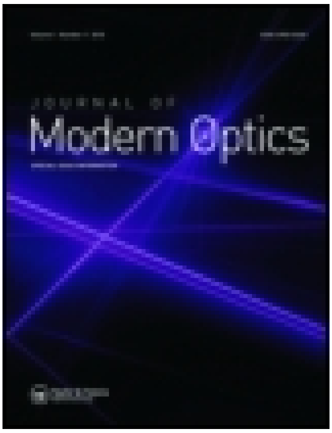

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On the Validity of the Method of Depolarization Factors an Analytical Study

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On the validity of the method of depolarization factors An analytical study

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Abstract. A comparison between two alternative scattering theories, the approximate method of depolarization factors and the exact Mie theory, is performed on a pure analytical basis for the case of spherical particles. It is shown that the usual assumption requiring that the radius be small compared with the interacting wavelength is insufficient to ensure the validity of the approximate method. The complete set of conditions are provided for both the scattered and absorbed fields.

1. Introduction

It is well known that the interaction of an ellipsoidal particle (small compared to wavelength) with an electromagnetic field can be described by the method of depolarization factors [1]—often known as the Rayleigh approximation in the case of spherical particles—which assimilates the particle to a dipole. In the case of ellipsoids, it is the only method which allows study of their scattering properties such as plasmon resonances [2] and interstellar light [3, 4], since an exact formulation is not available although some progress has been made [5]. However, the condition on size with respect to wavelength alone is insufficient to ensure the validity of the approximate method of depolarization factors. Moreover, the formulation based on integral solutions [6] requires in its most elementary approximation (the so-called Born approximation) a condition on the phase shift across the particle which involves the (real) refractive index. Kerker *et al.* [7] have performed a numerical investigation of this problem for the case of spherical particles which shows the influence of the complex index of refraction. In this paper, we present for the case of spherical particles, a full analytical comparison between the exact Mie theory and the approximate method of depolarization factors for both the scattered and the absorbed fields in the framework of electromagnetic optics. This is not possible in the context of the integral solution of the Helmholz equation since (i) an exact solution exists only for a medium of continuously varying (real) refractive index [8], and (ii) there exists only approximate solutions for small arbitrarily shaped particles with phase shift less than 2 resulting from an interative procedure [9].

In fact, an exact solution for a particle, i.e. a medium presenting a discontinuity of its complex index of refraction, requires the use of the mathematical theory of distributions.

We develop rigorous conditions for the validity of the approximate method and compare them with the Rayleigh–Gans–Born (RGB) approximation [6]. A few numerical examples for real materials illustrate the analytical results.

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2. The Rayleigh terms in Mie scattering theory

2.1. *Basic equations*

We shall consider an uncharged spherical particle of radius s in free space. Its material is supposed homogeneous, isotropic and non-magnetic. Its electrical properties are determined by its complex index of refraction $\hat{n}=n+ik$ (or its complex dielectric constant $\hat{\varepsilon}=\varepsilon_1+i\varepsilon_2$). The scattering of an incident monochromatic plane wave having linear polarization and wavelength λ is described by Maxwell's equations. Using the notation of Born and Wolf [10] and gaussian units for the expressions of the complex amplitude of the scattered electric field at a distance $r \gg \lambda$, in the spherical coordinates, we have

$$\mathscr{E}^s_\theta = -\frac{\lambda}{2\pi}\frac{\cos\phi}{r}\sum_{l=1}^{\infty}\left\{{}^{\mathrm{e}}B_l\zeta_l^{(1)'}\left(\frac{2\pi r}{\lambda}\right)P_l^{(1)'}(\cos\theta)\sin\theta - i\,{}^{\mathrm{m}}B_l\zeta_l^{(1)}\left(\frac{2\pi r}{\lambda}\right)P_l^{(1)}(\cos\theta)\frac{1}{\sin\theta}\right\} \quad (1)$$

for the transverse component parallel to the scattering plane, and

$$\mathscr{E}^s_\phi = -\frac{\lambda}{2\pi}\frac{\sin\phi}{r}\sum_{l=1}^{\infty}\left\{{}^{\mathrm{e}}B_l\zeta_l^{(1)'}\left(\frac{2\pi r}{\lambda}\right)P_l^{(1)}(\cos\theta)\frac{1}{\sin\theta} - i\,{}^{\mathrm{m}}B_l\zeta_l^{(1)}\left(\frac{2\pi r}{\lambda}\right)P_l^{(1)'}(\cos\theta)\sin\theta\right\} \quad (2)$$

for the transverse component normal to the scattering plane.

