Application of Gegenbauer analysis to light scattering from spheres: Theory

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The Mie and Rayleigh-Debye theories of homogeneous spheres are reformulated in terms of Gegenbauer functions to allow a comparison between the two treatments in the Rayleigh-Debye limit. This leads to reduced Mie equations that are valid for all particle sizes, whereas the Rayleigh-Debye equations are restricted to an outer size parameter $\alpha < 1$. The application of Gegenbauer analysis of experimental patterns is discussed and the possibility of pattern inversion is examined.

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I. INTRODUCTION

Two fundamental challenges exist in the study of light scattering from particulate matter. These are classified in the following ways.

(a) The direct problem. How can the scattering pattern be calculated when the properties of the particle are known?

(b) The inverse problem. How may the characteristics of the particle be determined when the scattering pattern is known?

The first type of problem has led to a considerable body of theoretical work over the years, and has resulted in rigorous solutions being developed for a few particle geometries that include spheres [1], circular cylinders [2], elliptical cylinders [3], prolate spheroids, and oblate spheroids [4]. A general computational method for any geometry, using an expansion of vector spherical waves, has even been proposed [5], but doubts about the proper convergence of the solutions have since been raised [6]. Less rigorous solutions have also been derived based on the Rayleigh-Debye theory of scattering. These are valid when the refractive indices of the particle and the ambient medium are nearly equal [7,8]. The treatment can be applied to all particle shapes; however, as will be shown in the present paper, an upper limit exists for the particle dimensions.

No significant progress has been made on the inverse problem for wide angle scattering. Since this case is of prime importance to experimentalists, they have resorted to applying heuristic techniques for particle characterization. Examples of such procedures are (i) the use of sets of calibration particles to construct instrument response curves, obtained by collecting most or part of the scattered light; and (ii) the matching of experimental scattering to a stored theoretical data base. The former method clearly restricts applications to particles of the same material as the reference particles, and ignores the possibility of an oscillatory response at sizes between the reference set. The latter technique, while not suffering the above difficulties, requires extensive prior computation of patterns and large amounts of computer memory, and raises the question of how the best fit solutions are to be recognized.

This paper will attempt, in part, to rectify this serious omission from the body of knowledge by investigating a procedure which is capable of allowing partial, and probably complete, inversion of experimental scattering patterns of spheres. The technique introduced here is the Gegenbauer analysis of scattering patterns. Optimum conditions for the application of the method are achieved when nearly complete quasicontinuous scattering patterns from spheres are available over 180° in a fixed plane of detection. This detector arrangement is fairly standard for wide angle scattering instruments. Data are, however, missing near scattering angles of 0° and 180° in such systems at the positions of the entrance and exit apertures of the irradiating laser beam. A remarkable feature of the analysis is that a procedure exists which allows the missing data to be almost completely restored. The processed data then give an immediate measure of particle radius, and offer a possible strategy for determining the particle's refractive index. Clearly any analysis capable of exhibiting these features is worthy of study.

To illustrate the power of the technique, we will reformulate the Mie and Rayleigh-Debye theories of spheres in terms of Gegenbauer polynomials. A theoretical comparison between the two treatments is then possible when the Rayleigh-Debye limit is imposed on the Mie theory. The feasibility of inverting the experimental patterns for Rayleigh-Debye spheres will be illustrated, and the case for the more general spheres will be discussed.

II. GEGENBAUER ANALYSIS

Gegenbauer polynomials $T_n^\beta(z)$ of degree β and order n belong to a particular class of solution of the hypergeometric function $F(a,b|c|x)$, and may be defined by [9]

$$T_n^\beta(z)=\frac{\Gamma(n+2\beta+1)}{2^\beta n!\Gamma(\beta+1)}F\left[-n,n+2\beta+1|\beta+1|\frac{1-z}{2}\right], \quad (1)$$

where $\Gamma(x)$ represents the gamma function. The choice of $a=-n(n=0,1,2,\dots)$ ensures that the series terminates to generate a polynomial. In general, β is an arbitrary parameter, but our interest will be confined to positive integer values $\beta=k$, and in particular to $k=1$. The association of these functions with the light scattering patterns of spheres arises since the angular functions $\pi_n(\cos\vartheta)$ and $\tau_n(\cos\vartheta)$ appearing in the Mie theory are expandable in terms of Gegenbauer functions. However, unlike the $\pi_n(z)$ and $\tau_n(z)$ functions, Gegenbauer functions of a fixed degree constitute a complete orthogonal set with respect to a weight function $(1-z^2)^k$:

$$\int_{-1}^{1}(1-z^2)^k T_m^k(z) T_n^k(z) dz = \frac{2(n+2k)!}{(2n+2k+1)n!}\delta_{m,n} . \quad (2)$$

Other useful properties of Gegenbauer functions are given by Morse and Feshbach [9].

A consequence of the orthogonality and completeness is that any piecewise continuous function $F(z)$ of z in the range $-1\le z\le 1$ may be written as a Gegenbauer series. Hence

$$F(z)=\sum_{n=0}^{\infty} c_n T_n^k(z) , \quad (3)$$

where

$$c_n=\frac{(2n+2k+1)n!}{2(n+2k)!}\int_{-1}^{1}(1-z^2)^k F(z) T_n^k(z) dz . \quad (4)$$

These two relations may be regarded as a transform pair

$$c_n=G[F(z)] ,$$

$$F(z)=G^{-1}[c_n] ,$$

similar to that of discrete Fourier series. The calculation of c_n as a function of order is therefore taken as the definition of the discrete Gegenbauer transform of $F(z)$. The presentation of the coefficients c_n as a discrete Gegenbauer spectrum of $F(z)$ is then, in all respects, representative of the initial function.

As both the amplitude and irradiance functions of the scattering patterns can be represented by Gegenbauer series in $z=\cos\vartheta$, where ϑ is the scattering angle, we have the basis of a method for theoretical and experimental analysis.

III. NOTATION AND CONVENTIONS

The scattering configuration which will be considered is given in Fig. 1. This shows a spherical particle of refractive index m relative to the ambient medium, and a radius a which is irradiated by a collimated monochromatic beam traveling along the z axis. The incident beam is plane polarized in either the x or y directions

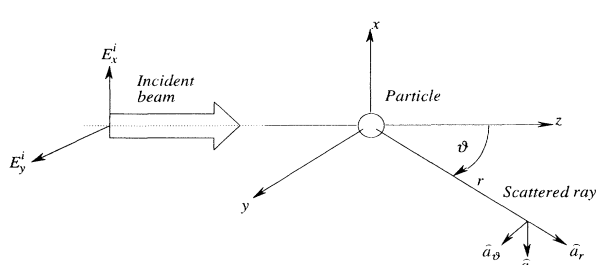

FIG. 1. Scattering geometry.

with electric fields

$$\mathbf{E}^i(\mathbf{r},t)=\begin{Bmatrix}\hat{\mathbf{a}}_x\\ \hat{\mathbf{a}}_y\end{Bmatrix}\times E_0 e^{i(kz-\omega t)} , \quad (5)$$

in which the propagation constant $k=2\pi/\lambda$ and wavelength λ are for the ambient medium. Scattering is assumed to be detected in the yz plane. Two solutions therefore exist for the scattered electric field at angle ϑ, corresponding to the two directions of incident polarization. For perpendicular polarization E_x^i the scattering amplitude is $S_\perp(\vartheta)$, while for parallel polarization E_y^i it is $S_\parallel(\vartheta)$. These are defined by specifying the scattered electric field at a distance r from the particle by

$$\mathbf{E}^s=-i\frac{e^{ikr}}{kr}E_0\times\begin{cases}\hat{\mathbf{a}}_\phi S_\perp(\vartheta)\\ -\hat{\mathbf{a}}_\vartheta S_\parallel(\vartheta) .\end{cases} \quad (6)$$

The associated irradiance functions are

$$H_p^s=\frac{H_0}{k^2r^2}|S_p(\vartheta)|^2, \quad p=\perp \text{ or } \parallel \quad (7)$$

for an incident beam of irradiance H_0.

