Towards photophoresis with the generalized Lorenz-Mie theory

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Abstract

Based on the adjoint boundary value problem proposed decades ago by Zulehner and Rohatschek [1], analytic and closed-form expressions for the photophoretic forces exerted by arbitrary-shaped beams on homogeneous and low-loss spherical particles is derived in both the free molecular and slip flow regimes. To do so, the asymmetry vector for arbitrary-index particles is explicitly calculated by expanding the internal electromagnetic fields with the aid of the generalized Lorenz-Mie theory (GLMT). The approach here proposed is, to the best of the authors' knowledge, the first systematic attempt to incorporate the GLMT *stricto sensu* into the field of photophoresis and might as well be extended, e.g. to spheroids and find important applications, among others, in optical trapping and manipulation of microparticles, in geoengineering, particle levitation, optical trap displays and so on.

Keywords: Generalized Lorenz-Mie theory, Photophoresis

1. Introduction

The determination of radiometric or photophoretic forces ($\mathbf{F}_{\mathrm{ph}}$) is not always an easy task. Because of that, the scientific community in the area of photophoresis suffers from the lack of an analytical theory capable of predicting such forces for light beams with arbitrary field profiles. In fact, the 'standard' solution procedure involves dealing simultaneously with the heat conduction equation and the Navier-Stokes equation with appropriate boundary conditions on the particle surface. Such conditions are based on physical grounds and dependent upon the Knudsen number $Kn = \ell/a$, where ℓ is the mean free path of molecules in the host fluid and a the radius of the illuminated particle.

After solving this set of equations for quantities such as temperature distribution in the particle and in the fluid, gas pressure and velocity fields, and photophoretic velocities, photophoretic forces are then evaluated from the stress tensor.

For plane waves and spherical particles with low losses, analytical solutions exist for what is known as the *asymmetry factor* J_1 (and, consequently, for $\mathbf{F}_{\rm ph}$, since they are proportional to each other), in the *slip-flow* and *free molecular* regimes. The formalism involves expansions of the electromagnetic fields internal to the particle using the Mie theory [2]. Qualitatively, however, it is known in advance that the resulting photophoretic forces will point either parallel (positive photophoresis) or anti-parallel (negative photophoresis) to the Poynting vector. For light beams with arbitrary spatial field profiles, we find most of the times attempts to approximate or use numerical methods [3, 4, 5, 6].

Boundary conditions depend upon the Knudsen number Kn. For $Kn >> 1$ (*free molecular* regime), the particle is much smaller than the mean free path ℓ of the gas and kinetic theory of gases applies. In 1967, using this theory, Hidy and Brock found an expression for the photophoretic force in this regime by assuming a solid, non-volatile and non-radiative homogeneous sphere [7]. Such an analysis was further improved by Tong in 1973, who introduced the additional effect of radiation from the surface of a black body caused by heating [8], and by subsequent works [2, 9, 10, 11, 12]. For $Kn < 1$ or $Kn << 1$ (*slip-flow* or *continuous* regime, respectively), the particle is larger or much larger than ℓ and the mechanical transport of the particle is given in terms of a continuous medium approach with appropriate slip-flow boundary conditions, the photophoretic force being then a direct consequence of thermal creep [13, 14]. In 1928, Hettner presented the first expressions for $\mathbf{F}_{\rm ph}$ in the continuous regime, assuming solid and non-volatile homogeneous spheres [15]. Also, a few decades after Rosen and Orr proposed an order of magnitude estimation for $\mathbf{F}_{\rm ph}$ [16] based on specific expressions for the temperature gradient at the surface of the particle previously deduced by Rubinowicz [17] and relying upon spheres illuminated only over a single hemisphere ($z < 0$). In a notorious work, Yalamov, Kutukov and Shchukin carried out a systematic study of the theory of photophoretic movement for volatile aerosols, considering the pressure on the particle surface caused by the asymmetric evaporation of the substance from the sphere [18]. Another interesting work was also published by Reed in almost the same period [19], who theoretically investigated photophoretic forces in the low Knudsen number regime for opaque particles, comparing his theoretical predictions with the most recent experimental results so far available [8, 20]. The dependence of the photophoretic force as a function of the size parameter was analyzed numerically by Arnold and Lewittes [21] and analytically by Mackowski [2] with the aid of the Mie theory for expressing the internal electric and magnetic fields. Studies involving photophoretic forces in the intermediate region $Kn < 1$ and how two extreme cases $Kn >> 1$ and $Kn << 1$ link to each other were initially carried out by Reed [19] and Mackowski [2]. In all previous works, as well as in the majority of publications to date, theoretical analysis has been restricted to uniform plane wave illumination (see, for instance, Refs. [14, 22, 23] for the period before 2013, to be complemented by Refs. [24, 25, 26, 27, 28] and references therein.). Very recently, photophoretic longitudinal and transverse asymmetry factors for dielectric and magnetodieletric cylinders and aggregates, including reflection from planar boudaries and corner spaces, have been investigated by Mitri, including incidence by waves and light-sheets with arbitrary polarization and incidence angle [29, 30, 31, 32].

The inclusion of arbitrary-shaped beams in photophoresis problems with spheres will certainly lead us to work within the formalism of the generalized Lorenz-Mie theory (GLMT) [33]. In the GLMT *strictu senso*, the incident, scattered and internal fields are expanded over a set of orthogonal spherical wave functions, the coefficients of such expansions - the beam shape coefficients (BSCs) - carrying all the information regarding the spatial field distribution of the incident wave. Because any solution to Maxwell's equations can be described within this context, we expect that any general theory on photophoresis for light-scattering by arbitrary-shaped beams and homogeneous spheres must inevitably incorporate GLMT into its mathematical foundations. In this path, Ambrosio has recently been able to extend the analysis beyond plane waves and dielectric particles, first by introducing arbitrary-index spheres in the case of plane wave illumination [34] and then by considering photophoretic forces exerted by on-axis axisymmetric beams [35], subsequently extended to higher-order Bessel beams by Wang *et al.* [36].

As stated by Fuchs [37] (also quoted in Ref. [14]), *"The main difficulty* *in calculating the radiometric force on a particle is the determination of the temperature gradient in the particle itself."*. Lamb's general solution, usually applied for plane wave illumination, might not be of much help beyond it [38, 39].

This paper deals with analytic and closed-form solutions to the photophoretic forces in both slip-flow and free molecular regimes with the aid of the GLMT. It incorporates into the theory of photophoresis, for the first time in the literature to the best of the author's knowledge, shaped beams beyond plane waves and arbitrarily located with respect to an opaque, non-radiative, non-volatile spherical scatterer. To do so, the method of the Adjoint Boundary Value Problem (ABVP) to the heat conduction equation proposed a few decades ago by Zulehner and Rohatschek [1] is here invoked in order to resolve for a vector generalization of J_1 called the *asymmetry vector* $\mathbf{r}_{\rm as}$, thus allowing us to solve for the photophoretic forces without the need for explicitly finding the temperature distribution within the particle itself. Expressions for both longitudinal and transverse components of $\mathbf{r}_{\rm as}$ exerted on arbitrary-index micro-spheres are then derived in terms of the BSCs, a feature which makes the present theory valid for any incident wave field in any optical regime (Rayleigh, Mie or geometric).

Section 2 presents a brief review on the method of calculation of $\mathbf{F}_{\rm ph}$ for spherical particles and plane waves, including the main aspects of the ABVP to be adopted in the subsequent sections. Section 3 concerns the derivation of $\mathbf{r}_{\rm as}$ for arbitrary beams with the aid of the GLMT, using the approach proposed by Zulehner and Rohatschek, for which $\mathbf{F}_{\rm ph} \propto \mathbf{r}_{\rm as}$. Here, both heat transfer from the particle and absorption of radiation within the fluid are neglected, and particles are restricted to non-volatile (solid) homogeneous spheres. Finally, conclusions are presented in Sec. 4.

2. Photophoresis for uniform plane wave illumination

2.1. The 'standard' procedure based on Lamb's general solution

Let us consider a homogeneous micro-particle of radius a and constant thermal conductivity k_s. The gas density, pressure and temperature distribution are represented by ρ_g, p_g and T_g, respectively.

The 'standard' procedure based on Lamb's general solution to the heat conduction equation [38, 39] says that in order to determine the photophoretic velocity and, consequently, the photophoretic force $\mathbf{F}_{\rm ph}$, the temperature distribution T_s within and on the surface of the sphere must be determined. For

$Kn < 1$, i.e. in the slip-flow regime, for example, the following set of equations must be solved

$$\nabla^2 T_s = -\frac{Q\left(r,\theta,\varphi\right)}{k_s}, \tag{1a}$$

$$\nabla^2 T_g = \frac{\rho_g c_p}{k_g}\left(\frac{v_\theta}{r}\frac{\partial T_g}{\partial \theta} + v_r \frac{\partial T_g}{\partial r}\right), \tag{1b}$$

$$\nabla^2 \mathbf{v} = \frac{1}{\eta_g}\nabla p_g, \tag{1c}$$

$$\nabla \cdot \mathbf{v} = 0. \tag{1d}$$

Equations (1a) and (1b) are the heat conduction equations for T_s and T_g, respectively. The function $Q(r,\theta,\varphi)$ is known as the heat source function (HSF) and depends on the internal field intensity distribution. Navier-Stokes equations are given by (1c) and (1d), where $\mathbf{v} = v_r\hat{r} + v_\theta\hat{\theta} + v_\varphi\hat{\varphi}$ is the fluid velocity vector according to a spherical coordinate system (r,θ,φ) whose origin coincides with the center of the sphere.

