

# Understanding light scattering by a coated sphere

## Part 1: Theoretical considerations

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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

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### 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  $p \geq 1$  [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. This

fact explains some of the discrepancies between ray theory and Aden–Kerker scattering obtained in [26]. 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  $k = 2\pi/\lambda$ , traveling in the positive  $z$  direction in an exterior medium (region 3) 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

$$N_n^{12} = \alpha\psi_n(m_2ka_{12})\psi'_n(m_1ka_{12}) - \beta\psi'_n(m_2ka_{12})\psi_n(m_1ka_{12}) \quad (1a)$$

$$D_n^{12} = \alpha\chi_n(m_2ka_{12})\psi'_n(m_1ka_{12}) - \beta\chi'_n(m_2ka_{12})\psi_n(m_1ka_{12}) \quad (1b)$$

$$N_n^{23} = \gamma\psi_n(m_3ka_{23})\psi'_n(m_2ka_{23}) - \delta\psi'_n(m_3ka_{23})\psi_n(m_2ka_{23}) \quad (1c)$$

$$D_n^{23} = \gamma\chi_n(m_3ka_{23})\psi'_n(m_2ka_{23}) - \delta\chi'_n(m_3ka_{23})\psi_n(m_2ka_{23}) \quad (1d)$$

$$P_n^{23} = \gamma\psi_n(m_3ka_{23})\chi'_n(m_2ka_{23}) - \delta\psi'_n(m_3ka_{23})\chi_n(m_2ka_{23}) \quad (1e)$$

$$Q_n^{23} = \gamma\chi_n(m_3ka_{23})\chi'_n(m_2ka_{23}) - \delta\chi'_n(m_3ka_{23})\chi_n(m_2ka_{23}), \quad (1f)$$

where  $\psi_n$  are Riccati-Bessel functions and  $\chi_n$  are Riccati-Neumann functions in the notation of [36],  $\alpha = m_1$ ,  $\beta = m_2$ ,  $\gamma = m_2$ , 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]

$$a_n, b_n = (D_n^{12}N_n^{23} - N_n^{12}P_n^{23}) / [(D_n^{12}N_n^{23} - N_n^{12}P_n^{23}) + i(D_n^{12}D_n^{23} - N_n^{12}Q_n^{23})]. \quad (2)$$

The scattered intensity for unpolarized incident light is

$$I(r, \theta) = [E_0^2 / (2\mu_0ck^2r^2)] [|S_1(\theta)|^2 + |S_2(\theta)|^2], \quad (3)$$

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

$$S_1(\theta) = \sum_{n=1}^{\infty} \{(2n+1)/[n(n+1)]\} [a_n\pi_n(\theta) + b_n\tau_n(\theta)] \quad (4a)$$

$$S_2(\theta) = \sum_{n=1}^{\infty} \{(2n+1)/[n(n+1)]\} [a_n\tau_n(\theta) + b_n\pi_n(\theta)], \quad (4b)$$

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

$$P_n^{12} = \alpha\psi_n(m_2ka_{12})\chi'_n(m_1ka_{12}) - \beta\psi'_n(m_2ka_{12})\chi_n(m_1ka_{12}) \quad (5a)$$

$$Q_n^{12} = \alpha\chi_n(m_2ka_{12})\chi'_n(m_1ka_{12}) - \beta\chi'_n(m_2ka_{12})\chi_n(m_1ka_{12}), \quad (5b)$$

as well as the expressions for the partial wave Fresnel reflection and transmission coefficients at each of the two interfaces. The quantities  $R_n^{323}$  and  $T_n^{32}$  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,  $R_n^{232}$  and  $T_n^{23}$  are the Fresnel coefficients for a radially outgoing spherical multipole wave in region 2 incident on the coating/exterior interface,  $R_n^{212}$  and  $T_n^{21}$  are the Fresnel coefficients for a radially incoming spherical multipole wave in region 2 incident on the core/coating interface, and  $R_n^{121}$  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

$$R_n^{323} = (-N_n^{23} + Q_n^{23} + iD_n^{23} + iP_n^{23}) / (N_n^{23} + Q_n^{23} + iD_n^{23} - iP_n^{23}) \quad (6a)$$

$$R_n^{232} = (-N_n^{23} + Q_n^{23} - iD_n^{23} - iP_n^{23}) / (N_n^{23} + Q_n^{23} + iD_n^{23} - iP_n^{23}) \quad (6b)$$