When the specimen is a homogeneous sphere, the scattering functions depend on only two factors, the external and internal size parameters $\alpha=ka$ and $\beta=m\alpha$, respectively. Our analysis, however, indicates that it is convenient to introduce a third factor—the effective size parameter

$$\gamma=(m^2-1)\alpha/2 . \quad (8)$$

When the relative refractive index approaches unity, this parameter becomes equal to the difference of the optical phase shifts in the internal and external media when a distance a is traversed. The usual Rayleigh-Debye condition $2(m-1)\alpha\ll 1$ is therefore equivalent to $2\gamma\ll 1$. A specimen for which the condition is satisfied will be referred to as a Rayleigh-Debye (RD) sphere.

IV. MIE SCATTERING

Mie scattering is derived by the rigorous application of electromagnetic theory to spheres. For perpendicular and parallel polarization, the scattering amplitudes are [10]

$$S_\perp(z)=\sum_{n=1}^{\infty}\frac{(2n+1)}{n(n+1)}\{a_n\pi_n(z)+b_n\tau_n(z)\} , \quad (9a)$$

$$S_\parallel(z)=\sum_{n=1}^{\infty}\frac{(2n+1)}{n(n+1)}\{a_n\tau_n(z)+b_n\pi_n(z)\} , \quad (9b)$$

where a_n and b_n are the electric and magnetic multipole coefficients and $z=\cos\vartheta$. The angular functions $\pi_n(\cos\vartheta)$ and $\tau_n(\cos\vartheta)$ are defined in terms of the associated Legendre functions, but may be expressed as first degree Gegenbauer polynomials $T_n^1(z)$:

$$\pi_n(z)=\frac{P_n^1(z)}{(1-z^2)^{1/2}}=T_{n-1}^1(z) , \quad (10a)$$

$$\tau_n(z) = -(1-z^2)^{1/2}\frac{d}{dz}\{P_n^1(z)\}$$
$$= \frac{1}{(2n+1)}\{n^2 T_n^1(z) - (n+1)^2 T_{n-2}^1(z)\} . \tag{10b}$$

Substitution into the scattering functions then yields

$$S_p(z) = \sum_{n=0}^{\infty} (c_n)_p T_n^1(z) , \tag{11}$$

where

$$(c_n)_\perp = \frac{n}{n+1} b_n + \frac{2n+3}{(n+1)(n+2)} a_{n+1} - \frac{n+3}{n+2} b_{n+2} , \tag{12a}$$

$$(c_n)_\parallel = \frac{n}{n+1} a_n + \frac{2n+3}{(n+1)(n+2)} b_{n+1} - \frac{n+3}{n+2} a_{n+2} . \tag{12b}$$

Alternative expressions can be derived for the scattering functions in terms of Gegenbauer polynomials of degree other than $k=1$, but these lead to more complicated formulas for $(c_n)_p$. In the case of $k=0$, the Gegenbauer polynomials reduce to the case of Legendre polynomials $T_n^0(z) = P_n(z)$, and the scattering function becomes Legendre series by the substitution of

$$T_n^1(z) = \sum_{r=0}^{[n/2]} (2n+1-4r) T_{n-2r}^0(z)$$

into Eq. (10), while, for $k=2$,

$$T_n^1(z) = \frac{1}{(2n+3)}\{T_n^2(z) - T_{n-2}^2(z)\}$$

should be used.

The multipole coefficients are related to the particle properties through the relations

$$a_n = \tfrac{1}{2}\{1 - e^{i2u_n}\} = -ie^{iu_n}\sin u_n , \tag{13a}$$

$$b_n = \tfrac{1}{2}\{1 - e^{i2v_n}\} = -ie^{iv_n}\sin v_n \tag{13b}$$

and

$$\tan u_n = \frac{\psi_n'(\beta)\psi_n(\alpha) - m\psi_n(\beta)\psi_n'(\alpha)}{\psi_n'(\beta)\chi_n(\alpha) - m\psi_n(\beta)\chi_n'(\alpha)} , \tag{14a}$$

$$\tan v_n = \frac{m\psi_n'(\beta)\psi_n(\alpha) - \psi_n(\beta)\psi_n'(\alpha)}{m\psi_n'(\beta)\chi_n(\alpha) - \psi_n(\beta)\chi_n'(\alpha)} , \tag{14b}$$

in which $\psi_n(\alpha)$ and $\chi_n(\alpha)$ are Riccati-Bessel functions. When the sphere is nonabsorbing, m and β are both real, giving real values for u_n and v_n, but for an absorbing sphere m and β are complex leading to values of u_n and v_n which are also complex. In the analysis here it will generally be assumed that the particle is nonabsorbing.

The behavior of $\psi_n(x)$ and $\chi_n(x)$ when x is fixed and n varies is of interest as such functions determine the multipole coefficients and hence the magnitudes of the scattering coefficients $(c_n)_p$. When x is large, $\psi_n(x)$ and $\chi_n(x)$ initially oscillate with a period ≈ 4 as the order increases. But for $n > x-2$, the functions are monotonic; $\psi_n(x)$ decreases rapidly from a positive maximum to zero, and $\chi_n(x)$ becomes increasingly negative from zero. Thus, for large spheres $\alpha > 2$, the magnitudes of the coefficients of the Gegenbauer series are expected to change erratically with order when $n < \alpha - 2$. This is followed by a region $\alpha - 2 < n < \alpha + 2$ over which the coefficients reduce rapidly in magnitude predominantly due to the factor $\psi_n(\alpha)/\chi_n(\alpha)$ which can be separated in Eq. (14). Asymptotic forms of $\psi_n(x)$ and $\chi_n(x)$ which are valid for $n > \alpha - 1/2$ may be used to obtain

$$\tan u_n \approx e^{(2n+1)(\tanh\xi - \xi)} \left\{ \frac{(1+2\gamma/\alpha)\sinh\xi}{\kappa+\lambda} - \frac{1}{2} \right\} , \tag{15a}$$

$$\tan v_n \approx e^{(2n+1)(\tanh\xi - \xi)} \left\{ \frac{\sinh\xi}{\kappa} - \frac{1}{2} \right\} , \tag{15b}$$

in which $\cosh\xi = (2n+1)/2\alpha$, $\kappa = m[\psi_{n-1}(\beta)/\psi_n(\beta)] - e^{-\xi}$ and $\lambda = (2\gamma/\alpha)\sinh\xi - (\gamma/\alpha^2)$.

As the arguments of the exponential factors are negative, the angles u_n and v_n become small when $n > \alpha + 2$. By Eqs. (13), we then have a_n, b_n, and $(c_n)_p \to 0$.

The Gegenbauer series for the scattered amplitude may therefore be terminated at $n \approx \alpha + 2$. In the case of small spheres $\alpha < 2$, $\psi_n(x)$, and $\chi_n(x)$ are monotonic for all orders, and a set of reducing coefficients $(c_n)_p$ is obtained.

The cutoff condition

$$n_{\text{CO}} \approx \alpha + 2 \tag{16}$$

for scattering amplitudes functions therefore appears to be generally valid.

V. RAYLEIGH-DEBYE SCATTERING

The physical basis of this theory is the condition that the refractive indices of the particle and the medium are nearly equal. As a consequence, refraction of the incident beam can be neglected, and the internal field of the particle is the same as the incident field. The internal field then induces dipoles throughout the specimen, and these radiate to generate the scattered field. The validity of the treatment is limited to particles with small optical thickness relative to the ambient medium $2(m-1)\alpha \approx 2\gamma \ll 1$.