The differential equations in Eq. (1) must satisfy the following boundary conditions:

$$T_g - T_s = c_t \ell \frac{\partial T_g}{\partial r}, \qquad r = a, \tag{2a}$$

$$k_g \frac{\partial T_g}{\partial r} = k_s \frac{\partial T_s}{\partial r}, \qquad r = a, \tag{2b}$$

$$T_g = T_0, \qquad r \to \infty, \tag{2c}$$

$$v_r = 0, \qquad r = a, \tag{2d}$$

$$\begin{aligned} v_\theta &= c_m \ell \left[r \frac{\partial}{\partial r}\left(\frac{v_\theta}{r}\right) + \frac{1}{r}\frac{\partial v_r}{\partial \theta}\right] + \frac{c_s \eta_g}{\rho_g T_0 a}\frac{\partial T_g}{\partial \theta} \\ &= c_m \ell \sigma_{\theta r} + \frac{c_s \eta_g}{\rho_g T_0 a}\frac{\partial T_g}{\partial \theta}, \qquad r = a, \end{aligned} \tag{2e}$$

$$v_\varphi = c_m \ell \left[r\frac{\partial}{\partial r}\left(\frac{v_\varphi}{r}\right) + \frac{1}{r\sin\theta}\frac{\partial v_r}{\partial \varphi}\right] + \frac{c_s\eta_g}{\rho_g T_0 a}\frac{\partial T_g}{\partial \varphi}$$
$$= c_m\ell\sigma_{\varphi r} + \frac{c_s\eta_g}{\rho_g T_0 a}\frac{\partial T_g}{\partial \varphi}, \qquad r = a, \tag{2f}$$

$$\mathbf{v} = \mathbf{V}_0, \qquad r \to \infty. \tag{2g}$$

In (1) and (2), η_g is the viscosity and k_g the thermal conductivity of the gas, c_t, c_m and c_s are constants calculated from the kinetic theory of gases with values 2.18, 1.14 and 1.17, respectively [40], and c_p is the specific heat at constant pressure. The elements of the stress tensor $\overline{\overline{\sigma}}$ are designated by σ_{ij}.

For details involving the solution of the set of equations in the free molecular regime, including its appropriate boundary conditions, see e.g. Refs. [2, 18]. Analytic solutions of (1) and (2) have been found when (i) the HSF has azimuthal symmetry [that is, $Q(r,\theta,\varphi) \equiv Q(r,\theta)$], which happens to be the case for unpolarized plane wave illumination), and (ii) when convection terms of the r.h.s. of (1b) are neglected, which means that T_g obeys a Laplace equation. For axisymmetric flow, it is easy to infer that $v_\varphi = 0$ and one can shown that for $+z$-propagating light, $\mathbf{F}_{\text{ph}} = F_z\hat{z}$.

The standard method for solving the set of equations (1) and (2) relies upon expansions of T_g, $\mathbf{v}$ and p_g in terms of spherical wave functions. For the axisymmetric plane wave case and using spherical coordinates, the general solutions to the thermodynamics [(1a) and (1b)] and hydrodynamics [(1c) and (1d)] equations can be obtained with the aid of Lamb's general solutions [38, 39] under the following form [2]:

$$\frac{T_s - T_0}{T_0} = \sum_{n=0}^{\infty}\left[A_n\zeta^n + G_n(\zeta)\right]P_n(\mu), \tag{3a}$$

$$\frac{T_g - T_0}{T_0} = \sum_{n=0}^{\infty} D_n\zeta^{-(1+n)}P_n(\mu), \tag{3b}$$

$$v_r = \sum_{n=1}^{\infty} f_{rn}(\zeta)P_n(\mu), \tag{3c}$$

$$v_\theta = \sum_{n=1}^{\infty} f_{\theta n}(\zeta)\, P_n^1(\mu)\,, \tag{3d}$$

$$p_g = \sum_{n=1}^{\infty} f_{pn}(\zeta)\, P_n(\mu)\,, \tag{3e}$$

where $P_n^m(x)$ are the associated Legendre functions $[P_n^0(x) = P_n(x)]$ according to Robin's notation [41] adopted in the GLMT convention. The constants A_n and D_n, as well as the r-dependent functions $\zeta = r/a$, f_{rn}, $f_{\theta n}$ and f_{pn}, are calculated after the imposition of the boundary conditions (2), see [2]. The function $G_n(\zeta)$ depends on the HSF $Q(r,\theta)$ according to

$$\begin{aligned} G_n(\zeta) =& \frac{1}{2}\Bigg[\zeta^n \int_\zeta^1 t^{1-n} \int_{-1}^{1} g(t,\theta)\, P_n(\cos\theta)\, d(\cos\theta)\, dt \\ &+ \zeta^{-(1+n)} \int_0^\zeta t^{n+2} \int_{-1}^{1} g(t,\theta)\, P_n(\cos\theta)\, d(\cos\theta)\, dt\Bigg], \end{aligned} \tag{4}$$

with $g(r,\theta) = a^2 Q(r,\theta)/k_s T_0$. After some algebra, one finds an expression for $\mathbf{F}_{\rm ph}$ [2, 9, 19]:

$$\mathbf{F}_{\rm ph} = -\frac{4\pi c_s \eta_g^2 I_\lambda a J_1}{\rho_g k_s T_0} \frac{1}{(1+3c_m\ell/a)(1+2c_t\ell/a+2k_g/k_s)}\hat{z}, \tag{5}$$

where J_1 is the *asymmetry factor*

$$J_1(x,M) = 3 n_{sp} m_{sp} x \int_0^1 \int_{-1}^{1} B\left(t = \frac{r}{a}, \mu\right) t^3 \mu\, d\mu\, dt. \tag{6}$$

In (5), $I_\lambda = |E_0|^2/2\eta_0$ is the intensity of the incident wave, E_0 its electric field strength and η_0 the intrinsic impedance of the gas. The size parameter of the particle is defined as $x = (2\pi/\lambda)a = ka$ and $M = n_{sp} - i m_{sp}$ is its complex refractive index with $\mu_{sp} = \mu' = \mu_0$ and $\epsilon_{sp} = \epsilon' - i\epsilon''$ its permeability (μ_0 is the permeability of free space) and permittivity, respectively. Parameters relative to the external medium carry a subscript 'r' (e.g., a relative permittivity $\epsilon_{sp,r} = \epsilon'_r - i\epsilon''_r$). Finally, $B(r,\theta) = |\mathbf{E}_{\rm int}(r,\theta)|^2/|E_0|^2$ is the dimensionless radiative intensity distribution function [2], source strength [42] or normalized source functions [3], with $\mathbf{E}_{\text{int}}$ the electric field *inside* the sphere.

The double integral in (6) can be evaluated explicitly for the case of a plane wave illumination using the Mie theory for scattering by dielectric particles. By expanding the internal fields into a sum of spherical wave functions, Mackowski [2] found an expression for J_1 which, in terms of the standard time harmonic factor $\exp(+i\omega t)$ used in the GLMT [33], can be written as:

$$J_1\left(x,M\right) = \frac{6n_{sp}m_{sp}}{\left|M\right|^2 x^3}\,\mathrm{Im}\sum_{n=1}^{\infty}\left\{\frac{n\left(n+2\right)}{M}\left(c_{n+1}c_n^*R_{n+1}+d_{n+1}d_n^*R_n\right)\right.$$
$$\left. - \left[\frac{n\left(n+2\right)}{n+1}\left(c_{n+1}^*c_n+d_{n+1}d_n^*\right)+\frac{2n+1}{n\left(n+1\right)}c_nd_n^*\right]S_n\right\}, \tag{7}$$

where the Mie coefficients c_n and d_n for internal fields and dielectric particles are [33]:

$$c_n = \frac{M\left[\xi_n\left(x\right)\psi_n'\left(x\right)-\xi_n'\left(x\right)\psi\left(x\right)\right]}{\xi_n\left(x\right)\psi_n'\left(Mx\right)-M\xi_n'\left(x\right)\psi_n\left(Mx\right)}, \tag{8a}$$

$$d_n = \frac{M^2\left[\xi_n\left(x\right)\psi_n'\left(x\right)-\xi_n'\left(x\right)\psi\left(x\right)\right]}{M\xi_n\left(x\right)\psi_n'\left(Mx\right)-\xi_n'\left(x\right)\psi_n\left(Mx\right)}. \tag{8b}$$

