An approach for a polychromatic generalized Lorenz-Mie theory

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Abstract

In this work a polychromatic version of the generalized Lorenz-Mie theory *stricto sensu* (GLMT) is derived. In this new formalism, arbitrary time-dependent fields are expanded into partial waves using Bromwich scalar potentials and, instead of the usual expansion coefficients – the beam shape coefficients (BSCs) – found in the monochromatic GLMT, now one finds *field shape spectra* (FSSs) which are intrinsically frequency-dependent. Expressions for the incident, scattered and internal fields are presented, and it is shown how physical quantities defined and expressed in the monochromatic GLMT in terms of the BSCs are modified and redefined in terms of the FSSs in polychromatic light scattering problems, like scattered intensities and phase angles, absorption, extinction and scattering cross-sections, and radiation pressure cross-sections.

Keywords: generalized Lorenz-Mie theory, light scattering, polychromatic light

1. Introduction

The experimental advent of the laser by Maiman, in 1960 [1], allowed physicists and engineers to produce, in a controlled manner, transversely localized coherent light, opening a new era of investigations in optics and photonics. Concomitantly, it also forced theoreticians to devise, among others, new formulations to incorporate such transversally-localized, Gaussian-like light into existing light scattering "theories", like the Lorenz-Mie theory (LMT) [2, 3, 4].

The LMT deals with the scattering and absorption of monochromatic plane waves by homogeneous, spherical particles of radius a of arbitrary value relatively to the wavelength λ of the incoming light. In this framework, the plane wave is expanded into a set of partial waves using spherical wave functions. Scattered and internal fields, absorption, scattering and extinction cross-sections and radiation pressure forces, among other physical quantities of possible interest, follows this infinite-sum structure, although truncations might become plausible depending on the ratio a/λ. Of course, the larger the number of non-negligible terms in the expansion, the more computationally demanding will be the calculations [5]. This number is determined by the size of the scatterer and its electromagnetic parameter relative to the host medium in which it is immersed, or might depend on the spatial domain of reconstruction, e.g., in computing the fields in a certain region.

The simplest extension of the LMT would be the one that replaces the non-physical plane waves by arbitrary-shaped beams like Gaussian-like beams (fundamental TEM_{00} mode) produced by laser sources [6, 7, 8, 9, 10, 11, 12]. Although light produced in this manner cannot rigorously be assumed monochromatic, today's technology allows us to speak of laser beams as single-frequency, temporal coherent wave fields without introducing observable theoretical deviations from practical light-matter problems. Instead of talking about a very narrow spectrum centered at λ, the laser source is assumed to emit light at that specific wavelength. This simplest extension of the LMT is widely known as the generalized LMT (GLMT) *stricto sensu* [13, 14, 15, 16], or simply GLMT in the context of the present work, see Refs. [17, 18, 19, 20, 21, 22] for review, historical and scientific facts.

From a theoretical point of view, the fundamental difference between the LMT and the GLMT lies on the expansion coefficients of the partial wave expansion, which now must be altered in magnitude and phase in order to accommodate for the new spatial field distribution [23, 24, 25, 26]. Such expansion coefficients, known in the literature as the beam shape coefficients (BSCs), carry, therefore, all the spatial nuances of an arbitrary-shaped, monochromatic beam, relative to a plane wave. Incidentally, the GLMT was first applied to Gaussian beams, since these were the most common type of laser sources available in optics laboratories during the 70's and 80's. It must be clear, however, that since the BSCs can be modeled at will, and that, regardless of their values, a true Maxwellian beam will always be produced, they apply to any envisioned structured light, contrary to past claims now firmly rebutted (see, for instance, the Introduction of Ref. [21]).

One of the most successful versions of the GLMT has its mathematical roots on Bromwich scalar potentials (BSPs) rather than on Hertz or magnetic vector potentials [15, 27, 28]. The BSPs can be defined for certain classes of coordinate systems for which the coordinates are separable and whose scale factors obey specific extra conditions. The spherical coordinate system meets all such required conditions. Although BSPs do not satisfy a wave equation, the electromagnetic fields can be easily extracted from them. In the GLMT, time-harmonic fields are taken for granted, so that the formalism is derived in terms of phasor fields starting from TM (Transverse Magnetic) and TE (Transverse Electric) BSPs assuming a fixed but otherwise arbitrary angular frequency ω.

Despite its recognized success over the past decades, the mathematical framework of the GLMT is still limited to monochromatic arbitrary-shaped beams. No systematic study is available showing the step-by-step procedure to incorporate polychromatic electromagnetic fields into the analysis of scattered intensities, phase angles, radiation pressure forces and torques, and so on. A formalism for such a *polychromatic* GLMT would help in the analysis, for instance, of the interaction between spherical particles with RGB (red, green, blue) laser system. Optical pulses with large bandwidth and their interaction with small particles could also be readily investigated, including femtosecond pulsed light generated by non-continuous wave lasers [29, 30, 31, 32, 33, 34, 35]. Finally, such a framework would also pave the way for extensions such as polychromatic Extended Boundary Condition Method (EBCM) – not to be confused with T-matrix method, insofar as GLMT, among others, is as well a T-matrix method [18, 20], polychromatic light-matter interactions with multilayered spheres, ellipsoidal and cylindrical particles, aggregates of scatterers and so on.

This paper is therefore devoted to the task of presenting a systematic approach to a polychromatic GLMT. The derivation is based on BSPs (which could be possibly translated to a more compact but less readable theory in terms of Vector Spherical Wave Functions, e.g. Ref. [18] and "Supplemental Material for the Second Edition" in Ref. [28]), and extended formulas for the incident, scattered and internal fields are presented in Section 2. As will be seen, the BSCs become, as they should, frequency-dependent, now called *field shape spectra* (FSSs). In Section 3, new expressions for scattered intensities and phase angles, extinction, scattering and absorption cross-sections, and radiation pressure cross-sections are derived which can be applied to continuous or discrete frequency-dependent incident optical fields. Finally, Section 4 is a conclusion.

2. Generalities

Let us start by assuming some basic notation for the GLMT for monochromatic arbitrary-shaped beams. In what follows, by GLMT we mean the GLMT *stricto sensu*, that is, valid for homogeneous spherical scatterers. A Cartesian coordinate system (x, y, z) is attached to a sphere of radius a, which is centered at its origin O_P. Corresponding cylindrical and spherical coordinate systems (ρ, φ, z) and (r, θ, φ) are assumed to be related to (x, y, z) as usual. The scatterer has a relative refractive index of $M = [\mu_{sp}\epsilon_{sp}/\mu\epsilon]^{1/2}$, μ and ϵ (μ_{sp} and ϵ_{sp}) being, respectively, the permeability and permittivity of the host medium (particle).

In the GLMT based on Bromwich Scalar Potentials (BSPs), we assume classes of orthogonal curvilinear coordinate systems with coordinates x^k ($k = 1, 2, 3$) and scale factors e_k, such that [28]:

$$e_1 = 1, \tag{1a}$$

$$\partial_1 \left(\frac{e_2}{e_3} \right) = 0, \tag{1b}$$

with $\partial_k \equiv \partial/\partial x^k$.

From Maxwell's equations in the time domain, BSPs $U(\mathbf{r}, t)$ must satisfy a differential equation of the form [28]:

$$\partial_1^2 U(\mathbf{r}, t) - \mu\epsilon\partial_t^2 U(\mathbf{r}, t) + \frac{1}{e_2 e_3} \left[\partial_2 \frac{e_3}{e_2} \partial_2 + \partial_3 \frac{e_2}{e_3} \partial_3 \right] U(\mathbf{r}, t) = 0. \tag{2}$$

For spherical coordinates, Eq. (2) can be specialized to read:

$$\partial_r^2 U(\mathbf{r}, t) - \mu\epsilon\partial_t^2 U(\mathbf{r}, t) + \frac{1}{r^2 \sin\theta} \partial_\theta \left[\sin\theta \partial_\theta U(\mathbf{r}, t) \right] + \frac{1}{r^2 \sin^2\theta} \partial_\varphi^2 U(\mathbf{r}, t) = 0. \tag{3}$$

As is well known from the GLMT, Eq. (3) is not a wave equation for $U(\mathbf{r}, t)$. However, this partial differential equation do admit solutions of the form:

$$U(\mathbf{r}, t) = rR(kr)\Theta(\theta)\Phi(\varphi)T(t). \tag{4}$$

Notice the explicit appearance of time in the present approach, which contrasts with the one adopted in the traditional GLMT for monochromatic fields [15]. Since we are working explicitly in the time domain with arbitrary instantaneous fields, a temporal function $T(t)$ has been explicitly considered in the method of separation of variables.

Substituting Eq. (4) in (3), dividing the resulting expression by $U(\mathbf{r},t)$ and temporarily omitting the arguments, one then has:

$$\frac{1}{rR}d_r^2\left(rR\right)-\mu\epsilon\frac{1}{T}d_t^2T+\frac{1}{r^2\sin\theta}\frac{1}{\Theta}d_\theta\left[\sin\theta d_\theta\Theta\right]+\frac{1}{r^2\sin^2\theta}\frac{1}{\Phi}d_\varphi^2\Phi=0. \quad (5)$$

In Eq. (5), the notation $d_k^i = d^i/dx^k$ has been introduced, in which x^k now stands for r, θ, φ and t. Isolating the term containing T,

$$\frac{1}{rR}d_r^2\left(rR\right)+\frac{1}{r^2\sin\theta}\frac{1}{\Theta}d_\theta\left[\sin\theta d_\theta\Theta\right]+\frac{1}{r^2\sin^2\theta}\frac{1}{\Phi}d_\varphi^2\Phi=\mu\epsilon\frac{1}{T}d_t^2T. \quad (6)$$

The r.h.s. of Eq. (6) depends exclusively on t, while the l.h.s. depends on other independent spatial variables. Therefore, both sides must be equal to a possibly complex constant which we shall call $-k^2$, so that:

$$\mu\epsilon\frac{1}{T}d_t^2T=-k^2 \quad (7)$$

After rearranging Eq. (7) and introducing the definition $k^2 \equiv k^2(\omega) = \omega^2\mu\epsilon$:

$$d_t^2T+\omega^2T=0. \quad (8)$$

As expected, the solutions to the temporal contribution in the method of separation of variables are simple harmonic functions $\exp(i\omega t)$, where ω is the angular frequency and, therefore, a continuous variable in general.

Having solved for $T(t)$, Eq. (5) is now multiplied by r^2 and rearranged so as to read as:

$$\frac{r}{R}d_r^2\left(rR\right)+k^2r^2=-\frac{1}{\sin\theta}\frac{1}{\Theta}d_\theta\left[\sin\theta d_\theta\Theta\right]-\frac{1}{\sin^2\theta}\frac{1}{\Phi}d_\varphi^2\Phi=0. \quad (9)$$

Except for the fact that we are not assuming time-harmonic functions with a particular frequency $\omega=\omega_0$, Eq. (9) is identical in form with Eq. (2.64) of Ref. [28], so that the solution to the above equation can be directly reproduced from Eq. (2.91) in the aforementioned reference.

