Mie theory for light scattering by a spherical particle in an absorbing medium

Qiang Fu and Wenbo Sun

Analytic equations are developed for the single-scattering properties of a spherical particle embedded in an absorbing medium, which include absorption, scattering, extinction efficiencies, the scattering phase function, and the asymmetry factor. We derive absorption and scattering efficiencies by using the near field at the surface of the particle, which avoids difficulty in obtaining the extinction based on the optical theorem when the far field is used. Computational results demonstrate that an absorbing medium significantly affects the scattering of light by a sphere. © 2001 Optical Society of America

1. Introduction

Light scattering and absorption by spherical particles in an absorbing medium occur in the earth–atmosphere system. Some examples are light scattering by cloud particles surrounded by water vapor in the atmosphere, by biological particles in the ocean, and by air bubbles in the ocean and sea ice. It is important for many applications to understand the effects of the absorbing medium on the scattering and absorption of light by the particles.

Light scattering by a spherical particle in a nonabsorbing medium is well understood based on Mie theory. When the host medium is absorbing, however, the Mie equations need to be modified for light scattering by particles to be described. Several studies have been carried out to develop a theoretical treatment of light scattering by particles embedded in absorbing media by using the far-field approximation. But, because of absorption in the host medium, extinction, scattering, and absorption efficiencies based on the far-field approximation depend on the radius of the conceptual integrating sphere. They do not represent the actual extinction, scattering, and absorption efficiencies of the particle.

In this paper, light scattering based on Mie theory is first formulated for a spherical particle in an absorbing medium. We then derive the single-scattering properties of the particle, including the absorption, scattering, and extinction efficiencies, by using the electromagnetic fields on the surface of the particle. This approach avoids difficulty in deriving the extinction from the optical theorem where the far field is used. Numerical results are presented to show the single-scattering properties for spherical particles in an absorbing medium. A wide range of cases is examined.

2. Formal Scattering Solution for a Sphere in an Absorbing Medium

The formal scattering solution for a sphere in a nonabsorbing medium based on Mie theory can be expressed in the form of equations that explicitly exhibit the refractive index of the medium as well as that of the sphere. These equations are formally identical to those obtained for the case of an absorbing medium, where both refractive indices can be complex. In this section the formal scattering solution is briefly reviewed by use of the notation of Bohren and Huffman.

Consider the scattering of a linearly polarized plane wave by a sphere with a radius \( a \) embedded in an absorbing medium. As shown in Fig. 1, we select the origin of a Cartesian coordinate system to be at the center of the sphere, with the positive \( z \) axis along the direction of propagation of the incident wave. The incident electric vector is polarized in the direction of the \( x \) axis. In the spherical coordinates, if the...
amplitude of the incident wave at the origin is $E_0$, the incident $i$, internal $t$, and scattered $s$ fields can be expressed in spherical harmonics in the form

$$E_i = \sum_{n=1}^{\infty} E_n[M_{01n}^{(1)} - iN_{01n}^{(1)}], \quad (1a)$$

$$H_i = -\frac{\mu}{\omega \mu_0} \sum_{n=1}^{\infty} E_n[M_{11n}^{(1)} + iN_{11n}^{(1)}], \quad (1b)$$

$$E_t = \sum_{n=1}^{\infty} E_n[c_nM_{01n}^{(1)} - id_nN_{01n}^{(1)}], \quad (2a)$$

$$H_t = -\frac{k}{\omega \mu_0} \sum_{n=1}^{\infty} E_n[d_nM_{11n}^{(1)} + ic_nN_{11n}^{(1)}], \quad (2b)$$

$$E_s = \sum_{n=1}^{\infty} E_n[i\alpha_nN_{10n}^{(3)} - b_nM_{01n}^{(3)}], \quad (3a)$$

$$H_s = \frac{k}{\omega \mu_0} \sum_{n=1}^{\infty} E_n[i\beta_nN_{01n}^{(3)} + a_nM_{10n}^{(3)}], \quad (3b)$$

where $i = \sqrt{-1}$, $E_n = \mathcal{E}^{n+1}/[n(n+1)]E_0$; $\omega$ is the angular frequency; $k = 2\pi n/\lambda_0$ and $k_t = 2\pi m_t/\lambda_0$, where $\lambda_0$ is the wavelength in vacuum and $m$ and $m_t$ are the refractive indices of the host medium and the scatterer, respectively; and $\mu$ and $\mu_t$ are the permeabilities of the host medium and the scatterer, respectively.

