COMPUTATIONAL ANALYSIS OF THE EFFECTS OF SURFACE FILMS ON THE OPTICAL SCATTERING PROPERTIES OF BUBBLES IN WATER

By

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With a Supplement by Professor P. L. Marston on pages 151-156.

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**Title:** Computational Analysis of the Effects of Surface Films on the Optical Scattering Properties of Bubbles in Water

**Authors:** Stuart C. Billette and Philip L. Marston

**Abstract:**

A scattering algorithm for coated spheres was developed using the partial wave series of Aden and Kerker to theoretically determine if surface films on air bubbles in water have a significant effect on optical scattering patterns. The air bubbles were modeled as spheres (of radius $a$) coated by a film of uniform thickness $h$ and complex refractive index $n_c$ surrounded by water, with refractive index $n_w$ equal to 4/3. Size parameters of $ka$ (where $2\pi/k$ equals the optical wavelength in water) equal to 100, 500, 1000, and 2500 (corresponding to radii $a = 7.5$ μm to 189 μm for a wavelength in air = 632.8 nm) were modeled for values of $h$ ranging from 0 to 3 μm. Irradiance results are plotted as a function of scattering angle $\theta$ with emphasis on the critical scattering region. Comparison is made with known results from Mie theory for noncoated spheres as well as a physical-optics approximation developed by Marston and Kingsbury. Some modeling is done in terms of geometric ray optics, in order to explain observed effects. Finally, observations regarding the...
19. ABSTRACT (continued)

Use of noncoated versus coated bubble results for the sizing and detection of microbubbles in various experimental applications is discussed.
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Abstract

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August 1986

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Comparison is made with known results from Mie theory for noncoated spheres as well as a physical-optics approximation developed by Marston and Kingsbury. Some modeling is done in terms of geometric ray optics, in order to explain observed effects. Finally, observations regarding the use of noncoated versus coated bubble results for the sizing and detection of microbubbles in various experimental applications is discussed.
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To the memory of my father;

Richard Joseph Billette
CHAPTER 1

INTRODUCTION

1.1. Overview

In recent years a great deal of experimental laboratory research and computational modeling has been done regarding the behavior of light scattered by air bubbles in water.1-8 In these investigations the bubbles have generally been modeled as a non-absorbing uncoated sphere of given radius \( a \) and relative refractive index \( m \) less than 1. Recent oceanographic \textit{in situ} experiments, however, have suggested that bubbles found outside the laboratory may possess coatings estimated to range from 10 nm to 1 \( \mu \)m in thickness.9-13 If this is indeed the case, it becomes important to understand and model the effects of these surface films in order to establish the range of validity of the uncoated laboratory bubble results. In addition, if coated bubble results were found to vary significantly, the modeling done here could be useful in determining the size and nature of the coating found on the bubble which, at present, is still unresolved.

The approach taken in this study may be broken down as follows; first, an outline of the partial wave solution used for the case of a coated sphere is presented. This mathematical development gives some insight into the complexities involved in the calculation of scattering coefficients and also serves as a precursor for the actual computational results which follow in Chapter 2. In Chapter 2, the computational considerations involved in writing and testing the coated sphere algorithm are discussed. The results for the range of parameters tested are then presented along with a discussion of noted effects. A comparison is also made with Mie results for a noncoated sphere as well as a physical-optics approximation developed by Marston and Kingsbury.2 Using this
technique it was possible to determine the range of validity for the noncoated bubble results. An effort has been made to isolate each parameter and analyze it individually following the relevant computations as a greater sense of continuity is thereby maintained. Finally, a discussion of the experimental application of the overall results is presented.

1.2. Coated Sphere Scattering

The exact solution to the problem of the scattering of electromagnetic waves from concentric spheres was first presented in a paper by Aden and Kerker in 1951.\textsuperscript{14} It was based on a development presented in 1908 by Gustave Mie regarding the field scattered by any spherically symmetric particle of arbitrary radius and refractive index (commonly referred to as Mie theory). Although the solution to the problem of scattering by coated and noncoated spheres has been available for many years, it is only since the advent of large digital computers that it has become practical to use in scattering calculations. The reason for this will become apparent in the following development for an electromagnetic wave incident on a coated sphere.

Consider an electromagnetic plane wave which is incident upon a coated sphere of radius \( a \) and outer radius \( b \) as is shown in Fig. 1.1. The relative refractive index of the core is \( m_1 = n_i/n_o \) while that of the coat is \( m_2 = n_c/n_o \) where \( n_o, n_c \) and \( n_i \) are the refractive indices of the outer medium, coating and inner medium respectively. For our particular application of a coated air bubble in water \( n_o = n_w \), the refractive index of water and \( n_i = n_{air} \). In order to determine the fields in the given regions; \( 0 \leq r \leq a, a \leq r \leq b \) (\( a \neq 0 \)) and \( r \geq b \) it is necessary to solve the pertinent vector wave equations given by Maxwell's relations using spherical geometry and the appropriate boundary conditions.

The mathematics required to do this is straightforward but extremely tedious and therefore steps have been omitted and emphasis placed on the solutions. The reader is referred to
Fig. 1.1.—Electromagnetic wave incident upon a coated sphere.
texts by van de Hulst\textsuperscript{15} and Bohren and Huffman\textsuperscript{16} for a more thorough description. The following outline was taken from Chapter 8 of the Bohren and Huffman text.

Considering again the situation shown in Fig. 1.1, the solution of the scalar wave equations in spherical polar coordinates yields:

\begin{align*}
\psi_{\text{emn}} &= \cos m\phi \ P_n^m (\cos \theta) z_n(kr), \\
\psi_{\text{omn}} &= \sin m\phi \ P_n^m (\cos \theta) z_n(kr),
\end{align*}

where \( P_n^m(\cos \theta) \) are the associated Legendre functions and \( z_n \) is any of the four spherical Bessel functions \( j_n, y_n, h_n^{(1)}, \) or \( h_n^{(2)} \) depending on the region of interest. The subscripts \( e \) and \( o \) denote even and odd. Due to the completeness of the functions \( \cos m\phi, \sin m\phi, P_n^m(\cos \theta) \) and \( z_n(kr) \), any function that satisfies the scalar wave equation in spherical polar coordinates may be expanded as an infinite series in the functions (1) and (2). The vector spherical harmonics generated by \( \psi_{\text{emn}} \) and \( \psi_{\text{omn}} \) are

\begin{align*}
M_{\text{emn}} &= \nabla \times (r \ \psi_{\text{emn}}), \\
M_{\text{omn}} &= \nabla \times (r \ \psi_{\text{omn}}), \\
N_{\text{emn}} &= (\nabla \times M_{\text{emn}})/k, \\
N_{\text{omn}} &= (\nabla \times M_{\text{omn}})/k,
\end{align*}

which are written out in component form in Bohren and Huffman.\textsuperscript{17} Finally, the incident wave is taken to be of the form \( E_i = E_0 \ e^{i(kr - \omega t)} \hat{e}_x \) where \( \hat{e}_x \) indicates the \( x \) unit vector in spherical coordinates. In the following description we omit the \( \exp(-i\omega t) \) time dependence for all fields.

With this information it is now possible to write out the fields for the various regions of the coated sphere. The electromagnetic field \( (E_1, H_1) \) in the region from \( 0 \leq r \leq a \) is given by
\[ E_1 = \sum_{n=1}^{\infty} E_n[c_n M_{01n}^{(1)} - i d_n N_{e1n}^{(1)}], \quad (3a) \]

\[ H_1 = -k_1/\omega \mu_1 \sum_{n=1}^{\infty} E_n[d_n M_{e1n}^{(1)} + i c_n N_{01n}^{(1)}], \quad (3b) \]

where \( E_n = i^n E_0 (2n+1)/(n(n+1)), \mu_1 \) is the permeability of region 1, \( k_1 \) is the wave number of the wave with frequency \( \omega \) in that region and \( c_n, d_n \) are coefficients determined by the boundary conditions. The scattered field \((E_s, H_s)\) is given by

\[ E_s = \sum_{n=1}^{\infty} E_n[i a_n N_{e1n}^{(3)} - b_n M_{01n}^{(3)}], \quad (4a) \]

\[ H_s = k/\omega \mu \sum_{n=1}^{\infty} E_n[i b_n N_{01n}^{(3)} + a_n M_{e1n}^{(3)}], \quad (4b) \]

where again \( a_n \) and \( b_n \) are coefficients which will be determined by the appropriate boundary conditions. The (3) superscript indicates that the outgoing wave solutions in Eqs. (1) and (2) with \( z_n(kr) = h_n^{(1)}(k r) \) are to be used. Due to the requirement of finiteness at the origin, the radial part of the functions in Eqs. (1) and (2) which generate the vector harmonics in the expansion of \((E_1, H_1)\) is constrained to be \( j_n \) [indicated by (1) superscript]. However, in the region \( a \leq r \leq b \) \((a \neq 0)\), both the spherical Bessel functions \( j_n \) and \( y_n \) are finite; as a consequence the field in region 2 \((E_2, H_2)\) is written as

\[ E_2 = \sum_{n=1}^{\infty} E_n[f_n M_{01n}^{(1)} - i g_n N_{e1n}^{(1)} + v_n M_{01n}^{(2)} - i w_n N_{e1n}^{(2)}], \quad (5a) \]

\[ H_2 = -k_2/\omega \mu_2 \sum_{n=1}^{\infty} E_n[g_n M_{e1n}^{(1)} + i f_n N_{01n}^{(1)} + w_n M_{e1n}^{(2)} + i v_n N_{01n}^{(2)}], \quad (5b) \]
where the vector harmonics $M_{e1n}^{(2)}$ and so on, are generated by functions of the form of Eqs. (1) and (2) with radial dependence $y_n(k_2r)$. The boundary conditions

\[(E_2 - E_1)\hat{e}_r = 0, \quad (H_2 - H_1)\hat{e}_r = 0, \quad r = a\]

\[(E_3 + E_4 - E_2)\hat{e}_r = 0, \quad (H_3 + H_4 - H_2)\hat{e}_r = 0, \quad r = b\]

yield eight equations in the coefficients $a_n, b_n, c_n, d_n, f_n, g_n, \phi_n, \psi_n$:

\[f_n m_1 \psi_n(m_2x) - v_n m_1 \chi_n(m_2x) - c_n m_2 \psi_n(m_1x) = 0, \quad (6a)\]

\[w_n m_1 \chi'_n(m_2x) - g_n m_1 \psi'_n(m_2x) + d_n m_2 \psi'_n(m_1x) = 0, \quad (6b)\]

\[v_n \mu_1 \chi'_n(m_2x) - f_n \mu_1 \psi'_n(m_2x) + c_n \mu_2 \psi'_n(m_1x) = 0, \quad (6c)\]

\[g_n \mu_1 \psi_n(m_2x) - w_n \mu_1 \chi_n(m_2x) + d_n \mu_2 \psi_n(m_1x) = 0, \quad (6d)\]

\[m_2 \psi'_n(y) - a_n m_2 \phi_n^2(y) - g_n \psi'_n(m_2y) + w_n \chi'_n(m_2y) = 0, \quad (6e)\]

\[m_2 b_n \phi_n^2(y) - m_2 \psi_n(y) + f_n \psi_n(m_2y) - \nu_n \psi_n(m_2y) = 0, \quad (6f)\]

\[\mu_2 \psi_n(y) - a_n \mu_2 \phi_n^2(y) - g_n \mu_2 \psi_n(m_2y) + w_n \mu_1 \chi_n(m_2y) = 0, \quad (6g)\]

\[b_n \mu_2 \phi_n^2(y) - \mu_2 \psi'_n(y) + f_n \mu_2 \psi'_n(m_2y) - \nu_n \mu_2 \chi'_n(m_2y) = 0, \quad (6h)\]

where $m_1$ and $m_2$ are the refractive indices of the core and coating relative to the surrounding medium; $\mu$, $\mu_1$, $\mu_2$ are the permeabilities of the surrounding medium, core, and coating; and $x = k_2a$, $y = k_2b$. The prime indicates differentiation with respect to $x$ or $y$ in the argument. The Riccati-Bessel function $\chi_n(z)$ is $-z\gamma_n(z)$. Let us assume for simplicity that $\mu = \mu_1 = \mu_2$ and solve the set of above equations for the scattering coefficients $a_n$ and $b_n$.
Thus we find that the determination of scattering coefficients $a_n$ and $b_n$ becomes an impressive task. However, with the aid of a computer the computation can be accomplished as will be shown in Chapter 2.