Letting $U(\mathbf{r},\omega) = rR(kr)\Theta(\theta)\Phi(\varphi)$ [remember that $k \equiv k(\omega)$], one then has:

$$U(\mathbf{r},\omega) = \sum_{n=1}^{\infty}\sum_{m=-n}^{+n} \frac{c_{nm}}{k} \begin{pmatrix} \psi_n(kr) \\ \xi_n(kr) \end{pmatrix} P_n^{|m|}(\cos\theta)e^{im\varphi}. \tag{10}$$

In Eq. (10), $\psi_n(z) = zj_n(z)$ and $\xi_n(z) = zh_n^{(2)}(z)$ are Ricatti-Bessel functions, with $j_n(z)$ and $h_n^{(2)}(z)$ being, respectively, spherical Bessel functions of the first kind and spherical Hankel functions of the second kind (the latter functions may also be called spherical Bessel functions of the fourth kind, see Chapter 2 in Ref. [28]). The associated Legendre functions $P_n^{|m|}(\cos\theta)$ are defined in accordance with Hobson's notation [36]. Finally, the pre-factors c_{nm} depend on n, m and possibly on k or, equivalently, ω.

Therefore, the general form for time-dependent BSPs $U(\mathbf{r},t)$ is given by:

$$\begin{aligned} U(\mathbf{r},t) =& U(\mathbf{r},\omega)e^{i\omega t} \\ =& \sum_{n=1}^{\infty}\sum_{m=-n}^{+n} \frac{c_{nm}}{k} \begin{pmatrix} \psi_n(kr) \\ \xi_n(kr) \end{pmatrix} P_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}, \end{aligned} \tag{11}$$

which is simply the usual BSP in the monochromatic GLMT multiplied by the time-harmonic factor $\exp(i\omega t)$.

At this point, one may define a *generalized* BSP by adding up all possible solutions to the partial differential equation, Eq. (6):

$$U(\mathbf{r},t) = \sum_{j}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} \frac{c_{nm}}{k} \begin{pmatrix} \psi_n(kr) \\ \xi_n(kr) \end{pmatrix} P_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}. \tag{12}$$

In Eq. (12), the symbol $\sum_j$ can actually represent either discrete or continuous (integrals) sums, or compositions of both. In general, therefore, one could then propose:

$$\begin{aligned} U(\mathbf{r},t) =& \sum_{j=1}^{J}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} \frac{c_{nm}^j}{k_j} \begin{pmatrix} \psi_n(k_j r) \\ \xi_n(k_j r) \end{pmatrix} P_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega_j t} \\ &+ \int_{-\infty}^{\infty}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} \frac{c_{nm}(\omega)}{k(\omega)} \begin{pmatrix} \psi_n[k(\omega)r] \\ \xi_n[k(\omega)r] \end{pmatrix} P_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \end{aligned} \tag{13}$$

or

$$U(\mathbf{r},t) = \sum_{j=1}^{J} U_d(\mathbf{r},\omega_j)e^{i\omega_j t} + \int_{-\infty}^{\infty} U_c(\mathbf{r},\omega)e^{i\omega t}d\omega. \tag{14}$$

In Eq. (13), $c_{nm}^j \equiv c_{nm}(\omega_j)$ and $k_j \equiv k(\omega_j) = \omega_j\sqrt{\mu(\omega_j)\epsilon(\omega_j)} = 2\pi/\lambda_j$, with $\lambda_j \equiv \lambda(\omega_j)$ being the wavelength of the j-th monochromatic field with angular frequency ω_j. Notice that we have explicitly assumed that the constituent parameters of the host medium might change with (angular) frequency. The lower indices "d" and "c" stands for *discrete* and *continuous* BSPs $U_d(\mathbf{r}, \omega_j)$ and $U_c(\mathbf{r}, \omega)$, respectively, whose general forms can be taken from Eq. (13).

Equation (13), or (14), is the most general form for BSPs for light fields composed of both sums of monochromatic (single frequency) beams and polychromatic optical waves. However, since

$$\sum_{j=1}^{J} U_d(\mathbf{r}, \omega_j) e^{i\omega_j t} = \int_{-\infty}^{\infty} \left[\sum_{j=1}^{J} U_d(\mathbf{r}, \omega_j)\delta(\omega - \omega_j)\right] e^{i\omega t} d\omega, \tag{15}$$

where $\delta(z)$ is the Dirac delta function, the term under brackets in Eq. (15) can be merged with $U_c(\mathbf{r}, \omega)$ in Eq. (14), allowing us to define the BSP spectra $U(\mathbf{r}, \omega)$ as follows:

$$U(\mathbf{r}, t) = \int_{-\infty}^{\infty} U(\mathbf{r}, \omega) e^{i\omega t} d\omega. \tag{16}$$

As given by either Eq. (14) or (16), $U(\mathbf{r}, t)$ can be viewed as a *generalized* BSP. The second term in the r.h.s. of Eq. (14) corresponds, except for an absent multiplicative factor of $1/2\pi$, to the inverse Fourier transform of $U_c(\mathbf{r}, \omega)$, that is, of the continuous part of $U(\mathbf{r}, t)$. Once the discrete part of the generalized BSP, $U_d(\mathbf{r}, \omega)$, is incorporated into a single spectra $U(\mathbf{r}, \omega)$, Eq. (16) is simply a Fourier transformation process of an arbitrary polychromatic BSP, as expected. Notice that, in writing the r.h.s. of Eq. (15) we have explicitly forced ω_j (and not ω) to emphasize that the BSP is discrete from the outset, although nothing prevents us to write $U_d(\mathbf{r}, \omega)$ instead. However, this would give the false idea to the reader that in the $\sum_{j=1}^{J}$-sum we are actually working with a BSP which is continuous in ω and then picking up specific contributions from $\omega = \omega_j$.

The generalized BSP can be split into TM and TE modes by simply appending lower indices "TM" and "TE", respectively. At this point, the formulation can be carried out in one of the two ways. First, we use the TM and TE generalized BSPs and try to find the polychromatic, physical fields. The second approach is to find the latter fields from the TM and TE monochromatic BSPs. As we will justify later, we prefer to follow the second path and reserve the superposition principle to be applied to the (physical) fields satisfying Maxwell's equation.

2.1. Incident EM fields in the polychromatic GLMT

From Maxwell's equations and the classes of orthogonal curvilinear coordinate systems constrained by Eq. (1), the following relations are established between EM fields $\mathbf{E}(\mathbf{r}, t)$ and $\mathbf{H}(\mathbf{r}, t)$, and the generalized BSP $U(\mathbf{r}, t)$, for both TM and TE modes, see Eqs. (2.23)–(2.35) in Ref. [28]:

$$E_{1,TM} = \partial_1^2 U_{TM} - \mu\epsilon\partial_t^2 U_{TM}, \tag{17}$$

$$E_{2,TM} = \frac{1}{e_2}\partial_1\partial_2 U_{TM}, \tag{18}$$

$$E_{3,TM} = \frac{1}{e_3}\partial_1\partial_3 U_{TM}, \tag{19}$$

$$H_{1,TM} = 0, \text{ (TM-definition)}, \tag{20}$$

$$H_{2,TM} = \frac{\epsilon}{e_3}\partial_3\partial_t U_{TM}, \tag{21}$$

$$H_{3,TM} = -\frac{\epsilon}{e_2}\partial_2\partial_t U_{TM}, \tag{22}$$

for TM modes, and

$$E_{1,TE} = 0, \text{ (TE-definition)}, \tag{23}$$

$$E_{2,TE} = -\frac{\mu}{e_3}\partial_3\partial_t U_{TE}, \tag{24}$$

$$E_{3,TE} = \frac{\mu}{e_2}\partial_2\partial_t U_{TE}, \tag{25}$$

$$H_{1,TE} = \partial_1^2 U_{TE} - \mu\epsilon\partial_t^2 U_{TE} \tag{26}$$

$$H_{2,TE} = \frac{1}{e_2}\partial_1\partial_2 U_{TE}, \tag{27}$$

$$H_{3,TE} = \frac{1}{e_3}\partial_1\partial_3 U_{TE} \tag{28}$$

for TE modes. In Eqs. (17)–(28), arguments have been omitted for simplicity, and the total electromagnetic field components are found from

$$E_k = E_{k,TM} + E_{k,TE}, \ k = 1, 2, 3 \tag{29a}$$

$$H_k = H_{k,TM} + H_{k,TE}, \ k = 1, 2, 3. \tag{29b}$$

As we mentioned in the previous section, using the superposition principle to define a generalized BSP is not very useful when $\mu \equiv \mu(\omega)$ and $\epsilon \equiv \epsilon(\omega)$, that is, when the constitutive parameters are frequency-dependent quantities in general. This can be readily seen from Eqs. (17), (21), (22), (24), (25) and (26), where multiplicative factors proportional to μ and ϵ are present which cannot be immediately absorbed inside the integral in Eq. (16) and become part of the integrand. Since it is $\mathbf{E}$ and $\mathbf{H}$ the physical fields of interest, we then first consider the monochromatic solutions to the electromagnetic field components and then use the superposition principle to establish their polychromatic, partial wave expressions.

Therefore, for the incident monochromatic fields, from Eq. (11) one readily writes,

$$U^i_{TX}(\mathbf{r}, t) = U^i_{TX}(\mathbf{r}, \omega)e^{i\omega t}, \tag{30}$$

where TX stands for either TM or TE.

Following the conventions in the monochromatic GLMT, let us further define:

$$c_{nm,TX} \equiv c_{nm,TX}(\omega) = c^{pw}_n(\omega)G^m_{n,TX}(\omega), \tag{31}$$

where

$$c^{pw}_n(\omega) = \frac{1}{ik(\omega)}(-i)^n \frac{2n+1}{n(n+1)}. \tag{32}$$

In Eq. (31), both field strengths E_0 and H_0 available from the monochromatic GLMT have been absorbed into $G^m_{n,TX}(\omega)$. Equation (32) can be seen as the natural generalization of c^{pw}_n in the monochromatic GLMT, see Eqs. (3.1)–(3.3) in Ref. [28], in order to deal with frequency-dependent fields.