The radical component of the scattered electric field is given by

$$\mathscr{E}^s_r = \left(\frac{\lambda}{2\pi}\right)^2\frac{\cos\phi}{r^2}\sum_{l=1}^{\infty} l(l+1)\,{}^{\mathrm{e}}B_l\zeta_l^{(1)}\left(\frac{2\pi r}{\lambda}\right)P_l^{(1)}(\cos\theta)$$

and is always negligible under the condition $r \gg \lambda$. In the above formulae $P_l^{(1)}(x)$ are Legendre polymials. ${}^{\mathrm{e}}B_l$ and ${}^{\mathrm{m}}B_l$ can be expressed in terms of spherical Bessel and Hankel functions:

$$\psi_l(z)=\sqrt{\left(\frac{\pi z}{2}\right)}J_{l+1/2}(z), \quad \chi_l(z)=-\sqrt{\left(\frac{\pi z}{2}\right)}N_{l+1/2}(z), \quad \zeta_l^{(1)}(z)=\psi_l(z)-i\chi_l(z)$$

and their derivatives, where z is a complex variable,

$${}^{\mathrm{e}}B_l = i^{l+1}\frac{2l+1}{l(l+1)}\frac{\hat{n}\psi_l'(q)\psi_l(\hat{n}q)-\psi_l(q)\psi_l'(\hat{n}q)}{\hat{n}\zeta_l^{(1)'}(q)\psi_l(\hat{n}q)-\zeta_l^{(1)}(q)\psi_l'(\hat{n}q)},$$

$${}^{\mathrm{m}}B_l = i^{l+1}\frac{2l+1}{l(l+1)}\frac{\hat{n}\psi_l(q)\psi_l'(\hat{n}q)-\psi_l'(q)\psi_l(\hat{n}q)}{\hat{n}\zeta_l^{(1)}(q)\psi_l'(\hat{n}q)-\zeta_l^{(1)'}(q)\psi_l(\hat{n}q)},$$

where $q=2\pi s/\lambda$.

Using the power series expansions of Bessel and Neumann's functions, we obtain

$$\psi_l(z)=\frac{z^{l+1}}{1\times 3\times\ldots\times(2l+1)}S_1(z),$$

where

$$S_1(z)=1-\frac{z^2}{2(2l+3)}+\frac{z^4}{2\times 4(2l+3)(2l+5)}-\ldots,$$

$$\psi_l'(z)=\frac{(l+1)z^l}{1\times 3\times\ldots\times(2l+1)}S_1(z)-\frac{z^{l+2}}{1\times 3\times\ldots\times(2l+1)(2l+3)}S_2(z),$$

where

$$S_2(z)=1-\frac{z^2}{2(2l+5)}+\frac{z^4}{2\times 4(2l+5)(2l+7)}-\dots,$$

$$\zeta_l(z)=\frac{z^{l+1}}{1\times 3\times\dots\times(2l+1)}S_1(z)-i\frac{1\times 3\times\dots\times(2l-1)}{z^l}S_3(z),$$

where

$$S_3(z)=1-\frac{z^2}{2(-2l+1)}+\frac{z^4}{2\times 4(-2l+1)(-2l+3)}-\dots,$$

$$\zeta_l'(z)=\frac{(l+1)z^l}{1\times 3\times\dots\times(2l+1)}S_1(z)-\frac{z^{l+2}}{1\times 3\times\dots\times(2l+1)(2l+3)}S_2(z),$$
$$+il\frac{1\times 3\times\dots\times(2l-1)}{z^{l+1}}S_3(z)+i\frac{1\times 3\times\dots\times(2l-1)}{(-2l+1)z^{l-1}}S_4(z)$$

where

$$S_4(z)=1-\frac{z^2}{2(-2l+3)}+\frac{z^4}{2\times 4(-2l+3)(-2l+5)}\dots.$$

Each series S_1, S_2, S_3, S_4 may be bounded by geometric series whose sums are readily obtained:

$$S_1(z)<1+\frac{|z|^2}{2(2l+3)}+\left[\frac{|z|^2}{2(2l+3)}\right]^2+\dots=\frac{1}{1-\dfrac{|z|^2}{2(2l+3)}},$$

$$S_2(z)<1+\frac{|z|^2}{2(2l+5)}+\left[\frac{|z|^2}{2(2l+5)}\right]^2+\dots=\frac{1}{1-\dfrac{|z|^2}{2(2l+5)}},$$

$$S_3(z)<1+\frac{|z|^2}{2}+\left[\frac{|z|^2}{2}\right]^2+\dots=\frac{1}{1-\dfrac{|z|^2}{2}},$$

$$S_4(z)<1+\frac{|z|^2}{2}+\left[\frac{|z|^2}{2}\right]^2+\dots=\frac{1}{1-\dfrac{|z|^2}{2}}.$$

The final expressions for ${}^{\mathrm{e}}B_l$ and ${}^{\mathrm{m}}B_l$ are therefore,

$${}^{\mathrm{e}}B_l=\frac{i^{l+1}}{l(l+1)}\frac{q^{2l+1}}{[1\times 3\times\dots\times(2l-1)]^2}\frac{{}^{\mathrm{e}}N_l}{{}^{\mathrm{e}}D_l},\tag{3}$$

$${}^{\mathrm{m}}B_l=\frac{i^{l+1}}{l(l+1)}\frac{q^{2l+1}}{[1\times 3\times\dots\times(2l-1)]^2}\frac{{}^{\mathrm{m}}N_l}{{}^{\mathrm{m}}D_l},\tag{4}$$

where

$$^{e}N_l=(l+1)(\hat{n}^2-1)S_1(q)S_1(\hat{n}q)+\frac{(\hat{n}q)^2}{2l+3)}[S_1(q)S_2(\hat{n}q)-S_2(q)S_1(\hat{n}q)],$$

$$^{e}D_l=i\left\{(\hat{n}^2l+l+1)S_3(q)S_1(\hat{n}q)+(\hat{n}q)^2\left[\frac{S_4(q)S_1(\hat{n}q)}{-2l+1}-\frac{S_3(q)S_2(\hat{n}q)}{2l+3}\right]\right\}$$
$$+\frac{q^{2l+1}}{[1\times 3\ldots\times(2l-1)]^2(2l+1)}\,{}^{e}N_l,$$