Also, R_n and S_n are functions of x and M according to the following relations (correcting for a typo in Eq. (61) of Ref. [2]),

$$R_n \equiv \int_0^x \left|\psi\left(M\rho\right)\right|^2 d\rho = \frac{\mathrm{Im}\left[M\psi_{n+1}\left(Mx\right)\psi_n^*\left(Mx\right)\right]}{\mathrm{Im}\left(M^2\right)}, \tag{9}$$

$$
\begin{aligned}
S_n &\equiv \int\limits_0^x \rho \psi_n^* (M\rho)\, \psi_n' (M\rho)\, d\rho \\
&= -\frac{i}{2\,\mathrm{Im}\,(M^2)} \Bigg\{ x \left(M |\psi_n (Mx)|^2 + M^* |\psi_{n+1} (Mx)|^2 \right) \\
&\quad - \left(M + 2\,(n+1)\, \frac{\mathrm{Re}\,(M^2)}{M} \right) R_n + (2n+1)\, M^* R_{n+1} \Bigg\}.
\end{aligned} \tag{10}
$$

In (8)-(10), $\psi_n(x)$ and $\xi_n(x)$ are Ricatti-Bessel functions, with a prime indicating a differentiation with respect to the argument [33]. Generalizations of (7) have been recently developed by Ambrosio for arbitrary refractive index spheres under plane wave illumination [34] and for on-axis axisymmetric beams (Gaussian and zero-order circularly symmetric Bessel beams) [35], with an extension to higher-order circularly symmetric Bessel beams by Wang *et al.* [36]. To the best of the author's knowledge, despite experimental advances, the only other work that attempts to analytically calculate $\mathbf{F}_{\mathrm{ph}}$ for arbitrary-shaped beams is the one presented by Desyatnikov *et al.* in 2009 [6] for low-loss aerosol particles manipulated via photophoretic forces using vortex beams (Laguerre-Gauss LG_{01} beam). In their approach, approximations are proposed based on the size of the particle with respect to the diffraction length l and assuming that the sphere is always placed along the optical axis (z axis). Theoretical results are shown to be in good agreement with experiments.

2.2. The ABVP method and the asymmetry vector

In 1994, Zulehner and Rohatschek [1] presented a method for calculating $\mathbf{F}_{\mathrm{ph}}$ for non-spherical particles based on an equivalent problem to the heat conduction equation. The analysis was separated according to the slip-flow or free molecular regime, which means that for each regime certain boundary conditions must be met.

In this method, $\mathbf{F}_{\mathrm{ph}}$ is expressed directly in terms of the HSF after applying Green's second identity to obtain an adjoint boundary value problem starting from the non-homogeneous heat conduction equation (1a). In the process, a weight function $\mathbf{w}(\mathbf{r})$ is introduced ($\mathbf{r} = x\hat{x}+y\hat{y}+z\hat{z}$ is the position vector) whose form depends on the geometry of the particle.

For the free molecular regime, a general linear boundary condition is assumed for (1b):

$$-k_s \frac{\partial T_s}{\partial n} = A + BT_s, \tag{11}$$

where $\partial/\partial n$ denotes the normal derivative with respect to the surface of the particle, $A = -hT_0$ and $B = h$, with h being the molecular heat transfer coefficient and given by $h = \alpha p_g \overline{v}/2T_0$ for monatomic and $h = 3\alpha p_g \overline{v}/4T_0$ for diatomic gases, where α is the thermal accommodation coefficient and $\overline{v}$ is the mean speed of gas molecules. In view of that, for $Kn >> 1$ [1],

$$\mathbf{F}_{\text{ph}} = -C \int_{V_p} Q(\mathbf{r})\, \mathbf{w}(\mathbf{r})\, dV, \tag{12}$$

where $C = \alpha p_g/4T_0$ and V_p is the volume of the arbitrary-shaped particle. In the case of a spherical particle, $\mathbf{w}(\mathbf{r}) = \mathbf{r}/(Ba + k_s)$ and (12) reduces to

$$\mathbf{F}_{\text{ph}} = -\frac{C}{Ba + k_s} \int_{V_p} \mathbf{r} Q(\mathbf{r})\, dV. \tag{13}$$

Similarly, in the slip-flow regime with boundary condition given by (2b), one has [1]:

$$\mathbf{F}_{\text{ph}} = -\frac{3c_s \eta_g^2}{\rho_g T_0 a^2 (k_g + k_s)} \int_{V_p} \mathbf{r} Q(\mathbf{r})\, dV. \tag{14}$$

It is seen from (13) and (14) that, instead of a scalar asymmetry factor J_1, one can now speak in terms of an *asymmetry vector*, $\mathbf{r}_{\text{as}}$ [1, 5] which, for our purposes and differing slightly from previous works, is here defined as:

$$\mathbf{r}_{\text{as}} = \int_{V_p} \mathbf{r} Q(\mathbf{r})\, dV. \tag{15}$$

3. Photophoretic forces for arbitrary-shaped beams in the GLMT

Equations (13) and (14) can be written in a more compact form:

$$\mathbf{F}_{\text{ph}} = -C_{Kn} \mathbf{r}_{\text{as}}, \tag{16}$$

where $C_{Kn} = C/(Ba + k_s)$ for $Kn >> 1$ and $3c_s\eta_g^2/[\rho_g T_0 a^2(k_g + k_s)]$ for $Kn < 1$. As is clear from (16), knowledge of $\mathbf{r}_{\rm as}$ for a given HSF or, in other words, for a given electromagnetic field distribution inside the sphere completely determines (except for a constant factor) the photophoretic force in both the slip-flow and free molecular regimes. Equation (16) shall be explicitly solved first for dielectric (or non-magnetic) particles having only electric losses. Then, we extend the calculations to incorporate scatterers having an arbitrary index of refraction, which encompasses magnetic, magnetodielectric, negative index scatterers and so on, for which both electric and magnetic losses can be present.

3.1. Dielectric/non-magnetic particles

For dielectric or non-magnetic particles in general, the HSF $Q(r, \theta, \varphi)$ can be written in terms of the electric field intensity as [34]:

$$Q\left(r,\theta,\varphi\right) = \frac{1}{2}\sigma|\mathbf{E}\left(r,\theta,\varphi\right)|^2 = k\epsilon_r'' I_\lambda B\left(r,\theta,\varphi\right). \tag{17}$$

In (17), $\sigma = \omega\epsilon_m\epsilon_r''$ is the electric conductivity of the sphere (ϵ_m is the permittivity of the host fluid), $I_\lambda = |\mathbf{E}_0|^2/2\eta_0$ is the intensity of the wave, η_m being the intrinsic impedance of the fluid. In addition, $B\left(r,\theta,\varphi\right) = |\mathbf{E}_{\rm int}\left(r,\theta,\varphi\right)|^2/|\mathbf{E}_0|^2$ is the dimensionless radiative intensity distribution function [2] (also called source strength [42] or normalized source function [3]).

The determination of $\mathbf{F}_{\rm ph}$ starts with the replacement of (17) into (15) and substituting $|\mathbf{E}_{\rm int}|^2/E_0$ considering the electric field components provided by the GLMT formalism [33] (see also [43, 44] and references therein), which may be rewritten as:

$$\frac{E_r}{E_0} = \sum_{n=1}^{\infty}\sum_{p=-n}^{n}\left(-i\right)^{n+1}\left(2n+1\right)c_n g_{n,\rm TM}^p \frac{\psi_n\left(k_{sp}r\right)}{k_{sp}^2 r^2} P_n^{|p|}\left(\cos\theta\right)e^{ip\varphi}, \tag{18a}$$

$$\begin{aligned}\frac{E_\theta}{E_0} =& \frac{1}{k_{sp}r}\sum_{n=1}^{\infty}\sum_{p=-n}^{n}\left(-i\right)^{n+1}E_n\Bigg\{c_n g_{n,\rm TM}^p \psi_n'\left(k_{sp}r\right)\tau_n^{|p|}\left(\cos\theta\right) \\ &+ p\left(\frac{\mu_{sp}k}{\mu k_{sp}}\right)d_n g_{n,\rm TE}^p \psi_n\left(k_{sp}r\right)\pi_n^{|p|}\left(\cos\theta\right)\Bigg\}e^{ip\varphi},\end{aligned} \tag{18b}$$

$$\begin{aligned}\frac{E_\varphi}{E_0} =& \frac{i}{k_{sp} r} \sum_{n=1}^{\infty} \sum_{p=-n}^{n} (-i)^{n+1} E_n \Bigg\{ p c_n g_{n,\mathrm{TM}}^{p} \psi_n' \left(k_{sp} r\right) \pi_n^{|p|} \left(\cos\theta\right) \\ &+ \left(\frac{\mu_{sp} k}{\mu k_{sp}}\right) d_n g_{n,\mathrm{TE}}^{p} \psi_n \left(k_{sp} r\right) \tau_n^{|p|} \left(\cos\theta\right) \Bigg\} e^{ip\varphi}.\end{aligned} \tag{18c}$$

In (18), $k_{sp} = Mk$, $E_n = (2n+1)/[n(n+1)]$, $\tau(\cos\theta) = dP_n^m(\cos\theta)/d\theta$ and $\pi_n^m(\cos\theta) = P_n^m(\cos\theta)/\sin\theta$ are generalized Legendre functions. The coefficients $g_{n,\mathrm{TM}}^m$ and $g_{n,\mathrm{TE}}^m$ are the *beam shape coefficients* (BSCs) for TM and TE modes, respectively. The BSCs contain all the information regarding the spatial field distribution of the incident beam relative to the plane wave.

