufficient, so a third one is necessary, which may quite well be fixed arbitrarily as the position of the maximum $\overline{\alpha}$. It must be verified that the Gaussian Normal distribution is represented by an exponential curve beyond the maximum (but of a stronger slope than the exponential 3.11 since the argument is twice as large). On Figure II. 16, the log-log plot of the power function $\alpha^{-4}$, chosen as the basis for comparison, is represented by the straight line of slope $-4$. The other distributions have been forced to cross it at $\alpha=10$ and $\alpha=100$ and the choice has been made to center the log-normal and the normal distributions at $\alpha = 1$. 
Figure II.16: Log-log plot of various types of distributions whose values coincide at $\alpha=10$ and $\alpha=100$.

1) Power law: $F(\alpha) = 10^{n+4} \alpha^{-4}$

2) Log-normal law: $F(\alpha)=10^n \exp(B) \exp(-B(\log_{10} \alpha)^2)$ with $B=\frac{4}{3} \cdot \log_{10} 10$ (3.12b)

3) Exponential law: $F(\alpha)=10^n \exp(10, B) \exp(-E\alpha)$ with $B = \frac{4}{90} \cdot \log_{10} 10$ (3.11b)

4) Gaussian law: $F(\alpha)=10^n \exp(A)\exp(-B\alpha^2)$ with $A=\frac{4}{120} \cdot \log_{10} 10$

and $B = \frac{1}{9^2} \log_{10} A$
Figure II.18: This figure presents the progressive values of the integral
\[ \int_{F(a)}^{a} i_T(\theta, \alpha) \, d\alpha, \]
as a function of the upper limit \( \alpha \) in the case of the preceding log-normal distribution. It is at the same values of \( \theta \) as the preceding Figure II.12.
Figure II.17: Scattering functions obtained using the log-normal distribution (equation 3.12b, Fig. II.16) and the Junge distribution with -4 exponent, truncated below $\alpha=10$ and above $\alpha=100$; the scattering functions have been plotted as the decimal logarithm of the normalized scattering coefficient $\bar{\beta}(\theta)$ as a function of $\theta$. The values $\bar{\beta}_1(\theta)$ and $\bar{\beta}_2(\theta)$ related to the vertical and horizontal polarization components have been marked with points and crosses respectively.

The points marked by the arrows correspond to the computations of O. B. Brown and H. R. Gordon (1971) for the same exponents, for limits 8.6 and 86.0 (diameters 1 to 10 $\mu$m, $\lambda=488$nm) and with index 1.05-0.0li. For comparison the values have been recomputed in order to be presented in the form of the normalized coefficient $\bar{\beta}(\theta)$. The upper limit is 86 and not 200, but considering the exponent, the asymptotic form has been practically achieved (cf. Section 3.2.1) and the comparison is justified.
On the side of small sizes the exponential distribution tends towards a finite value with zero slope and the condition (3.8) of convergence always holds. The log-normal distribution tends to zero, the negative slope (between \( \alpha = 1 \) and \( \alpha = 10 \)) being necessarily weaker than that of the power law; the condition (3.8) is a fortiori fulfilled since it holds for the power law \( (7 - m > 0) \). In order for the small particles, of sufficiently large number, to play a non-negligible role it is necessary to have a power law with a negatively large exponent; in practice the distributions under consideration, attribute small numbers to them, corresponding mathematically to a semi-truncation for the purposes of scattering. The conclusion is identical as far as the Gaussian distribution is concerned.

On the side of the large sizes, the negative slopes continuously increasing and greater than that of the straight line, are found to totally satisfy the conditions of convergence, whatever the scattering angle \( \Theta \) (Section 3.2.1). With these laws, the frequency of occurrence of large particles is smaller than for the corresponding power law; in practical terms this is tantamount to truncation, as before. Figure II.17 illustrates this effect; it permits comparisons of the scattering functions obtained through the log-normal law on the one had and the power law on the other; for the former, the distribution is extended to all the particles with sizes \( 0.2 \) to \( 200 \) (for \( \alpha \)) whereas for the latter, truncation has been performed below \( \alpha_m = 10 \) and above \( \alpha_M = 100 \). By superposition it appears that these two scattering functions overlay (which is not quite the case for the polarized components \( i_1 \) and \( i_2 \)).

It is not fruitful to multiply the computations and the examples of this type, given the arbitrary character of the laws used, in particular as far as the maximum of the distribution is concerned. It suffices to state that with the exponential and more emphatically with the log-normal law the results only slightly differ from those obtained using the equivalent power law (and in the second case, as closely as needed, according to the choice of parameters). With the Gaussian law, even truncated beyond the maximum, it is by contrast never so.

Finally between the sizes (10 and 100 for the example here) where the distributions have been taken to coincide, the curves present a convexity; in consequence, these distributions favor in a certain way the intermediate sizes (20 to 50), when compared to the power law, which play a comparatively more important role in scattering. An example of this conclusion will be given further (Part III, Section 3.1.2).

END OF PART II.
PART III

PREFACE

In the computation of the scattering properties of a poly-
disperse system, there enter on the one hand the parameters tied to
the distribution law and its limits and on the other hand the relative
index of refraction which will be called "mean" index since it is
assumed to be the same for all the particles. As an example, many
numerical applications have been presented in the preceding presenta-
tion but no discussion has been given on the values attributed to the
parameters used. A certain number of theoretical cases have been
treated systematically in view of the comparison with experimental
results and a choice ought to be made which it is appropriate to ex-
amine here.

For the index of refraction of the particles, only hypotheses
can be made since no direct measurement appears to have been
performed. Starting from the mineral content of the suspended particles
(calcium carbonate, silica, aluminosilicates, various hydroxides, . . .)
which are assumed to be in crystalline form, a mean index may be
deduced. The values often proposed from such considerations are of the
order of 1.15 or 1.20* (values relative to water, i.e. absolute values of
1.53 to 1.60); computations have been made for suspended clays with
the value 1.15 (and an imaginary part of 0.001; H. Pak, R. V. Zaneveld,
G. F. Beardsley, 1971). In a way it is probably not realistic to attri-
but to the whole particle the index appropriate to the crystalline form,
even if it is essentially mineral. Various insolubles or precipitates may
be present in a highly hydrated colloidal form or the mineral portion of
a detrital particle may only be a shell; such mineral particles produce
a "mean" index which is much closer to that of water than that deduced
from the composition alone (and excluding the water). On the other
hand, the organic portion in the suspended material is always important
and the index of these substances is very close to that of water. Recently
K. L. Carder et. al. (1972) have deduced the relative index, from scatter-
ing measurements on unicellular algal cultures (Isochrisis galbana), to be
of the order of 1.026 to 1.036 for this organic material. In relation to
the total mass of the particles the organic portion** which is large for

*See for example W.V. Burt (1956), Y.E. Otchakovsky (1965), N.G. Jerlov
(1968).

** It is the particulate organic carbon which is in fact given, the weight of
the organic substances being deduced in general by multiplication with a
factor of 2. (D.C. Gordon uses the factor 1.80).
the surface waters in general, remains large even in the deeper layers; between the surface and depths down to 4000m, the relative abundance of particulate organic matter remains higher than 25% (D. C. Gordon 1970), varies from 40% to 60% (L. A. Hobson, 1967), from 40% to 100%* (P. J. Kinney et. al. 1971), from 40% to 88% (C. Copin, G. Copin, 1972), from 26% to 49% (J. E. Harris, 1972). This suggests choosing values close to 1 for the "mean" relative index of the particles. Whatever its value is, the preliminary computations of the individual scattering functions have been made for 5 values of index, covering a sufficiently large domain, from small values (1.02 and 1.05) judged to be more realistic, up to values characteristic of the mineral fraction (1.15), the intermediate values having been chosen as 1.075 and 1.10.

Regarding the distribution law, in contrast to the index, experimental results do exist. The use of an electrical particle counter (Coulter-counter) in particular has permitted acquisition of new data (L. A. Hobson, 1967; R. W. Sheldon and T. R. Parsons, 1967; K. L. Carder, 1970). The measurements of H. Bader (1970), as well as those of J. C. Brun Cottan (1971), appear to show that the Junge law (i.e. a power law of \(d^{-m}\), \(d\) being the diameter), previously proposed for atmospheric particles (C. E. Junge 1963) seems to adequately describe the marine particle distributions too. This counter permits the granulometric study of particles whose equivalent diameters vary from 1\(\mu\)m to 15 or 20\(\mu\)m**, in many cases the reduction of number of particles with increasing diameters occurs according to two successive laws, one with an exponent between -3.3 and -3.9 in the interval 1\(\mu\)m to 4 or 5 \(\mu\)m followed beyond this size by a second law with a higher exponent (in absolute value) of -4 to -5 approximately.

Numerous measurements performed in very different zones of the Atlantic and the Pacific (R. W. Sheldon et. al., 1972) show that the distributions, in a general way, are like those of equal logarithmic size intervals (for example from 1 to 2\(\mu\)m, then 2 to 4\(\mu\)m, then 4 to 8\(\mu\)m, ...).

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*From 19 to 55% for the particulate carbon - cf. see previous footnote p. 85.
** Other intervals may be studied, corresponding however to larger sizes; 1\(\mu\)m is the approximate lower limit of detection of the Coulter system. The diameters deduced from the measurements are those of equivalent spheres, i.e., presenting the same volume as the real particles, regardless of their shape.
the volume of particles belonging to these classes remaining roughly constant. It is easy to verify that this property of logarithmic equipartition of volumes is that of the distribution with exponent -4. According to these authors, it extends over a very extensive range of sizes. In near-surface waters and in productive regions, maxima (around 16-20 μm for example) are superimposed on the preceding distribution, indicating the presence of phytoplanktonic particles. For the computations, the exponents (from -3 to -5 in increments of 0.1) have been chosen in an interval largely covering the experimental values. Systematic results for 8 values of the exponent in this interval and 5 values of the index of refraction have been presented elsewhere (A. Morel, 1973); results obtained with log-normal and exponential distributions are also shown. Even if it is found that the power laws are somewhat simplistic with respect to reality, more complex distributions may be decomposed into several successive laws of this type. In any case, they are convenient for the computations and can also serve as a basis for predicting the result in the case where other distributions must be considered (cf. Section 3.2.5, Part II).

There do not appear to exist any results concerning the laws governing the distribution of particles smaller than 1 μm; for those larger than 20 μm, laws with exponents -m continue to apply (m may become slightly greater than 4, because the relative volumes of the higher order classes have a tendency to decrease, R. W. Sheldon et. al., 1972). Limits must be determined for the needs of the computations, however arbitrary they may be from a physical point of view. Their influence, as far as can be predicted, has already been studied previously. The preliminary computations on the individual scattering functions have been performed with values of α between 0.2 and 200, which were also used for computations in polydisperse systems.*

From a practical point of view, this leads to recounting the correspondence among the parameters α and ρ and the diameter d. To repeat:

* The computation has also been done between the theoretical limit $\alpha = 0$ (zero size) and $\alpha = 0.2$ (cf. Appendix II and Section 3.2.2 in Part II).
\[ \alpha = \frac{\pi d n_e}{\lambda_o} \quad \text{and} \quad \rho = 2 \alpha (n_r - 1), \]

where \(d\) is the sphere diameter, \(\lambda_o\) the wavelength in vacuum, \(n_e\) the index of refraction of water (1.33), \(n_r\) the relative index of the particle to that of water. The computations performed between \(\alpha = 0.2\) and \(\alpha = 200\) correspond to particles whose diameters go from 0.02 \(\mu\)m to 20 \(\mu\)m, when \(\lambda = 419\) nm (thus \(\alpha = 10d\)), or equally well from 0.04 \(\mu\)m to 40 \(\mu\)m when \(\lambda = 838\) nm. It also happens that \(\rho = d\) if \(n_r\) is taken as 1.05 and \(\lambda = 419\) nm; this case is often taken as an example. In order to compare the various particle distributions, they are made to coincide at \(\alpha = 10\) and \(\alpha = 100\), (Section 3.2.5 Part II), for which \(d\) is 1 and 10 \(\mu\)m, when \(\lambda = 419\) nm, i.e. diameter values which lie in the interior of the range covered by experiments. In this respect, the log-normal law used, could have been approximated by two power laws, one with exponent -3.4 up to \(\alpha = 40\) (\(d = 4\mu m\)), and another with exponent -4.5, between \(\alpha = 40\) and 200 (\(d\) of 4 to 20 \(\mu\)m), which is a plausible example (see above).

In order to guide the comparisons between experimental and theoretical scattering functions it is necessary to examine in advance how the latter behave with index and distribution law exponent (Section 1). Various comparisons are performed, and followed the observed agreements, probable values of the index and the exponent are deduced (Section 2). Various applications of the theory are examined, in particular in order to examine the spectral selectivity of scattering and the polarization. Conclusions are also obtained regarding the different roles of the various particles, and the relations between scattering and the concentration of the suspended material (Chapter 3).

1. THEORETICAL VARIATIONS OF THE SCATTERING FUNCTIONS

1.1 INFLUENCE OF THE EXPONENT OF THE DISTRIBUTION LAW ON THE SCATTERING FUNCTION

Figure II.12 presented in PART II has already demonstrated the manner in which this influence is exerted. The separation among the various plateaus which characterize the final scattering function, varies
in fact with the exponent; and the way this occurs will be determined. The beginning of the plateaus takes place at fixed values of $\alpha$ which vary for each particular angle $\theta$. As a consequence these separations are directly determined by the initial slope which is the same for all the curves (whatever the angle) and whose value is $7-m$. This permits quantification** of what is qualitatively predictable: when the large sized particles are proportionately more abundant, i.e. when $m$ decreases in absolute value, then the initial slope of the curves is larger, the plateaus are more distant from each other and the scattering function is more asymmetric. This behaviour of the scattering function is shown in Figure III.1.

Figure III.1: The effect of the distribution law exponent on the scattering function; the index of refraction is 1.05. The size parameters corresponding to the limits of the distribution are 0.2 and 200.

** Figure III.3: The effect of index of refraction on the scattering function; the exponent of the distribution law is -3.5. The size limits remain unchanged (0.2 and 200).

* These are the values of $\alpha$ for which the curves $i_T(\theta) \alpha^{-4}$ begin to decrease, and which are independent of the index except if the angle $\theta$ is small ($2^\circ$ for example). This property finds its justification in the Rayleigh-Gans theory (cf. Part II, Section 2.2.3 and 3.1.2.b).

** For example, if $\alpha_1$ and $\alpha_2$ are the values of $\alpha$ at which the first dark ring appears at angles $\Theta_1$ and $\Theta_2$ following the Rayleigh-Gans theory, then the ratio of the scattered intensities at these angles $i(\Theta_1)/i(\Theta_2)$ varies with exponent according to $(\alpha_1/\alpha_2)^{7-m}$. A table (Section 2.2.3, Part II) provides the corresponding values of $\alpha$ and $\Theta$. 
Figure III. 2: The continuous curves correspond to computed scattering functions with size limits $\alpha_m = 0.2$ and $\alpha_m = 200$, & the dashed line is for the upper limit reduced to 50. The positioning of the two groups of curves marked 1 for a relative index of 1.05 and 2 for 1.10 is arbitrary. On the contrary in each of the two cases the continuous and dashed lines have been positioned correctly in relation to each other, so that the total scattering coefficient is the same (i.e. they are correctly positioned with respect to the normalized volume scattering function). The crosses and the triangles correspond to the horizontal polarized component $i_2$, when $\alpha_m = 200$ or $\alpha_m = 50$ respectively; the dotted lines correspond to the vertical component $i_1$; without any difference in the two cases.
Figure III.1 shows the values of the integral

\[ \int_{\alpha_m}^{\alpha_M} i^\theta (\theta, \alpha) \, \alpha^{-n} \, d\alpha, \]

as a function of \( m \) (varying between 3 and 5) for various angles \( \theta \); the upper and lower limits remaining constant at 200 and 0.2 respectively. The intensity at 0\(^{\circ}\) serves as a normalization value. From this figure it is determined that the important variation is that of the ratio \( i^\theta (2\, ^{\circ})/i^\theta (0\, ^{\circ}) \), which however has no meaning, since as has been seen, the integral at 0\(^{\circ}\) does not converge. The significant fact is the respective location of the curves for angles other than 0\(^{\circ}\). The scattering function (from 2\(^{\circ}\) to 180\(^{\circ}\)) covers 5 orders of magnitude for an exponent of -3, and only 3 orders of magnitude for an exponent of -5. Another significant statement can be made which will be of importance: between the exponent values of -3 to -4 the curves remain approximately parallel. This means that in this domain, the scattering function is least sensitive to variations in the distribution law, at least for the "medium" angles (since at 180\(^{\circ}\) there is an exception and for 0\(^{\circ}\) nothing can be said).

