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Second-harmonic generation in the surface layer of a dielectric spheroidal particle: I. Analytical solution

© V.N. Kapshai , A.A. Shamyna¶, A.I. Talkachov¶¶

Francisk Skorina Gomel State University,
246019 Gomel, Belarus

e-mail: kapshai@rambler.ru, ¶ anton.shamyna@gmail.com, ¶¶ anton.talkachov@gmail.com

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The problem of second-harmonic generation by a plane elliptically polarized electromagnetic wave in a thin optically nonlinear surface layer of a dielectric particle shaped as an ellipsoid of revolution is solved. The generalized Rayleigh–Gans–Debye approximation is used for an analytical description with taking into account the difference in refractive indices of the medium corresponding to the frequencies of the exciting and generated radiation. The limiting forms of functions are obtained, with the use of which the electric field strength of the generated radiation is expressed. The order of dependence of these functions on the linear dimensions is found, when the lengths of the semiaxes of the particle are small compared to the wavelength of the exciting radiation and their ratio remains constant. It was found that the power density of the generated radiation in this case is determined to a greater extent by the chiral components of the nonlinear dielectric susceptibility tensor and is proportional to the fourth power of the length of the semiaxis of the particle, if the shape of the spheroidal particle differs significantly from the spherical one. The solution of this problem, obtained by other authors, is supplemented for the possibility of applying to the description of generation in the surface layer of a dielectric particle not only in the form of a prolate, but also in the form of an oblate spheroid. Corrections of inaccuracies and misprints made in similar works by other authors are proposed. The relationships between the formulas used in these works are found, taking into account the corrections and the formulas used in this work.

Keywords: second-harmonic generation, dielectric spheroidal particle, generalized Rayleigh–Gans–Debye model, small particle approximation, chiral component.

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Introduction

Second-harmonic generation was proven to be indispensable in studies of micro- and nanosized objects (medical and biological research included). It provides an opportunity both to visualize live cells, tissue, and collagen fibers [1] and to examine the properties of surfaces of dielectric particles [2] and molecules adsorbed on them [3].

According to the results of earlier research, the nonlinear response of metal and dielectric nanoparticles is directly affected by their size and shape [4]. The majority of studies published to date are focused on second-harmonic generation in spherical particles, since this is the most simple and symmetrical shape [2–5]. Particles with a single symmetry axis (spheroidal [6] and cylindrical [7–10]) are examined much less frequently. This is attributable to the fact that such studies raise the requirements imposed on the mathematical framework and the computation capacity. Axially symmetric particles are used in the design of metamaterials that allow for amplification of a nonlinear signal [11] and control over the intensity of electromagnetic waves propagating through interfaces [12].

Particles shaped as an ellipsoid of revolution (spheroid) are a generalization of spherical ones. Under the influence of external forces, spherical particles may deform and assume a spheroidal shape. In fact, it is virtually impossible to form dielectric particles of an ideal spherical shape due to the presence of irregularities on their surface. This is especially relevant to the formation of ultrasmall particles (with diameters below 100 nm), when the emerging irregularities become comparable to the linear dimensions of particles [13]. As far as we know, a sufficiently complete analytical model of nonlinear generation in the surface layer of spheroidal dielectric particles has not been developed yet.

The aim of the present study is to characterize analytically and reveal the specific features of second-harmonic generation in the surface layer of dielectric particles, which are shaped as an ellipsoid of revolution, using the generalized Rayleigh–Gans–Debye model [4] that was proven to be efficient in analyzing nonlinear generation in spherical dielectric particles.

In the first part, we

– provide formulae characterizing the spatial distribution of generated radiation,

– analyze limit forms of the used mathematical functions for a small-sized spheroidal particle,

– find a relation between the obtained expressions and formulae corresponding to the problem of second-harmonic generation in the surface layer of a spherical particle, and

– compare the results with the data obtained in other studies.

The following topics are covered in the second part of the study:

– the spatial distribution of generated radiation at key values of the problem parameters is illustrated using three-dimensional directivity patterns,

– mathematical properties characterizing the symmetry of directivity patterns are indicated for the functions used,

– the influence of individual parameters on the shape of the directivity pattern is characterized, and

– methods for estimating selectively the independent components of the nonlinear dielectric susceptibility tensor using the conditions of zero generation and generation of linearly polarized radiation are proposed.

Problem formulation

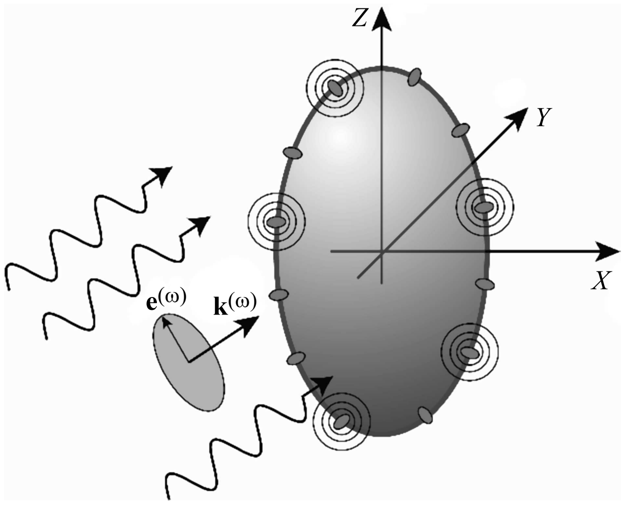
Let us assume that second-harmonic generation proceeds in a thin optically nonlinear layer distributed uniformly over the surface of a dielectric particle shaped as an ellipsoid of revolution. We denote the length of the semiaxis of an ellipsoid aligned with its symmetry axis as a_z and the length of the semiaxis perpendicular to the symmetry axis as a_x. The ratio of these quantities is denoted as $\rho = a_z/a_x$. If $\rho > 1$, a particle has the shape of an elongated ellipsoid of revolution, which may be produced by stretching a spherical shape along the prospective symmetry axis. If $\rho < 1$, a particle has the shape of an oblate ellipsoid of revolution, which may be produced by compressing a spherical shape along the prospective symmetry axis. If $\rho = 1$, the particle shape is spherical. Thickness d_0 of the optically nonlinear layer is chosen so that conditions $d_0 \ll a_x$, $d_0 \ll a_z$ are satisfied.

Following the line of reasoning from [5,8], we define the electric field vector of an incident plane electromagnetic wave at a point characterized by radius vector $\mathbf{x}$ as

$$\mathbf{E}(\mathbf{x}) = E_0 \mathbf{e}^{(\omega)} \exp(i\mathbf{k}^{(\omega)}\mathbf{x}), \tag{1}$$

where E_0 and $\mathbf{e}^{(\omega)}$ are the complex amplitude and a unit vector of polarization, respectively; the wave vector is denoted as $\mathbf{k}^{(\omega)}$. Temporal part $\exp(-i\omega t)$, where ω is the cyclic frequency of excitation radiation, is implied to be present here and elsewhere; if not a part of an index, symbol i denotes imaginary unit. The schematic diagram of the problem is presented in the figure.

Scattered electromagnetic waves are neglected in calculations within a model based on the generalized Rayleigh–Gans–Debye approximation. Such a model may be used if the refraction indices of a dielectric within and outside of the particle are fairly close [5]:



Schematic diagram of the problem of second-harmonic generation in the surface layer of a spheroidal particle.

$$\left|\frac{n_p}{n_m} - 1\right| \ll 1, \;\; 4\pi \frac{R}{\lambda} \left|\frac{n_p}{n_m} - 1\right| \ll 1, \tag{2}$$

where n_p and n_m are the refraction indices of the particle material and the environment, respectively, R is the characteristic particle size (e.g., major semiaxis length), and λ is the excitation radiation wavelength in vacuum.

The components of vector $\mathbf{P}^{(2)}$ (nonlinear part of the polarization vector enabling second-order nonlinear generation) may then be determined, in accordance with the dipole model, for a surface layer element using the rule of summation over repeated indices:

$$P_i^{(2)} = \chi_{ijk}^{(2)} E_j E_k, \tag{3}$$

where E_j, E_k are the jth and kth components of the electric field vector of excitation radiation, respectively. Tensor $\chi_{ijk}^{(2)}$ for second-harmonic generation contains only four independent components ($\chi_{1-3}^{(2)}$ are non-chiral, and $\chi_4^{(2)}$ is chiral) and may be written as follows in the component form:

$$\chi_{ijk}^{(2)} = \chi_1^{(2)} n_i n_j n_k + \chi_2^{(2)} n_i \delta_{jk} + \chi_3^{(2)} (n_j \delta_{ki} + n_k \delta_{ij}) + \chi_4^{(2)} n_m (n_k \varepsilon_{ijm} - n_j \varepsilon_{imk}). \tag{4}$$

Here, n_i, n_j, n_k, n_m are the components of a unit normal to a surface element, $\delta_{jk}, \delta_{ki}, \delta_{ij}$ are Kronecker delta symbols, and $\varepsilon_{ijm}, \varepsilon_{imk}$ are Levi-Civita symbols. The lower indices in formula (4) may assume values x, y, z that correspond to the axes of a right-hand orthonormal coordinate system. Only the case of real values of components $\chi_{1-4}^{(2)}$ is considered in the present study.

The objective is to derive and analyze formulae characterizing the spatial distribution and power of double-frequency radiation in the far-field region generated in the surface optically nonlinear layer of a spheroidal dielectric particle by a plane elliptically polarized electromagnetic wave.