Computing $|\mathbf{E}_{\mathrm{int}}|^2$ from (18) and replacing the resulting HSF from (17) in (16), one finds the following expression for $\mathbf{r}_{\mathrm{as}}$:

$$\mathbf{r}_{\mathrm{as}} = \frac{\epsilon_r''}{2} I_\lambda \left[\mathcal{I}_x \hat{x} + \mathcal{I}_y \hat{y} + \mathcal{I}_z \hat{z}\right], \tag{19}$$

where

$$\mathcal{I}_x = 2\frac{x}{a} \int_0^{2\pi} \int_0^{\pi} \int_0^{a} B\left(r,\theta,\varphi\right) r^3 \sin^2\theta \cos\varphi \, dr d\theta d\varphi, \tag{20a}$$

$$\mathcal{I}_y = 2\frac{x}{a} \int_0^{2\pi} \int_0^{\pi} \int_0^{a} B\left(r,\theta,\varphi\right) r^3 \sin^2\theta \sin\varphi \, dr d\theta d\varphi, \tag{20b}$$

$$\mathcal{I}_z = 2\frac{x}{a} \int_0^{2\pi} \int_0^{\pi} \int_0^{a} B\left(r,\theta,\varphi\right) r^3 \cos\theta \sin\theta \, dr d\theta d\varphi. \tag{20c}$$

The integrals with respect to the azimuth angle φ in (20) can be easily evaluated. It can be shown from (18) that they are of the form

$$\int_0^{2\pi} e^{i(p-q)} \cos\varphi \, d\varphi = \pi\left(\delta_{p,q+1} + \delta_{q,p+1}\right), \tag{21a}$$

$$\int_0^{2\pi} e^{i(p-q)} \sin\varphi \, d\varphi = i\pi\left(\delta_{p,q+1} - \delta_{q,p+1}\right), \tag{21b}$$

$$\int_{0}^{2\pi} e^{i(p-q)} d\varphi = 2\pi \delta_{p,q}, \tag{21c}$$

where $\delta_{i,j}$ is the Kronecker delta. Imposing (21) on (20), we get the double integrals:

$$\mathcal{I}_x = 2\frac{x}{a} \int_{0}^{\pi} \int_{0}^{a} B_x\left(r,\theta\right) r^3 \sin^2\theta dr d\theta, \tag{22a}$$

$$\mathcal{I}_y = 2\frac{x}{a} \int_{0}^{\pi} \int_{0}^{a} B_y\left(r,\theta\right) r^3 \sin^2\theta dr d\theta, \tag{22b}$$

$$\mathcal{I}_z = 2\frac{x}{a} \int_{0}^{\pi} \int_{0}^{a} B_z\left(r,\theta\right) r^3 \cos\theta \sin\theta dr d\theta d\varphi, \tag{22c}$$

where

$$B_{\left\{ \begin{array}{c} x \\ y \end{array} \right\}} = \sum_{j=1}^{5} B^j_{\left\{ \begin{array}{c} x \\ y \end{array} \right\}}, \tag{23}$$

with

$$\begin{aligned} B^1_{\left\{ \begin{array}{c} x \\ y \end{array} \right\}} =& \frac{i\pi}{\left|k_{sp}\right|^4 r^4} \sum_{n=1}^{\infty} \sum_{l=1}^{\infty} \left(-1\right)^{n+1} i^{n+l+2} \left(2n+1\right)\left(2l+1\right) c_n c_l^* \psi_n \psi_l^* \\ & \times \left[\sum_{q=-l}^{l} g_{n,\mathrm{TM}}^{q+1} g_{l,\mathrm{TM}}^{q*} P_n^{|q+1|} P_l^{|q|} \pm \sum_{p=-n}^{n} g_{n,\mathrm{TM}}^{p} g_{l,\mathrm{TM}}^{p+1*} P_n^{|p|} P_l^{|p+1|} \right], \end{aligned} \tag{24a}$$

$$
\begin{aligned}
B^2_{\left\{\begin{matrix} x \\ y \end{matrix}\right\}} =& \frac{i\pi}{\left|k_{sp}\right|^2 r^2} \sum_{n=1}^{\infty}\sum_{l=1}^{\infty} \left(-1\right)^{n+1} i^{n+l+2} E_n E_l c_n c_l^* \psi_n' \psi_l'^* \\
& \times \left[\sum_{q=-l}^{l} g_{n,\mathrm{TM}}^{q+1} g_{l,\mathrm{TM}}^{q*} \left(\tau_n^{|q+1|} \tau_l^{|q|} + q\left(q+1\right) \pi_n^{|q+1|} \pi_l^{|q|} \right) \right. \\
& \left. \pm \sum_{p=-n}^{n} g_{n,\mathrm{TM}}^{p} g_{l,\mathrm{TM}}^{p+1*} \left(\tau_n^{|p|} \tau_l^{|p+1|} + p\left(p+1\right) \pi_n^{|p|} \pi_l^{|p+1|} \right) \right],
\end{aligned} \tag{24b}
$$

$$
\begin{aligned}
B^3_{\left\{\begin{matrix} x \\ y \end{matrix}\right\}} =& \frac{i\pi}{\left|k_{sp}\right|^2 r^2} \left|\frac{\mu_{sp} k}{\mu k_{sp}}\right|^2 \sum_{n=1}^{\infty}\sum_{l=1}^{\infty} \left(-1\right)^{n+1} i^{n+l+2} E_n E_l d_n d_l^* \psi_n \psi_l^* \\
& \times \left[\sum_{q=-l}^{l} g_{n,\mathrm{TE}}^{q+1} g_{l,\mathrm{TE}}^{q*} \left(\tau_n^{|q+1|} \tau_l^{|q|} + q\left(q+1\right) \pi_n^{|q+1|} \pi_l^{|q|} \right) \right. \\
& \left. \pm \sum_{p=-n}^{n} g_{n,\mathrm{TE}}^{p} g_{l,\mathrm{TE}}^{p+1*} \left(\tau_n^{|p|} \tau_l^{|p+1|} + p\left(p+1\right) \pi_n^{|p|} \pi_l^{|p+1|} \right) \right],
\end{aligned} \tag{24c}
$$

$$
\begin{aligned}
B^4_{\left\{\begin{matrix} x \\ y \end{matrix}\right\}} =& \frac{i\pi}{\left|k_{sp}\right|^2 r^2} \left(\frac{\mu_{sp} k}{\mu k_{sp}}\right)^* \sum_{n=1}^{\infty}\sum_{l=1}^{\infty} \left(-1\right)^{n+1} i^{n+l+2} E_n E_l c_n d_l^* \psi_n' \psi_l^* \\
& \times \left[\sum_{q=-l}^{l} g_{n,\mathrm{TM}}^{q+1} g_{l,\mathrm{TE}}^{q*} \left(q\tau_n^{|q+1|} \pi_l^{|q|} + \left(q+1\right) \pi_n^{|q+1|} \tau_l^{|q|} \right) \right. \\
& \left. \pm \sum_{p=-n}^{n} g_{n,\mathrm{TM}}^{p} g_{l,\mathrm{TE}}^{p+1*} \left(\left(p+1\right) \tau_n^{|p|} \pi_l^{|p+1|} + p\pi_n^{|p|} \tau_l^{|p+1|} \right) \right],
\end{aligned} \tag{24d}
$$

$$B^5_{\left\{\begin{array}{c} x \\ y \end{array}\right\}} = \frac{i\pi}{|k_{sp}|^2 r^2} \left(\frac{\mu_{sp} k}{\mu k_{sp}}\right) \sum_{n=1}^{\infty} \sum_{l=1}^{\infty} (-1)^{n+1} i^{n+l+2} E_n E_l d_n c_l^* \psi_n \psi_l^{\prime *}$$
$$\times \left[\sum_{q=-l}^{l} g^{q+1}_{n,\mathrm{TE}} g^{q*}_{l,\mathrm{TM}} \left(q \tau_n^{|q+1|} \pi_l^{|q|} + (q+1) \pi_n^{|q+1|} \tau_l^{|q|} \right) \right. \tag{24e}$$
$$\left. \pm \sum_{p=-n}^{n} g^{p}_{n,\mathrm{TE}} g^{p+1*}_{l,\mathrm{TM}} \left((p+1) \tau_n^{|p|} \pi_l^{|p+1|} + p \pi_n^{|p|} \tau_l^{|p+1|} \right) \right],$$

and

$$B_z = \sum_{j=1}^{5} B_z^j, \tag{25}$$

with

$$B_z^1 = \frac{2\pi}{|k_{sp}|^4 r^4} \sum_{n=1}^{\infty} \sum_{l=1}^{\infty} \sum_{p=-n}^{n} (-1)^{n+1} i^{n+l+2} (2n+1)$$
$$\times (2l+1) c_n c_l^* g^{p}_{n,\mathrm{TM}} g^{p*}_{l,\mathrm{TM}} \psi_n \psi_l^* P_n^{|p|} P_l^{|p|}, \tag{26a}$$