$$^{m}N_l=\frac{q^2}{2l+3}[S_2(q)S_1(\hat{n}q)-\hat{n}^2S_1(q)S_2(\hat{n}q)],$$

$$^{m}D_l=-i\left\{(2l+1)S_3(q)S_1(\hat{n}q)+q^2\left[\frac{S_4(q)S_1(\hat{n}q)}{-2l+1}-\frac{\hat{n}^2S_3(q)S_2(\hat{n}q)}{2l+3}\right]\right\}$$
$$+\frac{q^{2l+1}}{[1\times 3\ldots\times(2l-1)]^2(2l+1)}\,{}^{m}N_l.$$

Since $r\gg\lambda$, we have

$$\zeta_l^{(1)}\left(\frac{2\pi r}{\lambda}\right)\simeq\exp\left[i\left(\frac{2\pi r}{\lambda}-\frac{\pi}{2}(l+1)\right)\right],$$

$$\zeta_l^{(1)\prime}\left(\frac{2\pi r}{\lambda}\right)\simeq-\exp\left[i\left(\frac{2\pi r}{\lambda}-\frac{\pi}{2}(l+2)\right)\right].$$

In the following, we shall always consider $q<1$ since we want to study the conditions for Rayleigh scattering in the general formalism of Mie.

2.2. *Analysis for the case when $|\hat{n}q|<1$ with $|\hat{n}|\lesssim 1$ and $q^2\ll 1$*

(*a*) General case: $\hat{n}^2l+l+1\neq 0$. The upper boundaries for S_1, S_2, S_3, S_4 allow us to simplify expressions (3) and (4). Then

$$^{e}B_l\simeq i^l\frac{q^{2l+1}}{l^2[1\times 3\times\ldots\times(2l-1)]^2}\,\frac{\hat{n}^2-1}{\hat{n}^2+\dfrac{l+1}{l}},\tag{5}$$

$$^{m}B_l\simeq i^l\frac{q^{2l+3}}{l(l+1)(1l+1)(2l+3)[1\times 3\times\ldots\times(2l-1)]^2}(\hat{n}^2-1).\tag{6}$$

The contribution to the total scattered field of the dipolar field corresponds to the terms $l=1$ in expressions (1) and (2); their components are given by

$$\mathscr{E}_\theta^{\mathrm{sd}}=\left[\left(\frac{2\pi}{\lambda}\right)^2s^3\,\frac{\hat{n}^2-1}{\hat{n}^2+2}\cos\theta+\left(\frac{2\pi}{\lambda}\right)^4\frac{s^5}{30}(\hat{n}^2-1)\right]\cos\phi\frac{\exp\left(i\dfrac{2\pi r}{\lambda}\right)}{r},$$

$$\mathscr{E}_\phi^{\mathrm{sd}}=-\left[\left(\frac{2\pi}{\lambda}\right)^2s^3\,\frac{\hat{n}^2-1}{\hat{n}^2+2}+\left(\frac{2\pi}{\lambda}\right)^4\frac{s^5}{30}(\hat{n}^2-1)\cos\theta\right]\sin\phi\frac{\exp\left(i\dfrac{2\pi r}{\lambda}\right)}{r}.$$

Since $q^2\ll 1$, these expressions may be simplified.

(i) When $\theta \neq \pm\pi/2$, the contribution of the magnetic wave is negligible in comparison with the electric wave ($|{}^{m}B_1| \ll |{}^{e}B_1|$:

$$\mathscr{E}_\theta^{sd} = \left(\frac{2\pi}{\lambda}\right)^2 s^3 \frac{\hat{n}^2-1}{\hat{n}^2+2} \cos\theta \cos\phi \frac{\exp\left(i\frac{2\pi r}{\lambda}\right)}{r}, \tag{7}$$

$$\mathscr{E}_\phi^{sd} = -\left(\frac{2\pi}{\lambda}\right)^2 s^3 \frac{\hat{n}^2-1}{\hat{n}^2+2} \sin\phi \frac{\exp\left(i\frac{2\pi r}{\lambda}\right)}{r}. \tag{8}$$

These are characteristic formulae for Rayleigh scattering as they present a λ^{-2} dependence upon wavelength.

The RGB condition $2q(n-1) \ll 1$ if $\hat{n}=n$ is real or $2q|\hat{n}-1| \ll 1$ if $\hat{n}$ is complex is in general more restrictive for the size of the particle than the conditions given at the beginning of this section. As an example, consider amorphous quartz at $\lambda = 0{\cdot}165$ μm with $n = 1{\cdot}67$ and $k = 4{\cdot}9 \times 10^{-6}$. Then $2q|\hat{n}-1| = 1{\cdot}34q$ and the RGB condition imposes $q \ll 1$ while the above conditions impose $q^2 \ll 1$ which is obviously less restrictive.

However, if following Van de Hulst [1], the additional condition $|\hat{n}-1| \ll 1$ is associated to the above RGB condition, then it is sufficient that $q < 1$ to ensure the inequality $2q|\hat{n}-1| \ll 1$ while the above conditions still require $q^2 \ll 1$. This wider range of possibilities for the size is obtained at the expense of a severe restriction upon the complex index of refraction.