Electric field of the second harmonic

The following expression may be used to determine the components of the electric field vector of the second harmonic in the far-field region:

$$E_i^{(2\omega)}(\mathbf{x}) = \mu_{2\omega}\frac{(2\omega)^2}{c^2}\frac{\exp(ik_{2\omega}r)}{r}d_0(\delta_{im} - e_{r,i}e_{r,m})e_j^{(\omega)}e_k^{(\omega)}$$

$$\times \int_S \exp(i\mathbf{x}'\mathbf{q}(\mathbf{x}))\chi_{mjk}^{(2)}(\mathbf{x}')dS_{\mathbf{x}'}. \quad (5)$$

Formula (5), which was derived in [4,5,8] with the use of the Green function method, is applicable in calculations of the electric field of radiation generated in the surface layer of dielectric particles of arbitrary shape. Integration should be performed over the entire area S of the particle surface covered with an optically nonlinear layer. The notation in formula (5) is as follows: $\mu_{2\omega}$ is the permeability of the environment, $k_{2\omega}$ is the modulus of wave vector $\mathbf{k}^{(2\omega)}$ of the generated wave, $r = |\mathbf{x}|$ is the distance from the geometric center of a dielectric particle to the observation point, $e_{r,i}$, $e_{r,m}$ are the components of unit vector $\mathbf{e}_r$ directed from the particle center to the observation point, $\mathbf{x}'$ is a vector directed from the center of a particle to its surface element, and $\mathbf{q}(\mathbf{x})$ is the scattering vector given by

$$\mathbf{q}(\mathbf{x}) = 2\mathbf{k}^{(\omega)} - \mathbf{k}^{(2\omega)}(\mathbf{x}),$$

$$\mathbf{k}^{(2\omega)} = \mathbf{k}^{(2\omega)}(\mathbf{x}) = k_{2\omega}\,\frac{\mathbf{x}}{r}. \quad (6)$$

When (4) is inserted into expression (5), it becomes necessary to calculate the following integrals over the surface of an ellipsoid of revolution:

$$I(n_i|\mathbf{x}) = \frac{1}{a_x^2}\int_S \exp(i\mathbf{x}'\mathbf{q}(\mathbf{x}))n_i(\mathbf{x}')dS_{\mathbf{x}'},$$

$$I(n_in_j|\mathbf{x}) = \frac{1}{a_x^2}\int_S \exp(i\mathbf{x}'\mathbf{q}(\mathbf{x}))n_i(\mathbf{x}')n_j(\mathbf{x}')dS_{\mathbf{x}'},$$

$$I(n_in_jn_k|\mathbf{x}) = \frac{1}{a_x^2}\int_S \exp(i\mathbf{x}'\mathbf{q}(\mathbf{x}))n_i(\mathbf{x}')n_j(\mathbf{x}')n_k(\mathbf{x}')dS_{\mathbf{x}'}. \quad (7)$$

Permutation symmetry is an important property of integrals (7):

$$I(n_in_j|\mathbf{x}) = I(n_jn_i|\mathbf{x}), \quad (8)$$

$$I(n_in_jn_k|\mathbf{x}) = I(n_in_kn_j|\mathbf{x}),\ \ I(n_in_jn_k|\mathbf{x}) = I(n_jn_in_k|\mathbf{x}). \quad (9)$$

The expression for the components of the electric field vector may then be written as

$$E_i^{(2\omega)}(\mathbf{x}) = \mu_{2\omega}\frac{(2\omega)^2}{c^2}\frac{\exp(ik_{2\omega}r)}{r}$$

$$\times d_0a_x^2(\delta_{im} - e_{r,i}e_{r,m})e_j^{(\omega)}e_k^{(\omega)}X_{mjk}^{(2\omega)}(\mathbf{x}), \quad (10)$$

where $X_{mjk}^{(2\omega)}$ is the effective nonlinear dielectric susceptibility tensor [5] that is expressed in terms of integrals (7) in the following way:

$$X_{mjk}^{(2\omega)}(\mathbf{x}) = \chi_1^{(2)}I(n_mn_jn_k|\mathbf{x}) + \chi_2^{(2)}I(n_m|\mathbf{x})\delta_{jk}$$

$$+ \chi_3^{(2)}\big(I(n_j|\mathbf{x})\delta_{km} + I(n_k|\mathbf{x})\delta_{mj}\big)$$

$$+ \chi_4^{(2)}\big(I(n_pn_k|\mathbf{x})\epsilon_{mjp} - I(n_pn_j|\mathbf{x})\epsilon_{mpk}\big). \quad (11)$$

The determination of the explicit form of integrals in formulae (7) is the most difficult part of calculation of the electric field of generated radiation.

Explicit form of auxiliary integrals

Let us introduce Cartesian coordinate system (x, y, z) with its center coinciding with the geometric center of a dielectric particle and axis Oz being aligned with the symmetry axis of this particle (see the figure). The equation characterizing the surface of an ellipsoidal particle with semiaxes a_x, a_x, a_z may then be written as

$$\frac{x^2}{a_x^2} + \frac{y^2}{a_x^2} + \frac{z^2}{a_z^2} = 1. \quad (12)$$

In the context of calculation of integrals (7), it is convenient to switch to the parametric form of this equation:

$$\mathbf{x}' = a_x\sin\theta'\cos\varphi'\mathbf{e}_x + a_x\sin\theta'\sin\varphi'\mathbf{e}_y + a_z\cos\theta'\mathbf{e}_z, \quad (13)$$

where $\mathbf{x}'$ is the radius vector of a surface element of an ellipsoid of revolution, θ', φ' are its angular coordinates, and $\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z$ are Cartesian vectors.

Using expression (13), we find the dependence of a unit normal to the particle surface on angular coordinates θ', φ':

$$\mathbf{n} = \frac{\left[\frac{\partial\mathbf{x}'}{\partial\theta'} \times \frac{\partial\mathbf{x}'}{\partial\varphi'}\right]}{\left|\frac{\partial\mathbf{x}'}{\partial\theta'} \times \frac{\partial\mathbf{x}'}{\partial\varphi'}\right|}$$

$$= \frac{a_z\sin\theta'\cos\varphi'\mathbf{e}_x + a_z\sin\theta'\sin\varphi'\mathbf{e}_y + a_x\cos\theta'\mathbf{e}_z}{\sqrt{a_z^2\sin^2\theta' + a_x^2\cos^2\theta'}}$$

$$= \frac{\rho\sin\theta'\cos\varphi'\mathbf{e}_x + \rho\sin\theta'\sin\varphi'\mathbf{e}_y + \cos\theta'\mathbf{e}_z}{\sqrt{\rho^2\sin^2\theta' + \cos^2\theta'}}. \quad (14)$$

The surface element area in integrals (7) is also calculated based on the expression for radius vector (13):

$$dS_{\mathbf{x}'} = |d\mathbf{S}_{\mathbf{x}'}| = \left|\frac{\partial \mathbf{x}'}{\partial \theta'} \times \frac{\partial \mathbf{x}'}{\partial \varphi'}\right| d\theta' d\varphi'$$

$$= a_x \sin\theta \sqrt{a_z^2 \sin^2\theta' + a_x^2 \cos^2\theta'}\, d\theta' d\varphi'. \qquad (15)$$

The components of scattering vector $\mathbf{q}$ are defined as

$$\mathbf{q} = q_x \mathbf{e}_x + q_y \mathbf{e}_y + q_z \mathbf{e}_z. \qquad (16)$$

Inserting (13)−(16) into (7), we obtain a more extensive form of auxiliary integrals depending on $\mathbf{x}$:

$$I(n_i|\mathbf{x}) = \frac{1}{a_x^2} \int\limits_S \exp(i\mathbf{x}'\mathbf{q}(\mathbf{x})) n_i(\mathbf{x}') dS_{\mathbf{x}'}$$

$$= \frac{1}{a_x^2} \int\limits_{-\pi}^{\pi} \int\limits_0^{\pi} \exp(iq_x a_x \sin\theta' \cos\varphi' + iq_y a_x \sin\theta' \sin\varphi'$$

$$+ iq_z a_z \cos\theta')$$

$$\times n_i(\theta', \varphi') a_x \sin\theta' \sqrt{a_z^2 \sin^2\theta' + a_x^2 \cos^2\theta'}\, d\theta' d\varphi'$$

$$= \int\limits_0^{\pi} \exp(iq_z a_z \cos\theta') \sqrt{\rho^2 \sin^2\theta' + \cos^2\theta'} \sin\theta'$$

$$\times \int\limits_{-\pi}^{\pi} \exp(iq_x a_x \sin\theta' \cos\varphi' + iq_y a_x \sin\theta' \sin\varphi')$$

$$\times n_i(\theta', \varphi') d\varphi' d\theta', \qquad (17)$$

$$I(n_i n_j|\mathbf{x}) = \int\limits_0^{\pi} \exp(iq_z a_z \cos\theta') \sqrt{\rho^2 \sin^2\theta' + \cos^2\theta'} \sin\theta'$$

$$\times \int\limits_{-\pi}^{\pi} \exp(iq_x a_x \sin\theta' \cos\varphi' + iq_y a_x \sin\theta' \sin\varphi')$$

$$\times n_i(\theta', \varphi') n_j(\theta', \varphi') d\varphi' d\theta', \qquad (18)$$

$$I(n_i n_j n_k|\mathbf{x}) =$$

$$= \int\limits_0^{\pi} \exp(iq_z a_z \cos\theta') \sqrt{\rho^2 \sin^2\theta' + \cos^2\theta'} \sin\theta'$$

$$\times \int\limits_{-\pi}^{\pi} \exp(iq_x a_x \sin\theta' \cos\varphi' + iq_y a_x \sin\theta' \sin\varphi')$$

$$\times n_i(\theta', \varphi') n_j(\theta', \varphi') n_k(\theta', \varphi') d\varphi' d\theta'. \qquad (19)$$