$$B_z^2 = \frac{2\pi}{|k_{sp}|^2 r^2} \sum_{n=1}^{\infty} \sum_{l=1}^{\infty} \sum_{p=-n}^{n} (-1)^{n+1} i^{n+l+2} E_n E_l c_n c_l^*$$
$$g^{p}_{n,\mathrm{TM}} g^{p*}_{l,\mathrm{TM}} \psi_n' \psi_l^{\prime *} \left(\tau_n^{|p|} \tau_l^{|p|} + p^2 \pi_n^{|p|} \pi_l^{|p|} \right), \tag{26b}$$

$$B_z^3 = \frac{2\pi}{|k_{sp}|^2 r^2} \left|\frac{\mu_{sp} k}{\mu k_{sp}}\right|^2 \sum_{n=1}^{\infty} \sum_{l=1}^{\infty} \sum_{p=-n}^{n} (-1)^{n+1} i^{n+l+2} E_n E_l$$
$$d_n d_l^* g^{p}_{n,\mathrm{TE}} g^{p*}_{l,\mathrm{TE}} \psi_n \psi_l^* \left(\tau_n^{|p|} \tau_l^{|p|} + p^2 \pi_n^{|p|} \pi_l^{|p|} \right), \tag{26c}$$

$$B_z^4 = \frac{2\pi}{|k_{sp}|^2 r^2} \left(\frac{\mu_{sp} k}{\mu k_{sp}}\right)^* \sum_{n=1}^{\infty} \sum_{l=1}^{\infty} \sum_{p=-n}^{n} p (-1)^{n+1} i^{n+l+2}$$
$$E_n E_l c_n d_l^* g^{p}_{n,\mathrm{TM}} g^{p*}_{l,\mathrm{TE}} \psi_n' \psi_l^* \left(\tau_n^{|p|} \pi_l^{|p|} + \pi_n^{|p|} \tau_l^{|p|} \right), \tag{26d}$$

$$B_z^5 = \frac{2\pi}{|k_{sp}|^2 r^2}\left(\frac{\mu_{sp}k}{\mu k_{sp}}\right)\sum_{n=1}^{\infty}\sum_{l=1}^{\infty}\sum_{p=-n}^{n} p\,(-1)^{n+1}\, i^{n+l+2}$$
$$E_n E_l d_n c_l^* g_{n,\mathrm{TE}}^{p} g_{l,\mathrm{TM}}^{p*} \psi_n \psi_l'^* \left(\tau_n^{|p|}\pi_l^{|p|} + \pi_n^{|p|}\tau_l^{|p|}\right). \tag{26e}$$

In Eqs. (24) and (26), $\Psi_n(k_{sp}r) \equiv \Psi_n$, the same being valid for the functions $P_n^m(\cos\theta)$, $\tau_n^m(\cos\theta)$ and $\pi_n^m(\cos\theta)$, where we omitted the arguments. The θ-integrals in (22a) and (22b) related to B_x and B_y are of the form:

$$\mathcal{I}_{\theta,1} = \int_0^\pi P_n^{|q+1|} P_l^{|q|} \sin^2\theta d\theta, \tag{27a}$$

$$\mathcal{I}_{\theta,2} = \int_0^\pi P_n^{|p|} P_l^{|p+1|} \sin^2\theta d\theta, \tag{27b}$$

$$\mathcal{I}_{\theta,3} = \int_0^\pi \left[\tau_n^{|q+1|}\tau_l^{|q|} + q\,(q+1)\,\pi_n^{|q+1|}\pi_l^{|q|}\right] \sin^2\theta d\theta, \tag{27c}$$

$$\mathcal{I}_{\theta,4} = \int_0^\pi \left[\tau_n^{|p|}\tau_l^{|p+1|} + p\,(p+1)\,\pi_n^{|p|}\pi_l^{|p+1|}\right] \sin^2\theta d\theta, \tag{27d}$$

$$\mathcal{I}_{\theta,5} = \int_0^\pi \left[q\tau_n^{|q+1|}\pi_l^{|q|} + (q+1)\,\pi_n^{|q+1|}\tau_l^{|q|}\right] \sin^2\theta d\theta, \tag{27e}$$

$$\mathcal{I}_{\theta,6} = \int_0^\pi \left[(p+1)\,\tau_n^{|p|}\pi_l^{|p+1|} + p\pi_n^{|p|}\tau_l^{|p+1|}\right] \sin^2\theta d\theta, \tag{27f}$$

and, for B_z in (22c),

$$\mathcal{I}_{\theta,7} = \int_0^\pi P_n^{|p|} P_l^{|p|} \cos\theta \sin\theta d\theta, \tag{28a}$$

$$\mathcal{I}_{\theta,8} = \int_0^\pi \left[\tau_n^{|p|}\tau_l^{|p|} + p^2\pi_n^{|p|}\pi_l^{|p|}\right] \cos\theta \sin\theta d\theta, \tag{28b}$$

$$\mathcal{I}_{\theta,9} = \int_0^\pi \left[\tau_n^{|p|}\pi_l^{|p|} + \pi_n^{|p|}\tau_l^{|p|}\right] \cos\theta \sin\theta d\theta. \tag{28c}$$

Some of the integrals in (27) and (28) can be found in Ref. [45], and others in the Appendix section of Ref. [33], the remaining ones being calculated from combinations of some of the integrals presented in the first aforementioned reference using several recurrence relations for the associated Legendre polynomials and their derivatives. For convenience, we list them in the Appendix with the appropriate notation. In using Ref. [45] we have introduced a multiplicative factor $(-1)^m$ to ensure the usual Robin's definition of the associated Legendre polynomials adopted the GLMT [33].

Substituting (25) in (22c) and making use of (26) with the corresponding integrals (28) whose solutions are given in (A.7)-(A.9), changing dummy variables and after some pages of calculations, one gets an expression for $\mathcal{I}_z$ in terms solely of integrals over r:

$$\begin{aligned}
\mathcal{I}_z =& \frac{16\pi x}{a|k_{sp}|^2} \sum_{n=1}^{\infty} \sum_{m=-n}^{n} \operatorname{Im} \Bigg\{ c_n^* c_{n+1} g_{n,\mathrm{TM}}^{m*} g_{n+1,\mathrm{TM}}^{m} \\
& \left[\frac{1}{|k_{sp}|^2} \frac{(n+1+|m|)!}{(n-|m|)!} \int_0^x \frac{\psi_n^*(M\rho)\,\psi_{n+1}(M\rho)}{\rho} d\rho \right. \\
& \left. + \frac{1}{k^2} \frac{1}{(n+1)^2} \frac{(n+1+|m|)!}{(n-|m|)!} \int_0^x \psi_n'^*(M\rho)\,\psi_{n+1}'(M\rho)\,\rho d\rho \right] \\
& + \left|\frac{\mu_{sp}k}{\mu k_{sp}}\right|^2 \frac{1}{k^2} \frac{1}{(n+1)^2} \frac{(n+1+|m|)!}{(n-|m|)!} d_n^* d_{n+1} g_{n,\mathrm{TE}}^{m*} g_{n+1,\mathrm{TE}}^{m} \\
& \times \int_0^x \psi_n^*(M\rho)\,\psi_{n+1}(M\rho)\,\rho d\rho \\
& + im \left(\frac{\mu_{sp}k}{\mu k_{sp}}\right)^* \frac{1}{k^2} \frac{2n+1}{n^2(n+1)^2} \frac{(n+|m|)!}{(n-|m|)!} c_n d_n^* g_{n,\mathrm{TM}}^{m} g_{n,\mathrm{TE}}^{m*} \\
& \times \int_0^x \psi_n^*(M\rho)\,\psi_n'(M\rho)\,\rho d\rho \Bigg\},
\end{aligned} \tag{29}$$

where $\rho = kr$. For $\mathcal{I}_x$ and $\mathcal{I}_y$, the expressions are more complicated. Setting $\psi_n(M\rho) \equiv \psi_n$ with $\rho = kr$, they can be put into the form

$$\mathcal{I}_{\left\{\begin{array}{c} x \\ y \end{array}\right\}} = 2\frac{x}{a}\sum_{j=1}^{3} \mathcal{g}^{j}_{\left\{\begin{array}{c} x \\ y \end{array}\right\}}, \tag{30}$$

with

$$\begin{aligned}
\mathcal{g}^{1}_{\left\{\begin{array}{c} x \\ y \end{array}\right\}} =& \frac{4\pi}{|k_{sp}|^2}\left\{\begin{array}{c} \mathrm{Im} \\ \mathrm{Re} \end{array}\right\}\sum_{n=1}^{\infty}\Bigg\{ \pm \sum_{m=0}^{n+1} c_n^* c_{n+1} g_{n,\mathrm{TM}}^{m+1*} g_{n+1,\mathrm{TM}}^{m} \\
& \times \frac{(n+m+1)!}{(n-m-1)!} - \sum_{m=0}^{n} c_n^* c_{n+1} g_{n+1,\mathrm{TM}}^{m+1} g_{n,\mathrm{TM}}^{m*} \frac{(n+m+2)!}{(n-m)!} \\
& \mp \sum_{m=-n-1}^{-1} c_n^* c_{n+1} g_{n,\mathrm{TM}}^{m+1*} g_{n+1,\mathrm{TM}}^{m} \frac{(n+|m|+1)!}{(n-|m|+1)!} \\
& + \sum_{m=-n}^{-1} c_n^* c_{n+1} g_{n+1,\mathrm{TM}}^{m+1} g_{n,\mathrm{TM}}^{m*} \frac{(n+|m|)!}{(n-|m|)!}\Bigg\} \\
& \times \int_0^x \left(\frac{\psi_n^* \psi_{n+1}}{|k_{sp}|^2 \rho} + \frac{\psi_n^{\prime *} \psi_{n+1}^{\prime}}{k^2 (n+1)^2}\rho \right) d\rho,
\end{aligned} \tag{31a}$$

