Phase function of a spherical particle when scattering an inhomogeneous electromagnetic plane wave

JEPPE REVALL FRISVAD[1,*]

[1] *Department of Applied Mathematics and Computer Science, Technical University of Denmark, Richard Petersens Plads, Building 321, DK-2800 Kongens Lyngby, Denmark*
[*] *Corresponding author: jerf@dtu.dk*

In absorbing media, electromagnetic plane waves are most often inhomogeneous. Existing solutions for the scattering of an inhomogeneous plane wave by a spherical particle provide no explicit expressions for the scattering components. In addition, current analytical solutions require evaluation of the complex hypergeometric function $_2F_1$ for every term of a series expansion. In this work, I develop a simpler solution based on associated Legendre functions with argument zero. It is similar to the solution for homogeneous plane waves but with new explicit expressions for the angular dependency of the far-field scattering components, that is, the phase function. I include recurrence formulae for practical evaluation and provide numerical examples to evaluate how well the new expressions match previous work in some limiting cases. The predicted difference in the scattering phase function due to inhomogeneity is not negligible for light entering an absorbing medium at an oblique angle. The presented theory could thus be useful for predicting scattering behavior in dye based random lasing and in solar cell absorption enhancement.

1. INTRODUCTION

Scattering of a light wave by a perfect sphere has been considered for more than a century. The formulae were originally derived by Lorenz [1], who considered light in a nonabsorbing medium scattered by a nonabsorbing sphere. He used the ratio $N = n'_{med}/n'_p$ of the real refractive index of the host medium to that of the spherical particle. Mie [2] derived the same formulae over again using Maxwell's equations. Mie was considering colloidal suspensions of metallic particles. Since metals have a very significant imaginary part in their index of refraction, he generalized the original formulae using a complex index of refraction $n_p = n'_p + in''_p$ for the spheres.

The theory of Lorenz is useful for modeling atmospheric phenomena such as skylight, clouds, mist, haze, and rainbows. Beyond this, the types of matter consisting of nonabsorbing particles in a nonabsorbing host medium seem rather few. Mie's extension of the theory was thus a considerable improvement in terms of materials that we are able to model. The theory of scattering by spherical particles is today called the Lorenz-Mie theory (or Mie theory). Kerker [3] gives an excellent historical review of the different contributions to the theory in its initial development (and of how little the different contributors knew of each other's work).

Even with the extension of Mie, the class of materials that we can model as possibly absorbing spheres in a nonabsorbing host is somewhat limited. Many particles are approximately spheres, but oftentimes they are embedded in an absorbing host. The blue color of the seas is caused by the weak absorption of water. Hence, even water is absorbing if we consider a large enough volume. Examples of absorbing spheres in a nonabsorbing host are materials such as paints and plastics, suspensions of metallic particles in a nonabsorbing solvent, colored glass, and the like. It is useful to include the more general case where the refractive index of the host medium is also allowed to be complex. This extension of the Lorenz-Mie theory was seemingly first considered by Mundy et al. [4]. These authors used a complex index of refraction for the host medium $n_{med} = n'_{med} + in''_{med}$. However, they retained the assumption of a homogeneous wave and therefore ended up with angular dependency of the scattering equivalent to that of Lorenz and Mie.

After the paper by Mundy et al. [4] there has been much discussion on scattering by particles in an absorbing host [5–15]. The discussion is concerned with the problem that we cannot really consider the scattering cross section to be an independent property of the material because there is an exponential attenuation term in the direction towards the observer. It means
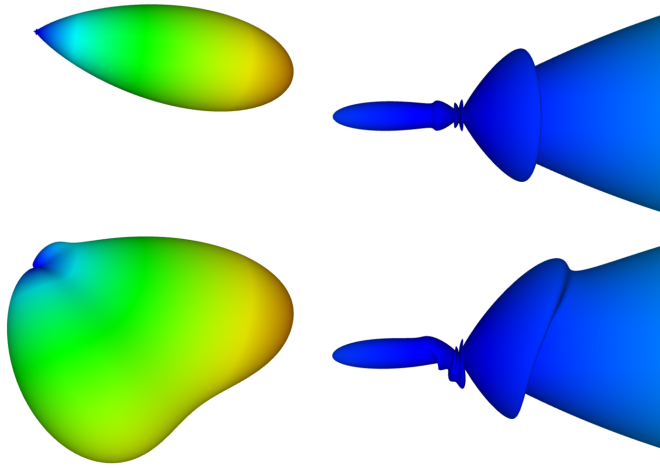
Fig. 1. The top row visualizes the Lorenz-Mie phase function for a homogeneous incident wave. The bottom row visualizes the same phase function, but taking significant inhomogeneity of the incident wave into account. Each column uses a different wavelength, type of particle, and host medium. To the left, we have the first case of Sec. 6, but with an absorbing host and a more inhomogeneous wave. To the right, we have one of the polarization components from the second case of Sec. 6.

that we cannot really tell how much light was scattered by the particle if we look at the light that reaches a far away observer. This problem is still unsolved, and I leave it as is. I work with a different problem, namely that the authors of this previous work all assume the incident wave to be a homogeneous plane wave. Some of them [8] explicitly mention that the light should be incident on the host medium along the normal direction to ensure that this assumption holds true.

The Lorenz-Mie formulae were derived for homogeneous waves of light, but waves are only very rarely homogeneous when propagating in an absorbing medium [16]. Light refracting into an absorbing material is only homogeneous at normal incidence [17]. Belokopytov and Vasil'ev [18] have found a solution for the scattering of plane inhomogeneous waves by an absorbing particle in a nonabsorbing host. This is the special case of an evanescent wave produced by total internal reflection in a nonabsorbing medium [17–19]. As opposed to this, the goal is here to find a solution for the scattering of an absorbing sphere in an absorbing host. To do it properly, we need to take inhomogeneous waves in an absorbing medium into account.

Numerical techniques, such as the finite element method [20], are commonly used for computing the scattering of an inhomogeneous plane wave by a spherical particle in an absorbing medium. Recently, Frezza and Mangini [21] presented a solution based on vector spherical harmonics expansion of the fully general expressions for inhomogeneous plane waves. Every term of such an expansion, however, requires evaluation of the complex hypergeometrical function $_2F_1$ [22], which involves an infinite sum of ratios of gamma functions. The function $_2F_1$ is considered notoriously difficult to evaluate [23, 24]. Nevertheless, this previous work enables us to evaluate the scattered electromagnetic field at different positions in an absorbing medium. For applications like radiative transfer, however, it is preferable to have a computationally simpler solution and an explicit expression for the scattering phase function.

No previous work that I am aware of provides explicit for-

mulae for the far-field scattering components when a spherical particle scatters an inhomogeneous wave of light. I present such formulae and provide practical recurrence relations for their evaluation. The terms of the expansion in my solution involve only sums with few terms. This makes the solution sufficiently simple to enable interactive inspection of the shape of the phase function as input parameters vary. Examples are provided in Fig. 1 and the reader is encourages to interactively explore phase function shapes.[1] My solution seems a useful addition to the theory for reflection and refraction of inhomogeneous plane waves by an interface between two absorbing media [25–27]. Perhaps it can also be combined with generalized Lorenz-Mie theory [28], which is concerned with solutions for arbitrary positions of the scattering sphere in an incident beam of arbitrary shape.

The inhomogeneity of light refracting into a thin film was originally investigated to measure the optical properties of highly absorbing materials such as metals [16, 17, 25, 26]. Scattering of an inhomogeneous wave by a particle in a nonabsorbing medium [18] is useful in total internal reflection microscopy [19], as this is based on the scattering by particles of an evanescent wave. Scattering by particles in absorbing media has especially received attention in random lasing [29–32] and solar cell absorption enhancement [33–37]. In the former area, the laser gain medium is often a dye solution with embedded particles, and researchers investigate the influence of scattering on the output character of the laser. In the latter area, particles can be embedded in the active layer of a solar cell to improve the light-harvesting efficiency. The intention of this paper is to take a step toward a better understanding of the scattering by particles in absorbing media such as dyes and solar cells.

2. PLANE WAVES OF LIGHT

An electromagnetic plane wave is described by the real parts of the two plane wave expressions:

$$\mathbf{E}_c(\mathbf{x}, t) = \mathbf{E}_0 e^{-i(\omega t - \mathbf{k} \cdot \mathbf{x})} \tag{1}$$

$$\mathbf{H}_c(\mathbf{x}, t) = \mathbf{H}_0 e^{-i(\omega t - \mathbf{k} \cdot \mathbf{x})}, \tag{2}$$

where $\text{Re}(\mathbf{E}_c)$ is the electric field vector and $\text{Re}(\mathbf{H}_c)$ is the magnetic field vector. These standard equations describe a harmonic plane wave of angular frequency ω with wave vector $\mathbf{k}$. The wave varies in time t and with location in space $\mathbf{x}$. Furthermore, $\mathbf{E}_0$ and $\mathbf{H}_0$ are complex vector amplitudes and $i = \sqrt{-1}$.