Because Ricatti-Bessel functions $\xi_n(z)$ cannot be used to describe incident fields since it is not finite at the origin, from Eqs. (11), (31) and (32), the TM and TE instantaneous (complex) incident BSPs for monochromatic fields are then written as:

$$U^i_{TX}(\mathbf{r}, t) = \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} c^{pw}_n(\omega)G^m_{n,TX}(\omega)\frac{\psi_n[k(\omega)r]}{k(\omega)} P^{|m|}_n(\cos\theta)e^{im\varphi}e^{i\omega t}, \tag{33}$$

which obviously coincides with Eqs. (3.4) and (3.5) in Ref. [28], except for the fact that the dependence on ω is explicitly put into evidence. Therefore, from the superposition principle, and making use of Eqs. (3.39)–(3.50) of Ref. [28], which corresponds to substituting Eq. (33) into (17)–(28) using spherical coordinates, one then obtains the general expressions for the partial wave expansion of polychromatic beams for TM modes:

$$\begin{aligned} E^i_{r,TM}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} E^i_{r,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega \\ &= \int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} k(\omega)c_n^{pw}(\omega)G^m_{n,TM}(\omega) \\ &\quad \times \left[\psi''_n[k(\omega)r] + \psi_n[k(\omega)r]\right] P_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \end{aligned} \tag{34}$$

$$\begin{aligned} E^i_{\theta,TM}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} E^i_{\theta,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega \\ &= \int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} c_n^{pw}(\omega)G^m_{n,TM}(\omega)\frac{\psi'_n[k(\omega)r]}{r}\tau_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \end{aligned} \tag{35}$$

$$\begin{aligned} E^i_{\varphi,TM}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} E^i_{\varphi,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega \\ &= i\int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} mc_n^{pw}(\omega)G^m_{n,TM}(\omega)\frac{\psi'_n[k(\omega)r]}{r}\pi_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \end{aligned} \tag{36}$$

$$H^i_{r,TM}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} H^i_{r,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega = 0, \tag{37}$$

$$\begin{aligned} H^i_{\theta,TM}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} H^i_{\theta,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega \\ &= -\int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} mc_n^{pw}(\omega)\frac{G^m_{n,TM}(\omega)}{\eta(\omega)}\frac{\psi_n[k(\omega)r]}{r}\pi_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \end{aligned} \tag{38}$$

$$H^{i}_{\varphi,TM}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} H^{i}_{\varphi,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega$$

$$= -\, i\int\limits_{-\infty}^{\infty}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} c_n^{pw}(\omega)\frac{G^{m}_{n,TM}(\omega)}{\eta(\omega)}\frac{\psi_n[k(\omega)r]}{r}\tau_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega. \tag{39}$$

Notice that Eqs. (34)-(36) are expressed in terms of $G^{m}_{n,TM} = E_0(\omega)g^{m}_{n,TM}$. Therefore, in Eqs. (38) and (39), we have to introduce the intrinsic impedance $\eta(\omega) = [\mu(\omega)/\epsilon(\omega)]^{1/2}$ so that, using Eq. (1.107) of Ref. [28], we have $G^{m}_{n,TM}/\eta = H_0 g^{m}_{n,TM}$. Note that, for similar reason, the impedance has to be introduced in Eqs. (41) and (42) below.

Similarly, for TE modes,

$$E^{i}_{r,TE}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} E^{i}_{r,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega = 0, \tag{40}$$

$$E^{i}_{\theta,TE}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} E^{i}_{\theta,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega$$

$$= \int\limits_{-\infty}^{\infty}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} m c_n^{pw}(\omega)\eta(\omega)G^{m}_{n,TE}(\omega)\frac{\psi_n[k(\omega)r]}{r}\pi_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \tag{41}$$

$$E^{i}_{\varphi,TE}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} E^{i}_{\varphi,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega$$

$$= i\int\limits_{-\infty}^{\infty}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} c_n^{pw}(\omega)\eta(\omega)G^{m}_{n,TE}(\omega)\frac{\psi_n[k(\omega)r]}{r}\tau_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \tag{42}$$

$$
\begin{aligned}
H^i_{r,TE}(\mathbf{r},t) &= \int_{-\infty}^{\infty} H^i_{r,TE}(\mathbf{r},\omega) e^{i\omega t} d\omega \\
&= \int_{-\infty}^{\infty} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} k(\omega) c_n^{pw}(\omega) G^m_{n,TE}(\omega) \\
&\quad \times \left[\psi''_n[k(\omega)r] + \psi_n[k(\omega)r]\right] P_n^{|m|}(\cos\theta) e^{im\varphi} e^{i\omega t} d\omega,
\end{aligned}
\tag{43}
$$

$$
\begin{aligned}
H^i_{\theta,TE}(\mathbf{r},t) &= \int_{-\infty}^{\infty} H^i_{\theta,TE}(\mathbf{r},\omega) e^{i\omega t} d\omega \\
&= \int_{-\infty}^{\infty} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} c_n^{pw}(\omega) G^m_{n,TE}(\omega) \frac{\psi'_n[k(\omega)r]}{r} \tau_n^{|m|}(\cos\theta) e^{im\varphi} e^{i\omega t} d\omega,
\end{aligned}
\tag{44}
$$

$$
\begin{aligned}
H^i_{\varphi,TE}(\mathbf{r},t) &= \int_{-\infty}^{\infty} H^i_{\varphi,TE}(\mathbf{r},\omega) e^{i\omega t} d\omega \\
&= i \int_{-\infty}^{\infty} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} m c_n^{pw}(\omega) G^m_{n,TE}(\omega) \frac{\psi'_n[k(\omega)r]}{r} \pi_n^{|m|}(\cos\theta) e^{im\varphi} e^{i\omega t} d\omega,
\end{aligned}
\tag{45}
$$

In Eqs. (34)–(45), $\pi_n^m(\cos\theta) = P_n^m(\cos\theta)/\sin\theta$ and $\tau_n^m(\cos\theta) = d_\theta P_n^m(\cos\theta)$ are generalized Legendre functions [28]. These equations are the polychromatic generalization of the monochromatic Eqs. (3.39)–(3.50) in Ref. [28].

It is seen from Eqs. (34)–(45) that $G^m_{n,TX}(\omega)$ are frequency-dependent functions. For discrete-frequency, polychromatic light, they are distributions proportional to the BSCs of the original, monochromatic GLMT, as will soon be shown. The proportionality depends on the field strengths $E_0(\omega_j)$ and $H_0(\omega_j)$, respectively for electric and magnetic fields, of the j-th monochromatic arbitrary-shaped beam.

For continuous frequency-dependent fields, $G^m_{n,TX}(\omega)$ are functions of a continuous variable ω and represent FSSs. They become, therefore, a generalization of the BSCs of the original GLMT, now extended to polychromatic beams.

2.2. Quadratures for the FSSs

In this section we shall find general expressions to evaluate the FSSs $G_{n,TX}^{m}(\omega)$ by assuming arbitrary polychromatic fields which can be either discretely or continuously dependent on ω. To do so, orthogonal relations for the φ-exponential, $\exp(im\varphi)$, and associated Legendre functions, $P_n^{|m|}(\cos\theta)$, are imposed, as usual. Like in the monochromatic GLMT, it suffices to consider the radial field components $E_r^i(\mathbf{r},t)$ and $H_r^i(\mathbf{r},t)$ in Eq. (29), now with the lower index k of Eq. (29) representing the radial spherical coordinate r.

The total instantaneous radial field components $E_r^i(\mathbf{r},t)$ and $H_r^i(\mathbf{r},t)$ are found from Eqs. (29), (34), (37), (40) and (43):

$$\begin{aligned} E_r^i(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} E_r^i(\mathbf{r},\omega)e^{i\omega t}d\omega \\ &= \int\limits_{-\infty}^{\infty} \left\{ \sum_{n=1}^{\infty}\sum_{m=-n}^{+n} n(n+1)c_n^{pw}(\omega)G_{n,TM}^{m}(\omega)\frac{\psi_n[k(\omega)r]}{k(\omega)r^2}P_n^{|m|}(\cos\theta)e^{im\varphi}\right\} e^{i\omega t}d\omega \end{aligned} \tag{46}$$

and

$$\begin{aligned} H_r^i(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} H_r^i(\mathbf{r},\omega)e^{i\omega t}d\omega \\ &= \int\limits_{-\infty}^{\infty} \left\{ \sum_{n=1}^{\infty}\sum_{m=-n}^{+n} n(n+1)c_n^{pw}(\omega)G_{n,TE}^{m}(\omega)\frac{\psi_n[k(\omega)r]}{k(\omega)r^2}P_n^{|m|}(\cos\theta)e^{im\varphi}\right\} e^{i\omega t}d\omega, \end{aligned} \tag{47}$$

where we have used Ricatti-Bessel's differential equation $z^2\psi_n''(z)+[z^2-n(n+1)]\psi_n(z)=0$ to simplify the terms under brackets in Eqs. (34) and (43).

After introducing the following orthogonality relations for exponentials and associated Legendre functions [37]:

$$\int\limits_{0}^{2\pi} \exp[i(m-m')\varphi]d\varphi = 2\pi\delta_{m,m'}, \tag{48}$$

and

$$\int_0^{\pi} P_n^{|m|}(\cos\theta) P_{n'}^{|m|}(\cos\theta)\sin\theta d\theta = \frac{2}{2n+1}\frac{(n+|m|)!}{(n-|m|)!}\delta_{n,n'}, \tag{49}$$

where $\delta_{i,j}$ is the Kronecker delta, Eqs. (46) and (47) can be recast under the form:

$$\begin{aligned}\int_{-\infty}^{\infty} & c_n^{pw}(\omega) G_{n,TM}^m(\omega)\frac{\psi_n[k(\omega)r]}{k(\omega)} e^{i\omega t} d\omega \\ &= \frac{r^2}{4\pi}\frac{2n+1}{n(n+1)}\frac{(n-|m|)!}{(n+|m|)!}\int_0^{\pi}\int_0^{2\pi} E_r^i(\mathbf{r},t) P_n^{|m|}(\cos\theta) e^{-im\varphi}\sin\theta d\varphi d\theta.\end{aligned} \tag{50}$$

and

$$\begin{aligned}\int_{-\infty}^{\infty} & c_n^{pw}(\omega) G_{n,TE}^m(\omega)\frac{\psi_n[k(\omega)r]}{k(\omega)} e^{i\omega t} d\omega \\ &= \frac{r^2}{4\pi}\frac{2n+1}{n(n+1)}\frac{(n-|m|)!}{(n+|m|)!}\int_0^{\pi}\int_0^{2\pi} H_r^i(\mathbf{r},t) P_n^{|m|}(\cos\theta) e^{-im\varphi}\sin\theta d\varphi d\theta.\end{aligned} \tag{51}$$

