Attenuation of light scattered by transparent particles

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We derive a simple integral form for the Mie cross section for an electromagnetic plane wave scattered by a transparent spherical particle. We show that this expression applies to any value of the diffraction parameter ka, and discuss the reasons for that behavior.

INTRODUCTION

One of the most frequently encountered phenomena in nature is the scattering of light by a spherical particle, as discussed by Mie.[1,2] It shows up in applications ranging from the rainbow and the glory right on through to astrophysics.[3] Scattering of this kind is also valuable as an analytical tool.[4]

The exact Mie formulae are multipole series of order l, a representation that could scarcely be less convenient in practical applications. They are therefore often replaced by simpler approximations, which are usually of somewhat restricted usefulness, and which tend to depend on the diffraction parameter $\alpha = ka$ (k here is the incident wave number and **a** is the particle radius).

For small α, the attenuation factor Q (or equivalently, the transverse scattering cross section in units of πa^2) is given by the Rayleigh formula

$$Q_R = \frac{8}{3} m \frac{(m^2-1)^2}{(m^2+2)} (ka)^4 \tag{1}$$

(for a transparent particle, the refractive index m relative to the ambient medium is a real quantity).

In the opposite limit $ka \gg 1$, van de Hulst's formula provides a better approximation[3]:

$$Q_H = 2\left[1 - 2\frac{\sin\delta}{\delta} + 2\frac{1-\cos\delta}{\delta^2}\right], \quad \delta = 2ka(m-1). \tag{2}$$

These expressions hold over rather different domains: Rayleigh's formula is very narrowly applicable ($ka \leqslant 0.5$), while van de Hulst's is somewhat broader ($ka \geqslant 5.0$). Unfortunately, these domains fail to overlap, and the intervening gap has been poorly bridged by a patchwork of largely inadequate approximations.

What we see here is an acute need for rediscussion of the results of Mie theory, and a search for more serviceable expressions for both the cross section Q and other physical quantities involving polarization, the forward- and back-scattering amplitudes, and so on.

2. SUMMARY OF REQUIRED FORMULAE

The transverse scattering (diffraction) cross section for an electromagnetic plane wave scattered by a spherical particle is given in general by[1–3]

$$Q = (2/\alpha^2) \sum_{l=1}^{\infty} (|a_l|^2 + |b_l|^2), \tag{3}$$

involving the partial amplitudes a_l and b_l,

$$a_l = (\mathrm{Re}\, g_l^E)/g_l^E, \quad b_l = (\mathrm{Re}\, g_l^M)/g_l^M. \tag{4}$$

These can be expressed in terms of Hankel and Bessel functions[5]

$$\zeta_l(z) = H^{(1)}_{l+1/2}(z)\sqrt{\frac{\pi z}{2}},$$

$$\psi_l(z) = \mathrm{Re}\, \zeta_l(z) = J_{l+1/2}(z)\sqrt{\frac{\pi z}{2}} \tag{5}$$

in the form

$$g_l^p = c^p \psi_l'(\beta)\zeta_l(\alpha) - d^p \psi_l(\beta)\zeta_l'(\alpha), \quad p = E, M, \tag{6}$$

$$c^E = d^M = \alpha, \quad c^M = d^E = \beta. \tag{7}$$

The arguments of these functions are the diffraction parameters

$$\alpha = ka, \quad \beta = mka, \tag{8}$$

where the diffractive index m normally depends on k.

One critical aspect of the following discussion is Perelman's[6] suggestion that the absolute value of g_l^p is independent of the multipole order and polarization,

$$|g_l^E|^2 \approx |g_l^M|^2 \approx K, \tag{9}$$

which reduces Q to the vastly simpler form

$$Q_P = \frac{2}{K\alpha^2} \sum_{l=1}^{\infty} (|\mathrm{Re}\, g_l^E|^2 + |\mathrm{Re}\, g_l^M|^2).$$

The simplification here consists in the fact that the cylindrical functions enter into Q polylinearly:

$$Q = \frac{2}{K\alpha^2} [(\alpha^2 + \beta^2)(A+B) - 4\alpha\beta C]. \tag{10}$$