Integrals (17)−(19) may be calculated analytically with the use of infinite series. An incomplete calculation of this kind was performed in [6]. All combinations of component values (with i, j, k equal to x, y, z) should be considered separately. Since transformations needed to derive an explicit form of the indicated integrals are rather cumbersome, we provide only the end result. Function $M_{2\underset{\cdot}{s},\underset{\cdot}{c},\underset{\cdot}{q}}(z_1, z_2, \rho)$ needs to be introduced for this purpose:

$$M_{2\underset{\cdot}{s},\underset{\cdot}{c},\underset{\cdot}{q}}(z_1, z_2, \rho) = \frac{4\pi i^{2\{\underset{\cdot}{q}/2\}-\underset{\cdot}{c}}}{\rho^{2\underset{\cdot}{s}+\underset{\cdot}{c}+\underset{\cdot}{q}-1}} \sum_{\underset{\cdot}{g}=0}^{\infty} \sum_{\underset{\cdot}{n}=0}^{\infty} \sum_{\underset{\cdot}{m}=0}^{\underset{\cdot}{s}}$$

$$\times \sum_{\underset{\cdot}{k}=0}^{\underset{\cdot}{s}-\underset{\cdot}{m}} \sum_{\underset{\cdot}{l}=0}^{2\underset{\cdot}{m}+\underset{\cdot}{c}} (-1)^{\underset{\cdot}{k}+\underset{\cdot}{l}+\underset{\cdot}{g}} \left(2(\underset{\cdot}{q}/2 + \{\underset{\cdot}{q}/2\} + \underset{\cdot}{n} + \underset{\cdot}{g} + \underset{\cdot}{k}) - 1\right)!!$$

$$\times \binom{-(\underset{\cdot}{c} + \underset{\cdot}{q} - 1)/2 - \underset{\cdot}{s}}{\underset{\cdot}{n}} \binom{\underset{\cdot}{s}}{\underset{\cdot}{m}, \underset{\cdot}{k}, \underset{\cdot}{s} - \underset{\cdot}{m} - \underset{\cdot}{k}}$$

$$\times \binom{2\underset{\cdot}{m} + \underset{\cdot}{c}}{\underset{\cdot}{l}} \left(\frac{1}{\rho^2} - 1\right)^{\underset{\cdot}{n}} (\underset{\cdot}{q}/2 + \{\underset{\cdot}{q}/2\} + \underset{\cdot}{n} + \underset{\cdot}{g} + \underset{\cdot}{k})_{\underset{\cdot}{l}}$$

$$\times \frac{j^{(2\underset{\cdot}{m}+\underset{\cdot}{c}-\underset{\cdot}{l})}_{\underset{\cdot}{q}/2+\{\underset{\cdot}{q}/2\}+\underset{\cdot}{n}+\underset{\cdot}{g}+\underset{\cdot}{k}}(z_1)}{z_1^{\underset{\cdot}{q}/2+\{\underset{\cdot}{q}/2\}+\underset{\cdot}{n}+\underset{\cdot}{g}+\underset{\cdot}{k}+\underset{\cdot}{l}}} \frac{z_2^{2\underset{\cdot}{g}+2\{\underset{\cdot}{q}/2\}}}{(2\underset{\cdot}{g} + 2\{\underset{\cdot}{q}/2\})!}. \qquad (20)$$

The notation in formula (20) is as follows:

– all symbols with a dot below them ($\underset{\cdot}{s}$, $\underset{\cdot}{c}$, $\underset{\cdot}{q}$, $\underset{\cdot}{g}$, $\underset{\cdot}{n}$, $\underset{\cdot}{m}$, $\underset{\cdot}{k}$, $\underset{\cdot}{l}$, $\underset{\cdot}{d}$) are auxiliary non-negative integer indices;

– $\{z\}$ designates the fractional part of z;

– $(\underset{\cdot}{n})_{\underset{\cdot}{l}} = \frac{\Gamma(\underset{\cdot}{n}+\underset{\cdot}{l})}{\Gamma(\underset{\cdot}{n})}$ is the Pochhammer symbol expressed in terms of a gamma function;

– $\binom{\underset{\cdot}{s}}{\underset{\cdot}{m}} = \frac{\underset{\cdot}{s}!}{\underset{\cdot}{s}!(\underset{\cdot}{s}-\underset{\cdot}{m})!}$ and $\binom{\underset{\cdot}{s}}{\underset{\cdot}{m}, \underset{\cdot}{k}, \underset{\cdot}{s} - \underset{\cdot}{m} - \underset{\cdot}{k}} = \frac{\underset{\cdot}{s}!}{\underset{\cdot}{m}!\underset{\cdot}{k}!(\underset{\cdot}{s}-\underset{\cdot}{m}-\underset{\cdot}{k})!}$ are binomial and multinomial coefficients, respectively;

– $j_{\underset{\cdot}{n}}^{(\underset{\cdot}{d})}(z) = \frac{\partial^{\underset{\cdot}{d}} j_{\underset{\cdot}{n}}(z)}{\partial z^{\underset{\cdot}{d}}}$ — is the derivative of order $\underset{\cdot}{d}$ of a spherical Bessel function of order $\underset{\cdot}{n}$.

The infinite series in $\underset{\cdot}{n}$ in (20) converges only at $\rho^2 > 1/2$. A proof of this is provided in Appendix A. It may be proven in a similar fashion that the infinite series in (20) converges at arbitrary values of z_1 and z_2 in summation over indices $\underset{\cdot}{n}$ and $\underset{\cdot}{g}$.

In the case when the series in formula (20) diverges, one needs to use another form of function $M_{2s,c,q}(z_1, z_2, \rho)$:

$$M_{2s,c,q}(z_1, z_2, \rho) = 4\pi i^{2\{q/2\}-c} \sum_{g=0}^{\infty} \sum_{n=0}^{\infty} \sum_{m=0}^{s} \sum_{k=0}^{s-m} \sum_{l=0}^{2m+c}$$

$$\times \sum_{d=0}^{n} (-1)^{k+l+g+d} \big(2(q/2 + \{q/2\} + d + g + k) - 1\big)!!$$

$$\times \begin{pmatrix} -(c+q-1)/2 - s \\ -(c+q-1)/2 - s - n,\, n-d,\, d \end{pmatrix}$$

$$\times \begin{pmatrix} s \\ m, k, s-m-k \end{pmatrix} \begin{pmatrix} 2m+c \\ l \end{pmatrix}$$

$$\times (\rho^2 - 1)^n (q/2 + \{q/2\} + d + g + k)_l$$

$$\times \frac{j^{(2m+c-l)}_{q/2+\{q/2\}+d+g+k}(z_1)}{z_1^{q/2+\{q/2\}+d+g+k+l}} \frac{z_2^{2g+2\{q/2\}}}{(2g + 2\{q/2\})!}, \tag{21}$$

where the notation is similar to the one used in (20). Index d and the other indices also assume integer non-negative values. The convergence domain of the infinite series in summation over n in (21) is bounded by condition $0 < \rho^2 < 2$. The proof of this is similar to the one presented in Appendix A.

Thus, at $\rho^2 \geq 2$, one should use form (20) of function $M_{2s,c,q}(z_1, z_2, \rho)$; at $0 < \rho^2 \leq 1/2$, only form (21) is applicable; if condition $1/2 < \rho^2 < 2$, $\rho \neq 1$ is satisfied, both formulae are applicable. When $\rho = 1$, 0^0 indeterminancy emerges in formulae (20) and (21), which renders them unusable. Since a particle is spherical in this case, the formulae from [5], where second-harmonic generation in the surface layer of a spherical particle was characterized, should be used. The following trend is observed when either one of formulae (20), (21) are applied: the lower the value of $|\rho^2 - 1|$ is, the smaller is the number of terms in infinite sums over n and g needed to achieve the required accuracy of the value of function $M_{2s,c,q}(z_1, z_2, \rho)$. Analyzing formulae (20), (21), one may notice that, depending on the values of indices, function $M_{2s,c,q}(z_1, z_2, \rho)$ assumes real ($c + q$ is even) or purely imaginary ($c + q$ is odd) values.