$$T_n^{32} = -2im_3/(N_n^{23} + Q_n^{23} + iD_n^{23} - iP_n^{23}) \quad (6c)$$

$$T_n^{23} = -2im_2/(N_n^{23} + Q_n^{23} + iD_n^{23} - iP_n^{23}) \quad (6d)$$

$$R_n^{212} = (-N_n^{12} + Q_n^{12} + iD_n^{12} + iP_n^{12})/(N_n^{12} + Q_n^{12} + iD_n^{12} - iP_n^{12}) \quad (6e)$$

$$R_n^{121} = (-N_n^{12} + Q_n^{12} - iD_n^{12} - iP_n^{12})/(N_n^{12} + Q_n^{12} + iD_n^{12} - iP_n^{12}) \quad (6f)$$

$$T_n^{21} = -2im_2/(N_n^{12} + Q_n^{12} + iD_n^{12} - iP_n^{12}) \quad (6g)$$

$$T_n^{12} = -2im_1/(N_n^{12} + Q_n^{12} + iD_n^{12} - iP_n^{12}). \quad (6h)$$

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

$$\begin{aligned} a_n, b_n &= [1 - R_n^{323} - T_n^{32} W_n T_n^{23} / (1 - W_n R_n^{232})] / 2 \\ &= \left[ 1 - R_n^{323} - \sum_{q=1}^{\infty} T_n^{23} (W_n R_n^{232})^{q-1} W_n T_n^{23} \right] / 2, \end{aligned} \quad (7)$$

where

$$W_n = R_n^{212} + T_n^{21} T_n^{12} / (1 - R_n^{121}) = R_n^{212} + \sum_{p=1}^{\infty} T_n^{21} (R_n^{121})^{p-1} T_n^{12}. \quad (8)$$

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  $R_n^{212}$ ,  $N^{121}$  of the reflections  $R_n^{121}$ , and  $N^{232}$  of the reflections  $R_n^{232}$  with the constraint

$$N^{212} + N^{121} + N^{232} = N. \quad (9)$$

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

$$(a_n, b_n)_{N=0} = (1 - R_n^{323} - T_n^{32} T_n^{21} T_n^{12} T_n^{23}) / 2. \quad (10)$$

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 [32] 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/external interface is large as for ray path B in Fig. 1, it can be transmitted into the coating, miss striking the core, and be

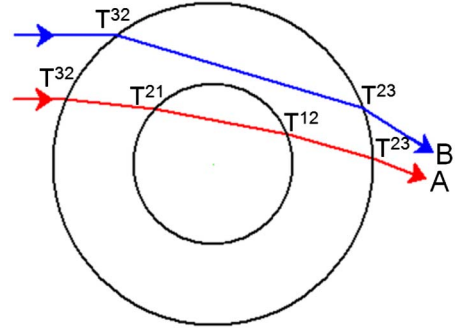


Fig. 1. (Color online) Ray path A passes through the core, whereas ray B misses the core.

transmitted back out again. But the term  $T_n^{32} T_n^{23}$  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 or be transmitted into and out of the core before it becomes a radially outgoing wave that can be transmitted from region 2 back into region 3. The situation described above is described rather by the  $N = 1$  term  $T_n^{32} R_n^{212} T_n^{23}$  in Eq. (11) below, where the partial waves  $n > ka_{12}$  corresponding to the incident rays that miss the core still interact with it via tunneling external reflection with the amplitude  $R_n^{212} \rightarrow 1$  [40]. But then it would seem in analogy to quantum electrodynamics that the set of ladder terms [41]  $T_n^{32} R_n^{212} R_n^{212} T_n^{23}$ ,  $T_n^{23} R_n^{212} R_n^{121} R_n^{212} T_n^{23}$ , etc. should also describe this situation. These terms with the additional adjacent factors of  $R_n^{121}$  are not present in the Debye series of Eqs. (7) and (8). Again this is because radially incoming and outgoing spherical multipole waves are being described. Such a wave initially in region 2 and reflected by the core/coating interface must next be reflected by the coating/external interface before it can be reflected by 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

$$\begin{aligned} (a_n, b_n)_{N=1} &= -T_n^{32} (R_n^{212} + T_n^{21} R_n^{121} T_n^{12} \\ &\quad + T_n^{21} T_n^{12} R_n^{232} T_n^{21} T_n^{12}) T_n^{23} / 2. \end{aligned} \quad (11)$$