It is also appropriate to examine the case in which the distribution follows a law whose exponent attains two successive values \( m_1 \) and \( m_2 \), instead of a single value. According to what has been observed, the second value is larger than the first (in absolute value); and corresponds to the law applying to particles of larger than 5\( \mu \)m diameter (\( \alpha > 50 \)). The integrals for the various angles (except 0\(^{\circ}\)) have in fact attained their asymptotic values for \( \alpha = 50 \) (See Figure III.2). Consequently, whether the particles are distributed according to an-\( m_1 \) or-\( m_2 \) law beyond \( \alpha = 50 \), has no significance on the final form of the scattering function, except at 0\(^{\circ}\). Figure III.2 shows this even more strongly: scattering functions corresponding to populations extending from \( \alpha_m = 0.2 \) to \( \alpha_M = 200 \) and from \( \alpha_m = 0.2 \) to \( \alpha_M = 50 \) have been plotted for two values of the index of refraction; in the latter case it may just as well be assumed that the value of the second exponent \( m_2 \) is infinite. Thus the scattering functions practically overlap except at 0\(^{\circ}\). In conclusion, by using the above distribution laws, the large particles play a very minor role and their greater or lesser numerical densities (\( m_2 \) larger or smaller) barely affect the shape of the scattering function.
1.2 INFLUENCE OF THE INDEX OF REFRACTION ON THE SCATTERING FUNCTION.

The value of the exponent affects the shape of the resulting scattering function, but the shape of the individual scattering functions appearing in the integrations is determined by the value of the index of refraction. Figure III.3 supplies an example of this influence; with the exponent remaining fixed \( m=3.5 \) the ratios \( i_T(\theta)/i_T(0^\circ) \) have been plotted this time as a function of index, with the upper and lower limits of the integrals remaining the same as before \( (200 \text{ and } 0.2) \). It had been pointed out in PART II (Section 2.2.5 and 2.2.6) that the influence of the index on the individual scattering functions is practically limited to the region of small \( (0^\circ-10^\circ) \) or large (around \( 180^\circ \)) angles. The summations which were performed do not modify this property which permits the following remarks:

---

the global asymmetry of the scattering function decreases as the index increases. Thus the ratio of the scattered intensities at \( 2^\circ \) and \( 140^\circ \) corresponds to 6 orders of magnitude for \( n=1.02 \) but only 4 when \( n=1.15 \).

---

But it can be stated that this variation of the asymmetry can be uniquely attributed to the variation of the ratio \( i_T(10^\circ)/i_T(2^\circ) \). In contrast the relative curves at angles between \( 10^\circ \) and \( 140^\circ \) (and even at \( 160^\circ \) which is not shown) remain essentially parallel and thus the middle portion \( (10^\circ - 160^\circ) \) of the scattering function hardly depends on the index of refraction.**

---

finally, the curve at \( 180^\circ \) shows that in relative terms, backscattering is enhanced as the index increases (following the enhancement of the reflection factor).

It may be noted by now, that based on the preceding statements, criteria exist permitting, at least in theory, extraction of a plausible index of refraction from measurements. The ratio of scattered intensities at \( 2^\circ \) and \( 10^\circ \) is a very sensitive criterion, especially if the index varies between \( 1.02 \) and \( 1.075 \); for the higher values \( 1.075 \) and \( 1.15 \), backscattering provides another one (unfortunately less exploitable experimentally). A detail, which may be seen on the continuous trace of

*Translator's note: By individual scattering function is meant the function \( i_T(\theta, \alpha) \) which holds for a single size \( \alpha \). The term scattering function is therefore used to imply that for the polydisperse ensemble, i.e., \( i_T(\theta, \alpha) \cdot \alpha \).

** This cannot be generalized; this property is tied to the values of index considered here, which are sufficiently close to 1 to render the Rayleigh Gans approximation valid and applicable (cf. Part II, Section 2.2.3).
the scattering function, may also serve as a criterion. The existence of a very weak relative maximum is determined, whose angular location is directly tied to the value of the index of refraction (at around 60° for n=1.05 and around 85° for n=1.10, Figure III. 2)*. The maximum is all the more pronounced as the computation takes into account the larger particles, which are the only ones responsible for this effect.

From a practical point of view, the measurements of scattering functions are rarely performed at a sufficiently high angular resolution to render this phenomenon unambiguously evident. It can be noted however that the mean scattering function obtained by Jerlov (1961) and by Otchakovski (1965) clearly demonstrate a convexity around 60°-70° (which would correspond to an index of around 1.05), (Part I, Fig I.1). An apparatus which would produce a continuous curve of the scattering coefficient vs angle, would make it possible to determine whether this effect appears more or less permanently, and thus whether it is possible to deduce a mean index of refraction, based on this observation. In this respect, the scattering function determinations by J. W. Reese and S. P. Tucker (1970) demonstrate that very frequently a weak maximum appears between 55° and 75°.

Is it possible to find criteria with respect to the exponent, such as those indicated for the index? On re-examination of Figure III.1, it is established that the total amplitude of the scattering function is a priori the quantity truly varying with the exponent. But for the reasons of no convergence already given, the value at 0° cannot be taken into consideration. In addition the angular region 2°-10° is particularly sensitive to the value of index. Using the ratio of intensities at 2° and 140° to define the amplitude, leads to a quantity for which the effects of exponent and index cannot be separated. On the contrary, as is also shown in Fig. III. 3, the ratio i(10°)/i(140°) is practically independent of the index. The amplitude of the scattering function, no longer the total one, but that restricted to the domain 10°-140° can well constitute the sought-after criterion.

*See also the scattering functions presented in Appendix II. The relation between the angle at which this concentration of energy appears and the index of refraction has been presented graphically (Part II, Figure II. 8).
1.3 CONCLUSIONS REGARDING THE THEORETICAL BEHAVIOR OF THE SCATTERING FUNCTION

A first conclusion may be drawn from the above. When the index of refraction varies within fairly broad limits, the scattering function is only very slightly affected, at least as far as the "medium" angles are concerned (10° to 140°, just to set some values); the same is true when the exponent of the distribution varies, within slightly more restricted limits (approximately -3 to -4). This may constitute a first explanation to what in fact is known experimentally, namely the small variability of the scattering function of marine particles --- it must be noted that this point has been established primarily for the "medium" angles ---. This explanation will be developed and slightly modified in the following section. The small angle case, where the variability is greater, will also be examined.

2. INTERPRETATION OF THE OBSERVATIONS AND APPLICATIONS

It is initially appropriate to demonstrate that the theoretical scattering functions which are not too variable can be said to coincide with the experimental scattering functions, and to examine whether within the whole range of possible variations some may more precisely account for the experimental scattering functions. Through this procedure, probable values of the index of refraction and of the exponent of the distribution will be extracted.

In order to carry out the comparisons, certain properties will be used (Section 2.1) --- such as the ratios \( \beta(10°) / \beta(20°) \) and \( \beta(140°) / \beta(10°) \) --- which have been found to be sufficiently variable, whether with index or exponent, to serve as criteria. In addition (Sec. 2.2) certain ratios of volume scattering function to total scattering coefficients* will be used --- i.e. normalized coefficients \( \tilde{\beta}(\theta) \) --- whose values depend on both index and exponent. If the experimental variability of these ratios appears weaker than the theoretical variability, it will signify that the real indices and exponents vary within a more restricted range than that envisioned in the computations. Each property leads to delimiting such a region; if all these regions are compatible or if they at least have a common portion, one may hope to deduce from experiments plausible values of index and ex-

*Translator's Note: The volume scattering function \( \beta(\theta) \) is called "coefficient angulaire de diffusion" in French, literally "angular scattering coefficient". I may have inadvertently used the literal rather than the formal translation of the term somewhere.
ponent. Scattering functions computed with these values are compared to experimental scattering functions. (Section 2.4)

Methods for evaluating the total scattering coefficient from angular measurements have been proposed by various authors. Examination of the theoretical variability of certain normalized volume scattering functions (at 90°, 45°, and 4° - Section 2.2 and 2.3) in order to determine the index and exponent is at the same time a way of evaluating whether such methods are well founded.

2.1 APPLICATION OF CRITERIA REGARDING THE INDEX AND THE EXponent OF THE DISTRIBUTION. RATIO OF VOLUME SCATTERING FUNCTIONS.

The ratio of the volume scattering function at 10° and 2° is, as has been seen (Sec. 1.2), much more sensitive to variations in index than exponent; conversely the ratio of the function at 140° and 10° is almost exclusively dependent on the exponent; the variables index and exponent are thus in some sense separable. The theoretical behaviour of these ratios with the two variables are presented in Fig. III.4. Experimental values of the same ratios are grouped in Table I. Measurements performed simultaneously at 2° and 10° or at 10° and 140° are very few. In addition, in order to be used here, the measurements at 140° must be expressed in absolute value in order to enable subtraction of the molecular scattering component which is often not negligible; it is only under this condition that the ratio $\beta(140°)/\beta(10°)$ is truly meaningful. Adopting the values $2 \times 10^{-2}$ and $5.5 \times 10^{-2}$ in order to fix the experimental limits of the ratios $\beta(10°)/\beta(2°)$ and plotting them on the graph of the theoretical values (Figure III.4) one finds a domain delimited by the intersections. This corresponds to a simultaneous combination of values of index and exponent for which, theory can account for the experiment. By reason of the nature of the criterion itself, the domain leaves the values of the exponent undetermined, but restricts the possible values of index; the values outside the interval 1.02-1.06 appear to have to be excluded. In the same way, choosing as limits the experimental values $0.6 \times 10^{-3}$ & $2.3 \times 10^{-3}$ for the second ratio another domain can be determined, which does not contain information about the index (which is however, smaller than 1.10) but defines the values of the exponent; for example, if the index is assumed equal to 1.05, the values of the exponent, compatible with the experimental results are approximately from -3.8 to -4.3.

* Translator's Note: There appears to be a lack of an appropriate term in the English literature to designate the value of the volume scattering function at a given angle, e.g. $\beta(10°)$. In French the term "coefficient angulaire de diffusion" is used, literally "angular scattering coefficient".
Table I: Experimental values of the ratios $\beta(10^\circ)/\beta(2^\circ)$ and $\beta(140^\circ)/\beta(10^\circ)$

<table>
<thead>
<tr>
<th>English Channel</th>
<th>méditerranéen</th>
<th>Atlant.</th>
<th>Pacif.</th>
</tr>
</thead>
<tbody>
<tr>
<td>(2)</td>
<td>(3)</td>
<td>(4a)</td>
<td>(4b)</td>
</tr>
<tr>
<td>2.6 (2.2 - 3.0)</td>
<td>3.0</td>
<td>3.2</td>
<td>4.7</td>
</tr>
<tr>
<td>4.8 (4.5 - 5.4)</td>
<td>3.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.35 (1.00 - 1.70)</td>
<td>0.60 - 1.90</td>
<td>1.66 - 2.28</td>
<td>(5)</td>
</tr>
</tbody>
</table>

1) Computed ratio of values borrowed from curves by Y. E. Otchakovsky (1965).

2) Mean values of ratios corresponding to mean particle scattering function. This has been obtained by relating values at angles between $30^\circ$ and $150^\circ$ (A. Morel, 1965) to values at small angles of $1.5^\circ$ and $14^\circ$ (D. Bauer, A. Morel, 1967).

3) R. E. Morrison (1970), ratios measured from curves traced according to what the author calls the "Duntley" extrapolation, (Fig. 4 in ref.)

4) Ratio corresponding to the mean of in situ measurements, at three wavelengths; (4b) to the mean of in vitro measurements at two laser wavelengths (F. Nyffeler, 1970). For the in situ measurements it is necessary to extrapolate from $3^\circ$ to $2^\circ$ in order to form the ratio.

5) Three measurements in the Atlantic (Bahamas) and two in the Pacific (outside San Diego) (T. J. Petzold, 1972). For $140^\circ$, the molecular scattering has been removed from the absolute values given by the author, before computing the ratio.

Figure III. 4: Theoretical variation of the ratios $\beta(10^\circ)/\beta(2^\circ)$ and $\beta(140^\circ)/\beta(10^\circ)$ as functions of the index of refraction, for the indicated values of the exponent. The experimental values are located in the bands indicated by the curved brackets.
For clarity, the two domains thus defined are shown on a $m-n$ plot (i.e., whose coordinates are exponent and index, see Fig. III.6). The common portion thus corresponds to plausible values of index and exponent taking into account the available experimental results.

Spilhaus (1968) has considered using the ratio $\beta(30^\circ)/\beta(45^\circ)$ to characterize the shape of the scattering function. Figures III.1 and III.3 show that the curves drawn for $20^\circ$ and $40^\circ$ remain essentially parallel as the index and the exponent vary. It therefore appears that this ratio (only slightly variable in theory) is, all things considered, a bad enough choice to be dismissed. Finally, the theoretical values perfectly corroborate the experimental values (under the condition that here too the molecular scattering is subtracted; this is another difficulty in the interpretation of the ratio which Spilhaus does not appear to have considered). Table II demonstrates concurrently the theoretical values and a certain number of experimental values of this ratio. Even if the values are significantly distinct statistically (A. F. Spilhaus, 1968) it is seen that it is difficult to draw conclusions from the small observed variations, except possibly where the exponent is concerned: these experimental values would show that it could essentially vary between -3.5 and -4.2, the most frequent value being -3.9, which is not in contradiction with the conclusions drawn by application of the previous criteria.

2.2 RELATIONS BETWEEN $\beta(45^\circ)$ or $\beta(90^\circ)$ and $b$

Jerlov had since 1953 (N.G. Jerlov, 1953) considered deducing the total scattering coefficient $b$ from measurements at $45^\circ$. Numerous authors have elsewhere submitted that the theoretical proof of the constance of the ratio of $\beta(45^\circ)$ to $b$ had been made by D. Deirmendjian (1963). In actual fact this theoretical work treated the case of scattering in fog and clouds, the computation being made with the index of refraction of water and special distribution laws. Deirmendjian's conclusion, according to which $\bar{\beta}(40^\circ)$ is practically constant (and in addition equal to $10^{-1}$) had not been extended to all cases, since it concerns not a proof, strictly speaking, but a statement of a numerical property belonging to the adopted distributions. Figure III.5 shows how $\bar{\beta}(44^\circ)$ varies with index for four values of the exponent of the distribution law*. In contrast to the distributions considered by Deirmendjian, the power laws carry the possibility of an important variation of the normalized coefficient $\bar{\beta}(44^\circ)$, for a fixed value of index.
TABLE II: Theoretical and experimental values of the ratio $\beta(30^\circ)/\beta(45^\circ)$

<table>
<thead>
<tr>
<th>Theoretical values</th>
<th>Experimental values (and standard deviations)</th>
</tr>
</thead>
<tbody>
<tr>
<td>exponent</td>
<td>Index of refraction</td>
</tr>
<tr>
<td></td>
<td>Ex. Ch. - Med.</td>
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<tr>
<td></td>
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<td></td>
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</tr>
<tr>
<td></td>
<td>Ocean Indien</td>
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<td></td>
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</tr>
</tbody>
</table>

(1a) Mean value for 27 measurements in the English Channel and 40 in the Mediterranean (A. Morel, 1965).

(1b) Mean values at wavelengths 546, 436, 366 nm, for 17 samples from the Indian Ocean (Madagascar, unpublished, A. Morel, 1967).

(2) Mean values for 5 water types between Woods Hole & Sargasso Sea (A. F. Spilhaus, 1968).

(3) R. E. Morrison, 1970.


(5a) Ratios obtained from observations by T. J. Petzold (1972).

(5b) Ratios obtained from the preceding values, reduced by the portion due to molecular scattering (1.67 x 10^-4 m^-1 at 90° for $\lambda$=510nm, A. Morel 1968).
Figure III. 5: Theoretical variation of the normalized volume scattering function for 4°, 6°, 44° and 90°, with index and exponent. The curved brackets correspond, as in Fig. III. 4, to the observed variation of the same coefficients. These theoretical values have been computed for relative sizes α extending from 0.2 to 200 as in Fig. III. 4.

Table III presents a certain number of values obtained for the ratio \( \beta(45^\circ)/b \) - i.e. \( \bar{\beta}(45^\circ) \) - which are quite dependable in the sense that the computation of \( b \) through integration is based on experimentally determined values at the small angles rather than extrapolated ones, and also in the sense that, wherever it exists, the part which arises from molecular scattering is subtracted from \( \beta(45^\circ) \).