For spherical particles, the scattering amplitudes are given by

$$\left.\begin{matrix} S_\perp \\ S_\parallel \end{matrix}\right\} = -i\frac{6\gamma\alpha^3}{(3\alpha+2\gamma)}\frac{j_1(u)}{u} \times \begin{cases} 1 & (17a) \\ \cos\vartheta , & (17b) \end{cases}$$

where $u = 2\alpha\sin(\vartheta/2)$ and $j_1(u)$ is the spherical Bessel function of order 1. However, these forms are unsuitable for comparison with the Mie theory, but, due to the completeness of Gegenbauer functions, they can be reexpressed as Gegenbauer series. Conversion is possible by using the relation

$$\frac{j_1(u)}{u} = \frac{1}{\alpha^2}\sum_{n=0}^{\infty} (2n+3) j_{n+1}^2(\alpha) T_n^1(\cos\vartheta) , \tag{18}$$

which allows Eq. (17a) to be converted directly. Equation (17b) requires an additional step due to the presence of $\cos(\vartheta)$. This factor may be incorporated by the application of

$$(2n+3)zT_n^1(z)=(n+1)T_{n+1}^1(z)+(n+2)T_{n-1}^1(z)\ , \quad (19)$$

where $z=\cos(\vartheta)$. The resulting expressions then have the desired forms

$$S_p=\sum_{n=0}^{\infty}(d_n)_p T_n^1(z)\ , \quad (20)$$

where

$$(d_n)_\perp=-\frac{i6\gamma\alpha}{(3\alpha+2\gamma)}(2n+3)j_{n+1}^2(\alpha)\ , \quad (21a)$$

$$(d_n)_\parallel=-\frac{i6\gamma\alpha}{(3\alpha+2\gamma)}\{nj_n^2(\alpha)+(n+3)j_{n+2}^2(\alpha)\}\ . \quad (21b)$$

VI. REDUCED MIE EQUATION

A comparison of Eqs. (11) and (20) now allows an examination to be made of the conditions required for the Mie formula to reduce to those given by Rayleigh-Debye theory. The most obvious difference is that $(c_n)_p$ are complex, while $(d_n)_p$ are imaginary. This can easily be dealt with by requiring u_n and v_n in Eqs. (14) to be small, then $a_n \to -iu_n$, $b_n \to -iv_n$, and

$$(c_n)_\perp \to -i\left\{\frac{n}{n+1}v_n+\frac{2n+3}{(n+1)(n+2)}u_{n+1}-\frac{n+3}{n+2}v_{n+2}\right\}\ , \quad (22a)$$

$$(c_n)_\parallel \to -i\left\{\frac{n}{n+1}u_n+\frac{2n+3}{(n+1)(n+2)}v_{n+1}-\frac{n+3}{n+2}u_{n+2}\right\}\ . \quad (22b)$$

Contributions from the real parts of a_n and b_n will be less than 5% of the imaginary parts for an upper limit on u_n and v_n of 0.05. Furthermore, the forms of $(d_n)_p$ encourage us to search for expressions of u_n and v_n in terms of squares of spherical Bessel functions $j_n^2(\alpha)$ or $\psi_n^2(\alpha)$. This leads us to apply the multiplication theorem of Bessel functions,

$$\psi_n(\beta)=\psi_n(m\alpha)=m^{n+1}\sum_{k=0}^{\infty}(-1)^k\frac{\gamma^k}{k!}\psi_{n+k}(\alpha)\ , \quad (23)$$

which, it should be noted, is valid for all values of α and γ. Substituting into Eq. (14) then yields

$$\tan u_n=\frac{\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}\bar{f}_n^k(\alpha)}{\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}\bar{g}_n^k(\alpha)}\ , \quad (24a)$$

$$\tan v_n=\frac{\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}f_n^k(\alpha)}{\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}g_n^k(\alpha)}\ , \quad (24b)$$

as shown in Appendix A, with the notation

$$f_n^k(\alpha)=\psi_{n+k-1}(\alpha)\psi_n(\alpha)-\psi_{n+k}(\alpha)\psi_{n-1}(\alpha)\ , \quad (25a)$$

$$\bar{f}_n^k(\alpha)=f_n^k(\alpha)+\frac{2k}{\alpha}\psi_{n+k-1}(\alpha)\psi_n'(\alpha)\ , \quad (25b)$$

$$g_n^k(\alpha)=\psi_{n+k-1}(\alpha)\chi_n(\alpha)-\psi_{n+k}(\alpha)\chi_{n-1}(\alpha)\ , \quad (26a)$$

$$\bar{g}_n^k(\alpha)=g_n^k(\alpha)+\frac{2k}{\alpha}\psi_{n+k-1}(\alpha)\chi_n'(\alpha)\ . \quad (26b)$$

The bar is used here only to distinguish between the two types of expression. These functions have limiting forms

$$f_n^k(\alpha)\to\frac{2k}{(2n+1)!!(2n+2k+1)!!}\alpha^{2n+k+1}\ ,$$

$$\bar{f}_n^k(\alpha)\to\frac{2k(n+1)}{(2n+1)!!(2n+2k-1)!!}\alpha^{2n+k-1}\ ,$$

$$g_n^k(\alpha)\to-\frac{(2n-1)!!}{(2n+2k-1)!!}\alpha^k\ ,$$

$$\bar{g}_n^k(\alpha)\to\frac{2kn(2n-1)!!}{(2n+2k-1)!!}\alpha^{k-2}\ ,$$

when $\alpha\to 0$, and

$$\bar{f}_n^k(\alpha)\to f_n^k(\alpha)\to\sin(k\pi/2)\ ,$$

$$\bar{g}_n^k(\alpha)\to g_n^k(\alpha)\to-\cos(k\pi/2)\ ,$$

when $\alpha\to\infty$. Thus the functions are generally well behaved over the entire range of α, and have magnitude limits of $\approx\pm 1$. One exception is $\bar{g}_n^1(\alpha)$, which has a simple pole at $\alpha=0$. The functions $f_n^1(\alpha)$ and $g_n^1(\alpha)$ are needed in the present analysis and are shown in Figs. 2–5. From the variation of $f_n^1(\alpha)$ with size parameter, it can be seen that the function is always positive with $f_n^1(\alpha)=0$ when $\alpha=0$, and $f_n^1(\alpha)\to 1$ when $\alpha\gg n$. Similar plots for $g_n^1(\alpha)$ are always negative with $g_n^1(\alpha)=0$ when $\alpha=0$, and $g_n^1(\alpha)\to 0$ for $\alpha\gg n$. Alternative displays of the functions against order, Figs. 4 and 5, reveal for $f_n^1(\alpha)$ the existence of a pedestal region which decreases irregularly from ≈ 1 at $n=0$ before reaching a

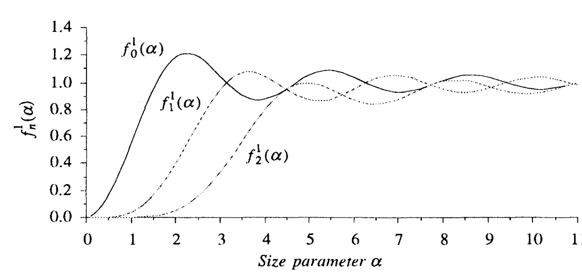

FIG. 2. Variation of $f_n^1(\alpha)$ with size parameter, $n=0$, 1, and 2.

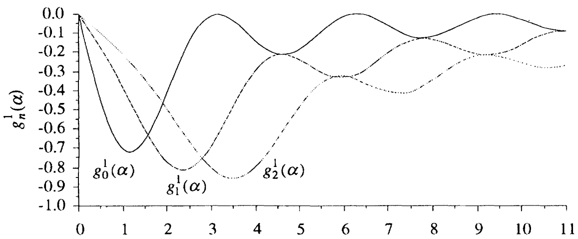

FIG. 3. Variation of $g_n^1(\alpha)$ with size parameter, $n=0$, 1, and 2.

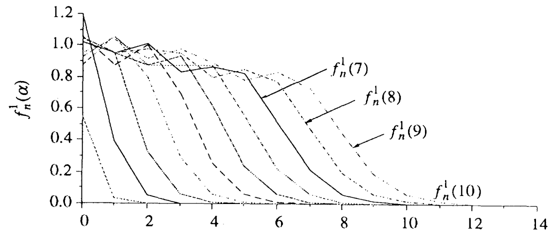

FIG. 4. Variation of $f_n^1(\alpha)$ with order, $\alpha=1,\ldots,10$.

cutoff edge centered at $n\approx\alpha-2$, while for $g_n^1(\alpha)$ a minimum is found at $n\approx\alpha-2$.