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/external 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/external 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  $T_n^{32} R_n^{232} T_n^{23}$ , 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

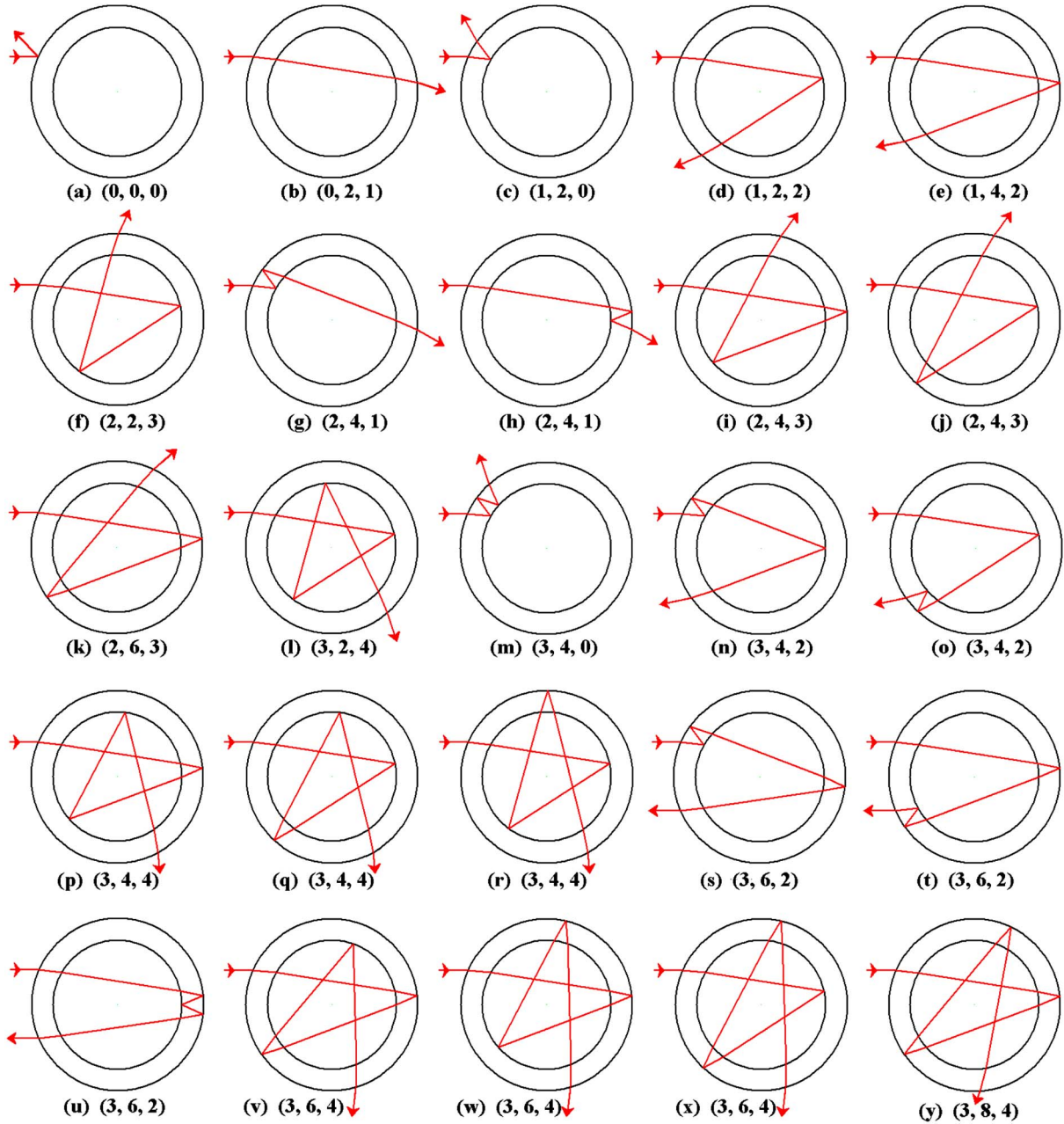


Fig. 2. (Color online) Ray paths for  $N \leq 3$  internal reflections showing the  $(N, A, B)$  values, where  $A$  is the number of chords in the coating and  $B$  is the number of chords in the core.

one-internal-reflection rainbow of a homogeneous sphere is now contained in the  $N = 3$  Debye term  $T_n^{32} R_n^{212} R_n^{232} R_n^{212} T_n^{23}$  via tunneling external reflection of large partial waves with the amplitude  $R_n^{212} \rightarrow 1$  both before and after the internal reflection at the coating/external interface.