Assuming isotropic materials and no charges moving freely through empty space, Maxwell's equations [38] simplify to the time-free form for plane waves [39]:

$$\mathbf{k} \times \mathbf{H}_0 = -\omega(\varepsilon + i\sigma/\omega)\mathbf{E}_0 \tag{3}$$

$$\mathbf{k} \times \mathbf{E}_0 = \omega\mu\mathbf{H}_0 \tag{4}$$

$$\mathbf{k} \cdot \mathbf{E}_0 = 0 \tag{5}$$

$$\mathbf{k} \cdot \mathbf{H}_0 = 0, \tag{6}$$

where ε is the permittivity, μ is the permeability, and σ is the conductivity. Collectively, these three parameters are called the isotropic material properties. The time-free Maxwell equations reveal that the plane waves are required to satisfy the following conditions:

$$\mathbf{k} \cdot \mathbf{E}_0 = \mathbf{k} \cdot \mathbf{H}_0 = \mathbf{E}_0 \cdot \mathbf{H}_0 = 0 \tag{7}$$

$$\mathbf{k} \cdot \mathbf{k} = \omega^2 \mu(\varepsilon + i\sigma/\omega), \tag{8}$$

[1]http://people.compute.dtu.dk/jerf/code/phase/ [This demonstrator requires a computer with a decent graphics processing unit (GPU).]

where all the vectors may be complex and $\varepsilon_c = \varepsilon + i\sigma/\omega$ is sometimes called the complex permittivity (or the complex dielectric constant). The latter equation is particularly interesting as it describes the relation between material and wave propagation.

Let us take a look at the real and imaginary parts of the wave vector **k**. We write

$$\mathbf{k} = \mathbf{k}' + i\mathbf{k}'' = k'\vec{s}' + ik''\vec{s}'',$$

where $k' = |\mathbf{k}'|$ and $k'' = |\mathbf{k}''|$ such that $\vec{s}'$ and $\vec{s}''$ are unit vectors in the directions of the real and imaginary parts of the wave vector, respectively. If the real part of the wave vector $\mathbf{k}'$ is parallel to the imaginary part $\mathbf{k}''$, the wave is said to be *homogeneous*. Otherwise, it is *inhomogeneous*. Of course, $\mathbf{k}'' = \mathbf{0}$ is parallel to any vector, why a wave is homogeneous if $\mathbf{k}$ is real-valued. If $\mathbf{k}$ is complex, the exponential term of the plane wave expressions (1–2) is as follows

$$e^{i\mathbf{k}\cdot\mathbf{x}} = e^{i\mathbf{k}'\cdot\mathbf{x}}e^{-\mathbf{k}''\cdot\mathbf{x}}.$$

Here, we may observe that $\mathbf{k}'$ is the vector normal to the surface of constant phase and $\mathbf{k}''$ is normal to the surface of constant amplitude. The phase velocity is then $v = \omega/k'$ and the amplitude is damped (or decays) in the direction $\vec{s}''$ at the rate k''. Thus, we have an inhomogeneous wave whenever the damping of the wave is not aligned with its direction of propagation.

The rule for propagation of plane waves in an isotropic medium (8) indicates the convenience of a phenomenological quantity defined by

$$n = n' + in'' = c\sqrt{\mu(\varepsilon + i\sigma/\omega)}. \qquad \textbf{(9)}$$

This is the (complex) index of refraction, or refractive index. Multiplication by c, the speed of light *in vacuo*, ensures that n is a unitless quantity. The index of refraction nicely collects the material properties and is a quantity measured as one of the key optical properties of materials.

The energy of the field is given by the magnitude of the Poynting vector **S** [40]. For isotropic materials, we have

$$|\mathbf{S}| = \varepsilon_0 c^2 \mu |\text{Re}(\mathbf{E}_c) \times \text{Re}(\mathbf{H}_c)|,$$

where ε_0 is the electric constant (also called the vacuum permittivity). The factor $\varepsilon_0 c^2 \mu$ is the relative permeability, which is one if the material is not magnetic. Inserting the plane wave expressions (1–2), we get

$$\begin{aligned}|\mathbf{S}| &= \varepsilon_0 c^2 \mu \, e^{-2k''\vec{s}''\cdot\mathbf{x}} \\ &\times \left|\text{Re}(\mathbf{E}_0 e^{-i(\omega t - \mathbf{k}'\cdot\mathbf{x})}) \times \text{Re}(\mathbf{H}_0 e^{-i(\omega t - \mathbf{k}'\cdot\mathbf{x})})\right|.\end{aligned}$$

Since the exponential terms which involve ωt and $\mathbf{k}' \cdot \mathbf{x}$ are only oscillations, it follows that $2k''$ is the exponential attenuation of the energy flux as the wave propagates through the material. This attenuation is called the *absorption coefficient* and it is often denoted by the symbol μ_a or σ_a (these should not be confused with the permeability μ or the conductivity σ).

The direction of the energy flow in the electromagnetic field is generally considered to be the time average of the Poynting vector over a period of oscillation $T = 2\pi/\omega$. This is [39]

$$\mathbf{S}_{\text{avg}} = \frac{\varepsilon_0 c^2 \mu}{2}\text{Re}(\mathbf{E}_c \times \mathbf{H}_c^*), \qquad \textbf{(10)}$$

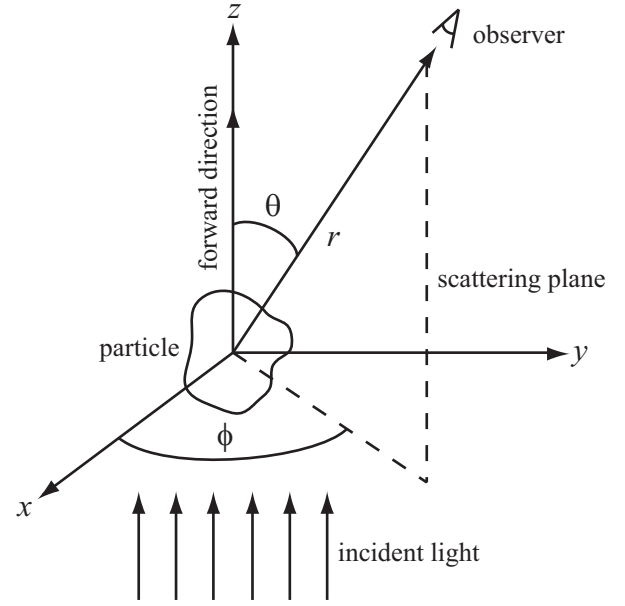where the asterisk * denotes the complex conjugate.

Fig. 2. Scattering by a particle.

3. SCATTERING BY A PARTICLE

Consider a particle in an electromagnetic field. To describe the effect, we split the field in two contributions: an incident field ($\mathbf{E}_i, \mathbf{H}_i$) and a scattered field ($\mathbf{E}_s, \mathbf{H}_s$). The sum of the two contributions makes up the total electromagnetic field:

$$\mathbf{E}_c = \mathbf{E}_i + \mathbf{E}_s, \quad \mathbf{H}_c = \mathbf{H}_i + \mathbf{H}_s.$$

Inserting in Eq. (10) to find the total time-averaged Poynting vector, we get [39]

$$\mathbf{S}_{\text{avg}} = \mathbf{S}_i + \mathbf{S}_s + \mathbf{S}_{\text{ext}},$$

where $\mathbf{S}_i$ and $\mathbf{S}_s$ describe the incident and scattered fields, each with an equation as Eq. (10), but with i or s as subscript instead of c. The term $\mathbf{S}_{\text{ext}}$ describes the interaction of the two fields.

To get an idea about the directions of the different vectors, we introduce a coordinate system; see Fig. 2. The direction of the incident light (that is, the direction of $\mathbf{S}_i$) defines the axis of incidence. Let us orient our coordinate system so that it has origin inside the particle and z-axis along the axis of incidence. The z-axis is then the *forward direction*. Together the forward direction and the direction in which we consider the scattered light define the *scattering plane*. If the two directions are parallel, we may choose any plane containing the axis of incidence. In this setting, a spherical coordinate system (r, θ, ϕ) is oriented such that θ is the angle between the forward direction and the scattered direction, while ϕ is the angle between the x-axis and the scattering plane. This description of the setup follows that of Bohren and Huffman [39] (Sec. 3.2).