Now, multiplying both sides of Eqs. (50) and (51) by $\exp(-i\omega' t)$, integrating over time, interchanging the order of the integrals in the l.h.s. of the resulting expression and using the fact that, for continuous variables ω,

$$\int_{-\infty}^{\infty} \exp[i(\omega-\omega')t] dt = 2\pi\delta(\omega-\omega'), \tag{52}$$

the following expressions for the FSSs $G_{n,TM}^m(\omega)$ and $G_{n,TE}^m(\omega)$ are then obtained:

$$\begin{aligned}G_{n,TM}^m(\omega) =& \frac{r^2}{8\pi^2 c_n^{pw}(\omega)}\frac{2n+1}{n(n+1)}\frac{(n-|m|)!}{(n+|m|)!}\frac{k(\omega)}{\psi_n[k(\omega)r]} \\ & \times \int_0^{\pi}\int_0^{2\pi}\left[\int_{-\infty}^{\infty} E_r^i(\mathbf{r},t) e^{-i\omega t} dt\right] P_n^{|m|}(\cos\theta) e^{-im\varphi}\sin\theta d\varphi d\theta\end{aligned} \tag{53}$$

and

$$G_{n,TE}^{m}(\omega) = \frac{r^2}{8\pi^2 c_n^{pw}(\omega)} \frac{2n+1}{n(n+1)} \frac{(n-|m|)!}{(n+|m|)!} \frac{k(\omega)}{\psi_n[k(\omega)r]} \times \int_0^{\pi} \int_0^{2\pi} \left[\int_{-\infty}^{\infty} H_r^i(\mathbf{r},t) e^{-i\omega t} dt \right] P_n^{|m|}(\cos\theta) e^{-im\varphi} \sin\theta d\varphi d\theta. \tag{54}$$

Since the time integrals in Eqs. (53) and (54) correspond, in accordance with Eq. (16) extended to the electric and magnetic fields, to 2π times the Fourier transforms $E_r^i(\mathbf{r},\omega)$ and $H_r^i(\mathbf{r},\omega)$ of $E_r^i(\mathbf{r},t)$ and $H_r^i(\mathbf{r},t)$, respectively, one finally gets the double quadrature formulae for the FSSs:

$$G_{n,TM}^{m}(\omega) = \frac{1}{4\pi c_n^{pw}(\omega)} \frac{2n+1}{n(n+1)} \frac{(n-|m|)!}{(n+|m|)!} \frac{k(\omega) r^2}{\psi_n[k(\omega)r]} \times \int_0^{\pi} \int_0^{2\pi} E_r^i(\mathbf{r},\omega) P_n^{|m|}(\cos\theta) e^{-im\varphi} \sin\theta d\varphi d\theta \tag{55}$$

and

$$G_{n,TE}^{m}(\omega) = \frac{1}{4\pi c_n^{pw}(\omega)} \frac{2n+1}{n(n+1)} \frac{(n-|m|)!}{(n+|m|)!} \frac{k(\omega) r^2}{\psi_n[k(\omega)r]} \times \int_0^{\pi} \int_0^{2\pi} H_r^i(\mathbf{r},\omega) P_n^{|m|}(\cos\theta) e^{-im\varphi} \sin\theta d\varphi d\theta. \tag{56}$$

Equations (55) and (56) represent the generalized quadrature procedure to evaluate the FSSs in the polychromatic GLMT. In comparison with the monochromatic GLMT, frequency-dependent quantities become explicitly evident and, when no analytical solution exists to the time integrals in Eqs. (53) and (54), an additional integral over time is present which must be solved numerically. For electromagnetic fields constructed from discrete superpositions of J monochromatic beams of distinct frequencies ω_j (for example, a colorful intensity pattern created from a RGB laser system), expressing the physical, instantaneous fields in the form seen in the first term of the r.h.s. of Eq. (14) allows us to write

$$E_r^i(\mathbf{r},t) = \sum_{j=1}^{J} E_r^i(\mathbf{r},\omega_j) e^{i\omega_j t} \tag{57}$$

$$H_r^i(\mathbf{r}, t) = \sum_{j=1}^{J} H_r^i(\mathbf{r}, \omega_j) e^{i\omega_j t}, \tag{58}$$

and, multiplying both sides of Eqs. (57) and (58) by $\exp(-i\omega t)$, integrating with respect to time and making use of Eq. (52), one obtains:

$$\begin{aligned} \int_{-\infty}^{\infty} \left\{ \begin{array}{c} E_r^i(\mathbf{r}, t) \\ H_r^i(\mathbf{r}, t) \end{array} \right\} e^{-i\omega t} dt =& 2\pi \sum_{j=1}^{J} \left\{ \begin{array}{c} E_r^i(\mathbf{r}, \omega_j) \\ H_r^i(\mathbf{r}, \omega_j) \end{array} \right\} \delta(\omega - \omega_j) \\ =& 2\pi \left\{ \begin{array}{c} E_r^i(\mathbf{r}, \omega) \\ H_r^i(\mathbf{r}, \omega) \end{array} \right\} \end{aligned} \tag{59}$$

In Eqs. (57)–(59), $E_r^i(\mathbf{r}, \omega_j)$ and $H_r^i(\mathbf{r}, \omega_j)$ correspond to the electric and magnetic field phasors of the monochromatic incident field of frequency $\omega = \omega_j$.

Therefore, from Eqs. (53), (54), (59),

$$\begin{aligned} G_{n,TM}^m(\omega) =& \sum_{j=1}^{J} \frac{1}{4\pi c_n^{pw}(\omega)} \frac{2n+1}{n(n+1)} \frac{(n-|m|)!}{(n+|m|)!} \frac{k(\omega) r^2}{\psi_n[k(\omega) r]} \\ & \times \int_0^{\pi} \int_0^{2\pi} E_r^i(\mathbf{r}, \omega_j) P_n^{|m|}(\cos\theta) e^{-im\varphi} \sin\theta d\varphi d\theta \delta(\omega - \omega_j) \\ =& \sum_{j=1}^{J} G_{n,TM}^m(\omega_j) \delta(\omega_j - \omega) \\ =& \sum_{j=1}^{J} E_0(\omega_j) g_{n,TM}^{m,mono}(\omega_j) \delta(\omega_j - \omega) \end{aligned} \tag{60}$$

and

$$
\begin{aligned}
G_{n,TE}^{m}(\omega) = &\sum_{j=1}^{J} \frac{1}{4\pi c_n^{pw}(\omega)} \frac{2n+1}{n(n+1)} \frac{(n-|m|)!}{(n+|m|)!} \frac{k(\omega)r^2}{\psi_n[k(\omega)r]} \\
&\times \int_0^{\pi} \int_0^{2\pi} H_r^i(\mathbf{r},\omega) P_n^{|m|}(\cos\theta) e^{-im\varphi} \sin\theta d\varphi d\theta \delta(\omega-\omega_j) \\
= &\sum_{j=1}^{J} G_{n,TE}^{m}(\omega_j)\delta(\omega_j-\omega) \\
= &\sum_{j=1}^{J} H_0(\omega_j) g_{n,TE}^{m,mono}(\omega_j)\delta(\omega_j-\omega),
\end{aligned}
\tag{61}
$$

where, for discrete frequencies ω_j, $g_{n,TX}^{m,mono}(\omega_j)$ are the usual BSCs in the monochromatic GLMT, $E_0(\omega_j)$ is a normalization electric field strength for TM modes and $H_0(\omega_j)$ is a normalization magnetic field strength for TE modes.

Equations (60) and (61) express the fact that the FSSs for discrete superpositions of distinct, arbitrary-shaped time-harmonic fields, each of which having a particular and arbitrary frequency ω_j, is the weighted sum of the BSCs associated with each time-harmonic field. The sum is actually weighted since $E_r^i(\mathbf{r},\omega_j)$ and $H_r^i(\mathbf{r},\omega_j)$ can possess different relative amplitudes. In other words, the FSSs $G_{n,TX}^{m}(\omega)$ for discrete sums of monochromatic electromagnetic fields of distinct frequencies ω_j can be expressed as a train of impulses at the discrete frequencies ω_j, each impulse weighted by a complex amplitude $X_0(\omega_j)g_{n,TX}^{m}(\omega_j)$, in which $X_0(\omega_j) = E_0(\omega_j)$ for TM modes and $X_0(\omega_j) = H_0(\omega_j)$ for TE modes. Equations (60) and (61), once substituted in Eqs. (34)–(45), recover Eqs. (57) and (58), as expected.

For $\omega_j = \omega_0$, $\forall j$, that is, when all distinct, arbitrary-shaped fields carry the same operating frequency (e.g., when generated by a single laser source), the ω_j arguments can be dropped and the FSSs then become simple weighted sums of equal-frequency BSCs representing a total monochromatic arbitrary-shaped beam, such a problem lying therefore in the realm of the monochromatic GLMT.

Before ending this section, it must be stressed that, as it stands, the monochromatic GLMT using BSPs expands the electromagnetic fields into partial waves using spherical wave functions in spherical coordinates. This leads to BSPs of the form given by Eq. (11) for a fixed but arbitrary ω, where $U(\mathbf{r},\omega)$ depends on both ω (an arbitrary but otherwise fixed frequency) and the spatial coordinates. However, it is common practice to express the instantaneous physical fields in terms of spectra of the form $S(\mathbf{k},\omega)$, that is, by also transforming the spatial coordinates and going to a reciprocal space in the wave vector $\mathbf{k}$. For instance, paraxial, scalar Gaussian pulses can be represented by a spectrum $S(k_\rho,\omega)$, where k_ρ is the transverse wave number which obeys the dispersion relation $k^2=\omega^2/c^2=k_\rho^2+k_z^2$, with k_z being the longitudinal wave number. Then, the corresponding scalar field $\psi(\rho,z,t)$ is determined from the following relation [38]:

$$\psi(\rho,z,t)=\int_0^\infty dk_\rho\int_{-\infty}^{+\infty}d\omega\,k_\rho S(k_\rho,\omega)J_0(k_\rho\rho)\exp\left(-i\sqrt{k^2-k_\rho^2}z\right)e^{i\omega t},\quad(62)$$

where $J_0(.)$ is the zero-order cylindrical Bessel function of the first kind.

Extending this process to vector fields then would lead to another representation of Eqs. (55) and (56) involving integrations over k_ρ (or, in view of the dispersion relation, over k_z) with arguments involving spectral functions $E_r^i(\mathbf{k},\omega)$ and $H_r^i(\mathbf{k},\omega)$.

2.3. Scattered and internal fields

The procedure to find the scattered and internal field components resembles that adopted for the incident fields.