In like manner, the  $N = 2$  portion of Eqs. (7) and (8) is

$$\begin{aligned}
 (a_n, b_n)_{N=2} = & -T_n^{32} (T_n^{21} R_n^{121} R_n^{121} T_n^{12} + R_n^{212} R_n^{232} T_n^{21} T_n^{12} \\
 & + T_n^{21} T_n^{12} R_n^{232} R_n^{212} + T_n^{21} T_n^{12} R_n^{232} T_n^{21} R_n^{121} T_n^{12} \\
 & + T_n^{21} R_n^{121} T_n^{12} R_n^{232} T_n^{21} T_n^{12} \\
 & + T_n^{21} T_n^{12} R_n^{232} T_n^{21} T_n^{12} R_n^{232} T_n^{21} T_n^{12}) T_n^{23}/2. \quad (12)
 \end{aligned}$$

The terms in Eq. (12) correspond to the ray paths shown in Fig. 2(f)–2(k), respectively.

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



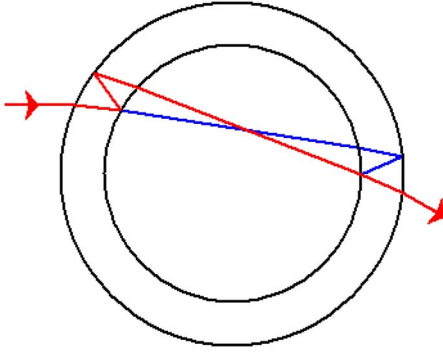


Fig. 3. (Color online) Two degenerate (2, 4, 1) ray paths.

$N = 2$  the two Debye terms  $T_n^{32} R_n^{212} R_n^{232} T_n^{21} T_n^{12} T_n^{23}$  and  $T_n^{32} T_n^{21} T_n^{12} R_n^{232} R_n^{212} T_n^{23}$  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  $\theta$ . 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  $\theta_{3i}$  of an incoming ray and the optical path length of the scattered rays as a function of  $\theta_{3i}$  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  $T_n^{32} T_n^{21} R_n^{121} T_n^{12} R_n^{232} T_n^{21} T_n^{12} T_n^{23}$  and  $T_n^{32} T_n^{21} T_n^{12} R_n^{232} T_n^{21} R_n^{121} T_n^{12} T_n^{23}$ , 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  $\theta_{2t}$ ,  $\theta_{2i}$ , and  $\theta_{1t}$  be the transmitted angle of a ray from region 3 into region 2 at the coating/ exterior 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

$$m_3 \sin(\theta_{3i}) = m_2 \sin(\theta_{2t}) \quad (13a)$$

$$m_2 \sin(\theta_{2i}) = m_1 \sin(\theta_{1t}), \quad (13b)$$

and the coated sphere geometry gives

$$a_{23} \sin(\theta_{2t}) = a_{12} \sin(\theta_{2i}). \quad (14)$$

As a corresponding ray propagates through the coated sphere, each  $T_n^{32}$  and  $T_n^{23}$  factor contributes a deflection angle of  $\theta_{3i} - \theta_{2t}$ , each  $T_n^{21}$  and  $T_n^{12}$  contributes  $\theta_{2i} - \theta_{1t}$ , each  $R_n^{232}$

