Mie-scattering formalism for spherical particles embedded in an absorbing medium

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Most of the Mie-scattering calculations have been done for a particle embedded in a nonabsorbing host medium. Generalization to an absorbing host medium can be achieved (a) by modifying the calculation of the spherical Bessel functions to account for a complex argument and (b) by accounting properly for the net rate of incident, scattered, and absorbed energy. We present an extended formalism of Mie scattering for the case of an absorbing host medium. Numerical calculations show that for a large spherical particle embedded in an absorbing host medium the extinction efficiency approaches 1 compared with 2 for a nonabsorbing host medium. We conjecture that this difference is due to the suppression of diffraction when the radius of the sphere is large. © 2001 Optical Society of America

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1. INTRODUCTION

Scattering of electromagnetic waves by a spherical particle embedded in a vacuum or in a homogeneous nonabsorbing medium is described by Mie-scattering formalism.[1–3] The exact solution is given by an expansion in vector spherical harmonics. Several authors have attempted to generalize the Mie theory for the case of a spherical particle embedded in an absorbing medium.[4–7] Mundy *et al.*[5] and Chylek[4] extended the Mie theory to take into account the effect of an absorbing medium. However, in their formalism they used a far-field approximation of the Mie solution, and they also integrated the radiative fluxes over a large sphere that included both the particle and the absorbing medium. Their approach was further modified by Quinten and Rostalski,[6] who did not use the far-field approximation. In these versions of the Mie-scattering generalization, the extinction is not a well-defined quantity owing to the integration over the surface of a large sphere concentric to the particle. As a result, the extinction is a function of the radius of the integrating sphere considered.

In this paper we calculate the extinction and other scattering characteristics of interest by integration over the surface of the spherical particle itself. This approach, similar to that proposed by Lebedev *et al.*,[7] removes the ambiguity in the definition of the extinction and provides methods useful for practical applications. The computer time required for numerical calculations in the case of an absorbing host medium is approximately the same as for the case of a nonabsorbing medium.

2. BACKGROUND THEORY

Formally, the generalized Mie solution is obtained by replacing the real size parameter x with a complex number. The form of the partial wave coefficients (a_n, b_n, c_n, and d_n) remains unchanged. The only modification is that all spherical Bessel functions and size parameters are now complex quantities.

A. Energy Scattered and Absorbed by a Sphere

1. *Scattered Energy*

Let us consider an incident plane wave propagating in the direction of the positive z axis with electric and magnetic field vectors $\mathbf{E}_i$ and $\mathbf{H}_i$ in an absorbing host medium. The scattered field is denoted $\mathbf{E}_{\text{sca}}$ and $\mathbf{H}_{\text{sca}}$. Fields outside the sphere are the sum of the scattered and incident fields:

$$\mathbf{E} = \mathbf{E}_i + \mathbf{E}_{\text{sca}},$$

$$\mathbf{H} = \mathbf{H}_i + \mathbf{H}_{\text{sca}}. \tag{1}$$

All fields are expanded in vector spherical harmonics.[1] The expansion coefficients a_n and b_n of the scattered wave and c_n and d_n of the field inside the particle are obtained by matching the boundary conditions on the surface of the sphere. The expansion coefficients a_n and b_n are given by[1]

$$a_n = \frac{m_r \psi_n(m_r x)\psi_n'(x) - \psi_n(x)\psi_n'(m_r x)}{m_r \psi_n(m_r x)\xi_n'(x) - \xi_n(x)\psi_n'(m_r x)}, \tag{2}$$

$$b_n = \frac{\psi_n(m_r x)\psi_n'(x) - m_r \psi_n(x)\psi_n'(m_r x)}{\psi_n(m_r x)\xi_n'(x) - m_r \xi_n(x)\psi_n'(m_r x)}, \tag{3}$$