Fig. 1. Geometry for the scattering of a linearly polarized plane wave by a spherical particle of radius $a$. The origin of the coordinate system is at the particle center. The positive z-axis is along the direction of propagation of the incident wave with the electric vector polarized in the direction of the $x$ axis. The direction of the scattered light is defined by polar angles $\theta$ and $\phi$.

Fig. 2. Extinction, scattering, and absorption efficiencies and asymmetry factor as functions of size parameters for a spherical particle embedded in a medium. A refractive index of 1.0 is used for the particle. The real refractive index of the medium is 1.34, and the imaginary refractive index of the medium is 0.0, 0.001, 0.01, and 0.05.

The vector spherical harmonics in Eqs. (1)–(3) are given by

$$M_{01n} = \cos \phi \tau_n(cos \theta)z_n(r)\hat{e}_\phi - \sin \phi \tau_n(cos \theta)z_n(r)\hat{e}_\theta, \quad (4a)$$

$$M_{11n} = -\sin \phi \tau_n(cos \theta)z_n(r)\hat{e}_\phi - \cos \phi \tau_n(cos \theta)z_n(r)\hat{e}_\theta, \quad (4b)$$

$$N_{01n} = \sin n(n+1)\sin \theta \tau_n(cos \theta)z_n(r)/\rho \hat{e}_r, \quad (4c)$$

$$N_{11n} = \cos n(n+1)\sin \theta \tau_n(cos \theta)z_n(r)/\rho \hat{e}_r, \quad (4d)$$

where $\tau_n = P_n/\sin \theta$ and $\tau_n = dP_n/\sin \theta$ with $P_n$ the associated Legendre functions of the first kind of de-
gree \(n\) and order 1; \(\hat{e}_o, \hat{e}_s, \hat{e}_r\) are unit vectors in spherical coordinates; and \(\rho = kr\) for the incident and scattered fields and \(\rho = \frac{k}{2}r\) for the internal fields.

Superscripts to \(M\) and \(N\) in Eqs. (1)–(3) denote the kind of spherical Bessel function \(z_n\): (1) denotes \(j_n(\rho)\), which is defined as \([\pi/(2\rho)]^{1/2}J_{n+1/2}(\rho)\), where \(J_{n+1/2}\) is a Bessel function of the first kind; (3) denotes \(h_n^{(1)}(\rho)\), which is defined as \(j_n(\rho) + iy_n(\rho)\), where \(y_n(\rho) = [\pi/(2\rho)]^{1/2}Y_{n+1/2}(\rho)\) with \(Y_{n+1/2}\) is a Bessel function of the second kind.

Using the boundary conditions at the particle–medium interface, i.e., \(E_{\theta} + E_{\theta} = E_{\theta}\) and \(H_{\theta} + H_{\theta} = H_{\theta}\), we can derive the coefficients \(a_n, b_n, c_n,\) and \(d_n\) in Eqs. (2) and (3) in the form

\[
a_n = \frac{m \psi_n(\alpha) \psi_n(\beta) - m \xi_n(\alpha) \psi_n(\beta)}{m \xi_n(\alpha) \psi_n(\beta) - m \xi_n(\alpha) \psi_n(\beta)},
\]

\[
b_n = \frac{m \psi_n(\alpha) \psi_n(\beta) - m \xi_n(\alpha) \psi_n(\beta)}{m \xi_n(\alpha) \psi_n(\beta) - m \xi_n(\alpha) \psi_n(\beta)},
\]

\[
c_n = \frac{m \xi_n(\alpha) \psi_n(\alpha) - m \xi_n(\alpha) \psi_n(\beta)}{m \xi_n(\alpha) \psi_n(\beta) - m \xi_n(\alpha) \psi_n(\beta)},
\]

\[
d_n = \frac{m \xi_n(\alpha) \psi_n(\alpha) - m \xi_n(\alpha) \psi_n(\beta)}{m \xi_n(\alpha) \psi_n(\beta) - m \xi_n(\alpha) \psi_n(\beta)},
\]

where \(\alpha = k \rho, \beta = k \rho\), and the Riccati–Bessel functions \(\psi_n(\rho) = \rho j_n(\rho)\) and \(\xi_n(\rho) = \rho h_n^{(1)}(\rho)\). Note that in the derivation of Eq. (5) we take the permeability of the particle and the surrounding medium to be the same.

After \(a_n, b_n, c_n,\) and \(d_n\) are obtained, we can simulate the internal and the scattered fields by using Eqs. (2)–(4). Note that the Mie solution in Eqs. (2)–(5) is general. It can be used for light scattering by a sphere embedded in either an absorbing or a nonabsorbing medium. In the former case, \(m\) is complex.