As the Riccati-Bessel functions obey recurrence relations, these may be employed to find similar formulas between $f_n^k(\alpha)$, $\bar{f}_n^k(\alpha)$ and $g_n^k(\alpha)$, $\bar{g}_n^k(\alpha)$. Such relations are derived in Appendix B. For the present purpose, the most important formulas are

$$f_n^0(\alpha)=\bar{f}_n^0(\alpha)=0\ , \tag{27}$$

$$g_n^0(\alpha)=\bar{g}_n^0(\alpha)=-1\ , \tag{28}$$

$$f_n^1(\alpha)=2\sum_{k=0}^{\infty}(4k+2n+3)j_{n+2k+1}^2(\alpha)\ , \tag{29}$$

$$\bar{f}_n^1(\alpha)=\frac{n+1}{2n+1}f_{n-1}^1(\alpha)+\frac{n}{2n+1}f_{n+1}^1(\alpha)\ , \tag{30}$$

$$g_n^1(\alpha)=-\frac{2n+1}{2\alpha}+2\sum_{k=0}^{\infty}(4k+2n+3)j_{n+2k+1}(\alpha)n_{n+2k+1}(\alpha)\ , \tag{31}$$

$$\bar{g}_n^1(\alpha)=\frac{2n}{(2n+1)\alpha}+\frac{n+1}{2n+1}g_{n-1}^1(\alpha)+\frac{n}{2n+1}g_{n+1}^1(\alpha)\ . \tag{32}$$

Note that the pole in $\bar{g}_n^1(\alpha)$ has been separated, and the apparent pole in $g_n^1(\alpha)$ is canceled by terms present in the summation. The $f_n^1(\alpha)$ and $\bar{f}_n^1(\alpha)$ functions have the form we require. Returning to Eq. (24), we impose the Rayleigh-Debye limit $\gamma\to 0$ to obtain

$$\tan u_n\approx\frac{\gamma\bar{f}_n^1(\alpha)}{1+\gamma\bar{g}_n^1(\alpha)}=\frac{\gamma\{(n+1)f_{n-1}^1(\alpha)+nf_{n+1}^1(\alpha)\}}{(2n+1)(1+\Delta_n)}+O(\gamma^2)\ , \tag{33a}$$

where $\Delta_n=[2n\gamma/(2n+1)\alpha]$ and

$$\tan v_n\approx\gamma f_n^1(\alpha)+O(\gamma^2)\ . \tag{33b}$$

These are the first order approximations. The Δ_n term arises by including the effect of the pole in $\bar{g}_n^1(\alpha)$. It is now apparent that the Rayleigh-Debye limit is a sufficient condition for u_n, $v_n\to 0$, with the criterion that both $\gamma f_0^1(\alpha)$ and $\gamma|g_0^1(\alpha)|\le 0.05$. The reduced Mie coefficients may be written as

$$(c_n)_\perp=-i\gamma\{(\kappa_n)_\perp f_n^1(\alpha)-(\lambda_n)_\perp f_{n+2}^1(\alpha)\}\ , \tag{34}$$

where

$$(\kappa_n)_\perp=\frac{(2n+3)\alpha+2n\gamma}{(2n+3)\alpha+2(n+1)\gamma}\ ,$$

$$(\lambda_n)_\perp=\frac{(n+2)(2n+3)\alpha+2(n+1)(n+3)\gamma}{(n+2)\{(2n+3)\alpha+2(n+1)\gamma\}}\ ,$$

and

$$(c_n)_\parallel=-i\gamma\{(\kappa_n)_\parallel f_{n-1}^1(\alpha)+(\lambda_n)_\parallel f_{n+1}^1(\alpha)-(\mu_n)_\parallel f_{n+3}^1(\alpha)\}\ , \tag{35}$$

with

$$(\kappa_n)_\parallel=\frac{n\alpha}{(2n+1)\alpha+2n\gamma}\ ,$$

$$(\lambda_n)_\parallel=\frac{(n+1)(2n+3)\alpha^2+2(5n^2+11n+3)\alpha\gamma+4n(2n+3)\gamma^2}{(n+1)\{(2n+1)\alpha+2n\gamma\}\{(2n+5)\alpha+2(n+2)\gamma\}}\ ,$$

$$(\mu_n)_\parallel=\frac{(n+3)\alpha}{(2n+5)\alpha+2(n+2)\gamma}\ .$$

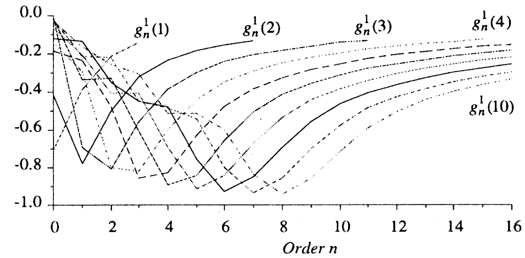

FIG. 5. Variation of $g_n^1(\alpha)$ with order, $\alpha=1,\ldots,10$.

Equations (34) and (35) are applicable for all values of α. They can be simplified further by specifying the condition $\alpha<1$ which ensures that $j_n(\alpha)$ decays monotonically with increasing order. Then $j_n^2(\alpha) \gg j_{n+1}^2(\alpha)$, and, by Eq. (29), $f_n^1(\alpha) \gg f_{n+2}^1(\alpha)$.

The Gegenbauer coefficients are now

$$(c_n)_\perp \approx -i2\gamma \left\{ \frac{(2n+3)\alpha+2n\gamma}{(2n+3)\alpha+2(n+1)\gamma} \right\} \times (2n+3) j_{n+1}^2(\alpha) + O(j_{n+3}^2(\alpha)), \quad n \ge 0 \tag{36}$$

and

$$(c_0)_\parallel \approx -i30\gamma \left\{ \frac{\alpha+2\gamma}{5\alpha+4\gamma} \right\} j_2^2(\alpha) + O(j_4^2(\alpha)) , \tag{37a}$$

$$(c_n)_\parallel \approx -i2\gamma \left\{ \frac{n\alpha}{(2n+1)\alpha+2n\gamma} \right\} (2n+1) j_n^2(\alpha) + O(j_{n+2}^2(\alpha)), \quad n \ge 1 . \tag{37b}$$

Even these restricted Mie coefficients differ from the equivalent Rayleigh-Debye values of

$$(d_n)_\perp = -i\frac{6\gamma\alpha}{3\alpha+2\gamma}(2n+3) j_{n+1}^2(\alpha), \quad n \ge 0 \tag{38}$$

and

$$(d_0)_\parallel = -i\frac{18\gamma\alpha}{3\alpha+2\gamma} j_2^2(\alpha) , \tag{39a}$$

$$(d_n)_\parallel \approx -i\frac{6\gamma\alpha}{3\alpha+2\gamma} n j_n^2(\alpha) + O(j_{n+2}^2(\alpha)), \quad n \ge 1 , \tag{39b}$$

although $(d_0)_\perp=(c_0)_\perp$ and $(d_1)_\parallel=(c_1)_\parallel$ when second order and higher terms are neglected. Nevertheless, the leading terms of the two sets $(c_n)_p$ and $(d_n)_p$ are in agreement when $\gamma/\alpha=(m^2-1)/2 \to 0$. Thus the criteria for the application of the Rayleigh-Debye (RD) theory to spheres are $\alpha<1$ and $\gamma/\alpha \to 0$.

A cutoff condition similar to that of the general sphere exists for the reduced Mie theory. This is evident from the behavior of $f_n^1(\alpha)$ with increasing orders, as displayed in Fig. 4, and can be made explicit by the asymptotic expression

$$f_n^1(\alpha) = \frac{e^{(2n+1)(\tanh\xi-\xi)}}{2\alpha \sinh^2(\xi)} , \tag{40}$$

where $\cosh\xi=(2n+1)/2\alpha$. As for the general sphere, $n_{\rm CO} \approx \alpha+2$.

