

https://doi.org/10.1070/QEL17998

Transformation of polarisation singularities in nonlinear mixing of light beams with frequency conversion in a medium with cubic nonlinearity

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Abstract. **We analyse the electric field transverse distributions in light beams with a tripled frequency and with a frequency of $2\omega_1 - \omega_2$, which arise in the bulk of an isotropic cubic-nonlinear medium upon the propagation of nonuniformly polarised beams of fundamental radiation with a multimode transverse structure described by a superposition of Laguerre–Gaussian modes. A relation is found between the total topological indices of the circular polarisation singularity points in the transverse sections of the generated signal beams and the corresponding total indices in the beams of fundamental radiation.**

Keywords: *cubic nonlinearity, polarisation singularity, topological index.*

1. Introduction

In 1950, S.I. Vavilov predicted that "…birefringence, dichroism and rotational polarisation strength depend on the light intensity" [1]. The intensity-dependent self-rotation of the polarisation ellipse, which increases with increasing degree of ellipticity in an incident plane wave and completely vanishes for linearly polarised light, was first observed by R. Terhune (Ford Motor Company, USA) [2] in 1964 in a medium with cubic nonlinearity. S.A. Akhmanov and V.I. Zharikov (Department of Physics, Lomonosov Moscow State University, USSR) in 1967 predicted the effect of nonlinear optical activity [3], i.e., intensity dependence of rotation of the polarisation plane of linearly polarised light incident on a medium with spatial dispersion of cubic nonlinearity. These studies stimulated further development of nonlinear polarisation optics.

Theoretical and experimental studies carried out to date allow a definite statement that the effects of polarisation self-action and wave interaction belong to subtle and widespread effects of nonlinear optics. A wave in devices of quantum electronics is generally elliptically polarised, and the degree of its ellipticity and the angle of inclination of the polarisation ellipse major axis can change when propagating through nonlinear crystals, reflecting from smooth surfaces, due to diffraction and dispersion effects. Moreover, when light beams interact in nonlinear media, their polarisation can change differently at different points of the cross section. In a number of cases, in an elliptically polarised pulse at the output of a nonlinear medium, it is possible to indicate its separate parts, in which the degrees of ellipticity differ significantly, the electric field vectors rotating in them in opposite directions.

An extensive list of spectroscopic schemes for the study of matter includes methods based on fixing changes in the polarisation states of waves during their interaction in a nonlinear medium. Being one of the most advanced, polarisation measurements make it possible to register rather weak changes in the degree of ellipticity and the angle of the major axis rotation of the signal wave polarisation ellipse and, therefore, to obtain high-accuracy spectroscopic data that are inaccessible to other research methods. The use of specially selected elliptically polarised waves of the fundamental radiation makes it possible to suppress the contributions of individual components of local, nonlocal, and surface nonlinear susceptibilities to the intensity and polarisation of the signal wave arising in the experiment.

In its development, nonlinear polarisation optics encountered singular optics that studies, in particular, singular points of the phase and polarisation of electric field. The former arise in a field uniformly polarised in space at points of its zero intensity. At these points, the field oscillation phase is indefinite. The points of phase singularity are characterised by the topological index I_Φ, which is equal to the change in the phase of field oscillations normalised to 2π during a roundtrip of the point along a small closed loop, which lies in a plane perpendicular to the direction of radiation propagation. Points of polarisation singularities arise in nonuniformly polarised harmonic light fields; at these points, indefinite is one of the polarisation ellipse characteristics. Generally, in non-paraxial electromagnetic fields, the polarisation ellipse is uniquely specified by two scalar and two vector quantities: the intensity, $|\boldsymbol{E}|^2$, the degree of ellipticity, $M = |\boldsymbol{E}^* \times \boldsymbol{E}|/|\boldsymbol{E}|^2$, the normal vector to the ellipse plane, $\boldsymbol{n} = \mathrm{Im}\{\boldsymbol{E}^* \times \boldsymbol{E}\}$, and the bidirectional vector (director) of its major axis, $\ddot{\boldsymbol{A}} = \pm \mathrm{Re}\{\boldsymbol{E}^* \sqrt{(\boldsymbol{EE})/|\boldsymbol{EE}|}\}$. Here $\boldsymbol{E}$ is the vector complex amplitude of a harmonically changing electric field. Using the bidirectional vector $\ddot{\boldsymbol{A}}$ is more convenient for determining the orientation of the major axis of the ellipse, since it does not have a preferred direction.

The vectors $\boldsymbol{n}$ and $\ddot{\boldsymbol{A}}$ are defined not for all states of polarisation of the electromagnetic field. If the polarisation ellipse turns into a circle ($M = 1$), the director $\ddot{\boldsymbol{A}}$ becomes indefinite, and if the polarisation is linear ($M = 0$), then the notion of a

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Received 6 December 2021
Kvantovaya Elektronika **52** (3) 247–253 (2022)
Translated by V.L. Derbov

normal to the ellipse plane loses sense. The points in space where this occurs form isolated lines known as C^T and L^T lines, or singularity lines of circular and linear polarisation, respectively [4]. In their vicinity, $\boldsymbol{n}$ and $\ddot{\boldsymbol{A}}$ change their orientation in space in a complicated way.