$$\begin{aligned}
\mathcal{g}^{2}_{\left\{\begin{array}{c} x \\ y \end{array}\right\}} =& \frac{4\pi}{|k_{sp}|^2}|\eta_r|^2\left\{\begin{array}{c} \mathrm{Im} \\ \mathrm{Re} \end{array}\right\}\sum_{n=1}^{\infty}\Bigg\{ \pm \sum_{m=0}^{n+1} d_n^* d_{n+1} g_{n,\mathrm{TE}}^{m+1*} g_{n+1,\mathrm{TE}}^{m} \\
& \times \frac{(n+m+1)!}{(n-m-1)!} - \sum_{m=0}^{n} d_n^* d_{n+1} g_{n+1,\mathrm{TE}}^{m+1} g_{n,\mathrm{TE}}^{m*} \frac{(n+m+2)!}{(n-m)!} \\
& \mp \sum_{m=-n-1}^{-1} d_n^* d_{n+1} g_{n,\mathrm{TE}}^{m+1*} g_{n+1,\mathrm{TE}}^{m} \frac{(n+|m|+1)!}{(n-|m|+1)!} \\
& + \sum_{m=-n}^{-1} d_n^* d_{n+1} g_{n+1,\mathrm{TE}}^{m+1} g_{n,\mathrm{TE}}^{m*} \frac{(n+|m|)!}{(n-|m|)!}\Bigg\} \int_0^x \frac{\psi_n^* \psi_{n+1}}{k^2 (n+1)^2} \rho d\rho,
\end{aligned} \tag{31b}$$

$$\mathcal{G}^3_{\left\{\begin{array}{c} x \\ y \end{array}\right\}} = \frac{4i\pi}{k^2|k_{sp}|^2} \left\{\begin{array}{c} \mathrm{Im} \\ \mathrm{Re} \end{array}\right\} \sum_{n=1}^{\infty} \frac{2n+1}{[n(n+1)]^2} \eta_r^* c_n d_n^*$$
$$\left\{ \sum_{m=0}^{n} \left(g_{n,\mathrm{TM}}^{m+1} g_{n,\mathrm{TE}}^{m*} \pm g_{n,\mathrm{TM}}^{m} g_{n,\mathrm{TE}}^{m+1*} \right) \frac{(n+m+1)!}{(n-m-1)!} \right.$$
$$\left. - \sum_{m=-n}^{-1} \left(g_{n,\mathrm{TM}}^{m+1} g_{n,\mathrm{TE}}^{m*} \pm g_{n,\mathrm{TM}}^{m} g_{n,\mathrm{TE}}^{m+1*} \right) \frac{(n+|m|)!}{(n-|m|)!} \right\} \int_0^x \psi_n^* \psi_n' \rho d\rho. \tag{31c}$$

Now, the following relations are invoked [2]:

$$\psi_n'^* \psi_{n+1}' = \psi_n \psi_n'^* - \frac{(n+1)^2}{|M|^2 \rho^2} \psi_n^* \psi_{n+1} + \frac{n+1}{M\rho} \psi_{n+1} \psi_{n+1}^*, \tag{32a}$$

$$\psi_n^* \psi_{n+1} = -\psi_n^* \psi_n' + \frac{n+1}{M\rho} \psi_n \psi_n^*. \tag{32b}$$

Replacing (32) in (31) and after reintroducing the definitions of R_n and S_n given in (9) and (10), we arrive at the final expressions for $\mathcal{G}_x$, $\mathcal{G}_y$ and $\mathcal{G}_z$:

$$\mathcal{G}_{\left\{\begin{array}{c} x \\ y \end{array}\right\}} = -\frac{8\pi a^3}{|M|^2 x^3} \left\{\begin{array}{c} \mathrm{Im} \\ \mathrm{Re} \end{array}\right\} \sum_{n=1}^{\infty} \left[A_n^m \left(S_n^* + \frac{n+1}{M} R_{n+1} \right) \right.$$
$$\left. + B_n^m \left(-S_n + \frac{n+1}{M} R_n \right) + i C_n^m S_n \right], \tag{33a}$$

$$\mathcal{G}_z = \frac{16\pi a^3}{|M|^2 x^3} \mathrm{Im} \sum_{n=1}^{\infty} \sum_{m=-n}^{m} \left[D_n^m \left(S_n^* + \frac{n+1}{M} R_{n+1} \right) \right.$$
$$\left. + E_n^m \left(-S_n + \frac{n+1}{M} R_n \right) + i F_n^m S_n \right], \tag{33b}$$

where the coefficients G_n^m $(G = A, B, C, D, E, F)$ are given as

$$\begin{aligned}
A_n^m =& \frac{1}{(n+1)^2} \left\{ \sum_{m=0}^{n-1} c_n c_{n+1}^* g_{n,\mathrm{TM}}^{m+1} g_{n+1,\mathrm{TM}}^{m*} \frac{(n+m+1)!}{(n-m-1)!} \right. \\
&+ \sum_{m=0}^{n} c_n^* c_{n+1} g_{n,\mathrm{TM}}^{m*} g_{n+1,\mathrm{TM}}^{m+1} \frac{(n+m+2)!}{(n-m)!} \\
&- \sum_{m=-n-1}^{-1} c_n c_{n+1}^* g_{n,\mathrm{TM}}^{m+1} g_{n+1,\mathrm{TM}}^{m*} \frac{(n+|m|+1)!}{(n-|m|+1)!} \\
&\left. - \sum_{m=-n}^{-1} c_n^* c_{n+1} g_{n,\mathrm{TM}}^{m*} g_{n+1,\mathrm{TM}}^{m+1} \frac{(n+|m|)!}{(n-|m|)!} \right\},
\end{aligned} \tag{34a}$$

$$\begin{aligned}
B_n^m =& \frac{|\eta_r|^2}{(n+1)^2} \left\{ \sum_{m=0}^{n-1} d_n d_{n+1}^* g_{n,\mathrm{TE}}^{m+1} g_{n+1,\mathrm{TE}}^{m*} \frac{(n+m+1)!}{(n-m-1)!} \right. \\
&+ \sum_{m=0}^{n} d_n^* d_{n+1} g_{n,\mathrm{TE}}^{m*} g_{n+1,\mathrm{TE}}^{m+1} \frac{(n+m+2)!}{(n-m)!} \\
&- \sum_{m=-n-1}^{-1} d_n d_{n+1}^* g_{n,\mathrm{TE}}^{m+1} g_{n+1,\mathrm{TE}}^{m*} \frac{(n+|m|+1)!}{(n-|m|+1)!} \\
&\left. - \sum_{m=-n}^{-1} d_n^* d_{n+1} g_{n,\mathrm{TE}}^{m*} g_{n+1,\mathrm{TE}}^{m+1} \frac{(n+|m|)!}{(n-|m|)!} \right\},
\end{aligned} \tag{34b}$$

$$\begin{aligned}
C_n^m =& \frac{2n+1}{[n(n+1)]^2} \eta_r^* c_n d_n^* \\
&\times \left\{ \sum_{m=0}^{n} \left(g_{n,\mathrm{TM}}^{m+1} g_{n,\mathrm{TE}}^{m*} \pm g_{n,\mathrm{TM}}^{m} g_{n,\mathrm{TE}}^{m+1*} \right) \frac{(n+m+1)!}{(n-m-1)!} \right. \\
&\left. - \sum_{m=-n}^{-1} \left(g_{n,\mathrm{TM}}^{m+1} g_{n,\mathrm{TE}}^{m*} \pm g_{n,\mathrm{TM}}^{m} g_{n,\mathrm{TE}}^{m+1*} \right) \frac{(n+|m|)!}{(n-|m|)!} \right\},
\end{aligned} \tag{34c}$$

$$D_n^m = \frac{1}{(n+1)^2} c_n^* c_{n+1} g_{n,\mathrm{TM}}^{m*} g_{n+1,\mathrm{TM}}^{m} \frac{(n+|m|+1)!}{(n-|m|)!}, \tag{34d}$$

$$E_n^m = \frac{|\eta_r|^2}{(n+1)^2} d_n^* d_{n+1} g_{n,\mathrm{TE}}^{m*} g_{n+1,\mathrm{TE}}^{m} \frac{(n+|m|+1)!}{(n-|m|)!}, \tag{34e}$$

$$F_n^m = m\eta_r^* \frac{2n+1}{[n(n+1)]^2} c_n d_n^* g_{n,\mathrm{TM}}^{m} g_{n,\mathrm{TE}}^{m*} \frac{(n+|m|)!}{(n-|m|)!}. \tag{34f}$$

Inserting (33) and (34) back into (19) for $\mathbf{r}_{\mathrm{as}}$ and then using the result in (16) provides us with an analytic and closed-form expression for $\mathbf{F}_{\mathrm{ph}}$ in both the slip-flow and free molecular regimes.