First, we introduce the instantaneous scattered and internal monochromatic TM and TE BSPs in accordance with Eq. (30):

$$U_{TX}^s(\mathbf{r},t)=U_{TX}^s(\mathbf{r},\omega)e^{i\omega t},\quad(63)$$

$$U_{TX}^{sp}(\mathbf{r},t)=U_{TX}^{sp}(\mathbf{r},\omega)e^{i\omega t}.\quad(64)$$

In Eqs. (63) and (64), the upper indices "s" and "sp" stands for scattered and internal (sphere) fields, respectively. In addition, $U_{TX}^s(\mathbf{r},\omega)$ and $U_{TX}^{sp}(\mathbf{r},\omega)$ are the BSP phasors in the monochromatic GLMT, see Eqs. (3.35)–(3.38), (3.84)–(3.87) of Ref. [28]. They are here reproduced for the convenience of the reader:

$$U_{TM}^s(\mathbf{r},\omega)=-\sum_{n=1}^{\infty}\sum_{m=-n}^{+n}\frac{1}{k}c_n^{pw}a_nG_{n,TM}^m\xi_n(kr)P_n^{|m|}(\cos\theta)e^{im\varphi},\quad(65)$$

$$U_{TE}^{s}(\mathbf{r},\omega) = -\sum_{n=1}^{\infty}\sum_{m=-n}^{+n}\frac{1}{k}c_n^{pw}b_n G_{n,TE}^{m}\xi_n(kr)P_n^{|m|}(\cos\theta)e^{im\varphi}, \tag{66}$$

$$U_{TM}^{sp}(\mathbf{r},\omega) = \sum_{n=1}^{\infty}\sum_{m=-n}^{+n}\frac{k}{k_{sp}^2}c_n^{pw}c_n G_{n,TM}^{m}\psi_n(k_{sp}r)P_n^{|m|}(\cos\theta)e^{im\varphi}, \tag{67}$$

$$U_{TE}^{sp}(\mathbf{r},\omega) = \sum_{n=1}^{\infty}\sum_{m=-n}^{+n}\frac{k}{k_{sp}^2}c_n^{pw}d_n G_{n,TE}^{m}\psi_n(k_{sp}r)P_n^{|m|}(\cos\theta)e^{im\varphi}. \tag{68}$$

where a_n, b_n, c_n and d_n are the classical scattering coefficients of the Mie theory [4, 28, 39]:

$$a_n = \frac{\mu_{sp}\psi_n(\alpha)\psi_n'(\beta) - \mu M\psi_n'(\alpha)\psi_n(\beta)}{\mu_{sp}\xi_n(\alpha)\psi_n'(\beta) - \mu M\xi_n'(\alpha)\psi_n(\beta)}, \tag{69}$$

$$b_n = \frac{\mu M\psi_n(\alpha)\psi_n'(\beta) - \mu_{sp}\psi_n'(\alpha)\psi_n(\beta)}{\mu M\xi_n(\alpha)\psi_n'(\beta) - \mu_{sp}\xi_n'(\alpha)\psi_n(\beta)}, \tag{70}$$

$$c_n = \frac{\mu_{sp}M\left[\xi_n(\alpha)\psi_n'(\alpha) - \xi_n'(\alpha)\psi_n(\alpha)\right]}{\mu_{sp}\xi_n(\alpha)\psi_n'(\beta) - \mu M\xi_n'(\alpha)\psi_n(\beta)}, \tag{71}$$

$$d_n = \frac{\mu M^2\left[\xi_n(\alpha)\psi_n'(\alpha) - \xi_n'(\alpha)\psi_n(\alpha)\right]}{\mu M\xi_n(\alpha)\psi_n'(\beta) - \mu_{sp}\xi_n'(\alpha)\psi_n(\beta)}, \tag{72}$$

where

$$\alpha = ka = \frac{2\pi a}{\lambda} \tag{73a}$$

$$\beta = k_{sp}a = Mka = M\alpha. \tag{73b}$$

Notice that Eqs. (69)–(73) have been written for a specific but otherwise arbitrary single operating frequency ω (arguments omitted). For a fixed scatterer, the Mie coefficients are frequency-dependent functions, since μ_{sp}, M, α and β might also become functions of ω.

Substitution of Eqs. (63)–(68) into (17)–(28) and making use of the superposition principle for the electromagnetic fields then leads to the scattered and internal field components for TM and TE modes.

For the scattered fields, one obtains

$$\begin{aligned} E^s_{r,TM}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} E^s_{r,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega \\ &= -\int\limits_{-\infty}^{\infty}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} k(\omega)c_n^{pw}(\omega)a_n(\omega)G^m_{n,TM}(\omega) \\ &\quad\times\left[\xi''_n[k(\omega)r]+\xi_n[k(\omega)r]\right]P_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \end{aligned} \tag{74}$$

$$\begin{aligned} E^s_{\theta,TM}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} E^s_{\theta,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega \\ &= -\int\limits_{-\infty}^{\infty}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} c_n^{pw}(\omega)a_n(\omega)G^m_{n,TM}(\omega)\frac{\xi'_n[k(\omega)r]}{r}\tau_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \end{aligned} \tag{75}$$

$$\begin{aligned} E^s_{\varphi,TM}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} E^s_{\varphi,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega \\ &= -i\int\limits_{-\infty}^{\infty}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} mc_n^{pw}(\omega)a_n(\omega)G^m_{n,TM}(\omega)\frac{\xi'_n[k(\omega)r]}{r}\pi_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \end{aligned} \tag{76}$$

$$H^s_{r,TM}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} H^s_{r,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega = 0, \tag{77}$$

$$\begin{aligned} H^s_{\theta,TM}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} H^s_{\theta,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega \\ &= \int\limits_{-\infty}^{\infty}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} mc_n^{pw}(\omega)a_n(\omega)\frac{G^m_{n,TM}(\omega)}{\eta(\omega)}\frac{\xi_n[k(\omega)r]}{r}\pi_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \end{aligned} \tag{78}$$

$$H^s_{\varphi,TM}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} H^s_{\varphi,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega$$
$$= i \int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} c_n^{pw}(\omega)a_n(\omega)\frac{G^m_{n,TM}(\omega)}{\eta(\omega)}\frac{\xi_n[k(\omega)r]}{r}\tau_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \tag{79}$$

for TM modes, and

$$E^s_{r,TE}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} E^s_{r,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega = 0, \tag{80}$$

$$E^s_{\theta,TE}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} E^s_{\theta,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega$$
$$= -\int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} mc_n^{pw}(\omega)b_n(\omega)\eta(\omega)G^m_{n,TE}(\omega)\frac{\xi_n[k(\omega)r]}{r}\pi_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \tag{81}$$

$$E^s_{\varphi,TE}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} E^s_{\varphi,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega$$
$$= -i\int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} c_n^{pw}(\omega)b_n(\omega)\eta(\omega)G^m_{n,TE}(\omega)\frac{\xi_n[k(\omega)r]}{r}\tau_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \tag{82}$$

$$\begin{aligned} H^s_{r,TE}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} H^s_{r,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega \\ &= -\int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} k(\omega)c_n^{pw}(\omega)b_n(\omega)G^m_{n,TE}(\omega) \\ &\quad \times \left[\xi_n''[k(\omega)r] + \xi_n[k(\omega)r]\right]P_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \end{aligned} \tag{83}$$

$$H^s_{\theta,TE}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} H^s_{\theta,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega$$
$$= -\int\limits_{-\infty}^{\infty}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} c_n^{pw}(\omega)b_n(\omega)G^m_{n,TE}(\omega)\frac{\xi'_n[k(\omega)r]}{r}\tau_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \tag{84}$$

$$H^s_{\varphi,TE}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} H^s_{\varphi,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega$$
$$= -i\int\limits_{-\infty}^{\infty}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} mc_n^{pw}(\omega)b_n(\omega)G^m_{n,TE}(\omega)\frac{\xi'_n[k(\omega)r]}{r}\pi_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \tag{85}$$

for TE modes.

Similarly, for the internal fields,

$$\begin{aligned} E^{sp}_{r,TM}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} E^{sp}_{r,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega \\ &= \int\limits_{-\infty}^{\infty}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} k(\omega)c_n^{pw}(\omega)c_n(\omega)G^m_{n,TM}(\omega) \\ &\quad\times\left[\psi''_n[k_{sp}(\omega)r]+\psi_n[k_{sp}(\omega)r]\right]P_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \end{aligned} \tag{86}$$

$$E^{sp}_{\theta,TM}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} E^{sp}_{\theta,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega$$
$$= \int\limits_{-\infty}^{\infty}\sum_{n=1}^{\infty}\sum_{m=-n}^{+n} \frac{k(\omega)}{k_{sp}(\omega)}c_n^{pw}(\omega)c_n(\omega)G^m_{n,TM}(\omega)\frac{\psi'_n[k_{sp}(\omega)r]}{r}\tau_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega, \tag{87}$$

$$
\begin{aligned}
E^{sp}_{\varphi,TM}(\mathbf{r},t) =& \int\limits_{-\infty}^{\infty} E^{sp}_{\varphi,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega \\
=& i\int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty}\sum_{m=-n}^{+n} m\frac{k(\omega)}{k_{sp}(\omega)}c_n^{pw}(\omega)c_n(\omega)G^m_{n,TM}(\omega) \\
&\times \frac{\psi_n'[k_{sp}(\omega)r]}{r}\pi_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega,
\end{aligned}
\tag{88}
$$

$$
H^{sp}_{r,TM}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} H^{sp}_{r,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega = 0,
\tag{89}
$$

$$
\begin{aligned}
H^{sp}_{\theta,TM}(\mathbf{r},t) =& \int\limits_{-\infty}^{\infty} H^{sp}_{\theta,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega \\
=& -\int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty}\sum_{m=-n}^{+n} m\frac{\mu(\omega)}{\mu_{sp}(\omega)}c_n^{pw}(\omega)c_n(\omega)\frac{G^m_{n,TM}(\omega)}{\eta(\omega)} \\
&\times \frac{\psi_n[k_{sp}(\omega)r]}{r}\pi_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega,
\end{aligned}
\tag{90}
$$

$$
\begin{aligned}
H^{sp}_{\varphi,TM}(\mathbf{r},t) =& \int\limits_{-\infty}^{\infty} H^{sp}_{\varphi,TM}(\mathbf{r},\omega)e^{i\omega t}d\omega \\
=& -i\int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty}\sum_{m=-n}^{+n} \frac{\mu(\omega)}{\mu_{sp}(\omega)}c_n^{pw}(\omega)c_n(\omega)\frac{G^m_{n,TM}(\omega)}{\eta(\omega)} \\
&\times \frac{\psi_n[k_{sp}(\omega)r]}{r}\tau_n^{|m|}(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega,
\end{aligned}
\tag{91}
$$

for TM modes, and

$$
E^{sp}_{r,TE}(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} E^{sp}_{r,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega = 0,
\tag{92}
$$