(ii) When $\theta = \pm\pi/2$, there remains only the term for the magnetic wave in the expression of $\mathscr{E}_\theta^{sd}$ and it is non-Rayleigh. However, its contribution to the total scattered intensity is small in comparison with the term for the electric wave in $\mathscr{E}_\phi^{sd}$.

(*b*) $\hat{n}^2 l + l + 1 \simeq 0$. The approximate expression (6) remains valid while the expression for the electric coefficient (3) presents a resonance and can be written

$${}^{e}B_l \simeq i^l \frac{q^{2l-1}}{l^2[1\times 3\times \ldots \times (2l-1)]^2} \frac{\hat{n}^2-1}{2\hat{n}^2 \dfrac{2l+1}{l(2l+3)(-2l+1)}}.$$

The above condition is satisfied if

(i) $l=1$, $\hat{n}^2+2 \simeq 0$. This implies $n^2 \simeq 0$ and $k^2 \simeq 2$, then

$$\mathscr{E}_\theta^{sd} = \left[s\frac{5(\hat{n}^2-1)}{6\hat{n}^2}\cos\theta + \left(\frac{2\pi}{\lambda}\right)^4 \frac{s^5}{30}(\hat{n}^2-1)\right] \cos\phi \frac{\exp\left(i\frac{2\pi r}{\lambda}\right)}{r},$$

$$\mathscr{E}_\phi^{sd} = -\left[s\frac{5(\hat{n}^2-1)}{6\hat{n}^2} + \left(\frac{2\pi}{\lambda}\right)^4 \frac{s^5}{30}(\hat{n}^2-1)\cos\theta\right] \sin\phi \frac{\exp\left(i\frac{2\pi r}{\lambda}\right)}{r},$$

these components do not present a λ^{-2} dependence and hence the scattering is not of the Rayleigh type.

(ii) $l=2$, $2\hat{n}^2+3 \simeq 0$. This implies $n^2 \simeq 0$ and $k^2 \simeq 3/2$; the quadripolar term ($l=2$) presents a λ^{-2} dependence and contributes to the Rayleigh scattering along with the dipolar term ($l=1$) which remains unchanged; neglecting the magnetic dipolar wave as in the general case we find

$$\mathscr{E}_\theta^{sd}=\left(\frac{2\pi}{\lambda}\right)^2 s^3\left[\frac{\hat{n}^2-1}{\hat{n}^2+2}\cos\theta-\frac{7}{20}\frac{\hat{n}^2-1}{\hat{n}^2}\cos 2\theta\right]\cos\phi\frac{\exp\left(i\frac{2\pi\lambda}{r}\right)}{r},$$

$$\mathscr{E}_\phi^{sd}=\left(\frac{2\pi}{\lambda}\right)^2 s^3\left[-\frac{\hat{n}^2-1}{\hat{n}^2+2}+\frac{7}{20}\frac{\hat{n}^2-1}{\hat{n}^2}\cos\theta\right]\sin\phi\frac{\exp\left(i\frac{2\pi\lambda}{r}\right)}{r}.$$

(iii) If $l>2$, the resonance does not create new Rayleigh terms among the multipolar terms and the dipolar term ($l=1$) alone contributes to the Rayleigh scattering as in the general case (§ 2.2 (a))

2.3. *Analysis for the case when* $|\hat{n}|q \gtrsim 1$ *with* $|\hat{n}| \gg 1$ *and* $q^2 \ll 1$

The simplified expression (5) for ${}^{e}B_l$ remains valid since $q^2 \ll 1$. This is no longer the case for the expression of ${}^{m}B_l$; indeed, in the expression (4) for ${}^{m}B_l$,

$$|S_2(q)S_1(\hat{n}q)| \ll |\hat{n}^2 S_1(q)S_2(\hat{n}q)|,$$

while in the expression for ${}^{e}D_l$,

$$\left|\frac{S_4(q)S_1(\hat{n}q)}{-2l+1}\right| \ll \left|\frac{\hat{n}^2 S_3(q)S_2(\hat{n}q)}{2l+3}\right|.$$

Consequently, the term

$$\left|\frac{(\hat{n}q)^2 S_3(q)S_2(\hat{n}q)}{2l+3}\right|$$

is not longer negligible in comparison with the term

$$|(2l+1)S_3(q)S_1(\hat{n}q)|.$$

Therefore,

$${}^{m}B_l \simeq \frac{i^l}{l(l+1)}\frac{q^{2l+1}}{[1\times 3\times\ldots\times(2l-1)]^2}\frac{\frac{(\hat{n}q)^2}{2l+3}S_1(q)S_2(\hat{n}q)}{[(2l+1)S_3(q)S_1(\hat{n}q)-(\hat{n}q)^2 S_3(q)S_2(\hat{n}q)]}.$$

As in § 2, the contribution of the electric wave to the Rayleigh scattering is due to the dipolar term ($l=1$); the contribution of the magnetic wave is also due to the dipolar term provided that

$$(2l+1)|S_1(\hat{n}q)| \ll |(\hat{n}q)^2 S_2(\hat{n}q)|, \tag{9}$$

then

$${}^{m}B_l \simeq -i^l\frac{q^{2l+1}}{l(l+1)(2l+3)[1\times 3\times\ldots\times(2l-1)]^2}.$$

If the above condition (9) is not satisfied, the magnitude of the magnetic wave (in λ^{-4}) is of the same order of magnitude as that of the electric wave and the total scattering is not of the Rayleigh type.