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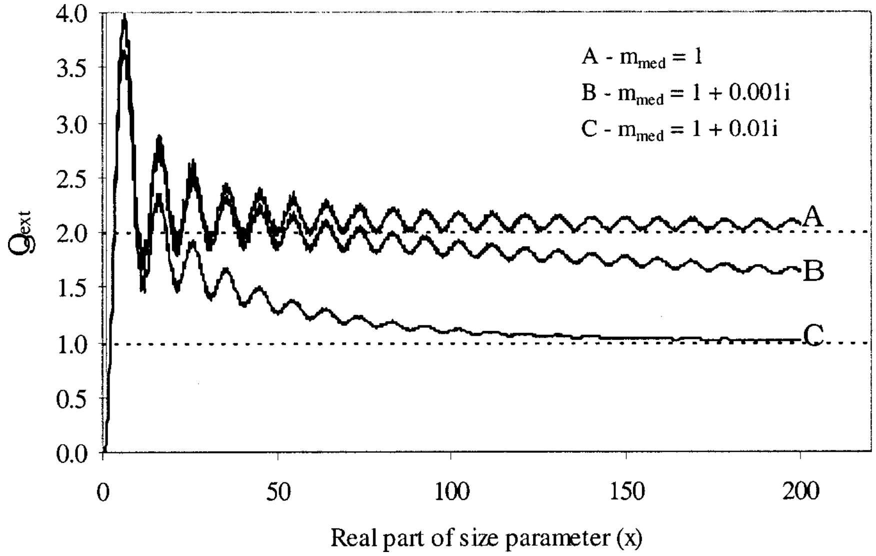

Fig. 1. Extinction efficiency (Q_{ext}) of a nonabsorbing sphere, with a refractive index $m_{\text{sphere}} = 1.33$, placed in a medium of a given refractive index (1; $1 + 0.01i$; $1 + 0.001i$) as a function of the real part of the size parameter x. The size parameter $x = 2\pi a m_{\text{med}}/\lambda$, where a is the radius of the sphere and λ is the wavelength of the considered radiation in vacuum.

where $\psi_n(\rho)$, $\psi_n'(\rho)$, $\xi_n(\rho)$, and $\xi_n'(\rho)$ are the Riccati–Bessel functions and their derivatives with respect to the argument ρ; m_r is the relative complex refractive index of a particle with respect to the host medium; and the size parameter $x = 2\pi a m_{\text{med}}/\lambda$, where a is the radius of the sphere, m_{med} is the refractive index of the host medium, and λ is the wavelength in vacuum. The Riccati–Bessel functions are defined as $\psi_n(\rho) = \rho j_n(\rho)$ and $\xi_n(\rho) = \rho(j_n(\rho) + iy(\rho))$, where $j_n(\rho)$ and $y_n(\rho)$ are the n-order spherical Bessel functions of the first and the second kind. The Riccati–Bessel functions and their derivatives are evaluated at a complex argument $\rho = x$ and $\rho = m_r x$.

The rate of the scattered energy is determined by the integral of the radial component of the Poynting vector over the surface of the sphere:

$$W_{\text{sca}} = \frac{1}{2}\text{Re}\int_{\text{sphere}} (\mathbf{E}_{\text{sca}} \times \mathbf{H}_{\text{sca}}^*) \cdot \hat{\mathbf{r}}\text{d}A, \tag{4}$$

where $\hat{\mathbf{r}}$ is the unit vector pointing in the direction of an outward normal to the surface of the sphere, Re means the real part, and the asterisk denotes the complex conjugate. After substituting for $\mathbf{E}_{\text{sca}}$ and $\mathbf{H}_{\text{sca}}$ and using the appropriate expansions in vector spherical harmonics, we obtain

$$W_{\text{sca}} = \frac{\pi|E_0|^2}{\mu\omega|k|^2}\text{Re}\Bigg\{k^*\sum_{n=1}^{\infty}(2n+1)[-i|a_n|^2\xi_n'(x)\xi_n^*(x) + i|b_n|^2\xi_n(x)\xi_n'^*(x)]\Bigg\}, \tag{5}$$

where E_0 is the amplitude of the electric field at $z = 0, \mu$ is the permeability of the host medium, ω is the angular frequency of the wave, and k is the wave number in the host medium.