At a large distance from the particle (in the far field) the scattered field may be expressed as a spherical wave [39, 41]:

$$\mathbf{E}_s(\mathbf{x}) = i\frac{e^{ikr}}{kr}\mathbf{Z}(\vec{s}), \quad \mathbf{H}_s(\mathbf{x}) = \frac{k}{\omega\mu}i\frac{e^{ikr}}{kr}\vec{s} \times \mathbf{Z}(\vec{s}), \qquad \textbf{(11)}$$

where $\mathbf{x} = r\vec{s}$ such that r is the distance to the observer and $\vec{s}$ is the direction toward the observer, while ω is the angular frequency, k is the (complex) wave number, and μ is the permeability of the medium surrounding the particle. As a consequence

of Eqs. (8–9) and complex number algebra, we have

$$k = k_0 n_{\text{med}} = \sqrt{\mathbf{k} \cdot \mathbf{k}} \neq k' + ik'',$$

where $k_0 = \omega/c$ is the wave number *in vacuo*. With λ as the wavelength *in vacuo*, we also have $k_0 = 2\pi/\lambda$. The vector $\mathbf{Z}$ denotes the strength of the light scattered in the direction $\vec{s}$. There must be a relation between the incident electric field $\mathbf{E}_i$ and the scattered field hidden in the directionally dependent *scattering vector function* $\mathbf{Z}(\vec{s})$. Let us formulate this relationship in the following paragraphs.

We consider two polarizations: one with the electric vector perpendicular to the scattering plane ($\perp$, TE) and one with the electric vector parallel to the scattering plane ($\parallel$, TM). Suppose we (in the case of the electric vector) denote the magnitudes of these two components by $E_{\perp i}$ and $E_{\parallel i}$ for the incident field and by $E_{\perp s}$ and $E_{\parallel s}$ for the scattered field. Then we introduce a *scattering matrix* $\mathbf{S}(\theta, \phi)$, which in general depends on both the angle with the forward direction θ and the location of the scattering plane ϕ. Putting all this notation together, we get a formula describing the relationship between the magnitudes of the scattered components and those of the incident components [39, 42]:

$$\begin{pmatrix} E_{\parallel s} \\ E_{\perp s} \end{pmatrix} = i \frac{e^{ikr}}{kr} \begin{pmatrix} S_2 & S_3 \\ S_4 & S_1 \end{pmatrix} \begin{pmatrix} E_{\parallel i} \\ E_{\perp i} \end{pmatrix} e^{-i\mathbf{k} \cdot \mathbf{x}}, \quad \textbf{(12)}$$

where the four-component matrix is the scattering matrix $\mathbf{S}(\theta, \phi)$ and $\mathbf{k}$ is the wave vector of the incident plane wave.

The scattering matrix gives an expression for the scattering vector function $\mathbf{Z}$:

$$\mathbf{Z} = \begin{pmatrix} \vec{e}_{\parallel} & \vec{e}_{\perp} \end{pmatrix} \begin{pmatrix} S_2 & S_3 \\ S_4 & S_1 \end{pmatrix} \begin{pmatrix} E_{\parallel i} \\ E_{\perp i} \end{pmatrix} e^{-i\mathbf{k} \cdot \mathbf{x}}, \quad \textbf{(13)}$$

where $\vec{e}_{\parallel}$ and $\vec{e}_{\perp}$ are unit vectors parallel and perpendicular to the scattering plane, respectively. They are chosen such that $\vec{e}_{\parallel} \times \vec{e}_{\perp} = \vec{s}$. Now that we have an expression connecting $\mathbf{Z}$ to the incident field, let us see how $\mathbf{Z}$ relates to the scattered energy. By insertion of the spherical wave equations (11) in the expression for the scattering Poynting vector (Eq. 10 with subscript s instead of c), we get

$$\mathbf{S}_s = \frac{\varepsilon_0 c \, n'_{\text{med}}}{2} \frac{e^{-2k_0 n''_{\text{med}} r}}{|k|^2 r^2} \text{Re}(\mathbf{Z}(\vec{s}) \times (\vec{s} \times \mathbf{Z}(\vec{s}))^*). \quad \textbf{(14)}$$

For comparison, the Poynting vector of the incident light field is:

$$\mathbf{S}_i = \frac{\varepsilon_0 c^2}{2\omega} e^{-2\mathbf{k}'' \cdot \mathbf{x}} \text{Re}(\mathbf{E}_{0i} \times (\mathbf{k} \times \mathbf{E}_{0i})^*),$$

Because the vectors are perpendicular, we get the following magnitudes of the scattering and incident Poynting vectors:

$$|\mathbf{S}_s| = \frac{\varepsilon_0 c \, n'_{\text{med}}}{2} \frac{e^{-2k_0 n''_{\text{med}} r}}{|k|^2 r^2} |\mathbf{Z}(\vec{s})|^2$$

$$|\mathbf{S}_i| = \frac{\varepsilon_0 c \, n'_{\text{med}}}{2} e^{-2\mathbf{k}'' \cdot \mathbf{x}} |\mathbf{E}_{0i}|^2.$$

Let us see if we can find out something about the ratio of scattered to incident light. We denote the surface of the particle A. The rate of energy transfer across a closed surface arbitrarily close to A is given by the integral [39]

$$\int \mathbf{S}_{\text{avg}} \cdot \vec{n} \, dA,$$

where $\vec{n}$ denotes the direction of the outward surface normal. The total scattering by a particle is then an integral over the surface of the particle. A patch on the surface of the particle can be considered an area ΔA or a solid angle $\Delta \Omega$. We have

$$\Delta \Omega = \frac{\Delta A}{r^2},$$

which means that the energy scattered by a particle is given by

$$W_s = \int \mathbf{S}_s \cdot \vec{s} \, dA = \int_{4\pi} \frac{\varepsilon_0 c \, n'_{\text{med}}}{2} \frac{e^{-2k_0 n''_{\text{med}} r}}{|k|^2} |\mathbf{Z}(\vec{s})|^2 \, d\Omega.$$

The conventional way of describing the ratio of scattered to incident light is to divide W_s by the incident energy flux $|\mathbf{S}_i|$. To have the ratio on a simple form, we introduce a scattering vector function $\mathbf{X}$, which is like $\mathbf{Z}$, but normalized so that its expression has the weights of the two polarizations ($\parallel$ and $\perp$) rather than $E_{\parallel i}$ and $E_{\perp i}$:

$$\mathbf{X}(\vec{s}) = \frac{\mathbf{Z}(\vec{s})}{|\mathbf{E}_{0i}|}.$$

The ratio is then

$$C_s = \frac{W_s}{|\mathbf{S}_i|} = \frac{e^{-2k_0 n''_{\text{med}} r}}{e^{-2\mathbf{k}'' \cdot \mathbf{x}}} \frac{1}{|k|^2} \int_{4\pi} |\mathbf{X}(\vec{s})|^2 \, d\Omega, \quad \textbf{(15)}$$

where

$$\mathbf{k}'' \cdot \mathbf{x} = (k'' \vec{s}'') \cdot (r\vec{s}) \approx k_0 n''_{\text{med}} r \frac{\vec{s} \cdot \vec{s}''}{\vec{s}' \cdot \vec{s}''} \approx k_0 n''_{\text{med}} r,$$

why we assume that the exponential term in the expression for C_s is 1. A ratio of total scattered energy to energy incident per unit area (15) gives an area as result. This particular area C_s is referred to as the *scattering cross section* of the particle. It is the area that would receive the same amount of energy as the particle scatters if we subtend it normal to the incident light.

If we normalize the directionally dependent quantity under the integral, we have the phase function of the particle:

$$p(\theta, \phi) = \frac{|\mathbf{X}(\theta, \phi)|^2}{|k|^2 C_s}. \quad \textbf{(16)}$$

Let us now turn to the idealized case of a spherical particle.

4. SCATTERING BY A SPHERE

Consider a plane wave scattered by a spherical particle embedded in a host medium. We take it that both the host medium and the particle are isotropic, homogeneous substances. Plane waves would otherwise not be a good approximation. Note that we make no assumptions about the absorption properties of the host medium and the particle, and we do not require the plane wave to be homogeneous or inhomogeneous.

We are aiming at a solution for the scattering phase function. As seen in Eq. (16), this requires the normalization factor $|k|^2 C_s$ and the components of the scattering matrix $\mathbf{S}(\theta, \phi)$.

In Sec. 3 (Fig. 2), we chose our coordinate system so that light is incident along the z-axis. This means that the z-axis points along the direction of $\mathbf{S}_i$. For homogeneous plane waves, the direction of $\mathbf{S}_i$ is parallel to both the real and imaginary parts of the wave vector, but in general this is not necessarily the case. This means that the wave vector $\mathbf{k}$ has two different directions: one for the real part $\mathbf{k}'$ and one for the imaginary part $\mathbf{k}''$.