3.2. Arbitrary-index particles

So far, only non-magnetic or dielectric particles have been considered. It is, however, possible to extend the analysis developed in the previous section in order to incorporate particles possessing magnetic responses and losses as well, or even metamaterial spheres, using the GLMT.

The procedure is similar to that presented by the author for on-axis axisymmetric beams in Ref. [35]. First, remember that the HSF $Q(r,\theta,\varphi)$ is related to the energy which is dissipated within the particle and that, when magnetic losses are presented, it is given as [3]

$$Q(r,\theta,\varphi) = -\frac{1}{2}\mathrm{Re}\left[\nabla\cdot\left(\mathbf{E}_{\mathrm{int}}(r,\theta,\varphi)\times\mathbf{H}_{\mathrm{int}}^*(r,\theta,\varphi)\right)\right], \tag{35}$$

where $\mathbf{H}_{\mathrm{int}}(r,\theta,\varphi)$ is the magnetic field distribution inside the particle which, according to the GLMT, reads as

$$\frac{H_r}{H_0} = \sum_{n=1}^{\infty}\sum_{p=-n}^{n}(-i)^{n+1}(2n+1)\, d_n g_{n,\mathrm{TE}}^{p} \frac{\psi_n(k_{sp}r)}{k_{sp}^2 r^2} P_n^{|p|}(\cos\theta)\, e^{ip\varphi}, \tag{36a}$$

$$\begin{aligned}\frac{H_\theta}{H_0} = \frac{1}{k_{sp}r}\sum_{n=1}^{\infty}\sum_{p=-n}^{n}(-i)^{n+1} E_n \Bigg\{ & d_n g_{n,\mathrm{TE}}^{p}\psi_n'(k_{sp}r)\,\tau_n^{|p|}(\cos\theta) \\ & - p\left(\frac{\mu_{sp}k}{\mu k_{sp}}\right)^{-1} c_n g_{n,\mathrm{TM}}^{p}\psi_n(k_{sp}r)\,\pi_n^{|p|}(\cos\theta)\Bigg\} e^{ip\varphi},\end{aligned} \tag{36b}$$

$$\frac{H_\varphi}{H_0} = \frac{i}{k_{sp}r}\sum_{n=1}^{\infty}\sum_{p=-n}^{n}(-i)^{n+1}E_n\Bigg\{pd_n g_{n,\mathrm{TE}}^{p}\psi_n'(k_{sp}r)\,\pi_n^{|p|}(\cos\theta) - \left(\frac{\mu_{sp}k}{\mu k_{sp}}\right)^{-1} c_n g_{n,\mathrm{TM}}^{p}\psi_n(k_{sp}r)\,\tau_n^{|p|}(\cos\theta)\Bigg\}e^{ip\varphi}. \tag{36c}$$

For an arbitrary-index particle having a complex permeability $\mu_{sp} = \mu' - i\mu''$, we introduce the vector identity $\nabla\cdot(\mathbf{E}\times\mathbf{H}^*) = (\nabla\times\mathbf{E})\cdot\mathbf{H}^* - \mathbf{E}\cdot(\nabla\times\mathbf{H}^*)$ and use Maxwell's equations to write $\nabla\times\mathbf{E}$ and $\nabla\times\mathbf{H}^*$ in terms of $\mathbf{H}$ and $\mathbf{E}^*$, respectively. From (35), one then obtains:

$$\begin{aligned}
Q(r,\theta,\varphi) &= -\frac{1}{2}\,\mathrm{Re}\left[-i\omega\mu_{sp}|\mathbf{H}_{\mathrm{int}}|^2 + i\omega\epsilon_{sp}^*|\mathbf{E}_{\mathrm{int}}|^2\right] \\
&= \frac{1}{2}\,\mathrm{Re}\left[\omega\mu''|\mathbf{H}_{\mathrm{int}}|^2 + \omega\epsilon''|\mathbf{E}_{\mathrm{int}}|^2\right] \\
&= \frac{1}{2}\,\mathrm{Re}\left[\omega\mu_m\mu_r''|\mathbf{H}_{\mathrm{int}}|^2 + \omega\epsilon_m\epsilon_r''|\mathbf{E}_{\mathrm{int}}|^2\right].
\end{aligned}$$

Extracting multiplicative factors of $|H_0|^2$ and $|E_0|^2$, using the relation $E_0 = H_0/\eta_0$ [33] and the definition of I_λ $(= |E_0|^2/2\eta_0)$, (35) can be recast under the form [34]:

$$\begin{aligned}
Q(r,\theta,\varphi) &= \left(\frac{\sigma_m}{\eta_m}\right)I_\lambda B_m(r,\theta,\varphi) + (\sigma_e\eta_m)\,I_\lambda B_e(r,\theta,\varphi) \\
&= k\mu_r'' I_\lambda B_m(r,\theta,\varphi) + k\epsilon_r'' I_\lambda B_e(r,\theta,\varphi),
\end{aligned} \tag{37}$$

where $\sigma_m = \omega\mu_m\mu_r''$ and $\sigma_e = \omega\epsilon_m\epsilon_r''$ are the electric and magnetic conductivities of the particle. In (37), the source strength $B_e(r,\theta,\varphi)$ coincides with the one appearing in (17), as expected for electric losses. A magnetic source strength $B_m(r,\theta,\varphi)$ in (37) indicates that radiation is absorbed in the particle due to magnetic losses. Equation (19) for $\mathbf{r}_{\mathrm{as}}$ is now replaced by a more general expression in which the electric term of (19) is complemented by a similar magnetic term:

$$\mathbf{r}_{\mathrm{as}} = \frac{\mu''}{2}I_\lambda\left[\mathcal{I}_{x,m}\hat{x} + \mathcal{I}_{y,m}\hat{y} + \mathcal{I}_{z,m}\hat{z}\right] + \frac{\epsilon_r''}{2}I_\lambda\left[\mathcal{I}_{x,e}\hat{x} + \mathcal{I}_{y,e}\hat{y} + \mathcal{I}_{z,e}\hat{z}\right], \tag{38}$$

where $\mathcal{I}_{x,e}$, $\mathcal{I}_{y,e}$ and $\mathcal{I}_{z,e}$ are exactly those calculated previously for a non-magnetic particle and given in (33). As for $\mathcal{I}_{x,m}$, $\mathcal{I}_{y,m}$ and $\mathcal{I}_{z,m}$, they can be found using the magnetic field expansions in (36) and following the steps that lead to (33).

There is, however, a clever way to calculate such integrals without re-doing all the calculations. Just as done in Refs. [34, 35], it is based on the observation that (18) and (36) are dual to each other, so that they are related according to the following replacements:

$$\begin{aligned} c_n &\rightarrow d_n, \\ d_n &\rightarrow -c_n, \\ \eta_r &\rightarrow \eta_r^{-1}, \\ g^p_{n,\mathrm{TM}} &\rightarrow g^p_{n,\mathrm{TE}}, \\ g^p_{n,\mathrm{TE}} &\rightarrow g^p_{n,\mathrm{TM}}, \end{aligned} \tag{39}$$

with $\eta_r = (\mu_{sp} k/\mu k_{sp})$. Instead of (8), c_n and d_n for arbitrary-index particles are now given by (see Eqs. (3.90) and (3.91) of Ref. [33]):

$$c_n = \frac{M\mu_r \left[\xi_n(x)\,\psi_n'(x) - \xi_n'(x)\,\psi(x)\right]}{\mu_r \xi_n(x)\,\psi_n'(Mx) - M\xi_n'(x)\,\psi_n(Mx)}, \tag{40a}$$

$$d_n = \frac{M^2 \left[\xi_n(x)\,\psi_n'(x) - \xi_n'(x)\,\psi(x)\right]}{M\xi_n(x)\,\psi_n'(Mx) - \mu_r \xi_n'(x)\,\psi_n(Mx)}. \tag{40b}$$