For future reference it is desirable to define the following complex scattering amplitudes which are descriptive of the scattered fields:

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

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

The angular functions $\pi_n$ and $\tau_n$ are defined by the relations

$$\pi_n(\cos \theta) = \frac{1}{\sin \theta} P_n^1(\cos \theta),$$

$$\tau_n(\cos \theta) = \frac{d}{d\theta} P_n^1(\cos \theta),$$
where $\theta$ is the scattering angle. The scattered irradiances proportional to $|S_1|^2$ and $|S_2|^2$. The subscripts $j = 1$ and $j = 2$ on $S_j$ refer to the two orthogonal directions of incident polarization which are defined in Chapter 2.
REFERENCES TO CHAPTER 1


11. V. K. Goncharov, S. N. Kuznetsova, G. G. Neuimin, and N. A. Sorokina,
"Determination of the diffusion constant of a gas in sea water from the solution of air


13. R. E. Glazman, "Damping of bubble oscillations induced by transport surfactants
between the adsorbed film and the bulk solution," J. Acoust. Soc. Am. 76, 890-896


16. C. F. Bohren and D. R. Huffman, Absorption and Scattering of Light by Small
Particles (Wiley, N.Y., 1983).

17. Ref. 16, p. 89.

18. Ref. 16, p. 183. Note also that $\psi_n$ and $\xi_n$ are Riccati-Bessel functions of the kind
which commonly appear in the Mie solution with an $\exp(-i\omega t)$ time dependence:
$\psi_n(\rho) = \rho j_n(\rho)$ and $\xi_n(\rho) = \rho h_n^{(1)}(\rho)$. See, e.g., Ref. 16, p. 101.

19. Ref. 16, p. 112.
CHAPTER 2

COATED BUBBLE SCATTERING COMPUTATIONS

2.1. Computational Considerations

A computational scheme is presented in this section for the calculations of the scattered irradiances $I_j$, where $j = 1$ indicates polarization of the incident electric field vector perpendicular to the scattering plane and $j = 2$ indicates the parallel case. It is based on similar schemes developed by Wiscombe\textsuperscript{1,2} and Bohren and Huffman\textsuperscript{3} but differs in that it computes the coated sphere scattering irradiance as a function of scattering angle rather than just the efficiency factors. Another difference is that it also uses down recursion for all logarithmic derivative calculations.

Adopting the notation of Bohren and Huffman, we may calculate irradiances $(I_j)$ by way of Eqs. (8a) and (8b) using the values of the coefficients $a_n$ and $b_n$ determined in Eqs. (7a) and (7b). The logarithmic derivatives of the Ricatti-Bessel functions found in Eqs. (7c) and (7d) were calculated by the down recursion technique for Mie scattering developed by Wiscombe, which is known to be stable. This technique consists of specifying a priori the last term in the partial wave series denoted by $N$ and then using recursion relations to calculate down to the $n = 0$ term. By doing this the computational error is decreased in each step. The problem with the method, however, lies in selection of the $N$th term. Wiscombe, through extensive testing, has determined $N = kb + 4.05$ $(kb)^{1/3} + 2$ (where here $b$ is the radius of the outer sphere shown in Fig. 1.1) is the appropriate relation to determine $N$ and hence this was the expression used in our computations. Another necessary consideration was the preliminary determination of angle step size $N_{ang}$, which increases in proportion to $ka$. In order to resolve the fine
structure shown in Mie results, the value of $N_{\text{ang}} < \lambda/8a$ rad (where $a$ is the inner sphere radius shown in Fig. 1.1) was taken from previously published bubble results.$^4$

The partial wave scattering coefficients $a_n$ and $b_n$ were computed using the IBM FORTRAN VS program which is given in Appendix A. All computations were performed on an IBM 3090-200 computer. The program was written using IBM quadruple precision (32 significant digits) as this insured that the effects of the coating would not cause numerical instabilities. The complex arithmetic was done using IBM FORTRAN VS subroutines again using quadruple precision which allowed both the real and imaginary components in all calculations to have 32 significant digit precision.

The program was tested in a wide variety of ways. Limiting cases were taken in which the coating was allowed to go to zero thickness which gave the expected noncoated bubble Mie results previously published.$^5$ The coating was also given the same index of refraction as the surrounding medium (water) which again yielded the correct Mie results. Next, results were checked against other coated sphere calculations done by Cooper et al.$^6$ and Brunsting.$^7$ We were able to duplicate Cooper's results exactly for all cases and Brunsting's for scattering angles ranging from approximately $\theta = 0^\circ$ to $90^\circ$. Scattering coefficients and efficiency factors calculated by our program were checked against results from the subroutine written by Bohren and Huffman$^8$ and were also in perfect agreement.

Due to the use of quadruple precision and the need to calculate three logarithmic derivatives by down recursion the program is rather slow. Typical times ranged from approximately 10 c.p.u. seconds for $ka = 100$ with a 1 $\mu$m coating to roughly 150 c.p.u. seconds for $ka = 2500$ with a coating of 1 $\mu$m. Angle step size was typically on the order of $(\lambda/14 a)$ rad to avoid sampling errors for any size coating calculation. Contrast this the physical-optics approximation program (given in Appendix A), which is to predict
the coarse structure, executes in a few seconds regardless of the size of $k_a$. The equations used to develop this program are listed at the end of Section II of Ref. 9. Comparison of coated bubble cases will be made with the physical-optics approximation for forward scattering ($\theta = 0^\circ$ to $90^\circ$) to determine its usefulness in coarse structure calculations for coated bubbles.

2.2. Results

A. Effect of Increasing Coating Thickness

A typical set of calculations for a coated air bubble in water with $k_a = 100$ ($a = 7.5 \mu m$ for $\lambda_w = 474.6 \text{ nm}$) over the $\theta = 0^\circ$ to $180^\circ$ range are shown in Figs. 2.1 and 2.2. The parameters used were the following: the index of refraction of the coat $n_c$ was real (nonabsorbing coat) and equal to 1.5, the index of refraction of the water $n_w$ was also real and equal to 4/3. The thickness of the coatings $h$ were 0.26 $\mu m$ and 1.14 $\mu m$ respectively. The values of the coating were determined by taking the ratio of the inner sphere radius $a$ and the outer sphere $b$ (described by expression AOB in the COATSPHR program in Appendix A). The coating thickness $h$ was then determined by the relation $h = b - a = b(1 - \text{AOB})$. The range of bubble sizes can be illustrated by considering HeNe laser illumination with a wavelength in air $\lambda_{\text{air}}$ equal to 632.8 nm. The wavelength in water is then given by $\lambda_w = \lambda_{\text{air}}/n_w = 474.6 \text{ nm}$. Using $\lambda_w$ to determine $a$ for $k_a = 100$ gives $a = 7.5 \mu m$. A list of bubble sizes in terms of $h$ and the actual AOB values used in the calculations for $\lambda_w = 474.6 \text{ nm}$ are given in Table 2.1. Some concern may be raised about using the approximate value of $n_w = 4/3$; however, computational testing over a range of $n_w$ from to 1.332 (water at 30°C for $\lambda_{\text{air}} = 589.2 \text{ nm}$) to 1.338 (high salinity) showed the 4/3 value to be a very good approximation. Representative results of this evaluation are given in Appendix C. Normalization in the figures was chosen so that $I_f(\theta) = 1$ represents perfect reflection from an uncoated sphere of radius $a$. 
Table 2.1

Coating Thickness Values for $\lambda_w = 474.6$ nm ($\lambda_{air} = 632.8$ nm)

This table may be used to find the actual value of AOB used in the computations except in certain cases where the value of AOB is noted in the figure caption.

<table>
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<th>ka</th>
<th>radius = a(µm)</th>
<th>h(µm)</th>
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<th>radius = b(µm)</th>
<th>AOB</th>
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Figure 2.1. Calculated normalized scattering irradiances for $ka = 100$, $n_w = 4/3$, $n_c = 1.5$, $	heta = 0^\circ$ to $180^\circ$ for the log of the perpendicular polarization $I_1$. The solid curve is from Mie theory. The thin-dashed curve is the calculated coated sphere result for (a) $h = 0.26 \mu m$, (b) $h = 1.14 \mu m$. All values of $h$ are for HeNe laser light in water with $\lambda_{air} = 632.8$ nm. The inner radius $a = 7.5 \mu m$. 
Fig. 2.1 (a) and (b)
Figure 2.2 Like Figure 2.1 but for the parallel polarization case, $I_2$. 
Fig. 2.2 (a) and (b)
according to geometric optics. At a distance \( R >> ka^2 \) from the center of the bubble, the actual \( j \)-polarization irradiance is incident \( j \)-polarized irradiance multiplied by 
\[ I_j[(a/R)^2/4]\] where \( I_j = 4\pi S_j^2 (ka)^{-2} \) with \( j \) again denoting the previously mentioned polarization cases.

Analysis of Mie results (indicated by solid curve in Figs. 2.1 and 2.2) has shown two distinct regions of interest for detecting and sizing of noncoated bubbles in water. The first of these regions known as the Brewster scattering angle region, is the region around the Brewster angle determined by the condition \( \theta_B = \pi - 2\tan^{-1}(m_1) \) \( \theta_B \) = 106.3° for a noncoated air bubble in water) where \( m_1 = n_\text{air}/n_\text{w} \). The second region is that near the critical angle given by \( \theta_c = \pi - 2\sin^{-1}(m_1) \) \( \theta_c \) = 82.8° for noncoated air bubbles in water). The critical angle region irradiance is generally used for size determination while the ratio of the two regions may be used to determine if the scattered irradiance is indeed from a bubble. Furthermore, data and inspection of Mie results for uncoated bubbles, suggests that the normalized irradiance for the \( j = 2 \) case is closer to the universal value of \( I_j = 1/4 \) predicted at \( \theta_c \) by the physical-optics approximation. For this reason and the fact that parallel polarization has a simpler fine structure the \( j = 2 \) case is preferred for sizing and detecting microbubbles in water. Therefore in further illustrations we shall use the \( j = 2 \) case while the supplementary \( j = 1 \) cases are given in Appendix B.