Here

$$A = \sum_{l=1}^{\infty} (2l+1)\psi_l'(\beta)\psi_l(\alpha)\psi_l'(\beta)\psi_l(\alpha),$$

$$B = \sum_{l=1}^{\infty} (2l+1)\psi_l(\beta)\psi_l'(\alpha)\psi_l(\beta)\psi_l'(\alpha), \tag{11}$$

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$$C=\sum_{l=1}^{\infty}(2l+1)\psi_l'(\beta)\psi_l(\alpha)\psi_l(\beta)\psi_l'(\alpha).$$

The three quantities A, B, and C can all be obtained from the single function

$$S(x_1,x_2,x_3,x_4)=\sum_{l=1}^{\infty}(2l+1)\psi_l(x_1)\psi_1(x_2)\psi_1(x_3)\psi_1(x_4), \tag{12}$$

where $x_1=x_3=\beta$ and $x_2=x_4=\alpha$, by differentiating with respect to its arguments:

$$A=\partial^2S/\partial x_1\partial x_3,\quad B=\partial^2S/\partial x_2\partial x_4,\quad C=\partial^2S/\partial x_1\partial x_4. \tag{13}$$

We calculate the function $S(x)$ in Appendix A and obtain the following expression [see Eq. (A6)]:

$$S=x_1x_2x_3x_4\int_{-1}^{1}g(x_1,x_2|t)g(x_3,x_4|t)dt/2. \tag{14}$$

Differentiating, substituting A, B, and C into Eq. (10), and simplifying, we arrive at our basic formula

$$Q=(\beta^2/K)(\beta^2-\alpha^2)^2\int_{-1}^{1}dt(1+t^2)g^2(\omega), \tag{15}$$

in which

$$g(\omega)=(\omega\cos\omega-\sin\omega)/\omega^3 \tag{16}$$

and

$$\omega=\sqrt{\alpha^2+\beta^2-2t\alpha\beta}. \tag{17}$$

Henceforth, following Ref. 6, we put $K=\alpha\beta$.

The integral (15) can be reduced to a combination of elementary and transcendental functions (the cosine integral). Details of the calculation can be found in Appendix B; the final result is

$$Q=\frac{(m^2-1)^2}{4m^2}\Bigg[k_1H(R)-k_2H(\delta)+2m\left(1+4\frac{\cos R-\cos\delta}{R^2-\delta^2}\right)+\left(\frac{1}{2\alpha^2}-m^2-1\right)\int_\delta^R dt\frac{1-\cos t}{t}\Bigg], \tag{18}$$

where $H(z)$ denotes the van de Hulst function encountered above,

$$H(z)=1-2\frac{\sin z}{z}+2\frac{1-\cos z}{z^2}, \tag{19}$$

k_1 and k_2 are given in Appendix B, $R=2(m+1)\alpha$, and $\delta=2(m-1)\alpha$.

Equations (15) and (18) form the basis for subsequent developments.

3. SURVEY OF IMPORTANT APPROXIMATIONS

In the Introduction we cited two fundamental formulae:

Rayleigh ($ka\ll1$)—for a given wavelength, this corresponds to small scattering particles;

van de Hulst ($ka\gg1$)—corresponding to "large" particles (in the same sense).

Both of these follow from our basic formula (15). We take the van de Hulst formula first.

3.1. van de Hulst scattering, $R\gg\delta$

The foregoing relationship between R and δ enables us to keep just the second term in Eq. (18),

$$Q_H^5=f(m)H(\delta), \tag{20}$$

where

$$f(m)=-k_2(m^2-1)^2/4m^2=\frac{(m+1)^2}{8m^2}[2m(m^2+1)-(m^2-1)^2]. \tag{21}$$

One usually considers scattering by so-called "soft" particles, for which $m\approx1$, so that $f(m)=2$. We then obtain van de Hulst's formula (2) and its asymptotic value $Q(\infty)=2$, a result known as the blackbody paradox: the scattering cross section of short electromagnetic waves for an absorbing sphere is twice its geometrical size, notwithstanding the naïve result given by geometrical optics, which is assumed to apply here. The resolution of the paradox can be found in Refs. 2 and 7, and amounts to taking account of that part of the cross section attributable to the narrow diffractive region near the boundary of the scatterer.

At moderate values $m=1$, the van de Hulst approximation applies to $\delta\geqslant0.5$, as is readily apparent in Fig. 1 (see below).