This is particularly pronounced in nonparaxial electromagnetic fields, where the isotropy parameters [5] are used to characterise polarisation singularities:

$$\Upsilon_{\mathrm{C}} = \frac{|\boldsymbol{E}^* \nabla(\boldsymbol{EE})|^2 - |\boldsymbol{E}\nabla(\boldsymbol{EE})|^2}{|\boldsymbol{E}^* \nabla(\boldsymbol{EE})|^2 + |\boldsymbol{E}\nabla(\boldsymbol{EE})|^2} \quad (1)$$

for C^T points and

$$\Upsilon_{\mathrm{L}} = \frac{e_{ilm} e_{jpq} T_{pl} T_{qm} D_{ij}}{\sum\limits_{r,s=x,y,z} [(\delta_{rl} - D_{rl})(\delta_{sm} - D_{sm}) T_{lm}]^2} \quad (2)$$

for L^T points. They generalise the topological index of the polarisation singularity, which is widely used in the study of singularity points in paraxial light fields. The index I_C is equal to the change in the major axis orientation angle of the polarisation ellipse normalised to 2π upon passing around the singularity along a small closed loop, which lies in the plane perpendicular to the direction of radiation propagation. In Eqns (1) and (2), $\nabla = (\partial_x, \partial_y, \partial_z)$ is a differential operator; $D_{ij} = E_i E_j^* / |\boldsymbol{E}|^2$ and $T_{ij} = \mathrm{Im}\{\sqrt{(\boldsymbol{EE})}\,\partial_j E_i^*\}$ are tensors (in the latter, the root sign is chosen arbitrarily); δ_{ij} and e_{ijk} are the Kronecker and Levi–Civita tensors; the summation is carried out over repeated indices $i, j, l, m, p, q \in \{x, y, z\}$; and x, y, and z are coordinates in an arbitrarily chosen Cartesian coordinate system.

The isotropy parameters coincide in sign with the topological indices of the patterns formed by the projections of the polarisation ellipses near the singularity points onto a specially chosen plane. For singularity points of circular polarisation, this plane coincides with the plane of rotation of the electric field strength vector, and for points of linear polarisation, it is perpendicular to the direction of oscillation of this vector. Polarisation singularities are also additionally characterised by the parameters

$$\eta_{\mathrm{C}} = \arg\{|\boldsymbol{E}\nabla(\boldsymbol{E}^*\boldsymbol{E}^*)|^3 \boldsymbol{E}\nabla(\boldsymbol{EE})\} \quad (3)$$

(for C^T points) and

$$\eta_{\mathrm{L}} = \arg\left\{\mathrm{Im}(\boldsymbol{E}^*[\nabla\times\boldsymbol{E}]) + \mathrm{i}\,\mathrm{Im}\left(\frac{0.5(\boldsymbol{E}^*\nabla)(\boldsymbol{EE}) - |\boldsymbol{E}|^2(\nabla\boldsymbol{E})}{\sqrt{(\boldsymbol{EE})}}\right)\right\} \quad (4)$$

(for L^T points), which are related to the spatial distribution of $\ddot{\boldsymbol{A}}$ and the director perpendicular to it, which specifies the orientation of the minor axis of the polarisation ellipse, in a neighbourhood of the singularity point.

Polarisation singularities are resistant to small perturbations of the electromagnetic field and transform during its propagation according to strictly defined laws [6]. The streamlines of the Umov–Poynting vector near the points of phase or polarisation singularity have a vortex structure; therefore, the study of optical singularities is closely related to the branch of optics that studies the transformation of the angular momentum of electromagnetic radiation (and separately its orbital and spin components) during propagation [7].

Few works are devoted to nonlinear processes in which polarisation singularities arise or interact. We can mention the analysis of the stabilisation of the propagation of a light beam with a polarisation singularity on its axis [8], the study of polarisation singularity formation during the propagation of circularly polarised radiation in a uniaxial KDP crystal, the symmetry of which is broken by an external electric field [9], and the classical nonlinear optical problem of second harmonic generation in a KTP crystal using radiation with a polarisation singularity [10]. The electric field of a third harmonic beam arising in an isotropic medium with cubic nonlinearity in the process of propagation of a monochromatic light beam containing a polarisation singularity of an arbitrary type is determined. The relationship between the characteristics of C^T points in the main and signal beams and the influence of wave vector detuning on the shape of C^T lines in a tripled-frequency beam is found [11].

From the point of view of singular polarisation optics, of particular interest are media with a nonlocal nonlinear optical response, which is exceptionally sensitive to the state of polarisation of the propagating light. The formation of nonuniformly polarised light beams in such media was theoretically predicted earlier [12–17] even in the case when the fundamental radiation beams have a Gaussian transverse profile and uniform polarisation.