The scattering plane is spanned by the z-axis (forward direction) and the direction of the scattered light. Since we are
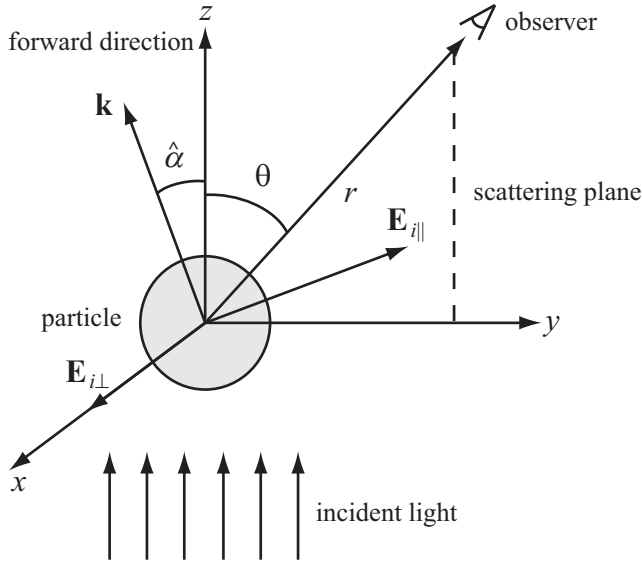
Fig. 3. Scattering by a spherical particle in an absorbing host.

working with a sphere, it is of no consequence with respect to particle geometry which way we choose to orient the x and y axes of the coordinate system. We are therefore always allowed to perform a change of basis and choose new directions for the x and y axes. Suppose we choose for any point of observation to align the yz-plane with the scattering plane through a change of basis. We can then find the coordinates of the vectors in the new basis through projection. In this coordinate system, where the yz-plane is aligned with the scattering plane, it is convenient to specify an arbitrary incident plane wave by

$$\mathbf{E}_i = \mathbf{E}_{0i} e^{-i(\omega t - k_x x)} e^{i(k_y y + k_z z)} . \tag{17}$$

We can think of it as a wave with wave vector $\mathbf{k} = (0, k_y, k_z)$ and amplitude that varies in time and with the location along the x-axis. Note that this wave vector corresponds exactly to the projection of an arbitrary wave vector on the scattering plane spanned by the forward direction and the direction toward the observer. Note also that, in the end, we are only interested in evaluating this wave at the point of observation, where $x = 0$ with our particular choice of basis.

Electromagnetic waves are transverse (due to Maxwell's equations). Any inhomogeneous transverse plane wave can be decomposed into a transverse electric wave (TE) and a transverse magnetic (TM) wave [26]. We now decompose our incident wave in this way. As before, we identify TE components with $\perp$-polarized components and TM components with $\parallel$-polarized components. The $\perp$-polarized component of the electric field vector is by definition perpendicular to the scattering plane. The $\parallel$-polarized component lies in the scattering plane. We have

$$\mathbf{E}_i = E_{\perp i} \vec{e}_x + E_{\parallel i} (a \vec{e}_y + b \vec{e}_z) , \tag{18}$$

where $\vec{e}_x$, $\vec{e}_y$, and $\vec{e}_z$ are unit vectors in the directions of the coordinate axes, $E_{\perp i}$ and $E_{\parallel i}$ are complex magnitudes of the two polarization components of $\mathbf{E}_i$, and a and b are the direction cosines of $\mathbf{E}_{\parallel i}$ in the y and z directions, respectively. See Fig. 3. For a homogeneous wave, the direction of $\mathbf{E}_{\parallel i}$ would also be perpendicular to the z-axis, but this is not true in general.

The direction cosines a and b are given by

$$a = E_{y \parallel i} / E_{\parallel i} \quad \text{and} \quad b = E_{z \parallel i} / E_{\parallel i} .$$

Using that the $\parallel$-polarized component is also transverse magnetic (TM), we have

$$H_{y \parallel i} = H_{z \parallel i} = 0 \quad \text{and} \quad H_{x \parallel i} = H_{\parallel i} = \sqrt{\mathbf{H}_{\parallel i} \cdot \mathbf{H}_{\parallel i}} .$$

Inserting this in Eq. (3) and considering the magnitude of the (complex) wave vector (8), we obtain

$$k_z H_{\parallel i} = -\frac{k^2}{\omega \mu} E_{y \parallel i} \quad \text{and} \quad k_y H_{\parallel i} = \frac{k^2}{\omega \mu} E_{z \parallel i}$$

from which the following expressions appear:

$$a = -\frac{k_z \omega \mu}{k^2} \frac{H_{\parallel i}}{E_{\parallel i}} , \quad b = \frac{k_y \omega \mu}{k^2} \frac{H_{\parallel i}}{E_{\parallel i}} .$$

The ratio of the amplitude magnitudes of the (complex) electric and magnetic vectors is obtained from Eqs. (4–5):

$$\sqrt{(\mathbf{k} \times \mathbf{E}_{\parallel i}) \cdot (\mathbf{k} \times \mathbf{E}_{\parallel i})} = k E_{\parallel i} = \omega \mu H_{\parallel i} .$$

Insertion leads to

$$a = -\frac{k_z}{k_0 n_{\text{med}}} = -\cos \hat{\alpha} , \quad b = \frac{k_y}{k_0 n_{\text{med}}} = \sin \hat{\alpha} , \tag{19}$$

where $\hat{\alpha}$ is a complex angle denoting the direction of the wave vector in the scattering plane (Fig. 3). In the following, I use a Greek letter with a hat for complex angles.

The structure of the electric vector of the incident wave (17) is quite different from the case of a homogeneous wave, where only one spatial coordinate is left in the exponential. For the inhomogeneous wave *the scattering is not axially symmetric around the forward direction*. This is counterintuitive as the geometry of the scatterer is perfectly symmetric. The inhomogeneity of the wave is enough to break the symmetry. As a consequence, we can use only part of the results of Lorenz and Mie (and many others) for inhomogeneous waves. To find a solution, we look to the work of Belokopytov and Vasil'ev [18].

Suppose we introduce the following slightly unusual set of spherical coordinates:

$$\begin{aligned} x &= r \cos \beta \\ y &= r \sin \beta \sin \theta \\ z &= r \sin \beta \cos \theta , \end{aligned}$$

where the x-axis is the polar axis such that the scattering plane is given by $\sin \beta = 1$ and the forward direction when also $\theta = 0$. The angle θ is shown in Fig. 3. With these spherical coordinates, the expression for our incident field is

$$\mathbf{E}_i = \mathbf{E}_{0i} e^{-i(\omega t - k_x x)} e^{ir(k_y \sin \beta \sin \theta + k_z \sin \beta \cos \theta)} .$$

Using the complex angle $\hat{\alpha}$, we can also write it as

$$\begin{aligned} \mathbf{E}_i &= \mathbf{E}_{0i} e^{-i(\omega t - k_x x)} e^{ikr \sin \beta (\sin \hat{\alpha} \sin \theta + \cos \hat{\alpha} \cos \theta)} \\ &= \mathbf{E}_{0i} e^{-i(\omega t - k_x x)} e^{ikr \sin \beta \cos(\theta - \hat{\alpha})} . \end{aligned} \tag{20}$$

This is very close to the form of incident wave for which Belokopytov and Vasil'ev found a solution [18]. The generalization is in the definition of $\hat{\alpha}$ (which is purely imaginary in their treatment). For an inhomogeneous wave, $\mathbf{k}'$ and $\mathbf{k}''$ are not parallel and a nonabsorbing medium has $\mathbf{k} \cdot \mathbf{k} = k_0^2 n_{\text{med}}^2$ to be a real number. This can be fulfilled by having k_z real and k_y purely imaginary, and then the structure of our incident wave becomes equivalent to the one addressed by Belokopytov and Vasil'ev.