contributes  $\pi - 2\theta_{2t}$ , each  $R_n^{121}$  contributes  $\pi - 2\theta_{1t}$ , and each  $R_n^{212}$  contributes  $-\pi + 2\theta_{2i}$ . 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  $m_3 a_{23} [1 - \cos(\theta_{3i})]$ , the optical length of each chord in the coating is  $m_2 [a_{23} \cos(\theta_{2t}) - a_{12} \cos(\theta_{2i})]$ , and the optical length of each chord in the core is  $2m_1 a_{12} \cos(\theta_{1t})$ . 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  $T_n^{32} T_n^{21} R_n^{121} R_n^{121} T_n^{12} R_n^{232} T_n^{21} T_n^{12} T_n^{23}$ . 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  $T_n^{32} T_n^{21} T_n^{12} R_n^{232} T_n^{21} T_n^{12} T_n^{23}$  while the two degenerate paths (3, 4, 2) are  $T_n^{32} R_n^{212} R_n^{232} T_n^{21} R_n^{121} T_n^{12} T_n^{23}$  and  $T_n^{32} T_n^{21} R_n^{121} T_n^{12} R_n^{232} R_n^{212} T_n^{23}$ . In addition to the partial wave Fresnel coefficients common to both terms, the (1, 4, 2) term has an extra  $T_n^{21} T_n^{12}$  while the (3, 4, 2) terms have a extra  $R_n^{212} R_n^{121}$ . According to the rules given above, both of these extra factors have the same deflection angle  $2\theta_{2i} - 2\theta_{1t}$  of the corresponding light rays. Thus the (1, 4, 2) and (3, 4, 2) rays have the same scattering angle as a function of  $\theta_{3i}$ . They both have a rainbow at exactly the same scattering angle, and they both have the same glory structure for  $\theta \approx 180^\circ$ . Since the optical path length as a function of  $\theta_{3i}$  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  $T_n^{21} T_n^{12}$  factor in (1, 4, 2) and the extra  $R_n^{212} R_n^{121}$  factor in (3, 4, 2). The extra  $T_n^{21} T_n^{12}$  factor contributes more strongly to the scattered field for small partial waves corresponding to near-paraxial rays, while the extra  $R_n^{212} R_n^{121}$  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  $\exp[-(m_3 k a_{23})^{1/3} 3^{1/2} X(\theta - \theta_c) / 2^{4/3}]$ , where  $\theta_c$  is the critical scattering angle

**Table 1. Debye Series Terms ( $N, A, B$ ) for  $N \leq 7$  Internal Reflections Showing the Number of Internal Reflections  $N^{232}$ ,  $N^{212}$  and  $N^{121}$ , and the Degeneracy Factor  $D^a$**

$(N, A, B)$	$N^{232}$	$N^{212}$	$N^{121}$	$D$
(0, 0, 0)	0	0	0	1
(0, 2, 1)	0	0	0	1
(1, 2, 0)	0	1	0	1
(1, 2, 2)	0	0	1	1
(1, 4, 2)	1	0	0	1
(2, 2, 3)	0	0	2	1
(2, 4, 1)	1	1	0	2
(2, 4, 3)	1	0	1	2
(2, 6, 3)	2	0	0	1
(3, 2, 4)	0	0	3	1
(3, 4, 0)	1	2	0	1
(3, 4, 2)*	1	1	1	2
(3, 4, 4)	1	0	2	3
(3, 6, 2)	2	1	0	3
(3, 6, 4)	2	0	1	3
(3, 8, 4)	3	0	0	1
(4, 2, 5)	0	0	4	1
(4, 4, 3)*	1	1	2	2
(4, 4, 5)	1	0	3	4
(4, 6, 1)	2	2	0	3
(4, 6, 3)*	2	1	1	6
(4, 6, 5)	2	0	2	6
(4, 8, 3)	3	1	0	4
(4, 8, 5)	3	0	1	4
(4, 10, 5)	4	0	0	1
(5, 2, 6)	0	0	5	1
(5, 4, 4)*	1	1	3	2
(5, 4, 6)	1	0	4	5
(5, 6, 0)	2	3	0	1
(5, 6, 2)*	2	2	1	3
(5, 6, 4)*	2	1	2	9
(5, 6, 6)	2	0	3	10
(5, 8, 2)	3	2	0	6
(5, 8, 4)*	3	1	1	12
(5, 8, 6)	3	0	2	10
(5, 10, 4)	4	1	0	5
(5, 10, 6)	4	0	1	5
(5, 12, 6)	5	0	0	1
(6, 2, 7)	0	0	6	1
(6, 4, 5)*	1	1	4	2
(6, 4, 7)	1	0	5	6
(6, 6, 3)*	2	2	2	3
(6, 6, 5)*	2	1	3	12
(6, 6, 7)	2	0	4	15
(6, 8, 1)	3	3	0	4
(6, 8, 3)*	3	2	1	12
(6, 8, 5)*	3	1	2	24
(6, 8, 7)	3	0	3	20
(6, 10, 3)	4	2	0	10
(6, 10, 5)*	4	1	1	20
(6, 10, 7)	4	0	2	15
(6, 12, 5)	5	1	0	6
(6, 12, 7)	5	0	1	6
(6, 14, 7)	6	0	0	1
(7, 2, 8)	0	0	7	1
(7, 4, 6)*	1	1	5	2
(7, 4, 8)	1	0	6	7
(7, 6, 4)*	2	2	3	3
(7, 6, 6)*	2	1	4	15
(7, 6, 8)	2	0	5	21
(7, 8, 0)	3	4	0	1