Recently, the specificity of some nonlinear optical processes involving beams with polarisation singularities in these media has been studied. In Ref. [18], the ranges of ellipticity degree values for uniformly polarised fundamental radiation pulses propagating coaxially in an isotropic gyrotropic medium are determined, at which the change in the angle between the major axes of polarisation ellipses determines the total topological index of circular polarisation singularities arising in the sum frequency pulse generated in the volume of the medium. It was found [19, 20] that in the course of propagation of a light beam, consisting of a left-hand polarised Gaussian mode and two coaxial right-hand polarised first-order Laguerre–Gaussian modes, through an isotropic gyrotropic medium (limiting symmetry group $\infty\infty$) with spatial dispersion of the quadratic nonlinearity, the ellipticity degree modulus of the emerging second-harmonic radiation is equal to the modulus of the isotropy parameter of the polarisation singularity located at the axis of the fundamental radiation pulse propagation.

A striking example of an experiment in such media was the detection of interference between the processes of three-wave and five-wave mixing during the generation of the second harmonic by femtosecond laser pulses [21]. In Ref. [22], for the ellipticity degree of a Gaussian beam normally incident on the surface of an isotropic gyrotropic medium with a spatial dispersion of quadratic nonlinearity, analytical expressions were obtained, in which a polarisation singularity line is present in any cross section of the reflected beam at a doubled frequency. It is shown that if a beam of fundamental radiation normally incident on the boundary of an isotropic gyrotropic medium with a spatial dispersion of quadratic nonlinearity contains a solitary polarisation singularity on its axis, then the number of polarisation singularities and their total topological index in the reflected doubled-frequency beam are determined by the absence or presence of a nonlinear response of the near-surface layer of the medium [23, 24]. It is found that the propagation of a Gaussian light beam having a uniform elliptical polarisation at the boundary of a medium with a cubic nonlinear response (electronic or orientational) can be accompanied by the formation of closed circular polarisation singularity lines that lie in planes perpendicular to the beam axis [25]. It was demonstrated that, upon self-focusing of an elliptically polarised Gaussian beam in the isotropic phase of a nematic liquid crystal near the mesophase transition temperature, polarisation singularities of its electric field are generated in the interior of this crystal for almost any polarisation of the incident radiation [26]. The main results of Refs [11, 18–20, 22–26] are presented in more detail in brief review [27].

Below, for the first time, we present the results of a study of the interaction of polarisation singularities in the case of nonlinear mixing of waves with frequency conversion in a medium with cubic nonlinearity. The process of third harmonic generation in an isotropic medium by a nonuniformly polarised beam of fundamental radiation, whose transverse structure is a superposition of circularly polarised Laguerre–Gaussian modes, is considered. We also analyse the transformation of polarisation singularities during the generation of radiation at a frequency of $2\omega_1 - \omega_2$ in a medium with spatial dispersion of cubic nonlinearity in the case of collinear interaction of two such beams with frequencies ω_1 and ω_2.

2. Third harmonic generation in a bulk isotropic medium

Let a paraxial monochromatic light beam propagate in a bulk isotropic medium. Let us direct the z axis of the cylindrical coordinate system $r\varphi z$ along its axis and write the equation for slowly varying complex amplitudes $E_\pm(r,\varphi,z)$ of its right- and left-hand polarised components:

$$2ik_\omega\partial_z E_\pm + \Delta_\perp E_\pm = 0, \quad (5)$$

where $\Delta_\perp = \partial_r^2 + r^{-1}\partial_r + r^{-2}\partial_\varphi^2$; $k_\omega = \omega n_\omega/c$ is the wavenumber; n_ω is the real-valued refractive index of the medium at frequency ω; and c is the speed of light in vacuum. We represent its solution as

$$E_\pm(r,\varphi,z) = \sum_{n,l} c_{nl\pm}\Lambda_n^{(l)}(r/w,\varphi,\beta(z)), \quad (6)$$

where $c_{nl\pm}$ are the complex coefficients determined by the initial conditions; n and l are integers, $n \geqslant 0$; w is the characteristic transverse size of the beam; $\beta(z) = 1 + i(z - z_0)/z_\omega$; $z_0 > 0$ is the beam waist coordinate in the medium; and $z_\omega = k_\omega w^2/2$ is the beam diffraction length. In (6), the functions

$$\Lambda_n^{(l)}(\rho,\varphi,\beta) = \frac{\rho^{|l|}}{\beta^{n+|l|+1}} L_n^{(|l|)}\left[\frac{\rho^2}{\beta}\right]\exp\left(il\varphi - \frac{\rho^2}{\beta}\right) \quad (7)$$

define the transverse modes of the beam. Here $\rho = r/w$, and $L_n^{(l)}$ are generalised Laguerre polynomials.