Using a formula due to Bauer [43], we now expand the exponential part of the plane inhomogeneous incident wave in spherical harmonics [1, 2, 18, 41]:

$$e^{ikr \sin \beta \cos(\theta - \hat{\alpha})} = \sum_{n=0}^{\infty} i^n (2n+1) \frac{\psi_n(kr)}{kr} P_n(\sin \beta \cos(\theta - \hat{\alpha})).$$

The functions ψ_n are Riccati-Bessel functions, which we will return to later. The wave number $k = k_0 n_{\text{med}}$ is complex for an absorbing host, but that does not invalidate the expansion. In the case of homogeneous waves, the argument for the Legendre polynomials P_n are real-valued. This is not the case for inhomogeneous waves. We would like to avoid evaluating the Legendre polynomials for complex arguments, as this would lead to the computationally difficult solutions employed in previous work [21, 22]. We therefore use the addition theorem for spherical harmoics to obtain [18]:

$$P_n(\sin \beta \cos(\theta - \hat{\alpha})) = P_n(\cos \beta) P_n(0)$$
$$+ 2 \sum_{m=1}^{n} \frac{(n-m)!}{(n+m)!} P_n^m(\cos \beta) P_n^m(0) \cos(m(\theta - \hat{\alpha})),$$

where P_n^m are the associated Legendre polynomials. Based on Abramowitz and Stegun [44], we find

$$P_n^m(0) = (-1)^{(n+m)/2} \frac{2^{-n}(n+m)!}{[(n-m)/2]![(n+m)/2]!} \quad \textbf{(21)}$$

for $n + m$ even and

$$P_n^m(0) = 0 \qquad \text{for } n + m \text{ odd.} \quad \textbf{(22)}$$

To shorten notation it is convenient to introduce

$$R_n^m = (-1)^{(n+m)/2} \frac{2^{1-n-\delta_{m0}}(n-m)!}{[(n-m)/2]![(n+m)/2]!} \quad \textbf{(23)}$$

for $n + m$ even and

$$R_n^m = 0 \qquad \text{for } n + m \text{ odd,} \quad \textbf{(24)}$$

where δ_{m0} is a Kronecker delta which is 1 when $m = 0$ and 0 otherwise. Then [18]

$$P_n(\sin \beta \cos(\theta - \hat{\alpha})) = \sum_{m=0}^{n} R_n^m P_n^m(\cos \beta) \cos(m(\theta - \hat{\alpha})).$$

What we are really interested in is the scattered wave. In particular, we would like to determine the four components of the scattering matrix $\mathbf{S}(\theta, \phi)$. It has been shown many times in the literature that all the components of the scattered field can be found using the Debye potentials [3, 41, 42, 45]. In the expansion of Belokopytov and Vasil'ev [18], the Debye potentials of the scattered wave outside the sphere are:

$$u = \frac{1}{k^2} \sum_{n=1}^{\infty} i^{n-1} \frac{2n+1}{n(n+1)} a_n \zeta_n(kr) U_n(\theta, \beta) \quad \textbf{(25)}$$

$$v = \frac{1}{kk_0\mu} \sum_{n=1}^{\infty} i^{n-1} \frac{2n+1}{n(n+1)} b_n \zeta_n(kr) V_n(\theta, \beta), \quad \textbf{(26)}$$

where

$$U_n(\theta, \beta) = (E_{\|i} W_{1n}(\theta, \beta) + E_{\perp i} W_{2n}(\theta, \beta)) e^{-i\mathbf{k} \cdot \mathbf{x}} \quad \textbf{(27)}$$

$$V_n(\theta, \beta) = (E_{\|i} W_{2n}(\theta, \beta) - E_{\perp i} W_{1n}(\theta, \beta)) e^{-i\mathbf{k} \cdot \mathbf{x}} \quad \textbf{(28)}$$

$$W_{1n}(\theta, \beta) = \sum_{m=0}^{n} m R_n^m P_n^m(\cos \beta) \sin(m(\theta - \alpha)) \quad \textbf{(29)}$$

$$W_{2n}(\theta, \beta) = \sum_{m=0}^{n} (n-m) R_{n-1}^m P_n^m(\cos \beta) \cos(m(\theta - \alpha)). \quad \textbf{(30)}$$

The exponential terms in Eqs. (27–28) appear to cancel the corresponding but reciprocal exponential terms in $E_{\|i}$ and $E_{\perp i}$. The spherical function ζ_n is another Riccati-Bessel function and the coefficients a_n and b_n are the traditional Lorenz-Mie coefficients. We will return to those shortly.

Far field expressions for the magnitudes of the two polarization components of the scattered field ($E_{s\|}$ and $E_{s\perp}$) lead to expressions for the components in the scattering matrix (see Eq. 12). We have [39, 42]

$$E_{\|s} = -E_\theta \qquad \text{and} \qquad E_{\perp s} = E_\beta \quad \textbf{(31)}$$

with flipped signs because of our unusual choice of spherical coordinate system, and using the Debye potentials we also have [3, 41]

$$E_\beta = \frac{1}{r} \frac{\partial^2 u}{\partial r \partial \beta} + ik_0 \mu \frac{1}{r \sin \beta} \frac{\partial v}{\partial \theta} \quad \textbf{(32)}$$

$$E_\theta = \frac{1}{r \sin \beta} \frac{\partial^2 u}{\partial r \partial \theta} - ik_0 \mu \frac{1}{r} \frac{\partial v}{\partial \beta}. \quad \textbf{(33)}$$

Considering the relations (31) between these components and the expressions for $E_{s\|}$ and $E_{s\perp}$ from Eq. (12), we obtain equations which involve the components of the scattering matrix. In the far field, we have [3]

$$\zeta_n(kr) \approx i^{-n-1} e^{ikr} \qquad \text{and} \qquad \frac{\partial \zeta_n(kr)}{\partial r} \approx i^{-n} k e^{ikr}. \quad \textbf{(34)}$$

Inserting this far field approximation (34) in the Debye potentials (25–26) and inserting the potentials in the expressions for E_β and E_θ (32–33), the equations with the components of the scattering matrix simplify to

$$(S_4 E_{\|i} + S_1 E_{\perp i}) e^{-i\mathbf{k} \cdot \mathbf{x}}$$
$$= \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left(-a_n \frac{\partial U_n}{\partial \beta} - b_n \frac{1}{\sin \beta} \frac{\partial V_n}{\partial \theta} \right)$$
$$(S_2 E_{\|i} + S_3 E_{\perp i}) e^{-i\mathbf{k} \cdot \mathbf{x}}$$
$$= \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left(a_n \frac{1}{\sin \beta} \frac{\partial U_n}{\partial \theta} - b_n \frac{\partial V_n}{\partial \beta} \right).$$

Using the expressions (27–30) for the U_n and V_n functions, we arrive at the following scattering matrix components:

$$S_1 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left(-a_n \frac{\partial W_{2n}}{\partial \beta} + b_n \frac{1}{\sin \beta} \frac{\partial W_{1n}}{\partial \theta} \right) \quad \textbf{(35)}$$

$$S_2 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left(a_n \frac{1}{\sin \beta} \frac{\partial W_{1n}}{\partial \theta} - b_n \frac{\partial W_{2n}}{\partial \beta} \right) \quad \textbf{(36)}$$

$$S_3 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left(a_n \frac{1}{\sin \beta} \frac{\partial W_{2n}}{\partial \theta} + b_n \frac{\partial W_{1n}}{\partial \beta} \right) \quad \textbf{(37)}$$

$$S_4 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left(-a_n \frac{\partial W_{1n}}{\partial \beta} - b_n \frac{1}{\sin \beta} \frac{\partial W_{2n}}{\partial \theta} \right). \quad \textbf{(38)}$$

Let us interpret the angles appearing in this generalized formulation of the Lorenz-Mie theory. Using a change of basis depending on the point of observation, we ensured that the x-axis is along the direction of the $\perp$-polarized component of the electric vector. The angle β is the polar angle between this component and the scattering plane. Since we are only evaluating the wave at the point of observation (where $x = 0$), this is a right angle, and thus $\beta = \pi/2$. The azimuth angle θ is the angle in the scattering plane between the forward direction and the

direction of the scattered light. This means that θ is the standard scattering angle. Another angle in the formulae is the complex angle $\hat{\alpha}$. It is a measure of the inhomogeneity of the incident wave, and it is given by the direction cosines in the scattering plane of the wave vector $\mathbf{k}$ of the incident light (Eq. 19).

It should be mentioned that for a homogeneous wave, we get $k_z = k$ and $k_y = 0$, and then $\hat{\alpha} = 0$. This simplifies the structure of the exponential term in the expression for the incident wave, and we get the components of the scattering matrix in the commonly available form [1–3, 42]:

$$S_1(\theta) \quad = \quad \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left(a_n \pi_n(\cos\theta) + b_n \tau_n(\cos\theta) \right) \quad \textbf{(39)}$$

$$S_2(\theta) \quad = \quad \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left(a_n \tau_n(\cos\theta) + b_n \pi_n(\cos\theta) \right), \quad \textbf{(40)}$$

where the functions π_n and τ_n are related to the Legendre polynomials P_n as follows:

$$\pi_n(\cos\theta) \quad = \quad \frac{P_n^1(\cos\theta)}{\sin\theta} = \frac{\mathrm{d}P_n(\cos\theta)}{\mathrm{d}(\cos\theta)} \quad \textbf{(41)}$$

$$\tau_n(\cos\theta) \quad = \quad \frac{\mathrm{d}P_n^1(\cos\theta)}{\mathrm{d}\theta}$$

$$= \quad \cos\theta\,\pi_n(\cos\theta) - \sin^2\theta\,\frac{\mathrm{d}\pi_n(\cos\theta)}{\mathrm{d}(\cos\theta)}. \quad \textbf{(42)}$$

Their numeric evaluation can be found in standard references on Lorenz-Mie theory [39, 46].