When condition (9) is satisfied

$$\mathscr{E}_\theta^{\mathrm{sd}}=\left(\frac{2\pi}{\lambda}\right)^2 s^3\left[\frac{\hat{n}^2-1}{\hat{n}^2+2}\cos\theta-\frac{1}{10}\right]\cos\phi\frac{\exp\left(i\dfrac{2\pi r}{\lambda}\right)}{r},$$

$$\mathscr{E}_\phi^{\mathrm{sd}}=\left(\frac{2\pi}{\lambda}\right)^2 s^3\left[\frac{\hat{n}^2-1}{\hat{n}^2+2}-\frac{\cos}{10}\right]\sin\phi\frac{\exp\left(i\dfrac{2\pi r}{\lambda}\right)}{r}.$$

The multipolar components of the electric field beyond the dipolar term do not give Rayleigh scattering since their dependence upon λ has the form λ^{-m} with $m>2$.

3. The absorbed electric field in the Mie formalism

In a manner similar to § 2.1, we now express the components of the electric field at a point r located inside the grain. The radial component

$$\mathscr{E}_r^{\mathrm{w}}=\left(\frac{\lambda}{2\pi}\right)^2\frac{\cos\phi}{r^2}\frac{1}{\hat{n}^2}\sum_{l=1}^{\infty} l(l+1){}^{\mathrm{e}}A_l\psi_l\left(\frac{2\pi\hat{n}r}{\lambda}\right)P_l^{(1)}(\cos\theta); \tag{10}$$

the transverse component parallel to the scattering plane

$$\mathscr{E}_\theta^{\mathrm{w}}=-\frac{\lambda}{2\pi}\frac{\cos\phi}{r}\frac{1}{\hat{n}}\sum_{l=1}^{\infty}\left\{{}^{\mathrm{e}}A_l\psi_l'\left(\frac{2\pi\hat{n}r}{\lambda}\right)P_l^{(1)'}(\cos\theta)\sin\theta.\right.$$
$$\left.-i{}^{\mathrm{m}}A_l\psi_l\left(\frac{2\pi\hat{n}r}{\lambda}\right)P_l^{(1)}(\cos\theta)\frac{1}{\sin\theta}\right\}. \tag{11}$$

and the transverse component perpendicular to the scattering plane

$$\mathscr{E}_\phi^{\mathrm{w}}=-\frac{\lambda}{2\pi}\frac{\sin\phi}{r}\frac{1}{\hat{n}}\sum_{l=1}^{\infty}\left\{{}^{\mathrm{e}}A_l\psi_p'\left(\frac{2\pi\hat{n}r}{\lambda}\right)P_l^{(1)}(\cos\theta)\frac{1}{\sin\theta}\right.$$
$$\left.-i{}^{\mathrm{m}}A_l\psi_l\left(\frac{2\pi\hat{n}r}{\lambda}\right)P_l^{(1)'}(\cos\theta)\sin\theta\right\}, \tag{12}$$

where

$${}^{\mathrm{e}}A_l=i^{l+1}\frac{2l+1}{l(l+1)}\frac{-i\hat{n}}{\hat{n}\zeta_l'(q)\psi_l(\hat{n}q)-\zeta(q)\psi_l'(\hat{n}q)},$$

$${}^{\mathrm{m}}A_l=i^{l+1}\frac{2l+1}{l(l+1)}\frac{i\hat{n}}{\hat{n}\zeta_l(q)\psi_l'(\hat{n}q)-\zeta'(q)\psi_l(\hat{n}q)}.$$

Using the notation of § 2.1, we obtain

$${}^{\mathrm{e}}A_l=i^l\frac{(2l+1)^2}{l(l+1)\hat{n}^{l-1}}\frac{1}{{}^{\mathrm{e}}D_l},$$

$${}^{\mathrm{m}}A_l=-i^l\frac{(2l+1)^2}{l(l+1)\hat{n}l}\frac{1}{{}^{\mathrm{m}}D_l}.$$

In the case of the scattered electric field, the condition $q<1$ was independent of the condition $r\gg\lambda$ (this allowed us to neglect the radial component of the field). This is no longer true for the absorbed electric field since the condition $q<1$ implies $r<\lambda$

and consequently (*i*) the radial component of the field inside the grain is no longer negligible, and (*ii*) the (λ/r) dependence for the transverse components and the $(\lambda/r)^2$ dependence for the radial component suggest that, for $r \ll \lambda$, the former are negligible in comparison with the latter.

It is not possible to explain further this behaviour through an analysis of the coefficients ${}^{e}A_l$ and ${}^{m}A_l$ identical to that performed in §§ 2.2 and 2.3 since these coefficients affect the three components with approximately equal weight. However, the inequality $r < \lambda$ implies that the functions ψ_l and ψ'_l of $(2\pi\hat{n}r/\lambda)$ for $r \leqslant s$ assume values close to those taken by the coefficients ${}^{e}A_l$ and ${}^{m}A_l$, especially when $s \ll \lambda$.