### 3. Absorption, Scattering, and Extinction Efficiencies

For a nonabsorbing host medium the far-field approximation for the electromagnetic field is usually used to calculate the particle scattering and extinction cross sections. The far-field approach has also been used by Mundy et al.,\textsuperscript{2} Chylek,\textsuperscript{3} and Bohren and Gilr\textsuperscript{4} to study scattering and absorption by a spherical particle in an absorbing host medium. When the medium is absorbing, however, the scattering and extinction based on the far-field approximation depend on the
radius of the conceptual integrating sphere, which is concentric with the scatterer but with $r \gg a$. In this study we derive the absorption, scattering, and extinction efficiencies of a sphere embedded in an absorbing medium in such a way that these efficiencies depend only on the size of the particle and on the optical properties of the particle and the surrounding medium. We achieved this goal by using the near field at the surface of the scatterer. The analytic expressions for the absorption, scattering, and extinction efficiencies are presented.

The flow of energy and the direction of the electromagnetic wave propagation are represented by the Poynting vector

$$ S = \frac{1}{2} \text{Re}(\mathbf{E} \times \mathbf{H}^*), \quad (6) $$

where $\text{Re}(\cdot)$ represents the real part of the argument; the asterisk denotes the complex conjugate value; $\mathbf{E}$ and $\mathbf{H}$ are the total electric and magnetic vectors, respectively; and $|S|$ is in units of flux density.

Using the Poynting vector, we can express the rate of energy absorbed by the spherical particle $W_a$ as

$$ W_a = -\frac{1}{2} \text{Re} \int (\mathbf{E} \times \mathbf{H}^*) \cdot \hat{n} ds, \quad (7) $$

where the integral is taken over the surface of the scatterer and $\hat{n}$ is an outward unit vector normal to the surface of the scatterer. Following the boundary conditions of the electromagnetic fields across the particle surface $r = a$, we have

$$ (\mathbf{E} \times \mathbf{H}^*) \cdot \hat{n} = [ (\mathbf{E}_i + \mathbf{E}_s) \times (\mathbf{H}_i + \mathbf{H}_s)^* ] \cdot \hat{n} $$

$$ = (\mathbf{E}_s \times \mathbf{H}_s^*) \cdot \hat{n} \quad (8) $$

at the surface of the scatterer. Thus we can use either the external fields (the scattered plus incident wave) or the internal fields to evaluate $W_a$ from Eq.

Fig. 5. Scattering phase function as a function of the scattering angle for a sphere with a refractive index of 1.0 embedded in a medium with a real refractive index of 1.34 and an imaginary refractive index of 0.0, 0.001, 0.01, and 0.05. The results are presented for size parameters of 5, 25, and 100.

Fig. 6. As in Fig. 5, but the refractive index of the particle is 1.34 + 0.01i and the refractive index of the medium is 1.0 + imi, where $m_i = 0.0, 0.001, 0.01,$ and 0.05.
(7). Here we employ the latter because of its simplicity. Therefore $W_a$ can be written in the form

$$W_a = -\frac{1}{2} \Re \int \int (\mathbf{E}_l \times \mathbf{H}_l^*) \cdot \hat{n} ds$$

$$= \frac{1}{2} \Re \int_0^{2\pi} \int_0^\pi (E_{l0}H_{l0}^* - E_{l1}H_{l1}^*) a^2 \sin \theta d\theta d\phi,$$

where the internal electromagnetic vector components are obtained from Eqs. (2) and (4). Note that

$$\int_0^\pi (\pi_n \pi_n + \pi_n^* \pi_n) \sin \theta d\theta = 0, \quad (10)$$

$$\int_0^\pi (\pi_n \pi_n + \pi_n^* \pi_n) \sin \theta d\theta = \delta_{nn'} \frac{2n^2(n + 1)^2}{2n + 1}, \quad (11)$$

where $\delta_{nn'}$ is unity if $n = n'$ and zero otherwise. We can then derive $W_a$ in the form

$$W_a = \frac{\pi |E_0|^2}{\omega \mu} \sum_{n=1}^{\infty} (2n + 1) \Im(A_n), \quad (12a)$$

where $\Im()$ represents the imaginary part of the argument and

$$A_n = \frac{|c_n|^2 \psi_0(\beta)\psi_n(\beta) - |d_n|^2 \psi_0(\beta)\psi_n(\beta)}{k}. \quad (12b)$$

The rate of energy scattered by the scatterer $W_s$ is given by

$$W_s = \frac{1}{2} \Re \int \int (\mathbf{E}_s \times \mathbf{H}_s^*) \cdot \hat{n} ds$$

$$= \frac{1}{2} \Re \int_0^{2\pi} \int_0^\pi (E_{s0}H_{s0}^* - E_{s1}H_{s1}^*) a^2 \sin \theta d\theta d\phi.$$  