2. Absorbed Energy

The rate of energy absorbed by the sphere is evaluated in a similar way:

$$W_{\text{abs}} = \frac{1}{2}\text{Re}\int_{\text{sphere}} (\mathbf{E} \times \mathbf{H}^*) \cdot \hat{\mathbf{r}}\text{d}A, \tag{6}$$

where $\mathbf{E} = \mathbf{E}_i + \mathbf{E}_{\text{sca}}$, $\mathbf{H} = \mathbf{H}_i + \mathbf{H}_{\text{sca}}$, and the integral is evaluated over the surface of a sphere. We obtain

$$\begin{aligned} W_{\text{abs}} = \frac{\pi|E_0|^2}{\mu\omega|k|^2}\text{Re}\Bigg\{k^*\sum_{n=1}^{\infty}(2n+1)[&i\psi_n^*(x)\psi_n'(x) \\ &- i\psi_n(x)\psi_n'^*(x) + ib_n\psi_n'^*(x)\xi_n(x) \\ &+ ib_n^*\psi_n(x)\xi_n'^*(x) + i|a_n|^2\xi_n'(x)\xi_n^*(x) \\ &- i|b_n|^2\xi_n(x)\xi_n'^*(x) - ia_n\psi_n^*(x)\xi_n'(x) \\ &- ia_n^*\psi_n'(x)\xi_n^*(x)]\Bigg\}. \end{aligned} \tag{7}$$

3. Extinction

Extinction is defined as the energy removed by the particle owing to absorption and scattering i.e., the sum of the scattered and the absorbed energy $W_{\text{sca}} + W_{\text{abs}}$.[1,2,4] In this definition, the energy absorbed by the host medium is not included. We have used the integrating sphere given by the surface of the particle to eliminate the energy absorbed by the host medium. This is convenient when we are interested in the extinction caused by the particle itself, not by the medium. The host medium causes the field to decay as it travels through the medium. Consequently, the host medium affects the amount of energy incident on the particle. This effect is accounted for in the calculation of an average intensity incident on the particle.

Adding Eq. (5) and Eq. (7) we obtain

$$W_{\text{ext}} = \frac{\pi |E_0|^2}{\mu\omega |k|^2} \operatorname{Re}\left\{ k^* \sum_{n=1}^{\infty} (2n + 1)[i\psi_n^*(x)\psi_n'(x) - i\psi_n(x)\psi_n'^*(x) + ib_n\psi_n'^*(x)\xi_n(x) + ib_n^*\psi_n(x)\xi_n'^*(x) - ia_n\psi_n^*(x)\xi_n'(x) - ia_n^*\psi_n'(x)\xi_n^*(x)] \right\}. \tag{8}$$

These results are equivalent to the expressions given by Lebedev *et al.*[7] and by Quinten and Rostalski[6] when the size of the integrating sphere is equal to the size of the particle. For a nonabsorbing medium, the scattering, absorption, and extinction reduce to the standard Mie-theory form.[1–3]

The physical meaning of the W_{abs}, W_{sca}, and W_{ext} is similar to that in the case of an ordinary Mie scattering. W_{abs} is the rate at which the energy is being absorbed by the particle, and W_{sca} is the rate at which the energy is being scattered through a spherical surface that is identical to the surface of the scattering particle. W_{ext} is the rate at which the energy is being removed from the beam owing to the absorption and scattering by the particle; it does not include the energy absorbed by the host medium itself.