If $|2\pi\hat{n}r/\lambda| \ll 1$, then $|2\pi\hat{n}r/\lambda| \ll l$ and we may write

$$\psi_l\left(\frac{2\pi\hat{n}r}{\lambda}\right) \simeq \frac{\left(\dfrac{2\pi\hat{n}r}{\lambda}\right)^{l+1}}{1 \times 3 \times \ldots \times (2l+1)}, \tag{13}$$

$$\psi'_l\left(\frac{2\pi\hat{n}r}{\lambda}\right) \simeq \frac{(l+1)\left(\dfrac{2\pi\hat{n}r}{\lambda}\right)^{l}}{1 \times 3 \times \ldots \times (2l+1)}. \tag{14}$$

Retaining the lowest order terms (r/λ), we obtain approximate expressions for the components of the absorbed field,

$$\mathscr{E}_r^{\mathrm{w}} = \frac{3 \cos\phi \sin\theta}{\hat{n}^2 + 2}, \tag{15}$$

$$\mathscr{E}_\theta^{\mathrm{w}} = \frac{3 \cos\phi \sin\theta}{\hat{n}^2 + 2}, \tag{16}$$

$$\mathscr{E}_\phi^{\mathrm{w}} = \frac{3 \sin\phi}{\hat{n}^2 + 2}. \tag{17}$$

These components have the same order of magnitude. If the above condition $|2\pi\hat{n}r/\lambda| \ll l$ is not satisfied, expressions (13) and (14) are no longer valid and higher order terms should be included in the expressions of the components (10), (11) and (12).

4. Rayleigh scattering described by the depolarization factors method

This alternative method allows us to simplify the analysis of the general problem of the scattering of a plane electromagnetic wave by a particle, generally of ellipsoidal shape. As in electrostatics, a relationship may be written between the complex amplitudes of the polarization $\mathscr{P}$ of the particle and of the incident field $\mathscr{E}^{\mathrm{i}}$:

$$\mathscr{P} = \frac{\chi(\nu)}{1 + 4\pi\chi(\nu) f} \mathscr{E}^{\mathrm{i}}, \tag{18}$$

where $\chi(\nu)$, the electrical suceptibility, is a function of frequency $\nu(=\omega/2\pi)$, provided that the incident field may be considered uniform throughout the particle at any time. f is a diagonal matrix whose components along the principal axes of the ellipsoid satisfy

$$f_x + f_y + f_z = 1.$$

The three quantities $4\pi f_x$, $4\pi f_y$ and $4\pi f_z$ are defined as the depolarization factors.

Considering now the case of a spherical particle ($f_x = f_y = f_z = 1/3$), this requires $s \ll \lambda/2n$ and $k \simeq 0$; these conditions are identical with those set in § 2.2. The suceptibility is simply $\chi = (\hat{\varepsilon} - 1)/4\pi$ and

$$\mathscr{P} = \frac{1}{4\pi} \frac{\hat{\varepsilon} - 1}{1 + (\hat{\varepsilon} - 1)/3} \mathscr{E}^{\mathrm{i}}.$$

As we only consider a first-order vectorial relationship (18), the scattered field is the same as that radiated by an oscillating dipole of instantaneous dipolar moment p, oscillating parallel to the direction of the incident field.

$$\mathbf{E}^{\mathrm{s}} = -\left(\frac{\omega}{c}\right)^2 \frac{1}{r^3} \mathbf{r} \wedge (\mathbf{r} \wedge \mathbf{p}),$$

where $\mathbf{E}^{\mathrm{s}}$ is the real part of $\mathscr{E}^{\mathrm{s}} \exp(-i\omega t)$,

$$p = s^3 \frac{\hat{n}^2 - 1}{\hat{n}^2 + 2} \mathbf{E}^{\mathrm{i}}$$

and $\mathbf{E}^{\mathrm{i}}$ is the real part of $\mathscr{E}^{\mathrm{i}} \exp(-i\omega t)$. This yields:

$$\mathscr{E}_r^{\mathrm{s}} = 0,$$

$$\mathscr{E}_\theta^{\mathrm{s}} = \left(\frac{2\pi}{\lambda}\right)^2 s^3 \frac{\hat{n}^2 - 1}{\hat{n}^2 + 2} \cos\theta \cos\phi \frac{\exp\left(i\dfrac{2\pi r}{\lambda}\right)}{r},$$

$$\mathscr{E}_\phi^{\mathrm{s}} = -\left(\frac{2\pi}{\lambda}\right)^2 s^3 \frac{\hat{n}^2 - 1}{\hat{n}^2 + 2} \sin\phi \frac{\exp\left(i\dfrac{2\pi r}{\lambda}\right)}{r},$$

which are identical with expressions (7) and (8). The approximate method of depolarization factors is therefore strictly equivalent to the case § 2.2 (*a*) of the Mie scattering theory.

Turning now to the absorbed electric field, we have the following expressions [8] under the conditions of § 3:

$$\mathscr{E}^{\mathrm{w}} = \frac{3}{\hat{n}^2 + 2} \mathscr{E}^{\mathrm{i}},$$

yielding

$$\mathscr{E}_r^{\mathrm{w}} = \frac{3 \sin\theta \cos\phi}{\hat{n}^2 + 2} \exp\left(i\frac{2\pi r}{\lambda} \cos\theta\right),$$

$$\mathscr{E}_\theta^{\mathrm{w}} = \frac{3 \cos\theta \cos\phi}{\hat{n}^2 + 2} \exp\left(i\frac{2\pi r}{\lambda} \cos\theta\right),$$

$$\mathscr{E}_\phi^{\mathrm{w}} = \frac{3 \sin\phi}{\hat{n}^2 + 2} \exp\left(i\frac{2\pi r}{\lambda} \cos\theta\right).$$

Since $r \ll \lambda$, $\exp(i2\pi r \cos\theta/\lambda) \simeq 1$ and the above expressions are similar to equations (15), (16) and (17) obtained in § 3.

