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Letter

A note on the spherical harmonic expansion of the Mie scattering kernel

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Abstract. Three methods of obtaining the Mie scattering function are considered and compared. Legendre polynomials and Clebsh–Gordon coefficients are employed and an exact expression for the expansion of the Mie kernel in spherical harmonics is given.

1. Introduction

The absorption and scattering of electromagnetic radiation by a sphere of arbitrary size and refractive index is an exactly solvable problem with an extensive literature [1–3]. The angular variation of intensity of the scattered light is the radial component of the Poynting vector of the radiation and has the form, for unpolarised light

$$I_s(\theta)=S_{11}(\theta)I_i, \tag{1}$$

where I_i is the incident intensity and the scattering kernel S_{11} is expressible as

$$S_{11}(\theta)=\frac{1}{2x^2}\sum_{n,m}\left[\frac{(2n+1)}{n(n+1)}\frac{(2m+1)}{m(m+1)}\right][A_{n,m}(x)F_{n,m}\cos\theta+B_{n,m}(x)G_{n,m}\cos\theta], \tag{2}$$

where

$$A_{n,m}(x)=a_n(x)a_m(x)+b_n(x)b_m(x), \tag{3 a}$$

$$B_{n,m}(x)=a_n(x)b_m(x)+b_n(x)a_m(x), \tag{3 b}$$

$$F_{n,m}(\mu)=\pi_n(\mu)\pi_m(\mu)+\tau_n(\mu)\tau_m(\mu), \tag{3 c}$$

$$G_{n,m}(\mu)=\pi_n(\mu)\tau_m(\mu)+\tau_n(\mu)\pi_m(\mu). \tag{3 d}$$

Here $\mu=\cos(\theta)$ and the Mie coefficients a and b are derived from the Ricatti–Bessel functions. The size parameter is given by $x=2\pi n_m r/\lambda$ for a particle radius r, with refractive index n_p, irradiated by light wavelength λ, in a medium with refractive index n_m. The coefficients a and b depend also on the relative refractive index $\nu=n_p/n_m$.

Also

$$\pi_j\cos\theta=\frac{P_j^1\cos\theta}{\sin\theta}, \tag{4 a}$$

and

$$\tau_j \cos\theta = \frac{d}{d\theta} P_j^1 \cos\theta, \tag{4b}$$

where $P_j^1(\mu)$ is the associated Legendre polynomial of order j and degree 1.

The summation in equation (2) is infinite, but in practical computation schemes an upper limit of $N_{max} = O(x)$ is used. Several authors have discussed the appropriate value of N_{max}; following Bohren and Huffman [3] we have used $N_{max} = x + 4x^{1/3} + 2$. The correctness of (1) has been demonstrated over a wide range of values of x and ν.

Often it is desirable to have an expression for S_{11} in terms of a series of spherical harmonics $Y_{jk}(\theta, \phi)$ which are related to the associated Legendre functions by

$$Y_j^k(\theta, \phi) = P_j^k \cos\theta \exp ik\phi \tag{5}$$

For example, some methods for finding solutions to the radiative transfer equation [4] make use of such an expansion [5–7] and for particle sizing experiments [8, 9] the singular spectrum of the scattering kernel may be developed in spherical harmonics [10].

Any function that is finite on a sphere is expandable in spherical harmonics. Since the Mie kernel is symmetric in the azimuthal angle, an expansion may be found in ordinary Legendre polynomials only ($k = 0$), also known as the Legendre transform of the function [11]. We thus seek the coefficients c_j in the expansion

$$S_{11} = \sum c_j P_j^0, \tag{6}$$

which are given by

$$c_j = (j + 1/2) \int_{-1}^{1} P_j^0(\mu) S_{11}(\mu)\, d\mu. \tag{7}$$

Since S_{11} is obtained numerically, a numerical integration is possible for the set $\{c_j\}$. It is simple to prove that the expansion will require Legendre polynomials of order up to $2N_{max}$ (see Appendix). Then the I/θ curve will contain $2N_{max}$ oscillations [12]. In general this will be quite a complex integration to achieve numerically. This is required, however, if the limits of integration are not $[-1, 1]$ in (7). Such an approach has usually been adopted by others [13].