Figures 2.1 and 2.2 show that even with the coating, the coarse and fine structure oscillations described by Langley and Marston still exist. However, with increasing coating thickness some structural changes become apparent. An example of these changes in the Brewster angle region is shown in Figs. 2.3(a)-(c) and 2.4(a)-(c) (thin-dashed curve). Through inspection of Figs. 2.3 and 2.4 one finds, in general, that the effect of the coating is to raise the baseline irradiance near \( \theta_B \) and to increase the magnitude of the
Figure 2.3. Calculated normalized irradiances for $ka = 100$ in the Brewster angle scattering region for $I_2$. The solid curve is from Mie theory and the thin-dashed curve is the calculated coated sphere result. With $\lambda_{\text{air}} = 632.8 \text{ nm}$ the coating thickness values are (a) $h = 0.26 \mu\text{m}$, (b) $h = 0.53 \mu\text{m}$, and (c) $h = 1.14 \mu\text{m}$. 
$I_2$

scattering angle (deg)

Fig. 2.3 (a)
Figure 2.4. Like Figure 2.3 but for \( k_a = 1000 \) (a = 75.5 \( \mu m \)) and (a) \( h = 0.23 \mu m \).
(b) \( h = 0.53 \mu m \), (c) \( h = 0.99 \mu m \).
ka = 1000

scattering angle (deg)

Fig. 2.4 (a)
$ka = 1000$

scattering angle (deg)

Fig. 2.4 (b)
$ka = 1000$

scattering angle (deg)

Fig. 2.4 (c)
fine structure oscillations (especially prominent in Fig. 2.4(c)). This behavior may affect the use of the Brewster angle region as a method of discriminating bubbles from particles in water since the ratio of the irradiances between it and the critical region vary non-uniquely for different coating values. The critical angle region also shows this behavior but due to the fact that irradiance values are generally several orders of magnitude higher the effects are of greater interest for the sizing of coated bubbles and hence further analysis shall be concentrated on this region.

It has been demonstrated by Marston and Langley\textsuperscript{14} that noncoated bubbles exhibit coarse irradiance oscillations as the scattering angle $\theta$ decreases below a critical value for total reflection $\theta_c = 82.82^\circ$ in water. With the introduction of a coating under the condition $n_c > n_w$, we find that this coarse structure begins to shift towards this noncoated critical angle with the degree of the angle shift dependent on the coating thickness $h$ and the coating index of refraction $n_c$. In addition to this coarse structure angle shift the magnitude of both $I_1$ and $I_2$ can generally increase. Examples of this angular shift and magnitude amplification for $k\alpha = 100$ with thickness values ranging from 0.25 $\mu$m to 3.0 $\mu$m are shown in Fig. 2.5(a)-(e). (Note that in these figures coarse structure comparison is also made with the physical-optics approximation.) The fact that the shifting of coarse structure is small near $\theta_c$ is noteworthy since noncoated results may be used to estimate the magnitude of the scattering from coated bubbles in many cases of interest. Note also that the physical-optics approximation does very well for coated bubbles in the critical angle region for thickness values less than 1 $\mu$m. The next structural change examined is the change in magnitude and period of the superposed fine structure.

Figure 2.6 illustrates the evolution of the fine structure in $I_2$ from $30^\circ$ to $90^\circ$ as a function of coating thickness. In this figure the effects of the coating can be seen as early as $h = 0.08 \mu$m (Fig. 2.6(a)). As the thickness increases, both the first and second peaks
Figure 2.5. Calculated normalized irradiances for \( ka = 100 \) in the critical angle scattering region for \( I_2 \). The solid curve is from Mie theory, the thin-dashed curve is the calculated coated sphere result, and the thick-dashed curve is from the Marston-Kingsbury physical-optics approximation. The coating thickness values for \( \lambda_{\text{air}} = 632.8 \text{ nm} \) are

(a) \( h = 0.25 \mu\text{m} \), (b) \( h = 0.5 \mu\text{m} \), (c) \( h = 1.0 \mu\text{m} \), (d) \( h = 2.0 \mu\text{m} \), and (e) \( h = 3.0 \mu\text{m} \).
Fig. 2.5 (b)

ka = 100

scattering angle (deg)

l_2
$ka = 100$

![Graph showing scattering angle (deg) for $ka = 100$](image)

**Fig. 2.5 (c)**
Figure 2.6. Evolution of the fine structure as a function of coating thickness for parallel polarized irradiance for $k_a = 100$. Coating thickness values for $\lambda_{\text{air}} = 632.8$ nm are

(a) 0 $\mu$m, (b) 0.08 $\mu$m, (c) 0.15 $\mu$m, (d) 0.19 $\mu$m, (e) 0.23 $\mu$m, (f) 0.48 $\mu$m,

(g) 0.66 $\mu$m, (h) 0.93 $\mu$m, and (i) 1.13 $\mu$m. The corresponding values of AOB specified in the computation were: (a) 1.0, (b) 0.990, (c) 0.980, (d) 0.975, (e) 0.970, (f) 0.940,

(g) 0.920, (h) 0.890, (i) 0.870.
from the left begin to decay into finer structure while the large number of smaller peaks increase in amplitude and move closer together. At \( h = 0.23 \, \mu \text{m} \) (Fig. 2.6(e)) the fine structure becomes quite prominent. An interesting effect was also noted certain values like \( h = 0.66 \, \mu \text{m} \) (Fig. 2.6(g)). At these particular values the fine structure amplitude is diminished as is shown. Possible reasons for this will be discussed in the next section.

B. Geometric Modeling

Interpretation of the results found in Section A may be done using Fig. 2.7. Adopting the notation of Langley we define rays using the notation \((p,l)\), where \( p \) is the number of chords within the bubble and \( l \) equals the number of crossings of the optic axis. Note that a subscript \( b \) will be used to indicate rays that pertain to the outer sphere only. Examining the two rays used in the physical-optics approximation, namely the \((0,0)\) and \((1,0)\) rays, and the \((2,1)\) ray the figure shows that the coating shifts the scattered ray towards higher values of \( \theta \) as well as increasing the distance between the \((0,0)\) and \((2,1)\) ray. The first mentioned effect partially accounts for the observed coarse structure angle shift while the second partially accounts for the change in the fine structure.

It has been shown that interference between the \((0,0)\) and \((2,1)\) rays is partially responsible for the fine structure oscillations. The quasi-period \( \Delta \theta_{fs} \) of this structure near the critical angle has been calculated in a first term approximation to be roughly equal to \( \lambda_w/B_2 \), where \( B_2 \) is the distance between the \((0,0)\) and \((2,1)\) ray. Thus, by increasing \( B_2 \) with the addition of the coating, the fine structure will have a smaller period which is clearly the case. The damped fine structure seen in Fig. 2.6(g) is most likely due to effect of the coating on reflection and transmission coefficients. The purpose of the geometric modeling is to give the reader some physical insight in the interpretation of the computational results presented. It is by no means meant to be a complete
Figure 2.7. Coated sphere geometric model. Rays are specified by \((p,l)\) where \(p\) = the number of chords within the bubble, and \(l\) = the number of crossings of the optic axis. Dashed lines indicate the uncoated sphere ray paths while solid lines represent coated sphere ray paths. Note that bending of the rays is exaggerated for effect. The subscript \(b\) indicates rays which pertain to the outer sphere only.
Fig. 2.7.--Coated sphere geometric model.
description of the rather complex phenomena of scattering of radiation by coated bubbles. Note also that the bending of rays is exaggerated for illustration of the effect of the coating.

C. Coating Effects on Larger Bubbles

The calculated \( j = 2 \) critical angle region results for a range of coating thicknesses for \( k_\alpha \) equal to 500, 1000, and 2500 (\( a = 37.8 \, \mu m \) to 189 \( \mu m \) for \( \lambda_w = 474.6 \, nm \)) respectively are shown in Figs. 2.8 to 2.10. The parameters used were similar to those used for Fig. 2.5. The effects of the coating on increasing bubble size indicate that as \( k_\alpha \) becomes larger with \( h \) fixed the angular phase shift in the coarse structure is diminished. Marston has calculated this shift in the thin film limit for the critical angle region to be proportional to \( h/a \) which agrees with the shown results. Another effect noted with increasing bubble size is that the changes in the fine-structure are less drastic for a given value of \( h \) as bubble size increases. As an example, comparison between Figs. 2.5(e) and 2.9(e) (the 3 \( \mu m \) thickness cases for \( k_\alpha = 100 \) and \( k_\alpha = 1000 \)) shows that in the smaller bubble case a 3 \( \mu m \) thickness (thin-dashed line) has drastically changed the intensity structure from the original noncoated case (solid line) while for the large bubble results, a lesser shift in the coarse oscillations and a slight increase in fine-structure amplitude has occurred. The original noncoated structure, however, has remained.

Another feature overlayed on these graphs is the physical-optics approximation. The approximation does well predicting the coarse structure for larger values of \( k_\alpha \) for thin coatings (less than 0.5 \( \mu m \)) over most of the graphed regions and is capable of accounting for coarse structure at most coating thickness values in the critical angle region (approximately 80° to 85°) due to the stationary behavior of the angular shift near \( \theta_c \).
Figure 2.8. Calculated normalized irradiances for \( ka = 500 \) (\( a = 37.8 \, \mu m \)) in the critical scattering region for \( I_2 \). The solid curve is from Mie theory, the thin-dashed curve is the calculated coated sphere result, and the thick-dashed curve is from the physical-optics approximation. The coating thickness values for \( \lambda_{air} = 632.8 \, nm \) are (a) \( h = 0.25 \, \mu m \), (b) \( h = 0.5 \, \mu m \), (c) \( 1.0 \, \mu m \), (d) \( h = 2.0 \, \mu m \), and (e) \( h = 3.0 \, \mu m \).
\[ \text{ka} = 500 \]

**Fig. 2.8 (a)**

- **Scattering Angle (deg)**

The graph shows a plot of scattering angle (degree) against various intensity levels. The figure illustrates the scattering behavior at an angular range of 60 to 90 degrees, with notable peaks and troughs indicating variations in scattering intensity.
Fig. 2.8 (b)
Figure 2.9. Like Figure 2.8 but for $ka = 1000$ with (a) $h = 0.25 \mu m$, (b) $h = 0.53 \mu m$, (c) $h = 0.99 \mu m$, (d) $h = 2.01 \mu m$, and (e) $h = 3.00 \mu m$. 
$ka = 1000$

![Graph](image.png)

Figure 2.9 (c)
Figure 2.10. Thickness values approximately equal to those in Figure 2.8 but for \( ka = 2500 \). See Table 2.1.
\[ \text{ka} = 2500 \]

Fig. 2.10 (a)

**scattering angle (deg)**
\[ ka = 2500 \]

Fig. 2.10 (e)

Scattering angle (deg)
D. Absorbing Coatings

Results for absorbing coatings are shown in Figs. 2.11 - 2.14. The range of absorbing coatings tested was $n_c = 1.5 + i1E-9$ (weakly absorbing) to $n_c = 1.5 + i1E-2$ (very strong absorption). The results in Figs. 2.11 - 2.13 indicate that the effects of absorbing coatings are negligible until the strong absorbing cases are reached ($n_c = 1.5 + i1E-3$). In Fig. 2.14 a typical result for the effects of increasing absorbing coating thickness is shown. As can be seen even with a 3.0 μm coat the effects are still small. This was even more apparent for the perpendicular case which is given in Appendix B. One notable effect was the decrease of the irradiance amplitude, which was greater at smaller values of $\theta$ and approached non-absorbing results near the critical angle. It should also be pointed out that the coarse structure angular shift is present in the absorbing coating cases also. Severe angle shift changes in the coarse structure amplitude and structure were observed in all cases for $n_c = 1.5 + i1E-2$ (Fig. 2.13).