The expressions for integrals (17)–(19) take the following form when function $M_{2s,c,q}(z_1, z_2, \rho)$ is used:

$$I((n_z)^m|\mathbf{x}) = M_{0,0,m}\big(q_\perp(\mathbf{x})a_x, q_z(\mathbf{x})a_z, \rho\big), \; m = 1, 2, 3, \tag{22}$$

$$I((n_z)^m n_i|\mathbf{x}) = \rho M_{0,1,m}\big(q_\perp(\mathbf{x})a_x, q_z(\mathbf{x})a_z, \rho\big)\nu_i, \; m=0, 1, 2, \tag{23}$$

$$I((n_z)^m n_i n_j|\mathbf{x}) = \rho^2 \Big(M_{0,2,m}\big(q_\perp(\mathbf{x})a_x, q_z(\mathbf{x})a_z, \rho\big)$$
$$- M_{2,0,m}\big(q_\perp(\mathbf{x})a_x, q_z(\mathbf{x})a_z, \rho\big)\Big)\nu_i\nu_j$$
$$+ \rho^2 M_{2,0,m}\big(q_\perp(\mathbf{x})a_x, q_z(\mathbf{x})a_z, \rho\big)\delta_{ij}, \; m = 0, 1, \tag{24}$$

$$I(n_i n_j n_k|\mathbf{x}) = \rho^3 \Big(M_{0,3,0}\big(q_\perp(\mathbf{x})a_x, q_z(\mathbf{x})a_z, \rho\big)$$
$$- 3M_{2,1,0}\big(q_\perp(\mathbf{x})a_x, q_z(\mathbf{x})a_z, \rho\big)\Big)\nu_i\nu_j\nu_k$$
$$+ \rho^3 M_{2,1,0}\big(q_\perp(\mathbf{x})a_x, q_z(\mathbf{x})a_z, \rho\big)\big(\nu_i\delta_{jk} + \nu_j\delta_{ki} + \nu_k\delta_{ij}\big). \tag{25}$$

The possible values of m are indicated in the corresponding formulae; indices i, j, k may assume values x or y; quantity $q_\perp$ is the modulus of component $\mathbf{q}$ of the scattering vector perpendicular to the particle symmetry axis; q_z is the projection of scattering vector $\mathbf{q}$ onto Cartesian axis Oz; and ν_i, ν_j, ν_k are the components of unit vector $\boldsymbol{\nu}$ codirectional with vector $\mathbf{q}_\perp$:

$$q_z = \mathbf{q}\mathbf{e}_z, \;\; \mathbf{q}_\perp = \mathbf{q} - q_z\mathbf{e}_z, \;\; q_\perp = |\mathbf{q}_\perp|, \;\; \boldsymbol{\nu} = \mathbf{q}_\perp/q_\perp. \tag{26}$$

Formulae (22)–(25) were verified by numerical integration at random parameter values for all possible combinations of indices i, j, k.

This compact form of formulae (22)–(25) eliminates the need to specify integrals (17)–(19) component-wise at different values of indices i, j, k. However, the values of integrals I at all the possible values of indices i, j, k, m are listed in Appendix B to make it easier to compare the results with the solution of a similar problem presented in [6].

Note that integrals $I(n_i|\mathbf{x})$ and $I(n_i n_j n_k|\mathbf{x})$ assume real values, while the values of integrals $I(n_i n_j|\mathbf{x})$ are purely imaginary. This is attributable to the specifics of functions $M_{2s,c,q}(z_1, z_2, \rho)$: functions $M_{0,0,1}$, $M_{0,1,0}$, which are used to define integrals $I(n_i|\mathbf{x})$, and functions $M_{0,0,3}$, $M_{0,2,1}$, $M_{2,0,1}$, $M_{2,1,0}$, $M_{0,3,0}$, which are used to define integrals $I(n_i n_j n_k|\mathbf{x})$, assume imaginary values, and functions $M_{0,1,1}$, $M_{0,0,2}$, $M_{2,0,0}$, $M_{0,2,0}$ found in the expressions for $I(n_i n_j|\mathbf{x})$ assume real values.

Limit forms of functions M

Limit forms of functions M ***at*** $\rho \to 1$

At limit values of certain parameters, function $M_{2s,c,q}(z_1, z_2, \rho)$ takes a simpler form and may be related to the already known functions used to solve problems of nonlinear generation.

If $\rho \to 1$, formulae (20) and (21) take the form

$$\lim_{\rho \to 1} M_{2s,c,q}(z_1, z_2, \rho) = 4\pi i^{2\{q/2\}-c} \sum_{g=0}^{\infty} \sum_{m=0}^{s} \sum_{k=0}^{s-m} \sum_{l=0}^{2m+c}$$

$$\times (-1)^{k+l+g} \big(2(q/2 + \{q/2\} + g + k) - 1\big)!!$$

$$\times \begin{pmatrix} s \\ m, k, s-m-k \end{pmatrix} \begin{pmatrix} 2m+c \\ l \end{pmatrix}$$

$$\times (q/2 + \{q/2\} + g + k)_l$$

$$\times \frac{j^{(2m+c-l)}_{q/2+\{q/2\}+g+k}(z_1)}{z_1^{q/2+\{q/2\}+g+k+l}} \frac{z_2^{2g+2\{q/2\}}}{(2g + 2\{q/2\})!}. \tag{27}$$

We managed to avoid summation over index n here, since all terms at $n > 0$ turn to zero.

At specific values of indices s, c, q, limit $\lim_{\rho\to 1} M_{2s,c,q}(z_1, z_2, \rho)$ in (27) assumes the following forms after simplification (here, $Z = \sqrt{z_1^2 + z_2^2}$):

$$\lim_{\rho\to 1} M_{0,0,1}(z_1, z_2, \rho) = 4\pi i \, \frac{z_2}{Z} \, j_1(Z), \tag{28}$$

$$\lim_{\rho\to 1} M_{0,0,2}(z_1, z_2, \rho) = 4\pi \left[\frac{1}{3}(j_0(Z) + j_2(Z)) - \left(\frac{z_2}{Z}\right)^2 j_2(Z) \right], \tag{29}$$

$$\lim_{\rho\to 1} M_{0,0,3}(z_1, z_2, \rho) = 4\pi i \left[\frac{3}{5}(j_1(Z) + j_3(Z)) \frac{z_2}{Z} - j_3(Z) \left(\frac{z_2}{Z}\right)^3 \right], \tag{30}$$

$$\lim_{\rho\to 1} M_{0,1,0}(z_1, z_2, \rho) = 4\pi i \, \frac{z_1}{Z} \, j_1(Z), \tag{31}$$

$$\lim_{\rho\to 1} M_{0,1,1}(z_1, z_2, \rho) = -4\pi \, \frac{z_1 z_2}{Z^2} \, j_2(Z), \tag{32}$$

$$\lim_{\rho\to 1} M_{0,1,2}(z_1, z_2, \rho) = 4\pi i \, \frac{z_1}{Z} \left[\frac{1}{5}(j_1(Z) + j_3(Z)) - j_3(Z) \left(\frac{z_2}{Z}\right)^2 \right], \tag{33}$$

$$\lim_{\rho\to 1} M_{2,0,0}(z_1, z_2, \rho) = 4\pi \, \frac{1}{3} \big(j_0(Z) + j_2(Z)\big), \tag{34}$$

$$\lim_{\rho\to 1} M_{2,0,1}(z_1, z_2, \rho) = 4\pi i \, \frac{1}{5} \, \frac{z_2}{Z} \big(j_1(Z) + j_3(Z)\big), \tag{35}$$

$$\lim_{\rho\to 1} M_{0,2,0}(z_1, z_2, \rho) = 4\pi \left(\frac{1}{3}\big(j_0(Z) + j_2(Z)\big) - j_2(Z) \left(\frac{z_1}{Z}\right)^2 \right), \tag{36}$$

$$\lim_{\rho\to 1} M_{0,2,1}(z_1, z_2, \rho) = 4\pi i \, \frac{z_2}{Z} \left(\frac{1}{5}\big(j_1(Z) + j_3(Z)\big) - j_3(Z) \left(\frac{z_1}{Z}\right)^2 \right), \tag{37}$$

$$\lim_{\rho\to 1} M_{2,1,0}(z_1, z_2, \rho) = 4\pi i \, \frac{1}{5} \, \big(j_1(Z) + j_3(Z)\big) \frac{z_1}{Z}, \tag{38}$$

$$\lim_{\rho\to 1} M_{0,3,0}(z_1, z_2, \rho) = 4\pi i \, \frac{z_1}{Z} \left(\frac{3}{5}\big(j_1(Z) + j_3(Z)\big) - j_3(Z) \left(\frac{z_1}{Z}\right)^2 \right). \tag{39}$$

Only the values of indices found in formulae for integrals (22)–(25) were considered in (28)–(39).

Inserting (28)–(39) into formulae (22)–(25) and the obtained result into formula (11), we find the expression for effective susceptibility tensor $X_{ijk}^{(2\omega)}$. In accordance with the correspondence principle, this expression matches a similar quantity in [5] for second-harmonic generation in the surface layer of a spherical particle.

Limit forms of function M at $z_2 \to 0$

Let us analyze the value of function $M_{2s,c,q}(z_1, z_2, \rho)$ at small values of z_1 and z_2. Condition $z_2 \to 0$ in the considered problem corresponds to the case when $q_z(\mathbf{x})a_z \to 0$. It is established if semiaxis length a_z is negligible comparable to the wavelength of excitation radiation or radiation is generated in directions where projection $q_z(\mathbf{x})$ of the scattering vector is close to zero. Condition $z_1 \to 0$ may be satisfied if $q_\perp(\mathbf{x})a_x \to 0$. When this is the case, semiaxis length a_x is small comparable to the wavelength of an incident electromagnetic wave or generation proceeds in directions where the modulus of the scattering vector projection onto the plane perpendicular to the particle symmetry axis tends to zero.