It remains in this discussion of scattering by a spherical particle to give expressions for the Lorenz-Mie coefficients a_n and b_n. In the far field, they are given by the same formulae that Lorenz [1] derived more than a century ago, only now the refractive indices are complex:

$$a_n \quad = \quad \frac{n_{\mathrm{med}}\psi_n'(y)\psi_n(x) - n_p\psi_n(y)\psi_n'(x)}{n_{\mathrm{med}}\psi_n'(y)\zeta_n(x) - n_p\psi_n(y)\zeta_n'(x)} \quad \textbf{(43)}$$

$$b_n \quad = \quad \frac{n_p\psi_n'(y)\psi_n(x) - n_{\mathrm{med}}\psi_n(y)\psi_n'(x)}{n_p\psi_n'(y)\zeta_n(x) - n_{\mathrm{med}}\psi_n(y)\zeta_n'(x)}, \quad \textbf{(44)}$$

where the primes $'$ denote derivative. The Riccati-Bessel functions $\psi_n(z)$ and $\zeta_n(z)$ are related to the spherical Bessel functions $j_n(z)$ and $y_n(z)$ as follows:

$$\psi_n(z) \quad = \quad z j_n(z)$$
$$\zeta_n(z) \quad = \quad z(j_n(z) + i y_n(z)).$$

One should use the complex conjugate of ζ_n if the time dependence for the wave had been taken to be $e^{i\omega t}$ instead of $e^{-i\omega t}$. The argument z is an arbitrary complex number, the arguments x and y used for the Lorenz-Mie coefficients are related to particle and host media as follows:

$$x = \frac{2\pi r n_{\mathrm{med}}}{\lambda} \quad \text{and} \quad y = \frac{2\pi r n_p}{\lambda},$$

where λ is the wavelength *in vacuo* and r is the radius of the spherical particle.

When computers came around, it turned out to be quite difficult to find a numerically stable way of evaluating the spherical functions ψ_n and ζ_n for complex arguments. Eventually, the numerical difficulties were solved for complex y [46–48]. This is sufficient for the traditional Lorenz-Mie theory with a nonabsorbing host medium. When people started considering spheres in an absorbing host, starting with Mundy et al. [4], it became

necessary to find a robust way of evaluating the Lorenz-Mie coefficients for complex x as well. A robust evaluation scheme has been proposed by Frisvad et al. [49].

To complete the phase function solution, we still miss the normalization factor $|k|^2 C_s$ and the calculation of the complex angle $\hat{\alpha}$ by means of projection. With the Lorenz-Mie coefficients, we can find the normalization factor for the phase function using [39]

$$|k|^2 C_s = 2\pi \sum_{n=1}^{\infty} (2n+1)(|a_n|^2 + |b_n|^2).$$

Suppose we let $\mathbf{k}_i$ denote the arbitrary wave vector of the incident field before projection to the scattering plane. The projected wave vector is then given by

$$\mathbf{k} = \mathbf{k}_i - \frac{\mathbf{k}_i \cdot (\mathbf{S}_i \times \vec{s})}{|\mathbf{S}_i \times \vec{s}|^2}\mathbf{S}_i \times \vec{s} \quad \textbf{(45)}$$

with $\mathbf{k} = \mathbf{k}_i$ if $|\mathbf{S}_i \times \vec{s}| = 0$. After this projection, we can find the complex angle $\hat{\alpha}$ using dot products and arcus cosine for complex numbers

$$\hat{\alpha} = \cos^{-1}\frac{\mathbf{S}_i \cdot \mathbf{k}}{|\mathbf{S}_i|\sqrt{\mathbf{k} \cdot \mathbf{k}}}.$$

In this setting, we can calculate $\vec{s}$ from the spherical coordinates θ and ϕ if we want angles as arguments for the phase function.

5. SIMPLIFICATION

Given that we always have $\beta = \pi/2$, we should try to simplify the solution presented in Equations 35–38. What we are interested in is the partial derivatives of the functions W_{1n} and W_{2n} (Equations 29–30). This means that we need the derivatives

$$\frac{\mathrm{d}}{\mathrm{d}\theta}(\sin(m(\theta - \hat{\alpha}))) \quad = \quad m\cos(m(\theta - \hat{\alpha}))$$

$$\frac{\mathrm{d}}{\mathrm{d}\theta}(\cos(m(\theta - \hat{\alpha}))) \quad = \quad -m\sin(m(\theta - \hat{\alpha}))$$

and

$$\frac{\mathrm{d}}{\mathrm{d}\beta}P_n^m(\cos\beta) = -\sin\beta\frac{\mathrm{d}P_n^m}{\mathrm{d}x}(x) = -\frac{\mathrm{d}P_n^m}{\mathrm{d}x}(0),$$

where $x = \cos\beta$. The recurrence relations for the associated Legendre polynomials provide excellent tools for evaluating the special case of zero as argument for the associated Legendre polynomials and their derivatives. Using the relations

$$(x^2 - 1)\frac{\mathrm{d}P_n^m}{\mathrm{d}x}(x) \quad = \quad \sqrt{1 - x^2}P_n^{m+1}(x) + mxP_n^m(x)$$

$$(x^2 - 1)\frac{\mathrm{d}P_n^m}{\mathrm{d}x}(x) \quad = \quad -(n+m)(n-m+1)$$
$$\times \sqrt{1 - x^2}P_n^{m-1}(x) - mxP_n^m(x),$$

we get the following simple relations for the special case of $\beta = \pi/2$:

$$\frac{\mathrm{d}}{\mathrm{d}\beta}P_n^m\left(\cos\frac{\pi}{2}\right) = -\frac{\mathrm{d}P_n^m}{\mathrm{d}x}(0) \quad = \quad P_n^{m+1}(0) \quad \textbf{(46)}$$

$$\frac{\mathrm{d}}{\mathrm{d}\beta}P_n^n\left(\cos\frac{\pi}{2}\right) = -\frac{\mathrm{d}P_n^n}{\mathrm{d}x}(0) \quad = \quad 0.$$

This means that, if we have the zero argument values of the associated Legendre polynomials (Equations 21–22, Fig. 4), the zero argument derivatives with respect to the angle are given

$n\backslash m$	-4	-3	-2	-1	0	1	2	3	4
0					1				
1				1/2	0	-1			
2			1/8	0	-1/2	0	3		
3		1/48	0	-1/8	0	3/2	0	-15	
4	1/384	0	-1/48	0	3/8	0	-15/2	0	105

Fig. 4. The zero argument values of the associated Legendre polynomials $P_n^m(0)$ for $n = 0, 1, 2, 3, 4$ and $m = -4, -3, \ldots, 3, 4$.

by a right hand shift (or inserting zero if there is no right hand neighbour).

The recurrence relations also provide simple formulae for finding the values of the associated Legendre polynomials themselves. Using the relations

$$
\begin{aligned}
P_0^0(x) &= 1 \\
P_{n+1}^n(x) &= x(2n+1)P_n^n(x) \\
\sqrt{1-x^2}\,P_n^{m+1}(x) &= (n-m)xP_n^m(x) - (n+m)P_{n-1}^m(x) \\
2mxP_n^m(x) &= \sqrt{1-x^2} \\
&\times \left[(n+m)(m-n-1)P_n^{m-1}(x) - P_n^{m+1}(x) \right],
\end{aligned}
$$

we get the following relations for the special case of zero argument:

$$
\begin{aligned}
P_0^0(0) &= 1 & \textbf{(47)} \\
P_{n+1}^n(0) &= 0 & \textbf{(48)} \\
P_n^{m+1}(0) &= -(n+m)P_{n-1}^m(0) & \textbf{(49)} \\
P_n^{m-1}(0) &= (n+m)^{-1}(m-n-1)^{-1}P_n^{m+1}(0). & \textbf{(50)}
\end{aligned}
$$

These four relations provide a simple forward recurrence for computing the zero argument values of all the associated Legendre polynomials. Placing the values in a triangle as illustrated in Fig. 4, the algorithm starts with the initial value (47), takes a diagonal step down and to the right to find the last element of the next row (49), inserts a zero before the last element of the row (48), fills out the remaining elements of the row (50), and goes back to the last element from where it takes a new diagonal step (49). This continues until all desired rows have been filled.