* The relative curve for the exponent -3.10 is not represented. In fact for this value of the exponent, the asymptotic value for the total scattering coefficient is far from being reached with the upper limit \( \alpha_M \) used (being \( \alpha = 200 \), corresponding to \( \rho \) between 8 and 60 as the index varies between 1.02 and 1.15); therefore \( \bar{\beta}(44^\circ) \) is overestimated in this case. This effect, of several percent, does not exist for the other exponent values (cf. Part II, Section 3.2.3 and Fig. II.15) The problem does not appear for Fig. III. 4 since the volume scattering functions, which form the ratio, have attained their asymptotic value even for the exponent -3.10.
Table III: Experimental values of $\beta(45^\circ)$ and $\beta(90^\circ)$ and standard deviations.

\[
\begin{array}{|c|c|c|c|}
\hline
& \text{Eng-Channel} & \text{Baltic} & \text{Medit.} & \text{Atlant.} \\
\text{Mean & Med.} & \text{(1a)} & \text{(2a)} & \text{(2b)} & \text{(3)} \\
\hline
2.55 \pm 0.5 & 2.4 \pm 0.9 & 2.5 \pm 0.7 & 1.6 \pm 0.4 \\
\hline
2.3 \pm 0.7 & 1.9 \pm 0.3 & 3.4 \pm 0.2 \text{ Black Sea} & 2.58 & 1.82 \\
\hline
\text{Medit.} & \text{Atlant.} & \text{(4)} & 3.05 & 1.78 \\
\text{Atlant. Pacif.} & \text{(5)} & 2.80 & & \\
\hline
\end{array}
\]

\[
\begin{array}{|c|c|c|}
\hline
\text{Eng-Channel} & \text{Baltic} & \text{Medit.} \\
\text{Mean & Med.} & \text{(1a)} & \text{(2b)} & \text{Atlant.} \text{ (3)} \\
\hline
2.25 \pm 0.4 & 3.0 \pm 1.3 & 2.9 \pm 0.6 \text{ Black Sea} & 3.16 & 2.00 \\
\hline
\text{Medit.} & \text{Atlant.} & \text{(4)} & 2.13 & 1.63 \\
\text{Atlant. Pacif.} & \text{(5)} & 1.85 & & \\
\hline
\end{array}
\]

(1) Mean particle scattering function (A. Morel, 1965) divided by the value 444 of the integral giving $b$, using $\beta(90^\circ)=1$ in the computation (A. Morel, 1968, and corrected computation 1970); when divided by 337 given by F. Nyffeler (1969) for the same integral, the values $2.0 \times 10^{-2}$ and $2.25 \times 10^{-3}$ become $2.6 \times 10^{-2}$ and $3.0 \times 10^{-3}$ respectively. There are 27 measurements form the English Channel and 40 from the Mediterranean.

(2) 16 measurements from the Baltic Sea (G. Kullenberg, 1969).

(2b) 14 measurements from the Mediterranean (G. Kullenberg, N. Olsen, 1972); the value at 150m, station A2, is separate from the mean.

(3) Inverse of the ratios $b/\beta(0)$ given by R. E. Morrison (1970), where $b$ is obtained by including the measurements at small angles (named "S" by the author).

(4) 23 measurements from the Mediterranean, 90 from the Atlantic, 104 from the Black Sea; measurements of V. I. Mankovsky (1971).

(5) Measurements by T. J. Petzold (1972); the molecular scattering has been removed from the values presented by the author, before taking the ratios.
These remarks are also valid for the experimental values of $\beta(90^\circ)$ appearing in the same table. Examination of this table, shows that the experimental values of $\beta(45^\circ)$ occur within a narrower interval than the theoretical values, implying that the actual variations of the index and the exponent are weaker than those considered in the computation. As previously, the corresponding domains are obtained by comparing the computed with the experimental values choosing as limits $1.5 \times 10^{-2}$ to $3.6 \times 10^{-2}$ for $\overline{\beta}(45^\circ)$ and $1.5 \times 10^{-3}$ to $4.0 \times 10^{-3}$ for $\overline{\beta}(90^\circ)$. The domains which are practically overlapping are given in the $m-n$ diagram (Fig. III.6).

Figure III.6: Plots on the $m-n$ plane ($n$ is the index of refraction on the abscissa, $m$ is the exponent of the distribution on the ordinate) demonstrating the domains where theoretical and experimental values are compatible. On the left: the band between the continuous lines is obtained by applying as a criterion the ratio $\beta(10^\circ)/\beta(20^\circ)$, and that between the dashed lines by using $\beta(140^\circ)/\beta(10^\circ)$. The common region of the two bands is transferred to the figure on the right. On this one are also shown the domain obtained by comparing the theoretical and experimental values of the normalized values $\overline{\beta}(44^\circ)$ (continuous lines) and $\overline{\beta}(90^\circ)$ (dashed lines).

In any case, if the relation between $b$ and $\beta(45^\circ)$ effectively demonstrates a certain invariance, this is not, as we would like to find, theoretically justifiable. On the contrary, it would rather prove a relative stability of the properties of marine particles, both in their distribution and in their mean composition, on which the index depends.
2.3 SPECIAL PROPERTIES OF THE COEFFICIENTS $\beta(4^0)$ and $\beta(6^0)$

The numerical property pointed out by Deirmendjian for a scattering angle of $40^0$ has an equivalent in the case of the indices and the Junge distribution laws considered here. If the scattering functions obtained for a variety of indices and exponents are examined, (A. Morel, 1973) where they are drawn as normalized values $\beta(\theta)$, it is determined that they have approximately the same values around $4^0$ and $6^0$ (of the order of 12 and 6 respectively). The values of $\beta(4^0)$ and $\beta(6^0)$ have been plotted as functions of the index for various values of the exponent on Figure III.5. It can be seen that for a given value of the index, the variations of the above terms with exponent are weak compared to those of $\beta(44^0)$. Even if the variations in index are also considered, between 1.02 and 1.10, the conclusion more of less still holds; thus for example $\beta(4^0)$ varies from 9.6 to about 14. This supports a recent observation by V. I. Mankovsky (1971) on the scattering function plots of normalized coefficients $\beta(\theta)$ (Fig. III 6B). All these scattering functions cross on the average around $\theta=4.5^0$ and the statistical study demonstrates that the correlation coefficient has its highest value when in the regression between b and $\beta(\theta)$, $\theta$ has the value $4.5^0$. This regression produces $\beta(4.5^0)=9.0\times b$. Assuming (from the slope of the scattering function) that $\beta(\theta)$ varies as $\theta^{-1.5}$, in this angular domain, the preceding value is changed for $4^0$ to $\beta(4^0)=10.7$. This value is identical to what is presented here: $4750/444=10.7(\pm 1.7\%)$, (D. Bauer, A. Morel, 1967 and Table II, Part II).

![Graph](image)

Figure III.6B: Reproduced from V.I. Mankovsky (1971). Normalized scattering functions $\beta(\theta)$ from the North Atlantic.
The fact that the normalized coefficient $\bar{\beta}(4^0)$ is only slightly dependent on the index and the exponent suggests that the comparison of experimental and theoretical values has little to suggest on the plausible values of index and exponent. However if for example the value $10.5 \pm 2$ is used, it excludes the possibility of indices greater than 1.10; on the other hand if it is assumed that the index has the value 1.05, all the exponents (between -3.2 and -5) are possible. It can be added that $\bar{\beta}(10^0)$, already more variable theoretically than $\bar{\beta}(4^0)$ or $\bar{\beta}(6^0)$, remains nevertheless close to 1; it varies from 0.8 to 1.8 when the index is between 1.02 and 1.05 and the exponent between -3.5 and -4.2, which the experiments confirm, without going into detail* (it becomes on the contrary greater than 2, whatever the exponent, if the index is equal to or greater than 1.075). For this $10^0$ angle, the volume scattering function and the total scattering coefficient present numerically similar values.

2.4 PLAUSIBLE VALUES OF THE INDEX OF REFRACTION AND OF THE EXPONENT OF THE DISTRIBUTION LAW

Each of the various comparisons that can be made, leads to delimiting a domain of the index and the exponent which renders the experimental and theoretical values compatible. These domains, as has been shown in Figure III.6, present a common region in which all the criteria used are simultaneously fulfilled. Thus the combination of values of index and exponent is found which may be considered plausible. It can be remarked that the values of the exponent (from -3.8 to -4.2) are in agreement with values supplied from direct particle counts (cf. Preface to Part II) and also that the determined values of index, from **1.02 to 1.05, appear realistic. On this last point,

* See page 18 $\bar{\beta}(10^0)$ = 1.126 and also Mankovski's values: 0.7 and 1.3 approximately, and Petzold's: 0.85, 1.13, 1.04 for three Atlantic measurements; 0.88, 0.99 for two measurements off San Diego.

** The central value of 1.04 is, to be noted, that which permits the greatest variations of exponent in the interior of the compatibility domain. The examples presented below, correspond however only to cases where the index is set equal to 1.02 or 1.05, this taking into account the preliminary computations of the individual scattering functions (Section 2.2, Part II).
H. R. Gordon and O. B. Brown (1972) have recently concluded that the scattering functions determined by G. Kullenberg (1968) in the Sargasso Sea, may be correctly interpreted if the value of 1.05-0.01 is given to the index. Using a very different method based on the selectivity of scattering, J. R. Zaneveld and H. Pak (1973) computed approximate values of the relative index between 1.02 and 1.04 for the particles of both surface as well as deep layers (3000m). It is appropriate to add here that there does not exist a unique solution; different combinations of index and exponent lead to comparable and acceptable scattering functions, taking into account the variability of the experimental scattering function. Several examples are demonstrated in what follows. The lack of knowledge of the physical limits of the population introduced in the computation, necessitates further examination of their influence on the preceding conclusions.

2.4.1 COMPARISON OF THE SCATTERING FUNCTIONS

The comparison can be performed for all angles, by plotting the scattering functions computed under the previous conditions. Several examples have been provided below (Fig. III.7); the mean experimental scattering function is also presented on the same scales with the tabulated normalized values of the volume scattering function (cf. Part I). Other examples of theoretical curves, computed for cases beyond the domain of realistic values, have been presented in a more complete form elsewhere (A. Morel, 1973).

The best agreement is obtained for the exponent taken equal to -4, the index being 1.05; in which case the experimental and theoretical curves are practically indistinguishable from each other. This agreement, which also holds well for neighboring values of index and exponent, concerns the scattering function with natural light. Two remarks may be made on this subject:

a) the agreement between the experimental and theoretical values for the total intensity \( \beta(\theta) \), does not imply in itself the coincidence of the values of the two polarized components; for example, if on the one hand \( n=1.05 \) and \( m=-3.9 \) and on the other hand \( n=1.02 \) and \( m=-4.20 \), the computed values for \( \bar{\beta}(\theta) \) are practically identical but the polarization is much stronger in the second case (it is total at 90° at almost 10^-3). This question will be examined in detail elsewhere (Section 4).
Figure III.7: The theoretical scattering functions are shown by the continuous lines as the logarithm (base 10) of the normalized volume scattering functions $\bar{\beta}(\theta)$ vs $\theta$, computed for the indicated values of index, exponent and size limits. The dots and the crosses correspond to the vertical $\beta_1(\theta)$ and horizontal $\beta_2(\theta)$ polarized components respectively. The experimental scattering function is presented in the same way (on the sixth graph). On the others it is shown by the dashed lines. "Efficiency" is a short form denoting the mean scattering efficiency factor $Q$ whose value has been computed (Part II, equation 3.2) and is indicated in each case.
(Figure III.7 continued)

\[ \tilde{\rho}(\theta) \]

<table>
<thead>
<tr>
<th>Index</th>
<th>Exponent</th>
<th>Limits</th>
<th>Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.050</td>
<td>-3.90</td>
<td>0.2 200</td>
<td>0.041248</td>
</tr>
</tbody>
</table>

\[ \tilde{\rho}(\theta) \]

<table>
<thead>
<tr>
<th>Index</th>
<th>Exponent</th>
<th>Limits</th>
<th>Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.050</td>
<td>-4.20</td>
<td>0.2 200</td>
<td>0.012798</td>
</tr>
</tbody>
</table>

\[ \tilde{\rho}(\theta) \]

<table>
<thead>
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<th>Index</th>
<th>Exponent</th>
<th>Limits</th>
<th>Efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.050</td>
<td>-4.00</td>
<td>0.2 200</td>
<td>0.027553</td>
</tr>
</tbody>
</table>

\[ \tilde{\rho}(\theta) \]

<p>| |</p>
<table>
<thead>
<tr>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>EXPERIMENTAL</td>
</tr>
</tbody>
</table>

\[ \theta^\circ \]

<table>
<thead>
<tr>
<th>0 30 60 90 120 150 180</th>
</tr>
</thead>
</table>

\[ \theta^\circ \]

<table>
<thead>
<tr>
<th>0 30 60 90 120 150 180</th>
</tr>
</thead>
</table>
b) as indicated in Fig. III.6, three out of four criteria may be satisfied if, as the index increases, the exponent decreases (in absolute value). Taking for example the index 1.10 and the exponent -3.3, a satisfactory curve is obtained* as far as the experimental scattering function is concerned, except obviously for the small angle domain. The theoretical ratio $\beta(10^\circ)/\beta(2^\circ)$ (0.101) is 2 to 5 times higher than the experimental ratio, and the slope in this angular region is very small.

This second remark generates additional emphasis on the usefulness of the criterion of the ratio $\beta(10^\circ)/\beta(2^\circ)$ to indirectly determine the mean index of refraction of the particles.

2.4.2 First remark on the plausible limits of the distribution

The theory demonstrates that there exists coherence among the experimental results relative to the particle distribution on the one hand and to scattering on the other, under the condition that the index of refraction is given certain values; this in any case appears reasonable. However, to reach this coherence, another category of hypotheses have had to be made; namely those which refer to the values of the two size limits, regarding which it may be asked whether they raise uncertainties about the conclusions. The possible effects are successively examined:

Upper limit: The value given to it very slightly affects the shape of the final scattering function except at $0^\circ$ angle, and the results concerning the validity domain, which were obtained without including $0^\circ$, still hold. The particular case of $0^\circ$ angle will be examined later. (Section 3.1.3), as well as the polarization problem (Section 3.3) for which as will be seen, the value given to this limit is determinate.

* Abstractly setting the maximum around $\theta = 85^\circ$ whose position is related to the value of the index of refraction, (Part II, Section 2.2.5) and whose amplitude increases as the exponent decreases in absolute value.
Lower limit: as the truncation* progresses towards increasing sizes, the asymmetry of the scattering function is accentuated (by reducing the scattering initially at large angles and then at smaller and smaller angles - cf. Part II, Section 3.2.2). Among the ratios which may serve as criteria, the first to be employed is $\beta(140^\circ)/\beta(10^\circ)$, and then $\beta(90^\circ)$; the ratio $\beta(44^\circ)$ is used except if the lower limit $\alpha_m$ goes beyond 5; finally $\beta(10^\circ)/\beta(2^\circ)$ or $\beta(4^\circ)$ is not used as long as $\alpha_m$ remains less than 10 (cf. Figure II.12). Under these conditions where the ratio $\beta(140^\circ)/\beta(10^\circ)$ is small, whatever the values of index or exponent, the curves describing its behaviour (Fig. III.4) are approximately subjected to a translation of at most one order of magnitude, if $\alpha_m$=10; similarly the curves for $\beta(90^\circ)$ are also displaced by almost an order of magnitude. Compared to the theoretical values diminished in this way, the experimental values cannot be explained unless both the index becomes large, greater than 1.15, and the exponent becomes large, above 5 or 6 (in absolute value). As a result the different domains shown in Fig. III.6 will become disjointed with the new hypotheses, and the common portion will no longer exist. It follows then, that no scattering function can be computed which can account for the experimental values, at all angles.

In conclusion, the coherence previously evoked also disappears. In order to conserve it therefore, the lower limit $\alpha_m$ must be fixed at a value no greater than 1. Without it constituting an irrefutable proof, there is still room for thought on whether this also occurs in reality; it suffices to assume that the distribution law which was experimentally established down to $1\mu m$ particles, be in fact extended down to particles of $0.1\mu m$ in size.

*Or if the distributions are not extended to small sizes through the Junge law but instead through the log-normal or exponential distribution, which is equivalent to truncation. (cf. Part II, Section 3.2.5).
3. OTHER CONCLUSIONS AND APPLICATIONS OF THE THEORETICAL COMPUTATIONS

The first contribution of the theory was to demonstrate that starting from the granulometric data and using certain hypotheses whose validity has been demonstrated, the general shape of the scattering function is given a satisfactory explanation. The theoretical analysis has also illuminated other conclusions concerning the different roles that the various classes of particles play in scattering, some having a predominant importance (Section 3.1). This idea of "effective" classes is essential in attacking the spectral selectivity problem and in facilitating the interpretation (Section 3.2). Questions touching on polarization, which appear in the comparison of theoretical and experimental scattering functions are to be specially examined (Section 3.3). Finally the relations between particle number density and scattering which can be deduced from the theory, are examined from a practical point of view (Section 3.4).

3.1 THE DIFFERENT ROLES OF THE VARIOUS PARTICLE CLASSES

3.1.1 Case of total scattering

It is conceivable that in the case of a particle distribution obeying a continuously decreasing law with size, i.e. in the case where it is impossible to determine a mean size, there exists nevertheless a class of particles having a dominant role in the scattering phenomenon. This originates from what has been examined in Part II which has led to the determination of the validity domain of the computation. It has been seen that the "small" particles, even though more numerous (and even of infinite number) can produce a weak and finite total scattering, since their efficiency factor Q is small; in contrast, the "large" particles, for which Q is constant and equal to 2, are too few according to the distribution law to notably contribute to the total scattering. Between these two extremes there exists a favorable case that is to say a category of particles, essentially due to which scattering occurs, since both their numbers and their efficiency are sufficiently large. In order to determine the sizes of these particles, it suffices to refer to Figure II.15 where, for various values of the exponent, one may find the value of the parameter \( \rho \) corresponding to a given fraction of the total scattering (The plots have been computed for particles whose sizes extend from zero to infinity). These values may be pre-
sent in a different manner which makes more apparent the dominant role of certain classes of particles. Thus on Fig. III. 8 the values of \( \rho \) corresponding to given fractions of the total scattering (1%, 5%, 10%, 50%, 90%, 95%, 99%) have been plotted as functions of the exponent.