At $z_2 \to 0$, all terms containing factor $z_2^{2g+2\{q/2\}}$ at $g > 0$ in the sum over index g are negligible comparable to the terms at $g = 0$. Therefore, function $M_{2s,c,q}(z_1, z_2, \rho)$ may be written as

$$\begin{aligned} M_{2s,c,q}(z_1, z_2, \rho) &= \frac{4\pi i^{2\{q/2\}-c}}{\rho^{2s+c+q-1}} \sum_{n=0}^{\infty} \sum_{m=0}^{s} \\ &\times \sum_{k=0}^{s-m} \sum_{l=0}^{2m+c} (-1)^{k+l} \big(2(q/2 + \{q/2\} + n + k) - 1\big)!! \\ &\times \binom{-(c+q-1)/2 - s}{n} \binom{s}{m, k, s-m-k} \\ &\times \binom{2m+c}{l} \left(\frac{1}{\rho^2} - 1\right)^n (q/2 + \{q/2\} + n + k)_l \\ &\times \frac{j^{(2m+c-l)}_{q/2+\{q/2\}+n+k}(z_1)}{z_1^{q/2+\{q/2\}+n+k+l}} \, \frac{z_2^{2\{q/2\}}}{(2\{q/2\})!}. \end{aligned} \tag{40}$$

Since expression (40) was derived from formula (20), it is applicable only at $\rho^2 > 1/2$. Extending this reasoning to formula (21), we find that function $M_{2s,c,q}(z_1, z_2, \rho)$ at

$\rho^2 < 2$ may be written as

$$M_{2s,c,q}(z_1, z_2, \rho) = 4\pi i^{2\{q/2\}-c} \sum_{n=0}^{\infty} \sum_{m=0}^{s} \sum_{k=0}^{s-m} \sum_{l=0}^{2m+c}$$

$$\times \sum_{d=0}^{n} (-1)^{k+l+d} \big(2(q/2 + \{q/2\} + d + k) - 1\big)!!$$

$$\times \binom{-(c+q-1)/2 - s}{-(c+q-1)/2 - s - n,\ n-d,\ d}$$

$$\times \binom{s}{m,\ k,\ s-m-k} \binom{2m+c}{l}$$

$$\times (\rho^2 - 1)^n (q/2 + \{q/2\} + d + k)_l$$

$$\times \frac{j^{(2m+c-l)}_{q/2+\{q/2\}+d+k}(z_1)}{z_1^{q/2+\{q/2\}+d+k+l}} \frac{z_2^{2\{q/2\}}}{(2\{q/2\})!}. \tag{41}$$

It follows from (40) and (41) that $M_{2s,c,q}(z_1, z_2, \rho) \propto z_2^{2\{q/2\}}$. This implies that function $M_{2s,c,q}(z_1, z_2, \rho)$ is independent of z_2 at even q values; at odd values, $M_{2s,c,q}(z_1, z_2, \rho) \propto z_2$.

***Limit forms of function** M **at** $z_1 \to 0$*

Let us analyze the dependence of function M on z_1. A series expansion of the corresponding factor containing a spherical Bessel function [14] and depending on z_1 is needed for this purpose:

$$\frac{j_n^{(d)}(z_1)}{z_1^m} = z_1^{-m} \frac{\partial^d}{\partial z^d} \left(\sum_{p=0}^{\infty} \frac{(-1)^p z_1^{n+2p}}{2^p p!(2n+2p+1)!!} \right)$$

$$= \sum_{p=0}^{\infty} \frac{(-1)^p (n-d+2p+1)_d}{2^p p!(2n+2p+1)!!} z_1^{n-d-m+2p}. \tag{42}$$

Inserting formula (42) into (20) and simplifying the obtained expression at $z_1 \to 0$, we find that $M_{2s,c,q}(z_1, z_2, \rho) \propto z_1^{2\{c/2\}}$. The transformations are not detailed here owing to their cumbersomeness. Thus, function $M_{2s,c,q}(z_1, z_2, \rho)$ is independent of variable z_1 at even values of c; at odd c values, it is proportional to z_1.

The indicated trends are easy to verify by analyzing the simplified form of functions $M_{2s,c,q}(z_1, z_2, \rho)$ at fixed values of parameters s, c, q, which are given in Appendix C, or at $\rho \to 1$ (formulae (28)–(39)) or via numerical calculations at arbitrary values of arguments of function $M_{2s,c,q}(z_1, z_2, \rho)$.

***Limit forms of function** M **at** $z_1 \to 0$ **and** $z_2 \to 0$*

If z_1 and z_2 both assume small values, the following formula holds true:

$$M_{2s,c,q}(z_1, z_2, \rho) \propto z_1^{2\{c/2\}} z_2^{2\{q/2\}}, \ z_1 \to 0, \ z_2 \to 0. \tag{43}$$

In addition to functions $M_{2s,c,q}(z_1, z_2, \rho)$, one should also consider linear combinations of these functions found in integrals (22)–(25):

$$M_{0,2,0}(z_1, z_2, \rho) - M_{2,0,0}(z_1, z_2, \rho), \tag{44}$$

$$M_{0,2,1}(z_1, z_2, \rho) - M_{2,0,1}(z_1, z_2, \rho). \tag{45}$$

If functions $M_{2s,c,q}(z_1, z_2, \rho)$ in their explicit form are inserted into formulae (44) and (45), it becomes evident that these two expressions are directly proportional to $(z_1)^2$ and $(z_1)^2 z_2$, respectively, at $z_1 \to 0$, $z_2 \to 0$. This is attributable to the canceling out of terms containing factor z_1 and $z_1 z_2$ in formulae (44) and (45), respectively.

Limit forms of integrals I

Let us consider the dependence of integrals (22)–(25) on the linear dimensions of an ellipsoidal particle and the components of scattering vector $\mathbf{q}$ at $|q_z(\mathbf{x})a_z| \ll 1$ and $q_\perp(\mathbf{x})a_x \ll 1$. Using (43), we obtain

$$I((n_z)^m|\mathbf{x}) \propto (q_z(\mathbf{x})a_z)^{2\{m/2\}}, \ m = 1, 2, 3, \tag{46}$$

$$I((n_z)^m n_i|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x)(q_z(\mathbf{x})a_z)^{2\{m/2\}}, \ m = 0, 1, 2, \tag{47}$$

$$I((n_z)^m n_i n_j|\mathbf{x}) \propto (q_z(\mathbf{x})a_z)^{2\{m/2\}}, \ m = 0, 1, \tag{48}$$

$$I(n_i n_j n_k|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x). \tag{49}$$

Indices i, j, k may assume values x or y. The sole exceptions are the values of integrals $I(n_x n_y|\mathbf{x})$ and $I(n_z n_x n_y|\mathbf{x})$. They contain expressions (44) and (45), respectively. Therefore, the following formulae hold true for the indicated integrals:

$$I(n_x n_y|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x)^2, \tag{50}$$

$$I(n_z n_x n_y|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x)^2 (q_z(\mathbf{x})a_z). \tag{51}$$

Formulae characterizing the dependence of integrals $I(n_i|\mathbf{x})$, $I(n_i n_j|\mathbf{x})$, $I(n_i n_j n_k|\mathbf{x})$ on $q_\perp(\mathbf{x})a_x$ and $q_z(\mathbf{x})a_z$ at $|q_z(\mathbf{x})a_z| \ll 1$ and $q_\perp(\mathbf{x})a_x \ll 1$ and all the possible combinations of indices i, j, k are given in Appendix D. Note that the discovered trends are not only valid at small particle sizes ($a_z/\lambda \ll 1$, $a_x/\lambda \ll 1$), but are also typical of directions where specific components of the scattering vector are close to zero ($q_\perp \to 0$, $q_z \to 0$) even at relatively large linear dimensions of a spheroidal particle. The indicated forms of dependence of integrals I on linear particle dimensions agree with the formulae for functions listed in Table 5 in [6].

Dependences (46)–(51) may be used to characterize the dependence of effective susceptibility tensor $X_{ijk}^{(2\omega)}$ on linear dimensions of a small ($k_\omega a_x \ll 1$) dielectric particle. The expressions for non-chiral components, which feature coefficients $\chi_{1-3}^{(2)}$, also contain integrals $I(n_i|\mathbf{x})$ and $I(n_i n_j n_k|\mathbf{x})$. The indicated integrals are, in turn, proportional to a_x at a fixed ρ value. In the far-field region where the generated radiation is characterized by equations for plane waves, the modulus of the Umov–Poynting vector of a generated wave may be calculated as

$$S_r^{(2\omega)} \approx |\mathbf{S}^{(2\omega)}(\mathbf{x})| = \frac{c}{8\pi} \frac{n_{2\omega}}{\mu_{2\omega}} |\mathbf{E}^{(2\omega)}(\mathbf{x})|^2. \tag{52}$$

Therefore, the power density of the generated radiation for a non-chiral layer ($\chi^{(2)}_{1-3} \neq 0,\ \ \chi^{(2)}_4 = 0$) is directly proportional to a_x^6 for a spheroidal particle with small linear dimensions. Similar dependences for non-chiral components have already been found earlier in solving the problems of second-harmonic generation [5] and sum-frequency generation [15] in a spherical layer.

The expressions for chiral components including $\chi^{(2)}_4$ also contain integrals $I(n_i n_j|\mathbf{x})$ that do not depend on a_x. Therefore, the modulus of the Umov–Poynting vector for a chiral layer ($\chi^{(2)}_4 \neq 0$, $\chi^{(2)}_{1-3} = 0$) is directly proportional to a_x^4. A similar degree of dependence was found in the examination of sum-frequency generation in the surface layer of a spherical particle [15].

It is worth noting that the Umov–Poynting vector is directly proportional to a_x^8 in the case of second-harmonic generation in a chiral surface layer of a spherical particle [5]. This differs considerably from the result for a spheroidal dielectric particle ($S_r^{(2\omega)} \propto a_x^4$).

