Understanding light scattering by a coated sphere
Part 1: Theoretical considerations

James A. Lock1,* and Philip Laven2

1Department of Physics, Cleveland State University, Cleveland, Ohio 44115, USA
29 Russells Crescent, Horley RH6 7Dj, UK
*Corresponding author: j.lock@csuohio.edu

Received April 9, 2012; revised May 30, 2012; accepted May 31, 2012; posted May 31, 2012 (Doc. ID 166285); published July 9, 2012

Although scattering of light by a coated sphere is much more complicated than scattering by a homogeneous sphere, each of the partial wave amplitudes for scattering of a plane wave by a coated sphere can be expanded in a Debye series. The Debye series can then be rearranged in terms of the various reflections that each partial wave undergoes inside the coated sphere. For a given number of internal reflections, it is found that many different Debye terms produce the same scattered intensity as a function of scattering angle. This is called path degeneracy. In addition, some of the ray trajectories are repeats of those occurring for a smaller number of internal reflections in the sense that they produce identical time delays as a function of scattering angle. These repeated paths, however, have a different intensity as a function of scattering angle than their predecessors. The degenerate paths and repeated paths considerably simplify the interpretation of scattering within the coated sphere, thus making it possible to catalog the contributions of the various paths. © 2012 Optical Society of America

OCIS codes: 080.5602, 290.4020.

1. INTRODUCTION

The exact solution to scattering of an electromagnetic plane wave by a coated sphere was first obtained in terms of an infinite series of partial wave contributions by Aden and Kerker in 1951 [1], and numerical computations of coated sphere scattering in the Mie regime appeared a decade later [2]. Since that time, research into coated sphere scattering has expanded in a number of directions. For example, it has been found that the one-internal-reflection rainbow of a homogeneous sphere evolves into two distinct components when a thin coating surrounds the sphere [3,4]. This phenomenon has found use in various liquid refractometry measurement methods [5,6]. The physical parameters of either the core or coating strongly affect both the relation between the wavelength and particle size for the excitation of morphology dependent resonances of a coated sphere and the structure of the interior source function at resonance [7–13]. Coated sphere scattering, averaged over a particle size distribution, has been used to study the effect of condensation of water on soot particles for the absorption of sunlight in the atmosphere [14]. Other research in coated sphere scattering has focused on improving the robustness of numerical computations. Potential instabilities in the numerical computation of the spherical Bessel functions appearing in the coated sphere partial wave scattering amplitudes for complex coating and core refractive indices have led to a series of improved scattering algorithms [15–18]. Lastly, the problem of scattering of a plane wave by a coated sphere has been used as a starting point for various enhancements to light scattering theory, such as scattering of a focused Gaussian beam [19–21] by a multilayer sphere [22–25]. Recently, ray scattering methods have been applied to coated sphere scattering, and were compared to the Aden–Kerker scattered intensity averaged over a narrow particle size distribution [26].

A more detailed comparison between wave scattering and ray scattering that takes into account interference between the various ray contributions would be obtained by using the Debye series, which serves as a bridge between Mie theory and the ray model for a particle whose size is much larger than the wavelength of the incident light. It decomposes each of the Mie partial wave scattering amplitudes into an infinite series of terms that describe diffraction, external reflection, and transmission following p – 1 internal reflections for p ≥ 1 [27]. In previous studies [3,28], the Debye series has been obtained for scattering by a coated sphere. In this paper and in a companion paper [29] we examine and interpret the behavior of many of the Debye terms that contribute to coated sphere scattering of an electromagnetic plane wave. We find that, in its original form, the coated sphere Debye series is not especially well suited to this task. Thus in this paper we reorganize the terms of the series into a succession of single-scattering contributions, in analogy to the expanded form [30] or the order of scattering formalism [31] of multiple scattering of waves from a collection of target particles, so as to simplify the comparison.

Such a reorganization might seem relatively uncomplicated. However, we found that expressing the partial wave scattering amplitudes directly in terms of single-interaction amplitudes was not straightforward, and along the way a number of unanticipated features were encountered. Specifically, we uncovered two geometric relationships that greatly simplify the job of cataloging and organizing all the Debye terms. First, most of the Debye terms have some degree of degeneracy, i.e., a number of different paths of the light rays (corresponding to the partial waves via van de Hulst’s localization principle [32]) have exactly the same scattered intensity as a function of scattering angle. These paths are thus effectively amplified in the scattered field by the degeneracy factor.
fact explains some of the discrepancies between ray theory and Aden-Kerker scattering obtained in [36]. Second, for a given number of internal reflections, many of the Debye terms encountered are “repeats” of terms that occurred for a smaller number of internal reflections in the sense that the scattering angle of the corresponding rays as a function of the incident ray impact parameter is the same as before. The scattered intensity as a function of scattering angle, however, is different than for fewer internal reflections.

The ideal context in which to examine and interpret the various Debye terms is time domain scattering where a short pulse of electromagnetic radiation is incident on the coated sphere and the scattered intensity is calculated as a function of both scattering angle and delay time of the scattered pulse. This method separates the contributions of the different Debye terms that occur at the same scattering angle but at different delay times [33-35] in much the same way that the Fourier transform of a two dimensional image separates all the different spatial frequencies contained in the image. It also removes much of the interference between the different contributions. Time domain analysis of coated sphere scattering is the subject of a companion paper [29].