Fig. III. 8: Values of the parameter \( \rho \) corresponding to a given percentage of the total scattering, as a function of the exponent. The total scattering has been computed for a unlimited population and thus has its maximum possible value. The scale on the right for the parameter \( \alpha \) corresponds to an index of refraction of 1.05.

All the curves tend towards infinity for the exponent value of -3, since with this value and the hypothesis of an unlimited distribution, the scattering is itself infinite. For the other values in contrast, it is observed that the greatest portion of the scattering (for example 90%, delimited by the curves 5% and 95%) is produced by intermediate-size particles; to provide an example, when the exponent of the distribution is -4, those particles with diameters between 0.4 and 20\( \mu \)m are responsible for 90% of the scattering, those between 0.6 and 10\( \mu \)m for 80% (assuming an index of refraction of 1.05 and a wavelength of 419\( \text{nm} \)). Table IV gives these values in a more complete form; it also contains the values of \( \rho \) corresponding to 50%. These latter values define in some way a median class with respect to scattering: the class of particles with larger dimensions than this class, and the class with smaller dimensions contribute equally to scattering. As already observed (Part II, Section 3.2.5) the log-normal and exponential distribution laws lead to a more restricted size interval for the same fraction (80%) of scattering.
Table IV: Values of $\rho^*$ corresponding to 10%, 50%, 90% of the total scattering:

<table>
<thead>
<tr>
<th>Exponents</th>
<th>-3.6</th>
<th>-3.8</th>
<th>-4.0</th>
<th>-4.2</th>
<th>-4.5</th>
<th>GL**</th>
<th>EX*</th>
</tr>
</thead>
<tbody>
<tr>
<td>10%</td>
<td>1.42</td>
<td>1.04</td>
<td>0.78</td>
<td>0.60</td>
<td>0.52</td>
<td>0.22</td>
<td>1.20</td>
</tr>
<tr>
<td>50%</td>
<td>5.60</td>
<td>3.80</td>
<td>3.01</td>
<td>2.46</td>
<td>2.10</td>
<td>1.22</td>
<td>2.70</td>
</tr>
<tr>
<td>90%</td>
<td>31.7</td>
<td>48</td>
<td>19</td>
<td>10.2</td>
<td>6.6</td>
<td>4.3</td>
<td>5.50</td>
</tr>
</tbody>
</table>

* when $n=1.05$ and $\lambda=419\text{nm}$, the diameter (in $\mu\text{m}$) is numerically equal to $\rho$.

** the two last columns correspond to values of $\rho$ in the case of a log-normal (GL) and an exponential (EX) distribution respectively, whose equations have been given in Part II (legend of Fig. II.16). These distributions approach that of the power law when the exponent is -4.

From the same Figure III.8 additional conclusions can be drawn. For example, the fact that the smaller particles are inaccessible to experiment and especially to counting, does not render the computation of scattering impossible since an upper limit to the error may be determined. Along the same level of ideas, if it can be determined that the power law does not extend to the very small sizes and that there do not exist particles which are continuously smaller and more numerous, which is a certainty, the abandonment of this hypothesis only slightly affects the result; the location of the curve at 1% determines the order of magnitude of this effect. The results obtained using the other distribution laws confirm this point.

On the other hand, the total scattering coefficient which enters in the phenomenon of light penetration in the ocean, i.e. in an infinite medium, is probably larger than that which can be measured with an apparatus utilizing a limited scattering volume. In this case the large particles which generate an intense and erratic signal are not integrated in the measurement. However it is reasonable to consider that the change corresponds to an increase of the coefficient by only a few percent.
3.1.2 Case of the volume scattering functions.

The preceding may be generalized, to consider not only the total scattering coefficient but the volume scattering function too. The notion of the "dominant" particle size class persists but the limits or the median value of this class vary depending on the angle. The progressive values of the integrals giving the volume scattering function (Part II, Fig. II.12) enable the determination of the values of $\alpha$ for which the scattering coefficient has attained a given fraction of its final value (assumed to be asymptotic). It is immediately evident that as far as the large angles are concerned ($\theta$ from $90^\circ$ to $180^\circ$), the greater portion of scattering is caused by the smaller particles since their asymptotic plateau is reached very rapidly; in the opposite case the effective class is displaced towards the larger sizes where small angle scattering is concerned. It may be stated therefore that each portion of the scattering function is the preferential expression of a given class of particles, the smaller angles corresponding to larger particles. The same Figure II.12 enables the determination that these classes cover an ever larger size interval, the smaller the initial slope is, i.e. the greater the exponent (in absolute value). This enlargement of a class is accomplished by a decrease of the lower limit & its displacement to smaller sizes.

It is convenient to use as before the median size, in placing this class in the size scale (it is the value of $\alpha$ for which the volume scattering function under consideration attains 50% of its final value). Figure III.9 valid for an index of refraction of 1.05, demonstrates the variations of this median size with exponent, for various values of the scattering angle $\theta$. The median size varies in the approximate ratio of 30 as the scattering angle goes from $140^\circ$ to $2^\circ$; the value of this ratio is little affected by the value of the exponent, at least when it is smaller than -3.70. The median size corresponds to a diameter of 0.1$\mu$m (for $\lambda=419$nm) for large angle scattering, $90^\circ$ or $140^\circ$, while the particle class around 3 to 4$\mu$m is dominant for scattering at $2^\circ$. 
Figure III. 9: Values of $\alpha$ corresponding to the median size, as a function of the exponent, for the indicated scattering angles. (The median size is that for which the volume scattering function under consideration attains 50% of its final value, when $\alpha = 200$). The index of refraction is 1.05.

Holding now the value of the exponent fixed, it is possible, to determine the "effective" class for each angle, in a better way than by its midpoint. It is convenient to limit for example this class by the values of $\alpha$ for which the volume scattering function attains approximately 10% and 90% of its final value. This is schematically illustrated in Figure III. 10 (the diameter scale is deduced from the $\alpha$ scale setting $\lambda = 419$nm).

Figure III. 10: The edges of the rectangles correspond to values of $\alpha$ for which the particular element of the volume scattering function has attained 10% and 90% of its final value (being 100%, when $\alpha = 200$). The median size is marked by an asterisk. The upper panel corresponds to the power law with a -3.9 exponent, while the lower panel to a log-normal law whose equation has been given (Part II, Fig. II. 16).
As far as the power law distribution is concerned, (in this example exponent -3.90), the displacement of the effective class and its enlargement occur progressively as $\theta$ increases. It is determined that it is the smaller particles with diameters less than 1$\mu$m which produce almost all the scattering at 140° or 90°, i.e. the particles which are most inaccessible to experiment. On the contrary the 2 to 5$\mu$m particles are predominant in scattering at 2°. If another wavelength is used these values will be proportionately modified, since the classes are defined in relation to $\alpha$; thus the previous diameters are doubled if the wavelength itself is doubled (838nm). This matter, directly related to spectral selectivity, will be taken up further (Section 3.2). The result is quite different with the log-normal distribution. The preference effect is less prominent than in the case of the Junge distribution. This is understandable, since in relation to the latter, the former distribution is in a sense truncated both at small and large sizes, and through this enhances the importance of the intermediate particles in scattering; it is a consequence of what was remarked previously (Part II, Section 3.2.5).

3.1.3 Special case at 0° angle

In all the preceding analysis, no mention has been made about scattering at 0° and more generally at very small angles. In fact when a computation is performed for a population whose exponent is less than 5 in absolute value, the integral expressing the scattering coefficient at 0° does not converge (cf. Part II, Section 3.2.1). This absence of an asymptotic value renders the definition of an effective class at this angle quite arbitrary; more accurately and through continuity, this class should correspond to the largest particles, i.e. the last ones to be included in the integration. However, several conclusions can be drawn from the theory. On Figures such as II.12 where the progressive values of the integrals

$$\int i_T(\theta, \alpha) F(\alpha) \, d\alpha$$

are given, the curve at 0° continues to grow with a slope of 5-m, whereas the curve at 2° enters a plateau for a value $\alpha_2$ of the parameter $\alpha$, which depends on the index* and is of the order of 40 if the index is

---

*As it has been seen previously, the value of $\alpha$ must be such that the parameter $\rho=2\alpha(n-1)$ be equal to 4.1 (Part II, Section 2.2.4 and 2.2.7).
1.05 for example. It is therefore immediately possible to deduce what the size $\alpha_0$ of the largest particles must be in order that the ratio $i(0^\circ)/i(2^\circ)$ attain a given value $k$; in fact:

$$\left( \frac{\alpha_0}{\alpha_2} \right)^{5-m} \approx \frac{i(0^\circ)}{i(2^\circ)} \approx k$$

An attempt can be made to at least approximately apply this relation to the measurements of T. J. Petzold (1971). The results of this author do not refer to $0^\circ$ but to $0.1^\circ$. The ratio of $i(0.1^\circ)$ to $i(2^\circ)$ is of the order of $10^2$ and assuming $k$ to be of the same order (using 1.05 for the index):

$$\left( \frac{\alpha_0}{40} \right)^{5-m} = 10^2$$

which leads to $\alpha_0 = 4000$ if $m=4$ and to $\alpha_0 = 862$ if $m=3.5$. This estimate, approximate as it may be, nevertheless demonstrates that the upper limit of integration must be pushed to values beyond $\alpha=200$, in order to account for the experimental results at very small angles.*

This also suggests emphasizing the relative or inadequately defined character of the scattering function in this particular angular domain because the shape probably depends on the spatial scale considered. Thus, the scattering function entering the computation of long-distance transmission or even the scattering function deduced from the transfer function of spatial modulation (W. H. Wells, 1970) could present a much more "pointed" lobe than does a scattering function determined using an experimental apparatus with a much shorter optical path. Simple geometrical considerations demonstrate that this conclusion does not contradict the fact that the total scattering coefficient does not increase at all when the scattering volume is increased (cf. Part III, Section 3.1.1). In fact, when the large particles enter, following the change of scale, this forward lobe which is ever more pointed, also becomes narrower. It is conceivable by continuity that in addition to scattering by particles in this case, scattering from inhomogeneities in index occurs, this time on a macroscopic scale, exactly similar to that of the fluctuations which induce them.

*Except perhaps for problems related to image transmission, the possibility of prediction makes the extension of the computations for $\alpha>200$ impractical before long, even with an adequately fast computer.
3.2 SPECTRAL DEPENDENCE OF SCATTERING

For a particle population, distributed according to a rectangular of triangular law, or equivalently according to a normal or log-normal law (cf. Part II, Section 3.2.5) the computation of the spectral selectivity of scattering presents no theoretical problem, since there is no problem of limits. For a distribution established in relation to true size (diameter) the change of wavelength implies an inversely proportional change of the relative size $\alpha$ --- and also of the parameter $\rho = 2\alpha(n-1)$ ---. This change affects simultaneously both the integrand and the limits of integration, in the expression giving the mean efficiency factor $\bar{\Omega}$ or the volume scattering function (Part II, equations 3.2 and 3.3 respectively). In order that the computation of $\bar{\Omega}$ be done with the parameter $\alpha$, or with the parameter $\rho$ to introduce the approximate expression of Van de Hulst (2.29, Part II), it is necessary to attribute a value to the index of refraction.

For an only slightly polydisperse population, the selectivity for (total) scattering is simple to predict. It depends on the size at which the distribution is centered, and also on the adopted value for the index; i.e. a judgement must be made with respect to the parameter $\rho$ and the value it obtains for the mean size. As shown on Figures II.10 and II.11 of Part II, if the mean size is such that the change of wavelength $\lambda$ always results in $\rho$ having values less than 4, the scattering will be selective with a negative exponent, i.e. $\lambda^{-\lambda}$. If for this variation of $\lambda, \rho$ remains very small, $\lambda$ will be equal to or close to 4; if $\rho$ remains between 0.1 and 2, $\lambda$ will be of the order of 2, it then becomes less than 2 and goes to zero as $\rho$ approaches 4. Finally if due to changes in wavelength, $\rho$ is between 4 and 7, the selectivity is inverted, i.e. it is expressed by a $\lambda^{+x}$ law. All these may be immediately deduced from the ondulations of the $\bar{\Omega}(\rho)$ curve. Finally when $\rho$ always remains high the selectivity is practically zero. If the population is no longer just slightly polydisperse (for example, if the geometric type growth is large enough when the distribution is expressed by a log-normal law) the preceding selectivities may persist but due to the medium, they are more attenuated as the distribution becomes more polydisperse.
Applications of these principles have been researched in optical oceanography, especially by W. V. Burt (1955, 56). The main idea, in the sought for interpretation, consists in assuming that the distribution is log-normal, and then centering its maximum in the zone where $\rho$ is exactly less than 4, this is done in order to obtain a selectivity in agreement with experiment; in order to do this, the index must be fixed. The disadvantage of this approach is in assuming that the distribution does have a maximum and in making the position of this maximum dependent on the value of the index. Thus from the maxima of numbers of particles in the range 0.6 to 1.2 $\mu$m, Burt concluded that they would be, for example, ten times smaller if an index of 1.015 was assumed instead of 1.15. If one abstains from hypothesizing on the limits of the distribution and on the existence of a maximum, the problem arises of whether the theory remains workable and whether any selectivity whatsoever remains plausible. Along with these conditions, the influence of the choice of index will be examined.

3.2.1 General Case: Selectivity in the case of a non-limited Junge distribution.

This $\alpha^{-m}$ distribution, is assumed to extend over all particles from zero to infinite size*, the index of refraction, still undetermined, must remain constant in the range of wavelengths under consideration, so that for a given class of particles, the variations of $\rho$ depend on $\lambda$ alone. The following argument can be made: with the population remaining unchanged, if the wavelength is modified, $\lambda$ becoming $\lambda'$ with $\lambda'=k\lambda$ ($k>1$ for example), it is the particles that are $k$ times larger than previously which play the same role in regard to scattering. More precisely, there existed a class of particles of diameter $d$, in an interval $\Delta d$ for which $Q$ had a certain value; when the wavelength changed, this same value of $Q$ is attributable to the class of particles of diameter $kd$ in an interval $k\Delta d$. According to the distribution law, the number of particles of dimension $kd$ is smaller than that of particles with dimension $d$ in the ratio $k^{-m}$, but the class itself is enlarged in the ratio $k$; finally, the number of particles in the "$kd$" class is $k^{1-m}$ times the corresponding class "$d$". These particles present a geometric cross-section $k^2$ times greater, thus the total surface presented by the class "$kd$" of width $k\Delta d$ is $k^3-m$ times that of the class which initially played the same role.

*The particle number is obviously infinite, which does not refute the statement that scattering can be finite, under certain conditions as has already been shown (Part II, Section 3.2).
Therefore \( Q \) is unchanged; thus the scattering produced by the surface through the efficiency coefficient, is itself multiplied by \( k^3 - m \). The argument applies to all the classes, and if all the classes exist, the selectivity of scattering by the ensemble of the population consequently follows a law:

\[
\lambda^{3-m}.
\]

This very simple result may appear to correspond to an ideal case and for this reason not to be appropriate for applications. The notion of "predominant" class with regard to scattering as developed previously (Section 3.1.1) enables the determination of application possibilities, by demonstrating the influence of hypotheses about the index and the limits. From this point on, it must be underlined that selectivity is not uniquely a property of weakly polydisperse suspensions.

3.2.2 Modification for the case of a limited distribution

The \( \lambda^{3-m} \) law applies to the integral

\[
\int_0^\infty F(\rho) Q(\rho) \rho^2 d\rho
\]

If the limits are no longer 0 and \( \infty \) but \( \rho_m \) and \( \rho_M \), the same continues to apply but an additional effect appears. In fact the value of the integral is itself modified since the change of wavelength implies a change in limits, which are tied to the diameters \( d_m \) and \( d_M \) rather than to \( \rho \); if \( \lambda \) becomes \( k\lambda \), \( \rho_m \) and \( \rho_M \) become \( \rho_m / k \) and \( \rho_M / k \). It is possible to predict what will happen, by going to Figure III.8, which clearly shows the so-called "effective" classes.