(13)

Substituting the scattered electromagnetic vector components from Eqs. (3) and (4) into Eq. (13) and using Eqs. (10) and (11) yield

$$W_s = \frac{\pi |E_0|^2}{\omega \mu} \sum_{n=1}^{\infty} (2n + 1) \Im(B_n), \quad (14a)$$

where

$$B_n = \frac{|a_n|^2 \xi_0^*(\alpha)\xi_n^*(\alpha) - |b_n|^2 \xi_0^*(\alpha)\xi_n^*(\alpha)}{k}. \quad (14b)$$

The rate of energy attenuated by the spherical particle is $W_e = W_a + W_s$, which can be written in the form

$$W_e = \frac{\pi |E_0|^2}{\omega} \sum_{n=1}^{\infty} (2n + 1) \Im\left(\frac{A_n}{\mu_t} + \frac{B_n}{\mu}\right). \quad (15)$$

Fig. 7. As in Fig. 5, but the refractive index of the particle is $1.4 + 0.05i$ and the refractive index of the medium is $1.2 + im$, where $m = 0.0, 0.001, 0.01,$ and $0.05$.

Following Mundy et al., the rate of energy incident on the scatterer in an absorbing medium is

$$f = \frac{2\pi a^2}{\eta} I_0 [1 + (\eta - 1)e^\eta], \quad (16)$$

where $\eta = 4\pi a / \lambda_0$ and $I_0 = |m_r/(2c\mu)| |E_0|^2$. Here $c$ is the speed of light in vacuum, and $m_r$ and $m_i$ are the real and the imaginary parts, respectively, of the complex refractive index of the host medium. It can be shown that $f = \pi a^2 I_0$ when $m_i = 0$. Therefore the absorption, scattering, and extinction efficiencies are

$$Q_a = W_a/f, \quad (17a)$$

$$Q_s = W_s/f, \quad (17b)$$

$$Q_e = W_e/f. \quad (17c)$$

Note that, although the results in this section are derived for a sphere embedded in an absorbing medium, these results are also valid for the nonabsorb-
ing situation. It can be shown, e.g., that $Q_s$ based on Eqs. (14), (16), and (17a) is

$$2/\alpha^2 \sum_{n=1}^{\infty} (2n + 1)(|a_n|^2 + |b_n|^2)$$

when $m$ is real, which is identical to that based on the far-field approximation in the nonabsorbing medium case. Here we have used $\xi_n = \psi_n - i\chi_n$ with $\chi_n = -\rho \gamma_n(p)$, and the functions $\psi_n$ and $\chi_n$ are real for the real argument so that $\chi_n \psi_n' - \psi_n \chi_n' = 1$.

When the surrounding medium is nonabsorbing, both scattering and extinction of a spherical particle are traditionally derived by use of the external fields. Then the absorption is obtained as the difference between the extinction and scattering. In this study it is the first time, to our knowledge, that the absorption has been directly derived by use of the internal fields, which leads to a concise analytic form of $W_a$.

Based on the far-field approximation, Mundy et al. defined the so-called unattenuated scattering and extinction efficiencies for a sphere in an absorbing medium, which are independent of the radius of the conceptual sphere. However, their unattenuated efficiencies do not represent the actual scattering and extinction efficiencies of the sphere. The same conclusion can be drawn for the extinction cross section defined by Bohren and Gilra based on the far-field approximation.

4. Scattering Phase Function and Asymmetry Factor
The scattering phase function represents the angular distribution of scattered energy at very large distances from the sphere, which can be derived by use of the far-field approximation. It is convenient to define amplitude functions $S_1$ and $S_2$ as in Ref. 6:

$$S_1 = \sum_{n=1}^{\infty} \frac{2n + 1}{n(n+1)} [a_n \tau_n(\cos \theta) + b_n \tau_n(\cos \theta)], \quad (18a)$$

$$S_2 = \sum_{n=1}^{\infty} \frac{2n + 1}{n(n+1)} [a_n \pi_n(\cos \theta) + b_n \pi_n(\cos \theta)]. \quad (18b)$$

For unpolarized incident light, since the scattered energy in the far-field is $I_s \propto |S_1|^2 + |S_2|^2$, the normalized scattering phase function can be derived as

$$P(\cos \theta) = \frac{|S_1|^2 + |S_2|^2}{\sum_{n=1}^{\infty} (2n + 1)(|a_n|^2 + |b_n|^2)}. \quad (19)$$