2. DEBYE SERIES FOR SCATTERING BY A COATED SPHERE

Consider a coated sphere consisting of a core (region 1) of radius $a_{12}$ and real refractive index $m_1$ that is concentrically surrounded by a coating (region 2) of refractive index $m_2$. The overall radius of the coated sphere is $a_{23}$. An electromagnetic plane wave of vacuum wave number $k = 2\pi/c$, traveling in the positive $z$ direction in an exterior medium of real refractive index $m_3$, and polarized in the $x$ direction is incident on the coated sphere. The partial wave scattering amplitudes $a_n, b_n$ for the transverse magnetic (TM) and transverse electric (TE) polarizations, respectively, and where $n$ is the partial wave number, may be conveniently written in terms of the six partial wave amplitudes

$$N_n^{12} = a_n(m_2ka_{12})\psi_n(m_1ka_{12}) - \beta\psi_n(m_2ka_{12})\psi_n(m_1ka_{12})$$

(1a)

$$D_n^{12} = \alpha\psi_n(m_2ka_{12})\psi_n(m_1ka_{12}) - \beta\psi_n(m_2ka_{12})\psi_n(m_1ka_{12})$$

(1b)

$$N_n^{23} = \gamma\psi_n(m_3ka_{23})\psi_n(m_2ka_{23}) - \delta\psi_n(m_3ka_{23})\psi_n(m_2ka_{23})$$

(1c)

$$D_n^{23} = \gamma\psi_n(m_3ka_{23})\psi_n(m_2ka_{23}) - \delta\psi_n(m_3ka_{23})\psi_n(m_2ka_{23})$$

(1d)

$$P_n^{23} = \gamma\psi_n(m_3ka_{23})\chi_n(m_2ka_{23}) - \delta\psi_n(m_3ka_{23})\chi_n(m_2ka_{23})$$

(1e)

$$Q_n^{23} = \gamma\psi_n(m_3ka_{23})\chi_n(m_2ka_{23}) - \delta\psi_n(m_3ka_{23})\chi_n(m_2ka_{23})$$

(1f)

where $\psi_n$ are Riccati-Bessel functions and $\chi_n$ are Riccati-Neumann functions in the notation of [36]. $\alpha = m_1$, $\beta = m_2$, $\gamma = m_3$, and $\delta = m_3$ for the TE polarization, and $\alpha = m_2$, $\beta = m_1$, $\gamma = m_3$, and $\delta = m_2$ for the TM polarization. The partial wave scattering amplitudes are expressed in terms of Eqs. (1a)-(1f) as [3]

$$a_n, b_n = (D_n^{12}N_n^{23} - N_n^{12}P_n^{23})/(D_n^{12}N_n^{23} - N_n^{12}Q_n^{23}) + i(D_n^{12}D_n^{23} - N_n^{12}Q_n^{23})].$$

(2)

The scattered intensity for unpolarized incident light is

$$I(r, \theta) = |E_0|^2/(2\mu_0c^2r^2)[|S_1(\theta)|^2 + |S_2(\theta)|^2],$$

(3)

where $E_0$ is the field strength of the incident plane wave, $\mu_0$ is the permeability of free space, $\theta$ is the scattering angle, $c$ is the speed of light, and $r$ is the distance from the center of the coated sphere to the detector. The scattering amplitudes $S_1(\theta)$ and $S_2(\theta)$ are

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

(4a)

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

(4b)

where $\pi_n(\theta)$ and $\tau_n(\theta)$ are the angular functions of Mie theory [37]. In the short wavelength limit, $S_1(\theta)$ asymptotically becomes the amplitude for TE-polarized scattering and $S_2(\theta)$ asymptotically becomes the amplitude for TM-polarized scattering.

The Debye series decomposes the partial wave scattering amplitudes into a sum of terms corresponding to diffraction, external reflection, and transmission accompanied by various numbers of internal reflections, and involves the two additional partial wave amplitudes

$$P_n^{12} = \gamma\psi_n(m_3ka_{12})\chi_n(m_1ka_{12}) - \beta\psi_n(m_2ka_{12})\chi_n(m_1ka_{12})$$

(5a)

$$Q_n^{12} = \alpha\psi_n(m_3ka_{12})\chi_n(m_1ka_{12}) - \beta\psi_n(m_2ka_{12})\chi_n(m_1ka_{12})$$

(5b)

as well as the expressions for the partial wave Fresnel reflection and transmission coefficients at each of the two interfaces. The quantities $R_n^{12}$ and $T_n^{12}$ are the Fresnel reflection and transmission coefficients for a radially incoming spherical multipole wave of partial wave number $n$ in region 3 incident on the coating/exterior interface. Similarly, $R_n^{23}$ and $T_n^{23}$ are the Fresnel reflection coefficients for a radially outgoing spherical multipole wave in region 2 incident on the coating/exterior interface. $R_n^{12}$ and $T_n^{12}$ are the Fresnel coefficients for a radially outgoing spherical multipole wave in region 1 incident on the core/coating interface. It can then be straightforwardly shown by matching the boundary conditions of the components of the electric and magnetic fields at the interfaces [38] that

$$R_n^{23} = (-N_n^{23} + Q_n^{23} + iD_n^{23} + iP_n^{23})/(N_n^{23} + Q_n^{23} + iD_n^{23} - iP_n^{23})$$

(6a)

$$R_n^{23} = (-N_n^{23} + Q_n^{23} - iP_n^{23} + iD_n^{23})/(N_n^{23} + Q_n^{23} + iD_n^{23} - iP_n^{23})$$

(6b)
\[ T_{n2}^{32} = -2im_2/(N_{n2}^{23} + Q_{n2}^{23} + iD_{n2}^{23} - iP_{n2}^{23}) \] (6c)
\[ T_{n3}^{23} = -2im_2/(N_{n3}^{23} + Q_{n3}^{23} + iD_{n3}^{23} - iP_{n3}^{23}) \] (6d)

\[ R_{n2}^{212} = (-N_{n2}^{12} + Q_{n2}^{12} + iD_{n2}^{12} + iP_{n2}^{12})/(N_{n2}^{12} + Q_{n2}^{12} + iD_{n2}^{12} - iP_{n2}^{12}) \] (6e)

\[ R_{n1}^{212} = (-N_{n1}^{12} + Q_{n1}^{12} - iD_{n1}^{12} - iP_{n1}^{12})/(N_{n1}^{12} + Q_{n1}^{12} + iD_{n1}^{12} - iP_{n1}^{12}) \] (6f)

\[ T_{n1}^{212} = -2im_1/(N_{n1}^{12} + Q_{n1}^{12} + iD_{n1}^{12} - iP_{n1}^{12}) \] (6g)

\[ T_{n1}^{21} = -2im_1/(N_{n1}^{12} + Q_{n1}^{12} + iD_{n1}^{12} - iP_{n1}^{12}) \] (6h)