5. Numerical examples

We now illustrate through some numerical examples, the deviation of the depolarization factors method from Mie theory. We calculated the scattering cross-section Q^s and the absorption cross-section Q^w for spherical grains of various materials of known optical properties (see the table) and plotted the relative differences

$$\frac{Q_{\mathrm{Depolarization}} - Q_{\mathrm{Mie}}}{Q_{\mathrm{Mie}}}$$

as a function of q in figures 1, 2, 3 and 4.

These particular examples have been chosen so as to illustrate the various cases delineated in the theoretical treatment.

(i) Amorphous quartz (figure 1). This is a typical example where the depolarization factors method gives an excellent accuracy for the scattering cross-section even for q close to 1 (figure 1). This is an agreement with the results in §§ 2.2 (a) and 4 since

Materials and complex indices of refraction for the numerical examples.

Material	$\lambda(\mu\mathrm{m})$	n	k
Amorphous quartz	0·165	1·67	$4{\cdot}9 \times 10^{-6}$
Lithium	0·306	0·346	1·21
Lithium	8·266	0·366	38
Iron	18	18·91	49

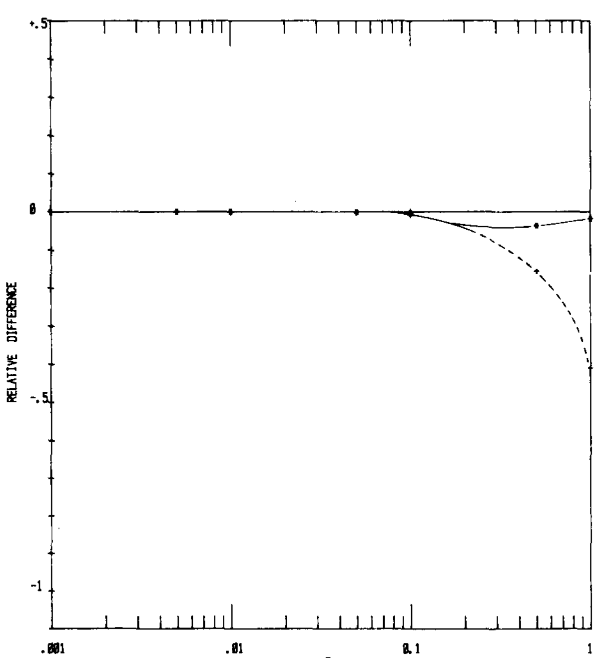

Figure 1. The relative difference between the prediction of the approximate formulation of depolarization factors and the exact Mie formulation for scattering (solid line) and absorption (broken line) cross-sections for amorphous quartz spheres at $\lambda = 0{\cdot}165\ \mu$m versus $q = 2\pi s/\lambda$.

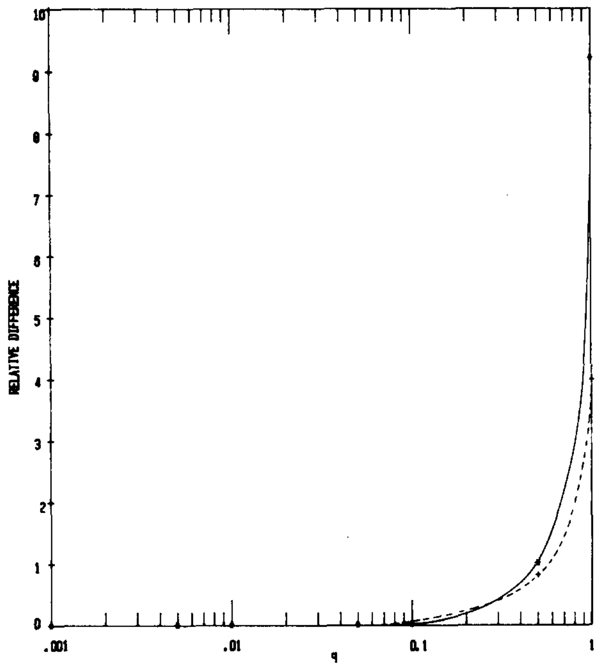

Figure 2. Same as figure 1 for lithium spheres at $\lambda = 0{\cdot}306\,\mu$m.

$|\hat{n}| \simeq 1$ with $k \simeq 0$. This is not the case for the absorption cross-section when $q > 0{\cdot}1$ since terms of order $l \geqslant 2$ are no longer negligible in the expressions (10), (11) and (12) and the condition $|2\pi\hat{n}r/\lambda| \ll l$ is not satisfied for all points inside the grain for $l \geqslant 1$.

(ii) Lithium (figure 2). We have $|\hat{n}| \simeq 1$ but k is no longer close to 0 and the approximated method (§4) is not valid. Furthermore, the value of k is close to the values giving resonances described in §2.2 (*b*) and which partly affect the absorbed field. In the case of the exact Mie formulation, the magnitude of the resonance is damped by the presence of multipolar terms. The larger negative deviation for Q^{w} found in the case of amorphous quartz still exists here and it modulates the resonance so that the difference for Q^{w} is less than that for Q^{s} (figure 2).