VII. SCATTERING PATTERNS

The scattered irradiance is proportional to the square of the scattering amplitude, hence we let

$$I(z) = \frac{k^2r^2}{H_0} H^s(z) = |S(z)|^2 = \sum_{m,n=0}^{\infty} c_m c_n^* T_m^1(z) T_n^1(z) , \tag{41}$$

in which Eq. (7) has been applied. But this function also may be expressed as a Gegenbauer series

$$I(z) = \sum_{l=0}^{\infty} C_l T_l^1(z) , \tag{42}$$

due to the relation that exists between C_l and $c_m c_n^*$ terms. This may be obtained by examining the product space of Gegenbauer functions which shows that the decomposition of a product can be carried out by

$$T_m^1(z) T_n^1(z) = \sum_{l=|m-n|}^{m+n} A_l^{m,n} T_l^1(z) , \tag{43}$$

where the summation variable l changes in increments of 2. An expression for the product coefficients may be obtained as a special case of those derived for the products of Jacobi polynomials [11], and yields

$$A_l^{m,n} = \frac{(2l+3)}{4(l+1)(l+2)} \frac{(s+1)!(s+2)!}{(2s+3)!} \frac{(2a+2)!}{a!(a+1)!} \times \frac{(2b+2)!}{b!(b+1)!} \frac{(2c+2)!}{c!(c+1)!} \tag{44}$$

where $s=(l+m+n)/2$, $a=(m+n-l)/2$, $b=(l+m-n)/2$, and $c=(l+n-m)/2$. Each product term $T_m^1(z)T_n^1(z)$ generates $n+1$ terms, $T_{m-n}^1(z)$, $T_{m-n+2}^1(z), \ldots, T_{m+n}^1(z)$ when $n \le m$ as is shown in Fig. 6. The coefficients C_l of the scattering patterns therefore have contributions from various $c_m c_n^*$ product terms starting from a leading set, as is illustrated for $C_3 T_3^1(z)$. Particular relations are

$$C_0 = \sum_{n=0} |c_n|^2 A_0^{n,n} , \tag{45a}$$

$$C_1 = 2\,\mathrm{Re}\left\{ \sum_{n=0} c_n c_{n+1}^* A_1^{n,n+1} \right\} , \tag{45b}$$

$$C_2 = 2\,\mathrm{Re}\left\{ \sum_{n=0} c_n c_{n+2}^* A_2^{n,n+2} \right\} + \sum_{n=1} |c_n|^2 A_2^{n,n} , \tag{45c}$$

$$C_3 = 2\,\mathrm{Re}\left\{ \sum_{n=0} c_n c_{n+3}^* A_3^{n,n+3} \right\} + 2\,\mathrm{Re}\left\{ \sum_{n=1} c_n c_{n+1}^* A_3^{n,n+1} \right\} . \tag{45d}$$

As an example we consider the scattering pattern in perpendicular polarization of a RD sphere with $\alpha \approx 1$. The cutoff condition $n_{\rm CO} \approx \alpha+2$ indicates that we need only

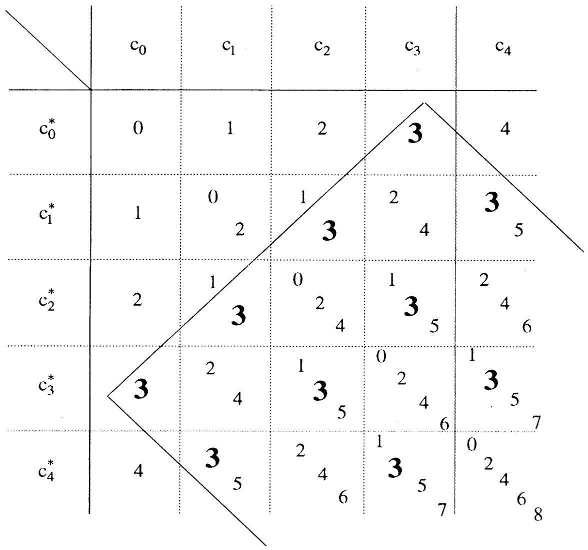

FIG. 6. Decomposition of the leading $T_m^1(z)T_n^1(z)$ products into $T_l^1(z)$ terms. Contributions to $C_3T_3^1(z)$ are shown bold.

to consider amplitude coefficients c_0 up to c_3, giving a 4×4 coefficient table. Referring to Eq. (34), we examine $f_n^1(\alpha)$ and obtain

$$f_0^1(\alpha)\approx0.545,\quad f_1^1(\alpha)\approx0.039\ ,$$

$$f_2^1(\alpha)\approx1.14\times10^{-3},\quad f_3^1(\alpha)\approx1.84\times10^{-5}\ .$$

However, the pattern coefficients depend on $f_n^1(\alpha)f_m^1(\alpha)$ terms giving a rapidly reducing sequence $C_0>C_1>C_2>C_3$, and hence we need consider C_l up to $l=2$ only. The scattering pattern therefore can be represented by

$$I_\perp(z)=C_0T_0^1(z)+C_1T_1^1(z)+C_2T_2^1(z)\ , \tag{46}$$

where

$$C_0=\gamma^2\{(\kappa_0)_\perp f_0^1(\alpha)\}^2\ ,$$

$$C_1=2\gamma^2(\kappa_0\kappa_1)_\perp f_0^1(\alpha)f_1^1(\alpha)\ ,$$

$$C_2=\gamma^2\{\tfrac{6}{5}[(\kappa_1)_\perp f_1^1(\alpha)]^2+2(\kappa_0\kappa_2)_\perp f_0^1(\alpha)f_2^1(\alpha)\}\ ,$$

since $A_0^{0,0}=A_1^{0,1}=A_2^{0,2}=1$ and $A_2^{1,1}=\frac{6}{5}$. This shows that the effective pattern table is 2×3, and suggests a general cutoff criterion for scattering patterns of

$$2\alpha-1<l_{\mathrm{CO}}<2\alpha+1\ . \tag{47}$$

The number of terms in the scattering which can be determined experimentally will, of course, depend on the sensitivity of the instrument. l_{CO} and n_{CO} only specify the orders at which the pattern and amplitude coefficients are small compared with lower order contributions.

VIII. DISCUSSION

The application of Gegenbauer analysis to light scattering from homogeneous spheres has shown that the Mie and Rayleigh-Debye theories can be formulated in terms of Gegenbauer functions. This allows a theoretical comparison to be made between the two treatments. From the analysis, it has been demonstrated that for Rayleigh-Debye scattering to be valid an upper limit of $\alpha<1$ must be placed on the particle size as well as the usual phase condition $2\gamma\approx(m^2-1)\alpha\ll1$. Under these conditions, the dominant amplitude terms is the same in the two theories, but other terms differ. The reduced Mie formulation is preferred for Rayleigh-Debye spheres since it is based on the rigorous application of electromagnetic theory and the particle size is unrestricted. Although the Rayleigh-Debye equations are simple, this simplicity cannot be used to justify their possible use, as only a few terms of the scattering pattern need be calculated when $\alpha<1$.

An attractive and powerful feature of the Gegenbauer formulation for the more general case of spheres of arbitrary size and refractive index is the ability to convert experimental scattering patterns $I(z)$ to a set of coefficients by the integration

$$C_l=\frac{2l+3}{2(l+1)(l+2)}\int_{-1}^{1}(1-z^2)I(z)T_l^1(z)dz\ , \tag{48}$$

where $z=\cos\vartheta$. The Gegenbauer spectrum of the pattern may then be generated by plotting the coefficients against order. Examples of computed scattering patterns for spectra are illustrated in Figs. 7–9. Despite the difference in the appearance of the patterns and their spectra, the two modes of representation are complementary and contain the same information. This follows since $I(z)$ and the set C_l constitute a Gegenbauer transform pair. Accordingly, $I(z)$ can be reconstructed by the inverse transform $I(z)=\sum_{l=0}C_lT_l^1(z)$. In general, the

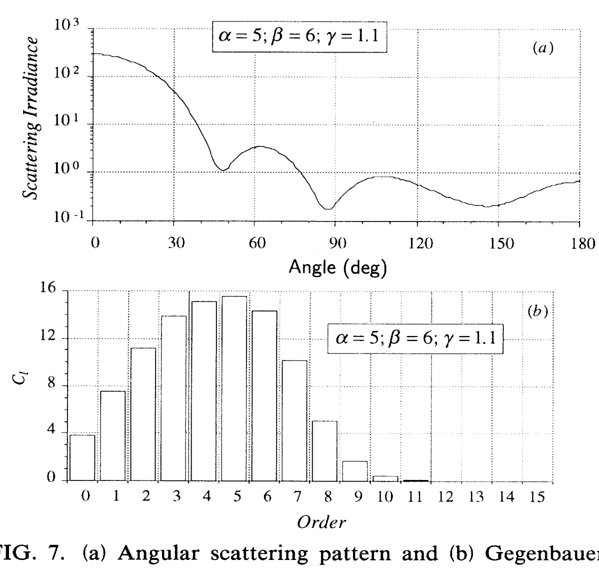

FIG. 7. (a) Angular scattering pattern and (b) Gegenbauer spectrum for particles $\alpha=5$, $\beta=6$, and $\gamma=1.1$.