In this communication an exact expression for the integral (7) is derived by considering orthogonality properties of the associated Legendre polynomials, and results for the products of such polynomials derived from the group representation analysis of rotations in three-dimensional space.

2. Method

Let L be the Legendre differential operator

$$L[f(\mu)] = \frac{d}{d\mu}(1 - \mu^2)\frac{d}{d\mu}[f(\mu)]. \tag{8}$$

Then the function $P_j^k(\mu)$ satisfies the associated Legendre equation

$$L[P_j^k(\mu)] = \left[\lambda_j + \frac{k^2}{(1 - \mu^2)}\right] P_j^k(\mu), \tag{9}$$

where λ_j is the eigenvalue of the ordinary Legendre equation ($k=0$) given by

$$\lambda_j = -j(j+1).$$

In the Mie kernel, with $k=1$, we have

$$F_{n,m}(\mu) = (1/2)[L - (\lambda_n + \lambda_m)]P_m^1 P_n^1, \tag{10}$$

$$G_{n,m}(\mu) = \frac{\mathrm{d}}{\mathrm{d}\mu}(P_m^1 P_n^1). \tag{11}$$

Thus substituting with (7) we obtain

$$c_j = \frac{(j+1/2)}{2x^2} \sum_{n,m}^{N_{\max}} \frac{(2n+1)(2m+1)}{\lambda_n \lambda_m} \times \int_{-1}^{1} P_j^0(\mu) \left\{ A_{m,n}(1/2)[L - (\lambda_n + \lambda_m)] + B_{n,m} \frac{\mathrm{d}}{\mathrm{d}\mu} \right\} P_m^1(\mu) P_n^1(\mu) \, \mathrm{d}\mu. \tag{12}$$

Since L is self-adjoint we have

$$\int_{-1}^{1} fLg = \int_{-1}^{1} gLf,$$

and therefore

$$c_j = \frac{(j+1/2)}{2x^2} \sum_{n,m}^{N_{\max}} \frac{(2n+1)(2m+1)}{\lambda_n \lambda_m} \times \int_{-1}^{1} A_{m,n}(1/2)[\lambda_j - (\lambda_n + \lambda_m)] P_j^0 P_m^1 P_n^1 - B_{n,m}\left(\frac{\mathrm{d}}{\mathrm{d}\mu} P_j^0\right) P_m^1 P_n^1 \, \mathrm{d}\mu, \tag{13}$$

where we have used integration by parts and the fact that $P_j^k(1) = P_j^k(-1) = 0 \, \forall \, k \neq 0$.

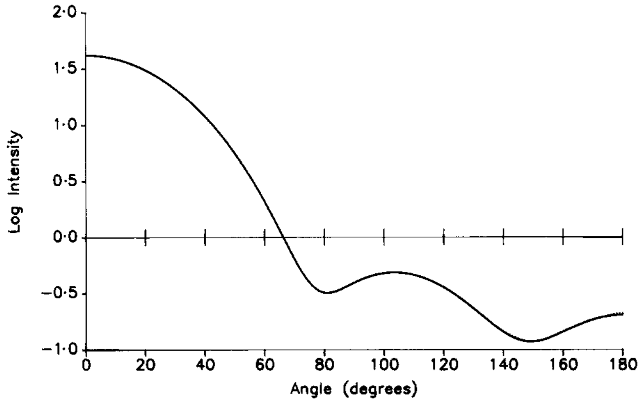
The first term in equation (13) is the integral of three associated Legendre polynomials. This is given straightforwardly by the application of the Clebsh–Gordon coefficients (also called the Wigner, or $3-j$ coefficients) familiar in quantum mechanics,

$$\int_{-1}^{1} P_j^k(\mu) P_m^l(\mu) P_n^p(\mu) \, \mathrm{d}\mu = \left[\frac{(l+m)!(j+n)!}{(l-m)!(j-n)!}\right]^{1/2} \times C(j,m,n;k,l,-p) C(j,m,n;0,0,0) \left[\frac{(k+p)!}{(k-p)!}\right], \tag{14}$$