The probable reason for this is as follows. In accordance with formula (11), the value of integrals $I(n_m n_k|\mathbf{x})$ and $I(n_m n_j|\mathbf{x})$, which contain zero-order and second-order spherical Bessel functions at $\rho \to 1$, is required in calculation of the contribution of chiral component $\chi^{(2)}_4$ to generation. One may verify this assertion by inserting the values of functions $M_{2\varsigma,\varsigma,q}(z_1, z_2, \rho)$ from formulae (29), (32), (34), (36) of the present study into formulae (81)–(86) from Appendix D. However, the terms containing zero-order spherical Bessel functions are canceled out in summation over indices j and k, which emerges after the insertion of (11) into formula (5). The terms proportional to second-order spherical Bessel functions, which are proportional to a_x^2 in the case of small-sized particles, become dominant. The canceling out of zero-order spherical Bessel functions occurs only in the case of second-harmonic generation in a spherical layer; it does not occur in sum-frequency generation [15] in a spherical layer or second-harmonic generation in a spheroidal layer.

Comparison with the works of other authors

The problem of nonlinear generation in the surface layer of a spheroidal dielectric particle has been analyzed earlier in [6]. However, the obtained solution suffered from several drawbacks and inaccuracies.

(1) The obtained expressions in the form of series for characterization of integrals, which are similar to the ones written in (7), converge only at $\rho^2 > 1/2$ and are thus inapplicable to the process of generation in sufficiently oblate spheroidal particles. Notably, the authors of [6] do not indicate the limits of applicability of their model.

(2) The values of integrals in [6] were determined on the assumption that scattering vector $\mathbf{q}$ lies in plane Oxz. Therefore, it is not possible to calculate the numerical values of components of the electric field vector of generated radiation outside of the plane containing vector $\mathbf{q}$ without additional rotation transformations, which complicate the process of analysis of the spatial distribution of generated radiation.

(3) Several typing errors were made in Table 3 in [6]:

– the expression for $B_{x'z'}$ should not feature an imaginary unit. In other words, the expression for $B_{x'z'}$ should be imaginary (in common with the expressions for $B_{x'x'}$, $B_{y'y'}$, and $B_{z'z'}$ listed in the same table); this is also corroborated by one characteristic feature noted in the present study: the values of integrals $I(n_i n_j|\mathbf{x})$ corresponding to functions B_{ij} are real;

– variable $a_{z'}$ in the expression for $B_{x'z'}$ should be substituted with $q_{z'}$. This is corroborated by the fact that, in contrast to $q_{z'}$, $a_{z'}$ is not found in any other expression in [6].

With these corrections, the expression for $B_{x'z'}$ in [6] should read

$$B_{x'z'} = -3VDq_{x'}q_{z'}\sum_{n,h}^{\infty}\frac{(2n-1)!!}{(2n)!!} \times \frac{(2n+2h+1)!!}{2h+1}M(n,h)\kappa_{n+h+2}(q_{x'}D). \quad (53)$$

(4) A typing error was made in the series expansion in formula (27) in [6]: variable ρ in the denominator of expression $\cos^{2n}(t)/\rho^2$ should not be squared. This is easy to verify by inserting numerical values into the formula. The correct form of the mentioned expansion is

$$\frac{1}{\gamma(\rho,t)} = \frac{1}{(\cos^2 t + \rho^2\sin^2 t)^{1/2}} = \sum_{n=0}^{\infty}\left(\frac{\rho^2-1}{\rho^2}\right)^n \frac{(2n-1)!!}{(2n)!!}\frac{\cos^{2n} t}{\rho}. \quad (54)$$

(5) Factor 3 was omitted in the expression for vector $\mathbf{C}$ in formula (19) in [6]. The formula should be written as

$$\mathbf{C} = 3i\mathbf{q}V\frac{j_1[(q_{x'}^2+\rho^2q_{z'}^2)^{1/2}D]}{(q_{x'}^2+\rho^2q_{z'}^2)^{1/2}D}, \quad (55)$$

which is corroborated by the values of components of vector $\mathbf{C}$ listed in Table 3 in [6] and agrees with the results obtained in the present study.

With the indicated corrections introduced, the relation between the functions in the present study and in [6] is as follows:

$$C_i = a_x^2 I(n_i), \quad (56)$$

$$B_{ij} = a_x^2 I(n_i n_j), \quad (57)$$

$$A_{ijk} = a_x^2 I(n_i n_j n_k). \quad (58)$$

The designations adopted in [6] are shown on the left, and the designations from the present study for a special case of $q_y = 0$ and $\rho^2 > 1/2$ are on the right.

It is easy to verify that the values of integrals $I(n_i)$, $I(n_in_j)$, and $I(n_in_jn_k)$ also adhere to the properties indicated in [6]:

$$I(n_in_kn_k) = I(n_i), \tag{59}$$

$$I(n_kn_k) = I(1). \tag{60}$$

Expression $I(1)$ in [6] is designated as $f(\mathbf{q})$ and referred to as a surface linear form factor.

Conclusion

Finding the solution of the problem of second-harmonic generation in the surface layer of a particle shaped as an ellipsoid of revolution by expansion into a series and integration is the only currently available approach to an analytical treatment of this phenomenon. A compact form of resulting expressions written with the use of function $M_{2s,c,q}(z_1, z_2, \rho)$ was obtained for the first time. It is fairly convenient for subsequent analysis of the properties of the spatial distribution of double-frequency radiation and remains applicable at arbitrary ratio of linear dimensions of a spheroidal particle. The use of two forms of function $M_{2s,c,q}(z_1, z_2, \rho)$ provided an opportunity to expand the applicability domain of the analytical solution obtained in [6] to the characterization of generation of double-frequency radiation in the surface layer of a particle shaped as an ellipsoid of revolution. In order to verify the correctness of this solution, the relation between special cases of functions given in the present study and the functions used in [6] to characterize second-harmonic generation was established. It was found that the general properties typical of tensor integral quantities, which were mentioned for the first time in [6], also apply to auxiliary integrals I obtained in the present study.

In the limit of a spherical shape of a spheroidal particle ($\rho \to 1$), the functions used to characterize second-harmonic generation in the surface layer of a spheroidal particle transform into functions characterizing the spatial distribution of double-frequency radiation generated in the surface layer of a spherical dielectric particle [5]. This is in line with the correspondence principle.

The explicit form of functions $M_{2s,c,q}(z_1, z_2, \rho)$ at fixed values of indices s, c, q (Appendix C) may be used in further analysis to shorten the time needed for calculating the components of the electric field vector of the second harmonic. The nature of dependence of functions $M_{2s,c,q}(z_1, z_2, \rho)$ on small arguments z_1 and z_2 corresponds to the formulae given in Appendix C and was used successfully to determine the behavior of auxiliary integrals for a small-sized dielectric particle shaped as an ellipsoid of revolution.

The dependences provided in Appendix D allow one to determine the dominant components of the double-frequency electric field vector. Specifically, it was found that chiral components associated with coefficient $\chi_4^{(2)}$ produce the dominant contribution to generation in the case of small linear dimensions of a spheroidal particle when ρ differs substantially from 1. The power density of generated radiation is directly proportional to a_x^4. Under the same conditions, non-chiral components associated with coefficients $\chi_{1-3}^{(2)}$ induce the generation of radiation with its power density proportional to a_x^6. Similar results with a predominant contribution of chiral components to generation have been obtained earlier in studies focused on second-harmonic generation [8] and sum-frequency generation [16] in surface layers of dielectric particles shaped as cylinders.

The developed approach to the characterization of second-harmonic generation in deformed spherical particles may find application in analytical treatment of sum-frequency generation and other nonlinear effects in dielectric particles of a similar shape. The method of searching for an explicit form of integral tensor quantities by expansion into a series may also be applied to problems of nonlinear generation in particles of a more complex shape (arbitrary ellipsoid, elliptical cylinder, hemisphere, and their elements). The use of this method furthers the prospect of developing systematic approaches for high-accuracy characterization of nonlinear generation in particles of an arbitrary shape (higher-order generation included).

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Conflict of interest

The authors declare that they have no conflict of interest.

Appendix A. Proof of convergence of the sum of infinite series

Let us consider one of the terms in the sum written in (20). In order to do that, we fix the values of indices s, c, q, g, m, k, l, d, (all indices except for n) and variables ρ, z_1, z_2. The following expression is then obtained:

$$F_n = \frac{(-1)^{k+l+g}\left(q + 2\{q/2\} + 2n + 2g + 2k - 1\right)!!}{(2g + 2\{q/2\})!}$$
$$\times (q/2 + \{q/2\} + n + g + k)_l \binom{-(c + q - 1)/2 - s}{n}$$
$$\times \binom{s}{m, k, s, -m - k} \binom{2m + c}{l} \left(\frac{1}{\rho^2} - 1\right)^n$$
$$\times j^{(2m+c-l)}_{q/2+\{q/2\}+n+g+k}(z_1) z_1^{-(n+g+k+l)} z_2^{2g}. \tag{61}$$

Let us use the d'Alembert's ratio test to identify the conditions for convergence of the sum in n over $n \to \infty$. With this aim in view, we take the ratio limit of values of expression (61) at two consecutive values of index n at $n \to \infty$:

$$\lim_{n\to\infty} (F_{n+1}/F_n). \tag{62}$$

We then consider the ratios of the corresponding factors in functions F_{n+1} and F_n.