The Debye series decomposition of the coated sphere partial wave scattering amplitudes is then

\[ a_n, b_n = [1 - R_{n2}^{212} - T_{n2}^{32}W_n T_{n2}^{23}]/(1 - W_n R_{n1}^{212})]/2 \]

\[ = [1 - R_{n2}^{212} - \sum_{q=1}^{\infty} T_{n2}^{32}(W_n R_{n1}^{212})q^{-1}W_n T_{n2}^{23}]/2, \] (7)

where

\[ W_n = R_{n1}^{212} + T_{n2}^{12}T_{n2}^{12}/(1 - R_{n1}^{212}) = R_{n2}^{212} + \sum_{p=1}^{\infty} T_{n2}^{32}(R_{n1}^{212})p^{-1}T_{n2}^{12}. \] (8)

Three different derivations of Eqs. (7) and (8) are given in [3,28,39].

3. REORGANIZATION OF THE DEBYE SERIES

Although Eqs. (7) and (8) express the partial wave scattering amplitudes in terms of the Fresnel transmission and reflection coefficients at the two interfaces, the specific sequence of transmissions and reflections of the different paths of the corresponding light rays is not readily evident in the equations. It was thus found to be useful to re-express the Debye series in terms of differing numbers of internal reflections \( N \), subdivided into \( N_{212} \) of the internal reflections \( R_{n2}^{212} \), \( N_{121} \) of the reflections \( R_{n1}^{212} \), and \( N_{232} \) of the reflections \( R_{n2}^{123} \) with the constraint

\[ N_{212} + N_{121} + N_{232} = N. \] (9)

The \( N = 0 \) portion of Eqs. (7) and (8) is then

\[ (a_n, b_n)_{N=0} = (1 - R_{n2}^{212} - T_{n2}^{32}T_{n2}^{12}T_{n2}^{23})/2. \] (10)

The first term of Eq. (10) when summed over partial waves quantitatively describes diffraction, the second term describes external reflection, and the third term describes transmission through the coating into the core and then transmission back out again, as indicated by ray path \( A \) in Fig. 1. The progression of superscripts in each term from left to right indicates the path of a corresponding light ray through the various regions from start to finish. In ray theory, if the core is small and the impact parameter of a ray incident on the coating/exterior interface is large as for ray path \( B \) in Fig. 1, it can be transmitted into the coating, miss striking the core, and be transmitted back out again. But the term \( T_{n2}^{32}T_{n2}^{23} \) that would seem to correspond to this situation does not occur in the \( N = 0 \) portion of the Debye series. This is because the Debye series describes radially incoming and outgoing spherical multipole waves. Once a radially incoming wave is transmitted from region 3 into region 2, it must either reflect off the core/coating interface again. The last two terms of Eq. (10) correspond to the ray paths shown in Figs. 2(a) and 2(b).

The \( N = 1 \) portion of Eqs. (7) and (8) is

\[ (a_n, b_n)_{N=1} = -T_{n2}^{32}(R_{n1}^{212} + T_{n2}^{12}R_{n1}^{212}T_{n2}^{12}) + T_{n2}^{32}T_{n2}^{12}T_{n1}^{23}T_{n1}^{23}/2. \] (11)

The three terms in Eq. (11) correspond to the ray paths shown in Figs. 2(c), 2(d), and 2(e). They describe, respectively, reflection from the core/coating interface with incidence from region 2, reflection from the core/coating interface with incidence from region 1, and reflection from the coating/exterior interface with incidence from region 2. Again, if a large impact parameter ray were incident on a coated sphere containing a small core, it could enter the coating, be internally reflected at the coating/exterior interface, and be transmitted back out without ever entering the core. An appropriate collection of such rays could participate in a one-internal-reflection rainbow entirely in the coating material, which would be identical to the one-internal-reflection rainbow of a homogeneous sphere composed of coating material. But again the term \( T_{n2}^{32}T_{n2}^{23} \), which would seem to correspond to this situation, does not occur in the \( N = 1 \) portion of the Debye series expansion. What had been the
one-internal-reflection rainbow of a homogeneous sphere is now contained in the $N$ Debye term $T_n^{12} R_{n}^{12} R_{n}^{21} T_{n}^{12}$ via tunneling external reflection of large partial waves with the amplitude $R_{n}^{12} \rightarrow 1$ both before and after the internal reflection at the coating/exterior interface.