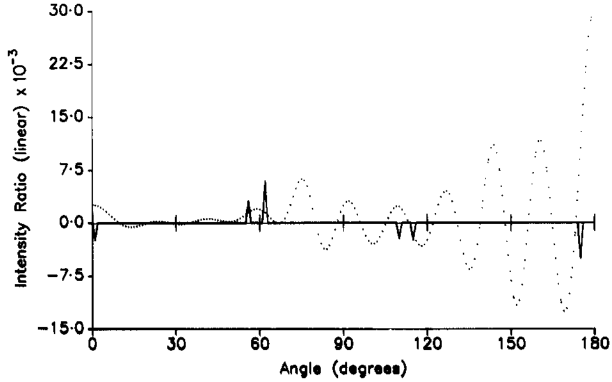
where $p = m + n$. The Clebsh–Gordon (C–G) coefficients arise naturally from consideration of the tensor product of irreducible representations of the Lie group SU(2) of rotation in space [14, 15]. They may be given explicitly by a variety of formulae, such as

$$C(j,m,n;k,l,p) = (-1)^{2j-m+p} \left[\frac{(j+m-n)!(n+j-m)!(m+n-j)!(n+p)!(n-p)!}{(j+m+n+1)!(j+k)!(j-k)!(m+l)!(m-l)!}\right]^{1/2} \times \sum_s (-1)^s \frac{(j+n-l-s)!(m+l+s)!}{s!(n+p-s)!(s+m-j-p)!(n-m+j-s)!}, \tag{15}$$

where the summation is understood to be over all values of s such that none of the factorial functions have negative argument. In addition they satisfy many symmetry



(*a*)



(*b*)

Figure 1. Mie scattering function, a comparison of methods. (*a*) The log intensity of scattered unpolarised radiation as a function of angle for a water droplet in air of size parameter 3, assuming the refractive index of water is $1{\cdot}33 + \mathrm{i} \times 10^{8}$. Three methods: (i) direct evaluation of the Mie function [3]—M_{D} (———); (ii) evaluation by summing the first 20 Legendre polynomials, weighted by coefficients produced by the C–G method (see text)—M_{CG} (—·—) (iii) by numerical integration of (i) with each Legendre polynomial—M_{NI} (·····). The difference in the curves is not visible in this diagram. (*b*) The ratio functions $(M_{\mathrm{CG}}/M_{\mathrm{D}} - 1) \times 10^{3}$ (solid) and $M_{\mathrm{NI}}/M_{\mathrm{D}} - 1)$ (dotted).

and recursion formulae. Since only $2N_{max}$ terms are included, the C–G coefficients can be pre-computed up to $j=2N_{max}$, $m=N_{max}$, $n=N_{max}$.

The second term in (13) involves the derivative $(P_j^0)'$. This may be obtained by considering the recurrence relation

$$(2j+1)P_j^0=(P_{j+1}^0)'-(P_{j-1}^0)', \tag{16}$$

to generate an expansion

$$(P_j^0)'=\begin{cases}\sum_{k=0}^{(j-1)/2}(4k+1)P_{2k}^0 & (j \text{ odd}),\\ \sum_{k=0}^{j/2-1}(4k+3)P_{2k+1}^0 & (j \text{ even}),\end{cases}$$