It was shown in Ref. [20] that the beams, which are similarly polarised at any point of the cross section and are expressed by the right-hand side of Eqn (6), contain phase singularity points in their cross section, at which the field intensity turns into zero. The total topological index I_Φ of these singularities is defined as the field phase change normalised to 2π, calculated along the closed contour Γ, which embraces all singular points in the beam cross section:

$$I_\Phi = \frac{1}{2\pi}\int_\Gamma d\arg E = \frac{1}{2\pi i}\int_\Gamma d\ln E, \quad (8)$$

where E is a slowly varying scalar amplitude of a uniformly polarised beam. The total topological indices $I_{\Phi\pm}$ of the phase singularities of the $E_\pm$ components of form (5) can be found separately [20] by rearranging the terms in Eqn (6):

$$E_\pm = \exp\left(-\frac{r^2}{w^2\beta}\right)\sum_{p=0}^{\tilde{p}_\pm}\left(\frac{r}{w}\right)^p\sum_{s=0}^{p} a_{ps\pm}(z)\exp[i(2s-p)\varphi]. \quad (9)$$

Here $\tilde{p}_\pm$ is the highest degree of the corresponding polynomial at the exponential function in Eqns (6), (7). The form of the functions $a_{ps\pm}(z)$ is determined by the coefficients $c_{nl\pm}$ and the coefficients that enter the Laguerre polynomials (see [20]).

The values of $I_{\Phi\pm}$ in this case are determined by simple relations

$$I_{\Phi\pm} = 2N_\pm - \tilde{p}_\pm \quad (10)$$

where $N_\pm$ is the number of complex roots of the equation

$$\sum_{s=0}^{\tilde{p}_\pm} a_{\tilde{p}\pm s\pm}(z)u^s = 0$$

satisfying the condition $|u| < 1$, and, in fact, do not depend on the coordinate z, since the functions $a_{\tilde{p}\pm s\pm}(z)$ depend on z in a similar way (see Ref. [20] for details). In our case, the beam under consideration is nonuniformly polarised, and each point of the phase singularity of the component E_+ (or E_-) with a specific value of the topological index I corresponds to a singularity point of the left-hand circular (or right-hand circular) polarisation with the topological index $I/2$ (or $-I/2$) [6]. As a result, the total topological indices of the right-hand (I_{C+}) and left-hand (I_{C-}) circular polarisation singularities in the beam at the frequency ω are determined by the relations

$$I_{C\pm} = \mp 0.5 I_{\Phi\mp}. \quad (11)$$

The problem of third harmonic generation in a bulk isotropic medium is classical for nonlinear optics due to the relative simplicity of its solution and the beauty of the results obtained [11, 28, 29]. It is important to emphasise that in the volume of an isotropic medium it is impossible to implement it with a circularly polarised beam of fundamental radiation, because circularly polarised components of the nonlinear polarisation field $P_\pm^{(3\omega)}$ of the substance at triple frequency are proportional to the product of amplitudes E_+E_-:

$$P_\pm^{(3\omega)} = \chi^{(3)}E_+E_-E_\pm, \quad (12)$$

where $\chi^{(3)}$ is a constant specifying all nonzero components of the isotropic medium material tensor $\chi_{ijkl}^{(3)}(3\omega;\omega,\omega,\omega)$, symmetric with respect to the permutation of the last three indices. Substituting (6) into (12) allows writing equations for slowly varying complex amplitudes $E_{3\pm}(r,\varphi,z)$ of the circularly polarised components of the electric field of the third harmonic beam arising in a nonlinear medium:

$$2\mathrm{i}k_{3\omega}\partial_z E_{3\pm} + \Delta_\perp E_{3\pm}$$

$$= -4\pi n_{3\omega}^{-2} P_{\pm}^{(3\omega)} \exp[\mathrm{i}(3k_\omega - k_{3\omega})z]. \quad (13)$$

Here $k_{3\omega} = 3\omega n_{3\omega}/c$; $n_{3\omega}$ is the real-valued refractive index of the medium at a frequency of 3ω; we neglect the back effect of the beam at the tripled frequency on the fundamental radiation beam. The solution of Eqns (13) with the initial conditions $E_{3\pm}(r,\varphi,z=0) = 0$ is well-known and can be written in terms of the Green function.

The determination of the total topological indices $I_{\Phi 3\pm}$ of the phase singularities of the found components $E_{3\pm}$ and, therefore, the total topological indices $I_{C3\pm}$ of the polarisation singularities in the tripled-frequency beam, at first glance, seems to be a qualitatively more difficult problem, since the solutions of Eqns (13), even in the simplest cases, can be written only in quadratures [11, 28]. However, in Ref. [20], where an equation absolutely similar to Eqn (13) was studied, it was shown that the indices $I_{\Phi 3\pm}$ of the components of the generated signal beam field $E_{3\pm}$ are equal to the corresponding total indices of the components of the generating nonlinear polarisation field $P_{\pm}^{(3\omega)}$. The latter are easily found due to the exceptional simplicity of Eqns (12), as well as the fact that the total topological index of phase singularities of a field proportional to the product of several fields is equal to the sum of the total topological indices of phase singularities of each field in the product (see the last expression in the definition (8)). Thus, taking Eqns (11) into account, we finally obtain the relations:

$$I_{\Phi 3\pm} = 2I_{\Phi\pm} + I_{\Phi\mp} \Rightarrow I_{C3\pm} = 2I_{C\pm} + I_{C\mp}. \quad (14)$$