(iii) The final examples for lithium (figure 3) and iron (figure 4) clearly shows the large deviations even for low values of q when the conditions on the complex refractive index are not satisfied. For Q^{s}, the growing negative difference as q increases is due to the magnetic wave which becomes of the same order as the electric wave. The sudden reversal as q approaches 1 is a consequence of the appearance of multipolar terms which efficiently counteract the above trend.

6. Conclusion

An analytical study of the formulations of Mie theory and the depolarization factors method has allowed us rigorously to compare these two methods and to determine the conditions of validity of the approximated method. We now summarize our main conclusions.

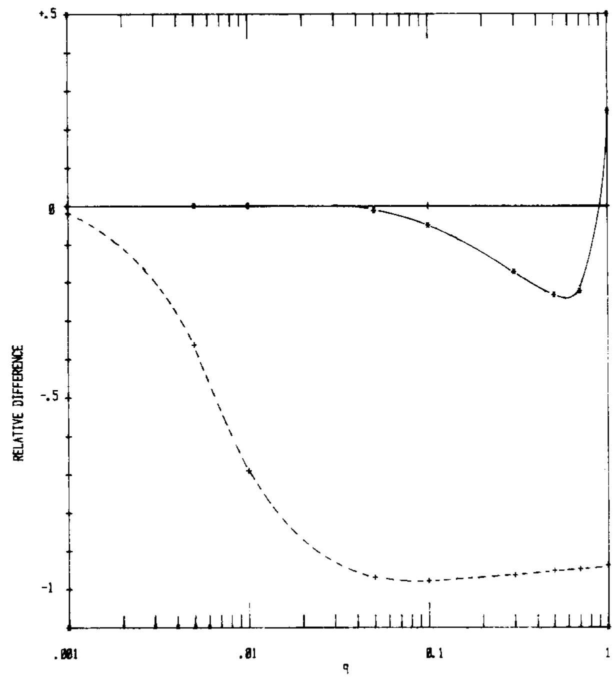

Figure 3. Same as figure 1 for lithium spheres at $\lambda = 8{\cdot}266\,\mu m$.

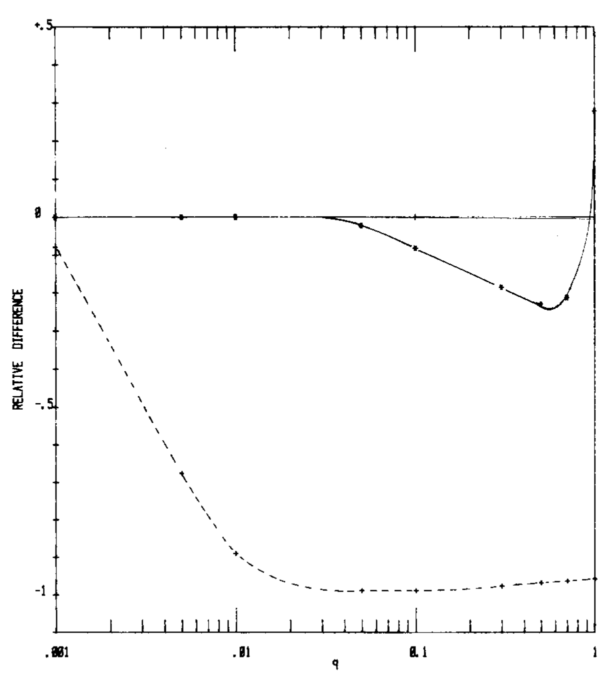

Figure 4. Same as figure 1 for iron spheres at $\lambda = 18\,\mu m$.

(i) *Diffracted field*

The approximate method of depolarization factors gives accurate results even for $q \simeq 1$ under the following conditions:

$$|\hat{n}|q < 1 \quad \text{with} \quad |\hat{n}| \lesssim 1 \quad \text{and} \quad q^2 \ll 1,$$

$$\hat{n}^2 l + l + 1 \neq 0 \quad \text{with} \quad k \simeq 0.$$

These conditions are in general less restrictive than those required by the RGB approximation which is a *sufficient* condition only [6], except when $|\hat{n}-1| \ll 1$.

(ii) *Absorbed field*

The approximate method gives accurate results under the sole restriction that $|\hat{n}|q \ll 1$.

Une comparaison entre deux théories de la diffusion, la méthode approchée des facteurs de dépolarisation et la théorie exacte de Mie, est effectuée sur une base purement analytique pour le cas de particules sphériques. On montre que l'hypothèse usuelle, impliquant que le rayon soit petit devant la longueur d'onde d'interaction, est insuffisante pour assurer la validité de la méthode approchée. L'ensemble complet des conditions requises est fourni pour le champ diffusé et le champ absorbé.

Ein Vergleich zwischen zwei alternativen Streutheorien, dem Approximationsverfahren der Depolarisationsfaktoren und der exakten Mietheorie, wird auf rein analytischer Basis für sphärische Teilchen durchgeführt. Es wird gezeigt, daß die übliche Forderung nach im Vergleich zur Wellenlänge kleinem Teilchenradius für die Gültigkeit des Approximationsverfahrens nicht hinreicht. Es wird der vollständige Satz von Bedingungen für die gestreuten als auch für die absorbierten Felder angegeben.

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