In analyzing the absorbing coating results it was found that the effects of absorption were minimal. Since most liquids have very low absorption values$^{15}$ and coating thickness values are generally presumed to be small it is doubtful whether absorption will have a major effect on actual coated-bubble measurements. Thus, modeling could be done using the non-absorbing cases. This would speed up computation time considerably as the imaginary components would not be involved in the calculations. Another point of interest is that coatings would have to be extremely thick (> 3 μm), especially for larger bubbles, in order to have any major effects even for strongly absorbing cases. Finally, the absorption value for water is in the $\text{Im}(n_w) = 1E-9$ region and the effects of such a small value of $\text{Im}(n_w)$ on the scattering pattern are expected to be small. Hence $n_w$ is always taken to be real.
Figure 2.11. Calculated normalized irradiance for $ka = 500$ and $n_c = 1.5 + i1E-3$. The solid curve is the non-absorbing case at $h = 1.01 \, \mu m$, the thick-dashed curve is the physical-optics approximation, and the thin-dashed curve is the calculated absorbing coating result for $h = 1.01 \, \mu m$. 
Figure 2.12. Like Figure 2.11 but the thin-dashed curve represents $n_c = 1.5 + i5E-3$

at $h = 1.01 \mu m$. 
$ka = 500$

**scattering angle (deg)**

Fig. 2.12
Figure 2.13. Like Figure 2.11 but the thin-dashed curve represents \( n_c = 1.5 + i1E-2 \) at \( h = 1.01 \text{ \mu m} \).
Figure 2.14. Calculated normalized irradiances for $k_a = 1000$ and $n_c = 1.5 + i1E-3$ for
(a) $h = 0.99 \mu m$, (b) $h = 2.01 \mu m$, (c) $h = 3.14 \mu m$, and (d) $h = 5.00 \mu m$. The solid
curves are non-absorbing results at the given values of $h$ and the thin-dashed curve is the
calculated absorbing coating results.
$ka = 1000$

![Graph showing scattering angle (deg) for $ka = 1000$](image)

Fig. 2.14 (a)
Fig. 2.14 (b)
E. Effect of Varying \( n_c \) Values

The final parameter investigated was the effect of varying real values \( n_c \). Figures 2.15 - 2.18 show the differences between the \( n_c = 1.5 \) case generally used and those of \( n_c \) equal to 1.45 and 1.55, respectively. In general, the effect of coatings with indices of refraction less than 1.5 was to diminish the changes in structure until the value \( n_c = n_w \) in which case the shell was water and gave Mie results. For \( n_c < n_w \) an angular shift of the coarse structure in the opposite direction towards lower values of \( \theta \) occurred. In the cases where \( n_c > 1.5 \) the effect was one of accelerating both the shift and breakdown in structure. The difference between values \( n_c = 1.45, 1.5, \) and 1.55 was very slight for thin coatings at all size parameters; i.e., Figs. 2.15 and 2.16 and even less noticeable at larger values of \( ka \) for thicker coatings (Figs. 2.17 and 2.18).

In general most surfactants found in sea water will most likely have indices of refraction in the 1.45 to 1.55 range. Based on the results in this section, the use of the value \( n_w = 1.5 \) will yield representative results. Although there are some small changes in the amplitude of the fine structure, the amount of coarse structure angular shift in this region is negligible for thinly coated bubbles at all sizes and thickly coated bubbles at large values of \( ka \). Using the results characterized here it would be difficult to determine from near-critical angle scattering data alone if the coating had a 1.45, 1.55, or 1.5 index of refraction.
Figure 2.15. The effect of changing $n_c$ with $ka = 100$ and $h = 0.26 \mu m$. The thick-dashed curve represents $n_c = 1.45$, the thin-dashed curve is for $n_c = 1.5$, and the solid curve represents the noncoated case.
Figure 2.16. Like Figure 2.15 but the thick-dashed curve represents $n_c = 1.55$. 
Figure 2.17. The effect of changing $n_c$ with $ka = 1000$ and $h = 3.14 \, \mu m$. The short-dashed curve represents $n_c = 1.5$, the long dashed curve represents $n_c = 1.45$, and the solid curve is for the noncoated case.
Figure 2.18. Like Figure 2.17 but for $n_c = 1.55$ (long-dashed curve).
REFERENCES TO CHAPTER 2


3. C. Bohren and D. Huffman, Absorption and Scattering of Light by Small Particles (Wiley, N.Y., 1983) Appendix B.


5. See Ref. 4.


8. See Ref. 3.


11. See Ref. 4.


15. See Ref. 3, p. 279.

CHAPTER 3

DISCUSSION

3.1. Interpretation and Analysis

Computational results indicate that for both perpendicular and parallel polarizations, $I_1$ and $I_2$ respectively, the irradiance patterns of the scattered light for coated bubbles in water are noticeably altered from noncoated results. While both the coarse and fine structure oscillations previously evident in noncoated Mie results remain, a prominent angular shift toward $\theta_c$ occurs in the coarse structure oscillations due to the coating. The degree of this shift is dependent upon three factors: the size of the bubble $a$, the index of refraction of the coating $n_c$ and finally the coating thickness $h$. For large bubbles (i.e., Fig. 2.10) the amount of shift is much less than for the smaller sized bubble (i.e., Fig. 2.5). The shift has been calculated by Professor Marston in the thin coating limit to be directly proportional to $h/a$.

Other effects induced by the coating are changes in the period and amplitude of the fine structure oscillations. As an illustration of this effect consider the fine structure oscillations evident in Fig. 2.6 at fixed scattering angle, say 60 degrees. In Fig. 2.6(b) ($h = 0.08 \mu m$) the angle between the two fine structure maxima peaks is roughly 2.6° while in Fig. 2.6(i) ($h = 1.12 \mu m$) the distance is roughly 1.7°. Thus, in general, this indicates that the fine structure quasi-period $\Delta \theta_f$ tends to decrease with increasing $h$. The plausible reason for this decrease is that the transverse distance between the far side, or (2,1) ray, and the surface reflected ray (0,0) appears to increase with $h$. Inspection of Fig. 2.6 also indicates that except for 2.6(g) the amplitude of the fine structure oscillations
COMPUTATIONAL ANALYSIS OF THE EFFECTS OF SURFACE FILMS ON THE SCATTERING... (U)
WASHINGTON STATE UNIV PULLMAN
DEPT OF PHYSICS S C BILLETTE ET AL. 01 OCT 86

UNCLASSIFIED N89614-86-K-0242
MICROCOPY RESOLUTION TEST CHART
NATIONAL BUREAU OF STANDARDS-1963-A
tends to increase with increasing \( h \). It is possible that this could be accounted for by a shift in the effective reflection or transmission coefficients for the (2,1) ray shown in Fig. 2.7. The observations made here were shown to be valid for both absorbing and non-absorbing coatings.

The computation results for the case of coatings with absorption may be understood as follows: analyzing Figs. 2.11 to 2.14 one finds that it is possible on physical grounds to anticipate that the effects of absorption are governed by the magnitude of a unitless parameter \( \sigma = 4\pi h \text{Im}(n_c)/\lambda_{\text{air}} \). The irradiance of a wave propagating through an absorbing medium a distance \( h \) is related to \( \sigma \) by the relation \( I = I_0 e^{-\sigma} \) where \( I_0 \) is the initial irradiance. This is explained in Section 2.8 of Ref. 1. Let us now illustrate the use of this parameter \( \sigma \) by looking at an actual case presented in the data. Analyzing Fig. 2.12 we find that \( h = 1.0 \mu m \) and \( \text{Im}(n_c) = 5E-3 \) giving a value of \( \sigma = 9.93E-2 \). Calculating \( \sigma \) for Fig. 2.14(d) with \( h = 5.0 \mu m \) we obtain the same value of \( \sigma = 9.93E-2 \). Note that the effect on the irradiance curves is similar. Finally, it should be noted that for higher \( \sigma \) values the effects due to absorption become more drastic.

Comparing Fig. 2.13 (\( \sigma = 1.99E-1 \)) with Figs. 2.11 and 2.14(a) (\( \sigma = 1.99E-2 \)) we find this is indeed the case. Thus for coatings with \( \sigma \) less than approximately 0.02 the effects of absorption are negligible and real values for \( n_c \) would best be used in computations since the use of real values reduces the computation time significantly.

Based on the results presented here and in Chapter 2, the following conclusions are noted for the case of \( j = 2 \) scattering from bubbles having nonabsorbing coatings. First, for a coating less than 1 \( \mu m \) in thickness on bubbles having \( ka \geq 500 \) there is very little effect of the coating on the principal features of the scattering pattern. As noted in Appendix B, the effect of a given coating on the coarse features of the scattering pattern is usually smaller for \( j = 2 \) than for \( j = 1 \). This may be because the reflection of light from the coating-water interface is smaller for \( j = 2 \) than \( j = 1 \) in the angular region of
interest. (See Appendix B.) For bubbles with \( ka = 100 \), changes in the fine structure can be pronounced for coating thickness values between 0.25 \( \mu m \) and 1.0 \( \mu m \).

### 3.2. Experimental Applications

If microbubbles in the ocean are proven to be coated by a thin film of surfactant, then it becomes important to determine if such parameters as thickness and complex refractive index of the coatings affect the reliability where by light scattering techniques can be used to determine the radius \( a \) of a gas bubble. Calculations given here for both coated and noncoated bubbles indicate that for their sizing and detection the coarse structure oscillations for the \( j = 2 \) case in the critical angle scattering region are best used for sizing. The basis for this observation is three-fold; first, the fine structure is generally much smaller in magnitude for both coated and noncoated bubbles for \( j = 2 \) than for \( j = 1 \). Secondly, the \( j = 2 \) case for coated values does not have the change in coarse structure (other than the angular shift which was noted for both cases) as was observed for \( j = 1 \); see e.g., Fig. B.3(e). Thirdly, unless coatings are found to be quite thick or highly absorbing previous observations and conclusions made using Mie theory and the physical-optics approximation could be used as an aid in the general design of apparatus. Thus, it is recommended that sizing and detecting of coated bubbles in experimental applications is best done by making use of the \( j = 2 \) coarse-structure and placing the maximum number of detectors as close as possible to the critical angle scattering region.
REFERENCES TO CHAPTER 3

APPENDIX A

COMPUTER PROGRAMS

Two programs were used to generate our plots. COATSPHR, using modified versions of Wiscombe's Mievo, and Bohren and Huffman's coated sphere subroutines produced the coated sphere results and ESCAT the approximation results. The data was stored in system files and manipulated using a Statistical Analysis System (SAS) routine which generated the plots presented. All plots were done on an HP7475a plotter using 0.5 mm and 0.7 mm Hewlett Packard drafting plotter pens. All programs are written in IBM FORTRANV.

COATSPHR

To use COATSPHR the following values must be input in the main calling program:

1. DT is the angle increment to be sampled. Typically it must be less than ($\lambda/7a$) rad where $a$ is the inner sphere radius.

2. ANG(N) is the initial angle at which scattering calculations begin. All calculations are symmetric about 90° meaning that if ANG(N) = 30° the range of 30° to 150° will be calculated.

3. NANG is the number of angles for which the coefficients are calculated. Typically for the entire range it is determined by NANG = 1/DT x (90° - ANG(N)) + 1. Note that for this particular program version NANG must be less than 1800. (This number may be increased by increasing all array sizes but for the cases presented 1800 was adequate.)
desired then the variable NANG should be determined by \( NANG = \frac{1}{DT} \times (\text{last angle}) + 1 \) (for example, to compute 0° to 5° and 175° to 180° with \( DT = 1/20 \) use \( NANG = 20 \times 5 + 1 = 101 \); \( \text{ANG}(N) = 0° \).

In addition to this the following values must be input in subroutine BHCP(X,Y).

1. \( \text{REFMED} \) is the refractive index of the surrounding medium.
2. \( \text{REFREI} \) is the real part of the index of refraction of the inner sphere medium.
3. \( \text{REFIM} \) is the imaginary component of the index of refraction of the inner sphere medium.
4. \( \text{REFRE2} \) is the real part of index of refraction of the coating.
5. \( \text{REFIM2} \) is the imaginary component of the coating index of refraction.
6. \( AOB \) is the ratio of the inner radius \( a \) divided by the outer radius \( b \) (see Fig. 1.1). The coating value is given by the relation \( h = b(1 - AOB) \).
7. \( X \) is the inner sphere size parameter \( ka = 2\pi a/\lambda_o \) where \( \lambda_o \) is the wavelength in the outer medium.