Formulae (14) obtained for an arbitrary transverse structure of the main radiation beam confirm the results of Ref. [11], in which a particular case was considered, when the circularly polarised components of the fundamental radiation beam were

$$E_+ = 2^{-1/2} E_{\mathrm{L}} (p\Lambda_0^{(1)} + q\Lambda_0^{(-1)}), \quad (15)$$

$$E_- = E_{\mathrm{G}} \Lambda_0^{(0)}, \quad (16)$$

where E_{L} and E_{G} are constants that determine the amplitude ratio of the circularly polarised beam components. Its left-hand polarised component is an ordinary Gaussian transverse mode, and its right-hand polarised component is a linear combination of two Laguerre–Gaussian modes with indices +1 and –1. As a result, the fundamental radiation beam contains a left-hand circular polarisation singularity on its axis. The constants p and q satisfying the normalisation condition $|p|^2 + |q|^2 = 1$, in accordance with Eqns (1) and (3), determine its characteristics

$$\Upsilon_{C0} = |p|^2 - |q|^2, \quad \eta_{C0} = \arg(q(p^*)^3). \quad (17)$$

An example of polarisation distribution in the cross section of such a beam is shown in Fig. 1a. The shape, relative size, and orientation of the polarisation ellipses are determined by the values of the beam electric field strength at spatial points coinciding with their centres. Filled and empty ellipses correspond to the right-hand and left-hand directions of electric field rotation; pluses and crosses, respectively, mark the polarisation singularity points with a positive and negative index. Naturally, the knowledge of the values of the total indices $I_{C3\pm}$ allows determining neither the number, nor the position and characteristics of polarisation singularities in the tripled-frequency signal beam cross sections. The analysis [11] of the expressions for $E_{3\pm}(r,\varphi,z)$, obtained by solving Eqn (13) analytically, showed that the signal beam contains one right-hand polarisation singularity point on the beam axis and two left-hand polarisation singularity points. The segment connecting them in any cross section of the beam crosses its axis and is divided in half at the point of intersection. Identical to each other in characteristics, these singularity points move in the cross section of the beam as it propagates (as the z coordinate increases). Thus, for $\Upsilon_{C0} > 0$, which corresponds to the situation $I_{C-} = 1/2$, $I_{C+} = 0$, we obtain the values $I_{C3+} = -1/2$, $I_{C3-} = 1$ (this is confirmed by the polarisation distribution shown in Fig. 1b in one of the cross sections of a beam generated in the bulk of the medium at a tripled frequency).

In the present work, by solving the equations for $E_{3\pm}(r,\varphi,z) = 0$ numerically, we not only determined the positions of singular points in the signal beam, but also found the dependence of the parameters Υ_C and η_C, characterising the distribution of polarisation ellipses near them, on the coordinate z . It is noteworthy that not only the topological indices of the generated polarisation singularities, but also their isotropy parameters Υ_C remain unchanged in the process of third harmonic generation and are equal to Υ_{C0} ($-\Upsilon_{C0}$) for the generated singularities having the left-hand (right-hand) circular polarisation. In turn, the parameters η_C change with increasing z, and these dependences are shown in Fig. 1c.

3. CARS signal generation in the bulk of an isotropic medium

Another example of a four-wave process that is possible in an isotropic nonlinear medium is the interaction of two light beams of fundamental radiation with frequencies ω_1 and ω_2, accompanied by the generation of a signal beam having the frequency $\omega_a = 2\omega_1 - \omega_2$. Such interaction of waves underlies coherent anti-Stokes Raman spectroscopy (CARS) [30].

Let us consider the collinear geometry interaction of the fundamental radiation beams in the approximation of their independent propagation along the z axis. In this case, the change in the slowly varying complex amplitudes $E_{m\pm}$ (hereinafter, $m = 1, 2$) of the circularly polarised components of the electric field of each beam is described by the linear diffraction equations:

$$2\mathrm{i}k_m \partial_z E_{m\pm} + \Delta_\perp E_{m\pm} = 0. \quad (18)$$

As in the previous section, we present their solutions as

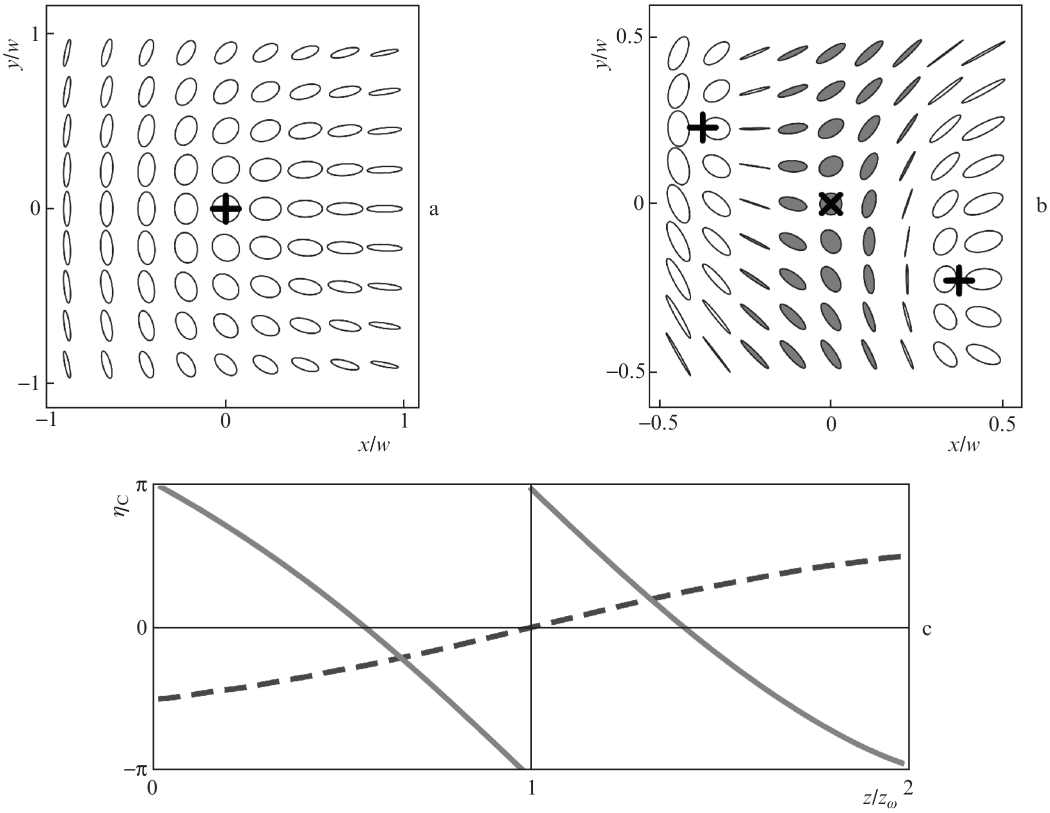

Figure 1. Polarisation distributions (a) in the beam waist of the fundamental radiation and (b) in the beam cross section at a tripled frequency, in which polarisation singularity points with a positive and negative index are indicated by pluses and crosses, respectively, as well as (c) the dependence on the z coordinate of the parameter η_C, characterising the distribution of polarisation ellipses near the singularity point of the right-hand polarisation (solid line) and one of the singularity points of the left-hand polarisation (dashed line).

$$E_{m\pm}(r,\varphi,z) = \sum_{n,l} c_{nl\pm}^{(m)} \Lambda_n^{(l)}(r/w_m, \varphi, \beta_m(z)). \quad (19)$$

All quantities entering Eqns (18), (19) are defined similarly to those in Eqns (5), (6), and the index m indicates that the corresponding quantity (e.g., k_m, w_m, β_m, etc.) is related to the mth beam. The multimode transverse structure of fundamental radiation waves leads to the appearance in their cross sections of phase singularity points of the circularly polarised components $E_{m\pm}$. In this case, the total topological indices $I_{\Phi m\pm}$ of individual components, as well as the total topological indices of the polarisation singularities of the vector fields of the beams $I_{Cm\pm}$, are determined by relations similar to (10) and (11). We emphasise that the structures of the fundamental radiation beams can be not at all related with each other and that the maximum values of $\tilde{p}_{m\pm}$ for the powers of r at the Gaussian exponential functions in the expressions for their amplitudes [see Eqn (9)] can be different.

The search for the slowly varying amplitudes $E_{a\pm}(r,\varphi,z)$ of the circularly polarised components of the signal beam electric field at the frequency ω_a implies knowing the circularly polarised components of the nonlinear polarisation of the substance $P_{\pm}^{(a)}(\omega_a)$, which are easily obtained from the constitutive equation [31]:

$$P_i^{(a)} = \chi_{ijpl} E_{1j} E_{1p} E_{2l}^{*} + \mathrm{i}(k_1 \gamma_{ijzlp}^{(1)} - k_2 \gamma_{ijzlp}^{(2)}) E_{2j}^{*} E_{1l} E_{1p}. \quad (20)$$

In Eqn (20), subscripts i, j, p, and l take the values x and y; the tensor $\chi_{ijpl}(\omega_a;\, \omega_1,\, \omega_1,\, -\omega_2)$ describes the local response of the medium; and two tensors $\gamma_{ijzlp}^{(1)}(\omega_a;\omega_1,\omega_1,-\omega_2)$ and $\gamma_{ijzlp}^{(2)}(\omega_a;\, -\omega_2,\omega_1,\omega_1)$ describe the spatial dispersion of the cubic nonlinearity. Taking into account (20), $P_{\pm}^{(a)}$ have the form

$$P_{\pm}^{(a)} = (\alpha_{\pm} E_{1\pm} E_{2\pm}^{*} + \beta_{\pm} E_{1\mp} E_{2\mp}^{*}) E_{1\pm}, \quad (21)$$

where

$$\alpha_{\pm} = 2\chi_1 \pm 4(k_2 \gamma_4^{(2)} - k_1 \gamma_4^{(1)}), \quad (22)$$

$$\beta_{\pm} = 2(\chi_1 + \chi_2) \pm 2(k_1 \gamma_1^{(1)} - k_2 \gamma_1^{(2)}). \quad (23)$$

