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Analysis of the spatial distribution of the second-harmonic radiation generated in a thin surface layer of a spheroidal dielectric particle

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Abstract
Based on Rayleigh–Gans–Debye model, the spatial distribution of the second-harmonic radiation generated in a thin surface layer of a spheroidal dielectric particle is presented using three-dimensional directivity patterns. The peculiarities of forms of the directivity patterns are described for the key values of the parameters. The symmetries of the directivity patterns are

revealed, as in the case of second-harmonic generation in a surface layer of a spherical particle. The relationship between the polarization of the generated radiation and the polarization of the incident wave is described.

Introduction

Second-order nonlinear optical effects are widely used for the investigation of anisotropic properties of molecules and different structures. The second-harmonic generation (SHG) as the special case of nonlinear optical phenomenon of the even order may be used to get information about the thin surface layer of dielectric particles. The investigations of SHG were mostly devoted to generation in the surface layer of spherical [1, 2] and cylindrical particles [2, 3]. Some authors described the SHG in spheroidal dielectric particles [2]. Despite that, the analytical solution of the latter problems cannot be used to determine the spatial distribution of the generated radiation in some partial cases: if the particle has the form of an oblate spheroid, if the exciting radiation has the elliptical polarization, if one has to account the dispersion properties of the media in the problem. Some of these issues are solved during the research described in this paper.

Problem statement

Let an elliptically polarized electromagnetic wave propagate through a spheroidal dielectric particle covered by an optically nonlinear layer. Using the dipole model, the SHG radiation can be described by the second-order term in the expression for the polarization vector expressed using components of the electric field strength $E_{j,k}$:

$$P_i^{(2)} = \chi_{ijk}^{(2)} E_j E_k. \tag{1}$$

The tensor $\chi_{ijk}^{(2)}$ is the second-order nonlinear susceptibility tensor that depends on the components of the unit vector n_i perpendicular to the surface of the layer as follows:

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

The coefficients $\chi_{1-4}^{(2)}$ are independent components of the tensor $\chi_{ijk}^{(2)}$. For SHG, there are only four independent components in $\chi_{ijk}^{(2)}$.

Spatial distribution of the second harmonic radiation

The expression for the electric field strength of the generated radiation may be written as in [1]:

$$\mathbf{E}^{(2\omega)}(\mathbf{x}) = \mu_{2\omega} \frac{(2\omega)^2}{c^2} \frac{\exp(ik_{2\omega} r)}{r} (1 - \mathbf{e}_r \otimes \mathbf{e}_r) \int_V \exp\left(-i\mathbf{k}^{(2\omega)}(\mathbf{x})\ \mathbf{x}'\right) \mathbf{P}^{(2)}(\mathbf{x}')\mathrm{d}\mathbf{x}' \tag{3}$$

where $\mathbf{k}^{(2\omega)}$ ($k_{2\omega} = \left|\mathbf{k}^{(2\omega)}\right|$) is the wave vector of the second-harmonic (SH) wave, the multiplier $(1 - \mathbf{e}_r \otimes \mathbf{e}_r)$ eliminates the radial component of the value of the integral, ω is the cyclic frequency of the incident wave, r is the distance from the particle to the point of observation. The integration is performed within the volume of the thin optically nonlinear layer on the spheroidal particle. The integral in (3) may be found analytically using infinite series like in paper [2] where the case of prolate spheroid was considered.

Let us consider the radiation patterns for the SHG from the particles of different forms.

One can see in Fig. 1 the directivity patterns for the size of minor semiaxis of spheroidal particle characterized by the condition $k_\omega a_x = 0.1$ (k_ω is the magnitude of the wave vector of the incident wave, a_x – the length of minor semiaxis), the ratio of semiaxes equals 0.1 (oblate spheroid). These radiation patterns are plotted for the case when the incident wave has linear polarization and the electric field vector (the red arrow) is parallel to the symmetry axis of the spheroidal particle. The wave vector of the incident wave (the black arrow) is oriented perpendicularly to the symmetry axis. The polarization of the second-harmonic radiation is also linear (white color of the radiation patterns). The forms of radiation patterns differ from each other for different values of coefficients $\chi_{1-4}^{(2)}$ and have their own mirror and axial symmetries.