In like manner, the $N$ Debye series terms rapidly increases as a function of $N$, there being $2$, $3$, $6$, $14$, $31$, $70$, $157$, $353$ terms for $0 \leq N \leq 7$, respectively. In order to simplify the cataloging of these Debye series terms, it will prove useful to parameterize them by the total number $N$ of internal reflections, the number of chords $A$ of the path of the corresponding light ray in the coating region between reflections, and the number of chords $B$ in the core region between reflections. Using this parameterization, the diffraction-plus-external reflection term in Eq. (10) is $(N, A, B) = (0, 0, 0)$ and the transmission term is $(0, 2, 1)$. The three terms in Eq. (11) are $(1, 2, 0)$, $(1, 2, 2)$, and $(1, 4, 2)$ respectively, and the six terms in Eq. (12) are $(2, 2, 3)$, $(2, 4, 1)$, $(2, 4, 1)$, $(2, 4, 3)$, $(2, 4, 3)$, and $(2, 6, 3)$, respectively. Figure 2 shows all the possible paths of the corresponding light rays for $N \leq 3$. It should be noted that for

The number of Debye series terms rapidly increases as a function of $N$, there being $2$, $3$, $6$, $14$, $31$, $70$, $157$, $353$ terms for $0 \leq N \leq 7$, respectively. In order to simplify the cataloging of these Debye series terms, it will prove useful to parameterize them by the total number $N$ of internal reflections, the number of chords $A$ of the path of the corresponding light ray in the coating region between reflections, and the number of chords $B$ in the core region between reflections. Using this parameterization, the diffraction-plus-external reflection term in Eq. (10) is $(N, A, B) = (0, 0, 0)$ and the transmission term is $(0, 2, 1)$. The three terms in Eq. (11) are $(1, 2, 0)$, $(1, 2, 2)$, and $(1, 4, 2)$ respectively, and the six terms in Eq. (12) are $(2, 2, 3)$, $(2, 4, 1)$, $(2, 4, 1)$, $(2, 4, 3)$, $(2, 4, 3)$, and $(2, 6, 3)$, respectively. Figure 2 shows all the possible paths of the corresponding light rays for $N \leq 3$. It should be noted that for

The terms in Eq. (12) correspond to the ray paths shown in Fig. 2(f)–2(k), respectively.
\( N = 2 \) the two Debye terms \( T_{21}^{E} R_{m}^{212} T_{21}^{R} T_{12}^{T} T_{23}^{T} \) and \( T_{21}^{E} T_{21}^{R} R_{m}^{212} R_{21}^{12} T_{21}^{T} T_{23}^{T} \) are both parameterized by \((2, 4, 1)\) and are reversed paths of each other, as is illustrated in Fig. 3. Each of these two Debye terms is merely a different ordering of the same collection of partial wave Fresnel transmission and reflection coefficients, which when summed as in Eqs. (4a) and (4b), produces the same scattered field as a function of the \( \theta \). Thus each of the \((2, 4, 1)\) Debye terms has the same scattering signature in Eq. (3). This is reminiscent of coherent back-scattering in the multiple scattering of light from a collection of many particles \([42, 43]\). If one considers the corresponding light rays for these two terms, the scattering angle \( \theta \) as a function of the angle of incidence \( \theta_{0} \) of an incoming ray and the optical path length of the scattered rays as a function of \( \theta_{0} \) are the same as well. Thus each \((2, 4, 1)\) Debye term has the same trajectory in scattering angle-delay time space for time domain scattering. This degeneracy also occurs for the \( N = 2 \) terms \( T_{21}^{E} T_{21}^{R} R_{m}^{211} T_{12}^{T} T_{23}^{T} \) and \( T_{21}^{E} T_{21}^{R} R_{m}^{212} R_{21}^{12} T_{21}^{T} T_{23}^{T} \), which are both parameterized by \((2, 4, 3)\). Thus we say that the degeneracy factor of \((2, 4, 1)\) and \((2, 4, 3)\) is \( D = 2 \) since two different Debye terms corresponding to two different ray paths have exactly the same scattering signature when the scattered intensity is plotted as a function of the scattering angle or the scattered pulse delay time is plotted as a function of the scattering angle.

The degeneracy factor was also determined for ray paths with larger \( N \). Let the angles \( \theta_{23}, \theta_{01}, \) and \( \theta_{12} \) be the transmitted angle of a ray from region 3 into region 2 at the coating/core interface, the angle of incidence in region 2 on the coating/core interface, and the transmitted angle from region 2 into region 1 at the coating/core interface, respectively. Then Snell’s law gives

\[
m_{1} \sin(\theta_{01}) = m_{2} \sin(\theta_{23}) \tag{13a}
\]

\[
m_{2} \sin(\theta_{23}) = m_{1} \sin(\theta_{12}) \tag{13b}
\]

and the coated sphere geometry gives

\[
a_{21} \sin(\theta_{23}) = a_{12} \sin(\theta_{12}) \tag{14}
\]

As a corresponding ray propagates through the coated sphere, each \( T_{21}^{E} \) and \( T_{21}^{R} \) factor contributes a deflection angle of \( \theta_{32} - \theta_{21} \), each \( T_{21}^{R} \) and \( T_{12}^{T} \) contributes \( \theta_{23} - \theta_{12} \), each \( R_{m}^{212} \) contributes \( \pi - 2 \theta_{23}, \) each \( R_{m}^{211} \) contributes \( \pi - 2 \theta_{12}, \) and each \( R_{m}^{12} \) contributes \( -\pi + 2 \theta_{12}. \) Applying this rule to all the ray paths for \( N \leq 7, \) the scattering angle was found to depend only on the number of chords \( A \) and \( B \) in the coating and core regions, the refractive indices \( m_{1}, m_{2}, m_{3} \), the radii \( a_{12} \) and \( a_{23} \), but not on \( N. \) Similarly, the optical path length associated with the incident ray and with the exiting ray in region 3 is \( m_{2} a_{23} \cos(\theta_{23}) \) and the optical length of each chord in the coating is \( m_{2} a_{23} \cos(\theta_{23}) - a_{12} \cos(\theta_{12}) \), and the optical length of each chord in the core is \( 2 m_{1} a_{12} \cos(\theta_{12}) \). The total optical path length also depends only on \( A \) and \( B, \) the refractive indices, and the radii, but not on \( N. \) As \( N \) increases, the complexity of the ray paths increases and the degeneracy factor of many of the \((N, A, B)\) terms also increases. As an example of increased degeneracy, one of the four \((4, 4, 5)\) Debye terms is \( T_{21}^{E} T_{21}^{R} R_{m}^{211} R_{m}^{12} R_{m}^{12} T_{12}^{T} T_{23}^{T} \) while the two degenerate \((3, 4, 2)\) terms have a extra factor in \((3, 4, 2). \) The extra \( \theta \) contributes \( \cos \). In addition to the partial wave \( T_{12}^{T} T_{23}^{T} \) and \( a \theta \) in the coating and core regions, each \( \theta \) and \( R \) of the \( B \) \( \sin \) further decreases the number of new paths that \( R \) \( a \) increases, the \( R \) \( \theta \) \( \sin \) and \( \theta \) are both parameterized by \((2, 4, 1)\) while the two degenerate \((2, 4, 3)\) terms have a extra factor in \((2, 4, 3). \) All three of the 121 reflections in this term occur before the 232 reflection. The other three degenerate \((4, 4, 5)\) terms have two, one, and zero of the 121 reflections occurring before the 232 reflection. Taking this path degeneracy into account, the number of independent Debye terms becomes 2, 3, 4, 7, 9, 13, 16, 21 for \( 0 \leq N \leq 7, \) as shown in Table 1.