Once these values are input COATSPHR generates the following output:

1. \( Y \) is size parameter of the outer shell of radius \( b \).
2. \( I_1(\theta) \) is the intensity for the electric field perpendicular to the scattering plane.
   It is given for each value of \( \theta \) input. It is normalized such that \( I_1(\theta) = 1 \)
   represents perfect reflection according to geometric optics.
3. \( I_2(\theta) \) is the intensity for the parallel case.
4. \( S_1(\theta) \) and \( S_2(\theta) \) are the partial wave angular scattering functions.
5. The average intensity.
6. Efficiency factors:
   a) Extinction coefficient \( Q_{ext} = C_{ext}/G \) where \( C_{ext} \) is the extinction cross section and \( G \) is defined below.
b) Scattering coefficient \( Q_{\text{sca}} = \frac{C_{\text{sca}}}{G} \) where \( C_{\text{sca}} \) = scattering cross section and \( G \) is the particle cross sectional area projected onto a plane perpendicular to the incident beam (e.g., \( G = \pi b^2 \) for coated sphere).

c) Absorption coefficient \( Q_{\text{abs}} \) which is simply \( Q_{\text{ext}} - Q_{\text{sca}} \).

7. The asymmetry factor is the average cosine of the scattering angle. For a particle that scatters light isotropically the asymmetry factor would be zero.

Being that the author is a novice in FORTRAN programming no claim is made as to the efficiency or lack thereof in COATSPHR. The program requires close to 1 Megabyte of virtual memory to run as there are 3 large arrays and the use of quadruple precision to contend with. Fortunately this is well within the IBM 3090-200's capability range and so no attempt was made to economize the program.

**ESCAT**

Values for the physical-optics approximation were generated using ESCAT FORTRAN. The program is virtually unchanged from the version used by Langley in 1982. Only a few modifications were necessary to convert it for use on the FORTVS compiler. To use ESCAT the following values must be entered:

1. \( M \) is the relative refractive index of the non-coated bubble.
2. \( KA \) is size parameter of the noncoated bubble.
3. \( \text{PHIBGN} \) is the value of the first scattering angle (in degrees) to be evaluated.
4. \( \text{PHIEND} \) is the last scattering angle (in degrees).
5. \( DPHI \) is the angle step size (analagous to \( DT \) in COATSPHR).

The output gives the irradiances \( I_j \) for both the \( j = 1 \) and \( j = 2 \) cases, with or without diffraction effects. The program only determines the coarse structure as has been shown in the figures but has the advantage that it executes extremely quickly for all bubble sizes, whereas similar Mie calculations require significantly more computation time.
FILE: COATSPHR FORTRAN AL

SCATTERING PROGRAM FOR COATED SPHERES
S. C. BILLETTE  NOVEMBER 1985

ANGULAR DIMENSIONS

COMPLEX=8 SBACF, S1, S2
REAL=16 ANG(3600), XMU, PI, CON, DT, XX, CONV, YY
REAL=16 I1, I2, INTEN, ANGLE, N2CUT
COMMON/IMOUT/S1(3600), S2(3600), XMU(3600), SBACF, XX, YY.

USE SUBROUTINE BHCP TO INITIALIZE FOR AN AND BM CALCULATIONS
AND OBTAIN CORE AND COAT SIZE PARAMETERS

CALL BHCP(XX,YY)

N2CUT = 0.000
PI = 3.14159265358979323846264338327950
CON = PI/180.00
NANG = 301
CONV = XX**2
DT = 1.000/20.000
N = 1
ANG(N) = 75.000

DO 1 J = 1, NANG
XMU(N) = QCMS(CON*ANG(N))
ANG(N+1) = ANG(N)+DT
N = N+1
NUMANG = 2*(N-1)
J = NUMANG+1

2 DO 1 I = N, NUMANG
ANG(1) = 180.00-ANG(J-I)
1 XMU(I) = -XMU(J-I)

CALL SUBROUTINE MIEVO TO DETERMINE S1 AND S2

CALL MIEVO

CONTINUE

QAMS = GEXT-GSCA
WRITE(6,1000) XX, YY

DO 10 I = 1, NUMANG
I1 = 4.0000*REAL(S1(I))**2-(AIMAG(S1(I))**2)/CONV
I2 = 4.0000*REAL(S2(I))**2-(AIMAG(S2(I))**2)/CONV
INTEN = .500*(I1-I2)
ANGLE = ANG(I)

WRITE STATEMENTS

10 WRITE(6,1001) ANGLE, I1, I2, S1(I), S2(I), INTEN
FILE: COATSPHER FORTRAN A1

WRITE(6,1002) QEXT, QSCA, QABS, QGSC
WRITE(6,1003) SBACX
STOP

FORMAT STATEMENTS
1000 FORMAT(5X,'CORE PARAMETER =',1D14.6,1X,'COAT PARAMETER =',1D16.6,
* '/U ANGLE',8X,'IL',11X,'IZ',20X,'SL',26X,
* 'SZ',15X,'INTENSITY')
1001 FORMAT(F8.3,1E14.6)
1002 FORMAT(130X,'EXTINCTION SCATTERING ABSORPTION'/7X,
* 'EFFICIENCY FACTORS',3F14.6/7X,'ASYMmetry FACTOR =',F9.6)
1003 FORMAT(7X,'S BACKSCATTER =',2E14.6)

END

SUBROUTINE MIEVO

************
NOTE: THIS SUBROUTINE USES THE SAME VARIABLE NAMES AS THOSE FOUND IN WISCOMBER'S MIEVO FROM WHICH IT IS DERIVED. SEE MIEVO FOR A COMPLETE LISTING.
************

COMMON/INOUT/S1(1600),S2(1600),XNU(1600),SBACX,XX,YY,
* QEXT, QSCA, QGSC, NUMANG, N2CUT/BLOCK/NAN, SN/BLOCK/NT
REAL*16 NP1N, N2CUT
REAL*16 XX,YY,XINV,FN,FNP1,BN,BNP1,THNP1,COEFF
REAL*16 PIN,FINNL,THP,TAUN,SNX
LOGICAL NOMAG, NOANGS
COMPLEX*8 SBACX, SL, S2
COMPLEX*32 SP,SM,SPS,SN
COMPLEX*32 ANFH,SNFH,ANP,BNP,ANML,BNML
COMPLEX*32 AN, BN
DIMENSION SP(1800),SN(1800),SPS(1800),SNX(1800),
* PIN(1800),FINNL(1800),TAUN(1800),THP(1800)

XINV = 1.00/XX
NN2 = NUMANG+1
NN = NN2/2
NOANGS = NUMANG.EQ.0

CALCULATE NUMBER OF TERMS IN PARTIAL WAVE SERIES
USING EMPIRICAL FORMULAE WHICH WERE FITTED FOR SIZE PARAMETERS UP TO 20,000
IF(YY.LE.8.000) NT = YY+4.0000+YY*(1.0000/1.0000)-1.000
IF(YY.GT.8.000) NT = YY+4.0000+YY*(1.0000/1.0000)-2.000
NP1 = NT+1
NP2 = NT+2

WRITE(6,8001) NT
8001 FORMAT(4X,' NT =',16/4X)

INITIALIZE QUANTITIES USED FOR EFFICIENT CALCULATION OF NUMERICAL COEFFICIENTS IN PARTIAL WAVE SERIES
FILE: COATS.FORTRAN  AL

FM = 1.000
RM = 1.000
MN = 1.
AMML = (0.000,0.000)
BMML = (0.000,0.000)

INITIALIZE SUMS FOR EFFICIENCIES, ASYMMETRY FACTOR, AND BACKSCATTERING AMPLITUDE

QEXT = 0.000
QSCA = 0.000
GQSC = 0.000
SBACK = (0.000,0.000)

INITIALIZE ANGULAR FCN PIN AND SUMS FOR S+, S- AT ALL ANGLES

DO 250 J = 1, NM
   S(J) = (0.000,0.000)
   SM(J) = (0.000,0.000)
   SPS(J) = (0.000,0.000)
   SMN(J) = (0.000,0.000)
   NJSML(J) = 0.000
250 CONTINUE

DO 500 N = 1, NT

COMPUTE THE VARIOUS NUMERICAL COEFFICIENTS NEEDED

FNPL = FM-1.000
TNWNP1 = FN+FNPL
RNPL = 1.000/RNPL
CQEFF = RM+RNPL
NPDLN = 1.000-RM

CALCULATE THE PARTIAL WAVE SERIES COEFFICIENTS AN AND BN

CALL BCOAI

INCREMENT SUMS FOR ASYMMETRY FACTOR, EXTINCTION EFFICIENCY, AND BACKSCATTERING AMPLITUDE

IF(NOANGS) GO TO 450

PUT SERIES COEFFICIENTS IN FORM NEEDED FOR COMPUTING S+, S-

GQSC = GQSC - (FM-RM)*REAL(AMML+QCONJG(AN)-BMML+QCONJG(BN))
   - - COEFF*REAL(AN+QCONJG(BM))
QSCA = QSCA - TWONP1*REAL(AN)**2 + (QIMAG(AN)**2
   - REAL(BM)**2 - (QIMAG(BN)**2
QEXT = QEXT - TWONP1*REAL(AN-BM)
SBACK = SBACK - (MN+TWONP1)*AN-BM

ANP = COEFF*(AN-BM)
BNP = COEFF*(AN-BM)
FILE: COATSPH FORTRAN AL

ANPH = MN*ANP
BNPH = MN*BNP

DO 400 J = 1, NN

ADD UP SUMS WHILE UPWARD RECURRING ANGULAR FUNCTIONS
LITTLE PI AND LITTLE TAU

TMP(J) = (XMU(J)*PIN(J)) - PINML(J)
TAIN(J) = FH*TMP(J) - PINML(J)
SP(J) = SP(J) - ANPH*(PIN(J) - TAIN(J))
SN(J) = SN(J) - BNPH*(PIN(J) - TAIN(J))
SPS(J) = SPS(J) - ANPH*(PIN(J) - TAIN(J))
PINML(J) = PIN(J)
PIN(J) = (XMU(J)*PIN(J)) - BNPH*TMP(J)

400 CONTINUE

UPDATE RELEVANT QUANTITIES FOR NEXT PASS THROUGH LOOP

450 NN = -MN
FN = FHNP
RN = RHNP
ANPH = AN
BNPH = BN

500 CONTINUE

MULTIPLY SUMS BY APPROPRIATE FACTORS TO GET QEXT, QSCA, QGSC

QEXT = 2.0Q0*(XINV*2)*QEXT
QSCA = 2.0Q0*(XINV*2)*QSCA
QGSC = 4.0Q0*(XINV*2)*QGSC
SBACK = 2.5Q0*SBACK

IF (NOIMGS) RETURN

DO 600 J = 1, NN
S1(J) = 0.5Q0*(SP(J) = SN(J))
S2(J) = 0.5Q0*(SP(J) - SN(J))
S1(NN2-J) = 0.5Q0*(SPS(J) - SHS(J))
S2(NN2-J) = 0.5Q0*(SPS(J) - SHS(J))

600 RETURN

END

SUBROUTINE BHCP(X,Y)