In Eqns (22) and (23), $\chi_{1,2}$ are constants defining nonzero components of the tensor $\chi_{ijpl}(\omega_a;\, \omega_1,\, \omega_1,\, -\omega_2)$; $\gamma_{1,4}^{(1)}$ are two of the six constants that determine the nonzero components of the tensor $\gamma_{ijzlp}^{(1)}(\omega_a;\, \omega_1,\, \omega_1,\, -\omega_2)$; and $\gamma_{1,4}^{(2)}$ are two of the four constants defining all nonzero components of the tensor $\gamma_{ijzlp}^{(2)}(\omega_a;\, -\omega_2,\, \omega_1,\, \omega_1)$, symmetric with respect to permutation of the last two indices. When writing $\alpha_{\pm}$ and $\beta_{\pm}$, we corrected inaccuracies in formulae (10) and (11) in [31], which do not affect the formulae that follow from them and the formulated results.

To find the values of the total topological indices of phase singularities of each of the components $P_{\pm}^{(a)}$ and, therefore, the values of $I_{\Phi a\pm}$ and $I_{Ca\pm}$ for the electric field at the frequency ω_a is a much more difficult problem compared to the case of third harmonic generation considered above. This is because Eqn (21) includes a linear combination of products $E_{1+}E_{2+}^*$ and $E_{1-}E_{2-}^*$. However, the values of the total indices will be affected generally by only one of them, which occurs to contain the maximum power of r before the Gaussian exponential function after substituting into these products the explicit form of $E_{1\pm}$ and $E_{2\pm}$. The choice of one product or another is thus determined by comparing the sums $\tilde{p}_{1+}+\tilde{p}_{2+}$ and $\tilde{p}_{1-}+\tilde{p}_{2-}$. If the first of them is greater than the second one, then it is the product $E_{1+}E_{2+}^*$ that will determine the total topological singularity index of each of the fields given by the expressions in square brackets in Eqn (21). As a result, in this case

$$I_{\Phi a\pm} = I_{\Phi 1+} - I_{\Phi 2+} + I_{\Phi 1\pm}, \tag{24}$$

$$I_{Ca\pm} = I_{C1\pm} \pm (I_{C2-} - I_{C1-}). \tag{25}$$

If, however, the inequality $\tilde{p}_{1-}+\tilde{p}_{2-} > \tilde{p}_{1+}+\tilde{p}_{2+}$ holds, then we obtain the relations slightly different from (24) and (25):

$$I_{\Phi a\pm} = I_{\Phi 1-} - I_{\Phi 2-} + I_{\Phi 1\pm}, \tag{26}$$

$$I_{Ca\pm} = I_{C1\pm} \pm (I_{C1+} - I_{C2+}). \tag{27}$$

It is easy to show that the minus sign before $I_{\Phi 2\pm}$ arises due to the complex conjugation of the beam field components at the frequency ω_2 in Eqn (21).

A special case when the sums $\tilde{p}_{1-}+\tilde{p}_{2-}$ and $\tilde{p}_{1+}+\tilde{p}_{2+}$ are equal deserves a separate discussion. In this case, the two terms in brackets in (21) have equal maximum powers of the coordinate r before the Gaussian exponential function, and it is possible to determine the values of $I_{\Phi a\pm}$ only by applying to Eqn (21) a procedure similar to that described by Eqns (9) and (10). In this case, in contrast to the considered above, no unambiguous relationship exists between $I_{\Phi a\pm}$, $I_{\Phi m\pm}$, and the numbers $\tilde{p}_{m\pm}$.

Let us demonstrate this fact by an example of fundamental radiation beams with a rather simple structure

$$E_{1\pm}(r,\varphi,z) = \mu_{\pm}\Lambda_0^{(0)}(r/w_1, \varphi, \beta_1(z)), \tag{28}$$

$$E_{2\pm}(r,\varphi,z) = \Lambda_0^{(\pm 1)}(r/w_2, \varphi, \beta_2(z)), \tag{29}$$

where $\mu_{\pm} = \sqrt{(1 \pm M)/2}$. Formula (28) describes a Gaussian beam with uniform elliptical polarisation (Fig. 2a) characterised by the degree of ellipticity M, which can vary from –1 (left-hand circular polarisation) to 1 (right-hand circular polarisation). Circularly polarised beam components do not contain phase singularities ($I_{\Phi 1\pm} = 0$). Formulae (29), in turn, describe a beam with a zero on-axis intensity, linearly polarised all over the cross section with the electric field strength oscillating along the direction specified by the angle φ at each point with coordinates r, φ, z. Its circularly polarised components contain such phase singularities that $I_{\Phi 2\pm} = \pm 1$ (Fig. 2b). Substituting Eqns (28), (29) into Eqn (21), we obtain:

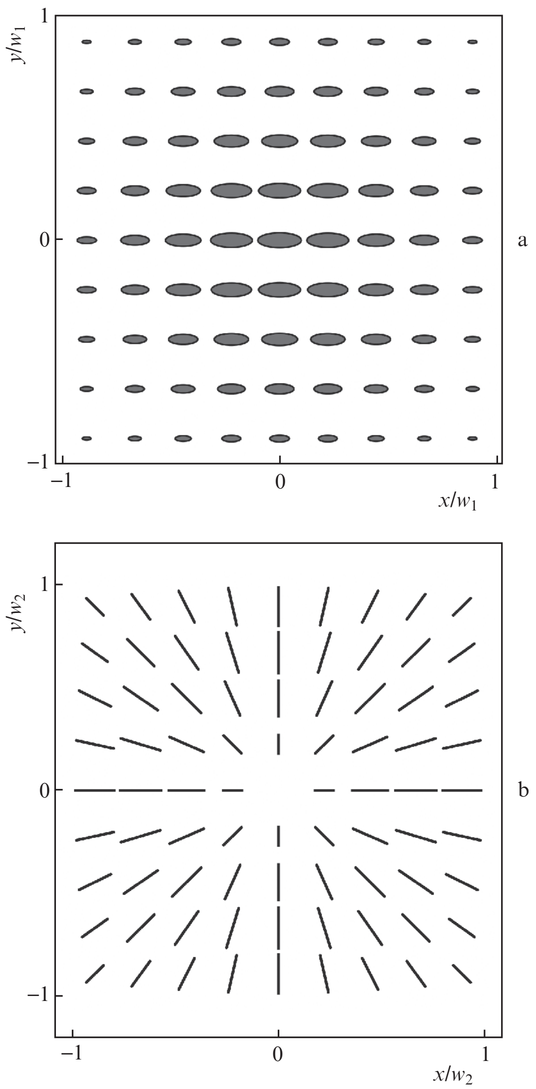

Figure 2. Example of the polarisation distribution in the cross sections of two beams of fundamental radiation at frequencies (a) ω_1 and (b) ω_2 in their waist planes, at which the total topological indices of their polarisation singularities are not related to the similar indices of the beam at a frequency of $2\omega_1 - \omega_2$. The shape, relative size and orientation of the ellipses in the figures are determined by the values of the electric field strength of the beam at the points located in their centre.

$$P_{\pm}^{(a)} = \left[\alpha_{\pm}\mu_{\pm}\exp(\mp i\varphi) + \beta_{\pm}\mu_{\mp}\exp(\pm i\varphi)\right] \times \frac{r}{w_2\beta_2}\exp\left(-\frac{r^2}{w_1^2\beta_1} - \frac{r^2}{w_2^2\beta_2}\right). \tag{30}$$

Comparing Eqn (30) with Eqns (9), (10), we arrive at the result

$$I_{\Phi a\pm} = \mathrm{sgn}\left(\frac{|\beta_{\pm}|^2 - |\alpha_{\pm}|^2}{|\beta_{\pm}|^2 + |\alpha_{\pm}|^2} \mp M\right). \tag{31}$$

It is seen that in the case of equal sums $\tilde{p}_{1-}+\tilde{p}_{2-}$ and $\tilde{p}_{1+}+\tilde{p}_{2+}$, the total indices $I_{\Phi a\pm}$ additionally depend on the interrelations between the material constants of the medium, present in the expressions for $\alpha_\pm$ and $\beta_\pm$, as well as on the polarisation state of the fundamental Gaussian beam containing no singular points.

4. Conclusions

We have analysed the previously obtained formulae for the transverse distribution of the electric field in the problems of third harmonic generation and the generation of the difference frequency $2\omega_1 - \omega_2$ in an isotropic medium in a coaxial interaction geometry. The multimode transverse structure of the incident beams of fundamental radiation gives rise to points of circular polarisation singularity in their cross sections and, as a consequence, in the cross sections of signal beams. Analytical expressions are obtained that relate the total topological indices of the singularity points of the left-hand and right-hand polarisations in the signal and incident beams (the values of the total topological indices were considered unchanged during beam propagation). The found relations turn out to be much more complicated than the 'expected' results. When the third harmonic is generated, the total indices do not triple, and when the frequency $2\omega_1 - \omega_2$ is generated, the values of the total topological indices of singularities in the signal beam can be affected by the ratios between the constants of the medium nonlinear response. It depends even on the state of polarisation of the fundamental radiation beam, containing no polarisation singularities at all. The found laws of transformation of the total topological indices allow one to get an idea of the fine details of these nonlinear optical processes and may be of interest for creating light beams and pulses with an inhomogeneous distribution of the electric field containing polarisation singularities of a given type by methods of nonlinear optics. The latter are promising for use in quantum information optical systems and can be used in problems of nonlinear bulk and surface spectroscopy of cubic-nonlinear media.

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