The first factor:

$$\frac{(-1)^{k+l+g}\left(q+2\{q/2\}+2(n+1)+2g+2k-1\right)!!}{(2g+2\{q/2\})!}$$
$$\times\left(\frac{(-1)^{k+l+g}\left(q+2\{q/2\}+2n+2g+2k-1\right)!!}{(2g+2\{q/2\})!}\right)^{-1}$$
$$= q+2\{q/2\}+2n+1+2g+2k. \tag{63}$$

The second factor:

$$\left(q/2+\{q/2\}+n+1+g+k\right)_l \Big/ \left(q/2+\{q/2\}+n+g+k\right)_l = \frac{q/2+\{q/2\}+n+1+g+k}{q/2+\{q/2\}+n+g+k}. \tag{64}$$

The third factor:

$$\binom{-(c+q-1)/2-s}{n+1}\Big/\binom{-(c+q-1)/2-s}{n}$$
$$=\frac{\Gamma(-(c+q-1)/2-s+1)}{\Gamma(n+2)\Gamma(-(c+q-1)/2-s-n)}\Big/\frac{\Gamma(-(c+q-1)/2-s+1)}{\Gamma(n+1)\Gamma(-(c+q-1)/2-s-n+1)}$$
$$=\frac{\Gamma(n+1)}{\Gamma(n+2)}\frac{\Gamma(-(c+q-1)/2-s-n+1)}{\Gamma(-(c+q-1)/2-s-n}$$
$$=\frac{-(c+q-1)/2-s-n}{1+n}. \tag{65}$$

Since the fourth and the fifth factors do not depend on n, their ratio at two consecutive values of n is unity. The sixth factor:

$$\left(\frac{1}{\rho^2}-1\right)^{n+1}\Big/\left(\frac{1}{\rho^2}-1\right)^{n} = \frac{1}{\rho^2}-1. \tag{66}$$

To perform calculations with the seventh factor, we first need to calculate limit $\lim_{n\to\infty} j^{(d)}_{n+1}(z)/j^{(d)}_n(z)$. The easiest way to do that is via a series expansion [14]:

$$j_n(z) = z^n\sum_{k=0}^{\infty}\frac{\left(-\frac{1}{2}z^2\right)^k}{k!(2n+2k+1)!!} = \sum_{k=0}^{\infty}\frac{(-1)^k z^{2k+n}}{2^k k!(2n+2k+1)!!}. \tag{67}$$

The following expansion then holds true for the derivative of order d:

$$j^{(d)}_n(z) = \sum_{k=0}^{\infty}\frac{(2k+n)!(-1)^k z^{2k+n-d}}{(2k+n-d)!2^k k!(2n+2k+1)!!}. \tag{68}$$

Therefore, the ratio of the corresponding functions at consecutive values of n may be written as

$$j^{(d)}_{n+1}(z)/j^{(d)}_n(z) = \frac{\sum_{k=0}^{\infty}\frac{(2k+n+1)!(-1)^k z^{2k+n+1-d}}{(2k+n-d+1)!2^k k!(2n+2k+3)!!}}{\sum_{k=0}^{\infty}\frac{(2k+n)!(-1)^k z^{2k+n-d}}{(2k+n-d)!2^k k!(2n+2k+1)!!}}$$
$$= z\frac{\sum_{k=0}^{\infty}\frac{(2k+n)!(-1)^k z^{2k}}{(2k+n-d)!2^k k!(2n+2k+1)!!}\frac{(2k+n+1)}{(2k+n-d+1)(2n+2k+3)}}{\sum_{k=0}^{\infty}\frac{(2k+n)!(-1)^k z^{2k}}{(2k+n-d)!2^k k!(2n+2k+1)!!}}. \tag{69}$$

At $n \to \infty$, we obtain

$$\lim_{n\to\infty}\frac{(2k+n+1)}{(2k+n-d+1)(2n+2k+3)} = \frac{1}{2n}. \tag{70}$$

Using (70) in (69), we find

$$\lim_{n\to\infty} z\frac{\sum_{k=0}^{\infty}\frac{(2k+n)!(-1)^k z^{2k}}{(2k+n-d)!2^k k!(2n+2k+1)!!}\frac{(2k+n+1)}{(2k+n-d+1)(2n+2k+3)}}{\sum_{k=0}^{\infty}\frac{(2k+n)!(-1)^k z^{2k}}{(2k+n-d)!2^k k!(2n+2k+1)!!}}$$
$$= \lim_{n\to\infty} z\frac{\frac{1}{2n}\sum_{k=0}^{\infty}\frac{(2k+n)!(-1)^k z^{2k}}{(2k+n-d)!2^k k!(2n+2k+1)!!}}{\sum_{k=0}^{\infty}\frac{(2k+n)!(-1)^k z^{2k}}{(2k+n-d)!2^k k!(2n+2k+1)!!}} = \frac{z}{2n}. \tag{71}$$

Note that the substitution of $\frac{(2k+n+1)}{(2k+n-d+1)(2n+2k+3)}$ with $\frac{1}{2n}$ in formula (71) is correct if $n \gg k$. However, the other terms of the sum for which n and k are of a similar order (or condition $k \gg n$ is satisfied) are negligible relative to the accurate value of the derivative corresponding to a spherical Bessel function.

The limit value of the ratio of derivatives of order d of $(n+1)$- and n-order spherical Bessel functions at $n \to \infty$ may be determined using the following expression:

$$\lim_{n\to\infty}\frac{j^{(d)}_{n+1}(z)}{j^{(d)}_n(z)} = \frac{z}{2n}. \tag{72}$$

Applying (72), we find

$$\lim_{n\to\infty}\frac{j^{(2m+c-l)}_{q/2+\{q/2\}+n+1+g+k}(z_1)}{j^{(2m+c-l)}_{q/2+\{q/2\}+n+g+k}(z_1)} = \frac{z_1}{2(q/2+\{q/2\}+n+g+k)}. \tag{73}$$

The ratio for the eighth and the ninth factors is

$$\frac{z_1^{-(n+1+g+k+l)}z_2^{2g}}{z_1^{-(n+g+k+l)}z_2^{2g}} = \frac{1}{z_1}. \tag{74}$$

Using (63)−(66), (73), and (74), we then find the limit of ratio F_{n+1}/F_n:

$$\lim_{n\to\infty}(F_{n+1}/F_n)=\lim_{n\to\infty}(q+2\{q/2\}+2n+2+2g+2k-1)$$
$$\times\frac{q/2+\{q/2\}+n+1+g+k}{q/2+\{q/2\}+n+g+k}\frac{-(c+q-1)/2-s-n}{1+n}$$
$$\times\left(\frac{1}{\rho^2}-1\right)\frac{z_1}{2(q/2+\{q/2\}+n+g+k)}\frac{1}{z_1}=-\left(\frac{1}{\rho^2}-1\right). \quad (75)$$

In accordance with the d'Alembert's ratio test, the series converges only if the following condition is satisfied:

$$\left|-\left(\frac{1}{\rho^2}-1\right)\right|<1. \quad (76)$$

This is possible if ρ^2 adheres to restriction

$$\rho^2>1/2. \quad (77)$$

Appendix B. Explicit form of integrals I

$$I(n_x|\mathbf{x})=\rho M_{0,1,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_x, \quad (78)$$

$$I(n_y|\mathbf{x})=\rho M_{0,1,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_y, \quad (79)$$

$$I(n_z|\mathbf{x})=\rho M_{0,0,1}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho), \quad (80)$$

$$I(n_xn_x|\mathbf{x})=\rho^2\big(M_{0,2,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_x\nu_x$$
$$+M_{2,0,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)(1-\nu_x\nu_x)\big), \quad (81)$$

$$I(n_xn_y|\mathbf{x})=\rho^2\big(M_{0,2,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)$$
$$-M_{2,0,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\big)\nu_x\nu_y, \quad (82)$$

$$I(n_yn_y|\mathbf{x})=\rho^2\big(M_{0,2,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_y\nu_y$$
$$+M_{2,0,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)(1-\nu_y\nu_y)\big), \quad (83)$$

$$I(n_zn_x|\mathbf{x})=\rho M_{0,1,1}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_x, \quad (84)$$

$$I(n_zn_y|\mathbf{x})=\rho M_{0,1,1}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_y, \quad (85)$$

$$I(n_zn_z|\mathbf{x})=M_{0,0,2}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho), \quad (86)$$

$$I(n_xn_xn_x|\mathbf{x})=\rho^3\big(M_{0,3,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_x\nu_x$$
$$+3M_{2,1,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)(1-\nu_x\nu_x)\big)\nu_x, \quad (87)$$

$$I(n_xn_xn_y|\mathbf{x})=\rho^3\big(M_{0,3,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_x\nu_x$$
$$+M_{2,1,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)(1-3\nu_x\nu_x)\big)\nu_y, \quad (88)$$

$$I(n_xn_yn_y|\mathbf{x})=\rho^3\big(M_{0,3,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_y\nu_y$$
$$+M_{2,1,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)(1-3\nu_y\nu_y)\big)\nu_x, \quad (89)$$

$$I(n_yn_yn_y|\mathbf{x})=\rho^3\big(M_{0,3,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_y\nu_y$$
$$+3M_{2,1,0}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)(1-\nu_y\nu_y)\big)\nu_y, \quad (90)$$

$$I(n_zn_xn_x|\mathbf{x})=\rho^2\big(M_{0,2,1}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_x\nu_x$$
$$+M_{2,0,1}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)(1-\nu_x\nu_x)\big), \quad (91)$$

$$I(n_zn_xn_y|\mathbf{x})=\rho^2\big(M_{0,2,1}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)$$
$$-M_{2,0,1}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\big)\nu_x\nu_y), \quad (92)$$

$$I(n_zn_yn_y|\mathbf{x})=\rho^2\big(M_{0,2,1}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_y\nu_y$$
$$+M_{2,0,1}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)(1-\nu_y\nu_y)\big), \quad (93)$$

$$I(n_zn_zn_x|\mathbf{x})=\rho M_{0,1,2}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_x, \quad (94)$$

$$I(n_zn_zn_y|\mathbf{x})=\rho M_{0,1,2}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho)\nu_y, \quad (95)$$

$$I(n_zn_zn_z|\mathbf{x})=M_{0,0,3}(q_\perp(\mathbf{x})a_x,q_z(\mathbf{x})a_z,\rho). \quad (96)$$