*The result may be easily rediscovered by a calculation beginning from the expression giving the mean efficiency \( \overline{Q} \) (Part II, Equation 3.2) and taking 0 and \( \infty \) as limits. It suffices to expand into a function of \( d \), \( n \) and \( \lambda \), in place of \( \rho \), and to substitute \( k\lambda \) for \( \lambda \). The denominator (total area) remains unchanged, the mean factor \( \overline{Q} \) is multiplied by \( k^3 - m \) if the distribution is expressed by \( F(d) = \text{const.} \cdot d^{-m} \).
To take a first example, assume a particle population extending between sizes 0 and \( \rho_M \) and that for a given wavelength \( \lambda \), \( \rho_M \) is such that scattering attains 95% of its possible maximum value (computed for \( \rho \) going to infinity). If the wavelength is doubled, \( \rho_M \) is divided by two, the integral will have a smaller value, corresponding to 90% for example. In consequence the selective effect of \( \lambda^{3-m} \) will be slightly modified (if the exponent 3-m is negative, the modification is in the direction of enhancing selectivity). The opposite example of a population for which \( \rho \) varies between \( \rho_M \) and infinity, shows that the same change in wavelength increases the value of the integral; the selectivity is diminished, in a more or less striking way, depending on the initial value of \( \rho_M \) (diminished whenever 3-m is assumed negative).

In a general way, it is conceivable that if the population encompasses mostly the effective classes, the selectivity law remains unchanged, otherwise it is more or less strongly affected. It remains the same, for example, if the range of sizes is extended in the following two cases: either when the relative index is assumed very close to 1 (which implies small values of \( \rho \)), or if the exponent of the distribution, as shown in Figure III. 8, approaches the value -3 (since then the notion of effective classes tends to disappear). In practice, following the conclusion regarding the plausible values of index and exponent (Section 2.4) and setting them at 1.05 and -4 respectively, the predominant classes responsible for 98% of the scattering (from 1% to 99%) correspond to diameters covering the range 0.1 to 100 \( \mu m \) (for \( \lambda = 419 \text{ nm} \)). Under these conditions, which are probably quite close to reality, the change in wavelength by several percent, will conserve the value of the integral and thus the selectivity law which will be approximately expressed by \( \lambda^{-1} \). Numerical examples are given elsewhere (Part III, Section 3.4).

3.2.3 Shape of the scattering function and wavelength

This discussion continues that of Part II (Section 3.2.1 and 3.2.2) regarding the influence of the limits on the result of the scattering function computation. As long as the convergence conditions are satisfied and the range of sizes is sufficiently broad, the shape of the scattering function is not essentially affected, as has already been seen, by the values attributed to the limits. The change of wavelength which implies a change of limits will therefore not have a noticeable effect, except again in the
immediate vicinity of 0° (if m < 5). In this respect, Fig. III 2 may be taken to represent the variation of the scattering function with variation of the wavelength in the ratio of 4 (more precisely, when $\alpha_M$ goes from 200 to 50, $\alpha_m$ would go to 0.05 and would not retain its value of 0.2, but this would not influence the result --- cf. Part II, Section 3.2.2 ---). If the Junge distribution is not effectively extended towards the small sizes but is truncated, a predictable effect would be the enhancement of the scattering function asymmetry as the wavelength decreases; the numerical values indicated previously* would permit its estimation.

3.3 POLARIZATION**

The polarization problem has been set aside up to this time; it has been only invoked when the experimental scattering function was compared to the theoretical one. As a commentary on the results obtained from the theoretical calculations, the following remarks will be given, which refer to both the individual scattering functions as well as those of polydisperse system:

a) The computations for the individual scattering functions have shown oscillations for the two polarized components $i_1(\theta)$ and $i_2(\theta)$ that are more prominent than those for $i_T(\theta)$, for which they are somewhat attenuated due to the averaging (cf. the examples in Appendix II). In addition, the polarization may be inverted, when $i_2$ ("horizontal" or "parallel" component) is greater than $i_1$ ("vertical" or "perpendicular" component); the degree of polarization (cf. Part II, Section 1.2) then

$$p(\theta) = \frac{i_1(\theta) - i_2(\theta)}{i_1(\theta) + i_2(\theta)}$$

becomes negative.

* cf. Part II, Section 3.2.2 "case where the truncation occurs beyond the Rayleigh domain".

** As has been mentioned in Part II, this matter is not seen in its broadest scope since in the preliminary calculations using Mie theory only the $i_1$ and $i_2$ components have been computed and not $i_3$ and $i_4$. Consequently, only the case where the incident light is natural will be considered and not the case where the incident light itself is polarized (linearly, circularly, ...).
b) the integration which permits consideration of a polydisperse system of particles produces smoothing. The normalized values \( \overline{\beta}_1(\theta) \) and \( \overline{\beta}_2(\theta) \) with respect to polarization (cf. Part II, equation 3.3) no longer produce the irregularity seen in \( \overline{\beta}(\theta) \) itself; whatever irregularity persists is probably due to the fact that the summation which in practice replaces the integration, is not carried to sufficiently many terms.

c) for the individual scattering functions, the polarization tends in a general way to be reduced for the small and large angles, and it vanishes theoretically at 0° and 180°. It is greatest at 90° in the small particle case, which comprises the Rayleigh and Rayleigh Gans domains. The integration extended to the total population does not eliminate these general traits. It should be remarked that, all things being equal (exponent, limits), the polarization is more pronounced as the index of refraction is closer to 1*; i.e. this is what is indicated by the separation between \( \overline{\beta}_1(\theta) \) and \( \overline{\beta}_2(\theta) \), particularly in the vicinity of 90°. As shown in Figures III.2 and III.7, the very accentuated minimum of \( \overline{\beta}_2(\theta) \) is centered around 90° for an index of 1.02, but for higher index values it is flatter and located at angles somewhat greater than 90°. Figure III.11 displays this effect.

![Graph](image)

Figure III.11: Theoretical values of the maximum degree of polarization as a function of the index of refraction. The population distribution and the limits remain unchanged (exponent -3.9, \( \alpha_m = 0.2, \alpha_M = 200 \)). The angles at which the degree of polarization is maximum are in parentheses.

* Which can be explained since the Rayleigh Gans domain extends to the relative sizes \( \alpha \) which can be large without violating the condition \( \rho < 1 \) (Part II, Section 2.2.3).
### 3.3.1 Influence of the limits and interpretation

Figure III. 2 illustrates the effect of the upper limit; when the upper limit $\alpha_M$ is taken from 50 to 200, the scattering function for natural light $\beta(\theta)$ is barely modified, and the same holds for the vertical component $\beta_1(\theta)$, whereas in contrast the maximum polarization decreases (and also the polarization at other angles), because the minimum of the horizontal component $\beta_2(\theta)$ is less accentuated.

Figure III. 12 demonstrates the opposite effect for the lower limit $\alpha_m$. As its value goes from 1.2 to 10, $\beta(\theta)$ is this time diminished, as has already been shown, except for the small angles, and the same holds for the component $\beta_1(\theta)$; in contrast the minimum of the scattering function for the $\beta_2(\theta)$ component remains unchanged.

Figure III. 12: Theoretical scattering functions computed for the indicated values of the parameters using the same scale and symbols as previously --- dots for $\beta_1(\theta)$, crosses for $\beta_2(\theta)$---. The two cases presented here approach that presented previously (Fig. III. 7) where the lower limit was $\alpha_m = 0.2$, while index and exponent are the same.

In summary, taking the typical case $\theta = 90^\circ$, the degree of polarization is in the first case reduced by augmentation of $\beta(90^\circ)$ while in the second case it is the result of the decrease of $\beta_1(90^\circ)$.

The interpretation of these effects is based on similar arguments to those given for the total intensity $i_T$ (it is limited to examination of polarization at $90^\circ$). The graph of the functions $i_1(90^\circ)\alpha^{-4}$ and $i_2(90^\circ)\alpha^{-4}$ demonstrates the different behaviour of these two quantities (Fig. III. 13).
Figure III.13: Variations of the functions $i_1(90^\circ) \alpha^{-4}$, $i_2(90^\circ) \alpha^{-4}$ and $i_T(90^\circ) \alpha^{-4}$ with the relative size $\alpha$, where $i_T = 1/2 (i_1 + i_2)$. The scales are logarithmic. This figure is similar to those presented in Part II (Fig. II.3 to II.5) or to that for an index of 1.05 (A. Morel, 1972a).

The decrease of $i_1(90^\circ) \alpha^{-4}$, on which $i_T(90^\circ) \alpha^{-4}$ is roughly based, occurs with a slope of around -2.3, for an index of refraction of 1.05. For the horizontal component, the corresponding quantity decreases more slowly, with a slope of about -1 which tends to zero for large values of $\alpha$, of the order of greater than 100. When the computation is made for the total population, the two integrals do not converge simultaneously (Part II, equation 3.3 and Section 32.1). The convergence is slower for the horizontal component (subscript 2) than for the vertical component (subscript 1); it may also not take place at all since the condition: $5+p-m<0$ (condition 3.4, Part II) may not be fulfilled in the first case (where $p<-1$ or 0) even though it might be in the second ($p>-2.3$). This is approximately the case illustrated in Figure III.14 (where $m = 3.9$);
the curve expressing the progressive values of the integral continues to increase where the horizontal component is concerned; in contrast the curve for the vertical component presents an asymptotic plateau.

Figure III.14: This figure is analogous to Fig. II.12 in Part II, but it is for the angle $\theta=90^\circ$ only. In addition to the curve giving the progressive value of the integral for the total intensity (dashed line), the two polarized components are also shown.

3.3.2 Conclusions - second remark on the values of the limits

In applying the above, $\bar{\beta}_1(90^\circ)$ is not essentially affected, just as $\bar{\beta}(90^\circ)$, except by a change of the lower limit $\alpha_m$, while $\beta_2(90^\circ)$ whose value is determined by the upper limit $\alpha_M$ is not affected by such a change. This is the situation in Figure III.12. On the contrary (the case of Figure III.2) the modification of the upper limit $\alpha_M$ affects only

*As previously, it must be assumed that the upper limit has a sufficient value, i.e. that the population is not just weakly polydisperse.
the component \( \bar{\beta}_2(90^\circ) \), to the exclusion of \( \bar{\beta}_1(90^\circ) \) and consequently of \( \bar{\beta}(90^\circ) \) also; as \( \alpha_\text{M} \) increases, \( \bar{\beta}_2(90^\circ) \) continues to grow and the polarization diminishes. In summary the influence of each of the two limits is exercised exclusively on one or the other of the two polarized components.

It can be said, that there exists an analogy between the forward scattering situation and the polarization at right angles, more precisely an analogy between \( \bar{\beta}(0^\circ) \) and \( \bar{\beta}_2(90^\circ) \), and for essentially the same reasons (cf. Section 3.1.3, Part III); their magnitudes continue to grow along with the upper limit (except if \( m > 5 \)).

When the experimental scattering function is compared to various theoretical ones it has been pointed out that at the same time, agreement may exist between the values for natural light --- \( \bar{\beta}(\theta) \) --- but not exist for polarization. This disagreement will not constitute an argument, and has not been considered in the conclusions regarding the values of exponent and index (Part III, Section 2.4); for example, the case in which the values of index and exponent are 1.02 and -4.2 respectively, leads to a very high degree of polarization, which can be much reduced by pushing the upper limit, without however affecting the values of \( \bar{\beta}(\theta) \). Thus the indicated values remain plausible, notwithstanding the disagreement regarding polarization. These considerations also have a negative aspect, by demonstrating the limits of the possibilities in interpreting the experimental results. The assumption and even the computation of a larger upper limit, as has been done to account for the observed scattering at small angles, is a procedure subject to caution whenever polarization is concerned. In fact, the extension of the \( i_2(90^\circ) \) curve, is not simply predictable as is that for \( 0^\circ \), the conclusions on the subject of the upper limit being for all practical purpose quite risky. It should also be mentioned, that the assumption of marine particles being spheres, even if it is justifiable for the total intensity, is less so in regard to polarization (cf. Preface, Part II and refer to Holland and Gagne, 1970).

3.4 RELATIONS BETWEEN SCATTERING AND PARTICLE CONTENT

The estimation of the concentration of material particles in seawater is justifiably one of the motivations invoked in undertaking systematic measurements of light scattering. However, in both theory and experiment, the establishment of quantitative relationships carries many
certainties. These arise from necessary hypotheses on the efficiency factor $\bar{Q}$, the index, the extremities of the size range considered, the total mass of the particles, etc. These difficulties have already been considered for example by Y.E. Otchakovsky (1965b) or G. F. Beardsley et. al. (1970), to cite only a few. It is useful to examine the influence of these hypotheses, before suggesting how and with what precautions the results of the computations may be applied.

3.4.1 General remark on the possible variations of the mean efficiency factor $\bar{Q}$.

The first problem consists of computing the total scattering coefficient, when the number of particles, counted between two limits, is known, as is the law which governs their distribution. The inverse problem is to deduce information on the particles (volume, surface, number) from observations of their scattering coefficient. In the first case, the scattering due to denumerable particles is perfectly computable, as long as the index is known (or is chosen); the problem nevertheless remains incompletely resolved because the experimentally observed scattering will be a priori greater, on the same sample, than the computed one, since it involves non-denumerable particles. The fact that there exist "effective" classes (cf. Section 3.1.1, Part III) demonstrates that in certain cases the influence of the limits is weak; an order of magnitude for the difference between computed and real scattering can be estimated. The resolution of the inverse problem implies even more hypotheses to the assumptions on the index and the limits is added that of the distribution law.

The relations that may be established are dependent on these hypotheses. To demonstrate this dependence, a resumption of the study of the variations in the mean efficiency factor $\bar{Q}$ is warranted, with varying index $n$, exponent $m$ and limiting diameters of the distribution $d_m$ and $d_M$ (or of the corresponding parameters, $\alpha_m \alpha_M$ or $\rho_m \rho_M$). For the index values considered, it is justified to use the limiting expression of Van de Hulst to compute $\bar{Q}$ (Part II, equation 2.29) and in this case the mean factor, more conveniently defined with respect to $\rho$, rather than $\alpha$, is written:
\[ \bar{Q} = \frac{\int_{\rho_m}^{\rho_M} \rho^2 \ F(\rho) \ \rho^2 \ d\rho}{\int_{\rho_m}^{\rho_M} F(\rho) \ \rho^2 \ d\rho} \]

Thus from a single computation, various applications may be performed, according to the value given to the index. (To recount --- cf. Part II, Section 2.2.10 --- the exact formula must be used to compute \( Q \) when \( \rho \) is small, in practice less than 0.2). Figure III.15 (A. Morel, 1972b) demonstrates the variation of \( \bar{Q} \) with the exponent of the distribution, where the various curves correspond to different pairs of values given to the limits \( \rho_m \) and \( \rho_M \).

Figure III.15: Behaviour of the mean scattering efficiency factor \( \bar{Q} \) with the exponent of the distribution. The numbers in parentheses correspond to the parameters \( \rho_m \) and \( \rho_M \) computed for the limits of the distribution.

When the exponent is equal to -2, all the particle classes present equal surfaces, the mean value \( \bar{Q} \) being in fact the average value of \( Q(\rho) \) (cf. Part II, Fig. II.10), therefore \( \bar{Q} \) is close to 2 as long as the upper limit is high enough \( (\rho_m = 10 \ \text{or} \ 20 \ \text{for example}) \) and, in contrast, whatever the value of the lower limit may be. \( \bar{Q} \) may be slightly greater than 2 if the chosen interval \( \rho_m - \rho_M \) is restricted and favors a maximum of the \( Q(\rho) \) curve. As the exponent increases (in absolute value), the increasing relative importance of the small particles, causes a decrease in \( \bar{Q} \). When \( m \) reaches and exceeds 4, the large sized particles and the upper limit \( \rho_m \) do not enter in practice, and the value of \( \bar{Q} \) is only dependent on the value of the lower limit.
Finally the factor $\bar{Q}$ may vary extensively with the exponent, with the limits and of course with the index and wavelength, since these parameters reflect on the values of the limits. This demonstrates the difficulties arising in applications, due to the hypotheses which have to be made, whereas the problem is perfectly resolved on the mathematical level.

3.4.2 Examples of applications

These various influences may be concretely determined by taking as an example a particle population distributed according to a power law in a size interval defined by the diameters $d_m = \mu m$ and $d_M = 20\mu m$. Taking into consideration the values attributed to the index and the wavelength, the corresponding values of $\alpha_m\alpha_M$ or $\rho_m\rho_M$ are given in the following table.

Table V: Correspondence between $d$, $\alpha$ and $\rho$ for two wavelengths and three index values.

<table>
<thead>
<tr>
<th>$d$ (m)</th>
<th>$\alpha$</th>
<th>$\rho$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$d_m = 1$</td>
<td>if $\lambda = 419\ nm$</td>
<td>$\alpha_m = 10$</td>
</tr>
<tr>
<td>$\rho_M = 20$</td>
<td>$\alpha_M = 200$</td>
<td>$\rho_M = 12$</td>
</tr>
<tr>
<td>$d_M = 4$</td>
<td>if $\lambda = 546\ nm$</td>
<td>$\alpha_m = 77$</td>
</tr>
<tr>
<td>$M = 20$</td>
<td>$\alpha_M = 154$</td>
<td>$\rho_M = 9.24$</td>
</tr>
</tbody>
</table>

Using the expression given above, it is possible to compute the mean factor $\bar{Q}$ between the limits provided by the pairs $\rho_m\rho_M$, for various exponents of the distribution law. The results of Table VI are obtained in this way. The indicated percentages correspond to the ratio of scattering, caused by particles in the interval $1-20\mu m$ to that of a hypothetical distribution of a non-limited population.
Table VI: Mean efficiency factor $\overline{Q}$ for various index of refraction and wavelength values, for particles between $1 \mu m$ and $20 \mu m$, distributed according to the power law with the indicated exponent.