(Table continued)

**Table 1. (Continued)**

$(N, A, B)$	$N^{232}$	$N^{212}$	$N^{121}$	$D$
(7, 8, 2)*	3	3	1	4
(7, 8, 4)*	3	2	2	18
(7, 8, 6)*	3	1	3	40
(7, 8, 8)	3	0	4	35
(7, 10, 2)	4	3	0	10
(7, 10, 4)*	4	2	1	30
(7, 10, 6)*	4	1	2	50
(7, 10, 8)	4	0	3	35
(7, 12, 4)	5	2	0	15
(7, 12, 6)*	5	1	1	30
(7, 12, 8)	5	0	2	21
(7, 14, 6)	6	1	0	7
(7, 14, 8)	6	0	1	7
(7, 16, 8)	7	0	0	1

<sup>a</sup>The terms marked \* correspond to repeated paths of terms with  $(N - 2, A, B)$ .

and  $-X$  is the first zero of the Airy function, with the numerical value  $X \approx 2.3381$ . 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  $\exp[-(m_2ka_{12})^{1/3}3^{1/2}X(\theta - \theta_c)/2^{4/3}]$ .

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. (8). 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 terms 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

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

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

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

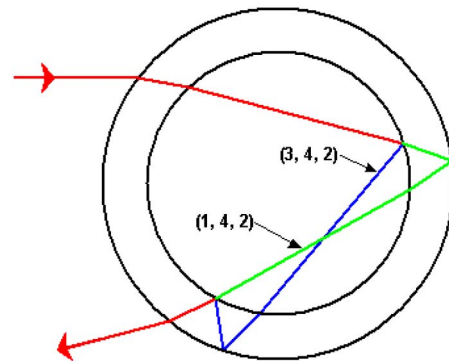


Fig. 4. (Color online) Ray path (1, 4, 2) is shown in green and the repeated path (3, 4, 2) is shown in blue.

or

$$A = 2N^{232} + 2, \tag{16a}$$

$$B = N + 1 - 2N^{212}. \tag{16b}$$

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

$$N^{232} = N - j, \quad N^{121} = j, \quad N^{212} = 0 \tag{17}$$

for  $0 \leq j \leq N$ , and

$$N^{232} = N - j, \quad N^{121} = 0, \quad N^{212} = j \tag{18}$$

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

$$a_n, b_n = \left( 1 - F_n^{323} - \sum_{N=0}^{\infty} T_n^{32} F_n^N T_n^{23} \right) / 2, \tag{19}$$

where

$$F_n^N = \sum_{N^{232}=0}^N \sum_{N^{212}=0}^{N_{\max}} D(N, N^{232}, N^{212}) (T_n^{21} T_n^{12})^{N^{232}+1-N^{212}} \times (R_n^{232})^{N^{232}} (R_n^{212})^{N^{212}} (R_n^{121})^{N^{121}} + G_n^N, \tag{20}$$

$N_{\max}$  is the smaller of  $N^{232}$  and  $N - N^{232}$ , and

$$G_n^N = (R_n^{232})^{(N-1)/2} (R_n^{212})^{(N+1)/2} \tag{21}$$

for odd  $N$ .