Appendix C. Simplified formulae for functions M

The following formulae hold true at $\rho^2>1/2$:

$$M_{0,0,q}(z_1,z_2,\rho)=\frac{4\pi i^{2\{q/2\}}}{\rho^{q-1}}$$
$$\times\sum_{g=0}^{\infty}\sum_{n=0}^{\infty}(-1)^g\big(2(q/2+\{q/2\}+n+g)-1\big)!!$$
$$\times\binom{-(q-1)/2}{n}\left(\frac{1}{\rho^2}-1\right)^n$$
$$\times\frac{j_{q/2+\{q/2\}+n+g}(z_1)}{z_1^{q/2+\{q/2\}+n+g}}\frac{z_2^{2g+2\{q/2\}}}{(2g+2\{q/2\})!}, \quad (97)$$

$$M_{0,1,q}(z_1,z_2,\rho)=\frac{4\pi i^{2\{q/2\}-1}}{\rho^q}\sum_{g=0}^{\infty}\sum_{n=0}^{\infty}\sum_{l=0}^{1}(-1)^{l+g}$$
$$\times\Big(2(q/2+\{q/2\}+n+g)-1\Big)!!\big(q/2+\{q/2\}+n+g\big)_l$$
$$\times\binom{-q/2}{n}\left(\frac{1}{\rho^2}-1\right)^n\frac{j^{(1-l)}_{q/2+\{q/2\}+n+g}(z_1)}{z_1^{q/2+\{q/2\}+n+g+l}}$$
$$\times\frac{z_2^{2g+2\{q/2\}}}{(2g+2\{q/2\})!}=\frac{4\pi i^{2\{q/2\}-1}}{\rho^q}$$

$$\times \sum_{g=0}^{\infty}\sum_{n=0}^{\infty}(-1)^{g+1}\Big(2(q/2+\{q/2\}+n+g)-1\Big)!!$$

$$\times \binom{-q/2}{n}\left(\frac{1}{\rho^2}-1\right)^{n}\frac{j_{q/2+\{q/2\}+n+g+1}(z_1)}{z_1^{q/2+\{q/2\}+n+g}}$$

$$\times \frac{z_2^{2g+2\{q/2\}}}{(2g+2\{q/2\})!}, \qquad (98)$$

$$M_{0,2,q}(z_1,z_2,\rho)=\frac{4\pi i^{2\{q/2\}-2}}{\rho^{1+q}}\sum_{g=0}^{\infty}\sum_{n=0}^{\infty}\sum_{l=0}^{2}(-1)^{l+g}$$

$$\times\big(2(q/2+\{q/2\}+n+g)-1\big)!!\binom{-(1+q)/2}{n}$$

$$\times\binom{2}{l}\left(\frac{1}{\rho^2}-1\right)^{n}(q/2+\{q/2\}+n+g)_l$$

$$\times\frac{j^{(2-l)}_{q/2+\{q/2\}+n+g}(z_1)}{z_1^{q/2+\{q/2\}+n+g+l}}\frac{z_2^{2g+2\{q/2\}}}{(2g+2\{q/2\})!}$$

$$=\frac{4\pi i^{2\{q/2\}-2}}{\rho^{1+q}}\sum_{g=0}^{\infty}\sum_{n=0}^{\infty}(-1)^{g}\big(2(q/2+\{q/2\}+n+g)-1\big)!!$$

$$\times\binom{-(1+q)/2}{n}\left(\frac{1}{\rho^2}-1\right)^{n}$$

$$\times\frac{j_{q/2+\{q/2\}+n+g+2}(z_1)-j_{q/2+\{q/2\}+n+g+1}(z_1)/z_1}{z_1^{q/2+\{q/2\}+n+g}}$$

$$\times\frac{z_2^{2g+2\{q/2\}}}{(2g+2\{q/2\})!}. \qquad (99)$$

The formulae for $0<\rho^2<2$ are as follows:

$$M_{0,0,q}(z_1,z_2,\rho)=4\pi i^{2\{q/2\}}\sum_{g=0}^{\infty}\sum_{n=0}^{\infty}\sum_{d=0}^{n}(-1)^{g+d}$$

$$\times\big(2(q/2+\{q/2\}+d+g)-1\big)!!$$

$$\times\binom{-(q-1)/2}{-(q-1)/2-n,\ n-d,\ d}(\rho^2-1)^{n}$$

$$\times\frac{j_{q/2+\{q/2\}+d+g}(z_1)}{z_1^{q/2+\{q/2\}+d+g}}\frac{z_2^{2g+2\{q/2\}}}{(2g+2\{q/2\})!}, \qquad (100)$$

$$M_{0,1,q}(z_1,z_2,\rho)=4\pi i^{2\{q/2\}-1}\sum_{g=0}^{\infty}\sum_{n=0}^{\infty}\sum_{l=0}^{1}\sum_{d=0}^{n}(-1)^{l+g+d}$$

$$\times\big(2(q/2+\{q/2\}+d+g)-1\big)!!$$

$$\times\binom{-q/2}{-q/2-n,\ n-d,\ d}(\rho^2-1)^{n}$$

$$\times(q/2+\{q/2\}+d+g)_l$$

$$\times\frac{j^{(1-l)}_{q/2+\{q/2\}+d+g}(z_1)}{z_1^{q/2+\{q/2\}+d+g+l}}\frac{z_2^{2g+2\{q/2\}}}{(2g+2\{q/2\})!},$$

$$=4\pi i^{2\{q/2\}-1}\sum_{g=0}^{\infty}\sum_{n=0}^{\infty}\sum_{d=0}^{n}(-1)^{1+g+d}$$

$$\times\big(2(q/2+\{q/2\}+d+g)-1\big)!!$$

$$\times\binom{-q/2}{-q/2-n,\ n-d,\ d}(\rho^2-1)^{n}$$

$$\times\frac{j_{q/2+(q/2)+d+g+1}(z_1)}{z_1^{q/2+\{q/2\}+d+g}}\frac{z_2^{2g+2\{q/2\}}}{(2g+2\{q/2\})!}, \qquad (101)$$

$$M_{0,2,q}(z_1,z_2,\rho)=4\pi i^{2\{q/2\}-2}\sum_{g=0}^{\infty}\sum_{n=0}^{\infty}\sum_{l=0}^{2}\sum_{d=0}^{n}(-1)^{l+g+d}$$

$$\times\big(2(q/2+\{q/2\}+d+g)-1\big)!!$$

$$\times\binom{-(q+1)/2}{-(q+1)/2-n,\ n-d,\ d}\binom{2}{l}(\rho^2-1)^{n}$$

$$\times(q/2+\{q/2\}+d+g)_l$$

$$\times\frac{j^{(2-l)}_{q/2+\{q/2\}+d+g}(z_1)}{z_1^{q/2+\{q/2\}+d+g+l}}\frac{z_2^{2g+2\{q/2\}}}{(2g+2\{q/2\})!}$$

$$=4\pi i^{2\{q/2\}}\sum_{g=0}^{\infty}\sum_{n=0}^{\infty}\sum_{d=0}^{n}(-1)^{g+d+1}$$

$$\times\big(2(q/2+\{q/2\}+d+g)-1\big)!!$$

$$\times\binom{-(q+1)/2}{-(q+1)/2-n,\ n-d,\ d}(\rho^2-1)^{n}$$

$$\times\frac{j_{q/2+\{q/2\}+d+g+2}(z_1)-j_{q/2+\{q/2\}+d+g+1}(z_1)/z_1}{z_1^{q/2+\{q/2\}+d+g}}$$

$$\times\frac{z_2^{2g+2\{q/2\}}}{(2g+2\{q/2\})!}. \qquad (102)$$

Appendix D. Dependence of auxiliary integrals on linear dimensions of a particle

$$I(n_x|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x), \tag{103}$$

$$I(n_y|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x), \tag{104}$$

$$I(n_z|\mathbf{x}) \propto (q_z(\mathbf{x})a_z), \tag{105}$$

$$I(n_xn_x|\mathbf{x}) \propto 1, \tag{106}$$

$$I(n_xn_y\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x)^2, \tag{107}$$

$$I(n_yn_y|\mathbf{x}) \propto 1, \tag{108}$$

$$I(n_zn_x|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x)(q_z(\mathbf{x})a_z), \tag{109}$$

$$I(n_zn_y|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x)(q_z(\mathbf{x})a_z), \tag{110}$$

$$I(n_zn_z|\mathbf{x}) \propto 1, \tag{111}$$

$$I(n_xn_xn_x|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x), \tag{112}$$

$$I(n_xn_xn_y|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x), \tag{113}$$

$$I(n_xn_yn_y|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x), \tag{114}$$

$$I(n_yn_yn_y|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x), \tag{115}$$

$$I(n_zn_xn_x|\mathbf{x}) \propto (q_z(\mathbf{x})a_z), \tag{116}$$

$$I(n_zn_xn_y|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x)^2(q_z(\mathbf{x})a_z), \tag{117}$$

$$I(n_zn_yn_y|\mathbf{x}) \propto (q_z(\mathbf{x})a_z), \tag{118}$$

$$I(n_zn_zn_x|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x), \tag{119}$$

$$I(n_zn_zn_y|\mathbf{x}) \propto (q_\perp(\mathbf{x})a_x), \tag{120}$$

$$I(n_zn_zn_z|\mathbf{x}) \propto (q_z(\mathbf{x})a_z). \tag{121}$$

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