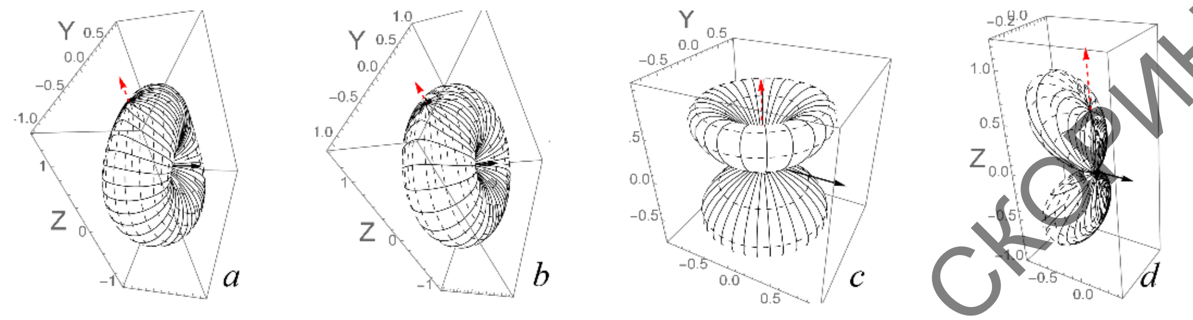

Fig. 1. The diagram of the second-harmonic radiation in the surface layer of the spheroidal particle:
$a - \chi_1^{(2)} \neq 0, \chi_{2-4}^{(2)} = 0$, $b - \chi_2^{(2)} \neq 0, \chi_{1,3,4}^{(2)} = 0$, $c - \chi_3^{(2)} \neq 0, \chi_{1,2,4}^{(2)} = 0$, $d - \chi_4^{(2)} \neq 0, \chi_{1-3}^{(2)} = 0$

The following directivity patterns in Fig. 2 are plotted for the elliptically polarized incident wave and the prolate form of the dielectric particle (the ratio of semiaxes is 1.5).

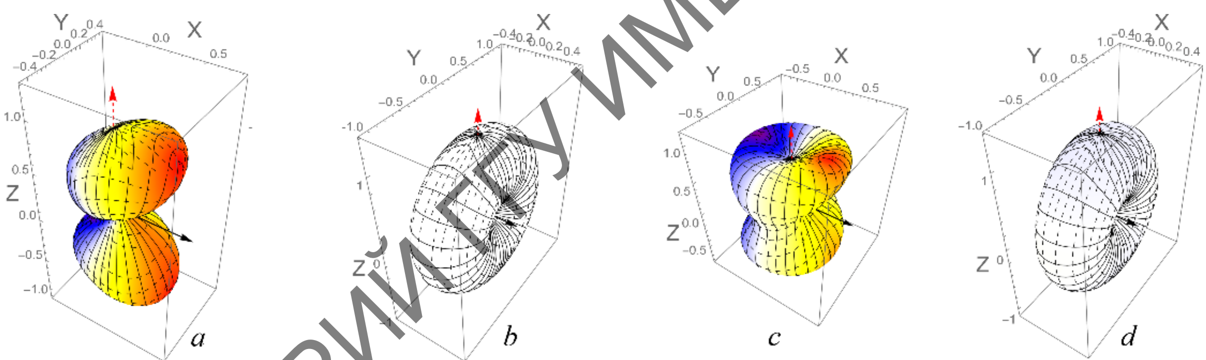

Fig. 2. The diagram of the SH-radiation spatial distribution caused by the elliptically polarized wave:
$a - \chi_1^{(2)} \neq 0, \chi_{2-4}^{(2)} = 0$, $b - \chi_2^{(2)} \neq 0, \chi_{1,3,4}^{(2)} = 0$, $c - \chi_3^{(2)} \neq 0, \chi_{1,2,4}^{(2)} = 0$, $d - \chi_4^{(2)} \neq 0, \chi_{1-3}^{(2)} = 0$

The main differences between the directivity patterns in Fig. 1 and Fig. 2 are:
– the polarization of the SH waves (elliptically/linearly polarized incident wave causes generation of elliptically/linearly polarized radiation (non-white color) for $\chi_1^{(2)} \neq 0$, $\chi_3^{(2)} \neq 0$ or $\chi_4^{(2)} \neq 0$);
– the form of the directivity pattern depends on the polarization of the incident wave and on the ratio of the semiaxes of the spheroidal particle.

Conclusion

The connection between the polarization of the incident waves and the polarization of the second-harmonic radiation is revealed. The dependence of the form of the directivity patterns on the form of the dielectric particle is also observed. The symmetries of the spatial distribution of the generated radiation are found for each considered case. Similar symmetries were previously discovered for the spherical and cylindrical form of the dielectric particle [1, 3].

This work was supported by Belarusian Republican Foundation for Fundamental Research (project No. F20M–011).

References

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