3.2. Rayleigh scattering, $R\approx\delta\ll1$

In this case, the basic formula (15) can be expanded in powers of $\alpha=ka$. The leading term of the series yields

$$Q_R=(8/27)m(m^2-1)^2(ka)^4, \tag{22}$$

which is often cited as the Rayleigh formula. Here, however, the factor $(m^2+2)^{-2}$ in the original Rayleigh expression (1) is simply replaced by the number 9. There is a simple explanation for this: the Rayleigh calculation is carried out in the dipole approximation, which can be obtained from the Mie formula (3) by keeping only the first term, with $l=1$; Eq. (22), on the other hand, encompasses all multipoles (in the approximation $\alpha\ll1$), so that the role played by the first multipole, i.e., the dipole, is "smeared out."

A careful analysis shows that the denominator $(m^2+2)^{-2}$ cannot be obtained in the Perelman approximation, since it is incompatible with the basic assumption (9).

3.3. Intermediate scattering, Rayleigh–Gans formula

Scattering with $m\approx1$ was closely examined by Rayleigh[8] and later by Gans.[9] Taking $R\approx4\alpha$, $\delta\approx0$ in (18), we have

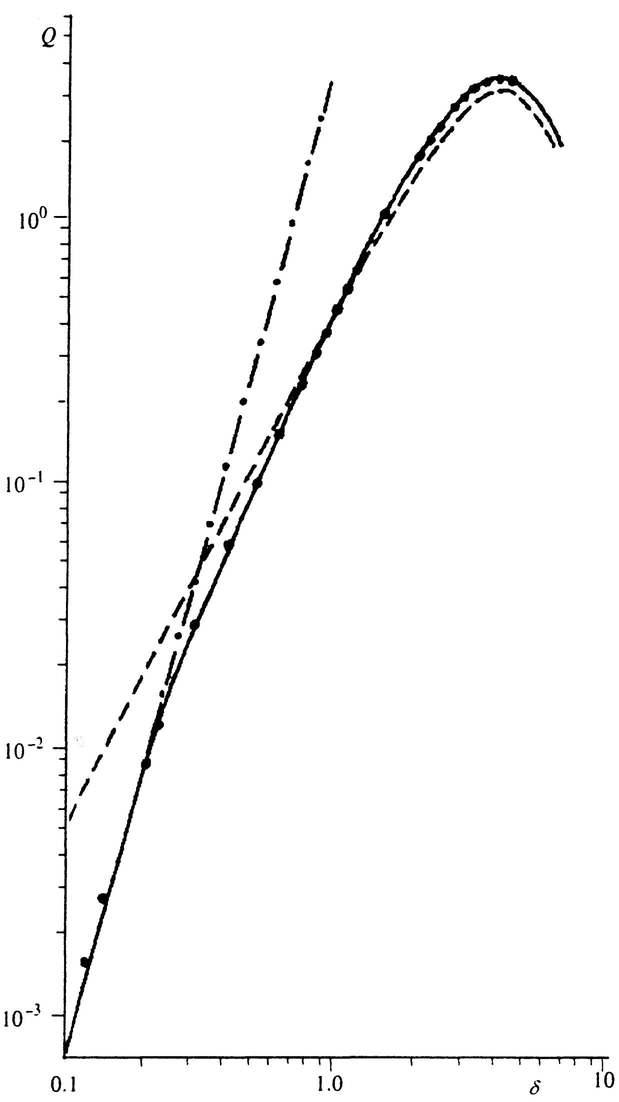

FIG. 1. The function $Q(\delta)$ from Mie theory (solid curve) in the Rayleigh (dot-dash) and van de Hulst (dashed) approximations. Data from the present paper are plotted as points. $Q_{as}=2.16$.

$$Q_{RG}=(m-1)^2\left[\frac{1}{2}H(R)-\lim_{m\to 1}k_2H(\delta)+2+8\frac{\cos R-1}{R^2}+\left(\frac{8}{R^2}-2\right)\int_0^R(1-\cos t)\frac{dt}{t}\right].$$

Substituting $\lim k_2H(\delta)=-R^2/8$ and Eq. (19) for $H(R)$, we obtain the Rayleigh–Gans formula,

$$Q_{RG}=(m-1)^2\left[\frac{5}{2}+7\frac{\cos R-1}{R^2}-\frac{\sin R}{R}+\frac{R^2}{8}+\left(\frac{8}{R^2}-2\right)\int_0^R(1-\cos t)\frac{dt}{t}\right]. \qquad (23)$$