<table>
<thead>
<tr>
<th>$\lambda = 419 \text{ nm}$</th>
<th>Exponents</th>
<th>(-3.6)</th>
<th>(-3.3)</th>
<th>(-4.2)</th>
<th>(-4.5)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n = 1.03$</td>
<td>72.3</td>
<td>1,282</td>
<td>79.2</td>
<td>1,089</td>
<td>76.9</td>
</tr>
<tr>
<td>$n = 1.05$</td>
<td>73.0</td>
<td>1,756</td>
<td>74.7</td>
<td>1,626</td>
<td>67.1</td>
</tr>
<tr>
<td>$n = 1.10$</td>
<td>63.3</td>
<td>2,307</td>
<td>57.5</td>
<td>2,338</td>
<td>46.0</td>
</tr>
<tr>
<td>$%$</td>
<td>$\overline{Q}$</td>
<td>$\overline{Q}$</td>
<td>$\overline{Q}$</td>
<td>$\overline{Q}$</td>
<td>$\overline{Q}$</td>
</tr>
<tr>
<td>$\lambda = 546 \text{ nm}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$n = 1.03$</td>
<td>69.7</td>
<td>1,028</td>
<td>76.3</td>
<td>0.828</td>
<td>79.3</td>
</tr>
<tr>
<td>$n = 1.05$</td>
<td>72.7</td>
<td>1,539</td>
<td>77.1</td>
<td>1,375</td>
<td>71.9</td>
</tr>
<tr>
<td>$n = 1.10$</td>
<td>67.8</td>
<td>2,171</td>
<td>64.1</td>
<td>2,148</td>
<td>58.0</td>
</tr>
</tbody>
</table>

This shows that in certain cases, especially when a large value must be attributed to the index, the portion "explained" through scattering is small. In a favorable case ($\lambda = 546 \text{ nm}$, $n = 1.05$, $m = 3.9$), at most 14% of the scattering would theoretically correspond to particles of a smaller diameter than $1 \mu m$ ($\rho < 0.77$), 9% to those of greater than $20 \mu m$ diameter ($\rho > 15.4$), while 77% of the total theoretical scattering would correspond to sizes between 1 and $20 \mu m$. This is tantamount to stating that in this case the particles under consideration essentially form the effective classes (cf. Figure III.8).

Figure III.16 presents the variations of the mean factor $\overline{Q}$ as a function of the index, for various exponents. Figure III.17 demonstrates that selectivity in scattering depends on the exponent. The curve corresponds to the theoretical non-limited distribution law, i.e. $\lambda^3 m$ (cf. Section 3.2.1), and the points locate the ratios $\overline{Q}_{419}/\overline{Q}_{546}$ characterizing the selectivity for the limited population* ($1-20 \mu m$).

* The percentages shown in Table VI permit in theory a determination of the values which would correspond to a non-limited distribution. The crosses on Fig. III.17 represent these values which would normally fall on the curve. A slight inaccuracy, produced by the method of computation, caused an underestimation and explains the slight disagreement.
Figure III.16: Variation of the mean efficiency factor $\bar{Q}$ with index of refraction, for the two indicated wavelengths. $\bar{Q}$ has been computed for a particle population with size limits of 1 $\mu$m and 20 $\mu$m which behaves according to a power law having the indicated exponents.

Figure III.17: Ratio of scattering at 419 nm to that at 546 nm demonstrating the selectivity which varies with the exponent of the population distribution (see text and the previous remarks).
The forward problem: computation of the scattering

An example is necessary for a practical application of the preceding: it is assumed that the total number of particles in unit volume, between two size limits of diameters \( d_m \) and \( d_M \) is known:

\[
N = \int d_m^M F(d) \, \text{d}(d)
\]

and it is also assumed that the distribution is expressed by \( F(d) = Ad^{-m} \). The total surface \( S \) of the geometrical cross-sections as well as the total volume \( V \) of the particles can be computed. To facilitate applications, it is convenient to divide \( S \) and \( V \) by the number of particles \( N \); thus the following relations are obtained:

\[
\frac{S}{N} = \frac{\pi}{4} \left( \frac{d_M^{m-1} - d_m^{m-1}}{d_M^{m-1} - d_m^{m-1}} \right) \quad \frac{\left(\frac{d_m}{d_M}\right)^{m-1}}{\left(\frac{d_m}{d_M}\right)^{m-1}}
\]

and

\[
\frac{V}{N} = \frac{\pi}{6} \left( \frac{d_M^{m-1} - d_m^{m-1}}{d_M^{m-1} - d_m^{m-1}} \right) \quad \frac{\left(\frac{d_m}{d_M}\right)^{m-1}}{\left(\frac{d_m}{d_M}\right)^{m-1}}
\]

(where the first equation assumes \( m > 3 \) and the second \( m > 4 \); if these conditions are not satisfied, modified expressions can be easily found).

Replacing \( d_m \) and \( d_M \) by 1 and 20\( \mu \text{m} \) and adopting \( m = 4.2 \), then \( S = N \times 2.04 \mu \text{m}^2 \) and \( V = N \times 5.77 \mu \text{m}^3 \). Chosing a concentration corresponding to clear water below the surface layer, \( N = 10^{10} \text{ m}^{-3} \) (being 10000 particles per \( \text{cm}^3 \), cf. J.C. Brun Cottan, 1971, H.R. Gordon, O.B. Brown, 1972) so that \( S = 2.04 \times 10^{-2} \text{ m}^2 / \text{m}^3 \) and \( V = 5.77 \times 10^{-8} \text{ m}^3 / \text{m}^3 \) (it may be noted that if a density of 1 is attributed to the particles, this volume concentration corresponds to 57.7\( \mu \text{g} / \text{liter} \)). Assuming the index to be 1.05 and using the values of \( Q \) from the preceding Table, the total scattering coefficient is estimated to be \( b = SxQ \) :

\[
b_{419} = 3.05 \times 10^{-2} \text{ m}^{-1}
\]

and

\[
b_{546} = 2.49 \times 10^{-2} \text{ m}^{-1}
\]
Returning to Figure III.8, it is seen that these computed coefficients for the particles between 1 and 20 \( \mu m \) can be 15 to 20% below the experimental coefficients, which in principle, correspond to a more extended distribution. This is the manner in which the scattering coefficients have been computed, assuming an index of 1.05, at 546nm for Mediterranean samples whose granulometry had been established by J. C. Brun Cottan (1971) --- the computation is in practice more complicated by the fact that in general two efficiency factors must be computed, one between 1 to 4\( \mu m \), the other between 4 and 20\( \mu m \) ---. Measurements of the volume scattering function at 30°, \( \beta(30°) \), have been made on the same samples, from which \( b \) can be deduced (cf. Part I, Section 6). The values of \( b \) are also in very good agreement with those obtained in situ using an integrating apparatus (D. Bauer, A. Ivanoff, 1971). The computed coefficients are compared to those deduced from measurement (Fig. III.18). The agreement is quite satisfactory for the measurements of November 1969, but is less so for those of June 1969, in particular for those samples withdrawn from 100 to 400 meters (an index of 1.03 is best suited to account for these measurements).

Figure III.18: Scattering coefficients computed from the granulometric information, compared to those obtained from measurements (see text). Bouée Laboratoire 42°14'N, 05°35'E.

* but without making the 15% to 20% correction which remains uncertain.
The inverse problem: use of the scattering measurements

Three hypotheses are necessary to resolve it. Choosing for example an index of 1.05, and assuming the distribution to have a -4 exponent and limits corresponding to sizes 0.5μm and 50μm (the lower limit is equivalent to truncation approximately produced by filtration through a millipore, HA filter), it is possible to establish the following relations, proposed with all the precautions imposed by the above choices. The mean efficiency factor for λ = 546nm is:

$$\overline{Q}(0.5 \rightarrow 50, 546) = 0.70$$

(which for λ=419 becomes Q=0.92). The total area of the geometrical cross-sections is

$$S(m^2/m^3) = b(m^{-1}) \times (1/0.70)$$

and from the expression giving S/N:

$$N(m^{-3}) = 10^{12} \times 1.715 \times S$$

and from the expression giving V/S --- which becomes V/S = 1/3 log 100, since the exponent in this case is -4, and the integral expressing V is a logarithm --- one obtains

$$V(m^3/m^3) = 10^{-6} \times 1.535 \times S$$

If for example it is desired to compare the results of a scattering measurement to a particle count between 1 and 20μm and also to that through filtration, assumed to be effective between the sizes 0.5 and 50μm, it is of interest to compare how the surface, volume and number is distributed in the three size classes 0.5 to 1μm, 1 to 20μm and 20 to 50μm. Assuming for example that b=0.07m⁻¹ at λ = 546nm, the total geometric cross-sectional area is 0.10 m²/m³ and for N, S and V the following values are obtained, expressed as a percentage of the total:
The relation between $b$ and $V$ ($b$ in m$^{-1} = 0.455 \times 10^{+6} \times V$ in m$^3$/m$^3$) is presented in Figure III.19 in which are plotted the scattering coefficients of samples (see also Fig. 6, A. Morel, 1970) as functions of their concentrations in dry particle weight ($10^{-9}$ g/g, i.e. in μg/liter). It appears that the ratio between dry weight and theoretical volume is much less than 1, which could be in any case explained by a high hydration of the particles. It is illusory to desire to make a more accurate determination, because on the one hand the relation has been established for the mean parameters (index and exponent) while on the other hand the filtration does not produce a true truncation (at $d=0.5\mu$m) but instead a progressive one which may also be variable.

![Figure III.19: Dry weight of particles retained on Whatman GF filters (measurements of G. Copin) compared to measured scattering coefficients --- through $B(30^\circ)$ --- on the same samples. The Atlantic measurements were taken during the Harmattan (1971) expedition.](image-url)
The Whatman G. F. filters produce a retention efficiency decreasing from 100% to 40% between the diameters of 4\(\mu\)m and 0.7\(\mu\)m respectively (R. W. Sheldon, W. H. Sutcliffe Jr., 1969). This point must not be forgotten when comparing dry weights, either among themselves, or with scattering measurements, especially when considering that an important fraction of particles in terms of weight or volume may be located in the small size domain. According to Lisitzin and Bogdanov (1968), 40% to 60% of the particle mass is from particles with less than 1\(\mu\)m diameter, which assumes an extension of the \(-4\) exponent law to diameters 0.02\(\mu\)m; with this exponent, there occurs logarithmic equipartition of the volumes, so that for example, the two classes 0.02-1\(\mu\)m and 1-50\(\mu\)m, have the same total volume.

END OF PART III
CONCLUSION

It would be an illusion to expect total agreement between experiment and theory, for the situations arising in the natural sciences, the theory being by necessity simplified. It is however useful to propose a somewhat simple theoretical model which may constitute a valid approximation to a complex reality, and in relation to which the experimental departures may be interpreted. To accomplish this it is necessary to clearly determine the effect of diverse factors; which explains the importance given to them in a previous section, as well as that given to the individual scattering functions.

This somewhat preliminary aspect of research is not always indispensable. In atmospheric optics for example, Deirmendjian (1969) has not done it, in the sense that experimental facts on aerosols are much more complete. In optical oceanography it is not so, and in fact the inverse problem is posed, which is that of obtaining presently lacking information on suspended particles, which would at least permit their rapid investigation through optical means, but whose meaningfulness must be justified.

Despite its limitations, the model accounts for a good number of properties satisfactorily. The shape of the computed scattering functions is compatible with what is already known about the particles; a simultaneous study of scattering and granulometry would permit determination of the effective index, which is to say determination of the particle nature and of the state they are found in. The intensity of the scattering phenomenon and its spectral dependence have received a satisfactory explanation, opening the way for applications. For some applications simultaneous measurement of all the parameters, granulometric, chemical and optical remains desirable, since it may be stated by paraphrasing, that it is that theory which matters least, the less so if it is easily perfectible.

This work is dedicated to the memory of George F. Beardsley.

André Morel
May 1973
APPENDIX I: DEFINITIONS

The "Committee on radiant energy in the sea" of the International Association of Physical Oceanography has defined the physical quantities and fixed the terminology of optical oceanography (Chronicle of UGCI, No. 57, 1964). A proposition for the translations into French has been made (A. Ivanoff, A. Morel, 1970). In describing scattering properties various quantities have been defined.

QUANTITIES DEFINED BY THE COMMITTEE ON RADIANT ENERGY IN THE SEA

These are defined considering a beam of monochromatic light consisting of parallel rays. The relative loss of flux $\frac{dF}{F}$ is proportional to the traversed path $dx$, assumed elementary, and the proportionality coefficient is the attenuation coefficient $c$ (coefficient d'attenuation);

$$c = -\frac{1}{F} \frac{dF}{dx}.$$

The loss of flux is related to the effects of absorption and scattering:

$$c = a + b$$

where $a$ is the absorption coefficient (coefficient d'absorption) and $b$ the total scattering coefficient (coefficient (total) de diffusion) where:

$$a = -dF(\text{abs}) \frac{1}{F} \frac{1}{dx} \quad \quad b = -dF(\text{scat}) \frac{1}{F} \frac{1}{dx},$$

where $dF(\text{abs})$ is the reduction of flux by absorption and $dF(\text{scat})$ the flux removed from that which would normally be transmitted in the direction of the beam, and which is in fact dispersed in space.

A section of the beam, of length $dx$ and volume $dv=Sdx$, observed from far enough, behaves like a point source of light, having an intensity $dI(\theta)$ in a direction forming an angle $\theta$ with the initial direction of the beam; $dI(\theta)$ being proportional to the volume scattering function $\beta(\theta)$ (coefficient angulaire de diffusion):

$$\beta(\theta) = 2 \frac{\int I(\theta) \frac{1}{F} \frac{1}{dx}}{\frac{1}{dx}}, \quad \text{or} \quad \beta(\theta) = \frac{dI(\theta)}{\frac{1}{F} \frac{1}{dv}}.$$
where $E$ is the illumination on the front surface $S$ of the scattering volume $\mathrm{dv}$ ($E \, \mathrm{dv} = F \, \mathrm{dx}$).

The coefficients $a, b, c$, with dimensions of inverse length, are expressed in oceanography in $\text{m}^{-1}$, and the volume scattering function in $\text{m}^{-1} \text{str}^{-1}$. Integrating over all space the flux scattered in all directions $\theta$, the following relation is obtained:

$$b = \int_{4\pi} b(\theta) \, d\theta, \text{which becomes} \quad b = 2 \pi \int_{0}^{\pi} b(\theta) \sin \theta \, d\theta,$$

if the scattering phenomenon has a symmetry of revolution around the axis of the initial beam. Integration over non-elementary paths, in a homogeneous medium (constant $a$ and $b$) leads to exponential decrease of flux by absorption or scattering or both.

The presentation of the volume scattering function $b(\theta)$ as a function of $\theta$ has been named in French "indicatrice de diffusion".

OTHER QUANTITIES USED IN THE THEORIES.

Intensity functions (Fonctions d'intensite) $i_1$ and $i_2$.

Denoting by $I_o$ the intensity of incident light, natural (and monochromatic), propagating as a plane wave (parallel beam of light), and by $I$ the intensity scattered in a direction $(\theta, \varphi)$ at a distance $D$ (assumed large) from the particle:

$$i = i_o \frac{1}{k^2 b^2} F(\theta, \varphi),$$

where $F(\theta, \varphi)$ is a dimensionless function of the direction $(\theta, \varphi)$, $k$ is the wavenumber $k = 2\pi/\lambda$, $k^2$ having dimensions $L^{-2}$. If there exists symmetry of revolution, as in the case of spherical particles considered previously, only one angle $\theta$ enters, measured between the directions of propagation of the incident and the scattered wave; these two directions define the "scattering plane" (plan de diffusion). If the incident light is natural, the scattered light is in general polarized,

$$i = i_o \frac{1}{k^2 b^2} \left( \frac{i_1(\theta) + i_2(\theta)}{2} \right).$$
where \( i_1(\theta) \) and \( i_2(\theta) \) are the intensity functions.

They are dimensionless quantities, obtained as the squared modulus of the complex amplitudes \( S_1(\theta) \) and \( S_2(\theta) \); the subscript 1 (or sometimes \( r \) or \( l \)) refers to the vibration perpendicular to the scattering plane, and the subscript 2 (or \( l \) or \( = \)) to vibrations in the scattering plane.

The ratio \( \frac{i_1 - i_2}{i_1 + i_2} \) expresses the degree of polarization, produced by scattering when the incident light is natural (cf. Part II, Section 1.1 and 1.2).

Angular Mie scattering coefficient. (Coefficients angulaires d'efficacite).

Scattering efficiency factor. (Coefficient d'efficacite pour la diffusion).