Another effect that decreases the number of different paths for time domain scattering occurs for \( N \geq 3. \) For example, the path \((1, 4, 2)\) is \( T_{21}^{E} T_{21}^{R} R_{m}^{211} R_{m}^{12} T_{12}^{T} T_{23}^{T} \) while the two degenerate \((3, 4, 2)\) terms are \( T_{21}^{E} T_{21}^{R} R_{m}^{211} T_{12}^{T} T_{23}^{T} \) and \( T_{21}^{E} T_{21}^{R} R_{m}^{212} R_{m}^{12} T_{12}^{T} T_{23}^{T} \) and \( T_{21}^{E} T_{21}^{R} R_{m}^{212} R_{m}^{12} T_{12}^{T} T_{23}^{T} \). In addition to the partial wave Fresnel coefficients common to both terms, the \((1, 4, 2)\) term has an extra \( T_{12}^{T} T_{23}^{T} \) while the \((3, 4, 2)\) terms have a extra \( R_{m}^{212} R_{m}^{211}. \) According to the rules given above, both of these extra factors have the same deflection angle \( 2 \theta_{23} - 2 \theta_{12} \) of the corresponding light rays. Thus the \((1, 4, 2)\) and \((3, 4, 2)\) rays have the same scattering angle as a function of \( \theta_{0} \). They both have a rainbow at exactly the same scattering angle, and they both have the same glory structure for \( \theta = 180^\circ. \) Since the optical path length as a function of \( \theta_{0} \) depends only on \( A \) and \( B, \) the \((3, 4, 2)\) time domain trajectories in scattering angle-delay time space are a repeat of the \((1, 4, 2)\) path. This is illustrated in Fig. 4. However, when the partial wave scattering amplitudes of these Debye terms are summed in Eqs. (4a) and (4b), the scattered field of \((1, 4, 2)\) as a function of \( \theta \) differs from that of \((3, 4, 2)\) due to the extra \( \frac{T_{21}^{R}}{T_{21}^{E}} \) factor in \((1, 4, 2)\) and the extra \( R_{m}^{212} R_{m}^{211} \) factor in \((3, 4, 2). \) The extra \( \frac{T_{21}^{R}}{T_{21}^{E}} \) factor contributes more strongly to the scattered field for small partial waves corresponding to near-paraxial rays, while the extra \( R_{m}^{212} R_{m}^{211} \) factor contributes more strongly for larger partial waves corresponding to rays having near-grazing incidence on the core. Similarly for \( N = 4, \) the \((4, 4, 3)\) and \((4, 6, 3)\) time domain paths are repeats of the \( N = 2 \) paths \((2, 4, 3)\) and \((2, 6, 3)\). The existence of repeated paths at higher values of \( N \) further decreases the number of new paths that need to be considered for time domain scattering to \( 2, 3, 4, 6, 7, 9, 10, 12 \) for \( 0 \leq N \leq 7, \) respectively.

It should be noted that rays having near-grazing incidence on the core generate electromagnetic surface waves at the core/coating interface in the same way that rays having near-grazing incidence on the coating generate surface waves at the coating/exterior interface. In \([40, 44]\) it was shown that the dominant coating/exterior surface wave scattered field has the approximate attenuation factor \( \exp[-(m_{3} ka_{23})^{1/3} 3^{1/2} \sqrt{\pi (\theta - \theta_{c})}/2^{1/3}], \) where \( \theta_{c} \) is the critical scattering angle.
and $-X$ is the first zero of the Airy function, with the numerical value $X = 2.3381$. A similar derivation, expanding the Debye series terms for partial waves in the edge region in terms of Airy functions, converting the sum over partial waves into a modified Fock function, and evaluating it via contour integration, gives the result that the dominant core/coating surface wave scattered field has the attenuation factor $\exp[-(m_2 k_0 \lambda_2)^{1/3} X(\theta - \theta_0)/2^{1/3}]$.

In order to determine the degeneracy factor of all Debye terms for arbitrary $N$, Eq. (7) was expanded in powers of $R_n^{232}$ and was combined with Eq. (9). The results were then organized into differing total numbers of internal reflections under the constraint of Eq. (9). For rays that are transmitted into the coating, the degeneracy of each $(N, A, B)$ Debye term in the resulting collection was found to be the product of two factors. The first factor was the binomial coefficient for the powers of $R_n^{232}$ and $R_n^{212}$ appearing in the $(N, A, B)$ term. The second factor was the total number of ways of organizing the remaining $R_n^{121}$ factors between the $R_n^{232}$ and $R_n^{212}$ factors. The results are

$$N^{232} = (A - 2)/2.$$  \hfill (15a)

$$N^{121} = (N + 1 - A + B)/2.$$  \hfill (15b)

$$N^{212} = (N + 1 - B)/2.$$  \hfill (15c)

![Fig. 4](image-url) (Color online) Ray path (1, 4, 2) is shown in green and the repeated path (3, 4, 2) is shown in blue.