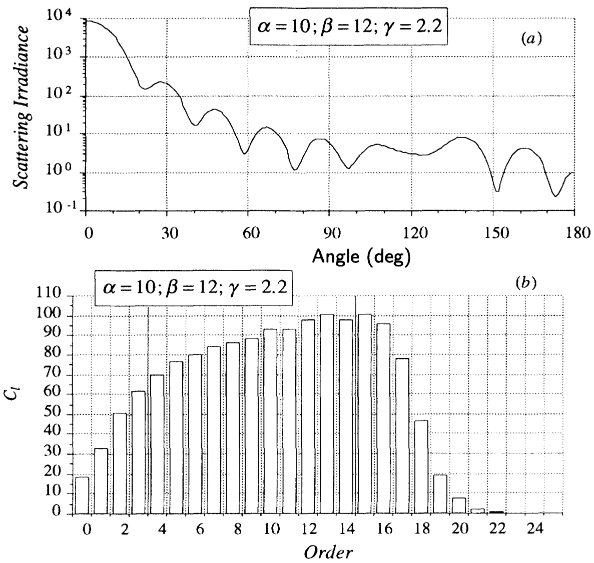

FIG. 8. (a) Angular scattering pattern and (b) Gegenbauer spectrum for particles $\alpha=10$, $\beta=12$, and $\gamma=2.2$.

spectra plots have simpler structure and require fewer data points than the scattering patterns. They also exhibit a high-order cutoff l_{CO} from which the external size parameter can be deduced.

For computed scattering patterns, the most direct procedure for calculating Gegenbauer spectra is from the Mie multipole coefficients a_n and b_n through Eqs. (12) and (45). Spectra derived from experimental data, on the other hand, can be obtained by integral (48), and introduce an instrumental factor for converting the scattering irradiance $H^s(z)$ into the scattering function $I(z)$:

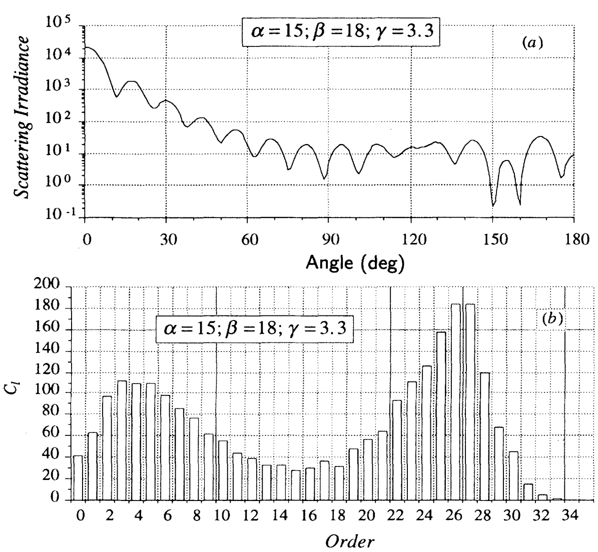

FIG. 9. (a) Angular scattering pattern and (b) Gegenbauer spectrum for particles $\alpha=15$, $\beta=18$, and $\gamma=3.3$.

$$I(z)=\frac{k^2r^2}{H_0}H^s(z) \; .$$

Evaluation of the instrumental factor is possible either from the instrument parameters or by conducting experiments on standard spherical reference particles. Yet another complication in the implementation of Eq. (48) arises from missing data near 0° and 180°. However, analysis shows that the effect of omitting data in the ranges 0 to ϑ_0 and $\pi-\vartheta_0$ to π is to superimpose a high-order ripple given by $-\vartheta_0^3 J_2(l\vartheta_0)/(l\vartheta_0)$, when l is large, on the Gegenbauer spectrum. Such a ripple can be detected for $l>l_{CO}$ and subtracted by fitting forward and backward scattering to the forms $A+B\sin^2(\vartheta/2)+C\sin^4(\vartheta/2)$. Further processing may also be introduced to eliminate random noise in the scattering pattern having high-order angular periodicity. The accuracy of the final spectra will clearly be highest when quasicontinuous scattering patterns are available.

A cutoff order $2\alpha-1<l_{CO}<2\alpha+1$ is introduced from the present analysis, but numerical studies have shown that $l_{CO}=2.1\alpha+0.12$, which is hardly affected by the particle's refractive index. This allows the direct determination of particle size. Particular equations for the cutoff order will, of course, depend on the cutoff criterion used.

The number of terms in the scattering pattern which can be detected experimentally will depend on the sensitivity of the instrument as well as the particle properties. The amplitude and pattern cutoff orders n_{CO} and l_{CO} only specify the orders at which the coefficients are small compared with lower order terms. This raises the possibility of a complete inversion of the scattering pattern.

We consider first the case of Rayleigh-Debye spheres. For perpendicular polarization, Eq. (48) yields

$$\frac{f_1^1(\alpha)}{f_0^1(\alpha)}=\frac{3}{2}\frac{(3+2m^2)}{(2+m^2)(4+m^2)}\frac{C_1}{C_0} \tag{49}$$

and

$$m^2=\frac{3\alpha f_0^1(\alpha)+4\sqrt{C_0}}{3\alpha f_0^1(\alpha)-2\sqrt{C_0}} \; , \tag{50}$$

in which

$$\frac{f_1^1(\alpha)}{f_0^1(\alpha)}=\frac{1+\sin\alpha\{\cos\alpha-(2\sin\alpha)/\alpha\}/\alpha}{1-(\sin\alpha\cos\alpha)/\alpha} \tag{51}$$

varies monotonically for $\alpha<2$. Hence α and m may be determined iteratively by starting with an initial value of $m=1$. A further equation

$$\frac{f_2^1(\alpha)}{f_1^1(\alpha)}=\frac{(4+m^2)(4+3m^2)}{(3+2m^2)(5+2m^2)}\left\{\frac{C_2}{C_1}-\frac{3C_1}{10C_0}\right\} \tag{52}$$

offers a useful check on the presence of experimental errors or deviations of the particle from the Rayleigh-Debye sphere model.