The values in Fig. 4 can also be calculated using Equations 21–22. From a numerical standpoint, the recurrence algorithm (47–50) is significantly more attractive as it avoids the factorial function. To make the evaluation of R_n^m equally attractive, we find from Equations 21–24 that

$$
R_n^m = 2^{1-\delta_{m0}}(-1)^m P_n^{-m}(0). \tag{51}
$$

These equations also inform us that when $n+m$ is an odd number, the values of $P_n^m(0)$ and R_n^m are zero. Thus $R_{n-1}^m P_n^m(0)$ are zero, and $R_n^m \frac{dP_n^m(0)}{d\beta}$ are also zero due to Equation 46. This means that the functions $W_{2n}(\theta, \pi/2)$ vanish unless we take the partial derivative with respect to β, whence $W_{1n}(\theta, \pi/2)$ would vanish instead. From this, we conclude that

$$
S_3 = S_4 = 0.
$$

Equation 21 also enables us to simplify the expressions for the scattering components S_1 and S_2 (35–36). In fact, we can
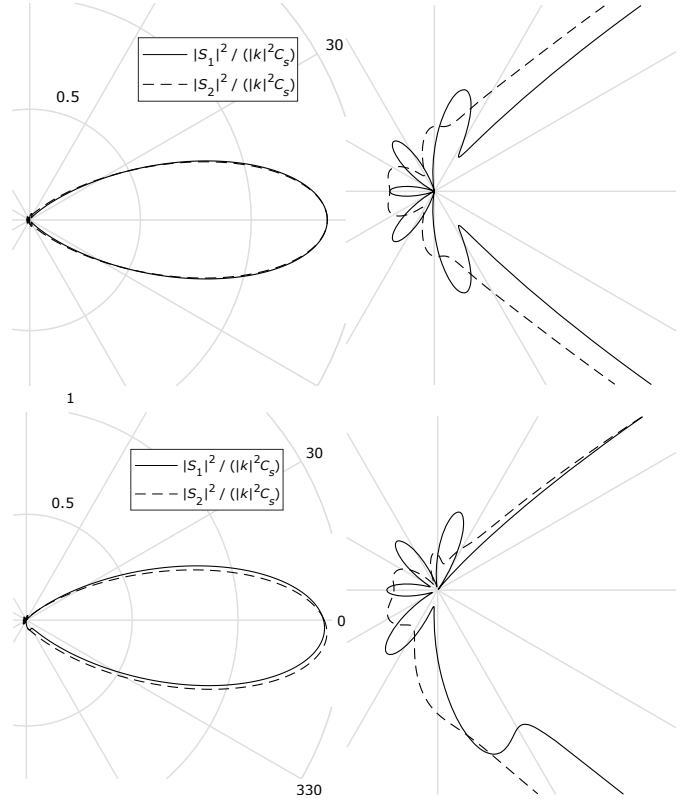

Fig. 5. The two polarization components of the phase function for a spherical particle with $x \approx 4$. The first row is the result for a homogeneous incident wave (Mie scattering). The second row is the result for an inhomogeneous wave with $\alpha \approx i\,0.1$. The plots in the right column are closeups of the region around the origin of the plots in the left column.

write down new expressions for the functions π_n and τ_n that are used in the standard expressions for the scattering components (39–40). For homogeneous and inhomogeneous plane waves, we have

$$
\begin{aligned}
\pi_n(\hat{\theta}) &= -\frac{\partial W_{2n}(\theta, \frac{\pi}{2})}{\partial \beta} \\
&= \sum_{m=0}^{n-1} (m-n)R_{n-1}^m P_n^{m+1}(0)\cos(m\hat{\theta}) & \textbf{(52)} \\
\tau_n(\hat{\theta}) &= \frac{\partial W_{1n}(\theta, \frac{\pi}{2})}{\partial \theta} \\
&= \sum_{m=1}^{n} m^2 R_n^m P_n^m(0)\cos(m\hat{\theta}), & \textbf{(53)}
\end{aligned}
$$

where $\hat{\theta} = \theta - \hat{\alpha}$ is allowed to be a complex number and the scattering components are given by the usual equations (39–40).

6. EVALUATION

To provide some verification of the new, generalized equations for the angular dependency of the scattering components (52–53), we first see if they reproduce results for homogeneous waves calculated using the standard equations (41–42). I do this using the same configuration as Belokopytov and Vasil'ev [18], namely $r = 0.4\,\mu m$, $n_{\text{med}} = 1$, $n_p = 1.47 + i\,10^{-6}$, and $\lambda = 632.8$ nm. Using 1000 equally distributed angular samples (and $\hat{\alpha} = 0$),
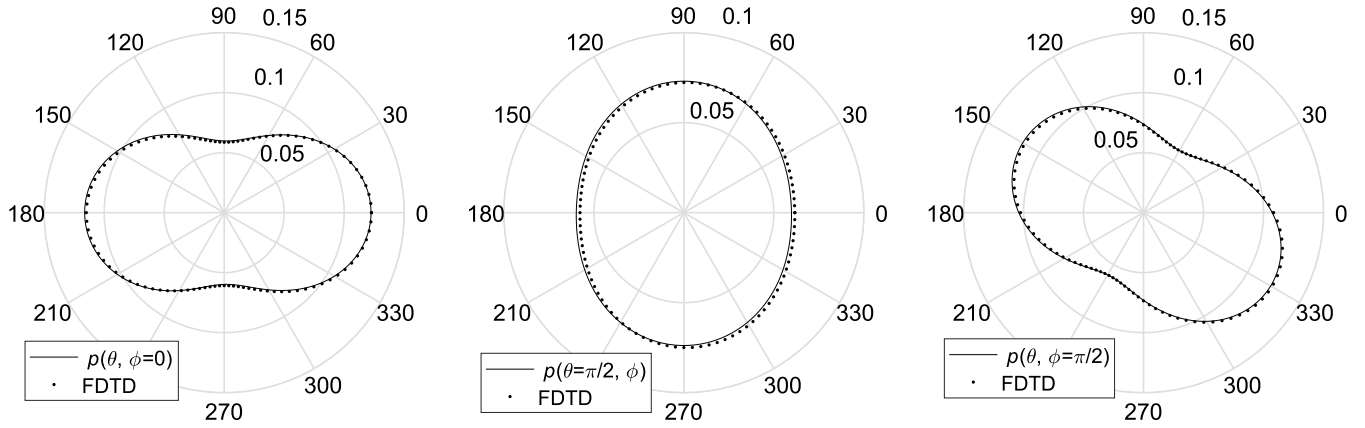
Fig. 6. Finite-difference time-domain (FDTD) simulation of the scattering by a gold particle in air of a plane wave of unpolarized light. Hypothetical wave inhomogeneity is $\hat{\alpha} = 0.5$. Three planar slices of the simulated result are compared with the phase function $p(\theta, \phi)$: (left) $\phi = 0$ and varying θ, (middle) $\theta = \pi/2$ and varying ϕ, (right) $\phi = \pi/2$ and varying θ.

the root-mean-squared difference between (S_1, S_2) calculated using the standard equations (41–42) and the new equations (52–53) was $2 \cdot 10^{-15}$, which is only one order of magnitude larger than the machine epsilon for double precision numbers. This is quite convincing given that we are working with expansions that require quite a few operations per function evaluation. The resulting scattering components are in the first row of Fig. 5.

The scattering of a homogeneous wave by a spherical particle is invariant to rotation around the forward direction. In the case of an inhomogeneous wave with wave vector $\mathbf{k}_i = \mathbf{k}_i' + i\mathbf{k}_i''$, the incident wave is determined by projecting $\mathbf{k}_i'$ and $\mathbf{k}_i''$ to the scattering plane (Eq. 45). If we produce an inhomogeneous wave by means of total internal reflection (an evanescent wave), it will exist along the surface with $\mathbf{k}_i'$ tangent to the surface and $\mathbf{k}_i''$ normal to the surface. In the tangent plane, the projected $\mathbf{k}''$ is the zero vector, and the phase function will be the same as for the homogeneous wave (first row of Fig. 5). In the plane perpendicular to the surface, we would have k_z real and k_y imaginary. The second row of Fig. 5 shows the result for $k_y = i1\ \mu m^{-1}$, which by insertion in Eq. (19) gives $\hat{\alpha} \approx i0.1$. The results in Fig. 5 (right column) are the same as in Fig. 2a of Belokopytov and Vasil'ev [18]. In the tangent plane, these authors find that the result for the inhomogeneous wave differs slightly from the result for the homogeneous wave. This deviation could be due to the fact that they are plotting wave intensities rather than the polarization components of the phase function.