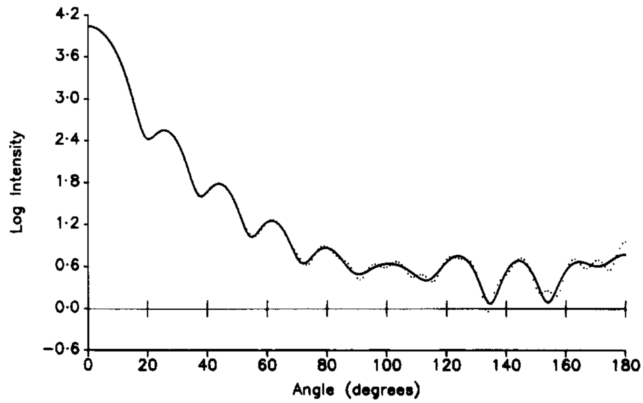
which allows the integral to be expressed as a summation over terms given by equation (14).

3. Results

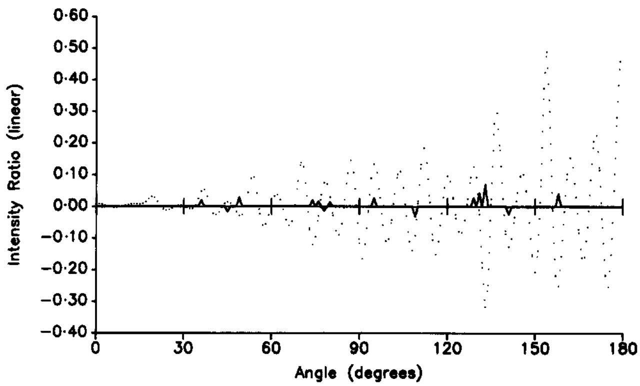
Two examples of the application of this method are given. In figure 1 (*a*) the Mie function is shown on a logarithmic scale for a water drop in air with size parameter $x=3$ and $n_p=1{\cdot}33+i\times 10^{-8}$. The function was obtained (i) from a Mie routine [3], (ii) by generating the first 20 ($N_{max}=10$) spherical harmonic coefficients $\{c_j\}$ from the C–G method, and summing the Legendre polynomials weighted accordingly, (iii) by generating the series $\{c_j\}$ by numerical integration using equation (7), on a one

Table 1. The coefficients of the Legendre polynomial expansion of the Mie function of figure 1 found by the Clebsh–Gordon method (equation (13) in the text), and by a quadrature approximation to equation (7). The right-hand figure in each column is the power of ten to scale by.

	C–G		Numerical integral	
Polynomial	Coefficient	Exponent	Coefficient	Exponent
0	3·945	00	3·946	00
1	9·270	00	9·271	00
2	1·080	01	1·080	01
3	9·131	00	9·131	00
4	5·487	00	5·486	00
5	2·184	00	2·185	00
6	6·881	−01	6·907	−01
7	1·583	−01	1·619	−01
8	2·729	−02	3·175	−02
9	3·638	−03	8·634	−03
10	3·863	−04	5·976	−03
11	3·352	−05	6·099	−03
12	2·421	−06	6·663	−03
13	1·477	−07	7·126	−03
14	7·676	−09	7·732	−03
15	3·404	−10	8·188	−03
16	1·279	−11	8·806	−03
17	3·976	−13	9·254	−03
18	1·014	−14	9·884	−03
19	−2·837	−15	1·032	−02



(a)



(b)

Figure 2. Mie scattering function, a comparison of methods. (*a*) The same as figure 1 (*a*) for a particle with refractive index 1·6, size parameter 10·3 in water. Here the difference between M_D (solid) and M_{NI} (dotted) is noticeable. (*b*) The ratio functions $(M_{CG}/M_D - 1) \times 10^4$ (solid) and $(M_{NI}/M_D - 1)$ (dotted).

degree quadrature interval. The differences are not noticeable in figure 1. In figure 1 (*b*) the difference from unity of the ratios between (i) and (ii) and between (i) and (iii) are shown. The former is scaled by 10^3 showing that the error is reduced by this order for the C–G method. The coefficients are tabulated in table 1. The C–G method gives zero coefficient above $j = 2N_{max}$ (by definition) whereas the numerical method gives terms higher than this.