SUBROUTINE TO INPUT COATED SPHERE PARAMETERS AND TO
INITIALIZE FOR LATER ENTRY INTO BCARTI

REAL*16 X,Y,PI,AQB
REAL*16 QEXT,QSCA,QBACK
REAL*16 REFMED,REFREL,REFN1,REFN2,REFM2
COMPLEX*12 RFREL1, RFREL2

REFMED = REFRACTIVE INDEX OF SURROUNDING MEDIUM
FILE: COATSPHR FORTRAN AL

**REFRACTIVE INDEX OF CORE = REFRL - i*REFN1**
**REFRACTIVE INDEX OF COAT = REFRL2 - i*REFN2**
**AOB = RATIO OF CORE RADIUS TO COAT RADIUS**
**X = CORE SIZE PARAMETER**
**Y = COAT SIZE PARAMETER**

REFMED = 4.000/3.000
REFRL = 1.0000
REFN1 = 0.0000
REFRL2 = 1.3000
REFN2 = 0.0000
AOB = 0.9900
X = 100.000
Y = X/100

DEFINE VARIABLES RFRELL, RFREL2
RFRELL = QCMLPX(REFRL,REFN1)/REFMED
RFREL2 = QCMLPX(REFRL2,REFN2)/REFMED

USE SUBROUTINE HCOCAT TO CALC. EFFICIENCIES FOR COATED SPHERES
CALL HCOCAT (X,Y,RFRELL,RFREL2)

WRITE STATEMENTS
WRITE (6,9)
WRITE (6,11)
WRITE (6,9)
WRITE (6,4)
WRITE (6,12) REFMED,REFREL,REFN1,REFREL2,REFN2
WRITE (6,14) AOB

**FORMAT STATEMENTS**
4 FORMAT('')
9 FORMAT('9X,' 'CALCULE SPHERE SCATTERING PROGRAM')
11 FORMAT('9X,' 'CORE SIZE PARAMETER')
12 FORMAT('5X,' 'REFMED = ',E14.9/5X,'REFREL = ',E14.9,
 9X,' 'REFN1 = ',E14.9/5X,'REFREL2 = ',E14.9,3X,'REFN2 = ',E14.9)
14 FORMAT('5X,' 'AOB = ',E14.9)

END
SUBROUTINE HCOCAT (X,Y,RFRELL,RFREL2)

SUBROUTINE HCOCAT TO CALCULATE LITTLE AN AND BN FOR
GIVEN SIZE PARAMETERS OF CORE AND COAT AND RELATIVE
REFRACTIVE INDICES, BASED ON SUBROUTINE GIVEN IN AP-
END 0.0% OF ABSORPTION AND SCATTERING OF LIGHT BY C.
BOHREN AND D. HUFFMAN (WILEY, 1983)

************************** CAUTION *********************
HCOCAT SHOULD NOT BE USED FOR LARGE, HIGHLY ABSORBING
FILE: COATSFER FORTRAN AL

COATED SPHERES X*REFM1, X*REFM2 AND Y*REFM2 SHOULD BE LESS THAN ABOUT 10.

***************CAUTION***************

COMMON/BLOCK2/AN, BN/BLOCK1/YSTOP
REAL*16 QSCA, QBACK, XEXT, PS1V, CH1T, DEL, RN, YSTOP
REAL*16 X, Y, PS1V, PS1V, CH1T, CH1T, CH1T
COMPLEX*12 RFRELL, RFREL3, X1, X2, Y2, RFREL
COMPLEX*12 D1X1, D0X1, D1X2, D0X2, D1Y2, D0Y2
COMPLEX*12 DX1, DX2, AY2, CH1T2, CH1T2, CH1T2, CH1T2, CH1T2, CH1T2, CH1T2
COMPLEX*12 CHIPX2, CHIPY2, ANCAP, BNCAP, CNYBAR, CNYBAR, AN, BN, CRACK, CRACK
COMPLEX*12 DBIGD1, BIGD1, DBIGD2, BIGD2, BIGD2, BIGD2
COMPLEX*12 ZINV, ZINV2, ZINV3
DIMENSION BIGD1(10150), BIGD2(10150), BIGD2(10150)

DEL IS THE INNER SPHERE DIVERGENCE CRITERION

DEL = 1.0Q=0
X1 = RFRELL*X
X2 = RFRELL*X
Y2 = RFRELL*Y
YSTOP = Y - 4.0Q0*Y*((1.0Q0/3.0Q0) - 2.0Q0)
RFREL = RFREL2/RFRELL
NSTOP = YSTOP
NTP1 = NSTOP
NTP2 = NSTOP

SERIES TERMINATED AFTER NSTOP TERMS

SUBROUTINE TO CALCULATE INITIAL HIGH ORDER BIGD1-N
CALL BIGD(X, RFRELL, DBIGD1)

BIGD1(NSTOP) = DBIGD1
ZINV = 1.0Q0/(RFRELL*X)

DO 70 N = 2, NSTOP
DBIGD1(IDNP1-N) = (NTP1-N)*ZINV-1.0Q0/((NTP2-N)*ZINV-DBIGD1)
70 BIGD1(NTP1-N) = DBIGD1
SUBROUTINE TO CALCULATE INITIAL HIGH ORDER BIGD2-N
CALL BIGD(X, RFRELL, DBIGD2)

BIGD2(NSTOP) = DBIGD2
ZINV2 = 1.0Q0/(RFRELL*X)

DO 80 N = 2, NSTOP
DBIGD2(IDNP1-N) = (NTP1-N)*ZINV2-1.0Q0/((NTP2-N)*ZINV2-DBIGD2)
80 BIGD2(NTP1-N) = DBIGD2
SUBROUTINE TO CALCULATE INITIAL HIGH ORDER BIGD3-N
CALL BIGD(Y, RFRELL, DBIGD3)
FILE: COATSPHER FORTRAN AL

BIGD3(NSTOP) = DBIGD3
ZNVJ = 1.0QG/(RFRELI+Y)

DO 90 N = 1,NSTOP
    DBIGD3 = (NTP2-N)*ZNVJ-1.0QG/(((NTP2-N)*ZNVJ)-DBIGD3)
90 BIGD3(NTP1-M) = DBIGD3

DOX1 = CCQGOS(X1)/COSIN(X1)
DOX2 = CCQGOS(X2)/COSIN(X2)
DOY2 = CCQGOS(Y2)/COSIN(Y2)
PSIY = QGOS(Y)
PSIY = QSIN(Y)
CHIY = -QSIN(Y)
CHIY = QGOS(Y)

XXY = QCMPX(PSTY,-CHIY)
XXY = QCMPX(PSTY,-CHIY)
CHIY2 = -QSIN(Y2)
CHIY2 = QCOS(X2)
CHIY2 = -QSIN(X2)
CHIY2 = QCOS(X2)

QCCA = 0.0QG
QEXT = 0.0QG
XBACK = QCMPX (0.0QG,0.0QG)
N = 1
IFLAG = 0

RETURN
ENTRY BCOAT1

200 RN = N
PSY = (2.0QG*RN-1.0QG)*PSIY/Y - PSIY
CHY = (2.0QG*RN-1.0QG)*CHIY/Y - CHIY
XXY = QCMPX(PSTY,-CHY)
OLY2 = BIGD3(N)
IF (IFLAG.EQ.1) GO TO 999
OLX1 = BIGD1(N)
OLX2 = BIGD2(N)

CHIY2 = (2.0QG*RN - 1.0QG)* CHIY2/X2 -CHIY2
CHIY2 = (2.0QG*RN - 1.0QG)* CHIY2/Y2 -CHIY2
CHIY2 = CHIY2 - RN*CHIY2/X2
CHIY2 = CHIY2 - RN*CHIY2/Y2

ANCAP = REFREL*OLX1-OLX2
ANCAP = ANCAP/(REFREL*OLX1-CHIY2 - CHIPX2)
ANCAP = ANCAP/(CHIY2*OLX2 - CHIPX2)
BRACK = ANCAP*(CHIY2*OLY2 - CHIPY2)

BNCAP = REFREL*OLX2 - OLX1
BNCAP = BNCAP/(REFREL*CHIY2 - OLX1*CHIX2)
BNCAP = BNCAP/(CHIY2*OLX2 - CHIPX2)
CRACK = BNCAP*(CHIY2*OLY2 - CHIPY2)
FILE: COATSPHER FORTRAN A1

ANESS1 = BRACK*CHIPY2
ANESS2 = BRACK*CHIPY2
ANESS3 = CRACK*CHIPY2
ANESS4 = CRACK*CHIPY2

IF (CQABS(ANESS1).GT.DEI.*CQABS(DLY2)) GO TO 999
IF (CQABS(ANESS2).GT.DEI.) GO TO 999
IF (CQABS(ANESS3).GT.DEI.*CQABS(DLY2)) GO TO 999
IF (CQABS(ANESS4).GT.DEI.) GO TO 999

BRACK = QCMPLX (0.001,0.000)
CRACK = QCMPLX (0.001,0.000)
IFLAG = 1

999 DNBAR = DLY2 - BRACK*CHIPY2
GNBAR = DNBAR/(1.000 - BRACK*CHIPY2)
GNBAR = DLY2 - CRACK*CHIPY2

AN = (DNBAR/RFREL2-RH/Y)*PSIX - PSIY
BN = (RFREL2*GNBAR-RH/Y)*XTY - XZLY
BN = BN/(RFREL2*GNBAR-RH/Y)*XTY-XZLY

PSI15 = PSI1Y
PSI1Y = PSI1X
CHI1X = CHI1Y
CHI1Y = CHI1X
XZLY = QCMPLX(PSI1Y,-CHI1X)

CHI1X2 = CHI1X2
CHI1X2 = CHI1X2
CHI1Y2 = CHI1Y2
CHI1Y2 = CHI1Y2

J0X1 = D1X1
J0X2 = D1X2
J0Y2 = D1Y2
N = N - 1

RETURN
END

SUBROUTINE SIGD(XX, IOR, DBIGD)

SUBROUTINE TO CALCULATE LOGARITHMIC DERivatives BY LENZ'S METHOD USING DOWN RECURSION

COMMON/BLOCK3/YSTOP
REAL*16 XINV,XX,YSTOP
COMPLEX*12 ZINV, IOR, FF, AK, DEN, NUM, MTN, DTD, TT, DBIGD
DATA EPS1/L.Q-2/EPS2/L.Q-8/
DATA MAXT/10000/
NT = YSTOP
NTP1 = NT - 1
NTP2 = NT - 2
FILE: COATSPHR FORTRAN AL

MAKE SURE BIGO ARRAY WILL BE BIG ENOUGH

IF(NTPL.LE.20150) GO TO 10
WRITE(*,8000) MT
8000 FORMAT(/**/"ESTIMATED LENGTH OF MIX SERIES NT=",I6)
STOP

COMPUTE BIGO

10 XINV = 1.0QQ/XX
     ZINV = XINV/ZOR

PREPARE FOR DOWN RECURRANCE-COMPUTE INITIAL HIGH ORDER
BIGO=M USING LENTZ METHOD

FF = NTPL-ZINV
MM = -1
KK = 2+NT-3
AK = (MM+KK)*ZINV
DEN = AK
NUM = DEN + 1.0QQ/FF
COUNT = 1

20 KOUNT = KOUNT+1
IF(KOUNT.GT.MAXIT) GO TO 40
IF(CQABS(NUM/AK).GT.EPSL.AND.CQABS(DEN/AK).GT.EPSL) GO TO 30
     MM = -MM
     KK = KK-3
     AK = (MM+KK)*ZINV
     NUM = AK+NUM + 1.0QQ
     DTD = AK+DEN + 1.0QQ
     FF = (NUM/DDTD)*FF
     MM = -MM
     KK = KK-3
     AK = (MM+KK)*ZINV
     NUM = AK = NUM/NIN
     DEN = AK = DEN/DDTD
     KOUNT = KOUNT + 1
     GO TO 20

30 TT = NUM/DEN
      FF = TT*FF

CHECK FOR CONVERGENCE

IF(QABS(QREAL(TT)-1.0QQ).LT.EPSL2 .AND. QABS(QIMAG(TT)).LT.EPSL2)
   * GO TO 30
     MM = -MM
     KK = KK+3
     AK = (MM+KK)*ZINV
     NUM = AK = 1.0QQ/NUM
     DEN = AK = 1.0QQ/DEN
     GO TO 20

40 WRITE(*,8001) NT,XX,ZOR,AK,NUM,DEN,TT,FF
FILE: COATSPHR FORTRAN A1

8001 FORMAT('CONTINUED FRACTION FOR A-SUB-MT FAILED TO CONVERGE':
   ' NT=' ,I6,' X=' ,2E20.8,' REF INDEX=' ,2E20.8,' AK=' ,2E20.8/
   ' NUM=' ,2E20.8,' DEN=' ,2E20.8,' TT=' ,2E20.8,' FF=' ,2E20.8)
STOP
50 DBXGD = FF
RETURN
C END
FILE: ESCAT FORTRAN A1

PROGRAM ESCAT TO COMPUTE COMPLEX SCATTERING AMPLITUDES S1 AND S2 USING A PHYSICAL OPTICS APPROXIMATION AND DIFFRACTION.