A dimensionless number, expressed in sterad\(^{-1}\), defined as follows:

\[
q(\theta) = \frac{i_1(\theta) + i_2(\theta)}{2} \frac{1}{k^2} \frac{1}{S},
\]

where \( S \) is the geometric cross-sectional area, i.e. the particle projection on the incident wave plane. For a spherical particle \( S = \pi r^2 \), and introducing the size parameter \( \alpha = 2\pi r/\lambda \).

\[
q(\theta) = \frac{1}{2\pi r^2} \left( i_1(\theta) + i_2(\theta) \right),
\]

which may be interpreted as the flux scattered by a single particle, per unit solid angle, in a direction \( \theta \), divided by the flux incident on the geometric cross-section (\( \pi r^2 \) here). Integrating over all space

\[
\int_0^{2\pi} \int_0^\pi q(\theta) d\Omega = \frac{1}{2\pi r^2} \int_0^\pi \left( i_1(\theta) + i_2(\theta) \right) \sin \theta \, d\theta = Q,
\]

the number \( Q \) is obtained, which is the scattering efficiency factor: it is the ratio of the total scattered flux to that incident on the particle.
Effective cross-section (section efficace pour la diffusion) and  
Differential cross-section (sections efficaces "partielles")

The differential cross-section in the direction $\theta$ is defined by

$$s(\theta) = S q(\theta) = \frac{\lambda^2}{8\pi^2} (i_1(\theta) + i_2(\theta)),$$

with dimension $L^2$ in units of $m^2\text{sterad}^{-1}$. The integral over $4\pi$ furnishes the effective cross-section $s$:

$$s = \frac{\lambda^2}{8\pi^2} \int_0^\pi (i_1(\theta) + i_2(\theta)) \sin \theta \ d\theta = \frac{\lambda^2}{4\pi} Q \ a^2,$$

where $s = \pi r^2 Q$ for a sphere and more generally $s = S Q$ where $S$ is the area of the geometric cross-section.

Volume scattering function (coefficient angulaire de diffusion) and  
Total scattering coefficient (coefficient total de diffusion).

The sum of the differential cross-sections, relative to $\theta$, of $N$ particles contained in unit volume, constitutes the volume scattering function. In the spherical particle case, which are also of identical radius $r$,

$$\beta(\theta) = N s(\theta) = N \pi r^2 q(\theta), \quad \beta(\theta) = \int \frac{\lambda^2}{8\pi^2} (i_1(\theta) + i_2(\theta))$$

where $N$ has dimensions $L^{-3}$, $\beta$ has $L^{-1}$, and units of $m^{-1} \text{sterad}^{-1}$. It is the ratio of the incident flux to that scattered in the direction $\theta$ per unit solid angle, assuming unit scattering volume.

Integrating over all space leads to the total scattering coefficient $b$:

$$b = \int_0^{\pi} B(\theta) \ d\theta = 2 \pi \int_0^\pi s(\theta) \sin \theta \ d\theta, \quad (B = \pi r^2 Q \text{ for spherical particles})$$

which is the ratio of the total scattered flux to that incident on unit volume; with dimension $L^{-1}$ and units $m^{-1}$. If the spherical particles are not of uniform size:

$$s(\theta) = \int_0^\infty \pi(r) \pi r^2 q_0(r) \ dr,$$
where

\[ \int_0^n n(r) \, dr = N \quad \text{and} \quad b = \int_0^n n(r) \pi r^2 q(r) \, dr . \]

Phase function (fonction de phase) and normalized volume scattering function (coefficient angulaire normalisé)

The phase function is also a dimensionless function defined as:

\[ P(\theta) = 4\pi \frac{\eta(\theta)}{q} = 2 \frac{i_1(\theta) + i_2(\theta)}{q a^2} \cdot \]

In addition we may define:

\[ P_1(\theta) = \frac{4 \pi i_1}{q a^2} \quad \text{et} \quad P_2(\theta) = \frac{4 \pi i_2}{q a^2} . \]

For a single particle:

\[ P(\theta) = 4\pi \frac{s(\theta)}{a} , \]

and for a scattering volume:

\[ P(\theta) = 4\pi \frac{\eta(\theta)}{b} \]

and in all cases:

\[ \int_0^{4\pi} P(\theta) \, d\Omega = 4\pi . \]

It is more convenient to use the normalized volume scattering function defined as:

\[ \overline{E}(\theta) = \frac{P(\theta)}{4\pi} = \frac{s(\theta)}{a} , \]

and consequently

\[ \int_0^{4\pi} \overline{E}(\theta) \, d\Omega \equiv 1 \quad , \quad \overline{E}_1(\theta) = \frac{i_1}{4\pi q a^2} \quad \text{et} \quad \overline{E}_2(\theta) = \frac{i_2}{4\pi q a^2} \cdot \]

\[ \overline{E}(\theta) = \frac{1}{2} \left( \overline{E}_1(\theta) + \overline{E}_2(\theta) \right) . \]
Remark concerning polarization.

All the above formulae correspond to the case of natural incident light; the more general case where the incident wave has some state of polarization has been examined (Section 1.2 Part II) and 4 functions are used instead of 2 in describing the scattering phenomenon (by isotropic spheres):

\[ i_1 = s_1 s_1^* \]
\[ i_2 = s_2 s_2^* \]
\[ i_3 = \frac{i}{2} (s_1 s_2^* + s_2 s_1^*) = \text{Re} \{ s_1 s_2^* \} \]
\[ i_4 = \frac{i}{2} (s_1 s_2^* - s_2 s_1^*) = -\text{Im} \{ s_1 s_2^* \} \]

The formulae are related to the intensity and not the phases of the scattered wave which do not enter since \( i_1 \) and \( i_2 \) remain unchanged; the same holds for \( q(\theta) \), \( s(\theta) \), \( \tilde{p}(\theta) \) and their integral. It is possible however using the same formulae to define for example \( \tilde{C}_1 \), \( \tilde{C}_2 \), \( \tilde{C}_3 \) and \( \tilde{C}_4 \), with \( \beta = 1/2(\beta_1 + \beta_2) \). In fact some authors define and use the four normalized functions \( P_1(\theta)/4\pi \) \ldots \( P_4(\theta)/4\pi \). In this general case the degree of polarization has a different expression as shown in Section 1.2, Part II.
LIST OF THE PRINCIPAL SYMBOLS

The symbols presented here are those not defined above.
The paragraphs and equations (in Part II) where they appear for the first time are indicated in parentheses.

\[ a : \text{particle size parameter} \] (2.2)
\[ a_n, b_n : \text{Mic coefficients} \] (2.2, 2.4)
\[ a_{ij} : \text{scattering matrix coefficients} \] (1.8)
\[ A_i, A_i^* : \text{Complex amplitudes. The asterisk denotes the complex conjugate.} \] (1.9, 1.11)
\[ A_1, A_2, B_1, B_2 : \text{parameters in the log-normal and exponential distribution laws.} \] (3.11, 3.12)
\[ a : \text{diameter} \]
\[ E_x, E_y : \text{components of the electric field } E \] (1.1)
\[ v_n, x_n, \zeta_n : \text{Ricatti-Bessel and Ricatti-Hankel functions} \] (2.4 and 2.8)
\[ r(\theta) : \text{angular distribution function (Rayleigh-Gans)} \] (2.21)
\[ r'(\theta) : \text{distribution function (diffraction)} \] (2.24)
\[ F(d), F(a), F(\rho) : \text{particle distribution functions} \] (Section 3.1, Part II)
\[ g(u) : \text{Gans function} \] (2.22)
\[ H_n, J_n, Y_n : \text{Hankel function of order } n, \text{Bessel functions of first and second kind of order } n. \]
\[ i_T : 1/2(i_1(\theta)+i_2(\theta)) \text{ see definitions} \] (Sec. 2.2.1, Part II)
\[ I_1, Q, U_1, V_1, I_2, U_2, V_2 : \text{Stokes parameters} \] (1.2, 1.11)
\[ k : \text{wavenumber } (=2\pi/\lambda) \] (Sec. 2.1, Part II)
\[ \lambda : \text{wavelength} \]
\[ \Lambda : \text{Lorentz-Lorenz term: } n^2 - 1/n^2 + 2 \] (2.15)
\[ m : \text{particle index of refraction relative to the medium in Part II.} \] (Sec. 2.1)
\[ m' : \text{Junge distribution law exponent for section 3.2} \] (Part II).
LIST OF THE PRINCIPAL SYMBOLS
(continued)

n: order of terms in the series  \( (2.2) \)

\( \eta \): relative index of refraction (see 'm' above)
beginning in Section 2.2 (Part II).  \( (\text{Sec. 3.1, Part II, and Sec. 3.4, Part III}) \)

N: total number of particles in unit volume

nm, \( \mu m \): nanometer and micrometer

p and \( p(\theta) \): degree of polarization  \( (\text{Sec. 2.1 and 3.2, PART I}) \)

p: slope of the \( i(\theta)\alpha^{-4} \) curves  \( (\text{Sec. 3.2, Part II}) \)

\( P_n \): Legendre polynomial of order \( n \)  \( (2.3) \)

Q: efficiency factor for absorption, scattering and attenuation

\( \bar{Q} \): mean scattering efficiency factor  \( (2.25 \text{ and } 2.28) \)

r: ratio of scattering of a sample to that of benzene.

\( R_p(\theta) \): characteristic ratio of particle scattering  \( (\text{Sec. 4.7, PART I}) \)

\( \rho \): parameter equal to \( 2\alpha|n-1| \)  \( (2.19) \)

\( S_1, S_2 \): complex amplitude functions

S and V: geometric cross-sectional surface and volume  \( (\text{Sec. 3.4, Part III}) \)

\( \theta \): scattering angle

END OF APPENDIX I
APPENDIX 2

COMPUTATIONAL PROCEDURE AND ADAPTATION
ONTO THE COMPUTER

1.0 COMPUTATION OF THE SCATTERING FUNCTIONS THROUGH
MIE THEORY

1.1 Useful relationships.

According to relations 2.2 from Mie theory, it has been seen
that the complex amplitudes of the scattered wave $S_1(\alpha, m, \theta)$ and $S_2(\alpha, m, \theta)$
are expressed in the form of series, which for each order combine:

--- the Mie coefficients $a_n$ and $b_n$ which depend on the relative size $\alpha$
and the relative index $m$ through the Ricatti-Bessel and Ricatti-Hankel
functions of order $n$ (2.4)

--- the functions $\pi_n$ and $\tau_n$, dependent only on the scattering angle $\theta$,
through functions containing Legendre polynomials of order $n$, using
$\cos \theta$ (2.3).

The generation of these functions is possible using the recurrence
relations, after setting the appropriate values to the first orders. This
computational method is similar to that used by R. Penndorf and
B. Goldberg (1956) and by D. Deirmendjian et. al. (1961); this author
has recently (1969) given a detailed description for the case where
the index of refraction is complex. A different computational procedure
utilizing the logarithmic derivatives of the Ricatti-Bessel functions can
be employed (see for example G. W. Kattawar and G. N. Plas, 1967).

The coefficients $a_n$ and $b_n$ - Recurrence for the Ricatti-Bessel and
Ricatti-Hankel functions

According to (2.5), (2.7), (2.9), the functions $\Psi_n$ and $\zeta_n$ and their
derivatives entering into $a_n$ and $b_n$ are expressed by:

$$\psi_n(x) = \left(\frac{x}{2}\right)^{1/2} J_{n+1/2}(x), \quad \zeta_n(x) = \psi_n(x) + i \chi_n(x),$$

with

$$\chi_n(x) = -\left(\frac{x}{2}\right)^{1/2} Y_{n+1/2}(x).$$

We set:

$$\psi_n(x) = S_n(x) \quad \text{et} \quad \chi_n(x) = \zeta_n(x).$$

Translator's Note:

* Equation numbers refer to Part II where MIE thoery is exposed.
Initially, we express these functions only through the Bessel function of the first kind, using

$$Y_{n+1/2}(x) = (-1)^{n+1} J_{-(n+1/2)}(x)$$

between the functions of the first kind $J$ and the second kind $Y$; thus $S_n(x)$ and $C_n(x)$ are written:

$$S_n(x) = \left(\frac{\pi x}{2}\right)^{1/2} J_{n+1/2}(x) \quad C_n(x) = (-1)^n \left(\frac{\pi x}{2}\right)^{1/2} J_{-(n+1/2)}(x)$$

For the functions of the first kind there exists the following relationship among three successive orders:

$$2 \frac{n}{x} J_n = J_{n-1} + J_{n+1} \quad \text{or} \quad J_n = 2 \frac{n-1}{x} J_{n-1} - J_{n-2}$$

and by replacing $n$ by $n + \frac{1}{2}$:

$$J_{n+1/2} = 2 \frac{n-1/2}{x} J_{n-3/2} - J_{n-1/2} \quad \text{or} \quad S_n(x) = \frac{2}{x} S_{n-1}(x) - S_{n-2}(x) \quad \text{(II.1 A)}$$

and by replacing $n$ by $-(n + \frac{1}{2})$:

$$J_{-n+1/2} = 2 \frac{n-3/2}{x} J_{-n+3/2} - J_{-n+1/2}$$

where:

$$(-1)^n \quad C_n(x) = 2 \frac{n-3/2}{x} \quad (-1)^{n-1} \quad C_{n+1} - (-1)^{n+2} \quad C_{n+2}$$

and downstepping by two orders:

$$C_n(x) = \frac{2}{x} C_{n-1}(x) - C_{n-2}(x) \quad \text{(II.1 B)}$$

which is identical to the relation established for $S_n$.

The computation can be conducted after setting the first two orders:

$$S_0(x) = \left(\frac{\pi x}{2}\right)^{1/2} J_{1/2}(x) = \sin x$$

$$S_1(x) = \left(\frac{\pi x}{2}\right)^{1/2} J_{3/2}(x) = \frac{\sin x}{x} - \cos x$$

$$C_0(x) = \left(\frac{\pi x}{2}\right)^{1/2} J_{-1/2}(x) = \cos x$$

$$C_1(x) = -(\pi x/2)^{1/2} J_{-3/2}(x) = \frac{\cos x}{x} + \sin x \quad \text{(II.2)}$$
In the expressions for $a_n$ and $b_n$, there also enter the derivatives $\Psi'$ and $\zeta'$, i.e., $S'$ and $S'+iC'$. There exists a recurrence relation between the Bessel function derivative of order $n$ and the functions themselves of order $n$ and $n-1$:

$$x J'_n(x) = -n J_n(x) + x J_{n-1}(x)$$

which, when applied directly to $S_n$ and $C_n$, provides the relations:

$$S'_n(x) = -\frac{n}{x} S_n(x) + S_{n-1}(x)$$

$$C'_n(x) = -\frac{n}{x} C_n(x) + C_{n-1}(x) \quad (II.3)$$

Finally, $a_n$ is expressed through $S_n$, $C_n$ and their first derivatives as:

$$a_n = \frac{S'_n(\infty) S_n(\infty) - n S_n(\infty) S'_n(\infty)}{S'_n(\infty) [S_n(\infty) + i C_n(\infty)] - n S_n(\infty) [S'_n(\infty) + i C'_n(\infty)]}$$

all of whose terms can be computed from the preceding formulae (the expression giving $b_n$ is analogous). If $m$ is real, all the functions have real arguments and during the computation of $a_n$, the real and imaginary parts are separable; setting:

$$p = S'(\infty) S(\infty) - m S(\infty) S'(\infty)$$

$$q = S'(\infty) C(\infty) - m S(\infty) C'(\infty) \quad (II.4)$$

permits writing:

$$a_n = \frac{p}{p + iq} ,$$

from which:

$$\text{Re} \{a_n\} = \frac{1}{1 + p^2/q^2} \quad \text{and} \quad \text{Im} \{a_n\} = \frac{-p/q}{1 + p^2/q^2} . \quad (II.5)$$

As has been seen, this implies that on the complex plane, all $a_n$ (or all $b_n$) lie on the circle of radius $1/2$, with the center at the point $\text{Im}=0$, $\text{Re}=0.5$ (see Fig. II.1). This point is important since it supplies a simple means for verifying any numerical computation. Another practical consequence relates to the adoption of a convergence criterion for the series $a_n$ or $b_n$: when for a sufficiently high order these numbers become small, the circle merges into a parabola and it necessarily happens that $\text{Im} \{a_n\} \sim \text{Re} \{a_n\}^{1/2}$. Assuming that convergence has been reached when the terms $a_n$ or $b_n$ become less than $10^{-7}$ for example (this is the value that was chosen), and applying this condition on the imaginary parts, the real part are then of the order of $10^{-14}$. 
The functions \( \pi_n \) and \( \tau_n \) - Recurrence for the derivatives of the Legendre polynomials.