$$
\begin{aligned}
E^{sp}_{\theta,TE}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} E^{sp}_{\theta,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega \\
&= \int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty}\sum_{m=-n}^{+n} m\frac{\mu_{sp}(\omega)}{\mu(\omega)}\frac{k^2(\omega)}{k^2_{sp}(\omega)}c^{pw}_n(\omega)d_n(\omega)\eta(\omega)G^m_{n,TE}(\omega) \\
&\quad\times\frac{\psi_n[k_{sp}(\omega)r]}{r}\pi^{|m|}_n(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega,
\end{aligned} \tag{93}
$$

$$
\begin{aligned}
E^{sp}_{\varphi,TE}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} E^{sp}_{\varphi,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega \\
&= i\int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty}\sum_{m=-n}^{+n} \frac{\mu_{sp}(\omega)}{\mu(\omega)}\frac{k^2(\omega)}{k^2_{sp}(\omega)}c^{pw}_n(\omega)d_n(\omega)\eta(\omega)G^m_{n,TE}(\omega) \\
&\quad\times\frac{\psi_n[k_{sp}(\omega)r]}{r}\tau^{|m|}_n(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega,
\end{aligned} \tag{94}
$$

$$
\begin{aligned}
H^{sp}_{r,TE}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} H^{sp}_{r,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega \\
&= \int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty}\sum_{m=-n}^{+n} k(\omega)c^{pw}_n(\omega)d_n(\omega)G^m_{n,TE}(\omega) \\
&\quad\times\left[\psi''_n[k_{sp}(\omega)r]+\psi_n[k_{sp}(\omega)r]\right]P^{|m|}_n(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega,
\end{aligned} \tag{95}
$$

$$
\begin{aligned}
H^{sp}_{\theta,TE}(\mathbf{r},t) &= \int\limits_{-\infty}^{\infty} H^{sp}_{\theta,TE}(\mathbf{r},\omega)e^{i\omega t}d\omega \\
&= \int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty}\sum_{m=-n}^{+n} \frac{k(\omega)}{k_{sp}(\omega)}c^{pw}_n(\omega)d_n(\omega)G^m_{n,TE}(\omega) \\
&\quad\times\frac{\psi'_n[k_{sp}(\omega)r]}{r}\tau^{|m|}_n(\cos\theta)e^{im\varphi}e^{i\omega t}d\omega,
\end{aligned} \tag{96}
$$

$$
\begin{aligned}
H^{sp}_{\varphi,TE}(\mathbf{r},t) =& \int\limits_{-\infty}^{\infty} H^{sp}_{\varphi,TE}(\mathbf{r},\omega) e^{i\omega t} d\omega \\
=& i\int\limits_{-\infty}^{\infty} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} m \frac{k(\omega)}{k_{sp}(\omega)} c_n^{pw}(\omega) d_n(\omega) G^m_{n,TE}(\omega) \\
& \times \frac{\psi'_n[k_{sp}(\omega)r]}{r} \pi_n^{|m|}(\cos\theta) e^{im\varphi} e^{i\omega t} d\omega,
\end{aligned}
\tag{97}
$$

for TE modes.

3. Physical quantities in the polychromatic GLMT

In the monochromatic GLMT, several physical quantities of practical importance can be expressed in terms of the usual BSCs. Among them we find, for instance, scattered intensities, phase angles, absorption, scattering and extinction cross sections, radiation pressure forces and torques.

It is instructive to investigate the changes occurred in such quantities and re-express them when one goes from single time-harmonic to arbitrary time-dependent fields. It must be stressed that, since physical quantities in the monochromatic GLMT are calculated from time-averaged fields, a general procedure for extending them to polychromatic fields can be devised, as we now show.

3.1. Scattered Intensities

The scattered intensities are found from time-averaging, for the far-fields, the instantaneous Poynting vector $\mathbf{S}(\mathbf{r},t) = \mathbf{E}^s(\mathbf{r},t) \times \mathbf{H}^s(\mathbf{r},t)$. Because instantaneous fields are real quantities, one can use the relations $\mathbf{E}^s(\mathbf{r},t) = [\mathbf{E}^s(\mathbf{r},t) + \mathbf{E}^{s*}(\mathbf{r},t)]/2$ and $\mathbf{H}^s(\mathbf{r},t) = [\mathbf{H}^s(\mathbf{r},t) + \mathbf{H}^{s*}(\mathbf{r},t)]/2$ so that, after some rearrangement, it follows that:

$$
\mathbf{S}(\mathbf{r},t) = \frac{1}{2}\,\mathrm{Re}\,[\mathbf{E}^s(\mathbf{r},t) \times \mathbf{H}^s(\mathbf{r},t)] + \frac{1}{2}\,\mathrm{Re}\,[\mathbf{E}^s(\mathbf{r},t) \times \mathbf{H}^{s*}(\mathbf{r},t)]\,. \tag{98}
$$

From the first lines of Eqs. (34)–(45), (74)–(85) and (86)–(97), one writes:

$$
\mathbf{E}^k(\mathbf{r},t) = \int\limits_{-\infty}^{\infty} \mathbf{E}^k(\mathbf{r},\omega) e^{i\omega t} d\omega, \tag{99}
$$

$$\mathbf{H}^k(\mathbf{r},t) = \int_{-\infty}^{\infty} \mathbf{H}^k(\mathbf{r},\omega) e^{i\omega t} d\omega, \tag{100}$$

where $k = i, s, sp$ for incident, scattered and sphere fields, respectively.

In view of Eqs. (99) and (100), the cross product $\mathbf{E}^s(\mathbf{r},t) \times \mathbf{H}^s(\mathbf{r},t)$ in Eq. (98) can be expressed as:

$$\mathbf{E}^s(\mathbf{r},t) \times \mathbf{H}^s(\mathbf{r},t) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \mathbf{E}^s(\mathbf{r},\omega) \times \mathbf{H}^s(\mathbf{r},\omega') e^{i(\omega+\omega')t} d\omega d\omega' \tag{101}$$

Once averaged over time, the term in the r.h.s. of Eq. (101) will be proportional to an integral of the form

$$\lim_{T\to\infty} \frac{1}{T} \int_{-T/2}^{T/2} e^{i(\omega+\omega')t} dt = \begin{cases} 1, & \omega = -\omega' \\ 0, & \text{otherwise} \end{cases}. \tag{102}$$

For scattered fields with non-negative frequencies, the cross product $\mathbf{E}^s(\mathbf{r},\omega) \times \mathbf{H}^s(\mathbf{r},\omega)$ will contain only positive frequency components, that is, ω and ω' will both be positive. Therefore, the use of Eq. (102) together with the fact that $\omega > 0$ and $\omega' > 0$, imply that Eq. (101) will average to zero.

Now, for the second term in the r.h.s. of Eq. (98), it is seen that

$$\mathbf{E}^s(\mathbf{r},t) \times \mathbf{H}^{s*}(\mathbf{r},t) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \mathbf{E}^s(\mathbf{r},\omega) \times \mathbf{H}^{s*}(\mathbf{r},\omega') e^{i(\omega-\omega')t} d\omega d\omega'. \tag{103}$$

Similarly, since

$$\lim_{T\to\infty} \frac{1}{T} \int_{-T/2}^{T/2} e^{i(\omega-\omega')t} dt = \begin{cases} 1, & \omega = \omega' \\ 0, & \text{otherwise} \end{cases}, \tag{104}$$

for physical scattered fields carrying no non-negative frequencies, once averaged over time, the term in the r.h.s. of Eq. (103) will simplify to a single integral (either in ω or ω') and, using Eq. (98), we will eventually be left with:

$$\begin{aligned}\langle \mathbf{S}(\mathbf{r},t)\rangle =&\lim_{T\to\infty}\frac{1}{T}\int\limits_{-T/2}^{+T/2}\mathbf{S}(\mathbf{r},t)dt\\ =&\left\langle \frac{1}{2}\,\mathrm{Re}\left[\mathbf{E}^s(\mathbf{r},t)\times\mathbf{H}^{s*}(\mathbf{r},t)\right]\right\rangle\\ =&\frac{1}{2}\,\mathrm{Re}\left\{\int\limits_{-\infty}^{\infty}\mathbf{E}^s(\mathbf{r},\omega)\times\mathbf{H}^{s*}(\mathbf{r},\omega)d\omega\right\},\end{aligned}\tag{105}$$

or, more compactly,

$$\langle \mathbf{S}(\mathbf{r},t)\rangle = \mathrm{Re}\left[\int\limits_{-\infty}^{+\infty}\mathbf{S}(\mathbf{r},\omega)d\omega\right].\tag{106}$$

with

$$\mathbf{S}(\mathbf{r},\omega)=\frac{1}{2}\mathbf{E}^s(\mathbf{r},\omega)\times\mathbf{H}^{s*}(\mathbf{r},\omega).\tag{107}$$

From Eqs. (74)–(85), after imposing the far-field asymptotic expressions [28]

$$\xi_n(z)\to i^{n+1}e^{-ikr},\tag{108a}$$

$$\xi_n''(z)+\xi_n(z)=0,\tag{108b}$$

one then has the following non-normalized scattered intensity for polychromatic fields:

$$\begin{aligned}S_c\equiv&S_c(\omega)=\mathrm{Re}\left[\mathbf{S}(\mathbf{r},\omega)\right]\\ =&\frac{1}{2}\,\mathrm{Re}\left[E^s_\theta(\mathbf{r},\omega)H^{s*}_\varphi(\mathbf{r},\omega)-E^s_\varphi(\mathbf{r},\omega)H^{s*}_\theta(\mathbf{r},\omega)\right]\\ =&\frac{1}{2}\frac{1}{\eta(\omega)k^2(\omega)r^2}(|\mathcal{S}_{2,c}|^2+|\mathcal{S}_{1,c}|^2)\\ =&\frac{1}{2\eta(\omega)k^2(\omega)r^2}|\mathcal{S}_{2,c}|^2+\frac{1}{2\eta(\omega)k^2(\omega)r^2}|\mathcal{S}_{1,c}|^2\\ =&I_{\theta,c}+I_{\varphi,c},\end{aligned}\tag{109}$$

where

$$I_{\theta,c} \equiv I_{\theta,c}(\omega) = \frac{1}{2\eta(\omega)k^2(\omega)r^2}|\mathcal{S}_{2,c}|^2 \tag{110}$$

and

$$I_{\varphi,c} \equiv I_{\varphi,c}(\omega) = \frac{1}{2\eta(\omega)k^2(\omega)r^2}|\mathcal{S}_{1,c}|^2 \tag{111}$$

In Eqs. (109)–(111), the $1/2\eta(\omega)$ comes from the fact that the normalization condition of Eq. (3.106) of Ref. [28] was not considered. Also, the generalized amplitude functions $\mathcal{S}_{1,c}(\omega)$ and $\mathcal{S}_{2,c}(\omega)$ are given by:

$$\begin{aligned}\mathcal{S}_{1,c} \equiv \mathcal{S}_{1,c}(\omega) = \sum_{n=1}^{\infty}\sum_{m=-n}^{n}\frac{2n+1}{n(n+1)}\Big[ma_n(\omega)G_{n,TM}^m(\omega)\pi_n^{|m|}(\cos\theta) \\ + ib_n(\omega)\eta(\omega)G_{n,TE}^m(\omega)\tau_n^{|m|}(\cos\theta)\Big]e^{im\varphi},\end{aligned} \tag{112}$$

$$\begin{aligned}\mathcal{S}_{2,c} \equiv \mathcal{S}_{2,c}(\omega) = \sum_{n=1}^{\infty}\sum_{m=-n}^{n}\frac{2n+1}{n(n+1)}\Big[a_n(\omega)G_{n,TM}^m(\omega)\tau_n^{|m|}(\cos\theta) \\ + imb_n(\omega)\eta(\omega)G_{n,TE}^m(\omega)\pi_n^{|m|}(\cos\theta)\Big]e^{im\varphi}.\end{aligned} \tag{113}$$

In Eqs. (112) and (113), the FSSs $G_{n,TM}^m(\omega)$ and $G_{n,TE}^m(\omega)$ can represent discrete or continuous frequency-dependent incident fields, or both. Notice that the second line of Eq. (109) corresponds to Eq. (3.105) of Ref. [28] for fixed but otherwise arbitrary positive frequencies, and that Eqs. (112) and (113) re-express their monochromatic versions as given by Eqs. (3.110) and (3.111) of Ref. [28], now in terms of the FSSs $G_{n,TM}^m(\omega)$ and $G_{n,TE}^m(\omega)$.