To consolidate the idea of projection to the scattering plane, I compare with a numerical finite-difference time-domain (FDTD) simulation of a plane wave being scattered by a gold particle in air ($n_{med} = 1$). The simulation tool is FDTD Solutions by Lumerical Inc. I use two sources to model unpolarized incident light. The wave is configured to have $\lambda = 525$ nm and $\hat{\alpha} = 0.5$, while the gold particle has radius $r = 0.05\ \mu m$ and refractive index $n_p = 0.6181 + i\,2.144$ [50]. The far-field scattered electric vector output from the numerical simulation was normalized so that it corresponded to a phase function. Results are in Fig. 6. The curves match each other well despite a very slight unexpected shift in the simulation (particularly noticeable in the slice that varies the ϕ angle). This comparison specifically tests the insight that we can use projection instead of varying the β angle in the solution. In this hypothetical case where $\hat{\alpha}$ is real-valued, the same result is obtained if the projection method is used together with the standard Lorenz-Mie equations (41–42).
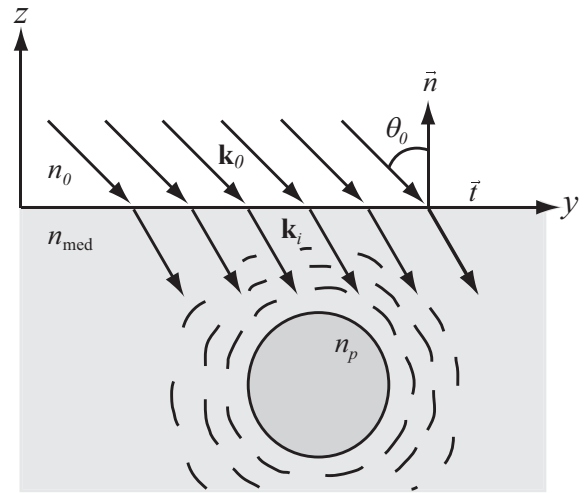

Fig. 7. Plane wave refracting into an absorbing medium at an oblique angle. The transmitted wave is then inhomogeneous, which influences how it is scattered by a particle.

The transmitted wave due to a homogeneous plane wave incident on an absorbing medium has its wave vector $\mathbf{k}_i$ in the plane of incidence. Even more specifically, the imaginary part $\mathbf{k}_i''$ is in the direction of the inward surface normal [17, 26]. In fact, we can find the wave vector of the transmitted wave using the generalized law of refraction [26, 27]:

$$\mathbf{k}_i = k_0 \left(\vec{t}\, n_0 \sin\theta_0 - \vec{n}\sqrt{n_{med}^2 - n_0^2 \sin^2\theta_0} \right),$$

where $\vec{n}$ is the surface normal and $\vec{t}$ is the surface tangent in the plane of incidence. The transmitted wave is most easily described by resolving it into a TE and a TM mode. The forward directions for these modes are given by the directions of the corresponding time-averaged Poynting vectors. These are $\mathbf{k}_i'$ for the TE mode and $\mathrm{Re}(\mathbf{k}_i/n_{med}^2)$ for the TM mode [26].

We can now construct an example of a plane wave refracting into an absorbing medium where it becomes inhomogeneous and is scattered by a particle (Fig. 7). Let us consider light of wavelength $\lambda = 530$ nm incident on a methanolic 0.4 mol/l rhodamine 6G solution with $n_{med} = 1.4 + i\,0.27$ [51]. Suppose this dye solution contains a 400 nm titanium dioxide particle of

refractive index $n_p = 2.5 + i\,1.4 \cdot 10^{-7}$ [52]. We then have a case that seems relevant in dye based random lasing [29–32]. The homogeneous wave in air ($n_0 = 1$) is incident on the surface of the dye solution at a 45° angle, thus $\theta_0 = \pi/4$. We choose a coordinate system with the surface normal in the z-direction and the yz-plane as the plane of incidence, such that $\vec{t}$ is in the y-direction. This is illustrated in Fig. 7, and we now have enough information to calculate $\mathbf{k}_i$, which we can insert in Eq. 45.

Let us illustrate the distribution of the scattered light using two special cases: (a) the scattering plane perpendicular to the plane of incidence but containing the forward direction; and (b) the scattering plane parallel to the plane of incidence. In the first case (a), the real and imaginary parts of the projected wave vector are both parallel with the forward direction. Thus the scattering components are the same as for a homogeneous wave of light ($\hat{\alpha} = 0$). The only difference in this case is in the slightly different forward directions for the TE and TM components. In the second case (b), we have $\mathbf{k} = \mathbf{k}_i$. In addition, the TE component defines $E_{\perp i}$ and the TM component defines $E_{\parallel i}$, so that the forward directions

$$\mathbf{S}_i^{\perp} = \mathbf{k}_i' \quad \text{and} \quad \mathbf{S}_i^{\parallel} = \mathrm{Re}\left(\frac{\mathbf{k}_i}{n_{\mathrm{med}}^2}\right)$$

relate to S_1 and S_2, respectively. These two cases (a and b) are plotted in Fig. 8, where the consequence of the wave inhomogeneity is seen both in the direction of the forward peak and in the shape of the smaller lobes. As opposed to the axial symmetry of conventional Lorenz-Mie theory, the scattering distribution here only exhibits mirror symmetry. The plane of incidence (yz) is the mirror plane in this example. In general, the mirror plane is the plane spanned by the real part and the imaginary part of the wave vector.

7. DISCUSSION AND CONCLUSION

I have introduced a practical solution for the scattering of an inhomogeneous plane wave by a particle in an absorbing medium. The solution simplifies to the existing solution for a homogeneous wave if inhomogeneity ($\hat{\alpha}$) is zero. It also simplifies to the existing solution for an inhomogeneous wave in a nonabsorbing medium if host medium absorption (n_{med}'') is zero. The new equations for the angular dependency of the scattering components (Equations 52–53) are easy to evaluate numerically using recurrence relations for the associated Legendre polynomials with argument zero (Equations 47–51). This solution seems a practical alternative to the numerical techniques for solving Maxwell's equations [36] and the more involved analytical solution [21, 22] that one would otherwise employ.

As is natural in a phase function context, my solution employs a far field approximation. This is done in two places: the assumption of a spherical wave (11) and the approximation of the distance-dependent Riccati-Bessel function ζ_n and its derivative (34). It is however worth noting that the far field assumption is not required for using the key insight that makes the solution practical. The key insight is that we can expand the incident wave in a way so that we only need to evaluate associated Legendre polynomials with argument zero (rather than complex valued argument). This practical expansion should also be applicable in a vector spherical harmonics context [22], where we would presumably get computationally attractive formulae for the electromagnetic field vectors without a far field approximation (but not expressions for the scattering components as
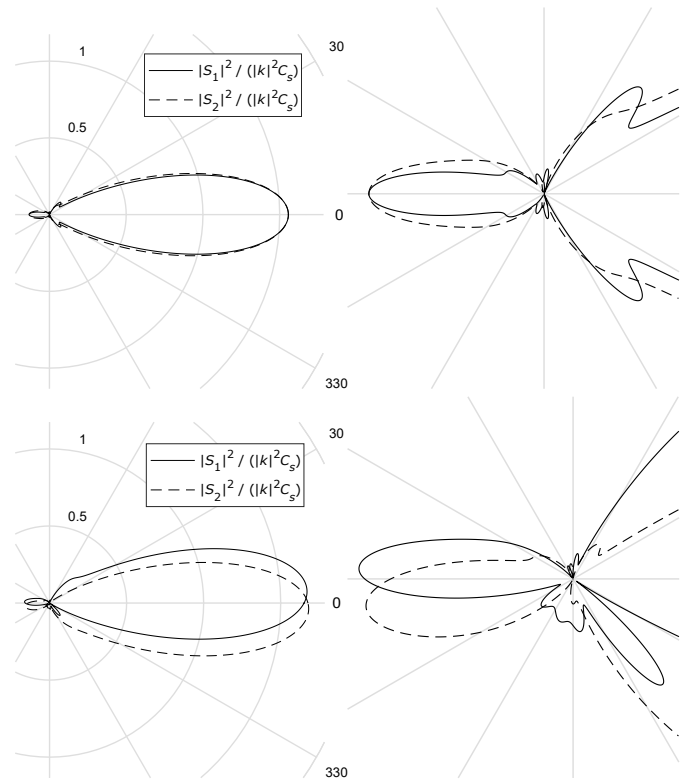

Fig. 8. Polarization components of the phase function for a spherical titanium dioxide particle submerged in a laser dye medium (as illustrated in Fig. 7). The rows contain the two cases (a and b) described in the main text. With the standard theory for homogeneous waves (Mie scattering), the result in the second row (b) would be the same as in the first row with swapped S_1 and S_2. The plots in the right column are closeups of the region around the origin of the plots in the left column.

presented here). I leave an investigation of a vector spherical harmonics solution based on this expansion for future work.

The provided phase function solution seems useful as plane waves are most often inhomogeneous when propagating in an absorbing medium. A radiative transfer simulation should account for the influence of this inhomogeneity on the directions of the scattered light. This is particularly relevant because the scattering of light by particles in absorbing media has recently found interesting applications in random lasing and solar cell absorption enhancement. I provided an example to illustrate how scattering is affected by wave inhomogeneity in a configuration similar to those seen in dye based random lasing.

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