B. Incident Intensity

As a consequence of an absorbing medium, the field incident on a sphere is different at different locations on the sphere. We need to calculate the total energy flux incident on the particle. This can be done by calculating the net energy that crosses the illuminated part of the sphere. We define an average incident intensity as the average energy flux crossing the cross-sectional area of the sphere, $I = W/(\pi a^2)$. Thus the average incident intensity is given by

$$I = \frac{1}{2\pi a^2} \operatorname{Re} \int_{\text{half sphere}} (\mathbf{E}_i \times \mathbf{H}_i^*) \cdot \hat{\mathbf{r}} \mathrm{d}A. \tag{9}$$

The integration is over the illuminated part of the sphere. We obtain

$$I = \frac{|E_0|^2}{\mu\omega} \operatorname{Re}\{k\} \left[\frac{\exp(2ak_I)}{2ak_I} + \frac{1 - \exp(2ak_I)}{(2ak_I)^2} \right], \tag{10}$$

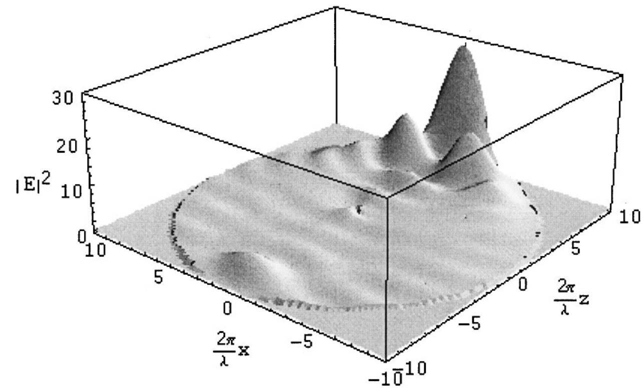

Fig. 2. Distribution of $|E|^2$ in an equatorial plane of a spherical particle with the real part of the size parameter $\operatorname{Re}(x) = 10$ ($x = 2\pi a m_{\text{med}}/\lambda$, a is the radius of sphere, and λ is the wavelength in vacuum) embedded in a nonabsorbing medium ($m_{\text{sphere}} = 1.33$ and $m_{\text{med}} = 1.0$). The spherical particle is illuminated by x-polarized electromagnetic plane waves propagating in the positive z direction.

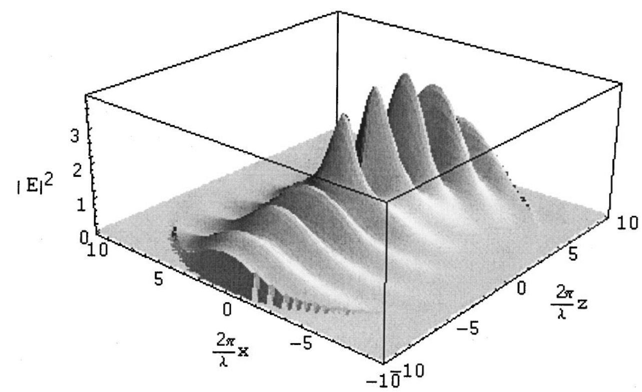

Fig. 3. Same as Fig. 2 except that the refractive index of the medium is $m_{\text{med}} = 1.0 + 0.5i$.