The formulae (II.3) can be written otherwise, to relate \( \pi_n \) to \( \tau_n \):

\[
\pi_n (\cos \theta) = \frac{1}{\sin \theta} P_n^\nu (\cos \theta) = \frac{d}{d \cos \theta} P_n (\cos \theta)
\]

where we set:

\[
 x = \cos \theta \quad , \quad \pi_n (x) = P'_n (x) \quad , \quad (II.6)
\]

we obtain

\[
\tau_n (\cos \theta) = \frac{d}{d \cos \theta} P'_n (\cos \theta) ,
\]

where

\[
\tau_n (\cos \theta) = \frac{d}{d \theta} (\pi_n (\cos \theta) \cdot \sin \theta) ,
\]

\[
\tau_n (\cos \theta) = \frac{d}{d \theta} \pi_n (\cos \theta) \sin \theta + \cos \theta \cdot \pi_n (\cos \theta) ,
\]

\[
\tau_n (\cos \theta) = - \frac{d}{d \cos \theta} \pi_n (\cos \theta) \sin^2 \theta + \cos \theta \cdot \pi_n (\cos \theta) ,
\]

where

\[
\tau_n (x) = x \pi_n (x) - (1 - x^2) \pi'_n (x) \quad , \quad (II.7)
\]

so that \( \tau \) is explicitly expressed in terms of the function \( \pi_n \) and its derivative \( \pi'_n \), through the argument \( x = \cos \theta \).

In order to compute (II.6), \( P'_n (x) \) must be expressed through the lower orders. Among three successive orders, there exist for the Legendre polynomials the following relations:

\[
(n-1) P_{n+1} (x) - (2n+1) x P_n (x) + n P_{n-1} (x) = 0 ,
\]
from which

\[(n+1) P'_{n+1}(x) - (2n+1) P_n(x) - (2n+1) x P'_n(x) + n P'_{n-1}(x) = 0\]

Thus between the polynomial and its derivatives, there exists the following relation:

\[x P'_n(x) - P'_{n-1}(x) = n P_n(x)\]

which permits elimination of \(P_n\) between the two equations; we have:

\[n \cdot P'_{n+1}(x) - (2n+1) x P'_n(x) + (n+1) P'_{n-1}(x) = 0\]

and by stepping down one order (replacing \(n+1\) by \(n\)); the recurrence relation for \(\tau_n\) is written

\[\tau_n(x) = \frac{1}{n-1} \left( (2n-1) x \tau_{n-1}(x) - n \tau_{n-2}(x) \right) \quad (II.8)\]

which permits computation of \(\tau_n(x)\) from the values of the two previous orders.

In an analogous fashion, a recurrence relation for \(\tau\) is found where \(\tau\) and the two functions of the immediately preceding order \(\pi_{n-1}\) and \(\tau_{n-1}\) appear:

\[\tau_n(x) = (\tau_{n-1}(x) + \tau_n(x)) x + (n-1) x^2 \tau_{n-1}(x) \quad (II.9)\]

Through these relations (II.7) and (II.8), the functions \(\pi_n\) and \(\tau_n\) of whatever order, can be computed, setting initially the values for the first two orders (See 2.9A and 2.9B, giving these values).

1.2 Organization of the computation:

The initial data for the computation are:

--- the extreme values for the scattering angle \(\theta\) and the chosen step \(\Delta \theta\) (for example: 0(2)180).

--- a variable number of values for the size parameters: \(\alpha_1', \alpha_2' \ldots\) between which the step \(\Delta \alpha\) has a fixed value \(\Delta \alpha_1', \Delta \alpha_2' \ldots\)

--- one value for the index of refraction \(m\), which can be incremented by \(\Delta m\), after it is given an initial value at the beginning.
Computation of the coefficients $a_n$ and $b_n$

The coefficients are the first to be computed since they are the ones which determine the convergence of the series and determine the order to be reached.

Given the index of refraction $m$ and the size parameter $\alpha$, the functions $S$ and $C$ are computed for the first two orders through equations (II.2), and their derivatives $S'$ and $C'$ are deduced from relations (II.3). The real and imaginary parts of $a_n$ and $b_n$ are computed from the expressions (II.4) and then (II.5) and are stored in the computer memory.

After stepping one order, the values $S, C, S', C'$ are computed anew utilizing this time the recurrence relations (II.1.A), (II.1.B) and (II.3) and after computing this loop the new values $a_{n+1}$ and $b_{n+1}$ are stored in memory. The loop is repeated as long as necessary to ensure convergence to within a given precision which is decided in advance; it has been considered previously that convergence has been obtained when the generated terms $a_n$ and $b_n$ become less than $10^{-7}$. It has been seen that it suffices to impose this condition only on the imaginary part. It may be added also that it would suffice to apply it only to $\text{Im}\{a_n\}$, since it appears that numerically $b_n$ is always less than $a_n$, more precisely of the order of $a_{n+1}$.

When the condition is satisfied, the loop is terminated, and the following are stored in memory:

--- the highest order reached, say $N$.
--- the $N$ successive values of $\text{Re}\{a_n\}$, $\text{Re}\{b_n\}$, $\text{Im}\{a_n\}$, $\text{Im}\{b_n\}$,
--- the sum $\sum (2n+1) \text{Re}\{a_n + b_n\}$ which is progressively accumulated and serves in computing $S(0)\alpha^{-2}$ and the efficiency factor $Q$.

Computation of the angular values

Given an initial value of $\theta$, ($0^0$ or $180^0$ for example), the argument $x = \cos \theta$ is computed as well as the first two orders for $\tau$ and $\tau$ using the initial formulae presented. The values of $a_n$ and $b_n$ of the same order are recalled and the real and imaginary parts of the following are computed:

$$\{a_n \tau_n(x) + b_n \tau_n(x)\} \frac{2n+1}{n(n+1)}$$

and of

$$\{a_n \tau_n(x) + b_n \tau_n(x)\} \frac{2n+1}{n(n+1)}.$$
The same computation is repeated \( N \) times, with the difference that \( \tau_n \) and \( \tau_n' \) are now computed using the recurrence relations (II.6) and (II.7). When the computation is done, the following are placed in storage.

\[
\begin{align*}
\frac{2}{n(n+1)} \left( \Re \{a_n\} \tau_n + \Re \{b_n\} \tau_n' \right) &= \Re \{S_1(\theta)\}, \\
\frac{2}{n(n+1)} \left( \Im \{a_n\} \tau_n + \Im \{b_n\} \tau_n' \right) &= \Im \{S_1(\theta)\},
\end{align*}
\]

and the analogous expressions for:

\[
\Re \{S_2(\theta)\}, \quad \Im \{S_2(\theta)\}.
\]

The intensity functions \( i_1(\theta) \) and \( i_2(\theta) \) for the angle \( \theta \) are then computed as the square of the modulus:

\[
i_1(\theta) = (\Re \{S_1(\theta)\})^2 + (\Im \{S_1(\theta)\})^2 \quad \text{and} \quad i_2(\theta) = (\Re \{S_2(\theta)\})^2 + (\Im \{S_2(\theta)\})^2.
\]

At this stage \( i_T = \frac{1}{2} (i_1 + i_2) \), the polarization \( p = i_1 - i_2 / i_1 + i_2 \) and the phase functions \( P_1(\theta)/4\pi = i_1/\pi Q \alpha \) and \( P_2(\theta)/4\pi \) (the functions \( P_3, P_4 \) have not been computed).

Examples of scattering functions computed in this way are graphically presented on the following two pages.

**Further computations**

The angle \( \theta \) is then incremented by \( \Delta \theta \), and the second part of the computation is totally repeated, while the results of the first part (the coefficient \( a_n \) and \( b_n' \)) are successively recalled. When all the scattering angles have been explored, the computation for a given particle size is completed and the results removed from memory: according to the options, which are not mutually exclusive, they are transferred to magnetic tape, on punched cards, on a printout or perhaps plotted.
See following page
Examples of volume scattering functions, computed for the relative index 1.075 and the indicated values of the parameter $\alpha$. These figures help illustrate the material in Section 2.1.2 (evolution of the scattering functions), 2.2.3 (Rayleigh-Gans domain), and 2.2.5 (reflection-refraction). The curve has been drawn normalized at 0° (the ordinate is logarithmic), and the value at this angle $i(0°)$ as well as the efficiency coefficient $Q$ are indicated. The two polarized components $i_1(\theta)$ and $i_2(\theta)$ are indicated by the points and the crosses respectively. The curve corresponds to the total intensity $i_T(\theta) = 1/2 (i_1(\theta) + i_2(\theta))$. 
The program increments the size parameter $\alpha$ by $\Delta \alpha$ (determined in advance) and the whole computation is repeated from the beginning (new coefficients $a_n$ and $b_n$) and this is repeated up to reaching a pre-determined maximum value for $\alpha$.

Having treated this whole series of cases, it is possible to re-initialize the parameter $\alpha$ in a new loop, and all other things remaining the same ($\Delta \alpha$, $\Delta \theta$, extreme values of $\alpha$ and $\theta$), to increment the index. The classical structure, permits extension of the computations to a more or less large number of cases, according to the initial values chosen.

**Precision**

The criterion for truncation of the series $a_n$ and $b_n$ adopted here ($10^{-7}$) is the same employed by Penndorf and Goldberg (1956, 1960) and also used by Deirmendjian, Classen and Viezee (1961) (in the case of a complex index, it is transformed to $a_n^* b_n + b_n^* a_n < 10^{-14}$). The value of the criterion determines the attained precision for the value of the functions $i(\theta)$ and of those derived from them; it corresponds to six significant digits whatever the absolute value of the number may be. The numerical analysis does not permit absolute certainty in predicting the precision since the rate of convergence for the terms that were not computed, is not a priori predictable; it is also necessary to compare with other published results, or even which has already been done, to conduct verification by modifying the value of the criterion; this is the way that the stated precision can be confirmed.

The convergence of the series $a_n$ and $b_n$ is all the slower, as $\alpha$ increases. For all cases treated (index less than 1.15), an approximate empirical relation has been verified: $N \approx 1.07 \alpha + 10$ relating $\alpha$ to the order $N$ necessary to satisfy the criterion $< 10^{-7}$. A relation of this type has also been proposed to limit a priori the number of iterations to be performed (Penndorf); it cannot be general however since the value of the index enters also.

It is necessary to conduct the computation of the Mie coefficients $a_n$ and $b_n$ in double precision (17 significant digits). Without entering into the details of the numerical aspect, in the recurrence relations computing $S_n^i$ and $C_n^i$, the subtractions in fact introduce numerical indeterminacies (but not mathematical); since the generated terms do not differ until the eighth significant digit, it is impossible to compute differences in simple precision (in which case the memory reserved for each number consists of eight decimal places). This leads to division by zero which cannot be executed and the loop is infinitely repeated without ever satisfying the convergence criterion. The computation in double precision, which is indispensable when $\alpha$ is greater than 20 or 25, eliminates this difficulty at the price of quadrupling the execution time. Deirmendjian (1969) has recently underlined the necessity for this precaution.
2. COMPUTATION FOR POLYDISPERSE SYSTEMS

2.1 Organization of the Computations

There is no particular difficulty to conduct this computation. It can be organized in various ways. The one used most often may be briefly summed up as follows:

The integrals of the type giving $\beta_1(\theta)$ and $\beta_2(\theta)$, (3.2 and 3.3, PART II) are replaced by summations of terms corresponding to the 60 individual scattering functions previously computed; this also means that $d\alpha$ is not constant, it is small where it corresponds to particle classes abundantly represented and increases with reduction in numbers as size increases. The term $C(\alpha)$ is computed from the adopted distribution law (Junge or other). Thus the sums that must be computed are

\[
\begin{align*}
(1) & \quad \sum_{k=1}^{61} i_{kj}(\theta, \alpha) \cdot c_k(\alpha) , \\
(2) & \quad \sum_{k=1}^{61} \alpha^2 \cdot q(\alpha) c_k(\alpha) , \\
(3) & \quad \sum_{k=1}^{61} \alpha^2 \cdot c_k(\alpha) ,
\end{align*}
\]

The summations are conducted simultaneously with the computation of the 91 angular values considered, ranging from 0 to $180^\circ$ in steps of 2° (expression 1) and similarly for the other two expressions (2) and (3). To the 60 values of $k$ there correspond discreet values of $\alpha$. The term $C_k$ of order $k$ having been calculated by the distribution law, the 182 values of $i_{kj}(\theta)$, the value of $Q$ and $\alpha$ corresponding to the same order $k$ are recalled, then multiplied by $C_k$ and then accumulated with the analogous values computed for the previous orders, into 184 memory spaces corresponding to 184 summations to be conducted.

Following this, the operations indicated by 3.2 and 3.3 (Part II) are performed, as well as the associated computations of

\[
\begin{align*}
\bar{\beta}(\theta) &= \frac{1}{2} (\bar{\beta}_1(\theta) + \bar{\beta}_2(\theta)) \quad \text{and} \\
p(\theta) &= \frac{\bar{\beta}_1(\theta) - \bar{\beta}_2(\theta)}{\bar{\beta}(\theta)}
\end{align*}
\]
Optional exits permit knowledge of the partial results corresponding to truncations at the lower end (k minimum > 1) or the upper end (k maximum < 6l), or both together.

An automatic re-initialization changing the parameters of the distribution permit treatment of all necessary cases.

2.2 Computations below the lower limit $\alpha_m = 0.2$

It is justifiable to use the Rayleigh theory (see Sec. 2.2.2, Part II) which leads to table I below, where values of $i_1$ from Mie and Rayleigh theory are compared, for $\alpha = 0.2$ and $\theta = 0^o$ (taken as examples). In this case the intensity functions are written (see Eq. 2.15, Sec. 2.2.2 and Eq. 2.31, Sec. 2.2.10).

$$i_1(\theta) = a^6 \lambda^2, \quad i_2 = a^6 \lambda^2 \cos^2 \theta \quad \text{and} \quad q = (\frac{8}{3}) a^4 \lambda^2,$$

and the integrals can be rigorously calculated, from the arbitrary size 0 up to 0.2, in order to evaluate the maximum error, when the distribution is expressed by a power law (or also an exponential law). The following expressions are computed

$$\lambda^2 \int_0^{0.2} a^6 \left| \frac{1}{\cos^2 \theta} \right| a^{-m} \ da \quad \text{and} \quad (\frac{8}{3}) \lambda^2 \int_0^{0.2} a^4 \cdot a^2 \cdot a^{-m} \ da,$$

corresponding to (1) and (2). The value is to a close approximation

$$\frac{1}{T-m} \left| a^{-m} \right|_0^{0.2}$$

whose value is finite if $m > 1$ (See Part II, Sec. 3.2.2). As indicated in table I, these quantities are always negligible compared to those computed between 0.2 and 200. (For the computation it was assumed that there existed a particle for $\alpha = 1$, and that the exponent of the distribution was 3.9).

Table I:

<table>
<thead>
<tr>
<th>Indice</th>
<th>$i_1$ (0°)</th>
<th>$i_2$ (0°)</th>
<th>$i_1$ (0.2)</th>
<th>$i_2$ (0.2)</th>
<th>200 $i(0)a^{-m}da$</th>
<th>200 $i(0)a^{-m}da$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Rayleigh</td>
<td>Mie</td>
<td>Rayleigh</td>
<td>Mie</td>
<td>0,102 $10^{-6}$</td>
<td>0,102 $10^{-6}$</td>
</tr>
<tr>
<td>1,02</td>
<td>0,1130 $10^{-7}$</td>
<td>0,1131 $10^{-7}$</td>
<td>0,75 $10^{-6}$</td>
<td>1,0 $10^{-6}$</td>
<td>0,0102 $10^{-8}$</td>
<td>0,162</td>
</tr>
<tr>
<td>1,05</td>
<td>0,6986</td>
<td>0,7001</td>
<td>4,66</td>
<td>5,0</td>
<td>0,0744</td>
<td>0,369</td>
</tr>
<tr>
<td>1,075</td>
<td>1,5567</td>
<td>1,5619</td>
<td>10,31</td>
<td>10,0</td>
<td>0,165</td>
<td>0,534</td>
</tr>
</tbody>
</table>

For the computation it was assumed that there existed a particle for $\alpha = 1$, and that the exponent of the distribution was 3.9.
Table II:

<table>
<thead>
<tr>
<th>Exponent</th>
<th>(\int_{0}^{100})</th>
<th>(\int_{100}^{\infty})</th>
</tr>
</thead>
<tbody>
<tr>
<td>-3.6</td>
<td>93.0</td>
<td>7.0%</td>
</tr>
<tr>
<td>-3.8</td>
<td>97.4</td>
<td>2.6%</td>
</tr>
<tr>
<td>-4.0</td>
<td>99.0</td>
<td>1.0%</td>
</tr>
<tr>
<td>-4.5</td>
<td>99.35</td>
<td>0.65%</td>
</tr>
</tbody>
</table>

| Gausse log. | 90% pour \(\int_{0}^{0.5}\) | Expon. | 90% pour \(\int_{0}^{3.1}\) |

2.3 Computations beyond the upper limit.

This problem is posed in Part II (Sec. 3.2.3). Taking \(\rho\) instead of \(\alpha\), and assuming \(Q\) to be constant and equal to 2 when \(\rho > 100\), the integrals from 100 to \(\infty\) over \(Q(\rho)/\rho^{2-m}\) are immediately computable, furnishing the desired asymptotic values. For various values of the exponent, the ratios to the total value of table II are obtained, expressed in %; also shown are the values of \(\rho\) corresponding to 90% of the total scattering using the log-normal and exponential distributions considered.

END OF APPENDIX II
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