Figure 2 shows the same results for a 1 µm radius polystyrene sphere ($n_p = 1{\cdot}6$) in water, irradiated by radiation of 783 nm. The size parameter is 10·3 and N_{max} is 21. Here the difference between (i) and (iii) gives a noticeable error in the function. In this case, for figure 2 (*b*), the C–G difference ratio was scaled by 10^4.

4. Discussion and conclusions

An exact expression for the expansion of the Mie kernel in spherical harmonics has been given. This formula may be used to generate the Mie function itself, but is more useful in applications requiring the integral of the function with spherical

Table 2. The coefficients of the Legendre polynomial expansion of the Mie function of figure 2 found by the Clebsh–Gordon method (equation (13) in the text), and by a quadrature approximation to equation (7). The right-hand figure in each column is the power of ten to scale by.

	C–G		Numerical integral	
Polynomial	Coefficient	Exponent	Coefficient	Exponent
0	1·044	02	1·046	02
1	2·885	02	2·893	02
2	4·415	02	4·427	02
3	5·570	02	5·585	02
4	6·400	02	6·417	02
5	6·972	02	6·990	02
6	7·350	02	7·367	02
7	7·540	02	7·554	02
8	7·615	02	7·626	02
9	7·564	02	7·571	02
10	7·408	02	7·411	02
11	7·153	02	7·150	02
12	6·803	02	6·796	02
13	6·323	02	6·312	02
14	5·762	02	5·747	02
15	5·082	02	5·066	02
16	4·264	02	4·250	02
17	3·445	02	3·434	02
18	2·502	02	2·499	02
19	1·577	02	1·583	02
20	8·585	01	8·742	01
21	3·512	01	3·753	01
22	1·304	01	1·593	01
23	4·133	00	7·324	00
24	1·142	00	4·536	00
25	2·797	−01	3·830	00
26	6·157	−02	3·763	00
27	1·232	−02	3·853	00
28	2·263	−03	3·990	00
29	3·837	−04	4·127	00
30	6·036	−05	4·275	00
31	8·837	−06	4·414	00
32	1·205	−06	4·563	00
33	1·528	−07	4·702	00
34	1·804	−08	4·852	00
35	1·974	−09	4·992	00
36	1·999	−10	5·143	00
37	1·633	−11	5·284	00
38	1·555	−12	5·436	00
39	−2·187	−12	5·577	00
40	7·625	−15	5·731	00
41	−2·420	−12	5·873	00

harmonics. The comparison with a numerical integration method as given in section 3, is of course dependent on the quadrature interval used. A very fine quadrature would give closer agreement. However this necessitates computing the Mie function itself at a finer resolution, with attendant space and time requirements. By contrast the C–G method requires only the coefficients a and b. An application which will be reported subsequently is the determination of the exact singular value spectrum of the Mie scattering kernel.

It must be noted that the use of equation (15) for deriving the C–G coefficients can equally well lead to numerical instability. The naive implementation used in the work reported here was stable up to size parameters of at least 11. More subtle formulae may be required for higher size parameters, but the analytic validity will still remain.

A program in C for evaluating Clebsh–Gordon coefficients and the Mie expansion is available from the author.

Acknowledgments

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Appendix

The Mie kernel contains terms in the products of two associated Legendre polynomials $P_j^1(\mu)$ up to $j = N_{max}$. Each P_j^1 is of the form $(1-\mu^2)^{1/2}Q(j-1)$ where $Q(n)$ is a polynomial of order n. Thus the maximum term in the function is a polynomial of order $2N_{max}$. But any polynomial order n is orthogonal to all Legendre polynomials of order greater than n [16]. Thus the series expansion of the Mie kernel contains terms up to $2N_{max}$ only.

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