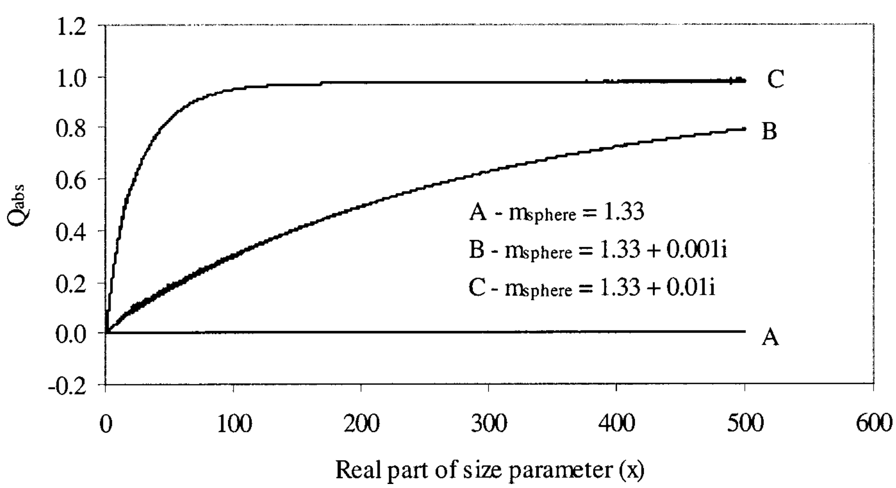

Fig. 4. Absorption efficiency (Q_{abs}) of a spherical particle with a given refractive index ($m_{\text{sphere}} = 1.33$; $1.33 + 0.01i$; $1.33 + 0.001i$) embedded in an absorbing medium ($m_{\text{med}} = 1 + 0.001i$) as a function of the real part of the size parameter x. A spherical particle with a real refractive index does not absorb any electromagnetic radiation even when its relative refractive index (with respect to the absorbing medium) is complex.

where k_I is the imaginary part of wave number k in the host medium. The cross sections are now obtained as $\sigma = W/I$ and the efficiencies are given by $Q = W/I\pi a^2$. For a nonabsorbing medium ($k_I = 0$) Eq. (10) reduces to the usual expression of intensity:[8]

$$I = \tfrac{1}{2}\epsilon c|E_0|^2. \tag{11}$$

This modification of an average intensity incident on a particle as far as we know has not been taken into account by previous authors (except by Mundy *et al.*[5]).

3. NUMERICAL RESULTS AND DISCUSSION

The effect of an absorbing medium on the extinction efficiency is shown in Fig. 1 (the particle itself is not absorbing). The interference[9,10] and the ripple[11,12] structures are significantly reduced with increasing absorption of the medium. With increasing radius, the extinction efficiency is approaching an asymptotic limit of 1. This is in contrast to the case of a nonabsorbing medium where the extinction is approaching a value of 2 (extinction paradox).[1] For a large spherical particle embedded in an absorbing medium, the incident radiation is absorbed by a medium before it can reach the edges of the sphere. Thus the diffraction contribution to the extinction is reduced, and in the asymptotic limit of the radius approaching infinity, there is no diffraction contribution to the extinction efficiency.

The distribution of $|E|^2$ in the equatorial plane of a spherical particle for the cases of a nonabsorbing and an absorbing host medium is shown in Figs. 2 and 3, respectively. The $|E|^2$ characterizes the energy density distribution within a particle. To emphasize the differences between the host media, we use a large imaginary part of the refractive index ($m_I = 0.5i$) for the absorbing host. The value of E_0 is chosen to be equal to 1 for the nonabsorbing medium and to $E_0 = \exp(-k_I a)$ for the absorbing medium. In this way the fields in front of the sphere at $z = -a$ are the same for the two cases. The strong peaks near the shadow part of a particle embedded in a nonabsorbing host (Fig. 2) are significantly reduced when the particle is placed in an absorbing host (Fig. 3). The wavelike structure of $|E|^2$ close to the surface of the particle (Fig. 2) is suppressed (Fig. 3).

For the case of a nonabsorbing sphere placed in an absorbing medium, the relative refractive index of the sphere is complex. Therefore one may expect that a particle embedded in an absorbing medium should absorb the radiation even if the particle by itself is nonabsorbing. This turns out, however, to be incorrect. The numerical calculations show (Fig. 4) that the absorption within a particle is zero when the refractive index of the particle is real.

The developed formalism may find some practical applications in cases of particles embedded inside absorbing solids or liquids. In atmospheric applications the absorption of the earth's atmosphere itself is too weak (even at strong water vapor or carbon dioxide absorption lines) to have any observable effect on the absorption and scattering of radiation by atmospheric aerosols or cloud droplets. This is true for both broadband and monochromatic radiation.

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