D.L. KINGSBURY AND P.L. MARSTON 1980
D.S. LANGLEY 1982
MODIFIED FOR CMS BY S.C. BILLETTE APRIL 7, 1986

**************************************************************

INPUT PARAMETERS

N = RELATIVE REFRACTIVE INDEX OF BUBBLE
KA = SIZE PARAMETER
PHIBGN = STARTING ANGLE IN DEGREES
PHIEND = LAST ANGLE (DEGREES)
DPHI = ANGLE STEP SIZE (DEGREES)

**************************************************************

REAL M, KA, S1, S2, SD1, SD2, P1, P2
COMPLEX S1, S2, S01, S02, S11, S12, F01, F02, F11, F12, SD1, SD2
DOUBLE PRECISION N, THC, ST1

DATA RHMN/0.0/
N = .75
KA = 100.0
PHIBGN = 30.0
PHIEND = 90.0
DPHI = 1.0/18.0

II > 1 GIVES P.O.A RESULTS USING ONLY (0,0) & (1,0) RAYS
JJ > 1 GIVES DIFFRACTION ONLY RESULTS
KK > 1 GIVES P.O.A. INCLUDING DIFFRACTION RESULTS

WRITE(1,1000) M, KA
1000 FORMAT(1X,'M = ',F4.4,' KA = ',F4.2)

compute critical angles for given refractive index

PI = 4.000*DATAN(1.000)
N = 1.000/M
THC = DATAN(1.000/DATAN((N**2-1.000)))
PHC = PI - 2.0*THC

BEGIN COMPUTATION FOR SCATTERING ANGLE PHI (RADIANS)

L = 0
PHID = PHIBGN
IF(PHID.GT.PHIEND) STOP
90 PHI = PHID/180.0

compute (0,: ray parameters
FILE: ESCAT FORTRAN AL

TH0 = INCIDENCE ANGLE
G01,G02 = PHASES FOR J = 1,2 (CENTER REF.)
F01,F02 = S.P.A. FAR ZONE COMPLEX AMPLITUDES FOR J = 1,2

TH0 = (PI-PHI)/2.0
CT0 = COS(TH0)
ST0 = SIN(TH0)
W = SIN(PHC-PHI)*SQRT(KA*COS(SNGL(THC))/(2.0*PI))

CALL FRES(W,FC,FS)
F01 = CNPLX(FC,FS)*CEXP(CNPLX(0.0,-1.0*PI/4.0))/SQRT(2.0)
F02 = F01
G01 = -1.0*KA*CT0
G02 = G01

INITIALIZE (1,0) RAY SCATTERING AMPLITUDES:
IF PHI EXCEEDS PI/2 THERE IS NO (1,0) RAY, AND NO
(0,0) REFLECTION PHASE SHIFT.

$11 = CNPLX(0.0,0.0)
$12 = CNPLX(0.0,0.0)
IF(PHI.GE.PHC) GO TO 60

COMPUTE (1,0) RAY PARAMETERS:
TH1 = INCIDENT ANGLE
R01 = REFRACTION ANGLE
G11,G12 = PHASES FOR J = 1,2 (CENTER REFERENCED)
D1 = GEOMETRIC DIVERGENCE FACTOR
RL1,RL2 = FRESNEL COEFFICIENTS FOR J = 1,2
F11,F12 = S.P.A. FAR ZONE COMPLEX AMPLITUDES

PH2 = PHI/2.0
TH1 = ATAN(N*SIN(PH2)/((1.0-M)*COS(PH2))
CT1 = COS(TH1)
ST1 = SIN(TH1)/CT1
R01 = DATAN(N*ST1/DSQRT(1.000-(N*ST1)**2))
CR1 = COS(R01)
G11 = 2.0*KA*(M*CR1-CT1)
G12 = G11
TAU = N
IF(CR1.GT.RMIN) TAU = TAU*CT1/CR1
D1 = N*CT1+2/(4.0*(Tau+1.0)*(1.3-M*COS(PH2);*COS(PH2))
DIF1 = R01 = TH1
SUM1 = RO1 = TH1
SDF = SIN(DIF1)
SSM = SIN(SUM1)
TDF = TAN(DIF1)
TSN = TAN(SUM1)

IF(SDF.LE.RMIN) TDF = 1.0-N
IF(SDF.LE.RMIN) TSN = 1.0-N
IF(SDF.LE.RMIN) SDF = 1.0-N
FILE: ESCAT FORTRAN A1

R11 = (SDF/SSM)**2
R12 = (TDF/TSM)**2
F11 = CMPLX(2.0*(1.0-R11)*SQRT(D1),0.0)
F12 = CMPLX(2.0*(1.0-R12)*SQRT(D1),0.0)

INCLUDE REFLECTION PHASE SHIFTS FOR (0,0) RAY:

DEL = SQRT(STG**2 - M**2)/C0
DEL1 = 2.0*ATAN(DEL)
DEL2 = 2.0*ATAN(N*M*DEL)
G01 = G01 - DEL1
G02 = G02 - DEL2

COMPUTE (1,0) AND (0,0) RAY SCATTERING AMPLITUDES IN
STANDARD UNITS (E.G. VAN DE HULST, WISCOMBE)

S11 = CMPLX(0.0,-1.0*KA/2.0)*F11*CEXP(CMPLX(0.0,G11))
S12 = CMPLX(0.0,-1.0*KA/2.0)*F12*CEXP(CMPLX(0.0,G12))
60 S01 = CMPLX(0.0,-1.0*KA/2.0)*F01*CEXP(CMPLX(0.0,G01))
S02 = CMPLX(0.0,-1.0*KA/2.0)*F02*CEXP(CMPLX(0.0,G02))

SUM THE (0,0) AND (1,0) RAY SCATTERING AMPLITUDES

S1 = S01 + S11
S2 = S02 + S12
ANG = PH1D
RS1 = REAL(S1)
CS1 = AIMAG(S1)
RS2 = REAL(S2)
CS2 = AIMAG(S2)
T1 = (2.0*CABS(S1)/KA)**2
T2 = (2.0*CABS(S2)/KA)**2
IF (II.LE.1) GO TO 888
WRITE(1,1100)ANG,II,T2
1100 FORMAT (F7.4,2E14.6)
888 CONTINUE

COMPUTE THE DIFFRACTED AMPLITUDES SD1 AND SD2
WITH CENTER REFERENCED PHASES

ARG = KA*SIN(PHI)
DP = 0.5
IF (ARG.LT.0.0) ARG = ABS(ARG)
IF (ARG.LE.RHCM) GO TO 90
IF (ABS(PH1D).GE.90.0) GO TO 95
80 DP = COS(PHI)*REJX(ARG)
90 SD1 = CMPLX(DP*KA**2,0.0)
95 IF (ABS(PH1D).GE.90.0) SD1 = CMPLX(0.0,0.0)
SD2 = SD1
ANG = PH1D
RS01 = REAL(SD1)
CS01 = AIMAG(SD1)
RS02 = RS01
CS02 = CS01
FILE: ESCAT  FORTRAN AL

ID = (2.0*CABS(S1)/KA)**2
IF(JJ.LE.1) GO TO 889
WRITE(1,1200) ANG, ID
1200 CONTINUE

COMPUTE TOTAL AMPLITUDES FOR F.O.A. PLUS DIFFRACTION

S1 = S1 - SDL
S2 = S2 - SD2
ANG = PHID
RS1 = REAL(S1)
CS1 = AIMAG(S1)
RS2 = REAL(S2)
CS2 = AIMAG(S2)
IT1 = (2.0*CABS(S1)/KA)**2
IT2 = (2.0*CABS(S2)/KA)**2
IF(KK.LE.1) GO TO 890
WRITE(1,1300) ANG, IT1, IT2
1300 CONTINUE

STEP PHI UP BY DPPI AND REPEAT UNTIL PHIEND

L = L - 1
PHID = PHID - L*DPHI
IF(PHID.LE.PHIEND) GO TO 50
STOP
END

SUBROUTINE FRES(WE, FS, FC)

COMPUTES THE FRESEL INTEGRAL FROM 0 TO WE

W = ABS(WE)
F = (1.0 - 0.926*W)/(2.0 - 1.792*W - 3.104*W**2)
G = 1.0/(2.0 - 4.142*W - 3.492*W**2 - 6.870*W**3)
A = 1.870798*W**2
C = COS(A)
S = SIN(A)
FC = 0.5 - F*S - G*C
FS = 0.5 - F*C - G*S
IF(WE.LE.0) GO TO 3
FC = FC + 0.5
FS = FS + 0.5
RETURN
3 FC = 0.5 - FC
FS = 0.5 - FS
RETURN
END

FUNCTION BJ1X(X)

COMPUTES BESSEL FUNCTION J1(X), AND DIVIDES BY X:
USING THE POLYNOMIAL APPROXIMATIONS FROM ABRAMOWITZ
& STEGEM
FILE: ESCAT FORTRAN AL

DIMENSION A(7), B(7), C(7)
DATA A/0.5,-0.56249985,0.31092573,-0.03954289,0.30448359, + 0.1761E-4,1.1053E-5/  
DATA B/0.79734564,1.56E-6,0.01699667,1.7105E-6,-0.00249511, + 0.00133511,-2.0033E-6/  
DATA C/-3.35419449,0.12499612,5.65E-5,-0.00637879,7.4248E-4, + 7.8824E-4,-2.9164E-6/  
IF(ABS(X).LT.3.0) GO TO 20
Z = ABS(3.0/X)
F = B(7)
T = C(7)
DO 10 K = 4,1,-1
F = B(K) + Z*F
T = C(K) + Z*T
10 CONTINUE
BJX = F*COS(X*T)/(X*SQRT(X))
RETURN
20 Z = (X/3.0)**2
BJX = A(7)
DO 30 K = 4,1,-1
BJX = A(K) + Z*BJX
30 CONTINUE
RETURN
END
APPENDIX B
SUPPLEMENTAL I₁ EXAMPLES

Figures B.1 - B.9 give the results for calculations for the \( j = 1 \) case (perpendicular E-field vector polarization) similar to those performed in Chapter 2. There are several differences in the features for the \( I₁ \) case which should be mentioned. First, the fine structure variation is much more prominent than for the \( I₂ \) case as can be seen in Figs. B.1 - B.4. This behavior is also shown for coated bubble cases. Uncoated bubble results show that for \( 30° < \theta < 90° \) the positions of the fine-structure intensity oscillations for \( j = 2 \) are typically shifted from those for \( j = 1 \) by one half of a fine-structure quasi-period, except where \( \theta \) is slightly less than \( \theta_c \). This fine structure shift is also present in the coated bubble results although it appears that the shift towards \( \theta_c \) in \( I₁ \) is slightly greater than for \( I₂ \) at comparable coating thickness values.