Therefore, application of (39) in (33) and (34) gives us for the magnetic contribution to the asymmetry vector:

$$\begin{aligned} \mathcal{I}_{\left\{\begin{array}{c} x,m \\ y,m \end{array}\right\}} = -\frac{8\pi a^3}{|M|^2 x^3} \left\{\begin{array}{c} \mathrm{Im} \\ \mathrm{Re} \end{array}\right\} \sum_{n=1}^{\infty} \Bigg[& \overline{A}_n^m \left(S_n^* + \frac{n+1}{M} R_{n+1}\right) \\ & + \overline{B}_n^m \left(-S_n + \frac{n+1}{M} R_n\right) \mp i\overline{C}_n^m S_n \Bigg], \end{aligned} \tag{41a}$$

$$\begin{aligned} \mathcal{I}_{z,m} = -\frac{16\pi a^3}{|M|^2 x^3} \,\mathrm{Im} \sum_{n=1}^{\infty} \sum_{m=-n}^{m} \Bigg[& \overline{D}_n^m \left(S_n^* + \frac{n+1}{M} R_{n+1}\right) \\ & + \overline{E}_n^m \left(-S_n + \frac{n+1}{M} R_n\right) + i\overline{F}_n^m S_n \Bigg], \end{aligned} \tag{41b}$$

where the coefficients $\overline{G}_n^m$ $(\overline{G} = \overline{A}, \overline{B}, \overline{C}, \overline{D}, \overline{E}, \overline{F})$ are given as

$$\overline{A}_n^m = |\eta_r|^{-2} B_n^m, \tag{42a}$$

$$\overline{B}_n^m = |\eta_r|^{-2} A_n^m, \tag{42b}$$

$$\overline{C}_n^m = \mp |\eta_r|^{-2} \left(C_n^m\right)^*, \tag{42c}$$

$$\overline{D}_n^m = |\eta_r|^{-2} E_n^m, \tag{42d}$$

$$\overline{E}_n^m = |\eta_r|^{-2} D_n^m, \tag{42e}$$

$$\overline{F}_n^m = -|\eta_r|^{2} \left(F_n^m\right)^*. \tag{42f}$$

For $+z$-propagating on-axis x-polarized axisymmetric beams, for which (see, for instance, Eq. (6.3) of Ref. [33]):

$$\begin{cases} g_{n,TM}^m = g_{n,TE}^m = 0, & |m| \neq 1 \\ g_{n,TM}^1 = g_{n,TM}^{-1} = i g_{n,TE}^1 = -i g_{n,TE}^{-1} = \frac{g_n}{2}, & \end{cases} \tag{43}$$

where g_n are known as the *special* BSCs, one infers that $\mathbf{F}_{\text{ph}} = F_{\text{ph}} \hat{z}$ and that $\mathbf{r}_{\text{as}}$ can be written in terms of J_1 as first deduced for arbitrary-index particles by Ambrosio in Ref. [35].

In addition, when the axisymmetric beam is a $+z$ propagating, x-polarized uniform plane wave, $g_n = \exp(ikz_0)$ [33], that is, the special BSCs are simple phase factors. In this case, (33) and (34) reveals that such BSCs appears in the force expressions under the form $|g_n|^2 = 1$. For arbitrary-index particles, such conditions allow us to recover Eqs. (9), (11) and (12) of Ref. [34] for plane wave incidence on arbitrary-index spherical particles.

It is also possible to extend the analysis and calculation of the asymmetry vector in order to incorporate concentric or multilayered spheres [46], or even geometries other than spherical, e.g., spheroidal particles [47], in particular by using the GLMT for spheroids [48] or cylindrical absorbers [49].

4. Conclusions

This work has proposed a theoretical framework within which photophoretic forces can be calculated for on- or off-axis arbitrary-shaped beams. The analysis is valid for both the free molecular and slip-flow regimes, for which particles are much smaller or much larger than the mean free path of gas molecules in the host medium, respectively. Incidentally, the continuum regime is also contemplated since it is a limiting case of the slip-flow regime of very small Knudsen numbers.

The analytic and closed-form expression for the asymmetry vector, besides involving an intricate dependence on the electromagnetic properties of the spherical micro-particle, incorporates arbitrary shaped beams with the help of the generalized Lorenz-Mie theory. It is now an easy and a computationally-efficient task to compute photophoretic forces for any light beam of interest, since everything that is required to know about it is embedded in the values of the beam shape coefficients, which can be easily calculated for a large number of laser beams of practical usage.

Several applications can benefit from this approach in the optical and infrared domains, including optical tweezers systems for trapping and manipulation of particles, atmospheric problems with suspended aerosols, transport mechanisms in combustion environments, particle levitation, optical trap displays for creating three-dimensional images in space, and so on.

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Appendix A.

In this Appendix, the solutions to the integrals (27) and (28) are listed. For $p \geq 0$ or $q \geq 0$ ($p < 0$ or $q < 0$), the integrals (27) carry a superscript '$+$' ('$-$').

For the integrals in (27):

$$\begin{aligned}\mathcal{G}^{+}_{\theta,1} =& \frac{2\,(n-q)}{(2n+1)\,(2n+3)}\frac{(n+q+1)!}{(n-q)!}\delta_{l,n+1}\\ &- \frac{2\,(l+q+2)}{(2l+1)\,(2l+3)}\frac{(l+q+1)!}{(l-q)!}\delta_{n,l+1},\end{aligned} \tag{A.1a}$$

$$\begin{aligned}\mathcal{G}^{-}_{\theta,1} =& -\frac{2}{(2n+1)\,(2n+3)}\frac{(n+|q|+1)!}{(n-|q|+1)!}\delta_{l,n+1}\\ &+ \frac{2}{(2l+1)\,(2l+3)}\frac{(l+|q|)!}{(l-|q|)!}\delta_{n,l+1},\end{aligned} \tag{A.1b}$$

$$\begin{aligned}\mathcal{G}^{+}_{\theta,2} =& -\frac{2\,(n+p+2)}{(2n+1)\,(2n+3)}\frac{(n+p+1)!}{(n-p)!}\delta_{l,n+1}\\ &+ \frac{2\,(l-p)}{(2l+1)\,(2l+3)}\frac{(l+p+1)!}{(l-p)!}\delta_{n,l+1},\end{aligned} \tag{A.2a}$$

$$\begin{aligned}\mathcal{G}^{-}_{\theta,2} =& \frac{2}{(2n+1)\,(2n+3)}\frac{(n+|p|)!}{(n-|p|)!}\delta_{l,n+1}\\ &- \frac{2}{(2l+1)\,(2l+3)}\frac{(l+|p|+1)!}{(l-|p|+1)!}\delta_{n,l+1},\end{aligned} \tag{A.2b}$$

$$\begin{aligned}\mathcal{G}^{+}_{\theta,3} =& \frac{2n\,(n+2)}{(2n+1)\,(2n+3)}\frac{(n+q+1)!}{(n-q-1)!}\delta_{l,n+1}\\ &- \frac{2l\,(l+2)}{(2l+1)\,(2l+3)}\frac{(l+q+2)!}{(l-q)!}\delta_{n,l+1},\end{aligned} \tag{A.3a}$$

$$\begin{aligned}\mathcal{G}^{-}_{\theta,3} =& -\frac{2n\,(n+2)}{(2n+1)\,(2n+3)}\frac{(n+|q|+1)!}{(n-|q|+1)!}\delta_{l,n+1}\\ &+ \frac{2l\,(l+2)}{(2l+1)\,(2l+3)}\frac{(l+|q|)!}{(l-|q|)!}\delta_{n,l+1},\end{aligned} \tag{A.3b}$$

$$\mathcal{G}_{\theta,4}^{+} = \frac{2n\,(n+2)}{(2n+1)\,(2n+3)} \frac{(n+p+2)!}{(n-p)!} \delta_{l,n+1} + \frac{2l\,(l+2)}{(2l+1)\,(2l+3)} \frac{(l+p+1)!}{(l-p-1)!} \delta_{n,l+1}, \tag{A.4a}$$

$$\mathcal{G}_{\theta,4}^{-} = \frac{2n\,(n+2)}{(2n+1)\,(2n+3)} \frac{(n+|p|)!}{(n-|p|)!} \delta_{l,n+1} - \frac{2l\,(l+2)}{(2l+1)\,(2l+3)} \frac{(l+|p|+1)!}{(l-|p|+1)!} \delta_{n,l+1}, \tag{A.4b}$$

$$\mathcal{G}_{\theta,5}^{+} = \frac{2}{2n+1} \frac{(n+q+1)!}{(n-q-1)!} \delta_{l,n}, \tag{A.5a}$$

$$\mathcal{G}_{\theta,5}^{-} = - \frac{2}{2n+1} \frac{(n+|q|)!}{(n-|q|)!} \delta_{l,n}, \tag{A.5b}$$

$$\mathcal{G}_{\theta,6}^{+} = \frac{2}{2n+1} \frac{(n+p+1)!}{(n-p-1)!} \delta_{l,n}, \tag{A.6a}$$

$$\mathcal{G}_{\theta,6}^{-} = - \frac{2}{2n+1} \frac{(n+|p|)!}{(n-|p|)!} \delta_{l,n}, \tag{A.6b}$$

while for (28):

$$\mathcal{G}_{\theta,7} = \frac{2}{(2n+1)\,(2n+3)} \frac{(n+|p|+1)!}{(n-|p|)!} \delta_{l,n+1} + \frac{2}{(2l+1)\,(2l+3)} \frac{(l+|p|+1)!}{(l-|p|)!} \delta_{n,l+1}, \tag{A.7}$$

$$\mathcal{G}_{\theta,8} = \frac{2n\,(n+2)}{(2n+1)\,(2n+3)} \frac{(n+|p|+1)!}{(n-|p|)!} \delta_{l,n+1} + \frac{2l\,(l+2)}{(2l+1)\,(2l+3)} \frac{(l+|p|+1)!}{(l-|p|)!} \delta_{n,l+1}, \tag{A.8}$$

$$\mathcal{G}_{\theta,9} = \frac{2}{2n+1} \frac{(n+|p|)!}{(n-|p|)!} \delta_{n,l}. \tag{A.9}$$

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