In conclusion, in view of Eqs. (106), (109)–(113), the total scattered intensity, S, can be expressed as:

$$S = \int_{-\infty}^{\infty} S_c d\omega = \int_{-\infty}^{\infty} \left[I_{\theta,c} + I_{\varphi,c}\right] d\omega = I_\theta + I_\varphi, \tag{114}$$

where

$$\begin{bmatrix} I_\theta \\ I_\varphi \end{bmatrix} = \frac{1}{r^2}\int_{-\infty}^{\infty}\begin{bmatrix} \frac{|\mathcal{S}_{2,c}|^2}{2\eta(\omega)k^2(\omega)} \\ \frac{|\mathcal{S}_{1,c}|^2}{2\eta(\omega)k^2(\omega)} \end{bmatrix} d\omega \tag{115}$$

This extends the scattered intensities in the monochromatic GLMT to the more general case of arbitrary frequency-dependent fields in the polychromatic GLMT.

3.2. Phase angles

Phase angles can be used to describe the state of polarization of a given scattered electromagnetic field in the far field. Since the angular field components are the only non-negligible components in the far field, the angle associated with the ratio E_θ^s/E_φ^s can be used as a measure of such polarization.

To see it clearly, consider Eqs. (74)–(85) in the far field, that is, with the asymptotic expressions given by Eq. (108). Following usual notation presented, e.g., in Sec. 3.10 of Ref. [28], omitting arguments and making use of Eqs. (32), (112) and (113), one finds:

$$E_\theta^s = i \int_{-\infty}^{\infty} \frac{e^{-ikr}}{kr} \mathcal{S}_{2,c} d\omega = \mathcal{E}_\theta, \tag{116a}$$

$$E_\varphi^s = -\int_{-\infty}^{\infty} \frac{e^{-ikr}}{kr} \mathcal{S}_{1,c} d\omega = \mathcal{E}_\varphi, \tag{116b}$$

In Eq. (116),

$$\mathcal{E}_\theta = i \int_{-\infty}^{\infty} \frac{e^{-ikr}}{kr} \mathcal{S}_{2,c} d\omega = i\mathcal{S}_2, \tag{117a}$$

$$\mathcal{E}_\varphi = -\int_{-\infty}^{\infty} \frac{e^{-ikr}}{kr} \mathcal{S}_{1,c} d\omega = -\mathcal{S}_1, \tag{117b}$$

with $\mathcal{S}_1$ and $\mathcal{S}_2$ being the non-normalized, total generalized amplitude functions.

Let δ denote the phase angle between the total field components E_θ^s and E_φ^s, which may be used to characterize the state of polarization of the total polychromatic beam. Then, from Eq. (117) and defining

$$\mathcal{E}_\theta = i\mathcal{S}_2 = A_\theta e^{i\varphi_2} \tag{118a}$$

$$\mathcal{E}_\varphi = -\mathcal{S}_1 = A_\varphi e^{i\varphi_1}, \tag{118b}$$

where A_θ and A_φ are real amplitude functions and φ_2 and φ_1 are the respective phase angles of $\mathcal{E}_\theta$ and $\mathcal{E}_\varphi$, it is then seen that

$$\begin{aligned} \tan\delta &= \tan(\varphi_2 - \varphi_1) \\ &= \frac{\mathrm{Re}(\mathcal{S}_1)\,\mathrm{Re}(\mathcal{S}_2) + \mathrm{Im}(\mathcal{S}_1)\,\mathrm{Im}(\mathcal{S}_2)}{\mathrm{Im}(\mathcal{S}_1)\,\mathrm{Re}(\mathcal{S}_2) - \mathrm{Re}(\mathcal{S}_1)\,\mathrm{Im}(\mathcal{S}_2)}. \end{aligned} \tag{119}$$

Equation (119) is identical in form with the one derived in the monochromatic GLMT, see Eq. (3.114) in Ref. [28]. However, now $\mathcal{S}_1$ and $\mathcal{S}_2$ are defined in accordance with Eq. (117), that is, they carry the spherical wave factor $\exp(-ikr)/kr$ inside the integral over ω.

When the incident field is of the form given by Eqs. (57) and (58), substitution of the corresponding FSSs of Eqs. (60) and (61) into (117), with the aid of (112) and (113), leads to the simplified forms

$$\begin{aligned}\mathcal{S}_1 &= \int_{-\infty}^{\infty} \frac{e^{-ik(\omega)r}}{k(\omega)r} \mathcal{S}_{1,c}(\omega) d\omega \\ &= \int_{-\infty}^{\infty} \frac{e^{-ik(\omega)r}}{k(\omega)r} \sum_{j=1}^{J} E_0(\omega_j) \mathcal{S}_1^{mono}(\omega_j) \delta(\omega - \omega_j) d\omega \\ &= \sum_{j=1}^{J} \frac{e^{-ik(\omega_j)r}}{k(\omega_j)r} E_0(\omega_j) \mathcal{S}_1^{mono}(\omega_j)\end{aligned} \tag{120}$$

and

$$\begin{aligned}\mathcal{S}_2 &= \int_{-\infty}^{\infty} \frac{e^{-ik(\omega)r}}{k(\omega)r} \mathcal{S}_{2,c}(\omega) d\omega \\ &= \int_{-\infty}^{\infty} \frac{e^{-ik(\omega)r}}{k(\omega)r} \sum_{j=1}^{J} E_0(\omega_j) \mathcal{S}_2^{mono}(\omega_j) \delta(\omega - \omega_j) d\omega \\ &= \sum_{j=1}^{J} \frac{e^{-ik(\omega_j)r}}{k(\omega_j)r} E_0(\omega_j) \mathcal{S}_2^{mono}(\omega_j),\end{aligned} \tag{121}$$

where $\mathcal{S}_1^{mono}(\omega_j) = \mathcal{S}_{1,c}(\omega = \omega_j)/E_0(\omega_j)$ and $\mathcal{S}_2^{mono}(\omega_j) = \mathcal{S}_{2,c}(\omega = \omega_j)/E_0(\omega_j)$ are the normalized, generalized amplitude functions in the monochromatic GLMT, see also Eqs. (3.106), (3.110) and (3.111) of Ref. [28], for a particular operating frequency ω_j. Obviously, for $J = 1$, the identification between the polychromatic and the monochromatic GLMT is complete, and the phase angle δ as given by Eq. (119) will coincide with that in the latter framework, as it should.

3.3. Extinction, scattering and absorption cross-sections

Other physical quantities of interest in the polychromatic GLMT include the scattering, extinction and absorbing cross sections $\mathcal{C}_{sca}$, $\mathcal{C}_{ext}$ and $\mathcal{C}_{abs}$, respectively, which are calculated by considering the total radial Poynting vector and integrating it over a spherical surface centered at the origin of the coordinate system (where the center of the scatterer is located).

The results are straightforward, but we shall present them for clarity. Let C_k^{mono} ($k = abs$, sca or ext) denote the normalized cross sections familiar from the monochromatic GLMT, see Eqs.(3.106), (3.124), (3.137) and (3.142) in Ref. [28]. Proceeding similarly to Sec. 3.1, one eventually establishes that, for arbitrary-shaped, polychromatic fields in free space,

$$\mathcal{C}_{abs} = \mathcal{C}_{ext} - \mathcal{C}_{sca} \tag{122}$$

$$\begin{aligned}\mathcal{C}_{sca} =& \int_{-\infty}^{\infty} \frac{|E_0(\omega)|^2}{2\eta(\omega)} C_{sca}^{mono}(\omega) d\omega \\ =& \frac{1}{\pi} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \frac{2n+1}{n(n+1)} \frac{(n+|m|)!}{(n-|m|)!} \\ & \times \int_{-\infty}^{\infty} \frac{\lambda^2(\omega)}{2\eta(\omega)} \left(|a_n(\omega)|^2 \left| G_{n,TM}^m(\omega) \right|^2 + |b_n(\omega)|^2 \eta^2(\omega) \left| G_{n,TE}^m(\omega) \right|^2 \right) d\omega,\end{aligned} \tag{123}$$

$$\begin{aligned}\mathcal{C}_{ext} =& \int_{-\infty}^{\infty} \frac{|E_0(\omega)|^2}{2\eta(\omega)} C_{ext}^{mono}(\omega) d\omega \\ =& \frac{1}{\pi} \operatorname{Re} \left[\sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \frac{2n+1}{n(n+1)} \frac{(n+|m|)!}{(n-|m|)!} \right. \\ & \left. \times \int_{-\infty}^{\infty} \frac{\lambda^2(\omega)}{2\eta(\omega)} \left(a_n(\omega) \left| G_{n,TM}^m(\omega) \right|^2 + b_n(\omega) \eta^2(\omega) \left| G_{n,TE}^m(\omega) \right|^2 \right) d\omega \right],\end{aligned} \tag{124}$$

with simplifications again being possible for discrete sums of monochromatic fields of different or equal frequencies, further simplified for single monochromatic fields.

3.4. Radiation pressure cross-sections

As a final example of a physical quantity extended to polychromatic fields, we consider radiation pressure cross sections $C_{pr,i}$ in Cartesian coordinates

$(i = x, y, z)$. In the monochromatic GLMT, such radiation pressure cross-sections can be found, e.g., in Eqs. (3.159), (3.181) and (3.185) of Ref. [28].