3.4. Asymptotic behavior

At large values of α, the cosine integral can be replaced by its asymptotic value

$$\int_\delta^R(1-\cos t)\frac{dt}{t}=\ln\frac{R}{\delta}-\frac{\sin t}{t}\int_\delta^R+\int_\delta^R\sin t\frac{dt}{t}\approx\ln\frac{m+1}{m-1}; \qquad (24)$$

in addition, we can assume

$$H(R)\approx H(\delta)\approx 1. \qquad (25)$$

As a whole, Eq. (18) for the cross section goes asymptotically as

$$Q_{as}=\frac{m^2+1}{2m}\left[m^2+1-\frac{(m^2-1)^2}{2m}\ln\frac{m+1}{m-1}\right]. \qquad (26)$$

Notably, the aforementioned "blackbody" cross section $Q_{as}=2$ can be obtained from this result only for $m=1$!

4. DISCUSSION

The foregoing results can most conveniently be analyzed using Fig. 1, where they have been plotted as dots. There we show the dependence of the cross section on the phase shift $\delta=ka(m-1)$, as obtained in the van de Hulst anomalous diffraction approximation (dashed curve), the Rayleigh dipole approximation (dash-dot line), and as given by the exact Mie calculation, without the Perelman approximation (solid curve).

We carried out the calculations by integrating Eq. (15) numerically, using Eq. (18) as a check. All curves were calculated for a refractive index $m=1.1$, corresponding, for example, to oil droplets in water.

These data show that over the full range of δ, our results are in good agreement (to within 1–2%) with the exact Mie theory. In particular, the asymptotic values are the same as those given by the latter. This means that Eq. (15), which was derived for "soft" scatterers, is also reasonably accurate for so-called "hard" particles.

The other approximations are more limited:

a) the Rayleigh dipole approximation, Eq. (2), can only be used for $\delta<0.1$–0.2;

b) the improved van de Hulst approximation, Eq. (20), is suitable for $\delta>0.8$. The basic van de Hulst result, Eq. (2) without the factor $f(m)$, lies somewhat lower than the exact result;

c) the Rayleigh–Gans approximation can only be used with sufficiently "soft" particles, and in the range $\delta\approx 0.4$–0.5 it leads to appreciable errors (in this regard see also Ref. 10).

Thus, Eq. (15) turns out to be an equitable approximation to the Mie theory, and it is mathematically simple as well. We have therefore confirmed Perelman's[9] hypothesis, and this raises the question of the latter's meaning and justification. Perelman himself[6] does not adduce any solid arguments on this score, and merely cites the structure of the Wronskian. In our view, the most important aspect of his hypothesis is not the specific form of the constant K (which is highly likely not to be equal to $\alpha\beta$), but the feasibility of summing the entire Mie series.

The issue here is that the Rayleigh, Rayleigh–Gans, and van de Hulst approximations operate only on selected "segments" of that series, thereby overlooking properties inherent to the series as a whole. In particular, this would explain their inability to interpret the glory,[11] which results from an intricate interplay among contributions from the various terms of the Mie series.

While the theory of the glory is intimately related to the complex angular momentum method, the alternative—albeit approximate—possibility of summing the multipole series is extremely interesting, timely, and expedient.

Equation (15), which is approximate in name only, and not in spirit, can also be applied to polydisperse colloidal mixtures, in which the interpretation of the measured scattering cross sections for light is closely tied to the statistical distribution of scattering particle size; this is why standard analysis leads to such complicated expressions. The expressions developed here simplify such calculations significantly.