A similar analysis for parallel polarization to that in Sec. VII leads to

$$I_{\|}=C_0T_0^1(z)+C_1T_1^1(z)+C_2T_2^1(z)+C_3T_3^1(z)+C_4T_4^1(z)\ , \tag{53}$$

where

$$C_0=\tfrac{3}{2}C_2=\tfrac{9}{5}\{\gamma(\kappa_1)_{\|}f_0^1(\alpha)\}^2\ ,$$

$$C_3=\frac{9C_1}{12+(\lambda_0/\kappa_2)_{\|}}=\tfrac{18}{7}\gamma^2(\kappa_1\kappa_2)_{\|}f_0^1(\alpha)f_1^1(\alpha)\ ,$$

$$C_4=\gamma^2\{\tfrac{10}{7}[(\kappa_2)_{\|}f_1^1(\alpha)]^2+\tfrac{8}{3}(\kappa_1\kappa_3)_{\|}f_0^1(\alpha)f_2^1(\alpha)\}\ .$$

The coefficients now follow the sequence $C_0\geq C_2>C_1\geq C_3>C_4$, and the iteration equations are

$$\frac{f_1^1(\alpha)}{f_0^1(\alpha)}=\frac{7}{20}\frac{(3+2m^2)}{(2+m^2)}\frac{C_3}{C_0} \tag{54}$$

and

$$m^2=\frac{3\alpha f_0^1(\alpha)+4\sqrt{5C_0}}{3\alpha f_0^1(\alpha)-2\sqrt{5C_0}}\ . \tag{55}$$

Additional relations

$$\frac{C_1}{C_3}=\tfrac{1}{6}(8+7m^2) \tag{56}$$

and

$$\frac{f_2^1(\alpha)}{f_1^1(\alpha)}=\frac{1}{4}\frac{(4+3m^2)}{(3+2m^2)}\left\{\frac{18}{7}\frac{C_4}{C_3}-\frac{C_3}{C_0}\right\} \tag{57}$$

may also be obtained.

The direct inversion of Rayleigh-Debye spheres is possible due to the imaginary amplitude coefficients and the rapid reduction of their magnitudes with order. In the case of the general sphere such conditions apply only near or above cutoff. Below cutoff the coefficients are complex and vary erratically in magnitude with order. If the instrument limits the coefficients to $c_0,\ldots,c_k,\ldots,c_N$, in which the first $k+1$ are complex and the last $N-k$ are imaginary, then the number of unknown variables to be found is $k+N+2$. This should be compared with the number of experimental values $2N+1$ of $C_0,\ldots,C_{2N}$. Thus the amplitude coefficients can be determined from Eq. (45) when $N\geq k+1$.

The equations unfortunately are not linear in c_n but involve products $c_nc_m^*+c_n^*c_m$, so that optimum algorithms for solutions will need to be investigated. Favorable factors are that (a) a reasonable value for α is known from l_{CO}; (b) the highest order coefficients have simple relations, e.g., $C_{2N}=A_{2N}^{N,N}|c_N|^2$ and $C_{2N-1}=2\,\mathrm{Re}\{A_{2N-1}^{N-1,N}c_{N-1}^*c_N\}$; and (c) asymptotic expressions are available for the highest order amplitudes.

Once the c_n coefficients are obtained the multipole coefficients a_n and b_n may be found prior to determining best fit solutions of α and β from Eqs. (13) and (14).

This paper concerns the theory and application of Gegenbauer analysis to light scattering from homogeneous dielectric spheres. The treatment is also applicable to absorbing and layered spheres. Other papers are in preparation covering studies based on theoretical and experimental scattering patterns.

APPENDIX A

The properties of Riccati-Bessel functions are as follows.

(a) Definitions

$$\psi_n(z)=zj_n(z)=\left[\frac{\pi z}{2}\right]^{1/2}J_{n+1/2}(z)\ ,$$

$$\chi_n(z)=zn_n(z)=\left[\frac{\pi z}{2}\right]^{1/2}N_{n+1/2}(z)\ .$$

(b) Recurrence formulas

$$(2n+1)\phi_n(z)=z\{\phi_{n-1}(z)+\phi_{n+1}(z)\}\ , \tag{A1}$$

$$\frac{d}{dz}\phi_n(z)=\phi_{n-1}(z)-\frac{n}{z}\phi_n(z)\ , \tag{A2}$$

$$\frac{d}{dz}\phi_n(z)=\frac{n+1}{z}\phi_n(z)-\phi_{n+1}(z)\ , \tag{A3}$$

if $\phi_n(z)=\psi_n(z)$ or $\chi_n(z)$ or any linear combination of the two.

(c) Wronskian

$$\psi_n(z)\frac{d}{dz}\chi_n(z)-\chi_n(z)\frac{d}{dz}\psi_n(z)=1 \tag{A4}$$

or, by using Eq. (A2),

$$\psi_n(z)\chi_{n-1}(z)-\chi_n(z)\psi_{n-1}(z)=1\ . \tag{A5}$$

(d) Multiplication theorem

$$\psi_n(\beta)=\psi_n(m\alpha)=m^{n+1}\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}\psi_{n+k}(\alpha)\ , \tag{A6}$$

where $\gamma=(m^2-1)\alpha/2$.

(e) Products. If we let $A=m\psi_n'(\beta)\phi_n(\alpha)-\psi_n(\beta)\phi_n'(\alpha)$, then, by Eq. (A2),

$$A=m\psi_{n-1}(\beta)\phi_n(\alpha)-\psi_n(\beta)\phi_{n-1}(\alpha)\ . \tag{A7}$$

On substituting (A6) into this, we obtain

$$A=m^{n+1}\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}B_n^k(\alpha)\ , \tag{A8}$$

where

$$B_n^k(\alpha)=\psi_{n+k-1}(\alpha)\phi_n(\alpha)-\psi_{n+k}(\beta)\phi_{n-1}(\alpha)\ .$$

For particular $\phi_n(\alpha)$,

$$B_n^k(\alpha)=\begin{cases}f_n^k(\alpha)\\ g_n^k(\alpha)\end{cases}\text{ when }\phi_n(\alpha)=\begin{cases}\psi_n(\alpha)\\ \chi_n(\alpha)\ .\end{cases}$$

In the case of $\overline{A}=\psi_n'(\beta)\phi_n(\alpha)-m\psi_n(\beta)\phi_n'(\alpha)$, rearrangement gives

$$\overline{A}=\frac{1}{m}(A-C)\ , \tag{A9}$$

where

$$C=(m^2-1)\psi_n(\beta)\phi_n'(\alpha) \ . \tag{A10}$$

This may be expanded using Eq. (A6) as

$$C=m^{n+1}\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}\frac{2\gamma}{\alpha}\psi_{n+k}(\alpha)\phi_n'(\alpha)$$
$$=-m^{n+1}\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}\frac{2k}{\alpha}\psi_{n+k-1}(\alpha)\phi_n'(\alpha) \ . \tag{A11}$$

Hence

$$\overline{A}=m^n\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}\overline{B}_n^k(\alpha) \ , \tag{A12}$$

in which

$$\overline{B}_n^k(\alpha)=B_n^k(\alpha)+\frac{2k}{\alpha}\psi_{n+k-1}(\alpha)\phi_n'(\alpha) \ .$$

For particular $\phi_n(\alpha)$,

$$\overline{B}_n^k(\alpha)=\begin{cases}\overline{f}_n^k(\alpha)\\ \overline{g}_n^k(\alpha)\end{cases} \text{when } \phi_n(\alpha)=\begin{cases}\psi_n(\alpha)\\ \chi_n(\alpha) \ .\end{cases}$$

(f) The scattering functions

$$\tan u_n=\frac{\psi_n'(\beta)\psi_n(\alpha)-m\psi_n(\beta)\psi_n'(\alpha)}{\psi_n'(\beta)\chi_n(\alpha)-m\psi_n(\beta)\chi_n'(\alpha)} \tag{A13}$$

has a numerator and denominator of the form of $\overline{A}$, so that

$$\tan u_n=\frac{\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}\overline{f}_n^k(\alpha)}{\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}\overline{g}_n^k(\alpha)} \ . \tag{A14}$$

Similarly, the function

$$\tan v_n=\frac{m\psi_n'(\beta)\psi_n(\alpha)-\psi_n(\beta)\psi_n'(\alpha)}{m\psi_n'(\beta)\chi_n(\alpha)-\psi_n(\beta)\chi_n'(\alpha)} \tag{A15}$$

has a numerator and denominator of the form of A. Hence

$$\tan v_n=\frac{\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}f_n^k(\alpha)}{\sum_{k=0}^{\infty}\frac{(-\gamma)^k}{k!}g_n^k(\alpha)} \ . \tag{A16}$$

APPENDIX B

The properties of scattering functions are as follows.