The degeneracy factor  $D$  of the time domain paths is

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

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 as a function of  $\theta$  with the sum of the 16 Debye series terms for  $N \leq 3$  taking account of the degeneracy factors  $D$  listed in Table 1. The agreement is very good, bearing in mind that we have neglected the Debye terms with  $N > 3$ . However, when degeneracy is ignored

(i.e., by setting  $D = 1$  for all Debye terms), the comparison is much worse—especially for  $90^\circ < \theta < 140^\circ$ . The parameters used to produce Fig. 5 (i.e.  $m_1 = 1.5$ ,  $m_2 = 1.33$ , and  $a_{12}/a_{23} = 0.8$ ) are similar to those in Fig. 2 of [26], which compared the results of ray tracing computations of coated sphere scattering with the Aden–Kerker scattered intensity. As in Fig. 5(a), the comparison in [26] was generally good except that the Aden–Kerker unpolarized scattered intensity was a factor of  $\sim 1.8$  higher than the ray tracing unpolarized intensity at the position of the  $\alpha\beta$  rainbow [29] at  $\theta \approx 100^\circ$ , and it was a factor of  $\sim 1.4$  higher than the ray tracing intensity for  $100^\circ < \theta < 140^\circ$  in the relatively dark region between the  $\alpha\beta$  and  $\alpha$  rainbows. The authors of [26] attributed these discrepancies to possible inaccuracies in the Aden–Kerker wave scattering computer program. Figure 5(b) shows that the Debye terms

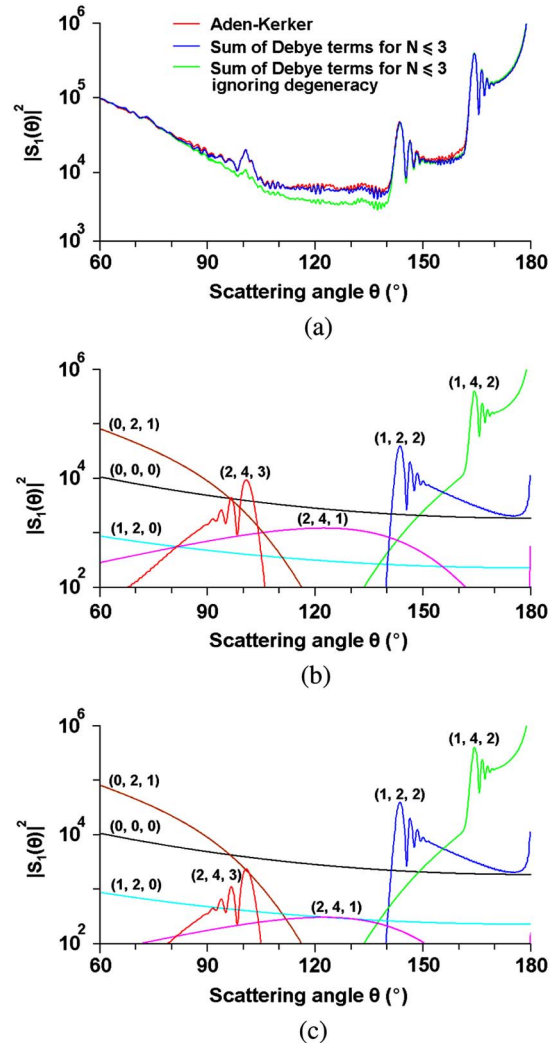


Fig. 5. (Color online) Transverse-electric polarized scattering from polydisperse coated spheres with median value of  $x_e = 2\pi a_{23}/\lambda = 600$ , variance  $v_e = 1/9$ ,  $a_{12}/a_{23} = 0.8$ ,  $m_1 = 1.5$  and  $m_2 = 1.33$  (as in Fig. 2 of [26]). Figure 5(a) compares the results of Aden–Kerker 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  $\theta \approx 100^\circ$ , while Fig. 5(c) shows that the (2, 4, 3) term is no longer dominant at  $\theta \approx 100^\circ$  when degeneracy is ignored. Since ray scattering methods typically perform an incoherent sum of all the ray theory paths, the coherent 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 incoherently 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 degeneracy cannot be ignored.

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

$$a_n, b_n = \left( 1 - R_n^{212} - \sum_{N=0}^{\infty} T_n^{21} F_n^N T_n^{12} \right) / 2, \quad (23)$$

where

$$F_n^N = D(N)(R_n^{121})^N \quad (24)$$

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

The Debye series for scattering by a coated sphere is in some sense intermediate between two limiting cases. For scattering by a homogeneous sphere, it assumes a simple form when written as Eqs. (23) and (24) in terms of single-scattering reflection and transmission amplitudes, whereas for scattering 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 transmission 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 expanded 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 complete 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 353 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 repeats of paths that occurred for smaller  $N$ . The consequences of this simplification will be studied in detail in [29].

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