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APPENDIX A

To sum the Mie series in the Perelman approximation, it is convenient to make use of the addition theorem for Bessel functions[5] in the form[6]

$$\frac{\pi}{2\sqrt{x_1x_2}}\sum_{l=0}^{\infty}(2l+1)J_{l+1/2}(x_1)J_{l+1/2}(x_2)P_l(t)=\frac{\sin w_1}{w_1},$$

$$w_1=\sqrt{x_1^2+x_2^2-2x_1x_2t},$$

or in the notation of Eq. (5),

$$\sum_{l=0}^{\infty}(2l+1)\psi_l(x_1)\psi_l(x_2)P_l(t)=x_1x_2\frac{\sin w_1}{w_1}. \quad \text{(A1)}$$

Similarly, with the corresponding expression for w_2,

$$\sum_{l=0}^{\infty}(2l+1)\psi_l(x_3)\psi_l(x_4)P_l(t)=x_3x_4\frac{\sin w_2}{w_2}. \quad \text{(A2)}$$

If we multiply (A1) by (A2), integrate over t, and take advantage of the orthogonality of the Legendre polynomials

$$\int_{-1}^{1}P_l(t)P_m(t)dt=2\delta_{lm}/(2l+1), \quad \text{(A3)}$$

we obtain on the left-hand side the S function of (12) with an additional $l=0$ term in the sum over l, while the right-hand side gives the integral. Transferring the additional term to the right-hand side, we obtain

$$S(x_1,x_2,x_3,x_4)=\frac{1}{2}\int_{-1}^{1}\left[x_1x_2x_3x_4\frac{\sin w_1}{w_1}\frac{\sin w_2}{w_2}-\sin x_1\sin x_2\sin x_3\sin x_4\right]. \quad \text{(A4)}$$

Let

$$g(x_1,x_2|t)=\frac{\sin w_1}{w_1}-\frac{\sin x_1}{x_1}\frac{\sin x_2}{x_2}; \quad \text{(A5)}$$

we then obtain (14),

$$S(x_1,x_2,x_3,x_4)=x_1x_2x_3x_4\int_{-1}^{1}g(x_1,x_2|t)g(x_3,x_4|t)dt/2. \quad \text{(A6)}$$

Differentiating this expression, according to (13), yields A, B, and C. Replacing the arguments with α and β, we arrive at the basic formula (15).

APPENDIX B

Here we reduce Eq. (15) to the form (18). This transformation is accomplished by introducing the van de Hulst function (19) into the integrand,

$$Q=\frac{(m^2-1)^2}{4m}\int_{-1}^{1}dt[(1+t^2)/(\sigma-t)^2]\left[H(2\omega)-\frac{1-\cos 2\omega}{2}\right], \quad \text{(B1)}$$

where $\sigma=(m^2+1)/2m$, and, according to (17),

$$\omega=\alpha\sqrt{m^2+1-2mt}=\alpha\sqrt{2m}\sqrt{\sigma-t}. \quad \text{(B2)}$$

Instead of t, we integrate over the variable $z=2\omega$:

$$Q=4\alpha^2(m^2-1)^2\int_\delta^R dz\left[\frac{1+\sigma^2}{z^3}-\frac{\sigma}{4mz\alpha^2}+\frac{z}{(8m\alpha^2)^2}\right]\times\left[H(z)-\frac{1-\cos z}{2}\right]. \quad \text{(B3)}$$

All integrals can be expressed in terms of the H function, the cosine integral, and elementary functions, as is clear from the following:

$$\int\left[H(z)-\frac{1-\cos z}{2}\right]\frac{dz}{z^3}=-\frac{H(z)}{4z^2},$$

$$\int\left[H(z)-\frac{1-\cos z}{2}\right]\frac{dz}{z}=-\frac{H(z)}{2}+\frac{J(z)}{2}, \quad \text{(B4)}$$

$$\int\left[H(z)-\frac{1-\cos z}{2}\right]zdz=-z^2\frac{H(z)}{4}+2J(z)+2\cos z+\frac{z^2}{2}.$$

We denote the cosine integral[5] by $J(z)$:

$$J(z)=\int(1-\cos z)dz/z. \quad \text{(B5)}$$

After a great deal of simplification, Eq. (B3) takes the final form

$$Q=(\sigma^2-1)\left[k_1H(R)-k_2H(\delta)+2m+\frac{\cos R-\cos\delta}{2\alpha^2}+\left(\frac{1}{2\alpha^2}-m^2-1\right)\int_\delta^R dz\frac{1-\cos z}{z}\right], \quad \text{(B6)}$$

where

$$k_1 = m\left(\sigma - \frac{1}{\sigma+1}\right), \quad k_2 = m\left(\sigma - \frac{1}{\sigma-1}\right). \tag{B7}$$

Equation (B6) is the same as (18).

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