### Table 1. Debye Series Terms $(N, A, B)$ for $N \leq 7$

<table>
<thead>
<tr>
<th>$(N, A, B)$</th>
<th>$N^{232}$</th>
<th>$N^{212}$</th>
<th>$N^{121}$</th>
<th>$D$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(0, 0, 0)</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>(0, 2, 1)</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>(1, 2, 0)</td>
<td>0</td>
<td>1</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>(1, 2, 2)</td>
<td>0</td>
<td>0</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>(1, 4, 2)</td>
<td>1</td>
<td>0</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>(2, 2, 3)</td>
<td>0</td>
<td>0</td>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>(2, 4, 1)</td>
<td>1</td>
<td>1</td>
<td>0</td>
<td>2</td>
</tr>
<tr>
<td>(2, 4, 3)</td>
<td>1</td>
<td>0</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>(2, 6, 3)</td>
<td>2</td>
<td>0</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>(3, 2, 4)</td>
<td>0</td>
<td>0</td>
<td>3</td>
<td>1</td>
</tr>
<tr>
<td>(3, 4, 0)</td>
<td>1</td>
<td>2</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>(3, 4, 2)</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>(3, 4, 4)</td>
<td>1</td>
<td>0</td>
<td>2</td>
<td>3</td>
</tr>
<tr>
<td>(3, 6, 2)</td>
<td>2</td>
<td>1</td>
<td>0</td>
<td>3</td>
</tr>
<tr>
<td>(3, 6, 4)</td>
<td>2</td>
<td>0</td>
<td>1</td>
<td>3</td>
</tr>
<tr>
<td>(3, 8, 4)</td>
<td>3</td>
<td>0</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>(4, 2, 5)</td>
<td>0</td>
<td>0</td>
<td>4</td>
<td>1</td>
</tr>
<tr>
<td>(4, 4, 3)</td>
<td>1</td>
<td>1</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>(4, 4, 5)</td>
<td>1</td>
<td>0</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>(4, 6, 1)</td>
<td>2</td>
<td>2</td>
<td>0</td>
<td>3</td>
</tr>
<tr>
<td>(4, 6, 3)</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>4</td>
</tr>
<tr>
<td>(4, 6, 5)</td>
<td>2</td>
<td>0</td>
<td>2</td>
<td>6</td>
</tr>
<tr>
<td>(4, 8, 3)</td>
<td>3</td>
<td>1</td>
<td>0</td>
<td>4</td>
</tr>
<tr>
<td>(4, 8, 5)</td>
<td>3</td>
<td>0</td>
<td>1</td>
<td>4</td>
</tr>
<tr>
<td>(4, 10, 5)</td>
<td>4</td>
<td>0</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>(5, 2, 6)</td>
<td>0</td>
<td>0</td>
<td>5</td>
<td>1</td>
</tr>
<tr>
<td>(5, 4, 4)</td>
<td>1</td>
<td>1</td>
<td>3</td>
<td>2</td>
</tr>
<tr>
<td>(5, 4, 6)</td>
<td>1</td>
<td>0</td>
<td>4</td>
<td>5</td>
</tr>
<tr>
<td>(5, 6, 0)</td>
<td>2</td>
<td>3</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>(5, 6, 2)</td>
<td>2</td>
<td>2</td>
<td>1</td>
<td>3</td>
</tr>
<tr>
<td>(5, 6, 4)</td>
<td>2</td>
<td>1</td>
<td>2</td>
<td>9</td>
</tr>
<tr>
<td>(5, 6, 6)</td>
<td>2</td>
<td>0</td>
<td>3</td>
<td>10</td>
</tr>
<tr>
<td>(5, 8, 2)</td>
<td>3</td>
<td>2</td>
<td>0</td>
<td>6</td>
</tr>
<tr>
<td>(5, 8, 4)</td>
<td>3</td>
<td>1</td>
<td>1</td>
<td>12</td>
</tr>
<tr>
<td>(5, 8, 6)</td>
<td>3</td>
<td>0</td>
<td>2</td>
<td>10</td>
</tr>
<tr>
<td>(5, 10, 4)</td>
<td>4</td>
<td>1</td>
<td>0</td>
<td>5</td>
</tr>
<tr>
<td>(5, 10, 6)</td>
<td>4</td>
<td>0</td>
<td>1</td>
<td>5</td>
</tr>
<tr>
<td>(5, 12, 6)</td>
<td>5</td>
<td>0</td>
<td>0</td>
<td>1</td>
</tr>
<tr>
<td>(6, 2, 7)</td>
<td>0</td>
<td>0</td>
<td>6</td>
<td>1</td>
</tr>
<tr>
<td>(6, 4, 5)</td>
<td>1</td>
<td>1</td>
<td>4</td>
<td>2</td>
</tr>
<tr>
<td>(6, 4, 7)</td>
<td>1</td>
<td>0</td>
<td>5</td>
<td>6</td>
</tr>
<tr>
<td>(6, 6, 3)</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>3</td>
</tr>
<tr>
<td>(6, 6, 5)</td>
<td>2</td>
<td>1</td>
<td>3</td>
<td>12</td>
</tr>
<tr>
<td>(6, 6, 7)</td>
<td>2</td>
<td>0</td>
<td>4</td>
<td>15</td>
</tr>
<tr>
<td>(6, 8, 1)</td>
<td>3</td>
<td>3</td>
<td>0</td>
<td>4</td>
</tr>
<tr>
<td>(6, 8, 3)</td>
<td>3</td>
<td>2</td>
<td>1</td>
<td>12</td>
</tr>
<tr>
<td>(6, 8, 5)</td>
<td>3</td>
<td>1</td>
<td>2</td>
<td>24</td>
</tr>
<tr>
<td>(6, 8, 7)</td>
<td>3</td>
<td>0</td>
<td>3</td>
<td>20</td>
</tr>
<tr>
<td>(6, 10, 3)</td>
<td>4</td>
<td>2</td>
<td>0</td>
<td>10</td>
</tr>
<tr>
<td>(6, 10, 5)</td>
<td>4</td>
<td>1</td>
<td>1</td>
<td>20</td>
</tr>
<tr>
<td>(6, 10, 7)</td>
<td>4</td>
<td>0</td>
<td>2</td>
<td>15</td>
</tr>
<tr>
<td>(6, 12, 5)</td>
<td>5</td>
<td>1</td>
<td>0</td>
<td>6</td>
</tr>
<tr>
<td>(6, 12, 7)</td>
<td>5</td>
<td>0</td>
<td>1</td>
<td>6</td>
</tr>
<tr>
<td>(6, 14, 7)</td>
<td>6</td>
<td>0</td>
<td>0</td>
<td>7</td>
</tr>
<tr>
<td>(7, 2, 8)</td>
<td>0</td>
<td>0</td>
<td>7</td>
<td>1</td>
</tr>
<tr>
<td>(7, 4, 6)</td>
<td>1</td>
<td>1</td>
<td>5</td>
<td>2</td>
</tr>
<tr>
<td>(7, 4, 8)</td>
<td>1</td>
<td>0</td>
<td>6</td>
<td>7</td>
</tr>
<tr>
<td>(7, 6, 4)</td>
<td>2</td>
<td>2</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>(7, 6, 6)</td>
<td>2</td>
<td>1</td>
<td>4</td>
<td>15</td>
</tr>
<tr>
<td>(7, 6, 8)</td>
<td>2</td>
<td>0</td>
<td>5</td>
<td>21</td>
</tr>
<tr>
<td>(7, 8, 0)</td>
<td>3</td>
<td>4</td>
<td>0</td>
<td>1</td>
</tr>
</tbody>
</table>