(a) Recurrence formulas of $B_n^k(z)$. The argument of all functions is z. Starting with

$$B_n^k=\psi_{n+k-1}\phi_n-\psi_{n+k}\phi_{n-1} \ , \tag{B1}$$

a downward recursion of ϕ_n and ϕ_{n-1}, using Eq. (A1), gives

$$B_n^k=\frac{2}{z}\psi_{n+k-1}\phi_{n-1}+\frac{(2n-3)}{z}B_{n-1}^{k+1}-B_{n-2}^{k+2} \tag{B2}$$

and

$$B_n^k=\frac{2}{z}\psi_{n+k}\phi_{n-2}+\frac{(2n-1)}{z}B_{n-1}^{k+1}-B_{n-2}^{k+2} \ . \tag{B3}$$

From these we derive

$$\frac{2}{z}\psi_{n+k}\phi_n=B_{n+1}^k-\frac{(2n-1)}{z}B_n^{k+1}+B_{n-1}^{k+2} \tag{B4}$$

and

$$\frac{2}{z}\psi_{n+k}\phi_n=B_{n+2}^{k-2}-\frac{(2n+3)}{z}B_{n+1}^{k-1}-B_n^k \ . \tag{B5}$$

An upward recursion of ψ_{n+k} and ψ_{n+k-1} in Eq. (B1) also leads to

$$\frac{2}{z}\psi_{n+k}\phi_n=\frac{(2n+2k+3)}{z}B_n^{k+1}-B_n^k-B_n^{k+2} \ . \tag{B6}$$

This can be combined with Eq. (B4) to give the recurrence relations of $B_n^k(z)$ as

$$B_n^{k+2}+B_{n+1}^{k+2}=\frac{2}{z}(2n+k+3)B_{n+1}^{k+1}-(B_{n+1}^k+B_{n+2}^k) \ . \tag{B7}$$

(b) Recurrence formulas of $f_n^k(z)$. When $\phi_n=\psi_n$, $B_n^k=f_n^k$, and $f_n^0=0$,

$$f_{n+k}^{-k}=-f_n^k \ . \tag{B8}$$

Various equations can be generated for particular values of k from Eqs. (B4) and (B5) using (B8), e.g.,

$$\frac{2}{z}\psi_n^2=f_{n-1}^2-\frac{(2n-1)}{z}f_n^1 \ , \tag{B9}$$

$$\frac{2}{z}\psi_n^2=\frac{(2n+3)}{z}f_n^1-f_n^2 \ , \tag{B10}$$

$$\frac{2}{z}\psi_n\psi_{n+1}=f_n^1-f_{n+1}^1 \ , \tag{B11}$$

and

$$\frac{2}{z}\psi_n\psi_{n+1}=f_{n-1}^3-\frac{(2n-1)}{z}f_n^2+f_{n+1}^1 \ . \tag{B12}$$

From Eq. (B11), we have $f_{n+1}^1-f_{n+2}^1=(2/z)\psi_{n+1}\psi_{n+2}$ and $f_n^1-f_{n+1}^1=(2/z)\psi_n\psi_{n+1}$, which are summed to obtain

$$f_n^1-f_{n+2}^1=\frac{2}{z}\{\psi_n+\psi_{n+2}\}\psi_{n+1}$$
$$=\frac{2}{z^2}(2n+3)\psi_{n+1}^2 \tag{B13}$$

by using Eq. (A1). Thus f_n^1 may be written as a series

$$f_n^1=\frac{2}{z^2}\sum_{s=0}^{\infty}(4s+2n+3)\psi_{2s+n+1}^2 \ , \tag{B14}$$

which is always positive. In a similar manner the recurrence relations within Eq. (B7) lead to

$$f_n^2=\frac{2}{z}\sum_{s=0}^{\infty}(-1)^s(2s+2n+3)f_{s+n+1}^1 \ , \tag{B15}$$

$$f_n^3+f_{n+1}^1=\frac{2}{z}\sum_{s=0}^{\infty}(-1)^s(2s+2n+4)f_{s+n+1}^2 \ , \tag{B16}$$

and

$$f_n^4+f_{n+1}^2=\frac{2}{z}\sum_{s=0}^{\infty}(-1)^s(2s+2n+5)f_{s+n+1}^3 \ . \qquad \text{(B17)}$$

General expressions for $\bar{f}_n^k$ are complicated, so we will examine $\bar{f}_n^1$ only. This is defined as

$$\bar{f}_n^1=f_n^1+\frac{2}{z}\psi_n\psi_n'$$

or

$$\bar{f}_n^1=f_n^1+\frac{2}{z}\psi_n\left[\psi_{n-1}-\frac{n}{z}\psi_n\right] \qquad \text{(B18)}$$

by Eq. (A2). The application of Eqs. (B11) and (B13) then gives

$$\bar{f}_n^1=\frac{(n+1)}{(2n+1)}f_{n-1}^1+\frac{n}{(2n+1)}f_{n+1}^1 \ . \qquad \text{(B19)}$$

(c) Recurrence formulas of $g_n^k(z)$. When $\phi_n=\chi_n$, $B_n^k=g_n^k$, and

$$g_n^0=\psi_{n-1}\chi_n-\psi_n\chi_{n-1}=-1 \ , \qquad \text{(B20)}$$

$$g_n^1+g_{n+1}^{-1}=-\frac{2n+1}{z} \ . \qquad \text{(B21)}$$

Letting $k=-1$ in Eq. (B4), and using Eqs. (B20) and (B21), we have

$$\frac{2}{z}\psi_{n-1}\chi_n=g_{n+1}^{-1}+g_{n-1}^1+\frac{(2n-1)}{z}$$

$$=g_{n-1}^1-g_n^1-\frac{2}{z} \ . \qquad \text{(B22)}$$

Thus

$$g_n^1-g_{n+1}^1=\frac{2}{z}\{1+\psi_n\chi_{n+1}\}=\frac{2}{z}\psi_{n+1}\chi_n \qquad \text{(B23)}$$

and

$$g_{n+1}^1-g_{n+2}^1=\frac{2}{z}\{1+\psi_{n+1}\chi_{n+2}\} \ , \qquad \text{(B24)}$$

which may be summed to obtain

$$g_n^1-g_{n+2}^1=\frac{2}{z}\left\{1+\frac{(2n+3)}{z}\psi_{n+1}\chi_{n+1}\right\} \ . \qquad \text{(B25)}$$

A more convenient form of this equation is

$$\left\{g_n^1+\frac{(2n+1)}{2z}\right\}-\left\{g_{n+2}^1+\frac{(2n+5)}{2z}\right\}$$

$$=\frac{2}{z^2}(2n+3)\psi_{n+1}\chi_{n+1} \ , \qquad \text{(B26)}$$

giving a series solution

$$g_n^1=-\frac{2n+1}{2z}+\frac{2}{z^2}\sum_{s=0}^{\infty}(4s+2n+3)\psi_{2s+n+1}\chi_{2s+n+1} \ . \qquad \text{(B27)}$$

Other functions may be obtained from the recurrence relations

$$g_n^2+g_{n+1}^2=2\left\{1+\frac{(2n+3)}{z}g_{n+1}^1\right\} \ , \qquad \text{(B28)}$$

$$g_n^3+g_{n+1}^3=2\frac{(2n+4)}{z}g_{n+1}^2-(g_{n+1}^1+g_{n+2}^1) \ , \qquad \text{(B29)}$$

and

$$g_n^4+g_{n+1}^4=2\frac{(2n+5)}{z}g_{n+1}^3-(g_{n+1}^2+g_{n+2}^2) \ . \qquad \text{(B30)}$$

For $\bar{g}_n^1$, we have $\bar{g}_n^1=g_n^1+(2/z)\psi_n\chi_n'$ and

$$\bar{g}_n^1=g_n^1+\frac{2}{z}\psi_n\left\{\chi_{n-1}-\frac{n}{z}\chi_n\right\} \ , \qquad \text{(B31)}$$

using Eq. (A2). The application of Eqs. (B23) and (B25) then yields

$$\bar{g}_n^1=\frac{2n}{(2n+1)z}+\frac{n+1}{(2n+1)}g_{n-1}^1+\frac{n}{(2n+1)}g_{n+1}^1 \ . \qquad \text{(B32)}$$

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