Assuming that the incident light propagates along $+z$, as usual, in the polychromatic GLMT the new $C_{pr,i}$ will read as:

$$
\begin{aligned}
C_{pr,z} =& \frac{1}{\pi} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \frac{1}{(n+1)^2} \frac{(n+1+|m|)!}{(n-|m|)!} \int_{-\infty}^{\infty} \frac{\lambda^2(\omega)}{2\eta(\omega)} \\
& \times \Bigg\{ \mathrm{Re} \Big[\big(a_n(\omega) + a_{n+1}^*(\omega) - 2a_n(\omega) a_{n+1}^*(\omega) \big) G_{n,TM}^m(\omega) G_{n+1,TM}^{m*}(\omega) \\
& + \big(b_n(\omega) + b_{n+1}^*(\omega) - 2b_n(\omega) b_{n+1}^*(\omega) \big) \eta^2(\omega) G_{n,TE}^m(\omega) G_{n+1,TE}^{m*}(\omega) \Big] \\
& + m \frac{2n+1}{n^2(n+1)^2} \frac{(n+|m|)!}{(n-|m|)!} \\
& \times \mathrm{Re} \left[i(2a_n(\omega) b_n^*(\omega) - a_n(\omega) - b_n^*(\omega)) G_{n,TM}^m(\omega) \eta(\omega) G_{n,TE}^{m*}(\omega) \right] \Bigg\} d\omega
\end{aligned}
\tag{125}
$$

for the longitudinal component, and

$$
\begin{aligned}
\left\{ \begin{array}{c} C_{pr,x} \\ C_{pr,y} \end{array} \right\} =& \frac{1}{2\pi} \sum_{p=1}^{\infty} \sum_{n=p}^{\infty} \sum_{m=p-1 \neq 0}^{\infty} \frac{(n+p)!}{(n-p)!} \\
& \times \Bigg\{ \left(\frac{1}{m^2} \delta_{m,n+1} - \frac{1}{n^2} \delta_{n,m+1} \right) \\
& \times \int_{-\infty}^{\infty} \frac{\lambda^2(\omega)}{2\eta(\omega)} \left\{ \begin{array}{c} \mathrm{Re} \\ \mathrm{Im} \end{array} \right\} \left[S_{mn}^{p-1}(\omega) + S_{nm}^{-p}(\omega) - 2U_{mn}^{p-1}(\omega) - 2U_{nm}^{-p}(\omega) \right] d\omega \\
& + \frac{2n+1}{n^2(n+1)^2} \delta_{n,m} \\
& \times \int_{-\infty}^{\infty} \frac{\lambda^2(\omega)}{2\eta(\omega)} \left\{ \begin{array}{c} \mathrm{Re} \\ \mathrm{Im} \end{array} \right\} \left[T_{mn}^{p-1}(\omega) - T_{nm}^{-p}(\omega) - 2V_{mn}^{p-1}(\omega) + 2V_{nm}^{-p}(\omega) \right] d\omega \Bigg\}
\end{aligned}
\tag{126}
$$

In Eqs. (125) and (126), the expressions for S_{nm}^p, T_{nm}^p, U_{nm}^p and V_{nm}^p are similar to those available from the monochromatic GLMT [see Eqs. (3.167), (3.168), (3.177) and (3.178) in Ref. [28], a typo being corrected in the last term of Eq. (3.178)]:

$$\begin{aligned} S^{p}_{nm}(\omega) = & \left[a_n(\omega) + a^*_m(\omega)\right] G^{p}_{n,TM}(\omega) G^{p+1*}_{m,TM}(\omega) \\ & + \left[b_n(\omega) + b^*_m(\omega)\right] \eta^2(\omega) G^{p}_{n,TE}(\omega) G^{p+1*}_{m,TE}(\omega), \end{aligned} \tag{127}$$

$$\begin{aligned} T^{p}_{nm}(\omega) = & - i \left[a_n(\omega) + b^*_m(\omega)\right] G^{p}_{n,TM}(\omega) \eta(\omega) G^{p+1*}_{m,TE}(\omega) \\ & + i \left[b_n(\omega) + a^*_m(\omega)\right] \eta(\omega) G^{p}_{n,TE}(\omega) G^{p+1*}_{m,TM}(\omega), \end{aligned} \tag{128}$$

$$\begin{aligned} U^{p}_{nm}(\omega) = & a_n(\omega) a^*_m(\omega) G^{p}_{n,TM}(\omega) G^{p+1*}_{m,TM}(\omega) \\ & + b_n(\omega) b^*_m(\omega) \eta^2(\omega) G^{p}_{n,TE}(\omega) G^{p+1*}_{m,TE}(\omega), \end{aligned} \tag{129}$$

$$\begin{aligned} V^{p}_{nm}(\omega) = & i b_n(\omega) a^*_m(\omega) \eta(\omega) G^{p}_{n,TE}(\omega) G^{p+1*}_{m,TM}(\omega) \\ & - i a_n(\omega) b^*_m(\omega) G^{p}_{n,TM}(\omega) \eta(\omega) G^{p+1*}_{m,TE}(\omega). \end{aligned} \tag{130}$$

For incident fields generated from discrete superpositions of monochromatic fields of arbitrary frequencies ω_j, substitution of Eqs. (60) and (61) into (125) and (126) allows us to perform the integral over ω and get

$$\begin{aligned} C_{pr,z} = & \frac{1}{\pi} \sum_{j=1}^{J} \sum_{n=1}^{\infty} \sum_{m=-n}^{+n} \frac{1}{(n+1)^2} \frac{(n+1+|m|)!}{(n-|m|)!} \frac{\lambda^2(\omega_j)}{2\eta(\omega_j)} \\ & \times \Bigg\{ \mathrm{Re} \Big[\big(a_n(\omega_j) + a^*_{n+1}(\omega_j) - 2 a_n(\omega_j) a^*_{n+1}(\omega_j) \big) \left|E_0(\omega_j)\right|^2 g^{m,mono}_{n,TM}(\omega_j) g^{m*,mono}_{n+1,TM}(\omega_j) \\ & + \big(b_n(\omega_j) + b^*_{n+1}(\omega_j) - 2 b_n(\omega_j) b^*_{n+1}(\omega_j) \big) \left|E_0(\omega_j)\right|^2 g^{m,mono}_{n,TE}(\omega_j) g^{m*,mono}_{n+1,TE}(\omega_j) \Big] \\ & + m \frac{2n+1}{n^2(n+1)^2} \frac{(n+|m|)!}{(n-|m|)!} \\ & \times \mathrm{Re} \Big[i (2 a_n(\omega_j) b^*_n(\omega_j) - a_n(\omega_j) - b^*_n(\omega_j)) |E_0(\omega_j)|^2 g^{m,mono}_{n,TM}(\omega_j) g^{m*,mono}_{n,TE}(\omega_j) \Big] \Bigg\} \end{aligned} \tag{131}$$

$$\left\{ \begin{array}{c} C_{pr,x} \\ C_{pr,y} \end{array} \right\} = \frac{1}{2\pi} \sum_{j=1}^{J} \sum_{p=1}^{\infty} \sum_{n=p}^{\infty} \sum_{m=p-1 \neq 0}^{\infty} \frac{(n+p)!}{(n-p)!} \frac{\lambda^2(\omega_j)}{2\eta(\omega_j)}$$
$$\times \Bigg\{ \left(\frac{1}{m^2} \delta_{m,n+1} - \frac{1}{n^2} \delta_{n,m+1} \right)$$
$$\left\{ \begin{array}{c} \mathrm{Re} \\ \mathrm{Im} \end{array} \right\} \left[S_{mn}^{p-1}(\omega_j) + S_{nm}^{-p}(\omega_j) - 2U_{mn}^{p-1}(\omega_j) - 2U_{nm}^{-p}(\omega_j) \right]$$
$$+ \frac{2n+1}{n^2(n+1)^2} \delta_{n,m}$$
$$\times \left\{ \begin{array}{c} \mathrm{Re} \\ \mathrm{Im} \end{array} \right\} \left[T_{mn}^{p-1}(\omega_j) - T_{nm}^{-p}(\omega_j) - 2V_{mn}^{p-1}(\omega_j) + 2V_{nm}^{-p}(\omega_j) \right] \Bigg\} \tag{132}$$

with the FFSs in Eqs. (127)–(130) being now replaced by $X_0 g_{n,TX}^{m,mono}(\omega_j)$. If $\omega_j = \omega_0$, $\forall j$, then the total field is a monochromatic field with operating frequency ω_0, therefore falling into the realm of the monochromatic GLMT. Of course, for a single monochromatic incident field, the j-sum can be discarded, and the radiation pressure cross-sections become the familiar cross-sections in the monochromatic GLMT. Torque expressions for polychromatic fields may be treated similarly following Refs. [40, 41].

4. Conclusions

In this work, a polychromatic version of the generalized Lorenz-Mie theory has been proposed in order to describe the interaction between an arbitrary-shaped, arbitrarily frequency-dependent optical field and a spherical, homogeneous particle, in terms of a partial wave expansion starting from Bromwich scalar potentials.

The monochromatic Bromwich scalar potentials have been expanded into spherical wave functions and their instantaneous, complex forms used to derive expressions for the incident electromagnetic fields. It was shown that the total incident field can be described in terms of field shape spectra, since frequency now becomes, in general, a continuous variable. When the total incident field becomes a discrete superposition of arbitrary-shaped, monochromatic light waves, the field shape spectra reduces to a train of impulses whose complex amplitude associated with each impulse is the beam shape coefficient of the original, monochromatic generalized Lorenz-Mie theory.

Expressions for several physical quantities of interest have been provided. They include incident, scattered and internal fields, scattered intensities and phase angles, extinction, scattering and absorption cross-sections, and radiation pressure cross-sections which are proportional to optical forces from non-thermal origin. Other quantities of potential interest, like optical torques and efficiency factors, could also be derived based on the formalism here developed, the latter requiring a more extended discussion (see Sec. 3.13 of Ref. [28]).

We expect that this work will serve its purpose in providing a framework for investigating polychromatic light scattering from spherical particles. It now incorporates arbitrary-shaped pulses and multichromatic light generated, for instance, from RGB laser sources, whose total field can be ideally described as a discrete superposition of three monochromatic fields of different frequencies. Also, natural extensions of it for multilayered spheres or spheres with inclusions, cylindrical and ellipsoidal particles can also be devised. EBCM, which is a T-matrix method (as other approaches such as GLMT beyond the "strict sense"), can also benefit from the formalism here presented. Finally, we expect that the polychromatic approach of this work will also eventually be incorporated into the analysis of thermal forces like photophoretic forces, now extended for polychromatic light scattering problems.

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