*(Table continued)*
or

\[ A = 2N^{232} + 2, \]  
(16a)

\[ B = N + 1 - 2N^{212}. \]  
(16b)

The ray paths for time domain scattering occurring for a given \( N \) that are not repeats of paths that occurred for \( N - 2 \) are

\[ N^{232} = N - j, \quad N^{121} = j, \quad N^{212} = 0 \]  
(17)

for \( 0 \leq j \leq N \), and

\[ N^{232} = N - j, \quad N^{121} = 0, \quad N^{212} = j \]  
(18)

for \( 1 \leq j \leq N/2 \) when \( N \) is even and for \( 1 \leq j \leq (N + 1)/2 \) when \( N \) is odd. The reorganized Debye series of the partial wave scattering amplitudes is then

\[ a_n, b_n = \left( 1 - R_n^{232} - \sum_{N=0}^{\infty} T_n^{232} e^{N} T_n^{232} \right) / 2, \]  
(19)

where

\[ F_n^N = \sum_{N^{232}=0}^{N} \sum_{N^{121}=0}^{N} D(N, N^{232}, N^{121})(T_n^{212} T_n^{121} N^{232} + 1 - N^{212}) \times (R_n^{232})^{N^{232}} (R_n^{121})^{N^{121}} + G_n^N, \]  
(20)

\( N_{\text{max}} \) is the smaller of \( N^{232} \) and \( N - N^{232} \), and

\[ G_n^N = (R_n^{232})^{(N-1)/2}(R_n^{121})^{(N+1)/2} \]  
(21)

for odd \( N \).

The degeneracy factor \( D \) of the time domain paths is

\[ D(N, N^{232}, N^{212}) = (N^{232} + 1)! \times (N - 2N^{212})! / [(N^{212})!(N^{232} + 1 - N^{212})] \times (N - N^{232} - N^{212})! / [(N^{232} - N^{212})!]. \]  
(22)

The \( G_n^N \) term in Eqs. (20) and (21) for odd \( N \) is the Debye term \((N, N + 1, 0)\) having \( D = 1 \). This term describes rays reflecting back and forth within the coating a number of times without ever entering the core. For a large core and small impact parameter of the corresponding light rays, this term gives a time delayed optical echoing of the external reflection term \((0, 0, 0)\), which is known in the modeling of mirages as ducting [45]. For a small core and large ray impact parameter, it contains the \( p - 1 \) internal reflection rainbow for a homogeneous sphere made of coating material. It should be noted that since the degeneracy factor appears in the expression for the partial wave scattering amplitudes, the scattered intensity for each Debye term is proportional to \( D^2 \), i.e., it is a coherent, rather than an incoherent, enhancement [42,43].