One prominent feature seen in the \( j = 1 \) case and not the \( j = 2 \) case is the anomalous modulation in coarse structure for large values of \( h \) in the \( \theta = 70° \) to \( 90° \) region as is shown in Figs. B.2(e) and B.3(e). This behavior was not seen in the \( j = 2 \) cases (compare Fig. 2.8(e) and 2.9(e)). The apparent cause of this modulation is the nature of the reflection coefficient at the water-coating interface for the two cases. Consider the reflection at the water-coating interface of ray \((0,0)_b\) shown in Fig. 2.7. From geometric considerations the local angle of incidence \( i \) is related to the scattering angle \( \theta \) by

\[
i = (180° - \theta)/2.
\]  

(B.1)

The relative amplitudes of the waves associated with this ray in the two cases \( j = 1 \) and
are given by comparison of the Fresnel (or plane surface) reflection coefficients $r_j$,

\[ r_1 = \frac{\sin(i - r)}{\sin(i + r)}, \quad r_2 = \frac{\tan(i - r)}{\tan(i + r)} \]  

(B.2a,b)

where $r$ denotes the local refraction angle and is given by $r = \sin^{-1}\left(\frac{n_w}{n_c}\sin i\right)$.

Consider the case $\theta = 82.82^\circ$ for which (B.1) gives $i = 48.59^\circ$. Then for $n_w = 4/3$ and $n_c = 1.5$ we have $r = 41.81^\circ$ and (B.2) gives $r_1 = 0.1181$ and $r_2 = -0.0008$; clearly $|r_1| > |r_2|$. For the case $j = 1$ it appears that the amplitude of the $(0,0)_b$ ray may be sufficient to interfere with that of the $(0,0)$ ray in Fig. 2.7 to cause the anomalous modulations. Because this effect only occurs in the $j = 1$ case, the choice of $j = 2$ for bubble sizing and detection is further supported.

Another feature which was much more prominent in $I_1$ was the presence of contrast modulations created by the superposing of the $(0,0)$ and $(3,1)$ ray amplitudes onto the fine-structure interference pattern of the $(0,0)$ and $(2,1)$ ray amplitudes (e.g., $\theta = 67^\circ$, $\theta = 85^\circ$ in Fig. B.2(a)). These modulations were discussed by Langley and Marston for the case of an uncoated bubble in Ref. 13 of Chapter 2. Note that as the thickness of the coating is increased other modulations occur (i.e., $\theta = 72^\circ$, $75^\circ$ in Fig. B.2(d)) which appear to be unique to that particular coating value. This effect is most prominent for smaller bubbles ($ka < 1000$). The apparent reason for this behavior is due to the fact that the approximate angular period of the contrast modulations $(\Delta \theta)_M$ is proportional to $\lambda/(B_3 - B_2)$ where $B_3$ is the distance between the $(0,0)$ and $(3,1)$ ray and $B_2$ is the distance between the $(0,0)$ and $(2,1)$ ray. The introduction of a coating invariably alters the distance between $B_3$ and $B_2$ resulting in the new modulations. For most other cases the behavior of $I_1$ was found to be similar to the analogous cases presented in Chapter 2 for $I_2$. These results are presented in Figs. B.5 - B.9.
Figure B.1. Calculated normalized scattering irradiances for $ka = 100$, $n_w = 4/3$, $n_c = 1.5$ for the perpendicular polarization $I_1$ in the critical angle scattering region. The solid curve is from Mie theory, the thick-dashed curve is the physical-optics approximation, and the thin-dashed curve is the calculated coated sphere results for (a) 0.25 μm, (b) 0.5 μm, (c) 1.0 μm, (d) 2.0 μm, and (e) 3.0 μm. Note the greater amount of fine structure as compared to the $I_2$ case in Figures 2.5 (a) - (e).
$ka = 100$

**Fig. B.1 (b)**

**scattering angle (deg)**

$I_1$
$ka = 100$

![Graph](c)

**scattering angle (deg)**

Fig. B.1 (c)
Fig. B.1 (d)

Scattering angle (deg)

$k_a = 100$

$d$

$I_1$

0 45 60 65 70 75 80 85 90

8 6 4 2 0
Figure B.2. Like Figure 2.8 but for $j = 1$ case.
Fig. B.2(a)

\( \text{scattering angle (deg)} \)

\( \text{ka} = 500 \)

\( I_1 \)
Fig. B.2 (c)

ka = 500

scattering angle (deg)
Figure B.3. Like Figure 2.9 but for j = 1 case.
Fig. 8.3 (a)

(a)
$ka = 1000$

**Fig. B.3 (c)**

scattering angle (deg)
Figure B.4. Like Figure 2.10 but for $j = 1$ case.
Figure B.5. The effect of changing $n_c$ with $ka = 100$ and $h = 0.26$. The thick-dashed curve represents $n_c = 1.45$, the thin-dashed curve is for $n_c = 1.5$, and the solid curve represents the noncoated case.
\[ ka = 100 \]

Scattering angle (deg)

Fig. B.5
Figure B.6. Like Figure B.5 but the thick-dashed curve represents \( n_c = 1.55 \).
$ka = 100$

Fig. B.6
Figure B.7. The effect of changing $n_c$ with $ka = 1000$ and $h = 3.14 \, \mu m$. The thin-dashed curve is for $n_c = 1.45$ and the solid curve is for $n_c = 1.5$. (The noncoated case is omitted.)
Figure B.8. Like Figure B.7 but thin-dashed curve represents $n_c = 1.55$. 
Figure B.9. Calculated normalized irradiances for a strongly absorbing coating
\( (n_c = 1.5 \pm \text{i} \times 10^{-3}) \) for \( k a = 1000 \) and (a) \( h = 0.99 \text{ \mu m} \), (b) \( h = 2.0 \text{ \mu m} \), (c) \( h = 3.14 \text{ \mu m} \),
and (d) \( h = 5.00 \text{ \mu m} \). The solid line is the non-absorbing coat result. Note that in case (a)
the absorbing case (dashed) is virtually the same as the non-absorbing case.
Fig. B.9 (b)

scattering angle (deg)

$ka = 1000$

$I_1$
APPENDIX C

EFFECTS OF THE REFRACTIVE INDEX OF WATER
ON SCATTERING PATTERNS

For the computations done in Chapter 2 and Appendix B the value of \( n_w = 4/3 \) was used. This value was chosen as it represents roughly the typical value for conditions found both inside and outside the lab. Tests were performed over the range of \( n_w = 1.332 \) (pure water at a temperature = 30°C) to \( n_w = 1.338 \) (high salinity in ocean water) in order to determine the range of validity of \( n_w = 4/3 \). Typical results are shown for the two extremes in Figs. C.1 and C.2. As can be seen in both cases there is a slight shift in the coarse structure while fine structure retains its approximate structure. This shift was found to be less prominent for larger bubbles. Typically values of \( n_w \) within \( \pm 0.002 \) (1.332 to 1.335) were found to have a negligible effect on all but the thickest coating values where a slight amount of added coarse structure shift was noted. Under unusual conditions such as extreme water temperatures or high salinity where variation of \( n_w \) is greater than 0.002 the measured value of \( n_w \) given in the tables is best used.

Figure C.3 compares the \( j = 2 \) irradiance values for \( n_w = 1.332, \) and \( n_w = 1.338 \) to \( n_w = 4/3 \) for a non-coated air bubble over the entire 180° range. As can be seen from the figure the critical angle region shows good agreement between all three values. In the majority of cases the approximation of \( n_w = 4/3 \) was valid for both coated and non-coated air bubble calculations as the behavior of all earlier described effects was unaltered. Since most values of sea water are within the \( \pm 0.002 \) range of \( n_w = 4/3 \) the results presented here may be used as accurate models of possible coating effects.
Figure C.1. Water index of refraction evaluation for the lower extreme case of
$n_w = 1.332$, (dashed curve). The solid curve is for $n_w = 4/3$. The other parameters
were $ka = 500, n_c = 1.5$. Note that the 4/3 value is a very good approximation in most
cases.
Fig. C.1 (a) and (b)
Figure C.2. Like Figure C.1 but for $n_w = 1.338$
Fig. C.2 (a) and (b)

scattering angle (deg)

ka = 500
Figure C.3. Comparison of irradiance profiles for $n_w = 4/3$ (long-dashed curve) versus $n_w = 1.332$ (solid curve) and $n_w = 1.338$ (short-dashed curve) for noncoated air bubble in water. Note the excellent agreement between the three curves in the $\theta = 60^\circ$ to $\theta = 85^\circ$ range.
ka = 100

![Graph showing scattering angle vs. log(l2)](image)

scattering angle (deg)

Fig. C.3.
SUPPLEMENT TO BILLETTE'S THESIS

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Following the completion of Stuart Billette's thesis (which constitutes the bulk of the present report) an error was discovered in the computer program used to evaluate the physical-optics approximation of Marston and Kingsbury. This error (which could be traced to a previous student) is only concerned with the physical-optics approximation for the scattering from an uncoated bubble. Hence all of the new results of this thesis, which concern the scattering from coated bubbles, were not affected by this error. Furthermore, the Mie theory results (which are exact for an uncoated spherical bubble) were not affected by this error. Indeed, the effect of the programming error on the computed physical-optics approximation (POA) to the normalized irradiances $I_j$ is typically only a few percent of the corrected POA values if $ka \geq 500$.

For completeness, the next few pages show the corrected (solid curve) and erroneous (dashed curve) POA predictions for $ka = 100, 500, 1000,$ and $2500$. The first set of four plots give $I_2$. The solid curves of the plots supersede the appropriate POA curves in Fig. 2.5 and 2.8 - 2.13. The second set of four plots give $I_1$. The solid curves of these plots supersede the appropriate POA curves in Fig. B.1 - B.4. Except for $ka = 100$ the differences resulting from this correction would probably not be noticeable.

These corrections do not affect any of the conclusions of this thesis.

The programming error in the POA algorithm (FILE: ESCAT) is on page 101. To correct the error change:
CALL FRES(W, FC, FS)

to read:

CALL FRES(W, FS, FC).

This error was not present in computer programs used in the original development of the
Marston, ibid 71, 358-361 (1981).] Consequently the POA predictions there are correctly
plotted [see, however, J. Opt. Soc. Am. 71, 917 (1981).]

It is appropriate to comment on an additional aspect of Billette's thesis. Recent
tests were performed which give additional support to the correctness of the algorithm
(FILE: COATSPHR) used to evaluate the exact partial-wave series for scattering from a
spherical bubble. The procedure was to consider the case of a shell with $n_o = 4/3$,
$n_c = 1.5$, and $n_i = 1$ in the limit $a/b \rightarrow 0$ so that the radius $a$ of the bubble was
negligible in comparison to both the wavelength and the coating radius $b$. In this case the
scattered irradiance went over to that of a uniform sphere (of relative refractive index
$n_c/n_o$) as predicted from Mie theory.
KA = 100

\[ I_2 \]

scattering angle (deg)

KA = 500

\[ I_2^2 \]

scattering angle (deg)
Scattering angle (deg)

KA = 1000

KA = 2500

I_2^2

scattering angle (deg)
$KA = 1000$

$\text{scattering angle (deg)}$

$\text{scattering angle (deg)}$

$KA = 2500$
Billette Report
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