Another often-used parameter in the radiative-transfer calculation is the asymmetry factor, which is defined as

$$g = \frac{1}{2} \int_{-1}^{1} P(\cos \theta) \cos \theta \, d\cos \theta. \quad (20)$$

Using Eq. (19), we can also express the asymmetry factor in analytic form:

![Fig. 8. Electric energy density $|E|^2$ within an air bubble embedded in a medium with a refractive index of 1.34 + $m_i$, where $m_i = 0.0, 0.001, 0.01$, and 0.05. $|E|^2$ is in relative units with $E_0 = 1$. The fields are shown on a plane that is through the center of the sphere and parallel to the incident light. The positive z axis is along the direction of propagation of the incident wave with the electric vector parallel to the plane. The size parameter is 25.](20 March 2001 / Vol. 40, No. 9 / APPLIED OPTICS 1359)
5. Computational Results
A computer program has been developed based on the theory presented in Sections 2–4. A suggestion of Bohren and Huffman\(^5\) has been followed to obtain a numerically stable scheme for calculating \(a_n, b_n, c_n,\) and \(d_n\). For the case of a nonabsorbing medium the results from the new program are the same as those from the standard Mie code developed by Wiscombe.\(^7\) In this section we present the computational results to examine how an absorbing medium affects the scattering and absorption of light by a sphere.

We have considered three different sets of refractive indices for a sphere embedded in an absorbing medium. In the first set we used the refractive index of 1.0 for the sphere and a real refractive index of 1.34 with several values of the imaginary part including 0, 0.001, 0.01, and 0.05 for the surrounding medium. This can be considered the situation of an air bubble embedded in water or ice. For the second set we used \(m_r = 1.34 + 0.01i\) and \(m = 1.0 + im_i,\) where \(m_i = 0.0, 0.001, 0.01,\) and 0.05, which is the situation covered by Mundy et al.\(^2\)

Figures 2–4 show the single-scattering properties of a sphere embedded in an absorbing medium for the three sets of refractive indices, respectively, as functions of size parameter \(\frac{2\pi a}{\lambda_0}\). The single-scattering properties include extinction, scattering, and absorption efficiencies and asymmetry factors. It is well known that for a particle in a nonabsorbing medium, the extinction efficiency approaches two when the size parameter increases. However, from Figs. 2–4, we can see that \(Q_e\) approaches one for an absorbing medium. For the case of an air bubble (Fig. 2) we have \(Q_s = Q_e\) because \(Q_a = 0.\) For a sphere with absorption (Figs. 3 and 4) \(Q_s\) approaches one for nonabsorbing media but zero for absorbing media. Figures 3 and 4 also show that the absorbing medium has little effect on absorption efficiency. The effect of absorbing media on the asymmetry factor becomes significant only when \(Q_s\) is small.

Figures 5–7 show the scattering phase function when three different sets of refractive indices are used. It is obvious that the effect of absorbing media
on the angular distribution of the scattered energy becomes more significant for a larger size parameter.

Figures 8 and 9 show the electric energy density $|E|^2$ within an air bubble. The $|E|^2$ presented here are in relative units with $E_0 = 1$. The fields are shown on a plane that is through the center of the sphere and parallel to the incident light. In Fig. 8 the incident electric vector is parallel to the plane, whereas in Fig. 9 the incident electric vector is perpendicular to the plane. We can see that the internal field pattern is sensitive to $m_i$ of the surrounding medium.

6. Summary and Conclusions
Light scattering and absorption by a spherical particle in an absorbing medium have been formulated in this study. Using the electromagnetic fields at the particle surface, we have derived analytic expressions for the single-scattering properties of the particle, which include the absorption, scattering, and extinction efficiencies. In particular, the absorption efficiency is directly derived by using the internal field, which leads to a concise analytic formula of $Q_a$. Our approach here, when the near field is used, avoids difficulty in deriving the extinction based on the optical theorem when the far field is used for the case of absorbing media. In this study the equations for the scattering phase function and asymmetry are based on the far-field approximation.

Numerical simulations have been carried out to examine how an absorbing medium affects the scattering and absorption of light by a sphere. The main conclusions from the computational results for the case of absorbing media are as follows:

(1) The extinction efficiency approaches one as the size parameter increases.
(2) For an absorbing particle, the scattering efficiency approaches zero as the size parameter increases.
(3) The absorbing medium has little effect on absorption efficiency.
(4) The absorbing medium has a more significant effect on the scattering phase function for a larger size parameter.

We thank P. Chylek for useful discussions. The research contained here was supported by U.S. Department of Energy grant DE-FG03-00ER62931.

References
5. C. F. Bohren and D. R. Huffman, Absorption and Scattering of Light by Small Particles (Wiley, New York, 1983).