Figure 5(a) compares the exact Aden–Kerker transverse-electric polarized scattered intensity from polydisperse coated spheres with median value of \( x_e = 2\pi n_e / \lambda = 600 \), variance \( v_e = 1/9 \), \( \sigma_{12'} \sigma_{22'} = 0.8 \), \( m_1 = 1.5 \) and \( m_2 = 1.33 \) (as in Fig. 2 of [26]). Figure 5(a) compares the results of Aden–Kerker calculations (shown in red) with the sum of the 16 Debye terms for \( N \leq 3 \); the blue curve has been calculated using the degeneracy factors \( D \) listed in Table 1, while the green curve has been calculated assuming that \( D = 1 \). The scattering contributions from individual terms of the Debye series terms in Fig. 5(b) take account of the fact that \( D = 2 \) for the \((2, 4, 1)\) and \((2, 4, 3)\) terms, whereas the results in Fig. 5(c) incorrectly assume that \( D = 1 \) for all of the terms.
(0, 0, 0), (0, 2, 1), (1, 2, 0), and (2, 4, 1) provide a background to
the $\alpha \beta$ rainbow of $(2, 4, 3)$ at $\theta = 100^\circ$, while Fig. 5(c) shows
that the $(2, 4, 3)$ term is no longer dominant at $\theta = 100^\circ$ when
degeneracy is ignored. Since ray scattering methods typically
perform an incoherent sum of all the ray theory paths, the co-
herent amplification of certain paths by their degeneracy
factors is not taken into account in ray calculations. The $\alpha \beta$
rainbow of the $(2, 4, 3)$ Debye term has a degeneracy of $D = 2$
and thus produces twice the scattered intensity as the inco-
herentely added intensity of the first $(2, 4, 3)$ path plus that of
the second $(2, 4, 3)$ path. This expected amplification factor of
2.0 is in good agreement with the $\sim 1.8$ amplification factor
measured in Figs. 2 and 4 of [26]. Similarly, the assumption
of $D = 1$ in Fig. 5(c) underestimates the contribution from
the $(2, 4, 1)$ term, resulting in the erroneous results shown
by the green curve in Fig. 5(a) when $105^\circ < \theta < 140^\circ$. These
results demonstrate that the coherent effects of path degen-
eracy cannot be ignored.

By way of comparison, coated sphere scattering is the sim-
plest case beyond scattering of a plane wave by a homo-
genous sphere (region 1) in an external medium (region 2).
Since only one type of internal reflection can occur for homo-
genous sphere scattering, its Debye series of the partial wave
scattering amplitudes is

\[ a_n, b_n = \left(1 - R_n^{212} - \sum_{N=0}^{\infty} T_n^N F_n^N T_n^2 \right) / 2, \]  

(23)

where

\[ F_n^N = D(N)(R_n^{21})^N \]  

(24)

and $D = 1$, while the reorganized Debye series for coated
sphere scattering is given in parallel form by the signifi-
cantly more complicated Eqs. (19)–(22).

The Debye series for scattering by a coated sphere is in
some sense intermediate between two limiting cases. For scat-
ering by a homogeneous sphere, it assumes a simple form
when written as Eqs. (23) and (24) in terms of single-scatter-
ing reflection and transmission amplitudes, whereas for scat-
ering by a sphere containing a large number of concentric
layers, the Debye series assumes the same simple form when
expressed in terms of multiple scattering reflection and trans-
mittance amplitudes that encompass many interfaces [38,
46–49]. Analogously to multiple scattering of waves from a
collection of target particles, expressions such as Eqs. (7)
and (8) are termed compact forms for the scattering amplitude
while expressions such as Eqs. (19)–(22) are termed ex-
panded forms [30] or order of scattering forms [31]. The
coated sphere problem has the unique status of allowing
the compact form to be tractable enough to permit the com-
plete cataloging and organization of the expanded form.

4. CONCLUSIONS

We found that the parameterization of the individual coated
sphere Debye terms by the total number of internal reflections
$N$, and the number of chords in the coating and core regions,$A$ and $B$, provides a simple and practical organization. This
organization leads in a natural way to both the degeneracy
of a number of the Debye terms and the repeat in time domain
scattering of some of the ray paths for $N$ internal reflections

that previously occurred for $N - 2$ internal reflections. These
two geometrical relationships greatly simplify the apparent
complexity of multiple internal reflection scattering within
the coated sphere, where for example the 335 Debye series
terms for $N = 7$ reduce down to 21 distinct terms and only
12 new ray paths for time domain scattering that are not re-
peats of paths that occurred for smaller $N$. The consequences
of this simplification will be studied in detail in [20].

REFERENCES

1. A. L. Aden and M. Kerker, “Scattering of electromagnetic waves from
functions for concentric spheres. Total scattering coefficients,
observation of rainbow scattering by a coated cylinder: twin primary
5. H. Hattori, H. Kakui, H. Kurniawan, and K. Kagawa, “Liquid re-
6. C. L. Adler, J. A. Lock, I. P. Rafferty, and W. Hickok, “Twin-
rainbow metrology. I. Measurement of the thickness of a thin
7. A. B. Pluchino, “Surface waves and the radiative properties of
8. R. Bhandari, “Tiny core or thin layer as a perturbation in scat-
9. R. L. Hightower and C. B. Richardson, “Resonant Mie scat-
10. J. A. Lock, “Interference enhancement of the internal fields at
structural scattering resonances of a coated sphere,” Appl.
11. T. Kaiser, S. Lange, and G. Schweiger, “Structural resonances in
a coated sphere: investigation of the volume-averaged source
12. T. M. Bambino and L. G. Guimarães, “Resonances of a coated
13. T. M. Bambino, A. M. S. Breitschaft, V. C. Barbosa, and L. G.
Guimarães, “Application of semiclassical and geometrical optics
14. G. W. Kattawar and D. A. Hood, “Electromagnetic scat-
ering from a spherical polydispersion of coated spheres,” Appl.
15. O. B. Toon and T. P. Ackerman, “Algorithms for the calculation
16. C. F. Bohren and D. R. Huffman, “Coated sphere” in Absorption
and Scattering of Light by Small Particles (Wiley-Interscience,
17. T. Kaiser and G. Schweiger, “Stable algorithm for the computa-
tion of Mie coefficients for scattered and transmitted fields of a
of light scattering by a coated sphere,” China Particul. 5,
by a coated sphere illuminated with a Gaussian beam,” Appl.
20. F. Onofri, G. Grehan, and G. Gouesbet, “Electromagnetic scat-
ering from a multilayered sphere located in an arbitrary beam,”
21. Z. S. Wu, L. X. Guo, K. F. Ren, G. Gouesbet, and G. Grehan,
“Improved algorithm for electromagnetic scattering of plane


